U.S. Production of Manufactured Gases: Assessment of Past Disposal Practices

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This study examines the history of the manufactured-gas industry of the United States,
its production processes, disposal trends, waste toxicity, methods of site investi-
gation, and the current status of manufactured-gas sites. The report is intended as
a guide to those who are examining and evaluating manufactured-gas sites for either
environmental risks or possible remediation.

Six manufactured-gas sites and one spent oxide disposal area were visited during
the project, and case studies were prepared for six former gas-manufacturing sites,
two byproduct tar utilization facilities, a creosoting plant and a coal tar processor.

The current status of manufactured-gas sites in the United States was determined
by contacting State and regional environmental officials and by discovering how their
regions were treating manufactured-gas sites.

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FOREWORD

Today's repidly developing and changing technologies and industrial products and practices frequently carry with them the increased generation of solid and hazardous wastes. These materials, if improperly dealt with, can threaten both public health and the environment. Abandoned waste sites and accidental releases of toxic and hazardous substances to the environment also have important environmental and public health implications. The Hazardous Waste Engineering Research Laboratory assists in providing an authoritative and defensible engineering basis for assessing and solving these problems. Its products support the policies, programs, and regulations of the Environmental Protection Agency, the permitting and other responsibilities of State and local governments, and the needs of both large and small businesses in handling their wastes responsibly and economically.

This report reviews the history of the U. S. manufactured-gas industry, the methods of production, wastes produced, disposal practices, potential environmental effects of disposed wastes, and methods of site investigation and remediation. Several specific manufactured-gas sites are examined, and a recent compilation of U. S. manufactured-gas sites is evaluated.

For further information,, please contact the Land Pollution Control Division of the Hazardous Waste Engineering Research Laboratory.

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Hazardous Waste Engineering Research Laboratory

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ABBREVIATIONS AND SYMBOLS

AGA   -- American Gas Association

bcf   -- Billion cubic feet

Btu   -- British thermal unit

CERCLA   -- Comprehensive Environmental Response, Compensation, and Liability Act (1980)

COD   -- Chemical oxygen demand

CP   -- Candle power

CWG   -- Carbureted water gas

DM_{30} -- Dimethyl sulfoxide

EC_{50} -- Effective concentration affecting 50 percent of test organisms

EM   -- Electromagnetic

EP   -- Extraction procedure

GAC   -- Granular-activated carbon

GC/MS   -- Gas chromatography/mass spectroscopy

g/kg   -- Grams per kilogram

gpm   -- Gallons per minute

H_2O_2/UV   -- Hydrogen peroxide/ultraviolet

HCN   -- Hydrogen cyanide

HPLC   -- High-performance liquid chromatograph

IARC   -- International Agency for Research on Cancer

LC_{50} -- Lethal concentration affecting 50 percent of test organisms

LP   -- Liquid petroleum

ABBREVIATIONS AND SYMBOLS (continued)

mg   -- Milligrams

mg/L   -- Milligrams per liter

Mcf   -- 1 million cubic feet (10^6 ft^3)

MSL   -- Mean seal level

NPL   -- National Priority List (Superfund)

O_3/UV -- Ozone/ultraviolet

PAH   -- Polynuclear aromatic hydrocarbon

PCAP   -- Preliminary contamination assessment plan

PCB   -- Polychlorinated biphenyl

PCP   -- Pentachlorophenol

ppb   -- Parts per billion

ppm   -- Parts per million

PVC   -- Polyvinyl chloride

RCRA   -- Resource Conservation and Recovery Act (1976)

RTI   -- Research Triangle Institute

SCS   -- Soil Conservation Service

SMR   -- Standardized mortality rate

TOC   -- Total organic carbon

μg/g -- Micrograms per gram

USGS   -- U.S. Geological Survey

VOC   -- Volatile organic compound

WHO   -- World Health Organization

EXECUTIVE SUMMARY

Former sites of gas manufacture present problems for remediation and reuse of the sites. In some cases, polluted groundwater and surface waters are near the sites. This study examines the history of the manufactured-gas industry of the United States, its production processes, disposal trends, waste toxicity, methods of site investigation, and the current status of manufactured-gas sites. The report is intended as a guide to those who are examining and evaluating manufactured-gas sites for either environmental risks or possible remediation.

The manufacture of gas for lighting and heating was performed in the United States from 1816 into the 1960's. Three major processes were used to manufacture gas: coal carbonization, carbureted water gas, and oil gas. Coal carbonization consisted of heating bituminous coal in a sealed chamber, with destructive distillation of gas from the coal and the formation of coke. The gases were collected, cleaned, and distributed while coke was removed and sold or used. The carbureted water-gas process used coke (or coal), steam, and various oil products to produce a combustible product gas. Steam was fed through a bed of incandescent coke, producing a gas containing hydrogen and carbon monoxide. This gas (blue gas) then passed through two chambers containing hot firebrick, where oil was sprayed into the gas and cracked into gaseous hydrocarbons and tar. Oil gas cracked oil alone into gaseous hydrocarbons, tar, and carbon (lampblack). A variety of oil-based feedstocks were used in the production of carbureted water gas and oil gas, including naphtha, gas oil, fuel oil, and residuum oils.

The byproducts from the three processes were similar, but there were important differences, which affect both the current character of wastes and their toxicity. Tars produced from coal carbonization contained substantial amounts of phenols and base nitrogen organics. The tars from carbureted water gas and oil gas contain only trace amounts of these compounds because they were not produced during the manufacture of gas. Coal carbonization also

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produced substantial amounts of cyanide in the gas, which was removed during gas cleaning and often appears in current wastes. Carbureted water gas and oil gas produced only trace amounts of cyanide, and cyanide does not appear in substantial quantities in wastes from these processes. Likewise, ammonia was produced by coal carbonization, but it was not produced by oil or carbureted water-gas manufacture. Wastes from the recovery of ammonia occur at plants that coked coal to produce gas, but not at plants producing only carbureted water gas or oil gas.

Gas production in the north central United States was principally coal carbonization, oil gas was predominant on the West Coast, and carbureted water gas was predominant in the South, the East Coast, and the Northeast. The variation in the production processes used in various areas of the United States reflects the relative cost of raw materials for production and markets for byproducts in the regions. The types of production employed changed with time, as did the materials used for gas production. This influenced both the types of wastes produced and the disposal practices of the plants. Plant size and access to markets were two major factors affecting the disposal practices of manufactured-gas plants.

Tars and oils were produced as byproducts from all three production processes. The tars and oils were generally recovered as byproducts from the production of town gas, and they were usually separated from condensate water by gravity separators. The tars could be either sold (as fuel or to tar refiners), refined at the plant site, or burned in the boilers of the gas plant. The recovered tars had a minimum value to the producing plants as fuel because the use of tars as fuel replaced other fuels used for steam production. Some tars were disposed very early in the production of coal-carbonization gas, but recovered tars during this period were also frequently burned in the coal-carbonization retorts. Smaller gas plants often produced tars in insufficient quantities to justify their recovery, and these were disposed with the waste condensate (this was particularly true of the carbureted water-gas plants). Emulsions of tar and water occurred with the production of carbureted water gas and oil gas, and because these were difficult to separate, they were frequently disposed. The waste sludge from

the purification of light oils was generally disposed on the plant "dump," along with other off-spec or difficult to handle tars.

The tars produced for carbureted water gas were usually less viscous and less dense than were the tars produced by coal carbonization. These tars are more mobile in the environment than are most coal tars. The properties of collected tars changed with respect to where the tai :s collected within the purification trains. The heavier tars condensed first within the gas purification system, and these were the most viscous and the densest tars. The tars that condensed later in the purification system were less viscous and dense. Volatile organics (such as benzene and toluenes) were either scrubbed from the gas as light oil or condensed in the gas holders or distribution pipes as "drip oil." The variety of tars and oils produced within manufactured-gas plants contributed to the wide range of organic contamination generally present at gas sites.

Leakage of petroleum oils, tars, and aqueous condensates occurred frequently from gas plants during plant operation. Early vessels used for the underground storage of liquids were constructed of wood or brick. Several historical references indicate that groundwater contamination was common near gas plants, caused both by unintentional leakage from the plants and intentional disposal practices.

The oils and tars from gas manufacture contain relatively high concentrations of polycyclic aromatic hydrocarbons and are carcinogenic, with numerous cases of skin cancer correlated with the occupational use of tars and tar products. Phenols (from coal carbonization) are toxic to human, animal, and plant life. Small concentrations of phenols cause taste and odor problems in drinking water, imparting a medicinal taste to the water. Spent oxides frequently develop low pH's and have relatively high concentrations of tars, and the iron cyanide complexes in spent oxide from coal carbonization appear very stable and have relatively low toxicity.

The site investigation techniques applied to manufactured-gas sites are not significantly different from those applied to other uncontrolled waste sites and appear adequate for site assessment. Surface geophysical techniques can be applied to help identify buried structures and the extent of possible contamination. The location of underground structures at a site is

particularly important because such structures frequently contain tars or oils that could eventually leak or be released during future actions on the site. Historical information on the operation, production, and layout of the gas plants is frequently available and should be used wherever possible. Maps of plant sites can be used to locate underground structures and possible dump areas around the sites. The types of production employed by a plant determine the nature and types of wastes produced, and the amount of gas produced frequently affects the amount of waste remaining on a site. Many of the sites examined to date are fairly stable (no wastes, currently observed moving off the site). These sites can often be adequately managed by taking no remedial actions until the site is to be redeveloped.

Six manufactured-gas sites and one spent oxide disposal area were visited during the project, and all showed visible contamination of soil by tars. Ferrocyanides were visible form spent oxide at plants that produced coal gas, but they were absent from those sites that produced carbureted water gas. The characteristic odor of gas-manufacturing plants was observed at all the sites examined. In addition to the visited sites, case studies were prepared for six former gas-manufacturing sites, two byproduct tar utilization facilities, a creosoting plant, and a coal-tar processor. These case studies were prepared primarily from articles reported in the literature and illustrate current methods of site assessment and remediation.

The current status of manufactured-gas sites in the United States was determined by contacting State and regional environmental officials and by discovering how their regions were treating manufactured-gas sites. Many States are examining manufactured-gas sites with other waste sites, and most of these are conducting preliminary assessments of the sites. Where the manufactured-gas sites have been ranked (by risk assessment), they have generally been ranked as posing a low hazard to both humans and the environment. Groundwater contamination has been reported as several sites, but it is not significant at many of the sites examined.

INTRODUCTION

> After we had gone to the trouble of eliminating the oil and tar from
> the stream, we met a difficulty not at all anticipated. Very near
> our works and about ten years after they were installed, an arti-
> ficial ice plant was erected. The owners decided to dig artesian
> wells and found water of excellent quality, and ample quantity which
> they used for three or four years with no evidence that we, their
> neighbors, would cause them any trouble. In the early days of the
> gas plant, the tar waste from the works had leaked through broken
> pipelines and from the wooden separator box used for waste disposal.
> The tars seeped through the ordinary fissures of rock into the
> ground around the well casing, and traces of oil began to appear in
> the well water. Needless to say, there was very serious trouble for
> a while and it is possible that other plants are storing up,
> unawares, difficulties of the same kind (Dutton, 1919).

Between 1816 and the 1960's, combustible gas for heating, cooking, and
lighting was manufactured from coke, coal, and oil at 1,000 to 1,500 sites in
the United States. These facilities were called gas plants, gasworks, or town
gas plants. For most areas of the country, manufactured gas was the major gas
fuel available for use during this period. Some regional natural gas pipe-
lines were established before World War II, but it was only after the war that
the technology was available for a national system of interstate gas pipe-
lines. As natural gas was introduced into areas previously served by natural
gas, the gas companies stopped the gas-manufacturing operations and became
distributors of natural gas. Most companies maintained the manufacturing
facilities for several years after natural gas was available so that gas could
be manufactured to meet peak demand. With better storage of gas and the
installation of multiple pipelines serving regions, there was no longer any
need for manufactured gas, and the plants were demolished.

The old gas manufacturers frequently disposed solid and liquid wastes
onsite, making the current sites difficult to redevelop and posing potential
environmental problems from either groundwater or surface water contamination,
as evidenced by L.R. Dutton's testimony given at the start of the chapter.

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This report reviews the history of the U.S. manufactured-gas industry, the methods of production, wastes produced, disposal practices, potential environmental effects of disposed wastes, and methods of site investigation and remediation. Several specific manufactured-gas sites are examined, and a recent compilation of U.S. manufactured-gas sites is evaluated.

Chapter 1 is a complete historical review of the U.S. manufactured-gas industry, principally using information generated by the manufactured-gas industry while it was in operation. The chapter reviews the production processes (1.2), gas purification methods (1.3), wastes produced and disposal methods 1.4), trends of the gas industry (1.5), and a comparison of U.S. practices and those used in Great Britain.

Chapter 2 describes the techniques previously used for site investigations (2.2) and site remediation (2.3). Chapter 3 reviews several specific town gas sites, both those visited by the Research Triangle Institute (RTI) (seven sites, Section 3.2), and sites reported in and reviewed through available literature (six gas-manufacturing sites and two tar-processing plants, Section 3.3). Chapter 4 examines a recent compilation of town gas sites and current handling of gas sites by individual States.

1.0 HISTORICAL REVIEW OF THE TOWN GAS INDUSTRY

1.1 INTRODUCTION

This chapter is a review of the processes, wastes, geographic trends, and historical trends of the U.S. town gas industry. The wastes produced from different production processes are frequently similar, but substantial differences in waste types, volumes, and disposal are dependent on the production method employed. The chapter is divided into several sections: 1.2 describes the production methods used to produce town gas in the United States; 1.3 describes the gas cleaning and purification processes; 1.4 describes the wastes, byproducts, and disposal practices of the industry; 1.5 details the temporal and geographic trends of gas production, and Section 1.6 describes the significant differences between U.S. and U.K. gas industries.

A review of literature from the town gas industry was conducted as a part of this project. The production of synthetic gases for use by consumers in cities and towns was once a substantial industry in the United States, and a large amount of information is available concerning the industry (although most of it was published before 1955). This material exists as books, manuals, journals, and conference reports. Much of it is not indexed, and virtually none of the information was ever placed into computer data bases for rapid access. Approximately 300 articles and books concerning the town gas industry were collected and examined during the project. Special emphasis was placed on materials concerned with waste generation and disposal practices in the town gas industry. Statistical information about town gas production was also collected to show geographic and time-dependent trends.

Table 1 is a list of the journals reviewed as part of the historical literature review. This historical review principally covers the production of flammable gas for distribution to consumers. The production of industrial fuel gases and coke is described but not reviewed in depth.

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TABLE 1. LIST OF MANUFACTURED-GAS PERIODICALS REVIEWED

American Gas Association Proceedings

Pennsylvania Gas Association Proceedings

Proceedings of the Southern Gas Association

Proceedings of the Pacific Gas Association

Proceedings of the Illinois Gas Association

Proceedings of the American Gas Institute

Proceedings of the American Gas Light Association Proceedings

Indiana Gas Association Proceedings

Proceedings of the New England Association of Gas Engineers

Gas Age (Gas Age Record)

American Gas Association Monthly

American Gas Journal

Progressive Age

Brown's Directory of Gas Production Plants[a]

[a]Data concerning several gas production sites were collected from Brown's Directory.

1.2 TOWN GAS PRODUCTION

1.2.1 Producer Gas Production

Producer gas was not distributed to towns for lighting or heating, but it was used extensively as a fuel gas within gas-manufacturing plants. Producer gas has a relatively low heating value and very few illuminants, and it was only used where the gas was burned near its production location. Producer gas was initially manufactured by burning coal or coke with insufficient air for complete combustion. This produces a flue gas high in carbon monoxide that was combined with additional air to complete the combustion wherever the heat is required. The early Siemens gas producer (1861) operated in this manner. Steam was later added to the air stream flowing into the coke bed to cool the bed and to add additional CO and H_2 to the producer gas using the two reactions, $H_2O + C = H_2 + CO$ and $2H_2O + C = CO_2 + 2H_2$. Figure 1 is a diagram of a producer gas bed and the relevant gas production reactions. The conditions and flows on this figure are only approximate because the actual numbers depend very highly on the operation of the gas producer.

Producer gas used either coke, bituminous coal, anthracite coal, or coke-coal mixtures for fuel. Producer gas composition varied with the fuel used, rate of air feed, and amount of steam used. Gas produced from coke or anthracite would contain no tar materials whereas some tar would be evolved from bituminous coal. If the gas were to be burned near the producer, these tars could be burned with the producer gas. When the gas was transported a short distance or was burned with orifice-type burners, coke or anthracite coal would be used to avoid problems of tars condensing in the pipes and burners. Any cleanup of the gas prior to combustion was performed with dry scrubbers (usually filled with woodchips). Additional cleaning was rarely performed because it would require cooling the gas with loss of the heat and combustible tars contained in the gas. It is possible to recover tar and ammonia from producer gas, but this was not widely practiced in the United States (Morgan, 1926). The tars (from bituminous fueled production) and the ash from the producer would be the primary waste products from producer gas manufacturing. Because the gas was burned for industrial uses, impurities in the gas (H_2S, HCN, CO_2) were not removed prior to combustion. Table 2 shows the approximate composition and characteristics of producer gas from bituminous coal.

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54 LB COAL PER SQ FT PER HOUR

GAS   SPACE
DECREASE IN HEATING VALUE

67 CU FT OF
154 BTU GAS
PER LB COAL
AT 1250 °F

DISTILLATION ZONE
VOLATILE MATTER OF FUEL
ADDED TO GAS

10'

15'

SECONDARY REDUCTION ZONE

$C + CO_2 = 2CO$ $-70,200\ BTU$

$CO + H_2O = CO_2 + H_2$ $-700\ BTU$

ZONE OF
PREHEAT

HEAT
FLOW

PRIMARY REDUCTION ZONE

$C + H_2O = CO + H_2$ $-70,900\ BTU$

$C + 2H_2O = CO_2 + 2H_2$ $-71,600\ BTU$

$C + CO_2 = 2CO$ $-70,200\ BTU$

HEAT
ABSORBED

10'

HEAT
FLOW

OXIDATION ZONE

$C + 3.76N_2 + O_2 CO + 3.76N_2 + 174,600\ BTU$

HEAT
EVOLVED

3" TO 5"

ASH   ZONE

3 LB AIR AND
0.3 LB STEAM
PER LB COAL

Figure 1. Producer gas production.

Source: *Gas Engineers Handbook*, 1934.

TABLE 2. COMPOSITION AND CHARACTERISTICS OF PRODUCER GAS

Kind of operation	Gas analysis			Characteristics of Gas		Appearance of fuel surface	Value of gas obtained and causes of poor operation
	Percent CO_2	Percent CO	Percent H_2	Temperature, F	Appearance		
Cold fire	Over 7	Below 20	13-17	1,200-1,300	Golden yellow;	Dull red or pale cherry	Very bad; combustion too slow; upper part much hotter than lower; too much steam or grate choked
Good fire	3-6	About 24	About 16	1,300-1,500	Yellow; still much tar; often a little smoky from decomposed tar	Cherry to bright red	Excellent operation
Good fire - with channels in bed	Over 6	Less than 24	16 or less	1,300-1,500	Smoke; tar decomposed	Cherry to bright red	Upper part much hotter than lower; poor operation, not enough poking, ashe too sticky, or too much fusible ash
Hot fire	3-4	Over 26	5-10	Well above 1,500	Smoke; tar entirely decomposed	Bright red to pale yellow rich- or low-ash coals changes this to next type	Not enough steam; operation possible, and sometimes to be recommended with channeling
Hot fire with channels in bed	Over 3 or 4	Less than 26	5-10	Well above 1,500	Smoke; tar entirely decomposed	Bright red to pale yellow	Not enough steam, too much ash; ash too fusible; not enough poking; very poor gas, distinguished by low H_2

SOURCE: Haslam and Russell, 1926.

Producer gas was manufactured for industrial use and for use within gas-manufacturing facilities. Because many installations were at industrial plants, there is little available data on the number of producer gas installations in the United States. An estimate of the number of producer gas facilities in the U.S. (about 1921) is in Table 3. This table does not include gas producers used with the production of town gas. There were many different types of machinery for producer gas manufacturing. Production equipment was classified by draft direction (up or down), production pressure (suction or positive), feed method (hand or mechanical), poking method (hand or mechanical), ash removal (hand, intermittent, or continuous), cleanliness of gas produced, and equipment location (attached as part of combustion equipment or centrally located). Figure 2 is a diagram of a typical gas producer, the Chapman. The body of the producer is stationary, and the bed is poked by a revolving agitator that floats on top of the coal bed. Air and steam is fed to the bottom of the bed, ash continuously removed from the bottom of the bed, and coal continuously fed into the top of the bed. The producer gas is removed through a pipe near the top of the apparatus.

In contrast to centrally located producers (like the Chapman), producer gas installations at town gasworks were frequently an integral part of the machinery with produced coke-oven gas and coal gas. Figure 3 shows a horizontal retort for coal-gas production with an attached producer gas generator. The producer gas is made in the chamber at the base of the apparatus and the gas produced combined with secondary air and combusted to heat the six horizontal retorts. This apparatus is described further in the coal-gas section (1.2.2.2).

1.2.2 Coal-Gas Production

1.2.2.1 Introduction--

The discovery that combustible gas could be produced from coal was first described by Dr. John Clayton, who between 1660 and 1670 heated coal and described the gas and tar produced. The first practical application of coal gas was when William Murdock, a Scottish engineer, illuminated his home in 1792 with gas from coal distilled in an iron retort. The basic method of producing coal gas has remained substantially the same ever since. A bituminous coal is placed in a closed vessel that is heated. The evolving gases are

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TABLE 3. ESTIMATED DISTRIBUTION OF GAS PRODUCERS IN THE
UNITED STATES

Industry	Number of producers
Steel	6,500
Glass	1,500
Ceramics and lime burning	1,500
Power generation	1,000
Metallurigical and other chemical fields	500
Total	11,000

SOURCE: Chapman, 1921.
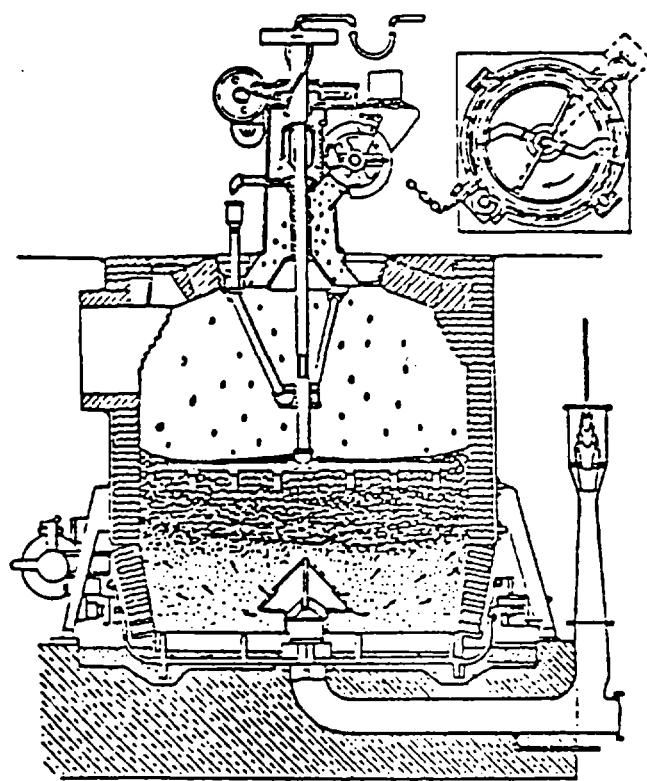
Figure   2. Chapman gas producer.

Source: Haslam and Russell, 1926.
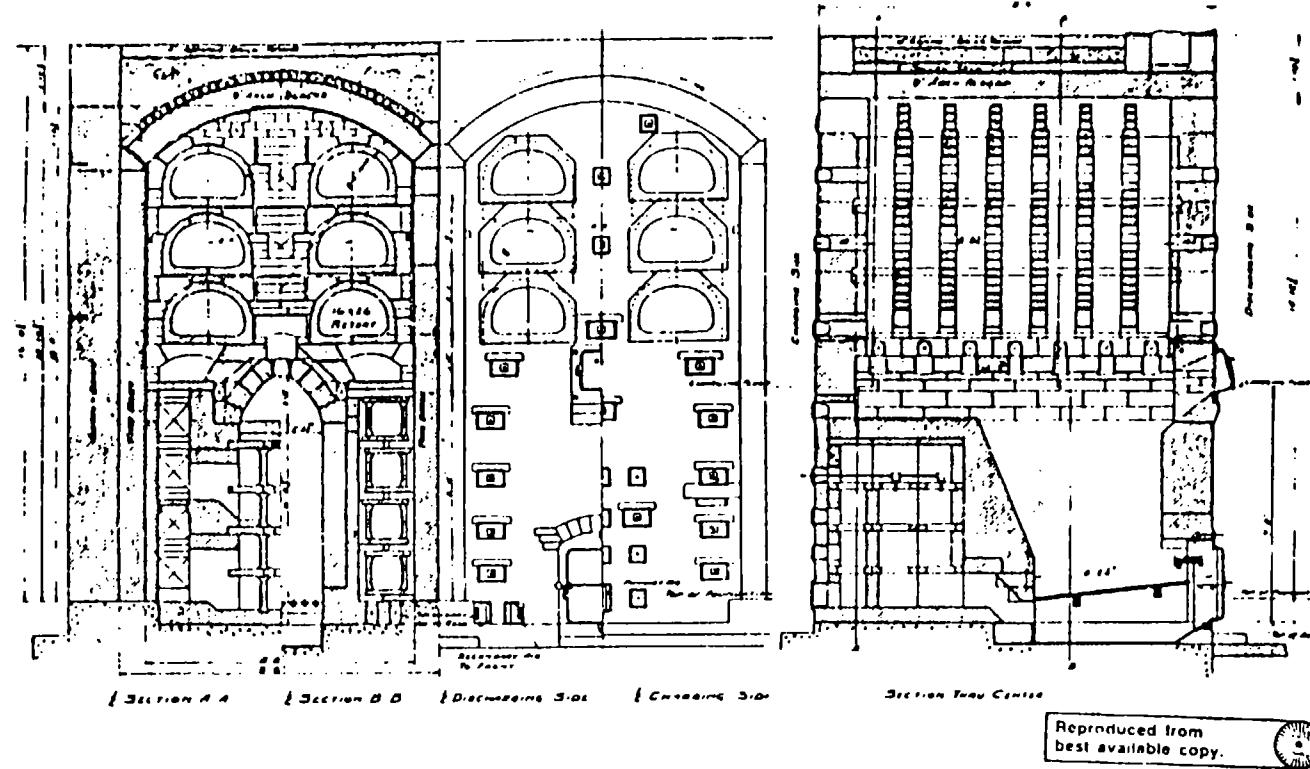
Figure   3.   Horizontal retort with internal producer gas generator.

Source: Morgan, 1926.

then removed and burned for heat or light. The coal remained in the vessel until all of its volatile materials evolved as gas, then the coke was removed from the vessel. This section reviews the various apparatus and methods that were used for the production of coal gas. It is divided into two classes of carbonizing apparatus--retorts and coke ovens.

1.2.2.2 Coal-Carbonization Retorts--

Coal-carbonization retorts were vessels in which bituminous coal was placed and heated external'y to destructively distill volatiles from the coal. The major features common to coal retorts are (1) a closed vessel containing coal, (2) a method of heating the vessel, (3) removal of volatiles from the retort, and (4) methods of filling the retort with coal and removing coke. Because the requirements for a retort were relatively simple, a wide variety of retorts for coal carbonization were manufactured and used for gas production. The major types were horizontal retorts, inclined retorts, and vertical retorts.

The earliest retorts were essentially cast iron kettles with lids. The kettles were filled with coal, covered, and heated by a coal fire. Gas from the coal was removed through a pipe, cooled, and distributed. Coke was removed from the kettle by hand, after the kettle had cooled. The kettles were rapidly replaced by horizontal retorts constructed of cast iron. These retorts were cylindrical or half cylindrical tubes about 7 feet long with one end sealed. The open end was used for charging with coal, and removing coke. During gas production, the open end was sealed by a door, and the coal gas removed through a tube at the door. The retorts were heated by fires below the tube or by producer gas to between 600 and 800 °C. The cast iron retorts had a relatively short lifetime (6 to 8 months) and required frequent replacement. Horizontal retorts of clay refractory materials replaced the cast iron retorts around 1850.

Horizontal retorts constructed of clay refractory were the major method of producing coal gas through the start of World War I. They were similar in construction to the cast iron horizontal retorts, but,were larger and carbonized coal at higher temperatures than the cast iron retorts (above 900 °C). They also had an average life of 2.5 years, compared to 6 to 8 months for a cast iron retort (Hughes and Richards, 1885). A typical horizontal retort is

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shown in Figure 4.   It consists of six retorts (the "D" shaped objects, approximately 16 to 26" wide x 18" high x 8 to 20" long) and a producer gas furnace for heating the retorts. A set of retorts, and their heating apparatus, is called a bench. Benches varied in the number of retorts per bench but were usually fewer than 10 retorts. The producer gas furnace has two doors; the small upper door is for charging with either coke from the retorts or coal, while the lower door is for poking the bed and ash removal. Primary air and steam is fed to the base of the producer bed. The producer gas is then burned with secondary air around and between the retorts for heat. The flue gas then exits through a stack at the rear of the retorts and sent to a waste heat boiler or exhausted. Each retort has a door and a standpipe that carries the tar and gas to a hydraulic main (essentially a water seal) above the bench. Typical operation of the horizontal retorts after starting the bench consisted of removing coke from the retorts and recharging them with coal. Periodically, coal or coke was added to the producer below the retorts, ash was removed from the producer, carbon buildup on the inside of the retorts was removed (scurfed), and the gas standpipes cleaned.

All of the retort operations were originally performed by hand, until machines for charging coal and discharging coke from horizontal gas retorts were developed. Figures 5 and 6 show machines for charging and pulling horizontal gas retorts. These machines usually used doors on each end of the retorts.

Several other types of retorts, similar to horizontal retorts, were used after 1900. They varied in the orientation of the retorts and were either inclined or vertical retorts. These two types were further divided into intermittent retorts (charged and discharged as a batch process) or continuous (with continuous feeding of coal and removal of coke). Inclined retorts have the same design as horizontal retorts except the retorts are inclined at about 30°, with doors at each end of the retort. The original concept was to feed the coal at the top of the retort and remove the coke from the bottom, with gravity assisting the operation. In actual operation the coke frequently jammed in the retorts and had to be removed by hand. It was also difficult to heat the retorts evenly, and few installations were made in the United States. .

Vertical retorts placed the retorts vertically, with coal fed to the top of the retort and coke removed from the bottom. They came into general use

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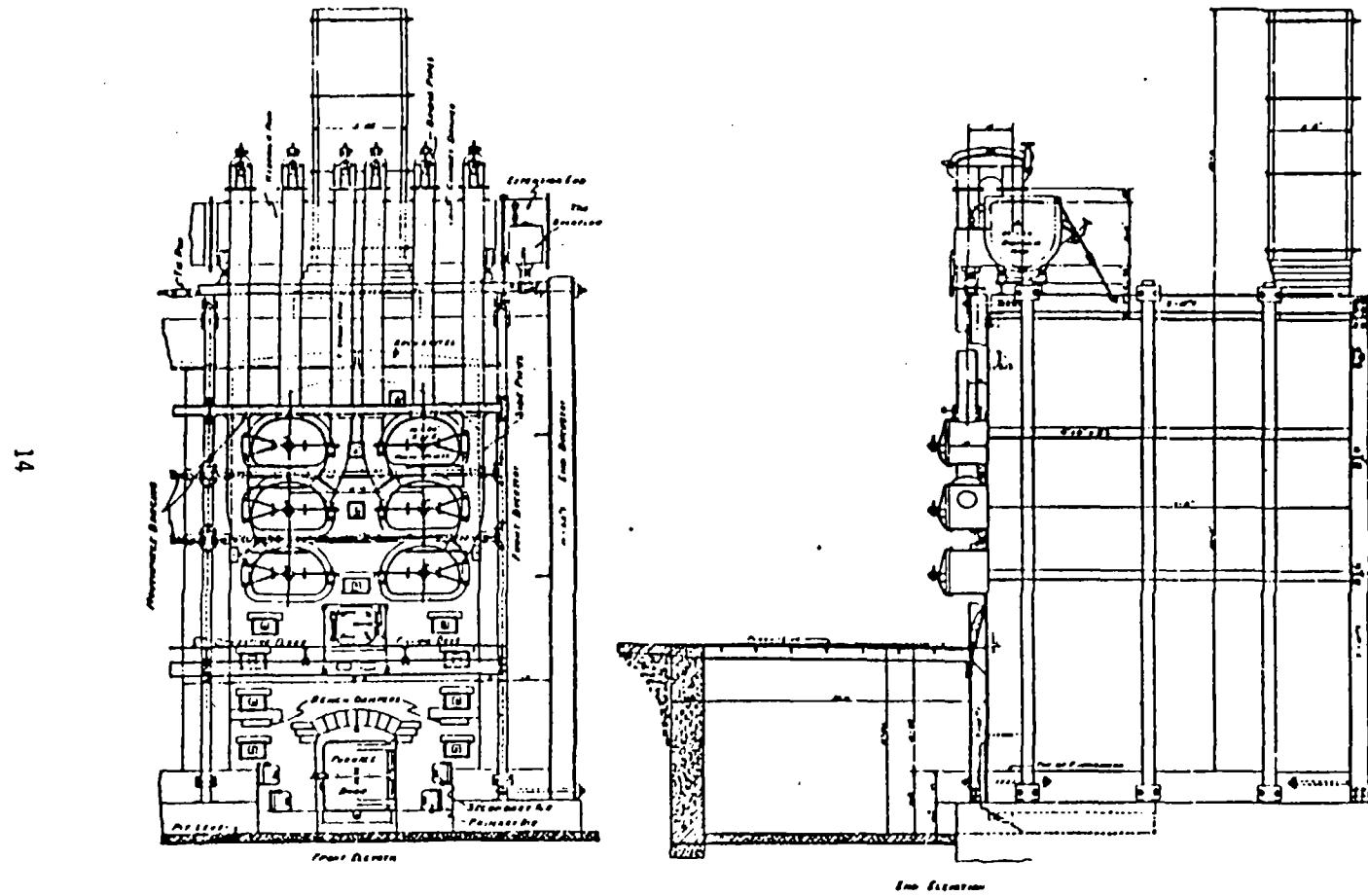
Figure   4. Horizontal retort.

Source: Morgan, 1926.

GATE

10 TON STORAGE HOPPER

1000 LBS WEIGH HOPPER

GATE

TELESCOPIC CHUTE

SCALE DIAL

20 FT. RETORT

GROOVED PULLEY

CHAIN DRIVE

ELECTRIC MOTOR

CHARGING BELT

DE BROUWER CHARGING MACHINE
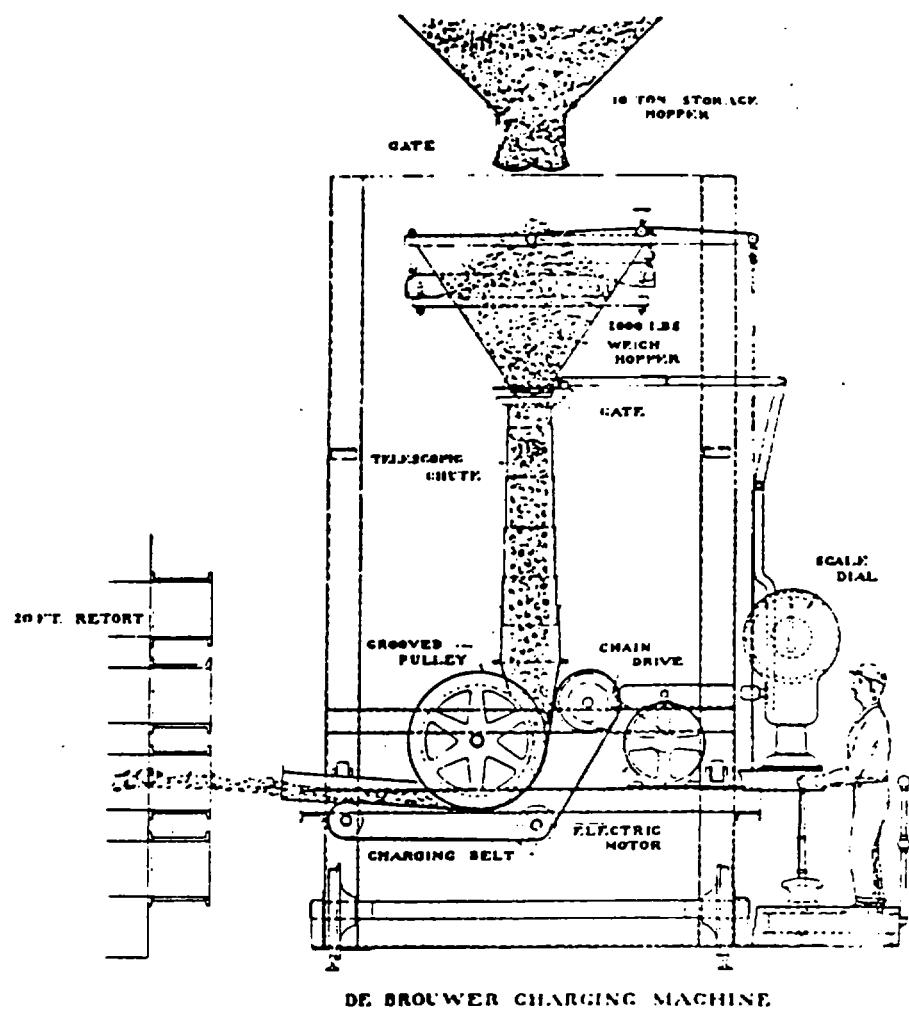
Figure 5. Charging machine.

Source: Morgan, 1926.

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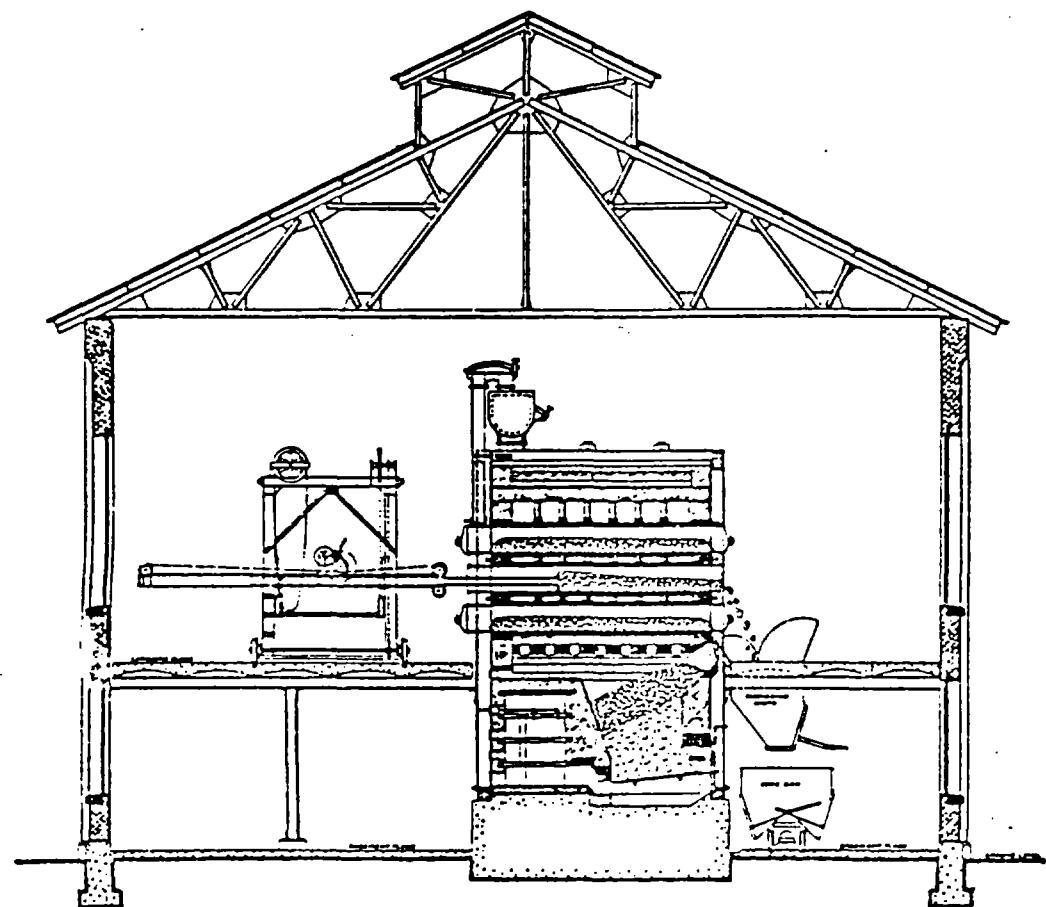
Figure   6. Coke pusher.

Source: Morgan, 1926.
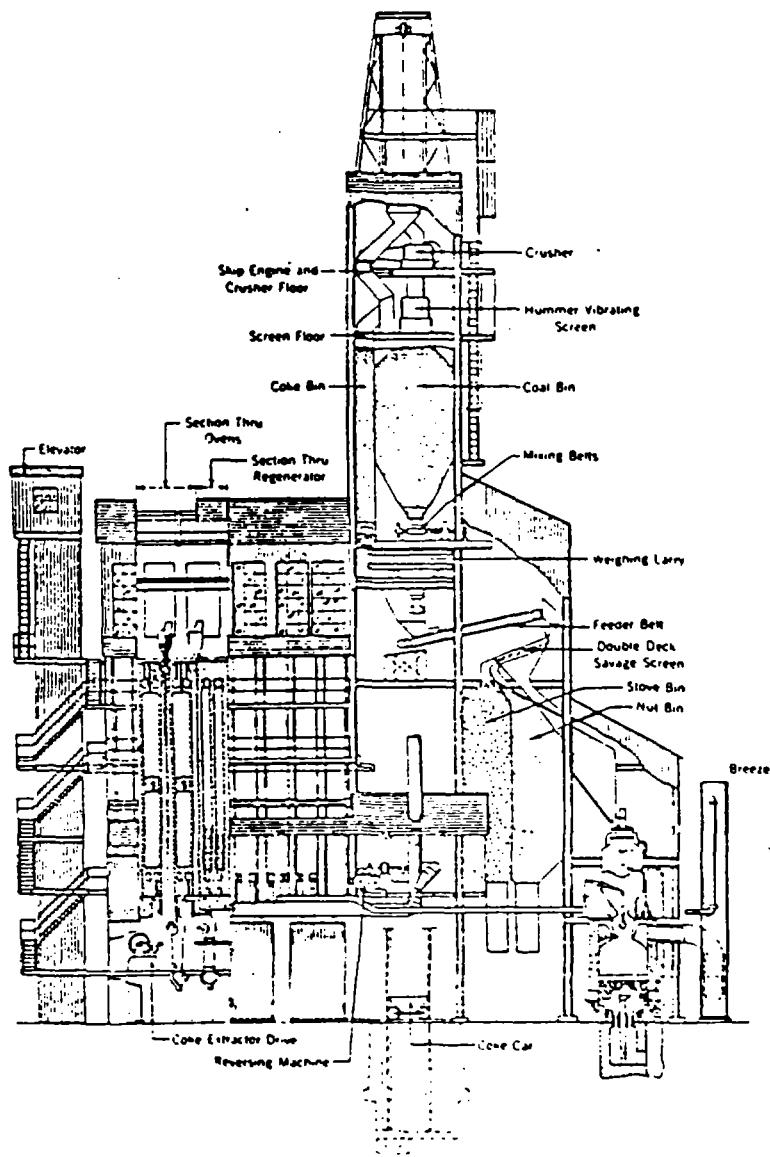
Figure 7. Continous vertical retort.

Source: Denig, 1945.

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tion. Prior to the introduction of byproduct coke ovens, most coke was manufactured in beehive coke ovens that produced coke from bituminous coal, with no collected byproducts. They did not produce gas for distribution but were the oldest form of oven for the carbonization of coal.

The name "beehive" comes from the oven's shape, which is similar to the old basket beehives. Figure 8 is a diagram of the beehive coke oven. The oven was charged with coal through the hole in the top, and the coal was coked by admitting air through openings in the side door. Volatiles from the coal were burned within the chamber, providing heat for devolatilizing the layer of coal on the bottom of the beehive oven. Charges of coal in the beehive ovens were typically 5 or 6 tons, with coking occurring oven 2 to 3 days. After coking, the coke was removed from the oven and quenched with water. Beehive ovens lose all of the volatiles of the coal, either to the air or by combustion, and were inefficient compared to coking methods that recovered these components. The major advantage to the beehive oven was its ability to produce high-grade coke with a minimum of capital investment. Waste heat ovens were similar to the beehive ovens but attempted to better utilize the waste gases from the coking chamber, which were collected and burned under the oven with air for additional heat. The only waste produced by the beehive and waste heat ovens is the coke quench water, which may have been contaminated with some of the organics remaining in the coked coal.

The substantial waste of heat, combustible gases, tars, ammonia, and volatiles from the operation of beehive coke ovens led to the development of coking processes that would produce a high-grade coke, conserve heat, and recover marketable byproducts. Byproduct coke ovens are basically large horizontal retorts, but with large rectangular coking chambers and more mechanized movement of coal and coke. Figure 9 shows a typical byproduct coke oven. The ovens are rectangular chambers that are approximately 40 feet long by 10 to 12 feet and 12 to 20 inches wide. They are charged from the top with coal, and heated by combustion in flues along the sides of each oven. After the coal is coked, doors at each end of the oven are opened, and the coke is mechanically pushed from the chamber and quenched with a water spray.

Byproduct coke ovens were constructed for the economical production of metallurgical coke and recovery of byproducts from the coking process. Exten-

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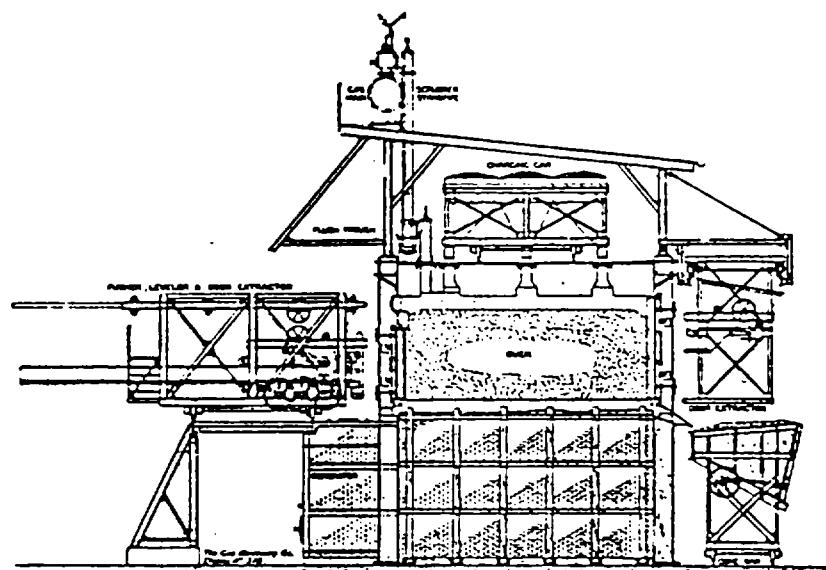
Figure   9. Byproduct coke oven.

Source: Morgan, 1926.

sive recovery and recycling of waste heat was practiced to reduce fuel consumption for heating the coke ovens. Coke ovens were produced in many models with variations in oven size, flue orientation (horizontal or vertical), method of heat recovery (recuperative or regenerative), and type of gas used to heat the ovens.

Byproduct coke ovens could be heated by 35 to 40 percent of the coal gas produced within their ovens. This left approximately 60 percent of the coal gas as a surplus that could be sold and distributed to industrial users or consumers. The coke-oven gas had a heating value of about 560 Btu/ft^3 (after being stripped of light oils) and was readily marketable as a fuel gas. Many coke ovens produced lower Btu gases (producer gas or blue gas) to heat the coke ovens, freeing a larger portion of the coal gas for sale. This allowed coke-oven facilities more flexibility in the quantity of gas they could sell. In periods of low gas demand, coal gas would be burned to heat the ovens, but in periods of higher gas demand all coal gas would be sold and the ovens heated with producer gas or blue gas.

Table 4 shows the gas composition of coal gas produced from byproduct coke ovens, horizontal retorts, and vertical retorts. These gases are all produced by the carbonization of bituminous coal and are very similar in composition and heating value. The cleanup processes, byproducts, and wastes from these coal-carbonization processes are also very similar and are discussed in Sections 1.3 (cleanup processes) and 1.4 (wastes and byproducts). The raw coal gas was cleaned to remove tar, ammonia, cyanide, and hydrogen sulfide. The byproducts from these cleanup processes were either sold, used, or disposed.

Because many products besides gas were produced from coal carbonization, there was a substantial overlap between coke-manufacturing companies selling gas as a byproduct and gas production companies selling coke, ammonia, and tar as byproducts. Some gas distribution companies purchased coke-oven gas for distribution but did not manufacture the gas. The distinction between coke companies and gas companies is not important from a process standpoint, but it is an important consideration when determining who will pay for site remediation (e.g., gas companies were absorbed by current gas distribution companies, while coke producers remained as a separate industry).

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TABLE 4.   COMPOSITION AND TOTAL HEATING VALUES OF TYPICAL COAL GASES

Kind of gas	Carbon dioxide	Illumi-nants	Oxygen	Carbon monoxide	Methane	Ethane	Hydrogen	Nitrogen	Btu/ft^3
Coal Vertical retorts	2.0	3.6	0.2	6.5	33.1	1.8	51.0	2.8	615
Coal Horizontal retorts	1.5	5.6	0.2	6.0	31.0	--	52.5	3.2	615
Coal Coke ovens	1.8	3.7	0.2	6.3	31.6	--	53.0	3.4	586

SOURCE: Morgan, 1926.

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1.2.3 Carbureted Water Gas

Blue gas is prepared by passing steam through a bed of incandescent carbon. The steam reacts with the carbon to produce a fuel gas composed primarily of carbon monoxide and hydrogen. This gas is also known as water gas or blue water gas. When liquid hydrocarbons are thermally cracked into the water gas, a fuel gas known as carbureted water gas (CWG) is produced. Blue gas was sometimes produced as an industrial fuel but was not distributed to consumers because of its low fuel value (about 300 Btu/ft^3) and lack of illuminants (bright-burning hydrocarbons). The shortcomings of blue gas were overcome by the thermal cracking of liquid hydrocarbons into the gas to produce carbureted water gas. This both increased the heating value of the gas and its illuminating power. CWG was a very good fuel gas and was widely produced and distributed to consumers.

The discovery of blue gas is attributed to Fontana in 1780. He passed steam over incandescent carbon and produced a flammable gas. Blue gas was only rarely produced until Lowe's invention of carbureted water gas in 1875. Liquid hydrocarbons were sprayed into the blue gas (carbureted) and thermally cracked to form gases and tars. Carbureted water gas became the predominant form of gas production in the United States and was produced until the demise of the manufactured-gas industry. The production of carbureted water gas was economically possible because of the growth of the U.S. petroleum industry after the 1880's. The petroleum industry supplied the inexpensive hydrocarbon feedstocks required for the production of carbureted water gas. The availability of cheap petroleum-based feedstocks for gas production created a gas industry based on oil instead of coal. The gas industries of Great Britain and Europe did not have cheap oil products and subsequently did not adopt oil-gas and carbureted water-gas production to the same extent as did those in the United States.

Figure 10 is a diagram of a blue-gas generator. Blue gas is produced in a cyclical manner: (1) air is blown through the bed, burning coke and heating the bed; (2) the air is cut off, and steam is blown through the bed, producing
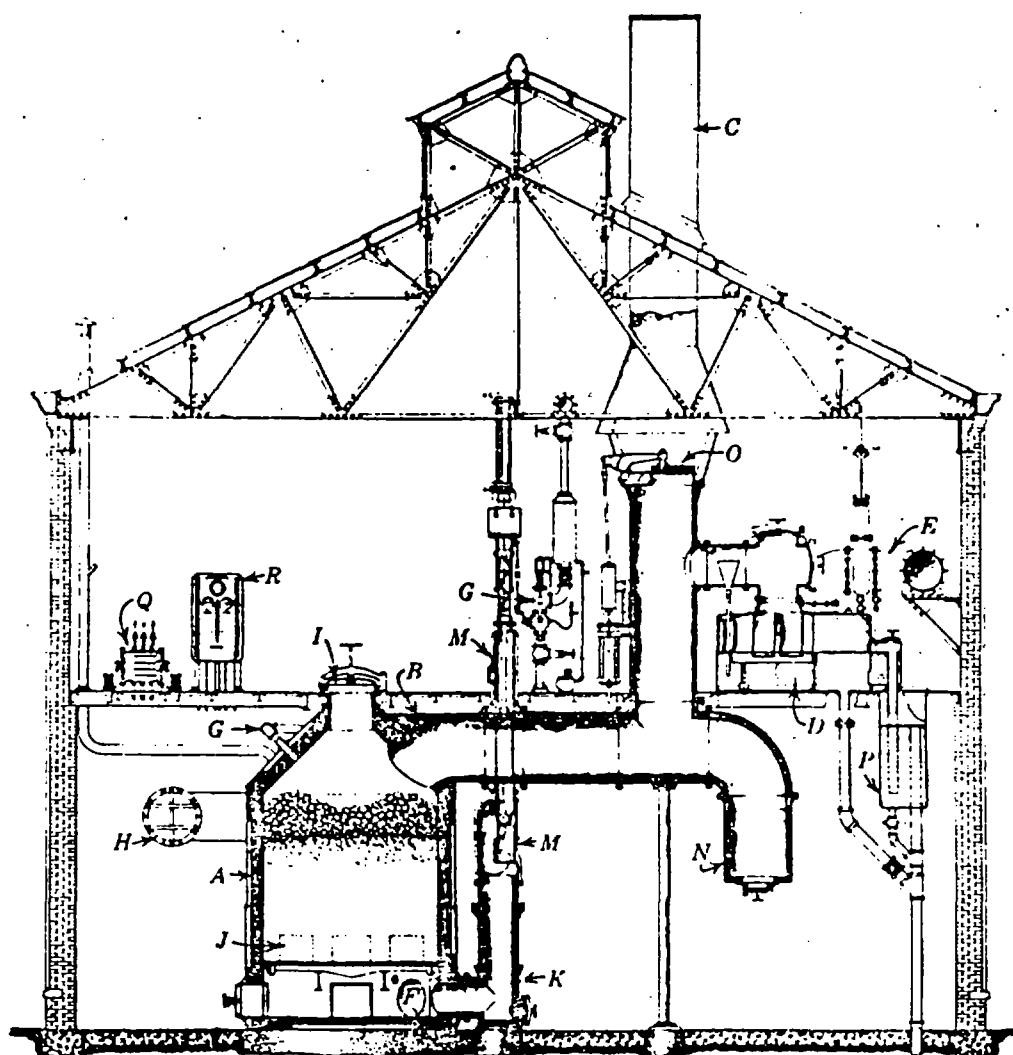
Source: Morgan, 1945.

A	= Generator.	I	= Coaling door.
B	= Gas off-take or	J	= Clinkering doors.
	hydrogen pipe.	K	= Bottom gas off-take.
C	= Stack.	M	= Heat valves.
D	= Wash-box, or seal	N	= Dust catcher.
	separator.	O	= Stack valve.
E	= Hot main connection.	P	= Seal pot or drain tank.
F	= Blast connection.	Q	= Controls.
G	= Steam connection.	R	= Instrument board.
H	= Explosion door.		

Figure 10. Blue gas generator.

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blue gas and cooling the bed; then (3) the cycle is repeated. This is the simplest cycle that can be used to produce blue gas, and variations of this cycle were employed to improve the production of gas. During the "runs" (with steam), carbon monoxide and hydrogen are produced, principally from the water gas shift reaction:

$$H_2O + C = CO + H_2 \ .$$
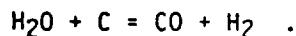
This reaction is endothermic and rapidly cools the coke bed. When the bed has cooled, the steam is stopped and air is blown through the bed ("blow") to reheat the coke. The cyclical process is made more heat efficient by recovering heat in the flue gases during the blow and by preheating the air used in the blow. During the blow, the coke bed tends to form carbon monoxide from incomplete combustion. This gas was similar to producer gas and could frequently be burned when additional combustion air was added. A complete set of the reactions occurring during the blow-and-make periods of blue-gas production is given in Table 5.

A blue-gas producer is the front third of apparatus used to produce carbureted water gas. Figure 11 shows a three-shell water-gas set. The first shell is a blue-gas generator, and the second shell (carburetor) and a third shell (superheater) are attached to it. The carburetor and superheater are checkerbricked with firebricks. The bricks are arranged so that a large surface area of the bricks is exposed to gases flowing through the shell, but with a relatively low pressure drop.

This apparatus was also operated in a cyclical manner, with alternate blows to heat the coke bed and the checkerbrick, followed by runs in which blue gas was produced and hydrocarbons cracked into the gas from oils sprayed onto the hot firebrick of the carburetor. The blow and run parts of the cycle are described below and illustrated in Figures 12, 13, and 14.

Blow: Air is blown through the coke bed to heat the bed. Air enters from the bottom of the bed and flows upward through the coke. Air is admitted to the top of the carburetor, then it burns carbon monoxide in the gas from the generator, supplying additional heat for the checkerbricks. The gases flow downward through the carburetor, then upward through the superheater, exiting from the top of the superheater and flowing to a waste heat boiler.

TABLE 5. REACTIONS DURING BLUE GAS MANUFACTURE

Blow:

$$O_2 + C = CO_2$$
$$O_2 + 2C = 2CO$$
$$CO_2 + C = 2CO$$
$$2CO + O_2 = 2CO_2$$

Make:

$$H_2O + C = H_2 + CO$$
$$2H_2O + C = 2H_2 + CO_2$$
$$H_2O + CO = H_2 + CO_2$$
$$CO_2 + C = 2CO$$

ELEVATION - SECTION A-A
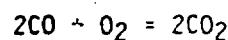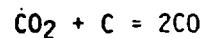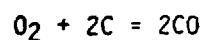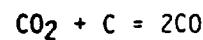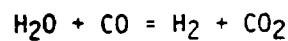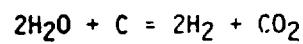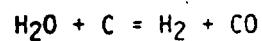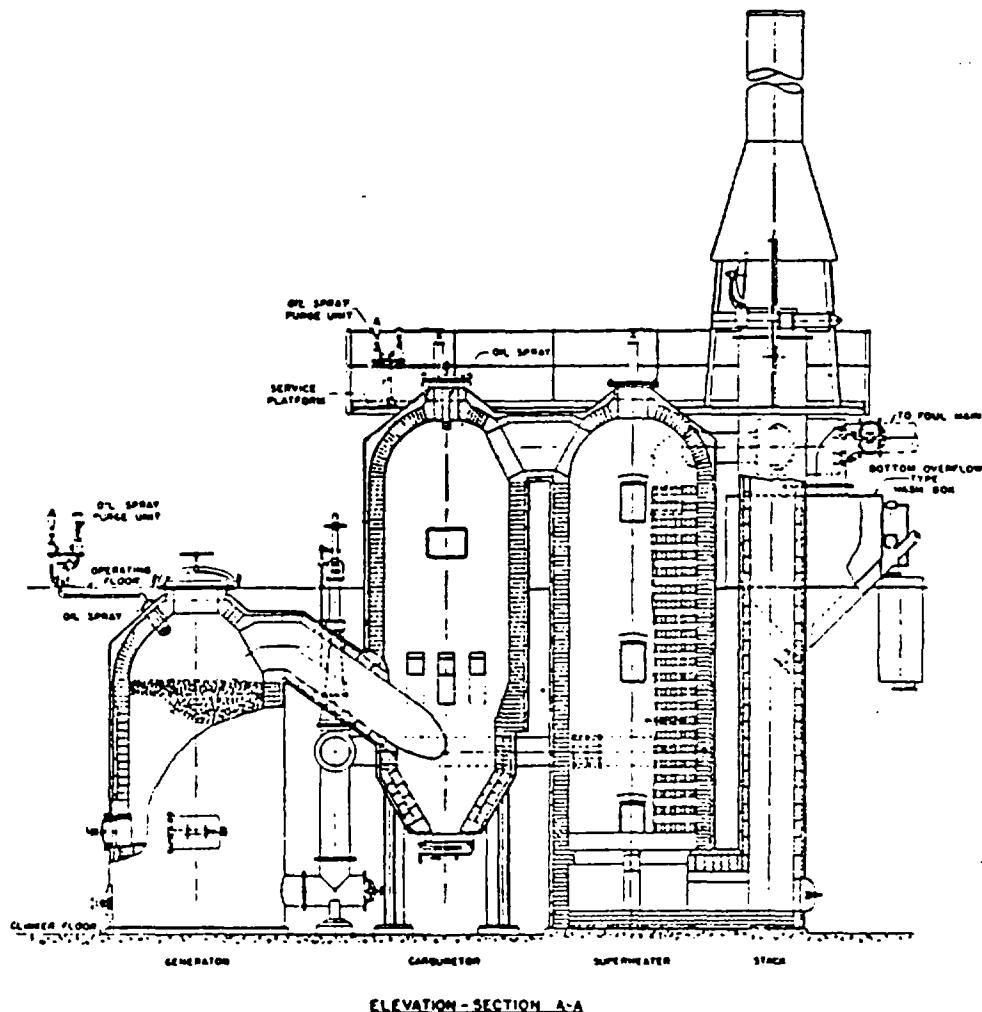
Figure 11. Typical three-shell water-gas set.

Source: Morgan, 1945.

Blow, To Heat Apparatus
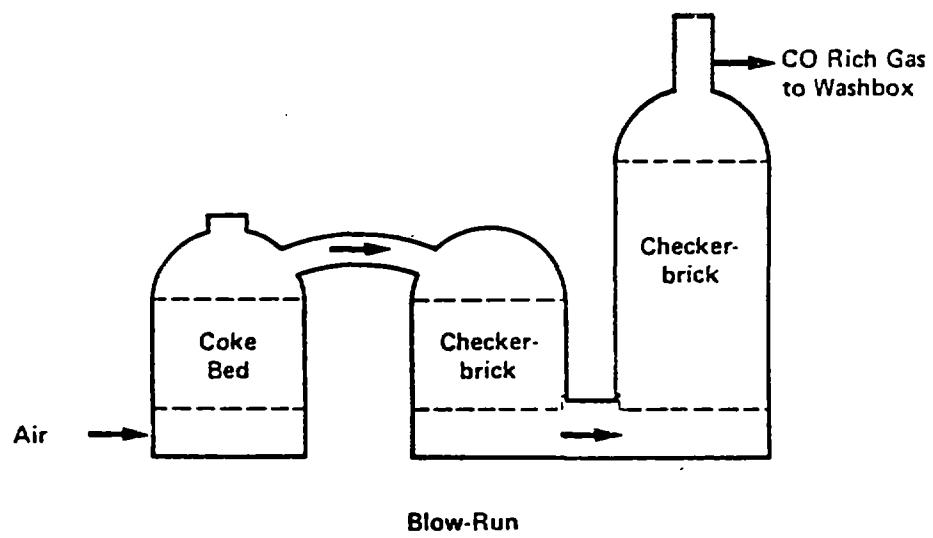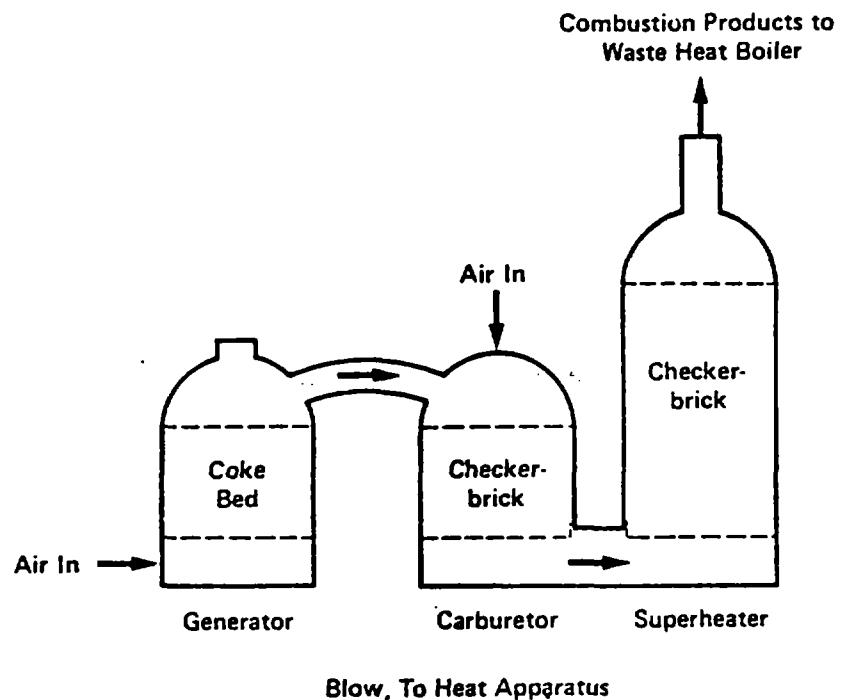
Blow-Run

Figure   12. Blow and blow-run.

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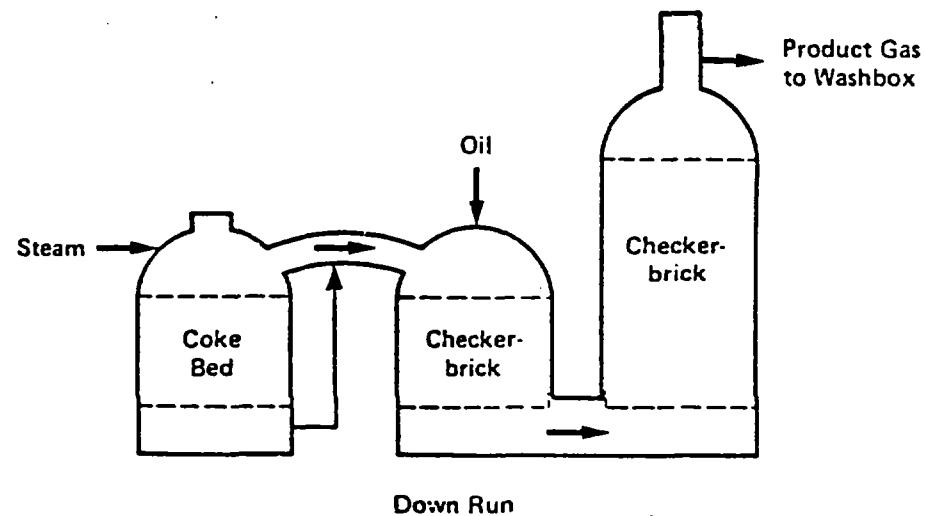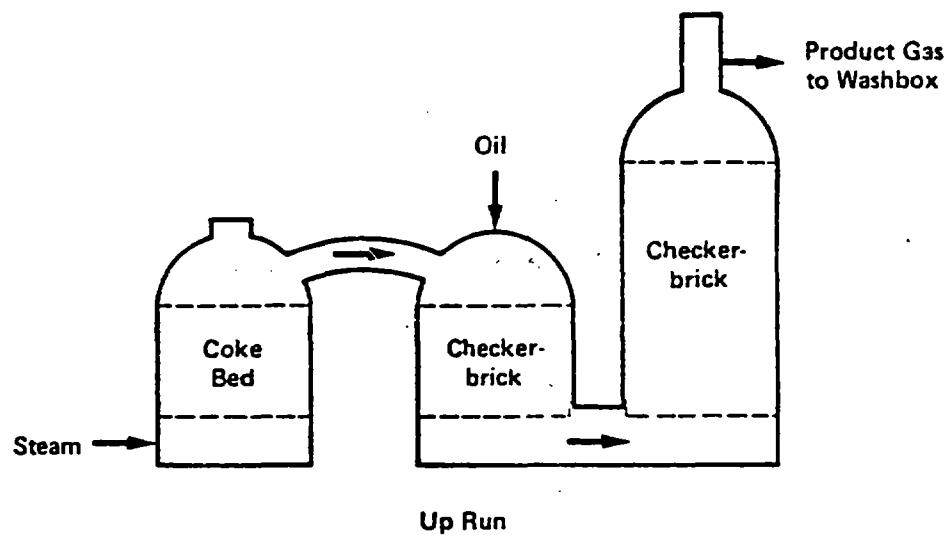
Figure 13. Up and down runs.
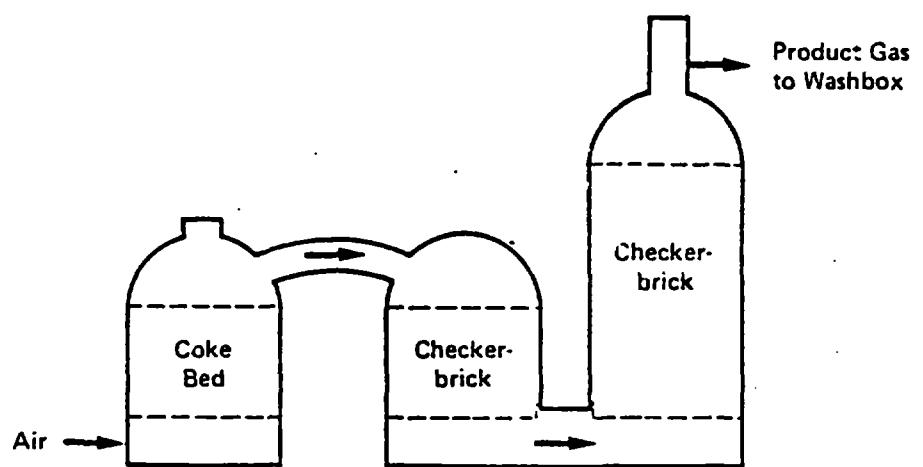
Figure 14. Air purge.

Blow-run: This part of the cycle collects the carbon-monoxide-rich gas from the generator bed and adds it to the product gas. The air flow is the same as that during the blow except no air is added at the top of the carburetor and the gases are routed through the wash-box to the gas mains.

Up-run: During the up-run, steam is admitted to the base of the generator, flows upward through the bed of incandescent coke (form-ing blue gas), out the top of the generator to the top of the carbu-retor (where oil is sprayed into the gas and onto the checkerbrick, cracking the hydrocarbons), down through the carburetor, upward through the superheater (where additional cracking of the hydrocar-bons occur), and out through the top of the superheater and washbox to the gas mains. During the up-run, the bottom of the coke bed cools faster than does the top.

Down-run: The down-run (or back-run) is identical to the up-run except that steam is introduced at the top of the generator bed, flows down through the bed, and then to the top of the carburetor. The top of the bed is cooled during the down-run, maintaining a hot area in the center of the bed. More efficient operation of the generator is obtained with split runs (up and down) than if the entire run were performed in the same direction.

Air purge: The air purge actually starts the blow, but gas from the superheater is sent to the gas mains. This purges the apparatus of higher Btu gases and recovers them as part of the product gas.

Table 6 shows some typical compositions of blue gas and carbureted water gas. The carbureted water-gas process was used to produce gases of widely varying Btu contents. This was accomplished by varying the amount of oil cracked into the blue gas. The specific heating value of carbureted water gas produced by individual companies was determined by economic considerations, but it was usually set between 500 and 600 Btu/ft^3. Higher Btu-carbureted water gas could be mixed with lower grades of gas (producer, blue gas, or coal gas) to produce a mixed gas for distribution. This had the net effect of increasing gas production capacity without increasing the number of water- gas sets used to produce the gas. The highest Btu-carbureted water gas could be mixed with natural gas without reducing the heating value of natural gas (both natural gas and high Btu-carbureted water gas have heating values of about 1,000 Btu/ft^3). The higher heating value comes from increased use of carburetion oils, increasing the cost of the gas.

A variety of feedstocks were used in the production of carbureted water gas, and these raw materials varied both with time and location of individual

TABLE 6. TYPICAL COMPOSITIONS OF BLUE GAS AND CARBURETED WATER GAS

	Blue gas	Carbureted water gas			
Btu/ft^3	287	540	695[a]	850	1,010
Oil, gas/1,000 ft^3	--	3.0	4.2	8.6	13.1[b]
Fuel, lb/1,000 ft^3	34.7	30.0	30.8	32.0	26.5[c]
Steam, lb/1,000 ft^3	61.9	30.0	30.9	19.8	15.0
Carbon dioxide, percent	5.4	3.4	4.3	1.6	4.4
Illuminants, percent	0	8.4	12.6	18.9	27.4
Oxygen, percent	0.7	1.2	0.7	0.2	1.1
Carbon monoxide, percent	37.0	30.0	30.2	21.3	9.1
Hydrogen, percent	47.3	31.7	29.3	28.0	19.9
Methane, percent	1.3	12.2	17.8	20.7	21.8
Ethane, percent	--	--	--	4.3	5.3
Propane, percent	--	--	--	--	0.3
Nitrogen, percent	8.3	13.1	5.1	5.0	10.7
Specific gravity	--	0.64	--	0.69	0.85

SOURCE: Morgan, 1946.

[a] 23.5-candlepower gas.

[b] Heavy oil, 7.6 percent Conradson carbon, specific gravity 13.7 °Baume.

[c] In addition 42 lb of steam per 1,000 ft^3 were used in the top of the generator as carrier steam and 36 lb per 1,000 ft^3 of exhaust steam as superheater cooling steam.

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gas plants. Two types of raw material are required for carbureted water-gas production: (1) a solid carbon material for the generator and (2) a liquid hydrocarbon for the carburetor. Because several petroleum fractions and sources of carbon could be used, the specific feedstocks employed at individual plants were selected based on economic factors.

The original carbon materials used in CWG production were anthracite coal or coke from bituminous coal. Both were considered ideal generator fuels because they had very high carbon and low volatile contents. Consequently, they were very clean fuels to use for blue gas, producing hot fires and little smoke during blows. Increasing prices for anthracite coal after the turn of the century and shortages of coke during World War I encouraged modifications of the standard carbureted water-gas process to allow the use of bituminous coals in the generator. Because bituminous coal was cheaper than coke, many plants replaced the use of coke or anthracite by bituminous coals after the war. The conversion to bituminous coals was not universal because some plants had coking facilities onsite and some difficulties occurred with the conversion. The use of bituminous coals reduced the gas production capacity of carbureted water-gas apparatus, entrainment of coal from the generator into the carburetor occurred, and smoke was frequently produced during the air blows of the gas cycles. Some of the problems were reduced by modifying the operation of the sets, primarily through the "pier" process and the use of reversed air blasts through the carburetor and superheater during blows.

Table 7 presents an analysis of fuels frequently used in the generator for the production of blue gas and water gas. The use of raw bituminous coal instead of coke or anthracite introduced some coal constituents into the tars and waste liquids of CWG plants. Coke and anthracite coals have very low volatile contents, and tar acids (phenols), tar bases, and cyanides were produced in only trace amounts from CWG when these generator fuels were used. When bituminous coal was used, the coal actually coked within the generator, releasing coal gas and volatile constituents into the product gas. About 58 percent of the coal gas from the bituminous coal was added to the carbureted water gas, while the remainder was burned during the blows (Murdock, 1926). About 8 percent of the final product gas was from coal gas, and the amount of tar acids, tar bases, and cyanides produced would also be about

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TABLE 7. ANALYSES OF FUELS FOR MANUFACTURE OF BLUE GAS AND CARBURETED WATER GAS

Kind of fuel	Moisture as received	Analysis on dry basis				Btu per pound	Ash fusion point (°F)	Size of fuel (inches)
		Volatile matter	Fixed carbon	Ash	Sulfur			
Anthracite, Pittston broken coal	4.61	6.07	83.94	9.99	0.91	--	2,600+	3 7/16 to 4 7/16
Anthracite, broken	3.30	5.23	81.74	13.03	0.91	13,042	--	--
Anthracite	2.77	5.44	84.19	10.37	0.86	12,836	--	--
Horizontal and inclined retort coke	10.09	1.91	87.92	10.17	0.73	12,746	--	--
Byproduct oven coke	3.13	1.99	89.17	8.84	0.63	13,081	--	--
Water gas coke	1.67	2.21	87.32	10.47	1.11	13,004	2678	--
Spokane gas house coke	1.30	8.3	70.3	21.3	--	11,150	--	--
Denver gas house coke	--	2.88	79.58	17.54	0.62	11,899	--	--
Boone-Chilton coal	1.82	36.48	58.73	4.79	0.58	14,380	2825	--
Fairmont gas coal, average	1.06	34.67	58.16	7.17	1.00	--	--	3 to 6
Elkhorn gas coal	1.94	37.77	59.11	3.12	0.54	14,750	--	3 to 6
Franklin County, IL	7.95	36.08	53.71	10.21	1.31	--	--	6 x 3 lump
Whatcom County, WA, Sub-bituminous coal	8.01	43.44	37.23	19.33	0.35	10,760	--	Washed pea-size
Portland lampblock briquettes	3.40	9.0	90.7	0.3	--	15,100	--	--

8 percent (per volume of gas manufactured) of that produced from coal carbonization.

Any liquid hydrocarbon that could be thermally cracked into gaseous, liquid, and solid products could be used in the production of carbureted water gas. This included many of the distilled fractions of petroleum oils, but in practice gas-manufacturing companies used inexpensive oil fractions that had only limited alternative markets. As the petroleum industry changed between 1880 and the 1950's, the gas industry modified its use of petroleum products. Table 8 shows the general classes of products distilled from petroleum. The fractions are listed in the order of distillation temperatures, with the lower boiling fractions at the top and the higher boiling fractions toward the bottom of the table. Although any of the fractions could be used in the production of carbureted water gas, three fractions were principally used. These were naphtha, gas oil, and residual fuel oil. Crude oil and "topped" crude oils were also used to a more limited extent.

These fractions each have different distributions of hydrocarbons, and the specific composition of any carburetion oil was dependent on both the source of the original crude oil and its processing during distillation. The carburetion oils differed substantially in their compositions, which in turn influenced the amount of byproducts from the process and the character of the byproducts.

The early carbureted water-gas processes used naphtha fractions of petroleum as the carburetion oil. Naphtha was rich in short-chain alphatic and light aromatic hydrocarbons. It vaporized readily in the carburetor and superheater, with almost all of the naphtha cracking to gaseous hydrocarbons. Tar produced from carbureted water gas using naphtha was 1.7 to 3.5 percent of the original naphtha (McKay, 1901). The early oil refiners produced principally lamp oil (kerosene) and lubricating oils. The naphtha fraction (during this period) was the liquid hydrocarbon fraction that boiled at temperatures above gaseous hydrocarbons and below the kerosene fraction. There was little demand for the naphtha fractions until the invention and use of internal combustion engines. The gas industry used naphtha for the carburetion of water gas and the enrichment of coal gas from about 1880 through World War I, when other uses of naphtha increased the price of this petroleum fraction. As the price of naphtha increased, gas manufacturers

TABLE 8. GENERAL CLASSES OF PETROLEUM PRODUCTS

Source: Biggs and Woolrich, 1925.

switched to other oils, and the use of naphtha ended altogether about 1930 (Dashiell, 1944). Table 9 shows a typical distillation curve for a naphtha fraction, used for carbureted water- gas production in 1897.

Although naphtha was the preferred fraction for the production of carbureted water gas, a fraction boiling between kerosene and lubricating oils was increasingly used after about 1895. This fraction came to be known as gas oil and was a more viscous and heavier petroleum fraction than was naphtha. It also produced more tars in cracking, 12 to 18 percent by volume of the original carburetion oils (McKay, 1901). Table 10 presents a distillation curve for a typical gas oil used for the production of carbureted water gas in 1897. This fraction was the predominant carburetion oil until increased demand for gasoline and the invention of catalytic cracking of the gas-oil fraction into gasoline and residual fuel oil (the heavy residue left from the cracking process). The use of gas oil as a cracking stock for gasoline meant that the price and availability of gas oil was linked to the price and demand of gasoline. Gas-oil supplies became more expensive and less available as the demand for gasoline increased. The gas industry began to switch from gas oil to fuel oils around 1930. The great variability of oils used for the manufacture of carbureted water gas is shown in Table 11. Each oil sample was analyzed and divided into four constituents: aromatics, olefins, parafins, and naphthenes. A similar study of 50 gas-making oils showed the following ranges of properties (Kugel, 1947):

Specific gravity (60 °F)	1.049 to 0.754
Viscosity (100 °F)	27 to 288 S.S.
Flash point	Below 62 to 75 °F
Pour point	Trace to 14.0 percent
Sulfur	0 to 3.7 percent.

As the price of fuel oils increased during the late 1940's, some facilities switched to heavier fuels oils, such as residual oils with high carbon contents. With fuel oils and heavy residual oils, the tar byproducts from the carbureted water-gas process increased to up to 25 percent of the original oil

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TABLE 9. NAPHTHA DISTILLATION

Naphtha					
Specific gravity = 0.6930, or 72° Baume at 60 °F (color white)					
Fraction (°F)	Vol. (÷)	Wt. (%)	Sp. Gr.	Beaumé	Color
100-150	10.90	10.34	0.6579	83°	White
150-200	54.09	53.69	0.6885	73.5	White
200-250	28.00	29.07	0.7196	64.5	White
250-300	4.20	4.45	0.7370	60	White
Above 300	1.75	1.91	0.7560	55	Clear red-brown
Residue	None appreciable				
	98.94	99.46			

SOURCE: McKay, 1901.

TABLE 10. GAS-OIL DISTILLATION

Gas oil

Specific gravity = 0.8462, or 35° 0.3 Baume at 60 °F (color dark)

Fraction (°F)	Vol. (%)	Wt. (%)	Sp. Gr.	Beaumé	Color
180-300	4.40	3.83	0.7369	60°	White
300-350	4.55	4.10	0.7639	53.5	Nearly white
350-400	3.50	3.24	0.7823	49	Nearly white
400-450	6.30	5.95	0.8001	45	Nearly white
450-500	6.95	6.73	0.8108	40.75	Slightly yellow
500-550	10.45	10.27	0.8320	38.5	Pale yellow
550-600	16.35	16.46	0.8491	35	Pale yellow
600-650	21.95	22.17	0.8625	32.25	Pale yellow
650-700	18.35	18.99	0.8764	29.75	Yellow
Above 700	7.50	7.98	0.9009	25.5	Dark
Residue	--	0.30	solid	--	Black
	100.30	100.02			

SOURCE: McKay, 1901.

TABLE 11. TYPICAL CARBURETION OILS

	Spec. Grav. API°	Conradson Carbon %	Sulphur %	Viscosity at 122°F. Saybolt-Furol Seconds	Ash %	Hydrocarbons, %				Carbureting Value by Laboratory Set	
						A	B	C	D	B.t.u./gal.	Temp. °F.
(1) Fuel Oil	12.3	8.2	1.0	83	0.05	14	26	40.8	19.2	85,000	1525
(2) Blend of heavy uncracked residuum, like No. 4, fluxed with low viscosity tar, like No. 3	16	8	.5	300	1.5	8	28	50	14	91,400	1400-1500°
(3) Cracking Plant Tar	3.4	8.4	1.43	21		12	44	25	19	67,600	1594
(4) Straight Run Residue, too viscous to be sold under Fuel Oil specifications	10.7	12.4	0.75	5,000 to 10,000	1.67	10	30	44	16	101,400	1375
(5) Gas Oil of fair quality	24.2	1.6	0.45	13		8	8	63.8	20.2	105,000	1450
(6) Fuel Oil "X"	22.3	4.84	2.17	18		10	16	62	12	104,000	1400-1500°
(7) Fuel Oil "Y"	19.7	5.06	1.67	53		8	24	53	15	104,000	1486
(8) Fuel Oil "Z"	14.7	7.14	0.68	281		10	26	48.6	15.4	96,500	1400
(9) Cracked Gas Oil	22.5	0.14	.26	10	Nil	14	20	44.9	21.1	80,000	1400
(10) Blend of material like No. 4 with material like No. 11, except enriching value is higher	13	11.7	1.03	1500	1.66	10	18	54.5	17.5	111,632	1580
(11) Bunker C Oil Low viscosity	22.3	2.68	0.53	14	.06	12.4	24.0	54.6	9.0	101,000	1430-1570°
(12) Gasoline—											
Cracked	58.5	Nil	0.03		Nil	16	22	54.6	7.4	102,957	1514
Straight Run	65	Nil	0.03		Nil	8	4	81.8	6.2	116,900	1450
(13) Heavy Naphtha	38	.18	.27		.02	16	30	51.3	2.7	98,935	1604
(14) (a) Gasoline	58.2	Nil	Trace	348	Nil	10	6	76.4	7.6	124,217	1326
(b) Bunker C	15.4	8.4	.70	(Over 10000 at 33°F.) Too low	.72	12.4	29.2	52.1	6.3	108,000	1465
(c) Mixture—27.5% (a) 72.5% (b)	24	6.5	.56	(253 at 33°F.)	.55	12	22.4	59.6	6	112,000	1305-1400

Source: Dashiell, 1944.

A – Aromatics; B – Olefins; C – Paraffins; D – Naphthalenes

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fed to the process. Larger plants, which consumed large quantities of oil, changed to less expensive oil types faster than did the smaller plants. Changes in oil type were accompanied by changes in the production apparatus and operating procedures of the plant, and these costs were better absorbed by larger production plants.

The major byproduct from the production of carbureted water gas was the uncracked portion of the liquid hydrocarbons fed to the carburetor. This tar was produced in varying amounts from the process, and both the amount of tar produced and its characteristics were dependent on the original hydrocarbon feed material and the operation of the gas apparatus. These tars contain many of the compounds that are present in coal tar, but they contain no tar acids (phenolic compounds) and only traces of coal nitrogen compounds. The use of bituminous coals as a generator fuel increased the amount of these compounds in the water-gas tar, but they are still present in relatively small amounts when compared to coal tars. Because of the generally small nitrogen content of coke and petroleum products, very small amounts of ammonia and cyanide appeared in the gas from carbureted water-gas operations, and this is reflected by low concentrations of these compounds in byproducts.

1.2.4 Oil-Gas Production

1.2.4.1 Introduction--

The production of carbureted water gas required only two raw materials, carbon and an oil. Transporting coal or coke to certain areas of the United States (mainly the Pacific Coast) was expensive for the gas companies. States along the Pacific Coast had ample supplies of inexpensive oil products after 1890, but coal materials had to be transported from the East. This led to modifications of the water-gas process that eventually eliminated the need for coal or coke in the generator. The production of gas from oil was invented in England in 1815, and gas from whale oil was produced in some U.S. cities in the early 1800's (see Section 1.2.5). It was L. P. Lowe, the son of the inventor of carbureted water gas, who invented an oil-gas process using refractory material in 1889. Ten years passed before the first "modern" oil-gas plant was constructed in California, and it was 1902 when an oil-gas plant was installed in Oakland, California, for lighting purposes.

In addition to the production processes used on the Pacific Coast, several other oil-gas processes were used for the production of gas. When natural gas became available to manufactured-gas areas, some carbureted water-gas facilities were converted to high Btu oil-gas production. The high Btu oil gas had a heating value very close to that of natural gas, and it could be either mixed with natural gas for peak loads or substituted for natural gas when natural gas supplies were interrupted. Other relatively minor oil-gas processes were used to provide gas for institutional structures that could not receive gas service from larger sources. This section is divided into two sections; the first covers the California oil-gas processes, and the second describes the high Btu oil-gas processes. The other minor oil-gas production methods are included with the miscellaneous gas production methods at the end of the section.

1.2.4.2 Pacific Coast Oil-Gas Processes--

The major oil-gas process used in the United States was the Pacific Coast oil-gas process that was based on the gasification of oils and steam by passing them through a chamber of heated checkerbricks. The process is cyclical, with alternate heating and gas-manufacturing parts of the cycle. Equipment for the Pacific Coast oil-gas process were of two main types--the single-shell apparatus and the two-shell apparatus. Figure 15 shows the single-shell apparatus, and Figure 16 shows the two-shell or "improved oil-gas apparatus."

The single-shell heat-up and make-down type (also know as a straight shot generator) was heated with air and oil injected into the base of the apparatus, passing through the checkerbrick, and exiting at the top of the shell. Gas was produced by injecting steam and oil at the top of the apparatus, which passed through the checkerbrick and exited at the base of the apparatus.

The single-shell heat-down and make-down apparatus injected oil and air for heating (and oil and steam for gas manufacture) at the top of the apparatus, with stack and product gas removed from the base of the apparatus. This type of process was used in small plants because it had a very low capital cost.

The two-shell apparatus placed two generators in series and achieved better utilization of heat and carbon in the generators than did the single-shell designs. This design permits more efficient heating of the generators by using primary air and secondary air in the generators.

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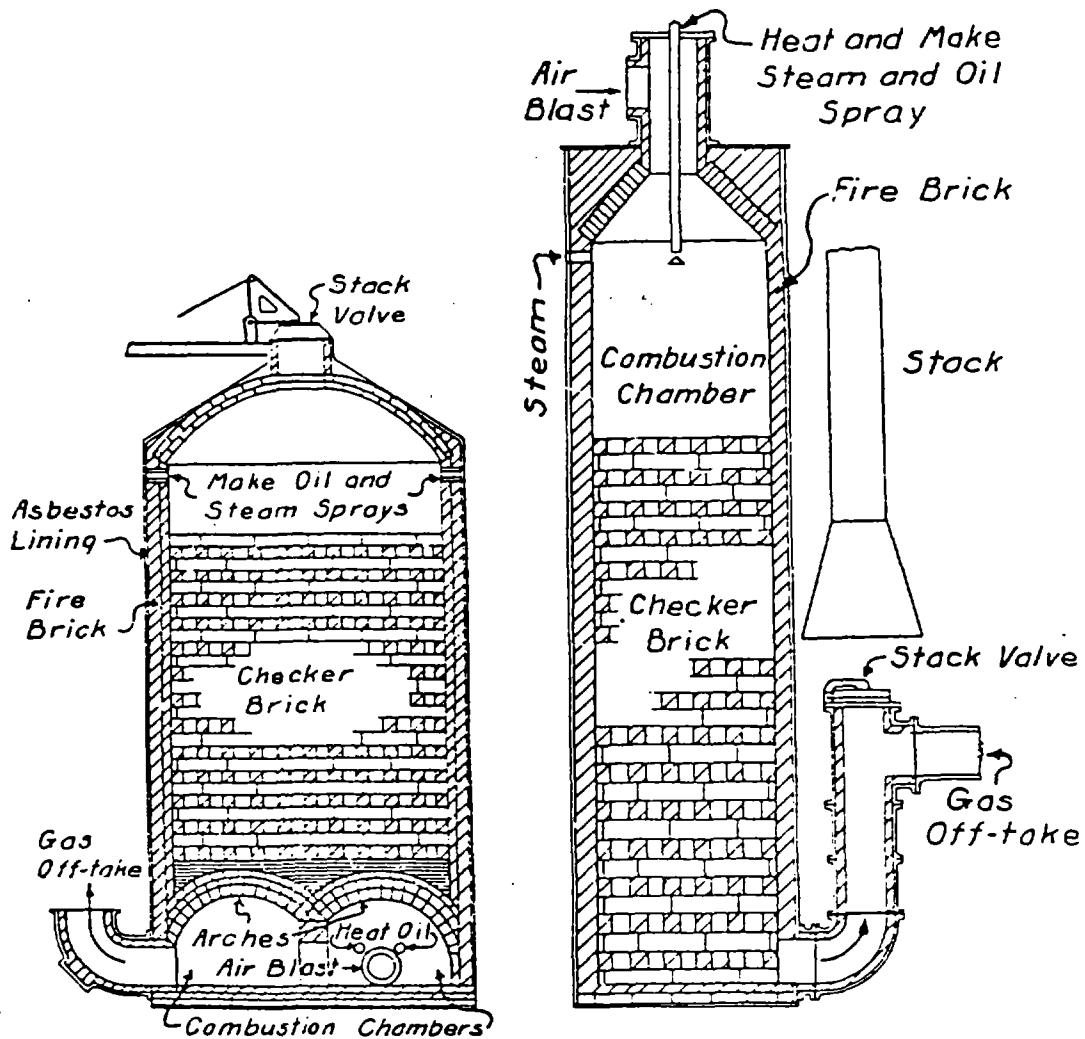
Figure 15. Single-shell oil-gas apparatus.

Source: Morgan, 1926.

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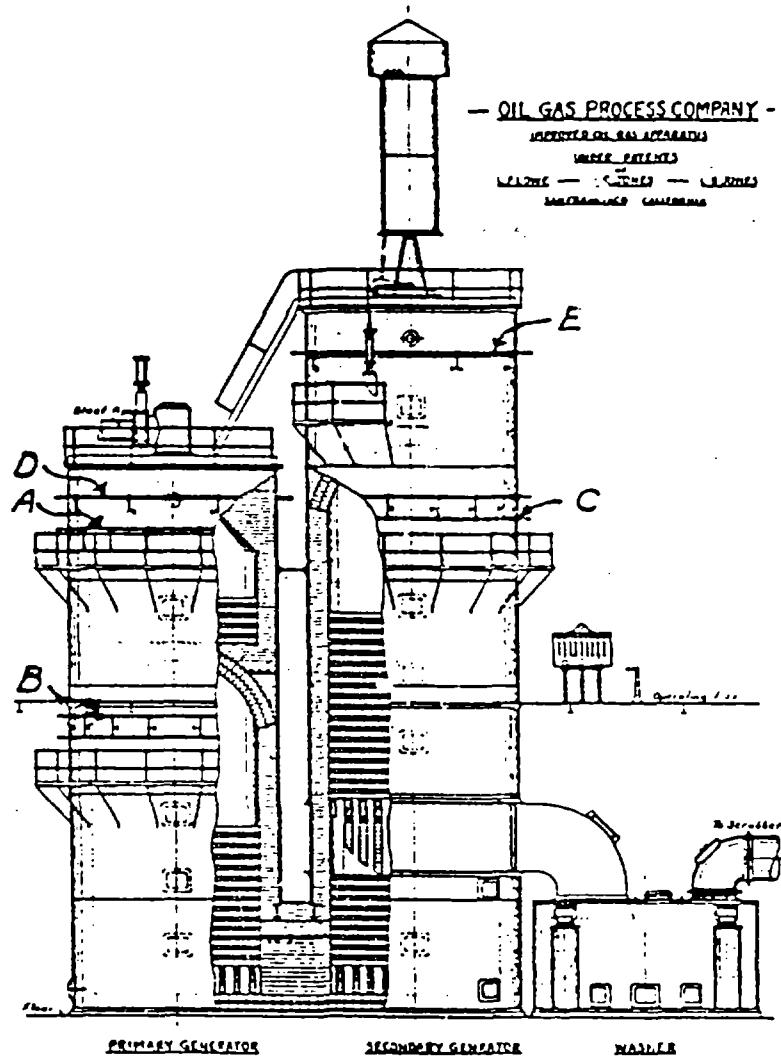
Figure 16. Two-shell, improved oil-gas apparatus.

Source: Morgan, 1926.

Table 12 shows representative operating cycles for each type of oil-gas apparatus. Each part of the cycle is described below.

Blow with air: Air is blown through the apparatus to burn off carbon on the checkerbrick, heating the apparatus. The combustion products are vented to the stack or a waste heat boiler.

Heat with air and oil: Air and oil are turned on and sprayed into the generator. The combustion of the oil heats the checkerbrick to a temperature of about 1,600 °F.

Make with steam: The air and and oil are shut off and steam is fed to the apparatus. The steam cools the bricks somewhat as it reacts with carbon on the bricks to form blue gas. The blue gas is sent through the washbox and mixed with product gas. The cooling of the bricks is desirable because high temperatures cause excessive carbon formation, and the highest brick temperatures occur at the combustion chamber. This part of the cycle is used with the single-shell heat and make-down apparatus and the two-shell apparatus. It was generally not employed with the straight shot apparatus.

Make with oil and steam: After the apparatus is heated, the high-pressure steam is used to atomize the oil. The stack valve is shut, and the valve to the gas take off is opened so that the product gases will be collected. The atomized oil and steam becomes a mixture of gaseous hydrocarbons, fixed gases, tar, and lampblack. The gas mixture leaves the generator and flows through a water-sealed washer.

Steam purge: After the make cycle, the apparatus is purged with steam to remove combustible gases from the apparatus. The gases from the purge are mixed with the product gas. The purge is necessary to prevent the formation of flammable mixture within the apparatus when air is admitted as the next step of the cycle.

Table 13 shows a comparison of operating data for five oil-gas facilities. The two-shell apparatus (Jones) results are from two plants, and the single-shell straight shot apparatus results are from three plants. All of the plants were located in California. The two-shell process produced substantially less lampblack than did the straight shot process (12.5 vs. 21.2 lbs/Mcf gas), while the straight shot process produced less tar (4.3 vs. 1.8 lbs/Mcf gas).

The major byproducts from the oil-gas process are lampblack, tar, and light oil. As in the carbureted water-gas process, only very small amounts of ammonia, cyanides, tar bases, and tar acids (phenols, creysols) are produced. The major difference between the byproducts from oil-gas manufacture and those

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TABLE 12. OPERATING CYCLES OF OIL-GAS PRODUCERS

	Heat up make down	Heat down make down	Two-shell
Blow with air (min)	3	2	5
Heat with oil + air (min)	9	8	5
Total heating (min)	12	10	10
Make with steam (min)	0	2	1
Make with oil + steam (min)	16	6	7
Purge with steam (min)	8	2	2
Total making period (min)	24	10	10

SOURCE: Morgan, 1926.

TABLE 13. COMPARISON OF FIVE OIL-GAS PLANTS

Weight in Pounds[1]

Items	Jones		Single-shell up blast		
	Potrero	San José	Santa Barbara	Southern California Gas Company	Los Angeles Gas and Electric Corp.
Weight of Materials into Generator					
Make period					
Make oil	50.48	51.03	58.51	57.94	59.06
Steam	24.80	23.50	14.20	12.20	10.50
Steam (purge)	6.40	3.50	2.00	7.60	6.00
Combustion products	3.30	5.30	5.40	5.30	3.80
Totals	84.98	83.33	80.11	83.04	79.38
Blast period					
Air	112.00	44.00	86.00	48.00	76.00
Steam	1.50	1.20	0.80	0.80	0.90
Totals	113.50	45.20	58.80	48.80	76.90
Heat period					
Air	97.00	129.00	140.00	119.00	153.00
Oil	6.88	7.30	8.08	5.23	8.07
Steam	5.90	5.00	4.00	2.70	3.00
Totals	109.78	141.30	152.08	126.93	164.07
Totals of all materials in	308.26	269.83	320.99	258.77	320.33
Weight of Materials out of Generator					
Make period					
Purified gas	35.77	37.06	29.77	31.80	33.25
Lampblack	12.00	13.00	22.00	19.60	21.70
Tar	4.00	4.50	2.50	1.50	1.50
Water vapor	22.40	17.60	12.80	14.70	9.00
Totals	74.17	72.16	67.07	67.60	65.45
Blast period					
Stack gases	112.00	42.00	86.00	48.00	77.00
Water vapor	1.50	1.20	0.80	0.80	0.90
Totals	113.50	43.20	86.80	48.80	77.90
Heat period					
Stack gases	94.00	122.20	129.60	114.00	149.40
Water vapor	13.05	12.33	11.16	7.83	10.35
Totals	107.05	134.53	140.76	121.83	159.75
Totals of all materials out	294.72	249.80	294.63	238.23	303.10
Difference (including carbon deposited on bricks, H₂S, naphthalene, and losses)	13.54	19.94	26.36	20.54	17.23
Per cent difference	4.4	7.3	8.2	7.9	5.4

[1] From Final Report of the Joint Committee on Efficiency and Economy of Gas of the R. R. Commission of the State of California.

Source: *Gas Engineers Handbook*, 1934.

from water-gas manufacture is the large amount of lampblack (petroleum coke) produced from the oil-gas process. This lampblack was deposited in the wash-boxes or scrubbers of the plant and was disposed by burning, briquetting and sale, or dumping. The generators also required frequent rebuilding when they became clogged with carbon.

The fuels used for the Pacific Coast oil-gas process came principally from the oil fields in southern California or from the processing of the California crudes. This crude oil had an asphaltic base instead of the paraffinic-based crudes of Pennsylvania. The raw crude oil was used directly for oil-gas production until about 1919, when "topped" crudes or residual oils started replacing the raw crude oil. Topped crude oils were those in which the more volatile and valuable fractions were distilled from the crude, leaving a residual fuel of higher boiling components and a high carbon content. Rather than continuing to distill the residue to heavy asphalts and coke, the refiners sold the residue to gas companies, which used it for the manufacture of oil gas. Table 14 shows the distillation curves for a typical California crude and a refinery residuum. The crude oil would have been a much better feedstock for the manufacture of oil gas, in that the lower boiling components would be readily cracked into the gas, while the residuum would produce much larger quantities of lampblack and require more oil to produce gas comparable to that produced from the crude oil. Because the residuum was less expensive than the crude oil, gas manufacturers preferred the use of residuum oil.

1.2.4.3 High Btu Oil-Gas Processes--

The introduction of natural gas to areas previously served by manufactured gas brought substantial changes in the operations of the manufactured-gas companies. Initially, the gas pipelines installed in manufactured-gas regions were for base capacity. The purchasing gas company was required to buy a fixed amount of natural gas from the pipeline, with financial penalties for using more gas than originally contracted. Consequently, natural gas was purchased for the base load of the gas company, i.e., the amount of gas used everyday by the gas consumers. The gas company then had to provide whatever additional gas was required to meet peak demands of the population they served. This meant either storing large quantities of natural gas to smooth

TABLE 14. DISTILLATION OF OILS COMMONLY USED FOR OIL GAS

| Temperature (°C) | Percent of distillate by volume | |
	Crude oil	Residuum
Up to 150	8.5	--
150-200	13.5	1.3
200-250	15.0	5.7
250-300	20.0	11.9
Total to 300	57.0	18.9
300-332	35.7	--
300-350	--	15.1
350-400	--	35.2
400-407	--	22.4
Residue	7.3	8.4
Gravity, Baume at 60°F	18.4	18.0

SOURCE: Morgan, 1926.

out the peaks and valleys of demand or manufacturing whatever gas was required in excess of the purchased natural gas.

When gas companies switched from manufactured gas (Btu content of approximately 550 Btu/ft^3) to natural gas (Btu content of about 1,000 Btu/ft^3), virtually every gas appliance had to be readjusted for the higher Btu fuel. Only two types of gas had heating values in the same range of natural gas and could be successfully mixed with natural gas in peak demand periods--a high Btu oil gas (approximately 1,000 Btu/ft^3) and LP gas. LP gas was the the distilled petroleum fraction that is a gas at atmospheric pressures and temperatures, but it could be stored as a liquid under pressure and was vaporized into the gas distribution system when needed. It contributed no byproducts or wastes at sites where the process was used.

Existing apparatus for the production of carbureted water gas were frequently converted for the production of high Btu oil gas. This allowed the gas companies to produce a manufactured gas for mixing with natural gas during peak loads and a plant that could provide manufactured gas whenever the natural gas supplies were interrupted. Because the conversions did not involve the purchase of additional equipment, it was a cost-effective method of providing gas for peak loads.

The simplest conversion of carbureted water-gas apparatus for the production of high Btu oil gas was the refractory screen oil process. This conversion consisted of replacing the coke in the generator of the water-gas apparatus with a high-temperature refractory brick and adding additional oil sprays and oil-handling equipment. Figure 17 is a diagram of the converted apparatus. The apparatus is operated in a manner similar to the Pacific Coast processes, with a 3- to 6-minute cycle. This process was successfully demonstrated with a wide variety of hydrocarbon feedstocks with up to 16 percent carbon and between 10 and 39° A.P.I. (Johnson, 1932). Table 15 shows the results of the process when using fuel oil and gas oil. All of the tars produced by the process could be recycled back into the process, reducing the overall fuel requirements. The refractory screen oil-gas process involved a minimum modification of existing carbureted water-gas apparatus and could produce high Btu oil gas for peak loads at relatively low costs to the gas companies.

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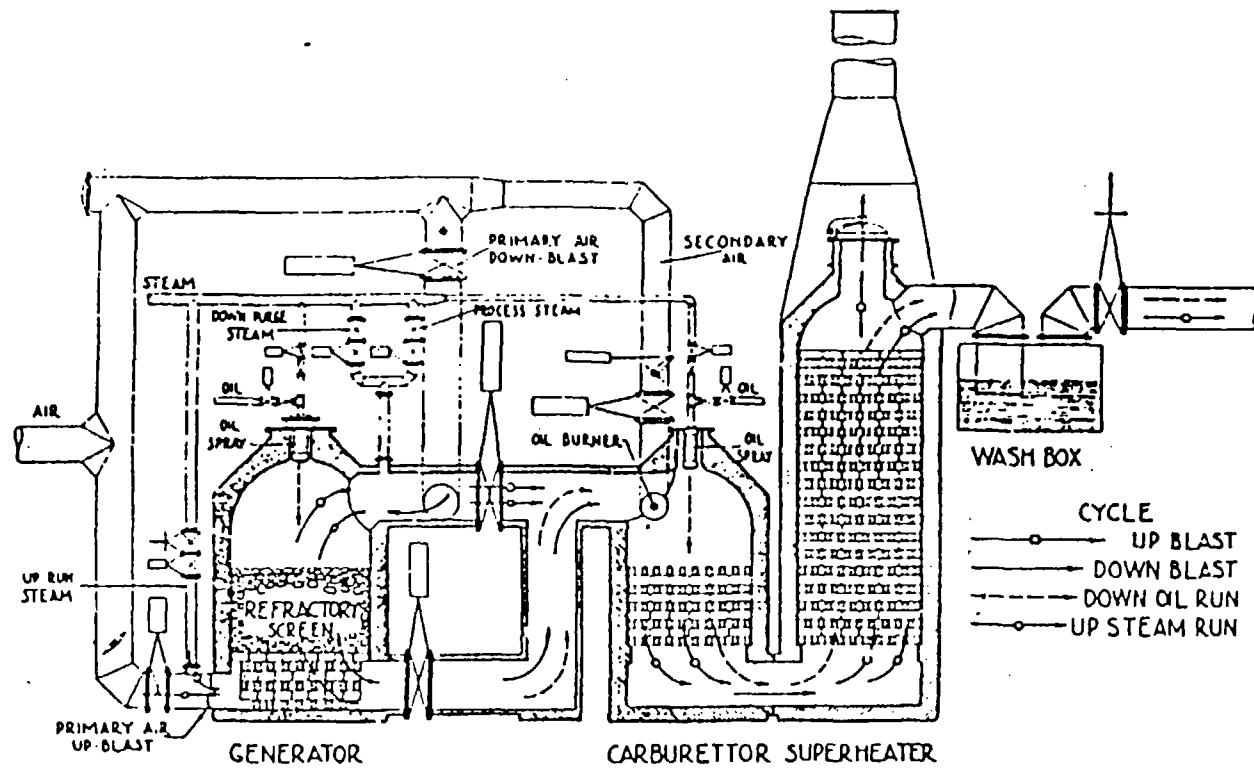
Figure 17. Refractory screen oil-gas process.

Source: Johnson, 1932.

TABLE 15. RESULTS OF REFRACTORY SCREEN OIL-GAS PROCESS

Heating Value—B.t.u. per cu. ft.	1100	1000	900	800
Specific Gravity	0.740	0.682	0.631	0.579
Gas Analyses				
Carbon Dioxide—Per cent	2.0	2.4	2.6	2.9
Illuminants	28.6	24.2	20.1	15.9
Oxygen	0.4	0.4	0.4	0.4
Carbon Monoxide	5.4	6.6	7.7	8.8
Hydrogen	27.7	32.0	36.2	40.4
Methane	28.5	27.0	25.2	23.7
Ethane	4.9	4.4	3.8	3.4
Nitrogen	2.5	3.0	4.0	4.5
Naphthalene—Grs. per 100 cu. ft.	1.39	...	...	
Hydrogen sulphide—Grs. per 100 cu. ft.	80	...	...	
Organic sulphur—Grs. per 100 cu. ft.	2.9	...	...	
Oil requirements—No tar return				
Fuel Oil—Gals. per MCF	4.63	5.17	5.72	6.18
Gas Oil—Gals. per MCF	7.60	6.25	4.95	3.60
Total Oil—Gals. per MCF	12.23	11.42	10.67	9.78
Oil requirements—With tar return				
Total Oil—Gals. per MCF	10.3	9.7	9.2	8.5
Steam—lbs. per MCF	32.0	36.7	40.7	44.5
Tar—Gals. per MCF	1.95	1.74	1.54	1.30
Overall Thermal Efficiency	75.5	72.6	69.2	67.0

Basis of Figures:
Fuel oil—12-18 deg. A.P.I. Cracked Mid-Continent Residuum—0.7 per cent sulphur
Gas oil—34-38 deg. A.P.I. Pennsylvania gas oil

Source: Johnson, 1932.

Another adaptation of carbureted water-gas apparatus was the Hall high Btu oil-gas process. It utilized the carburetors and superheaters of two adjacent carbureted water-gas sets to form a single oil-gas set. Figure 18 shows the configuration of the equipment of the Hall process. The apparatus was operated with a fairly complex cycle that captured more of the heat created during blows, with resulting increases in thermal efficiency and reduced fuel consumption. Table 16 shows the operating results of the process for five different oils. The light oil recovered from the process was approximately 0.35 gallons/MCF, with its characteristics comparable to that produced using carbureted water gas (Utermohle, 1948a,b and Utermohl, 1948b).

1.2.5 Miscellaneous Gas Production Methods

Besides the three major types of gas production processes (coal, carbureted water gas, and oil), there were several minor processes that were commonly used, principally by small manufacturers. These processes are listed with their uses and waste products in Table 17. These processes were typically employed for the lighting of small towns, hotels, or factories. Because they were, in general, small producers who used processes with minimal wastes, sites using exclusively these processes will probably pose only minimal hazards. The production of rosin gas or whale oil gas was primarily used prior to the discovery of bituminous coal in the United States in 1840.

1.3 MANUFACTURED-GAS CLEANING AND PURIFICATION PROCESSES

1.3.1 Introduction

The raw gas from manufactured-gas processes contained many components that were removed prior to gas distribution. Components that would condense within the distribution system, corrode pipes, or produce noxious gases when burned were removed by various processes. Cleaning and purification processes removed undesirable materials from the raw gas. These processes were employed sequentially, with the gas flowing through the entire purification train prior to distribution.

The processes employed to clean the gas were dependent on the method of gas production and sometimes on the specific raw materials used in gas production. Table 18 shows the general temperatures and impurities in manufactured gases as they enter the purification train. The specific concentrations of
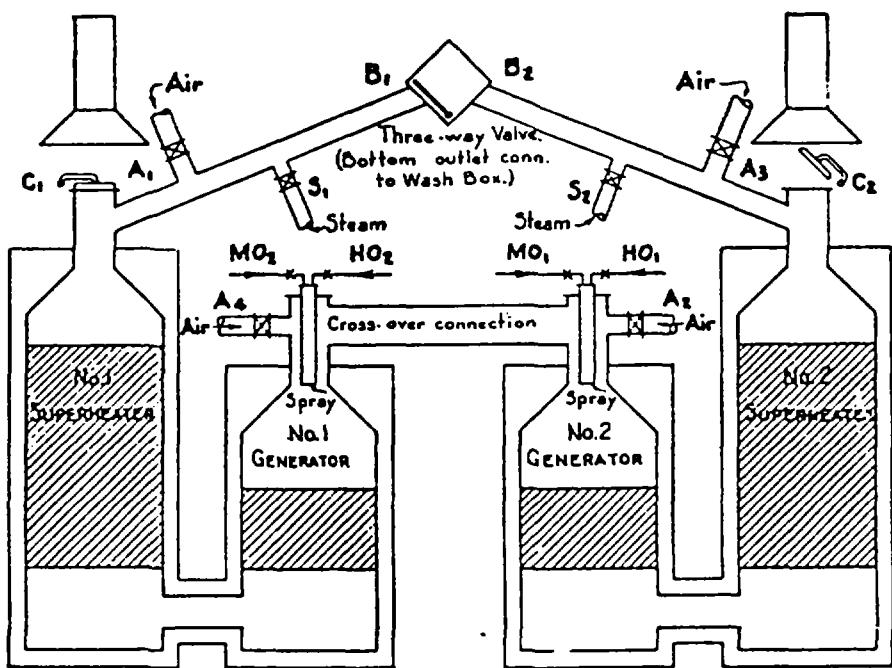
Figure 18. Schematic diagram of Hall oil-gas process.

TABLE 16. HALL OIL-GAS PROCESS OPERATING RESULTS

Oil Analysis

Conradson carbon, %	.20	3.16	6.02	13.03	12.56
Ash content, %	.02	.01	.04	.16	.16
Enriching value, M Btu per gallon (Dick Method) (avg. of 1400, 1500, 1600° F results)	103.6	98.8	102.5	93.8	95.2

Operating Results

Btu of gas per cubic foot	1046	1006	1047	966	974
Specific gravity of gas	.855	.866	.834	.867	.833
Gals. Heat oil per MCF†	.99	.80	.95	.28	69
Gals. Make oil per MCF†	11.50	11.46	10.67	11.69	13.31
Gals. Total oil per MCF†	12.49	12.26	11.62	11.97	14.00
Gals. Tar per MCF†	2.56	2.73	2.30	2.63	*
Gas made per day, MCF†	5,952	5,088	5,088	3,576	3,504
Thermal efficiency, %	79.2	80.9	81.8	80.6	

†Million cubic feet.

TABLE 17.   MISCELLANEOUS GAS PRODUCTION PROCESSES

Gas type	Production method	Gas composition	Residues
Whale oil gas	Pyrolysis of whale oil in heated retorts	Light hydrocarbons, CO, H_2	A waste tar would remain in the retort after gas production. This could be burned or disposed.
Resin gas	Pyrolysis of pine rosin in heated retorts	Light hydrocarbons, CO, H_2	A very heavy, solid tar would remain in the retort after gas reduction.
Gasoline-air gas	Air is bubbled through naptha or gaoline, producing a flammable gas. Burned in special burners with metal gauze to prevent flashback.	Light hydrocarbons	The devolatilized naptha would probably be burned as fuel.
Acetylene gas	Produced from calcium carbide and water $CaC_2 + H_2O = Ca(OH)_2 + C_2H_2$	C_2H_2	The hydrated lime is the only residue.
Pintch gas and Blan gas	Cracking of petroleum oils in iron or clay retort	Light hydrocarbons	Tar.
Dayton gas	Petroleum oil partially combusted	CO_2, hydrocarbons, CO, H_2, N_2	Some tar (0.3 gal/$1,000$ ft^3).

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TABLE 18. TEMPERATURE AND IMPURITIES IN RAW GASES AT OUTLET OF
HYDRAULIC MAIN OR WASHBOX

	Coal gas	Blue gas	Carbureted blue gas	Pacific Coast oil gas
Temperature °F	140-190	160-200	160-200	150-200
Impurities				
Percent by volume				
Water vapor	19-30	32-78	32-78	25-78
Ammonia	1-2	--	--	--
Tar and oil vapors	2-3.5	a	1-2	1-1.5
Parts per million (ppm)				
Cyanogen	1,007-1,410	b	b	--
Naphthalene	3,700-9,300	--	1,490-4,660	2,790-11,200
Hydrogen sulfide	8,000-12,800	1,500-3,200	1,920-4,800	3,200-4,800
Organic sulfur	594-850	b	170-510	340-510

aSmall amounts with bituminous coal.

bNo definite figures available but amounts are small.

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the impurities were dependent on the raw materials used to manufacture the gas (e.g., sulfur content of coal or oil) and the operation of the gas production process. Table 19 shows the types of gas purification processes and whether they were used with specific gas production processes. This section is divided into descriptions of specific purification processes followed by descriptions of general purification systems for coal gas and carbureted water gas or oil gas.

1.3.2 Condensers

After the raw gas leaves the production apparatus, it passes through a water-sealed hydraulic main or a washbox where the gas is initially cooled and some of the heavy tars are condensed and removed. The purpose of condensing the gas is to cool it to ambient temperature and remove all constituents that are not gases. The condenser causes water vapor and tars to condense from the gas and form a liquid, which is then removed from the condenser. Air condensers (condensers that transferred heat from the product gas to air) were the first type employed for the cooling of gas. It was originally believed that slow cooling of the gas allowed more of the illuminants to be retained in the gas and hence be distributed. These condensers were frequently lengths of pipe that zig-zagged across the wall of the retort house.

Water-cooled condensers replaced the air-cooled versions about 1900. These condensers were basically shell and tube construction, with cooling water passing through the shell and the gas flowing through the the tubes. The heat from the gas was transferred from the gas through the tubes and to the water.

Direct cooling (or scrubbing) of the gas by direct contact with recirculated condensate began about 1907 and spread rapidly to both carbureted water-gas plants and coal-carbonization plants. It is also the method currently used for cooling of coke-oven gas. In direct cooling of the gas, it is contacted with cooled recycled water. The water is heated as it absorbs heat from the gas, and additional condensed water vapor and tars are removed in the water. The tars are then separated from the condensate water, the water is cooled, and then reused in the gas cooler. The direct cooling of the gas is usually accomplished in a counter-current packed scrubber, as shown in Figure 19.

TABLE 19. MAJOR GAS PURIFICATION PROCESSES USED WITH PRODUCTION PROCESSES

Gas purification process	Condensers for water removal	Tar removal and recovery	Light oil scrubbing	Ammonia recovery	H₂S removal Lime (before 1898)	Iron oxide (after 1898)	Thylox or liquid scrubbing (after 1927)	Phenol recovery	Additional HCN removal	Lamp-black recovery
Production process										
Coal carbonization										
1. Retorts	U	U	U	S	U	U	S	S	S	R
2. Byproduct coke ovens	U	U	U	U	-	U	S	S	S	R
Producer gas	R	R	R	R	R	R	-	R	R	R
Carbureted water gas										
1. Using naphtha	U	R	R	R	S	U	R	R	R	R
2. Using gas oil	U	U	S	R	S	U	S	R	R	R
3. Using fuel oils	U	U	S	R	S	U	S	R	R	R
4. Using crude oil	U	U	S	R	S	U	S	R	R	S
Oil gas										
1. Using naphtha	U	R	R	R	S	U	R	R	R	R
2. Using gas oil	U	U	S	R	S	U	S	R	R	S
3. Using fuel oils	U	U	S	R	S	U	S	R	R	U
4. Using crude oil	U	U	S	R	S	U	S	R	R	S

U = Usually used.
S = Sometimes used.
R = Rarely used.
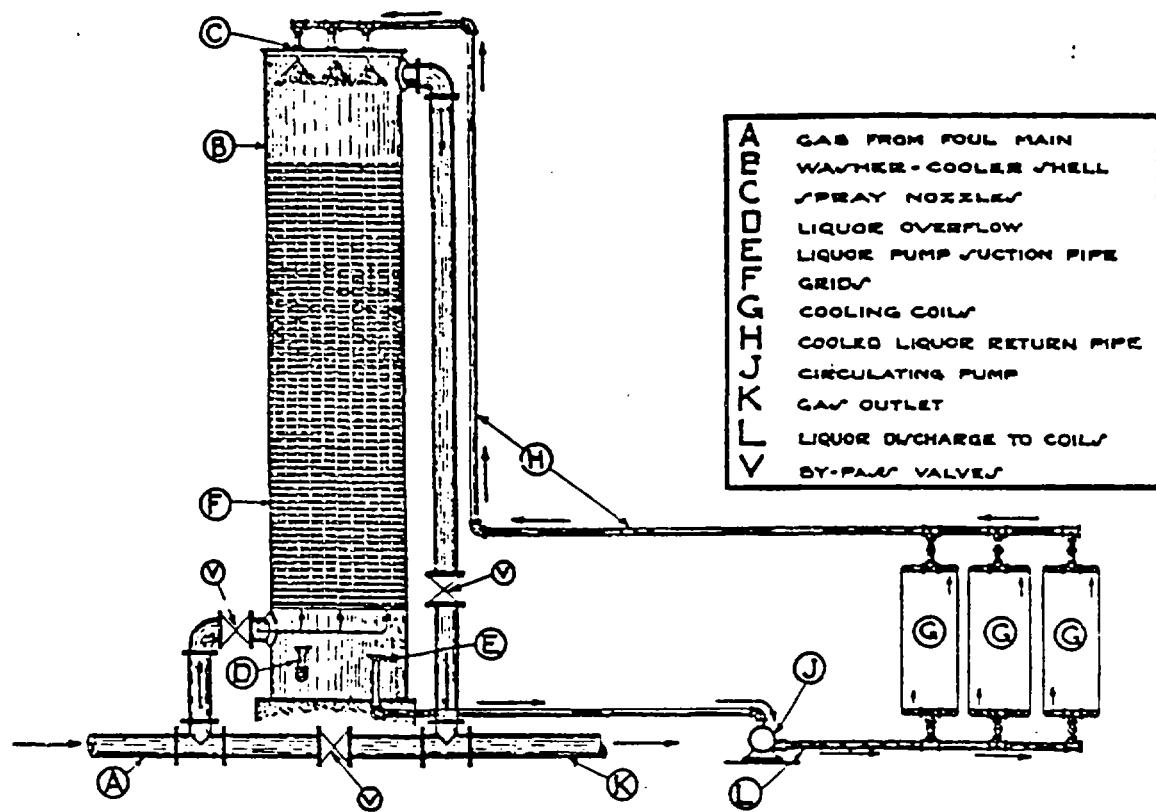
Figure 19. Direct contact cooler.

Source: Morgan, 1926.

The legend in the figure reads:

A	GAS FROM FOUL MAIN
B	WASHER-COOLER SHELL
C	SPRAY NOZZLES
D	LIQUOR OVERFLOW
E	LIQUOR PUMP SUCTION PIPE
F	GRIDS
G	COOLING COILS
H	COOLED LIQUOR RETURN PIPE
J	CIRCULATING PUMP
K	GAS OUTLET
L	LIQUOR DISCHARGE TO COILS
V	BY-PASS VALVES

1.3.3 Tar Removal

Tar is a complex mixture of carbon and hydrocarbons that forms when either petroleum is thermally cracked or coal is carbonized. When raw manufactured gas is cooled, the tar condenses from the gas and usually separates from the condensed water. The distinguishing feature of tars (in the manufactured-gas industry) is that they have a specific gravity greater than 1.0 and sink when placed into water. Organic hydrocarbons that have specific gravities less than 1.0 and float on water are considered oils. Tars were condensed and recovered with condensate at several locations within the purification train. The heaviest tars condensed in the washbox or hydraulic main. The lighter tars were condensed with water from the gas either in indirect or direct condensers. Tar fog (aerosols of tar remaining in the gas after scrubbing) are removed with either a P. and A. (Pelouze and Audouin) tar extractor or an electrostatic precipitator (ESP). Figure 20 shows a P. and A. tar extractor. The gas flows through a pipe in the center of the apparatus, then through several concentric perforated inverted bells. As the gas flows through the perforations in the first bell, the tar aerosols impact on the metal of the second bell, removing the tar from the gas. The counterweight attached to the bells allows the bells to move up and down within the apparatus, exposing more perforations when the gas flow is high and avoiding excessive pressure drops across the extractor. ESP's were introduced about 1924 for the removal of tar fog from gas (Downing, 1934). Figure 21 is a Cottrell ESP. It consists of a steel shell containing vertical tubes. A charged wire runs down the center of each tube. As the gas flows through the tubes, the tar aerosols become charged and impact on the tube walls, removing the tar from the gas. The ESP's were very efficient for the removal of the tar fog, and they were installed on many of the larger coke ovens and carbureted water-gas plants.

A common method for the removal of tar aerosols was the use of shavings scrubbers. These were basically towers or boxes that were filled with wood shavings (or sometimes other materials, such as oyster shells, coke, or slag).

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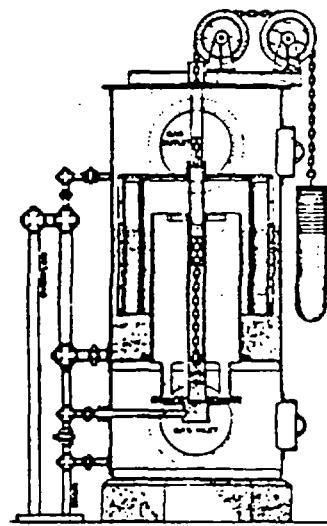
Figure 20. P+A tar extractor.

Source: Morgan, 1926.
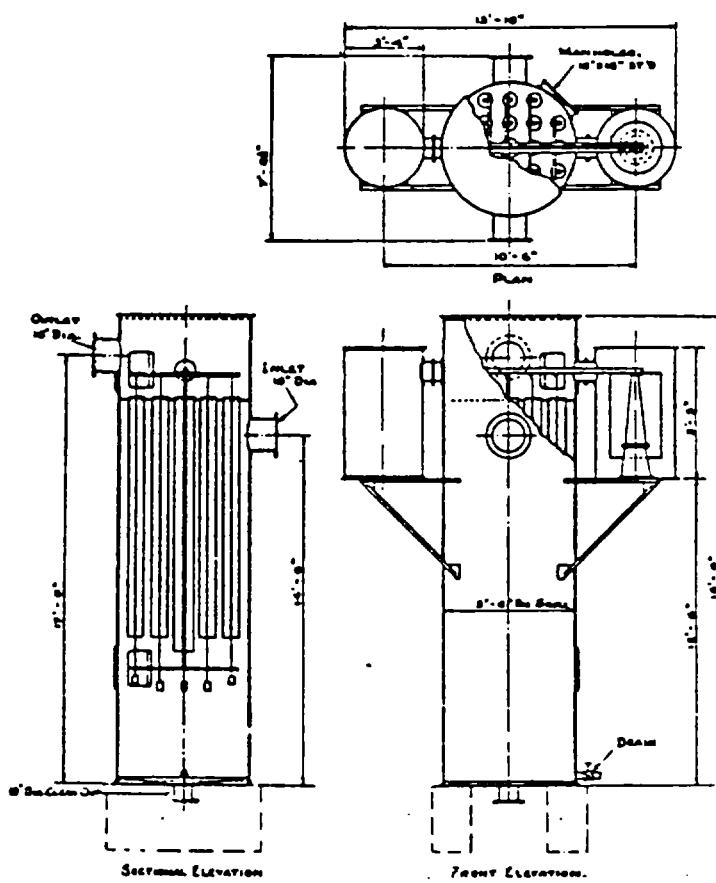
Figure 21. Cottrell ESP.

Source: Morgan, 1926.

The gas would flow through the woodchips, and entrained tar would hit the wood and be removed from the gas. The tar-contaminated shavings would be periodically removed and discarded or burned. The shavings scrubbers were used principally after condensers or scrubbers and removed entrained tar aerosols. The removed tar was prevented from entering the iron oxide boxes, extending the useful life of the iron oxide. Small carbureted water-gas plants and small oil-gas plants were most likely to use shavings scrubbers because their gas production was small and the tar loadings were relatively low.

The tar was usually separated from the condensates by gravity in tar separators similar to that shown in Figure 22. The tar/condensate mixture flows into the separator and separates into three distinct layers by gravity. An oil layer of lighter hydrocarbons floats to the top of the liquid and is retained by oil skimmers. The tar sinks to the bottom of the tank and is removed. Water is the middle layer, and it flows through the gaps in the baffles and exits through the water outlet. The tar separator produces three distinct products, which receive different treatments, depending on the production process. Table 20 lists what was generally done with these three products. Because carbureted water gas and oil gas produce very little ammonia or phenolic compounds, these were not recovered from oil-gas and carbureted water-gas condensates. The oils from the separator were frequently not recovered, particularly with oil and carbureted water gas. In these cases, such oil would be disposed with the condensate.

This type of tar separator had one major problem: The tar, oil, and condensate had to separate relatively rapidly and form the three distinct layers. This usually did not present a problem for coal-gas plants, but carbureted water-gas and oil-gas plants frequently formed oil/tar/water emulsions. These emulsions were relatively stable and were difficult to separate. An emulsion would quickly fill the tar separator, with little or no separation of the tar. The emulsion would then flow out of the separator through both the tar outlet and condensate outlet. In addition to gravity-based tar separators, several other methods were employed for the separation of condensates, oil, and tar. These are listed in Table 21.

In situations where the entire oil/tar/water mixture was disposed instead of recovered, the mixture separated into the three fractions after disposal.

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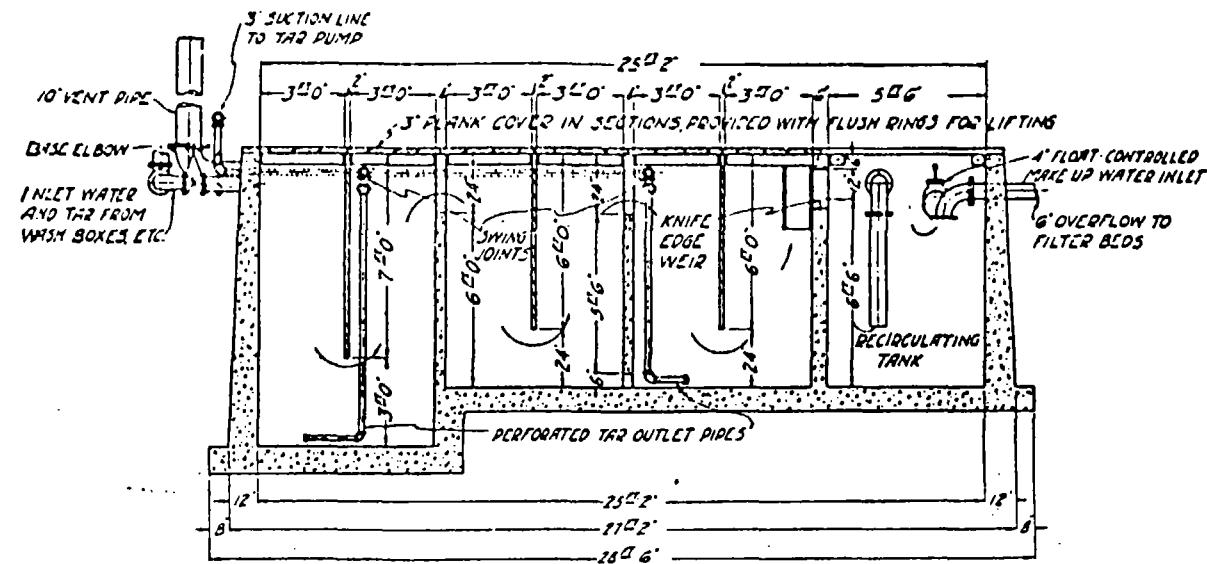
Figure 22. Tar and water separator.

Source: Sperr, 1921.

TABLE 20.   STREAMS FROM TAR SEPARATOR

Stream	Treatment
Tar	Burned as fuel, sold to refiners, distilled on site, mixed with carburetion stocks, or disposed
Oil	Recovered and mixed with light oils, mixed with carburetion stocks, or disposed with condensate water
Water (condensate)	Disposed into stream, treated for recovery of phenols and ammonia (coal gas only), flowed through coke beds prior to disposal, used as coke quench water, recycled to cooler-scrubbers

TABLE 21.   OTHER METHODS OF TAR-WATER SEPARATION

Method	Description
Steam still	Steam is used to distill water from the tar. High cost due to high steam consumption but will handle emulsions containing any concentration of water.
Centrifugal process	Water and tar are separated by density in a centrifuge. Fairly low cost of operation but requires frequent cleaning of tars from the equipment.
Warner tar dehydration system	A modification of the steam still in which water is distilled from the tar.
R.S. de-emulsifying System	Tar-water emulsion is placed into a tank, 30 lb of soda ash is added (for 5,000 gal tank), and the emulsion is heated to 312 °F under pressure. Most emulsions then separate in 30 min - 13 hr. Water is flashed from the tank to cool it to 212 °F.

SOURCE: Seely, 1928.

In rivers or streams, the oil and water would be carried downstream, with some
of the oil depositing on the banks of the stream. The tar fraction would sink
to the bottom of the stream and was usually stopped by fine silts along river
bottoms. In the ground, however, the mixture separates so that oils float on
the groundwater surface, the water soluble components dissolve in the ground-
water, and the tar layer sinks through the groundwater until stopped by a low
permeability layer of ground.

1.3.4 Naphthalene and Light-Oil Scrubbers

After the tars were removed from raw manufactured gases, naphthalene and
light oil were frequently removed from the gas. Naphthalene is a fairly vola-
tile PAH compound, which frequently was not completely removed with the tar.
Naphthalene would crystallize within the gas distribution system, plugging
orifices and reducing flow through pipes. It would often drop out of the gas
as the gas passed through iron oxide purifiers, decreasing the life of the
oxide. The naphthalene could be easily removed from the gas by scrubbing with
a relatively small amount cf a petroleum oil. The naphthalene-enriched oil
could then be either distilled for the recovery of naphthalene or used in the
carburetion of water gas or the production of oil gas. Figure 23 shows a
naphthalene scrubber that consists of two stages: The first stage scrubs the
gas with a recirculated oil, and the second stage uses a small amount of fresh
oil for the scrubbing. The use of two stages allows most of the naphthalene
to be removed in the first stage, with almost complete removal of the naphtha-
lene in the second stage. Used oil from the second stage is added to the
recirculating oil of the first stage. Some of the recirculating oil is con-
tinuously removed. The naphthalene-containing oil from the process was never
considered a waste product, in that the fuel value of the original oil was
enhanced by the naphthalene, and the oil could be either sold or used at the
plant. The naphthalene could be recovered from the oil (if profitable under
market conditions) by distilling the naphthalene-containing oil. Recovered
oil could then be reused in the process.

Any fluid petroleum oil could be used to scrub naphthalene from the gas,
and the most common oils were gas oil and fuel oil. Because the naphthalene
had a large affinity for the oil, relatively low oil flowrates were used for
the removal of naphthalene. Table 22 shows typical operating results for a
naphthalene scrubber.

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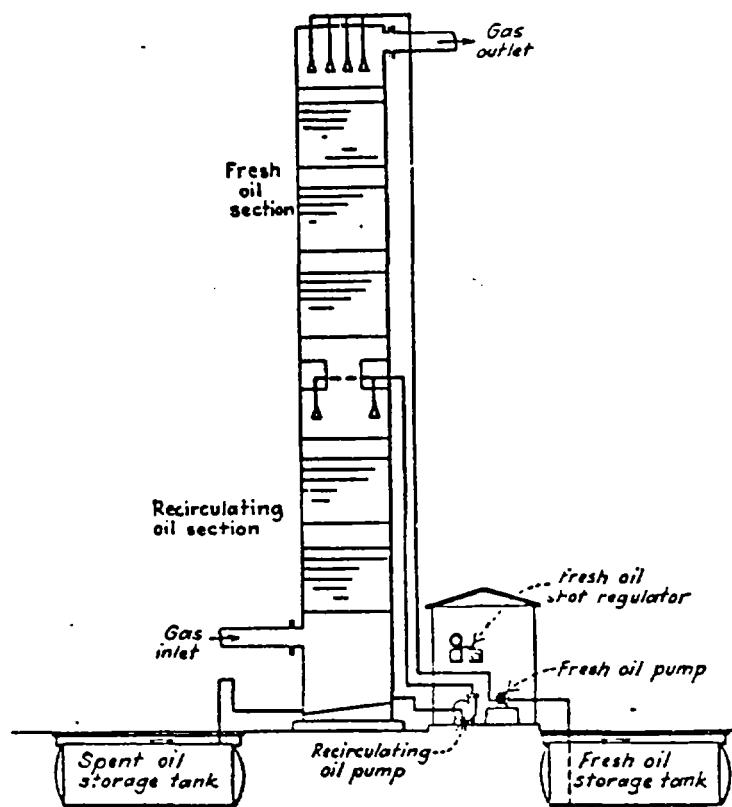
Figure   23. Naphthalene scrubber.

Source: *Gas Engineers Handbook*, 1934.

TABLE 22. RESULTS OF NAPHTHALENE SCRUBBER AT SEABOARD BYPRODUCT
COKE CO., KEARNY, NEW JERSEY

Inlet naphthalene (ppm)	
Max	577
Min	298
Average	436
Naphthalene in outlet gas (ppm)	
Average	69
Oil consumption	17.5 gal/10^6 ft^3
Spent oil	
Specific gravity (22 °C)	0.875
Light oil (to 200 °C)	20.1%

SOURCE: Gas Engineers Handbook, 1934.

Light oil consisted of the light aromatic compounds contained in the gas. They were primarily benzenes, xylenes, and related compounds. These compounds were originally considered beneficial in the gas because they burned with a brighter flame than did other gas constituents. With the invention of the gas mantle and the switch from light to heating standards for gas, the illuminants were no longer necessary for the gas quality. During World War I, the demand for benzene and xylene chemicals increased greatly, and many gas plants began to recover the light oils from the gas. The method of removing light oils from the gas is very similar to that for the removal of naphthalene, except that the light oils were always recovered. (The recovery of light oils was a purely economic decision when the recovered oils were worth more than their heating value in the gas. When not recovered, the light oils enriched the distributed gas and caused no problems in the distribution system.) Figure 24 shows a representative light-oil scrubber. The entering gas is scrubbed counter-currently, first by recirculated oil, then by fresh oil. Spent oil is removed from the recirculating oil and distilled to produce the light oil and regenerated scrubbing oil. A variety of oils was used in the scrubbing of light oil, including gas oil, green oil, fuel oils, tetralin, and lighter tar fractions.

Light oil contains a variety of intermediate boiling hydrocarbons. Table 23 shows a typical analysis of a coke-oven light oil, divided into distillation fractions. Table 24 is a list of compounds commonly found in light oil from coke ovens. Constituents of light oil from oil gas or coke oven gas would have a subset of these constituents, excluding the phenols and base nitrogen compounds. Light oil was used as a feedstock for the production of benzene, toulene, xylene, and other organic chemicals, or it was mixed with gasolene to increase its octane. A complete history of light-oil recovery was prepared by Glowacki (1945).

Light oils were recovered at most coal-carbonization plants, large carbureted water-gas plants, and large oil-gas plants. Small gas production plants would usually not recover the light oils (they did not produce enough to make their recovery profitable). When the light oils were not recovered, they passed through additional gas purifiers, then into the distribution system, and were ultimately burned with the product gas.
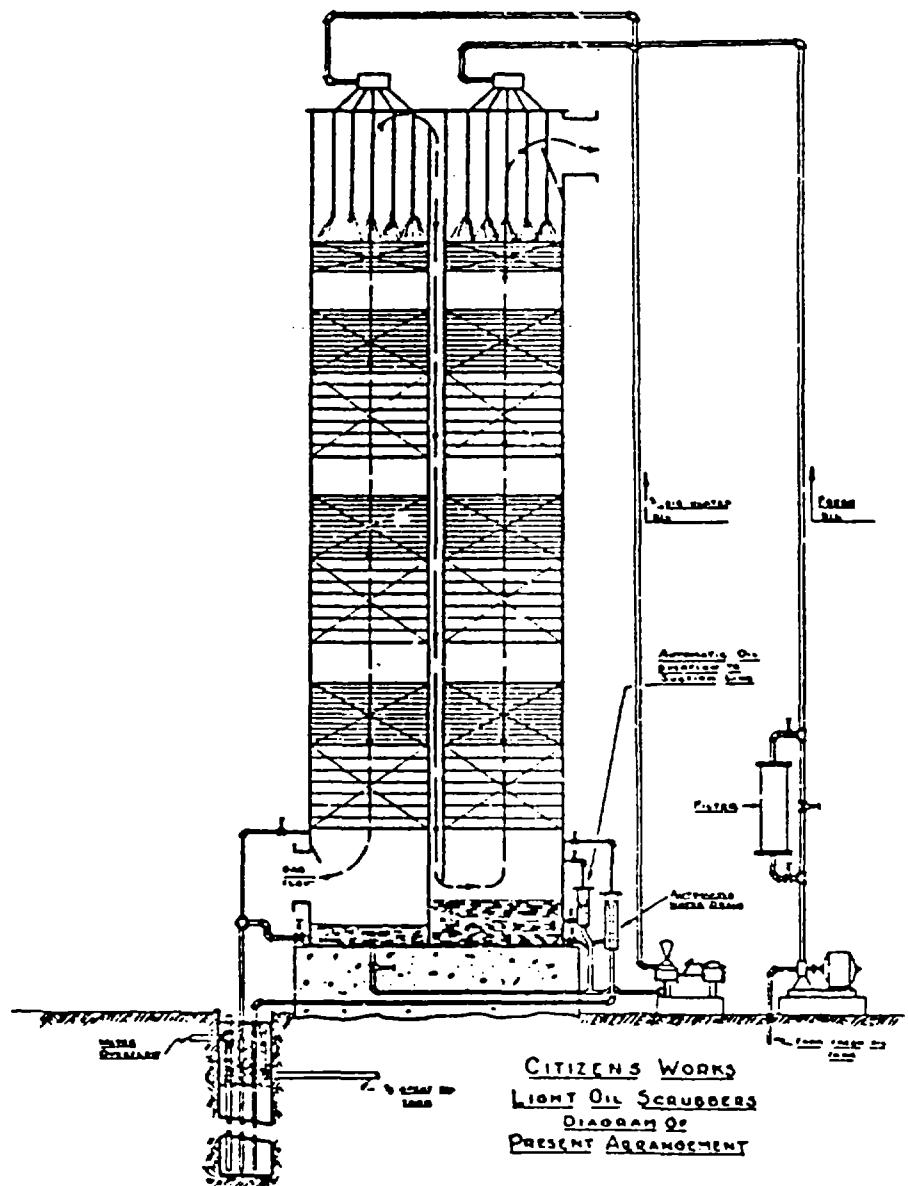
Figure   24. Light oil scrubber.

Source: Green, 1939.

TABLE 23. ANALYSIS OF A TYPICAL CRUDE COKE-OVEN LIGHT OIL

	Percentages by Volume
I. Forerunnings.	
Cyclopentadiene	0.5
Carbon disulfide	0.5
Amylenes and unidentified	1.0
II. Crude benzol.	
Benzene	57.0
Thiophene	0.2
Saturated nonaromatic hydrocarbons, unidentified	0.2
Unsaturates, unidentified	3.0
III. Crude toluol.	
Toluene	13.0
Saturated nonaromatic hydrocarbons, unidentified	0.1
Unsaturates, unidentified	1.0
IV. Crude light solvent.	
Xylenes	5.0
Ethyl benzene	0.4
Styrene	0.8
Saturated nonaromatic hydrocarbons	0.3
Unsaturates, unidentified	1.0
V. Crude heavy solvent.	
Coumarone, indene, dicyclopentadiene	5.0
Polyalkyl benzenes, hydrindene, etc.	4.0
Naphthalene	1.0
Unidentified "heavy oils"	1.0
VI. Wash oil	5.0 *
Total	100.0

* The amount of wash oil present depends greatly upon the performance and design of the debenzolization apparatus as well as upon the nature of the wash oil employed.

Source: Glowacki, 1945.

TABLE 24. CHEMICALS FOUND IN LIGHT OIL FROM COKE OVENS

Aromatic hydrocarbons

benzene	C_6H_6
toluene	$C_6H_5CH_3$
o-xylene	$C_6H_4(CH_3)_2$
m-xylene	$C_6H_4(CH_3)_2$
p-xylene	$C_6H_4(CH_3)_2$
ethyl benzene	$C_6H_5C_2H_5$
hydrindene	$C_6H_4CH_2CH_2CH_2$

isopropyl benzene	$C_6H_5CH(CH_3)_2$
o-ethyl toluene	$CH_3C_6H_4C_2H_5$
m-ethyl toluene	$CH_3C_6H_4C_2H_5$
p-ethyl toluene	$CH_3C_6H_4C_2H_5$
n-propyl benzene	$C_6H_5CH_2CH_2CH_3$
mesitylene	$C_6H_3(CH_3)_3$
pseudocumene	$C_6H_3(CH_3)_3$
hemimellitene	$C_6H_3(CH_3)_3$
naphthalene	$C_{10}H_8$
(1,2-dihydronaphthalene) †	$C_{10}H_{10}$
(1,4-dihydronaphthalene) †	$C_{10}H_{10}$
tetrahydronaphthalene	$C_{10}H_{12}$
(o-cymene) †	$CH_3C_6H_4CH(CH_3)_2$
(m-cymene) †	$CH_3C_6H_4CH(CH_3)_2$
(p-cymene) †	$CH_3C_6H_4CH(CH_3)_2$
durene	$C_6H_2(CH_3)_4$
isodurene	$C_6H_2(CH_3)_4$

Paraffin hydrocarbons

n-pentane	C_5H_{11}
n-hexane	C_6H_{14}
n-heptane	C_7H_{16}
n-octane	C_8H_{18}
n-decane	$C_{10}H_{22}$

Saturated cyclic hydrocarbons

cyclohexane	C_6H_{12}
methylcyclohexane	$C_6H_{11}CH_3$
(1,1-dimethylcyclohexane) †	$C_6H_{10}(CH_3)_2$
(1,2-dimethylcyclohexane) †	$C_6H_{10}(CH_3)_2$
(1,3-dimethylcyclohexane) †	$C_6H_{10}(CH_3)_2$
(1,4-dimethylcyclohexane) †	$C_6H_{10}(CH_3)_2$

Olefin hydrocarbons

1-butene	C_4H_8
1-pentene	C_5H_{10}
1-hexene	C_6H_{12}
cyclohexene	C_6H_{10}
1-heptene	C_7H_{14}

Diolefin hydrocarbons

1,3-butadiene	$CH_2:CHCH:CH_2$
2-butyne :	$CH_3C:CCH_3$
cyclopentadiene-1,3	$CH:CHCH:CHCH_2$

(cyclohexadiene) :	C_6H_8
(cyclohexadiene 1,3) :	C_6H_8
(cyclohexadiene 1,4) :	C_6H_8
dicyclopentadiene	$C_{10}H_{12}$

Source: Glowacki, 1945.

TABLE 24. (con.)

Aromatic hydrocarbons with un-
saturated side chains

styrene	$C_6H_5CH:CH_2$
indene	$C_6H_4CH_2CH:CH$

(2-methyl indene) †	$C_{10}H_{10}$
(3-methyl indene) †	$C_{10}H_{10}$

Neutral oxygen compounds

acetone	CH_3COCH_3
methylethyl ketone	$CH_3COC_2H_5$
coumarone	$C_6H_4OCH:CH$

acetophenone †	$C_6H_5COCH_3$
(2-methyl coumarone) †	C_9H_8O
(3-methyl coumarone) †	C_9H_8O
(5-methyl coumarone) †	C_9H_8O
(6-methyl coumarone) †	C_9H_8O
(7-methyl coumarone) †	C_9H_8O

Neutral and acidic nitrogen
compounds

hydrogen cyanide	HCN
acetonitrile	CH_3CN
benzonitrile	C_6H_5CN

Phenols

phenol	C_6H_5OH
o-cresol	$CH_3C_6H_4OH$
m-cresol	$CH_3C_6H_4OH$
p-cresol	$CH_3C_6H_4OH$
2,3-dimethyl phenol	$(CH_3)_2C_6H_3OH$
2,4-dimethyl phenol	$(CH_3)_2C_6H_3OH$
2,5-dimethyl phenol	$(CH_3)_2C_6H_3OH$
2,6-dimethyl phenol	$(CH_3)_2C_6H_3OH$
3,4-dimethyl phenol	$(CH_3)_2C_6H_3OH$
3,5-dimethyl phenol	$(CH_3)_2C_6H_3OH$
o-ethyl phenol	$C_2H_5C_6H_4OH$
m-ethyl phenol	$C_2H_5C_6H_4OH$
p-ethyl phenol	$C_2H_5C_6H_4OH$

Basic nitrogen compounds

pyrrole	$NHCH:CHCH:CH$

pyridine	C_5H_5N
aniline	$C_6H_5NH_2$
2-methyl pyridine	$CH_3C_5H_4N$
3-methyl pyridine	$CH_3C_5H_4N$
4-methyl pyridine	$CH_3C_5H_4N$
o-toluidine	$CH_3C_6H_4NH_2$
2,3-dimethyl pyridine	$(CH_3)_2C_5H_3N$
2,4-dimethyl pyridine	$(CH_3)_2C_5H_3N$
2,5-dimethyl pyridine	$(CH_3)_2C_5H_3N$
2,6-dimethyl pyridine	$(CH_3)_2C_5H_3N$
3,4-dimethyl pyridine	$(CH_3)_2C_5H_3N$
(3,5-dimethyl pyridine) †	$(CH_3)_2C_5H_3N$
dimethyl aniline	$C_6H_5N(CH_3)_2$

TABLE   24. (con.)

2,4,5-trimethyl pyridine	$(CH_3)_2C_5H_2N$
2,4,6-trimethyl pyridine	$(CH_3)_2C_5H_2N$
(2,3,4-trimethyl pyridine) †	$(CH_3)_2C_5H_2N$
(2,3,5-trimethyl pyridine) †	$(CH_3)_2C_5H_2N$
(2,3,6-trimethyl pyridine) †	$(CH_3)_2C_5H_2N$

Sulfur compounds

sulfur	S
hydrogen sulfide	H_2S
carbonyl sulfide	COS
carbon disulfide	CS_2
methyl mercaptan	CH_3SH
ethyl mercaptan	C_2H_5SH
dimethyl sulfide	$(CH_3)_2S$
diethyl sulfide	$(C_2H_5)_2S$
thiophene	C_4H_4S
2-methyl thiophene	$CH_3C_4H_3S$
3-methyl thiophene	$CH_3C_4H_3S$
(2,3-dimethyl thiophene) †	$(CH_3)_2C_4H_2S$
(2,4-dimethyl thiophene) †	$(CH_3)_2C_4H_2S$
(2,5-dimethyl thiophene) †	$(CH_3)_2C_4H_2S$
(3,4-dimethyl thiophene) †	$(CH_3)_2C_4H_2S$
(2,3,4-trimethyl thiophene) †	$(CH_3)_3C_4HS$
thionaphthene	$C_6H_4SCH{:}CH$

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1.3.5 Removal of Ammonia and Recovery

The production of ammonia, cyanides, and phenolic compounds occurred with gas produced by coal carbonization. These compounds were produced in trace amounts by carbureted water gas and oil gas and were not removed or recovered from these processes. Prior to the Haber process for the synthetic production of ammonia, coal carbonization was the principal source of fixed nitrogen. The removal of ammonia from the gas was always accomplished by scrubbing the gas with water, condensate, or sulfuric acid. Ammonia has a very high affinity for both water and acid solutions and is readily removed by aqueous scrubbing.

During coal carbonization, a portion of the nitrogen in the coal is converted to ammonia, and other nitrogen forms cyanides, organic nitrogen compounds, or remains in the coke. Table 25 shows the average distribution of nitrogen compounds from high-temperature carbonization of coal. Approximately 18 percent of the nitrogen in coal is converted to ammonia during carbonization. This is about 1.1 percent by volume of the raw coal gas.

There were three basic processes for the removal of ammonia from coal gas. These were the direct method, the indirect method, and the semidirect method. They differ primarily in the treatment of condensate containing the ammonia and are described in detail in several commonly available references (Wilson and Wells, 1945; Kohl and Riesenfeld, 1985; Hill, 1945). In the direct method, the raw coal gas was scrubbed directly with a solution of sulfuric acid. The ammonia was absorbed into the solution, reacted with the sulfuric acid, and the resulting ammonium sulfate precipitated. This method was the simplest method of removing ammonia as a product from the gas, but the resulting ammonium sulfate was of p. - quality and generally contained substantial impurities. An additional drawback to the process was the degradation of the coal tar from contact with the sulfuric acid.

The indirect process, as shown in Figure 25, removes ammonia from the coal gas by first absorbing the NH_3 into water, then releasing the ammonia as a gas in an ammonia still. The raw coal gas first contacts recirculated

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TABLE 25. DISTRIBUTION OF NITROGEN IN COAL CARBONIZATION PRODUCTS

	% of nitrogen originally in coal
Ammonia	18.0
Cyanide	1.2
In tar	3.3
Free in gas	27.5
In coke	50.0
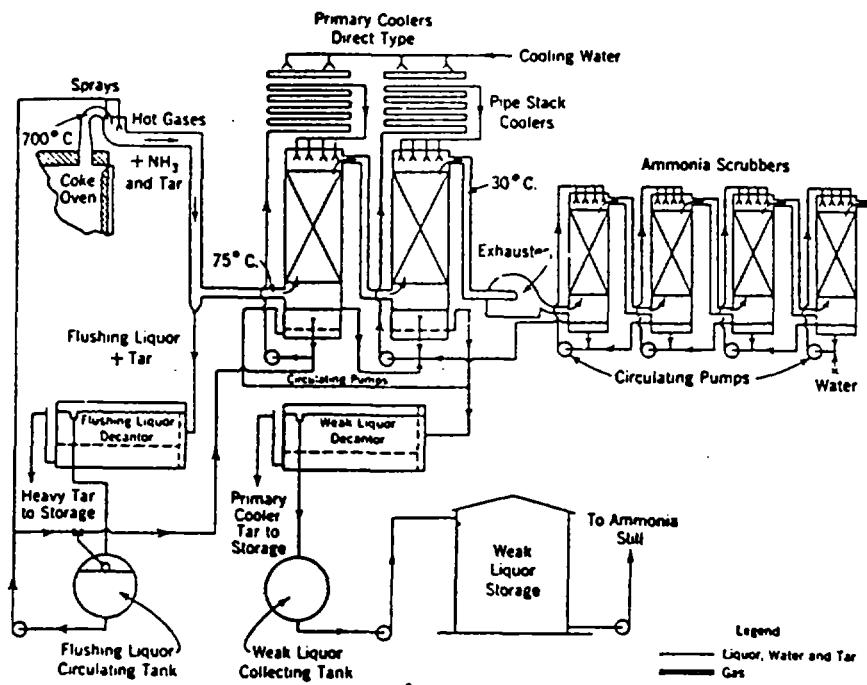
Figure 25. Indirect process for ammonia recovery.

Source: Wilson and Wells, 1945.

flushing liquor in the hydraulic main. The gas is cooled to a certain extent, and the heavy tars condense. The fixed ammonia compounds (those that do not release ammonia when the solution is boiled, such as $(NH_4)_2SO_4$ and NH_4Cl, are dissolved into the flushing liquor. This gas is then further cooled either by direct or indirect condensation, with most of the tar and water being condensed from the gas. The condensate, which has a high ammonia concentration, is separated from the tar in gravity separators. The remaining tar aerosols in the gas are removed by an ESP, and the remaining ammonia in the gas is removed by scrubbing with water.

The condensate and ammonia scrubber water are mixed and fed to an ammonia still that uses lime and heat to decompose ammonia salts and free the ammonia as a gas. Figure 26 is a diagram of the ammonia stills that were generally used. The ammonia still is constructed of a fixed still, volatile still, and lime keg. The volatile still removes all of the free ammonia and other volatile compounds from the crude ammonia liquor. The fixed still decomposes fixed ammonia salts in the liquor and liberates the ammonia gas. Lime water is fed to the lime keg while ammonia still waste is removed from the base of the fixed still. The free ammonia and steam that exit the top of the volatile still were scrubbed either with water (to reabsorb the ammonia as an aqueous ammonia product) or with sulfuric acid (to produce ammonium sulfate).

The semidirect process (Figure 27), patented by the Koppers Company in 1909, was a variation of the indirect process. The processes were identical except the indirect process did not use water scrubbers to remove the final amounts of ammonia from the gas. Instead, the coal gas (after complete tar removal) was bubbled through sulfuric acid with the ammonia from the lime still. This reduced the amount of crude ammonia liquor that was processed through the ammonia still and allowed for better heat utilization in the saturator. There were also reductions in capital and operating costs with the semidirect process, with only marginal effects on the quality of ammonium sulfate product.

The lime still would have been effective at removing volatile organics that were dissolved in the liquor, but tar acids (principally phenols) were retained in the still waste and frequently constituted a major disposal problem for the gas plants. The phenols have a very low taste threshold in water,

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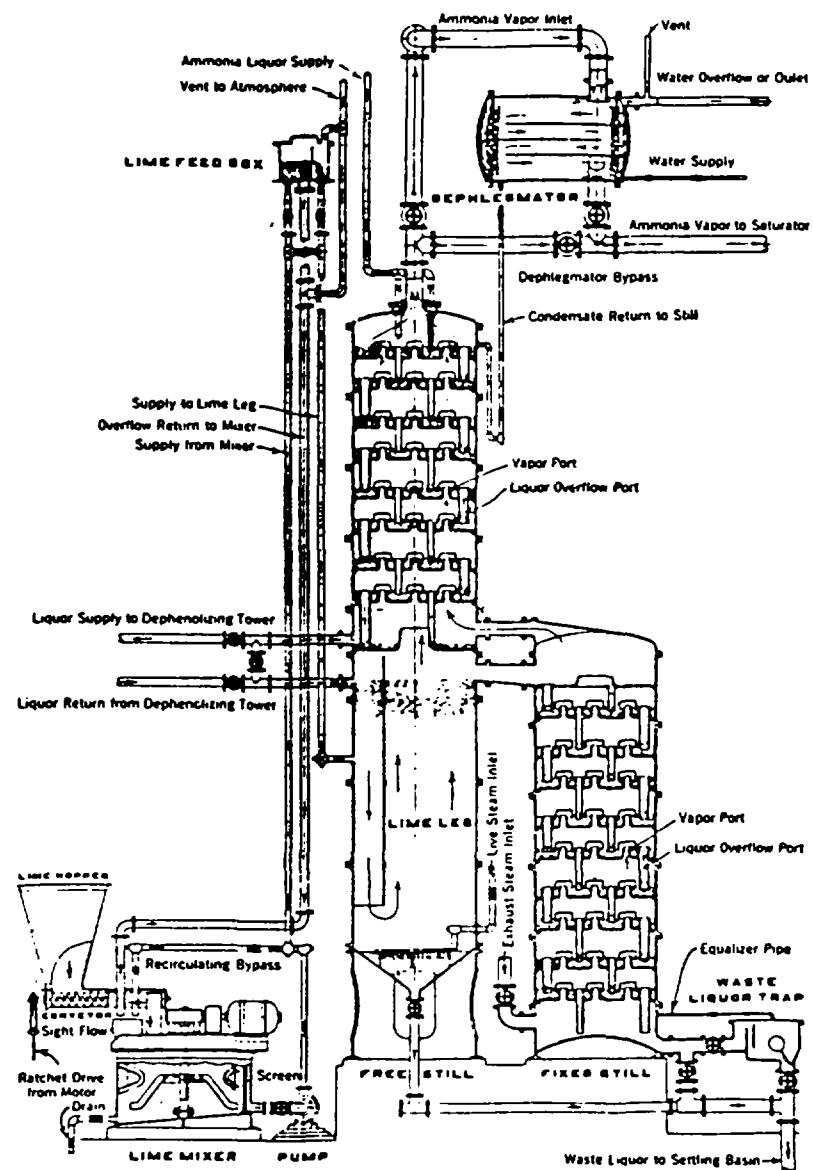
Figure 26. Ammonia still.

Source: Hill, 1945.
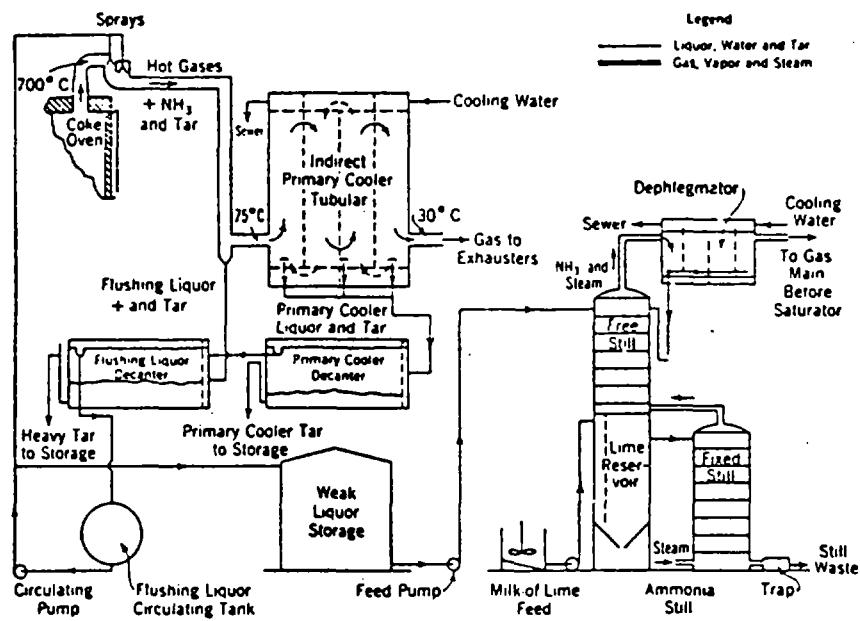
Figure 27. Semidirect process for ammonia recovery.

Source: Wilson and Wells, 1945.

particularly when the water is chlorinated. The removal and treatment of phenols in the still waste is discussed in Section 1.3.5.

Sometimes the gas plant would sell the ammonia liquor directly to a chemical company for the production of ammonium sulfate. The ammonia liquor could be used directly as scrubbing liquor to absorb SO_3 produced by burning sulfur. This is essentially the process for producing sulfuric acid, except that by using ammonia liquor, ammonium sulfate can be produced directly as a product.

1.3.6 Phenol Removal and Recovery

Phenol was produced in the carbonization of coal. As an acidic compound, it was readily absorbed in the condensate and ammonia liquor during the purification of the coal gas. The phenol remained in the ammonia still waste and had to be removed from this waste stream before disposing of the water. The phenolic compounds were very noticeable in water, imparting a medicine taste to it. This occurred even at low concentrations and was exacerbated when the water was chlorinated. There were several methods that were commonly used for the removal of phenol from the ammonia still waste.

The simplest method of disposing of phenol containing liquid wastes was to discharge the water directly into the city sewer system (if one were available). The phenol in the wastes was rapidly degraded by organisms in the sewage and by the activated sludge method of sewage disposal.

A common method of disposal was to use the water to quench coke as it was removed from the ovens. This method substantially reduced the volume of the wastes, but it degraded the value of the coke, greatly increased the corrosion of steel in the coke-quenching area, and evaporated phenols into the air. These evaporated phenols generally killed any remaining plant life around the coke plant and may have been washed into surface water.

If recovery of the phenols were desired, the phenol was extracted from the raw ammonia liquor by washing the liquor with benzene or light oil, then recovering the phenol from the benzene by washing it with a solution of sodium hydroxide. This process is shown in Figure 28. The process uses benzene or light oil, which continuously absorbs phenol in one tower, while the solution is continuously regenerated by contact with a sodium hydroxide solution in a second tower. The sodium phenolate was then usually converted to raw phenols by "springing" the solution with carbon dioxide. The process actually removed

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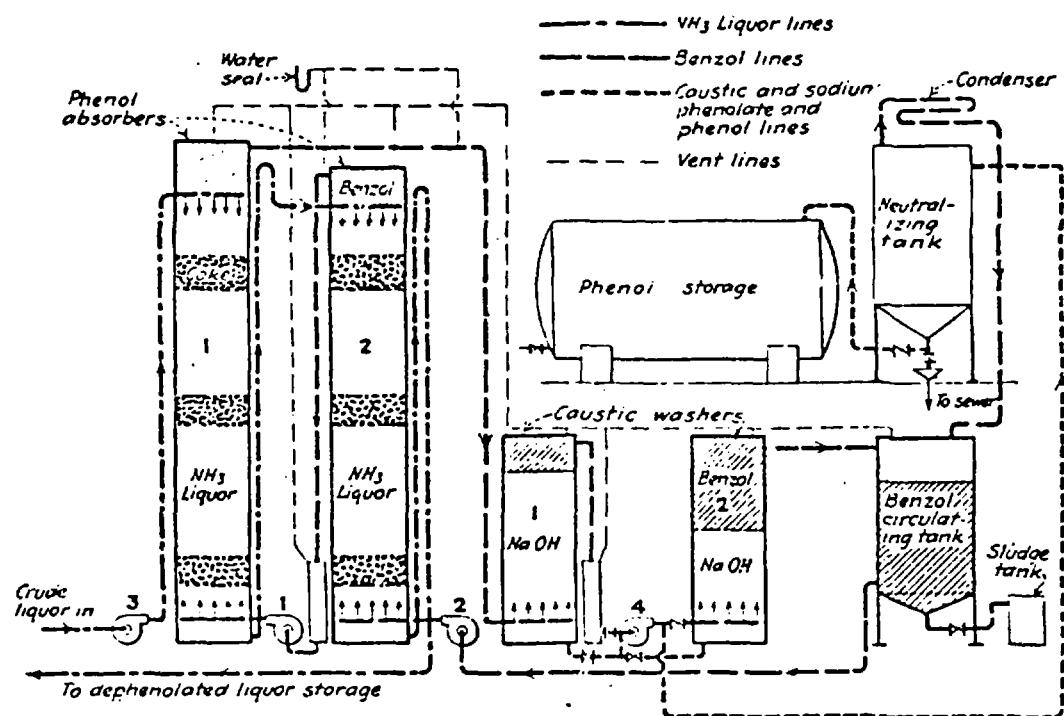
Figure 28. Benzene extraction of phenols.

Source: Jones, 1928.

a variety of tar acid compounds from the liquor, although the recovered product was primarily phenol. The recovered tar acids from one plant were analyzed (dry basis) as 57 percent phenol, 13 percent o-cresol, 8 percent m-cresol, 10 percent p-cresol, and 10 percent higher tar acids (Wilson and Wells, 1945). This process generally removed about 75 percent of the phenols contained in the ammonia liquor, but higher removal efficiencies were obtained when the phenols were separated from the benzene by distillation instead of extraction with caustic.

A second common method of recovering phenols was the Koppers vapor recirculation process. In this process (shown in Figure 29), ammonia liquor was removed from the base of the free still (after removal of the free ammonia, but before the fixed ammonia salts are decomposed) and was stripped by steam. The steam-stripped ammonia liquor was then returned to the lime keg section of the ammonia still for the decomposition of fixed ammonia salts. The steam and phenols were then scrubbed by a solution of sodium hydroxide, removing the phenols as sodium phenolate. The sodium phenolate could then be sprung as phenol-using carbon dioxide. This process had higher removal efficiencies than did extraction of phenols, and it generally gave about 97 percent removal. Inlet concentrations of phenol were about 2.5 g/L.

Wilson and Wells (1945) mention the disposal of waste ammoniacal liquors into the ground but advise:

> Discharge into an opening, such as a disused well, is dangerous, because the final fate of the liquor is unknown. It may be gradually dissipated and purified as it seeps through the soil. On the other hand, it may find its way into some water-bearing strata or percolate unchanged through the layers of soil to drain into a stream. In such a case, the pollution would not appear immediately, but when it did, deposits of the material in the contaminated soil would cause the trouble to persist over a long period of time.

The ammoniacal liquors could also be discharged directly in a stream or bay, if the water were not used for drinking purposes. This would have been more common along coastal areas, where the discharges could flow directly into the ocean, and complaints would be minimal. Evaporation of the liquors by flue gas or steam was also suggested as a method for disposal, but it was not generally employed.

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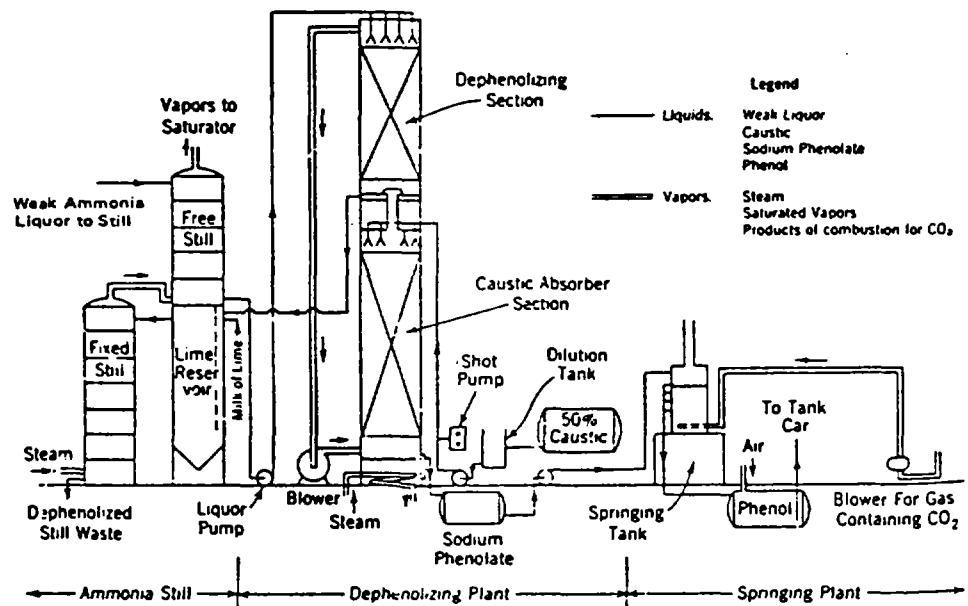
Figure 29. Koppers vapor recirculation process for phenol removal.

Source: Wilson and Wells, 1945.

1.3.7 Removal of Hydrogen Sulfide

1.3.7.1 Introduction--

The need to remove hydrogen sulfide from town gases was recognized very early in the industry. If left in the gas, the H_2S would cause corrosion in the distribution system and appliances, be a nuisance to the consumer, and be an odor problem with even small leaks of gas. Hydrogen sulfide was produced by all major gas production methods, so its removal was universal within the industry. The concentration of hydrogen sulfide in the raw gas (and hence the amount of H_2S to be removed) was proportional to the original sulfur concentration in the gas feedstocks. For coal carbonization, the sulfur concentration in the original coal determined the gas H_2S concentration; for carbureted water gas and oil gas, the sulfur concentration of the oil used was the primary variable. Table 26 shows typical concentrations of hydrogen sulfide in town gases, although these numbers would vary considerably, depending on the sulfur concentration of the feedstocks used to produce the town gases.

The sulfur removed from the gas could either be recovered as a salable byproduct, discharged as H_2S to the air, or discarded as waste. Lime was the original material used for the purification of gas until the process was widely replaced by iron oxides after about 1885. Iron oxides were universally used for the removal of hydrogen sulfide from coal gas, water gas, and oil gas until about 1927, when several liquid purification processes for hydrogen sulfide removal became available (primarily the Seaboard and Thylox processes).

1.3.7.2 Hydrogen Sulfide Removal by Lime--

Hydrated lime was one of the earliest techniques used to remove H_2S, CO_2, and cther impurities from coal gas. This lime was produced by calcining limestone, then slaking the lime with water to form calcium hydroxide. The relevant reactions for the purification of coal gas with hydrated lime are:

$$Ca(OH)_2 + H_2S = CaS + 2H_2O$$

$$Ca(OH)_2 + CO_2 = CaCO_3 + H_2O.$$
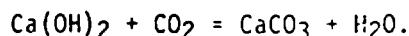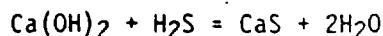
The lime also removed some cyanides (which reacted with iron impurities in the lime to form ferrocyanides) and some tar materials. Stoichiometrically, each mole of lime could remove one mole of CO_2 or H_2S. Actual

TABLE 26. TYPICAL CONCENTRATIONS OF HYDROGEN SULFIDE IN
TOWN GASES

Gas	H_2S concentration (ppm)
Coal gas	3,200-7,990
Carbureted water gas	800-2,400
Pacific Coast oil gas	3,200-4,000

SOURCE: Morgan, 1926.

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conversion of the lime to sulfide was usually about 40 percent, so that large quantities of lime were required for purification of the gas (Veley, 1885). The spent lime could not be regenerated, and it usually had a foul odor from the tars and a blue color from the ferric cyanides. After disposal, the CaS would slowly combine with CO_2 to rerelease H_2S by the reaction:

$$CaS + CO_2 + H_2O = CaCO_3 + H_2S.$$
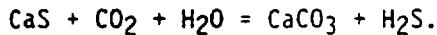
Although some spent lime was sold or given away for agricultural purposes, much of it was discarded. Because it could only be used once for purification, it was a costly purification method to use. The discovery and use of the iron oxide process for removing H_2S around 1885 replaced almost all the use of lime for gas purification. The iron oxide process did not remove CO_2 from the gas, and CO_2 gave a gas with poor lighting and burning properties. Some lime was frequently used in a bed directly after the iron oxide purifiers to remove CO_2 from the gas. This use of lime involved much smaller quantities of lime than were previously employed at operating gas plants.

1.3.7.3 Removal of Hydrogen Sulfide by Iron Oxide--

Iron oxide removed H_2S from the gas, was regenerated with oxygen from air, then reused to remove more H_2S. The iron oxide could be regenerated until it was between 40 and 50 percent sulfur by weight, at which time it was generally discarded. This regeneration allowed iron oxide to remove much more H_2S than did lime and substantially reduced the cost of gas manufacture.

The relevant reactions for the removal of hydrogen sulfide and regeneration of the spent oxide are below:

H_2S REMOVAL

(1) $Fe_2O_3 + 3H_2S = Fe_2S_3 + 3H_2O$

(2) $Fe_2O_3 + 3H_2S = 2FeS + S + 3H_2O$

REGENERATION

(3) $2Fe_2S_3 + 3O_2 = 2Fe_2O_3 + 6S$

(4) $4FeS + 3O_2 = 2Fe_2O_3 + 4S$

DEACTIVATION

(5) $FeS + S = FeS_2.$
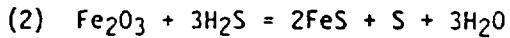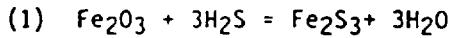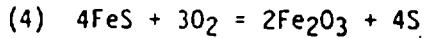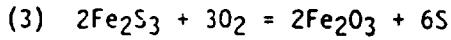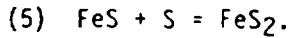
Reaction (1) was the most desirable for gas purification, and it occurred under slightly alkaline conditions. Reaction (2) occurred under slightly acidic conditions. The formation of ferrous sulfide (FeS) was undesirable because it combines with free sulfur to form FeS_2 (reaction 5), which cannot be regenerated. During revivification, some sulfuric acid is formed by the reactions:

(6) $FeS + 2O_2 = FeSO_4$

(7) $FeSO_4 + H_2O = H_2SO_4 + FeO$.
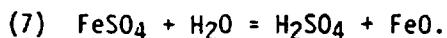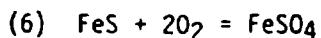
Some hydrated lime or soda ash (Na_2CO_3) was added to the iron oxide to keep it in an alkaline state. Some ammonia was usually present (or added) to the gas passing through the iron oxide to keep the oxide alkaline and to promote the removal of cyanide from the gas. A small concentration of ammonia apparently promoted the removal of cyanides as ferrocyanide while a high ammonia concentration caused the cyanides to be removed as thiocyanates.

The iron oxide used for the removal of hydrogen sulfide was of three major types: rusted iron borings, bog ore, and precipitated iron oxides. Each of these materials was usually mixed with a fluffing material to provide for better gas flow through the iron oxide (after 1930, however, some plants stopped adding fluff material to the iron oxide). The fluffing material was primarily woodchips, but blast furnace slag and corn cobs were also used. The iron borings were usually added to the woodchips, then sprayed with water and exposed to air to rust the borings. Salt or ferrous sulfate was often added to the water to promote the rusting. Most plants used the rusted iron borings, but some used bog ore (naturally precipitated iron oxide) during World War I and World War II, and some plants switched to precipitated iron oxides after they were introduced about 1930.

The oxides were placed into boxes, and the town gas flowed through the box. Several oxide boxes were connected in series, and the order in which the gas contacted the boxes rotated so that gas contacted the most fouled oxide first and boxes of fresh oxide last. This permitted maximum utilization of the oxides, while removing the H_2S concentration in the product gas to very low levels. The oxides that contacted the gas first were periodically discarded, the box refilled with fresh oxide, and the box added as the last oxide to purify the gas.

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Originally, the oxides were revived by physically removing them from the box, exposing them to air, and then replacing the material into the box. This was very labor intensive, and because the regeneration of the oxide was exothermic, considerable care was required to prevent the oxides from becoming deactivated or igniting the tars and bulk material with the oxide. This was replaced by the practice of reviving the oxide continually while the oxide was removing H_2S. A small amount of air (approximately 2 percent) was added to the gas prior to the gas entering the iron oxide purifiers. The oxygen continuously regenerated the oxide in the boxes and greatly reduced the labor required for the purification. The major disadvantage of this method was that the nitrogen added to the gas with the air reduced the heating value of the gas. The oxide was sometimes revived by switching the box out of the combustible gas and blowing air through the oxide.

1.3.7.4 Liquid Scrubbing for Hydrogen Sulfide Removal--

Lime water was the original method of removing impurities from coal carbonization gases. It was principally used in Great Britain, but its use was fairly rapidly replaced by use of hydrated lime in beds. The basic process was to use a solution of hydrated lime in water (milk of lime) and bubble the raw coal gas through the liquid. Lime removed the hydrogen sulfide as CaS, carbon dioxide as $CaCO_3$, and other impurities by their solubility in water. Tars and oils were also condensed into the lime water. The contaminated lime water was generally run directly into the nearest river, much to the displeasure of those downstream. The CaS reacted with carbon dioxide and water to rerelease hydrogen sulfide while the oils and phenols contaminated the water and killed fish.

Lime water was not used at a significant level in the United States because, by the time gas was produced, beds of hydrated lime were used instead of the lime solutions.

1.3.7.4.1 Seaboard process--The first major liquid purification process for the removal of hydrogen sulfide was the Seaboard process, which was named for the plant in New Jersey where it was developed. This process used a solution of sodium carbonate to scrub H_2S from the coal gas and release the H_2S into the air when the solution was regenerated. This process was invented in 1920 and installed in 6 plants (with 12 under construction) by 1923 (Bird,

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1923). It was used on coal carbonization plants, carbureted water-gas plants, and oil-gas plants.

Figure 30 is a diagram of the Seaboard process. The process used either two packed columns or a single packed column divided into two sections. In this figure, the gas is scrubbed in the upper half of the column by a solution of sodium carbonate (1 to 3 percent). The solution is introduced at the top of the column and flows down the packing in the column. The gas enters the .niddle of the column and flows out through the top of the column. As it progresses through the column, the hydrogen sulfide and cyanide gases are absorbed into the. solution. The solution then flows to the top of the bottom column. There it flows over another set of packing and contacts air (blown into the base of the column and removed from the top of the lower column). The air strips the H_2S from the solution, reviving the solution (actification). The reactivated solution is then removed from the base of the column and returned to the absorber (the upper column). The solution is continuously recycled, but it must be replenished periodically by adding fresh solution. The cyanide in the gas is removed as sodium thiocyanate, which cannot be regenerated to sodium carbonate. Sodium thiosulfate and sodium sulfate were also formed by side reactions in the scrubber liquid.

The actifier air contains the H_2S that was originally in the product gas. This stream was usually just vented to the environment, although sometimes it was used as boiler air so that the H_2S would be oxidized to SO_2 and reduce odor problems created by H_2S. Table 27 shows some typical operating parameters for the Seaboard process. The removal efficiency of the Seaboard process was generally between 70 and 95 percent. The remaining hydrogen sulfide in the gas was removed by a bed cf iron oxide that immediately followed the Seaboard process. The Seaboard process was extremely efficient at removing hydrogen cyanide, so that no cyanide would be removed in the iron oxide that was used with the Seaboard process.

The Seaboard process greatly reduced the amount of iron oxide purification required to remove hydrogen sulfide from town gas. Because it discharged all of the sulfur it removed to the atmosphere, processes were developed that were similar to the Seaboard process, but that recovered the sulfur as a byproduct.

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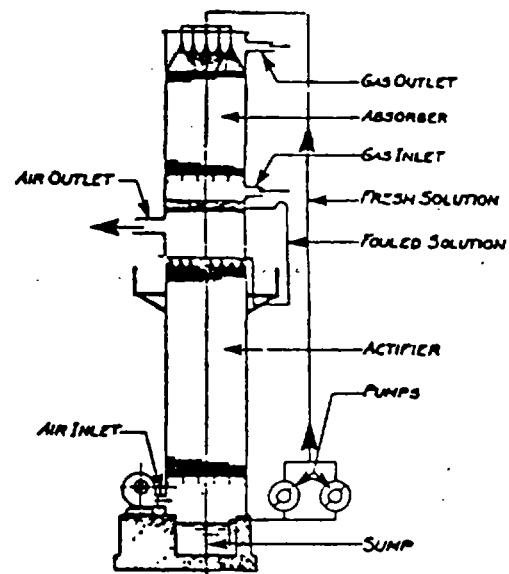
Figure 30. Seaboard process for H_2S removal.

Source: Morgan, 1926.

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TABLE 27. OPERATION OF SEABOARD PROCESS

	Plant		
	A	B	C
Gas purified (10^6 ft^3/day)	5,317	2,557	353
Inlet H_2S (ppm)	2,760	6,950	7,100
Outlet H_2S (ppm)	145	304	17
H_2S removed/day (lb)	10,250	15,166	29,920
Na_2CO_3 used/day (lb)	1,000	2,005	149
% removal of H_2S	94.7	95.6	99.8

SOURCE: Herbst, 1931.

1.3.7.4.2 Thylox, Nickel, and Ferrox processes--The Thylox process was developed shortly after the Seaboard process, and it recovered the sulfur. Two other processes, the Nickel process and the Ferrox process, used the same apparatus as the Thylox process, but they used different scrubber solutions. Figure 31 is a diagram of the equipment used for the three processes. The gas is scrubbed counter-currently with the absorber solution in the absorber. For the Thylox process, this solution was a mixture of arsenic trioxide and sodium carbonate. The Ferrox process used an iron compound suspended in soda ash, and the Nickel process used a solution of a nickel salt in soda ash (Downing, 1934). The foul solutions were then pumped with compressed air into the thionizer, where the oxygen in the air oxidized the H_2S to sulfur crystals. Table 28 lists some typical operating data for the Thylox and Nickel processes. The arsenic, iron, and nickel act as a catalyst for the oxidation. The sulfur slurry is then drawn from the top of the thionizer, and the sulfur is recovered by filtration. The Arsenic and Ferrox processes could be used with either carbureted water gas, oil gas, or coal-carbonization gas. The nickel catalyst in the Nickel process was poisoned rapidly by cyanide, and the process could be used only on gases that had low cyanide concentrations. This limited the process to use only with oil and water gas.

The Thylox, Nickel, and Ferrox processes were all very efficient in the removal of cyanide, as was the Seaboard process. Cyanide was converted to thiocyanates in all four processes. Each liquid process also required the periodic replacement of the scrubber solutions. This was accomplished either through normal fluid losses of the system (carryover to the iron oxide beds, spills, evaporation, and liquid loss with the filtered sulfur product), the continuous withdrawal and replacement of spent solution, or the periodic draining and fluid replacement of all the scrubber liquid.

The three sulfur recovery processes were fairly efficient in the removal of hydrogen sulfide (about 98 percent) but were generally followed by an iron oxide bed to remove the last traces of the H_2S. The spent iron oxide from this type of operation would be expected to contain some of the scrubber solution that would be carried over from the liquid purification processes. The arsenic or nickel salts could occur in the spent oxides.

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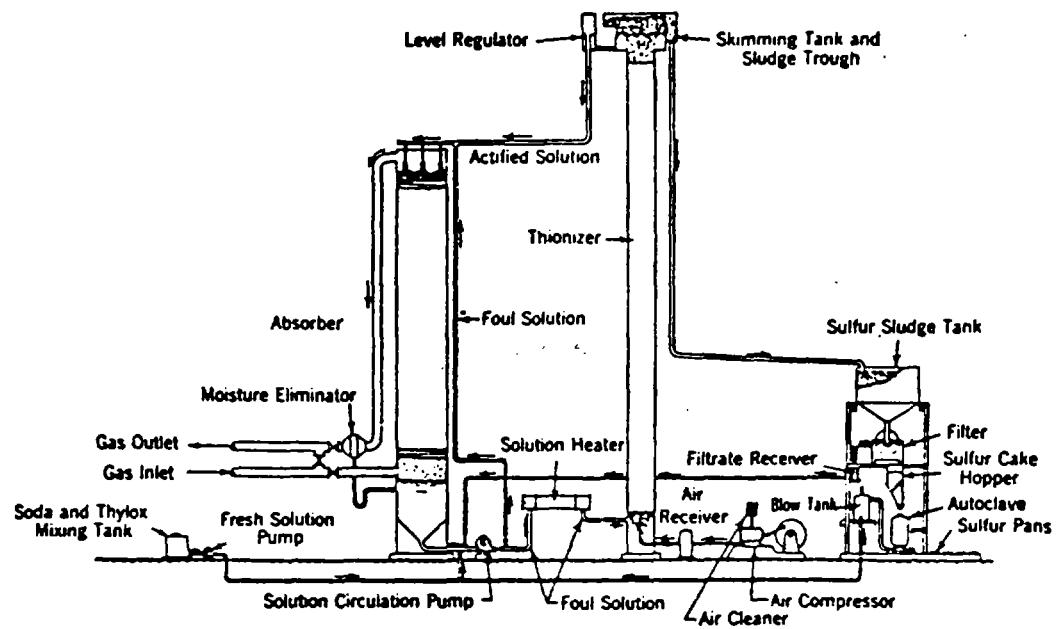
Figure 31. Thylox process for H$_2$S removal.

Source: Gollmar, 1945.

TABLE 28. OPERATION OF NICKEL AND THYLOX PROCESSES

	Thylox process		Nickel process
	Avg. of 3 coal-gas plants	Coal and water-gas plant	Avg. of 4 oil-gas plants
Inlet H_2S (ppm)	4,794	4,315	--
Outlet H_2S (ppm)	85.2	112	--
H_2S removal efficiency (%)	98.2	97.4	70-100
HCN inlet (ppm)	322	81	--
HCN outlet (ppm)	0	0	--
HCN removal efficiency (%)	100	100	--
Na_2CO_3 consumption ($lb/10^6 ft^3$)	0.07	0.06	0.102
As_2O_3 consumption ($lb/10^6 ft^3$)	0.022	0.024	0
Total gas volume purified ($10^6 ft^3$/day)	3,000-8,000	14,000	--
Nickel salt consumed ($lb/10^6 ft^3$)	--	--	0.023

SOURCES: Gas Engineers Handbook, 1934; Cundall, 1927.

1.3.8 Cyanide Removal

Cyanide was also an impurity in gas produced by coal carbonization, but it was produced in only trace quantities by carbureted water gas and oil gas. The recovery of cyanides for sale was only profitable at the larger coal-gas plants and only prior to the Haber process for ammonia production (cyanide can be produced from ammonia and coke). Table 29 shows representative concentration of cyanide in coal gas, carbureted water gas, and oil gas. The cyanide in coal gas was either recovered as a product or was removed with hydrogen sulfide. Because both hydrogen sulfide and cyanide are acid gases, processes that removed hydrogen sulfide generally removed cyanide as well.

The concentration of cyanogen in coal gas was generally between 0.12 and 0.20 percent (Hill, 1945). Because cyanide was rarely recovered from coal gas, the recovery processes will be described in only general details. Additional details of specific processes may be found in articles by Hill (1945) and Powell (1922). The Bueb process used a scrubbing solution of ferrous sulfate in ammonia liquor. Hydrogen sulfide in the gas reacted with the ferrous sulfate to form ferrous sulfide. This in turn reacted with cyanide to form ammonium-iron-cyanide complexes. The discharge from this process is a light-colored mud, which turns blue on exposure to air. It has a cyanogen content (as Prussian blue) of 13.5 percent and an ammonia content of 6 to 7 percent (Hill, 1945). This product is then boiled and filtered, producing an ammonium sulfate solution and a filter cake of about 30 percent Prussian blue. The blue mud product can then be converted to calcium ferrocyanide by boiling with lime (driving off the ammonia), or potassium ferrocyanide by adding KCl to a solution of the calcium ferrocyanide.

In the Foulis process, a water-ferrous carbonate slurry (from sodium carbonate and ferrous chloride) is contacted with the coal gas. The cyanide reacts with the ferrous carbonate to yield a product of sodium ferrocyanide. The Burkheiser purification process used a slurry of iron oxide in water to simultaneously remove both HCN and H_2S. Dissolved ammonia keeps the liquid alkaline and helps remove the cyanide as thiocyanide compounds.

Cyanide was generally not recovered from the coal gas, but was instead removed with the hydrogen sulfide. The removal of hydrogen cyanide by iron oxide purification, the Seaboard process, the Thylox process, and lime purifi-

TABLE 29. CONCENTRATION OF HCN IN VARIOUS GASES

Gas	HCN concentration in raw gas (ppm)
Vertical retort	886
Coke oven	516-947
Carbureted water gas	Trace to 26 ppm
Oil gas	a

[a]Not listed but known to be comparable to carbureted water gas.

cation are described below. The purification processes themselves are described in Section 1.3.7.

The earliest method of removing hydrogen sulfide was to run the raw coal gas from the condensers directly through a bed cf hydrated lime. The lime removed the hydrogen sulfide, and the cyanides in the gas would be removed by iron impurities in the lime. This caused the formation of Prussiar blue in the lime and "gave rise to the technical term blue billy" (Veley, 1875) for the spent lime wastes.

If cyanide were not removed by a specific process before iron oxide purification, then the iron oxide would remove the cyanide. Hill gave the following possible reactions for the removal of cyanide with iron oxide:

$$Fe(OH)_2 + 2HCN = Fe(CN)_2 + 2H_2O \quad and/or$$
$$FeS + 2HCN = Fe(CN)_2 + H_2S.$$
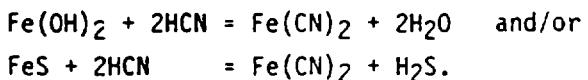
The ferrous cyanide then combines with ammonium cyanide to form complex compounds such as $(NH_4)_4Fe(CN)_6$ and $(NH_4)_2Fe_2(CN)_6$. The final form of the cyanide is as complex ferri-, ferro-, and ferri-ferro ammonium cyanide complexes. These chemicals are best identified by their intense blue color. A large amount of ammonia in the gas, or strong fixed alkali in the oxide, caused the cyanide to be removed as thiocyanates (either sodium, potassium, or ammonium thiocyanate). The cyanides were generally disposed with the spent oxides, although several methods for the removal and recovery of ferrocyanides and ferricyanides from the spent oxide were developed. These methods usually removed the sulfur from the spent oxide, then treated the remaining mass with strong alkalies.

The Seaboard process, which removed H_2S by absorption into a solution of Na_2CO_3, was a very efficient process for removing HCN. The HCN was originally absorbed as sodium cyanide, which is then converted to sodium thiocyanate. Each mole of cyanide removed requires a mole of sodium carbonate, and the thiocyanate could be recovered as a byproduct. This was not generally done, however, because it was usually discarded with spent scrubber solution.

The Thylox process used a solution of sodium carbonate and arsenic trioxide solution to remove hydrogen sulfide and recover it as sulfur. The process also removed cyanide as thiocyanate in a manner similar to the Seaboard

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process. The thiocyanates would accumulate in the solution and were removed with a side stream of scrubber liquid.

1.3.9 Tar and Light Oil Treatment

Many gas production plants did not refine or process their byproduct tars; instead, they sold them to processors, sold them as fuel, used them onsite, or discarded them. It is beyond the scope of this study to review tar processing in detail, but several aspects of tar treatment should be mentioned because they could occur at many of the gas sites. Table 30 is a list of the tar fractions and major components present in coal tar. The component list for tar from water-gas and oil-gas processes would be similar to this, except that there would be no tar acids, tar bases, or nitrogen heterocyclics as major components. Rhodes (1945) prepared a list of about 350 chemicals that were identified in coal tar, and estimates of the actual number of compounds run to 5,000 (Smith and Eckle, 1966). The chemicals contained in water-gas tars and oil-gas tars would be a subset of this list, with many of the tar acids and tar bases being present in coal tar appearing only as trace constituents in water-gas and oil tars.

Raw tars generally did not have very much product value. They could be burned in the plant boilers for steam production, burned under the benches used for coal carbonization, sold as boiler fuel to a local company, or discarded. Tars were a resource to most companies, a byproduct that was sold and produced income. Near the beginning of the industry, tars were disposed because uses had not yet been developed for them; later, tar/water emulsions were disposed when they could not be separated. Small plants that did not produce sufficient tar for recovery or use would discard it rather than spend money to prevent its release.

Tars were distilled into fractions that could be marketed as products, and the fractions were frequently treated with acid and caustic washes to improve the tar quality and remove undesirable components. The gas purification system separated the recovered hydrocarbons into two fractions--the tar and light oil. The tar condensed with water or was removed with an ESP. The light oil was scrubbed out of the gas after the ammonia was removed.

The crude light oils (either recovered by the process described in Section 1.3.3 or distilled as the highest boiling fraction of the tar) were

TABLE 30. PRINCIPAL COMPONENTS IN COAL TAR FRACTIONS

Tar fraction	Boiling range[a] (°C)	Major components
Light oil	To 210	Benzene
		Toluene
		Xylene
		Tar acids
		Tar bases
		Solvent naphtha
Middle oil	210-230	Tar acids
		Tar bases
		Naphthalene
Methylnaphthalene	230-270	Mixed methylnaphthalenes
Light creosote	270-315	Acenaphthene
		Diphenylene oxide
		Fluorene
Middle creosote	315-355	Phenanthrene
		Anthracene
		Carbazole
Heavy creosote	Above 355	Chrysene
		Fluoranthene
		Pyrene

SOURCE: Smith and Eckle, 1966.

[a]As determined by ASTM test D20-56.

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usually treated with sulfuric acid prior to additional refining. The light oil was charged to an agitator (5,000 to 13,000 gallons) to which strong sulfuric acid was slowly added (66 deg Baume). It was frequently added in small amounts, followed by removal of the acid and the sludge it contained. The total acid consumed was about 0.4 pounds of 66 deg Baume sulfuric acid per gallon light oil treated. The acid layer was removed after 6 to 8 hours of treatment in the agitator, and the remaining acidity of the oil was neutralized by adding 0.06 to 0.12 pounds of sodium hydroxide per gallon of oil. Several beneficial reactions occurred during the acid treatment of light oil. These included oxidation and/or removal of sulfur compounds, the removal of nitrogen bases into the acid, the polymerization of unsaturated organic compounds, the sulfonation of aromatic compounds, the oxidation of unstable hydrocarbons, and the polymerization of certain aromatic hydrocarbons (Glowacki, 1945).

The acid sludge waste is a waste product from plants that produced the light oils. Although the volume produced by the midsized plants was not particularly large, its acid character and high concentration of tar bases is cause for concern. This sludge was sometimes treated for the recovery of the unused sulfuric acid, but it was frequently just dumped or poured somewhere and burned. It was not burned in boilers because of the high sulfur content (placing sulfuric acid into boilers is usually not a recommended practice because of the resulting corrosion). The acid sludge from light oils recovered from oil or carbureted water gas would be of substantially different character from that of coal-carbonization plants. The nitrogen bases would be present in the acid sludge from coal carbonization, but they would be absent from acid sludge produced from oil-gas and carbureted water-gas production.

The basic technique for separating the tar and light oil into marketable fractions was distillation. The distillation could be performed either continuously or by batch distillation. In both types of distillation, the oil or tar was separated into fractions with similar boiling points. The batch still was first charged with tar, and the still was heated slowly. The lower boiling fractions of the tar vaporized preferentially at lower temperatures, and these components were condensed and recovered as a liquid. Condensed fractions of the tar were removed at various times (corresponding to different

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still pot temperatures). Each of the collected fractions (they were recovered as the fractions described in Table 30) had compositions and properties that were generally more desirable than those of the original raw tar.

In continuous distillation, the tar is fed continuously to a distillation column containing multiple fractionation trays. The bottoms of the column are continuously boiled, producing vapor that flows up through the trays. The vapors from the top of the column are condensed and a portion of the condensate returned to the top of the column. This liquid (and the feed) flow down the column from tray to tray. A temperature profile exists within the column, and the liquid composition existing on each tray is different, with the higher trays having a higher concentration of volatile components and the lower trays containing more compounds that boil at high temperatures. Various fractions of the tar can be removed at several of the trays. Because it operates in a continuous manner, continuous distillation was usually employed at the larger tar-processing facilities. Batch distillation was used early in the industry and at smaller processors.

Tar-processing operation sites would have had much more handling and treatment of the tars than did plants that merely recovered tar and sold it to tar processors. In many cases, a tar processor was located adjacent to the gas plant and could receive the tar byproducts directly from the gas production plant.

1.3.10 Gas Storage

This section describes how gas was stored at town gas facilities. Tanks that were used for the storage of product gas were also frequently used for the storage of tars and waste condensates at gas production plants. Because these tanks frequently leaked, they were a significant source of contamination.

The operating basis for the early gas holders was originally discovered by the French chemist Lavoisier in 1781. His lab-scale gas holder consisted of an inverted cylindrical bucket in a tub of water. The bucket was suspended from a cord attached to its bottom, where the cord was run through a pulley and attached to a counterweight. When gas was placed into the holder, the bucket rose. The water in the tub formed a seal around the bucket. When gas was removed from the bucket, it dropped farther into the water. This arrange-

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ment allowed gases to be collected and removed for experimentation. The earliest gas holders used by the manufactured-gas industry were of this same basic design but larger.

Figure 32 shows a diagram of an early single-lift gas holder. The water-holding portion of the gas holder was usually placed underground or partly underground. This allowed the earth to support the walls of the water-holding tank and reduced construction costs. The plant operators soon discovered that tars could be stored in the gas holder instead of water. This reduced corrosion of the tanks and allowed the gas holder to serve as a tar tank in addition to its use for gas storage. Even when tar was not stored in the tanks, the water contained in the tanks became fouled by water-soluble and organic compounds in the gas.

The early gas holders used masonry tanks for the water and iron plates for the bell itself. Alrich (1934) describes the early masonry tanks:

> The important consideration of holder tanks in the earlier years of our Industry was the necessity for water tightness; not only did foul water leaking from the tanks contaminate the water in wells upon which even populous communities relied for their supplies, but the holders [were] frequently located closely adjacent to dwellings, [and] the buildings were rendered uninhabitable by the foul water entering through cellars.

He also states that the soils in England were much better suited to the construction of watertight masonry tanks, and when the same designs were applied in the United States they leaked rather badly.

Many plants also lost substantial quantities of condensate water through leakage. Because this water was generally recycled to the scrubbers, the loss of water had to be made up from other external sources. "The question frequently arose, 'Why does one gas plant have an excess of water and another plant apparently have none?' Upon investigating this question we found that in every case where a gas plant had no excess water there was a pit holder or some other leaking underground structure through which excess water was undoubtedly leaking into the ground" (Bains, 1921).

The single-lift gas holder had one obvious problem, the depth of water in the tank had to be the same as the height of the bell. To increase the size of the gas holder without increasing the size of the tank, the telescopic or multiple-lift gas holder was used. Figure 33 is a diagram of a multiple

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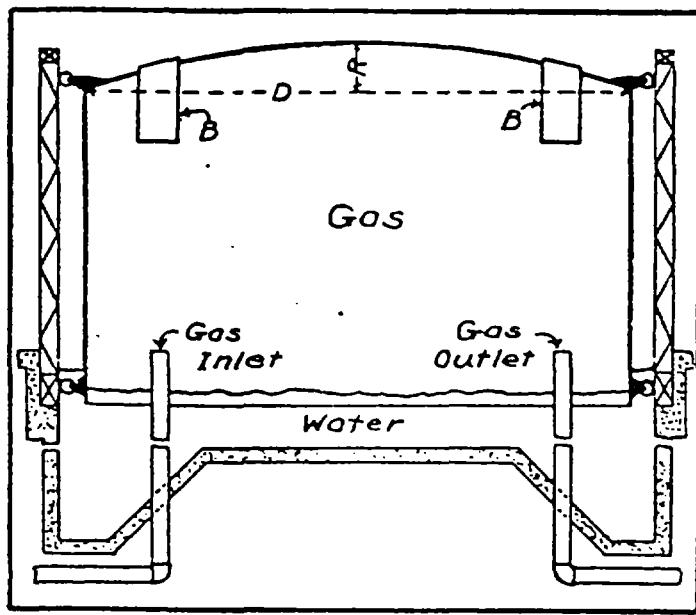
Figure 32. Cross section of single-lift gas holder.
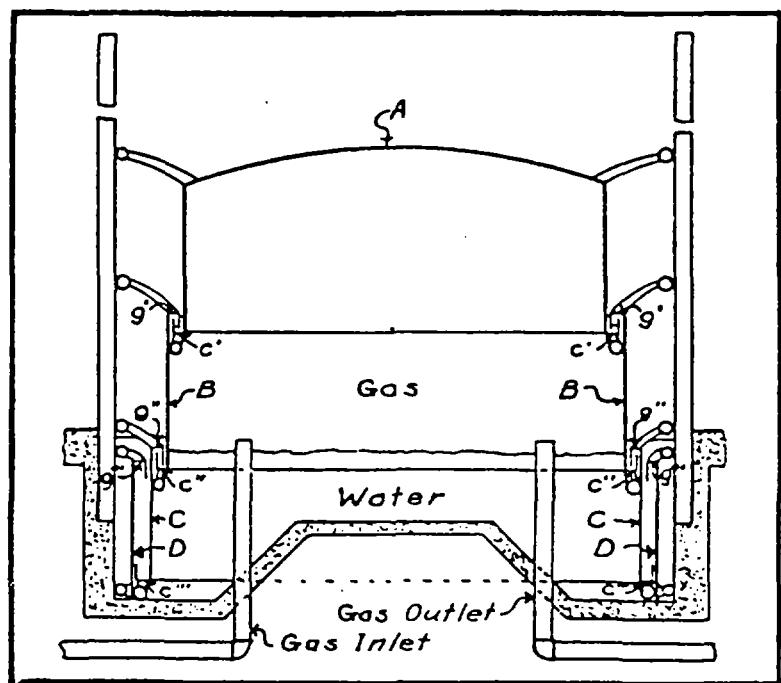
Source: Morgan, 1926.

**Figure   33. Cross section of multiple-
            lift gas holder.**

Source: Morgan, 1926.

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(four-lift) gas holder. The top section (A) would fill with gas first, and its base would reach the water level in the tank. The top section would then form a seal with a second lift (B), and together sections A and B would hold the gas. Subsequent sections would automatically be picked up by the gas holder as it filled, and the sections would each collapse into the tank as gas was withdrawn. This allowed greatly increased storage capacity over single-lift gas holders. There could in principle be any number of lifts, but in general fewer than five were used. The raised area of the concrete tank (dumpling) for Figures 32 and 33 allowed the tanks to be constructed with less excavation of the plant site and the tank to operate with less water. More concrete is required for this construction than for flat-bottomed tank construction. The tank bottom was usually flat for the early gas holders or smaller gas holders.

By 1926, the use of brick to construct the water-holding tank was obsolete (Morgan, 1926). Tanks during this period were constructed of steel plates, and the water tanks could be either below ground, semiburied, or above ground. For very large tanks, buried or semiburied concrete construction was used. Small gas tanks were typically constructed above ground, with the entire tank structure resting on a concrete slab. Any leakage from this type of tank would be readily visible to the operators.

Waterless gas holders were used at some plants after about 1925. These were cylindrical tanks that contained a free-floating piston that would move up and down within the tank as the volume of gas stored changed. The piston was usually sealed around the edges of the tank by a tar seal (a seal applied by some mechanical means with a layer of tar above the mechanical seal). This tar would slowly leak down the inside walls of the tank, collect at the bottom of the tank, and be pumped back to the floating piston seal. Waterless gas holders were generally used for very large (500,000 to 15,000,000 ft^3) tanks, and the water-sealed gas holders were used for smaller tanks. The tar used to form the seal was generally produced somewhere within the plant.

The gas holders previously described held gas at constant pressures slightly greater than atmospheric pressure. The volumes of the tanks were required to change as the amount of gas stored changed. High-pressure gas storage tanks were installed at some plants during the 1920's, but they were

not in common use until after World War II. These steel tanks store gas under high pressures so that larger volumes of gas can be stored in smaller-sized tanks. With high-pressure storage, the pressure of the gas in the tank can be changed as the amount of gas stored is varied, rather than having the tank volume change.

1.3.11 General Purification Trains for Town Gases

The processes for the production of town gases are described in Section 1.2, and Sections 1.3.1 through 1.3.9 describe the various methods of purifying the raw gases prior to distribution. This section integrates the production and purification processes by examining several complete town gas production facilities. These descriptions are not intended to be representative of all of the plants using a given production process, but they will help to give readers generic descriptions of town gas plants.

Figure 34 shows a material flowsheet for a typical coal gas plant. This flowsheet indicates a plant in which bituminous coal is carbonized to produce coke, tar, and ammonia. Bituminous coal is first crushed and fed to the coal-carbonizing apparatus (Section 1.2.2). The coal is carbonized to produce coke and gas (containing tars and other byproducts). The coke is used to manufacture producer gas (Section 1.2.1) to heat the coal-carbonization apparatus; it can also be sold or used to produce carbureted water gas (Section 1.2.3). Coke breeze (coke of small particle size) is used in the boiler room. The raw gas is scrubbed with weak ammonia liquor in the hydraulic and foul main, then it is cooled in the primary condenser (Section 1.3.2), blown through the exhauster, and tars are removed by the tar extractor (Section 1.3.3). The collected tars and condensate are combined and fed to a tar-liquid separator. Weak ammonia liquor and tar are separated, and the tar is either processed further or sold as raw coal tar. The ammonia is then scrubbed from the gas (Section 1.3.5), and H_2S is removed by liquid or iron oxide purifiers (Section 1.3.7). The purified gas is then metered, stored, and distributed to consumers.

Figure 34 shows a flowsheet that would be typical of small-to-midsized coal-carbonization plants whose primary purpose was the production of fuel gas. No recovery of light oils was performed, but the organics that condensed in the storage and distribution system were recovered as drip oils. The light

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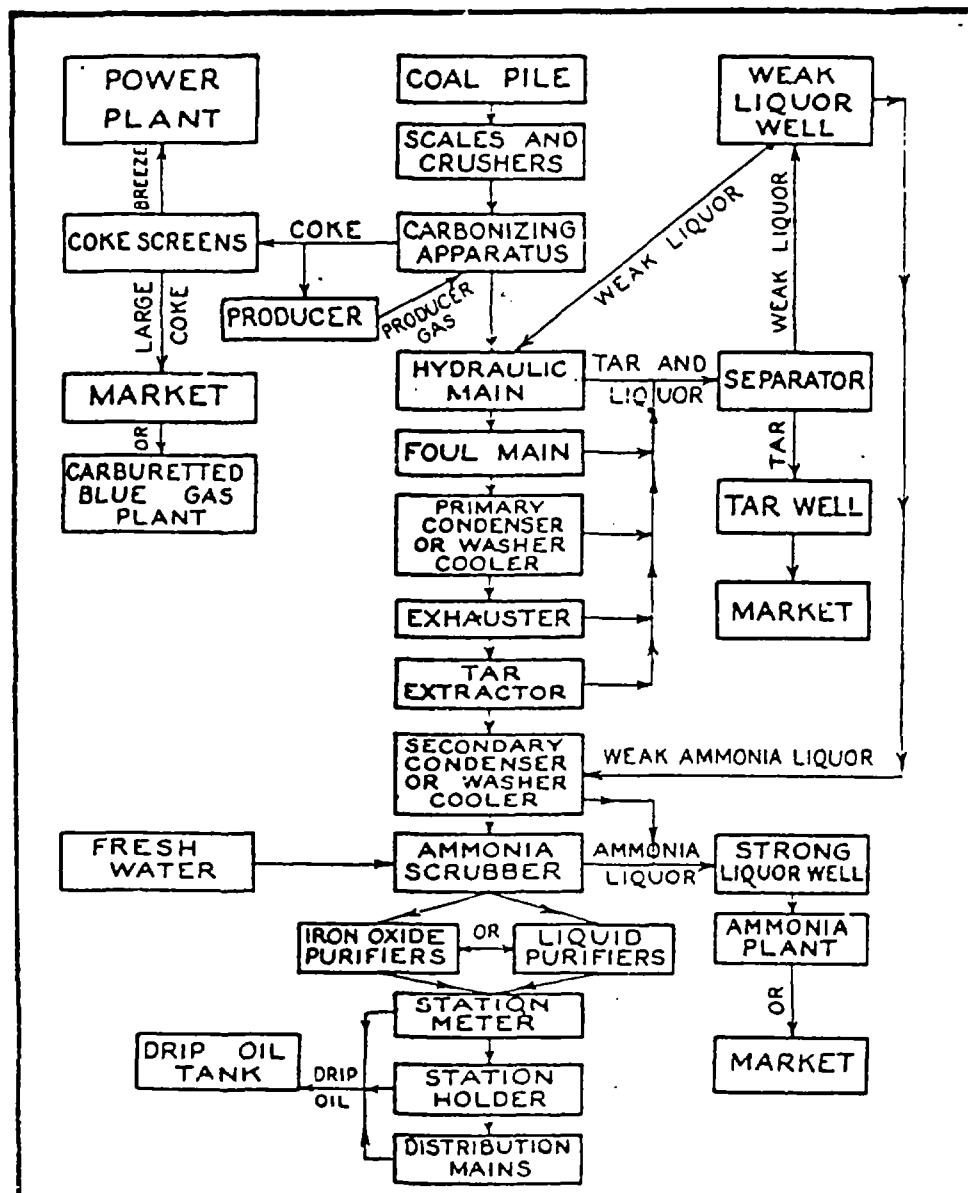
Figure 34. Flowsheet for a coal carbonization gas plant.

Source: Morgan, 1926.

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organics that did not condense would just enrich the fuel value of the gas when the gas was burned. Phenols would be in the ammonia liquor, and the ammonia liquor could either be sold in this form or the ammonia could be recovered onsite (Section 1.3.5). The ammonia recovery in this figure is the indirect process. Cyanide was removed in the iron oxide or liquid scrubbers, and it was not recovered as a product.

Figure 35 is a flowsheet similar to the one shown in Figure 34, but it is for a modern (1945) byproduct coke-oven plant. It is indentical to Figure 34 except that (1) phenol is shown recovered from the ammonia liquor (Section 1.3.6), (2) ammonia is recovered by the semidirect process (Section 1.3.5), (3) light oil is recovered, and (4) liquid purification (Section 1.3.7.4) is employed for the removal of H_2S and HCN, with the recovery of both sulfur and thiocyanates. This flowsheet would be typical of large byproduct coke ovens. The products of the process are coke, gas, tar, sodium phenolates, ammonium sulfate, light oils, sulfur, and ammonium thiocyanate. Although some plants would recover all the byproducts as indicated by this figure, there would be many variations of this basic design. As an example, some plants would not recover light oils, use iron oxide purifiers for H_2S removal, or use the Seaboard process for H_2S removal and not recover sulfur. Moreover, some plants would not recover thiocyanates as a product, would not recover phenols (they would dispose of them instead), or would not recover ammonia.

Figure 36 shows a material flowsheet for the production of carbureted water gas, which is described fully in Section 1.2.3. The generator contains a carbon fuel (either coke, anthracite coal, bituminous coke, or petroleum coke briquets). Air to the generator, superheater, and carburetor is supplied by a blower. Carburetion oil is pumped from storage, preheated, and sprayed into the carburetor. The carburetion oil could be naptha, gas oil, fuel oil, or heavy residual oils. Waste heat produced during the blows is passed through a waste heat boiler, which produces the steam sprayed through the generator. Raw gas is passed through a washbox and condenser (Section 1.3.2). Because the production of gas is not continuous, a relief holder (Section 1.3.10) is used to dampen the gas flowrate changes and provide a relatively constant flow through the exhauster, tar extractor (Section 1.3.3), purifiers (Section 1.3.7.3), and finally to the metering and distribution system. Tars

COAL

HYDRAULIC MAIN PRIMARY COOLER EXHAUSTER TAR EXTRACTOR & REHEATER SATURATOR ACID SEPARATOR FINAL COOLER BENZOL WASHER LIQUID PURIFICATION PLANT THIOCYANATE PLANT GAS HOLDER

OVENS GAS GAS GAS

SULFURIC ACID

AMMONIA VAPOR

TAR & AMMONIA CONDENSATE

STRIPPED WASH OIL

ENRICHED WASH OIL

DRAIN TABLE

DECANTER

LIGHT OIL STILL

AMMONIA STORAGE WEAK AMMONIA LIQUOR AMMONIA STILL DRIER

LIGHT OIL

AGITATOR

PURE STILL

TAR STORAGE DEPHENOLIZER SALT STORAGE

STORAGE

MOTOR FUEL BENZENE, TOLUENE SOLVENT NAPHTHA

COKE TAR SODIUM PHENOLATE AMMONIUM SULFATE SULFUR AMMONIUM THIOCYANATE GAS

Figure 35. Condensing and collecting system of a modern byproduct coke plant.

Source: Rhodes, 1945.

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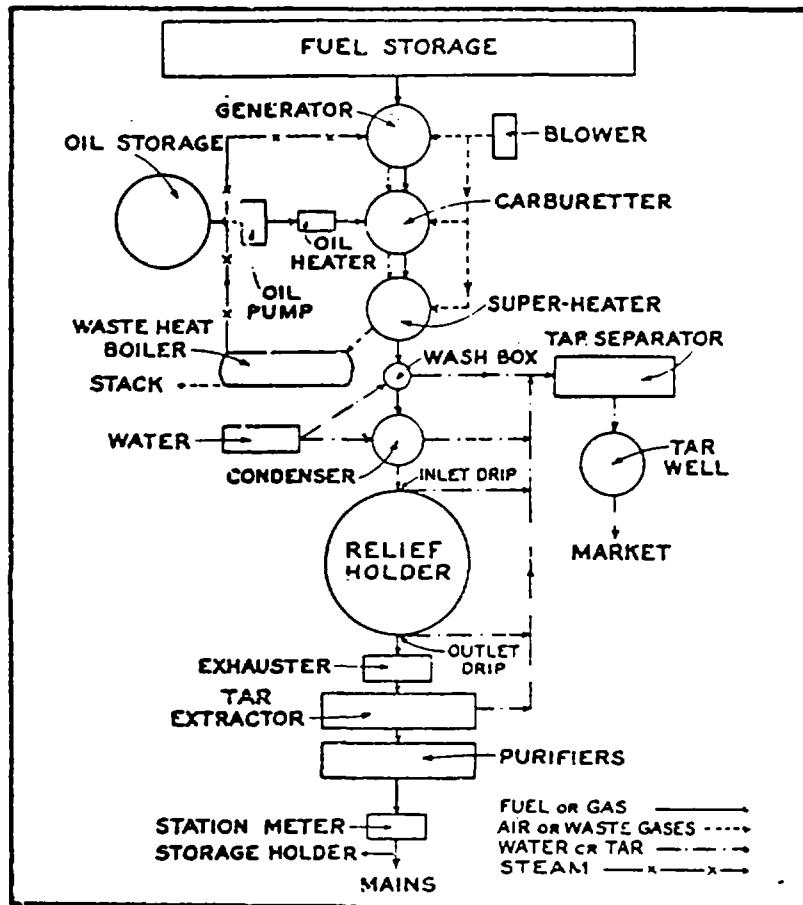
Figure 36. Flowsheet for a carbureted water-gas plant.

Source: Morgan, 1926.

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and condensate are collected from the washbox, condenser, relief holder, and tar extractor. The tar and condensate are then separated in the tar separator (Section 1.3.3). The product tar was frequently sold as a boiler fuel, burned in the plant boilers, or remixed with the carburetion oils.

This flowsheet is much simpler than are those for the coal-gas production. No ammonia is produced or recovered, no phenols are produced or recovered, and no cyanide is produced or recovered from the carbureted water-gas process. In fact, some small amounts of phenols, ammonia, and cyanides were produced by the process, but they were not in recoverable quantities and were in much smaller concentrations than were those in gas from coal carbonization. The purifiers generally used iron oxides, although liquid purification could be employed. Sulfur recovery was practical at some of the larger plants or at those that used carburetion oils contain.ng a high concentration of sulfur. Light oils were not recovered in this flowsheet, but the organics condensing as liquids in the relief holder are collected and recycled to the tar separator. The tar extractor was frequently a tower packed with wood shavings in which entrained tar aerosols would either condense or be removed by impact with the shavings. The tars condensing in the tar extractor would drip to the base of the tower, then they would be removed and mixed with the other plant tars. The wood shavings required periodic replacement because heavier tars would eventually build up on the shavings and plug the shavings scrubber. This process was much better suited than coal carbonization for use in small gas plants. Less labor was required to produce gas, the gas was of generally high quality, and there were fewer byproducts (no ammonia, phenols, cyanides, and organic nitrogen compounds) to recover or dispose.

Very small gas plants producing carbureted water gas might only operate the gas production equipment during part of the day and rely on the gas storage holder to supply gas when gas was not being produced. The larger plants, however, usually operated several separate units (similar to that shown in Figure 36) to produce the quantities of gas required. Individual units (or sets) would be started up or taken out of production depending on gas demand.

Figure 37 shows a flow diagram for a typical oil-gas production plant, the Portland Gas and.Coke Company works. This diagram does not show the steam

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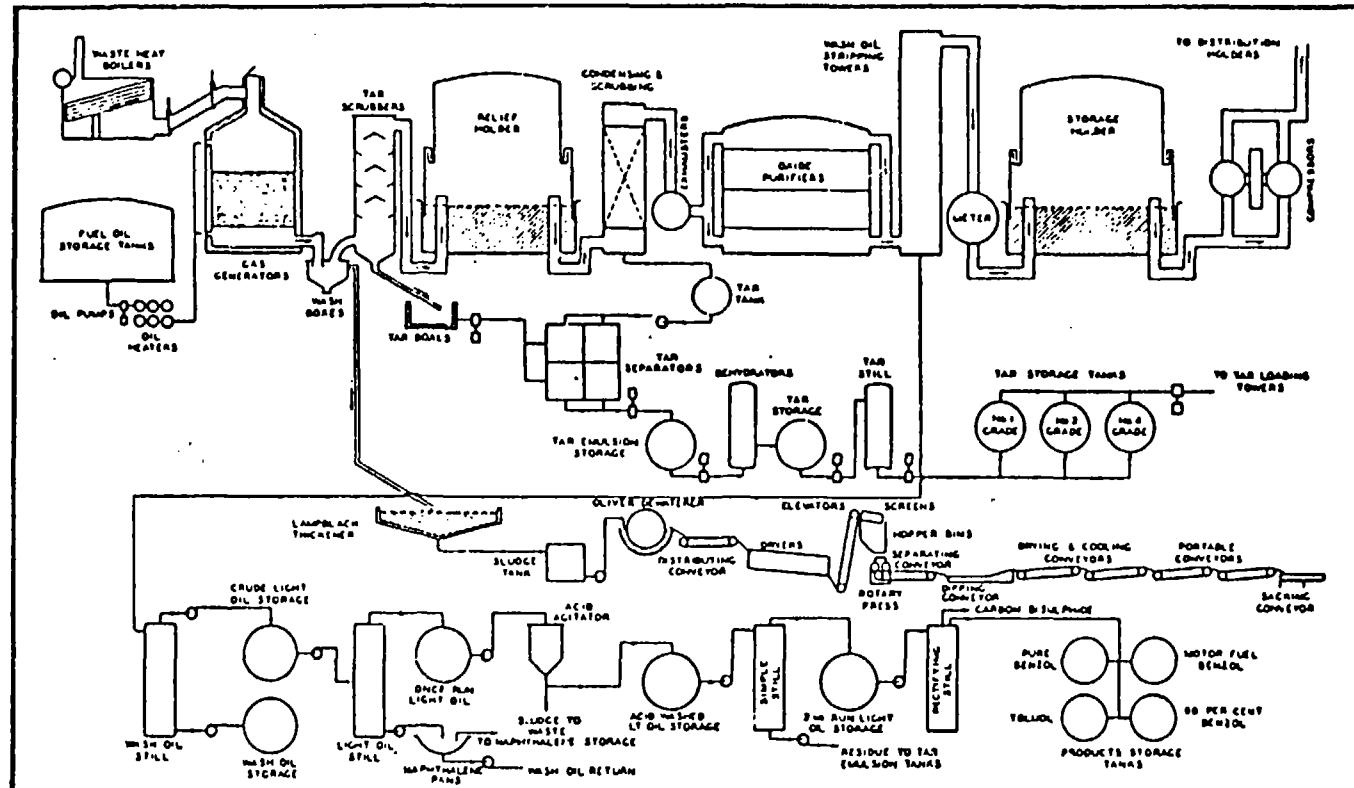
Figure   37. Flow diagram for gas manufacturing and byproducts of
Portland Gas and Coke Company works.

and air inputs that are required for the gas generators, but it does adequately show the gas-cleaning and byproduct recovery operations. Oil, steam, and air are used to produce gas in the generator (Section 1.2.4.2). The raw gas is initially cooled in the washboxes. Most of the lampblack that is produced by the process deposits in the washbox, along with the heaviest of the tars. The gas is then scrubbed at lower temperatures in the tar scrubber to remove tars (the scrubbing liquid is not shown on the figure, but it would usually be recycled water and condensate from the tar separators). The gas is temporarily stored in a relief holder, and it is then scrubbed to remove more tars prior to being exhausted into the iron oxide purifiers. After this step, the light oils are scrubbed from the gas, which is followed by the storage and distribution of the purified gas. Steps including the wash-oil to the light-oil scrubbers and the recycled condensate to the second condenser-scrubber are not shown.

The lampblack-heavy tar-water mixture is fed to a thickener to remove some of the water from the mixture. The thickened sludge is then dewatered, dried, packaged, and sold. The lampblack product could be sold as fuel, briqueted (for use in water-gas generators or sold as fuel), or burned in the boilers of the plant. The lampblack could also be slightly dewatered prior to burning in the plant boilers. Sometimes the lampblack was not recovered at all; instead, it was merely routed from the washboxes to an appropriate lagoon. The raw tar and condensates were separated in gravity tar separators (Section 1.3.3), which was followed by the dewatering of the collected tars. The product tars were then distilled into marketable fractions and sold. Wash oil containing light oils and naphthalene was regenerated by distilling the light oil and naphthalene from the oil. The light oil and naphthalene were then separated in a second still. The recovered light oil was then acid-washed and distilled into marketable fractions. The acid washing of the light oil produced a waste acid sludge, but this sludge would be substantially different from the acid sludge produced from the acid washing of light oil from coal carbonization (Section 1.3.4).

The oil-gas plants along the West Coast frequently operated as extensions of oil refineries. The petroleum refiners would sell residual oils with high carbon contents to the gas companies, and the gas companies would use it to

produce a wide array of petroleum products in addition to gas. The light oils and tars that were cracked from the oils had sufficient market value that the plants recovered and sold them, rather than recycle the light oils into the gas generation oils. The units were operated essentially as thermal crackers of petroleum, producing lampblack, tars, light oils, and gas as products.

The production of oil gas was not accompanied by the production of significant amounts of phenols, cyanides, ammonia, and base nitrogen organics. These constituents would not be recovered at plants that produced exclusively oil gas, and they would be present only in trace amounts in any wastes from the process. The amount of hydrogen sulfide produced in the gas was proportional to the sulfur content of the oils used in the generators. Sulfur recovery processes could be used to remove hydrogen sulfide from oil gas.

Many possible variations are possible for this flow diagram. Smaller plants whose primary purpose was the production of gas would probably not recover the light oils from the gas. Most of the light oils would remain in the gas and enrich the heating value of the distributed gas. Many plants would sell the raw tars to distillers, rather than distill it onsite. The recovery of lampblack could vary and would range between disposing of the washbox sludge and condensate to complete recovery and use of the lampblack. If the plant also produced gas by coal carbonization (e.g., as was done at the one in Seattle, Washington), the lampblack sludge could be mixed with bituminous coal prior to coking. The tar in the lampblack would be added to the recovered coal tars, and the carbon would be added to the coke produced.

1.4 BYPRODUCTS AND WASTES FROM TOWN GAS PRODUCTION

1.4.1 Introduction

Each of the three processes for the production of town gas also produced nongas materials that were not directly related to the production and distribution of combustible gas to consumers. These materials could frequently be recovered, recycled, or sold but were also disposed at some production plants. The only difference between byproducts and wastes is that, if a material could be sold or given away, it was considered a byproduct, but if the material were discarded it was considered a waste. This distinction between byproducts and wastes is somewhat unimportant for the types of waste disposed on or near gas

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sites because some byproducts would spill or leak at the site and off-spec byproducts may be disposed. It is important for the quantity of wastes disposed, however, because some materials were always disposed while others were frequently recovered.

Several factors affected whether a given byproduct was recovered or disposed. If there were no market or use for a material, it was considered a waste for disposal. Sometimes these wastes did have a value as fill (such as spent oxide, ash, lampblack, clinker, and broken firebrick) and were used as fill around the plant or given away as fill. Some potential wastes such as ammonia, phenol, lampblack, and tars could be recovered and sold, but they were often not recovered because the price for the material did not justify its recovery. Any material that was recovered at a gas site was a potential waste because some of the products would not meet marketable standards.

1.4.2 Description of Wastes

1.4.2.1 Coal Tar, Water-Gas Tar, and Oil-Gas Tar--

When most people think of tar, they generally remember the tars that they have seen. These are principally either road or roofing tar, which is usually a solid but pliable material that softens as its temperature is increased. The prospect of this tar flowing through the ground or contaminating water is remote, even to the casual observer. However, the raw tars produced by town gas processes were frequently liquids at ambient temperatures with viscosities sometimes not too different from water. Tars were considered to be any organic liquid that was more dense than water (density > 1 g/cm^3). The tars would sink to the bottom of the tar separators, with the water forming a separate layer above it. The tars that collected in this manner generally had organic compounds normally associated with light oils, but they were dissolved in the heavier tar layer. The range of tars produced for the manufacture of town gases was considerable, ranging from tars that were slightly more dense and viscous than water, to tars that were solid at ambient temperatures and required heating before they could flow. Raw tar properties varied substantially within individual production processes because the heavier tars usually condensed in the washbox and lighter tars in the condensers.

Tar was usually defined as a nonaqueous viscous liquid of very complex composition produced by the destructive distillation or partial combustion of

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organic matter. The tars produced by town gas processes fit into three general categories, depending on the production process. Coal tars were tarry liquids produced by the partial combustion or destructive distillation of coal. They were usually further classified by the specific process that produced the tar, but they were divided into two major classes: high-temperature tars and low-temperature tars. Coal tars contained principally aromatic hydrocarbons: benzene, naphthalene, anthracene, and related compounds. They also contained phenolics and tar bases.

Oil tars were tarry fluids produced by the destructive distillation or thermal cracking of petroleum oils. The tars produced by the major oil-gas processes were high-temperature oil tars. They were composed principally of aromatic hydrocarbons; benzene, toluene, naphthalene, phenanthrene, and methyl anthracene were reported components. Other complex aromatic hydrocarbons are also present. "No true anthracene has been identified in any of the American oil tars. They are further characterized by the almost entire absence of tar acids and tar bases, and this seems to constitute the chief difference between this type of tar and high temperature coal tar" (Bateman, 1922).

Water-gas tar is the tar produced from the oil that is cracked from petroleum oils in the carburetor of carbureted water-gas (CWG) machines. Water-gas tar is very similar to tar produced by oil-gas manufacture. It also is very similar to coal tar but "could be distinguished from coal tar only by its lack of phenolics and tar bases" (Bateman, 1922).

Table 31 shows typical analyses for various types of raw coal tars. The tars from horizontal retorts, vertical retorts, inclined retorts, and coke-oven coal tars are listed. Table 32 compares the properties of two CWG tars to three types of coal tars. Table 33 lists the properties of three oil tars produced by the Pacific Coast oil-gas process. The properties listed in these tables are those of the raw tars produced by the processes. These properties reflect the mixing of the tars that condensed in various parts of the purification train. The properties of the tar condensing in various parts of the purification train would be substantially different. Tars condensing in the hotter portions of the purification train (e.g., the washboxes) would be higher boiling and more viscous than would be tars condensing in the cooler sections of the purification system (e.g., secondary scrubbers or the tar extractor).

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TABLE 31. PROPERTIES OF COAL TARS[1]

Test No.[2]		Gas-works coal tars			Coke-oven coal tar	Blast-furnace coal tar	Gas-producer coal tar	Low-temp. coal tar
		Horizontal retorts	Inclined retorts	Vertical retorts				
1	Color in mass	Black	Black	Black	Black	Brownish-black	Brownish-black	Brownish-black
2a	Homogeneity to the eye at 77°F	Gritty	Gritty	Smooth	Smooth to gritty	Smooth to gritty	Smooth to gritty	Smooth
2b	Appearance under microscope	Lumpy	Lumpy	Sl. lumpy	Sl. lumpy	Lumpy	Lumpy	Sl. lumpy
7d	Specific gravity at 77°F	1.18-1.33	1.15-1.25	1.08-1.16	1.15-1.26	0.95-1.10	1.12-1.20	0.95-1.12
Ni	Engler viscosity at 212°F (100 cc)	150-650	100-300	25-30	30-110	75-100	100-∞	25-50
15c	Fusing point (cube method)	Below 40°F	Below 30°F	Below 25°F	Below 25°F	Below 25°F	Below 50°F	Below 25°F
19	Fixed carbon, per cent	15-40	15-40	15-30	14-40	10-25	10-30	8-15
20a	Distillation test:							
	Distillate to 315°C, by vol., per cent	10-35	15-45	25-55	20-45	35-55	30-50	30-80
	Sp gr ditto at 60°/60°F	1.02-1.04	1.02-1.04	1.00-1.01	1.02-1.05	0.94-0.98	0.95-0.97	0.95-1.00
	Tar acids in ditto, per cent	5-20	10-25	20-30	0-12	20-30	5-15	20-50
	Residue at 315°C, by vol., per cent	60-90	55-85	40-70	55-80	45-65	50-70	20-70
21	Soluble in carbon disulphide, per cent	60-85	80-85	94-98	83-97	65-80	60-95	90-100
	Nonmineral matter insoluble, per cent	16-40	15-20	2-5½	3-17	10-25	5-35	0-7
	Mineral matter, per cent	0-½	0-½	0-½	0-½	10-15	0-25	0-1½
22	Carbenes	0-2	0-2	0-2	0-2	0-2	0-2	0-1
23	Soluble in 88°F petroleum naphtha, per cent	20-40	20-60	60-80	40-75	50-70	50-75	75-90
24	Free carbon, per cent	20-45	10-25	2-8	2-20	15-30	5-30	3½-10
25a	Water in "dehydrated" tar, per cent	0-5	0-5	0-5	0-5	0-5	0-5	0-5
28	Sulphur, per cent	Tr.-1	Tr.-1	Tr.-1	Tr.-1	Tr.-1	Tr.-1	Tr.-1
31	Tar acids in tar, per cent	1-4	4-6	5-11	1-5	5-15	3-9	10-30
32	Naphthalene, per cent	3-8	2-5	0-4	7½-15	0-2	0-5	0-2
33	Solid paraffins, per cent	0-Tr.	0-2	0-5	0-Tr.	3-15	0-3	3-15
34a	Soluble in conc H2SO4, per cent	98-100	96-99	95-98	98-100	90-95	98-100	90-95
34b	Sulfonation residue of distillate 235°-315°C, per cent	0-5	5-10	5-10	0-5	15-25	0-5	5-25
37c	Saponifiable matter, per cent	2-5	2-5	7½-15	2-5	10-15	5-15	15-40
39	Diazo reaction	Yes	Yes	Yes	Yes	Yes	Yes	Yes
40	Anthraquinone reaction	Yes	Yes	Yes	Yes	No	Yes	No

[1] From Abraham, "Asphalts and Allied Substances," 3d ed., p. 277, 1929; courtesy D. Van Nostrand Co., Inc.

[2] For details of test methods see above source reference.

Source: *Gas Engineers Handbook*, 1934.

TABLE 32. COMPARATIVE ANALYSES OF TYPICAL, DEHYDRATED, WATER-GAS TARS AND COAL TARS PRODUCED IN 1921 to 1922

Property	Water-gas tar No. 1	Water-gas tar No. 2	Low-temperature coal tar	Coke-oven coal tar	Horizontal-retort coal tar
Specific gravity at 15.5°C	1.061	1.125	1.105	1.196	1.240
Free carbon, per cent by weight	0.26	1.64	2.9	0.9	22.1
Tar acids, per cent by volume	0.00	0.00	31.7	4.2	4.4
Sulfonation residue, per cent by volume	8.3	5.2	1.9	Trace	0.7
Specific viscosity, Engler, 50 cc at 40°C	1.7	11.8	7.3	163.0	—
Float test, seconds at 50°C	—	—	—	26.0	24.0
Distillate (Engler) per cent by weight					
to 170°C	2.5	—	1.4	0.1	0.5
to 235°C	30.0	10.0	27.8	10.9	10.1
to 300°C	57.6	40.0	45.0	25.4	21.8
to 355°C	74.6	60.3	65.5	38.5	34.6
Specific gravity of distillate to 300°C	0.983	1.011	1.011	1.045	1.039
Specific gravity of distillate to 355°C	1.005	1.038	1.040	1.070	1.073
Specific gravity of fraction 300–355°C	1.080	1.093	1.099	1.121	1.135
Distillation residue at 300°C, per cent by weight	42.2	59.8	54.9	74.3	77.8
Specific gravity of 300° residue at 15.5°C	1.192	1.204	1.190	1.252	1.305
S.P. of 300° residue, C.W.	54°C	48°C	58.5°C	61.4°C	74.0°C
Free carbon in 300° residue, per cent by weight	1.5	6.1	0.2	12.1	33.0
Distillation residue at 355°C, per cent by weight	24.9	38.9	32.9	61.2	64.9
Specific gravity of 355° residue at 15.5°C	1.266	1.280	1.257	1.285	1.347
S.P. of 355° residue, C.A.	114°C	118.5°C	113°C	91.0°C	124°C
Free carbon in 355° residue, per cent by weight	18.5	20.8	25.2	18.5	41.8

Source: Rhodes, 1966b.

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TABLE 33. COMPARISON OF SOME PACIFIC COAST OIL-GAS TARS

Sample	1	2	3	4
Specific gravity at 60°F	1.206	1.297	1.334	1.317
Insoluble in CS₂, per cent by wt	12.5	24.2	30.7	28.7
Specific viscosity Engler, 50 cc at 40°C (104°F)	13.2			
Float test, sec at 32°C		247		
Softening point (ring and ball), °C			33.8	32.6
Distillation, per cent by wt.				
To 210°C (410°F)	5.8	2.7	1.2	2.7
To 235°C (455°F)	16.6	15.5	4.4	10.8
To 270°C (518°F)	26.1	20.6	8.0	14.5
To 315°C (599°F)	33.6	24.2	13.4	18.4
To 355°C (671°F)	41.3	31.0	23.2	27.6
Residue and loss	58.7	69.0	76.8	72.4
Softening point residue (R. and B.), °C	105.5	140.0	137.0	148.5
°F	222	284	279	299
Specific gravity total distillate at 60°F	1.071	1.115	1.120	1.110
Sulphonation residue, total distillate (per cent vol.)	2.55	0.30	trace	1.62

Sample 1: Med.-temp. fuel oil-gas tar.
Samples 2 and 3: High-temp. fuel oil-gas tar.
Sample 4: Mixture of fuel oil- and reformed-gas tar.
Sample 5: Reformed-gas tar, 625 Btu operation.

Source: Pacific Coast Gas Association, 1926.

In general, the raw CWG tars were less dense and less viscous than were tars produced by coal carbonization. The low-temperature coal tar in Table 31 has a lower density and viscosity than do the coal tars, and this reflects the lower temperature of carbonization. Low-temperature coal carbonization was not employed in the United States to any great extent, however. The lower viscosity of the CWG tars means that they are generally more mobile and flowable than are the raw coal-gas tars. They generally also have a much lower carbon content than do the coal tars. The specific properties of the CWG tars depended substantially on how the plant operated the CWG apparatus. Because gas production occurred in cycles, the carburetor and superheater started out very hot when the oil was first injected into them to produce gas. They cooled relatively rapidly, requiring that the production of gas be stopped and the apparatus reheated. Hence, the cyclical nature of the process actually alternated between heating the apparatus and cooling the apparatus while producing gas. When the gas production part of the cycle began, the apparatus was at its highest temperature. The high temperature tended to overcrack the oil, producing very heavy tars, carbon, and gas. As the apparatus cooled, the lower temperatures tended to undercrack the oils, merely vaporizing the oils. An apparatus that was operated at higher temperatures produced tars that were higher boiling, denser, more viscous, and had higher carbon contents than did an apparatus operated at lower temperatures. Apparatus operated at low temperatures produced tars that more resembled the original feed oils.

The oil-gas tars (Tables 32 and 33) highly resembled CWG tars because both were produced principally by the thermal cracking of petroleum products. The discussion above regarding the properties of CWG tars as related to the operation of the gas-manufacturing apparatus applies to the production of oil-gas tars as well.

The tars produced by oil-gas and CWG production are very similar, and it would be very difficult to distinguish between the two. The oil-gas tars, however, would generally have higher carbon contents than would CWG tars. The petroleum-based tars (CWG and oil tars) can be distinguished from coal tars by the presence of phenols and nitrogen-containing organics in the coal tars.

The amount of tar produced by CWG or oil-gas production depended on the oil used for the gas manufacture. The first carburetion oils used were the

naphtha fraction of petroleum. This was the fraction that was between gaseous
hydrocarbons and kerosene. It made an excellent carburetion oil and produced
only a small amount of tar (which was probably not worth recovering). The tar
produced when using naphtha was only 1.7 to 3.5 percent of the original carbu-
retion oil (McKay, 1901). No analyses of the tar from naphtha were dis-
covered, but it would be very similar to that of the CWG tar in Table 32. The
tar from naphtha would probably be slightly less dense, less viscous, and
contain more lower-boiling hydrocarbons than would be tar from gas oils, and
it would be fairly mobile. After World War I, the increased demand for
gasoline (produced from the naphtha fractions of petroleum) led gas producers
to switch from naphtha to gas oils. The gas-oil fraction of petroleum was
between kerosene and lubricating oils. The gas oils produced more tars than
did the use of naphtha, CWG tars (produced using gas oils) were between 12.3
and 18.3 percent of the original oil volume (McKay, 1901). The use of gas
oils became less attractive after catalytic cracking of the gas-oil fraction
into heavy fuel oils and gasoline was adopted by petroleum refineries. This
alternative use of the gas oils competed with the gas industries' use of the
oils, increasing prices and causing some shortages of gas oil. The industry
subsequently switched from gas oil to heavy fuel oils for the manufacture of
CWG. The tars produced from the use of gas oils became know as light water-
gas tars, and those from heavy fuel oils or residuum oils were called heavy
water-gas tars. Table 34 is a comparison of light water-gas tars, heavy
water-gas tar, and coke-oven tar. The heavy water-gas tar was denser and more
viscous than was the light water-gas tar. It had much more carbon than did
the light CWG tar and fewer low-boiling organics. The use of heavy fuel oils
for carburetion also increased the amount of tar formed from the production of
CWG to up to 25 percent of the oil fed to the process.

Odell (1922) described water-gas tars as follows:

> In the carburetion of water-gas the aim is always to convert as much
> of the oil used as possible into fixed gases; the conversion, how-
> ever, is never 100 per cent complete, but invariably appreciable
> amounts of tarry condensable matter or carbon, or both, form in the
> checkered chambers. This condensable matter, which is water gas
> tar, may be composed of substances resulting chiefly from the crack-
> ing of oil, or it may consist, in part of some of the ingredients of
> the original oil which resisted cracking. As produced by the var-
> ious plants water gas tars are not uniform in character, but may
> very materially differ in their chemical and physical properties.

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TABLE 34. COMPARATIVE ANALYSES OF TYPICAL, LIGHT, AND HEAVY WATER-GAS TARS AND COKE-OVEN COAL TAR

Property	Light water-gas tar	Heavy water-gas tar	Coke-oven coal tar
Water, per cent by volume	0.5	1.1	—
Specific gravity at 15.5/15.5°C	1.089	1.212	1.198
Specific viscosity, Engler, 50 cc at 40°C	2.0	—	—
Float test at 32°C, seconds	—	74	38
CS₂ insoluble, per cent by weight	1.1	8.9	7.8
Distillation, Engler, per cent by weight			
to 170°C	1.0	0.2	7.3
to 235°C	12.1	4.2	—
to 300°C	44.6	16.4	21.5
to 355°C	67.6	31.8	35.8
Residue at 300°C, per cent by weight	53.4	83.1	—
Residue at 355°C, per cent by weight	31.9	67.9	—
S.P. of residue at 300°C, R & B	34°C	61°C	44°C
S.P. of residue at 355°C, R & B	99°C	96°C	75°C
Sulfonation index, 0 to 300°C	1.2	1.6	Trace
Sulfonation index, 300° to 355°C	0.2	0.8	Trace
Tar acids, per cent by volume	None	None	1.53

Source: Rhodes, 1968b.

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The tar may be brown in color, thin or watery in consistency, contain a large percentage of light oils, and have a specific gravity but slightly greater than 1.00, or it may be a black liquid of the consistency of molasses, containing a much smaller percentage of light oil and a specific gravity as high as 1.15. Furthermore, the percentages of free carbon and naphthalene are different in the various tars, varying from almost zero to a relatively high percentage.

The amount and character of coal tar produced by coal carbonization varies substantially with the temperature at which the coal is carbonized. As the carbonization temperature is increased, the amount of gas produced increases because more of the tars are converted to coke and gas. Figure 38. shows how the yields of gas, light oil, liquor and ammonia, tar, and coke change as the carbonizing temperature is increased. The carbonization temperature also affects the composition of the tar produced by the process. Figure 39 shows the effect of temperatures on the tar composition. As the amount of pitch residue in the tar is increased, the tar density and viscosity also increase. The carbonization temperature for coke ovens was about 850 to 900 °C, and the horizontal retorts operated at higher temperatures of 1,000 to 1,100 °C. Figure 39 shows that the tar produced from byproduct coke ovens would have more tar acids and less pitch than would tars produced from horizontal retorts. Rhodes (1945) gives a complete account of the effects of coal-carbonization temperature on the byproducts produced during the coking of coal.

Table 35 shows the amount of tar produced by each of the gas production processes. The estimates of the amount of tar produced for each process should be considered approximate but representative of the amount of tar produced by each process. This table shows that the production of CWG by naphtha produced very little tars, while oil-gas production was a very large tar producer. The large amount of tar produced by the oil-gas process reflects the use of high-boiling residuum oils for the production of oil gas. When heavy fuel oil was used for CWG production, the tar production increased relative to the tar production when gas oil was used.

The coal, CWG, and oil tars each had substantial uses that generally justified their recovery and use. It is not within the scope of this study to review the multitude of uses for tar products, but some mention of the major

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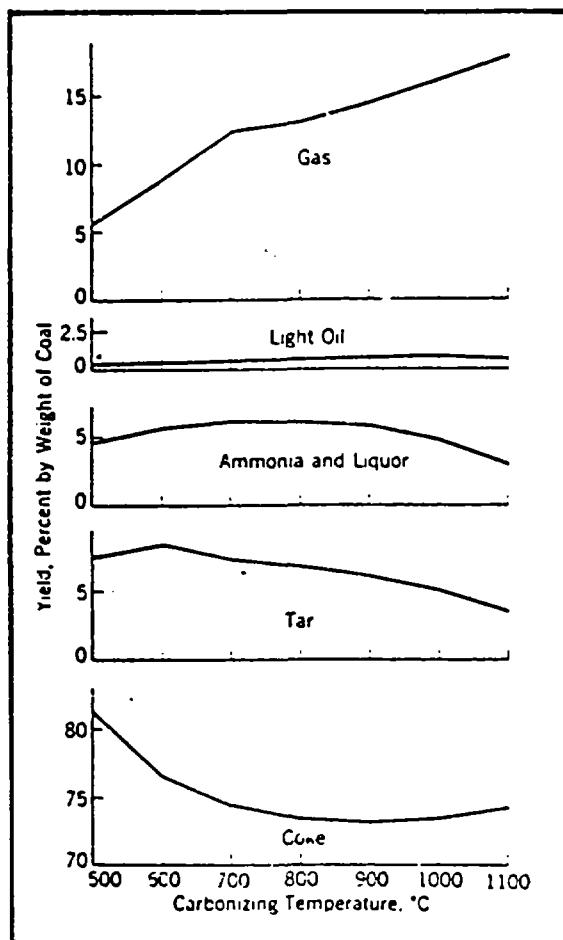
Figure 38. Effects of carbonizing
temperatures on yields of
primary products from Pratt
coal.

Source: Rhodes, 1945.

Note: Yields of primary products plotted
individually.

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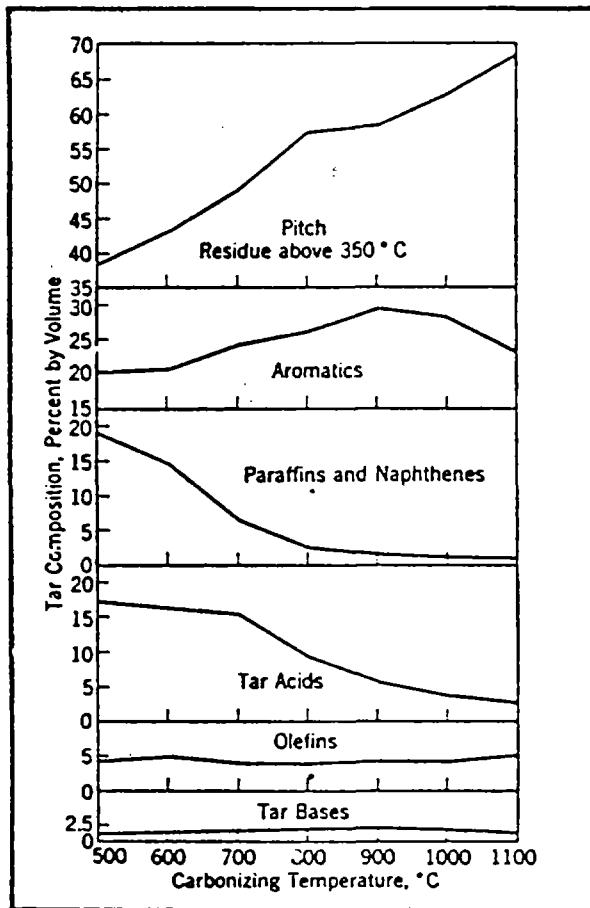
Figure 39. Effects of carbonizing temperatures on composition of tar from Pratt coal. Source: Rhodes, 1945.

Note: Concentrations of tar constituents plotted individually.

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TABLE 35. APPROXIMATE QUANTITIES OF TAR PRODUCED IN THE MANUFACTURE OF TOWN GAS

	Tar production		
Process	Gallons/ton coal carbonized	Gallons/10^6 ft^3 gas produced	Gallons/gallons oil fed
Coal carbonization			
Byproduct coke oven	8-12[a]	760-1,090[a]	--
Horizontal gas retorts	11-15[a]	820-1,090[a]	--
Vertical gas retorts (continuous, 10% steam)	11-22[a]	820-1,005[a]	--
Carbureted water gas			
Naphtha	--	50-120[b,d]	0.017-0.035[b]
Gas oil	--	470-640[b,d]	0.16-0.18[b]
Heavy fuel oil	--	850-1,080[d]	0.23[d]
Oil gas (Pacific Coast)			
Single-shell	--	2,100[c]	0.24[c]
Double-shell	--	3,000-4,000[c]	0.47-0.62[c]

[a]Gas Engineers Handbook, 1934.
[b]McKay, 1901.
[c]Morgan, 1926.
[d]Morgan, 1945.

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uses of tars is clearly appropriate. Rhodes (1945) divides the products from coal tar into two classes: the principal crudes and chemicals. The principal crudes are products produced directly from the distillation of raw coal tars. They include wood-preserving oils, road tars, industrial pitches. and pitch coke. The major basic chemicals produced from coal tar are naphthalene, tar acids (phenolics), and tar bases (nitrogen-containing organics). Paw coal tar also contains some light oils (about 2 percent) with lower-boiling organics such as benzene, toluene, and xylenes. The manufacture of coal-tar products has been reviewed by Smith and Eckle (1966), and the commercial aspects of coal-tar pitch have been studied by Doerr and Gibson (1966). Rhodes (1966a) examined the history of coal-tar and light-oil use, and he also examined the uses of coal and water-gas tars. The uses of heavy CWG tar and oil tar and the uses of light CWG tar are listed in Table 36. The principal use of CWG tars was as a fuel. The CWG tars could be burned in the plant boilers, replacing the coal that would normally have to be consumed. The tars therefore had a minimum value as fuel to the plant and would be burned if they could not be sold for a price that exceeded the fuel value of the tars.

The tars produced by town gas processes were generally recovered as a byproduct of the plant operations. There were several reasons why tars were disposed rather than recovered at gas production plants:

- Early plant operators disposed of tars rather than make attempts at recovery.

- The production of off-spec tars that could not be sold occurred, and these tars were either burned or discarded.

- Small gas plants were likely to dispose of tars in that they frequently did not produce enough tar to make recovery practical.

- Tars (particularly water-gas and oil-gas tars) frequently formed emulsions when the tars condensed with the steam. These emulsions could usually be broken, but when several attempts to break the emulsion into separate tar and water fractions failed, it was disposed. In some cases, plants would not even attempt to separate the emulsions. Instead, if the tar did not separate from the water in the gravity separator, it was disposed. (Emulsions are discussed in Section 1.4.2.3).

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TABLE 36. USES OF CARBURETED WATER-GAS TARS AND OIL-GAS TARS

Light carbureted water-gas tars (from gas oil)	Industrial fuel (burned in boilers) Road tars Waterless gas holder sealant (see Section 2.3.10) Fiber conduits (for electrical lines) Wood preservation Lampblack manufacture Paints and coatings Animal dips Mineral separation
Heavy carbureted water-gas tars (from heavy fuel oil) and oil-gas tars	Fuel Road tar Coal dispersions Pitch coke Electrode pitch Creosote (for wood preservation with PCP) Naphthalene Light oils

SOURCE: Rhodes, 1966b.

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Prior to the discovery that coal tar could be used to preserve wood in 1838, there were very few uses for the raw coal tars except as a fuel. Coal tar was not distilled in the United States prior to the early 1860's; hence, there was really no market for the raw tars. Many of the early plants disposed of the tar with the condensates from coal carbonization. This was done by whatever methods were most convenient for the plant, which generally meant running the wastes into the nearest body of water. Because most of the early plants were located along the coast, this was often done without causing noticeable problems. If the wastes could not be discharged to water, a pit or lagoon would often serve as a substitute.

Raw coal tar could be burned at the plant rather than be merely disposed. Undoubtedly, some plants did recover and burn much of the tar they produced during this early period. Hughes and Richards (1885) states that:

> When there is not a sale for the tar, or when there is a great demand for the coke, tar may be employed advantageously for heating the retorts, thus entirely replacing coke for that purpose...In a works having only six benches, or settings, the yield of tar would be sufficient to heat one of them.

Although Hughes' book was published in England, it was a seventh edition (the first edition being published around 1850), and it was probably common knowledge that the raw coal tar could be burned to heat some of the horizontal retorts. In any event, the early production of town gas was principally for the lighting of streets and shops, and then only during a certain portion of the evening. The gas produced was too expensive for people to use in their homes, and the amount of gas produced was relatively small until after 1865, when people started to use gas lights in their homes (Stotz and Jamison, 1938). It is probably impossible to reliably estimate either the total amount of tar produced during the early years of the industry or to determine how much of the tar was burned as fuel. The amount of tar disposed by methods in which it could be a hazard today would also be very difficult to estimate because much of the early waste tars were dispersed.

Some tars that were disposed by the plants early in their operation would not continue to be disposed during later operation. In 1896, Grimwood described the recovery of CWG tars:

> Aside from its commercial value and supposing it to be deficient in
> all the compounds which make coal tar a marketable residue, there
> arises the question of how to dispose of it without cost and annoy-
> ance to the neighborhood...In 1891 and 1892 all, or nearly all, our
> tar was a waste product; now we have a good and sufficient market
> for what we do not use...The raw tar is of no value except as
> fuel...Mixed with anthracite screening and coke breeze it makes a
> very fierce fire and serves admirably as a boiler fuel--a use to
> which I believe it is universally out.

Plants that recovered tars either for sale as raw tars or refined onsite into products would often produce tars not of marketable quality. Sometimes these tars could be mixed with better tars to produce an acceptable product, they could be burned or mixed with the coal prior to carbonization, or they could be discarded. The most common tar product likely to be discarded is the coal-tar pitch remaining in the still after the lighter fractions have been distilled from the raw tar. This tar had to be handled hot, in that it would solidify at ambient temperatures. Burning the tar meant that it had to be fed to a fire somehow, and the equipment for burning this heavy a tar would not be commonly available. Holding the tar for any length of time meant either heating it continuously or letting it solidify and then remelting it at a later time. Hence, the most expedient way of dealing with off-spec heavy tars was merely to add them to the waste dump.

Small gas plants had substantially less incentive to recover tars than did the larger gas plants. First, the small gas plants generally produced much less volume of tars than did larger plants. The least expensive way of dealing with the tars and condensate was to run them into a stream, or along the railroad tracks, or into a lagoon or pit. Because the volumes were gener-ally small, this method of disposal would not create immediate problems. Because CWG or oil gas was generally less expensive to produce at small plants, the disposal of waste condensates by this method was more common at these small CWG plants. Vincent (1907), in a discussion on the removal and disposal of tar, stated:

> I have noticed in a rather superficial investigation that probably
> the large majority of quite small gas companies are allowing the tar
> to run to waste, generally creating a nuisance in the community and
> also wasting a very valuable product...Tar can be burned under the
> boilers with equipment any ordinary workman can make: and while they
> cannot make enough to run the plant, the whole year around, they can
> make so much of it that it will ultimately reduce the cost.

J.A. Brown (1926) discusses the economics of removal and disposal:

> In the small plant the expenditure of every dollar is of such impor-
> tance in the monthly cost sheet that extraordinary caution is taken
> and resourcefulness exercised to avoid the expenditure. The small
> gas plant has nearly the same equipment as the large one, differing
> mainly in size. Any lack, loss, or failure to function results in
> much large loss in efficiency in the small gas operation. Any addi-
> tional labor or repair expense so looms up in the cost of gas in the
> holder that the small plant operator is particularly skilled at
> avoiding such expense.

Small gas plants were also more likely to use CWG using naphtha as a car-
buretion oil than were larger plants. They were slower in converting from one
oil feedstock to another because of the high capital cost of the conversion
relative to the quantities of carburetion oil they consumed. Naphtha produced
only a small amount of tar, and disposal of both the condensates and the tar
were very likely.

1.4.2.2 Oils--

   1.4.2.2.1 Carburetion oils--The carburetion oils used in the production
of oil gas and CWG were not intentionally disposed, but it was normal for some
of the feed oils to be spilled while transferring the oil or to leak from
storage tanks. These oils ranged from low-boiling naphtha fractions to
higher-boiling, high-carbon residuum oils. These oils would be, in general,
much more mobile in groundwater than would be the tars produced from the oils.
It is possible that at some gas production plants the major contamination
could come from either an old spill of the oils or a steady unnoticed leak
from oil storage tanks. The carburetion oils are described with their
respective processes in Section 1.2.

   1.4.2.2.2 Light oil--Light oils were recovered from oil gas, CWG, and
coal-carbonization gases (the process is described in Section 1.3.4). Light
oils were scrubbed out of the gas by a relatively heavy oil, then the light
oil was separated from the heavier oil by distillation. Light oils would not
be disposed as a waste, but leakage and spills of the scrubbing oils, or dis-
tilled light oils, could create local areas of contamination at gas plants.
The composition of light oils is described in Section 1.3.4. They are com-
posed principally of light aromatic hydrocarbons (benzenes, toluenes, and
xylenes).

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1.4.2.2.3 Drip oils--Drip oils were any hydrocarbons that condensed as a liquid in the gas holder, meter, or mains. After the gas was produced and cleaned, some hydrocarbons remained in the gas. As the gas cooled further to ambient temperatures, some of the heavier organics condensed out as a liquid. This organic condensate (or drip oil) was collected in special tanks at the low end of the gas mains. The drip oil was collected and either mixed with the raw tar or recovered light oils. It had a composition similar to the recovered light oil. Because the drip oils were collected in a separate tank (usually underground), some of the drip-oils could leak from the tanks and into the surrounding soil. Some of the drip-oil tanks remaining at gas sites may also be intact and may possibly contain the drip oils. Drip oils were not considered wastes because they could always be added to either the raw tar, light oils, or carburetion oils.

1.4.2.3 Tar-Oil-Water Emulsions--

The difficulties of recovering tars from tar-oil-water emulsions were one of the major headaches that plagued the operators of CWG and oil-gas plants. A tar with a high water content could not be sold (buyers specified low water contents for purchased tars), could not be burned (a water content below 25 percent is required for the combustion of the tar), and could not be disposed offsite (local sewer authorities would not permit the disposal of the emulsion in the sewer system, and the emulsions would contaminate a large amount of water if dumped into a river or lake). The emulsions were, in short, a problem nobody particularly wanted to deal with. As a relatively difficult material to separate, some of the gas plants disposed of the emulsions, rather than spend the time and effort to break the emulsion. The emulsions are tars that were very likely to be either stored for long periods of time or discarded.

Emulsions were not usually a problem in the production of coal-carbonization gas. The coal tar separated relatively cleanly from the condensates, and each could be recovered using only gravity separators. The formation of emulsions was a problem occurring primarily in the production of CWG and oil gas. There was really no problem with the formation of emulsions when paraffinic-based oils were used as the carburetion oils. The tars produced almost always separated cleanly. The emulsion problem began when the manufacturers of CWG

had to switch from paraffinic-based petroleum oils (produced in the East) to asphaltic-based oils (produced in the Texas Gulf region and California). The emulsion problems became even more acute when the oil feedstocks were switched from gas oils to heavy fuel oils. As described by Bennett (1935):

> Since 1903 gas oils of asphaltic base have been used. Lately heavy fuel oils have attained a wide-spread use as enriching material. Their use has invariably resulted in permanent emulsions which do not respond readily to the ordinary method of separation, i.e., settlement. The reason for this disturbing condition can be found in a brief examination of the petroleum industry. Pennsylvania crude oils (paraffine base) present no dehydraticn problem to the oil producer nor to the tar producer since emulsions in the field are unusual. As the field progress westward crude-oil emulsions steadily become worse and the ratio of asphalt to paraffine base oils becomes greater. The California fields in general produce the most stable emulsions and contain the highest quantity of asphalt bases. The carbureted water gas industry's shift, since 1903, has been a change from gas oils, principally of paraffine base, to oils which contain and produce more asphaltic constituents.

Numerous papers deal specifically with the problems of the formation and separation of tar-oil-water emulsions (Barlow and Kennedy, 1922; Hauschidt, 1922; Odell, 1922; Simmons, 1924b; Seely, 1927; Seely, 1928; Carswell, 1928; Morgan and Stolzenbach, 1934a; Morgan, 1934b; Parke, 1934b; Parke, 1935a; Dashiell, 1935; Bennett, 1935; Leuders, 1942; Petrino, 1947; Young, 1947; Hall, 1947; Glover, 1951; Laudani, 1952; Costigan, 1953; Costigan, 1954; Schwarz and Keller, 1955). The volume of information specifically addressing the problem of emulsions indicates both the size of the problem to the industry and the efforts expended to solve the problem.

According to Odell (1922), the emulsions are formed when the raw gas is cooled, and the water, tars, and carbon are removed simultaneously. The presence of uncracked oils in the tar and finely divided carbon made the emulsion more stable. Rapid cooling of the gas created emulsions because the droplet size of tar and condensate is very small, creating a more stable emulsion. The practice of dumping all of the plant tar and oils into a common receiver also assisted in the formation of emulsions. When the tars collected from different parts of the purification train were collected and treated separately, emulsion problems decreased.

A poll of 50 large CWG producers (Seeley, 1927) revealed that most of the plants had experienced emulsion problems at one time or another. Sixty-eight percent of the CWG manufacturers using coke as generator fuel reported emulsion problems, and 100 percent of the plants using bituminous coal in their generators reported problems with emulsions. All of the plants using oil with greater than 1.5 percent carbon reported emulsion problem, while only 80 percent of the plants using oil with less than 1.5 percent carbon reported emulsion problems. Of the 78 percent of the total plants reporting emulsion problems, only 32 percent had overcome the problems, while the other 68 percent still had emulsion problems. The most common solution to the problems was to raise the superheater temperature or change the grade of carburetion oil used.

The scope of the emulsion problems faced by individual plants can best be understood by examining the amount of emulsion produced by the plants. According to Morgan and Stolzenbach (1934):

> Carbureted blue gas plants using heavy oils produce on the average two to four gallons of emulsions per thousand cubic feet of gas. These emulsions contain about 60 percent of water. A medium sized plant producing 10 million cubic feet of carbureted water gas per day will produce from 20,000 to 40,000 gallons of tar emulsion. The usual ways of disposing of this tar are as fuel under boilers in the plant, and by sale to tar refiners. In either case, the tar emulsions must be dehydrated to a greater or lesser extent before disposal can be made of it. On account of the low value of the tar for either purpose, the dehydration process must be one that can be operated at low cost...Attempts to dehydrate the emulsions by the methods which have been developed in connection [with] ordinary water gas tar emulsions or oil field emulsions have not been successful in the case of most types of heavy oil tar emulsions. The heavy oil tar emulsions are better stabilized, and appear to be quite different from the types of emulsions which have previously been studied.

A plant producing emulsions would quickly find all of its liquid storage tanks filled, with nowhere else to store the emulsion. When all of the tanks at the plant had been filled, the plant operators were faced with either dumping the emulsions into pits or lagoons at the plant site or stopping gas production while they dealt with the emulsion problem. Very few plants would have shut down. Some of the heavier tars from the washboxes separate from the tar-water emulsion, reducing the higher-boiling organic content of the emulsion.

The tars contained in the emulsions would have essentially the same composition of the tars described in Section 1.4.2.1, except that some of the heaviest tar components would separate and be removed. Eventually, the disposed emulsions would separate into tar, water, and oil fractions. The tar fraction would sink in water, and the oil fraction would float on the surface of water.

Several methods were commonly used in the separation of tar-oil-water emulsions. The method that always worked for the separation of tar from water is the steam still. Water is simply distilled from the tar, leaving behind a dehydrated tar product. This method had two major drawbacks. First, it was relatively expensive, in that about 1.1 pounds of steam was required for each pound of water evaporated. Second, the temperatures involved caused substantial crosslinking of tar constituents, degrading the chemical value of the tars. Centrifuges were frequently used to separate tar from water. The spinning centrifuge basket separated the tar and water by density differences. The operation of the centrifuge was relatively expensive because it required frequent cleaning. The Warner tar dehydrator was essentially a steam still that heated the emulsion to 240 °F to cause a separation of the tar and water. The R.S. Dehydrator treated the emulsion with heat, pressure, and chemical reaction to separate the emulsions. A tank was filled with emulsion, and soda ash was added to the tank. The emulsion was heated to a steam pressure of 65 psig, then a valve to the tank was opened and part of the water in the emulsion flashed to steam and was withdrawn. The tar layer then usually separated and produced a tar with a water content of 10 to 12 percent water. In actual practice, the plants would try one or two methods of separating the tar from the emulsion, but they would probably dispose of the tar if their normal methods of tar handling were ineffective.

1.4.2.4 Waste Sludges--

1.4.2.4.1 Water purification sludges--One method used to purify waste condensate at many plants was to treat the aqueous waste stream with lime (or soda ash) and copperas (ferric sulfate) prior to discharging of the water. This process added 5 pounds of lime and 4 pounds of ferrous sulfate to the effluent water. The solids in the waste coagulated as small particles and settled as a sludge with about 10 percent solids and 90 percent oil and water.

This process produced about 1 ton of sludge per day when 72,000 gallons of water were treated per day at the Brooklyn Union Gas Company. Approximately one-third of the operating costs was for the removal of the sludge from the plant site (Murphy, 1928). This sludge would apparently be very similar to currently produced API separator sludge. The water purification sludge could be mixed with coke and burned in the plant's boilers, or it could be disposed at or near the gas production site.

1.4.2.4.2 Acid sludge from light oil agitators--Light oil from either the distillation of tar or scrubbing the gas was frequently treated with sulfuric acid to remove basic compounds and to improve the quality of the light oil. The recovery of light oil and its treatment by sulfuric acid is discussed in Section 1.3.9. Consequently, this section will deal principally with the characteristics and disposal of the sludge.

According to Powell (1929):

> In plants that recover and purify light oil, the acid sludge resulting from the sulfuric acid treatment constitutes a waste disposal problem. Willien (1920) has described the usual method of disposing of the material. It is placed in an acid resisting vessel which is heated with direct steam. The light oil given off during this heating may or may not be recovered by a condenser. The heating causes the solid matter to separate at the top as a spongy, carbonaceous material. The amount of this solid material produced in a medium sized plant is not large, and it may usually be discarded on the dump or burned out in the yard. Because of its sulfur content it is better not to burn it under boilers.

The acid layer forms under the solid matter and is withdrawn. The recovered acid can be slowly fed to the saturators for the recovery of ammonia as ammonium sulfate; however, because the recovered acid is almost black, it should be added slowly. Glowacki (1945) describes the waste and its treatment as follows:

> The acid sludge drained from the agitator during the washing process is an intimate mixture of unused oil, entrained light oil, and reaction products: "resins." In modern practice, the material is taken to some convenient spot and burned. In the past, fairly elaborate acid reclaiming plants have been devised and built; in general the value of the reclaimed material failed to justify the labor, maintenance, and investment costs of the reclamation equipment. A few plants can still be found at American installations.

Either the waste acid sludge, or the carbonaceous material from the reclamation of the acid would be wastes for disposal. The burning of the waste would consist of digging a small trench in the ground at the disposal area, filling the trench with the waste, and then burning the waste in the trench. Only a portion of the waste would actually burn, and the residue would remain in the waste disposal area. Whether the acid was worth recovering from the waste depended primarily on the cost of sulfuric acid. Because the sulfuric acid recovered was a very low grade, its recovery would have been practiced primarily at larger plants.

The waste itself would be very acidic, and the base nitrogen compounds in the light oils from coal-carbonization plants would be extracted into the waste and generally disposed with the waste.

1.4.2.4.3 Tar decanter settlings and saturator sludge--Two types of solid or semisolid, black, and pitchy sludges were produced in the tar decanters and the saturators (used for ammonium sulfate manufacture). The tar-decanter settlings are the solid materials that come from the tar and and flushing liquors. They consist primarily of coal and solid matter carried with the gas into the washboxes. The saturator sludge is a hard pitchy material that forms in the saturators used for the production of ammonium sulfate. "Its exact nature is not known, but it is supposed to be formed by the action of sulfuric acid on the small quantities of tar that are carried by the gas into the saturator" (Powell, 1929). This sludge was probably produced by the acid-catalyzed polymerization of unsaturated hydrocarbons in the saturator. "Fortunately, the quantities of these solid, pitchy wastes are not large. It is usual practice to discard them on the dump or in an excavation" (Powell, 1929). The tar-decanter sludge would be produced by coal-gas, CWG, and oil-gas plants. The saturator sludge would only be produced by coal-gas plants using the semidirect process for the production of ammonium sulfate.

1.4.2.5 Ammonia Recovery Wastes--

Ammonia recovery was practiced only at coal-carbonization plants; when ammonia stills were used to release fixed ammonia salts, ammonia still waste was produced. The recovery of ammonia is described in Section 1.3.5, and the removal of phenolic compounds from the ammonia still wastes is examined in Section 1.3.6. Table 37 shows the composition of the original ammonia liquor

TABLE 37. ANALYSES OF AMMONIACAL LIQUORS AND THE STILL WASTES THEREFROM

ANALYSES OF AMMONIACAL LIQUORS AND THE STILL WASTES THEREFROM				
Type of carbonizing operation	Coke Ovens [a]		Vertical Retorts	
Plant	A	B	A [a]	B [a]
Weak liquor composition, grams per liter				
NH₃, total	6.54	7.06	14.5	13.3
free	3.35	......	9.0	10.0
fixed	3.19	......	5.5	3.3
Sulfide as H₂S	.....	0.138	1.9	1.5
Carbonate as CO₂	.....	0.81		
Cyanide as HCN	.....	......	0.12	0.05
Thiosulfate as H₂S₂O₃	.....	......	0.46	1.78
Thiocyanates as HCNS	.....	......	1.22	2.03
Phenols as C₆H₅OH	.....	......	3.55	3.5
Oxygen absorption (4-hr test), ppm	.....	......	....	16,500
Recovery of fixed ammonia			Limed	Unlimed
Waste composition, grams per liter				
NH₃, total	0.041	0.0034	....	3.1
free	.....	0.0034	....	0.1
fixed	.....	......	....	3.0
Alkalinity as CaO	1.57	1.14	3.6	...
Sulfide as H₂S	.....	0.075	....	Trace
Carbonate as CO₂	.....	0.37	....	...
Thiosulfate as H₂S₂O₃	.....	......	0.21	1.60
Thiocyanate as HCNS	.....	......	0.54	1.83
Phenols as C₆H₅OH	.....	......	1.8	1.7
Oxygen absorption (4-hr test), ppm	.....	......	5,800	10,080

Source: Wilson and Wells, 1945.

and the compositions of the still wastes from two coke-oven plants and two vertical retort plants. The ammonia still waste was first treated for the removal of phenols (by extraction, if the Koppers vapor recirculation process was not used; see Section 1.3.6). "The waste is generally discharged into baffled sumps. Here solid matter settles out and the liquid cools. Accumulations of sediment are removed from the sumps by bypassing them periodically for cleaning" (Wilson and Wells, 1945). "The quantity of lime settlings is not large with good operating conditions, and the material is usually disposed of on the dump" (Powell, 1929).

The wastewater from the separation tanks could be either recycled as scrubber water or discharged into the nearest stream. Its discharge generally created only minor problems if the phenols were removed to adequate levels. Wastewater containing phenols could be run directly to the city sewers or used in coke-quenching operations (as described in Section 1.3.5).

The amount of ammonia still wastes that were produced varied with the ammonia recovery process employed by the plant. Coal-carbonization plants using the direct process of ammonia recovery produced between 20 gallons (Marquard, 1928) and 30 gallons (Powell, 1929) of waste per ton of coal carbonized. The indirect ammonia recovery plants produced about 90 gallons of waste per ton of coal carbonized (Marquard, 1928; Powell, 1929). The semi-direct process for the production of ammonia would produce some intermediate volume of ammonia still wastes.

1.4.2.6 Hydrogen Sulfide Removal Wastes--

   1.4.2.6.1 Spent lime--The disposal of spent lime was a substantial problem for the early gas plants. The spent lime contained a relatively high concentration of CaS, which upon exposure to the atmosphere slowly reacted with water and carbon dioxide to form $CaCO_3$ and H_2S. The spent lime also contained substantial amounts of tar from the gas, and the tar was also very odorous. "The residue from dry lime purification is under certain conditions readily disposed of, being valuable in many cases for agricultural purposes." (Hughes and Richards, 1885). The spent lime for gas plants would have been either disposed near the plant or sold as an agricultural lime. The spent lime, once dispersed, would release hydrogen sulfide and then perform as normal lime in the soil. The tars would be sufficiently dilute to biodegrade,

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and any other constituents in the spent lime would be diluted below noticeable levels. The amount of slaked lime required to purify coal gas was about 1 bushel for every 5,000 to 9,000 ft^3 gas (i.e., 4,020 to 7,230 ft^3 gas/ft^3 slaked lime) (AGLA, 1875).

1.4.2.6.2 Spent oxide--The spent oxide from removal of hydrogen sulfide from town gases is a waste generally found at any previous gas site. The use of iron oxide quickly replaced lime for H_2S removal, and it was the dominant method of hydrogen sulfide removal until the demise of the industry. The use of liquid purification was employed at some of the larger plants after about 1925, but iron oxide was still used at smaller works. The use of iron oxide purification, the types of oxide used, methods of regeneration, and the fillers mixed with the iron oxides are discussed in Section 1.3.7.3.

The composition of spent iron oxide varied substantially among town gas production plants. According to Auebach (1897):

> The gas purifying material...varies in the most extraordinary way, from one works to another; the water varies from 2 to 40, the sulphur from 10 to 55, the sulphocyanides from 0 to 16, the ammonia from 0 to 8, and the Prussian blue from 0 to 15 per cent; the colors vary from yellow to black with all shades of blue, some are dry powders, some are wet masses, and some are half sawdust and chips; and the value varies accordingly.

Water-gas processes produce only small amounts of ammonia and cyanides, so the spent oxide from water-gas production contains only small amounts of sulpho-cyanides, ammonia, and Prussian blue. The spent oxides from coal-carboniza-tion plants would contain substantial amounts of both sulfocyanides and ferri-ferrocyanides.

Spent oxide wastes were universally disposed in the United States, although sulfur and Prussian blue were recovered from spent oxides in the United Kingdom. Spent oxides were usually disposed by using the material as fill, either around the plant, at the local dump, or on private property. Downing (1932) stated that:

> The disposal of spent oxide is a vexatious problem for many gas plants. Because of a possibility of fires starting through heat generated by revivification, it is necessary to hold the spent mate-rial at the plant until this danger is past. As soon as city authorities learn of this menace the material is prohibited at pub-lic dumps. Continuous storage on gas works land eventually becomes

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impossible. The material makes excellent filling for roads or private property when properly handled. It should be covered with ashes or dirt immediately to prevent the access of air and consequent combustion.

Consequently, spent iron oxide wastes are a major waste material remaining on and around the manufacturing sites of manufactured gas. Morgan (1926) described the utilization of spent oxides in the United States:

> In England and on the European Continent, considerable work has been done on the utilization of spent oxide. When cyanogen is not removed from the gas previous to the purifiers, the spent oxide contains considerable ferrocyanide which was formerly recovered in Europe, but which it does not pay to recover in this country. In Europe, also, large quantities of spent oxide are used for the manufacture of sulfuric acid. In one sulfuric acid plant it is claimed that the burnt oxide regenerated by a special process produces a purifying material of good mechanical condition and special activity. At present, however, in the United States there is a plentiful supply of cheap brimstone for the manufacture of sulfuric acid, and the spent oxide has no market value.

The spent iron oxide wastes contain tar, some volatile organics, iron oxide, Fe_2S_3, FeS, FeS_2, sulfur, fluff materials (usually woodchips), ferric ferrocyanide (Prussian blue) $Fe_4[Fe(CN)_6]_3$, and thiocyanates. The cyanide compounds would be almost absent from oxides from CWG, but they would be in oxides from coal gas or mixed coal/water-gas operations. Spent oxide wastes degrade somewhat after disposal. The FeS oxidizes to form sulfuric acid, which helps to rust and dissolve the remaining iron oxides in the waste. Depending on the amount of tar in the waste, the woodchips may or may not be broken down by the acid. The highly acidic conditions do not appear to decompose the ferric-ferrocyanide compounds. When the iron oxides dissolve away, the ferric-ferrocyanide compounds become small unattached particles that can migrate short distances from the waste to stain wood, rocks, soil, and other materials not originally in the spent oxide. This bright blue color is characteristic of cyanide-containing wastes from coal-gas processes, but it is usually absent from water-gas spent oxide waste. A recent analysis of disposed spent oxide wastes in Massachusetts reveals that:

pH	1.7-3.8
Cyanide, total	7,500. ppm
Cyanide, water soluble	0.7 ppm

PAH compounds (mg/kg dry material)

Naphthalene	0.83
Benzo(a)anthracene	1.70
Benzo(a)pyrene	2.10
3,4-Benzofluoranthene	5.10
Benzo(k)fluoranthene	2.20
Chrysene	1.60
Benzo(ghi)perylene	2.00
Dibenzo(a,h)anthracene	0.75
Ideno(1,2,3-cd)pyrene	2.00
Pyrene	2.80
Fluoranthene	6.10
bis(2-ethylhexyl)phthalate	1.70

The high concentration of bound cyanides (and its blue color) identifies the spent oxides as being produced as a waste from coal carbonization. Table 38 is an analysis of a spent iron oxide, listing the chemicals identified in the spent oxide. This analysis should not be thought of as typical for a spent oxide waste because of the very high variation of spent oxides from plant to plant. This oxide was obviously not mixed with woodchips or other fluff, and consequently it has a low organic matter content. The presence of ferrocyanide compounds indicates that the oxide was used to purify coal gas, and the low tar content and high sulfur content of the oxide indicate a very efficient tar-removal system was in place prior to the oxide purifiers. A more typical spent oxide would have a larger tar content, a substantial amount of organic matter (from woodchips), and a smaller amount of free sulfur. Table 39 shows the average composition of spent oxides from eight water-gas plants that operated in Illinois and Indiana in 1921. The tar content of these oxides ranged between 0.6 and 19.0 percent of the dry spent oxides. The conversion of the oxides used with CWG was much lower than was the predicted use of oxides reported in the literature of the time. The predicted conversions of spent oxide have sulfur concentrations of 50 to 60 percent.

The amount of oxide used by an individual plant to remove hydrogen sulfide was proportional to both the amount of gas produced by the plant and to the hydrogen sulfide content of the gas purified. Table 40 shows the

TABLE 38. AN ANALYSIS OF SPENT OXIDE

An Analysis of Spent Oxide [47]	
	Percent
Free sulfur	44.70
Moisture	17.88
Ferric monohydrate	5.26
Ferrous monohydrate	6.25
Basic ferric sulfate	1.25
Ferric ammonium ferrocyanide	3.80
Ferrosoferric ammonium ferrocyanide	2.50
Ferric pyridic ferrocyanide	1.20
Organic matter peat fiber	4.68
Tar	1.21
Silica	1.05
Naphthalene	0.72
Pyridine sulfate	0.77
Ammonium sulfate	2.06
Calcium sulfate	0.12
Ferrous sulfate	0.02
Ammonium thiocyanate	1.30
Sulfur otherwise combined	1.33
Organic matter soluble in alkalies (humus)	1.54
Combined water and loss (by difference)	2.36
	100.00

Source: hill, 1945.

TABLE 39. SPENT OXIDE COMPOSITIONS FROM 16 GAS PLANTS IN ILLINOIS AND INDIANA

	8 carbureted water-gas plants	8 mixed (coal and carbureted water-gas) plants
Density of spent oxide (lb/ft^3)	33.0	52.3
Sulfur in spent oxide (% dry basis)	21.7	37.4
Tar in spent oxide (% dry basis)	6.9	3.6
Total gas purified (10^3 ft^3 gas/ft^3 oxide)	39.6	73.1
Average inlet H$_2$S concentration (%)	0.21	0.36
Range of average daily production (10^6 ft^3)	0.17-3.20	0.20-1.50
Oxide types		
Rusted borings	5	8
Natural (bog ore)	3	0

SOURCE: Dunkley, 1921.

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TABLE 40. ESTIMATED GENERATION OF SPENT OXIDE WASTES FROM GAS PRODUCTION

	H_2S concentration in gases[a] (%)	Spent oxide generated per 10^6 ft^3 gas[b] (tons)	(ft^3)
Coal gas	0.30-0.75	0.31-0.77	15-39
Carbureted water gas	0.075-0.22	0.007-0.22	3.9-11
Pacific Coast oil gas	0.30-0.38	0.31-0.39	15-20

[a]From F.W. Speer (1921) as reported by Morgan, 1923.
[b]Assumes: (1) Original bulk density of iron oxide with woodchips = 20 lb/ft^3.
(2) All of H_2S removed.
(3) Final spent oxide is 40% sulfur by weight, with bulk density = 40 lb/ft^3.

approximate weight and volume of oxides produced from the three major gas production processes. The assumptions and locations of the data used to generate this information are on the table. This table indicates that the production of CWG generally produced less iron oxide waste than did either oil gas or coal-carbonization gas. The estimates in Table 40 are useful for rough estimates of the amount of spent iron oxide-generated gas production.

1.4.2.6.3 Liquid scrubbing wastes--The solutions used for the liquid scrubbing of hydrogen sulfide from town gases could not be used indefinitely. The solutions generally became deactivated by side reactions that produced inert salts. The products of these side reactions had to be removed and either recovered or discarded. The four significant liquid purification processes (Seaboard, Nickel, Thylox, and Ferrox processes) are described in Section 1.3.7.4.

The Seaboard process uses a solution of 3 to 3.5 percent Na_2CO_3 to absorb hydrogen sulfide from manufactured gas. The solution is regenerated by blowing air through the H_2S rich solution, releasing the hydrogen sulfide to the air. The use of air to strip the H_2S from the gas also oxidizes some of the absorbed H_2S to sodium thiosulfate ($Na_2S_2O_5$). This occurs with about 5 percent of the H_2S, which is absorbed by the process. The cyanide in the gas is also absorbed and oxidized to form sodium thiocyanate (NaSCN). This process required between 20 and 60 pounds of sodium carbonate for every 10^6 ft^3 of gas purified. The sodium thiocyantes were sometimes recovered from the scrubbing liquia, but they were usually disposed rather than recovered. The thiocyanates would be formed when the process was applied to coal-gas production, but they would have been formed only in small amounts when the process was applied to oil gas or CWG (because of the small amount of cyanide in these gases).

The Thylox, Ferrox, and Nickel processes each used solutions of sodium carbonate for the removal of hydrogen sulfide. The metals added to the solutions (arsenic, iron, and nickel) served as catalysts in the regeneration of the solutions. In the presence of the catalysts, the H_2S is oxidized to f·e sulfur and water. Cyanides were removed and oxidized to sodium thiocyanate by both the Ferrox and Thylox processes. In the Nickel process, the cyanide reacted with the nickel catalyst, deactivating it. This process was not used

for the removal of H_2S from coal-carbonization gases because of the cyanide present in the gas. It was used with either oil gas or CWG.

The Ferrox process used a solution of 3.0 percent sodium carbonate and 0.5 percent ferric hydroxide. The sulfur prcduced by the process entrapped both the ferric hydroxide and sodium carbonate in the product sulfur [the product sulfur had 20 to 40 percent (dry basis) total impurities attributable to these compounds]. This reduced the marketablity of the product sulfur and also required relatively large amounts of makeup sodium carbonate (about 350 lb 10^6 ft^3 gas treated) and ferric hydroxide (about 280 lb/10^6 ft^3 gas treated) (Kohl and Riesenfeld, 1985).

The Thylox process used a solution of sodium carbonate and arsenic trioxide to absorb hydrogen sulfide from the gas and recover it as sulfur crystals. The sulfur produced was of a high grade and was usually marketed for agricultural purposes. Sodium carbonate consumption from the process was 60 to 120 lb/10^6 ft^3 treated, and arsenic trioxide consumption was 15 to 27 lb/10^6 ft^3 gas treated (Gas Engineers Handbook, 1934). The process required that a small portion of the recycling solution be continously withdrawn from the system to prevent the accumulation of sodium thiosulfate and thicyanate salts. The arsenic in this purge stream could be recovered by acidifying the solution and recovering the arsenic as arsenic sulfide crystals. The recovered arsenic could then be returned to the scrubber with additional sodium carbonate solution. Because the recovery and recycling of the arsenic was an economic decision, some plants may have disposed of the purge stream rather than attempt recovery of the arsen.. "If feasible, the solution removed can merely be discarded, or, if necessary, it can be acidified and filtered to remove its arsenic as arsenic sulfide before being discarded" (Gollmar, 1945). Some of the arsenic also remained in the recovered sulfur product, but at levels too low to cause problems when the sulfur was used for agricultural purposes.

The Nickel process used a colloidal solution of nickel sulfide and sodium carbonate to scrub hydrogen sulfide from gas and recover the sulfur. Like the Thylox and Seaboard processes, sodium thiosulfate and sodium thiocyanide accumulate in the solution. The consumption of the nickel sulfide was 23 lb 10^6 ft^3 gas oil or CWG treated, and sodium carbonate consumption was 51 to 120 lb 10^6 ft^3 gas treated (Cundall, 1927).

The disposal of waste scrubber solutions was generally performed by discharging the liquid wherever it was practical. No references were found as to the disposal practices in articles that reviewed the operations of the processes, but a survey of gas manufacturers did report disposal practices for scrubber liquids. This survey (in 1930) sent questionnaires to 100 large gas companies with production of greater than 500×10^6 ft^3/year. Of the 57 companies that responded, 12 used some type of liquid purification, 5 indicated that they discharged their waste liqours to ponds, sand flats, or cinder fills and the remaining 7 said they discharged to either city sewers or to river tidewater (Wardale, 1930). This survey indicates that some plants using liquid purification could have substantial contamination from arsenic or nickel if they disposed these scrubber solutions onsite.

1.4.2.7 Lampblack Wastes--

The production of gas by the Pacific Coast oil-gas process was accompanied by the generation of large amounts of lampblack. The feedstocks for the production of oil gas were asphaltic-based oils and had high carbon-to-hydrogen ratios. When these oils were thermally cracked for the production of oil gas, much of the original carbon in the oils formed elemental carbon. This carbon (lampblack) usually washed out in the washboxes, where the heaviest tars also condensed. The material recovered in the washbox was a sludge with large amounts of free carbon, some heavy tars, and water from the washbox.

Morgan (1926) states that:

From 12 to 24 pounds of lampblack are formed per 1000 cubic feet of gas made, and practically all of this is thrown out in the wash-box. The water from the wash-box containing this lampblack in suspension passes off through large overflow pipes. In smaller plants this water suspension of lampblack flows into small settling pits, from which after settling the clear water is drawn off. The lampblack is then mixed with tar and used for boiler fuel. In larger plants the lampblack in the overflow may be separated from the water by an Oliver continuous rotary filter. It may then be briqueted with a small amount of tar and sold as a superior boiler fuel.

The briqueted lampblack was sometimes used as generator fuel for the production of CWG. Although the lampblack had value as a fuel, many small plants would dispose of the lampblack rather than recover it, and large plants might

produce so much lampblack that they disposed of the material they could not use. The organic tars removed from the washbox with the lampblack would be relatively heavy tars, with the composition dependent on the temperature of the washbox.

1.4.2.8 Ash, Clinker, and Coke--

Ash, slag, and coke were wastes produced in the production of town gases. The ash was produced from boilers, CWG generators, and producer gas generators. The coke or coal placed into producer gas generators or CWG generators could not be combusted completely to ash. The requirement that air and steam be able to flow through the coke beds meant that the ash had to be removed with a substantial amount of unburned coke remaining in the ash. The ash was then usually run through a coarse grate to remove any large pieces of coke (which were recycled to the generators), and the material falling through the grate was discarded. This both recovered usable coke and decreased the carbon content of the ash, making the ash more suitable as a fill material. The ash produced by CWG and producer gas had substantial amounts of unburned coke. Ash from the boilers, however, was combusted much more completely. Within the generator bed, some large agglomerations of ash would form. These were called "clinkers" and were removed from the generators at regular intervals. "Water gas generator clinker, and boiler house and producer gas ash are normally disposed of by using [them] for fill and grading purposes" (Powell, 1929). Although ash was apparently used in Europe for the manufacture of brick or cement, this was not done in the United States because of the relative cheapness of other raw materials.

The amount of ash produced by gas manufacturers was directly proportional to the ash content of the coals and coke used for gas production. The ash produced at oil-gas plants would be a petroleum ash and would have a different composition than the coal ash.

1.4.2.9 Firebrick and Building Materials--

The apparatus for the production of oil gas and CWG was lined with firebricks that were alternately heated and cooled during the manufacture of gas. Coal-carbonization apparatus used firebricks for linings and heat exchangers. The apparatus periodicall. had to be relined with new firebricks because of the wear associated with gas manufacture. Broken firebricks were used as fill

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material wherever needed around the plant or they were added to the dump. Buildings were sometimes removed during plant operations, and the final clearing of the site occurred after manufactured gas was replaced by natural gas. These building materials were also used to fill areas on or near the site.

1.4.3 Specific Articles on Waste Disposal

During the literature review, several articles that specifically addressed the waste disposal practices of the industry were discovered. These articles, which take a fairly wide view of waste disposal practices, are described in this section.

Shelton (1897) reviews "The Nuisance Question in Gas Works." He describes the sources of odor, noise, smoke, and offensive drainage from gasworks. Methods of reducing the problems created by operating a gas plant are described, as are methods of improving the plant appearance. Shelton states that:

> Offensive refuse drainage may come from: 1) unintercepted scrubber water or condenser water saturated or laden with ammonia, tar or oily scum; 2) tar or oil wasted; 3) the rain washings of spent lime or old oxide; 4) general gas works and surface drainage; 5) drip water not properly disposed of.

Hansen (1916) describes the "Disposal of Gas House Wastes" in which he describes the objectionable effects of gas house waste disposal and describes methods for preventing these effects. Because Hansen's work was presented to a group of gas plant operators, it was not especially well-received according to the reviewer comments. Hansen states that:

> Wastes vary greatly in quantity and character due to variable recovery of useful constituents and to the use of variable quantities of water. Generally speaking, the quantity of wastes per million cubic feet of gas manufactured is greater and more offensive in the smaller plants than in the larger ones because of the smaller recovery of marketable products and greater waste due to leaky tanks and defective apparatus.

He lists several cases of stream and water pollution attributable to gas plant wastes and how the disposed wastes give fish gassy odors and impart medicinal tastes to water. At Centralia, Illinois, according to Hansen:

> Much complaint was made of tarry wastes adhering to the legs of cattle, and to injury of soil and crops by tarry deposits...Another

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bad effect of gas house wastes which has here and there given rise
to more or less se ious trouble is the pollution of the soil, which
in turn gives rise to gassy tastes in well waters and to gassy odors
in cellars. A striking example of this occurred at Joliet, where
one of the public water supply wells was affected with a gassy taste
which could be explained on no other basis than contamination from a
gas plant near by...At the town of Carthage, in southern Ohio...pol-
lution was occasioned by coal tar wastes used at a tar paper fac-
tory. These wastes were permitted to flow into a pit at least 2,000
feet from the affected wells.

An estimate that 1,600,000 gallons of tar and oily wastes exist underground at
Lowell, Massachusetts, is presented. When some contaminated areas were tapped
by excavations. the wastes "flowed out in springs." Methods of removing oils
and tars from aqueous wastes by coagulation with ferrous sulfate and lime are
described, as is the use of sand and coke filters.

Brown (1919) describes how the chlorination of water containing trace
amounts of gas plant waste produces objectionable tastes in the water. The
levels of organic material themselves did not produce objectionable tastes,
but the tastes became noticeable after chlorination.

The American Gas Association (AGA) had a standing committee on waste
disposal from gas plants during the 1920's. Their articles (as reported in
the annual proceedings of the AGA) detail the wastes produced and the normal
methods of waste disposal.

Willien (1920) documented the injurious effects attributed to the waste
from gas plants and described the types of waste produced from coal-gas and
CWG plants. The effects of gas plant waste included driving away fish and
contaminating oyster beds, damaging paint on pleasure boats, objectionable
odors, pollution of wells, deposits in sewer systems, and pollution of drink-
ing water. According to Willien, "Pollution of wells...is caused by the seep-
age of gas plant waste through the ground and contaminating the ground water.
This may result from a crack in a tar well or holder pit through which tar
leaks, or from leaky tar, oil, and ammonia pipes."

Sperr (1921) describes methods of tar separation that can be applied to
aqueous gas plant wastes. Typical systems for the gravity separation of tar
from water are described, as is the use of centrifuges for the dehydration of
tar emulsions.

Willien (1923) describes the formation, treatment, and storage of tar
emulsions and tars.

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Powell (1929) classifies and describes the wastes produced by gas manufacturing. Dividing the wastes into two classes (solid and liquid wastes), he describes the wastes and the usual methods of disposal. "It must be realized," states Powell, "that gas plant wastes are really by-products whose value is too low to make direct sale feasible." Table 41 lists the wastes as listed by Powell.

The only survey of waste disposal methods was published by Wardale in 1930. A survey was sent to 100 gas companies in the United States with gas production greater than 500×10^6 ft^3/year. Answers were received from 57 companies, 10 of which were no longer producing gas (they had converted to natural gas). Table 42 summarizes the questions and answers most related to waste disposal. Although this survey was not comprehensive of the entire industry (smaller gas plants were not even contacted), it is the only reported survey of gas plant disposal practices.

One possible method of waste disposal that was originally thought to be commonly used by plants was the disposal of waste by injecting it into wells. Only two references to the use of wells for the disposal of wastes were uncovered during this investigation. The first is an article listed in a bibliography on plant waste disposal. The bibliography was published in the 1955 AGA proceedings, and the referenced article was titled "Underground Disposal of Process Waste Water," by L.K. Cecil (1950). A summary of Cecil's article states: "Underground disposal of brines and chemical wastes water. Acidizing the injection well semi-annually maintains disposal capacity. Cooling tower blowdown containing chromates is similarly handled." The second reference is by Wilson and Wells (1945), who state that:

> Disposal of ammoniacal liquors or waste by discharge into the ground is seldom possible except in very small carbonizing operations. Discharge into an opening, such as a disused well, is dangerous, because the ultimate fate of the liquor is unknown. It may be gradually dissipated and purified as it seeps through the soil. On the other hand, it may find its way into some water bearing strata or percolate unchanged through the layers of soil to drain into a stream. In such a case the pollution would not appear immediately, but when it did, deposits of the material in the contaminated soil would cause the trouble to persist over a long period of time.

TABLE 41. GAS PLANT WASTES

I.   Solid wastes

     1. Ash and clinker
     2. Spent oxide
     3. Tar decanter settlings and saturator sludge
     4. Lime settlings

II. Liquid wastes

     1. Phenol-bearing wastes
         a. Ammonia still waste
         b. Other phenol-bearing wastes

     2. Wastewater not containing phenol
         a. Coke quenching water
         b. Producer gas cooler water
         c. Water-gas tar separator overflow

     3. Acid sludge from light oil agitators

     4. Tar emulsions

SOURCE: Powell, 1929.

TABLE 42. RESPONSES TO WASTE DISPOSAL SURVEY

Do you produce ammonia? What disposal is made of still wastes?

    22 - Ammonia plants
     5 - Settling basins or coke filters
     1 - Phenol removal equipment
    13 - Discharge untreated weak liquor or still waste
     1 - Sells weak ammonia liquor

What is done with spent oxide from purifiers?

    24 - Use as fill onsite or given away as fill
     1 - Sold for sulfuric acid manufacture
     1 - Dumps it at sea
    13 - Haul it to city dump
     1 - Dumps it into river at flood level
     1 - Gives it to stable for horse bedding
Several - Mention need to cover or mix spent oxide with dirt

Do you use liquid purification? How are waste liquors disposed of?

    12 - Use liquid purification
     5 - Discharge to ponds, sand flats, cinder fills
     7 - Discharge to city sewers or river tidewater

What disposition is made of wastes containing oil?

     3 - Pump into relief holder
     8 - Use baffle separators and coke filters
    15 - Use separators or settling basins, remove oil by skimming, burning
          it in boiler, or mixing it with tar
     4 - Run wastes to sewers or creeks without treatment

What other wastes do you dispose of besides waters from scrubbers, washboxes,
purifiers, and sanitary and surface water drains?

    10 - Ammonia still waste or weak ammonia solution
     1 - Shavings from tar scrubber, which are burned after dark
     2 - Coke quench water
     1 - Water-softening residue

What methods of treatment before discharge to sewers?

    18 - Baffled separators
    13 - Baffled separators and coke or cinder filters
     1 - Oliver-Borden filters
     1 - Ferrous sulfate and soda ash treatment before coke filters
     1 - Recirculates water to washboxes
     6 - Discharge without treatment

SOURCE: Wardale, 1930.

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1.5 PRODUCTION AND HISTORICAL TRENDS OF THE U.S. TOWN GAS INDUSTRY

1.5.1 Introduction

This section examines the historical trends of the U.S. town gas industry. By studying the production trends for various parts of the country, the predominant methods of gas production can be identified, the amount of gas produced can be examined, and the approximate time that the manufacture was replaced by natural gas use can be determined. The gas production processes, feedstocks, and innovations in the industry affected both the quantities and disposal practices for wastes. By tracing the changes that occurred in the industry, additional insight to the problems of current gas sites can be acquired.

Most of the statistical data on gas production, employment, and feedstock use were collected during the operation of the manufactured-gas industry by AGA. RTI's effort to collect and examine this data is probably the first time the industry has been examined since the late 1950's.

Section 1.5.2 reviews the historical production trends within the U.S. and individual regions. Section 1.5.3 shows how the feedstocks for gas manufacture changed with time. Section 1.5.4 plots the historically significant events of the industry.

1.5.2 U.S. Gas Production Trends

The production trends of the U.S. manufactured-gas industry show the amounts of gas produced, the types of gas manufactured, and when the manufacture of gas stopped. The types of wastes from gas production varied with the manufacturing processes (coal gas, CWG, and oil gas), and the amounts of waste produced are approximately proportional to the the amount of gas manufactured. The gas production within a region can be used to estimate (in a qualitative manner) the waste types that would be found at former gas-manufacturing sites.

The gas production trends can be studied for either the entire country or for separate regions. Examining the entire United States allows overall trends to be studied, whereas regional trends are more relevant for applications to local trends. Statistical data were compiled from the information collected and reported annually by the AGA. The original data were collected on a State-by-State basis, with regional totals. The regions used by the AGA,

and the States within each region, are listed in Table 43. Attempting to compile and analyze the statistics on a State-by-State basis is feasible, but it was not performed on this project because of the substantial effort required. Most of the earlier data on gas production were reported on an Mcf basis (10^6 ft^3), and data after 1945 were in millions of therms (1 therm = 100,000 Btu). Table 44 shows the gas heat values and conversion factors used for each type of gas.

Figure 40 shows the total U.S. manufactured-gas sales between 1821 and 1956. This figure includes manufactured gas that was mixed with natural gas and distributed as a mixed gas product. This figure indicates that U.S. gas production was relatively small before 1900, increased rapidly to 400 billion cubic feet (bcf) between 1900 and the beginning of the Great Depression (1929), then fell about 25 percent during the Depression but recovered during World War II. The production of gas peaked shortly after World War II, before declining about 50 percent between 1947 and 1956. The apparent drop in gas production in 1920 did not actually occur. The data prior to 1920 came from a source (Fulweiler, 1921) different from the information between 1920 and 1956 (AGA, 1961).

Figure 41 shows how the manufactured gas was produced between 1919 and 1956. This figure does not include gas manufactured for mixing with natural gas, and the production of retort gas was included with coke-oven gas prior to 1928. This plot shows several interesting trends. There was a steady rise in purchases of coke-oven gas between 1920 and 1930, reflecting increased production of metallurgical coke by byproduct coke ovens during the period. There was a steady decline in retort gas production by gas companies during the period, displaying a tendency of smaller coal-carbonization plants (using retorts) to switch to other forms of manufactured gas as existing retorts wore out. The large drop in oil-gas production in 1928 occurred because much of California switched to natural gas that year. The production of coke-oven gas, oil-gas, and coke-oven gas purchases remained relatively constant between 1930 and 1950, and CWG production showed a substantial decline and increase during the same period. This shows that CWG production was more sensitive to gas demand than was coal-gas production. In relative amounts of gas produced, this figure indicates that the production of CWG was approximately equal to

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TABLE 43. STATES LOCATED WITHIN EACH GAS PRODUCTION REGION

New England States
  Connecticut
  Maine
  Massachusetts
  New Hampshire
  Rhode Island
  Vermont

Middle Atlantic States
  New Jersey
  New York
  Pennsylvania

East North Central States
  Illinois
  Indiana
  Michigan
  Ohio
  Wisconsin

West North Central States
  Iowa
  Kansas
  Minnesota
  Missouri
  Nebraska
  North Dakota
  South Dakota

South Atlantic States
  Delaware
  District of Columbia
  Florida
  Georgia
  Maryland
  North Carolina
  South Carolina
  Virginia
  West Virginia

East South Central States
  Alabama
  Kentucky
  Mississippi
  Tennessee

West South Central States
  Arkansas
  Louisiana
  Oklahoma
  Texas

Mountain States
  Arizona
  Colorado
  Idaho
  Montana
  Nevada
  New Mexico
  Utah
  Wyoming

Pacific Coast States
  California
  Oregon
  Washington

TABLE 44.   GAS HEAT VALUES USED TO CONVERT BETWEEN FT^3 AND THERMS[a]

Gas type	Btu/ft^3	10^6 therm/10^9 ft^3
Coke-oven gas	540	5.4
Retort gas	520	5.2
Carbureted water gas	600	6.0
Oil gas	600	6.0

[a]1 therm = 100,000 Btu.
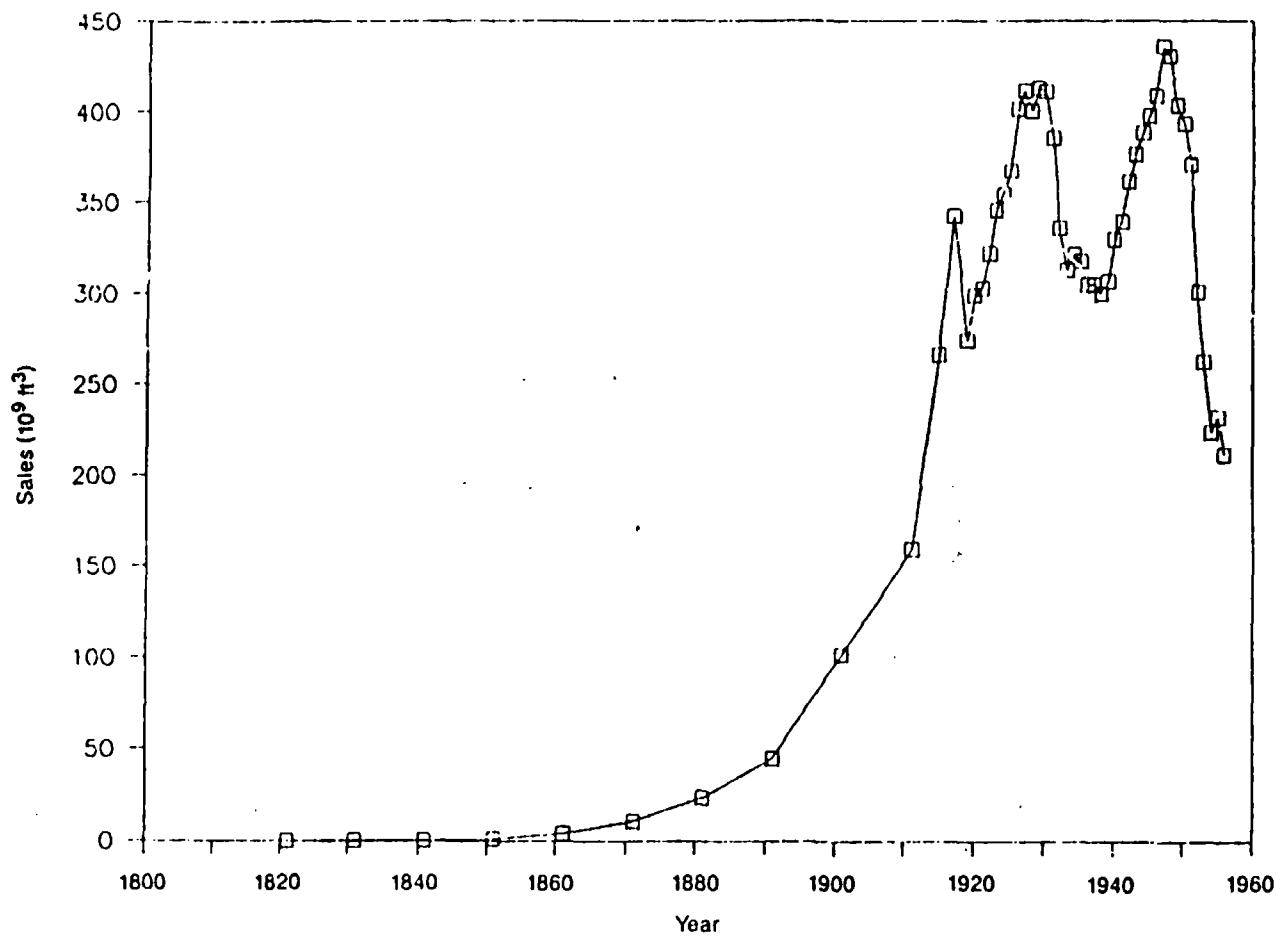
Figure 40. Total U.S. manufactured-gas sales, 1821 to 1956.
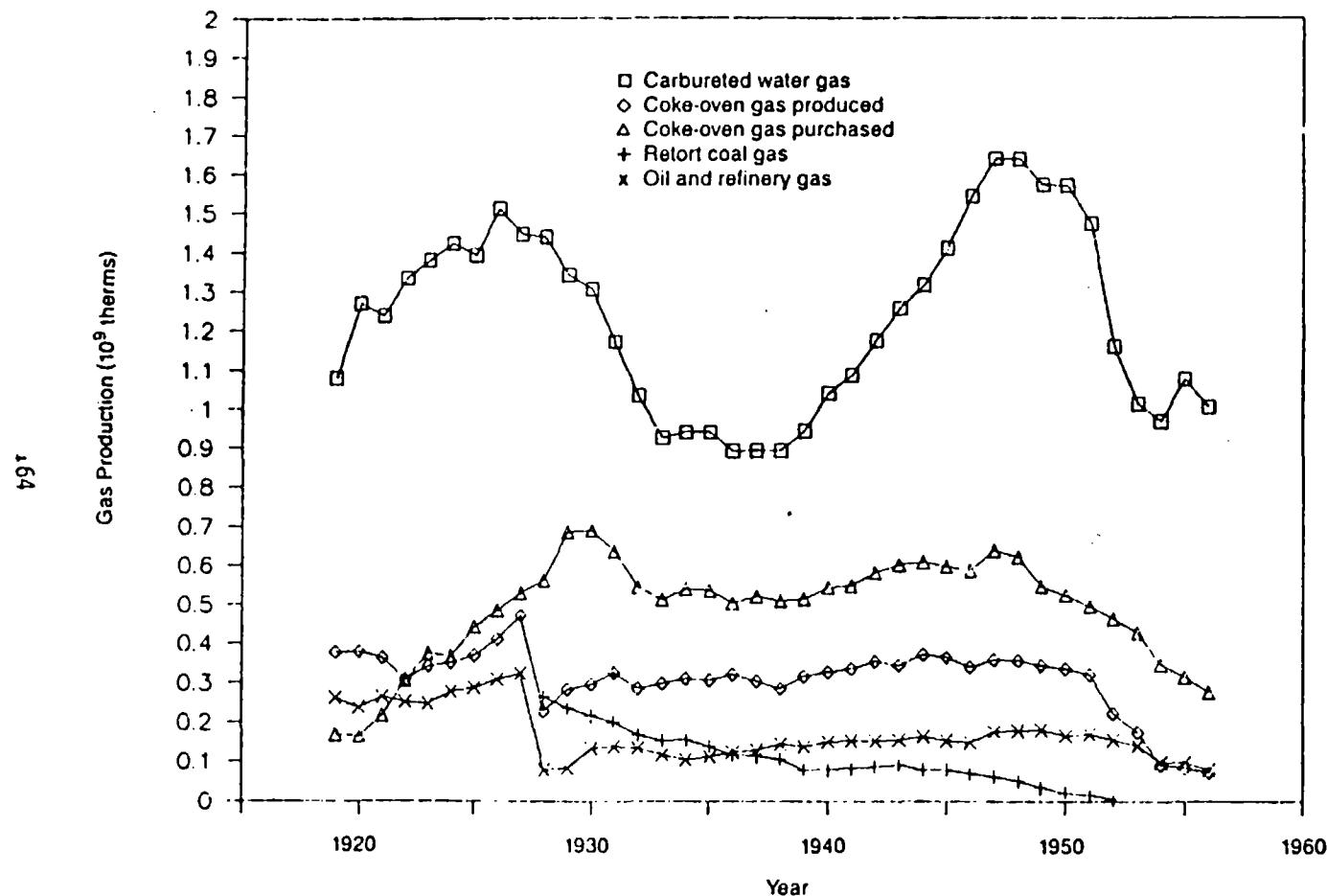
Figure 41. Total U.S. manufactured-gas production, by type, 1919 to 1956.

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the production and purchases of coal gas. The production of CWG was much more
suited to smaller plants, so the relative number of plants producing the two
gases was greatly different. In 1920, the AGA reported the following distri-
bution of gas-manufacturing plants (as compiled from Brown's Directory):

Coal-gas plants	189
CWG plants	429
Combined gas plants	
Coal and CWG	150
CWG and oil	3
Coke oven and CWG	9
Coal, CWG, and coke oven	5
Coal and oil	3
Coal, CWG, and purchased.	5
CWG and purchased	12
CWG and natural	4
Oil and natural	3
Reformed natu il gas	4
Type not listed	1
Purchased, no mf .	99
Total manufactured-gas plants	987
Byproduct coke ovens	82

This distribution shows that 43 percent of the U.S. gas plants in 1920
produced exclusively CWG and 62 percent of the plants produced at least some
CWG. The 82 byproduct coke ovens sold gas to companies for distribution.

Figure 41 also snows a decrease in all types of gas production, beginning
about 1950. The decrease in the coke-oven gas produced in 1928 is an artifact
of the way the data were collected. Retort coal gas was included with the
produced coke-oven gas prior to 1928, but it was collected separately after
1928.

The U.S. manufactured-gas production for each region is shown in Figures
42a and 42b. These figures do not include gas that was manufactured and mixed
with natural gas. Hence, whenever a company acquired natural gas, but still
produced gas for peak loads, its production was excluded from the data. The
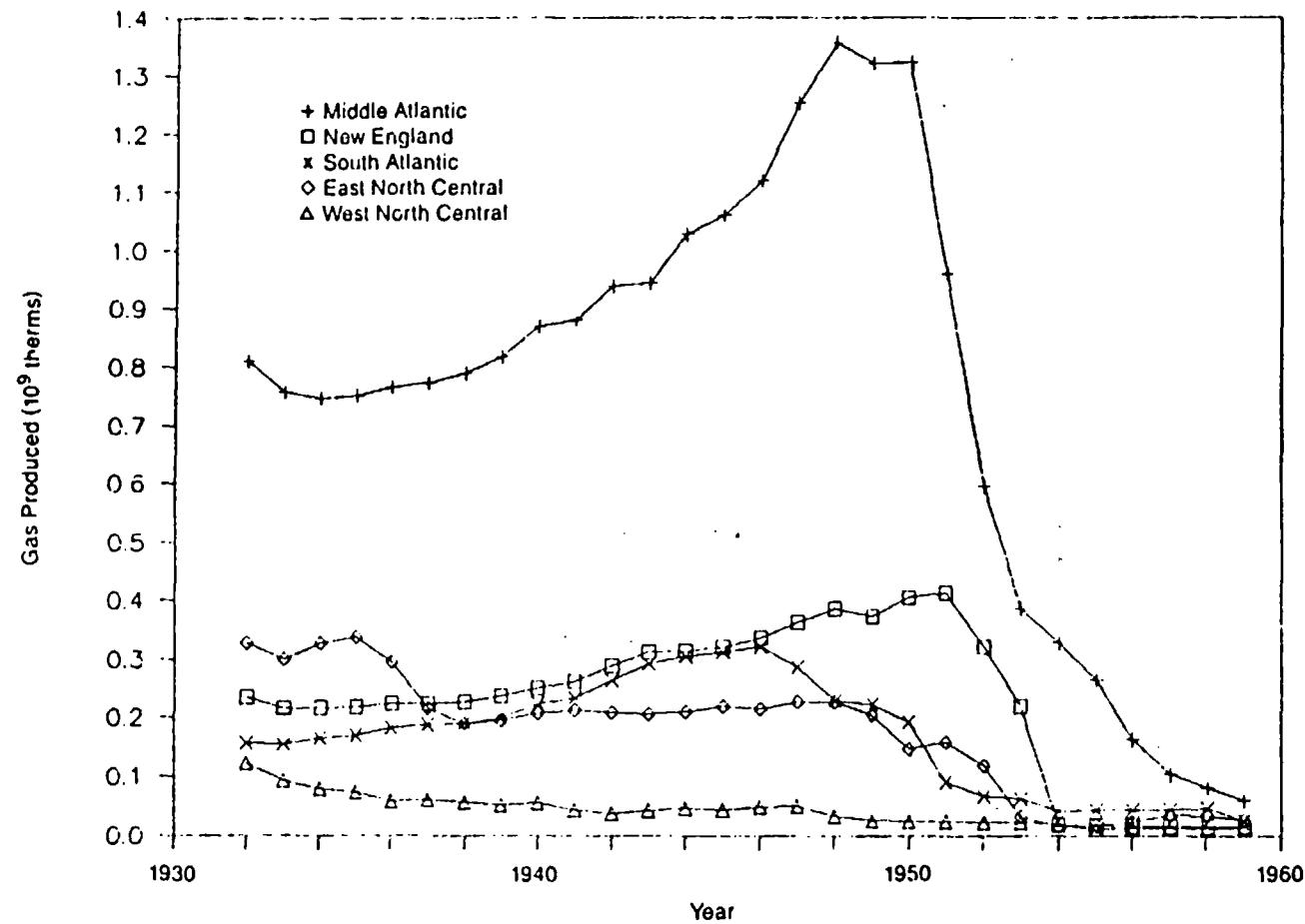
Figure 42a. U.S. manufactured-gas sales, by region, 1932 to 1959.
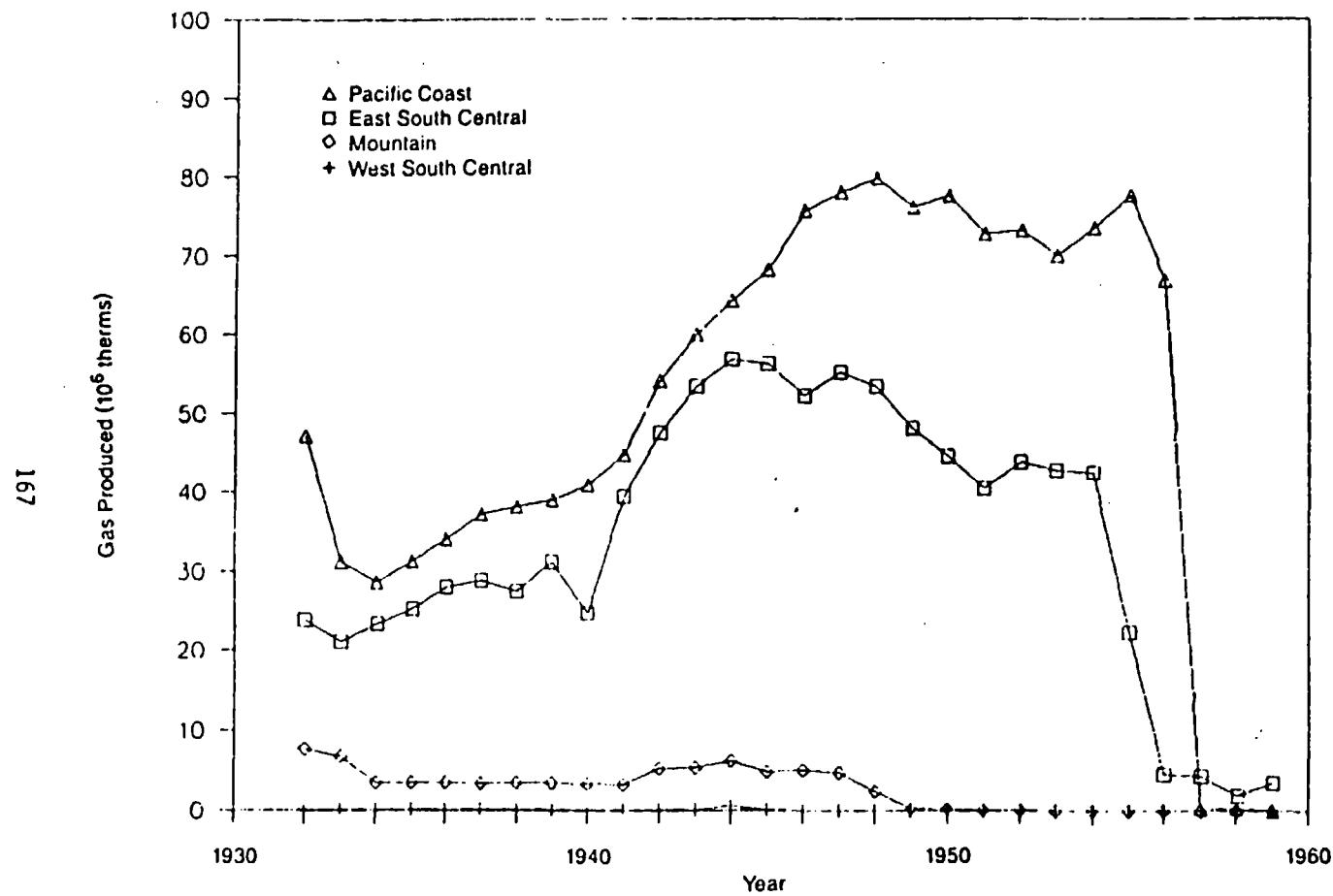
Figure   42b. U.S. manufactured-gas sales, by region, 1932 to 1959.

data do include coke-oven gas purchased for resale by gas companies selling
manufactured gas. These figures show the relative amount of manufactured gas
produced by various regions in the United States. The Middle Atlantic region,
composed of New York, New Jersey, and Pennsylvania, produced about twice the
amount of gas of any other region during this period. The New England, South
Atlantic, and East North Central regions produced comparable amounts of gas
during this period, and each of the other regions produced gas at levels
smaller than 1/10 the production of the Middle Atlantic states. There is a
large change in scale between Figures 42a and 42b, which allows the gas
production in the smaller gas production regions to be examined. The West
South Central region produced no gas during this period because of the avail-
ability of natural gas in the region. The figures indicate the introduction
of natural gas to the regions by the resulting drops in records of manu-
factured-gas sales. The start of the production declines for the regions is
listed below:

Region	Year
West South Central	?
Mountain	1948
South Atlantic	1951
Middle Alantic	1951
East North Central	1952
New England	1952
East South Central	1955
Pacific	1956

The employment trends of the gas industry tracked the production trends.
Figure 43 shows employment in the U.S. gas industry, divided by the type of
gas sold by companies. It shows a dip in all employment during the Great
Depression, with increases in employment during World War II and until 1950.
Between 1950 and 1955, employment in companies selling manufactured gas drop-
ped sharply, and employment in companies selling mixed gas increased during
the period, prior to decreasing after 1955. This indicates that manufactured-
gas companies switched to distributing mixed gases after natural gas pipelines
were installed in their regions. The employment in companies producing or
distributing natural gas increased steadily after World War II.
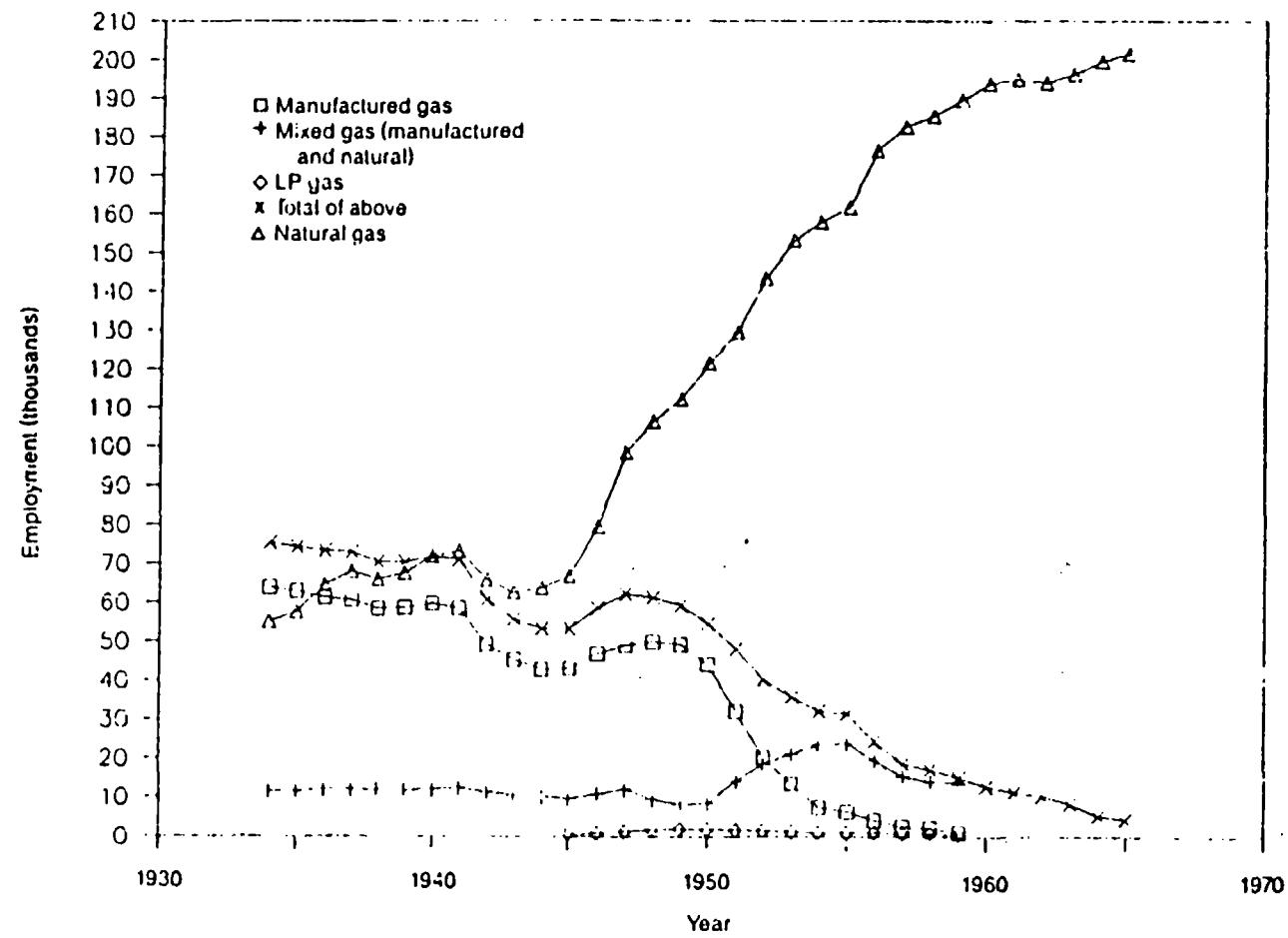
Figure 43. U.S. gas industry, by gas type, 1934 to 1965.

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Figures 44 through 52 show regional gas production by gas type for the nine U.S. regions between 1928 and 1956. They are ordered by the total amount of gas produced within each region, so that Figure 44 is for the Middle Atlantic (with the largest gas production) and Figure 52 is for the West South Central States (which had very little gas production). These figures include gas that was manufactured and mixed with natural gas for distribution. Specific features of the figures are described below:

- Figure 44: Middle Atlantic States--CWG was the major gas type manufactured in this region. The rate of CWG production doubled between 1935 and 1952, and production of other gas types remained relatively constant during the period. Relatively little retort coal gas and oil gas was produced, and the production of coke-oven gas was equally divided between that produced by gas companies and that purchased from coke companies. Natural gas became available in the area after 1951, resulting in the decline of coke-oven gas produced by gas companies and CWG production. The gas companies continued to purchase coke-oven gas during this period. A comparison of this figure and Figure 42a shows that the CWG was mixed with natural gas for distribution by companies in this region (gas manufactured and mixed with natural gas is not shown in Figure 42a).

- Figure 45: New England States--CWG and coke-oven gas were the major production processes. CWG production increased during and after World War II, and coke-oven gas production and purchases remained relatively constant. Natural gas was introduced to the region in 1952, resulting in declines in all gas-manufacturing production. Oil-gas production increased between 1945 and 1952 and fell to zero later. This indicates that gas utilities in the region either converted CWG apparatus to oil gas or installed oil-gas apparatus. High Btu oil gas had a heating value close to natural gas and was used to supplement natural gas for peak loads.

- Figure 46: South Atlantic States--CWG was the major gas produced in this region. Some coke-oven gas was purchased, and a small amount of retort gas and oil gas was produced, but the total of the gas from these sources was less than half of the CWG production. Gas production dropped steadily after 1945, but some increase in oil-gas production is observed. The oil-gas production would principally be from converted CWG apparatus and used for gas production during peak loads.

- Figure 47: East North Central States--Purchases of coke-oven gas exceeded the other types of gas production between 1929 and 1948. The production of coal gas (both produced and purchased)
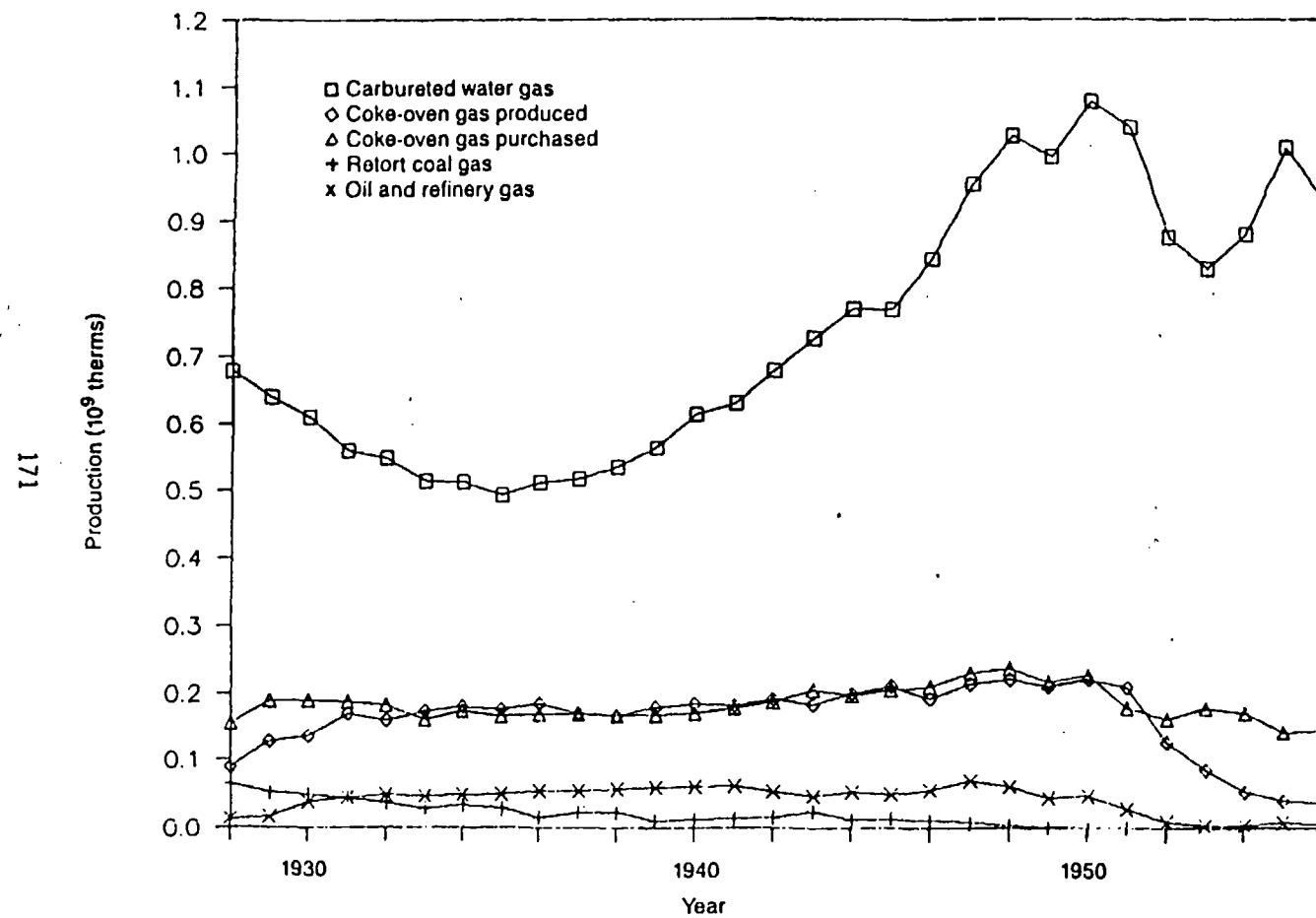
Figure 44. Gas production in the Middle Atlantic States, 1928 to 1956.

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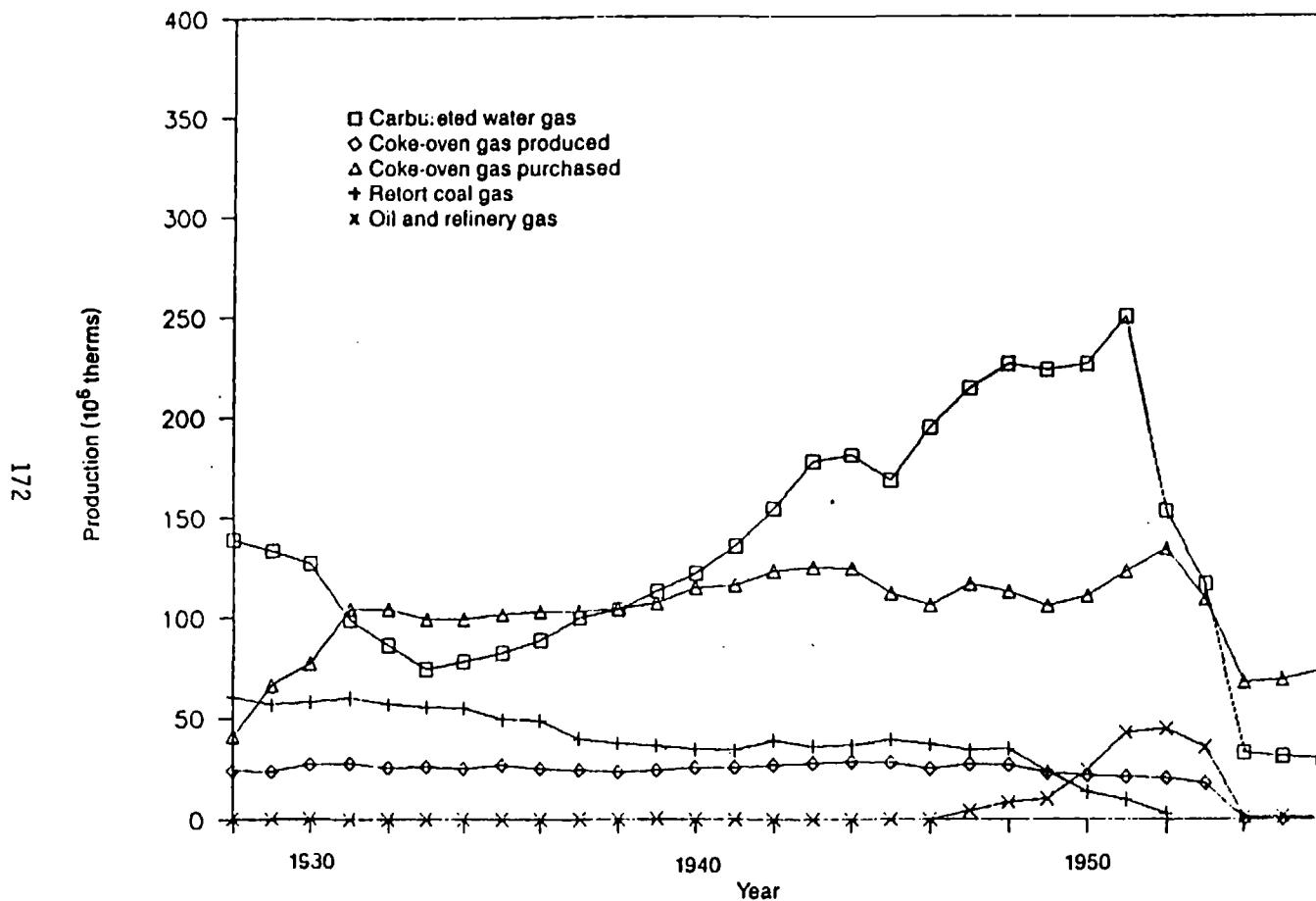
Figure 45. Gas production In the New England States, 1928 to 1956.
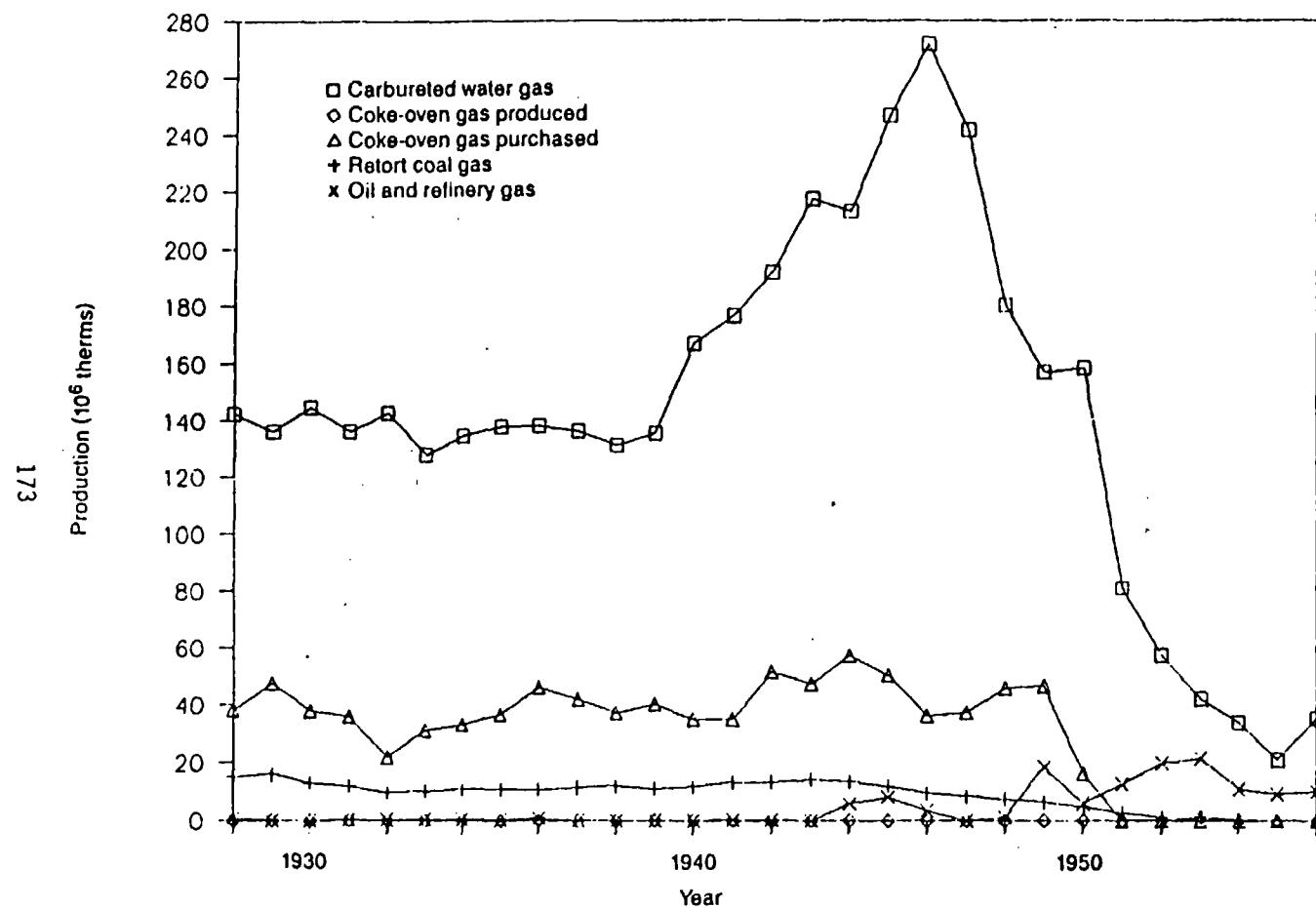
Figure 46. Gas production in the South Atlantic States, 1928 to 1956.

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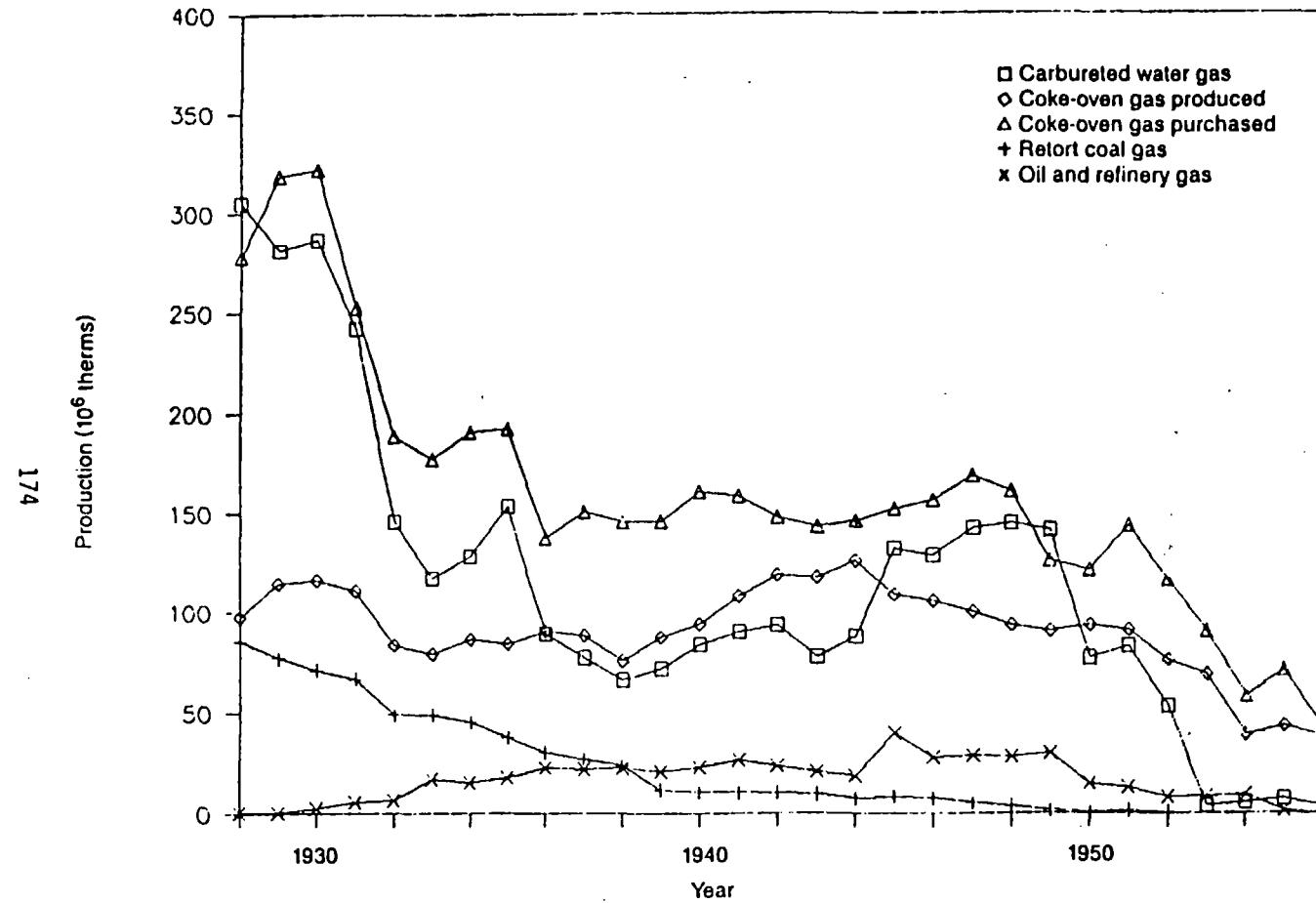
Figure 47. Gas production in the East North Central States, 1928 to 1956.

174
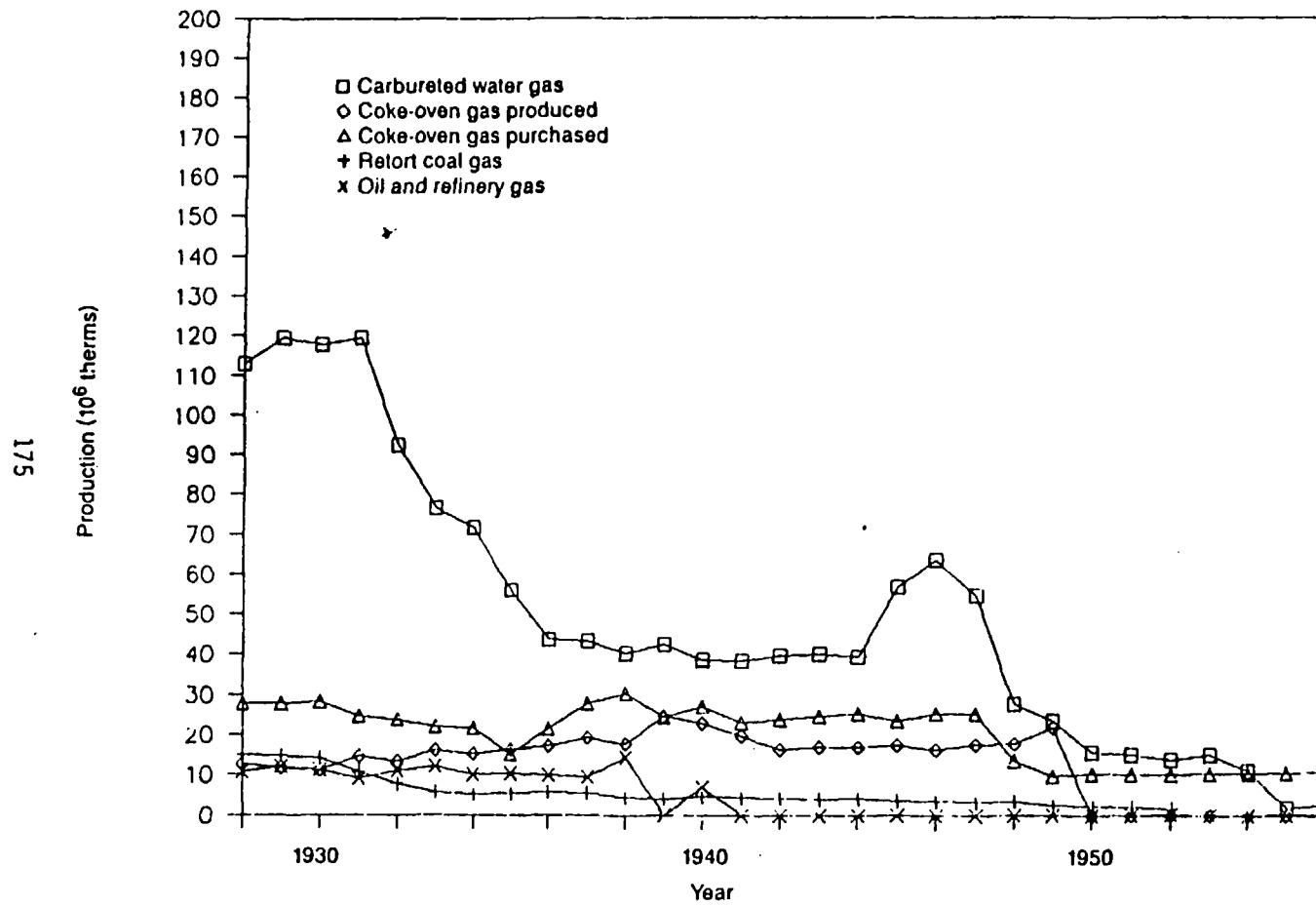
Figure 48. Gas production in the West North Central States, 1928 to 1956.

175
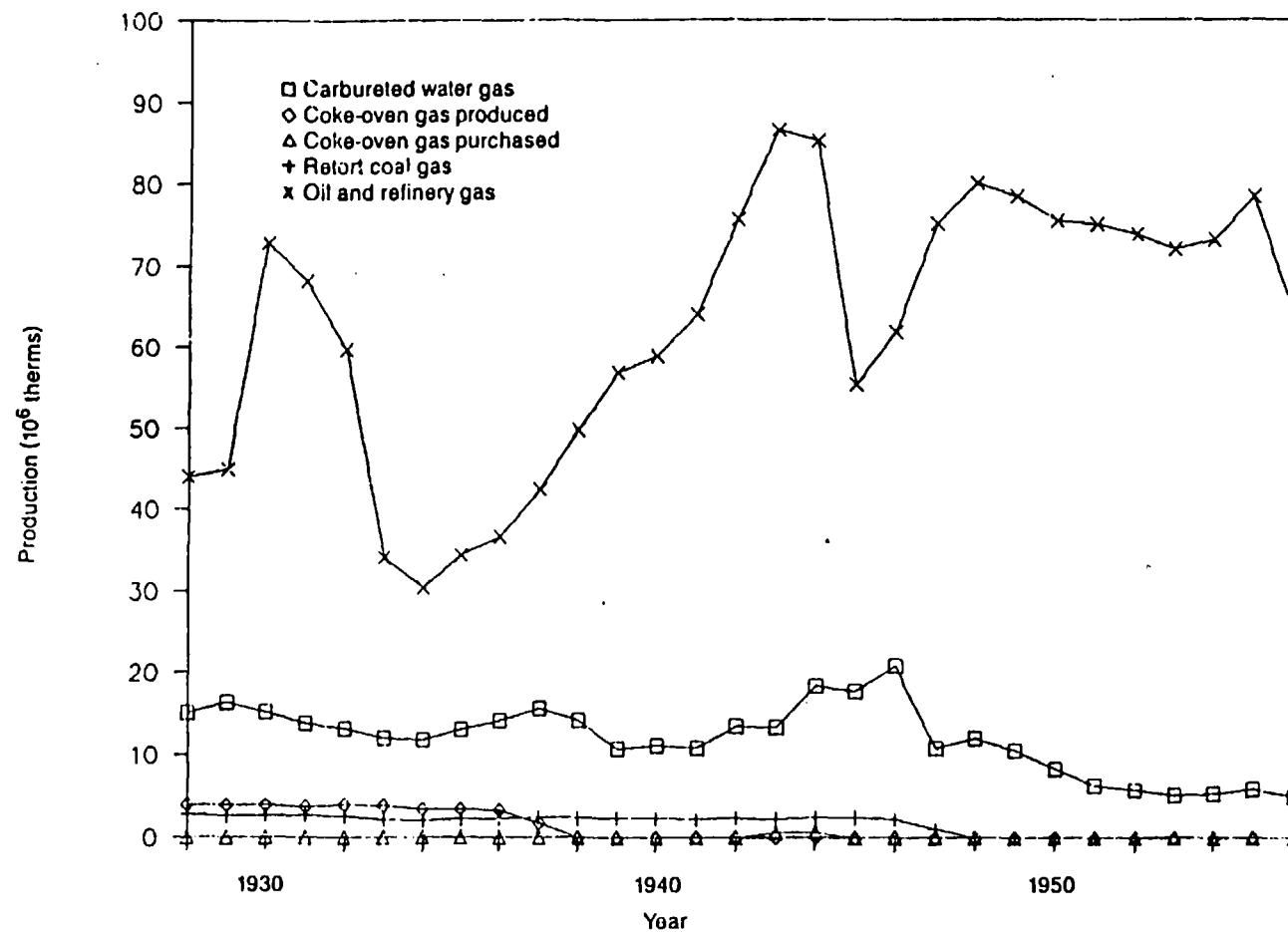
Figure 49. Gas production in the Pacific Coast States, 1928 to 1956.

176
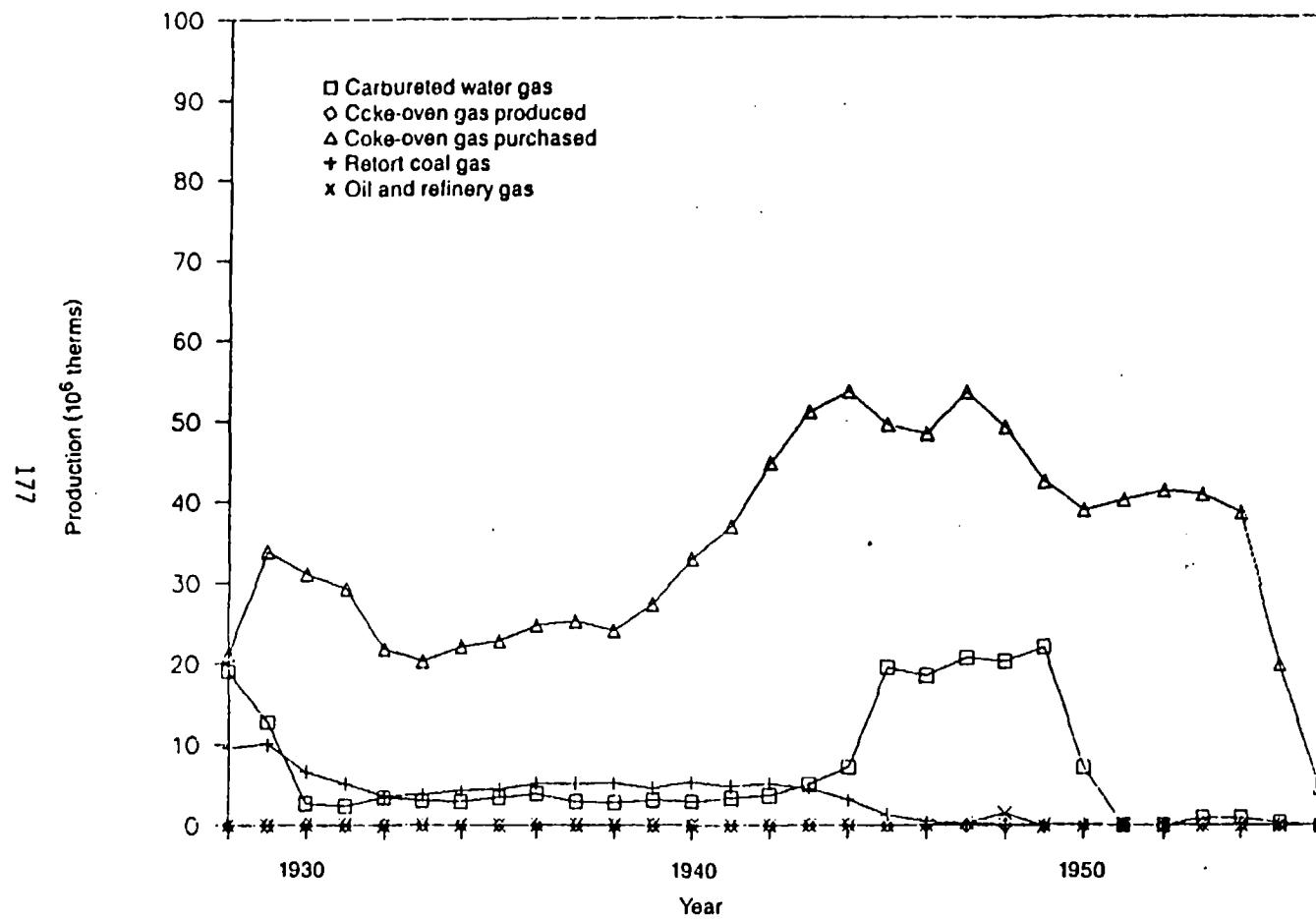
Figure 50. Gas production in the East South Central States, 1919 to 1956.

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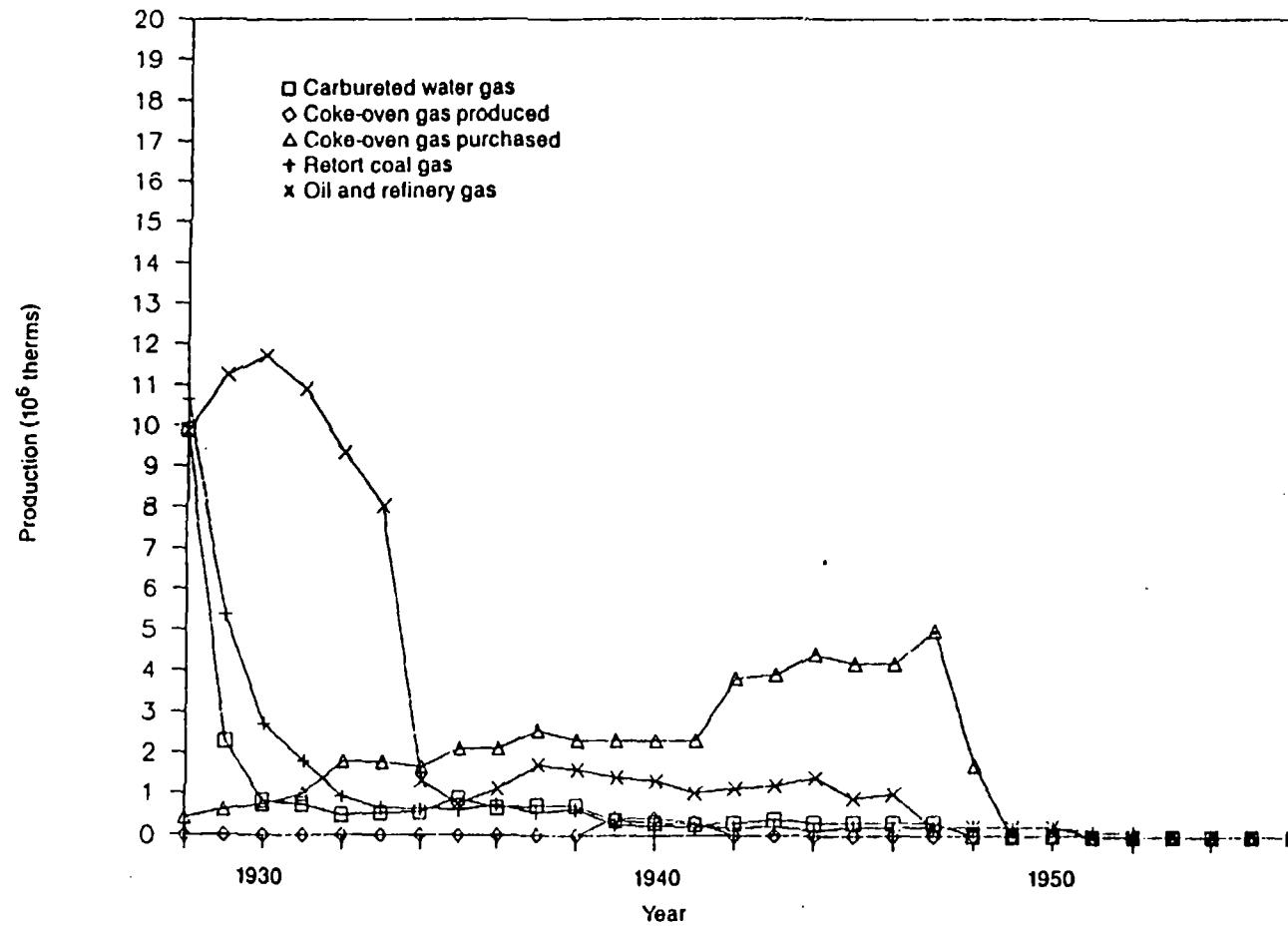
Figure 51. Gas production in the Mountain States, 1928 to 1956.

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Production (10⁶ therms)

50
- □ Carbureted water gas
- ◇ Coke-oven gas produced
- △ Coke-oven gas purchased
- + Retort coal gas
- × Oil and refinery gas

40

30

20

10

0

1930

1940
Year

1950

Gas production in the West South Central States, 1928 to 1956.
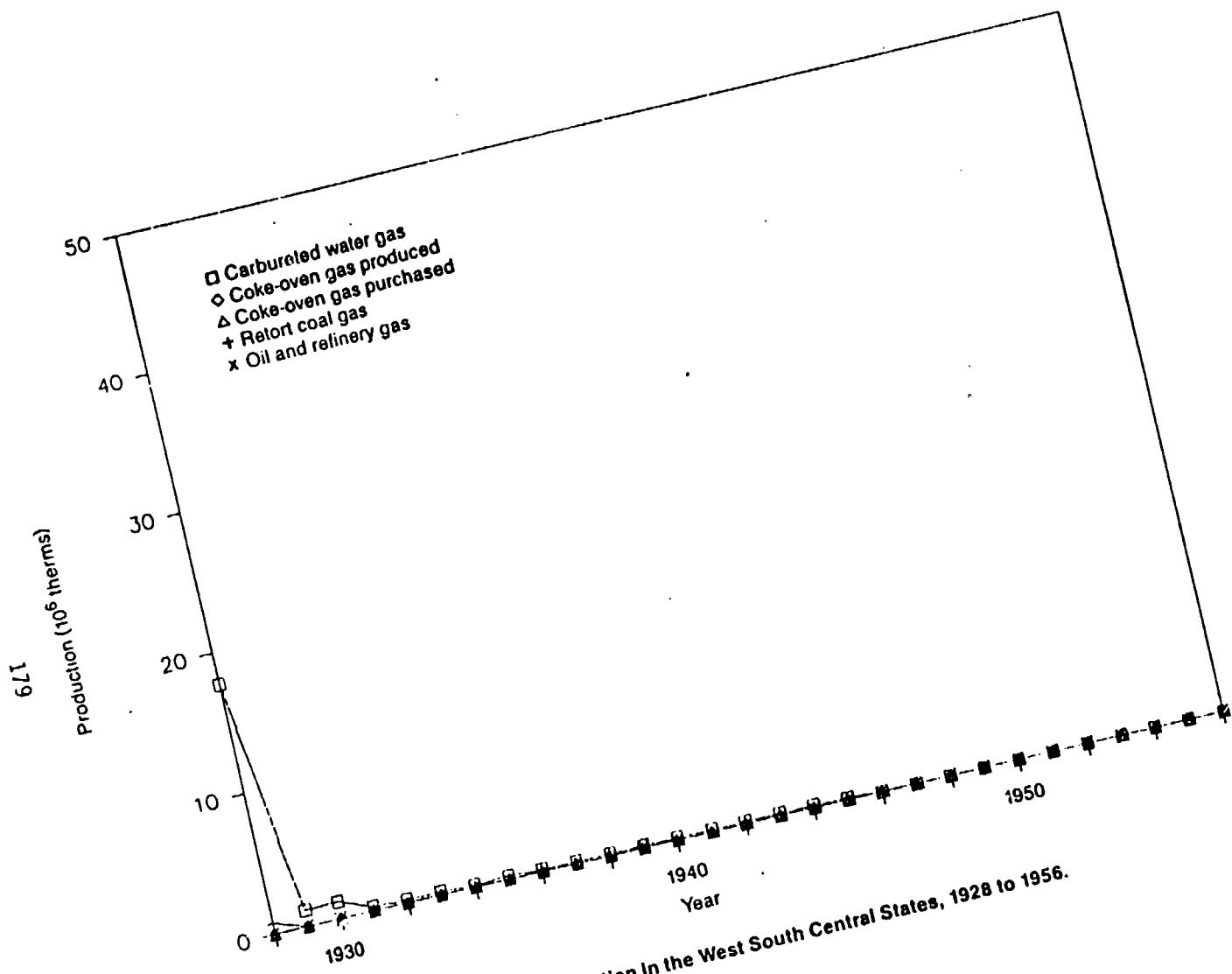
was about twice the CWG production in the region. Sharp drops in CWG production occurred after 1949, and coke-oven gas production and purchases dropped at slower rates. A steady decline in retort gas production occurred between 1928 and 1950.

- Figure 48: West North Central States--CWG production was the major gas produced by gas companies in this region, but CWG production dropped sharply after 1930 and showed a small increase after World War II. Coke-oven gas purchases dropped after 1948, and coke-oven gas production dropped to zero in 1950. Only a small amount of oil gas was produced in the region.

- Figure 49: Pacific Coast States--Gas production in States bordering the Pacific Ocean was principally by the oil-gas process during this period. This figure is somewhat misleading in that by this period California was producing and distributing natural gas, and Oregon and Washington continued to manufacture gas. Some CWG was produced and very little coal-carbonization gas was produced in this region. The oil-gas production shows a very rapid decline at the end of World War II (1945). This is because the oil-gas plants were operated at relatively high levels during the war so that by-products needed for the war effort could be produced. Gas was still being produced at substantial levels through 1956.

- Figure 50: East South Central States--This is the only region examined where coke-oven gas purchases were the major source of manufactured gas. The purchases of coke-oven gas dwarfed the gas production by gas distributors, although CWG was produced for several years after World War II. CWG production declined sharply in 1950, and coke-oven gas was still purchased (probably for mixing with natural gas) through about 1955. No oil gas or coke-oven gas was produced by gas companies during this period.

- Figure 51: Mountain States--This region had very low levels of gas production. Retort gas and CWG were produced in 1928 but declined sharply after 1928. Oil gas and purchases of coke-oven gas predominated between 1931 and 1948. Gas production essentially stopped in 1949.

- Figure 52: West South Central States--There was no significant gas production in this region after 1929. There would be some gas production before this period, however.

Figures 53 and 54 show some early information on gas production in Massachusetts (Grimwood, 1896). Figure 53 shows the amounts of coal gas, CWG, and oil gas produced between 1886 and 1900. This figure clearly shows the

2.0 INVESTIGATION AND REMEDIATION OF TOWN GAS SITES

The investigation and remediation of abandoned town gas sites is a large task, considering the large number of sites that have been discovered and the even larger number that remain undiscovered. Contacts made with State and Federal agencies during the course of this project indicated that, of the sites that have been discovered, only a few have progressed beyond preliminary assessments, and fewer still have had remedial actions implemented to address contamination. Thus, site investigation activities and remedial action activities at town gas sites should increase markedly over the next few years.

As with any uncontrolled site contaminated with potentially hazardous chemicals, site investigation activities should focus on determining threats to human health and the environment posed by the site and on generating the information necessary to evaluate and select remedial alternatives. Selection of remedial alternatives should concentrate on cost-effective alternatives that effectively mitigate the threat, with an emphasis on treatment or destruction alternatives that eliminate the hazardous nature of the wastes. This chapter discusses the behavior of contaminants commonly occurring at abandoned town gas sites, reviews current practices in investigating and remediating these sites, and presents recommended practices based on this review. The case studies, presented in Chapter 3, provide background information supporting the information presented in this chapter.

2.1 CONTAMINANT BEHAVIOR AND FATE

The most commonly occurring and environmentally significant contaminants at abandoned town gas sites are byproduct tars and oils and spent oxide wastes. Significant aspects of the behavior of these contaminants in the subsurface environment are discussed in the following sections.

2.1.1 Byproduct Tars and Oils

Byproduct tars and oils represent multiple-density contaminants at gas-works sites. For the purpose of this discussion, byproduct oils are defined as liquid hydrocarbon from gas manufacture with densities less than water; byproduct tars are defined as liquid hydrocarbons with densities greater than water. These substances are of concern environmentally because of their potential to contain high concentrations of carcinogenic compounds, such as PAH's and nitrogen heterocyclics. From the standpoint of groundwater contamination, the byproduct oils are of most concern because of their higher solubilities and tendency to float on the watertable, where soluble components may be leached out by infiltration. The byproduct tars are also of concern, however, because of their potential to flow in density currents through subsurface fractures and coarse-grained deposits. A discussion of the hydrogeologic behavior of these immiscible, variable density contaminants adapted from Alexander (1984) follows.

Byproduct tars and oils from gas manufacture a e immiscible fluids and as such do not readily mix with groundwater. The flow of immiscible fluids is more complex than is the flow of soluble contaminants. An immiscible fluid that is more dense (e.g., tar) than water will migrate according to the combined effects of relative density and the fluid-fluid and fluid-solid interfacial pressures. Because of the density contrast, the fluid will generally sink within the groundwater. Lighter hydrocarbons, such as byproduct oil, will generally "float" on the water table or on the tension-saturated zone. The existence of capillary pressure in a two-phase flow system means that the migration of an immiscible fluid is not entirely dependent on the flow of groundwater and, as a result, can migrate in an opposite direction of the dominant flow system. It is not uncommon in spills of low-density fluids, for example, for the fluid to migrate "upgradient" of the groundwater flow system within the capillary fringe. The theoretical aspects of multiple-phase flow of hydrocarbons in the subsurface are discussed in detail by van Dam (1967).

One of the biggest problems associated with the release of the lighter hydrocarbons into the subsurface is that their relative solubility increases

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the volume of groundwater that is contaminated. Rainwater that percolates through the "pancake" of light hydrocarbons typically formed over the groundwater body eventually weakens the concentration of oil causing dissolved components of the oil to enter and be transported by the flow of groundwater (Dietz, 1971).

An example showing the soluble component of an immiscible contaminant is provided in Figure 58 with the following designation of zones (from Pfannkuch, 1982):

- Zone I is the above-ground and surface zone where leaked or projected oil runs off and collects in surface depressions, thus forming the area from which infiltration takes place. The configuration of this area depends on the local topography, the amount spilled, and the conditions of release or eruption.

- Zone II is the soil profile. From Zone I the oil starts infiltrating into the subsurface via the organic soil layer, if such a layer is present. This zone is characterized by its high organic content and high moisture content due to soil structure. If the soil is oleophilic, it has a much higher oil retention capacity than do the underlying nonorganic deposits.

- Zone III is the vadose or unsaturated zone. This is the most important zone for oil retention. Water saturates the pore space only partially and ranges in value from zero to field capacity. Oil, as the nonwetting phase, moves downward under the forces of gravity. At first it moves as a more or less continuous phase or "oil body," displacing excess water from the larger pores. When all oil has infiltrated from the surface, the "oil body" will move downward by translation, but small amounts of oil will be left behind the trailing end, trapped as insular disconnected droplets. The oil body continues to move in a disintegrated fashion until all of the oil is trapped in the pore spaces of the vadose zone if its total retention capacity exceeds the infiltrated spill volume. Any oil in excess of this total retention capacity reaches the groundwater body and spreads on the water table through the capillary fringe.

- Zone IV is the capillary fringe that is partially watersaturated, directly connected with the groundwater body vertically, but continuous laterally. When excess oil reaches this zone, it will spread laterally under its own hydrostatic pressure and form a lens on the water table. The spreading will halt when the hydrostatic forces in the oil phase are counterbalanced by the capillary forces at the outer edges of the spreading oil lens. This movement is governed by the phenomena of relative permeabilities and multiphase flow in porous media.
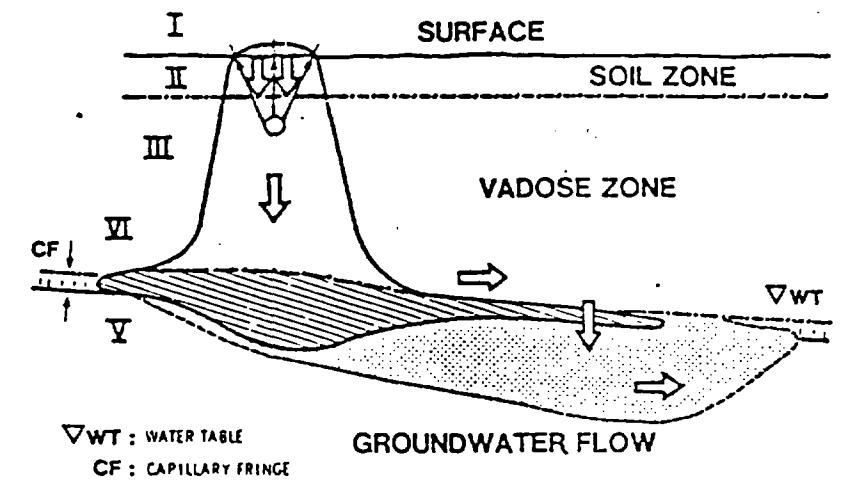
I   Surface zone
II   Soil profile
III Vadose zone (unsaturated)
IV Capillary fringe
V   Groundwater body

Source: Pfannduch, 1982.

Figure 58. Subsurface propagation of a nonmiscible containment.

If the porous medium is homogeneous and isotropic and the water table is horizontal, then the oil lens would be perfectly circular around the center of infiltration. In most realistic cases, the water table has a slope that gives rise to an elliptically elongated lens extending in the direction of the flow. The shape of this lens depends on the water-table gradient, groundwater flow velocities, the capillary properties of the multiphase flow system, and the shape and orientation of the original infiltration area.

* Zone V is the groundwater body. Most hydrocarbon compounds in a spill are lighter than water and therefore tend to float on the water table. Under the hydrostatic head of the continuous oil column, an actual depression and penetration of the groundwater body below the water table occurs. This inverted mound will dissipate as the overlying oil body spreads laterally. The penetration and subsequent retraction may result in leaving trapped insular oil behind in the groundwater body. The most important feature of Zone V in the emplacement stage is the formation of an interface between the bottom of the oil lens and the free-flowing groundwater. It is at this interface that small but significant amounts of hydrocarbon compounds go into solution with the water and are spread by convective and dispersive transport mechanisms.

Model experiments have been useful for studying the mechanism of low-density oil spread in porous media above the water table (Schwille, 1967). The seepage and spreading of heating oil in layers of varying hydraulic conductivity and hydraulic gradients are shown in Figure 59. The oil seeps downward under the influence of gravity, and its geometry is influenced by the rate of infiltration, the hydraulic conductivity, capillarity, and the hydraulic gradient.

Multiple discharges of different kinds of chemicals can lead to a complex pattern of contaminant plumes (Figure 60). In this example, the heavy petroleum product that is denser than water is flowing down the slope of the confining bed in an opposite direction to the flow of dissolved and low-density products. Migration of heavy coal-tar derivatives through density currents is illustrated by a case described by Berggreen (1985), in which creosote has migrated along slickensides (fractures) in a low-permeability clay to bedrock at a depth of 120 feet. Byproduct tar migration through density currents is illustrated by the Brattleboro, Vermont, and St. Louis Park, Minnesota, case studies in Chapter 3.
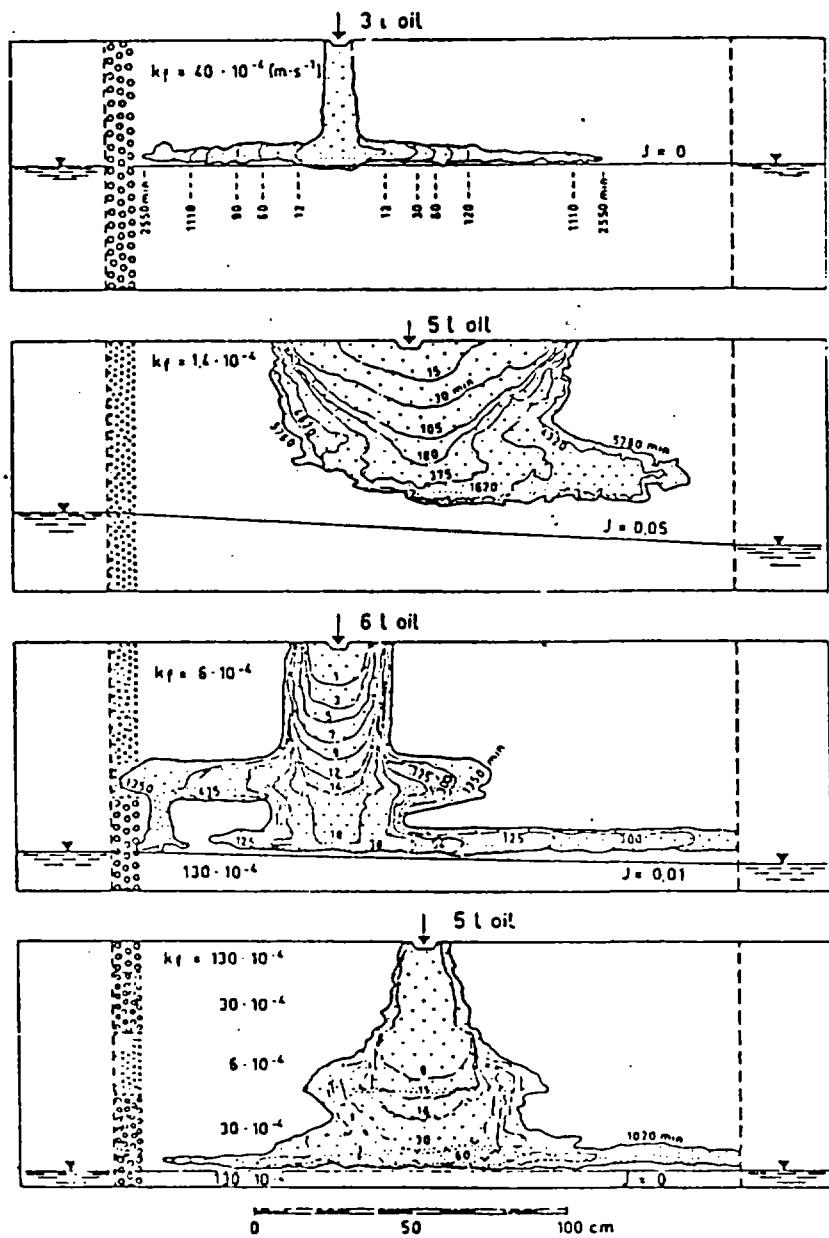
Source: Schwille. 1967.

Figure 59. Seepage and spreading of heating oil in porous media
above the water table.

Figure labels:

SOURCE OF PRODUCT
(Greater density than water)

SOURCE OF PRODUCT
(Lesser density than water)

UNSATURATED ZONE

WATER TABLE

PRODUCT FLOW

DIRECTION OF GROUND-WATER FLOW

FLOW OF DISSOLVED PRODUCT

PRODUCT FLOW

CONFINING BED
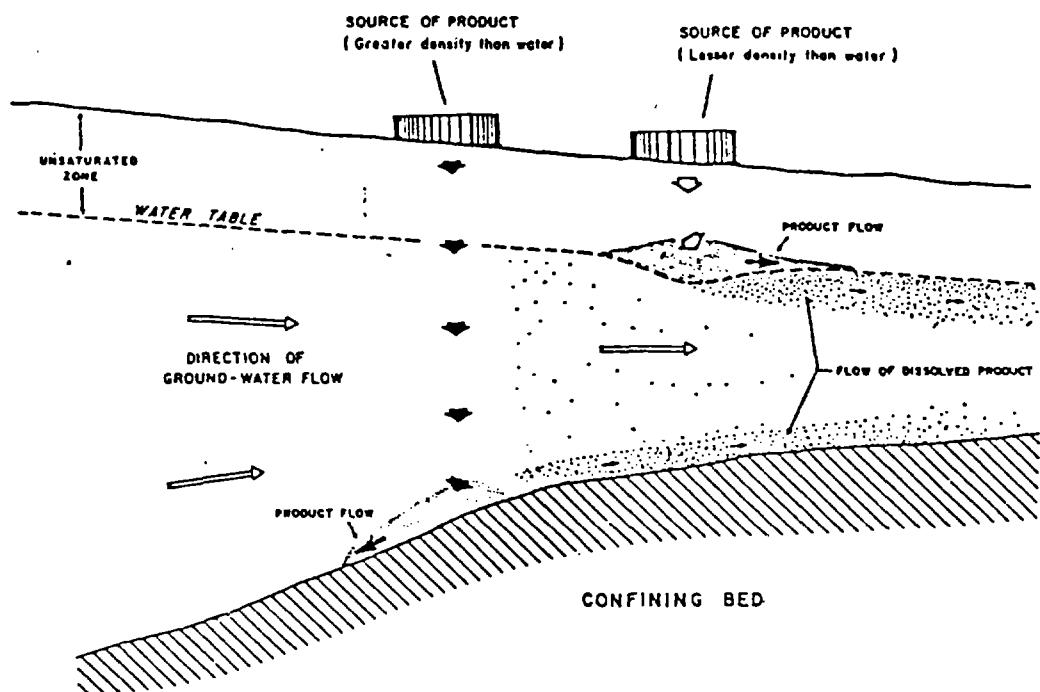
Source: Miller, 1983.

Figure 60. Effects of variable density migration in the subsurface.

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2.1.2 Spent Oxides

Spent oxides are extremely heterogeneous and variable in nature, as discussed in Chapter 1. The most significant contaminants in spent oxide wastes are sulfuric acid, arsenic, and complexed iron cyanides. These complexed cyanides occur in the form of ferric ferrocyanide, imparting a blue color to the spent oxide wastes.

There has been considerable research on the fate and transport of cyanide compounds in the environment by the mining and mineral-processing industry, which uses cyanides to leach metal-containing ores. A recent symposium (van Zyl, 1984) summarized the state of knowledge on this subject, but it also pointed out many gaps in the knowledge necessary to predict environmental impacts accurately. Many of these gaps concerned iron cyanide complexes. Conclusions from this symposium of relevance to this study are:

- Low levels of free cyanides do not persist to soils because of biological and chemical degradation. Biological degradation in soil is inhibited by concentrations of 2 ppm free cyanide under anaerobic conditions and 200 ppm free cyanide under aerobic conditions.

- Ferro- and ferricyanide complexes in solution are photodecomposed to free cyanide. Their toxicity in water is related to the degree of decomposition.

- When KCN in municipal landfill leachate is passed through saturated, anaerobic soil, Prussian blue (ferric ferrocyanide) precipitates and accumulates in the uppermost soil layers. This suggests that Prussian blue is quite immobile in soil.

- Free cyanide migration in saturated, anaerobic soils increases with increasing $CaCO_3$ content and decreases with increasing concentrations of Mn and hydrous iron oxides.

- Complexed iron cyanide ($Fe(CN)_6^{-3}$) migration in saturated, anaerobic soils is retarded by high free FeO_3 and increases with increasing pH and $CaCO_3$ content. At low pH, iron cyanide mobility decreases with increasing clay content.

This information suggests that complex iron cyanides are relatively immobile in a municipal landfill environment and that chemical treatments may be developed for complexed iron cyanides that will limit releases of free cyanides in the soil environment to levels that can be biologically degraded.

2.2 SITE INVESTIGATION

2.2.1 Introduction

Our review of case studies (Chapter 3) and assessment of past disposal practices (Chapter 1) have indicated that appropriate procedures for conducting hydrogeological investigations of town gas facilities are not significantly different from those used for investigating uncontrolled chemical and industrial waste sites. The primary difference is that town gas sites generally tend to be older, and less background information is available about past site activities. In many cases, the present-day site has been cleared, and little or no evidence of past site activities is visible at the ground surface. As a result, research into historical records often is necessary to determine the physical layout and operating history of the plant. As with any investigation of an industrial site, it is extremely important to utilize process information to help determine what contaminants may be present at the site and where these materials may be located.

2.2.2 Current Practices

Most investigations of manufactured-gas plant sites rely on conventional site investigation methods that are not significantly different from contamination investigations of other industrial sites. These methods include surface water sampling, shallow soil and groundwater sampling (from borings and test pits), and, when necessitated by the results of these sampling activities, more extensive groundwater monitoring. In many instances, these methods appear adequate for an initial understanding of the potential for adverse impacts on human health and the environment. A typical approach used in the investigation of manufactured-gas plant sites is summarized in Table 46. Actual case studies are presented in Chapter 3 of this report.

It is apparent from RTI's review of relevant case studies (Chapter 3) that other potentially useful (and often cost-effective) alternative techniques of investigation, such as geophysics and soil-gas sampling, have not been extensively employed at manufactured-gas sites to date. However, based on limited use at manufactured-gas sites and more extensive utilization at industrial waste sites, these techniques show potential utility for screening sites to optimize sampling and analysis plans.

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TABLE 46. SUMMARY OF TYPICAL INVESTIGATIVE APPROACHES FOR MANUFACTURED-GAS SITES

Method	Purpose	Typical activities
Review of historical data	To facilitate the delineation of former plant operations and waste disposal areas.	Review of company records, including business records, construction and utility location plans, tax and insurance records, and geotechnical reports.
	To gain insight to the suspected chemical nature of waste products formed from the gasification process.	Interviews with long-time and former employees concerning past gasification processes and waste disposal practices.
	To assist in the development of a site assessment plan.	Review of older local zoning maps, topographic maps, and soil survey reports, and comparison with more recent versions.
		Review of old aerial photographs and old site photographs.
		Review of any data concerning the locations of water supply and/or groundwater monitoring wells (U.S. Geological Survey, State governments).
Development of site assessment plan	To determine the media of concern and the optimal sampling locations.	Description of site and past site activities.
	To ensure representative and valid sample collection.	Site reconnaissance to identify sampling stations and required sampling equipment and procedures.
	To ensure precision, accuracy, and completeness of sampling data.	Determination of the analytes of interest at each sampling
		Development of a comprehensive quality assurance/quality control plan to include sampling, analysis, chain-of-custody, and data evaluation.
	To identify potential hazards and fulfill health and safety requirements.	Determination of the number of samples required at each station, the volume of sampling material needed for the determination of each analyte, and appropriate collection, preservation, and storage criteria for each sample.
		Development of a detailed health and safety plan that includes an evaluation of potential hazards, the level of protection required for onsite personnel, and emergency contacts.
		Development of an equipment checklist for field and laboratory activities.
Sampling and analysis of surface waters	To identify background water quality conditions in the area.	Collection and preservation of surface water and/or sediment grab samples from sampling stations identified in the site assessment plan in accordance with procedures outlined in the site assessment plan.
	To determine the extent and degree of surface-water contamination.	Analysis of the samples in accordance with the procedures and quality assurance specifications outlined in the site assessment plan.

(continued)

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TABLE 48 (continued)

Method	Purpose	Typical activities
Sampling and analysis of shallow soils	To characterize the shallow-soil media in the vicinity of the site. To identify the location of former waste disposal areas and the extent of soil contamination. To characterize the physical properties of the medium as they relate to fluid transport in unsaturated conditions.	Development of a sampling grid based on the suspected location of former waste disposal or other areas where residuals will occur. Collection, onsite characterization, and preservation of samples from the appropriate depths using hand auger, backhoe, or drilling techniques. Analysis of the samples in accordance with the procedures and quality assurance measures outlined in the site assessment plan. Analysis of soil moisture variations above the water table (using soil moisture probes).
Sampling and analysis of deep soils	To characterize the deeper subsurface lithologic and chemical conditions in the vicinity of the site. Conversion of the soil test borings to groundwater monitoring wells for the purpose of groundwater sampling and analysis. To characterize the physical properties of the medium, particularly as they relate to fluid transport.	Development and implementation of a soil-test drilling program in conjunction with sampling locations specified in the site assessment plan. Collection and onsite characterization of soil/rock samples at specified depth intervals. Collection of geologic data during drilling activities (penetration resistance, water zones, lithologic conditions, etc.). Appropriate preservation of any samples to be analyzed for chemical constituents or physical properties. Analyses of chemical constituents and physical properties as detailed in the site assessment plan. Appropriate field measures for conversion of soil test borings to groundwater monitoring wells, in accordance with State and Federal regulations. Appropriate abandonment procedures for borings not intended for monitoring well conversion. Estimation of field hydraulic conductivity values for specified depth intervals below the water table (slug tests, hydrologic monitoring, etc.).

(continued)

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TABLE 46 (continued)

Method	Purpose	Typical activities
Sampling and analysis of groundwater	To characterize the chemical and physical conditions at or below the water table in the vicinity of the site.	Water sampling in open soil-test borings or in test pits. Conversion of the soil-test borings to groundwater monitoring wells in accordance with State and Federal regulations. Careful consideration of the exact position and depth of screened intervals, the appropriate well-construction and back-filling materials, proper grouting procedures, and installation of protective caps and well-identification labels. Appropriate well-purging measures with consideration of any drilling fluids that have been added. Collection of static-water-level information prior to purging activities and subsequent to well recovery. Collection, preservation, storage, and analysis of groundwater samples from the appropriate depth intervals and in accordance with the procedures specified in the site assessment plan.

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In a manufactured-gas site investigation in Wallingford, Connecticut, ground-penetrating radar was used to estimate the location, extent, and character of tar ponds, in cases where no records were available. The ground-penetrating radar demonstrated that the tar had migrated well beyond the original pond location and the site boundary. Magnetometer surveys were used to locate buried pipes extending from the tar pond to a former lake bed, which could later be investigated by a grid of soil-test borings. Additional geophysical tools used in this investigation included seismic refraction to assist in the definition of the depth to bedrock (a potential controling factor in the subsurface migration of high-density contaminants; see Brattleboro case study, Chapter 3) and electrical resistivity to outline locations of potential groundwater contamination (Quinn et al., 1985). Ground-penetrating radar also has potential for estimating the location and extent of lighter hydrocarbons that may be floating on the groundwater table (Stanfill and McMillan, 1985).

Soil-gas sampling has potential for delineating contamination at a gas plant site when the more volatile fractions of gasifier tar (e.g., benzene, toluene, xylenes, naphthalene) are present at a site. An investigation conducted at the Spencer, Massachusetts, town gas site illustrates this potential applicability. During test pit excavation, site air was screened for volatile organics using a photoionization meter. These measurements were made to assess potential air quality impacts of excavation activities, which were demonstrated to be minimal. However, air in the test pits had substantial concentrations of volatile organics (>200 ppm), levels of concern from the standpoint of occupational safety (Perkins Jordan, 1984). Although the small size of this site would limit the value of using soil-gas sampling as a site investigation technique, the levels of volatile organics suggest that it may be used to help guide sampling and analysis activities at larger, more complex sites.

A discrepancy commonly encountered in the gasworks site investigations reviewed by RTI is insufficient information on the processes that operated at the specific sites. Most site assessments reported that gas was produced by coal pyrolysis or carbonization (i.e., retort or coke-oven gas); most of these sites actually were carbureted water-gas (CWG) plants. The difference is

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significant, both in terms of waste characteristics and byproduct utilization practices (see Chapter 1). For instance, nitrogen and sulfur compounds are more prevalent in coal carbonization tars than in tars from CWG processes. Tar emulsions produced by CWG processes were hard to dewater. As a result, they were not reused and were disposed onsite, especially in smaller plants. Spent oxides from CWG cleanup processes often do not have the brilliant blue color often considered a characteristic of spent oxides because of the absence of significant levels of ferrous ferricyanides. One site assessment report reviewed under this study identified a mixture of yellow and red cinders, but it failed to recognize the material as spent oxide from the small CWG plant. It was not sampled or analyzed, but it could have been a source of contaminants at the site. Historical background information of the gas industry is invaluable in planning and conducting gas plant site investigations because it can provide data on the characteristics and likely disposition of potential contaminants at site.

2.2.3 Recommendations for Site Investigations

2.2.3.1 Introduction--

As discussed in the previous section, site investigation techniques employed for hazardous waste site investigations are generally applicable to former manufactured-gas sites. However, some special considerations should be taken into account when conducting site investigations in order to focus the investigations on characteristic features of these sites. First, as described in Chapter 1 of this report, contaminants, especially gasifier tar and oil, often are contained in below-ground structures that were covered over and left when the plant was decommissioned. Gasworks site investigations initially should concentrate on identifying these structures because they often contain almost pure contaminants. Because such contaminants are contained, they are relatively easy to remove, and because they may be relatively pure, the materials may be reused as supplementary fuel or chemical feedstocks (see Platts-burgh Case Study, Chapter 3). In addition, it is especially important to take extreme care not to damage these structures during site investigation or remediation because this could result in the release and spread of contaminants, complicating and increasing the expense of cleanup operations.

Second, it is important to determine the real extent of contamination on and off a site as wastes, especially solid wastes from gas cleanup operations (e.g., woodchips, spent oxides). Such wastes were often disposed in areas adjacent to but not actually on the original gas plant site. In addition, gas plant sites were usually sited in low-lying areas (to facilitate gas distribution) and were adjacent to streams, lakes, or wetlands. In many cases, wastes were accidentally or deliberately discharged into these areas; recent releases into streams, lakes, and rivers have resulted in site discoveries in many cases. It is important, therefore, to investigate wetlands and waterbodies adjacent to gas plant sites for potential contamination.

Third, it is important to recognize that organic contaminants with various densities commonly occur at gasworks sites. Multiple-density contaminants can result in complex contaminant migration patterns in the subsurface (Section 2.1.1) and can complicate the design and implementation of site investigation and groundwater monitoring. The relative density of potential contaminants should be known, at least qualitatively, during the planning stages of site investigation activities. .

Fourth, it is important to understand the variety of methods used to produce the gas and the resulting variability of byproducts and waste products. By knowing the gas production processes used at a given manufactured-gas site, it is possible to determine the most appropriate chemical analyses for development of the site investigation plan, thereby resulting in lower investigation costs. For example, an assessment plan being developed for a site that used a coal-carbonization process should include analysis of phenolic compounds, nitrogen heterocyclics, ammonia, and cyanides. The analysis of these substances at carbureted water-gas and oil-gas production sites is less important because they usually were produced in low amounts in these processes. In addition, it is important to determine the potential toxicity and other hazards that may be associated with gas plant wastes (e.g., the carcinogenicity of coal tar and the tendency of spent oxides to spontaneously combust) so that adequate provisions may be made for the health and safety of onsite workers and the general public during site investigation and remediation. .

The following is a general approach for planning and conducting site investigations at abandoned town gas sites. Most of the site investigation

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techniques and procedures are the same as those applied to investigate any ground contamination situation; therefore, details of the techniques are not addressed. The approach below recommends a chronological sequence of optional activities that may have applicability to gasworks sites. The discussion focuses on describing how characteristics of gasworks could influence the planning of a site investigation. Because of the heterogeneity of gasworks sites, specific and detailed site investigation plans must be developed on a site-by-site basis.

2.2.3.2 Information Collection and Review--

Because of the age of these sites and the fact that most of the visible evidence on the site (including storage tanks and waste disposal areas) have been destroyed, it is important to review as much available information as possible. Information collection efforts should concentrate on the following:

- Identification of the processes and operating practices that were used at a site, including plant size, gas production processes, types of feedstocks, gas cleanup processes, waste types, waste disposal practices, and byproduct recovery operations. The entire history of the site should be covered, if possible.

- Locations of structures such as retort houses, water-gas production facilities, gas cleanup facilities, storage tanks, etc. Also, locations of waste disposal and fuel stockpiles.

- Information on the activities and historical condition of properties adjacent to the plant, focusing on likely areas for waste disposals (e.g., wetlands).

- Information on the geology of the site (e.g., from old construction borings) and regional geological information.

- Past incidents of contamination release into adjacent bodies of water or encounters with contaminants during construction on the site.

This information can be very helpful in developing a field investigation plan (e.g., locating surface geophysical survey lines, soil borings, and monitor wells). By collecting this information early on during site assessment efforts, one can maximize the efficiency and effectiveness of subsequent site investigation efforts, both in terms of cost and utility of the data collected.

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Information collection and review should begin by obtaining the actual records of the town gas site, including business records, construction plans, geotechnical reports, tax and insurance records, utility location plans, and town plat maps. Old insurance maps can be especially valuable for determining the locations of old buildings and other structures on the site. These maps were published for most towns in the East and in California until the 1950's. They were published by street address, have a scale of 1 inch for 10 feet, and were updated at 10-year intervals. The most recent versions of these maps can ·be obtained from the Sanborn Map Company, Plattsville, New York. Earlier versions are available from the Library of Congress on microfilm.

An excellent source of information about past practices at manufactured-gas plant sites is interviews with old-timers who worked at these sites. Often these persons can provide a wealth of information that is not recorded anywhere. In several of the case studies reviewed, old-timers supplied valuable information on past waste disposal practices, especially information on the locations of old waste disposal areas.

Another important source of information to review when investigating abandoned town gas facilities is old aerial or ground-level photographs of the site and surrounding area. These old photographs generally provide the best record of past site activities. If one is fortunate to obtain photographs spanning several years of the town gas operation, it may be possible to accurately locate sources of potential contamination. As an example, Figure 61 shows the Seattle gas plant on Lake Union late in its operational period (1959); Figure 62 shows it more recently after it was developed into a park. By comparing these photographs, one can associate areas of vegetational stress in Figure 62 with gas plant operations in Figure 61.

U.S. Geological Survey (USGS) and Soil Conservation Service (SCS) maps and publications, information from State geological surveys, geotechnical records, and geological publications should be consulted during a site investigation for background information on local and regional hydrogeology.

Finally, a walk around the site often can prove valuable during information review efforts. Even if structures have been removed above ground, often ground-level evidence remains, such as circular features marking the sites of old gas holders. Often waste disposal areas can be identified, as can surficial contamination by spent oxides (especially when they contain ferric ferro-
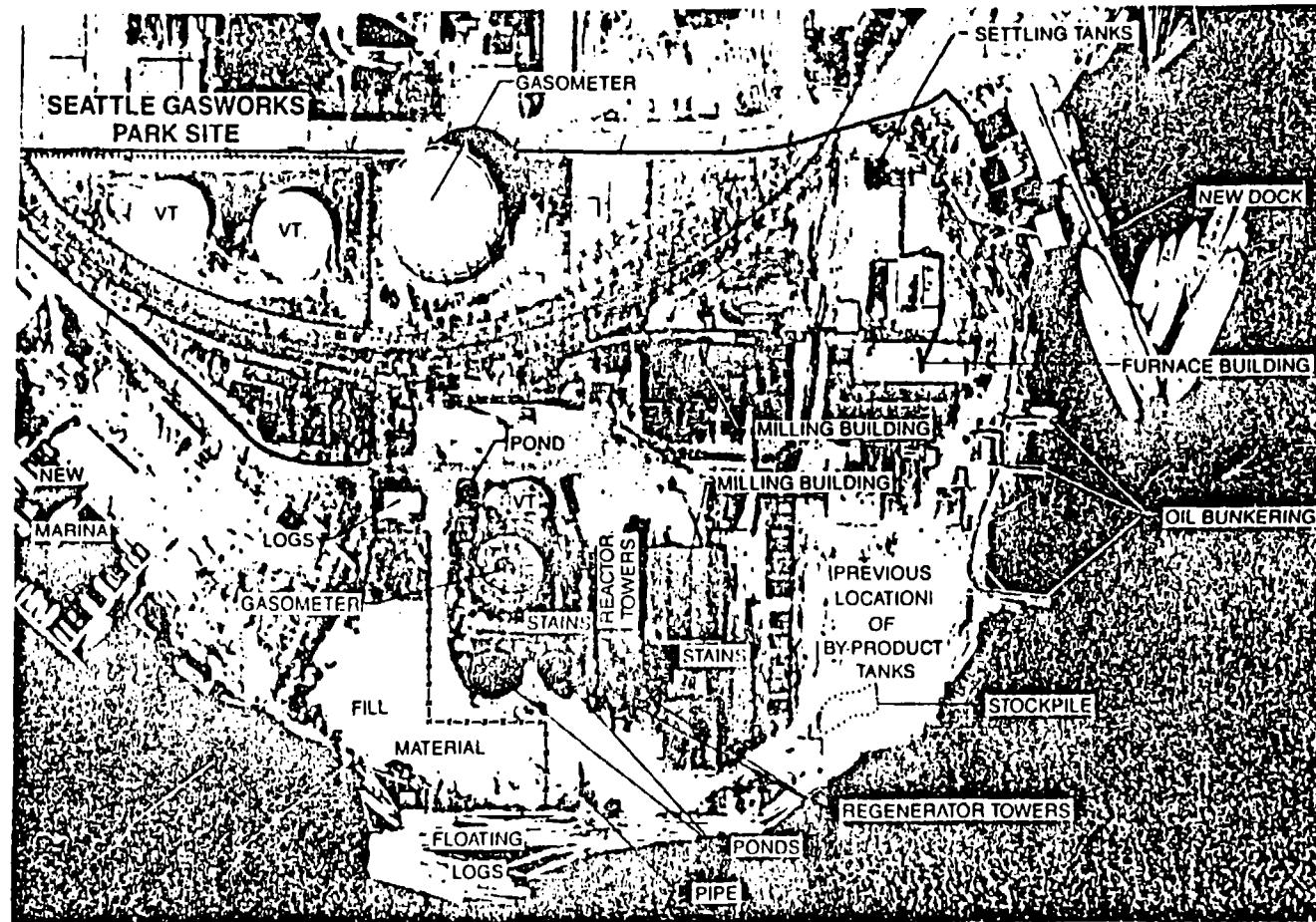
Figure 61. Seattle Gas Works Park site (1959).
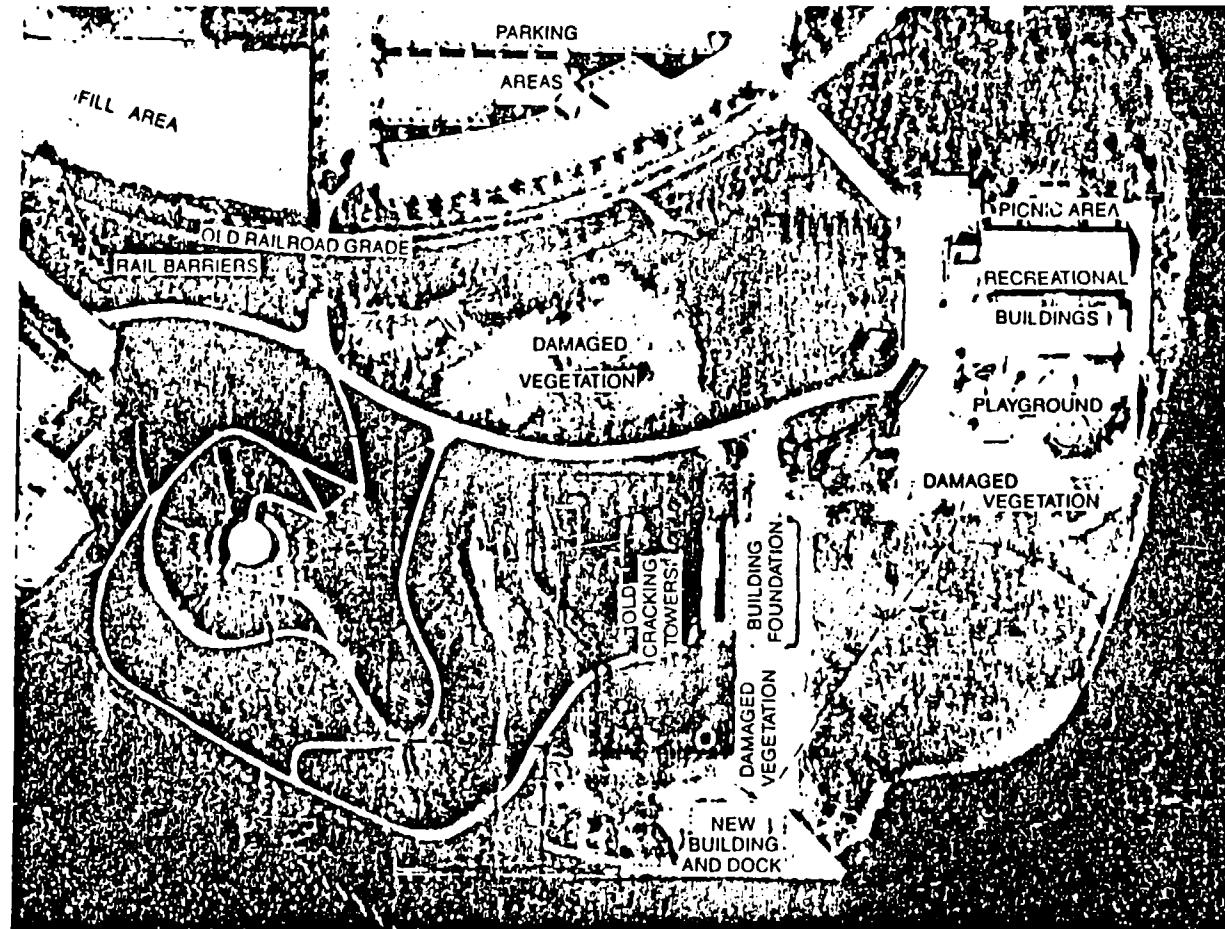
Figure 62. Gas Works Park (Seattle).

cyanides) and tars and oils. Odor also can be used to identify areas of likely contamination during these walk-around preliminary site assessments.

2.2.3.3 Field Investigation Plan Development--

Once background information has been obtained for a site, a field investigation plan should be prepared. This effort should be closely coordinated with local, State, and Federal environmental agencies to ensure that all environmental concerns are properly addressed and that State and Federal site investigation requirements are satisfied. Prior to beginning the hydrogeological site investigation, it may be necessary to submit the field investigation plan to the various environmental agencies for their review, comment, and possibly their written approval.

The plan should consist of a detailed site description, past site activities (including a list of known chemicals used or produced at the site), statement of work objectives, description of proposed fieldwork activities, and proposed laboratory analyses. Also, a detailed health and safety plan should be included.

The health and safety plan should be prepared by a qualified industrial hygienist who should characterize the site for the potential risk to human health by field personnel conducting the site investigation. Safety precautions, including the level of respiratory and dermal protection, should be addressed. Emergency plans and procedures also should be included in the health and safety plan.

The following text describes the field activities that are specified in the field investigation plan. The actual field investigation may deviate from the original plan if unexpected site conditions warrant.

2.2.3.3.1 Surface geophysical survey--Conducting a surface geophysical survey can be an excellent "first step" in a field investigation because it can provide preliminary information about the subsurface conditions of the site. This information may be used to modify the field investigation plan by locating areas where more detailed subsurface investigation may be necessary.

The surface geophysical survey is a valuable tool for investigating old town gas sites for two reasons:

- It provides a method for locating buried storage tanks, buried lagoons, and other buried structures that may contain contaminants.

- It provides a method for delineating contaminants (coal tar and other chemicals) in the soil and in groundwater. However, the ability to detect hydrocarbon compounds in soil and groundwater is limited generally to areas where only high concentrations of these compounds are present.

A number of surface geophysical techniques (ground-penetrating radar, electromagnetics, electrical resistivity, magnetometry, and seismic surveys) can be used to provide preliminary information about subsurface conditions at contaminated sites.

Ground-penetrating radar can be and has been used to detect and delineate pools of organic compounds below ground. However, site conditions can interfere with the operation of this equipment, and it is difficult to predict where it can be used successfully. Applicability, cost, and equipment availability may be factors determining its utilization at specific sites.

Electromagnetic (EM) conductivity is an excellent technique for making a fast and efficient site survey of subsurface anomalies. It can locate old excavations (buried lagoons), buried tanks, pipes, and other metal objects. This equipment also can detect hydrocarbon compounds (tars and oils) in the ground if the compounds are present in high concentrations. Such concentrations are typically represented by low-conductivity measurements at the ground surface because these compounds inherently have very low electrical conductivities. Although EM equipment can locate subsurface anomalies, it may not be able to determined accurately the size, depth, and subsurface condition causing an anomaly.

An electrical resistivity survey can be conducted in conjunction with an EM survey to confirm the EM anomalies and to better define the size and depth of the anomalies. Also, utilizing the electrical resistivity equipment in a sounding and profiling array can help to define subsurface geologic conditions at a site. Electrical resistivity surveying can be used to delineate the depth of the water table as well as the presence of subsurface layers or lenses of different permeability that have contrasting resistivities (e.g., clay and sand layers). However, electrical resistivity methods cannot be applied in certain geologic settings where general subsurface resistivity is relatively high; these methods are best used in areas (e.g., the Atlantic Coastal

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Plain) where electrical resistivities of subsurface materials contrast strongly (White and Brandwein, 1982). Further information on electrical surveying may be found in reports by the U.S. Environmental Protection Agency (1978) and Freeze and Cherry (1979).

Magnetometry may be used to detect buried metal objects at a site. Pipes, drums, buried tanks, and other metal objects may be detected by this method. At one gasworks site, a magnetometer survey was used to locate outfall pipes running from a waste lagoon to a lake adjacent to the site.

Seismic refraction surveys can give valuable information about the depth to bedrock, the subsurface bedrock topography, and the condition (fracturing) of the bedrock (Cichowicz et al., 1981). In addition, the seismic velocity of a geologic material is altered by the degree of weathering and water saturation and therefore can provide information about the variability of these parameters in the subsurface. However, because of the multitude of variables that can affect a material's characteristic seismic velocity, seismic results can be difficult to interpret, especially in areas with complex subsurface geology or in areas where there is little contrast in seismic propagation velocities in the subsurface. For this reason, limited exploratory drilling usually will be necessary in conjunction with seismic surveys to confirm interpretations based on this technique (Cichowicz et al., 1981). More detailed information on seismic refraction surveying may be found in Dobrin (1960).

The selection of geophysical techniques depends to a large degree on the geologic setting (White and Brandwein, 1982) and local site conditions. In general, surface geophysical methods can be utilized on most town gas facilities. However, there are certain sites where geophysical methods may not be appropriate because of local site conditions. Proximity to power lines, metal fences, railroad tracks, and buried utilities may make it difficult to properly interpret geophysical data. In many cases, the type of geophysical tool best suited for a specific site is often difficult to determine without onsite testing. Further information on the application of surface geophysics to groundwater investigation may be found in Zohdy et al. (1974).

2.2.3.3.2 Soil sampling--Soil sampling includes soil-test borings and test pits, soil-water sampling, and soil-gas sampling. These activities are

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the most important means to determine the extent and nature of contamination at a gasworks site. They provide samples for contaminant analyses and document the subsurface conditions at the site; extensive soil sampling is necessary prior to planning remedial actions at a site. A soil sampling program should be directed toward determining subsurface stratigraphy, properties of the subsurface materials that are important to contaminant transport (e.g., permeability, clay content, primary and secondary porosity), and obtaining representative samples of wastes and contaminated soil and water for analytical characterization. This section briefly reviews the important aspects of a soil sampling program. For more information on soil sampling and monitoring, see U.S. EPA (1984a).

A particularly important activity in a soil sampling program is to determine the proper number, location, and depth of the soil borings. Existing information collected during the initial phase of a site investigation as well as surface geophysical results are extremely valuable in planning a site-specific test-boring program. This program should be directed toward delineating the extent and characteristics of contamination at the site and in determining the characteristics of the subsurface soil and rock material. Soil-test borings are typically drilled using hollow stem augers so that the borings can be converted easily to groundwater monitor wells. Also, this drilling technique minimizes the potential for aquifer contamination compared to other drilling processes.

Down-hole geophysical methods can be utilized in soil-test borings where complex geology (including multiple aquifer systems) is anticipated. Various geophysical tools can be used to provide a variety of continuous down-hole data that is useful in determining the presence of contamination and interpreting soil stratigraphy. Down-hole geophysical methods are especially helpful in delineating relatively thin clay and sand layers that may not be detected by discontinuous soil-boring sampling methods (Keys and MacCary, 1971).

Test pits, usually constructed using backhoe excavators, allow for more complete inspection of subsurface conditions than do soil borings. Features such as vertical fractures or sand lenses, which may present pathways for contaminant transport and can be difficult to detect in soil boring, can be readily observed in test pits. Test pits offer a means to determine the

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continuity and persistence of such features in the subsurface. They also may be used to delineate pockets of contamination and to investigate buried structures on the site. Test pits require the excavation of considerable amounts of soil. Because this soil can be contaminated, adequate provisions should be made prior to excavation for the safe handling, transportation, and storage of contaminated soil.

Other reconnaissance techniques that may be used during soil sampling efforts are soil-gas monitoring and soil-water sampling in open boreholes and in the vadose zone. Soil-gas monitoring is generally accomplished in one of two ways. One method involves penetrating the partially saturated and capillary fringe zones above the water table with a pressure-driven probe or auger through which soil gas is withdrawn and collected. Soil-gas samples are then analyzed for volatile components onsite, commonly with mobile gas chromatography, or taken to a laboratory for later analysis. An alternative soil-gas sampling method requires that passive vapor collectors be installed within 5 feet of the ground surface. The vapor collectors remain buried for a period of days to weeks; when exhumed, they are taken to a laboratory where the vapors are released and analyzed. Although both methods are relatively quick and inexpensive ways of qualitatively characterizing subsurface organic contaminants, they are limited to compounds with relatively low water solubilities and high vapor pressures that are capable of diffusing through porous media. In general, soil-gas monitoring has little utility at sites that lack the more volatile fractions of coal tar, e.g., benzene, toluene, xylene, or naphthalene. If these components are present, however, soil-gas monitoring may prove successful in qualitatively characterizing the extent of contamination at a site.

Soil-water sampling is very similar to soil-gas sampling except that a water sample is collected. Drill-stem sampling collects the sample in open boreholes at the top of the water table. Drill-stem sampling offers some advantages over soil-gas sampling in that dissolved nonvolatile and volatile organic and inorganic contaminants can be measured. The method offers cost savings when compared to conventional groundwater monitoring techniques using permanent well installations. Soil-moisture profiling in the partially saturated or vadose zone can be accomplished by a modified soil-gas sampling probe

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or by a number of geophysical methods (e.g., neutron scattering or gamma-ray absorption). Once a sufficiently moist horizon is located, suction lysimeters with porous clay cups can be installed in the vadose zone for sampling soil water. In practice, soil-gas sampling probes have a water-sampling capability, so the advantages of both methods can be combined.

As stated previously, it is very important to take special care when using invasive site investigation techniques (e.g., borings, test pits) at abandoned gasworks sites to avoid penetrating or otherwise damaging buried structures such as tanks, gas holder foundations, or tar separators. These structures often contain tars, oils, or other contaminants. Structural damage could result in their release and spread of contaminants, complicating cleanup efforts.

2.2.3.3.3 Groundwater monitoring--The major objectives for installing a groundwater monitoring system are to:

- Measure water levels for the purpose of determining gradient and direction of groundwater movement

- Perform in-situ permeability tests

- Sample groundwater for chemical analysis.

This section discusses the means to achieve these objectives with specific emphasis on monitoring considerations for abandoned gasworks sites. More detailed information on the design and installation of groundwater monitoring systems may be found in Barcelona et al. (1983), Barcelona et al. (1985), Todd (1980), Fetter (1980), Freeze and Chowy (1979), Johnson Division (1975), Villaume (1985), and NWWA/API (1984).

The number, spacing, depth, and well screen length of monitoring wells may be determined based on background information collected about a site and on the findings of the soil sampling and surface geophysical monitoring programs.

It is important to properly space the monitor wells across the site so that the gradient and direction of groundwater movement can be measured to determine groundwater flow directions and velocity at a site. On small sites it may be necessary to locate monitor wells offsite to discern measurable differences in groundwater levels. If multiple aquifers or perched water

table conditions are suspected, it is suggested that nested piezometers be installed at selected depths to measure vertical gradients. In-situ variable head permeability tests should be performed in selected monitor wells representing various geologic conditions across the site. The permeability measurements along with the groundwater gradient data are useful in estimating the average velocity of groundwater movement across the site.

For groundwater quality sampling and analysis, it is important to have a good distribution of monitor wells upgradient and downgradient from the suspected source of groundwater contamination. The upgradient monitor wells provide the background (uncontaminated) water sample. It may be necessary on small old town gas sites to use offsite wells upgradient of the site as background wells. The downgradient monitor wells should be well spaced and have variable-depth well screens for the purpose of determining the vertical and lateral extent of groundwater contamination. It is also suggested that a downgradient monitor well be placed near the property boundary to determine if the suspected contaminant plume has migrated offsite.

Variable density contaminants have been observed in the subsurface investigations of several manufactured-gas sites and can result in complex contaminant migration patterns in aquifers. The potential for variable density fluids needs to be recognized to the appropriate design of groundwater monitoring systems at manufactured-gas sites. Adequate groundwater monitoring in flow fields with significant density contrasts requires careful monitoring well design and placement to avoid costly redrilling efforts or the creation of undesirable conduits for contaminant migration. Although single well installations that are properly screened within a groundwater flow system may be adequate for some variable density situations, it may be necessary to supplement single wells with multiple-level sampling to fully characterize the vertical extent of contamination. It is also important to compensate measurements and sampling activities for differences in density where significant contrasts exist. Because the variable density contaminants commonly occur at abandoned town gas plants, special monitoring considerations for immiscible, multiple density fluids in groundwater are discussed below.

The relative density of potential contaminants at a gasworks site should be understood, at least qualitatively, before implementation of a groundwater

monitoring program. In some cases, the relative density contrast may be obvious, such as with low-density (coal oil) or high-density (coal tar) immiscible contaminants. However, soluble components of the contaminant also may be present, especially when low-density immiscible contaminants occur (as discussed in Sections 2.1.1 and 2.3.3.3.5), and these need to be considered in the design of the monitoring system (Figure 63). In this example, the downgradient well closest to the source area may encounter immiscible and soluble compounds, whereas further downgradient, the monitoring well will encounter only soluble compounds. A multilevel groundwater sampler would be useful in this example to detect migration of the soluble component and its stratification within the groundwater. Lysimeters or similar in-situ pore-water samplers might be useful in delineating the dimensions of the contaminant plume above the water table (Figure 63).

In other situations, contaminants migrating from a gasworks site may consist of constituents with multiple densities (Figure 64). In this example, downgradient well A will detect an intermediate density zone, and well B will detect the higher density zone. A multilevel sampler (well C) can be used to further delineate the two relative density zones.

The position of the screened interval of monitoring wells (or intake ports of multilevel samplers) is one of the most important aspects of detecting variable density contaminants in the subsurface. This is illustrated in Figure 65 where examples of appropriate and inappropriate monitoring techniques are compared for variable density contaminant situations in a uniform flow field. In example 1, the high-density contaminant solution could be overlooked as a result of shallow screen settings of the monitoring wells. Deeper-screened settings would be more appropriate in this example (nested wells A, B, and C), or a multilevel sampler (well D) would allow for more complete definition of the vertical extent of contamination. For example 2, the low-density immiscible contaminant could be largely overlooked if screened intervals were too deep below the water table. Shallow monitoring wells would be more appropriate in this situation, particularly for defining the depth of the depressed water table. In example 3, the contaminant solution has a similar relative density as the groundwater, but it is not detected by the shallow screen setting of well A. The long screen interval of well B intercepts the
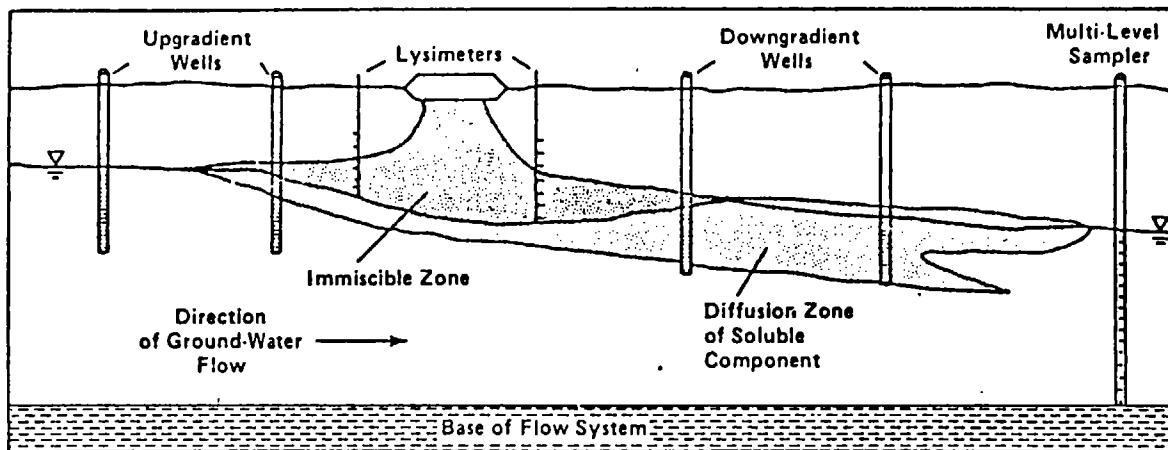
Source: Alexander, 1984.

Figure 63. Monitoring of low relative density containment with immiscible and soluble components.
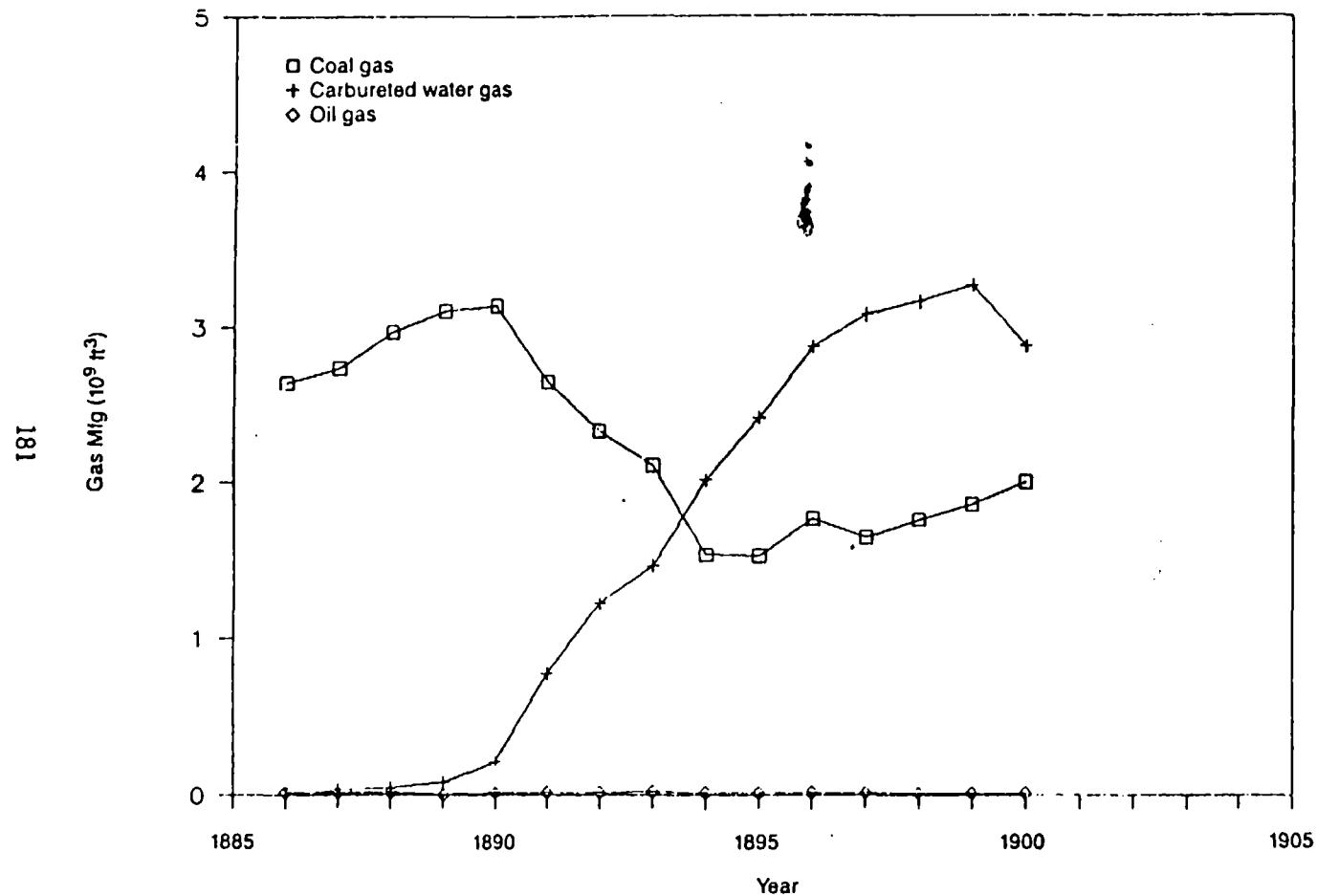
Figure   53. Gas production in Massachusetts, 1886 to 1900.
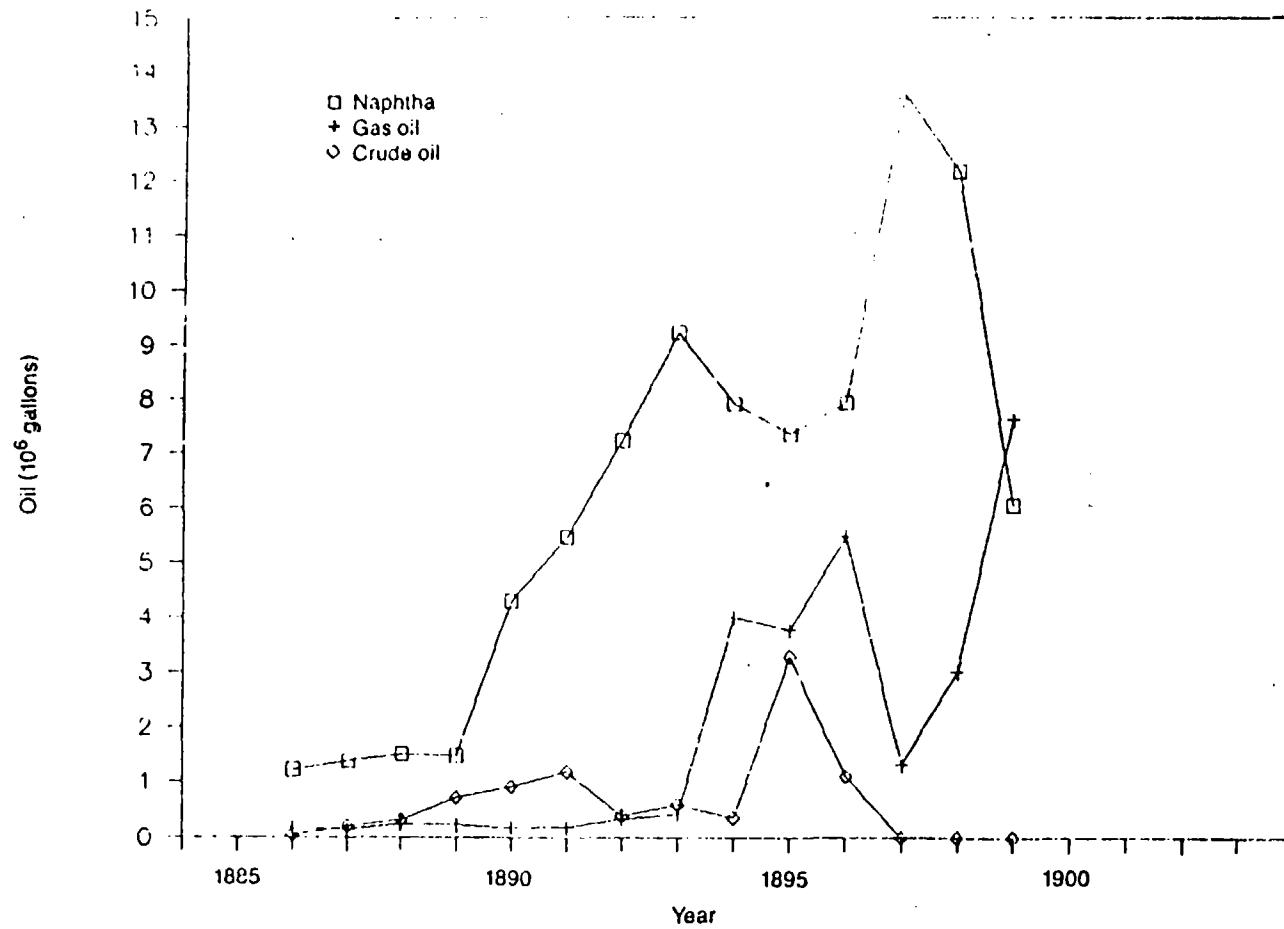
Figure   54. Use of enriching oil in Massachusetts, 1686 to 1900.

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increase in CWG production after 1890 while coal gas production fell, due principally to the replacement of coal-gas retorts by CWG apparatus. Similarly, Figure 54 shows the use of enriching oils increasing with increased CWG production. Naphtha was the major carburetion oil used during this period, but gas oil and crude oil were also used. The changes in gas-oil use and naphtha use between 1896 and 1899 are exactly opposite. When gas-oil use increased, naphtha use decreased; likewise, when naphtha use increased, gas-oil use decreased. This indicates that either of the two feedstocks could be used, with the amounts of each purchased dependent on price and availability.

The regional gas production shown in this section shows clear patterns of variation with respect to the production methods employed in the various U.S. regions and in the relative amounts of gas produced within the regions.

1.5.3 U.S. Gas Feedstock Trends

Just as there were trends with respect to the types of gas produced, there were also variations of the types and amounts of raw materials used in the production of gas. Two major types of feedstocks were used in the production of town gas--solid carbon-based fuel and liquid oils. Figure 55 shows the use of solid fuel for gas manufacture between 1919 and 1965. Two types of coal (anthracite and bituminous) and coke produced from bituminous coal were used in the manufacture of gas. Anthracite coal was used as both generator fuel (for CWG and producer gas) and as boiler fuel. The use of anthracite declined before 1930 because reduced supplies of anthracite increased costs of the fuel. Coke was used primarily in the gas generators of CWG apparatus, and some of the coke was used for producer gas and as boiler fuel. The rise in coke use prior to 1930 is from the increased production of CWG. Coke was produced from bituminous coal in either retorts or coke ovens. Figure 55 also shows the characteristic drop in fuel use during the Great Depression and increasing fuel purchases during World War II. The decline in solid-fuel purchases after 1950 parallels that of the gas-manufacturing trends.

Figure 56 shows the total oil used in gas manufacturing between 1919 and 1965. Oils were used primarily for the carburetion of CWG and for the production of oil gas, but they were also used as boiler fuels by the gas producers. Figure 57 shows the types of oils used between 1945 and 1952. The major trend shown in this figure is the substantially increased use of other heavy oils

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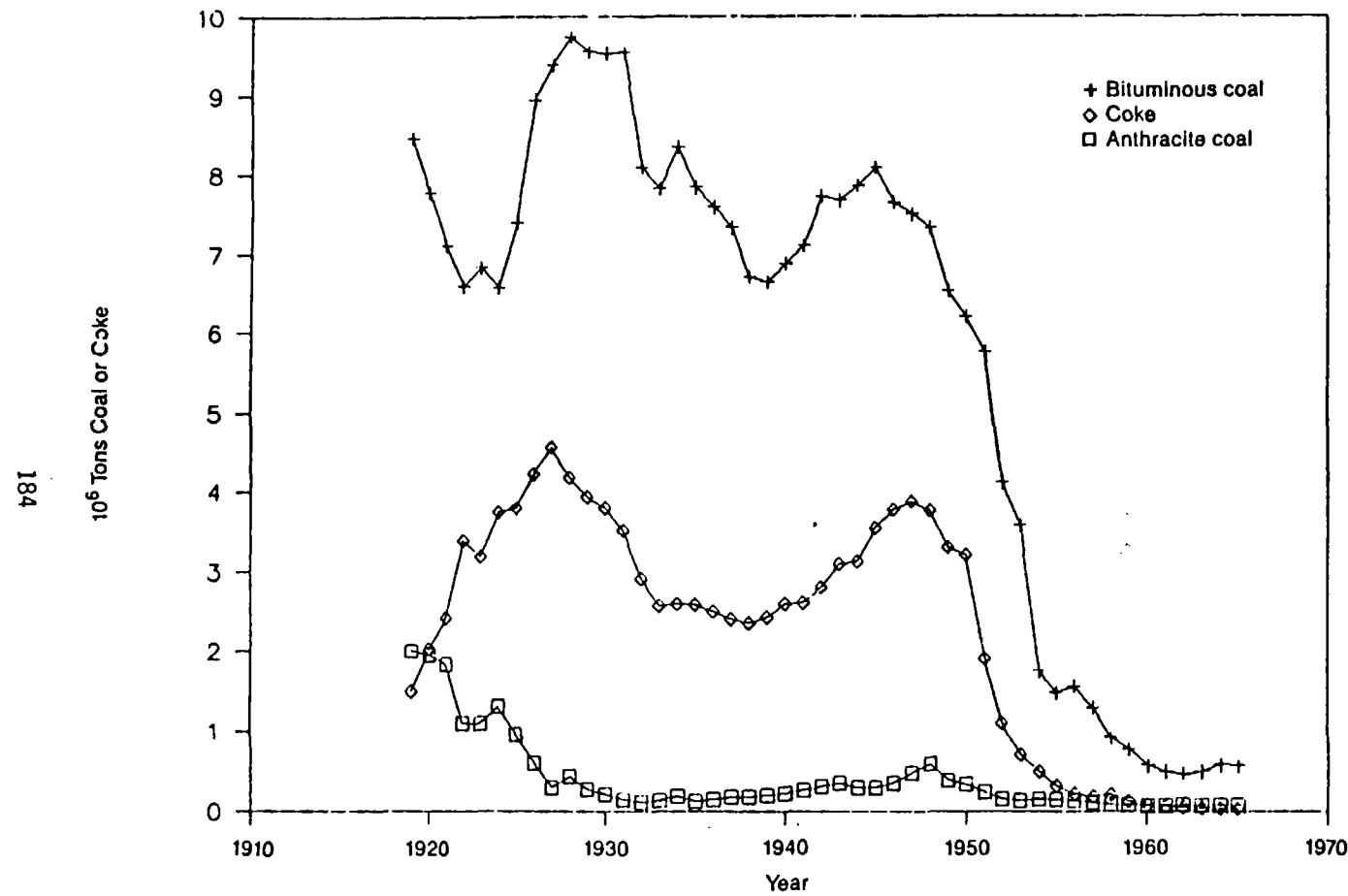
Figure 55. Solid fuel used for gas manufacture, 1919 to 1965.

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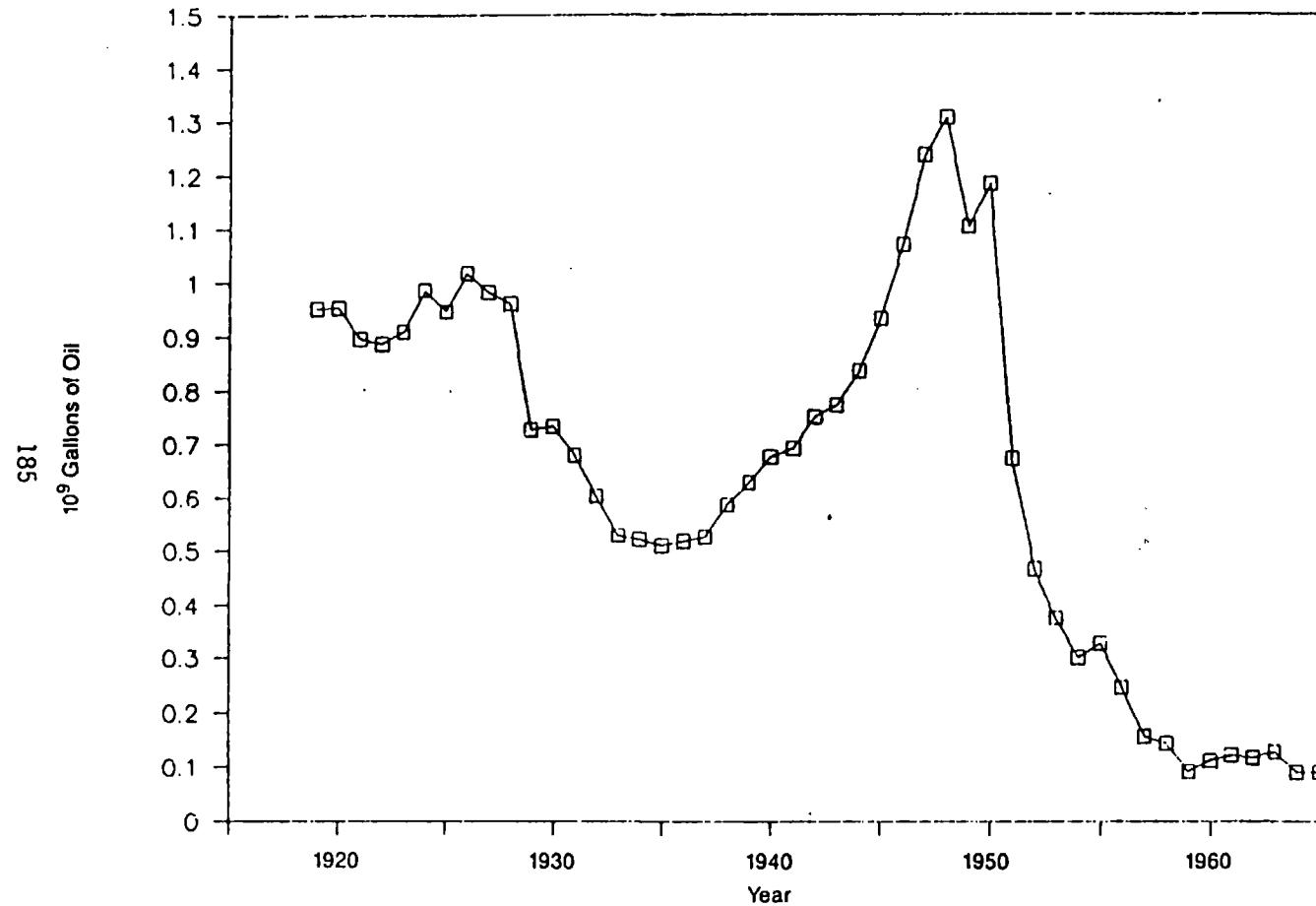
Figure 56. Total oil used in gas manufacturing, 1919 to 1965.
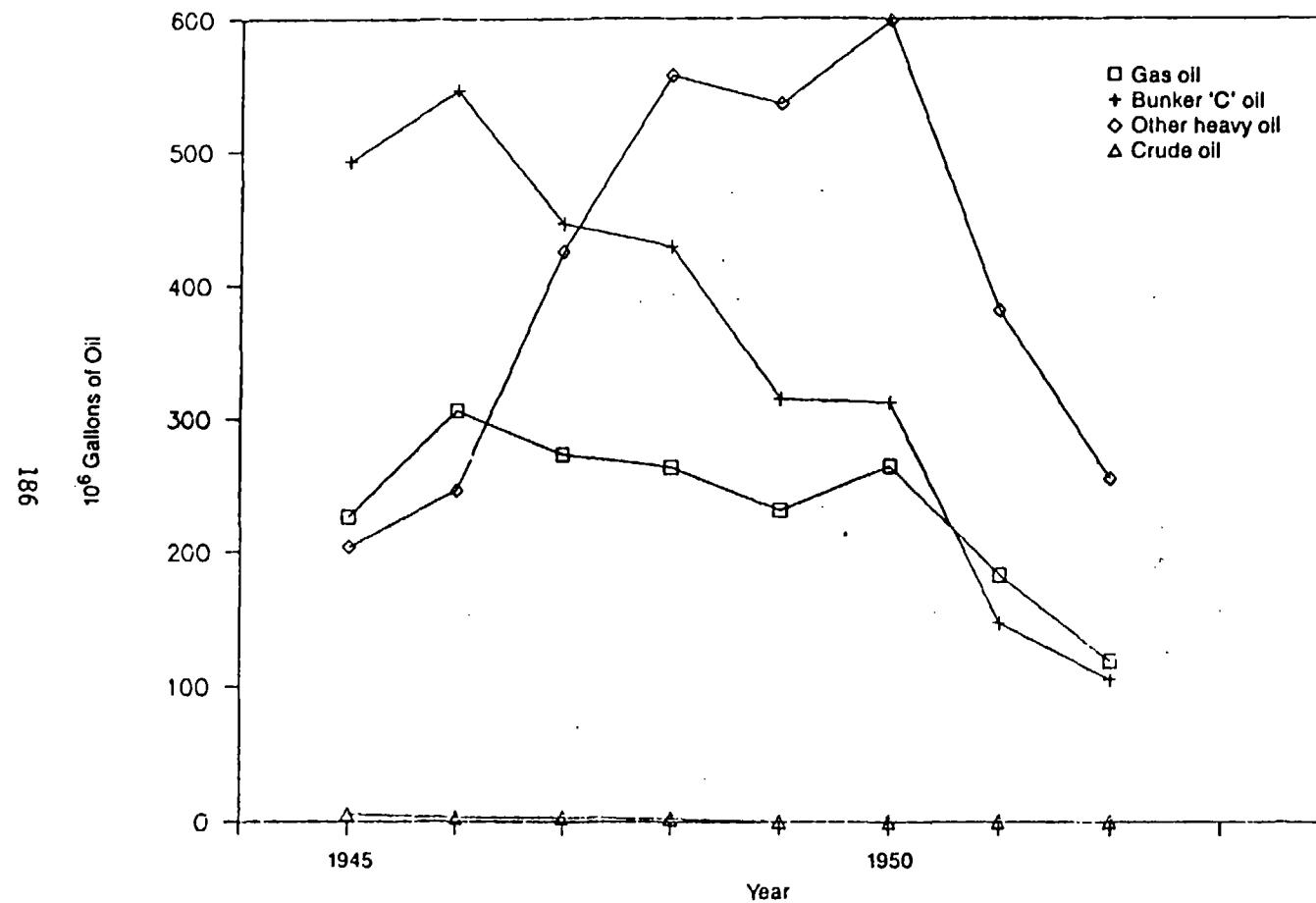
Figure   57. Oil use for gas manufacture, by oil type, 1945 to 1952.

between 1945 and 1950. Because the production of CWG also increased during the same period, most of this increased production used other heavy oils (which were principally the heavy residuum oils that remained after the catalytic cracking of gas oils). The other use of other heavy oils inc ied as the use of lighter Bunker "C" oils decreased during the period, indicating that gas manufacturers switched from tne C oils to heavier oils. Because there were more tars and lampblack created and more emulsion problems associated with the use of the residuum oils, this change in oil feedstocks increased the amount of waste produced by the industry.

1.5.4 Historical Events of the U.S. Gas Industry

Table 45 is a listing of the significant events in the manufact ired-gas industry. This listing includes many of the developments in gas production, purification, markets, and feedstock usage that affected the types ano character of waste produced by the town gas industry.

1.6 DIFFERENCES BETWEEN THE U.S. AND BRITISH GAS INDUSTRIES

The redevelopment of gas production sites has occurred much more fre- quently in Great Britain than it has in the United States. The Harwell report on the problems arising from the redevelopment of gas sites (Wilson and Stevens, 1981) was published several years before a somewhat similar work was published in the United States (Handbook on Manufactured Gas Sites, Environmental Research and Technology [ERT], 1984). There is a tendency to apply the information from the British work on site redevelopment directly to U.S. sites. This section outlines the major differences between the U.S. and British gas industries, and it relates those differences to current waste problems at U.S. sites.

In the United States, the availability of petroleum and petroleum distillates encouraged their use for the production and enrichment of town gas. British gas was primarily coal gas and coke-oven gas, reflecting the abundance of coal in the United Kingdom and the absence of significant oil resources. Because the tars produced from oil-gas and CWG production are generally less viscous than coal tars, the problems of tar migration from the U.S. facilities are probably greater than are the tar migration problems associated with the U.K. coal-gas plants.

TABLE 45. SIGNIFICANT EVENTS OF THE TOWN GAS INDUSTRY

Year	Event	Reference[a]
1806	A home and street lighted by manufactured gas in Newport, RI	Tunis, 1933; Morgan, 1926
1809	Milk of lime used for H_2S removal in Britain	Powell, 1945a and 1945b
1812	Company chartered to light London streets	Rhodes, 1966a
1815	English patent for oil-gas production issued	Rhodes, 1966b
1815	English patent for oil-gas process issued	Hull and Kohloff, 1952; Rhodes, 1966b
1816	First U.S. coal-gas company incorporated	Hull and Kohloff, 1952
1816	Coal-gas plant installed in Baltimore, MD	Rhodes, 1966a; Morgan, 1926
1816	First public display of gas lighting in Baltimore, MD	Tunis, 1933
1820	First coal-tar distillation plant started in England	Rhodes, 1966a
1822	Coal-gas plant installed in Boston, MA	Rhodes, 1966a; Morgan, 1926
1825	Coal-gas plant installed in New York, NY	Rhodes, 1966a; Morgan, 1926

(continued)

TABLE 45 (continued)

Year	Event	Reference[a]
1829	Water-sealed gas holder used in England; masonry tanks were used to hold the water	Alrich, 1934
1838	First timber treated with coal tar in England	Rhodes, 1966a
1838	Heavy oil (creosote) first used to preserve wood from decay and marine worms	Stover and Chung, 1979
1847	First benzene recovered from coal tar in England	Rhodes, 1966a
1849	Iron oxide process for H_2S removal patented	Powell, 1945a and 1945b
Around 1850	Horizontal firebrick retorts were commonly used for coal-gas production	Rhodes, 1966a
Before 1850	Cast iron retorts used for coal-gas manufacture, 600-800 °C	Rhodes, 1966a
1850	Clay retorts used for coal-gas production instead of cast iron	Morgan, 1926
1856	Dye from light-oil fraction of coal tar discovered; analine dyes follow this discovery	Stover and Chung, 1979
1856	First coke ovens with byproduct recovery installed in France	Morgan, 1926
1857	Dye manufactured from coal-tar products in England	Rhodes, 1966a
1860	British "Sulfur Act of 1860" limited sulfur in gas to 22 grains per 100 cubic feet	Powell, 1945a and 1945b
1859- 1900	Air-cooled condensers used to cool manufactured gas	Downing, 1934

(continued)

TABLE 45 (continued)

Year	Event	Reference[a]
Early 1860's	First U.S. coal tar distilled in Boston, MA	Lane, 1921
1861	Three-lift holder tank introduced in England	Alrich, 1934
1865	Phenol recovered from coal-gas liquids for antiseptic purposes in England	Rhodes, 1966a
1869	Dyes manufactured from coal tar	Stover and Chung, 1979
1870	Fontana identified Blue Gas by passing steam over incandescent carbon	Morgan, 1945
1870	Water gas (blue gas) discovered; 330 Btu/ft^3, very poor luminosity	Rhodes, 1966b; Morgan, 1926
1870	Iron oxide purification introduced to U.S.	Powell, 1945a and 1945b
1872	T.S.C. Lone invents carbureted water gas; it has higher heating value and luminosity than does coal gas	Rhodes, 1966b; Morgan, 1926
1876	First iron gas holder tank installed in U.S.	Alrich, 1934
1877	Antiseptic and deodorizing solutions produced from tar-acid oils in England	Rhodes, 1966a
1880	Indigo produced from coal tar	Stover and Chung, 1979
1882	A considerable percentage of the gas output of the country was carbureted water gas	Morgan, 1926
1884	Use of down stream for carbureted water-gas production introduced	Morgan, 1926

(continued)

TABLE 45 (continued)

Year	Event	Reference[a]
Before 1885	Lime used as purifying agent to remove CO_2, H_2S, and organic sulfur	Downing, 1934
1886	Mantles introduced for gas lighting	Forstall, 1934
1887	First U.S. tar distillation plant installed in Philadelphia, PA	Rhodes, 1966a
1888	First steel gas holder tank installed in U.S.	Alrich, 1934
1889	L.P. Lowe patents oil-gas process in the U.S.	Rhodes, 1966b; Morgan, 1926; Hull and Kohloff, 1952
1885 - 1890	Development of rusted iron borings (iron oxide) process for H_2S removal	Downing, 1934
1892	First U.S. byproduct coke oven installed in Syracuse, NY (10 years after England and Germany)	Rhodes, 1966a; Morgan, 1926
1894	First three-lift holder tank installed in U.S.	Alrich, 1934
1894	Byproduct coke plant erected in Johnstown, PA	Lane, 1921
1900	Pacific Coast oil-gas process developed	Hull and Kohlhoff, 1952
Before 1900	Tar removal by bubbling gas through strong ammonia solution (Livisey washer)	Downing, 1934
After 1900	Water-cooled condensers used to cool manufactured gas	Downing, 1934

(continued)

TABLE 45 (continued)

Year	Event	Reference[a]
Before 1900	Luminous flame burners used for lighting	Forstall, 1934
Around 1900	Vertical retorts used to produce coal gas	Morgan, 1934
Early 1900's	Light-oil recovery scrubbers introduced	Downing, 1934
Early 1900's	Direct-contact washer-cooled with P and A tar extractor introduced for tar removal	Downing, 1934
1901	Steel gas holder tanks preferred to brick holder tanks; steel tanks were now cheaper	Alrich, 1934
1902	First use of crude oil in a carbureted water-gas plant in California	Morgan, 1926
1902	First oil-gas plant installed in Oakland, CA	Morgan, 1926
1902	First oil-gas plant in U.S. installed in Oakland, CA; uses the Pacific Coast oil-gas process	Rhodes, 1966b
1903	Carbureted water-gas industry begins change from paraffinic-based oils to asphaltic-based oils	Fischer, 1933
1905	Lime scrubbing replaced by Iron Oxide Purification in Britian	Powell, 1945
1907	Centrifuges introduced for separation of emulsions	Fischer, 1933
1907	Washer-cooler introduced; contacted gas directly with recirculated condensate from gas	Downing, 1934
1910	Turbo exhauster; used to increase the pressure of manufactured gas flowing to scrubbers	Downing, 1934
1910	Aluminia from bauxite used for H_2S removal; this process was not used very much	Downing, 1934
1910	First use of water-gas tar to preserve railraod ties; tar mixed with ZnCl prior to wood treatment	Fulweiler, 1921

(continued)

TABLE 45 (continued)

Year	Event	Reference[a]
1912	Refiners start cracking petroleum oils to increase the production of gasoline	Rhodes, 1966b
1915	World War I spurs development of tar recovery and use in the U.S.; demand for tar products increases	Rhodes, 1966a
1916	Water purification process using lime and copperas ($FeSO_4$) followed by coke filter described	Hansen, 1916
1916	Dry-gas holders introduced	Alrich, 1934
1919-1920	Production and prices of coal-tar chemicals dropped after World War I	Rhodes, 1966a
1920	Out of the 917 gas plants in the U.S., 596 of them are carbureted water gas	Rhodes, 1966b
1920-1929	Growing use of phenolic and alkyd resins promotes the recovery of naphthalene and phenol	Rhodes, 1966a
1921	Seaboard process for H_2S removal introduced	Denig and Powell, 1933
1921	Seaboard liquid process for H_2S and HCN removal developed by the Koppers Co.	Sperr, 1923
1925	Nickel process for H_2S removal and sulfur recovery invented	Cundall, 1927
1929-1932	Great Depression cuts deeply into prices and production of tar-based chemicals	Rhodes, 1966a
1929-1932	Horizontal and vertical retorts abandoned or replaced by oil gas, water gas, or natural gas	Rhodes, 1966a
Around 1930	Use of heavy fuel oils for oil and carbureted water gas begins	Rhodes, 1966a
1930	High surface area iron oxide sponges introduced; they had double the S removal of homemade FeO	Downing, 1934

(continued)

TABLE 45 (continued)

Year	Event	Reference[a]
Early 1930's	Electrostatic precipitation for tar removal introduced	Downing, 1934
Early 1930's	Tetralin (tetrahydronaphthalene) used to remove naphthalenes from gas	Downing, 1934
1932-1945	World War II greatly increased demand and production of tar-based chemicals	Rhodes, 1966a
1933	Seaboard H_2S removal process installed at 30 plants	Denig and Powell, 1933
1938	Catalytic cracking of crude-oil residuals by refineries produces high yields of gasoline and gas oil	Pew, 1940
1949	Federal Power Commission allows certain pipelines that previously transported oil to carry natural gas	Rhodes, 1966b

Land area for the production of gas was generally more available for the U.S. plants. There was more area for onsite disposal of waste products and less need to use underground structures for storage (and placing other structures directly over underground structures).

British town gas sites closed when North Sea natural gas became available (1967 through 1974). U.S. plants had closed much earlier when pipeline natural gas from western fields became available (1945 through 1955). Because the U.K. plants closed later, during a period of increased environmental consciousness, they were generally better decommissioned than were the U.S. plants.

Britain, a relatively small country, was more homogenous in the production techniques and purification processes employed. In the United States, different production processes were employed in various areas of the country to take advantage of local resources and markets. Markets for byproducts were frequently more accessible in Great Britain than they were in the United States. This meant that the recovery of byproducts was practiced more extensively in the United Kingdom than it was in the United States. Products discarded for economic reasons in the United States would frequently be recovered in the United Kingdom.

Sale and recovery of sulfur from spent oxide was practiced (and profitable) in Great Britain. Spent oxide was viewed as a usable byproduct from the manufacture of gas. The sale and recovery of spent oxide was employed at very few U.S. plants, and spent oxide was universally viewed as a waste for disposal. Because spent oxide was utilized in Great Britain, gas plants disposed less of it and had much less incentive to switch to liquid purification processes for H_2S removal. The quantities of spent oxide wastes disposed in the United States were consequently a larger percentage of the spent oxides produced than were those disposed in the United Kingdom.

Tars and oils recovered from town gas production were more valuable in Great Britain than they were in the United States (due to higher petroleum prices in Great Britain). Disposal of tars and oils was much less likely in Great Britain than it was in the United States. Because coal tar was generally regarded as more valuable than CWG tars or oil tars, more of the tars produced in the United Kingdom would have been recovered.

The United States was much slower than was Great Britain in distilling coal tar and recovering coal-tar byproducts. The United States did not start recovering coal-tar chemicals on a large scale until World War I. This was due in part to the importation of coal-tar chemicals from Germany and Europe and also to the use of CWG in the United States. Because CWG tars did not contain many of the most valuable chemicals in coal tar (e.g., anthracene, used in the production of dyes), there was less incentive to process the tars for recovery.

1.7 CONCLUSIONS FROM THE HISTORICAL REVIEW

Three major processes were used for the production of town gas in the United States. These were (1) coal carbonization, (2) carbureted water gas (CWG), and (3) oil gas. In general, all three processes were employed in all areas of the United States, but each process became predominant in specific geographical areas in the United States. Gas plants along the West Coast started as coal-gas plants, switched to CWG, then converted to oil-gas production. Plants along the East Coast were generally CWG, with some coal-gas production, and coal-gas production was predominant in the Middle States. Because the gas purification processes, byproducts, and wastes from the gas production varied with each production method, it is important to understand the specific production methods and associated byproduct recovery operations of individual gas sites.

The feedstocks used in gas production changed during the operation of gas plants. The coal used for coal carbonization did not change substantially over time, but the carbon and hydrocarbons used for CWG production and oil-gas production changed substantially over time, which had a significant effect on the wastes produced. CWG production originally used coke or anthracite coal in the generator and low-boiling naphtha fractions as hydrocarbon feedstock. Later, bituminous coal often was used directly in the generator, and the hydrocarbon feed was switched first to gas-oil fractions, and later to heavy fuel oils and residual oils. Oil gas originally utilized either gas-oil fractions of petroleum or crude oil, but later switched to heavier fuel oils and residual oils. The choice of feedstocks was determined by the prevalent economics of the oil industry during the production of town gas. The conversion from lower-boiling petroleum fractions (naphtha and gas oil) to heavier oils

(fuel oil and residual oil) was accompanied by increases in the tars produced by the processes and the increased formation of tar-water emulsions. For oil-gas production, the amount of lampblack produced per 10^6 ft^3 gas manufactured increased with the conversion to feedstocks with higher carbon contents. The emulsions that formed were often difficult to separate, and they were often discarded when separation attempts failed.

Coal carbonization produced a fuel gas containing substantial amounts of ammonia, cyanide, phenolic compounds, and hydrogen sulfide. The presence of these chemicals determined the cleanup processes for their removal from the gas and any recovery processes. They also appeared in the wastes from coal carbonization. In contrast, both CWG and oil gas contained only small amounts of nitrogen compounds (ammonia and cyanide) and only trace quantities of phenols. All three processes produced gas containing hydrogen sulfide. Ammonia and phenol were not produced, removed, or recovered from CWG and oil gas, but they were from coal-carbonization gases. This relatively simple correlation explains much of the variation seen currently at sites. The absence of phenols in tars from Stroudsburg, Pennsylvania (oil and CWG), and Ames, Iowa (CWG), are two more prominent examples. Iron oxide was used almost universally to remove hydrogen sulfide from town gases. The iron oxide also reacted with hydrogen cyanide in the gas to produce blue iron cyanide complexes. These ferriferrocyanides are relatively stable, and they persist at gas sites that produced coal gas and disposed spent oxides onsite (an almost universal practice). They are the most visible waste at plants that produced coal gas, but they re absent from plants that produced only oil gas or CWG.

The removal of hydrogen sulfide was required for all three gas production processes, with the amount of hydrogen sulfide removal required being dependent on the coal sulfur concentration for coal-carbonization gases or the sulfur concentration in oil for oil gas and CWG. Between 1816 and 1855, lime was used for the removal of hydrogen sulfide and other impurities from town gas. Lime use was characterized by low conversion of the lime to CaS, difficult disposal problems, and high cost. The use of lime was essentially replaced by iron oxide purification after 1890. Both the lime and spent iron oxide were considered wastes; although there were many attempts to use them

for some productive purpose, they were universally disposed. Lime use occurred primarily during a period when the cost of town gas was very high, and it was used principally to light only streets and shops in cities. With the introduction of iron oxide purification, gas prices dropped and gas became a larger consumer item. Spent lime wastes were not a significant problem at most U.S. sites because of the low gas production rates during the time that lime was used. Spent lime was also used for agricultural purposes, which reduced the amounts of spent lime that had to be discarded. Because lime was also used in the recovery of ammonia from coal gas, spent lime sludges from ammonia recovery are possible at most coal-gas plants that recovered ammonia (but it would be present in much smaller quantities than if used for hydrogen sulfide removal). Spent iron oxides, however, are the predominant waste from the removal of hydrogen sulfide.

Spent iron oxides were universally regarded as wastes, and they were often used as a general fill material around gas plants. They constitute a major discarded waste that can be located on most sites. Unfortunately, there is wide variation in the composition of spent oxide wastes, which hinders characterization efforts. Organic hydrocarbon content, sulfur content, cyanide content, and mixtures with woodchips are all variables affecting the current composition of spent oxide wastes.

Alternatives to the use of iron oxide for hydrogen sulfide removal were introduced after 1921. The Seaboard process used a solution of sodium carbonate to scrub hydrogen sulfide from the gas. Solutions were regenerated by blowing air through the scrubbing liquid, rereleasing the hydrogen sulfide to the atmosphere. A process using a solution of arsenic salts to remove hydrogen sulfide and recover it as a sulfur was introduced around 1925. This process would be accompanied by possible arsenic contamination of sites, especially if spent solutions were disposed. This process was frequently used upstream of iron oxide beds (the arsenic process would remove most of the hydrogen sulfide, and the iron oxide would reduce the hydrogen sulfide content of the gas to very low concentrations). The spent oxide waste from this type of operation would have potential arsenic contamination resulting from carryover of the scrubber solution.

The composition and characteristics of coal- and water-gas tars varied substantially among plants. Water-gas tars and oil-gas tars tend to be very

similar in composition and properties because both are essentially produced by the thermal cracking of petroleum fractions. They tend to be less viscous than are coal gas tars, and they contain only trace amounts of phenolic and base nitrogen compounds.

The formation of tar-water emulsions was a major problem of the industry, and it frequently resulted in the disposal of these oily materials when the emulsions could not be broken. Water and tar are condensed simultaneously in the purification of town gas. The resulting mixture of tar, oils, and water would usually separate into layers, and the tar and oil could be recovered. When emulsions formed, the tar would not separate from the water, and the gravity separators frequently used for the separation would not function. Emulsions were rarely formed from production of coal gas, but were a frequent problem for both carbureted water-gas production and oil-gas production. Emulsions could generally be separated by mechanical and thermal methods, but occasionally emulsions would form that defied all attempts at separation. These emulsions were disposed by any means available, including the use of open, unlined lagoons, direct discharge to bodies of water (where feasible), or into any convenient unused well. Lagoons were frequently used for storage of emulsions. This allowed additional time for the emulsions to separate by gravity or for alternative batch methods of separation to be used. The plant at Plattsbugh, New York, utilized lagoons for the storage and disposal of tar-water emulsions.

The formation of emulsions became more prevalent when oil and CWG producers switched from lower-boiling petroleum fractions to heavier and higher carbon-content residual oils.

Tars and oils were generally recovered from the production of town gases. Although early plants disposed essentially all of their tars and waste condensates (usually to the nearest body of water), they rapidly discovered that this waste was worth recovering. Coal tars could be separated by gravity from the condensate and oils. These tars could then be either burned (as fuel in the retorts or boilers), refined and sold, or sold as a raw byproduct. Water-gas tars were recovered and sold as a liquid fuel, burned in the plant's own steam boiler, or recycled back into the hydrocarbons used for cracking into the gas. All tars had a minimum value to the plant as fuel because the tars

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could replace a portion of the coal that would normally be burned at the plant.

Several specific practices contributed to the contamination of gas production sites by tars and oils. Many of the original gas holders for plants were partly buried below ground and frequently filled with ccal tar. They were usually not well sealed at the base, and some of the tar contained in them leaked into the ground. Tar wells (tar storage tanks) and tar separators were frequently constructed underground of masonry or cement, and they often leaked. Some storage tanks were constructed of wood. Wastes were usually disposed either at the plant site or adjacent to the plant. These practices indicate that any former gas site will probably have some tar and oil contamination, with the extent of contamination being dependent on the specific practices of the plant.

Most of the byproducts from town gas production could be considered either products or wastes, depending on the prevailing price that could be obtained for the byproduct. Spent iron oxide was always considered a waste, in spite of continuing attempts to develop uses for the material. Recovered tars could be sold, but they had a minimum fuel value that determined their value as a fuel. Plant size and access to markets were two of the primary factors that influenced the waste disposal practices of gas production plants. Smaller plants did not have the same economy of scale as did the larger plants, and frequently they did not recover materials that the larger plants recovered extensively. This was particularly true of small water-gas and oil plants, which sometimes let the tars and condensates flow to waste rather than attempt to recover any of the tar. Transportation costs of shipping tars or ammonia liquors to appropriate markets frequently prevented the sale of byproducts that might have been worth recovering.

There is a substantial tendency to apply the work done in the United Kingdon with old town gas sites to U.S. plants. There are, however, several substantial differences between plants in these two countries. First, the United States had abundant petroleum resources, which made the use of CWG and oil gas practical. The United Kingdom had only limited petroleum resources and produced coal gas almost entirely. Coal tars and tar products also commanded a higher price in the United Kingdom than they did in the United

States, thereby encouraging United Kingdom plants to recover these byproducts. The market for spent oxides in the United Kingdom was well developed (it was used for the manufacture of sulfuric acid); low sulfur prices in the United States prevented the development of any markets for spent oxides. Similarly, liquid-scrubbing methods for the removal of hydrogen sulfide from gas were developed in the United States, but the United Kingdom plants continued to use iron oxides because they could market the spent oxides. Gas plants in the United Kingdom also were generally placed on smaller sites than were those in the United States. Consequently, wastes from U.K. plants would be more likely to be hauled away to disposal sites, rather than discarded onsite.

After the first natural gas pipelines were installed in an area formerly served by manufactured gas, the natural gas was generally used to meet baseline demand, and the manufactured-gas plant was modified to produce gas for mixing with the natural gas to meet peak demands. As larger pipelines were installed for natural gas delivery and better storage methods for natural gas became available, the need for a standby gas production facility evaporated. The manufacturing plants were generally idle for several years before they were decommissioned. The most frequent reason for decommissioning the plants was to remove structures from the site and reduce the site valuation for tax purposes. The purpose of site decommissioning was to remove surface structures from the site. Gas storage tanks were cut off at ground level, and the tanks were filled with debris from the plant site. Underground tanks and structures were rarely removed, and some tanks and tar separators were left filled with tar or liquid wastes. Many gas companies still own the original sites used for the manufacture of gas, in that it is generally much cheaper to keep the site as unused land than it would be to clean the site for sale.

During the literature review, RTI discovered that the literature describing the operations of gas plants is very substantial. This is not surprising in that the manufacture of town gas was once a large industry. Several references were discovered that deal specifically with the waste disposal practices and problems of the U.S. industry. These articles indicate that groundwater contamination in areas around gas sites was common while the plants were in operation and that contamination of downstream water supplies was also a common problem.
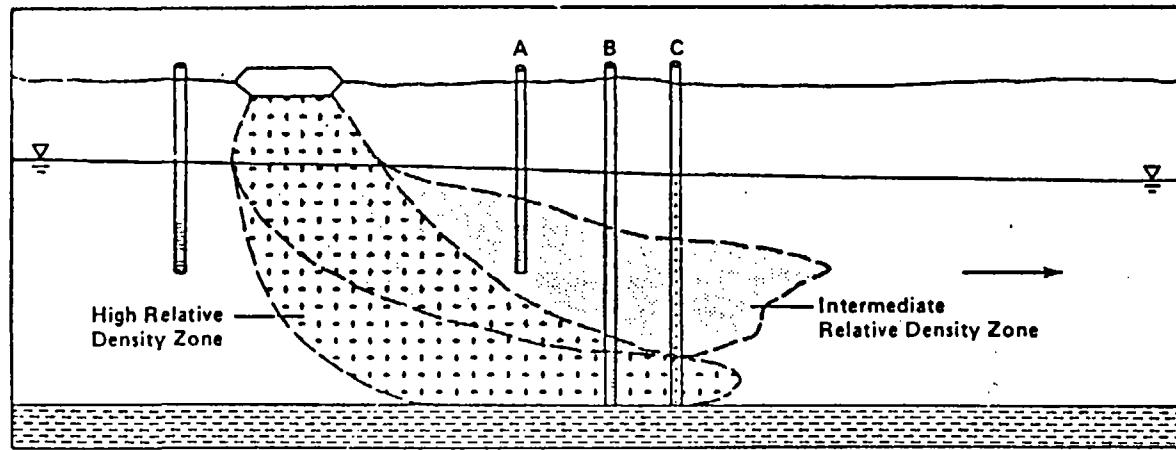
Source: Alexander, 1984.

Figure 64. Groundwater monitoring of multiple-density components originating from single source area.
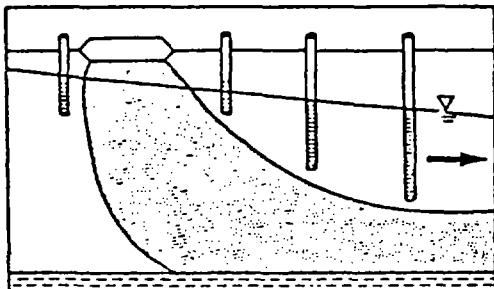
Inappropriate Monitoring   Appropriate Monitoring
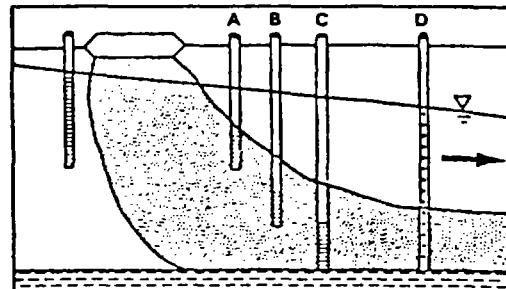
Example 1. High relative density contaminant solution.
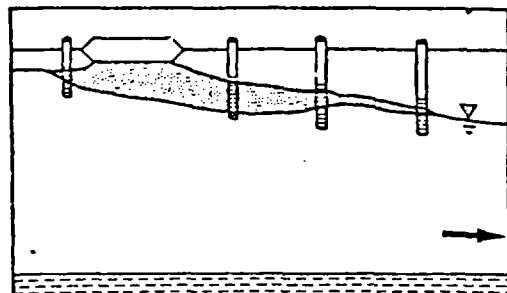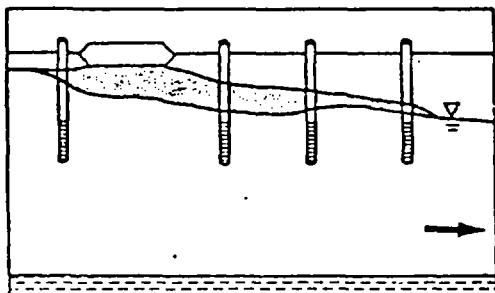
Example 2. Low relative density immiscible contaminant.
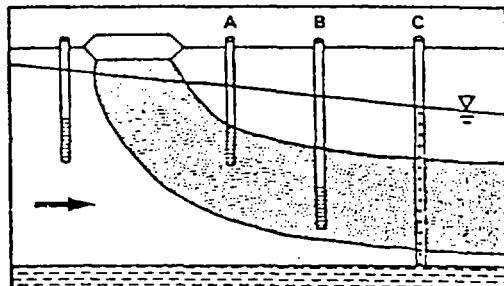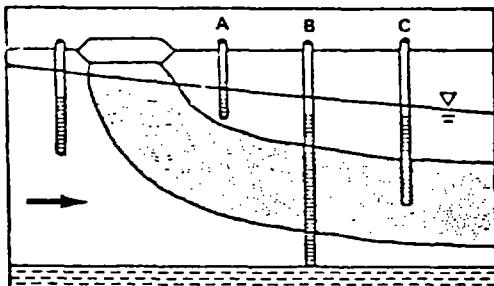
Example 3. Contaminant solution with similar relative density as ground water.
Source: Alexander, 1984.

Figure 65. Comparative groundwater monitoring of variable density
contaminants in uniform flow field.

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contaminant plume, but it also draws in uncontaminated groundwater, as does well C. The results of groundwater analysis from these wells may not reveal the presence of contamination because of dilution of the samples. More appropriately, the carefully screened intervals of wells A and B would detect the contamination, but that would require prior knowledge of the plume's vertical extent. This knowledge could be gained by the installation of a multilevel sampler (well C).

The presence of high-density tars at gas sites in the subsurface requires special care when constructing monitoring wells into deeper aquifers below a site. These wells can provide pathways for such tars to move, under density gradients, into deeper aquifers, even against an upward hydraulic gradient between the confined aquifer and the surface. At St. Louis Park, Minnesota, coal tar flowed down a multiaquifer well, resulting in contamination of multiple aquifers (see Chapter 3). Because of this contamination potential, monitoring wells for aquifers beneath a gasworks site should not pass through zones of tar contamination. If tar is encountered during the construction of such a well, the well should be moved to an area with no underlying coal tar. If this is not possible, extreme care should be taken to seal off the tar-containing zone to prevent migration of tar into the borehole and down into the aquifer.

2.3 SITE REMEDIATION

2.3.1 Introduction

Remediation options for gasworks sites are basically the same as those for other industrial hazardous waste sites: no action; onsite containment, with or without stabilization or fixation; removal and disposal of contaminated material; in-situ treatment; removal and treatment or destruction of contaminated materials. The selection and implementation of remedial alternatives for specific gasworks sites are the same as for other hazardous waste sites. This discussion does not go into detail about site remediation. Instead, it concentrates on the unique features of gasworks sites that may affect site remediation, case studies of actual gasworks site remediation, and listing remedial action alternatives for specific gasworks wastes. For more information on the selection and evaluation of remedial action alternatives

for specific sites, the reader is referred to Cochran and Hodge (1985a, 1985b), Boutwell et al. (1985), U.S. EPA (1982), Ehrenfeld and Bass (1983), and Sims et al. (1984).

2.3.2 Factors Affecting Site Remediation

Gasworks sites have certain unique features that can influence the selection of remedial alternatives. First, the sites are old: Many were abandoned more than 50 years ago, and almost all are more than 30 years old. This age can affect remediation in several ways. It can result in a low-priority ranking for the site in terms of cleanup. If the site owner can demonstrate that there is no history of contaminant migration and that wastes currently are remaining onsite, it is possible that site remediation efforts could be postponed without damage to human health or the environment. The fact that a site has existed for decades without problems may be taken as evidence that postponing remediation will cause no further problems. If cleanup is postponed, however, groundwater monitoring should be employed to detect contaminant release, and measures such as restricted site access should be taken to avoid exposure of the public to contaminants at the site.

On the other hand, the age of these sites can afford a long period of time for contaminants to move offsite, thereby resulting in a significant spreading of contaminants and an increase in the volume of material that must be cleaned up. This was the case at Brattleboro, Vermont, where coal tar has moved through a porous gravel layer along a bedrock surface, underneath a river adjacent to the former plant site. At St. Louis Park, Minnesota, where a coal tar refinery operated for more than 50 years, contaminants have spread to several aquifers to a depth of over 900 feet, and a plume of contaminants extends over one-half mile from the site. At Ames, Iowa, lighter tar constituents from a gas plant closed in the 1930's have contaminated the municipal well field, resulting in the closure of five municipal wells since contamination was first detected in 1927. In contrast, at Stroudsburg, Pennsylvania (Brodhead Creek), favorable geological conditions resulted in the containment of over 8,000 gallons of free coal tar in the subsurface for about 40 years, until excavation of the adjacent creek bank caused release of the tar into the creek.

Many of the case studies reviewed in this study (see Chapter 3) illustrate the fact that gasworks were often built in low-lying areas adjacent to waterbodies or wetlands. In some cases, discharges into these waterbodies resulted in a site discovery. Proximity to waterbodies or wetlands can require barrier construction to prevent surface water contamination during site remediation. In addition, contaminants may have been disposed of or migrated into these waterbodies, which can result in accumulation in river or lake sediments. This could necessitate underwater cleanup operation, complicating and increasing the cost of site remediation.

Gasworks also usually occur in downtown areas or old industrial districts. The recent trend to redevelop these areas has resulted in the discovery of many former gasworks sites across the country. Redevelopment pressures and priorities can affect site remediation efforts and vice versa. The following cases illustrate how redevelopment and remediation were handled in different areas of the country.

In Newport, Rhode Island, two multimillion dollar apartment buildings were being constructed across the street from one another when tar from a former gas plant was discovered in the subsurface at both construction sites. One building was being constructed on pilings. The only contaminated material removed from this site was that actually excavated for the pilings. It was disposed offsite, and the lower floors of the building were designated for nonresidential use (parking garage). At the other site, a buried concrete structure was discovered and accidentally ruptured during construction of the foundation. It was full of coal tar. In this case, the structure was repaired, the coal tar left in place, and a ventilation system installed to prevent organic vapors from accumulating in the basement of the apartment building.

In San Francisco, California, coal-tar contamination was encountered during construction of an addition to EPA's Region 9 headquarters. This material was removed and disposed in a secure landfill. There was suspicion that the soil under the existing building also could be contaminated, but this has not been verified.

Cases of contamination discovery under existing buildings constructed after a gas plant was removed were not uncovered in this study. However, the

downtown location of most plants makes the existence of such a situation possible, if not probable. The presence of an existing building over a contaminated gasworks site would be considerably complicated and could prevent remediation of a site. In such cases, onsite containment may be the best option.

Case studies in Chapter 3 that illustrate the interaction of redevelopment and site remediation are GasWorks Park, Seattle, Washington; Brattleboro, Vermont; Plattsburgh, New York; Everett, Massachusetts; and Mendon Rd., Attleboro, Massachusetts.

When gasworks were decommissioned, surface structures often were removed, but structures below the surface usually were left in place. These structures often contain contaminants, usually tars, oils, or tar/water emulsions. Because of this, it is important to determine the locations of these structures during a site investigation and to consider their locations when planning site remediation activities. In some cases, free tars and oils occur in these structures; such gasification byproducts may be reused as supplementary boiler fuel or chemical feedstocks. If reuse is not a viable alternative, careful recovery of the material from the structures results in a more concentrated waste stream for treatment or disposal. If subsurface structures are damaged during remediation efforts, contamination can spread into surrounding soils, increasing the expense and complexity of remediation efforts.

Another feature of gasworks sites that can affect remediation efforts is the presence of injection wells that were used for waste disposal (e.g., for tar residues and emulsions). At least one site reviewed in this study, Stroudsburg, Pennsylvania, may have had one of these wells. Research by the Stroudsburg site investigators suggested that other gasworks in the area may have used wells for waste disposal. Maps for the Lowell, Massachusetts, plant showed a "deep well" on the site. However, it is not clear whether this well was used for waste disposal. Additionally, it is important when reviewing old site maps not to confuse tar wells, which are underground structures containing tar, with injection wells used for disposing of wastes.

The location and depth of all wells on a site should be determined during remedial investigations. These wells may be reopened and sampled for contamination. Care should be taken during reopening to prevent them from adding to the spread of contaminants. If no contamination is detected, they should be

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properly closed and sealed to prevent them from becoming pathways for contaminant migration. If contaminated, they can complicate site remediation efforts. However, if wastes were pumped down a well, it may be possible to pump them back out. This was accomplished at Stroudsburg, where over 8,000 gallons of free coal tar was removed from the subsurface. However, considerable tar remains bound up in subsurface material at Stroudsburg; this necessitated containment (slurry wall) to prevent migration of contaminants offsite.

2.3.3 Remedial Action Alternatives

2.3.3.1 Introduction--

As previously stated, remedial action alternatives for gasworks sites are similar to those for other uncontrolled hazardous waste sites. Containment, removal and disposal, and treatment all are applicable. Some containment generally will be required for all remedial actions to prevent the release and spread of contaminants. Slurry walls and caps have been used to contain gasworks wastes. Removal and disposal is a simple but expensive option that also has been used to clean up gasworks sites. Treatment to stabilize, detoxify, or destroy gasworks wastes has not been employed to a great extent, but it is attractive because it can destroy a waste's hazardous nature, enabling safe disposal of residues in nonhazardous waste landfills and eliminating future liability. Treatment alternatives with potential applicability to gasification wastes are summarized in Table 47.

The following discussion focuses on remediation techniques actually applied to gasification wastes or similar substances. For more general information on the evaluation and selection of remedial action alternatives, the reader is referred to the references listed at the end of Section 2.3.1.

2.3.3.2 Oils, Tars, and Lampblack--

The most prevalent and persistent contaminants at gasworks sites are organic byproducts of the gas manufacturing process--tars, oils, and lampblack. Tars and oils could be produced in any process; lampblack was most commonly produced in oil-gas processes. Tars and oils can contaminate soils and groundwater (see following sections), but they also occur as free products at gasworks sites, especially in buried tanks and other structures, buried lagoons, and in coarse sands and gravel in the subsurface. Lampblack may

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TABLE 47. POTENTIAL TREATMENT TECHNOLOGIES FOR CONTAMINATED SOILS

Method	Description	Applicability
In-situ methods		
Neutralization	Addition of base to soil to neutralize acid; base could be spread or injected into soil as a solution or spread as a powder and tilled into soil	Acids or acid-forming wastes
Solvent extraction	Flush with chemical solution to remove contaminants, then collect and treat solvent; solvent could be acidic, basic, or surfactant, injected or percolated into soil and collected in drain or withdrawal wells	Organics or metals, depending on solvent
Chemical oxidation	Addition of chemicals such as ozone or peroxide to break down compounds into harmless forms or forms more readily attenuated by natural microbial activity; lack of selectivity may lead to high dosage requirements	Primarily organics, may mobilize metals, requiring leachate collection and treatment
Immobilization	Reduces rate of release of contaminants into environment; pH adjustment or chemical addition promotes sorption or precipitation onto organic materials such as sawdust or agricultural byproducts; may have already occurred at gas manufacturing sites through reaction with organic "fluff"	Metals and organics
Attenuation	Mixing of contaminated soils with clean soil, municipal refuse, or sewage sludge; may be acceptable for low-risk wastes, also may promote natural biological degradation	Compatible wastes of low mobility and toxicity

(continued)

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TABLE 47 (continued)

Method	Description	Applicability
Biological oxidation	Occurs naturally in soils; may be enhanced by addition of nutrients, oxygen, or specially developed microbes; contaminants are metabolized by bacteria and/or fungi to harmless forms	Primarily organics although sulfur and nitrogen also may be oxidized

Methods requiring excavation

Thermal treatment

Evaporation	Contaminated soils heated to drive off volatiles that are destroyed in an afterburner	Organics, cyanides, sulfides; auxiliary fuel required
Incineration	Entire waste matrix heated to over 1,000 °C to destroy contaminants	Same as evaporation
Chemical treatment	Neutralization, extraction, oxidation, immobilization similar to description under in-situ methods, carried out in a reactor under controlled conditions	Various wastes

Biological treatment

Landfarming	Waste incorporated into upper layers of soil, biological degradation stimulated, cover or livestock feed crops grown	Organics, cyanides; not suitable for wastes containing heavy metals, which may build up in soil or crops
Composting	Waste biologically stabilized above ground, may be mixed with municipal refuse or sewage sludge; result may be used as a soil amendment	Primarily organics or cyanides; disposal of metals depends on final disposition of product

SOURCES: Sims et al., 1984; Hoogendoorn, 1984.

occur at or below the surface. If these contaminants can be recovered in pure form, they may be reused as supplementary fuels or chemical feedstocks. Alternatively, they are easily incinerated.

Removal of tars or oils from underground containment structures is relatively straightforward, but care must be exercised to avoid rupturing the structure. Once the substances have been removed from tanks, the tanks can be either cleaned using steam or aqueous surfactants or removed and disposed of.

At Stroudsburg, 8,000 gallons of free tar was pumped from the ground using techniques developed for control of distribution of tar in the surface (Villaume et al., 1983; Roberts et al., 1982). Poor understanding of these phenomena at Stroudsburg resulted in overestimation of free coal tar in the subsurface and overscaling of the coal tar recovery system. Original estimates of 35,000 gallons of free tar resulted from a failure to realize that the tar was present in several different "phases" or zones. Much of the tar was held up in the subsurface by capillary forces so that no coal tar could be removed by pumping, or it was associated with water in a fashion that would result in recovery of coal tar and water if this zone was for heavy oil recovery (see case study in Chapter 3). In this case, recovery by pumping was possible because the tar was contained in a coarse, highly permeable aquifer that enabled it to move relatively freely. The feasibility of this approach at other sites may be determined from the characteristics of the porous medium (e.g., porosity and permeability), the characteristics of the tar (e.g., viscosity, density, interfacial tension between tar and water, and wetting angle of tar on aquifer material in the presence of water), and an awareness of how viscous and capillary pressure forces can be pumped. Figure 66 illustrates the zoned distribution of water and coal tar in the subsurface at Stroudsburg, inferred from capillary pressure theory, and it indicates the types of material that may be pumped from the different zones. Failure to perform this sort of analysis can result in overestimation of the amount of free tar in the subsurface; tar in water emulsions and tar held by capillary forces in the subsurface material may be included in the free coal tar estimates.

As previously mentioned, free products recovered from gasworks sites may be used as fuel (as at Stroundsburg) or as chemical feedstocks. It also may

PUMP PRODUCT	LIQUID PHASE(S)	POROUS MEDIUM
WATER	GROUND WATER WITH DISSOLVED ORGANICS	SAND AND GRAVEL
WATER	GROUND WATER AND TRAPPED COAL TAR	
WATER AND COAL TAR	GROUND WATER AND COAL TAR	
COAL TAR	COAL TAR AND "IRREDUCIBLE" WATER	
WATER	GROUND WATER WITH DISSOLVED ORGANICS	SILTY SAND

Source: Villuame et al., 1983.

Figure 66. Ideal distribution of coal tar in porous materials at the Stroudsburg contamination site, as inferred from capillary pressure theory.

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be easily incinerated (heating value ~17,736 Btu/lb) or may be amenable to land treatment, as described in the following section on contaminated soil.

Lampblack is solid, sooty material that was commonly produced in oil-gas plants. It is composed of very heavy organic compounds, including PAH. It is essentially immobile and insoluble in the subsurface. Because of this, it can be safely contained onsite, as was done at an unnamed site in southern California. If it is removed, it can be briquetted and used as solid fuel or possibly used as a blackening agent in certain industrial processes. Alternatively, it may be easily incinerated. Lampblack contains PAH's and is carcinogenic; its powdery form makes it necessary to exercise care to prevent dust emissions when excavating and handling the material. Inhalation and skin contact also should be avoided.

2.3.3.3 Spent Oxide Wastes--

Spent oxide wastes, as described in Section 2.1.2, are extremely heterogeneous in nature from site to site and within specific sites. This variability occurs both in terms of the wastes' physical characteristics and types of contaminants that may be present. Because of this variability, and because they have not been extensively characterized by composition or occurrence, it is difficult to evaluate remedial alternatives for these wastes. This discussion concentrates on the characteristics of the wastes that can affect their treatment and handling during remedial actions and on two cases in which sites containing spent oxide wastes were remediated.

Spent oxide wastes are pyrophoric, i.e., when exposed to air they have a tendency to self-heat and spontaneously combust. For instance, Downing (1932) reports:

> The disposal of spent oxide is a vexatious problem for many gas plants. Because of a possibility of fires starting through heat generated by revivification, it is necessary to hold the spent material at the plant until this danger is past. As soon as city authorities learn of this menace the material is prohibited at public dumps. Continuous storage on gasworks land eventually becomes impossible. The material makes excellent filling for roads or private property when properly handled. It should be covered with ashes or soil immediately to prevent the access of air and consequent combustion.

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This pyrophoric nature probably is due to the presence of reduced sulfur compounds that oxidize exothermically when exposed to air. At one unnamed gasworks site, a gas cleanup box that was left full of oxide material years ago when the plant closed was opened during site cleanup activities. It subsequently caught on fire. In this case, the fire was easy to extinguish because it was contained. However, care should be taken to avoid combustion when excavating, moving, or storing spent oxide wastes at a gas plant site. The material should be covered as much as possible with soil, plastic, or other material to prevent contact with air. In addition, when it is to be stored or transported, it should be carefully placed and compacted into the pile or transportation vehicles to prevent air from permeating the waste materials. Alternatively, it may be possible to separate combustible materials (e.g., woodchips) from the sulfur-containing oxides to prevent combustion of these materials. Physical separation, followed by incineration of the combustible material, may be an appropriate alternative for treating these wastes.

Spent oxides can have elevated levels of arsenic associated with wastes from the Thylox gas cleanup process. They also have significant acid-generating potential, leachates from these wastes having a pH of 1.5. This low pH can result in release of arsenic or other trace metals. At the Birmingham, Alabama, gasworks site, arsenic levels of 8.0 mg/L were reported for 1.5 pH leachate from spent oxide wastes that contained 160 ppm arsenic (Harry Hendon and Associates, Inc., 1982).

Total cyanide levels as high as 8,900 ppm were measured in spent oxides at the Birmingham site. However, the highest levels of free cyanides in water reported at sites contaminated with these spent oxides was 2.6 ppm for a sample with a pH of 1.5 (Harry Hendon and Associates, Inc., 1982); free cyanide levels less than 1 ppm were more commonly associated with spent oxide wastes at Birmingham. This is because most of the cyanides are present as complex iron cyanides. These compounds are very stable in the environment and have a low toxicity. They do appear to release small concentrations of free cyanides; however, these concentrations are well below the 200 ppm level that limits degradation of free cyanides in aerobic soils, and most are below the 2 ppm limit for the anaerobic degradation of free cyanide (Fuller, 1984). The persistence of complexed ferric ferrocyanides remaining for decades in spent

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oxide wastes disposed at or near the surface is further evidence of their stability in the soil environment.

The persistence of the cyanide materials in spent oxides and the relative stability of ferric ferrocyanide compounds is an encouraging observation from the standpoint of treating these wastes. Although the complete destruction of cyanides in spent oxide might be the most ideal solution, the cost associated with destruction options, along with potential for the liberation and release of free cyanide during treatment, may make stabilization or fixation a more desirable choice. The long-term survival of ferric ferrocyanides at gas plant sites, along with the use of this material in table salt, highway deicing salt, paints, pigments, and laundry bluing, suggests that treatments to elimi- nate any hazards under the Resource Conservation and Recovery Act (RCRA), and containment onsite or disposal in a municipal landfill may be an environmen- tally acceptable and cost-effective alternative for dealing with these wastes.

At the Alabama Gas Corporation Gas Works site in Birmingham, Alabama, in-place stabilization was selected as the remedial alternative for an onsite spent oxide disposal area (Harry Hendon and·Associates, 1982). Stabilization of the 2.4-acre site involved excavating and stockpiling the contaminated material, then mixing agricultural lime ($CaCO_3$) and soil in 1-foot lifts across the site, not exceeding 80 tons of lime per acre-foot of soil. In addition to lime, fertilizer and sewage sludge was added to the top 6 inches to promote the growth of vegetation. The lime neutralized the acidic condi- tions formerly present at the site, thereby reducing trace metal (As) release to environmentally safe levels. The remediation plan was successful: The once barren site has been revegetated, and soil samples indicate that acidic conditions and high arsenic concentrations have abated. The cost of remedia- tion was about $100,000; removal, disposal in a secure landfill, and refilling was estimated to cost $2 million to $5 million.

At the Mendon Road site in Attleboro, Massachusetts, 1,083 yd^3 (about one-third of the volume of material at Birmingham) of spent oxide material from gasworks manufacture had been disposed in an abandoned gravel pit. The site was discovered during residential development of the area. The waste was similar to that found at Birmingham (pH = 1.61; total cyanide = 7,500 ppm, free CN^- = 0.7 ppm) except that high arsenic levels were not detected and low

ppm levels of PAH compounds were found. The waste was excavated, removed from the site, and disposed in a secure hazardous waste disposal facility at a cost of over $1.6 million.

The difference in costs in the two spent oxide site remediations is not insignificant. In-place stabilization appears to be a desirable remedial alternative for cyanide-containing spent oxide wastes on both technical and cost bases. If site use plans rule out onsite stabilization as a viable alternative (as at Mendon Road), removal, stabilization, and disposal at a nonhazardous waste landfill may be an environmentally acceptable alternative that is more economical than disposal as a hazardous waste. Studies demonstrating the low mobility in soils of ferrocyanides in municipal waste leachate suggest that stabilization and disposal in municipal landfills may be acceptable (Fuller, 1984). However, more research is needed on the mobility of complex iron cyanides before this can be proven safe. In addition, the extreme variability and heterogeneity of spent oxide wastes necessitate waste-specific evaluations of remedial alternatives. Other methods for treating cyanide-containing wastes are discussed in the following section on remediating contaminated soils.

The characteristic blue color of complex ferric ferrocyanides can be used both to identify areas of spent oxide contamination during site investigations and to guide remediation efforts; however, some question exists as to color-threshold-contaminated levels. At the Mendon Road site, color was used to delineate contaminated soil with greater than 2 ppm total cyanide during cleanup efforts. Wilson and Stevens (1981) report that blue color may be detected in soils containing about 270 ppm total cyanide (or 500 ppm ferric ferrocyanide). Further analyses of samples of soil contaminated with complex iron cyanides is necessary to resolve this discrepancy.

Spent oxide wastes that do not contain complex cyanides are usually red to yellow. They may be more common at U.S. gas plant sites than are cyanide-containing wastes because of the prevalence of water-gas and oil-gas processes that produced gas that characteristically had low levels of cyanide compounds. The major hazards associated with these wastes is their acid-producing potential and their potential to release toxic trace elements. These hazards may be reduced by additives, such as $CaCO_3$, that can reduce acid and limit trace metal release.

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Spent oxide materials may be contaminated with tar and/or may have been codisposed with tar-contaminated shavings from shavings scrubbers used during gas cleanup to tar mist prior to the oxide boxes. At one site visited by the authors, oyster shells contaminated with tar were seen onsite; these probably were used in place of shavings for tar removal. Methods for treating solid materials contaminated with tars and oils are discussed in the following section.

2.3.3.4 Contaminated Soil--

Our review of gas plant site investigations revealed that the most commonly occurring soil contaminants are byproduct tars and oils from gas manufactured. Spent oxide waste containing complex iron cyanides, sulfur compounds, and arsenic is another significant but less prevalent soil contaminant.

Treatment techniques that may have applications at gas plant sites are summarized in Table 46. A complete review of treatment technologies for contaminated soils is beyond the scope of this study. The following discussion considers techniques actually applied on contaminated soil from gasworks plants or on soils contaminated with substances similar to gas plant wastes (i.e., creosote). More information on soil treatment techniques in general may be found in Sims et al. (1984), Hoogendoorn (1984), Cullinane and Jones (1984), Spooner (1984), Rulkens and Assnik (1984), and Wagner and Kosh (1984).

2.3.3.4.1 Land treatment--The land treatability of PAH-contaminated soils and PAH-containing sludges has been demonstrated for petroleum refinery wastes (API, 1983) and for creosote used by the wood-preserving industry (Sims, 1984; Sims and Overcash, 1983; Umfleet et al., 1984; Patnode et al., 1985; Ryan and Smith, 1986). The fractional distillation of creosote from coal tar (creosote has a 200 to 400 °C distilling range), suggests that land treatment will be effective in treating soils contaminated with gasifier tars and oils. Comparison of contamination removal rates for creosote wastes and refinery wastes shows good agreement (Ryan and Smith, 1986); this implies that the land treatability of PAH-containing hydrocarbons is similar regardless of their source.

Currently, the wood-treating industry and the U.S. EPA are sponsoring studies to demonstrate the land treatability of creosote sludge and creosote-

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contaminated soils [Ryan and Smith, 1986; R. C. Sims, Utah Water Research Laboratory (UWRL), personal communication, 1986]. At one site in Minnesota, bench-scale and pilot-scale field tests have demonstrated the feasibility of land treatment of creosote-contaminated soils (Patnode et al., 1985; Ryan and Smith, 1986). Important results of this study are:

- Percent removals of benzene-extractable hydrocarbons averaged about 40 percent over 4 months, with a corresponding first-order kinetic constant of 0.004.

- Complete toxicity reduction appeared to fall between 2.5 and 5.0 percent benzene-extractable content. Two out of five test plots were nontoxic after 4 months (those with lowest initial application rates). All plots showed significant degradation.

- Microbial assays suggested that initial concentrations of creosote compounds would kill soil microorganisms and inhibit degradation. This did not occur. In addition, seeding plots with adapted microorganisms did not significantly enhance degradation. This implies that an active, adapted microbial population naturally developed in the contaminated soil.

- Within the range of loading rates tested (4 to 10 percent benzene extractables), no correlation between loading rates and kinetic rates was observed, with the exception of 4+ ring PAH compounds, which showed a slight inverse relationship between loading rates and kinetic rates.

- All loading rates tested (4 to 10 percent benzene extractables) were feasible.

- Toxicity reduction occurred at a faster rate at 4 to 5 percent initial loading rate than at higher loading rates.

- Greater kinetic rates were observed after waste reapplication to a treated soil.

- At this site, 3 to 5 years would be necessary to treat 12,500 tons of contaminated soil.

- Waste application rates of 2 to 3 pounds of benzene extractables per ft^3 of soil per 2 months can be degraded.

This study demonstrates the feasibility of land treating sandy soils contaminated with creosote wastes in Minnesota. Treatment times should be lower in warmer areas with a longer growing period. Preliminary results from an ongoing study in California suggest similar kinetic degradation rates in clayey soils (Ryan and Smith, 1986).

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TABLE 48. COST ESTIMATES FOR REMEDIAL ACTION ALTERNATIVES
AT A CREOSOTE IMPOUNDMENT

Alternative	Unit cost ($/ton)	Total cost[a] ($1,000)
Land treatment (onsite)	51	738
Landfill	200	2,500
Incineration (onsite)	184	2,300
Incineration (offsite)	1,900	23,750

[a]12,500 tons contaminated material.

SOURCE: Patnode et al., 1984.

One of the most significant results of the Minnesota study is that onsite land treatment is very cost-effective. Table 48 compares the cost estimates of land treatment with other options (i.e., landfill and incineration); land treatment cost estimates were lowest at $51/ton. If onsite conditions are not amenable to land treatment, costs will increase as a result of transportation costs to a suitable treatment site. However, even if this results in costs higher than landfilling, land treatment will still be preferable because it can detoxify the waste, thereby eliminating long-term liability. Comparison of onsite land treatment costs with onsite incineration (Table 48) demonstrates that land treatment is more cost-effective.

Our review of remedial alternatives for soil contaminated with tars and oils from gas plant manufacture indicate that land treatment is the best demonstrated treatment technology. It appears to be cost-effective, as well as effective in detoxifying the wastes. The age of all gasworks sites further supports this conclusion because soil microbes capable of degrading tar and oil compounds will have had time to evolve. The Ames, Iowa, case study (see Chapter 3) demonstrates this; organisms capable of degrading PAH compounds have evolved in the groundwater at Ames.

Several questions remain unanswered with respect to applying the results of the creosote studies to gas plant residuals. First, creosote is a distillate fraction of coal tar; the tars and oil at former gas plants tend to have a broader boiling point range. In addition, creosote is derived from coal tar; most gas plants operated water-gas processes, which produced tars with different composition (e.g., no tar acids or bases), it is not clear how this will affect soil toxicity and degradation rates. It does seem possible that soil microbes will have adapted to whatever tar constituents are present at a site. Other soil contaminants present at gas plant sites also could affect the land treatability of contaminated soil. Complex iron cyanides are not amenable to land treatment (Hoogendorn, 1984); free cyanides are rapidly broken down by soil microbes at concentrations below 200 ppm; and, as long as complex iron cyanides do not release free cyanides at rates sufficient to elevate soil levels to above 200 ppm, they may not affect degradation. Sulfur and arsenic compounds also may be present and could influence degradation rates. Another question is the volatilization of volatile components in coal

tars and oils during land treatment operations. These questions can be addressed by site-specific land treatment demonstrations such as those required for permitting a facility under RCRA (40 CFR 264).

Studies to demonstrate the treatability of contaminated soils and tars and oils should include bench-scale and pilot-scale tests to evaluate the effect of various design and operational parameters on the treatability of the wastes in question. These parameters include:

- Soil characteristics
- Waste characteristics
- Treatment supplements
- Climate
- Initial loading rate
- Reapplication rate
- Soil lift thickness
- Frequency of tilling.

Treatability studies should be directed toward determining the effects of these parameters on the reduction of organics, PAH's, and toxicity for the wastes or contaminated soils to be treated.

When conducting a treatability study, soil conditions that promote the degradation of hydrocarbons should be maintained. These conditions include (Ryan and Smith, 1986):

- Soil pH of 6.0 to 7.0 in the treatment zone
- Soil carbon-to-nitrogen ratios of 25:1
- Soil moisture near field capacity.

Other criteria that have been recommended for land treatment of creosote wastes include:

- Small and frequent fertilizer applications
- Waste reapplication only after initial applications have been effectively degraded.

U.S. EPA has published general guidance on land treatment demonstrations (EPA, 1984a; EPA, 1983a; and EPA, 1983b). EPA also has released a draft technical guidance manual on hazardous waste land treatment demonstrations for

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public comment (EPA, 1984b). This latter document currently is being revised to address and incorporate the public comments (R. C. Sims, UWRL, personal communication, 1986). It should be stressed that each of these EPA documents presents guidance only and not regulations.

The detailed design of a land treatment unit for gasifier wastes will depend on the conditions at the specific site. Although onsite land treatment is most economical, the location of many former gas plants in populated, urban areas may preclude onsite treatment.

Regardless of whether treatment is to be conducted onsite or offsite, the contaminated soil to be treated must be excavated and stockpiled at the treatment site. The stockpile may be covered and placed on a liner to prevent spread of contamination. The treatment area should be lined, and a leachate collection system installed, to prevent migration of leachate. The contaminated soil is then laid down in 1 to 1.5 foot lifts, and soil amendments and water are added as necessary to reach and maintain optimum soil condition for degradation (determined in bench-scale and pilot-scale studies). It may be necessary to blend clean soil with the waste or contaminated soil to achieve the desired contaminant loading rate. The soil should be cultivated regularly during the treatment process; soil conditions (moisture, pH, nutrients, etc.) should be carefully monitored and controlled. Once the initial lift has been detoxified, a second lift is placed on the previous lift, and so on until all the soil is treated.

Leachate collected from the land treatment facility may be treated or discharged without treatment, depending on the level of contaminants. At the Minnesota creosote treatment site, the State and EPA permitted discharge of leachate either into the Mississippi River or into the municipal sewage system, depending on the level of PAH compounds in the leachate. This implies that dissolved PAH's may be successfully treated in municipal wastewater treatment plants.

Land treatment is therefore a well demonstrated, effective technology for degrading PAH compounds. Field and bench-scale treatability studies on creosotes have demonstrated that a range of initial loading rates are acceptable and that degradation time increases with increasing loading rate. The selection of loading rate should balance land area requirements and time require-

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ments for completing the treatment process (Ryan and Smith, 1976). Lower loading rates decrease the time required for degradation, and higher loading rates decrease the land area requirements. Further information on the design and demonstration of land treatment may be found in Overcash and Pal (1979), API (1983), and EPA (1983a and b, 1984a, b, and c).

2.3.3.4.2 Extraction or thermal treatment of excavated contaminated soil--Hoogendoorn (1984) and Rulkens and Assnik (1984) reported on the successful pilot-scale use of a hot aqueous alkali solution to clean gasworks soil contaminated with free and complexed cyanides. The process (Figure 67) has been scaled-up to 25 tonnes/hr and is estimated to treat soils at a cost of $24.80 to $99.20/m^3. Soil is pretreated to remove large objects (wood and stones) and to break up clods. It is then extracted with a lye solution, the soil and cleaning agent are separated, and the extraction agent is cleaned by pH adjustment, coagulation, flocculation, sludge separation, sludge dewatering, and a second pH adjustment. The sludge, containing free and complexed cyanides, may be landfilled or incinerated; hydrolysis also may be practical. However, there is little experience in applying incineration and hydrolysis to these sludges. The alkali extraction process should be applicable to soils contaminated with PAH compounds as well (Hoogendoorn, 1984). Current applications are limited to clean sands; difficulty in applying extraction techniques to loamy soils include difficulty in separating clay/silt suspensions and strong adsorption of contaminants and clay particles.

The excavation and extraction of contaminated soils is economical in the Netherlands because of the high cost and intensive utilization of land and the high demand for clean fill. In the United States, this alternative may not be the most cost-effective one. The in-situ extraction of organics by alkali solutions has been demonstrated for industrial sludges (Kosson et al., 1986). This technique should be more economical than excavation and extraction, may be applicable to organic-contaminated soils at gasworks, and may be more cost-effective than excavation and extraction. However, in-situ alkali extraction should not be used when cyanide contamination is present at a site because strong alkalies can dissociate complex iron cyanides into free cyanide compounds.

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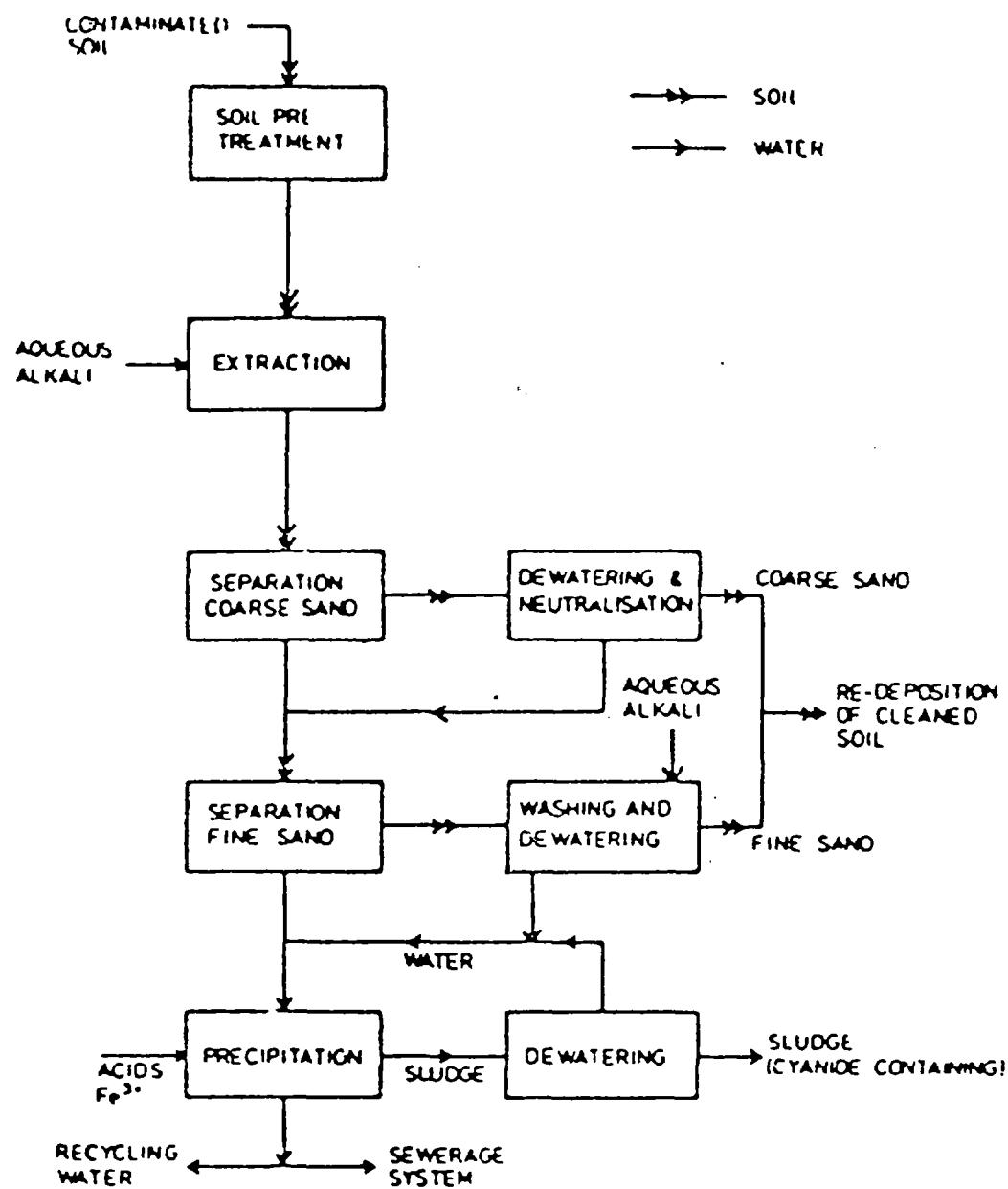
Source: Hoogendoorn, 1984.

NOTE: The arrows indicate the level of original coal tar injection.

Figure 67. Treatment of soil by extraction with an aqueous solution.

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Thermal treatment methods (high-temperature evaporation and incineration) also are applicable to soils contaminated with cyanides and PAH's (Hoogendoorn, 1984). Unlike the alkali extraction process, both sandy and clayey soils are amenable to thermal treatment methods. Evaporation at 850 °C has been used to clean cyanide and PAH-contaminated soil excavated from a gasworks site at Tilberg in the Netherlands. However, these techniques require excavation of the soil and are more expensive (after excavation) than is alkali extraction (Hoogendoorn, 1984). Thus, they may not be cost-effective even though they are technically effective.

2.3.3.4.3 Fixation--A novel, patented process for fixating wastes has been applied to gasworks wastes at Dortmund in the Federal Republic of Germany (U.S. Patent 4,456,400). Remedial investigations at the Dortmund site revealed extensive contamination. Liquid coal tar was clearly visible to a depth of 10 meters along with volatile hydrocarbons and sulfur compounds. Large quantities of spent iron oxide (containing sulfur and complexed cyanides) from gas purification were also present.

Remediation at this site involved excavating and treating the contaminated soil, contaminated water, and waste by mixing it (onsite) with lignite fly ash using the patented process (Heide and Werner, 1984). The treated material was finally disposed in a specially designed plastic-lined pit located on the site. This site cleanup was the first application of the technology on such a large scale. This cleanup approach is expected to result in considerable cost savings over an alternative plan involving removal of the contaminated material to an offsite licensed disposal facility.

The treatment/solidification process relies on the pozzolonic properties of the brown lignite fly ash. The ash used at this site was obtained from local power plants burning brown lignite coal. The contaminated soil, tars, and water are mixed with the ash in a three-stage reactor along with additional water. The exothermic reaction must be controlled carefully to maintain a continuous flow through the mixers. The product exiting the final mixing stage is a freely flowable slurry and is conveyed directly to the lined pit. Within approximately 30 minutes, the slurry hardens to a solid material that is claimed to be virtually impermeable to water ($<10^{-8}$ cm/sec). Data from numerous tests indicate that metals, sulfates, cyanides, and organics are

bound tightly in the treated material and are not leached even under rigorous conditions. Solid wastes, fluid suspensions, and sludges can all be treated by this process, being combined with the fly ash in amounts up to 50 percent by weight. Between 20 and 40 percent water is required in the process. From the standpoint of gasworks waste, the process is attractive because it can fix organic contaminants, cyanides, and sulfates.

The German governmental authorities granted approval for the site cleanup plan after 2 years of reviewing the data to support the proposed process and considering other alternatives. Protection of groundwater was the major concern. After the remediation is completed, the site will be used again for heavy industry. The pit containing the solidified waste will be monitored to ensure that there is no leaching of contaminants.

One limiting factor in the process is the availability of sufficient quantities of the lignite fly ash, which must be trucked in from local power plants. Brown coal ash is different from the ash of U.S. bituminous or anthracite coals because of its higher content of alkali metals (e.g., Na, K) and alkaline Earth elements (e.g., Ca, Mg). Brown coal ash contains about 10 percent CaO; it also contains calcium ferrite and calcium sulfate (Heide and Werner, 1984). It is this high concentration of calcium that is responsible for its pozzolanic properties. The ash of Western coals also tends to have higher calcium contents; however, the availability of fly ash from these coals is limited. It is possible that other fixation agents could be identified with similar properties or could be made up (e.g., by combining conventional coal fly ash and lime). The effectiveness of the fixation process may be evaluated by leaching tests such as EPA's EP or TCLP in soils. It may be the method of choice for remediating contaminated soil at gas sites.

2.3.3.5 Contaminated Groundwater--

The most significant groundwater contaminants at gasworks sites are light aromatics (e.g., benzene, toluene, xylene, ethylbenzene, naphthalene, acenaphthene indene). Incidents of significant offsite migration of gasworks contaminants in groundwater (e.g., Ames, Iowa; Dover, Delaware) have involved the lighter components of gasworks tars and oils that are easy to detect at ppb levels by the water's taste and odor. The concentrations of the heavier PAH compounds (three or more aromatic rings) in groundwater are generally lower,

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being controlled principally by their aqueous solubilities. PAH concentrations tend to drop off rapidly beyond the coal tar source; the persistence of these heavier compounds in groundwater beyond the immediate site area has not been documented.

2.3.3.5.1 Source control--The most important step in the remediation of contaminated groundwater is destruction or removal of the source of contamination. Until this is successfully accomplished, the success of groundwater cleanup will be limited by continuing contaminant release at the source. It is especially important to identify and remove any lighter organics (i.e., oils) present at a gasworks site because their higher solubilities and usual occurrence above the water table give these organics a high potential to contaminate groundwater. The heavier tars tend to cause localized groundwater contamination that is localized around the area of tar contamination. However, it is important to clean up free tars or to ensure that they will be effectively contained onsite; free tars can migrate significant distances frcm the site under certain subsurface conditions (see Section 2.1). Coal tars, produced in processes that involve coal pyrolysis, have more potential to contaminate groundwater than do water-gas or oil-gas tars because they contain significant quantities of more soluble tar acids (e.g., phenols, cresols, and xylenols.

Inorganic contaminants that can contaminate gasworks sites include sulfates (which can acidify groundwater) and trace elements (e.g., arsenic) associated with gas manufacture. The source of these contaminants includes spent oxide wastes and other solid waste from gas manufacture. Control of these contaminant sources may be accomplished by removal or treatment; in many cases, pH adjustment with limestone may be adequate treatment. Neutralization reduces acidity, raises pH, and thereby controls trace metal release. The potential for groundwater contamination by cyanides from solid wastes at gasworks sites also must be considered; however, no cases of significant contamination of groundwater by cyanides was found in this study. At the Birmingham, Alabama, site, leachate from untreated spent oxide wastes had free cyanide levels well below the level that can be effectively degraded by soil microbes (200 ppm), in spite of a pH of 1.5. In-situ treatment by limestone addition reduced free cyanide levels further, to below 20 ppm.

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2.3.3.5.2 Selection of groundwater treatment alternatives--In devising remedial actions for contaminated groundwater, one must consider the following:

- Containment control to prevent the further spread of contaminants and to collect groundwater for treatment

- Treatment to destroy or remove contaminants in the groundwater.

Both of these factors must be addressed when devising remedial actions for groundwater contaminants because the long times required to treat contaminated groundwater necessitate the containment activities, and it is often necessary to collect the groundwater prior to treatment.

Groundwater control measures for contaminant containment include physical barriers and hydrologic barriers. Selection of appropriate technologies depends on the hydrogeologic characteristics of the site and the extent of contamination. For instance, physical barriers such as slurry walls, grout curtains, and sheetpile cutoff walls and hydrologic barriers such as interceptor trenches or subsurface drains are appropriate for sites where contamination is confined to the near surface (25 to 50 feet deep) and underlain by a low-permeability layer into which the barrier may be keyed. Examples of the use of physical barriers (slurry walls) during gasworks site remediation may be found in the case studies for Stroudsburg, Pennsylvania, and Plattsburgh, New York, in Chapter 3. When contamination extends to greater depths, or where there is no natural barrier to vertical (downward) migration of the contaminant plume, hydrologic barriers using pumping wells may be the only appropriate control strategy. A hydrologic barrier using pumping wells was employed to control contamination from the Ames, Iowa, gasworks (see Case Studies, Chapter 3).

Groundwater collection strategies include subsurface drains and interceptor trenches, which are appropriate for shallow contamination, and pumping wells, which may be used for shallow or deep contaminated groundwater. Subsurface drains were used at Plattsburgh, New York, to collect incoming groundwater to prevent breaching of the slurry wall. The drain system also served to collect contaminated groundwater leaving the site (see Chapter 3). Pumping wells were used to collect free coal tar at Stroudsburg, Pennsylvania, and

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contaminated groundwater at Ames, Iowa, and may be employed to control and collect contaminated groundwater at St. Louis Park, Minnesota (see Chapter 3).

For further information on the selection, evaluation, and design of groundwater control strategies, see U.S. EPA (1982), Ehrenfield and Bass (1983), U.S. EPA (1984d), Boutwell et al. (1985), Schafer (1984), Xanthakos (1979), and D'Appolonia (1980).

Treatment alternatives for groundwater contaminated with aromatics from byproduct tars and oils include physical.methods (e.g., carbon adsorption, reverse osmosis), chemical methods (e.g., wet air oxidation, ozonation), and biological methods (Ehrenfield and Bass, 1983). At St. Louis Park (see Chapter 3), the groundwater remediation plan includes the use of granular-activated carbon to clean up contaminated groundwater. At Ames, Iowa, recovered contaminated groundwater was used, without treatment as boiler make-up water at a nearby power plant. Microbes capable of degrading PAH compounds were discovered in the contaminated Ames groundwater (see Case Study, Chapter 3). This suggests that, where groundwater is contaminated with organic compounds from gas plant wastes, indigenous microbes capable of degrading these organics may have evolved. In these cases, in-situ remediation may be possible by containing the groundwater and allowing natural degradation to take its course, with or without enhancement through the addition of oxygen (or air) and nutrients. For more information on biological treatment methods for contaminated groundwater, see Parkin and Calabria (1985).

2.3.4 Conclusions

The following conclusions can be made concerning the investigation and remediation of town gas sites.

- Site investigation techniques used at abandoned town gas plants do not differ significantly from those used at other uncontrolled hazardous waste sites.

- Because of the age of the sites, collection of historical information from company records, insurance maps, interviews with plant personnel, aerial photos, etc., is an important first step in site investigations at abandoned town gas plants.

- Surface geophysical techniques can be used to identify buried structures, pipes, and subsurface zones of coal-tar contamination at abandoned town gas plants, and they can help guide further site investigation activities.

- It is important to identify buried structures because these can contain tars, oils, emulsions, and other contaminants. Care should be exercised to avoid damage to these structures when using invasive site investigation techniques or when conducting remedial actions. If care is not taken, these substances may be released.

- The probable presence of multiple-density contaminants (i.e., tars and oils) should be considered when planning site investigation activities and when evaluating remedial action alternatives.

- The long-term stability (i.e., no release of hazardous substances over a period of years) of some sites may make no-action a viable alternative at some sites.

- Free tars and oils recovered at a site often may be sold for beneficial use as fuel or chemical feedstocks.

- Land treatment has been proven effective in treating soil contaminated with byproduct tars and oils. Other treatments used for removing or destroying heavy organics in soils also may be applicable.

- Spent oxide wastes and soils contaminated with complex cyanide compounds have been treated successfully by immobilizing with lime, or with a combination of lime and pozzolonic material, and evaporation at elevated temperatures.

- The presence of indigenous microbes capable of degrading aromatic compounds in the groundwater at Ames, Iowa, suggests that in-situ biological treatment may be feasible for groundwater contaminated with compounds from byproduct tars and oils.

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3.0 SITE INVESTIGATIONS OF SPECIFIC TOWN GAS SITES

3.1 INTRODUCTION

This chapter describes the specific town gas sites reviewed by RTI. It is divided into two sections: Section 3.2 describes the sites visited by RTI personnel, and Section 3.3 discusses case studies of town gas sites that have been described in recent literature. This chapter is designed as a overview of existing town gas sites, types of contaminants, and remedial actions.

In its review, RTI also collected some historical data from pre-1960 sources on specific sites that sometimes conflicted with the site information reported by other investigators. These contradictions are also examined in this chapter.

3.2 SITE VISITS PERFORMED BY RTI

3.2.1 Introduction

Mr. Scott Harkins of RTI visited six gas sites and one iron oxide disposal site to permit RTI personnel to collect data and site assessments on specific sites during the course of the project. Site assessments were available for only two of these gas sites (Lowell, Massachusetts, and Spencer, Massachusetts) and the spent oxide disposal site (near Attleboro, Massachusetts). One site was chosen because the authors were familiar with it, and because many of the structures were still present on the site (Richmond, Virginia). One other site (Taunton, Massachusetts) was recommended by the Massachusetts Department of Environment Quality Engineering (DEQE), and the other two were selected because they were within traveling distance of the other sites examined (Pawtucket, Rhode Island, and Worchester, Massachusetts). All of these sites and the information obtained during the site visits are described in the next section.

3.2.2 Colonial Gas Company, Lowell, Massachusetts

This Colonial Gas Company in Lowell, Massachusetts, was visited on March 3, 1986. The site examination consisted of reviewing the Phase 1 site assessment, visually examining the plant site and surrounding area (without entering the site), and collecting an early site map of the plant. This 17-acre site produced coal, water, oil, and LP gas for the town of Lowell, Massachusetts. The plant began as a coal-gas plant in 1849, added carbureted water gas during the 1870's, converted to oil gas between 1950 and 1951, and was placed on standby in 1951. It operated intermittently between 1951 and 1975 to supplement natural gas supplies. The site is currently used as an operations center and storage and gas distribution center by the Colonial Gas Company (formerly the Lowell Gas Light Company). The site is approximately 300 feet from the Pawtuckett Canal, which removes water from the Merrimack River, flows through the town of Lowell, and then returns to the river.

An 1876 map of Lowell (available at a local national park gift shop) clearly shows the plant layout, with five large buildings and four masonry gas holders. Two buildings on this map currently remain onsite. A vacant area is seen next to the plant and is now part of the plant site.

A Phase 1 site investigation (problem definition and site history) of the site was completed in December 1985 by M. Anthony Lally Associates, and a Phase 2 site investigation (problem evaluation and field investigation) is currently planned. These investigations were in response to observed volatile contamination of soil and groundwater during an investigation of PCB contamination on the property adjacent to the site. VOC's were detected at 65.1 mg/L in groundwater flowing from the gas site.

Soil samples were taken and organic vapor concentrations measured from shallow depths (0 to 3 feet) around the plant. Organic vapor concentrations from the probe hole varied between 0 and 96 ppm, and soil concentrations were between 0 and 37 ppm. Analyzed soil samples showed contamination by benzene (0.013 mg/g), toluene (0.004 mg/g), ethyl benzene (0.030 mg/g), xylenes (0.23 mg/g), and assorted PAH compounds (1.09 mg/g). RTI's examination of the site area found two small sources of oil flowing into the canal from the canal wall nearest the gas site. The water in the canal was lowered for routine maintenance during the visit. The canal itself, and several areas around the plant,

had fairly strong gaseous odors, probably from gas plant wastes. Diagrams of the plant site contained in the Phase 1 site assessment indicate a "deep well" was present. This well was possibly used for waste condensate disposal because any liquid wastes dumped into the canal would flow through the center of town and pass through water-powered factories.

A literature review by RTI revealed that several articles were written by engineers working at the Lowell plant. One article on oxide purification of gas stated, "Because of the possibility of fires starting through the heat generated by revivification, it is necessary to hold the spent material at the plant until this danger is past. As soon as city authorities learn of this menace the material is prohibited at public dumps. Continuous storage on gasworks land eventually becomes impossible. The material makes excellent filling for roads or private property when properly handled. It should be covered with ashes or dirt immediately to prevent the access of air and consequent combustion. ...The plant is indeed fortunate it has a place to store the spent oxide and doubly so if a transportation company will agree to remove it without charge because of its value as a filling material" (Downing, Superintendent of Manufacturing, Lowell Gas Light Company, 1932).

Evidence of tar and oil contamination of the site was also located in an article on gas plant wastes. "That large quantities of gas house waste can enter the ground is strikingly shown by investigations made at the Lowell, Massachusetts, gas works in 1905 and 1906 by A.T. Stafford and W.H. Clark, who estimated that there existed within the ground and within an area of a few acres 1,600,000 gallons of tarry and oily wastes. Some of these consisted of accumulations in old drains and porous gravel, which when tapped by excavations flowed out in springs. Much waste was regularly finding its way into sewers, and from the sewers it entered cellars along the lines of sewers at even remote distances from the works" (Hansen, 1916). RTI has yet to locate the articles Hansen referred to, but if accurate, they indicate possible widespread contamination from the facility.

3.2.3 Massachusetts Electric Company, Spencer, Massachusetts

The Massachusetts Electric Company in Spencer, Massachusetts, was examined on March 4, 1986. The site examination consisted of viewing the fenced portion of the site through the fence, making an examination of the perimeter

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of the site, and examining the site assessment prepared by Perkins Jordan in January 1984.

The site came to the attention of the Massachusetts DEQE when a truckload of soil (removed so that a drainage culvert could be installed) was delivered to a landfill during a routine inspection of the landfill. The inspector recognized the materials as being gas production wastes and ordered that they be returned to the site. A subsequent site investigation by Perkins Jordan used nine test pits, seven borings, and two test wells.

This also was the site of a very small carbureted water-gas plant. (Brown's Directory and the 1917 report of the Massachusetts Board of Gas and Light Commissioners show it to be a carbureted water-gas plant, but the site assessment identifies it as coal-gas plant.) It was constructed between 1885 and 1887 and operated into the 1950's. The site is approximately 0.4 acres and adjacent to a small stream, the "Muzzy Meadow Brook." It is currently fenced off and is the site of a power substation. Wastes typical of carbureted water-gas plants (coal, coke, ash, slag and tars) were identified at the site. About 15 to 20 feet of soil rests on top of the bedrock at the site. The depths of the test well and pits were limited by the bedrock under the site.

Soil samples were found to be contaminated with toluene, benzene, ethylbenzene, PAH compounds, and xylenes. Groundwater samples contained low levels of PAH and volatile compounds. Table 49 shows the measured concentrations of volatile and semivolatile compounds in soil and water samples from test pits and brook samples. Table 50 shows the same analysis for soil samples from borings, and Table 51 shows concentrations from monitoring wells. The brook flowing beside the site showed no detectable contamination.

3.2.4 Fulton Gas Works, Richmond, Virginia

Fulton Gas Works in Richmond, Virginia, was visited on February 7, 1986. The site examination consisted of touring the site structures, guided by an employee of the Richmond gas company, examining the site perimeter for wastes and dumping locations, and visiting the Richmond public library for information about the manufactured-gas site. The 8-acre site, which began as a coal-gas plant and later switched to carbureted water gas around 1895, produced coal and carbureted water gas for the city from before 1860 to 1952.

TABLE 49. VOLATILE AND SEMIVOLATILE ORGANIC ASSAY RESULTS FROM TEST PIT AND BROOK SAMPLES
(FROM PERKINS JORDAN, 1984)

Chemical compounds	Soil samples (mg/kg) ppm					Water samples (mg/L) ppm				
	TP-1 S-2	TP-2 S-6	TP-6 S-17	TP-9 S-19	TP-9 S-30 & 31	TP-2 S-9	TP-2 S-10 & 11	TP-7 S-20	Brook upstream	Brook downstream
Volatile organics										
Toluene	NA	NA	NA	NA	0.360	NA	0.0070	NA	NA	NA
Benzene	NA	NA	NA	NA	0.065	NA	--	NA	NA	NA
Ethylbenzene	NA	NA	NA	NA	>7.00	NA	0.0052	NA	NA	NA
Xylenes	NA	NA	NA	NA	>20.0	NA	0.037	NA	NA	NA
Semivolatile organics										
Acenapthylene	--	2.50	--	130	NA		NA	0.240	--	--
Acenaphthene	--	1.00	--	48.0	NA	--	NA	--	--	--
Benzo(k)fluorene	--	0.01	--	6.60	NA	--	NA	--	--	--
Benzo(a)pyrene	--	0.93	--	7.90	NA	0.040	NA	0.064	--	--
Chrysene	--	1.40	--	18.0	NA	0.079	NA	0.116	--	--
Fluoranthene	--	3.20	--	91.0	NA	0.200	NA	0.170	--	--
Fluorene	--	3.60	--	120	NA	--	NA	0.270	--	--
Phenanthrene	--	6.20	--	160	NA	0.200	NA	0.760	--	--
Pyrene	--	2.80	--	66.0	NA	0.150	NA	0.250	--	--
Naphthalene	--	--	--	400	NA	--	NA	0.330	--	--
Diethylphthalate	0.070	--	--	--	NA	--	NA	--	--	--
Bis(2 ethylhexyl) phthalate	1.60	1.40	0.010	--	NA	--	NA	--	--	--
Di-n-butylphthalate	0.020	--	--	--	NA	--	NA	--	--	--
Total polynuclear aromatic hydrocarbons (PAH's)	--	23.24	--	1,037	NA	0.829	NA	2.194	--	--

SOURCE: Perkins Jordan, 1984.

NA = Not analyzed.
-- = Not detected.
TP = Test pit identification.
S = Sample number.

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TABLE 50. VOLATILE AND SEMIVOLATILE ORGANIC ASSAY
RESULTS FROM BORING SAMPLES

	Soil samples (mg/kg) ppm			
Chemical compounds	B-2 S-42 & 43	B-2 S-41	B-3C S-48	B-4 S-52
Volatile organics				
Toluene	5.80	NA	NA	NA
Benzene	75.0	NA	NA	NA
Ethylbenzene	41.0	NA	NA	NA
Xylenes	53.0	NA	NA	NA
Semivolatile organics				
Acenapthylene	NA	80.0	4.60	--
Acenaphthene	NA	46.0	9.30	--
Benzo(k)fluorene	NA	11.0	--	--
Benzo(a)pyrene	NA	100	1.30	0.520
Chrysene	NA	31.0	--	0.720
Fluoranthene	NA	130	6.70	2.00
Fluorene	NA	170	8.40	--
Phenanthrene	NA	370	21.0	1.60
Pyrene	NA	100	8.0	1.40
Naphthalene	NA	670	39.0	--
Diethylphthalate	NA	--	--	--
Bis(2 ethylhexyl)phthalate	NA	--	--	--
Di-n-butylphthalate	NA	--	--	--
Total polynuclear aromatic hydrocarbons (PAH's)	NA	1,708	98.3	6.24

SOURCE: Perkins Jordan, 1984.

NA = Not analyzed.
-- = Not detected.
B = Boring identification.
S = Sample number.

TABLE 51. VOLATILE AND SEMIVOLATILE ASSAY RESULTS FROM MONITORING WELLS

| Chemical compounds | Groundwater samples (mg/L) ppm (taken on 11-30-83) | |
	MW-1	MW-2
Volatile organics		
Toluene	0.0095	0.120
Benzene	0.071	0.410
Ethylbenzene	0.015	0.480
Xylenes	0.068	0.610
Semivolatile organics		
Acenaphthylene	ND	0.041
Acenaphthene	ND	0.032
Anthracene	ND	0.004
Fluorene	ND	0.030
Naphthalene	ND	1.000
Total polynuclear aromatic hydrocarbons (PAH's)	ND	1.147

SOURCE: Perkins Jordan, 1984.

MW = Monitoring well.
ND = Not detected.

A complete investigation (documented in transcripts in "The Affairs of the Richmond Gas Works," [no author] 1896) followed a major scandal at the gas plant shortly after the conversion, thereby marking that year. The scandal involved several plant supervisors who were dumping ash-coke mixtures from the water-gas generators before most of the coke was converted to gas and ash. This allowed the ash to be hauled from the plant and the coke recovered and sold for the profit of those who stole it. Also during this period, the plant operated for 9 months with no down-run on the carbureted water-gas apparatus (the down-run valve had burned out). This caused poor heating of the apparatus and resulted in the inadequate cracking of carburetion oils. The creek behind the plant was reportedly full of oil that overflowed from the relief holder with condensate.

The report states that the fences were rotted, the roofs decayed, the coal benches were clogged and had to be rebuilt, the water-gas plant needed to be relined, new castings and valves were needed, the purifying house oxide boxes were rusted and leaking, the condensers were broken, employees were mismanaged, and coke was constantly stolen. The report indicates that the plant sold coke, tar, sulfate (probably ammonium sulfate), lime, and junk (the type of junk was not defined).

When the plant switched to carbureted water gas, they also switched from lime purification to the use of iron oxide (the new purifier house was erected in 1894). They had previously used 9,000 to 10,000 bushels of lime per month (415 to 460 ft^3). This use dropped to 80 bushels per month after converting to iron oxide purification. This gas plant, also referred to as the lower gasworks, is shown on maps in the library. One map from 1888 clearly shows another gas plant along the river, closer to the center of the city. An 1876 map of the gasworks (Figure 68) clearly shows the plant layout and structure. The round object below the coal shed is labeled as a retort by the mapmaker, but it is actually a gas holder for the plant.

The plant continued to produce water gas until the early 1950's, when the plant was converted to LP gas for peak loads and standby operation. Most of the buildings present in 1950 remain on the site, i.e., the gas house, compressor building, purifier buildings, coal shed, and gas holder. The purifier building has been converted into a welding shop and classrooms for the

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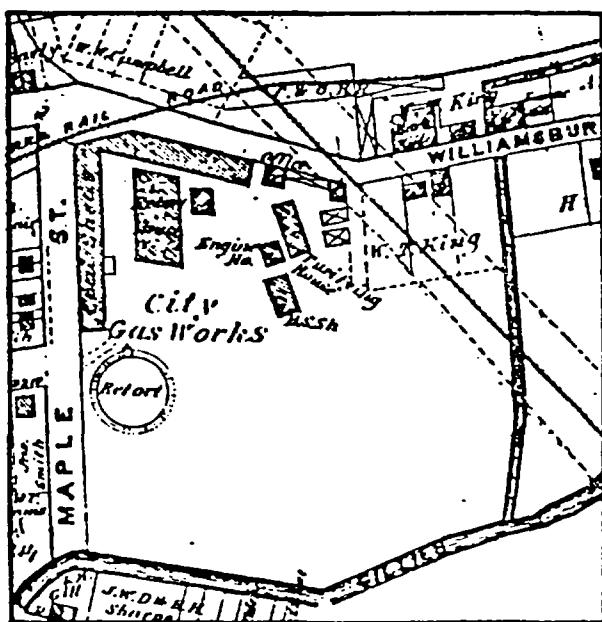
Figure 68. Fulton Gas Works (1876).

current, municipally owned gas company. The other buildings are not used and probably would have been removed many years ago if the plant were not city owned. (Cities do not collect property taxes from their buildings.) All of the buildings, except the purifier building and the gas holder, were to be removed during the spring of 1986.

The site is adjacent to a concrete culvert (formerly a creek) that flows into the James River about 600 feet from the site. The area between the gas plant and the creek shows substantial signs of being a dump area for the plant, with contaminated woodchips, ash, coke, firebricks, and tar present. No site or environmental assessment of the plant is currently planned prior to removing site structures. The entire site was flooded with about 6 feet of water from the James River during the fall of 1985.

3.2.5 Mendon Road Spent Iron Oxide Disposal Site, near Attleboro, Massachusetts

The Mendon Road Spent Iron Oxide Disposal Site near Attleboro, Massachusetts, was visited on March 3, 1986. The site was evaluated by visiting the site during site remediation and by examining two reports of the site, a geohydrologic study by Clean Harbors, Inc. (May 1985) and a hazardous waste evaluation by Hydrosample (November 1984). This was not a gas site, but a site where some spent oxide waste was disposed. The site was originally a gravel pit, but it later became a dump and was recently filled and houses were constructed on the site. When the land was purchased, the buyers sent a sample of the waste to the State health department to approve construction. The perk tests revealed additional waste, and this information was sent to the Massachusetts DEQE. After two subsequent site investigations, removal of the spent oxide wastes began with funding from the State Superfund.

The waste is spent iron oxide (mixed with woodchips) from coal-gas manufacture. The waste was apparently used as fill at the site, with other fill material above and below the waste "seam." The waste material consisted of contaminated woodchips with high concentrations of PAH compounds, iron cyanides (total CN 7,500 ppm, soluble CN 0.7 ppm), and low pH (1.7 to 3.8). It passes the EP toxicity but has a high total metal content. The waste was a seam of material with a maximum thickness of about 3 feet, covered by between 1 and 4 feet of clean topsoil. The site remediation was to remove all cyanide

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to a concentration of 2 ppm in the soil. Clean topsoil was removed and put aside; the waste and an additional foot of soil below the waste were removed, stabilized with calcium sulfate, and transported to a hazardous waste landfill in Alabama. The resulting holes were filled with clean fill dirt. An early estimate of the necessary remediation was removal of 2,500 ft^3 of contamination at a cost of $1.6 million.

The solubility of the iron cyanide compounds in water was evidently very small. The cyanide wastes were removed from the equipment used in the remediation by physical means only. The equipment was hosed off with water, and the water was drained into a holding tank (approximately 200 gallons). The solids were allowed to settle to the bottom of the tank, and the clear water was removed from the top of the tank. This water was then run through a sand filter, and the resulting water was discharged without further treatment. The cyanides were essentially all removed by settling and filtration with sand. The material that settled in the tank, and the tank itself, were to be discarded in the Alabama landfill at·the end of the remediation.

A similar disposal of spent oxide wastes is on the ground surface just across the Rhode Island border.

3.2.6 Pawtucket, Rhode Island

The Pawtucket, Rhode Island, site was examined on March 5, 1986. It was evaluated by only a visual examination of the plant site and by data from Brown's Directory. This is a fairly large gas site that produced both coal and water gas during operation and had an electrical power plant as part of the site. The site occupies 20 to 40 acres between a residential neighborhood and the Seekonk tidal basin, just soutn ɔ̣̃ Pawtucket, about 3.5 miles from the Attleboro road site in Massachusetts. Part of the site is currently used as an electrical substation and for the distribution of natural gas. There were several areas of the site that contained spent oxide wastes similar to that at Mendon Road (e.g., woodchips from spent oxide, and blue areas of soil from ferrocyanides). A substantial amount of waste from the gas production and power generation was visible on and around the site, evidently as fill.

3.2.7 Taunton, Massachusetts

The Taunton, Massachusetts, site was visited on March 3, 1986, with a representative of the Massachusetts DEQE. The site was examined visually. It is a mid-sized gas plant that primarily produced coal gas but later produced water gas (Brown's Directory, as reported by Radian Corp). Constructed around 1890, the plant added water gas around 1920. The site, approximately 15 acres, is in an industrial area south of Taunton, adjacent to the Taunton River. All of the structures were removed in the early 1960's, but the site has never been properly decommissioned. Gas-holding tanks were cut off at ground level and filled with soil from the site. It is very unlikely that any underground structures were removed. The plant was located at the northern end of the site, and the southern part of the site was evidently used as a waste disposal area. The State DEQE was called by the gas company in the early 1970's to stop waste materials from eroding into the river. Eventually, the southern half of the site was capped with a layer of clay soil and top-soil. This southern half has a small stream that crosses it and currently flows through a lined culvert. The northern half has remained uncovered. No waste materials have been removed from the site, and approximately 1 to 3 feet of mixed wastes are under the capped area. Heavy tars, ash, and spent oxide wastes are visible in the uncapped area. The site is currently fenced, and the local gas company operates a standby LP gas facility across the street. No additional remedial actions or in-depth site studies are currently planned for the site.

3.2.8 Worchester, Massachusetts

The Worchester, Massachusetts, site was visited on March 4, 1986. The city was chosen because it is large, happens to be close to Spencer, and is listed in Radian's 1984 compilation of U.S. gas sites. Some information and maps of the plant site were located in the Worchester public library. The Worchester Gas company, chartered in 1849, moved to a 9-acre site on Quinsigamond Avenue in 1869. It produced both coal and water gas. Currently, the site is used by the Commonwealth Gas Co. as a gas storage and distribution facility. The entire site has been capped with approximately 3 feet thick of construction refuse and fill. The site has no noticeable wastes and only a

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slight odor. An EPA pollution control project (Project C250.347-04), a new
$7.5 million ($5.5 million Federal, $1 million State) sewage treatment
facility, is to be newly constructed on the gas site (as indicated by a sign
on the property).

3.3 CASE STUDIES OF TOWN GAS SITES

3.3.1 Introduction

The case studies in this chapter were selected to demonstrate the types,
modes of occurrence, and persistence of contaminants at abandoned manufactured-
gas sites, as well as applicable remedial measures for these sites. The case
studies are presented to support the material discussed in the preceding chap-
ters. They were collected from published literature, State and Federal agen-
cies, and previous work at RTI. Differences in detail between the studies
reflect different amounts of information available for specific sites. In
addition to six former gasification sites, two byproduct tar utilization
facilities, a creosoting plant (Pensacola, Florida), and a coal-tar processor
(St. Louis Park, Minnesota) are included. These two studies offer well-
documented evidence of migration and degradation of coal-tar derivatives in
the subsurface that is relevant to contamination at gas plants.

The case studies were compiled from the references presented at the
beginning of each study.

3.3.2 Norwich, Great Britain (Wood, 1962)

The Norwich, Great Britain, site is the oldest site found during this
study, having groundwater contamination from tar present for over a century.
It illustrates the potential persistence of gasworks tar in the subsurface
environment, both in terms of the tar's appearance and its potential to con-
taminate groundwater.

In 1950-1951, a 36-inch bore was sunk into the chalk aquifer underlying
Norwich for water-supply purposes. Although it produced water of sufficient
quality for its intended use, the well's yield was inadequate. To remedy
this, a horizontal adit was drilled from the bore into the chalk at a depth of
150 feet below the surface. Shortly after, the water acquired a tarry taste
and thus was rendered unusable. Subsequent colorimetric analysis indicated

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that the water contained about 0.2 ppm total phenols, which appeared to be largely cresols. Thiocyanates were below the detection limit of 0.01 ppm.

Inspection of the adit by descent into the well showed black tarry matter exuding from the adit roof. Samples of the tar contained a small proportion of volatile matter, which had a trace of phenols, but was mainly composed of a yellow oil with a blue fluorescence in benzene solution (suggesting the presence of aromatic compounds). The larger portion of the tar sample was non-volatile, tarry in consistency and odor, and contained particles of solid carbon.

The source of the tar was originally a mystery because the site was far from the Norwich gasworks. However, subsequent investigation revealed that the first gasworks plant in Norwich was constructed over this site. That plant, which operated from 1815 to 1830, produced gas from destructive distillation of whale oil by the Taylor process. Thus, the well constructed in 1951 was polluted by tar that had been lying in the ground for over 120 years.

This case study illustrates that tar from town gas processes can persist and retain its potential for environmental damage for over a century. The amount of tar degradation that may have occurred is impossible to estimate because there is little information on the original tar composition. However, of significance is that at least some of the tar acids (phenols and cresols) have persisted in spite of their high solubility, and they have contaminated groundwater. The absence of thiocyanates is expected because of the low sulfur content of whale oil. The tar's appearance and odor is similar to that of coal tar, illustrating that, with the exception of the formation of sulfur and nitrogen compounds, the gas production process is more important than feedstock composition in influencing tar formation. The "steam-volatile matter" reported by Wood (1962) probably corresponds to the naphtha or light oil fraction of tar, and it may be responsible for much of the observed groundwater contamination.

3.3.3 Ames, Iowa (Siudyla, 1975; Yazicigil, 1977; Yazicigil and Sendlein, 1981; Burnham et al., 1972; Burnham et al., 1973; Ogawa et al., 1981)

The Ames, Iowa, case study illustrates long-term contamination of a water supply by town gas wastes from a relatively small gas plant that served about 15,000 customers. Groundwater contamination was first detected by taste and

odor problems in 1927 and has persisted into the 1980's. This case study illustrates the following:

- Site discovery through odors in water caused by ppb levels of dissolved tar constituents in groundwater

- Contamination of groundwater by lighter tar fractions (tar oils) that are less dense than water and more soluble than heavier tar components

- Contaminant sources resulting from town gas waste disposal practices

- Contamination by tar wastes from a water-gas process, notable by their lack of tar acids (phenols, cresols, xylenols)

- Migration of contaminants through cracks in soil to the water table

- Influence of pumping wells on the migration of dissolved coal tar constituents in the groundwater

- Use of historical data in a site investigation

- Degradation of PAH's by microbes naturally occurring in groundwater at Ames

- Remediation through removal of contaminant sources, installation of barrier wells, and controlled municipal well pumpage.

3.3.3.1 Site History--

According to Siudyla (1975), who interviewed long-time residents of Ames, town gas was produced in Ames from 1911 until 1927. The original gas plant was in operation from 1911 until 1920, and it was located in the western sector of the Ames wellfield. In 1920, the plant was moved to its final location. Although there was a waste pit at the original plant site, 70 feet of glacial drift isolated this source from the underlying buried channel-sand aquifer. However, the drilling of a municipal well in 1968 through the pit and into the underlying aquifer resulted in some contamination of the aquifer by PAH's. Contaminant levels at the well have decreased over the years because the well has been pumped (Siudyla, 1975).

Brown's Directory indicates that the Ames plant operated from 1912 until about 1932 when gas lines were completed from Boone, Iowa. There is no mention of the plant's 1920 move, but it is indicated that Iowa Railway and Light

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purchased the gasworks in 1925. According to Brown's, the plant produced carbureted water gas over most of its history, with some coal gas being produced from 1916 to 1918. The directory notes that bituminous coal was used to fuel the plant after 1924. The operating data from Brown's is compiled in Table 52. No mention is made of fuel type prior to this entry, although coke, anthracite, or bituminous coals would be used in the generator, and gas oil or fuel oil would be used in the carburetor.

The appearance of a disagreeable taste and odor in groundwater from two city wells first occurred in 1927. The timing of the appearance could be related to the change in plant management in 1925, which could have affected waste disposal practices, or it may just reflect the time it took the contaminants to reach the wells from the source. In the early 1930's, three auger holes showed increasing concentrations of contaminants toward the waste pit at the second gas plant site, which was then recognized as the source of contamination. At that time, investigators determined that abandoning contaminated wells and drilling new wells farther from the source was the best solution. This practice was followed until 1961, when the wastes from the second pit were removed to a sanitary landfill in an attempt to mitigate the problem. It did not. By the late 1960's, five wells had been abandoned and several were restricted to limited pumping.

In 1975, Siudyla interviewed a former gas plant employee and discovered that an overflow channel not visible on any city maps had once flowed from the waste pit to the Skunk River. Although now buried with fill, the channel was described as once being "odorous...containing pools of coal tar wastes" (Siudyla, 1975). Subsequent sampling showed that oils had collected in two low areas in the former channel and were floating on top of the water table at these locations. These areas were identified as the contamination sources of the city's water supply aquifer. The type of organic contamination was thus discovered, and its oily nature is consistent with the disposal of waste condensate (and floating oils) from carbureted water-gas manufacture.

3.3.3.2 Extent of Contamination--

As previously described, the taste and odor problems in Ames' groundwater have existed since 1927. Originally attributed to phenolic compounds, analytical work in the early 1970's showed a notable lack of phenolics. The

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TABLE 52. GAS PRODUCTION AT AMES, IOWA

Year	Process	Production (10^6 ft^3/yr)		Population	Customers	Other data
1889	Not listed					
1891	Not listed					
1892	Not listed					
1894	Not listed					
1900	Not listed					
1902	Not listed					
1904	Not listed					
1906	Not listed					
1908	Not listed					
1910	Not listed					
1912	Lowe (Tenney)	7.5	650 Btu 20 CP	5,000	300	Ames Gas Co. President listed as C.I. Tenney.
1914	Water (Tenney)	14	20 CP 650 Btu	4,223 Ames 2,139 Nevada	817	Iowa Public Service (Consolidation of Ames Gas Co. serving Nevada).
1916	Coal and water (Tenney)	25	16 CP 600 Btu	4,223 Ames 2,138 Nevada 3,000 Ames College	1,150	13,500 holder capacity.
1918	Coal and water (Tenney)	27	600 Btu 16 CP	11,500	1,400	Byproducts: 65,000 gallons tar 2,400 tons coke.
1920	Water gas (Tenney)	36	660 Btu	7,900	1,400	
1922	Water gas (Tenney)	35	560 Btu	9,500	1,400	
1924	Water (Tenney)	44	560 Btu	15,950	1,584	105,000 holder capacity. Used for gas production: 9,000 tons bituminous coal 135,000 gallons gas oil.

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TABLE 52 (continued)

Year	Process	Production (10^6 ft^3/yr)		Population	Customers	Other Data
1928	Water gas (Tenney)	?	550 Btu	16,200	1,854	Aquired by Iowa Railway & Light in July 1925. Used for gas production: 334 tons bituminous coal 62,571 gallons gas oil.
1928	Water gas (Tenney)	1.3	525 Btu	12,143	2,017	Iowa Railway & Light Corp. Used for gas production: 9,904 gallons gas oil 44.5 tons coke in generators 215 tons coal in boilers. Byproducts: 5,760 gallons tar.
1930	Carbureted water gas	56.5	525 Btu	9,332	1,817	
1932	Carbureted water gas	60.9	525 Btu	10,261	1,975	Gas purchased from Boone division of Iowa Railway & Light.

SOURCE: Brown's Directory.

Lowe = Carbureted water gas.
Holder capacity in ft^3.
CP = Candle power.
Btu = Btu/ft^3.

predominant contributor to taste and odor was found in the neutral component of the groundwater organics, which included several aromatic compounds. Table 53 presents concentrations of these organics. No basic organic compounds were found. The lack of tar acids (i.e., phenolics) is consistent with the water-gas process that operated at the site; water gas does not produce significant tar acids.

Recent analysis of Ames' groundwater for heavier PAH compounds [e.g., phenanthrene, benzo(a)pyrene] has shown these compounds to be present, but at very low concentrations (Tom Neumann, Ames Municipal Water Department, personal communication, 1986). The concentrations of heavier PAH's in water from the dewatering wells were slightly higher than those in water use wells, but no wells showed total PAH levels above 100 ± 80 ng/L, and all levels were below levels of concern and World Health Organization (WHO) water quality criteria. The low level of the heavier PAH's is consistent with their low solubility in waters.

The source of contaminants in the Ames' aquifers was the waste pit and the overflow channels. There is no information on the type and disposition of contaminants in the original disposal pit prior to its removal in 1961. The overflow channel did receive some pit wastes, but these may largely represent the lighter floating components of the tar and wastewater disposed in the pit. Soil auger borings and test pits were used to investigate the overflow channel. The borings showed four levels of contamination: (1) odor, oil, and tar; (2) odor and oil; (3) odor alone; and (4) no odor.

Determination of the vertical extent of contamination from the soil borings was difficult because of contamination of the auger as it passed through the upper levels of oily and tarry materials. Test pits, dug to 10 feet, showed that the contaminants had moved downward through vertical cracks in the alluvial materials and that oil was floating on the groundwater table (Yazicigil and Sendlein, 1981). Subsequent excavation of the contaminated material indicated that heavier contaminants (heavy oil and tar) had moved below the water table and that pockets of tar in an almost solid state existed in the excavated material. Excavation depths were limited to 15 feet because of the high water table (at 8 feet). However, the lighter oil, floating on the water table, was probably largely responsible for the taste and odor

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TABLE 53.   NEUTRAL COMPOUNDS IN A CONTAMINATED AMES, IOWA, WELL

Compound	Concentration (ppb)	Std. Dev.
Acenaphthylene	19.3	1.4
1-Methylnaphthalene	11.0	0.6
Methylindenes	18.8	0.8
Indene	18.0	1.5
Acenaphthene	1.7	0.2
2-2-Benzothiophene	0.37	0.11
Isopropylbenzene		
Ethyl benzene		
Naphthalene		
2,3-Dihydroindene	15	
Alkyl-2,3-dihydroindene		
Alkyl benzenes		
Alkyl benzothiophenes		
Alkyl naphthalenes		

SOURCE: Burnham et al., 1972.

problems in groundwater, and this was removed by the excavation of the channel area.

The wellfield for Ames, to the north of the site, has been contaminated by tar constituents in spite of a regional hydrologic gradient to the southeast. Pumping of municipal wells appears to have locally reversed the gradient, causing contaminants to flow northward from the source to the municipal wellfield. Burnham et al. (1973) demonstrated that total concentrations of aromatics at a given well in 1972 were directly proportional to the demand placed on the well (total pumpage) over a period from 1935 to 1972. Drawing on this conclusion, Yazicigil and Sendlein (1982) modeled the Ames' aquifer system and various remediation alternatives. Based on their investigation, they suggested removal of the source materials, installation of pumping wells to create a hydrologic barrier between the source and the wellfield, and controlled municipal well pumpage to control the problem and prevent further well contamination.

Ogawa et al. (1981) studied the degradation of aromatic compounds in samples of Ames' groundwater. They found that, at a 25 to 150 μg/L concentration, acenaphthylene, acenaphthene, 2-methylnaphthalene, 2-methylindene, 3-methylcindene, and indene were almost totally degraded at ambient temperature within 3 days. Decay rates were highest for acenaphthylene and lowest for indene. Additionally, acenaphthylene was degraded even when spiked into the Ames' well water at its solubility limit (3 mg/L). Degradation could be prevented by filtering the groundwater through a 0.45-μm filter. Samples of distilled water and uncontaminated Ames' groundwater that were spiked with acenaphthylene (at 80 μg/L) showed no degradation of this compound after 18 days. However, when similarly spiked samples when inoculated with water from a contaminated Ames' well, the acenaphthylene was degraded within 9 days. Inoculation with anaerobic and aerobic bacteria from a sewage treatment plant resulted in no degradation.

These results suggest that a population of microbes capable of degrading aromatic compounds has adapted in the contaminated Ames' groundwater. Cell mass measurements and microorganism counts further support this conclusion. Correlated with the decrease in aromatic constituents, Ogawa et al. (1981)

observed an increase in both cell mass (from 2 to 20 mg/L) and microorganism count (from 102 to 104 cells/mL).

Ogawa et al. (1981) also measured the degradation of heavier PAH compounds spiked into aged Ames' groundwater that was formerly contaminated. Acenaphthene, phenanthrene, and fluoranthene (added at a 150 μg/L level) were degraded within 36 days. Pyrene, which had the same concentration, was 56 percent degraded in the same period. Thus, degradation rate of the PAH compounds decreases as the number of rings increases, as was also illustrated by degradation rates for the lighter PAH compounds (discussed previously).

The Ogawa et al. (1981) study demonstrates that dissolved PAH compounds, at concentrations up to their solubility limit, can be degraded by microbes naturally occurring in groundwater and that these microbes do not normally occur in groundwater, but may adapt in groundwater contaminated with PAH compounds. These conclusions are important for the remediation of abandoned coal gasification sites. Degradation of compounds by microbes suggests that cleanup of groundwater contamination may be possible by somehow enhancing this degradation, either by aeration and adding nutrients to the groundwater and/or by enhancing the degradation rates of these microbes by breeding more active strains. Additionally, groundwater with no PAH-degrading microbes may be inoculated with water from groundwater systems where microbial degradation is occurring.

3.3.3.3 Site Remediation--

To date, site remediation at Ames has consisted of following the recommendations of Yazicigil and Sendlein (1982), i.e., removal of the source of contamination, installation of two dewatering wells to form a hydrological barrier between the source and the wellfield, and careful management of the pumpage in the individual city wells.

The sources in the overflow channel were removed in 1980-1981 by excavating a 30 x 70 x 15 foot deep trench, removing contaminated material to a landfill, and replacing it with clean fill along the length of the channel. Two dewatering wells, installed to the north of the channel to permit this excavation, are now pumped to create a hydrologic barrier between the overflow channel and the wellfield to the north. Water from these wells is used at a nearby power plant. It is too early to assess the effectiveness of the removal

action in mitigating the contamination, but increases in contaminant levels have not been observed in either the barrier wells or in the last two wells closed in the municipal wellfield (Tom Neumann, Ames Municipal Water Department, personal communication, 1986).

Questions remain about whether the source of contamination has been sufficiently removed because the depth of the excavation of the channel was limited to 15 feet due to a high water table. If the source were removed or reduced to a size that results in a slower, low-level release of contaminants, it is possible that microbial degradation may eventually reduce contaminant levels in the aquifer. Otherwise, it may be necessary to continually pump the dewatering wells and carefully manage pumping of the aquifer to control contaminant migration.

3.3.4 STROUDSBURG, PENNSYLVANIA (Adaska and Cavalli, 1984; Berg, 1975; Campbell et al., 1979; Hem, 1970; Hult and Schoenberg, 1981; Lafornara et al., 1982; McManus, 1982; Schmidt, 1943; Unites and Houseman, 1982; Villaume, 1982; Villaume et al., 1983)

The Stroudsburg, Pennsylvania, town gas site, located next to Brodhead Creek, was in operation from the mid-1800's until 1939. During plant operations, the production byproducts (mainly byproduct tars) were disposed in open trenches and later in an underground injection well located onsite. After severe flooding in 1955 from Hurricane Diane, the Army Corps of Engineers modified the Brodhead Creek Channel. In 1980, the channel was deepened to prevent undercutting of the levee. At this time, black tarry globules were observed emanating from the base of the dike along the western bank of Brodhead Creek. The site was reported to the National Response Center, and the EPA initiated an investigative study. The study found that tar was present in the subsurface at the site; the tar was confined primarily to coarse clean gravels and had collected in a large depression underlain by a fine silty sand. The site was listed as a priority Superfund site and was the first one in the nation to receive emergency Superfund money. This case study illustrates the following:

- Site discovery through discharge into an adjacent stream

- Role of capillary pressure in controlling the movement of coal tar

- Recovery of free coal tar in the subsurface by pumping through a 30-inch gravel-packed well

- Increasing the efficiency of tar recovery by pumping the overlying groundwater to create a negative pressure and make the tar upwell

- Construction of a 648-foot bentonite-cement slurry cutoff wall on the streamside of the western levee to contain the contamination and prevent further seepage into the streambed

- Possible misinterpretation of historical data, leading to erroneous conclusions about the site, the nature of the contamination, and site remediation (see the next section).

3.3.4.1 Site History--

In light of the information collected during RTI's historical literature review, some of the previous site historical information about the Stroudsburg plant appears to be incorrect. This section compiles the site history and processes reported in the current literature, and Section 3.3.4.4 addresses the contradictions between this section and data collected by RTI.

The Stroudsburg coal gasification site is located in the borough of Stroudsburg, Pennsylvania, along the western bank of Brodhead Creek (Figure 69). The geology of the area consists of limestone bedrock overlain by a valley-fill-type deposit. The valley-fill-type deposit is made up of an underlying, well-sorted, fine, silty sand overlain by both stratified and unstratified, well-sorted, coarse glacial gravels. Inside the western levee is a single, steep-sided, gravel-filled depression, probably a kettle feature. The median depth to groundwater previous to any remediation was 10 feet, the hydraulic gradient was 0.015 foot per foot, and the groundwater generally flowed to the southwest at the rate of about 2 feet per day (Villaume et al., 1983) (also see Figure 70).

The plant was built in the mid-1800's and was in operation until 1939. The coal gas was manufactured by heating pulverized coal in a reaction vessel to drive off the volatiles. Superheated steam was then passed over the hot coal to produce a gas-steam mixture that was blown into a large holding tank. In this tank, the steam condensed, leaving the gas at the top and a liquid containing coal tar at the bottom. The major byproducts of this procedure
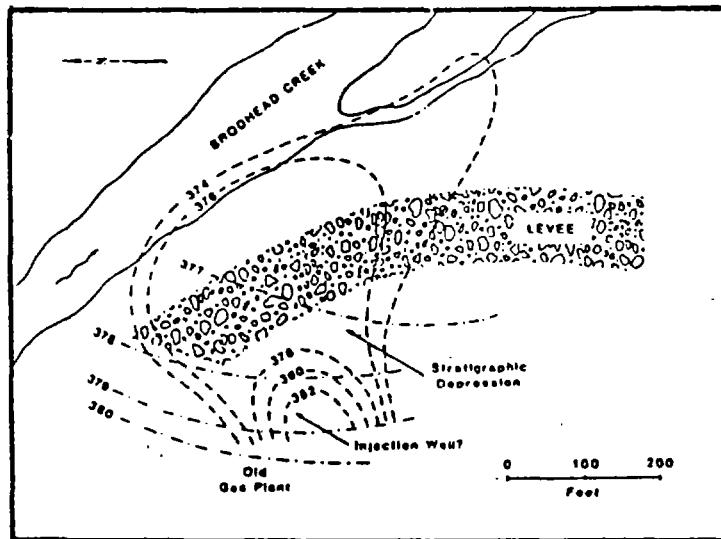
Figure 69. Stroudsburg site map with top-of-contamination (dash) and
groundwater (dot-dash) contours (in feet) shown. The
groundwater data are for June 12, 1981, prior to slurry wall
construction. Almost no free coal tar occurs beyond the
374-foot contour.

Source: Villaume et al., 1983.
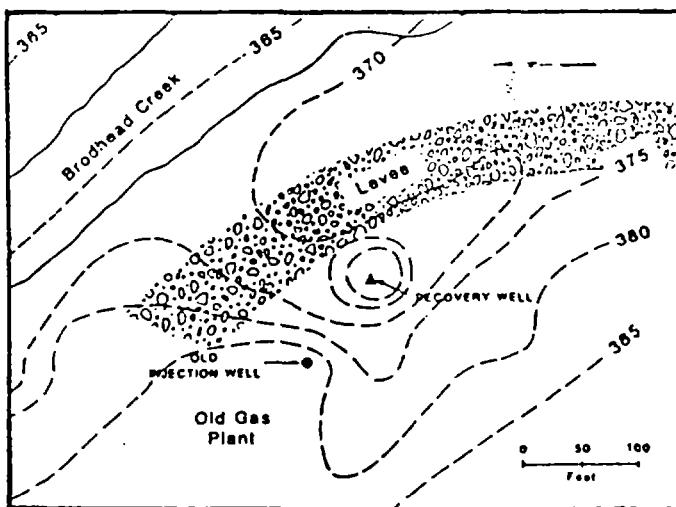
Figure 70. Top-of-sand contours (in feet) for the Stroudsburg
coal-tar contamination site.

Source: Villaume et al., 1983.

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were the coal tar left in the reaction vessel and the liquid containing coal tar in the holding tank.

Lafornara et al. (1982) estimate that as much as 16 million gallons of coal-tar could have been produced over the 100-year operating life of the Stroudsburg gas plant. Initially, the reaction vessel coal tar was disposed in open trenches that ran along the western edge of the site, eventually discharging into Brodhead creek, and the water and tar that collected in the holding tanks were blown down onto the ground next to the tanks (Lafornara et al., 1982). In the early 1900's, as coal-tar reprocessing technology developed, the coal-tar wastes were purified onsite to remove the commercially valuable constituents. The remaining wastes were disposed in an underground injection well onsite. This method of disposal continued until the plant shut down in 1939.

Brodhead Creek experienced severe flooding in 1955 as a result of Hurricane Diane. Between 1958 and 1960, the Army Corps of Engineers had to modify the stream channel by straightening several reaches of the stream and placing the channel within a floodway lined by riprapped levees. Within the next 20 years, the levees experienced significant downcutting, causing officials to deepen the riprap another 10 feet in 1980 to protect the levees from undercutting. During this work, coal tar was identified in the open trenches along the western bank of Brodhead Creek.

In 1981, the site was reported to the National Response Center. The EPA ordered all affected property owners to conduct a study to determine the extent of the contamination and a method of rectifying the damage. The Stroudsburg, Pennsylvania, site appears on the expanded list of 418 priority Superfund sites (which currently number 388) and was the first site in the nation to receive emergency Superfund money (Lafornara et al., 1982; McManus, 1982; Unites and Houseman, 1982; Villaume, 1982).

3.3.4.2 Extent of Contamination--

Based on the 1981 investigative studies, up to 1.8 million gallons of free coal tar is estimated to be distributed over an 8-acre area (Figure 50). The contamination extends vertically downward only to the top of the silty sand deposit. This deposit currently cannot be penetrated by the coal tar because of the extreme capillary-pressure forces that must be overcome. An

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accumulation of up to 35,000 gallons of nearly pure coal tar was estimated to occur in a single stratigraphic depression located just below the old gasification plant (Figure 51).

Capillary pressure (P) is defined by the equation:

$$P = 2\gamma \cos O / R$$

where

γ = the interfacial tension between the coal tar and water

O = the contact (wetting) angle formed by the coal tar against a solid surface in the presence of water

R = the radius of the water-filled pore that the coal tar is trying to enter.

The displacement of water by coal tar is most difficult when the capillary pressure is high, by definition indicating a high interfacial tension and low contact angle. Once the interfacial tension and contact angle are set, the pore size of the rock determines whether the coal tar can move into the media. Using Hobson's Formula (Berg, 1975), the critical height of coal tar needed to overcome the capillary pressure is calculated to be more than 10 meters. The maximum thickness of coal tar in the contaminated zone at any location onsite does not exceed 5.5 meters. The high capillary pressure and lack of critical column height of the coal tar explains why the silty-sand deposit serves as an effective barrier to the coal tar.

Hydrodynamic dispersion would be expected under onsite groundwater flow conditions. Shallow groundwater samples from throughout the site indicate the presence of dissolved contaminants. Partial analysis of the Stroudsburg coal tar is shown in Table 54. The polynuclear aromatics were generally detected at the ppb level or within the range of known aqueous solubilities of the individual chemical species involved. Table 55 shows that the principal control on the concentrations of these contaminants in the groundwater is their aqueous solubility and not their concentration in the coal tar. There is not enough data at this time to determine whether a relationship exists between solubility and distance of transport; however, there appears to be a rapid decrease in concentration just beyond the free coal-tar plume in the downgradient direction. The only contaminant detected at this point is naphthalene at less than 10 ppb.

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TABLE 54. PARTIAL ANALYSIS OF THE STROUDSBURG COAL TAR

Parameter	Value	Units
Naphthalene	3.60	%
Fluoranthene	3.20	%
Phenanthrene	2.30	%
Anthracene	2.30	%
Dimethyl naphthalenes	2.15	%
Trimethyl naphthalenes	1.78	%
Methyl phenanthrenes	1.50	%
Trimethyl benzene	1.30	%
Fluorene	0.98	%
Acenaphthylene	0.74	%
Acenaphthene	0.72	%
Pyrene	0.56	%
Benzo(a)anthracene	0.31	%
Chrysene	0.31	%
Benzo(a)pyrene	0.10	%
Other	7.84	%
Total	29.69	%
Acidity	0.62	mg KOH
pH	4.6	standard
Free carbon (Carbon I)	<0.01	%
Ash	0.00	%
Total carbon	90.77	%
Total hydrogen	8.12	%
Total nitrogen	0.17	%
Sulfur	0.65	%
Chloride	50.0	ppm
Ammonia	0.26	ppm
Cyanide	0.18	ppm
Iron	50.3	ppm
Copper	2.48	ppm
Manganese	2.11	ppm
Zinc	0.13	ppm
Nickel	0.19	ppm
Cadmium	0.01	ppm
Lead	0.5	ppm
Arsenic	12.7	ppm
Aluminum	22.4	ppm
Vanadium	1.6	ppm
Barium	0.5	ppm

SOURCE: Villaume et al., 1983.

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TABLE 55. ORGANIC CONTAMINANTS IN SHALLOW GROUNDWATER

Contaminant	Formula	Molecular weight	Aqueous solubility (mg/L)	Conc. in coal tar (mg/L)	Max. conc. in groundwater (mg/L)
Base-neutral fraction					
Naphthalene	$C_{10}H_8$	128.16	31.7[a]	36,000.	3.525
Acenaphthylene	$C_{12}H_8$	152.21	--	7,400.	0.428
Acenaphthene	$C_{12}H_{10}$	154.21	3.93[a]	7,200.	0.275
Fluorene	$C_{13}H_{10}$	166.21	1.98[a]	9,800.	0.218
*Anthracene	$C_{14}H_{10}$	178.22	0.073[a]	23,000.	0.085
*Phenanthrene	$C_{14}H_{10}$	178.22	1.29[a]	23,000.	0.330
Fluoranthene	$C_{16}H_{10}$	202.26	0.26[a]	32,000.	0.038
Pyrene	$C_{16}H_{10}$	202.24	0.135[a]	5,600.	0.063
*1,2-Benzoanthracene	$C_{18}H_{12}$	228.28	0.014[a]	3,100.	0.023
*Chrysene	$C_{18}H_{12}$	228.28	0.002[a]	3,100.	0.031
3,4-Benzopyrene	$C_{20}H_{12}$	252.30	0.0038[a]	1,000.	0.013
3,4-Benzofluoranthene	$C_{20}H_{10}$	252.32	0.0015[b]	370.	0.015
Benzo(ghi)perylene	$C_{22}H_{12}$	276.34	0.00026[a]	<250.	<0.010
Indeno(1,2,3-cd)pyrene	$C_{22}H_{12}$	276.34	0.0002[b]	<250.	<0.010
Volatile fraction					
Benzene	C_6H_6	78.11	1,780.[c]	--	0.241
Toluene	C_7H_8	92.13	583.[c]	--	0.960
Ethylbenzene	C_8H_{10}	106.16	159.[c]	--	1.193

SOURCE: Villaume et al., 1983.

Notes: *Indicates isomers that are indistinguishable by gas chromatography/mass spectroscopy (GC/MS).
[a]Data from Mackay and Shiu, 1977.
[b]Data from National Bureau of Standards (NBS), 1982.
[c]Data from McAuliffe, 1963.

Of the volatile organic fractions, only benzene, ethylbenzene, and toluene were found in the shallow groundwater. No acid fraction organics, most notably phenol, were found in the shallow groundwater. These materials were also detected in extremely low levels in the coal tar itself and were attributed by Villaume (1982) to either their original absence or to prolonged leaching by groundwater. Although the latter interpretation was supported by the Villaume, our investigation found that the plant operating at the site was a water-gas plant, which would produce tars with very low levels of tar acids (phenols, cresols, and xylenols), supporting the hypothesis that these compounds were not initially present in tars.

Elevated levels of certain metals and traces of cyanide were detected in the shallow groundwater at the site. In some of the sampled wells, aluminum, iron, manganese, and cyanide were detected at levels as high as 218, 460, 25.5, and 0.30 ppm, respectively. By comparison, these contaminants were measured in the raw tar at levels of 22.4, 50.3, 2.11 and 0.184 ppm, respectively. Sodium also was found in the groundwater at 26.2 ppm, but it was never analyzed in the tar. Cyanide, probably as either HCN or NH_4CN, is a byproduct of the gas cleanup and was typically removed from an iron salt (see Chapter 1). The source of the aluminum, on the other hand, is more problematical and, at such high concentrations, is probably present as a precipitated solid (Hem, 1970). The high sodium levels may be the result of sodium hydroxide usage at the plant. Even higher levels were found in the aquifers around the coal-tar distillation plant studied by Hult and Schoenberg (1981), who attributed them to such a source.

The toxic effects of tar seepage into Brodhead Creek were assessed using a macroinvertebrate and fish survey, tissue analysis, and in-situ toxicity testing of caged trout. These analyses revealed no apparent biological accumulation of the tar constituents. Also, tar contaminants were not found in the mixed stream flow as measured by gas chromatograph/mass spectrographic analysis.

3.3.4.3 Site Remediation--

In 1981, the State's investigative study recommended the construction of a slurry trench cutoff wall to contain the coal tar and prevent further migration into the streambed. Also recommended was the installation of a recovery

well system to collect tar wastes for removal. Because of the nature and extent of contamination, the State applied for and received funds for the remedial work under the Superfund program.

The cutoff wall was constructed of a bentonite-cement slurry. The completed wall is 648-feet long, 1-foot wide, and 17-feet deep. The wall extends down through the contaminated gravel stratum and 2 feet into the silty sand layer, which serves as an effective barrier to the coal tar. The upstream end of the wall is tied into a sheet-piling gate that is part of the existing flood dike, and the downstream end is tied into an impermeable cement-bentonite grout curtain (Adaska and Cavalli, 1984).

Initially, it was estimated that 35,000 gallons of free pumpable tar had accumulated in the single stratigraphic depression below the old coal gasification plant at Stroudsburg. This is tar that has displaced virtually all of the initial pore water in the gravel. Some tar also occurs above the pure coal tar in the depression, but it is associated with free water (water not held by strong capillary pressure forces), which could be picked up during any pumping operation.

To recover the full tar, a 30-inch gravel-packed well cluster was installed at the deepest point in the depression. It consists of four 6-inch wells screened only in the coal-tar layer. In the center is a single 4-inch monitoring well, which is screened over its entire length. Originally, product recovery was accomplished by pumping only the tar at a very slow rate. Using this method, approximately 100 gallons per day of nearly pure material were recovered, although this rate decreased drastically over time as the volume of tar in the vicinity of the well was depleted.

To increase the efficiency of the coal-tar recovery, the central monitoring well was modified by the installation of a packer at a depth between the static groundwater and static tar levels, thus isolating the lower part of the well. When groundwater is pumped from the uppermost layer, the resulting pressure reduction combined with the density difference between the two fluids causes the tar to upwell. If the tar is pumped at the same time as the overlying groundwater, the tar flows into the recovery well at an increased rate. Using this setup, a two-fold increase in the recovery rate

was achieved. To date, approximately 8,000 gallons of product with less than 1 percent water content has been recovered.

The initial estimate of total free coal-tar contamination at Stroudsburg is probably too high because it was based on an assumed 30 percent porosity for the contaminated gravels and on the assumption of complete coal-tar saturation. The majority of this porous material is probably only poorly saturated. This is evidenced by field observations that could not be explained at the time they were made, but they are consistent with the capillary pressure model presented by Villaume et al. (1983). Had this been understood earlier, justification for the expense of building the containment wall may have been questioned.

The amount of tar in the stratigraphic depression below the old gasification plant also was overestimated. The overestimation occurred because of well-screening practices that did not account for the characteristics of the various coal-tar phases and because these phases are virtually indistinguishable in split-spoon samples. Had the estimation been closer to the actual amount present, the recovery operation may not have been undertaken or may have been scaled down considerably.

Currently, the pumping operations have been stopped, with a total of 10,000 gallons of tar recovered. The site is still on the National Priority List (NPL) (ranked at 388), and it is uncertain whether further cleanup action will be required.

3.3.4.4 New Historical Data on Stroudsburg--

During RTI's historical literature review of the town gas industry, several items were uncovered that will result in reevaluations of previously reported information about the Stroudsburg site. These observations concern (1) the gas production processes used at the plant, (2) the previously reported method of waste disposal (injection well), (3) the source of the tar contamination, and (4) the nature of tar products from the site.

The Stroudsburg site has always been reported as a coal-gas production site. Table 56 shows the gas production at the site as compiled from Brown's Directory, which lists the gas production process as oil and steam (1891 to 1894), Van Syckel oil process (1894 to 1904), and Lowe carbureted water gas (1912 to 1952). The process specifics for the oil and steam gas production

TABLE 68. GAS PRODUCTION AT STROUDSBURG, PENNSYLVANIA

Year	Process	Production (10^6 ft^3/yr)		Population	Customers	Other data
1889	?			2,000		Stroudsburgh Gas and Light Co.
1892	Oil and steam	1	20 CP	4,200		Monroe County Gas Co.
1894	Van Syckel	3	22 CP	5,000		
1900	Van Syckel	4.5	22 CP	7,000		
1902	Van Syckel (oil gas)	6	22 CP	7,000	415	
1904	Van Syckel (oil gas)	7	22 CP	7,200	440	Citizens Gas Co. of Stroudsburgh.
1906	Carbureted water gas	6	22 CP	7,000	625	
1908	Carbureted water gas	7	22 CP	8,000	625	Citizens Gas & Electric Co. of Stroudsburgh.
1910	Lowe (Gas Mach. Co.)	8	22 CP	5,000	646	51,000 holder capacity.
1912	Lowe (Gas Mach. Co.)	10	22 CP	10,000	700	
1914	Lowe (Gas Mach. Co.)	12	22 CP	10,000	900	70,000 holder capacity.
1916	Lowe (Gas Mach. Co.)	12	22 CP	10,000	1,000	
1920	Lowe (Gas Mach. Co.)	1?	20 CP	10,000	1,200	
1924	Lowe (Gas Mach. Co.)	34	18 CP 530 Btu	12,000	1,526	170,000 holder capacity. Used for gas production: 110,800 gallons gas oil.
1928	Carbureted water gas	54.9	530 Btu	10,000	2,000	Used for gas production: 906 tons grate coal 139,793 gallons gas oil 9.4 tons coke as generator fuel 487 tons anthracite as boiler fuel.

(continued)

TABLE 58 (continued)

Year	Process	Production (10^6 ft^3/yr)		Population	Customers	Other data
1932	Carbureted water gas	63.0	525 Btu	12,038	2,039	Citizens Gas Co. (Subsidiary of Central Gas & Electric Co.). Used for gas production: 171,872 gallons gas oil 683 tons boiler fuel.
1936	Carbureted water gas	61.4	522 Btu	12,050	2,715	Used for gas production: 153,578 gallons gas oil used 96 tons of bituminous coal as generator fuel. Boiler fuel: 753 tons bituminous coal 1 ton coke 14,707 gallons tar.
1940	Carbureted water gas	62.7	518 Btu	13,675	2,400	Used for gas production: 145,260 gallons gas oil 661 tons bituminous coal used in boilers 1,166 tons bituminous coal used in generators.
1944	Carbureted water gas	65.6	520 Btu	13,750	2,974	Used for gas production: 153,632 gallons gas oil 1,217 tons coal used in generators. Boiler fuel: 722 tons bituminous coal 8 tons anthracite coal.
1948	Carbureted water gas	94.8	522 Btu			Used for gas production: 276,494 gallons gas oil 1,798 tons anthracite used in generators 1,183 tons anthracite used in boilers.
1952	Carbureted water gas					
1956	Natural gas					

SOURCE: Brown's Directory.

Lowe = Carbureted water gas.
Holder capacity in ft^3.
CP = Candle power.
Btu = Btu/ft^3.

(and the Van Syckel oil-gas process) were not found during this study, but processes of this type generally sprayed oil and steam into an externally heated retort. The oil cracked into lighter gaseous hydrocarbons, and the steam reacted with carbon to produce CO and H_2. The Lowe carbureted water-gas process is described in Section 1.2.3 of this report. There is no indication in Brown's that coal carbonization ever occurred at the Stroudsburg site. Brown's also shows the Stroudsburg plant as operating into the 1950's, with natural gas being installed sometime between 1952 and 1956. According to these data, the plant was operated primarily as a carbureted water-gas plant.

It has been reported that an injection well was used to dispose of waste tars at Stroudsburg. When tar was produced and separated from town gases, it was usually stored in an underground tank until sold or used. These tanks were called "tar wells," in that tar was placed into the tanks and pumped out as if one were removing water from a well. The tar wells were labeled as "tar well" on plans and maps of the sites. They were also sometimes completely underground, with only a pipe visible from the surface for removal and filling of tars from the tar well. Unless the notation on the site map was clearly labeled as a tar disposal well or an injection well, it is possible that it was actually a tar storage well.

There are two other possible sources that could have caused the subsurface tar contamination. Leaks of tar and oils from carbureted water-gas plants were very common. Underground tar wells (for tar storage) were often constructed of masonry and leaked. Underground liquid storage tanks were sometimes constructed of wood. Tars were frequently placed in the gasholder for storage (gas sometimes blew around the tar-water seal for the holder, blowing tar out of the holder and onto the ground). The bottom of the gasholder was frequently below the groundlevel and also was prone to leaks. Underground pipes also leaked oil and tar materials into the ground.

The second likely source of the tar contamination is the disposal trench described by Lafornara et al. (1982). The tars and emulsions draining into the ground from the trench would flow downward until stopped, and they would have accumulated in the area where the subsurface tars were located. The amount of tar produced by the plant in 1936 was 15,000 gallons (this is about 10 percent of the gas oil used that year). Thus, finding 10,000 gallons of

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free tar underground (and maybe 5,000 to 20,000 gallons of tar [this estimate is a guess) left in the ground] is approximately 1 to 2 years of tar production during this period.

The Stroudsburg tar (as described by Villaume, 1982) is a carbureted water-gas tar, not a coal tar. It is only slightly more dense than water (P = 1.02 g/cm^3), contains very little nitrogen (0.17 percent), has no tar acids, and has a viscosity of 19 cp (45 °F). Coal tar would be denser (1.1 to 1.2 g/cm^3), contain more nitrogen, have some phenols, and be more viscous. The density of the tar is so close to that of water that it would be very difficult to separate a tar-water emulsion. Lafornara states that "Treatability studies performed on a coal tar/water emulsion pumped from the backwater revealed that no cost-effective method could be found to separate the emulsion and treat the water." This is precisely why the water gas tar was originally disposed during plant operation. Such an emulsion would frequently be disposed. The distillation curve (90 percent at 662 °F) shows that the tar did not contain very much heavier boiling-organics, which probably indicates they were removed in the washbox and not disposed with this tar.

If this tar could have been successfully recovered at the plant, it either would have been burned or added to the carburetion oils. The water-gas plant bought large quantities of oil that were poorer carburetion oils than was the recovered Stroudsburg tar.

3.3.5 Plattsburgh, New York (Thompson et al., 1983)

The coal-gas and carbureted water-gas plant in Plattsburgh, New York, was in operation from 1896 to 1957. The plant was located cn 11 acres of land on the south bank of the Saranac River. Byproduct tar was disposed in unlined ponds just above the river. Over several decades, ccal tar could be periodically observed on the south side of the riverbed as globules and as a film along the riverbank. This case study illustrates the following:

- Site discovery through discharge into an adjacent waterbody

- Coal-tar migration during active disposal by slow downward movement through subsurface soils along a dense till layer and from occasional overflow of the ponds during heavy rainfall

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- Various influences on contaminant migration including seasonal groundwater fluctuations causing changes in pore pressure, increased temperatures during summers causing coal tar to become more mobile due to decreased surface tension and viscosity, and increased river flow causing a flushing of the contaminants from the soil

- Remediation by containing the contaminants onsite (Two containment structures include cells built of a soil-bentonite slurry wall keyed into an underlying, low-permeability till layer and capped with a 36-mil Hypalon liner covered with 15 centimeters of sand, topsoiled, and seeded, and a second cement-bentonite wall built along the riverfront to prevent migration of contaminant not contained within the soil-bentonite cells.)

- Remediation with a groundwater collection system being built to collect waters draining from the uncontained contaminated site (These waters will be pumped to water treatment equipment, treated, and discharged into the Saranac River.)

- Use of the 4 acres of reclaims that lie along the river as part of the City of Plattsburgh's riverfront park system.

3.3.5.1 Site History--

A coal and carbureted water-gas plant was operated within the city limits of Plattsburgh, New York, from 1896 to 1957. The New York State Electric and Gas Corporation (NYSEG) purchased the site and coal gasification plant from Eastern New York Electric and Gas Corporation in 1929. The plant was located on 11 acres of land on the south bank of the Saranac River. The topography falls gently in steps from an approximate elevation of 125 to 130 feet mean sea level (MSL) along the south edge of the site to 102 to 107 feet MSL along the Saranac riverbank. Other than a narrow band of trees and bushes adjacent to the river, most of the site has been cleared and filled. Two structures that cross the site are a 24-inch diameter concrete sanitary sewer and an active transmission line (owned by the Plattsburgh Municipal Lighting District) (see Figure 71).

This land consists of two parcels. The larger parcel (approximately 9 acres owned by NYSEG) lies uphill to the south and is the old site of the gas plant. The smaller parcel (approximately 2 acres) is a long narrow strip of land that fronts the Saranac River just downhill (to the north) of the NYSEG gas plant. This parcel was given to the City of Plattsburgh in 1981 by NYSEG

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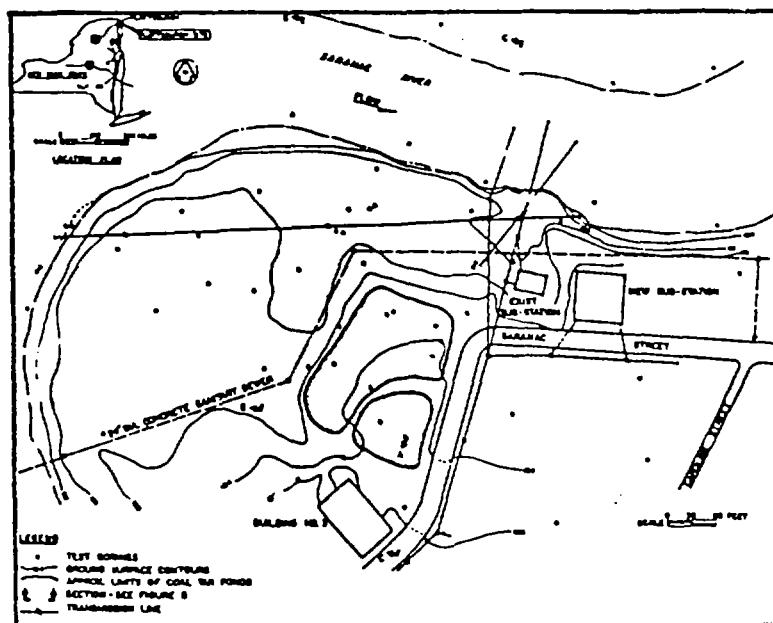
Figure 71. Plattsburgh, New York, general site plan.

Source: Thompson et al., 1983.

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as a contribution to the city's long-range plan for recreational development of the Saranac River inside the city.

Table 57 is a list of the gas productions as recorded in Brown's Directory. This plant produced primarily water gas over its history, although notations in 1906, 1924, and 1936 indicated that coal was also carbonized at the plant.

Byproduct tar and condensate from the gas production was disposed in unlined ponds on the NYSEG property just uphill from the Saranac River. No records of the amount and times of tar disposal into the unlined ponds could be found. After the plant shut down in 1957, the ponds were filled with random material and covered with layers of cinders and ash. Over the years, this coal tar migrated downhill across the property now owned by the city and into the Saranac River. This migration occurred via two routes: by slow downward movement through subsurface soils, and from occasional overflow of the ponds during periods of heavy rainfall. Tar can be observed periodically on the south side of the riverbed both as globules of coal tar and as film along the riverbank. This problem, which has been in existence for some years, has been attributed to seepage of the tar from the previously existing tar-ponding areas on the site.

To address the problem, NYSEG conducted a geotechnical investigation during the summer of 1979. This fieldwork and laboratory testing, together with preliminary, alternative strategies for site remediation, were completed in early 1980. Following review of this work, a supplementary program of soil boring and testing was undertaken in November 1980. Actual site remediation occurred between September 1981 and September 1982. Remediation activities were coordinated with the City of Plattsburgh's long-range plans for recreational development of the Saranac riverbank, including the parcel given to the city by NYSEG. Construction plans include building scenic overlooks for fishing during trout season and a pedestrian bridge to cross the river.

3.3.5.2 Extent of Contamination--

To define the site geology, hydrology, and area of contamination, a total of 53 boreholes were drilled across the site. In addition to these boreholes, three test pits were excavated to obtain bulk samples of the tar and soil for

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TABLE 57. GAS PRODUCTION AT PLATTSBURGH, NEW YORK

Year	Process	Production (10^6 ft^3/yr)		Population	Customers	Other data
1889	Lowe	?	25 CP	8,000		
1891	Lowe	4	25 CP	8,000		Plattsburgh Light, Heat & Power Co.
1892	Lowe	4	25 CP	8,000		
1894	Lowe	4	22-24 CP	8,000		
1900	Lowe	8.25	22-24 CP	10,000	413	
1902	Lowe	13	22-24 CP	10,000	502	
1904	Lowe	15	22-24 CP	10,000	387	
1906	Lowe and coal	20	20-24 CP	10,000	765	
1908	Lowe	21	20-24 CP	10,000	937	
1910	Lowe	24	21 CP 652 Btu	11,000	1,040	
1912	Lowe	28.2	20 CP 625 Btu	11,000	1,060	
1914	Lowe	33.1	22 CP 618 Btu	11,000	1,106	
1915	Lowe	38.4	20 CP 618 Btu	10,138	1,219	
1920	Lowe (UGI)	46.5	19 CP 620 Btu	10,138	1,451	
1924	Lowe (UGI)	53.2	604 Btu	10,138	1,692	Used for gas production: 1,541 tons coal carbonized 197,920 gallons gas oil used.
1928	Lowe (UGI)	63.5	540 Btu	12,138	1,853	Eastern NY Electric & Gas Co. Used for gas production: 276,554 gallons gas oil used 635 tons bituminous coal in boilers.

(continued)

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TABLE 57 (continued)

Year	Process	Production (10^6 ft^3/yr)		Population	Customers	Other data
1930	Lowe (UGI)	71	539 Btu	13,333	1,862	Used for gas production: 287,926 gallons gas oil 858 tons bituminous coal for generators 677 tons bituminous coal for boilers.
1932	Lowe (UGI) Production after 1932 included with Ithaca, NY					New York State Electric & Gas Co.
1936	Water gas and coal gas	Max. sent out from plant: 232,400 ft^3/day				
1940	Water gas					
1944	Water gas					
1948	Water gas					

SOURCE: Brown's Directory.

Lowe = Carbureted water gas.
Holder capacity in ft^3.
CP = Candle power.
Btu = Btu/ft^3.
UGI = United Gas Improvement Co.

laboratory testing. Nineteen standpipe piezometers were installed to monitor groundwater levels across the site.

The borings indicated the presence of an extremely dense till underlying the entire site. This till consists of silt and fine sand intermixed with medium- to coarse-gr·ined sand and gravel. The till appears to have served as a barrier over the years, halting vertical migration of the coal tar on the site. No tar was observed below this till anywhere on the site.

However, in the sandy soil and fill layers above this till, tar contamination was found over most of the site. In the area of the original tar ponds, contaminated soils were found as deep as 4 meters. From this region of maximum soil contamination, the thickness of the contaminated soil gradually lessened toward the NYSEG property boundaries except for a layer of contamination extending across the city's parcel to the north and into the riverbed of the Saranac River. The data from the borings indicated that the subsurface movement of tar from the ponds had been downward through the permeable sands and gravels and then laterally along the top of the till toward the river. No tar was observed below the till layer (Thompson et al., 1983).

A laboratory testing program was undertaken to further characterize the contamination. Tar content (percent dry weight) in contaminated soils was found to be as high as 9.6 percent with an average content of 1.5 percent. Tests to determine total leachable salts in the soil/coal tar showed low concentrations of metals (although leachable arsenic was reported at 2 and 3 ppm and lead at 0.9 and 1 ppm in two samples). Determination of total leachable salts in tar reported for three samples showed high chemical oxygen demand (COD) and total organic carbon (TOC) at 850, 900, and 935 ppm. Leachable phenol was as high as 4 ppm in a tar sample taken from the Saranac River (Thompson et al., 1983).

The investigations determined that tar migration has decreased exponentially since disposal of tars was halted in 1957. When active disposal was in progress, the sands, silts, and gravels beneath the ponds became saturated with tar. The higher viscosity of the tar and its immiscible properties allowed the tar to migrate in density currents as a separate phase from the groundwater. With continued disposal, movement of the tar occurred relatively rapidly downgradient along the top of the till layer into the river. Once the

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tar disposal stopped, the rate of migration gradually decreased. Thompson et al. (1983) believe that the majority of the tar currently onsite is being retained within the pores and matrix structure of the soil grains by capillary forces, and that the mechanism causing the tar migration today is different from that when the ponds were in operation. Although difficult to quantify, the mechanism causing tar migration today is most likely influenced by one or more factors, including seasonal groundwater fluctuations causing changes in pore-water pressure, increased ground and groundwater temperature during summer causing the tar to become more mobile due to decreased surface tension and viscosity, and increased riverflow causing a flushing of the contaminants from the soil.

3.3.5.3 Site Remediation--

Site remediation occurred in two phases. The Phase I Project focused on arresting the subsurface migration of coal tar away from the area of the original disposal ponds. The Phase II Project addressed the cleanup of the Saranac River and the city-owned property to the north.

Phase I began in the fall of 1981 with the installation of a soil-bentonite slurry wall around the main tar pond area (735 feet in perimeter). This wall was keyed into the underlying impervious till that was 4 to 6 meters below grade in the main-pond area. This main-pond area was then capped with a temporary 20-mil polyvinyl chloride (PVC) liner. It was estimated that approximately 80 percent of the onsite coal tar was encapsulated within this containment cell. A well was placed within the cell to monitor the effectiveness of isolation.

Phase II remediation activities began in June 1982 with the installation of a temporary, portable fabric cofferdam in the Saranac River. Behind this cofferdam, tar contamination in the riverbed was excavated in the dry. Water was pumped from the area of excavation into a triple-compartment settlement tank before being discharged back into the river. Riverbed cleanup was performed in two stages moving from upstream to downstream.

The temporary PVC liner that had been placed as a cap over the previously constructed containment cell was perforated, and the contaminated material excavated from the river was placed on top. Additional contaminated materials were placed in an area just to the southwest of the original containment cell.

Later, this additional area was also surrounded with a soil-bentonite slurry wall and thus represented an enlargement (almost a doubling) of the size of the original containment cell.

After excavation of all visible contamination in the riverbed and along the riverbank, the riverbed and bank were reestablished to grade with imported clean fill. To prevent continued migration of remaining uncontained tar into the riverbed area, a cement-bentonite cutoff wall was constructed through the clean fill for approximately 213 meters along the riverbank. A cement-bentonite wall was used in this area (instead of soil-bentonite wall used previously on the NYSEG property) because a higher strength wall was considered necessary to meet the city's plans for recreational development of this area.

To intercept drainage of groundwater from the uphill area above the cement-bentonite wall paralleling the river, a groundwater collection system was installed. This system consists of a 15-centimeter perforated drainpipe 0.6 meters below grade and 3 meters upgradient of the cement-bentonite wall. This drainpipe discharges into a precast manhole at the midpoint of the line. Water collected by this system is pumped back uphill to water treatment equipment located in the vicinity of the coal-tar containment cell. Treated groundwater has been discharged into the Saranac River since September 1982.

After grading the contaminated soil in the areas inside the walls of the containment cells, the cells were permanently capped with a 36-mil Hypalon liner. This liner was then covered with 15 centimeters of sand, topsoiled, and seeded. This site work was completed in September 1982.

Because so much tar contamination has simply been contained onsite, future use of both the NYSEG and City of Plattsburgh parcels will have to be carefully guarded. Specifically, certain restrictions to onsite development have been mandated by the NYSDEC, and other restrictions have been suggested by NYSEG, who will remain responsible for maintaining the slurry walls, containment cell, groundwater collection and treatment system, and monitoring network on both parcels. These restrictions are:

- Sale of the lands on which the containment cell was constructed is prohibited by NYSDEC.

- No structures or other activities that could result in rupture to the Hypalon membrane may be placed or performed on the containment cell.

- All trees or shrubs will be maintained at a distance from the slurry walls such that their mature drip line will not intersect the slurry walls.

- All construction on or near the cement-bentonite partial cutoff wall and/or groundwater collection system must have prior engineering approval cf NYSEG.

3.3.6   Seattle, Washington (Cole, 1972a and b; Cole and Machno, 1971; Drew, 1984; Haag, 1971; Royer, 1984; Mayor's Committee on Gas Works Park, 1984; Orth, 1984; Steinbrueck, 1971)

The Seattle Gas Works plant was in operation for approximately 50 years. A large portion of the waste byproducts were disposed offsite, but large quantities of lampblack were disposed onsite, building up the shoreline into the adjacent Lake Union in Seattle, Washington. This case study illustrates the following:

- Site discovery through redevelopment as a park

- Large stockpiling of lampblack filling in Lake Union

- Conversion of the site into a public park by partial building demolition, composting of contaminated soils in preparation for planting, without removal of onsite contaminants

- Closing of park

- Present ongoing investigations to determine whether further remediation is necessary.

3.3.6.1   Site History--

The Gas Works Park is located on a point projecting into Lake Union in Seattle, Washington. The park occupies about 20.5 acres, which includes some 1,900 linear feet of waterfront. The surrounding area is mainly industrial property.

The Lake Union site known as Brown's Point, once a popular spot for picnicking, was developed in 1906 by the Seattle Lighting Company as a gas plant. The location of the plant on Lake Union made it ideal for the barge delivery of local and imported coal (and later, oil) for gas production. Eventually,

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the site became known as the Gas Company Peninsula, built by a slow process of filling in Lake Union with cinders, unusable coal and coke, and gas production wastes. The Seattle Lighting Company became the Seattle Gas Company in 1930 and eventually was made part of the Washington Natural Gas Company (WNG).

The original plant on Lake Union produced illuminating, heating and cooking, and industrial gases for the growing Seattle community. Coke ovens were operated, and retort gas and carbureted water gas were produced. During the mid-1930's, six water-gas sets were in operation with a total daily capacity of 6,600,000 ft^3 of gas (Steinbrueck, 1971). The byproducts of the gas plant operations were ammonia, light oils (benzene, toluene, xylenes), various other hydrocarbons, and tar, which was refined into creosote. Tar and creosote produced by the Seattle Gas Company were delivered to the American Tar Company, which was located adjacent to the Seattle Gas Company until about 1920. The tar company refined the coal tar into various grades of tars and pitches using steam distillation (Orth, 1984).

In 1937, oil replaced coal carbonization as the basis for gas production. The plant continued to produce water gas.

Table 58 shows the gas production and byproducts from Seattle as compiled from Brown's Directory. Oil-gas tars contained more asphaltene-type compounds than did the coal tars produced earlier and were not suitable for the products derived from the coal tars. Thus, the oil-gas tars were generally used as fuel for steam production. The tar emulsion from the Jones crackers was over 90 percent water and had to be concentrated before it could be burned. Naphthalene and related aromatic oils were collected in the condensation from this process. The naphthalene was sometimes combined with creosote oils and sold, but it often was simply dumped offsite (Orth, 1984).

The lampblack from the oil-gas cracking operation was dried for briquetting and used to replace coke in the water-gas sets. However, the briquets would often break during the firing. As a result, there was considerable waste. The lampblack production far exceeded the use, and the excess was piled next to the lake. The pile of lampblack grew to nearly 100 feet high and covered several acres (Orth, 1984). There were frequent complaints of odors from the plant and from the wind dispersal of the lampblack.

TABLE 58. GAS PRODUCTION AT SEATTLE, WASHINGTON

Year	Process	Production (10^5 ft^3/yr)		Population	Customers	Other data
1889	Coal			25,000		
1893	Coal			40,000		Seattle Gas Electric, Light and Motor Co.
1892	Coal			40,000		
1894	Coal	70		60,000		
1900	Coal	85	18 CP	50,000	4,500	
1902	Coal and Lowe	125	18 CP	50,000	5,667	
1904	Coal and Lowe	175	20 CP	60,000	7,500	Seattle Gas & Elec. Co.
	Coal and Lowe	140	25 CP	6,000	3,60?	Citizens Light & Power Co.
1906	Coal and Lowe	350	20 CP	100,000	13,100	Seattle Lighting Co. (consolidation of the 1904 companies listed)
1908	Coal and Lowe	500	20 CP	100,000	21,000	
1910	Coal-40% Lowe-60%	776	20 CP	220,000	36,500	
1912	Coal-40% Lowe-60%	675	20 CP	180,000	27,600	
1914	Coal-34.5% Lowe-65.5%	968	19 CP 600 Btu	250,000	41,539	
1916	Coal	783	600 Btu	250,000	45,419	Byproducts produced: 334,000 gallons tar 566,000 pounds ammonium sulfate 531,700 gallons ammonia liquor.
	Lowe	361	600 Btu			
	Coke ovens	495	675 Btu			
1920	Coal	877	607 Btu	250,000	53,539	Byproducts produced: 337,000 gallons tar 152,000 gallons ammonia liquor 33,500 tons coke.
	Lowe	609				
	Coke oven	46.1				

(continued)

TABLE 58 (continued)

Year	Process	Production (10^6 ft^3/yr)	Population	Customers		Other data
1924	Coal Lowe Coke oven	142 1,109 505 Btu 523	320,000	55,873		Used for gas production: 70,333 tons coal 2,565,614 gallons gas oil. Byproducts produced: 563,000 gallons oil tar 541,562 gallons coal tar 457,312 lbs ammonia 31,081 tons coke sold.
1930	Coal and Lowe Coke oven	1,279 505 Btu 875	350,000	58,354		Used for gas production: 64,928 tons coal carbonized 2,970,197 gallons gas oil 25,988 tons coke used in generators 3,533 tons coke used in boilers 13,986 tons bituminous coal used in boilers. Byproducts produced: 41,864 tons coke 955,001 gallons tar 636,030 lbs ammonia.
1934	Water gas Coal gas	1,400 519 Btu 840	390,630	45,480		Used for gas production: 55,735 tons coal carbonized 5,532,000 gallons gas oil 28,491 tons coke used in generators 6,124 tons used in boilers. Byproducts produced: 36,228 tons coke.
1936	Water gas Coke ovens	1,253 507 Btu 570	390,630	67,730		Used for gas production: 47,288 tons coal carbonized 201,721 gallons diesel oil 5,514,407 gallons heavy oils 8,535 tons coke used in generators 3,152 tons coke used in boilers 110,451 gallons heavy oil used in boilers.

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TABLE 58   (continued)

Year	Process	Production (10^6 ft^3/yr)		Population	Customers	Other data
1936 (continued)						1,911,281 gallons water-gas tar used in boilers. Byproducts produced: 28,733 tons coke 358,741 gallons coke oven tar.
1940	Water gas Oil gas	810 1,663	459 Btu 517 Btu	390,630	69,625	Used for gas production: 198,452 gallons diesel oil 17,228,824 gallons heavy oil 1,834 tons coke used in generators. Boiler fuel: 2,323,705 gallons heavy oil 751,394 gallons water-gas tar 807,326 gallons oil tar. Byproducts produced: 20,409 tons lampblack 630,612 gallons benzol 1,787,341 gallons tar 3,349 pounds napthalene.
1944	Water gas Oil gas	1,220 1,749	464 Btu 518 Btu	390,630	69,533	Used for gas production: 99,491 gallons diesel oil 17,962,000 gallons heavy oil. Generator fuel: 2,340 tons coke 16,830 tons petroleum briquets. Boiler fuel: 2,810,636 gallons heavy oil 1,475,672 gallons water-gas tar 777,986 gallons oil tar. Byproducts produced: 22,815 tons lampblack 392,084 gallons benzol 64,499 gallons toluol 5,778 gallons xylol 2,089,295 gallons tar.

TABLE 58 (continued)

Year	Process	Production (10^6 ft^3/yr)		Population	Customers	Other data
1948	Water gas	285	475 Btu			Used for gas production:
	Oil gas	3,728	502 Btu			626,352 gallons diesel oil
	Propane-air	42.7	737 Btu			31,559,799 gallons heavy oil.
						Generator fuel:
						859 tons coke
						5,826 tons petroleum coke
						briquets
						540 tons coal.
						Boiler fuel:
						5,050,128 gallons heavy oil
						540,345 gallons water-gas tar
						1,310,208 gallons oil-gas
						tar.
						Byproducts produced:
						47,818 tons lampblack
						1,040,551 gallons benzol
						56,832 gallons toluol
						6,680 gallons xylol
						1,733,156 gallons tar.
1952	Water gas	159	448 Btu			Used for gas production:
	Oil gas	3,971	521 Btu			207,593 gallons diesel oil
	Propane-air	10.2	758 Btu			33,445,740 gallons heavy oil.
						Generator fuel:
						3,592 tons petroleum
						briquets.
						Byproducts produced:
						34,342 tons lampblack
						1,111,883 gallons benzol
						132,496 gallons toluol
						3,856 gallons xylol
						3,607,000 gallons tar
						62,216 gallons crude solvent
						naptha.
1956	Natural gas					Seattle Gas Co. merged with
						Washington Gas & Electric
						Co. (Tacoma, WA) to form
						Washington Natural Gas Co.

SOURCE: Brown's Directory.

Lowe = Carbureted water gas.
Holder capacity in ft^3.
CP = Candle power.
Btu = Btu/ft^3.

The company continued to produce gas until 1956, when a natural gas pipe-line was extended to Seattle. After that, WNG used the site for storage and other activities. During the plant's operation, the shoreline on the penin-sula had been extended some 24 meters into Lake Union. Eventually, the site was almost flat down to the lake's edge where there was a 2.4 meter drop.

In 1962, the City of Seattle purchased the peninsula for development as a public park. A bond resolution passed in 1968, providing funds for park development, and planning for the park was initiated. The city hired a land-scape architect, Mr. Richard Haag, to propose a master plan for the park. After a study of the site, Haag determined that traditional park development would be impractical and proposed a controversial plan that allowed for the restoration and reuse of some of the gasworks structures. The plan for the site demolition (to be done by WNG in 1971 under the 1962 purchase agreement) called for leaving six generator towers, the pre-cooler towers, a boiler house, and an exhauster building. Haag concluded that it would not be pos-sible to remove all of the underground piping and existing soil from the site, nor to cover the entire site sufficiently to permit the growth of large trees essential to a more traditional park design. Despite the controversy over allowing the former plant structures to remain, the city council finally approved Haag's plan in 1972.

3.3.6.2 Extent of the Contamination--

Some 50 years of heavy industrial use at a time when there was little concern for environmental contamination had left the site on Lake Union heav-ily contaminated with residues from production, spills, waste materials, and air pollution fallout. Haag, the landscape architect, expressed concern for the ability of the site to support vegetation, noting that there was no "natu-ral" soil on the site. He described the condition of the soil as a sterile layer cake of hydrocarbon contamination that supports no vegetation (Haag, 1971). Studies were undertaken by the Seattle Engineering Department and by Dr. Dale Cole and Peter Machno of the University of Washington to characterize drainage patterns and soil conditions at the site.

3.3.6.3 Site Remediation--

The description of the remediation activities below is summarized from information contained in a document made available by the site manager in the

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U.S. EPA Regional Office. The document is not commonly available but was probably prepared in 1984.

After the removal of the above-ground structures by WNG in 1971, considerable site preparation work was still needed. The primary intent was to stockpile and/or bury onsite much of the excavated material and demolition rubble. The stockpiling was in the central portion of the site. Portions of the stockpile were later buried onsite. Several existing structures considered potential safety hazards were removed. WNG was required to purge certain pipes in 1973.

The mound area in the southwest portion of the site consisted of excavation materials from offsite. This fill had been brought to the site during the 1960's and early 1970's. It was thought at one time that this fill material could be used to cover the entire site following the demolition of the above-ground structures. However, the "Great Mound" became a major element of the master plan for the park, and it was cleared, grassed, and opened to the public for the purpose of viewing the ongoing park development.

Work contracted by the Parks Department included the following tasks:

- Demolition and burial in the northwest section of the rubble from 13 concrete purifiers that were located just east of the tower area

- Removal and stockpiling of the contents of the purifiers (i.e., woodchips coated with iron oxide and residue from the purification process)

- Removal and burial in the northwest section of the concrete slab remaining from the 2 million ft^3 storage holder

- Demolition of remaining concrete foundations and piping

- Excavation and removal or stockpiling onsite of approximately 20,000 to 30,000 yd^3 of badly contaminated soils

- Regrading of demolition areas to match the surrounding ground level.

In the process of removing contaminated material and burying rubble and debris, there was concern of increased pollution to surrounding areas, particularly Lake Union. Of particular concern was the excavation of the contaminated soil in the southwest area. The contract specifications cautioned the

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contractor responsible for this work of the conditions there. The contract stated, "Excavating oil-gas contaminated material at the southwest property edge shall be performed with extreme care. This excavation extends to the lake level and shall commence 30 feet or more inland from the water's edge. Demolition work and pipe removal shall be completed prior to any excavating of this 30 foot wide levee. When the inland area is excavated, filled and/or graded to the proposed grade the levee at the lake's edge shall be removed."

One part of the site preparation work involved efforts to improve growing conditions by an application of a compost-like mixture containing dewatered-sludge cake as the primary ingredient. The mixture was applied over approximately 10 to 12 acres of the southerly half of the site (about 100 tons per acre, wet) and then worked into the top 18 to 24 inches by periodic plowing. Sawdust and leaves were also applied and worked into the surface soil. The surface was reworked, fertilized, and sown with a cover crop of grass about 2 weeks after the compost treatment. The first crop was plowed under, and the area was finally rehydroseeded.

The actual park improvements were undertaken upon completion of the site preparation work. Phase I of the park development consisted of the following actions:

- Renovation of the former boiler house for use as an indoor/outdoor picnic shelter

- Renovation of the former exhaust building for use as a "Play Barn"

- Creation of a grassed picnic "Bowl" projecting to the water's edge

- Construction of paths

- Further development of an existing 170-car parking area

- Deter access to the towers and remove miscellaneous structures

- Regrade mound and hydroseed

- Plant trees and shrubs and provide sod in one small section of the picnic area.

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The work delineated above was completed, and the official park opening was held during the summer of 1976. Additional improvements were completed in 1978. Plans for further improvements were being finalized when the U.S. EPA began an investigation of contamination at the site.

Soil testing during the park development was directed primarily at horticultural aspects of the design. The park did not include any significant amounts of fill. Cuts were made primarily in the southeast quadrant and between the mound and tower areas. Considerable soil was removed from the site, part of which was known to contain arsenic. No work was undertaken in the water areas surrounding the site. According to the Gas Works Park history:

> It appears that the development was directed at reusing the site in what was felt at the time to be an environmentally sensitive manner. Both the general design concept and the budget were important factors in the decisions that were made. The major controversial issues centered on the retention and reuse of structures associated with the former gas plant. Most of the discussion concerning the levels of pollution centered on what would and would not grow on the site. Public health was an issue, more in terms of access to the towers, aquatic activity from the park, and use of the Play Barn, than in terms of general use of the site (Gas Works Park, no date).

Recognizing the severity of the buried contamination at the gasworks site, concern was expressed by some members of the community that opening up the soils of the Gas Company Peninsula could only worsen the potential for irreversible ecological damage to Lake Union. Notable among those voicing this concern was Mr. Otto Orth, Jr., a distinguished chemist and lifelong citizen of Seattle, who in 1984 recounted in a letter to the Seattle Times a history of the operations at the gasworks (Orth, 1984).

During 1983 and 1984, Environmental Protection Agency and University of Washington-investigators began to sample for toxic materials in offshore sediments and surface and subsurface soils. Because of the high levels of polyaromatic hydrocarbons [i.e., benzo(a)pyrene] and other contaminants reported, Mayor Charles Royer temporarily closed the park on April 21, 1984. He established a Health Advisory Committee that reopened portions of the park considered safe for the public. The committee agreed it would be prudent to conduct additional testing and investigations at the site. Tetratech, a consulting firm, was hired to carry out soil-sample and groundwater

investigations. A summary of the maximum polyaromatic-hydrocarbon concentrations found onsite is presented in Table 59. The groundwater investigation is still in progress.

3.3.7 Brattleboro-Hinsdale Bridge: Brattleboro, Vermont (E. C. Jordan Co., 1984)

The Brattleboro, Vermont, site illustrates the following:

* Site discovery during site investigation for a road construction project

* Movement of dense tar components by the action of gravity along a subsurface bedrock surface, from the original disposal area to beneath a riverbed

* Movement of tar in a coarse sand and gravel deposit

* Limited groundwater contamination from the wastes

3.3.7.1 Site History--

During initial site explorations associated with constructing a bridge across the Connecticut River, the State of New Hampshire discovered "odorous, oily materials" in soil borings. Subsequent analysis indicated that the materials were similar in composition to coal tars. Further investigation indicated that the site was the location of a town gas facility that was closed around 1949. One of the original gasworks buildings remains in use as a distribution center for bottled gas. The planned bridge abutment is to be built between this building and the river.

No detailed site history has been compiled on this plant. Table 60, which give the gas production data as compiled by the Radian Corp., shows that the plant produced carbureted water gas.

Currently, a site contamination audit has been completed, including recommendations on how to remove and safely dispose of contaminated materials encountered during construction of the bridge.

3.3.7.2 Extent of Contamination--

The initial exploratory borings indicated that there might be tar contamination at the site, and the site contamination audit confirmed this hypothesis. This investigation showed that the site was underlain by 5 to 15 feet of fill material that grades into alluvium as one proceeds out under the

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TABLE 59. MAXIMUM CONTAMINANT LEVELS:
GAS WORKS PARK, SEATTLE, WASHINGTON

Compound	Soil (ppm)
Naphthalene	1.6
Acenaphthylene	U 10
Acenaphthene	U 20
Fluorene	7.4
Anthracene	10
Phenanthrene	26
Fluoranthene	65
Benzo(a)pyrene	28
Pyrene	100
Benzo(b)fluoranthene	28
Benzo(a)anthracene	26
Chrysene	33
Benzo(k)fluoranthene	11
Benzo(g,h,i,)perylene	29
Dibenz(a,h)anthracene	3.1
Indeno(1,2,3-c,d)pyrene	25

U = Undetected at the detection limit shown.

TABLE 60. GAS PRODUCTION AT BRATTLEBORO, VERMONT

Year	Gas process	Production (10^6 ft^3/yr)
1890	Lowe	6
1900	Lowe	5
1910	Lowe	15
1920	Lowe	24
1930	Lowe	41

Lowe = Carbureted water gas.

SOURCE: Radian Corp. from Brown's Directory.

river (Figure 72). Underlying this layer is about 10 feet of sand and gravel that rest upon weathered bedrock (phyllite) and also extends out under the river. The bedrock surface slopes downward under the western portion of the river from about 220 feet above mean sea level (AMSL), to the eastern bank where the bedrock surface is at about 70 feet AMSL.

Figure 72 illustrates the extent of contamination under the old gasworks site and under the river, as far as Bridge Pier 1. It also illustrates how the contaminants have collected in the coarse sand and gravel immediately overlying the bedrock under the old gasworks building, and it shows that the contaminants have migrated through this coarse layer, down the bedrock surface, and under the river to the site of Bridge Pier 1. A borehole to the east, at the site of Bridge Pier 2, indicates that the coal tar has continued to migrate along the bedrock surface under the eastern portion of the river, where it occurs under 45 feet of sediments. This contaminant distribution clearly illustrates that the tar moved by way of density currents along the surface of the bedrock. The high permeability of the sand and gravel layer above the bedrock has enabled this migration to occur. Migration distance is at least 360 feet laterally and 150 feet deep from the contaminant source.

Maximum contaminant levels for soil, river sediment, and groundwater are presented in Table 61. Maximum levels in soil were found to the east and to the west of the gasworks building (B-107, B-108, B-110). Maximum levels in sediment were found at the site of Pier 1 (B105, B106). Maximum groundwater contaminant levels occurred both onshore (MW-107) and at the Pier 1 site (B-105, B-106). Sediment contamination levels at the site of Pier 2 were about five times lower than those presented in Table 61; no PAH's were detected in the groundwater at this location.

3.3.7.3 Site Remediation--

To address the contamination previously described, the following recommendations were made:

- Any contaminated soils excavated during construction of Abutment A or Pier 1 should be removed and disposed in a secure hazardous waste landfill.

- Suspended soil and visible contamination in water removed from the above construction areas should be removed. The water may then be discharged into the river without further treatment; no

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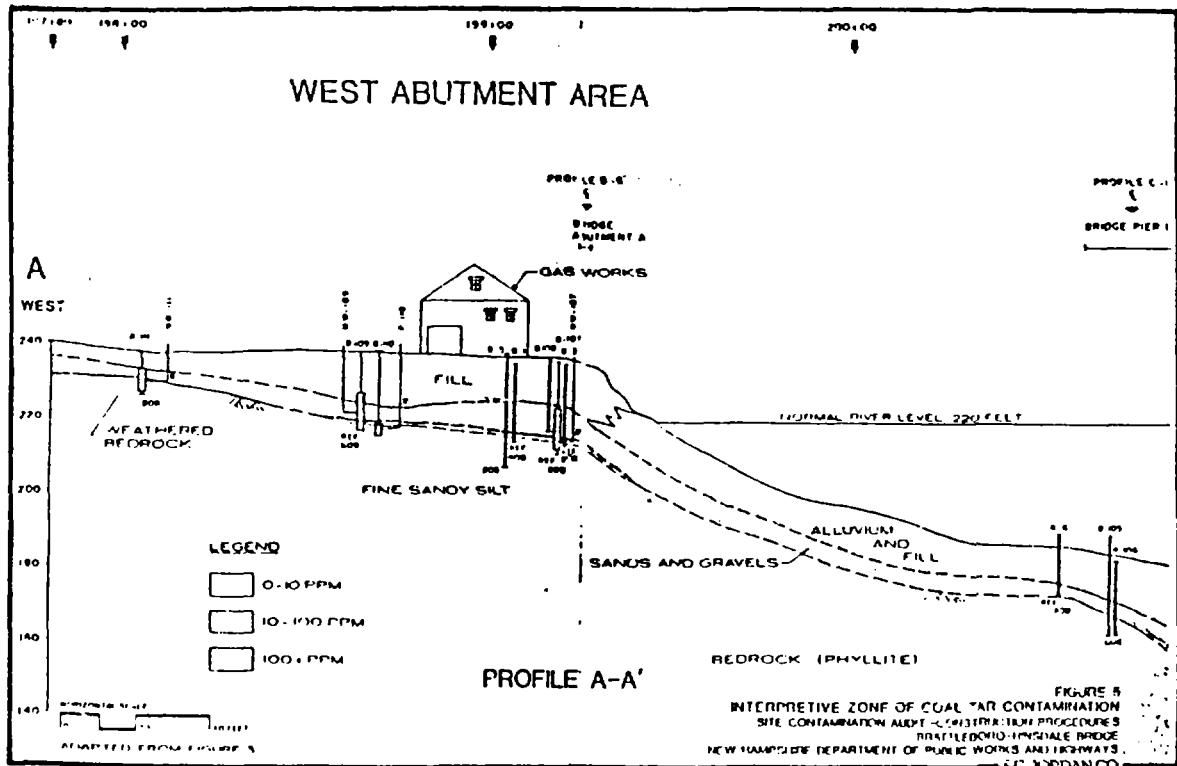
Figure 72. Brattleboro—Hinsdale Bridge.

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TABLE 61. MAXIMUM CONTAMINANT LEVELS: BRATTLEBORO, VERMONT

Compound	Soil (ppm)	River sediment (ppm)	Groundwater (ppm)
Benzene	1.3	0.025	0.15
Toluene	4.8	--	0.20
Ethylbenzene	32	0.130	0.27
Xylenes	64	0.27	0.79
Naphthalene	140	180	5.5
Acenaphthylene	85	1.3	0.27
Acenaphthene	140	28	0.84
Fluorene	100	22	0.051
Anthracene, Phenanthrene	190	240	0.037
Fluoranthene	64	72	0.0097
Benzo(a)pyrene	9.8	4.8	0.011
Pyrene	43	77	0.0094
Benzofluoranthene(b,k)	10	4.8	0.01
Benzo(a)anthracene/Chrysene	21	8.5	0.0095
Benzo(g,h,i,)perylene	2.3	1.4	--
Indeno(1,2,3,-c,d)pyrene	2.2	1.1	--

NPDES permit will be required (New Hampshire Water Supply and Pollution Control Commission).

- Pilings should be used to sup   : the bridge at Pier 1 to mini-mize the removal of contaminatuj material.

- Site safety and contingency plans should be developed to mini-mize worker and public exposure to contaminated material.

The report concluded that the bridge could be constructed without signi-ficant environmental or public health impacts and that removal of all contami-nated materials would not be necessary.

Since the report, the New Hampshire Department of Public Works and High-ways has decided to use pilings for both Abutment A and Pier 1, thereby avoid-ing any excavation. However, there is also the possibility of moving the bridge site upstream (for reasons other than site contamination), thus avoid-ing the contaminated area entirely. Vermont's Agency of the Environment con-siders the site to be of low priority because of low potential for release and contamination of groundwater, surface water, or air.

3.3.8 St. Louis Park, Minnesota (Barr Engineering Co., 1976; Ehrlich et al., 1982; Harris and Hansel, 1983; Hickok et al., 1982; Hult and Schoenberg, 1984; May et al., 1978; Minnesota Department of Health, 1938, 1974; Rittman et al., 1980; Schwartz, 1936; Schwarz, 1977; Sutton and Calder, 1975; U.S. Forest Products Laboratory, 1974)

The Reilly Tar and Chemical Corporation operated a coal-tar distillation and wood preserving plant (80-acre site) in St. Louis Park, Minnesota, from 1918 to 1972. The plant wastes, consisting of solutions of phenolic compounds and a water-immiscible mixture of PAH's, were discharged into a network of ditches emptying into an adjacent wetland. The contaminants entered under-lying aquifers via the wetlands and multiaquifer wells in the area. In 1932, the first well was shut down due to contamination, followed by others until over 35 percent of St. Louis Park's water supply was shut down. In 1975, the Minnesota Pollution Control Agency conducted a study to assess the extent and magnitude of the contamination. Since then, the Reilly site has been desig-nated as the State of Minnesota's highest priority Superfund site. This case study illustrates the following:

- Site discovery through groundwater contamination

- Contaminant transport via spill of drippings onsite, surface runoff, plant process-water discharge into adjacent wetlands, and movement of coal tar directly into bedrock aquifers through one or more deep wells used to drain creosote from the site and through one well that had experienced a spill into the well

- Contamination of several aquifers due to other water wells in the area extending through several aquifers, thereby providing a pathway for the contamination to travel between aquifers

- Contaminant migration in aquifers influenced by pumpage of water supply wells

- Removal of phenolic compounds in groundwater by biodegradation and naphthalene concentrations being reduced due to sorption

- Plan of remediation including a gradient-control well pumping system, a granular-activated carbon-filtering system, repair of leaking multiaquifer wells, removal of coal tar from any contaminated wells (in particular W23), establishment of source control wells, and monitoring of all contaminated aquifers over a set period of time.

3.3.8.1 Site History--

The Reilly Tar and Chemical Corporation operated a coal-tar distillation and wood preserving plant (80-acre site) in St. Louis Park, Minnesota, from 1918 to 1972 (Figure 73). The plant wastes, consisting of solutions of phenolic compounds and a water-immiscible PAH mixture, were discharged into a network of ditches discharging into an adjacent wetland. The contaminants entered underlying aquifers via the wetlands and a 909-foot deep, plant site well (W23) (see Figure 73). Well W23 was drilled in 1917 as a source of cooling water for the plant.

In 1932, the first St. Louis Park village well was drilled 3,500 feet from the plant. After only several weeks of operation, the well was shut down because of odors attributed to phenols. An investigation done by McCarthy Well Company (USGS files) concluded that the contaminants were entering the groundwater through old wells used to drain creosote from the site. One of the wells, W23, had experienced a spill of tar into the well, leading to contamination of several aquifers. By 1938, the Minnesota Department of Health (MDH) reported nine wells contaminated with phenolic or tar-like taste. The
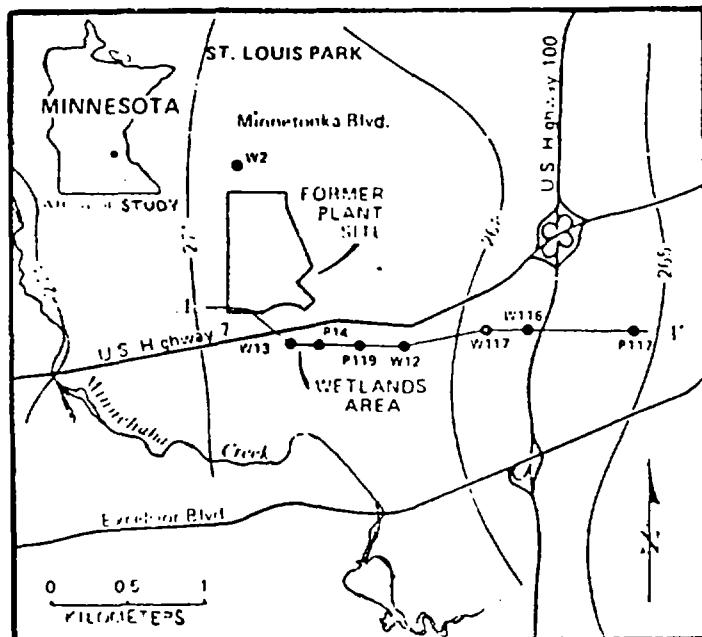
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Figure 73. Location of former plant site, wetlands
area, hydrologic section, water table
configurations, and location of key
wells at St. Louis Park, Minnesota.
Generalized potentiometric surface,
June 5, 1979, shown.

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well farthest from the plant site was originally 280-feet deep (into the St. Peter aquifer; Schwartz, 1936). This well was deepened another 130 feet, extending into the Prairie du Chien-Jordan aquifer, and it immediately yielded a distinct tar-like taste.

Throughout the 1960's and 1970's, the MDH and St. Louis Park monitored municipal, commercial, and industrial wells for phenol. In 1975, the Minnesota Pollution Control Agency (MPCA) conducted a study to assess the extent and magnitude of contamination. The study concluded that soil and shallow unconsolidated sandy aquifers near the old Reilly site were seriously contaminated and were the source of contamination to deeper bedrock aquifers. In 1978, PAH's, including benzo(a)pyrene, were found in several St. Louis Park municipal wells located 1/4 to 1/2 miles north of the site. These wells were closed down, followed by more well closures in 1979 and 1981 until over 35 percent of the city's water supply capacity was shut down.

In 1978, a USGS study of private wells in the St. Louis Park area, including Reilly's deep Well W23, revealed a down-hole flow of contaminated water from shallow aquifers to the Prairie du Chien-Jordan aquifer. The flow was estimated at greater than 150 gallons per minute (gpm). The well was plugged to stop continuing downward water contamination. In 1982, the MPCA cleaned out Well W23, removing over 150 feet of coal-tar wastes and debris.

All of the closed municipal wells draw from the Prairie du Chien-Jordan aquifer, as does 80 percent of the water supply to Minneapolis-St. Paul, of which St. Louis Park is a suburb. The Reilly site is designated as the State of Minnesota's highest priority Superfund site.

3.3.8.2 Extent of Contamination--

The vertical strata, including five major aquifers in the area, are shown in Figure 74. The Platteville Limestone is a nearly flat-lying, dolomite limestone. Fractures and solution channels contain water that yield small supplies to wells. The Glenwood Shale underlies the Platteville Limestone and serves as a confining bed except in locations where the shale has been eroded away. Glacial drift consisting of glacial till, outwash sand and gravel, lake deposits, and alluvium of several ages and provenances overlies the Platteville Limestone. The detailed stratigraphy of the drift at St. Louis Park is complex, but three areally persistent units have been identified. Directly

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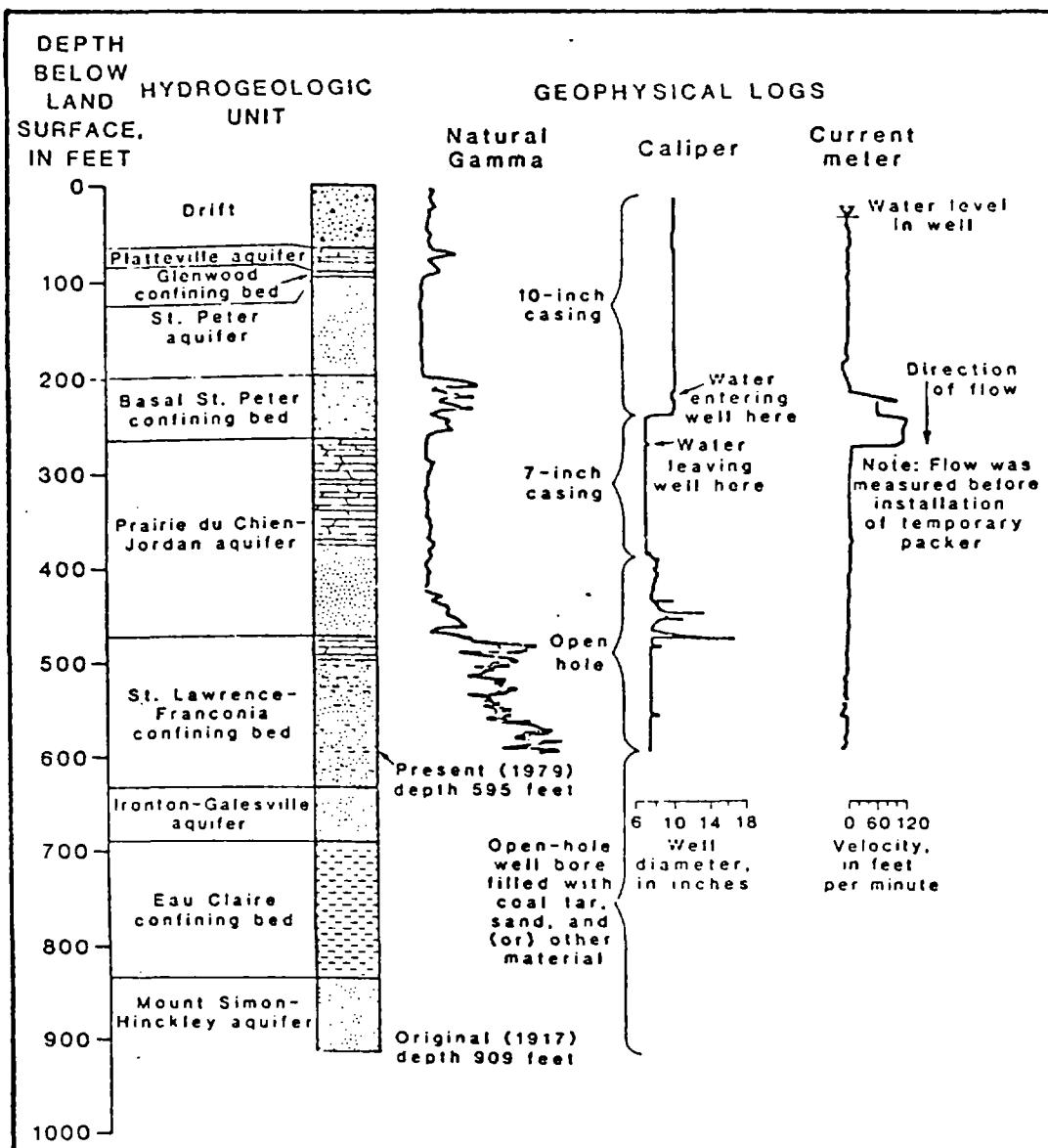
Figure 74. Hydrogeologic and geophysical logs of Well W23 ("Hinckley" well on the site).

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overlying the Platteville Limestone are (1) a unit of till, outwash, valley-fill deposits, and deeply weathered bedrock; (2) a middle unit of glacial sand and gravel called the Middle Drift aquifer; and (3) an uppermost unit of lake deposits and till. Below the Glenwood confining bed lies the St. Peter aquifer, the Basal St. Peter confining bed, the Prairie du Chien-Jordan aquifer, the St. Lawrence-Franconia confining bed, the Ironton-Galesville aquifer, the Eau Claire confining bed, and the Mount Simon-Hinckley aquifer.

The movement of the groundwater and, consequently, contaminants over the 50 years of plant operation has most probably varied with time because of a number of factors. A major control in groundwater movement is the draw-down created by water demand in communities as they have grown and diminished in population. The continuity of confining beds plays an important role in that a conduit for water and contaminant exchange between aquifers occurs where confining beds have been eroded. The presence of glacial valleys filled with coarse-grained deposits may provide preferential pathways for movement of groundwater or contaminants. Also, multiaquifer wells (wells hydraulically connecting two or more aquifers) provide an avenue of transport for contaminants and water, and they can locally change potentiometric surfaces of connecting aquifers. Multiaquifer wells result from original open-hole construction, leaks in casing, or flow in annular space between casing and borehole. In the St. Louis Park area, Hult and Schoenberg (1984) found that the water level in each aquifer is higher than the level in the underlying aquifers, causing water flow through multiaquifer wells to be downward.

The major contaminant from the Reilly plant was creosote, a complex mixture of chemical compounds. Typically, creosote contains 85 percent PAH [i.e., naphthalene, anthracene, phenanthrene; some of which are carcinogenic (at least 12 have been identified as carcinogenic, U.S. EPA, 1980a)] and 2 to 17 percent phenolics (i.e., phenol, methylated phenols). The remaining contents consist of various nitrogen- and sulfur-containing heterocyclic compounds (U.S. Forest Products Laboratory, 1974).

In addition to creosote, the Reilly plant discharged approximately 80,000 gallons of 70 percent NaOH into ponds from 1940 to 1943, as well as some sulfuric acids. [For more detail, see Table 4 in Hult and Schoenberg (1984).]

The distinction between transport processes of most natural constituents of groundwater and transport of coal tar is that many compounds of coal tar are relatively insoluble (Sutton and Calder, 1975; Schwarz, 1977). PAH's tend to adsorb strongly to soil particles and have low aqueous solubilities (Hickok et al., 1982). Phenolic compounds are generally more soluble in water than PAH's. The solubility of phenol is more than 10 g/L at 25 °C and pH 7.0, while the solubility of naphthalene under the same conditions is only 0.032 g/L (May et al., 1978). Solubility behavior of hydrocarbons is poorly under-stood. In Hult and Schoenberg (1984), dissolved constituents are defined as those not removed by filtration through a 0.45-micrometer filter. Many coal-tar derivatives are non-ionic and may exist as microscopic aggregates of individual monomers known as micelles. Micelles are considered part of the aqueous phase, and their movement is controlled by critical pore size. Micelles may move as though they were ideal solutes or become trapped, forming a hydrocarbon fluid phase at some distance from the source. This complicates contaminant movement and explains the wide variation of contaminant concen-tration throughout the area.

When creosote is mixed with water, two phases generally emerge: a light-er aqueous phase enriched in phenolics and a more dense hydrocarbon phase enriched in PAH's. Because the second phase has different properties (i.e., density and viscosity) from the aqueous phase, the hydrocarbons may move at a different rate and in a different direction than does the groundwater. At St. Louis Park, the dense hydrocarbon phase has percolated downward relative to the direction of groundwater flow, allowing contaminants to dissolve in the flowing groundwater and to be transported downgradient. The major transport mechanism is in the aqueous phase, whether as solutes or as micelles (Hult and Schoenberg, 1984).

There are three major paths for contaminant transport. The first is by spill or drippings onsite, which infiltrated and percolated through the unsat-urated zone to the water table. This has resulted in extensive contamination of the unsaturated zone on the 80-acre Reilly site. The contaminants reaching the groundwater vary in composition from area to area because the coal tar used throughout the plant's operation came from different suppliers and

subaereal decomposition of the coal-tar constituents produced degradation products dissimilar to those produced in the saturated zone.

The second path for contaminant transport is surface runoff and plant process-water discharge to depressions and wetlands found south of the plant site. Natural surface drainage was toward the site and south to Minnehaha Creek. Since approximately 1938, the drainage has been disrupted by roads and other manmade structures. Therefore, surface runoff and plant process-water were discharged through ditches and culverts to water table ponds near Well W13 (see Figure 73). If the rate of discharge becomes greater than the rate of evaporation, mounding in the water table occurs and vertical movement of the contaminated water and hydrocarbon-fluid phase into the underlying, confined drift aquifers occurs. Visible contamination extends at least 50 feet below the water table south of the plant site near Well W13 (Minnesota Department of Health, 1974; Barr Engineering Co., 1976). Since approximately 1938, surface water inflow to the ponds recharged to underlying peat and the Middle Drift aquifer. Inflow included 30 to 60 gpm of wastewater (Minnesota Department of Health, 1938) and as much as several hundred gpm of runoff during peak periods, increasing the vertical leakage. Also included in the plant discharge were sodium hydroxide and sulfuric acid occasionally used in plant processing.

The third path for contaminant transport is movement of coal tar directly into bedrock aquifers through one or more deep wells onsite. The main pathway is through the 909-foot deep Well W23, drilled in 1917. At some time, a coal-tar spill into this well occurred and is probably the source of early contamination reported in the Prairie du Chien-Jordan aquifer. The well was temporarily plugged and is now 595-feet deep. An unsuccessful removal of the viscous material was attempted in 1958.

3.3.8.3 Site Remediation--

In 1980, the available data were studied to assess the feasibility of (1) controlling movement of contaminated groundwater by pumping wells, (2) excavating or otherwise remedying contaminated soils, and (3) treating and disposing the residual waste products. A system of 12 to 15 wells in 5 to 6 aquifers was designed to flush the groundwater system. Hickok et al. (1982) estimated that the contaminated areas could be flushed in a few decades with

minimal sorption effects. However, leakage from the overlying drift, and especially from the "source zone," could continue to cause significant contamination of the bedrock aquifers for thousands of years, even with gradient control wells.

Ideally, management of the "source zone" would include excavating the highly contaminated surficial peat, removing the associated fluid, and pumping out the body of hydrocarbon fluid generally underlying the peat in the Middle Drift aquifer. Hickok et al. (1982) surmised that, at the time of their study, too little information on the actual contaminant distribution was available to design a complete remedial program for the "source zone."

As far as disposal of the "source" material, Hickok et al. (1982) concluded that the hydrocarbon fluid could not feasibly be treated for discharge to the Mississippi River or other surface waters. They concluded disposal would probably entail transport by truck or rail tank car to a secure landfill, a reprocessing plant, or another option depending on the total volume of hydrocarbon fluid. The disposal of the peat-associated fluid probably would be similar.

In a subsequent study, Harris and Hansel (1983) completed an evaluation of groundwater treatment and potable water supply alternatives for the City of St. Louis Park. As part of this study were bench-scale tests conducted to determine the efficiency of various water-treatment technologies in removing PAH's and other coal-tar derivatives from groundwater. Of all the technologies tested, only three were shown to be effective in removing PAH compounds to below the treatment goal of 280 ng/L total "other" PAH compounds. These three technologies were: granular-activated carbon (GAC), ozone/ultraviolet (O_3/UV), and hydrogen peroxide/ultraviolet (H_2O_2/UV). At raw-water concentrations of about 7,000 ng/L, GAC appears to be the most cost-effective, and a GAC pilot plant was set up and successfully operated in the pump station at one of St. Louis Park's contaminated wells. These three technologies achieved compliance with project-specific treatment goals and provided effluent water quality adequate for use in a potable water distribution system.

Phenolic compounds and naphthalene are disappearing downgradient from source points (i.e., Wells W13 and W23) faster than expected if only dilution were occurring. A study by Ehrlich et al. (1982) concludes that phenolic

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compounds in groundwater are being converted to methane and carbon dioxide by anaerobic bacteria. Naphthalene also shows an attenuation in concentration, but this appears to be due to sorption rather than biodegradation. Ehrlich et al. (1982) believe that the contaminated drift is acting as a treatment zone for removal of phenolic compounds that have penetrated the aquifer. They characterize this zone as a continuous flow bioreactor consisting of a fixed-film microbial population fed by a multiple nutrient stream as envisioned by Rittmann et al. (1980).

To date, a portion of the surface contamination has been removed and infilled with clean topsoil. The State of Minnesota is planning to build a highway interchange that would cover an area of contamination that has not yet been removed. If the State builds the interchange, the construction plans will include removal of the contaminated soils. If the interchange is not built, the Reilly Tar and Chemical Corporation is responsible for this surface contamination removal.

Upon approval by all parties involved, a remedial action plan will go into effect. The plan includes a gradient-control well pumping system, a GAC filtering system, repair of leaking multiaquifer wells, removal of coal tar from any contaminated wells (in particular W23), establishment of source control wells, and monitoring of all contaminated aquifers over a set period of time. The entire remedial action plan has not been completed and is still being drafted. The Minnesota Pollution Control Agency is coordinating the remedial action planning.

3.3.9 Pensacola, Florida (Ehrlich et al., 1982; Franks et al., 1985; Mattraw and Franks, 1984; McCarty et al., 1984; Troutman et al., 1984; Wilson and McNabb, 1983)

American Creosote Works Inc., an abandoned wood-treatment plant near Pensacola, Florida, was chosen by the U.S. Geological Survey in 1983 as a field laboratory to study the transport and environmental fate of creosote constituents in groundwater and surface water. Also, the site was chosen as being appropriate to apply the latest techniques for characterizing hazardous waste problems. To quote the National Priority List (NPL) description:

> The American Creosote Works, Inc., Site covers 1.5 acres in Pensacola, Florida, about 0.3 miles north of where Bayou Chico and Pensacola Bay meet. The facility treated wood with creosote and penta-

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chlorophenol (PCP) from the early 1900s to late 1981 or early 1982. PCP-contaminated waste water was discharged into two unlined 80,000-gallon percolation ponds. In February, 1981, the U.S. Geological Survey identified phenols in ground water associated with American Creosote Works. At present, no drinking supply wells are within the known zone of contamination.

This case study illustrates the following:

- Contamination of a sand-and-gravel aquifer from direct contact with creosote waste

- Insignificant attention of contaminants through sorption onto aquifer materials

- Anaerobic degradation of phenolic compounds in the groundwater environment

- Degradation of quinative to 2-quinolinone in groundwater by microbial oxidation

- Utilization of novel onsite groundwater sampling and analysis method to map the extent of microbes responsible for contaminant degradation, and by reference, the extent of contamination (Report is a selective summary of the USGS findings and is entirely based on the three referenced documents).

3.3.9.1 Site History--

The wood-treatment facility located within Pensacola, Florida, had been in operation from 1902 to 1981. Over this time, wood-preserving chemicals were discharged into two, unlined surface impoundments. Prior to dewatering and capping in 1982, the impoundment wastewaters were in direct hydraulic contact with an underlying sand-and-gravel aquifer. The aquifer was up to about 300-feet thick and consisted of deltaic, fine-to-coarse quartz sand deposits interbedded with locally confining, discontinuous clays and silts (Troutman et al., 1934). The impoundment wastes, in general, consisted of the wood preservative creosote, a coal-tar derivative. In addition to creosote, diesel fuel and pentachlorophenol (PCP) were discharged to the surface waste impoundments.

3.3.9.2 Methods of Investigation--

3.3.9.2.1 Soils and groundwater sampling--Nine test borings were drilled in 1981 to investigate the hydrostratigraphy beneath the site and to survey

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groundwater quality close to the facility. Borings were later (1983) completed and developed as groundwater monitoring wells. At each boring site, a well cluster of two to five wells was constructed with each well set at different depths. Details of well construction and materials, sampling protocols, and the results of groundwater sampling for creosote constituents and PCP's are given in Troutman et al. (1984) and Mattraw and Franks (1984).

3.3.9.2.2 Microbiological investigations--The aerobic degradation of quinoline in soils derived from the site was evaluated by standard laboratory batch techniques. The anaerobic degradation of phenolic compounds was also studied using enriched bacterial cultures from contaminated groundwaters at the facility (Mattraw and Franks, 1984).

3.3.9.2.3 Experimental/innovative investigative techniques--The research site was used to test the practicability of several experimental, nonconventional groundwater sampling methods:

- A multilevel "bundle" piezometer for sampling groundwater and measuring hydraulic heads at discrete vertical intervals within an aquifer (Mattraw and Franks, 1984)

- A reconnaisance groundwater sampling method, whereby groundwater within the hollow-stem auger is sampled and analyzed by an onsite high-performance liquid chromatograph (HPLC) for dissolved methane (Troutman et al., 1984; Franks et al., 1985).

3.3.9.3 Extent of Contamination Findings--

Results of the 1983 groundwater analyses by gas chromatography/mass spectroscopy (GC/MS) indicate the presence of approximately 80 organic contaminants in groundwaters near the facility. For classification purposes, three compound groups were identified: phenols (up to 2 ppm); PAH's (up to 2 ppm); and heterocyclic compounds containing oxygen, nitrogen, or sulfur (up to 1.5 ppm). Based on these general groupings, two contaminant zones were observed at the waste site:

- A highly contaminated water-table aquifer plume to approximately 36 feet depth

- A relatively less contaminated, confined, or semiconfined aquifer plume extending to a maximum depth of 75 feet.

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Questions concerning the transport of pure creosote within the unsaturated zone and within the aquifer were not directly addressed in any report. However, pools of denser-than-water, black, oily material were reported to be seeping from a stream approximately 450 feet downgradient of the waste impoundment (Mattraw and Franks, 1984).

PCP was not observed to be present in groundwater downgradient of the waste site at concentrations greater than 0.01 ppm.

Vertical distributions of contaminants at well clusters near the impoundments and approximately 450 feet downgradient show that contaminants have, in general, moved en masse (though in a dissolved state) with little or no "chromatographic separation" of compounds because of their differential retention on the aquifer media. Based on these observations, the reports conclude that retardation of organics because of sorption on aquifer materials and soil organic matter provides little or no control of contaminant transport at the site. This is not surprising considering that aquifer materials are predominantly clean sands, with minimal clays and organic matter.

Individual contaminants such as phenols do, however, decrease in concentration downgradient, presumably because of microbial degradations. Phenol biodegradation under anaerobic aquifer conditions is well established (Ehrlich et al., 1982; Wilson and McNabb, 1983; McCarty et al., 1984), and results at the Pensacola creosote site replicate these findings specifically. Godsy and Goerlitz (Mattraw and Franks, 1984, pp. 77-84), found a sequential disappearance of C^3 through C^6 carboxylic acids, phenol and benzoic acid, 3- and 4-methylphenol, and 2-methylphenol "during downgradient movement within the aquifer." In laboratory digesters containing enriched bacterial cultures from contaminated groundwaters at the site, the same sequential disappearance was observed with concomitant methane and carbon dioxide production.

The extent of the dissolved methane plume, and thus the extent of methane-generating bacteria and their degradation products, was later addressed in 1985 using an innovative drill-stem groundwater sampling technique and an onsite HPLC analysis (Franks et al., 1985). These findings indicate a much wider distribution of methane in the aquifer and that some of the byproducts of microbial degradation may have migrated farther in the aquifer than did the more readily degraded organic contaminants. Thus, selected

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contaminant plumes may extend well beyond the trace of the specific target (or "indicator") compounds (e.g., total phenols) if lower molecular weight organic and inorganic byproducts of the target compounds are considered.

No evidence was presented for the anaerobic microbial degradation of PAH's or heterocyclics, nor were any studies undertaken to examine the aerobic microbial degradation of any compound except quinoline. In one study by Bennett et al. (Mattraw and Franks, 1984, pp. 33-42), groundwater samples were collected and found to contain appreciable amounts of 2-quinolinone, a principal aerobic degradation product of quinoline. Subsequent soil samples and surface water and groundwater samples were found to contain large numbers of aerobic bacteria that convert quinoline to 2-quinolinone. These organisms were identified and counted.

3.3.9.4 Site Remediation (as of July 1983)--

According to the NPL description:

In March, 1982, American Creosote sold all the equipment onsite and later filed for bankruptcy under Chapter 11 of the Federal Bankruptcy Act. The state has negotiated a Consent Order requiring American Creosote to restore the discharge areas and install onsite monitoring wells. The company constructed higher berms around the ponds to prevent overflow during heavy rainfall.

EPA recently completed a remedial plan outlining the investigations needed to determine the full extent of cleanup required at the site. EPA plans to fund (1) a $290,000 remedial investigation/feasibility study to determine the type and extent of contamination at the site and identify alternatives for remedial action and (2) an $85,000 initial remedial measure involving fencing the site, posting warning signs, reconstructing the berms, and controlling flooding from the waste ponds. The work is scheduled to start in the third quarter of 1986.

3.4 CONCLUSIONS

Each of the gas sites visited showed surface contamination by tars, ash, and other wastes associated with gas manufacture. The amount of visible contamination varied from site to site, but it appeared more widespread at the larger sites.

Blue ferrocyanide contamination was visible at the Mendon Road, Taunton, and Pawtucket sites. Each of these sites was known to produce gas by coal carbonization. Spent oxides were discovered at the Spencer and Richmond

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plants. This spent oxide showed signs of sulfur and iron, but no ferro-cyanides. Both of these were principally water-gas plants.

Some oil contamination of the water in the Pawtucket canal (in Lowell, Massachusetts) was visible. This contamination was from the general direction of the gas plant. No other oil contamination of surface waters was seen at the other former gas sites.

Substantial gas odors were noted at the Lowell, Richmond, Taunton, Pawtucket, and Mendon Road sites. The odors indicate that contamination may be substantial at these sites. Only slight odors were noted at the Spencer and Worchester sites. The plant at Spencer was very small, and the Worchester site was capped with construction refuse and soil.

The case studies indicate that sites are "discovered" when (1) surface water is contaminated, (2) construction activities disturb the site or ground around the site, (3) redevelopment of the site is attempted, or (4) municipal groundwater sources are contaminated.

Phenol and PAH compounds appear to degrade in the groundwater when they are present in dilute concentrations. In raw tars, however, the microorganisms cannot survive, and the tar components do not degrade. This means that tars can remain substantially unchanged over time.

Tars (heavier than water) sink within groundwater systems until stopped by low permeability strata. Oils can float and spread on the surface of groundwater, contaminating a band of soil and thereby serving as a source of contamination to underlying groundwater. Cases of significant groundwater contamination usually can be attributed to the lighter, more soluble aromatics found in oils.

Local pumping of groundwater wells can affect the flow and transport of tars and contaminated water. Controlled pumping can be used to limit the spread of groundwater contamination.

Much of the historical data reported about the Stroudsburg site appears to be incorrect. The "coal tar" at Stroudsburg actually appears to be a tar from the production of carbureted water gas. The density of the tar is very close to water, which later separated. The low carbon content and absence of high-boiling organics imply that the tar was condensed after the washbox removed the higher boiling organics. The lack of phenols and the low nitrogen

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content of this tar identify it as a water-gas tar. The existence of an injection well for tar disposal also has been questioned because the term "tar well" was frequently used to describe underground tar storage tanks.

None of the case studies examined a plant that produced gas only by coal carbonization. Possible explanations for this include the fact that the coal carbonization plants produce tars that are not as prone to tar migration, it may only indicate the widespread adoption of the carbureted water-gas process, or coincidence. Coal carbonization tars were generally more dense and more viscous than carbureted water-gas and oil-gas tars.

Tar viscosity decreases with temperature, and surface tars generally become more mobile during the summer months.

The principal remediation employed at town gas sites is containment. Slurry walls, caps, and collection wells have been used.

Site contamination differs with the processes employed for gas manufacture. The principal contamination at the Seattle plant was lampblack, which was produced in substantial amounts by oil-gas production. At carbureted water-gas plants, the principal contaminant of concern was relatively mobile tar.

The waste disposal practices at the sites examined were generally quite poor. Although tars were frequently recovered, the liquids that disposed were either placed into the nearest body of water or, if they could not be disposed into water, placed into lagoons, trenches, or allowed to flow across the soil until absorbed. Solid wastes either were used to fill in areas along the shoreline or piled in a dump beside the plant.

4.0 STATE STATUS OF MANUFACTURED-GAS SITES

4.1 INTRODUCTION

This portion of the project was undertaken to determine the current status of manufactured-gas sites on a national basis. Originally, this determination was to be made by comparing the Radian list of manufactured-gas sites (compiled from Brown's Directory of American Gas Companies) to the national Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) CERCLIS list of sites (reported to EPA by the individual States). Such a comparison would have produced a list of manufactured-gas sites that individual States viewed as sufficiently hazardous for inclusion in CERCLIS. The resulting list could then be used to assist in planning further EPA efforts in the area.

The organization and nature of the information on the two lists prevented approaching the problem as planned, and an alternative approach was used to determine the status of manufactured-gas sites within States. Each EPA region was contacted to identify which States had placed manufactured-gas sites on CERCLIS and to determine what the status of the sites was. For most regions, the persons responsible for placing State sites on CERCLIS within individual States had to be consulted. Section 4.2 explains why the originally planned list comparison was impractical, and Section 4.3 describes the information acquired on the status of gas sites within States. Section 4.4 discusses the Radian list of manufactured-gas sites.

4.2 COMPARISON OF THE RADIAN LIST AND CERCLIS

The original task of comparing the Radian list and CERCLIS of manufactured-gas sites proved infeasible because the data included in each were incompatible. Figure 75 illustrates the type of data contained in the Radian list of town gas manufacturing sites. These data were compiled from Brown's Directory at 10-year intervals between 1890 and 1950. The information

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IDENTIFICATION OF TOWN GAS MANUFACTURING SITES - ALL PLANTS (1830-1950)

STATE: ALABAMA

NO.	CITY	COMPANY	YEAR	STATUS	GAS TYPE	GAS PRODUCTION RATE (MM cu. ft./YR) Coal	Water	Oil	Coke	Total	GASIFIER/ PROCESS	BY-PRODUCTS Coke	Tar	Ammonia	Other	MISCELLANEOUS INFORMATION
1	Birmingham	GEL&P Co	1890	*	C	40				40						
			1900	*	C	54				54						
			1910	*	C	244				244						
			1920	*	C,W	200	590			790	Lowe					
			1930	P												
			1940	P												
			1950	P												
			AVERAGES:			124	590			271						
2	Decatur	L&P Co	1890	*	C	1										
			1900	*	C	4				4						
			1910	*	C	5				5						
			1920	*	C	8				8		8	8			
			1930	*	C	30				30		2	25			
			1940	*	C	21				21		1	23			
			1950	*	C	24				24		1	25			
			AVERAGES:			15				15		1	20			
3	Eufaula	GEL&P Co	1890	*	C	0				1						
			1900	*	C	5				5						
			1910	*	C	2				2						
			1920	*	C	2				2						
			1930	O												
			1940	O												
			1950	-												
			AVERAGES:			4				4						
4	Huntsville	G&Coke Co	1890	*	C	1										
			1900	*	C	10				10						
			1910	*	C	12				12						
			1920	*	C	12				12		1	15			
			1930	*	C,CW	26				26		1	31			
			1940	*	C	26				26		1				
			1950	N												
			AVERAGES:			17				17		1	24			

Source: Radian, 1974.

Figure 75. Radian list of manufactured gas sites.

reported includes the city where the plant was located, company name, plant status, production, and byproducts. The only information recorded on plant location is the city name where the plant was situated. Figure 76 provides data from CERCLIS. These data indicate the EPA identification number, site name, address, county, latitude, and longitude for each site. The list includes no information on the type of contamination at the sites or on any operations at the site resulting in contamination. The site name of sites on CERCLIS can be used to determine if listed sites were former manufactured-gas sites, but only when the site is listed specifically as a gas plant or as owned by a gas company. Many of the sites in the list have names that do not indicate anything about the source of site contamination. Thus, merely compiling a list of the sites with site names that indicate they might be manufactured-gas sites would produce many omissions and inaccuracies.

The only basis that could be used to compare the Radian list and CERCLIS would be to compare the cities on each list and produce a list of CERCLIS sites in cities that also had manufactured-gas sites. Table 62 shows the number of sites resulting from this approach for the State of Alabama. There were 164 CERCLIS sites in cities that had manufactured-gas sites in the Radian list. The inability to match Radian and CERCLIS sites within cities made this type of comparison essentially worthless, so an alternative approach had to be found to examine the status of manufactured-gas sites in the States.

4.3 EXAMINATION OF MANUFACTURED-GAS SITE STATUS IN STATES

As an alternative, individual EPA regions and States were contacted to collect information on manufactured-gas sites within States. Table 63 lists the results of the inquiries and the current status of sites within each State. The information was collected from employees of either the EPA or State agencies who were "in a position to know" the status of CERCLIS waste sites within their areas. Consequently, the absence of known gas-manufacturing sites on CERCLIS may either indicate that there are actually none on the list for that State, or merely that the individuals contacted were not aware of any.

Table 63 summarizes the information collected from regions and States on the status of manufactured-gas sites. Tables 63 through 72 list the sites

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EPA ID	SITE NAME STREET CITY COUNTY NAME	COUNTY CODE ZIP CODE	LATITUDE	LONGITUDE	SMSA	HYDRO UNIT

***

| | PRATTVILLE | 001 | | | | |
| | AUGUSTA | 36067 | 322748.0 | 862830.0 | 5240 | 3150201 |

| ALD950710370 | CALLAHAN PROPERTY HWY 82 ROUTE 4 BOX 2 66 PRATTVILLE | 001 | | | | |
| | AUTAUGA | 36067 | 322748.0 | 862830.0 | 5240 | 3150201 |

| ALD980556245 | SOUTHERN RAILWAY DERAILMENT SITE MP 178.9 FREEMONT | 001 | | | | |
| | AUTAUGA | 36784 | 315442.0 | 874424.0 | | 3150203 |

| ALD005557004 | UNION CAMP CORP MONTGOMERY MILL SITE JENSEN RD PRATTVILLE | 001 | | | | |
| | AUTAUGA | 36067 | 322520.0 | 862520.0 | 5240 | 3150201 |

| ALD980495667 | BALDWIN COUNTY LANDFILL PO BOX 150 BAY MINETTE | 003 | | | | |
| | BALDWIN | 36507 | 305300.0 | 874624.0 | 5160 | 3140106 |

| ALD980495709 | BAY MINETTE CITY DUMP W 7TH ST BAY MINETTE | 003 | | | | |
| | BALDWIN | 36507 | 305300.0 | 874624.0 | 5160 | 3140106 |

| ALD980727929 | BOLON PROPERTY RABUN RD BAY MINETTE | 003 | | | | |
| | BALDWIN | 36507 | 305300.0 | 874624.0 | 5160 | 3140106 |

| ALD980727747 | BRANTLEY E R NEWPORT PARKWAY BAY MINETTE | 003 | | | | |
| | BALDWIN | 36507 | 305300.0 | 874624.0 | 5160 | 3140106 |

| ALD000652941 | DSI TRANSPORTS INC HWY 47 N BAY MINETTE | 003 | | | | |
| | BALDWIN | 36507 | 305300.0 | 874624.0 | 5160 | 3140106 |

| ALD001874254 | KAISER ALUMINUM & CHEMICAL CORP HWY 31 S BAY MINETTE | 003 | | | | |

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Figure 76. CERCLIS waste sites.

TABLE 62. COMPARISON OF RADIAN TOWN GAS SITES TO
CERCLIS FOR ALABAMA

County	City	Number of CERCLA sites in city
Barbour	Eufaula	1
Calhoun	Anniston	13
Colbert	Sheffield	6
Dallas	Selma	10
Etowah	Gasden	6
Jefferson	Bessemer	5
Jefferson	Birmingham	34
Lauderdale	Florence	4
Madison	Huntsville	16
Mobile	Mobile	29
Montgomery	Montgomery	18
Morgan	Decatur	14
Talladega	Talladega	1
Tuscaloosa	Tuscaloosa	7
	Total	164

TABLE 63.   STATUS OF MANUFACTURED-GAS SITES WITHIN STATES

State	Contacted	Gas sites on CERCLIS	No.	Comments
Alabama	Yes	Yes	1	Disposal area for "Blue Mud" in Birmingham recently remediated.
Alaska	No			
Arizona	Yes	None		
Arkansas	Yes	None		
California	Yes	Yes	?	No one knows how many gas sites are on CERCLIS. 136 gas sites have been identified. Sixty were connected to Southern California Gas, and 76 were connected to Pacific Gas and Electric. A PG+E site in Marin County was recently remediated for $2 million. One lampblack site in southern California was remediated. No groundwater contamination has been reported from any of the sites. See Table 64 for list of PG+E sites.
Colorado	Yes	None		
Connecticut	Yes			No successful reply to inquiries.
Delaware	Yes	Yes	8	Used Radian list to locate sites. Cover Gas Light site on NPL. One site inspection report is complete, and five other preliminary assessments are in preparation. Groundwater contamination present at the Dover site. See Table 65 for list of Delaware sites.
Florida	Yes	Yes	23	Used Radian list to locate sites. The State has recommended that each site owner prepare a Preliminary Contamination Assessment Plan (PCAP) to sample soil, groundwater, and surface water. Six sites have received walkover inspections, and two have PCAP's. See Table 66 for list of Florida sites and current status.
Georgia	Yes	Yes	1	Rome Coal Tar Pit in Rome, Georgia. Discovered May 1985. No assessment. Tar contamination was removed.
Hawaii	No			
Idaho	Yes[a]	None		
Illinois	Yes	None		No efforts currently being made to locate sites.
Indiana	Yes	None		

See notes at end of table.   (continued)

TABLE 63 (continued)

State	Contacted	Gas sites on CERCLIS	No.	Comments
Iowa	Yes[b]	Yes	1	Mason City site on CERCLIS. Preliminary assessment being prepared. Other sites on Radian list are currently being examined by utility companies. Preliminary assessments currently planned for Belle Plaine and Fairfield sites.
Kansas	Yes[b]	None		
Kentucky	Yes	None		Two gas sites were investigated in Owensboro, Kentucky (Goodlowe School and a VFW site). Determined that no action was necessary for the sites.
Louisiana	Yes	None		
Maine	Yes	Yes	3	Preliminary assessments being prepared for one site. Sites are in Portland, Lewistown, and Bangor.
Maryland	Yes	Yes	21	Used Radian list of sites. Have completed preliminary assessments on about half of the sites. See Table 67 for list of Maryland sites.
Massachusetts	Yes	None currently		Completed one site remediation (Mendon Rd. site near Attleboro) on iron oxide fill area. Cost paid for by State Superfund (approx. $2 million). Currently designing State program to examine gas sites. Preliminary assessments have been prepared on sites in Everett, Spencer, and Lowell, Massachusetts.
Michigan	Yes	None		Fifteen known sites that scored very low with risk assessments. Sites were not placed on CERCLIS. Michigan Consolidated Gas Co. (MichCon) has performed preliminary assessments of all sites. Remediation planning for two sites (Detroit riverside and Greenville) is in progress. The information from these sites will be applied to other sites in the State. Table 68 lists the MichCon sites.
Minnesota	Yes	None		
Mississippi	Yes	None		
Missouri	Yes[b]	None		
Montana	Yes	None		
Nebraska	Yes[b]	None		
Nevada	Yes	None		

See notes at end of table.   (continued)

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TABLE 83 (continued)

State	Contacted	Gas sites on CERCLIS	No.	Comments
New Hampshire	Yes	Yes	4	Preliminary assessments currently being prepared on all four sites.
New Jersey	Yes	Yes	?	Fifty-seven gas sites and 3 disposal areas have been located in New Jersey. Approximately 35 of the sites are currently under investigation. Table 69 lists the New Jersey sites.
New Mexico	Yes	None		
New York	Yes	Yes	?	Sixteen sites in New York are currently listed by the State as inactive hazardous waste sites. See Table 70 for these sites.
North Carolina	Yes	None		
North Dakota	Yes	None		
Ohio	Yes	None		Currently have no efforts to locate or examine sites.
Oklahoma	Yes	None		Had checked Radian list. Gas sites are considered low priority.
Oregon	Yes[a]	Yes	1	One plant in Astoria owned by Pacific Power and Light.
Pennsylvania	Yes	Yes	50+	Over 50 sites from the Radian list. Site inspection currently being prepared on four sites. Stroudsburg site is on NPL. See Table 71 for list of Pennsylvania sites.
Rhode Island	Yes	Yes	2	Two sites in Providence, Rhode Island, are on CERCLIS. They have an iron oxide disposal area (Cumberland) near the Massachusetts border. Remediated and built over one site in Newport, Rhode Island.
South Carolina	Yes	None		
South Dakota	Yes	None		
Tennessee	Yes	None		
Texas	Yes	None		One preliminary assessment done by Radian for a gas site in Austin, Texas.
Utah	Yes	None		

See notes at end of table.

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(continued)

TABLE 63 (continued)

State	Contacted	Gas sites on CERCLIS	No.	Comments
Vermont	Yes	Yes	2	Brattleboro, Vermont, site is on CERCLIS. Preliminary assessment currently being prepared by E.C. Jordan. Burlington, Vermont, site is on NPL, and a cleanup plan is currently being reviewed. One coal tar site (Barry, Vermont) is not on CERCLIS. Three other sites are known to exist in Montpelier, St. Albans, and Rutland. Nothing is currently planned for these sites.
Virginia	Yes	Yes	11	Currently performing preliminary assessments on sites. Table 72 is a list of the Virginia sites.
Washington	Yes[a]	Yes	4	(1) Seattle Gas Works Park--ranked below threshold for NPL, city is currently leading remediation. (2) Tacoma Tar Pits on NPL (3) Boulevard Park, Bellingham, Washington, preliminary assessment performed.
West Virginia	No			
Wisconsin	Yes	None		Using 42 sites in Radian list. Have received preliminary assessments on eight sites: Two Rivers, Sheboygan, Stevens Point, Green Bay, Oshkosh, Milwaukee (2), and Beaver Dam. Other sites are currently under investigation.
Wyoming	Yes	None		

[a]Data for EPA Region 10 (Washington, Oregon, and Idaho) were collected principally from regional EPA sources.

[b]Data for EPA Region 7 (Kansas, Iowa, Missouri, and Nebraska) were collected principally from regional EPA sources.

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TABLE 64. GAS SITES IN CALIFORNIA COMPILED BY
PACIFIC GAS AND ELECTRIC COMPANY

Site	No.	County
Gilroy	408-1	Santa Clara
Hollister	418-9	San Benito
Monterey	418-1	Monterey
Salinas	418-2	Monterey
San Luis Obispo	418-4	San Luis Obispo
Santa Cruz	408-7	Santa Cruz
Watsonville	408-8	Santa Cruz
Bakersfield	335-1	Kern
Chico	210-1	Butte
Chico	210-1A	Butte
Coalinga	325-8	Fresno
Colusa	212-1	Colusa
Fowler	325-2	Fresno
Fresno	325-3	Fresno
Fresno	325-3A	Fresno
Grass Valley	215-1	Nevada
Grass Valley	215-1A	Nevada
Lodi	316-6	San Joaquin
Madera	325-4	Madera
Marysville	212-2	Yuba
Marysville	212-2A	Yuba
Merced	325-5	Merced
Modesto	316-2	Stanislaus
Nevada City	215-3	Nevada
Oakdale	316-3	Stanislaus
Oroville	212-3	Butte
Reu ʾluff	213-1	Tehena
Redding	213-2	Shasta
Sacramento	206-2	Sacramento

(continued)

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TABLE 64 (continued)

Site	No.	County
Sacramento	206-2A	Sacramento
Selna	325-6	Fresno
Stockton	316-4	San Joaquin
Tracy	316-7	San Joaquin
Turlock	316-5	Stanislaus
Willows	210-2	Glenn
Woodland	206-3	Yolo
Eureka	119-1	Humboldt
Eureka	119-1A	Humboldt
Eureka	119-1B	Humboldt
Santa Rosa	104-6	Sonoma
Santa Rosa	104-6A	Sonoma
Okiah	104-B	Mendocino
Benicia	104-1	Solano
Daly City	508-2	San Mateo
Livermore	601-1	Alameda
Los Gatos	408-3	Santa Clara
Napa	104-3	Napa
Napa	104-3A	Napa
Oakland	601-2	Alameda
Oakland	601-2A	Alameda
Petaluma	104-4	Sonoma
Pittsburg	601-3	Contra Costa
Redwood City	508-1	San Mateo
San Francisco	502-1	San Francisco
San Francisco	502-1A	San Francisco
San Francisco	502-1B	San Francisco
San Francisco	502-1C	San Francisco
San Francisco	502-1D	San Francisco
San Francisco	502-1E	San Francisco

(continued)

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TABLE 64 (continued)

Site	No.	County
San Francisco	502-1F	San Francisco
San Francisco	502-1G	San Francisco
San Francisco	502-1H	San Francisco
San Francisco	502-1I	San Francisco
San Francisco	502-1J	San Francisco
San Francisco	502-1K	San Francisco
San Jose	408-5	Santa Clara
San Jose	408-5A	Santa Clara
San Leandro	601-4	Alameda
San Rafael	104-5	Marin
San Rafael	104-5A	Marin
Santa Clara	408-6	Santa Clara
St. Helena	104-7	Napa
Vallejo	104-9	Solano
Vallejo	104-9A	Solano

TABLE 65. DELAWARE GAS SITES

Dover Gas Light (DES7)
Wilmington Coal Gas Co.
Coal Gas Holder Site
New Castle Gas Co.
Smyrna Gas-Coke Co.
Georgetown Gas Co.
Lewes Gas Co.
Sussex Gas Co.

TABLE 66. FLORIDA GAS SITES

Location	Walkover inspection	(PCAP)[a]	Comments
DER District Office			
NW District			
Pensacola (Municipal)	Yes	No	No visible problem.
Tallahassee (Municipal)	Yes	No	No visible problem; known as Cascades Park.
NE District			
Jacksonville (Peoples/ Container Corp.)	Yes	Yes	Coal tar present onsite, CAP's being prepared.
Gainesville (Gainesville Gas Co./Poole Roofing Co.)	No	No	
Palatka (Municipal)	No	No	Location not known.
St. Augustine (Municipal)	No	No	Location not known.
SW District			
Tampa (Peoples)	No	No	Coal tar was shipped offsite.
Lakeland (Peoples)	No	No	Field and parking lot.
St. Petersburg (Peoples; site owned by City)	No	No	Coal tar may have been barged offsite; stadium constructed onsite.
Bradenton (Southern Co.)	No	No	
Clearwater (Municipal)	No	No	Coal tar sold and decomposed by bacteria. Now a parking lot.
Winter Haven (Central Florida Gas)	No	No	Adjacent to lake.

(continued)

TABLE 66 (continued)

Location	Walkover inspection	(PCAP)[a]	Comments
St. Johns River District			
Orlando (Peoples)	No	No	
Sanford (FL Public Utilities)	No	No	Office and parking lot.
Ocala (Gulf Natural Gas Corp.)	No	No	Up for sale.
Deland (FL Public Utilities)	No	No	
Daytona Power & Light	No	No	
South Florida District			
Key West	No	No	Location not known.
Ft. Myers (Municipal)	Yes	No	No visible problem.
SE Florida District			
Miami (Peoples)	No	No	
Ft. Lauderdale (Peoples)	Yes	Yes	Soil and groundwater sampling by ERM; no visible problem, low concentrations of coal tar constituents in groundwater.
Miami Beach (Peoples)	Yes	Yes	CAP has been prepared, but not approved by DER and DERM.
West Palm Beach (FL Public Utilities)	No	No	Office and parking lot

[a]PCAP = Preliminary contamination assessment plan.

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TABLE 67. MARYLAND GAS SITES

Annapolis Plant (MD141)

Bayard Station (MD166)

Canton Station (MD159)

Spring Garden Station (MD145)

First Plant (MD147)

Second Plant (MD148)

Scots St. Station (MD191)

Cranberry Run Substation

Westminster Plant (MD146)

Cambridge Town Gas (MD165)

Fredrick Town Gas (MD164)

De Grace Town Gas (MD162)

Salisbury Town Gas (MD163)

Cumberland Gas Light (MD190)

Frostbury Gas Light

Elkton Gas Light

Chesterton Gas Light

Hyattsville Gas & Electric

Crisfield Gas and Light

Easton Gas and Light

Hagerstown Gas and Electric

TABLE 68. MICHIGAN GAS SITES (INVESTIGATED BY MichCon)

Site name	Additional soil samples	Additional offsite groundwater samples	Site development guidelines	Access control	Additional groundwater investigation	Investigate drainage structures	2-year monitoring program	No further action
Group 1:								
Riverside Park							X	
Station B			X				X	
River Rouge							X	
Station J								X
Station H								X
Wealthy Annex			X	X			X	
Wealthy Street							X	
Group 2:								
Belding					X		X	
Ludington					X		X	
Old Ann Arbor		X			X		X	
Broadway					X	X	X	
Mt. Pleasant	X				X		X	
Big Rapids		X			X		X	
Greenville	X				X		X	
Muskegon					X		X	

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TABLE 69. NEW JERSEY GAS SITES

A. South Jersey Gas Company[a]

1. Atlantic City
2. Atlantic City
3. Pleasantville
4. Egg Harbor
5. Hammonton
6. Bridgeton
7. Millville
8. Glassboro
9. Paulsboro
10. Swedesboro
11. Penns Grove
12. Salem

B. New Jersey Natural Gas Company/Jersey Central Power & Light

1. Dover in Morris County
2. Belmar
3. Cape May City
4. Ocean City
5. Long Branch
6. Lakewood
7. Toms River
8. Wildwood
9. Asbury Park
10. Atlantic Highlands
11. Boonton

C. Elizabethtown Gas

1. Elizabeth
2. Elizabeth
3. Perth Amboy
4. Rahway
5. Flemington
6. Newton

7. Lambertville
8. Washington Boro (Warren)[b]
9. Newton
10. Phillipsburg

D. Public Service Electric & Gas

1. Hobart Avenue Gas Works
2. Camden Gas Plant
3. Camden Coke Plant
4. Gloucester Gas Works
5. Hackensack Gas Works
6. Harrison Gas Plant
7. Hoboken Gas Works
8. Halladay Street Works
9. Old Provost Street Works
10. West End Gas Plant
11. Mount Holly Works
12. Front Street Works
13. New Brunswick Works
14. Paterson Gas Plant
15. Plainfield Gas Works
16. Central Gas Plant
17. Ridgewood Gas Works
18. Riverton Works
19. South Amboy Gas Works
20. Trenton Gas Plant
21. Trenton Gas
22. Woodbury Works

E. Other Sites

1. Kearney--Koopers Coke
2. Tuckahoe
3. West Paterson[c]
4. Hawthorne[c]
5. Hawthorne[c]

[a]Present owners of former coal-gasification plant.

[b]Site located in the service territory of Elizabethtown Gas, but never owned or operated by the company.

[c]Disposal sites.

TABLE 70. NEW YORK STATE GAS SITES

New York State Electric and Gas

    Oneonta site

    Mechanicville sites (2)

    Plattsburgh site

    Cayuga Inlet site

    Cortland-Homer site

    Ithaca-Court Street site

    Ithaca-First Street site

    Elmira site

    Geneva site

Niagara-Mohawk Power Corporation

    South Glens Falls site

    Glens Falls site

    Gloversville site

    Saratoga site

    Harbor Point site

Rochester Gas and Electric

    Lower Falls site

TABLE 71. PENNSYLVANIA GAS SITES

Site Name	Street	City	County	CERCLA ID No.
UGI Corp Gas Mfg Plt - Allentown	Second & Union Sts.	Allentown	Lehigh	PAD980538748
Altoona Town Gas	100 E. Belwood Ave	Altoona	Bla'r	PAD980706857
Penn Gas & Wtr Co	W 9th & Oak St.	Berwick	Columbia	PAD980554818
Allied Chem Corp Bethlehem Tar Plt	Coke Works Rd.	Bethlehem	Lehigh	PAD980540025
UGI Corp Gas Mfg Plt - Bethlehem	Harvard Ave.	Bethlehem	Northampton	PAD980538771
Penn Pwr & Lgt Co Bloomsburg Gas Plt	Seventh St.	Bloomsburg	Columbia	PAD980539720
UGI Corp Gas Mfg Plt	Sixth & Wash Sts.	Boyertown	Berks	PAD980538912
Penn Gas & Wtr Co 8th St Plt	Eighth St.	Carbondale	Lackawanna	PAD980829675
UGI Corp Gas Mfg Plt	E. High & York Rd.	Carlisle	Cumberland	PAD980639597
Penn Pwr & Lgt Carlisle Gas Plt	E. Louther St.	Carlisle	Cumberland	PAD980539167
UGI Corp Gas Mfg Plt - Carlisle	Main & Chestnut Sts.	Carlisle	Cumberland	PAD980639951
UGI Corp Gas Mfg Plt - Catasauqua	Front & Walnut Sts.	Catasauqua	Lehigh	PAD980538839
UGI Corp Gas Mfg Plt	S. Front & Mill Sts.	Columbia	Lancaster	PAD980539126
Keystone Coke Co	442 River Rd.	Conshohocken	Montgomery	PAD088810239
Penna. Power & Light Co - Brunner Isl.	Brunner Island Station	East Manchester 133 Twp	York	PAD000797787
Easton Plant	640 N. 13th St.	Easton	Northhampton	PAD980832430
UGI Corp Gas Mfg Plt - W. Easton	Front & Green St	Easton	Northampton	PAD980538896
UGI Corp Gas Mfg Plt	Plymouth Ave.	Edwardsville	Luzerne	PAD980539738
UGI Corp Gas Mfg Plt	W. High & Hess Sts.	Elizabethtown	Lancaster	PAD980539654
Elrama Works Town Gas	Unobtainable	Elrama	Washington	PAD980706915
UGI Corp Gas Mfg Plt	Third & Mulberry Sts.	Harrisburg	Dauphin	PAD980638888
Penn Pwr & Lgt Co Harwood	PA Rte. 924 & Inrste Hwy 81	Harwood Mines	Luzerne	PAD980539191
UGI Corp Gas Mfg Plt	31 N. Poplar St.	Hazleton	Luzerne	PAD980539258

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TABLE 71 (continued)

Site Name	Street	City	County	CERCLA ID No.
UGI Corp Gas Mfg Plt - Hellertown	Crest Ave. & Signet Sts.	Hellertown	Northampton	PAD980539019
Penn Pwr & Lgt Honesdale Gas Plt	Church St.	Honesdale	Wayne	PAD980539704
UGI Corp Hunlock Power Plt	Unobtainable	Hunlock	Luzerne	PAD983434948
Taintor Tar Seeps	Taintor Drive (TR 323)	Lafayette Twp	Mckean	PAD981037997
UGI Corp Gas Mfg Plt	262 Conestoga St.	Lancaster	Lancaster	PAD003926441
Penn Pwr & Lgt Lehighton Gas Plt	State Rte. 443	Lehighton	Carbon	PAD980539829
Lewistown Gas Plant	Fleming Ave.	Lewistown	Mifflin	PAD981037443
UGI Corp Gas Mfg Plt	Front & Locust Sts.	Lititz	Lancaster	PAD980539183
Mayburg Tar Plt	Unobtainable	Mayburg	Forest	PAD980832612
Penn Gas & Wtr Co Filbert St Gas	Filbert St.	Milton	Northumberland	PAD980552715
Penn Gas & Wtr Co 3rd Wagner Site	Third St. & Wagner Ave. E.	Montgomery	Lycoming	PAD980539480
Penn Gas & Wtr Co Wash Holding	Washington St.	Montoursville	Lycoming	PAD980552772
Mt. Carmel Gas Plant	Railroad & Vine Sts.	Mt. Carmel	Northumberland	PAD981037674
Penn Pwr & Lgt Co Mt Joy Gas Plt	235 W. Main St.	Mt. Joy	Lancaster	PAD980538862
Penn Gas & Wtr Co Muncy Holder Site	Market St.	Muncy	Lycoming	PAD980539548
UGI Corp Nanticoke Gas Plt	Walnut St.	Nanticoke	Luzerne	PAD980539431
Butler Gas Prod Co	1103 13th St.	New Brighton	Beaver	PAD014449219
UGI Corp Gas Mfg Plt	200 Block S. 7th St.	Perkasie	Bucks	PAD980538904
Penn Coal Products Koppers Co Inc	#8 on Koppers Map	Petrolia	Butler	PAD980893626
Passyunk Town Gas	Passyunk Ave. & Schuylkill Riv.	Philadelphia	Philadelphia	PAD980708972
Point Breeze Town Gas	23rd & Market Sts.	Philadelphia	Philadelphia	PAD980707095
Richmond Town Gas	W. Delaware Ave. & N. Verangost	Philadelphia	Philadelphia	PAD980707038

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TABLE 71 (continued)

Site Name	Street	City	County	CERCLA ID No.
Pottsville Gas Plant	Coal & Railroad Sts.	Pottsville	Schuylkill	PAD981037617
UGI Corp Gas Mfg Plt	Fifth & Canal St.	Reading	Berks	PAD980539169
Equitable Gas Co	Rt. 21, East of Rogersville	Rogersville	Greene	PAD980430288
Penn Gas & Wtr Co Brg St Gas	Bridge St.	Scranton	Lackawanna	PAD980554851
Penn Gas & Wtr Co Greenridge Holder	Albright St.	Scranton	Lackawanna	PAD980538078
Penn Pwr & Lgt Co Shamokin Gas Plt	Vine St.	Shamokin	Northumberland	PAD980537955
Penn Pwr & Lgt Co Shenandoah Gas Plt	S. Main St.	Shenandoah	Schuylkill	PAD980539969
UGI Corp Gas Mfg Plt	Main & Frankl.. Sts.	Steelton	Dauphin	PAD980538821
Brodhead Creek	S. of Main St. Brg.	Stroudsburg	Monroe	PAD980691760
Penn Gas & Wtr Co Alley & Arch St	Gashouse Alley & Arch Sts.	Sunbury	Northumberland	PAD007917818
Penn Pwr & Lgt Co Sunbury Gas Plt	Vine St.	Sunbury	Northumberland	PAD980539662
Penn Pwr & Lgt Co Tamaqua Gas Plt	Greenwood St.	Tamaqua	Schuylkill	PAD980537831
Penna Power Co	To be obtained	Taylor Twp	Lawrence	PAD007912736
Carpentertown Coal & Coke Works	Rd. #1	Templeton	Armstrong	PAD004370898
UGI Corp Gas Mfg Plt	Franklin & Rasberry Sts.	W. Easton	Northampton	PAD980538953
UGI Corp Wyoming Holding Sta	Sixth St.	W. Wyoming	Luzerne	PAD980539910
Carnegie Natural Gas Prop Camden	Hellena St.	West Mifflin	Allegheny	PAD980537658
Penn Gas & Wtr Co. Wilkes-Barre	Water St. & North St.	Wilkes-Barre	Luzerne	PAD980539670
Penn Gas & Wtr Co N Riv St Plt	N. River St.	Wilkes-Barre	Luzerne	PAD980539795
Penn Power & Light: Wilkes-Barre Gas	Darling St.	Wilkes-Barre	Luzerne	PAD980539613
Penn Gas & Wtr Co Darling St Plt	Darling St.	Wilkes-barre	Luzerne	PAD980552533
Pennwalt Corp Easton Plt	Heller Town P'. Vly. Ave.	Williams Twp	Northampton	PAD980537591
Penn Gas & Wtr Co Rose St Gas Plt	Rose St.	Williamsport	Lycoming	PAD980428399
Penn Gas Wtr Co Mulberry St Gas	Mulberry St.	Williamsport	Lycoming	PAD980554935

TABLE 72. VIRGINIA GAS SITES

Site	Status
Danville Town Gas Craghess St. RR Depot Danville, VA 24541	Discovery (PA)
Fredericksburg Town Gas 400 Charles Street Fredericksburg, VA 22401	Discovery (PA)
Fulton Bottom Town Gas Fulton & Williamsburg Road Richmond, VA 23201	Discovery (PA)
Lynchburg Town Gas Black Water Street Lynchburg, VA 24501	Discovery (PA)
Newport News Town Gas Terminal Blvd. & 22nd Street Newport News, VA 23601	Discovery (PA)
Norfolk Town Gas Monticello & VA Beach Rd. Norfolk, VA 23501	Discovery (PA)
Portsmith Town Gas Gust Lane Portsmouth, VA 23701	Discovery (PA) Site inspection
Roanoke Town Gas NE Kimbell & Rutherford Ave. Roanoke, VA 24001	Discovery (PA)
Suffolk Town Gas Hill Street Suffolk, VA 23434	Discovery (PA)
Alexandria Town Gas	
City Yard Town Gas	

PA = Preliminary assessment.

that have been located, are currently under investigation, or have been listed by the States.

4.4 EVALUATION OF THE RADIAN LIST OF MANUFACTURED-GAS SITES

The list of gas production sites compiled by Radian is a faithful compilation of the site material contained in Brown's, but it has several shortcomings, most of which result from the way Brown's compiled and reported information on the manufactured-gas industry.

Sites were listed in Brown's corporate designation. Whenever two plants merged their management, Brown's usually stopped listing one plant, even though it was often still in production. In Radian's compilation of the data from Brown's, plants that merged with larger plants showed no production at the site, even though gas was still produced there. The listing for Plattsburgh, New York, is a good example. The plant merged with New York State Electric and Gas Corporation in 1932, and subsequently its production was included with that of Ithaca, New York. The Radian compilation shows that no gas was produced under the Plattsburgh listing in 1940 and 1950, although the plant actually operated into the 1950's.

Brown's Directory includes only gas producers who sold their gas to consumers. Facilities that supplied gas to a limited market (e.g., a large hotel or an individual factory) did not appear in the directory. Many universities also had their own gas plants at one time; however, because they did not sell gas to consumers, they were not listed in Brown's. Brown's also did not list gas production at factories that generally manufactured producer gas for onsite heating purposes. An estimated 11,000 such gas producers were in operation in 1921 (Chapman, 1921). Most sites using producer gas would probably have several gas producers on each site, so the actual number of possible sites would be much lower than 11,000. Brown's Directory, however, reported none of these.

Brown's Directory also did not record the movement of plant operating sites. It was common for gas companies to operate a small plant initially, outgrow it, and then expand to a larger facility. Brown's recorded the company's production as occurring at a single site rather than at two sites and, as a result, the records Radian compiled indicate only a single site.

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Brown's generally included substantial information on plant byproducts marketed by individual companies (in later operating years), but Radian did not generally compile this information. The data available in Brown's could be very useful in evaluating individual sites, but a very large effort would be required to compile the data for all listed sites.

The Radian compilation apparently did not include any gas purchased by gas companies from byproduct coke ovens. This was gas produced by coal carbonization, which was not manufactured by a gas company, but was sold (generally locally) to a gas company by a coke manufacturer. From a waste or site standpoint, it makes no difference if the gas were produced by a coke company selling gas as a byproduct or by a gas company selling coke as a byproduct. A town having a gas company that produced some gas and purchased additional gas from a local coke manufacturer would have had at least two gas production sites, but it would be reported only as one in the Radian compilation.

When the data were compiled from Brown's at 10-year intervals, significant variations in rates of gas production were overlooked. The production of gas dropped sharply after 1930, and it did not recover until World War II. This would have produced errors in the total amounts of gas reported, particularly for the production of carbureted water gas.

4.5 CONCLUSIONS

Many States currently have active programs to examine manufactured-gas sites specifically for possible environmental hazards. In most cases, the existing owners are requested to perform preliminary site assessments to determine the extent of site contamination. Any necessary remedial actions are determined only after the extent of contamination is known. Several States have used the Radian list of manufactured-gas sites to assist them in locating gas sites within their States.

In most States, the environmental authorities are initially satisfied with determinations that no significant amounts of waste materials are moving off a site and that no significant groundwater contamination has occurred. Remediation is generally not performed at sites until some waste material moves offsite or additional use of the manufactured-gas site is planned. The site owners are generally content with leaving the sites as monitored (but

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unremediated) because the cost of carrying the site as undeveloped land is small compared to the costs of remediation and redevelopment. In many cases, the sites have remained undeveloped land since the surface structures were removed.

In summary, the Radian list of manufactured-gas sites presented several problems. Not all gas-manufacturing sites appeared in Brown's; hence, the list is incomplete. Brown's listed gas manufacturers by corporate designation, so some companies listed as single sites in Brown's were actually composed of several operating plants. In addition, several plant sites were listed as only one when plants moved within cities. Cities having operating coke plants (which produced gas that was sold to gas companies) and gas companies were reported as having only a single gas production site.

The Radian list is a good starting point for locating gas plants because most of the towns listed had a gas-manufacturing plant. Local sources of information, however, should not be overlooked, and they should take precedence over both information in Brown's Directory and in the Radian list.

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