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October 30, 2002

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Re: United States v. Aeronca, Inc. (Skinner Landfill CERCLA Litigation)

Dear Annette and Mike:

I have enclosed the expert report of William J. Deutsch in the above-captioned matter.

Yours truly,

Dave

David E. Northrop

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Occurrence, Geochemistry and Fate of Manganese

Skinner Landfill, West Chester, Ohio

Prepared by:

William J. Deutsch

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Prepared for:

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October 16, 2002

Occurrence, Geochemistry and Fate of Manganese
Skinner Landfill, West Chester, Ohio

The purpose of this report is to present information and opinions regarding the occurrence, geochemistry and fate of manganese at the Skinner Landfill, West Chester, Ohio. This report is based on my twenty-three years experience as a groundwater geochemist and review of the technical data and other information you have provided, including but not limited to the material identified in Exhibit A.

Introduction

Manganese is a naturally occurring metal that is common in soil, sediment, groundwater and surface water. Permanganate (MnO_4^-), the highly oxidized form of manganese, is used as a cleaning agent for metals. At an industrial facility operated by Aeronca, Inc. in Middletown, Ohio, a solution containing potassium permanganate was used in a cleaning bath for stainless steel parts. It is alleged that spent cleaning solution containing residual potassium permanganate and its byproducts (described below) was sent to the Skinner Landfill for disposal. The landfill was used for the disposal of municipal refuse for approximately 56 years (1934 – 1990). As early as 1964, industrial wastes were also accepted for disposal. This landfill is now on the National Priorities List for cleanup because of contamination of soil and water by a wide variety of organic and inorganic compounds. The site has been characterized by Phase I and Phase II Remedial Investigations conducted by Roy F. Weston, Inc. (1989) and WW Engineering & Sciences Inc. (1991), respectively.

Manganese has been identified as one of the contaminants of potential concern in groundwater and the surface water of Skinner Creek. The source(s) of manganese in these waters may be either (or both) naturally occurring or landfill-disposed manganese. The occurrence of elevated manganese in groundwater and surface water is a function of the geochemistry of manganese and the local environmental conditions. This report discusses the following topics as they apply to manganese and the Skinner Landfill:

- Manganese Geochemistry
- Impact of Landfills on Manganese Mobility
- Occurrence of Manganese at the Skinner Landfill
- Potassium Permanganate Chemistry, Uses and Byproducts
- Fate of Potassium Permanganate in the Landfill Environment
- Is Potassium Permanganate Likely to Increase/Decrease Manganese Concentrations in Groundwater and/or Surface Water

The final section of this report is a summary of these topics and provides conclusions relative to the contribution of disposed permanganate waste to the occurrence of manganese contamination at a level of concern at the Skinner Landfill.

Manganese Geochemistry

Manganese is a common minor element in soil with a normal range in concentration of 20 to 3,000 mg/kg (Lindsay 1979). It is redox-sensitive and occurs naturally in the environment in three valence states: Mn(II), Mn(III), and Mn(IV). [The valence state of an element reflects the electrical charge on the species of the element; for example Mn(II) is the +2 charge species of manganese. The higher the charge, or valence, the more oxidized the element; and the lower the charge, the more reduced the element.] Common manganese minerals that form from each of its naturally-occurring valence states are: Mn(II) - rhodochrosite ($MnCO_3$); Mn(III) – manganite (γ-MnOOH); and Mn(IV) - pyrolusite (β-MnO_2). It has been shown that microorganisms can obtain energy by the oxidation of organic matter to carbon dioxide with Mn(IV) as the sole electron acceptor (Lovley 1991). The source of Mn(IV) in an aquifer is the minerals, such as pyrolusite, that contain this valence state of the element. The oxidation/reduction process for pyrolusite can be represented by the following reaction:

$$2\ MnO_2(\text{pyrolusite}) + 4\ H^+ + CH_2O \rightarrow 2Mn^{2+} + 3\ H_2O + CO_2 \qquad (1)$$

where CH_2O (carbohydrate) is used to represent organic compounds, which are oxidized by Mn(IV) in MnO_2. The Mn^{2+} released to solution by this reaction may partially precipitate under reducing conditions (that is, conditions that favor the lower valence state species of manganese) as rhodochrosite according to:

$$Mn^{2+} + CO_3^{2-} \rightarrow MnCO_3(\text{rhodochrosite}) \qquad (2)$$

The resulting dissolved concentration of manganese in equilibrium with the manganese mineral will be a function of many factors including groundwater pH, Eh, carbonate concentrations and ionic strength, however rhodochrosite is generally more soluble than pyrolusite, thus the dissolved Mn concentration will be higher in groundwater limited by rhodochrosite formation than one limited by pyrolusite formation. [Eh is a measure of the redox potential of the aquifer. A low Eh reflects reducing conditions that favor lower valence states of manganese, while a high Eh reflects oxidizing conditions and favors higher valence states.] Typical oxidizing groundwater conditions (pH =7, Eh > 600mV) result in pyrolusite limiting dissolved manganese to less than 0.05 mg/L, whereas typical reducing groundwater conditions (pH =7, Eh < 400 mV) result in rhodochrosite limiting dissolved manganese in the range of 5.5 to 30 mg/L (Lindsay 1979).

Reoxidation of a groundwater with high dissolved Mn^{2+} commonly leads first to the formation of an Mn^{3+} mineral (Eary and Schramke 1990) by reactions of the type:

$$2\ Mn^{2+} + \tfrac{1}{2}\ O_2 + 3H_2O \rightarrow 2\ MnOOH(\text{manganite}) + 4\ H^+ \qquad (3)$$

The solubility of MnOOH may be 100x less than that of $MnCO_3$ under the more oxidizing conditions, consequently the dissolved Mn^{2+} concentration may be on the order of 0.05 mg/L when limited by MnOOH under oxidizing conditions.

As a consequence of the above redox reactions, the dissolved manganese concentration is controlled by natural site conditions and the formation of stable manganese minerals, which have different solubilities depending on the site conditions. Application of these geochemical processes to the landfill environment is discussed next.

Impact of Landfills on Manganese Mobility

A landfill is a source of reactive organic matter like the carbohydrate that reduced pyrolusite in reaction (1) above. Because of this, placing a landfill on a typical oxidized soil creates a localized reducing condition that commonly dissolves manganese minerals in the soil as the landfill leachate moves through the subsurface. This leads to elevated manganese concentrations in groundwater beneath and immediately downgradient of the landfill (Deutsch 1997). As the groundwater flows away from the landfill and reoxidizes, dissolved manganese concentrations decrease due to physical dispersion processes and the geochemical process of mineral formation described above (reaction 3).

Several examples of manganese geochemistry at landfills have been published. Baedecker and Back (1979) showed that at the Army Creek landfill in Delaware dissolved manganese increased from less than 0.01 mg/L under background conditions to 4.2 mg/L adjacent to the landfill. Downgradient of the landfill, the concentration began to decrease and it had returned to background about 800m from the landfill. Baedecker and Apgar (1984) described the geochemical processes active at the Army Creek landfill and noted the presence of the reduced Mn mineral rhodochrosite adjacent to the landfill and an oxidized MnO_2 mineral farther downgradient. They equated the reducing conditions at the landfill to the degradation of landfill organic matter. Nicholson et al. (1983) found a similar situation at the Borden landfill (Ontario, Canada) where the background Mn concentration was 0.28 mg/L and the concentration in the plume downgradient of the landfill was 11 mg/L. They also discuss the change in redox condition along the flow path and the manganese minerals that control dissolved Mn concentrations.

Mirecki and Parks (1994) studied the Shelby County landfill (Memphis, Tennessee) and found that the average Mn in background wells in the alluvial aquifer was 0.185 mg/L; whereas in the wells downgradient of the landfill it was 2.5 mg/L. The redox potential was very reducing at about 0 mV near the landfill and it was more oxidizing downgradient. Nielsen et al. (1995) measured the following dissolved Mn concentrations associated with the Vejen City landfill (Jutland, Denmark):

landfill perimeter - 5.7 mg/l;
135m downgradient - 0.67 mg/L;
250m downgradient - 0.46;
350m downgradient - <0.1 mg/L.

Deutsch (1997) discusses Mn mobility at the Fort Lewis (Washington) landfill where the dissolved Mn increases to 11 mg/L due to the influence of landfill leachate, but the concentration returns to background (<0.05 mg/L) 650m downgradient of the landfill. At this site, the landfill depressed the pH to 6.25 from a background value of 7.25. Downgradient of the landfill, the pH increased to background over the distance required for manganese mineral formation.

The mobilization of manganese by organic matter has also been shown for conditions other than found at landfills. At a crude oil spill site in Minnesota, dissolved Mn increases from < 0.03 mg/L upgradient of the spill to 6.6 mg/L in the vicinity of the product floating on the water table. In this case, the reducing conditions are produced by the soluble, oxidizable components of the crude oil that dissolve into the groundwater. As the groundwater naturally reoxidizes downgradient of the spill site, the dissolved Mn concentration returns to background within 100 to 150 meters of the source (Baedecker et al. 1993). Note that the soluble components of petroleum hydrocarbon include benzene, ethylbenzene and toluene, which are present at high concentrations in the waste lagoon sediments and groundwater at the Skinner Landfill. These compounds were also found in drums near the waste lagoon and landfill (WWES 1992).

Occurrence of Manganese at the Skinner Landfill

The concentrations of manganese have been characterized during the Skinner Landfill Remedial Investigation for a wide variety of media (groundwater, surface water, soil and sediment). This section discusses the concentration ranges relative to background levels. The background concentrations were measured in media unaffected by the landfill.

Groundwater

Groundwater beneath the Skinner Landfill flows in a shallow aquifer comprised of unconsolidated sand and gravel deposits. Underlying this shallow aquifer is the bedrock aquifer. Groundwater samples were collected and analyzed for potential organic and inorganic contaminants from 39 monitoring wells installed in the two aquifers. Table 2-4 of the Baseline Risk Assessment lists the manganese range of detections in groundwater as 0.0104 to 18 mg/L with a background concentration range of 0.021 to 0.712 mg/L. The higher levels of manganese concentration in impacted groundwater are consistent with the solubility of manganese minerals affected by leachate from a landfill described above.

Surface Water

Surface waters were collected and analyzed from Mill Creek, Skinner Creek, Dump Creek, Diving Pond and Trilobite Pond. As shown in Table 2-1 of the Baseline Risk Assessment, manganese was not considered a contaminant of concern in any of these locations, except one, because its concentration was similar to background values. The one exception is Skinner Creek surface water where manganese concentration was measured in the range of 0.0163 to 0.0715 mg/L (Table 2-5, Baseline Risk Assessment), which was considered elevated compared to the background manganese concentration of 0.0094 mg/L for the creek.

Soil

Sixty-two soil samples were collected in the vicinity of the buried waste lagoon and analyzed for a wide variety of potential contaminants, including manganese. The range of detected manganese concentrations in soil near the waste lagoon is 168 to 2,430 mg/kg (Phase II RI Table 5.3). Seven soil samples were collected in the vicinity of the buried pit and analyzed for manganese. The range of detected manganese concentrations in soil near the buried pit is 639 to 3,630 mg/kg (Phase II RI Table 5.4). Nineteen additional soil samples were collected and analyzed for manganese from monitoring well boreholes throughout the site. The range of detected manganese soil concentrations from these samples is 337 to 1,830 mg/kg (Phase II RI Table 5.5). The range of background concentrations for manganese in soil has been reported as 542 to 1,180 mg/kg (Phase II RI Table 5.3, 5.4 and 5.5). Manganese was not considered a chemical of concern for soil in the Baseline Risk Assessment when the measured soil concentrations were compared to this background range (Baseline Risk Assessment Table 2-1).

Sediment

Sediment samples from three creeks, three ponds and three leachate seeps associated with the site were collected and analyzed. The range of manganese concentrations for the 32 sediment samples is 470 to 3,520 mg/kg (Phase II RI Table 5.13, 5.15, 5.17 and 5.19). The range of background concentrations for manganese in sediment has been reported as 805 to 3,250 mg/kg (Phase II RI Table 5.13, 5.15, 5.17 and 5.19). Manganese is not considered a chemical of concern for sediments based on a comparison with background levels (Baseline Risk Assessment Table 2-1).

Summary of Manganese Detections

Based on the sampling described above, manganese is only considered elevated in groundwater and Skinner Creek surface water. Skinner Creek may be partially fed by groundwater with elevated dissolved manganese. The other surface water bodies at the [illegible] the soil and sediment are not considered to be contaminated with manganese.

[illegible]sium Permanganate Chemistry, Uses and Byproducts

Manganese in permanganate (MnO_4^-) is in the +7 valence state [Mn(VII)]. This is not a naturally occurring valence state for manganese. Permanganate is produced by an industrial process for the purpose of creating a strong oxidizing agent. Mn(VII) is not a thermodynamically stable form of manganese, and it will slowly oxidize water with the evolution of oxygen:

$$4\ MnO_4^- + 4\ H^+ \rightarrow 4\ MnO_2 + 2\ H_2O + 3\ O_2 \qquad (4)$$

In this oxidation-reduction reaction Mn(VII) in the permanganate ion is reduced to Mn(IV) and it forms the stable +4 manganese mineral pyrolusite.

Potassium permanganate is a strong oxidant that has been used for approximately 100 years in the drinking water, wastewater and chemical manufacturing industries. [An oxidant is a chemical compound that has the ability to raise the valence state (oxidize) a contaminant. For example, hydrogen sulfide is a contaminant in which sulfur is in the –2 valence state. An oxidant will oxidize this sulfur to the +6 valence state and change the hydrogen sulfide contaminant to the relatively benign sulfate compound.] In potable water it is used to control taste and odor problems by oxidizing iron, manganese and hydrogen sulfide. In the wastewater field, it is used to oxidize hydrogen sulfide and other odor causing compounds, prevent corrosion, and destroy phenol and other toxic pollutants. In the manufacturing industry it is used for metals surface treatment and equipment cleaning. This was the application of permanganate at the Aeronca plant in Middletown.

Rather than being considered a source of groundwater contamination, potassium permanganate has been identified, tested and used as a reagent to treat groundwater contaminated with organic contaminants. The strong oxidizing ability of permanganate is used to oxidize the carbon in the organic compounds to carbon dioxide and decompose the contaminant to innocuous inorganic compounds. For example, potassium permanganate has been emplaced in the aquifer at the Portsmouth Gaseous Diffusion plant in central Ohio to test its ability to oxidize trichloroethylene (TCE) that is contaminating the groundwater (Siegrist, Lowe et al. 1999). The oxidation of TCE (C_2HCl_3) by permanganate can be represented by the reaction:

$$C_2HCl_3 + 2\ MnO_4^- + 2\ H_2O \rightarrow 2\ MnO_2 + 2\ HCO_3^- + 3\ Cl^- + 3H^+ \qquad (5)$$

At the recent Battelle Conference on Remediation of Chlorinated and Recalcitrant Compounds (May 20-23, 2002; Monterey, California), at least 18 papers were presented on the use of permanganate for aquifer remediation and an entire session was devoted to permanganate oxidation of organic compounds.

The primary manganese byproducts of permanganate are the manganese minerals describe above (pyrolusite, manganite and rhodochrosite). The type of natural manganese solid that forms in the environment depends on the pH, redox state and concentrations of other ions in the groundwater or surface water. The resulting dissolved concentration of manganese is a functioning of the solubility of the manganese solid in the environment.

Fate of Potassium Permanganate in the Landfill Environment

As discussed above, it is alleged that spent cleaning solution containing residual potassium permanganate and its byproducts was sent to the Skinner Landfill for disposal. Reaction (4) shows that the primary manganese byproduct of permanganate decomposition is manganese dioxide, MnO_2. The disposal of MnO_2 in the landfill adds to the natural concentration of manganese in the soil, which is likely present as MnO_2 and/or MnOOH. The effect on groundwater concentration of adding additional MnO_2 to the soil is discussed in the following section. The residual permanganate added to the landfill produces reduction/oxidation (redox) reactions because of the strong oxidizing ability of this compound.

Redox reactions require that one compound be oxidized while another is being reduced. The addition of an oxidizing agent, like permanganate, to a landfill environment with reduced organic compounds, such as benzene and toluene, produces redox reactions. The Remedial Investigation reports document the presence of oxidizable organic compounds in the soil and groundwater at the landfill. A representative redox reaction between organic compounds and permanganate is benzene (C_6H_6) oxidation:

$$C_6H_6 + 10\ MnO_4^- + 4\ H^+ \rightarrow 10\ MnO_2 + 6\ HCO_3^- + 2\ H_2O \qquad (6)$$

In this reaction, carbon is oxidized from the –1 valence state in benzene to the +4 valence state in HCO_3^- (bicarbonate) as manganese is reduced from the +7 valence state in permanganate to the +4 valence state in MnO_2. Permanganate thus oxidizes many of the organic contaminants co-disposed in the landfill producing innocuous, inorganic forms of carbon such as bicarbonate. This is the same process that occurs when permanganate is used as a remediation agent for organic compounds described above. The manganese that is reduced forms the solid MnO_2 or some other manganese solid (MnOOH or $MnCO_3$) depending on the final pH and Eh of the environment.

Is Potassium Permanganate Likely to Increase/Decrease Manganese Concentrations in Groundwater and/or Surface Water

The dissolved manganese concentration in groundwater or surface water is a function of the solubility of the manganese mineral that is limiting the dissolved level. As discussed above, manganese is a redox-sensitive element and at normal environmental pH values the solubilities of its minerals are primarily a function of the redox potential of the environment. Under relatively oxidizing Eh conditions (> 600 mV), pyrolusite (MnO_2) limits dissolved manganese to low concentrations typically less than 0.05 mg/L, while under relatively reducing Eh conditions (< 400 mV) rhodochrosite ($MnCO_3$) limits the dissolved manganese concentration, but at relatively high levels of about 5 to 30 mg/L. As a consequence, because the dissolved concentration of manganese in groundwater or surface water is limited by the formation of these reaction products, the concentration in water has little to do with the amount of manganese solids in the soil or sediment. As long as a mineral is effectively limiting the dissolved manganese concentration, adding or removing the mineral containing the element from the solid phase will not affect the dissolved concentration. Consequently, the addition of permanganate and its byproduct manganese minerals to the landfill will not directly affect the concentration of manganese in the groundwater or surface water, except as it affects the Eh of the environment.

Because permanganate is a strong oxidizing agent, it will tend to raise the Eh of the environment. This will favor the formation of the byproduct manganese minerals pyrolusite or manganite over rhodochrosite, which will limit the dissolved manganese to lower levels. However, if the reducing capacity of the environment is greater than the oxidizing capacity of the permanganate, then the Eh of the environment may not be raised by the addition of permanganate. This is the typical condition in a landfill or waste lagoon with large quantities of organic matter, which acts as a reducing agent. Where there is more reducing agent than oxidizing agent, the result will be that the system becomes reducing and the Eh is depressed. This is the case at the Skinner Landfill. Although the Eh was not measured during the investigations at the landfill, the dissolved iron concentrations were determined. Dissolved iron can be used to estimate Eh conditions, because iron concentrations are low under oxidizing conditions and high under reducing conditions at normal groundwater pH values. Iron was detected in the range of 4.23 to 19 mg/L in the contaminated sand and gravel aquifer monitoring wells compared to a background range of 0.009 to 0.018 mg/L. This strongly suggests that the landfill has generated reducing conditions in the groundwater. The elevated manganese in the water is not a result of the permanganate or its byproduct manganese minerals added to the landfill, but is a response to the reducing environment created by the organic matter disposed in the landfill. The addition of permanganate to the landfill would work to reduce the dissolved manganese level in water because of its potential to make conditions more oxidizing.

Summary and Conclusions

Spent permanganate cleaning solution was allegedly disposed of at the Skinner Landfill. The Remedial Investigation conducted at the landfill showed elevated dissolved manganese in groundwater beneath and downgradient of the landfill and Skinner Creek surface water, but manganese concentrations were not elevated in soil, sediment or other surface waters.

Manganese is a reactive element in the environment. Under oxidizing conditions found in most shallow groundwaters, mineral formation will limit dissolved Mn to low dissolved concentrations (<0.05 mg/L). If reducing conditions are imposed on the natural system by a landfill or other source of oxidizable organic matter, the Mn minerals will become more soluble and will maintain a higher level (tens of mg/L) of dissolved Mn in groundwater.

The manganese in permanganate is in the highly oxidized +7 valence state making permanganate a strong oxidizing agent, which is used in many commercial applications including groundwater treatment. The byproducts of permanganate oxidation are the relatively oxidizing manganese minerals, such as pyrolusite and manganite. The disposal of permanganate and its byproduct manganese minerals at a landfill would tend to counter the reducing ability of the co-disposed organic contaminants thereby making the system less reducing. This would lower rather than raise the dissolved manganese concentrations in groundwater and surface waters impacted by the landfill.

The elevated manganese in groundwater and surface water at the Skinner Landfill is a result of the reducing reactions between disposed organic compounds and the naturally occurring manganese minerals in the soil and sediment. The addition of permanganate and its byproducts to the Skinner Landfill does not produce the elevated dissolved manganese levels, and may actually lower manganese concentrations in groundwater and surface water.

Exhibit A

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