Fate of Brine Applied to Unpaved Roads at a Radioactive Waste Subsurface Disposal Area

Larry C. Hull* and Carolyn W. Bishop

ABSTRACT

Between 1984 and 1993, MgCl₂ brine was used to suppress dust on unpaved roads at a radioactive waste subsurface disposal area. Because Cl⁻ might enhance corrosion of buried metals in the waste, we investigated the distribution and fate of Cl⁻ in the vadose zone using pore water samples collected from suction lysimeters and soluble salt concentrations extracted from sediment samples. The Cl/Br mass ratio and the total dissolved Cl⁻ concentration of pore water show that brine contamination occurs primarily within 13 m of treated roads, but can extend as much as 30 m laterally in near-surface sedimentary deposits. Within the deep vadose zone, which consists of interlayered basalt lava flows and sedimentary interbeds, brine has moved up to 110 m laterally. This lateral migration suggests formation of perched water and horizontal transport during periods of high recharge. In a few locations, brine migrated to depths of 67 m within 3 to 5 yr. Elevated Cl⁻ concentrations were found to depths of 2 m in roadbed material. In drainage ditches along roads, where runoff accumulates and recharge of surface water is high, Cl⁻ was flushed from the sediments in 3 to 4 yr. In areas of lower recharge, Cl⁻ remained in the sediments after 5 yr. Vertical brine movement is directly related to surface recharge through sediments. The distribution of Cl⁻ in pore water and sediments is consistent with estimates of vadose zone residence times and spatial distribution of surface water recharge from other investigations at the subsurface disposal area.

Magnesium Chloride brine was applied to unpaved roads at a radioactive waste subsurface disposal area at the Idaho National Engineering and Environmental Laboratory (INEEL) between 1984 and 1993 to suppress airborne dust emissions. Brines stabilize unpaved roads and provide significant benefits in reduced maintenance costs, control of particulate air emissions, and reduced storm-water runoff pollution (Hansen, 1982; Skorseth, 2000). Salts applied to roads can also have negative effects by increasing the conductivity of runoff water, which induces and accelerates corrosion (Keating, 2001). Corrosion can affect critical vehicle parts, damage bridge decks, and compromise the structural integrity of parking structures (Transportation Research Board, 1991). Chloride ions penetrate concrete and corrode reinforcing rods, causing the surrounding concrete to crack and fragment. Other roadside hardware and some nonhighway objects near salt-treated roads also are affected by the corrosive properties of road salts (Transportation Research Board, 1991). Enhanced corrosion due to salt is also a concern for subsurface disposal at INEEL because waste containers used for the disposal of radioactive waste and buried activated metals are subject to degradation by underground corrosion (Piculio et al., 1985). Corrosion in soils is an electrochemical process that is influenced by factors such as water content, redox potential, pH, soil resistivity, soluble ionic species, and microbiological activity (Roberge, 2000). Elevated salt concentration enhances corrosion by decreasing the resistivity of the soil and increasing ionic current flow associated with soil corrosion. The chloride ion is particularly harmful because it participates directly in the anodic dissolution of metals and causes pitting in stainless steel (Roberge, 2000). Environmental impacts from road-salt applications have mainly been evaluated for NaCl used in deicing applications (Forman and Deblinger, 2000; Williams et al., 2000; Lofgren, 2001; Foos, 2003). A few studies have evaluated the effects of Ca²⁺ and MgCl₂ used for deicing (Transportation Research Board, 1991; Lewis, 1999). However, little is known about the fate of brine used for road stabilization and dust suppression. This paper describes the distribution of brine in the vadose zone for a period of 18 yr after MgCl₂ brine was applied to unpaved roads for dust control at a radioactive waste disposal area.

Site Description

Since 1952, the Radioactive Waste Management Complex at the INEEL has been used for disposal and storage of radioactive waste generated from experiments associated with development of nuclear power reactors, reprocessing of spent nuclear fuel, and nuclear weapons construction. The Radioactive Waste Management complex consists of two operational areas: the Subsurface Disposal Area (SDA) and the Transuranic Storage Area. The SDA was opened in 1952 for shallow land disposal of solid radioactive waste. Waste is buried in cardboard boxes, plywood boxes, carbon steel drums, and some bulky items are placed directly into pits or trenches (Holdren et al., 2002). The SDA is still open for disposal of low-level radioactive waste. The Transuranic Storage Area was added in 1970 to store transuranic waste above ground; this waste is being processed and shipped to the Waste Isolation Pilot Plant in New Mexico for disposal.

The INEEL is in southeastern Idaho (Fig. 1) and occupies 2305 km² in the northeastern region of the Snake River Plain. The surface of the INEEL is a relatively flat, semiarid, sagebrush desert. Elevations on the INEEL range from 1460 m in the south to 1802 m in the northeast, with an average elevation of 1524 m above sea level. INEEL’s climate is arid to semiarid with average annual

Abbreviations: INEEL, Idaho National Engineering and Environmental Laboratory; SDA, Subsurface Disposal Area.

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breccia. The interbeds are called the A–B, B–C, and C–D sedimentary layers after the basalt flow groups (i.e., A, B, C, and D) that bound the layers above and below. The C–D interbed is by far the most continuous. However, each of the interbeds is known to be stratigraphically discontinuous. The A–B interbed is of limited aerial extent and generally occurs only beneath the northern half of the SDA.

Hydrology

The Big Lost River, an intermittent stream, passes 3 km north of the SDA (Fig. 1). A diversion dam diverts part of the river flow to playas, referred to as the spreading areas, 1.5 km west of the SDA, to prevent flooding of downstream facilities at the INEEL. The spreading areas west of the SDA represent a significant potential source of recharge to the vadose zone and groundwater near the SDA (Nimmo et al., 2002). The SDA is separated from the playas by a lava ridge and man-made dikes, and has not been flooded by the river since waste disposal operations began. However, the SDA has been flooded by local runoff three times because of rapid snowmelt combined with rain. Dikes and drainage channels were constructed around the perimeter of the SDA in 1962 in response to the first flood. The height of the dike was increased and the drainage channel around the perimeter was enlarged following a second flood in 1969. The dike was breached by accumulated snowmelt in precipitation of 22.1 cm (Clawson et al., 1989). Winter snowfall normally occurs from November through April and ranges from a low of 17.3 cm yr⁻¹ to a high of 151.6 cm yr⁻¹, with an annual average of 70.1 cm. Potential annual evaporation is 109 cm, and about 80% of this occurs between May and October. Evapotranspiration by the sparse native vegetation of the Snake River Plain is estimated to be between 15 and 23 cm yr⁻¹, or four to six times less than the potential evaporation. Periods when the greatest quantity of precipitation water is available for infiltration (late winter to spring) coincide with periods of relatively low evapotranspiration rates.

Geology

The INEEL is on the Eastern Snake River Plain, a northeast trending structural basin about 325 km long and 80 to 110 km wide. The plain is underlain by a layered sequence of Tertiary and Quaternary volcanic rocks and sedimentary deposits (Anderson and Lewis, 1989). Volcanic rocks in this sequence consist of basaltic lava flows and cinder beds. During periods of volcanic quiescence, fluvial, lacustrine, and eolian sediments were deposited. Alternating periods of volcanic activity and sediment deposition formed a complex sequence of layers (Fig. 2). Radioactive wastes are buried in unconsolidated sediments in a natural topographic depression surrounded by basalt lava flows. Undisturbed surficial sediments at the SDA range in thickness from 0.6 to 7.6 m and consist primarily of fine-grained playa and alluvial material. Irregularities in sediment thickness generally reflect the undulating surface of underlying basalt flows. The interbeds consist of unconsolidated sediments, cinders, and
Fig. 2. Cross section trending west to east through the Subsurface Disposal Area.

Fig. 3. Map of the Subsurface Disposal Area showing the distribution of ponded water from snowmelt in the spring of 1995. Locations of neutron access tubes (NAT) used to measure moisture infiltration are also shown.

Perched water bodies have been identified at three depth intervals. Perched water has been observed in surficial sediments immediately above the underlying basalt flow at depths between about 3 and 9 m. Deeper perched water zones occur at depths of 24 to 27 m and 61 to 67 m, which correspond to the sedimentary B–C and C–D interbeds, respectively (Fig. 2). Perched water typically occurs in fractured basalt above the interbeds. Two principal sources of perched water at the SDA are surface infiltra-
tion and water moving laterally from the spreading areas west of the SDA.

Very high water flow rates and transport of tracers over large distances have been measured in the vadose zone at the INEEL where surface recharge is high. In the summer of 1994, a vertical moisture movement rate of 5 m d$^{-1}$ was measured from land surface to a depth of 55 m through the fractured basalt during an infiltration test where the infiltration rate was 4526 m$^3$ d$^{-1}$ (Dunnivant et al., 1998). The infiltration test was conducted approximately 2.1 km south of the SDA. In May and June 1999, 2.6 × 10$^7$ m$^3$ of water were diverted into the spreading areas west of the SDA. A tracer, 1,5-naphtalene sulfonate, was mixed into the water in the spreading areas, and vadose zone and aquifer wells were monitored for arrival of the tracer (Nimmo et al., 2002). Tracer was detected on top of the B–C interbed 1.3 km from the spreading areas at significant concentrations. Tracer was detected in one perched zone monitoring well under the SDA, Well USGS-92. This well is on the order of 1.4 km from the spreading areas. The tracer test provided conclusive evidence of significant lateral flow of water in perched water zones near the SDA. Transport velocities calculated from the tracer arrival curves are on the order of 10 m d$^{-1}$ for horizontal saturated flow in perched water.

**Magnesium Chloride Brine Treatments**

Heavy equipment moving on unpaved roads at the SDA has generated large quantities of airborne dust that limit visibility. To control the dust, a chemical solution of MgCl$_2$ brine was experimentally applied to 4 km of dirt roads at and around the SDA in August 1984. The treatment met the objective of controlling dust, and the brine was subsequently reapplied in 1985, 1992, and 1993. Applying MgCl$_2$ brine to stabilize roads, control fugitive dust emissions, and minimize suspended solids contamination of storm-water runoff is a well accepted practice (Hansen, 1982; Skorseth, 2000). Road surface preparation includes grading and smoothing the road surface to set the grade and remove potholes, wash boarding, and furrowing. Before application, the road is scarified and prewetted to a depth of 12 cm. Brine is then applied at a recommended rate of 3 kg m$^{-2}$ (Hansen, 1982). One to four additional applications of water are used to aid in the hardening process after the brine is applied. When brine is applied properly, the salt is tightly bound into the roadbed material and forms a dense, low permeability layer.

The MgCl$_2$ brine used on the roads at SDA is primarily Mg$^{2+}$ and Cl$^-$, with large concentrations of Na$^+$, SO$_4^{2-}$, and Br$^-$ (see Table 1). No Ca$^{2+}$ or CO$_3^{2-}$ concentrations are given in the analysis provided by the manufacturer, but concentrations of these constituents are likely to be low, controlled by carbonate solubility. Chloride/bromide mass ratios are commonly used to identify water from different sources (Davis et al., 1998). From the range in typical percentages by weight provided by the manufacturer (Table 1), the Cl/Br mass ratio in the brine could fall anywhere between 105 and 540. The brine was not analyzed at the time of application to determine the actual concentrations.

The roads selected for treatment varied somewhat from application to application (Hull and Bishop, 2003). Dust suppressant was applied in 1993, but no map for this application was found, so it is assumed that the 1993 application was made in the same areas as the 1992 application. We estimated the amount of brine applied to roads inside and around the SDA based on the application maps for each year and the assumptions that the roads were 9 m wide and the brine was applied at 3 kg m$^{-2}$. We estimated that 600 t of brine were applied inside and immediately around the SDA (Table 2). The brine is about 27% (w/w) Cl$^-$ ion. Therefore, 161 t of Cl$^-$ would have been added to SDA roads between 1984 and 1993. Based on the recommended brine application practices described above, the initial Cl$^-$ concentration in the roadbed would be on the order of 2500 to 3500 mg kg$^{-1}$, depending on the bulk density of the roadbed material. Two applications of brine, 1 yr apart, would double this roadbed concentration.

**METHODS**

We studied the distribution of brine in the vadose zone at the SDA by interpreting chemical analyses of water samples collected from suction lysimeters and perched water monitoring wells, and by collecting sediment samples and extracting the soluble ions from the sediment. No water samples were specifically collected for this investigation. Lysimeters and perched zone monitoring wells have been sampled since 1986, and the existing database of analytical data was used to evaluate brine migration. This section discusses the sampling networks and the methods employed to collect samples.

**Table 1. Chemical composition of MgCl$_2$ brine applied to the Subsurface Disposal Area.**

<table>
<thead>
<tr>
<th>Component</th>
<th>Typical weight percent</th>
<th>Estimated</th>
</tr>
</thead>
<tbody>
<tr>
<td>Magnesium</td>
<td>7.1–9.2</td>
<td>114 530</td>
</tr>
<tr>
<td>Calcium</td>
<td>Not given</td>
<td>120†</td>
</tr>
<tr>
<td>Sodium</td>
<td>0.2–1.0</td>
<td>11 200</td>
</tr>
<tr>
<td>Potassium</td>
<td>0.1–0.8</td>
<td>8 400</td>
</tr>
<tr>
<td>Lithium</td>
<td>0.1–0.2</td>
<td>2 500</td>
</tr>
<tr>
<td>Chloride</td>
<td>21–27</td>
<td>356 000</td>
</tr>
<tr>
<td>Sulfate</td>
<td>1.2–2.0</td>
<td>26 300</td>
</tr>
<tr>
<td>Bromide</td>
<td>0.05–0.2</td>
<td>742</td>
</tr>
<tr>
<td>Bicarbonate</td>
<td>Not given</td>
<td>384†</td>
</tr>
</tbody>
</table>

† Calcium and bicarbonate are calculated assuming the brine is saturated with respect to calcite at atmospheric $P_{\text{CO}_2}$. The brine has a density of 1.32 kg L$^{-1}$.

<table>
<thead>
<tr>
<th>Year</th>
<th>Inside the SDA</th>
<th>Around the SDA</th>
<th>Total</th>
<th>Other total estimate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1984</td>
<td>85 530</td>
<td>14 800</td>
<td>100 326</td>
<td>108 000†</td>
</tr>
<tr>
<td>1985</td>
<td>44 040</td>
<td>47 380</td>
<td>91 420</td>
<td>—</td>
</tr>
<tr>
<td>1992</td>
<td>54 930</td>
<td>148 000</td>
<td>202 920</td>
<td>—</td>
</tr>
<tr>
<td>1993</td>
<td>54 930‡</td>
<td>148 000‡</td>
<td>202 920‡</td>
<td>259 100‡</td>
</tr>
<tr>
<td>Total</td>
<td>239 430</td>
<td>358 180</td>
<td>597 610</td>
<td>—</td>
</tr>
</tbody>
</table>

† Estimated from 4 km of roads, 9 m wide, at 3 kg m$^{-2}$ from a 1984 newspaper article.
‡ Taken from 1993 purchase requisition for 259 100 kg of brine.
§ Assumed to cover the same area as the 1992 application.
Vadose Zone Pore-Water Monitoring

Suction lysimeters have been used to collect moisture periodically from unsaturated sediment and basalt in the SDA since 1986 (Rawson et al., 1991). An inventory of lysimeters installed at the SDA is given in Hull and Bishop (2003), and the locations of wells containing lysimeters are shown in Fig. 4. From 1985 through 1987, 32 suction lysimeters with ceramic cups were installed in surficial sediments in and around the SDA, and seven deep lysimeters were installed in sedimentary interbeds (Hubbell et al., 1987; Laney et al., 1988). A slurry of silica flour or mud was placed around the lysimeter cups to ensure good hydraulic connection with the native rock or sediment. In 1986 and 1987, a 10 mg L\(^{-1}\) KBr tracer was added to the silica flour to determine when water used in the slurry was flushed from the silica flour and valid samples of pore water could be collected. Seven additional lysimeters were installed in surficial sediments between 1994 and 1998. During November 1999 through March 2000, 22 deep lysimeters were installed in and around the SDA to monitor vadose zone water near the B–C and C–D interbeds. The porous cups of these lysimeters are stainless steel.

Samples were collected approximately three times a year from 1986 to 1989. Routine sampling was discontinued in 1990, with only sporadic sampling conducted until 1998. Regular monitoring for radionuclide migration began in 1998, and the network of lysimeters for the B–C and C–D interbeds was significantly expanded in 1999. Routine sampling of the lysimeters is currently conducted quarterly.

Perched water is present in isolated lenses above the sediments comprising the B–C and C–D interbeds. Samples from the water perched above the C–D interbed are obtained with a bailer from Wells USGS-92 and 8802D (locations shown in Fig. 2). Well USGS-92 was installed in 1972 and Well 8802D in 1988. These perched water wells have slow recharge rates and thin depths of water, often limiting the volume of water collected in the bailer. Generally, samples can be collected at USGS-92, but success is sporadic at 8802D.

Soluble Salts in Sediments

We collected samples of surficial sediments at the SDA and analyzed the samples for soluble cations and anions as a method to test for brine persistence. We gathered these samples in two ways. First, we drilled new auger holes and collected samples using a split-spoon sampler at 12 locations in the SDA (Fig. 5) in January and February 1998. Second, we sampled cores of surficial sediments stored in the INEEL Core Library. Selected sedimentary material was sampled from the Core Library from three additional wells drilled between 1986 and 1990. This provided older sample material from locations near where the new wells were drilled to provide a comparison among the time intervals. Because the objective was to evaluate persistence of Cl\(^{-}\) in SDA surficial sediments and to evaluate the potential for lateral movement of the brine from the roads to buried waste, boreholes were located where geophysical measurements indicated elevated levels of ground conductance associated with roads, or where Cl\(^{-}\) contamination in suction lysimeters indicated brine migration and persistence. Details of the site selection process are given in Hull and Bishop (2003).
We collected 50 core samples from new boreholes in the SDA and eight samples from material in the Core Library. The sediment samples were extracted following the procedure for soluble salts outlined in Methods of Soil Analysis, Part 2 (Rhoades, 1982), using a ratio of one part sediment to five parts deionized water by weight. The sediment and water mixture was shaken on a shaker table for 1 h and then filtered. The extracts were analyzed for Na\(^+\), Ca\(^{2+}\), K\(^+\), Mg\(^{2+}\), Li\(^+\), Cl\(^-\), Br\(^-\), NO\(_3\)\(^-\), and SO\(_4\)\(^2-\). From these measured concentrations, we calculated the concentration of soluble salt in the sediment.

**RESULTS AND DISCUSSION**

**Brine in Vadose Zone Pore Water**

High levels of Cl\(^-\) detected in suction lysimeters were not initially linked to the brine applications. Once the possibility of the correlation was raised, we evaluated the possible link from chemical composition and the proximity of the elevated Cl\(^-\) to roads where brine was applied.

**Comparison of Pore Water to Brine Chemistry**

The brine contains Cl\(^-\) and Br\(^-\), both of which should be conservative species in vadose zone pore water (Hem, 1992; Davis et al., 1998). Because both chemicals are conservative, Cl/Br ratios are commonly used to identify the sources of Cl\(^-\) in groundwater (Edmunds, 1996; Davis et al., 2001). The correlation of Cl\(^-\) and Br\(^-\) in vadose zone pore water at the SDA is shown in Fig. 6. Two distinct linear trends are apparent in this figure. One group of samples with low Cl\(^-\) concentrations and...
Br\(^-\) concentrations between 0 and 10 mg L\(^{-1}\) plots along the Br\(^-\) axis. These data are from early samples from lysimeters where KBr was added to the silica flour as a tracer. The second distinct trend is for Cl\(^-\) and Br\(^-\) increasing to very high concentrations. We removed the early samples with very low Cl/Br mass ratios from the data set and fit a line to the remaining samples. The trend line, the solid line in Fig. 6, has a Cl/Br mass ratio of 480 \pm 28 (2 sigma). This is consistent with typical MgCl\(_2\) brine compositions (Table 1) as reported by the manufacturer.

Two conclusions can be drawn from Fig. 6. First, the early samples containing KBr tracer can be differentiated from other lysimeter samples based on the Cl/Br mass ratio. Second, we conclude that the samples with elevated Cl\(^-\) concentrations and a Cl/Br mass ratio of about 480 represent brine contamination. Based on this conclusion, essentially all water samples from the SDA with 350 mg L\(^{-1}\) or more Cl\(^-\) have been contaminated by brine. This latter conclusion is important because many of the samples taken in the past 18 yr were analyzed for Cl\(^-\) but not for Br\(^-\). The samples with brine contamination can be identified from the total Cl\(^-\) concentration as well as from the Cl/Br ratio.

The Cl/Br mass ratio vs. Cl\(^-\) for water samples collected from the SDA is plotted in Fig. 7. The SDA has three end-member waters: brine, the tracer used when the lysimeters were installed, and natural recharge water. Only the tracer is defined independently—it’s composition is known because it was added to the system during lysimeter installation. The compositions of the other two waters are, at least in part, inferred from the results of the SDA measurements. Although the manufacturer’s data (Table 1) give the ranges of Cl\(^-\) and Br\(^-\) concentrations in the brine, we estimate its Cl/Br mass ratio from measured pore water chemistry.

In general, pore water chemistry is derived from atmospheric deposition and reaction with minerals in the surficial sediments. For Cl\(^-\) and Br\(^-\), however, we assume no significant mineral sources in the sediment. Atmospheric deposition is recorded at Craters of the Moon National Monument, 38 km southwest of the SDA. The 20-yr precipitation-weighted average Cl\(^-\) concentration (Table 3) is 3.3 mg L\(^{-1}\) (0.093 mmol L\(^{-1}\)). No water in the SDA has a Cl\(^-\) concentration as low as 3.3 mg L\(^{-1}\). No Br\(^-\) has been measured for atmospheric deposition at Craters of the Moon. Potable groundwater away from coastal areas generally has Cl/Br mass ratios <200 (Davis et al., 1998, 2001). Davis et al. (2001) estimated the average Cl/Br ratio of recharge in Idaho to be between 100 and 200 with a single measured value of 168, so we assumed a Cl/Br mass ratio of 168 for recharge to the SDA. A number of different Cl\(^-\) concentrations were tried in mixing calculations. Based on the fit of the mixing equation to the data in Fig. 7, we adopted 8 mg L\(^{-1}\) as the Cl\(^-\) concentration for recharge. Using a Cl/Br mass ratio of 168, the Br\(^-\) concentration for natural recharge is then 0.05 mg L\(^{-1}\). Mixing between each pair of the three waters is plotted on Fig. 7. These lines bound most of the water analyses from the SDA. Many of the samples plot along the line between the brine composition and natural background water composition.

Water samples from the lysimeters were divided into three groups based on the Cl/Br ratio and the total Cl\(^-\) concentration: the early samples, which still contained KBr tracer; samples with >350 mg L\(^{-1}\) Cl\(^-\) and Cl/Br mass ratios near 480 that are identified as brine contaminated; and samples that, we conclude, represent natural pore water at the SDA. The brine-contaminated samples plot along the mixing line between brine and natural recharge, or somewhat below the mixing line where mixing with KBr tracer may have decreased the Cl/Br mass ratio (Fig. 7). The range of natural pore waters presents a wide range in Cl/Br mass ratios. Some of this may be because samples classified as pore water on the basis of the dominant chemical pattern contain a small fraction of brine or KBr tracer. There is also a significant uncertainty, and likely variation, in the Cl/Br ratio of atmospheric deposition. The natural pore waters do not appear to show a simple pattern that reflects a dominant process, such as evaporation, controlling the water chemistry.

### Spatial Distribution

Because the brine was used on roads, there should be a correlation between proximity to roads and brine contamination. A map of the SDA showing the roads

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**Table 3. Precipitation-weighted average chemical composition of atmospheric deposition at Craters of the Moon National Monument for 1980 to 2000.**

<table>
<thead>
<tr>
<th>Cations</th>
<th>Anions</th>
<th>mmol L(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca(^{2+})</td>
<td>SO(_4^{2-})</td>
<td>0.061</td>
</tr>
<tr>
<td>NH(_4^+)</td>
<td>NO(_3^-)</td>
<td>0.108</td>
</tr>
<tr>
<td>Na(^+)</td>
<td>Cl(^-)</td>
<td>0.094</td>
</tr>
<tr>
<td>Mg(^{2+})</td>
<td>Cl(^-)</td>
<td>0.013</td>
</tr>
<tr>
<td>K(^+)</td>
<td></td>
<td>0.008</td>
</tr>
</tbody>
</table>
and well locations, and identifying the wells contaminated with brine, is shown in Fig. 8. The wells with contamination are primarily along the main east–west road and in the southeast corner of the SDA. For wells in surficial alluvium, the median distance to contaminated wells from treated roads is less than the median distance to uncontaminated wells (Table 4). All the wells <13 m from a treated road are contaminated. All the wells >33 m from a treated road are uncontaminated. For wells between 13 and 33 m, three are uncontaminated and two are contaminated. From the distribution of distances in Table 4, we conclude that brine can readily move lateral distances on the order of 13 m in the surficial alluvium, and may move up to 33 m.

Chloride from MgCl₂ applications was detected in the deep vadose zone below the surficial sediments at two perched zone monitoring wells and two lysimeters within a few years after brine was applied to roads. The perched water monitoring wells are Well 77-2, completed at a depth of 27 m and at a distance of 22 m from the road, and Well 8802D, completed at a depth of 67 m and a distance of 7 m from the road. Migration of water from the road to these wells is mainly downwards. The two lysimeters are DL02 in Well D-06 at a depth of 13 m and DL04 in Well TW-1 at a depth of 31 m. These lysimeters are 80 to 110 m north of the nearest treated road, so there is an appreciable lateral component in the subsurface flow reaching these lysimeters. While some lateral movement of water may occur by capillary forces for unsaturated sediments, this degree of lateral migration indicates saturated flow across the top of perching layers such as the A basalt flow, the A–B interbed, and possibly the B–C interbed.

For lysimeters installed in the deep vadose zone below surficial sediments, brine contamination is aligned along the main east–west road through the SDA. These areas underlie areas of brine contamination in the surficial alluvium. North of the road, Wells D-06, TW-1, I-4D, and I-4S in the deep vadose zone have elevated Cl⁻

<table>
<thead>
<tr>
<th>Well</th>
<th>Distance to nearest road</th>
<th>Well</th>
<th>Distance to nearest road</th>
</tr>
</thead>
<tbody>
<tr>
<td>98-1</td>
<td>17</td>
<td>TH05</td>
<td>188</td>
</tr>
<tr>
<td>98-4</td>
<td>9</td>
<td>W02</td>
<td>104</td>
</tr>
<tr>
<td>PA01</td>
<td>12</td>
<td>W04</td>
<td>17</td>
</tr>
<tr>
<td>PA02</td>
<td>5</td>
<td>W06</td>
<td>29</td>
</tr>
<tr>
<td>TH04</td>
<td>11</td>
<td>W08</td>
<td>33</td>
</tr>
<tr>
<td>W03</td>
<td>1</td>
<td>W09</td>
<td>121</td>
</tr>
<tr>
<td>W05</td>
<td>25</td>
<td>W13</td>
<td>60</td>
</tr>
<tr>
<td>W23</td>
<td>33</td>
<td>W25</td>
<td>13</td>
</tr>
<tr>
<td>Median</td>
<td>11</td>
<td>Median</td>
<td>46</td>
</tr>
</tbody>
</table>

Table 4. Distances from wells in the surficial alluvium to the nearest road where brine was applied.
levels and underlie the contaminated Wells PA-01 and PA-02 in the surficial alluvium. At the west end of the SDA, Wells I-1S and O-4 have elevated Cl\(^{-}\) levels and underlie the shallow contaminated Wells 98-5 and W-23. Snowmelt accumulates along the main east–west road (Fig. 3), and recharge has been estimated to be significant in these areas (Martian, 1995; Bishop, 1998). The vertical migration of Cl\(^{-}\) to the B–C and C–D interbeds is attributed to significant infiltration along the drainage ditches.

**Temporal Changes**

Figure 9 shows the Cl\(^{-}\) concentration with time at four depths in two adjacent wells in the surficial alluvium at the west end of the SDA. Well 23, drilled in 1985, has lysimeters at 5.7, 3.6, and 2.4 m; Well 98-5, drilled nearby in 1998, has a lysimeter at 3.2 m. Chloride was elevated at depths of 2.4 and 3.6 m in the late 1980s. (Brine was applied to roads between 1984 and 1993.) A number of samples collected in the intervening years show a declining trend in Cl\(^{-}\) at both depths after 1993. Lysimeter 98-5-L39, installed in 1998, shows concentrations consistent with results from Well W23. Recent samples collected from Lysimeter L07, at 5.7 m, are also low in Cl\(^{-}\), indicating that the Cl\(^{-}\) has been flushed from this depth also. The Cl/Br mass ratios of the samples confirm that the elevated concentrations represented brine contamination. The ratio was around 480 for the early samples and has dropped to <400, and as low as 160, for recent samples. These trends indicate that brine takes several years to flush from the shallow vadose zone in the west end of the SDA.

The lysimeters in Wells D-06 and TW-1 were installed in the deep vadose zone in the late 1980s, and water samples have been collected periodically from Lysimeter DL02 (depth 13.4 m) and DL04 (depth 31 m). Both lysimeters showed increasing Cl\(^{-}\) concentrations very soon after sampling began (Fig. 10). The initial low Cl\(^{-}\) concentration in Lysimeter DL04 at 31 m is associated with Cl/Br mass ratio near 0 (Fig. 11) and reflects the KBr tracer solution that was used to wet the silica flour when the lysimeter was installed. For Lysimeter DL02, at 13.4 m depth, the initial Cl\(^{-}\) concentrations are well above 350 mg L\(^{-1}\) and the early Cl/Br ratio is near 480 (Fig. 11). However, the first two samples collected from Lysimeter DL02 were not analyzed for Br\(^{-}\), so the Cl/Br ratio cannot be calculated for two Cl\(^{-}\) values near 1600...
Fig. 12. Plot of soluble Cl\(^{-}\) and soluble Br\(^{-}\) in Subsurface Disposal Area surficial sediments.

and 3200 mg L\(^{-1}\). Therefore, while the early samples reflect mixing, we cannot determine if this is mixing of brine with the KBr tracer solution or the native pore water. Because of this, we cannot interpret the rise in Cl\(^{-}\) in these two lysimeters as initial breakthrough of brine or for the purpose of defining flow rates to these depths. A couple of samples from Lysimeter DL01 (depth 26.8 m), starting in 1998, show similar levels of Cl\(^{-}\) to Lysimeter DL04 at about the same depth. Brine was first applied to roads in the SDA in 1984. By 1987, the brine was in lysimeters at depths of 13 and 31 m. Chloride has persisted in these lysimeters since that time. It appears that Cl\(^{-}\) has started to move out of the 13-m level (Lysimeter DL02).

Brine Extracted from Sediments

To collect data more directly associated with treated roads, we augered boreholes through roadbeds, in drainage ditches along roads, and in the surficial sediment adjacent to the drainage ditches. We collected sediment in split-spoon samplers and extracted soluble salts from the sediment. Some older sediment samples, taken from sample archives, were also evaluated to determine concentrations of soluble salts.

Soluble salt concentrations in the sediments varied widely among boreholes, and among depths in the same borehole. Chloride concentrations ranged from <1 to 2104 mg kg\(^{-1}\). If the elevated soluble salts are from brine, then the Cl/Br mass ratio should be close to 480. A plot of Br\(^{-}\) against Cl\(^{-}\) shows a good correlation between Br\(^{-}\) and Cl\(^{-}\) (Fig. 12). A regression of Br\(^{-}\) on Cl\(^{-}\) gives a Cl/Br mass ratio (shown by the line) of 433 ± 82 (2 sigma), a value that is not significantly different than the ratio for lysimeter samples of 480 ± 28. This value is also consistent with the range of brine concentrations shown in Table 1.

Soluble Chloride Distribution in Sediments

Main East–West Road

Three boreholes were drilled and sampled along the main east–west road through the SDA, and a core from a borehole drilled near Pit 9 was sampled (Fig. 5). Samples from the boreholes (SDA-11, SDA-12, 98-5) had only background or very low levels of Cl\(^{-}\) (Fig. 13). The Cl\(^{-}\) concentrations near the surface in Borehole 98-5 are slightly above background, indicating some residual brine in the sediment. Boreholes SDA-11 and SDA-12 were drilled in a location where water accumulates in drainage ditches along roads (Fig. 3) and that has been identified as an area of elevated infiltration (Bishop, 1998). If Cl\(^{-}\) had been introduced to these sediments from brine, it was flushed from the system between 1993 and 1998. At the eastern end of the SDA, the sediments contained elevated Cl\(^{-}\) in 1990, when the borehole Pit 9 P05 was drilled. Chloride was not flushed from this borehole between 1985 and 1990, a period equivalent to the period between brine application and sampling for the other boreholes. The elevated Cl\(^{-}\) in borehole Pit 9 P05 confirms water samples collected from a lysimeter in nearby Well W03 in October 1989, which contained Cl\(^{-}\) concentration of 13 000 mg L\(^{-1}\). Well W03, and its lysimeter, were destroyed by construction of the Pit 9 project, so there is no information from this area to evaluate changes that may have occurred since then.

Central Portion of the SDA

Extractable Cl\(^{-}\) concentrations in Boreholes SDA-02, SDA-03, and Acid Pit 4, in the central portion of the SDA, are shown in Fig. 14. SDA-02 was drilled in the middle of the road 5 yr after the last brine application and shows moderately elevated Cl\(^{-}\) concentrations in the roadbed to a depth of 2 m. Below 3 m, the samples reflect background Cl\(^{-}\) concentrations. In Boreholes SDA-03 and Acid Pit 4, drilled adjacent to the road, Cl\(^{-}\) concentrations are within background or slightly elevated. At this location, there is little evidence of lateral spread of Cl\(^{-}\).

Boreholes SDA-07 and 98-4 were drilled in and just behind a drainage ditch (Fig. 5) along a road treated...
with brine. Chloride concentrations in these boreholes are slightly above background levels at all depths sampled (Fig. 14). Boreholes 98-4 and SDA-07 show similar patterns to SDA-03 and Acid Pit 4. For this part of the SDA, it appears that the roadbed retains some of the MgCl₂ brine. Brine has migrated to depths several meters below drainage ditches, and laterally several meters. Concentrations in the sediment, however, are on the order of a few dozen milligrams per kilogram and are relatively low.

Well 98-3 was drilled in an area where snowmelt runoff can accumulate (Fig. 3), but is not an area of high recharge (Martian, 1995). Some 5 yr after brine application, Cl⁻ was in the sediment at Well 98-3 all the way down to the sediment–basalt interface at a depth of about 7 m. This distribution seems to support Martian’s conclusion that this is not an area of high recharge. In areas of high recharge such as at Boreholes SDA-11 and SDA-12, Cl⁻ was flushed from the sediment by 1998 (Fig. 13). Here, sufficient recharge has occurred to carry Cl⁻ to a depth of 7 m, but not enough to flush Cl⁻ out of the sediments.

**Southern Boundary of the SDA**

Samples taken along the southern boundary of the SDA show the highest levels of extractable Cl⁻ (Fig. 15). Borehole W05 was drilled in 1986, and Borehole 98-1 was drilled nearby in 1998. Peak Cl⁻ concentrations in the two wells are similar, and significant Cl⁻ remains...
in the pore water and in the sediment. The persistence of Cl\(^{-}\) in pore water and sediment indicates that this is an area of relatively low recharge.

Brine was applied to the road exiting the southern SDA boundary at least three, and possibly four, times. Samples collected from the middle of the roadway in Borehole SDA-05 show that the roadbed has high levels of Cl\(^{-}\). Borehole SDA-04, drilled in the drainage ditch along the road, has even higher soluble Cl\(^{-}\) levels, up to 2100 mg kg\(^{-1}\) (Fig. 15). The sediment in Borehole 98-2, drilled behind the drainage ditch, contains elevated extractable Cl\(^{-}\) at a depth of 1.5 to 2 m, but the concentrations are an order of magnitude lower than in the drainage ditch. At this location, Cl\(^{-}\) migrated into the drainage ditch, but very little Cl\(^{-}\) has migrated further away from the road.

**SUMMARY**

Between 1984 and 1993, an estimated 600 t of MgCl\(_2\) brine were applied to roads in and around the INEEL's radioactive waste Subsurface Disposal Area to control airborne dust. The locations where brine was applied were obtained from company records. Data on the persistence and distribution of Cl\(^{-}\) were gathered using water samples from suction lysimeters and soluble ions extracted from sediments. Suction lysimeters at the SDA have been sampled periodically for major cation and anion composition since 1986. Chloride/bromide mass ratios show that the MgCl\(_2\) brine significantly affects vadose zone water chemistry at the SDA. Elevated Cl\(^{-}\) concentrations were found in the surficial alluvium along many of the roads in the SDA. Lateral brine migration in the surficial sediments is commonly on the order of 13 m, with migration up to 33 m in some locations. Evidence of brine has been found in lysimeters and perched water monitoring wells to depths of 67 m. Chloride from the brine moved downwards very rapidly and was detected in a lysimeter at 31 m depth in 1987, no more than 3 yr after the brine was first applied. Lateral migration to lysimeters installed in the A–B and B–C interbeds 80 to 110 m away from roads indicates development of perched water zones and lateral transport beneath the SDA. These results demonstrate that conclusions drawn from larger-scale infiltration and tracer tests (Dunnivant et al., 1998; Nimmo et al., 2002) conducted near the SDA are also applicable for the smaller-scale and natural recharge conditions beneath the SDA.

Samples collected from roadbeds in Boreholes SDA-02 and SDA-05 show that Cl\(^{-}\) has penetrated up to 2 m into the roadbed, and is present in concentrations on the order of one to several hundred milligrams per kilogram. This is at least an order of magnitude lower than initial concentrations calculated on the basis of brine application practices. At SDA-02, where samples were collected several meters below the roadbed, the deeper samples are at background levels. Samples collected from drainage ditches along roadbeds show more vertical movement of brine contamination. Where recharge through the ditch is high, such as along the main east–west road, brine has been flushed out of the sediments (98-5, SDA-11 and SDA-12). Where recharge is lower, the amount of Cl\(^{-}\) under drainage ditches is highly variable. However, in sediments from Borehole SDA-06 and Well 98-3, there are high concentrations of Cl\(^{-}\) to depths of up to 7 m. Chloride concentrations are significantly lower on the side of the drainage ditch away from the roadbed (98-2, 98-4) than in the road or drainage ditch. For the most part, vertical migration of brine appears to be occurring under drainage ditches adjacent to the roads where brine was applied. Locations where Cl\(^{-}\) persists in surficial sediments generally agree with locations where recharge is calculated to be lower through the surficial sediment. Areas where Cl\(^{-}\) has been flushed from the surficial sediment are in areas identified as high-recharge areas (Martian, 1995; Bishop, 1998).

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**REFERENCES**


Hansen, E.D. 1982. The use of magnesium chloride for dust treatment for surface stabilization of forest service roads. USDA, Forest Service. Intermountain Region, Ogden, UT.


Hubbell, J.M. 1995. Perched groundwater monitoring in the subbur-


Skorseth, K. 2000. Gravel roads, maintenance and design manual. South Dakota Local Transportation Assistance Program, South Dakota State University, Brookings, SD.
