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Chloride-Mass-Balance for Predicting Increased Recharge after Land-Use Change

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ABSTRACT

The chloride-mass-balance (CMB) method has been used extensively to estimate recharge in arid and semi-arid environments. Required data include estimates of annual precipitation, total chloride input (from dry fallout and precipitation), and pore-water chloride concentrations. Typically, CMB has been used to estimate ancient recharge but recharge from recent land-use change has also been documented. Recharge rates below a few mm/yr are reliably detected with CMB; however, estimates above a few mm/yr appear to be less reliable. We tested the CMB method against 26 years of drainage from a 7.6-m-deep lysimeter at a simulated waste-burial ground, located on the Department of Energy's Hanford Site in southeastern Washington State, USA where land-use change has increased recharge rates. Measured drainage from the lysimeter for the past 26 years averaged 62 mm/yr. Precipitation averaged 190 mm/yr with an estimated chloride input of 0.225 mg/L. Initial pore-water chloride concentration was 88 mg/L and decreased to about 6 mg/L after 26 years, while the drainage water decreased to less than 1 mg/L. A

recharge estimate made using chloride concentrations in drain water was within 20% of the measured drainage rate. In contrast, recharge estimates using 1:1 (water: soil) extracts were lower than actual by factors ranging from 2 to 8 or more. The results suggest that when recharge is above a few mm/yr, soil water extracts can lead to unreliable estimates of recharge. For conditions of elevated recharge, direct sampling of pore water is the preferred method, because chloride concentrations are often 20 to 50 times higher in directly-sampled pore water than in pore-water extracts.

INTRODUCTION

In the chloride mass balance (CMB) method, measurements of chloride in pore water are used to estimate the recharge rate when both precipitation and chloride inputs are known. The CMB for a soil profile at steady state can be written as:

$$P\ (Cl_p) = R\ (Cl_s) \qquad (1)$$

where P = average annual precipitation (mm/yr)

Cl_p = average chloride input from all sources (mg/L)

Cl_s = average chloride concentration of pore water below the root zone (mg/L)

R = average annual recharge rate (mm/yr).

Key assumptions are: 1) steady influx of water and chloride; 2) steady, vertical efflux (recharge) of chloride below the root zone; 3) no soil sources or sinks for chloride, and

4) piston flow of chloride such that point measurements of solute concentrations can be used to represent a true spatial average of the soil chloride flux. CMB has been used to estimate recharge in arid and semi-arid regions throughout the world (Allison et al. 1994; Prych 1995; Murphy et al. 1996; Hendrickx and Walker 1997; Scanlon et al. 1997). The CMB method appears to be useful in estimating paleoclimate recharge rates dating back thousands of years (Stone 1984; Murphy et al. 1996; Tyler et al. 1996), but CMB also has been used for estimating modern recharge rates, including those that have increased in response to land-use change (Jolly et al. 1989; Walker et al. 1991). Because of its simplicity, CMB is an attractive method. With proper assumptions about average annual precipitation and chloride inputs, the only direct measurement required is the chloride concentration of the pore water.

The general principles of the CMB method have been known and practiced in irrigation management for years (U.S. Department of Agriculture [USDA] Handbook 60, 1954). In a manner similar to the CMB method, salt mass balance in irrigation water is often expressed as the leaching requirement (LR), defined as the applied water volume (Q) times the input salt concentration (Ci) divided by the drainage volume (D) times the salt concentration (Cs) of the drainage water i.e.,

$$\mathrm{LR} = \frac{\mathrm{Q} \times \mathrm{Ci}}{\mathrm{D} \times \mathrm{Cs}} \qquad (2)$$

For an LR of 1 and considering only chloride, Equation 1 is identical to Equation 2.

Pore water chloride is typically obtained from soil samples taken at depths well below the root zone and analyzed first for water content (gravimetrically) and then for chloride with chloride determined typically from a 1:1, 2:1 or sometimes 3:1 (solution: soil) extract (Murphy et al. 1997; Scanlon and Goldsmith 1997). The chloride concentration, Cl_s (in mg Cl/L soil solution), is subsequently computed by dividing the measured extract concentration by the gravimetric water content (g water/ g oven-dry soil) and multiplying by the solution: soil ratio.

This method works best at high-salt concentrations and low recharge rates because, as the chloride concentration approaches the detection limit of the analytical method used, analytical errors significantly increase the uncertainty of the recharge estimate (Tyler et al. 1999). Also, at low concentrations, systematic contamination of sample and extracts can become an important source of error. Because most analytical methods have an operational resolution for chloride between 0.02 mg/L and 1 mg/L, it should be apparent that for estimates of soil pore water chloride, uncertainties mount rapidly as the recharge rate increases above 1 mm/yr and extract concentrations drop below 1 mg/L. In many soils, there are chloride minerals that have varying dissolution rates. In addition, treatment of a soil low in chloride with deionized water can potentially release mineral chloride (Murphy et al. 1996). For finer textured soils, such as silts or clays, the practical recharge limit increases because the field water content increases. Another consideration for soils with much clay is the possibility of anion exclusion, which could adversely affect the recharge estimate using the CMB method (Gee and Hillel 1988). This is true because anion exclusion from the clay double-layer creates elevated chloride concentrations in the pore water resulting in an underestimation of recharge rate. The impact is greatest when the water content is lowest. For clay-rich soils, pore water samples obtain from direct extraction (e.g., centrifugation) would be impacted the most.

In some coarse-textured soils there is evidence that unstable (e.g., finger) flow, funnel flow or preferential flow through worm holes, or root channels, may cause non-piston type flow of solutes in and below the root zone. Allison et al. (1994) provide examples of extreme heterogeneities in field soils giving rise to highly variable chloride concentrations. McCord et al. (1997) showed that textural variations can lead to zones of slowly leached soil under steady-flux conditions. Roth (1995) also showed that flow channeling can occur in mildly heterogeneous soils, which can significantly increase the time required for solute concentrations to equilibrate throughout the soil profile. The successful application of CMB at a given site is therefore dependent upon site conditions which may or may not cause preferential flow of solutes.

Chloride mass balance has been applied for years to estimate recharge, but in situations where there has been land-use change and recharge has increased, there has been few if any attempts to compare CMB with independent estimates of recharge. In this paper, we tested the CMB method for predicting recharge in a semi-arid climate setting, where land use has changed and where the chloride flux could be quantitatively checked with lysimetry, a method that can be used to directly measure the percolation of water through soils and determine both the flux rate and soluble constituents removed in the drainage. (Gee and Hillel 1988; Gee et al. 2003).

METHODS

The study was conducted at the U. S. Department of Energy’s Hanford Site, near Richland, Washington, USA. The Hanford Site has a northern, steppe (semi-arid) climate typified by dry, hot summers and cool, wet winters (Hoitink et al. 2003). For the past 25 years, the precipitation at the Hanford Meteorological Station has averaged 180 mm/yr, about two thirds of this amount coming in winter months (Nov. through Mar). Potential evaporation, controlled by the aridity of the climate, is about 1600 mm/yr, nearly 9 times the annual precipitation (Gee et al. 1989). Actual evaporation (AE) is significantly different than potential at Hanford. For undisturbed sites, with shrub-steppe vegetation, AE is approximately equal to annual precipitation, so we would expect little drainage. Chloride measurements made at the Hanford site in areas of undisturbed shrub-steppe vegetation growing on coarse soils have shown significant bulges of high chloride (> 100 mg/L) at shallow depths with corresponding recharge rates estimated to be less than 1 mm/yr (Prych 1995, Murphy et al. 1996, Fayer et al. 1999). In contrast, for disturbed sites with little or no vegetation and coarse soils, AE can be less than two-thirds of the annual precipitation (Gee et al. 1992) resulting in drainage rates that exceed 50 mm/yr for the Hanford Site.

The Hanford Meteorological Station (HMS) is located about 15 km northwest of the simulated waste site. Continuous records of precipitation have not been kept at the lysimeter test site because of a hiatus in project funding over the years. However, precipitation at the lysimeter test site was previously found to be about 6% more than at the HMS (Gee et al. 1989), so we have estimated the precipitation to be 190 mm/yr for the past 20 years. Effective chloride concentration in precipitation has been studied extensively by Murphy et al. (1996) and found to range from 0.220 mg/L to 0.226 mg/L. For the purposes of this study we selected 0.225 mg/L for the effective chloride concentration of precipitation at the lysimeter site.

Soil samples were taken at a simulated waste-burial-ground about 5 km north of Richland from two sand-filled, 7.6-m-deep, lysimeters that were kept vegetation free for the past 25 years. The sand is screened material containing only 1% gravel (material between 2 mm and 10 mm) taken from the lysimeter excavation and is Hanford formation material that is largely granitic in origin (Baker et al. 1991). The lysimeters were installed in 1978 and began draining in 1981 (Gee et al. 1989, Tyler et al. 1999). . Figure 1 shows the cross-section of the lysimeters along with an instrument caisson that provided entry for collecting drainage water and for soil sampling. Most of the sampling was done in the south lysimeter (Figure 1) by coring through the side ports (in 1996, 1998) or by direct coring using a split-spoon sampler (in 2002). Some check samples for chloride analysis of the pore water were also taken from the north lysimeter. The lysimeter soil is classified as a sand, and key hydraulic properties are listed in Table 1. During the past 20 years, the lysimeter drainage has averaged 34% of the total precipitation or about 55% of the winter precipitation.

In December 1996, soil cores were taken from the south lysimeter at 10 depths, from the surface to a depth of 7.1 m. Each soil sample was analyzed for water content and then a 1:1 (solution:solid) extract was prepared and analyzed for chloride using a Dionex ion chromatograph with a resolution of ±0.02 mg/L chloride. Similarly, samples were taken in March 1998 and then again in September of 2002. Samples in 1996 and 1998 were taken from the side ports of the south lysimeter. Samples taken in 2002 were taken using a split-spoon sampler from the center of the south lysimeter. The samples taken in March 1998 were analyzed in an aseptic environment free from significant potential for chloride contamination. The samples taken in March 1998 were also not oven dried but brought to 1:1 dilution using known water contents obtained from sub samples. The other two sampling sets (December 1996 and September 2002) were oven dried and analyzed in conventional laboratory settings.

RESULTS AND DISCUSSION

Table 2 shows the chloride concentrations for the soil samples and drainage water collected at three dates. Recharge estimates from soil cores are presented in Table 3 along with recharge estimates based on chloride in the drainage water. These values are compared to the measured drainage rate. It is apparent that the soil-core estimates underestimate the recharge by a factor of 7 or more. In an attempt to explain the underestimation, we compared the soil pore-water estimates with numerical simulations.

The simulations were done for a period of 26 years (1978 through 2003). Using the available soil hydraulic properties from the lysimeter and assuming a constant recharge flux of 62.5 mm/year, numerical simulations of the leaching of the lysimeter were conducted using STOMP (White and Oostrom, 2004), which is designed to solve a variety of nonlinear, multiple-phase, multi-dimensional flow and transport problems for unsaturated porous media. The transport was modeled as a one-dimensional process and we assumed transport to be controlled by advection and dispersion alone. The initial chloride concentration in the lysimeter in 1978 was taken to be 88 mg/L based on chloride analysis of archived soil from the lysimeter, and the chloride input was assumed to be steady at 0.225 mg/L based on the analysis of Murphy et al. (1996).

We ran the simulations two ways. Case 1 assumed the net infiltration was steady at 62.5 mm/yr. Case 2 assumed the net infiltration varied with precipitation input, with the net infiltration mimicking the variability observed in the annual drainage rates. Figure 2 shows the simulated chloride concentrations in the drainage water as a function of time and Figure 3 the chloride concentration profiles at two selected times. The variable net infiltration case (case 2) yielded the lowest chloride concentrations and approached a steady state lowest value more quickly than when the net infiltration was assumed to be steady. By 1998 and subsequently as late as Sept. 2002, the drainage water concentrations simulated in the model ranged from 0.8 mg/L to 0.7 mg/L. The measured value in the drainage water for the same time period ranged from 0.8 to 1.0 mg/L. In contrast the 1:1 pore water extracts gave concentration estimates of 5 mg/L chloride, more than 6 times that measured in the drainage water.

To further investigate the discrepancy between the soil chloride concentrations and the observed drainage flux, chloride transport was modeled using a mobile/immobile water-transport approach (van Genuchten and Wagenet, 1989). Simulations were conducted with CXTFIT (Toride et al., 1995) under conditions of steady drainage (65 mm/year) over 19 years of drainage (1978 through 1996). Previous laboratory data (Gee and Campbell, 1980) suggested that the ratio of mobile water to total water content was approximately 0.65. Simulations using this ratio failed to produce any similarities to the observed drainage and soil-sample concentrations. Only when the ratio of mobile to total water content was increased to 0.9 and an extremely slow exchange between the mobile and immobile phase was chosen, could simulated soil-water and drainage-water concentrations be well matched to those observed in the 1:1 dilutions and drainage water. It should also be noted that the simulated lower boundary condition was that of unit gradient condition; however, this is not sufficient to explain the need for very small immobile water contents nor the extremely slow exchange between the phases.

We also ruled out preferential flow as an explanation of the high chloride concentrations found in the soil samples. The chloride concentrations of side port samples taken in 1996 were nearly the same as those found in borehole samples taken from the center of the lysimeter in 2002. If there were wall effects or other preferential flow occurring, it should have shown up as marked differences in the two tests.

The reported 1:1 soil chloride concentrations are always too high to correctly predict the drainage. To match the measured recharge rate, the 1:1 extract chloride concentrations would have to be 0.041 mg/L or lower, compared to measured values, which were as high as 0.5 mg/L or more. Table 4 shows what the pore water and 1:1 concentrations would be for various recharge rates. It is clear that as the recharge rate increases, the pore water chloride becomes more dilute. At 100 mm/yr, 1:1 extracted chloride is at the detection limit for the Dionex ion chromatograph. Care must be taken to obtain reliable results because the 1:1 chloride values are in the few mg/L range or less for all recharge values above 1 mm/yr. The data from Mar. 1998, where samples were run in an aseptic manner in a low chloride environment, suggests that chloride additions from sources other than precipitation and fallout is likely the explanation for the higher chloride numbers found in Dec. 1996 and Sept. 2002. Even the 1998 soil chloride data underestimate the measured drainage rate by more than a factor of 2.

We have considered mineral dissolution as a possible mechanism for some of the high chloride numbers. Fayer et al. (1999) reported chloride concentrations in Hanford formation minerals ranging from 100 ppm to 230 ppm. If 0.1% of the chloride leached from the mineral, this would account for a soil-solution concentration ranging from 0.100 mg/L to 0.230 mg/L. Such levels of chloride could possibly have leached from the samples, particularly after they had been dried and subsequently wetted with DI water. The fact that the samples in 1998 were not oven dried and gave the best results, suggest that oven drying may release chloride from the sediments tested. While we have not exhaustively tested this hypothesis, we suspect that mineral dissolution contributed to the high chloride values obtained with the 1:1 extracts. Additional chloride contamination from outside sources at the very low levels of chloride is also possible.

Table 2 shows the chloride results of samples taken from the lysimeter in 1996, 1998, and 2002, 5 years apart, and Table 3 shows the estimates of recharge using the soil cores and the drainage water. Differences in soil-water concentrates are attributed to differences in laboratory analysis. Samples taken in March 1998 were run in an aseptic environmental laboratory in Reno, Nevada; the other two sets were run in more conventional laboratory settings. Table 3 shows the estimated recharge values determined from the soil cores and the drain water. The average values of these recharge estimates are then compared to the measured value (62.5 mm/yr). The drainage water is within 22% of the measured value, while the pore water underestimates the actual drainage by values ranging from more than a factor of 2 to more than 8 fold. Scanlon and Goldsmith (1997) observed similar underestimation of recharge from 3:1 chloride extracts in a playa lake study in Texas.

Based on these observations, we recommend caution in using soil pore-water sampling in the CMB method for recharge rates much above a few mm/yr. It is further recommended that minimum soil-water dilutions be used when measuring chloride concentrations in soils to reduce the impacts of analytical errors and possible dissolution. It is possible that dilution errors can be eliminated in the laboratory by centrifuging field-most samples (when the sample is wet enough), or by obtaining pore water directly in the field using solution sampling or wick lysimetry (Gee et al. 2003). It is clear for our study that at concentrations below 1 mg/L, chloride sources other than from precipitation and fallout may contribute to errors in estimating recharge using the CMB method.

CONCLUSIONS

The CMB method was tested using a deep lysimeter at a site where land-use changes increased recharge from a very low rate (less than a few mm/yr) to a much higher rate (more than 60 mm/yr). Using the chloride data from the lysimeter drainage as the true estimate of the pore water, recharge estimates agreed within 20% of the measured drainage rate. In contrast, soil samples subjected to leaching (1:1) yielded pore-water estimates that always underestimated the drainage by a factor of 2 or more. The apparent error in pore-water chloride concentrations is attributed to dilution effects coupled with mineral dissolution and possibly other external sources of chloride in sampling low concentration chloride. The data show that the CMB method can be used where land use has caused increased recharge rates over relatively short time periods (decades) but that collecting pore-water samples from drainage is preferred over conventional soil-water extraction procedures. When recharge rates are much above a few mm/yr, it appears that dilution effects and possible chloride contamination from mineral dissolution and others sources can cause pore-water extracts to give unreliably low estimates of recharge.

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Figures

Figure 1. Schematic of the cross-section of two deep lysimeters at a simulated waste burial ground at the U. S. Department of Energy's Hanford Site near Richland, Washington.

Figure. 2. Time course of the simulated chloride concentration at the drainage.

Figure 3. Simulated chloride concentration profile in March 1998 and September 2002.

Tables

Table 1. Soil texture and hydraulic properties of the south lysimeter soil.

Table 2. Pore-water chemistry of soils taken from the south lysimeter for selected times.

Table 3. Comparison of measured and estimated drainage rate (mm/yr) from CMB recharge estimates using drain-water and pore-water data sets. Assumes 190 mm/yr average precipitation with a) 0.225 mg/L average chloride input or b) 0.275 mg/L average chloride input.

Table 4. Recharge rates and chloride concentrations expected for 1:1 extracts under Hanford climate and estimated chloride input.

Table 1. Soil texture and hydraulic properties of the south lysimeter soil.

Texture (USDA)	K sat (cm/s)	K unsat (cm/s) at–5 kPa	Theta at–5 kPa
Sand*	2E-03	1E-07	0.10

* Separates: Gravel, 1%; Sand 95%, Silt 3%, Clay 1%

Table 2. Chloride concentrations (mg/L) found in soils and drain water taken from the south lysimeter at selected times.

Sample Depth (m)	Dec. 1996	Mar. 1998	Sept. 2002
0.0–0.5 m	7.0	-	-
0.5–1.0 m	5.0	-	-
1.0–1.5 m	3.2	1.5	6.1
1.5–2.0 m	4.8	1.6	5.6
2.0–2.5 m	5.8	-	3.9
3.0–3.5 m	6.7	-	6.7
4.0–4.5 m	5.7	1.5	4.6
5.5–6.0 m	7.8	1.6	4.9
6.5–7.0 m	7.4	-	4.7
7.0–7.5 m	4.5	-	5.2
Drain Water	1.2	0.8	0.9

Table 3. Comparison of measured and estimated drainage rate (mm/yr) from CMB recharge estimates using drainage water and 1:1 extract (pore water) data sets. Assumes 190 mm/yr average precipitation with a) 0.225 mg/L average chloride input or b) 0.312 mg/L average chloride input.

a) 0.225 mg/L Chloride Input	Dec. 1996	Mar. 1998	Sept. 2002	Average
Soil Samples	7	25	8	13
Drain Water	37	53	48	46
Measured	64	66	61	62*
b) 0.275 mg/L Chloride Input				
Soil Samples	10	37	11	19
Drain Water	50	74	66	62
Measured	64	66	61	62*

* 26 year average

Table 4. Recharge rates and chloride concentrations expected for 1:1 extracts under Hanford climate and estimated chloride input.

Recharge Rate (mm/yr)	Pore Water Chloride (mg/L)	1:1 Extract (mg/L)
1	40	2.0
3	14	0.68
10	4	0.20
30	1.4	0.068
100	0.4	0.020

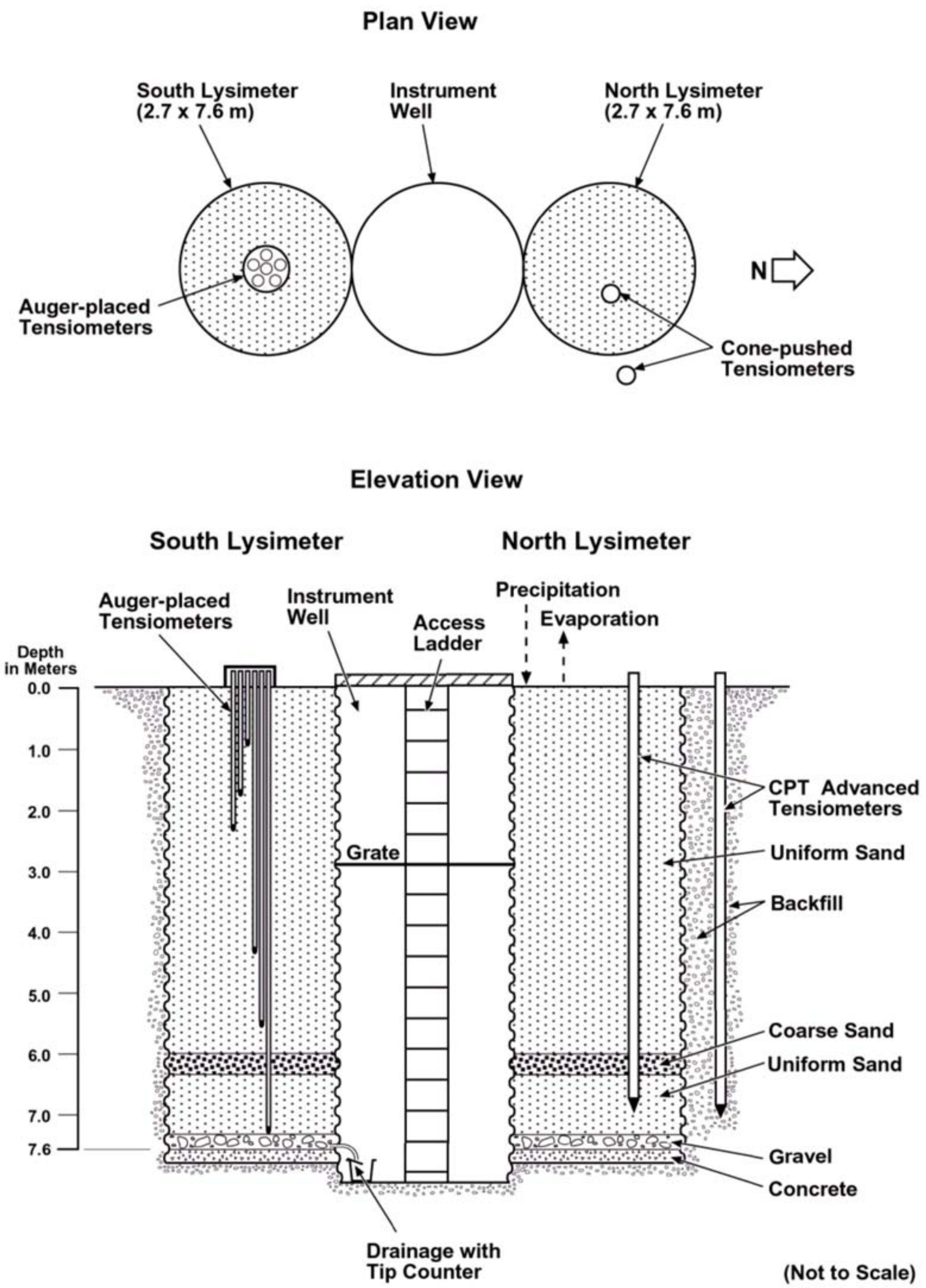

Figure 1. Schematic of the cross-section of two deep lysimeters at a simulated waste burial ground at the U.S. Department of Energy’s Hanford Site near Richland, Washington.

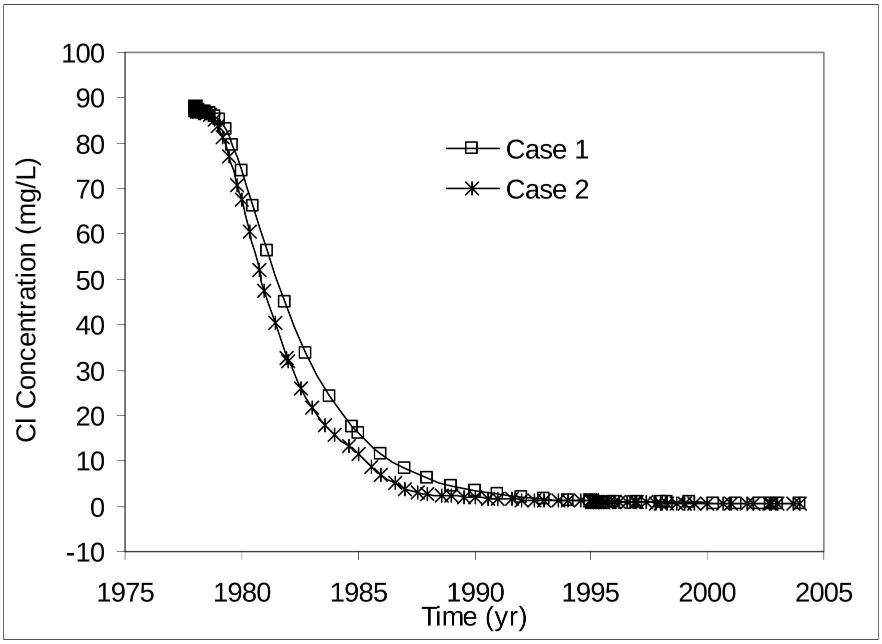

Figure. 2. Time course of the simulated chloride concentration at the drainage.

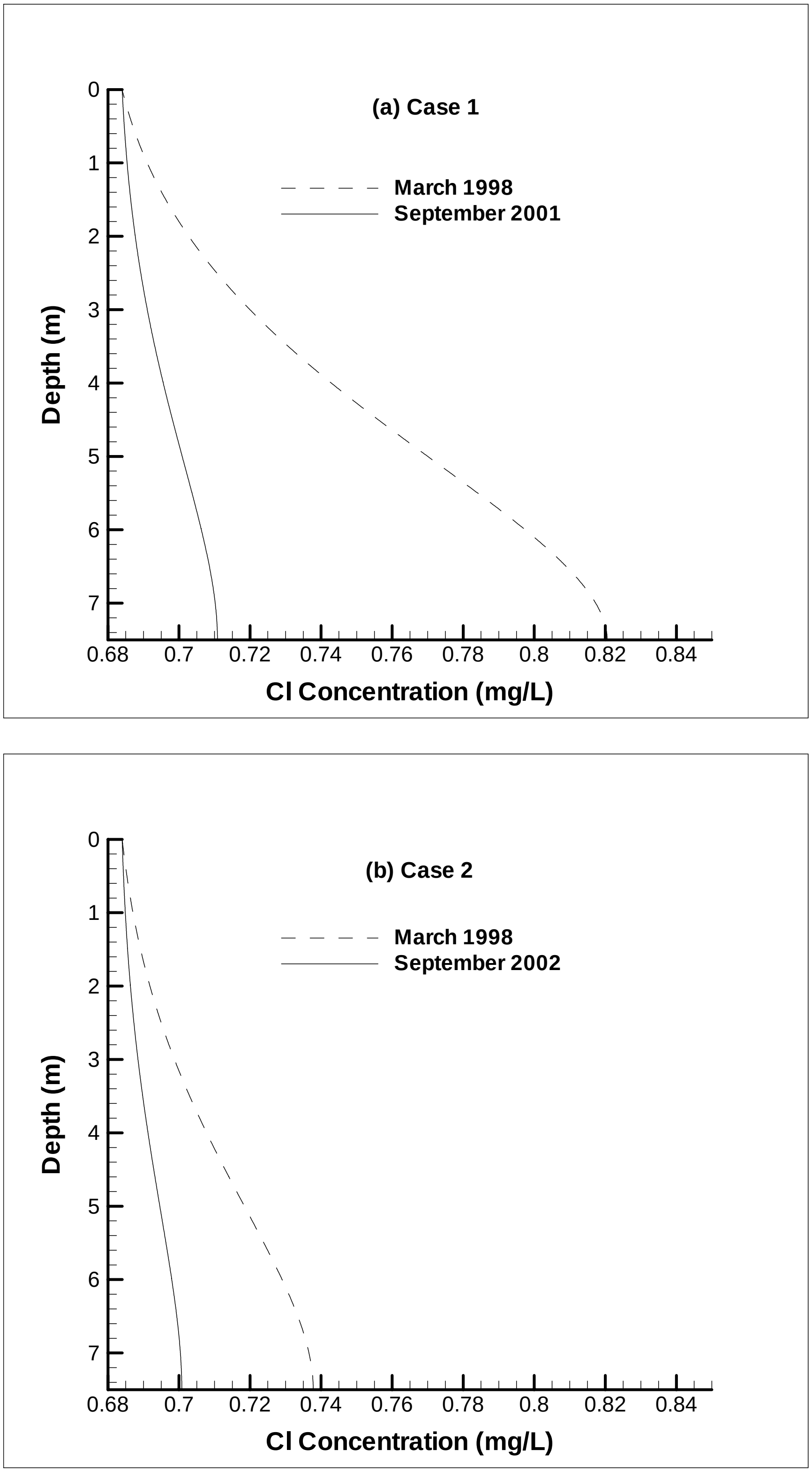

Figure 3. Simulated chloride concentration profile in March 1998 and September 2002.