WATER MIGRATION IN COVERED WASTE ROCK, INVESTIGATIONS USING DEUTERIUM AS A TRACER.[1]

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Abstract. A deuterium tagged rainfall event with a δD of +213 was applied to the top surface of an experimental covered waste rock pile in May of 2003. The tracer experiment was designed to resolve the spatial variability of infiltrating water and to estimate the magnitude and rate of flow within covered waste rock. The five meter high pile was deconstructed one year later and waste rock was sampled along vertical profiles at 10cm increments. Pore waters were extracted from the waste rock using the centrifugal method. The measured δD values range from -90 to background levels of approximately -130 with a few zones of locally high δD values deeper than three meters in the pile. Variability between individual vertical δD profiles are observed within the 8x8 meter area. The combination of the spatial variability and the deep δD values yield strong evidence for intermediate scale (~15 cm) preferential flow.

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Introduction

Soil covers are a common strategy used to reduce infiltration into waste rock stockpiles, and thereby reduce long-term acidic drainage and metal loading to the receiving environment. Soil covers can be designed to: (1) slow down internal weathering reactions, and (2) limit the flushing of stored weathering products from a stockpile. Detailed discussions on the design and effectiveness of soil covers are available (U.S. EPA, 1994, Albright et al., 2002, Milczarek et al., 2003, O'Kane and Waters, 2003, MEND 2.21.4, 2004); however, information about the systematics of the water flow and of the transport of weathering products through covered waste rock is limited. Such information may lead to better ARD predictive methods and refined source control strategies.

Scope

In this paper we describe the use of a stable isotope tracer to characterize the flow dynamics within an experimental waste rock pile. The tracer was applied following the placement of a compacted waste rock cover. The compacted cover reduced infiltration from approximately 50 % of rainfall to less than 6 % of rainfall. This reduction in infiltration is consistent with the estimates of Timms and Bennett, (2000) for infiltration into uncovered and covered waste rock at the Rum Jungle mine. The initial expectation was to see a transition from a pre-cover flow regime with a high degree of spatial variability and distinctive preferential flow characteristics to one dominated by more uniform flow in the finer-grained matrix materials. Data are presented here to examine this hypothesis.

Preferential flow in unsaturated media

Preferential flow paths in waste rock are often described as a system of large, interconnected voids or pore spaces (Beven and Germann, 1982, Morin et al., 1991, Flury et al., 1994, Newman, 1999, Li, 2000, Bellehumeur, 2001). The link between large-scale preferential flow paths and permeability variations or soil heterogeneity has been documented numerically (Trapp et al., 1995, Birkholzer and Tsang, 1997, Eriksson and Destouni, 1997, Fala et al., 2003) and observed experimentally (Eriksson et al., 1997, Fines et al., 2003, Nichol, 2003).

Small-scale heterogeneities have also been shown to significantly affect flow and solute transport in unsaturated matrix material (Buczko, et al., 2001, Hangen et al., 2004). Parlange and Hill (1976) and Baker and Hillel, (1990) document small scale preferential flow resulting from water flowing across a transition from fine to coarse texture materials.

Preferential flow is also observed in materials that are devoid of obvious structural or physical heterogeneity (Parlange and Hill, 1976, Jury et al., 2003, Ghodrati and Jury, 1992, Wang and Jury, 2003, Cho et al., 2005). Preferential flow that is not directly related to the material properties or texture can occur at the redistribution wetting front as the hydraulic gradient reverses following an infiltration event into a drier medium (Diment and Watson, 1985, Wang and Jury, 2003). Wang et al. (2004) and Jury et al. (2003) suggest that all soils are likely to experience such preferential flow, or flow fingers. Wang et al. (2003) observed that flow fingers could be as wide as 13cm and that the flow paths reappear in the same location during subsequent infiltration events. Waste rock beneath a soil cover may also exhibit this type of preferential flow. Waste rock often has a high air entry value and will remain close to residual water content except at shallow depths during and shortly following an infiltration event. The majority of water that infiltrates through the cover will be constrained within the finer matrix material and experience a gradient reversal due to evaporation above as the matric potential switches. At the equivalence point of the gravitational and evaporative matric potentials, lateral redistribution will occur. If water reaches coarse areas that act as capillary breaks before hydrostatic equilibrium occurs, then preferential flow will be initiated.

Chemical tracers in waste rock

Tracers have been used to investigate flow and transport processes in uncovered mine waste rock and soils. Murr (1979) and Nichol (2003) used chloride, Eriksson et al., (1997) used bromide, Bellehumeur (2001) used dye staining, and Hangen et al., (2004) used Br, terbuthylazine, and deuterium. Other than Guebert and Gardner, (2001) who used a fluorescent dye, few tracer studies have been conducted on covered mine material.

Nichol (2003) described flow and transport of a chloride tracer through the same experimental waste rock pile that is discussed in this paper. The waste rock pile was 8x8m in plan and 5m high. The test pile was built upon a grid of 16 contiguous lysimeters, each 2x2m in area. No cover was placed on the waste rock. Outflow at the base of the pile was recorded on a continuous basis. Nichol (2003) concluded that multiple flow paths exist in as small as a 2x2m area and that some of these flow paths are not dominated by capillary forces.

The experimental waste rock pile was covered with a lower permeability cover in September 2002. A deuterium tracer was applied in May 2003 during an artificial rainfall event. The pile was deconstructed in May 2004. Deuterium was chosen as a tracer based on a low analysis cost, a low detection limit and the lack of chemical interference with other solutes in the pore water. A bromide tracer was also applied to the covered waste rock pile in 2003; however we were unable to accurately measure bromide in pore water. High SO_4^{-2} concentrations (400-40,000 ppm) in pore water masked bromide in ion chromatography analyses and sample sizes were too small for colorimetric analysis. This study describes the observations of deuterium tracer distributions in pore water obtained from within the waste rock pile during deconstruction. At the time the pile was deconstructed, no tracer had yet been observed in the basal drainage system.

Experimental Methods

Field design and sampling

The deuterium tracer experiment was conducted on a constructed waste rock pile located at the Cluff Lake Mine in northern Saskatchewan. Several earlier experiments included evaluation of water and solute transport through the 5m high uncovered waste rock pile. Details of the pile construction and earlier experiments can be found in Nichol et al., (2000), Nichol (2003), Marcoline et al., (2003), Nichol et al. (2003), and Wagner et al., (2006 this issue).

Lower permeability surface layer

A compacted waste rock layer was constructed on the top surface of the waste rock pile in September 2002. Waste rock sieved to minus 10cm was compacted slightly dry of optimum water content to an average field density of 2114±47 kg/m^3. Density was measured in six locations with a Troxler nuclear density gauge with a 15cm measurement depth. Small excavations through the compacted layer were logged to visually inspect the degree and depth of compaction. Compaction appeared homogeneous to a depth of approximately 12cm with a slightly lesser degree of compaction between 12cm and 15cm.

In the spring of 2004, the physical properties of the compacted surface layer were re-evaluated. Based on visual inspection, compaction appeared homogenous to a depth of approximately 20cm. Sixteen samples were collected for measurement of volumetric water content and soil density from the top 25cm of the waste rock pile (Fig. 1). The average density was 1984±140 kg/m^3 and the volumetric water content was 7±0.8 %. It is unclear if the lower soil density measured in 2004 was related to averaging over a 25cm depth as opposed to 15cm in

2002; if it is related to measurement technique, or to temporal changes in the material characteristics.

Figure 1. Photo of the top covered surface of the waste rock pile. The large grid (orange) lines approximate the 2×2m boundaries of underlying lysimeters. The smaller (16 red and 8 blue) squares mark the locations of sample pits and density test pits respectively.

Experimental procedures

A 49mm artificial rainfall event tagged with a deuterium tracer was applied to the surface of the waste rock pile over a 6.5 hour period on May 13, 2003. Approximately 12 % of the rainfall was recorded as runoff, resulting in an estimated infiltration of 43mm. The rainfall event had a deuterium value (δD) of +213 per mil and a $\delta^{18}O$ value of -15.47 per mil. In June of 2004 the waste rock pile was deconstructed in four lifts (Fig. 2) each 1 to 1.5 m in height. Waste rock samples were collected at random locations along vertical trench faces excavated across the waste rock pile. All samples were collected below the lower-permeability cover. Each sample was collected in 10 cm increments by hand excavating an area approximately 15x15cm wide by 10cm high from the trench faces. Waste rock samples were sealed with nitrogen and transported to the University of British Columbia where pore waters were extracted using the centrifugal method. Stable isotopes $\delta^{18}O$ and δD were determined from pore waters at the New Mexico Tech Stable Isotope Lab with a Thermo Finnigan Delta Plus XP isotope ratio mass spectrometer. The hydrogen and oxygen isotopes are reported in delta (δ) notation, as per mil (‰) differences relative to the Vienna-Standard Mean Ocean Water (V-SMOW) international. Based on duplicate results, the analytical precision was (+/-) 0.6‰ for δD and as much as 0.3‰ for the very small (0.05 to 0.15 ml) $\delta^{18}O$ samples based on lab standards.

Figure 2. Photo of a vertical trench face at the base of the experimental waste rock pile. Vertical (red) lines approximate boundaries of underlying lysimeters. Note the heterogeneity in waste rock texture.

Discussion

Isotopic background

A water sample can be enriched in deuterium (heavier) by either mixing with the deuterium tracer or by evaporative fractionation. Evaporative fractionation is the enrichment of water in which lighter isotopes are disproportionally removed leaving heavier residual water. We can distinguish between the evaporative and mixing effects in our pore water samples using O_2 isotopes of the water molecule, since the initial O_2 isotopic composition of the applied tracer was similar to that of the local water. Any variations in the O_2 isotopic composition are due to evaporation alone. Any deuterium in a sample beyond that which can be explained by evaporation must therefore come from the applied tracer. The meteoric water lines, developed next, are used to establish the degree of evaporative fractionation.

The Global meteoric water line (GMWL) is defined by $\delta^{18}O$ and δD values of global precipitation and is approximated by $\delta D = 8 \times \delta^{18}O + 10$ (Craig, 1961) (Fig. 3). Regional or local meteoric water lines (LMWL) are often developed as part of isotopic studies. A LMWL may have a slightly different slope and intercept than the GMWL, as a result of factors such as the degree of evaporation, local climate, humidity, distance from the ocean, latitude, and altitude (Allison, 1982, Gat, 1996, Clark and Fritz, 1997).

Cluff Lake is located at 58º latitude in the center of the continent and in an area without significant topography. Data to create a true LMWL based on seasonal variations in precipitation are not available for the Cluff Lake area. To establish background isotope values for the study, the $\delta^{18}O$ and δD values of 10 pre-tracer, pore water samples were collected between 0 and 4.5 meters deep within the experimental pile and from an artificial precipitation event. Pore water samples were collected over a period of two years using standard vacuum soil water solution samplers. Our measured $\delta^{18}O$ pore water and artificial precipitation water values are within the range of values predicted for the geographic area by Bowen and Wilkinson, (2002). The $\delta^{18}O$ pore water values are plotted against their δD values (Fig. 3) and define an average pre-tracer isotope value with depth in the experimental waste rock pile. For the purposes of this study, this line will be referred to as the Cluff Lake pore water line (CPWL). The CPWL differs from a local meteoric water line in that the samples are not derived exclusively from precipitation over a period of time. It is important to note that any fractionation of pore water subsequent to precipitation would have resulted in an evaporation trend line with a shallower slope than the GMWL. While the entire CPWL is shifted to the right of the GMWL, the CPWL has a very similar slope suggesting that all background samples have undergone the same amount of evaporation relative to the GMWL with minimal fractionation. Based on the slope and the location of the CPWL relative to the GMWL, we believe that the CPWL adequately serves as proxy for a LMWL for our experiments. Our estimated background δD value is in the range from -125 to -131 parts per mil, depending on depth. We propose that any larger values observed after the tracer application are either a result of fractionation or contain water from the tracer application.

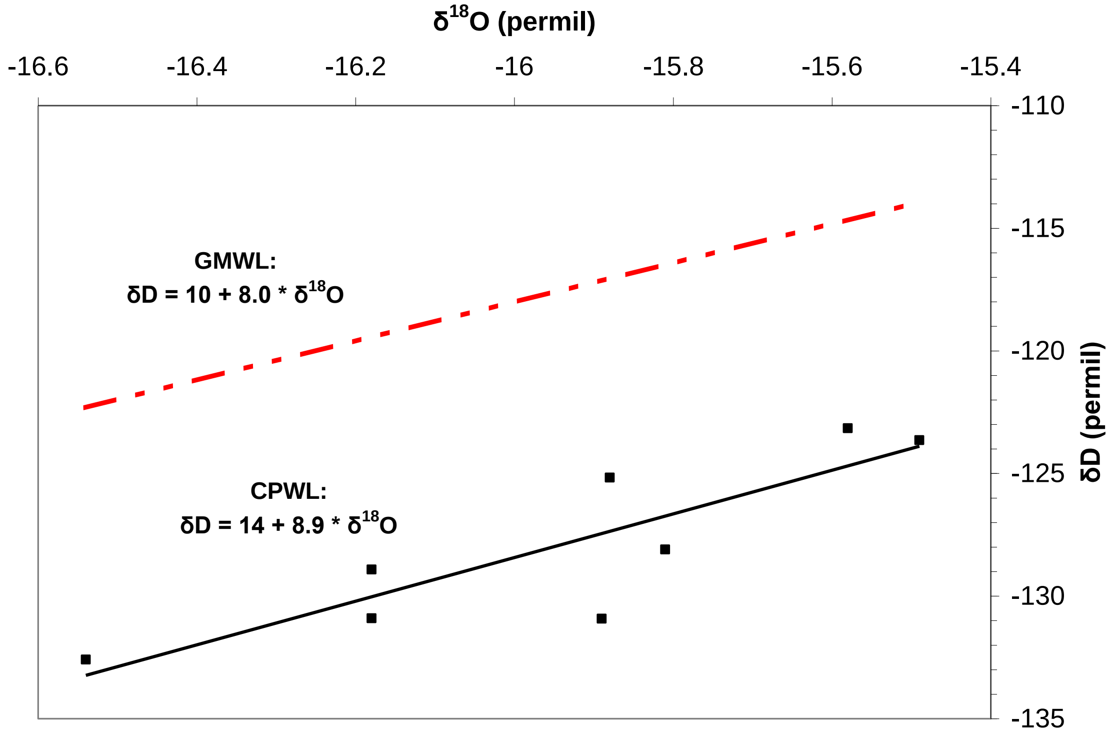

Figure 3. The solid line (CPWL) represents the average background pore water isotope values measured from within the experimental waste rock pile in comparison to the Global meteoric water line (GMWL) published by Craig (1961).

<u>Evaporative effects</u>

The δ^{18}O values were measured for 26 of our 150 post tracer pore water samples in order to assess the degree of fractionation resulting from evaporation. Figure 4 shows the 26 δ^{18}O and δD values relative to the CPWL. All samples are isotopically heavier, shifted to the right of the CPWL, suggesting that a degree of fractionation has occurred since the application of the tracer. The data are representative of waters that have undergone evaporation in air with low relative humidity (Gat, 1996). Due to the high degree of spatial variability of the waste rock and the flow, and the low number of samples, a single evaporative trend line cannot be estimated. An evaporative trend line consisting of six samples is observed within only one of the vertical profiles collected and exhibits a slope of 4.7. The slope of 4.7 is comparable with other slopes for evaporation of pore water in the unsaturated zone including 2 to 5 determined by Allison (1982) and 4.0 determined by Ortega-Guerrero et al. (1997).

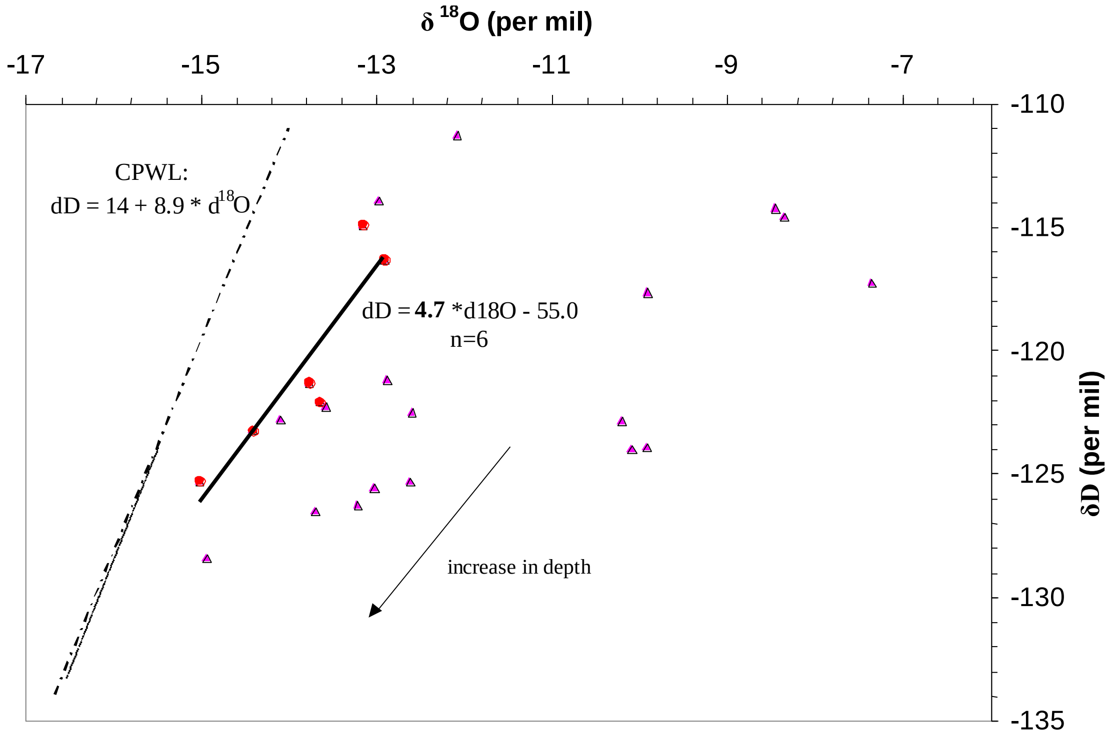

Figure 4. Plot of 18 $\delta^{18}O$ and δD pore water values of samples collected one year after tracer application. The dotted line is the CPWL. The solid line is an evaporative trend line defined by 6 samples collected along a vertical profile.

To further investigate the significance of evaporation on our δD values within the waste rock pile, $\delta^{18}O$ was plotted with depth (Fig. 5). In general, the samples have increasing $\delta^{18}O$ values with a decreasing sample depth suggesting that fractionation has occurred and the process is more pronounced near the surface of the test pile.

It is unclear why the large range in $\delta^{18}O$ values is observed between depths of 2 and 3.5 meters. Possible explanations are that the heavier samples were collected from areas with larger amounts of air circulation leading to higher evaporation and resulting in preferential fractionation, or that subsequent rainfall events displaced near surface, pre-fractionated water to this depth. The second scenario is unlikely since it appears that little water passed through the cover following the tracer application. The values for samples collected at the base of the pile plot adjacent to the background line and are unaffected by evaporation.

Deuterium distribution

During the deconstruction of the experimental pile, 300 waste rock samples at 10 cm intervals were collected along vertical profiles. Each sample was collected from an area of approximately 15 by 15 cm wide by 10 cm high from vertical trench faces. The intent was to map continuous tracer profiles with depth. The vertical profiles were located in zones where the matrix material exhibited vertical continuity.

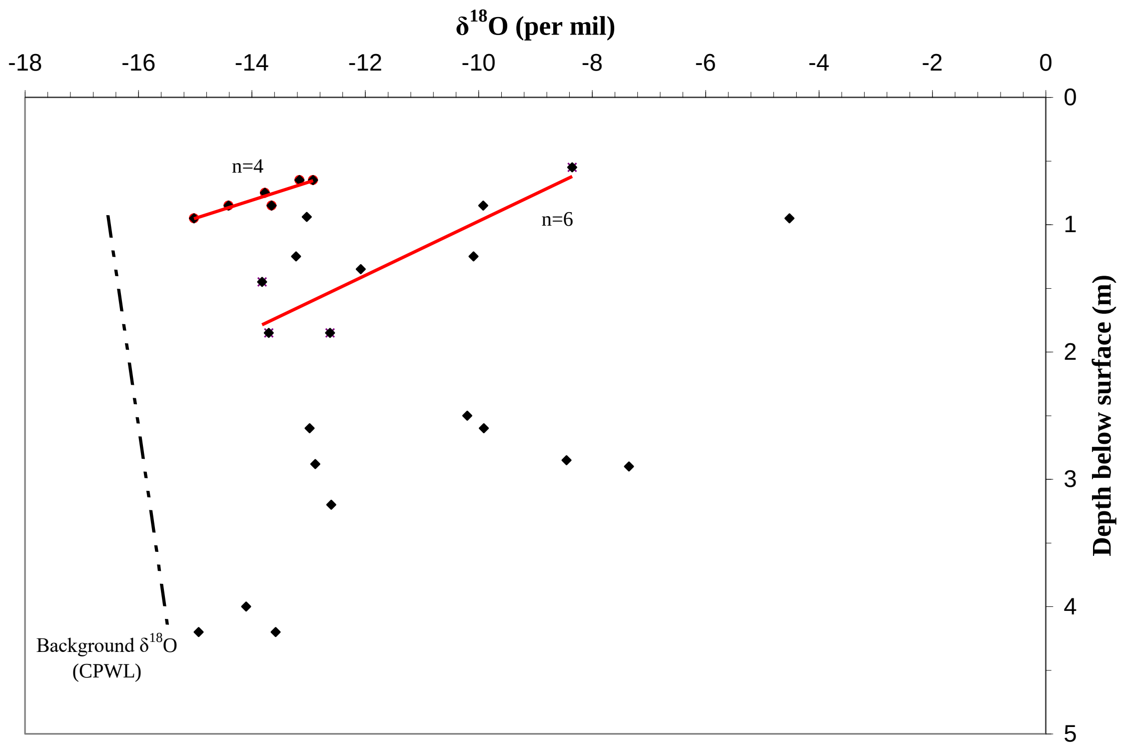

Figure 5. Plot of 26 $\delta^{18}O$ values relative to depth in the waste rock pile. The dotted line represents the pre-tracer $\delta^{18}O$ values from the CPWL plotted with depth. The two red lines represent potential evaporative trend lines from samples collected within two different sample profiles. The apparent trends of increased isotopic ratios are observed in the upper two meters of the profile.

Pore waters that are enriched in δD and not in $\delta^{18}O$ have clearly mixed with tracer water with little evaporative fractionation. The distinction between evaporation and mixing cannot be made in pore waters that are enriched in both $\delta^{18}O$ and δD. In the top one to two meters of the pile, it is difficult to distinguish between the two processes. As a result, both the $\delta^{18}O$ and δD pore water values must be examined individually for each sample.

Only four out of 150 δD values plot to the left of the CPWL (Fig. 6); further supporting the idea that the CPWL adequately defines an upper depleted boundary for post tracer isotopic values within the waste rock pile. Pockets of pore water exist, at all depths within the pile, which have not been affected by either mixing with tracer or by evaporation. This is evident from the data that plot immediately adjacent to the CPWL. At the same time, heavier δD values which were observed at all depths above four meters indicate that enrichment relative to the CPWL has occurred as a result of both tracer application and / or evaporation.

The δD values from 150 pore water samples range from as heavy as -63 per mil to the background level of -132 per mil. The vertical and horizontal spatial distribution of pore water δD values in the waste rock pile is highly variable. There is no clear trend in δD values with depth at the 8x8m horizontal scale (Fig. 6). It is clear that tracer has migrated to a depth of four meters in one year following the tracer application. There is an absence of heavier δD values below four meters, suggesting that the tracer did not migrate past this depth. Similarly, there are numerous shallow samples that plot adjacent to the CPWL suggesting that very limited amounts of mixing or evaporation have occurred. Based on the heterogeneous tracer distribution in the waste rock pile it is clear that even after cover placement, preferential flow paths have continued to transmit water within the test pile.

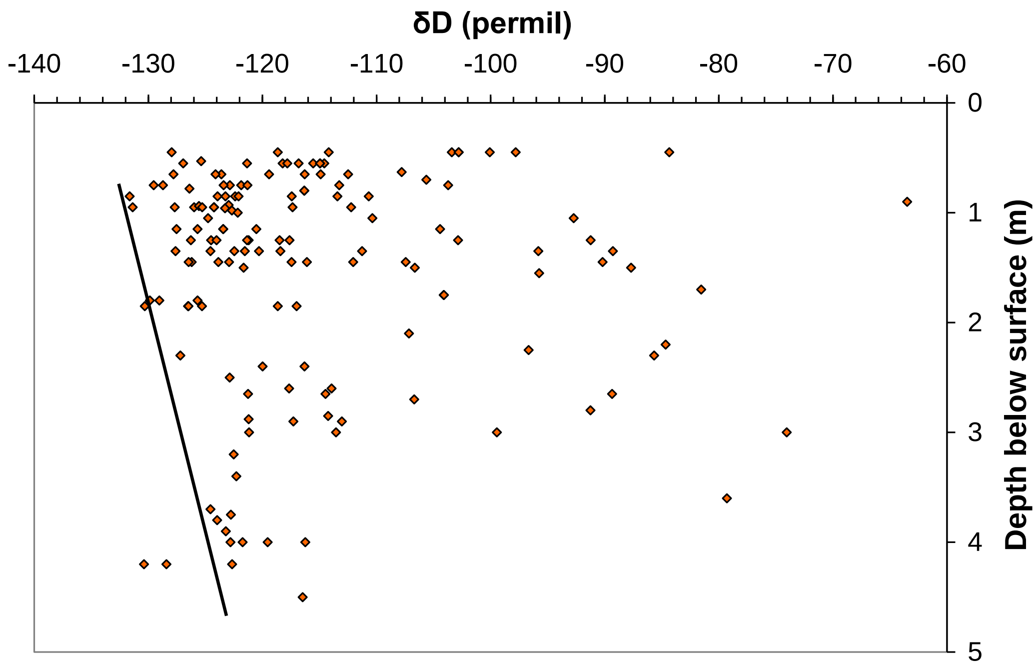

Figure 6. Scatter plot of δD values measured from post tracer pore water samples plotted against sample depth. The solid line marks the background δD values from pore water with depth. The background samples were collected prior to tracer application and are the same data used to determine the background CPWL shown in Fig. 3.

<u>Scale of preferential flow</u>

While clear trends in δD values with depth are not observable at the 8x8m horizontal scale, they appear to be evident within the 15x15cm wide scale defined by sample profiles. Vertical sampling profiles were located no further than 2m apart. Several of the sampling profiles were as close as 25cm. Contoured δD values of pore water extracted from these individual vertical profile samples are shown in Fig. 7. Caution is needed when interpreting tracer values along these vertical profiles since discontinuities and truncations in the data exist. The truncations and discontinuities result from either localized coarse zones, an inability to extract sufficient water from the samples for analysis or from misaligned profiles between lifts during deconstruction.

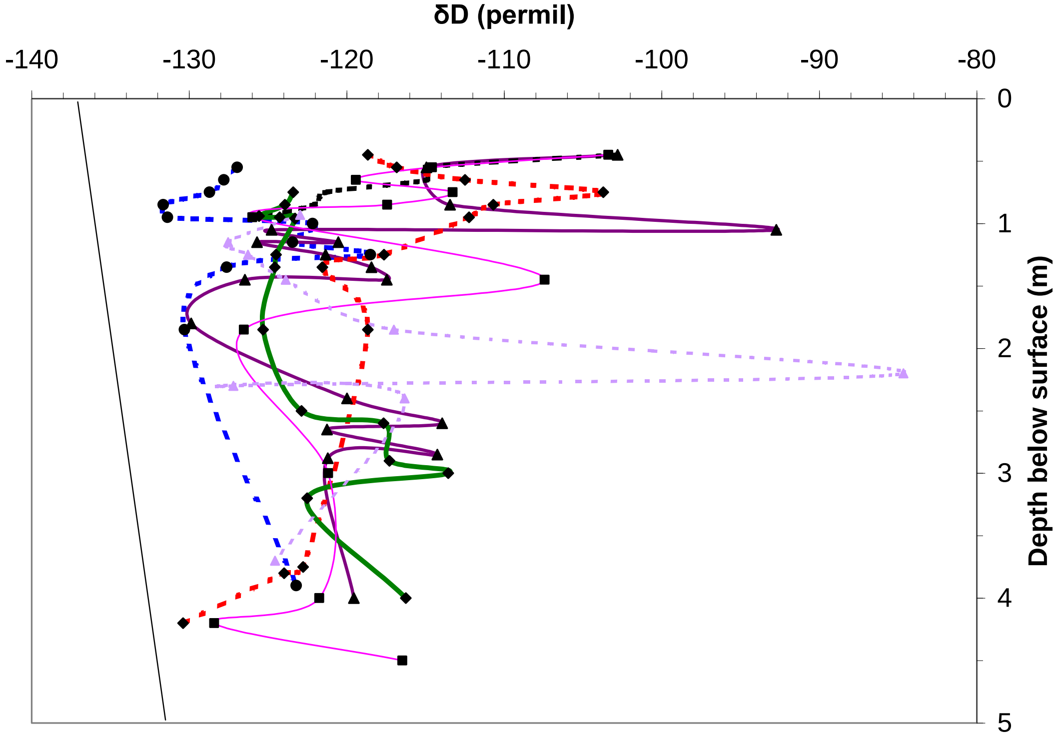

Figure 7. Patterns in δD values with depth in the waste rock pile. For clarity, only six profiles are shown. The straight line is the CPWL defined by average background δD values. Of particular interest is the variability of depth of the maximum and the slope of the curves above and below the maximum. Contoured δD values from three profiles within 40 cm are shown by the dotted lines.

The isotopic ratios of water samples collected in the top one meter of waste rock below the cover are highly variable and difficult to interpret. The pore water samples are a mix of pre-tracer water, tracer water and fresh, post tracer infiltration water. The samples in the top meter also show a range in the degree of evaporative fractionation from sample to sample. The near surface samples that are heavier with respect to δD and depleted in $\delta^{18}O$ are believed to consist primarily of old pre-tracer water. Samples that are heavier with respect to δD and have near background $\delta^{18}O$ values consist of non-fractionated tracer water. A few near surface samples have isotopic ratios characteristic of fresh rain water with no subsequent fractionation. The fresh water near the surface is progressively mixed with tracer water with depth along these few profiles resulting in a negative slope. Only a few profiles show a negative slope suggesting that the infiltration of fresh water between the tracer application and the deconstruction was highly localized.

The majority of samples from the top meter of the waste rock pile consist of a mix of post tracer and tracer water that has experienced evaporative fractionation as evident by the positive slopes in δD with depth. As a result of the complexity of the data in the top meter of waste rock, only general conclusions can be drawn based on these samples. One, it appears that some water infiltrated through the cover to a depth of up to one meter during rainfall events that followed the large tracer event. These pathways were highly localized. Two, it appears that the majority of the evaporative flux occurred to a maximum depth of 1m below the cover and that the evaporation within the top meter of the waste rock pile was spatially variable.

The pore water samples collected below a depth of one meter were not influenced by mixing with post tracer infiltration water and show a localized and lesser degree of evaporative fractionation. While the interpretations may be speculative, several patterns are evident (Fig. 7) that can be attributed to individual flow paths. Each profile exhibits a δD maximum at a different depth, most between 0.7 and 1.5 m. The maximum values identify the approximate depth of the wetting front following tracer application in that specific profile. While localized lateral flow is expected, no lateral correlations in δD values are observed in the data. In addition to maxima at 0.7 to 1m, contoured δD values from several different profiles have maximum values between 2 and 3m. The fact that the depth of the maximum δD value is different for all profiles suggests that several different scales of preferential flow were occurring within the covered waste rock. Since the depth of maximum δD values is different even in profiles located immediately adjacent to each other, we conclude that the dominant scale of preferential flow occurs at less than 15cm (the size of the sample taken for pore water extraction).

Conclusions

The tracer experiment documents the existence of event-driven preferential flow within a waste rock pile with a lower-permeability cover. For conditions prior to placement of the lower permeability cover, Nichol (2003) estimated an average pore water velocity through the matrix materials of approximately 1.5 m/yr. Nichol (2003) and Marcoline et al., (2003) documented maximum preferential flow velocities as high as 5 m/d. Following placement of the cover we observed little to no flow within the waste rock between rainfall events. Maximum tracer concentrations were observed between 0.7 and 1.5 meters below the cover one year after tracer application. Since the tracer maxima are attributed to preferential flow during rainfall events, 0.7 to 1.5 m/y serves as an upper estimate for the average pore water velocity. We have also observed tracer to a maximum depth of four meters in the waste rock which is attributed primarily to preferential flow velocities as high as 2-4 m/d. Interpretations of pore water tracer values are complicated in the top 1 to 1.5 meters of the pile as a result of fractionation and mixing. At all depths between 1.5 and 4m, fractionation is minimal and the enriched water can only be from the applied tracer. The high variability in tracer values result from different amounts of mixing with pre-tracer water. The combination of the lateral spatial variability of δD values and the δD values deep within the pile yield strong evidence for preferential flow. Based on the tracer values obtained from adjacent vertical profiles, distinctive preferential flow paths appear to be contained within an area less than the 15cm diameter profiles. The observation that preferential flow occurs in covered and drained waste rock has significant implications for prediction of water residence times, wetted surface area, and characterizing how metals are transported through reclaimed waste rock.

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