Long-Term Risk from Actinides in the Environment:
Modes of Mobility

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Shawki Ibrahim, Ph.D.
Department of Radiological Health Sciences
Colorado State University
Fort Collins, Colorado 80523

PUBLICATIONS:

- Abstract for Health Physics Society's 45th Annual Meeting:
Whicker, R.D.; Ibrahim, S.A.; Whicker, F.W. 2000. Vertical Migration of plutonium in soils from WIPP and Rocky Flats. Health Physics, Vol. 78, no.6, supplement 3. Pg. 158. ISSN 0017-9078.
- Research paper:
Vertical Migration of plutonium in soils from the Waste Isolation Pilot Plant and the Rocky Flats Environmental Technology Site (Whicker, R.D.; Ibrahim, S.A.; Whicker, F.W.). In preparation for submission. Expected completion date: Dec. 31, 2000.

PRESENTATIONS

- Vertical Migration of plutonium in soils from WIPP and Rocky Flats (Whicker, R.D.; Ibrahim, S.A.; Whicker, F.W.). Presentation given by R.D. Whicker at the American Radiation Safety Conference and Exposition (Health Physics Society's 45th Annual Meeting), Adams Mark Hotel, Denver, CO, June 29, 2000.

TECHNOLOGY TRANSFER

- Following concepts previously used by German researchers (Bachhuber, H., et al., 1981), a non-destructive method was developed for measuring the vertical migration of soil particles labeled with ¹³⁴Cs tracer in soil column experiments. The methodology involved monitoring ¹³⁴Cs transport from outside the column walls (external monitoring) with a BGO detector encased in a specially constructed lead collimator coupled to a multi-channel analyzer. The collimator was positioned perpendicular to the walls of the columns (in-situ). Supporting the collimator was an adjustable lab jack that allowed external monitoring at various soil depths.

Unlike the design used by the German researchers, our collimator "window" was a narrow (3 mm) horizontal slit. This helped reduced the angular scatter of photons impinging on the detector surface and constrained incoming signals to a more perpendicular geometry. The result was improved resolution of ¹³⁴Cs depth profiles (to within 2 or 3 mm) and greater ability to detect subtle changes in these profiles.

Future studies of this kind could benefit not only from the design of the detection system, but from prior knowledge of the methodology-related problems encountered during this study (e.g. adequacy of ¹³⁴Cs concentration levels, edge effect problems, etc.).

VERTICAL MIGRATION OF PLUTONIUM IN SOILS FROM THE WASTE
ISOLATION PILOT PLANT AND THE ROCKY FLATS ENVIRONMENTAL
TECHNOLOGY SITE.

PREPUBLICATION
COPY

R.D. Whicker, S.A. Ibrahim and F.W. Whicker

Department of Radiological Health Sciences
Colorado State University, Fort Collins, CO 80523.

Abstract – laboratory soil column experiments were conducted to investigate the migration of plutonium (using ^{134}Cs tracer as a surrogate for Pu) in soils from the Waste Isolation Pilot Plant (WIPP) and the Rocky Flats Environmental Technology Site (RFETS). For RFETS columns subjected to cycles of irrigation and drying (treatment columns), the mean estimated percent loss of ^{134}Cs inventory ($\pm$ one standard error) from the labeled-soil layer at the soil surface was 3.1 ± 0.6 % cycle $^{-1}$. In RFETS columns subjected to cycles of irrigation only (control columns), the mean estimated loss of ^{134}Cs inventory was 0.7 ± 0.6 % cycle $^{-1}$. WIPP treatment and control columns had estimated losses of 1.3 ± 1.2 % cycle $^{-1}$ and 0.5 ± 0.2 % cycle $^{-1}$ respectively. Respective cesium migration depth estimates for RFETS treatment and control columns were 0.27 ± 0.04 cm cycle $^{-1}$ and 0.10 ± 0.01 cm cycle $^{-1}$, while for WIPP, they were 0.08 ± 0.02 cm cycle $^{-1}$ and 0.04 ± 0.003 cm cycle $^{-1}$. Significant relationships existed between estimates of losses and migration depths despite differing methods of estimation ($r^2 = 0.76$ and 0.86 for RFETS and WIPP columns respectively). Differences in ^{134}Cs migration between treatment and control columns were statistically significant ($\alpha = 0.05$) for RFETS but not for WIPP. Between-site comparisons indicated significant differences in migration depth estimates, but not in ^{134}Cs loss estimates. Bypass migration of bulk soil particles through soil cracks was apparently responsible for about 60 to 70 % of total migration in RFETS soils, while colloidal migration through interstitial pore spaces accounted for the remainder. Significant differences in migration depth estimates between control columns ($p = 0.002$) from the two sites indicated that colloidal migration may have been greater in RFETS soils despite having much smaller pore spaces. Within-column, between-column and cycle-to-cycle variability was high, particularly for RFETS treatment columns.

INTRODUCTION

Sound policies regarding cleanup activities and environmental management practices at Department of Energy (DOE) facilities in arid or semi-arid environments requires dynamic models for the terrestrial transport of soils contaminated with actinides. The mobility of plutonium-contaminated soils is a particularly high priority issue. Environmental factors can vary widely between these facilities, and thus the adequacy of any Pu transport model depends greatly on site-specific parameter assessments. Of the basic mechanisms for transporting Pu in the environment, wind and water erosion acting on the soil surface are by far the most important. Vertical migration of Pu into soil profiles is normally a relatively minor mechanism. On time scales of a few decades or less, generally only the top few centimeters of soil profiles are subject to wind and water erosion. Thus, Pu residing deeper in the soil profile is not immediately subject to erosional processes. The purpose of this study was to quantify the vertical migration of plutonium (using ^{134}Cs tracer as a surrogate for Pu) in soils from the Waste Isolation Pilot Plant (WIPP) and the Rocky Flats Environmental Technology Site (RFETS). This information is crucial for developing credible, site-specific loss rate expressions for Pu transport models.

An assumption fundamental to the design of this study was that infiltrating precipitation would be the primary driving force for the vertical migration of Pu. Other potential driving forces such as bioturbation or freeze-thaw action were not considered due to practical constraints on the scope and duration of the study. The effectiveness of precipitation in driving vertical migration may be governed by factors such as soil type, vegetation, and topography, all of which ^a vary widely between sites, and can vary considerably within each site. Within the climatic regimes at either WIPP or RFETS, it is relatively common for drought-like conditions to be punctuated by occasional large precipitation events. Such extreme climatic sequences can result in large amounts of erosion at the soil surface, but how they affect the vertical migration of soil particles or colloids is not well known. This is an important issue for conservative actinide transport modeling at these sites. The scope of this study was limited to examining vertical migration as a function of climatically extreme sequences and soil type, both between and

within sites. Our hypothesis was that any vertical migration would result from the transport of bulk soil particles through macropores (cracks) in the soil profile (bypass migration), or from the transport of colloidal materials through the soil matrix itself (colloidal migration).

METHODS

Conceptual framework

There is substantial evidence that under normal environmental conditions, both Cs and Pu bind strongly with soil particles and that Cs can be used to estimate Pu concentrations in soil (White, et al., 1981; Lee, et al., 1996;). The characteristic adsorption of both Cs and Pu as a function of particle size is very similar (White, et al., 1981). Because migration for both Cs and Pu is governed by the transport of soil particles through physical processes (Higley, 1994; Hakonson and Bostick, 1975;), both radionuclides are good tracers of sediment particle movement (Hakonson, et al., 1985).

The experimental design for this study entailed measuring the vertical migration of ^{134}Cs labeled soils applied to the surface of uncontaminated soil cores when subjected to simulated cycles of climatic extremes. The intention was to make inferences about Pu migration at each site based on indications of ^{134}Cs migration in soil column experiments. Soil cores from both sites were extracted in the field as sample pairs, with each pair representing a given soil type. From each sample pair, one core was subjected to alternating cycles of irrigation (simulated precipitation) and drying (treatment column) while the other was subjected to cycles of irrigation only (control column).

Treatment cycles were intended to simulate the effects of extensive drought followed by heavy precipitation on vertical migration. Drying can promote the formation of networks of cracks, particularly in clay soils (Mermut, 1992; Tariq and Durnford, 1993). The more extensive the drying, the more invasive these macropores are likely to be. In clay soils, cracks can reach depths of more than 0.5 meters (Bouma and Dekker, 1978). When

precipitation intensity is higher than the (bulk) soil infiltration capacity, soil cracks provide conduits for preferential drainage (bypass flow) of runoff water. If turbulent velocities of runoff water are relatively high in such circumstances, bulk soil particles (particles of many characteristic sizes) can be entrained and thus flow into soil cracks.

Control cycles were intended to simulate only the effects of extreme amounts of precipitation on vertical migration. Control columns were covered with plastic sheets between irrigations to prevent drying and soil cracking. In the absence of cracks, the transport of soil colloids through interstitial soil pore spaces by infiltrating water is presumably the only other possible physical mechanism for the vertical transport of ^{134}Cs . Conceptually, for each site and given soil type, treatment columns would provide information about the degree of total vertical migration that might result from climatically extreme sequences. Comparisons of treatment and control columns, intern, would help identify the relative effectiveness of bypass migration versus colloidal migration in vertically transporting ^{134}Cs at each site.

Soil core sampling

Representative soil core samples (~ 30-cm length $\times$ 20-cm diameter) from areas near the WIPP and RFETS facilities were collected in PVC tubes (June, 1998 and May, 1999 respectively). At each site (WIPP and RFETS), two replicate soil cores (sample pairs) were taken from four different sample locations representing a variety of soil types. Replicate cores were obtained by simultaneously driving side-by-side PVC tubes into the pre-saturated ground. The tubes were beveled on one end to make driving easier and to help reduce disturbance to the soil profiles. Core extraction involved removing soil on one side of the cores in order to free them without disturbing the soil profile left behind on the other side of the cores.

After core removal, layered samples were collected from the exposed soil profile left behind (in 3-cm increments). Laboratory analyses of these layered samples helped characterize each profile (soil texture, chemistry, etc.). Additional samples from the soil

surface (top 3 cm) at each sample location were obtained for ^{134}Cs labeling in the lab. After the cores were extracted, they were capped on one end for transport. The cap, designed for use in the laboratory soil column experiments, involved a special screen inserted into a perforated end cap – the design was intended to optimize a balance between unrestricted water flow and maximum soil particle retention.

Soil column preparation

Soil labeling. Surface soils obtained near each respective site were irreversibly labeled with ^{134}Cs in several steps. First, DI water was added to a known volume of soil to form a soil / water slurry. Next, a HCl acid solution containing a known concentration of ^{134}Cs was rapidly and thoroughly mixed in to the slurry. The slurry was kept wet and mixed often for several days before being dried in an oven. After drying, water was again added to reconstitute the soil / water slurry, which was again periodically mixed over several days before re-drying. This process was repeated several times. In a background solution of water, clay minerals will readily adsorb ^{134}Cs ions between silicate layers while excluding most other hydrated cations by size (EPA, 1999). A large amount of cesium can be almost irreversibly adsorbed by a soil containing only a small amount (e.g. 1-2 % by weight) of such clay minerals (Douglas, 1989). Repeated shrinking and swelling of the soil was intended to ensure complete and essentially permanent adsorption of ^{134}Cs ions on exchange sites within the crystal lattices of clay minerals. These soils were considered irreversibly labeled when water from small aliquots of the slurry mixture (obtained in a centrifuge) contained no ^{134}Cs activity. Finally, dry ^{134}Cs -labeled soils were conditioned for use by breaking up aggregates with a mortar and ~~pedestal~~^{pestle} until only constituent soil particles remained. The activity density of ^{134}Cs in labeled soils was about 496 Bq g^{-1} for RFETS soils and 364 Bq g^{-1} for WIPP soils.

Application of labeled soils. In some cases, soil cores surfaces were not quite perpendicular to the column walls and / or had significantly irregular topography. When subjected to simulated rainfall, labeled soils would tend to erode from high spots and accumulate in low spots. This would make the spatial distribution of ^{134}Cs at the surface

uneven and could potentially distort the results. To help prevent this, the topography of original soil core surfaces were artificially leveled where necessary before applying ^{134}Cs labeled soils. Low spots were filled with excess surface soil collected at each core's respective sampling location. Fill soils were compacted by irrigation until core surfaces were level. A consistent amount (weight in grams) of ^{134}Cs labeled soil was then evenly distributed on the surface of each soil core (to a depth of about 0.6 cm).

Experimental treatment cycles

Irrigation cycles involved irrigating each column with a documented volume of water (in ml) until approximately 400 ml of leachate was recovered in collection reservoirs supporting the columns. Intended to simulate rainfall, irrigation was conducted by pouring water into a perforated cup that was held about 10 cm above the soil surface. All leachate was monitored for ^{134}Cs activity in a HPGe well detector. Leachate was recycled as new irrigation water provided that any ^{134}Cs activity present in the leachate was negligible in relation to the total ^{134}Cs inventory within each column. This was done for two reasons: 1) to alleviate problems with liquid waste disposal and 2) to help prevent potential flow problems related to the leaching of salts from the soil system (Higley, 1994). Drying cycles involved drying columns in a large oven at about 55 degrees Celsius until thoroughly dry, as determined by the initial dry weight of the columns.

In order to help insure valid ^{134}Cs migration results, it was necessary to address "edge effects" along the walls of the soil columns. When subjected to wetting / drying cycles, soils can shrink and pull away from the column walls, leaving large, artificial migration pathways around the edges. In order to minimize ^{134}Cs migration through such pathways, a latex foam sealant was injected into any cracks formed between the soil and the column walls after each drying cycle. Because it was unlikely that ^{134}Cs could migrate the entire length of the column through the soil matrix itself, the presence of ^{134}Cs activity in leachate was a possible indicator of edge effect migration.

Measurement of ^{134}Cs profiles

General approach. After the application of the labeled-soil layer, estimates of vertical migration as a function of treatment or control cycles were generated by analysis of changes in ^{134}Cs depth distributions (i.e. net decay-corrected activity per unit of soil profile depth). Two methods were used to measure these changes: passive external monitoring of ^{134}Cs depth profiles (external profiles) after each treatment or control cycle, and destructive internal sampling of ^{134}Cs depth profiles (internal profiles) at the end of the experiment. For external profiles, ^{134}Cs measurements are expressed as the number of counts per 100 seconds in the energy region between 505 and 908 KeV. For internal profiles, where a different detector and counting geometry was used, measurements are expressed as the number of counts per minute in the energy region between 520 and 890 KeV.

External monitoring. Following concepts of a detection system previously used by German researchers to investigate ^{137}Cs migration in soil columns (Bachhuber, et al., 1982), external monitoring involved a Bicron BGO detector encased in a specially constructed lead collimator. To limit measurements to regions of interest for ^{137}Cs , the detector was coupled to a Davidson multi-channel analyzer. The detector / collimator apparatus was positioned outside (and perpendicular to) the walls of the columns (in-situ). Unlike the collimator used in the German study, our collimator “window” had a narrower (0.3 cm) vertical slit width. This helped reduce the vertically oriented angular scatter of photons impinging on the detector surface, and constrained incoming signals to a more perpendicular geometry. The intention was to maximize the resolution of ^{137}Cs depth profiles so that subtle changes in the distributions could be detected.

Supporting the collimator was an adjustable lab jack that allowed external monitoring at various soil depths. Measurements were taken at depth intervals of 1 mm for the top centimeter of the profile, 2 mm for the second centimeter, and depth intervals of 5 mm or more below that. At any given soil depth, measurements were taken on four sides of the

column and averaged. Thus, each external ^{134}Cs depth profile used in the data analysis represented an average of four profiles taken over the length of a given column. Other than initial measurements on dry columns, all subsequent measurements were made on wet columns after all gravitational drainage of irrigation water had ceased and soil water contents were at “field capacity” (Veihmeyer and Hendrickson, 1931). Slight differences in soil water content could result in different degrees of photon attenuation, and soil water content was more likely to be consistent from one measurement to the next at field capacity.

Internal sampling. Internal sampling was accomplished by taking 6 random sub-cores within each column. Because such sampling is destructive, direct measurements of initial internal profiles for individual columns were not possible. Initial internal profiles were instead derived empirically by internally sampling separate, but identically prepared representative columns (one for RFETS and one for WIPP) that were not subjected to any experimental cycles. Sub-coring involved a 1.1-cm diameter brass tube pushed into the soil surface to a depth of 10 cm or until rocks prevented further depth of penetration. The coring tube had depth gage markings etched on the outside to document depth of penetration. Soil columns were moist during sampling to insure soil cohesion during core extraction and extrusion.

During coring, sub-cores were usually compacted, especially in RFETS soils. The degree of compaction for each sub-core was measured with a separate depth gage designed to fit inside the coring tube and rest freely / independently on the surface of the soil during coring. This allowed documentation of the actual length of the soil core relative to the penetration depth of the coring tube (information later used to adjust internal ^{134}Cs profile depths to account for compaction). Sub-core sectioning and preparation for counting involved extruding soil cores with a special plunger in small increments, slicing with a thin blade into sections, placing into pre-weighed / labeled scintillation vials, and obtaining wet and dry (dried in oven) sample weights. Sections were sliced in increments of 1 mm for the top centimeter of the profile, 2 mm for the second centimeter, and 5 mm

below that. Samples were counted for 15 minutes each with a Packard Minaxi γ Auto-Gamma 5000 Series gamma counter.

Data analysis

General approach. As mentioned previously, estimates of vertical migration as a function of treatment or control cycles were generated by analysis of changes in both “internal” and “external” ^{134}Cs depth distributions. External profiles provided quantitative evidence of distributional changes from one cycle to the next. Internal profiles provided direct evidence of the *overall* (beginning to end of experiment) depth of ^{134}Cs migration, and by comparing final and empirically derived initial profiles, provided indirect evidence of overall distributional changes. Overall distributional changes ^{in turn} ~~intern~~, provided estimates of overall losses of ^{134}Cs inventory from the initial labeled-soil layer. Estimates of average losses and depths of migration on a per cycle basis were obtained simply by dividing overall estimates by the number of cycles run.

To evaluate trends and variability for losses and migration depths across individual cycles, a simple model was developed to estimate losses and migration depths for individual cycles (based on external profile data). This model involved relating (through linear regression) overall distributional changes in internal profiles to corresponding changes in external profiles, and to estimates of overall losses and migration depths. These relationships, along with external profile changes from one cycle to the next, were then used to predict losses and migration depths for individual cycles.

External profiles. Allowing for small irregularities that may have resulted from measurement or counting error, there were some basic characteristics typical of most external profiles. One was a “peak” ^{134}Cs reading centered approximately in the middle of the labeled-soil layer (figure 1). This was probably because despite the narrow (3 mm) slit width of the collimator, some degree of angular impingement of photons on the detector (photons originating at angles slightly greater or less than perpendicular to the detector) could still occur. Such angular impingement would presumably reach a

Figure 1. Example ^{134}Cs depth profile with soil column superimposed (note: in this example profile, only counts in the energy region between 505 and 700 KeV are expressed).

maximum at the center of the labeled-soil layer, resulting in a peak reading. Below the ^{134}Cs -labeled soil layer, profiles exhibited exponential decline culminating asymptotically at levels near background.

Preliminary evaluations of these profiles revealed that quantitative indications of migration would be very subtle and primarily limited to the top two centimeters of ^{134}Cs distributions (figure 1). In-depth analyses of distributional changes therefore focused only on this region of ^{134}Cs profiles. Indications of such changes involved two aspects of ^{134}Cs distributions – the depth of the peak and the depth of center of mass. Of these two aspects, the depth of the center of mass appeared to be a stronger indicator of migration. Signs of ^{134}Cs migration always involved changes in the depth of the center of mass (i.e. depletion of ^{134}Cs inventory from the labeled-soil layer), but did not necessarily involve changes in the depth of the peak. A single indicator, however, sensitive to both aspects, was used to represent distributional changes for all analysis or

modeling purposes. This indicator was the slope of a linear regression of all data within the top two centimeters of external profiles (external slope). Conceptually, this slope would become smaller (less steep) over subsequent cycles if significant downward migration of ^{134}Cs were to occur.

Internal profiles. Prior to ^{data} analysis, these profiles were adjusted to account for compaction during sub-coring using a factor obtained by dividing the actual length of the sub-core by the penetration depth of the coring tube. The actual adjustment entailed dividing the lower limit of each internal profile depth increment by this fractional value. Although compaction was seldom uniform in sub-cores after extraction (i.e. compaction tended to increase with depth), core extrusion usually produced additional compaction in the upper layers resulting in more uniformity. This provided reasonable justification for the simplified approach of using a constant factor to adjust an entire profile for compaction.

Like external profiles, below the interface between the labeled-soil layer and underlying soil horizons, internal profiles declined exponentially with depth. Estimating the depth of ^{134}Cs migration involved plotting counts for this receding limb of the distribution on a logarithmic scale and fitting an exponential curve to the data. The depth at which this exponential curve dropped below a certain baseline (100 counts per minute – about twice background) was considered a reasonable estimate of the gross depth of migration for ^{134}Cs . The ^{rational} ~~rational~~ for using this baseline as the minimum evidentiary level for ^{134}Cs migration was that during sub-coring, a small degree of profile “smearing” (artificial entrainment of ^{134}Cs -contaminated particles by coring tube walls) appeared to occur. Under such circumstances, using a baseline value closer to background levels could easily result in erroneous overestimates of the depth of migration. The “net” depth of migration (depth of migration into the originally uncontaminated soil profile) was obtained by subtracting the thickness of the labeled soil layer (approximately 6 mm) from the gross migration depth estimate.

As discussed earlier, overall losses of ^{134}Cs inventory from the initial labeled-soil layer were estimated indirectly by comparing distributional changes between final and empirically derived initial profiles. Preliminary evaluations of final and initial profiles, however, revealed discrepancies with respect to their mass-balances for ^{134}Cs . This was not surprising since initial profile characteristics were not measured directly but were instead inferred from empirical evidence. Typically, final profiles had a lower total ^{134}Cs inventory than respective initial profiles. This, along with possible inaccuracies in the shape of initial ^{134}Cs distributions, was a potential source of error with respect to estimates of losses. In order to generate the best possible estimates of overall losses of ^{134}Cs from the labeled-soil layer, it was necessary to address potential inaccuracies in these empirically derived initial profiles where possible.

To account for mass balance discrepancies in ^{134}Cs inventories, empirical initial profiles were normalized to actual final profiles without changing their basic shape relative to profile depth. This involved multiplying initial profiles by a factor that would result in mass-balance agreement between initial and final inventories. Because there was no scientifically rational method to address potential inaccuracy with respect to initial profile shape, the shape of normalized empirical profiles was necessarily assumed to provide a reasonable hypothetical approximation.

Once initial profiles were individually normalized to each column, overall losses of ^{134}Cs from the labeled-soil layer were estimated by subtracting the ^{134}Cs inventory found in the labeled-soil region of the final distribution from that found in the initial distribution. These ^{134}Cs inventories were calculated by summing the mean activities of each (1-mm) depth segment in this region. There were two columns (one from RFETS and one from WIPP) in which final inventories were greater than initial inventories, suggesting a net gain of ^{134}Cs in the labeled-soil layer during the course of the experiment. Perhaps error in the hypothetical shape of respective initial distributions was responsible for this result. Because possible errors in initial profile shapes could easily have inflated loss estimates from other columns, however, there was no real justification for excluding these findings from the statistical analyses.

Finally, like external profiles, the slope of the top 2 cm of internal profiles (internal slopes) provided a measure of two distributional characteristics (depth of peak and center-of-mass) affected by ^{134}Cs transport. Since internal sampling provided estimates only of initial and final internal profiles, however, only overall changes in internal slopes could be estimated. With the exception of evaluations involving ^{134}Cs migration depths, changes in internal slopes (as with external slopes) were used to represent distributional changes for all other analysis and modeling purposes.

Statistical analysis. Linear regressions were used to establish trends or relationships for a variety of analytical or modeling purposes. Differences in mean estimates of various parameters between treatment and control columns, both within and between sites, entailed one-tailed paired t-tests. A standard level of significance ($\alpha = 0.05$) was used to evaluate the statistical significance of both t-test and linear regression results. The coefficient of variation (CV), calculated as the standard deviation divided by the mean, provided a measure of within-column, between-column, and cycle-to-cycle variability. Estimation uncertainty was evaluated by the standard error of the mean.

RESULTS AND DISCUSSION

Internal Profiles

Cesium losses. Analysis of internal profiles for RFETS columns revealed evidence that losses of original ^{134}Cs inventory from the labeled-soil layer were greater for treatment columns than for control columns. Mean estimated percent losses per cycle ($\pm$ one standard error) for RFETS treatment and control columns were $3.1 \pm 0.6 \text{ \% cycle}^{-1}$ and $0.7 \pm 0.6 \text{ \% cycle}^{-1}$ respectively (fig. 2). T-tests indicated a significant difference in mean losses per cycle between treatment and control columns ($p = 0.02$). It is clearly evident that much of this apparent difference is due to bypass migration in treatment columns.

Figure 2. Mean estimated loss of ^{134}Cs inventory per cycle for treatment and control columns.

Because of the high clay content in RFETS soils (clay contents averaged $48 \pm 1.3\%$), the formation of networks of cracks in columns subjected to drying cycles was pervasive. These cracks often reached widths at the surface up to 5 mm. Contrary to previous findings (Bouma and Dekker, 1978; Horn et al., 1994), cracks appeared to occur at random locations from one cycle to the next, rather than repeatedly forming along the same planes (fig. 3). During the first few seconds of irrigation, soil particles at the surface were rapidly mobilized and flushed into soil cracks that provided large preferential flow paths for infiltrating water. After a few seconds, however, cracks would clog with sediments and infiltration rates would slow towards some steady state. This non-linear “pulse” of particle transport in response to irrigation was a clearly visible process in RFETS treatment columns. Presumably, any subsequent vertical ^{134}Cs transport after soil cracks were sealed was limited to colloidal migration, and the rate of such migration would be comparable to that of control columns. In a previous study, bypass flow was found to be a more effective mechanism than interstitial flow for transporting actinide contaminated soil particles through homogeneous soils (Higely, 1994).

Figure 3. Example of soil cracking patterns for successive cycles in RFETS treatment columns.

For WIPP columns, mean estimated percent losses per cycle for treatment and control columns (fig. 2) were 1.3 ± 1.2 and 0.5 ± 0.2 % cycle⁻¹ respectively. This difference in mean losses per cycle between treatment and control columns was not significant ($p = 0.50$). Because of the small clay content in WIPP soils (clay contents averaged 6.0 ± 0.4 %), treatment columns seldom formed cracks. When cracks did form, they were extremely small and did not visually appear to extend to any significant depth. A reasonable interpretation of these results and observations is that colloidal migration was the main process governing ¹³⁴Cs transport in both treatment and control columns, and despite differing experimental cycles, its relative effectiveness in each case was similar. The numerical difference may have simply resulted from differences in volumes of irrigation water applied. On average, treatment columns required about 64 % more irrigation water per cycle in order to completely saturate the column and obtain sufficient leachate for analysis from the bottom.

Although the mean estimated loss of ¹³⁴Cs per cycle was almost three times greater for RFETS treatment columns (3.1 ± 0.6 % cycle⁻¹) than for WIPP treatment columns (1.3 ± 1.2 % cycle⁻¹), this difference was not statistically significant ($p = 0.27$). If colloidal

migration were similarly effective at each site, the additional mechanism of bypass migration occurring at RFETS would presumably result in significant differences between sites. However, colloidal migration may not be equally effective at each site. Sandy soils have much larger pore spaces than clay soils and thus have much greater infiltration capacities (Dunne and Leopold, 1978). Generally, soil colloids are considered to be particles less than 10 micrometers in diameter and can sometimes move faster than average ground-water flow velocities due to effects such as size exclusion from smaller pore spaces (Puls, et al., 1991). Theoretically, colloidal migration through interstitial pore spaces should thus be appreciably more effective in sandy soils. Estimated mean losses per cycle in control columns, however, were actually slightly higher for RFETS soils relative to WIPP soils (despite averaging 31 % less irrigation water applied per cycle). Although this relative numerical difference for control columns was contrary to expectations regarding “colloidal” migration, the statistical difference was not significant ($p = 0.77$).

In an attempt to isolate only losses resulting from bypass migration, mean estimated losses per cycle in control columns were subtracted from those in paired treatment columns. When the resulting ‘bypass’ loss estimates were compared, no significant differences between sites were found ($p = 0.34$). Given that during irrigation, bypass migration was only apparent in RFETS treatment columns, estimation error could easily be responsible for this result. For all of the above comparisons involving estimates of ^{134}Cs losses, there is some potential for uncertainty in the results because initial profiles were derived empirically and thus have some inherent potential for error. Nevertheless, there was some basis for having a reasonable general level of confidence in ^{134}Cs loss estimates. Estimates of ^{134}Cs migration depths, generated directly from measured final profiles and thus having considerably less potential for error, showed a significant relationship with ^{134}Cs loss estimates for both sites (figure 4).

Figure 4. Relationship between mean estimates of total depth of ^{134}Cs migration and total percent loss of ^{134}Cs inventory from the soil surface.

Migration depths. For RFETS columns, final internal profiles showed direct evidence that ^{134}Cs had migrated into originally uncontaminated soil to greater depth for treatment columns than for control columns. Mean estimated migration depths per cycle for RFETS treatment and control columns (fig. 5) were 0.27 ± 0.04 and 0.10 ± 0.01 cm cycle⁻¹ respectively. The difference in these estimates was highly significant ($p = 0.001$). This strongly support the earlier finding of significant differences in mean ^{134}Cs losses, as well as the interpretation that bypass migration is primarily responsible for differences in ^{134}Cs transport between treatment and control columns. If so, differences in migration depth estimates between 'paired' treatment and control columns suggests that about 60 to 70 % of total vertical transport at RFETS would be due to bypass migration processes.

For WIPP columns, mean estimated migration depths per cycle for treatment and control columns (fig. 5) were 0.08 ± 0.02 and 0.04 ± 0.003 cm cycle⁻¹ respectively. The difference between these estimated migration depths, as with ^{134}Cs losses, was not significant ($p = 0.09$). This further strengthens the idea that colloidal migration was not only governing ^{134}Cs transport in both treatment and control columns from WIPP, but

Figure 5. Mean estimated depths of ^{134}Cs migration per cycle for treatment and control columns.

that it was similarly effective in each case. Again, the numerical difference probably stems from differences in volumes of irrigation water applied.

Results from between-site comparisons of mean estimated migration depths consistently differed from those involving ^{134}Cs losses. For example, the difference in mean estimated ^{134}Cs migration depths between treatment columns for RFETS ($0.27 \pm 0.04 \text{ cm cycle}^{-1}$) and WIPP ($0.08 \pm 0.02 \text{ cm cycle}^{-1}$) was significant ($p = 0.01$). When respective uncertainties between estimated ^{134}Cs losses and migration depths are considered, this suggests that considerable differences in ^{134}Cs migration between sites are probable. Furthermore, when estimated 'bypass' migration depths (treatment minus control migration depths) between sites were compared, significant differences were also found ($p = 0.03$) indicating that bypass migration alone could account for differences in ^{134}Cs migration between sites.

Finally, mean estimated migration depths in RFETS control columns (0.10 ± 0.01 cm cycle⁻¹) were significantly greater ($p = 0.002$) than those in WIPP control columns (0.04 ± 0.003 cm cycle⁻¹). This result strengthens the possible (and unexpected) indication of greater ¹³⁴Cs losses due to colloidal transport in RFETS soils mentioned earlier. The reason why such (reverse) differences in “colloidal” migration between sites might exist is unclear. As mentioned, colloidal migration should be more effective at WIPP because sandy soils have larger interstitial pore spaces. One possible explanation is that ¹³⁴Cs transport in RFETS control columns may not have been restricted only to colloidal migration between interstitial pore spaces. Although soil cracking at the surface of control columns was not visually apparent, small internal macropores could still have developed providing conduits for “limited” bypass migration (i.e. bypass migration limited to very small or even colloidal-sized particles). Several characteristics of RFETS soils could conceivably result in the development of such small internal conduits.

First, soils at RFETS (unlike WIPP soils) have a high degree of soil structure. In clay soils, soil aggregates – bound by both organic and inorganic agents – often form as columnar structures and can sometimes be coated with a cemented clay layer (Horn et al., 1994). If soil moisture dropped slightly below field capacity (some minor evaporation probably did occur between cycles), small internal cracks could have developed between aggregates as clay minerals began to shrink slightly. Secondly, RFETS soils generally contained many large rocks (up to 1 cm diameter or more). Like the interface between soil cores and column walls, the interface between large rocks and bulk soil could be a potential location for the development of internal macropores with just a slight reduction in soil moisture. Finally, mean organic matter content in the top 9 cm of RFETS soils (3.0 ± 0.3 %) was greater than for WIPP soils (0.6 ± 0.1 %). Vegetation is more pervasive at RFETS and small channels left by decayed vegetative roots could provide conduits for limited bypass migration, even without any evaporative reduction in soil moisture.

Variability. Because of the uncertainty associated with ¹³⁴Cs loss estimates, only estimated ¹³⁴Cs migration depths were used to evaluate variability for ¹³⁴Cs transport.

TREATMENT					CONTROL			
	Column number	Mean estimated migration depth per cycle (cm cycle ⁻¹)	Within columns		Column number	Mean estimated migration depth per cycle (cm cycle ⁻¹)	Within columns	
			CV	n			CV	n
RFETS	RF-1	0.32	0.56	6	RF-2	0.12	0.27	6
	RF-3	0.20	0.32	6	RF-4	0.08	0.38	6
	RF-5	0.23	0.35	6	RF-6	0.09	0.39	6
	RF-7	0.36	0.59	6	RF-8	0.11	0.34	6
	Between columns	CV	0.28		CV	0.18		
		n	4		n	4		
WIPP	WP-3	0.14	0.39	6	WP-4	0.04	0.50	6
	WP-5	0.08	0.57	6	WP-6	0.03	0.77	6
	WP-7	0.04	0.39	6	WP-8	0.03	0.53	6
	WP-9	0.07	0.43	6	WP-10	0.04	0.47	6
	Between columns	CV	0.53		CV	0.14		
		n	4		n	4		

Table 1. Coefficient of variation (CV) values for mean estimated migration depths per cycle, both within columns (vertical) and between columns (horizontal).

Coefficient of variation values for estimated migration depths within individual columns ranged from 0.32 to 0.59 for RFETS treatment columns, and from 0.27 to 0.39 for RFETS control columns (table 1). While within-column variability, on average, was somewhat greater in treatment columns (perhaps due to variability in soil cracking), there was no statistical difference with control columns ($p = 0.32$). For WIPP treatment and control columns, CV values ranged from 0.39 to 0.57 and from 0.50 to 0.77 respectively. Within-column variability was significantly greater for control relative to treatment columns ($p = 0.03$), which was unexpected since differences in soil cracking between treatment and control columns were not evident. Estimated migration depths for control columns, however, were comparatively small and since the CV measures relative variability, a small degree of variation can sometimes appear comparatively large relative to the mean.

Regarding variability between columns (i.e. within-site variability), CV values among RFETS treatment columns, and among RFETS control columns, were 0.28 and 0.18 respectively (table 1). Coefficient of variation values among WIPP treatment columns,

and among WIPP control columns were 0.53 and 0.14 respectively. While many different factors are possible contributors to between-column variation (differences in soil cracking, soil type, rock content, core extraction, measurement error, etc.), the data garnered within the scope of this study was insufficient to adequately partition such variability.

External Profiles

Distributional (slope) changes. A typical example of how the top two centimeters of ^{134}Cs profiles for paired treatment and control columns changed over subsequent cycles and how such changes were reflected by changes in respective (external) slopes is shown in figure 6. Also shown are the corresponding relationships between external slope and cycle number. RFETS treatment columns consistently showed decreasing trends in external slopes over subsequent cycles that were, in general, statistically significant (r^2 values averaged 0.79). The magnitude of these trends, or more precisely, the magnitude of the slope of their respective linear relationships (termed “trend slopes” to avoid confusion with “external slopes”), averaged $-239 \text{ counts} \cdot (100 \text{ g})^{-1} \cdot \text{cm}^{-1} \cdot \text{cycle}^{-1}$. For RFETS control columns, trends in external slopes were always non-significant (r^2 values averaged 0.22) and trend slopes were extremely small relative to RFETS treatment columns (averaging $-1 \text{ count} \cdot (100 \text{ g})^{-1} \cdot \text{cm}^{-1} \cdot \text{cycle}^{-1}$). While treatment columns from WIPP displayed consistent decreasing trends in external slopes that were generally significant (r^2 values averaged 0.68), the average trend slope was less than half that of RFETS treatment columns ($-100 \text{ counts} \cdot (100 \text{ g})^{-1} \cdot \text{cm}^{-1} \cdot \text{cycle}^{-1}$). Finally, trends in external slopes for WIPP control columns, though typically significant, displayed very shallow trend slopes (averaging only $-9 \text{ counts} \cdot (100 \text{ g})^{-1} \cdot \text{cm}^{-1} \cdot \text{cycle}^{-1}$).

For all columns, there was a significant relationship between trend slope and estimated migration depth ($r^2 = 0.71$). When evaluated for migration depth categories, however, this relationship was highly significant for migration depths greater than 1 cm ($r^2 = 0.91$) but not for migration depths less than 1 cm ($r^2 = 0.02$). This result clearly suggests that a certain minimum degree of ^{134}Cs transport was required before external monitoring could

Figure 6. Top: profile changes over successive cycles and respective changes in external slope for paired treatment and control columns. Bottom: corresponding trends in external slope versus cycle number.

definitively resolve associated changes in ^{134}Cs distributions. Relative to the potential attenuation of photons by soil minerals and water, and to the overall sensitivity of the collimated detection system, the activity density of ^{134}Cs in labeled-soil particles may have thus been somewhat inadequate for tracing small changes.

Modeling losses / migration depths for individual cycles. The amount of uncertainty in model predictions for ^{134}Cs losses and migration depths may be considerably greater for WIPP columns than for RFETS columns. This is because the relationship between overall changes in internal and external slopes for RFETS columns ($r^2 = 0.61$) was

significant, while the relationship for WIPP columns ($r^2 = 0.14$) was not. Nevertheless, both of these relationships were used to predict internal changes in slope for individual cycles based on respective external slope changes. The other relationships necessary for modeling losses and migration depths for individual cycles were all highly significant for both RFETS and WIPP columns. Linear regressions of overall internal slope change versus overall ^{134}Cs losses had r^2 values of 0.93 and 0.95 for RFETS and WIPP columns respectively. Overall internal slope changes versus overall migration depths for RFETS and WIPP columns had respective r^2 values of 0.79 and 0.86.

Compared to mean losses and migration depths (on a per cycle basis) estimated from internal sampling, model results for individual cycles, on average, over-predicted losses and migration depths by about 28 %. Nevertheless, model over-predictions should not affect variability from one cycle to the next. Cycle-to-cycle variability was similar for both losses and migration depths. Model predictions for migration depths alone (and respective CV values) are presented in table 2.

Coefficient of variation values indicated that the variation from one cycle to the next for both losses and migration depths was generally very high. Although variability was somewhat greater on average for control columns versus treatment columns, again, the CV is a measure of variation relative the mean, and control columns had considerably smaller means. When comparing numerical differences in losses and migration depths for individual cycles, it was clear that the absolute magnitude of cycle-to-cycle variation was much greater in treatment columns. Like within-column variability, the reason for this in RFETS columns seems clear (because of soil cracking), but for WIPP columns it remains uncertain.

		TREATMENT				CONTROL			
		RFETS				RFETS			
column #:		1	3	5	7	2	4	6	8
cycle number		predicted ¹³⁴ Cs migration depth (cm)				predicted ¹³⁴ Cs migration depth (cm)			
1		0.4	0.6	0.5	0.2	0.1	0.2	-0.1	0.1
2		0.2	0.1	0.2	0.6	0.1	0.0	0.1	0.2
3		0.2	0.2	0.3	0.2	0.2	0.1	0.1	0.1
4		0.8	0.1	0.2	0.3	0.1	0.3	0.4	0.2
5		0.1	0.1	0.2	0.3	0.1	0.2	0.1	0.1
6		0.3	0.3	0.4	0.4	0.1	0.1	0.1	0.1
7		0.4	0.3	0.1	0.3	0.1	0.1	0.1	0.2
8						0.2	0.2	0.2	0.1
9						0.2	0.2	0.2	0.2
mean		0.36	0.26	0.29	0.34	0.15	0.15	0.12	0.14
CV		0.59	0.66	0.51	0.42	0.21	0.57	1.10	0.26

		WIPP				WIPP			
column #:		3	5	7	9	4	6	8	10
cycle number		predicted ¹³⁴ Cs migration depth (cm)				predicted ¹³⁴ Cs migration depth (cm)			
2		0.2	0.1	0.0	0.1				
3		0.1	0.1	0.1	0.2	0.1	0.0	0.0	0.0
4		0.2	0.2	0.2	0.1	0.0	0.0	0.0	0.0
5		0.1	0.1	0.1	0.1	0.0	0.1	0.0	0.0
6		0.1	0.0	0.1	0.1	0.1	0.0	0.0	0.0
7		0.1	0.2	0.1	0.1	0.0	0.0	0.0	0.0
8		0.1	0.1	0.1	0.1	0.0	0.0	0.0	0.0
9						0.0	0.0	0.0	0.0
10						0.0	0.1	0.0	0.0
11						0.0	0.0	0.1	0.0
12						0.0	0.0	0.1	0.0
13						0.0	0.0	0.1	0.1
14						0.0	0.0	0.1	0.1
15						0.1	0.1	0.1	0.1
16						0.1	0.1	0.1	0.1
mean		0.12	0.11	0.10	0.12	0.05	0.04	0.05	0.05
CV		0.45	0.59	0.49	0.43	0.67	0.74	0.76	0.59

Table 2. Predicted migration depths for individual cycles (based on external monitoring data) and associated CV values. Cycle-to-cycle variabilities for predicted ¹³⁴Cs losses were similar.

Migration rate estimates

Although this study provided only estimates of vertical ¹³⁴Cs migration based on cycles of alternating wetting and drying, this information could be used to develop credible estimates of *rates* of migration due to physical processes. In-depth investigations of weather data from each site (precipitation, temperature, humidity, etc.), along with field studies to determine the meteorological sequences necessary to produce “cycles” of soil cracking and precipitation analogous to our treatment cycles, would be a reasonable approach. While these endeavors were beyond the scope of this study, a cursory supplemental investigation, conducted only for the RFETS site, provided some idea of what migration rates might look like.

Fifty years of daily precipitation data from a location near RFETS (EarthInfo, 2000) were sorted by contiguous incremental categories of precipitation event magnitude (e.g. 2.5-5.0 cm, 5.0-7.5 cm, etc.) for frequency analysis. Though not realistic, a greatly simplified investigative approach was to assume a certain 'baseline' event magnitude would, in effect, represent a wet / dry cycle. The average number of "cycles" per year for all precipitation events greater than each baseline magnitude was calculated. These values were then multiplied by our estimated mean migration depth per cycle for RFETS columns to obtain 'hypothetical' migration rates (cm yr^{-1}) for each baseline event magnitude. Next, these hypothetical rates were compared with a migration rate inferred through evaluation of actual field data from earlier research conducted at Rocky Flats in order to determine which one (if any) might be reasonably credible. Assuming a migration rate was thus selected, the number of cycles used to generate that hypothetical rate would provide a plausible estimate of the number of cycles that might occur at RFETS in a given year.

Air sampling data from the late 1960's, indicated that the peak year of Pu contamination at RFETS was about 1969 (Little and Whicker, 1977). Plutonium distributions for soil samples taken roughly four years later (~ 1973), indicated that Pu had migrated as deep as 20 centimeters into soil profiles (Little and Whicker, 1977). If Pu took 4 years to migrate to this depth, and migration rates were relatively constant, then the annual rate of migration was about 5 centimeters per year. By matching this annual rate to a similar hypothetical rate calculated from the precipitation data, we determined that on average, wet / dry cycles comparable to our 'treatment' cycles, might be expected to occur about 18 times per year at RFETS. If so, the magnitude of a baseline event that might produce rates of migration similar to our treatment cycles was about 0.8 cm of precipitation.

If in-depth studies were to support the above number of cycles per year, then a Pu migration rate of about 5 cm yr^{-1} at RFETS would be a credible estimate of the combined affects of colloidal and bypass migration processes. At present, however, it is a highly uncertain estimate because of the inadequacy of the data and the superficial nature of the analysis. Daily precipitation data alone provides no information about rainfall intensity

or drying sequences between events. Furthermore, some studies have provided possible evidence that little migration has occurred since the early 1970's (Webb, et al., 1993; Webb, et al., 1997). Why plutonium may have migrated to depth within a few years and then remained relatively immobile since is uncertain. Perhaps this depth corresponds to the average annual maximum depth of infiltrating precipitation. Comprehensive research would be required to address these issues and provide concrete evidence of the average number of comparable cycles expected to occur annually at each site. In terms of total Pu losses from the soil surface, it would also be necessary to address biological processes affecting Pu migration at both sites. Once we have a better understanding of such issues, the findings of this study could easily be used to develop realistic, site specific loss rate constants for Pu transport models.

CONCLUSIONS

- There is substantial evidence in the literature that under normal conditions, both Cs and Pu behave similarly in the environment. Both radionuclides bind strongly with soil particles, and terrestrial migration for both is governed largely by soil particle transport. Therefore, using ^{134}Cs as a tracer of soil particle movement is a valid approach for investigating Pu transport.
- Clay contents in RFETS soils ($48 \pm 1.3 \%$) were much greater than in WIPP soils ($6 \pm 0.4 \%$). Consequently, RFETS treatment columns developed extensive networks of large (up to 5 mm wide) cracks. Soil cracks tended to occur at random rather than forming repeatedly along the same planes. During irrigation, bypass migration of bulk soil particles through soil cracks was visually apparent. Significant soil cracking was not evident in other columns.

- Mean estimated losses of ^{134}Cs inventory from the labeled-soil layer at the surface of soil columns were as follows:
 - RFETS treatment columns: $3.1 \pm 0.6 \text{ \% cycle}^{-1}$
 - RFETS control columns: $0.7 \pm 0.6 \text{ \% cycle}^{-1}$
 - WIPP treatment columns: $1.3 \pm 1.2 \text{ \% cycle}^{-1}$
 - WIPP control columns: $0.5 \pm 0.2 \text{ \% cycle}^{-1}$
- Mean estimated ^{134}Cs migration depths into originally uncontaminated underlying soil horizons were as follows:
 - RFETS treatment columns: $0.27 \pm 0.04 \text{ cm cycle}^{-1}$
 - RFETS control columns: $0.10 \pm 0.01 \text{ cm cycle}^{-1}$
 - WIPP treatment columns: $0.08 \pm 0.02 \text{ cm cycle}^{-1}$
 - WIPP control columns: $0.04 \pm 0.003 \text{ cm cycle}^{-1}$
- Cesium loss estimates had more potential for uncertainty because they were, in part, derived from empirical evidence while migration depth estimates were generated directly from measured ^{134}Cs profiles. Despite potential uncertainty, however, significant relationships existed between loss and migration depth estimates (r^2 values were 0.76 and 0.86 for RFETS and WIPP columns respectively).
- Differences in loss estimates between treatment and control columns were statistically significant for RFETS soils ($p = 0.02$), but not for WIPP soils ($p = 0.50$). Statistical comparisons of corresponding migration depth estimates ($p = 0.001$, and $p = 0.09$ respectively) supported these findings. Between-site comparisons had mixed results – differences were significant for migration depth estimates, but not for ^{134}Cs loss estimates.
- The evidence indicated that bypass migration of bulk soil particles through soil cracks was responsible for about 60 to 70 % of the total migration in RFETS soils, while colloidal migration through interstitial pore spaces apparently made up the rest.

- Ostensibly, colloidal migration was similarly effective for transporting ^{134}Cs in both treatment and control columns from WIPP. Contrary to expectations, however, a between-site comparison of control column migration depths indicated that colloidal migration was significantly greater ($p = 0.002$) in RFETS soils than in WIPP soils. While certain RFETS soil characteristics may provide plausible explanations, the reason(s) why this may have occurred remains unclear.
- In general, cycle-to-cycle variability relative to mean migration depths was high, and on average, was similar among all columns (mean CV values ranged from 0.49 to 0.69). Clearly, however, the absolute magnitude of variation in migration from one cycle to the next was much greater in RFETS treatment columns (presumably due to soil cracking).
- Both within-column variability (mean CV values ranged from 0.34 to 0.57) and between-column variability (CV values ranged from 0.14 to 0.53) for vertical migration were generally high.
- A cursory supplemental investigation produced an estimate of the average number of comparable cycles that might occur annually at RFETS ($18 \text{ cycles yr}^{-1}$), as well as an estimated migration rate for Pu in RFETS soils (5 cm yr^{-1}). While these estimates are highly uncertain, they serve to illustrate possible general magnitudes. Furthermore, this brief investigation served to highlight the nature of research necessary to produce well-substantiated estimates.

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