Geomorphic control of radionuclide diffusion in desert soils

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[1] Diffusion is a standard model for the vertical migration of radionuclides in soil profiles. Here we show that diffusivity values inferred from fallout ^{137}Cs profiles in soils on the Fortymile Wash alluvial fan, Nye County, Nevada, have a strong inverse correlation with the age of the geomorphic surface. This result suggests that radionuclide-bound particles are predominantly transported by infiltration rather than by bulk-mixing processes such as wetting/drying, freeze/thaw, and bioturbation. Our results provide a preliminary basis for using soil-geomorphic mapping, point-based calibration data, and the diffusion model to predict radionuclide transport in desert soils within a pedotransfer-function approach. **Citation:** Pelletier, J. D., C. D. Harrington, J. W. Whitney, M. Cline, S. B. DeLong, G. Keating, and K. T. Ebert (2005), Geomorphic control of radionuclide diffusion in desert soils, *Geophys. Res. Lett.*, *32*, L23401, doi:10.1029/2005GL024347.

1. Introduction

[2] Climatic, tectonic, and drainage-adjustment events often combine to form a distinct set of terraces that rise like a flight of stairs from the active channel on alluvial fans [*Bull*, 1991]. Soil-geomorphic mapping makes use of relative-age indicators such as landscape position (i.e. height above active channels), drainage patterns (tributary vs. distributary), and desert pavement and varnish development to group terrace surfaces into distinct age ranges. The pedological, hydrological, and biological properties of these surfaces are often quite distinct. As such, soil-geomorphic mapping provides a framework for characterizing many diverse landscape properties. Soil-geomorphic mapping has long been used by geomorphologists to delineate flood hazards [e.g., *Pearthree*, 1991] and map fault activity [e.g., *Gerson et al.*, 1993]. More recently, *Young et al.* [2004] documented correlations between hydrologic conductivity, water-storage potential, and surface age on alluvial-fan terraces in the Mojave Desert. These authors found that more well-developed A_v horizons on older surfaces inhibit infiltration and enhance water storage potential. This correlation provides the basis for using a pedotransfer-function approach for predicting soil hydrologic conductivity using point-based calibrations and a soil-geomorphic map.

[3] In this paper, we use soil-geomorphic mapping as a framework for predicting radionuclide diffusivities in soil profiles on the Fortymile Wash alluvial fan in Nye County, Nevada. This study was motivated by the need to predict radionuclide transport rates on the Fortymile Wash alluvial fan for the Yucca Mountain Project. The Fortymile Wash fan is of special interest because it is the depozone for most of the contaminated tephra transported by fluvial processes in the event of a volcanic eruption that intersects the repository. In addition, the Fortymile Wash fan is the location at which the radiological dose to a hypothetical individual is calculated for comparison to the regulatory standard. As such, quantifying the redistribution of radionuclides within soils of this fan is critical to calculating the expected dose, as well as to identify which dose pathways (e.g., inhalation, direct radiation from the ground, etc.) are most potentially hazardous in the event of an eruption.

2. Field Measurements

[4] The Fortymile Wash alluvial fan is located in northern Amargosa Valley, Nevada (Figure 1). The fan drains a 970 km^2 drainage basin and has a slope of approximately 1%. The alluvial-surface material is dominated by sandy, gravelly material. The eolian silt input to the soil is likely sourced by nearby playas, especially Franklin Lake playa, but the dominant eolian input is fine to medium-grained sand derived from active channels along Fortymile Wash and the Amargosa River. This sand locally forms coppice dunes and is visible as bright, NNW-directed streaks on aerial photographs. Silt-dominated vesicular A_v horizons on Fortymile Wash alluvial fan are less than 5 cm in thickness, with thinner and more discontinuous horizons on younger fan surfaces.

[5] Soil-geomorphic mapping of the Fortymile Wash fan was performed using the elevation above active channels, terrace dip, depth of dissection, degree of planarity, drainage pattern (tributary versus distributary), and the development of pavement, varnish, and calcic soils as relative-age indicators [*Bull*, 1991] (Figure 1). Holocene and Pleistocene units were readily distinguishable using pavement and varnish development, as is the case elsewhere in Amargosa Valley [*Whitney et al.*, 2004]. The maximum relief from the oldest unit (Qa3) to the active channels (Qa7) is 1–2 m, decreasing down-fan. Relief between units of adjacent age is typically 0.5 m, and often the intra-surface bar-and-swale relief approaches the relief between surface units. The degree of planarity was particularly useful for distinguishing between middle-to-late Pleistocene (Qa3) and late Pleistocene (Qa4) units on the Fortymile Wash fan. Drainage patterns were particularly useful for distinguishing between Qa4 (tributary drainage) and late Pleistocene-to-early Holocene (Qa5) surfaces (distributary drainage). Approximate ages have been assigned based on correlation with the regional chronology of *Whitney et al.* [2004], developed

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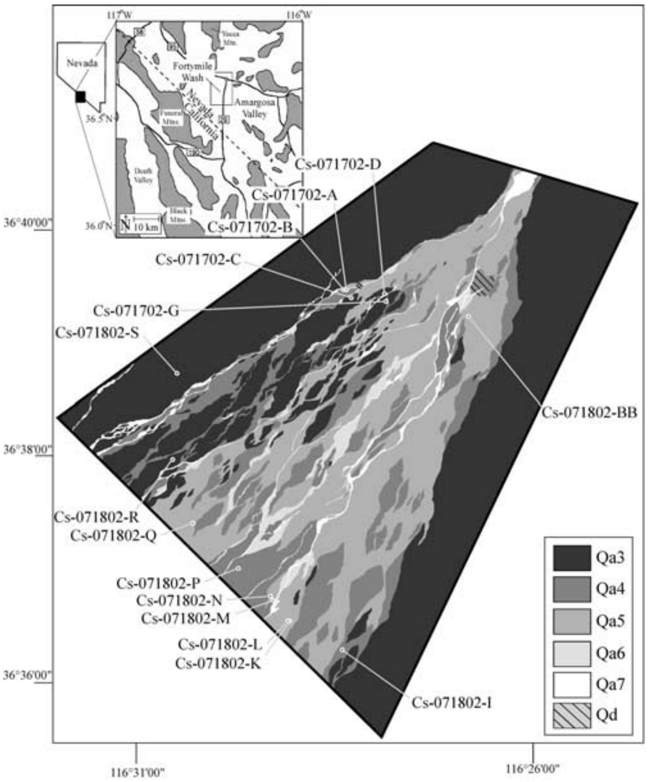

Figure 1. Soil-geomorphic map of the Fortymile Wash alluvial fan, with ^{137}Cs sample locations shown. Approximate surface ages are Qa3 - middle to late Pleistocene, Qa4 - late Pleistocene, Qa5 - latest Pleistocene to early Holocene, Qa6 - mid to late Holocene, and Qa7 - late Holocene to active, based on correlation with the *Whitney et al.* [2004] chronology.

by integrating soil-geomorphic mapping and geochronology on nearby Yucca and Crater Flats.

[6] Beginning in the mid-1950s and continuing through the mid-1960s, world-wide above-ground nuclear tests introduced radioactive fallout ^{137}Cs into the soil [*He and Walling*, 1997]. Fourteen ^{137}Cs profiles from the mapped area of Fortymile Wash fan were collected and analyzed (Figure 1 and Table 1). Bulk samples were collected between 0–3 cm and 3–6 cm. The fraction of total ^{137}Cs in the upper half of the profile typically varied between 80 and 95%, suggesting that very little ^{137}Cs has diffused below 6 cm in these profiles.

[7] The radionuclide profile shape may reflect the influence of both surface erosion/deposition and redistribution within the soil column. Except for the active channel, all of the older surfaces have not experienced significant flooding for several thousand years. Eolian deposition rates inferred for the Fortymile Wash fan by *Reheis et al.* [1995], suggest that eolian erosion/deposition is less than or equal to several millimeters over the 50-yr time scale since the introduction of fallout ^{137}Cs. Due to the relative stability of the Fortymile Wash fan surfaces to both fluvial and eolian erosion/deposition, we expect the shape of the ^{137}Cs profile to predominantly reflect redistribution processes. The exception is the active channel. In this case the Cs profile may be strongly influenced by fluvial mixing in addition to infiltration and other mixing processes.

3. Model Description

[8] The convection-diffusion equation (CDE) is a classic model for radionuclide dispersion in the soil profile [*He and Walling*, 1997; *Toso and Velasco*, 2001]. ^{137}Cs can be adsorbed onto soil particles and transported into the soil profile by infiltration and bulk-mixing processes (i.e. wetting/drying and freeze/thaw cycles, and bioturbation). The CDE model does not represent these processes explicitly, but instead treats soil-transport processes as analogous to the hydrodynamic dispersion of a passive tracer, or to the Brownian motion of molecules in a liquid.

[9] The equation describing the evolution of radionuclide concentration C by diffusive processes is given by the solution to the diffusion equation in a semi-infinite medium with no-flux boundary condition at the surface and a fallout mass $C_0 d_w$ input at $z = 0$ at $t = 0$ [*Carslaw and Jaeger*, 1959]:

$$\frac{C(z,t)}{C_0 d_w} = \frac{1}{\sqrt{\pi D t}} e^{-z^2/4Dt} \qquad (1)$$

where C_0 is the initial concentration within a thin surface layer of thickness d_w, z is depth in the soil profile, D is the diffusivity, and t is time following deposition. Equation (1) assumes that the convective component of transport is negligible (reducing the CDE model to a simple diffusion model). This is an appropriate assumption in cases where

Table 1. ^{137}Cs Data and Inferred Diffusivity Values for Geomorphic Surfaces on the Fortymile Wash Alluvial Fan

Sample ID	Total ^{137}Cs (pCi/g)	Fraction at 3 cm	D (cm^2/yr)	Geomorphic Unit
Cs-071802-A	0.313	0.8274	0.047	Qa6
Cs-071802-B	0.271	0.5387	0.165	Qa7
Cs-071802-C	0.258	0.8100	0.052	Qa3
Cs-071802-E	0.208	0.7644	0.063	Qa3
Cs-071802-G	0.118	0.9593	0.021	Qa3
Cs-071802-I	0.388	0.9614	0.021	Qa4
Cs-071802-J	0.155	0.6387	0.108	Qa6
Cs-071802-K	0.340	0.9558	0.022	Qa5
Cs-071802-N	0.218	0.9082	0.031	Qa5
Cs-071802-P	0.245	0.9428	0.024	Qa4
Cs-071802-Q	0.205	0.9951	0.011	Qa5
Cs-071802-R	0.237	0.9578	0.021	Qa3
Cs-071802-S	0.285	0.8807	0.037	Qa3
Cs-071802-BB	0.321	0.7943	0.056	Qa5

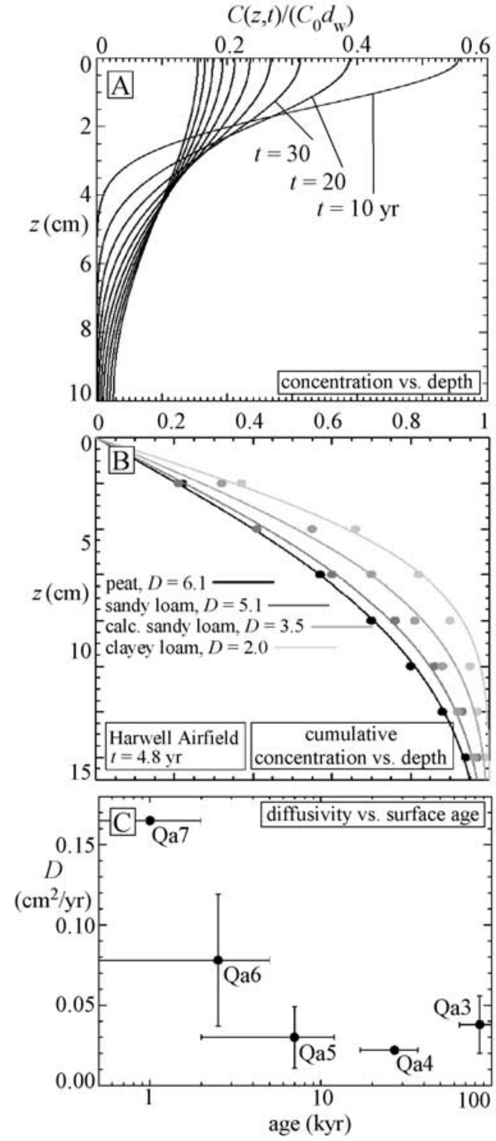

Figure 2. (a) Diffusion-model results for normalized concentration vs. depth from t = 10 to 100 yr with D = 0.1 cm^2/yr. (b) Cumulative ^{137}Cs concentration as a function of depth in four different soil types at Harwell, England (data from *Gale* [1964]), along with best-fit results for the diffusion model. (c) Plot of diffusivity values vs. surface age for the ^{137}Cs profiles on Fortymile Wash alluvial fan. Horizontal "error" bars represent the range of age estimates for each surface based on *Whitney et al.* [2004]. Vertical bars represent the standard deviation of diffusivity values for all of the sampled collected on each surface.

the maximum radionuclide concentration is observed to be at or very close to the surface. In cases with significant downward migration (i.e. with high convection rates or "old" profiles), the concentration maximum migrates deeper into the profile and the effects of convection and diffusion must both be inferred. Equation (1) also approximates the permeable soil layer as semi-infinite. This is an accurate approximation for man-made fallout profiles, even in a soil with a calcic horizon at depth, because radionuclides do not penetrate more than several cm into desert soils over decadal time scales. It should also be noted that radioactive decay need not be considered explicitly in this analysis because decay does not affect the relative radionuclide concentration at different depth intervals, only their absolute values. Figure 2a gives an example of the diffusion model (equation (1)) using D = 0.1 cm^2/yr and t = 10–100 yr.

[10] Radionuclide concentrations measured in the field are bulk measurements rather than point measurements because the concentration within different depth intervals is measured. For the purposes of extracting model parameters from bulk measurements, it is most accurate to represent measured data cumulatively as the fraction of total concentration to a given depth. To compare the diffusion model predictions to this normalized cumulative curve, it is necessary to integrate (1) to give

$$\int_0^z \frac{C(\eta, t)}{C_0 d_w} d\eta = \mathrm{erf}\left(\frac{z}{\sqrt{4Dt}}\right) \tag{2}$$

where erf is the error function and η is an integration variable for depth. Figure 2b compares the diffusion model prediction to ^{137}Cs profiles measured during a 4.7-yr field experiment at Harwell Airfield in Oxfordshire, England [*Gale et al.*, 1964]. Although these data are from a very different climatic regime than Fortymile Wash, the results illustrate the precision of the diffusion model in a way that profiles in desert soils cannot show because shallow penetration limits the spatial resolution of desert-soil profiles. To obtain the results in Figure 2b, the measured ^{137}Cs profiles were fit to (2) using the Levenberg-Marquardt algorithm, which yields a least-squares fit to the error function [*Press et al.*, 1992]. Diffusivity values are lowest for clayey and calcareous soils at Harwell, increasing in value in sandy soils and peat.

[11] Table 1 summarizes the data and results from Fortymile Wash. To calculate D, the fraction of total activity at 3 cm was first computed by dividing the activity from 0–3 cm by the total activity from 0–6 cm. Equation (2) was then used to infer the value of the error function argument, equal to $z/\sqrt{4Dt}$, corresponding to the fraction of activity at 3 cm after 50 yr of diffusion following nuclear testing. A table of calculated error function values was used for this purpose. The value of the error function argument was then used to solve for D (column 3 in Table 1). Figure 2c illustrates the relationship between diffusivity values and geomorphic-surface age on the Fortymile Wash fan. The vertical bars are the standard deviation of D values obtained on different profiles of the same soil-geomorphic unit. The horizontal bars correspond to the age range for each unit based on the available age control [*Whitney et al.*, 2004]. The plot exhibits an inverse relationship between diffusivity and age, with values increasing slightly on Qa3 (the oldest surface) relative to those on Qa4.

4. Discussion

[12] Radionuclides may be redistributed within the soil profile through entrainment and redeposition by infiltrating runoff and/or by bulk-mixing processes including freeze/thaw, wetting/drying, and bioturbation. The rate of transport by infiltration can be expected to correlate positively with

hydrologic conductivity, and hence negatively with surface age based on the results of *Young et al.* [2004]. Transport rates associated with bulk-mixing processes can be expected to correlate positively with surface age because water-storage potential and clay fraction both increase with age, and because water storage and clay fraction promote freeze/thaw and wetting/drying cycles. The inverse correlation between diffusivity values and surface age we found at Fortymile Wash fan is most consistent with the hypothesis that radionuclide transport is dominated by infiltrative transport. In fact, the relationship between diffusivity and age in Figure 2c closely parallels the relationship between hydrologic conductivity and age documented by *Young et al.* [2004], with both exhibiting the largest differences between the youngest (Qa5–Qa7) surfaces compared to the oldest (Qa3–Qa5) surfaces. The slight increase in diffusivity on the oldest surface may reflect the role of a more gravelly parent material, since older Pleistocene alluvial-fan deposits are often coarser and more poorly sorted than latest Pleistocene and Holocene deposits [*Mayer and Bull*, 1981]. The greater permeability associated with coarser parent material on this surface may offset the effect of a thicker A_v horizon on this surface.

[13] Our conclusion that radionuclide transport is related primarily to runoff infiltration rather than bulk-mixing processes differs somewhat from the conclusions of previous research. *Romney et al.* [1970] argued that radionuclide migration in montmorillonite and illite-bearing soils is predominantly caused by freeze/thaw and wetting/drying cycles that granulate the soil by aggregation and dispersion, enhancing the mechanical movement and downward migration of high-density particles. The reason for the predominant role of infiltration in Fortymile Wash is most likely that the sandy texture and lower clay content of these soils (relative to those in more temperate environments) increases their hydrologic conductivity and hence their infiltrative-transport efficiency.

5. Conclusions

[14] Modeling the transport and fate of radionuclides in the environment is a critical problem in applied geoscience. Our work emphasizes the importance of the geomorphic context in quantifying rates of radionuclide migration in soils. The inverse correlation between radionuclide migration and geomorphic-surface age we documented provides a basis for using point-based calibrations together with soil-geomorphic mapping to predict diffusivity values across the entire landscape. More data is needed to test the robustness of this relationship, however. Our results illustrate that infiltrative transport of radionuclide-bearing particles is the key transport mechanism in the sandy alluvial soils typical of the arid southwestern U.S.

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