Short-term soil mixing quantified with fallout radionuclides

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ABSTRACT

Soil mixing plays a significant role in contaminant transport, carbon sequestration, and landscape evolution, yet the rates and driving mechanisms are poorly constrained. Here we use depth profiles and advection-diffusion modeling of fallout nuclides to quantify differences in short-term (<100 yr) physical soil mixing across contrasting landscapes. We constrain advection in soils using the distribution of cosmogenic ^{7}Be and weapons-derived isotopes, and quantify mixing with a steady-state model of vertical ^{210}Pb transport. On a forested landscape in the Bega Valley in southeastern Australia and on grasslands in Marin County, California, where bioturbation is documented as the dominant sediment transport mechanism, we calculate diffusion-like mixing coefficients of 1–2 cm^2 yr^{-1}. In montane forest soils of northern New England, we observe little field evidence of short-term mixing, and find that the traditional advection-diffusion model fails to describe ^{210}Pb profiles. Because nuclide profiles here can be described with a simple model of litterfall, organic matter decay, and radioactive decay, we argue that diffusion-like processes are barely active on short time scales, and that the advection-diffusion model overestimates diffusion-like transport. While animal bioturbation and soil freezing cycles have little effect on the fate of elements in New England, physical soil mixing drives transport at Bega Valley and Marin County. We suggest that the absence of soil stirring that we quantify in New England forests may explain the slow physical erosion here (~0.2 cm/k.y.) relative to the actively bioturbated soils of Bega Valley and Marin County (5–10 cm/k.y.).

Keywords: bioturbation, erosion, fallout, mixing, soil.

INTRODUCTION

Animal burrowing, plant rooting, freeze-thaw cycles, and tree-throw processes mix soil directly by moving particles, and indirectly by creating conduits through which water can wash soil to depth. Because soil mixing directly drives creep—one of the most pervasive forms of erosion on upland, soil-mantled landscapes—a quantitative understanding of mixing processes is critical to accurately describe landscape evolution (Gabet, 2000; Yoo et al., 2005a). Furthermore, since soils are a large reservoir of carbon, essential nutrients, and contaminants, soil mixing can play a major role in the biogeochemical cycling of elements in the terrestrial environment (Yoo et al., 2005b).

Despite the importance of characterizing and quantifying soil mixing, studies doing so are rare. Humphreys (1994) used a detailed depth analysis of soil fabric to map the distribution and pattern of bioturbation in soil profiles in southeastern Australia. Disturbed soil horizons and a general lack of pedogenesis can also be applied to estimate the age of a tree-throw disturbance in forest soils (Ulanova, 2000). Heimsath et al. (2002) used single-grain optically stimulated luminescence dating to document vertical mixing rates of 10–40 cm/k.y. in southeastern Australia. Similarly, if dateable volcanic deposits are present, soil mixing rates can be determined by measuring the incorporation of the layer within the soil column (Roering et al., 2002).

Many such studies are resource intensive or rely on discrete, dateable horizons in the soil column. Here we exploit nuclides with short lives ($t_{1/2}$ < 500 yr) that are deposited to Earth's surface by precipitation to quantify soil mixing on decadal time scales. We show that the diffusion-like transport of radionuclides in soil varies significantly across contrasting field sites, and we attribute this variation to mechanistic differences in the physical mixing processes. The short-term diffusion-like mixing rates we quantify here correlate well with physical denudation rates determined by independent methods, which underscores the potential significance of processes operating on this time scale. We also demonstrate that the widely used model for describing nuclide transport via advection-diffusion processes can fail under coniferous forest cover in New England.

FALLOUT AS TRACERS OF SURFACE PROCESSES

Fallout radionuclides are used extensively in erosion and sediment transport studies on both agricultural and forested landscapes. Atmospherically delivered ^{210}Pb ($t_{1/2}$ = 22 yr, ^{210}Pb$_{ex}$, "in excess" of that supported directly by soil ^{222}Rn decay), cosmogenic ^{7}Be ($t_{1/2}$ = 53 d), and weapons-derived ^{137}Cs ($t_{1/2}$ = 30 yr) and ^{241}Am ($t_{1/2}$ = 433 yr) can be used alone or simultaneously to quantify and trace erosional processes (Wallbrink and Murray, 1996; Walling et al., 1999; Whiting et al., 2001), date and source sediments (Appleby and Oldfield, 1992), and determine sediment transit times (Bonniwell et al., 1999). Fallout radionuclides are useful geomorphic tools because of their unique atmospheric source term, but the technique relies on the assumption that the radionuclides are geochemically immobile and thus effective particle tracers.

Because fallout nuclides are deposited at the soil surface, mixing processes increase the dispersion of nuclides with depth and the overall downward transport rate (Dörr, 1995; Tyler et al., 2001). For example, on agricultural landscapes, where tilling homogenizes the soil to the depth of the plow, a unique, well-mixed ^{137}Cs profile captures the process (Walling et al., 1999). Field evidence suggests that some soils are homogenized naturally by burrowing organisms (Black and Montgomery, 1991), which would mix fallout isotopes to a depth governed by the flora and fauna at the site. However, in undisturbed forest soils where bioturbation is less obvious, sediment transport mechanisms can be more elusive and difficult to quantify. By measuring the vertical distribution of fallout radionuclides in soils and calculating diffusion-like coefficients, we quantify mixing rates and sediment transport mechanisms that operate on the short but important time scale of 10–100 yr.

Soils were collected from clean faces of excavated pits. We avoided sites obviously disturbed by tree-throw, as the radionuclides we measure only capture processes operating over the last century: animal bioturbation, freeze-thaw, plant rooting, etc. We sampled only on upland, convex hillslopes, where advective transport (i.e., erosion via overland flow) was unlikely. Mineral samples were sieved <2 mm, and organic samples were ground for the direct determination of ^{241}Am, ^{7}Be, ^{137}Cs, ^{210}Pb, and ^{226}Ra by gamma ray analysis; we used corrections for photon self-absorption when appropriate (Cutshall et al., 1983). Excess ^{210}Pb was calculated at each point in the soil profile using the depth distributions of ^{210}Pb and ^{226}Ra (Wallbrink and Murray, 1996).

FIELD SITES

We quantify soil mixing on three well-studied landscapes with contrasting climate, geology, and vegetation. One is a subbasin of the Nunnock River in the Bega Valley of southeastern Australia (Heimsath et al., 2000, 2002). Here, a thin layer of *Sclerophyll* litter is underlain by mineral soils

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ranging from 10 cm thick on convex noses to 90 cm thick in convergent hollows. Evidence of bioturbation by wombats, worms, and insects is observed in soil profiles. We also sampled a grassland basin in Tennessee Valley, Marin County, California, where pocket gopher burrowing is the dominant sediment transport mechanism (Black and Montgomery, 1991; Yoo et al., 2005a). Soil thickness generally ranges from 20 cm on the ridge crests to >1 m in the hollows. Marin County and Bega Valley have Mediterranean and temperate climates, respectively, and each typically receives between 0.8 and 1 m of rainfall annually. Our third site was in northern New England, where we sampled from the coniferous zone of the Hubbard Brook Experimental Forest (Likens and Bormann, 1995) and adjacent areas in the White Mountains of New Hampshire, USA. Our New England site stands apart from Marin County and Bega Valley due to the erosional effects of the last continental glaciation and its colder climate. Precipitation here averages 1.4 m yr^{-1}, with ~30% falling as snow, and temperatures typically range from –24 to 32 °C. These cold and acidic immature soils are relatively inhospitable to mesofauna (e.g., earthworms) common to other soils (Li et al., 2002). Mammals, arthropods, and insects were not observed to be a major factor affecting soil development. Tree-throw creates a pit-and-mound topography, and the soils are subjected to freeze-thaw cycles. Soil thickness ranges from 30 to 60 cm, with a 3–15 cm thick organic "forest floor" horizon.

SOIL MIXING FROM NUCLIDE DEPTH PROFILES

The steady-state distribution of $^{210}Pb_{ex}$ with depth (z) in soils can be described by solving the traditional advection-diffusion model (e.g., DeMaster and Cochran, 1982), where the amount of $^{210}Pb_{ex}$ in a particular volume of soil (A, in Bq cm^{-3}) is controlled by the initial amount (A_0), an advection term (v, in cm yr^{-1}), which is defined here as downward leaching of nuclides sorbed to colloidal materials, a diffusion-like mixing coefficient (D, in cm^2 yr^{-1}), and radioactive decay (λ, in yr^{-1}):

$$A(z) = A_0 \exp\left[\frac{v - \sqrt{v^2 + 4\lambda D}}{2D}(z)\right]. \quad (1)$$

Radioactive equilibrium is a reasonable assumption at our sites: They have remained free from anthropogenic disturbance for more than three half-lives of ^{210}Pb; precipitation (which controls deposition) has remained relatively constant during the twentieth century; and measured inventories are consistent with those predicted by fallout models. By modeling the distribution of a single nuclide in the soil column, it isn't possible to solve for unique values of v and D, and it is unrealistic to ignore either process in soils. While our goal is to quantify D to measure soil mixing, we need to constrain advection that might occur. For this, we use the distribution of weapons-derived nuclides (^{137}Cs, ^{241}Am) in the soil profile, which had a pulse-like input to soils during the 1950s and 1960s, with a strong global depositional maximum in 1963–1964 (U.S. ERDA, 1977). Using the precise position of the subsurface concentration maximum of weapons fallout in our soils, we determine v (Fig. 1).

While advection-diffusion models are the most common method for describing the depth distribution of fallout nuclides (e.g., Walling et al., 1999, and references therein), an alternative model for describing radionuclide depth profiles may be required for our sites in New England since >70% of the total $^{210}Pb_{ex}$ inventory resides in organic matter. Organic matter decomposition must control the concentration of $^{210}Pb_{ex}$ here, since the half-life of ^{210}Pb is nearly an order of magnitude higher than the half-life of organic matter. As organics decompose, $^{210}Pb_{ex}$ concentrations will increase from relative carbon loss. A modified constant initial concentration model (CIC; Appleby and Oldfield, 1992) can be applied to our measured $^{210}Pb_{ex}$ profiles, assuming that litterfall and ^{210}Pb flux is relatively constant over time, and that the contribution of root mass in the O horizon is relatively low. The $^{210}Pb_{ex}$ concentration (C) over time is defined by

$$C(t) = \frac{C_o e^{-\lambda t}}{f(t)}, \quad (2)$$

where C_0 is the initial $^{210}Pb_{ex}$ concentration measured in fresh litter, λ is the ^{210}Pb decay constant (0.031 yr^{-1}), and $f(t)$ is the fraction of organic matter remaining at time t. We posit that the only difference between the CIC model and our data is an exponential-based organic matter decomposition function, $f(t)$. This steady-state model assumes that total decomposition in the O horizons is equivalent to litterfall inputs, and the model is run for 125 yr to allow for an equilibrium $^{210}Pb_{ex}$ profile and organic matter pool. The decomposition functions, $f(t)$, that provide the best match between equation 2 and our observed $^{210}Pb_{ex}$ concentration-depth profiles can be evaluated using our measurements of forest floor mass and with additional data on organic matter turnover reported for similar forests (Currie and Aber, 1997; Berg, 2000).

We first tried to apply a simple advection-decay model (e.g., Roberts et al., 1997) to our $^{210}Pb_{ex}$ profiles, using v determined with the weapons isotopes and assuming $D = 0$. Advection alone failed to describe the vertical distribution of $^{210}Pb_{ex}$ at all of our sites. We then kept advection constant and identified the best-fit D by minimizing the percentage of the inventory that the advection-diffusion model misplaced (Fig. 2). We optimized the fit on the upper 15–20 cm of soil,

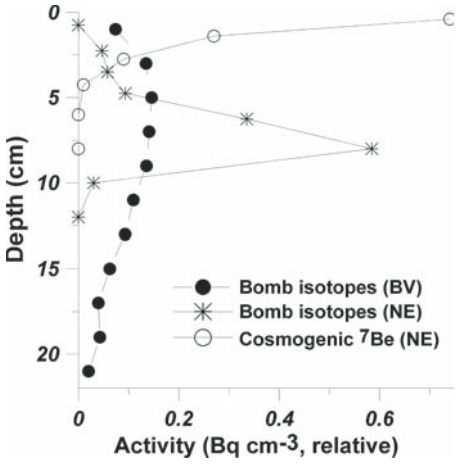

Figure 1. Depth profiles of cosmogenic ^{7}Be and weapons-fallout nuclides at New England (NE) and Bega Valley (BV). ^{7}Be is largely (>80%) retained in the upper 2 cm of soil. Weapons-fallout isotopes display a sharp subsurface peak at New England because of limited mixing. At Bega Valley, similar isotopes are more dispersed presumably from bioturbation. We calculate v by using the position of the subsurface maximum and assume that it tracks the 1963 deposition spike: v ranged from 0.1 to 0.2 cm yr^{-1} at New England, and from 0.07 to 0.11 cm yr^{-1} at Bega Valley and Marin County.

the portion of soil with the majority of the nuclide inventory and the portion most likely to be influenced by organisms. At Bega Valley and Marin County, the advection-diffusion model described the $^{210}Pb_{ex}$ profiles well, as total misplaced inventory was typically <18% (Fig. 2; Appendix in the GSA Data Repository[1]). Diffusion coefficients D can then be used to calculate mixing time constants τ for a soil of thickness L ($\tau = L^2D^{-1}$). A soil 35 cm thick at Bega Valley is "turned over," or mixed, every 1200 yr, and a similar thickness of soil at Marin County is mixed every 660 yr. If we use the mixing time scale for a soil of 35 cm thickness at Bega Valley and assume that a particle travels 0.5L during τ, we calculate grain-scale vertical displacement velocities of 1–2 cm per century. Surprisingly, these values are consistent with the 1–4 cm per century grain-scale velocities calculated here by luminescence dating (Heimsath et al., 2002).

In contrast with Bega Valley and Marin County, a simple advection-diffusion model did not fit the data well in New England; our best model-data fits had misplaced inventory >30% (Fig. 2C). In addition, D values calculated here were on the order of 1 cm^2 yr^{-1}, a

[1]GSA Data Repository item 2007054, Appendix 1, Testing ^{210}Pb-derived D values, is available online at www.geosociety.org/pubs/ft2007.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

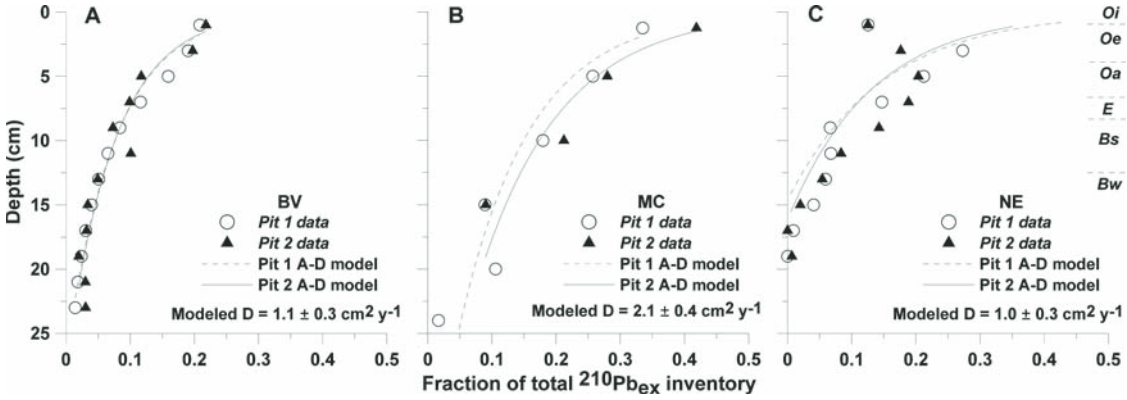

Figure 2. $^{210}Pb_{ex}$ data and A-D (advection-diffusion) model given by equation 1; *v* was calculated using weapons-derived isotopes (Fig. 1); we then solved for a *D* that best matched our data: 80%–90% agreement at Bega Valley (BV) (A) and Marin County (MC) (B); in New England (NE) (C), fits were typically <70%. See the Appendix (see footnote 1) for more details on fits and modeling. Soil horizons observed at New England given; no equivalent horizon boundaries exist at Bega Valley and Marin County. Oi—fresh litter; Oe—moderately decomposed litter; Oa—humus; E—strongly leached zone; Bs—zone with strong illuviation of iron oxides; Bw—chemically altered zone with weak illuviation.

rate inconsistent with the preservation of the weapons spike (Fig. 1; Figs. DR2 and DR3 in Appendix). It is possible that the initial infiltration of rainwater during intense storms through the porous O horizon could result in subsurface $^{210}Pb_{ex}$ peaks not accounted for by a constant *v* model (Fig. 2C). Profiles of cosmogenic ^{7}Be would capture this process well because of its short life and its association with large rain events (Olsen et al., 1985). We find instead that episodic vertical transport is low at all three sites: ^{7}Be activity declines rapidly with depth, and the upper 2 cm of soil typically retains >80% of the ^{7}Be inventory (Fig. 1). ^{7}Be was completely retained in the upper 1–2 cm of soil at Bega Valley and Marin County.

Decomposition functions required to fit the CIC model to the $^{210}Pb_{ex}$ data are consistent with organic matter turnover times and litter decomposition rates reported for similar forests (Berg, 2000). Specifically, Currie and Aber (1997) determined steady-state O horizon turnover times of ~40 yr using a model of CO_2 efflux and dissolved losses for coniferous forests in New Hampshire. Our calculated decomposition functions yield steady-state O horizon turnover times of 25–45 yr for sites in the same region (Fig. 3), and allow for a steady-state O horizon mass of 7–9 kg m^{-2}, consistent with our measurements. Because $^{210}Pb_{ex}$ profiles can be explained well with a model accounting for only radioactive decay, burial, and organic matter decomposition, we argue that random bioturbation and stirring, processes that can drive sediment transport, are minimal here. This conclusion is supported by field evidence: In Spodosols of New England, the 3–16 cm thick O horizon is well defined by distinct layers of fresh litter (Oi), moderately decomposed litter (Oe), and humus (Oa). Beneath the O horizon is typically a strongly leached E horizon, which is underlain by various spodic horizons (Bs), where iron and aluminum have accumulated (Fig. 2C). Soils at Bega Valley and Marin County lack such distinct pedogenic boundaries.

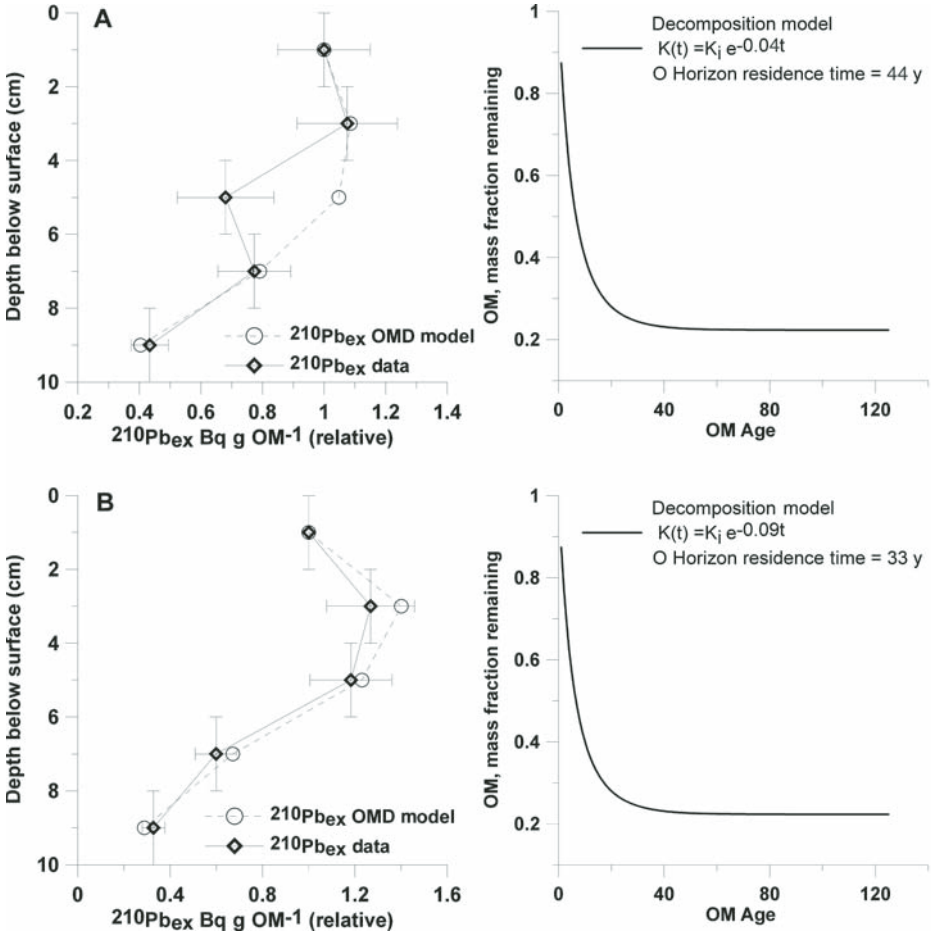

Figure 3. Depth profiles of $^{210}Pb_{ex}$ in O horizons sampled from two coniferous forest plots (A and B) in New England and steady-state $^{210}Pb_{ex}$ model results, $C(t) = C_0 e^{-\lambda t}/f(t)$, where *f(t)* = fraction of organic matter remaining. Best-fit exponential-based organic matter decomposition functions *f(t)* given at right, where *K* is the organic matter decay term and K_i is the initial decomposition rate (in yr^{-1}, K_i = 0.06–0.14 for A and B). We iteratively solved for K_i and the fractional rate loss constant of *K* (–0.04 and –0.09 for A and B, respectively) by searching for the best match between the $^{210}Pb_{ex}$ data and equation 2. OM—organic matter; OMD—organic matter decay.

Our work demonstrates quantitatively how process rates can vary in different geological settings on a time scale that is traditionally difficult to capture. Short-term diffusion-like mixing rates range by an order of magnitude across our sites. Neither animal bioturbation nor freeze-thaw cycles, both postulated as diffusion-like processes (Anderson, 2002; Yoo et al., 2005a), effectively mix the soil column in New England. The persistence of an insulating snowpack cover

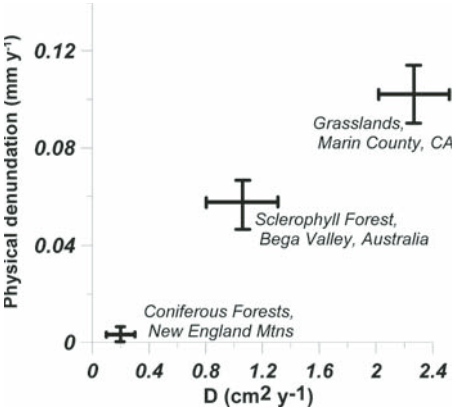

Figure 4. Landscape denudation measured by sediment traps and cosmogenic nuclides (reported elsewhere) versus short-term soil mixing rates (calculated here). A maximum *D* of 0.2 cm² yr⁻¹ was calculated for New England soils via a numerical simulation of the diffusion of the weapons peak (Appendix; see footnote 1). Bar width represents variation (as standard deviation) that we measured in individual profiles at each site; bar height represents one standard deviation reported for the erosion rate.

during the winter months probably minimizes the frequency of soil freezing cycles here (Likens and Bormann, 1995). In contrast, short-term diffusion-like processes can homogenize the soil at Bega Valley and Marin County on time scales of ~0.5–1 k.y. We also provide insight into the potential significance of short-term diffusion-like processes in landscape evolution. Mixing rates determined here are proportional to physical denudation rates measured by others (Fig. 4), and the short-term mixing rates we measure at Bega Valley are comparable to the longer-term mixing rates reported by Heimsath et al. (2002). These results are interesting because tree-throw and landslides, which can occur episodically on time scales >100 yr, are thought to be significant sediment transport mechanisms (e.g., Dietrich et al., 2003). The presence of a fibrous, porous organic horizon in northern New England that is not randomly stirred may protect mineral soil and probably limits physical denudation here. Our work suggests that relatively short-term soil mixing processes may limit erosion rates, and that there is a rough steady state between the continuous processes of soil mixing and long-term landscape evolution. We conclude that short-term diffusion-like processes are significant and can play an important role in landscape evolution and the fate of any element delivered to the land surface.

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APPENDIX: Testing the ^{210}Pb-derived diffusion values (*D*) with a numerical simulation of the subsurface weapons-spike profile.

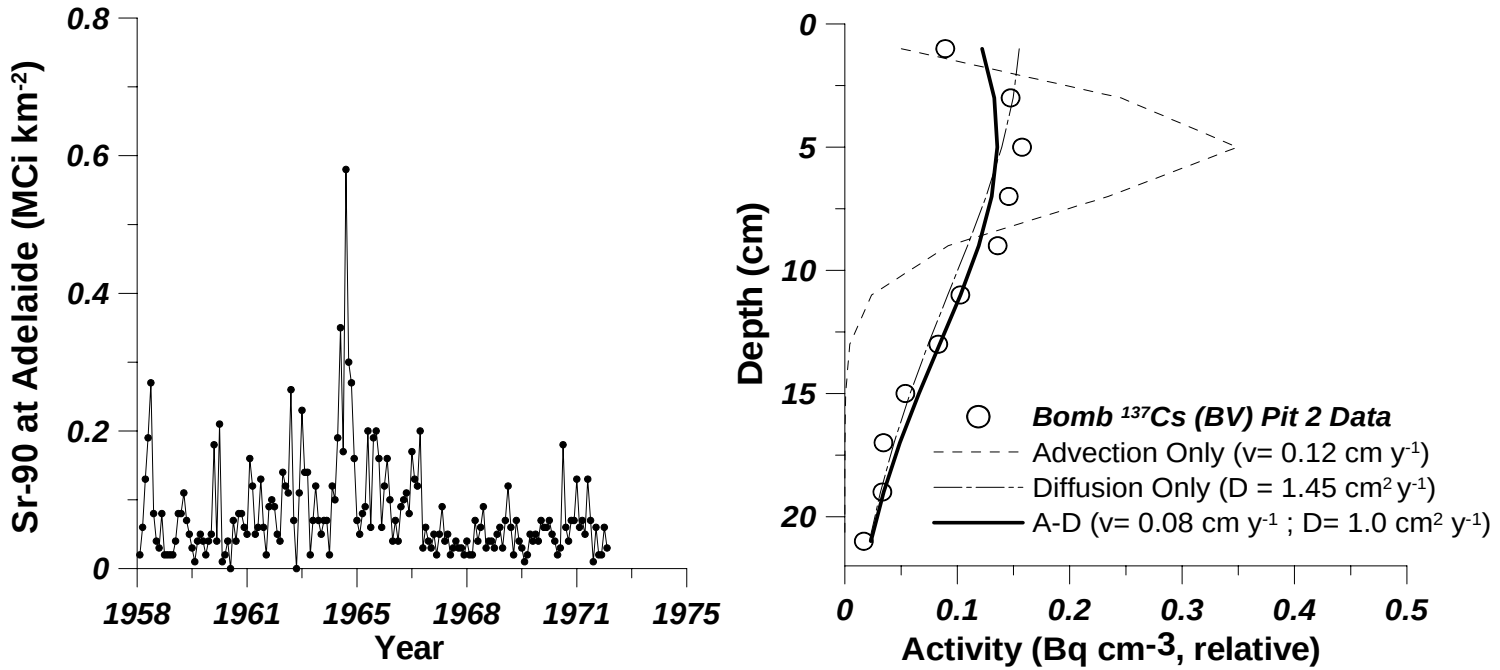

Figure DR1. Monthly Sr-90 deposition measured in Adelaide, Australia between 1958 and 1974 (U.S. ERDA, 1977). We use this input function (left) to feed a dynamic transport model (right) of the ^{137}Cs profile in soils at BV. We normalized the ^{90}Sr input data to 1, so that that transport model is not dependent on the absolute amount of weapons fallout measured. Instead, the model relies on the measured temporal distribution of the fallout. The transport model was run from 1958 (earliest record of radioactive fallout at Adelaide) until the samples were collected from the field (2001 or 2005). We assume zero weapons fallout after 1974 (end of the US ERDA record), and run the model in 0.1 year time increments to minimize artifacts that could be introduced by numerical diffusion. The "advection-only" model simulates the position of the sub-surface ^{137}Cs peak well (at ~5 cm), but does not accurately recreate the broad shape of the observed profile. The best-fit "diffusion-only" model provides an upper limit of $D = 1.45$ cm^2 y^{-1} (an upper limit because of the zero advection assumption). However, the "diffusion-only" model fails to recreate the observed subsurface peak. In this case, the best-fit A-D model with $D = 1$ cm^2 y^{-1} explains 85% of the variance of the data, and the velocity and *D* values agree well with those derived via the steady-state model of ^{210}Pb transport (Figure 2 in manuscript).

APPENDIX: Advection-diffusion modeling of $^{210}Pb_{ex}$ profiles in NE: Detailed fitting and residuals

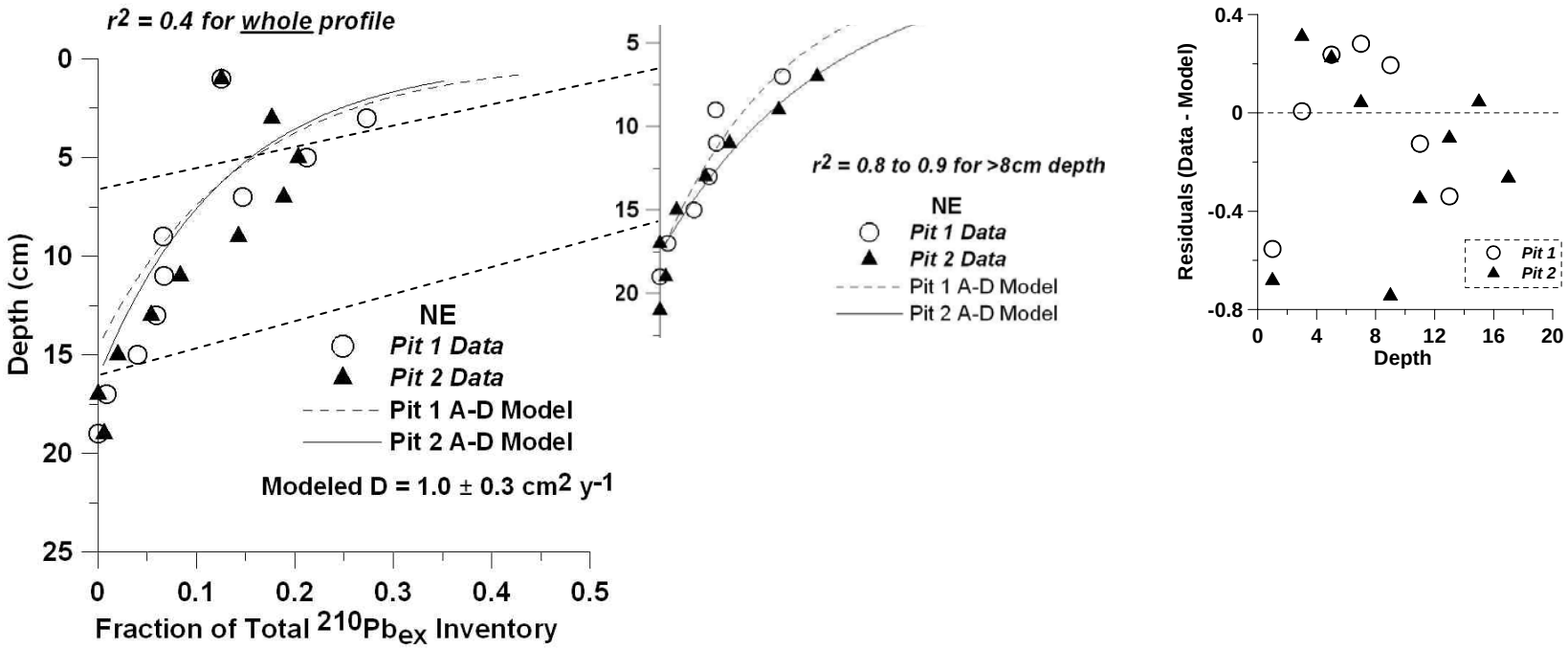

Figure DR2. The advection-dispersion model does not accurately describe the form of the $^{210}Pb_{ex}$ depth-profiles in the whole soil column in NE. This is most apparent by looking at the residuals, which are depth dependent (upper most samples have the largest deviation from the model) and do not sum to zero, indicating that they are not random. However, below 6 cm, the profiles take on a more true exponential form, where the a-d model can account for >80% of the data. This probably represents a change in dominant process: below 6 cm, organic matter is no longer decomposing at a rate that controls $^{210}Pb_{ex}$ concentration, and leaching (advection) probably begins to dominate in the soil profile.

APPENDIX: Testing the ^{210}Pb-derived diffusion values (*D*) at NE with a numerical simulation of the subsurface weapons-spike profile.

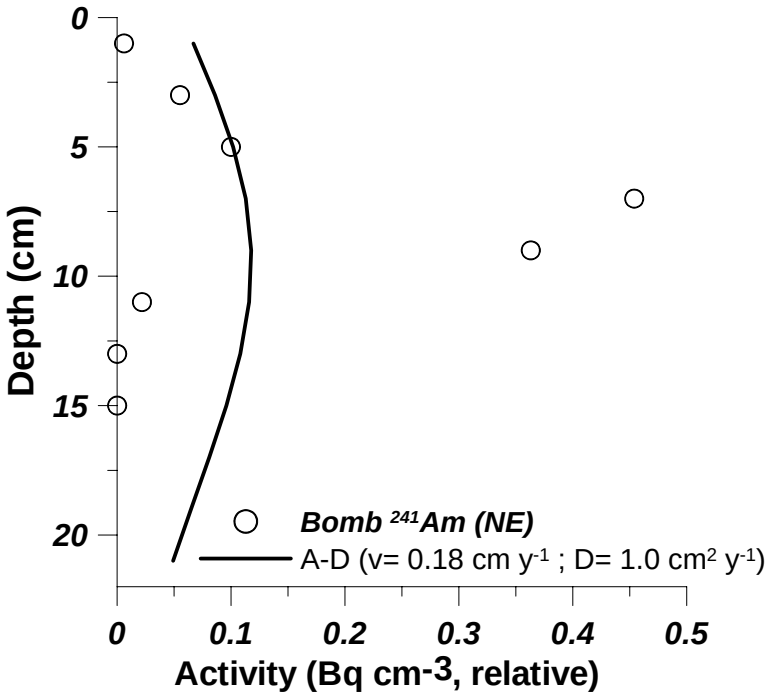

Figure DR3. Dynamic numerical model simulating the weapons fallout profile at a pit in NE using the best fit *D* derived from the steady-state ^{210}Pb A-D model (Figure 2, Data Repository Figure A2). The model itself is similar to that described in Figure A3, although we use input data measured in New York from 1956-1975 (U.S. ERDA, 1977). This simulation shows how poorly the traditional A-D model describes our data in surface soils in NE.