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Soil mixing depth after atmospheric deposition. I. Model development and validation

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ABSTRACT

Knowledge of a soil mixing depth, or the migration depth of various pollutants in soil, is necessary to assess the soil chemical concentration resulting from atmospheric deposition of a specific air emission source. A mathematical model has been developed that describes the depth and time behavior of the soil concentration of an inert chemical after atmospheric deposition on surfaces. The soil mixing model is based on one-dimensional diffusion theory, and analytic solutions have been derived for the cases of: (1) instantaneous surface deposition; (2) continuous surface deposition; and (3) a finite period of continuous surface deposition, followed by a deposition-free time period. Comparisons of the model with measured soil depth profiles resulting from atmospheric deposition showed good agreement for lead, cesium, and dioxins. The best-fit effective diffusion coefficients in undisturbed soils varied from 0.5 $cm^2 yr^{-1}$ to 2 $cm^2 yr^{-1}$. The soil mixing depth was found to be a strong function of the atmospheric deposition time period. Calculated soil mixing depths in undisturbed soils were 2 cm after one year, 5 cm after five years, and 10 cm after 20 years of continuous atmospheric deposition on the soil surface.

1. Introduction

Particulates released from an air emission source will deposit on nearby soil surfaces as a function of particle size, particle density, meteorological parameters, and surface conditions. Many air dispersion models, such as the widely-used AERMOD and CALPUFF models recommended by the U.S. Environmental Protection Agency (USEPA, 2005a), can predict the atmospheric deposition rate from a specific emission source on surfaces in terms of mass per unit time per unit surface area (*e.g.*, $g\,yr^{-1}\,cm^{-2}$). Even in undisturbed soils, the chemicals deposited onto the surface will mix downward into the soil column over time, as a result of various processes. These processes include physical mixing (*e.g.*, through the freeze-thaw cycle), chemical processes (*e.g.*, dissolution at the surface and adsorption to soil particles at some depth), and biological processes (*e.g.*, plant rooting and mixing by earthworms and other bioturbators).

As a result of these mixing processes, chemicals deposited on the surface of undisturbed soils are slowly distributed downward through the soil column as a function of time. Knowledge of the depth over which soil mixing occurs is necessary to assess a soil chemical concentration resulting from atmospheric deposition of a specific air emission source over time. The calculation of a soil concentration resulting from atmospheric deposition, without any removal or degradation of the chemical in soil, is very straightforward:

$$C_s = \frac{QT}{z_d} \tag{1}$$

where:

C_s = soil chemical concentration ($g\,cm^{-3}$)
Q = surface atmospheric deposition rate ($g\,yr^{-1}\,cm^{-2}$)
T = time period of deposition (yr)
z_d = soil mixing depth (cm)

The soil concentration C_s as calculated in Eq. (1) represents an average over the assumed soil mixing depth, and in any validation study, should be compared with measured soil data averaged over a similar depth. Since Eq. (1) does not contain any removal mechanisms, predicted soil concentrations will continue to increase over time, which will be valid only for highly immobile chemicals. A more complex version of Eq. (1) has been developed that includes chemical removal or degradation from soil by incorporating a soil

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loss parameter (USEPA, 2005b; Barton et al., 2010). The soil loss parameter as defined by USEPA (2005b) is primarily applicable to organic compounds and can consist of a combination of five separate removal mechanisms: biotic and abiotic degradation; soil erosion; surface runoff; leaching; and volatilization. The atmospheric deposition rate (Q), which can be a combination of dry and wet deposition, is a standard output from many air models such as AERMOD or CALPUFF, and the deposition time period (T) for industrial sources is usually well known. However, an appropriate soil mixing depth (z_d) is more uncertain. Intuitively, one would expect an increasing mixing depth in soil with increasing time. For human health risk assessments, the U.S. Environmental Protection Agency recommends 2 cm as a mixing depth for untilled soil to use with calculated surface deposition rates from an air model (USEPA, 2005b), but does not consider any time dependence of the 2-cm mixing depth. This paper shows that soil mixing depths can be much larger, and depend strongly on the atmospheric deposition time period.

Several technical papers have measured soil depth profiles of nuclear testing fallout of ^{137}Cs and naturally occurring fallout of ^{210}Pb (Barisic et al., 1999; Blagoeva and Zikovsky, 1995; Doering et al., 2006; He and Walling, 1997; Miller et al., 1990; VandenBygaart et al., 1999); the ^{210}Pb soil profiles are typically presented as “excess” ^{210}Pb, after correction for natural background ^{210}Pb from radon decay. The more recent ^{137}Cs deposition from the Chernobyl incident in 1986 has been monitored by Rosen et al. (1999) as a function of depth in Sweden soils from one to nine years after the incident. Fernandez et al. (2008) measured industrial soil lead *vs.* depth profiles near the site of a zinc smelter complex in France that operated from 1900 to the early 1960s, and Cernik et al. (1994) measured zinc and copper soil profiles near a brass smelter in Switzerland. Brzuzy and Hites (1995) have measured polychlorinated dibenzo-*p*-dioxins and dibenzofurans as a function of soil depth in Michigan. In general, the measured soil concentrations from surface deposition are found to migrate downward with increasing time, and to decrease with depth in an approximately exponential manner. Several empirical equations, which typically employ a decreasing exponential term with depth, have been developed to approximate the measured soil concentration *vs.* depth profiles (Barisic et al., 1999; Blagoeva and Zikovsky, 1995; Miller et al., 1990).

The measured soil depth profiles have also been modeled by one-dimensional diffusion theory, using an effective diffusion coefficient to approximate the soil mixing processes in undisturbed soil (Cernik et al., 1994; He and Walling, 1997; Kaste et al., 2007). Based on diffusion equation solutions from Lindstrom and Boersma (1971) that contained a vertical velocity term, He and Walling (1997) derived effective diffusion coefficients for UK soils of 0.4–0.5 cm^2 yr^{-1}, which were applicable for both ^{137}Cs and ^{210}Pb. Kaste et al. (2007), using a numerical solution of a similar advective-diffusion equation, found best-fit effective diffusion coefficients for ^{210}Pb of 0.2 cm^2 yr^{-1} in New England soils, 1 cm^2 yr^{-1} in Australian soils, and 2 cm^2 yr^{-1} in Marin County, CA grasslands.

2. Model development

Our objective was to derive analytic diffusion equation solutions for soil concentrations as a function of time for several realistic atmospheric deposition scenarios, and to compare these solutions with measured data. The one-dimensional diffusion equation was solved for the cases of: (1) instantaneous surface deposition; (2) continuous surface deposition; and (3) a finite period of continuous surface deposition, followed by a deposition-free time period. The basic assumption in this analysis is that the mixing processes in soil after surface deposition can be represented by the classic one-dimensional diffusion equation,

$$\frac{\partial C_s}{\partial t} = D_{eff}\frac{\partial^2 C_s}{\partial z^2} \tag{2}$$

where:

C_s = soil chemical concentration (g cm^{-3})
D_{eff} = effective diffusion coefficient (cm^2 yr^{-1})
z = depth (cm), with a surface at $z = 0$ and increasing z with depth
t = time (yr)

2.1. Soil concentration from instantaneous surface deposition

The solution to Eq. (2) for a semi-infinite solid with an instantaneous surface deposition source, M_0 (g cm^{-2}), applied at $t = 0$ over the surface at $z = 0$, is a simple expression for the case of a constant diffusion coefficient. The soil concentration as a function of depth and time ($t > 0$) is (Crank, 1975):

$$C_s(z,t) = \frac{M_0}{\sqrt{\pi D_{eff} t}}\exp\left(\frac{-z^2}{4D_{eff}t}\right) \tag{3}$$

where:

M_0 = instantaneous surface deposition mass per unit area (g cm^{-2})

2.2. Depth-averaged soil concentration from instantaneous surface deposition

The average soil concentration over any specific depth interval is calculated by integrating Eq. (3) over a given depth interval from $z = L_1$ to $z = L_2$ ($L_2 > L_1$):

$$C_{s,ave} = \frac{\int_{z=L_1}^{z=L_2} C_s \mathrm{d}z}{\int_{z=L_1}^{z=L_2} \mathrm{d}z} = \frac{M_0}{(L_2 - L_1)\sqrt{\pi D_{eff} t}}\int_{z=L_1}^{z=L_2}\left[\exp\left(\frac{-z^2}{4D_{eff}t}\right)\right]\mathrm{d}z \tag{4}$$

The solution of Eq. (4) for the depth-averaged soil concentration between $z = L_1$ and $z = L_2$ ($L_2 > L_1$) for an instantaneous surface deposition source becomes:

$$C_{s,ave} = \frac{M_0}{(L_2 - L_1)}\left[erf\left(\frac{L_2}{2\sqrt{D_{eff}t}}\right) - erf\left(\frac{L_1}{2\sqrt{D_{eff}t}}\right)\right] \tag{5}$$

where:

$erf(x)$ = error function.

2.3. Soil concentration from continuous surface deposition

The solution for a continuous surface deposition source for a semi-infinite solid is derived by integrating the instantaneous solution over time. For a constant surface deposition rate, Q (g yr^{-1} cm^{-2}), applied at $t \geq 0$ over the surface $z = 0$, the solution for soil concentration as a function of depth and time ($t > 0$) was presented by Carslaw and Jaeger (1959) as: