ASSESSING INHALATION EXPOSURE FROM AIRBORNE SOIL CONTAMINANTS

An Overview of Particle Transport and Resuspension Studies

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Abstract

A method of estimation of inhalation exposure to airborne soil contaminants is presented. This method is derived from studies of airborne soil particles with radioactive tags. The concentration of contaminants in air (g/m^3) can be derived from the product of M , the suspended respirable dust mass concentration (g/m^3), S , the concentration of contaminant in the soil (g/g), and E_f , an enhancement factor. Typical measurement methods and values of M , and E_f are given along with highlights of experiences with this method.

Introduction

Human exposure to airborne soil contaminants occurs primarily through inhalation of particles in the "respirable range", that is, with aerodynamic diameters less than $10\text{ }\mu\text{m}$ (PM-10). Other potential mechanisms of exposure are dermal uptake of contaminants from deposited particles, and ingestion of particles deposited on edible plants. But in this paper we consider only the potential inhalation of contaminants from suspended soil. While this exposure mechanism is unusual, there are cases where it is known to be important or needs to be assessed. Most of the information we have gained about this exposure mechanism comes from the investigation of alpha radioactivity attached to soil particles. In principle, what we have learned applies to other types of soil-borne contamination as well.

The transport of particles from a soil surface to air at the point of inhalation is a process of wind erosion, whereby particles are continuously freed from the aggregates of the surface soil, even at lower wind velocities than those required to initiate soil saltation at a threshold of about 6.5 m/s , or 14.5 mph . While basic studies of the wind erosion process have not provided us with a practical model, observations have helped to provide us with some tools for assessment of potential health effects (Shinn, Homan, and Robison, 1997).

Deposition and Resuspension

The atmospheric deposition of particles onto soil is usually the originating source of contamination, as for example, on soil downwind of a polluting

stack. We have found that most particles from atmospheric deposition do not readily migrate into the soil, but become firmly attached to soil aggregates. The speciation of the particle and chemistry of the soil play a role of course, but unless the soil is sterile sand, the top soil tends to retain the deposited particles. Countering the tendency for metals to migrate downward is the effect of mixing of the top soil layers by the impact of raindrops and by soil-stirring due to roots and animals. The effect of vegetative cover on the deposition process is minimal unless the intercepting vegetation is harvested or removed prior to rain events that will wash off and transport the particles to the soil. Eventually the particles reside in the top soil, however, the resulting concentrations of soil contaminants are never found to be horizontally homogeneous because of the complexity of the interception and deposition process. Thus, soil sampling is usually most representative when it is integrated over an area 10 m to 100 m in radius. It matters little whether the soil was contaminated by atmospheric deposition or by some other means, since the wind erosion process occurs from the top surface of soil which soon becomes well mixed in the first 2 cm in either case.

Resuspension is the term given to the process by which airborne contamination is transported from the soil surface by means of wind erosion, and by traffic or similar fugitive dust emissions. Although there are many useful studies of wind erosion, the release of contaminants by disaggregation in the soil and resuspension remains unpredictable in a mechanistic sense. Airborne contaminant concentrations may depend upon cycles of soil moisture, soil shrinking and swelling, soil freezing and thawing, daily cycles of heating and cooling, amount and type of vegetative cover, soil microtopography and roughness, history of tilling, traffic destruction of aggregates, and so on. The multitude of factors controlling the structure of soil, and the nature of adherence of contaminants to soil aggregates, prohibit a general and practical model. Observations of radionuclide resuspension, on the other hand, have assisted us for the purposes of assessment of risks to humans by inhalation exposure.

Estimation Of Airborne Contamination Concentrations.

Our field studies have utilized the principle that the airborne suspended mass, known as "total suspended particulates" (TSP), is more or less in steady-state equilibrium with the mechanisms of resuspension and deposition. So given that these processes are all local, and that the observations are not biased by long-range transport from some upwind source, the concentration, C , of contaminant species in air (g/m^3) is the product

$$C = E_f S M \quad (1)$$

where E_f is a factor representing the fraction of soil contaminant suspended, S is the contaminant concentration in the top soil (g/g) integrated across all aggregate sizes, and M is the suspended mass or TSP (g/m³) across all suspended particle sizes. The TSP that originates from soil, and contains no imported particles such as combustion products, has been found to be approximately log-normally distributed with diameter, having very little mass below 0.5 μm , and having a median aerodynamic diameter in the range 2 to 5 μm . This shows that most of the soil-originated TSP is in the respirable range (PM-10), but would be split down the middle of the distribution if we measured only a fine particle fraction defined as those particles less than 2.5 μm (PM-2.5). In actual cases, the particle size distribution will display deviations from this ideal log-normal distribution, as for example in high winds when there may be a second maximum above 10 μm . Usually, however, the ratio TSP/PM-10 is less than 2, typically 1.7. Rural annual TSP values typically have a median of 30 $\mu\text{g}/\text{m}^3$ with 98% less than 75 $\mu\text{g}/\text{m}^3$.

The factor E_f is a measure of the degree that the contaminant concentration in the respirable size range reflects the distribution of contaminants across all soil aggregate sizes. In fact, we also have found through observations that E_f is a sort of "enhancement factor", reflecting the availability of contaminants to be resuspended, and changing as the surface is disturbed by natural cycles, traffic, or construction. On a mass basis it can be shown that soil particles in the PM-10 size range are a small fraction, 1 to 5%, of the soil-texture size distribution. But observations show that, at least in the case of radionuclides as soil tags, the range of values of E_f is from 0.6 to 7, having the lowest values in the undisturbed state. Table 1 shows E_f values observed for radionuclide soil tags. Table 1A gives E_f observed under normal soil conditions and Table 1B shows how E_f varied with top soil disturbances (Shinn, 1992). It should be noted that during disturbance, both E_f and M would change independently.

Table 1A. Enhancement factors under normal conditions.

Site	E_f
Nevada Test Site, Area 5	0.87
Nevada Test Site, Area 11	1.04
Bikini fields	0.80
South Carolina, Field 1	0.21
South Carolina, Field 2	0.49
California, garden plot	0.73
mean (std. dev.)	0.7 (0.3)

Table 1B. Enhancement factors under disturbed conditions.

Disturbancetype	E_f
Soil thawing in springtime	6.5
Bulldozer blading of soil	3.9
After 60% vacuum cleanup	3.6
Wildfire; removing grasses	3.5
Raked off desert pavement	2.2
Soil dried, eroded 2 weeks	0.8
geometric mean	2.9

The problem of assessment of human health risk by inhalation exposure in average or disturbance conditions, therefore begins with predicting the TSP concentration (M), and the enhancement factor, E_f, but using measurements of soil contamination concentration, S, where available. The problem can be bounded by air sampling to obtain min and max values of M and E_f over soils of known S, or by comparison against the measurements made at similar sites where metals or radionuclides served as a particle tag. If airborne particle size distribution is beyond the scope of measurement, the ratio TSP/PM-10 is simpler to measure to correct for the respirable fraction.

Estimation of Airborne Particle Flux

The ultimate measure of the stability of an eroding surface is the characteristic vertical flux, F, of contaminant particles (PM-10) from the surface (g m⁻² s⁻¹). This is also the contaminant emission rate. In many cases where both the contaminant downward migration and the resuspension time-scales are observed, the latter is more important than the former. This is because the deposited contaminant is quickly immobilized in the soil, but the soil may erode continuously and rapidly, particularly if the surface is bare or disturbed. For long-lived contaminants, the exposure of the population in the long term due to resuspension may also need to be considered in a risk assessment. A method of estimation of the flux is therefore derived from

$$F = K (dC/dz) \quad (2)$$

where K is the turbulent diffusivity for heat in the atmospheric surface boundary layer, and dC/dz is the vertical gradient of airborne contaminant concentration in the layer from 0.25 m to 2 m above the soil surface. The hourly values of K can be measured in the field by conventional micrometeorological methods, and the vertical gradient dC/dz can be assumed to have similarity properties:

$$dC/dz = p(C/z) \quad (3)$$

where p is a parameter observed as the least-squares slope of measured $\log C$ vs $\log z$. The parameter p reflects the stability of the soil surface, and the assumed similarity means that locally and within a period of unchanging top soil conditions, the vertical gradient changes with C while p is constant. That dC/dz should scale as C/z is to be expected in surface boundary layer flows near the boundary ($z = 0$). Furthermore, the flux F is typically constant with height in the surface boundary layer between 0.25 m and 2 m heights, since K , the turbulent diffusivity:

$$K = \alpha u z \quad (4)$$

where u is wind speed at height z , and α is a coefficient (equal to the product of the Karman constant, the local drag coefficient, the ratio of diffusivity for heat to that for momentum, and the inverse local stability influence function). Taking K integrated over several days, and measuring u and C at the breathing height z_1 between 0.25 m and 2 m the flux is approximately:

$$F = \alpha p u_1 C_1 \quad (5)$$

where the coefficient, α , evaluated locally will be about 0.2 for typical bare soils, and the p values measured via air samplers over the height interval 0.25 m to 2 m. We have observed p -values in the range -0.06 to -0.6 with typical values -0.25. (Here the sign indicates that the concentration decreases with height and the gradient shows loss of contaminant from the soil.) It should be noted that u_1 and C_1 are not independent because C will also depend on wind speed. At a minimum we expect from observations that at lower wind speeds, C will be proportional to u^2 . When saltation begins to influence the erosion rate, and at higher wind speeds, C will be proportional to greater powers of u because the amount of TSP is stimulated by saltation.

Since the flux F is the aerosol emission rate it may need to be determined over seasonal periods to determine the annual emission rate and to model the risk of atmospheric transport of the contaminant to the local human population.

Verification Measurements

Specialized equipment may be useful to verify the airborne contaminant concentrations, aerosol size distributions, and the vertical flux, although many alternative standardized methods may be available. We utilize the standard high volume (HV) air samplers (TSP or PM-10 types) running at 68 m³/h (40 cfm) and use fiber filters weighed before and after exposure at

constant humidity and temperature to avoid hygroscopic errors. We then determine the contaminant species chemically. This approach allows us to separately obtain M , C , and E_f provided the surface soil contamination levels are known and one can be assured of no aerosols imported from upwind. Generally a relatively uniform level of surface contamination is required within a 100 m radius of the samplers.

A quick calculation will show how much air needs to be sampled to obtain the minimum detectability limit for the particular contaminant. In many cases, to minimize the length of sampling time a number of simultaneous air samplers would be necessary. For example, if the $E_f = 1$, total suspended particulates $M = 10 \mu\text{g}/\text{m}^3$ (clean air), and the concentration of contaminant in soil $S = 1\mu\text{g}/\text{g}$, the concentration, $C = 10^{-12} \text{ g}/\text{m}^3$. Assuming an analytical minimum detectability, $\text{MDA} = 10^{-9} \text{ g}$, of contaminant per sample, and sampling 10 times more volume than needed to attain the MDA, $V = 10 \text{ MDA}/C$, it would require a volume $V = 10^4 \text{ m}^3$ of air sampled. For HV operated at $68 \text{ m}^3/\text{h}$ that should require a sampling time of $147 \text{ h} = 6.1 \text{ days}$. To shorten this time we have operated as many as 6 simultaneous HV samplers with standard 8- by 10-inch fiber filters, or alternatively, we have operated an ultra-high-volume (UHV) air sampler running at $900 \text{ m}^3/\text{h}$ (530 cfm) using a 1 m^2 fiber filter. These large filters were ashed before weighing to obtain M .

Recommendations

It is possible to assess the health risk by inhalation exposure for soil-borne contaminants by considering the independent components of suspended mass and contaminant enhancement factors, and by obtaining the wind speeds and the concentration gradient factor p for particular sites. These are observable variables and are also a starting point for assessment of health risk by dermal uptake of airborne particle contaminants or by ingestion of contaminant particles on edible plants. Although mechanistic models are lacking, we have obtained sufficient information and experience to provide first-order estimates of the enhancement factors and p -parameters, and recommend that each site include an air sampling verification study.

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