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Measuring Plutonium Concentrations in Respirable Dust

In their report on the plutonium hazard in respirable dust, Johnson *et al.* (*1*) state that "the respirable fraction of surface dust was separated by ultrasonic dispersion and a standard water-sedimentation procedure." It is apparent that their respirable fraction includes particulate that is too large to fall within the respirable size range, and that the analytical results obtained after the sample preparation techniques described will not show the concentration of plutonium associated with the in situ respirable surface dust. My criticism has as its basis the following reasons.

1) Wet digestion with hydrogen peroxide and particle dispersion by sonication reduces or eliminates the binding mechanisms that hold respirable-size plutonium particles to nonrespirable-size dust particles in the surface soil. After altering the real in situ particle associations, it is wrong to assign the final value for the soil concentration of plutonium to the original respirable size fraction of the surface soil.

2) In using the sedimentation technique to isolate the respirable size fraction, it is wrong to base "threshold parameters" on particles having an effective maximum diameter of 5 μm and a density of 11.45 g/cm^3 because (i) A 5-μm PuO_2 particle having a density of 11.45 g/cm^3 has an equivalent aerodynamic size of about 17 μm, which is well above the respirable size range. (ii) By using the above threshold parameters, one includes in the respirable fraction much of the ordinary dust present that is well beyond the respirable size. For example, by Stokes' law, dust particles with a density of 1.5 g/cm^3 and a size of 23 μm may be shown to have sedimentation characteristics similar to those of PuO_2 with a size of 5 μm and a density (ρ) of 11.45 g/cm^3. A 23-μm (ρ = 1.5 g/cm^3) dust particle has an aerodynamic diameter of about 28 μm. It is the aerodynamic diameter that determines respirability.

This selection of threshold parameters may or may not give conservative results in assessing the hazard of plutonium in soil. The plutonium attached to host dust particles that are well above the respirable size range is included as respirable particulate, while nonrespirable dust particles with no attached plutonium are also included in the respirable dust fraction.

In any event, these methods of sample preparation and data analysis will not yield valid results. The most conservative approach would be to call all of the plutonium respirable, since that which is outdoors is virtually all in the respirable size range (the mean size at Rocky Flats is on the order of 0.3 μm or less, depending on source) when considered as unassociated with host soil particles. If one wants to know the concentration of plutonium actually associated with respirable dust particles, then a valid technique must be used. One such technique would be to sample by vacuum and collect by impaction, using an impactor that classifies the dust according to its aerodynamic size.

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We believe that it is valid to disperse the in situ particle associations. The procedure is used in an effort to overcome the variables associated with microaggregate stability, in order to achieve reproducible results and provide data that are comparable from season to season or site to site. Our definition of the respirable size fraction is that fraction of soil that includes plutonium oxide particles of the given size. This fraction does include other mineral particles of lower density and larger diameter (to 12.6 μm, based on an average mineral particle density of 2.65 g/cm^3, according to Stokes' equation).

It is irrelevant whether these other mineral particles are ever retained within the lung, although there is evidence of some retention (*1*). However, that does not render it unacceptable to use the weight of the entire fraction as a basis for expressing the concentration of the plutonium. This fraction does comprise the orders of particle sizes of concern for health.

We agree that the selection of threshold parameters could be based on an appropriate equivalent aerodynamic size in place of the actual particle size. However, this is not necessarily a more conservative approach for the conditions of this study. It is probably true that nearly all of the plutonium on the soil is in the respirable size range (*2*), and we have probably measured nearly all of the plutonium on the surface of the soil. A minor adjustment in threshold parameters as proposed would result in a small change in the weight basis for expressing concentration. We believe that this concentration difference is trivial, particularly when compared with the difference between the weight of the respirable fraction (following our definition), which we used, and the weight of the whole soil collected to arbitrary depths, which it has been the practice to use in the past.

Employing a vacuum device for sample collection may be a useful modification of our method, if the device is equipped to avoid loss of submicrometer particles. The respirable fraction may be separated by any procedure capable of performing the separation effectively. However, the separation procedure that we utilized to isolate the respirable fraction will probably yield more reproducible results (*3*).

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