MEASUREMENTS OF THE AEROSOL SIZE DISTRIBUTION OF URANIUM AND THORIUM AND ITS IMPLICATIONS FOR DOSE CALCULATIONS

R. Z. Leifer[1], E. M. Jacob[2], S. F. Marschke[3] and D. M. Pranitis[4]

Abstract: A rotating drum impactor was co-located with a high volume air sampler for ~ 1 y at the fence line of the U. S. Department of Energy's Fernald Environmental Management Project site. Data on the size distribution of uranium and thorium bearing atmospheric aerosols were obtained and used to compute inhalation dose using several different models. During most of the year, the mass of ^{238}U above 4.3 μm exceeded 80% of the total uranium mass from all particulates. During any sampling period the size distribution was bimodal. Thorium concentrations were comparable to the uranium concentrations during the late spring and summer period and decreased to ~25% of the ^{238}U concentration in the late summer. The seasonal average of the activity median aerodynamic diameter (AMAD), based on the impactor data was approximately 6.5 μm. The current calculational method used to demonstrate compliance with regulations assumes that the airborne particulates are characterized by an activity median diameter of 1 μm. This assumption results in an overestimation of the inhalation dose to offsite individuals by as much as a factor of seven relative to values derived using the latest ICRP 66 lung model with more appropriate particle sizes. Further evaluation of the size distribution for each radionuclide would substantially improve the dose estimates.

Introduction: To better quantify the air component of the dose of an offsite individual in the U.S. Department of Energy's (DOE) Fernald Environmental Management Project (FEMP) area, the Environmental Measurements Laboratory (EML) set up a Davis rotating universal size-cut monitoring sampler (DRUM)(University of California, Davis, CA). The sampler is an inertial aerosol impactor that separates and collects particulate samples in a number of size-segregated fractions (50% aerodynamic cut diameters of 8.54 μm, 4.26 μm, 2.12 μm, 1.15 μm, 0.56 μm, 0.34 μm, 0.24 μm, and 0.069 μm, respectively). The inlet rain hat removes particles above 15 μm. The DRUM impactor is designed to run unattended for 4 weeks. The samples are deposited on a Mylar™ (Du Pont, Wilmington, DE) strip that has been coated with a 2% solution of Apiezon grease (Apiezon-L, Apiezon Products, London, U.K.) Every 4 weeks EML shipped a set of eight coated drums to FEMP for installation in the impactor to coordinate with the FEMP's changing of high volume filter samplers. The Mylar™ foils are then removed from the drums and divided into 2-week segments, dissolved in nitric acid using microwave heating, and analyzed using an inductively-coupled plasma/mass spectrometer (ICP/MS) for quantification (Pranitis 1999) of ^{238}U. Quality assurance (QA) samples, including blanks and spikes were included with the samples. In May 1999, it was decided to add ^{232}Th to the analyses to explore whether we could resolve thorium from the small samples collected.

Results and Discussion: Using the DRUM data and the filter data from the high volume samplers co-located with the DRUM impactor, a series of plots were generated to characterize the time history of the data set as well as size distribution plots. The ^{238}U above 15 μm accounts for more than 73% of the total mass concentration. The mass fraction > 4.3 μm exceeds 87% for

[1]Physical Scientist, Environmental Measurements Laboratory, U.S. Department of Energy, 201 Varick Street, 5th Floor, New York, NY 10014-4811, Ph 212-620-3626, Fx 212-620-3600, Rleifer@eml.doe.gov. (corresponding author).
[2]Computer Specialist, Environmental Measurements Laboratory, U.S. Department of Energy, 201 Varick Street, 5th Floor, New York, NY 10014-4811, Ph 212-620-3626, Fx 212-620-3600, Jacob@eml.doe.gov
[3]Health Physicist, Gemini Consulting Company,44 Washington Ave, New York, NY 10014
[4]Chemist, David Pranitis, Avon, Inc., Avon Place, Suffern, NY 10901

the whole data set. The largest concentrations of ^{238}U occurred during the late summer period. The winter months showed the lowest values and may be associated with inactivity during this period. Figure 1 provides a detailed picture of the time history of the ^{238}U concentration below 100 µm and 15 µm respectively obtained from the DRUM impactor. It is evident from these data that using the total suspended particulate (TSP) concentration to calculate the dose to off-site individuals, under the assumption of an AMAD of 1µm, will result in a gross overestimation in the amount of material deposited in the lung, and, hence, in the inhalation dose.

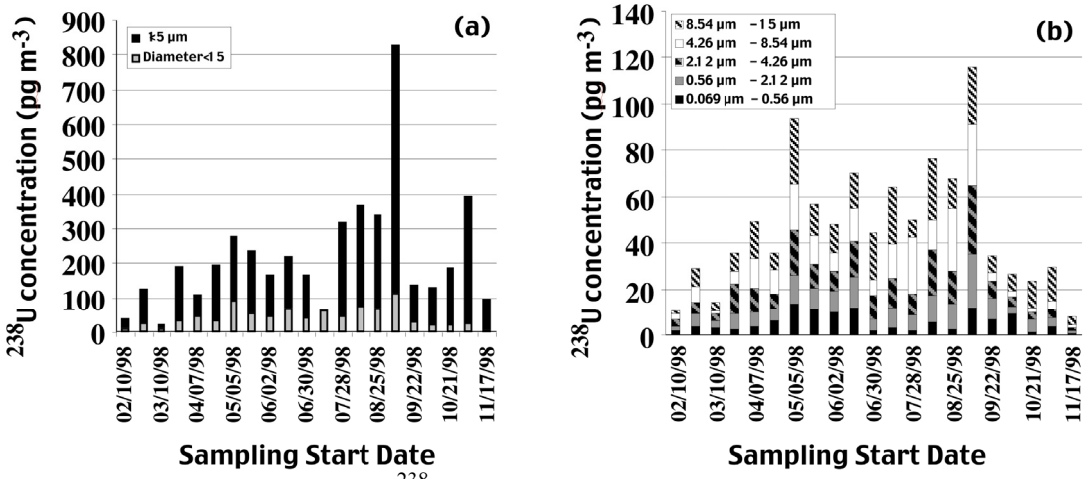

Figure 1. The size dependent ^{238}U concentration as a function of sampling period for aerosol sizes less than: a) 100 µm; b) 15 µm.

Activity Median Aerodynamic Diameter: To extract an activity median aerodynamic diameter (AMAD) from our uranium and thorium data requires a detailed knowledge of the size distribution from 0.06 µm to 100 µm. Although the DRUM impactor provides the necessary size

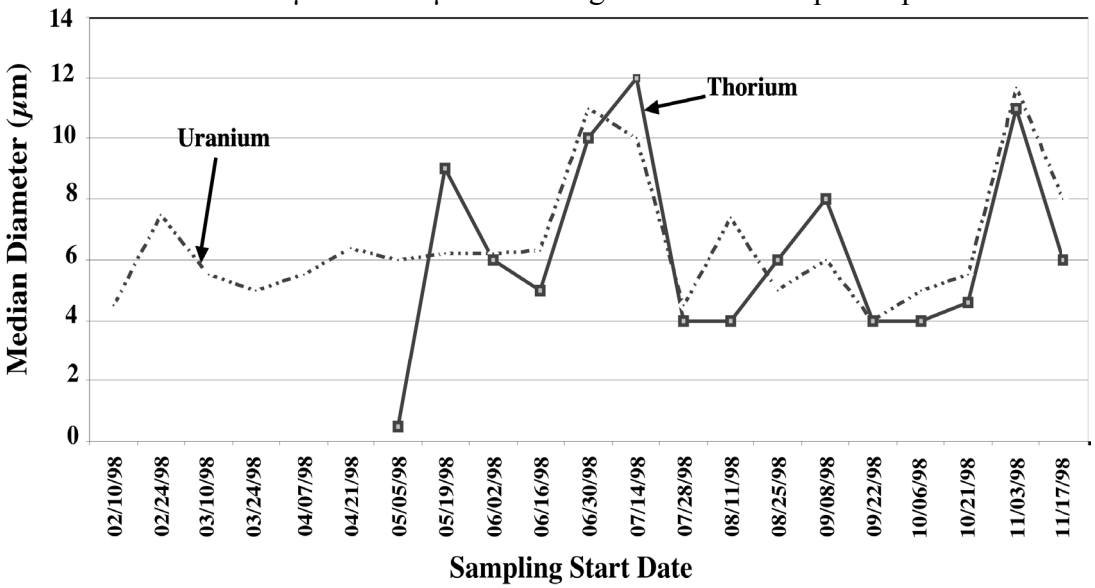

Figure 2. The seasonal AMAD of uranium and thorium as a function of sampling time period .

data below 15 µm, the data from the high volume or TSP sampler does not. Because of the uncertainty in the upper size value of the TSP and lack of size data between 15 µm and 100 µm, only the DRUM data are used to calculate the AMAD. These calculated values represent a lower limit to the actual AMAD. The calculations are based on using the cumulative distribution format. The mass fraction, was calcuated for the size intervals of the DRUM impactor. A best estimate of the diameter for both uranium and thorium was obtained from the 50% percentile value. The resulting seasonal AMAD are plotted as a function of sampling time period (Figure 2). The analysis of the samples for thorium did not start until the 5 May sampling period. Except for the May sampling periods, the two nuclides track exceptionally well. The average AMAD for the period from 5 May through 17 November was 6.9 µm and 6.3 µm for uranium and thorium, respectively. The average for all seasons for uranium was 6.5 µm. The June and November periods show elevated AMAD reaching a maximum of 12 µm. If the high volume data were included in these calculations the AMAD would be larger.

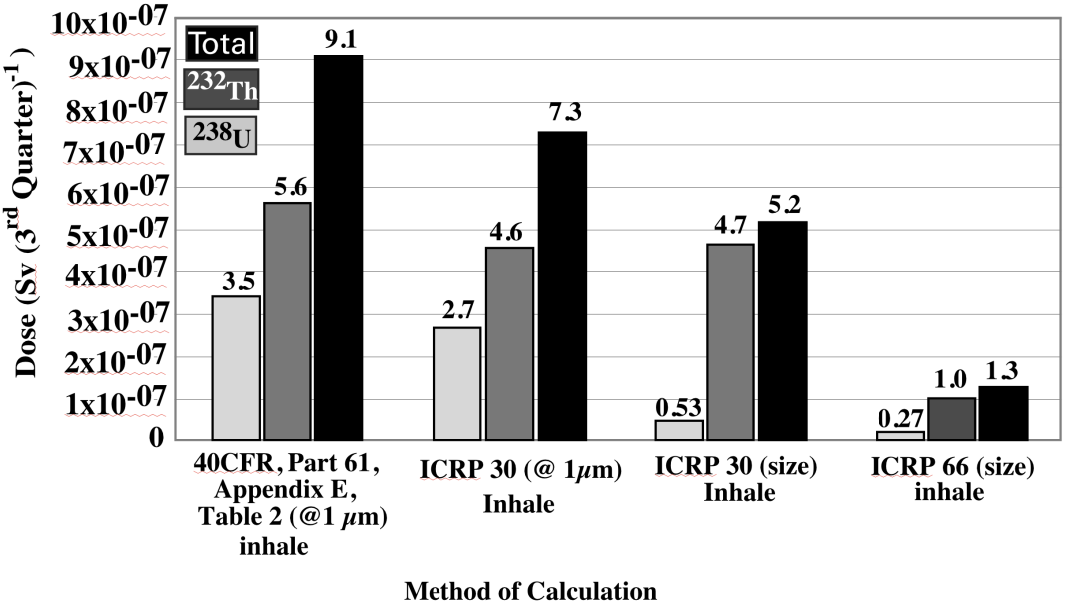

Figure 3. The ^{238}U and ^{232}Th air compartment dose caculations for the 3rd quarter of 1998, using three different model calculations, 40CFR, ICRP 30, and ICRP 66.

Air Compartment, Dose Calculation: Using the clearance time dose factor that is "largest" for a given size of particles and the size dependent airborne concentrations measured at Fernald, the 3rd quarter annual effective dose equivalent (EDE) based on the ICRP 66 respiratory tract model was calculated to be 2.7×10^{-8} Sv from ^{238}U, and 1.0×10^{-7} Sv from ^{232}Th, for a total of 1.3×10^{-7} Sv or ~14% of the FEMP value of 9.1×10^{-7} Sv (see Figure. 3). Included in the figure but not discussed here are calculations from the ICRP 30 model (1 µm and size dependent) and from 40CFR, p. 61 Appendix E, Table 2. The air compartment, dose calculation overestimates the dose of an off-site individual by a large factor because the calculations use data obtained from high volume samplers collecting more than 70% of the uranium mass above 15 µm.
For more detailed information on thorium and uranium size distributions and dose calculations see Leifer et al. (2000).

Reference: Leifer, R. Z. E. M. Jacob, S.F. Marschke, D.M. Pranitis and H-R Kristina Jaw ^{238}U and ^{232}Th (2000) *Dose Calculations and Size Distribution Measurements of Atmospheric Aerosols at Fernald*, Ohio, U.S. DOE Report EML-606.
Pranitis, D. (1999) Private Communication, Avon, Inc., Avon Place, Suffern, NY.