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²³⁸U and ²³²Th Dose Calculations and Size Distribution Measurements of Atmospheric Aerosols at Fernald, Ohio



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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 bearing atmospheric aerosols from 0.065 µm to 100 µm in diameter were obtained and used to compute dose using several different models. During most of the year, the mass of ²³⁸U above 15 µm exceeded 70% of the total uranium mass from all particulates. Above 4.3 µm, the ²³⁸U mass exceeded 80% of the total uranium mass from all particulates. During any sampling period the size distribution was bimodal. In the winter/spring period, the modes appeared at 0.29 µm and 3.2 µm. During the summer period, the lower mode shifted up to $\sim 0.45 \ \mu m$. In the fall/winter, the upper mode shifted to $\sim 1.7 \ \mu m$, while the lower mode stayed at 0.45 µm. These differences reflect the changes in site activities. Thorium concentrations were comparable to the uranium concentrations during the late spring and summer period and decreased to ~25% of the ²³⁸U concentration in the late summer. The thorium size distribution trend also differed from the uranium trend. 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 overestimate of the dose to offsite receptors 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.

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Introduction

The U. S. Department of Energy's (DOE) Fernald Environmental Management Project (FEMP), located 18 miles northwest of Cincinnati, OH, was constructed in the early 1950's to produce uranium metal products for use by the government. Activities at the site, formerly called the "Feed Materials Production Center," were suspended in July 1989. As part of the national environmental restoration program, the FEMP site managers needed to implement an environmental monitoring plan to characterize the radionuclide emissions at the site and how they impact the population (DOE 1988). The National Emissions Standard for Hazardous Air Pollutants (NESHAP 1979) requires an annual assessment of the dose to offsite receptors due to radionuclide emissions. There are additional guidelines for detection limits, and site/facility boundary definitions (DOE 1990,1992). The maximum allowable dose from airborne emissions, excluding radon, cannot exceed 10 mrem y-1 (NESHAP 1979). There are more than 10 additional regulatory drivers having air monitoring implications and they are summarized in a FEMP report (1996).

The original environmental monitoring plan established a program using a high volume filter sampler at a few sites and combined it with air dispersion modeling to calculate the dose to offsite receptors. In order to best monitor the emissions from several large area sources that will be active during the full scale remediation of the site, the FEMP implemented a new monitoring plan that relied on fence line environmental measurements in order to estimate the dose for the purpose of NESHAP compliance. Fence line environmental measurements were preferred because it is difficult to accurately characterize emissions from area sources using computer models.

The FEMP monitoring stations are used to characterize air concentrations at the site fence line (16 locations) and at background locations (two locations). The air sampling instruments, known as high volume samplers, provide continuous sampling of airborne particulates during a 2-week sampling period. A portion of the 2-week sample is used to monitor fence line total uranium concentrations. Quarterly composites of the 2-week filters are analyzed for radionuclides identified as the major contributors to dose from airborne emissions (²³⁴U, ²³⁵U, ²³⁸U, ²²⁸Th, ²³⁰Th, ²³²Th, and ²²⁶Ra). In 1997, biweekly uranium concentrations at the fence line ranged from 0 to 1.8 ng m⁻³. This high value corresponds to ~ 1% of the DOE Derived Concentration Guide.

Dorrian (1997) provided a detailed discussion and review of the available radionuclide size distribution data published in the open literature. He found only 21 papers containing 181 measurements of the activity median aerodynamic diameter (AMAD). He also indicated that for

resuspension processes using an AMAD of 1 μ m is inappropriate. During resuspension processes, the AMAD shifts from 1 Fm to a larger value, close to the 6 Fm for Dorian's (1997) analysis of the available data. From the results of his survey Dorrian reported, "The need for characterization of activity size distributions of environmental aerosols to enable realistic doses to be calculated for members of the public is evident."

Current NESHAP dose calculation methods used at FEMP do not account for the effects of particle size when estimating dose. All of the airborne uranium measured at the high volume monitors is assumed to contribute to dose. To better quantify the air component of the dose of an offsite individual in the FEMP area, the Environmental Measurements Laboratory (EML) set up specialized aerosol sampling equipment at the site.

Experimental Design

To help the site managers better understand the dose to the offsite receptor, EML set up a Davis Rotating Universal Size-cut Monitoring Sampler (DRUM; a cascade impactor on a loan from Dr. Thomas Cahill, University of California, Davis) to characterize the size distribution of the atmospheric aerosols at one of the FEMP fence line stations. The cascade impactor is an inertial aerosol sampler that separates and collects particulate samples in a number of sizesegregated fractions for the characterization of the aerodynamic particle size distribution. All diameter values used in this report refer to aerodynamic diameters unless otherwise stated to be physical diameters. The site chosen for the cascade impactor was AMS-9C where a high volume sampler was co-located (see Figure 1). The impactor (shown in Figure 2) fractionates the sampled aerosol, by particle size, into successively smaller sizes. Each size has a characteristic aerodynamic diameter (the diameter of a sphere of unit density having the same terminal settling velocity in air as the particle of interest) cut size corresponding to a collection efficiency of 50%. For an infinitely steep collection efficiency, all particles above the aerodynamic cut diameter would be removed from the air stream and all particles below would pass to the next stage. The ideal impactor does not exist and all efficiency curves have some slope, allowing some cross over of sizes between stages. The Davis impactor efficiency curves are quite steep providing good particle separation. The Davis impactor contains eight stages with 50% aerodynamic cut diameters of $8.54 \mu m$, $4.26 \mu m$, $2.12 \mu m$, $1.15 \mu m$, $0.56 \mu m$, $0.34 \mu m$, $0.24 \mu m$, and $0.069 \mu m$, respectively. The inlet rain hat removes particles above 15 µm. These cut diameters were obtained using monodispersed fluorescent aerosols, and the resulting experimental data were in good agreement with theoretical calculations and are fully discussed in Raabe (1988). The impactor was operated at 1.1 L min⁻¹. Ambient pressure and temperatures were available from a meteorological tower at the Fernald site.

In support of the use of impactors for air sampling, we refer to Marple and Willike (1976), who report that "impactor stages, which are properly designed and operated, will provide sharp classification between the particles collected and those which are not." Furthermore, a recent study, using the same DRUM impactor as EML used for this study, showed the DRUM's suitability to characterize the time history of atmospheric aerosol size distributions (Pitchford and Green 1997).

Instrument Operation

The DRUM impactor is designed to run unattended for 4 weeks. The samples are deposited on a MylarTM (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 a set of eight coated drums arrived at FEMP for installation in the impactor. The time was coordinated to match FEMP's changing of their filter samplers. The completed set of drums was returned to EML for sample preparation and analysis.

Sample Preparation and analysis

The MylarTM foils were removed from the drums in a Class 100 laminar flow hood and divided into 2-week segments. Each segment was dissolved in nitric acid using microwave heating. The digestate, in 4<u>M</u> nitric acid, is pumped through a column packed with Eichrom TRU Resin (Eichrom Industries, Inc., 8205 Cass Ave., Suite 107, Darien IL 60561) that retains dissolved uranium. After a fixed collection time, the uranium is back-flushed through an ultrasonic nebulizer into an inductively-coupled plasma/mass spectrometer (ICP/MS) for quantification (Pranitis 1999) of ²³⁸U. Quality assurance (QA) samples, including blanks and spikes were included with the samples. Samples from February, March and April were analyzed at EML.

All subsequent samples were sent to Key Laboratories, Inc. (Grand Junction, CO 81505), which was under contract to EML. The procedures used by Key Laboratories, Inc. (standard operating procedure # 38, Uranium and Thorium on MylarTM, Inductively Coupled Plasma - Mass Spectroscopy) were agreed to by EML and Key. Blind spikes and blanks were included in each shipment. In May 1999, it was decided to add ²³²Th to the analyses to explore whether we could resolve thorium from the small samples collected.

Results

Table 1 contains the results of the analyses of more than 160 impactor samples by EML and Key Laboratories, Inc. Key analyzed more than three fourths of the samples. The table includes a unique EML sample number, the laboratory analyzing the sample, the collection stage size range, sample length, and starting and stopping date and time, ²³⁸U and ²³²Th analyzed mass per foil section, and the analytical error associated with the analyses. The small variations in the length reflect variations in drum rotation speed, as well as the additional blank MylarTM strips included in the sample. Samples were sent to Key in two separate batches. Initially, 40 samples were sent to Key to evaluate the capability of the laboratory to handle our samples. The results received were encouraging and a formal contract was drawn with Key for analyzing an additional 106 samples.

The uranium mass per sample varied from a low of 2.1 pg cm⁻¹ and 1.7 pg cm⁻¹ per foil for 238 U and 232 Th, respectively, to a high of 130 pg cm⁻¹ and 69 pg cm⁻¹ for 238 U and 232 Th, respectively. One sample showed a 232 Th value > 250 pg cm⁻¹ per foil. No analytical problems were found with this sample.

Quality Assurance Data

Table 2 contains the QA data for all of the samples analyzed. We sent approximately 10% of the samples as blanks and spikes to Key laboratories, Inc. The method detection limit (MDL) for the ²³⁸U is 0.77 pg cm⁻¹, while the MDL for ²³²Th is 1.15 pg cm⁻¹. The difference between EML and Key (batch 1) in average blank values was negligible (see Table 3). Key could not account for the increase in the blank values between the two shipments. Calculations of the final concentrations are based on the respective blank values for each shipment. ²³²Th was not analyzed at EML. Blind samples spiked with 100 pg of ²³⁸U were submitted to Key. The spike was placed on blank foils and could not be discriminated from regular samples. There is an average 9% analytical difference between the laboratories in the measurement of ²³⁸U.

DISCUSSION OF RESULTS

URANIUM DATA

The data from Table 1 was converted to concentration values (Table 4) using a volume corrected to 25°C and 1 atmosphere pressure. The overall error associated (including, analytical

sample volume and sample volume errors) with the concentration data is estimated to average better than 11%. Published impactor losses are < 10% (Raabe et al. 1988).

Using the data from Table 4 and the filter data from the high volume samplers (Table 5) colocated 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. To obtain the ²³⁸U concentration below 100 μm (Figure 3), the sums of all DRUM stages were subtracted from the total ²³⁸U reported by FEMP. The ²³⁸U above 15 μ m accounts for more than 73% \pm 4% of the total mass concentration. The mass fraction > 4.3 μ m exceeds 87% \pm 4% for the whole data set (Figure 4). The average percent concentration of ²³⁸U for the total data set is included in Figure 5. Only during the 2-week period starting on July 14, 1998 was the size distribution dominated by particles below 15 um. The largest concentrations of ²³⁸U occurred during the late summer period. The winter months showed the lowest values and may be associated with inactivity during this period. Still seen in the winter months are significant concentrations of 238 U in the $> 15 \mu m$ size range, which may be associated with resuspension processes especially if the ground is not snow covered. Figure 4 shows a comparison of the percent ²³⁸U above 4.26 µm and 15 µm. During most of the year, the mass of ²³⁸U above 15 µm exceeded 70% of the total suspended particulate (TSP) values. Above 4.26 µm the ²³⁸U mass exceeded 80% of the total TSP values. Figure 6 provides a detailed picture of the time history of the ²³⁸U concentration below 15 µm obtained from the DRUM impactor. For this size range, no more than 13% (Figure 5) of the ²³⁸U mass is below 4.26 µm. It is evident from these data that using TSP to calculate the dose to off-site individuals, under the assumption of an AMAD of 1 µm, will result in a gross overestimate in the amount of material deposited in the lung, and, hence, in the doses. In the Inhalation Dose Calculation section, and specifically the sections on dose calculations comparisons using measured size distributions, we discuss the reasons for this gross over estimate. That is, the effective dose conversion factors are very size dependent.

The results of plotting the data as a normalized size distribution are shown in Figure 7a,b. The plots are normalized using the TSP values to allow easier comparison when plotting distributions. The mean value of each size interval is used for plotting the distributions. The upper interval is obtained by subtracting the TSP values from the total impaction values and the resulting data plotted at 57.5 μ m (midpoint between 15 mm and 100 μ m). The upper limit of the TSP high volume air sampler is taken to be 100 μ m (EPA 1999). The accumulated mass on the two-week filter sample for the July 14, 1998 sampling period was extremely low and close to the blank filter mass (see Figure 7a). Figure 7a contains the combined DRUM and TSP data. The mass above 15 μ m dominates the normalized size distribution and tends to suppress the visualization of the lower modes. Figure 7b provides a picture of the size distribution below 15 μ m. The upper mode is not resolved because only one size range was measured above 15 μ m (Figure 7a).

The seasonal dependence of the size distribution is obtained by separating the data into a winter/spring period (Figure 8a), summer period (Figure 8b), and fall/winter period (Figure 8c). In the winter/spring period, the fine particulate mode peaks at \sim .29 μm and the upper coarse mode is centered at \sim 3.2 μm . During the summer period the lower mode shifted to \sim 0.45 μm , while the upper mode remained the same. Two time periods, the weeks of June 2, 1998 and August 25, 1998, showed a disappearance of the lower mode and a shifting of the upper mode to atmospheric size distribution sizes above 6 μm . The fall/winter period shows a reduction in the upper mode that is consistent with site inactivity. Frozen ground and snow cover reduced resuspension which affects the atmospheric size distribution. The shifting modes in the size distribution with time emphasized the need to calculate the dose using realistic size distribution data.

THORIUM DATA

Figures 9 to 11 contain ²³²Th data plotted in formats similar to the uranium plots. Since only uranium was measured in a 2-week sampling period, no TSP comparison could be made with our thorium measurements. The trend in the thorium data (Figure 9) is quite different from the earlier discussed uranium trends. The thorium concentration decreased from May to December 1998, while the uranium concentration, during the same period, stayed elevated until the early fall. At the present time, there is no clear explanation of this difference.

The size distribution data for thorium (Figure 10) as well as for uranium show a number of discrete peaks. Except for the June 16, 1998 and October 21, 1998 data, the diameters at which the peaks appear for both radionuclides are the same. Also, the thorium and uranium size distributions differ. These differences reflect changing of the source strengths as well as site activities.

The fraction of uranium to thorium (Figure 12) reaches a maximum during the summer/early fall period (~607%), while during the spring the uranium is comparable to the thorium. The uranium to thorium ratio decreases in the late fall. Since no upwind data exists, it is possible that during the spring we are seeing background thorium concentrations.

INHALATION DOSE CALCULATION

The U.S. Nuclear Regulatory Commission (NRC) in 10 CFR Part 20 and the U.S. Environmental Protection Agency (EPA) in 40 CFR Part 61 (NESHAP 1979) base their exposure limits on the dosimetric models of the International Commission on Radiation Protection (ICRP) Publication 30 (1981). FEMP uses these exposure limits to calculate the annual average dose to an off-site individual at a specific location. These calculations are very conservative and not only include the dose from inhalation but also the dose from ingestion of food that is assumed to be grown by the individual at the site.

The EPA has published inhalation dose conversion factors, based on ICRP 30 (1981), in Federal Guidance Report (FGR) No. 11 (EPA 1988), which can be used to demonstrate compliance with NRC and EPA regulations. These inhalation dose factors are based on an activity median aerodynamic diameter (AMAD) of 1 μm, and on lung clearance times on the order of days, weeks, and years. The clearance time is a function of the chemical species of the radionuclide of interest. For example, FGR No. 11 states that uranium radionuclides that have the chemical form UO₂ or U₃O₈ have a clearance time of years, while those with the chemical forms UO₃, UF₄ and UCl₄ have a clearance time of weeks, and those with the chemical forms UF₆, UO₂F₂ and UO₂(NO₃)₂ have a clearance time of days. Likewise FGR No. 11 states that thorium radionuclides that are oxides or hydroxides have a clearance time of years, while all other chemical forms of thorium have a clearance time of weeks. When the chemical form of the radionuclides is unknown, it is usually conservatively assumed that the largest inhalation dose factor applies.

INHALATION DOSE FACTORS FOR ²³⁸U AND ²³²TH

The FGR No. 11 inhalation dose factors for 238 U are 6.62×10^{-7} , 1.90×10^{-4} and 3.20×10^{-5} Sv Bq⁻¹ for clearance times of days, weeks and years, respectively, while the 232 Th FGR No. 11 factors are 4.43×10^{-4} and 3.11×10^{-4} Sv Bq⁻¹ for clearance times of weeks and years, respectively. These dose factors correspond to a log-normal distribution of the aerodynamic diameter characterized by a median of 1 μ m and a sigma of 2.5μ m.

DOSE CALCULATION COMPARISON (3RD QUARTER): ICRP 30 AT 1 mm

Intakes based on the 3rd quarter 238 U and 232 Th airborne concentrations (232 Th concentrations are not available for the whole year) measured at Fernald and the largest FGR No. 11 inhalation dose factors gives an effective dose equivalent (EDE88)¹ of 2.7 x $^{10^{-7}}$ Sv from 238 U and $^{4.6}$ x $^{10^{-7}}$ Sv from 232 Th, for a total of 7.3 x $^{10^{-7}}$ Sv. For this same period, FEMP obtained 9.1 x $^{10^{-7}}$ Sv or a 20% difference.

DOSE CALCULATION COMPARISON (3RD QUARTER): ICRP 30 USING THE MEASURED SIZE DISTRIBUTION

In reality, the 238 U and 232 Th concentrations were measured at Fernald for various particle sizes, ranging from 0.069 μ m to 15 μ m. The computer program DFINT (Eckerman 1994) was used to calculate the size dependent inhalation dose conversion factors shown as the dashed line

¹Dose equivalent is the product of the absorbed dose, the quality factor and any other modifying factors, while the EDE is the sum over specified tissues of the products of the dose equivalent in a tissue or organ and is the weighting factor for that tissue.

in Figure 13 for 238 U and in Figure 14 for 232 Th, based on the ICRP 30 dosimetric models. Using the clearance time with the largest dose factor for a given size of particles and the size dependent airborne concentrations measured at Fernald, the 3rd quarter EDE was calculated to be 5.3 x $^{10^{-8}}$ Sv (3rd quarter) $^{-1}$ from 238 U and 4.7 x $^{10^{-7}}$ Sv (3rd quarter) $^{-1}$ from 232 Th, for a total of 5.2 x $^{10^{-7}}$ Sv (3rd quarter) $^{-1}$ or 57% of the FEMP value of 9.1 x $^{10^{-7}}$ Sv (3rd quarter) $^{-1}$

ICRP Publication 66

In 1990, the ICRP (1990) revised the tissue weighting factors used to calculate the effective dose (ICRP Publication 61), and in 1994 they replaced the ICRP 30 inhalation dosimetric model with a new respiratory tract model, which is presented in ICRP Publication 66. The respiratory tract model revision was motivated by the availability of an increased knowledge of the anatomy and physiology of the respiratory tract and of the deposition, clearance and biological effects of inhaled radioactive particles, and by greatly expanded dosimetry requirements. These revisions to the inhalation dosimetric model have been incorporated into the LUDEP (1988) computer program (National Radiological Protection Board). LUDEP was used to calculate the size dependent inhalation dose conversion factors shown as solid lines in Figure 13 for ²³⁸U and in Figure 14 for ²³²Th. (Note: While FGR No. 11 does not present a dose factor for the "day's" clearance time for ²³²Th, dose factors for "fast" clearance times were calculated with LUDEP based on the fact that the IAEA's RasaNet Web Site provided the dose factor for 1 µm AMAD and a "fast" clearance time.) Additionally, the dosimetric models can distinguish between radionuclides that are bone volume seekers and those that are bone surface seekers. From published ICRP 66-based inhalation dose factors, it was determined that a split of 78% bone volume seekers and 22% bone surface seekers should be used for both ²³⁸U and ²³²Th. Table 7 summarizes the effective dose coefficients for the inhalation uptakes by an adult.

DOSE CALCULATION COMPARISON: ICRP 66 USING THE MEASURED SIZE DISTRIBUTION

Again, 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 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 15).

ANNUAL AVERAGE DOSE CALCULATION COMPARISON: 238U

Using the size dependent EDE Factor (Figure 13) and the annual average size distribution (Figure 16) of 238 U the annual average EDE is 7.2×10^{-8} Sv, which is 8% of the FEMP calculated annual average EDE of 9.0×10^{-7} Sv (see Figure 17).

AVERAGE YEARLY DOSE USING ICRP 66 AND THE MEASURED SIZE DISTRIBUTION

Shown in Figure 18 is the size dependence of the annual average dose for ²³⁸U. More than 74% of the dose is accounted for in the 4.26 to 100 µm region (Figure 18). It is in this region that the dose calculations need more work as most of the assumptions we used are very conservative. That is, we used the largest clearance time dose factor for the mean of the size interval.

Conclusions

At the fence line of the FEMP site, a rotating drum impactor was co-located with a high volume sampler for ~1 y. Data on the size distribution of uranium bearing atmospheric aerosols from 0.065 μ m to 100 μ m in diameter were obtained. During most of the year, the mass of 238 U above 15 μ m exceeded 70% of the total TSP values. Above 4.3 μ m the 238 U mass exceeded 80% of the total TSP values. Only during the week of July 14, 1998 was the contribution from the size range above 15 μ m negligible. During any sampling period two modes appeared. In the winter/spring period, the modes appeared at 0.29 μ m and 3.2 μ m. During the summer period, the lower mode shifted up to ~0.45 μ m. During the winter/fall, the upper mode shifted down to ~1.7 mm. These changes reflect the changes in activities at the site. Thorium concentrations were comparable to the uranium concentration during the late spring and summer period and decreased to ~25% in the late summer. The thorium signature also differed from the uranium signature.

Using the 238 U averaged annual size distribution data and the ICRP 66 respiratory tract model, we showed that the annual EDE for 238 U was 7.2 x 10^{-8} Sv compared to the FEMP calculated annual EDE of 9.0 x 10^{-7} Sv or a 92% decrease in the EDE. Although we only had thorium data overlapping one sampling quarter (3rd quarter), we showed that when applying a size distribution to the calculations, the 232 Th 3rd quarter EDE was 1.0×10^{-7} Sv compared to the FEMP calculated EDE of 5.6×10^{-7} Sv or an 82% decrease in the EDE. Combining the 238 U and 232 Th data produces an EDE of 1.3×10^{-7} Sv compared to the FEMP calculated EDE of 9.1×10^{-7} Sv or an 86% decrease in EDE. These data are summarized in Table 6, where the reduction in calculated dose is evident when incorporating the newer ICRP models with size information.

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 μ m.

Finally, our measurements support the results of Dorrian (1997), that is, activity size distribution data are needed in order to improve the dose calculations to the public.

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TABLE 1SAMPLING DATA

Sample No.	Analysis Laboratory	Size Cut (µm)	Sample Length (cm)	Starting Date	Starting Time	Ending Date	Ending Time	²³⁸ U (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)	²³² Th (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)
0953	EML	8.54 - 15	5.6	02/10/98	12:00 pm	02/24/98	12:00 pm	55	5.5	-	
0954	EML	4.26 - 8.54	5.6	02/10/98	12:00 pm	02/24/98	12:00 pm	78	7.8	-	
0955	EML	2.12 - 4.26	5.6	02/10/98	12:00 pm	02/24/98	12:00 pm	100	10.0	-	
0956	EML	1.15 - 2.12	5.6	02/10/98	12:00 pm	02/24/98	12:00 pm	64	6.4	-	
0957	EML	0.56 - 1.15	5.2	02/10/98	12:00 pm	02/24/98	12:00 pm	30	3.0	-	
0958	EML	0.34 - 0.56	5.2	02/10/98	12:00 pm	02/24/98	12:00 pm	29	2.9	-	
0959	EML	0.24 - 0.34	5.6	02/10/98	12:00 pm	02/24/98	12:00 pm	53	5.3	-	
0960	EML	0.069 - 0.24	5.2	02/10/98	12:00 pm	02/24/98	12:00 pm	30	3.0	-	
0961	EML	8.54 - 15	5.6	02/24/98	12:00 pm	03/10/98	12:00 pm	202	20.2	-	
0962	EML	4.26 - 8.54	5.6	02/24/98	12:00 pm	03/10/98	12:00 pm	190	19.0	-	
0963	EML	2.12 - 4.26	5.6	02/24/98	12:00 pm	03/10/98	12:00 pm	140	14.0	-	
0964	EML	1.15 - 2.12	5.6	02/24/98	12:00 pm	03/10/98	12:00 pm	97	9.7	-	
0965	EML	0.56 - 1.15	5.2	02/24/98	12:00 pm	03/10/98	12:00 pm	38	3.8	-	
0966	EML	0.34 - 0.56	5.2	02/24/98	12:00 pm	03/10/98	12:00 pm	57	5.7	-	
0967	EML	0.24 - 0.34	5.6	02/24/98	12:00 pm	03/10/98	12:00 pm	45	4.5	-	
0968	EML	0.069 - 0.24	5.2	02/24/98	12:00 pm	03/10/98	12:00 pm	60	6.0	-	
0969	EML	8.54 - 15	5.2	03/10/98	12:00 pm	03/24/98	12:00 pm	108	10.8	-	
0970	EML	4.26 - 8.54	5.2	03/10/98	12:00 pm	03/24/98	12:00 pm	57	5.7	-	
0971	EML	2.12 - 4.26	5.2	03/10/98	12:00 pm	03/24/98	12:00 pm	94	9.4	-	
0972	EML	1.15 - 2.12	5.2	03/10/98	12:00 pm	03/24/98	12:00 pm	55	5.5	-	
0973	EML	0.56 - 1.15	5.2	03/10/98	12:00 pm	03/24/98	12:00 pm	42	4.2	-	
0974	EML	0.34 - 0.56	5.2	03/10/98	12:00 pm	03/24/98	12:00 pm	22	2.2	-	
0975	EML	0.24 - 0.34	5.2	03/10/98	12:00 pm	03/24/98	12:00 pm	11	1.1	-	
0976	EML	0.069 - 0.24	5.2	03/10/98	12:00 pm	03/24/98	12:00 pm	82	8.2	-	
0977	EML	8.54 - 15	5.2	03/24/98	12:00 pm	04/07/98	12:00 pm	201	20.1	-	

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Sample No.	Analysis Laboratory	Size Cut (µm)	Sample Length (cm)	Starting Date	Starting Time	Ending Date	Ending Time	²³⁸ U (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)	²³² Th (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)
0978	EML	4.26 - 8.54	5.2	03/24/98	12:00 pm	04/07/98	12:00 pm	144	14.4	-	
0979	EML	2.12 - 4.26	5.2	03/24/98	12:00 pm	04/07/98	12:00 pm	317	31.7	-	
0980	EML	1.15 - 2.12	5.2	03/24/98	12:00 pm	04/07/98	12:00 pm	129	12.9	-	
0981	EML	0.56 - 1.15	5.2	03/24/98	12:00 pm	04/07/98	12:00 pm	74	7.4	-	
0982	EML	0.34 - 0.56	5.2	03/24/98	12:00 pm	04/07/98	12:00 pm	37	3.7	-	
0983	EML	0.24 - 0.34	5.2	03/24/98	12:00 pm	04/07/98	12:00 pm	44	4.4	-	
0984	EML	0.069 - 0.24	5.2	03/24/98	12:00 pm	04/07/98	12:00 pm	36	3.6	-	
0985	EML	8.54 - 15	5.2	04/07/98	12:00 pm	04/21/98	12:00 pm	396	39.6	-	
0986	EML	4.26 - 8.54	5.2	04/07/98	12:00 pm	04/21/98	12:00 pm	308	30.8	-	
0987	EML	2.12 - 4.26	5.2	04/07/98	12:00 pm	04/21/98	12:00 pm	272	27.2	-	
0988	EML	1.15 - 2.12	5.2	04/07/98	12:00 pm	04/21/98	12:00 pm	107	10.7	-	
0989	EML	0.56 - 1.15	5.2	04/07/98	12:00 pm	04/21/98	12:00 pm	67	6.7	-	
0990	EML	0.34 - 0.56	5.2	04/07/98	12:00 pm	04/21/98	12:00 pm	62	6.2	-	
0991	EML	0.24 - 0.34	5.2	04/07/98	12:00 pm	04/21/98	12:00 pm	60	6.0	-	
0992	EML	0.069 - 0.24	5.2	04/07/98	12:00 pm	04/21/98	12:00 pm	35	3.5	-	
0993	EML	8.54 - 15	5.2	04/21/98	12:00 pm	05/05/98	12:00 pm	189	18.9	-	
0994	EML	4.26 - 8.54	5.2	04/21/98	12:00 pm	05/05/98	12:00 pm	256	25.6	-	
0995	EML	2.12 - 4.26	5.2	04/21/98	12:00 pm	05/05/98	12:00 pm	181	18.1	-	
0996	EML	1.15 - 2.12	5.2	04/21/98	12:00 pm	05/05/98	12:00 pm	91	9.1	-	
0997	EML	0.56 - 1.15	5.2	04/21/98	12:00 pm	05/05/98	12:00 pm	55	5.5	-	
0998	EML	0.34 - 0.56	5.2	04/21/98	12:00 pm	05/05/98	12:00 pm	90	9.0	-	
0999	EML	0.24 - 0.34	5.2	04/21/98	12:00 pm	05/05/98	12:00 pm	69	6.9	-	
1000	EML	0.069 - 0.24	5.2	04/21/98	12:00 pm	05/05/98	12:00 pm	54	5.4	-	
1021	Key 1	8.54 - 15	5.2	05/05/98	10:00 am	05/19/98	09:50 am	638.8	12.78	271.9	13.60
1025	Key 1	4.26 - 8.54	5.2	05/05/98	10:00 am	05/19/98	09:50 am	460.1	9.20	222.6	11.13
1019	Key 1	2.12 - 4.26	5.2	05/05/98	10:00 am	05/19/98	09:50 am	457.6	9.15	250.4	12.52
1039	Key 1	1.15 - 2.12	5.2	05/05/98	10:00 am	05/19/98	09:50 am	194.3	3.89	107.9	5.40

Sample No.	Analysis Laboratory	Size Cut (µm)	Sample Length (cm)	Starting Date	Starting Time	Ending Date	Ending Time	²³⁸ U (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)	²³² Th (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)
1030	Key 1	0.56 - 1.15	5.2	05/05/98	10:00 am	05/19/98	09:50 am	113.7	2.27	71.0	3.55
1017	Key 1	0.34 - 0.56	5.2	05/05/98	10:00 am	05/19/98	09:50 am	77.9	1.56	23.7	1.18
1023	Key 1	0.24 - 0.34	5.2	05/05/98	10:00 am	05/19/98	09:50 am	195.9	3.92	1318.3	65.92
1028	Key 1	0.069 - 0.24	5.2	05/05/98	10:00 am	05/19/98	09:50 am	90.9	1.82	58.5	2.92
1020	Key 1	8.54 - 15	5.2	05/19/98	10:00 am	06/02/98	09:40 am	329.8	6.60	358.6	17.93
1024	Key 1	4.26 - 8.54	5.2	05/19/98	9:50 am	06/02/98	09:40 am	293.3	5.87	173.1	8.65
1018	Key 1	2.12 - 4.26	5.2	05/19/98	9:50 am	06/02/98	09:40 am	261.8	5.24	137.1	6.86
1026	Key 1	1.15 - 2.12	5.2	05/19/98	9:50 am	06/02/98	09:40 am	142.9	2.86	113.1	5.66
1040	Key 1	0.56 - 1.15	5.2	05/19/98	9:50 am	06/02/98	09:40 am	98.9	1.98	55.5	2.78
1029	Key 1	0.34 - 0.56	5.2	05/19/98	9:50 am	06/02/98	09:40 am	113.1	2.26	61.2	3.06
1022	Key 1	0.24 - 0.34	5.2	05/19/98	9:50 am	06/02/98	09:40 am	109.3	2.19	85.8	4.29
1027	Key 1	0.069 - 0.24	5.2	05/19/98	9:50 am	06/02/98	09:40 am	80.5	1.61	49.4	2.47
1012	Key 1	8.54 - 15	5.2	06/02/98	10:40 am	06/16/98	09:43 am	282.7	5.65	181.1	9.06
1005	Key 1	4.26 - 8.54	5.2	06/02/98	10:40 am	06/16/98	09:43 am	205.5	4.11	73.3	3.66
1015	Key 1	2.12 - 4.26	5.2	06/02/98	10:40 am	06/16/98	09:43 am	215.3	4.31	122.2	6.11
1003	Key 1	1.15 - 2.12	5.2	06/02/98	10:40 am	06/16/98	09:43 am	125.6	2.51	83.5	4.18
1001	Key 1	0.56 - 1.15	5.2	06/02/98	10:40 am	06/16/98	09:43 am	112.3	2.25	56.3	2.82
1008	Key 1	0.34 - 0.56	5.2	06/02/98	10:40 am	06/16/98	09:43 am	138.7	2.77	26.9	1.34
1004	Key 1	0.24 - 0.34	5.2	06/02/98	10:40 am	06/16/98	09:43 am	77.0	1.54	17.2	0.86
1011	Key 1	0.069 - 0.24	5.2	06/02/98	10:40 am	06/16/98	09:43 am	61.6	1.23	13.0	0.65
1007	Key 1	8.54 - 15	5.2	06/16/98	9:43 am	06/30/98	08:45 am	353.8	7.08	274.0	13.70
1010	Key 1	4.26 - 8.54	5.2	06/16/98	9:43 am	06/30/98	08:45 am	326.3	6.53	217.8	10.89
1016	Key 1	2.12 - 4.26	5.2	06/16/98	9:43 am	06/30/98	08:45 am	358.6	7.17	185.1	9.26
1009	Key 1	1.15 - 2.12	5.2	06/16/98	9:43 am	06/30/98	08:45 am	200.2	4.00	172.9	8.65
1006	Key 1	0.56 - 1.15	5.2	06/16/98	9:43 am	06/30/98	08:45 am	140.9	2.82	86.5	4.32
1014	Key 1	0.34 - 0.56	5.2	06/16/98	9:43 am	06/30/98	08:45 am	129.6	2.59	29.2	1.46
1013	Key 1	0.24 - 0.34	5.2	06/16/98	9:43 am	06/30/98	08:45 am	77.3	1.55	19.6	0.98

Sample No.	Analysis Laboratory	Size Cut (µm)	Sample Length (cm)	Starting Date	Starting Time	Ending Date	Ending Time	²³⁸ U (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)	²³² Th (pg foil ⁻¹)	Counting Error (pg foil-1)
1002	Key 1	0.069 - 0.24	5.2	06/16/98	9:43 am	06/30/98	08:45 am	114.0	2.28	20.0	1.00
1038	Key 1	8.54 - 15	5.6	06/30/98	9:50 am	07/14/98	09:44 am	473.0	9.46	315.3	15.76
1089	Key 2	4.26 - 8.54	5.6	06/30/98	9:50 am	07/14/98	09:44 am	224.9	4.50	176.7	8.83
1078	Key 2	2.12 - 4.26	5.6	06/30/98	9:50 am	07/14/98	09:44 am	292.3	5.85	147.1	7.36
1076	Key 2	1.15 - 2.12	5.6	06/30/98	9:50 am	07/14/98	09:44 am	142.9	2.86	85.4	4.27
1098	Key 2	0.56 - 1.15	5.6	06/30/98	9:50 am	07/14/98	09:44 am	92.6	1.85	77.4	3.87
1083	Key 2	0.34 - 0.56	5.6	06/30/98	9:50 am	07/14/98	09:44 am	83.6	1.67	18.9	0.94
1086	Key 2	0.24 - 0.34	5.6	06/30/98	9:50 am	07/14/98	09:44 am	70.4	1.41	19.6	0.98
1094	Key 2	0.069 - 0.24	5.6	06/30/98	9:50 am	07/14/98	09:44 am	73.1	1.46	36.8	1.84
1093	Key 2	8.54 - 15	5.6	07/14/98	9:44 am	07/28/98	09:37 am	615.5	12.31	343.0	17.15
1075	Key 2	4.26 - 8.54	5.6	07/14/98	9:44 am	07/28/98	09:37 am	402.1	8.04	130.3	6.52
1088	Key 2	2.12 - 4.26	5.6	07/14/98	9:44 am	07/28/98	09:37 am	343.9	6.88	137.0	6.85
1037	Key 1	1.15 - 2.12	5.6	07/14/98	9:44 am	07/28/98	09:37 am	165.7	3.31	85.5	4.28
1081	Key 2	0.56 - 1.15	5.6	07/14/98	9:44 am	07/28/98	09:37 am	117.7	2.35	33.7	1.69
1084	Key 2	0.34 - 0.56	5.6	07/14/98	9:44 am	07/28/98	09:37 am	87.8	1.76	18.9	0.94
1087	Key 2	0.24 - 0.34	5.6	07/14/98	9:44 am	07/28/98	09:37 am	72.7	1.45	21.9	1.10
1095	Key 2	0.069 - 0.24	5.6	07/14/98	9:44 am	07/28/98	09:37 am	91.3	1.83	26.0	1.30
1092	Key 2	8.54 - 15	5.6	07/28/98	9:37 am	08/11/98	09:30 am	236.1	4.72	142.8	7.14
1090	Key 2	4.26 - 8.54	5.6	07/28/98	9:37 am	08/11/98	09:30 am	615.8	12.32	105.5	5.28
1091	Key 2	2.12 - 4.26	5.6	07/28/98	9:37 am	08/11/98	09:30 am	276.1	5.52	144.7	7.24
1077	Key 2	1.15 - 2.12	5.6	07/28/98	9:37 am	08/11/98	09:30 am	161.9	3.24	83.9	4.20
1097	Key 2	0.56 - 1.15	5.6	07/28/98	9:37 am	08/11/98	09:30 am	104.2	2.08	72.7	3.64
1082	Key 2	0.34 - 0.56	5.6	07/28/98	9:37 am	08/11/98	09:30 am	93.6	1.87	51.9	2.60
1085	Key 2	0.24 - 0.34	5.6	07/28/98	9:37 am	08/11/98	09:30 am	56.6	1.13	19.6	0.98
1096	Key 2	0.069 - 0.24	5.6	07/28/98	9:37 am	08/11/98	09:30 am	72.1	1.44	51.5	2.58
1051	Key 2	8.54 - 15	5.6	08/11/98	10:00 am	08/25/98	09:30 am	659.3	13.19	86.1	4.30
1058	Key 2	4.26 - 8.54	5.6	08/11/98	10:00 am	08/25/98	09:30 am	352.4	7.05	98.0	4.90

Sample No.	Analysis Laboratory	Size Cut (µm)	Sample Length (cm)	Starting Date	Starting Time	Ending Date	Ending Time	²³⁸ U (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)	²³² Th (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)
1043	Key 2	2.12 - 4.26	5.6	08/11/98	10:00 am	08/25/98	09:30 am	507.6	10.15	128.1	6.40
1049	Key 2	1.15 - 2.12	5.6	08/11/98	10:00 am	08/25/98	09:30 am	255.3	5.11	245.9	12.30
1057	Key 2	0.56 - 1.15	5.6	08/11/98	10:00 am	08/25/98	09:30 am	130.4	2.61	30.1	1.50
1063	Key 2	0.34 - 0.56	5.6	08/11/98	10:00 am	08/25/98	09:30 am	90.3	1.81	40.3	2.02
1047	Key 2	0.24 - 0.34	5.6	08/11/98	10:00 am	08/25/98	09:30 am	120.7	2.41	74.9	3.75
1066	Key 2	0.069 - 0.24	5.6	08/11/98	10:00 am	08/25/98	09:30 am	102.4	2.05	28.9	1.44
1044	Key 2	8.54 - 15	5.6	08/25/98	9:30 am	09/08/98	09:00 am	349.7	6.99	169.2	8.46
1048	Key 2	4.26 - 8.54	5.6	08/25/98	9:30 am	09/08/98	09:00 am	679.5	13.59	131.3	6.57
1064	Key 2	2.12 - 4.26	5.6	08/25/98	9:30 am	09/08/98	09:00 am	378.7	7.57	119.6	5.98
1045	Key 2	1.15 - 2.12	5.6	08/25/98	9:30 am	09/08/98	09:00 am	216.1	4.32	80.9	4.05
1069	Key 2	0.56 - 1.15	5.6	08/25/98	9:30 am	09/08/98	09:00 am	160.2	3.20	67.5	3.38
1067	Key 2	0.34 - 0.56	5.6	08/25/98	9:30 am	09/08/98	09:00 am	79.7	1.59	10.8	0.54
1074	Key 2	0.24 - 0.34	5.6	08/25/98	9:30 am	09/08/98	09:00 am	76.5	1.53	36.1	1.81
1061	Key 2	0.069 - 0.24	5.6	08/25/98	9:30 am	09/08/98	09:00 am	79.8	1.60	22.6	1.13
1070	Key 2	8.54 - 15	5.6	09/08/98	10:00 am	09/22/98	10:00 am	600.7	12.01	184.7	9.24
1065	Key 2	4.26 - 8.54	6.25	09/08/98	10:00 am	09/22/98	10:00 am	662.3	13.25	130.0	6.50
1042	Key 2	2.12 - 4.26	5.6	09/08/98	10:00 am	09/22/98	10:00 am	728.5	14.57	129.2	6.46
1056	Key 2	1.15 - 2.12	5.6	09/08/98	10:00 am	09/22/98	10:00 am	416.3	8.33	105.0	5.25
1060	Key 2	0.56 - 1.15	5.6	09/08/98	10:00 am	09/22/98	10:00 am	227.6	4.55	43.1	2.16
1053	Key 2	0.34 - 0.56	5.6	09/08/98	10:00 am	09/22/98	10:00 am	208.5	4.17	13.1	0.66
1055	Key 2	0.24 - 0.34	5.6	09/08/98	10:00 am	09/22/98	10:00 am	117.4	2.35	30.1	1.50
1054	Key 2	0.069 - 0.24	5.7	09/08/98	10:00 am	09/22/98	10:00 am	122.5	2.45	9.8	0.49
1052	Key 2	8.54 - 15	5.6	09/22/98	10:00 am	10/06/98	10:00 am	221.8	4.44	98.2	4.91
1071	Key 2	4.26 - 8.54	6.25	09/22/98	10:00 am	10/06/98	10:00 am	150.2	3.00	94.5	4.73
1062	Key 2	2.12 - 4.26	5.6	09/22/98	10:00 am	10/06/98	10:00 am	230.0	4.60	84.8	4.24
1046	Key 2	1.15 - 2.12	5.6	09/22/98	10:00 am	10/06/98	10:00 am	179.5	3.59	78.8	3.94
1059	Key 2	0.56 - 1.15	5.6	09/22/98	10:00 am	10/06/98	10:00 am	151.3	3.03	66.2	3.31

Sample No.	Analysis Laboratory	Size Cut (µm)	Sample Length (cm)	Starting Date	Starting Time	Ending Date	Ending Time	²³⁸ U (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)	²³² Th (pg foil ⁻¹)	Counting Error (pg foil-1)
1050	Key 2	0.34 - 0.56	5.6	09/22/98	10:00 am	10/06/98	10:00 am	135.3	2.71	48.5	2.43
1068	Key 2	0.24 - 0.34	5.6	09/22/98	10:00 am	10/06/98	10:00 am	89.2	1.78	33.3	1.66
1041	Key 2	0.069 - 0.24	5.7	09/22/98	10:00 am	10/06/98	10:00 am	109.7	2.19	59.9	3.00
1128	Key 2	8.54 - 15	5.6	10/06/98	10:00 am	10/20/98	10:00 am	219.0	4.38	93.3	4.66
1136	Key 2	4.26 - 8.54	5.6	10/06/98	10:00 am	10/20/98	10:00 am	120.0	2.40	58.8	2.94
1118	Key 2	2.12 - 4.26	5.6	10/06/98	10:00 am	10/20/98	10:00 am	146.3	2.93	84.7	4.24
1121	Key 2	1.15 - 2.12	5.6	10/06/98	10:00 am	10/20/98	10:00 am	118.9	2.38	62.0	3.10
1117	Key 2	0.56 - 1.15	5.6	10/06/98	10:00 am	10/20/98	10:00 am	78.5	1.57	39.8	1.99
1129	Key 2	0.34 - 0.56	5.6	10/06/98	10:00 am	10/20/98	10:00 am	253.0	5.06	53.5	2.68
1119	Key 2	0.24 - 0.34	5.6	10/06/98	10:00 am	10/20/98	10:00 am	50.2	1.00	24.2	1.21
1133	Key 2	0.069 - 0.24	5.6	10/06/98	10:00 am	10/20/98	10:00 am	59.3	1.19	30.3	1.52
1126	Key 2	8.54 - 15	5.6	10/21/98	10:00 am	11/03/98	10:00 am	295.0	5.90	106.9	5.35
1134	Key 2	4.26 - 8.54	5.6	10/21/98	10:00 am	11/03/98	10:00 am	97.4	1.95	41.3	2.06
1131	Key 2	2.12 - 4.26	5.6	10/21/98	10:00 am	11/03/98	10:00 am	130.1	2.60	77.5	3.88
1123	Key 2	1.15 - 2.12	5.6	10/21/98	10:00 am	11/03/98	10:00 am	137.5	2.75	55.5	2.78
1132	Key 2	0.56 - 1.15	5.6	10/21/98	10:00 am	11/03/98	10:00 am	96.6	1.93	82.1	4.10
1122	Key 2	0.34 - 0.56	5.6	10/21/98	10:00 am	11/03/98	10:00 am	56.5	1.13	32.3	1.62
1125	Key 2	0.24 - 0.34	5.6	10/21/98	10:00 am	11/03/98	10:00 am	70.3	1.41	29.1	1.46
1127	Key 2	0.069 - 0.24	5.6	10/21/98	10:00 am	11/03/98	10:00 am	69.9	1.40	29.2	1.46
1112	Key 2	8.54 - 15	5.6	11/03/98	10:00 am	11/17/98	12:30 pm	382.3	7.65	232.3	11.62
1106	Key 2	4.26 - 8.54	5.6	11/03/98	10:00 am	11/17/98	12:30 pm	141.4	2.83	39.1	1.96
1110	Key 2	2.12 - 4.26	5.6	11/03/98	10:00 am	11/17/98	12:30 pm	140.1	2.80	64.3	3.22
1099	Key 2	1.15 - 2.12	5.35	11/03/98	10:00 am	11/17/98	12:30 pm	100.9	2.02	66.8	3.34
1107	Key 2	0.56 - 1.15	5.35	11/03/98	10:00 am	11/17/98	12:30 pm	88.7	1.77	36.4	1.82
1111	Key 2	0.34 - 0.56	5.6	11/03/98	10:00 am	11/17/98	12:30 pm	136.9	2.74	71.2	3.56
1115	Key 2	0.24 - 0.34	5.6	11/03/98	10:00 am	11/17/98	12:30 pm	57.2	1.14	27.6	1.38
1109	Key 2	0.069 - 0.24	5.6	11/03/98	10:00 am	11/17/98	12:30 pm	67.5	1.35	66.3	3.32

TABLE 1 (Cont'd.)

Sample No.	Analysis Laboratory	Size Cut (µm)	Sample Length (cm)	Starting Date	Starting Time	Ending Date	Ending Time	²³⁸ U (pg foil ⁻¹)	Counting Error (pg foil ⁻¹)	(pg foil ⁻¹)	Counting Error (pg foil ⁻¹)
1105	Key 2	8.54 - 15	5.6	11/17/98	12:30 pm	12/01/98	10:00 am	134.4	2.69	59.5	2.98
1108	Key 2	4.26 - 8.54	5.6	11/17/98	12:30 pm	12/01/98	10:00 am	95.1	1.90	63.5	3.18
1102	Key 2	2.12 - 4.26	5.6	11/17/98	12:30 pm	12/01/98	10:00 am	83.7	1.67	41.3	2.06
1114	Key 2	1.15 - 2.12	5.35	11/17/98	12:30 pm	12/01/98	10:00 am	77.7	1.55	40.8	2.04
1113	Key 2	0.56 - 1.15	5.35	11/17/98	12:30 pm	12/01/98	10:00 am	71.5	1.43	37.4	1.87
1104	Key 2	0.34 - 0.56	5.6	11/17/98	12:30 pm	12/01/98	10:00 am	45.5	0.91	14.3	0.72
1100	Key 2	0.24 - 0.34	5.6	11/17/98	12:30 pm	12/01/98	10:00 am	54.7	1.09	31.7	1.58
1103	Key 2	0.069 - 0.24	5.6	11/17/98	12:30 pm	12/01/98	10:00 am	43.2	0.86	22.8	1.14

TABLE 2 QA DATA FOR ALL SAMPLES ANALYZED

Sample No.	Analysis Laboratory	Sample length (cm)	(pg foil-1)	²³⁸ U (pg cm ⁻¹)	²³² Th (pg foil ⁻¹)	²³² Th (pg cm ⁻¹)
Blank Data						
D1005	EML	4.2	18	4.2	-	-
D1006	EML	4.2	19	4.5	-	-
1031	Key 1	8.8	35.8	4.1	21.4	2.43
1033	Key 1	8.7	26.6	3.1	11.6	1.33
1036	Key 1	8.3	36.0	4.3	11.0	1.33
1072	Key 2	4.0	67.8	17.0	35.5	8.88
1073*	Key 2	4.0	37.5	9.4	-0.3	-0.08
1101	Key 2	4.0	44.1	11.0	19.8	4.95
1116	Key 2	4.0	30.7	7.7	22.8	5.70
1120	Key 2	4.0	33.0	8.2	25.5	6.38
1161	Key 2	4.0	61.4	15.4	41.1	10.28
1162	Key 2	4.0	41.4	10.4	38.5	9.62
1164	Key 2	4.0	46.3	11.6	30.9	7.72
1165	Key 2	13.5	142.0	10.5	51.2	3.79
1166	Key 2	19.0	150.1	7.9	64.2	3.38
1167	Key 2	19.3	150.1	7.8	56.4	2.92
1168	Key 2	29.7	308.7	10.4	102.9	3.46
1163*	Key 2	4.0	459.3	114.8	45.5	11.375
Analysis Labo	oratory San	nple Type	²³⁸ U (pg cm ⁻¹)	SD**	²³² Th (pg cm ⁻¹)	SD**
Blank Data Sun	<u>mmary</u>					
EML		Blank	4.4	0.1	_	_
Key: Batc		Blank	3.8	0.3	1.9	.3
Key: Batc		Blank	10.6	0.8	6.5	.6

^{*}Outlying data was not used in the calculations.
** Standard deviation.

TABLE 3

DIFFERENCE BETWEEN EML AND KEY (BATCH 1) IN AVERAGE BLANK VALUES

Sample No.	Analysis Laboratory	Sample Length (cm)	²³⁸ U Spike (pg)	²³⁸ U (pg foil ⁻¹)	% Deviation	²³² Th Spike (pg)	²³² Th (pg foil ⁻¹)
Spike Data							
1032	Key 1	8.7	100	118.4	0.15	0	21.0
1034	Key 1	8.6	100	140.4	0.08	0	27.2
1035	Key 1	8.5	100	114.6	0.18	0	6.7
1079	Key 2	4.0	100	135.1	0.07	0	13.3
1080	Key 2	4.0	100	141.5	0.01	0	13.5
1124	Key 2	4.0	100	139.0	0.03	0	27.0
1130	Key 2	4.0	100	121.9	0.20	0	21.7
1135	Key 2	4.0	100	136.1	0.06	0	22.2
1139	Key 2	4.0	100	150.3	0.08	0	39.8
1140	Key 2	4.0	100	133.8	0.09	0	28.2
1141	Key 2	4.0	100	135.8	0.07	0	37.2
1142	Key 2	4.0	100	131.9	0.10	0	30.4

	²³⁸ U (pg foil ⁻¹)	(pg foil ⁻¹) blank corrected	% difference *
Spike Summary Data			
Key: Batch 1	124.5	91.6	13
Key: Batch 1	136.2	93.8	8

^{*}Each sample spiked with 100 pg $^{238}\mathrm{U}$

TABLE 4

CONCENTRATION DATA

Sample No.	Standard Sample Volume (m³)	Average Sampling Temperature (°k)	Average Sampling Pressure (Atm)	²³⁸ U (pg m ⁻³)	Error (pg m ⁻³)	²³² Th (pg m ⁻³)	Error (pg m ⁻³)
0953	23.35	277.0	0.98	2.1	0.4	-	
0954	23.35	277.0	0.98	3.1	0.5	-	
0955	23.35	277.0	0.98	4.0	0.6	-	
0956	23.35	277.0	0.98	2.5	0.4	-	
0957	23.35	277.0	0.98	1.0	0.2	-	
0958	23.35	277.0	0.98	1.0	0.2	-	
0959	23.35	277.0	0.98	2.0	0.3	-	
0960	23.35	277.0	0.98	1.0	0.2	-	
0961	23.13	279.3	0.98	8.5	1.2	-	
0962	23.13	279.3	0.98	7.9	1.2	-	
0963	23.13	279.3	0.98	5.8	0.9	-	
0964	23.13	279.3	0.98	3.9	0.6	-	
0965	23.13	279.3	0.98	1.4	0.3	-	
0966	23.13	279.3	0.98	2.2	0.4	-	
0967	23.13	279.3	0.98	1.7	0.3	-	
0968	23.13	279.3	0.98	2.3	0.4	-	
0969	23.61	275.0	0.99	4.3	0.7	-	
0970	23.61	275.0	0.99	2.2	0.4	-	
0971	23.61	275.0	0.99	3.7	0.6	-	
0972	23.61	275.0	0.99	2.1	0.3	-	
0973	23.61	275.0	0.99	1.5	0.3	-	
0974	23.61	275.0	0.99	0.7	0.2	-	
0975	23.61	275.0	0.99	0.2	0.1	-	
0976	23.61	275.0	0.99	3.2	0.5	-	
0977	22.51	286.9	0.98	8.7	1.3	-	
0978	22.51	286.9	0.98	6.1	0.9	-	
0979	22.51	286.9	0.98	13.8	2.0	-	

TABLE 4 (Cont'd.)

Sample No.	Standard Sample Volume (m³)	Average Sampling Temperature (°k)	Average Sampling Pressure (Atm)	²³⁸ U (pg m ⁻³)	Error (pg m ⁻³)	²³² Th (pg m ⁻³)	Error (pg m ⁻³)
0980	22.51	286.9	0.98	5.5	0.8	-	
0981	22.51	286.9	0.98	3.0	0.5	-	
0982	22.51	286.9	0.98	1.4	0.3	-	
0983	22.51	286.9	0.98	1.7	0.3	-	
0984	22.51	286.9	0.98	1.3	0.2	-	
0985	22.69	284.5	0.98	17.2	2.5	-	
0986	22.69	284.5	0.98	13.3	1.9	-	
0987	22.69	284.5	0.98	11.7	1.7	-	
0988	22.69	284.5	0.98	4.4	0.7	-	
0989	22.69	284.5	0.98	2.7	0.4	-	
0990	22.69	284.5	0.98	2.5	0.4	-	
0991	22.69	284.5	0.98	2.4	0.4	-	
0992	22.69	284.5	0.98	1.3	0.2	-	
0993	22.61	286.4	0.98	8.1	1.2	-	
0994	22.61	286.4	0.98	11.0	1.6	-	
0995	22.61	286.4	0.98	7.7	1.1	-	
0996	22.61	286.4	0.98	3.7	0.6	-	
0997	22.61	286.4	0.98	2.2	0.4	-	
0998	22.61	286.4	0.98	3.7	0.6	-	
0999	22.61	286.4	0.98	2.8	0.4	-	
1000	22.61	286.4	0.98	2.1	0.4	-	
1021	22.12	292.5	0.98	28.0	1.7	11.8	1.1
1025	22.12	292.5	0.98	19.9	1.2	9.6	0.9
1019	22.12	292.5	0.98	19.8	1.2	10.9	1.0
1039	22.12	292.5	0.98	7.9	0.5	4.4	0.4
1030	22.12	292.5	0.98	4.2	0.3	2.8	0.3
1017	22.12	292.5	0.98	2.6	0.2	0.6	0.1
1023	22.12	292.5	0.98	8.0	0.5	59.2	5.4

TABLE 4 (Cont'd.)

Sample No.	Standard Sample Volume (m³)	Average Sampling Temperature (°k)	Average Sampling Pressure (Atm)	²³⁸ U (pg m ⁻³)	Error (pg m ⁻³)	²³² Th (pg m ⁻³)	Error (pg m ⁻³)
1028	22.12	292.5	0.98	3.2	0.2	2.2	0.2
1020	22.02	293.8	0.98	14.1	0.9	15.8	1.5
1024	22.02	293.8	0.98	12.4	0.8	7.4	0.7
1018	22.02	293.8	0.98	11.0	0.7	5.8	0.6
1026	22.02	293.8	0.98	5.6	0.4	4.7	0.5
1040	22.02	293.8	0.98	3.6	0.3	2.1	0.2
1029	22.02	293.8	0.98	4.2	0.3	2.3	0.3
1022	22.02	293.8	0.98	4.1	0.3	3.4	0.4
1027	22.02	293.8	0.98	2.8	0.2	1.8	0.2
1012	22.1	291.4	0.98	11.9	0.8	7.7	0.7
1005	22.1	291.4	0.98	8.4	0.5	2.9	0.3
1015	22.1	291.4	0.98	8.8	0.6	5.1	0.5
1003	22.1	291.4	0.98	4.8	0.3	3.3	0.3
1001	22.1	291.4	0.98	4.2	0.3	2.1	0.2
1008	22.1	291.4	0.98	5.4	0.4	0.8	0.1
1004	22.1	291.4	0.98	2.6	0.2	0.3	0.1
1011	22.1	291.4	0.98	1.9	0.2	0.3	0.1
1007	21.72	297.8	0.98	15.4	1.0	12.2	1.1
1010	21.72	297.8	0.98	14.1	0.9	9.6	0.9
1016	21.72	297.8	0.98	15.6	1.0	8.1	0.8
1009	21.72	297.8	0.98	8.3	0.5	7.5	0.7
1006	21.72	297.8	0.98	5.6	0.4	3.5	0.4
1014	21.72	297.8	0.98	5.1	0.3	0.9	0.1
1013	21.72	297.8	0.98	2.6	0.2	0.4	0.1
1002	21.72	297.8	0.98	4.3	0.3	0.5	0.1
1038	21.91	296.2	0.98	20.6	1.3	13.9	1.3
1089	21.91	296.2	0.98	7.5	0.6	6.4	0.7
1078	21.91	296.2	0.98	10.5	0.7	5.1	0.6

TABLE 4 (Cont'd.)

Sample No.	Standard Sample Volume (m³)	Average Sampling Temperature (°k)	Average Sampling Pressure (Atm)	²³⁸ U (pg m ⁻³)	Error (pg m ⁻³)	²³² Th (pg m ⁻³)	Error (pg m ⁻³)
1076	21.91	296.2	0.98	3.7	0.3	2.2	0.4
1098	21.91	296.2	0.98	1.4	0.2	1.9	0.3
1083	21.91	296.2	0.98	1.0	0.2	0.3	0.2
1086	21.91	296.2	0.98	0.4	0.1	0.3	0.2
1094	21.91	296.2	0.98	0.5	0.1	0.3	0.2
1093	21.91	296.6	0.98	25.3	1.6	14.0	1.4
1075	21.91	296.6	0.98	15.5	1.0	4.3	0.6
1088	21.91	296.6	0.98	12.9	0.9	4.6	0.6
1037	21.91	296.6	0.98	6.6	0.4	3.4	0.4
1081	21.91	296.6	0.98	2.6	0.3	0.3	0.2
1084	21.91	296.6	0.98	1.2	0.2	0.3	0.2
1087	21.91	296.6	0.98	0.5	0.1	0.3	0.2
1095	21.91	296.6	0.98	1.4	0.2	0.3	0.2
1092	21.93	296.9	0.99	8.0	0.6	4.9	0.6
1090	21.93	296.9	0.99	25.3	1.6	3.2	0.5
1091	21.93	296.9	0.99	9.8	0.7	4.9	0.6
1077	21.93	296.9	0.99	4.6	0.4	2.2	0.4
1097	21.93	296.9	0.99	1.9	0.2	1.7	0.3
1082	21.93	296.9	0.99	1.5	0.2	0.7	0.2
1085	21.93	296.9	0.99	0.2	0.1	0.3	0.2
1096	21.93	296.9	0.99	0.5	0.1	0.7	0.2
1051	21.95	296.0	0.98	27.2	1.8	2.3	0.4
1058	21.95	296.0	0.98	13.2	0.9	2.8	0.4
1043	21.95	296.0	0.98	20.3	1.3	4.2	0.5
1049	21.95	296.0	0.98	8.8	0.6	9.5	1.0
1057	21.95	296.0	0.98	3.1	0.3	0.3	0.2
1063	21.95	296.0	0.98	1.3	0.2	0.3	0.2
1047	21.95	296.0	0.98	2.7	0.3	1.8	0.3

TABLE 4 (Cont'd.)

Sample No.	Standard Sample Volume (m³)	Average Sampling Temperature (°k)	Average Sampling Pressure (Atm)	²³⁸ U (pg m ⁻³)	Error (pg m ⁻³)	²³² Th (pg m ⁻³)	Error (pg m ⁻³)
1066	21.95	296.0	0.98	1.9	0.2	0.3	0.2
1044	21.91	295.3	0.98	13.1	0.9	6.1	0.7
1048	21.91	295.3	0.98	28.2	1.8	4.3	0.6
1064	21.91	295.3	0.98	14.5	1.0	3.8	0.5
1045	21.91	295.3	0.98	7.1	0.5	2.0	0.3
1069	21.91	295.3	0.98	4.5	0.4	1.4	0.3
1067	21.91	295.3	0.98	0.8	0.2	0.3	0.1
1074	21.91	295.3	0.98	0.7	0.2	0.3	0.2
1061	21.91	295.3	0.98	0.8	0.2	0.3	0.2
1070	22.09	293.9	0.98	24.4	1.6	6.7	0.8
1065	22.09	293.9	0.98	26.9	1.7	4.0	0.5
1042	22.09	293.9	0.98	30.2	1.9	4.2	0.5
1056	22.09	293.9	0.98	16.1	1.1	3.1	0.4
1060	22.09	293.9	0.98	7.5	0.6	0.3	0.2
1053	22.09	293.9	0.98	6.7	0.5	0.3	0.1
1055	22.09	293.9	0.98	2.5	0.3	0.3	0.2
1054	22.09	293.9	0.98	2.7	0.3	0.3	0.1
1052	22.34	291.2	0.98	7.2	0.5	2.8	0.4
1071	22.34	291.2	0.98	3.6	0.3	2.4	0.4
1062	22.34	291.2	0.98	7.5	0.6	2.2	0.4
1046	22.34	291.2	0.98	5.3	0.4	1.9	0.3
1059	22.34	291.2	0.98	4.0	0.4	1.3	0.3
1050	22.34	291.2	0.98	3.3	0.3	0.5	0.2
1068	22.34	291.2	0.98	1.2	0.2	0.3	0.2
1041	22.34	291.2	0.98	2.1	0.2	1.0	0.3
1128	22.78	286.5	0.99	6.9	0.5	2.5	0.4
1136	22.78	286.5	0.99	2.6	0.3	1.0	0.3
1118	22.78	286.5	0.99	3.7	0.3	2.1	0.4

TABLE 4 (Cont'd.)

Sample No.	Standard Sample Volume (m³)	Average Sampling Temperature (°k)	Average Sampling Pressure (Atm)	²³⁸ U (pg m ⁻³)	Error (pg m ⁻³)	²³² Th (pg m ⁻³)	Error (pg m ⁻³)
1121	22.78	286.5	0.99	2.5	0.3	1.1	0.3
1117	22.78	286.5	0.99	0.7	0.2	0.3	0.2
1129	22.78	286.5	0.99	8.4	0.6	0.8	0.2
1119	22.78	286.5	0.99	0.2	0.1	0.3	0.2
1133	22.78	286.5	0.99	0.2	0.1	0.3	0.2
1126	21.38	283.4	0.99	10.9	0.8	3.3	0.5
1134	21.38	283.4	0.99	1.7	0.2	0.3	0.2
1131	21.38	283.4	0.99	3.2	0.3	1.9	0.3
1123	21.38	283.4	0.99	3.6	0.3	0.9	0.2
1132	21.38	283.4	0.99	1.6	0.2	2.1	0.4
1122	21.38	283.4	0.99	0.2	0.1	0.3	0.2
1125	21.38	283.4	0.99	0.4	0.1	0.3	0.2
1127	21.38	283.4	0.99	0.4	0.1	0.3	0.2
1112	23.45	279.5	0.98	13.7	0.9	8.4	0.9
1106	23.45	279.5	0.98	3.4	0.3	0.3	0.2
1110	23.45	279.5	0.98	3.3	0.3	1.2	0.3
1099	23.45	279.5	0.98	1.8	0.2	1.4	0.3
1107	23.45	279.5	0.98	1.3	0.2	0.3	0.2
1111	23.45	279.5	0.98	3.2	0.3	1.5	0.3
1115	23.45	279.5	0.98	0.2	0.1	0.3	0.2
1109	23.45	279.5	0.98	0.3	0.1	1.3	0.3
1105	22.98	281.3	0.99	3.2	0.3	1	0.2
1108	22.98	281.3	0.99	1.5	0.2	1.2	0.3
1102	22.98	281.3	0.99	1.0	0.2	0.3	0.2
1114	22.98	281.3	0.99	0.8	0.2	0.3	0.2
1113	22.98	281.3	0.99	0.6	0.1	0.3	0.2

TABLE 4 (Cont'd.)

Sample No.	Standard Sample Volume (m³)	Average Sampling Temperature (°k)	Average Sampling Pressure (Atm)	²³⁸ U (pg m ⁻³)	Error (pg m ⁻³)	²³² Th (pg m ⁻³)	Error (pg m ⁻³)
1104	22.98	281.3	0.99	0.2	0.1	0.3	0.1
1100	22.98	281.3	0.99	0.2	0.1	0.3	0.2
1103	22.98	281.3	0.99	0.2	0.1	0.3	0.2

TABLE 5FERNALD TSP ²³⁸U CONCENTRATION DATA*

Starting Date	Ending Date	Concentration (pg m ⁻³)
2/10/98	02/24/98	43
02/24/98	03/10/98	126
03/10/98	03/24/98	23
03/24/98	04/07/98	195
04/07/98	04/21/98	113
04/21/98	05/05/98	196
05/05/98	05/19/98	281
05/19/98	06/02/98	240
06/02/98	06/16/98	170
06/16/98	06/30/98	220
06/30/98	07/14/98	170
07/14/98	07/28/98	8.3
07/28/98	08/11/98	320
08/11/98	08/25/98	370
08/25/98	09/08/98	340
09/08/98	09/22/98	830
09/22/98	10/06/98	140
10/06/98	10/20/98	132
10/20/98	11/03/98	191
11/03/98	11/17/98	393
11/17/98	12/01/98	97

^{*}Analyzed by Fernald Management Group.

TABLE 6

PERCENT DIFFERENCE* BETWEEN THE FERNALD DOSE CALCULATIONS**

AND THE ICRP MODELS

Dose Model Description	²³⁸ U	²³² Th	Total
3 rd Quarter			
ICRP 30 (@ 1 µm) Inhalation	-21%	-19%	-20%
ICRP 30 (with size) Inhalation	-85%	-17%	-43%
ICRP 66 (with size) Inhalation / Ingestion	-92%	-82%	-86%
Annual			
ICRP 30 (@ 1 µm) Inhalation	-21%		
ICRP 30 (with size) Inhalation	-83%		
ICRP 66 (with size) Inhalation / Ingestion	-92%		

^{*}Percent difference = [(Fernald dose calculation - the ICRP model calculation)/Fernald dose calculation]*100.

^{**}Fernald dose model is based on the CFR40, Part 61 Appendix E, Table 2.

TABLE 7 $EFFECTIVE\ DOSE\ COEFFICIENTS\ (Sv\ Bq^{\text{--}1})\ FOR\ INHALATION$ INTAKES BY AN ADULT (AMAD = 1 $\mu m)$

Nuclide	Class D	Class W	Class Y
ICRP-30 (FGR-11)			
238U	6.62 x 10 ⁻⁷	1.90 x 10 ⁻⁶	3.20 x 10 ⁻⁵
²³² Th	0.02 x 10	4.43×10^{-4}	3.11 x 10 ⁻⁴
III	NA	4.43 A 10	J.11 X 10
ICRP-71			
^{238}U	5.0×10^{-7}	2.9 x 10 ⁻⁶	8.0×10^{-6}
²³² Th	1.1 x 10 ⁻⁰⁴	4.5 x 10 ⁻⁵	2.5 x 10 ⁻⁵

NA = not applicable.

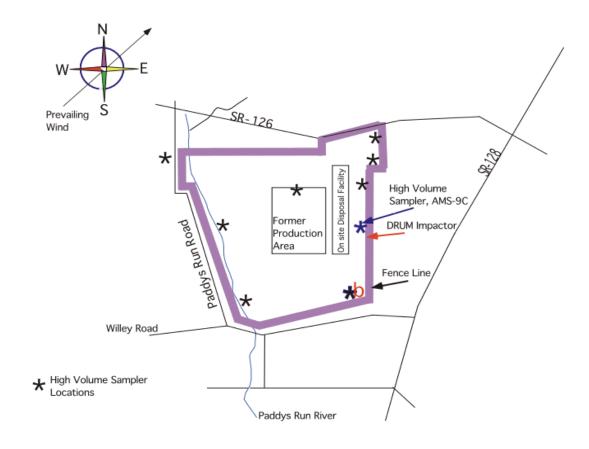


Figure 1. The Fernald Environmental Management site where EML co-located a cascade impactor at the fence line next to a High Volume Sampler.

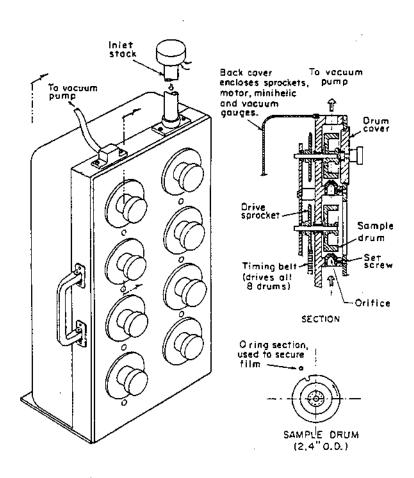


Figure 2. Schematic drawing of the Davis Rotating Universal Size-cut Monitoring Sampler (DRUM) from Raabe et al. (1988).

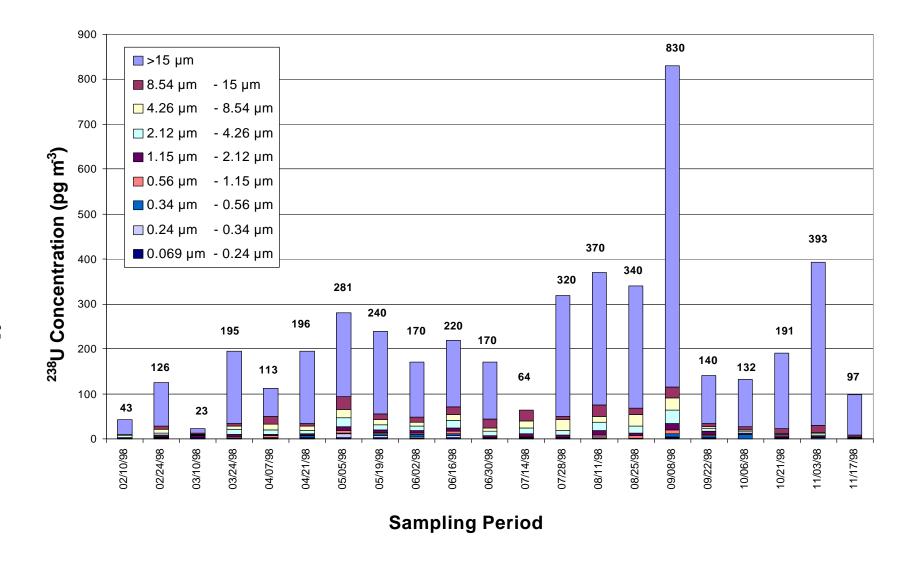


Figure 3. The ²³⁸U concentration below 100 μm (TSP) graphed for each sampling period.

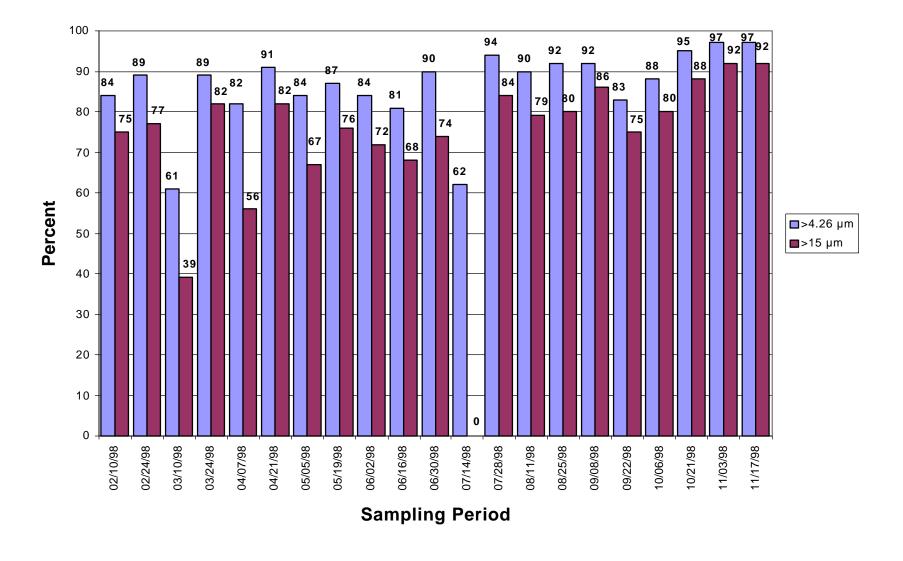


Figure 4. The percent concentration of 238 U above 4.26 μ m (mean = 87%; SD = 4.4%) and 15 μ m (mean = 73%; SD 4.4%) for each sampling period.

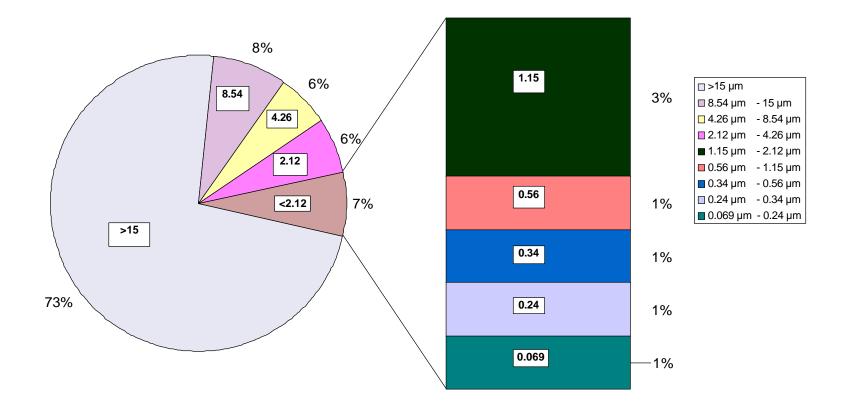


Figure 5. The average percent concentration of ²³⁸U in each sampling size range. The value in each square is the lowest particle size of the size interval.

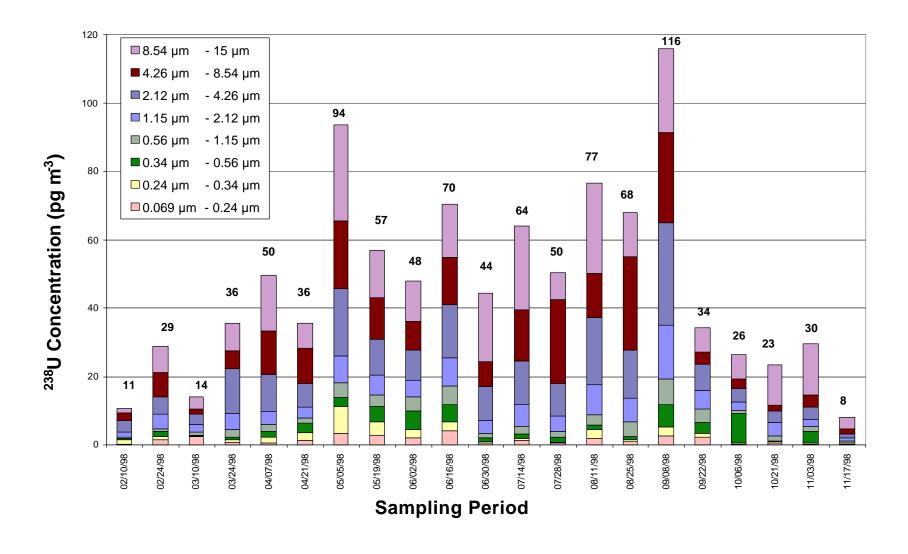
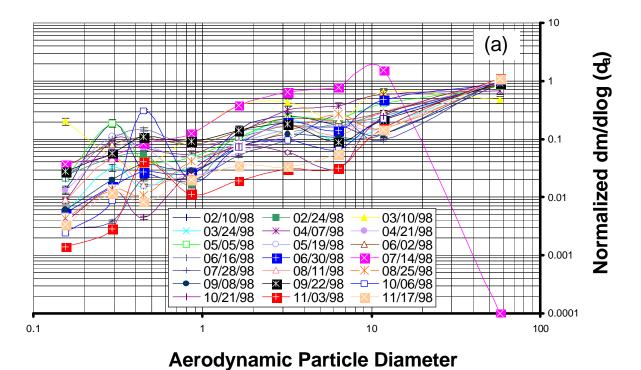


Figure 6. The 238 U concentration below 15 μm for each sampling period. The number above the bar is the total concentration.



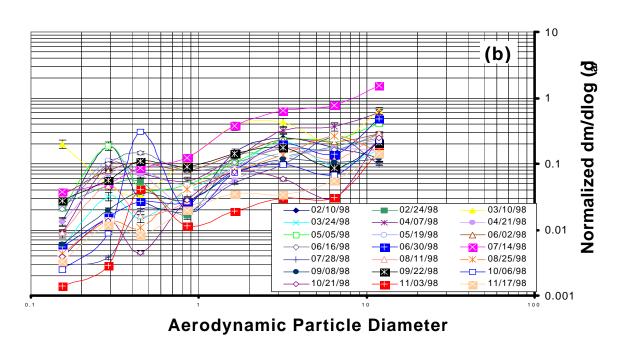
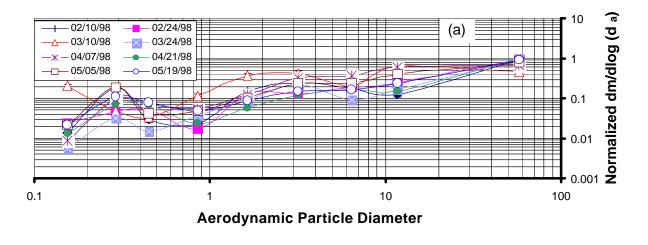


Figure 7. The normalized 238 U size distributions: (a) <100 μ m - the anomalous data from July 14, 1999 are discussed in the text, and (b) <15 μ m.



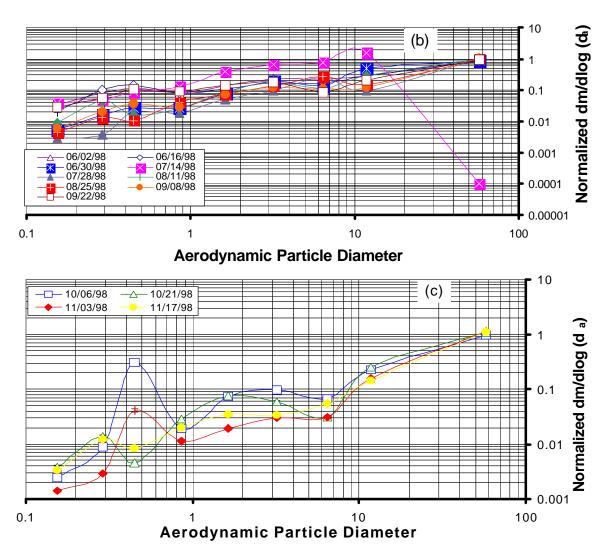


Figure 8. The normalized ²³⁸U size distributions for: (a) winter/spring, (b) summer, and (c) fall/winter samples.

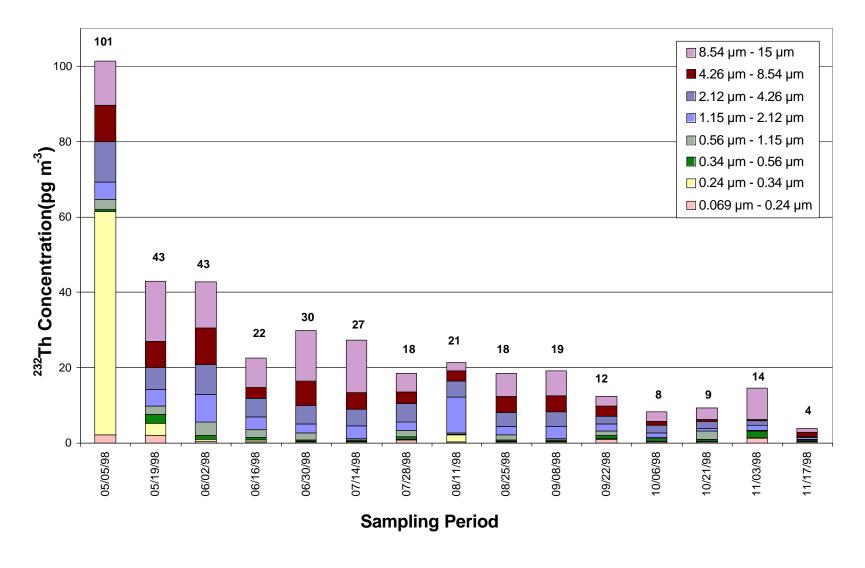


Figure 9. The ²³²Th concentration below 15 μm for each sampling period. The number above the bar is the total concentration.

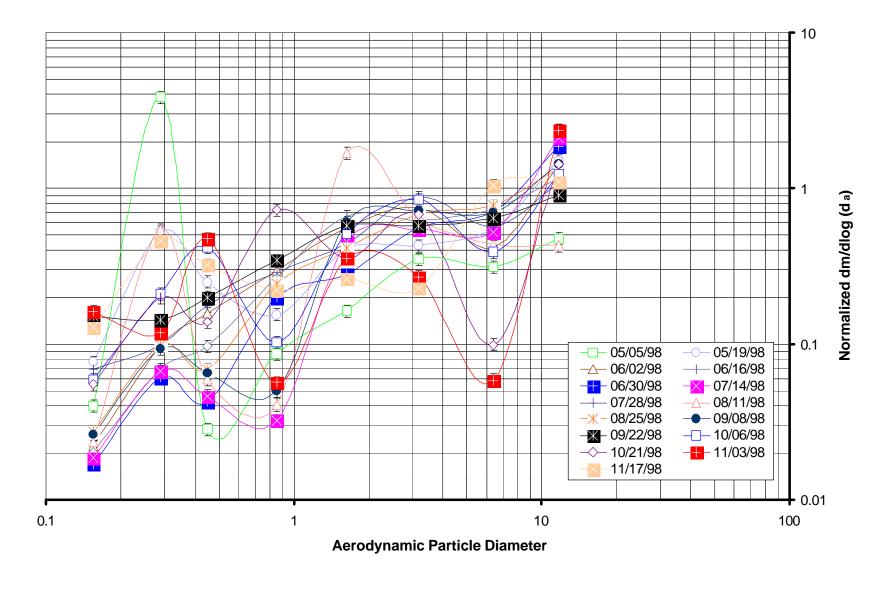


Figure 10. The normalized 232 Th size distributions <15 μ m.

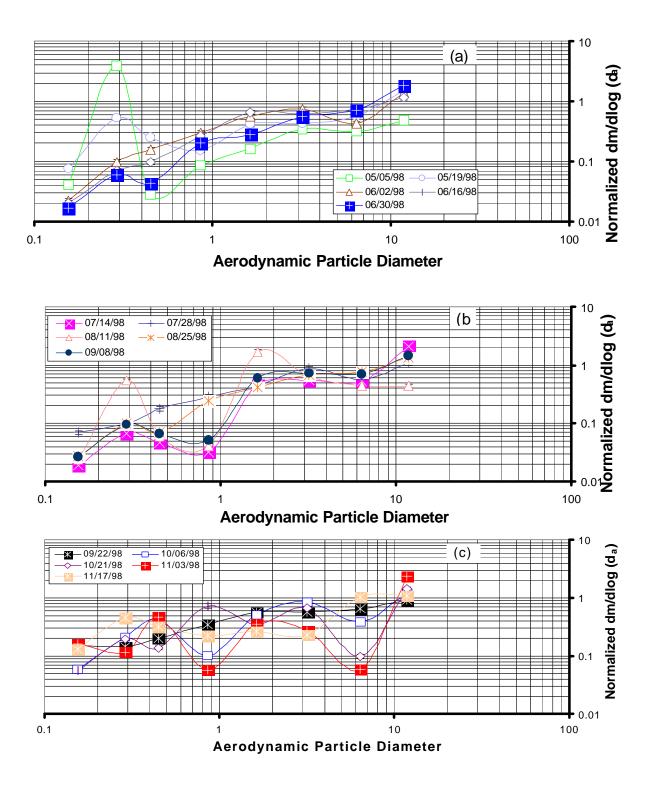


Figure 11. The normalized ²³²Th size distributions for: (a) spring/early summer, (b) early summer/fall, and (c) fall/winter.

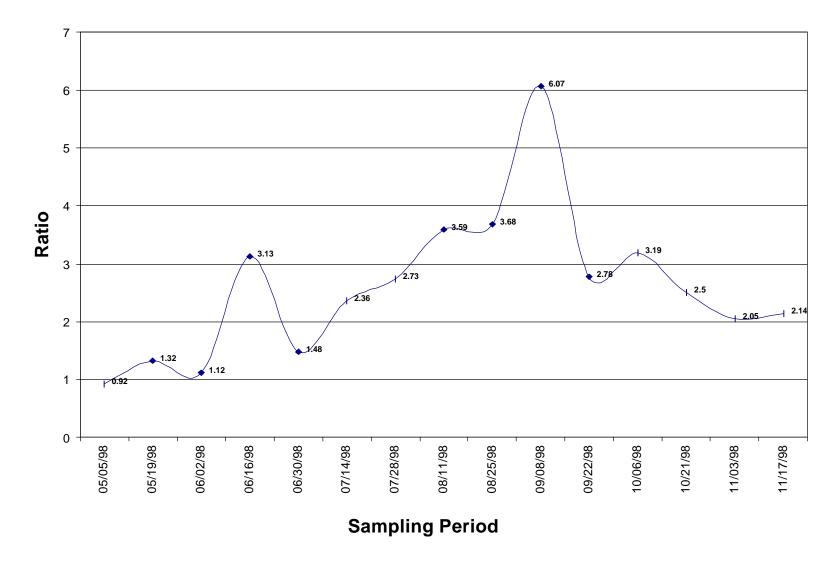


Figure 12. The 238 U/ 232 Th concentration ratio below 15 μ m graphed for each sampling period.

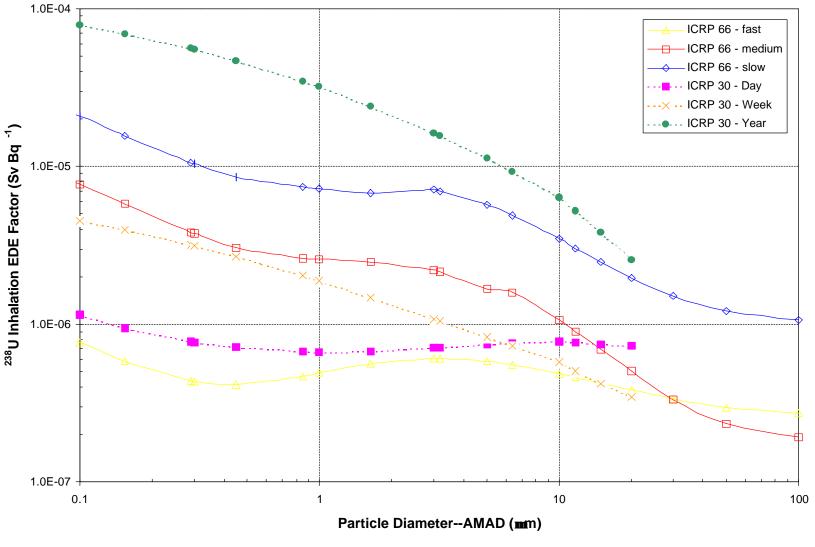


Figure 13. The size dependent inhalation dose conversion factors for ²³⁸U. EDE's calculated with ICRP 66 lung model assume that uranium is 78% bone volume seeker and 22% bone surface seeker.

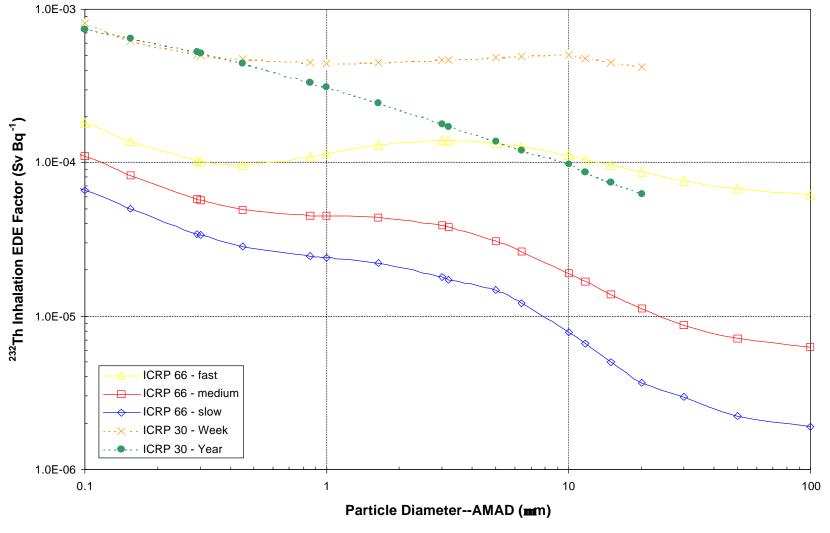


Figure 14. The size dependent inhalation dose conversion factors for ²³²Th. EDE's calculated with ICRP 66 lung model assume that thorium is 78% bone volume seeker and 22% bone surface seeker. No thorium dose factors are given for ICRP 30 Day case.

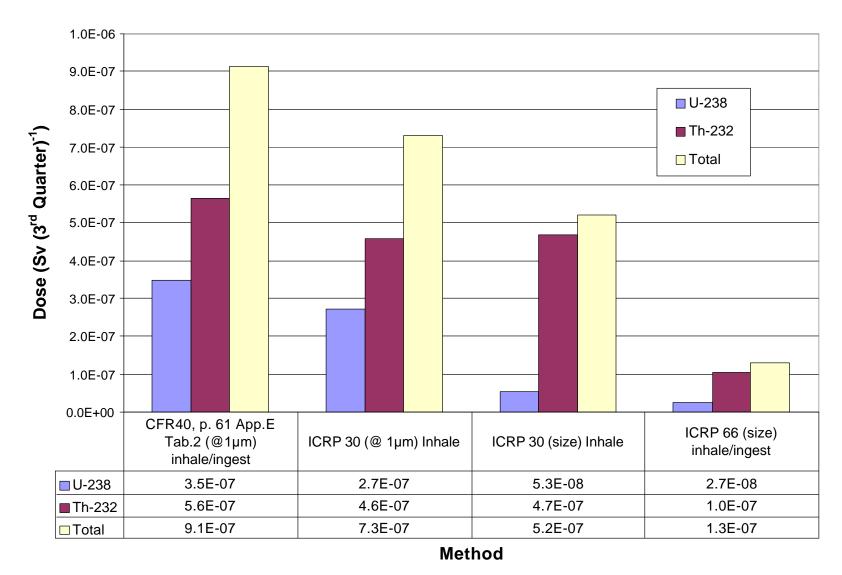


Figure 15. The third quarter dose results for ²³⁸U and ²³²Th comparing multiple methods of calculations.

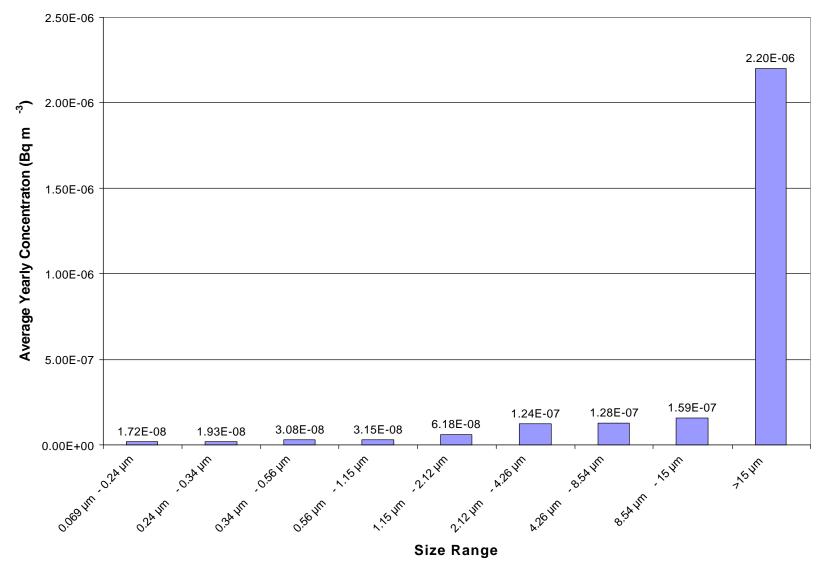


Figure 16. The average annual concentration of ²³⁸U for each size range.

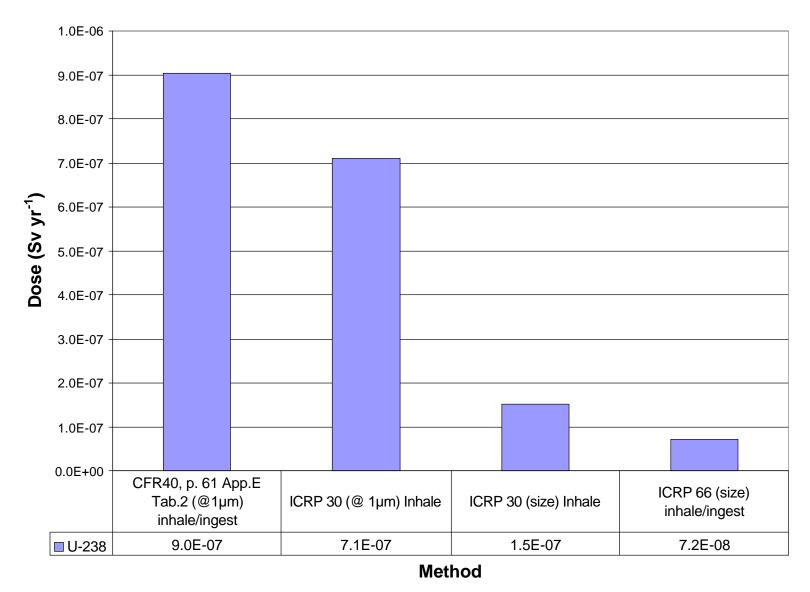


Figure 17. Annual average dose for ²³⁸U comparing multiple methods of calculation.

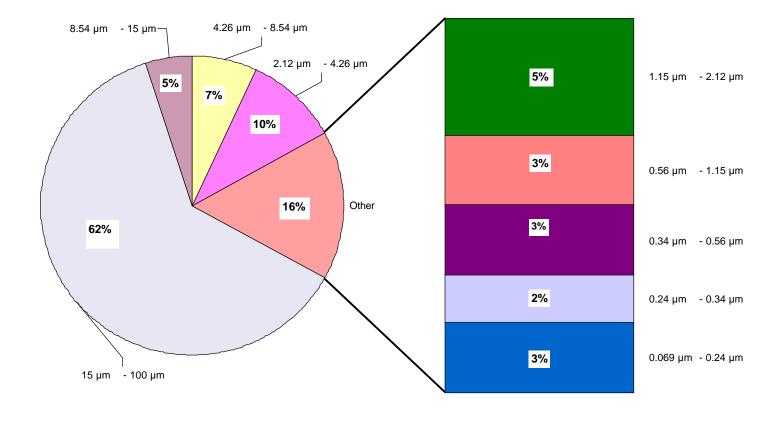


Figure 18. The average percent annual dose of ²³⁸U in each sampling size range.