An Investigation of the Degree of Equilibrium of the Long-Lived Uranium-238 Decay Chain Members in Airborne and Bulk Uranium Ore Dusts

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ABSTRACT

The degree of disequilibrium among ^{238}U decay chain members in some airborne dusts and typical ores has been established by precise radiochemical analyses. This information is necessary to evaluate the lung dose model currently used for estimating the effect of the inhalation of uranium ore dust. The particle size distributions of airborne decay chain components in dusts at one uranium mill have been investigated. Statistically significant disequilibria were observed for ^{230}Th, ^{226}Ra, and ^{210}Pb in both airborne dusts and composite ore samples. With the exception of ore from one mill in the United States, most of the daughter concentrations in powdered ore composites were within 10% of ^{238}U. In airborne dusts, the concentration of ^{226}Ra was typically below ^{238}U; the minimum ^{226}Ra concentration observed for airborne ore dusts was 56% of equilibrium. A statistically significant particle size dependence was observed for ^{226}Ra/^{238}U ratios in several airborne dusts collected at a uranium mill.

FIGURES

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INTRODUCTION AND OBJECTIVES

The regulation of occupational exposure to uranium ore dusts has been based on the uranium content of this dust, assuming that the long-lived daughters are in radioactive equilibrium with ^{238}U. It has also been assumed that elimination from the lung would proceed at the same rate for all long-lived members of the ^{238}U chain (Code of Federal Regulations, 1980). Since the technical validity of these assumptions was questionable, the U.S. Nuclear Regulatory Commission (NRC) initiated studies to permit an evaluation of the basis for new concentration limits.

Several authors (Rosholt 1964, 1965; Sill 1977; Winchester 1960) have reported a significant disequilibrium of the uranium decay chain in ores and dusts from some U.S. mining areas. In addition, only a limited number of measurements (Sill 1977) have been reported showing the particle size distributions of long-lived radionuclides in airborne ore dust at uranium mills.

To provide basic information required in evaluation of lung dose from inhalation of ore dust, the current investigation was undertaken to establish the degree of disequilibrium in ores and dusts associated with uranium milling and to determine the particle size of the long-lived uranium chain members in airborne milling dusts. The program was a co-operative effort for NRC by the Environmental Measurements Laboratory (EML) and the Pacific Northwest Laboratory (PNL) of the Department of Energy. Because of their experience in the collection of size fractionated dust samples, personnel from EML were selected to collect airborne dust samples at various locations in an operating uranium mill. Their preliminary observations and calibrations have been reported elsewhere (Knuth 1979). The radionuclide measurements were made at PNL. This is the final report documenting the PNL measurement results and the procedures used.

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A planned final phase of this program at PNL was the development
and application of a model which used the concentration, particle size,
distribution, and solubility classifications for the long-lived uranium
chain members to define the lung dose associated with inhalation of the
ore dust. At the request of the NRC staff, the program was terminated
before completion of this phase of the work.

EXPERIMENTAL

SAMPLE TYPES

Samples of two general types were received for analysis. Most consisted of Whatman-41 filter paper used for covering cascade impactor stages and for backup filters. Some of these had been saturated with white mineral oil prior to sampling in order to improve the collection properties of the impactors.

The second type of samples consisted of ground, homogenized ore dust which was collected by uranium mill operators as a part of their normal assaying operations. These samples were requested from thirteen operating mills located in six states; samples were received from eight mills. Each mill was requested to submit three samples collected on three separate working days. In addition, the mills were requested to select samples representing as wide a variety as possible of ore types or of mine locations. Additionally, the mill from which our air particulate samples were collected submitted ten samples of the ores which were being milled during the interval of air sample collections.

Two additional samples consisting of the filter media from dust mask canisters worn in that uranium mill were also submitted by EML and were analyzed.

All filter samples plus ten of the bulk ore samples from one mill were analyzed primarily by radiochemical separation and alpha particle counting techniques for each radionuclide. The remaining bulk ore dust samples were analyzed without chemical processing, using photon spectrometry with an intrinsic germanium detector.

RADIOCHEMICAL SEPARATIONS

Filter paper samples were double bagged in glazed paper envelopes immediately after collection. For analysis, the outer envelopes were removed and both the filter paper and inner envelope were analyzed intact

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to assure that any ore dust which rubbed off in transit would not be lost. The approximate uranium chain content of each sample was estimated initially by measuring the 0.61 MeV photons from ^{214}Bi using an 18 cm NaI(Tl) well counter with an anticoincidence background and Compton shield. The quantities of ^{234}Th, ^{133}Ba, and freshly prepared ^{232}U tracers added to each sample were adjusted based on the ^{214}Bi measurement, using as a criterion that each tracer should be readily measurable but not excessive in comparison with the background expected from sample radionuclides.

Sample Dissolution

Samples were wet-ashed prior to analysis to assure complete dissolution and destruction of organic matter while minimizing loss of the relatively volatile polonium and lead radionuclides. This commonly used technique has been successfully applied at PNL to biological specimens and various ores as well as the air filters.

After addition of the tracers, wet digestion with perchloric and nitric acid and small amounts of 30% hydrogen peroxide was continued until organic matter was destroyed. Samples were transferred to Teflon beakers and solid residues were attacked with a mixture of concentrated nitric, hydrofluoric, and hydrochloric acids. The silicon fluorides were volatilized by heating until the original perchloric acid started to fume. This attack was repeated two more times. Sample residues were dissolved by warming with 2M HNO_3. Then samples were filtered through a 0.45 μm membrane filter. The gross beta and gross alpha particle emission rates of the filters were measured and compared with the total activity of sample plus added tracers. If the measured emission rates exceeded 1% of the expected total, the filter was re-ashed and combined with the filtrate. The filtering process was repeated until filterable activity represented less than 1% of the total. (Normally it represented <0.1%.) Samples were adjusted to 4M HNO_3 and diluted to volume before storage in polyethylene bottles.

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The same general process was used for powdered ore samples. Blank samples of each type of filter packaged in the glazed paper envelopes were also ashed using this technique. Appropriate blank corrections were determined for each type of sample. Several reagent blanks were also analyzed for comparison with ore samples and to test for cross-contamination problems with the equipment.

Lead-210

The scheme for separating ^{210}Pb from other natural radioactive species was based on a combination of methods reported by others (Sill 1965; Gibson 1961; and Figgins 1961). Lead was separated after adding ^{212}Pb to trace the radiochemical recovery. Diethylammonium diethyldithiocarbamate extraction was used to concentrate lead from the dissolved sample. Ion exchange columns of Dowex Ag-1 resin were used to remove ^{210}Po from the separated lead. The ^{212}Pb recovery was measured with a sodium iodide detector using a multichannel gamma-ray energy spectrometer system. Polonium-208 tracer was then added and the samples were evaporated and stored in 4M HNO_3 for 138 days or longer to permit ingrowth of ^{210}Po. Then the samples were evaporated and polonium was deposited on silver discs from a 0.4 M HCl solution. The ^{208}Po and ^{210}Po content were both measured by alpha particle energy spectrometry. The ^{210}Pb activity was calculated from the ^{210}Po content determined by isotope dilution after correcting for the fractional ^{210}Po ingrowth, ^{212}Pb recovery, sample aliquot, and the appropriate blank.

Uranium-234/238 - Thorium-230

The initial separations steps for these nuclides were based on a method reported by Beasley (1965). Both uranium and thorium were co-precipitated with calcium ammonium phosphate and ferric hydroxide. Then they were extracted into a 10% (w/v) alamine-336 (dissolved in xylenes) from aluminum nitrate salting solution. Thorium was back-extracted with 7M HCl and uranium with 0.1M HNO_3. Normally the uranium was electro-plated at this point.

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If too many salts were present when the sample was evaporated, prior to electroplating the uranium, they were dissolved in 7M HCl and converted to the acetate form by repeated evaporation with acetic acid. Uranium was purified as described by Korkisch (1969) by collection on an acetate form anion exchange column before electroplating. The thorium fraction was further purified from traces of uranium and sample matrix ions by co-precipitation with neodymium fluoride and by thenoytrifluoracetone extraction (Perkins and Kalkwarf 1956) before electroplating. Alpha particle spectrometry of electroplated uranium yielded the content of ^{232}U, ^{234}U, and ^{238}U. An isotope dilution method was used for the uranium measurements, which automatically provided both correction for counting efficiency and radiochemical recovery. A beta particle count taken on the thorium mount yielded the radiochemical recovery of ^{234}Th, and the ^{230}Th was then measured by alpha particle spectrometry. Since a small fraction of the total measured ^{234}Th content was from the sample itself, a correction was made using the ^{238}U content of the sample to estimate the initial ^{234}Th. For ^{230}Th, a correction for the alpha particle spectrometer counting efficiency was also made. Standard volumetric and blank corrections were applied to both fractions.

Radium-226

Radium was concentrated and purified by coprecipitation with barium sulfate using a variation of a method suggested by Sill (1969). After dissolution with diethylene-triamine-pentacetic acid and piperiaine, barium sulfate was reprecipitated and washed. Following the barium sulfate purification steps, the ^{226}Ra was measured using a technique devised at PNL. The method involved dissolution of the sulfate precipitate by conversion to the chloride salt, and transferring the barium chloride solution to a small flask which was internally coated with a zinc sulfide scintillator, evacuated, and sealed. The flasks were stored for a month or more to permit the ingrowing ^{222}Rn to equilibrate with the ^{226}Ra in the flask. The ingrowing ^{222}Rn diffused from the solution into the

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evacuated volume of the flask. Alpha particles from the radon and its daughters were detected as scintillations of the zinc sulfide using photo-multiplier tubes.

The radiochemical recovery was determined by measuring the gamma-rays from the ^{133}Ba tracer which had been added prior to ashing the samples. The detection system was calibrated by adding standard ^{226}Ra solution directly to counting flasks and following the normal procedure.

Polonium-210

It was advantageous to also separate a ^{210}Po fraction from samples because spot checks had indicated that some of the initial ^{210}Pb measurements were in error. Subsequent investigations indicated that an inadvertent modification of the original lead separation method had been made which produced erroneously high results. Polonium-210 could be used to give a rapid measurement of ^{210}Pb because at least 1.3 years and in most cases 1.7 years had passed since the samples had been collected. No reasonable estimate of the initial disequilibrium of ^{210}Po would produce a significant error in the assumption that the ^{210}Pb content was equal to the ^{210}Po at the time of measurement. A known quantity of ^{208}Po was added to each aliquot at the time of the analyses. The analyzed aliquots were evaporated and converted to a 0.4M HCl solution. The ^{208}Po and ^{210}Po were then collected by spontaneous deposition on silver discs and ^{210}Po was determined by the isotope dilution technique using alpha particle energy spectrometry.

DIRECT PHOTON SPECTROMETRY

The ^{238}U/^{234}Th, ^{230}Th, ^{226}Ra, and ^{210}Pb were measured in most bulk ore samples by direct photon spectrometry using an intrinsic germanium detector. Fifteen grams of pulverized ore dust were mixed with two grams of cellulose powder and pressed into a pellet 5 cm in diameter by about 4.5 mm thick. Each pellet was positioned on an intrinsic germanium detector and the photon spectrum collected by counting overnight with a multichannel analyzer. A second pellet was also prepared but was counted

for a shorter interval to check the uniformity of the samples. The photon spectrometer was calibrated using aliquots of pitchblende with well established radionuclide concentrations. Varying portions of pitchblende were mixed with blank sand (dunite) and cellulose. Each was then pressed into a pellet, and counted with the spectrometer.

The sensitivity limitations of the direct counting method are estimated in Table 1. For comparison purposes, the estimated sensitivities of our chemical separation techniques are also shown. Although these limitations based on counting statistics indicate that it should be possible to directly measure most long-lived uranium chain members to an accuracy better than ±3% (relative standard deviation), cross comparisons with other methods indicate that accuracies for direct counting are in the area of 5% to 10% because of sample density and uniformity variations.

QUALITY ASSURANCE PROGRAM

CALIBRATIONS

All radioisotope measurement systems have been calibrated using radiation sources traceable to the National Bureau of Standards or to a pitchblende reference standard (Sill, 1977) supplied by the Idaho National Engineering Laboratory (INEL) of the DOE. Analytical balances were routinely tested with calibrated weight sets traceable to the National Bureau of Standards. Volumetric flasks were certified by the manufacturers as passing Bureau of Standards criteria. Volumetric pipettes were retested regularly by weighting replicate pipetted aliquots of distilled water using an analytical balance. Pipettes having an error of greater than 1% were adjusted or discarded.

REFERENCE SAMPLES

To evaluate the effectiveness of our control programs, and the accuracy of our methods, referee samples submitted by INEL were analyzed and the results were reported to INEL. The first referee sample submitted was the pitchblende reference standard which was reported to contain 6060 dis/min of ^{238}U, essentially in equilibrium with its daughters. Two portions of this ore were dissolved for analysis. Duplicate aliquots of each portion were taken after dissolution. The results of these analyses are shown in Table 2. It can be seen that the relative standard deviations of replicates ranged from 0.7% to 3.6% and that there was no significant bias relative to the known concentrations of the standard.

Since the first test of our techniques involved a standard with concentrations known in advance, a second set of standards was prepared by INEL. Three samples were submitted for which the radionuclide concentrations were unknown to us. The first of the samples had been prepared as a dilution of the pitchblende standard with blank soil. The second was an ore sample which had been analyzed at INEL, and the third was a dilution of the second with blank soil. Table 3 shows a comparison of the PNL

results with those of INEL. The agreement was generally within the sample-to-sample variability indicated by the replicate analyses made by INEL. The PNL measurements were normally within two standard deviations of the INEL measurements.

One comparison which can be made to show the reproducibility of measurements taken at PNL is to compare the ratio of results for the second and third unknown with the dilution factor used by INEL. This comparison shown in Table 4 demonstrated that, excepting ^{226}Ra, the methods were not significantly biased relative to the precision estimates. Later tests demonstrated that the reproducibility inherent in the ^{226}Ra procedure was about 4% relative standard deviation exclusive of propagated counting errors. Thus, the apparent bias for that method was also insignificant.

In the case of those samples assayed by direct gamma-ray spectrometry, some of the inherent errors were related to the uniformity of the prepared sample configurations in addition to the usual counting statistics. Significant changes of density or composition can alter the self-absorption of low energy photons used in analysis of the ^{238}U decay chain. Thorough mixing and uniform pellet formation were necessary to minimize the variance with a single ore sample. Calibration curves were prepared with varying proportions of the standard pitchblende diluted with dunite. When the three unknown samples used to evaluate PNL's chemical methods were analyzed by direct photon spectrometry, we found that the least bias relative to the chemical methods was obtained when the efficiency factors had been determined from standards diluted to 30 weight percent or less of pitchblende at which point the efficiency no longer changed with composition. Apparently that standard, which was very high grade ore, was sufficiently dense and contained sufficient high atomic weight elements to cause a different self-shielding than the more typical ores being analyzed. In Table 5, a comparison of direct photon spectrometry results with the results of chemical separations is shown. It can be seen that the agreement for those samples was within 12.5% for all measurements

except the ^{226}Ra. The ^{226}Ra results were all biased high at the time of this comparison.

A homogenized ore sample was pressed into three pellets to determine the reproducibility of the mounting technique. Recounts were taken on one of the pellets to establish the stability of the counting geometry. The relative standard deviations for three pellets was as follows: ^{238}U - 6%, ^{230}Th - 6%, ^{226}Ra - 5%, and ^{210}Pb - 8%. The relative standard deviations from recounting a single pellet were: ^{238}U - 1.4%, ^{230}Th - 2.%, ^{226}Ra - 2.8%, ^{210}Pb - 4.5%.

The counting statistics for these measurements accounted for less than 1% of the variation except for ^{230}Th which had a relative standard deviation of 3% from this source. Thus, the reproducibility of counting geometry accounted for a standard deviation of no more than 4.5%, and pellet inhomogeneity caused less than 8% variation. Our experience using this method for a large number of samples is in agreement with this limited study. The reproducibility is typically in the range of 5% to 10% relative standard deviation, when it is not limited by counting statistics.

STATISTICAL EVALUATIONS OF MEASUREMENT RESULTS

In the discussions of our quality assurance experiments, we showed the results of intercomparison studies performed prior to the analyses of airborne dust samples. In general, the methods were capable of producing unbiased results in comparison with the small propagated errors which resulted primarily from the counting statistics of our measurements. However, since we have received several replicate samples, collected simultaneously within about a meter of one another, we could test for the overall errors from sampling and analyses. This has the advantage of detecting any variations in measurements which might occur from changes in the analytical methods with time and from personnel changes. In addition, the real effects of variations in sampler characteristics are included. We examined replicate sample variations for the cases of both the overall disequilibrium measurements and for their particle size dependence.

For the overall disequilibrium measurements, we have taken the total radioactivity of each radionuclide on all stages and filters collected simultaneously at the same location prior to determining the ratio. This process will tend to reduce random stage-to-stage and sampler-to-sampler variations. To check the precision of these ratios, the sum of each radionuclide in the stages of the individual impactors was used to determine ratios for each total impactor sample, along with the ratio determined for the reference filter. These give several sets of three replicate determinations for statistical comparison at the secondary crusher area of one uranium mill and sets of two for areas which had only one impactor sample with its reference filter. The variations of the replicate ratios were pooled for all sampling periods after we demonstrated the homogeneity of the variances using a modification of "Bartlet's" test. By pooling, the degrees of freedom of our standard deviation estimates for the overall disequilibrium measurements were increased to 18. From those estimates, a 95% confidence interval for the overall total activity ratio of the sum of all replicate samples was derived using the student "t" distribution.

Since we had the propagated counting errors for our original concentration measurements, we could further propagate them to produce standard deviation estimates for the total activity ratios determined as described above and compare the propagated errors with the actual pooled standard deviation measurements. Table 6 compares standard deviations determined by each method. It can be seen that the actual replication errors are somewhat larger than those from the propagation process. This is to be expected since they contain additional sources of variation from sampling and analysis. The largest relative standard deviation was about 8% for the $^{210}Pb/^{226}Ra$ ratio. It is apparent that the variability of ^{210}Pb measurements was considerably higher than can be explained by propagation of known error source terms. This was partly caused by analytical variations resulting from the behavior of polonium in solution. After most of the analyses had been performed, we discovered that the ^{208}Po tracer used in this analysis had partly formed filterable species and was not truly homogeneous. Because of this, for ^{210}Pb a disequilibrium must exceed $\pm15\%$ of the ^{238}U content to be considered significant relative to the replication error.

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The same techniques were used for evaluating the variability of ratios measured on individual impactor stage results. The data at the secondary crusher area gave duplicate sets of impactor stages for which the replication errors could be determined. A variance was calculated for each stage for each sampling interval. Those data were pooled to obtain estimates of the replication errors, including those involved with sampling. The standard deviations for single measurements were as follows: $^{234}U/^{238}U$ - 0.025; $^{230}Th/^{238}U$ - 0.048; $^{226}Ra/^{238}U$ - 0.069; $^{210}Pb/^{238}U$ - 0.088; $^{210}Pb/^{226}Ra$ - 0.054. Note that these ratios are based on a pooled estimate which assumes a homogeneous variance when, in fact, there was a dependence of a part of the variance of each stage on the counting rate of the radionuclides deposited there. In most cases other than ^{234}U, the variations from sources other than counting errors tend to dominate the variance when the activity on all stages in an impactor are totaled, since the total activity is relatively large. Thus, statistical tests based on these values were relatively insensitive to the observed counting rates, making the pooled estimates valid.

RESULTS AND DISCUSSION

AIRBORNE DUST

Analyses were performed on 18 sets of cascade impactor stages and a total of 26 general air filters collected at a uranium mill in New Mexico. Located in the Appendix is a tabulation of the measured radionuclide concentrations and estimates of the relative standard deviations (%) propagated from measurement variations associated with the radiation counting process for samples and standards. A separate small group of filtered dust samples was collected by PNL at a different mill in New Mexico to provide specimens for methods testing.

We have evaluated all results in terms of the measured disequilibrium of the long-lived daughters from ^{238}U. Interpretations based on the particle size characteristics and sampling locations are not covered in depth since they can best be interpreted by EML who performed the sampling program and particle size analyses. Two questions that we will attempt to answer are:

1. Was there a significant disequilibrium of any of the long-lived uranium decay chain members in airborne dusts and ores?

2. Was the disequilibrium related to the particle size of airborne dusts?

DEGREE OF EQUILIBRIUM

The ratios of the daughters to ^{238}U concentrations for each combined impactor sample or general air filter represent the overall degree of equilibrium in airborne dusts for the location sampled and sampling time. Three cascade impactor samples were collected simultaneously. Gross general air filter samples were also collected at the same locations as the impactors. Most of the sets contained duplicate impactor samples plus a reference filter all of which had been stationed within about 1.3 meters of one another. A third impactor and its reference filter were stationed in another area not adjacent to the first sets. In addition, individual

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general air filter samples were collected at various places around the uranium mill.

Table 7 shows the results for thirteen series of particulate samples where replicates were collected. It can be seen that there was only one instance of a significant disequilibrium of ^{234}U, and at 97.5% of the ^{238}U content, that result was probably an outlier and not truly significant. Most ^{234}U concentrations averaged within 1% of the ^{238}U. There were five occasions when a significant disequilibrium of ^{230}Th was noted. In two samples the ^{230}Th was less than 90% of the ^{238}U. Most of the ^{230}Th measurements were within 5% of the ^{238}U content. All of the ^{226}Ra and ^{210}Pb measurements showed a significant disequilibrium from ^{238}U. In all but one sample the ^{226}Ra was below the ^{238}U content; the high value was associated with a ^{230}Th measurement which was higher than 1.0. High ^{226}Ra values were also measured in some bulk ore samples from this mill. Most radium concentrations were in the range from 75% to 90% of the ^{238}U, as were the ^{210}Pb concentrations. Two sample sets had relative concentrations of about 60% of uranium for both ^{226}Ra and ^{210}Pb. Since the relatively short half-life of ^{210}Pb allows a more rapid re-establishment of equilibrium with ^{226}Ra than for the other tabulated ratios, the ratio of ^{210}Pb/^{226}Ra gives indications of relatively recent events which might cause disequilibrium. Although the ^{210}Pb content of the dusts was normally within 10% of the ^{226}Ra content, in most cases the ^{210}Pb concentrations were low. They were significantly low in seven of the thirteen cases. A probable cause for these low ratios is the partial loss of the ^{222}Rn parent of ^{210}Pb from the ore body for an extended interval during the past 50 years. One significantly high ^{210}Pb/^{226}Ra ratio was observed.

As shown in Table 8, the general air filters (samples 126-136) collected in several other locations around the mill, and two respirator filters (samples 137 and 138) worn at the mill, showed, in most cases, the same range of disequilibrium as was observed in the cascade impactor samples. Three notable exceptions (samples 127, 132, and 136) were apparently collected in a yellowcake handling area since the proportions

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of ^{230}Th, ^{226}Ra, and ^{210}Pb were very low. Other than the yellowcake areas and sample 126 which appears to be a spurious result, ^{210}Pb concentrations were again lower than the ^{226}Ra. In two samples the ^{210}Pb was more nearly in equilibrium with ^{238}U than was ^{226}Ra. This indicates that the disequilibrium of ^{226}Ra in those dusts is of relatively recent origin.

In summary, we have measured in some of the airborne dust from this mill a significant degree of disequilibrium for all long-lived ^{238}U daughters except ^{234}U. In all samples the ^{226}Ra and ^{210}Pb are in significant disequilibrium. Only one case of a significantly positive disequilibrium was noted. Thus, most of these samples indicated that the dose from the inhalation of airborne dust would be lower than that estimated from the ^{238}U content, assuming secular equilibrium.

PARTICLE SIZE RELATIONSHIPS

Although a rigorous analysis of the particle size distributions for each radionuclide in air samples is beyond the scope of this report, we have attempted to test the significance of observed relationships by means of analysis of variance techniques.

To evaluate the hypothesis that there is no particle size dependence in the ratios of daughter radionuclides to ^{238}U, a series of one-way analysis of variance tests were performed for each duplicate set of cascade impactor samples. For each daughter, the variance between the impactor stages of the radionuclide ratios was compared with a pooled estimate of the variations of duplicate measurements from within each stage. When the ratio of these errors ("F" values) exceeded the expected values at the 1% probability level, the hypothesis was rejected and the result interpreted to indicate a significant functional relationship between particle size and activity ratio. The use of a 1% level rather than 5% insures that possible inhomogeneity of the pooled variances will not prejudice our hypothesis tests.

The results of these tests for the secondary crusher area, where duplicate impactor samples were collected, are shown in Table 9. Most of the $^{234}U/^{238}U$ samples showed a significant dependence on the particle size in this test. However, this appears to be an artifact resulting from the interaction of rounding errors where numerous cases were rounded to values of nearly 1.00. The high precision made the test overly sensitive to small variations. There was only one case of a ^{230}Th disequilibrium which was significantly related to particle size. The $^{226}Ra/^{238}U$ ratios showed a significant dependence on particle size in three of the five tests. This dependence on particle size could not be demonstrated for the ^{210}Pb daughter. This difference was only partly caused by the generally poorer measurement precision for ^{210}Pb. The high degree of disequilibrium of ^{226}Ra along with a unique particle size dependence suggests that ^{226}Ra was separated from the bulk of the original parent particles and redeposited over different substrate components of the ore. The failure of ^{210}Pb to follow the size dependence of ^{226}Ra suggests that this process has occurred only relatively recently in the history of the ore body.

To evaluate the size dependencies of disequilibrium at locations where only a single impactor set was collected at a given time, we compared the pooled replication error estimates from the duplicate sets taken at the secondary crusher with the stage-to-stage variances of each single sample. For samples with I.D. number above 106, a significant part of the measurement variation results from counting statistics, and the pooled variance estimates for replication error are, no doubt, too low; this gives rise to the large number of significantly large ratios. Nonetheless, the analyses of variance results also shown in Table 9 demonstrate relationships similar to those observed at the secondary crusher.

A graphical presentation of these data is helpful to confirm the statistical findings. Figure 1, which is typical of many of the impactor samples, shows the measured ratios of each daughter to ^{238}U for one

cascade impactor set collected at the secondary crusher on 8/23/77 from 1310 to 1555. The size dependence can be inferred since the median particle size retained on the impactor stages decreases with increasing stage number. Thus, the significant relationship between the $^{226}Ra/^{238}U$ ratios and particle size can be seen to result from an increasing proportion of ^{226}Ra as the particle sizes decrease. On the other hand, the statistically significant particle size dependence of the $^{234}U/^{238}U$ ratios is not apparent in the figure. In samples displaying this size distribution characteristic, when the $^{226}Ra/^{238}U$ ratios were increasing with decreasing particle size, the $^{210}Pb/^{238}U$ ratios tended to be significantly below them and showed less particle size dependence.

The relationship of $^{226}Ra/^{238}U$ ratios to particle size changed several times at the secondary crusher, and was somewhat different than the distribution observed at the primary crusher. Cases where ratios significantly increased or decreased with particle size or where the proportions changed directions were observed. Figure 2 typifies another set of distributions observed for a sample taken at the secondary crusher a day after the set shown in Figure 1 was collected. In this set, when the $^{226}Ra/^{238}U$ ratios were relatively independent of particle size, they were significantly below 1.0 and the $^{210}Pb/^{238}U$ ratios were not significantly different from them.

These patterns can be explained by processes which occurred at different periods in the history of the various uranium ores being milled. Most of them must result after a portion of the ^{226}Ra is dissolved in the ore body. The most common characteristic of an increasing proportion of ^{226}Ra on finer particulates would occur if a redeposition of dissolved ^{226}Ra had a surface area dependence or if a fine particle substrate had a chemical or physical ion exchange affinity for ^{226}Ra. This assumption also implies that the size distribution of airborne particulates is related to that of the accreted particulates making up the ore. The occasional failure of ^{210}Pb to follow the patterns of ^{226}Ra suggests that this solution and deposition process may have occurred only relatively

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recently - within the past fifty years or so - in those ore samples. The very low ratios measured for some samples as in Figure 2 suggest an older or longer-term process in which ^{226}Ra is dissolved and removed from the ore without redeposition. Perhaps in those cases the ore no longer contains the necessary substrate for redeposition. A characteristic supporting the recent process pattern was also noticed in some general air filters which showed a severe loss of ^{226}Ra. In those cases, the ^{210}Pb content of the dust tended to be higher than the ^{226}Ra content. Since the principal occurrence in the recent history of the ore body is the opening of the uranium mines, these findings suggest the possibility that ^{226}Ra was being dissolved and transported by ground water which flowed through the ore in the typical mine dewatering process. It is possible that, for some ores, the missing ^{226}Ra will be present in the mine dewatering ponds.

Airborne dusts were also collected by PNL at a different uranium mill in New Mexico. These data resulted from a preliminary evaluation of our procedures and are included in this final report primarily to demonstrate similarities and differences between the particle size dependence at the two mills. In Table 10 the composition of the measured radionuclides and ratios of the progeny to ^{238}U are summarized. It can be seen that in these samples the ^{226}Ra also shows the largest degree of disequilibrium of the daughters. Here, however, the patterns all show a slightly decreasing proportion of ^{226}Ra with decreasing particle size. Judging by the precision estimates, there were no significant instances of ^{230}Th disequilibrium. In these samples, the proportion of ^{210}Pb was generally higher than ^{226}Ra, indicating that at least a part of the ^{226}Ra disequilibrium is of recent origin.

The attempt we have made to interpret the particle size relationships demonstrated in these samples is, necessarily, incomplete since we lack specific information about the sampling program. The interaction of sampling and particle generation at the mill may have contributed significantly to these results in ways that are difficult to predict. Particle size distributions of the ^{238}U decay chain members were not always

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identical when simultaneous samples were collected at the various loca-
tions throughout the crushing areas of the mill. Some distributions also
changed from time to time at a given location. Although we have not es-
tablished precise particle size information from the samples, the data
demonstrate that significant size dependencies do exist, and that their
nature is quite complex. The effect of these dependencies to lung dosim-
etry will depend on whether or not the size parameters used in lung
deposition models will be significantly modified.

Ore Dust Samples

Ten samples of uranium ore dust being processed at the mill at the
time of air sample collections were analyzed using the wet chemical
methods to determine ^{238}U, ^{234}U, and ^{210}Pb. Photon spectrometry was used
to determine ^{230}Th and ^{226}Ra when it was discovered that there was the
possibility of a systematic pipetting error in the wet chemical analysis
of these nuclides in this set of samples. The results of the analyses
numbers 139 through 148 can be found in the Appendix. The measured
ratios of daughters to ^{238}U are shown in Table 11. Values differing
significantly from 1.00 are shown with an asterisk. No cases of signifi-
cant disequilibrium were noted for ^{234}U or ^{230}Th, but there were several
for ^{226}Ra and ^{210}Pb.

It is of interest to compare the radioactivity ratios determined
for bulk ore samples with those for airborne dusts shown in Tables 7 and
8. One can note that all but one of the $^{230}Th/^{238}U$ ratios in ore were
greater than 1.00. Taken as a set, the average ratio was significantly
greater than 1.00. This reflects a small systematic bias known to be
possible for the instrumental methods. Most of the $^{226}Ra/^{238}U$ ratios
were higher in the bulk ore samples than in air samples collected at the
time of ore processing, and often showed no significant disequilibrium
from ^{238}U. This finding indicates that the airborne dust does not neces-
sarily reflect the average composition of bulk ore, and indicates that
the particles suspended in air may reflect a fraction of the ore

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which is easily pulverized. It may also indicate that a substantial re-suspension of dusts occurred about the milling area, caused by local activities. The former conclusion is supported by the unique particle size dependence which we observed for some ^{226}Ra/^{238}U ratios in airborne dust.

It can also be noted that the ratios for airborne dusts at the primary crusher area are generally closer to those of the bulk ores than ratios measured at either the secondary crusher area or the lower level of the secondary crusher.

The ^{210}Pb/^{238}U ratios in the bulk ore tended to be slightly lower than the corresponding ^{226}Ra/^{238}U ratios. This is probably an indication of the continual loss of a fraction of ^{222}Rn from the ore body in the last 50 years.

These results indicate that if the sampling conditions at this mill are typical, a dose rate derived from analyses of uranium chain members in bulk ore will give a slight over-estimate since the proportions of ^{226}Ra and ^{210}Pb, and possibly ^{230}Th, were less in airborne dust than in the bulk ore.

Samples of pulverized uranium ore were received from eight uranium mills located in five states. Three samples were requested from each mill. They were selected by mill operators from their normal assay samples to represent a broad cross-section of the types of ore being processed in their mills. The measured degree of equilibrium between the long-lived uranium decay chain members in these samples represents a snapshot of the majority of the United States industry. The radio-nuclide concentrations reported in Table 12 were measured by direct photon spectrometry using intrinsic germanium detectors. Duplicate aliquots of each ore sample were measured. The variations shown result from both the uniformity of the constituents in each aliquot and from self-absorption variations inherent in the counting process.

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The ratio of the averaged daughter radionuclide concentrations to that of ^{238}U is shown in Table 13. A number of the samples showed significant degrees of disequilibrium relative to the precision of the measurements. The most striking incidence of disequilibrium was at Mill A in Wyoming where all daughters were significantly lower in concentrations than the ^{238}U. Lung dose estimates based on the assumption of equilibrium concentrations of uranium daughters would be higher than the actual dose.

A second common factor of most samples was the slightly low $^{226}Ra/$ ^{238}U ratios. This phenomenon possibly caused by leaching of ^{226}Ra from the ore bodies would also result in lower doses than is predicted assuming that ^{226}Ra is in equilibrium with ^{238}U. It is interesting to note that the ^{210}Pb concentration was generally higher than the ^{226}Ra, indicating that the ^{226}Ra disequilibrium is of relatively recent origin. This might have occurred during mine dewatering.

Although there were some instances where the concentration of ^{210}Pb or ^{230}Th exceeded that of ^{238}U significantly relative to the measurement precision, our experience with direct gamma-ray measurements for those nuclides indicates that detection efficiencies are sensitive to variations in the density and average atomic number of the ore so that small biases of about 5% - 10% can occur. Thus, it cannot be established that their enrichment is real in these samples.

These measurements indicate that with the exception of samples from a single milling area in Wyoming the disequilibrium among the long-lived ^{238}U decay chain members was less than 20% among ores from eight mills in five states representative of most of the United States production facilities. In fact, only eight measurements from the remaining mills showed a disequilibrium of any nuclide greater than 10%. Thus, the use of a lung dose model which assumes complete equilibrium among the decay chain members will produce relatively small errors in most cases unless the

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dust aerosols formed during milling are significantly different from
the bulk ore composition. The geologic process which produced the severe
disequilibrium in the small milling area of Wyoming appears to be rela-
tively unique in this country.

SUMMARY AND CONCLUSIONS

Concentrations of the long-lived members of the ^{238}U decay chain have been measured in uranium ore samples from major mining areas of the United States and in airborne ore dusts from two uranium mills in the New Mexico area. Significant degrees of disequilibrium were found in both homogenized ore samples and airborne dusts. There was a small but significant difference in the composition of respirable airborne dusts and the bulk ore dust at one uranium mill. The concentrations of ^{226}Ra were significantly below those of ^{238}U in most samples, being as low as 50% of the equilibrium concentration in some airborne dusts. In only a relatively few instances were the concentrations of the progeny significantly higher than ^{238}U. Thus, the use of the ^{238}U content of ore to estimate the lung dose from inhalation of the dust in a mill will generally produce an over-estimate if one assumes a total equilibrium among the decay chain members. The magnitude of these errors will be considerably less than a factor of two in most instances since the disequilibrium of the major constituent ^{230}Th is relatively insignificant in almost all ore samples analyzed except those from a single region of Wyoming.

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TABLE 1

Sample Quantities Needed for Analysis of Uranium Chain Members

Radionuclide			Chemical Analysis				Direct Photon Spectrometry		
			Weight Required (mg)[1,4]					Weight Required (mg)[1,4]	
	Recovery %	Counting Efficiency %	1% Std. Dev.	3% Std. Dev.	10% Std. Dev.	Energy, keV	Efficiency %	3% Std. Dev.	10% Std. Dev.
^{238}U	88	33	84	8.4	0.84	63(^{234}Th)	0.49	400	70
^{234}U	88	33	84	8.4	0.84	52	0.14	(3)	(3)
^{230}Th	78	33	95	9.5	0.95	68	0.055	3000	800
^{226}Ra	84	175	16	1.6	0.16	186	0.30	400	130
^{210}Pb	44(2)	33	170	17.0	1.7	46	0.54	200	50

NOTES:

1. 0.2% U_3O_8 ore, 1000-minute count, 1/3 aliquot.

2. Combination of 95% Pb recovery, 92% Po recovery, and 50% ^{210}Po ingrowth.

3. Normal accuracy is at least ∿20% at any weight.

4. Weight required to achieve the indicated % relative standard deviation.

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TABLE 2

Replicate Analyses of Pitchblende Standard

dis/min per 100 mg

Sample No. [1]	^{238}U	^{234}U	^{230}Th	^{210}Po	^{210}Pb	^{226}Ra
PBA-1	615	615	654	594	590	617
-2	620	625	646	616	618	633
PBB-1	615	617	637	597	601	583
-2	629	623	602	576	603	603
$\bar{x}$ ± standard deviation	620 ± 7	620 ± 5	634 ± 23	596 ± 16	603 ± 12	609 ± 21
Relative standard deviation	1.1%	0.7%	3.6%	2.7%	2.0%	3.5%
Ratio to accepted value [2]	1.02 ± .01	1.02 ± .01	1.05 ± .04	0.98 ± .03	1.00 ± .02	1.00 ± .035

NOTES

1. PBA, PBB are two portions of the pitchblende standard which were dissolved and analyzed in duplicate.

2. Accepted value is from Claude W. Sill of the Idaho National Engineering Laboratory who supplied this standard. Extensive analyses have shown the pitchblende to be essentially at equilibrium with ^{238}U at 606 ± 4 dpm per 100 mg.

TABLE 3

Analysis of Referee Samples by Pacific Northwest Laboratory (PNL)
and the Idaho National Engineering Laboratory (INEL)

Sample I.D.[1]	Nuclide	PNL dis/min per gram ±1 Std. Dev.	INEL dis/min per gram ±1 Std. Dev.	Ratio: PNL/INEL ±1 Std. Dev.
1	^{238}U	561 ± 10	563 ± 4 [2]	.996 ± .019
	^{234}U	560 ± 10	563 ± 4	.995 ± .019
	^{230}Th	549 ± 14	563 ± 4	.975 ± .026
	^{226}Ra	544 ± 8	563 ± 4	.966 ± .016
	^{210}Pb	584 ± 16	563 ± 4	1.037 ± .029
	^{210}Po [3]	557 ± 6	563 ± 4 [5]	.989 ± .012
2	^{238}U	1390 ± 33	1368 ± 24 [4]	1.016 ± .030
			1345 ± 24	1.033 ± .030
	^{234}U	1314 ± 32	1278 ± 22	1.028 ± .031
			1218 ± 22	1.078 ± .032
	^{230}Th	2122 ± 48	2133 ± 19	.995 ± .024
			2128 ± 18	.997 ± .027
	^{226}Ra	2344 ± 42	2211 ± 26	1.060 ± .023
			2181 ± 26	1.075 ± .023
	^{210}Pb	2055 ± 48	1940 ± 26	1.059 ± .028
			1737 ± 25	1.183 ± .032
	^{210}Po	2004 ± 32	1940 ± 26 [5]	1.033 ± .021
			1737 ± 25	1.153 ± .024
3	^{238}U	626 ± 21	591 ± 10 [4]	1.059 ± .040
			581 ± 10	1.077 ± .041
	^{234}U	571 ± 19	552 ± 10	1.034 ± .042
			526 ± 10	1.086 ± .042
	^{230}Th	920 ± 18	921 ± 8	.999 ± .021
			919 ± 8	1.001 ± .021
	^{226}Ra	941 ± 14	955 ± 11	.985 ± .019
			942 ± 11	.999 ± .019
	^{210}Pb	920 ± 26	837 ± 11	1.099 ± .034
			750 ± 11	1.227 ± .039
	^{210}Po	877 ± 10	837 ± 11 [5]	1.048 ± .018
			750 ± 11	1.169 ± .022

NOTES:

1. Sample 1 was a dilution of standardized pitchblende ore. Dilution was with subsurface earth material of known low uranium decay chain content.

   Sample 2 was a sample of ore dust collected at a uranium mill.

   Sample 3 was a dilution of sample 2 with subsurface earth material. (sample 3 = 0.4319 x sample 2 concentrations)

2. Precision by INEL for sample 1 is based on repeated determination of the uranium content of the ore and the assumption of radioactive equilibrium of the daughters of uranium. It does not indicate the precision of measuring the individual nuclides.

3. Polonium-210 analyses were performed in addition to ^{210}Pb because it was necessary to shorten the normal ingrowth period used in the ^{210}Pb procedure. The samples were known to have been stored for more than one year and the ^{210}Po should be a reliable indicator of the ^{210}Pb content.

4. Two fractions of samples 2 and 3 were analyzed by INEL and the individual results of both are reported.

5. No analyses for ^{210}Po were performed by INEL. Lead-210 results are assumed to be the same as ^{210}Po for comparison purposes.

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TABLE 4

Comparison of Related Referee Sample Results

Nuclide	Sample 3/(Sample 2)(0.4319)
^{238}U	1.043 ± .043
^{234}U	1.006 ± .041
^{230}Th	1.004 ± .030
^{226}Ra	0.929 ± .021
^{210}Pb	1.037 ± .038
^{210}Po	1.013 ± .020

NOTE: Sample 3 was prepared by diluting Sample 2 with soil
known to be low in uranium chain content. The dilution
factor was Sample 3 = 0.4319 x Sample 2.

TABLE 5. Results From Direct Photon Spectrometry
and Chemical Analysis

Sample I.D.	Nuclide	Direct Spectrometry dis/min per gram	Ratio to Chemical Analysis PNL	INEL [1]
1	^{238}U	575	1.025	1.021
1	^{230}Th	541	0.985	0.960
1	^{226}Ra	620	1.140	1.101
1	^{210}Pb	564	.966	1.002
2	^{238}U	1415	1.018	1.044
2	^{230}Th	2126	1.002	0.998
2	^{226}Ra	2614	1.115	1.190
2	^{210}Pb	2061	1.003	1.121
3	^{238}U	608	0.971	1.038
3	^{230}Th	903	0.982	0.982
3	^{226}Ra	1153	1.225	1.215
3	^{210}Pb	882	0.959	1.125

NOTES:
1. Average of reported values.

TABLE 6. Comparison of Radioactivity Ratio
Error Estimates

Method of Error Estimate	Standard Deviations				
	$\frac{^{234}U}{^{238}U}$	$\frac{^{230}Th}{^{238}U}$	$\frac{^{226}Ra}{^{238}U}$	$\frac{^{210}Pb}{^{238}U}$	$\frac{^{210}Pb}{^{238}U}$
Propagation of Counting Statistics [1]	0.013	0.018	0.024	0.016	0.039
Pooled Replication Error [2]	0.016	0.035	0.046	0.054	0.075

NOTES:

1. Errors include those derived from the statistical uncertainty of radiation counting of samples and standards.

2. Standard deviations were determined by pooling the variance measurements of thirteen sets of replicate samples.

TABLE 7. Radioactivity Ratios in Airborne Dusts at Various Locations in a Uranium Mill

Location	Date-Time			Radioactivity Ratio [1,2,3,4]				
				$\frac{234U}{238U}$	$\frac{230Th}{238U}$	$\frac{226Ra}{238U}$	$\frac{210Pb}{238U}$	$\frac{210Pb}{226Ra}$
Secondary	8/23/77	1310	- 1533	1.001±0.019	0.944±0.042*	0.896±0.055*	0.779±0.066*	0.869±0.091*
Crusher	8/23-24/77	1705	- 0730	0.991±0.019	0.998±0.042	0.869±0.055*	0.785±0.066*	0.903±0.091*
	8/24/77	0905	- 1355	0.984±0.019	0.995±0.042	0.590±0.055*	0.578±0.066*	0.980±0.091
	8/24/77	1440	- 1950	1.007±0.019	0.951±0.042*	0.897±0.055*	0.811±0.066*	0.904±0.091*
	8/24-25/77	2055	- 0745	0.993±0.019	0.963±0.042	0.752±0.055*	0.757±0.066*	1.006±0.091
Primary	8/23/77	1320	- 1605	0.975±0.023*	1.036±0.052	1.140±0.068*	0.898±0.080*	0.788±0.111*
Crusher	8/23-24/77	1710	- 0735	0.993±0.023	1.014±0.052	0.759±0.068*	0.861±0.080*	1.134±0.111*
	8/24/77	0915	- 1305	1.022±0.023	0.950±0.052	0.746±0.068*	0.713±0.080*	0.956±0.111
Lower Level								
Secondary	8/24/77	1350	- 1950	1.004±0.023	0.934±0.052*	0.728±0.068*	0.631±0.080*	0.865±0.111*
Crusher	8/24-25/77	2050	- 0730	0.989±0.023	1.051±0.052	0.614±0.068*	0.663±0.080*	1.080±0.111
Near Ore Bin	8/25/77	0930	- 1425	0.998±0.023	0.887±0.052*	0.849±0.068*	0.725±0.080*	0.853±0.111*
Near Primary Crusher Belt	8/25/77	0920	- 1400	1.008±0.023	0.953±0.052	0.751±0.068*	0.696±0.080*	0.928±0.111
Sample Prep Room B	8/25/77	0900	- 1400	0.998±0.023	0.833±0.052*	0.745±0.068*	0.735±0.080*	0.987±0.111

NOTES:

1. Samples collected by EML.

2. Radioactivity ratios were determined after summing the activity of each radionuclide over all impactor stages and filters collected simultaneously at a given site.

3. Precision estimates are at the 95% confidence level, based on the pooled variance determined from all replicated measurements for the ratio of each radionuclide to ^{238}U.

4. Asterisks denote cases of significant disequilibrium at the 95% confidence level.

TABLE 8. Uranium Decay Chain Disequilibrium in General Air Filters Collected at a Uranium Mill

	Radioactivity Ratios[1,2]				
Sample I.D.	$^{234}U/^{238}U$	$^{230}Th/^{238}U$	$^{226}Ra/^{238}U$	$^{210}Pb/^{238}U$	$^{210}Pb/^{226}Ra$
126	1.04	0.98	0.95	1.49*	1.57*
127	0.98	0.0097*	0.0026*	0.0091*	3.44*
128	1.01	1.00	1.03	0.98	0.95
129	1.02	1.00	0.90	0.89	0.98
130	0.96	0.99	0.91	0.95	1.04
131	0.98	1.04	0.61*	0.71*	1.16*
132	0.99	0.023*	0.016*	0.040*	2.53*
133	0.98	0.96	0.56*	0.80*	1.42*
134	1.00	0.94	0.87	0.81*	0.94
135	1.00	0.92	0.86*	0.80*	0.94
136	0.99	0.008*	0.0038*	0.0015*	0.34*
137[2]	1.03	0.99	0.77*	0.76*	0.99
138[2]	0.97	0.86*	0.70*	0.66*	0.94
95% Confidence Limits	±0.05	±0.10	±0.14	±0.18	±0.11

NOTES:

1. Samples collected by EML.

2. Asterisks indicate active ratios significantly different from 1.0 at the 95% confidence level.

3. Respirator filter elements.

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TABLE 9. Analysis of Variance Tests of the Particle-Size Dependence
of Radionuclide Radioactivity Ratios

Location	Date/Time Sampled		Derived "F" Values[4]				
			$^{234}U/^{238}U$	$^{230}Th/^{238}U$	$^{226}Ra/^{238}U$	$^{210}Pb/^{238}U$	$^{210}Pb/^{226}Ra$
Secondary Crusher	8/23/77	1310 - 1555	7.03*[1]	1.54[3]	31.5*[3]	3.17[1]	10.0*[1]
	8/23-24/77	1705 - 0730	4.28*[2]	0.35[3]	6.32*[3]	0.78[2]	3.82*[2]
	8/24/77	0905 - 1355	0.26[2]	1.12[3]	2.29[3]	0.41[2]	2.40[2]
	8/24/77	1440 - 1950	6.10*[2]	3.56*[3]	4.26*[3]	1.08[2]	0.91[2]
	8/24-25/77	2055 - 0745	9.08*[2]	1.14[3]	2.57[3]	0.99[2]	2.39[2]
Primary Crusher	8/23/77	1320 - 1605	8.4*[2]	2.4[3]	10.4*[3]	0.76[2]	7.2*[2]
	8/23-24/77	1710 - 0735	0.77[2]	1.3[3]	1.5[3]	0.55[2]	10.1*[2]
	8/24/77	0915 - 1305	2.6[2]	0.24[3]	3.5*[3]	1.2[2]	3.5*[2]
Lower Level Secondary Crusher	8/24/77	1350 - 1950	0.73[2]	0.50[3]	2.2[3]	0.17[2]	1.7[2]
	8/24-25/77	2050 - 0730	1.6[2]	2.3[3]	0.83[3]	0.48[2]	0.74[2]
Near Ore Bin Area	8/25/77	0930 - 1425	13.6*[2]	1.8[3]	2.6[3]	1.4[2]	10.4*
Near Primary Conveyer Belt	8/25/77	0920 - 1400	4.9*[2]	2.6[3]	3.9*[3]	1.8[2]	5.1*[2]
Sample Prep Room "B"	8/25/77	0900 - 1400	5.1*[2]	7.9[3]	5.9*[3]	1.9[2]	4.0*[2]

NOTES:

1. Upper 99% limit for "F" is 3.73.

2. Upper 99% limit for "F" is 3.50.

3. Upper 99% limit for "F" is 3.47.

4. Asterisks indicate "F" values which demonstrate a significant size dependence for activity ratio.

TABLE 10. Airborne Uranium Ore Dust Composition Versus Particle Size
at the Primary Crusher Area of a Second Uranium Mill

	dis/min per sample					Isotope Ratios			
Sample No.	^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb	^{234}U/^{238}U	^{230}Th/^{238}U	^{226}Ra/^{238}U	^{210}Pb/^{238}U
Run 1 [2]									
Stage 1	856±1.8[5]	872±1.8	906±2.1	752±4.8	828±2.8	1.02	1.06	.88	.97
Stage 2	344±1.7	351±1.7	338±2.6	270±4.8	313±2.4	1.02	.98	.78	.91
Stage 3	211±2.6	214±2.6	215±2.3	173±4.8	190±3.8	1.01	1.02	.82	.90
Stage 4	149±2.4	146±2.4	143±2.8	103±4.8	125±2.8	.98	.96	.69	.84
Back Up	151±3.3	149±3.3	137±2.9	104±4.8	129±3.5	.99	.91	.69	.86
Run 2 [3]									
Stage 1	985±1.0	977±1.0	986±2.7	758±4.8	720±3.4	.99	1.01	.77	.73
Stage 2	329±1.7	313±1.7	310±2.3	197±4.8	196±4.4	.97	.94	.60	.60
Stage 3	214±2.1	211±2.1	219±2.7	126±4.8	154±3.5	.99	1.02	.59	.72
Stage 4	170±2.4	161±2.4	165±3.6	106±4.8	122±3.2	.95	.97	.62	.72
Back Up	312±2.1	300±2.1	306±2.3	178±4.8	226±2.5	.96	.98	.57	.72
Run 3 [4]									
Stage 1	3689±.80	3859±.80	3999±3.0	2210±4.8	2850±2.0	1.05	1.08	.60	.77
Stage 2	1392±1.4	1407±1.4	1497±3.5	752±4.8	1031±2.5	1.01	1.07	.54	.74
Stage 3	985±1.1	957±1.1	986±2.1	507±4.8	702±2.2	.97	1.00	.51	.71
Stage 4	715±2.0	673±2.0	650±2.8	399±4.8	469±1.7	.94	.91	.56	.66
Back Up	1542±2.0	1456±2.0	1555±2.6	762±4.8	1063±1.8	.94	1.01	.49	.69
Reagent Blank	1.12±17	1.23±18	.680±4.4	.109±18	2.03±8.0	(1.10)	(.61)	(.097)	(1.81)

NOTES:

1. Samples collected by PNL with a 20 cfm 4 stage Andersen Cascade Impactor. Particle size decreases with increasing stage number.

2. Collected at the second level of the crusher area   6/?3/78 @ 10:38 a.m. Limestone type ore being crushed (very dusty).

3. Collected at the second level of the crusher area   6/23/78 @ 14:12 p.m. Sandstone type ore being crushed (wet, less dusty).

4. Collected at the second level of the crusher area   6/23/78 @ 15:14 p.m. Sandstone type ore being crushed (wet, less dusty).

5. Errors expressed in units of percent relative standard deviation from propagation of counting errors. Radium-226 includes an inherent measurement error of 4%.

Sample Number	Date and Time Processed		Radioactive Ratios [2,3]				
			$^{234}U/^{238}U$	$^{230}Th/^{238}U$	$^{226}Ra/^{238}U$	$^{210}Pb/^{238}U$	$^{210}Pb/^{226}Ra$
139	8/24/77	0250 - 0630	0.96	1.06	1.01	0.91	0.90
140	8/24/77	0630 - 1100	1.02	1.09	0.95	0.86	0.91
141	8/24/77	1500 - 1655	0.96	1.08	0.83*	0.77*	0.93
142	8/24/77	1655 - 1900	0.95	1.04	0.84*	0.77*	0.92
143	8/24/77	2045 - 2325	1.00	0.99	0.81*	0.81*	0.99
144	8/25/77	1100 - 1415	0.99	1.07	1.15*	0.94	0.82*
145	8/25/77	1415 - 1935	0.99	1.06	1.00	0.93	0.93
146	8/25-26/77	2310 - 0550	1.05	1.14	1.00	0.81*	0.81*
147	8/24-25/77	1930 - 0300	1.02	1.10	1.04	1.07	1.09
148	8/24/77	1900 - 0250	0.98	1.07	0.99	0.95	0.95

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NOTES:

1. These composite ore samples were collected from production runs contemporary with air sampling by EML.

2. Asterisks demonstrate significant disequilibrium.

3. Precision estimates for ^{234}U and ^{210}Pb were based on pooled variances of air samples. Estimates for ^{230}Th and ^{226}Ra are based on pooling the variances of aliquots analyzed by photon spectrometry. Statistical tests for the significance of the measured disequilibrium were based on the student "t" test at the 95% confidence level.

TABLE 12. Uranium Decay Chain Concentrations in Ore Samples from Uranium Mills in the United States

Mill	State[2]	Sample[1]	Concentrations (dis/min per gram)				
			^{238}U	^{230}Th	^{226}Ra	^{210}Pb	^{214}Pb
A	WY	1a	312	233	166	205	142
A	WY	1b	309	203	177	214	150
A	WY	2a	411	345	272	309	233
A	WY	2b	491	413	342	372	282
A	WY	3a	453	377	378	384	306
A	WY	3b	579	532	338	453	308
B	WY	1a	552	521	486	577	366
B	WY	1b	552	542	489	564	368
B	WY	2a	766	724	836	815	486
B	WY	2b	782	801	763	863	481
B	WY	3a	478	491	498	524	312
B	WY	3b	472	502	499	523	315
C	WY	1a	906	1006	876	955	695
C	WY	1b	926	1035	878	951	709
C	WY	2a	863	916	792	869	540
C	WY	2b	806	977	759	840	503
C	WY	3a	887	837	851	914	672
C	WY	3b	871	936	870	925	662
D	NM	1a	538	620	609	591	445
D	NM	1b	538	555	519	546	402
D	NM	2a	564	591	560	594	441
D	NM	2b	563	536	585	596	455
D	NM	3a	547	551	565	579	438
D	NM	3b	571	676	563	570	426
E	NM	1a	532	569	500	543	395
E	NM	1b	530	536	536	568	407
E	NM	2a	415	425	404	448	318
E	NM	2b	406	382	415	448	336
E	NM	3a	830	889	788	874	650
E	NM	3b	817	847	762	874	644

TABLE 12 (continued)

Mill	State[2]	Sample[1]	Concentrations (dis/min per gram)				
			^{238}U	^{230}Th	^{226}Ra	^{210}Pb	^{214}Pb
F	CO	1a	788	763	682	784	538
F	CO	1b	760	767	734	778	551
F	CO	2a	911	914	833	933	655
F	CO	2b	787	774	746	811	586
F	CO	3a	874	867	767	839	603
F	CO	3b	936	950	798	909	633
G	WA	1a	580	576	569	568	414
G	WA	1b	587	659	561	575	411
G	WA	2a	859	1015	779	791	623
G	WA	2b	871	1011	752	796	621
G	WA	3a	874	990	804	843	669
G	WA	3b	861	949	850	859	670
H	TX	1a	562	499	444	521	473
H	TX	1b	569	592	487	525	481
H	TX	2a	861	912	859	850	781
H	TX	2b	856	874	902	845	802

NOTE:

1. Two aliquots of ore dust (a and b) were analyzed.

2. WY = Wyoming; NM = New Mexico; CO = Colorado; WA = Washington, TX = Texas.

TABLE 13. Uranium Decay Chain Disequilibrium in Ore Samples Collected from Uranium Mills

Mill	State	Sample	Radioactivity Ratio[1]				
			$^{230}Th/^{238}U$	$^{226}Ra/^{238}U$	$^{210}Pb/^{238}U$	$^{230}Th/^{226}Ra$	$^{210}Pb/^{226}Ra$
A	WY	1	0.702*	0.553*	0.689*	1.278*	1.246*
A	WY	2	0.840*	0.680*	0.755*	1.238*	1.130*
A	WY	3	0.875*	0.708*	0.815*	1.285*	1.195*
B	WY	1	0.963	0.883*	1.034	1.090	1.192*
B	WY	2	0.984	1.034	1.084*	0.958	1.070
B	WY	3	1.042	1.052	1.110*	0.996	1.068
C	WY	1	1.114*	0.958	1.040*	1.165	1.086*
C	WY	2	1.138*	0.930	1.024	1.222*	1.102*
C	WY	3	1.009	0.979	1.046*	1.030	1.069
D	NM	1	1.091	1.047	1.058*	1.044	1.030
D	NM	2	1.000	1.016	1.055*	0.986	1.040
D	NM	3	1.096	1.010	1.029	1.088	1.018
E	NM	1	1.041	0.976	1.048*	1.069	1.092*
E	NM	2	0.984	1.000	1.094*	0.986	1.094*
E	NM	3	1.054	0.941	1.062*	1.120	1.128*
F	CO	1	0.990	0.916*	1.008	1.082	1.144*
F	CO	2	0.992	0.930	1.030	1.068	1.126*
F	CO	3	1.004	0.866*	0.966	1.160	1.136*
G	WA	1(2)	1.057	0.968	0.978	1.094	1.011
G	WA	2(3)	1.172*	0.886*	0.921*	1.324*	1.037
G	WA	3(3)	1.117*	0.954	0.980	1.174	1.029
H	TX	1(4)	0.964	0.822*	0.925	1.170	1.164*
H	TX	2(5)	1.040	1.026	0.987	0.016	0.976
95% Confidence Interval:			±0.091	±0.083	±0.037	±0.174	±0.085

NOTES

1. Asterisks indicate a value significantly different from 1.00 at the 95% confidence level based on pooled variance estimates from counts of duplicate pellets.

2. Ore was noted as being oxidized.

3. Ore was noted as being reduced.

4. Ore was noted as beign in lignitic sand.
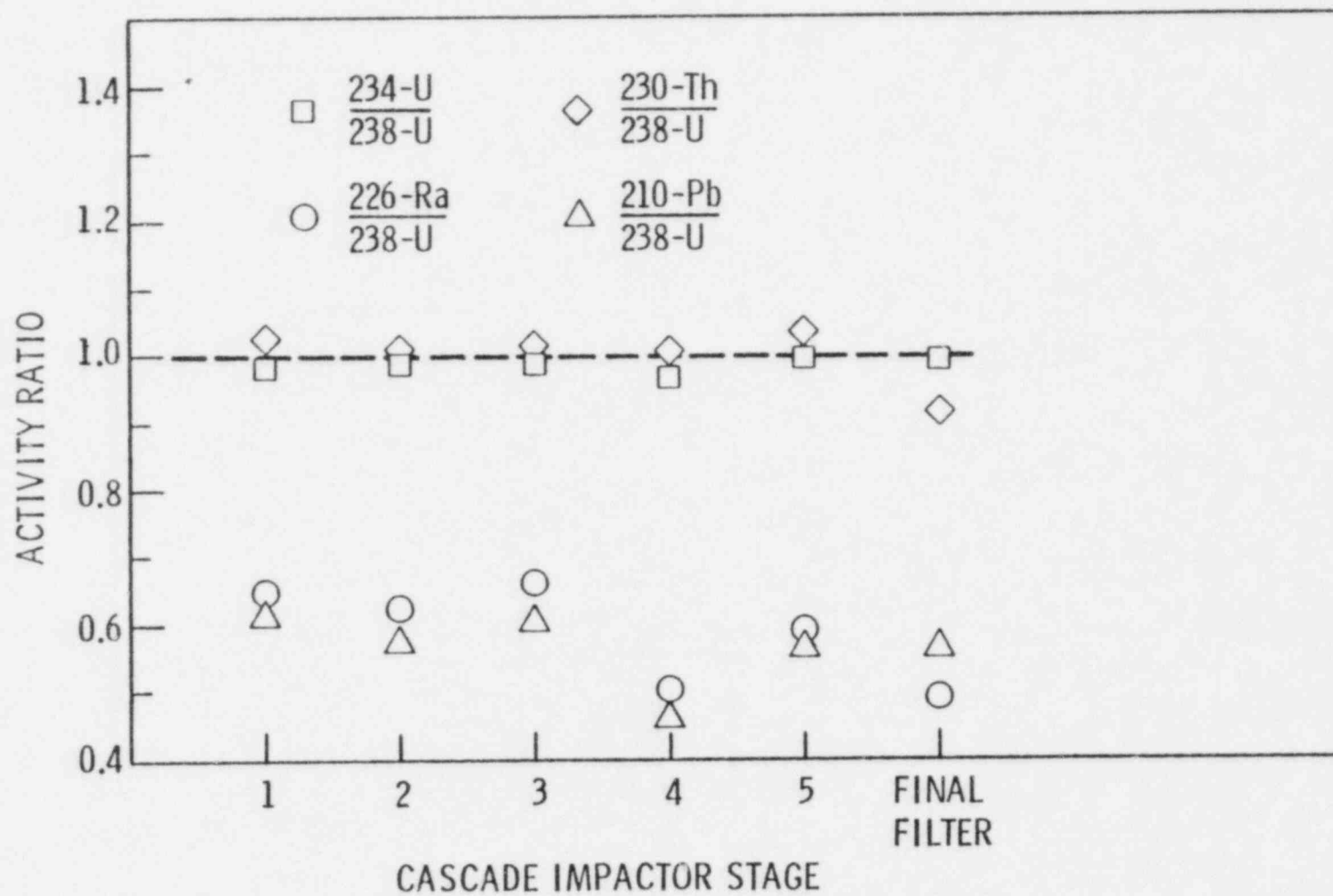
5. Ore was noted as being high in lime.

FIGURE 1. Particle Size Dependencies of Radioactivity Ratios Measured at the Secondary Crusher on 8-23-77 from 1310 to 15:55
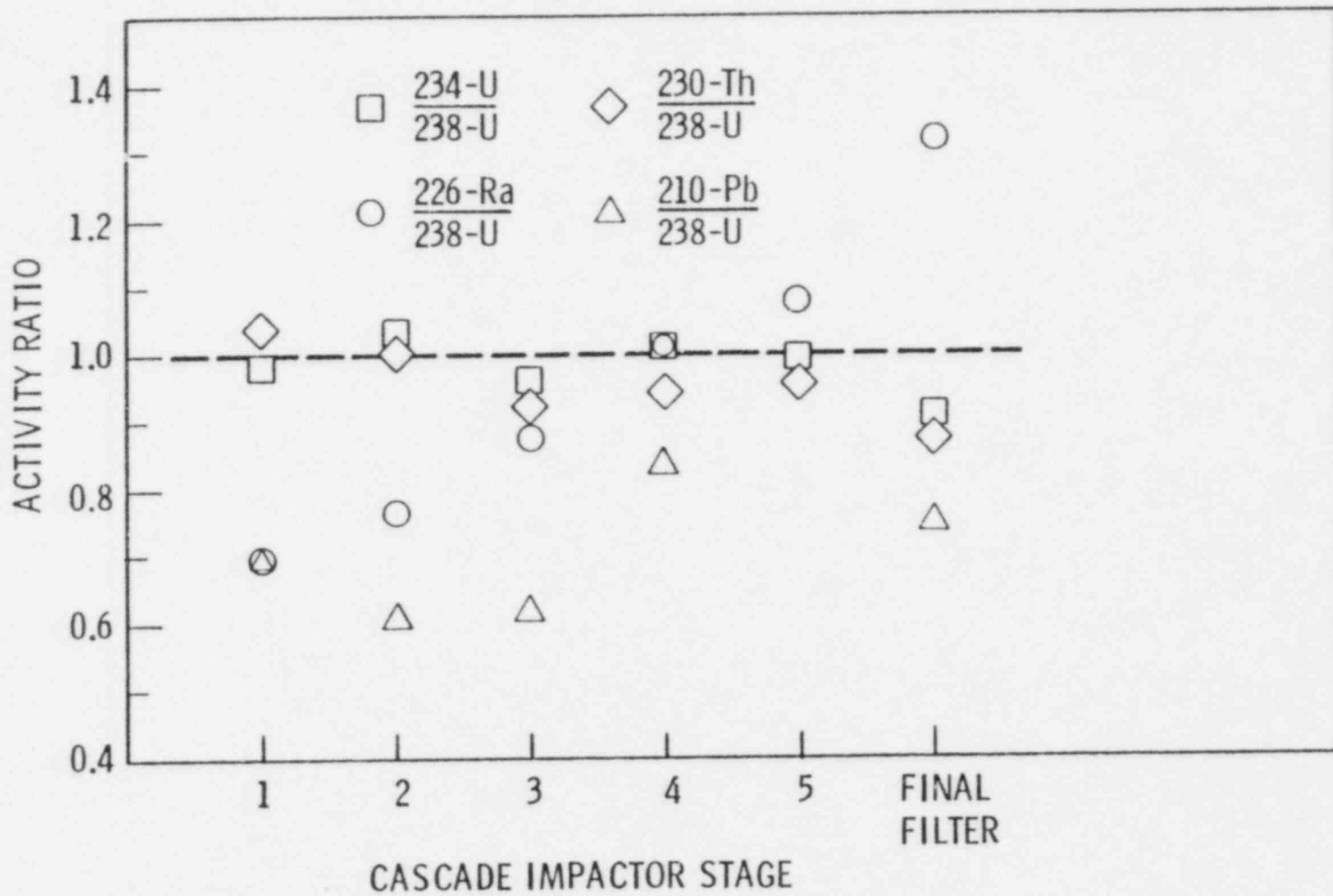
FIGURE 2. Particle Size Dependencies of Radioactivity Ratios Measured at the Secondary Crusher on 8-24-77 from 09:05 to 13:55

APPENDIX A

Uranium Ore Dust Composition On Cascade Impactor Stages

dis/min per filter

Sample No.[1]		^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb
1	82310 Ref	1000 ± 2.2	1000 ± 2.2	940 ± 3.2	950 ± 4.8	842 ± 1.5
2	82311 1-1	359 ± 3.0	355 ± 3.2	316 ± 2.9	237 ± 4.8	275 ± 2.5
3	82312 1-2	257 ± 1.9	273 ± 1.8	236 ± 3.2	220 ± 4.8	213 ± 2.1
4	82313 1-3	148 ± 2.0	144 ± 2.0	127 ± 3.1	151 ± 4.8	107 ± 3.0
5	82314 1-4	91.5 ± 2.6	92.1 ± 2.6	83.0 ± 3.2	105 ± 4.8	65.5 ± 3.1
6	82315 1-5	59.3 ± 1.7	58.3 ± 1.8	65.4 ± 3.9	77.3 ± 4.8	57.6 ± 3.9
7	82316 ff	51.6 ± 3.1	54.2 ± 3.2	48.1 ± 3.7	80.4 ± 4.8	60.0 ± 4.0
8	82317 Env Blank	2.42 ± 12	2.16 ± 12	1.91 ± 16	.68 ± 4.8	2.86 ± 11
9	82318 Control Filter	3.19 ± 3.5	4.40 ± 3.0	3.65 ± 3.1	3.81 ± 4.8	5.62 ± 13
10	82321 2-1	309 ± 1.6	305 ± 1.6	321 ± 2.5	216 ± 4.8	216 ± 2.2
11	82322 2-2	254 ± 2.3	261 ± 2.3	257 ± 2.3	193 ± 4.8	154 ± 2.5
12	82323 2-3	154 ± 2.5	148 ± 2.6	141 ± 3.4	134 ± 4.8	96.0 ± 4.1
13	82324 2-4	83.3 ± 2.5	83.9 ± 2.5	79.2 ± 3.8	84.1 ± 4.8	70.0 ± 4.0
14	82325 2-5	67.3 ± 2.4	66.7 ± 2.4	64.8 ± 3.7	72.0 ± 4.8	2
15	82326 ff	45.2 ± 3.2	40.9 ± 3.2	39.7 ± 4.2	59.2 ± 4.8	34.0 ± 4.8
Reagent & Procedure Blank		.31 ± 29	.33 ± 27	.48 ± 18	.28 ± 8.6	.28 ± 40
16	82330 Ref	1090 ± 2.0	1060 ± 2.0	1150 ± 2.8	1240 ± 4.8	1030 ± 1.5
17	82331 3-1	64.0 ± 2.8	62.5 ± 2.8	61.4 ± 5.6	70.2 ± 4.8	54.0 ± 3.6
18	82332 3-2	116 ± 3.8	116 ± 3.8	116 ± 2.9	129 ± 4.8	94.0 ± 2.5
19	82333 3-3	78.5 ± 1.4	77.3 ± 1.4	72.4 ± 3.4	84.1 ± 4.8	71.8 ± 2.5
20	82334 3-4	24.8 ± 4.0	25.4 ± 3.9	26.6 ± 4.6	29.8 ± 4.8	16.9 ± 5.0
21	82335 3-5	16.9 ± 3.2	15.1 ± 3.4	15.9 ± 4.7	27.1 ± 4.8	13.2 ± 5.5
22	82336 ff	13.6 ± 7.9	12.8 ± 9.5	11.6 ± 8.9	19.9 ± 4.8	11.0 ± 5.2
Reagent & Procedure Blank		<.074	.71 ± 16	.08 ± 50	.20 ± 9.4	.75 ± 20
23	84710 Ref	2350 ± 1.4	2340 ± 1.4	2400 ± 2.9	2030 ± 4.8	1740 ± 1.4
24	84711 1-1	903 ± 1.6	840 ± 1.6	854 ± 2.9	662 ± 4.8	727 ± 1.4
25	84712 1-2	707 ± 2.3	724 ± 2.2	746 ± 2.5	631 ± 4.8	633 ± 1.4
26	84713 1-3	306 ± 2.0	305 ± 2.0	316 ± 2.6	292 ± 4.8	255 ± 1.8

Uranium Ore Dust Composition On Cascade Impactor Stages

dis/min per filter

Sample No.[1]			^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb
27	84714	1-4	182 ± 3.5	183 ± 2.5	184 ± 2.8	193 ± 4.8	161 ± 1.2
28	84715	1-5	103 ± 2.1	101 ± 2.2	104 ± 4.2	103 ± 4.8	83.1 ± 1.7
29	84716	f?	129 ± 2.4	132 ± 2.4	136 ± 3.1	147 ± 4.8	131 ± 1.4
30	84720						
Slotted filter blank			12.6 ± 3.2	12.2 ± 3.3	2.45 ± 37	1.29 ± 5.1	1.07 ± 5.9
31	84721	2-1	823 ± 2.5	809 ± 2.5	823 ± 2.5	625 ± 4.8	624 ± 1.9
32	84722	2-2	563 ± 3.0	575 ± 3.0	544 ± 2.7	526 ± 4.8	429 ± 2.0
33	84723	2-3	268 ± 3.0	270 ± 3.0	280 ± 2.5	246 ± 12	202 ± 1.6
34	84724	2-4	134 ± 2.2	137 ± 2.2	133 ± 1.2	134 ± 4.8	97.1 ± 3.2
35	84725	2-5	96.6 ± 2.4	96.6 ± 2.4	97.3 ± 3.5	101 ± 4.8	70.4 ± 3.7
36	84726	ff	128 ± 2.1	127 ± 2.2	117 ± 2.6	128 ± 4.8	105 ± 3.0
37	84730	Ref	15000 ± 1.0	14900 ± 1.0	14900 ± 3.1	10700 ± 4.8	12600 ± 1.5
38	84731	3-1	5020 ± 1.1	5010 ± 1.1	5280 ± 3.7	4000 ± 4.8	4540 ± 1.4
39	84732	3-2	4170 ± 1.0	4140 ± 1.1	4380 ± 3.0	3480 ± 4.8	3580 ± 1.8
40	84733	3-3	862 ± 1.9	835 ± 1.9	845 ± 3.1	816 ± 20	831 ± 1.7
41	84734	3-4	385 ± 1.1	379 ± 1.1	388 ± 2.5	313 ± 4.8	330 ± 2.6
42	84735	3-5	116 ± 1.6	115 ± 1.6	127 ± 2.6	93.2 ± 4.8	98.7 ± 2.4
43	84736	ff	92.6 ± 2.3	93.6 ± 2.3	87.4 ± 3.6	63.6 ± 4.8	93.3 ± 9.0
44	82410	Ref	1660 ± 2.2	1610 ± 2.2	1650 ± 2.4	1000 ± 4.8	911 ± 1.5
45	82411	1-1	277 ± 1.9	274 ± 1.9	284 ± 2.4	177 ± 4.8	174 ± 1.6
46	82412	1-2	354 ± 1.7	350 ± 1.7	354 ± 2.6	219 ± 4.8	206 ± 1.8
47	82413	1-3	199 ± 2.4	197 ± 2.5	200 ± 1.2	132 ± 4.8	122 ± 1.9
48	82414	1-4	200 ± 3.3	193 ± 3.3	200 ± 2.6	99 ± 4.8	92.1 ± 4.6
49	82415	1-5	110 ± 2.2	110 ± 2.2	113 ± 3.2	64.7 ± 4.8	62.6 ± 3.4
50	82416	ff	144 ± 2.3	144 ± 2.3	133 ± 2.7	68.5 ± 4.8	80.2 ± 3.0
51	Whatman Control Blank		<.84	<.96	<1.2	1.28 ± 16	.212 ± 13
52	82421	2-1	262 ± 2.2	260 ± 2.2	272 ± 2.4	162 ± 4.8	166 ± 1.0
53	82422	2-2	333 ± 2.0	330 ± 2.0	330 ± 2.3	196 ± 8.3	217 ± 1.3
54	82423	2-3	225 ± 1.5	226 ± 1.5	224 ± 2.7	148 ± 4.8	139 ± 2.5
55	82424	2-4	146 ± 1.7	147 ± 1.7	143 ± 3.0	66.7 ± 4.8	85.1 ± 3.7

Uranium Ore Dust Composition On Cascade Impactor Stages

dis/min per filter

Sample No. [1]			^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb
56	82425	2-5	124 ± 3.3	125 ± 3.3	119 ± 3.0	58.0 ± 4.8	75.7 ± 2.5
57	82426	ff	156 ± 1.6	155 ± 1.6	145 ± 3.3	81.8 ± 4.8	91.4 ± 1.8
58	82430	Ref	3250 ± 2.0	3280 ± 2.0	3100 ± 2.8	2350 ± 4.8	2270 ± 1.0
59	82431	3-1	347 ± 2.5	367 ± 2.6	336 ± 3.4	274 ± 4.8	275 ± 1.3
60	82432	3-2	630 ± 2.2	674 ± 2.2	592 ± 2.7	529 ± 4.8	486 ± 1.1
61	82433	3-3	333 ± 2.9	341 ± 2.9	309 ± 2.7	270 ± 4.8	249 ± 1.2
62	82434	3-4	82.3 ± 2.2	83.4 ± 2.2	75.1 ± 3.8	59.3 ± 4.8	56.4 ± 2.3
63	82435	3-5	34.6 ± 2.8	36.0 ± 2.8	32.9 ± 3.8	19.3 ± 4.8	18.4 ± 6.3
64	82436	ff	38.1 ± 3.2	40.8 ± 3.2	34.5 ± 3.6	20.8 ± 4.8	24.7 ± 5.7
65	82440	Ref	874 ± 1.6	872 ± 1.6	823 ± 1.3	745 ± 4.8	747 ± 2.2
66	82441	1-1	139 ± 2.0	140 ± 2.0	129 ± 4.0	107 ± 4.8	113 ± 3.0
67	82442	1-2	252 ± 1.5	249 ± 1.5	237 ± 3.0	220 ± 4.8	215 ± 2.4
68	82443	1-3	137 ± 1.4	147 ± 1.4	130 ± 4.4	128 ± 4.8	114 ± 3.1
69	82444	1-4	87.8 ± 1.7	89.4 ± 1.7	81.2 ± 4.1	88.8 ± 4.8	76.0 ± 3.4
70	82445	1-5	46.6 ± 2.2	46.5 ± 2.2	50.2 ± 3.4	50.2 ± 4.8	40.6 ± 3.0
71	82446	ff	51.7 ± 2.0	49.0 ± 2.0	45.7 ± 3.9	52.3 ± 4.8	49.1 ± 3.3
72	82451	2-1	272 ± 1.1	276 ± 1.1	258 ± 4.8	232 ± 4.8	176 ± 2.3
73	82452	2-2	246 ± 1.3	250 ± 1.3	257 ± 4.8	236 ± 4.8	177 ± 2.1
74	82453	2-3	105 ± 1.6	108 ± 1.6	93.5 ± 4.9	101 ± 4.8	83.2 ± 3.0
75	82454	2-4	55.1 ± 1.9	56.8 ± 1.9	54.5 ± 4.8	58.2 ± 4.8	44.6 ± 3.7
76	82455	2-5	42.6 ± 2.4	42.6 ± 2.4	42.4 ± 3.7	46.8 ± 4.8	33.4 ± 4.6
77	82456	ff	51.1 ± 2.0	50.2 ± 2.0	41.8 ± 3.9	52.2 ± 4.8	45.8 ± 5.5
78	82460	Ref	6030 ± 1.0	6060 ± 1.0	5700 ± 3.1	4300 ± 4.8	3690 ± 1.5
79	82461	3-1	990 ± 1.5	997 ± 1.5	888 ± 3.5	674 ± 4.8	649 ± 2.0
80	82462	3-2	544 ± 1.1	547 ± 1.1	486 ± 3.9	433 ± 4.8	382 ± 1.5
81	82463	3-3	252 ± 1.4	246 ± 1.4	227 ± 3.5	236 ± 4.8	173 ± 2.3
82	82464	3-4	126 ± 1.6	126 ± 1.6	123 ± 3.5	120 ± 4.8	95.2 ± 3.1
83	82465	3-5	103 ± 1.7	102 ± 1.7	93.8 ± 3.6	92.2 ± 4.8	72.3 ± 3.1
84	82466	ff	117 ± 1.6	115 ± 1.6	104 ± 1.7	94.2 ± 4.8	86.4 ± 2.9
85	84910	Ref	4400 ± 1.0	4490 ± 1.0	3990 ± 2.7	3250 ± 4.8	3270 ± 1.7
86	84911	1-1	1360 ± 1.0	1340 ± 1.0	1320 ± 2.4	1080 ± 4.8	974 ± 2.0

Uranium Ore Dust Composition On Cascade Impactor Stages

dis/min per filter

Sample No. [1]			^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb
37	84912	1-2	1430 ± 1.5	1360 ± 1.5	1390 ± 2.5	1020 ± 4.8	1160 ± 1.5
88	84913	1-3	604 ± 1.0	579 ± 1.0	603 ± 2.6	487 ± 4.8	477 ± 1.3
89	84914	1-4	295 ± 3.5	307 ± 3.5	325 ± 2.6	245 ± 4.8	215 ± 2.2
90	84915	1-5	170 ± 1.0	168 ± 1.0	181 ± 2.6	138 ± 4.8	131 ± 2.4
91	84916	ff	147 ± 2.8	140 ± 2.9	139 ± 2.8	107 ± 4.8	102 ± 1.0
92	84921	2-1	1450 ± 1.0	1450 ± 1.0	1510 ± 2.5	1010 ± 4.8	1150 ± 1.4
93	84922	2-2	1060 ± 1.4	1030 ± 1.4	1000 ± 3.3	763 ± 4.8	775 ± 1.0
94	84923	2-3	415 ± 2.0	399 ± 2.0	425 ± 2.6	421 ± 20	325 ± 2.1
95	84924	2-4	190 ± 2.4	195 ± 2.4	193 ± 3.2	156 ± 4.8	141 ± 2.5
96	84925	2-5	149 ± 2.6	139 ± 2.7	152 ± 4.0	113 ± 4.8	111 ± 1.0
97	84926	ff	136 ± 2.0	127 ± 2.0	142 ± 2.8	92.5 ± 9.5	106 ± 2.5
98	84930	Ref	21900 ± 1.0	21900 ± 1.0	23300 ± 2.7	13500 ± 4.8	14400 ± 1.3
99	84931	3-1	6030 ± 1.4	5800 ± 1.4	6310 ± 2.8	3540 ± 4.8	3950 ± 1.5
100	84932	3-2	2110 ± 1.3	2020 ± 1.3	2100 ± 2.5	1290 ± 4.8	1400 ± 1.0
101	84933	3-3	731 ± 1.3	701 ± 1.3	669 ± 2.8	480 ± 4.8	562 ± 1.1
102	84934	3-4	316 ± 1.0	319 ± 1.0	340 ± 2.9	234 ± 4.8	251 ± 2.0
103	94935	3-5	262 ± 1.3	258 ± 1.3	261 ± 2.5	184 ± 4.8	206 ± 2.4
104	94936	ff	226 ± 1.3	218 ± 1.5	202 ± 2.2	164 ± 4.8	170 ± 2.5
Reagent & Procedure Blank			.430 ± 25	.538 ± 20	.517 ± 17	.676 ± 16	.625 ± 33
105	82510	Ref	302 ± 1.5	302 ± 1.5	278 ± 2.6	269 ± 4.8	226 ± 2.8
106	82511	1-1	43.8 ± 4.3	45.4 ± 4.4	36.3 ± 4.9	36.7 ± 4.8	31.2 ± 4.7
107	82512	1-2	54.9 ± 2.1	55.2 ± 2.1	46.4 ± 3.5	45.7 ± 4.8	35.8 ± 4.2
108	82513	1-3	20.9 ± 2.7	19.4 ± 2.9	15.6 ± 4.5	12.4 ± 4.8	10.4 ± 6.5
109	82514	1-4	12.6 ± 3.4	12.5 ± 3.4	11.5 ± 4.8	10.4 ± 4.8	9.42 ± 8.7
110	82515	1-5	7.92 ± 7.4	8.46 ± 7.9	7.10 ± 5.8	6.26 ± 4.8	5.55 ± 7.4
111	82516	ff	24.1 ± 3.0	22.2 ± 3.1	18.7 ± 4.1	15.4 ± 4.8	19.5 ± 4.5
112	82520	Ref	517 ± 1.2	527 ± 1.2	495 ± 2.5	390 ± 4.8	356 ± 2.2
113	82521	2-1	181 ± 1.5	177 ± 1.5	173 ± 2.8	143 ± 4.8	138 ± 2.0
114	82522	2-2	105 ± 1.6	106 ± 1.6	102 ± 2.9	82.1 ± 4.8	71.3 ± 3.2
115	82523	2-3	34.7 ± 3.1	33.9 ± 3.1	31.3 ± 6.4	20.1 ± 4.8	23.2 ± 5.0

A-4

Uranium Ore Dust Composition On Cascade Impactor Stages

dis/min per filter

Sample No.			^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb
116	82524	2-4	11.8 ± 3.7	12.6 ± 3.7	9.47 ± 4.6	5.95 ± 4.8	6.81 ± 8.4
117	82525	2-5	8.58 ± 2.5	8.72 ± 2.5	7.79 ± 4.8	4.41 ± 4.8	3.83 ± 9.0
118	82526	ff	6.36 ± 7.1	6.26 ± 7.2	5.01 ± 13	3.40 ± 4.8	3.21 ± 9.5
119	82530	Ref	572 ± 1.5	₋70 ± 1.5	477 ± 2.4	409 ± 4.8	431 ± 2.2
120	82531	3-1	34.4 ± 3.5	33.6 ± 3.6	31.4 ± 4.2	30.3 ± 4.8	28.1 ± 5.4
121	82532	3-2	48.9 ± 2.5	49.1 ± 2.5	45.8 ± 3.8	48.9 ± 4.8	36.3 ± 4.0
122	82533	3-3	43.6 ± 2.8	42.1 ± 2.9	35.6 ± 3.7	35.0 ± 4.8	26.9 ± 4.8
123	82534	3-4	22.3 ± 5.3	23.5 ± 5.3	18.3 ± 5.4	16.3 ± 4.8	15.5 ± 6.1
124	82535	3-5	19.7 ± 3.8	20.8 ± 4.0	14.2 ± 5.4	16.7 ± 4.8	11.8 ± 6.3
125	82536	ff	19.6 ± 6.0	19.8 ± 6.0	11.2 ± 4.9	10.0 ± 4.8	9.16 ± 6.6

NOTES:

1. Samples collected by EML.

   In addition to the PNL identification number, the EML log number, cascade impactor stages, reference high volume filters and blanks are tabulated. The format used is PNL number, EML number, Impactor-Stage number; ff = final filter; Ref is a reference filter at the same locations as impactors which precede it (1 and 2 or 3). Results reported ± the relative standard deviation (%) from the propagation of radioactivity counting measurement errors.

   Appropriate blank corrections have been made to all reported results. In this report, the uranium content of the slotted filters was assumed to be the average of the ^{230}Th, ^{210}Pb, and ^{226}Ra contents, and corrections were made, since there was more uranium in the slotted filter paper blanks than in some exposed samples. The inherent variation of the ^{226}Ra emanation counting technique for these samples was approximately ±4% for a single replicate. Propagating this error with other inherent errors yielded a value of ±4.8%--unless limited by counting statistics as in the case of blanks.

2. Sample 14-82325 2-5: The ^{210}Pb fraction was lost.

APPENDIX B

Uranium Ore Dust Composition

General Air Filters [1]

dis/min per filter [2]

Sample No.	^{238}U	^{234}U	^{230}Th	^{226}Ra	^{210}Pb
126	79.8 ± 2.6	82.6 ± 2.6	77.8 ± 3.7	75.6 ± 4.8	119 ± 2.3
127	1930 ± 1.9	1900 ± 1.9	18.7 ± 4.3	5.08 ± 13	17.5 ± 9.0
128	4070 ± 1.5	4120 ± 1.5	4060 ± 2.4	4210 ± 4.8	4000 ± 1.8
129	426 ± 1.3	433 ± 1.3	425 ± 3.2	385 ± 4.8	378 ± 1.0
130	156 ± 1.8	150 ± 1.8	154 ± 3.3	142 ± 4.8	148 ± 1.1
131	458 ± 1.5	451 ± 1.5	477 ± 2.5	281 ± 4.8	326 ± 1.9
132	1450 ± 1.7	1430 ± 1.7	33 ± 3.5	22.9 ± 4.8	57.9 ± 1.7
133	1730 ± 1.4	1690 ± 1.4	1660 ± 3.1	974 ± 4.8	1380 ± 1.1
134	296 ± 1.6	297 ± 1.6	278 ± 2.8	257 ± 4.8	241 ± 1.3
135	8960 ± 1.6	9000 ± 1.6	8200 ± 2.8	7690 ± 4.8	7140 ± 1.3
136	3230 ± 1.0	3210 ± 1.0	24.6 ± 5.0	12.2 ± 4.8	4.15 ± 11
137	63.4 ± 3.1	65.0 ± 3.1	62.5 ± 4.3	48.5 ± 4.8	48.2 ± 4.2
138	79.8 ± 1.2	77.1 ± 1.2	68.8 ± 3.7	56.1 ± 4.8	52.6 ± 3.9
Reagent and Procedure Blank	7.53 ± 16	7.53 ± 16	.23 ± 18	.28 ± 16	2.37 ± 19

NOTES:

1. General air filters were collected by EML personnel at various locations in the uranium mill. These samples were not accompanied by cascade impactors.

2. All samples have been corrected for the reagent blank.

3. Previous estimates are percent relative standard deviations based on counting statistics.

Composition of Uranium Ores Being Milled At the
Time of Air Sample Collection[1]

Sample No.	dis/min per gram[3]				
	^{238}U	^{234}U	^{230}Th[2]	^{226}Ra[2]	^{210}Pb
139	1070 ± 1.1	1030 ± 1.1	1130 ± 7.9	1080 ± 3.1	976 ± 7.4
140	927 ± 1.3	948 ± 1.3	1010 ± 7.9	880 ± 3.1	798 ± 3.4
141	416 ± 1.1	400 ± 1.1	434 ± 7.9	345 ± 3.1	321 ± 1.9
142	496 ± 1.2	472 ± 1.2	514 ± 7.9	415 ± 3.1	383 ± 2.3
143	549 ± 1.5	551 ± 1.5	545 ± 7.9	445 ± 3.1	442 ± 1.6
144	1050 ± 1.5	1040 ± 1.5	1120 ± 7.9	1204 ± 3.1	983 ± 4.0
145	757 ± 1.4	746 ± 1.4	805 ± 7.9	757 ± 3.1	702 ± 1.4
146	1180 ± 1.3	1240 ± 1.2	1340 ± 7.9	1180 ± 3.1	959 ± 1.3
147	1270 ± 1.3	1290 ± 1.3	1400 ± 7.9	1110 ± 3.1	1360 ± 2.5
148	4380 ± 1.2	4280 ± 1.2	4700 ± 7.9	4356 ± 3.1	4150 ± 1.0

NOTES

(1) Collected for EML.

(2) The ^{230}Th and ^{226}Ra results are based upon direct counting with an intrinsic germanium diode. The error estimates for direct counting include propagated counting errors (RMS error) and estimates of the accuracy of calibration (linear error). They are in units of % relative standard deviation.

(3) Appropriate reagent blank corrections have been applied to these results.

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The degree of disequilibrium among ^{238}U decay chain members in some airborne dusts and typical ores has been established by precise radiochemical analyses. This information is necessary to evaluate the lung dose model currently used for estimating the effect of the inhalation of uranium ore dust. The particle size distributions of airborne decay chain components in dusts at one uranium mill have been investigated. Statistically significant disequilibria were observed for ^{230}Th, ^{226}Ra, and ^{210}Pb in both airborne dusts and composite ore samples. With the exception of ore from one mill in the United States, most of the daughter concentrations in powdered ore composites were within 10% of ^{238}U. In airborne dusts, the concentration of ^{226}Ra was typically below ^{238}U; the minimum ^{226}Ra concentration observed for airborne ore dusts was 56% of equilibrium. A statistically significant particle size dependence was observed for ^{226}Ra/^{238}U ratios in several airborne dusts collected at a uranium mill.

17. KEY WORDS AND DOCUMENT ANALYSIS

17a. DESCRIPTORS

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