AIRBORNE PARTICLE SIZE DISTRIBUTION MEASUREMENTS AT USDOE FERNALD

N.H. Harley[1] , P. Chittaporn[1], Heikkinen, M.[1], R. Medora[1], R. Merrill[2], ([1]New York University School of Medicine, Nelson Inst. of Environmental Medicine, 550 First Avenue, New York, NY 10016, [2] Fluor Fernald Radiation Control Section, 7400 Wiley Road, Hamilton, OH)

Abstract

There is presently no other atmospheric aerosol sampler available, commercially or otherwise, that will sample continuously for up to 2 months in outdoor conditions and provide particle size distribution information. This new sampler does so using environmental concentrations of natural radioactivity as a tracer (Harley et al. 2000). This aerosol sampler has been developed for personal or area use specifically for exposure assessment at the DOE Fernald, OH site in the EMSP program. Measurements taken at Fernald are presented along with measurements at a suburban New Jersey location used for quality control.

A New Particle Size Sampler

The Fernald Feed Materials Production Center (FMPC), 16 miles from Cincinnati, processed uranium ore and thorium during the period from 1951 through 1988. FMPC processed raw ore (feed material) using nitrate digestion, followed by oxide reduction, tetrafluoride production, and then reduction to the metal. The uranium was separated from the nitrate digestion by solvent extraction, the radium was separated by barium sulfate coprecipitation, and the resulting salt cake was stored initially in drums, and later transferred to 2 silos. The remaining liquid residue (raffinate) containing the other radionuclides in the uranium natural activity series was ponded in raffinate pits. They also recovered scrap from the metal fabrication. The 2 large silos each contain about 7.4×10^{13} Bq (2000 Ci) of radium. This site is now undergoing restoration for future return of the land to the State for public use. The purpose of our measurements is to provide an accurate personal exposure assessment of inhaled radioactivity during the removal of relatively large quantities of radium and thorium bearing ore residues. Also, the measurements are to provide detailed data on the airborne transport of material within or outside the boundaries of the site, and assess current dispersion models.

The present sampler configuration uses a miniature commercial sampler head (4 cm. x 2 cm. diam./height, weight 55 gms.) modified to contain a ZnS alpha phosphor inlet impaction stage, followed by four, very fine mesh stainless steel screen filters, and an exit Millipore filter to collect all residual particles. A low flow pump (4-6 lpm) draws the atmosphere sampled through the sampler for periods of up to 2 months for an integrated particle size sample. Six units have sampled outdoors at the USDOE Fernald site for about 2 years. The locations are, on top of and between the two radium storage silos, near the raffinate waste pits, and several portable locations during the ongoing decontamination process.

Radon and thoron measurements are always made simultaneously with the particle size

measurements using the miniature 4 chamber alpha track detector also developed for exposure assessment measurements at Fernald under EMSP. Radon gas is present in all atmospheres, and most of its short lived ^{222}Rn decay products attach immediately to the ambient aerosol particles, ultimately decaying to long lived ^{210}Pb/^{210}Po. The alpha emitting ^{210}Po is measured directly and used as the aerosol particle tracer. The ^{210}Po on all 6 particle filtration stages is measured in our ultra low background scintillation counters (5 counts per day) (Hallden and Harley 1957) and a deconvolution program estimates the particle size distribution from the measurements. Extensive calibrations were performed using standard latex particles, and freshly formed (unattached) ^{222}Rn decay products, to determine the deposition characteristics of the sampler over the size range from ultrafine (< 1 nm) to micrometer sized particles. Three size modes are evident in outdoor field samples, an ultrafine, a small sized (100-200 nm), and occasionally a micron sized mode.

The screens and backup filter are counted repetitively over several months to measure the build up of ^{210}Po from the ^{210}Pb deposited directly on these filtration elements, and the ^{210}Po arising from the deposited short-lived ^{222}Rn decay product chain. To show that ^{210}Pb could be measured accurately by direct counting of the filters, about 4 months of data were accumulated and measured both by alpha counting and radiochemical analysis of the filters. The alpha counting of the backup filters with low background counters was shown to provide excellent agreement with dissolution of the filter material and radiochemical measurement of the ^{210}Pb (Fisenne et al. 1996).

The collection efficiency of particulates on fine mesh screens has been studied theoretically and experimentally for many years (Thomas and Hinchliffe, 1972; Cheng et al. 1985; Reineking, Porstendorfer 1986; Holub et al. 1988; Hopke et al. 1992;). Two publications provide data on intercomparison exercises using various screen diffusion batteries (Cheng et al 1994; EML 1994). A deconvolution program is necessary to generate the particle size distributions from the alpha counting data on the 6 collection stages. This is accomplished using an Extreme Value Estimation program (EVE) that provides an accurate evaluation of the particle size distribution (Paatero 1990).

Two background locations in New Jersey are monitored synoptically for the normal, natural particle size distribution. These data are used for quality control and comparative purposes.

Results

Figure 1 shows the particle size sampler. Figure 2 and 3 show the particle size measurements made on top of the ^{226}Ra silos at Fernald from March to August, 2002, and from March to June, 2002 at the New Jersey quality control site.

The Fernald site outdoor measurements show a median particle diameter of about twice that of the New Jersey location (0.2 versus 0.1 micrometer). The reason for this is not known but may be related to the ongoing remediation activities with considerable traffic compared with a residential New Jersey area. The ultrafine mode diameter also differs by a factor of two with Fernald having the smaller size. The dosimetric consequence of such size difference is that, for equal concentrations of

radioactivity, the bronchial dose at Fernald will be less than that at the residential home in New Jersey.

The personal aerosol particle size sampler we have developed is capable of measuring any deposited alpha emitting radionuclides, or any substance of interest depending upon the measurement technique applied for the 6 stages. Thus the sampler can measure not only ^{210}Pb decay product aerosol size distribution and air concentration, even at normal background concentrations, but any long lived alpha emitting nuclides, if they are present when sampling.

Very little has been done for personal exposure assessment with regard to the inhaled particle size distribution. Reasons for this, are the practical difficulties in assessing particle size. The measurements with alternative instruments can be very labor intensive, cover only short time periods, mainly require real time instrumentation, are not suitable for personal use, and are expensive.

The inhaled aerosol particle size is the major determinant of the radiation dose to the lung, because this determines the fraction that will be deposited on the airways and in the lower lung (Harley et al 1996). As yet, there are no long term personal size distribution data for the inhaled aerosol particle size distribution of any radioactive nuclide during DOE remediation activities. In most cases when occupational lung dose is calculated the particle size is assumed to be of nominal value, namely 1 micrometer. This new particle size sampler can fill an important gap in exposure assessment at DOE facilities.

We have redesigned the present particle size sampler into a lightweight plastic version of with more filtration stages, and thus somewhat greater size resolution. It is expected that this inexpensive instrument will be fabricated and available in 2003.

Acknowledgment. Support for the project under USDOE Environmental Management Science Program (EMSP) Contract DE FG07 97ER62522, is gratefully acknowledged .

References

CHENG, YS., YEH, HC., BRINSKO, J., Use of wire screens as a fan filter. Aero Sci. Tech 4;(1985) 165-174.
CHENG, Y.S., YU, C.P., TU, K.W. Intercomparison of activity size distributions of thoron progeny by alpha and gamma counting methods. Health Physics 66: 72-79; 1994.
EML. International intercalibration and intercomparison measurements of radon progeny particle size distribution. USDOE, Environmental Measurement Laboratory Report EML-589, July 1997.
FISENNE, IM., KELLER, HW, KELLER, EW. ^{210}Pb in Indoor Air by Total Alpha Measurement Health Physics 71, (1996) 723-726.
HALLDEN, N.A.; HARLEY, J.H. An Improved Alpha-Counting Technique. Anal. Chem. S32 : 1861 (1960.
HARLEY, N.H.; COHEN, B S.; ROBBINS E. S. The Variability in Radon Decay Product Bronchial Dose. Environment International 22: S959-964; 1996.
HARLEY, NH., CHITTAPORN, P., FISENNE, IM., PERRY, P. ^{222}Rn decay products as tracers of indoor and outdoor aerosol particle size . J. of Environmental Radioactivity 51:27-35;2000.
HOLUB, R.F., KNUTSEN, E.O., SOLOMAN, S. Tests of the graded wire screen technique for measuring the amount and size distribution of unattached radon progeny. Rad. Prot. Dosimetry 24: 265-268; 1988.
HOPKE, P.K., RAMAMURTHI, M., KNUTSEN, E.O., TU, K.W., SCOFIELD, P., HOLUB, R.F., CHENG, Y.S., SU,. Y.F., WINKLMAYR, W. The measurement of activity weighted size distributions of radon progeny: Methods and laboratory intercomparison studies. Health Physics 63: 560-570;1992.
PAATERO, P. The Extreme Value Estimation Deconvolution Method with applications in aerosol research. University of Helsinki Report Series in Physics, HU-P-250, Helsinki, Finland 1990.
REINEKING, A., PORSTENDORFER, P. High volume screen diffusion batteries and alpha spectrometry for measurement of the radon daughter activity size distributions in the environment. J. Aero. Science 17: 873-880; 1986.
THOMAS, J.W., HINCHLIFFE, L.E. Filtration of 0.001 µm particles by wire screens. Aerosol Science 3, 387-393, 1971.

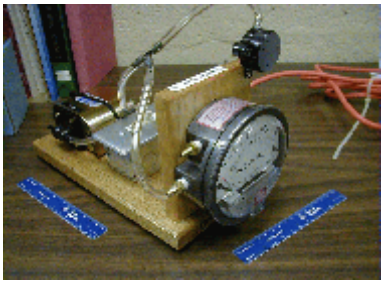

Figure 1. Continuous particle size sampler

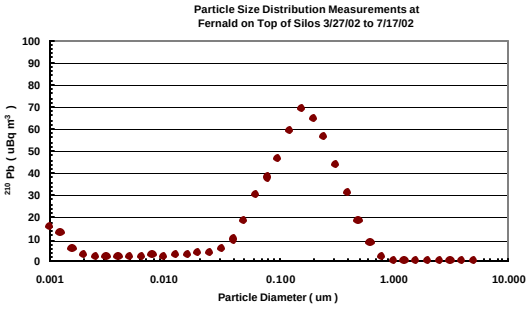

Figure 2. Measured particle size sampler at Fernald radium silos

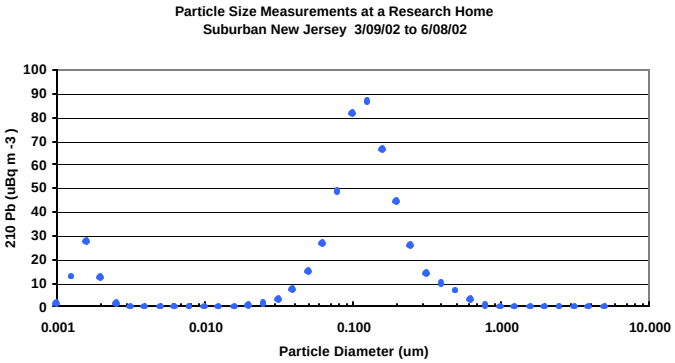

Figure 3. Measured particle size distribution at a suburban New Jersey Home