A TAGGED AEROSOL GENERATOR FOR TESTING CTBT ATMOSPHERIC RADIONUCLIDE AEROSOL SAMPLERS

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ABSTRACT

Gamma-ray spectrometry of air particulates collected on filter paper can be a very useful technique for determining the presence of nuclear fission products in the environment. Such airborne fission products could indicate clandestine nuclear weapons testing, nuclear fuel reprocessing or the catastrophic failure of a nuclear reactor. These atmospheric aerosols are collected and analyzed by high-volume air sampler/analyzers at many locations around the world to monitor compliance with the Comprehensive Nuclear-Test-Ban Treaty (CTBT).

For airborne radionuclide samplers, the CTBT specifications require a total collection efficiency of 80% or greater for 0.2-µm particles as well as a total collection efficiency of 60% or greater for 10-µm particles. To verify that CTBT samplers meet these specifications, a Tagged Aerosol Generator will produce aerosol particles with two known size distributions: a polydisperse distribution with diameters around ***0.25*** µm and a monodisperse distribution with a diameter of 10 µm. For calibration, one size distribution is generated; these particles are tagged with short-lived radon progeny (^{218}Po), then injected into the apparatus to be tested. Sampler collection efficiency for a given aerosol can be determined by measuring the activity of the radon progeny deposited on the air filter.

Particles of the required diameter are generated by commercially available instruments (Palas AGF 2.0 for the 0.25-µm diameter particles, and the TSI 3450 Vibrating Orifice Generator for the 10-µm diameter particles). These particles are neutralized in a TSI 3054 high-flow neutralizer and injected into a 100 l Attachment Volume, along with radon gas from a 1.5-MBq radon source. The Attachment Volume is a cylinder 150 cm tall and 30 cm in diameter; the major design constraint is provided by the requirement to produce 10-µm diameter tagged particles. The settling velocity of such particles is 18 cm min^{-1}; given the attachment rate of ^{218}Po, this implies a tall attachment vessel with low flow rates. Assuming a particle concentration of 10^5 cm^{-3} for the 0.25-µm diameter particles and 50 cm^{-3} for the 10-µm diameter particles (Liu et al., 1974), the attached fractions are 0.98 and 0.55, respectively. This is computed using standard attachment theory (Fuchs, 1964; Postendörfer, 1979, Raabe, 1969) using a diffusion coefficient of 0.07 cm^2 sec^{-1}, which is appropriate for neutral polonium dioxide (Goldstein and Hopke, 1998). A screen (Cheng, et al., 1980) at the exit of the attachment volume is used to remove the unattached aerosol from the air stream. For a sample time of 15 minutes, this should produce an activity on the sampler filter of 140 Bq and 70 Bq, for each particle size, respectively (Evans, 1969).

Key Words: radioactive aerosol, tagged aerosol, radon, aerosol sampler

Introduction

Radionuclide-labeled particles are often used as tracers in lung deposition, pharmacokinetic, and scientific studies. For example, Smith and Cheng (1998) used monodisperse silver particles ($10<d_p<40$ nm) tagged with ^{212}Pb, a decay product of ^{220}Rn with a 10.6 hr half life, for gamma scintigraphy in respiratory tract deposition studies. The decay products of ^{222}Rn also emit gamma rays (Table 1) and can also be used as tracers for calibration and research. Given their high activity and short half-lives, which minimizes counting time and any possibility of contamination, they are a reasonable choice in this application where gamma detection is already used to measure airborne radionuclides.

Methodology

Fig. 1 shows a schematic of a proposed instrument, which consists of the following components:

- the aerosol generator(s), used to create aerosols with a known particle size distribution,
- the charge neutralizer, needed to produce high, stable aerosol concentrations,
- the radon source, used to produce a short lived radioactive element (^{218}Po) to tag the aerosol,
- and the attachment chamber, in which the ^{218}Po becomes attached to the aerosol particles.

The characteristics of each of these components will be examined in some detail.

Aerosol Generators

The aerosol material is sebacic acid dioctyl ester doped with fluorescine, a fluorescent dye. The aerosol would be tagged with both ^{218}Po and fluorescine, providing two independent methods of verifying the sampler efficiency (assuming that the filter material could be removed from the instrument). Using the radioactive aerosol would test both the aerosol collection efficiency and the gamma counting efficiency of the sampler, while a fluorescence measurement would verify aerosol collection efficiency only.

No one piece of equipment can be used to generate particles over the size range of interest (***0.2*** to 10 µm). A vibrating orifice will be used to generate large particles and a nebulizer to generate small diameter particles. Each is sufficiently simple so that having two different aerosol generators is not a design issue.

A vibrating orifice generator, in this case the TSI Model 3450, produces a monodisperse aerosol by applying a periodic disturbance to a liquid jet. Droplet diameters between 10 µm and 200 µm are possible and are determined by the orifice diameter, the flow rate of the fluid through the orifice and the vibration frequency of the orifice plate. The liquid is a mixture of solvent (propanol), which evaporates rapidly from the droplet, and solute (sebacic acid), which remains. Final particle diameter is determined by the solute to solvent ratio. The generator has been tested with a pure propanol solution using both a 20 µm orifice and a 10 µm orifice. The former is easier to use but operates at a lower frequency and generates fewer particles (lower particle concentration). Concentration of the 10 µm particles (N, cm^{-3}) is:

$$N = \frac{f \; 60 \; e}{Q_V}$$

f = orifice vibration frequency (275 kHz)

e =efficiency (0.5)

Q_V = volumetric flow rate (25 lpm)

For the parameters listed above, the particle concentration in the attachment volume is expected to be about 330 cm^{-3}, well above the value of 50 cm^{-3} assumed in the design study.

Droplet diameter (d) is:

$$d = \left(\frac{6 \; Q}{\pi \; f}\right)^{\frac{1}{3}}$$

Q =liquid flow rate (0.08 $cm^3 min^{-1}$)

f = vibrating orifice frequency (275 kHz)

which for the listed parameters is 20.97 µm. This implies a solvent to solute ratio (equal to the ratio of the generated particle diameter to the desired particle diameter to the third power) of 9.24:1. Using a syringe with a 20 cm^3 capacity gives an operating time of 4 hrs, which is more than adequate to perform the calibration.

An atomizer (nebulizer) produces a polydisperse aerosol with a count mean diameter of ***0.25*** µm and a geometric standard deviation ≤2. In the atomizer, compressed air is used to form a high velocity jet flowing over a tube leading to the liquid reservoir. Liquid is drawn up through the tube and atomized by the jet of air. Large particles are removed from the air stream by impaction and in a cyclone, and the resulting aerosol leaves the atomizer through a fitting at the top of the reservoir. A commercially available instrument, the Palas AGF 2.0, has been tested for this application; for a flow rate of 15 lpm, particle concentrations of $>10^6$ cm^{-3} are easily obtained. Once again, final particle size is controlled by the solvent to solute ratio.

Radon Source

Sources with activities of up to 3700 kBq are available commercially, and can be transported without special permits. A source with an activity of 3000 kBq would be sufficiently large for this application. At a flow rate of 25 lpm in the attachment chamber, such a source would produce an average radon concentration of 15 kBq m^{-3}. Assuming 50 percent attachment of the ^{218}Po, this will provide a signal far above background. For example, the activity of a NIST calibration source for an NaI well detector using ^{137}Cs at 662 keV, half life of 30.17 years, is 164 Bq. For comparison, the maximum activity of ^{214}Pb in the instrument to be tested would be about 800 Bq, assuming a ^{218}Po concentration of 3700 Bq m^{-3} on the attached aerosol, a flow rate of 25 lpm, and a run time of 120 minutes (Evans, 1969).

Neutralizer

Both the nebulizer and the vibrating orifice produce particles that are charged or multiply charged, and thus have lower diffusion coefficients than uncharged particles. Particle charge depends on many factors, such as particle diameter and composition; high particle charge can cause drastically reduced particle output and unstable particle concentration. Neutralizers using ^{85}Kr in a sealed stainless steel tube for flow rates up to 150 lpm are available commercially, and routinely used to produce an aerosol of known charge distribution, and high, stable particle concentrations.

Attachment Volume

The attachment coefficient β (cm^3 sec^{-1}) can be written as (Postendorfer et al., 1979):

$$\beta = \frac{\pi v_{avg} d^2 (\Delta + 0.5d) D}{4D(\Delta + 0.5d) + 0.25 v_{avg} d^2}$$

Here d is the target aerosol diameter, v_{avg} is the average velocity and D the diffusion coefficient of the ultrafine polonium aerosol, and Δ the mean apparent free path, modified for diffusion effects at larger particle diameters (Fuchs, 1964). For aerosol particles with diameters greater than 1 µm, the attachment coefficient is proportional to dD while for particles with diameters less than 0.1 µm, the coefficient is proportional to $d^2 v_{avg}$.

For these calculations the diffusion coefficient (D) of the polonium aerosol is taken to be 0.07 cm^2 sec^{-1}, 34 nm $\leq \Delta \leq$ 63nm, and v_{avg} is 1.30×10^4 cm sec^{-1}. The attached fraction of the aerosol (A) as a function of residence time in the attachment chamber is given by (Raabe, 1969):

$$A = 1 - [\frac{\lambda_1}{\lambda_1 + \lambda_a} \ (1 - \exp - (\lambda_1 + \lambda_a)t)].$$

Here λ_1 is the decay constant for ^{218}Po (0.227 min^{-1}), and λ_a is the attachment constant for polonium-aerosol attachment (ß•N, the attachment coefficient times the aerosol concentration). For the system parameters quoted above, ß•N>2.5 min^{-1} for both the 300 µm and the 10 µm distributions, indicating high attachment fraction (>0.8) and excellent system performance.

A 100 mesh screen at the end of the attachment chamber would serve as a filter to remove from the exiting flow the unattached aerosol which has a particle size (0.5 nm) far outside the operating range of the sampler. The high diffusivity ultrafine aerosol would be captured on the screen while the larger diameter particles pass through into the instrument.***[; it must be removed since its diameter of 0.5 nm]*** Aerosol losses from impaction and interception on the screen can be computed (Cheng et al., 1980) as follows:

Interception efficiency (n_r) is given by:

$$n_r = (2k)^{-1}[(1+R)^{-1} + (1+R) + 2(1+R)\ln(1+R)]$$

while impaction efficiency (n_i) is given by:

$$n_i = (2k)^{-2} I \ St;$$

$$I = (29.6 - 28a^{0.62})R^2 - 27.5R^{2.8}$$

$$St = \frac{2ra_p^2 CU}{9ma}$$

For the system under consideration, k=0.22, St≈0.068, I≈0.1, and n_r≈0.04, n_i=0.03.

One problem with using radon progeny to tag the aerosol is that, given the high flow rates of the atmospheric samplers (8000 lpm) the activity from the radon progeny normally in the atmospheric may, under some circumstances, approach that of the tagged aerosol on the filter. This will need to be determined for each sampler being tested; it can be handled either by taking a background measurement before and after the calibration, or by supplying filtered air to the sampler itself. While this complicates the measurement, it does not make it significantly more complex or less accurate.

CONCLUSION

By using commercially available equipment the design and assembly of the tagged aerosol generator is greatly simplified. Generator stability and running time appears to be more than adequate for this application. This provides a simple and cost effective solution to the problem of performance verification of the atmospheric aerosol samplers.

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Table 1. Gamma Ray Emission From Radon Progeny

Gamma Ray Energy (keV)	Radionuclide	Emission Probability
242	^{214}Pb	0.04
295	^{214}Pb	0.19
352	^{214}Pb	0.36
609	^{214}Bi	0.46
1120	*^{214}Bi*	*0.15*
1765	*^{214}Bi*	*0.16*

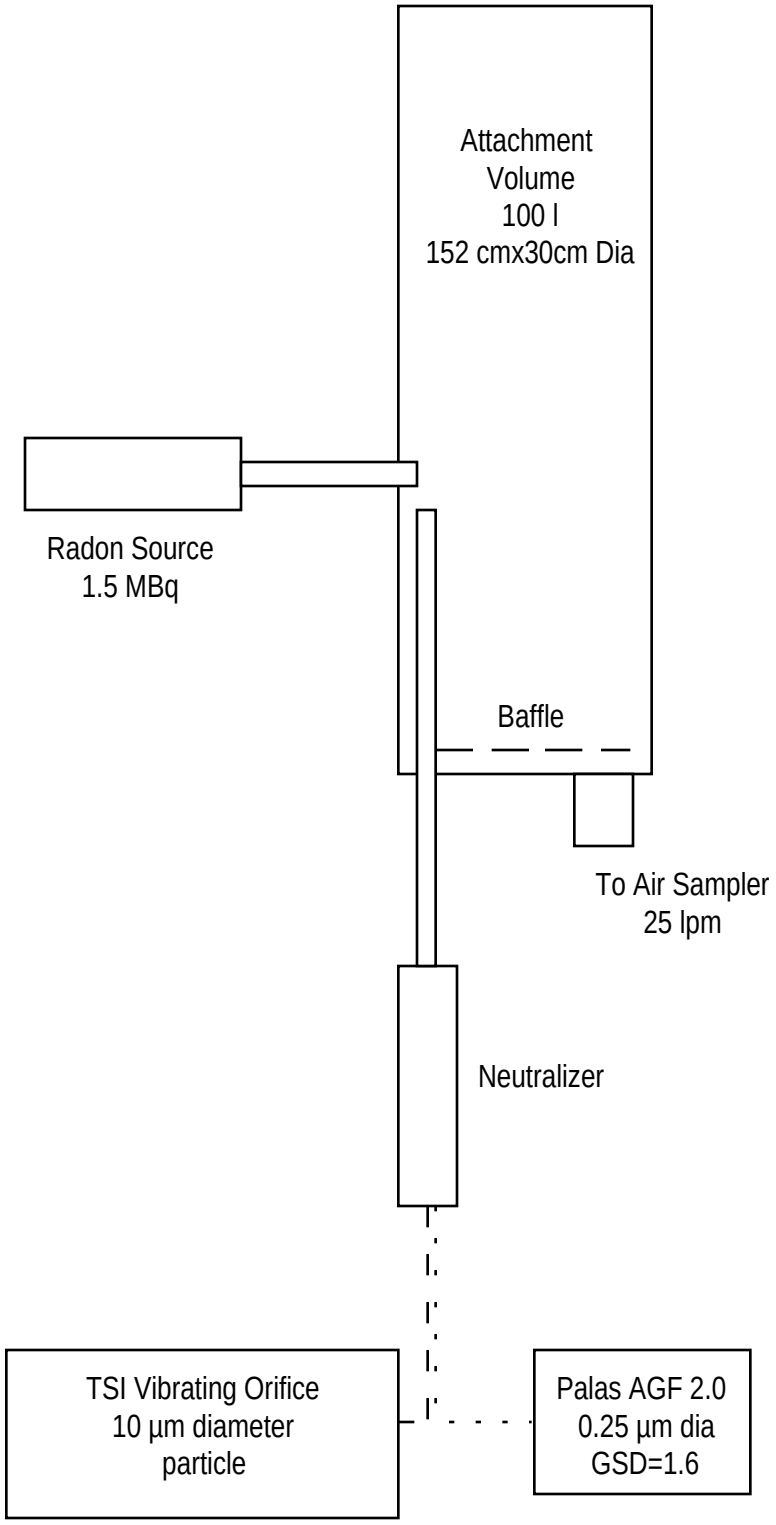

Figure 1. Schematic of the Tagged Aerosol Sampler, showing the particle generators, neutralizer, radon source and attachment volume.