A TRANSURANIC AEROSOL MEASUREMENT SYSTEM: PRELIMINARY RESULTS

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Abstract

We have completed the design, fabrication, and assembly of a computer-based prototype system for the measurement of transuranic aerosols in the workplace and environment. This system (called WOTAMS for Workplace Transuranic Aerosol Measurement System) incorporates two detectors: (1) an in-line solid-state alpha detector that sends out an alarm the moment a transuranic release occurs, and (2) an in-vacuum detector that increases off-line-analysis sensitivity. The in-line sensitivity of the system is better than 5.0 MPC-h, and the in-vacuum sensitivity exceeds 0.5 MPC-h.

Introduction

Systems which use alpha spectroscopy to monitor airborne transuranic aerosols must do so against a large background of natural alpha radiation. Although ^{222}Rn and ^{220}Rn are noble gases, their charged daughter products readily attach to particles in air, and are thus collected along with the transuranics. Under normal conditions, radon (^{222}Rn) and thoron (^{220}Rn) concentrations at locations 1 m above ground level range from 0.04 to 0.4 pCi/l (Ref. 1). Since this is from 20 to 200 times the 40-h occupational maximum permissible concentration (MPC) of soluble ^{239}Pu, the radon and thoron background constitutes a formidable obstacle to the detection of transuranics.

Since most alpha spectroscopic detection is done in air, the energy degradation of the alpha particles lowers the energy resolution and thus limits the system's sensitivity. Earlier articles have described a WOTAM system which improves the resolution by performing the spectroscopy in a vacuum.[2,3] In this paper we report on this system's sensitivity and describe an algorithm that can account for radon and thoron background interference.

WOTAMS

The WOTAMS uses either a house vacuum or external pump to draw air through a filter medium over a predetermined period of time. An in-line detector measures alpha activity on the filter as the sample is being collected. Should a detectable release occur at this time, the detector will very quickly send out an alarm. Once the collection has been completed, we shut off the air flow and advance the filter medium through the vacuum system for a more sensitive analysis.

Figure 1 shows the in-line and vacuum-spectroscopy detectors, along with the air-monitoring and filter-transport systems. Before the filter medium can be advanced, it is necessary to close the sample-line vacuum valve, vent the vacuum chamber, and release the lower pressure plate. It takes 52 seconds after a count before the next in-line count can be made; during this time, a vacuum pump rapidly lowers the operating pressure of the detector chamber.

The WOTAM system is controlled by an NSC 800 computer system, which processes data and displays it on a 64 × 256 dot-matrix, liquid-crystal display (the display may consist of spectral plots, status-including alarms, or trend data) The computer also controls filter transport, as well as monitoring other points in the system to ensure reliable operation. The keypad may be used to (a) select display modes, (b) control the system, and (c) change the system's calibration. A small front-panel alarm sounds whenever the system detects transuranics or equipment malfunction (one of two front-panel lights indicate which type of alarm has occurred). WOTAMS uses an algorithm developed specifically for the subtraction of background from radon and thoron activity.

Figure 2 shows an external view of the WOTAMS (the alarm panel is visible to the left), and Fig. 3 presents a similar view with the door open.

Radon and thoron background

The radon decay chain begins with uranium-238, which decays through five steps to ^{226}Ra with the heavy nuclides fixed in place. The next decay product, radon, does not stay fixed. A noble gas with a 3.8-day half-life, radon diffuses readily through the material containing the ^{238}U and thus escapes into the surrounding atmosphere.

Thoron originates from the decay chain which starts with naturally-occurring thorium-232. This too is a noble gas which readily diffuses into the atmosphere.

Figure 4 shows the decay schemes for radon and thoron. These inert gases do not directly interfere with the detection of plutonium or other transuranics, but their charged daughters do attach to the filter medium and so produce a background of alpha activity. There are five isotopes of these daughters that emit alpha particles: RaA, RaC', ThA, ThC, and ThC'. The other daughters emit non-interfering beta particles. The short-lived daughter products of radon and thoron can approach radioactive equilibrium whenever there is no opportunity for separation from the parent elements. This situation may obtain for outdoor inversions, though it is much more likely to occur in unventilated buildings, since ventilation tends to replace the contaminated air with fresh air from outside and so reduces the radon and daughter product concentrations markedly. This reduction only occurs, of course, if the ventilation rate is high enough to remove the radon before it can reach equilibrium with its daughters. Because the half-life of the radon daughters is shorter than that of the thoron daughters (30 min compared with 10.6 h), they reach sampling equilibrium on the filter more rapidly. Thoron is normally present

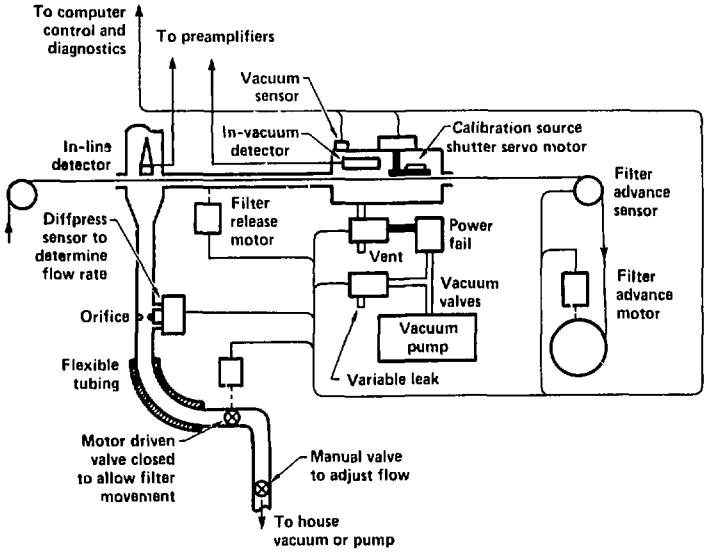


Figure 1. WOTAMS detectors (in-line and vacuum-spectroscopy), air-monitoring and filter-transport systems.

Figure 2. External view of WOTAMS.

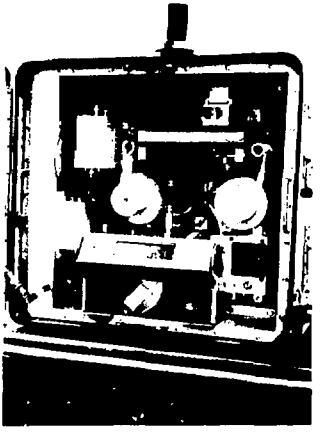
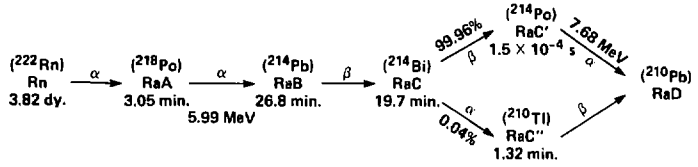
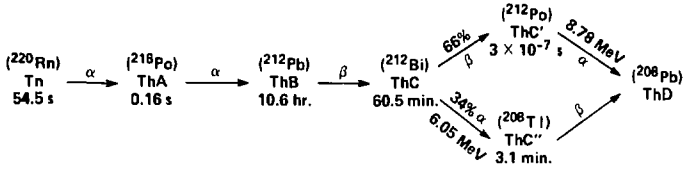


Figure 3. View of WOTAMS with door open.



(a) radon (^{222}Rn)



(b) thoron (^{220}Rn)

Figure 4. Decay schemes of radon (^{222}Rn) and thoron (^{220}Rn).

in the range of one-tenth to one-hundredth the radon concentrations.

During sampling at a flow rate V, the radon daughter atoms that collect on a filter medium obey the following time relations:

$$\frac{dN_i}{dt} = \lambda_{(i-1)}N_{(i-1)} + Q_iV - \lambda_iN_i$$

where

N_i = number of atoms of the i^{th} isotope on the filter,
t = time in min,
λ_i = decay constant of the i^{th} isotope in min^{-1},
Q_i = air concentration of the i^{th} isotope in atoms/l,
V = sampling flow rate in l/min,
i = radon or thoron daughter products.

Solving this set of differential equations for an initial atom concentration of zero is tedious but straightforward. For the period of high-sensitivity off-line analysis after the sampling is terminated, a similar set of differential equations describes the buildup of daughter atoms on the filter medium. In this description, V = 0 and the initial atom concentration is given by the final sampling-period concentration.

Having obtained th solutions to these differential equations, we could next introduce them into a computer code for calculating the daughter activity on the filter paper. The code describes this activity as a function of time from the start of sample collection. Assuming initial air concentrations of 0.1 pCi/l for each of the radon daughters, and one of 0.01 pCi/l for the thoron daughters, and introducing collection and counting times of 60 min, we were able to illustrate the variation in alpha-emitter activities with time.

Figure 5 presents the results of this calculation. ThA (not shown in the figure) is the first daughter product of thoron, but since it has only a 0.16-s half-life it decays before it can be collected on the filter medium and is thus of no concern. RaA has a 3.05-min half-life and thus reaches an equilibrium value within a few minutes of the start of collection. The RaC', ThC, and ThC' activities on the filter

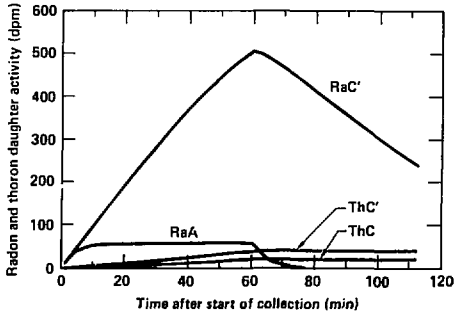


Figure 5. Radon and thoron daughter activity on a porous teflon filter medium.

paper continue to increase throughout sample collection. Once the collection is complete (60 min from the start of collection), the RaA and RaC' isotopes begin to decay away, and continue to do so over the next 60 min. The ThC and ThC' isotopes, on the other hand, remain almost constant, growing slightly during the 60-min post-collection period.

Figure 6 shows a typical background spectrum. The alpha particle background in the plutonium counting window is made up of 5.99-MeV alpha particles from RaA, 6.05-MeV alpha particles from ThC, and 7.68-MeV alpha particles from RaC'. The easiest method for eliminating the RaA interference in the in-vacuum system is to wait about 30 min for the element to decay. This way, while the previously collected sample is being counted in the vacuum, the in-air detector can be used to detect any immediate release of activity. A fraction of the RaA, ThC, and RaC' peaks can be subtracted from the count in the plutonium window to account for the radon and thoron background contribution.

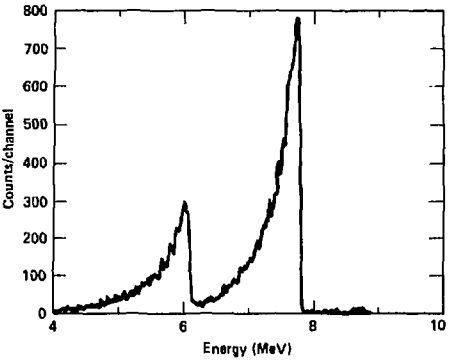


Figure 7. In-line spectrum.

Spill-over factors

To determine the fraction of the radon and thoron daughters which spill over into the counting window of interest, we made background measurements at two locations having different radon-to-thoron ratios. (The variation in these ratios was important because it enabled us to solve two simultaneous equations with two unknowns in determining the spill-over factors.) At the first location we collected and counted background for 72 h using a 1-h cycle time. More specifically, we collected radon and thoron daughter activity for 60 min, advanced the filter medium to the vacuum system, and then started the collection of a new 60-min sample, reiterating the process over a period of 72 h. During collection, four consecutive 15-min counts were made, whereas two consecutive 30-min counts were made after advancing the filter medium to the vacuum counting chamber. At the second location we collected and counted background for 24 h, using the same 1-h cycle time. We then combined the individual spectra from each 15-min in-line detection period at both locations into a single spectrum having less statistical variation than any of its 15-min segments. A similar combination was done for the two 30-min intervals of off-line (vacuum) analysis. This gave us six spectra at each location: four in-line and two in-vacuum.

The 0- to 15-min count for one of the locations (see Fig. 7) shows the effect of energy degradation in air: the tail that appears in the spectrum clearly spills over into any lower energy window that may be of interest. Figure 8 shows the 30- to 60-min spectrum of the same sample, only this time measured in the vacuum system. The reduced widths at half maximum of this spectrum shows that the energy resolution of the vacuum system has greatly improved. We used all of the spectra to calculate the fraction of the background peaks which spilled over into the plutonium window for both the in-line and in-vacuum detectors. The fractions for the in-vacuum case were determined only for the 30- to 60-min count, since by then the RaA isotope (3.05-min half-life, 5.99-MeV peak energy) had all decayed

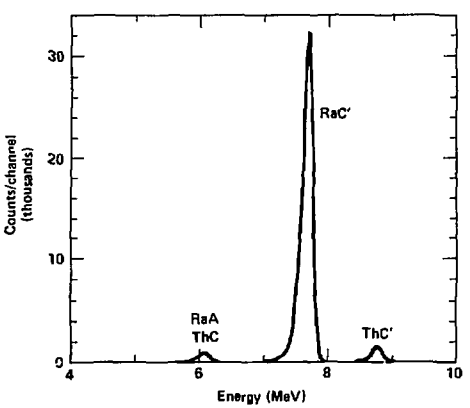


Figure 6. A typical background spectrum.

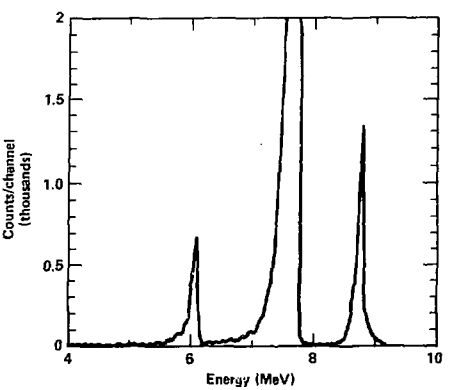


Figure 8. In-vacuum spectrum.

away and only two of the background peaks (ThC at 6.05 MeV and RaC′ at 7.68 MeV) produced any spillover into the plutonium window. After 2.83% of the counts in the RaC′ window are subtracted from the RaA and ThC window, we subtract 37.6% of the remaining counts from the plutonium window. In addition, 0.63% of the RaC′ counts must be subtracted from the plutonium window. The remaining counts in the plutonium window are produced by plutonium.

Sensitivity

The first requirement in calculating the system's sensitivity is to determine the geometry factor for the disk source and disk detector. The geometry factor, G, is defined as the fraction of the alpha particles emitted from the source that produces counts in the detector. The sensitive area of both detectors now used in WOTAMS is 490 mm^2, and the source is a 2027-mm^2 disk (radius 25.4 mm). The in-line detector-to-source distance is 10.3 mm and the in-vacuum distance is 3.5 mm. Since the sensitive surface for the detectors is recessed 2.5 mm, and this recession provides some collimation, a Monte Carlo radiation-transport code called TART[4] was used to calculate the geometry factors for both the in-line (G = 0.0662) and in-vacuum (G = 0.0796) detectors.

From the measured backgrounds in the two locations of differing radon and thoron activity we calculated the minimum detectable MPC-h, assuming a 2 cubic-foot per min flow rate, and using Pasternak's equation[5] for the minimum detectable counts (with errors of the first and second kind equal to 0.05). The results of this calculation are shown in Table 1. For the in-line detector, the MPC-hours did not vary significantly over each of the 15-min intervals. In the case where the radon background was as high as 0.324 pCi/l, we were able to see better than 2.7 MPC-h, and we could see better than 1.3 MPC-h for the case where the background was relatively low (0.043 pCi/l). With the in-vacuum detector we were able to see 0.29 MPC-h in the presence of high-radon background, and 0.15 MPC-h in the low-radon background case. Neither the counting time nor energy intervals were optimized for these calculations, though we do not expect that optimization would produce much improvement over these results.

Table 1. MPC-h for the in-line and in-vacuum detectors at two locations having different backgrounds.

Background	In-Line			In-Vac	
(pC/liter)	0-15 min	15-30 min	30-45 min	45-60 min	30-60 min
0.043	1.07	1.21	1.27	1.30	0.13
0.324	2.29	2.62	2.65	2.71	0.23

Acknowledgment

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