BE-7 CROSS-TALK IN RASA CONTINUOUS AIR SAMPLERS

Richard J. Arthur, Harry S. Miley, and Lindsay C. Todd

Pacific Northwest National Laboratory

Sponsored by National Nuclear Security Administration
Office of Nonproliferation Research and Engineering
Office of Defense Nuclear Nonproliferation

Contract No. DE-AC06-76RLO 1830

ABSTRACT

Several experiments were run to determine the extent of cross talk on existing RASA systems. The most definitive experiment consisted of shutting off power to the blower motor and advancing the filter media so that no exposed media was visible to the HPGe detector. The MDL for detecting Be-7 was thereby confirmed. A series of 5 blank filters were counted before power was restored to the blower motor.

Comparison of several spectra for the blank filters confirmed that levels of Be-7 were below detection levels. The blank filters were subsequently removed and counted on an ultra-low-background HPGe detector. The presence of Be-7 was clearly visible in several of the blank filters, but not in others.

Several theories are advanced to account for the presence of trace levels of Be-7 in the blank filters. Changes to the commercially available RASA are proposed based on the findings of this study. While these possibilities are put forward by the authors in the spirit of producing the best possible scientific data for atmospheric science research, the authors do not feel that cross talk or any other minor problems materially reduce the value of the RASA or any similar system for detecting and reasonably quantifying the size and nature of an atmospheric nuclear event.

OBJECTIVE

The Radionuclide Aerosol Sampler/Analyzer[1] (RASA) detects natural radionuclides in air samples by collecting a large volume sample (>15,000 m^3) through a high efficiency filter, decaying the sample to reduce radon daughters, then advancing the filter automatically into a lead shield containing a large germanium detector. This arrangement allows the RASA to monitor the air for anthropogenic or natural radioactive materials in an automatic, remote, and unattended way.

Considering the use of the RASA in monitoring for fission debris in the air from use of nuclear weapons or reactor accidents, certain fission products are considered key. The RASA was designed to achieve better than 10 $\mu Bq/m^3$ of Ba-140, one such key fission isotope. In reality, however, RASA installations typically only detect primordial radionuclides (from potassium, uranium, or thorium present in the Earth's composition) such as K-40, Pb-212, Pb-214, Bi-212, Bi-214, and Tl-208 plus Be-7 that is produced in the upper atmosphere from cosmic ray proton reactions on Ar-40. A typical RASA gamma-ray spectrum is shown in Figure 1.

Be-7 is present in most RASA air samples, and with sufficient intensity to have very small statistical errors. Figure 2 shows the results of automated Be-7 analyses on a large number of RASA samples. The results have been histogrammed by the size of the statistical error bar on the Be-7 peak at 477 keV. Because most radon decay products have fairly short half lives (less that 11 hours), after a few days the only remaining material on a filter may likely be Be-7 (53 days) and low levels of the long-lived species K-40 (1.3×10^9 y). In addition, Be-7 contamination of surfaces tends to linger if caused by electrostatic precipitation from the atmosphere or contact with an unsealed filter.

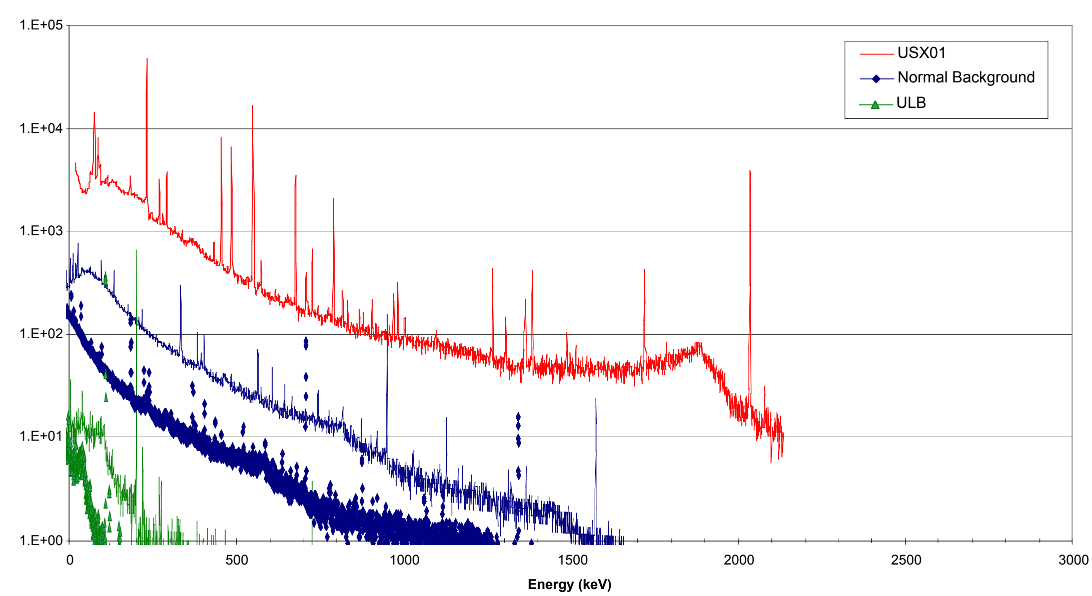

Figure 1. A typical RASA spectrum taken in Richland, Washington. Also shown is a system blank and a well decayed filter measured in an ultra-low background counter. Substantial radon daughters associated with aerosols are visible in the RASA spectrum.

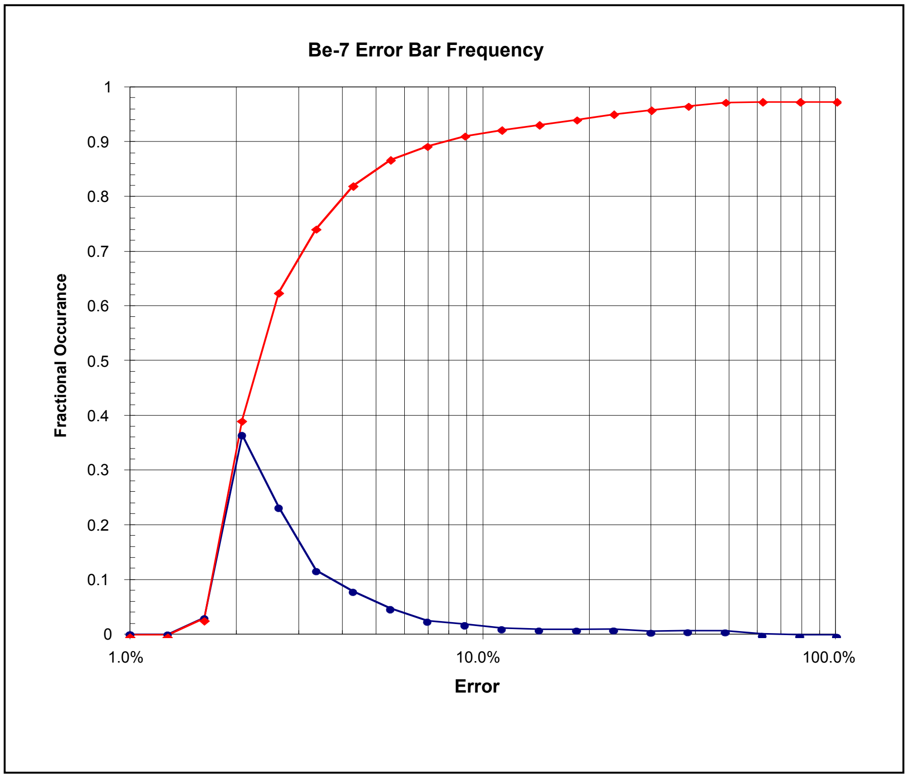

Figure 2. Plot is showing probability vs. size of statistical error bar.

The mechanics of the RASA are such that the filter area is large at the time of sampling and smaller during the radionuclide measurement. To accomplish this, the RASA employs six parallel continuous filter strips played off of long rolls, through the sample head, into a decay position, then wrapped around the germanium detector. The six strips are widely separated in the sample head, then brought together in a bundle and sealed as they advance into the decay position. In principle, then, the exposed filter samples have no opportunity to contaminate the detector area. A pair of motorized rollers advances the entire continuous filter bundle from the output side of the lead shield. Figure 3 shows a schematic layout of part of the RASA mechanical apparatus. It should be noted that substantial openings in the lead shield facilitate the entry and exit of the filter bundle.

Observation of the Cross Talk Effect

During normal operation of the RASA or any monitoring device it is occasionally required to perform a system blank. A system blank, in the case of the RASA, consists of measuring the background radiation recorded by the detector when a filter bundle is present in the shield that had zero accumulated air volume. In recent operations of this type, a slight Be-7 signal was detected. Several immediate possibilities presented themselves: that the volume of air within the shield contained that much Be-7, that Be-7 contamination within the sample head was becoming attached to blank filters, and that the substantial quantity of Be-7 on the filter before and after the blank were contributing to the blank spectrum, an effect casually labeled "cross-talk" for the purposes of this discussion.

A cursory examination of the position of the filters in the decay and output locations (labeled A and C in the diagram) shows that 7.5% and 20% of these filters, respectively, have a line-of-sight view of the detector, but at a substantially greater distance (about 5 times greater) than the filter under examination.

Original design estimates were that about 1% of the signals recorded for a given sample would come from the next sample, mostly due to short-lived radon daughters. No consideration of the contribution to a blank was made.

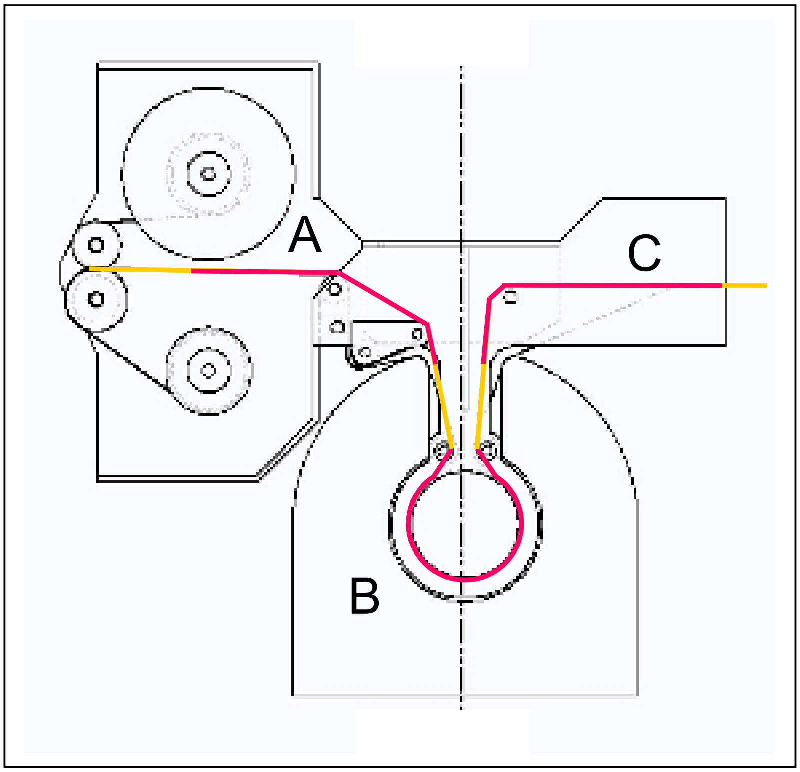

Figure 3. The basic layout of the RASA showing the decay position (A), the measurement location (B) and the output location (C). The rollers in the left hand portion of the diagram apply a polyester film to the filter to seal it.

The size of the cross talk from the decay position was estimated by comparing the integral of the Be-7 counts for a blank with a normal atmospheric sample in the decay position. The next day, the normal sample was in the shield. Only 320 net Be-7 counts were observed in the blank vs.170142 net counts when the normal sample was counted. This yields a 2.8% effect, but does not completely explain the Be-7 signals. In tests where blanks were advanced in the normal daily method into the decay, count, and output positions, some traces of Be-7 have been reported by RASA users. This seems to indicate that possibly more than one cross-talk effect is occurring.

A commercially produced RASA in operation in Richland, Washington was set to reproduce the results by disconnecting the blower, manually advancing filters until a blank was in each location, then 5 blanks were allowed to advance in the normal daily manner. After this, the blower was reconnected and the system ran in the usual manner. The samples were measured by the RASA itself, but to improve the sensitivity the samples were later counted on an ultra-low background spectrometer[2].

Table 1. Results of ultra-low background testing of RASA blanks. Note: No cross talk of gamma-rays from one sample to another is possible in this scenario since the samples were out of the RASA and in another counting system.

Filter ID	Comments	ULB Be-7 Rate	ULB Activity
02002 02704	Normal loading		
05	“		
06	“		
07	“		
08	Light loading		
09	Blank, blower off	12.5 cts/day	145 µBq
10	“	9.38 cts/day	109 µBq
11	“	<1 cts/day	<12 µBq
12	“	<1 cts/day	<12 µBq
13	“	<1 cts/day	<12 µBq
14	Blank, blower on	<1 cts/day	<12 µBq
15	“	24.6 cts/day	285 µBq
16	“	24.4 cts/day	282 µBq
17	Normal loading		
18	“		

The automated analysis of the blanks (09-16) showed either no Be-7 or a very questionable result consistent with no Be-7. The ultra-low level counting did yield interesting results, however.

CONCLUSIONS AND RECOMMENDATIONS

From the ultra-low level counting summarized in Table 1, we may conclude several facts.

First, contamination of blank filters while in the sample head, if it occurs, contributes less than 1 count per day of Be-7. Similarly, contamination from one filter to another via scraping or rubbing is limited to less than one count per day.

Second, the operation of the blower seems to cause a low level of contamination on adjacent blank filters. This contamination is 5%-10% the size of the original cross talk effect. Two theories are suggested as to the source of this contamination.

1. Be-7 on particulate adheres to the outside of the polypropylene envelope as it passes into a small leak in the sample head.

2. Be-7 in room air is filtered through a presumably blank filter between regular samples due to a leak in the sample head.

Remedies to the effects listed above include a redirected filter path to greatly reduce or eliminate the gross (~3%) effect of cross talk. This effect and remedy are rather certain. Another, so-far untested remedy would be to seal the known gaps in the commercial RASA sample head so no air is allowed to flow over or through media in or near the decay position.

REFERENCES

S.M. Bowyer, et al, *IEEE Transaction on Nuclear Science*, Vol. **44**, No. 3 (1997) 551-556.

H.S. Miley, et al, *J. of Radionanlytical and Nuclear Chemistry, Articles*, Vol. **160**, No. 2 (1992) 371-385;

R.J. Arthur, et al, *J. of Radionanlytical and Nuclear Chemistry, Articles*, Vol. **160**, No. 2 (1992) 297-304.