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Environmental measurements with a Comprehensive Nuclear Test Ban Treaty radionuclide particulate monitor

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A Radionuclide Aerosol Sampler/Analyzer (RASA Mark 4) has been developed at PNNL for use in verifying the Comprehensive Nuclear Test Ban Treaty (CTBT). The RASA Mark 4 collects about 20,000 m^3 of air per day on a 0.25 m^2 filter. This filter is automatically decayed for 24 hours, then advanced to a germanium detector for a 24 hour count. This system has been operated in Richland, WA for a limited period of time in a pre-deployment testing phase. The germanium-detector gamma-ray spectra have been analyzed by automatic spectral analysis codes to determine Minimum Detectable Concentrations (MDC) for a number of isotopes of interest. These MDC's have been compared to other atmospheric measurements in the field and in the laboratory.

Introduction*

The Comprehensive Nuclear Test Ban Treaty (CTBT) is intended to eliminate the testing of nuclear weapons. A key to the world-wide implementation of this treaty is verification technology designed to assure participants that all parties are complying with the treaty. To verify the CTBT, many technologies will be used: seismic, infrasound, hydroacoustic, and radionuclide. Radionuclide methods differ from vibrational methods because the detection of a suite of short-lived fission isotopes like ^{140}Ba, ^{99}Mo, and others in appropriate ratios is positive proof of a nuclear detonation.

To fulfill CTBT measurement requirements, a system has been developed at the Pacific Northwest National Laboratory (PNNL) to automatically collect and measure radioactive aerosol debris, then communicate spectra data to a central data center. The development has proceeded through several design iterations which began with the demonstration of achievable measurement sensitivity (<10 μBq/SCM ^{140}Ba-^{140}La) and resulted in a fully automatic measurement system of minimal size (2 m high × 2 m wide × 1 m deep) and minimal power usage (1600 watts). This fully automatic system referred to as the RASA Mark 4 prototype (see Fig. 1) is currently the subject of an U.S. Air Force procurement with private industry to partially fulfill U.S. treaty obligations under the CTBT. It is planned that the system will be available for purchase from a manufacturer in late 1997.

All of the radionuclide and other CTBT sensors will comprise the International Monitoring System (IMS). The IMS will be comprised of in part of eighty radionuclide aerosol systems. Each one of these systems will be collecting and reporting data on a daily basis to the International Data Center (IDC). Although the primary purpose of the IMS radionuclide aerosol systems will be to monitor the air for short lived fission products, the presence of normal levels of natural isotopes confirms the proper operation, calibration, and security of aerosol systems in the IMS.

The primary objective of the Radionuclide Aerosol program at PNNL is to develop CTBT verification technology for remote, automatic, rear-real-time collection and measurement of radioactive aerosol debris. The system must achieve or exceed CTBT-mandated sensitivity as a primary objective and also must provide robust network infrastructure.

The basic characteristic of the RASA Mark 4 is its synchronized sampling, decay and counting times. For the CTBT, the mandatory reporting period is 72 hours. The RASA accomplishes this in the following way: Air is drawn through a filter for 24 hours, then the filter paper is advanced to the decay position and a new air sample is taken. After the 24 hour decay, the filter paper is advanced again so that the original sample is now in the counting position. At this time the second sample is moved into the decay position and a third air sample is begun. A sample is counting while it is wrapped around the circumference of a mechanically cooled 90% relative efficiency germanium gamma-ray detector. The power consumed by the whole system (including the air blower) is 1600 watts. More details of the operation of the RASA have been published elsewhere.[1]

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Fig. 1. RASA Mark 4

Experimental

Fission spectra

A germanium detector was chosen for its selectivity. The reason for this choice can be clearly seen in Fig. 2. Shown in Fig. 2 are three spectra, the first is a routine air sample of 12,000 SCM. The second spectrum is a 12,000 SCM air sample that has been added to fission products created in the laboratory (a uranium compound activated in a low flux neutron facility) and measured with a HPGe detector. The third spectrum is the same as the second, but only measured with a NaI(Tl) detector.

The higher resolution of a Ge vs. NaI(Tl) detector produces a measurement that allows gamma lines from the fission products to be clearly resolved. This provides high signal-to-noise ratios for these gamma-lines. In addition it minimizes (but not eliminates) the effect that radon concentrations will have on the Minimum Detectable Concentration (MDC) calculations of fission products in an air sample. The presence of radon in an air sample will elevate the Compton continuum under peaks of interest, thus raising the MDC computed for the radionuclide of interest.

Radon decay

Radon daughters are part of the naturally occurring environmental background for a CTBT particulate sensor. Increasing the shielding around a detector can compensate for elevated amounts of most environmental radionuclides (e.g., ^{40}K in a building's concrete floors). A larger shield is not an effective solution to solving the radon problem, radon daughters are actually trapped by the filter media and therefore become part of the sample. Once on the filter media these radon daughters are placed directly in front of the detector, thus rendering any amount of shielding ineffective. Another approach to partially solving the radon problem is to increase the decay time of the sample.

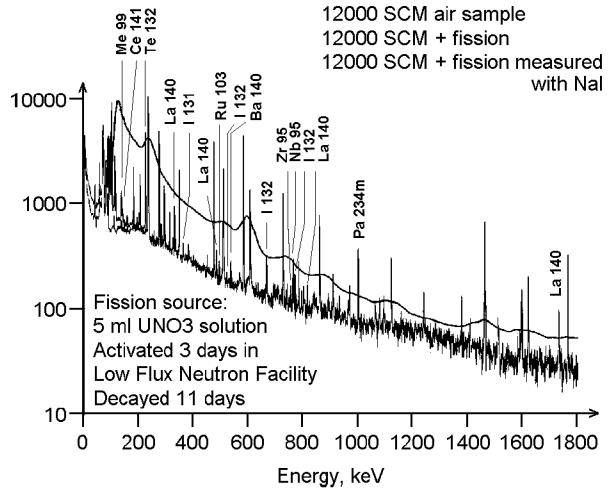

Fig. 2. Germanium/NaI comparison

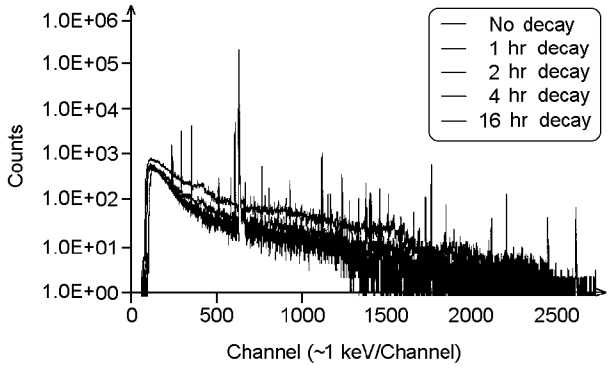

Fig. 3. Effects of radon decay. One hour counts at various intervals

Table 1. Effects of radon decay

Decay, hr	Continuum counts, ^{99}Mo	Continuum channel ^{140}Ba
None	737	109.3
1	501	51.4
2	447	36.4
4	435	28.6
16	421	28.2

The ^{222}Rn decay chain (from ^{238}U) is dominated by fairly short half-life nuclides compared to interesting fission products. Thus increasing the decay time will allow these radon daughters to decay away, while still having a negligible affect on the activity of the fission products of interest. The results of one experiment shows the effect of various decay times. An air sample was drawn and then consecutive one hour counts of the sample were taken. In Fig. 3 one can readily see a drop in the count rate as the short-lived radon daughters decay away. In Table 1 the continuum count rate is listed for the ^{99}Mo 140.41 keV and ^{140}Ba 537.38 keV gamma-lines of interest. In the first few hours the height of the continuum in these regions drops significantly. The count rate will continue to drop as the sample is decayed for even longer periods of time. Traditionally, long (e.g., a week) aerosol samples have been taken, then these samples have been decayed for three or four days to allow for the ^{220}Rn decay chain to decay. The near-real-time reporting requirements of the CTBT constrain the length of the decay and so typically the CTBT samples will only be decayed for 24 hours, although the RASA can support any designated period for its sample, decay, and counting times.

Minimum Detectable Concentrations (MDC)

There are a wide variety of ways that Minimum Detectable Concentrations (MDC) are computed. In principle each method quantifies the maximum amount of a radionuclide than can go undetected in a given spectrum given a specified confidence level. Several methods were used to calculate a MDC for a given spectrum taken by the RASA Mark 4.

A typical hand calculation of the MDC begins with the determination of the background area of interest. This is chosen as the sum of the counts in a background. For the 30 October 1996 sample, the full width half maximum (FWHM) at the ^{140}Ba 537.38 keV line is two channels, and the background area is chosen as twice the FWHM, summing these four channels (418, 400, 405, and 385) results in a sum of 1608 counts. For this background region. The square root of 1608 is 40.10 and at a two sigma level confidence factor 80.30 counts may go undetected. The efficiency of the detector at this energy is 0.02905, resulting in a gamma count of 2,760.75. Folding in the branching ratio of 0.199 results in 13,873 disintegrations. The sample was counted for 75,900 seconds of live time, so the minimum detectable activity is 1.828e-1 Bq. Since the sample volume was 15,684 SCM the MDC is 11.68 μBq/SCM.

For comparison CURRIE's critical level[2] is computed by the formula $L_c = 2.33 \times \text{sqrt}(\text{BKG})$. For the 10 October 1996 sample, this yields 13.43 μBq/SCM. CURRIE's detection limit, L_d (L_d (counts) = 2.71+4.65×sqrt(BKG)) is 27.55 μBq/SCM. A PNNL version of the code RAYGUN computes an MDC based upon the formula, $\text{MDC}_{\text{counts}} = 1.0645 \times 2.5 \times \text{sqrt}\,(2 \times \text{avgBKG}) \times \text{FWHM} \times \text{SIGMA}$, which results in a value of 38.0 μBq/SMC. As one can see these numbers vary from 11.68 to 38.0 μBq/SCM, with the hand calculation being the lowest and the RAYGUN estimate being the most conservative.

Table 2. Average RASA sensitivities

Operating procedure:	Location: Richland, WA, USA
• 24-hr sample collection	Detector: 90% rel. eff. P Type Ge
• 24-hr decay	Sample volume: ~15,000 SCM
• 24-hr acquisition	(February – March 1997 Data)

Fission product	Raygun MDC's, µBq/SCM	Radon daughter	Observed, mBq/SCM
^{95}Zr	8.74	^{208}Tl	653.0
^{99}Mo	44.28	^{212}Pb	11.6
^{103}Ru	10.44	^{214}Pb	27.6
^{131}I	20.30	^{212}Bi	50.3
^{132}I	64.29	^{214}Bi	4.56
^{132}Te	14.45		
^{140}Ba	41.14		Observed,
^{140}La	12.79	Cosmic	mBq/SCM
^{141}Ce	11.35	^{7}Be	3.4

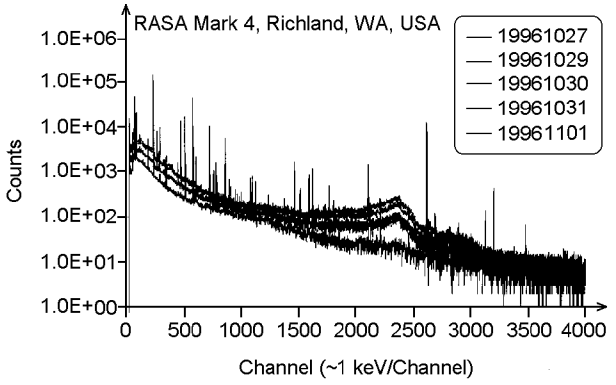

Fig. 4. RASA Mark 4 spectra. Volume: ~15,000 SCM, sample time: 24 hr, decay time: 24 hr, count time: 24 hr, detector: 90% rel. eff. Ge, filter: 0.25 m^2 (triple layer SBMF). Note: The low activity on 27 Oct 1997 is due to a rainy day

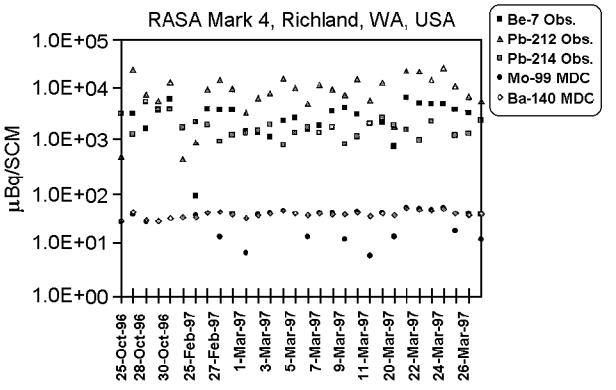

Fig. 5. RASA Mark 4 observed concentrations and MDC's. Volume: ~15,000 SCM, sample time: 24 hr, decay time: 24 hr, count time: 24 hr, detector: 90% rel. eff. Ge, filter: 0.25 m^2 (triple layer SBMF)

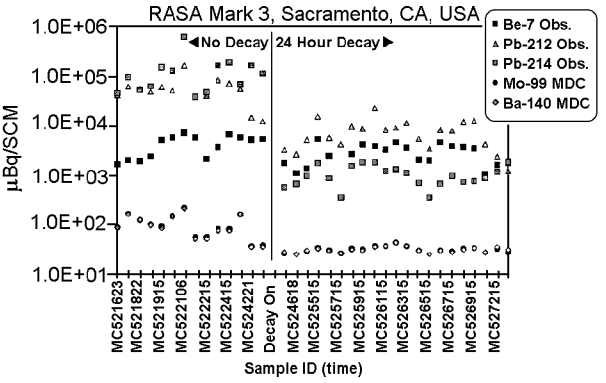

Fig. 6. RASA Mark 3 observed concentrations and MDC's. Volume: ~27,000 SCM, sample time: 24 hr, decay time: 0/24 hr, count time: 24 hr, detector: 90% rel. eff. Ge, filter: 0.25 m^2 (single layer SBMF)

Environmental data

The RASA Mark 4 has been in operation for a limited amount of time in a predeployment test phase. An earlier prototype, the RASA Mark 3, was in operation for an extended six month field test at McClelland Air Force Base in Sacramento, California, USA. Data from these sensors has been summarized in Table 2 and Figs 4, 5 and 6.

The average RASA Mark 4 MDC's shown in Table 2 are quite conservative estimates of the MDC's achievable by the RASA Mark 4. The reason for the conservative nature of these MDC's is that they were computed using RAYGUN's very conservative MDC formula. In addition, the air flow during the predeployment testing was rather minimal, achieving on average only 15,000 SCM per 24 hour sample. This low air flow is due to a constricted air ducting required to site the sampler in a pre-existing trailer. In laboratory testing, we were able to achieve over 18,000 SCM/day with a better ducting configuration.

It should be noted that average MDC values do not tell the whole story of the RASA Mark 4. In Fig. 4 five spectra are shown that were taken in the same week. One can easily see the large variation in these samples. In particular the sample taken on a rainy day shows a very low count rate in comparison to the other spectra. Trend data from the RASA Mark 4 shown in Fig. 5, illustrate how widely the observed concentrations of ^{7}Be, ^{212}Pb and ^{214}Pb and MDC values for ^{99}Mo and ^{140}Ba change over time. The level of radon in the air can vary drastically from day to day depending upon the weather (e.g., rain, wind and barometric pressure), which results in correlated changes in the limits of detection for the fission product isotopes of interest.

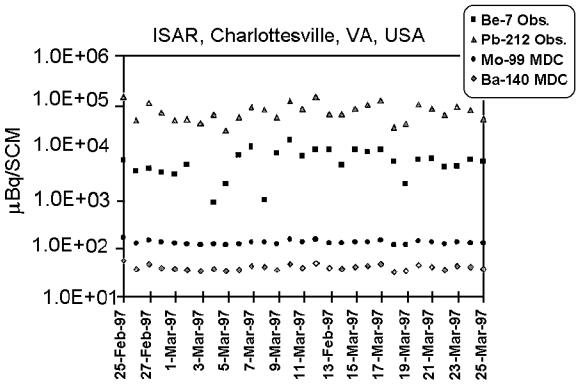

Fig. 7. ISAR observed concentratations and MDC's. Volume: ~27,000 SCM, sample time: ~24 hr, decay time: ~24 hr, count time: ~24 hr, detector: 40% rel. eff. Ge

The wide variations in observed radon daughters and therefore fission product MDC's is not limited to only the RASA Mark 4 data. The RASA Mark 3 data shown in Fig. 6 show similar variations. This data was taken over the course of several months in Sacramento, CA. The ISAR data[3] from Charlottesville, VA also shows variations in radon, (see Fig. 7). In addition to weather effects, the radon levels will vary from location to location in the International Monitoring System, due to the fact that some radionuclide stations will be located in radon rich geological areas and other locations will be radon poor.

Discussion

The detection sensitivity of the RASA Mark 4 is greatly affected by the radon concentration levels at the time of sample collection. As the radon concentration goes up the level of sensitivity of the system goes down. Thus the same system might have a sensitivity of 20 µBq/SCM on one day and later in the same week the sensitivity might be 50 µBq/SCM. The geology of a location and the weather around a sampler will effect the day to day radon concentrations of a system.

The CTBT community needs to establish a set of qualification guidelines for radionuclide sensors that are weather and geography neutral. Without the adoption of weather neutral guidelines a system might be approved for use when the testing is done on a rainy day, yet be failed if the system were tested as a low pressure zone moved through the region. Geography neutral guidelines are needed to allows for economies of scale when building samplers. Without these guidelines systems might pass when operated in a radon poor region, yet fail when operated in a radon rich region.

One possible solution for a weather and geography neutral qualification guideline is to base the acceptance of a system on a derived MDC calculation based upon a system blank. A system blank will be completely weather neutral, in that the only items being measured are contaminants on the filter media and room background that is not adequately shielded. To this blank sample a spectrum corresponding to a given amount of environmental radionuclides would be added to yield a pseudo-environmental sample. Once this pseudo-environmental sample is generated, the MDC's for the radionuclides of interest would be computed. If these MDC's are within limits the system would be approved for use in the IMS. Conversely, the same result would be achieved by adjusting the CTBT MDA's to correspond to an appropriate level for a blank.

Conclusions

A completely automated particulate system has been built that meets the requirements of the CTBT. Though designed primary to meet the CTBT requirements, valuable environmental data may be gathered from these systems. There is a wide variety of methods that are used to compute minimum detectable concentration values. Various communities have "standardized" upon given methods, but as yet the CTBT community has not selected a given method. The selection of one method or another may change the published sensitivity measurements for a given sensor.

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