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Parallel radioisotope collection and analysis in response to the Fukushima release

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Abstract Two independent radionuclide aerosol air samplers were operated at Pacific Northwest National Laboratory in close proximity during the Fukushima reactor releases. One system was an automated aerosol collection and analysis unit, whereas the other was a manual sampler of simpler design. The samples collected from each sampler showed correlation in radionuclide activity, although some variations were observed. During this unique event, the small variations observed between the co-located air samplers illustrate the effectiveness of a way to acquire useful parallel samples for scientific purposes. The results in radionuclide activity concentration show that, in some circumstances, use of a manual high volume air sampler in parallel to a complex automated sampler can produce results that are of comparable quality to International Monitoring System samples.

Keywords Fukushima · Radionuclide · Atmospheric aerosol sampling

Introduction

The Comprehensive Nuclear-Test-Ban Treaty calls for the International Monitoring System (IMS) to include 80 stations to be outfitted with high-volume aerosol samplers [1, 2]. These stations are key to confidently detecting atmospheric evidence of a treaty violation. These aerosol sampling stations are required to sample 12,000 m^3/24 h of air with a minimum sensitivity of 30 μBq/m^3 or better for Ba-140 [3]. Following the earthquake and tsunami that seriously compromised the integrity of the Fukushima Daiichi nuclear power station's containment, PNNL and other organizations deployed both IMS air samplers and "IMS-like" samplers that either reported the presence of airborne radionuclides [4–7] or collected particulate samples, respectively, to monitor for the presence of airborne radionuclides passing over the United States. Deployment of these sensitive atmospheric sampling devices allowed accurate computation of isotopic ratios to confirm the release source of the plume to be a commercial power reactor. The collection and calculation of local radionuclide measurements coupled with atmospheric backtracking models can be utilized to determine both the source location and severity of the release.

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Experimental

Pacific Northwest National Laboratory (PNNL) in Richland, Washington deployed two separate radionuclide air sampling systems in the immediate aftermath of the Fukushima release. These systems were the radionuclide aerosol sampler/analyzer (RASA) and the high volume air sampler (HVAS) [8]. The RASA system is permanently housed in a climate-controlled, sealed ROHN^{TM} communication shelter. The HVAS system was located approximately 1.6 km south of the RASA system in an open vehicle parking lot, selected as the closest placement with

sufficient infrastructure to allow continuous operation. Both system locations were free of building obstructions.

RASA system

The RASA is a particulate collection and analysis system utilized as a significant component in the IMS network (shown in Fig. 1a). It provides highly reliable, near real–time, continuous radionuclide activity concentrations for a given location [9]. The RASA collects airborne aerosol samples on 0.25 m^2 of 3 M-BMF20F filter paper arranged in six strips. The filter paper has >80 % collection efficiency for particle sizes of >0.2 μm at 120 cm/s face velocity. Samples from the PNNL RASA were collected for 24 h with an average air collection rate of 510 m^3/h, computed by an anemometer in the exhaust stream. These samples were then allowed to age for an additional 24 h before counting, in order to reduce the activity levels of collected radon decay products. Unlike those from the IMS-deployed RASA systems, the samples collected at PNNL during this period were prepared and analyzed in a laboratory setting as described below.

HVAS system

The HVAS system utilized for the collection of the aerosol samples at PNNL is illustrated in Fig. 1b. Airflow through the HVAS (1,500 m^3/h) system was higher than the set flow through the RASA during the collection of airborne radionuclides. The filter material is the same 3 M-SBMF-40VF filter paper utilized in the RASA system; however, the filter configuration is a large, single-ply sheet wrapped around an 8-in. diameter cylindrical filter holder. The HVAS utilizes an anemometer in the exhaust stream to measure the collected air volume rate in real time. Table 1 shows the collected air volumes for both the RASA and HVAS systems. All values are assumed to have an accuracy of ±10 %, which reflects variations in the average flow velocity through the exhaust stream and the location of the anemometer. The collected air volumes for both the RASA and HVAS system were calibrated to each other using the activity concentration of Be-7.

Sample processing

Following the Fukushima event, aerosol collection filters from both the RASA and HVAS were collected daily. The

Table 1 Collected air volumes for the HVAS and RASA air samplers

Collection begin (pacific standard time)	RASA air volume (m^3)	HVAS air volume (m^3)
3/19/11 15:33	19,149	31,506
3/20/11 15:15	19,728	39,055
3/21/11 15:40	19,351	37,938
3/22/11 15:37	19,095	33,789
3/23/11 15:15	19,998	40,420
3/24/11 17:00	18,355	34,476
3/25/11 15:43	19,822	34,883
3/26/11 16:15	18,786	33,933
3/27/11 15:30	19,796	36,272
3/28/11 16:00	18,853	34,928
3/29/11 15:20	19,459	37,684
3/30/11 15:25	19,863	32,658
3/31/11 16:00	19,163	32,296
4/1/11 15:43	19,149	33,627
4/2/11 15:25	19,392	35,422

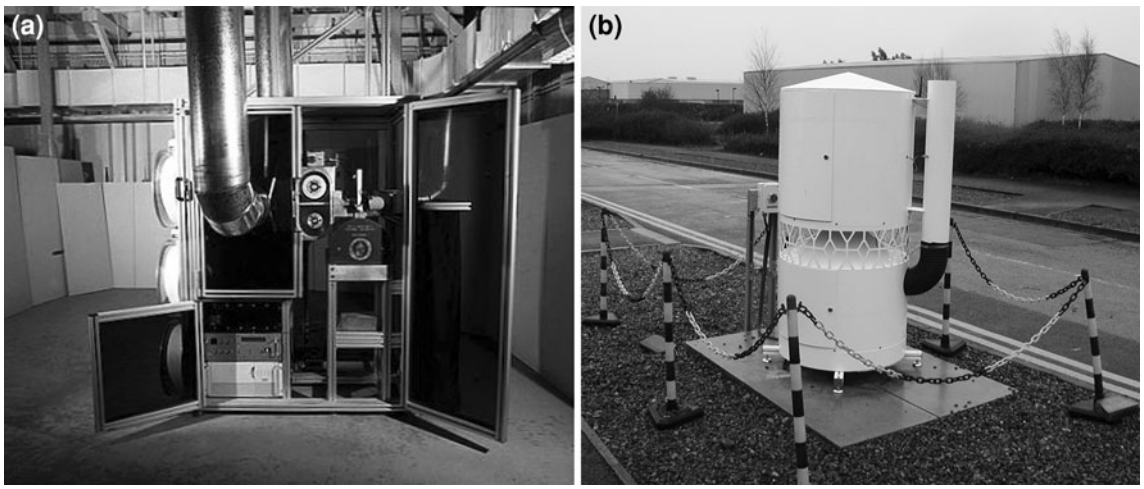

Fig. 1 **a** The radionuclide aerosol sampler/analyzer (RASA) located at Pacific Northwest National Laboratory; **b** the high volume air sampler (HVAS) co-located at Pacific Northwest National Laboratory

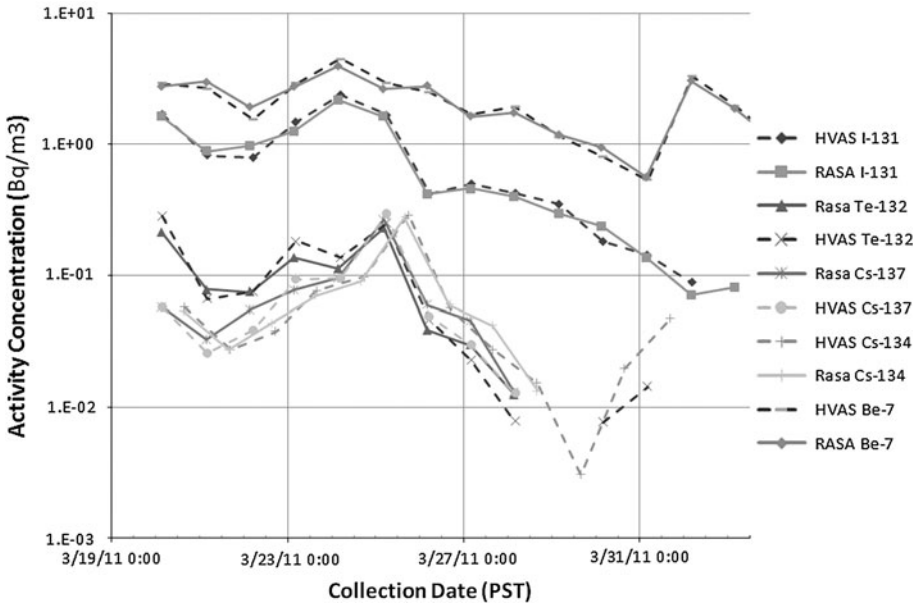

Fig. 2 Collected radionuclide concentrations in the aftermath of the Fukushima event as collected by the RASA and HVAS system for 5 days

air filter samples were delivered to the PNNL low-level gamma spectroscopy laboratory, where they were logged and prepared for counting. The HVAS filters were folded into a 10×10 cm^2, which was then sealed in a clean polyethylene bag. The RASA filters were sealed in a polyethylene bag and placed against the inside of the outer wall of a 4-L Marinelli beaker. Prior to counting, the gamma-ray detectors were calibrated in the same geometries the samples were to be counted. The counting geometry standards for the RASA and HVAS samples were created using a commercially available mixed radioisotope solution containing nine gamma-ray emitting isotopes. A blank RASA filter was impregnated with known amounts of a mixed isotope standard, placed in the counting geometry then used to calibrate the detector. The HVAS filter was impregnated with the calibration solution and then folded into the same 10×10 cm^2 geometry used for the HVAS filters and counted on a gamma-ray detector.

For the counting process, the sample was placed into a plastic holder that had been constructed so as to obtain a repeatable position with $\sim$2 mm of plastic between the detector face and sample. Unlike the RASA samples that were aged for 24 h prior to counting, the HVAS samples were brought to the counting laboratory the same day they were collected. Therefore, the HVAS samples still had the radon decay products present, yielding a higher total activity.

Two different HPGe detectors were used to count the RASA and HVAS samples. The detectors' efficiencies ranged from 40 to 130 % (relative to a 3-in.^{2}. NaI at 25 cm distance). The detectors were housed in lead caves (4-in.-thick lead wall with a copper liner) for counting. The samples each were counted for a half day on two consecutive days. The acquired data were collected using Canberra LYNX® multichannel analyzers networked to a data acquisition PC. The spectral data collected via the Canberra Lynx analyzers were analyzed using Canberra's GENIE 2000 analysis software and to provide an MDA based on the Currie method [3]. Figure 2 shows the results of the counting for the HVAS and RASA.

Results

Absolute activity concentrations

Samples from the HVAS and RASA systems were collected and analyzed for any detectable gamma-ray emitting radionuclides. Of the collected nuclides, Be-7, I-131, I-132, Te-132, Cs-137 and Cs-134, exhibited the highest activity concentration. The correlated radionuclide activity concentration levels are shown in Figs. 3 and 4, with a dashed line indicating the position of an exact 1:1 ratio between the systems. Given the 10 % assigned uncertainty in the collected air volume, the observed results in the activity concentrations generally agree between the two sampling units. Activity concentrations for the lower activity isotopes ($<$0.5 mBq/m^3) are shown in Fig. 3b. Once again, the measurements between the two sampling systems are within the 10 % error associated with the collected volume uncertainty.

The correlation of activity concentrations between the two sampling devices is relatively good for the I-131, I-132, and Te-132 samples. However, the Cs-137 shows a

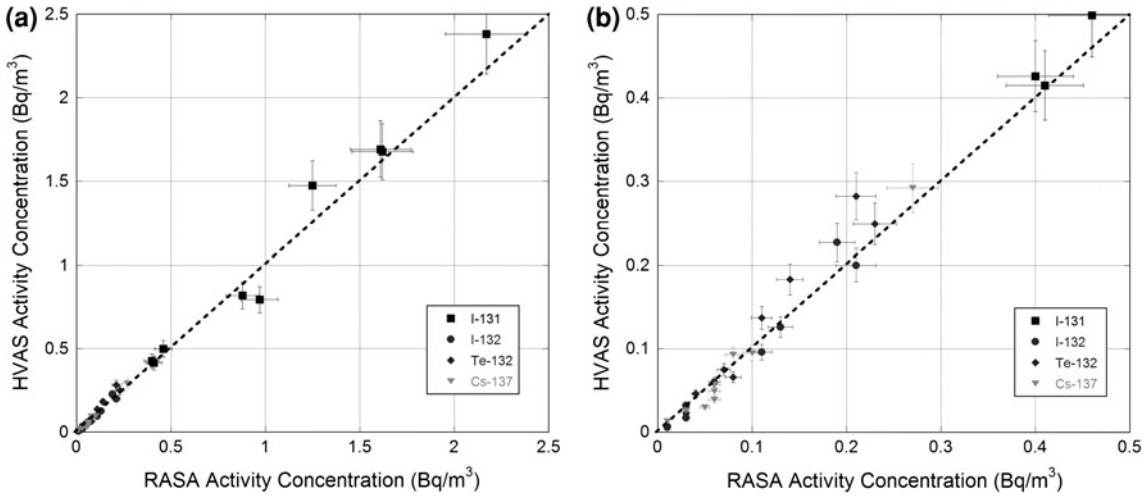

Fig. 3 **a** Radionuclides collected and analyzed using the RASA and HVAS systems. The *solid line* indicates the 1:1 ratio between the two systems; **b** expanded scale of **a** for lower concentration regions

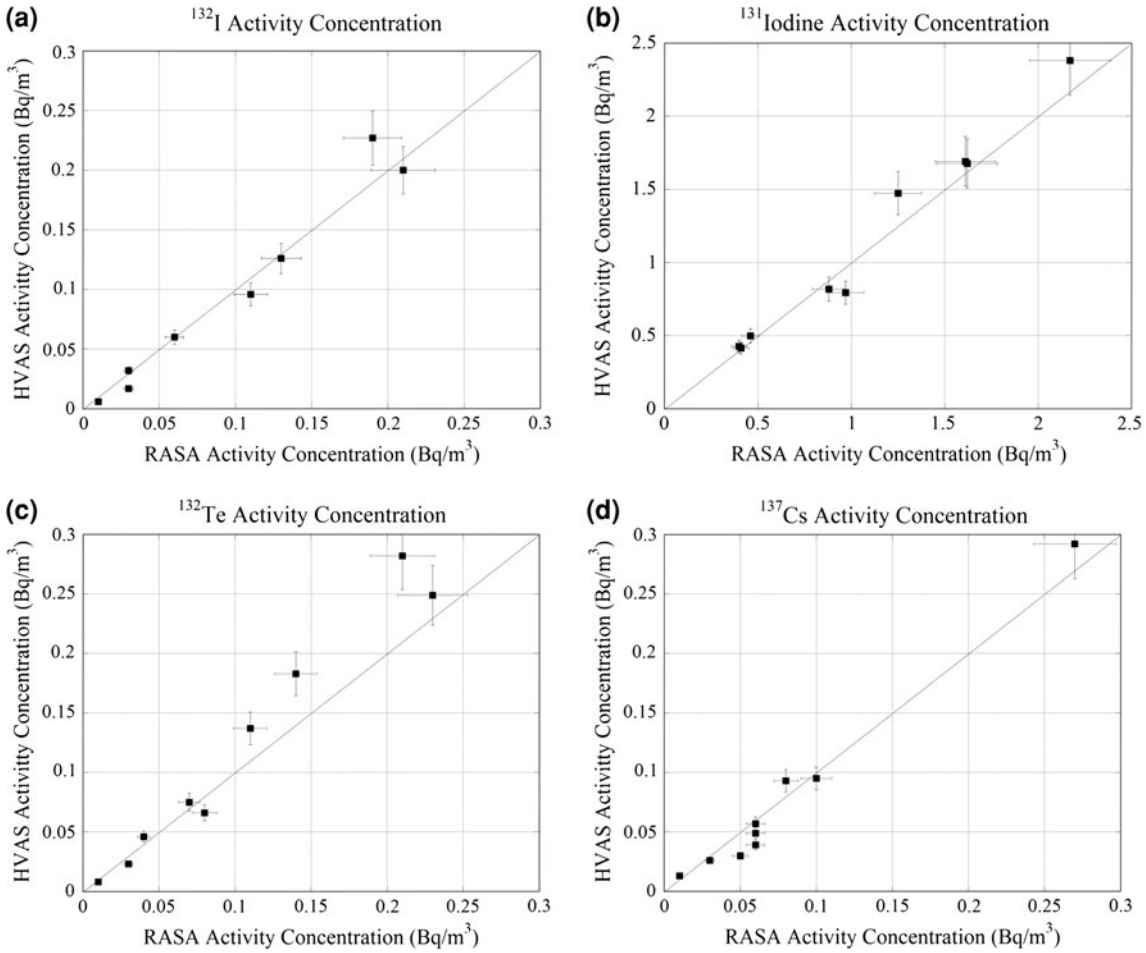

Fig. 4 Individual radionuclide activity concentrations for **a** I-132, **b**I-131, **c** Te-132 and **d** Cs-177. The *error bars* indicate the uncertainty associated with the detector counting

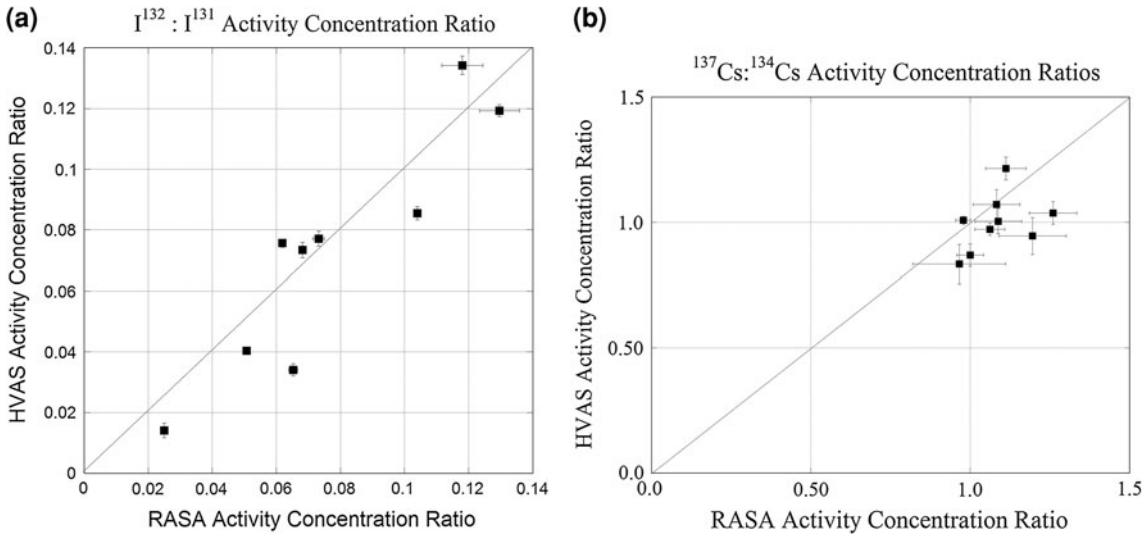

Fig. 5 Correlation of the radioisotope ratio **a** I-132 to I-131 and **b** Cs-137 to Cs-134 for both HVAS and RASA samplers

high degree of deviation in the lower activity concentrations. This deviation is addressed in the Discussion section of this submission.

Concentration ratios

Comparison of the isotopic ratios is crucial to determine the type of event that resulted in radionuclide releases. As shown in Fig. 5, the ratio of I-132/I-131 displays significant variations. While these values are plotted with 1σ uncertainties, the ratios fall outside of the calculated uncertainty It should be noted, as modeled by Biegalski et al. [6], these isotopic ratios clearly indicate a civilian power reactor as the source of the release.

Discussion

In general, the activity concentrations between the HVAS and RASA systems agree within a 10–20 % error, indicating that reasonably good correlation can be achieved by parallel system deployment. However, given that one possible motivation for parallel measurements is the acquisition of a "copy" of collected radionuclides, one must examine the variations between the systems for answers related to the systematic difference in measured radionuclide activity. One particularly interesting source of deviation can be illustrated in the Cs-137 data. Below 0.5 mBq/m^3, there is a significant shift towards higher concentrations in the RASA collection.

The most interesting observations were the ratios of iodine and cesium. The relatively large uncertainties in air volumes in this study are canceled out in these ratios. The ratio variation between samplers then calls into question whether the air parcel sampled $\sim$1.6 km away is truly the same or if some other unforeseen variation could be at play.

Conclusions

Two independent samplers were operated at PNNL during the collection of samples from the Fukushima reactor releases. This presented a unique opportunity to compare the results of near side-by-side measurements during an event in which significant radionuclide activity concentrations were released. In general, the agreement of the radionuclide concentration was between 10 and 20 % for concentrations and ratios of the radionuclides I-131, I-132, Te-132, Cs-137 and Cs-134. However, relatively large variations in the isotopic ratios indicate that the samplers, located 1.6 km apart, may not have measured identical air packets. Multiple possible explanations exist for the variations in the results. They include localized variations in surface atmospheric conditions, unanticipated variations in the collection efficiency of the sampling unit geometries, uncertainty in the actual collected air volumes, and collection variations for the different filter geometries. In order to further understand the utilization of spectator samplers for on-site inspection purposes, careful near receptor modeling should be applied to clarify the proper placement of spectator samplers. Until this issue is well understood, spectator samplers should be co-located in close proximity. This close proximity co-location will minimize the potential for the effects of variations in surface atmospheric conditions, potential obstruction due to near-by surface features (i.e., buildings) and maximize the possibility for measuring identical air packets.

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