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Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

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Robert Jubin[1], Arden Dougan[2], Paula Cable-Dunlap[1], Brian Ticknor[1], Joshua Hewitt[1], Benjamin Manard[1], Wendy Kuhne[3], Kelly McHugh[4], Megan Nims[4], Sean Stave[4], Nathan Stevens[5]

[1]Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA
[2]US Department of Energy, National Nuclear Security Administration, Washington, DC, USA (retired)
[3]Savannah River National Laboratory, Aiken, South Carolina, USA
[4]Pacific Northwest National Laboratory, Richland, Washington, USA
[5]Idaho National Laboratory, Idaho Falls, Idaho, USA

ABTRACT

The National Nuclear Security Administration's (NNSA) Office of Nonproliferation and Arms Control (NA-24) is evaluating Wide-Area Environmental Sampling (WAES) as an additional safeguards verification tool for the International Atomic Energy Agency to detect undeclared nuclear activities. The NNSA is evaluating strategies for conducting a generic WAES campaign, the cost of a WAES campaign, and the effect of technological advancements that have occurred since the last major WAES review in 1999. Until now, the NNSA effort has focused on tabletop exercises (TTXs) in which high-performance computing allows for advanced modeling and simulation efforts to be applied to the WAES question. Although the modeling and simulations used in the TTXs are extremely valuable, field campaigns are still needed to validate the assumptions that underpin the models and the modeling process itself.

During a 7 week period beginning in May 2023 and ending in June 2023, which included 4 weeks of active field collections, a multilaboratory team conducted its first in-field validation exercise. Prior to the in-field exercise, abbreviated TTXs were conducted to estimate the performance of all collection systems to be used during the field test. These TTXs guided the selection of materials to be released and the placement of the collection system. Based on these determinations, materials were procured to use in the field test, and an injection/release system was designed, built, and installed at the test facility. Background samples were collected during weeks one and four, and environmental collections against active releases were conducted during weeks two and three. The goals of this validation exercise included a demonstration of (1) the ability to provide controlled releases of particulates of surrogate materials, (2) the fielding and operation of collection systems (including deposition and active air collectors), and (3) the flexibility to revise equipment and campaign plans in the field. This paper presents the results and preliminary conclusions for this initial validation test. Based on these results, subsequent field campaigns are anticipated and will include the addition of other released materials.

INTRODUCTION

Article 9 of the Model Additional Protocol—INFCIRC/540 (Corrected) provides for the use of Wide-Area Environmental Sampling (WAES) by the International Atomic Energy Agency (IAEA) to assist in drawing conclusions about the absence of undeclared nuclear activities but only after its use and associated procedural arrangements have been approved by the IAEA Board of Governors. A demonstration of both technical feasibility and cost effectiveness is

anticipated to be needed before such approval. The last major review of WAES, completed in 1999, concluded that WAES was too costly for implementation at that time. However, advancements over the past two decades in laboratory analytical capabilities, meteorological modeling, collector technology, and high-performance computing support a contemporary assessment to reevaluate the practicality of implementing WAES as an IAEA verification tool. The overall scope of the WAES project is described in a companion paper [1] at this meeting and will not be repeated here.

The WAES project team elected to use tabletop exercises (TTXs) that utilize extensive modeling and simulation to understand the technical viability of using WAES to obtain the data required to verify the absence of undeclared nuclear activities. Using sophisticated computer models, the TTXs provide a means to rapidly explore a wide range of parameters to evaluate their effect on the ability to observe a possible undeclared nuclear facility. The results from these TTXs have been encouraging, but to support the conclusions drawn from the simulations, this project must validate the results generated in the TTXs. The Lost River Modeling and Sampling Validation Exercise performed in 2023 was the first exercise conducted that would allow a comparison of the results from the models with actual samples collected in the field by a suite of prototype and commercial off-the-shelf collectors.

The purpose of the Lost River Modeling and Sampling Validation Exercise was to demonstrate (1) the controlled release of particulates from the test facility and (2) particulate sample collections in and around that facility and at various distances from the facility. The results from the analysis of these samples also provides background characterization data that will serve as ground truth for future TTXs and enables correlation between modeling activities and field collections. Finally, this exercise provided an initial demonstration of the WAES prototype collector for use in the field. This demonstration includes operational ruggedness and the verification of its ability to collect WAES-relevant samples.

Lost River Modeling and Sampling Validation Exercise Planning

The release scenario planned for the Lost River Modeling and Sampling Validation Exercise was based on a semicontinuous release of particulates containing nonradioactive $Ru(NO_3)_2$ and $Fe(NO_3)_3$ from the stack of the Moran facility located on the Idaho National Laboratory (INL) site. Based on the climatology for this location, the particles were projected to be dispersed primarily to the southwest and to the northeast of the Moran facility. Six collection locations, designated in Figure 1, were available to the WAES team. Two locations were to the southwest of the Moran facility. Magic Kingdom (MK) was the furthest to the southwest at 4.6 km, and Off-Road (OR) at 1.3 km was relatively close to the facility. The four sites to the northeast were in increasing distance: Fenceline (FL) at 0.6 km, Optimus Prime-T3 (T3) at 6.2 km, K-Boom (KB) at 21.7 km, and Mud Lake (ML) at 36.5 km.

The particulate release concentrations for the exercise were determined through a series of three mini-TTXs conducted 6 to 8 months prior to the exercise. The mini-TTXs assumed three 2-day collections accruing during each of two collection weeks. The mini-TTXs explored a range of particle release quantities and particle size distributions for the suite of collectors that were considered for deployment in this exercise. The TTXs also considered a range of potential background conditions and multiyear climatology. A summary of the results is shown in Table 1 and Table 2. As shown in Table 1, low-flow collectors (i.e., less than 250 L/min) should only be used at relatively short distances from the facility. The high-flow prototype WAES collectors

should be viable at distances across the INL site. Table 1 shows the predicted results from the simulated analysis of the material accumulated on collectors located at each of the six collection locations. Based on multiyear climatology, these results indicate that for a 100 L/min collector, all samples collected at the FL location should provide positive indications for ruthenium released from the facility. At the OR and MK locations, the probability of detection decreased with increasing distance, but at OR, a better than 60% probability was found for all six collections being above the set detection limit. However, at greater distances, such as those for T3, KB, and ML, the probability was significantly reduced. As shown in Table 2 , the high-flow collectors significantly increased the probability of positive observations with probabilities approaching 100% out to the T3 location. KB and ML showed somewhat lower probabilities but were still in general above 50% probability.

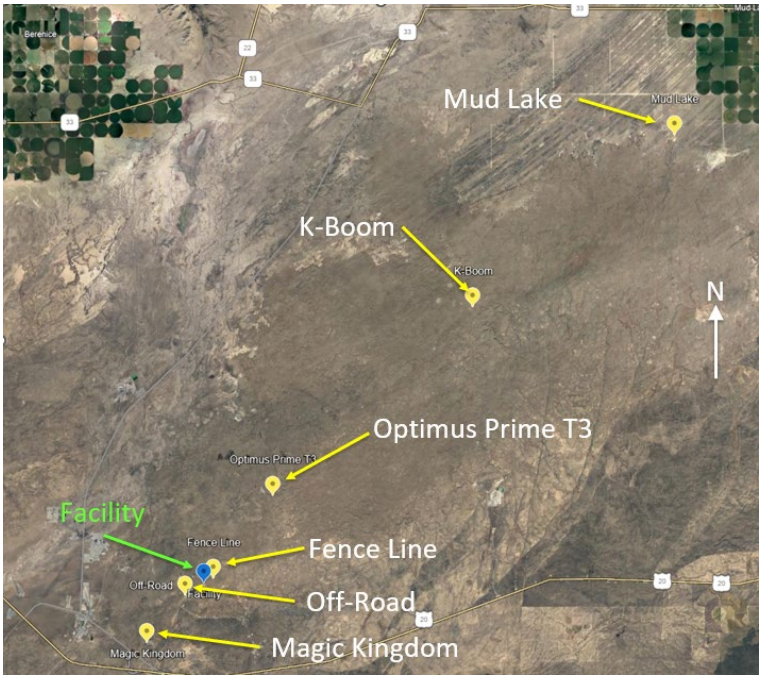

Figure 1. Collector locations relative to the Moran facility for the WAES Modeling and Sampling Validation Exercise.

Table 1. Modeled percentage of positive indications for ruthenium at 100 L/min at initial planned levels

Percentage of Positive Indications (> 4 Sigma)						
Particle Description	Location					
	Fence	OR	MK	T3	KB	Mud
bin1	96%	69%	64%	25%	0%	0%
bin2	96%	68%	62%	21%	0%	0%
bin3	95%	66%	56%	16%	0%	0%
bin4	95%	62%	50%	11%	0%	0%
bin5	96%	68%	63%	22%	0%	0%
bin6	95%	66%	57%	16%	0%	0%
bin7	95%	66%	57%	16%	0%	0%
bin8	95%	67%	57%	15%	0%	0%
bin9	95%	67%	58%	14%	0%	0%
bin10	98%	80%	73%	39%	0%	0%

Table 2. Modeled percentage of positive indications for ruthenium at 15,000 L/min at initial planned levels

Percentage of Positive Indications (> 4 Sigma)						
Particle Description	Location					
	Fence	OR	MK	T3	KB	Mud
bin1	99%	96%	93%	96%	82%	68%
bin2	99%	96%	93%	96%	79%	66%
bin3	99%	96%	92%	95%	78%	63%
bin4	98%	96%	92%	95%	75%	57%
bin5	99%	96%	93%	96%	80%	66%
bin6	98%	96%	92%	95%	77%	62%
bin7	98%	96%	92%	95%	77%	60%
bin8	98%	96%	92%	95%	75%	57%
bin9	98%	96%	92%	95%	69%	45%
bin10	99%	98%	93%	96%	76%	55%

Lost River Modeling and Sampling Validation Exercise Execution

The Lost River Modeling and Sampling Validation Exercise was conducted over a 7-week period beginning May 15, 2023, and ending on June 30, 2023. The first 2 weeks were dedicated to the installation of the TSI Inc. 8108 Large Particle Generator in the Moran facility at INL as well as the testing of the newly installed unit and supporting equipment. During this same 2-week period, collectors were placed in the field at six locations across the INL site, which are identified in Figure 1. Week three of the exercise was dedicated to background collections using all the collectors that had been placed in the field the previous week. Operational releases were conducted during weeks four and five. During the active release period, particles generated by the TSI 8108 were injected directly into the Moran facility stack via a manifold. The feedstock for the particle generator was an aqueous mixture of two metal nitrates. Week six was used for postrelease background collection. The nominal collection duration was a 48-hour period, which allowed three collections during each of the two 6-day background collections and the two 6-day release periods. The seventh week was used to pack up the collectors and return the facility to standby conditions.

Table 3 provides information on the six types of collectors placed in the field. The deposition collector, magnetic collector, and first article electrostatic collector were all prototype units. Figure 2 shows the Oak Ridge National Laboratory (ORNL) deposition collector, which consisted of nine Teflon sheets that provided a total collection area of approximately 1 m^2. The SKC Inc. collectors were very low-volume collectors and were placed on the stack and on the injection manifold of the facility to provide data on the source term at the stack and data on losses in the stack and blower. The Next Generation Impactor (NGI) collectors were also low-volume collectors with a collection volume of 100 L/min. These collectors were placed at near-field locations FL, OR, and MK. The Aerosol Contaminant Extractor (ACE) electrostatic collector had a flow rate of nominally 150 L/min for each collection plate and was located at the same locations as the NGI. A single magnetic collector was initially placed at MK and was later relocated near the Moran facility. The Savannah River National Laboratory (SRNL high-volume first article electrostatic collectors (Figure 3) had a collection capacity of 15,000 L/min for each collection chamber and were located at FL, MK, T3, KB, and ML. These locations and the collectors at each location are shown in Table 3 and Figure 1. Surface soil samples were also collected at the Moran facility, FL, OR, MK, and T3.

Over 30 individuals participated in this modeling and sampling validation exercise, including personnel from INL, Lawrence Livermore National Laboratory, ORNL, Pacific Northwest National Laboratory, SRNL, and the Y-12 National Security Complex. During the operational release period, personnel from IAEA as well as senior laboratory and NNSA management personnel visited the facility.

The analysis of the particles collected on the various samplers are being conducted at Pacific Northwest National Laboratory, ORNL, SRNL, and Lawrence Livermore National Laboratory using mass spectrometry and other techniques. The discussions presented below represent the findings from the initial sets of analytical results.

Figure 2. ORNL deposition collector.

Figure 3. SRNL high-volume WAES prototype electrostatic collector.

DISCUSSION

Active Operations

The initial week of active releases was significantly affected because of poor weather conditions and the limited supply of $Ru(NO_3)_2$. Rain occurred virtually every day, which resulted in the need to either cover the deposition collectors or terminate collections on some of the active collectors. Notably, despite the adverse weather conditions experienced during the collection periods, all collectors performed as expected. Some arcing was experiences in both the ACE and high-volume first article electrostatic collectors and the deposition collectors were covered to limit rain washing away the samples from the Teflon sheets.

Based on the forecasted weather for the week, the active releases were also limited to three 8-hour release windows and one 24-hour release rather than the planned three 48-hour release periods modeled in the planning TTXs. The released levels during this first week were set to 5.5 times the TTX-modeled release levels (i.e., ~0.05 g/h ruthenium) represented in Table 1 and Table 2. The higher releases should improve the probability of observing the emission from the facility. However, the 8108 Large Particle Generator experienced large accumulation of the $Ru(NO_3)_2$ materials on the walls of the generator. As a result, the release levels were believed to be degraded by ~90% from the target levels.

Based on the observations of material accumulation in the 8108 Large Particle Generator an attempt was made to reduce the accumulation of material in the generator and manifold with several expedient modifications between the first and second weeks of active releases.

The second week of active operations was also affected by poor weather conditions, albeit slightly improved. An additional supply of the $Ru(NO_3)_2$ was obtained, and researchers were able to increase the release levels to approximately 13 times the targeted levels modeled in the

TTX. The facility successfully completed three 48-hour release periods during the second week. Although improved, the Large Particle Generator continued to experience accumulation of the metal nitrates on the generator walls, and as a result, significant degradation of the release amounts was anticipated.

Table 3. Collectors placed in the field during the Lost River Modeling and Sampling Validation Exercise

Collector name	Collector type	Air rate (L/min for each cell)	Collectors available	Locations	Laboratory
Deposition collector	Passive	NA	10	FL, OR, MK	ORNL
Sioutas five-stage cascade impactor (SKC Inc.)	Low-volume; Active	9	3	Stack, Manifold, OR, FL	ORNL
Next Generation Impactor (NGI), seven-stage cascade impactor (TSI Inc.)	Low-volume; Active	100	3	FL, OR, MK,	ORNL
Aerosol Contaminant Extractor (ACE)	Low-volume; Active	150	5	FL, OR, MK,	ORNL
Magnetic collector	Medium-volume; Active	1,200 (120 through course fraction/1,080 through fine fraction)	1	MK	ORNL
First article collector—electrostatic	High-volume; Active	15,000	5	FL, MK, T3, KB, ML	SRNL

Sample Analysis of Field Collection Samples

SKC Samples

The SKC collectors were used to collect samples from the stack discharge during the second week of active releases. To date, only one set of the stack SKC samples has been analyzed. This sample collection covered the release period from 16:26 to 20:30 on June 14, 2023. The results show that the ruthenium was released to the environment at a rate of 0.066 g/h and had iron-to-ruthenium ratios consistent with the feed solution to the TSI 8108 Large Particle Generator. Based on the feed solution to the 8108 Large Particle Generator, this result would indicate that at this point in the exercise, approximately 91% of the ruthenium feed material was lost in the generator, the manifold leading to the stack, or in the stack itself. This result was not unexpected based on the observations of the continued particulate accumulation within the generator. Scanning electron microscopy analysis of the SKC plates show a range of particle sizes consistent with the method of generation. In the case of the larger particles, some indication was present that these particles were not fully dried or were “sticky,” which is also consistent with the generation of metal nitrate particles (see Figure 4).

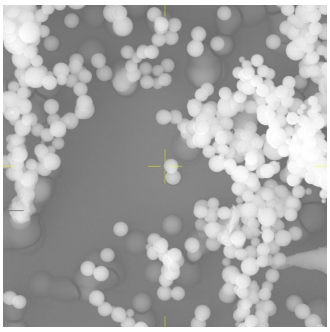

Figure 4. Scanning electron microscopy image of particles on SKC disk 2.

Deposition Samples

Analysis of the Teflon sheet samples from selected deposition collectors located at FL, OR, and MK collected during the week prior to active release and the week following active release has been completed. Deposited material was removed from Teflon sheets used in the collector and analyzed via inductively coupled plasma (ICP) mass spectrometry (MS). These results (see Table 4) showed the ruthenium deposition rate to be <0.01 ng/h. (Note that results that appear anomalous are highlighted in yellow.) The analysis of the deposition samples collected at FL and OR collected during the second week of active release showed a significant elevation in the deposition rate of ruthenium on the collector (0.1-0.3 ng/hr) which is ~10–100 times those observed during the background collects. Soil samples collected at the Moran facility during the second active week of releasee and a 2-week cumulative sample show elevated Ru levels during the second active week and a 2-week cumulative sample. All other soils were either <LOD or at background levels.

Table 4. Initial analytical results from the deposition collections during the Lost River Modeling and Sampling Validation Exercise

Location	Sample ID	Type	Start time	End time	Ruthenium deposition rate (ng/h)	Notes
FL	DBFL-4	active	6/12/2023 15:52	6/16/2023 16:29	0.288195	—
FL	DBFL-5	active	6/16/2023 16:56	6/18/2023 14:04	0.10274	—
FL	DOFL-1	background	5/29/2023 16:47	5/30/2023 20:11	0.29135	Does not compare well with NGI background for same period
FL	DOFL-4	active	6/12/2023 15:43	6/17/2023 14:32	0.180451	—
FL	DOFL-6	background	6/21/2023 17:12	6/23/2023 13:25	0.006265	—
FL	DRFL-5	active	6/12/2023 15:45	6/17/2023 22:43	1.407866	Appears high— Overlaps collection period with DBFL-4 and DBFl-5 above.
OR	DBOR-1	background	5/29/2023 15:05	5/30/2023 16:05	0.01044	—
OR	DBOR-6	background	6/21/2023 15:42	6/23/2023 12:46	<LOD	—
MK	DRMK-1	background	5/29/2023 21:55	6/1/2023 20:00	<LOD	—
MK	DRMK-3	active	6/12/2023 14:19	6/17/2023 19:22	0.014851	Appears low but within a factor of 2-4 based on atmospheric dilution.
MK	DRMK-4	background	6/19/2023 14:28	6/21/2023 15:06	0.009417	—
MK	DSMK-1	background	5/29/2023 21:55	6/1/2023 20:20	0.003132	—

Notes:

Samples collected before or active releases to determine background are shown in red

NGI Samples

Laser ablation (LA) ICP-MS imaging, an analytical capability newly established at ORNL and under active development, was applied to the NGI samples to rapidly (in just hours versus weeks to months) screen the collected samples for ruthenium, iron, and uranium. Results from screens of NGI plate D from FL, OR, and MK locations collected between 15:39 on June 12, 2023, and 16:27 on June 16, 2023, which was the first part of the second week of active releases, are shown in Figure 5 through Figure 7, respectively. At FL, approximately 40 particles contained

ruthenium; in a single pocket at OR, approximately 13 contained ruthenium, and MK contained approximately 4. This result implies a decrease in the airborne concentration of approximately one order of magnitude across these three locations, albeit with some uncertainty in counting statistics.

Currently, LA-ICP-MS cannot be used for quantification, so the samples were sent for bulk ICP-MS analysis of the entire NGI sample sets of eight plates. Additional NGI collections from the active release period and background collection were also sent for bulk ICP-MS analysis. The results for the FL location are shown in Figure 8, and the results for the three locations are summarized in Table 5 along with a comparison with a preliminary prediction of the anticipated ruthenium amount that could have been collected based on a meteorological reconstruction of the transport from the stack to each of the collectors.

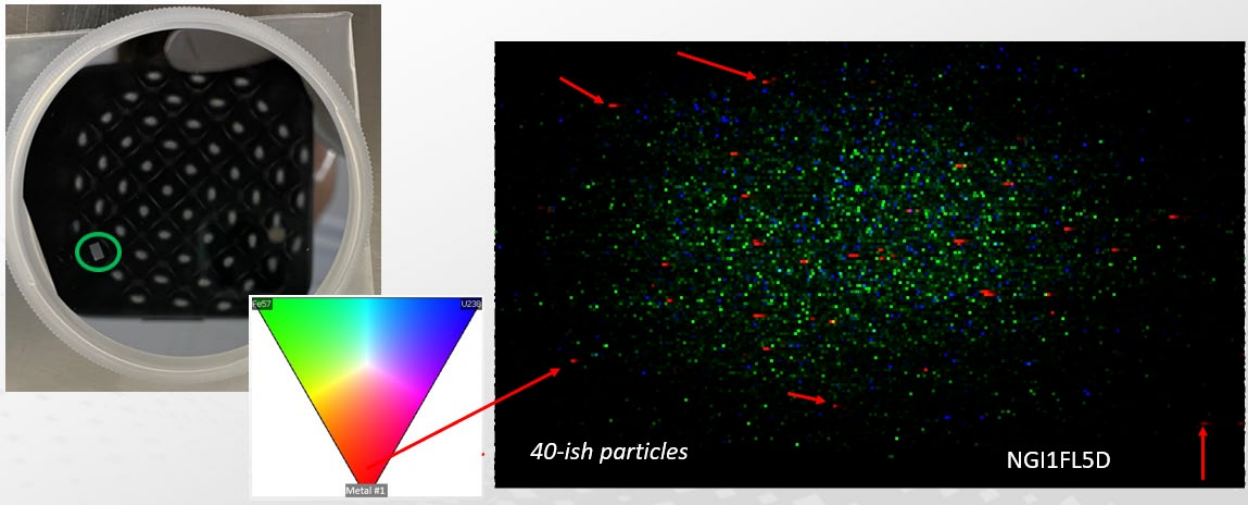

Figure 5. LA-ICP-MS analysis of a single pocket from FL NGI collector plate D. Red dots are particles containing ^{101}Ru. Green are ^{57}Fe-containing particles, and blue dots note particles that contain ^{238}U.

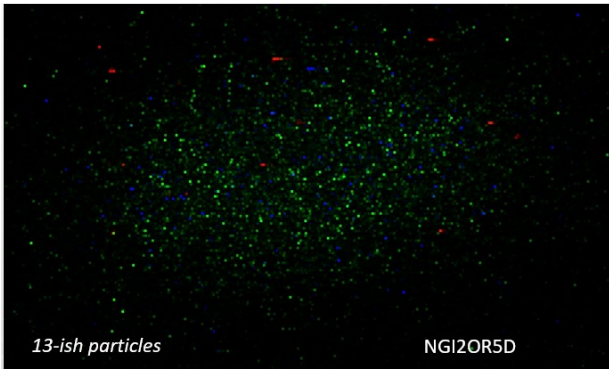

Figure 6. LA-ICP-MS analysis of a single pocket from OR NGI collector plate D. Red dots are particles containing ^{101}Ru. Green indicate ^{57}Fe-containing particles, and blue particles contain ^{238}U.

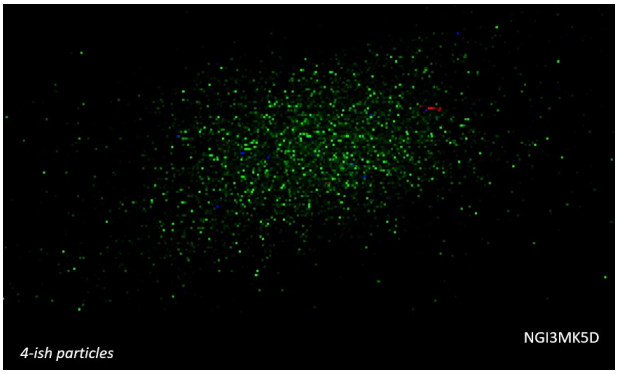

Figure 7. LA-ICP-MS analysis of a single pocket from MK NGI collector plate D. Red dots are particles containing ^{101}Ru. Green indicate ^{57}Fe-containing particles, and blue particles contain ^{238}U.

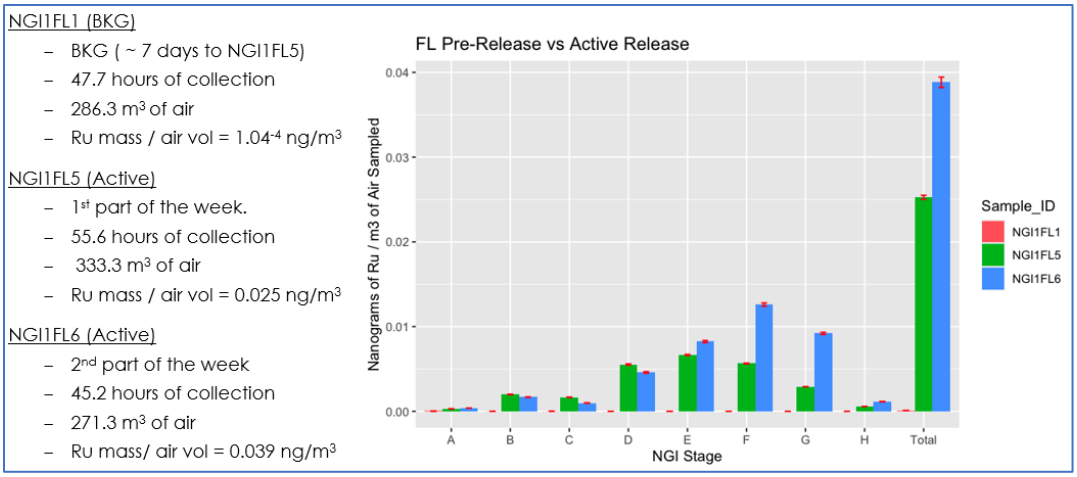

Figure 8. ICP-MS analysis of two FL NGI collection during the second week of active releases.

High-Volume Electrostatic Samples

The samples from the SRNL high-volume collectors are still in the process of being analyzed. However, preliminary results suggest that ruthenium was detected at both T3 (~6.2 km) and KB (~22 km) (see Table 6), which is consistent with the estimates shown in Table 2. FL data for NGI and high flow agree well at 0.038 ng/m^3 and 0.018 ng/m^3, respectively.

Table 5. Summary of near-field NGI collector results for second week of active collection and comparison with predicted loadings

Location	Sample ID	Measured Ru mass (ng)	Predicted Ru mass (ng) from met modeling (No hold up / 9% release)	Collection time (h)	Ru average concentration over collection period (ng/m^3)	Type
FL	NGI1FL1	0.0298	—	47.7	—	Background
FL	NGI1FL5	8.42	51 / 4.6	55.6	0.025	Active
FL	NGI1FL6	10.5	60 / 5.4	45.2	0.039	Active
OR	NGI2OR5	2.49	17 / 1.6	56.7	0.0073	Active
OR	NGI2OR6*	0.43	13 / 1.2	28.5	0.0025	Active
MK	NGI3MK5	8.62	16 /1.44	58.6	0.024	Active
MK	NGI3MK6*	0.48	5.3/ 0.48	28.5	0.0027	Active

Notes:

Samples collected before or active releases to determine background are shown in red

Table 6. Initial analytical results from the high-flow collector deployed during the Lost River Modeling and Sampling Validation Exercise

Location	Type (Date)	Collection time (h)	Ru (ng/m^3)	Average flow rate across one cell (L/min)
FL	First background (5/31–6/2)	47.33	<LOD	15,527
FL	Second release (6/16–6/19)	71.32	1.84×10^{-2}	15,232
FL	Second background (6/19–6/21)	48.00	1.72×10^{-5}	15,270
T3	First background (5/31–6/5)	116.40	<LOD	13,710
T3	Second release (6/16–6/19)	71.67	3.57×10^{-4}	14,866
T3	Second release (6/14–6/16)	47.08	4.41×10^{-5}	14,866
KB	Second release (6/16–6/19)	71.75	1.31×10^{-4}	14,945

Notes:

LOD: limit of detection

Samples collected before or active releases to determine background are shown in red.

CONCLUSION

Based on the results to date, the Lost River Modeling and Sampling Validation Exercise was a successful initial demonstration of WAES techniques. Analysis of samples collected during the initial background collection periods indicated the absence of ruthenium via both ICP-MS and LA-ICP-MS techniques. The samples from multiple collectors taken during the pre- and postactive release periods consistently show ruthenium at or below the levels of detection. Deposition samples showed positive indications at both FL and OR for ruthenium. Particles containing ruthenium were identified in samples from FL for both the NGI and high-volume collectors and have been confirmed by multiple analytical techniques. These releases are attributed to the facility operations. Particles containing ruthenium were also identified in OR and MK samples by LA-ICP-MS on the plates taken from the NGI samplers. These results were also confirmed by the bulk analysis of the sample. These indications are also attributed to the facility release. Preliminary data from the high-volume electrostatic collectors indicate that ruthenium was observed at least as far as KB with the ML sample still pending. For the most part, the results are generally consistent with the initial metrological reconstructions of what should have been observed. The NGI, the ACE electrostatic, and the high-volume electrostatic collectors, despite the very adverse weather conditions experienced during the exercise, all performed as anticipated, albeit with some arcing issues in the electrostatic units because of the rain.

Over the upcoming months, analysis of the remaining samples will be completed and a reconstruction will be conducted using the observed meteorological conditions to provide a direct comparison with the observations at the collection sites.

ACKNOWLEDGMENTS

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[1] Paula Cable-Dunlap, Arden Dougan, Robert Jubin, Diane Fischer, Brian Ticknor, Sean Stave, Kelly McHugh, Jill Cooley, “Initial Findings of a Contemporary Assessment of Wide-Area Environmental Sampling,” Proceedings of the INMM 65th Annual Meeting, July 2024.