PREX: AN EXPERIMENT TO INVESTIGATE DETECTION OF NEAR-FIELD PARTICULATE DEPOSITION FROM A SIMULATED UNDERGROUND NUCLEAR WEAPONS TEST VENT

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Abstract—A radioactive particulate release experiment to produce a near-field ground deposition representative of small-scale venting from an underground nuclear test was conducted to gather data in support of treaty capability development activities. For this experiment, a CO_2-driven "air cannon" was used to inject ^{140}La, a radioisotope of lanthanum with 1.7-d half-life and strong gamma-ray emissions, into the lowest levels of the atmosphere at ambient temperatures. Witness plates and air samplers were laid out in an irregular grid covering the area where the plume was anticipated to deposit based on climatological wind records. This experiment was performed at the Nevada National Security Site, where existing infrastructure, radiological procedures, and support personnel facilitated planning and execution of the work. A vehicle-mounted NaI(Tl) spectrometer and a polyvinyl toluene-based backpack instrument were used to survey the deposited plume. Hand-held instruments, including NaI(Tl) and lanthanum bromide scintillators and high purity germanium spectrometers, were used to take in situ measurements. Additionally, three soil sampling techniques were investigated and compared. The relative sensitivity and utility of sampling and survey methods are discussed in the context of on-site inspection.
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INTRODUCTION

THE DETECTION of underground nuclear explosions is typically performed using remote detection methods, such as those implemented by the Comprehensive Nuclear-Test-Ban Treaty Organization's (CTBTO) International Monitoring System, to detect seismic waves and airborne radioactivity vented from the subsurface (Bowyer et al. 1997). Techniques performed locally (i.e., within an area of several hundred square kilometers surrounding a nominal ground-zero point) are far more intrusive but also far more confirmative than remote techniques. Such on-site inspection (OSI) techniques are also allowed under the Comprehensive Nuclear-Test-Ban Treaty (CTBT), which has not yet entered into force (Schulze et al. 2000). An OSI can include many radionuclide techniques, including gamma radiation survey, gamma spectrometry, and environmental sampling and analysis (of solids, liquids, and gases). A good understanding of the possible contamination distribution, along with the most valuable equipment and search techniques for locating accessible radioactive contamination, would dramatically improve the likelihood of conducting a successful OSI. In support of such work, Pacific Northwest National Laboratory (PNNL) conducted research to understand a realistic deposition of radionuclides that might occur under venting conditions from an underground nuclear test.

In May 2013, PNNL, in collaboration with National Security Technologies LLC (NSTec), performed an atmospheric release of ~40 μm particulate lanthanum oxide (La_2O_3) after neutron activation in the Washington State University TRIGA research reactor produced 3.7×10^7 kBq (at release) of short-lived ^{140}La within the powder. The radioisotope ^{140}La was chosen as a representative fission fragment due to its relatively short half-life, prominent gamma-ray emissions, and ease of production through reactor-based neutron activation of stable lanthanum. The short-lived radioactive particulate material was successfully injected into the lowest levels of the atmosphere to simulate the release from a weakly vented underground nuclear test. Following the release, project personnel used a variety of radiation sampling and survey techniques to characterize the downwind deposition of the material. The Remote

Sensing Laboratory (RSL) supported this effort by performing a fixed-wing aerial survey shortly after the release. The techniques applied to this experiment are typical of those being planned for use in the event of an international on-site inspection under the Comprehensive Nuclear Test Ban Treaty.

During the planning stages of the PRex release, project personnel had the opportunity to participate in the Defence Research and Development Canada (DRDC) "Full-Scale Radiological Dispersal Device (FSRDD) Field Trials" (Green et al. 2016). Participation in the RDD Field Trials provided the PRex project team with significant insights into the scale of logistics required, as well as radiological protection and control issues. The successful production and use of ^{140}La, with low levels of radioactive contaminants, initially shifted the PRex focus from longer-lived isotopes (e.g., on the order of 30-d half-lives) to isotopes with half-lives on the order of a few days. This first shift was driven by recognition of the long-term storage of contaminated equipment that would be driven by isotopes with ~30-d half-lives. In the final stages leading up to the PRex release, commercial production of lanthanum-loaded aluminosilicate microspheres, as well as gold-coated aluminosilicate microspheres (targeting reactor neutron activation of ^{197}Au to ^{198}Au), was unsuccessful, and the project reverted to a backup plan based on the successful DRDC trials with lanthanum oxide powder.

An objective of the particle release experiment (PRex) was to measure the resulting downwind ground contamination using several sampling and survey techniques. One of the challenges in an OSI is to locate radioactive debris that has escaped an underground nuclear explosion and settled on the surface near and downwind of ground zero (GZ). Successful search and identification of such debris will enable confirmation that a nuclear test occurred. Positive results will also facilitate locating the testing site. To develop a robust OSI capability, it is necessary to understand the surface source term created by intermediate and long half-life ($t_{1/2}$ in the weeks to years range) particulate nuclear explosion debris. To facilitate study of the source term without the attendant long-term contamination associated with mixed fission products, this experiment used a shorter-lived radionuclide (^{140}La $t_{1/2}$ = 1.678 d), a strong gamma-emitting fission product.

Overview of PRex

PRex was carried out on Tuesday, 14 May 2013 with local preparation completed during the 8 d leading up to the experiment. NSTec personnel established considerable infrastructure preparation work in advance of initial PNNL staff arrivals on Monday, 6 May. At the time of execution, ~3.7 × 10^7 kBq of ^{140}La was released as La$_2$O$_3$. The powder was used to provide a precursor for reactor-based production of ^{140}La through neutron capture on the only stable lanthanum isotope, ^{139}La. The powder was sieved to select 32–45 μm particles prior to irradiation in the Washington State University (WSU) TRIGA reactor. This limited size range of particles was selected to ensure a large fraction of the source activity released would deposit within the first several kilometers, which was desirable for the logistics of source production as well as for health and safety reasons (Rishel et al. 2016). The irradiation was conducted on Friday, 10 May 2013; the ^{140}La source was shipped from WSU two days later and arrived at the experiment site on Monday morning, the day before the experiment. Witness plates and air samplers were deployed in an array to the north of the release point in advance of the release. The radioactive particles were lofted ~10 m in the air using an "air cannon" with the release occurring over an approximately 2-min time span. The site of the release was at latitude 36.99668° and longitude −116.00679° at the northern edge of Area 6 in the Nevada National Security Site (Fig. 1).

After the release, a number of research activities were conducted, including aerial survey, vehicle-based survey, in situ measurements, handheld and backpack survey, air sampling, soil sampling, and field laboratory measurements at a "Base of Operations." Witness plate collections were used to establish a baseline debris deposition activity estimate.

MATERIALS AND METHODS

Site selection

A number of possible sites at the NNSS were visited in January 2012 to locate an ideal site for the PRex. The desired location attributes were as follows:

1. Low anthropogenic gamma-ray background;
2. Sufficient distance from site infrastructure and site borders so that projected transport and deposition would not present any health and safety issues;
3. A nearby downwind area exhibits gamma-ray background contributions stemming from historic nuclear tests. This would provide a dataset useful in testing algorithms and procedures intended to discriminate recent tests from old tests under a CTBT OSI;
4. Downwind access for vehicle radiation surveys; and
5. Nearby infrastructure availability (e.g., lodging, electricity, etc.).

There were no suitable locations with access to electricity, forcing the use of a generator during the experiment. In the end, criteria 4 and 5 trumped criterion 3, and a site in the northern end of Area 6 was chosen at latitude 36.99668° and longitude −116.00679°. The site is at the southern end of the Yucca Flats area, immediately to the south of where many underground nuclear tests occurred in the past but
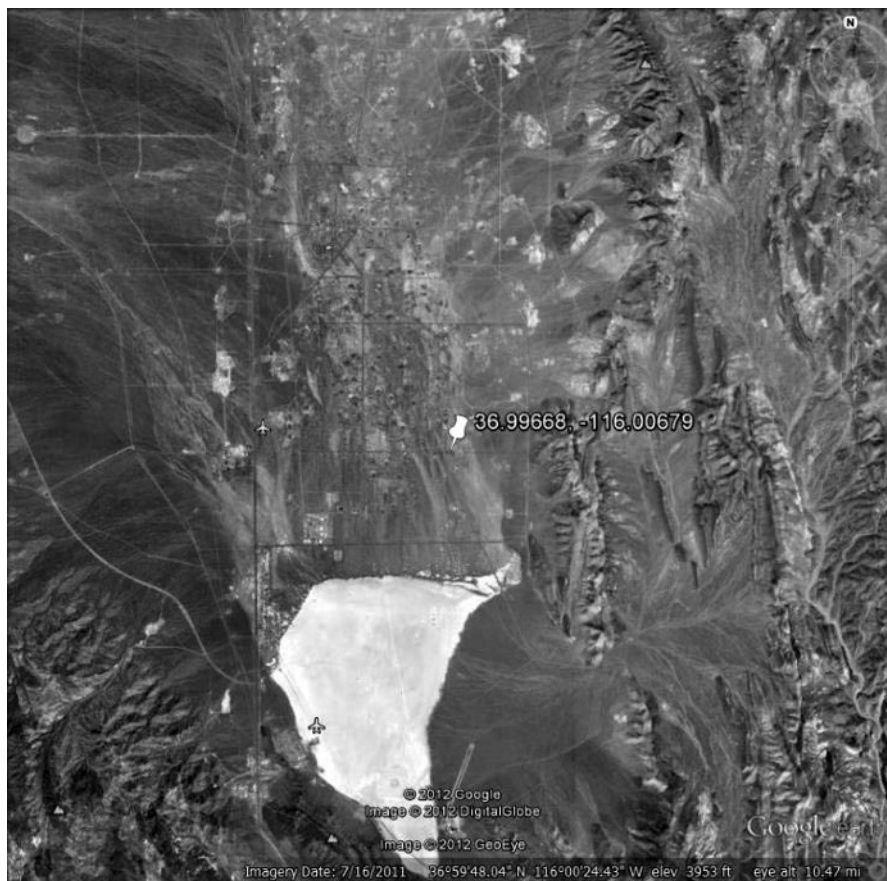
Fig. 1. PRex location on the Nevada National Security Site.

on the eastern edge away from the main road (Mercury Highway). The location is very flat with light desert grass and small shrubs, providing for straightforward off-road and aerial surveys (Fig. 2). Although there are radiologically controlled areas downwind (north) of the release site, particularly those associated with the tests Adze and Seaweed, these do not have surface radiation from old tests but are fenced to prevent the disturbance of buried contamination.

A vehicle-based background survey of the site was performed in advance of the experiment using a Radiation Solutions Inc. (RSI) RS 700 spectrometer system with two 5.1 cm × 10.2 cm × 40.6 cm NaI(Tl) detectors mounted in the bed of a Kawasaki (Kawasaki Motors Corp., USA, Santa Ana, CA, USA) Mule All-Terrain Vehicle (ATV). The RS 700 has an integrated global positioning system (GPS) that recorded coordinates along the path where the ATV was driven. An additional guidance system was tested in conjunction with the built-in GPS. A Raven RGL 600 system, (Raven Industries, Inc., Sioux Falls, SD, USA) designed to guide the precision driving of agricultural equipment, and an ancillary GPS unit were combined to enable surveying parallel lines of arbitrary separation. The ATV was driven at a nominal 1.4 m s^{-1}, with line spacing of

50 m. Successful completion of this vehicle background survey demonstrated that the several-square-kilometer site was suitable for vehicle survey using the Kawasaki ATV.

Fig. 2. NNSS release site. While not visible in the photo, several craters and un-collapsed test locations exist in close proximity to the north of the release point. Craters are more concentrated to the northwest; south-southwesterly winds would result in the fewest obstacles for ground survey.

Radioactive particulate source production

Lanthanum oxide powder, La$_2$O$_3$, was purchased from Materion (Materion Corporate Headquarters, Mayfield Heights, OH, USA) at 99.99% purity and −200 mesh (e.g., containing particles with diameter less than 74 μm). The material was separated by particle size using a Retsch (Retsch GmbH, Haan, Germany) AS200 sieve shaker. After sieving, the particle size of the material selected for irradiation and release was in the 32–45 μm range.

The lanthanum oxide powder was contained in a sealed plastic container inside of a roll-sealed aluminum cannister during irradiation in the WSU TRIGA research reactor and remained in this canister during transport to NNSS. A test irradiation producing the full planned ^{140}La activity was conducted in advance to confirm that long-lived contaminant isotopes were below acceptable levels for release. The 36-min irradiation for the release was performed on Friday, 10 May, with a target ^{140}La production of 8.1×10^7 kBq at noon Pacific daylight time (PDT) on Sunday, May 12th. The irradiation was performed with the reactor operating at a power level of 1 MW thermal, with a neutron flux previously characterized as 5.3×10^{12} n cm^{-2} s^{-1}. Gamma assay performed after the irradiation confirmed production of the desired 8.1×10^7 kBq of ^{140}La activity.

Site preparation

Witness plate sampling array. In the days leading up to the experiment, sampling equipment was placed at predetermined locations and prepared for collection of the PRex plume. The sampling network consisted of both air samplers and witness plates for the collection of debris. After the release and sample collection, gamma spectrometry measurements were performed quickly in a field laboratory onsite before decay of ^{140}La would make it undetectable.

The witness plates were placed downwind at nearly 110 locations in a fan pattern extending to the north of the release point, covering north-northwest through northeast transport directions, consistent with climatological morning winds from the south or south-southwest. Witness plates were placed on east-west lines with 100-m north-south spacing for the first 600 m downwind of the release, 200-m spacing from 600 m to 1 km, and 500-m spacing from 1 km to 2 km. East-west spacing was 100 m for the first 300 m and 200 m beginning at 400 m north of the release. During setup, some witness plate locations were modified due to the presence of subsidence craters or fenced-off areas. Since the exact path the radioactive plume would take was unknown, it was necessary to cover the full range of plume directions that were consistent with acceptable (e.g., predefined go/no-go) wind conditions. This was done with the expectation that many of the sampling locations would not collect positive samples and would be excluded

from laboratory measurement based on both the measured wind direction at release and the results from the vehicle survey. The range of acceptable wind directions was based on climatology for the site and influenced by the presence of the Mercury Highway to the west of the release. The sample locations in alternating west-east rows were offset relative to neighboring rows to reduce the risk of a narrow plume missing most sampling points. Ten of the witness plate sites were set up as "comprehensive sampling sites" (CSS), with five witness plates placed in the manner described below. After the release, several of the CSSs were also used as soil sampling and in situ measurement locations.

To collect radioactive lanthanum oxide powder for rapid measurement, witness plates comprised of "tacky mats" adhered to foam-core board were staked to the ground with aluminum tent stakes (Fig. 3) and took several days for installation of the witness plate array. The tacky mats used were 45.7 cm × 76.2 cm Criti Clean CRN-1836-30. The witness plates were placed lengthwise in the north-south direction; staking them to the ground usually resulted in a slight bowing in the shorter east-west direction. The day before the release, the top non-sticky layer was removed from the tacky mats so that they would be ready for collection. The southernmost rows—those closest to the release point—were uncovered on the morning of the release; there was insufficient time to uncover all 100+ sites on the morning of the experiment.

As noted earlier, some sampling sites were set up with five witness plates. The purpose of this was to investigate deposition variability on the order of a few meters. The five witness plates were placed in a "+" pattern, with the ends of the "+" pointing in the four cardinal directions and the distance from each end to the center of the "+" being 3 m (Fig. 3).

Fig. 3. CSS with five witness plates.

Air samplers. Two related types of air samplers were set up to collect the radioactive tracer; the intake/filter system was the same for both types. Six samplers set to run with an airflow of 0.47(3) $m^3 s^{-1}$ were deployed as follows:

- Two battery-powered, radio-controlled units were set up near the centerline at 1.5 km down range. These units were remotely activated just before the release and ran for 20 min; and
- Four large gas-powered leaf blowers with modified intakes holding filter material were located 2 km down range. These high volume blowers were used previously in Canada for a similar tracer release study (Okada et al. 2016).

Source transport

The transport shielding was designed based on 9.2 × 10^7 kBq of ^{140}La activity at the time of pickup by the commercial shipping company. Monte Carlo N-Particle (MCNP) Version 5 (Forster et al. 2004) transport calculations showed that if centered in a type "A" 55-gallon drum, 10 cm of lead shielding around the source would reduce drum surface dose to less than 2 mSv h^{-1}—below the "Yellow III" transport maximum surface dose level.

The procedure to extract the source began with removal of a lead-filled transport plug at the top of the shield. The bolts were loosened using an extended socket, and then a 4.9-m bar with a cable and hook was used to lift the plug from the shield. While the shield was open, staff remained at the ends of the bar, outside of the upward-directed radiation shine path. A second 4.9-m bar fitted with the "source extraction mechanism" in the middle was then lowered onto the source can. The extraction mechanism consisted of a 9.1-kg steel cylinder suspended by two stiff rods from the bar. The bottom of the steel cylinder included two ~0.75-cm-diameter "needles," which were used to puncture the source cannister and provide access to the La_2O_3 powder. Dry nitrogen was forced into the source canister through one of the needles and out the other; La_2O_3 powder was entrained in the exiting dry nitrogen. Using a remote valve control switch,

the operator was able to flow nitrogen through the source container, evacuating a majority of the radioactive La_2O_3 particles. The exit-port needle emptied into the inlet airstream of the air cannon described below.

The PRex used an air cannon device (Fig. 4) was expected (based on testing at PNNL) to loft particulate materials over 35 m into the air at the lower acceptable limit of wind speed (1.8 m s^{-1}) and ~10 m into the air at the high acceptable limit of wind speed (6.7 m s^{-1}). This type of air cannon is powered with liquid CO_2 and uses the Coanda effect (Coanda 1936), created by a nozzle structure and expansion of the CO_2 into gas to produce an upward-moving air column. For meteorological transport calculations (Rishel et al. 2016), the model was run with the assumption that particles were injected into the atmosphere at the maximum height of the plume column. The authors' goal was to extend the simulated "vent" to last for several minutes, requiring the use of several bottles of CO_2 linked through a manifold.

Meteorological conditions

Fig. 5 shows wind observations across the NTS site on 14 May 2013 at 10:45 a.m., approximately 10 min prior to the PRex release. Winds within the Yucca Flat basin were generally southerly, which is typical for the area and time of day in the absence of any large-scale weather systems. In Area 6, winds were generally from the south at 4.5 m s^{-1}, and became more south-southeasterly farther to the north. Upper-air wind measurements were made ~1 km to the west of the release using a weather balloon capable of providing reliable vertical profiles of wind, temperature, and humidity. Three weather balloons were launched on the day of the PRex release, including an early morning launch, and pre- and post-PRex release launches. The vertical profile indicated that winds were southerly at the surface (from 180°) and became slightly south-southeasterly (i.e., ~170°) up to 1,400 m above mean sea level (ASL), where they became more south-southwesterly (i.e., between 190° and 230°) with height. Wind speeds

Fig. 4. Left: Cross-sectional view of the source shipping/transfer system. This image shows the system with the source extraction plug installed. Right: the PRex release occurred with surface winds of 4.5 m s^{-1}; the dual cone air cannon injected the ^{140}La particulate and orange smoke to 10(1) m, with the particulates introduced through the upwind cone (e.g., the particulates were riding on the upper half of the visible plume. A test run in light winds 1 wk prior to the test injected material more than 40 m into the air.

ranged between 4.5–6.7 m s^{-1} up to ~2,400 m ASL. Temperature decreased with height and relative humidity increased with height through this layer. The released particulate material was transported by the near-surface average winds, which were approximately 175° at 5.4 m s^{-1}. This wind direction is consistent with the observed plume footprint.

Source dispersal

The PRex release began at ~10:55 a.m. on Tuesday, 14 May 2013 and lasted approximately 2 min. The release system did not contain an on-board radiation monitor, so the exact time period over which the material was released is not known. The supply of nitrogen carrier gas was cycled on and off several times during the first minute of the release, then was left flowing (on) for several minutes. Smoke candles were used during the release, with the smoke entrained along with the radioactive source particles. This allowed visual observation of the plume; the release height was estimated as 10(1) m based on video recordings of the event.

Sampling and laboratory measurements

Collection of witness plates. After an initial collection transit along the 1 km north row, when the plume location was still not fully known, the witness plates in general were collected from north to south. This sequence reflected an
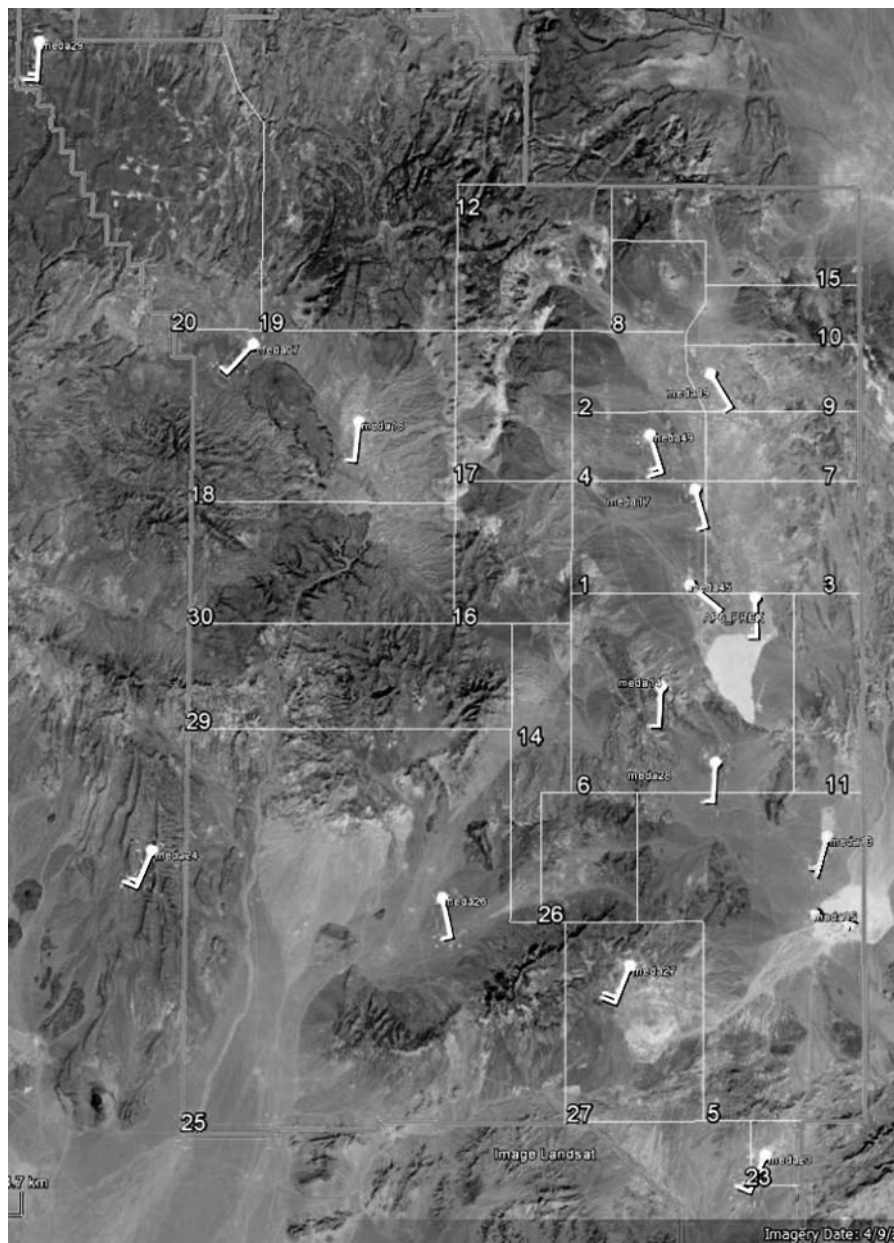

Fig. 5. Wind observations across the NTS site on May 14, 2013, 10:45 a.m.

effort to avoid resuspending particles in the more active regions so they would not be deposited onto uncollected witness plates further north. The most active regions of the plume area were also off-limits during the first day of survey activities until radiological control technicians completed procedures to verify acceptable levels of removable contamination. The area immediately around the release point remained off-limits for several weeks. Witness plates at a given location were collected at the same time, and collection of samples from all the sites was accomplished over 1.5 d, Tuesday afternoon and Wednesday. The witness plates remained uncovered until they were collected, leaving open the possibility of additional collection of radioactive materials due to resuspension. Windy conditions occurred throughout the late-morning and afternoon after the release, with steady wind speeds in the vicinity of 10 m s^{-1}, gusting to 15 m s^{-1}; dust devils were also observed to transit the plume area over the coming days (including prior to the collection of witness plates). The vehicles used to collect the witness plates could also potentially cause resuspension and to the extent possible were driven downwind (north) of the immediate sampling locations.

Consistently strong and gusty winds were present during sample collection, and it appeared that the daytime temperatures, winds, and personal protective equipment (PPE) would introduce a risk of personnel overheating while working in the field with no shade. To mitigate the impact of these conditions, samples were collected as quickly as possible in the field, individually bagged, and moved to a steel shipping container positioned near the exit to the CA. Gloves were changed between sample collections. The high winds often made it difficult to place the witness plates into the plastic bags, as did the bowed corners (Fig. 6).

The witness plate samples were processed on a table in the shipping container; each sample was removed and placed on top of its own plastic bag to minimize cross contamination. All of the tacky mat layers were removed from the foam and folded in a predetermined and consistent manner to fit inside a new, sealable sample bag for measurement on a high-purity germanium (HPGe) detector in the field laboratory. The samples were folded into a square having area similar to the HPGe detector face, so the sample bag could be laid flat on the detector face for counting. Although this process resulted in extra unexposed tacky-mat material being included in the sample, it was more expedient to include all of the tacky-mat layers in the sample bag rather than trying to separate only the top-most layer.

Soil sampling. In addition to the witness plates, at several sampling sites, three different soil sampling techniques were used during the PRex experiment. The principle motivator was that the three techniques were under consideration by the CTBTO for soil sampling during an OSI. While the above-mentioned witness plates are ideal
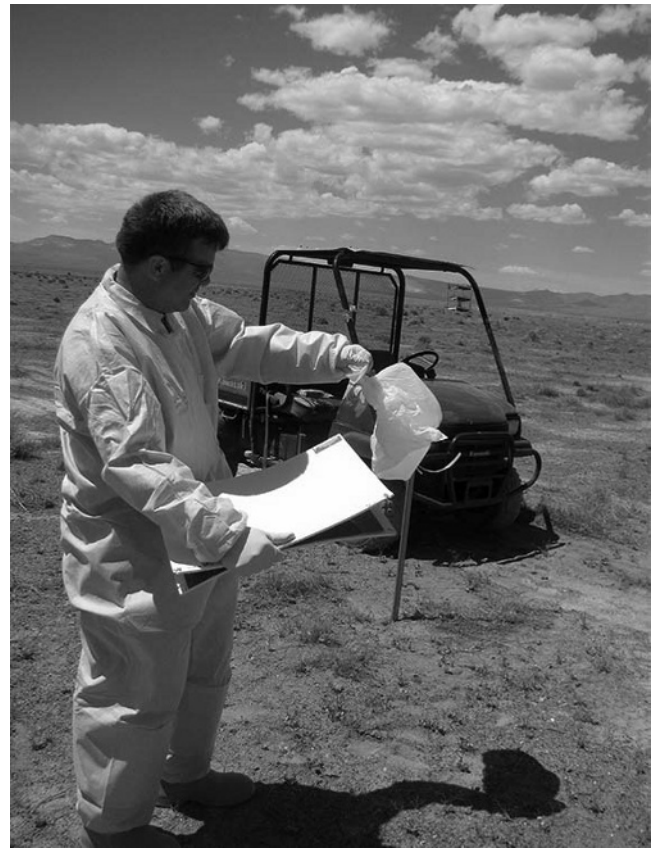

Fig. 6. Collection of tacky mat witness plate hampered by the bowed shape and the wind.

for pre-planned characterization of a release event, soil sampling is an obvious method of choice for a post-event inspection of possible ground deposition. Soon after an event, before significant weathering or precipitation, one can expect deposited radioisotopes to be on the very top of the soil surface and not to have migrated significantly into the soil matrix. Three techniques were compared to study whether limiting samples to the top-most soil material would improve detection. The techniques tested were troweling, collecting small particulates from the surface with an inverted tacky mat, and collecting small particulates with a vacuum cleaner. Samples using the different techniques were collected adjacent to each other, near witness plates to allow for comparison of techniques.

A simple hand trowel was used to scrape and scoop the topmost (as thin as the collector can manage) layer of soil. To do this, a 50.8 cm × 50.8 cm polyvinyl chloride (PVC) pipe square template was placed on the ground. (The sampling area was taken to be 45.7 cm × 45.7 cm; the idea being that one could only get to within about 2.5 cm from the sides with the trowel.) After removal of large rocks, the topmost soil was troweled into a 0.454-L sample jar of a diameter similar to the HPGe detector face. The top layer of soil was collected to within ~2.5 cm of the PVC template, providing a sample covering a 45.7 cm × 45.7 cm

Fig. 7. Soil sampling with a trowel.

area (Fig. 7). Hardpan surfaces were broken up by chopping the soil with the trowel. Inexpensive plastic trowels were used to collect a single sample to prevent cross contamination between samples.

The second technique was to place a 45.7 cm × 76.2 cm tacky mat (the same as the ones used as witness plates but without foam backing) sticky side down on the surface to be sampled after removing large rocks and avoiding plants. A single layer of tacky mat was typically used, pulled off a larger pad of the material. The mat was patted and rubbed down on the soil surface (Fig. 8) and then folded in a fashion similar to that of the witness plates. Due to the large number of small pebbles typically collected by this method, these folded mats did not fit into plastic bags like the witness plate samples, so they were placed in sample jars like those used for the troweled samples.

The third technique involved the use of a handheld vacuum cleaner [a Dirt Devil™ (TTI Floor Care North America, Glenwillow, OH, USA) brand Handy Duo]. These

units are battery powered and were observed to last for only three to four sample collections before needing recharge. This sampling technique used the same square PVC template as was used for troweling. To prevent coarse material from reaching the finer filters, a pre-filter was improvised by inserting a knee-high nylon stocking into the vacuum cleaner and holding it around the mouth of the vacuum cleaner while the panty hose toe was sucked inside. (No vacuum cleaner attachments were used.) The vacuum cleaner was then moved back and forth over the soil surface as close as possible (Fig. 9). In areas where the soil was loose enough to be sucked up en masse, it was necessary to keep the vacuum cleaner off of the surface a bit. As in the tacky mat case, the numerous small pebbles typically present were a challenge for this technique. The entire surface area was vacuumed twice. Next, the vacuum cleaner was disassembled over a sample jar like those used for the previous techniques. The pre-filter (stocking) and the coarse material inside were placed in the sample jar, along with the material in front of the vacuum cleaner filter. This filter was removed and shaken into the jar, along with the finer material between it and a second filter. The outer vacuum cleaner tube and initial collection area could be mostly wiped down with wet wipes between samples, but this was not true for the inner filters, which trapped moisture in their filter pores. Thus this technique appeared to be substantially more susceptible to cross-contamination in addition to its logistical difficulties relative to the other two soil sampling techniques.

Air sampling. The gasoline-powered high-volume samplers placed 2 km downwind of the dispersal were started at approximately 9:30 a.m. and allowed to run until the samples were collected (remote start and stop was not available on these samplers). The battery-powered high-volume samplers at 1.5 km downwind were remotely
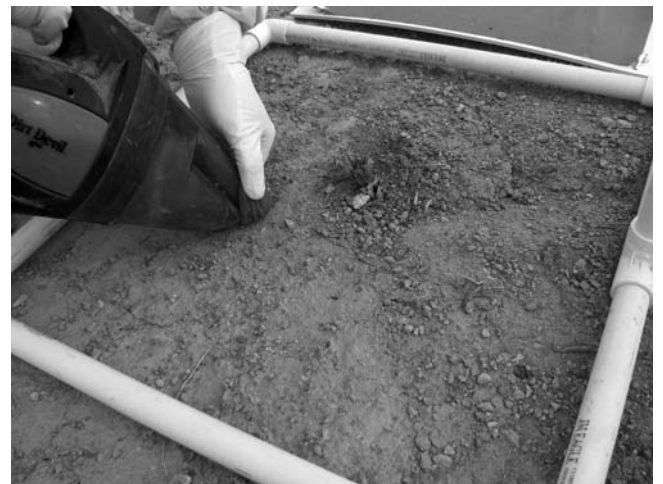

Fig. 8. Soil sampling with a tacky mat.

Fig. 9. Soil sampling with battery-powered vacuum cleaner.

started a few minutes before the source release and allowed to run for 20 min. Filter paper samples from both the gasoline- and battery-powered samplers were collected between 3 p.m. and 4 p.m.

In situ measurements

Handheld and human-portable radiation detectors and spectrometers were used to take in-situ measurements at several of the CSS locations. The instruments included a Canberra Falcon (Canberra Industries, Inc., Meriden, CT, USA) (HPGe), an ORTEC (ORTEC, Oak Ridge, TN, USA) detective (HPGe), a FLIR Identifinder (FLIR Systems, Wilsonville, OR, USA) (NaI(Tl)), and a Ludlum 711 (LaBr$_3$). A Thermo Packeye (Thermo Fisher Scientific, Waltham, MA, USA) backpack PVT-based survey system was also used to collect survey data along several transects of the deposited plume; the Packeye detector is based on a cylindrical PVT detector of ~800 cm^3. Although PVT has some minimal energy windowing capability, the instrument was used in gross count/dose rate mode.

Survey

Ground vehicle survey. The ground survey was conducted using two RSI 5.1 cm × 10.2 cm × 40.6 cm NaI (Tl) log spectrometers. The spectrometers were loaded into the back of a Kawasaki Mule, a small diesel 4 × 4 ATV (Fig. 10). Two separate GPS units were used for these measurements. One GPS unit fed directly into the RSI 700 system, which combined and archived the NaI(Tl) data. A second GPS was used for the steering indicator, which guided the driver along parallel lines with a known separation. Navigation heading information came from the Raven light bar. The Raven light bar displayed course corrections

Fig. 10. Survey equipment following the release of ^{140}La. The two NaI(Tl) detectors were packaged in large Igloo coolers, and are next to each other, oriented side-to-side in the bed of the Kawasaki Mule. The coolers were used to limit thermal gradients; rapid temperature changes can crack NaI(Tl) crystals.

based upon differentially corrected GPS. The system was originally designed for guiding farm vehicles; e.g., for a tractor planting a field. The selected mode of the system was to produce swaths parallel to the road just south of the release point. For this survey, the lines were driven at 50 m separation near the release point and at 100 m separation at larger downwind distances. Although it was possible to perform the ground survey with only a driver, it was advantageous to have the driver focused on driving and watching the steering indicator while an equipment operator monitored collection of radiation survey data on a laptop. The advantages included both more complete data collection and improved safety.

Aerial survey. RSL performed a fixed-wing survey starting within approximately 1.5 h of the ^{140}La release. RSL's Beechcraft B-200 was equipped with an RSI survey system consisting of six 5.1 cm × 10.2 cm × 40.6 cm NaI (Tl) spectrometers. Flight lines were flown 150 m above ground level (AGL) at a speed of 260 km h^{-1} at a line spacing of 150 m. These flight parameters limited the sensitivity of the aerial survey; however, they are realistic for a large area, fixed-wing survey during an OSI.

Field laboratory. A field laboratory was set up in a Quonset hut several miles to the southwest of the release point. The laboratory included three ~50% relative efficiency high resolution germanium HPGe gamma spectrometers (Canberra), as well as two low resolution 7.62 cm × 12.7 cm NaI(Tl) spectrometers. The lab was staffed by two people during the first day of measurements and by three people after sample collections were completed.

Straightforward 10.2-cm-thick lead shields were built on stands to accommodate two of the HPGe detectors (Fig. 11) and the two NaI(Tl) detectors. The third HPGe detector, supplied by NSTec, was housed in a Canberra portable shield. This detector was calibrated on the Canberra ISOCS system (Venkataraman et al. 2005), and models were developed to establish detection efficiencies for the sample geometries during buildup and preparation of the laboratory. Efficiency calibrations were also prepared for the PNNL-supplied HPGe detectors using the ISOCS method.

The first samples received into the field laboratory were collected by the high volume air samplers 2 km downwind from the release and were brought to the laboratory late in the day on Tuesday, 14 May. A large fraction of the samples were received into the lab on Wednesday, with additional samples received Thursday and Friday morning. Counting was performed beginning late Tuesday and was completed Friday morning, followed by tear-down and loading of the laboratory late Friday morning. Samples were typically counted for 15 min; samples that had weak positive indications for ^{140}La were extended to 30 min of counting to improve the result.

Fig. 11. Two ~50% relative efficiency HPGe detectors inside of 10.2-cm-thick lead shields. These detectors are set up in the field laboratory.

The two NaI(Tl) detectors were included in the field laboratory because they were expected to be more reliable than the HPGe detectors; the utility and sensitivity of the NaI(Tl) detectors were unclear. Early sample measurements

made it clear that the NaI(Tl) sensitivity was far inferior to that of the HPGe detectors due to the significant background rates relative to the ^{140}La activity [in the NaI(Tl) spectra, natural ^{40}K was a significant interference for the 1596 keV ^{140}La line]. One of the NaI(Tl) detectors also suffered from degraded energy resolution and was essentially not useable. After early attempts to use the NaI(Tl) detectors in a screening mode, they were no longer used as they were not providing added value. It seems likely that scintillators, with their significantly poorer energy resolution compared to HPGe, would only provide added value in the field laboratory if employed in a high detection efficiency coincidence configuration that would allow the NaI(Tl) to provide improved isotopic discrimination despite the lower energy resolution (Wogman et al. 1969).

RESULTS

Air sampling

The highest activity air sample of ^{140}La was collected by a gas high volume sampler at 2 km, which was in the path of the narrow plume trajectory measured by ground
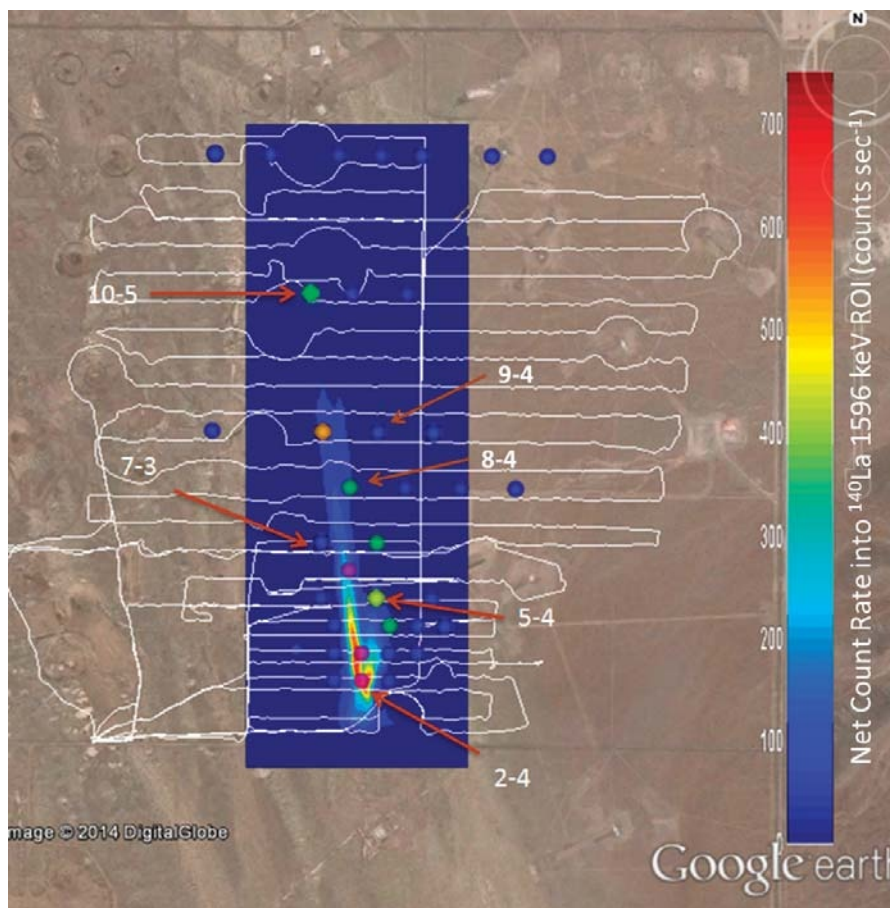

Fig. 12. Tacky mat locations (filled circles) overlaid on contours of ^{140}La count rate extracted from the ground-based vehicle survey data. The contour scale is plotted as counts per second into the ^{140}La ROI. The color scale for the tacky mat activity concentration results was set to be similar to the scale for the vehicle survey results (to guide the eye); however, specific results should be read from Table 1.

surveys. This sample (furthest east) is ~33 times higher in activity than the other three high-volume gas samplers located at 2 km. The next most active air sample of ^{140}La was collected by a battery powered high-volume sampler at 1.5 km, also in line with the observed ground deposition. All of the high-volume air samples collected during the period of the release resulted in positive collections; the air samplers were significantly more sensitive than the vehicle-based ground survey due to the high volume of air sampled. However, in the event of an OSI, high-volume air samplers would only be expected to collect resuspended material—sampling from the original plume passage would not be available.

Vehicle survey

Fig. 12 provides an example of the vehicle survey results when limited to background-subtracted count rate collected in the 1,554–1,674 keV region of interest (ROI) containing the ^{140}La 1,596 keV gamma line. The vehicle survey data is presented in counts per second (cps) for the ROI. Fig. 12 also presents a selection of witness plate locations (discussed below).

A MCNP model was constructed to estimate the conversion from the observed ^{140}La 1,596 keV count rate to the areal activity concentration. The model assumed a uniform disk surface source with 40 m radius, included a crude model of major mass components of the Kawasaki Mule, and included the NaI(Tl) crystals in aluminum cans 1 m above the ground surface in the bed of the Kawasaki Mule. The conversion factor estimated using this MCNP model for the survey data is 17(4) cps into the ROI per kBq m^{-2} of deposited activity.

The vehicle survey established the presence of a narrow deposited plume of ^{140}La activity extending to the NNW from the release point. This main plume was on the order of 200 m in width when measured down to the detection limits available from the vehicle survey. A smaller lobe of the deposition can be seen extending to the NNE. This portion of the plume may have been deposited during a brief wind shift that occurred during the release (apparent in the video record of the release) or perhaps via resuspension of activity initially deposited in the near vicinity of the release. Integration of data from the vehicle survey provided a rough estimate that only 7% of the expected ^{140}La activity was deposited, while comparison of meteorological transport modeling and the collected data provided an estimate closer to 20% of the activity (Rishel et al. 2016).

Comparison of atmospheric dispersion modeling to radiometric results

Post-analysis of the release was performed with HotSpot 9 (Homann and Aluzzi 2013) using observed weather data and a source injection height of 10 m and 40-μm particle diameter. The modeled and surveyed plume directions were in agreement, although the lobe that extended in a more northerly direction was not indicated in the model. It appears likely that this lobe was created by dispersal of material from the release point at a later time, after the winds shifted more northerly (or perhaps by material transported during a brief wind shift that occurred during the release, visible in video of the release).

Deposition contours modeled with an assumed source release of 3.7×10^7 kBq indicated that the deposited plume measurable by the RSI vehicle survey, with a minimum detectable concentration (MDC—see discussion under "Comparison of in-situ and RSI survey results" below) of ~1 kBq m^{-2}, would have extended beyond 5 km downwind. However, the plume mapped by the RSI vehicle survey extended only 1.1(1) km downwind. Deposition contours based on a 7.4×10^6 kBq (20% of the original source) release resulted in a HotSpot simulation of deposition of the particulate La$_2$O$_3$ that was more consistent with both the RSI-measured ground deposition and the witness plate results. Both the integration of the vehicle survey data and the comparison of HotSpot simulated ground deposition to measured deposition contained significant uncertainties; combining the results from the two methods gave an estimate of 14% of the original source activity being deposited in the near-field plume, with an estimated uncertainty on the order of 30–35% relative error. The measured plume was also narrow compared to the modeled plume, by roughly half. This is likely due to the short duration of the release: the HotSpot model integrates over a 10-min period, while the release lasted only 2 min.

Comparison of vehicle survey and witness plate results

The surface area and measured activity of the tacky mats was used to determine the deposited activity concentration. The equivalent RSI measurements are based upon the interpolation of the survey data to the location of the tacky mats. The results have been corrected to a consistent set of units; however, the tacky mat results represent a measurement of deposition on a 0.35 m^2 surface area, while the MCNP detection efficiency simulations indicate that the RSI survey system is sensitive to radioactivity extending to significant distances from the measurement point (on the order of 40 m). The deposited plume activity concentration derived from field laboratory measurement of witness plates is compared to the vehicle survey result for various witness plate locations, as shown in Table 1 and graphically in Fig. 12. With the exception of site 7–3 (discussed in the next section), all of these sites indicate reasonably good agreement between samples collected at a given site and between the tacky mat and vehicle survey results.

Table 1. Comparison of activity concentration determined by field laboratory measurement of tacky mat witness plates and via the vehicle based NaI(Tl) mobile survey.

Location	Tacky mats (kBq m^{-2})	RSI (kBq m^{-2})
2–4	19.6 (4)	29 (7)
5–4	3.4 (1)–4.4 (1)	3.7 (9)
7–3	0.019 (2)–0.118 (8)	7 (2)
10–5	1.85 (6)–2.13 (6)	2.2 (5)

Variability at comprehensive sampling sites

The data from CSS 7-3 are an interesting case to consider. In this instance, there were five tacky mats collected and measured in the field laboratory. The results had a wide range, from 6.6(7) Bq to 41(3) Bq. The neighboring site to the west (7–2) had a limit value result, while the site to the east (7–4) recorded 825(23) Bq. Closer examination of the data at site 7–3 indicates that the lowest value, 6.6(7) Bq, was observed at the western sampling point. Relatively consistent values of 24 (2) and 17(1) Bq were observed at the center and southern sampling points, respectively. Finally, values of 34 (2) and 41(3) Bq were observed at the northern and eastern points. While not conclusive, these results are consistent with a factor of six increase in ^{140}La deposition over the scale of a CSS, ~6 m. Inspection of Fig. 12 shows that this sampling site was in a location where there was a steep upward gradient of the plume when moving from west to east. The corresponding RSI vehicle survey result was 6.7(16) kBq m^{-2}, significantly higher than results from the tacky mats. This likely indicates that the survey value was biased higher due to the higher activity nearby to the east.

Aside from the variation in witness plate results at sampling site 7–3, the results from the RSI vehicle survey and the witness plate samples are in good agreement, as seen in Table 1. Two more of these sampling locations were also CSSs, 5–4 and 10–5. Data from the tacky mats collected at CSS 5–4 indicate there may be a minor variation of deposited activity across the sampling site, while the results from 10–5 indicate a consistent deposition across the CSS. The activity levels at both sites were higher than at 7–3. Neither of these sites appears to lie on a steep activity gradient (Fig. 12), so the good agreement between samples does not rule out the possibility that the results at 7–3 are evidence of a significant deposition gradient across the 6 m of a CSS location.

Aerial survey

The aerial survey provided a positive detection of the source dispersal location, and a weak indication of the presence and direction of the deposited plume. The strong detection of the source dispersal location and shipping barrel did indicate that a significant fraction of the original source activity remained in the nearby vicinity of the dispersal system, consistent with the observation that the downwind plume contained only 10–20% of the source activity. A helicopter-based survey, flown at lower elevation and closer line spacing, would have provided a much more detailed map of the plume.

Backpack survey

Two transects of the plume measured with the Thermo Packeye system provided dose-rate cross sections of the plume that were consistent with vehicle survey results collected along the same transects. The results of the backpack survey were only compared in a qualitative way against the RSI data and resulted in a similar plume profile and width. In plots of the plume profile, the peak dose rate reported by the backpack survey instrument was lower (roughly half the amplitude, in one case) than the count rate profile reported by the RSI NaI(Tl) vehicle survey. The backpack survey also provided a less detailed view of the plume profile. After this preliminary examination of data from the backpack, data analysis resources were committed to the vehicle survey data, and no further work was done with the backpack data.

Results from soil sampling methods

There were three CSSs where samples taken by placing tacky mats sticky-side-down on the ground (Fig. 8) resulted in measurable activity (5–4, 7–3, and 10–5), with two such samples at the first two locations but only one at 10–5. The tacky mat samples measured activities of between 6–12% of those of the initial witness plates in the same CSSs, implying an order of magnitude reduction in collection of the radioactive particulates relative to "ground truth" as measured by deposition on witness plates. It would have been possible to apply the tacky mats to more than a single area to increase the total activity collection, which was not done in this experiment. It appeared that the technique could have been doubled in its soil collection before filling up the tacky mat with too much sample. This approach could improve the sensitivity of the method; however, it would not correct the difference in observed activity concentration.

Positive detections were noted in troweled soil samples taken at two locations corresponding to witness plates: 5–4 and 10–5. In one of these locations (5–4), more than one soil sample was taken. In that case, the measured activity for the two samples agreed to within 7%. Results from these soil samples indicated activity concentrations between 15–20% of those of the witness plates. Again, it would have been possible to collect larger soil samples than was done with the template, which was not done in the experiment. Soil collection using this technique could have been doubled before filling the sample jars; however, sensitivity gains would be limited because detection efficiency for soil in the upper half of the jar is substantially lower than for the lower half.

Vacuum samples were taken at a number of locations corresponding to witness plates; positive detections were noted at sites 2–5, 5–4, and 6–3. Limited battery life prevented the collection of more than one sample at each location. The vacuumed sample at 2–5 measured 10 times the activity concentration of the "ground truth" witness plate, while at the other two locations the vacuumed samples measured 10–15% of the activity. A suitable explanation for the site 2–5 result was not determined. This technique could have easily been increased a factor of four to five times in its collection area before acquiring a full sample, although power and cross contamination issues would remain to be solved.

Based on the above discussion, the troweling and vacuum sampling techniques performed better in terms of collecting more recently-deposited surface activity for a given area than applying tacky mats. When one considers the total amount of recently deposited sample that can be collected, a vacuum approach is clearly favored. Drawbacks to the vacuum approach, however, include cross-contamination of equipment and the inability to provide power to the equipment adequate for the time needed to collect samples. Further thought and design is needed in terms of how to include sample filters in the vacuum cleaners. This technique would also be degraded more so than troweled soil samples if there were intervening periods of precipitation that moved radionuclides deeper into the soil. Soil sampling, even though it obtains much more soil depth than necessary for surface deposition, still provided reasonably robust samples in this experiment.

Minimum Detectable Activities (MDAs) (Currie 1968) based on Canberra Genie 2000 reporting for the limit-value soil and inverted tacky mat samples indicated a typical range of ~3.7 to 11.1 Bq, although some samples were found to have MDAs nearly an order of magnitude lower. Converting these MDAs to MDCs (activity per unit area) based on the $0.21 \, m^2$ sampling area gives a range of ~17.6 to 52.8 $Bq \, m^{-2}$. Finally, if one assumes that only 20% of the deposited activity was collected using these sampling methods (relative to the "ground truth" witness plate results), the sampling achieved a typical MDC range of ~88 to 264 $Bq \, m^{-2}$ detection sensitivity for samples brought to the field laboratory. Several of the field laboratory sample measurements exhibited lower natural background levels associated with the sample, resulting in MDAs roughly an order of magnitude better than the stated range (e.g. ~10 $Bq \, m^{-2}$).

Comparison of in situ and RSI survey results

Measurements with the LaBr$_3$, NaI(Tl) and HPGe handheld instruments were performed at a number of locations. These locations are included in Fig. 12 and listed in

Table 2. Comparison of deposited activity measured by handheld instruments, compared to the vehicle survey RSI results. The values for the 3 handhelds are for 10-min measurements, while the RSI values are for a 1 s drive-by measurement. Where shown as a range of values, the table provides the range of results for 5 measurements at a CSS location.

Sample location	NaI ($kBq \, m^{-2}$)	LaBr3 ($kBq \, m^{-2}$)	HPGe ($kBq \, m^{-2}$)	RSI ($kBq \, m^{-2}$)
2–4	26 (5)	26.7 (5)	18.5 (40)	28 (6)
			Ortec	
5–4	3.7 (9)	3.5 (9)	2.6 (6)	3.6 (9)
			Canberra	
8–4	3.6 (9)	4.5 (11)	3.3 (8)	2.8 (7)
			Ortec	
9–6	<0.5	0.7 (2)	0.7 (2)	<1.1
			Canberra	
Typical MDC	0.5	0.46	0.074	1.1

Table 2. The conversion of the count rate in the 1,596 keV peak (^{140}La) to deposited activity was based upon MCNP simulation of the detection efficiency for each detector, calculated in a manner similar to that described previously for the vehicle survey. The source was simulated as uniform activity deposited on a 40-m-radius disk, and detectors were simulated at 1 m above the ground, consistent with the experimental emplacement.

Judging from the results of Table 2, it appears that there is a minor calibration issue between these various detectors, particularly with the HPGe detectors typically arriving at lower values when compared to the other instruments. In general, these instruments are in reasonable agreement.

Table 2 provides typical Minimum Detectable Concentrations (MDCs) observed for the four portable detectors for 10-min measurements, as well as for the RSI vehicle survey system with 1 s integration time. The hand-held detectors were placed approximately 1 m above the ground in areas of active ^{140}La contamination and spectra were acquired. Background counts in the 1,596 keV ^{140}La peak were determined from these spectra, and those values were used in the Curie equation to determine the MDCs:

$$MDC = \frac{4.65 \times \sqrt{N_B} + 2.71}{\gamma \times \varepsilon \times T}, \qquad (1)$$

where N_B is the background count rate, γ is the 1,596 keV ^{140}La gamma emission intensity (0.954), and ε is the detection efficiency. Detection efficiency was determined by MCNP modeling over a uniform ground deposition (disk source at ground level in the model, with 40-m radius) with the detectors modeled at 1 m above ground and relatively simple detector geometry models (correct active volume

dimensions and major attenuation layer). The Canberra HPGe handheld had an MDA nearly an order of magnitude more sensitive than the scintillator-based handheld detectors. Note that the RSI survey MDC reported in Table 2 is 1.1 kBq m^{-2} for a 1-s measurement, only a factor of 2 poorer than the 10-min MDCs for the scintillator-based handheld instruments. This performance over short integration times is due to the much larger NaI(Tl) detectors in the vehicle-based survey system.

DISCUSSION

The vehicle ground survey was found to provide the most comprehensive data set for characterizing the location and deposition activity in the plume. This method also allowed the ground-based experiment team to cover the most area relative to time invested. However, the ground-based vehicle survey was not as sensitive as the in situ and sampling techniques employed. The narrowness of the plume, only on the order of 200 m in the vehicle survey data, showed that survey and sampling techniques must cover a comparably tight spacing or risk missing a plume for a radioactive vent.

In situ measurements with HPGe detectors were found to be roughly an order of magnitude more sensitive than comparable measurements with scintillator-based detectors, which were themselves more sensitive than the vehicle ground survey. The sensitivity of soil samples measured in the field laboratory was roughly comparable to the in situ HPGe measurements, although some laboratory sample measurements achieved more sensitive results. From this work, it appears that the selection of emphasis between in situ vs. sampling/field laboratory measurements should be based on throughput capability and other operational considerations (e.g., ability to archive samples). Both methods provide useful capability to an inspection team. However, the data provided is sparse compared to a vehicle-based ground survey; if possible, sampling and survey resources should be focused on areas identified as interesting through other techniques.

The backpack survey results did not provide significant sensitivity enhancement over vehicle survey in the Kawasaki mule. Further, this survey method was very slow compared to vehicle-based survey. A backpack survey should only be used in situations where vehicle survey is not possible or when the backpack is carried by an inspection team member while performing other duties.

All of the soil sampling methods were observed to provide surface activity concentration level estimates that were low compared to the "ground truth" values determined through measurement of the witness plate samples, roughly in the range of 6% to 20%. This likely indicates incomplete collection of the deposited activity within the

sampling area. A rough ordering of the techniques in terms of percent of "ground truth" activity collected for the same surface area was 1) trowel soil collection, 2) vacuum cleaner, and 3) inverted tacky mat; however, the latter two techniques could easily be designed to collect from larger areas than were used in our soil collection. The vacuum cleaner method appeared to provide a good technique for collecting recent debris but suffered from poor battery life and high potential for cross-contamination. The tacky-mat method also collected a sample from mainly the top layer of soil but is challenging to accomplish during windy conditions. Calibration of the measurement geometry for the bulky tacky mats with soil is also challenging. Troweling a surface layer of soil was relatively straightforward, allows for good contamination control, and provides a repeatable sample geometry for laboratory measurement. This is also likely the best approach for inspections that occur at longer times after a detonation event when radionuclide debris may have migrated downward into the soil.

CONCLUSION

Experimental dispersal and atmospheric transport of radioactive particulates

The PRex radionuclide release was successfully conducted during the week of 13 May 2013. The experiment, designed to mimic a small vent of radioparticulates from an underground nuclear explosion, involved lofting approximately 3.7×10^7 kBq of ^{140}La in the form of 32-45 μm diameter La$_2$O$_3$ powder via an air cannon. The contamination plume deposited was transported by natural atmospheric processes, primarily deposited within the first 2 km downwind, and appeared to be a distribution representative of deposition that could be experienced after small-scale venting from an underground nuclear test. A narrow plume was measured both with a vehicle ground survey as well as inferred from laboratory analysis of witness plates. Estimates of the total activity deposited in the near-field plume, based on integration of the vehicle survey data and through comparison to HotSpot meteorological modeling of the plume, indicate that only ~14% of the source activity was successfully injected into the atmosphere, transported, and deposited in the near field contamination plume. While this was unfortunate in terms of establishing the intended conditions, sufficient ^{140}La activity was transported to complete the planned survey, sampling, and measurement tests.

Implications for OSI

The deposited activity afforded an opportunity to observe significant gradients in the ground deposition, with measurements at one site showing a factor of six increase in activity concentration over a 3-m distance. This observation suggests that soil sampling at a sparse array of locations

could miss nearby evidence of a nuclear test, despite being directed into the correct vicinity by other techniques like visual observation.

In assessing sampling and survey results, HPGe detectors provided the best minimum detectable concentration results at specific locations; in situ HPGe survey and field laboratory HPGe measurements provided comparable results. The handheld scintillators were not competitive with HPGe in terms of sensitivity but likely represent a more reliable detection capability. Ground vehicle-based survey with large NaI(Tl) detectors provided the most comprehensive plume characterization and an efficient method of data collection relative to the commitment of personnel resources. The airborne survey flown at relatively high altitude and line spacing rapidly covered the plume area, however, and might easily have missed the deposited contamination if not for the high concentration in the immediate vicinity of the release. Choice of the optimal detector and technique for a particular OSI mission will vary depending on terrain, area to be covered, and available personnel.

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