Radionuclide observables during the Integrated Field Exercise of the Comprehensive Nuclear-Test-Ban Treaty

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Abstract

In 2014 the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) undertook an Integrated Field Exercise (IFE14) in Jordan. The exercise consisted of a simulated 0.5 – 2 kT underground nuclear explosion triggering an On-site Inspection (OSI) to search for evidence of a Treaty violation. This research paper evaluates two of the OSI techniques used during the IFE14, laboratory-based gamma-spectrometry of soil samples and in-situ gamma-spectrometry, both of which were implemented to search for 17 OSI relevant particulate radionuclides indicative of nuclear explosions. The detection sensitivity is evaluated using real IFE and model data. It indicates that higher sensitivity laboratory measurements are the optimum technique during the IFE and within the Treaty/Protocol-specified OSI timeframes.

Keywords: Integrated Field Exercise, OSI, CTBT

1. Introduction

Together with States Signatories, the Provisional Technical Secretariat (PTS) of the Preparatory Commission for the Comprehensive Nuclear-Test-Ban Treaty Organization (CTBTO) undertook the Integrated Field Exercise (IFE) in Jordan during 3 November to 9 December 2014. The purpose of the exercise was to assess progress in the development of On-site Inspection (OSI) capabilities and to test crucial aspects of an on-site inspection [1]. The capability to undertake an OSI is a key aspect of the Treaty's verification regime [2], and aims to clarify whether a nuclear explosion has been carried out in violation of the

Comprehensive Nuclear-Test-Ban Treaty (CTBT). The OSI aims to gather any facts which might assist in identifying any possible violator [3], including techniques to identify radioactive signatures produced from a nuclear explosion. The IFE consisted of a non-compliance scenario involving a simulated 0.5 – 2 kT underground explosion that provided the basis for OSI activities.

The IFE included laboratory and in-situ gamma-spectrometry of samples within a 1000 km² inspection area. These measurements were performed for 17 OSI relevant radionuclides (⁹⁵Zr, ⁹⁵Nb, ⁹⁹Mo, ^{99m}Tc, ¹⁰³Ru, ¹⁰⁶Rh, ¹³²Te, ¹³¹I, ¹³²I, ¹³⁴Cs, ¹³⁷Cs, ¹⁴⁰Ba, ¹⁴⁰La, ¹⁴¹Ce, ¹⁴⁴Ce, ¹⁴⁴Pr, ¹⁴⁷Nd) that are particulate in nature with half-lives ranging from 17.3 m to 30.1 y. These radionuclides allow for discrimination of a nuclear explosion from other radiation sources, such as reactor accidents or releases, nuclear waste and medical isotopes [4]. This list is designed to allow an OSI between 1 week and 2 years after the nuclear explosion, whilst being concise and avoiding disclosure of confidential information not related to the OSI. Each radionuclide has a specific reason for inclusion, and is measurable in field conditions with no common or otherwise expected interferences that could allow misidentification. The radionuclides are all fission products that are produced directly from the nuclear explosion or by activation resulting from the explosion, and pairs of some of them can provide timing information or good discrimination from reactor accidents. As an example, ¹³⁴Cs is produced in a nuclear reactor mainly by neutron capture of ¹³³Cs (¹³³Cs(n, γ)¹³⁴Cs). The ratio of ¹³⁴Cs with ¹³⁷Cs is different between nuclear explosions and reactor accidents, allowing the ratio to be used to differentiate between the Treaty-compliant and non-compliant scenarios.

This research provides an assessment of the potential to detect these OSI relevant radionuclides during the timeframe of 2 to 5 weeks for the conduct of the IFE, and during the wider timeframe of approximately 1 week to 2 years after the simulated nuclear explosion for the duration of the OSI relevant radionuclides. In doing so it evaluates two of the 17 techniques permitted under the CTBT for terrestrial environments [2]. This includes the use

of environmental sampling and analysis (soil and other samples) and in-situ gamma spectroscopy. The assessment was based on the IFE release scenario and uses a model that incorporates radionuclide production, radioactive decay, venting, atmospheric dispersion and deposition, sampling and measurement [7].

2. Modelling

2.1. Scenario

The fictional scenario, which was unknown to the Inspection Team, included the detonation of a low-yield 0.5 – 2.0 kT ^{239}Pu device within a vertical borehole at a depth of 200 m in an abandoned quarry in Jordan (named Maridia for exercise purposes). The simulation explosion zero time was 05:54:34 UTC on 28 October 2014, and occurred in limestone geology, with the formation of a cavity ($R = 13\text{ m}$) and rubble chimney that extended to within approximately 100 m of the surface. The explosion resulted in seismic aftershocks and radionuclide releases that were detected by the International Monitoring System (IMS) resulting in an OSI request on 3 November 2014. This was undertaken within a 1000 km² area from 3 November to 9 December 2014 to search for evidence of the nuclear explosion and confirm whether a nuclear explosion in violation of the Treaty [1].

2.2. Yield

The yield of a nuclear explosion is the amount of explosive energy discharged from the detonation of fissionable nuclear material (e.g. ^{235}U , ^{239}Pu) plus any fusion reactions that may occur during the explosion. It is typically measured in trinitrotoluene (TNT) equivalents ranging from < kiloton (kT) to > megaton (MT). One kT is equivalent to 10^{12} calories or 1.45×10^{23} fissions, and if the fission probability of a particular fission product is f , and its decay

constant is $\lambda \text{ s}^{-1}$, its activity following a 1 kT nuclear explosion is $1.45 \times 10^{23} f \lambda \text{ Bq}$ [8]. Using this formula the activities of each fission product for a given amount of fission energy can be calculated (using the data in Table 1), although there may be uncertainties in the contributions of slow and fast fission and in the relative contributions of fission from actinides present, e.g. ^{235}U and ^{238}U . In accordance with the IFE scenario a ^{239}Pu device with explosive yield of 2 kT and 0.5 kT was modelled (to cover the scenario range of 0.5 – 2 kT) using pooled fast neutron fission decay data [9].

2.3. Containment

The IFE scenario included venting of small amounts of radioactive material 24 hours after the explosion, with release of 1% of the noble gas inventory ($5.5 \times 10^{13} \text{ Bq}$ of xenon isotopes). This material reached the surface through various escape paths typical of an underground explosion (Fig. 1). The extent of containment is a function of various parameters, including yield, burial depth and geological factors such as rock strength, porosity, water content, carbonate and iron content, and the ability of the compressed rock to form a containment cage around the cavity after detonation [10]. Stemming measures are also typically used to prevent the emplacement hole becoming a venting pathway for radioactive material. To enable an assessment of the exercise detection capability, the particulate release was assumed to follow the noble gas release, with 1% of the particulate inventory escaping after 24 hours. This compares with a worst case scenario for prompt massive venting that would be in the range of 1 – 10% of the total radiation yield [10,11]. Based on the moderate containment measures implemented in the scenario, a prompt vent lasting 10 minutes was considered. This duration compares well with the early stages of the US nuclear weapons programme, where releases lasting up to 30 minutes were observed for underground nuclear explosive tests such as Chena (10 October 1961), Gnome (10 December 1961) and Mad (13 December 1961) [11].

2.4. Dispersion and deposition

Radionuclide concentrations vary spatially and temporally from the nuclear explosion. Atmospheric concentrations are highest close to the explosion and immediately downwind, and become reduced due to atmospheric mixing processes. The activity is also reduced by radioactive decay, with short-lived fission products with half-lives of minutes and hours disappearing quickly (although they may have longer-lived radioactive decay products). Radionuclides are also removed by deposition process, including gravitational settling, diffusion and turbulent transport [12]. These removal mechanisms are affected by a wide range of complex atmospheric conditions that may include wind, rain, snow, air density and temperature changes and variations in surface conditions. Aerosol particles $> 0.1 \mu\text{m}$ are largely removed by deposition processes to the earth's surface. For accumulation mode particles, which are those with a diameter of $0.1 - 1.0 \mu\text{m}$, removal by rain and snow (wet deposition) predominates, and for larger particles $> 10 \mu\text{m}$, gravitational settling becomes more dominant.

Radionuclide dispersion and deposition was estimated using the HYbrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model for the IFE scenario (Table 2). This uses a combined Lagrangian and Eulerian approach, where advection and diffusion calculations are made in a moving frame of reference from their initial venting location, with radionuclide concentrations calculated on a fixed three-dimensional grid as a frame of reference [13,14]. Real meteorological data that occurred during the IFE was used for model predictions.

2.5. Sampling

Particulate material that has been deposited onto surfaces can be collected during an OSI using a surface-soil sampling technique [15]. This involves using a metal template to collect

surface-soil to a depth of 0.5 cm from a 500 cm² area (~ 250 g). This technique was used during the IFE, though with smaller 2 oz (56.7 g) and 4 oz (113.4 g) aliquots being taken for gamma-spectrometry analysis. For in-situ measurements there was no sample collection, with direct measurements of the deposited surface activity within the detector field of view with results expressed as Bq m⁻² [16]. To enable a comparison between measurement techniques, the assumption was made that soil aliquots were representative of a 1 m² sampling area (although a 0.05 m² area was actually sampled). Considering the timeframe of 7 days to 2 years following the event, it is unlikely that all deposited particulate material would remain available for sampling throughout the OSI inspection timeframe, due to removal mechanisms such as surface runoff, re-suspension, vegetation growth and bioturbation [17, 18]. This study therefore provides a best-case scenario for detection of vented OSI relevant radionuclides.

2.6. Detection

The Inspection Team collected samples in various areas using the relevant CTBT On-Site Inspection procedures and equipment. Measurement of sampled particulate material was undertaken in a mobile laboratory set up in the Inspection Area using three 100 – 110% relative efficiency extended p-type HPGe detectors fitted in lead shields. The detectors were controlled using the Canberra Lynx Multichannel Analyser [19] with pre-amplifiers (model 2007). In-situ gamma spectroscopy was performed using a Canberra Falcon 5000 Portable HPGe-Based Radionuclide Identifier. In both instances, data acquisition and analysis was undertaken using the Canberra Genie Gamma Acquisition and Analysis software (version 3.3) and detection efficiency calculated using mathematical ISOCS modelling [20] for the 2 and 4 oz samples (density 1.25 g cm⁻³). To evaluate detection sensitivity, the Minimum Detectable Activity (MDA) was calculated using the Currie formula [21] with a region of interest (ROI) of 1.25 channels Full Width Half Maximum (FWHM) around the centre of the hypothetical peak. Five of the seventeen radionuclides are decay products of other relevant radionuclides

with half-lives of 17.3 m to 35.0 d and were considered in combination with their parent radionuclide. This includes $^{95}\text{Zr}/^{95}\text{Nb}$, $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$, $^{132}\text{Te}/^{132}\text{I}$, $^{140}\text{Ba}/^{140}\text{La}$ and $^{144}\text{Ce}/^{144}\text{Pr}$.

3. Results and discussion

3.1. Released activity

The detonation of a ^{239}Pu device with a 0.5 kT and 2 kT explosive yield produces 2.28×10^{18} and 9.10×10^{18} Bq of OSI relevant radionuclides, respectively. Assuming that 1% of the inventory escapes after 24 hours, this would release 6.51×10^{14} and 2.61×10^{15} Bq. Many of these radionuclides decay rapidly during the timeframe of 1 week to 2 years (Fig. 2), as the initial vented activity decreases from 2.8% (1 day after release), to 0.8% (1 week), to 0.006% (1 year) and to 0.002% (2 years). During this time the composition of the radionuclides also changes dramatically as the shorter half-life radionuclides (e.g. $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$, $^{132}\text{Te}/^{132}\text{I}$, ^{131}I) decay and the longer half-life radionuclides (e.g. ^{137}Cs , ^{134}Cs , ^{144}Ce) persist. After 1 day, the three most abundant radionuclides are $^{99\text{m}}\text{Tc}$ (26%), ^{140}La (20%) and ^{99}Mo (15%), changing after 1 week to ^{140}La (15%), $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ (12%) and ^{132}I (12%). From 1 to 2 years, two of the three main radionuclides remain consistent as ^{106}Rh and $^{144}\text{Ce}/^{144}\text{Pr}$, with only changes in their relative amounts. This changes from 27% to 38% for ^{106}Rh , and from 24% to 27% for $^{144}\text{Ce}/^{144}\text{Pr}$. During this time, the third main radionuclide changes from ^{95}Nb (14%) at 1 year to ^{137}Cs (7%) at 2 years.

During the IFE, soil samples were collected from 12 November 2014 to 2 December 2014. This corresponds to 2 – 5 weeks after the hypothetical nuclear explosion. During this time, the initial vented activity would have decreased from 0.4% (2 weeks) to 0.1% (5 weeks), and the three most abundant radionuclides changed from ^{140}La (17%), ^{140}Ba (14%) and ^{131}I (10%) to ^{95}Nb (18%), ^{103}Ru (18%) and ^{141}Ce (14%). Notably at 2 weeks, there are three radionuclides

(^{99m}Tc, ¹³²I and ¹⁴⁴Pr) that are < 1% of the initial vented activity, and at 5 weeks there are five radionuclides (⁹⁹Mo, ^{99m}Tc, ¹³²Te, ¹³²I, ¹⁴⁴Pr). This represents a loss of greater than 99% by radioactive decay. As another example, there are six radionuclides (⁹⁹Mo, ^{99m}Tc, ¹³²Te, ¹³²I, ¹⁴⁰La, ¹⁴⁴Pr) that are < 10% of the initial vented activity at 2 weeks; and seven radionuclides (⁹⁹Mo, ^{99m}Tc, ¹³¹I, ¹³²Te, ¹³²I, ¹⁴⁰La, ¹⁴⁴Pr) at 5 weeks.

3.2. Deposited activity

The authors have calculated a surface deposition footprint that is complementary to the scenario of IFE14. In this calculation, the deposited activity of a ²³⁹Pu device with a 0.5 – 2.0 kT explosive yield and 1% venting was estimated using the HYSPLIT model for the IFE location (Fig. 3). Deposition was estimated for a 24 hour dispersion period using actual meteorological data immediately following the assumed/calculated time of the underground explosion. The bulk of the activity was deposited south of the assumed/calculated explosion location as a plume. Immediately at the nuclear explosion location there was 9.4×10^{-9} Bq m⁻² of the initial activity deposited (0.0000009%). Surface deposition of 1×10^{-9} Bq m⁻² (0.0000001%) of the initial vented activity extended up to 3.8 km from the nuclear explosion location and covered an area of approximately 7 km². For the purposes of evaluating soil and in-situ measurements this area was referred to as the detection zone. (Note: as actually implemented in the IFE14 scenario, the detection zone was much smaller [1].) Beyond this area were concentric areas of lower deposition, with 1×10^{-10} Bq m⁻² (0.00000001%) to 12 km (width 2.5 km), 1×10^{-11} Bq m⁻² (0.000000001%) to 35 km (width 12 km) and 1×10^{-12} Bq m⁻² (0.0000000001%) to 65 km (width 35 km).

The modelled deposition rate within the detection zone (1×10^{-9} Bq m⁻²) was applied to the released activity to provide concentration estimates during the inspection period (Fig. 4). During the period of 1 week to 2 years, the total activity of OSI relevant radionuclides decreased from 7.25×10^5 Bq m⁻² to 1.87×10^3 Bq m⁻². As with released activity calculations,

only ^{106}Rh , ^{137}Cs and $^{144}\text{Ce}/^{144}\text{Pr}$ remained after 2 years. During the IFE (2 – 5 weeks), the deposited activity decreased from $3.26 \times 10^5 \text{ Bq m}^{-2}$ to $1.23 \times 10^5 \text{ Bq m}^{-2}$ with significant loss of the shorter-lived $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ and $^{132}\text{Te}/^{132}\text{I}$ radionuclides.

3.3. Detection

Multiple gamma-spectrometry measurements were made of environmental soil samples and in-situ during the IFE. To simulate an actual release from the fictitious underground explosion, surrogate radionuclides were used instead of OSI relevant radionuclides. This included placing sealed $^{110\text{m}}\text{Ag}$ sources inside selected soil samples that reached the Inspection Team mobile laboratory, and burying sealed ^{60}Co sources in the inspection area for survey detection systems to find and characterize. Whilst the measurements performed searched for these surrogate radionuclides, they also provided information on the sensitivity of both techniques for the actual 17 relevant radionuclides of interest (Table 3). The average MDA was calculated based on typical measurements during the inspection. For soil samples, 31 measurements with typical count duration of 50,000 – 60,000 s were used to calculate average MDA values of 2.6 Bq m^{-2} to 189.6 Bq m^{-2} . The variation within these measurements was consistent with an average relative standard deviation of 38.5%. The average MDA of in-situ measurements was typically an order of magnitude higher and ranged from 26.2 Bq m^{-2} to 2000.9 Bq m^{-2} . This was based on 21 measurements with typical count duration of 1,800 – 3,600 s and average relative standard deviation of 52.2%. The reduced precision and sensitivity of in-situ measurements was attributed to uncontrolled detector backgrounds, variable detector solid angles, inhomogeneous source distribution and lower count durations [22-24].

Both techniques provided sufficient detection sensitivity for the initial measurement of all OSI relevant radionuclides for a nuclear explosion with a 2 kT yield. However, the increased sensitivity of laboratory-based gamma-spectrometry provided the opportunity to measure all

radionuclides up to 40 days. Seven radionuclides also remained detectable throughout a 2 year OSI period, including $^{95}\text{Zr}/^{95}\text{Nb}$, ^{106}Rh , ^{134}Cs , ^{137}Cs and $^{144}\text{Ce}/^{144}\text{Pr}$. This compared to in-situ measurements, where all OSI relevant radionuclides were detectable up to 31 days, and only four radionuclides were detectable throughout the 2 year period, including ^{106}Rh , ^{134}Cs , ^{137}Cs and ^{144}Ce . Radionuclides useful for dating a nuclear event ($^{95}\text{Zr}/^{95}\text{Nb}$ and $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$) remained detectable throughout their useful times since detonation for both techniques [25,26]. This was also possible for $^{134}\text{Cs}/^{137}\text{Cs}$ needed for differentiation of signals from nuclear reactors and explosions [5,6]. Both techniques provided sufficient sensitivity to allow detection of all OSI relevant radionuclides during the IFE timeframe of 2 – 5 weeks, although the deposited ^{99}Mo activity decayed below the average in-situ MDA after 31 days.

Detection of a ^{239}Pu explosion with a 0.5 kT yield was more problematic, and only laboratory measurements allowed for the initial measurement of all OSI relevant radionuclides. This was achievable up to 34 days. Only four radionuclides were detectable throughout a 2-year inspection period including ^{106}Rh , ^{134}Cs , ^{137}Cs and ^{144}Ce . In-situ gamma-spectrometry did not provide sufficient sensitivity for the initial measurement of ^{144}Pr ; and only ^{134}Cs and ^{137}Cs were detectable from 18 months to 2 years. Measurement of soil samples provided sufficient sensitivity for the majority of radionuclides during the IFE timeframe, although ^{99}Mo and $^{99\text{m}}\text{Tc}$ decayed below MDA after 30 and 34 days respectively. Both radionuclides fell below the MDA at 22 and 32 days for in-situ measurements, in addition to ^{144}Pr which was not detectable at any time.

3.4. Fractionation effects

The authors' complementary model has assumed a best case scenario where the effects of fractionation have not been included. In reality fractionation may typically alter the relative activities of refractory (R) and volatile (V) radionuclides in close proximity fallout by a factor of 3-4 compared with initial atmospheric concentrations [27,28]. This occurs as radionuclides

become chemically separated due to their condensation rates within the fireball, with high-boiling point refractory elements condensing at 1500 – 3000°C. The volatile radionuclides condense only as the cloud cools further and approaches ambient temperatures (~50°C). Further fractionation is attributable to physical separation due to the settling velocities of different sized particles. This causes larger particles containing a greater proportion of refractory radionuclides to be deposited close to the explosion [28]. Significant fractionation also occurs within the underground cavity and throughout the soil column [29]. The fractionation process is complex and not fully understood, with factors exceeding 100 being reported [27-32].

Applying a fractionation ratio of 0.37 [27] reduces the amounts of refractory relative to volatile radionuclides. This reflects the higher degree of containment expected for the less mobile refractory radionuclides. The effect is to reduce the amount of deposited activity and the days available for measurement. For soil samples the average reduction is 12 days and for in-situ is 16 days. Notably for in-situ measurements, including fractionation effects makes $^{144}\text{Ce}/^{144}\text{Pr}$ undetectable at any inspection time for a 0.5 kT yield explosion (only ^{144}Pr is undetectable for a 2 kT yield). Fractionation effects also reduce the time available to measure ^{99}Mo (30 days) and $^{99\text{m}}\text{Tc}$ (34 days) in soil samples for a 0.5 kT yield during the IFE timeframe. The days available for in-situ measurements of both radionuclides are also reduced for a 2 kT (27 and 33 days) and 0.5 kT yield (22 and 28 days).

4. Conclusions

Detection of the IFE simulated 0.5 – 2 kT underground explosion is dependent on a multitude of complex factors, including elapsed time, device yield, containment, radionuclide dispersion and deposition, sampling strategy and measurement. Selection of the correct measurement technique is essential to optimise the probability of detecting a Treaty violation. Using a

combination of model and IFE measurements, two of the permitted OSI techniques have been evaluated, including laboratory-based measurement of soil samples and in-situ gamma-spectrometry. The detection sensitivity for soil samples is typically an order of magnitude better than in-situ measurements, with MDA values of $2.6 - 189.6 \text{ Bq m}^{-2}$ compared to $26.2 - 2000.9 \text{ Bq m}^{-2}$. This provides greater opportunity for measuring all OSI relevant radionuclides within the IFE timeframe of 2 to 5 weeks and OSI timeframe of 1 week to 2 years. Only soil samples allow detection of all relevant radionuclides within these timeframes and when the effects of fractionation are considered. This contrasts to in-situ measurement where $^{144}\text{Ce}/^{144}\text{Pr}$ is not detectable at any time for a 0.5 kT yield with fractionation. This suggests laboratory-based gamma-spectrometry is the optimum technique for IFE and OSI timeframes.

5. Acknowledgements

The authors thank the IFE14 host country, the Hashemite Kingdom of Jordan, for making this data available, and the Provisional Technical Secretariat (PTS) for providing IFE data and support, and particularly Dr. Xavier Blanchard of the PTS staff for detailed explanations on how to interpret the in-situ reported values. Further, the authors thank the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and Real-time Environmental Applications and Display sYstem (READY) website (<http://www.ready.noaa.gov>) used in this research. The views expressed here do not necessarily reflect the opinion of the United States Government, the United States Department of Energy, or the Pacific Northwest National Laboratory. Pacific Northwest National Laboratory is operated for the U.S. Department of Energy by Battelle under Contract DE-AC05-76RL01830.

330 **6. References**
331

- 332 1. CTBTO (2015) Development and Implementation of the 2014 Integrated Field
333 Exercise Scenario. Comprehensive Nuclear-Test-Ban Treaty Organisation (Austria).
334 CTBT/PTS/TR/2015-1.
- 335 2. United Nations (1996) The Comprehensive Nuclear-Test-Ban Treaty. Resolution
336 CTBT/MSS/RES/1.
- 337 3. Hawkins W L (2011) Observables in Comprehensive Nuclear-Test-Ban Treaty On-
338 Site Inspections: An Illustrative Guide. Los Alamos National Laboratory (USA). LA-
339 UR-11-06276.
- 340 4. CTBTO (2012) Refined List of Refined List of Radionuclides of Interest for OSI.
341 Comprehensive Nuclear-Test-Ban Treaty Organisation (Austria). CTBT/WGB/TL-
342 4/42.
- 343 5. IAEA (2008) Handbook of Nuclear Data for Safeguards: Database Extensions.
344 International Atomic Energy Agency (Austria). INDC(NDS)-0534.
- 345 6. CTBTO (2000) Recommended Standard List of Relevant Radionuclides for IDC
346 Event Screening. Comprehensive Nuclear-Test-Ban Treaty Organisation (Austria).
347 CTBT/WGB/TL-2/40.
- 348 7. Burnett J L and Davies A V (2014) On-site inspection for the radionuclide
349 observables of an underground nuclear explosion. J. Radioanal. Nucl. Chem. 303:
350 2073-2079.
- 351 8. Chamberlain A C (2004) Radioactive Aerosols. Cambridge Environmental Chemistry
352 Series (UK).
- 353 9. England T R and Rider B F (1993) Evaluation and Compilation of Fission Product
354 Yields. Los Alamos National Laboratory (USA). LA-UR-94-3106, ENDF-349.
- 355 10. US Congress, Office of Technology Assessment (1989) The Containment of
356 Underground Nuclear Explosions. OTA-ISC-414 US Government Printing Office
357 Washington.

- 358 11. DOE (1996) Radiological Effluents Released From US Continental Tests 1961
359 through 1992. Department of Energy (United States). DOE/NV-317(Rev. 1).
- 360 12. Schery S D (2001) Understanding Radioactive Aerosols and Their Measurement.
361 Kluwer Academic Publishers (United States).
- 362 13. Draxler R R and Rolph G D (2014) HYSPLIT (HYbrid Single-Particle Lagrangian
363 Integrated Trajectory) Model access via NOAA ARL READY Website
364 (<http://ready.arl.noaa.gov/HYSPLIT.php>). NOAA Air Resources Laboratory (United
365 States).
- 366 14. Rolph G D (2014) Real-time Environmental Applications and Display sYstem
367 (READY) Website (<http://ready.arl.noaa.gov>). NOAA Air Resources Laboratory
368 (United States).
- 369 15. CTBTO (2014) Using Surface Soil Sampling Techniques. Comprehensive Nuclear-
370 Test-Ban Treaty Organisation (Austria). IFE-RNS-WIN-003.
- 371 16. CTBTO (2014) Working with Canberra Falcon. Comprehensive Nuclear-Test-Ban
372 Treaty Organisation (Austria). IFE-GAM-WIN-001.
- 373 17. Wogman N A, Sifert C E, Payne R, Bowyer T W, Milbrath B D, Friese J I, Miley H
374 S, Hanlen R C (2010) A Discussion of Factors Affecting CTBT OSI Environemntal
375 Sampling and Analysis. Pacific Northwest National Laboratory (United States):
376 PNNL-SA-72065.
- 377 18. Kato H, Onda, Y and Teramage M (2012) Depth distribution of ¹³⁷Cs, ¹³⁴Cs, and
378 ¹³¹I in soil profile after Fukushima Dai-ichi Nuclear Power Plant Accident. Journal
379 of Environmental Radioactivity 111: 59-64.
- 380 19. Burnett J L and Davies A V (2011) Investigating the time resolution of a compact
381 multidimensional gamma-spectrometer. J. Radioanal. Nucl. Chem. 288: 699-703.
- 382 20. Venkataraman R, Bronson F, Abashkevich V, Young B M and Field M (1999)
383 Validation of in-situ object counting system (ISOCS) mathematical efficiency
384 calibration software. Nuclear Instruments and Methods in Physics Research A 422(1-
385 3): 450-454.

- 386 21. Currie L A (1968) Limits for Qualitative and Quantitative Determination –
387 Application to Radiochemistry. *Analytical Chemistry* 40: 586-593.
- 388 22. Wimmer N G and Kreek S A (2014) OSI-GAM-SOP-00x In-situ High-resolution
389 Gamma Spectroscopy. Lawrence Livermore National Laboratory (USA): EP-
390 DRAFT-1.1 2014-01-19.
- 391 23. Kreek S A (2015) Lessons Learned & Observations on the 2014 Integrated Field
392 Exercise. Lawrence Livermore National Laboratory (United States): LLNL-TR-
393 666289.
- 394 24. Al-Masri M S and Doubal A W (2013) Validation of in-situ and laboratory gamma
395 spectrometry measurements for determination of ^{226}Ra , ^{40}K and ^{137}Cs in soil. *Applied*
396 *Radiation and Isotopes* 75: 50-57.
- 397 25. Harms A, Johansson L and MacMahon D (2009) Decay correction of ^{95}Nb . *Applied*
398 *Radiation and Isotopes* 67:641-642.
- 399 26. Nir-El Y (2004) Dating the age of a nuclear event by gamma spectrometry. *Applied*
400 *Radiation and Isotopes* 60(2-4): 197-201.
- 401 27. Trabalka J R and Kocher D C (2007) Bounding Analysis of Effects of Fractionation
402 of Radionuclides in Fallout on Estimation of Doses to Atomic Veterans. Defense
403 Threat Reduction Agency (USA): DTRA01-03-C-0064.
- 404 28. NRC (2003) A Review of the Dose Reconstruction Program of the Defense Threat
405 Reduction Agency. National Academies Press (United States). ISBN: 978-0-309-
406 08902-9.
- 407 29. OTA (1989) The Containment of Underground Nuclear Explosions. Office of
408 Technology Assessment (USA): OTA-ISC-414.
- 409 30. Hicks H G (1981) Results of Calculations of External Gamma Radiation Exposure
410 Rates from Fallout and the Related Radionuclide Compositions. Lawrence Livermore
411 National Laboratory (United States): UCRL-53152.
- 412 31. Hicks H G (1982) Calculation of the concentration of a radionuclide deposited on
413 the ground by off-site fallout from a nuclear detonation. *Health Physics* 42: 585-600.

32. Hicks H G (1990) Additional calculation of radionuclide production following nuclear explosions and Pu isotopic ratios for Nevada Test Site events. Health Physics 59: 515-524.

Table captions

Table 1. Fission product yields per 100 fissions for ^{239}Pu pooled fast neutron fission decay for OSI relevant radionuclides. Each radionuclide is categorised as volatile (V), refractory (R) or intermediate (V-R). Fission product yields are not given for ^{134}Cs as it is produced predominantly by neutron capture of ^{133}Cs .

Table 2. HYSPLIT model parameters.

Table 3. Minimum detectable activity (MDA) for soil samples recalculated by the authors from IFE14 sample spectra and in-situ gamma-spectrometry measurements of the Inspection Team and time available before the deposited activity has decayed below the MDA. Calculations have been made for a ^{239}Pu device with a 2 kT and 0.5 kT yield. Radionuclides that are detectable throughout the 2 year OSI period are denoted by (>) and undetectable by (--). Values given in parenthesis are for refractory radionuclides when a fractionation ratio is applied.

Figure captions

Fig. 1. Escape paths of radioactive material to the surface (not to scale). Modified after [1].

Fig. 2. The relatively proportions of OSI relevant radionuclides during the OSI timeframe. The vented activity (%) is shown by the dashed line.

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443 Fig. 3. Deposited activity released from a ^{239}Pu device with a 0.5 - 2 kT explosive yield and
444 1% venting for the IFE scenario. Values given are fractional amounts of the initial vented
445 activity.

446

447 Fig. 4. Deposited activity released from a ^{239}Pu device with a 0.5 - 2 kT explosive yield and
448 1% venting for the IFE scenario.

449

Figure 1

Figure 2

Figure 3

Figure 4

Table 1

Nuclide	Half-life (Days)	Energy (keV)	Abundance (%)	Ind. Yield	Cum. Yield	Volatile or Refractory
⁹⁵ Zr	64.03	756.7	54.4	9.30×10^{-2}	4.67	R
⁹⁵ Nb	34.99	765.8	99.8	1.92×10^{-4}	4.67	R
⁹⁹ Mo	2.75	739.5	12.1	1.31×10^{-2}	5.98	R
^{99m} Tc	0.25	140.5	89.1	1.49×10^{-6}	5.26	R
¹⁰³ Ru	39.26	497.1	91.0	1.29×10^{-3}	6.83	V
¹⁰⁶ Rh	373.61	621.9	9.9	3.85×10^{-1}	4.36	V
¹³² Te	3.20	228.2	88.0	3.00	3.88	V
¹³¹ I	8.02	364.5	81.7	2.01×10^{-2}	5.15	V
¹³² I	0.10	667.7	98.7	1.74×10^{-1}	5.33	V
¹³⁴ Cs	754.17	604.7	97.6	Activation	Activation	V
¹³⁷ Cs	10972.22	661.7	85.1	1.00	6.58	V
¹⁴⁰ Ba	12.75	537.3	24.4	9.17×10^{-1}	5.32	V-R
¹⁴⁰ La	1.68	1596.2	95.4	9.87×10^{-3}	5.33	V-R
¹⁴¹ Ce	32.51	145.4	48.3	2.46×10^{-4}	5.15	V-R
¹⁴⁴ Ce	284.95	133.5	11.1	1.64×10^{-1}	3.67	R
¹⁴⁴ Pr	0.01	696.5	1.3	5.10×10^{-5}	3.67	R
¹⁴⁷ Nd	10.98	91.1	27.9	1.72×10^{-3}	1.99	R

Table 2

Parameter	Value	Comments
Release top	50 m	Top height of the pollutant source
Release bottom	0 m	Bottom height of the pollutant source
Release duration	10 min	Duration of venting
Runtime duration	24 hours	Duration of model calculation
Dry deposition velocity	0.001 ms ⁻¹	

Table 3

Nuclide	Soils (Bq m ⁻²)	r.s.d. (%)	2 kT Day	0.5 kT Day	In situ (Bq m ⁻²)	r.s.d. (%)	2 kT Day	0.5 kT Day
⁹⁵ Zr	4.7	32.5	> (666)	629 (537)	50.1	54.6	539 (447)	411 (319)
⁹⁵ Nb	3.5	39.3	>	730 (638)	28.6	55.5	663 (572)	535 (443)
⁹⁹ Mo	26.5	41.3	40 (36)	34 (30)	226.4	55.0	31 (27)	26 (22)
^{99m} Tc	9.9	40.3	43 (39)	38 (34)	44.7	40.8	37 (33)	32 (28)
¹⁰³ Ru	3.0	34.1	539	461	32.6	59.2	404	325
¹⁰⁶ Rh	24.9	31.9	>	>	285.1	60.9	>	468
¹³¹ I	3.6	35.0	120	104	34.0	42.8	94	78
¹³² Te	4.3	40.5	53	47	35.6	58.7	43	37
¹³² I	3.2	37.4	55	48	28.7	56.1	44	38
¹³⁴ Cs	3.4	42.9	>	>	31.2	57.6	>	>
¹³⁷ Cs	3.3	44.7	>	>	33.2	54.8	>	>
¹⁴⁰ Ba	11.4	33.3	167	142	117.0	58.6	124	99
¹⁴⁰ La	2.6	29.5	197	171	26.2	52.0	154	129
¹⁴¹ Ce	7.2	43.9	401	336	78.6	41.6	289	224
¹⁴⁴ Ce	31.9	44.4	>	>	356.6	41.3	> (467)	306 (--)
¹⁴⁴ Pr	189.6	32.4	> (726)	565 (157)	2000.9	55.9	167 (--)	-- (--)
¹⁴⁷ Nd	17.5	51.6	124 (108)	102 (86)	188.4	42.2	86 (70)	64 (49)