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Comparison of radionuclide ratios in atmospheric nuclear explosions and nuclear releases from Chernobyl and Fukushima seen in gamma ray spectrometry

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Abstract The Comprehensive Nuclear Test Ban Treaty has remote radionuclide monitoring followed by an On Site Inspection (OSI) to clarify the nature of a suspect event as part of its verification regime. An important aspect of radionuclide measurements on site is the discrimination of other potential sources of similar radionuclides such as reactor accidents or medical isotope production. The Chernobyl and Fukushima nuclear reactor disasters offer two different reactor source term environmental inputs that can be compared against historical measurements of nuclear explosions. The comparison of whole-sample gamma spectrometry measurements from these three events and the analysis of similarities and differences are presented. This analysis is a step toward confirming what is needed for measurements during an OSI under the auspices of the Comprehensive Test Ban Treaty.

Keywords CTBT · Chernobyl · Fukushima · Radionuclide monitoring · Gamma spectroscopy

Introduction

The use of environmental monitoring of radionuclides as an indication of nuclear accidents and explosions has a history as long as the nuclear age itself. One aspect of such monitoring for the Comprehensive Nuclear Test Ban Treaty (CTBT) verification regime is On Site Inspection (OSI). In an OSI, besides visual observation, seismological investigations, and radiological surveys, a variety of environmental sample types can be collected and analyzed by gamma spectrometry. Sample types may include soil, vegetation, water, aerosol air samples, and whole air samples processed to allow detection of noble gasses. The goal of this sampling and analysis is to determine if a nuclear explosion has occurred, sometimes referred to as the 'smoking gun' of the inspection. However, there are many mechanisms through which radionuclides can enter the environment besides nuclear tests, and a method is needed to discriminate between nuclear explosions and these other sources. One such source is a reactor accident. Two large reactor events over the last quarter-century injected large amounts of radionuclides into the environment; Chernobyl in 1986 and Fukushima in 2011.

These reactor events had very different mechanisms for injecting radionuclides into the environment. During the Chernobyl accident, the entire reactor core exploded and breached its primary containment, whereas during Fukushima the radionuclides were released through the coolant system. The difference in release mechanism has an impact on the radionuclides that were released. The radionuclides released from the Fukushima accident were primarily volatile in nature, whereas the list of fission products released during Chernobyl was quite broad. In principle, a nuclear explosion could also have different mechanisms for releasing radionuclides into the environment. An atmospheric or surface explosion will typically inject the entire fission product inventory into the environment; in contrast, an underground explosion may only leak gases and volatile elements.

Methods

Environmental samples collected against these nuclear reactor accidents and nuclear explosions can be compared to

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determine which radionuclides are likely useful to discriminate between types of events. There is a vast literature resource from the Chernobyl accident and historical nuclear explosions, and a growing data set for Fukushima releases. However, the goal is not to cover the all available data sets, but to demonstrate which isotopes that are likely to be measureable in the environment have discriminating power for an OSI. There will always be exceptions to any conclusions based on a particular test scenario because each scenario will have differences. A subset of data from Chernobyl [1, 2], Fukushima [3], US nuclear test Buggy [4] and Chinese test #19 [5] will be compared to illustrate isotopic ratios from real-world examples. The Chernobyl data used is from references [1] and [2] and is comprised of gamma spectrometry measurements on single hot particles collected in Sweden. This data set was chosen because hot particles are likely to have minimal environmental contributions and a higher degree of scatter in the isotopic data because the measurement is not an average over many particles. The Fukushima data used was published by the National Nuclear Security Administration (NNSA) [3]. The data chosen consisted of gamma spectrometry collected from air filters. The data was further selected to include only filters on which ^{140}Ba was detected because when this isotope was detected, many other radionuclides were also present; this presented a method of reducing the volume of data.

Two nuclear explosions were chosen as example data sets: the Buggy test in 1968 at the Nevada test site [4] and the 19th Chinese nuclear test in 1976 [5]. The Buggy event was part of the Plowshare test series in which the intent was to move soil, for a so-called Peaceful Nuclear Explosion (PNE). This event was nominally underground, a trench cratering experiment, but by design broke through the surface in a shower of soil and dust. It dispersed more radioactive material than a gas-only leak from an underground nuclear explosion, but not as much as an atmospheric explosion, because the falling soil could intercept the cooling nuclear debris and partially clean it from the atmosphere. The 19th Chinese nuclear test was an atmospheric explosion we chose for contrast.

All data from the nuclear tests are high-volume air collections unless otherwise noted. All ratios are activity ratios decay-corrected to the event time using the best nuclear data available at the time. In some cases, detection limits are used where available. Data from particle size fractions were measured from the Buggy and Chinese explosions and this data was selection for reasons similar to the Chernobyl data from hot particles.

Results and discussion

Table 1 lists all of the radionuclides detected in the selected data sets. In general, the trend in the various events is as expected. Chernobyl's releases exhibited the entire radionuclide inventory of both refractory (^{95}Zr, ^{144}Ce) and volatile (^{132}Te, ^{131}I, Cs isotopes) elements, whereas Fukushima's atmospheric releases that transported long distances were essentially only volatile elements. The radionuclides measured from the Buggy test have lower concentrations of refractory elements due to its emplacement in contrast to the atmospheric test, which has both volatile and refractory elements.

Table 1 Radionuclides detected in selected Chernobyl, Fukushima and nuclear explosions

Isotope	Chernobyl	Fukushima	Buggy	Chinese #19
^{95}Zr	X			X
^{95}Nb	X			X
^{99}Mo	X		X	X
^{103}Ru	X		X	X
^{106}Ru	X		X	
^{132}Te/^{132}I	X	X	X	X
^{129m}Te		X		
^{131}I	X	X	X	X
^{134}Cs	X	X		
^{136}Cs	X	X		
^{137}Cs	X	X	X	X
^{140}Ba	X	X	X	X
^{141}Ce	X		X	X
^{144}Ce	X			X

The activity ratio of one radionuclide to another is the best way to discriminate between various scenarios. In general the measured ratio will be a function of two processes: the production mechanism and the environmental chemistry of the elements. The production mechanism gives rise to some differences mostly due to cumulative fission yield production in a reactor compared to the fission yield produced in just an instant in an explosion. All of the data used in this study came from environmental collections, therefore the elements interacted with the environment and fractionated due to differences in elemental chemistry. For this reason, the most robust ratios are of isotopes of an element or radionuclides of elements with similar chemistry. These ratios will minimize the variation in ratios due to environmental chemistry interactions and uniquely identify the production mechanism. Large scatter in the data should be expected when ratios are made with two elements with significantly different chemistry.

Figure 1 shows the ratio of ^{131}I to ^{132}Te. These two elements are volatile in nature, but their environmental chemistry is very different, giving rise to about two orders of magnitude difference in the ratio. In addition, the data suggests that there is little difference between reactor incidents and nuclear explosions. This indicates that if

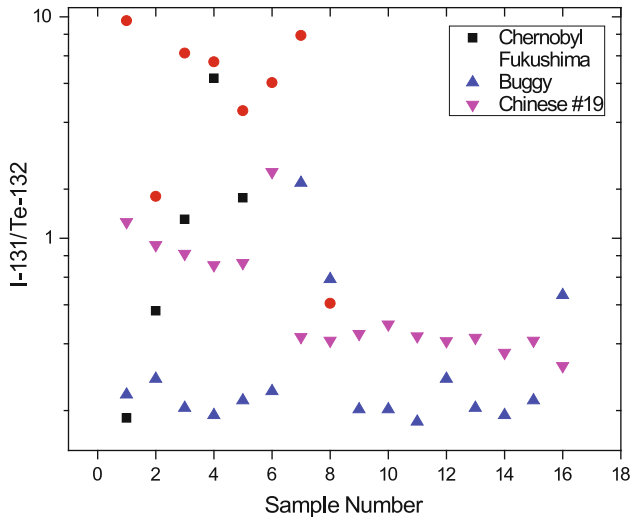

Fig. 1 Iodine-131 and ^{132}Te isotopic ratio for the four scenarios

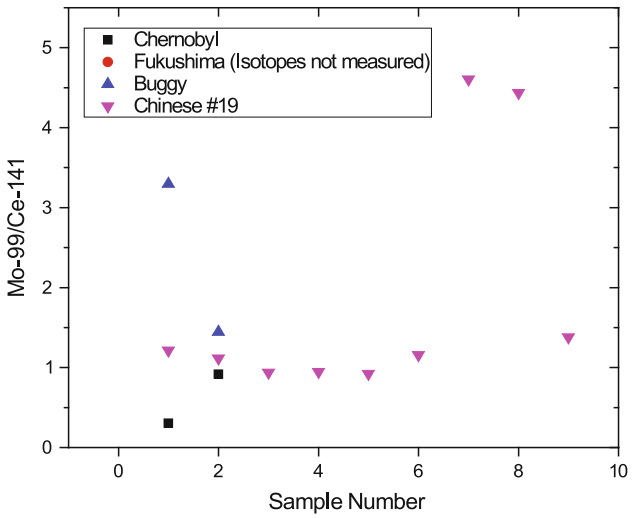

Fig. 3 Molybdenum-99 to ^{141}Ce isotopic ratios for the four scenarios

iodine and tellurium were to be measured in an environmental sample, it would not be possible to discriminate between scenarios. Figure 2 illustrates a similar situation with ^{141}Ce and ^{140}Ba; this ratio also shows variation of over two orders of magnitude. Cerium-141 was not measured in the Fukushima incident because it is refractory in nature and was not released in significant quantities.

Figure 3 plots two refractory elements together: molybdenum and cerium. The variation in the ratio is significantly less than seen in the volatile elements, with a range of about five. This indicates that these elements interacted similarly with the environment, as opposed to the more volatile elements represented in Figs. 1 and 2. However, despite their similar chemistry, there still is little difference between Chernobyl and nuclear explosions in this limited data set.

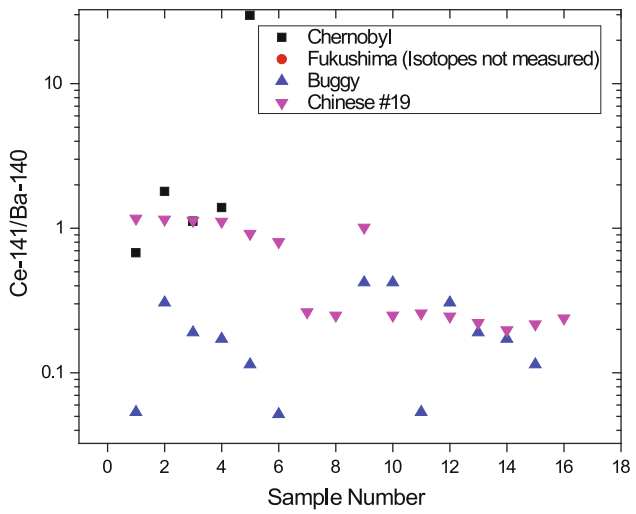

Fig. 2 Ceruim-141 to ^{140}Ba isotopic ratios for the four scenarios

Figure 4a, b show isotopic ratios within the same elements (cerium and ruthenium). The environmental chemistry of these isotopes should be nearly identical. Cerium is a refractory element, thus it was only observed in significant quantities in Chernobyl and the atmospheric explosion. However, even though the environmental chemistry is similar, the measured ratio is similar for both events. In contract, the isotopic ratios for the ruthenium isotopes show a small divergence between reactors and explosion. The ratio measured from the Buggy event is smaller than that measured from Chernobyl. This difference is likely due to the difference in the production mechanism for the two ruthenium isotopes. In a reactor like Chernobyl, the longer half-life of ^{103}Ru and ^{106}Ru isotopes allows the ratio to build up over time, thus when the ruthenium is injected into the environment, the ratio should be larger than instantaneous production from a nuclear explosion. Figure 4b shows this effect, but it is a small difference and a difficult measurement to make via gamma spectrometry. A larger data set over many different explosions is needed to quantify how this ratio could or could not be used for discrimination, but the principle is established.

Figure 5a, b show the cesium isotopic ratios. Like ruthenium, the environmental chemistry of the three cesium isotopes will be nearly identical, thus minimizing the perturbation of their ratios by environmental effects. As observed in Fig. 5, the ratios observed in the reactor releases are different from those seen in nuclear explosions. The difference is due to their respective production mechanisms; the effect from chemistry is minimized. In Fig. 5 one sees that ^{134}Cs and ^{136}Cs are not found in nuclear explosion data, so the minimum detectable activity (MDA) is used in the figures. In nuclear explosions, the production of ^{134}Cs and ^{136}Cs are shielded by stable precursors and independent production only; however, in reactors, these

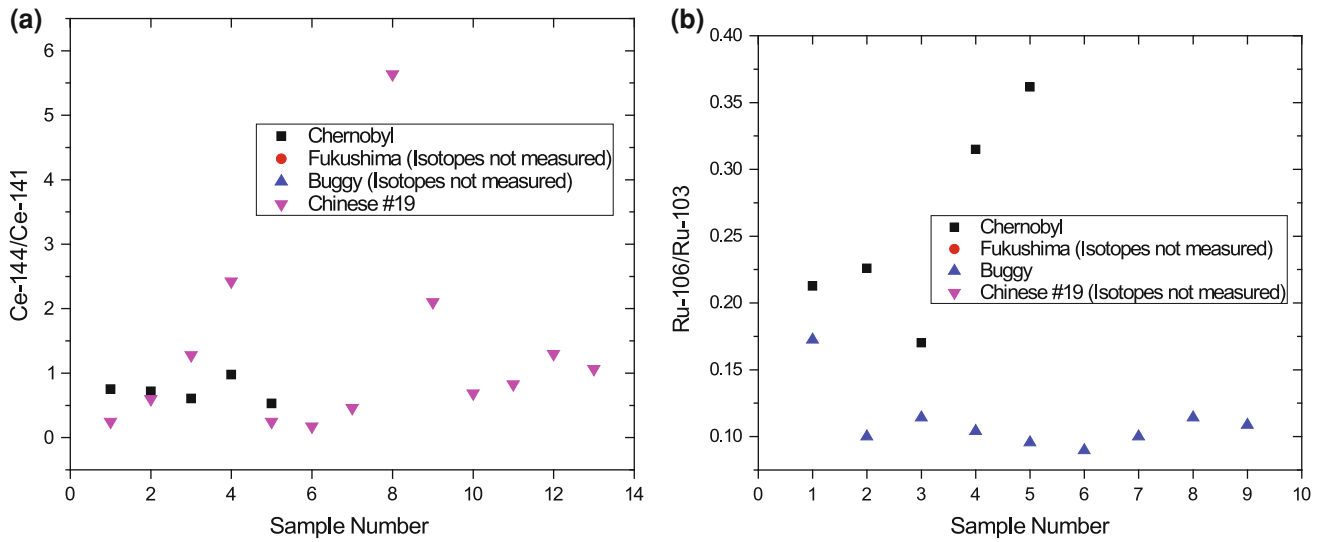

Fig. 4 **a** Cerium-144 to ^{141}Ce; **b** ^{106}Ru to ^{103}Ru isotopic ratios for the four scenarios

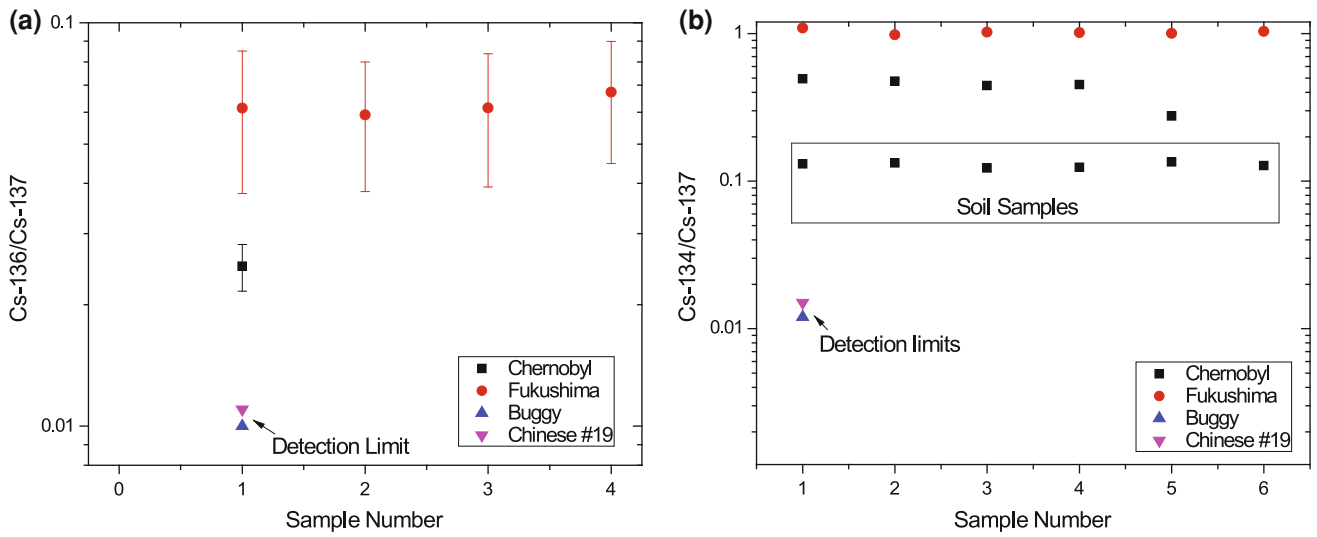

Fig. 5 **a** Cesium-136 to ^{137}Cs; **b** cesium-134 to ^{137}Cs isotopic ratios for the four scenarios

isotopes build up from activation of ^{133}Cs and ^{135}Cs. The result of this difference is an order of magnitude difference in the cesium ratios such that ^{134}Cs and ^{136}Cs are likely to be detected in reactor environmental releases, but unlikely to be observed in nuclear explosions by gamma spectrometry. The longer half-life of ^{134}Cs makes it of greater value to an OSI than ^{136}Cs, with both isotopes being equal in their ability to provide differentiation.

Conclusion

Gamma spectrometry applied to environmental samples collected against two nuclear reactor releases and two nuclear explosions shows similarities and differences in their resulting isotopic ratios. The use of inter-element volatile elements such as iodine and tellurium proves not to be useful for discrimination between reactor and explosion source terms. However, they are likely still useful in an OSI in that they could flag a location for further inspection. The use of refractory inter-elements ratios such as molybdenum and cerium have less scatter in the data; however, the differences between the source terms for isotopes of these elements are smaller than scatter in the data due to environmental chemistry effects. Intra-element ratios are more consistent because isotopic fractionation doesn't occur in the environment. Intra-element ratios and the use of isotopes that build up in reactors such as ^{134}Cs will be the best source term discriminators. However, a conclusive study would require larger data sets and the inclusion of more scenarios.

Some of the fission products that are in particulate form (not gases) that may have these characteristics are ^{94}Nb and ^{95}Nb, ^{103}Ru and ^{106}Ru, ^{129m}Te and ^{132}Te, ^{129}I and ^{131}I, ^{134}Cs and ^{137}Cs, ^{154}Eu and ^{155}Eu. Of these sets of fission products, only ^{103}Ru, ^{106}Ru, ^{134}Cs, ^{137}Cs were observed in this study in both reactor releases and nuclear explosions. The other isotope pairs were not easily measured by gamma spectrometry due to long half-lives or poor nuclear decay schemes. The ruthenium isotopes do show a slight differentiation between reactors and nuclear explosion, but the difference is small and a large number of nuclear explosions would need to be considered to identify whether this isotope pair is robust enough to be reliable for use in an OSI. However, the cesium isotope ratios show sufficiently large differentiation that even the detection limits were useful in discrimination. In general, more data for each type of scenario, and the addition of more scenarios, such as medical isotope production, will be important in the evaluation of this technique's robustness.

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