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The potential detection of low-level aerosol isotopes from new civilian nuclear processes

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A B S T R A C T

As the world faces a challenging future in maintaining the commercial availability of radioactive isotopes for medical use, new methods of medical isotope production are being pursued. Many of these are small in size and could effectively operate continuously. With the potential for much shorter retention times, a new suite of isotopes may soon be found in the environment. The authors estimate that many more aerosols containing low-level isotopes of gas/volatile origin could be detectable at short range and times, and a few at longer ranges and times as compared to those released in more common nuclear reactor operations.

1. Introduction

Environmental monitoring is done for many reasons: health and safety of populations, regulatory monitoring of industrial processes, evaluation of foodstuffs, and scientific research. It is expected to find contributions of natural radioactivity as well as certain legacy anthropogenic isotopes, for instance from Chernobyl and Fukushima disasters (Biegalski et al., 2012; Devell et al., 1986). Expert analysts have developed refined expectations for these anthropogenic isotopes, and use them regularly in earth science and biology. However, a new class of nuclear process has been proposed which could release a new suite of isotopes into the atmosphere. Where the source is sufficiently close or the measurement sufficiently sensitive, these isotopes could confound analysis or could prove to be useful for scientific research.

There is more than one new method of producing medical isotopes under study at present, but several of them share significant design features: a modest-sized mass of water containing a dissolved uranium compound in which batch or continuous mode irradiation with neutrons occurs (Piefer et al., 2011; Pillai et al., 2013). This is very different from current production methods using reactor irradiation of aluminum-clad low enriched uranium targets (Saey et al., 2010). As a result of the time delay for volatile species to escape through compromised fuel cladding and then out of large nuclear reactor vessels, escaping gas will experience a retention time that is long compared to the half-life of many short-lived volatile radioisotopes. Following target irradiation, short-lived fission products are allowed to decay for a cooling period before the uranium is dissolved in a hot-cell, at which point noble gases and other fission products can be released and further processed (Salacz, 1989). These time delays generally inhibit the large fractional release of short-lived volatile radionuclides when compared to potential short-time releases possible for some small alternate medical isotope production processes being considered. The present study considers the possible signatures resulting from an accidental release due to failure of containment during or shortly after neutron irradiation of an aqueous uranium solution. For the purposes of this work, the authors assume the retention time of escaping species to be 1 second for a small novel production method and 3 hours for a more traditional reactor containment vessel.

As novel isotope production methods would remain cool, with no hot fuel (perhaps not even boiling temperature), it is arguable that any leakage from these operations would be quite different than, say, the preponderance of the aerosol radionuclides (cesium, tellurium, and iodine) released at high temperature and while volatile from the Fukushima accident. The authors examine the list of detectable aerosol isotopes that have gaseous or highly volatile precursors, including very short-lived precursors, and compare simple retention time cases for novel medical isotope production and nuclear reactor-based production. Minimum detectable activities for these isotopes are computed for gamma detection in underground detectors.

2. Release estimate

Comparison is made of two generic, representative nuclear processes. The first model is that of a low-enriched uranium fueled nuclear

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research reactor operating at 20 MW. Retention time in such a reactor is difficult to assess in a general way – different leak locations and magnitudes within containment can obviously result in vastly different gas transport pathways and rates. For purposes of this work, it is assumed that the leak is small and has little net effect on the reactor operating dynamics. The average retention time of leaking gas in this scenario is assumed to be about 3 hours before gas escape. In the second case, a subcritical loading of distributed low-enriched uranium is irradiated by a constant thermal neutron flux of 10^{14} cm^{-2} s^{-1}. This generic scenario is representative of a number of the alternative small and subcritical isotope generating methods being pursued as potential future medical isotope production pathways (Pillai et al., 2013). As a much smaller vessel, retention time in this case is assumed to be 1 second in a hypothetical accidental leakage. These two medical isotope production cases will serve as the basis for signature comparison.

The Fortran isotopic production and depletion code Origen 2.2 was used to calculate radionuclide inventories for a simple release scenario from each of the two cases loaded with approximately 20 kg of 19% enriched uranium (Croff, 1983). To provide the largest general relevance, it was chosen to model each case leak as occurring in the middle of what can be considered normal steady state operation, significantly removed from recent startup. In the case of a classic reactor, fuel burnup was simulated for 20 days at 20 MWt, and then the isotopic inventory was allowed to cool 3 hours before leakage. In the novel isotope production case, a constant 10^{14} thermal flux irradiation was simulated for 7 days before a 1-second-delayed release. While examination of possible leakage mechanisms is beyond the scope of this work, as a way of normalizing the two scenarios, leakage fractions were chosen so that each case corresponds to roughly equal estimated ^{133}Xe atmospheric concentration at one day, assuming an initial uniform distribution throughout the respective reactor vessel volumes of roughly 0.1 m^3 and 100 m^3 for the novel and classic cases Leakage fractions of 0.1% and 10^{-8} were assumed for the novel isotope production case and the classic reactor case, respectively. Release fraction chosen here is inherently directly correlated with whether estimated leakages fall above or below MDAs.

Upon simulated leakage, only gaseous bromine, krypton, iodine and xenon fractions were carried through the process and allowed to evolve naturally through radioactive decay (Table 1). The activity of each radioisotope was then multiplied by a dilution factor (D_f) to estimate the effects of atmospheric transport. The atmospheric transport dilution was estimated as a function of time using a power law relation

$$D_f = 4.62 \times 10^{-11} \times T^{-2.19}$$

describing median atmospheric dilution (m^{-3}) where T is time in hours after release (Eslinger et al., 2015). Using the leakage fractions discussed previously, dilution estimates yield roughly 1×10^{-5} Bq/m^3 of ^{133}Xe in both scenarios after 24 h of atmospheric transport, at a median distance of roughly 600 km. For longer times in the atmosphere, this dilution can be a much greater factor than radioactive decay.

3. Detection systems

The volatile noble gases and halogens released from an aqueous fission solution vessel decay to produce a range of isotopes suitable for environmental measurement. Airborne particulates can be collected using high-volume air sampling techniques with typical sampling rates of 1000 m^3 h^{-1} (Bowyer et al., 1997; Miley et al., 1998), and are useful for providing integrated samples with a minimized natural background (Rosenberg et al., 1999). After collection, the filters containing the extracted particulate material are compacted into an optimized geometry for measurement (halogens can also be trapped using activated materials such as charcoal). Deposited activity can be collected using a surface-soil sampling technique (Baciak et al., 2012; CTBTO, 2014), which produces a sample (e.g. from 1 m^2) with minimum matrix interferences from the undisturbed deeper soil. Vegetation, including lichens and mosses (e.g. 0.5–1 kg), can also be sampled and ashed for integrated signals as well as water samples, including aquatic particulates, ionic species, and colloids (Wogman, 2013).

Multidimensional gamma-spectrometry is a powerful technique for radionuclide detection (Burnett and Davies, 2011; Keillor et al., 2009). A high-sensitivity system is being developed using two Broad Energy Germanium (BEGe) detectors (model BE5030 from Canberra Industries, Inc.) aligned in an up-down configuration. These high-performance BEGe detectors combine the spectral advantages of low energy and coaxial detectors, with an energy range from 3 keV to 3 MeV and optimized energy resolution and efficiency. The detectors are situated within a 20 cm thick low-background lead cave equipped with a cosmic veto system that reduces background by 34% (Burnett and Davies, 2014; Burnett et al., 2013). Background contributions are further reduced by locating the system within the Class 1000 cleanroom of the shallow underground laboratory at PNNL (Aalseth et al., 2012). The laboratory has a calculated 30 m of water equivalent (mwe), which results in approximately 100 times fewer fast neutrons and 6 times fewer muons.

To evaluate detection sensitivity, the Minimum Detectable Activity (MDA) was calculated using the Currie formula (Currie, 1968) with a region of interest (ROI) of 1.25 channels Full Width Half Maximum (FWHM) around the center of the hypothetical peak. Analysis was performed using the Canberra Genie Gamma Acquisition and Analysis software (version 3.3), which was also used to normalize and combine the spectra from each BEGE detector (the normal and split REXX commands). MDA values were calculated for count times ranging from 3600 s to 800,000 s using the start of acquisition as the reference time. Optimum values were selected according to the half-life of the radionuclide of interest (Table 2).

4. Results

The estimated activity concentration levels of the isotopes of interest with atmospheric transport are shown for the two release cases in Table 3. Those levels that are above the respective estimated MDCs are indicated in the table. The inhibiting effect of the 3-hour coolant retention time in the traditional nuclear reactor production case is clear – only ^{88}Rb is estimated in this release scenario to remain detectable while all of the others have substantially decayed away by the time leakage occurs. In the case of the generic novel production leakage scenario, many of the isotopes of interest are either estimated to be detectable or nearly detectable. Fig. 1 illustrates the results visually by showing a bar chart summarizing the estimated detectable time windows for each isotope of interest for the two cases. While there are dramatically more isotopes present shortly after the release in the novel case, after an extended period of time (several hours), both facilities see the activity of the aerosols drop below the estimated detection limits of the detection systems. Especially for shorter-lived species, the time taken to sample and measure each isotope becomes an important consideration and might require collection/measurement optimization to allow detection.

The assumed leakage fraction of each facility is a difficult parameter to constrain and one that has a direct correlation with the results of the estimated activity levels that could result in the environment. The assumed leakage fraction of the much smaller, novel isotope production case is five orders of magnitude greater than that of the traditional neutron-generating reactor. The authors chose this so that the signatures can be compared more directly. This says little about the realistic probability of each release scenario happening. Such an analysis would require much closer examination of specific facility and operations details, which in turn would necessitate a greater attention to details of the isotopic source model, and, along with a variety of starting