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Detection of Radioactive Isotopes from the Fukushima Daiichi Nuclear Power Plant in the Kittitas Valley of Washington State

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Abstract

Following the Fukushima Daiichi nuclear power plant disaster in March, 2011, various radioactive isotopes were emitted from the plant [Ewing, 2011]. We hypothesized that emissions would bind to particulate matter, which then could be dispersed by wind currents and deposited across the planet. We analyzed an aerosol sample collected with a high volume cascade impactor in the Kittitas Valley of Washington state. NaI(Tl) gamma spectrometry revealed the presence of the isotopes ^{132}Te and ^{131}I, consistent with the type of nuclear accident at Fukushima Daiichi.

Keywords

Acknowledgements

INTRODUCTION

In a typical nuclear power plant, controlled nuclear reactions of Uranium (^{235}U) are used to heat water to produce energy by adiabatically expanding steam to do work on generators that produce electricity. On March 11, 2011, an earthquake struck Japan that caused three of the six Fukushima Daiichi reactors to stop the nuclear chain reaction by insertion of control rods. The other three reactors were already shut down for inspection and maintenance [Ewing, 2011]. The design of Fukushima Daiichi was inadequate for the subsequent tsunami, damaging critical systems of the nuclear reactors. The tsunami cut off electricity to backup pumps and when diesel fuel was exhausted, battery power fueled the pumps [Ewing, 2011]. When these batteries died, a catastrophic sequence of events followed.

The loss of power to maintain adequate flows of cooling water to a nuclear reactor and spent fuel rods can have a number of serious consequences that could result in a release of radioactive isotopes, including damage to fuel rods and the reactor core from overheating, chemical reactions that liberate explosive hydrogen gas, and ignition of the zirconium metal alloy that clads the fuel rods [Ewing, 2011]. Fukushima Daiichi experienced at least the first two of these scenarios at various sites and reactors at the plant [Ewing, 2011].

From the well-known fission products of ^{235}U and reports of previous reactor accidents, notably Chernobyl, we identified radioactive isotopes that would be likely to be released into the atmosphere in a nuclear accident of the kind that occurred at Fukushima Daiichi. These isotopes include ^{131}I, ^{132}Te, ^{137}Cs, ^{140}Ba, and ^{141}La [Nuclear Energy Agency, 2002]. The goal of this work was to determine whether radioisotopes from the Fukushima Daiichi accident could be detected in ambient aerosol samples from the Kittitas Valley of Washington state.

METHODS AND MATERIALS

Following the Fukushima Daiichi accident on March 11, a high volume cascade impactor (HVCI) located on top of Dean Hall at Central Washington University in the Kittitas Valley of Washington state was set up to collect ambient aerosols from March 16-23, 2011 [Demokritou, 2001]. During this sampling period approximately 6.9×10^6 liters of air passed through the HVCI. NOAA Hysplit data for this period show wind currents at various altitudes travelled and possibly transported particulate matter between Fukushima Daiichi and the Kittitas Valley of Washington state [NOAA, 2011]. The trajectories at various altitudes are shown in Figure 1.

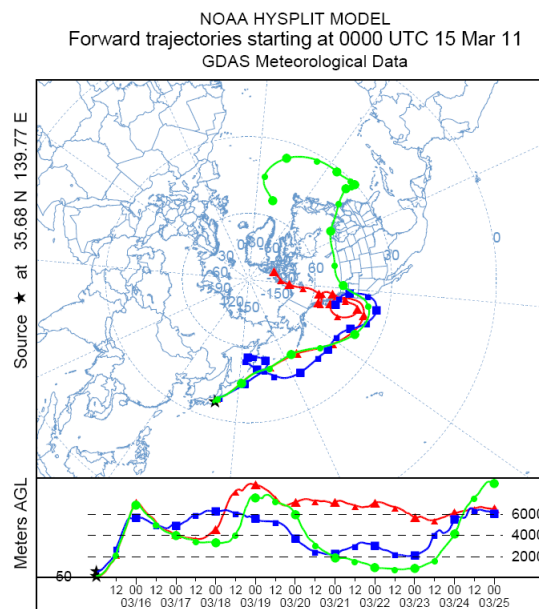The HVCI pumps air from the atmosphere through a system of 4

Figure 1. This map and graph illustrate the trajectory of air mass at various altitudes while the HVCI was collecting the sample. Each point is a 12 hr increment.

acceleration platforms to collect aerosols in 4 size fractions. One of these collection platforms is shown schematically in Figure 2. As the sample moves from stage to stage, the filters collect finer and finer material. On March 23, the fine and ultrafine sampling substrates, which include all particles with aerodynamic diameter of < 1 μm, was removed from the collector, placed in a sealed bag, and subsequently analyzed with gamma spectrometry. The sample is designated CWU-08411-f-uf.
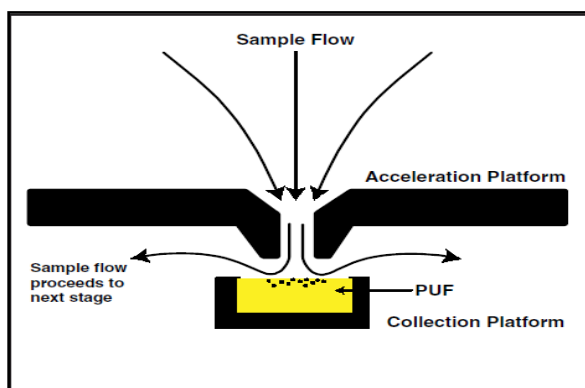

Figure 2. Schematic of a single filter at one stage in the HVCI.

Multiple gamma spectrometry runs were performed on sample CWU-08411-f-uf over a ten day period using a typical NaI(Tl) scintillation gamma spectrometer arrangement, shown schematically in Figure 3.
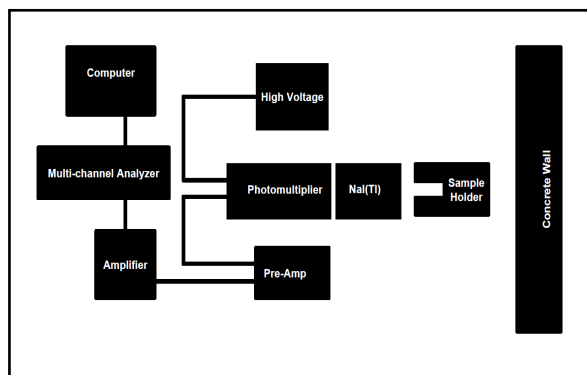

Figure 3. Schematic of the apparatus used to perform the gamma spectrometry measurements for HVCI sample CWU-08411-f-uf.

The spectrometer utilizes a 2 inch NaI(Tl) crystal for detecting and measuring the energy of gamma rays. We estimated the detector efficiency for different energy gammas and the configuration used for sample CWU-08411-f-uf based on a measured efficiency of 0.5 % for the 662 keV photopeak of a 1 μCi, ^{137}Cs source placed in a similar configuration and the energy dependence of the photoabsorption coefficient of NaI(Tl) [Knoll, 1979]. Isotopes (gamma energies) used to calibrate the spectrometer energy scale were ^{137}Cs (662 keV), ^{54}Mn (835 keV), ^{22}Na (511 keV), and ^{133}Ba (81 keV) [Lederer, 1978].

Sample CWU-08411-f-uf was marked for reproducible placement at the detector, and the plastic bag was placed in contact with the detector face for spectrometer runs. The spectrometer gain was adjusted so that the full scale MCA channel (1024) yielded E_γ=967 keV. Gamma spectrum runs on sample CWU-08411-f-uf ranged in duration from 12 to 45 hours. The first gamma spectrum run was initiated approximately 30 minutes after the HVCI stopped collecting the sample. In between spectrum runs, background measurements and energy calibrations were performed with the spectrometer. Calibration sources were isolated behind lead brick shields for sample runs. Background runs were taken at multiple intervals during data collection and were summed and time scaled for subtraction from sample runs for analysis.

RESULTS

The small excess in counts in the first spectrometer run of sample CWU-08411-f-uf compared with the background is shown in Figure 4. The small sample contribution above a relatively much larger background illustrates a significant source of uncertainties in these measurements.
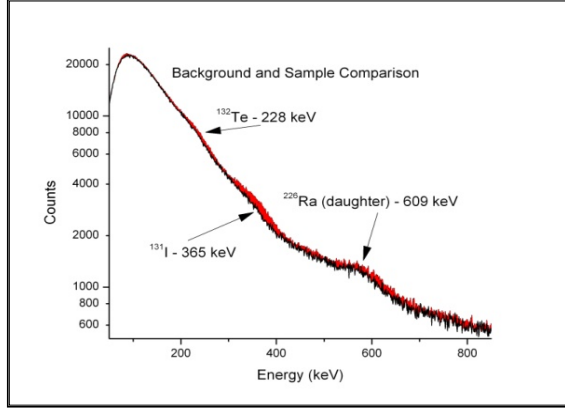
Figure 4. A scaled background run (black) and the small excess in counts above background from the initial gamma spectrum for sample CWU-08411-f-uf (red). Features pointed out in the spectrum are consistent with detections of radioactive isotopes ^{131}I and [132]Te from releases at Fukushima Daiichi and a naturally occurring [226]Ra daughter.
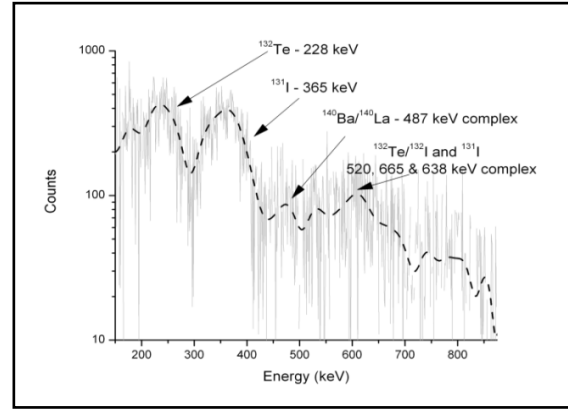

Figure 5. The energy calibrated and background subtracted gamma spectrum obtained from the initial gamma spectrum run for sample CWU-08411-f-uf showing our identification of prominent features in the spectrum. The dashed line is smoothed data.
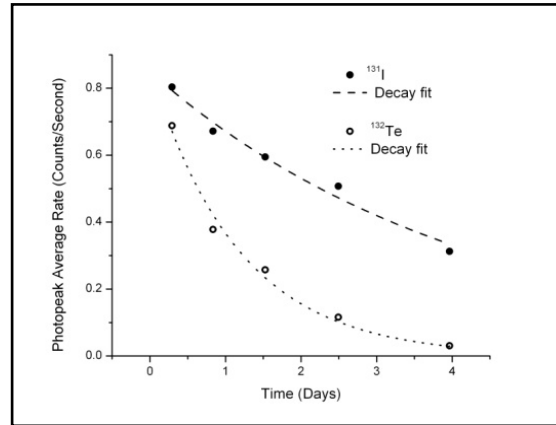
A background subtracted spectrum is shown in Figure 5. Background subtracted spectra were used to obtain the number of counts in the photopeaks that we identified as the [131]I 365 keV gamma ray and the [132]Te 228 keV gamma ray which were the most prominent photopeaks in the background subtracted spectra. The relative uncertainties due solely to counting statistics in the amplitude of the ^{131}I and [132]Te signals identified in the initial gamma spectrum for sample CWU-08411-f-uf are 5.4 % and 8.4% respectively. The decay curves for these two photopeaks are shown in Figure 6.

We used the photopeak counts for [131]I (365 keV) and [132]Te (228 keV) gammas to estimate the activity of these isotopes in the HVCI sample (see Table 1). We estimated a_0, the activity of ^{131}I and [132]Te in HVCI sample CWU-08411-f-uf at the time the filters were removed from the HVCI, using the following expression:

$$a_0 = \frac{r\, e^{\frac{t_{mr}\,\ln 2}{T_{1/2}}}}{\varepsilon\, b\, C} \qquad (1)$$

where r is the average rate of counts in a region of interest centered on the photopeak (365 keV for ^{131}I; 228 keV for [132]Te) for the

Figure 6. Detected average count rates in the photopeaks for the 365 keV gamma from ^{131}I and for the 228 keV gamma from [132]Te as a function of mid-run time for five gamma spectrum runs performed on sample CWU-08411-f-uf. Time 0 corresponds to removal of sample CWU-08411 from the HVCI. The fits are standard exponential decay curves, e.g., $r = c\, e^{-\alpha t}$.

intial gamma spectrum for sample CWU-08411-f-uf, t_{mr} is the midrun time of the intial gamma spectrum for sample CWU-08411-f-uf, $T_{1/2}$ is the accepted value for the half-life of the radioactive isotope, ε is the estimated efficiency of the detector for the sample configuration and photopeak energy, b is the branching ratio of the isotope that produces the detected gamma, and C is the definition of the Curie, 3.70×10^{10} decays/second.

Detected Isotopes	Prominent photopeak centroid energy, Measured/[Actual] (keV)	Estimated activity of isotope in sample when removed from HVCI (pCi)	Half-Life Measured/ [Actual] (hours)
^{131}I	361 / [365]	2000 (600<a<6000)	71 / [192]
^{132}Te	239 / [228]	800 (300<a<3000)	19 / [77]
^{132}I	*	*	* / [2.3]
^{140}Ba & ^{140}La	*	*	* / [300 & 40]

Table 1. Results of radioactivity and half-life calculations of sample CWU-08411-f-uf. The numbers in parentheses - (x<a<y) - represent the range of the uncertainties in the activity measurements. An asterisk - *- indicates insufficient signal or resolution.

DISCUSSION

The principal sources of uncertainties in our measurements are statistical counting uncertainties as a result of subtraction of a large background to obtain a relatively weak signal, background in detected radioactive isotope photopeaks contributed by other radioactive isotopes that were almost certainly present in the sample but which were below detection threshold, and electronic drift in the gamma spectrometer over the ten day period during which gamma spectra were obtained.

The quantitative half-lives obtained from the measurements and analysis of what we identified as the ^{131}I 365 keV and ^{132}Te 228 keV photopeaks in the sample spectra are in significant disagreement with actual values for these half-lives, though the qualitative trends and relative decay rates for ^{131}I and ^{132}Te are what we would expect [Firestone, 2011]. The presence of the isotopes shown in Table 1 strongly suggests that the sample also contained other radioactive isotopes that were below detection threshold due to detector resolution and efficiency. For instance, ^{132}I is a radioactive daughter of ^{132}Te and must be present if there is a detection of ^{132}Te Lederer, 1978]. Members of this group of radioactive isotopes with short half-lives and gamma energies either similar to or above

those of the detected isotopes (e.g., ^{132}I) would, given our methods, result in measured half-lives that are too short suggesting this as one possible explanation for the quantitative disagreement of the measured half-lives. The properties of isotopes relevant to this discussion are shown in Table 2.

Isotope	Gamma Rays (keV)	Half-Life
^{132}I	523,630,668	2.3 hours
^{140}Ba	163,305,424	12.8 days
^{140}La	329,398,432	40.2 hours
^{137}Cs	662	30 years

Table 2. Gamma energies and half-lives associated with isotopes relevant to the discussion in the text. All data in the table are from [Heath, 1997].

The fact that we did not detect ^{137}Cs (which would have been indicated in the NaI(Tl) gamma spectra by a photopeak at 662 keV) even though it is one of the most likely radioactive isotopes to be released by a damaged nuclear reactor is, in fact, consistent with the ^{131}I signal we were able to detect. Because the half-life of ^{137}Cs (30 years) is much longer than the half-life of ^{131}I (8 days), if comparable masses of these two isotopes were present in sample CWU-08411-f-uf the activity of the ^{131}I would be on the order of 1000 times greater than the activity of the ^{137}Cs in the sample, and thus the ^{137}Cs signal would have been well below our detection threshold.

Because of common perception of, and potential for overreaction to the public health risks especially associated with radioactivity, we think it is very important to briefly comment on the (lack of) public health implications for the Kittitas Valley of Washington state associated with the detected radioactive isotopes from the Fukushima Daiichi accident that are specifically identified in this report. The graph in Figure 3 compellingly illustrates that the radioactive isotopes associated with the Fukushima Daiichi accident and collected in sample CWU-08411-f-uf contribute only a very slight excess in activity above the naturally occurring background. In the energy interval that we identified as the ^{131}I photopeak for the initial gamma spectrum, which was the most prominent Fukushima related signal obtained from sample CWU-08411-f-uf, the count rate for the ^{131}I photopeak was less than a tenth of the background count rate. To elaborate on this point, while we reported that sample CWU-08411-f-uf contained approximately 2,000 pCi of ^{131}I, by way of comparison the typical adult has approximately 100,000 pCi of naturally occurring radioactive ^{40}K in their body at any given time [Peterson, 2007]. Finally, sample CWU-08411-f-uf was obtained by sampling a volume of air that is the same order of magnitude as the volume that the typical adult breathes over an entire year. The now clearly protracted nature of the Fukushima Daiichi accident suggests that continued monitoring of its impact in the US was and is prudent, but sample CWU-08411-f-uf obtained almost at the beginning of these events showed a signal that is consistent with posing no public health risk in the Kittitas Valley.

We conclude that our methods were able to detect radioactive isotopes ^{131}I and ^{132}Te associated with fine particulate matter in the Kittitas Valley of Washington state due to releases from the Fukushima Daiichi nuclear plant in March 2011. The amounts of these radioactive isotopes that were detected in HVCI sample CWU-08411-f-uf pose no hazard to residents of the Kittitas Valley.

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