Activities of $\gamma$-ray emitting isotopes in rainwater from Greater Sudbury, Canada following the Fukushima incident

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Abstract

We report the activity measured in rainwater samples collected in the Greater Sudbury area of eastern Canada on 3, 16, 20, and 26 April 2011. The samples were $\gamma$-ray counted in a germanium detector and the isotopes $^{131}$I and $^{137}$Cs, produced by the fission of $^{235}$U, and $^{134}$Cs, produced by neutron capture on $^{133}$Cs, were observed at elevated levels compared to a reference sample of ice-water. These elevated activities are ascribed to the accident at the Fukushima Dai-ichi nuclear reactor complex in Japan that followed the 11 March earthquake and tsunami. The activity levels observed at no time presented health concerns.

1 Introduction

The nuclear accident in the Fukushima area in Japan released radioisotopes to the atmosphere which have been measured in several locations in Asia [Bolsunovsky et al., 2011; Fushimi et al., 2011; Momoshima et al., 2011], North America [Bowyer et al., 2011; Leon et al., 2011; Norman et al., 2011; Sinclair et al., 2011; MacMullin et al., 2011], and Europe [Clemenza et al., 2011; Manolopoulou et al., 2011; Pittauerová et al., 2011], as the radioactivity spread around the Earth. We report here the measurement of several isotopes in water samples collected in eastern North America during April 2011, from 3 to 7 weeks after the Fukushima incident.

2 Experimental Methods

To investigate the dispersal of radioactivity from the Fukushima incident we collected samples of rainwater in aluminum trays in Greater Sudbury, Ontario on 3, 16, 20, and 26 April, the first rainy days that followed 11 March. A reference sample of ice water from Meatbird Lake in Lively, Ontario was also collected on 3 April. Within 1–2 d of their collection the water samples were passed through Whatman Grade 1 filters (medium porosity, > 11 $\mu$m) and then poured into 1 L polyethylene Marinelli beakers. Filtration was necessary to remove particulate material that was present in the samples because they were collected at ground level under windy conditions. The volume of
all samples was very close to 1 L. The beakers were sealed, encapsulated in nearly air-tight bags, and transported to the SNOLAB underground laboratory where they were γ-ray counted by a high-purity germanium detector. To minimize the background from ambient $^{222}$Rn in the mine air [Lawson and Cleveland, 2011], the samples began to be counted immediately after their arrival underground. The duration of counting was 1 d except for the ice sample which was counted for 2 d. Data on the samples and counting periods are given in Table 1.

Table 1: Data on samples. Dates of sample collection and start of counting are given in day of year 2011 in Eastern Standard Time (GMT-5 h). Dead time during counting was negligible.

| Sample     | Date collected (d) | Volume (mL) | Date counting began (d) | Counting time (d) |
|------------|--------------------|-------------|-------------------------|-------------------|
| Ice 3 April| 93.46              | 935         | 96.315                  | 2.01              |
| Rain 3 April| 93.96             | 857         | 95.345                  | 0.93              |
| Rain 16 April| 106.48            | 935         | 109.275                 | 1.02              |
| Rain 20 April| 110.57            | 1015        | 123.314                 | 0.97              |
| Rain 26 April| 116.44            | 1050        | 124.356                 | 1.0               |

The dimensions of the Ge detector crystal are 63-mm length by 67-mm diameter and its efficiency for the 1333-keV γ-rays from a $^{60}$Co source is 47% relative to a 3-inch by 3-inch NaI(Tl) detector. The FWHM resolution of the detector at 1333 keV is 1.9 keV. To reduce local background the detector is shielded by 2 inches of high-purity copper and 8 inches of lead. The detector shield is enclosed in a sealed copper box through which pure nitrogen from liquid nitrogen boil-off is flowed at 2 L/min to purge $^{222}$Rn. The efficiency of the detector for γ-rays has been measured with standard sources of known decay rate.

Table 2: Specific activity of radionuclides detected in the ice water and rainwater samples. Uncertainty includes statistical and systematic components and is given with 68% confidence.

| Sample      | $^{137}$Cs (mBq/kg) | $^{134}$Cs (mBq/kg) | $^{131}$I (mBq/kg) | $^{238}$U progeny (mBq/kg) | $^{232}$Th progeny (mBq/kg) | $^{7}$Be (mBq/kg) |
|-------------|---------------------|---------------------|---------------------|-----------------------------|----------------------------|------------------|
| Ice 3 April | $< 0.4$            | $0.4^{+0.4}_{-0.4}$ | $22.8 \pm 3.7$       | $26.1 \pm 3.1$              | $2.4 \pm 1.7$              | $80 \pm 18$     |
| Rain 3 April| $11.0 \pm 4.1$     | $8.3 \pm 2.4$       | $668 \pm 44$         | $32.8 \pm 6.2$              | $101 \pm 11$              | $1900 \pm 180$  |
| Rain 16 April| $22.7 \pm 4.9$   | $16.8 \pm 2.9$      | $64.0 \pm 8.9$       | $9.9 \pm 4.8$               | $9.4 \pm 2.9$             | $835 \pm 95$    |
| Rain 20 April| $19.1 \pm 4.7$    | $13.1 \pm 2.5$      | $31 \pm 11$          | $< 4.8$                     | $5.9 \pm 2.7$             | $770 \pm 90$    |
| Rain 26 April| $0.9^{+1.7}_{-0.9}$| $0.7 \pm 0.7$       | $2.4^{+4.6}_{-4.4}$  | $3.3 \pm 2.6$               | $< 0.3$                   | $2700 \pm 240$  |

This detector is usually used to measure the activity of samples of materials that are being considered for use in one of the SNOLAB experiments, all of which must be made from extremely low-background components. The detector sensitivity is 1 mBq/kg (0.1 ppb) for $^{226}$Ra, 1.5 mBq/kg (0.3 ppb) for $^{228}$Th, and 21 mBq/kg (0.7 ppm) for $^{40}$K. Further information on the detector and its use is given in [Lawson and Cleveland, 2011].

The samples were collected close to the SNOLAB laboratory which is located at 46°28.5' N latitude, 81°12.0' W longitude. The counting facility is 2092-m underground where the cosmic-ray muon flux is $3.31\times10^{-10}/(\text{cm}^2\text{s})$ [SNO Collaboration, 2009].
Figure 1: Energy spectra of ice water sample from Meatbird Lake and rainwater samples.
### 3 Results and Interpretation

The raw energy spectra from each sample are shown in Figure 1. Peaks are evident from the emission of $\gamma$-rays by $^{137}\text{Cs}$ at 661.7 keV; by $^{134}\text{Cs}$ at 569.3, 604.7, and 795.9 keV; and by $^{131}\text{I}$ at 284.3, 364.5, 637.0, and 722.9 keV. $\gamma$-ray lines with intensity greater than background are also apparent at 477.6 keV from $^7\text{Be}$ (mainly produced by cosmic-ray spallation on $^{14}\text{N}$ and $^{16}\text{O}$) and at several other energies from the decays of the progeny of $^{226}\text{Ra}$ and $^{228}\text{Th}$, which were present in the water samples as impurities.

The region of a peak in a typical spectrum is shown in Fig. 2. The number of counts above background in each peak is determined by counting the number of events in a 2-FWHM region centered at the peak and subtracting half the number of events in regions of equal energy both above and below the peak. $^{137}\text{Cs}$ is an exception as there also must be subtracted a constant background in the peak of $1.72 \pm 0.13$ counts/d. This latter activity is contamination internal to the Ge detector crystal housing.

The specific activities of the isotopes observed in the waters samples are given in Table 2. Because of its short half-life the $^{131}\text{I}$ activities have been corrected to the time of sample collection. For those isotopes that produced more than one peak we checked that the activity inferred from each peak was in agreement within uncertainty and we give their weighted average.

The observation of the short-lived isotope $^{131}\text{I}$ and the high concentrations of $^{137}\text{Cs}$ and $^{134}\text{Cs}$ indicate a recent release into the atmosphere of typical reactor-produced isotopes.

Figure 3 shows the decay of activity of $^{131}\text{I}$. The half-life of the observed decay is 3.7 d, considerably less than the 8.0 d half-life of $^{131}\text{I}$. We presume this is due to the transport of the radioactivity over our measuring location and washout of the isotope from the atmosphere.
Figure 3: Specific activity of $^{131}$I vs time. The solid line is a fit of the specific activity $A$ as a function of time $t$ to the function $A(t) = A(0) \exp(-t/t_1)$ where $A(0)$ and $t_1$ are constants whose best fit values are $A(0) = 1183 \pm 99$ mBq/kg and $t_1 = 5.33 \pm 0.30$ d.

The ratio of activity of $^{134}$Cs to $^{137}$Cs is shown in Figure 4. The weighted average ratio for the four measurements is $0.72 \pm 0.13$, in agreement with the value of $\sim 0.7$ reported in [Leon et al., 2011] and the measurements given in [Momoshima et al., 2011]. This ratio is approximately constant because both of these isotopes are products of nuclear fission, $^{137}$Cs directly, and $^{134}$Cs by fission production of $^{133}$Cs followed by neutron capture.

Some laboratories [Leon et al., 2011, Norman et al., 2011, MacMullin et al., 2011] have detected $^{132}$Te from the Fukushima incident, but we did not observe this isotope. Our supposition is that this is because of its short half-life of 3.2 d, the appreciable delay between the release and our first measurements, and the low volatility of Te.

At no time during our measurements were the activities of the isotopes we detected from Fukushima of any radioactive concern to the inhabitants of northern Ontario. The average radioactivity levels were much less than what is received by normal background radiation and we were only able to observe these isotopes because of our extremely sensitive well-shielded low-background apparatus.

4 Summary

Several nuclear reactor fission products were observed in rainwater samples collected in Greater Sudbury and $\gamma$-ray counted in a high-purity germanium detector at SNO-LAB. The short-lived isotope $^{131}$I and the longer-lived isotopes $^{134}$Cs and $^{137}$Cs, were detected at concentrations much higher than in a background sample. The presence of all these isotopes is associated with their release to the atmosphere from the nuclear
The accident at the Fukushima Dai-ichi reactors in Japan. These data, along with measurements made in other places around the world, may aid our understanding of the release of radioactive fission products and their transport in the atmosphere.

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