



Short communication

Iodine isotopes in precipitation: Four-year time series variations before and after 2011 Fukushima nuclear accident



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ABSTRACT

Rainwater samples were collected monthly from Fukushima, Japan, in 2012–2014 and analysed for ¹²⁷I and ¹²⁹I. These are combined with previously reported data to investigate atmospheric levels and behaviour of Fukushima-derived ¹²⁹I before and after the 2011 nuclear accident. In the new datasets, ¹²⁷I and ¹²⁹I concentrations between October 2012 and October 2014 varied from 0.5 to 10 µg/L and from 1.2×10^8 to 6.9×10^9 atoms/L respectively, resulting in ¹²⁹I/¹²⁷I atomic ratio ranges from 3×10^{-8} to 2×10^{-7} . The ¹²⁷I concentrations were in good agreement with those in the previous period from March 2011 to September 2012, whereas the ¹²⁹I concentrations and ¹²⁹I/¹²⁷I ratios followed declining trends since the accident. Although ¹²⁹I concentrations in five samples during the period of 2013–2014 have approached the pre-accident levels, ¹²⁹I concentrations in most samples remained higher values in winter and spring-summer. The high ¹²⁹I levels in winter and spring-summer are most likely attributed to local resuspension of the Fukushima-derived radionuclide-bearing fine soil particles deposited on land surfaces, and re-emission through vegetation taking up ¹²⁹I from contaminated soil and water, respectively. Long-term declining rate suggests that contribution of the Fukushima-derived ¹²⁹I to the atmosphere would become less since 2014.

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1. Introduction

After the accident of Fukushima Dai-ichi nuclear power plant (FDNPP) caused as a consequence of the tsunami resulted from the giant earthquake on March 11, 2011, the rainwater samples were monthly collected from Fukushima City, and analysed for ¹²⁷I and ¹²⁹I to assess the atmospheric level and behaviour of the Fukushima-derived ¹²⁹I (Xu et al., 2013). The ¹²⁹I concentration of 10^8 atoms/L in 2010 before the accident dramatically increased about four orders of magnitude to 7.6×10^{11} atoms/L in March 2011 immediately after the accident with a ¹²⁹I/¹²⁷I ratio up to 6.9×10^{-5} . Afterwards the ¹²⁹I concentrations in precipitation decreased exponentially, with several fluctuations, to $\sim 4 \times 10^8$ atoms/L in November 2012. Such a temporal fluctuation has been explained as a result of continuous removal of the ¹²⁹I released to the atmosphere in the Fukushima accident combined with re-suspension of

the Fukushima-derived ¹²⁹I deposited on the land surface. In comparison with the ¹²⁹I pre-accident level of $(1.7 \pm 0.8) \times 10^8$ atoms/L averaged from rainwater samples between November 2010 and February 2011, the value of ¹²⁹I concentration in November 2012 (3.6×10^8 atoms/L) remained more than two times higher. Therefore, it is necessary to understand whether the Fukushima-derived ¹²⁹I in atmosphere has subsequently declined to pre-accident levels. If this is a case, it is also worthwhile to investigate temporal variations since December 2012, because later elevated ¹²⁹I events may be an indicator of new releases associated with the decommission of FDNPP and resuspension caused by natural processes and/or decontamination (Hirose, 2013; Tsuruta et al., 2014). This work aims to extend the investigation on levels and behaviour of Fukushima-derived ¹²⁹I in the atmosphere by analysis of a further 2-year time-series of precipitation samples collected from Fukushima.

2. Materials and methods

The rainwater samples were monthly collected at campus of the

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Fukushima University (37°41′00″N, 140°27′16″E), located about 60 km northwest of the FDNPP. Detailed description on rainwater sample collection and analytical methods for ^{127}I and ^{129}I has been described previously (Xu et al., 2013).

The ^{127}I concentrations in the precipitation samples were determined using inductively coupled plasma mass spectrometer at the Technical University of Denmark. Repeat analysis of a reference material indicates that the typical uncertainty of ^{127}I concentrations is about 1%.

A modified method was used to separation of iodine from precipitation and accelerator mass spectrometry measurement of ^{129}I . Firstly, instead of NaOH used previously, $\text{K}_2\text{S}_2\text{O}_5$ is used to treat the sample in this study in order to completely convert organic iodine in rainwater to inorganic form (Dang et al., 2013). Secondly, instead of $^{129}\text{I}^{5+}$ ion detection at 3 MV terminal voltage in previous study, the $^{129}\text{I}^{3+}$ was chosen for detection in this study so that the ion transmission can be significantly improved for the low-level ^{129}I determination. In this operational condition, interference of $^{97}\text{Mo}^{4+}$ (disassociated from the injected MoO_2^-) to the measurement of $^{129}\text{I}^{5+}$ can be removed. Instead, there are two main interferences of $^{86}\text{Sr}^{2+}$ and $^{43}\text{Ca}^+$ (e.g., disassociated from injected SrCO_2^- and CaClO_3^- or CaSO_3^- , respectively) to $^{129}\text{I}^{3+}$, but they can be completely separated using the gas ionization detector, resulting in no need of any interference corrections.

The measured $^{129}\text{I}/^{127}\text{I}$ ratios is normalized to a standard with $^{129}\text{I}/^{127}\text{I}$ ratio of 1.098×10^{-10} prepared by dilution of the NIST 4949B standard reference material with ^{127}I carrier (Woodward) with $^{129}\text{I}/^{127}\text{I}$ ratio of 10^{-14} . The $^{129}\text{I}/^{127}\text{I}$ ratios in the prepared target of rainwater samples are in range of 10^{-12} – 10^{-11} , which are more than one order of magnitude higher than those of procedure blank (10^{-13}). Repeat measurements of a secondary standard with $^{129}\text{I}/^{127}\text{I}$ ratio of 1.063×10^{-11} indicated better than 3% precision and accuracy.

3. Results

Table 1 lists ^{127}I and ^{129}I concentrations, and $^{129}\text{I}/^{127}\text{I}$ ratios newly measured in rainwater samples collected from December 2012 to October 2014 together with previously reported ^{127}I data from March 2011 to September 2012 and ^{129}I data from November 2010 to November 2012 (Xu et al., 2013). Fig. 1 shows temporal variations of ^{127}I and ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios in the rainwater samples from the whole period from November 2010 to October 2014.

The newly measured ^{127}I concentrations between October 2012 and October 2014 varied from 0.5 $\mu\text{g/L}$ in July 2014 to 10 $\mu\text{g/L}$ in March 2013, which is consistent with the previous period between March 2011 and September 2012 ranging from 0.8 to 2.3 $\mu\text{g/L}$. Combination of the two datasets gives an average ^{127}I concentration of $1.6 \pm 1.5 \mu\text{g/L}$ in the period between March 2011 and October 2014. As shown in Fig. 2, these data fall into the reported range in the literature (0.2–12 $\mu\text{g/L}$, Aldahan et al., 2009). Such natural variation of ^{127}I is reflected by rainwater samples from Japan (Muramatsu and Ohmomo, 1986; Xu et al., 2016a), China (Zhang et al., 2011), Europe (Buraglio et al., 2001; Reithmeier et al., 2005; Hou et al., 2009; Gómez-Guzmán et al., 2012) and USA (Moran et al., 1999). Overall, there is no apparent trend on temporal variation of ^{127}I concentration in Fukushima through the whole period.

The newly ^{129}I concentrations in rainwater samples between December 2012 and October 2014 varied from 1.2×10^8 atoms/L in October 2014 to 6.9×10^9 atoms/L in March 2013. The highest value is comparable with those in August–September 2011, whereas the lowest value is consistent with those observed before the accident. The ^{129}I concentrations in 2012–2014 showed a declining trend after the accident, but were significantly lower compared to the

previous period in 2011–2012. Overall, as shown in Fig. 2, the ^{129}I concentrations in precipitation from Fukushima are comparable with those observed in North Europe (Germany, Denmark, Sweden) and South Europe (Spain), but significantly higher than those from USA (Moran et al., 1999). The elevated ^{129}I concentrations in Europe have been attributed to contributions from the nuclear reprocessing plants (Aldahan et al., 2009; Hou et al., 2009; Gómez-Guzmán et al., 2012).

The newly measured $^{129}\text{I}/^{127}\text{I}$ atomic ratios in this study vary within relatively narrow bands from 1.3×10^{-8} to 2.0×10^{-7} . The lowest $^{129}\text{I}/^{127}\text{I}$ ratio of 1.3×10^{-8} observed in October 2014 is consistent with those observed in pre-accident soil samples nearby (Matsunaka et al., 2015). If the average ^{127}I in the whole period $1.6 \pm 1.5 \mu\text{g/L}$ as described above can be assumed for the pre-accident rainwaters, the corresponding pre-accident $^{129}\text{I}/^{127}\text{I}$ atomic ratios would be $(1-4) \times 10^{-8}$. Clearly, although six samples collected in the period of 2012–2014 showed $^{129}\text{I}/^{127}\text{I}$ ratios $(3-4) \times 10^{-8}$ consistent with the pre-accident values, $^{129}\text{I}/^{127}\text{I}$ ratios in most samples are 2–10 times higher than the pre-accident values.

Overall, ^{129}I and $^{129}\text{I}/^{127}\text{I}$ show gradually declining trends through the whole period (Fig. 1b and c). The ^{129}I concentrations are averagely 1×10^{11} atoms/L in 2011 (since the accident), 2×10^9 atoms/L in 2012, 6×10^8 atoms/L in 2013 and 3×10^8 atoms/L in 2014, whereas the corresponding $^{129}\text{I}/^{127}\text{I}$ ratios are 2×10^{-5} in 2011, 3×10^{-7} in 2012, 9×10^{-8} in 2013 and 6×10^{-8} in 2014. In addition, a roughly seasonal variation on ^{129}I and $^{129}\text{I}/^{127}\text{I}$ can be observed. The high ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios are generally observed in winter and spring-summer, whereas the low values are found in autumn.

Table 1 also lists the calculated ^{127}I and ^{129}I depositions by combining the precipitation with ^{127}I and ^{129}I concentrations. The ^{127}I and ^{129}I depositions are in range of 0.7–14 $\mu\text{g/m}^2/\text{d}$ and 2×10^8 – 1×10^{11} atoms/ m^2/d , respectively. With the exception of early periods of 2011, both ^{127}I and ^{129}I depositions overlap those observed in North Europe (0.9–4.1 $\mu\text{g/m}^2/\text{d}$ for ^{127}I and $(8-80) \times 10^8$ atoms/ m^2/d for ^{129}I , Aldahan et al., 2009) and South Europe ($(0.1-8) \times 10^8$ atoms/ m^2/d for ^{129}I , Gómez-Guzmán et al., 2012).

One of the most striking features in the present dataset is that both ^{127}I and ^{129}I concentrations in March 2013 are higher than those in previous and later months by one order of magnitude. The high ^{127}I and ^{129}I concentrations coincide with the lowest precipitation in this sample (Table 1). The similar high ^{127}I and ^{129}I concentrations are also observed in sample collected in August 2012 with the relatively low precipitation. In contrast, the relatively low iodine concentrations (in particular ^{127}I) are observed in other samples with high precipitation. This is mainly attributed to the washing out and trapping process of iodine during precipitation. Iodine in the atmosphere presents in different forms including particle associated, inorganic species (I_2 , HI, HIO, etc.) and organic gaseous species (e.g., alkyl-iodide). Of them particle associated iodine and inorganic species of iodine are easily trapped by droplet of rain or during the formation of cloud, causing a high removal rate of iodine from the atmosphere in the beginning of rainfall event. While organic iodine species is not easily trapped or converted to soluble iodine in the droplet of rain, therefore removed from atmosphere slowly. This causes a high iodine concentration in a low precipitation event compared to a high precipitation event which iodine in the rainwater is highly diluted. The similar pattern has been previously observed in a precipitation in USA (Moran et al., 1999).

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