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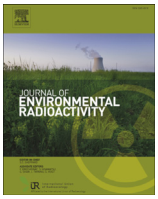

Short communication

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 ^{127}I and ^{129}I. These are combined with previously reported data to investigate atmospheric levels and behaviour of Fukushima-derived ^{129}I before and after the 2011 nuclear accident. In the new datasets, ^{127}I and ^{129}I 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 ^{129}I/^{127}I atomic ratio ranges from 3×10^{-8} to 2×10^{-7}. The ^{127}I concentrations were in good agreement with those in the previous period from March 2011 to September 2012, whereas the ^{129}I concentrations and ^{129}I/^{127}I ratios followed declining trends since the accident. Although ^{129}I concentrations in five samples during the period of 2013–2014 have approached the pre-accident levels, ^{129}I concentrations in most samples remained higher values in winter and spring-summer. The high ^{129}I 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 ^{129}I from contaminated soil and water, respectively. Long-term declining rate suggests that contribution of the Fukushima-derived ^{129}I to the atmosphere would become less since 2014.

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 ^{127}I and ^{129}I to assess the atmospheric level and behaviour of the Fukushima-derived ^{129}I (Xu et al., 2013). The ^{129}I 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 ^{129}I/^{127}I ratio up to 6.9×10^{-5}. Afterwards the ^{129}I concentrations in precipitation decreased exponentially, with several fluctuations, to ~4×10^8 atoms/L in November 2012. Such a temporal fluctuation has been explained as a result of continuous removal of the ^{129}I released to the atmosphere in the Fukushima accident combined with re-suspension of the Fukushima-derived ^{129}I deposited on the land surface. In comparison with the ^{129}I 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 ^{129}I 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 ^{129}I 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 ^{129}I 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 ^{129}I 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, $K_2S_2O_5$ 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 $^{129}I^{5+}$ ion detection at 3 MV terminal voltage in previous study, the $^{129}I^{3+}$ 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 $^{97}Mo^{4+}$ (disassociated from the injected MoO_2^-) to the measurement of $^{129}I^{5+}$ can be removed. Instead, there are two main interferences of $^{86}Sr^{2+}$ and $^{43}Ca^{+}$ (e.g., disassociated from injected $SrCO_2^-$ and $CaClO_3^-$ or $CaSO_3^-$, respectively) to $^{129}I^{3+}$, but they can be completely separated using the gas ionization detector, resulting in no need of any interference corrections.

The measured $^{129}I/^{127}I$ ratios is normalized to a standard with $^{129}I/^{127}I$ ratio of 1.098×10^{-10} prepared by dilution of the NIST 4949B standard reference material with ^{127}I carrier (Woodward) with $^{129}I/^{127}I$ ratio of 10^{-14}. The $^{129}I/^{127}I$ 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 $^{129}I/^{127}I$ 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 $^{129}I/^{127}I$ 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 $^{129}I/^{127}I$ 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 μg/L in July 2014 to 10 μg/L in March 2013, which is consistent with the previous period between March 2011 and September 2012 ranging from 0.8 to 2.3 μg/L. Combination of the two datasets gives an average ^{127}I concentration of 1.6 ± 1.5 μg/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 μg/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 $^{129}I/^{127}I$ atomic ratios in this study vary within relatively narrow bands from 1.3×10^{-8} to 2.0×10^{-7}. The lowest $^{129}I/^{127}I$ 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 ± 1.5 μg/L as described above can be assumed for the pre-accident rainwaters, the corresponding pre-accident $^{129}I/^{127}I$ atomic ratios would be $(1–4) \times 10^{-8}$. Clearly, although six samples collected in the period of 2012–2014 showed $^{129}I/^{127}I$ ratios $(3–4) \times 10^{-8}$ consistent with the pre-accident values, $^{129}I/^{127}I$ ratios in most samples are 2–10 times higher than the pre-accident values.

Overall, ^{129}I and $^{129}I/^{127}I$ 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 $^{129}I/^{127}I$ 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 $^{129}I/^{127}I$ can be observed. The high ^{129}I concentrations and $^{129}I/^{127}I$ 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 μg/m^2/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 μg/m^2/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 presentation 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).

Table 1
^{127}I and ^{129}I concentrations in rainwaters collected from Fukushima.[a]

Sampling date	Rainfall (mm)	^{127}I[b] (μg/L)	^{129}I[b] (atoms/L)	^{129}I/^{127}I (atomic ratio)	^{127}I deposition (μg/m^2/d)	^{129}I deposition (atoms/m^2/d)
Nov 2010	72		$(1.16 \pm 0.10) \times 10^{8}$			2.8×10^{8}
Dec 2010	142		$(1.70 \pm 0.10) \times 10^{8}$			8.1×10^{8}
Jan 2011	22		$(2.77 \pm 0.10) \times 10^{8}$			2.0×10^{8}
Feb 2011	65		$(9.60 \pm 1.00) \times 10^{7}$			2.1×10^{8}
Mar 2011	57	2.32 ± 0.02	$(7.58 \pm 1.99) \times 10^{11}$	$(6.89 \pm 0.19) \times 10^{-5}$	4.4	1.4×10^{12}
1–20 Apr 2011	46		$(1.79 \pm 0.02) \times 10^{11}$			2.7×10^{11}
21 Apr - 9 May 2011	49		$(1.04 \pm 0.03) \times 10^{11}$			1.7×10^{11}
3 Jun 2011		1.04 ± 0.01	$(3.90 \pm 0.09) \times 10^{10}$	$(7.93 \pm 0.21) \times 10^{-6}$		
15 Jun 2011		2.23 ± 0.01	$(7.62 \pm 0.17) \times 10^{10}$	$(7.20 \pm 0.17) \times 10^{-6}$		
30 Jun 2011		0.917 ± 0.002	$(1.71 \pm 0.01) \times 10^{10}$	$(3.94 \pm 0.03) \times 10^{-6}$		
15 Jul 2011		0.943 ± 0.004	$(2.10 \pm 0.03) \times 10^{10}$	$(4.69 \pm 0.08) \times 10^{-6}$		
16 Jul - 5 Aug 2011	97		$(1.08 \pm 0.01) \times 10^{10}$			3.5×10^{10}
Aug 2011	94		$(1.07 \pm 0.01) \times 10^{10}$			3.4×10^{10}
Sep 2011	322		$(3.45 \pm 0.06) \times 10^{9}$			3.7×10^{10}
Oct 2011	127		$(3.29 \pm 0.07) \times 10^{9}$			1.4×10^{10}
Dec 2011	25	0.854 ± 0.014	$(4.42 \pm 0.05) \times 10^{9}$	$(1.09 \pm 0.02) \times 10^{-6}$	0.7	3.7×10^{9}
Jan 2012	63	1.43 ± 0.02	$(4.46 \pm 0.05) \times 10^{9}$	$(6.58 \pm 0.11) \times 10^{-7}$	3.0	9.4×10^{9}
Feb 2012	167	0.836 ± 0.010	$(1.36 \pm 0.02) \times 10^{9}$	$(3.42 \pm 0.07) \times 10^{-7}$	4.7	7.6×10^{9}
Mar 2012	51	0.808 ± 0.014	$(8.98 \pm 0.20) \times 10^{8}$	$(2.34 \pm 0.07) \times 10^{-7}$	1.4	1.5×10^{9}
Apr 2012	32	2.72 ± 0.02	$(3.51 \pm 0.04) \times 10^{9}$	$(2.72 \pm 0.03) \times 10^{-7}$	2.9	3.7×10^{9}
May 2012	120	1.60 ± 0.02	$(1.61 \pm 0.02) \times 10^{9}$	$(2.12 \pm 0.04) \times 10^{-7}$	6.4	6.4×10^{9}
Jun 2012	186	1.14 ± 0.01	$(1.32 \pm 0.02) \times 10^{9}$	$(2.45 \pm 0.05) \times 10^{-7}$	7.0	8.2×10^{9}
Jul 2012	128	0.921 ± 0.010	$(1.27 \pm 0.02) \times 10^{9}$	$(2.92 \pm 0.06) \times 10^{-7}$	3.9	5.4×10^{9}
Aug 2012	15	2.19 ± 0.02	$(5.13 \pm 0.06) \times 10^{9}$	$(4.94 \pm 0.07) \times 10^{-7}$	1.1	2.6×10^{9}
Oct 2012	67	1.67 ± 0.01	$(5.45 \pm 0.10) \times 10^{8}$	$(6.88 \pm 0.14) \times 10^{-8}$	3.7	1.2×10^{9}
Nov 2012	71	1.59 ± 0.01	$(3.57 \pm 0.10) \times 10^{8}$	$(4.73 \pm 0.14) \times 10^{-8}$	3.8	8.4×10^{8}
Dec 2012	54	1.32 ± 0.01	$(4.23 \pm 0.27) \times 10^{8}$	$(6.76 \pm 0.44) \times 10^{-8}$	2.4	7.6×10^{8}
Jan 2013	60	1.28 ± 0.01	$(1.19 \pm 0.02) \times 10^{9}$	$(1.95 \pm 0.04) \times 10^{-7}$	2.6	2.4×10^{9}
Feb 2013	36	1.25 ± 0.01	$(5.88 \pm 0.12) \times 10^{8}$	$(9.94 \pm 0.20) \times 10^{-8}$	1.5	7.1×10^{8}
Mar 2013	2	10.0 ± 0.1	$(6.94 \pm 0.16) \times 10^{9}$	$(1.46 \pm 0.03) \times 10^{-7}$	0.7	4.6×10^{8}
Apr 2013	88	1.28 ± 0.01	$(6.83 \pm 0.12) \times 10^{8}$	$(1.12 \pm 0.02) \times 10^{-7}$	3.8	2.0×10^{9}
May 2013	34	2.04 ± 0.01	$(1.15 \pm 0.02) \times 10^{9}$	$(1.18 \pm 0.02) \times 10^{-7}$	2.3	1.3×10^{9}
Jun 2013	76	1.14 ± 0.01	$(7.45 \pm 0.13) \times 10^{8}$	$(1.37 \pm 0.03) \times 10^{-7}$	2.9	1.9×10^{9}
Jul 2013	286	1.51 ± 0.03	$(2.93 \pm 0.07) \times 10^{8}$	$(4.10 \pm 0.13) \times 10^{-8}$	14.4	2.8×10^{9}
Aug 2013	104	1.10 ± 0.01	$(4.37 \pm 0.09) \times 10^{8}$	$(8.38 \pm 0.18) \times 10^{-8}$	3.8	1.5×10^{9}
Sep 2013	174	1.26 ± 0.01	$(3.25 \pm 0.08) \times 10^{8}$	$(5.46 \pm 0.13) \times 10^{-8}$	7.3	1.9×10^{9}
Oct 2013	257	0.966 ± 0.004	$(2.42 \pm 0.07) \times 10^{8}$	$(5.29 \pm 0.14) \times 10^{-8}$	8.3	2.1×10^{9}
Nov 2013	15	2.71 ± 0.02	$(3.77 \pm 0.13) \times 10^{8}$	$(2.93 \pm 0.10) \times 10^{-8}$	1.4	1.9×10^{8}
Dec 2013	71	1.06 ± 0.01	$(1.57 \pm 0.06) \times 10^{8}$	$(3.10 \pm 0.12) \times 10^{-8}$	2.5	3.7×10^{8}
Jan 2014	16	1.76 ± 0.01	$(4.49 \pm 0.11) \times 10^{8}$	$(5.39 \pm 0.13) \times 10^{-8}$	0.9	2.4×10^{8}
Feb 2014	134	0.927 ± 0.005	$(2.83 \pm 0.20) \times 10^{8}$	$(6.44 \pm 0.47) \times 10^{-8}$	4.1	1.3×10^{9}
Mar 2014	158	0.968 ± 0.008	$(1.94 \pm 0.12) \times 10^{8}$	$(4.23 \pm 0.26) \times 10^{-8}$	5.1	1.0×10^{9}
Apr 2014	74	0.824 ± 0.005	$(1.57 \pm 0.10) \times 10^{8}$	$(4.02 \pm 0.27) \times 10^{-8}$	2.0	3.9×10^{8}
May 2014	70	1.09 ± 0.01	$(3.55 \pm 0.15) \times 10^{8}$	$(6.86 \pm 0.29) \times 10^{-8}$	2.6	8.3×10^{8}
Jun 2014	196	1.15 ± 0.01	$(3.92 \pm 0.39) \times 10^{8}$	$(7.21 \pm 0.73) \times 10^{-8}$	7.5	2.6×10^{9}
Jul 2014	196	0.483 ± 0.006	$(1.63 \pm 0.11) \times 10^{8}$	$(7.11 \pm 0.51) \times 10^{-8}$	3.2	1.1×10^{9}
Aug 2014	138	1.24 ± 0.01	$(4.02 \pm 0.16) \times 10^{8}$	$(6.82 \pm 0.28) \times 10^{-8}$	5.7	1.8×10^{9}
Sep 2014	31	1.38 ± 0.01	$(4.42 \pm 0.19) \times 10^{8}$	$(6.74 \pm 0.30) \times 10^{-8}$	1.4	4.6×10^{8}
Oct 2014	252	0.596 ± 0.010	$(1.22 \pm 0.12) \times 10^{8}$	$(4.33 \pm 0.43) \times 10^{-8}$	5.0	1.0×10^{9}

[a] Data of ^{127}I and ^{129}I/^{127}I prior to Oct 2012 and those of ^{129}I prior to Dec 2012 from Xu et al. (2013).

[b] The uncertainties of concentrations of ^{127}I and ^{129}I are estimated considering the statistics error of measurement as well as the contributions of all steps they are expanded uncertainties using a coverage factor of $k = 1$.

4. Discussion

4.1. Sources of ^{129}I in rainwaters

There is no doubt that source of the elevated ^{129}I exceeded the baseline of ^{129}I in Fukushima atmosphere results mainly from the ^{129}I released from the FDNPP. It is particularly true in the early period of the post-accident. With the cessation of atmospheric release from the FDNPP and following rapid dispersion of Fukushima-derived radioactive plumes in atmosphere, processes causing elevated atmospheric ^{129}I in later period may include resuspension of the fine particles carrying deposited Fukushima-derived ^{129}I on land surfaces, re-emission through vegetation taking up ^{129}I from contaminated soil and water, re-emission of Fukushima-derived ^{129}I discharged and deposited in the sea, and possible releases from the damaged reactors during decommissioning.

It is difficult to assess the role of ongoing releases from the damaged reactors during the decommissioning process with the current datasets, due to the low resolution of our sampling period. Post-accident monitoring results during last 4 y have not shown significant new releases from the FDNPP site. Aerosol samples have been collected beside our rainwater sampling site every 3 d and atmospheric ^{137}Cs was monitored from May 2011 to August 2014. The measured atmospheric ^{137}Cs activities seem unlikely to suggest significant releases from the decommissioning processes of FDNPP (Watanabe, 2015). Even though there was some releases in very short period during the decommission process, they might not be captured in our monthly rainwater samples. It is nonetheless recognized that decommissioning carries potential risks of further release, and therefore it is important that sampling and monitoring should be continued.

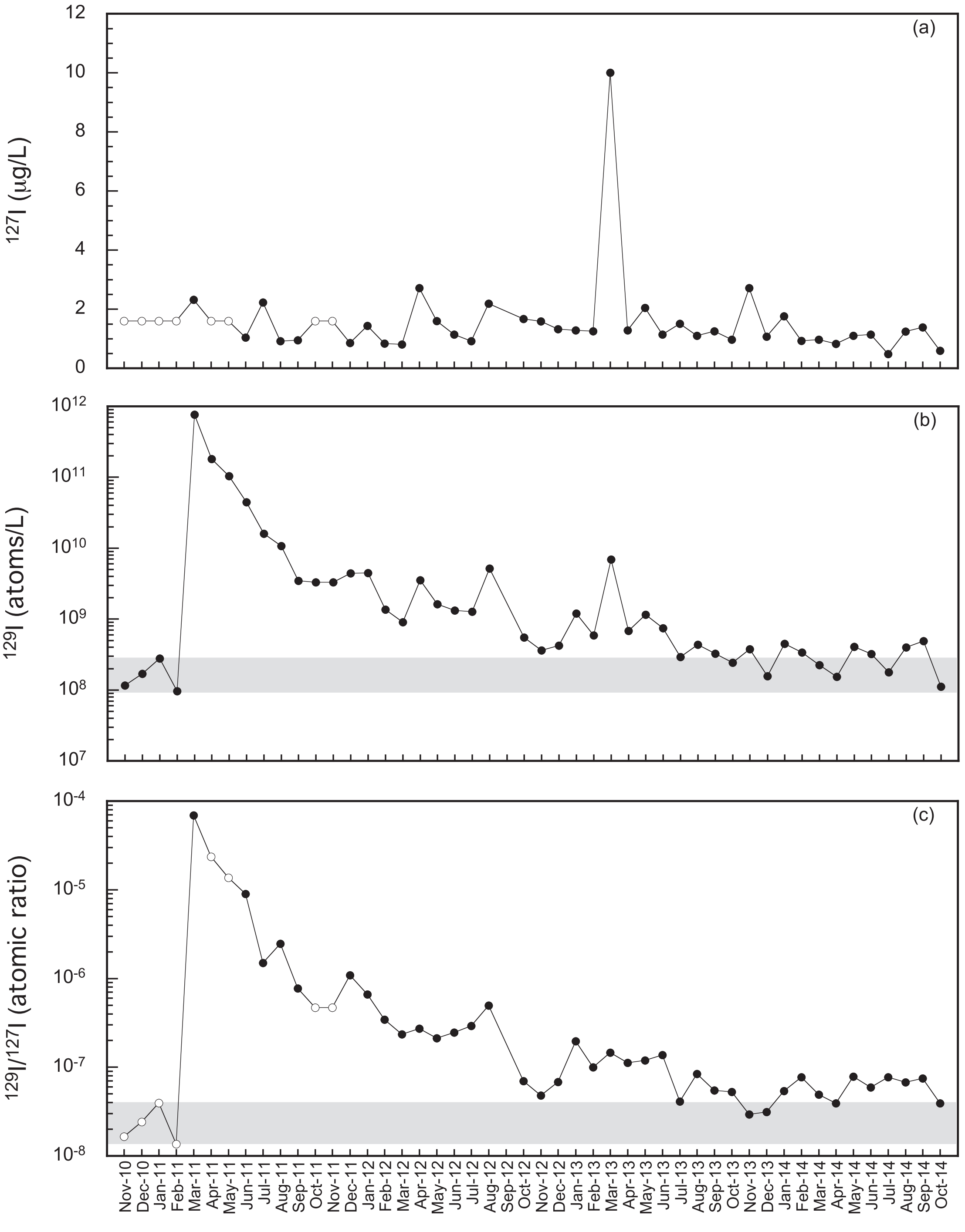

Fig. 1. Temporal variations of ^{127}I (a), ^{129}I (b) and ^{129}I/^{127}I (c) in rainwater samples from Fukushima. The open circles denote samples with an assumed ^{127}I concentration (1.6 μg/L) deduced from the averaged ^{127}I of the whole period. This value is used to calculate the corresponding ^{129}I/^{127}I ratio. The grey zones show ranges of the measured ^{129}I concentrations and estimated ^{129}I/^{127}I ratios prior to the accident.

Investigations of ^{129}I in precipitation in Europe have confirmed that ^{129}I discharged to the seas from nuclear reprocessing plants is the major source of ^{129}I in the atmosphere in Europe, especially in the North Europe (Aldahan et al., 2009). In Fukushima, in addition to gaseous release of radionuclides from the damaged reactors, indeed large amount of radioactive water has also been discharged to the sea from the FDNPP in March–April 2011, which has induced a significant increase of the concentration of radionuclides in the coast water nearby the FDNPP. The ^{129}I concentrations in seawaters in the offshore Fukushima have been reported with the highest ^{129}I concentration of 6×10^8 atoms/L and ^{129}I/^{127}I ratio of 2.1×10^{-9} (Hou et al., 2013). In comparison with the measured ^{129}I level in the rainwater in this study, the ^{129}I/^{127}I ratios in seawater are one order of magnitude lower. This seems to imply that the high dilution factors in seawater may limit the contributions of ^{129}I in the atmosphere from this pathway, although the transfer of

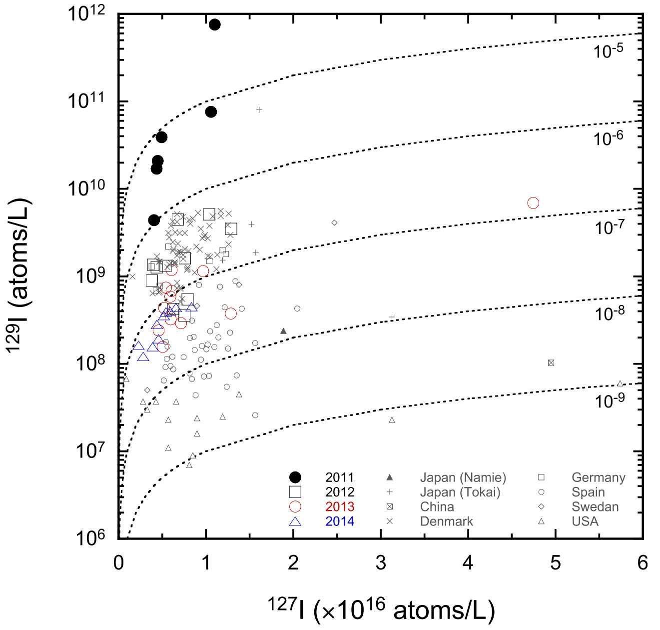

Fig. 2. Correlation between ^{127}I and ^{129}I in rainwater samples from Fukushima in this study (large symbols) and other sites worldwide (small symbols). The dotted lines stand for the isolines of ^{129}I/^{127}I ratios. Data source: Japan (Namie) from Xu et al. (2016a); Japan (Tokai) from Muramatsu and Ohmomo (1986); China from Zhang et al. (2011); Denmark from Hou et al. (2009); Germany from Reithmeier et al. (2005); Spain from Gómez-Guzmán et al. (2012); Sweden from Buraglio et al. (2001); and USA from Moran et al. (1999).

radionuclides from sea to land via the air plays an important role on terrestrial environments along the coastal regions (McKay and Pattenden, 1990).

Fig. 2 shows relationship between ^{127}I and ^{129}I concentrations in rainwater since March 2011. Overall there is no clear correlation between the two isotopes through the whole period. The similar pattern is observed in the European atmosphere, which was attributed to different sources for ^{127}I and ^{129}I (Hou et al., 2009). This is particularly true for the rainwater samples collected soon after the accident during which ^{127}I and ^{129}I originate from different sources/reservoirs. However, if focussing on later rainwater samples in 2013 and 2014, it is found that there are somewhat positive correlations between the two isotopes, although it is significantly driven by the March 2013 sample. Such correlations might suggest a common reservoir for ^{127}I and ^{129}I with a ^{129}I/^{127}I atomic ratio ~1 × 10^{-7}. It is highly considerable that this reservoir might consist of the anthropogenic Fukushima-derived ^{129}I and natural ^{127}I, which were well mixed, in environment. With the cessation of release from the FDNPP, Fukushima-derived ^{129}I in atmosphere is continuously removed by deposition (dry and wet), causing that the concentration of ^{129}I in the atmosphere is gradually declined with ecological half-lives of 29–44 d before October 2011 (Xu et al., 2013). Meanwhile, the Fukushima-derived ^{129}I deposited on land surface and sea is accumulated. Miyake et al. (2015) have reported ^{129}I/^{127}I ratios in 50 surface soils collected in the areas within 60 km radius from FDNPP in April 2011, varying from 3 × 10^{-9} to 1 × 10^{-5} with an average of (7 ± 18) × 10^{-7}. Muramatsu et al. (2015) investigated 82 surface soil samples within 80 km from FDNPP collected in June 2011 which have ^{129}I/^{127}I ratios from 2 × 10^{-8} to 4 × 10^{-6} with an average of (5 ± 6) × 10^{-7}. Matsunaka et al. (2015) analysed three surface soil samples nearby the FDNPP. Two samples collected 4.3 km and 8.2 km from FDNPP in November 2012 showed ^{129}I/^{127}I ratios of 2.14 × 10^{-6} and 4.18 × 10^{-7}, respectively. One sample collected 7.5 km from FDNPP in June 2013 gave ^{129}I/^{127}I ratio of 2.33 × 10^{-6}. Averaging the three samples results in ^{129}I/^{127}I ratio of (1.6 ± 1.0) × 10^{-6}. These values of ^{129}I/^{127}I ratio are much higher than that observed in most of precipitation sample collected after January 2013 in this work (>2 × 10^{-7}). Suspension of the highly contaminated soil particles can therefore significantly enhance the ^{129}I concentration and ^{129}I/^{127}I ratio in the atmosphere. In addition, organic activity in the soil can emit gaseous iodine to the atmosphere from the soil, which results in an enhanced ^{129}I concentration and ^{129}I/^{127}I ratio in the atmosphere (Bostock et al. 2003). In addition, two Japanese cedar frond samples collected in Iwaki in January 2013, 50 km southwest of the FDNPP show ^{129}I/^{127}I ratios of 2.46 × 10^{-7} and 8.38 × 10^{-7} (Xu et al. 2016b), which is also much higher than that in the most of precipitation samples after January 2013. Due to the slow dynamic processing of iodine in trees compared to vegetables, a similar ^{129}I/^{127}I ratio in the vegetable and soil it grows is expected. It have been reported that many vegetation (e.g., rice plant and grass) can release gaseous iodine (e.g., alkyl-iodide, HI, etc.) to the atmosphere. It has been estimated that the releases from vegetation is one of the major sources of iodine in the atmosphere (Muramatsu et al. 1991; Redeker et al. 2000). Therefore the release of iodine from vegetables in the contaminated land to the atmosphere might be another source of ^{129}I in the atmosphere. In addition, pollen released by plants in the contaminated field might be also important source of ^{129}I in the atmosphere.

Therefore, it can be concluded that the post-accident elevated ^{129}I in Fukushima can be mainly attributed to resuspension of the Fukushima-derived ^{129}I deposited on land surfaces to atmosphere in the form of fine soil particles and pollen, and to re-emission through vegetation which uptake ^{129}I from the contaminated soil and water as well as the biological activities in the contaminated soil.

In addition to the Fukushima-derived radionuclides, it should be pointed out that other sources such as re-emission and transportation from the European reprocessing plants (in particular La Hague in France and Sellafield in UK) might count for the elevated ^{129}I in this study. This may be particular true for the later periods after the accident. For example, since July 2013, the ^{129}I values varied from 1.2 × 10^{8} atoms/L to 4.5 × 10^{8} atoms/L of which seven samples are below the pre-accident level. The similar results are also found in five samples having ^{129}I/^{127}I below 5 × 10^{-8}, close to or below the pre-accident level. Toyama et al. (2012, 2013) reported the long-term atmospheric ^{129}I/^{127}I ratios in Japan between 1963 and 2005. In their dataset, the atmospheric ^{129}I/^{127}I ratios were in range of (1–11) × 10^{-8} in May and (0.7–3) × 10^{-8} in September during the period of 1995–2005. The high depositions of ^{129}I in May are attributable to the transport of the continental air mass directly affected by the European sources and other possible plant located between Europe and Japan (Toyama et al., 2012). Therefore, it implies that contribution of ^{129}I from European reprocessing plants cannot be ruled out for Fukushima samples in spring-summer such as May–June in 2013 and 2014. To clarify these alternatives, long-term observation is required. Moreover, considering the possible spatial variations, observation near the FDNPP would be helpful to distinguish the different sources.

4.2. *Factors affecting resuspension and re-emission of the deposited ^{129}I*

Variations of the ^{129}I concentration in atmosphere after the FDNPP accident are governed by several factors, which include the atmospheric emission history of ^{129}I at the FDNPP, resuspension of Fukushima-derived ^{129}I-bearing soil particles, meteorological factors and migration of Fukushima-derived ^{129}I deposited in land surface. Fig. 3 compares four-year variations of ^{129}I in rainwaters with ^{137}Cs in aerosols (Watanabe, 2015) both of which were

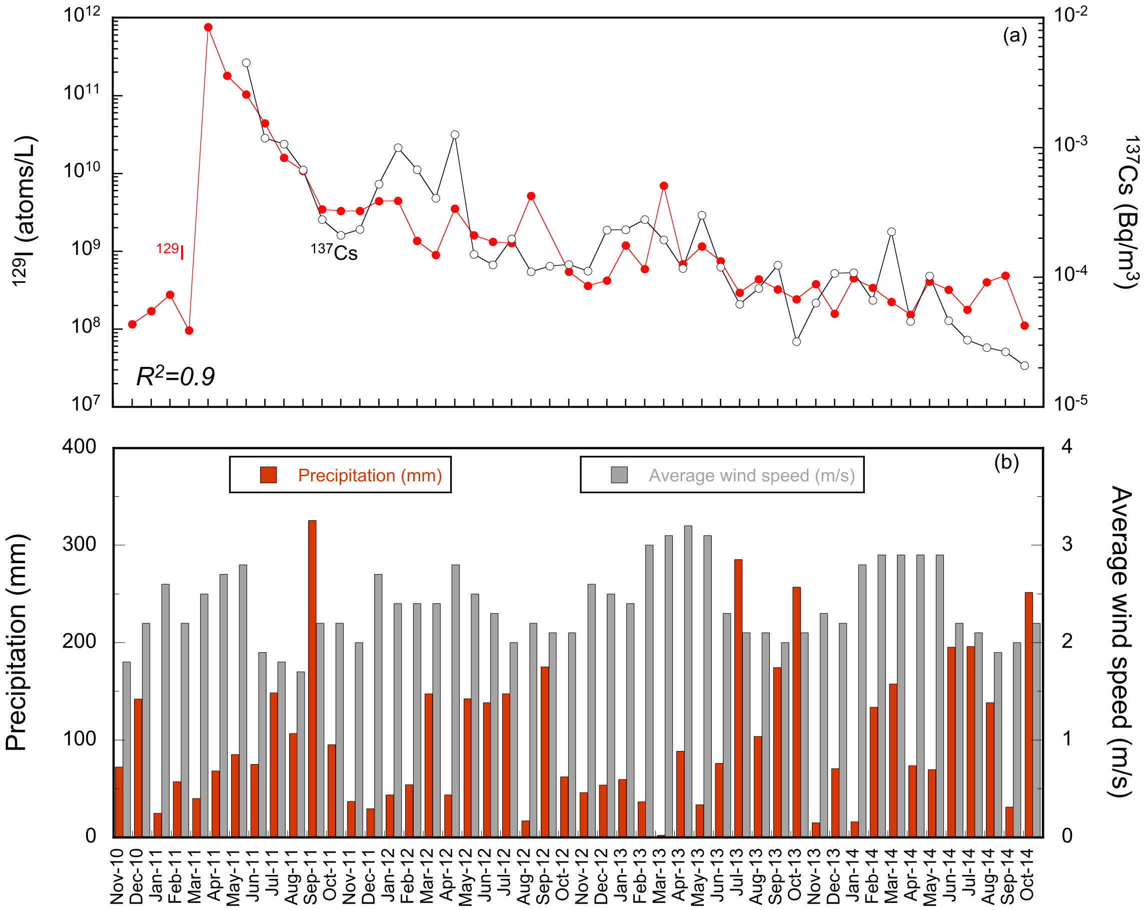

Fig. 3. (a) Comparison of temporal variation of ^{129}I in rainwaters and ^{137}Cs in aerosols from Fukushima. The monthly ^{137}Cs data were averaged from the measured values of every 3 d (Watanabe, 2015). (b) variation of average monthly precipitation and wind speed in Fukushima City (data sources: http://www.jma.go.jp/jma/index.html).

collected in the same site. Overall, there is positive correlation between atmospheric ^{129}I and ^{137}Cs with correlation coefficient $R^2 = 0.9$ (Fig. 3a). The high correlation should be attributed to the same source of ^{129}I in the precipitation and ^{134}Cs in aerosols. A significantly amount of radioiodine and radiocesium have been released to the atmosphere during Fukushima accident because of high volatility of both iodine and cesium, causing a tremendously increased level of ^{131}I and ^{137}Cs in the atmosphere after the Fukushima accident. A relative constant ^{129}I/^{137}Cs activity ratio of ~4 × 10^{-7} from the FDNPP have been reported (Tumey et al., 2013; Xu et al., 2015). The high coincident of ^{129}I in the precipitation and ^{137}Cs in the aerosols also indicates their similar speed of removal from the atmosphere. This might be partly attributed to that ^{137}Cs and some fractions of ^{129}I present in particle associated form, which can be removed from the atmosphere in the same process. Iodine in the gas form, especially in inorganic form can be also quickly removed from the atmosphere with a similar speed as the particle associated forms.

There are several ^{129}I and ^{137}Cs pulses shown in Fig. 3a. It mainly occurred in winter season (e.g., December 2011 to February 2012, December 2012 to March 2013, December 2013 to March 2014), and spring-summer seasons (e.g., April 2012, June 2012, May 2013, May 2014). Fig. 3b illustrates the monthly precipitation rate and wind speed. In comparison with meteorological factors, the radionuclides have weakly negative correlation with precipitation in 2012–2014. The relationship between radionuclides and average wind speed is not as visible as that between radionuclide and precipitation, which is probably due to frequent changes of wind directions. The similar patterns were neither seen between atmospheric radionuclides and the maximum or maximum instantaneous wind speed. Nevertheless, it can be found that high wind speed and low precipitation roughly coincides with the high ^{129}I concentration in precipitations and ^{137}Cs concentrations in aerosols. Therefore, it can be concluded that the dry and windy weather conditions favour the resuspension of the deposited Fukushima-derived radionuclides on land surfaces.

It has been indicated that the Fukushima-derived ^{129}I and ^{137}Cs deposited in the top surface soils are mainly bound to humus substances and clay minerals (i.e., Ohta et al., 2012; Tanaka et al., 2012; Saito et al., 2014). Furthermore, analysis of size distribution showed that both ^{129}I and ^{137}Cs dominate in fine particles (μm scale) in soils. Therefore, in dry and windy climatic conditions which are typical of winter in Fukushima (Fig. 3b), such fine soil particles can be easily suspended into the atmosphere unless large amounts of snow covers the land. This is particularly demonstrated in the rainwater sample collected in March 2013 in which both very high ^{127}I and ^{129}I are associated with the lowest precipitation and high wind speeds (Fig. 3b). In addition, dust-bearing radionuclides can be deposited on the surface of leaves of plants. When they fall to the ground and degrade in winter, such dust can also be suspended into the atmosphere. Local biomass burning, which occurs in November–December in the eastern Japan plain, is also considered to be a potential source of suspended ^{129}I and ^{137}Cs, following processed which have been recognized for Chernobyl fallout (Lujanienė et al., 2009). These observations can explain the elevated ^{129}I and ^{137}Cs concentrations observed in the winter season.

In case of the elevated radionuclides in spring-summer seasons, prevalence of biological activity might play an important role in the re-emission of ^{129}I from vegetation. For example, high radiocesium concentration has been reported in pollen of Japanese cedar, the most popular evergreen tree in Japan (MAFF, 2014). Kanasashi et al. (2015) demonstrated uptake and translocation of significant components of the intercepted activity by investigating radiocesium concentrations in Japanese cedar needles from pre-2010, 2011 and 2012. They have also shown similar radiocesium concentrations in cedar pollen and flowers from 2012, which suggests that seasonal pollen emission may be linked to the seasonal remobilization pulses observed in this study. Therefore, such radiocesium can be released to the atmosphere with pollen in the early spring season. On the other hand, iodine isotope trace experiments have revealed that iodine volatilization from plants is a pathway for radioiodine re-emission into the atmosphere from a ^{129}I-contaminated area (Amiro and Johnston, 1989; Muramatsu and Yoshida, 1995). Although iodine emissions from living vegetation contributes ~0.1% to the ^{127}I concentration in the atmosphere above terrestrial areas (Amiro and Johnston, 1989), this pathway was thought to be a direct route for ^{129}I transport from contaminated soils to the atmosphere (Muramatsu and Yoshida, 1995). As a result, the Fukushima-derived ^{129}I can be released as gaseous form through vegetation that takes up ^{129}I from contaminated soil and water.

4.3. Long-term variation of atmospheric ^{129}I

As described above, the average ^{129}I concentrations in 2011, 2012, 2013 and 2014 are approximately 1×10^{11}, 2×10^9, 6×10^8 and 3×10^8 atoms/L, respectively. These result in ^{129}I decline rates of 98%, 71% and 49% between 2011 and 2012, 2012 and 2013, and 2013 and 2014, respectively. Similar decline rates are also observed for ^{137}Cs activities in aerosols (Watanabe, 2015).

In addition, temporal ^{129}I variation can be further assessed by analysis of exponential function fits. We have examined models of the single exponential, the single exponential plus constant, and double exponential functions, but found that they gave poor fits to the whole data set. This suggests the complex decay across the whole period, and seems to indicate that the exchange mechanisms between environmental subsystems were still undergoing dynamic changes over the study period. The observation that ^{129}I activity concentrations are now approaching pre-accident levels may argue for inclusion of a term to represent non-Fukushima-derived ^{129}I, however at this stage we do not have sufficient data to indicate whether or not this is well represented by a constant term, or whether it will also show seasonal fluctuations. Nevertheless, we have chosen to use single exponential fits in each individual year in order to demonstrate that the exponential decay constant decreases each year in a systematic manner. With the exception of two samples (August 2012 and March 2013), the fitting results for 2011, 2012, 2013 and 2014, respectively are expressed as follows.

$$^{129}I = (7.53 \pm 0.22) \times 10^{11}\ e^{(-2.3\pm0.9)t},\ R^2 = 0.9$$

$$^{129}I = (3.35 \pm 0.75) \times 10^{9}\ e^{(-0.19\pm0.02)t},\ R^2 = 0.5$$

$$^{129}I = (1.07 \pm 0.17) \times 10^{9}\ e^{(-0.12\pm0.04)t},\ R^2 = 0.6$$

$$^{129}I = (3.17 \pm 0.14) \times 10^{8}\ e^{(-0.02\pm0.05)t},\ R^2 = 0.01$$

where ^{129}I is concentration in precipitation (atoms/L) and t is time (month). The ^{129}I decline rate can be simply expressed by the decay constant in the equations due to very low ^{129}I radioactive decay constant (4.4×10^{-8} y^{-1}) and therefore are 28, 2.3, 1.4 and 0.2 y^{-1} in 2011, 2012, 2013 and 2014, respectively. These values indicate the decrease of the ^{129}I declining rate from 2011 to 2014. Poor exponential fitting observed in 2014 ($R^2 = 0.01$) suggests that variation of ^{129}I in precipitation likely overlaps the range of natural variability, which implies that contribution of the Fukushima-derived ^{129}I to the atmosphere would become less since 2014.

The total amounts of suspended materials from the local surfaces might change little every year. However, radionuclides in the surface soil and forest/vegetation might gradually decrease since the accident, probably due to processes such as downward migration, resuspension and dispersion of fine particles from surface soils and forests (MAFF, 2014; Matsuda et al., 2015; Takahashi et al., 2015). Mikami et al. (2015) have constructed distribution maps of the inventories of ^{134}Cs and ^{137}Cs deposited onto ground soil using portable gamma-ray spectrometers at nearly 1000 locations within 80 km from FDNPP in March, September and December 2012. No apparent temporal change of the radionuclide inventories was observed from March to December 2012. Matsuda et al. (2015) measured ^{137}Cs activity in soil profiles in 84–85 locations with a 100 km radius from FDNPP during three periods from December 201 to December 2012. These results suggest that radioactive cesium has moved downwards in these soils, but remained within 5 cm of the ground surface at most study sites. Takahashi et al. (2015) investigated ^{137}Cs in soil profiles under eight different land uses in July 2011, January 2012, August 2012 and December 2012. There is no clear trend observed in depth profile on any site – probably due to spatial variability not being picked up in single measurements. Honda et al. (2015) examined the variation of depth profiles from the same upland field (Kawauchi village, 20 km southwest of the FDNPP) with an interval of one year (April 2012, October 2011, December 2011 and March 2012). The downward migration rates for ^{129}I and ^{137}Cs during the interval were estimated to be 0.81 ± 0.32 and 0.19 ± 0.17 g/cm^2/yr, respectively. Furthermore, considering factors such as post-accident decontamination activities (i.e., removal of topmost 5 cm soils) carried out by local government, post-accident agricultural activities, and the quick downward migration which can be enhanced during the rainfall events, activity concentrations of Fukushima-derived radionuclides in the topmost soils are expected to be progressively lower. As a result, these observations suggest the gradual decreases of the ^{129}I and ^{137}Cs concentrations in the topmost soils from 2011 to 2014.

It should be pointed out that variations of ^{129}I and ^{137}Cs are not very well synchronous in 2013–2014, particularly demonstrated by the amplitude of variation. This might be attributed to the difference of baseline of ^{129}I and ^{137}Cs level in Fukushima. The atmospheric ^{137}Cs activity has been reported to be <1 μBq/m^3 in Tsukuba (Igarashi et al., 1999). This value is in good accord with the reported data in the mid-latitudes of the Northern Hemisphere. The level of ^{137}Cs in Tsukuba could be regarded as a typical value for the mid-latitudes of the Northern Hemisphere in the 1990s (Igarashi et al., 1999). This ^{137}Cs level in the air seems to be resulted from resuspension of the global fallout from atmospheric nuclear bomb tests, etc. and can be regarded as the background of atmospheric ^{137}Cs in Japan. In comparison with this background level, the Fukushima atmospheric ^{137}Cs concentrations in 2013–2014 (10–100 μBq/m^3) are still 10–100 times higher. In contrast, large amounts of ^{129}I discharged from the nuclear reprocessing facilities in Europe have resulted in largely elevated ^{129}I in global atmosphere (Aldahan et al., 2009), although the present ^{137}Cs concentration in the European air is <1 μBq/m^3 (i.e., Nielsen et al., 2015). This added ^{129}I flux has caused enhancement of baseline of global ^{129}I level (i.e., ^{129}I/^{127}I>10^{-7} in Denmark, Hou et al., 2009). As a result, the Fukushima ^{129}I concentrations in 2013–2014 are only up to four times higher than the pre-accident level, except for one sample in March 2013, which shows typical resuspension of local Fukushima-derived ^{129}I, and natural ^{127}I in land surface. Thus, as a result of higher background level of ^{129}I in the atmosphere, with the gradually decreasing of Fukushima-derived ^{129}I in the suspended matter, effect of Fukushima-derived ^{129}I might become less contribution to the present ^{129}I level in respective with the ^{137}Cs since 2014.

5. Conclusions

Variations of iodine isotopes in four-year time series rainwater collected in Fukushima, Japan over 2010–2014 reveal that Fukushima-derived ^{129}I has gradually declined from 7.6×10^{11} atoms/L in March 2011 immediately after the accident to $(1–4) \times 10^8$ atoms/L in 2014, having intermittently approached the pre-accident level $(1–3) \times 10^8$ atoms/L. The atmospheric ^{129}I and ^{137}Cs showed roughly a season variation, high values in winter and low in autumn. Enhancement of ^{129}I during the studied period can be mainly attributed to resuspension of the Fukushima-derived radionuclide bearing fine particles deposited on land surfaces under the winter dry and windy climatic conditions, and re-emission through vegetation taking up ^{129}I from contaminated soil and water in spring-summer. Long-term declining rate suggests that contribution of the Fukushima-derived ^{129}I to the atmosphere would become less since 2014.

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