



Regional and global contributions of anthropogenic iodine-129 in monthly deposition samples collected in North East Japan between 2006 and 2015



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ABSTRACT

We measured the monthly atmospheric deposition flux of ¹²⁹I at Rokkasho, Aomori, Japan—the location of a commercial spent nuclear fuel reprocessing plant—from 2006 to 2015 to assess the impact of the plant on environmental ¹²⁹I levels. The plant is now under final safety assessment by a national authority after test operation using actual spent nuclear fuel. During cutting and chemical processing in test operations from April 2006 to October 2008, ¹²⁹I was discharged to the atmosphere and detected in our deposition samples. ¹²⁹I deposition fluxes largely followed the discharge pattern of ¹²⁹I from the plant to the atmosphere, and most of the deposited ¹²⁹I originated from the plant. In and after 2009, ¹²⁹I deposition fluxes decreased dramatically to reach the background level; the ¹²⁹I deposition fluxes at Rokkasho were almost the same as those at Hirosaki, where an additional sampling point was set up as a background site 85 km from the plant in 2011. The background ¹²⁹I deposition fluxes showed seasonal variation—high in winter and low in the other seasons—at both Rokkasho and Hirosaki. The results of a backward trajectory analysis of the air mass at Rokkasho suggested that reprocessing plants in Europe were the origins of the high ¹²⁹I flux in winter. The contribution of ¹²⁹I released from the Fukushima Dai-ichi Nuclear Power Plant accident to the ¹²⁹I deposition flux at Rokkasho in 2011 was small on the basis of the ¹²⁹I/¹³¹I activity ratio.

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1. Introduction

Iodine-129, which has a long half-life of 1.57×10^7 years, is produced naturally via the interaction of cosmic rays with Xe in the atmosphere; spontaneous fission of ²³⁸U and ²³⁵U in the lithosphere (Eissenbud and Gesell, 1997). The atomic ratio of ¹²⁹I/¹²⁷I resulting from these productions range from 6×10^{-13} to 1×10^{-12} (Fabryka-Martin et al., 1985; Killius et al., 1992). The natural inventory of ¹²⁹I has been estimated to be ~1.5 TBq, with most of it occurring in the deep oceans (Rao and Fehn, 1999).

In addition to ¹²⁹I occurring naturally, anthropogenic ¹²⁹I has been produced by fission through civil and military nuclear activities, including accidents such as those at the Chernobyl Nuclear

Power Plant in April 1986 and the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) in March 2011. Nuclear weapons testing, mainly in the 1960s, dispersed ¹²⁹I at 0.3 to 1.0 TBq into the atmosphere (Raisbeck et al., 1995; Wagner et al., 1996); a further ~40 GBq was released in the Chernobyl accident (Gallagher et al., 2005) and 8 GBq in the FDNPP accident (Hou et al., 2013).

Discharge from nuclear fuel reprocessing plants is now the most important contributor of ¹²⁹I to the environmental inventory. The total amount of ¹²⁹I now discharged from two major European commercial reprocessing facilities—the Magnox and Thorp reprocessing plants at Sellafield in the United Kingdom and the AREVA La Hague plant in France—now accounts for >90% of the global inventory (Aldahan et al., 2007). In an inventory of ¹²⁹I discharged from reprocessing plants, Michel et al. (2012) showed that the total atmospheric and marine discharges up to 2005 were 0.9 and 9.0 TBq, respectively, from the Sellafield plants and 0.45 TBq and 21.8 TBq from the La Hague plant. The latest annual releases of ¹²⁹I to the atmosphere were reported as 11 GBq (as radioiodine)

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from the Sellafield plants in 2015 (Sellafield Ltd, 2016) and 5.8 GBq from the La Hague plant in 2015 (AREVA, 2016). There are limited reports about ^{129}I discharge from military reprocessing plants. Plutonium production at the Hanford site (USA) released 1.7 TBq into the atmosphere between 1944 and 1972, and the operation of production reactors at the Savannah River site (USA) also released 0.21 TBq into the atmosphere from 1953 until about 1990 (Hu and Moran, 2011). Those releases have increased the background $^{129}\text{I}/^{127}\text{I}$ atomic ratio in the general environment to 1×10^{-10} to 1×10^{-8} (Handl et al., 1993).

Japan's first commercial spent nuclear fuel reprocessing plant, operated by Japan Nuclear Fuel Limited (JNFL) in Rokkasho, in Aomori Prefecture, is now under safety assessment by a national authority before it starts full operation. From March 2006, when final testing of the plant with actual spent nuclear fuels was started, until October 2008, spent fuel rods containing 425 t of U were cut and chemically processed as part of the testing. Iodine-129 was discharged to the atmosphere from April 2006 to April 2009 (referred to hereafter as the gaseous discharge period) with occasional breaks, resulting in a total discharge of 740 MBq.

Iodine forms various compounds (e.g., I_2 , HI, HOI, CH_3I)—including water soluble and volatile ones—and can easily enter hydrological and atmospheric cycles. Iodine-129 discharged from nuclear facilities has been detected not only around the facilities but far away from them. Toyama et al. (2012) reported that the atmospheric fallout collected in Tokyo after 1979 was contributed to by atmospheric ^{129}I discharge from the Tokai Reprocessing Plant located about 130 km northeast of Tokyo. Herod et al. (2015) found a pulse of ^{129}I derived from the FDNPP accident in precipitation in Vancouver, Canada, 6 days after the accident. Therefore, ^{129}I is considered to be a powerful tracer in both local and global environments.

Here, we collected atmospheric deposition samples in the village of Rokkasho from 2006 to 2015 and analyzed their ^{129}I concentration and $^{129}\text{I}/^{127}\text{I}$ atomic ratio to evaluate the influence of ^{129}I discharged from the JNFL reprocessing plant during the gaseous discharge period. To clarify the contribution of secondary local sources of ^{129}I , such as resuspended soil, after the gaseous discharge period, we set up another sampling site in the city of Hirosaki, 85 km southwest of the plant, and we collected deposition samples from 2011 to 2015. We continuously collected monthly atmospheric deposition samples over an experimental period of 9 years at Rokkasho and 4 years at Hirosaki. To the best of our knowledge, this is the first paper to report the continuous monthly deposition flux of ^{129}I in the Asian region for a period of more than 5 years. The results from both sites are presented in this report. We also discuss the effects of the FDNPP accident and reprocessing at the JNFL plant, as well as the mechanism of seasonal variation in background values.

2. Materials and methods

2.1. Study site

Atmospheric deposition samples were collected at the Institute for Environmental Sciences (IES; $40^\circ 57' \text{ N}$, $141^\circ 21' \text{ E}$) in the village of Rokkasho, in Aomori Prefecture, Japan, and at Hirosaki University ($40^\circ 35' \text{ N}$, $140^\circ 28' \text{ E}$) in the city of Hirosaki, in Aomori Prefecture (Fig. 1). The IES is located approximately 2.6 km east of the main stack of the JNFL reprocessing plant, whereas Hirosaki University is located approximately 85 km southwest of the main stack.

In Aomori Prefecture, a northwestern monsoon from the Asian continent blows from winter to spring, whereas high-pressure systems developed in the Pacific Ocean in summer bring air masses from the ocean. As a result, either easterly or westerly

winds prevail throughout the year (Kawabata et al., 2002; Nibe, 1989). In Rokkasho, the mean annual temperature from 2011 to 2015 was $10.0 \pm 0.4^\circ \text{C}$; over the same period the mean accumulated precipitation was $1363 \pm 89 \text{ mm}$, and the maximum snow depth $88 \pm 39 \text{ cm}$. In Hirosaki the equivalent values were $10.5 \pm 0.5^\circ \text{C}$, $1343 \pm 264 \text{ mm}$, and $117 \pm 32 \text{ cm}$.

2.2. Sample collection and measurement methods

Bulk atmospheric deposition samples, including dry deposition, were collected for approximately 1 month (every 24–39 d) by using a deposition sampler with an open surface area of 0.0314 m^2 (ST-1F, Suntecno Co. Ltd., Toyama, Japan) from April 2006 till October 2015 at the Rokkasho site, and from April 2011 till October 2015 at the Hirosaki site. The sampler has a funnel with a deicing heater and a sampling bottle. The funnel output was connected to the bottle with a silicone tubing. A 50 g aliquot of tetramethyl ammonium hydroxide (TMAH) was poured into the sampling bottle at the beginning of every sampling period to prevent loss of iodine during the sampling period. The addition of TMAH was decided by a preliminary experiment comparing ^{127}I concentrations in total deposition samples collected by using simple bucket type samplers (27 cm ϕ) with 0.25% TMAH solution or only pure water. Three samples for each TMAH solution and pure water groups have been collected 1, 3, 4, 5 and 6 d after the beginning of collection and analyzed for ^{127}I . The mean ^{127}I concentration ratio of (pure water)/(TMAH solution) were gradually decreased with time from 0.89 ± 0.02 at 1 d to 0.59 ± 0.06 at 6 d (\pm standard deviation; detailed data are not shown). We concluded that iodine in the bucket type collector was not kept in the pure water. For safety sake, TMAH was also used in the present collector, ST-1F.

Iodine was extracted from the sample with toluene in accordance with the method of Schmidt et al. (1998). An 80 mL aliquot of sample was transferred to a separation funnel, and 2 mg of an iodine carrier in iodide form (Woodward Iodine Co., OK, USA) was added. After toluene (30 mL) had been added to the sample solution, the aqueous phase was acidified to pH 1 with nitric acid to convert iodate to iodide. Then, an aliquot of 4 mL 10% NaNO_2 solution was added to oxidize the iodide to I_2 , which was extracted to the toluene phase. Extraction was performed twice, and the toluene phases were combined. Iodine in the combined toluene phase was back-extracted to 21 mL 0.02% NaHSO_3 solution and then 10 mL water. The back-extracted iodide was precipitated as AgI by the addition of 1.5 mL 30% NH_3 solution and 1 mL of 1 M AgNO_3 . The precipitated AgI was dried with Nb powder and pressed into a cathode target as the ion source. Iodine-129 in the sample was measured by using an accelerator mass spectrometry (with an upgraded HVEC model FN-8MV) at the PRIME Laboratory (Purdue University). The lowest raw $^{129}\text{I}/^{127}\text{I}$ in the measurement sample in this study was $3.9 \pm 0.4 \times 10^{-13}$ and approximately eight times higher than our procedure blank value with the Woodward iodine. Stable iodine was analyzed by using inductively coupled plasma mass spectrometry (Agilent-7700, Agilent Technologies Inc., USA).

3. Results and discussion

The deposition flux of ^{129}I and the $^{129}\text{I}/^{127}\text{I}$ atomic ratios in atmospheric deposition samples collected at Rokkasho from April 2006 to October 2015 are shown in Table S1 in the supplementary materials, together with the ^{129}I and ^{127}I concentrations in precipitation and the cumulative precipitation during each sampling period. Data for samples collected at Hirosaki from April 2011 to October 2015 are given in Table S2.

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