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Temporal variation of ^{134}Cs and ^{137}Cs activities in surface water at stations along the coastline near the Fukushima Dai-ichi Nuclear Power Plant accident site, Japan

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We present our April to December 2011 observations of ^{134}Cs and ^{137}Cs activities in surface water at Hasaki, a coastal station 180 km south of the Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident site. We also investigate trends by using published data from several other coastal stations, including the accident site. The maximum in radiocaesium activity at Hasaki was observed in June 2011, representing a delay of two months from the corresponding maximum in April 2011 at FNPP1. Directly discharged ^{134}Cs and ^{137}Cs were transported dominantly southward along the coastline of northeastern Honshu, at least in May and June 2011. The reasons for the two-month delay at Hasaki are not yet clear, but clockwise current associated with a warm water eddy of which center located at 36.5 N, 141.4 E off Iwaki between Onahama and Hasaki in mid of May 2011 might prevent southward transport of ^{134}Cs and ^{137}Cs released from FNPP1 to Hasaki until the end of May 2011.

Keywords: radiocaesium, ocean, coast, Fukushima, geochemistry

INTRODUCTION

On 11 March 2011, an extraordinary earthquake of magnitude 9.0 occurred centred about 130 km off the Pacific coast of Honshu, Japan's main island, at 38.3°N, 142.4°E. It was followed by a huge tsunami with waves reaching up to 40 m height in the Iwate region and about 10 m in the Fukushima region (The 2011 Tohoku Earthquake Tsunami Joint Survey Group, 2011; Mori *et al.*, 2011). These events caused great loss of life (about 16000 confirmed dead and about 4000 missing) and extensive damage. One of the consequences was a total loss of AC electric power at the Fukushima Dai-ichi Nuclear Power Plant (hereafter FNPP1). The station blackout developed into a disaster that left three of the six FNPP1 reactors heavily damaged and caused radionuclides to be discharged into the atmosphere and ocean (Chino *et al.*, 2011; Tsumune *et al.*, 2011).

In this paper, we focus on the trend of ^{134}Cs and ^{137}Cs radioactivities at Hasaki, a coastal station on the east coast of Honshu, and other coastal stations, including the FNPP1 site. Because these stations are located in the densely populated Tohoku and Kanto areas of Honshu, the behaviour of ^{134}Cs and ^{137}Cs (radiocaesium) in coastal waters is important for understanding the fate of ^{134}Cs and ^{137}Cs in the environment. These radionuclides were released from the FNPP1 reactors and directly discharged into the ocean and released into the atmosphere; other minor contributions may have arisen from riverine outflow carrying sediment from contaminated lands. Radiocaesium is a serious concern for people involved in coastal fisheries and seafood safety.

METHODS

We collected 2-litre surface seawater samples once a week at the Hazaki Oceanographical Research Station of the Port and Airport Research Institute, (station Hasaki,

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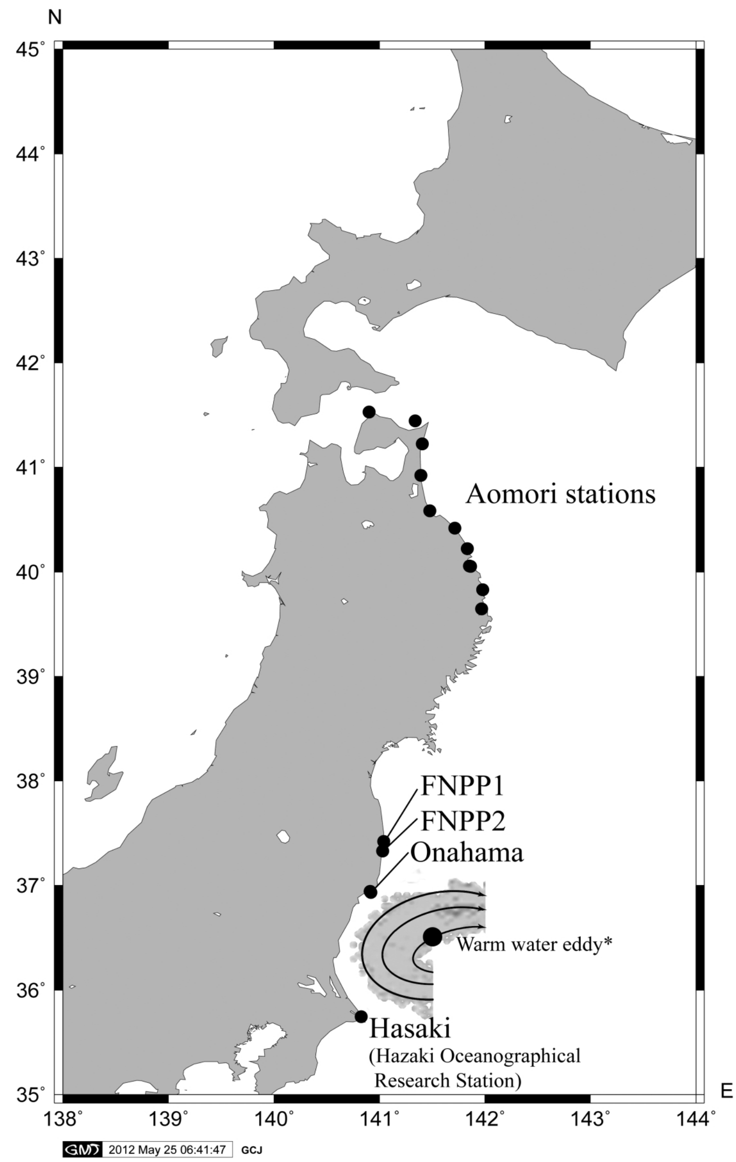

*Fig. 1. Sampling locations for ^{137}Cs and ^{134}Cs in surface water. *Center of warm water eddy in mid of May 2011 (see detail in the main text).*

35.84°N, 140.76°E) from 25 April 2011 to 5 December 2011. The samples were treated as described by Aoyama and Hirose (2008), and their activities were measured at the underground facility of the Low Level Radioactivity Laboratory of Kanazawa University using ultra low background Ge-detectors (Hamajima and Komura, 2004). We also used online data published by Tokyo Electric Power Company (TEPCO) and Fukushima Prefecture for coastal stations including FNPP1, Fukushima Dai-ni Nuclear Power Plant (FNPP2) and Onahama as shown in Fig. 1. The data from TEPCO and Fukushima Prefecture were not shown with the analytical uncertainty, but the MEXT guidelines state that "not detected" is defined when counting error exceeds one-third of the measured activity. We therefore assumed that the uncertainty of the published data is less than 33%, and we display these data with a 33% error bar.

Results and Discussion

^{134}Cs activity in surface water at Hasaki was around 40–110 Bq m^{-3} until the end of May 2011, thereafter it suddenly increased and reached 2080 ± 150 Bq m^{-3} on 6 June 2011 (Table 1 and arrow in Fig. 2). Then ^{134}Cs ac-

Table 1. Temporal variation of radiocaesium activity at Hasaki

Sample ID	Year	Month	Day	^{134}Cs Bq m^{-3}			^{137}Cs Bq m^{-3}		
HA-001	2011	4	25	107	±	8	114	±	6
HA-003	2011	5	9	66	±	5	77	±	4
HA-005	2011	5	23	41	±	3	54	±	3
HA-007	2011	6	6	1880	±	130	2020	±	130
HA-008	2011	6	13	2080	±	150	2290	±	120
HA-009	2011	6	20	1590	±	110	1760	±	90
HA-010	2011	6	27	447	±	35	538	±	30
HA-011	2011	7	4	1120	±	70	1290	±	70
HA-012	2011	7	11	935	±	65	1070	±	60
HA-013	2011	7	18	709	±	47	844	±	43
HA-014	2011	7	25	507	±	38	603	±	33
HA-015	2011	8	1	468	±	31	541	±	27
HA-016	2011	8	8	397	±	32	455	±	26
HA-017	2011	8	15	323	±	21	402	±	20
HA-019	2011	8	29	132	±	9	163	±	9
HA-020	2011	9	5	74	±	6	94	±	5
HA-021	2011	9	12	114	±	8	143	±	8
HA-022	2011	9	22	147	±	11	186	±	10
HA-023	2011	9	26	71	±	5	90	±	5
HA-025	2011	10	10	102	±	7	126	±	7
HA-027	2011	10	24	58	±	4	76	±	4
HA-029	2011	11	7	52	±	4	76	±	4
HA-031	2011	11	21	46	±	3	63	±	3
HA-033	2011	12	5	44	±	4	57	±	3

Radioactive decay was corrected at a time of collection. One sigma of counting error is shown in this table.

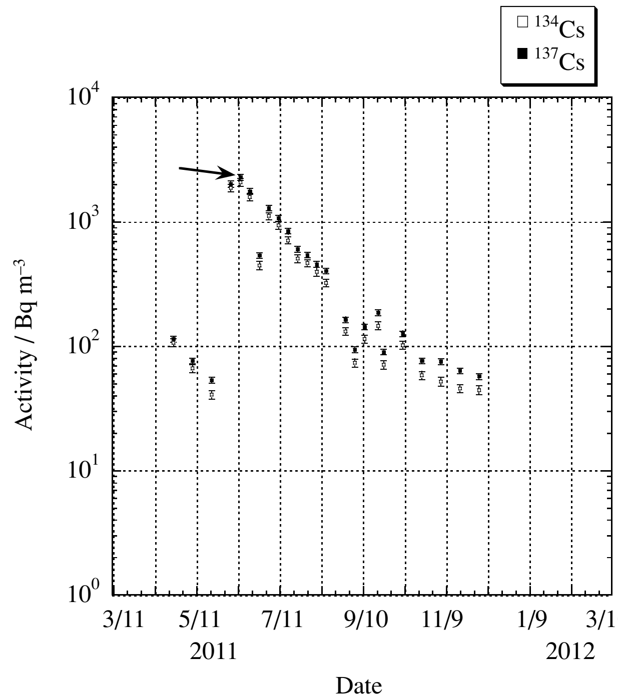

Fig. 2. Temporal variations of ^{137}Cs and ^{134}Cs at Hasaki.

tivity decreased steadily with an apparent half-residence time of about 21 days until the end of August 2011. After that ^{134}Cs activity decreased with an apparent half-residence time of about 60 days until December 2011, at which time ^{134}Cs activity in surface water was 40–50 Bq m^{-3}.

^{137}Cs activity showed the same trend as ^{134}Cs (Fig. 2). Before the FNPP1 accident, ^{137}Cs activity in the surface water of the western North Pacific Ocean was around 1–2 Bq m^{-3} (Aoyama *et al.*, 2008, 2011), therefore ^{137}Cs activity in June 2011 was 1000 times higher than that before the FNPP1 accident.

^{134}Cs/^{137}Cs activity ratios observed at Hasaki were close to 1 (Fig. 3) when we take into account the ^{134}Cs half-life of 2.1 years. This ratio is consistent with the ^{134}Cs/^{137}Cs activity ratio of 0.99 ± 0.02 observed at FNPP1 in March and April 2011 (Buesseler *et al.*, 2011) and in the western North Pacific in April and May 2011 (Honda *et al.*, 2012). This ratio also indicates that the ^{134}Cs and ^{137}Cs observed at Hasaki originated from the FNPP1 accident.

The sudden increase of radiocaesium activity in surface water at Hasaki on 6 June 2011 came two months after the maximum of radiocaesium activity in surface water at FNPP1, which was observed on 6 April 2011 (upper arrow in Fig. 4). At Onahama, 30 km south of FNPP1 (Fig. 1), a delayed maximum of ^{134}Cs activity in surface water was also observed at the beginning of June 2011 (lower arrow in Fig. 4) as was the case at Hasaki. In contrast, Inoue *et al.* (2011) showed little increase of ^{134}Cs activity in surface water north of Fukushima at ten coastal stations, shown as "Aomori" in Fig. 4. The ^{134}Cs activity in May and June 2011 at these stations north of FNPP1 was only a few Bq m^{-3}, and three orders of magnitude lower than those observations at Hasaki and Onahama south of FNPP1.

The trend of radiocaesium activity in surface water north and south of FNPP1 might be regulated by characteristics of direct discharge of ^{134}Cs and ^{137}Cs at the FNPP1 site, transport processes in the coastal zone, and characteristics of atmospheric deposition of ^{134}Cs and ^{137}Cs released from FNPP1 into the atmosphere. The main source of the ^{134}Cs and ^{137}Cs measured at these coastal stations was the variable flux of radiocaesium from FNPP 1; Buesseler *et al.* (2011) reported a peak in ocean discharge in early April, one month after the earthquake, and a decrease by a factor of 1000 by the following month. Concentrations through the end of July at FNPP1 remained several orders of magnitude higher than levels in coastal waters measured in 2010, implying continuing releases from the reactors or other sources (Buesseler *et al.*, 2011). ^{134}Cs activity in surface water at the FNPP1 and FNPP2 sites remained at 10^3–10^4 Bq m^{-3} in December 2011 (Fig. 4). ^{134}Cs activity at Hasaki (Fig. 2) was about two orders of magnitude lower than that at FNPP1

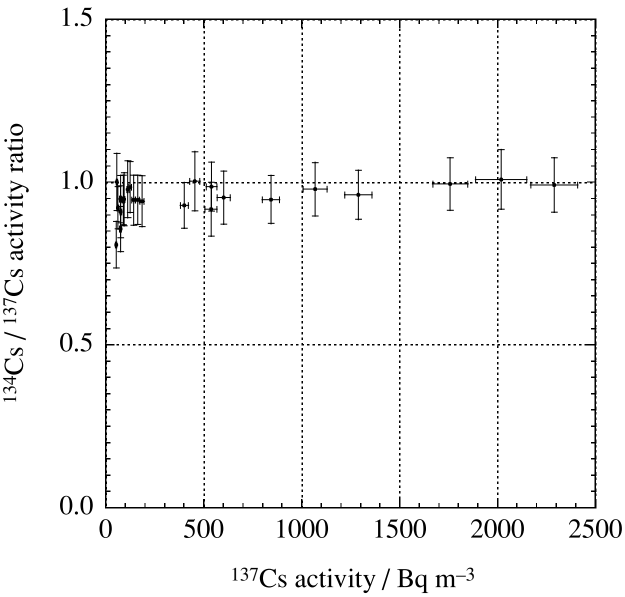

Fig. 3. ^{134}Cs/^{137}Cs activity ratio vs. ^{137}Cs activity at Hasaki. Radioactive decay is corrected on 11 March 2011.

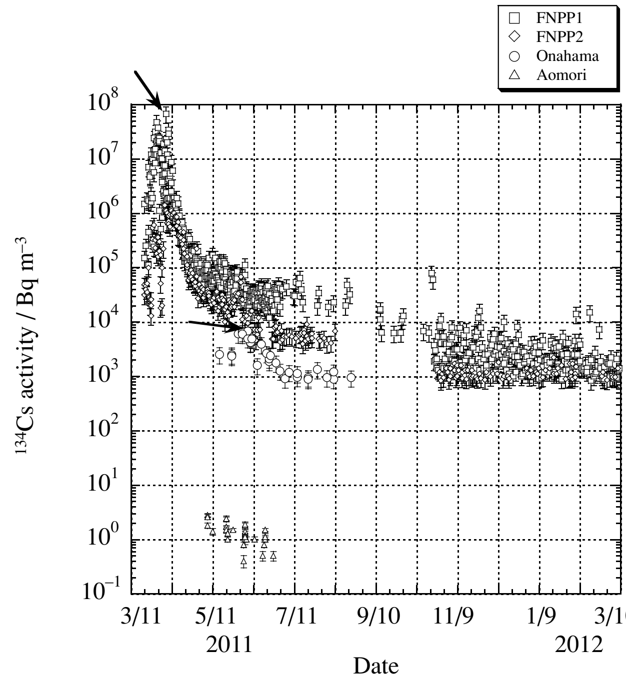

Fig. 4. Temporal variations of ^{134}Cs at the FNPP 1 and FNPP2 sites, Onahama, and stations "Aomori".

and FNPP2 from June 2011 to December 2011 and similar to that at Onahama, whereas ^{134}Cs activity in Aomori stations was four orders of magnitude lower than that at FNPP1 and FNPP2. This pronounced difference in activity to the south and north of FNPP1 shows that transport of directly discharged ^{134}Cs and ^{137}Cs was dominantly southward, at least in May and June 2011 off north eastern Honshu.

Sudden increase of ^{134}Cs and ^{137}Cs activities in surface water at Hasaki occurred between 23 May 2011 and 6 June 2011 as shown in Fig. 2 and Table 1. Before this sudden change, ^{134}Cs and ^{137}Cs activities at Onahama already exceeded 2000 Bq m^{-3} while those at Hasaki were only around 50–100 Bq m^{-3} in mid of May 2011 (Table 1; Figs. 2 and 4). This indicates that southward transport of ^{134}Cs and ^{137}Cs released from FNPP1 to Hasaki were relatively limited rather than southward transport to Oanahama until the end of May 2011. Coast transport processes are very complex in this sea area and these might be controlled by Kuroshio, meso-scale eddies associated with Kuroshio and fresh water flux from land. Therefore, it is interesting and important to discuss about the sudden increase of ^{134}Cs and ^{137}Cs activities at Hasaki regarding with hydrographic conditions near the coast of this region. In fact, there was a warm water eddy of which center located at 36.5 N, 141.4 E off Iwaki between Onahama and Hasaki in the middle of May 2011 as shown in Fig. 1 (http://www.data.kishou.go.jp/kaiyou/db/tokyo/archive/2011/05_2/tokyo_current/tokyo_current.html). Clockwise current associated with this warm water eddy, which means northward current east of this warm water eddy, might be able to prevent southward transport of ^{134}Cs and ^{137}Cs as we stated previously. This warm water eddy disappeared on 30 May 2011 (http://www.data.kishou.go.jp/kaiyou/db/tokyo/archive/2011/05_3/tokyo_current/tokyo_current.html) and as a consequence sudden increase of ^{134}Cs and ^{137}Cs activities in surface water at Hasaki and Onahama were observed. Just before these changes, a difference of ^{134}Cs and ^{137}Cs activities in surface water between Hasaki and FNPP1 was five hundred times or more. After the sudden changes, the difference of ^{134}Cs and ^{137}Cs activities in surface water between both stations decreased to only 30 times indicating that increased southward transport reaching at Hasaki made less activity difference between these two stations. We also can see small decrease of ^{134}Cs and ^{137}Cs activities in surface water at FNPP1 and FNPP2 between end of May and beginning of June 2011 as shown in Fig. 4. These small decreases occurring at the similar period with increases of Hasaki and Onahama may also indicate enhanced southward transport as discussed above, if we can assume less change on ^{134}Cs and ^{137}Cs fluxes at FNPP1.

In general, radiocaesium was transported to the south, then to the east after it was released directly from the FNPP1 site as already described by a model simulation study (Tsumune *et al.*, 2011). It might be also necessary to conduct more detailed model simulations that include coastal processes such as meso-scale eddy behavior and freshwater flux from land to simulate a sudden increase of ^{134}Cs and ^{137}Cs activity in the surface water observed at Hasaki on 6 June 2011.

Conclusion

We compared our results at Hasaki to published radiocaesium trends at the FNPP1 site and several coastal stations to its south and north. The maximum in radiocaesium activity at Hasaki was observed in June 2011 representing a delay of two months from the corresponding maximum in April 2011 at FNPP1. Directly discharged ^{134}Cs and ^{137}Cs was transported dominantly southward along the coastline of northeastern Honshu, at least in May and June 2011. The reasons for the two-month delay at Hasaki and Onahama are not yet clear, but clockwise current associated with this warm water eddy of which center located at 36.5 N, 141.4 E off Iwaki between Onahama and Hasaki in mid of May 2011 might prevent southward transport of ^{134}Cs and ^{137}Cs released from FNPP1 to Hasaki until the end of May 2011.

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