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ARTICLE

Fukushima radionuclides in the marine environment from coastal region of Japan to the Pacific Ocean through the end of 2016

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The authors present total amount of released radiocaesium from FNPP1 accident which satisfy mass balance in all components of the environment. The authors also present consensus values of Fukushima Dai-ichi Nuclear Power Plant (FNPP1) derived ^{137}Cs in the environment. Total amount of atmospheric release was 15-20 PBq and direct discharge to the ocean was 3-6 PBq, respectively. 12-15 PBq was deposited in the North Pacific and 3-6 PBq was deposited on land, respectively. Therefore 15-18 PBq was injected as sum of atmospheric deposition and direct discharge in the North Pacific Ocean. The authors also present temporal variation of ^{137}Cs in coastal region close to FNPP1 site. Based on radiocaesium activity concentration and ^{134}Cs/^{137}Cs activity ratios decay corrected to 11 March 2011 in surface water, major source of radiocaesium is still FNPP1 site and contribution from global fallout ^{137}Cs increased at Tomioka and Hasaki depending on the distances from FNPP1 site. A part of radiocaesium in the Sub Tropical Mode Water (STMW) in the Pacific Ocean is already obducted from ocean interior and enter into northern part of East China sea and in Japan Sea.

Keywords: Fukushima accident; radiocaesium; mass balance; inventory; North Pacific

1. Introduction

On 11 March 2011, an extraordinary earthquake of magnitude 9.0 centered about 130 km off the Pacific coast of Japan's main island, at 38.3ºN, 142.4ºE, was followed by a huge tsunami with waves reaching up to 40 m height in Iwate region and about 10 m in Fukushima region [1]. These events caused the loss of about 16000 lives, missing of about 4000 lives and extensive damage. One of the consequences was a station blackout (total loss of AC electric power) at the Fukushima Dai-ichi Nuclear Power Plant (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 air and ocean [2].

Total amount of release radionuclides to the environment from the FNPP1 accident in March 2011 might be one of the big concerns about the FNPP1 accident as well as impact to human and non-human biota. In this paper, 1) we summarize total amount of released radiocaesium from the FNPP1 accident and mass balance of FNPP1 derived radiocaesium in the North Pacific Ocean and 2) present latest status of radiocaecium in the coastal region in Japan.

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2. Total amount of released radiocaesium from the FNPP1 accident and mass balance of FNPP1 derived radiocaesium in the North Pacific Ocean

Observed ^{134}Cs to ^{137}Cs activity ratio at the time of accident was close to 1 (0.99 ± 0.03 in the FNPP1 north and south discharge channels) and extremely uniform [3], and ^{134}Cs to ^{137}Cs activity ratio in the estimated core inventory was also close to 1 [4], therefore in this article we assume that the total amount of ^{134}Cs released to the atmosphere and that of direct discharge to the ocean can be considered the same as the total amount of ^{137}Cs released to the atmosphere and that of direct discharge to the ocean. Total deposition of ^{134}Cs from the atmosphere on ocean surface, deposition of ^{134}Cs from the atmosphere on Japanese land and total amount of ^{134}Cs in the North Pacific Ocean also can be considered the same as those of ^{137}Cs.

Estimates of the released amount of the most widely studied radionuclide, ^{137}Cs, have varied from 3 to 30 PBq [5]. One of causes of the large variability is that most of the researchers did not consider mass balance of radiocaesium, the law of conservation of mass, in the environment. Many researchers did estimate only a part of the environment such as only to the air, only to the ocean etc. Mass balance of radiocaesium is a strong constrain to understand environmental impact of FNPP1 released radionuclides as below,

$$\Sigma R_i = \Sigma I_j \quad (1)$$

where R_i are released amount to each domain and I_j are inventory in each domain and
i: 1=atmosphere, 2=direct discharge
j: 1=atmosphere (now zero due to short residence time), 2= land, 3=ocean, 4=sediment (known as small), 5= biota (negligible)

$$R_1+R_2=I_1+I_2+I_3+I_4+I_5=<\text{total in the core} \quad (2)$$

We can also ignore I_1 due to short residence time of radionuclides in the air around a few weeks. We can ignore I_4 because small amount radiocaesium in the sediment as reported to be around 130 ± 60 TBq [6,7].We also can ignore I_5 because maximum estimate of radiocaesium in biota based on fish catch amount of 20×10^6 kg around Fukushima and assuming an activity of 1×10^4 Bq kg^{-1} give us order around GBq. Finally we can simplify the equation (2) as below;

$$R_1+R_2=I_2+I_3 \quad (3)$$

$$R_1-I_2=I_3-R_2 \quad (4)$$

Therefore, major portion of mass balance of FNPP1 derived radiocaesium is that sum of atmospheric release and direct discharge to the ocean, R_1+R_2, must be very close to sum of inventories on land and in the ocean, I_2+I_3, as shown in Eq. (3). Tsumune et al. reported that total amount of direct discharge to the ocean, R_2, was 3.5 ± 0.7 PBq [8]. Aoyama et al. reported based on the mass balance as shown Eqs. (3) and (4) and observed inventory in the North Pacific Ocean that atmospheric release, R_1, was 15.2-20.4 PBq and total amount of radiocaesium deposited in the North Pacific Ocean, I_3-R_2, was 11.7-14.8 PBq [9]. Total amount of deposited radiocaesium on land, I_2, was estimated to be 3.4-6.2 PBq. Finally as a total 15.2-18.3 PBq, I_3, was estimated to be discharged to the North Pacific [9]. These discharges amount from Fukushima accident represent about 22-27 % of the total input of 69 PBq delivered to the North Pacific in the 1950s and 1960s from atmospheric nuclear weapons tests [9].

The inventory of FNPP1-released ^{134}Cs in the North Pacific Ocean, I_3, was also estimated to be 15.3 ± 2.6 PBq [10] based on observational value and by Optimal Interpolation (OI) analysis which showed in good agreement with an estimate above. About half of this activity (8.4 ± 2.6 PBq) was found in the coastal region near the FNPP1 at the time of accident [10]. Based on a linear least-squares regression between simulated and observed ^{134}Cs activity, the total amount of ^{134}Cs injected into the North Pacific, I_3, was estimated at 16.1 ± 1.4 PBq [11]. This estimation also showed in good agreement with an estimate of 15.2-18.3 PBq [9] and an estimate of 15.3 ± 2.6 PBq [10].

In the ocean interior, radiocaesium maxima were observed at mode waters which indicate that subduction is main transport process of Fukushima derived radicaesium into the ocean interior. In the North Pacific Ocean 7.9 ± 1.4 PBq is in surface layer (Inomata unpublished data) and 4.2 ± 1.1 PBq is in Sub Tropical Mode Water (STMW) and FNPP1 derived radiocaesium was spread entire subtropical gyre in 2012 [12].

Therefore we propose consensus values of Fukushima derived radiocaseum in the environments as below combining results with other researchers.
1) R_1:Atmospheric release: 15-20 PBq [13-16]
2) R_2:Direct discharge: 3-6 PBq [17-19]
3) Atmospheric deposition in the North Pacific: 12-15 PBq [11]
4) I_2:Total on land: 3-6 PBq [9]
5) I_3:Total in the North Pacific Ocean: 15-18 PBq [11,20]
6) I_4:In the sediment: 130 ± 60 TBq [6,7]

As shown above, total amount of atmospheric release was 15-20 PBq and direct discharge to the ocean was 3-6 PBq, respectively. 12-15 PBq, 80 % of atmospheric release, was deposited in the North Pacific and 3-6 PBq was deposited on land, respectively. Therefore 15-18 PBq was injected as sum of atmospheric deposition and direct discharge in the North Pacific Ocean.

3. Dynamics of FNPP1 derived radiocaesium

3.1. Sampling and methods

We conducted enhanced surface water sampling at 84 stations in coastal regions on both the Japan Sea and Pacific Ocean sides of Japan from November 2015 to March 2016 to examine the recirculation behaviour of FNPP1-derived radiocaesium in the surface layer 5 years after the 2011 FNPP1 accident. We have also been collecting coastal water samples, basically once per month, at two coastal stations: Tomioka, Fukushima, Japan (37.335º N, 141.031ºE) since June 2014 and Hasaki, Ibaraki, Japan (35.842ºN, 140.763ºE) since April 2011. Sample volumes ranged from a few litres to 10 L, and radiocaesium in the sample seawater was extracted by an improved ammonium phosphomolybdate (AMP)/Cs compound method [21]. The AMP/Cs compound was measured by using ultra-low-background gamma-ray detectors at the Low Level Radioactivity Laboratory, Kanazawa University [22], because the total amount of ^{134}Cs in each sample was expected to be only a few mBq and the amount of ^{137}Cs was expected to range from a few to 20 mBq.

With this improved AMP procedure, the weight yield of AMP/Cs compound, as well as the radiochemical yield of radiocaesium, generally exceeds 95% for less than 100 L samples [23]. Because the reagents used to extract radiocaesium from seawater can contain trace levels of ^{134}Cs and ^{137}Cs and we were using small-volume samples for the measurements, we needed to know the specific activity of ^{134}Cs and ^{137}Cs in the reagents. The ^{137}Cs activity in the AMP, which we synthesized ourselves, was 0.024 mBq g^{-1}; thus, we subtracted from our results the expected amount of ^{137}Cs

in the AMP used (4 to 6 g) to extract radiocaesium from the samples. The ^{137}Cs activity in the CsCl reagent was measured as 0.03 mBq g^{-1} by extremely low background ɣ-spectrometry at the Ogoya underground laboratory of Low level radioactivity laboratory, Kanazawa University. We can neglect this ^{137}Cs contribution in the CsCl reagent because we used only 0.26 g of CsCl as a carrier for 4 g of AMP, or 0.39 g of CsCl as a carrier for 6 g of AMP. No other reagents were seriously contaminated with ^{137}Cs produced before the accident. We did not detect ^{134}Cs contamination in any of the reagents, which were produced before the FNPP1 accident.

3.2. Temporal variation in east coast of northern Honshu Island

Temporal variation of radiocaesium activity at Hasaki, Ibaraki, Japan, 35.842ºN, 140.763ºE is shown in **Table 1** since April 2011 until June 2016. Temporal variation of radiocaesium activity at Tomioka, Fukushima, Japan, 37.335º N, 141.031ºE is also shown in **Table 2** since June 2014 until June 2016. In addition to our own data as shown in Tables 1 and 2, we compiled monitoring data from TEPCO to examine ^{137}Cs and ^{134}Cs activity trends in the surface water at the FNPP1 accident site (http://www.tepco.co.jp/nu/fukushima-np/f1/smp/2015/images/seawater-newest01-j.csv).

Figure 1 shows activities of ^{137}Cs in surface water at North 56 canal of FNPP1 until 26 December 2016, at Tomioka until 30 June 2016 and at Hasaki until 13 June 2016. Analyses of the ^{137}Cs activities and ^{131}I/^{137}Cs activity ratios suggest that the major direct release of ^{137}Cs from the FNPP1 reactors occurred during the 12 days from 26 March 2011 to 6 April 2011 (15 to 26 days after the earthquake) [24]. After that, the amount released decreased considerably although radiocaesium continued to be released about a few GBq day^{-1} until at least December 2016 because the ^{137}Cs activity at North 56 canal of FNPP1 still around 100 Bq m^{-3} (Figure 1)

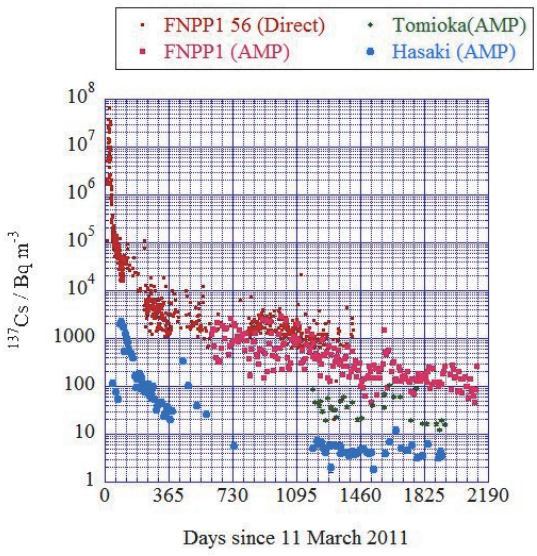

Figure 1. Activities of ^{137}Cs in surface water at North 56 canal of FNPP1, Tomioka and Hasaki.

and previous coastal model simulation results [25].

The ^{137}Cs activity observed at Hasaki in 2011 ranged from 32.1 ± 1.7 to 2290 ± 120 Bq m^{-3} and was much higher than that before the FNPP1 accident. The ^{137}Cs activity in surface water at Hasaki was 50-110 Bq m^{-3} until the end of May 2011 (81 days after the earthquake), and it suddenly increased to a maximum of 2080 ± 150 Bq m^{-3} on 13 June 2011 (94 days after the earthquake). Then, ^{137}Cs activity decreased steadily with an apparent half-residence time of about 21 days until the end of August 2011 (173 days after the earthquake). Subsequently, ^{137}Cs activity decreased with an apparent half-residence time of about 60 days until December 2011 (295 days after the earthquake), at which time ^{137}Cs activity in the surface water was 32-98 Bq m^{-3} [26]. A second increase at Hasaki occurred between 26 March and 28 May in 2012. The ^{137}Cs activity in surface water at Hasaki was 20-36 Bq m^{-3} from 5 March to 26 March (360 to 381 days after the earthquake), and it increased to a maximum of 347 ± 16 Bq m^{-3} on 28 May 2012 (444 days after the earthquake). Then ^{137}Cs activity decreased steadily and was 5.7 ± 0.3 Bq m^{-3} on 19 March 2013 (739 days after the earthquake) [26] From 9 June 2014 to 13 June 2016 (1186 to 1921 days after the earthquake), ^{137}Cs activity at Hasaki ranged from 1.9 ± 0.1 to 12.1 ± 0.7 Bq m^{-3} and generally did not decreased. Comparison of the ^{137}Cs activities near the FNPP1 site and at Hasaki (Figure 1) implies that the ^{137}Cs activity observed at Hasaki might have originated from the ongoing leakage of radiocaesium from the FNPP1 site and from global ^{137}Cs fallout, and a small amount might have originated from recirculated seawater which was contaminated just after the accident.

At Tomioka, the ^{137}Cs activity in the surface water ranged from 12.5 ± 0.7 to 105.4 ± 5.6 Bq m^{-3} from 9 June 2014 to 30 June 2016 (1186 to 1938 days after the earthquake) (Figure 1). Because Tomioka is situated between the FNPP1 site and Hasaki and the coastal current generally flows southward in this region [26], radiocaesium activities at Tomioka were between those at the FNPP1 site and those at Hasaki during the period of this study (Figure 1).

^{134}Cs/^{137}Cs activity ratios decay corrected to 11 March 2011 in surface water at North 56 canal of FNPP1 close to the site were around 1 throughout the period as shown in **Figure 2**. And activity of ^{137}Cs in surface water at North 56 canal of FNPP1 is still highest [28]. This means that source of radiocaesium observed at North 56 canal of FNPP1 is contaminated water within the FNPP1 site. At Tomioka ^{134}Cs/^{137}Cs activity ratios decay corrected to 11 March 2011 in surface water ranged from 0.85 ± 0.09 to 1.10 ± 0.12 as shown Figure 2. These activity ratios indicate that most of the radiocaesium observed at Tomioka might be originated from FNPP1 site itself. At Hasaki, ^{134}Cs/^{137}Cs activity ratios decay corrected to 11 March 2011 in surface water ranged from 0.81 ± 0.07 to 1.08 ± 0.08 until October 2012, then during the period from June 2014 to June 2016 it ranged from 0.46 ± 0.19 to 1.10 ± 0.12. The contribution of global fallout ^{137}Cs to radiocaesium activity

Table 1. Temporal variation of radiocaesium activity at Hasaki, Ibaraki, Japan, 35.842ºN, 140.763ºE. Uncertainty shows one sigma of counting error plus uncertainty of calibration standard. Radioactivities in this table are decay corrected at the time of sampling.

date	^{134}Cs Bq m^{-3}	^{137}Cs Bq m^{-3}	data sources
20110425	107 ± 8	114 ± 6	a
20110509	66 ± 5	77 ± 4	a
20110523	41 ± 3	54 ± 3	a
20110606	1880 ± 130	2020 ± 130	a
20110613	2080 ± 150	2290 ± 120	a
20110620	1590 ± 110	1760 ± 90	a
20110627	447 ± 35	537 ± 30	a
20110704	1120 ± 70	1290 ± 70	a
20110711	935 ± 65	1070 ± 60	a
20110718	709 ± 47	844 ± 43	a
20110725	494 ± 37	588 ± 32	a
20110801	468 ± 31	541 ± 27	a
20110808	397 ± 32	455 ± 26	a
20110815	323 ± 21	402 ± 20	a
20110922	147 ± 11	186 ± 10	a
20110829	132 ± 9	163 ± 9	a
20110905	74 ± 6	93.6 ± 5.1	a
20110912	114 ± 8	143 ± 8	a
20110922	147 ± 11	186 ± 10	a
20110926	71 ± 5	90.0 ± 4.9	a
20111003	138 ± 7	155 ± 7	b
20111010	102 ± 7	126 ± 7	a
20111017	80.4 ± 4.6	90.1 ± 4.6	b
20111024	58.0 ± 4.0	76.3 ± 4.2	a
20111031	91.9 ± 5.0	105 ± 5	b
20111101	89.3 ± 50.3	101 ± 5	b
20111107	50.6 ± 4.0	73.6 ± 4.1	a
20111114	93.7 ± 5.2	113 ± 6	b
20111121	46.0 ± 3.0	63.4 ± 3.4	a
20111128	65.8 ± 3.6	76.1 ± 3.7	b
20111205	43.3 ± 3.5	55.8 ± 3.2	a
20111212	82.5 ± 4.6	98.1 ± 4.9	b
20111219	41.4 ± 2.9	54.6 ± 2.9	b
20111226	25.0 ± 1.7	32.1 ± 1.7	b
20120103	27.9 ± 2.1	40.8 ± 2.2	b
20120110	31.5 ± 2.4	41.5 ± 2.3	b
20120116	32.6 ± 2.4	47.5 ± 2.5	b
20120123	31.7 ± 2.3	45.1 ± 2.4	b
20120130	32.0 ± 2.3	46.1 ± 2.5	b
20120206	15.8 ± 1.1	23.8 ± 1.3	b
20120213	16.7 ± 1.4	26.0 ± 1.5	b
2010220	17.7 ± 1.3	27.7 ± 1.4	b
20120227	25.2 ± 2.0	38.5 ± 2.1	b
20120305	26.7 ± 2.0	36.2 ± 2.0	b
20120312	22.3 ± 1.6	32.0 ± 1.7	b
20120319	11.7 ± 1.1	20.1 ± 1.2	b
20120326	19.6 ± 1.4	31.6 ± 1.7	b
20120528	246 ± 13	347 ± 16	b
20120625	75.6 ± 4.2	107 ± 5	b
20120813	26.0 ± 1.7	40.0 ± 3.0	b
20121009	18.3 ± 1.4	26.3 ± 1.4	b
20130319	2.9 ± 0.3	5.7 ± 0.3	b
20140609	1.4 ± 0.2	5.1 ± 0.3	b
20140623	2.1 ± 0.3	7.4 ± 0.4	b
20140714	1.7 ± 0.2	6.4 ± 0.4	b
20140728	1.9 ± 0.2	6.9 ± 0.4	b
20140824	1.4 ± 0.3	4.2 ± 0.3	b
20140922	ND ± NA	2.0 ± 0.4	b
20141005	1.5 ± 0.2	5.9 ± 0.3	b
20141104	1.5 ± 0.2	5.8 ± 0.4	b
20141116	1.2 ± 0.2	5.6 ± 0.3	b
20141214	0.5 ± 0.2	3.6 ± 0.2	b
20150119	0.7 ± 0.2	4.5 ± 0.3	b
20150202	1.0 ± 0.2	3.7 ± 0.3	b
20150216	0.9 ± 0.2	3.9 ± 0.3	b
20150302	1.0 ± 0.2	4.7 ± 0.3	b
20150316	1.1 ± 0.2	4.9 ± 0.3	c
20150330	1.0 ± 0.2	4.9 ± 0.3	c
20150413	0.8 ± 0.3	4.1 ± 0.3	c
20150427	0.7 ± 0.2	4.0 ± 0.3	c
20150511	0.9 ± 0.2	4.2 ± 0.3	c
20150525	0.4 ± 0.1	1.9 ± 0.1	c
20150727	0.5 ± 0.2	4.0 ± 0.3	c
20150824	1.8 ± 0.2	7.1 ± 0.4	c
20150928	3.3 ± 0.4	12.1 ± 0.7	c
20151026	0.9 ± 0.2	5.1 ± 0.3	c
20151130	0.7 ± 0.1	4.6 ± 0.3	c
20151228	1.0 ± 0.2	6.0 ± 0.3	c
20160125	0.8 ± 0.1	3.3 ± 0.2	c
20160229	0.5 ± 0.1	3.6 ± 0.2	c
20160328	1.1 ± 0.1	6.3 ± 0.4	c
20160530	0.5 ± 0.1	3.3 ± 0.2	c
20160606	0.7 ± 0.1	4.4 ± 0.3	c
20160613	0.5 ± 0.1	3.6 ± 0.2	c

a: Aoyama et al., 2012 [26].
b: Aoyama et al., 2016 [27].
c: This study.

Table 2. Temporal variation of radiocaesium activity at Tomioka, Fukushima, Japan, 37.335º N, 141.031ºE. Uncertainty shows one sigma of counting error plus uncertainty of calibration standard. Radioactivities in this table are decay corrected at the time of sampling.

Date	^{134}Cs Bq m^{-3}	^{137}Cs Bq m^{-3}	data sources
20140610	30.8 ± 2.1	87.8 ± 4.6	a
20140623	14.6 ± 1.2	45.7 ± 2.4	a
20140714	12.6 ± 1.1	39.0 ± 2.1	a
20140722	16.9 ± 1.3	48.5 ± 2.6	a
20140806	10.0 ± 1.0	30.1 ± 1.7	a
20140821	6.7 ± 0.6	19.5 ± 1.1	a
20140902	17.4 ± 1.6	55.9 ± 3.0	a
20140918	12.4 ± 1.9	37.0 ± 2.7	a
20140918	11.8 ± 1.2	34.4 ± 1.9	a
20141003	10.2 ± 1.0	32.2 ± 1.8	a
20141013	11.3 ± 1.1	32.9 ± 1.8	a
20141021	6.5 ± 0.6	22.7 ± 1.2	a
20141112	13.2 ± 1.2	48.8 ± 2.6	a
20141202	19.4 ± 1.6	61.0 ± 3.2	a
20141224	9.9 ± 1.0	37.0 ± 2.0	a
20150119	12.7 ± 1.1	46.9 ± 2.5	a
20150217	9.3 ± 0.9	20.2 ± 1.2	a
20150317	6.2 ± 0.6	21.9 ± 1.2	a
20150519	10.2 ± 1.1	40.0 ± 2.2	b
20150724	7.8 ± 0.7	35.3 ± 1.9	b
20150724	18.5 ± 1.4	68.7 ± 3.6	b
20150724	17.8 ± 1.8	67.9 ± 3.8	b
20150824	25.4 ± 2.2	105.4 ± 5.6	b
20150915	16.2 ± 1.6	60.4 ± 3.3	b
20151020	14.3 ± 1.2	60.5 ± 3.2	b
20151222	3.7 ± 0.4	19.4 ± 1.1	b
20160119	18.1 ± 1.5	89.1 ± 4.7	b
20160224	3.4 ± 0.4	16.9 ± 0.9	b
20160324	3.0 ± 0.4	16.4 ± 0.9	b
20160516	2.9 ± 0.3	16.3 ± 0.9	b
20160602	2.3 ± 0.3	12.5 ± 0.7	b
20160614	3.3 ± 0.4	19.3 ± 1.1	b
20160630	3.1 ± 0.4	16.0 ± 0.9	b

a: Aoyama et al., 2016 [27].
b: This study.

observed at Hasaki varied much during the period from June 2014 to June 2016 due to change of coastal current [26].

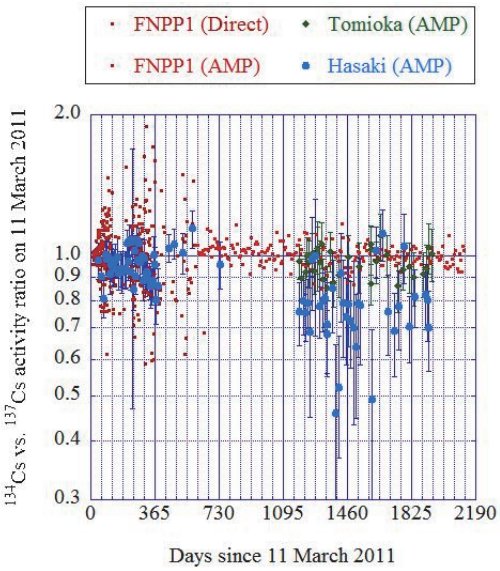

Figure 2. $^{134}Cs/^{137}Cs$ activity ratio in surface water at North 56 canal of FNPP1, Tomioka and Hasaki. Activity ratio was decay corrected to 11 March 2011.

3.3. Eastward surface transport

From the end of March to early April 2011, extremely high activities were observed in the coastal surface seawater near the FNPP1. The high activities spread to a region near 165°E in the western North Pacific Ocean, with a latitudinal center of 40°N. Atmospheric deposition also caused high activities in the region between 180° and 130°W in the North Pacific Ocean [10].

In summer 2012, the radiocaesium derived from the Fukushima Dai-ichi Nuclear Power Plant accident in March 2011 was found in the Bering Sea and western North Pacific between 25°N and 63°N, which agrees with model simulation results of atmospheric deposition. A semi-synoptic view suggests that a main body of radiocaesium discharged directly had been transported eastward to 170°W, northward to 50°N, and southward to 30°N by summer 2012, about one and half years after the accident [29]. The eastward speed was estimated to be 7 km day^{-1} [30].

3.4. Re-circulation

A part of radiocaesium in the STMW is already obducted from ocean interior and re-circulated [28]. ^{137}Cs activities in surface water during the period from Nov. 2015 to May 2016 are shown in **Figure 3**. Higher ^{137}Cs activities in surface water ranged from 2.0 to 2.2 Bq m^{-3} were observed in the northern part of East China Sea, west of Kyushu Islands and Tsushima straight. On the other hands, ^{137}Cs activities in surface water in the southern part of the East China Sea were relatively low compared with those in the northern part of East China Sea and ranged from 1.2 to 1.8 Bq m^{-3} . This indicate that a source of relatively higher ^{137}Cs activities in surface water observed during the period from Nov. 2015 to May 2016 might be ocean interior in the Pacific side and obducted seawater of which ^{137}Cs activities are relatively high in the STMW [12-31] rather than surface water.

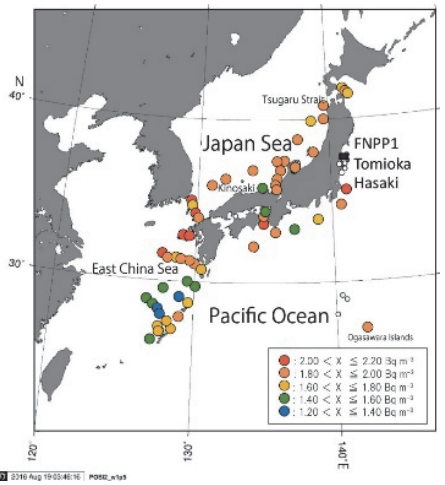

Figure 3. ^{137}Cs activities in surface water during the period from Nov. 2015 to May 2016.

4. Conclusion

Total amount of release of radiocaesium in each domain were reliably determined as consensus by our results and combining many researchers results as 15-20 PBq for atmospheric release, 3-6 PBq for direct discharge to the ocean, respectively. 12-15 PBq was deposited in the North Pacific and 3-6 PBq was deposited on land, respectively. Therefore 15-18 PBq was injected as sum of atmospheric deposition and direct discharge in the North Pacific Ocean. These values well satisfied the mass balance of Fukushima derived radiocaesium in the environment.

In the coastal region close to FNPP1 site, based on radiocaesium activity and $^{134}Cs/^{137}Cs$ activity ratios decay corrected to 11 March 2011 in surface water revealed that major source of radiocaesium is still FNPP1 site and contribution from global fallout ^{137}Cs increased at Tomioka and Hasaki depending on the distances from FNPP1 site.

A part of radiocaesium in the STMW in the Pacific Ocean is already obducted from ocean interior and enter into northern part of East China sea re-circulated in Japan Sea.

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References

[1] The-2011-Tohoku-Earthquake-Tsunami-Joint-Survey-Group. Nationwide Field Survey of the 2011 Off the Pacific Coast of Tohoku Earthquake Tsunami, J. J. Soc.Civil Eng. , Ser B2 (Coastal Engineering). 67, (2011), pp. 63-66.

[2] P. P. Povinec, K. Hirose and M. Aoyama, *Fukushima Accident: Radioactivity Impact on the Environment*, Elsevier (2013), pp. 1-400.

[3] K. Buesseler, M. Aoyama and M. Fukasawa, Impacts of the Fukushima nuclear power plants on marine radioactivity, *Environ. Sci. Technol.* 45 (2011), pp. 9931-9935.

[4] K. Nishihara, H. Iwamoto and K. Suyama, Estimation of Fuel Compositions in Fukushima-Daiichi Nuclear Power Plant, *JAEA-Data/Code* 18 (2012), pp. 1-190.

[5] K. Buesseler, M. Dai, M. Aoyama, C. Benitez-Nelson, S. Charmasson, K. Higley, V. Maderich, P. Masque, P. J. Morris, D. Oughton and J. N. Smith, Fukushima Daiichi-Derived Radionuclides in the Ocean: Transport, Fate, and Impacts, *Ann. Rev. Mar. Sci.* 9 (2017), pp. 173-203.

[6] M. Kusakabe, S. Oikawa, H. Takata and J. Misonoo, Spatiotemporal distributions of Fukushima-derived radionuclides in nearby marine surface sediments, *Biogeosciences 10* (2013), pp.5019-5030.

[7] E. E. Black and K. O. Buesseler, Spatial variability and the fate of cesium in coastal sediments near Fukushima, Japan, *Biogeosciences* 11 (2014), pp.5123-5137.

[8] D. Tsumune, T. Tsubono, M. Aoyama and K. Hirose, Distribution of oceanic ^{137}Cs from the Fukushima Dai-ichi Nuclear Power Plant simulated numerically by a regional ocean model, *J. Environ. Radioact.* 11 *(*2012), pp. 100-108.

[9] M. Aoyama, M. Kajino, T.Y. Tanaka, T.T. Sekiyama, D. Tsumune, T. Tsubono, Y. Hamajima, Y. Inomata and T. Gamo, ^{134}Cs and ^{137}Cs in the North Pacific Ocean derived from the March 2011 TEPCO Fukushima Dai-ichi Nuclear Power Plant accident, Japan. Part two: estimation of ^{134}Cs and ^{137}Cs inventories in the North Pacific Ocean, *J. Oceano.* 72 (2016), pp. 67-76.

[10] Y. Inomata, M. Aoyama, T. Tsubono, D. Tsumune and K. Hirose, Spatial and temporal distributions of ^{134}Cs and ^{137}Cs derived from the TEPCO Fukushima Daiichi Nuclear Power Plant accident in the North Pacific Ocean by using optimal interpolation analysis, *Environ. Sci. Processes & Impacts* 18 (2016), pp. 126-136.

[11] T. Tsubono, K. Misumi, D. Tsumune, F. O. Bryan, K. Hirose and M. Aoyama, Evaluation of radioactive cesium impact from atmospheric deposition and direct release fluxes into the North Pacific from the Fukushima Daiichi nuclear power plant, *Deep Sea Res. Part I: Oceano. Res. Papers* 115 (2016) pp. 10-21.

[12] H. Kaeriyama, Y. Shimizu, T. Setou, Y. Kumamoto, M. Okazaki, D. Ambe and T. Ono, Intrusion of Fukushima-derived radiocaesium into subsurface water due to formation of mode waters in the North Pacific, *Sci. Rep.* 6 (2016), pp. 22010, DOI: 10.1038/srep22010.

[13] A. Mathieu, I. Korsakissok, D. Quelo, J. Groell, M. Tombette, D. Didier, E. Quentric, O. Saunier, J.P. Benoit and O. Isnard, Atmospheric Dispersion and Deposition of Radionuclides from the Fukushima Daiichi Nuclear Power Plant Accident, *Elements* 8 (2012), pp. 195-200.

[14] V. Winiarek, M. Bocquet, N. Duhanyan, Y. Roustan, O. Saunier and A. Mathieu, Estimation of the caesium-137 source term from the Fukushima Daiichi nuclear power plant using a consistent joint assimilation of air concentration and deposition observations, *Atmos. Environ.* 82 (2014), pp. 268-279.

[15] O. Saunier, A. Mathieu, D. Didier, M. Tombette, D. Quélo, V. Winiarek and M. Bocquet, An inverse modeling method to assess the source term of the Fukushima nuclear power plant accident using gamma dose rate observations, *Atmos. Chem. Phys.* 13 (2013), pp. 11403-11421.

[16] G. Katata, M. Chino, T. Kobayashi, H. Terada, M. Ota, H. Nagai, M. Kajino, R. Draxler, M. Hort and A. Malo, Detailed source term estimation of the atmospheric release for the Fukushima Daiichi Nuclear Power Station accident by coupling simulations of an atmospheric dispersion model with an improved deposition scheme and oceanic dispersion model, *Atmos. Chem. Phys.* 15 (2015), pp. 1029-1070.

[17] H. Kawamura, T. Kobayashi, A. Furuno, T. In, Y. Ishikawa, T. Nakayama, S. Shima and T. Awaji, Preliminary numerical experiments on oceanic dispersion of ^{131}I and ^{137}Cs discharged into the ocean because of the Fukushima Daiichi Nuclear Power Plant Disaster, *J. Nucl. Sci. Technol.* 48 (2011), pp. 1349-1356.

[18] C. Estournel, E. Bosc, M. Bocquet, C. Ulses, P. Marsaleix, V. Winiarek, I. Osvath, C. Nguyen, T. Duhaut, F. Lyard, H. Michaud and F. Auclair, Assessment of the amount of cesium-137 released into the Pacific Ocean after the Fukushima accident and analysis of its dispersion in Japanese coastal waters, *J. Geophys. Res.* 117 (2012), pp. C11014.

[19] Y. Miyazawa, Y. Masumoto, S. Varlamov, T. Miyama, M. Takigawa, M. Honda and T. Saino, Inverse estimation of source parameters of oceanic radioactivity dispersion models associated with the Fukushima accident, *Biogeosciences* 10 (2013), pp. 2349-2363.

[20] Y. Inomata, M. Aoyama, T. Tsubono, D. Tsumune and K. Hirose, Spatial and temporal distributions of ^{134}Cs and ^{137}Cs derived from the TEPCO Fukushima Daiichi Nuclear Power Plant accident in the North Pacific Ocean by using optimal interpolation analysis, *Environ. Sci.: Processes & Impacts* 18 (2016), pp.126-136.

[21] M. Aoyama and K. Hirose, Radiometric determination of anthropogenic radionuclides in seawater. In *Radiometric Determination of Anthropogenic Radionuclides in Seawater*, (Ed. P.P. Pavel), Elsevier, Hungary, (2008), pp.137-162.

[22] Y. Hamajima and K. Komura, Background components of Ge detectors in Ogoya underground laboratory, *Appl. Radiat. Isot.* 61 (2004), pp.179-183.

[23] M. Aoyama, *Guidline of Ocean Observations 9 Natural and Artificial Radioactivity*, The Oceanographic Society of Japan, Tokyo, (2018), pp. G901EN:001-013.

[24] D. Tsumune, M. Aoyama, K. Hirose, F. O. Bryan, K. Lindsay and G. Danabasoglu, Transport of ^{137}Cs to the Southern Hemisphere in an ocean general circulation model, *Prog. In Oceanog.* 89 (2011), pp.38-48.

[25] D. Tsumune, T. Tsubono, M. Aoyama, M. Uematsu, K. Misumi, Y. Maeda, Y. Yoshida and H. Hayami, One-year, regional-scale simulation of 137Cs radioactivity in the ocean following the Fukushima Daiichi Nuclear Power Plant accident, *Biogeosciences* 10 (2013), pp.5601-5617.

[26] M. Aoyama, D. Tsumune, M. Uematsu, F. Kondo and Y. Hamajima, 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, *Geochem. J.* 46 (2012), pp.321-325.

[27] M. Aoyama, Y. Hamajima, M. Hult, M. Uematsu, E. Oka, D. Tsumune and Y. Kumamoto, ^{134}Cs and ^{137}Cs in the North Pacific Ocean derived from the March 2011 TEPCO Fukushima Dai-ichi Nuclear Power Plant accident, Japan. Part one: surface pathway and vertical distributions, *J. Oceanogr.* 72 (2016), pp.53-65.

[28] M. Aoyama, Y. Hamajima, Y. Inomata and E. Oka, Recirculation of FNPP1-derived radiocaesium observed in winter 2015/2016 in coastal regions of Japan, *Appl. Radiat.Isot.* 126 (2017), pp.83-87.

[29] Y. Kumamoto, M. Aoyama, Y. Hamajima, S. Nishino, A. Murata and T. Kikuchi, Meridional distribution of Fukushima-derived radiocesium in surface seawater along a trans-Pacific line from the Arctic to Antarctic Oceans in summer 2012, *J. Radioanal. and Nucl. Chem.* 307 (2016), pp.1703-1710.

[30] M. Aoyama, M. Uematsu, D. Tsumune and Y. Hamajima, Surface pathway of radioactive plume of TEPCO Fukushima NPP1 released ^{134}Cs and ^{137}Cs. *Biogeosciences* 10 (2013), pp.3067-3078.

[31] Y. Kumamoto, M. Aoyama, Y. Hamajima, T. Aono, S. Kouketsu, A. Murata and T. Kawano, Southward spreading of the Fukushima-derived radiocesium across the Kuroshio Extension in the North Pacific, *Sci. Rep.* 4 (2014), pp.4276, DOI:10.1038/srep04276.

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