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Mass balance and latest fluxes of radiocesium derived from the fukushima accident in the western North Pacific Ocean and coastal regions of Japan

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ABSTRACT

This article summarizes and discusses mass balance calculations of the activities of Fukushima-derived ^{137}Cs released to the atmosphere and ocean prior to 2018 as well as the ^{137}Cs inventories on land and in the ocean, biota, and sediment. We propose that the consensus value of the total amount of ^{137}Cs released to the atmosphere was 15–21 PBq; atmospheric deposition of ^{137}Cs on land was 3–6 PBq; atmospheric deposition of ^{137}Cs on the North Pacific was 12–15 PBq; and direct discharge of ^{137}Cs to the ocean was 3–6 PBq. We also evaluated the movement of ^{137}Cs from one domain to another for several years after the accident. We calculated that the amount of ^{137}Cs transported by rivers might be 40 TBq. The annual deposition of ^{137}Cs due to resuspension at Okuma during the period 2014–2018 was 4–10 TBq year^{-1}. The ^{137}Cs discharged to the ocean was 0.73–1.0 TBq year^{-1} in 2016–2018. The integrated amount of FNPP1-derived ^{137}Cs that entered the Sea of Japan from the Pacific Ocean from 2011 until 2017 was 270 ± 20 TBq, 6.4% of the estimated amount of FNPP1-derived ^{137}Cs in Subtropical Mode Water in the North Pacific. The integrated amount of FNPP1-derived ^{137}Cs that returned to the North Pacific Ocean through the Tsugaru Strait from the Sea of Japan was 110 ± 10 TBq. Decontamination efforts removed 134 TBq of ^{137}Cs from surface soil prior to February 2019, an amount that corresponded to 4% of the ^{137}Cs deposited on land in Japan.

1. Introduction

The total amount of radionuclides released to the environment from the Fukushima Dai-ichi Nuclear Power Plant, hereafter FNPP1, as a result of the accident in March 2011 as well as the impact of those radionuclides on biota, and especially humans, have been among the major concerns related to the FNPP1 accident. The radionuclide of principal concern with respect to human health has been radiocesium, and it is thus particularly important to know how much radiocesium was released to the environment. Many articles and several review articles have already been published concerning this issue (Buesseler et al., 2017; IAEA, 2015; Mathieu et al., 2018; Smith, 2014), but there has been no discussion based on mass balances between the atmosphere, land, and ocean. It is important to consider mass balances in discussions of the total amount of radionuclides released to the environment because the law of conservation of mass is a basic principle, and mass balance is one of the strongest constraints on estimates of the total amount of radionuclides released to the environment and to inventories in the air, on the land, and in the North Pacific Ocean.

In this paper, we have summarized the results of studies of the amounts of Fukushima-derived ^{137}Cs that were released to the atmosphere and ocean as well as estimates of the ^{137}Cs inventories on land, in the ocean, in biota, and in sediments. We propose consensus values of these inventories based on mass balance considerations. Finally, we discuss the fluxes of ^{137}Cs between domains for several years after the FNPP1 accident. We consider in particular fluxes from the land to the ocean via rivers, releases from the accident site to the ocean, and delayed effects of the accident associated with resuspension from the land to the atmosphere, deposition from the atmosphere onto the land and ocean, and transport of FNPP1-derived ^{137}Cs from the North Pacific Ocean to the Sea of Japan. Finally, we consider the total amount of ^{137}Cs in surface soil removed by human activity as a part of decontamination work.

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2. Mass balance of FNPP1 derived radiocesium in the environment

2.1. Summary of estimates of sources, deposition, and inventories of radiocesium released from the FNPP1 to the land and ocean

Mass balances of radiocesium are among the strongest constraints on the environmental impact of radiocesium released from the FNPP1. Equation (1) is a simple mass balance equation for FNPP1-derived radiocesium and expresses the fact that the sum of all the radiocesium released to the environment must equal the inventories on the land, in the ocean, in sediment, and in biota.

$$\Sigma \, Ri = \Sigma \, Ij \quad (1)$$

Where Ri is the amount of radiocesium released to domain i, and Ij is the inventory in domain j.

i: 1 = atmosphere, 2 = direct discharge
j: 1 = atmosphere (except for resuspension, now zero due to short residence time), 2 = land, 3 = ocean, 4 = sediment, 5 = biota (negligible, details are shown in the next chapter)

We have already proposed (Aoyama et al., 2019) consensus values of Fukushima-derived radiocesium in these environments, and the values in that previous proposal are confirmed in this paper with a very slight change for atmospheric release.

2.2. Mass balance and consensus values

Table 1 summarizes the results of this study. To obtain consensus values, we did not include several results from previous reviews (Buesseler et al., 2017) that were obviously obtained by inappropriate methods, did not represent the whole region, or included the amount of fallout as part of the direct discharge. Estimates of the total amount of radiocesium released to the atmosphere ranged from 8.1 PBq (Yumimoto et al., 2016) to 36 (with a range from 23 to 50) PBq (Stohl et al., 2012). The lowest reported values are 8–10 PBq by three researchers (Achim et al., 2014; Hirao et al., 2013; Yumimoto et al., 2016). Those values are too small because the mass balance of observed land deposition in Japan is 3 PBq (Japanese Ministry of Education, Culture, Sports, Science and Technology, 2011), and the estimated total land deposition is 3–6 PBq. The confirmed inventories in the North Pacific are 15–16 PBq by two methods (Inomata et al., 2016; Tsubono et al., 2016) and 3–6 PBq via direct discharge. These values indicate that at least 15 PBq should have been released to the atmosphere based on mass balance considerations. The estimate of 36 PBq released to the atmosphere (Stohl et al., 2012) is too large because 30 PBq (36 PBq to the atmosphere minus a maximum of 6 PBq to the land) is almost double the reliable radiocesium inventory in the North Pacific Ocean. We therefore conclude that 15–21 PBq might be the consensus value for the total atmospheric release, an estimate that is supported by five studies (Aoyama et al., 2016; Katata et al., 2015; Mathieu et al., 2012; Saunier et al., 2013; Winiarek et al., 2014). The consensus value for R1, the atmospheric release of radiocesium, is therefore 15–21 PBq and is only slightly changed from the previously proposed range of 15–20 PBq (Aoyama et al., 2019).

We conclude that 3–6 PBq might be the consensus for R2, direct discharge to the land, based on reported values that range from 3 to 6 PBq (Estournel et al., 2012; Kawamura et al., 2011; Kumamoto et al., 2019; Tsumune et al., 2012).

The total inventory in the atmosphere, I1, might be negligible because of the short residence time (about several weeks) of radiocesium in the atmosphere. The estimated land deposition, I2, was based on observed land deposition in Japan of 3 PBq (Japanese Ministry of Education, Culture, Sports, Science and Technology, MEXT, 2011) and estimated total land deposition of 3–6 PBq (Aoyama et al., 2016). We conclude that total land deposition was 3–6 PBq. Reported values of the total radiocesium inventory in the North Pacific by different methodologies are in good agreement and lie in the range 15–18 PBq (Aoyama et al., 2016; Inomata et al., 2016; Tsubono et al., 2016). We therefore conclude that 15–18 PBq was the consensus for the total inventory of radiocesium in the North Pacific.

For atmospheric deposition to the North Pacific, reported values are in good agreement and fall in the range 12–15 PBq (Aoyama et al., 2016; Tsubono et al., 2016). We therefore conclude that 12–15 PBq was the

Table 1
Source, deposition and inventory estimates for 137Cs from the Fukushima Daiichi nuclear power plants to land and ocean.

	Total atmospheric release (PBq)	Atmospheric deposition on land (PBq)	Atmospheric deposition on the North Pacific (PBq)	Direct discharge to ocean (PBq)	Total in the North Pacific (PBq)	Reference
			*	4		Kawamura et al. (2011)
	20.6					Mathieu et al. (2012)
	36 (23–50)					Stohl et al. (2012)
			*	4.3 ± 0.2		Estournel et al. (2012)
				3.5 ± 0.7		Tsumune et al. (2012)
	15.5					Saunier et al. (2013)
	9.6					Hirao et al. (2013)
				5.6 ± 0.2		Miyazawa et al. (2013)
	11.6–19.3					Winiarek et al. (2014)
	10.8					Achim et al. (2014)
	9.8					Katata et al., 2015 (land data only)
	14.5					Katata et al., 2015 (land and sea data)
	8.1					Yumimoto et al. (2016)
	15.2–20.4	3–6	11.7–14.8	3.5 ± 0.7***	15.2–18.3	Aoyama et al. (2016)
					15.3 ± 2.6	Inomata et al. (2016)
			10.5 ± 0.9		16.1 ± 1.4	Tsubono et al. (2016)
				3–6		Kumamoto et al., 2019
		2.65				MEXT (2011)
		3				Aoyama unpublished (Integrated Japaned land only)
Consensus	**15–21**	**3–6**	**12–15**	**3–6**	**15–18**	

*:They estimated but not covered for the North Pacific Ocean.
**: JAEA group published several estimates and this one is latest of their group.
***: Adopt Tsumune et al., 2012 estimation when estimate all domain.

consensus value for atmospheric deposition on the North Pacific.

There are radiocesium inventories in three domains of the interior of the North Pacific Ocean: the surface layer, Subtropical Mode Water (STMW), and Central Mode Water (CMW). The FNPP1-derived radiocesium inventory has been estimated to be 4.2 ± 1.1 PBq in the STMW (I3-2) by Kaeriyama et al. (2016) and 7.9 ± 1.4 PBq in the surface layer (I3-1) by Inomata et al. (2018b). In the CMW (I3-3), the FNPP1-derived radiocesium inventory has been estimated to be 2.5 ± 0.9 PBq by Inomata et al. (2018b) based on a mass balance between the total inventory in the Pacific Ocean and inventories in the three domains. In the sediment (I4), we assumed an inventory of 0.13 ± 0.06 PBq (Kusakabe et al., 2013). The inventory in the biota (I5) might be less than 200 GBq because a maximum estimate of radiocesium in biota can be obtained from the fish catch of 20 × 10^6 kg around the Fukushima coast and an assumed activity of no more than 1 × 10^4 Bq kg^{-1} in the fish (Aoyama et al., 2019).

Finally, Eq. (1) can be expressed as Eq. (2) based on the details of the mass balance, including the ocean interior, and the inventories as stated above.

$$R1 + R2 = I1 + I2 + (I3\_1 + I3\_2 + I3\_3) + I4 + I5 \quad (2)$$

Where.

R1: Atmospheric release 15–21 PBq
R2: Direct discharge 3–6 PBq
I1: Total in the atmosphere negligible
I2: Total on land 3–6 PBq
I3_1: Surface water 7.9 ± 1.4 PBq
I3_2: Subtropical Mode Water 4.2 ± 1.1 PBq
I3_3: Central Mode Water 2.5 ± 0.9 PBq
I4: Sediment 0.13 ± 0.06 PBq
I5: Biota negligible

3. Estimate and summary of latest transport amounts/fluxes between domains

There are several issues that have remained unresolved during the eight years since the accident. There are still uncertainties in several fluxes as described below.

3.1. Radiocesium transport from land to ocean by rivers

The amount of ^{137}Cs transported from the land to the ocean by 14 rivers around the FNPP1 (estimated to be 10–12 TBq $year^{-1}$ in 2011–2012) has been very small compared to the total deposition on land (3–6 PBq). The river transport includes particulate and dissolved forms of radiocesium (Kitamura et al., 2014). In 2015–2016, the flux by 16 rivers was estimated to be 0.4–0.6 GBq day^{-1} or 0.15–0.2 TBq $year^{-1}$ for dissolved radiocesium only (Tsumune et al., 2020). Fluvial transport of ^{137}Cs to the ocean from the Abukuma River, which is the largest river in the Fukushima region, was estimated to be 12 TBq from June 2011 to August 2015, and almost all of this radiocesium (96.5%) was transported in particulate form (Taniguchi et al., 2019). It has also been reported that after one year, the concentrations of radiocesium activity were lower and exhibited a more gradual secondary decline that was associated with reduced radiocesium losses from paddy fields, farmland, and urban areas, whereas forest areas continued to be more consistent sources of radiocesium (Taniguchi et al., 2019). Use of the ratio of activities of particle and dissolved forms of radiocesium calculated by Taniguchi et al. (2019) gives an estimated flux by 16 rivers of particulate and dissolved forms of 3.6–4.8 TBq $year^{-1}$ in 2015–2016. We estimate the apparent half-life of fluvial transport by rivers from 2011 to 2012 to 2015–2016 to be about 3 years. In sum, because the integrated annual flux of 10–12 TBq $year^{-1}$ in 2011–2012 had an apparent half-life of 3 years. we conclude that the total amount of ^{137}Cs transported from land to the ocean by all the rivers in the heavily contaminated region during June 2011 to 2016 might be 40 TBq (0.04 PBq). This maximum fluvial flux of 0.04 PBq corresponds to less than 1.3% of the ^{137}Cs deposited in the Fukushima region of Japan.

3.2. Land to atmosphere (resuspension) and atmosphere to land (aftereffect deposition)

There is a long history of observations and studies of radioactive fallout in Japan. Monthly ^{137}Cs deposition data at Tsukuba, Japan during the period from April 1957 to March 2018 was already published (Aoyama, 2019). For monthly ^{137}Cs deposition data at Fukushima-shi and Okuma-cho, observed data can be obtained at Fukushima prefectures web site (https://www.pref.fukushima.lg.jp/site/portal/genan225.html). These fallout data were summarized in Table 3 as annual deposition and shown in Fig. 1. The larger fallout of ^{137}Cs was a result of atmospheric nuclear weapons tests conducted mainly in the late 1950s and early 1960s by the USA and former USSR, fallout from Chinese atmospheric nuclear weapons tests from 1964 until 1980, and fallout from the Chernobyl accident in 1986 (Fig. 1) (Aoyama, 1999; Aoyama et al., 1987, 2006; Igarashi et al., 2003; Katsuragi and Aoyama, 1986). The decay-corrected cumulative deposition of ^{137}Cs in Tokyo/Tsukuba was about 6 kBq m^{-2} in 1966 (Aoyama, 1999) and 2.6 kBq in 2011, just before the Fukushima accident. After the Fukushima accident, the annual deposition of ^{137}Cs at Tsukuba, which is located about 270 km southwest of the FNPP1 site (Fig. 2) was 25.5 kBq m^{-2} in 2011. Thereafter, the annual deposition of ^{137}Cs at Tsukuba decreased rapidly to 39 Bq m^{-2} in 2015 (Table 3 and Fig. 2). Since 2015, the annual deposition of ^{137}Cs at Tsukuba has been more-or-less constant and fell in the range 36–39 Bq m^{-2} during 2015–2017. At Okuma, which is located 10 km west of the FNPP1 site (Fig. 2), the annual depositions in 2011, 2012, and 2013 were 3.57 MBq m^{-2}, 59 kBq m^{-2}, and 312 kBq m^{-2}, respectively. The annual deposition of ^{137}Cs at Okuma has varied more-or-less randomly from 4 to 10 kBq m^{-2} during the period 2014–2018. In contrast, at Fukushima-shi, which is located about 80 km northwest of the FNPP1 site (Fig. 2), the annual deposition of ^{137}Cs has followed a decreasing trend similar to the trend observed at Tsukuba and was 300 Bq m^{-2} in 2017. Radiocesium deposited on the ground and vegetation could return to the atmosphere, and the relatively high rates of deposition at Okuma and Fukushima-shi suggest that there has been resuspension of radiocesium deposited on the land.

Resuspension might be one of the important aftereffects of the FNPP1accident. Although radioactive particles might be resuspended by the wind or as a result of activities such as biomass burning, the dominant process responsible for resuspension in contaminated areas of Fukushima is not fully understood. Kinase et al. (2018) have examined the processes responsible for resuspension of radiocesium based on long-term measurements of the atmospheric concentration of radiocesium activity at four sites in the contaminated areas of Fukushima as well as of the aerosol characteristics. Their results differed from previous studies based on data at urban sites and suggested that the resuspension processes and the particles associated with the resuspended radiocesium changed seasonally. Biological activities in forest ecosystems can contribute considerably to radiocesium resuspension during the summer and fall. During the winter and spring, soil, mineral, and vegetation debris account for most of the coarse particles in the atmosphere, and radiocesium resuspension during these seasons can be attributed to mobilization of these particles by the wind (Kinase et al., 2018). Kinase et al. (2018) found that the concentrations of bioaerosols fluctuated around an average of roughly 10^6 particles m^{-3}; the phyla Basidiomycota and Ascomycota (true fungi) accounted for approximately two-thirds of the bioaerosols; and the fungal spore concentration in the air was positively correlated with the radiocesium concentration at Namie in the summer of 2016 (Igarashi et al., 2019).

In terms of mass balance, the annual deposition of ^{137}Cs at Okuma, which ranged from 4 to 10 kBq m^{-2} during 2014–2018, indicates that a

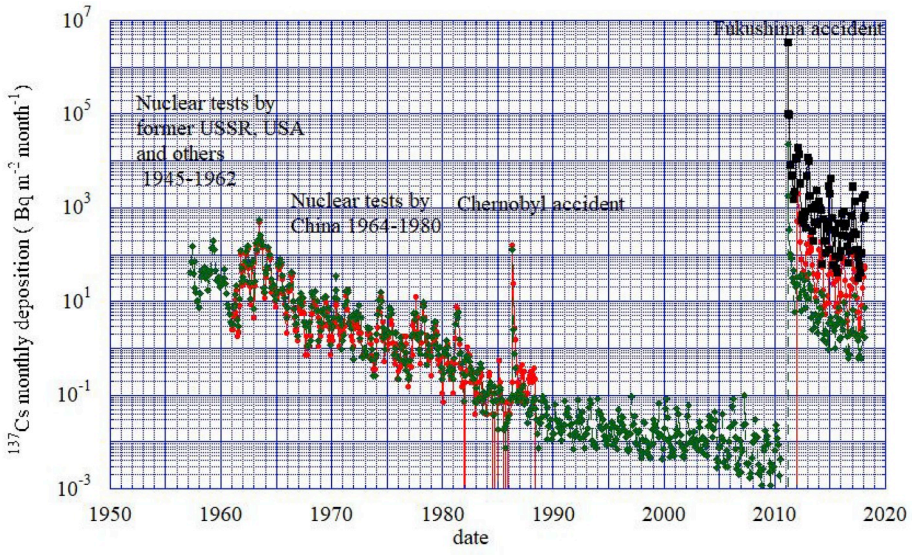

Fig. 1. Observed monthly deposition of ^{137}Cs at Tokyo/Tsukuba, Fukushima-shi and Okuma
Blue diamond: Tokyo/Tsukuba, Red circle: Fukushima-shi, Black square: Okuma. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

flux of 4–10 TBq year^{-1} from the land to the atmosphere would be required if it can be assumed that this much radiocesium is deposited within a 50 km × 20 km area (i.e., 1 × 10^9 m^2) of the coastal region each year. A flux of 4–10 TBq year^{-1} corresponds to about 0.1%–0.3% of the total deposition on land in Japan. This radiocesium would just be recycled via resuspension if it were deposited on land, but it would be a net flux from the land to the ocean if it were deposited on the ocean. We discuss this issue in section 3. x.

3.3. *Continuous radiocesium flux from the accident site to the ocean*

The temporal variations of ^{137}Cs activity concentration in surface water inside the port (i.e., Monoageba-landing area) and it for in the 56N canal of the FNPP1 have been similar. The ^{137}Cs activity concentration in surface water has varied seasonally: since 2015, it has been low in the winter (January–February) and high in the summer and fall (July–October) (Fig. 3). The ^{137}Cs activity in the port dropped by about one order of magnitude when a seaside barrier wall was installed in the middle of 2015. However, the ^{137}Cs activity concentration in surface water at the 56N canal followed a slightly different trend before the middle of 2015 (Fig. 3). The source of contaminated water to the port and to the 56N canal might therefore be similar, but the routes of leakage to the two places may have differed. Between 2013 and 2016, the annual average of the ^{137}Cs activity at the 56N canal decreased with an apparent half-life of one year (Aoyama, 2018). However, from 2016 to 2018 the annual average of the ^{137}Cs activity concentration in surface water at the

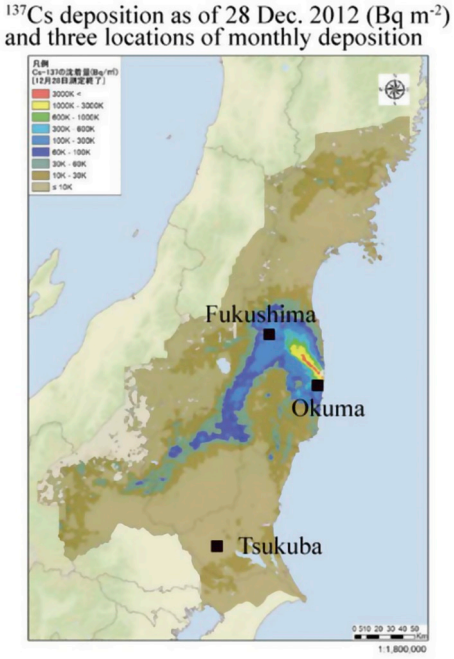

Fig. 2. ^{137}Cs deposition as of Dec. 28, 2012 and three locations of monthly deposition Deposition map is in a report of JAEA at https://radioactivity.nsr.go.jp/ja/contents/14000/13591/24/.pdf.

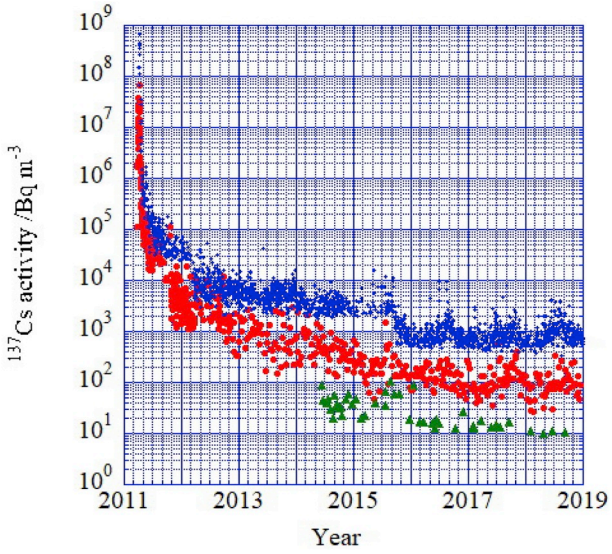

Fig. 3. Temporal variations of ^{137}Cs activity concentrations in surface water at Monoageba in the port of FNPP1, 56North canal of FNPP1 and Tomioka 30 km south of FNPP1.
Blue Diamond: Monoageba in the port of FNPP1, Red Square: 56North canal, Green Triangle: Tomioka. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 2
Annual average of ^{137}Cs activity concentration in surface water at 56N canal of FNPP1.

Year	^{137}Cs Bq m^{-3}		
2013	1019	±	708
2014	580	±	442
2015	233	±	216
2016	138	±	60
2017	140	±	104
2018	106	±	64

Weekly values can be obtained at TEPCO web site below and the authors made annual average of them.
http://www.tepco.co.jp/decommission/data/analysis/pdf_csv/2020/1q/seawater-newest01-j.csv.

56N canal ranged from 100 to 140 Bq m^{-3} and did not show a decreasing trend with time (Table 2 and Fig. 3). Based on the relationship between the ^{137}Cs activity concentration in surface water at the 56N canal and the flux to open water, the ^{137}Cs activity concentration in surface water at the 56N canal in 2016–2018 corresponded to 2.0–2.8 GBq day^{-1}, or 0.73–1.0 TBq year^{-1} of ^{137}Cs discharged to open water from the FNPP1 site (Tsumune et al., 2012). At Tomioka, which is located 30 km south of the FNPP1 site, the ^{137}Cs activity concentration in surface water showed a trend (Fig. 3) similar to that of the ^{137}Cs activity concentration in surface water at the 56N canal. This similarity confirmed that there was a change of the rate of direct discharge from the FNPP1 site to ocean in 2016 .

The discharge of about 1 TBq year^{-1} from the FNPP1 site to the ocean during 2016–2018 was very small compared with the discharge of 3.5 ± 0.7 PBq until May 2011 (Tsumune et al., 2012). However, the discharge from the FNPP1 site to the ocean in 2016–2018 was still larger than the flux of 0.15–0.20 TBq year^{-1} by the river stated in chapter 3.1.

In contrast, ^{137}Cs deposition of 4–10 TBq year^{-1} was the net flux to the ocean if we assume that deposition of 4–10 kBq m^{-2} occurred within a coastal region of 50 km × 20 km (i.e., 1 × 10^9 m^2). Direct release was estimated to be 1.0 TBq year^{-1} in 2016–2018. The potential atmospheric deposition of 4–10 TBq year^{-1} was therefore larger than the amount of direct discharge. The resultant ^{137}Cs activity in the water due to this deposition in 2016–2018, however, might have been only 0.75 Bq m^{-3} based on an assumed deposition rate is 15 Bq m^{-2} per day, a mixing depth of 20 m, and a residence time of the coastal water of roughly a few days. Therefore, direct discharge would still account for most of the ^{137}Cs activity (~100 Bq m^{-3}) close to the FNPP1 site.

3.4. Radiocesium transport from STMW to the sea of Japan

Several years after the FNPP1 accident, ^{137}Cs activities gradually increased in the East China Sea (ECS) and Sea of Japan (SOJ) (Aoyama et al., 2017; Inomata et al., 2018). Inomata et al. (2018a) have already reported the amounts of FNPP1-derived ^{137}Cs transported from STMW to the SOJ during 2012–2016, and we estimated that flux during 2012–2017 using the same methodology as Inomata et al. (2018a). We estimated the integrated amount of FNPP1-derived ^{137}Cs that entered the SOJ until 2017 to be 270 ± 20 TBq, which is 6.4% of the estimated total amount of FNPP1-derived ^{137}Cs in the STMW. We estimated the integrated amount of FNPP1-derived ^{137}Cs that returned to the North Pacific Ocean through the Tsugaru Strait to be 110 ± 10 TBq, 42% of the total amount of FNPP1-derived ^{137}Cs transported to the SOJ and 2.7% of the estimated total amount of FNPP1-derived ^{137}Cs in the STMW (Inomata et al., submitted to Proceedings of ENVIRA, 2019).

3.5. Results of decontamination work

We evaluated the total amount of ^{137}Cs that was removed from surface soil as a result of the decontamination efforts of the Japanese government. We used a report at http://josen.env.go.jp/chukanchozou/action/safety_commission/pdf/safety_commission_02_190327.pdf (accessed on July 12, 2019, Japanese document) as the basis of our calculations.

Table 3
Annual 137Cs deposition at Tsukuba, Fukushima-shi and Okuma-cho.

Year	Tsukuba	Fukushima-shi	Okuma-cho
	Bq m-2	Bq m-2	Bq m-2
2011	25,500	N.A.	3,570,000
2012	260	4370	59,700
2013	130	1020	312,000
2014	60	645	8710
2015	39	446	8490
2016	37	372	4660
2017	36	304	9830
2018	N.A.	N.A.	4020

N.A.:not available

Table 4
Results of decontamination.

	Bq kg-1	kg	Bq
	500	6.24 × 10^8	3.12 × 10^11
	2000	1.83 × 10^9	3.66 × 10^12
	4000	6.86 × 10^8	2.74 × 10^12
	6500	4.48 × 10^8	2.92 × 10^12
	14,000	5.36 × 10^8	7.51 × 10^12
	40,000	2.77 × 10^8	1.11 × 10^13
Total			2.82 × 10^13
137Cs			2.26 × 10^13
134Cs*			5.65 × 10^12

*:decay corrected on March 2015 Already transported amount of soil was 2.2 × 10^6 m3 and density is assumed 2.0 × 10^6 kg m-3.

Because no density data were included in the report, we assumed that the density of the soil was 2.0 × 10^3 kg m^{-3}. The volume of soil transported was reported to be 2.2 × 10^6 m^3, and the total mass of soil was therefore 4.4 × 10^{12} g. In the report, the total activity of ^{134}Cs and ^{137}Cs in a bag was assigned to 7 activity categories, and the activities were corrected for radioactive decay to March 2015. Because the ratio of ^{134}Cs–^{137}Cs activities was about 1 at the time of the accident (Nishihara et al., 2012), the ratio of ^{134}Cs–^{137}Cs activity was 0.25 in March 2015. Based on these assumptions, we estimated the total amount of radiocesium and the total activity of ^{134}Cs and ^{137}Cs in the soil that was removed and transported (Table 4): the total amount of soil removed from the surface was 1.4 × 10^7 m^3, and the reported amount of soil transported was 2.36 × 10^6 m^3. We assumed that the characteristics of the removed soil that remained and the removed soil that was transported were the same. We therefore conclude that 134 TBq of ^{137}Cs (i.e., 22.6 TBq × (14.0 × 10^6/2.36 × 10^6) was removed from the surface soil. The total amount of ^{137}Cs removed from the surface soil corresponded to 4.5% of the ^{137}Cs deposited on land in Japan until February 2019.

4. Conclusions

We propose that the consensus amount of total atmospheric release of ^{137}Cs was 15–21 PBq. The fallout of ^{137}Cs from the atmosphere was 3–6 PBq onto the land and 12–15 PBq onto the North Pacific. The direct discharge of ^{137}Cs to the ocean was 3–6 PBq. The total inventory of ^{137}Cs in the North Pacific was 15–18 PBq. We also estimated the inventory of ^{137}Cs in the surface layer and CMW to be 7.9 ± 1.4 and 2.5 ± 0.9 PBq, respectively.

The amount of ^{137}Cs transported by rivers from land to the ocean for several years after the FNPP1 accident might be 0.04 PBq, which corresponds to 1.3% of the ^{137}Cs deposited in the Fukushima region of Japan. The annual deposition of ^{137}Cs at Okuma during the period 2014–2018 means that 4–10 TBq year^{-1} was resuspended from the land

to the atmosphere, an amount that corresponds to about 0.1–0.3% of the total amount of ^{137}Cs deposited on land in Japan.

The ^{137}Cs activity at the 56N canal in 2016–2018 corresponded to 0.73–1.0 TBq year^{-1} of ^{137}Cs discharged to open water from the FNPP1 site. The integrated amount of FNPP1-derived ^{137}Cs that entered the SOJ from the North Pacific Ocean until 2017 was estimated to be 0.270 ± 0.002 PBq, whereas 0.11 ± 0.01 PBq returned to the North Pacific Ocean through the Tsugaru Strait. Decontamination efforts were estimated to have removed 0.134 PBq of ^{137}Cs from surface soil, an amount that corresponds to 4.5% of ^{137}Cs deposited on land in Japan prior to February 2019.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2020.106206.

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