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Impact of fluvial discharge on ^{137}Cs in the ocean following the Fukushima Daiichi Nuclear Power Station accident

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Abstract: After the Fukushima Daiichi Nuclear Power Station (F1NPS) accident, ^{137}Cs activity concentrations have not yet decreased to pre-accident levels because of direct release from the site and fluvial discharges of ^{137}Cs deposited on land. It is necessary to consider dispersion processes in the coastal area to understand the impact of multiple river discharges and direct release. To achieve this goal, we carried out oceanic dispersion simulations that considered direct release and fluvial discharges and compared the results with the annual averages of observed data. We assumed that particulate ^{137}Cs discharged from rivers to the ocean quickly resuspended and re-leached after coagulation and precipitation, and that all of the ^{137}Cs was dispersed. The reproducibility of results was improved by considering fluvial discharges of particulate ^{137}Cs at all sites between 2013 and 2016, except near the F1NPS. In other words, particulate ^{137}Cs discharged from rivers was found to influence the results of ocean surface activity concentrations within a relatively short period of time. The impact of direct release was dominant for the observed ^{137}Cs activity concentrations adjacent to the F1NPS, which was used to estimate direct releases.

Keywords: ^{137}Cs; fluvial discharge; Fukushima Daiichi Nuclear Power Station accident; marine environment; regional ocean model.

Introduction

A series of accidents at the Fukushima Daiichi Nuclear Power Station (F1NPS) following the Great East Japan Earthquake and Tsunami of 11 March 2011 resulted in the release of ^{137}Cs into the ocean. The primary release pathways were direct release from the F1NSP site and atmospheric deposition [1].

In previous studies, estimates of direct release of ^{137}Cs have been narrowed to 3–6 PBq [1, 2]. However, those estimates did not distinguish between atmospheric deposition and direct releases. The estimates based on analysis of ^{131}I/^{137}Cs activity ratios, which considered only the effects of direct release, were 3.5 ± 0.7 PBq by the end of May 2011 [3] and 3.6 ± 0.7 PBq by the end of 2019 [4]. The direct release rate estimated using this numerically simulated seawater exchange rate and observations near the F1NPS is consistent with both the direct release rate determined from the volume of seawater exchanged and observations in the harbor [5] as well as the direct release rate determined from the volume of leaked water estimated visually and the concentration of ^{137}Cs in the

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leaked water [6]. The direct release rates were estimated until 2019; since 2016, the rate has stopped decreasing and has remained approximately constant [4].

Uncertainties remain regarding atmospheric deposition because of insufficient understanding of rainfall phenomena at sea and other factors. The simulation targets for reproduction by the atmospheric model have only been releases on land that could be verified with observed values [7]. There has been significant uncertainty in estimates based on the process of descent from the atmosphere to the ocean. However, the effect of atmospheric fallout on coastal areas has been limited to the initial month [8], and the effect of recirculation of ^{137}Cs deposited on the North Pacific has not been seen in coastal areas [9]. Atmospheric deposition was therefore not addressed in this study.

Another release pathway is fluvial discharge of ^{137}Cs deposited on land into the ocean. Tsumune et al. [9] have evaluated the impact of dissolved ^{137}Cs supplied from rivers on the ocean. Regression equations set up from data on particulate ^{137}Cs concentrations in several rivers [10, 11] have been used to estimate the flux of dissolved ^{137}Cs from the distribution coefficient (K_d). The impact on the ocean was evaluated after estimating the flux of dissolved ^{137}Cs. To estimate annual average concentrations, we focused on the period 2013–2016, when the temporal change of direct release rate was stable. Particulate ^{137}Cs was assumed to coagulate and precipitate immediately after being supplied from rivers to the ocean, and the ^{137}Cs was assumed to have no impact on the ocean [12]. The fluvial discharge of dissolved ^{137}Cs was found to have little effect on ^{137}Cs concentrations in the ocean. However, the underestimation of ^{137}Cs activities at coastal stations far from the F1NPS suggested that there was an influence of re-leaching and resuspension of particulate ^{137}Cs. The effects of particulate ^{137}Cs on the ocean therefore remained to be clarified.

Studies have also been carried out on the transport of radioactive material from atmospheric nuclear tests from rivers to the sea. Elevated concentrations of ^{90}Sr were observed in the Columbia River discharge in the USA. On the other hand, although elevated concentrations of ^{137}Cs were observed in estuarine areas, there was no evidence of riverine inputs [13]. Subsequent ocean general circulation modeling studies suggested that this was due to advection of ^{137}Cs that had fallen into the western North Pacific [14]. Thus, the supply of ^{137}Cs from the Columbia River to the ocean was negligible. In the Kara Sea, elevated concentrations of ^{137}Cs have also been observed in estuarine sediments due to the contribution of the Ob and Yenisey Rivers. This is thought to be a result of coagulation and precipitation of ^{137}Cs from freshwater into seawater [15]. Increased concentrations in seawater and sediments due to the influx of ^{137}Cs from the land fallout of the Chernobyl NPP accident into the Black Sea via the Danube and Dnieper rivers are also a long-term effect [16]. Furthermore, ^{137}Cs sediments in the Rhône estuary in the Mediterranean have been found to be affected by atmospheric nuclear testing, fallout from the Chernobyl accident and effluents from nuclear facilities along the river [17]. Modeling of sedimentation processes due to changes in salinity has also been carried out in the Rhone estuary [18].

For the F1NPS accident, Uchiyama et al. [19] have developed a high-resolution, quadruple-nested 3D ocean circulation and sediment transport model to quantify the process of particulate ^{137}Cs transport. The model was validated using *in situ* sediment core data for the Niiida River estuary for the storm and subsequent flooding associated with Typhoon Wipha, which passed by the coast of Fukushima, Japan, in October 2013. The model results showed that nearly 32 GBq of river-derived particulate ^{137}Cs from the flooding associated with Typhoon Wipha was supplied in two days, and about 47 % was still deposited near the estuary 1.5 months later. In other words, about half the particulate ^{137}Cs was resuspended and re-leached. The impact of multiple rivers in the Fukushima coastal area will be addressed in future studies.

Delaval et al. [20] have summarized studies of the desorption that occurs at the interface of the river and ocean due to changes of physicochemical conditions such as ionic strength and solution composition. For particles containing ^{137}Cs released from environmental contamination, the desorption ranged from 0 to 64 % of the particulate activity, with a median at only 3 %. Desorption starts at low salinities of 3–4 and rapidly reaches a threshold at higher salinities of roughly 10–15. The two primary reactions that dominate the kinetics of this process have half-life reaction times of 1 hour and several days. These two reactions are probably related to the adsorption of ^{137}Cs onto sites on different kinds of particles. The dynamics of ^{137}Cs desorption also depend on the initial distribution of ^{137}Cs on these different sites and the relationship of that distribution to contamination history and aging. In other words, quantifying the desorption that occurs at the interface between rivers and the

ocean is a major challenge for chemistry, and quantitative assessment in the case of the Fukushima accident has not been achieved.

Precipitation increases the flux of ^{137}Cs from rivers. The antecedent precipitation index (API) to determine the relationship between rainfall and runoff was used [21]. The API is a weighted sum of daily precipitation for previous days and is used as an indicator of soil moisture. It can be used to determine how many days have passed since there was much rain. Results from the Fukushima coast (Ukedo, Tomioka, Iwasawa, and Onahama) have indicated a high correlation between the 7-day API and the level of ^{137}Cs radioactivity in surface seawater. This API corresponds to the slow reaction time determined by Delavel et al. [20]. The implication is that particulate ^{137}Cs discharged to the ocean might affect the ocean by resuspension and re-leaching with a time delay of about 7 days.

Kakehi et al. [12] have made observations in the Abukuma River from the estuary to the ocean and have estimated that more than 80 % of the particulate ^{137}Cs supplied from the river is deposited off the mouth of the river. However, their observations amount to a snapshot, and the temporal changes remain to be investigated. Takata et al. [22], who studied seven rivers south of the Fukushima Daiichi NPP for the period before and after Typhoon Hagibis in October 2019, noticed possible dissolution during river-to-sea supply, but they did not investigate the involvement of sediments near the river mouth.

The proportion of dissolved and particulate forms of ^{137}Cs supplied from rivers has been reported to vary with land use based on human activities in the catchment [11]. Overall, however, particulate ^{137}Cs makes a significant contribution to runoff flux, especially during typhoons. This discharge is expected to persist because the cumulative amount of ^{137}Cs discharged by rivers to date is lower than the quantity of atmospheric deposition of ^{137}Cs on land.

Sakuma et al. [23] have developed a simple model to assess and predict ^{137}Cs runoff from catchments. They used a tank model and load-discharge (L-Q) equations to estimate fluxes of dissolved and particulate ^{137}Cs in the runoff from seven major rivers. They confirmed consistency with some observations, although they did not consider land use and other factors in detail. The majority of the ^{137}Cs discharged from the watershed was particulate ^{137}Cs rather than dissolved ^{137}Cs.

In this study, we used the fluxes associated with fluvial discharges determined by Sakuma et al. [23] and focused on annual mean values to investigate whether the reproducibility of ^{137}Cs activities in the coastal ocean could be improved if resuspension and re-leaching were taken into account. In other words, we carried out simulations to determine whether resuspension and subsequent deposition of particulate ^{137}Cs off river mouths had an impact. In the ocean, the chemical process of re-leaching is considered more significant than resuspension because the proportion of dissolved ^{137}Cs is higher than that of particulate ^{137}Cs. We expected that this simulation would enhance understanding of the significance of chemical re-leaching of particulate ^{137}Cs, which is supplied from rivers to the ocean following coagulation and precipitation.

Section 2 describes the regional ocean model, the method for estimating direct release rates and fluvial discharge rates, and the observational data. Section 3 describes the results of the direct release and fluvial discharge estimates, the reproducibility of the estimates for the period 2013–2016, and the impact of fluvial discharge. Section 4 provides a summary and prospective for the future.

Methods

Regional ocean model

We used the Regional Ocean Modeling System (ROMS) [24] to simulate ^{137}Cs concentrations in the ocean. We considered direct release from the F1NPS site and fluvial discharge. The model setup was the same as that used previously [9]. The ROMS is a three-dimensional Boussinesq free-surface ocean circulation model formulated using a topography-tracking coordinate system. In this study, the area off Fukushima (35°54′–40°00′N, 139°54′–147°00′E) was used as the model domain. The horizontal resolution was set to 1/120° in both the zonal and meridional directions. The vertical resolution of the s-coordinates was 30 layers. To reduce the computational

resources required for the simulation, the maximum depth was assumed to be 1000 m. The model was forced at the sea surface by wind stresses and heat and freshwater fluxes, the values of which were obtained by a real-time nested atmospheric simulation system. The horizontal resolution of the atmospheric model was 5 km in both the zonal and meridional directions, and the time step of the real-time simulation was 1 hour. During the simulation, horizontal currents, water temperature, salinity, and sea level along the open boundary were obtained from the Japanese Coastal Ocean Prediction Experiment 2 (JCOPE2) reanalysis data [25, 26] with a horizontal resolution of 1/10°. To represent mesoscale eddies during the simulation period, a higher resolution (1/120°) of ROMS water temperatures and salinities were nudged into the JCOPE2 reanalysis results, which did not include coastal areas where depths exceeded 250 m, to simulate the effects of freshwater fluxes from rivers. The nudging parameter was set to 1 day^{-1} to provide a strong constraint on the representation of mesoscale eddies and the Kuroshio Current. Initial conditions of water temperature, salinity, horizontal current velocity, and sea level were set using the JCOPE2 reanalysis results. ^{137}Cs was modeled as a passive tracer whose migration into the ocean interior was controlled by advection and diffusion. The radioactivity of ^{137}Cs in seawater was assumed to decrease by radioactive decay with a half-life of 30.1 years.

Estimation of direct release

The rates of direct release of ^{137}Cs were estimated using the simulation results and the ^{137}Cs activity concentrations measured adjacent to the F1NPS site using the following equation [4].

$$\left[\text{Direct release rate}\left(\text{Bq day}^{-1}\right)\right] = \left[\text{Water exchange rate}\left(\text{m}^3\,\text{day}^{-1}\right)\right] \times \left[\text{Measured } ^{137}\text{Cs activity concentration } \left(\text{Bq m}^{-3}\right)\right]$$

where the water exchange rate was equated to the reciprocal of the simulated ^{137}Cs activity concentration with a release of 1.0 Bq day^{-1}.

The ^{137}Cs activity concentration in the area adjacent to the F1NPS site, which is measured weekly by Tokyo Electric Power Company Holdings, Inc. (TEPCO), was used for estimation purposes [27]. The area for determining the water exchange rate was approximately 1 km × 1 km, including the harbor at the F1NPS site. This area encompassed the assumed release sources and observation points and was adequate for estimating the direct release rate, although the detailed release pathways were unknown. The direct release rate was assumed to be proportional to an exponential approximation of the measured ^{137}Cs activity concentration adjacent to the F1NPS.

Estimation of fluvial discharge

The results of a simplified model that could assess and predict ^{137}Cs (particulate and dissolved forms) runoff from catchments using the tank model and the L-Q equation [23] were used to simulate fluvial fluxes in the regional ocean model. The target rivers were seven rivers with high deposition in the catchment area (Abukuma, Odaka, Ukedo, Takase, Maeda, Kuma, and Tomioka) (Fig. 1; refer to Sakuma et al. [23] for information on each basin). However, because the Ukedo and Takase rivers merge at their mouths, their runoff fluxes were summed. The parameters were changed for the Abukuma River to be more in line with observations [23]. As a result, the particulate ^{137}Cs flux from the Abukuma River increased by 1.4-fold between 2013 and 2016. The freshwater flux concurrent with the ^{137}Cs flux was specified for each river. Relatively high freshwater fluxes enhance southward flow, but differences in the distributions of flow fields caused by variable freshwater fluxes [9] did not affect the annual mean distribution of concentrations.

After being discharged from rivers to the ocean, the ^{137}Cs in particles may be leached, dispersed, resuspended, and re-leached after coagulation, precipitation, sedimentation. The proportional contributions of these processes are unknown; Tsumune et al. [9] have simulated the effect of dissolved ^{137}Cs discharged from rivers on the ^{137}Cs concentrations in the ocean, but they significantly underestimated the ^{137}Cs concentrations. In this study, the

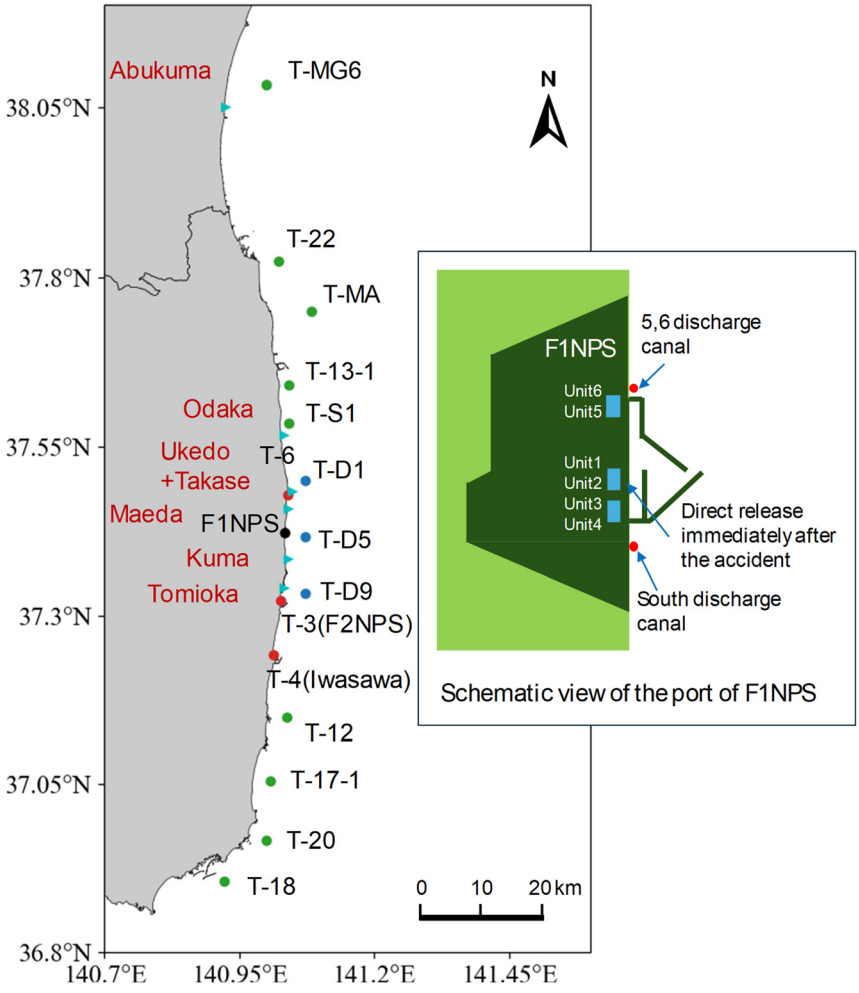

Fig. 1: Map of rivers and observation points and schematic view of port of the F1NPS. Black circle shows observed point adjacent to the F1NPS site (average of the observed values of 5, 6 and south discharge canals), red circles show observed points of the T-6, F2NPS and Iwasawa, blue circles show observed points of T-D1, T-D5 and T-D9, green circles show other coastal observed points. Light blue triangles show the river mouths.

processes of coagulation-sedimentation, resuspension, and re-leaching were not considered, and all particulate ^{137}Cs discharged from rivers was assumed to be dispersed in the ocean. The timeframe for resuspension and re-leaching after coagulation and sedimentation has been estimated to be ~7 days [21], but because we considered annual averages, we did not take time delay into account in this study. Because day-to-day variations were large, we based comparisons between observations and calculations on annual averages [9].

Observed data and period

The observation points along the coast considered to be affected by direct release and fluvial discharge were selected for comparison with simulated results (Fig. 1). All observations were at the surface. The observations adjacent to the F1NPS were at the 5, 6 and the south discharge canals, but because the model could not reproduce the difference between the 5, 6 and the south discharge canals, the average of the 5 and 6 and the south discharge canals, was equated to the point of the F1NPS.

The study covered the period of time beginning in 2013, when the fluctuations of the direct releases of ^{137}Cs were relatively small, to 2016, when the rates of direct release of ^{137}Cs stopped decreasing. Because the fluvial discharge of ^{137}Cs continued to decrease after 2016, we considered 2016 to be the year when the impact of fluvial discharges was highest compared to direct releases.

Results and discussion

Input sources

Direct release

The rate of direct release of ^{137}Cs was based on observations near the F1NPS and was compared to the releases reported by Tsumune et al. [9], with slight changes [4]. The order of magnitude of the direct release of ^{137}Cs decreased from 10^{10} Bq day^{-1} with an apparent half-life of 1 year to 2.3×10^{9} Bq day^{-1} in June 2016 (Fig. 2). Since then, there have been fluctuations around the annual average but no decreasing trend [4].

Immediately after the accident, a leak was visually confirmed to have occurred directly from the Unit 2 intake in the port (Fig. 1). However, the route of the subsequent release was unclear. It has been pointed out that the harbor was damaged by the tsunami and that after 2013, fluxes into the port [5, 28] and out of the port differed significantly and that sources may have existed outside the port [4]. The direct release rates of ^{137}Cs that we determined were not necessarily released within the harbor. They are estimates of releases from the vicinity of the F1NPS. These releases of ^{137}Cs may have included the impact of discharges from groundwater [29]. If runoff from rivers in the vicinity of the F1NPS was affecting observations near the F1NPS (the 5 and 6 and the south discharge canals), the fluxes of ^{137}Cs from this runoff should be subtracted from the estimates of direct release.

Fluvial discharge

The fluvial discharges of particulate ^{137}Cs are shown in Fig. 2. Fluvial discharges varied by more than one order of magnitude. There was a trend towards pulsed increases due to large precipitation events [23]. Typhoon Wipha, Typhoon Vongfong, and Tropical Storm Etau struck the basins in October 2013, September 2014, and September 2015, respectively. During each event, the fluvial discharges of particulate ^{137}Cs increased (Fig. 2).

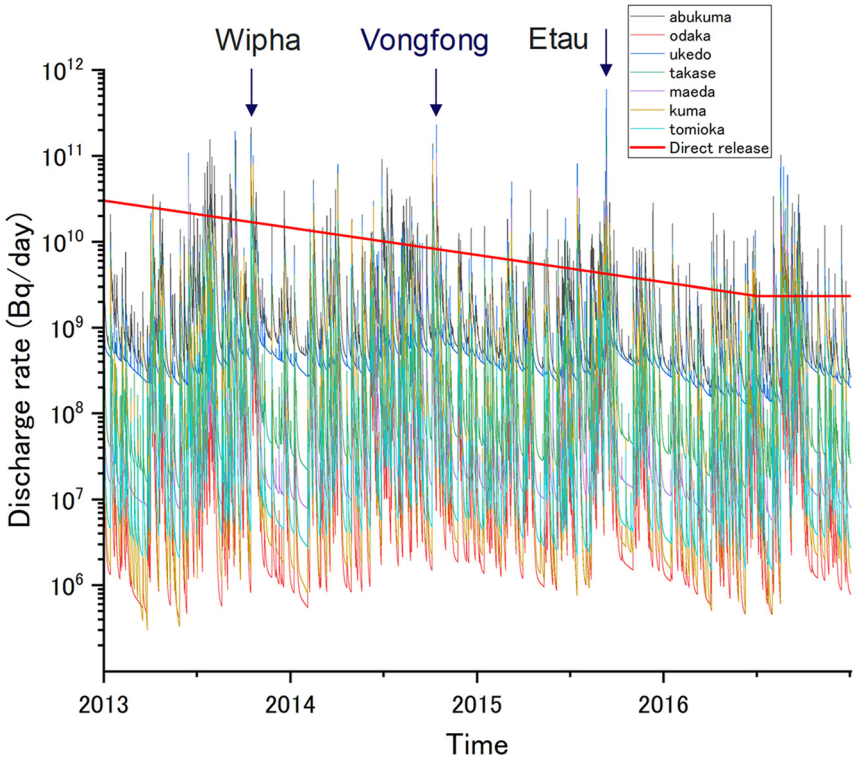

Fig. 2: Direct release rates of ^{137}Cs and rates of fluvial discharges of particulate ^{137}Cs from seven rivers between 2013 and 2016.

Table 1 shows the average discharge rates of dissolved and particulate ^{137}Cs from the river and their apparent half-lives between 2013 and 2016. The maximum average leakage rate of particulate ^{137}Cs was 3.0×10^9 Bq day^{-1} from the Abukuma River. This rate was less than 30 % of the rate of direct release of dissolved and particulate ^{137}Cs from the F1NPS site. The discharge rate of particulate ^{137}Cs from the Ukedo River was 40 % of that from the Abukuma River, although the rate of water discharge from the Ukedo River was 4 % of that from the Abukuma River. The discharge rate of dissolved ^{137}Cs was 20 % of the discharge rate of particulate ^{137}Cs in the Odaka River and less than 10 % in all other rivers. The discharge rates of dissolved and particulate ^{137}Cs from rivers are the products of the dissolved and particulate ^{137}Cs activity concentrations and the river water flow rate. The dissolved and particulate ^{137}Cs activity concentrations have been decreasing with an apparent half-life, whereas the river water flow rates have been highly variable with time but with no apparent trend. It was possible to ascertain the apparent half-lives of the discharge rates of dissolved and particulate ^{137}Cs from the rivers, although the temporal variations of the discharge rates of dissolved and particulate ^{137}Cs from rivers were large. The ranges of apparent half-lives were 3.2–9.5 years for the particulate form and 3.2–4.7 years except 23.7 years at Odaka River for the dissolved form. In the Odaka River, the apparent half-life is longer than in other rivers due to the large initial decrease in particulate ^{137}Cs fluxes and the relatively low ^{137}Cs concentrations since 2013. The apparent half-life of particulate ^{137}Cs concentrations in the Odaka River, determined from observations since June 2016, is also longer than in other rivers, at 11.4 years [30]. The apparent half-life associated with fluvial discharge was longer than the 1-year half-life associated with direct release. However, the direct release rate has not shown a clear downward trend since June 2016.

In all rivers, the discharge flux was larger for particulate than for dissolved ^{137}Cs. Particulate ^{137}Cs discharged by rivers coagulates and precipitates as a result of salinity effects [12], but some is resuspended and re-leached [13, 16]. Analysis of API data indicated that there was a time delay of about 7 days after a precipitation event before impacts became apparent in the ocean [15]. While the dissolved ^{137}Cs activity concentration in the ocean, originating from fluvial fluxes of dissolved ^{137}Cs, has been estimated to be negligibly small in comparison to the observed ^{137}Cs activity concentration in the ocean [9], what fraction of the discharge flux of particulate ^{137}Cs is leached and what fraction is resuspended and re-leached after coagulation and precipitation are unknown. In this study, our numerical simulations assumed that all of the particulate ^{137}Cs that entered the ocean was observed in the ocean surface waters, even if there was a temporal delay. In other words, because the temporal delay was unknown, we assumed that the particulate ^{137}Cs that entered the ocean was dispersed without modification, and we made comparisons based on annual average of observed concentrations. The observed data in this case were considered to have a large, dissolved component, but the unfiltered water may also have contained particulate ^{137}Cs.

Table 1: Average fluvial discharge rates of particulate and dissolved ^{137}Cs, apparent half-lives of particulate ^{137}Cs, and average water discharge rates.

Rivers	Particulate		Dissolved		
	Averaged ^{137}Cs discharge rate (Bq day^{-1})	Apparent half-life (year)	Averaged ^{137}Cs discharge rate (Bq day^{-1})	Apparent half-life (year)	Averaged water discharge rate (m^3 s^{-1})
Abukuma	3.0E + 09	3.2	1.1E + 08	3.2	138.2
Odaka	4.7E + 07	23.7	9.2E + 06	4.7	2.3
Ukedo	1.3E + 09	4.7	9.3E + 07	3.8	5.5
Takase	8.4E + 08	6.3	4.7E + 07	4.7	8.8
Maeda	3.3E + 08	9.5	2.5E + 07	3.8	1.6
Kuma	4.6E + 08	9.5	2.4E + 07	3.8	2.6
Tomioka	1.2E + 08	4.7	1.0E + 07	3.8	2.2
	Particulate and dissolved				
Direct release	9.8E + 09	1.0			

Annual averaged concentrations

2013

The distribution of surface activity concentrations of directly released ^{137}Cs and particulate ^{137}Cs discharged by rivers and their sum for the annual average surface ^{137}Cs activity concentration in 2013 are shown in Fig. 3. The influence of dissolved ^{137}Cs is not shown here, because dissolved ^{137}Cs has been confirmed to be very small [9]. The annual mean distribution of ^{137}Cs activity concentrations due to direct release showed that the ^{137}Cs was advected mainly in a southerly direction. The effects of fluvial discharge were significant in the case of the Abukuma River and the rivers adjacent to the F1NPS. The ^{137}Cs discharged by rivers was found to be advected in a southerly direction in the same way as the ^{137}Cs directly released. We also considered freshwater discharged by rivers in cases where only direct release was considered. In other words, in both of these comparisons, the effects of freshwater fluxes were taken into account in both flow fields. We found that the distributions of activity concentrations when we considered effects of both direct release and fluvial discharge (dissolved + particulate) fit the coastal observations well. Note that background concentrations are about 1 Bq m^{-3} in the seas around Japan because of the re-circulation of ^{137}Cs that fell into the North Pacific Ocean because of the F1NPS accident [30] and the global fallout from atmospheric nuclear tests conducted mainly in the 1960s [31]. Figure 3 does not take this effect into account and should therefore be treated with caution. However, all observed activity concentrations exceeded 10 Bq m^{-3}, and the influence of global fallout could be ignored for those activity concentrations.

A comparison of all the observed and simulated surface ^{137}Cs activity concentrations in 2013 is shown in Fig. 4. In order to be able to ascertain the observation points indicated by each point in Fig. 4, Table 2 shows the results of the observations and calculations for each observation point. We found that the concentrations were underestimated (except in the vicinity of the F1NPS) in the direct release case, but reproducibility improved in the case of the particulate ^{137}Cs discharged by rivers. The residual sum of squares (RSS) decreased from 610.8 Bq m^{-3} in the direct release case to 473.8 Bq m^{-3} in the case of particulate ^{137}C fluvial discharge. The improved reproducibility of fluvial effects especially from Abukuma River was particularly significant at the northern sites (T-MG6, T-22, T-MA) of the F1NPS. In 2013, the impact of direct release was larger, and the agreement was

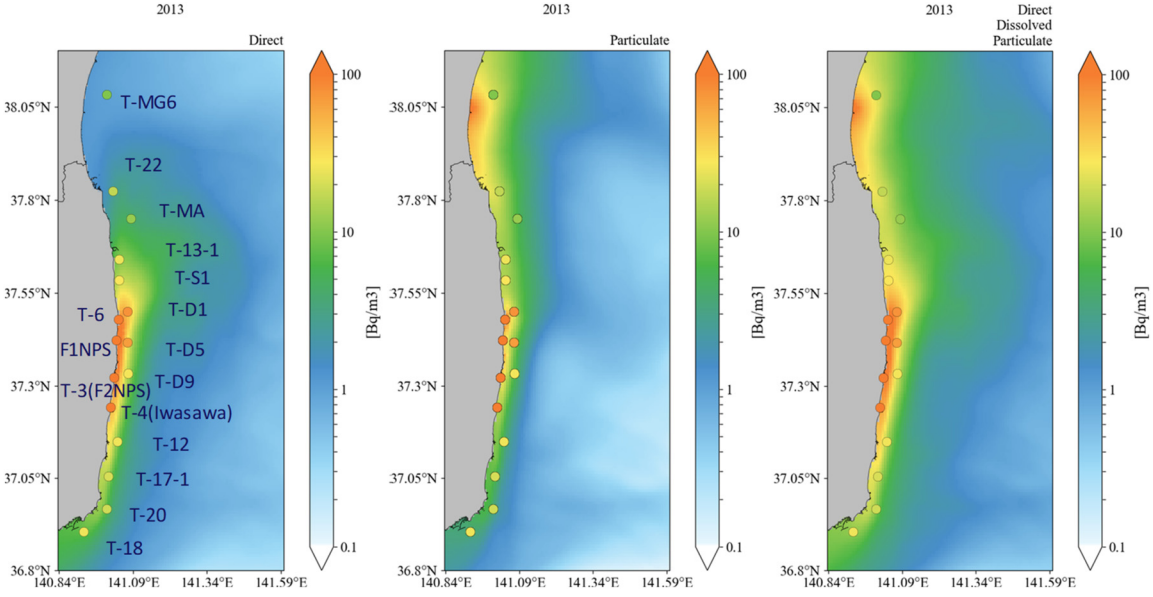

Fig. 3: Comparison of annual average observed (circles) and simulated (contours) distributions of surface ^{137}Cs activity concentrations in 2013. Direct release case (left), particulate ^{137}Cs fluvial discharge case (middle), direct release + dissolved and particulate ^{137}Cs fluvial discharge case (right).

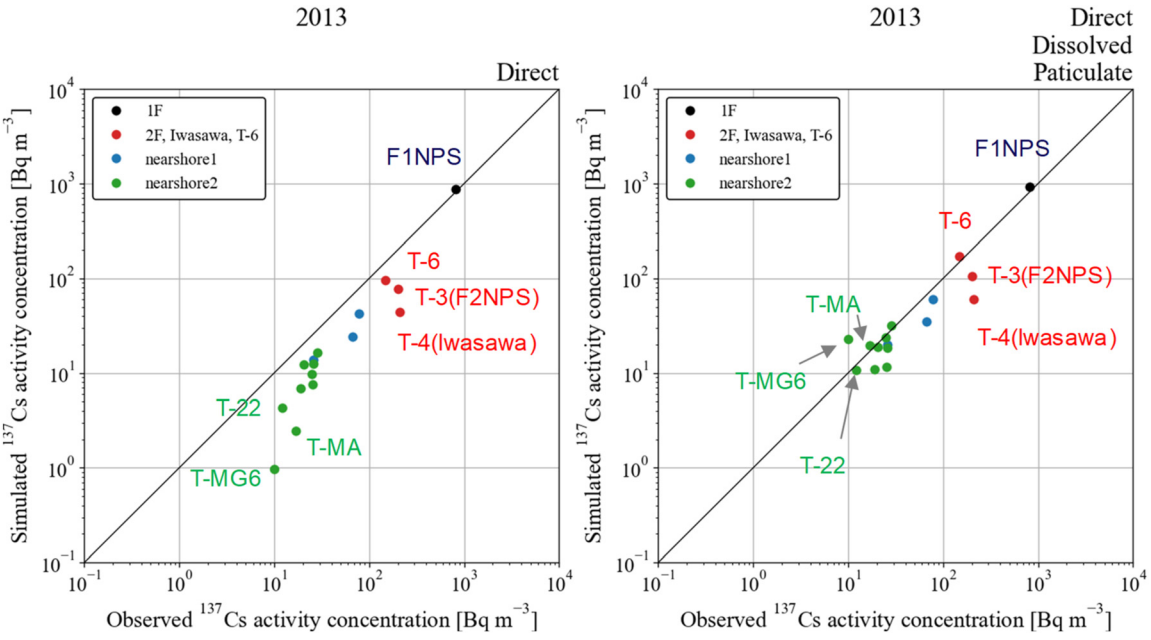

Fig. 4: Comparison of annual average observed and simulated surface ^{137}Cs activity concentrations at each observation point in 2013. Direct release case (left), direct release + dissolved and particulate ^{137}Cs fluvial discharge case (right). 1F (F1NPS), 2F (T-3, T-4, T-6), nearshore1 (T-D1, T-D5, T-D9), and nearshore2 (T-MG6, T-22, T-MA, T-13-1, T-S1, T-12, T-17-1, T-20, T-18).

Table 2: Comparison of annual average observed and simulated surface ^{137}Cs activity concentrations at each observation point in the direct release case and the case of direct release + dissolved and particulate ^{137}Cs fluvial discharge (all). The RSSs (residual sum of squares) were obtained for each year.

	2013			2014			2015			2016		
	Obs	Sim (direct)	Sim (all)	Obs	Sim (direct)	Sim (all)	Obs	Sim (direct)	Sim (all)	Obs	Sim (direct)	Sim (all)
F1NPS	812.3	881.3	918.4	563.6	453.6	491.7	269.3	222.7	256.1	152.5	100.8	120.9
T-3	200.2	77.6	105.9	117.7	39.5	63.8	64.4	20.7	37.3	48.5	11.0	25.5
T-4	211.0	44.6	59.9	99.0	25.7	39.2	61.4	13.4	21.4	42.7	6.9	14.9
T-6	146.9	95.5	171.0	80.9	40.2	134.4	60.7	15.4	100.8	48.3	8.1	50.6
T-D1	78.0	42.4	60.4	21.3	18.9	39.4	13.7	7.7	18.0	9.7	3.6	10.6
T-D5	67.5	24.0	35.0	19.2	12.1	23.8	11.4	6.4	20.8	7.9	3.3	8.8
T-D9	26.0	13.7	19.9	16.3	7.9	14.1	11.1	4.9	16.5	9.3	2.5	7.6
T-MG6	9.9	1.0	22.7	8.6	0.7	18.9	6.1	0.2	11.7	5.3	0.2	6.5
T-22	17.0	2.4	19.5	10.7	1.1	12.7	11.1	0.4	7.0	5.8	0.2	5.8
T-MA	12.1	4.3	10.7	14.2	2.2	8.4	7.7	1.2	4.1	5.0	0.4	2.8
T-13-1	24.9	9.6	23.9	14.1	4.1	11.8	16.1	1.2	8.6	7.0	0.5	5.0
T-S1	28.5	16.3	31.7	18.1	6.7	15.3	13.6	2.6	12.2	12.1	0.9	5.4
T-12	25.8	12.6	18.5	40.6	8.4	14.5	13.5	4.7	9.2	8.6	2.5	7.0
T-17-1	20.6	12.3	18.7	27.8	8.5	14.0	12.7	4.0	7.2	9.1	2.3	6.1
T-20	18.9	6.9	10.9	17.6	4.2	7.8	12.9	2.1	4.1	9.1	1.3	3.7
T-18	25.3	7.6	11.5	15.5	4.0	7.1	9.2	2.4	4.4	7.7	1.2	3.2
RSS		610.8	473.8		447.2	345.4		284.9	185.1		243.1	114.8

relatively good in simulations that considered only direct release and fluvial discharge of dissolved ^{137}Cs [9]. Improvements due to fluvial discharge of particulate ^{137}Cs were apparent at stations in the northern part of the F1NPS. The results were consistent with the scenario that particulate ^{137}Cs discharged from rivers did not

accumulate over a long period of time but was resuspended and re-leached on a timescale of less than 1 year. However, at station T-6 at the mouth of the Ukedo River (~7 km north of the F1NPS), the RSS increased when fluvial effects were taken into account. In contrast, the underestimation was not improved by considering the fluvial supply of particulate ^{137}Cs at stations near the F2NPS (T-3) and on the Iwasawa coast (T-4), which are 10 and 16 km south of the F1NPS, respectively. Those stations (T-3, T-4, and T-6) are close to the coast and require higher resolution to improve reproducibility [8]. There was also no improvement in the reproducibility for T-4 and T-6 in 2013. It should be noted that this underestimation could have resulted from overestimation of the impact of the river and/or the possible impact of dispersion.

2014

The annual average distribution of surface ^{137}Cs activity concentrations as direct release and particulate ^{137}Cs in fluvial discharge and their sum in 2014 is shown in Fig. 5. The ^{137}Cs activity concentration was lower in 2014 than in 2013. As was the case in 2013, the directly released ^{137}Cs was advected to the south. The impact of rivers was significant near the Abukuma River and F1NPS, and from there the impact extended southward.

A comparison of the observed and simulated surface ^{137}Cs activity concentration at each observation point is shown in Fig. 6. The simulated results considering only direct release were underestimated in all cases, but consideration of the supply of particulate ^{137}Cs by the river increased the calculated concentrations, and the reproducibility was improved. The RSS decreased from 439 Bq m^{-3} in the direct release case to 335 Bq m^{-3} in the case of particulate ^{137}C fluvial discharge. As in 2013, the improvement effect was particularly pronounced at the northern stations (T-MG6, T-22, T-MA) and also at T-13-1, due to the influence of the Abukuma River supply. No improvement was observed on the southern coast (T-3, T-4).

2015

The annual average distribution in 2015 of surface ^{137}Cs activity concentrations due to direct release and particulate ^{137}Cs in fluvial discharge and their sum is shown in Fig. 7. The reduction of the rate of direct release resulted in a more narrowly defined impact.

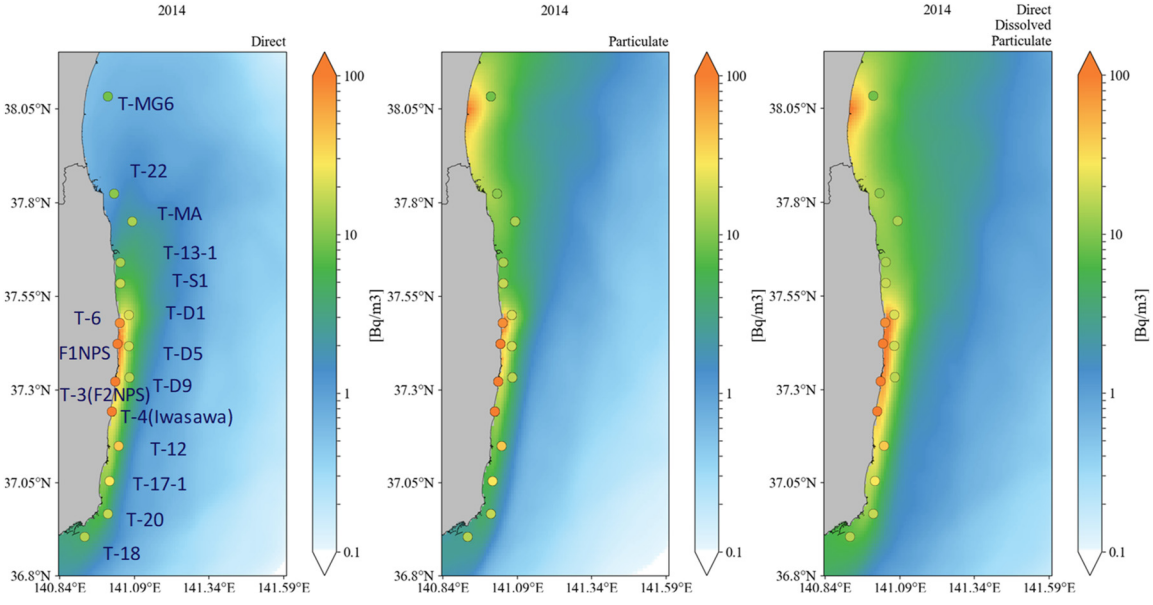

Fig. 5: Comparison of annual average distributions of observed (circles) and simulated (contours) surface ^{137}Cs activity concentration in 2014. Direct release case (left), particulate ^{137}Cs fluvial discharge case (middle), direct release + dissolved and particulate ^{137}Cs fluvial discharge case (right).

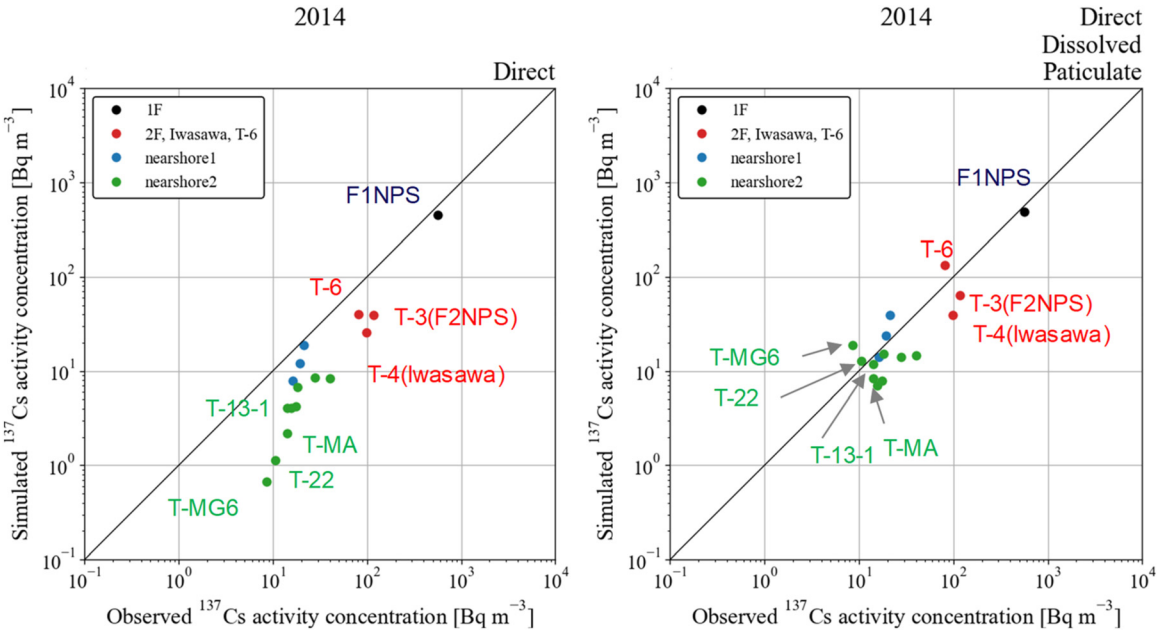

Fig. 6: Comparison of annual average observed and simulated surface ^{137}Cs activity concentrations at each observation point in 2014. Direct release case (left), direct release + dissolved and particulate ^{137}Cs fluvial discharge case (right). 1F (F1NPS), 2F (T-3, T-4, T-6), nearshore1 (T-D1, T-D5, T-D9), and nearshore2 (T-MG6, T-22, T-MA, T-13-1, T-S1, T-12, T-17-1, T-20, T-18).

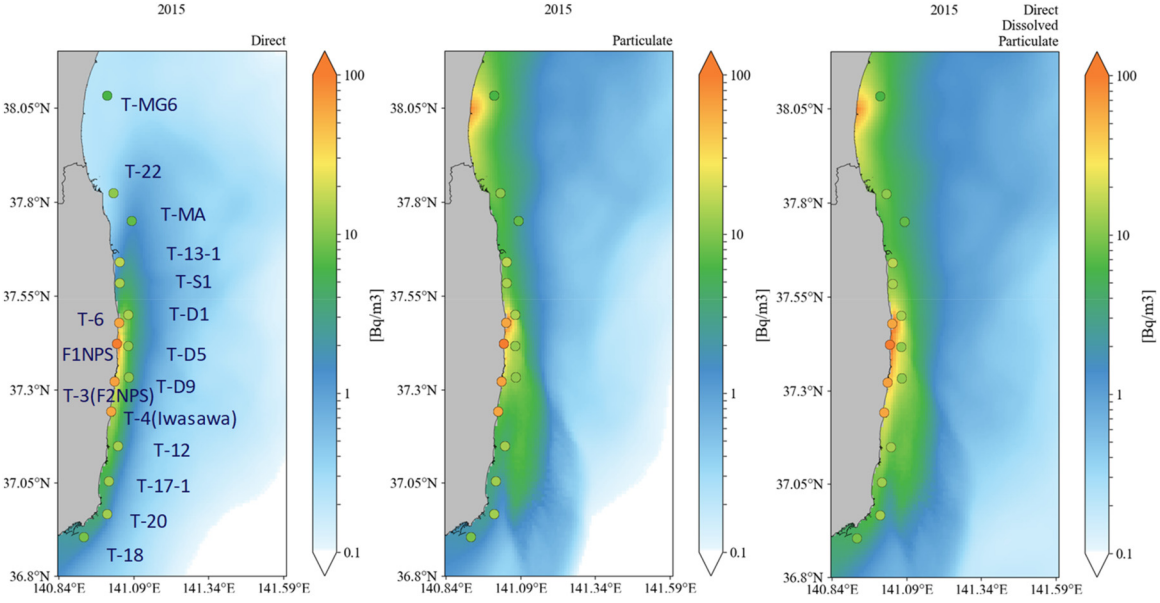

Fig. 7: Comparison of annual average distributions of observed (circles) and simulated (contours) surface ^{137}Cs activity concentrations in 2015. Direct release case (left), particulate ^{137}Cs fluvial discharge case (middle), and direct release + dissolved and particulate ^{137}Cs fluvial discharge case (right).

A comparison of observed and simulated surface ^{137}Cs activity concentration at each observation point is shown in Fig. 8. The RSS decreased from 279 Bq m^{-3} in the direct release case to 180 Bq m^{-3} in the case of particulate ^{137}C fluvial discharge. As in 2013 and 2014, the improvement effect was particularly pronounced at the northern stations (T-MG6, T-22, T-MA) and also at T-13-1, due to the influence of the Abukuma River supply.

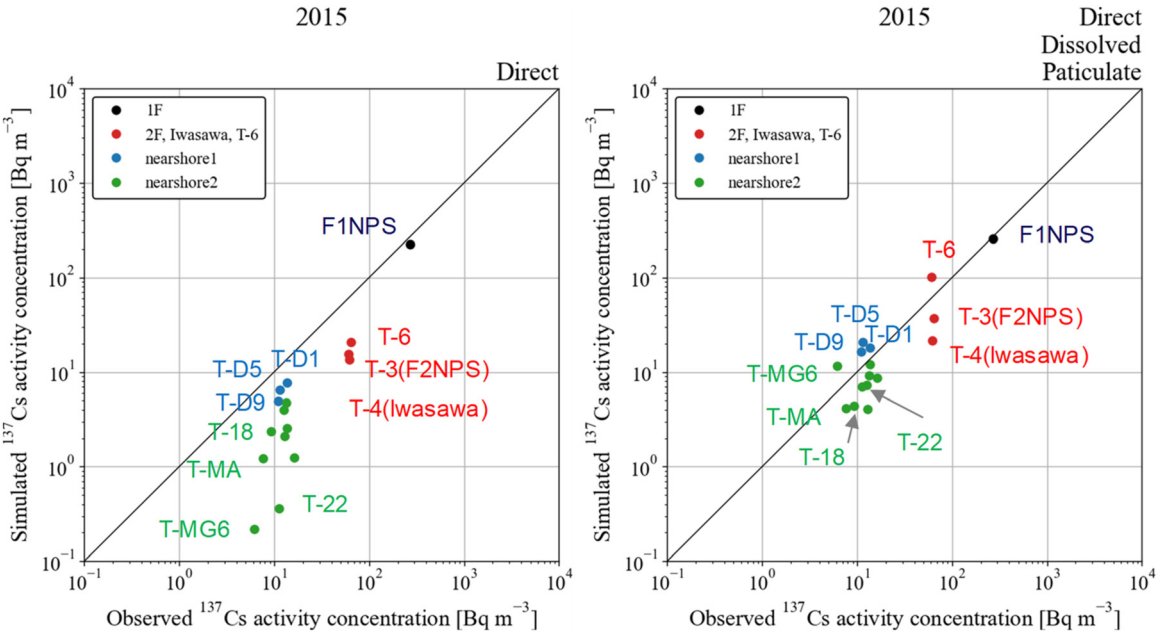

Fig. 8: Comparison of annual average observed and simulated surface ^{137}Cs activity concentrations at each observation point in 2015. Direct release case (left), direct release + dissolved and particulate ^{137}Cs fluvial discharge case (right). 1F (F1NPS), 2F (T-3, T-4, T-6), nearshore1 (T-D1, T-D5, T-D9), nearshore2 (T-MG6, T-22, T-MA, T-13-1, T-S1, T-12, T-17-1, T-20, T-18).

No improvement was observed on the southern coast (T-3, T-4). Improvement of the reproducibility at stations along the coast (T-3, T-4, and T-6) remained a challenge. The ^{137}Cs activity concentrations at the nearshore1 stations (T-D1, T-D5, and T-D9), which were further offshore than the nearshore2 stations, were slightly overestimated by consideration of the fluvial particulate ^{137}Cs discharge. In contrast, at the nearshore2 stations, the reproducibility was improved by considering fluvial particulate ^{137}Cs discharge.

2016

The annual average distribution in 2016 of surface ^{137}Cs activity concentrations as direct release and fluvial particulate ^{137}Cs discharge and their sum is shown in Fig. 9. There was a decrease in the area of impact of the fluvial particulate ^{137}Cs discharge. The rate of decrease of fluvial discharge was smaller than the decrease for direct release from 2013 to 2016 (Fig. 2). There was no decrease after July 2016 in the observed ^{137}Cs activity concentration adjacent to the F1NPS, which remained almost constant [4]. The implication is that there was no decrease in the direct release rate after July 2016. In contrast, the fluvial particulate ^{137}Cs discharge was decreasing, although there was a large variation in the apparent half-life, from 3 to 25 years. The impact of fluvial particulate ^{137}Cs discharge was larger than that of direct releases in 2016.

A comparison of observed and simulated surface ^{137}Cs activity concentrations at each observation point is shown in Fig. 10. The RSS decreased from 238 Bq m^{-3} in the direct release case to 114 Bq m^{-3} in the case of fluvial particulate ^{137}C discharge. In the case of direct release only, the underestimation was greater for the northern stations (T-MG6, T-22, T-MA and T-13-1). However, the improvement effect of the Abukuma River supply was significant. However, the Abukuma River estuary (T-MG6) was still slightly underestimated. Despite the overall good agreement, the underestimation in the coastal area south of the F1NPS (T-3, T-4) was not improved, suggesting the need for higher resolution.

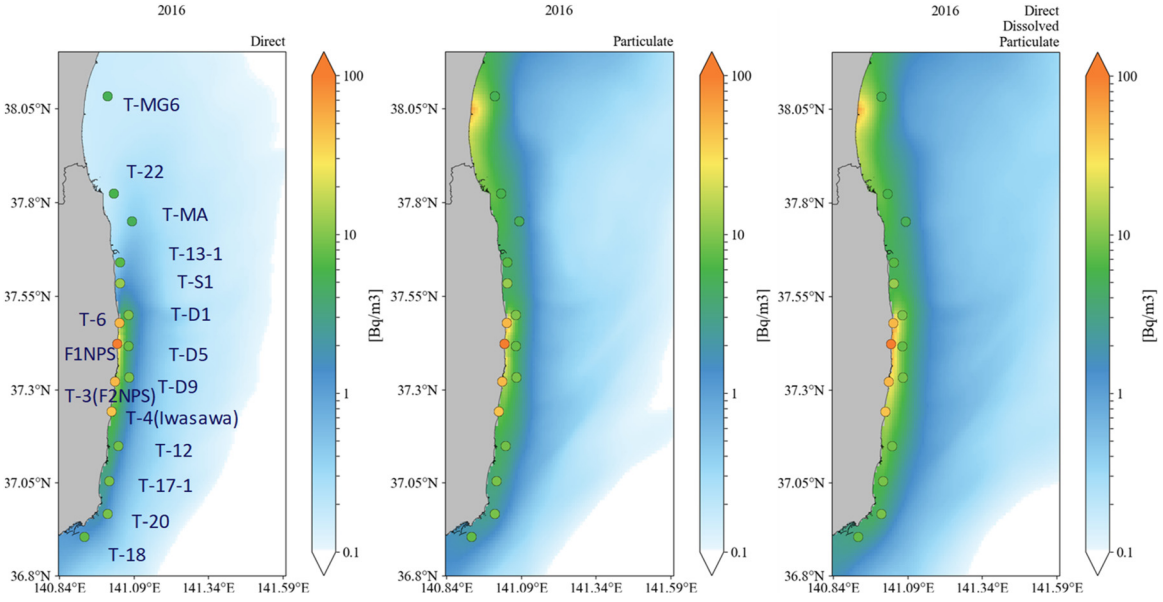

Fig. 9: Comparison of annual average distributions of observed (circles) and simulated (contours) surface ^{137}Cs activity concentrations in 2016. Direct release case (left), particulate ^{137}Cs fluvial discharge case (middle), direct release + dissolved and particulate ^{137}Cs fluvial discharge case (right).

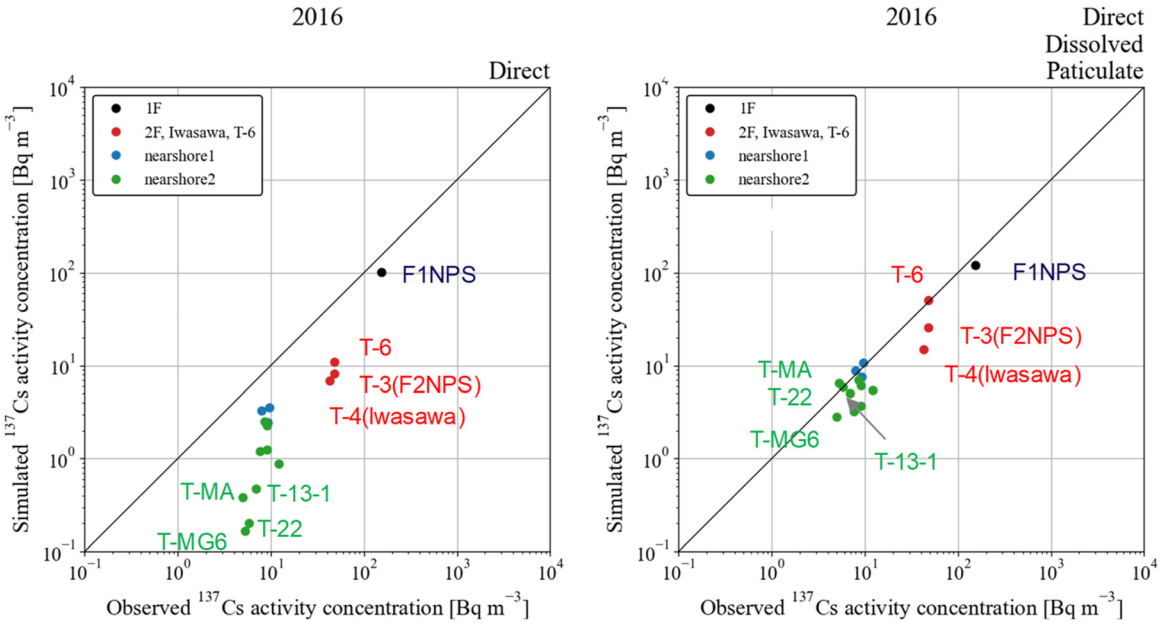

Fig. 10: Comparison of annual average observed and simulated surface ^{137}Cs activity concentrations at each observation point in 2016. Direct release case (left), direct release + dissolved and particulate ^{137}Cs fluvial discharge case (right). 1F (F1NPS), 2F (T-3, T-4, T-6), nearshore1 (T-D1, T-D5, T-D9), nearshore2 (T-MG6, T-22, T-MA, T-13-1, T-S1, T-12, T-17-1, T-20, T-18).

Impacts of fluvial discharge of particulate ^{137}Cs

The discharge of fluvial particulate ^{137}Cs increased the ^{137}Cs activity concentrations in the area of the sea where the ^{137}Cs activity concentrations were underestimated in the direct release case. This improvement of the reproducibility at all stations suggested that the fluvial discharge of particulate ^{137}Cs affected the observed surface

^{137}Cs activity concentrations. Table 3 shows the ratio of the impact of the fluvial particulate ^{137}Cs discharge on the overall ^{137}Cs activity concentration in the simulated results. The impact at the F1NPS stations was ~4 % in 2013. It then increased to 7 %, 12 %, and 15 % in 2014, 2015, and 2016, respectively, based on rates of direct release estimated from F1NPS observations. The implication is that in 2016 the ^{137}Cs activity concentration may have been overestimated by 15 % because of riverine impacts. However, it can be said that impacts of direct release were dominant from 2013 to 2016. The estimated amount of direct release was 3.5 ± 0.7 PBq by the end of May 2011 [3], and it had increased to 3.6 ± 0.7 PBq by the end of 2012 [8]. Direct release continued, but the total amount did not increase until the end of 2019 [4]. The error in fitting the initial observations has been estimated to be less than 20 % [3]. The impact of fluvial particulate ^{137}Cs discharge (15 %) was smaller than this error. The implication is that there was little need to consider fluvial effects when using the observed results of ^{137}Cs near the F1NPS for the estimation of direct release in 2013–2016. Similarly, there is little need to consider impacts of fluvial particulate ^{137}Cs discharge from 2017 onwards because the direct release rate remained unchanged, and the relative fluvial particulate ^{137}Cs discharge decreased.

For stations along the coast, T-3 (F2NPS), T-4 (Iwasawa coast), and the mouth of the T-6 at Ukedo River mouth, the contribution to the ^{137}Cs activity from advection increased from 25 % in 2013 to more than 50 % in 2016 for southern T-3 and T-4. However, this contribution was always an underestimate, and we considered it necessary to improve the simulation of advection through, for example, higher resolution [8]. In contrast, at T-6 at the mouth of the Ukedo River in the northern part of the FINPS, the ratio of particulate ^{137}Cs fluvial discharge increased from 40 % to 80 % between 2013 and 2016. As a result, reproducibility improved, but the ratio was sometimes overestimated. Similar results were observed at stations slightly offshore (T-D1, T-D5, and T-D9) but at T-D5 the fluvial influence decreased from 2015 to 2016. This decrease may have been due to differences in the advection process and will require more detailed analysis with a higher resolution model in the future.

In 2013, the ratio of fluvial particulate ^{137}Cs discharge at the northern station in front of Abukuma river mouth (T-MG6) was 89 %. The ratio of particulate ^{137}Cs fluvial discharge was also 40–60 % for T-MA, T-13-1, and T-S1. At stations slightly offshore to the south (T-D1, T-D5, and T-D9), the fluvial influence was less than 30 %. At points further south (T-17-1, T-20, and T-18), it was slightly larger, 33–35 %. This trend was maintained, and the fluvial influence increased until 2016. The fluvial influence at T-MG6 was 97 %, and the impact of direct release was therefore very small. To accurately replicate the observed ^{137}Cs concentrations off the Fukushima coast, it is evident that the particulate ^{137}Cs fluvial discharge must be considered. From 2017 onwards, direct release remained unchanged, and particulate ^{137}Cs fluvial discharge decreased. However, even at this point in time, the ^{137}Cs activity concentrations have not decreased to the levels prior to the F1NPS accident, and there is a need to continue to consider direct release and riverine impacts.

Table 3: Ratio of the impact of particulate ^{137}Cs fluvial discharge on the overall ^{137}Cs activity concentration in the simulated results.

	2013	2014	2015	2016
F1NPS	0.04	0.07	0.12	0.15
T-3	0.25	0.35	0.41	0.53
T-4	0.24	0.32	0.34	0.50
T-6	0.40	0.64	0.79	0.77
T-D1	0.28	0.49	0.54	0.62
T-D5	0.30	0.46	0.67	0.59
T-D9	0.30	0.96	0.68	0.64
T-MG6	0.96	0.96	0.98	0.97
T-22	0.85	0.88	0.91	0.93
T-MA	0.58	0.71	0.67	0.84
T-13-1	0.57	0.62	0.82	0.87
T-S1	0.46	0.52	0.75	0.79
T-12	0.30	0.40	0.46	0.61
T-17-1	0.33	0.37	0.41	0.60
T-20	0.35	0.44	0.46	0.63
T-18	0.33	0.40	0.43	0.59

Summary

The ^{137}Cs activity concentrations in coastal areas exceeded background concentrations for several years after the F1NPS accident. This exceedance may have been due to ongoing direct releases from the site and the fluvial discharge of ^{137}Cs that deposited on land. It is necessary to assess the combined effects of discharges from multiple rivers and direct releases through the process of dispersion in the ocean. Ocean dispersion simulations that considered direct releases and river discharges were therefore carried out from 2013 to 2016. The simulation assumed that particulate ^{137}Cs from rivers was dispersed directly into the ocean without a time lag. The reproducibility of the annual mean ^{137}Cs activity concentration was improved by considering the fluvial discharge of particulate ^{137}Cs in addition to direct releases. Particularly at sites north of the F1NPS, where the impact of direct releases was small, the impacts of discharge from the Abukuma River were more pronounced and improved the reproducibility. We found that in 2016, riverine influences accounted for more than 50 % of the total ^{137}Cs activity at all stations referred in this study, except for the F1NPS.

We confirmed that ^{137}Cs discharged from rivers to the ocean coagulated and precipitated and had only a low potential for long-term deposition. There were no observations of long-term increases of ^{137}Cs activity concentrations in estuarine sediments due to particulate ^{137}Cs supplied from rivers to the ocean along the Fukushima coast [32].

The impact of fluvial discharge on observed ^{137}Cs activity concentrations adjacent to the F1NPS, which are used to estimate direct release rates, was 15 % in 2016. We confirmed that it is not necessary to consider the impact of fluvial discharge to estimate direct release rates.

This study pointed out that most of the ^{137}Cs discharged from rivers to the ocean coagulates and precipitates as particulate ^{137}Cs and is subsequently observed at the surface on a timescale of at least 1 year or less. The processes of resuspension and re-leaching by coagulation-precipitation of particulate ^{137}Cs discharged from rivers to the ocean are complex. It is clear that these chemical processes have an impact on the ocean. This impact is an issue that needs to be resolved for future fluvial discharges along the Fukushima coast. In this study, the reproduction of annual mean values was targeted in order to get a complete picture [9]. In the future, attention should also be paid to temporal changes of ^{137}Cs activity concentrations in the ocean after heavy rainfalls.

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