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Continuing ^{137}Cs release to the sea from the Fukushima Dai-ichi Nuclear Power Plant through 2012

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Abstract. The rate of cesium-137 (^{137}Cs) release to the sea from the Fukushima Dai-ichi Nuclear Power Plant for the period until September 2012 was estimated. Publicly released data on ^{137}Cs radioactivity in seawater near the power plant by Tokyo Electric Power Company strongly suggest a continuing release of radionuclides to the sea. The plant has an artificial harbour facility, and the exchange rate of harbour water with surrounding seawater was estimated by the decrease in radioactivity immediately after an intense radioactive water release. The estimated exchange rate of water in the harbour was 0.44 d^{-1} during the period from 6 to 19 April. The ^{137}Cs radioactivity in the harbour water was substantially higher than that of seawater outside and remained relatively stable after June 2011. A quasi-steady state was assumed with continuous water exchange, and the average release rate of ^{137}Cs was estimated to be 93 GBq d^{-1} in summer 2011 and 8.1 GBq d^{-1} in summer 2012.

1 Introduction

The Fukushima Dai-ichi Nuclear Power Plant released a significant amount of radionuclides into the environment from crippled reactors that lost cooling capabilities following the earthquake and tsunami on 11 March 2011. Because the plant is located on the Pacific coast, a major portion of the radionuclides released to the atmosphere was deposited into the sea (Morino et al., 2011). Radionuclides were also transferred to the sea via direct release of radioactive water, which started at the end of March, peaked during early April and continued in smaller amounts at least through May (Tsumune et al., 2012). The direct release had a large effect on the coastal environment (Tsumune et al., 2012); however, the period of intense release was relatively short and the open nature of the Fukushima coast resulted in rapid flushing of radionuclides away from the coastal seawater (Yoshida and Kanda, 2012).

Owing to the rapid flushing, radioactivity in coastal seawater declined sharply in April and May of 2011; however, the decline was apparently curbed after June and the remaining radioactivity at a relatively higher level near the power plant indicates a continuous release of radionuclides from the power plant (Buesseler et al., 2011). Additionally, radioactivity in coastal sediment and marine biota was still present in 2012, which may be the result of a continuous input of radionuclides from the plant and/or from river and groundwater discharge (Buesseler, 2012).

In this study, cesium-137 (^{137}Cs) radioactivity in seawater around the power plant was examined. The measured ^{137}Cs radioactivity in the harbour facility of the power plant was especially informative and was used to determine an exchange rate of harbour water with outer seawater. The exchange rate and relatively stable ^{137}Cs radioactivity inside the harbour enabled estimation of the continuous ^{137}Cs release from the harbour to the sea.

2 Materials and methods

The data used in this study were publicly released by the Tokyo Electric Power Company (TEPCO) and are available on websites of TEPCO (TEPCO, 2012) and the Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan (MEXT, 2012). ^{137}Cs radioactivity in seawater was measured by TEPCO under direction of the national government by applying water samples directly to gamma

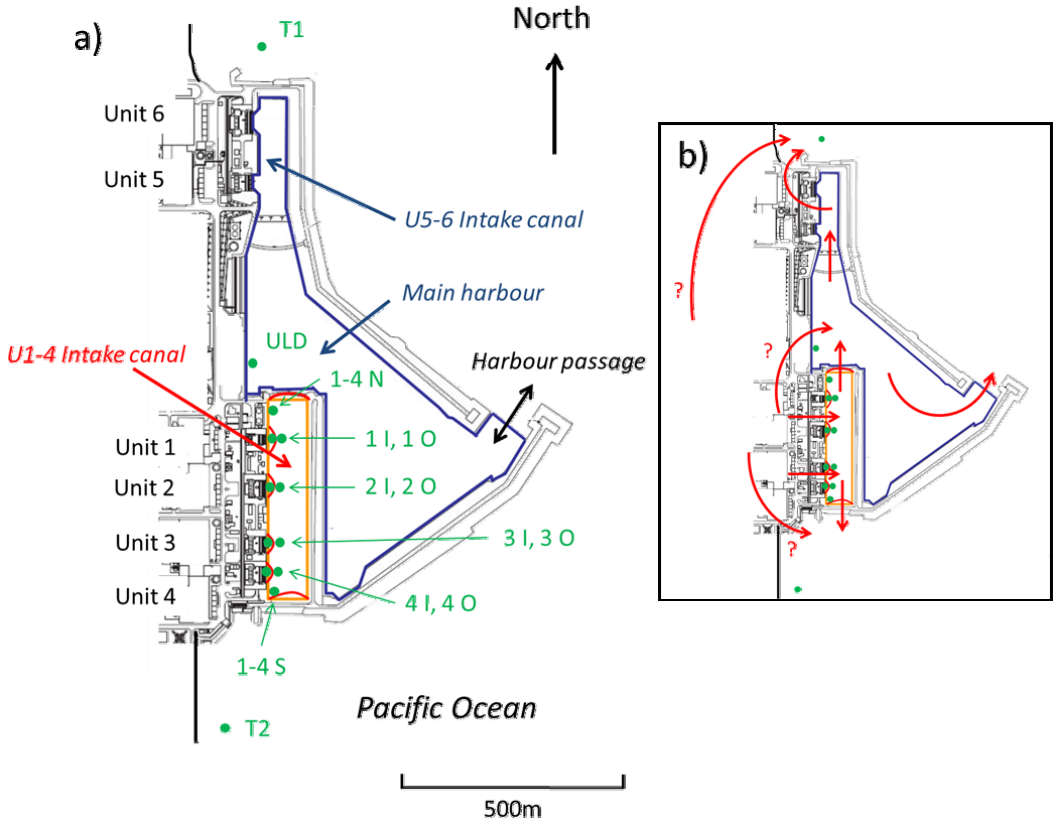

Fig. 1. (a) Artificial harbour facility of the Fukushima Dai-ichi Nuclear Power Plant and TEPCO monitoring points (redrawn from a TEPCO document ; TEPCO, 2011), **(b)** possible routes of radioactive water transfer. T1: north discharge gate, ULD: unloading dock, 1–4 N: north of Unit 1–4 intake, 1 I: inside Unit 1 screen, 1 O: outside Unit 1 screen, 2 I: inside Unit 2 screen, 2 O: outside Unit 2 screen, 3 I: inside Unit 3 screen, 3 O: outside Unit 3 screen, 4 I: inside Unit 4 screen, 4 O: outside Unit 4 screen, 1-4 S: south of Unit 1-4 intake, T2: south discharge gate.

spectrometers (Government of Japan, 2011). Figure 1a shows a map of facilities of the Fukushima Dai-ichi Nuclear Power Plant and TEPCO's seawater monitoring points.

As shown in Fig. 1a, the power plant has an artificial harbour facility composed of a pair of jetties for maritime transportation. Seawater for cooling purposes was designed to be drawn from intake canal areas in the harbour. Two intake canal areas are located in front of the housings for the reactor units, one in the southern part of the harbour for Units 1, 2, 3 and 4, and another in the northern part for Units 5 and 6. Thus, the harbour water can be separated into three areas: the main harbour area, the Unit 1–4 intake canal area, and the Unit 5–6 intake canal area. The intake canals are surrounded by jetties and connected to the main harbour water through curtain wall facilities. Water in the main harbour area is exchanged with outer seawater through a harbour passage. While seawater intake for the damaged Units 1 through 4 has been stopped, cooling water is still taken continuously from the intake canal for the two undamaged reactors (Units 5 and 6). The cooling water for these units is then released to the outer sea through a discharge gate located north of the plant.

Possible routes of the radioactive water around the plant are depicted in Fig. 1b. The majority of the radioactive water presumably originates from damaged Units 1 through 4. Some of the radioactive water was confirmed to flow into the Unit 1–4 intake canal (Government of Japan, 2011). From the Unit 1–4 intake canal, radioactive water likely diffused into the main harbour, and subsequently out to the sea through the harbour passage and the north discharge gate. The jetty between the Unit 1–4 intake canal and the outer sea was damaged by the earthquake and/or tsunami (ICANPS, 2011), and the radioactive water was likely released to the outer sea through the damaged portion of the jetty. Release that bypasses the Unit 1–4 intake canal or discharges either directly into the main harbour or the outer sea has not been documented. However, because the south discharge gate area facing the outer sea and the unloading dock area in the main harbour are both relatively close to the damaged reactor units, the possibility of such direct release cannot be ruled out.

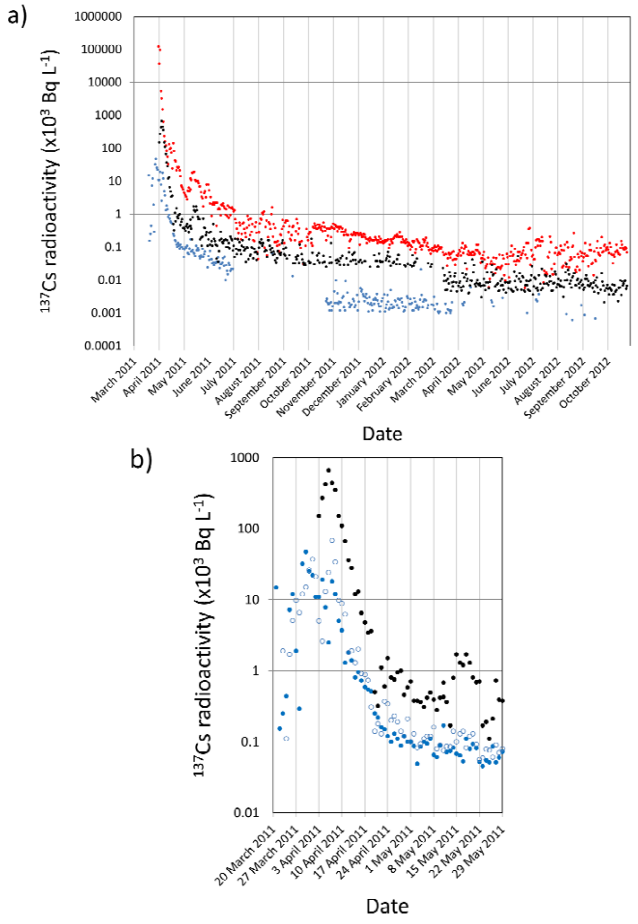

Fig. 2. (a) ^{137}Cs radioactivity in seawater taken at monitoring points T2 (blue dots), ULD (black dots), and 2I (red dots) for March 2011 through October 2012. The vertical grid line indicates date intervals of one month. **(b)** An expansion of ^{137}Cs radioactivity at monitoring points ULD and T2 for the period from 20 March through 29 May 2011, together with data obtained at T1 (open blue circles). The vertical gridline indicates intervals of one week.

3 Results and discussion

3.1 Variation of ^{137}Cs radioactivity around the harbour

Figure 2a shows seawater ^{137}Cs observed on the inside of the Unit 2 screen (monitoring point 2I) in the Unit 1–4 intake canal area, at the unloading dock (monitoring point ULD) in the main harbour area, and off the south discharge gate (monitoring point T2) outside of the harbour (Fig. 1a) from March 2011 to October 2012. Figure 2b is an expansion for the period from 20 March through 29 May 2011 showing data at ULD and T2 together with data obtained at T1 near the north discharge gate (Fig. 1a). Seawater sampling was started on 21 March at T2 and on 23 March at T1. Sampling at 2I and ULD was started on 2 April and 3 April, respectively. There are several other sampling points in the Unit 1–4 intake canal, but sampling was not started at these stations until late April or early May 2011. ULD is the only point at which seawater has been sampled daily in the main harbour area.

^{137}Cs radioactivity in seawater outside the harbour (T1 and T2) shows distinct elevations at the end of March through early April in 2011, which likely correspond to major release events of radioactive water (Fig. 2b). The largest leak event among those officially confirmed by TEPCO took place from 1 to 6 April at the cooling water intake in front of Unit 2 (Government of Japan, 2011). TEPCO estimated that approximately 520 m^3 of water and 0.94 PBq of ^{137}Cs were released (Government of Japan, 2011). The increase of radioactivity in March before the April leak is not related to the confirmed release events. Because data describing radioactivity in the harbour in March are not available, the route of the radioactivity release cannot be assessed. The ^{137}Cs radioactivity in March was higher at T2 than at T1 (Fig. 2b); thus, a release into the sea south of the harbour may have prevailed during this period. The highest radioactivity at T2 was recorded on 30 March, with two smaller peaks of radioactivity following on 4 and 7 April. At T1, a radioactivity peak was observed on 1 April, but the radioactivity was lower than the peak radioactivity observed on 30 March at T2. Monitoring from 3 April at ULD clearly showed that ^{137}Cs radioactivity increased sharply until 6 April, then began decreasing exponentially (Fig. 2b). This drastic change on 6 April must have reflected a sudden decrease in ^{137}Cs input to the main harbour, and the only corresponding event was TEPCO's operation that stopped the leak in front of Unit 2 early on 6 April (Government of Japan, 2011). Following the radioactivity peak at ULD on 6 April, the largest peak of radioactivity appeared at T1 near the north discharge gate the next day (7 April) (Fig. 2b). The peak radioactivity at T1 on 7 April was much higher than that on 1 April. A peak of radioactivity was also identified at T2 on 7 April, but the level of radioactivity in this peak was smaller than those recorded earlier at T2. The order of peak appearance indicates a successive transfer of the radioactive water from the main harbour to the sea. Another leak event occurred on 10–11 May at the intake of Unit 3, and ^{137}Cs released at this time was estimated to be 9.8 TBq (Government of Japan, 2011). This event is reflected in a slight elevation of harbour water radioactivity in May (Fig. 2b). The 10–11 May and the 1–6 April events are the only leak events that have been officially confirmed by TEPCO for the period from March to May 2011. TEPCO also deliberately discharged water with moderate radioactivity (total volume of 10 393 m^3 and 0.042 TBq of ^{137}Cs) from 4 to 10 April (Government of Japan, 2011). This water was released near the south discharge gate (Fig. 1a) to the outer sea, not into the harbour water.

^{137}Cs radioactivity at 2I in the Unit 1–4 intake canal was consistently larger than that at ULD in the main harbour (Fig. 2a). Moreover, ^{137}Cs radioactivity at ULD was consistently larger than at T1 and T2 in the outer sea (Fig. 2a, b). The average ^{137}Cs radioactivity at monitoring points in the Unit 1–4 intake canal and at ULD is shown in Table 1

Table 1. Average ^{137}Cs radioactivity in seawater around the Fukushima Dai-ichi Nuclear Power Plant (Bq L^{-1}, ± standard deviation). Values reported as ND (not detected) were excluded from the calculations.

	Harbour	Unit 1–4 Intake Canal									
	ULD	1–4 N	1 O	1 I	2 O	2 I	3 O	3 I	4 O	4 I	1–4 S
1 June–31 August 2011	114 ± 68	313 ± 232	305 ± 194	340 ± 200	318 ± 195	1030 ± 1320	391 ± 240	1650 ± 1680	404 ± 240	732 ± 398	391 ± 205
1 April–30 September 2012	9.9 ± 6.9	17.1 ± 8.7	17.6 ± 8.0	22.3 ± 10.8	20.2 ± 10.1	68.1 ± 53.1	35.6 ± 29.6	209 ± 189	56.0 ± 37.0*	83.4 ± 57.1	47.1 ± 27.6

* Likely overestimated because a significant amount of data are reported as ND.

for the periods from 1 June through 31 August 2011 and 1 April through 30 September 2012. ^{137}Cs radioactivity at other points in the Unit 1–4 intake canal area was also higher than at ULD (Table 1). The ^{137}Cs radioactivity levels inside the screened intake facilities (or in water closer to the reactor housing) are consistently higher than those outside of the screen (Table 1). Following the leak event of 1–6 April, TEPCO placed silt fences at the screen facility of each reactor intake and at the curtain screen between the main harbour and the Unit 1–4 intake canal on 11–14 April (ICANPS, 2011). These fences may have retarded exchange between the canal water and the inner water of the intake, as well as exchange between the main harbour water and the canal water. Nevertheless, the consistent gradient of radioactivity between these waters indicates that radionuclides are still being released continuously from somewhere around the reactor housings.

Figure 3 shows the ^{137}Cs radioactivity in seawater collected at monitoring points 1I, 2I, 3I and 4I (inside the screen of the intake for Units 1, 2, 3 and 4) for November 2011 through October 2012. Periodic elevations of ^{137}Cs radioactivity were observed after March 2012 at all of these monitoring points, while the amplitude and frequency of the radioactivity fluctuation varied. The reason for the appearance of such fluctuation of radioactivity after March 2012 is not clear. Pavement of the seabed of the Unit 1–4 canal was conducted by TEPCO from 14 March through 11 May 2012 (ICANPS, 2011), but the radioactivity fluctuation cannot be explicitly related to the pavement operation or its completion. The occasional elevations of radioactivity at those monitoring points located closer to the reactor housings strongly indicate that events of radioactivity input are occurring. Thus, both the gradient and the fluctuation in ^{137}Cs radioactivity support the view that ^{137}Cs is still being released to the intake canal.

3.2 Evaluation of water exchange in the plant harbour

The exchange rate of water in the main harbour area was estimated by the decrease of radioactivity immediately after the intense release of highly radioactive water on 1–6 April. The sharp decline of ^{137}Cs radioactivity at ULD after this release event fits an exponential curve well (Fig. 4a). Thus, this decrease is described by the following simple first-order model:

$$\frac{dC}{dt} = -kC, \tag{1}$$

where C is the ^{137}Cs radioactivity in the harbour water, t is the time in days, and k is a rate constant. Curve fitting for 6 to 19 April yielded a value of 0.44 d^{-1} for k. A coefficient of determination (R^2) of 0.9841 was obtained in this regression, and the 95 % confidence interval of the estimated k was between −0.40 and −0.47. If the ^{137}Cs release after 6 April is negligibly small with respect to the ^{137}Cs in the main harbour, and if water exchange is the only process of ^{137}Cs removal from the harbour water, k corresponds to an exchange rate of the harbour water with the outer seawater.

The mass balance of ^{137}Cs in the harbour water can be expressed as

$$V\frac{dC}{dt} = I - kVC, \tag{2}$$

where V is the volume of the harbour water, and I is the input of ^{137}Cs to the harbour water. At steady state, the ^{137}Cs input or I can be given as

$$I = kVC. \tag{3}$$

A value of V is implicitly shown in a TEPCO document (TEPCO, 2011) in which both the average radioactivity per unit of water volume and an estimated total radioactivity are shown for the harbour area. The water volume of the main harbour area plus the Unit 5–6 intake canal area (the area shown in a blue line in Fig. 1) was calculated to be 1.88×10^6 m^3 (see Supplement). The area was estimated to be about 2.7×10^5 m^2 on a map; thus, the average depth was calculated to be 7.0 m. While a detailed depth contour of the harbour was not available to the author, the harbour passage is known to have a water depth of 8 m (Tsuzuki et al., 1992), and the water depth at the unloading dock is reportedly 6.5 m (Fukushima Prefecture Government, 2013). Thus, the estimated volume and average depth are reasonable. Because cooling water is continuously drawn from the main harbour through the Unit 5–6 intake canal, use of the combined water volume of the main harbour area and the Unit 5–6 intake canal as V can be warranted.

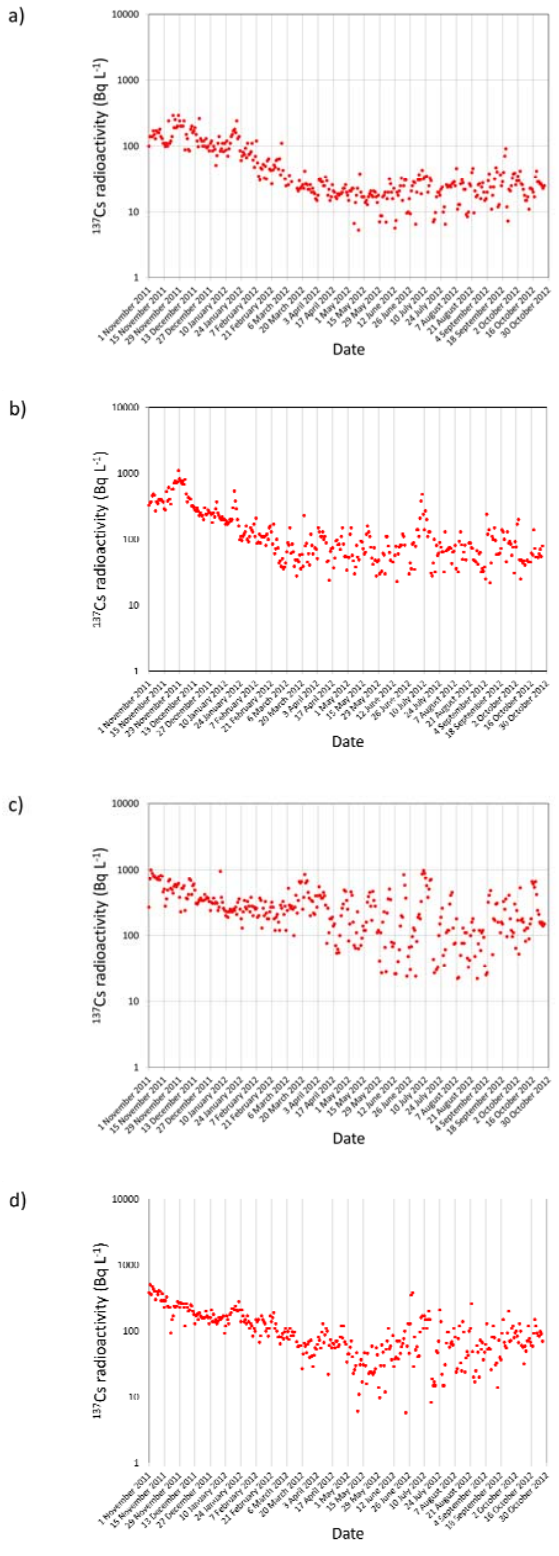

Fig. 3. ^{137}Cs radioactivity in seawater collected at 1I **(a)**, 2I **(b)**, 3I **(c)** and 4I **(d)** for the period from November 2011 through October 2012. The vertical gridline indicates intervals of two weeks.

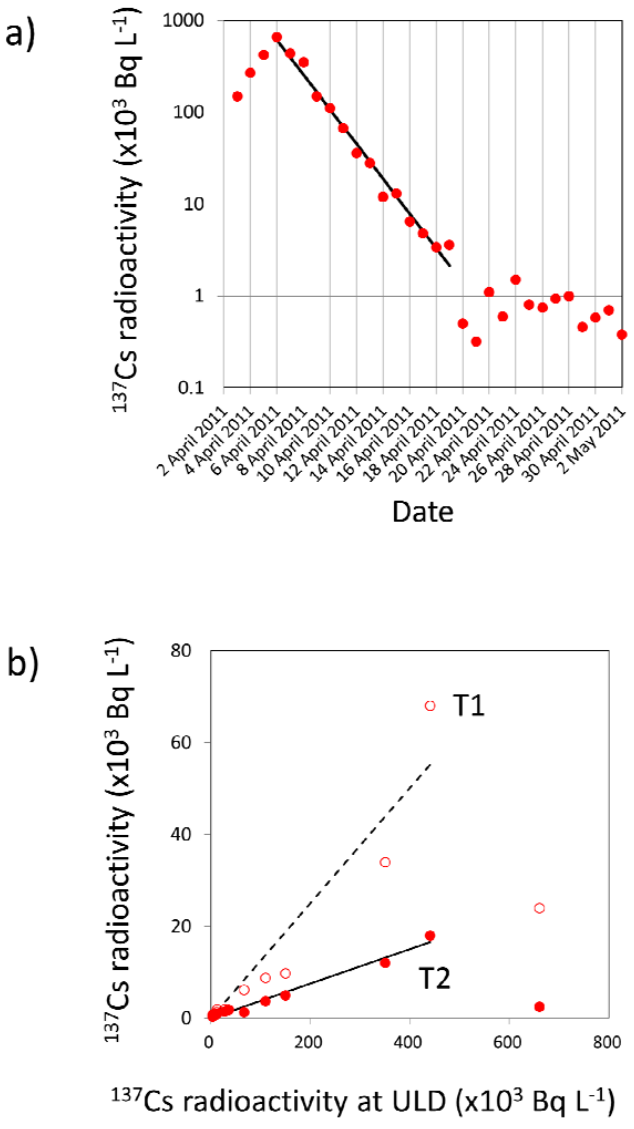

Fig. 4. (a) ^{137}Cs radioactivity at ULD in April 2011. Solid line indicates a least square fitting of an exponential curve for 6 April through 19 April. **(b)** ^{137}Cs radioactivity at T1 (open circles) and at T2 (closed circles) plotted against radioactivity at ULD for 6–19 April. The linear regression lines were obtained by excluding values on 6 April and generated a slope of 0.125 for T1 and a slope of 0.038 for T2.

If ^{137}Cs input after 6 April is significant in the main harbour, the value of k will underestimate the water exchange rate. As described above, the rapid increase of ^{137}Cs radioactivity at ULD suddenly turned into a rapid decrease on 6 April (Fig. 4a), and hence a sudden and substantial reduction of ^{137}Cs input to the main harbour is inferred. If we assume that ^{137}Cs input to the main harbour continued at a constant rate after 6 April and that the removal of ^{137}Cs from the harbour is proportional to the ^{137}Cs radioactivity in the harbour as suggested in Eq. (2), the decrease of ^{137}Cs in the main

harbour would have continued until the removal rate was reduced to balance the input rate. The decrease of ^{137}Cs in the main harbour continued at least until the radioactivity was below 10 % of the average level during 6–19 April (Fig. 4a); thus, the balanced rate of input and removal was less than 10 % of the inferred average removal rate during 6–19 April. Accordingly, it is assumed that underestimation of the water exchange should be less than 10 %. When water in the surrounding sea has a non-negligible value of ^{137}Cs radioactivity, the water exchange will also be underestimated. Figure 4b shows the ^{137}Cs radioactivity at T1 and T2 plotted against radioactivity at ULD for the period from 6 to 19 April. The lower radioactivity at T1 and T2 on April 6 likely reflected the lower radioactivity in the port water before the peak on 6 April. Excluding values on 6 April, the linear regression suggests that ^{137}Cs radioactivity was 12.5 and 3.8 % of ULD at T1 and T2, respectively. The radioactivity at T1 near the north discharge gate can be affected by discharge of cooling water for Units 5 and 6, which is originally drawn from the intake canal in the harbour. The typical seawater around the harbour is expected to have a lower level of radioactivity than at T1. Thus, the underestimation should be significant, but is still less than 10 %. The combined underestimation of the water exchange should be less than about 20 %.

If ^{137}Cs removal other than water exchange (e.g. adsorption by bottom sediment) is significant, then the value of k overestimates the water exchange rate. The ^{137}Cs radioactivity of sediment at ULD from 6 to 19 April 2011 was not available, but TEPCO reported a value of 87 000 Bq kg-wet^{-1} on 29 April and 150 000 Bq kg-wet^{-1} on 12 July 2011 (TEPCO, 2012). From the latter value, the ^{137}Cs inventory in the main harbour sediment could be as high as 8.1 TBq by conservatively assuming a specific wet density of 2 g cm^{-3} and a homogeneous ^{137}Cs distribution within the top 10 cm of the sediment. However, the inventory is less than 0.1 % of the estimated release for the April leak event (0.94 PBq). Thus the sediment adsorption will be insignificant as far as the water exchange during 6–19 April is concerned.

The average difference between the high and low tide at the Fukushima coast is 0.7 m, and the tidal cycle typically repeats twice a day. With an average water depth of 7 m, the tidal water exchange will be about 0.2 d^{-1}. Water exchange will be further facilitated by wind driven currents and waves. The reactors of Units 5 and 6 had been shut down for regular maintenance at the time of the earthquake, but the cooling capability for residual heat of reactors and spent fuel pools were lost in the tsunami. The residual heat removal systems were replaced with temporally installed submersible pumps (ICANPS, 2011), but their rate of seawater intake was not available to the author. At full operation of reactors, the cooling water intake at Units 5 and 6 was designed to be 112 m^3 s^{-1} (Tsuzuki et al., 1992), which would correspond to a water exchange of 5.1 d^{-1}. Seawater intake for residual heat removal should be much smaller, but even 1 % of the full intake corresponds to a water exchange of 0.05 d^{-1}. Thus, overall water exchange of 0.44 d^{-1} is possible.

Table 2. Estimated ^{137}Cs release from plant harbour by water exchange (GBq d^{-1}) and total release (PBq).

Period	Total release during the period (PBq)	Average daily release (GBq d^{-1})
−2 April 2011	?	–
3 April–31 May 2011 (58 days)	2.25[a]	–
1 June–31 August 2011 (92 days)	0.00858	93.2
1 September 2011–31 March 2012 (213 days)	0.00708	33.2[b]
1 April–30 September 2012 (183 days)	0.00148	8.1
Total (546 days)	2.27	–

[a] Release by water exchange was calculated for each day and then summed.
[b] Average ^{137}Cs radioactivity during this period (40.6 Bq L^{-1}) was calculated by linear interpolation between data of 1 ∼ 31 August 2011 and 15 March ∼ 30 April 2012 to avoid overestimate due to exclusion of ND data.

3.3 Estimate of ^{137}Cs release to the sea

The derivation of I with Eq. (3) requires an assumption that the main harbour water is relatively homogeneous with respect to ^{137}Cs, and that ULD represents the entire area of the main harbour. Because the water exchange of 0.44 d^{-1} is fairly rapid, this assumption should be realistic. However, it may not be valid when ^{137}Cs input to the harbour varies much more rapidly than the timescale of the water exchange.

Using the average ^{137}Cs radioactivity for periods when variation of ^{137}Cs was relatively small and assuming a quasi-steady state (Table 1), values of I were obtained from Eq. (3) (Table 2). A value of I corresponds to a daily rate of ^{137}Cs input to the harbour water, and equals the daily release rate of ^{137}Cs to the outer sea by the water exchange. There were several activities by TEPCO that might have changed the water exchange of the plant harbour (ICANPS, 2011). In addition to placement of silt fences mentioned earlier, the damaged part of the jetty between the Unit 1–4 intake canal and the outer sea was filled with 62 “large sandbags”. Filling of the area started on 5 April 2011 and was completed on 8 April. The damaged part was further reinforced by inserting steel plates inside the jetty on 12 July through 28 September 2011. These activities likely reduced the water exchange between the Unit 1–4 canal and the sea, but it is assumed that they had no impact on water exchange in the main harbour area.

For 1 June–31 August 2011, an average daily release of 93.2 GBq day^{-1} was estimated from the data set, giving a total release during the 92 day period of 8.58 TBq. For 1 April–30 September 2012, the average daily release was 8.1 GBq d^{-1} and the total release during this 183 day period was 1.48 TBq. During the 213 day period from 1 September 2011 to 31 March 2012, there were many data points for which ^{137}Cs at ULD was ND (not detected). ^{137}Cs determination by TEPCO apparently reduced the detection limit in March 2012, and little data points were listed as ND after this

period. The average ^{137}Cs radioactivity calculated by excluding ND cases may be an overestimate. Table 2 shows the average ^{137}Cs radioactivity during the 213 day period estimated by linear interpolation between data for 1–31 August 2011 and 15 March–30 April to avoid overestimation. The average daily release during this period would be 33.2 GBq d^{-1} with a total release of 7.08 TBq. These values are not small if we compare them with the release during the May 10–11 leak event (9.8 TBq) or the deliberate release on 4–10 April (0.042 TBq).

Finally, this approach was applied to the 58 day period from 3 April to 31 May 2011, when radionuclide input was varied substantially. As suggested above, the rapid variation of ^{137}Cs radioactivity may have increased uncertainty of the estimate. Even though the error range will be large, the calculation would be helpful to evaluating the validity of the approach of this study. An I value was obtained for each day in the period, and the total release was estimated by summation of each day's I. The estimated total release was 2.25 PBq, which compares well with the value obtained by Tsumune et al. (2012), who estimated that the total ^{137}Cs release to the sea from 26 March to 31 May was 3.5 PBq, as well as with the value of 4 PBq estimated by Kawamura et al. (2011) for the period of 21 March to 30 April. Because the estimate in this study does not include the release before 3 April and does not account for the release that bypasses the main harbour area (for example, a direct release that likely took place from the Unit 1–4 canal to the sea), the approach of the present study appears to have provided a fairly consistent estimate with previous studies. Moreover, the results of this study indicate that the ^{137}Cs release from June 2011 to September 2012 was significant, but that it will not substantially elevate the estimates of total ^{137}Cs release as a result of the accident at the Fukushima Dai-ichi Nuclear Power Plant.

4 Conclusions

The estimated water exchange between the artificial harbour of the Fukushima Dai-ichi Nuclear Power Plant and the outer seawater was 0.44 d^{-1}. Accordingly, the relatively stable ^{137}Cs radioactivity of the harbour water cannot be explained without a continuous input of radioactivity into the harbour. Assuming a steady state, the continuous input to the harbour and hence the continuous release to the outer sea, was calculated to be 93 GBq d^{-1} for June through August 2011 and 8.1 GBq d^{-1} for April through September 2012, respectively.

Supplementary material related to this article is available online at http://www.biogeosciences.net/10/6107/2013/bg-10-6107-2013-supplement..pdf.

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