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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

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

1 Introduction

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

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With the rapid flushing, radioactivity in coastal seawater declined sharply in April and May 2011. The decline was apparently curbed after June. 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). Radioactivity in coastal sediment and marine biota remains persistently even in 2012; a continuous input of radionuclides from the plant and/or from river and groundwater discharge might be one possible explanation for the resiliency (Buesseler, 2012).

In this paper, I examined cesium-137 (^{137}Cs) radioactivity in seawater around the power plant. ^{137}Cs radioactivity in the harbour facility of the power plant was especially informative. From these data, I obtained an exchange rate of harbour water with outer seawater. With the exchange rate and a relatively stable ^{137}Cs radioactivity inside the harbour, I estimated a rate of continuous ^{137}Cs release from the harbour to the sea.

2 Materials and methods

The data I use are obtained and publicly released by Tokyo Electric Power Company (TEPCO). The data are available on websites of TEPCO (TEPCO, 2012) and Ministry of Education, Culture, Sports, Science and Technology (MEXT), Japan (MEXT, 2012). ^{137}Cs radioactivity in seawater was determined by TEPCO under direction by the national government (Government of Japan, 2011). The water sample was directly applied to gamma spectrometers. Figure 1a shows a map of facilities of the Fukushima Dai-ich Nuclear Power Plant and TEPCO's seawater monitoring points.

As shown in Fig. 1a, the power plant has an artificial harbour facility with 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 reactor units; one is in the southern part of the harbour for units 1–4, and another is 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

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jetties and connected to 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 is stopped, cooling water is 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 in the north of the plant.

Possible routes of the radioactive water around the plant were depicted in Fig. 1b. The majority of the radioactive water is presumably originated from the 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 went out to the sea through the harbour passage and through the north discharge gate. The jetty between the unit 1–4 intake canal and the outer sea was known to be 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 discharge either directly into the main harbour or directly into the outer sea is not documented. Since the south discharge gate area facing to the outer sea and the unloading dock area in the main harbour are both relatively close to the damaged reactor units, possibilities of such direct release cannot be ruled out.

3 Results and discussion

Figure 2a shows seawater ^{137}Cs observed at 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) during the period from March 2011 to October 2012. Figure 2b is an expansion for the period from 20 March through 29 May 2011; in Fig. 2b, data obtained at T1 near the north discharge gate (Fig. 1a) is also shown. Seawater samplings were started on 21 March at T2 and on 23 March at T1. Samplings at 2I and

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ULD were started on 2 April and 3 April, respectively. There are several other sampling points in the unit 1–4 intake canal, but samplings at these stations were started in late April or in early May 2011. ULD is the only point that 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 the ones officially confirmed by TEPCO took place from 1 to 6 April, at cooling water intake in front of unit 2 (Government of Japan, 2011). TEPCO estimated that the amount of water released was 520 m^3 and the amount of ^{137}Cs released was 0.94 PBq (Government of Japan, 2011). The increase of radioactivity in March precedent to the April leak event is not related to the confirmed release events. Since radioactivity data in harbour water are not available in March, the route of the radioactivity release is unknown to the author. The ^{137}Cs radioactivity in March is higher at T2 than at T1 (Fig. 2b), thus a release into the sea south of the harbour likely prevailed during this period. The monitoring from 3 April at ULD clearly showed that ^{137}Cs radioactivity increased sharply until 6 April and then started decreasing exponentially (Fig. 2b). This drastic change on 6 April should reflect a sudden decrease of ^{137}Cs input to the main harbour, and the only documented event that could be related is the TEPCO's operation that stopped the leak in front of unit 2 in the early morning of 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 on the next day (7 April) (Fig. 2b). A peak of radioactivity was also identified at T2 on 7 April while the level of radioactivity was smaller than that in March. The order of peak appearance may indicate 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 98 TBq (Government of Japan, 2011). This event is likely 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 are officially confirmed by TEPCO for

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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) during a period 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 at ULD in the main harbour (Fig. 2a). Also, ^{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 for the periods from 1 June through 31 August 2011 and from 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 inside the screen facilities of intake (or in water closer to the reactor housing) is consistently larger than that at the outside of the screen (Table 1). Following the leak event of 1–6 April, TEPCO placed a "silt fence" at the screen facility of each reactor intake and at the curtain wall between the main harbour and the unit 1–4 intake canal on 11–14 April (ICANPS, 2011). The silt fences may have retarded exchange between the canal water and the inner water of the intake, and 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 somewhere around the reactor housings.

Figure 3 shows ^{137}Cs radioactivity in seawater taken at a monitoring point 3I (inside the screen of unit 3 intake) for the period from November 2011 through October 2012. Periodical elevations of ^{137}Cs radioactivity were observed after March 2012. The reason for the appearance of such fluctuation of radioactivity is unknown to the author; this could be due to pavement operation by TEPCO (14 March ~ 11 May 2012) for seabed of the unit 1–4 canal (ICANPS, 2012). Whatever the reason is, both the gradient and the occasional elevations of ^{137}Cs radioactivity indicate that ^{137}Cs is still released to the intake canal.

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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 2011. The sharp decline of ^{137}Cs radioactivity at ULD after this release event can be fitted well with an exponential curve (Fig. 4a). Thus, the decrease is well described by a simple first-order model, or

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

where C is ^{137}Cs radioactivity in harbour water, t is time in unit of day, and k is a rate constant. For the period from 6 to 19 April, a curve fitting yielded a value of 0.44 day^{-1} for k. 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.

A 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 a 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 an average radioactivity per unit 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 and thus the average depth is calculated to be 7.0 m. While detailed depth

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contour of the harbour is not available to the author, the harbour passage has a water-depth of 8 m (Tsuzuki et al., 1992), and the water-depth at the unloading dock is stated to be 6.5 m (Fukushima Prefecture Government, 2013). Thus the estimated volume and average depth seems to be reasonable. Since 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.

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 in proportion 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. I thus presume the underestimate of water exchange should be less than 10 %. When water in the surrounding sea has a significant value of ^{137}Cs radioactivity, the water exchange will be also underestimated. Figure 4b shows ^{137}Cs radioactivity at T1 and T2 plotted against radioactivity at ULD for the period from 6 to 19 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 underestimate could be significant but less than at least about 10 %. The combined underestimate of the water exchange should be less than about 20 %.

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If ^{137}Cs removal other than water exchange, for example, adsorption by bottom sediment, is significant, then the value of k overestimates the water exchange rate. The ^{137}Cs radioactivity of sediment at ULD during the period of 6–19 April 2011 is 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 would 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 a top 10 cm of the sediment. The inventory, however, is less than 1 % of the estimated release in 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 the average water-depth of 7 m, the water exchange by tide will be about 0.2 day^{-1}. Water exchange will be further facilitated by wind driven current and wave. 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 by the tsunami. The residual heat removal systems were managed to be resumed with "temporally installed submersible pumps" (ICANPS, 2011) but the rate of seawater intake is unknown to the author. At the 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); the intake would correspond to a water exchange of 5.1 day^{-1}. Seawater intake for residual heat removal should be much smaller, but even 1 % of the full intake corresponds to water exchange of 0.05 day^{-1}. The overall water exchange of 0.44 day^{-1} is thus fairly possible.

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 represent the whole area of the main harbour. As the water exchange of 0.44 day^{-1} is fairly rapid, this assumption seems realistic. However, it may not be valid when ^{137}Cs input to the harbour varies much more rapidly than the time scale of water exchange.

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With an average ^{137}Cs radioactivity for periods when variation of ^{137}Cs is relatively small and a quasi-steady state could be assumed (Table 1), values of / were obtained from Eq. (3) (Table 2). A value of / corresponds to a daily rate of ^{137}Cs input to the harbour water, and it equals a 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 plant harbour (ICANPS, 2011). In addition to the placement of "silt fences" as 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". The work was started on 5 April 2011 during the April leak event, 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 works likely reduced the water exchange between the unit 1–4 canal and the sea, but I assume the water exchange in the main harbour area was not affected.

For the period from 1 June to 31 August 2011, an average daily release of 93.2 GBq day^{-1} was estimated with the data set. The total release during the 92-day period would be 8.58 TBq. For the period from 1 April ~ 30 September 2012, the average daily release was 8.1 GBq day^{-1} and the total release during the 183-day period was 1.48 TBq. In the period in between these two periods or the 213-day period from 1 September 2011 to 31 March 2012, ^{137}Cs data at ULD contain fairly large cases of ND (not detected). ^{137}Cs determination by TEPCO apparently reduced the detection limit in March 2012, and data later on contain little ND cases. The average ^{137}Cs radioactivity calculated by excluding ND cases would be an overestimate. In Table 2, an average ^{137}Cs radioactivity during the 213-day period was estimated by linear interpolation between data of 1 ~ 31 August 2011 and 15 March ~ 30 April in order to avoid the overestimate. The average daily release in this period would be 33.2 GBq day^{-1} with a total release of 7.08 TBq. These values are significant if we compare them with the release of the May 10–11 leak event (98 TBq) or the deliberate release of 4–10 April (0.042 TBq).

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Finally, this approach was applied to the 58-day period from 3 April to 31 May 2011 when radionuclide input was substantially varied. As suggested above, the rapid variation of ^{137}Cs radioactivity may increase uncertainty of the estimate. Even though the error range will be large, the calculation would be helpful in evaluating 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. This compares well with the value obtained by Tsumune et al. (2012), who estimated total ^{137}Cs release to the sea from 26 March to 31 May was 3.5 PBq, and also with the value of 4 PBq by Kawamura et al. (2011) for the period of 21 March to 30 April. Because my estimate 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), I conclude that the approach of the present study resulted in a fairly consistent estimate with previous studies. I also conclude that ^{137}Cs release from June 2011 until September 2012 is significant, but that it will not substantially elevate the estimates of total ^{137}Cs release by the accident of the Fukushima Dai-ichi Nuclear Power Plant.

Supplementary material related to this article is available online at: http://www.biogeosciences-discuss.net/10/3577/2013/bgd-10-3577-2013-supplement.pdf.

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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) are excluded from calculation.

	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 Jun ~ 31 Aug 2011	114 ± 68	313 ± 232	305 ± 194	340 ± 200	318 ± 195	1030 ± 1320	391 ± 240	1650 ± 1680	404 ± 240	732 ± 398	391 ± 205
1 Apr ~ 30 Sep 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[a]	83.4 ± 57.1	47.1 ± 27.6

[a] Likely overestimated as a significant number of data is reported as ND.

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

Period	Total release during the period (PBq)	Average daily release (GBq day^{-1})
~ 2 April 2011	?	–
3 Apr ~ 31 May 2011 (58 days)	2.25[a]	–
1 Jun ~ 31 Aug 2011 (92 days)	0.00858	93.2
1 Sep 2011 ~ 31 Mar 2012 (213 days)	0.00708	33.2[b]
1 Apr ~ 30 Sep 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 up.
[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 in order to avoid overestimate due to exclusion of ND data.

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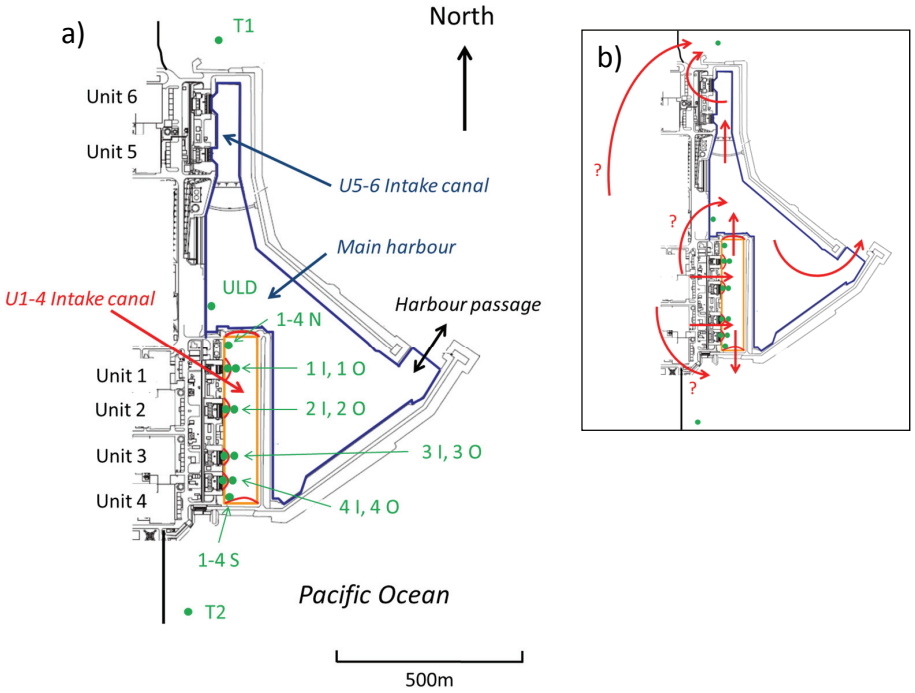

Fig. 1. (a) The artificial harbour facility of the Fukushima Dai-ichi Nuclear Power Plant and TEPCO monitoring points (redrawn from a TEPCO document TEPCO, 2011), **(b)** possible routes of radioactive water transfer. T1: North discharge gate, ULD: unloading dock, 1–4 N: North of unit 1–4 intake, 1 I: 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.

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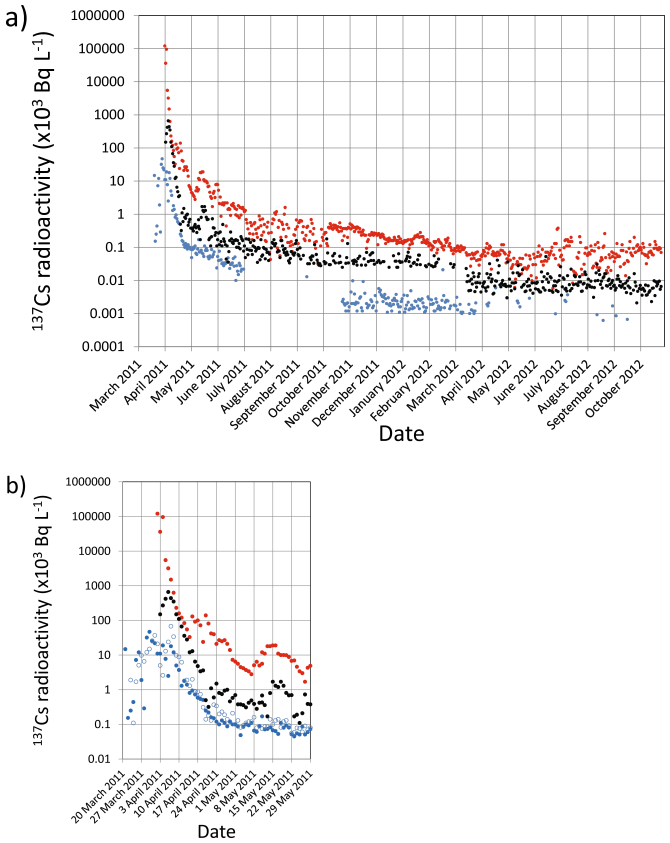

Fig. 2. ^{137}Cs radioactivity in seawater taken at monitoring points T2 (blue dots), ULD (black dots), and 2I (red dots) for the period from March 2011 through October 2012 **(a)**, and an expansion for the period from 20 March through 29 May 2011 together with data obtained at T1 (open blue circles) **(b)**.

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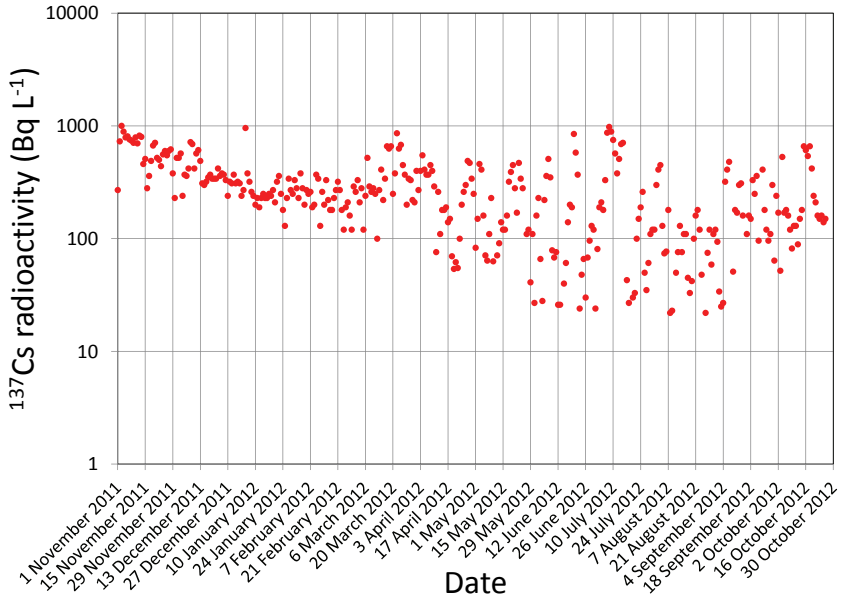

Fig. 3. ^{137}Cs radioactivity in seawater taken at 3I for the period from November 2011 through October 2012.

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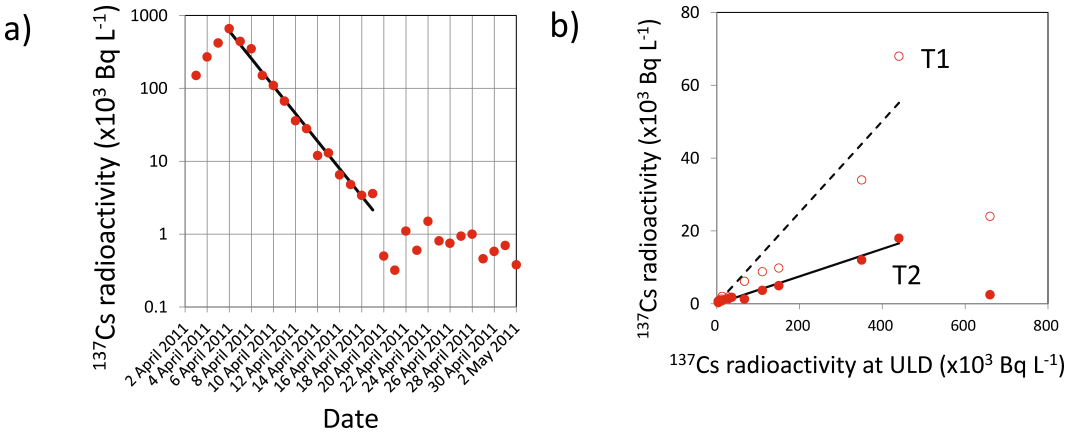

Fig. 4. (a) ^{137}Cs radioactivity at ULD in April 2011. Solid line indicates a least square fitting of an exponential curve for the period from 6 April through 19 April. **(b)** ^{137}Cs radioactivity at T1 (open circles) and at T2 (closed circles) plotted against radioactivity at ULD for the period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

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