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^{90}Sr and ^{89}Sr in seawater off Japan as a consequence of the Fukushima Dai-ichi nuclear accident

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Abstract

The impact of the earthquake and tsunami in the east coast of Japan in 11 March 2011 caused a loss of power at the Fukushima Dai-ichi Nuclear Power Plant (NPP) that resulted in one of the most important releases of artificial radioactivity to the environment. Although several works were devoted to evaluate the atmospheric dispersion of radionuclides, the impact of the discharges to the ocean has been less investigated. Here we evaluate the distribution of Fukushima-derived ^{90}Sr and ^{89}Sr throughout waters 30–600 km offshore in June 2011. Concentrations of ^{90}Sr and ^{89}Sr in both surface waters and shallow profiles ranged from 0.8 ± 0.2 to 85 ± 3 Bq m^{-3} and from 19 ± 6 to 265 ± 74 Bq m^{-3}, respectively. Because of its short half-life, all measured ^{89}Sr was due to the accident, while the ^{90}Sr concentrations can be compared to the background levels in the Pacific Ocean of about 1.2 Bq m^{-3}. Fukushima-derived radiostrontium was mainly detected north of Kuroshio Current, as this was acting as a southern boundary for transport. The highest activities were associated with near-shore eddies, and larger inventories were found in the closest stations to Fukushima NPP. The data evidences a major influence of direct liquid discharges of radiostrontium compared to the atmospheric deposition. Existing ^{137}Cs data reported from the same samples allowed us establishing a ^{90}Sr/^{137}Cs ratio of 0.0256 ± 0.0006 in seawater off Fukushima, being significantly different than that of the global atmospheric fallout (i.e. 0.63) and may be used in future studies to track waters coming from the east coast of Japan. Liquid discharges of ^{90}Sr to the ocean were estimated, resulting in an inventory of 53 ± 1 TBq of ^{90}Sr in the inshore study area in June 2011 and total releases of ^{90}Sr ranging from 90 to 900 TBq, depending upon the reported estimates of ^{137}Cs releases that are considered.

BGD
10, 2039–2067, 2013

1 Introduction

Radiostrontium, and in particular ^{90}Sr ($T_{1/2}$ = 28.9 a), is an artificial radionuclide that has been liberated to the environment in the last decades mainly due to the atmospheric nuclear weapon testing performed in the 50's and 60's and the direct releases to the sea from nuclear reprocessing facilities. Given that oceans cover more than 70 % or the Earth surface, much of these releases of ^{90}Sr now reside dissolved in seawater (IAEA, 2005). Concentrations of ^{90}Sr in oceans are summarized in the World Marine Radioactivity Studies (WOMARS), and a global ^{137}Cs/^{90}Sr ratio of 1.5 has been established. As its chemical behavior is similar to that of calcium, strontium is deposited in biological systems, especially in those tissues with high calcium content such as bone. The highly energetic 0.546 MeV beta particles, and the additional exposure to the 2.2 MeV beta particles released during decay of its short-lived ^{90}Y daughter isotope, makes ^{90}Sr a radionuclide of health concern (ICRP, 1993). However, not much literature exists on the presence of ^{89}Sr ($T_{1/2}$ = 50.52 d) in the oceans. This radionuclide is also a primary fission product, but with a much shorter half-life than ^{90}Sr.

As a result of the earthquake on 11 March 2011, and subsequent tsunami, radionuclide contaminants were accidently released to the environment from the Fukushima Dai-ichi nuclear power plant (NPP). The emissions of artificial radionuclides both to the atmosphere (Chino et al., 2011) and the marine environment (i.e. Povinec et al., 2012; Buesseler et al., 2011) have been broadly reported. In particular, ^{134}Cs and ^{137}Cs concentrations at the nuclear power plant ocean discharge channels peaked in early April at levels that reached several millions times higher compared with the preexisting levels in this area (Buesseler et al., 2011), thus representing the most important accidental artificial radioactive release into the sea (Bailly du Bois et al., 2011). Simulations of regional ocean models using ^{137}Cs concentrations in the ocean off Fukushima allowed estimating the total amount of ^{137}Cs directly discharged to the ocean, ranging from 1 to more than 20 PBq (Kawamura et al., 2011; Tsumune et al., 2012; Bailly du Bois et al., 2011).

BGD
10, 2039–2067, 2013

Although temporal changes of ^{131}I, ^{134}Cs and ^{137}Cs concentrations in surface water adjacent to Fukushima have been well documented, releases of radiostrontium to the sea are poorly document (Povinec et al., 2012). The main reason for this lack of data resides on the difficulties of pre-concentrating and analyzing ^{90}Sr in seawater. The recent study by Povinec et al. (2012) has been the first report assessing the impact of radiostrontium released from the Fukushima Dai-ichi nuclear power plant to the marine environment. Their data on ^{90}Sr in coastal seawater (near the discharging channel in Fukushima Dai-ichi NPP) were obtained from the Ministry of Education, Culture, Sports, Science and Technology (MEXT). Concentrations of ^{90}Sr reached up to four orders of magnitude greater than the pre-Fukushima levels (1 Bq m^{-3}). Between March and November 2011, maxima activities of ^{90}Sr and ^{89}Sr were of 69 and 9.3 kBq m^{-3}, respectively, but even higher values were reported after a December 2011 accidental discharge (400 and 140 kBq m^{-3} for ^{90}Sr and ^{89}Sr, respectively) (Povinec et al., 2012).

This study is part of an international project, which aimed at understanding the amount, type and fate of radioactive materials released to the North West Pacific Ocean and see whether the Kuroshio Current was acting as southern boundary for radionuclide transportation in the marine environment. This is the first survey of ^{90}Sr and ^{89}Sr offshore Japan after the Fukushima NPP accident in March 2011. In particular, here we evaluate the distributions of ^{90}Sr and ^{89}Sr released from Fukushima in the water column in an area encompassing 30–600 km off Japan in May–June 2011.

2 Materials and methods

2.1 Seawater sampling

A total of 57 20 L seawater samples, both from surface and vertical profiles, were collected from the R/V *Ka'imikai-o-Kanaloa* (KOK) between 4 and 18 June, 2011 (Fig. 1). Sample codes, collection date, locations, depths, salinity and temperature of each sample are provided in Table 1.

BGD
10, 2039–2067, 2013

Surface waters were collected using a deck mounted pump and hose which was placed about 2 m below the sea surface. A pre-filter cartridge was used to remove the suspended matter. Deeper samples (10–200 m) were collected with Niskin bottles mounted on a CTD/Rosette at stations #3, #19, #21, #23, #25, #27 and #29 (see Table 1). Samples were spiked with 200 mg of Sr^{2+} and stored for its subsequent analysis at the Universitat Autònoma de Barcelona and Universidad de Sevilla.

2.2 ^{90}Sr purification and quantification through ^{90}Y counting by proportional counter

Once in the laboratory, samples were weighted and acidified with concentrated HCl. The purification method for the analysis of ^{90}Sr was based on the protocol described in Waples and Orlandini (2010), and adapted to seawater samples by Casacuberta et al. (2013). The method consists on the measurement of the daughter of ^{90}Sr, ^{90}Y ($T_{1/2}$ = 64 h), which is in equilibrium with ^{90}Sr, by beta counting. Samples were spiked with 10 mg of stable Y, and 30 mg of Fe^{3+} as ferrous sulfate was added to the samples. After letting samples equilibrate overnight, pH was raised up to 8 to coprecipitate yttrium with the iron hydroxides. The precipitate was then collected onto a nitrocellulose filter (0.22 µm, 14 cm, Whatman) and subsequently dissolved in 20 mL HNO_3 8 M. Isolation of yttrium from other beta emitters (mainly Th) was performed by passing the solution through an anion ion-exchange column (Bio-Rad AG1-X8, 100–200 mesh). The eluate, 40 mL of HNO_3 8 M, was then evaporated to reconstitute the sample in 20 mL 1 M HCl. A further separation on a cation exchange column (Bio-Rad AG50W-X8, 100–200 mesh) was required for the final purification of yttrium from lead isotopes. Sample was transferred to the column and 80 mL of 1 M HCl were added to wash the resin. Finally, yttrium was eluted by adding 30 mL 6 M HCl recovered by precipitation using ferric hydroxides and filtration on to a nitrocellulose filter (0.22 µm, 47 mm, Whatman), which was dried and placed onto a beta-holder for the measurement of the ^{90}Y beta emissions. Samples were measured on a A 5-position beta counter manufactured

BGD
10, 2039–2067, 2013

by RISØ National Laboratories (Roskilde, Denmark) during 60 min intervals over approximately 60–64 h. Filters were re-measured after 1 week and 3 weeks to verify that detected counts corresponded to the decay of ^{90}Y and no other beta emitters, including ^{91}Y ($T_{1/2}$ = 58.5 d), interfered. The chemical recovery of yttrium, which ranged from 10 to 85 %, was assessed by determining the stable Y concentrations by ICP-MS on aliquots of the digested filters.

2.3 ^{89}Sr purification and quantification by proportional counter

The supernatants from the first separation of iron hydroxides described in Sect. 2.2 were stored and sent to Universidad de Sevilla to proceed with an alternative direct determination of ^{90}Sr in the same samples. The initial objective was to perform these measurements as a quality control exercise to validate the results obtained by the method described previously in Sect. 2.2. However, the method used for the validation of ^{90}Sr allowed for the determination of both, ^{89}Sr and ^{90}Sr (the first one only in some samples due to the long time delay between sampling and measuring).

The conventional radiochemical procedure (Harvey et al., 1989) applied for isolation and purification of the ^{89}Sr and ^{90}Sr in the supplied supernatants was based in an initial oxalate precipitation, followed by a transformation of the oxalate precipitate into carbonate by calcination to 550 °C. Subsequently, calcium was mostly eliminated by fuming acid precipitation, followed by the dissolution of the precipitate in water, its evaporation to dryness and its posterior re-dissolution in nitric acid.

Barium was afterwards separated from strontium by formation of barium chromate precipitation, and yttrium was removed by two successive hydroxide precipitations. Strontium was finally purified by its precipitation with a saturated solution of sodium carbonate, subsequent conversion of the precipitate to strontium nitrate by adding concentrated HNO_3, the dissolution of the precipitate with hot water, and the final precipitation of the Sr as $SrCO_3$, by adding again a saturated solution of sodium carbonate. The final $SrCO_3$ precipitate was homogeneously distributed over the counting planchet and dried until constant weight in a desiccator.

BGD
10, 2039–2067, 2013

The radiochemical yields were determined gravimetrically and double-checked by ICP-MS in a selected number of samples. The presence of natural Sr in samples was considered after its determination in aliquots by ICP-MS.

The quantification of ^{89}Sr and ^{90}Sr was performed by using a 10-channel low level proportional counter LB790 (Berthold Technologies). Two measurements at different time intervals were performed to determine the concentrations of ^{89}Sr and ^{90}Sr in the sample under study at the time of sampling. The first one was performed 36–72 h just after the ^{90}Y separation and the second one at least 18 days after the first measurement. In all cases, each measurement was performed during at least two cycles of 600 min. From the net counting rates obtained in the two measurements, the activities of ^{89}Sr and ^{90}Sr were determined, following the methodology described in Herranz et al. (2011), which implies the previous calibration of the counter for determination of the counting efficiencies of ^{90}Sr, ^{90}Y and ^{89}Sr.

3 Results

Concentrations of ^{90}Sr and ^{89}Sr in seawater samples collected at the different stations and depths ranged from 0.8 ± 0.2 to $85 \pm 3\,\mathrm{Bq\,m^{-3}}$ and from 19 ± 6 to $265 \pm 74\,\mathrm{Bq\,m^{-3}}$, respectively (see Table 1).

3.1 ^{90}Sr in surface seawaters

Activities of ^{90}Sr in surface seawater samples ranged from 1.1 ± 0.2 to $85 \pm 3\,\mathrm{Bq\,m^{-3}}$. The highest concentrations were found along the northern coast of Japan, being particularly enhanced about 100–200 km SE from Fukushima, at stations 29 and 30 (Fig. 2a). The concentration of ^{90}Sr in surface samples from near-shore stations ranged from 10 to $85\,\mathrm{Bq\,m^{-3}}$. The lowest activities were located in the southeastern areas of the studied area (i.e. stations 1, 2, 3 and 10) and northern from Fukushima NPP (stations 20 and 24). These concentrations were close to the background value of $1.2\,\mathrm{Bq\,m^{-3}}$

BGD
10, 2039–2067, 2013

reported for this area before the Fukushima NPP accident (IAEA, 2005). The distribution pattern of ^{90}Sr concurs with the surface ^{137}Cs and ^{134}Cs dispersal reported in Buesseler et al. (2012), from the same cruise (Fig. 2b, c).

3.2 ^{90}Sr in shallow profiles

Specific activities of ^{90}Sr in shallow profiles (down to 200 m) taken at stations 19, 21, 23, 25, 27 and 29 are detailed in Table 1 and summarized in Fig. 3. In all cases, the majority of ^{90}Sr was found in the first 50 m depth horizon. In general, ^{90}Sr concentrations measured at the deepest waters of each profile correspond to the expected for background levels (1.2 Bq m^{-3}) of ^{90}Sr in this area. The only exception was station 23, where concentrations at 100 meters depth were still enhanced (i.e. 4.8 ± 0.7 Bq m^{-3}). In general, specific activities of ^{90}Sr decrease with depth, yet some differences are noticed in the profile trends: whereas concentrations of ^{90}Sr in stations 19, 21, 27 and 29 decrease exponentially with depth, the two closest stations of Fukushima Dai-ichi NPP (23 and 25) showed high ^{90}Sr concentrations between 20 and 50 m depth.

The particular behavior of ^{90}Sr at stations 23 and 25 corresponds to an area affected by near shore eddies. This is observed in the section represented in Fig. 4a, which comprises stations 25 (left), 23, 21, 19 and 11 (right). ^{90}Sr concentrations in the station 23 profile are significantly higher (from 5 to 37 Bq m^{-3}) than the concentrations in the two profiles closer to shore (stations 25 and 21), due to the presence of this nearshore eddy (see Fig. 4b, c). Distribution of ^{90}Sr according to water masses is further discussed in Sect. 4.2.

Inventories of ^{90}Sr in the upper 100 m were calculated for the six profiles closer to the coast (Fig. 5). These inventories were in all instances higher than the expected inventory of ^{90}Sr for this area prior to the Fukushima accident, calculated assuming a background ^{90}Sr concentration of 1.2 Bq m^{-3} in the water column (i.e. 120 Bq m^{-2}). Station 23 holds the greatest inventory of ^{90}Sr (2511 ± 102 Bq m^{-2}), followed by stations 27 and 29, these last ones corresponding to the stations that presented the highest ^{90}Sr concentrations at surface.

BGD
10, 2039–2067, 2013

3.3 ^{89}Sr results in surface and shallow profiles

^{89}Sr was quantified in 19 samples (Table 1), and concentrations ranged from 19 ± 6 to $265 \pm 74\,\mathrm{Bq\,m^{-3}}$ in surface waters, reaching a maximum subsurface value of $137 \pm 43\,\mathrm{Bq\,m^{-3}}$ at 50 m depth at station 23. Due to the short half-life of ^{89}Sr (50.53 d) and the long time delay between sampling and counting, (up to 5–6 months) uncertainties were larger than 40 % in some of the measured samples.

4 Discussion

4.1 Impact of the Fukushima Dai-ichi nuclear accident in seawaters off Japan

Contrary to other radionuclides such as ^{137}Cs, ^{134}Cs and ^{131}I (Hisamatsu, 2012), the amounts of radiostrontium in the form of liquid radioactive wastes released to the sea after the Fukushima Dai-ichi nuclear accident are less known, mostly due to the more complex and time-consuming analytical procedures used for its determination. However, as the cooling water directly interacted with ruptured nuclear fuel rods, large amounts of radiostrontium may have been released directly to the ocean as well (Povinec et al., 2012). Indeed, our results of ^{90}Sr and ^{89}Sr in both, surface seawater and depth profiles, showed a clear impact of the Fukushima nuclear accident in the waters off Japan. Before April 2011, the main ^{90}Sr source in the Japanese waters was due to global fallout from atmospheric weapons testing, including the influence of 11 Chinese atmospheric nuclear weapons tests in the 1970s (Ikeuchi, 2003). Just after these events, the average ^{90}Sr concentrations in the eastern coast off Japan were between 4.3 and $5.7\,\mathrm{Bq\,m^{-3}}$. Average concentrations measured during the 1980s and 1990s ranged between 2.5 and $2.8\,\mathrm{Bq\,m^{-3}}$, and the last ^{90}Sr concentrations reported ranged from 1.4 to $1.8\,\mathrm{Bq\,m^{-3}}$ in surface waters (Ikeuchi et al., 1999; Povinec et al., 2000) and between 0.21 and $0.58\,\mathrm{Bq\,m^{-3}}$ in deep waters (> 2000 m) (Ikeuchi et al., 1999). Decay corrected concentrations of ^{90}Sr in the West-Pacific Ocean in 2011

BGD
10, 2039–2067, 2013

would therefore have decreased to a range of 1.1 to 1.4 Bq m^{-3} (IAEA, 2005; Povinec et al., 2012), agreeing with the lowest value found in this study for surface waters (i.e. 1.1 ± 0.2 Bq m^{-3}). The Fukushima Dai-ichi nuclear accident produced an increase of the ^{90}Sr concentrations in the waters off Japan, such that three months after the accident values were 1 to 2 orders of magnitude higher than the background in some areas. Fukushima-derived radiostrontium was present not only in surface waters but at depth, reaching maximum values of 37 ± 3 Bq m^{-3} at 20 m.

The presence of ^{90}Sr in the waters off Japan derived from the Fukushima nuclear accident was accompanied by the presence of ^{89}Sr. Given its short half-life (50.57 d), the occurrence of this radionuclide is a faultless signal of the Fukushima-derived release. Moreover, the ^{89}Sr/^{90}Sr ratio pointed to be Fukushima-derived radiostrontium, as the decay corrected ratio measured in this study (see Sect. 4.3) is in excellent agreement with the ratio reported for the reactor core inventory on 11 March 2011 (Nishikara et al., 2012).

4.2 Distribution of ^{90}Sr in the coast off Japan

Three assumptions were made for discussing the ^{90}Sr distribution in the waters off Japan in May–June 2011: (i) the Sr peak in direct oceanic release to the Pacific Ocean took place in 6 April, as it did for Cs isotopes (Buesseler et al., 2011); (ii) both Cs and Sr behave geochemically similarly (Fig. 2) in the waters off Japan, as both are dissolved in seawater and have a weak interaction to sediment (IAEA, 2005; Povinec et al., 2003); and (iii) most of the ^{90}Sr left in the KOK sampling area at the time of the cruise came from the direct oceanic discharges, as it was the case for Cs (Rypina et al., 2013).

After the radionuclide peak release, ^{90}Sr in seawaters would be dispersed and distributed through the Japanese coast according to advection processes occurring in this specific area of the Pacific Ocean. In particular, this is a region where three main currents converge: the Oyashio, the Kuroshio and the Tsugaru currents (Talley et al., 1995). The Oyashio front is defined to be the southern limit of subpolar waters with very

BGD
10, 2039–2067, 2013

low salinity, which are driven from the North to the South coast of Japan (Fig. 6). On the contrary, the Kuroshio Current originates in the subtropical gyre, consisting in low density, warm, high salinity surface waters, coming from the Southwest of the Japanese coast (Fig. 6). Finally, the Tsugaru current (not present in May–June) is a source of warm and saline water that enters the Sea of Japan through the Tsushima and Tsugaru straits (Talley et al., 1995).

Using data from surface drifters released during the KOK cruise, Rypina et al. (2013) performed a numerical model to investigate the dominant mechanisms governing the short-term spread of radionuclides in this area. From their model, and similar to ^{137}Cs results, it can be stated that: (i) the absence of the ^{90}Sr at the southernmost stations (stations 1, 2, 3, 14 and 28) was attributed to the Kuroshio Current, which prevents for a southward progression of Fukushima contaminants such as ^{90}Sr; (ii) the largest concentration of ^{90}Sr were shown to be associated with the semi-permanent nearshore eddy and stirring them for a long time around the eddy perimeter (stations 29, 30 and 31); and (iii) the intermediate ^{90}Sr concentrations at the westernmost row of stations (stations 25 and 27) were explained by the coolant water that continued to leak from the reactor after the peak release.

In addition to the dispersion of radiostrontium in surface waters off Japan, ^{90}Sr may also have penetrated into deep waters, either by diffusion or advection processes, as shown by our measurements of Fukushima-derived radiostrontium down to 100–200 m depth.

BGD
10, 2039–2067, 2013

4.3 $^{89}Sr/^{90}Sr$ ratio

By using the $^{89}Sr/^{90}Sr$ net ratio (subtracting ^{90}Sr background concentrations), the time delay between sampling and accident was calculated by using the following Eq (1):

$$t = \frac{\mathrm{Ln}\left[\frac{\left(\frac{^{89}Sr}{^{90}Sr}\right)_{obs}}{\left(\frac{^{89}Sr}{^{90}Sr}\right)_{reactor}}\right]}{-\lambda_{^{89}Sr}} \quad (1)$$

where $(^{89}Sr/^{90}Sr)_{obs}$ is the net ratio measured at each sample at sampling time, $(^{89}Sr/^{90}Sr)_{reactor}$ is the ratio at reactor (i.e. 11.8 ± 0.4 according to Povinec et al., 2012) and $\lambda_{^{89}Sr}$ is the decay constant of ^{89}Sr ($0.0137\,d^{-1}$). Time delay between the accident and the sampling calculated for each sample is reported in Table 2. Resulting values range from 74 to 135 d, which in average is 98 ± 18 d in good agreement with the elapsed time since the accident.

4.4 Estimate of ^{90}Sr discharge to the sea

Several works have been devoted to quantify the amount of ^{137}Cs released to the sea due to the Fukushima Dai-ichi nuclear accident, ranging from 3.5 to 34 PBq (Tsumune et al., 2012; Buesseler et al., 2012; Bailly-du-Bois et al., 2011; Charette et al., 2013). Here we calculated the release of ^{90}Sr by using the $^{90}Sr/^{137}Cs$ ratio data obtained from the analyzed samples (Buesseler et al., 2012), once corrected for background concentrations (1.2 and 2.5 $Bq\,m^{-3}$ for ^{90}Sr and ^{137}Cs, respectively) (IAEA, 2005) (Fig. 7). The $^{90}Sr/^{137}Cs$ ratio for the samples collected during the KOK cruise was 0.0265 ± 0.0006 which is higher than the ratio of 0.01 reported by Povinec et al. (2012). Their value was the average of the data obtained in April 2011 at site 1, close to one of the discharging channels. Indeed if calculating the average value at their sites 1 and 2 (close to discharging channels), 7 and 8 (15 km off the Fukushima NPP) between April and

BGD
10, 2039–2067, 2013

June, the resulting ratio $^{90}Sr/^{137}Cs$ is 0.0282, in very good agreement with our data. Therefore, ratio $^{90}Sr/^{137}Cs$ calculated here (0.0265 ± 0.0006) is representative of the average ratios found in Povinec et al. (2012) once surface waters off the coast of Japan were homogenized.

The total amount of ^{90}Sr in the study area in June 2011, taking the inventory of 2 PBq of ^{137}Cs estimated by Buesseler et al. (2012) and using the $^{90}Sr/^{137}Cs$ ratio of 0.0265 ± 0.0006 would then be of 53 ± 1 TBq. This figure is constrained within the inshore box, but the actual total releases of ^{90}Sr would have been greater. Given the range of published estimates of ^{137}Cs releases due to direct discharge from the NPP, the total amount of ^{90}Sr released could range from 93 ± 3 and 901 ± 20 TBq (Table 3).

It is worth noticing that in December 2011, the Tokyo Electric Power Company reported an accidental leakage from the evaporative condensation apparatus at Fukushima Dai-ichi NPP of approximately 150 L of water containing 11 GBq of ^{89}Sr and 15 GBq of ^{90}Sr. In fact, activities of ^{90}Sr measured at the discharge site in December 2011 exceeded ^{137}Cs concentrations. Thus the $^{90}Sr/^{137}Cs$ ratio that we measured cannot be used to predict the total amounts of ^{90}Sr released through direct ocean discharge after December 2011.

The amount of ^{90}Sr released because of the Fukushima Dai-ichi NPP accident can be compared with the most important nuclear accidents and the discharges of nuclear reprocessing fuel plants. The releases of ^{90}Sr from Chernobyl were quantified in 10 PBq (IAEA, 2003), of which 277 TBq were discharged to the Black Sea through the Pripyat and Dnieper Rivers (Vakulovsky et al., 1994). However, the direct inputs to the ocean were considered to be negligible as the radioactivity release was mainly of atmospheric origin (IAEA, 2005). On the contrary, ^{90}Sr releases to the Pacific Ocean due to the nuclear accident in Fukushima are comparable with discharges from the nuclear reprocessing plants in Sellafield and Cap de la Hague. Indeed, the total discharges of ^{90}Sr to the oceans (decay corrected to year 2000) due to the releases comprised between 1970 and 1998 were of 3195 and 744 TBq for Sellafield and La Hague, respectively (IAEA, 2005). Thus, the inventory of ^{90}Sr Fukushima-derived in the oceans

BGD
10, 2039–2067, 2013

represent between 3 and 23 % of the total amount due to direct discharges. It is important to note here that the Fukushima discharges, although comparable to the ones from Sellafield, have occurred over a much shorter time scale. Compared to the ocean inventory of ^{90}Sr due to global fallout, of about 116 PBq estimated in 2011 (IAEA, 2005), the Fukushima-derived ^{90}Sr directly released to the sea would represent less than 1 % of ^{90}Sr of the total inventory.

4.5 Direct releases of ^{90}Sr to the sea versus atmospheric deposition

The amount of radiostrontium measured in the study area during the KOK cruise could be a result of two main sources: (i) the localized direct discharge of contaminated coolant waters into the near-shore ocean, and (ii) the fallout of airborne released to the atmosphere during the Fukushima NPP explosions, with subsequent deposition onto the ocean.

Strontium is much less volatile than other fission products such as cesium and iodine isotopes, and thus smaller amounts of ^{90}Sr compared to ^{137}Cs were expected to be released to the atmosphere. Indeed, TEPCO published a ^{90}Sr/^{137}Cs ratio in soils of 0.001, which is far below the average ratio measured in seawater in this study (i.e. 0.0265). If the ^{90}Sr/^{137}Cs ratio of liquid discharges in April–May 2011 was 0.0265 and the ratio from atmospheric release was 0.001, it is clear that the ocean samples had a much stronger influence of the direct discharges of cooling water into the sea and the oceanic background concentration reported for the Pacific Ocean before Fukushima NPP accident (Fig. 8).

5 Conclusions

This study is the first survey of ^{90}Sr and ^{89}Sr offshore Japan after the Fukushima NPP accident in March 2011. Concentrations of ^{90}Sr and ^{89}Sr detected at 32 stations, including both surface and shallow profiles, ranged from 0.8 ± 0.2 to 85 ± 3 Bq m^{-3} and

BGD
10, 2039–2067, 2013

from 19 ± 6 to $265 \pm 74\,\mathrm{Bq\,m^{-3}}$, respectively. While the occurrence of ^{89}Sr is an evident signal of Fukushima-derived releases, the activities of ^{90}Sr measured in some stations reached values two orders of magnitude higher than the background levels reported in this area (i.e. $1.2\,\mathrm{Bq\,m^{-3}}$). The ^{90}Sr/^{137}Cs ratio has been calculated to be 0.0265 ± 0.0006 (in May–June 2011) although it may have varied, especially after a significant leakage of contaminated waters that occurred in December 2011. This ratio is unique and significantly different than that of the global atmospheric fallout produced by nuclear weapon tests, which was 0.63 and it may be used in future studies to track waters coming from Fukushima. Results of the samples analyzed here evidenced a much stronger influence of direct discharges of cooling water into the sea and the oceanic background concentration, rather than atmospheric deposition. Direct discharges have been quantified on the basis of the estimates of ^{137}Cs discharges and the ^{90}Sr/^{137}Cs ratio, resulting in a range between 90 and 900 TBq of ^{90}Sr.

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BGD
10, 2039–2067, 2013

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BGD
10, 2039–2067, 2013

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BGD
10, 2039–2067, 2013

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BGD
10, 2039–2067, 2013

Table 1. Concentrations of ^{90}Sr and ^{89}Sr in seawater samples collected off Fukushima in June 2011. Values were decay corrected to sampling date. Information on collection date location, depth, salinity and temperature is also provided. Uncertainties coverage is $k = 1$.

Station #	Sample code	Collection date	Depth (m)	Longitude	Latitude	Salinity (psu)	Temperature (°C)	^{90}Sr (Bq m^{-3})	^{89}Sr (Bq m^{-3})
1	Sr-1	6 Jun 2011	2	146.5924	33.5967	34.72	20.46	1.3 ± 0.2	
2	Sr-2	7 Jun 2011	2	147.0002	34.2989	34.69	28.81	1.4 ± 0.9	
3	Sr-3	7 Jun 2011	200	147.0004	34.9980	34.65	16.23	1.5 ± 0.6	
	Sr-4	7 Jun 2011	100	147.0004	34.9980	34.76	18.37	2.3 ± 1.4	
	Sr-5	7 Jun 2011	50	147.0004	34.9980	34.72	19.12	4.8 ± 5.7	
	Sr-6	7 Jun 2011	20	147.0004	34.9980	34.72	20.66	1.5 ± 0.2	
4	Sr-7	8 Jun 2011	2	147.0026	35.2986	34.33	17.29	10.1 ± 0.4	24 ± 4
5	Sr-8	8 Jun 2011	2	147.0227	36.0090	34.71	18.23	2.7 ± 1.2	
6	Sr-9	8 Jun 2011	2	147.0068	36.2998	34.57	15.95	3.4 ± 0.2	
7	Sr-10	9 Jun 2011	2	147.0000	37.5947	34.18	15.53	6.3 ± 0.3	
8	Sr-11	9 Jun 2011	2	146.5999	37.3010	34.34	15.21	5.6 ± 0.4	
9	Sr-12	9 Jun 2011	2	147.0124	37.5901	33.59	10.85	3.2 ± 0.3	
10	Sr-13	10 Jun 2011	2	143.5978	37.5986	33.92	12.27	9.3 ± 2.5	27 ± 5
11	Sr-14	10 Jun 2011	2	144.0018	37.3066	34.24	13.77	7.8 ± 2.0	26 ± 6
12	Sr-15	10 Jun 2011	2	143.5953	36.5623	33.86	11.71	3.5 ± 0.3	
14	Sr-16	11 Jun 2011	2	144.0033	35.5976	34.73	21.15	1.1 ± 0.2	
17	Sr-17	12 Jun 2011	2	143.0061	36.3021	34.53	20.96	1.1 ± 0.2	
18	Sr-18	12 Jun 2011	2	142.5998	36.5994	34.27	15.62	6.1 ± 0.3	
19	Sr-19	12 Jun 2011	2	142.5972	37.3029	34.20	14.96	8.8 ± 0.4	27 ± 6
	Sr-20	12 Jun 2011	200	142.9980	37.4940	33.81	5.90	0.8 ± 0.2	
	Sr-21	12 Jun 2011	100	142.9980	37.4940	34.15	9.42	1.7 ± 0.2	
	Sr-22	12 Jun 2011	50	142.9980	37.4940	33.99	10.03	4.7 ± 0.4	
	Sr-23	12 Jun 2011	20	142.9980	37.4940	34.19	14.85	8.1 ± 0.4	25 ± 4
20	Sr-24	13 Jun 2011	2	143.0016	37.6000	34.53	14.25	1.7 ± 0.2	
21	Sr-25	13 Jun 2011	2	142.2947	37.3008	34.18	14.88	7.4 ± 0.4	22 ± 9
	Sr-26	13 Jun 2011	200	142.5020	37.5010	33.71	4.66	1.0 ± 0.3	
	Sr-27	13 Jun 2011	100	142.5020	37.5010	33.69	6.46	1.2 ± 0.3	
	Sr-28	13 Jun 2011	50	142.5020	37.5010	33.98	9.39	2.6 ± 0.2	
	Sr-29	13 Jun 2011	20	142.5020	37.5010	34.19	14.87	7.3 ± 0.4	
22	Sr-30	13 Jun 2011	2	142.3005	38.0001	34.34	18.31	6.9 ± 0.4	19 ± 6
23	Sr-31	14 Jun 2011	2	141.6000	37.2974	34.79	17.14	26 ± 2	57 ± 14
	Sr-32	14 Jun 2011	100	142.0050	37.4810	33.73	7.32	4.8 ± 0.7	
	Sr-33	14 Jun 2011	50	142.0050	37.4810	34.32	12.83	34 ± 2	137 ± 43
	Sr-34	14 Jun 2011	20	142.0050	37.4810	34.67	16.06	37 ± 3	132 ± 34
	Sr-35	14 Jun 2011	10	142.0050	37.4810	34.79	17.17	28 ± 1	65 ± 21
24	Sr-36	14 Jun 2011	2	141.5984	38.0004	34.48	19.21	2.5 ± 0.2	
25	Sr-37	15 Jun 2011	2	141.2673	37.3187	33.98	15.08	17 ± 1	29 ± 13
	Sr-38	15 Jun 2011	90	141.4410	37.5170	33.89	8.98	3.6 ± 0.9	
	Sr-39	15 Jun 2011	50	141.4410	37.5170	33.89	9.30	12 ± 4	43 ± 16
	Sr-40	15 Jun 2011	20	141.4410	37.5170	33.88	9.60	12 ± 2	
	Sr-41	15 Jun 2011	10	141.4410	37.5170	34.20	14.11	11 ± 1	
	Sr-42	15 Jun 2011	120	141.4410	37.5170	33.89	8.98	1.9 ± 0.2	
	Sr-43	15 Jun 2011	100	141.4410	37.5170	33.90	9.74	3.3 ± 0.3	
27	Sr-45	15 Jun 2011	100	141.4000	36.5010	34.20	10.13	2.2 ± 0.4	
	Sr-46	15 Jun 2011	50	141.4000	36.5010	34.13	11.59	16 ± 6	
	Sr-47	15 Jun 2011	20	141.4000	36.5010	34.14	13.74	12 ± 1	35 ± 14
	Sr-48	15 Jun 2011	10	141.4000	36.5010	34.11	14.80	18 ± 1	45 ± 18
	Sr-50	15 Jun 2011	2	141.2390	36.2981	34.11	14.78	33 ± 8	67 ± 27
28	Sr-49	16 Jun 2011	2	141.2391	36.0033	34.38	20.63	1.1 ± 0.2	
29	Sr-51	16 Jun 2011	2	142.0058	36.2986	34.03	17.06	85 ± 3	265 ± 74
	Sr-52	16 Jun 2011	100	142.0030	36.4990	34.20	10.37	1.2 ± 0.2	
	Sr-53	16 Jun 2011	50	142.0030	36.4990	34.38	12.50	4.4 ± 0.3	
	Sr-54	16 Jun 2011	20	142.0030	36.4990	34.28	14.52	12 ± 1	32 ± 7
	Sr-55	16 Jun 2011	10	142.0030	36.4990	34.03	17.06	15 ± 1	
30	Sr-56	17 Jun 2011	2	142.2972	36.2975	33.94	17.49	28 ± 3	
31	Sr-57	17 Jun 2011	2	142.3001	36.5992	33.91	16.55	15 ± 2	
32	Sr-58	17 Jun 2011	2	142.0030	36.5989	34.19	17.66	11 ± 1	41 ± 14

BGD
10, 2039–2067, 2013

Table 2. $^{89}Sr/^{90}Sr$ ratio in seawater samples and resulting ages obtained from Eq. (1).

Station #	Depth (m)	Sample code	$^{89}Sr/^{90}Sr$	Water age (days)
4	2	Sr-7	2.7 ± 0.4	107 ± 17
10	2	Sr-13	3.2 ± 1.1	95 ± 31
11	2	Sr-14	3.9 ± 1.3	81 ± 28
19	2	Sr-19	3.5 ± 0.8	88 ± 21
19	20	Sr-23	3.6 ± 0.7	86 ± 16
21	2	Sr-25	3.4 ± 1.5	90 ± 39
22	2	Sr-30	3.3 ± 1.1	92 ± 30
23	2	Sr-31	2.3 ± 0.6	120 ± 30
23	50	Sr-33	4.1 ± 1.3	76 ± 24
23	20	Sr-34	3.7 ± 1.0	84 ± 23
23	10	Sr-35	2.4 ± 0.8	116 ± 39
25	2	Sr-37	1.8 ± 0.8	135 ± 62
25	50	Sr-39	4.1 ± 2.0	78 ± 38
27	20	Sr-47	3.1 ± 1.3	97 ± 40
27	10	Sr-48	2.6 ± 1.1	109 ± 46
27	2	Sr-50	2.1 ± 1.0	127 ± 59
29	2	Sr-51	3.2 ± 0.9	96 ± 28
29	20	Sr-54	2.9 ± 0.7	102 ± 24
32	2	Sr-58	4.3 ± 1.5	74 ± 26

BGD
10, 2039–2067, 2013

Table 3. Amounts of ^{90}Sr released to the sea from the Fukushima NPP by June 2011, in TBq ($\cdot 10^{12}$ Bq), calculated using the published estimates of ^{137}Cs releases and a ^{90}Sr/^{137}Cs ratio of 0.0265 ± 0.0006. The leakage ^{90}Sr occurred in December 2011 is not included in these estimates.

Reference	^{137}Cs (PBq)		^{90}Sr (TBq)	
	Range	Average	Range	Average
Tsumune et al. (2011)		3.5 ± 0.7		93 ± 19
Bailly du Bois et al. (2011)	10–34.	22	265–901	583 ± 13
Rypina et al. (2013)	9.5–22	16.2 ± 1.6	252–583	429 ± 43
Charette et al. (2012)	11–16		292–424	

BGD
10, 2039–2067, 2013

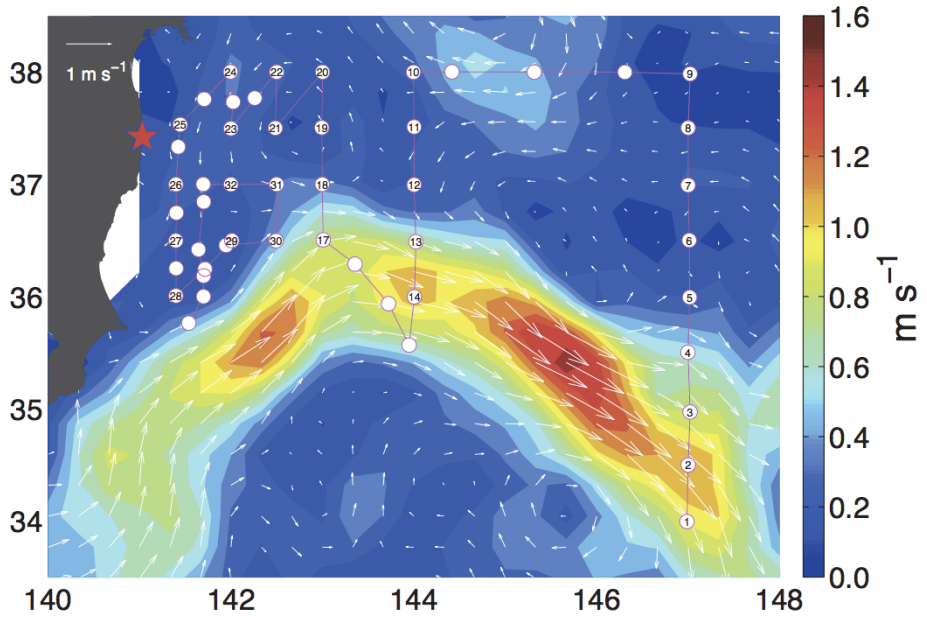

Fig. 1. Sampling stations, with their corresponding stations number (see Table 1 for more details), superimposed on surface velocities (from Buesseler et al., 2012).

BGD
10, 2039–2067, 2013

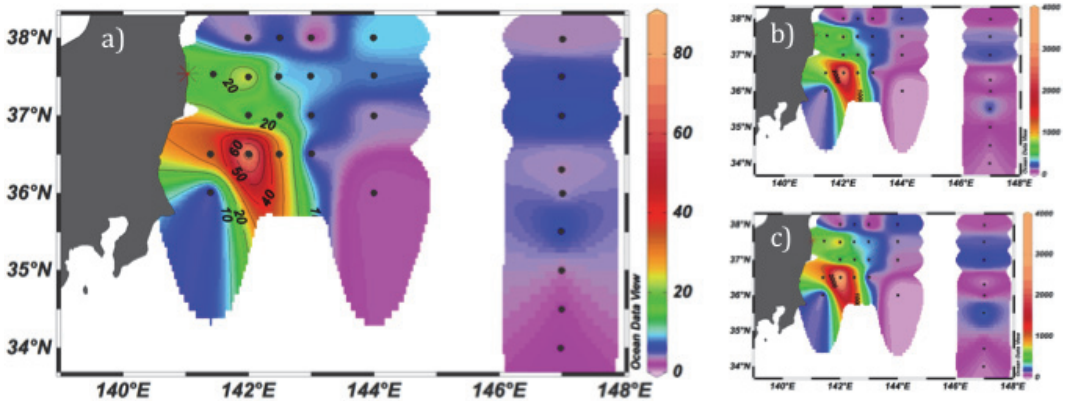

Fig. 2. (a) Activities of ^{90}Sr in surface seawater samples (expressed in Bq m^{-3}). The right hand plots show the specific activity of ^{137}Cs **(b)** and ^{134}Cs **(c)** in surface waters from Buesseler et al., 2012.

BGD
10, 2039–2067, 2013

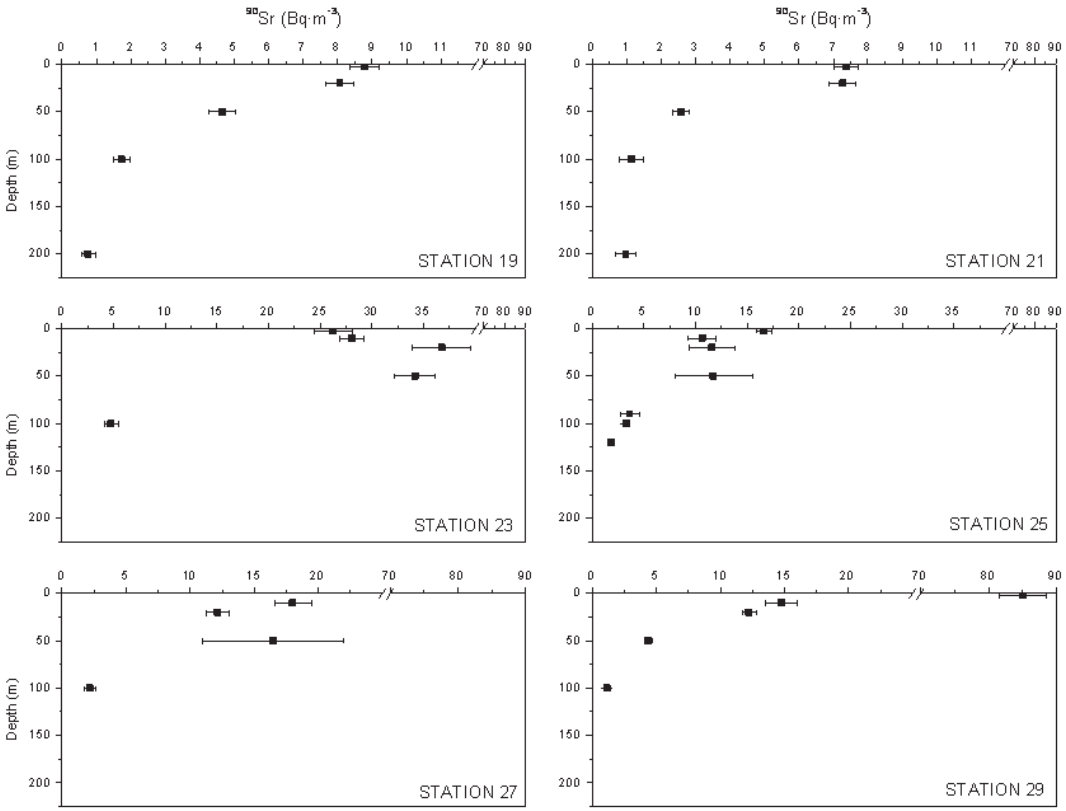

Fig. 3. Specific activities of ^{90}Sr in shallow profiles at the 6 closest stations of Fukushima Dai-ichi NPP.

BGD
10, 2039–2067, 2013

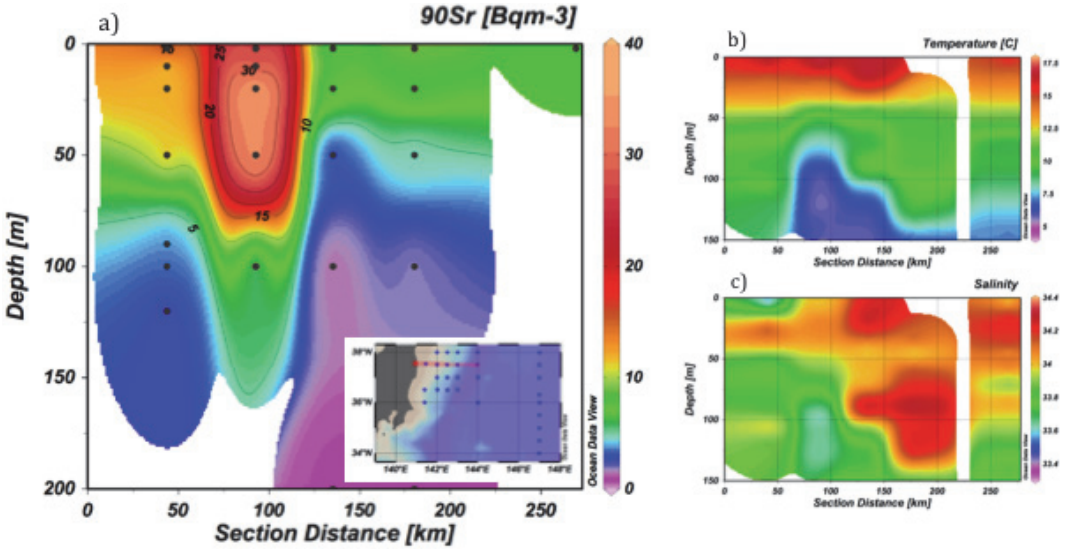

Fig. 4. Specific activities of ^{90}Sr (expressed in Bqm^{-3}) and temperature and salinity contour plots in a section at Fukushima NPP latitude. It comprises stations 25 (left hand, closest to Fukushima), 23, 21, 19 and 11 (right hand).

BGD
10, 2039–2067, 2013

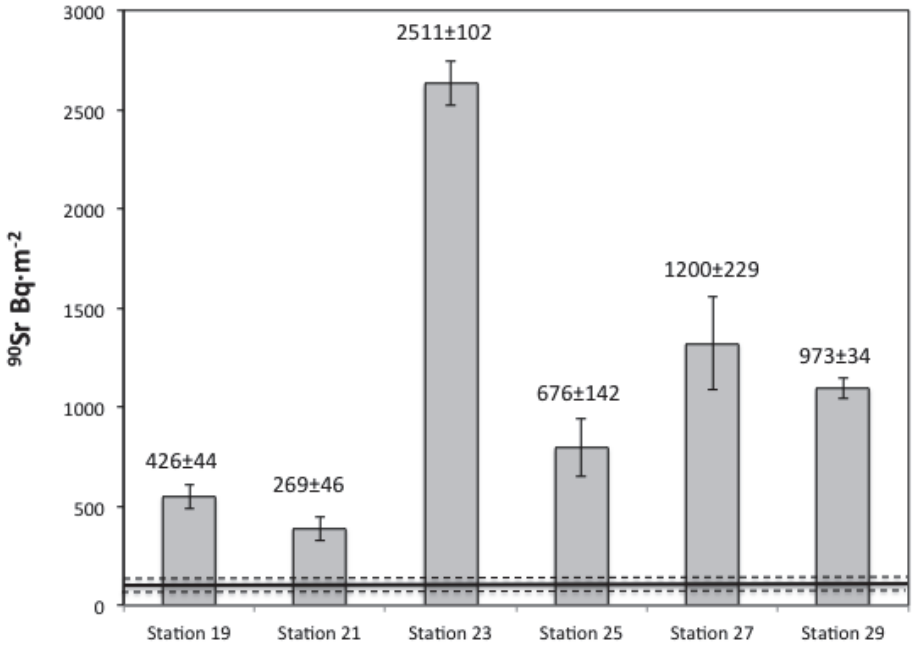

Fig. 5. Total inventories of ^{90}Sr in the upper 100 m of the water column at stations 19, 21, 23, 25, 27 and 29. The solid line represents the background value for this area (120 ± 40 Bq m^{-2}) and the numbers above the bars indicate the Fukushima-derived inventory (subtracting the background).

BGD
10, 2039–2067, 2013

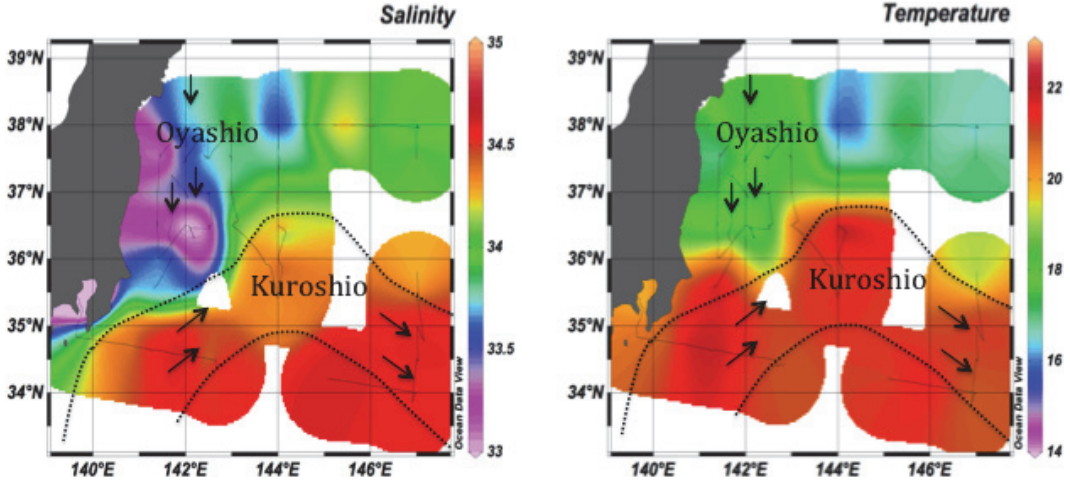

Fig. 6. Surface salinity and temperature measured during the KOK cruise in June 2011. The Oyashio and Kuroshio currents are depicted.

BGD
10, 2039–2067, 2013

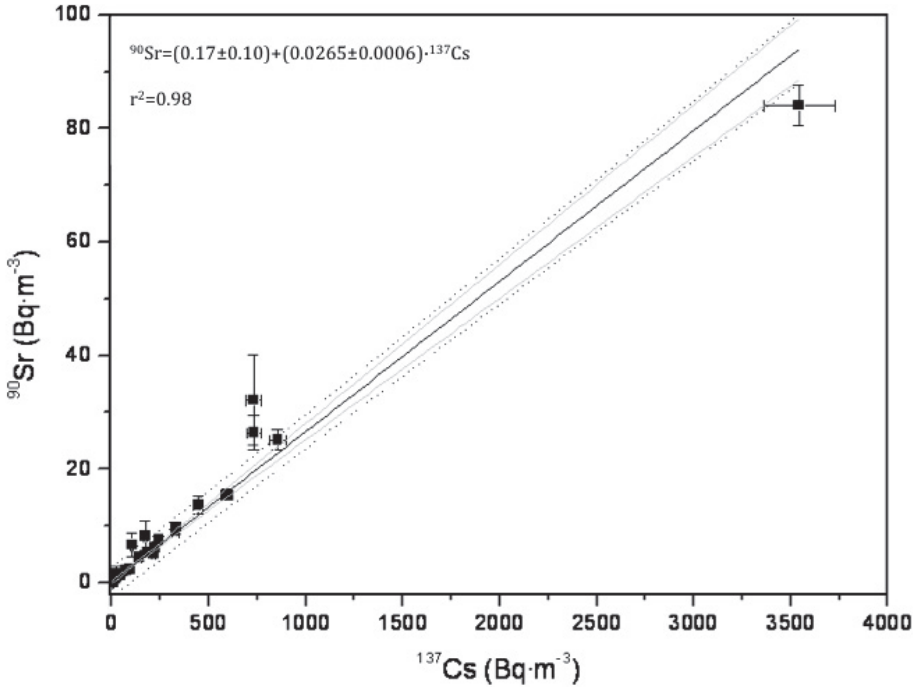

Fig. 7. Fukushima-derived ^{90}Sr/^{137}Cs ratio in surface waters. Background concentrations of both ^{137}Cs ($2.1 \pm 0.5\,\mathrm{Bq\,m^{-3}}$) and ^{90}Sr ($1.2 \pm 0.2\,\mathrm{Bq\,m^{-3}}$) have been subtracted. ^{137}Cs values taken from Buesseler et al. (2012), which were collected simultaneously with the samples for ^{90}Sr presented in this study. The average ^{90}Sr/^{137}Cs ratio is 0.0265 ± 0.0006.

BGD
10, 2039–2067, 2013

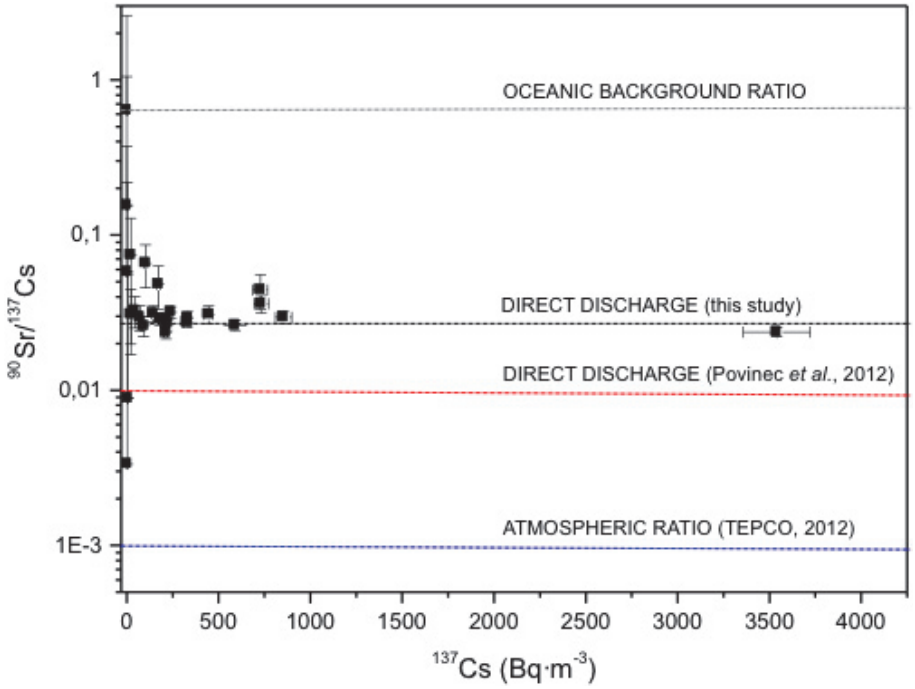

Fig. 8. The $^{90}Sr/^{137}Cs$ ratio as a function of ^{137}Cs specific activities. The grey dashed line represents the oceanic background ratio decay corrected to 2011. Also shown are the $^{90}Sr/^{137}Cs$ ratios corresponding to the oceanic background ratio, the direct discharges from Fukushima Dai-ichi and the atmospheric deposition.

BGD
10, 2039–2067, 2013