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Cesium, iodine and tritium in NW Pacific waters – a comparison of the Fukushima impact with global fallout

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Abstract. Radionuclide impact of the Fukushima Dai-ichi nuclear power plant accident on the distribution of radionuclides in seawater of the NW Pacific Ocean is compared with global fallout from atmospheric tests of nuclear weapons. Surface and water column samples collected during the *Ka'imikai-o-Kanaloa* (*KOK*) international expedition carried out in June 2011 were analyzed for ^{134}Cs, ^{137}Cs, ^{129}I and ^{3}H. The ^{137}Cs, ^{129}I and ^{3}H levels in surface seawater offshore Fukushima varied between 0.002–3.5 Bq L^{-1}, 0.01–0.8 µBq L^{-1}, and 0.05–0.15 Bq L^{-1}, respectively. At the sampling site about 40 km from the coast, where all three radionuclides were analyzed, the Fukushima impact on the levels of these three radionuclides represents an increase above the global fallout background by factors of about 1000, 50 and 3, respectively. The water column data indicate that the transport of Fukushima-derived radionuclides downward to the depth of 300 m has already occurred. The observed ^{137}Cs levels in surface waters and in the water column are compared with predictions obtained from the ocean general circulation model, which indicates that the Kuroshio Current acts as a southern boundary for the transport of the radionuclides, which have been transported from the Fukushima coast eastward in the NW Pacific Ocean. The ^{137}Cs inventory in the water column is estimated to be about 2.2 PBq, what can be regarded as a lower limit of the direct liquid discharges into the sea as the seawater sampling was carried out only in the area from 34 to 37° N, and from 142 to 147° E. About 4.6 GBq of ^{129}I was deposited in the NW Pacific Ocean, and 2.4–7 GBq of ^{129}I was directly discharged as liquid wastes into the sea offshore Fukushima. The total amount of ^{3}H released and deposited over the NW Pacific Ocean was estimated to be 0.1–0.5 PBq. These estimations depend, however, on the evaluation of the total ^{137}Cs activities released as liquid wastes directly into the sea, which should improve when more data are available. Due to a suitable residence time in the ocean, Fukushima-derived radionuclides will provide useful tracers for isotope oceanography studies on the transport of water masses during the next decades in the NW Pacific Ocean.

1   Introduction

The largest amount of radionuclides released into the atmosphere up to now (Table 1), representing the main source of anthropogenic radionuclides in the world ocean, has been from atmospheric tests of nuclear weapons (global fallout) carried out mainly in 1950s and early 1960s (UNSCEAR, 2008). Recently, large quantities of radioactive materials were released into the atmosphere and coastal waters of the NW Pacific Ocean following a nuclear accident at the Fukushima Dai-ichi nuclear power plant (NPP), which increased ^{137}Cs concentrations in coastal seawater off Fukushima up to eight orders of magnitude above the global fallout background (TEPCO, 2011; MEXT, 2011).

The Fukushima accident happened due to the failure of the cooling system of the NPP after the Tohoku earthquake, and subsequent unexpectedly high tsunami waves on 11 March 2011. It was classified by the government of Japan on the INES scale (International Nuclear and Radiological Event Scale; IAEA, 2011) at the maximum level of 7, similar to the Chernobyl accident, which happened in 1986 in the former Soviet Union (present Ukraine) (IAEA, 2003).

The atmospheric radionuclide releases during the Fukushima accident were estimated to be the highest for ^{131}I (153–160 PBq) and ^{137}Cs (13–15 PBq) (Chino et al., 2012). Stohl et al. (2012) estimated even higher atmospheric releases for ^{137}Cs (23–50 PBq). The discharged radioactive material, in addition to ^{131}I and ^{137}Cs, also included ^{134}Cs, ^{132}Te, ^{132}I, ^{136}Cs, and other radionuclides, as well as radioactive noble gases (^{133}Xe, ^{135}Xe) (Bowyer et al., 2011). The contribution of ^{134}Cs was similar to ^{137}Cs as the ^{134}Cs / ^{137}Cs activity ratio was close to one (Masson et al., 2011).

Apart from the contamination of Japan (and the Sea of Japan, Inoue et al., 2012a), due to prevailing western winds, the radionuclides emitted to the atmosphere were mainly transported from Fukushima over the Pacific Ocean, then to the Americas, the Atlantic Ocean, Europe, and back to the Asian continent. At the beginning of April, the global atmosphere was labeled with Fukushima-derived radionuclides (Masson et al., 2011; Hernández-Ceballos et al., 2012; Povinec et al., 2013). The released radionuclides were mostly deposited over the North Pacific Ocean (about 80 %), about 20 % over Japan, and less than about 2 % over the Atlantic and Europe (Stohl et al., 2012; Yoshida and Kanda, 2012; Morino et al., 2011).

Except atmospheric radionuclide releases, which occurred mostly due to hydrogen explosions at the Fukushima NPP (unit 1 and 3), large amounts of liquid radioactive wastes were directly discharged from the Fukushima Dai-ichi NPP into the ocean. A large volume of contaminated water was produced during emergency cooling of reactors using fresh water, and later also by seawater. Some of this water was unintentionally discharged directly into the sea, which widely contaminated coastal waters off the Fukushima NPP, as re-

ported by the Tokyo Electric Power Company (TEPCO) and Ministry of Education, Culture, Sports, Science and Technology (MEXT), and other investigators (TEPCO, 2011; MEXT, 2011; Aoyama et al., 2012; Buesseler et al., 2012; Tsumune et al., 2012; Povinec et al., 2012a). The total amounts of ^{137}Cs directly released into the sea have been estimated to be 3.5–4 PBq (Tsumune et al., 2012, 2013; Kawamura et al., 2011), 11–16 PBq (Charette et al., 2013), 16.2 ± 1.6 PBq (Rypina et al., 2013), and even up to 27 ± 15 PBq (Bailly du Bois et al., 2012).

As the cooling water directly interacted with ruptured nuclear fuel rods, it was estimated that 0.1–1 PBq of ^{90}Sr was also released into the ocean (Povinec et al., 2012a). This evaluation is, however, strongly dependent on the estimation of the ^{137}Cs liquid releases as the ^{90}Sr / ^{137}Cs activity ratios observed in seawater offshore Fukushima were used for the evaluation of the ^{90}Sr source term. Recently, Casacuberta et al. (2013), analyzing ^{90}Sr in seawater samples collected in the NW Pacific Ocean during June 2011 (the *KOK* cruise, Buesseler et al., 2012), estimated the ^{90}Sr source term to be 0.09–0.9 PBq, which is very similar to that of Povinec et al. (2012).

The direct discharge of contaminated water into the sea has significantly elevated radionuclide concentrations in coastal seawater, as well as in the NW Pacific Ocean. The peak ^{137}Cs values were observed at the discharge point of the Fukushima NPP into the sea on 30 March (47 kBq L^{-1}) and on 6 April (68 kBq L^{-1}) (TEPCO, 2012). Several papers have already discussed ^{134}Cs and ^{137}Cs concentrations in surface waters of the open NW Pacific Ocean, where the ^{137}Cs activity concentrations ranged from a few mBq L^{-1} to a few Bq L^{-1} (Buesseler et al., 2011, 2012; Aoyama et al., 2012; Honda et al., 2012; Inoue et al., 2012b).

The aim of the present study is to discuss radionuclide records in North Pacific waters, to evaluate radionuclide source terms and budgets, to assess an impact of the Fukushima accident on radionuclide levels in the NW Pacific waters, to explain the observed radionuclide patterns, and to compare them with global fallout data and post-Chernobyl measurements carried out in the NW Pacific Ocean.

2   Radionuclides of interest and their pre-Fukushima levels in North Pacific waters

Although ^{131}I was released after the Fukushima accident at the largest amounts, it does not represent a radionuclide frequently studied in the marine environment due to its short half-life ($T_{1/2} = 8.02$ d). The most important radionuclide in the Fukushima case is ^{137}Cs, as it was released in large quantities, and it has a relatively long half-life ($T_{1/2} = 30.17$ yr). The relatively short-lived isotope of cesium (^{134}Cs, $T_{1/2} = 2.06$ yr) can clearly identify cesium of the Fukushima NPP origin, as there is no remaining contribution from global fallout and the 1986 Chernobyl accident.

Table 1. Radionuclide inventories in the atmosphere and ocean (in PBq).

| Nuclide | Half-life | Natural inventory[a] Atmosphere[a] | Global fallout Atmosphere[a] | Global fallout inventory in the ocean[b] | | Discharges from reprocessing facilities to the ocean[b] | | Chernobyl | | Fukushima | |
				Total inventory in the ocean	Inventory in 2010 in the ocean	Total inventory	Inventory in 2010	Atmosphere[c]	Ocean[b]	Atmosphere	Ocean
^{131}I	8.02 d	–		–	–	39	–	1760		153–160 [e,f]	62[i]
											120[j]
^{137}Cs	30.17 yr	–	950	600	170	40	26	85	16	13–15[e,f]	0.9–3.5[k]
									9[d]	23–50[g]	3.6 ± 0.7[l]
											4[m]
											4.1–4.5[n]
											5.5–5.9[o]
											11–16[p]
											16.2 ± 1.6[r]
											27 ± 15[s]
											5[t]
^{3}H	12.32 yr	2200	186 000	113 000	8000	410	45				0.1–0.5[u]
^{14}C	5730 yr	1	213	130	130	2	2				
^{129}I	15.7×10^6 yr	0.6×10^{-3}	0.4×10^{-3}	0.3×10^{-3}	0.3×10^{-3}	0.04	0.04	0.013×10^{-3}			2.4–7×10^{-6}[v]
											4.6×10^{-6}[x]
^{90}Sr	28.78 yr	–	600	380	105	7	3	1		0.14[h]	0.1–1[y]
											0.09–0.9[z]

[a] UNSCEAR (2008), [b] IAEA (2005), [c] IAEA (2003), [d] decay-corrected inventory in 2010, [e] TEPCO (2011), [f] Chino et al. (2011), [g] Stohl et al. (2012), [h] JG (2011), [i] Hou et al. (2013), [j] Morino et al. (2011), [k] Dietze and Kriest, (2012), [l] Tsumune et al. (2013), [m] Kawamura et al. (2011), [n] Estournel et al. (2012), [o] Miyazawa et al. (2012b), [p] Charette et al. (2013), [r] Rypina et al. (2013), [s] Bailly du Bois et al. (2012), [t] atmospheric deposition (Kawamura et al., 2011), [u] liquid discharges into the sea plus atmospheric deposition (this work), [v] liquid discharges into the sea (this work), [x] atmospheric deposition (this work), [y] Povinec et al., 2012a, [z] Casacuberta et al. (2013)

Anthropogenic tritium and ^{129}I have been recognized as ideal short-term (^{3}H half-life $T_{1/2} = 12.32$ yr) and long-term (^{129}I half-life $T_{1/2} = 15.7$ Myr) oceanographic tracers, important for investigation of water circulation in the ocean (Schlosser et al., 1999; Raisbeck and Yiou, 1999; Hou et al., 2000, 2013; Povinec et al., 2000, 2010, 2011; Shima et al., 2006; He et al., 2013; Suzuki et al., 2010, 2013).

2.1 Cesium-137

The ^{137}Cs has been recognized radioecologically as one of the most important long-lived radionuclides of anthropogenic origin, which has accumulated in the marine environment (Livingston and Povinec, 2000, 2002). It has been considered to be the most important for the long-term radiological impact because of large releases, relatively long half-life, and its relative high bioavailability. Because of its accumulation in tissues, it has been important for delivering radiation doses to the public from the consumption of seafood (Aarkrog et al., 1997).

The most dominant source of ^{137}Cs in the western North Pacific has been global fallout originating from atmospheric nuclear weapons testing (Table 1). The main input into this part of the ocean occurred in the early 1960s due to wet and dry deposition of ^{137}Cs, after large-scale atmospheric nuclear weapons tests were carried out during 1961–1962 by the former Soviet Union at Novaya Zemlya in the Kara Sea (Livingston and Povinec, 2002). The major deposition of ^{137}Cs occurred in the mid-latitudes of the Northern Hemisphere, and specifically in the NW Pacific Ocean where large wet depositions were observed (Aoyama et al., 2006). The NW Pacific Ocean has therefore been well known as the area with the highest deposition of global fallout radionuclides into the ocean (Povinec et al., 2005a; Inomata et al., 2009).

Large amounts of ^{137}Cs were also released from nuclear reprocessing facilities in Sellafield (situated on the western coast of England) and in La Hague (situated in the English Channel) (Table 1), which have had mainly impact on the European seas (Povinec et al., 2003b).

After its release into the atmosphere, the ^{137}Cs has been rapidly associated with aerosols, which represent a major reservoir of pollutants in the atmosphere (Lujanienė et al., 2009). The ^{137}Cs present in the atmosphere mainly from global fallout and the Chernobyl accident has primarily been deposited on the earth's surface, including the ocean by wet and dry deposition (Pham et al., 2011a, 2013).

As ^{137}Cs is a conservative oceanographic tracer, it is primarily dissolved in seawater, and only < 1 % is attached to marine particles. The sediment distribution coefficient (K_d) for ^{137}Cs in coastal and open ocean waters is 4000 and 2000, respectively (IAEA, 2004). The removal of ^{137}Cs from the water column is mainly due to its radioactive decay and diffusion. It has been used frequently in water transport/mixing studies and in investigations of water column processes (Livingston and Povinec, 2002; Ito et al., 2003; Povinec et al., 2003a,b; Lujanienė et al., 2006).

Fortunately, the pre-Fukushima ^{137}Cs concentrations in NW Pacific waters were well established (Povinec et al., 2005a; Aoyama et al., 2006). Two marine radioactivity databases were developed: one at the International Atomic Energy Agency's Environment Laboratories in Monaco (the GLOMARD/MARIS database; Povinec et al., 2004, 2012b) and the second one at the Meteorological Research Institute

in Tsukuba (the HAM database; Aoyama and Hirose, 2004). The ^{137}Cs data can be downloaded from www.iaea.org/monaco/.

Long-term changes in surface ^{137}Cs concentrations in two latitudinal belts at 25–40° N (divided by the line at 180° E to the western and the eastern belt) of the North Pacific Ocean (representing upstream and downstream of the Kuroshio Extension, respectively, where the water masses of the Kuroshio and Oyashio currents mix) are shown in Fig. 1. The surface ^{137}Cs concentrations in the western and eastern belt decreased exponentially during the period 1970–2000 (except a small peak observed in 1986 due to the Chernobyl accident), with effective half-lives of 16.5 ± 0.9 yr and 10.6 ± 0.5 yr, respectively. Higher ^{137}Cs concentrations in surface water occurred in the 1980s, probably due to liquid radioactive discharges from the nuclear fuel reprocessing plant at Tokai, which were, however, by several orders of magnitude lower than releases from the European reprocessing plants (Mizutani et al., 2009). By combining all ^{137}Cs data, the mean effective half-life of ^{137}Cs in North Pacific surface waters was estimated to be 13 ± 1 yr (Povinec et al., 2005a).

Some of the ^{137}Cs water profiles measured in the NW Pacific Ocean are presented in Fig. 2. Samples collected in November 1997 during the IAEA'97 expedition at 30–35° N and 150–170° E showed surface ^{137}Cs concentrations around 2.5 mBq L^{-1}, subsurface ^{137}Cs maxima at around 3 mBq L^{-1}, a decrease down to 2 mBq L^{-1} at 500 m, and levels < 0.1 mBq L^{-1} at water depths below 1000 m (Povinec et al., 2003a), corresponding to concentrations of 1.2, 1.4, 0.95, and < 0.05 mBq L^{-1} in 2011, based on an effective half-life of 13 yr of ^{137}Cs in the Pacific Ocean. On the basis of the data stored in the MARIS database (www.iaea.org/monaco/, the average ^{137}Cs concentrations in NW and NE Pacific surface waters for the year 2010 are estimated to be 1.0 ± 0.1 mBq L^{-1} and 0.9 ± 0.2 mBq L^{-1}, respectively.

2.2 Iodine-129

Iodine-129 has been introduced to the global ocean mainly from nuclear reprocessing facilities (Table 1). Because of its long half-life, it represents an alternative tracer to global fallout radionuclides (Raisbeck and You, 1999; Hou et al., 2000, 2007, 2013; Povinec et al., 2000, 2010, 2011; Shima et al., 2006; Toyama et al., 2012; He et al., 2013). As it is a soft beta-emitter with maximum beta energy of 154 keV, and very long half-life, it is less important in the view of radiation protection of humans. However, it is an excellent analogue to reconstruct levels and for distribution of short-lived ^{131}I in the environment, which is usually released at largest quantities during nuclear accidents (Hou et al., 2003). ^{129}I is a useful environmental tracer for investigation of atmosphere–ocean interactions, as well as of biogeochemical cycles of stable iodine through chemical speciation studies of ^{129}I and ^{127}I in the environment (Hou et al., 2001, 2009a, 2012).
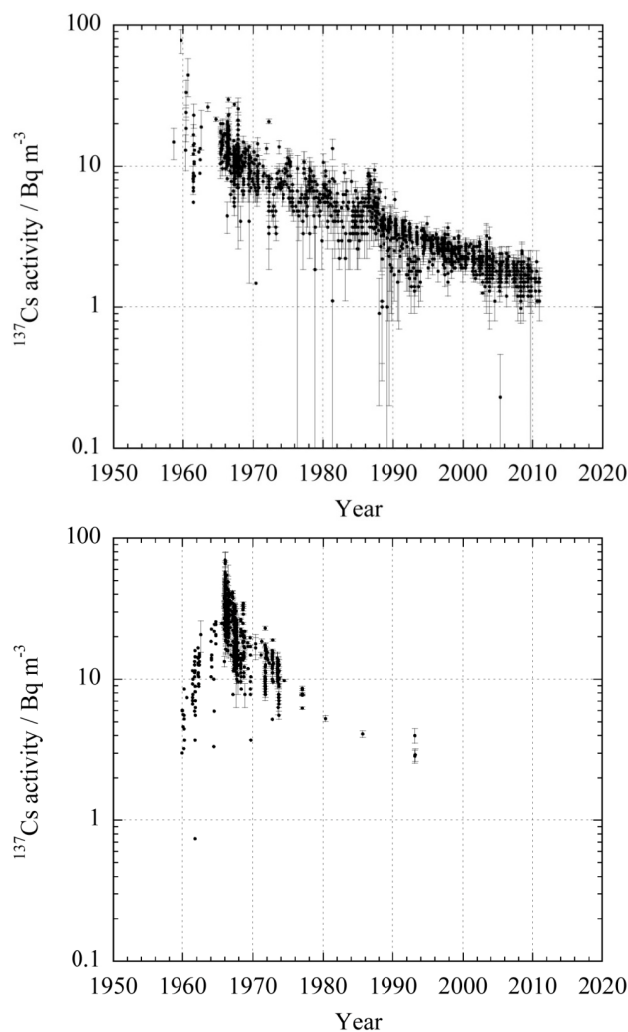

Fig. 1. Long-term changes of ^{137}Cs concentrations in surface seawater in two latitudinal belts at 25–40° N, divided at 180° E for the western (top) and the eastern (bottom) belt of the North Pacific Ocean.

Unfortunately, there are only a few data available on the distribution of ^{129}I in NW Pacific waters. Povinec et al. (2000) reported results on ^{129}I concentrations in a water profile collected in 1995 during the Japan–South Korea–Russia–IAEA expedition on investigation of radioactive waste dumping sites in the NW Pacific Ocean. The samples collected offshore Kamchatka (52°30′ N, 159°10′ E) showed ^{129}I concentrations up to two orders of magnitude higher than in the open ocean, which were probably caused by leakages from the dumping site (similar to that observed in the Kara Sea, Arctic Ocean; Povinec et al., 2000). The IAEA'97 results in the NW Pacific Ocean (Fig. 3) showed surface ^{129}I values around 1.5×10^7 atoms L^{-1}, subsurface maxima at around 2.1×10^7 atoms L^{-1}, a decrease down to 1.5×10^7 at 500 m, and a fast decrease $\leq 0.8 \times 10^7$ atoms L^{-1} at depths below 1000 m (Povinec et al., 2010). Higher ^{129}I
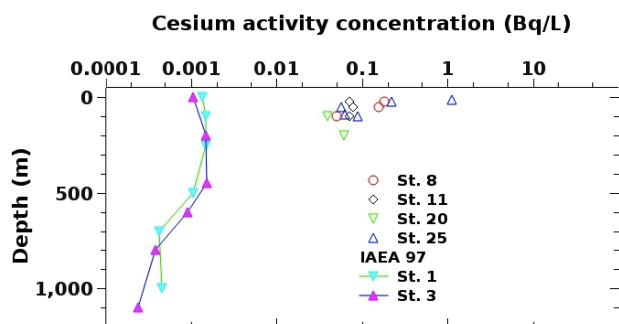
Cesium activity concentration (Bq/L)

Fig. 2. A comparison of ^{137}Cs profiles as obtained from the IAEA'97 expedition (global fallout impact; Povinec et al., 2003a) with the results presented in this paper, which are based on analysis of seawater samples collected during the *KOK* expedition (Fukushima impact) in the NW Pacific Ocean. IAEA'97 data were decay-corrected to June 2011.
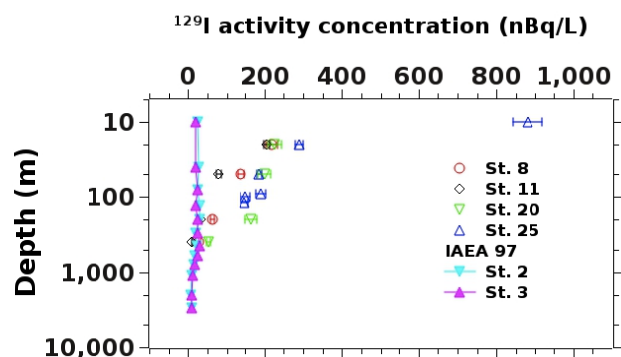
^{129}I activity concentration (nBq/L)

Fig. 3. A comparison of ^{129}I profiles as obtained from the IAEA'97 expedition (global fallout impact; Povinec et al., 2010) with the results presented in this paper, which are based on analysis of seawater samples collected during the *KOK* expedition (Fukushima impact) in the NW Pacific Ocean.

levels (up to 3×10^7 atoms L^{-1} at 200 m water depth) were observed close to the Enewetak and Bikini atolls (at around 110° N, 162° E), where ^{129}I concentration minima at around 500 m and secondary maxima (1.8×10^7 atoms L^{-1}) at about 1000 m were observed. It has been concluded that the NW Pacific stations could be influenced by a local impact of nuclear weapons testing carried out on the Enewetak and Bikini atolls during the 1950s. Suzuki et al. (2010) reported ^{129}I concentrations in the NW Pacific waters decreasing from the surface (2×10^7 atoms L^{-1}) to 100 m water depth (0.7×10^7 atoms L^{-1}), and then $< 0.2 \times 10^7$ atoms L^{-1} at water depths below 100 m. The corresponding ^{129}I/^{127}I atom ratios were 0.7×10^{-10}, 0.25×10^{-10}, and $< 0.07 \times 10^{-10}$, respectively. More ^{129}I data can be found in papers of Shima et al. (2006) and Toyama et al. (2012). He et al. (2013) reviewed recently the global distribution of ^{129}I in seawater.

Suzuki et al. (2013) found a weak dependence of ^{129}I concentration in seawater on latitude in the NW Pacific Ocean (between 32° and 44° N). The pre-Fukushima ^{129}I concentration in surface NW Pacific waters in the latitude belt of 37–38° N (where most of seawater samples studied in this paper were collected) is expected to be $(1.3 \pm 0.1) \times 10^7$ atoms L^{-1}.

High solubility and long residence time of iodine in the ocean makes ^{129}I an ideal oceanographic tracer for the investigation of water circulation on the global scale. Clearly, we need to understand how much its concentrations have changed in the Pacific Ocean after the Fukushima accident.

2.3 Tritium

Tritium is an ideal tracer used extensively in oceanographic studies, as it is directly incorporated into the water molecule, usually as HTO (or T_2O), and has a suitable half-life (12.32 yr). It is produced not only naturally by interactions

of cosmic rays with nitrogen and oxygen atoms in the upper troposphere and lower stratosphere, but it has also been produced in large amounts in atmospheric nuclear weapons tests (186 000 PBq, Table 1). Its peak concentration in the atmospheric moisture in 1963 was 1000 times higher than its natural cosmogenic background. It has also been released in large quantities from nuclear reprocessing facilities. As it is a soft beta-emitter with maximum beta energy of 18.6 keV only, it is less important in the view of radiation protection of humans (when compared with ^{137}Cs). The penetration of bomb tritium from surface waters into deeper layers of the ocean was used to study pathways and timescales of deep and bottom water formation (e.g., Schlosser et al., 1999).

Tritium distribution in the NW Pacific Ocean was investigated recently in the framework of the WOMARS (Worldwide Marine Radioactivity Studies) project, carried out by IAEA during 1995–2005 (Povinec et al., 2004, 2005a, 2010). The most comprehensive study was, however, the WOCE (World Ocean Circulation Experiment) program conducted in the 1980s and 1990s, which represents the most extensive coverage of ^{3}H in the world ocean (www.eWOCE.org).

The IAEA'97 results in the NW Pacific Ocean (Fig. 4) showed surface ^{3}H values around 1 TU (tritium data are usually expressed in tritium units: 1 TU is ^{3}H/^{1}H atom ratio of 1×10^{-18}, corresponding to activity concentration of 0.118 Bq L^{-1} of water), subsurface maxima (at 200–300 m water depth) around 1.3 TU, a decrease down to 0.7 TU at 500 m, and a fast decrease down to 0.1 TU at water depths below 1000 m (Povinec et al., 2010). On the basis of the data stored in the GLOMARD/MARIS database, the pre-Fukushima ^{3}H concentration in surface NW Pacific waters can be estimated to be 0.6 ± 0.1 TU.

Although tritium is a not so important radionuclide from a radioecological point of view, as it has been used as an oceanographic tracer (also in climate change studies), we
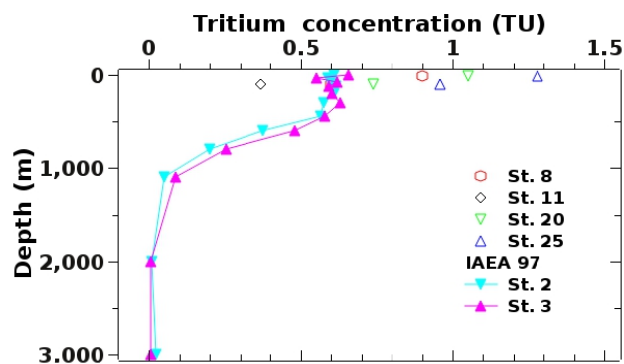
Fig. 4. A comparison of ^{3}H profiles as obtained from the IAEA'97 expedition (global fallout impact; Povinec et al., 2010) with the results presented in this paper, which are based on analysis of seawater samples collected during the *KOK* expedition (Fukushima impact) in the NW Pacific Ocean. IAEA'97 data were decay-corrected to June 2011.

need to know how its concentrations have changed in the NW Pacific Ocean after the Fukushima accident.

3 Pre- and post-Fukushima radionuclide source terms

3.1 Global fallout

Estimated radionuclide releases and their inventories into the world ocean (Table 1) indicate that the most significant source of anthropogenic radionuclides in the ocean is still global fallout. From the total of 950 PBq of ^{137}Cs released during atmospheric nuclear weapons testing (UNSCEAR, 2008), about 600 PBq was deposited on the ocean. The estimated ^{137}Cs inventory in the world ocean in 2010 is about 170 PBq, of which about 90 PBq is in the Pacific Ocean, and around 20 PBq in its latitudinal belt of 25–40° N. The ^{137}Cs global fallout inventories in the ocean could be compared with ^{90}Sr ones, which were by about a factor of 1.6 lower.

Tritium had the highest total inventory in the ocean from global fallout, about 113 000 PBq. The present inventory in the ocean is still the highest one, about 8000 PBq (Table 1). Iodine-129, on the other hand, had the lowest inventory from global fallout, about 300 GBq. Due to its long half-life and specific behavior in the ocean, this inventory did not change with time. The main sources of ^{129}I in the European seas have been, however, authorized discharges from reprocessing nuclear facilities in Sellafield and La Hague (Hou et al., 2009b).

3.2 Chernobyl accident

The Chernobyl accident was the biggest short-term release of ^{137}Cs into the environment (85 PBq), of which about 16 PBq was deposited on the ocean (IAEA, 2003). The present inventory of ^{137}Cs in the ocean from the Chernobyl accident

has been estimated to be around 9 PBq, which is mostly localized in the European seas (the Baltic, the Black, the North, the Mediterranean, the Norwegian and the Barents seas).

For comparison, about 42 PBq of ^{137}Cs has been released into the marine environment from reprocessing facilities in Europe (Sellafield and La Hague), which has mostly influenced its levels in the Irish, the North, the Norwegian and the Barents seas (Povinec et al., 2003b). The amounts of ^{131}I and ^{129}I released during the Chernobyl accident were 1760 PBq and 13 GBq, respectively (IAEA, 2003).

3.3 Fukushima accident

3.3.1 Atmospheric radionuclide releases

Large amounts of radionuclides were released into the atmosphere due to venting of gases from the damaged nuclear reactors, hydrogen explosions and the fires in the Fukushima NPP (JG, 2011). Major radionuclides released into the atmosphere were ^{131}I (153–160 PBq) and radiocesium (^{134}Cs and ^{137}Cs – each 13–15 PBq; Table 1) (TEPCO, 2011; NISA, 2011; IAEA, 2011; Chino et al., 2011). Even higher ^{137}Cs atmospheric releases (23–50 PBq) were estimated by Stohl et al. (2012).

The main atmospheric radionuclide releases occurred between 12 and 16 March, 2011, with smaller contributions up to 24 March, which have been then distributed globally (Masson et al., 2011), and due to wet and dry deposition contaminated the terrestrial and marine environments. If we compare the Fukushima atmospheric radionuclide releases (as estimated at present) with those estimated for the Chernobyl accident (Table 1), we see that they were for ^{131}I and ^{137}Cs lower by about an order of magnitude.

3.3.2 Radionuclide releases into the ocean

Direct liquid radionuclide releases into the marine environment consisted mainly of ^{134}Cs, ^{137}Cs and ^{90}Sr. These are all fission products, with relative fission yields of 6.8, 6.3, and 4.5 %, respectively.

Radiocesium preliminary estimations for direct releases into the sea were made by the TEPCO (JG, 2011), suggesting that 0.94 PBq of ^{137}Cs was discharged during the five-day period from 1 to 6 April 2011 (using a release rate of 1.9×10^{14} Bq day^{-1}). A later estimation of TEPCO (2012) for the period from 26 March to the end of September 2011 was 3.55 PBq. Kawamura et al. (2011) using the TEPCO data and numerical simulations estimated the total ^{137}Cs release for the period from 21 March to 30 April 2011 to be 4 PBq. Bailly du Bois et al. (2012), interpolating ^{137}Cs data obtained for surface waters off Fukushima, estimated the ^{137}Cs release for the period from 25 March to 18 July 2011 to be 27 ± 15 PBq. This approach, as discussed by Dietze and Kriest (2012) using numerical simulations, could probably overestimate the direct ^{137}Cs releases into coastal waters, as their

estimate was also only in the range 0.94–3.5 PBq. Predictions made using global ocean circulation models and the available ^{137}Cs monitoring data gave for the direct ^{137}Cs release between 26 March and 31 May 2011 a value of 3.5 ± 0.7 PBq (Tsumune et al., 2012), which has been recently refined for the period from 26 March to 29 February 2012 to be 3.6 ± 0.7 PBq (Tsumune et al., 2013). Estournel et al. (2012), using an inversion method based on measurements adjacent to the Fukushima NPP, estimated the ^{137}Cs direct release from 20 March to 30 June to be in the interval 4.1–4.5 PBq, adding 1 PBq to their estimation because the simulated results underestimated the ^{137}Cs activities measured offshore Fukushima. Miyazawa et al. (2012, 2013), using an inversion method based on ^{137}Cs data offshore Fukushima and numerical modeling, estimated the direct release of ^{137}Cs from 21 March to 30 April 2011 to be 5.5–5.9 PBq. Both the Estournel et al. (2012) and Miyazawa et al. (2013) estimations could be higher due to evaluating larger ^{137}Cs data sets (not only around the Fukushima Dai-ichi NPP, but also covering larger areas that could be influenced by the coastal current system).

Charette et al. (2013), using a different approach based on Ra isotopes measured in seawater samples collected in June 2011 during the *Ka'imikai-o-Kanaloa* (*KOK*) cruise, which was carried out in the region 34–37° N and 142–147° E (Buesseler et a., 2012), estimated the ^{137}Cs source term due to direct liquid discharges to be 11–16 PBq. Recently, Rypina et al. (2013), using numerical simulations and the ^{137}Cs data of Buesseler et al. (2012) from the same *KOK* sampling cruise, estimated the oceanic source to be 9.1–17.8 PBq, with the mean and standard deviation of 16.2 ± 1.6 PBq. These values were determined by minimizing the data–model mismatch over the *KOK* survey area.

We see that there are still large discrepancies in the estimated direct ^{137}Cs releases into the ocean. Generally, numerical simulations carried out for the coastal waters suggest ^{137}Cs releases between 3 and 6 PBq, while the simulations of Rypina et al. (2013) for a larger sea area suggest releases starting around 9 PBq. The results of Charette et al. (2013) are within the source term predicted by Rypina et al. (2013), while the interpolating method of Bailly du Bois et al. (2012) suggests the highest releases. Therefore, more work needs to be done for more precise estimation of ^{137}Cs liquid releases into the ocean.

The estimation of the atmospheric deposition of ^{137}Cs over the Pacific Ocean is an even more difficult task in the evaluation of its source term in the ocean (Kawamura et al., 2011; Morino et al., 2011; Buesseler et al., 2012; Stohl et al., 2012; Honda et al., 2012; Rypina et al., 2013). Dietze and Kriest (2012) estimated the accumulated atmospheric deposition of ^{137}Cs in the range of 50–200 kBq m^{-2}. The ^{131}I / ^{137}Cs activity ratio in surface seawater indicates that most of the radiocesium observed in coastal waters has been the result of a direct discharge into the ocean, rather than resulting from atmospheric deposition (Tsumune et al., 2012, 2013). Re-

cently, Rypina et al. (2013) using the *KOK* data estimated mean and standard deviation for the atmospheric source to be only 0.5 ± 2.7 PBq (the range 0–11 PBq). The *KOK* data are, however, not well-suited for constraining the atmospheric source strength because most of the fallout ^{137}Cs had left the survey area by mid-June. Therefore, the reported atmospheric amplitude, especially its mean value, is possibly severely underestimated. The oceanic source amplitude, on the other hand, is well-constrained by the observed *KOK* ^{137}Cs concentrations, which are due almost entirely to the oceanic discharge (more than 95 % of ^{137}Cs remaining in the water within ~ 600 km of Fukushima in mid-June 2011 was due to the direct oceanic discharge).

The ^{129}I, similar to ^{137}Cs, is a fission product with a relative low fission yield (0.8 %). On the basis of ^{129}I measurements in seawater offshore Fukushima, Hou et al. (2013) estimated that about 2.3 GBq of ^{129}I was released into coastal waters directly as liquid wastes, and about 1.1 GBq was deposited offshore Fukushima in the area 34–41° N and 137–145° E.

Tritium production in the Fukushima boiling water reactors was mainly due to fission with a yield of 0.01 %, as well as from the neutron activation in the ^{7}Li(n,α)^{3}H reaction (Hou, 2005). There were no data available on release rates of tritium into the sea offshore Fukushima.

4 Samples and methods

4.1 Seawater samples

Seawater samples were collected from offshore Fukushima during the international research cruise with participation of 13 institutions, which was organized by US scientists on 3–17 June 2011 using the research vessel *Ka'imikai-o-Kanaloa* (*KOK*) of the University of Hawaii (Buesseler et al., 2012). The cruise track extended from 34 to 37° N, and from 142 to 147° E (from about 30 to 600 km off Japan), visiting altogether 50 sampling stations (Fig. 5). Both surface and water profile samples were collected. Of these samples, 11 depth profiles were analyzed in this work for ^{137}Cs and ^{134}Cs, and 4 depth profiles were analyzed for ^{129}I and ^{3}H. The collected seawater samples for tritium and ^{129}I analyses were stored in 1 L glass bottles with air-tight covering so no exchange with the surrounding air was possible. The samples for cesium analysis were stored in plastic containers from 1 L (profiles) to 30 L (surface) volumes.

4.2 Analytical methods

All chemical reagents used during sample preparations were of analytical grade. All solutions were prepared using deionized water. The data quality was assured by regular participation in intercomparison exercises, and by analysis of reference materials. The IAEA reference materials IAEA-381 (Povinec et al., 2002) and IAEA-443 (Pham et al., 2011b)

were used for quality control of ^{137}Cs and ^{3}H analyses, and the IAEA-418 (Pham et al., 2010) and the NIST standard reference material (NIST-SRM-4949c) were used for quality control of ^{129}I analyses.

4.2.1 Tritium analysis

Pre-screening of ^{3}H levels in seawater samples (to avoid possible contamination) was carried out by direct ^{3}H counting (after triple distillation) in water-liquid scintillator cocktails using Packard liquid scintillation spectrometer. Tritium was then precisely analyzed in seawater samples using the ^{3}He in-growth method (Palcsu et al., 2010).

The method consists of three major steps:

(i) The water sample is put into a stainless steel vessel, and the dissolved gases including helium are then removed from the water by vacuum pumping.

(ii) The samples are stored for several months so that ^{3}He atoms are produced by tritium decay.

(iii) The helium fraction is admitted to a dual collector noble gas mass spectrometer; the abundance of the tritiogenic ^{3}He is then measured, from which ^{3}H activity is calculated. The sensitivity of the method is 0.01 TU.

4.2.2 Iodine-129 analysis

The method for preparation of AgI targets for accelerator mass spectrometry (AMS) measurements has already been described (Biddulph et al., 2000; Povinec et al., 2000, 2010; Hou et al., 2012). Therefore, it will be only briefly mentioned here. A total of 200–500 mL of seawater samples was spiked with ^{125}I$^-$ and transferred to a separation funnel. After addition of 0.5 mg ^{127}I carrier (Woodward Inc., with a ^{129}I/^{127}I ratio lower than 1×10^{-13}), 3 mL of 1 M NaHSO$_3$ solution and 3 M HNO$_3$ were added to convert all inorganic iodine to iodide. After addition of NaNO$_2$ to oxidize iodide to I$_2$, iodine was then extracted into CHCl$_3$. Iodine in CHCl$_3$ phase was then back-extracted to the water phase using 10 mL of 0.1 mM NaHSO$_3$ solution. A total of 0.5 mL of 1.0 mol L^{-1} AgNO$_3$ solution was added to the back-extracted aqueous phases to precipitate iodide as AgI, which was separated by centrifuge. The obtained AgI precipitate was dried at 70 °C and used for AMS measurement of ^{129}I. ^{125}I in the precipitate was counted using a NaI(Tl) gamma-detector to monitor the chemical yield of iodine in the separation (Hou et al., 2007). An ICP-MS system (X Series II, Thermo Fisher Scientific, Waltham, MA, USA) equipped with an Xs-skimmer cone and standard concentric nebulizer was used for measurement of ^{127}I. Cs$^+$ (to 2.0 ppb) as internal standard and 1 % NH$_3$ media were applied for measurements.

The ^{129}I/^{127}I atom ratios in total iodine samples were determined by AMS at the Vienna Environmental Research Accelerator (VERA) of the University of Vienna using a 3 MV Pelletron (Wallner et al., 2007), and at the 3 MV Pelletron AMS facility of the University of Arizona (Biddulph et al., 2000). The machine ^{129}I/^{127}I background ratio was 3×10^{-14}. The blanks prepared using the same procedure as the samples gave for the ^{129}I/^{127}I ratios values below 1×10^{-13}, which was by about three orders of magnitude lower than measured ^{129}I/^{127}I ratios in seawater samples.

4.2.3 Cesium-137 and ^{134}Cs analysis

The method for ^{137}Cs analysis in seawater has been recently described in detail (Levy et al., 2011; Povinec et al., 2012b). Therefore, it will be only briefly mentioned here. The seawater samples were nondestructively counted directly using high-efficiency HPG spectrometers placed in the CAVE (Counting Laboratory for Environmental Radionuclides) underground facility of IAEA-EL (Povinec et al., 2005b). The counting periods were between 24 and 72 h.

4.2.4 Modeling the water mass transport

The LAMER (Long-term Assessment Model of Radionuclides in the oceans) code was used for simulation of the dispersion of ^{137}Cs released from the Fukushima accident into the Pacific and Indian oceans (Nakano and Povinec, 2012). The LAMER is the robust diagnostic OGCM code of the medium resolution with grids of two degrees for latitudinal and longitudinal directions (Nakano and Povinec, 2003a). The parameters used, as well as the complete model, were validated using global ^{137}Cs data from the atmospheric nuclear weapons tests. In the vertical direction, 15 layers from the ocean surface to its bottom were used for determination of the velocity fields. A particle-tracking model with a random walk is used for simulation of advection and diffusion processes.

The model can also deal with scavenging processes (Nakano and Povinec, 2003b). However, in the case of cesium, scavenging processes are not of importance because it behaves in seawater as a conservative tracer. As the transport of Fukushima radionuclides in the Pacific Ocean using the LAMER model has already been described (Nakano and Povinec, 2012), we shall not present details in this paper. We shall focus here on the description of the dispersion of ^{137}Cs from the Fukushima NPP in waters of the NW Pacific Ocean, and on the comparison of predicted ^{137}Cs levels with recent measurements.

5 Results and discussion

5.1 ^{137}Cs in seawater of the NW Pacific after the Fukushima accident

The distribution of the Fukushima-derived ^{137}Cs in surface waters 30–600 km offshore the Fukushima NPP (Fig. 5), based on samples collected during the *KOK* cruise and
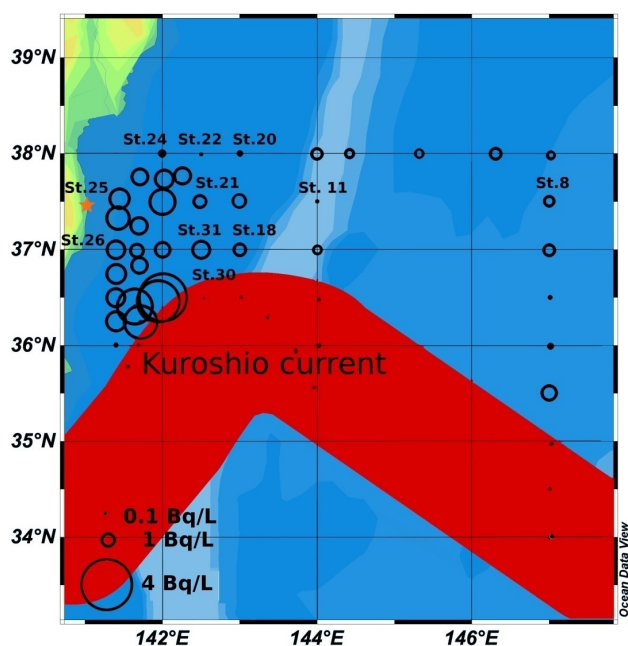
Fig. 5. Distribution of ^{137}Cs in surface waters offshore Fukushima. Data obtained in the present work (identified by St. numbers), as well as those of Buesseler et al. (2012), both from the *KOK* expedition, are presented.

reported by Buesseler et al. (2012), as well as on the results presented in this paper (identified by St. numbers), shows that the measured ^{137}Cs activity concentrations in surface waters ranged from $1.8 \, \text{mBq L}^{-1}$ to $3500 \, \text{mBq L}^{-1}$, up to 3500 times higher than the global fallout background, although the cruise track did not go closer than 30 km from the coast. The highest activities were observed at sampling sites around 36.5° N, 142° E, southeast of the Fukushima NPP. It is evident that the Kuroshio Current acts as a southern boundary for the transport of the radionuclides in the NW Pacific Ocean. The sampling sites around 37.5° N, 141.5° E, east of the Fukushima NPP, showed lower ^{137}Cs levels, only around $1 \, \text{Bq L}^{-1}$. The elevated ^{137}Cs levels covered an area of around 150 000 km^2 (south of 38° N and west of 147° E). Even at distances around 600 km off Fukushima, ^{137}Cs activity concentrations of around $0.3 \, \text{Bq L}^{-1}$ were found (i.e., by about a factor of 300 above the global fallout background of $1 \, \text{mBq L}^{-1}$).

We noticed that a considerable decrease in surface ^{137}Cs levels took place between the measurements, which were carried out earlier and during the *KOK* cruise. About $24 \, \text{Bq L}^{-1}$ of ^{137}Cs were measured in seawater 30 km offshore Fukushima on 23 March 2011, which could be compared to $3.5 \, \text{Bq L}^{-1}$ measured during the *KOK* expedition. Figure 5 also indicates that an atmospheric deposition of ^{137}Cs occurred within the 600 km zone offshore Fukushima.

The ^{134}Cs and ^{137}Cs water profiles measured in this work (plus the ^{137}Cs profiles already shown in Fig. 2, which were

compared with the pre-Fukushima profiles measured during the IAEA'97 expedition in the NW Pacific Ocean) and presented in Fig. 6 indicate that mostly surface water maxima were observed, although at some stations (e.g., St. 24, 22, 18, 31) subsurface maxima (at 20–50 m) were observed as well. The Fukushima-derived ^{134}Cs and ^{137}Cs penetrated up to June 2011 to at least 200 m water depth (St. 30 and 20). This figure also confirms that the ^{134}Cs / ^{137}Cs activity ratio in the analyzed seawater samples was close to 1, clearly indicating that the source of these radionuclides was the Fukushima accident. The ^{137}Cs water profiles obtained for St. 25, 20, 11 and 8 (Fig. 2) also confirm that the Fukushima signal was well visible in all four stations, especially at St. 25, where surface ^{137}Cs levels exceeded the global fallout values by about three orders of magnitude.

The ^{137}Cs inventory in the water column of the NW Pacific due to the Fukushima accident, estimated using the Buesseler et al. (2012) data and those presented in this paper, is $2.2 \pm 0.3 \, \text{PBq}$, confirming an essential contribution to its total ^{137}Cs inventory in the NW Pacific Ocean (Table 1). This value can be regarded as a lower limit of the direct ^{137}Cs liquid discharges into the sea as the seawater sampling was carried out only in the area from 34 to 37° N, and from 142 to 147° E.

5.2 Comparison of modeling and experimental ^{137}Cs results in NW Pacific waters

Because of its low resolution (2 by 2 degrees), the LAMER code is suitable only for simulating the open ocean distribution of ^{137}Cs at the surface and in the water column, about one year after the Fukushima accident (Nakano and Povinec, 2012). As indicated in Table 1, on the basis of modeling exercises of Kawamura et al. (2011), we expect that the cumulative deposition of ^{137}Cs over the western NW Pacific Ocean was around 5 PBq. The total source term of 9 PBq (4 PBq from direct liquid releases, and 5 PBq from atmospheric deposition) was used therefore in the oceanic dispersion model for prediction of ^{137}Cs levels in the open NW Pacific Ocean.

Unfortunately, there are not many recent ^{137}Cs data available for surface waters of the North Pacific Ocean. Surface seawater samples were collected by 17 cargo ships and by several research vessels in 2011 and 2012, as described by Aoyama et al. (2013). Although the available data set is still very small, a comparison of predicted (isolines, calculated using the LAMER code) and measured ^{137}Cs activity concentrations (dots) in surface waters of the NW Pacific Ocean is shown in Fig. 7. The measured ^{137}Cs concentrations in surface seawater of the NW Pacific Ocean (Aoyama et al., 2013) are within the range of simulated ones. Background ^{137}Cs concentration of $1 \, \text{mBq L}^{-1}$, which is due to global fallout, was deducted from the measured ^{137}Cs concentrations. The sampling dates were from January to March 2012; the model output was set on 1 March 2012. The highest ^{137}Cs concentration ($25.5 \, \text{mBq L}^{-1}$) was found at 46.6° N, 152° E on
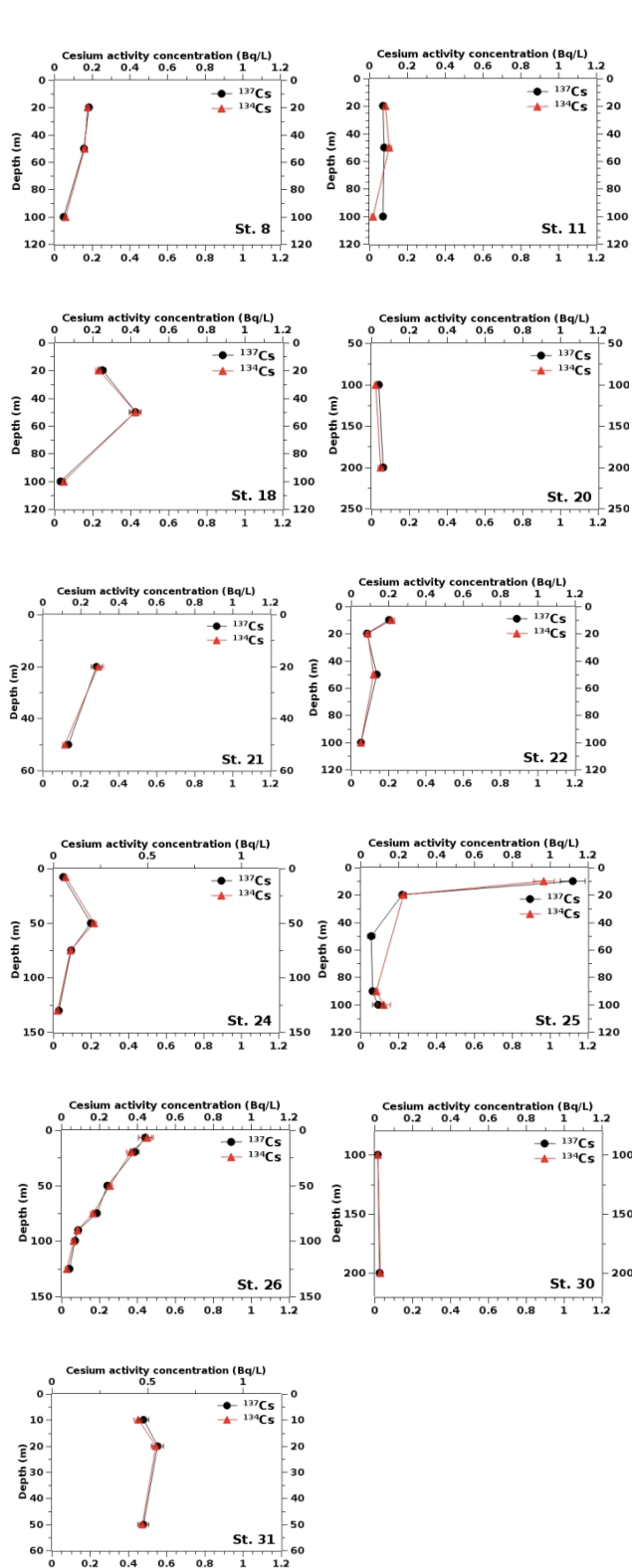
Fig. 6. ^{134}Cs and ^{137}Cs water profiles measured in the present work on samples collected during the *KOK* expedition. Positions of sampling stations are shown in Fig. 5.
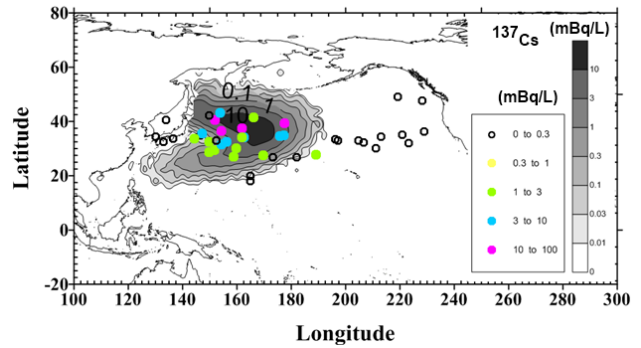

Fig. 7. A comparison of predicted (isolines, calculated using the LAMER code) and measured ^{137}Cs activity concentrations (dots) in surface water of the NW Pacific Ocean ($1 \, \mathrm{mBq \, L^{-1}}$ from the global fallout background was deducted from the measured ^{137}Cs concentration). The date for the model output was set as 1 March 2012. The experimental ^{137}Cs data from Aoyama et al. (2013) were from January to March 2012.

24 January 2012; the other three high concentrations (Fig. 7) were between 10 and 15 $\mathrm{mBq \, L^{-1}}$. The LAMER code predictions were for all these stations below 16 $\mathrm{mBq \, L^{-1}}$.

The development of the Fukushima-derived ^{137}Cs concentration in the water column of the NW Pacific Ocean with time, and its comparison with global fallout, is presented in Fig. 8a. It can be seen that the maximum predicted ^{137}Cs concentration due to the Fukushima accident (about 25 $\mathrm{mBq \, L^{-1}}$) should be observed in the NW Pacific Ocean in 2012, which will be comparable with the levels estimated for the middle 1960s from global fallout (Nakano and Povinec, 2003a; Inomata et al., 2009). However, after ten years this concentration will not be distinguishable from global fallout.

The chronological change of the maximum ^{137}Cs concentrations at the surface (0–100 m) and at the 200–300 m water depth (at 38° N, 164° E) presented in Fig. 8b indicates that the surface concentration will gradually decrease, and the ^{137}Cs concentrations at 200–300 m water depth will reach the surface levels within a few years. Presently observed ^{137}Cs levels in the water column along the 144° E line offshore Fukushima (Aoyama et al., 2013) are within the range predicted by the LAMER simulations.

A better comparison of both surface and water column data with simulations will be possible when more data on the distribution of ^{137}Cs in the water column of the North Pacific Ocean are available. Such a comparison will help to test the hypotheses on the ^{137}Cs source terms in the NW Pacific Ocean, which will also improve the simulation results, as more precise input data will be available for the oceanic circulation models.
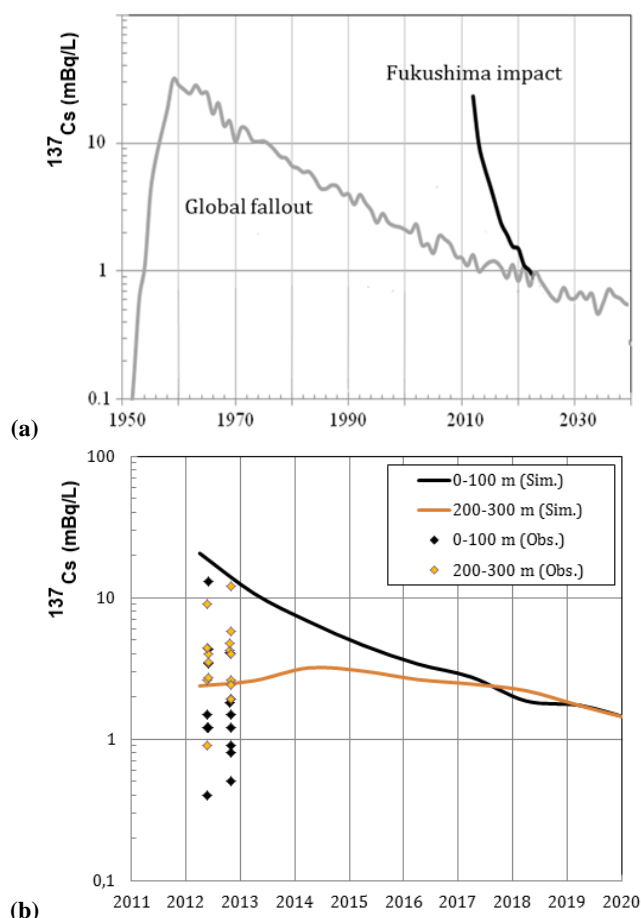
Fig. 8. (a) A comparison of simulated ^{137}Cs activity concentrations of Fukushima origin in surface seawater in the NW Pacific Ocean (at 38° N, 164° E) with global fallout data (after Nakano and Povinec, 2003a) (top). **(b)** A comparison of simulated and observed ^{137}Cs levels in surface water (0–100 m) and at 200–300 m water depth with time (bottom). The seawater samples were collected along the 144° E line offshore Fukushima (Aoyama et al., 2013). The global fallout background (1 mBq L^{-1}) has already been deducted from the experimental data.

5.3 ^{129}I in seawater of the NW Pacific after the Fukushima accident

The ^{129}I seawater profiles offshore Fukushima presented in Fig. 3 show that the highest surface levels were obtained at St. 25 (up to 62×10^7 atoms L^{-1}), which is situated about 40 km from the Fukushima NPP. The other three stations (St. 8, 11, and 20) show the ^{129}I concentrations of $(14–16) \times 10^7$ atoms L^{-1} at 20 m water depth, slightly lower than that at St. 25 (20×10^7 atoms L^{-1}). It can be also seen that the ^{129}I concentrations were decreasing with depth at all stations. The lowest ^{129}I concentrations (0.77×10^7 atoms L^{-1}) were observed in 400 m water depth at St. 11, which is about 260 km off Japan, and at St. 8 (1.9×10^7 atoms L^{-1}), which is the most distant location (530 km off Japan). The

distribution of ^{129}I / ^{127}I atom ratios follows the same trend as the ^{129}I concentrations. The highest ^{129}I / ^{127}I atom ratio (22×10^{-10}) was observed at 10 m depth at St. 25, while the lowest one (0.26×10^{-10}) in 400 m at St. 11. For a better comparison with other radionuclides, the observed range of ^{129}I levels in the unit of activity concentration was 0.01–0.8 µBq L^{-1}, which were comparable with those observed by Tumey et al. (2013), who analyzed a different set of samples from the same cruise. Recently, Suzuki et al. (2013) published ^{129}I data for seawater samples collected offshore Fukushima during April–October 2011, which were 8–70 times higher than the pre-Fukushima level.

Comparing the ^{129}I results presented in Fig. 3 with published data (Povinec et al., 2010; Suzuki et al., 2010, 2013), it can be concluded that St. 25 and 20 have been heavily influenced by the Fukushima accident, as the observed ^{129}I levels were higher by a factor of about 50 and 15, respectively. However, St. 11 and 8, which are the most distant stations from Japan, were also impacted by the Fukushima accident as their surface ^{129}I concentrations were by about a factor of 10 higher than the pre-Fukushima levels. Station 25 (measured down to 125 m) and St. 20 (down to 400 m) clearly indicate vertical transport of ^{129}I in the water column, which can also be seen at St. 8, as at 400 m water depth the ^{129}I level is by about a factor of three higher than a value expected from global fallout.

The ^{129}I / ^{137}Cs activity ratios at four stations varied by $(0.5–5) \times 10^{-6}$ indicating that all stations at depths up to 200 m were influenced by the Fukushima accident. A similar range in the ^{129}I / ^{137}Cs activity ratios observed in other set of samples collected during the *KOK* cruise was reported by Tumey et al. (2013). A ^{131}I / ^{137}Cs activity ratio of about 17.8 was measured in the water discharged directly into the sea from the Fukushima NPP. From the estimated atmospheric releases of ^{131}I (157 PBq) and ^{137}Cs (14 PBq), we get a value of 11.2 for the ^{131}I / ^{137}Cs activity ratio, which is not far from a median of 15 estimated by Hirose (2012) in precipitation over the Japan. As ^{131}I and ^{129}I have similar behavior in the environment, the amount of ^{129}I directly discharged into the sea can be estimated from the measured ^{131}I / ^{137}Cs ratio, and the estimated amount of ^{137}Cs discharged into the sea. If we take as a lower limit for direct liquid discharges of ^{137}Cs into the sea a value of 4 PBq (obtained by coastal modeling), the amount of ^{131}I directly discharged into the sea will be then 62 PBq. Based on these data, and the measured ^{129}I / ^{131}I ratio for released radioiodine from the Fukushima NPP, the amount of ^{129}I directly discharged into the sea was estimated to be 2.4 GBq (Hou et al., 2013). The middle values of ^{137}Cs source terms estimated by Rypina et al. (2013) and Charette et al. (2013) are around 12 PBq. If we use this value as a possible ^{137}Cs source term, then the amount of ^{129}I directly discharged into the sea could be around 7 GBq.

Morino et al. (2011) estimated that about 120 PBq of ^{131}I was deposited over the Pacific Ocean, of which 29 PBq was deposited in the area of 34–41° N and 137–145° E. Using

these values and the measured ^{129}I / ^{131}I ratio, we may estimate that about 4.6 GBq of ^{129}I released into the atmosphere was deposited in the Pacific Ocean, and 1.1 GBq in the area 34–41° N and 137–145° E.

The Chernobyl accident released into the atmosphere about 13 GBq of ^{129}I (IAEA, 2003), which had, however, a negligible impact on the marine environment. The European reprocessing plants at La Hague (France) and Sellafield (UK) discharged together much higher ^{129}I amounts into the sea (35 PBq) and to the atmosphere (3 PBq) (Hou et al., 2009b). As noted earlier, the liquid releases primarily affected the European seas and the Arctic Ocean. However, the ^{129}I released into the atmosphere was transported over the globe, and partially was also deposited in the Pacific Ocean.

5.4 Tritium in seawater of the NW Pacific after the Fukushima accident

The tritium seawater profiles (Fig. 4) generally follow the cesium and ^{129}I water profiles, although surprisingly the ^{3}H levels (in the range of 0.4–1.3 TU, equivalent to 0.05–0.15 Bq L^{-1}) are only by about a factor of 3 above the global fallout background. The lower ^{3}H signal in the seawater offshore Fukushima should be attributed to less release of ^{3}H from the Fukushima Dai-ichi NPP, due to much lower fission yield of ^{3}H compared to ^{137}Cs and ^{129}I, as well as due to the fact that the boiling water reactors produce much less tritium than the pressurized water reactors. All stations (except St. 11, sampling depth of 100 m) were affected by the Fukushima tritium. From the measured ^{3}H / ^{129}I and ^{3}H / ^{137}Cs activity ratios in the water column, and the previously estimated ^{129}I and ^{137}Cs releases in the sea, we may estimate the total ^{3}H activity released and deposited over the NW Pacific Ocean to be in the range 0.1–0.5 PBq.

The estimations of ^{129}I and ^{3}H source terms in the western NW Pacific Ocean presented above depend strongly on the estimations of direct liquid releases and atmospheric depositions of ^{137}Cs on the studied sea areas. Therefore, with improving our knowledge of ^{137}Cs source terms, the estimations of ^{129}I and ^{3}H source terms will also improve.

6 Conclusions

The contribution of ^{137}Cs, ^{129}I and ^{3}H released from the damaged Fukushima NPP into the sea has been remarkable, as it has considerably influenced their concentrations in surface seawater as well as in the water column of the NW Pacific Ocean. The main conclusions obtained in this work may be summarized as follows:

1. The ^{137}Cs, ^{129}I and ^{3}H levels in surface seawater observed offshore Fukushima (approximately from 30 km to 600 km from the coast) varied between 0.002–3.5 Bq L^{-1}, 0.01–0.8 µBq L^{-1} and 0.05–0.15 Bq L^{-1}, respectively. At the sampling site about 40 km from the coast, where all three radionuclides were analyzed, the Fukushima impact represents an increase above the global fallout background by factors of about 1000, 50 and 3, respectively. The water column data indicate that the transport of Fukushima-derived radionuclides has already reached depths of around 300 m.

2. The observed ^{137}Cs levels in surface waters and in the water column of the NW Pacific Ocean were within the range of predictions obtained from the ocean general circulation model. A better comparison will be possible, however, when more data on the distribution of ^{137}Cs in the water column of the North Pacific Ocean are available.

3. The ^{137}Cs inventory due to the Fukushima accident in the water column of the investigated NW Pacific Ocean (the area from 34 to 37° N and from 142 to 147° E) was estimated to be 2.2 ± 0.3 PBq, what can be regarded as a lower limit of the direct liquid discharges into the sea as the seawater sampling was carried out only in the area 34–37° N and 142–147° E.

4. The amount of ^{129}I directly discharged as liquid wastes into the sea was estimated to be in the range 2.4–7 GBq, and about 4.6 GBq of ^{129}I was deposited from the atmosphere into the NW Pacific Ocean. The total ^{3}H activity released and deposited over the NW Pacific Ocean was estimated to be in the range of 0.1–0.5 PBq. The evaluations of ^{129}I and ^{3}H source terms depend strongly on the estimations of direct liquid releases and atmospheric depositions of ^{137}Cs into the studied sea areas. Therefore, with improving our knowledge of ^{137}Cs source terms, the estimations of ^{129}I and ^{3}H source terms will also improve.

The radionuclides discussed in the present work will be useful tracers for studying horizontal and vertical transport of water in the North Pacific Ocean during the next decades.

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