

Cesium, iodine and tritium in NW Pacific waters

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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 seawater samples collected during the international expedition in June 2011 were analyzed for ^{134}Cs , ^{137}Cs , ^{129}I and ^3H . The ^{137}Cs , ^{129}I and ^3H levels in surface seawater offshore Fukushima varied between $0.002\text{--}3.5\text{ Bq L}^{-1}$, $0.01\text{--}0.8\text{ }\mu\text{Bq L}^{-1}$, and $0.05\text{--}0.15\text{ Bq L}^{-1}$, 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 represent an increase above the global fallout background by factors of about 1000, 30 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 in reasonable agreement with predictions obtained from the Ocean General Circulation Model, which indicates that the radionuclides have been transported from the Fukushima coast eastward. The ^{137}Cs inventory in the water column (the area from 34 to 37° N , and from 142 to 147° E) due to the Fukushima accident is estimated to be about 2.2 PBq . The amount of ^{129}I and ^3H released and deposited on the NW Pacific Ocean after the Fukushima accident was estimated to be about 7 GBq and 0.1 PBq , respectively. 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 in the NW Pacific Ocean.

1 Introduction

The largest amount of radionuclides ($\sim 950\text{ PBq}$) released to the atmosphere up to now, representing the main source of anthropogenic radionuclides in the World Ocean, has been global fallout resulting from atmospheric tests of nuclear weapons carried

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out mainly in fifties and early sixties (UNSCEAR, 2008). Recently, large quantities of radioactive materials were released to the atmosphere and coastal waters 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 11th 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, similarly as the Chernobyl accident, which happened in 1986 in the former Soviet Union (presently 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 total 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 $^{134}\text{Cs}/^{137}\text{Cs}$ activity ratio was close to one (Masson et al., 2011).

Apart from the contamination of Japan (and the Japan Sea, 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 into the Asia. At the beginning of April, the global atmosphere has been 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 % was deposited over the 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, large amounts of liquid radioactive wastes were

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5 directly discharged from the Fukushima Dai-ichi NPP into the ocean. 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 to the sea, which widely contaminated coastal waters off the Fukushima NPP, as reported 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 from 3.5 ~ 4 PBq (Tsumune et al., 2012; Kawamura et al., 2011) up to 27 ± 15 PBq (Bailly du Bois et al., 2012). As the cooling water directly interacted with ruptured nuclear fuel rods, it has been estimated that 0.1 ~ 1 PBq of ^{90}Sr has also been released to the ocean (Povinec et al., 2012a).

15 The direct discharge of contaminated water to 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 to the sea on 30th March (47 kBq L^{-1}) and on 6th April (68 kBq L^{-1}) (TEPCO, 2011). Several papers have already discussed ^{134}Cs and ^{137}Cs concentrations in surface waters of the NW Pacific Ocean. In the open ocean 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).

20 The aim of the present study has been 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.

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2 Radionuclides of interest and their pre-Fukushima levels in the 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) because of its shorter half-life can clearly identify cesium of Fukushima origin, as there is no remaining contribution from global fallout and the 1986 Chernobyl accident. Anthropogenic tritium and ^{129}I have been recognized as ideal short-term (^3H 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; Povinec et al., 2010, 2011).

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 the atmospheric nuclear weapons testing (Table 1). The main input into this part of the ocean occurred in early sixties due to wet and dry deposition of ^{137}Cs , after large-scale atmospheric nuclear weapons tests 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

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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).

5 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).

10 After its release to the atmosphere, the ^{137}Cs is rapidly associated with aerosols, which represent a major reservoir of pollutants in the atmosphere (Lujaniené 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., 2011, 2013).

15 As ^{137}Cs is a conservative oceanographic tracer, it is 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). Its removal 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; Lujaniené et al., 2006).

20 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 IAEA'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/MARIS.

25 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,

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respectively, where the water masses of the Kuroshio and Oyashio Currents mix) are shown in Fig. 1. The surface ^{137}Cs concentrations 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 and 10.6 ± 0.5 , respectively. Higher ^{137}Cs concentrations in surface water occurred in the 1980s, which could be due to liquid radioactive discharges from the nuclear fuel reprocessing plant at Tokai (Mizutani et al., 2009). By combining all ^{137}Cs data, the mean effective half-life of ^{137}Cs in North Pacific surface waters was determined 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 $30\text{--}35^\circ\text{N}$ and $150\text{--}170^\circ\text{E}$, 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 $< 0.1\text{ mBq L}^{-1}$ at water depths below 1000 m (Povinec et al., 2003a), corresponding to concentrations of 1.2, 1.4, 0.95, and $< 0.05\text{ mBq L}^{-1}$ in 2011, based on an effective half-life of 13 yr of ^{137}Cs in Pacific Ocean. On the basis of the data stored in the MARIS database (www.iaea.org/MARIS), the average ^{137}Cs concentrations in NW and NE Pacific surface waters for the year 2010 are estimated to be $1.0 \pm 0.1\text{ mBq L}^{-1}$ and $0.9 \pm 0.2\text{ mBq L}^{-1}$, 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; Povinec et al., 2010, 2011). 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 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 interaction of atmosphere and seawater, as well as biogeochemical cycles of stable

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iodine through chemical speciation analysis of ^{129}I and ^{127}I (Hou et al., 2001, 2009a, 2012).

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 ($52^{\circ}30' \text{N}$, $159^{\circ}10' \text{E}$) showed ^{129}I levels up to two orders of magnitude higher than the open ocean values, which were probably caused by leakages from the dumping site (similarly to that observed in the Kara Sea (Arctic Ocean, Povinec et al., 2000). The IAEA'97 results in the NW Pacific Ocean showed surface ^{129}I values around $1.5 \times 10^7 \text{ atoms L}^{-1}$ (Fig. 3), subsurface maxima at around $2.1 \times 10^7 \text{ atoms L}^{-1}$, a decrease down to 1.5×10^7 at 500 m, and a fast decrease $\leq 0.8 \times 10^7 \text{ atoms L}^{-1}$ at depths below 1000 m (Povinec et al., 2010). Higher ^{129}I levels (up to $3 \times 10^7 \text{ atoms L}^{-1}$ at 200 m water depth) were observed close to the Enewetak and Bikini atolls (about 11°N , 162°E). Stations at both atolls showed concentration minima at around 500 m, and secondary maxima ($1.8 \times 10^7 \text{ atoms L}^{-1}$) at about 1000 m. 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 fifties. Suzuki et al. (2010) reported ^{129}I concentrations in the NW Pacific waters decreasing from the surface ($2 \times 10^7 \text{ atoms L}^{-1}$) to 100 m water depth ($0.7 \times 10^7 \text{ atoms L}^{-1}$), and then $< 0.2 \times 10^7 \text{ atoms L}^{-1}$ at water depths below 100 m. Corresponding $^{129}\text{I}/^{127}\text{I}$ ratios were 0.7×10^{-10} , 0.25×10^{-10} , and $< 0.07 \times 10^{-10}$, respectively.

The previous investigations thus showed that the expected pre-Fukushima ^{129}I concentration in surface NW Pacific waters should be $(2 \pm 0.2) \times 10^7 \text{ atoms L}^{-1}$. 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.

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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 T₂O), and has a suitable half-life (12.32 yr). It is produced either 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. 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 re-processing facilities (Table 1). 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 ¹³⁷Cs). The penetration of bomb tritium from surface waters into deeper layers of the ocean was used to study pathways and time scales 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; 2005, 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 ³H in the World Ocean (www.eWOCE.org).

The IAEA'97 results in the NW Pacific Ocean (Fig. 4) showed surface ³H values around 1 TU (tritium data are usually expressed in Tritium Units: 1 TU is ³H/¹H atomic ratio of 1×10^{-18} , corresponding to activity concentration of 0.118 Bq L⁻¹ of water), sub-surface maxima (at 200–300 m) around 1.3 TU, a decrease down to 0.7 TU at 500 m, and a fast decrease down to 0.1 TU at depths below 1000 m (Povinec et al., 2010). On the basis of the data stored in the MARIS database, the pre-Fukushima ³H concentration in surface NW Pacific waters can be estimated to be (0.6 ± 0.1) TU. Although tritium is not an important radionuclide from the radioecological point of view, as it has been used as an oceanographic tracer (also in climate change studies), we need to know

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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

5 Estimated radionuclide releases and their inventories in 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
10 88 PBq is in the Pacific Ocean, and around 17 PBq in its latitudinal belt of 25–40° N.

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
15 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).
20 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 Sea). For comparison, about 42 PBq of ^{137}Cs has been released to the marine environment

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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 Sea (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 to 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 total ^{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 March 24, which have been then distributed globally (Masson et al., 2011), and due to wet and dry deposition contaminated the terrestrial and marine environments. When comparing the Fukushima atmospheric radionuclide releases with the Chernobyl ones, we see that they were lower for ^{131}I and ^{137}Cs by about a factor of 12 and 7, respectively (Table 1).

3.3.2 Radionuclide releases to the ocean

Radionuclides released directly to 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. The total amount of directly released ^{137}Cs to the sea was estimated by the Japanese Government (JG, 2011) to be 0.94 PBq. Predictions made using a global ocean circulation models and the available ^{137}Cs monitoring data gave 3.5 ± 0.7 PBq (Tsumune et al., 2012), and 4 PBq (Kawamura et al., 2011). Bailly du Bois

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et al. (2012) using a dispersion model supported with ^{137}Cs results measured in surface water off Fukushima estimated the ^{137}Cs releases to be in the range 10–34 PBq. Dietze and Kriest (2012) using numerical simulations indicated, however, that the model of Bailly du Bois et al. (2012) probably overestimated the direct ^{137}Cs releases to coastal waters, as their estimate was only 0.94–3.5 PBq. Recently Rypina et al. (2013) using the ^{137}Cs data of Buesseler et al. (2012) and numerical simulations estimated the strength of the oceanic source to be in the range 9–18 PBq. We see that there are still large discrepancies in the estimated direct ^{137}Cs releases to the ocean, therefore, more work needs to be done. The ^{90}Sr releases were estimated to be in the interval 0.1–1 PBq (Povinec et al., 2012a).

The estimation of the atmospheric deposition of ^{137}Cs over the Pacific Ocean is the most 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). Dietze and Kriest (2012) estimated the accumulated atmospheric deposition of ^{137}Cs in the range of 50–200 kBq m⁻². The $^{131}\text{I}/^{137}\text{Cs}$ activity ratio in surface seawater indicates that most of the radiocesium observed in coastal waters has been the result of a direct discharge to the ocean, rather than resulting from atmospheric deposition (Tsumune et al., 2012). On the basis of modeling exercises of Kawamura et al. (2011) we expect that the cumulative deposition of ^{137}Cs over the 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 in the oceanic dispersion model for prediction of ^{137}Cs levels in the open NW Pacific Ocean, which is described later.

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 to 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 $^7\text{Li}(n, \alpha)^3\text{H}$ reaction (Hou, 2005).

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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'2011) 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 analysis, and 4 depth profiles were analyzed for ^{129}I and ^3H . 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 15 L (surface) volumes.

4.2 Analytical methods

All chemical reagents used during sample preparations were of analytical grade. All solutions were prepared using deionised 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-418 (Pham et al., 2010) were analyzed for ^{137}Cs , ^{129}I and ^3H , and the NIST standard reference material (NIST-SRM-4949c) was used for ^{129}I analysis.

4.2.1 Tritium analysis

Pre-screening of ^3H levels in seawater samples (to avoid possible contamination) was carried out by direct ^3H counting (after triple distillation) in water-liquid scintillator cocktails using Packard Liquid Scintillation Spectrometer. Tritium was then precisely

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analyzed in seawater samples using the ^3He 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 ^3He 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 ^3He is then measured, from which ^3H 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 AMS measurements has already been described (Hou et al., 2012), therefore it will be only briefly mentioned here. 200–500 mL of seawater samples was spiked with $^{125}\text{I}^-$ and transferred to a separation funnel. After addition of 0.5 mg ^{127}I carrier (Woodward Inc., with a $^{129}\text{I}/^{127}\text{I}$ ratio lower than 1×10^{-13}), 3 mL of 1 M NaHSO_3 solution and 3M HNO_3 were added to pH 1–2 to convert all inorganic iodine to iodide. Iodine was then extracted into CHCl_3 after addition of NaNO_2 to oxidize iodide to I_2 . Iodine in CHCl_3 phase was then back-extracted to the water phase using 10 mL of 0.1 mM NaHSO_3 solution. 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, 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.

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The $^{129}\text{I}/^{127}\text{I}$ 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 $^{129}\text{I}/^{127}\text{I}$ background ratio was $\sim 3 \times 10^{-14}$. The blanks prepared using the same procedure as the samples gave for the $^{129}\text{I}/^{127}\text{I}$ ratios values below 1×10^{-13} , which was by about three orders of magnitude lower than measured $^{129}\text{I}/^{127}\text{I}$ 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. High activity seawater samples were nondestructively counted directly on HPGe spectrometers. In low activity samples radiocesium was pre-concentrated by adsorption onto ammonium molybdophosphate (AMP) at pH2. The AMP suspension was allowed to settle and the supernatant solution was decanted or siphoned away. The AMP solid particle in the remaining suspension was separated by centrifuge. The separated AMP solid was washed with water. The Cs recovery was obtained by determination of stable ^{133}Cs concentration in water before and after AMP-absorption by ICPMS. The AMP sample was analyzed for the presence of ^{134}Cs and ^{137}Cs in the CAVE underground facility of IAEA-EL using large efficiency HPGe detectors (Povinec et al., 2005b).

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 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

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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 the results from the KOK'2011 cruise (Bueseler et al., 2012), and the results presented in this paper shows that the measured ^{137}Cs concentrations in surface waters ranged from 1.8 mBq L^{-1} to 3500 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 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 Bq L^{-1} . The elevated ^{137}Cs levels covered an area of around $150\,000 \text{ km}^2$ (south of 38° N and west of 147° E). Even at distances around 600 km off Fukushima, ^{137}Cs activity concentrations of around

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0.3 Bq L⁻¹ were found, i.e. by about a factor of 300 above the global fallout background of 1 m Bq L⁻¹.

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

The ¹³⁴Cs and ¹³⁷Cs water profiles measured in this work (Fig. 6) indicate that mostly surface water maxima were observed, although at some stations (e.g. St. 29, 26, 19, 40) subsurface maxima (at 20–50 m) were observed as well. The Fukushima-derived ¹³⁴Cs and ¹³⁷Cs penetrated up to June 2011 to at least 200 m water depth (St. 4 and 22). This figure also confirms that the ¹³⁴Cs/¹³⁷Cs activity ratio in the analyzed seawater samples was close to 1, clearly indicating that the source of these radionuclides was the Fukushima accident.

The ¹³⁷Cs water profiles at St. 31, 22, 14 and 11, which are compared with the pre-Fukushima profiles measured in 1997 during the IAEA'97 expedition in the NW Pacific Ocean (Fig. 2), show that the Fukushima signal is well visible in all four stations, especially at St. 31, where surface ¹³⁷Cs levels exceeded the global fallout values by about three orders of magnitude.

The ¹³⁷Cs inventory in the water column of the NW Pacific due to the Fukushima accident, estimated using the Buessler et al. (2012) data and those presented here, is 2.2 ± 0.3 PBq, confirming an essential contribution to its total ¹³⁷Cs inventory in the NW Pacific Ocean (Table 1).

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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, the ^{137}Cs source-term offshore Fukushima used in our simulations was 9 PBq, represented by 4 PBq from direct liquid releases, and 5 PBq from atmospheric deposition.

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). It can be seen from Fig. 7 that a reasonable compromise has been found between the measured and the simulated ^{137}Cs concentrations in surface seawater of the NW Pacific Ocean.

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 20 mBq L^{-1}) should be observed in the NW Pacific Ocean (at 38° N , 164° E) in 2012, which will be comparable with the levels in the Oyashio region 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 concentrations at 200–300 m will be increasing from zero up to the surface level in about 5 yr. 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.

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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. 31 (up to 62×10^7 atoms L^{-1}), which is situated about 40 km from the Fukushima NPP. The other three stations (St. 11, 14, and 22) show the ^{129}I concentrations of $(14\text{--}16) \times 10^7$ atoms L^{-1} at 20 m water depth, slightly lower than that at St. 31 (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) were observed in 400 m water depths at St. 14, which is about 260 km off Japan, and at St. 11 (1.9×10^7 atoms L^{-1}), which is the most distant location (530 km off Japan). The distribution of $^{129}\text{I}/^{127}\text{I}$ ratio follows the same trend as the ^{129}I concentrations. The highest $^{129}\text{I}/^{127}\text{I}$ atom ratio (22×10^{-10}) was observed at 10 m depth at St. 31, while the lowest one (0.26×10^{-10}) in 400 m at St. 14. For a better comparison with other radionuclides, the observed range of ^{129}I levels in the unit of activity concentration was $0.01\text{--}0.8 \mu\text{Bq L}^{-1}$.

Comparing the ^{129}I results presented in Fig. 3 with published data (Povinec et al., 2010; Suzuki et al., 2010), it can be concluded that St. 31 and 22 have been heavily influenced by the Fukushima accident, as the observed ^{129}I levels were higher by a factor of 30 and 10, respectively. However, St. 14 and 11, which are the most distant stations from Japan were also impacted by the Fukushima accident as their surface ^{129}I concentrations were almost by about a factor of 10 higher than the pre-Fukushima levels. Station 31 (measured down to 125 m) and St. 22 (down to 400 m) clearly indicate vertical transport of ^{129}I in the water column, which can also be seen at St. 11, as at 400 m water depth the ^{129}I level is by about a factor of two higher than expected.

The $^{129}\text{I}/^{137}\text{Cs}$ activity ratios at four stations varied between $(0.5 \sim 5) \times 10^{-6}$ indicating that all stations at depths up to 200 m were influenced by the Fukushima accident. A $^{131}\text{I}/^{137}\text{Cs}$ activity ratio of about 17.8 was measured in the water discharged directly to the sea from the Fukushima NPP. From the estimated atmospheric releases of ^{131}I ($\sim 157 \text{PBq}$) and ^{137}Cs ($\sim 14 \text{PBq}$), we get a value of 11.2 for the $^{131}\text{I}/^{137}\text{Cs}$ activity

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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 to the sea can be estimated from the measured $^{131}\text{I}/^{137}\text{Cs}$ ratio, and the estimated amount of ^{137}Cs discharged to the sea. Thus if we take as an example 4 PBq for direct liquid discharge of ^{137}Cs to the sea, the amount of ^{131}I directly discharged to the sea will be 62 PBq. Based on these data, and the measured $^{129}\text{I}/^{131}\text{I}$ ratio for released radioiodine from the Fukushima NPP, the amount of ^{129}I directly discharged to the sea can be estimated to be 2.4 GBq (Hou et al., 2013). 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 34–41° N and 137–145° E. Using these values and the measured $^{129}\text{I}/^{131}\text{I}$ ratio, we may estimate that about 4.6 GBq of ^{129}I released to 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 to the atmosphere about 13 GBq of ^{129}I (IAEA, 2003), that is almost by a factor of two higher than the total release of about 7 GBq of ^{129}I from the Fukushima accident. The ^{129}I released from the Chernobyl accident 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 to 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 to the atmospheric was transported over the globe, and partially was also deposited on 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 ^3H 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 ^3H signal in the seawater offshore Fukushima should be attributed to less release of ^3H from the Fukushima Dai ichi NPP, due to much lower fission yield of ^3H compared

to ^{137}Cs and ^{129}I . All stations (except St. 14–15, sampling depth of 100 m) were affected by the Fukushima tritium. From the measured $^3\text{H}/^{129}\text{I}$ and $^3\text{H}/^{137}\text{Cs}$ activity ratios in the water column, and the previously estimated ^{129}I and ^{137}Cs releases to the sea we may estimate the total ^3H activity released and deposited over the NW Pacific Ocean to be of the order of 0.1 PBq.

6 Conclusions

The contribution of ^{137}Cs , ^{129}I and ^3H released from the damaged Fukushima NPP to 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 ^3H levels in surface seawater observed offshore Fukushima (approximately from 30 km to 600 km from the coast) varied between $0.002\text{--}3.5\text{ Bq L}^{-1}$, $0.01\text{--}0.8\ \mu\text{Bq L}^{-1}$ and $0.05\text{--}0.15\text{ Bq L}^{-1}$, respectively. At the sampling site about 40 km from the coast, where all three radionuclides were analyzed, the Fukushima impact represent an increase above the global fallout background by factors of about 1,000, 30 and 3, respectively. The water column data indicate that the transport of Fukushima-derived radionuclides has already reached a depth of 300 m.
2. The observed ^{137}Cs levels in surface waters and in the water column of the NW Pacific Ocean are in a reasonable agreement with predictions obtained from the Ocean General Circulation Model.
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) is estimated to be $2.2 \pm 0.3\text{ PBq}$. About 4.6 GBq of ^{129}I was released to the atmosphere and deposited in the NW Pacific Ocean, and about 2.4 GBq was directly

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discharged as liquid wastes to the sea offshore Fukushima. The total amount of ^3H released and deposited on the NW Pacific Ocean was estimated to be of the order of 0.1 PBq.

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

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

Nuclide	Half-life	Natural inventory ^a	Global fallout Atmosphere ^a	Global fallout inventory in the ocean ^b		Discharges from reprocessing facilities ^b		Global fallout Atmosphere ^a Ocean ^b		Chernobyl Atmosphere ^c Ocean ^b		Fukushima Atmosphere Ocean	
				Total inventory in the ocean	Inventory in 2010 in the ocean	Total inventory	Inventory in 2010	Atmosphere ^a	Ocean ^b	Atmosphere ^c	Ocean ^b	Atmosphere	Ocean
³ H	12.32 yr	2200		113 000	8000	410	45					0.1 ^p	
¹²⁹ I	15.7 × 10 ⁶ yr	0.6 × 10 ⁻³		0.3 × 10 ⁻³	0.3 × 10 ⁻³	0.04	0.04			0.013 × 10 ⁻³		0.0024 × 10 ^{-3g}	0.0046 × 10 ^{-3ah}
¹³¹ I	8.02 d	–			–	–	39	–			1760	153–160 ^{6,e}	62 ^f 120 ^g
¹³⁷ Cs	30.17 yr	–	950	600	170	40	26	950	600	85	16	13–15 ^{4,h} 23–50 ⁱ	3.5 ± 0.7 ^j 4 ^k 0.94–3.5 ^l 10–34 ^m 9–18 ⁿ 5 ^o

^a UNSCEAR, 2008 ^b IAEA, 2005, ^c IAEA, 2003, ^d TEPCO, 2011, ^e Chino et al., 2011, ^f Stohl et al., 2012,

^g Hou et al., 2013, ^h atmospheric deposition (this work), ⁱ Morino et al., 2011, ^j Tsumune et al., 2012,

^k Kawamura et al., 2012, ^l Dietze and Kriest, 2012, ^m Bailly do Bois et al., 2012, ⁿ Rypina et al., 2013,

^o atmospheric deposition (Kawamura et al., 2011), ^p liquid discharges with atmospheric deposition (this work).

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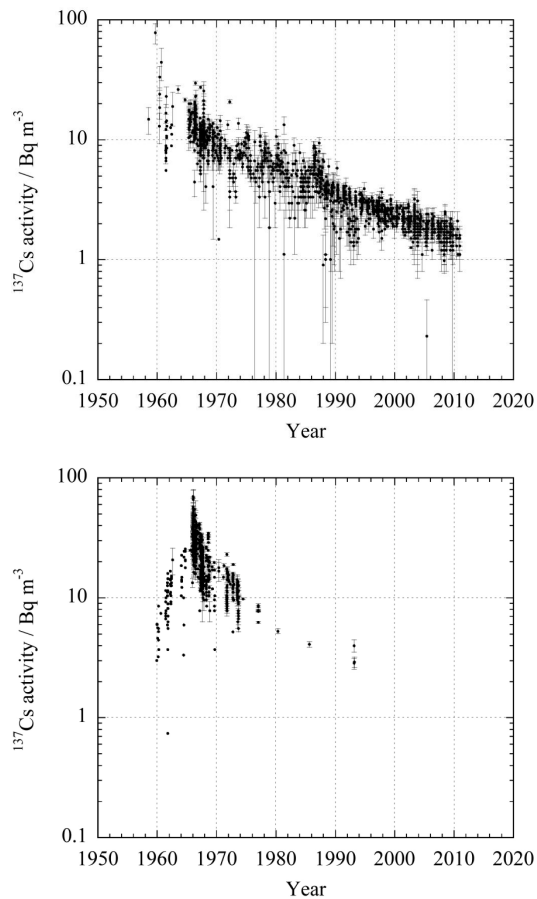


Fig. 1. Long-term changes of ^{137}Cs concentrations in surface seawater in two latitudinal belts at 25° – 40° N, divided at 180° E for the western (top) and the eastern (bottom) belt of the North Pacific Ocean.

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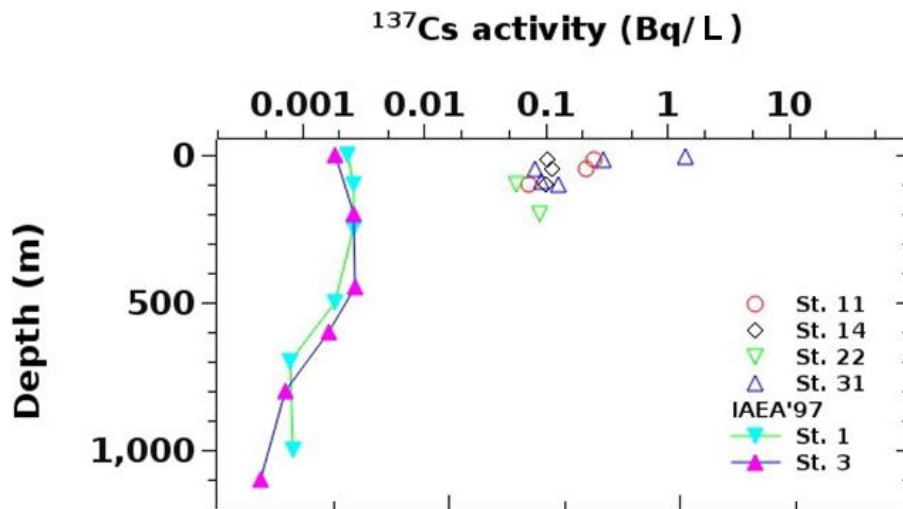


Fig. 2. A comparison of ^{137}Cs profiles as obtained from the IAEA'97 (global fallout impact) and the KOK'2011 expeditions (Fukushima impact) in the NW Pacific Ocean. IAEA'97 data were decay corrected to June 2011.

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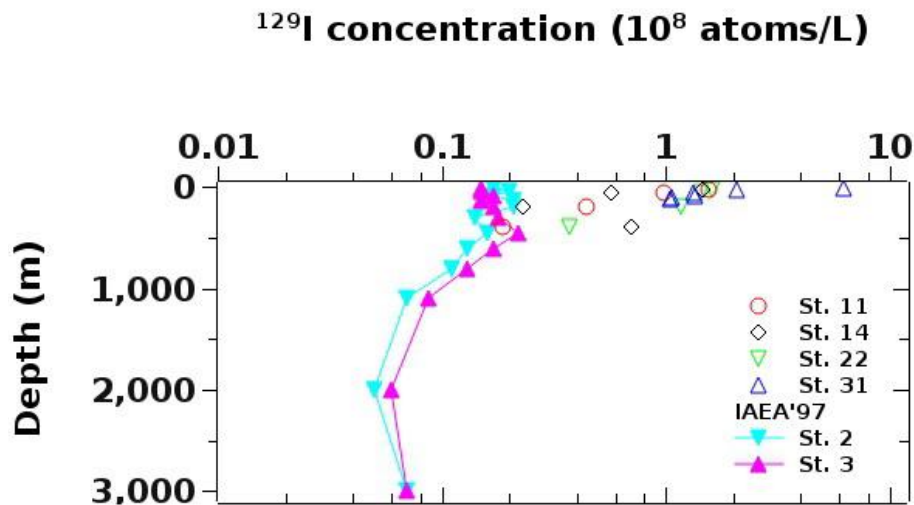


Fig. 3. A comparison of ^{129}I profiles as obtained from the IAEA'97 (global fallout impact) and the KOK'2011 expeditions (Fukushima impact) in the NW Pacific Ocean.

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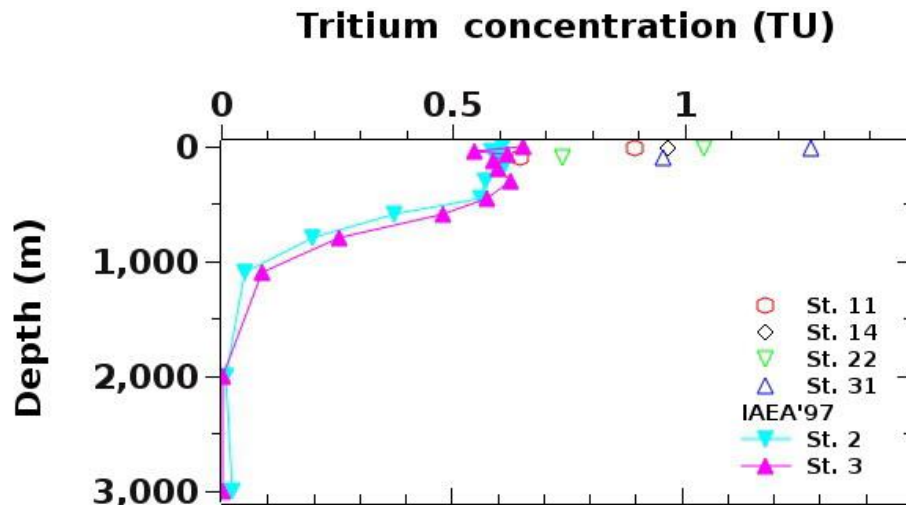


Fig. 4. A comparison of ^3H profiles as obtained from the IAEA'97 (global fallout impact) and the KOK'2011 expeditions (Fukushima impact) in the NW Pacific Ocean. IAEA'97 data were decay corrected to June 2011.

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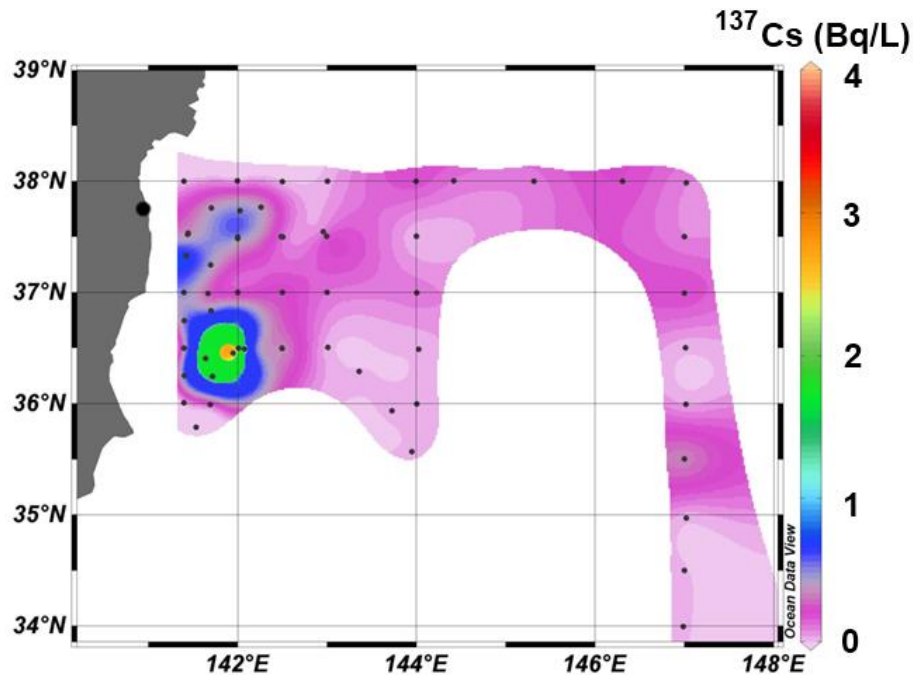


Fig. 5. Distribution of ^{137}Cs in surface waters offshore Fukushima. Data from Buesseler et al. (2012) and the present work (both from the KOK'2011 expedition).

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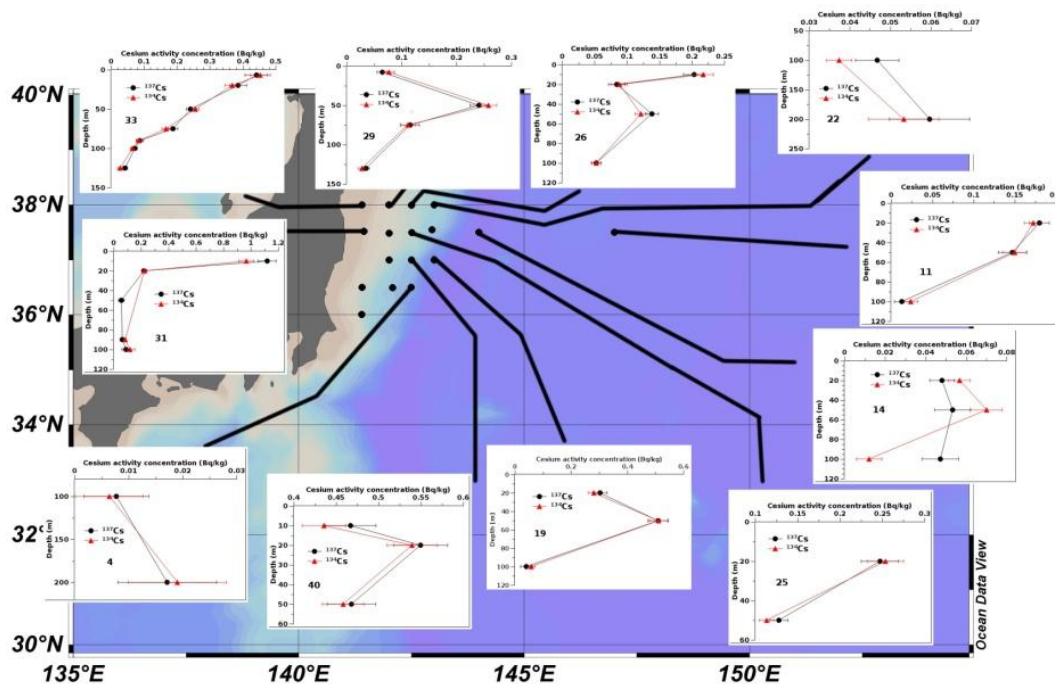


Fig. 6. ^{137}Cs water profiles as measured offshore Fukushima during the KOK'2011 expedition.

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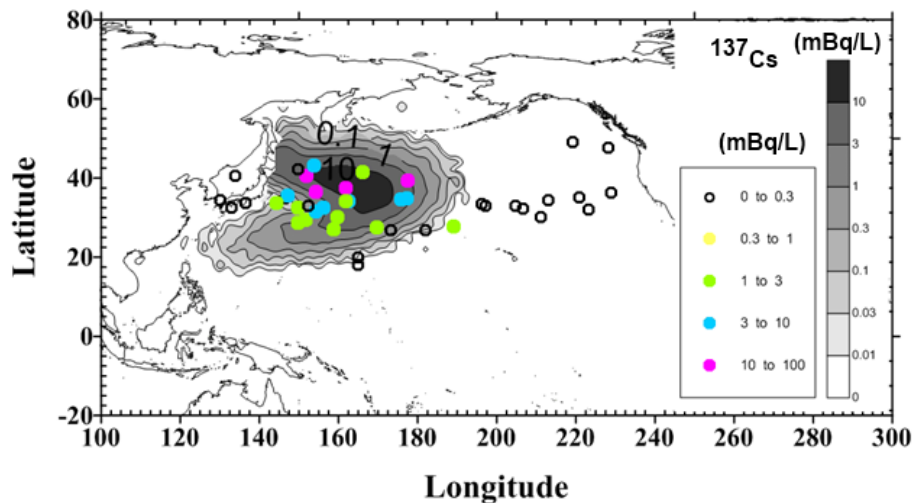


Fig. 7. A comparison of predicted (isolines, calculated using the LAMER code) and measured ^{137}Cs activity concentrations (dots) in surface water of the NW Pacific Ocean in March 2012 (^{137}Cs experimental data from Aoyama et al., 2013). In the case if ^{134}Cs was not detected, the ^{137}Cs concentration was set at zero. If ^{134}Cs was detected, 1 Bq m^{-3} from the global fallout background was deducted from the measured ^{137}Cs concentration.

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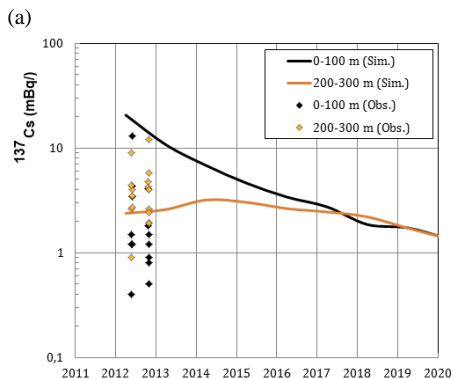
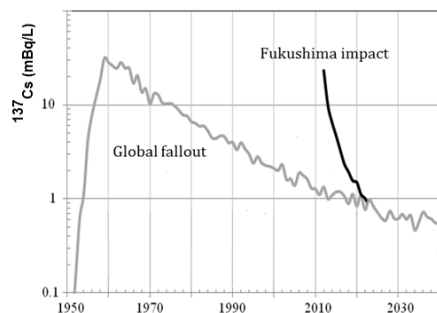
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(b)

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 , Oyashio region) 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 (1Bq m^{-3}) has already been deducted from the experimental data.