Impact of the Fukushima accident on tritium, radiocarbon and radiocesium levels in seawater of the western North Pacific Ocean: A comparison with pre-Fukushima situation

Pavel P. Povinec, Laval Liong Wee Kwong, Jakub Kaizer, M. Molnar, Hartmut Nies, L. Palcsu, Laszlo Papp, Mia Khanh Pham, P. Jean-Baptiste

P.P.Povinec, L.Liong Wee Kwong, J.Kaizer, M.Molnár, H.Nies, L.Palcsu, L.Papp, M.K.Pham, P.Jean-Baptiste

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ABSTRACT: Tritium, radiocarbon and radiocesium concentrations in water column samples in coastal waters offshore Fukushima and in the western North Pacific Ocean collected in 2011-2012 during the Ka'imikai-o-Kanaloa (KoK) cruise are compared with other published results. The highest levels in surface seawater were observed for ^{134}Cs and ^{137}Cs in seawater samples collected offshore Fukushima (up to 1.1 Bq L^{-1}), which represent an increase by about three orders of magnitude when compared with the pre-Fukushima concentration. Tritium levels were much lower (up to 0.15 Bq L^{-1}), representing an increase by about a factor of 6. The impact on the radiocarbon distribution was measurable, but the observed levels were only by about 9% above the global fallout background. The ^{137}Cs (and similarly ^{134}Cs) inventory in the water column of the investigated western North Pacific region was (2.7 ± 0.4) PBq, while for ^{3}H it was only (0.3 ± 0.2) PBq. Direct releases of highly contaminated water from the damaged Fukushima NPP, as well as dry and wet depositions of these radionuclides over the western North Pacific considerably changed their distribution patterns in seawater. Presently we can distinguish Fukushima labeled waters from global fallout background thanks to short-lived ^{134}Cs. However, in the long-term perspective when ^{134}Cs will decay, new distribution patterns of ^{3}H, ^{14}C and ^{137}Cs in the Pacific Ocean should be established for future oceanographic and climate change studies in the Pacific Ocean.

Keywords: Fukushima accident; radionuclides in seawater; KoK cruise; GEOSECS; WOCE; WOMARS

1. Introduction

Large quantities of radionuclides were released during the Fukushima Dai-chi Nuclear Power Plant (NPP) accident (starting on 11 March 2011) to the atmosphere (Chino et al., 2011; Hirose, 2012) and to coastal waters (Kawamura et al., 2011; Povinec et al., 2013a; Aoyama et al. 2015a, b). From a radiological point of view, ^{137}Cs has been considered to be the most important anthropogenic radionuclide in the environment because of its large releases, relatively long half-life, and its relative high bioavailability. Due to its accumulation in tissues, it has been relevant for delivering radiation doses to the public from the consumption of food (Aarkrog et al., 1997; Livingston and Povinec, 2000). Another radioecologically important radionuclide, which may be released in large quantities during nuclear accidents is ^{131}I, however, because of its short half-life (8.02 days), its contribution is dominant to terrestrial radiation doses during first days after the accident (Povinec et al., 2013a).

About 15—20 PBq of radiocesium (for ^{134}Cs and ^{137}Cs each) was released during the Fukushima accident to the atmosphere (Chino et al., 2011; Povinec et al., 2013a, b; Aoyama et al. 2015a), which occurred mainly between 12 and 16 March, with smaller contributions up to March 24. Most of the radionuclides released to the atmosphere were rapidly associated with aerosols, representing a major form of pollutants in the atmosphere, which have been then distributed globally (Masson et al., 2011; Povinec et al., 2013a, b). The radionuclides moved with the prevailing western winds mainly from Fukushima over the Pacific Ocean (Povinec et al., 2013c), with only a partial contamination of the Japanese land (Hirose, 2012) and the Japan Sea (Inoue et al., 2012).

After passing the Pacific Ocean, North America, the Atlantic Ocean, and Europe, the radioactive clouds went back across the Asian continent. At the beginning of April 2011, the northern hemisphere was labeled with Fukushima-derived radionuclides (Masson et al., 2011; Hernández-Ceballos et al., 2012; Povinec et al., 2013a, b). The aerosols with captured radionuclides were then deposited on the Earth's surface including the ocean by wet and dry deposition. About 80 % was deposited over the North Pacific Ocean, about 20 % over Japan, and less than 2% over the Atlantic and Europe (Morino et al., 2011; Stohl et al., 2012; Yoshida and Kanda, 2012).

We shall discuss in this paper radionuclide variations in the water column of the western North Pacific Ocean after the Fukushima accident. Distribution of several radionuclides dissolved in seawater (e.g., ^{3}H, ^{90}Sr, ^{129}I, ^{134}Cs, ^{137}Cs) has already been under investigations (e.g., Aoyama et al., 2012, 2013, 2015a; Buesseler et al., 2012; Hou et al. 2013; Povinec et

al., 2012, 2013a, d). We shall focus in this paper on Fukushima inpact on ^{3}H, ^{14}C and radiocesium levels in water column of the western North Pacific Ocean.

Cesium radioisotopes, namely ^{137}Cs (half-life $T_{1/2}$ = 30.17 yr) and ^{134}Cs ($T_{1/2}$ = 2.06 yr) have been the most frequently studied radionuclides in seawater after the Fukushima accident. Presence of ^{134}Cs in seawater clearly indicates its Fukushima origin as because of the short half-life ^{134}Cs from global fallout and the 1986 Chernobyl accident has already decayed. The nuclear accident at the Fukushima Dai-ichi NPP increased ^{137}Cs concentrations in coastal seawater in March 2011 up to 68 kBq L^{-1}, eight orders of magnitude above the global fallout background (Povinec et al., 2013a; Povinec and Hirose, 2015). This increase was due to direct liquid releases of ^{137}Cs contaminated fresh and later also seawater to coastal waters, which was estimated to be around 4 PBq (Kawamura et al., 2011; Tsumune et al., 2012, 2013; Aoyama et al., 2016a, b). The $^{131}I/^{137}Cs$ activity ratio in surface seawater offshore Fukushima indicated that most of the radiocesium observed in coastal waters was the result of a direct discharge to the ocean (Hou et al., 2013). Due to the transport of water masses from the Japanese coast to the open western North Pacific by the coastal currents (Nakano and Povinec, 2003, 2012; Tsumune et al., 2012, 2013), the high ^{137}Cs concentrations observed in 2011 in coastal seawater decreased rapidly with apparent half-life of about 1 year (Povinec and Hirose, 2015). The typical ^{137}Cs activity concentrations in coastal waters in 2011-2012 were from a few mBq L^{-1} to a few Bq L^{-1} (Aoyama et al., 2012, 2013; Buesseler et al., 2012; Honda et al., 2012; Povinec et al. 2013a, d; Kameník et al., 2013; Kumamoto et al., 2013a).

Only a little attention has been given till now to tritium (Povinec et al., 2013a, d; Matsumoto et al., 2013; Steinhauser, 2014), mainly because of the fact that liquid tritium releases when compared with radiocesium were much lower (0.1-0.5 PBq; Povinec et al., 2013d). Radiological impact due to tritium is low because of its short physical half-life (12.32 y), its short effective biological half-life in the human body (10 days, Kim et al., 2011), the low energy of emitted beta-decay electrons (18.6 keV) and its low production rates in nuclear reactors (Chudý and Povinec, 1982; Povinec et al., 2013a).

Similarly a radiological impact of radiocarbon on the marine environment and on humans has been expected to be low due to its low production rates in nuclear reactors (Chudý and Povinec, 1982). When compared with the Chernobyl accident, where due to ^{14}C production in the graphite moderator in the $^{13}C(n, \gamma)^{14}C$ reaction, and the graphite burning during the accident, the ^{14}C release to the atmosphere was estimated to be around 44 TBq (Povinec et al., 2013a). Elevated ^{14}C levels (up to 124 pMC (% Modern Carbon) or 281.6 Bq kg^{-1} of carbon) were measured in tree rings collected about 2.5 km from the Chernobyl NPP (Buzinny et al.

1998). However, the ^{14}C effect was not measurable in Central Europe (Povinec et al., 1988). There are no data available on radiocarbon in the atmosphere or in seawater after the Fukushima accident.

The aim of the present study has been to assess, compare and evaluate tritium, radiocarbon and radiocesium records in the water column of the western North Pacific Ocean after the Fukushima accident. It has been important to estimate changes in ^{3}H, ^{14}C and ^{137}Cs concentrations in Pacific waters after the Fukushima accident not only from a radioecological point of view, but also due to the fact that these radionuclides have been frequently used as tracers of ocean and ocean-atmosphere processes. We need to know how radionuclide concentrations have changed in the Pacific Ocean after the Fukushima accident, and how we can apply pre-Fukushima and post-Fukushima data in oceanographic research. The observed radionuclide patterns and their comparison with pre-Fukushima data will contribute to future oceanographic and climate studies using these radionuclides as tracers of processes in the marine environment.

The radionuclide data presented in this paper were obtained from analyses of seawater samples collected during the KoK (Ka'imikai-o-Kanaloa) cruise, carried out in June 2011 offshore Fukushima. Preliminary data on tritium and radiocesium (mostly surface samples) were discussed in our previous paper (Povinec et al., 2013d). In the present paper we discuss the final data set on tritium (21 results) and radiocesium (45 results), together with 12 radiocarbon results (Table 1). A comprehensive radiocesium data set, covering the KoK seawater sampling, has been published by Buesseler et al. (2012). In this paper we report complementary ^{3}H, ^{14}C, ^{134}Cs and ^{137}Cs data, and compare them with recently published post-Fukushima radiocesium data (Aoyama et al., 2012a,b; 2013; 2015a, b; Buesseler et al., 2012; Kaeriyama et al., 2013; Kumamoto et al., 2013a, 2014, 2015), as well as with pre-Fukushima ^{3}H, ^{14}C and ^{137}Cs data obtained for the western North Pacific Ocean (Ostlund et al., 1987; Watanabe et al., 1991; Tsunogai et al., 1995; Aramaki et al., 2001; Kumamoto et al., 2002; Povinec et al. 2003, 2004c, 2010).

2. Direct releases and atmospheric deposition of ^{137}Cs, ^{3}H and ^{14}C during the Fukushima accident to the western North Pacific Ocean

2.1. Radiocesium

The largest radionuclide releases to the coastal ocean were due ^{137}Cs because of its large production rates in the damaged Fukushima nuclear reactors (yield 6.2%). The ^{137}Cs inventory in the damaged nuclear reactors has been estimated to be about 700 PBq (230 PBq recovered), and 140 PBq in stagnant water (Povinec et al., 2013a). The ^{134}Cs production in the damaged Fukushima nuclear reactors was similar to that of ^{137}Cs, therefore its inventory and the direct ^{134}Cs releases to the sea were estimated to be similar to ^{137}Cs ones on the basis of their activity ratio close to one (Kirchner et al., 2012).

2.1.1. Direct releases of radiocesium to the sea

Kawamura et al. (2011) using the TEPCO (Tokyo Electric Power Company) data and numerical simulations estimated the total ^{137}Cs release for the period from 21 March to 30 April 2011 to be 4 PBq. Tsumune et al. (2013) using a global ocean circulation model and the available ^{137}Cs monitoring data estimated the direct ^{137}Cs releases between 26 March and 31 May 2011 to be 3.5 ± 0.7 PBq. Similar estimations were obtained using various oceanic models: 1-3.5 PBq by Dietze and Kriest (2012), 4.1-4.5 PBq by Estournel et al. (2012), and 5.5–5.9 PBq by Miyazawa et al. (2012a, b). Recently Aoyama et al. (2015a, b) critically reviewed the different ^{137}Cs release rate scenarios, and their best estimation is 3.6 ± 0.7 PBq.

2.1.2. Deposition of radiocesium over the Pacific Ocean

The estimation of the atmospheric deposition of ^{137}Cs over the Pacific Ocean is 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}. It has been expected therefore that the wet and dry deposition over the huge Pacific Ocean may dominate over the radiocesium transport from coastal waters to the open ocean (Aoyama et al., 2015a, b). Rypina et al. (2013) using the KoK data estimated the contribution from the atmospheric source to be in the range of 0 – 11 PBq. The KoK data are, however, not well-suited for constraining the atmospheric source strength because of the restricted sampling area (only about 600 km east from the coast), and the fact that most of the Fukushima fallout ^{137}Cs had left the survey area by mid-June when the KoK sampling cruise was carried out. Aoyama et al. (2015b) recently reviewed the available data

and came with the estimation that the ^{137}Cs wet and dry deposition over the North Pacific Ocean should be in the range of 12-15 PBq from the total atmospheric release of 14-17 PBq.

2.2. Tritium

Tritium production in the Fukushima boiling-water reactors was mainly due to a ternary fission of ^{235}U (yield of 0.013%) and of ^{239}Pu (0.023%), with the estimated annual ^{3}H production of about 1 PBq/GW_e (Povinec et al., 2013a). Tritium was also produced in nuclear reactions with ^{10}B in boron carbide control rods

^{10}B (n, 2α) ^{3}H

^{10}B (n, α) $^{7}Li \Rightarrow {}^{7}Li$ (n, αn) ^{3}H.

Further, the ^{3}H production was also due to the neutron capture reaction on deuterium present in the water coolant (^{2}H (n, γ) ^{3}H) and in the reaction ^{7}Li (n, αn) ^{3}H on lithium impurities in the water. The nuclear reaction contributions to the annual ^{3}H production were, however, much smaller (about 0.1 TBq/GW_e) than the ternary fission. Tritium will not be released at once even if the fuel and the boron control rods would be damaged. Generally, boiling-water nuclear reactors produce less tritium than pressurized water reactors and much less than heavy water reactors (Chudý and Povinec, 1982; Povinec et al., 2013a).

The ^{3}H inventory in three damaged nuclear reactors was estimated to be about 3.4 PBq (1-1.2 PBq in each reactor; Nishihara et al., 2012; http://www.meti.go.jp/earthquake/nuclear/pdf/140424/140424_02_008.pdf), much higher than the value of 0.018 PBq reported by Schwantes et al. (2012). From the total of 3.4 PBq produced tritium, about 0.8 PBq was stored in tanks, about 0.05 PBq was in stagnant water accumulated in the reactors and turbines buildings, and about 0.05 PBq was in seawater pipe trenches. Rest of the tritium inventory should be in the fuel debris, and part of it was released directly to the sea.

The ^{3}H activity concentration in stagnant water in June 2011 was in the range 0.7–20 MBq L^{-1} (lower by a factor of about 10^{5} when compared e.g. with heavy water reactors). The release ratio from nuclear cores to the stagnant water was estimated to be the highest for ^{3}H (46%), ^{131}I (32%), and ^{137}Cs (20%), but only 1.6% for ^{90}Sr (Nishihara et al., 2012). This would indicate that the maximal direct releases to the sea could be for ^{131}I, ^{137}Cs, ^{90}Sr and ^{3}H, of about 1900, 140, 8 PBq and 1.6, respectively (if all the stagnant water would be released to the sea). The $^{3}H/^{137}Cs$ and $^{3}H/^{90}Sr$ activity ratios in the stagnant water were $7x10^{-3}$ and 0.2, respectively, which could be expected in coastal waters as well. Additionally, during the

accident, external cooling with fresh and later also with seawater was carried out, which could also increase radionuclide levels in coastal waters.

At present the total water volume in storage tanks at the Fukushima NPP site is about 460,000 m^3, which is undergoing decontamination (except for tritium) in a Multi-Nuclide Removal System (ALPS), with typical ^3H activity concentrations of 800 kBq L^{-1} (Povinec et al., 2013a). It is very likely that the tritiated water will be discharged to the sea, or evaporated and released to the atmosphere.

Unfortunately, there are no data available on tritium levels during the early stage of the Fukushima NPP accident at the seaport, close to discharge points. The TEPCO reported for August 2013 a value of 68 Bq L^{-1} (www.TEPCO.com). We shall estimate the total amount of ^3H released to coastal waters and deposited over the North Pacific Ocean later when we shall calculate both the ^{137}Cs and ^3H inventories in the water column. A preliminary estimation of the total ^3H releases to the marine environment (using a very limited data set) was in the range of 0.1-0.5 PBq (Povinec et al., 2013d). Estimation of the ^3H source term in the western North Pacific Ocean depends 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 the ^{137}Cs source term, the estimation of the ^3H source term will also improve.

Generally, the tritium inventory should be much smaller when compared with global fallout and with natural production of tritium. The total bomb produced tritium inventory was estimated to be around 186 EBq in the atmosphere and 113 EBq in the ocean, from which 8 EBq was still present in 2010 (Povinec et al., 2010, 2013a). The inventory of ^3H discharged from reprocessing nuclear facilities to the ocean was 410 PBq, from which 45 PBq was still present in 2010. The annual cosmic-ray production, predominantly in the reactions

^{14}N (n, ^3H) ^{12}C and ^{16}O (n, ^3H) ^{14}N

is much lower, around 0.15 EBq, varying with cosmic-ray flux. The world steady-stay natural inventory is about 2.2 EBq.

2.3. Radiocarbon

There are no data available on radiocarbon source term, and on its gaseous and liquid releases after the Fukushima accident. Radiocarbon is an activation product formed mainly in the ^{14}N (n, p) ^{14}C and ^{17}O (n, α) ^{14}C nuclear reactions on target nuclei present in the fuel, and on impurities in the fuel rods and in the coolant/moderator (Chudý and Povinec, 1982). The ^{14}C is also produced in ternary fission (with yield of 1.6×10^{-4} %), however, its contribution is

negligible when compared with nuclear reactions. For the BWR reactors the dominant annual production is on ^{14}N impurities in the fuel, in the rods and in the water coolant (1.3 TBq/GW_e) and on ^{17}O present in the fuel and in the water coolant (0.7 TBq/GW_e). The total annual ^{14}C production in BWRs is estimated to be around 2 TBq/GW_e, from which about 1.2 TBq/GWe should be present in the fuel rods, and about 0.8 TBq/GWe in the water coolant (Povinec et al., 2013a).

Generally, the production rates of Fukushima-derived radiocarbon should be negligible when compared with bomb produced radiocarbon, and its natural production by cosmic rays. The radiocarbon inventory due to atmospheric nuclear bomb tests was about 213 PBq in the atmosphere and about 130 PBq in the ocean (UNSCEAR, 2008). The ^{14}C inventory from discharges from reprocessing nuclear facilities was about 2 PBq, mostly in the marine environment (Povinec et al., 2000). The natural ^{14}C is produced in the atmosphere by cosmic-ray neutrons in the same reactions as mentioned above, the dominant reaction being ^{14}N (n, p) ^{14}C. The annual ^{14}C production is about 1.4 PBq, which vary with the cosmic-ray flux. The total inventory is about 8.5 EBq, of which about 0.14 EBq is in the atmosphere and rest is mostly in oceans (UNSCEAR, 2008).

3. Samples and methods

3.1. Seawater samples

Seawater samples were collected offshore Fukushima during the US expedition using the research vessel Ka'imikai-o-Kanaloa (KoK) of the University of Hawaii (3—17 June, 2011) from about 30 km to about 600 km (from 34 to 38° N, and from 141.5 to 147° E) east of the Japan coast (Buesseler et al., 2012). Locations of KoK sampling stations used in this study are shown in Fig. 1a. Water column sampling stations, visited during other cruises (which have been used for comparison) are shown in Fig. 1b.

Surface samples for radionuclide analyses were collected using a water pump. Water column samples were collected at different depths using a Rosette sampling system equipped with Niskin bottles. Half liter of seawater was used both for ^{3}H and ^{14}C analyses, while ^{137}Cs was analyzed in 1 L seawater samples. The bottles had double plugs to prevent a penetration of air into the water sample during transport and storage. Surface seawater samples were collected at 17 stations, and water profile samples at 12 stations.

2.2. Tritium analysis

Pre-screening of ^{3}H levels in seawater samples (to avoid possible contamination) was carried out in the Bratislava laboratory 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 in the Debrecen and Saclay laboratories using the ^{3}He in-growth mass spectrometry method. The procedure used in Debrecen (Palcsu et al., 2010) consists of three major steps: First, the water sample is put into a stainless steel vessel, and the dissolved gases including helium are removed from the water by vacuum pumping. The samples are then stored for several months so that ^{3}He atoms are produced by tritium decay. The helium fraction is finally admitted to a dual collector noble gas mass spectrometer, where the abundance of the tritiogenic ^{3}He is measured, and ^{3}H activity is calculated.

In the procedure used in Saclay (Jean-Baptiste et al., 2010) the water samples were transferred and sealed in low helium diffusivity glass bulbs that had been previously baked at 600°C under a stream of argon. After an equilibrium time of about 100-150 days, the bulbs were then connected to a MAP-215-50 noble gas mass spectrometer equipped with a stainless steel low blank inlet system (^{3}He blank < $3x10^{-20}$ mol) for measurements that were calibrated against an air standard (He =5.24 ppm, $^{3}He/^{4}He = 1.38x10^{-6}$) drawn from a 5 L tank filled with clean air at known pressure, temperature and relative humidity conditions, through a precisely calibrated volume of 122.2 ± 0.1 (at 2 sigma) mm^3. The ^{3}He ingrowth method can measure ^{3}H levels down to 0.01 TU (precision at 1 σ is around 0.005 TU).

It has been widely accepted in oceanographic studies that tritium levels has been reported in Tritium Units (TU). One TU represents an isotope ratio of 1 ^{3}H atom to 10^{18} protium (^{1}H) atoms, which is equivalent to 0.118 Bq L^{-1} of water.

2.3. Radiocarbon analysis

The ^{14}C measurements from dissolved inorganic carbon (DIC) of the seawater samples were carried out using an accelerator mass spectrometry (AMS) in the Debrecen laboratory. However, to avoid a possible ^{14}C contamination of the AMS system, a few seawater samples with highest ^{137}Cs levels were checked for ^{14}C content in the Bratislava laboratory by a gas proportional counting technique (Povinec et al., 2013e). For AMS analyses, the dissolved CO_2 was extracted from 20 mL seawater samples by adding 85% phosphoric acid, followed by 2 hours heating at 75 °C in a heating block. The degassed CO_2 was extracted and

306 cryogenically cleaned in a vacuum line (Molnár et al., 2013a). The obtained pure CO_2 gas
307 was then converted to graphite by sealed tube graphitization method (Rinyu et al., 2013). The
308 ^{14}C measurements from the graphite targets were performed with a MICADAS type AMS in
309 the Debrecen laboratory (Molnár et al., 2013b). The overall measurement uncertainty for each
310 sample was below 5‰, including normalization, background subtraction, and counting
311 statistics. The ^{14}C results were corrected for decay of the standard and for isotope
312 fractionation using ^{13}C measurements.

313 The ^{14}C concentration is given as $\Delta^{14}C$ (in ‰), relative to the NIST (National Institute of
314 Standards and Technology, Gaithersburg, USA) ^{14}C oxalic acid standard (Stuiver and
315 Ostlund, 1983)

316 $$\Delta^{14}C = \delta^{14}C - 2(\delta^{13}C + 25)(1 + \delta^{14}C/1000)\ (‰) \quad \text{(eq. 1)}$$

317 $$\delta^{14}C = \delta^{14}C = [(^{14}C_{sample} - {}^{14}C_{standard})/{}^{14}C_{standard}] \times 1000\ (‰) \quad \text{(eq. 2)}$$

318 The $\delta^{13}C$ is the $^{13}C/^{12}C$ ratio measured by the AMS simultaneously with the ^{14}C measurement.

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320 2.4. Analysis of radiocesium

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322 The seawater samples were nondestructively counted directly using high efficiency HPGe
323 spectrometers placed in the in the CAVE (Counting Laboratory for Environmental
324 Radionuclides) underground facility of IAEA-EL (Povinec et al., 2004a, b, 2005b, 2012b).
325 The counting periods were between 24 and 72 hours.

326 The data quality of radionuclide analyses was assured by regular participation in
327 intercomparison exercises, and by analysis of IAEA (International Atomic Energy Agency)
328 reference materials (Irish Sea water: IAEA-381, Povinec et al., 2002; Mediterranean Sea
329 water: IAEA-418, Pham et al., 2010; and Irish Sea water: IAEA-443, Pham et al., 2011).

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331 4. Results and discussion

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333 4.1 Radiocesium

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335 The measured ^{137}Cs activity concentrations in surface waters collected 30-600 km offshore
336 the Fukushima NPP ranged from 15 mBq L^{-1} to 1120 mBq L^{-1} (Table 1, Fig. 2a). They were
337 by about three orders of magnitude higher than the global fallout background, although the
338 cruise track did not go closer than 30 km from the coast. Even at distances around 600 km off
339 Fukushima, ^{137}Cs activity concentrations of around 0.2 Bq L^{-1} were found, i.e. by about a

factor of 200 above the global fallout background of 1 mBq L^{-1}. The observed elevated ^{137}Cs levels covered an area of around 150 000 km^2 (south of 38° N and west of 147° E).

A considerable decrease in surface ^{137}Cs levels took place between the KoK cruise measurements (3.5 Bq L^{-1}, June 2011) and those carried out earlier when about 24 Bq L^{-1} of ^{137}Cs were measured in seawater 30 km offshore Fukushima (Povinec et al., 2013a). The ^{134}Cs and ^{137}Cs data presented in Table 1 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.

Figure 2a clearly separates the western KoK stations with higher ^{137}Cs levels from the eastern stations with smaller activity concentrations. The geographical distribution of ^{137}Cs levels in the water column indicates that an atmospheric deposition of ^{137}Cs occurred within the 600 km zone offshore Fukushima. For example, the increased ^{3}H, ^{134}Cs and ^{137}Cs activities found at St. 8 (Table 1), in agreement with other KoK data (Buesseler et al., 2012), should not be affected by direct liquid releases from the Fukushima NPP, but an atmospheric deposition took place there.

At some stations (e.g., St. 18, 24) subsurface maxima at 50 m water deth have already been observed three months after the accident (Table 1, Fig. 2a). At other stations (e.g., St. 22, 25 and 26) located closer to the Fukushima coast, the maximum radiocesium leves were observed in surface seawater, especially at St. 25, where surface ^{137}Cs levels exceeded the global fallout values by about three orders of magnitude.

Distribution of ^{137}Cs in the water column of the western North Pacific Ocean was investigated in the framework of the WOMARS (Worldwide Marine Radioactivity Studies) project, carried out by IAEA during 1995-2005 (Livingston and Povinec, 2005; IAEA, 2005; Povinec et al., 2005b, 2010). On the basis of the data stored in the GLOMARD/MARIS database (Povinec et al. 2004b, 2005b, 2006; www.iaea.org/monaco), and taking into account an effective half-life of ^{137}Cs in surface waters of 13±1 yr (Povinec et al., 2005b), the background (global fallout) ^{137}Cs level for the western North Pacific Ocean was estimated to be (1.1 ± 0.1) mBq L^{-1}. Unfortunately the IAEA'97 station (Fig. 1b) is not in the KoK region (called the Mixed-Water Region or Transition Zone), but it was under influence of the Kuroshio Current (Povinec et al., 2003). However, as we do not have better pre-Fukushima ^{137}Cs profiles we compare in Fig. 2b the KoK profiles with the IAEA'97 (St. 1, decay corrected to June 2011) profile. All KoK ^{137}Cs data are well above the IAEA'97 data, confirming thus Fukushima impact on all visited stations.

Fig. 2b compares the ^{137}Cs water profiles with other post-Fukushima data (Ramzaev et al. 2014; Kumamoto et al., 2014, 2015; Aoyama et al. 2015a), which were measured in the Transition Zone, however, some of the stations were outside of the KoK region. The highest ^{137}Cs levels were measured at the KoK stations (St. 25 and 23) as well as at the UM and KT stations situated closest to the coast. The surface ^{137}Cs levels in the KoK region are mostly between 0.1 and 1 Bq L^{-1}, while the levels at other stations are below 0.1 Bq L^{-1}. Although the P10 stations (e.g. St. P10-098) are at larger distances from the coast than the RGS stations (e.g. St. 7), their ^{137}Cs levels are higher, indicating possible deposition of ^{137}Cs from radioactive clouds.

The inventory of a radionuclide in a seawater column was calculated by interpolating the radionuclide concentration measured at each depth using the following equation

$$I_R = \frac{1}{2}\left\{\sum_{i=1}^{N}(W_{i+1}+W_i)(d_{i+1}-d_i) + 2W_1 d_1 + 2W_N(d_B - d_N)\right\}, \quad \text{(eq. 3)}$$

where I_R is the inventory of the radionuclide R in the seawater column (Bq/m^2), N is the number of sampling depths, W_i is the radionuclide concentration in seawater at depth i, d_i is the i-th sampling depth of seawater, and d_B is the total water depth to the bottom. For example the ^{137}Cs inventory calculated for the water column at St. 26 was (30 ± 3) kBq m^{-2}. The total inventories were calculated by integrating water column inventories and multiplying them by the corresponding areas.

Our preliminary ^{137}Cs inventory calculated for the KoK region was (2.2 ± 0.3) PBq (Povinec et al., 2013d), in reasonable agreement with previous estimation of Buesseler et al. (2012). In the present paper we included all the available ^{137}Cs data measured during 2011-2012 in the region (Honda et al., 2012; Kaeriyama et al., 2013; Kameník et al., 2013; Ramzaev et al., 2014; Kumamoto et al., 2013a, 2014, 2015; Aoyama et al., 2016a, b), and we got for the total inventory a value of (2.7 ± 0.4) PBq. This lower value when compared with the recent estimation of the direct liquid release inventory of (3.6 ± 0.7) PBq (Aoyama et al., 2015b), may be due to the fact that the investigated region covered only a limited area, 36 - 39°N and 141.5° - 148°E, as well as that part of the direct liquid discharges have already been transported out of the investigated region when seawater samling in 2011-2012 was carried out.

Unfortunately we do not know a pre-Fukushima ^{137}Cs inventory for the investigated region. We may expect, however, that it will be lower than for the western North Pacific due to the presence of Oyashio Intrusion waters in the western part of the Transition Zone (Tatebe

and Yasuda, 2004; Fig. 1a). Upwelling of deeper waters (with lower radionuclide levels) occurs in the Oyashio region, which are then transported by the Oyashio Intrusion to the western part of the Transition Zone, causing there a dilution of radionuclide levels (including ^{137}Cs). Our estimation is that the pre-Fukushima ^{137}Cs inventory in the western part of the KoK region should be around 200 Bq m^{-2} (for the top 200 m of the water column), i.e. by about a factor of two lower than the value estimated for the open western North Pacific Ocean (Aoyama end Hirose, 2004; Povinec et al. 2010). This global fallout background is negligible (~0.01 PBq) when compared with the Fukushima impact on the ^{137}Cs inventory in western North Pacific waters.

4.2. Tritium

Tritium levels measured in the KoK samples varied in the range of 0.4-1.3 TU (Table 1), with the maximum at St. 25 (sampled at 10 m water depth) situated 30 km off the Fukushima coast (the closest station to the Fukushima NPP, Fig. 1a), where the radiocesium levels were also at their maxima (Table 1). All KoK sampling stations were affected by the Fukushima-derived tritium, the lowest tritium activity (0.37 ± 0.02 TU) was found at St. 11 at 100 m water depth. As we already mentioned in the radiocesium section, the western Kok stations were under the influence of the Oyashio Intrusion, which should also dilute tritium concentrations in this region (Figs. 1a and 3).

The tritium seawater profiles (Fig. 3) generally follow the ^{137}Cs water profiles presented in Fig. 2a. The average ^{3}H/^{137}Cs activity ratio is 0.5 ± 0.3 (Table 1), with high values at the depth of 100 m at St. 20, 26 and 25 (2.2, 2.0 and 1.2, respectively), indicating a penetration of tritium into deeper layers of the water column. On the other hand, the lowest ^{3}H/^{137}Cs activity ratios (0.1-0.3) observed in surface waters (7-20 m) at several stations (e.g., St. 18, 27, 29, 30, 31, 32) have been confirming the dominance of radiocesium in surface waters. Generally, the observed lower tritium than radiocesium levels may be attributed to lower tritium releases during the Fukushima accident, due to much lower fission yield of ^{3}H compared to ^{137}Cs, as well as due to the fact that the boiling water reactors produce much less tritium than the pressurized water reactors.

Although the most comprehensive oceanographic study with the most extensive coverage of ^{3}H in the World Ocean was the WOCE (World Ocean Circulation Experiment, www.eWOCE.org) program conducted in the 1980s and 1990s, there were no sampling stations in the KoK region, which could be compared with our data. Tritium distribution in

the western North Pacific Ocean was also investigated in the framework of the WOMARS project. The background tritium level for the western North Pacific Ocean can be estimated from the GLOMARD/MARIS database (Povinec et al., 2004b, 2005b) to be (0.4 ± 0.1) TU (decay corrected to June 2011).

The only available pre-Fukushima tritium profile in the KoK region is from Watanabe et al. (1991), measured in 1988 (Fig. 3, decay corrected to June 2011). Their results are generally lower when compared with the IAEA'97 station (Povinec et al., 2010), although the ^{3}H profiles should be between the GEOSECS (Ostlund et al., 1987) and the IAEA'97 profiles. This may be due to the fact that the Watanabe et al. (1991) station was located in the western Transition Zone (with influence of the Oyashio Intrusion), while the GEOSECS and IAEA stations were in the Subtropical Zone and in the Kuroshio region, respectively (Fig. 1b). Following the Watanabe et al. (1991) profile (Fig. 3), the pre-Fukushima tritium background concentration in surface water of the western KoK region can be as low as 0.2 TU, increasing thus the Fukushima impact on tritium levels by about a factor of six.

There was generally agreement between the GEOSECS, IAEA and Watanabe et al. stations regarding the vertical transport of tritium in the water column. A penetration of global fallout ^{3}H down to about 1000 m depth has been observed, which also predicts the future transport of Fukushima tritium in the water column. The future penetration of Fukushima-derived tritium from surface waters into deeper layers of the ocean will be useful for studying pathways and time scales of deep-water formation (e.g. Povinec et al., 2010, 2011).

The tritum inventory in the water column at St. 26, situated close to the Fukushima coast, calculated using eq. 3 and the water depth profile data is (17 ± 2) kBq m^{-2}, which is aproximately half of the ^{137}Cs inventory in the same location, thus leading to the $^{3}H/^{137}Cs$ activity ratio of ~0.5. A preliminary estimation of the total ^{3}H releases to the marine environment was in the range of 0.1-0.5 PBq (Povinec et al., 2013d). From the full set of $^{3}H/^{137}Cs$ activity ratios in the water column, and the previously estimated ^{137}Cs releases to the sea, we may estimate the total ^{3}H activity released and deposited offshore of the Fukushima coast to be (0.3 ± 2) PBq.

Matsumoto et al. (2013) estimated the ^{3}H concentration in the air during an early stage of the accident in March 2011 to be around 1.5 kBq m^{-3}. The ^{3}H levels in precipitation collected at distances 170-220 km from the Fukushima NPP during 21-23 March 2011 were up to 19 Bq L^{-1} of water, about 27 times above the pre-Fukushima levels (Matsumoto et al. 2013), but still by a factor of 5 below the EU limit for ^{3}H concentration in drinking water. Similarly, Liu et al. (2013) observed in Taiwan rains during May-December 2011 ^{3}H levels below 7 Bq L^{-1}

of water. All these levels have been much lower than the tritium maximum (up to 1180 Bq L^{-1} of water) observed in 1963 in the Northern Hemisphere precipitation after the large-scale atmospheric nuclear bomb tests carried out during 1961 and 1962 (Povinec et al., 2013a).

Measured tritium concentrations and the calculated tritium inventories confirm that the distribution of ^{3}H in the western North Pacific Ocean was influenced by the Fukushima accident. The pre-Fukushima tritium data, e.g. those previously gathered in the framework of the WOCE and WOMARS projects do not represent anymore global fallout distribution of this radionuclide in the western North Pacific Ocean.

4.3. Radiocarbon

The observed ^{14}C levels in KoK water profiles (Fig. 4) show typical features: the concentrations were highest usually at the sub-surface layers (around -20‰ at 100-200 m water depth), and then they decreased down to around -115‰ at 400-500 m water depth. All $\Delta^{14}C$ values observed offshore Fukushima were negative, much lower when compared with the IAEA'97 (Aramaki et al., 2001; Povinec et al., 2004c), GEOSEC (Stuiver and Östlund, 1983) and WOCE stations (Key et al. 1996, 2002, 2004). The ^{14}C of global fallout origin observed at these stations penetrated down to about 1000 m (Stuiver and Ostlund, 1983; Povinec et al., 2004c), similarly as it was observed in the case of ^{3}H (Ostlund et al., 1987; Povinec et al., 2010). The observed decrease in surface $\Delta^{14}C$ levels after GEOSECS (~150‰ in 1973; Stuiver and Östlund, 1983) and IAEA'97 (~90‰ in 1997; Povinec et al., 2004c) would predict $\Delta^{14}C$ levels in 2011 around 50‰. However, all the measured $\Delta^{14}C$ levels in 2011 in the KoK samples were below 0‰, indicating that a comparison with IAEA and GEOSECS stations has not been appropriate because of different position of sampling stations. We have already noticed that ^{3}H levels in the western KoK region were influenced by Oyashio Intrusion, which brought low tritium activity waters to this region. In the case of ^{14}C the situation could be similar as the negative $\Delta^{14}C$ values were observed at all visited stations, including water profiles at St. 11 and 20 down to 500 m water depth (Fig. 4). Pre-Fukushima ^{14}C water profiles, measured in the western North Pacific by Tsunogai et al. (1995) and Kumamoto et al. (2002) also showed negative $\Delta^{14}C$ values (Fig. 4), however, these stations were located in the Oyashio region (Fig. 1b).

Surface ^{14}C concentration in seawater is regulated mainly by the CO_2 air – sea exchange, and the residence time of surface water in the region (Kumamoto et al., 2013b). Because of

the upwelling of deeper waters with lower ^{14}C levels in the Oyashio region, and their transport by the Oyashio Intrusion to the western part of the Transition zone, the surface ^{14}C levels in this region are lower than in the Kuroshio region as a mixing of Oyashio and Kuroshio waters occur in the Transition Zone (Kumamoto et al., 2002). All KoK ^{14}C stations located in the western Transition Zone showed negative $\Delta^{14}C$ values, indicating that these stations were under the influence of the Oyashio Intrusion waters. On the other hand, St. C22 and C23 sampled in 1993 by Aramaki et al. (2005) showed positive $\Delta^{14}C$ values (around 70‰), confirming that these stations were outside of the Oyashio Intrusion region (Fig. 1a, b).

The data presented in Fig. 4 show that the ^{14}C levels measured in the KoK seawater samples are higher than the pre-Fukushima results measured in the Oyashio region (44°00'N, 155°00'E) in 1997 by Kumamoto et al. (2002), but comparable with the results of Tsunogai et al. (1995) measured in 1985, also in the Oyashio region (43°00´N, 150°14´E). As we do not have pre-Fukushima ^{14}C data for the KoK region, we may accept the Kumamoto et al. (2002) surface ^{14}C data as a possible background for the KoK stations (after appropriate residence time corrections between 1997 and 2011). This assumption seems to be reasonable for surface St. 19, 21, 25 and 29 (the average $\Delta^{14}C$ value beeing -55‰), however, the water profiles at St. 11 and 20 showed at 100 m and 200 m water depths increased $\Delta^{14}C$ values to -20‰, while at water depths of 400 m and 500 m they decreased to around -115‰. If we take these deeper values as a possible background (as at the 400-500 m water depth we do not expect a Fukushima impact already in June 2011), the Fukushima contributions to ^{14}C levels in the investigated region can be 6% and 9% for surface and 100-200 m water depths, respectively. As the KoK ^{14}C stations were not located close to the coast, we may expect (similarly as in the case of ^{137}Cs and ^{3}H) that the Fukushima impact on the ^{14}C levels in the western part of the transition Zone could be even stronger.

There are no additional data available on radiocarbon levels in the atmosphere or in seawater after the Fukushima accident. Possible ^{14}C contributions of Fukushima origin were discussed in tree rings of the Japanese cedar grown in Iwaki (Fukushima Prefecture, Xu et al., 2015), and in ginkgo leaf samples from Korea (Park et al., 2013). Both samples showed for 2011 a smaller than expected decrease in ^{14}C from the exponential trend, which may imply a ^{14}C release during the Fukushima accident.

5. Conclusions

539 The analysis of ^{137}Cs, ^{3}H and ^{14}C in surface and water column samples collected in the
540 western North Pacific Ocean after the Fukushima accident showed that the Fukushima impact
541 has been mainly on ^{137}Cs in the marine environment. The ^{137}Cs concentrations observed in
542 surface seawater of the western North Pacific Ocean were due to the Fukushima accident
543 higher by about three orders of magnitude. On the other hand the ^{3}H levels were above the
544 global fallout background only by about a factor of 6. The ^{14}C data suggest that the impact on
545 radiocarbon levels in the western North Pacific Ocean was measurable, but only around 9%
546 above the background. The water column data indicated that the Fukushima-derived
547 radionuclides already in June 2011 reached depths down to around 200 m.
548 The ^{137}Cs inventory in the water column of the investigated western North Pacific Ocean
549 affected by the Fukushima accident was estimated to be (2.7 ± 0.4) PBq, which can be
550 regarded as a lower limit of the direct ^{137}Cs liquid discharges to the ocean. This lower value
551 when compared with the recent estimation of the direct liquid release inventory of (3.6 ± 0.7)
552 PBq (Aoyama et al., 2016b) may be due to the fact that the region under study did not cover
553 the full area affected by the Fukushima liquid releases. On the basis of $^{3}H/^{137}Cs$ activity ratios
554 in the water column profiles, the total ^{3}H inventory was estimated to be (0.3 ± 0.2) PBq.
555 Measured tritium and radiocarbon concentrations in the western North Pacific Ocean
556 confirm that their distribution has been influenced by the Fukushima accident. The pre-
557 Fukushima tritium and radiocarbon data, e.g. those previously gathered in the framework of
558 the WOCE and WOMARS projects, do not represent anymore the global fallout distribution
559 of these radionuclide in the western North Pacific Ocean.
560 Further studies of these radionuclides in the water column of the North Pacific Ocean are
561 necessary for better estimation of the atmospheric input (via a wet and dry deposition) to their
562 total inventory. The investigated radionuclides will be useful tracers during the next decades
563 for studying pathways, ocean currents and time scales of deep and bottom water formation,
564 for studying biogeochemical processes in the water column, circulation of water masses in the
565 North Pacific Ocean, and transport of North Pacific waters to the South Pacific and the Indian
566 Oceans.
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580 **References**

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Table 1

Tritium, radiocarbon and radiocesium in seawater samples collected offshore Fukushima during the KoK cruise.

Station number	Position of the sampling station	Water depth (m)	^{3}H (TU)	$\Delta^{14}C$ (‰)	^{134}Cs (Bq L^{-1})	^{137}Cs (Bq L^{-1})	$^{3}H/^{137}Cs$
2	34°30´ N, 147°00´ E	100			0.029 ± 0.009	0.027 ± 0.009	
		200			0.016 ± 0.006	0.015 ± 0.006	
6	36°30´ N, 147°00´ E	20				0.026 ± 0.009	
8	37°30´ N, 147°00´ E	5	0.90 ± 0.06				
		20			0.178 ± 0.009	0.184 ± 0.010	
		50			0.160 ± 0.012	0.158 ± 0.014	
		100			0.059 ± 0.006	0.051 ± 0.007	
11	37°30´ N, 144°00´ E	20			0.085 ± 0.008	0.072 ± 0.009	
		50			0.105 ± 0.011	0.080 ± 0.009	
		100	0.37 ± 0.02	-19.5 ± 2.9	0.018 ± 0.009	0.037 ± 0.009	1.18± 0.03
		200		-10.4 ± 2.6			
		400		-89.9 ± 3.2			
		500		-113.4 ± 3.0			
18	37°00´ N, 143°00´ E	20	0.74± 0.055		0.234 ± 0.017	0.252 ± 0.021	0.35 ± 0.03
		50			0.426 ± 0.029	0.425 ± 0.031	
		100			0.048 ± 0.009	0.035 ± 0.009	
19	37°33´ N, 142°57´ E	5		-61.5 ± 3.0			
		20			0.188 ± 0.014	0.171 ± 0.016	
20	38°00´ N, 143°00´ E	5	1.05 ± 0.05				
		100	0.74 ± 0.05	-21.0 ± 3.5	0.023 ± 0.005	0.039 ± 0.009	2.24 ± 0.2
		200		-20.5 ± 3.0	0.015 ± 0.005	0.019 ± 0.007	
		400		-102.3 ± 3.5			
		500		-116.8 ± 2.9			
21	37°30´ N, 142°30´ E	5		-50.6 ± 3.1			
		20			0.292 ± 0.027	0.135 ± 0.013	
		50	0.96 ± 0.09		0.117 ± 0.011	0.135 ± 0.013	0.84 ± 0.08
22	38°00´ N, 142°30´ E	8			0.836 ± 0.052	0.841 ± 0.060	
		10			0.217 ± 0.016	0.203 ± 0.016	
		20			0.087 ± 0.009	0.083 ± 0.009	
		50	0.78 ± 0.09		0.122 ± 0.009	0.138 ± 0.010	0.67 ± 0.08
		100			0.053 ± 0.007	0.052 ± 0.008	
24	38°00´ N, 142°00´ E	8			0.064 ± 0.009	0.054 ± 0.007	
		50			0.215 ± 0.013	0.201 ± 0.014	
		75			0.091 ± 0.009	0.094 ± 0.009	
		130			0.023 ± 0.006	0.029 ± 0.005	
25	37°31´ N, 141°27´ E	5		-38.5 ± 3.2			
		10			0.968 ± 0.054	1.120 ± 0.065	
		20	1.28 ± 0.09		0.225 ± 0.018	0.221 ± 0.019	0.68 ± 0.07
		50				0.057 ± 0.012	
		90			0.083 ± 0.009	0.064 ± 0.009	
		100	0.96 ± 0.08		0.120 ± 0.036	0.091 ± 0.030	1.24 ± 0.1
26	37°00´ N, 141°24´ E	7	0.75± 0.06		0.453 ± 0.031	0.441 ± 0.034	0.20 ± 0.02
		20			0.368 ± 0.023	0.386 ± 0.026	
		50	0.68 ± 0.03		0.256 ± 0.012	0.241 ± 0.012	0.33 ± 0.02
		75	0.73 ± 0.02		0.167 ± 0.013	0.188 ± 0.015	0.46 ± 0.02
		90	0.62 ± 0.03		0.085 ± 0.009	0.086 ± 0.009	0.85 ± 0.04
		100	1.22 ± 0.03		0.064 ± 0.006	0.072 ± 0.007	2.00 ± 0.06

		125			0.028 ± 0.008	0.042 ± 0.008	
		173	0.76 ± 0.25				
27	36°30´ N, 141°24´ E	10	0.90 ± 0.09		0.741 ± 0.034	0.748 ± 0.038	0.14 ± 0.02
28	36°01´ N, 141°24´ E	10	0.63 ± 0.06				
29	36°30´ N, 142°05´ E	0		-70.8 ± 3.4			
		10	0.79 ± 0.06		0.377 ± 0.016	0.438 ± 0.019	0.21 ± 0.02
30	36°30´ N, 142°30´ E	10	0.64 ± 0.02		0.765 ± 0.034	0.798 ± 0.040	0.095 ± 0.005
31	37°00´ N, 142°30´ E	10	0.70 ± 0.02		0.452 ± 0.023	0.480 ± 0.027	0.172 ± 0.007
		20			0.546 ± 0.027	0.555 ± 0.029	
		50			0.473 ± 0.022	0.481 ± 0.025	
32	37°00´ N, 142°00´ E	7	0.69 ± 0.10		0.287 ± 0.021	0.311 ± 0.022	0.26 ± 0.04

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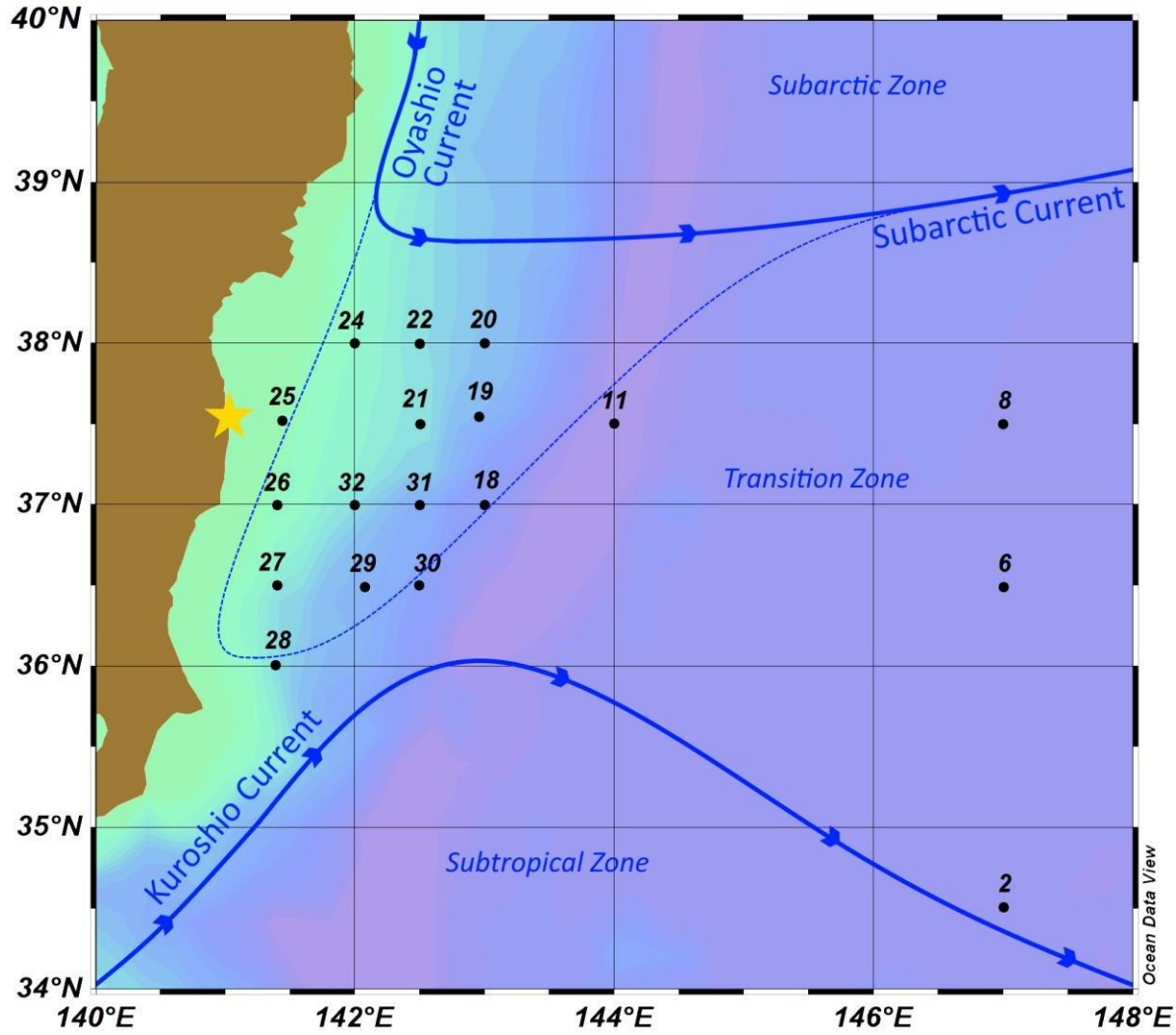

903

904 **Fig. 1a.** Locations of water sampling stations in the western North Paciifc Ocean during the
905 KoK expedition used in this study. Fukushima Dai-ichi NPP accident location is indicated
906 with the star. Approximate positions of the Kuroshio and Oyashio Currents are shown as well.
907 The dashed line represents the southward intrusion of the Oyashio Current (Tatebe and
908 Yasuda, 2004).

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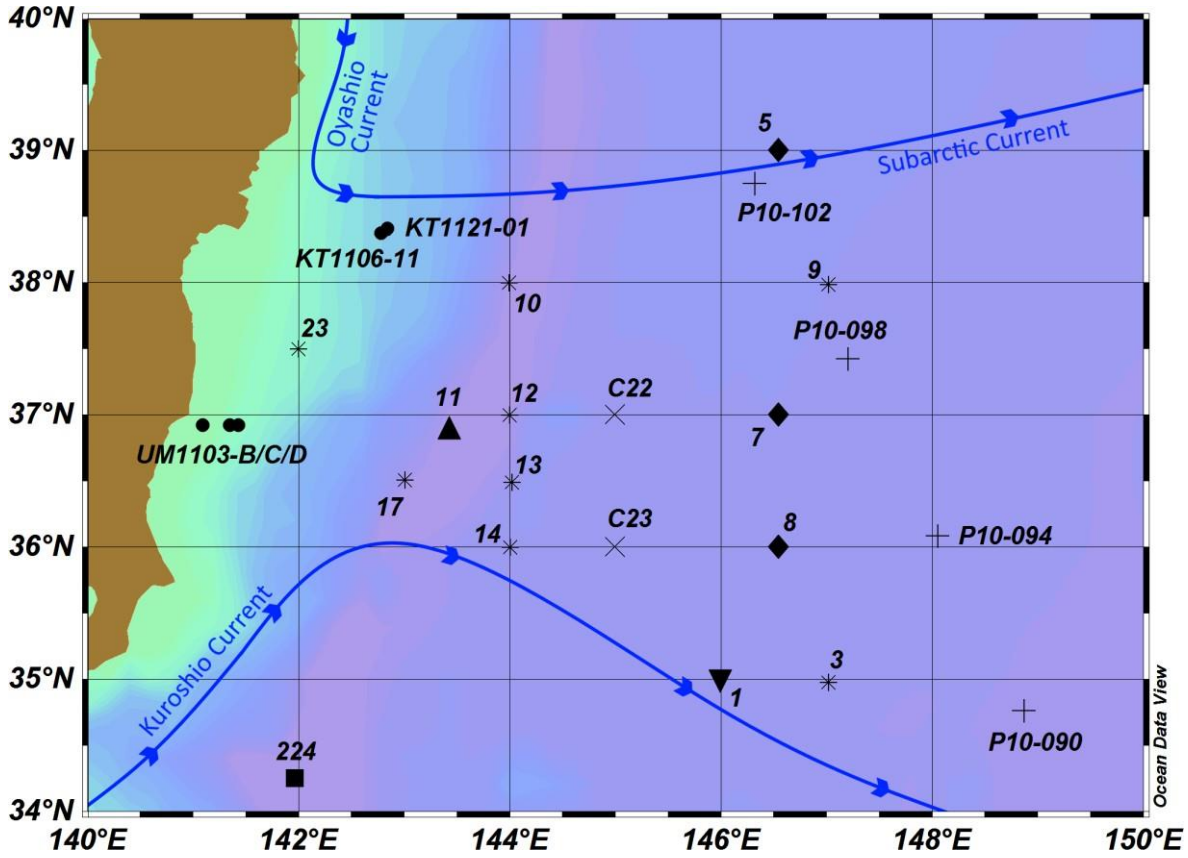

910

911 **Fig. 1b.** Locations of sampling stations used for data comparison: ● - UM11 and KT11
912 stations sampled for ^{137}Cs in 2011 (Aoyama et al., 2016a); ✱ - selected KoK stations sampled
913 for ^{137}Cs in June 2011 (Buesseler et al., 2012); ✕ - KH93 stations sampled for ^{14}C in 1993
914 (Aramaki et al., 2005); ■ - GEOSECS station sampled for ^{3}H (Ostlund et al., 1987) and for
915 ^{14}C in 1973 (Stuiver and Ostlund, 1983); ▼ - IAEA´97 station sampled in 1997 for ^{137}Cs
916 (Povinec et al., 2003), ^{3}H (Povinec et al., 2010) and ^{14}C (Povinec et al., 2004c); ✚ - P10-
917 stations sampled for ^{137}Cs in 2012 (Kumamoto et al., 2015); ◆ - RGS stations sampled for
918 ^{137}Cs in 2012 (Ramzaev et al., 2014); ▲ - KH88 station sampled for ^{3}H in 1988 (Watanabe et
919 al., 1991; The KH85 station sampled for ^{14}C in 1985 (43° 00´N 150°14´E; Tsunogai et al.,
920 1995), and the MR97 stations sampled for ^{14}C in 1997 (44°00'N, 155°00'E; Kumamoto et al.,
921 2002) were situated outside of the KoK region.

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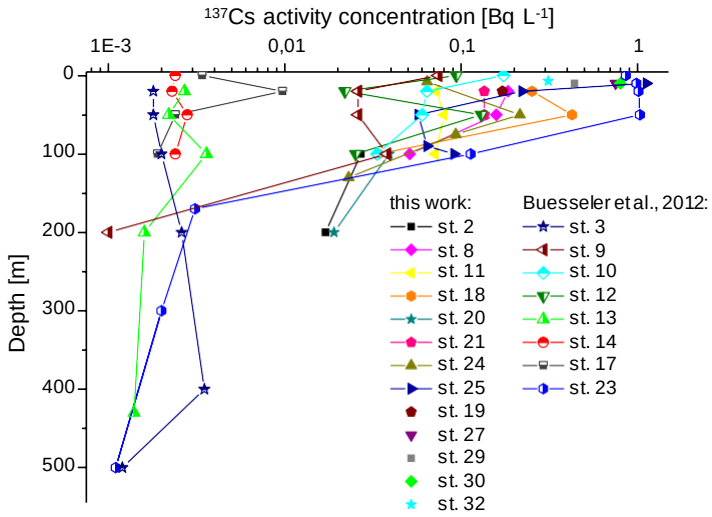


Fig. 2a. ^{137}Cs activity concentration in seawater samples collected offshore Fukushima NPP during the KoK cruise, compared with published data measured during the KoK cruise by Buesseler et al. (2012). For locations of the sampling stations see Figs. 1a, b.

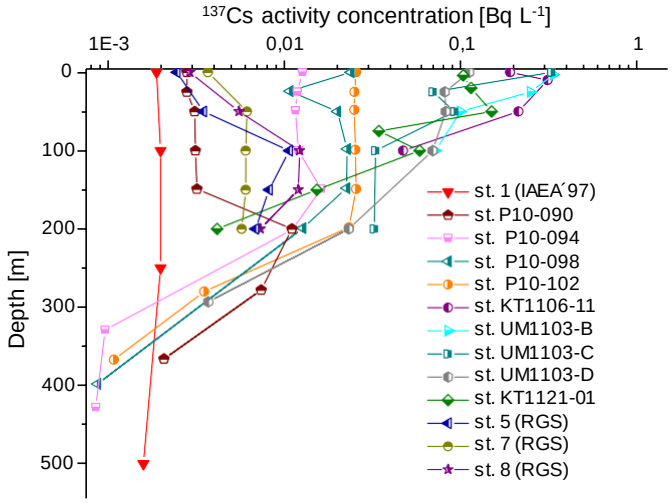
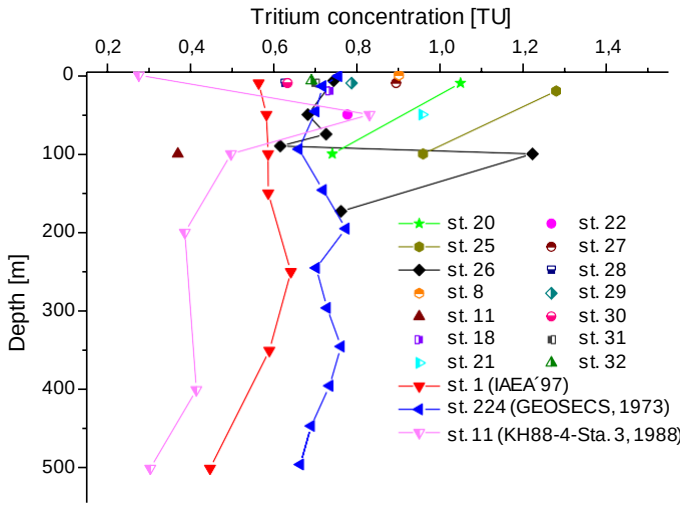


Fig. 2b. ^{137}Cs activity concentration in seawater samples collected offshore Fukushima NPP during the KoK cruise (decay corrected to June 2011), compared with published data measured during the IAEA´97 cruise (Povinec et al., 2003), the RGS cruise (Ramzaev et al., 2014), the P10 cruise (Kumamoto et al., 2015) and the UM11 and KT11 cruises (Aoyama et al. (2016a). For locations of the sampling stations see Figure 1b.



940

941 **Fig. 3.** Tritum concentration in seawater samples collected offshore Fukushima during the
942 KoK cruise (decay corrected to June 2011), compared with the results from the
943 GEOSECS (Ostlund et al., 1987), KH88 (Watanabe et al., 1991) and IAEA´97 (Povinec
944 et al., 2003) expeditions. For locations of the sampling stations see Figure 1b.
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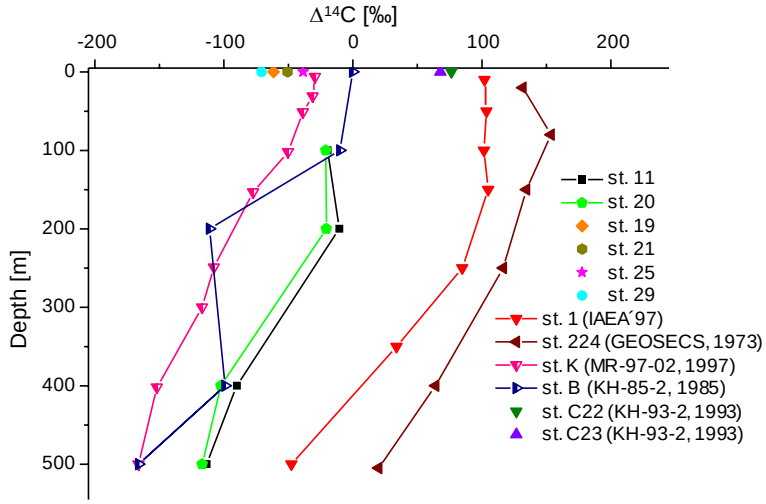

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949 **Fig. 4.** Radiocarbon concentration in seawater samples collected offshore Fukushima during
950 the KoK cruise (decay corrected to June 2011), compared with the results from the
951 GEOSECS (Stuiver and Otlund, 1983), IAEA'97 (Povinec et al., 2004c), KH-85 (Tsunogai et
952 al., 1995), KH93 (Aramaki et. al., 2005) and MR97 (Kumamoto et al., 2002) expeditions. For
953 locations of the sampling stations see Figure 1b.

954

955