



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

► To cite this version:

Pavel P. Povinec, Laval Liong Wee Kwong, Jakub Kaizer, M. Molnar, Hartmut Nies, et al.. 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. *Journal of Environmental Radioactivity*, 2017, 166 (Part 1), pp.56-66. 10.1016/j.jenvrad.2016.02.027 . hal-01584220

HAL Id: hal-01584220

<https://hal.science/hal-01584220>

Submitted on 24 Jun 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

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

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 $(2.7 \pm 0.4) \text{ PBq}$, while for ^3H it was only $(0.3 \pm 0.2) \text{ PBq}$. 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 ^3H , ^{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

<http://www.elsevier.com/open-access/userlicense/1.0/>

1. Introduction

Large quantities of radionuclides were released during the Fukushima Dai-ichi 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., ^3H , ^{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 impact on ^3H , ^{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 $^{131}\text{I}/^{137}\text{Cs}$ 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 ($0.1\text{-}0.5 \text{ PBq}$; 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 $^{13}\text{C}(\text{n}, \gamma)^{14}\text{C}$ 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 ^3H , ^{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 ^3H , ^{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 ^3H , ^{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 , ^3H 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.

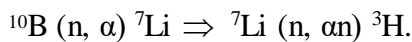
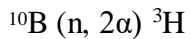
2.1.2. Deposition of radiocesium over the Pacific Ocean

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 ^3H 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



Further, the ^3H production was also due to the neutron capture reaction on deuterium present in the water coolant ($^2\text{H} (n, \gamma) ^3\text{H}$) and in the reaction $^7\text{Li} (n, \alpha n) ^3\text{H}$ on lithium impurities in the water. The nuclear reaction contributions to the annual ^3H 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 ^3H 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 ^3H activity concentration in stagnant water in June 2011 was in the range 0.7–20 MBq L⁻¹ (lower by a factor of about 10⁵ 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 ^3H (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 ^3H , of about 1900, 140, 8 PBq and 1.6, respectively (if all the stagnant water would be released to the sea). The $^3\text{H}/^{137}\text{Cs}$ and $^3\text{H}/^{90}\text{Sr}$ 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 m³, 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](http://www.tepco.co.jp)). 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 ¹³⁷Cs 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 ¹³⁷Cs on the studied sea areas. Therefore with improving our knowledge of the ¹³⁷Cs 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



is much lower, around 0.15 EBq, varying with cosmic-ray flux. The world steady-state 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 ¹⁴N (n, p) ¹⁴C and ¹⁷O (n, α) ¹⁴C 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 ¹⁴C 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/GW_e should be present in the fuel rods, and about 0.8 TBq/GW_e 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 $^{14}\text{N}(\text{n}, \text{p})^{14}\text{C}$. 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 ^3H 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 ^3H levels in seawater samples (to avoid possible contamination) was carried out in the Bratislava laboratory by direct ^3H 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 ^3He 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 ^3He 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 ^3He is measured, and ^3H 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 (^3He blank $< 3 \times 10^{-20}$ mol) for measurements that were calibrated against an air standard ($\text{He} = 5.24$ ppm, $^3\text{He}/^4\text{He} = 1.38 \times 10^{-6}$) 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 ^3He ingrowth method can measure ^3H 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 ^3H atom to 10^{18} protium (^1H) 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

cryogenically cleaned in a vacuum line (Molnár et al., 2013a). The obtained pure CO₂ gas was then converted to graphite by sealed tube graphitization method (Rinyu et al., 2013). The ¹⁴C measurements from the graphite targets were performed with a MICADAS type AMS in the Debrecen laboratory (Molnár et al., 2013b). The overall measurement uncertainty for each sample was below 5‰, including normalization, background subtraction, and counting statistics. The ¹⁴C results were corrected for decay of the standard and for isotope fractionation using ¹³C measurements.

The ¹⁴C concentration is given as Δ¹⁴C (in ‰), relative to the NIST (National Institute of Standards and Technology, Gaithersburg, USA) ¹⁴C oxalic acid standard (Stuiver and Ostlund, 1983)

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

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

The δ¹³C is the ¹³C/¹²C ratio measured by the AMS simultaneously with the ¹⁴C measurement.

2.4. Analysis of radiocesium

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

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

4. Results and discussion

4.1 Radiocesium

The measured ¹³⁷Cs activity concentrations in surface waters collected 30-600 km offshore the Fukushima NPP ranged from 15 mBq L⁻¹ to 1120 mBq L⁻¹ (Table 1, Fig. 2a). They were by about three orders of magnitude higher than the global fallout background, although the cruise track did not go closer than 30 km from the coast. Even at distances around 600 km off Fukushima, ¹³⁷Cs 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 $150\,000 \text{ km}^2$ (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 $^{134}\text{Cs}/^{137}\text{Cs}$ 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 ^3H , ^{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 depth 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 levels 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 $13 \pm 1 \text{ yr}$ (Povinec et al., 2005b), the background (global fallout) ^{137}Cs level for the western North Pacific Ocean was estimated to be $(1.1 \pm 0.1) \text{ mBq L}^{-1}$. 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/m²), 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 m². 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 sampling 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 and Hirose, 2004; Povinec et al. 2010). This global fallout background is negligible ($\sim 0.01 \text{ PBq}$) 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 ($0.37 \pm 0.02 \text{ TU}$) 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 $^3\text{H}/^{137}\text{Cs}$ 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 $^3\text{H}/^{137}\text{Cs}$ 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 ^3H 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 ^3H 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 ^3H 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 ^3H 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 tritium 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 approximately half of the ^{137}Cs inventory in the same location, thus leading to the $^3\text{H}/^{137}\text{Cs}$ activity ratio of ~ 0.5 . A preliminary estimation of the total ^3H releases to the marine environment was in the range of 0.1-0.5 PBq (Povinec et al., 2013d). From the full set of $^3\text{H}/^{137}\text{Cs}$ activity ratios in the water column, and the previously estimated ^{137}Cs releases to the sea, we may estimate the total ^3H activity released and deposited offshore of the Fukushima coast to be (0.3 ± 2) PBq.

Matsumoto et al. (2013) estimated the ^3H concentration in the air during an early stage of the accident in March 2011 to be around 1.5 kBq m^{-3} . The ^3H 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 ^3H concentration in drinking water. Similarly, Liu et al. (2013) observed in Taiwan rains during May-December 2011 ^3H 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 ³H 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 ¹⁴C 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 $\Delta^{14}\text{C}$ 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 ¹⁴C 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 ³H (Ostlund et al., 1987; Povinec et al., 2010). The observed decrease in surface $\Delta^{14}\text{C}$ levels after GEOSECS (~150‰ in 1973; Stuiver and Östlund, 1983) and IAEA'97 (~90‰ in 1997; Povinec et al., 2004c) would predict $\Delta^{14}\text{C}$ levels in 2011 around 50‰. However, all the measured $\Delta^{14}\text{C}$ 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 ³H levels in the western KoK region were influenced by Oyashio Intrusion, which brought low tritium activity waters to this region. In the case of ¹⁴C the situation could be similar as the negative $\Delta^{14}\text{C}$ 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 ¹⁴C water profiles, measured in the western North Pacific by Tsunogai et al. (1995) and Kumamoto et al. (2002) also showed negative $\Delta^{14}\text{C}$ values (Fig. 4), however, these stations were located in the Oyashio region (Fig. 1b).

Surface ¹⁴C concentration in seawater is regulated mainly by the CO₂ 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 $\Delta^{14}\text{C}$ 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 $\Delta^{14}\text{C}$ 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 $\Delta^{14}\text{C}$ value being -55‰), however, the water profiles at St. 11 and 20 showed at 100 m and 200 m water depths increased $\Delta^{14}\text{C}$ 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 ^3H) 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

The analysis of ^{137}Cs , ^3H and ^{14}C in surface and water column samples collected in the western North Pacific Ocean after the Fukushima accident showed that the Fukushima impact has been mainly on ^{137}Cs in the marine environment. The ^{137}Cs concentrations observed in surface seawater of the western North Pacific Ocean were due to the Fukushima accident higher by about three orders of magnitude. On the other hand the ^3H levels were above the global fallout background only by about a factor of 6. The ^{14}C data suggest that the impact on radiocarbon levels in the western North Pacific Ocean was measurable, but only around 9% above the background. The water column data indicated that the Fukushima-derived radionuclides already in June 2011 reached depths down to around 200 m.

The ^{137}Cs inventory in the water column of the investigated western North Pacific Ocean affected by the Fukushima accident was estimated to be (2.7 ± 0.4) PBq, which can be regarded as a lower limit of the direct ^{137}Cs liquid discharges to the ocean. This lower value when compared with the recent estimation of the direct liquid release inventory of (3.6 ± 0.7) PBq (Aoyama et al., 2016b) may be due to the fact that the region under study did not cover the full area affected by the Fukushima liquid releases. On the basis of $^3\text{H}/^{137}\text{Cs}$ activity ratios in the water column profiles, the total ^3H inventory was estimated to be (0.3 ± 0.2) PBq.

Measured tritium and radiocarbon concentrations in the western North Pacific Ocean confirm that their distribution has been influenced by the Fukushima accident. The pre-Fukushima tritium and radiocarbon data, e.g. those previously gathered in the framework of the WOCE and WOMARS projects, do not represent anymore the global fallout distribution of these radionuclide in the western North Pacific Ocean.

Further studies of these radionuclides in the water column of the North Pacific Ocean are necessary for better estimation of the atmospheric input (via a wet and dry deposition) to their total inventory. The investigated radionuclides will be useful tracers during the next decades for studying pathways, ocean currents and time scales of deep and bottom water formation, for studying biogeochemical processes in the water column, circulation of water masses in the North Pacific Ocean, and transport of North Pacific waters to the South Pacific and the Indian Oceans.

Acknowledgments

The authors are thankful to Dr. S. Sheppard (Chief Editor of JER) for useful comments, and to prof. K. Hirose and dr. Y. Kumamoto for recent radiocesium data for the western North Pacific Ocean. We also thank Dr. K. O. Buesseler for provision of seawater samples collected during the A/V Ka'imikai-o-Kanaloa (KoK) cruise in the western North Pacific

Ocean, the Gordon and Betty Moore Foundation, and the Chemical Oceanography Program of the US National Science Foundation for funding the KoK expedition. The Bratislava group acknowledges a support provided by the EU Research and Development Operational Program (funded by the ERDF, Project No. 26240220004). The International Atomic Energy Agency is grateful to the Government of the Principality of Monaco for support provided to its Environment Laboratories.

References

- Aarkrog, A., Baxter, M.S., Bettencourt, A.O., Bojanowski, R., Bologna, A., Charmasson, S., Cunha, I., Delfanti, R., Duran, E., Holm, E., Jeffree, R., Livingston, H.D., Mahapanyawong, S., Nies, H., Osvath, I., Pingyu, L., Povinec, P.P., Sanchez, A., Smith, J.N., Swift, D., 1997. A comparison of doses from ^{137}Cs and ^{210}Po in marine food: A major international study. *J. Environ. Radioact.* 34, 69–90.
- Aoyama, M., Hirose, K., 2004. Artificial radionuclides database in the Pacific Ocean: HAM database. *ScientificWorldJournal* 4:200–215.
- Aoyama, M., Tsumune, D., Hamajima, Y., 2012a. Distribution of ^{137}Cs and ^{134}Cs in the North Pacific Ocean: impacts of the TEPCO Fukushima-Daiichi NPP accident. *J. Radioanal. Nucl. Chem.* 296, 535–539.
- Aoyama, M., Tsumune, D., Uematsu, M., Kondo, F., Hamajima, Y., 2012b. Temporal variation of ^{134}Cs and ^{137}Cs activities in surface water at stations along the coastline near the Fukushima Dai-ichi Nuclear Power Plant accident site, Japan. *Geochem. J.* 46, 321–325.
- Aoyama, M., Uematsu, M., Tsumune, D., Hamajima, Y., 2013. Surface pathway of radioactive plume of TEPCO Fukushima NPP1 released ^{134}Cs And ^{137}Cs . *Biogeosciences*, 10, 3067–3078.
- Aoyama, M., Hamajima, Y., Hult, M., Uematsu, M., 2016a. ^{134}Cs and ^{137}Cs in the North Pacific Ocean derived from the March 2011 TEPCO Fukushima Dai-ichi Nuclear Power Plant accident, Japan. Part one: surface pathway and vertical distributions. *J. Oceanography* 72, 53-65.
- Aoyama, M., Kajino, M., Tanaka, T.Y., Sekiyama, T.T., Tsumune, D., Tsubono, T., Hamajima, Y., Inomata, Y., Gamo, T., 2016b. ^{134}Cs and ^{137}Cs in the North Pacific Ocean derived from the TEPCO Fukushima Dai-ichi Nuclear Power Plant accident, Japan in March 2011: Part Two – Estimation of ^{134}Cs and ^{137}Cs inventories in the North Pacific Ocean. *J. Oceanography* 72, 67-76.
- Aramaki, T., Mizushima, T., Kuji, T., Povinec, P.P., Togawa, O., 2001. Distribution of radiocarbon in the southwestern North Pacific. *Radiocarbon* 43, 857-867.
- Aramaki, T., Togawa, O., Watanabe, S., Tsunogai, S., Taira, K., 2005. Temporal change of radiocarbon in the Western North Pacific. *JAERI Report*, Mutsu.
- Buesseler, K.O., Jayne, S.R., Fisher, N.S., Rypina, I.I., Baumann, H., Baumann, Z., Breier, C. F., Douglass, E.M., George, J., Macdonald, A.M., Miyamoto, H., Nishikawa, J., Pike, S.

614 M., Yoshida, S., 2012. Fukushima-derived radionuclides in the ocean and biota off Japan.
615 Proc. Nat. Acad. Sci. USA 109, 5984–5988.

616 Buzinny, M., Likhtarev, I., Los, I., Talerko, N., Tsigankov, N., 1998. ^{14}C analysis of annual
617 tree rings from the vicinity of the Chernobyl NPP. Radiocarbon 40, 373–379.

618 Chino, M., Nakayama, H., Nagai, H., Terada, H., Katata, G., Yamazawa, H., 2011.
619 Preliminary estimation of release amounts of ^{131}I and ^{137}Cs accidentally discharged from
620 the Fukushima Daiichi nuclear power plant into the atmosphere. J. Nucl. Sci. Technol.,
621 48, 1129–1134.

622 Chudý M., Povinec P., 1982. Radiocarbon production in CO_2 coolant of nuclear reactor.
623 Acta Physica Univ. Comen. 22, 127–131.

624 Dietze, H., Kriest, I. 2012. ^{137}Cs off Fukushima Dai-ichi, Japan: model based estimates of
625 dilution and fate. Ocean Sci. 8, 319–332.

626 Estournel, C., Bosc, E., Bocquet, M., Ulses, C., Marsaleix, P., Winiarek, V., Osvath, I.,
627 Nguyen, C., Duhaut, T., Lyard, F., Michaud, H., Auclair, F., 2012. Assessment of the
628 amount of Cesium-137 released into the Pacific Ocean after the Fukushima accident and
629 analysis of its dispersion in Japanese coastal waters. J. Geophys. Res. 117, C11014. 630
<http://dx.doi.org/10.1029/2012JC007933>.

631 Hernández-Ceballos, M.A., Hong, G.H., Lozano, R.L., Kim, Y.I., Lee, H.M., Kim, S.H., Yeh,
632 S.W., Bolívar, J.P., Baskaran, M., 2012. Tracking the complete revolution of surface
633 westerlies over Northern Hemisphere using radionuclides emitted from Fukushima, Sci.
634 Total Environ. 438, 80–85.

635 Hirose, K., 2012. 2011 Fukushima Dai-ichi nuclear power plant accident: summary of
636 regional radioactive deposition monitoring results. J. Environ. Radioact. 111, 13–17.

637 Honda, M., Aono, T., Aoyama, M., Hamajima, Y., Kawakami, H., Kitamura, M., Masumoto,
638 Y., Miyazawa, Y., Takigawa, M., Saino, T., 2012 Dispersion of artificial caesium-134
639 and -137 in the Western North Pacific one month after the Fukushima accident.
640 Geochem. J. 46, 1–9.

641 Hou, X.L., Povinec, P.P., Zhang, L. Y., Biddulph, D., Chang, C.-C., Fan, Y.K., Golser, R.,
642 Jeřkovský, M., Jull, A.J.T., Liu, Q., Shi, K. L., Steier, P., Zhou, W.J., 2013. Iodine-129 in
643 seawater offshore Fukushima: Distribution, speciation, sources, and budget. Environ. Sci.
644 Technol. 47, 3091–3098.

645 IAEA, 2005. Worldwide marine radioactivity studies (WOMARS). Radionuclide levels in
646 oceans and sea. IAEA-TECDOC-1429, Vienna, Austria.

647 Inoue, M., Kofuji, H., Nagao, S., Yamamoto, M., Hamajima, Y., Yoshida, K., Fujimoto, K.,
648 Takada, T., Isoda, Y., 2012, Lateral variation of ^{134}Cs and ^{137}Cs concentrations in surface
649 seawater in and around the Japan Sea after the Fukushima Dai-ichi Nuclear Power Plant
650 accident. J. Environ. Radioact. 109, 45–51.

651 Jean-Baptiste, P., Fourré, E., Dapoigny, A., Baumier, D., Baglan, N., Alanic, G., 2010. ^3He
652 mass spectrometry of very low-level measurement of organic tritium in environment
653 samples J. Environ. Radioact. 101, 185–189.

654 Kaeriyama, H., Ambe, D., Shimizu, Y., Fujimoto, K., Ono, T., Yonezaki, S., Kato, Y.,
655 Matsunaga, H., Minami, H., Nakatsuka, S., Watanabe, T. Direct observation of ^{134}Cs and

656 ^{137}Cs in surface seawater in the western and central North Pacific after the Fukushima Dai-
657 ichi nuclear power plant accident (2013) *Biogeosciences*, 10 (6), pp. 4287-4295.

658 Kameník, J., Dulaiova, H., Buesseler, K.O., Pike S.M., Šťastná, K., 2013. Cesium-134
659 and 137 activities in the central North Pacific Ocean after the Fukushima Dai-ichi 660
Nuclear Power Plant accident. *Biogeosciences* 10, 6045–6052.

661 Kawamura, H., Kobayashi, T., Furuno, A., In, T., Ishikawa, Y., Nakayama, T., Shima, S.,
662 Awaji, T., 2011. Preliminary numerical experiments on oceanic dispersion ^{131}I and ^{137}Cs
663 discharged into the ocean because of the Fukushima Dai-ichi nuclear power plant
664 disaster. *J. Nucl. Sci. Technol.* 48, 1349–1356.

665 Key, R., Quay, P., Jones, G., McNichol, A., von Reden, K., Schneider, R., 1996. WOCE
666 AMS radiocarbon I: Pacific Ocean results (P6, P16 and P17). *Radiocarbon* 38, 425-518.

667 Key, R., Quay, P., Schlosser, P., McNichol, A., von Reden, K., Schneider, R., Elder, K.,
668 Stuiver, M., Ostlund, H., 2002. WOCE radiocarbon IV: Pacific Ocean results; P10,
669 P13N, P14C, P18, P19 & S4P. *Radiocarbon* 44, 239-392.

670 Key, R.M., Kozyr, A., Sabine, C.L., Lee, K., Wanninkhof, R., Bullister, J.L., Feely, R.A.,
671 Millero, F.J., Mordy, C., Peng, T.-H., 2004. A global ocean carbon climatology: Results
672 from Global Data Analysis Project (GLODAP). *Global Biogeochem. Cy.* 18(4), GB4031.
673 <http://dx.doi.org/10.1029/2004GB002247>.

674 Kim, H.G., Kong, T.Y., Lee, G.J., Jeong, W.T., Kim, S.T., 2011. Analysis of metabolism and
675 effective half-life for radiation workers' tritium intake at pressurized heavy water
676 reactors. *Progress Nucl. Sci. Technol.* 1, 545-548.

677 Kirchner, G., Bossew, P., De Cort, M., 2012. Radioactivity from Fukushima Dai-ichi in air
678 over Europe; part 2: what can it tell us about the accident? *J. Environ. Radioact.* 114, 35-
679 40. <http://dx.doi.org/10.1016/j.jenvrad.2011.12.016>.

680 Kumamoto, Y., Murata, A., Saito, C., Honda, M., Kusakabe, M., 2002. Bomb radiocarbon
681 invasion into the northwestern North Pacific. *Deep Sea Res. II* 49, 5339–5351.

682 Kumamoto, Y., Murata, A., Kawano, T., Aoyama, M., 2013a. Fukushima-derived
683 radiocesium in the northwestern Pacific Ocean in February 2012. *Appl. Radiat. Isotopes*
684 81, 335–339.

685 Kumamoto, Y., Murata, A., Kawano, T., Watanabe, S., Fukasawa, M., 2013b. Decadal
686 changes in bomb-produced radiocarbon in the Pacific Ocean from the 1990s to 2000s.
687 *Radiocarbon* 55, 1641–1650.

688 Kumamoto, Y., Aoyama, M., Hamajima, Y., Aono, T., Kouketsu, S., Murata, A., Kawano, T.,
689 2014. Southward spreading of the Fukushima-derived radiocesium across the Kuroshio
690 Extension in the North Pacific. *Sci. Rep.* 4, 4276. <http://dx.doi.org/10.1038/srep04276>.

691 Kumamoto, Y., Aoyama, M., Hamajima, Y., Murata, A., Kawano, T., 2015. Impact of
692 Fukushima-derived radiocesium in the western North Pacific Ocean about ten months
693 after the Fukushima Dai-ichi nuclear power plant accident. *J. Environ. Radioact.* 140,
694 114-122.

695 Livingston, H.D., Povinec, P.P., 2000. Anthropogenic marine radioactivity. *Ocean and*
696 *Coastal Management* 43, 689-712.

697 Livingston, H.D., Povinec, P.P., 2002. A millennium perspective on the contribution of global
698 fallout radionuclides to ocean science. *Health Phys.* 82, 656-668.

699 Masson, O., Baeza, A., Bieringer, J., Brudecki, K., Bucci, S., Cappai, M., Carvalho, F.P.,
 700 Connan, O., Cosma, C., Dalheimer, A. Didier, D., Depuydt, G., De Geer, L.E., De
 Vismes, A., Gini, L., Groppi, F., Gudnason, K., Gurriaran, R., Hainz, D., Halldorsson, O.,
 702 Hammond, D., Hanley, O. Holey, K. Homoki, Zs., Ioannidou, A. , Isajenko, K.,
 703 Jankovick M., Katzlberger, C., Kettunen, M., Kierepko, R., Kontro, R. Kwakman, P.J.M.,
 704 Lecomte, M. , Leon Vintro, L. Leppänen, A.- P., Lind, B. , Lujaniene, G., Mc Ginnity, P.,
 705 Mc Mahon, C., Mala H., Manenti, S., Manolopoulou, M., Mattila, A., Mauring, A.,
 706 Mietelski, J.W., Møller, B.S., Nielsen, P., Nikolick J., Overwater, R.M.W., Palsson, S. E.,
 707 Papastefanou, C., Penev, I., Pham, M.K., Povinec, P.P., Ramebäck, H., Reis, M.C. ,
 708 Ringer, W., Rodriguez, A., Rulík, P., Saey, P.R.J. , Samsonov, V., Schlosser, C., Sgorbati,
 709 G. , Silobritiene, B. V., Söderström, C., Sogni, R., Solier, L., Sonck, M., Steinhauser, G.,
 710 Steinkopff, T. , Steinmann, P., Stoulos, S., Sykora, I., Todorovic, D., Tooloutalaie, N.,
 711 Tositti, L., Tschiersch, J., Ugron, A., Vagena, E., Vargas, A., Wershofen, H., Zhukova O.,
 712 2011. Tracking of airborne radionuclides from the damaged Fukushima Dai-Ichi nuclear
 713 reactors by European networks. *Environ. Sci. Technol.* 45, 7670-7677.
 714 Matsumoto, T., Maruoka, T., Shimoda, G., Obata, H., Kagi, H., Suzuki, K.; Yamamoto, K.,
 715 Mitsuguchi, T., Hagino, K., Tomioka, N., Sambandam, C., Brummer, D., Klaus, P.M.,
 716 Aggarwal, P., 2013. Tritium in Japanese precipitation following the March 2011
 717 Fukushima Daiichi nuclear plant accident. *Sci. Total Environ.* 445-446, 365-370.
 718 Miyazawa, Y., Masumoto, Y., Varlamov, S. M., Miyama, T., 2012a. Transport simulation of
 719 the radionuclide from the shelf to open ocean around Fukushima. *Continental Shelf Res.*
 720 50-51, 16-29.
 721 Molnár, M., Hajdas, I., Janovics, R., Rinyu, L., Synal, H., Veres, M., Wacker, L., 2013a. C-14
 722 analysis of groundwater down to the milliliter level. *Nucl. Instrum. Meth. Phys. Res. B*
 723 294, 573-576.
 724 Molnár, M., Janovics, R., Major, I., Orsovszki, J., Gönczi, R., Veres, M., Leonard, A.G.,
 725 Castle, S.M., Lange, T.E., Wacker, L., Hajdas, I., Jull, A.J.T., 2013b. Status report of the
 726 new AMS 14C sample preparation lab of the Hertelendi Laboratory of Environmental
 727 Studies (Debrecen, Hungary). *Radiocarbon* 55, 665-676.
 728 Morino, Y., Ohara, T., and Nishizawa, M., 2011. Atmospheric behavior, deposition, and
 729 budget of radioactive materials from the Fukushima Daiichi nuclear power plant in
 730 March 2011. *Geophys. Res. Lett.* 38, L00G11. <http://dx.doi.org/10.1029/2011GL048689>.
 731 Nakano, M., Povinec, P.P., 2003. Oceanic general circulation model for the assessment of the
 732 distribution of ¹³⁷Cs in the world ocean. *Deep-sea Res. II* 50, 2803-2816.
 733 Nakano, M., Povinec, P.P., 2012. Long-term simulations of the ¹³⁷Cs dispersion from the
 734 Fukushima accident in the world ocean. *J. Environ. Radioact.* 111, 109-115.
 735 Nishihara, K., Yamagishi, I., Yasuda, K., Ishimori, K., Tanaka, K., Kuno, T., Inada, S.,
 736 Gotoh, Y., 2012. Radionuclide release to stagnant water in Fukushima-1 Nuclear Power
 737 Plant. *Transactions of the Atomic Energy Society of Japan* 11, 13–19.
 738 Ostlund, G.H., Craig, H., Broecker, W.B., Spencer, D., 1987. *GEOSECS Atlantic, Pacific and*
 739 *Indian ocean expeditions, vol.7, Shorebased data and graphics*, National Science
 740 Foundation, Washington, D.C., 200 pp.

741 Palcsu, L., Major, Z., Köllő, Z., Papp, L., 2010. Using an ultrapure ^4He spike in tritium
742 measurements of environmental water samples by the ^3He -ingrowth method. *Rapid*
743 *Comm. Mass Spectr.* 24, 698-704.

744 Park, J.H., Hong, W. Park, G. Sung, K.S., Lee, K.H., Kim, Y.E., Kim, J.K., Choi, H.W., Kim,
745 G.D., Woo, H.J., Nakanishi, T., 2013. A comparison of distribution maps of $\Delta^{14}\text{C}$ in
746 2010 and 2011 in Korea. *Radiocarbon* 55, 841-847.

747 Pham, M. K., Betti, M., Povinec, P. P., Alifimov, V., Biddulph, D., Gastaud, J., Kieser, W. E.,
748 Lopez Gutierrez J. M., Possnert, G., Sanchez-Cabeza, J. A., Suzuki, T., 2010. Certified
749 reference material IAEA-418: ^{129}I in Mediterranean Sea water. *J. Radioanal. Nucl. Chem.*
750 286, 121–127.

751 Pham, M.K., Betti, M., Povinec, P.P., Benmansour, M., Bünger, V., Drefvelin, J., Engeler, C.,
752 Flemal, J.M., Gasco, C., Guillevic, J., Gurriaran, R., Groening, M., Happel, J.D.,
753 Herrmann, J., Klemola, S., Kloster, M., Kanisch, G., Leonard, K., Long, S., Nielsen, S.,
754 Oh, J.-S., Rieth, P.U., Östergren, I., Pettersson, H., Pinhao, N., Pujol, L., Sato, K.,
755 Schikowski, J., Varga, Z., Varti, V.P., Zheng, J., 2011. A certified reference material for
756 radionuclides in the water sample from Irish Sea (IAEA-443). *J. Radioanal. Nucl. Chem.*
757 288, 603–611.

758 Povinec, P., Chudý, M., Sýkora, I., Szarka, J., Pikna, M., Holý, K., 1988. Aerosol
759 radioactivity monitoring in Bratislava following the Chernobyl accident. *J. Radioanal.*
760 *Nucl. Chem. Lett.* 126, 467-478.

761 Povinec, P.P., Oregioni, B., Jull, A.J.T., Kieser, W.E., Zhao, X.-L., 2000. AMS measurements
762 of ^{14}C and ^{129}I in seawater around radioactive waste dump sites. *Nucl. Instrum. Methods*
763 *Phys. Res. B* 172, 672-678.

764 Povinec, P.P., Badie, C., Baeza, A., Barci-Funel, G., Bergan, T.D., Bojanowski, R., Burnett,
765 W., Eikenberg, J., Fifield, L.K., Serradell, V., Gastaud, J., Goroncy, I., Herrmann, J.,
766 Hotchkis, M.A.C., Ikaheimonen, T.K., Jakobson, E., Kalimbadjan, J., La Rosa, J.J., Lee,
767 S.-H., Liong Wee Kwong, L., Lueng, W.M., Nielsen, S.P., Nouredine, A., Pham, M.K.,
768 Rohou, J.-N., Sanchez-Cabeza, J.A., Suomela, J., Suplinska, M., Wyse, E., 2002.
769 Certified reference material for radionuclides in seawater IAEA-381 (Irish Sea water). *J.*
770 *Radioanal. Nucl. Chem.* 251, 369-374.

771 Povinec, P.P., Livingston, H.D., Shima, S., Aoyama, M., Gastaud, J., Goroncy, I., Hirose, K.,
772 Huyhn-Ngoc, L., Ikeuchi, Y., Ito, T., La Rosa, J., Liong Wee Kwong, L., Lee, S.-H.,
773 Moriya H., Mulsow, S., Oregioni, B., Pettersson, H., Togawa, O., 2003. IAEA'97
774 expedition to the NW Pacific Ocean – results of oceanographic and radionuclide
775 investigations of the water column. *Deep-Sea Res. II* 50, 2607-2638.

776 Povinec, P.P., 2004a. Developments in analytical technologies for marine radioactivity
777 studies. In: H.D. Livingston (Ed.) *Marine Radioactivity*. Elsevier, Amsterdam, 237-294.

778 Povinec, P.P., Comanducci, J.F., Levy-Palomo, I., 2004b. IAEA-MEL's underground
779 counting laboratory in Monaco – background characteristics of HPGe detectors with anti-
780 cosmic shielding. *Appl. Rad. Isotopes* 61, 85-93.

781 Povinec, P.P., K. Hirose, T. Honda, T. Ito, E.M. Scott, Togawa, O., 2004b. Spatial
782 distribution of ^3H , ^{90}Sr , ^{137}Cs and $^{239,240}\text{Pu}$ in surface waters of the Pacific and Indian
783 Oceans – GLOMARD database. *J. Environ. Radioact.* 76, 113-137.

Povinec, P.P., Aramaki, T., Burr, G.S., Jull, A.J.T., Liong Wee Kwong, L., Togawa, O.,
 2004c. Radiocarbon in the water column of the southwestern North Pacific Ocean – 24
 years after GEOSECS. *Radiocarbon* 46, 583-594.
 Povinec, P.P., Comanducci, J.F., Levy-Palomo, I., 2005a. IAEA-MEL's underground
 counting laboratory (CAVE) for the analysis of radionuclides in the environment at very
 low-levels. *J. Radioanal. Nucl. Chem.* 263, 441-445.
 Povinec, P. P., Aarkrog, A., Buesseler, K.O., Delfanti, R., Hirose, K., Hong, G.H., Ito, T.,
 Livingston, H.D., Nies, H., Noshkin, V.E., Shima, S., Togawa, O., 2005b. ^{90}Sr , ^{137}Cs and
 $^{239,240}\text{Pu}$ concentration surface water time series in the Pacific and Indian Oceans –
 WOMARS results. *J. Environ. Radioact.* 81, 63-87.
 Povinec, P.P., Scotto, P., Osvath, I., Ramadan, H. 2006. The Marine Information System
 (MARIS). In: *Isotopes in Environmental Studies*. IAEA, Vienna, 68–69.
 Povinec, P. P., Lee, S. H., Liong Wee Kwong, L., Oregioni, B., Jull, A. J. T., Kieser, W. E.,
 Morgenstern, U., Top, Z., 2010. Tritium, radiocarbon, ^{90}Sr and ^{129}I in the Pacific and
 Indian Oceans. *Nucl. Instrum. Methods Phys. Res. B* 268, 1214-1218.
 Povinec, P.P., Breier, R., Coppola, L., Groening, M., Jeandel, C., Jull, A.J.T., Kieser, W.E.,
 Top, Z., 2011. Tracing of water masses using a multi-isotope approach in the southern
 Indian Ocean. *Earth Planetary Sci. Lett.* 302, 14-26.
 Povinec, P.P., Hirose, K., Aoyama, M., 2012a. Radiostrontium in the western North Pacific:
 Characteristics, behavior, and the Fukushima impact. *Environ. Sci. Technol.* 46, 10356-
 10363.
 Povinec, P. P., Eriksson, M., Scholten, J., Betti, M., 2012b. Marine Radioactivity Analysis.
 In: M.F. L'Annunziata (Ed.) *Handbook on Radioactivity Analysis*. Academic Press,
 Amsterdam, 770-832.
 Povinec, P.P., Hirose, K., Aoyama, M., 2013a. Fukushima Accident: Radioactivity Impact on
 the Environment. Elsevier, New York, 382 pp.
 Povinec, P.P., Gera, M., Hirose, K., Lujanienė G., Nakano, M., Plastino, W., 2013b.
 Dispersion of Fukushima radionuclides in the global atmosphere and the ocean. *Appl.*
Rad. Isotopes 81, 383–392.
 Povinec, P.P., Sýkora, I., Gera, M., Holý, K., Brestáková, L., Kováčik, A., 2013c.
 Fukushima-derived radionuclides in ground-level air of Central Europe: a comparison
 with simulated forward and backward trajectories. *J Radioanal Nucl Chem* 295, 1171–
 1176. <http://dx.doi.org/10.1007/s10967-012-1943-3>.
 Povinec, P.P., Aoyama, M., Biddulph, D., Breier, R., Buesseler K., Chang, C.C., Golser, R.,
 Hou, X.L., Jeřkovský, M., Jull, A.J.T., Kaizer, J., Nakano, M., Nies, H., Palcsu, L., Papp,
 L., Pham, M.K., 10, Steier, P., Zhang, L.Y., 2013d. Cesium, iodine and tritium in NW
 Pacific waters – a comparison of the Fukushima impact with global fallout.
Biogeosciences 10, 5481–5496.
 Povinec, P.P., Ženišová, Z., Šivo, A., Ogrinc, N., Richtáriková, M., Breier, R., 2013e.
 Radiocarbon and stable isotopes as groundwater tracers in the Danube river basin of SW
 Slovakia. *Radiocarbon* 55, 1017-1028.
 Povinec, P.P., Hirose, K., 2015. Fukushima radionuclides in the NW Pacific, and assessment
 of doses for Japanese and world population from ingestion of seafood. *Sci. Rep.* 5, 9016.
<http://dx.doi.org/10.1038/srep09016>.

828 Ramzaev, V., Nikitin, A., Sevastyanov, A., Artemiev, G., Bruk, G., Ivanov, S., 2014.
 829 Shipboard determination of radiocesium in seawater after the Fukushima accident:
 830 Results from the 2011-2012 Russian expeditions to the Sea of Japan and western North
 831 Pacific Ocean. *J. Environ. Radioact.* 135, 13-24.
 832 Rinyu, L., Molnár, M., Major, I., Nagy, T., Veres, M., Kimák, Á., Wacker, L., Synal, H.A.,
 833 2013. Optimization of sealed tube graphitization method for environmental C-14 studies
 834 using MICADAS. *Nucl. Instrum. Meth. Phys. Res. B* 294, 270-275.
 835 Rypina, I.I., Jayne, S.R., Yoshida, S., Macdonald, A.M., Douglass, E., Buesseler, K., 2013.
 836 Short-term dispersal of Fukushima-derived radionuclides off Japan: modeling efforts and
 837 model-data intercomparison. *Biogeosciences* 10, 4973-4990.
 838 <http://dx.doi.org/10.5194/bg-10-4973-2013>, 2013.
 839 Steinhauser, G., 2014. Fukushima's Forgotten Radionuclides: A Review of the Understudied
 840 Radioactive Emissions. *Environ. Sci. Technol.* 48, 4649-4663.
 841 Stohl, A., Seibert, P., Wotawa, G., Arnold, D., Burkhart, J. F., Eckhardt, S., Tapia, C.,
 842 Vargas, A., Yasunari, T. J., 2012. Xenon-133 and caesium-137 releases into the
 843 atmosphere from the Fukushima Dai-ichi nuclear power plant: determination of the
 844 source term, atmospheric dispersion, and deposition. *Atmos. Chem. Phys.* 12, 2313–
 845 2343.
 846 Stuiver, M., Ostlund, H.G., 1983. GEOSECS Indian and Mediterranean radiocarbon.
 847 *Radiocarbon* 25, 1-29.
 848 Schwantes, J.M., Orton, C.R., Clark, R.A., 2012. Analysis of a nuclear accident: fission and
 849 activation product releases from the Fukushima Daiichi Nuclear Facility as remote
 850 indicators of source identification, extent of release, and state of damaged spent nuclear
 851 fuel. *Environ. Sci. Technol.* 46, 8621-8627.
 852 Tatebe, H., Yasuda, I., 2004. Oyashio Southward Intrusion and Cross-Gyre Transport Related
 853 to Diapycnal Upwelling in the Okhotsk Sea. *J. Phys. Oceanogr.* 34, 2327-2341.
 854 Tsumune, D., Tsubono, T., Aoyama, M., Hirose, K., 2012. Distribution of oceanic ¹³⁷Cs from
 855 the Fukushima Dai-ichi Nuclear Power Plant simulated numerically by a regional ocean
 856 model. *J. Environ. Radioact.* 111, 100–108.
 857 Tsumune, D., Tsubono, T., Aoyama, M., Uematsu, M., Misumi, K., Maeda, Y., Yoshida, Y.,
 858 Hayami, H., 2013. One-year, regional-scale simulation of ¹³⁷Cs radioactivity in the ocean
 859 following the Fukushima Daiichi Nuclear Power Plant accident. *Biogeosciences* 10,
 860 5601–5617.
 861 Tsunogai, S., Watanabe, S., Honda, M., Aramaki, T., 1995. North Pacific Intermediate Water
 862 studied chiefly with radiocarbon. *J. Oceanogr.* 51, 519–536.
 863 UNSCEAR, 2008. United Nations Scientific Committee on the Effects of Atomic Radiation:
 864 Sources and Effects of Ionizing Radiation. Report to the General Assembly. United
 865 Nations, New York, USA.
 866 Watanabe, Y.W., Watanabe, S., Tsunogai, S., 1991. Tritium in the northwestern North
 867 Pacific. *Journal of the Oceanographical Society of Japan* 47 (3), 80-93.
 868 Xu, S., Cook, G.T., Cresswell, A.J., Dunbar, E., Freeman, S.P.H.T., Hastie, H., Hou, X.,
 869 Jacobsson, P., Naysmith, P., Sanderson, D.C.W., 2015. Radiocarbon concentration in
 870 modern tree rings from Fukushima, Japan. *J. Environ. Radioact.* 146, 67-72.

871 Yoshida, N., Kanda, J., 2012. Tracking the Fukushima Radionuclides. Science 336, 1115-
872 1116.
873
874

875

876 **Table 1**877 Tritium, radiocarbon and radiocesium in seawater samples collected offshore Fukushima during the
878 KoK cruise.

879

| Station number | Position of the sampling station | Water depth (m) | ³ H (TU) | $\Delta^{14}\text{C}$ (‰) | 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 |

880

881

882

883

884

885

886

887

888

889

890

891

892

893

894

895

896

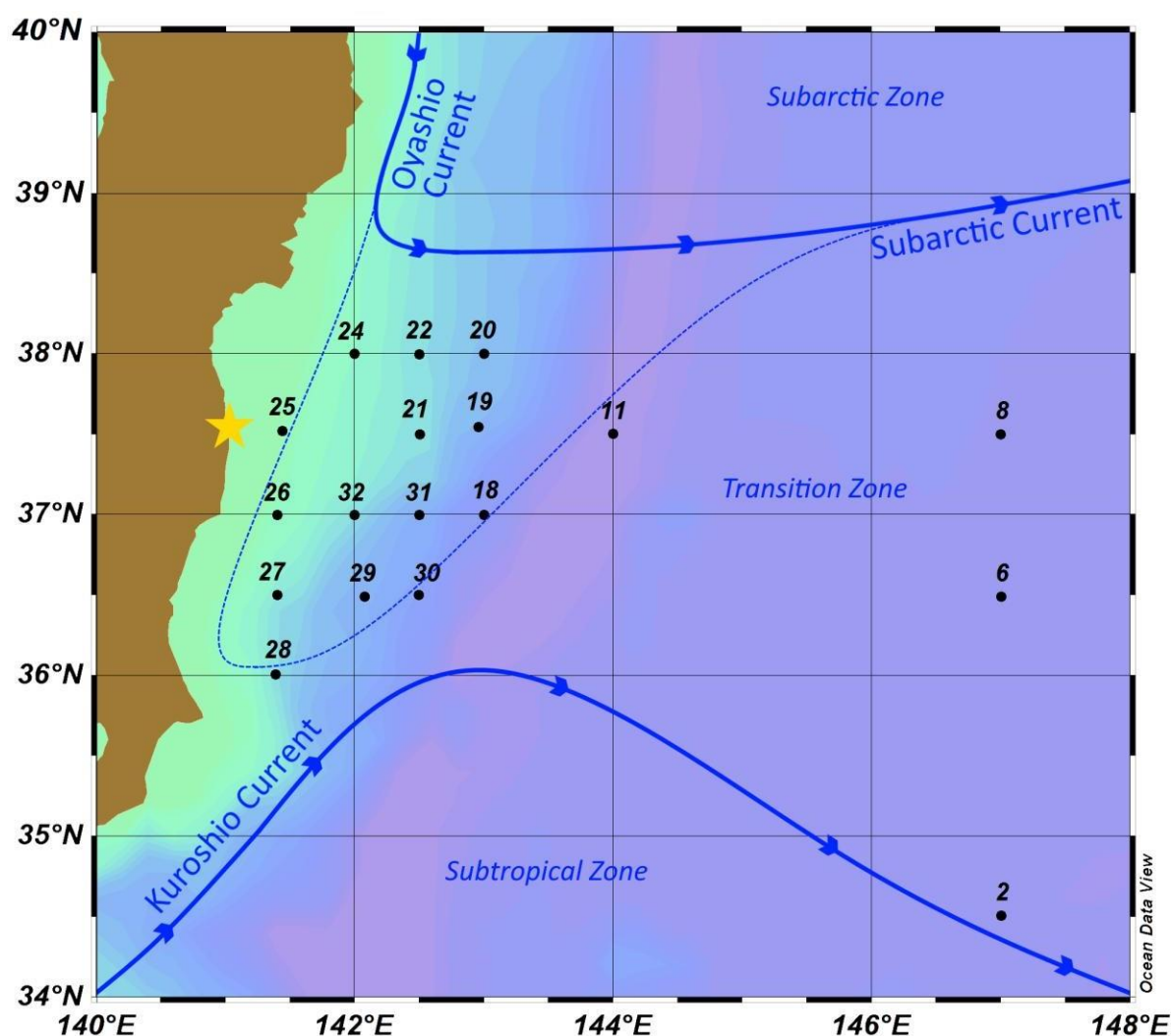
897

898

899

900

901



903

904 **Fig. 1a.** Locations of water sampling stations in the western North Pacific 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).

909

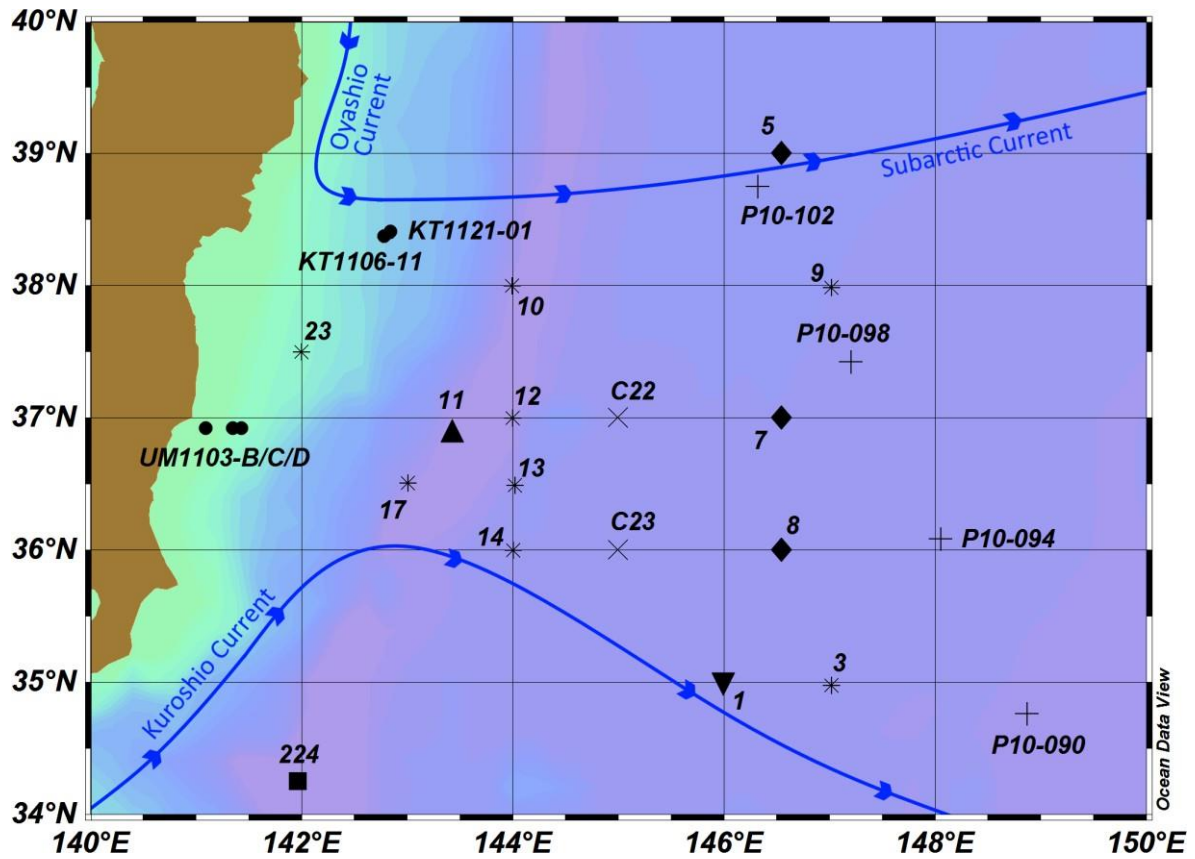


Fig. 1b. Locations of sampling stations used for data comparison: ● - UM11 and KT11 stations sampled for ^{137}Cs in 2011 (Aoyama et al., 2016a); * - selected KoK stations sampled for ^{137}Cs in June 2011 (Buesseler et al., 2012); × - KH93 stations sampled for ^{14}C in 1993 (Aramaki et al., 2005);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

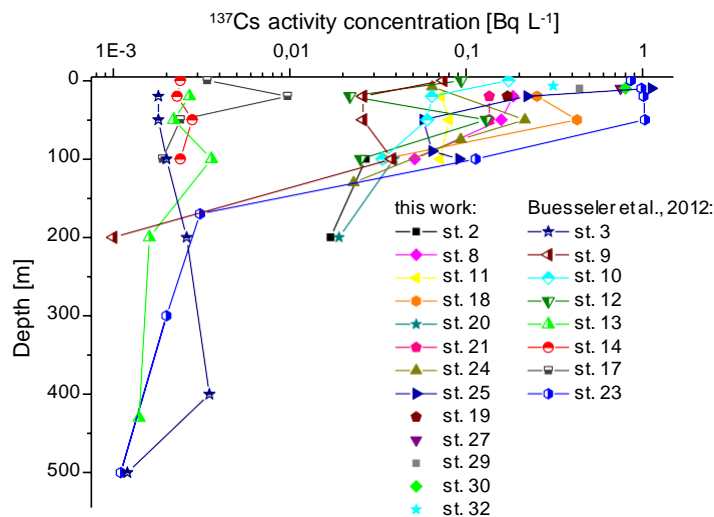


Fig. 2a. ^{137}Cs activity concentration in seawater samples collected offshore Fukushima NPP during the KoK cruise, compared with published data measured during the KoK cruise by Buesseler et al. (2012). For locations of the sampling stations see Figs. 1a, b.

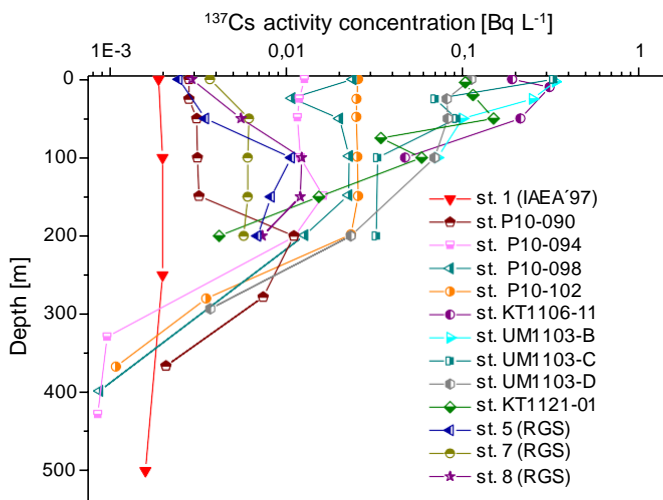


Fig. 2b. ^{137}Cs activity concentration in seawater samples collected offshore Fukushima NPP during the KoK cruise (decay corrected to June 2011), compared with published data measured during the IAEA'97 cruise (Povinec et al., 2003), the RGS cruise (Ramzaev et al., 2014), the P10 cruise (Kumamoto et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

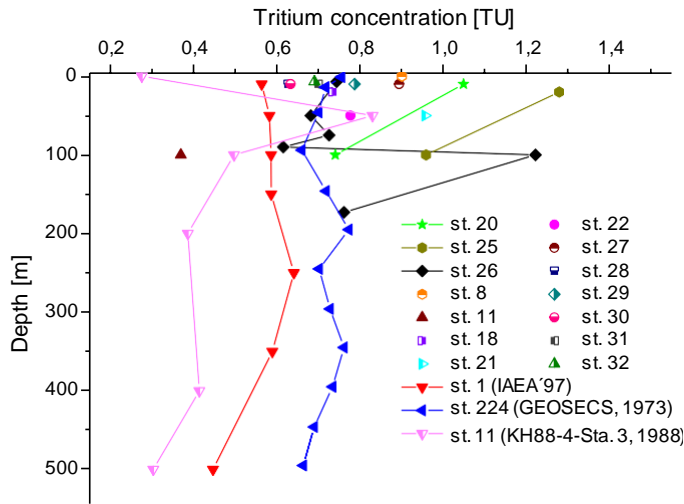


Fig. 3. Tritium concentration in seawater samples collected offshore Fukushima during the KoK cruise (decay corrected to June 2011), compared with the results from the GEOSECS (Ostlund et al., 1987), KH88 (Watanabe et al., 1991) and IAEA'97 (Povinec et al., 2003) expeditions. For locations of the sampling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

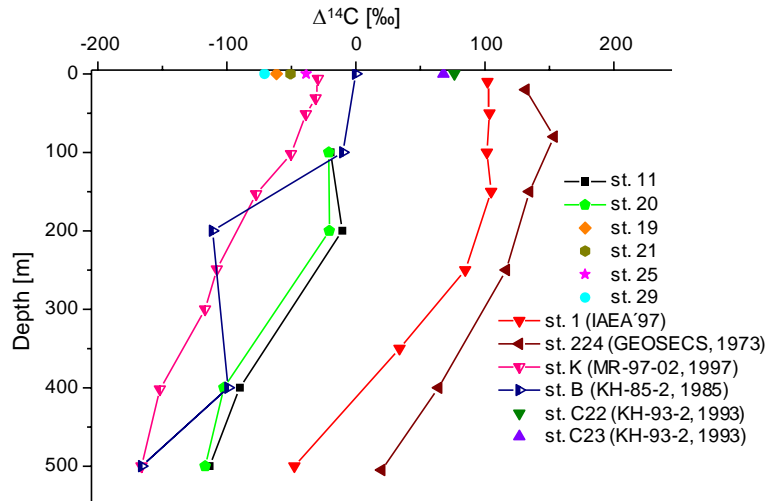


Fig. 4. Radiocarbon concentration in seawater samples collected offshore Fukushima during the KoK cruise (decay corrected to June 2011), compared with the results from the GEOSECS (Stuiver and Otlund, 1983), IAEA'97 (Povinec et al., 2004c), KH-85 (Tsunogai et al., 1995), KH93 (Aramaki et al., 2005) and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