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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

P.P. Povinec [a, *], L. Liong Wee Kwong [b], J. Kaizer [a], M. Molnár [c], H. Nies [b, 1], L. Palcsu [c], L. Papp [c], M.K. Pham [b], P. Jean-Baptiste [d]

[a] Department of Nuclear Physics and Biophysics, Faculty of Mathematics, Physics and Informatics, Comenius University, 84248 Bratislava, Slovakia
[b] Environment Laboratories, International Atomic Energy Agency, MC 98000 Monaco
[c] Institute for Nuclear Research (ATOMKI), 4026 Debrecen, Hungary
[d] LSCE, CEA-CNRS-UVSQ, CEA/Saclay, 91191 Gif-sur-Yvette, France

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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.

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., 2016a,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 only during the 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., 2016a), 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

* Corresponding author.
E-mail address: povinec@fmph.uniba.sk (P.P. Povinec).
[1] Present address: Bundesamt fuer Seeschifffahrt und Hydrographie, Federal Maritime and Hydrographic Agency, 22589 Hamburg, Germany.

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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., 2012a,b, 2013, 2016a; Buesseler et al., 2012; Hou et al., 2013; Povinec et al., 2012a,b, 2013a,d). We shall focus in this paper on Fukushima impact 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., 2012a,b, 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 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, γ)^{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'i-mikai-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, 2016a,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

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