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Tritium and radiocarbon in the western North Pacific waters: post-Fukushima situation

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ABSTRACT

Impact of the Fukushima Dai-ichi Nuclear Power Plant (FNPP1) accident on tritium (^{3}H) and radiocarbon (^{14}C) levels in the water column of the western North Pacific Ocean in winter 2012 is evaluated and compared with radiocesium ($^{134,137}Cs$) data collected for the same region. Tritium concentrations in surface seawater, varying between 0.4 and 2.0 TU (47.2–236 Bq m^{-3}), follow the Fukushima radiocesium trend, however, some differences in the vertical profiles were observed, namely in depths of 50–400 m. No correlation was visible in the case of ^{14}C, whose surface $\Delta^{14}C$ levels raised from negative values (about −40‰) in the northern part of transect, to positive values (~68‰) near the equator. Homogenously mixed ^{14}C levels in the subsurface layers were observed at all stations. Sixteen surface (from 30 in total) and 6 water profile (from 7) stations were affected by the Fukushima tritium. Surface and vertical profile data together with the calculated water column inventories indicate that the total amount of the FNPP1-derived tritium deposited to the western North Pacific Ocean was 0.7 ± 0.3 PBq. No clear impact of the Fukushima accident on ^{14}C levels in the western North Pacific was observed.

1. Introduction

On 11 March 2011, large tsunami waves, caused by the Tōhoku earthquake with the epicenter located approximately 130 km offshore of the eastern coast of Japan, stroke the Fukushima Dai-ichi Nuclear Power Plant (FNPP1), resulting in a series of severe mishaps and eventually in a release of large amounts of radioactive material to the atmosphere and seawater (Chino et al., 2011; Aoyama et al., 2013; Povinec et al., 2013a; Thakur et al., 2013). The government of Japan classified the event on the International Nuclear and Radiological Event Scale (INES) at the level 7, the same as it was in the case of the Chernobyl accident (IAEA, 2011). Due to western winds, prevailing at the time of the FNPP1 accident, the radionuclides emitted to the atmosphere were mostly transported over the North Pacific Ocean where about 80% of the released radioactivity was deposited by processes of dry and wet deposition, while the rest contaminated the Japanese land (Hirose, 2012), the Japan Sea (Inoue et al., 2012), the Atlantic Ocean and Europe (Masson et al., 2011; Morino et al., 2011; Stohl et al., 2012; Yoshida and Kanda, 2012; Povinec et al., 2013a). The total amount of the atmospheric emission was estimated to be 6.2–13.0 EBq, though almost all of it can be attributed to short-lived fission products ^{133}Xe (92–97%) and $^{131,133}I$ (1–5%; IAEA, 2015).

Direct discharges of liquid radioactive wastes were identified as the second relevant pathway of radionuclides from the damaged FNPP1 to the Pacific Ocean. These wastes mainly originated from emergency cooling of the three melted nuclear reactors by injection of huge amounts of sea- and fresh-water onto them. Several events occurred during March to May 2011 which resulted in significant intentional and unintentional releases of radioactive waters directly to the ocean. However, even after this period, highly contaminated stagnant water has continued to enter the ocean, though with a much lower rate. During heavy rains, runoffs of radionuclides from the areas around the damaged reactors, transported radioactive waters from the reactor basements and trenches, which has also been considered as the likely source of the persisting releases (IAEA, 2015). Even though the direct discharges affected only the coastal region offshore Fukushima, due to the nature of the Pacific Ocean and its currents, diluted levels of radionuclides were spread over long distances (Nakano and Povinec,

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2012; Tsumune et al., 2013; Kaeriyama et al., 2014; Kumamoto et al., 2015; Aoyama et al., 2016a). The elevated concentrations of radionuclides in the FNPP1 coastal area have been discussed in several papers (e.g., Buesseler et al., 2012; Povinec et al., 2013b, 2017; Aoyama et al., 2011, 2016a).

Radiocesium (^{134}Cs and ^{137}Cs) has been by far the most studied radioelement after the FNPP1 accident which may be reasoned by the fact that the release rates were relatively high, a suitable measuring techniques were available, and the decay-corrected ^{134}Cs/^{137}Cs activity ratio, which was equal to almost one in the FNPP1 reactor core inventories at the time of the meltdown (Povinec et al., 2013a), has been effectively exploited to trace the origin of radioactive contamination, either in air or water (Thakur et al., 2012; Steinhauser et al., 2013; Povinec et al., 2013c,d). Due to a relatively short half-life of ^{134}Cs ($T_{1/2}$ = 2.07 y), the ^{134}Cs/^{137}Cs signal has been slowly but surely fading. Moreover, because of its unique properties (fairly long half-life of $T_{1/2}$ = 30.05 y, very good solubility in seawater, reasonably detectable levels even on mBq scale), ^{137}Cs has been recognized as an excellent tracer, especially for investigation of different oceanic processes (Cochran et al., 1995; Ito et al., 2003; Tsumune et al., 2003; Aoyama et al., 2011).

In the last century, the inventory of radiocesium in the North Pacific Ocean was increased mainly due to atmospheric nuclear weapons testing in the Northern Hemisphere, the Chernobyl accident and operation of nuclear fuel reprocessing plants (Aoyama and Hirose, 2004; Aoyama et al., 2006; Povinec et al., 2013a). However, the FNPP1 accident logically brought up the need of re-validation of the established data. First estimates on releases of radiocesium from the damaged FNPP1 were quite inconsistent, though with more and more determinations of Fukushima-derived radiocesium, calculations of the amount of ^{137}Cs deposited to the ocean have improved. Aoyama et al. (2016b) have recently reviewed already published estimates of released radiocesium concluding that the combined input of ^{137}Cs to the North Pacific (deposition from the atmosphere plus direct discharges) was in the range of 15–18 PBq.

Another advantageous aspect of radiocesium is that it can be used as a proxy for estimation of releases of additional radionuclides present in the boiling-water reactor (BWR) cores of the FNPP1. Hence, with better understanding of the source term of radiocesium and its distribution in the North Pacific it may be possible to study the fate of other radionuclides of interest, such as ^{3}H, ^{14}C, ^{90}Sr, ^{129}I and others (Hou et al., 2013; Povinec et al., 2013b, 2017).

Tritium (^{3}H; $T_{1/2}$ = 12.33 y) is a pure beta-emitter (E_{max} = 18.6 keV) which enters the environment in the form of water molecules (mostly as HTO), thus representing an ideal tracer for oceanographic observations (e.g., Schlosser et al., 1999; Povinec et al., 2010, 2011). Worldwide steady-stay inventory of natural ^{3}H (2.2 EBq; UNSCEAR, 2008) was severely disturbed during the nuclear weapons testing era when around 113 EBq was deposited into the ocean, from which some 5.4 EBq has still not decayed (IAEA, 2005). Another important source of anthropogenic tritium, specific for the western North Pacific, are nuclear fuel reprocessing plants located on the eastern coast of Japan from which relatively large amounts of liquid radioactive waste were directly discharged into the sea. According to Anzai et al. (2008), approximately 1.8 PBq of ^{3}H was released from the Rokkasho plant (41.0°N, 141.3°E) during the active test period in 2006–2008. This led to temporary increased tritium concentration in coastal seawater up to 39 TU, though the majority of the measurements showed results around or well below 4 TU, and one year after the tests only slightly above 1 TU (Muranaka et al., 2011). Furthermore, the total controlled releases from the Tokai facility (36.4°N, 140.6°E), which was ceased in 2007 after thirty years of operation, were declared to be roughly 4.5 PBq, with ~90% of the ^{3}H activity discharged before 1997 (Kokubun et al., 2011).

Tritium in a FNPP1-type BWR reactor was dominantly produced by ternary fission of ^{238}U and ^{239}Pu, and to a much lesser extent, by neutron capture on deuterium, and in the reactions with ^{7}Li(n,αn)^{3}H and ^{10}B(n,2α)^{3}H, with an annual production rate of approximately 1 PBq GW_e^{-1} (Hou, 2005; TEPCO, 2014). According to calculations by Nishihara et al. (2012), three damaged reactor cores contained about 3.4 PBq of tritium, however, only part of this total amount could be directly released to the ocean through stagnant water discharges. The ^{3}H release ratio from core to stagnant water was estimated to be about 46% (Povinec et al., 2017). From measurements of ^{3}H concentration in precipitation samples collected south-west off the FNPP1 site, it was estimated that tritium concentrations in the air immediately after the accident reached in the vicinity of the FNPP1 about 1.5 kBq m^{-3} (13 TU) (Matsumoto et al., 2013), and part of this tritium was precipitated over the ocean. Based on the limited data set covering only the coastal region, Povinec et al. (2017) estimated that 0.1–0.5 PBq of ^{3}H was deposited and/or discharged in the coastal region offshore Fukushima.

While there is at least some information about tritium source terms and activity measurements in connection with the FNPP1 accident, unfortunately, ^{14}C ($T_{1/2}$ = 5730 y) has been even less explored. As a cosmogenic radionuclide, natural ^{14}C is almost exclusively produced in the lower stratosphere and the upper troposphere by interaction of cosmic rays with nitrogen and oxygen atoms, where it rapidly oxidizes first to carbon monoxide, and then slowly to CO_2; it decays to stable ^{14}N by the emission of beta-particles (E_{max} = 156 keV). From the total inventory originating from global fallout and nuclear reprocessing plants, around 132 PBq was deposited into the ocean (Povinec et al., 2000, 2001), which is lower by one order of magnitude if compared to the ^{14}C natural inventory in the marine environment (6.9 EBq; Key et al., 2004).

In nuclear reactors, radiocarbon is mainly produced in the reactions ^{14}N(n,p)^{14}C and ^{17}O(n,α)^{14}C in the fuel, and on impurities of the fuel rods, coolant and moderator (Chudý and Povinec, 1982). However, because of the way the fuel rods for the FNPP1 reactors were fabricated (evacuation and consequent backfilling with helium, thus removing most of the air composed of nitrogen and oxygen), radiocarbon could only be produced in a noticeable amount on aforementioned impurities (Steinhauser, 2014). Without considering the previous statement, Xu et al. (2016b) calculated that the total ^{14}C inventory of 1.0–1.6 TBq might have been accumulated in three FNPP1 reactors, though actual releases of radiocarbon have not been discussed in literature at all. Determination of ^{14}C concentration in seawater samples from the Fukushima coastal area from June 2011 showed only a small impact of the FNPP1accident on the investigated region (6–9% increase above the background level; Povinec et al., 2017).

In this paper, we report ^{3}H and ^{14}C concentrations which were determined in the western North Pacific between 42°N and 4°S, approximately along the 149°E meridional line about ten months after the FNPP1 accident. To identify possible ^{3}H and ^{14}C signals from the damaged FNNP1 units, we shall compare our results with the radiocesium data from the same sample set reported by Kumamoto et al. (2015), who discussed the influence of the FNPP1 accident on the bomb-derived radiocesium levels in seawater. From this comparison, and from the recent estimate of the radiocesium activity released to the North Pacific (Aoyama et al., 2016b), we shall estimate total releases of tritium and radiocarbon to the North Pacific Ocean. As both ^{3}H and ^{14}C have been often used for tracing different ocean and ocean-atmosphere processes, it is crucial to evaluate changes in their concentrations in the North Pacific Ocean after the FNPP1 accident, so all future oceanographic or climate change studies will be based on correct data sets.

2. Samples and methods

2.1. Seawater sampling

Seawater samples for ^{3}H and ^{14}C measurements with the volume of 1 L were collected during the cruise MR11-08 of the Research Vessel *Mirai*, which was directed from the north (42°N) of the western North