Post-Fukushima concentrations of tritium and radiocarbon in the western North Pacific Ocean

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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. 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. According to calculations 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, while the total ^{14}C inventory of 1.0-1.6 TBq might have been accumulated in three FNPP1 reactors.

2. SAMPLING AND MEASUREMENTS

Seawater samples for ^{3}H and ^{14}C measurements with the volume of 1 L were collected during the cruise, which was directed from the north (42°N) of the western North Pacific towards the south (4°S), approximately following the 149°E meridian (Fig. 1.), from December 2011 to February 2012. The radiocesium data from the same sample set were reported by Kumamoto et al. (2015). Concentration of tritium in seawater samples was determined using the ^{3}He in-growth mass spectrometry method (Palcsu et al., 2010). After distilation, the water samples were stored for several months to produce ^{3}He atoms from the decay of tritium. The tritiogenic helium samples were then admitted into a system where the helium fraction was separated and purified from any contaminants and spiked with a small amount of ultra-pure ^{4}He prior the measurements. Analyses of radiocarbon in the dissolved inorganic carbon (DIC) fraction were carried out by accelerator mass spectrometry (AMS). The DIC fraction was extracted from samples and cleaned CO_2 was graphitized. Radiocarbon content in the graphite targets was determined using the MICADAS (Molnár et al., 2013).

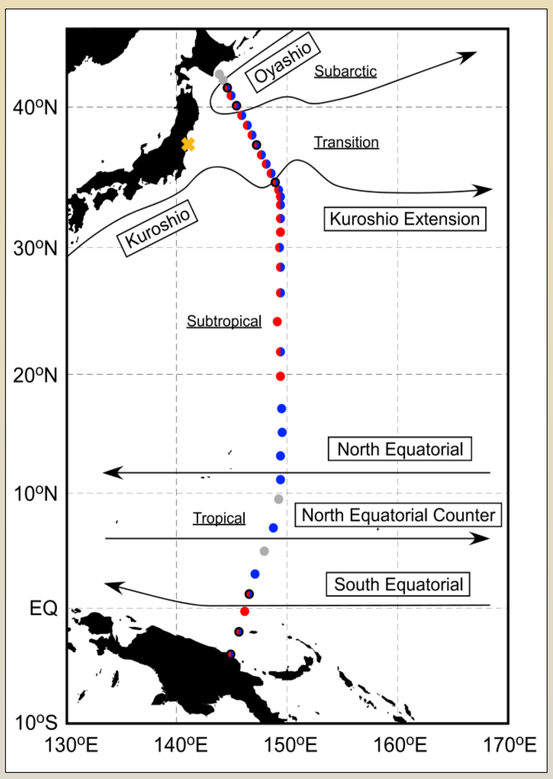

Fig. 1. Locations of the sampling sites visited during the winter 2012 cruise in the western North Pacific Ocean. Blue and red color denotes stations where tritium and radiocarbon was determined, respectively. The FNPP1 site is marked as a yellow cross (Kaizer et al., 2018).

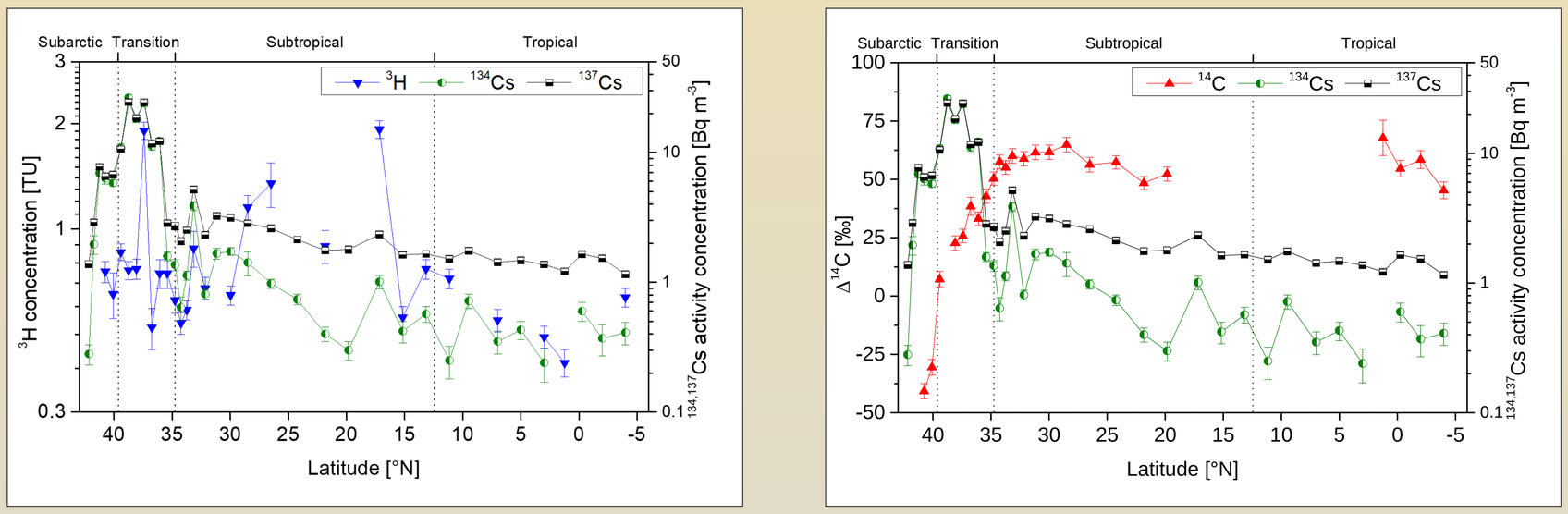

Fig. 2. Latitudinal distribution of ^{3}H concentration (TU; left), $\Delta^{14}C$ (‰; right) and radiocesium activity concentration (Bq m^{-3}) in surface waters of the western North Pacific Ocean roughly along the 149°E meridional line.

3. RESULTS AND DICUSSION

Tritium in surface seawater, varying between 0.4-2.0 TU (47.2-236 Bq m^{-3}), seemed to fairly follow the FNPP1-derived radiocesium signal, though we observed some differences in the vertical mixing as the shapes of the profiles in the 50-400 m water depths were quite different. Surprisingly, increased ^{3}H levels were not only found in the regions closest to the FNPP1 (37-40°N) but also in very distant regions (~1°N) with no subsurface radiocesium detected, suggesting an important role of precipitation in spreading of the FNPP1-derived tritium. The total amount of ^{3}H released to the western North Pacific Ocean and calculated water column inventories from surface and vertical profile data were estimated to be 0.7 ± 0.3 PBq.

No apparent correlation between radiocarbon levels and radiocesium activity concentrations was found in surface seawater samples. Going from the north to the south, the ^{14}C concentrations changed from quite negative (about -40‰) to positive (~68‰) values near the equator. Low $\Delta^{14}C$ levels observed around the 39-40°N latitudes might be associated with the Oyashio current, bringing subarctic waters with low $\Delta^{14}C$ values to the region. Regarding vertical distribution, radiocarbon in the surface mixed layers at all stations was almost uniformly mixed, similarly to radiocesium. The southern-most area showed by about 10‰ higher $\Delta^{14}C$ values in the surface mixed layer than the rest of the region The ^{14}C data did not show a clear impact of the FNPP1 accident on the radiocarbon levels, therefore we were not able to estimate ^{14}C release rates to the western North Pacific Ocean.

Besides the evaluation of connection with the FNPP1-derived radiocesium and influence of the accident on their concentrations in the western North Pacific Ocean, tritium and radiocarbon can be used to study different oceanographic processes, pathways, ocean currents and time scales of deep and bottom water formation.

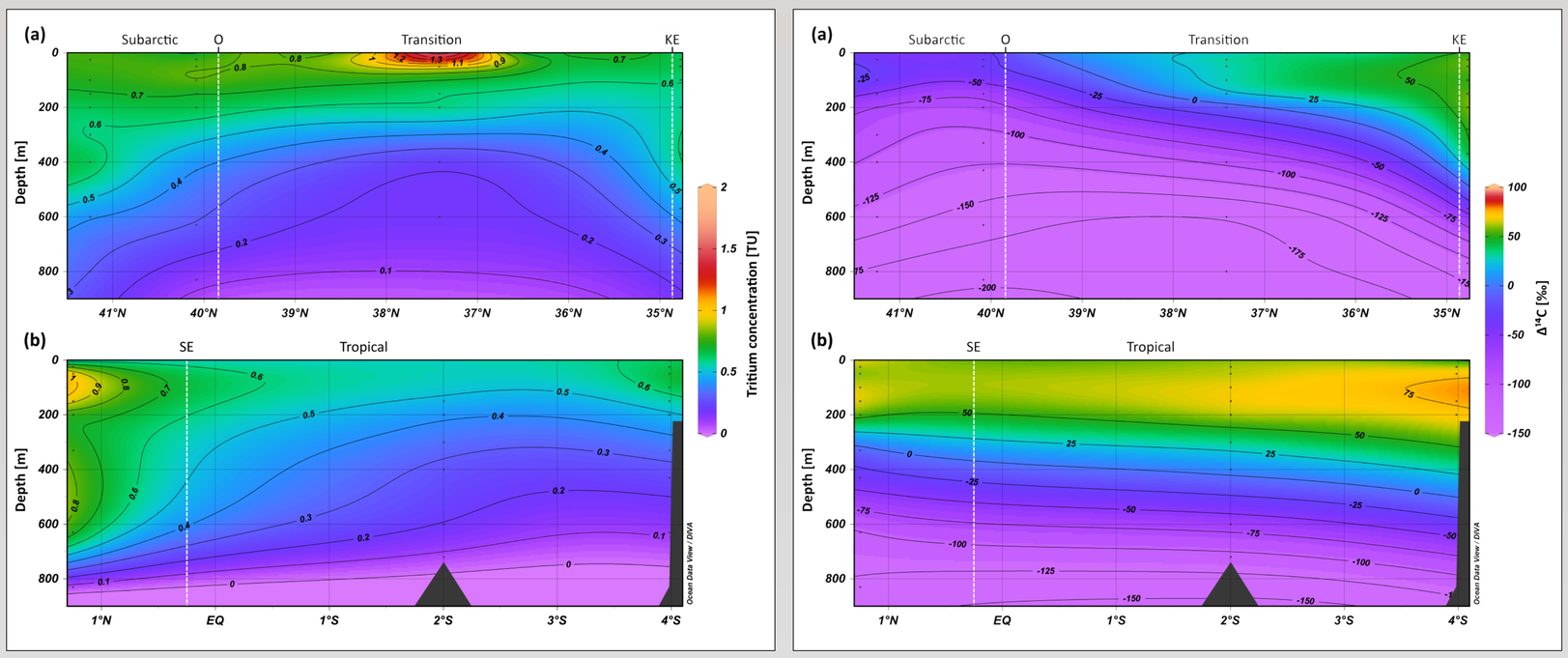

Fig. 3. Vertical distribution of tritium (TU; left) and radiocarbon ($\Delta^{14}C$ in ‰; right) in the (a) subarctic and transition zone (34.8-41.5°N) and (b) tropical region (4.1°S-1.3°N) along the 149°E meridian of the western North Pacific Ocean.

Rerferences

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