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Plutonium and ^{137}Cs in surface water of the South Pacific Ocean

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Abstract

The present plutonium and ^{137}Cs concentrations in South Pacific Ocean surface waters were determined. The water samples were collected in the South Pacific mid-latitude region (32.5 °S) during the BEAGLE expedition conducted in 2003–04 by JAMSTEC. ^{239,240}Pu concentrations in surface seawater of the South Pacific were in the range of 0.5 to 4.1 mBq m^{-3}, whereas ^{137}Cs concentrations ranged from 0.07 to 1.7 Bq m^{-3}. The observed ^{239,240}Pu and ^{137}Cs concentrations in the South Pacific were almost of the same level as those in the North Pacific subtropical gyre. The surface ^{239,240}Pu in the South Pacific subtropical gyre showed larger spatial variations than ^{137}Cs, as it may be affected by physical and biogeochemical processes. The ^{239,240}Pu/^{137}Cs activity ratios, which reflect biogeochemical processes in seawater, were generally smaller than that observed in global fallout, except for the most eastern station. The ^{239,240}Pu/^{137}Cs ratios in the South Pacific tend to be higher than that in the North Pacific. The relationships between anthropogenic radionuclides and oceanographic parameters such as salinity and nutrients were examined. The ^{137}Cs concentrations in the western South Pacific (the Tasman Sea) and the eastern South Pacific were negatively correlated with the phosphate concentration, whereas there is no correlation between the ^{137}Cs and nutrients concentrations in the South Pacific subtropical gyre. The mutual relationships between anthropogenic radionuclides and oceanographic parameters are important for better understanding of transport processes of anthropogenic radionuclides and their fate in the South Pacific.

Keywords: Plutonium; ^{137}Cs; ^{239,240}Pu/^{137}Cs activity ratio; South Pacific; Nutrients; Temporal change

1. Introduction

Long-lived anthropogenic radionuclides (plutonium isotopes and ^{137}Cs) in seawater of the South Pacific were introduced on the ocean surface by global fallout due to atmospheric nuclear weapons testing, from which the major fallout occurred in the mid 1960s. Global fallout amounts of anthropogenic radionuclides in the Southern Hemisphere were about one third or less than in the Northern Hemisphere (UNSCEAR, 2000; Aoyama et al., 2006a). Anthropogenic radionuclides (mostly plutonium) were also injected into seawater by close-in fallout from French nuclear explosions conducted at the French Polynesia (Chiappini et al., 1999).

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As a result, the South Pacific waters have been widely contaminated by anthropogenic radionuclides (Hamilton et al., 1996; Hirose and Aoyama, 2003; Povinec et al, 2005).

In order to have better understanding of present levels of anthropogenic radionuclides in the marine environment, it is necessary to determine their concentrations in seawater over the wide areas of the South Pacific Ocean. However, the available data for the South Pacific are very sparse when compared with that in the Northern Hemisphere seas (Aoyama and Hirose, 2004; Hirose and Aoyama, 2003; Povinec et al., 2004).

In 2003–2004, JAMSTEC conducted a 7-month round the world Blue Earth Global Expedition (BEAGLE), visiting the South Pacific (winter), Atlantic (late spring) and Indian (summer) Oceans (Uchida and Fukasawa, 2005). This cruise provided rare opportunity to collect a lot of water samples for radioactivity measurements in Southern Hemisphere oceanic waters, as did the most precious hydrographic data set to investigate an ocean response to climate change.

At present we can determine concentrations of anthropogenic radionuclides (^{239,240}Pu and ^{137}Cs) in small volume seawater samples thanks to recent developments in radioanalytical techniques which have sensitivities of more than one order of magnitude better than before (Aoyama et al., 2000; Hirose et al., 2005; Kim et al., 2002). Therefore, we can present here a comprehensive data set on ^{239,240}Pu and ^{137}Cs concentrations in seawater, which are also useful as oceanographic tracers.

In this paper, we describe geographic distributions and temporal trends of ^{239,240}Pu and ^{137}Cs concentrations, together with oceanographic parameters such as salinity and nutrients, in surface waters in the mid-latitude region of the South Pacific, and compare their distribution in surface waters of the North Pacific.

2. Sampling and methods

Surface water samples were collected using a submersible pump at about 5 m depth during the BEAGLE cruise on board of the R/V Mirai organized by Japan Agency for Marine-Earth Science and Technology (JAMSTEC). Large volume (80 L) surface seawater samples were collected in the South Pacific at 50 stations (Fig. 1). The sampling spacing was about 2.5°. All water samples were filtered through a fine membrane filter (Millipore HA, 0.45 μm pore size) immediately after sampling. Filtered water samples were transported to the Meteorological Research Institute (MRI) and subjected to ^{137}Cs and plutonium analyses. Small volume samples (less than 10 L) were transported to the Korean Institute of Nuclear Safety (KINS) for ICPMS (Inductively Coupled Plasma Mass Spectrometry) analysis of plutonium.

^{137}Cs was concentrated in 20 L seawater samples by adsorption onto AMP (ammonium molybdophosphate) using a method described in detail elsewhere (Aoyama et al., 2000; Hirose et al., 2005). ^{137}Cs activities were determined by γ-spectrometry with high efficiency HPGe detectors (Hirose et al., 2005). Plutonium for α-spectrometry measurements was concentrated by co-

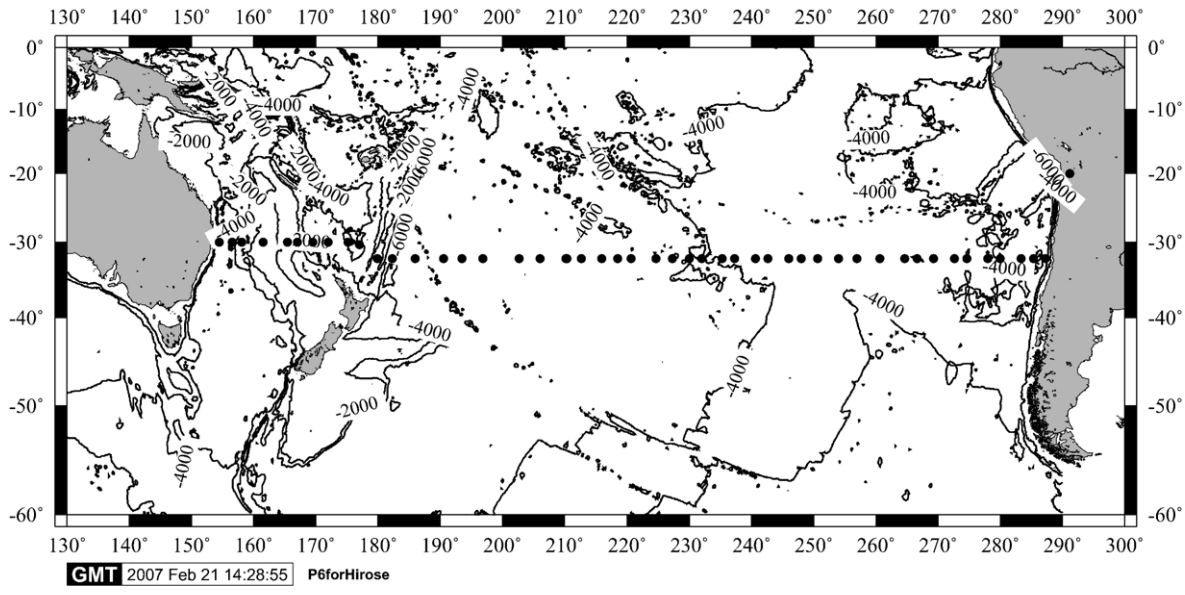

Fig. 1. Sampling locations in the South Pacific.