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Radioactive status of seawater and its assessment in the northeast South China Sea and the Luzon Strait and its adjacent areas from 2011 to 2014

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ABSTRACT

To understand the impact of the Fukushima nuclear accident (FNA), ^{137}Cs, ^{134}Cs, ^{90}Sr, and gross beta were analyzed in the northeast South China Sea (NSCS), the Luzon Strait (LS) and its adjacent areas. ^{137}Cs, ^{90}Sr, and gross beta values in the NSCS were similar to those prior to the FNA. ^{90}Sr and ^{137}Cs in the LS and its adjacent areas were consistent with those in the NSCS. The high ^{137}Cs-peak values occurred at depth of 150 m whereas the high ^{90}Sr-peak values occurred at depth of 0.5 m. The ^{137}Cs and gross beta mean values in Cruise I were higher than those in Cruise II whereas the ^{90}Sr mean value was just the reverse. ^{134}Cs in all seawater were below the minimum detectable activity. The past and present data since the 1970s suggested ^{137}Cs and ^{90}Sr in the study areas still originated from global fallout and the FNA influence were negligible.

1. Introduction

The Tohoku earthquake and the subsequent tsunami on 11 March 2011 damaged the Fukushima Dai-ichi nuclear power plant (FNA) on the east coast of Japan. Significant amounts of radionuclides (i.e., ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{129}I, ^{110m}Ag, ^{60}Co, and so on) were released to the atmosphere and, by direct discharge or leakage, to the ocean (Kamenik et al., 2013; Men et al., 2015). The FNA was one of the biggest environmental disasters in recent years. It is estimated that nearly 12–15 PBq of ^{137}Cs (observed ^{134}Cs to ^{137}Cs activity ratio at the time of the accident was close to one with extremely uniform ^{134}Cs concentrations) was released by the FNA resulting in atmospheric deposition. Additionally, another 3.5 ± 0.7 PBq was added by direct discharge which increased the total ^{137}Cs inventory in the North Pacific Ocean by 22–27% (Stan-Sion et al., 2015; Aoyama et al., 2016a, b). The Japanese government set a limit on radiation levels in the seafood and began screening fish. India and China recently banned imports of food products from certain areas of Japan and the US Food and Drug Administration began conducting its own radiation screens on seafood imports from Japan (Reardon, 2011). The concentrations of ^{137}Cs and ^{90}Sr in the squid sampled from the west Pacific waters in June 2012 were 11 and 2.7 times higher, respectively than those before the FNA. Additionally, the concentrations of ^{110m}Ag were also found to have increased (State Oceanic Administration (SOA) of P.R. China, 2013). Therefore, understanding the fate, transport, and ecological consequences of long-lived radionuclides in the oceanic environment is of paramount importance. In order to understand the influence of FNA generated radioactive pollutants on the marine environment, the risks to public health and marine biota have been studied and assessed (Buesseler et al., 2011, 2012; Buesseler, 2012; Bailly du Bois et al., 2012; Madigan et al., 2012; Zheng et al., 2012; Fisher et al., 2013; Lin et al., 2015; Tateda et al., 2017; Povinec et al., 2013).

To understand their impact on in the northeast South China Sea (NSCS) as well as the Luzon Strait (LS) and its adjacent areas from the FNA, many radioactive monitoring campaigns have been executed by the State Oceanic Administration of China from 2011 to 2014. It was very expensive and difficult to perform the monitoring work. There were seldom large-scale monitoring of the radioactive status in the NSCS, the LS and its adjacent areas more than three year after the FNA. This paper reports the concentrations of ^{137}Cs, ^{134}Cs, ^{90}Sr, and gross beta radioactivity in seawater sampled from the surface (0.5 m) and the medium-depth layers (≤600 m) of the NSCS, the LS and its adjacent sea areas from 13 March 2011 to 31 December 2014. Thus, all the data in this paper are important and provide essential information needed to assess the impacts (the amounts, distribution, transport, and ecological consequences) of the radioactive pollutants released from the FNA and also provide a baseline for future comparisons.

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2. Materials and methods

2.1. Sampling

After the FNA, in order to obtain environmental information regarding radioactivity in the South China Sea as well as to understand the consequent influence and risk, radioactive monitoring campaigns sampling only the surface water (0.5 m depth) were promptly conducted by the SOA of P.R. China since 13 March 2011. A long-term Station LS6, was repeatedly sampled till 31 December 2014. Surface seawater was sampled every three, seven or fifteen days followed by transport to the laboratory to conduct further analyses with respect to tracing radioactive influence from the FNA. The sampling details are shown in Fig. 1.

In addition, as shown in Fig. 2, two large and special cruises, i.e., Cruise I (11 May–3 June 2014) and Cruise II (25 September–18 October 2014) were implemented in the Luzon Strait and its adjacent sea areas. Seawater was sampled from three layers at depths of 0.5 m, 150 m, and 600 m. Seawater from two layers at depths of 0.5 m and 40 m in Station LS1 was sampled due to the limited depth of 46 m. No seawater at depth of 600 m in Station LS2, but seawater at depth 300 m was sampled because its depth was only 330 m. The water depth of Station LS3-LS15 was 1000 m or deeper.

Surface seawater samples (~120 L) at depth of 0.5 m were collected using a submersible pump. Other seawater samples (~120 L) at depths of 150 m and 600 m were collected by a CTD-rosette assembly with Niskin bottle samplers (Model Sea-Bird SBE17 plus, Sea Bird Electronics, Inc., Bellevue, Washington, USA). All seawater samples were collected in polyethylene barrels, acidified onboard using concentrated hydrochloric (HCl) acid to obtain a pH < 2 and, then stored for further transportation into the laboratory. The pH was rechecked before analyses to verify successful acidification. If not successfully acidified, any sample stored in polyethylene barrels should be re-acidified; analysis was performed at least 16 h after acidification. All sample treatment and measurements were implemented at the radioactivity monitoring laboratory of South China Sea Environment Monitoring Center (SEMC), SOA.

2.2. Measurement methods

2.2.1. ^{134}Cs and ^{137}Cs

The ^{134}Cs and ^{137}Cs special activities were measured using the AMP (ammonium phosphomolybdate) coprecipitation-γ spectrometry method according to the Technical Specification for Marine Radioactivity Monitoring issued by the Division of Marine Environmental Protection, SOA of P.R. China (Number 10 [2011] Haihuanzi). Thirty milligrams of CsCl and 18 g of AMP were added to 60 L of seawater and the mixture was stirred and adjusted to obtain a pH ≤ 3. The mixture was stirred for 30 min until it was well mixed. After allowing it to stand for > 24 h, the supernatants were removed by siphoning and the residual was filtered. After elution three times using deionized water, a filter membrane was placed into the muffle and incandesced at 450 °C for 2 h. The residue, including ^{134}Cs and ^{137}Cs, was measured using a High-purity Germanium (HPGe) γ-spectrometer with Model 747 Lead Shield and Model BE5030 (Broad Energy Ge; Canberra, USA) detector with its crystal diameter at 80.5 mm and thickness 31.0 mm. The BE5030 detector, with energy ranging from 3 keV to 3 MeV, combines the spectral advantages of Low Energy and Coaxial HPGe detectors. The multi-channel analyser used was Accuspec interface board coupling with microcomputer and the conversion gain and memory were 8192. Genie-2000 software was used to analyze spectrum data (Zhou et al., 2015).

2.2.2. ^{90}Sr

The ^{90}Sr special activity was measured by the di-(2-ethylhexyl) phosphoric acid (HDEHP) extraction-β counting method according to the Technical Specification for Marine Radioactivity Monitoring issued by the Division of Marine Environmental Protection, SOA of P.R. China (Number 10 [2011] Haihuanzi). A total of 2.00 mL of 100 mg/mL Sr $(NO_3)_2$, 1.00 mL of 20 mg/mL $Y(NO_3)_2$, 60 g of NH_4Cl, and 400 g of Na_2CO_3 were added to 40 L of seawater while stirring. The mixture was

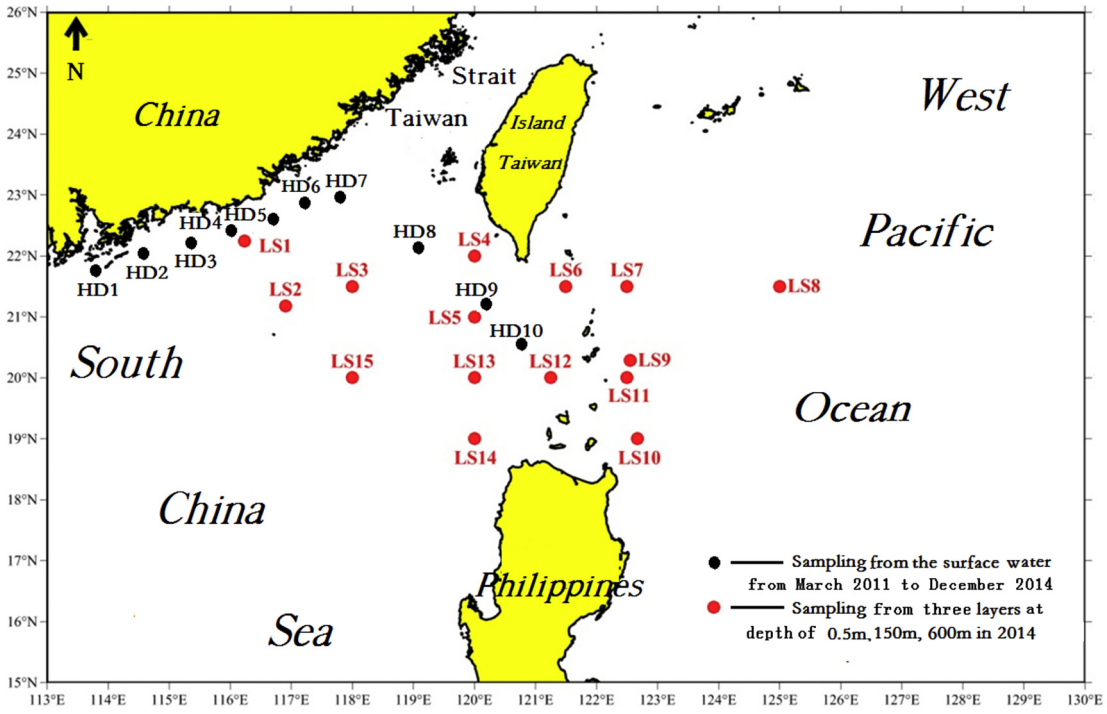

Fig. 1. Sampling stations at the northeast South China Sea and the Luzon Strait and its adjacent sea areas.
(Black dots signify the surface stations in 2011, especially Station HD6 was sampled every three, seven or fifteen days between 21 March 2011 and 31 December 2014; Red dots signify the vertical stations sampled between May and October 2014). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)