



Sources and accumulation of plutonium in a large Western Pacific marginal sea: The South China Sea



Junwen Wu^a, Minhan Dai^{a,*}, Yi Xu^a, Jian Zheng^b

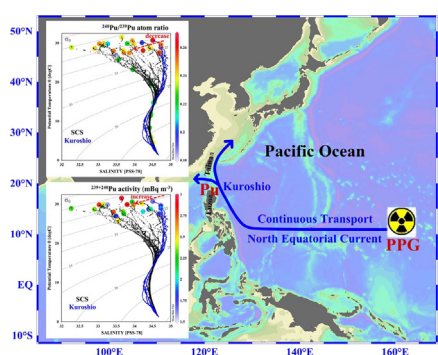
^a State Key Laboratory of Marine Environmental Science, Xiamen University, Xiang'an District, Xiamen 361102, China

^b Fukushima Project Headquarters, National Institute of Radiological Sciences, National Institutes for Quantum and Radiological Science and Technology, 491 Anagawa, Inage, Chiba 263-8555, Japan

HIGHLIGHTS

- ~41% of Pu in the SCS is sourced from the PPG via the NEC and Kuroshio Current.
- There occurred enhanced scavenging of Pu sourced from the PPG.
- There occurred significant accumulation of Pu in the SCS.

GRAPHIC ABSTRACT



ARTICLE INFO

Article history:

Received 26 May 2017

Received in revised form 25 July 2017

Accepted 25 July 2017

Available online xxx

Editor: Jay Gan

Keywords:

Plutonium

South China Sea

Pacific Proving Grounds

$^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio

Scavenging

Accumulation

ABSTRACT

In order to examine the sources of plutonium (Pu) and elaborate its scavenging and accumulation processes, $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios and $^{239} + ^{240}\text{Pu}$ activities in the water column of the South China Sea (SCS) were determined and compared with our previously reported data for the sediments. Consistently high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios that ranged from 0.184–0.250 (average = 0.228 ± 0.015), indicative of non-global fallout Pu sources were observed both in the surface water and at depth during 2012–2014. The spatial distribution of the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in the SCS showed a decreasing trend away from the Luzon Strait, which was very consistent with the introduction pathway of the Kuroshio Current. The Kuroshio had an even heavier Pu isotopic ratio ranging from 0.250–0.263 (average = 0.255 ± 0.006), traceable to the non-global fallout Pu signature from the Pacific Proving Grounds (PPG). Using a simple two end-member mixing model, we further revealed that this PPG source contributed $41 \pm 17\%$ of the Pu in the SCS water column. The $^{239} + ^{240}\text{Pu}$ activities in the SCS surface seawater varied from 1.59 to 2.94 mBq m^{-3} , with an average of $2.34 \pm 0.38 \text{ mBq m}^{-3}$. Such an activity level was ~40% higher than that in the Kuroshio. The distribution of $^{239} + ^{240}\text{Pu}$ in the surface seawater further showed a general trend of increase from the Kuroshio to the SCS basin, suggesting significant accumulation of Pu within the SCS. The $^{239} + ^{240}\text{Pu}$ inventory of the water column in the SCS basin at the SEATS station with a total depth of ~3840 m was estimated to be ~29 Bq m^{-2} , which was substantially higher than the sediment core estimates made for the SCS basin (3.75 Bq m^{-2}) but much lower than the sediment core estimates made for the shelf of the northern SCS (365.6 Bq m^{-2}). Such differences were determined by the lower scavenging efficiency of Pu in the SCS basin compared to the northern SCS shelf.

© 2017 Elsevier B.V. All rights reserved.

* Corresponding author.

E-mail address: mdai@xmu.edu.cn (M. Dai).

1. Introduction

The input of plutonium (Pu), a man-made element, into the oceanic environment is mainly through above ground nuclear weapons testing (Sholkovitz, 1983), accidental releases (Zheng et al., 2012) and discharges from reprocessing plants (Kershaw et al., 1995; Dai et al., 2005). Because of their high toxicity, long half-lives and a large risk for internal radiation exposure, the fate of Pu isotopes in the ocean is thus of great environmental concern. Note that, when deposited in the ocean, Pu takes part in a series of oceanic processes and can be transported quite far from the source point. Such environmental impact can be beyond a regional scale because of this transport (Buesseler et al., 2017).

The relative abundances of Pu isotopes, typically ^{239}Pu and ^{240}Pu , can be commendably used to trace the special sources of Pu because $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios vary with reactor types, neutron flux and energy, nuclear fuel burn-up time, and for fallout from nuclear detonations, weapon types and yield (Wu et al., 2014 and references therein). Our previous study concerning the $^{239} + ^{240}\text{Pu}$ activities and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in sediments of the Northern South China Sea (NSCS) and its adjacent Pearl River Estuary (PRE) infers that Pu in the NSCS is sourced from a combination of global fallout with a characteristic $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio of 0.180 ± 0.014 (Kelley et al., 1999) and close-in fallout traceable to the Pacific Proving Grounds (PPG) in the Marshall Islands that has a very high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio of 0.30–0.36 (Buesseler, 1997; Muramatsu et al., 2001) derived from the above-ground nuclear weapons testing carried out during the period 1952–1958 (Wu et al., 2014 and references therein). The present study aimed to further examine if there is a continuously supplying Pu source from the PPG transported via the North Equatorial Current (NEC) and the Kuroshio Current into the SCS through determination of the Pu activity and its isotopic ratio in the water column of the SCS, and to elaborate the scavenging and accumulation of Pu, which is of great importance to understanding the fate of this artificial radionuclide of environmental concerns. We point out that information on Pu activity levels and isotopic ratios would also help in establishing a baseline for future environmental risk assessment related to nuclear power plant operations that see a dramatically increasing rate along the coast of the region (Zeng et al., 2016).

2. Materials and methods

2.1. Study area

The SCS, with a total surface area of $3.5 \times 10^6 \text{ km}^2$, is the largest marginal sea of the North Pacific Ocean. The East Asian monsoon prevails in the SCS leading to a seasonal alternation of surface circulation with a cyclonic gyre in winter (November to March) and an anti-cyclonic gyre in summer (June to September) (Shaw and Chao, 1994). The basin-wide surface circulation gyres effectively reduce the influence of terrestrial inputs on the SCS proper. As a result, the SCS basin has oligotrophic characteristics similar to those of major ocean basins, with low surface chlorophyll-a levels and low primary production rates (Liu et al., 2002; Dai et al., 2013). In contrast, the nearshore area and the continental shelf are more productive (Han et al., 2012; Dai et al., 2014). In addition, the SCS has a unique circulation pattern with dynamic exchanges with the western North Pacific Ocean (WNPO) via the Luzon Strait with a sill depth of ~2000 m. A branch of the Kuroshio Current intrudes from the WNPO into the SCS in the upper layer (~400 m) and transports westward along the northern continental slope of the SCS, the SCS water outflows into the WNPO in the intermediate layer (~500–1500 m) and the WNPO deep water flows again into the SCS in the deeper layer (~1500 m) (Tian et al., 2006; Liu et al., 2014; Wu et al., 2015).

The Kuroshio is the important western ocean boundary current. It originates from the northward bifurcation of the NEC which flows westward to the Philippine Sea (Wang et al., 2011; Centurioni et al., 2004).

The Kuroshio intrusion to the SCS through the Luzon Strait presents a seasonal pattern with the intrusion being stronger in winter than in summer (Shaw, 1991), and significantly modulates the surface seawater chemistry of the NSCS (Du et al., 2013; Wu et al., 2015).

2.2. Sample collection

Sampling was conducted onboard the R/V *Dongfanghong II* in spring and summer 2012, and in spring 2014. The sampling locations are shown in Fig. 1 and in Table 1. Depth profile samples were collected at the South-East Asian Time Series Stations (SEATS) located in the northern basin of the SCS in April (0–300 m, SEATS I) and in August 2012 (0–1000 m, SEATS II). The remaining samples were all collected from the surface. The surface samples were collected in clean plastic buckets using a built-in pumping system, and subsurface samples were collected in Niskin bottles on a CTD rosette system. Upon collection, about 100 L unfiltered seawater samples were acidified with concentrated HNO_3 to a pH of about 1.6 for the subsequent sample processing and Pu measurements. Adding concentrated HNO_3 to a pH of about 1.6 can destroy any hydrolysis products or complexes of Pu and favors the formation of oxidized Pu (V–VI) in the seawater (Choppin, 2006, 2007).

2.3. Sample processing and Pu isotope analysis

The analytical procedure used for $^{239} + ^{240}\text{Pu}$ in seawater was in accordance with Bu et al. (2014a). Briefly, a known amount of ^{242}Pu (IRMM-085, European Commission Joint Research Centre, Belgium) was added to the seawater samples as a yield monitor. The Pu in the spiked sample was co-precipitated with ferric hydroxide ($\text{Fe}(\text{OH})_3$) by adding ~10 mL Fe^{3+} solution (15.5 mg mL^{-1}) and adjusting to pH ~9 with the addition of concentrated NH_4OH . The precipitate was dissolved in 20 mL 8 M HNO_3 and Pu was adjusted to Pu^{4+} through adding NaNO_2 . The Pu in the sample solution was subsequently purified with two-stage anion-exchange columns using AG 1-X8 and AG MP-1 M (Bio-Rad) (Bu et al., 2014b). The final sample solution, evaporated to near dryness was dissolved in 4% ultrapure HNO_3 (1.0 mL) and filtered for MC-ICP-MS analysis. The determination of Pu isotopes was conducted, using MC-ICP-MS (Nu plasma HR, Nu Instruments Ltd., England) in a low resolution mode in order to obtain the maximal sensitivity, in the State Key Laboratory of Marine Environmental Science, Xiamen University. The DSN-100 high efficiency sample introduction system with a membrane de-solvation unit and a conical concentric nebulizer was used. The chemical yield for Pu was $62.1 \pm 10.7\%$. We obtained high precisions of $^{239} + ^{240}\text{Pu}$ activity and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in seawater, i.e., their relative standard deviations of the uncertainties were $1.6 \pm 1.0\%$ and $2.4 \pm 1.8\%$. This was significantly superior to the values measured using SF-ICP-MS (corresponding to $11.9 \pm 5.6\%$ and $13.0 \pm 4.3\%$) (Bu et al., 2014a). In addition, for Pu measurements with MC-ICP-MS, the most significant interferences are usually caused by the formation of polyatomic uranium hydrides ($^{238}\text{UH}^+$) and peak tailing from the $^{238}\text{U}^+$ peak, resulting in overestimation of the ^{239}Pu signal. The analytical procedure we employed in our work was capable of effectively eliminating the U interferences by achieving an extremely high U decontamination factor of 6.0×10^7 , which was comparable to previously reported values (3.0×10^7 – 1.0×10^8) (Bu et al., 2014a).

The data quality and the mass bias correction were assured by regular analyses of the IAEA-443 (Irish Seawater) (International Atomic Energy Agency) certified reference material ($^{240}\text{Pu}/^{239}\text{Pu} = 0.228 \pm 0.004$, $n = 18$, verified value: 0.229 ± 0.006). The analytical method was also validated by analyzing with other reference materials: IAEA-384 (Fangataufa Lagoon Sediment) and IAEA-385 (Irish Sea Sediment). The accuracies of the $^{239} + ^{240}\text{Pu}$ activities and the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios were in good agreement with the certified and previously reported values (Table 2). In addition, the operational blank count rates for ^{239}Pu and ^{240}Pu were analyzed through measuring 100 L pure water following the same chemical procedure for Pu determination in seawater. The

Download English Version:

<https://daneshyari.com/en/article/5750186>

Download Persian Version:

<https://daneshyari.com/article/5750186>

[Daneshyari.com](https://daneshyari.com)