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Sources and scavenging of plutonium in the East China Sea

Junwen Wu*

Institute of Marine Biology, College of Science, Shantou University, Shantou 515063, China
State Key Laboratory of Marine Environmental Science, Xiamen University, Xiang'an District, Xiamen 361102, China

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ABSTRACT

The $^{240}Pu/^{239}Pu$ atom ratio and $^{239+240}Pu$ activity of seawater in the East China Sea (ECS) was measured in order to examine the Pu sources and elaborate Pu scavenging process. High $^{240}Pu/^{239}Pu$ atom ratios (0.187–0.243, average = 0.221 ± 0.017) in the surface water and water column were observed during 2011, implying of non-global fallout Pu sources. The distribution of $^{240}Pu/^{239}Pu$ atom ratio in the ECS was in agreement with the introduction pathway of the Kuroshio, showing a decreasing trend away from the outer shelf. An even higher $^{240}Pu/^{239}Pu$ atom ratios (0.243–0.263, average = 0.253 ± 0.007) were observed in the Kuroshio, indicating the non-global fallout Pu signal from the Pacific Proving Grounds (PPG). Using a two end-member mixing model, the Pu source from the PPG contribution was calculated to be 36 ± 9% in the ECS seawater. The $^{239+240}Pu$ activities of surface seawater were in the range of 2.00–2.95 mBq m^{-3} in the ECS. The spatial distribution of $^{239+240}Pu$ activity in the surface seawater showed an increasing trend from the outer shelf to the nearshore. Moreover, $^{239+240}Pu$ inventory of water column at the station DH23 in the ECS was calculated to be ~0.29 Bq m^{-2}, which was 1–3 orders of magnitude lower than the estimates of sediment cores in the ECS shelf (9–407 Bq m^{-2}). Such differences were determined by the high degree Pu scavenging efficiency in the ECS and high Pu input carried by terrestrial sediments from the Yangtze River. Finally, both $^{240}Pu/^{239}Pu$ atom ratios and $^{239+240}Pu$ activities were identical before and after the Fukushima nuclear accident (FNA), suggesting that the impact of the FNA on the ECS was negligible.

1. Introduction

Plutonium (Pu), a man-made radionuclide, has been inputted into the marine environment as a consequence of atmospheric nuclear testing (Sholkovitz, 1983), nuclear reprocessing plants discharge (Kershaw et al., 1995; Dai et al., 2005) and nuclear accidents (Zheng et al., 2012). Pu isotopes in the ocean are of great environmental and societal concerns since they have long half-lives, high toxicity and the exposure risk of internal radiation. In particular, the growing concerns were again aroused by the Fukushima nuclear accident (FNA) occurred in 2011. The input of Pu will participate in the subsequently oceanographic processes such as water mass movement and particle scavenging, and can be transported far away from the released point. This transport will result in the environmental risk beyond a local source area (Buesseler et al., 2017). In the East China Sea (ECS), the introduction ways of radioactive contaminants from the FNA are mainly through rapid atmospheric deposition and slow water masses transport (Wu et al., 2013). Wu et al. (2013) evaluated the impact of ^{137}Cs from the FNA on the China Seas due to rapid atmospheric transport, and concluded that such impact was minute. The Kuroshio, a strong west boundary ocean current, was thought to the greatest barrier to prevent the radioactive contaminants from the FNA transporting into the ECS. Nevertheless, recent studies suggested the radio-cesium originating from the FNA could cross the Kuroshio extension along the North Pacific Subtropical Mode Water and then southwardly spread in the North Pacific (Kaeriyama et al., 2014; Kumamoto et al., 2014; Men et al., 2014). The amount of $^{239+240}Pu$ released from the FNA is

* Corresponding author at: Institute of Marine Biology, College of Science, Shantou University, Shantou 515063, China.
E-mail address: wujw@stu.edu.cn.

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reported to be 1.0×10^9–3.5×10^9 Bq (Zheng et al., 2012, 2013; Schneider et al., 2013; Sakaguchi et al., 2014; Shinonaga et al., 2014; Yamamoto et al., 2014). Under the assumption that Pu behaviors similar to Cs and about 80% of the atmospheric Cs released deposited in the ocean (Buesseler et al., 2017), the maximum Pu released into the ocean through atmospheric deposition was calculated to be 0.8×10^9–2.8×10^9 Bq. However, the impact of Pu from the FNA on the ECS via slow water masses transport has not been addressed.

The "fingerprint" of Pu is widely applied to trace the Pu sources in the ocean because the different Pu sources have their own $^{240}Pu/^{239}Pu$ characteristic atom ratios (Wu et al., 2014 and references therein). For example, the $^{240}Pu/^{239}Pu$ atom ratio of global fallout is typically 0.180 ± 0.014 (Kelley et al., 1999), while the $^{240}Pu/^{239}Pu$ atom ratios of close-in fallout from the Pacific Proving Grounds (PPG) in the Marshall Islands where a large-scale USA nuclear testing was carried out in 1950s were featured by 0.30–0.36 (Buesseler, 1997; Muramatsu et al., 2001). $^{240}Pu/^{239}Pu$ atom ratios of the FNA were characterized by 0.30–0.38 (Zheng et al., 2012; Schneider et al., 2013). Previous studies concerning the $^{240}Pu/^{239}Pu$ atom ratios in the ECS sediments infer that the Pu sources are from global fallout and close-in fallout from the PPG (Wang and Yamada, 2005; Liu et al., 2011).

The objective of this study is to examine whether the supply of Pu from the PPG transported through the North Equatorial Current (NEC) and the branch of Kuroshio into the ECS is continuous, to elaborate the Pu scavenging process in the ECS and to evaluate the FNA's impact on the ECS. It is particular importance to understand the fate of the associated with man-made radionuclides at the key areas in the Marshall Islands. It is worthwhile that information on Pu atom ratios and activity levels would also build up a background for future risk assessment with respect to nuclear power plants that see a distinctly increasing rate in this region (Zeng et al., 2016).

2. Materials and methods

2.1. Study area

The ECS, a marginal sea, has a very wide and flat continental shelf (total area of about 7.7×10^5 km^2, a maximum width of 640 km and a mean water depth of 72 m). The Yangtze River, one of the largest rivers in the world, is the major supplier of freshwater, sediments and nutrients to the ECS (Chen, 1996). The surface circulation in the ECS is primarily governed by the East Asian Monsoon, which the strong northeast monsoon prevailed from September to April and the relatively weak southwest monsoon appeared from May to August (Liu et al., 2003). This leads to a seasonal alternation, namely, coastal current flowing to the southward direction in winter and the Taiwan current flowing to the northward direction in summer (Lee and Chao, 2003). In addition, the ECS dynamically exchanges with the western North Pacific (WNP) via the Kuroshio. The Kuroshio Current, characterized by high salinity, is from the northward bifurcation of the NEC flowing westwards to the Philippine Seas (Wang et al., 2011; Centurioni et al., 2004). At the northeast corner of Taiwan, the branch of Kuroshio intrudes into the ECS, and the main stream of Kuroshio continues to flow along the Japan coast in a northeast direction (Ichikawa and Beardsley, 2002).

2.2. Sample collection

Sampling in this study was carried out onboard the R/V *Dongfanghong II* in the cruises of 2011 and 2014–2015. Basic information of samples is listed in Table 1 and the sampling stations are presented in Fig. 1. After the FNA, six surface samples and one water column sample in the ECS were collected in 2011 cruise. Meanwhile, the samples along the Kuroshio mainstream starting from the Luzon Strait to south of Japan during 2014–2015 cruises were collected in order to test the Pu isotopes characteristics of Kuroshio, which is described elsewhere (Wu et al., 2018 and Wu et al. unpublished data). The surface samples were collected in clean plastic containers based on a submersible pumping system. The profile samples were collected in Niskin bottles using an oceanographic rosette system. Upon collection, the concentrated HNO_3 was added into about 60–100 L unfiltered seawater samples acidified a pH of about 1.6.

Table 1
$^{239+240}Pu$ activities and $^{240}Pu/^{239}Pu$ atom ratios in surface seawater of the East China Sea and the Kuroshio.

Area	Station	Latitude (°N)	Longitude (°E)	Bottom depth (m)	Sampling date	Layer (m)	Temperature (°C)	Salinity	$^{239+240}Pu$ activities (mBq m^{-3})	$^{240}Pu/^{239}Pu$ atom ratios
ECS	HH11	36.048	123.498	77	2011-06-08	0.5	15.836	31.890	2.33 ± 0.01	0.187 ± 0.009
	DH32	29.643	123.038	62	2011-06-02	0.5	21.931	33.652	2.78 ± 0.02	0.229 ± 0.010
	DH51	27.880	121.676	36	2011-05-31	0.5	20.340	31.333	2.20 ± 0.03	0.218 ± 0.014
	PN05	29.336	125.521	102	2011-06-06	0.5	20.450	32.051	2.00 ± 0.02	0.217 ± 0.014
	DH02	31.999	123.999	43	2011-06-07	0.5	16.610	31.404	2.65 ± 0.02	0.203 ± 0.010
	DH05	32.006	126.481	102	2011-06-07	0.5	19.592	33.382	2.95 ± 0.03	0.232 ± 0.012
	DH23	28.331	123.006	77	2011-06-01	0.5	22.536	34.129	2.91 ± 0.03	0.235 ± 0.011
Kuroshio	K1	19.000	123.000	3657	2014-05-29	0.5	30.726	34.506	1.64 ± 0.02	0.263 ± 0.007
	K2	19.994	122.994	4412	2014-11-12	0.5	28.121	34.448	1.36 ± 0.02	0.259 ± 0.008
	K3	21.000	123.000	3410	2014-05-27	0.5	29.652	34.714	1.75 ± 0.02	0.250 ± 0.005
	K4	21.129	121.683	2740	2014-05-28	0.5	28.951	34.730	1.93 ± 0.02	0.251 ± 0.006
	K5	22.998	122.999	3150	2014-05-26	0.5	27.525	34.819	1.76 ± 0.02	0.255 ± 0.003
	K6	27.615	125.423	550	2015-04-02	0.5	20.549	34.430	1.78 ± 0.04	0.249 ± 0.008
	K7	31.003	134.857	1640	2015-04-30	0.5	21.488	34.599	1.15 ± 0.01	0.243 ± 0.006