Chemosphere 207 (2018) 130–138

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Sedimentary record of plutonium in the North Yellow Sea and the response to catchment environmental changes of inflow rivers

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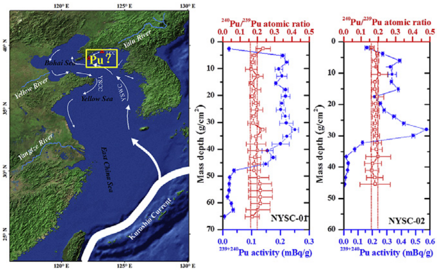
HIGHLIGHTS

- Sedimentary records of Pu reflected well the catchment environmental changes.
- The atmospheric fallout is the dominant source of Pu in the North Yellow Sea.
- PPG-derived Pu has no significant influence on the North Yellow Sea.
- Pu is a good chronological tracer for studying modern costal sedimentary process.
- Pu inventories indicated the riverine input source other than the direct deposition.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 January 2018
Received in revised form
25 April 2018
Accepted 13 May 2018
Available online 18 May 2018

Handling Editor: Martine Leermakers

Keywords:
Plutonium
North Yellow Sea
Sediment
Isotopic ratio
Chronological marker
Riverine input

ABSTRACT

Plutonium (Pu) isotopes were first determined in surface and core sediment samples collected from the northern North Yellow Sea (NYS) to elucidate their source terms and deposition process as well as the response to catchment environmental changes of inflow rivers. ^{240}Pu/^{239}Pu atom ratios in all sediments showed the typical global fallout value of ~0.18 without any influences from the nuclear weapons tests conducted recently in the North Korea or early in the Pacific Proving Ground. The large variation of $^{239+240}$Pu activities (0.022–0.515 mBq/g) observed in surface sediments should be mainly attributed to the re-suspension and transportation of fine sediments influenced by the Liaonan Costal Current. Based on the two $^{239+249}$Pu depth profiles with easily observed onset fallout levels (1952) and global fallout peaks (1963), $^{239+240}$Pu served as a valid time mark in the coastal sedimentary system. Riverine input Pu contributed only 15–27% to the total global fallout inventory (92.5–108.8 Bq/m^2) in the northern NYS, much lower than that in the Yangtze River estuary (77–80%), indicating a better soil conservation in the northeast China due to higher forest coverage compared to the Yangtze River's drainage basin. The increase of riverine input Pu after 1980s reflected the more intense soil erosion degree caused by the land use and cover change due to the increment of human activities in the northeast China at the same period.

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https://doi.org/10.1016/j.chemosphere.2018.05.082

Our results demonstrated that plutonium is a good indicator for studying sedimentary process and its response to the environment in the coastal area.

1. Introduction

Plutonium isotopes (^{239}Pu and ^{240}Pu) have been substantially introduced to the marine environment mainly from the global fallout of extensive atmospheric nuclear weapons tests (NWT) since 1945. The global fallout has not equally dispersed on the ocean surface, but had a maximum deposition over the mid-latitude in the western North Pacific Ocean due to the geographic and climatological conditions (Hirose et al., 2001; Gatsaud et al., 2011). In addition to the global fallout, close-in fallout from the Pacific Proving Ground (PPG), where a series of U.S. nuclear tests were conducted in the 1940s-1950s, was a second significant source of Pu in the North Pacific Ocean. ^{239}Pu ($T_{1/2} = 24110$ yr) and ^{240}Pu ($T_{1/2} = 6563$ yr) in the marine environment are of great environmental concern due to their high chemical toxicity and long half-lives. Besides to be good indicators for radioactive pollution, Pu isotopes can also serve as useful geochemical tracers for better understanding a variety of marine processes (Lindahl et al., 2010).

With the high particle affinity, Pu isotopes entered into the oceans can be effectively scavenged by settling particulate matter from the water column to the seafloor sediments. This process is well known as “boundary scavenging” and generally enhanced in ocean boundaries/margins, where are usually areas with high biological productivity, high particle flux and sediment accumulation (Hong et al., 2011). Seafloor sediments are therefore the ultimate sink of Pu entering the marine environment, and preserve Pu deposition history, from which valuable information of impact of human activities on the marine environment and its changes over time can be deduced. With the well constrained release history of the global fallout Pu from the NWTs, which began in the late 1940s and had a distinct deposition maximum in 1963 (UNSCEAR, 2000), undisturbed sediment recording these two events can be identified and thus used for dating purpose (Corcho-Alvarado et al., 2014). With the recent rapid development in analytical procedures and mass spectrometric measurements (Qiao et al., 2009), $^{239+240}$Pu has been suggested as a chronostratigraphic marker for modern sediments dating in both freshwater bodies (e.g. lakes and river estuaries) (Ketterer et al., 2004; Zheng et al., 2008; Wu et al., 2010; Pan et al., 2011) and coastal areas (Hancock et al., 2011; Corcho-Alvarado et al., 2014), and shown some advantages to the conventional ^{137}Cs method, such as more precise determination especially in marine sediments (Hancock et al., 2011) and additional chronometers associated with changes in ^{240}Pu/^{239}Pu atomic ratios for Pu isotopes fallout during the 1950s and 1960s (Koide et al., 1985) as well as fallout from the 1986 Chernobyl accident in some European areas (Ketterer et al., 2004).

In the past decades, numerous studies have been carried out to investigate source terms, transport, and scavenging and deposition processes of Pu isotopes in China marginal seas (Nagaya and Nakamura, 1992; Huh and Su, 1999; Su and Huh, 2002; Lee et al., 2003; Wang and Yamada, 2005; Hong et al., 2006; Zheng and Yamada, 2006; Liu et al., 2011; Pan et al., 2011; Wu et al., 2014; Wang et al., 2017). However, most of these studies concentrated in the East China Sea and the Yangtze River Estuary. Regarding Pu isotopes in the Yellow Sea sediments, only several data points have been reported in the South Yellow Sea and Jiangsu tidal flats (Nagaya and Nakamura, 1992; Hong et al., 2006; Liu et al., 2013). No Pu data in the North Yellow Sea (NYS) sediments is available. The Yellow Sea located between mainland China and the Korean Peninsula is a semi-closed shallow marginal sea. It is normally divided into two parts: the South Yellow Sea is connected to the East China Sea, while the NYS, considered in the present study, is adjacent to the Bohai Bay and surrounded by the Liaodong Peninsula, Shandong Peninsula and Korean Peninsula. The coastal areas of the NYS are densely populated areas, where are very close to North Korea. Nuclear activities including NWTs in North Korea have caused a great concern on the radiation exposure to the public in the past years. The Hongyanhe nuclear power plant (NPP) with 4 units located in Dalian (39.792°N, 121.475°E) on the Liaodong Peninsula has already been operating from 20th September 2016. Potential releases of radioactive materials including Pu from the NPP to the surrounding environment and the consequences are also a major concern of the local inhabitants and the authorities.

In the north coast of the NYS, major rivers flowing into the sea include Yalu River, Dayang River and Biliu River. These rivers play important roles in the transportation of terrigenous sediments to the northern NYS. The fluvial sediments providing significant source of sediments in the coastal sea (Liu et al., 2016), are related to the soil erosion degree in river basins and can vary in response to climate changes and human activities, which can be well reflected in the coastal sedimentary system. Thus investigation of the sediment deposition process in the coastal sea and its dynamics is of great significance for evaluating water and soil loss in surrounding river basins and comprehensive understanding the land-ocean interaction within the coastal areas.

In this work, surface sediments and sediment cores were collected in the northern NYS (Fig. 1) and analyzed for ^{239}Pu and ^{240}Pu, aiming (1) to investigate the source term, distribution and deposition process of Pu in the study area; (2) to quantify the contribution of fluvial sediments to the total Pu deposition, as well as to evaluate the water loss and soil erosion in surrounding drainage systems and its response to human activities; (3) to provide baseline data of Pu for environmental risk assessment in relation to the nuclear activities in North Korea and the Hongyanhe NPP.

2. Materials and methods

2.1. Study area

The study area was located along the southeastern coast of the Liaodong Peninsula, in the northern NYS (Fig. 1). Many islands are scattered along the coast (e.g. Shicheng Island, Changshan Island, etc.), resulting in the complex subwater topography in the study area (Chen et al., 2013). There are a number of rivers flowing into the NYS, among which the Yalu River is the largest one with the sediment discharge of 113×10^4 tons/a (State Oceanic Administration, 1998). Besides, the Dayang River and Biliu River with the sediment discharges of 69.3×10^4 tons/a and 53.3×10^4 tons/a also contribute significant amounts of sediments to the study area (Bao and Du, 1999). The water circulation in the study area is dominated by the Liaonan Coastal Current (LCC), which is formed by the Yalu River Diluted Water Mass and flows constantly along the southeast coast of the Liaodong Peninsula to the Bohai Strait. It has been reported that the Yalu River derived sediments were re-suspended and then transported by the LCC southwestward for a long distance and re-deposited along the southeast coast