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*Published in:*
Chemosphere

*Link to article, DOI:*
10.1016/j.chemosphere.2018.05.082

*Publication date:*
2018

*Document Version*
Peer reviewed version

# Sedimentory records of plutonium ~~Plutonium~~ in ~~sediments of~~ the North Yellow Sea and ~~its~~ response to catchment environment of inflow rivers

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ABSTRACT: Plutonium isotopes were first determined in surface and core sediment samples collected from the northern North Yellow Sea to elucidate their source terms and deposition process as well as response to the environment. $^{240}Pu/^{239}Pu$ atomic ratios in all sediments showed the typical global fallout value of ~0.18 without any influences from the nuclear weapons tests

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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) in surface sediments was mainly attributed to the re-suspension and transportation of fine sediments influenced by the Liaonan Costal Current. Based on the two Pu depth profiles with easily observed onset (1952) and global fallout peaks (1963), $^{239+240}$Pu served as a valid time mark in the coastal sedimentary system. Riverine input plutonium contributes only 15-27% to the total global fallout inventory (92.5-108.8 Bq/m$^2$) in the study area, much lower than that in the Yangtze River estuary (77-80%), indicating a better soil conservation in the northeast China compared to the Yangtze River's drainage basin. Riverine input Pu increased after 1980s reflected the increased soil erosion degree caused by the land use and cover change due to the increased human activities in the northeast China. These results demonstrated that plutonium is a good indicator for studying sedimentary process and its response to the environment in the coastal area.

**Key words:** Plutonium, North Yellow Sea, sediment, isotopic ratio, chronological marker, riverine input

## Highlights

**1. The atmospheric fallout is the dominant source of Pu in the North Yellow Sea.**

**2. PPG input Pu has no influence on the North Yellow Sea.**

**3. Riverine input Pu contributed about 15-27% to the northern North Yellow Sea**

**4.Riverine input Pu contributed much less in the NYS than in the Yangtze River estuary.**

**5. Pu is a good indicator to study the sediment process in coast sea.**

## 1. ~~INTRODUCTION~~Intrduction

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 (Gatsaud et al., 2011; Hirose et al., 2001).[1-2] 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).[3]

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).[4] 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),[5] undisturbed sediment recording these two events can be identified and thus

used for dating purpose (Corcho-Alvarado et al., 2014).[6] With the recent rapid development in analytical procedures and mass spectrometric measurements (Qiao et al., 2009),[7-8] $^{239+240}$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; Wu et al., 2010; Zheng et al., 2008) [9-12] and coastal areas (Corcho-Alvarado et al., 2014; Hancock et al., 2011),[6, 13] and shown some advantages to the conventional $^{137}$Cs method, such as more precise determination especially in marine sediments(Hancock et al., 2011)[13] and additional chronometers associated with changes in $^{240}$Pu/$^{239}$Pu atomic ratios for Pu fallout during the 1950s and 1960s (Koide et al., 1985)[14] as well as fallout from the 1986 Chernobyl accident in some European areas (Ketterer et al., 2004). [9]

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 (Hong et al., 2006; Huh & Su, 1999; Lee et al., 2003; Liu et al., 2011; Nagaya & Nakamura, 1992; Pan et al., 2011; Su & Huh, 2002; Wang et al., 2005; Wang et al., 2017; Wu et al., 2014; Zheng & Yamada, 2006).[15-27] However, most of these studies concentrated in the East China Sea ~~ESC~~ and the Yangtze River Estuary. Regarding Pu isotopes in the Yellow Sea ~~(YS)~~ sediments, only several data points have been reported in the South~~thern~~ Yellow Sea ~~(SYC)~~ and Jiangsu tidal flats (Hong et al., 2006; Liu et al., 2013; Nagaya & Nakamura, 1992).[15, 21, 25] No Pu data in the North Yellow Sea (NYS) sediments is available. The ~~YS~~ 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 ~~SYC~~ South Yellow Sea is connected to the ~~ESC~~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 (39.792ºN, 121.475ºE) on the Liaodong Peninsula has already being in operation from 20th September 2016. ~~In addition, there are plans to build 4 more NPPs in China's three northeast provinces.~~ Potential releases of radioactive materials including Pu from the NPP~~s~~ 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 sediment discharges providing significant source of sediments in the coastal sea (Liu et al., 2013), are related to the soil erosion degree in river basins and can vary in response to climate changes and human activities in the catchment, 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 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 (Figure 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 identify the contributions of direct deposition and riverine input by the deposition history of Pu, as well as to evaluate the water loss and soil erosion in the coastal major watersheds and its response to ~~climate change and~~ 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 METHODSaterials and methods

### 2.1 Study Area

The study area was located along the southeastern coast of the Liaodong Peninsula, in the northern NYS (Figure 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).[28] There are a number of rivers flowing into the NYS, among which the Yalu River is the largest one with the sediment discharge of $113\times10^4$ tons/a (State Oceanic Administration, 1998).[29] Besides, the Dayang River and Biliu River with the sediment discharges of $69.3\times10^4$ tons/a and $53.3\times10^4$ tons/a also contribute significant amounts of sediments to the study area (Bao & Du, 1999).[30] 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 of Liaodong Peninsula (Chen et al., 2013).[28] In addition to the LCC, the strong Yellow Sea Warm Current (YSWC) in winter, the Yellow Sea Coastal Current (YSCC) and the West Korean Coastal Current (WKCC) might also affect the material transport in the study area.

### 2.2 Sample collection and pretreatment

Sampling was conducted onboard in July 2016 and March 2017, respectively. Eight surface sediments (0-2 cm depth) and two sediment cores (NYSC-01 with water depth of 10 m and NYSC-02 with water depth of 20 m) were collected using the grab buckets and a gravity sampler, respectively. The sediment cores were sliced in 2 cm intervals and weighed

immediately, 2 g of sediment was taken from each surface and interval sediment sample for the analysis of the grain size composition by the Malvern Mastersize 2000 laser granulometer (measuring range 0.02-2000 μm, with a duplicate measurement error of < 3%). The remained sediment samples were freeze-dried and reweighed to determine mass depths in the laboratory. Dried samples were then ground to fine powder for Pu isotopes analysis.

## 2.3 Determination of Pu isotopes

The method for Pu isotopes analysis is similar as described in ~~our previous publications~~Xu et al., 2014.~~[31-34]~~ Briefly, approximately 5 g of dried sediments for each sample were weighed to a beaker and ashed at 550 °C overnight to decompose the organic matter as well as to obtain organic matter ~~(OM)~~ content by calculating the mass loss in this process. After being spiked with a known amount of $^{242}$Pu (ca. 1 mBq) as a chemical yield monitor (the $^{242}$Pu added to the ashed aliquot does not cover the losses during the mineralization, but they should be negligible), the samples were leached using *aqua regia* for 2 h at 200 °C, Pu in the leachate was co-precipitated with iron hydroxides to remove the major matrix components. After centrifuge, the precipitate was dissolved with concentrated HCl. Then Pu in the solution was reduced to Pu(III) by adding about 300 mg of $K_2S_2O_5$ and then oxidized to Pu(IV) with conc. $HNO_3$. A 15 mL AG1-×4 anion-exchange column and a 2 mL TEVA column were successively used for separation and purification of Pu isotopes. The final sample solution prepared in 3 mL of 0.5 mol $L^{-1}$ $HNO_3$ was analyzed for Pu isotopes ($^{239}$Pu, $^{240}$Pu, $^{242}$Pu) using a section field ICP-MS (SF-ICP-MS) (Element 2, Thermo Scientific, Bremen, Germany). An APEX-Q high efficiency sample introduction system (Elemental Scientific Inc., Omaha, NE, USA) combined with a membrane desolvation unit (ACM) and a conical concentric nebulizer was used to introduce sample solution to the ICP-MS. All the measurements were made in the self-aspiration mode with an

uptake rate of ~ 0.2 mL $min^{-1}$ to reduce the risk of contamination from the long peristaltic pump tubing. 0.1 ng $mL^{-1}$ U standard solution was used for tuning SF-ICP-MS before the samples were measured each time. The $^{242}Pu$ standard solution in the same media as the samples and procedure blanks were measured with the samples. The concentrations of $^{239}Pu$ and $^{240}Pu$ in the sample were calculated by the measured signal intensities of $^{239}Pu$, $^{240}Pu$ and $^{242}Pu$ and the amount of $^{242}Pu$ added to the sample before chemical separation. $^{242}Pu$ was used in this procedure as both dilution isotope and chemical yield monitor.

## 3. Results and discussions ~~ESULTS AND DISCUSSION~~

### 3.1 Source terms of Pu isotopes in the northern NYS

Since the isotopic composition of Pu is related to its production and release, the atomic ratio of $^{240}Pu/^{239}Pu$ in environmental samples could be used to identify the source term of plutonium contamination. It has been well proved that the global fallout has an average $^{240}Pu/^{239}Pu$ ratio of 0.18 (Kelley et al., 1999; Warneke et al., 2002).~~[35-37]~~ $^{240}Pu/^{239}Pu$ atomic ratios for weapons-grade Pu were reported to be very low (~~0.01-~~<0.07) (Lee & Clark, 2005),~~[38-39]~~ while much higher values were reported in materials of nuclear power reactors (0.2-0.8, depending on the reactor type and fuel burn-up) (Muramatsu et al., 2000; Warneke et al., 2002).~~[37, 40]~~ It has also been found that different test series have different atomic ratio. For example, the $^{240}Pu/^{239}Pu$ atomic ratios in the close-in fallout at the Nevada test site are generally very low with an average of 0.035 (Buesseler, & Sholkovitz, 1987),[41] while close-in fallout in the PPG of the Marshall Islands in the 1950s is characterized by higher ratios of 0.30-0.36 (Buesseler, 1997; Koide et al., 1985; Muramatsu et al., 2001).~~[14, 42-43]~~ Such characteristic of PPG-derived plutonium has been intensively employed to trace transportation of PPG-derived pollutions in the North Pacific Ocean and its marginal seas including the ~~ESC~~ East China Sea and ~~SYS~~South Yellow Sea. However, it is still unclear

whether the PPG-derived pollution has entered to the ~~YS~~Yellow Sea, especially the NYS due to lack of information on Pu isotopic compositions in this area.

As presented in Table A.~~S~~1, A.~~S~~2 (~~Supporting~~ Supplementary material~~information~~) and depicted in Figure 2, the measured $^{240}Pu/^{239}Pu$ atomic ratios ranged from 0.175 to 0.190 with an average of 0.184 ± 0.005 in all surface sediment samples. In the two sediment soil cores, $^{240}Pu/^{239}Pu$ atomic ratios varied from 0.164 to 0.233 with an average of 0.194 ± 0.02 in NYSC-01 core, and from 0.163 to 0.206 with an average of 0.185 ± 0.011 in NYSC-02 core, which were relative constant and similar to those determined in surface sediments. It is clear that the atomic ratios of $^{240}Pu/^{239}Pu$ obtained in all sediment samples agreed very well with the average value of ~0.18 for the global atmospheric fallout, as well as with ~~our~~ previous reported values (~0.18) in soils collected from Liaodong Bay coastal regions (Xu et al., 2013),[~~31, 33-34~~] which are quite close to the present study area. Similar $^{240}Pu/^{239}Pu$ atomic ratios have also been reported in a sediment core from the Lake Sihailongwan (42.29°N, 126.6°E) (Wu et al., 2010),[~~11~~] which is also located in the northeast China and not very far (<400 km) from our study area. Combined these results with our previous conclusion regarding Pu isotopes in soils of northeast China that the major source of Pu in northeast China is the global atmospheric fallout, and any other possible contributions should be very small, Pu isotopes in the sediments collected from the present study area should be also mainly from the global atmospheric fallout.

It is well known that several NWTs have been conducted in the North Korea in the past ten years. However, Pu is a refractory element and is not easily to be released to the atmosphere during underground NWT, thus contributions of Pu from the NWTs in North Korea should be negligible in spite of the short distance between the North Korea and our study area. Plutonium released from the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident might have

potential impact on the surrounding areas including the ~~YS~~Yellow Sea. ~~Since~~ However, the FDNPP-derived Pu has been reported to have much higher $^{240}Pu/^{239}Pu$ atomic ratios (0.303 to 0.~~381~~330) (Zheng et al., 2012)[~~44-45~~] than the measured values in present studied sediments collected in 2017, and some other reports (Bu et al., 2014; Hain et al., 2017; Oikawa et al., 2015;) have also indicated that the influence of the FDNPP-diverved Pu to the marine environment would be limited. ~~-~~Thus it is reasonable to believe that the contribution from the FDNPP-source Pu to the total Pu inventory in our study area is negligible.

Numbers of studies have reported that the close-in fallout Pu from the PPG has been transported from the Marshall lands through the North Equatorial Current followed by the Kuroshio Current and its extensions to the ~~SCS~~ South China Sea and ~~ESC~~East China Sea (Liu et al., 2011; Wang et al., 2005; Wang et al., 2017; Wu et al., 2014) .[~~19, 24, 26-27~~] Significant higher $^{240}Pu/^{239}Pu$ atomic ratios (> 0.20) were usually observed in surface sediments or bottom layers of sediment cores in these marginal seas due to the contributions of PPG-sourced Pu with high $^{240}Pu/^{239}Pu$ atomic ratios of 0.30-0.36. But this is not the case in our study area, since no abnormal higher ratios of $^{240}Pu/^{239}Pu$ were observed in both surface sediment samples and bottom layers of the two sediment cores. This indicates at least that our study area did not receive any PPG derived Pu from either the early direct close-in fallout or oceanic current transportation. Although earlier studies regarding Pu in surface seawater samples from the central ~~YS~~ Yellow Sea and ~~SYS~~ South Yellow Sea has shown some PPG-sourced Pu signals in surface seawater with relative higher ratios (0.18-0.33 with an average of 0.23) (Kim et al., 2004),[46] the impact of the PPG close-in fallout Pu on the Yellow Sea~~S~~ is still doubtful. Liu et al.（2013）[~~25~~] studied the Pu isotopes in the North Jiangsu tidal flats of the ~~YS~~ Yellow Sea and they concluded that the tidal flats did receive mainly global fallout Pu but no PPG-derived Pu in any pathways, which

are similar as our conclusions for the northern NYS. Therefore, the influence of the PPG-derived Pu on the ~~YS~~Yellow Sea, especially the NYS might be very limited.

All above results suggested that the global atmospheric fallout is the major source of Pu in the northern NYS, and any other possible contributions such as releases from NWTs of the North Korea, the FDNPP accident, as well as early direct deposition or later oceanic current transportation of PPG derived Pu are negligible. Since Feb. 2013, the Hongyanhe NPP has started in operation of four units successively. The measured Pu isotopic composition in this work will serve as a reference data for the future monitoring and assessment of environmental impact of this NPP.

### 3.2 Spatial distribution of Pu isotopes in surface sediments in the northern NYS

The $^{239+240}$Pu activities in surface sediments collected along the southeast coast of the Liaodong Peninsula in the NYS are presented in Table A.~~S~~1 and Figure 3. A large variation of $^{239+240}$Pu concentrations from 0.022 to 0.515 mBq/g with an average of 0.122 ± 0.021 mBq/g ($n$ = 10) was observed in these sediment samples (Figure 2). Such $^{239+240}$Pu activity levels are comparable to the reported values observed in the NYS (0.045 to 0.901 mBq/g) (Hong et al., 2006; Nagaya & Nakamura,1992)~~[15, 21]~~ and the ~~ESC~~ East China Sea (0.048-0.759 mBq/g) (Liu et al., 2011; Wang et al., 2017).~~[24, 27]~~ It can be observed from the Figure 3 that the $^{239+240}$Pu concentrations generally show a westwards increasing trend along the coast with the highest values near the northeast cost of Dalian Bay, which is similar to the direction of the LCC.

The distribution of Pu concentrations in surface sediments might be influenced by many factors such as organic matter and particle size of the sediment, coastal currents and riverine input. Since the differences of organic matter content in the surface sediments excluding the surface sample of the NYSC-02 sediment core are very small, the influence of the organic matter

contents on the $^{239+240}$Pu activities was not significant. ~~It has been reported by~~Based on the analysis of the rare earth element compositions of the surface sediments in the southeast coastal area of the Liaodong Peninsula, Chen et al.(2013)[28] have reported that after re-suspension and transportation by the LCC, the Yalu River-derived fine-grained sediments were re-deposited in water depth of 20-40 m in the area, where is 180-300 km away from the Yalu River mouth and extends along the southeast coast of the Liaodong Peninsula between the northeast of Dalian Bay and southwest of the Changshan Islands (as shown in Figure 1). It is obvious that the two sampling sites of SS05 and SS06 in this study are just located in the reported Yalu River-derived fine-grained deposition region. ~~Our previous results~~ Researches regarding the association of Pu isotopes with natural soil particles have proved that Pu isotopes tend to associate with finer particles due to their bigger specific surface areas (Xu et al., 2017).[34] Therefore, significant higher $^{239+240}$Pu activities in the sampling sites SS05 and SS06 are more likely to be attributed to their much smaller particle sizes (SS05 and SS06 with mean grain sizes of 3.2 and 2.4 μm respectively)., ~~although there was no direct linear correlation between mean grain sizes and $^{239+240}$Pu activities in all surface sediments.~~ The lowest Pu concentrations in the east part of the study area might be in relation to the dilution of Pu-depleted river-borne sediments from the Yalu River and Dayang River. While in the most west two sampling sites SS07 and SS08, which are close to the Bohai Strait, their very low $^{239+240}$Pu concentrations (0.025 and 0.040 mBq/g) are likely to be related to the interaction influence of the YSWC and YSCC. Hence, the transport of coastal currents and sediment dynamics of fine-particles should play an important role in controlling the spatial distribution of Pu isotopes in the northern NYS.

### 3.3 Temporal variations of Pu isotopes in the northern NYS

The vertical profiles of $^{239+240}$Pu activities in the two sediment cores are illustrated in Figure 4 and summarized in Table A.S2. The $^{239+240}$Pu concentrations in the NYSC-01 sediment core ranged from 0.006 to 0.251 mBq/g with an average of 0.141 ± 0.061 mBq/g, which are nearly 2 times lower than those in the NYSC-02 sediment core with a range of 0.002 to 0.576 mBq/g and an average of 0.231 ± 0.052 mBq/g. This difference may be partly attributed to the effect of particle size on the association of Pu with sediments. Considering that the NYSC-02 sampling site with water depth of 20 m and fine size grain sediment of a mean grain size of 6.04 μm is located in the fine grained deposition region, higher Pu concentrations would be expected at this site due to the stronger adsorption of plutonium by finer grain sediments. While the NYSC-01 sampling site with water depth of 10 m and relative coarser grain sediment of a mean grain size of 6.82 μm is located in the pathway of the LCC and much closer to the mouth of the Yalu River. In the near shore zone, due to the strong hydrodynamic conditions, fine particles adsorbed more Pu isotopes are easily re-suspended in water column and further transported westwards through the LCC, but coarse sediments with weaker adsorption of Pu isotopes were deposited locally, resulting in the lower Pu concentrations in the NYSC-01 site. Moreover, the difference in organic matter contents may also be one reason of different Pu concentrations in these two sediment cores. Although linear regression analyses showed that there was no obvious correlation between Pu concentrations in the NYSC-02 core and its relative constant organic matter contents from the top to the bottom layers. While, the relationship between the organic matter contents and Pu concentrations in the NYSC-01 core is significant ($r^2$= 0.64, $p$<0.05) as shown in Figure A.S1. The organic matter content in the NYSC-02 (with an average of 7.32%) was much higher than that in the NYSC-01 core (average of 5.84%).

It can be observed from the Figure 4 that $^{239+240}$Pu activities in both sediment cores showed unambiguous maximum values and background levels. Based on the previous discussions that Pu isotopes in the study area are mainly from the global fallout and the well-defined global fallout history, it is reasonable to relate the pronounced peaks to the historical global fallout maximums in 1963 and the positions of background values to the 1952 horizons (onset fallout). Using the 1963 horizon as a discrete time marker, we obtained similar mean sedimentation rate of 0.46 cm/yr for both sediment cores, but two different sediment accumulation rates of 0.61g/cm$^2$·yr for the NYSC-01 core and 0.52 g/cm$^2$·yr for the NYSC-02 core, respectively. While taking the 1952 horizons as a chronological marker for sediment dating, the mean sedimentation rates and mean sediment accumulation rates were calculated to be 0.6057 and 0.51cm/yr and 0.774 and 0.56 g/cm$^2$·yr respectively for the NYSC-01 and NYSC-02 cores, which are slightly higher but still comparable to the results obtained from the 1963 marker. The difference between the results obtained from the two time markers has been evaluated using a statistic *t*-test ($t_{stat}$=-3.181.34<$t_{crit}$ for $n$=4), which suggests that both 1963 and 1952 horizons are valid chronological markers for the sediment dating in the present study area. In fact, our results are also within the range of the results (0.06-1.18 g/cm$^2$·yr) reported by Qi et al.(2004)[47] using $^{210}$Pb$_{ex}$ dating method regarding the sediment cores in the NYS. The mean sediment accumulation rates obtained in the NYSC-01 core from the both time markers were obviously higher than those in the NYSC-02 core, indicating that the sedimentary environment in the latter sampling site are relatively stable in the past decades. This was reasonable and coincided well with the previous discussions.

Further insight into the temporal variation of Pu isotopes in the northern NYS was provided by the distribution patterns of Pu concentrations from the surface to the 1963 horizon in both sediment cores. According to the report of UNSCEAR (2000),[5] the temporal variation of the

annual global fallout deposition of $^{137}Cs$ in northern hemisphere (Figure 5) showed a maximum in 1963 and then significantly decreased to a low level especially after 1980. The historical pattern of the global fallout deposition of $^{239+240}Pu$ is believed to be similar. According to the estimation of Nakano and Povinec (2003),[48] the annual global fallout deposition of $^{239+240}Pu$ in marine environment was also found to be maximal in 1963 and then decrease dramatically. While the direct deposition of the global fallout Pu was recorded in undisturbed lakes, as depicted in the Figure 5, typical $^{239+240}Pu$ concentration peaks corresponding to the maximum deposition of 1963-1964 and followed by sustained declines were observed in lake Qinhai and lake Hongfeng which are located respectively in northwest and southwest of China (Wu et al., 2011; Zheng et al., 2008).[10, 12] However, A constant high $^{239+240}Pu$ activities were observed in the sediment core NYSC-01 corresponding to the period of 1979- 2009 (Figure 4 and 5), and significant higher $^{239+240}Pu$ activities were observed in the sediment core NYSC-02 from the mass depth of 15.13 g/cm$^2$ to 5.99 g/cm$^2$ corresponding to the period of 1988 to 2005. This indicates that besides the direct atmospheric fallout, there are other inputs of plutonium to the sediment, especially after 1980. The riverine input sediments which are mainly sourced from the soil erosion in the river catchments should be the main source of the excessive plutonium in the sediment cores. Plutonium in the riverine sediments in the northeast China should also originate from the global fallout according to the previous studies based on the constant $^{240}Pu/^{239}Pu$ ratios in the whole soil and sediment cores (Wu et al., 2010; Xu et al., 2013).[11, 31-34] In conclusion, the unusual distributions of global fallout Pu in the upper layers of the two sediment cores is attributed to the riverine input sediments over ~~time,~~time and this will be further discussed in the next section.

## 3.4 Contribution of Pu from riverine input and its response to catchment environment

Inventory of $^{239+240}$Pu in sediment cores can help to interpret the source function and the transport of Pu, thus providing a useful indicator for assessing the sedimentary process in the marine environment. In this study, how riverine input sediment influenced the global fallout Pu depositions in the sediments could be further deduced from the inventories of Pu in the two sediment cores. The total Pu inventories were estimated to be 92.5 ± 1.6 Bq/m$^2$ for the NYSC-01 core and 108.8 ± 1.2 Bq/m$^2$ for the NYSC-02 core, respectively. These values are nearly twice the average values of global atmospheric fallout (42-58 Bq/m$^2$) expected at the similar latitudinal belts of 30-50ºN (UNSCEAR, 1982; 2000) ,[5, 49] and also generally higher than those (55.5-90 Bq/m$^2$, Table A. ~~S~~3) obtained from the undisturbed soil and sediment sampling sites with similar average annual precipitations (600-800 mm) at the similar latitude of ~39 ºN worldwide (Hardy et al., 1973; Wu et al., 2010; Xu et al., 2013; 2015),[11, 31, 33, 50] which are believed to receive solely global fallout Pu. This indicates that besides the direct deposition from the global fallout, riverine input should contribute to some Pu depositions in the study area. Since the integrated global fallout values of 42 and 58.1 Bq/m$^2$ respectively for 30-40ºN and 40-50ºN published by UNSEAR (1982; 2000)[5, 49] were average values without considering the influence of annual precipitation on Pu fallout in specific sampling sites, they may not indeed represent the direct deposition of global fallout in the study area. Comparing global fallout Pu inventory data obtained from undisturbed soil and sediment cores with similar annual precipitation and latitudes as the study area (~39 ºN), an average value of 78.9 Bq/m$^2$ (Figure 6-a) was believed to be more close to the real direct deposition of global fallout Pu in the study area, and thus is taken into account in the calculation of the contributions of Pu from both direct deposition and riverine input.

The calculation results showed that the riverine input Pu contributes to about 15% and 27% of the total Pu inventories in the NYSC-01 and NYSC-02 cores, respectively (Figure 6-b). Since plutonium has high affinity to particle and can be easily adsorbed to soil particles after deposition on the earth surface, contributions of Pu from the riverine input should directly reflect the degree of soil erosion in river catchments. In order to evaluate whether this reflection is reliable, we did a comparative study between the Yangtze River and the Yalu River catchments. Based on the previous published Pu data from the sediment cores of SC07 and SC18 collected in the Yangtze River estuary(Liu et al., 2011; Pan et al., 2011),[23-24] the contribution of Pu from the Yangtze River input to the total global fallout Pu inventory in the Yangtze River estuary was estimated to be 77-80% as shown in Figure 6-b (note that the total global fallout Pu inventories in the SC07 and SC08 cores were 244 and 217 Bq/m$^2$, respectively, corresponding to the total Pu subtracted the PPG-derived Pu, and the direct deposition of global fallout for the Yangtze River estuary was estimated to be 49.8 Bq/m$^2$ based on the data reported by Dong et al.(2010)[51]). Obviously, the riverine Pu contributions in the Yangtze River estuary are nearly 3 to 4 times higher than that in our study area, implying that the status of soil erosion in the Yangtze River's catchments might be much more serious than that of the Yalu River and other rivers in northeast China.

The above conclusion was further supported by a comprehensive comparison of the catchment area, average annual runoff and sediment discharge between these rivers as shown in Table A. S4. The average annual runoff per unit of catchment area in the Yangtze River catchment is about 1.2 times higher than that in the three major river catchments of the study area, which is reasonable and agrees with the relatively higher precipitation in the Yangtze River catchment (~1000 mm) compared to that in the Yalu River catchment (~800 mm). While the

average annual sediment discharge of the Yangtze River was estimated to be 7.5 times the total average annual sediment discharges of the three rivers in the study area, indicating the soil erosion amount in the Yangtze River catchment should be much larger than that in the river catchments of northeast China, which is consistent with the conclusion deduced from the contributions of riverine Pu. The great difference of soil erosion between Yalu River's catchment in the northeast China and Yangtze River's drainage basin in the central China should be mainly attributed to the relatively higher forest coverage and less vegetation breakage by human activities in the northeast China compared to the central China.

In addition, the sudden increase of riverine input Pu after 1980s could be generally observed in both sediments cores as shown in Figure 5, ~~which might provide more insights on evaluation of the catchment environment in the northeast China. That riverine input Pu increased rapidly after 1980s should reflect~~ this might results from the increase of soil erosion degree in the northeast China at the same period. ~~, which could be mainly attributed to land use and cover change (LUCC) in the area.~~ According to the studies of Li et al. (2010), due to the increased human activities, the land use changed intensively in the northeast China from the 1980s to 2000. During this period, cropland increased substantially, while woodland, grassland, water body and marsh decreased sharply, and among all land use types, paddy fields increased ~~fastest~~significantly (by 20 %), conversely, grassland and marsh decreased rapidly ~~fastest~~, by 17 % and 12 %, respectively. Such land use and cover change ~~should~~ result in much more soil loss in the northeast China.

## 4. Conclusions

The present work for the first time investigated the sedimentary records of $^{239}$Pu and $^{240}$Pu in the NYS. From isotopic compositions of the two Pu isotopes in surface and core sediments, we

concluded that plutonium in the northern NYS is mainly from the global atmospheric fallout without any significant contributions from NWTs of the North Korea, the FDNPP accident, as well as PPG sourced Pu. From the depth profiles of $^{239+240}$Pu activities with easily observed onset and fallout peaks in the two sediment cores, $^{239+240}$Pu was found to be a useful time marker in sediments of the northern NYS. The riverine Pu was estimated to contribute 15-27% to the total global fallout Pu inventory in the study area, which are much lower than that in the Yangtze River estuary, indicating the soil erosion in the northeast China was much less than that in the Yangtze River's drainage basin. The sudden increase of riverine input Pu after 1980s reflected the increased soil erosion degree caused by the land use and cover change due to the increased human activities in the northeast China. In addition, the activity level and the atomic ratio of Pu isotopes reported in this work can serve as a baseline for future environmental monitoring and risk assessment related to the operation of Hongyanhe NPP or any other potential nuclear activities.

**Acknowledgments**

This work was jointly supported by the National Natural Science Foundation of China (41671466, 41501286, and 41576043), Natural Science Foundation of Jiangsu Province (BK20150578) and the Fundamental Research Funds for the Central Universities (020914380010, 020914380001). Authors thank Dr. Zhongtang Wang and Wenna Huang for their supports to sample analysis, as well as Dr. Zuo Wang for his help with map drawing.

**Appendix A. Supplementary material**

**Supplementary material related to this article can be found online at**

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**Figure captions**

Figure 1 a) Overview map of the Yellow Sea with the principal currents, plotted after Huh & Su, 1999 and Su & Huh, 2002. b) Detail of the Yellow Sea and its coasts with the location of the study area being highlighted by a red rectangle. c) Detailed sampling sites in the North Yellow Sea. The red oval dotted frame indicates the location of the Yalu River-derived clinoform according to Chen et al. (2013). YSWC: Yellow Sea Warm Current; YSCC: Yellow Sea Coastal Current; LCC: Liaonan Coastal Current; WKCC: West Korea Coastal Current.

Figure 2 Distribution of $^{240}Pu/^{239}Pu$ atomic ratios in surface sediments and sediment cores in the northern NYS. The red dDashed lines indicate the global fallout value of ~0.18±0.02.

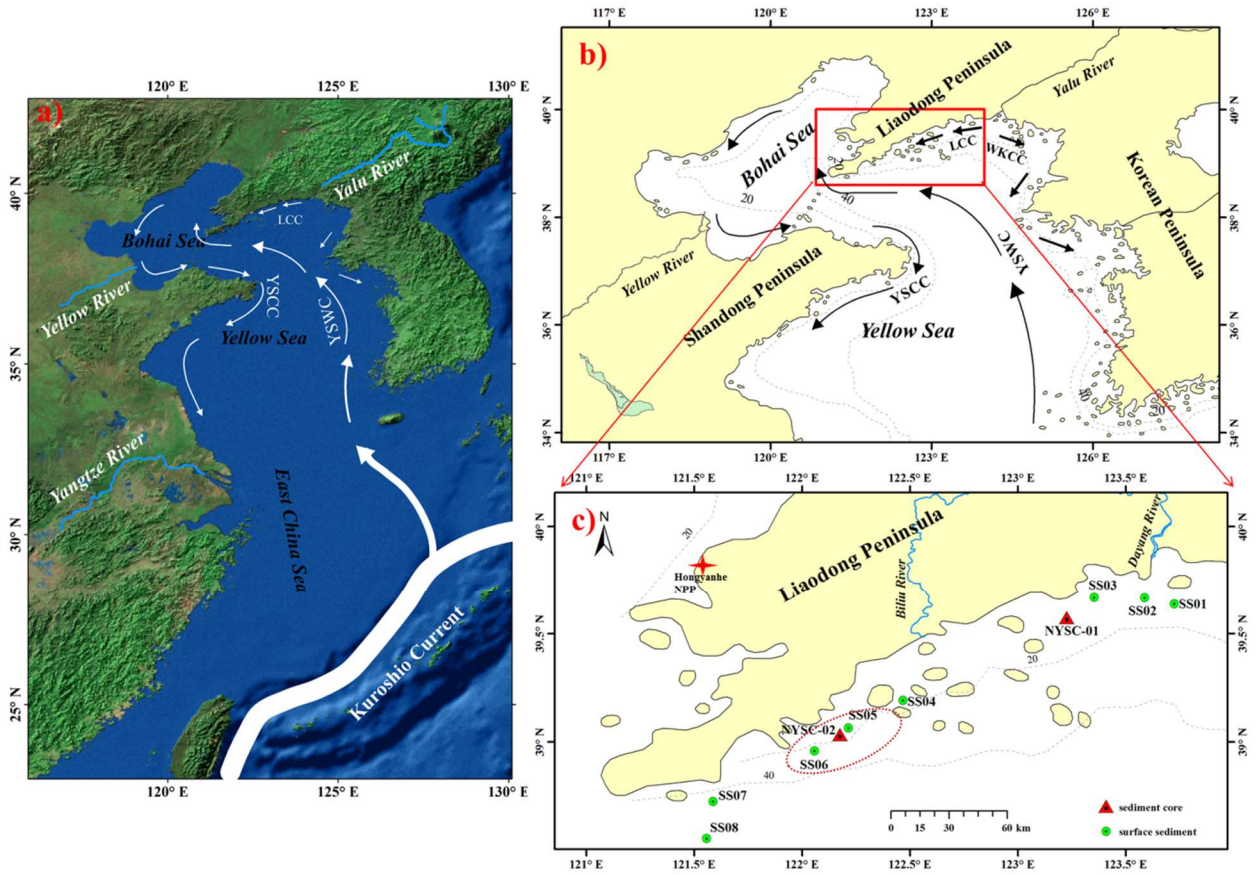
Figure 3 Distribution of the $^{239+240}Pu$ activities in all surface sediments (including the surface layer (0-2 cm) of the two sediment cores NYSC-01 and NYSC-02) along the southeast coast of the Liaodong Peninsula. The red oval dotted frame indicates the location of the Yalu River-derived clinoform according to Chen et al. (2013).

Figure 4 Vertical profiles of $^{239+240}$Pu activities in sediment cores collected from the northern NYS.

Figure 5 Abundance of annual deposition of $^{137}$Cs in northern hemisphere and abundance of deposition of $^{239+240}$Pu in each layer of sediments. The annual deposition data of $^{137}$Cs in northern hemisphere were cited from the UNSCEAR (2000).[5] Data of Pu in Lake Hongfeng and Qinghai were respectively cited from Zheng et al.(2008)[10] and Wu et al.(2011)[11] and mainly originated from the global fallout direct deposition.

Figure 6 a) Comparison of $^{239+240}$Pu inventories between the northern NYS and the Yangtze River estuary. $^{239+240}$Pu inventories for ~31ºN and ~39ºN were calculated as the averages of the data reported respectively by Dong et al.(2010)[51] and by Xu et al.(2013, 2015) [31, 33] and Hardy et al.(1973) [50] as shown in Table A.S3. b) Comparison of the relative contributions of Pu from the direct global fallout deposition and riverine input between the northern NYS and the Yangtze River estuary.

Figure 1

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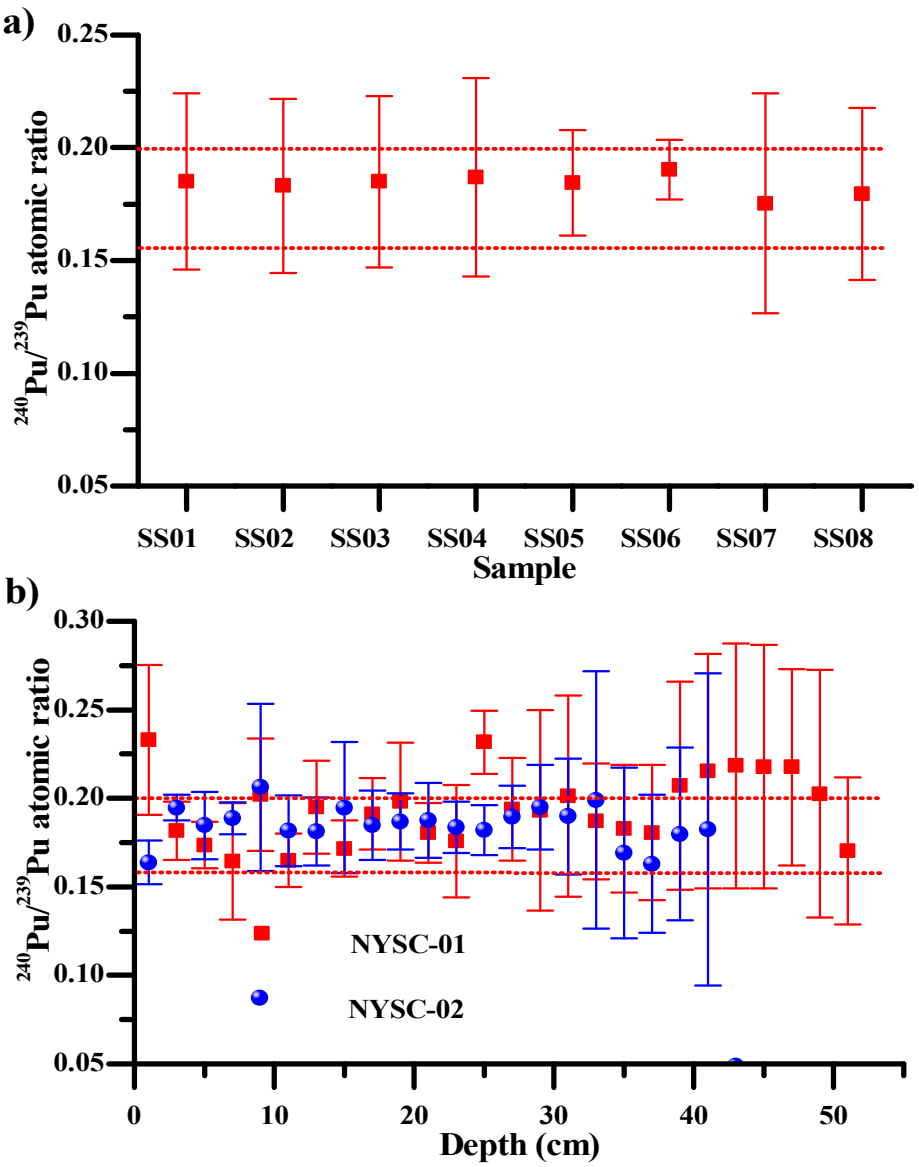
600 Figure 2

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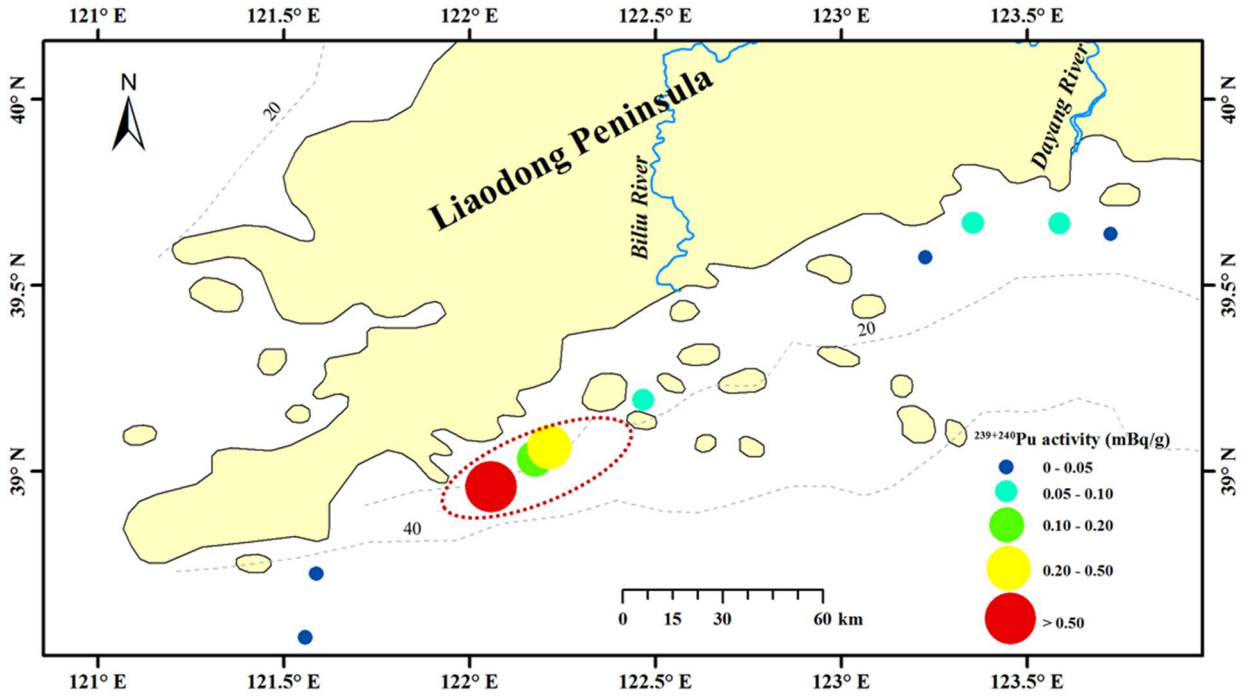
603 Figure 3

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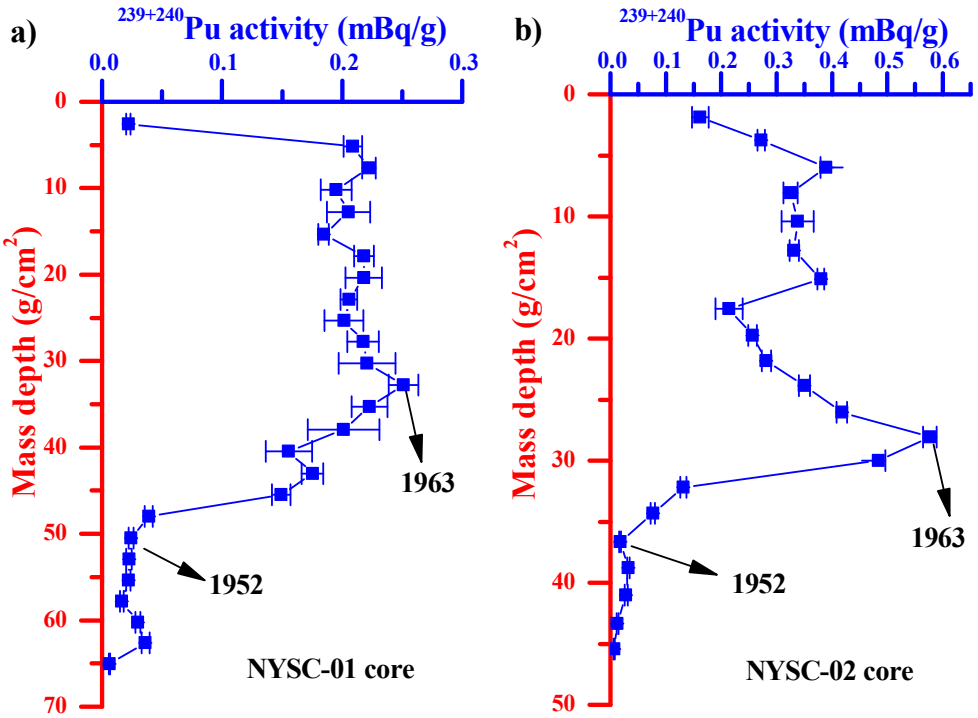


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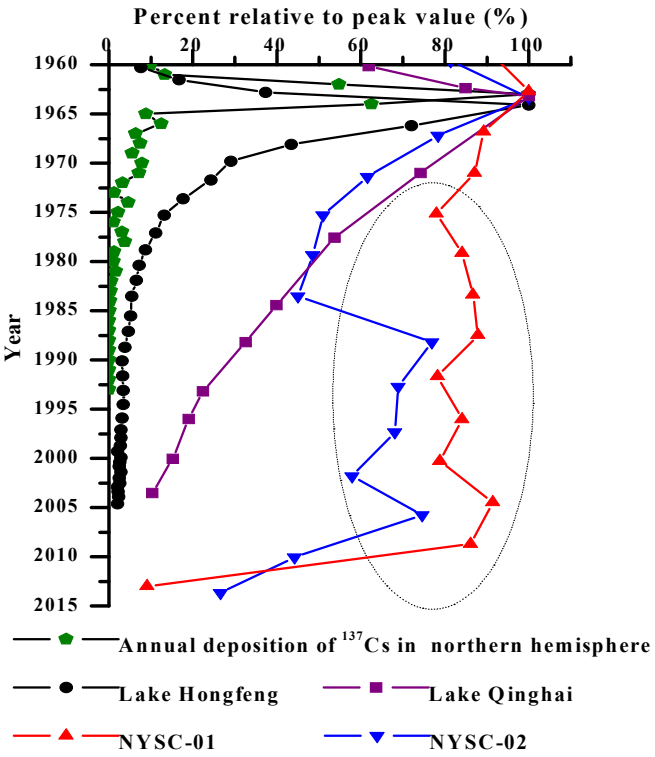


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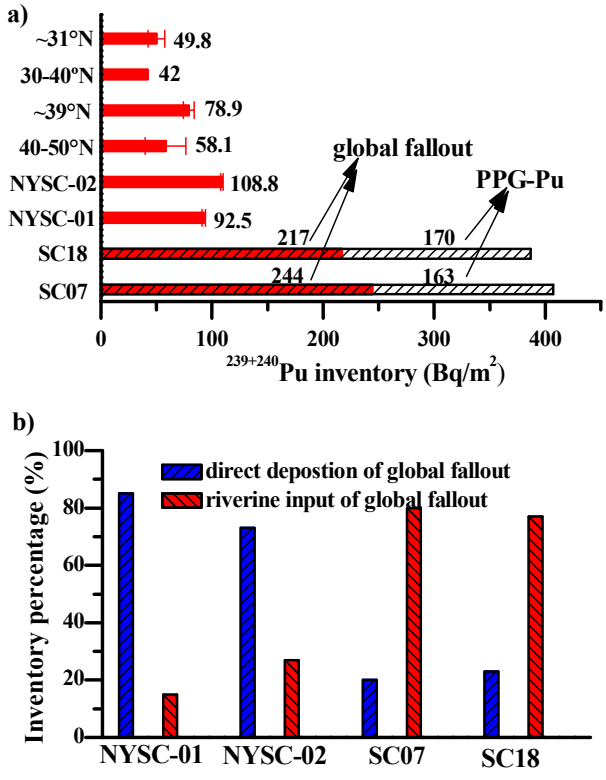
610 Figure 6

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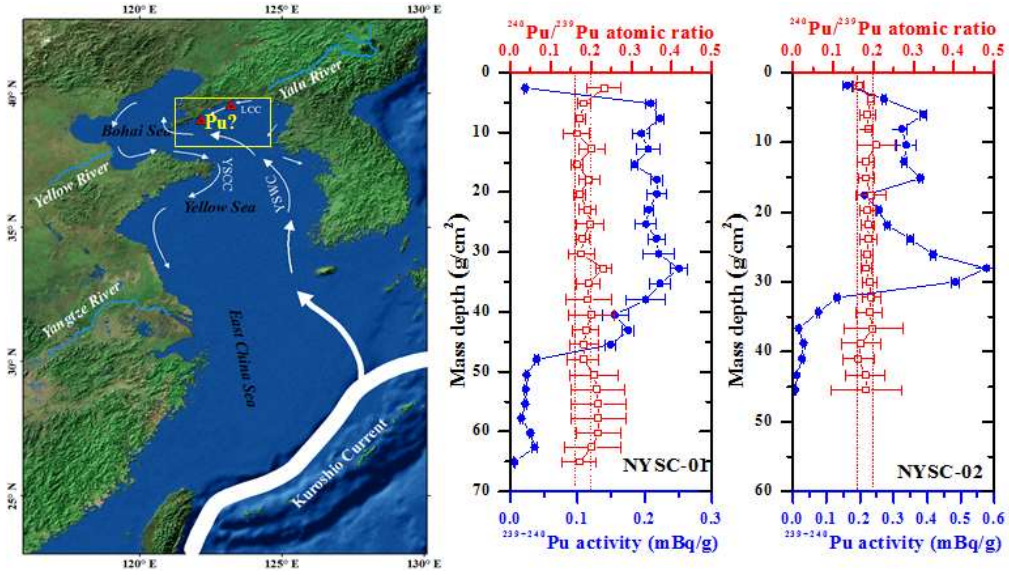


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