^{129}I record of nuclear activities in marine sediment core from Jiaozhou Bay in China

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Abstract

Iodine-129 has been used as a powerful tool for environmental tracing of human nuclear activities. In this work, a sediment core collected from Jiaozhou Bay, the east coast of China, in 2002 was analyzed for ^{129}I to investigate the influence of human nuclear activities in this region. Significantly enhanced ^{129}I level was observed in upper 70 cm of the sediment core, with peak values in the layer corresponding to 1957, 1964, 1974, 1986, and after 1990. The sources of ^{129}I and corresponding transport processes in this region are discussed, including nuclear weapons tests at the Pacific Proving Grounds, global fallout from a large numbers of nuclear weapon tests in 1963, the climax of Chinese nuclear weapons testing in the early 1970s, the Chernobyl accident in 1986, and long-distance dispersion of European reprocessing derived ^{129}I . The very well ^{129}I records of different human nuclear activities in the sediment core illustrate the potential application of ^{129}I in constraining ages and sedimentation rates

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of the recent sediment. The releases of ^{129}I from the European nuclear fuel reprocessing plants at La Hague (France) and Sellafield (UK) were found to dominate the inventory of ^{129}I in the Chinese sediments after 1990, not only the directly atmospheric releases of these reprocessing plants, but also re-emission of marine discharged ^{129}I of these reprocessing plants in the highly contaminated European seas.

1. Introduction

^{129}I , as an environmental tracer, has attracted much attention of researchers during the past decades due to its anthropogenic origin associated with human nuclear activities, long half-life (15.7 Ma), and ready mobility in the environment (high volatility and high water solubility). ^{129}I in environmental samples provide valuable information for the assessment of environmental impact of human nuclear activities. Recent studies have unraveled the complicated physical and chemical pathways that distinguish ^{129}I from natural ^{127}I in a range of environments. Speciation analysis of both isotopes from the same samples provides a comprehensive picture of the sources and pathways followed by the two isotopes (He et al., 2014; He et al., 2013; Hou et al., 2007; Hou et al., 2000; Yi et al., 2012). The levels observed in surface environmental samples are, in general, stacked signals with contributions from a number of historical sources. These have been exploited in some case to extract valuable time series information, using for example, precipitation or seaweed samples collected at regular intervals (Hou et al., 2009a; Hou et al., 2000; Toyama et al., 2013; Xu et al., 2013). Natural archives offer an alternative means of extracting time series data that avoids the need

41 to collect samples periodically. Examples include tree rings, ice cores and corals, all
42 of which accumulate ^{129}I continuously (Hauschild and Aumann, 1985; Biddulph et al.,
43 2006; Reithmeier et al., 2006). These types of samples are widely distributed and have
44 been used successfully to study historical releases of ^{129}I into the environment in a
45 number of locations around the globe. Marine and lake sediment cores from Europe
46 have shown to preserve valuable archives of ^{129}I (Aldahan et al., 2007; Englund et al.,
47 2008; Santos et al., 2007). Sediment cores from the Baltic Sea, Kattegate, and
48 Swedish lakes all reflect the fact that the two European nuclear fuel reprocessing
49 plants (NFRPs) are the dominant source of environmental ^{129}I in the Europe.

50 Time series records are particularly important in understanding the spread of ^{129}I
51 following accidental releases. The Fukushima Daiichi nuclear accident that occurred
52 in March 2011 spurred a number of timely investigations to study the dispersion of
53 ^{129}I into the environment (Hou et al., 2013; Masson et al., 2011; Shi et al., 2014; Xu et
54 al., 2015; Guilderson et al., 2014; Honda et al., 2015; Tumey et al., 2013; Zhang et al.,
55 2014). ^{129}I time series were determined from precipitation samples collected over a
56 two years period (2010-2012) at Fukushima, about 70 km from the Fukushima
57 Daiichi nuclear power plant facility. A sudden increase in ^{129}I levels of 3-4 orders of
58 magnitude was observed immediately following the accident, with an exponential
59 decrease with time, excepting some sporadic increases after about one year. These
60 were attributed to re-emission or re-suspension of ^{129}I from the local environment (Xu
61 et al., 2013). Aside from the studies of the Fukushima disaster, environmental ^{129}I
62 studies from East Asia are rare, and no time series record of ^{129}I in China is available.

This work aims to investigate the environmental history of ^{129}I deposited in North China since 1930, and the sources and transport processes of this isotope within the hydrosphere by analyzing a sediment core collected in Jiaozhou Bay, North China.

2. Experimental section

2.1 Sample collection and characterization

A sediment core of 116 cm length was collected using a gravity corer in Sep. 2002 at Jiaozhou Bay (36.09°N, 120.24°E), Yellow Sea, China (Fig. 1). Jiaozhou Bay is characterized by high runoff input, with more than 10 rivers contributing to the sediment load into the bay. It has been reported that the marine input to Jiaozhou Bay was approximately 1×10^4 tons while the terrestrial discharge reached up to 163.5×10^4 tons (Wang et al., 1982). High content of suspended particulate matter in the rivers flowing into the bay contributes to a rapid sedimentation rate in the Jiaozhou Bay. A sedimentation rate of 1.64 cm/yr at the sampling site has been estimated based on ^{210}Pb and ^{137}Cs dating (Liu et al. 2008). A peak value of ^{137}Cs in the sediment core from Jiaozhou Bay has been observed at a depth of 62.7 cm, corresponding to the maxima in atmospheric fallout in 1964 (Liu et al. 2008). The entire sediment core consists of grey clay with low detritus content (Yi, 2006). The top 36 cm of the sediment core was sliced into 1-cm intervals, and the lower part (36-116 cm) was sliced into 2 cm intervals for analysis.

2.2 Separation of iodine from sediment

The sliced sediment samples were first freeze-dried, then ground and sieved through a 200 mesh sieve. 3.0 gram of sediment samples was taken to in a quartz boat, 200 Bq ^{125}I as a chemical yield tracer was spiked. Iodine in the sample was then separated by combustion in a four-tube furnace (Catalytic Pyrolyser-4 Trio^{TM} furnace, Raddec LTD, Southampton, UK) at 800°C for 3 hours. The released iodine was trapped in 30 ml of 0.5 M NaOH-0.02 M NaHSO₃ solution. The detailed combustion system and method have been reported elsewhere (Zhou et al., 2010). 3.0 mL of the trapped solution was taken to measure ^{125}I using a gamma counter (model FJ-2021, Xi'an Nuclear Instrument factory, Xi'an, China) for monitoring the recovery of iodine in this step. The measured recoveries of iodine were better than 95%. After measurement of ^{125}I , the trap solution was analyzed for stable iodine (^{127}I) using inductively coupled plasma mass spectrometry (ICP-MS). The remaining trap solution was used to separate iodine for ^{129}I measurement. A 0.2 mmol of NaHSO₃ and 0.50 mg ^{127}I carrier (prepared from iodine crystal provided by Woodward Iodine Corporation, MICAL Specialty Chemicals, New Jersey, $^{129}\text{I}/^{127}\text{I}$ atomic ratio is $<2 \times 10^{-14}$) were added to the solution, and then 6 M HNO₃ was added to adjust the pH < 2, to convert all of the iodine to iodide. 0.5 mmol of AgNO₃ was added to each sample solution to precipitate iodide as AgI, which was then separated from the supernatant by centrifuge. The precipitate was washed with HNO₃ to remove excessive Ag₂SO₃, and rinsed repeatedly with H₂O until neutral. The separated AgI was dried, ground to a powder

and then mixed with Nb powder with a mass ratio of AgI:Nb about 1:5. The mixtures were pressed into copper target holders for AMS measurement of ^{129}I . A procedural blank was prepared using the same method as for samples for each batch of samples (normally every 7 samples with 1 blank).

2.3 Measurement of ^{129}I and ^{127}I

$^{129}\text{I}/^{127}\text{I}$ atomic ratios in the prepared targets were measured by accelerator mass spectrometry (AMS) using the 3 MV Tandem AMS system at the Xi'an AMS center. Iodine ion (I^-) is sputtered from the AgI target in the ion source, and stripped to the I^{5+} charge state in the tandem accelerator. The charge (current) produced by $^{127}\text{I}^{5+}$ was measured in a Faraday cup and $^{129}\text{I}^{5+}$ was measured using a gas ionization detector. Two ^{129}I standards with $^{129}\text{I}/^{127}\text{I}$ atomic ratios of 9.95×10^{-12} and 1.14×10^{-10} were prepared by dilution of an ^{129}I standard solution (NIST-SRM-4949c) with ^{127}I carrier solution, and prepared as AgI precipitate. These were measured along with samples and blanks to correct the $^{129}\text{I}/^{127}\text{I}$ atomic ratios in the samples. The measured $^{129}\text{I}/^{127}\text{I}$ ratios in the procedural blanks were lower than 2×10^{-13} , which are more than 2 orders of magnitude lower than those in the samples. The procedure blank in each batch of samples was used to calculate blank-corrected ^{129}I concentrations for the corresponding batch of samples.

The trapping solution was diluted 10 times using deionized water (18.2 M Ω) and a Cs^+ solution was added to the samples, blanks and standards to a final concentration of 2.0 ppb as an internal standard. Iodine (^{127}I) concentrations in the trap solutions

were determined by ICP-MS using an X SeriesII ICP-MS (Thermal Electron Corporation), under hot plasma conditions, with an Xt interface. The detection limit of this method for iodine is estimated as 3 times blank to be 0.02 ppb. An 0.15 mol/L $\text{NH}_3 \cdot \text{H}_2\text{O}$ solution was used to rinse the samples during the ICP-MS measurements.

3. Results

The analytical results of ^{127}I and ^{129}I in the sediment core collected in Jiaozhou Bay are shown in Table 1. ^{127}I concentrations in sediments from Jiaozhou Bay range from 15.4 to 70.7 $\mu\text{g/g}$ with an average of 27.3 $\mu\text{g/g}$. ^{129}I concentrations in the sediment in Jiaozhou Bay vary from 1.3×10^5 atoms/g to 199.1×10^5 atoms/g in the top layer with an average of 76.0×10^5 atoms/g. Fig. 3 presents depth profiles of ^{129}I and ^{127}I concentrations as well as $^{129}\text{I}/^{127}\text{I}$ atomic ratios in the sediment core. The ^{127}I concentrations are relatively consistent in the entire core with one exceptional high value (70.7 $\mu\text{g/g}$) at 24 cm depth, and two exceptional low values (15-17 $\mu\text{g/g}$) in the top layer (2.8 cm) and deep layer (78.5 cm). ^{129}I concentrations in the sediment core show a large variation from 1.3×10^5 in the deep layer to 199.0×10^5 atoms/g in the top layer. $^{129}\text{I}/^{127}\text{I}$ ratios in the sediment core show similar variations as ^{129}I concentrations with the lowest level in the deep layer (1.2×10^{-12}) and the highest level in the top layer (189.0×10^{-12}) because of the relative constant ^{127}I concentrations in the core. Both ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios decreased from top to bottom with strong fluctuations at depths of 0-75 cm, reflecting varied input during the past decades.

4. Discussion

4.1 ^{127}I and ^{129}I level in the sediment core

^{127}I concentrations in sediments from Jiaozhou Bay (15.4-70.7 $\mu\text{g/g}$, mostly 20-36 $\mu\text{g/g}$) are compared with the ^{127}I concentrations in sediments from other sites in Fig. 2a, including the Baltic Sea, Mississippi River and the Eastern Pacific Ocean (Aldahan et al., 2007; Moran et al., 1998; Oktay et al., 2000). ^{127}I concentrations in most marine sediments are higher than in our samples, aside from several exceptions. ^{127}I concentrations for samples from the Mississippi River are lower than all other samples, even those from Jiaozhou Bay. This is because iodine concentrations are generally much higher in seawater compared to fresh water, and marine sediments typically contain more iodine than freshwater sediments. The large terrestrial input to Jiaozhou Bay effectively dilute the supply of ^{127}I , with only a minor pure marine component, resulting a relatively lower ^{127}I concentration compared to most of other marine sediment samples. The low ignition loss (about 2-4%) in the investigated sediments confirms a significant input of continental inorganic material (Table 2). The ^{127}I concentrations are relatively consistent in the entire core with one exceptional high value (70.7 $\mu\text{g/g}$) at 24 cm depth, and two exceptional low values (15-17 $\mu\text{g/g}$) in the top layer (2.8 cm) and deep layer (78.5 cm). This constant ^{127}I level suggests a stable source of sediment material. This can be also interpreted by the high proportion (>80%) of fine particles (<0.063 mm) with less variation in all the layers in the investigated sediment core (Table 2).

The average (76.0×10^5 atoms/g) and range ($(1.3-199) \times 10^5$ atoms/g) of ^{129}I concentrations in the sediment in Jiaozhou Bay, China are distinctly lower than sediments from Europe, including Tinto River (Spain), the Baltic Sea and Loppesjön Lake (Sweden) (Fig.2b). This difference should be attributed to the strong regional influence in the European sites by the European NFRPs. A 2-3 orders of magnitude higher ^{129}I level in the atmospheric samples in the Europe compared to those in Asia has been reported. (Hou et al. 2009a, Toyama et al. 2013, Zhang et al. 2011; 2015)

4.2 Possible sources of ^{129}I in the sediment core

Relatively constant $^{129}\text{I}/^{127}\text{I}$ ratios were observed in the deep layers (about 76-116 cm depth), in the range of $(1.21-4.48) \times 10^{-12}$. They are lower than values obtained in most environmental samples, suggesting a minor influence of anthropogenic ^{129}I , and representing a pre-nuclear value in this core. The pre-nuclear ratio of $^{129}\text{I}/^{127}\text{I}$ has been extensively investigated in the past. An estimated value of $(3-4 \times 10^{-12})$ and a predicted value of $(5.5-6.5 \times 10^{-13})$ were reported by early efforts to use ^{129}I as an environmental tracer (Edwards, 1962; Fabryka-Martin et al., 1985). Baseline $^{129}\text{I}/^{127}\text{I}$ ratios of $(1.2-1.5) \times 10^{-12}$ have been reported for recent marine sediment cores in the East Pacific and Atlantic Oceans (Fehn et al., 1986), and a ratio of 1.5×10^{-12} has been widely adopted as the “initial value” of $^{129}\text{I}/^{127}\text{I}$ atomic ratio for ^{129}I dating of geological samples (Fehn et al., 1986; Fehn et al., 2007; Fehn et al., 2000; Lu et al., 2008; Moran et al., 1998; Perez-Fodich et al., 2014; Snyder and Fehn, 2002). Our data of $^{129}\text{I}/^{127}\text{I}$ ratios measured in the deep sediment layer (76-116 cm depth) are

comparable with this “initial value”. The consistency within such a large spatial scale (from both the Pacific and Atlantic Oceans) demonstrates a uniform global distribution of iodine isotopes in the marine system with a long residence time ($\sim 4 \times 10^4$ years) relative to an ocean mixing time of roughly 1000 years (Fabryka-Martin et al., 1985). This result also confirms that once fixed in the sediments iodine rarely moves vertically (Moran et al., 1998), and migration of high level ^{129}I from the top layers to the deep ones can be excluded. A significantly enhanced ^{129}I level in the upper 75 cm of sediment in Jiaozhou Bay must result from anthropogenic input through human nuclear activities.

The possible sources of the anthropogenic ^{129}I in the upper part of the sediment core in Jiaozhou Bay might include: 1) direct discharges of ^{129}I from local nuclear facilities, including those located in the vicinity of the bay and in catchment of the rivers which drain into the bay; 2) atmospheric fallout, including atmospheric releases from nuclear weapons testing (NWT) as well as direct and indirect atmospheric releases from nuclear facilities; 3) ocean current transport of ^{129}I released to the seawater from the NWT in the Pacific Ocean and nuclear facilities such as nuclear fuel reprocessing plants (NFRP) and nuclear power plants (NPP) located nearby the sampling region. Since there were no nuclear facilities in operation at or close to Jiaozhou Bay as well as in the catchment of the rivers flowing into the Bay earlier than the sampling date (2002), the influence of local nuclear facilities to ^{129}I in the sediment in the Jiaozhou Bay can be excluded. Due to active chemical properties, iodine is easy to exchange

among various reservoirs in the environment. ^{129}I released by the nuclear activities including atmospheric NWT in 1950's-1970's, NFRPs, and nuclear accidents to the atmosphere could be transported to remote regions via atmospheric dispersion and deposited as atmospheric fallout, resulting in increased ^{129}I levels in remote areas (Biddulph et al., 2006; He et al., 2013; Negri et al., 2013; Oktay et al., 2000; Smith et al., 2011; Xing et al., 2015; Zhang et al., 2011).

Based on ^{137}Cs data measured in the same sediment core, Liu et al. (2008) has estimated an average sedimentation rate of 1.64 cm/yr, which was used to date each slice of this sediment core. Fig. 4a shows the variation of ^{129}I level with time. As a comparison, the estimated ^{129}I releases from NWTs, the annual discharges of ^{129}I into the marine and atmosphere from the two European NFRPs and Japanese NFRP in Tokai as well as temporal variations of ^{137}Cs activity (decay corrected to the time of deposition) (Liu et al., 2008) are also presented in Fig. 4. These data provide a framework to understand ^{129}I sources in each segment of the core within a 70-year time frame (1931-2000).

4.3 Contribution of nuclear weapons testing

Four ^{129}I peaks were observed in the sediment core from depths of 76 cm to 19 cm, corresponding to 1955-1990 (Fig. 4a). An obvious but relatively small peak occurs around 1957 (depth 75 cm), which might be attributed to the contribution of NWT in 1955-1957 at the Marshall Islands in the South Pacific Ocean. Besides the direct

229 atmospheric fallout, ^{129}I can be also dispersed in a large area and transported to
230 remote areas through sea currents because of the very high water solubility of iodine.
231 (He et al., 2013; Hou, 2004; Hou et al., 2009a; Hou et al., 2007; Wong, 1991). ^{129}I
232 discharged from NFRPs at La Hague and Sellafield to the English Channel and Irish
233 Sea have been transported a long distance to the Arctic and Greenland Seas through
234 the North Sea and Norwegian Sea (Hou et al., 2007). ^{129}I discharged from the NFRP
235 in Marcoule (France) has also been transported to the North Atlantic through the
236 Mediterranean Sea and Strait of Gibraltar (He et al., 2013). In 1954-1957, 67 NWT
237 were conducted at the Pacific Proving Grounds (PPG) in the Marshall Islands, mostly
238 at Bikini Island and Enewetak Atolls as atmospheric tests
239 (https://en.wikipedia.org/wiki/Nuclear_testing_at_Bikini_Atoll#Weapons_tests). The
240 largest ones among them were carried out in 1954 (about 30 Mt TND), 1956 (total
241 about 20 Mt TNT) and 1958 (total about 30 Mt TNT). The ^{129}I injected from these
242 nuclear weapon tests to the sea through close-in fallout in the atmospheric tests and
243 direct contact with seawater during underwater tests could be transported to the
244 Jiaozhou Bay area by surface ocean currents through the northwest Pacific Ocean (Fig.
245 1). The Yellow Sea Warm Current (YSWC), a branch of the Kuroshio Current, which
246 moves along the west coast of the Korean Peninsula towards the north, turns to the
247 south at Liaodong Peninsula, and finally flows into the Jiaozhou Bay. This current
248 could therefore bring the ^{129}I released from the NWTs in PPG to the Jiaozhou Bay
249 region. A plutonium signal from NWTs at the PPG has been observed in the Yangtze
250 River estuary in the East China Sea, about >600 km south of Jiaozhou Bay, which was

supposed being transported to there by sea currents (Wang and Yamada, 2005). A measurement of ^{129}I depth profiles in coral samples collected in the east and west coastal areas of the Philippines has shown an obvious ^{129}I signal in year 1956-1958 (Bautista et al., 2015), confirming that radionuclides released from nuclear weapons tested in the Marshall Islands in 1954-1957 has been transported to distant areas and could reach the South China Sea and China coast at Jiaozhou Bay through sea currents.

The second ^{129}I peak in the sediment core from Jiaozhou Bay occurs at depth of 62.7 cm, corresponding to around 1964. A peak value of ^{137}Cs found in the same layer in this sediment core has been regarded as maxima atmospheric fallout in 1964 in the sediment and was used as time marker for the whole sediment core (Liu et al., 2008, mentioned in section 2.1). The fallout signal in 1964 has been recorded in many other sediment cores collected in China and other locations all over the world by other anthropogenic radionuclides (Appleby, 2013; Baskaran et al., 2014; Liu et al., 2013; Rowell et al., 2015; Santos et al., 2007; Oktay et al., 2000; Wu et al., 2010). The global NWTs in the early 1960s by USA and USSR also released considerable amount of radioactive substance to the atmosphere including ^{129}I into the atmosphere, which is characterized by the distinct peak in the estimated annual ^{129}I releases from the NWTs (Fig. 4c). Therefore, the peak value observed in the layer related to 1964 should refer to the contribution of the NWTs.

The third ^{129}I peak in the sediment core from Jiaozhou Bay is observed at depth of

272 42-46 cm, corresponding to 1972-1976. A ^{137}Cs peak at a similar depth in this
273 sediment was also observed (Liu et al., 2008). ^{137}Cs peaks around 1974 have been
274 reported in many Chinese lake sediments, including Hongfeng, Dianchi, Jianhu, and
275 Donghu lakes (Chen et al., 2007; Norris et al., 1994; Wan et al., 1990; Xiang et al.,
276 1996; Yang et al., 2004), and this signal was attributed to the fallout from some large
277 atmospheric NWTs (1-4 Mt TNT) completed in 1974 and 1976 at Lop Nor (41°N,
278 88°E) in northwestern China.
279 (https://en.wikipedia.org/wiki/List_of_nuclear_weapons_tests_of_China).

280 Jiaozhou Bay is located in the downwind direction of Chinese NWT site in Lop Nor,
281 with westerly winds dominating in this area (Carter and Moghissi, 1977). The
282 radionuclides released from these weapons testing in the northwestern China can be
283 directly transported to Jiaozhou Bay by wind. Atmospheric deposition on land,
284 especially in the catchments of the rivers that flow into the Jiaozhou Bay such as
285 Dagou, Yang and Xin'an river in east China, can also transport fallout from Chinese
286 NWTs to Jiaozhou Bay. The fallout of the weapons testing which deposited in the
287 catchments of the Yellow River could be transported to the Bohai Bay, where it is
288 mixed with the seawater, and might be transported towards south to Jiaozhou Bay via
289 the Yellow Sea Coastal Current (YSCC, Fig. 1). The Yellow River, more than 5000
290 km in length and flowing from west to east, is the second longest river with a
291 catchment covering a big area in west and north part of China where the fallout of
292 Chinese NWT deposited.

4.4 Record of the Chernobyl nuclear accident

The fourth ^{129}I peak in the sediment core from Jiaozhou Bay appears around 1985-1987. After the last atmospheric NWT in 1980, the anthropogenic source of ^{129}I to the atmosphere from the NWTs was cut off. This was disrupted by the nuclear accident at Chernobyl which happened in April 1986. This event released large amounts of radioactive substances including 1.3-6 kg ^{129}I into the environment (Hou et al., 2009b). Although most of this radioactive material was deposited in the vicinity of Chernobyl NPP, an enhanced signals of ^{129}I and ^{137}Cs have been observed at locations of great distances (Englund et al., 2008; Hou et al., 2003; Michel et al., 2015; Paul et al., 1987). Chernobyl derived radionuclides including ^{131}I have also been measured in a large area in China in various environmental samples including aerosols, vegetables and milk, and relatively high levels of ^{131}I were found the north part of China after the accident, including the Jiaozhou Bay area and the catchments of the Yellow River and others flowing into the bay. The highest daily deposition rate of 100 Bq/m²/day was observed in Northeast China, and a total deposition of 1.2-3.0 PBq ^{131}I (0.5-0.7% of total ^{131}I releases from the Chernobyl accident) in China has been estimated (Zhu et al. 1992). The Chernobyl-derived ^{129}I should also been transported to Jiaozhou Bay, because it follows the same pathways as ^{131}I . Therefore, the fourth ^{129}I peak in the sediment core from Jiaozhou Bay should result from the Chernobyl accident in 1986. The early occurred ^{129}I peak corresponding to 1985 might be attributed to the low resolution inherent in the dating of the sediment core

(segment responding to 2-3 years was sampled for ^{129}I analysis) and the assumption of a constant deposition rate. It should be mentioned that no ^{137}Cs peak corresponding to 1986 was observed in the sediment core. This may be due to the different chemical properties and environmental behaviors of radiocesium and radioiodine. Besides aerosol associated form, a large fraction of radioiodine exists in gaseous form which can be transported a longer distance in the air. Meanwhile radioiodine is water-soluble compared to radiocesium, and therefore can be leached to water in the catchment area to be finally transported a long distance through rivers.

4.5 Contribution of nuclear fuel reprocessing plants

^{129}I level in the sediment core significantly increased from 1990 and remained at high levels until the sampling date in 2001. In this period, ^{137}Cs levels in the core followed a steady decrease. The atmospheric nuclear weapon tests ceased after 1980, and no accidental radioactive releases after 1990 have been reported. This increased ^{129}I signal should be attributed to continuous releases from other nuclear activities. NPPs produce large quantities of ^{129}I as a fission product in the fuel elements used in nuclear power reactors. However, the ^{129}I produced in the spent fuel elements is normally well sealed inside the fuel elements and could only leak into the environment during a nuclear accident. This has been confirmed by the measurement of ^{129}I level in the vicinity of nuclear power plants. The $^{129}\text{I}/^{127}\text{I}$ ratios in seawater collected near a Chinese NPP have been reported to be 8.29×10^{-11} - 9.45×10^{-10} , which is comparable to background environmental water samples in China (He et al., 2011;

Snyder et al., 2010). The primary source of the ^{129}I to the environment since 1990 should be attributed to NFRP facilities. Spent fuel elements are processed there to extract useful radionuclides. As a result, ^{129}I is released from the fuel elements into the environment. NFRPs are now the dominant source of ^{129}I on the planet, primarily from liquid discharges to the ocean and from gaseous discharges to the atmosphere (Hu et al., 2010; Fan et al., 2013; Snyder et al., 2010; Xing et al., 2015).

The nearest NFRP to the sampling site in Jiaozhou Bay is the Tokai-mura NFRP, 130 km northeast of Tokyo, Japan. This NFRP facility has been operated in 1977-2007 and released ^{129}I of 0.65-0.97 GBq/yr in 1977-1986 and less than 0.32 GBq/yr in 1988-1997 to the atmosphere (Fig. 4f). Coincident increases of ^{129}I in the precipitation collected in Tokyo and Tsukuba (150 km and 100 km down wind direction to the Tokai NFRP) have been reported (Toyama et al., 2013; Toyama et al., 2012). However, the effect of the Tokai BFRP on ^{129}I levels in precipitation from Akita, on the west coast of Japan (600 km northwest of Tokai NFRP), and Ishigaki, on the south coast of Japan (2300 km to Tokai NFRP) have not been detected. This was attributed to the relatively small releases from the Tokai NFRP. Meanwhile the dominant wind direction at these two locations are not from the Tokai NFRP. Jiaozhou Bay and the catchment area of the rivers flowing into this Bay are located in the distant area (> 1000 km to the Tokai NFRP) and in the west and northwest of the Tokai NFRP, with the dominant westerly wind in this area. Consequently, the contribution of the Tokai NFRP to the ^{129}I in the sediment core in Jiaozhou Bay

should be relatively small. Furthermore, no coincidence between ^{129}I levels in the sediment core and releases from the Tokai NFRP in 1977-1986 are observed.

The two European NFRPs at La Hague (France) and Sellafield (UK) have contributed more than 90% of the ^{129}I in the present environment through marine and atmospheric discharges. Significantly enhanced ^{129}I levels have been observed in European seawater (North Sea, Baltic Sea, Northern North Atlantic and the Arctic) and the European continent (Raisbeck et al., 1995; Jabbar et al., 2013; Snyder et al., 2010). The increased ^{129}I levels observed in Akita in Japan since 1980 were attributed to ^{129}I releases from the two European NFRPs (Toyama et al., 2013). Increased levels of ^{129}I observed in precipitation in the U.S. in the 1990's were also attributed to the releases of these two European NFRPs (Moran et al., 1999). Atmospheric releases of ^{129}I from the two reprocessing plants (Fig. 4e) show consistently high levels from 1980 to 1997. This might explain a relatively elevated ^{129}I levels in our core in 1981-1987, but cannot explain the increased levels of ^{129}I since 1990.

An increased and steadily large amount of ^{129}I was discharged into the seas from the two European NFRPs at La Hague (France) and Sellafield (UK) since 1990 (Fig. 4d). The annual marine discharges of ^{129}I after 1990 are about 2 orders of magnitude higher than the direct atmospheric discharges from these two NFRPs. Once discharged to the seas, ^{129}I has been transported northwards by surface ocean currents to the Arctic through the North Sea and Norwegian coast. A high level of ^{129}I with $^{129}\text{I}/^{127}\text{I}$ ratios of 10^{-8} - 10^{-6} has been observed in the North Sea, Norwegian Sea and

the Arctic Ocean. These are 3-5 orders of magnitude higher than those from other locations (Cooper et al., 2001; Hou et al., 2007; Smith et al., 2011; Yiou et al., 2002). The ^{129}I discharged into sea can be re-emitted to the atmosphere through volatile iodine molecules formed on the sea surface. This re-emission has elevated $^{129}\text{I}/^{127}\text{I}$ ratios to 10^{-8} - 10^{-6} in rain water and aerosols in Denmark and Sweden (Aldahan et al., 2009; Hou et al., 2009a; Zhang et al., 2015). The increased ^{129}I observed in the sediment core from Jiaozhou Bay agrees well with the increased marine discharges from the European NFRPs at that time. It can be argued that re-emission of ^{129}I into the atmosphere is actually the primary source of ^{129}I in this period. Re-emitted ^{129}I to the atmosphere can be transported to Asia through westerly winds, and become deposited in the area of Jiaozhou Bay and in the river catchments upstream from the bay. More works on the investigation of ^{129}I in river and sediment as well as soil are going to be implemented in our laboratory in the near future for a better understanding of the spatial ^{129}I variation and environmental impact of nuclear activities in China.

The annual rate of ^{129}I re-emitted from sea surface to the atmosphere has been reported to be around 0.3% of the ^{129}I inventory in the upper layer of the ocean (Reithmeier et al., 2006). Due to the large marine discharges from the European NFRPs (22.7 TBq or 1600 kg up to 2000), ^{129}I re-emitted from the sea surface can be estimated to be 68 GBq in 2000, which is 2-3 times higher than the annual atmospheric releases. In addition, the different species of ^{129}I between those released directly to the atmosphere (primarily inorganic forms, e.g. I_2 and HI) and those

re-emitted from seawater (primarily organic forms, e.g. CH₃I) (Hou et al., 2011) have different atmospheric residence times. ¹²⁹I species released directly to the atmosphere have a shorter residence time than those re-emitted from seawater, leading to enhanced local deposition of atmospherically released ¹²⁹I (Frechou and Calmet, 2003; Wershofen and Aumann, 1989).

The well preserved ¹²⁹I records of the human nuclear activities in the sediment core showed in this work suggest a potential application of ¹²⁹I for dating recent sediments. Liking other anthropogenic radionuclides such as ¹³⁷Cs and ^{239, 240}Pu (Appleby, 2013; Baskaran et al., 2014; Oktay et al., 2000), ¹²⁹I can also act as a time marker in the sediment core. The high mobility of ¹²⁹I, together with its long half-life, and the fact that it is still being released to the environment offer potential advantages over other anthropogenic time markers. The application of this approach depends on location. In western and northern Europe, ¹²⁹I recorded in sediments is dominated by NFRP releases. The signals of the NWTs and nuclear accidents (Englund et al., 2008) are overlapped by the reprocessing signals. Minor or absent ¹²⁹I signals of nuclear bomb signal have been observed in Ireland and Sweden (Englund et al., 2007; Gallagher et al., 2005). In Ireland, even the large releases of radioactive iodine during the Windscale accident in 1957 have been obscured by the ¹²⁹I releases from NFRPs (Gallagher et al., 2005). While in the U.S. and Spain, the ¹²⁹I imprint of nuclear weapon tests can be resolved in sediments (Oktay et al., 2000; Santos et al., 2007).

5. Conclusion

In this work, we analyzed ^{129}I in a sediment core from Jiaozhou Bay, east coast of China and observed $^{129}\text{I}/^{127}\text{I}$ atomic ratios of $(1.21-189)\times 10^{-12}$, representing both pre-nuclear levels and anthropogenic input. Nuclear activities in the past a few decades were well recorded in this sediment core, including nuclear weapon tests at the Pacific Proving Grounds, maximum global fallout of nuclear weapons testing in 1963, the climax in early 1970s due to the Chinese nuclear weapon tests, and the Chernobyl accident in 1986. Taken together, these illustrate the dating potential of anthropogenic ^{129}I as a time marker in sediments. Two European NFRPs have a significant influence on the Chinese sediments after 1990, and re-emission of ^{129}I from European seas, which has been highly contaminated by liquid discharges from the NFRPs, is a likely major contributor to modern day sediment ^{129}I .

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Table 1. Information and Results of ^{129}I and ^{127}I concentrations as well as $^{129}\text{I}/^{127}\text{I}$ ratios in the sediments in Jiaozhou Bay.

Sample code	Depth* cm	Sedimentation date [#]	^{129}I concentration $\times 10^5$ atoms/g	^{127}I concentration ppm	$^{129}\text{I}/^{127}\text{I}$ ratio $\times 10^{-12}$
JZ3	2.8	2000	145 $\pm$ 7	17.3 $\pm$ 0.5	177 $\pm$ 9
JZ4	3.9	2000	181 $\pm$ 7	20.2 $\pm$ 0.4	189 $\pm$ 7
JZ5	5.0	1999	157 $\pm$ 6	30.9 $\pm$ 0.2	107 $\pm$ 4
JZ8	8.4	1997	119 $\pm$ 6	23.7 $\pm$ 0.5	106 $\pm$ 5
JZ9	9.5	1996	199 $\pm$ 8	24.4 $\pm$ 0.0	172 $\pm$ 7
JZ15	15.3	1993	151 $\pm$ 6	31.3 $\pm$ 0.5	102 $\pm$ 4
JZ17	17.3	1991	106 $\pm$ 4	23.5 $\pm$ 0.1	95.6 $\pm$ 3.8
JZ19	19.5	1990	27.9 $\pm$ 4.1	33.3 $\pm$ 2.8	17.7 $\pm$ 2.6
JZ20	20.6	1989	21.7 $\pm$ 2.2	26.0 $\pm$ 0.3	17.6 $\pm$ 1.8
JZ23	24.0	1987	98.0 $\pm$ 8.9	70.7 $\pm$ 4.4	29.2 $\pm$ 2.7
JZ27	27.3	1985	199.3 $\pm$ 9.3	23.3 $\pm$ 0.3	181 $\pm$ 8
JZ29	30.6	1983	47.1 $\pm$ 5.2	22.9 $\pm$ 0.8	43.4 $\pm$ 5.8
JZ30	31.8	1983	121 $\pm$ 4	43.9 $\pm$ 4.0	58.0 $\pm$ 6.7
JZ33	35.1	1981	19.1 $\pm$ 1.7	26.9 $\pm$ 2.1	15.0 $\pm$ 1.3
JZ37	39.5	1978	23.7 $\pm$ 2.1	36.7 $\pm$ 2.9	13.6 $\pm$ 1.2
JZ39	43.0	1976	129 $\pm$ 1	33.3 $\pm$ 2.6	82.0 $\pm$ 7.1
JZ40	44.9	1975	97.3 $\pm$ 5.3	25.6 $\pm$ 1.0	80.1 $\pm$ 4.4
JZ42	48.9	1972	157.5 $\pm$ 0.5	31.9 $\pm$ 1.9	104 $\pm$ 7
JZ45	54.8	1969	27.3 $\pm$ 1.6	22.0 $\pm$ 0.7	26.2 $\pm$ 1.6
JZ46	56.8	1967	81.1 $\pm$ 3.1	25.0 $\pm$ 0.4	68.3 $\pm$ 2.6
JZ49	62.7	1964	89.5 $\pm$ 7.7	24.3 $\pm$ 1.7	77.8 $\pm$ 6.7
JZ50	64.7	1963	32.2 $\pm$ 1.5	20.2 $\pm$ 0.2	33.7 $\pm$ 1.5
JZ51	66.7	1961	12.9 $\pm$ 0.8	23.4 $\pm$ 1.0	11.7 $\pm$ 0.8
JZ52	68.6	1960	6.9 $\pm$ 0.4	28.6 $\pm$ 0.5	5.1 $\pm$ 0.3
JZ55	74.6	1957	13.7 $\pm$ 0.8	20.8 $\pm$ 0.4	13.9 $\pm$ 0.8
JZ56	76.5	1955	3.3 $\pm$ 0.3	21.5 $\pm$ 0.5	3.3 $\pm$ 0.3
JZ57	78.5	1954	1.8 $\pm$ 0.1	15.4 $\pm$ 0.2	2.5 $\pm$ 0.2
JZ67	98.3	1942	6.3 $\pm$ 0.7	29.6 $\pm$ 2.4	4.5 $\pm$ 0.5
JZ74	108.1	1936	1.3 $\pm$ 0.1	22.7 $\pm$ 0.1	1.2 $\pm$ 0.1
JZ76	116.0	1931	2.8 $\pm$ 0.3	20.5 $\pm$ 0.7	2.9 $\pm$ 0.3

*Refer to the depth of the middle of each slice to the surface of the sediment core.

684 Table 2. Proportion of different grain size and loss of ignition in the sediment core.

Depth cm	<0.063 mm %	0.063-0.125 mm %	>0.125 mm %	Loss on ignition %
0.56	82.20	13.95	3.85	3.04
12.26	87.08	10.80	2.12	2.50
21.73	90.23	7.88	1.89	2.19
32.87	91.41	6.66	1.93	2.14
40.98	92.79	5.89	1.31	3.68
50.85	93.67	4.26	2.07	3.81
60.73	98.34	1.24	0.43	4.17
70.60	97.25	1.96	0.79	3.56
80.48	96.80	2.39	0.81	3.61
90.35	96.95	2.32	0.73	3.24
100.20	98.93	0.79	0.28	3.96
110.10	98.36	1.27	0.37	3.94

685 *The loss on ignition was obtained by combustion of sediment samples under a
686 maxima temperature of 450°C. All these data adopted from Yi, 2006.

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696 **Figures captions**

697 Fig. 1. Sampling site, possible sources of ^{129}I and relevant geographical settings.

698

699 Fig. 2. Comparison of the measured ^{127}I (a) and ^{129}I (b) concentrations in the
700 sediments from Jiaozhou Bay with the reported values in the modern sediment from
701 other locations. (Data from Aldahan et al. (2007), Englund et al. (2008), Moran et al.
702 (1998), Oktay et al. (2000) and Santos et al. (2007)). In plot b, each bar corresponds
703 to the range of the observed ^{129}I concentrations in the related sediment cores.

704

705 Fig. 3. Depth profiles of ^{129}I (a), ^{127}I (b) concentration and $^{129}\text{I}/^{127}\text{I}$ atomic ratio (c) in
706 the sediment core collected from Jiaozhou Bay in 2002. The bars of analytical
707 uncertainty ($k=1$) are shown.

708

709 Fig. 4. Comparison of the measured $^{129}\text{I}/^{127}\text{I}$ ratios in the sediment core from
710 Jiaozhou Bay (a) with ^{137}Cs activity concentration in the same sediment core
711 (b) (Liu et al., 2008), estimated ^{129}I releases from the NTWs (c) (Reithmeier
712 et al., 2006), sum of marine discharges of ^{129}I (d) and sum of air releases of
713 ^{129}I (e) from the two European NFRPs in Sellafield (Britain) and La Hague
714 (France) (Hou et al., 2009b), as well as gaseous releases of ^{129}I from the
715 Japanese Tokai-mura reprocessing plant (Toyama et al., 2013).

716

Figure 1

Figure 2

Figure 3

Figure 4