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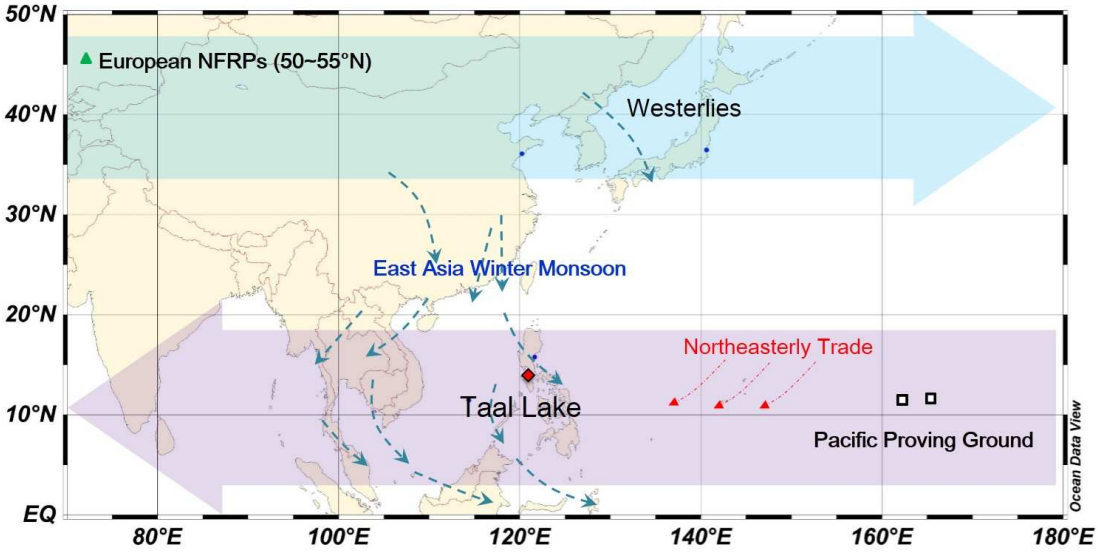

A 60-year record of ^{129}I in Taal Lake sediments (Philippines): influence of human nuclear activities at low latitude region

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Abstract. The influence of human nuclear activities on environmental radioactivity is not well known at low latitude region that are distant from nuclear tests sites and nuclear facilities. A sediment core collected from Taal Lake in the central Philippines was analyzed for ^{129}I and ^{127}I to investigate this influence in a low-latitude terrestrial system. A baseline of $^{129}I/^{127}I$ atomic ratios was established at (2.04 - 5.14) × 10^{-12} in the pre-nuclear era in this region. Controlled by the northeasterly equatorial trade winds, increased $^{129}I/^{127}I$ ratios of (20.1 - 69.3) × 10^{-12} suggest that atmospheric nuclear weapons tests at the Pacific Proving Grounds in the central Pacific Ocean was the major source of ^{129}I in the sediment during 1956 - 1962. The $^{129}I/^{127}I$ ratios, up to 157.5 × 10^{-12} after 1964, indicate a strong influence by European nuclear fuel reprocessing plants. The East Asian Winter Monsoon is found to be the dominant driving force in the atmospheric dispersion of radioactive iodine (^{129}I) from the European nuclear fuel reprocessing plants to Southeast Asia, which is also important for dispersion of other airborne pollutants from the middle-high, to low latitude regions. A significant $^{129}I/^{127}I$ peak at 42.8 cm in the Taal Lake core appears to be the signal of the Chernobyl accident in 1986. In addition, volcanic activities are reflected in the iodine isotope profiles in the sediment core, suggesting the potential of using iodine isotopes as an indicator of volcanic eruptions.

Keywords: Iodine-129 in sediment, Taal Lake, Human nuclear activities, Northeasterly trade winds, East Asian Winter Monsoon, Indicator of volcanic activity

1. Introduction

With the advent of the nuclear age, human nuclear activities (HNAs) mainly including nuclear weapons tests (NWTs), reprocessing of spent nuclear fuel, and the operation of nuclear power and research reactors, have produced a huge amount of radioactive materials. Only a small fraction of these radioactive substances have been released to the environment, but they can be found around the globe. Among these radioactive substances, ^{129}I, owing to its long half-life of 15.7 million years, high fission yield (0.706% per fission of ^{235}U) and volatility, serves as an excellent indicator of HNAs (Buraglio et al., 2001; Fehn and Snyder, 2000; Reithmeier et al., 2010). The global dispersion and distribution of ^{129}I in the environment are also a primary concern for its applications as a tracer for dispersion of gaseous pollutants, water masses movement and geochemical cycles of stable iodine in the atmosphere, hydrosphere and biosphere (Fehn, 2012; Hou et al., 2007; Zhang et al., 2016).

The global inventory of environmental ^{129}I was about 6900 kg by 2009 (Hou et al., 2007). Naturally occurred ^{129}I was estimated to be at about 250 kg, resulting in a natural background $^{129}I/^{127}I$ atomic ratio of (1.5×10^{-12}) (Fehn et al., 2007) in the marine system. Anthropogenic input has increased environmental ^{129}I levels by several orders of magnitude (Hou et al., 2009). Atmospheric NWTs and nuclear accidents have released 50 - 150 kg and 7.2 kg ^{129}I, respectively

(about 6 kg from the Chernobyl accident and 1.2 kg from the Fukushima accident) (Hou et al., 2009; Hou et al., 2013; Hu and Moran, 2010). More than 95% of ^{129}I (about 5900 kg by 2009) in the environment was discharged by two nuclear fuel reprocessing plants (NFRPs) at Sellafield (UK) and La Hague (France) (Hou et al., 2007). In recent years, the marine discharge rate from the two European NFRPs remains at a similar level as in 2009, approximately 200 kg/yr, causing the global ^{129}I inventory of about 8300 kg by 2016. Due to the remarkable discharges of ^{129}I from the European NFRPs, $^{129}I/^{127}I$ ratios of up to 10^{-6} have been reported in the European Seas (Alfimov et al., 2004; Hou et al., 2007; Yiou et al., 2002), six orders of magnitude higher than the natural background for $^{129}I/^{127}I$ ratio (1.5×10^{-12}) (Fehn et al., 2007). In the regions far away from the HNA point sources, $^{129}I/^{127}I$ ratios have remained much lower, in the range of $(1 - 43.5) \times 10^{-10}$ in precipitation, river water, soil and vegetation in the USA and central China (Moran et al., 1999; Oktay et al., 2001; Zhang et al., 2011). Despite the reduced influence of HNAs, $^{129}I/^{127}I$ ratios in Antarctic surface seawater and snow ($(0.06 - 9.5) \times 10^{-10}$) were still more than 4 times higher than the natural background level (1.5×10^{-12}), indicating a predominant anthropogenic source of ^{129}I in the Antarctic environment (Xing et al., 2015).

Sediments are ideal archive for ^{129}I time series records, which can provide useful information to evaluate influences from HNAs, and to understand environmental processes (Aldahan et al., 2007). Some marine and terrestrial sediment cores

have been analyzed to explore the historic ^{129}I records, either from NFRPs or nuclear accidents (Englund et al., 2008; Gallagher et al., 2005; Oktay et al., 2000; Santos et al., 2007). A recent study on ^{129}I in sediment from Jiaozhou Bay, China showed five $^{129}I/^{127}I$ peaks, reflecting atmospheric NWTs at the Pacific Proving Ground (PPG), global fallout of atmospheric NWTs, Chinese atmospheric NWTs, the Chernobyl accident, and NFRPs-derived ^{129}I transported by the westerly wind (Fan et al., 2016). However, the investigations of ^{129}I have mainly focused on middle-high latitude areas of the Northern Hemisphere (Englund et al., 2008; Gallagher et al., 2005; Oktay et al., 2000; Santos et al., 2007). It is well known that hundreds of megatons of nuclear weapons were detonated over the Pacific Ocean between the equator and the Tropic of Cancer during 1948 - 1962 (Andrews et al., 2016). Evaluation of the impact of these nuclear activities and knowledge of the distribution pattern of ^{129}I are lacking at low latitudes (Reithmeier et al., 2010; Snyder et al., 2010). Only a few data of ^{129}I in low latitude region are available in marine system. Time series records of ^{129}I in corals collected in the South China Sea, Con Dao (Vietnam), Guam, and Rabaul (Papua New Guinea) have been reported. These results revealed bomb-produced ^{129}I carried primarily through surface ocean currents to the investigated sites (Bautista et al., 2016; Chang et al., 2016). Currently, ^{129}I measurements in only corals and oceanographic samples in low-latitude region

have been reported, there are no measurement of time series of ^{129}I in terrestrial samples is reported.

This work aims to investigate temporal variations, sources and transportation pathways of ^{129}I in the low latitude terrestrial environment, in order to understand the influence of HNAs in the region. This is implemented by analysis of a sediment core collected in Taal Lake, central Philippines. Since Taal Lake is a volcanic lake, the potential application of iodine isotopes in volcanic eruptions is also explored.

▪ 2. Material and methods

2.1 Geological setting

Taal Lake resides in the Taal Volcano system, located in the southwest portion of Luzon Island, central Philippines (14°00.1'N, 120°59.1'E) (Fig. 1 and Fig. S1). The climate at the study site is monsoonal. The prevailing surface winds vary seasonally. When the western North Pacific subtropical high begins to move northeastward around mid-May, the Asian summer monsoon brings southwesterly winds that propagate over the Philippines. Subsequently, the East Asian winter monsoon is established around November and brings northeasterly surface winds to the Philippines (Villafuerte et al., 2014). The trade winds are the prevailing northeasterly surface winds found in the Philippines, and in the tropics of the Northern Hemisphere.

Taal Lake sediments are primarily sourced from the watersheds that cover about 350 km^2 in the surrounding land area, and are affected by the eroding lake slope and extruded tephra (Ramos, 2002).

Taal Volcano with 33 recorded eruptions is considered one of the most active volcanic centers in the Philippines. The most recent period of activity lasted from 1965 to 1977 with the area of activity concentrated in the vicinity of Mount Tabaro (Global volcanism program; Moore et al., 1966; Philippine Institute of Volcanology and Seismology).

2.2 Sampling and core chronology

A gravity core of 120 cm length was collected at a water depth of 15 m and 20 m away from Taal lake shoreline, central Philippines in November, 2007. The core is mainly dark brown-black clay, containing shells and volcanic ash. The chronology of the sediment core was established by $\Delta^{14}C$ values determined from plant remains, and compared with an atmospheric $\Delta^{14}C$ bomb curve (dating model in the supplementary information, Fig. S2 and S3). Ages fall within the period 1947 - 2004 (Li and Xu, 2008; Hua et al., 2013). The sedimentation rate was calculated to be 2.04 ± 0.01 cm y^{-1}. Based on assumption of a constant sedimentation rate and the sampling resolution of 2 - 3 cm for each subsample, the age uncertainties for entire core might be up to 5 years.

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

147 The sediment samples were sectioned into 2-3 cm intervals, freeze-dried, ground
148 and sieved through a 200 mesh sieve. Combustion followed by solvent
149 extraction was used to separate iodine from the sediments. The analytical
150 method has been reported in detail elsewhere (Hou et al., 2010; Zhang et al.,
151 2011; Zhou et al., 2010). In brief, 2 g of sediment was weighed into a quartz
152 boat. 500 Bq of ^{125}I was spiked to the sample to monitor chemical yield. The
153 sample was combusted at 800 ℃ in a four-tube Pyrolyser® furnace in an
154 atmosphere of O_2 and N_2 to release iodine (Zhou et al., 2010). The released
155 iodine was trapped in a 0.5 M NaOH - 0.02 M $NaHSO_3$ solution. One mL of the
156 trap solution was used reserved for ^{127}I determination, 3 ml of trap solution was
157 transferred to a counting tube and measured for 125I using a NaI gamma
158 detector for monitoring chemical yield of iodine. After measurement, the
159 solution was combined with the remaining trap solution and was used to
160 separate ^{129}I by solvent extraction. The solution was transferred into a separatory
161 funnel and 0.5 mg of ^{127}I carrier (Woodward Inc., USA) was added. $NaHSO_3$
162 and HNO_3 were added to reduce iodine to iodide, and then $NaNO_2$ was used to
163 oxidize iodide to I_2 that was extracted to chloroform ($CHCl_3$) phase. After
164 discarding the water phase, the I_2 in $CHCl_3$ phase was back extracted to water
165 phase by reducing I2 to iodide by $NaHSO_3$solution. This procedure was repeated
166 as an additional purification step. $AgNO_3$ was added to the back-extracted
167 aqueous solution and the iodide was precipitated as AgI. The AgI precipitate

was washed by HNO_3 once, rinsed with deionized water twice, and dried at 60°C in an oven. The dried AgI precipitate was ground to fine powder and mixed with niobium powder (325 mesh, Alfa Aesar) in a mass ratio of 1:3, and pressed into a copper holder for AMS measurement.

The trap solutions were diluted ten-fold with 1% $NH_3 \cdot H_2O$, and measured for stable iodine (^{127}I) using ICP-MS (X Series II ICP-MS, Thermal Electron Corporation). Cs^+ was added to the diluted solution as internal standard to a final concentration of 2 μg/L. The detection limit of ^{127}I was calculated as 3 times SD of blanks to be 0.02 μg/L.

^{129}I was measured by a 3 MV Tandem AMS system (High Voltage Engineering Europa) at the Xi'an AMS center. I^{5+} ion was chosen for the measurements. Stable $^{127}I^{5+}$ currents were measured using a Faraday cup, and ^{129}I ions were measured using a gas ionization detector. All samples were measured for 6 cycles and 5 min per sample in each cycle. The procedural blanks are 1.5×10^{-13} for $^{129}I/^{127}I$ ratios, which are more than 10 times lower than the measured $^{129}I/^{127}I$ ratios in samples. The final results of ^{129}I were calculated by subtracting the blank value.

▪ 3. Results and discussion

3.1 Depth profiles of ^{129}I and ^{127}I in Taal Lake sediment core

Large variations of the concentrations of iodine isotopes were observed in the sediment core (Fig. 2 and Table S1). The concentrations of ^{127}I range from 2.97 μg/g to 54.7 μg/g with a mean value of (11.1 ± 8.9) μg/g. The maximum ^{127}I concentration was observed at the depth of 7.1 cm (corresponding to the layer of 2002). ^{129}I concentrations range from 0.50×10^5 atoms/g to 215.9×10^5 atoms/g, and in most samples, ^{129}I concentrations were lower than 84.7×10^5 atoms/g except for three samples at depths of 4.8 - 9.5 cm. The ^{129}I peak was found at the same subsurface layer as that of ^{127}I (7.1 cm). Relatively lower ^{129}I and ^{127}I concentrations, as compared to adjacent layers, were observed in two depths that date to 1965 - 1970 (73.2 - 77.0 cm) and 1976 - 1977 (57.7 - 59.7 cm). $^{129}I/^{127}I$ ratios gradually increased from 2.04×10^{-12} in 1952 (109.5 cm) to a peak value of 157.4×10^{-12} in 1984 (42.8 cm), then slightly decreased to 69.3×10^{-12} in 1988 followed by a few minor fluctuations.

A significantly positive correlation was observed between ^{129}I and ^{127}I concentrations in the sediment (r= 0.81 for the whole core) (Table S1), while no positive correlation or a weak correlation was reported in other sediment samples (r=0.15 in the Jiaozhou Bay sediment, and r=0.29 in the Kattegat sea sediment) (Fan et al., 2016; López-Gutiérrez et al., 2004). Given the possible influence of high values on correlation, elevated ^{129}I and ^{127}I concentrations during 2000 - 2004 were excluded. No significant correlation could be observed during 1948 - 1998 (r=0.10). However, highly significant correlation was

observed during 1964 - 1998 (r=0.93). Values of ^{129}I and ^{127}I during 1948 - 1954 and 1956 - 1963 fell below the regression line of data during 1964 - 1998 (Fig. S4). The environmental implication of the correlation are discussed below, in the section 3.3.3.

3.2 Level and variation of $^{129}I/^{127}I$ in Taal Lake sediment compared with other sediment cores

^{127}I concentrations in Taal Lake sediment were comparable to those in coastal sediments from the Mississippi River Bight (3.4 - 34.3 μg/g) and Chinese marginal seas (mean values of 14 - 22 μg/g), but much lower than those in marine sediments from the Baltic Sea, Pacific and Arctic Ocean (20 - 139 μg/g) (Aldahan et al., 2007; Fan et al., 2016; Gao et al., 2003; Oktay et al., 2000). The peak values of ^{129}I and ^{127}I concentrations occurred at a depth of 7.1 cm (in 2002). Similar patterns of ^{129}I and ^{127}I concentrations were reported for cores from the Mississippi River and the Baltic Sea, in which ^{129}I peaks occurred at depths of 7 - 8 cm and 3 - 4 cm, respectively (Aldahan et al., 2007; Oktay et al., 2000). Enrichment of iodine in subsurface sediment was attributed to anaerobic degradation of organic matter in this layer, and re-adsorption of the released inorganic iodine to the subsurface layer (Aldahan et al., 2007; Price and Calvert, 1973; Ullman and Aller, 1980). This is also supported by a high water content

(87%) and abundant residual plants in this layer of the Taal Lake sediment core (Table S1) (Li and Xu, 2008).

$^{129}I/^{127}I$ ratios are distinctly different from ^{129}I and ^{127}I concentrations, and no subsurface maximum was observed (Fig. 2). Long time series ^{129}I records in sediment cores have also been reported in other locations (Fan et al., 2016; López-Gutiérrez et al., 2004; Oktay et al., 2000; Santos et al., 2007); and it is generally agreed that $^{129}I/^{127}I$ ratios are more useful than ^{129}I concentrations, when comparing ^{129}I levels in different sites and sample media. This is because iodine concentrations in sediment cores vary significantly, and are generally affected by environmental factors and sediment sources. Figure S5 compares the $^{129}I/^{127}I$ ratios in sediment cores from marine and terrestrial systems. As expected, $^{129}I/^{127}I$ ratios in the Taal lake sediment fell well within those areas far away from HNAs, e.g., Jiaozhou Bay in China and the Mississippi River in USA (Fan et al., 2016; Oktay et al., 2000). The $^{129}I/^{127}I$ ratios from Sweden, Ireland and Spain (23.4×10^{-12} to 7.69×10^{-7}) are about 1 - 3 orders of magnitude higher than those from the low latitude lake and river sediments, doubtless due to the direct influence of European NFRPs (Aldahan et al., 2007; Englund et al., 2008; Gallagher et al., 2005; Santos et al., 2007). In contrast, marine sediments collected from the Baltic Sea (1.07×10^{-8} to 7.50×10^{-7}) and Kattegat Sea (3.69×10^{-10} to 2.77×10^{-8}) showed a similar range of $^{129}I/^{127}I$ ratios as lake sediments from Central Sweden, both 3 - 5 orders of magnitude higher than that in Taal

Lake. This is a quantitative indication that $^{129}I/^{127}I$ ratios in lake sediments are mainly dependent on the proximity of HNAs and ^{129}I transport pathways.

3.3 Historic ^{129}I records in low latitude terrestrial environment

Characterized by a peak in the early 1960s followed by continuous decline, a typical Northern Hemisphere bomb-produced $\Delta^{14}C$ signal was found in residual plants extracted from Taal lake sediments (Fig. 3a, Li and Xu, 2008). Unlike $\Delta^{14}C$, the variation of $^{129}I/^{127}I$ ratios in the sediment core is quite complicated, due to variable ^{129}I sources through time, including nature occurred, NWTs, NFRPs and nuclear accidents releases over past decades. In order to clarify the sources of ^{129}I and HNAs influence on low latitude regions, the 60-year ^{129}I record in the Taal lake core was divided into three periods, including the period with consistently low $^{129}I/^{127}I$ ratios during 1948 - 1954, the first rise of $^{129}I/^{127}I$ ratios during 1956 - 1980, and the second rise leading up to 1984 and followed by fluctuating $^{129}I/^{127}I$ ratios (Fig. 3a).

3.3.1 A baseline of pre-nuclear $^{129}I/^{127}I$ ratios

This period (1948 - 1954) features a platform of low $^{129}I/^{127}I$ ratios with an average of $(3.66 \pm 1.14) \times 10^{-12}$, measured in the bottom layers (105 - 120 cm) of the sediment core. The results are close to the natural marine background level of $^{129}I/^{127}I$ ratio ($(1.50 \pm 0.15) \times 10^{-12}$) (Fehn et al., 2007). This indicates that ^{129}I in this period mainly originated from natural processes and that very limited

vertical migration of iodine occurred in the sediment core. This is also supported by the pre-nuclear $\Delta^{14}C$ values from -26.8‰ to -40.3‰, at the same depths (Fig. 3a).

The environmental background level cited above was derived by analyzing marine sediments from South Carolina and along the Western continental margin of the Americas (Fehn et al., 2007). As noted, $^{129}I/^{127}I$ ratios in 105-120 cm depth in the Taal Lake sediment core ($(2\text{-}7\text{-}4.5)\times10^{-12}$) are slightly higher than the documented pre-nuclear $^{129}I/^{127}I$ value (1.50×10^{-12}) in the marine system. Tt is well known that stable ^{127}I concentrations in terrestrial surface waters (typically a few μg/L) are generally an order of magnitude lower than those of seawater (40 - 60 μg/L). If production rates of ^{129}I in the air are the same, this implies that the pre-nuclear $^{129}I/^{127}I$ ratios in Taal Lake should be an order of magnitude higher than 1.50×10^{-12}. Furthermore, addition of ^{129}I to the terrestrial sediment by spontaneous fission of ^{238}U in the crust could likewise raise the pre-nuclear ratio. That possibility is easily dismissed because the produced ^{129}I is only 0.22×10^{5} atoms/g if assuming a maximum uranium concentration of 15 μg/g for calculation (Fabryka-Martin, 1988), which accounts for less than 15% of pre-anthropogenic ^{129}I level in the Taal sediment. As shown in the Tinto river sediment (Spain), the naturally produced $^{129}I/^{127}I$ ratio was 2×10^{-11}, which is one order of magnitude higher than the pre-nuclear $^{129}I/^{127}I$ ratio (Santos et al., 2007). However, even though Taal Lake is terrestrial, it was reported that Taal Lake was

connected with the sea by a navigational channel, while volcanic eruption in 1749 blocked the only channel and turned Taal Lake from marine environment to a brackish water reservoir (Ramos, 2002). This resulted in relatively high concentrations of ^{127}I in the lake sediment (2.97 - 54.7 μg/g), which were about one order of magnitude higher than those in Chinese stream sediments (0.36 - 2.4 μg/g) (Cheng et al., 2011). Therefore, the pre-nuclear Taal Lake $^{129}I/^{127}I$ ratios that are identical with those in marine sediment might be attributed to relatively high ^{127}I concentrations due to the evolution of Taal Lake. In addition, it should be mentioned that the nuclear weapons tests in the PPG started from June 1946 until 1962. The fall out of the atmospheric tests in this site in 1946-1954 should be another reason causing slightly higher $^{129}I/^{127}I$ ratios in this period.

3.3.2 Atmospheric NWT signal from the PPG through the northeasterly trade winds

The $^{129}I/^{127}I$ ratios in the Taal Lake sediments increased by 10 - 50 times over the baseline values since 1956. The main feature of this period is a minima value in 1963 (34.3×10^{-12}) separating two peaks; the first one shows rapidly increased $^{129}I/^{127}I$ from 20.2×10^{-12} in 1956 to 69.3×10^{-12} in the early 1960s, and the second is a quasi-constant $^{129}I/^{127}I$ peak of $(79.3 - 109) \times 10^{-12}$ during 1964 - 1980.

The significantly increased $^{129}I/^{127}I$ ratios in the Taal Lake sediment core since the mid-1950s indicate that the site received significant input of ^{129}I produced by atmospheric NWTs. At similar latitudes in the Northern Hemisphere (11°N), 104 aboveground NWTs were carried out in the PPG in the Marshall Islands area during 1945 - 1962, with a total yield of 152 Mt, which accounts for 34.6% of the total yield of worldwide atmospheric NWTs (440 Mt) (UNSCEAR, 2000). Of these, 67 nuclear tests from July 1946 to August 1958 were conducted in the Enewetak and Bikini Atolls (Department of Energy Nevada Operations Office, 2000), 4500 km and 4800 km east of Taal lake, respectively. There were no NWTs at the PPG during 1959 - 1961. Due to a global atmospheric fallout lag to the earth's surface of about 1 - 2 years (Hua et al., 2013) and dating uncertainty of the core, the increased $^{129}I/^{127}I$ ratios in Taal Lake sediments during 1956 - 1962 correspond to atmospheric NWTs before 1960. The apparent drop of $^{129}I/^{127}I$ ratio in the sediment core in 1963 (Fig. 3a) might correspond to the no-tests period of 1959 - 1961. More atmospheric NWTs were conducted in north of 30°N by the USA (Nevada, 37°N), and Soviet Union (Semipalatinsk, 50°N and Novaya Zemlya, 73 - 74°N). The total explosion yield at the PPG was about four times higher than those north of 30°N during 1945 - 1958, but five times lower than the latter area during 1961 - 1962 (Fig. S6). Therefore, atmospheric NWT at the PPG predominantly affected the study area before 1958.

329 The $^{129}I/^{127}I$ signals of atmospheric NWTs were also reported in the Jiaozhou
330 Bay sediment and Parola coral samples (Fig. 3b) (Bautista et al., 2016; Fan et al.,
331 2016). During 1956 - 1962, the peak value of the $^{129}I/^{127}I$ ratio in the Taal Lake
332 sediment is approximately five-fold higher than those reported in the Jiaozhou
333 Bay sediment and coral samples from Parola and Baler (Fig. 3b). The relatively
334 lower $^{129}I/^{127}I$ ratios in the marine system might be attributed to high ^{127}I
335 concentrations in the ocean. It has been reported that close-in fallout around the
336 PPG could be carried by the North Pacific Equatorial current to the east coast of
337 Philippines and South China Sea as seen in coral ^{129}I records from Con Dao and
338 Xisha Islands (Chang et al., 2016). Although the possible seawater intrusion into
339 the Taal Main Crater Lake (in Taal Volcano) (Delmelle et al., 1998) might carry
340 PPG-derived ^{129}I into Taal Lake, the relatively low $^{129}I/^{127}I$ ratios in the Baler
341 corals (equilibrium with $^{129}I/^{127}I$ in seawater) (Bautista et al., 2016) imply that
342 seawater intrusion was not the major source of ^{129}I in the Taal Lake sediment.

343 The high $^{129}I/^{127}I$ ratios in Taal Lake sediments were thus attributed to deposition
344 of airborne ^{129}I of atmospheric NWTs. Meteorological observations have shown
345 often brisk northeast trade winds at the PPG in the Marshall Islands. Radioactive
346 substances dispersed to the west as they fell through the trade winds (Kunkle
347 and Ristvet, 2013). Furthermore, the HASL aerial surveys have suggested that
348 radioactive contamination from the PPG was found in the islands of Yap
349 (9°32′N, 138°05′E) and Iwo Jima (24°47′N, 141°20′E) in the west Pacific

(Kunkle and Ristvet, 2013). This suggests that northeasterly trade winds play a key role in transporting ^{129}I from the PPG to Taal Lake, Philippines. Although fallout of the atmospheric NWTs conducted north of 30°N also dispersed and deposited iodine at low latitudes, the $^{129}I/^{127}I$ ratio in 1964 layer of the Taal Lake sediment was only 1.5 times higher than that in 1960, indicating that they were minor contributors due to longer transportation distances.

With the signing of the Partial Test Ban Treaty in 1963, no further atmospheric NWTs were conducted at the PPG. Since then, all the atmospheric NWTs were conducted north of 35°N and the South Pacific region, including 26 tests in Lop Nor, China during 1964 - 1980, and 46 tests in French Polynesia during 1966 - 1974 (Fig. S6) (Norris, 1996; Ribbe and Tomczak, 2006). Unlike the Jiaozhou Bay sediment core with two peaks during 1963 - 1980, rather constant $^{129}I/^{127}I$ ratios of (79 - 109) × 10^{-12} were observed in the Taal Lake sediment core in 1964-1980, which are less than 1.4-fold higher than the peak in 1960. The contribution of Chinese tests cannot be completely ruled out, but it is apparent that these tests were not a major contributor to the low latitude area. This is because 1) no peaks can be identified during 1963 - 1980, and 2) the total yield of Chinese tests was very low compared to that conducted at the PPG. The contribution from the French tests can be easily ruled out because all the tests were conducted to the south of 21°S, where ^{129}I is hardly carried to the north hemisphere. The results are indicative of a uniform input of ^{129}I from the

middle-high latitude regions. European NFRPs started to release gaseous ^{129}I to the atmosphere from the early 1950s, and reached about 100 GBq/y in the 1960s (Fig. 3c) (Bautista et al., 2016; Reithmeier et al., 2010), which is about 10 - 100 times higher than the total ^{129}I released from all atmospheric NWTs after 1963 (Fig. 3c). Thus, the addition of airborne ^{129}I originated from the NFRPs became the major source of ^{129}I during 1964 - 1980, indicating the influence of NFRPs on low latitude region.

3.3.3 Nuclear facilities-released ^{129}I after 1980: influence of the East Asian Winter Monsoon

It is notable that $^{129}I/^{127}I$ ratios in the Taal Lake sediment rapidly increased after 1980, reaching a maximum value (157.5×10^{-12}) for the whole core in 1984, and followed by four small peaks in ~1989, ~1995, ~1997 and ~2004 (Fig. 3a). No atmospheric NWTs has occurred after 1980, whereas the total airborne ^{129}I released from the NFRPs in Europe, Former Soviet and United States greatly increased to a maximum value of 200 GBq/y in 1984 (Bautista et al., 2016; Reithmeier et al., 2010). The rapid increase in $^{129}I/^{127}I$ ratios in Taal Lake during 1980 - 1984 could be attributed to gaseous releases of ^{129}I from the European NFRPs (Fig. 3c). Since 1996, atmospheric releases of ^{129}I from the two major NFRPs at Sellafield and La Hague dramatically decreased to about 30 GBq/y (Bautista et al., 2016), while liquid discharge of ^{129}I from the two NFRPs into the

391 sea significantly increased from 20 GBq/y in the 1950s to around 2200 GBq/y
392 (350 kg/y) in the 2000s (Hou et al., 2007). Studies on ^{129}I in aerosols and
393 rainwater have shown that re-emission of ^{129}I from contaminated seawater has
394 become a key contributor to ^{129}I in the atmosphere (Englund et al., 2010;
395 Reithmeier et al., 2006; Zhang et al., 2016). It can be estimated that the amount of
396 re-emitted ^{129}I from the sea to the atmosphere has increased to 90 GBq/y (13.8
397 kg/y) in the 2000s (Fig. 3c), assuming a 0.3% annual re-emission rate of the total
398 ^{129}I inventory in the upper layer of the ocean (Reithmeier et al., 2006). This
399 implies that re-emission of liquid discharged ^{129}I from the NFRPs become the
400 major ^{129}I source to Taal Lake sediments after 1996, instead of direct gaseous
401 releases.

402 The ^{129}I pulse from the Chernobyl nuclear accident occurred in 1986 had been
403 widely observed in sediment cores and in corals (Bautista et al., 2016; Fan et al.,
404 2016; Hou et al., 2003). However, the maximum $^{129}I/^{127}I$ ratio in the sediment
405 core in this study was dated to 1984, this might be attributed to the uncertainty of
406 the dating method and the contribution of other sources. The dating uncertainty
407 due to the application of a uniform sedimentary rate of 2.04 cm/y for the core
408 chronology can explain this discrepancy. In particular, we expect a fast
409 sedimentary rate during rapid deposition of volcanic ash and tephra during
410 volcanic eruptions (Moore et al., 1966). Hence, the peak $^{129}I/^{127}I$ ratios that we

observe around 1984 likely reflect an integrated signal of airborne NFRPs-derived ^{129}I with the Chernobyl signal.

After the 1984 peak value, $^{129}I/^{127}I$ ratios in the sediment core declined significantly. Three small $^{129}I/^{127}I$ peaks (Fig. 3a), in ~1989, ~1995, ~1997 were observed, which are correspond well to the releases of ^{129}I from the two European NFRPs in 1989, 1994 and 1996 (Fig. 3c). These peaks were not observed in the coral sample from Parola, Philippines, likely because the coral signals have been smoothed by dilution of high level ^{127}I in seawater. The difference in ^{129}I records between the Taal Lake sediment and Parola coral suggests that terrestrial sediments may provide more sensitive ^{129}I information than marine archives. As discussed above, air transport is the primary pathway that carries iodine to low latitude region. However, in spite of higher seawater ^{127}I concentrations and subsequent dilution, the $^{129}I/^{127}I$ values in Jiaozhou Bay sediments are still higher than those of Taal Lake after 1990 (Fig. 3b). This difference is attributed to the prevailing winds that affect each site. Jiaozhou Bay is located in mid-latitude region (39°N), and exactly in the pathway of the westerlies, which is known to carry re-emitted ^{129}I from European seas (Fan et al., 2016) to east direction. The Tall Lake (14°N) is not directly affected by the westerlies which extend only to about 30°N. Whereas the westerlies with an enriched ^{129}I air mass pass over the west Pacific are relatively depleted in ^{129}I. Toyama et al. (2013) has showed relatively low $^{129}I/^{127}I$ ratios in atmospheric samples (precipitation

containing airborne particulate dust) from Ishigaki Island (24°20′N, 124°9′E), approximately 1200 km northeast of Taal Lake. Due to the monsoonal climate in the Philippines, the sampling site is variably affected by ^{129}I-enriched East Asian winter monsoon and ^{129}I-poor air during East Asian summer monsoon (An et al., 2015; Loo et al., 2015; Villafuerte et al., 2014) (Fig. 1). There is evidence that eolian dust input related to the East Asian winter monsoon is the main provenance of sediments in the Philippine Sea (Xu et al., 2013). Therefore, the influence of European NFRPs on Taal Lake, Philippines is modulated by the summer and winter East Asian monsoon. Although the East Asian winter monsoon brings ^{129}I from the mid-high latitude regions into Taal Lake, the direct contribution of European NFRPs at this low latitude is lower than in the mid-latitude regions of the North Hemisphere.

In the surface layer of the Taal Lake sediment core, the fourth ^{129}I/^{127}I peak in 2004 (Fig. 3a) might be related to an enhanced East Asian winter monsoon with a high intensity index of 0.40 (relative to weak East Asian winter monsoon in 2000 - 2003 with a low average intensity index of -0.39) (He and Wang, 2012) that carried more re-emitted ^{129}I from European seas (the 4′ peak in Fig. 3c) to the low latitude region. More study is needed to further confirm this assumption.

It is worth noting the significant positive correlation between ^{129}I and ^{127}I during 1964 - 1998 (the period with increased ^{129}I signal mainly from the European

NFRPs) (Fig. S4), indicating that both ^{129}I and ^{127}I in Taal Lake sediments were mainly transported by the East Asian winter monsoon (Xu et al., 2013). While no correlation between ^{129}I and ^{127}I during 1956 - 1963 reflected that the two isotopes were from different air masses, ^{129}I controlled by the northeasterly trade and ^{127}I by the East Asian winter monsoon in that period.

Other nuclear activities might also contribute ^{129}I to Taal Lake. As of 2004, 68 nuclear power reactors were in operation in Japan, South Korea and China adjacent to the Philippines, with a net capacity of 60,519 MWe (Table S2) (World Nuclear Association, 2017). ^{129}I generated by nuclear power production was stored in the spent fuel. Our previous determinations of $^{129}I/^{127}I$ ratios in surface seawater in the vicinity of Chinese NPPs did not show a significant $^{129}I/^{127}I$ gradient in seawater samples collected from locations with a distance to the outlet of 2 - 7 km, and all at background level in this region (He et al., 2011; Zhang et al., 2012), indicating no measurable contribution to the environmental ^{129}I from these NPPs. A NFRP located in Tokai, Japan has released about 1.0 kg ^{129}I since its operation from 1977 until 2005, which results in a maximum ^{129}I release of 0.15 kg/y in 1985 (JAEA, 2006; Shinohara, 2004), less than 10% of ^{129}I re-emitted from NFRPs-influenced oceans (1.5 - 13.8 kg/y) during 1980 - 2004. $^{129}I/^{127}I$ ratios of (40 - 80) $\times 10^{-12}$ in surface seawater collected from Japan Basin, Yamato Basin and offshore of Kushiro in 2007 were reported (Suzuki et al., 2010), showing an insignificant contribution of the Japanese nuclear

reprocessing plant on ^{129}I in the Taal Lake. Therefore, the contribution of ^{129}I from the adjacent NPPs and the Tokai NFRP is considered negligible in the Taal Lake sediment record.

3.4 Relation of volcanic activities with iodine isotopes

Relatively lower concentrations of ^{127}I (about 5 μg/g) and ^{129}I (< 29.4×10^5 atoms/g) were observed in the sediment samples during 1965 - 1977. These decreases in ^{127}I and ^{129}I concentrations well correspond to the Taal volcanic eruptions (Fig. 2). Microscope observation of the sediment showed that these layers contained high amount of volcanic glass, but less water (55%), organic matters and carbonates (Li and Xu, 2008) (Table S1), confirming the contribution of the volcanic sources. Due to mixing of iodine-free volcanic materials (iodine has evaporated in high temperature during volcanic eruption), iodine concentrations can be potentially used to record historic volcanic activities.

▪ 4. Conclusions and perspectives

Distribution of ^{129}I and ^{127}I in a sediment core collected in Taal Lake, Philippines shows three distinct ^{129}I depositional periods: (1) pre-nuclear ^{129}I, providing a baseline to evaluate the influence of HNAs; (2) atmospheric NWTs-originated ^{129}I, showing a strong influence from the atmospheric NWTs at the PPG through the northeasterly trade; (3) NFRPs-derived ^{129}I, featuring by

high resolution ^{129}I records of direct gaseous releases and secondary emission of ^{129}I from oceans contaminated by liquid discharge of NFRPs. It was also found that East Asian winter monsoon plays a significant role in the transport of ^{129}I and influence of European NFRPs at the study site. It could also transport other volatile gaseous pollutants from mid- to low-latitude areas. In addition, this work suggests that iodine isotopes have potential to trace volcanic activities.

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Additional Information

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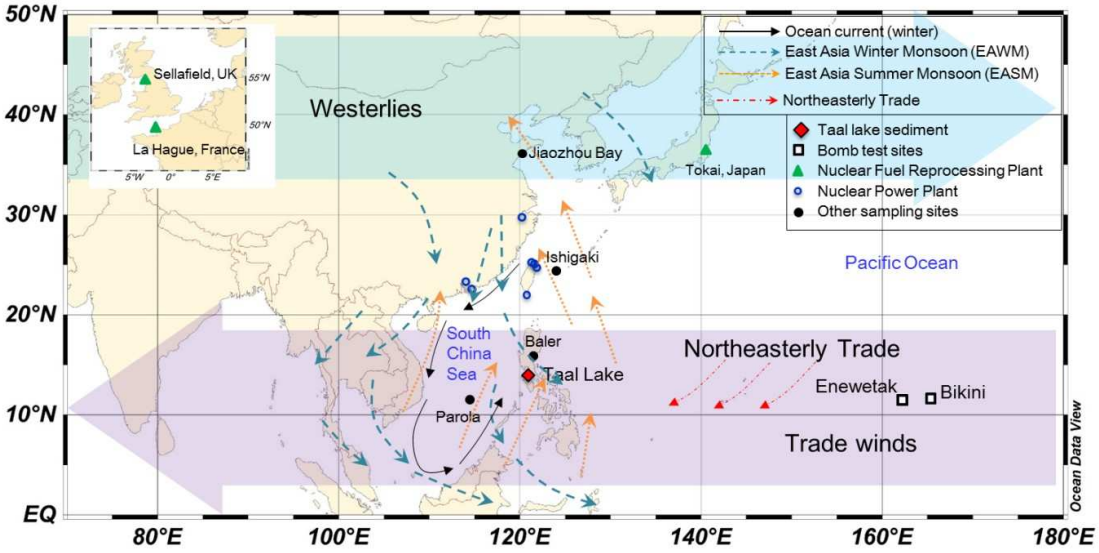
Caption of figures

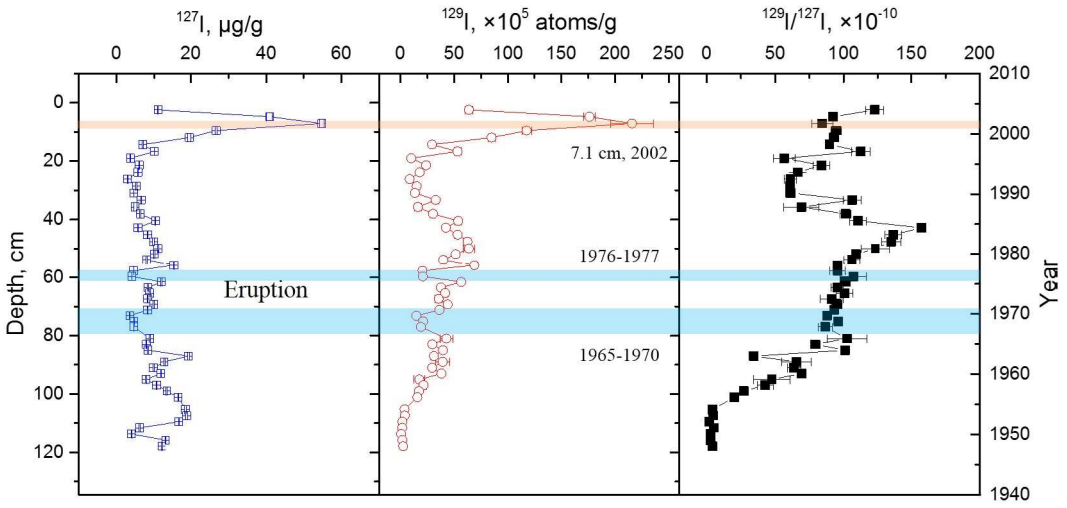
Fig. 1 Locations of Taal Lake in the central Philippines (red diamond), nuclear power plants (blue circles), nuclear fuel reprocessing plants in United Kingdom, France and Japan (green triangles), major bomb testing sites in the Pacific Ocean (hollow squares) and other sampling sites for corals from Parala and Baler, and precipitation from Ishigaka (black dots). The insert shows the locations of two nuclear fuel reprocessing plants, Sellafield and La Hague in Europe. The prevailing winds are westerly (light blue arrow) at mid-high latitudes and equator trade winds (light purple arrow). Regional winds are the East Asia Monsoon in winter (aqua dash-line arrows), summer (orange dot-line arrows), and northeasterly trade winds (red dash dot arrows). Ocean currents in the South China Sea during winter are denoted by black line arrows.

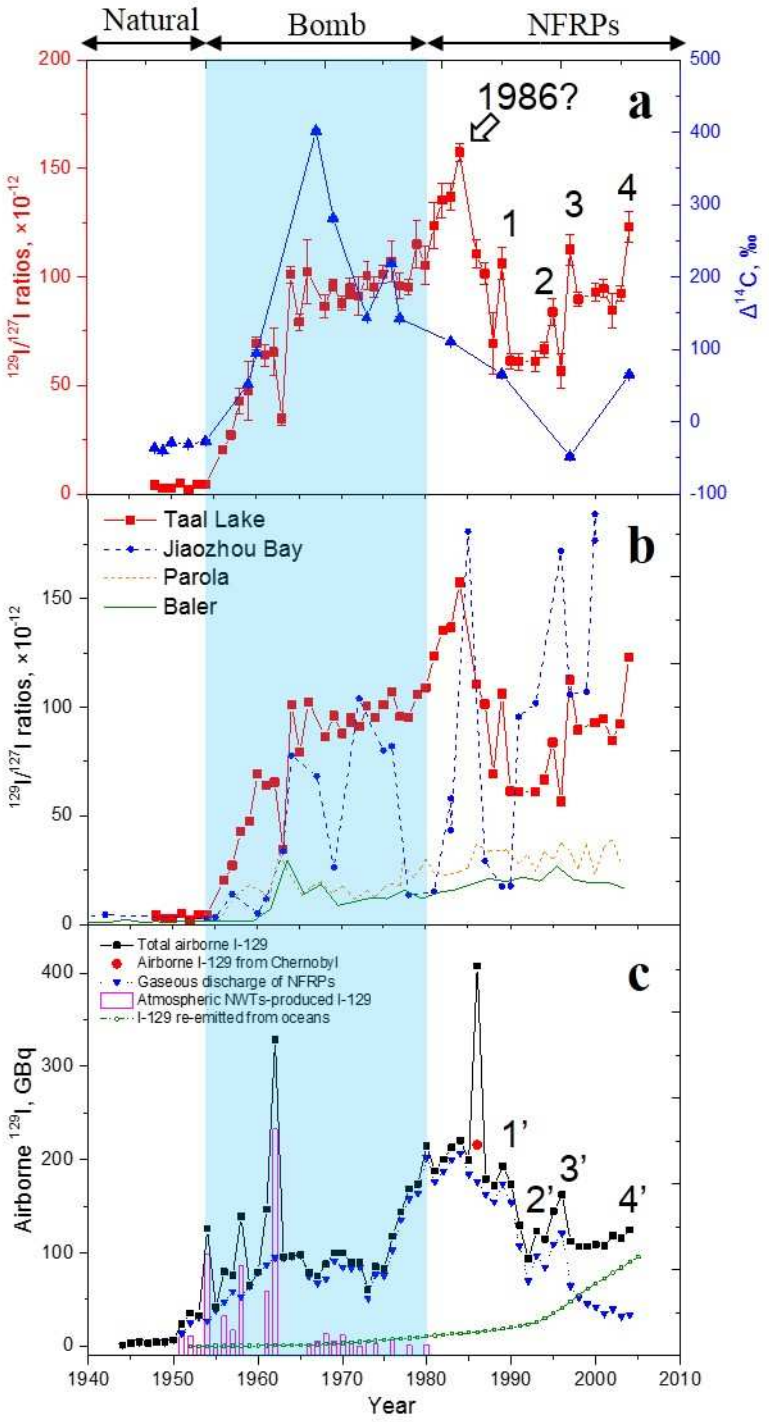
Fig. 2 Concentrations of ^{127}I and ^{129}I and the ratios of $^{129}I/^{127}I$ in the Taal Lake sediment core. The peaks of ^{127}I and ^{129}I concentrations occur at the depth of 7.1 cm (orange band). Events of volcanic eruptions during 1965-1977 are shown as blue bands.

Fig. 3 (a) $^{129}I/^{127}I$ ratios and $\Delta^{14}C$ in the sediment core from Taal Lake, Philippines and (b) comparison to $^{129}I/^{127}I$ ratios of Jiaozhou Bay, China (Fan et al., 2016), and coral from Parola, Philippines from the South China Sea side and Baler, the Philippines at the west Pacific Ocean side

(Bautista et al., 2016). (c) ^{129}I releases (GBq) from atmospheric NWT, NFRPs, the Chernobyl accident and re-emission from contaminated oceans (compiled from Fan et al., 2016 and Bautista et al., 2016). The highest peak of $^{129}I/^{127}I$ ratios around 1984 appears to be the signal of the Chernobyl accident in 1986. The lesser peaks (1, 2 and 3) correspond well the releases of ^{129}I from the two European NFRPs in 1989, 1994 and 1996, respectively. The fourth peak might be related to the stronger East Asia wither monsoon in 2004.



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Parola
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$^{129}I/^{127}I$ ratios, ×10^{-12}
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Gaseous discharge of NFRPs
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▪ Highlights

The highlights of this paper involve:

i. A 60-year ^{129}I profile in Taal Lake sediment is reported.

ii. Human nuclear activities at low latitudes are well recorded by ^{129}I profile;

iii. Northeasterly trade and East Asian winter monsoon are important driving forces;

iv. Iodine isotopes are potentially applied as a tracer for volcanic eruptions;