

Iodine isotopes (¹²⁹I and ¹²⁷I) in the hydrosphere of Qinghai-Tibet region and South China Sea

Yi, Peng; Chen, Xuegao; Wang, Zixia; Aldahan, Ala; Hou, Xiaolin; Yu, Zhongbo

Published in: Journal of Environmental Radioactivity

Link to article, DOI: 10.1016/j.jenvrad.2018.06.005

Publication date: 2018

Document Version Peer reviewed version

Link back to DTU Orbit

Citation (APA): Yi, P., Chen, X., Wang, Z., Aldahan, A., Hou, X., & Yu, Z. (2018). Iodine isotopes (¹²⁹I and ¹²⁷I) in the hydrosphere of Qinghai-Tibet region and South China Sea. *Journal of Environmental Radioactivity, 192*, 86-94. https://doi.org/10.1016/j.jenvrad.2018.06.005

General rights

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.

- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Iodine isotopes (¹²⁹I and ¹²⁷I) in the hydrosphere of Qinghai-Tibet region and South China Sea

1 Abstract

The radioactive isotope 129 I, with a half-life of 1.57×10^7 years, is widely used as a tracer to 2 3 assess nuclear safety, to track environmental and geological events and to figure out the details of the stable iodine geochemical cycle. This work investigated the ¹²⁹I and ¹²⁷I 4 distribution in water samples collected from the terrestrial (rivers, lakes and springs) and 5 marine water systems (estuary and sea) in China. The measured ¹²⁹I concentrations of (1-6 51) $\times 10^6$ atoms/L and 129 L/ 127 L ratios of (0.03-21) $\times 10^{-10}$ shows the variability of 129 L level 7 in the water systems. The local permafrost and seasonal frozen environment play a key role 8 9 in groundwater recharge in the Qinghai-Tibet region, which is reflected in the ¹²⁹I distribution in surface water. The depth distribution of ¹²⁹I in the water column of the South 10 China Sea reflects the effluence of different currents. The results also indicate that the 11 hydrosphere of China contains one to three orders of magnitude lower ¹²⁹I concentrations 12 compared to those reported in Europe. Despite the large distance, the European nuclear 13 fuel reprocessing facilities represent the major source of ¹²⁹I in the hydrosphere of China 14 through atmospheric transport. The contribution of the Fukushima nuclear accident to ¹²⁹I 15 16 levels in the hydrosphere of China was negligible.

17

18 Keywords

19 Iodine-129; Hydrosphere; permafrost; Tibet, China Sea;

1 1. Introduction

Iodine-129, is a radioisotope of iodine with a half-life of 1.57×10^7 years, that is naturally 2 3 produced by the spontaneous fission of uranium in the Earth's crust and cosmic-rays 4 induced spallation of xenon in the atmosphere (Aldahan et al., 2007a; He et al., 2013). These processes resulted in a ratio of 129 I/ 127 I in the marine reservoir that was between 2× 5 10^{-12} and 6×10^{-13} (Fabryka-Martin et al., 1985; Kilius et al., 1992). The natural inventory 6 of ¹²⁹I was estimated to be approximately 230 kg (Rao and Fehn, 1999), however, this 7 amount is very small compared to recent anthropogenic releases of ¹²⁹I. Major sources of 8 9 anthropogenic ¹²⁹I include releases from: 1) atmospheric nuclear weapon tests, 2) nuclear 10 accidents, 3) nuclear power plants operation and 4) nuclear fuel reprocessing facilities. Atmospheric nuclear weapons tests in 1945-1980 released about 43-150 kg of ¹²⁹I (Carter 11 and Moghissi, 1977; Eisenbud and Gesell, 1997a). Nuclear accidents at Chernobyl in 1986 12 13 released 6.0 kg of ¹²⁹I (Aldahan et al., 2007a) and at Fukushima in 2011 about 1.2 kg of ¹²⁹I (Hou et al., 2013). Emission of ¹²⁹I from the routine operation of nuclear power plants 14 is suggested to be insignificant (Jin et al., 2009; Zhang et al., 2011). Most of anthropogenic 15 ¹²⁹I in the environment has been released from nuclear fuel reprocessing facilities through 16 atmospheric and marine discharges. Besides atmospheric releases (about 400 kg by 2007), 1718 the Sellafield (UK) and La Hague (France) nuclear facilities have discharged about 6500 kg 129 I to the seas (up to 2007)), with an annual discharge of 129 I still remaining at a very 19 high level of about 250 kg/y (Liu et al., 2016a). This ¹²⁹I has contaminated large areas via 20 21 transport by ocean currents. Furthermore, the re-emission of the reprocessing derived ¹²⁹I 22 from the contaminated seawater (about 3% of the marine discharges) to the atmosphere has become an additional source of ¹²⁹I to the atmosphere in recent years (Zhang et al., 2016). 23 Despite the large amount of ¹²⁹I in the environment, direct health hazards are minimal 24 25 owing to its low specific radioactivity (long half-life) (Li et al., 2005). The criteria of the long half-life have, however, strengthened the use of ¹²⁹I as environmental tracer of both 26 27 environmental processes and anthropogenic activities.

There has been many published data on the ¹²⁹I distribution in different Earth reservoirs (atmosphere, hydrosphere, biosphere and soil and sediments) portraying a general picture

of ¹²⁹I contamination and sources in Europe and USA, but such research is meager in China. 31 32 Some investigations have been performed in selected areas (Hou et al., 2000; Li et al., 33 2005; Zhou et al., 2010; Zhang et al., 2011; Ma et al., 2013; Zhang et al., 2014) such as the analysis of ¹²⁹I in seaweed from the south coast region of China and human thyroid samples 34 35 (Hou et al., 2000), as well as grass, seaweed, seawater and pine needles (Li et al., 2005) 36 and local vegetation, soil and precipitation (Zhang et al., 2011). Although these investigations provided valuable data on ¹²⁹I concentrations, the regional distribution 37 pattern of ¹²⁹I in China is far from complete. Consequently, further research concerning ¹²⁹I 38 39 spatial patterns in China is needed to provide a base line for environmental analysis and prediction. In the investigation presented here, we focus on the distribution of ¹²⁹I and ¹²⁷I 40 41 in some parts of the hydrosphere of China including water samples collected from rivers 42 and lakes in the Qinghai-Tibet region, Yangtze Estuary and South China Sea (SCS). In addition, we explore the sources of ¹²⁹I in these regions, assess environmental hazards and 43 establish the possibility of using ¹²⁹I as a chronological indicator. The choice of these water 44 45 systems is based on the fact that none of them has been analyzed for ¹²⁹I and that: 1) The 46 Qinghai-Tibet region is the birthplace of the Yellow River, the second-longest river in Asia 47 and the sixth-longest river system in the world, including Gyaring and Ngöring (Sisters) 48 Lakes (Jin et al., 2009). 2) The Yangtze River is the longest river in Asia and the third-49 longest in the world, and its river basin is home to one-third of the population of China. 3) 50 The South China Sea is a marginal sea that is part of the Pacific Ocean, encompassing an 51 area from the Karimata and Malacca Straits to the Strait of Taiwan of around 3,500,000 52 square kilometers.

53

54 **2. Sampling and Analytical Methods**

55 **2.1 Sampling and Sites**

56 Sixteen freshwater samples from the inland water system of the Qinghai-Tibet region, and 57 2 samples from the Yangtze Estuary were collected. A seawater section in the South China 58 Sea with 10 samples covering the depth interval from 0 m to 3800 m (Figure 1 and Table 59 1) was also analyzed in this work. The inland water samples were collected during 15th -

19th April and 21st - 25th July in 2014 from Oinghai-Tibet region (Figure 1), the source area 60 61 of the Yellow River (SAYR), in four watersheds, Madoi, Ngöring lake, Chalaping and 62 Maqu. These samples were selected to represent landscapes of three different frozen 63 ground types (discontinuous permafrost and seasonal frozen region, talik, and continuous 64 permafrost region) (Jin et al., 2009) and environmental conditions (details in Figure 2). 65 Among these samples, 6 were collected from rivers, 6 from lakes and 4 from groundwater 66 springs. The water samples of the rivers and lakes were taken from the surface (0-5 cm), 67 and the spring water was collected from spring discharge points. The seawater depth profile 68 samples were collected in the SCS in June, 2014. The 2 Yangtze estuary water samples 69 were collected in the estuary area of the East China Sea in June, 2014.

70

71 **2.2 Methods**

The water samples were collected from spring, lake, estuary and sea. However, some of the river water samples contain high amount of suspending material (mud or sand), which was first removed by decantation. The supernatant of these river samples and all other water samples were then filtered through a 0.45 μ m membrane and stored in a clean polyethylene container for analysis. The analytical methods reported by Zhou et al. (2010) for determination of ¹²⁷I and ¹²⁹I in water and sediment samples were used.

For ¹²⁹I measurement, a one liter of water sample was transferred to a beaker, 1.0 mg ¹²⁷I 78 carrier (prepared from Woodward iodine, with a 129 L/ 127 L atomic ratio of 2 × 10⁻¹⁴) and about 79 500 Bq of 125 I were added, and then $K_2S_2O_8$ was added to a concentration of 30 mg/g, 80 81 mixed and covered. The sample was digested at 60°C for 20 h on a hotplate to convert all 82 organic iodine to inorganic form. The solution was then transferred to a separation funnel, 83 3 ml of 1.0 mol/L NaHSO₃ was added, and then HNO₃ was added to lower the sample pH 84 to 1-2 to convert all iodine to iodide. CHCl₃ was added, and then NaNO₂ was added to 85 oxidize iodide to I₂, which was extracted to CHCl₃ phase by shaking. CHCl₃ phase was 86 separated and new CHCl₃ was added to the aqueous phase to extract the remaining iodine 87 to organic phase. CHCl₃ phases were combined and transferred to a new separation funnel. 88 0.1 mol/L NaHSO₃ was added to back extract iodine to water phase. This extraction and back-extraction steps were repeated to purify iodine. ¹²⁵I in the separated solution was 89

- measured using a NaI detector for calculation of the recovery of iodine during separation
 (mean recovery: 85-95%). The separated iodine solution was transferred to a centrifuge
- 92 tube, 2 ml of 3.0 mol/l HNO₃ and 1.0 mL of 1.0 mol/L AgNO₃ were added to precipitate
- 93 iodide as AgCl. The AgI precipitate was separated by centrifuge, then washed with 3.0
- 94 mol/L HNO₃ and H_2O .
- Two ¹²⁹I standard solutions were prepared by diluting ¹²⁹I standard solution (NIST-SRM-95 4949c) using ¹²⁷I solution that was prepared from low ¹²⁹I level iodine carrier (Woodward 96 Inc., ${}^{129}I/{}^{127}I$ ratio < 2 × 10⁻¹⁴). The ${}^{129}I/{}^{127}I$ ratios of the standards are 9.954 × 10⁻¹² and 97 1.138×10^{-10} , respectively. The ¹²⁹I standards as AgI for AMS measurement were prepared 98 99 using above two standard solutions by directly addition AgNO₃ to the standard solution 100 after acidified to pH 2 using HNO₃ and addition of KHSO₃ to convert all iodine to iodide. 101 The prepared AgI precipitate was dried at 70 °C, ground to fine powder mixed with niobium powder (200 mesh) in a mass ratio of 1:5. The mixture was pressed into a copper 102 holder. ¹²⁹I/¹²⁷I ratios in the prepared targets of samples, standards and blanks were 103 measured using 3MV Tandem AMS system in the Xi'an AMS center. I⁵⁺ ions were selected 104 for the measurement. ¹²⁷I⁵⁺ was measured as charges with a Faraday cup, while ¹²⁹I⁵⁺ was 105 106 measured with a gas ionization detector. Each sample was measured for 6 cycles, and 5 107 minutes in each cycle.

108 The concentrations of total iodine (127 I) in the filtered water samples were directly 109 measured using ICP-MS. Thermo X Series^{II} ICP-MS was used under the conditions of hot 110 plasma and Xt interface. The detection limit for 127 I under this condition was calculated to 111 be 0.02 ng/ml.

Procedure blanks were prepared with each batch of samples by the same procedure as the samples, to monitor the laboratory background and cross-contamination in period of separation and measurement. The analytical results showed that the ${}^{129}I/{}^{127}I$ ratios in the procedure blanks (1 × 10⁻¹³) are 2 orders of magnitude lower than that in the most of samples. The blank value was subtracted from measured ${}^{129}I/{}^{127}I$ in the samples.

117

118 **3. Results and Discussion**

120 **3.1 Spatial distribution of iodine isotopes**

The ¹²⁷I and ¹²⁹I concentrations and ¹²⁹I/¹²⁷I atomic ratios in water samples collected from 121 the Qinghai-Tibet region, Yangtze Estuary and SCS are presented in Table 1. The ¹²⁹I/¹²⁷I 122 ratios in the inland water of the Qinghai-Tibet region range from $0.18-21.34 \times 10^{-10}$ with a 123 mean ratio of 8.04×10^{-10} (n=16), in which a range of $0.80-19.77 \times 10^{-10}$ in the river water, 124 $0.23-21.34 \times 10^{-10}$ in the lake water, and $0.18-2.40 \times 10^{-10}$ in the spring water. As for ¹²⁹I 125concentrations, they span 5.36-42.68 \times 10⁶ atoms/L in the river water, 2.04-51.22 \times 10⁶ 126 atoms/L in the lake water, and $1.26-4.34 \times 10^6$ atoms/L in the spring water. River water 127 128 samples No. 2, 3 and 6 were collected from the main stream of the Yellow River, while 129 samples No. 1, 4, 5 were collected from the tributaries (Figure 2). The data show that there is no significant difference between the main stream and tributary samples with respect to 130 ¹²⁷I concentrations. Lake water samples No. 9, 11, 12 collected from a saltwater lake have 131 higher ¹²⁹I concentrations than those water samples collected from a freshwater lake 132 (Figure 1 and Figure 2). The ¹²⁹I concentrations in the spring water generally show the 133 134 lowest value compared to the river and lake water in Qinghai-Tibet region, except for one 135lake water sample that had a very low ¹²⁹I concentration.

In the South China Sea (SCS), ¹²⁹I concentrations in the upper 1000 m of the depth profile 136 are higher than those of the deep water (Figure 3). The ¹²⁹I concentrations steeply decreased 137 in the depth from 1000 m to 2000 m. At depths below 2000 m, the ¹²⁹I concentration slightly 138 increases to 3.16×10^6 atoms/L at depth of 3000 m, but the concentration drops again below 139 3000 m. At the depth of 3800 m, the concentration of 129 I is extremely low (0.99 × 10⁶ 140 atoms/L), corresponding to a $^{129}I/^{127}I$ ratio of 0.03×10^{-10} . The trend of ^{129}I concentrations 141 against depth is like that of ¹²⁹I/¹²⁷I ratio, but an inverse trend is observed for the ¹²⁷I 142 concentrations (Figure 3). In the SCS, the lowest ¹²⁷I concentration appears in the surface 143 144 water, and then gradually increases from 56 μ g/L at surface to 68.6 μ g/L at 1500 m, then 145 remains relative constant at 65-68 µg/L from 1500- to 3800 m. This depth distribution pattern of ¹²⁷I is comparable with the stratification effect of the ocean current (Liu et al., 146 2016b). The slightly low ¹²⁷I in surface water may be attributed to the dilution of the low 147 ¹²⁷I precipitation (Reithmeier et al., 2010) and/or loss of iodine from the surface water by 148 formation and emission of volatile iodine species (He et al., 2013). Overall, the data cluster 149 150 into three different layers at 0 - 1000 m, 1000 - 2000 m and below 2000 m (Figure 3).

151 The ¹²⁹I depth profile distribution (Figure 3) at the sampling location in the SCS indicates 152that the occurrence of 3 different layers might correspond to different water sources at 153different depths. This is supported by other observations of ocean currents at different 154 depths in the SCS. It was reported that SCS surface water movement is controlled by a 155strong winter monsoon (Northeast monsoon drift) from November to April, and the weaker 156summer monsoon (Southeast monsoon drift) from May to August (Figure 4) (Liu et al., 2016b). The higher 129 I concentration in the upper layer (<1000m) compared to the deep 157layer water might be attributable to a higher contribution of anthropogenic ¹²⁹I input 158through global fallout from 1945-1980 and riverine input from the inland of China (Liu et 159al., 2016a). The relatively constant distribution of ¹²⁹I in the upper 1000 m water may be 160 161 caused by mixing induced by the summer and winter monsoon (Liu et al., 2016a). The steeply decreasing ¹²⁹I concentration from 1000 m to 2000 m can be attributed to limited 162 transfer of ¹²⁹I from the above water layer. Schink et al. (1995) has proposed that the flux 163 164 of water mass from the surface to the deep-water inventory is guite small and has essentially 165 no bearing on the vertical distribution and thus leading to little mixing of the upper 1000 166 m water with the 1000 m to 2000 m deep waters. A southward deep contour current (2000 167 to 2500 m) from Luzon Strait along the continental margin off southeast China has been reported (Ou et al., 2006). This water mass, carrying low ¹²⁹I concentration from the Pacific 168 deep water, enters the SCS leading to the low ¹²⁹I level in this layer. The slightly increased 169 ¹²⁹I level at 3000 m depth suggests the intrusion of other water masses. One possibility is 170 171that a deep cyclonic current below 2400 m which has been reported in the SCS (Wang et al., 2011), which may bring some high ¹²⁹I surface water from other locations to the deep 172 part of the SCS. This process was also indicated by the distribution of bomb produced Δ^{14} C 173 174 and tritium in nearby East Sea. Activities of radiocarbon decrease from a maximum in the 175upper mixed layer to 2000 m depth, increase to a peak value at 2800 m and then decrease 176 again towards the bottom, which is comparable to the tritium depth distribution (Watanabe 177et al., 1991; Cooper et al., 2001).

178 The ¹²⁹I concentrations in the two samples collected from the Yangtze Estuary ((17-21)

179 $\times 10^6$ atoms/L) are higher than those in the surface seawater of the SCS ((11-14) $\times 10^6$

- 180 atoms/L). This suggests different sources of 129 I in the Yangtze estuary of the ECS and the
- 181 SCS. ¹²⁹I in the ECS mainly originates from the input of the Yangtze River, while ¹²⁹I in

the SCS reflects mainly global fallout, and to lesser extent riverine input. In addition to data presented here (Table 1 and Figure 3), reported values of iodine isotopes in the surface water of the East China Sea (ECS) (Liu et al., 2016a) were used to plot the distribution of iodine isotopes in surface water of the ECS (Figure 4).

186 The measured values of iodine isotopes in the SCS and Yangtze River estuary reported here fit well with the distribution of ¹²⁷I and ¹²⁹I concentrations and ¹²⁹I/¹²⁷I ratios in the 187 ECS and the water circulation patterns of the SCS (Figure 4d). It has been reported that the 188 Yangtze River water enters the ECS through the Yangtze Estuary where ¹²⁷I concentrations 189 increase seaward, while ¹²⁹I decreases seaward (Figure 4) (Liu et al., 2016a). This feature 190 suggests decoupling of the ¹²⁹I and ¹²⁷I in the Yangtze Estuary that is related to different 191 sources of water. The high-level ¹²⁹I in the fresh water is diluted by the low ¹²⁹I seawater, 192 while the low-level ¹²⁷I in the riverine water is diluted by the high ¹²⁷I concentration 193 seawater when the river water enters the sea. The increased 129 I concentrations and 129 I/ 127 I 194 195 ratios from the SCS to the ECS was observed (Figure 4) and might be attributed to the different sources of ¹²⁹I in the ECS and SCS, and the water mass exchange processes driven 196 by the monsoons in this region. Due to the anthropogenic high ¹²⁹I deposition in the middle 197 198 and northern North Hemisphere, the ¹²⁹I deposition and input from the rivers to the ECS is 199 higher than that in the SCS. The surface currents in the SCS are mainly controlled by the 200 monsoon drift, with the southwest current direction prevailing in summer and the northeast 201 in winter (Daryabor et al., 2016). This pattern results in that the major movement of surface 202 water follows the monsoon's pathways along the northeast direction and providing a 203 monsoon-related mixing process.

204 The results presented above provide a general fingerprinting of the different water types (inland water, estuary water and seawater) with respect to their ¹²⁷I concentrations and 205 ¹²⁹I/¹²⁷I ratios (Figure 5). The inland water, namely, the river water, lake water and spring 206 water, tends to have higher ¹²⁹I/¹²⁷I ratios, but lower ¹²⁷I concentrations, while the seawater 207 tends to have lower ¹²⁹I/¹²⁷I ratios but higher ¹²⁷I concentrations. The ¹²⁷I concentrations 208 and ¹²⁹I/¹²⁷I ratios of the estuary water lies between that of inland water and seawater. The 209 data for ¹²⁹I/¹²⁷I versus ¹²⁹I in the SCS sea water track the correlation line of seawater 210 samples and extend to lower ¹²⁹I/¹²⁷I values. In the terrestrial hydrosphere the iodine data 211 212 plot along line of the surface and groundwater of Europe and again extend the European data to a lower ¹²⁹I/¹²⁷I ratios (Figure 6). The same holds for the ¹²⁹I/¹²⁷I versus ¹²⁷I plot,
except for a discrepancy between the ¹²⁷I concentrations in this study and that presented by
Michel et al. (2012). The results suggest dominance of the emission from the European
NFRPs on the ¹²⁹I global distribution.

217

218 **3.2**¹²⁹I in hydrology of permafrost area in Qinghai-Tibet region

The results of ¹²⁹I in surface water in the Oinghai-Tibet region show relatively higher ¹²⁹I 219 220 levels in the main river samples (No. 2, 3, 6) than the tributaries (No. 1, 4, 5). This 221 difference may be attributed to the complex and varied sources of water in the main stream 222 that receives contributions from the Ngöring lake with many small tributaries and sewer 223 water from Madoi Town. These sources are all located within a discontinuous permafrost, 224 seasonally frozen ground and talik (thermokarst lakes) dominated landscape where surface 225 and subsurface water interacts in a complex pattern. Furthermore, the ¹²⁹I concentrations and ¹²⁹I/¹²⁷I ratios of the main river water (sampling site 2 and 6) were identical to the lake 226 227 water nearby (sampling site No. 9), indicating a common source from the unfrozen 228 wetlands in the area as well as collapsed peatlands (Jin et al., 2009). During warm periods, 229 thawing of active topsoil layer enhances interflow and shallow groundwater discharge 230 increases (Wang et al., 2009). This process enhances the precipitation-coupled transfer of 231 ¹²⁹I deposited on the soil surface into the river. Tributaries water samples (No. 1 and 5), 232 which were mainly generated from the melt snow or recent rainfall that contained relatively low ¹²⁹I concentration. The thaw lake water was also characterized by a relatively high 233 isotopic ratio (sample No 9 in Xingxinghai lake) compared to other lakes. A high ¹²⁹I value 234 235 of tributary water was also noticed in Chalaping (sample No. 4), where a large number of 236 thaw lakes are developed. Consequently, wetland water may recharge the rivers (Gibson et al., 2016), which may become enriched in ¹²⁹I owing to accumulation in the organic-rich 237 238 active layer of soil (Herod et al., 2016). The association of iodine with soil organic matters has been suggested by several investigations (Hou et al., 2003; Luo et al., 2013). 239

The ¹²⁹I concentrations ((1.2-4.3) ×10⁶ atoms/L) and ¹²⁹I/¹²⁷I ratio ((0.2-2.4) × 10⁻¹⁰) in the spring water samples (Table 1) is much lower than those in the river and lake water.

However, they are higher than the reproted pre-nuclear level $(1.5 \times 10^{-12} \text{ for } {}^{129}\text{L})^{127}$ ratio)

243 in the marine system. This trend suggests that spring water in the Oinghai-Tibet region is 244 mainly recharged from deep groundwater that was partly isolated from the modern 245 environment by the permafrost as an impermeable layer (Yang et al., 2016). Fast transport 246 through most surface water reservoirs and isotopic equilibrium characterize naturally produced iodine with a pre-nuclear era concentration of ¹²⁹I in surface water at (0.1-2) \times 247 10⁴ atoms/L (Kohman and Edwards, 1966; Fabryka-Martin et al., 1985; Kilius et al., 1992; 248 249 Rao and Fehn, 1999). The isotopic equilibrium of iodine isotopes in surface reservoirs has 250 been disturbed since the start of the nuclear era activities in the 1940s. Consequently, the concentration of ¹²⁹I measured in the spring waters is two or three orders of magnitude 251252 higher than the pre-nuclear level (Table 1). This fact indicates that the spring water in 253 Qinghai-Tibet region could be young (< 70 years) or it reflects mixing with groundwater 254 recharged by post-nuclear era precipitation.

255

256 **3.3 Sources of ¹²⁹I in the hydrosphere of China**

The pre-nuclear ratio of ${}^{129}\text{I}/{}^{127}\text{I}$ in the marine environment was estimated to be 1.5×10^{-12} 257 (Moran et al., 1998; Schmidt et al., 1998; Fehn et al., 2013). With an average concentration 258 259 of stable iodine in the SCS of 60 µg/L (Hou et al., 2002), the contribution from natural source would be < 5 % of ¹²⁹I in the SCS. A pre-nuclear ¹²⁹I/¹²⁷I ratio of 2×10⁻¹¹ in terrestrial 260 system was estimated by determination of ¹²⁹I in the soil of China (Fan et al. 2018). 261 Assuming equilibrium of natural ¹²⁹I and ¹²⁷I in the soil and terrestrial water in the pre-262 nuclear age, the contribution of natural ¹²⁹I to the most river and lake water in the Qinghai-263 Tibet region can be negligible. However, for some river and lake water with low ¹²⁹I/¹²⁷I 264 ratio of (2.3-7.9) $\times 10^{-11}$ and all spring water with ${}^{129}\text{L}/{}^{127}\text{I}$ ratios of (1.7-24) $\times 10^{-11}$, the 265 266 natural ¹²⁹I might be the major source.

267 Another source of ¹²⁹I is related to the nuclear weapons tests (NWTs), mostly took place in

268 1945-1963 and released 43-150 kg of ¹²⁹I to the environment (Carter and Moghissi, 1977;

- 269 Chamberlain, 1991; Eisenbud and Gesell, 1997b). As most testing sites were situated at
- 270 mid-latitudes of the northern hemisphere, atmospheric ¹²⁹I was mainly deposited in the
- 271 northern hemisphere (Reithmeier et al., 2010; Snyder et al., 2010; He et al., 2013). It has
- been reported that ¹²⁹I level in rivers in USA was elevated to $1-5 \times 10^7$ atoms/L (Moran et
- al., 2002) as a result of the NWTs. This range is comparable to the values in the rivers of

the Qinghai-Tibet region $(0.54-4.27 \times 10^7 \text{ atoms/L})$ and the waters in Yangtze Estuary (1.77-2.11 × 10⁷ atoms/L). The contribution from NWTs to the SCS is complicated and is associated with a large uncertainty, but an increase to $1.22 \times 10^6 \text{ atoms/L}$ in surface water due to atmospheric nuclear weapons tests may provide a small contribution of the total measured ¹²⁹I.

- 279 The Chernobyl accident happened in 1986 and Fukushima accident are two serious nuclear accident in history and released about 6.0 kg and 1.2 kg¹²⁹I to the environment, 280 respectively (Aldahan et al., 2007a; Hou et al. 2013). Considering that most of ¹²⁹I released 281 from the Chernobyl accident was deposited in Europe, its contribution as a remote source 282 of 129 I in the Oinghai-Tibet region is very limited. Measurement of 129 I/ 127 I ratios in river 283 waters in Xi'an region before the Fukushima accident indicated values of $7.4-37.3 \times 10^{-10}$ 284 (Zhang et al., 2011). The 129 I/ 127 I ratios of 0.8-19.8 × 10⁻¹⁰ in the river waters measured in 285 286 the Qinghai-Tibet region (Table 1) in this work (after the accident) do not show any visible 287 change due to more recent inputs from Fukushima, which indicates that the contribution 288 from the Fukushima accident to the Qinghai-Tibet region is negligible.
- 289 As of 2017, 36 nuclear power reactors are in operation in China. It was found that surface 290 seawater samples collected at sites with distance of 0.5-10 km to the outlet of a nuclear power plant in China had ${}^{129}I/{}^{127}I$ ratios of ((5.7-9.5) × 10⁻¹⁰), which are similar to values 291 292 measured in the seawater collected at the discharge outlet (Zhang et al., 2012; Zhang et al., 2014). The ¹²⁹I/¹²⁷I ratios ((0.8-2.6) \times 10⁻¹⁰) at other distant sites (2-7 km to the outlet) 293 294 indicated the same levels as of those reported elsewhere in China which are far away from any nuclear facility. These data suggest insignificant ¹²⁹I discharge from the power plant 295 (He et al., 2011). In addition, there are no reports of ¹²⁹I releases from other NPPs in Asia 296 or other parts of the world. Therefore, the contribution from NPPs to the ¹²⁹I inventory in 297 298 China is considered negligible.
- Nuclear fuel reprocessing plants, especially Sellafield (UK) and La Hague (France), have released around 5700 kg of ¹²⁹I (up to 2009) to the environment, which account to more than 90% of ¹²⁹I inventory in the present environment (Aldahan et al., 2007b; He et al., 2013). Regions near these nuclear fuel reprocessing plants have shown high ¹²⁹I concentrations (Figure 7) (Chen et al., 2015). For instance, the reported ¹²⁹I concentrations
- in precipitation from North Europe reached up to 300×10^8 atoms/L (Aldahan et al., 2009).

- Releases of ¹²⁹I from La Hague and Sellafield to the atmosphere occur via direct atmospheric emission and remission from seawater, resulting in a global-scale contamination. Due to its relatively long atmospheric residence time of 2-3 weeks (Englund et al., 2010; Reithmeier et al., 2010).
- 309 The source of water to the lakes in the Qinghai region include mainly precipitation with 310 addition of some groundwater. Differentiating the contribution of groundwater to the lakes is neglected here and we consider only ¹²⁹I signal from precipitation. Most ¹²⁹I found in 311 312 lakes of the Qinghai region and Yangtze Estuary originates from precipitation, thus the 313 portion of atmospheric loading that may have been largely sourced from the NFRP's 314 atmospheric emissions and re-emission of mainly the NFRP's marine discharges should be 315 a major source in the lake and river water in Qinghai region. For the SCS, its average ¹²⁹I concentration is about 6.6×10^6 atoms/L, it is lower than the ¹²⁹I concentration in the 316 Qinghai-Tibet region (18×10^6 atoms/L) and Yangtze River Estuary (19×10^6 atoms/L). It 317 has been reported that the discharges of ¹²⁹I from the European NFRP has a significant 318 319 contribution to the ECS through riverine input and directly atmospheric deposition (Liu et 320 al., 2016a). Meanwhile, the NFRP might also contribute to the 129I inventory in the 321 seawater in the SCS. A significant contribution of NFRP to 129I in seawater in the Pacific 322 Ocean and the Japan Sea have also been reported (Suzuki et al., 2010).
- 323

324 3.4 Comparison with the global trends

The comparison of our results with other published data (Figure 7) shows that the ${}^{129}I/{}^{127}I$ 325 326 ratios measured in the hydrosphere of China are consistent with those observed in other locations in Asia and North America. The ¹²⁹I/¹²⁷I ratio of many rivers shows comparable 327 values, such as 7.7×10^{-10} in the Pearl River, $10.7-24.4 \times 10^{-10}$ in the Atchafalaya River 328 (both located at U.S South Coast), and $7.4-37.2 \times 10^{-10}$ in river water in the Xi'an region, 329 China (Moran and Oktay, 2002; Zhang et al., 2011). However, the ratio in these rivers is 330 1-3 orders of magnitude lower than in those of western Europe, such as 1.84×10^{-8} in Rhine 331 River and 1.88×10^{-8} in River Thames (Moran and Oktay, 2002; Snyder and Fehn, 2004), 332 where a significant influence from the European NFRPs was received. These distribution 333 patterns follow the trend of global distribution of ¹²⁹I in the surface environment where 334

335 sampling sites near NFRPs tend to have relatively high 129 I concentrations (around 10^{10} -

 10^{11} atoms/L). At the time, releases from nuclear accident (e.g. Chernobyl and Fukushima

337 accidents) could be highly localized but comparatively negligible when considering the

influences on global scale (Nagai et al., 2015; Chen et al., 2016).

339

340 **4. Conclusion**

341 Based on the results and discussion above, it can be concluded that:

342 1. The ¹²⁹I concentrations in the hydrosphere of China reflect anthropogenic input with

343 varying degrees being more on land hydrological system (rivers and lakes) than in the sea.

344 2. The ¹²⁹I depth profiles in the SCS reflect distribution pattern largely controlled by
 345 monsoon drift in the surface and cyclonic currents at depth.

346 3. Major source of ¹²⁹I in China is attributed to atmospheric transport from the NFRPs

emissions (direct atmospheric emission and re-emission from the marine water) and globalfallout of nuclear weapons tests.

349 4. The effect of Fukushima nuclear accident on the ¹²⁹I concentration in the hydrosphere of
350 China is negligible.

351

352 Acknowledgements

353 This research was funded in part by the Special Fund of State Key Laboratory of

354 Hydrology-Water Resources and Hydraulic Engineering [Grant No. 20165042512 and No.

355 20155045612], the Fundamental Research Funds for the Central Universities [Grant No.

356 2017B10314 and No. 2016B04214], and Postgraduate Research & Practice Innovation

³⁵⁷ Program of Jiangsu Province [Grant No. KYZZ16_0277]. The first two authors contributed

acceleration and should be considered co-first authors.

359

360 **References**

- Aldahan, A., Alfimov, V., & Possnert, G. (2007a). ¹²⁹I anthropogenic budget: major
 sources and sinks. Applied Geochemistry, 22(3), 606-618.
- Aldahan, A., Possnert, G., Alfimov, V., Cato, I., & Kekli, A. (2007b). Anthropogenic ¹²⁹I
 in the baltic sea. Nuclear Inst & Methods in Physics Research B, 259(1), 491-495.
- Aldahan, A., Persson, S., Possnert, G., & Hou, X. L. (2009). Distribution of ¹²⁷I and ¹²⁹I in
 precipitation at high European latitudes. Geophysical Research Letters, 36(11), 192 200.
- Carter, M. W., & Moghissi, A. A. (1977). Three Decades of Nuclear Testing. Health
 Physics, 33(1), 55-71.
- 370 Chamberlain, A. C. (1991). Radioactive aerosols. Radioactive Aerosols, 51(07), 11–58.
- Chen, X. G., Gong, M., Yi, P., Aldahan, A., Yu, Z. B., & Possnert, G., et al. (2015).
 Distribution of ¹²⁹I in terrestrial surface water environments. Nuclear Instruments &
 Methods in Physics Research, 361, 604-608.
- Chen, X. G., Liu, X., Yi, P., Aldahan, A., Yu, Z. B., & Chen, L., et al. (2016). Estimation
 of ¹²⁹I inventory in the oceans. Journal of Radioanalytical & Nuclear Chemistry,
 308(1), 59-65.
- Cooper, L. W., Hong, G. H., Beasley, T. M., & Grebmeier, J. M. (2001). Iodine-129
 concentrations in marginal seas of the north pacific and pacific-influenced waters of
 the Arctic Ocean. Marine Pollution Bulletin, 42(12), 1347-1356.
- Daryabor, F., Ooi, S. H., Samah, A. A., & Akbari, A. (2016). Dynamics of the water
 circulations in the southern South China Sea and its seasonal transports. Plos One,
 11(7), e0158415.
- 383 Eisenbud, M., Gesell, T. (1997a). Chapter 9 Nuclear Weapons. Environmental
 384 Radioactivity, 266-315.
- Eisenbud, M., Gesell, T. (1997b). Chapter 6 Natural Radioactivity. Environmental
 Radioactivity, 6(3), 134-200.
- Englund, E., Aldahan, A., Hou, X. L., Possnert, G., & Söderström, C. (2010). Iodine (¹²⁹I
 and ¹²⁷I) in aerosols from northern Europe. Nuclear Instruments & Methods in Physics
 Research, 268(7–8), 1139-1141.
- Fabryka-Martin, J., Bentley, H., Elmore, D., & Airey, P. L. (1985). Natural iodine-129 as
 an environmental tracer. Geochimica Et Cosmochimica Acta, 49(2), 337-347.

- Fan, Y.K., Zhou W.J., Hou X.L. (2018). Pre-nuclear level of ¹²⁹I in Chinese loess-paleosol
 sections: A search for the natural ¹²⁹I level for dating in terrestrial environments.
 Geochim. Cosmochim. Acta, 231, 64-72.
- Fehn, U., Snyder, G., & Egeberg, P. K. (2000). Dating of pore waters with ¹²⁹I: relevance
 for the origin of marine gas hydrates. Science, 289(5488), 2332.
- Fehn, U., Holdren, G. R., Elmore, D., Brunelle, T., Teng, R., & Kubik, P. W. (2013).
 Determination of natural and anthropogenic ¹²⁹I in marine sediments. Geophysical
 Research Letters, 13(2), 137-139.
- Gibson, J. J., Birks, S. J., & Yi, Y. (2016). Higher tritium concentrations measured in
 permafrost thaw lakes in northern Alberta. Hydrological Processes, 30(2), 245-249.
- He, C., Hou, X., Zhao, Y., Wang, Z., Li, H., & Chen, N., et al. (2011). ¹²⁹I level in seawater
 near a nuclear power plant determined by accelerator mass spectrometer. Nuclear
 Instruments & Methods in Physics Research, 632(1), 152-156.
- He, P., Aldahan, A., Possnert, G., & Hou, X. L. (2013). A summary of global ¹²⁹I in marine
 waters. Nuclear Instruments & Methods in Physics Research, 294, 537-541.
- Herod, M. N., Li, T., Pellerin, A., Kieser, W. E., & Clark, I. D. (2016). The seasonal
 fluctuations and accumulation of iodine-129 in relation to the hydrogeochemistry of
 the wolf creek research basin, a discontinuous permafrost watershed. Science of the
 Total Environment, 569-570, 1212-1223.
- Hou, X. L., Dahlgaard, H., Nielsen, S. P., & Ding, W. J. (2000). Iodine-129 in human
 thyroid and seaweed in China. Science of the Total Environment, 246(2-3), 285-291.
- Hou, X. L., Dahlgaard, H., Nielsen, S. P., & Kucera, J. (2002). Level and origin of iodine129 in the Baltic Sea. Journal of Environmental Radioactivity, 61(3), 331-343.
- Hou, X. L., Fogh, C. L., Kucera, J., Andersson, K. G., Dahlgaard, H., & Nielsen, S. P.
 (2003). Iodine-129 and caesium-137 in Chernobyl contaminated soil and their
 chemical fractionation. Science of the Total Environment, 308(1), 97-109.
- Hou, X. L., Povinec, P. P., Zhang, L., Shi, K., Biddulph, D., & Chang, C. C., et al. (2013).
 Iodine-129 in seawater offshore fukushima: distribution, inorganic speciation, sources,
- 420 and budget. Environmental Science & Technology, 47(7), 3091-8.
- Jin, H. J., He, R. X., Cheng, G. D., Wu, Q. B., Wang, S. L., & Chang, X. L. (2009). Changes
 in frozen ground in the source area of the yellow river on the Qinghai-Tibet plateau,

- 423 China, and their eco-environmental impacts. Environmental Research Letters, 4(4),
 424 045206.
- Kilius, L. R., Litherland, A. E., Rucklidge, J. C., & Baba, N. (1992). Accelerator massspectrometric measurements of heavy long-lived isotopes. International journal of
 radiation applications and instrumentation. Part A. Applied radiation and isotopes,
 43(1-2), 279-287.
- Kohman, T. P., & Edwards, R. R. (1966). ¹²⁹I as a geochemical and ecological tracer.
 Progress Report, December 1, 1965-October 31, 1966 (No. NYO--3624-1). Carnegie
 Inst. of Tech., Pittsburgh, Pa. Dept. of Chemistry.
- Li, B., Zhang, P. Q., Chen, C. Y., He, M., & Chai, Z. F. (2005). Determination of ¹²⁹I levels
 in Chinese biological and environmental specimens by accelerator mass spectrometry.
 Chinese Journal of Analytical Chemistry, 33(7), 904-908.
- Liu, D., Hou, X., Du, J., Zhang, L., & Zhou, W. (2016a). ¹²⁹I and its species in the East
 China Sea: level, distribution, sources and tracing water masses exchange and
 movement. Scientific reports, 6, 36611.
- Liu, Z., Zhao, Y., Colin, C., Stattegger, K., Wiesner, M. G., & Huh, C. A., et al. (2016b).
 Source-to-sink transport processes of fluvial sediments in the South China Sea. EarthScience Reviews, 153, 238-273.
- Luo, M., Hou, X., Zhou, W., He, C., Chen, N., & Liu, Q., et al. (2013). Speciation and
 migration of ¹²⁹I in soil profiles. Journal of Environmental Radioactivity, 118, 30.
- Ma, X., Song, Y., Liu, S., Jiang, L., & Hong, F. (2013). Origin and evolution of waters in
 the Hancheng coal seams, the Ordos Basin, as revealed from water chemistry and
 isotope (H, O, ¹²⁹I) analyses. Science China Earth Sciences, 56(11), 1962-1970.
- Michel, R., Daraoui, A., Gorny, M., Jakob, D., Sachse, R., & Tosch, L., et al. (2012).
 Iodine-129 and iodine-127 in European seawaters and in precipitation from northern
 Germany. Science of the Total Environment, 419(3), 151-169.
- Moran, J. E., Fehn, U., & Teng, R. T. D. (1998). Variations in ¹²⁹I/¹²⁷I ratios in recent
 marine sediments: evidence for a fossil organic component. Chemical Geology,
 152(1–2), 193-203.
- Moran, J. E., Oktay, S. D., & Santschi, P. H. (2002). Sources of iodine and iodine 129 in
 rivers. Water resources research, 38(8).

- 454 Nagai, H., Hasegawa, A., Yamagata, T., Kumamoto, Y., Nishino, S., & Matsuzaki, H.
 455 (2015). Anthropogenic ¹²⁹I in the North Pacific, Bering and Chukchi Seas, and Arctic
 456 Ocean in 2012-2013. Nuclear Inst & Methods in Physics Research B, 361, 680-684.
- 457 Qu, T., Girton, J. B., & Whitehead, J. A. (2006). Deepwater overflow through Luzon strait.
 458 Journal of Geophysical Research: Oceans, 111(C1).
- Rao, U., & Fehn, U. (1999). Sources and reservoirs of anthropogenic iodine-129 in western
 New York. Geochimica Et Cosmochimica Acta, 63(13–14), 1927-1938.
- 461 Reithmeier, H., Lazarev, V., Rühm, W., & Nolte, E. (2010). Anthropogenic ¹²⁹I in the
 462 atmosphere: overview over major sources, transport processes and deposition pattern.
 463 Science of the Total Environment, 408(21), 5052-5064.
- Schink, D. R., Santschi, P. H., Corapcioglu, O., Sharma, P., & Fehn, U. (1995). ¹²⁹I in gulf
 of mexico waters. Earth & Planetary Science Letters, 135(1–4), 131-138.
- Schmidt, A., Schnabel, C., Handl, J., Jakob, D., Michel, R., & Synal, H. A., et al. (1998).
 On the analysis of iodine-129 and iodine-127 in environmental materials by
 accelerator mass spectrometry and ion chromatography. Science of the Total
 Environment, 223(2–3), 131-156.
- Snyder, G., & Fehn, U. (2004). Global distribution of ¹²⁹I in rivers and lakes: implications
 for iodine cycling in surface reservoirs. Nuclear Instruments & Methods in Physics
 Research, 223-224, 579-586.
- Snyder, G., Aldahan, A., & Possnert, G. (2010). Global distribution and long-term fate of
 anthropogenic ¹²⁹I in marine and surface water reservoirs. Geochemistry Geophysics
 Geosystems, 11(4).
- Suzuki, T., Minakawa, M., Amano, H., & Togawa, O. (2010). The vertical profiles of
 iodine-129 in the Pacific Ocean and the japan sea before the routine operation of a
 new nuclear fuel reprocessing plant. Nuclear Instruments & Methods in Physics
 Research, 268(7–8), 1229-1231.
- Wang, G. X., Hu, H. C., & Li, T. B. (2009). The influence of freeze-thaw cycles of active
 soil layer on surface runoff in a permafrost watershed. Journal of Hydrology, 375(3),
 438-449.
- Wang, G., Xie, S., Qu, T., & Huang, R. X. (2011). Deep South China Sea circulation.
 Geophysical Research Letters, 38(5), 3115-3120.

- Watanabe, Y. W., Watanabe, S., & Tsunogai, S. (1991). Tritium in the japan sea and the
 renewal time of the japan sea deep water. Marine Chemistry, 34(1–2), 97-108.
- Yang, Y., Wu, Q., Yun, H., Jin, H., & Zhang, Z. (2016). Evaluation of the hydrological
 contributions of permafrost to the thermokarst lakes on the Qinghai–Tibet Plateau
 using stable isotopes. Global and Planetary Change, 140, 1-8.
- Zhang, L., Zhou, W., Hou, X., Chen, N., Liu, Q., & He, C., et al. (2011). Level and source
 of ¹²⁹I of environmental samples in Xi'an region, China. Science of the total
 environment, 409(19), 3780-3788.
- Zhang, H., Chen, Y., Hou, X., Sun, H., Lu, Y., & Zhang, L. (2012). The distribution and
 source apportionment of ¹²⁹I in coastal seawater of Shenzhen Dapeng Peninsula.
 Journal of Shenzhen University Science & Engineering, 29(1), 1-6 (in Chinese).
- Zhang, R., Zhang, H., Hou, X., Chai, Z., Chen, Y., & Fan, Y. (2014). ¹²⁹I assessment
 reveals the impact of Fukushima incident on Dapeng Peninsula, Shenzhen, China.
 Journal of Radioanalytical and Nuclear Chemistry, 301(1), 57-63.
- Zhang, L., Hou, X., & Xu, S. (2016). Speciation of ¹²⁷I and ¹²⁹I in atmospheric aerosols at
 Risø, Denmark: insight into sources of iodine isotopes and their species
 transformations. Atmospheric Chemistry and Physics, 16(4), 1971-1985.
- Zhou, W. J., Hou, X. L., Chen, N., Zhang, L. Y., Liu, Q., & He, C. H., et al. (2010).
 Preliminary study of radioisotope ¹²⁹I application in China using Xi'an accelerator
 mass spectrometer. INCS News, 7(1), 8-23.

520 Figure captions

- 521 Figure 1 Locations of sampling sites. Water samples reported in literature (Zhang et al.,
- 522 **2011**) are marked by grey circles. The ¹²⁹I concentrations in the samples are 523 shown as the size of the circles.
- 524 **Figure 2** Distributions of ¹²⁹I concentrations in the water samples of rivers, lakes and 525 springs in the source area of the Yellow River (SAYR), Qinghai-Tibet region.
- Figure 3 Variations of ¹²⁷I concentrations (a), ¹²⁹I/¹²⁷I ratios (b) and ¹²⁹I concentrations (c)
 in seawater with depth in a water section of the SCS. The red dash lines show
 water layer boundaries.
- Figure 4 ¹²⁷I concentration (a), ¹²⁹I concentration (b) and ¹²⁹I/¹²⁷I ratio (c) distributions of
 Yangtze Estuary towards the East China Sea (ECS) (data from Liu et al. (2016a))
 and the SCS. SCS water movement (d) is controlled by the strong winter monsoon
 (Northeast monsoon drift) from November to April and weaker summer monsoon
 (Southeast monsoon drift) from May to August, for the surface water; the deep
 water is controlled by deep contour and cyclonic currents (Liu et al., 2016b).
- Figure 5 Relationship between ¹²⁷I concentrations and ¹²⁹I/¹²⁷I atomic ratios in inland
 water, estuary water and seawater.
- Figure 6¹²⁹I/¹²⁷I isotopic ratios versus ¹²⁷I and ¹²⁹I concentrations, respectively, of all 537 538 samples (a, big size) and results of Michel et al. (2012) (b, small size). (a) Blue 539 squares are surface water, blue circles are ground water, magenta triangles are 540 estuary water and red triangles are SCS sea water. (b) Published data contains: 541 seawater (triangles), precipitation (open circles and shaded circles), surface water 542 (orange squares and open squares), ground water (black circles), and topsoil 543 (black diamonds). Eye-guides for European seawater were plotted with red dash 544 lines (Michel et al., 2012).
- Figure 7 Comparison of ¹²⁹I level in this study (triangles) with the reported data in terrestrial surface water in other places of the world (circles) (Chen et al., 2015).
- 547
- 548
- 549

Sampling ID	Sampling site		Depth	¹²⁷ I concentration	¹²⁹ I concentration	¹²⁹ I/ ¹²⁷ I ratio
	°N	°E	m	μg/L	$\times 10^{6}$ atoms/L	×10 ⁻¹⁰
River						
1	34°39'59.37"	98°08'16.35"		14.3±0.2	5.36 ± 0.55	$0.792{\pm}0.082$
2	34°52'21.94"	98°11'16.93"		7.16±0.17	25.0±1.9	$7.40{\pm}0.56$
3	34°52'01.56"	97°30'21.95"		7.81±0.19	42.7±3.2	11.6 ± 0.9
4	34°19'33.13"	97°55'28.19"		4.45 ± 0.14	27.0±1.7	12.8 ± 0.8
5	34°27'26.33"	97°43'9.22"		$2.28{\pm}0.07$	$11.0{\pm}0.9$	10.6 ± 0.9
6	35°06'34.66"	97°49'55.29"		3.95±0.11	36.4 ± 0.9	19.8 ± 0.5
Lake						
7	34°18'56.22"	98°37'16.43"		6.17±0.15	6.58±1.37	2.26 ± 0.44
8	34°51'22.61"	98°07'51.63"		19.1±0.3	2.04 ± 0.26	0.225 ± 0.030
9	34°46'52.61"	98°09'42.86"		7.58±0.19	45.8±2.6	12.8 ± 0.7
10	33°54'38.79"	102°49'1.15"		2.41 ± 0.07	8.21±0.76	$7.10{\pm}0.66$
11	35°05'6.16"	96°33'26.61"		2.18 ± 0.06	17.7±1.6	16.9 ± 1.5
12	36°33'15.98"	100°39'1.38"		5.14 ± 0.08	51.2±1.5	21.3±0.6
Spring						
13	34°22'29.15"	98°32'22.27"		5.17±0.14	3.46 ± 0.32	1.41 ± 0.13
14	35°24'16.69"	99°25'47.95"		15.3±0.3	1.26 ± 0.21	0.173 ± 0.031
15	35°02'58.18"	96°14'42.27"		4.28 ± 0.10	2.30 ± 0.48	1.13±0.23
16	35°16'13.88"	96°44'43.33"		4.40±0.11	4.34±0.33	2.40±0.19
Estuary						
17	32°48'0.00"	122°14'44.99"		54.9 ± 0.7	21.1±0.8	$0.808 {\pm} 0.029$

Table 1 Analytical results of ¹²⁷I and ¹²⁹I and ¹²⁹I/¹²⁷I of water samples collected from rivers, lake and springs in the Qinghai-Tibetregion, Yangtze Estuary and South China Sea.

18	31°25'43.97"	122°30'7.67"		46.1±0.7	17.7±0.5	0.810 ± 0.023
Seawater						
19	18°00'14.04"	116°00'23.4"	0	55.9 ± 0.8	12.1±0.6	$0.457 {\pm} 0.022$
20	18°00'14.04"	116°00'23.4"	75	57.7±0.9	14.1 ± 0.4	$0.516 {\pm} 0.016$
21	18°00'14.04"	116°00'23.4"	100	58.5 ± 0.9	12.0±0.3	$0.432 {\pm} 0.010$
22	18°00'14.04"	116°00'23.4"	200	56.6±0.7	11.1±0.3	0.414 ± 0.013
23	18°00'14.04"	116°00'23.4"	400	61.6±0.9	15.4±1.3	$0.527 {\pm} 0.046$
24	18°00'14.04"	116°00'23.4"	1000	64.2 ± 0.9	13.5±0.6	$0.443 {\pm} 0.019$
25	18°00'14.04"	116°00'23.4"	1500	68.6 ± 0.9	7.15±0.25	$0.220{\pm}0.008$
26	18°00'14.04"	116°00'23.4"	2000	68.6±1.0	1.20 ± 0.14	$0.037{\pm}0.005$
27	18°00'14.04"	116°00'23.4"	3000	65.8 ± 0.9	3.16±0.16	$0.101 {\pm} 0.006$
28	18°00'14.04"	116°00'23.4"	3800	66.3±1.0	0.992 ± 0.110	$0.0315{\pm}0.0034$

1 Figure 1



Figure 2



6 Figure 3





9 Figure 4











Figure 7

