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Atmospheric iodine (^{127}I and ^{129}I) record in spruce tree rings in the northeast Qinghai-Tibet Plateau

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ABSTRACT: Atmospheric iodine isotopes have significant impacts on climate change and human health. However, the sources, transport pathway, and transfer process of atmospheric iodine are still not well understood. Tree rings of spruce collected from the east edge of Qinghai-Tibet Plateau were analyzed for iodine isotopes (^{127}I and ^{129}I). The results show that the levels and temporal variation of atmospheric iodine were well recorded in the spruce tree rings, and iodine concentrations in tree rings increased three times from 1960 to 2015, reflecting the increased releases of iodine to the atmosphere in the past decades due to the human activities. The anthropogenic ^{129}I in the tree rings presents the record of the human nuclear activities in the past 55 years. The sources and the transport pathways of radioactive substances could be extracted from the ^{129}I recorded in the tree rings in Qinghai-Tibet region. They are fallout of the global nuclear weapons tests in 1961-1962, releases of the Chinese atmospheric nuclear weapons tests in 1964-1980 transported through the tropospheric northwest wind, the releases of the Chernobyl accident dispersed through Westerlies and the continuous air releases before 1997 and the re-emission of marine discharges from the European nuclear fuel reprocessing plants transported through Westerlies.

Key words: Atmospheric iodine, Iodine-129, source term, transport pathway, Spruce, Tree rings

INTRODUCTION

Iodine is an essential trace element of mammals. Iodine deficiency can result in irreversibly neurological damage and developmental retardation in newborns. The major source of iodine in terrestrial environment is from atmospheric deposition.[1] The atmospheric iodine has a significant impact on climate through tropospheric ozone depletion and aerosol formation, and was

observed several times to be changed in recent decades.[1,2,3] Although some investigations have been carried out by measuring iodine in soil, sediment and ice cores,[3,4] the sources, the transport pathway and transfer process of atmospheric iodine in the ecosystem are still not well understood due to its multi-sources and the difficulties on discrimination of the different sources.

^{129}I, a long-lived radioisotope of iodine ($T_{1/2}$=15.7 Ma), has been released to the environment by human nuclear activities, including nuclear fuel reprocessing plants (NFRPs), nuclear weapons tests (NWTs) and nuclear accidents (NA). The anthropogenic ^{129}I (NFRPs: 7400 kg; NWTs: 150 kg; NAs: 7.2 kg) has highly overwhelmed the natural inventory in surface environment (250 kg, with a pre-nuclear $^{129}I/^{127}I$ atomic ratio of 1.5×10^{-12}).[5,6] Due to unique source of anthropogenic ^{129}I, volatile property and biophilic characteristics of iodine, the anthropogenic ^{129}I has been widely applied as an environmental tracer for investigation of regional radioactive sources and the related atmospheric transport pathways by determination of ^{129}I preserved in time-serial samples (sediment, ice core and coral samples).[6-12] However, due to the insufficient resolutions of sediment samples caused by low deposition rates in many locations, the sampling difficulties and specified locations of ice core and coral samples, they are not sufficient for the investigation in large areas, especially in mid-low latitude terrestrial environment.

Tree ring is an excellent time-serial material to monitor past climate changes as well as anthropogenic activities due to its wide distribution, easy access, annual resolution, accurate chronology and environmental sensitivity,[13] which might preserve the environmental radioactivity information both in pre-nuclear and nuclear age and in different locations. Dendrochemistry based on stable isotopic composition and elements concentrations in annual ring as retrospective proxy data has been successfully used to investigate the past environmental

change, such as past temperature and precipitation ($\delta^{18}O$, $\delta^{13}C$, etc.),[13] and anthropogenic release of toxic elements to atmosphere (Pb, Cd, Hg, etc.).[14] A few tentative works on regional radioactive deposition (including sources and transport) have also been reported by determination the concentration of radionuclides, such as ^{40}K, ^{137}Cs and ^{134}Cs.[15] Two investigations on ^{129}I in tree ring samples collected near the West Valley NFRP in USA and the Kalsruhe NFRP in Germany were reported.[16,17] The variation trend of ^{129}I levels in the ring-porous tree (elm, oak and locast), in which vessels are localized in the outmost growth ring, followed the regional deposition pattern of ^{129}I released from local NFRPs.[16] While, the tree rings of semidiffuse to diffuse-porous wood (cherry and maple), where vessels are disseminated throughout the trunk, showed a less matched ^{129}I pattern to the local NFRP releases.[16] ´This indicates a critical function of vessels in the iodine cross-ring translocation. However, high values of $^{129}I/^{127}I$ atomic ratio up to 3.6×10^{-9} were also measured in pre-nuclear rings from ring-porous tree (elm), indicating a cross-ring translocation of iodine occurred too.[16] It is probably due to the negative hydrostatic pressure in vessels compared to that in adjacent parenchymal cells and tracheid cells.[18] Therefore, coniferous tree might be more suitable for less cross-ring translocation of iodine due to no vessels in this species of tree.

The Qinghai-Tibet plateau (QTP) has an average altitude of 4000 m and cold ecosystem with widely distributed permafrost,[19] it is therefore quite sensitive to environment changes. Meanwhile, this region is also important for the climate and ecosystem of a large area in the world because it acts as the largest fresh water reservoir in Asia including over 2×10^{11} m^3 of glacier resource, 3×10^{10} m^3 of annual runoff and 1800 lakes. In 1964 -1980, 22 atmospheric nuclear tests with the total yield of 22.72 Mt of TNT were conducted at Lop Nor NWTs site in Northwest China.[20] Due to the prevailing Westerlies at Lop Nor, the released ^{129}I from the

atmospheric nuclear weapons tests at this site might be transported to the QTP which is located in the 1100 km downwind area of Lop Nor.

This work aims to investigate the sources, transport pathway and transfer of atmospheric iodine in the QTP by determining ^{127}I and ^{129}I in tree rings of spruce. The feasibility using tree rings of spruce to record the temporal variation of iodine isotopes in the atmosphere will be investigated in order to obtain a historic record of ^{129}I and ^{127}I levels in the atmosphere; this is useful for reconstruction of the levels of radioactive fallout and providing fundamental database for regional environmental change research.

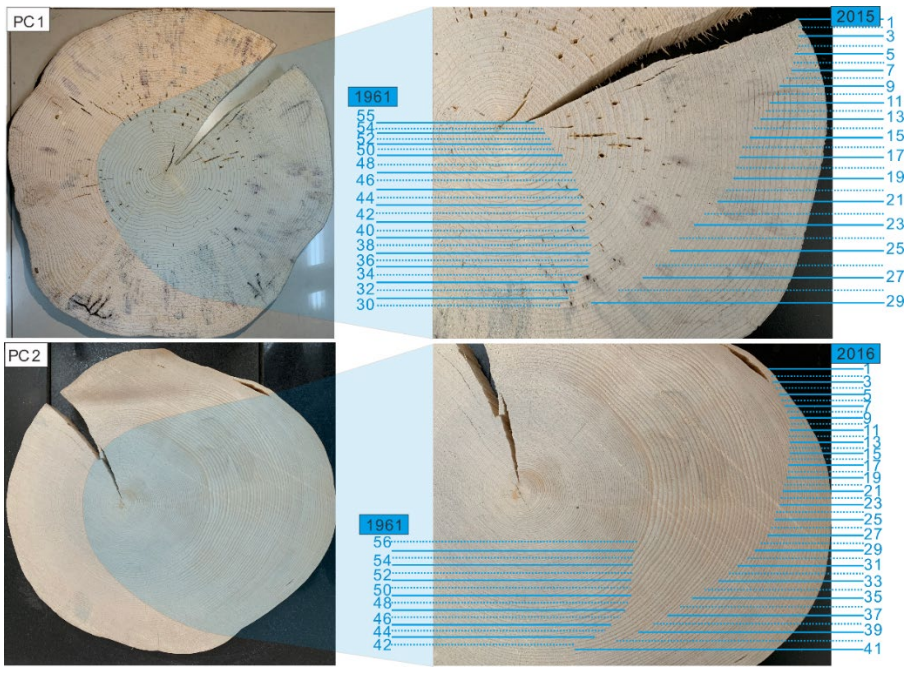
MATERIALS AND METHODS

Materials and dendrochronology. Two tree disks of Qinghai spruce PC1 and PC2 were collected from Qunjia (36º 16′ 44.04″ N, 101º 40′ 5.76″ E) and Maixiu National Park (35º 16′ 26.46″ N, 101º 55′ 1.26″ E) in Qinghai province, China, respectively. The sampling sites are located in the northeast edge of the QTP with average altitude over 3000 m and typical plateau continental climate. The Westerly is the dominant wind in this region.[19]

Qinghai spruce (*Picea crassifolia* Kom.), a coniferous species, is the dominant forest species in this area. Due to banned logging in the virgin forests in China in the past two decades, only the trees that naturally fell down can be sampled. In June 2017, one disk (PC1) was sampled from a spruce tree, which fell down in Dec 2015 in the foot of a slope of 10-20º with low canopy density. Another disk (PC2) was sampled from a spruce, which fell down in Jan 2017 in the middle of a slope of 20º with moderate canopy density. The sampling site of PC2 is located 110

103 km southeast of PC1. A disk of about 5 cm thickness was taken from the trunk of each tree at a
104 height of 30 cm above the ground and transported to the laboratory for analysis.

105 The tree disks were naturally air dried in a cool and ventilated room in Xi'an, China. The
106 surface of the wood disks was polished with sandy papers to easily identify the annual ring by
107 different colors between early and late wood (Fig. 1). No visible multiple rings with abnormal
108 thin width or missing rings with discontinuous boundary were observed in the transection of both
109 disks. Therefore, they were dated by counting rings from outside according to date of fall. The
110 age span is from 1961 to 2015 in PC1, and 1961 to 2016 in PC2 (Fig. 1). An uncertainty of ±1
111 year for each tree disk was estimated due to the possibility of visual inspection error. The center
112 part of the disk could not be used due to limited massive ring with super thin width. The clearest
113 part with arrayed rings was selected to dissect the annual ring from the outmost layer (Fig. 1).
114 Each dissected sample was dried at 60°C until constant weight, and then cut into small pieces.



115

116 Fig. 1 Wood disks of PC1 tree (up) and PC2 tree (bottom), showing the annual rings

Analytical Methods. Iodine in the dried tree ring samples was separated using an oxidative combustion method modified from Hou and Wang.[21] 5 g of dried sample was combusted in a tube furnace (Raddec LTD, Southampton, UK) with a stepwise temperature increasing protocol as shown in Fig S1. Due to the ignition point of about 270 °C in the major part of this type of wood, a slow increase of temperature during 230-350 °C was used to avoid production of a large amount of combustion gases (CO_2, CO, NO_x) in a short time. A second carbonization was implemented by increasing the temperature from 350 to 550 °C in a ramp speed of 2.5 °C /min to combust the remained residues (including rosin with ignition point of 480-500 °C). Afterwards, the temperature was increased to 800 °C and remained for 3 hours to completely decompose the sample. Iodine released during combustion was collected in a bubbler with 30 ml trap solution of 0.4 mol/l NaOH-0.05 mol/l $NaHSO_3$. The detailed method was presented in the supporting information. 1.0 ml of iodine trap solution was diluted with deionized water for measurement of ^{127}I using ICP-MS (Agilent 8800 ICP-MS). Cs^+ was used as internal standard, 0.15 mol/L $NH_3 \cdot H_2O$ was used as rinse solution. The detection limit of this method for ^{127}I was estimated to be 0.012 ng/ml. Due to large dilution (more than 30 times) of iodine trap solution, the iodine concentrations in some samples from PC2 are close to the detection limit (0.006 μg/g), causing a large measurement uncertainty (> 50%). Therefore, data of these samples are not included for further discussion. After addition of stable iodine carrier (prepared from iodine crystal purchased from Woodward company with $^{129}I/^{127}I$ atomic ratio $<2\times10^{-14}$) into the remaining trap solution, iodine (iodide) was precipitated as AgI for determination of ^{129}I by a 3 MV Tandem AMS system (HVEE) at Xi'an AMS Center, China. The details of AMS system and measurement method for ^{129}I have also been reported elsewhere,[22] and briefly described in Supporting Information. $^{129}I/^{127}I$ ratios in the procedure blanks prepared using the same procedure as the samples were

140 measured to be below 2×10^{-13}, which are at least one order of magnitudes lower than that of
141 samples.

142

143 ## RESULTS AND DISCUSSION

144 *^{127}I and ^{129}I level in the tree rings.* The measured ^{127}I concentrations are 0.01-0.10 μg/g with the
145 most data fall into the range of 0.02-0.05 μg/g in tree rings of PC1, and 0.008-0.025 μg/g with
146 the most fall into the range of 0.01-0.02 μg/g in tree rings of PC2 (Fig. 2 a). The higher iodine
147 concentration from PC1 might be attributed to its sampling location, at forest edge with
148 sufficient water, sunlight and fresh air, confirmed by its larger ring width than that of PC2. The
149 iodine concentrations in the two spruce trees are much lower than the reported levels in
150 deciduous trees, such as 0.1-0.6 μg/g in Black locust collected from Karlsruhe, Germany in
151 1954-1983, 0.7-1.2 μg/g in Elm from New York State, USA in 1950-1986 and 0.04-0.09 μg/g in
152 Red Oak from New York State, USA in 1961-1987 (Fig. 2 a).[16,17] This probably caused by low
153 uptake capacity of iodine from environment in this region with low temperature and associated
154 low growth rate.

155 Significant variations of ^{129}I concentrations were observed in two tree rings, i.e. (0.7-3.5) $\times10^6$
156 atoms/g in PC1 and (0.5-3) $\times10^6$ atoms/g in PC2 (Fig. S3, Table S1). The over fivefold
157 difference in 55-56 years might be attributed to the varied sources and concentrations of ^{129}I in
158 the environment. In addition, the variation might also result from the varied accumulation/uptake
159 rate of iodine into tree rings in different periods. To overcome this problem, the $^{129}I/^{127}I$ atomic
160 ratios were applied to represent the level of ^{129}I in the tree rings.

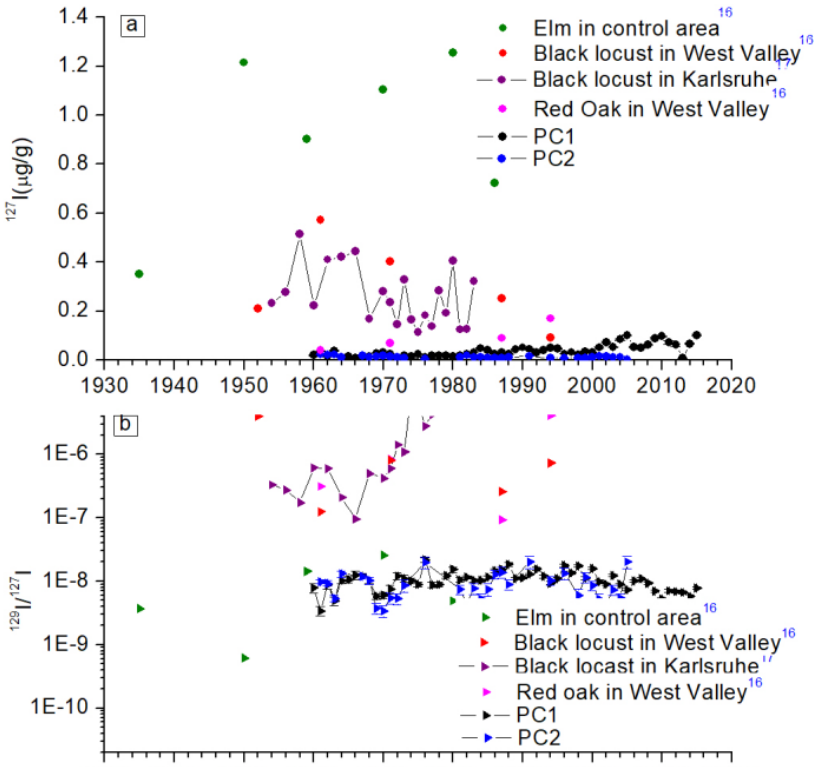


Fig. 2 Comparison of measured ^{127}I concentrations (a) and ^{129}I/^{127}I ratios (b) in the tree rings in this work with the reported values in other locations[16,17, 23, 24]

The ^{129}I/^{127}I atomic ratios of (6-15)×10^{-9} were obtained in the tree ring of PC1 and (4-20)×10^{-9} in PC2 (Fig. 2 b), which are more than two orders of magnitude higher than the pre-nuclear level of ^{129}I/^{127}I ratios (1.5×10^{-12} for marine system and 2.0×10^{-11} in terrestrial environment),[5,25] indicating anthropogenic sources of ^{129}I. However, this level is at least two orders of magnitude lower than the values reported outside the nuclear fuel reprocessing sites in Germany (10^{-7}-10^{-5}) and USA (10^{-7}-10^{-6}), but similar to those observed in the areas without direct influenced by nuclear activities (10^{-9}-10^{-8}) (Fig. 2 b).[16,17] The average ratio of ^{129}I/^{127}I in the two rings in 2009-2015 is around 6×10^{-9}, in the same magnitude with the values measured in herbs (4.5×10^{-9}) collected in 2015[23] and surface soils (3×10^{-9}) collected in 2013[24] in north China, representing the level of ^{129}I in this region.

Iodine uptake and integration in the tree rings from atmosphere. The measured $^{129}I/^{127}I$ ratios (average of 6.6×10^{-9}) during 2009-2015 in the rings of PC1 tree are more than two times higher than the reported values in surface soil (average: 3×10^{-9}, collected in 2009-2013) and river water (1.1×10^{-9}, collected in 2016-2017) in nearby locations (Fig. 2).[24,26] During 1963-1977, the ratios (average of 9.5×10^{-9}) in the rings of both trees are similar to the reported $^{129}I/^{127}I$ ratios (average of 9×10^{-9}) in the atmospheric fallout in Tokyo (35 ºN) before the operating of Tokai NFRPs (Fig. 4a).[27] No ^{129}I data in atmospheric sample from the study region was reported. Since the ^{129}I level in atmosphere in the study region (36 ºN) should be similar with that in the same latitude region as Tokyo, the result might indicate that iodine uptake from atmosphere is the dominant way entering into the tree rings compared to that from soil system.

The Qinghai spruce uptakes inorganic elements and water from soil through root, and translocates it to other parts through the pits on the trachieds cells wall relying on transpiration stream (Fig. S2). This slow process could induce a time lag of months to years.[28] Meanwhile, spruce can also uptake the nutrient from atmosphere through foliar absorption, and phloem transport, then to inner xylem through radially aligned ray parenchyma cells (the only radial transport tissue of tree trunk), which is sustainable by photosynthesis energy without much time lag.[29]

Iodine in soil mainly originates from the atmospheric deposition, decomposition of dead plants and animals (absorbed iodine from atmosphere or soil), and weathering of parent material of soil.[30] The anthropogenic ^{129}I presents mainly in the upper 50 cm of soil profiles in the background region (far away from any nuclear facilities),[30] and the natural $^{129}I/^{127}I$ ratios in the deep soil ($<10^{-11}$) are 2-3 orders of magnitude lower than the present environmental level (e.g. atmosphere, surface soil, etc.).[25] Considering the deep root of spruce (>>50 cm), lower $^{129}I/^{127}I$

ratios observed in the surface soil in this region compared to that in the tree ring, as well as the similar $^{129}I/^{127}I$ ratios of the tree rings with that in atmospheric fallout in Japan, iodine in the investigated tree rings should be dominantly taken from atmosphere. The similar pattern of higher $^{129}I/^{127}I$ ratios in precipitation (3.8×10^{-9}) than that in surface soil (1.9×10^{-9}) was also reported in Xi'an.[31] Due to the non-essential element for trees, iodine was also demonstrated to be hardly translocated to the aboveground compartments from the tree root system.[29]

In atmosphere, iodine presents mainly in gaseous form and partly associated with aerosol.[32,33] Gaseous iodine can be trapped directly through stomata in the tree leaves. Aerosol deposited on tree leaves might be adhered on the surface through wax on the spruce needles, and the associated iodine can be leached out by water vapor. Then soluble iodine can be taken up to the tree leaves through hydrophilic holes on leaf's surface and/or active absorption by plasmodesmata.[34] The absorbed iodine is translocated through phloem, ray parenchyma cells and then fixed in xylem. The Fukushima derived ^{129}I was observed to be well recorded in *Japanese cedar* (coniferous species) through atmosphere absorption.[35] Much higher ^{129}I concentration was measured in litter than in surrounding soil collected in nearby area of a NFRP, implying a more direct uptake way of iodine in tree/vegetation from atmosphere by foliage.[17]

The translocation pathway of the atmospheric uptake iodine into the xylem relies on the only radial tissue (ray parenchyma cells) and the pits on longitudinal tracheid cell walls (Fig. S2). The ray parenchyma cells, is alive only in sapwood (usually the out most 10-12 rings) but dead in heartwood.[36] A considerable amount of radial moving of water solution in alive ray parenchyma cells was detected based on fluorescein dyes.[18] However the migration depth was limited only in the outmost ring of mature xylem from the anatomical observations by scanning electron microscope.[18] A similar low crossing rings movement of water soluble fluorine (similar

chemistry with iodine) was also reported in Norway spruce.[37] The pits in longitudinal tracheid cells are usually less developed, both the size and quantity, from late wood than that from early wood, which will primarily limit iodine moving across one single growth ring. The water transport by tracheid cells was only observed in early wood, but not in late wood based on single growing spruce ring.[37] Combined with the fluctuated variation of the observed $^{129}I/^{127}I$ ratios over rings in both trees (Figure. 3a and b) instead of monotonous gradient distributions, it indicates that the crossing rings translocation of iodine hardly happens in the selected Qinghai spruce.

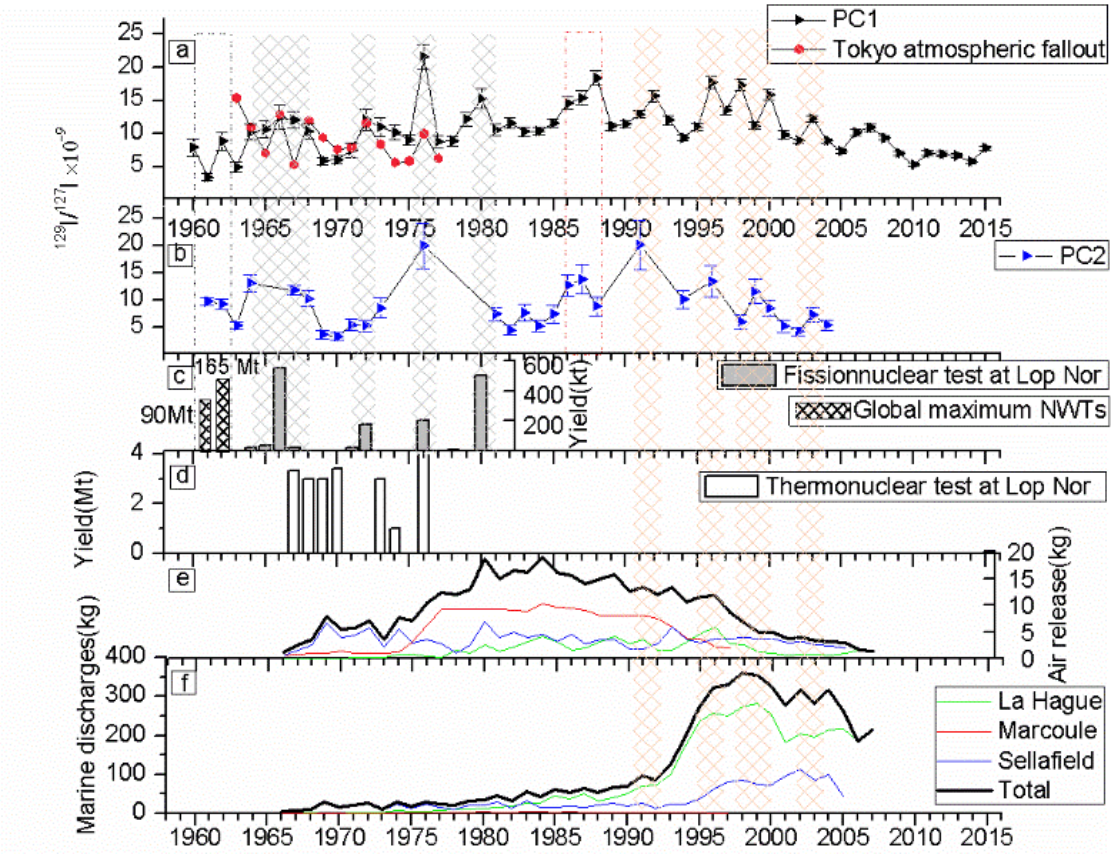
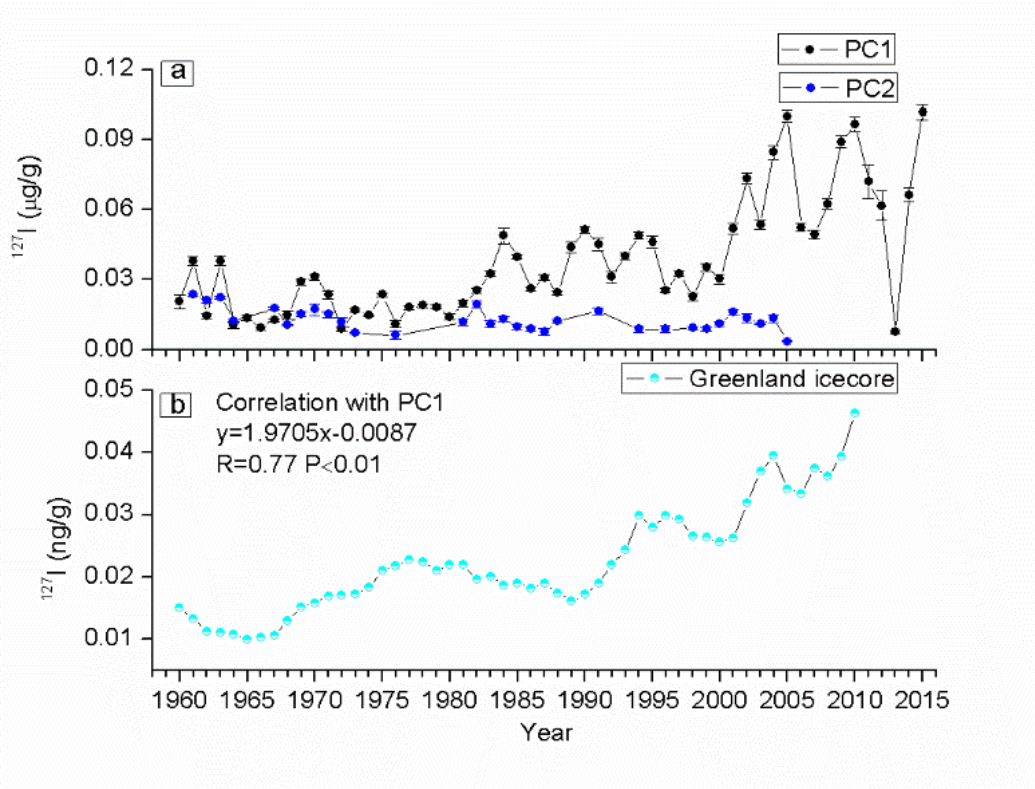


Fig. 3 Temporal variation of the measured $^{129}I/^{127}I$ ratios in the tree rings from two sites in the QTP (a), in comparison with the reported variation of $^{129}I/^{127}I$ in atmospheric fallout from Tokyo (b) [27], yield of fission NWTs (c), yield of thermonuclear weapons tests at Lop Nor

(d) [20], air releases of ^{129}I from major NFRPs (e) and marine discharges of ^{129}I from NFRPs (f).[33]

Historic records of atmospheric iodine level. ^{127}I concentrations in the tree rings of PC1 (Fig. 4a) were 0.010-0.033 μg/g with an average of 0.020 μg/g in 1961-1983, and increase to 0.023-0.051 μg/g with an average of 0.039 μg/g in 1984-2001, then to 0.049-0.100 with an average of 0.069 μg/g in 2001-2016. A threefold increase was observed, but no such variation in PC2 (Fig. 4a). This increased iodine concentrations might reflect the increased iodine concentrations in atmosphere based on the inferred atmospheric absorption pathway. A similar three folds elevation of atmospheric iodine has been observed in Alpine ice core,[4] and Greenland ice core [3] from 1950-2000s. Significant correlation of ^{127}I concentrations in the tree rings with that in Greenland ice core[3] (directly deposition from atmosphere) ($R=0.77$, $P<0.01$) are observed (Fig. 4b). However, no such kind of correlation was found in PC2. The lack of partial data in PC2 or/and the sampling site located in the middle of forest with higher canopy density than that in PC1 and lower environmental sensitivity might attribute to this phenomenon.[38] Although the uptake capacity of iodine through the foliar absorbing way can be affected by water condition, sunlight, temperature, etc., a less variation of temperature (<1°C of the annual average temperature), fluctuating variation of annual precipitation (from the nearest Xining weather station), and almost steady sunlight condition were reported in the past 50-60 years in the studied time scale,[39] excluded effect of these parameters on the uptake of iodine into the tree region in this location.



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253 Fig. 4 Variation of iodine (^{127}I) concentration in PC1 and PC2 tree (a), in comparison with the
254 temporal variation of iodine concentrations in ice core in Greenland[3]

255 Iodine in atmosphere might originate from oceanic emissions of gaseous iodine (through
256 biological, chemical and photochemical process), terrestrial emissions of gaseous iodine from
257 salt marches, rice fields, peatland and forest system, and combustion of fossil fuel and biomass,
258 of which the oceanic emission dominates.[1,3,4] The increased atmospheric iodine level might be
259 attributed to the increased releases from these processes. The oceanic emission of molecular
260 iodine (I_2) was observed to increase with the tropospheric ozone (O_3) content,[1] which increased
261 by a factor of two from the 1950s to early 1990s due to the elevated global combustion of fossil
262 fuel.[40,41] The oceanic emission iodine was reported to be enhanced by the sub-ice phytoplankton
263 production that associated with the recent thinning of sea ice.[3] Meanwhile, the increased

consumption of fossil fuels (from 1000 TWh/y in 1980s to 3200 TWh/y in 2000s) in the past decades could also directly contribute to the increased iodine concentration in the atmosphere, which was estimated to be 5000 tons/y in 2005.[42, 43]

Records of human nuclear activities in the tree rings. The distribution of $^{129}I/^{127}I$ atomic ratios (Fig. 3a and b) in the two tree rings show similar variation trend, e.g. the $^{129}I/^{127}I$ peaks in 1962, 1964-1968, 1976, 1996 and 2003 occurred in both tree rings, as well as the peaks in 1972/1973, (1986-1988)/(1986-1987), 1992/1991, 1998 and 2000/1999 occurred in PC1/PC2 respectively. The one-year discrepancy between the records in two tree rings could be caused by the uncertainty of dating (±1) from counting rings. In addition to that, due to the lack of data in 1980 in PC2 rings compared to an obvious peak in 1980 from PC1 rings, and a separate high values period of 2006-2008 in PC1 rings, might be attributed to the different sampling locations and/or uncertainty of dating results (±1 year) from counting rings.

The similar peaks in the two tree rings should be attributed to the peak values of ^{129}I occurred in atmosphere during these years, resulting from the anthropogenic releases. Fig. 3 shows the variation of $^{129}I/^{127}I$ ratios in two tree rings in comparison with that in atmospheric fallout records in Tokyo (Fig. 3a and b), the time distribution of explosion yields of atmospheric NWTs (Fig. 3c and d), and the annual discharges (air and marine) of ^{129}I from the three European NFRPs which accounts for the most of ^{129}I inventory in current environment (Fig. 3e and f).

Atmospheric nuclear weapons tests. Several peaks of $^{129}I/^{127}I$ atomic ratios in 1962, 1964-1968, 1972, 1976 and 1980 were observed in the tree rings of PC1, and in 1961-1962, 1964-1968 and 1976 in the rings of PC2. The atmospheric nuclear weapon tests are the only source of ^{129}I during this period since the release amount from NFRPs were negligible.[20]

286 The twofold increased $^{129}I/^{127}I$ ratios (8.8×10^{-9}) in 1962 compared to the value of (3-4) $\times10^{-9}$
287 in 1961 and 1963 was observed in tree rings of PC1, and 9×10^{9} in 1961-1962 compared to 5×10^{9}
288 in 1963 in PC2 rings, corresponding with the date of the largest numbers of atmospheric NWTs
289 in 1961-1962. The observed value in the tree rings of 1961-1962 (average: 9×10^{9}) agrees with
290 the value recorded in atmospheric fallout samples in 1963 from Tokyo (15.5×10^{9}).[27] This signal
291 was widely detected by peaks of $^{129}I/^{127}I$ ratio, ^{137}Cs and $^{239,240}Pu$ activities in north China.[7,44]
292 The 1-2 years lag among these records could be attributed to the hysteresis effect of samples
293 or/and the dating uncertainty (±1 year).

294 A broad peak of $^{129}I/^{127}I$ ratios corresponding to 1964-1968 was observed in both tree rings,
295 which matches well with several high fission yield tests at Lop Nor (Fig. 3c), such as the first
296 one in 1964 (20 kt) and the second one in 1965 (40 kt). The highest value of 12×10^{9} in 1966 in
297 PC1 rings agrees well with the boosted fission tests carried in 1966 with fission yield of 250 kt of
298 TNT, and the similar high $^{129}I/^{127}I$ ratio (12×10^{9}) was also recorded in atmospheric fallout
299 samples in 1966 collected from Tokyo.[27] The constant high ^{129}I level in both trees in 1967-1968,
300 as well as the high value in the atmospheric fallout in Tokyo in 1968, should be attributed to a
301 test of fission weapon in 1967 at Lop Nor and the first two Chinese thermonuclear weapons tests
302 in 1967 and 1968, where a detonator with high ^{235}U fuel load might be applied, causing releases
303 of fission products including ^{129}I.

304 The radioactive substances from atmospheric NWTs were injected into different height of
305 atmosphere depending on the total explosion yield, the height of burst, and the meteorological
306 condition after the detonation.[20] The radioactive substances from fission weapons with relative
307 lower yields and some part from thermonuclear weapons were mainly injected into troposphere
308 to disperse regionally.[20] The sampling sites in northeast QTP are located about 1100 km

southeast of Lop Nor. The dominant Westerlies in the troposphere in the Lop Nor region can transport these radioactive substances (including ^{129}I) to the sampling sites in several days and produce $^{129}I/^{127}I$ ratio peaks in the tree rings. The simulated forward trajectories of air masses by the HYSPLIT model (Fig. S4) confirmed that the air masses in the troposphere at the Lop Nor after the tests on 16th Oct. 1964, 14th May 1965, 28th Dec. 1966, 17th Jul. 1967, and 28th Dec. 1968 reached to the sampling region in a few days.[45]

One $^{129}I/^{127}I$ ratio peak, observed in 1972 in PC1 rings, is in accord with the high fission yield of 170 kt of TNT from a fizzled thermonuclear test conducted on 18th Mar. 1972 at Lop Nor. No significant peak of $^{129}I/^{127}I$ ratio was observed in 1972 but a high value in 1973 (8.5×10^{-9}) in PC2, which could be due to the ±1 year's uncertainty of dating. A similar peak was also measured in atmospheric fallout samples in 1972 collected from Tokyo (11.5×10^{9}) (Fig. 3a).[27] The highest value of $^{129}I/^{127}I$ ratios of 20×10^{-9} was observed in 1976 in both tree rings that matches very well with the date of the fizzled thermonuclear test with a high fission yield of 200 kt of TNT on 26th Sep. 1976 at Lop Nor, as well as a similar peak in atmospheric fallout in Tokyo.[27] A peak value of $^{129}I/^{127}I$ ratios (15.3×10^{9}) was observed in 1980 in PC1 rings in consistent with the time of the last atmospheric test at Lop Nor. The forward trajectory analysis of air mass also confirmed the stratospheric air masses at the Lop Nor after the tests on 18th Mar. 1972, 26th Sep. 1976, and 16th Oct. 1980 arrived in the sampling region in a few days (Fig. S4)

No significantly high ^{129}I signals were observed after 5 high yield thermonuclear tests conducted on 29th Sep. 1969 (3 Mt), 14th Oct. 1970 (3.4 Mt), 27th Jun. 1973 (3 Mt), 17th Jun. 1974 (1 Mt) and 17th Nov. 1976 (4 Mt) (Fig. 3d), reflecting less ^{129}I release due to the less fission fuel loaded as detonator.

Releases from nuclear accidents**.** A peak of $^{129}I/^{127}I$ ratios covers 3 years in 1986-1988 ((14.6-18.4) $\times 10^9$) in PC1, and 2 years in 1986-1987 ((12.7-13.8) $\times 10^9$) in PC2 were observed. Since atmospheric NWTs have already been ceased from 1980, and the almost steady discharges of ^{129}I from the main NFRPs (air: 13-14 kg/y; marine: 70 kg/y), this might be attributed to the Chernobyl accident on 26th April 1986 with 1.3-6 kg ^{129}I released to the environment.[33] The Chernobyl accident derived radioactive fallout had dispersed to north China through the Westerlies, and the level decreased from northwest towards east and south.[46] The total β activity in atmospheric fallout collected in Urumchi (1400 km in the northwest of sampling sites, 43º N), and the short lived radioactive ^{131}I activity in milk was observed to elevate over 50 times compared to the background value.[46,47] A 15-times elevation of the total β activity was also measured in atmospheric fallout in Xi'an (600 km in the east of sampling sites, 33º N).[47] Although no direct monitor data of radioactivity after the Chernobyl accident is available in the sampling site, it should receive the Chernobyl derived ^{129}I because the sampling site is located between Xi'an and Urumchi. The Chernobyl derived $^{129}I/^{127}I$ peak was even observed in sediment samples from Jiaozhou Bay in the similar latitude but 1600 km east to the study area.[7] A broad ^{129}I peaks in both tree ring profiles (PC1: 3 years; PC2: 2 years) might be contributed to the re-emission of the deposited ^{129}I in the high contaminated soil to atmosphere in the following 2-3 years.

No significant increased $^{129}I/^{127}I$ value was observed in 2011 (7.1×10^9) compared to 6.9×10^9 in 2012 and 5.2×10^9 in 2010 in PC1 rings, indicting an insignificant contribution from the Fukushima accident with about 1.2 kg ^{129}I releases (including 0.35 kg direct marine discharge).[7] The atmospheric releases from Fukushima accident mainly dispersed eastwards and deposited in the North Pacific Ocean, and the marine discharges including ^{129}I were mainly transported

354 eastwards by North Pacific Current.[48] Although some radionuclides (mainly ^{131}I, ^{137}Cs and ^{134}Cs)
355 in aerosol samples collected in North China after Fukushima were measurable, but the level is
356 much lower compared to those in North America and Europe, and at least two orders of
357 magnitudes lower than that from Chernobyl.[49,50,51] Therefore, the contribution to the study area is
358 negligible.

359 Releases from nuclear fuel reprocessing plants**.** A few peaks of $^{129}I/^{127}I$ ratios were observed
360 since 1991 in the rings of both trees, i.e. 1992 (PC1)/1991 (PC2), 1996 (PC1 and PC2), 1998 and
361 2000 (PC1)/1999 (PC2) and 2003 (PC1 and PC2) (Fig. 3a and b). Since the atmospheric NWTs
362 ceased in 1980, no reported nearby nuclear facilities and nuclear accidents can release ^{129}I to the
363 sampling region. The most possible source of ^{129}I during this period should be the releases from
364 NFRPs, which dominate the ^{129}I inventory in the present environment.[33] Although the
365 reprocessing plants have been in operation in USA, Russia, Japan, China, Germany, France and
366 UK, the European reprocessing plants in Marcoule (1959-1997), La Hague (1966-), and
367 Sellafield (1951-) are the most important ones due to their high capacities and large scales in
368 reprocessing operation. The contribution from two Chinese reprocessing plants in Guangyuan
369 (1975-1991) and Jiuquan (1968-1984) was insignificant due to the small capacity and ^{129}I release
370 compared to the European ones.[20] The investigation of ^{129}I distribution in surface soil from a
371 large area of China did not show any unexpected increase level, except the local region of these
372 two NFRPs (<100 km).[52] The contribution from Tokai NFRPs (1977-2005) in Japan should be
373 negligible because of the small volume of ^{129}I release (1 kg), the far distance, and non-major
374 direction of wind.[20]

375 The total atmospheric release of ^{129}I from the three European NFRPs gradually increased to
376 10 kg/y in 1976 from lower than 5 kg /y in late 1960s, and then kept at a relative constant high

377 level of about 15 kg /y until 1996. Afterwards, it gradually decreased to a quite low value of 1.4
378 kg/y in 2007 due to the shutdown of NFRP at Marcoule in 1997 and the updated system in other
379 reprocessing plants for capture of gaseous radioiodine (Fig. 3e).[33] The high value in 1980 (18 kg)
380 compared to that in 1979 (13 kg) might add a small background value on the peak produced by
381 the last Chinese NWT. The marine discharges, mainly from the two European reprocessing
382 plants at La Hague (France) and Sellafield (UK), gradually increased to 65 kg/y in 1980s from
383 less than 40 kg/y before that, and significantly increased after 1990 and keep at a high value of
384 250 kg/y (Fig. 3f).

385 The ^{129}I peak observed in both tree rings in 1991/1992 matches well with the first increased
386 total marine discharges (to 100 kg/y in 1991 from 70 kg/y in 1990), when air discharges of 13 kg
387 were steady. The peak observed in 1996 matches with the most drastically elevated marine
388 discharges up to 320 kg/y from below 200 kg/y in 1995, under the almost steady air discharges
389 of (9-12) kg/y. The peaks observed in 1998-2000 in the two tree rings agree with the first peak of
390 marine discharges of 350 kg/y in 1998-1999, while the air discharges continuously reduced by
391 half (1997: 9 kg; 1999: 5 kg). The peak in 2003 matched well with the high value of 360 kg/y of
392 marine discharges in 2002 compared to the value of 280 kg/y in 2001 and 2003, under the almost
393 steady air discharges of 4 kg/y (Fig. 3e and f).

394 Some exceptional events were not recorded in tree rings, such as the elevation of total air
395 discharges from 16 kg/y in 1983 to 19 kg/y in 1984, and the three-time elevation of total marine
396 discharges from 100 kg in 1991/1992 to 325 kg in 1996. The small increment could be
397 responsible for the former one. The decreased air discharges from Marcoule NFRPs from 8 kg/y
398 in 1991/1992 to 2 kg/y in 1996 could explain the later one due to the closer latitude of 43 °N
399 compared to that of La Hague (49° N) and Sellafield (54° N).

The marine discharges of ^{129}I can be re-emitted to the atmosphere in gaseous form confirmed by the similar high $^{129}I/^{127}I$ ratios observed in aerosols and rainwater samples collected in the North Europe (Denmark, Germany and Sweden) and the contaminated seawater samples ($^{129}I/^{127}I$ = 10^{-8}-10^{-6}).[32,53] The re-emitted ^{129}I can enter into lower troposphere, and to higher altitude by intensive vertical motions, then further transport eastwards through the Westerlies (around 1500 m-12000 m, mainly in 35° N - 65°N) to East Asia. The European NFRPs derived ^{129}I signal has been observed in sediment,[6,7] precipitation,[54] and ground water[26] in Westerly area of north China (> 33º N). The ^{129}I was finally taken up by tree leaves and recorded as $^{129}I/^{127}I$ ratio peaks in both tree rings within 1 year in consideration of the long-distance transport.

Above all, the sources, transport pathway and transfer process of atmospheric iodine in the northeast part of the QTP were well traced. The terrestrial iodine from northwest upwind area (traced by ^{129}I released from NWTs at Lop Nor and from Chernobyl Accident), and marine iodine from the mid-high latitude of Atlantic Sea (traced by ^{129}I released from NFRPs at Sellafield, La Hague and Marcoule) can be transported to the studied area through the Westerlies, which contribute significantly for the iodine in atmosphere and then ecosystem.

SUPPORTING INFORMATION

The detailed chemical separation procedure, preparation and measurement of ^{127}I and ^{129}I, the tissues, the microscopic structures of selected wood species are presented; The analytical results of ^{129}I and ^{127}I concentration in two tree rings (Table S1), protocol of combustion for iodine separation from woof (Fig. S1), diagram of tree rings and microstructure of nonporous and porous tree (Fig. S2), temporal variations of ^{129}I concentrations in PC1 and PC2 (Fig. S3), and simulated forward trajectories of air mass after the major Chinese atmospheric nuclear weapons tests (Fig. S4) are included.

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Caption of the figures

Fig. 1 Wood disks of PC1 tree (up) and PC2 tree (bottom), showing the annual rings

Fig. 2 Comparison of measured 127I concentrations (a) and 129I/127I ratios (b) in the tree rings in this work with the reported values in other locations16,17, 23, 24

Fig. 3 Temporal variation of the measured $^{129}I/^{127}I$ ratios in the tree rings from two sites in the QTP (a), in comparison with the reported variation of $^{129}I/^{127}I$ in atmospheric fallout from Tokyo (b) [27], yield of fission NWTs (c), yield of thermonuclear weapons tests at Lop Nor (d) [20], air releases of ^{129}I from major NFRPs (e) and marine discharges of ^{129}I from NFRPs (f).[33]

Fig. 4 Variation of iodine (^{127}I) concentration in PC1 and PC2 tree (a), in comparison with the temporal variation of iodine concentrations in ice core in Greenland[3]