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Levels and behavior of environmental tritium in East Asia

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Abstract For a more systematic understanding of the levels of environmental tritium and its behavior in East Asia, a database on environmental tritium was established based on the literature published in the past 30 years. Subsequently, the levels and behavior of the environmental tritium were further studied by statistical analyses. The results indicate that the distribution of environmental tritium is inhomogeneous and complex. In areas without nuclear facilities, the level of environmental tritium has decreased to its background level, even though a certain number of atmospheric nuclear tests were performed before 1980. In general, the level of atmospheric tritium was marginally higher than the levels in precipitation and surface water; the levels in shallow groundwater and seawater were considerably lower. Furthermore, the levels of tritium in the atmosphere, precipitation, and inland surface water were strongly correlated with latitude and distance from the coastline. In soil and living organisms, the level of tissue-free water tritium (TFWT) was comparable to the tritium levels in local rainfall, whereas the persistence of organically bound tritium (OBT) in the majority of organisms resulted in an OBT/TFWT ratio greater than one. Conversely, extremely high levels of environmental tritium were observed near certain nuclear power plants and the Fukushima accident sites. These results highlight the requirement to know the tritium baseline level and its behavior in the environment beforehand to better assess the impact of tritium discharge. Further investigations of environmental tritium in East Asia using more efficient and adequate monitoring methods are also required.

Keywords Tritium · Background · Nuclear facility · Nuclear accident · East Asia

1 Introduction

Tritium is a rare isotope of hydrogen with one proton and two neutrons in its nucleus. It is the radioactive form of hydrogen and is typically represented by the symbol T or ^{3}H. Despite the difference in mass between tritium and hydrogen (H), they occur in the same physicochemical form and are ubiquitous in the environment.

Naturally occurring tritium is produced primarily by the nuclear reactions of cosmic neutrons with nitrogen-14 in the upper atmosphere; the annual tritium production on the Earth's atmospheric surface has been estimated to be 7.2×10^{16} Bq [1]. However, tritium decays during generation and the equilibrium inventory of natural tritium is estimated to be approximately 1.29×10^{18} Bq (approximately 3.6 kg) at present [2]. Several human activities including atmospheric nuclear tests, nuclear reactor operation, spent fuel reprocessing, tritium manufacturing, and controlled thermonuclear reaction tests have produced large amounts of tritium in the past decades. In particular, significant amounts of anthropogenic tritium (520–550 kg) were injected into the stratosphere by atmospheric nuclear weapon tests in the mid-last century [3]. Consequently, the level of environmental tritium significantly increased in the 1950s and 1960s [4]. With the implementation of the

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Treaty Banning Nuclear Weapon Tests in the Atmosphere, in Outer Space, and Under Water (simplified as Partial Test Ban Treaty, PTBT) in 1963, Nuclear Nonproliferation Treaty (NPT) in 1968, and Comprehensive Nuclear Test Ban Treaty (CNTBT) in 1996, the influence of tritium originating from nuclear weapon tests has steadily decreased. At present, the main anthropogenic source of tritium is considered to be the operation of nuclear power plants [5]. Boyer et al. estimated that the global generation rate of anthropogenic tritium from nuclear power plants in the past 30 years was approximately 0.08 kg a^{-1}, which is marginally less than its natural yield (approximately 0.15–0.2 kg a^{-1}) [6].

Tritium in the environment can be cycled through physical/chemical exchange and biotransformation. After natural production, the uniform mixing of bomb tritium and natural tritium in the stratosphere allows them to be injected into the tropopause through stratospheric–tropospheric mass exchange (STEM). Subsequently, elemental tritium is gradually converted into tritiated water (HTO) vapor through oxidation and isotope exchange. Through dry deposition and precipitation, atmospheric HTO vapor from the tropopause is transported to the surface land, mixed with HTO generated by anthropogenic pathways, and eventually participates in the global water cycle. Therefore, tritium has been recognized as an ideal tracer for studying the global water cycling process [7–10]. Through the natural water cycle, atmospheric tritium can be introduced into rivers, lakes, seas, soil, plants, and food chains. Moreover, tritium is a low-energy beta emitter (average energy of 5.7 keV with a maximum of 18.6 keV) with a half-life of 12.33 years [11]. When tritium is inhaled, ingested, or penetrates through the skin, sustaining internal radiation exposure to humans could pose a potential health risk. Several animal experiments have reported that severe tritium contamination can cause acute radiation sickness. The effects of low-dose tritium on the central nervous system of offspring mice and rats have also been reported [12–14]. Therefore, environmental tritium has attracted wide attention in radiation protection communities [15–17]. In this context, environmental tritium monitoring is not only critical for reviewing and checking the operation status of nuclear facilities but is also helpful for assessing their long-term environmental impacts. Over the past decades, numerous investigations on environmental tritium have been performed in different regions of the world for the purposes of radiation safety and/or hydrogeological research [18–21].

East Asia is one of the most prosperous and highly populated regions in the world [22]. With the rapid development of industrialization and urbanization in China, Japan, and South Korea, the significant dependence on energy supplies is self-evident. Currently, 111 nuclear reactors are being operated in East Asia, providing a net electrical capacity of 1.06×10^5 MW per year, accounting for more than 25% of the nuclear energy assembly in the world [23]. Another 22 nuclear reactors are under construction in East Asia (China: 16; Japan: 2; South Korea: 4) [23]. The operation of nuclear reactors and the reprocessing of nuclear fuels can result in an increase in tritium release. In addition, it has been reported that approximately 1.25 million tons of tritium-contaminated wastewater will soon be released from the Fukushima Daiichi nuclear power plant into the Pacific Ocean [24, 25]. Moreover, huge amounts of tritium inventory (1000 times greater than the release of conventional fission reactors) will be loaded into fusion facilities in the future, which could cause a higher anthropogenic tritium release [26]. Consequently, East Asia is considered a region with high tritium emission densities both now and in the future. On the other hand, the extensive latitudinal and longitudinal spans and significant altitude differences induce differences in the natural sources of tritium distributed in East Asia [27]. Furthermore, the remarkable climatic differences resulting from such geographical differences can exacerbate the spatial and temporal variances in natural tritium profiles [27]. Therefore, East Asia is regarded as an ideal region for studies on tritium behavior. Over the last three decades, significant efforts have been made to conduct environmental tritium monitoring in this region; however, regional levels remain unclear because a large amount of sporadic data has not yet been systematically compared and analyzed.

In this study, the reported data on environmental tritium in East Asia over the past 30 years were collected through literature retrieval, and a database including tritium contents in the atmosphere, precipitation, surface water, groundwater, soil, and living organisms was established for further analysis and discussion. The main objective of this study is to evaluate the tritium levels and particularities of environmental behavior in East Asia. The findings can be helpful for future environmental studies on developing appropriate monitoring strategies and objective evaluation of environmental radiation safety.

2 Data collecting and analysis

2.1 Literature retrieval

Several literature databases were used to collect the related information and data on environmental tritium in China, Japan, and South Korea. For papers published in English, Web of Science (Thompson Scientific, USA) was used. For papers published in Chinese, the China National Knowledge Infrastructure (CNKI), WANFANG database, and China Science and Technology Journal Database

(CQVIP) were used. Google Scholar was used to retrieve papers written in Japanese and Korean. By limiting the sampling sites to within China, Japan, and South Korea, and sampling time or period to the 1990s to 2020s, approximately 112 papers were extracted for use in this study.

2.2 Database construction

A database containing the sampling site, sampling time, sampling medium, chemical forms of tritium, and their measured results was constructed in this study. The data entry was performed independently by two persons to reduce input errors. For the data on tritium in environmental media, if the papers provided detailed information on the sampling site, sampling time, and corresponding results of the environmental tritium, the data were extracted and entered into the database directly. For those that only provided the mean values of tritium contents or described their ranges in graphs, the values were inversely extracted using a digitizer function in Origin 9.3 (OriginLab, USA). In addition, data on tritium in precipitation in China, Japan, and South Korea for the period 1990–2020 were also obtained from the unified data source of the Global Network of Isotopes in Precipitation (GNIP) [28]. As a result, 6743 sets of data were registered in the database for further analysis.

2.3 Data analysis

To probe the temporal variation in environmental tritium levels in different regions in China, Japan, and South Korea, the data monitored in the vicinity of nuclear facilities were first excluded considering that they could be largely influenced by releases from the facilities. Based on the sampling years, the data monitored in the regions without nuclear facilities were categorized into three periods: 1990s, 2000s, and 2010s. For more rational comparisons, data from the same sampling site in the same period were combined and described by the median and extreme range if the dataset could not pass the normality test.

To reveal the spatial profile of natural tritium levels in the three countries in different periods, correlation analyses between the reported tritium level and latitude of the sampling sites were performed for cities with no less than five sets of data. If only the name of the sampling site was available, the latitude was retrieved from Google Earth. For those where only the names of provinces were reported, the capitals of the provinces were defined as the sampling site. The relationship between the tritium level and latitude was tested using SPSS Statistics 27 software (IBM, USA). The Pearson correlation test was selected for data following a normal distribution; otherwise, the Spearman correlation test was employed. Significance was defined as $P < 0.05$ for all analyses.

3 Results and discussion

3.1 Levels of environmental tritium in East Asia in past 30 years

In the atmosphere, tritium typically exists in three chemical forms: HTO, tritiated hydrogen (HT), and tritiated methane (CH_3T), which are eventually brought into different water pools in the form of HTO. In soil, tritium exists in the form of loose water tritium (LWT) and OBT. In living organisms such as plants, tritium exists in TFWT and in the form of OBT. After intake, HTO can be absorbed entirely and evenly distributed throughout the body within an hour, resulting in internal radiation exposure. In comparison, only approximately 0.01 and 1% of inhaled HT and CH_3T, respectively, are converted into HTO and absorbed by the human body [29]. Therefore, the high percentage, mobility, and transferability of HTO make its radiation impact considerably greater than those of HT and CH_3T, which in turn leads to the monitoring of HTO being more commonly performed. To monitor HTO in the atmosphere, atmospheric water vapor can be first collected using the desiccant adsorption, frozen, bubbling, or dehumidified methods [1]. Then, the desorbed water is typically transferred to a beta-ray counter or spectrometer to determine the activity of tritium in the sample. When monitoring HT and CH_3T in the atmosphere, LWT and OBT in soil, or TFWT and OBT in organisms, more cumbersome and complicated pretreatment is required to obtain the water sample for tritium measurement. The activity concentration of tritium in a water sample is typically expressed as Bq L^{-1}. When the absolute humidity of the air during sampling is avaliable, the activity concentration of tritium in the atmosphere can be expressed as Bq m^{-3}.

Table 1 summarizes the amount of the data extracted from China, Japan, and South Korea. China had the largest amount of data (n = 3035), followed by Japan (n = 2821), and South Korea (n = 887). Among these environmental samples, the number of samples from the atmosphere, precipitation, surface water, and shallow groundwater were relatively sufficient; therefore, the level comparison and profile study on tritium in the four media were performed in this review.

Table 2 lists tritium activity concentrations monitored in different environmental media in East Asia over the past 30 years. As indicated in Table 2, tritium in the atmosphere, precipitation, and water bodies was mainly measured in the form of HTO. This reflects both the importance

Table 1 Reported tritium data in different media in China, Japan, and South Korea in past 30 years

Media (Form of tritium)	Amount of data			Sub-total
	China	Japan	South Korea	
Atmosphere (HTO)	374	575	91	1040
Atmosphere (HT)	7	337	ND	344
Atmosphere (CH_3T)	168	127	ND	295
Precipitation (HTO)	1222	694	270	2186
Surface water (HTO)	511	545	48	1104
Shallow groundwater[a] (HTO)	344	209	34	587
Deep groundwater[a] (HTO)	44	No Data	11	55
Unspecified groundwater[a] (HTO)	18	No Data	85	103
Soil (LWT)	151	31	31	213
Soil (OBT)	29	12	No Data	41
Living organisms (TFWT)	82	171	158	411
Living organisms (OBT)	85	120	159	364
Total	3035	2821	887	6743

[a] For groundwater, a sampling depth of 150 m was set as a threshold to divide the groundwater into shallow or deep groundwater

of HTO and relative convenience of the HTO measurements [5, 19, 27]. For tritium in soil and living organisms, even though the measurements were relatively difficult and time-consuming, the data still accounted for 36% of the total in the vicinity of nuclear facilities. This implies that the ecological impact of anthropogenic tritium is also a concern in East Asia.

As indicated in Table 2, the tritium levels in East Asia change considerably with the environmental media and vary with different measurements even for the same medium. Compared with areas without nuclear facilities, the maximum values of tritium concentrations measured in regions with nuclear facilities were typically one to five orders of magnitude greater, and the ranges of tritium concentrations were also more variable. Moreover, in comparisons of the median and mean values of environmental tritium, it can also be observed that the difference in the vicinities of nuclear facilities was considerably greater than that in areas without nuclear facilities, indicating that different amounts of anthropogenic tritium were released in the vicinity of nuclear facilities. Furthermore, in comparisons of the median or mean values of tritium concentrations in soil and organisms, it was found that the levels in the vicinity of nuclear facilities were approximately one order of magnitude greater than those in areas without nuclear facilities. This implies that vicinities near nuclear facilities were contaminated by tritium release. When comparing the median values in the atmosphere, precipitation, and water, no significant difference was found among locations near nuclear facilities and areas without nuclear facilities. This could be explained by the fact that tritium was bound to the abundant organic components in the soil and organisms through isotopic exchange, which consequently led to the historical accumulation of tritium generated by anthropogenic pathways. While both the atmospheric and water environments are dynamic, the organic components in the atmosphere, precipitation, and water are typically negligible. The above results suggest significant differences in the patterns of source-sink competition for tritium in different environmental media. In fact, the same phenomenon in which tritium levels vary in different environmental media has also been reported in other regions of the world [20, 21].

3.2 Tritium levels in atmosphere in East Asia

3.2.1 Atmospheric HTO in areas without nuclear facilities

Among the different environmental media, except for the liquid discharge of anthropogenic tritium, the majority of the tritium first appears in the atmosphere. In the atmosphere, approximately 99% of tritium exists in the chemical form of HTO; its dose conversion factor (1.8×10^{-11} Sv Bq^{-1}) is more than two magnitudes greater than that of HT or CH_3T [30]. Furthermore, the measurement of HTO is considerably easier than that of HT or CH_3T. Consequently, atmospheric HTO has been the most frequently studied in countries or regions of interest for a long time.

In East Asia, China conducted several atmospheric weapons tests before 1980, and Japan began operating the first nuclear power plant in 1963. To assess the large-scale

Table 2 Tritium concentrations in different media in East Asia reported in past 30 years

Areas	Media (Form of tritium)	Amount of data	Concentration			
			Range	Median	Mean	Unit
Without nuclear facilities	Atmosphere (HTO)	416	0.1 ~ 31.5	0.9	1.7	Bq L^{-1}
	Atmosphere (HTO)	151	0.4 ~ 79.6	10.1	16.0	mBq m^{-3}
	Atmosphere (HT)	137	1.1 ~ 68.0	26.3	21.8	mBq m^{-3}
	Atmosphere (CH_3T)	130	0.4 ~ 34.0	10.6	9.3	mBq m^{-3}
	Precipitation (HTO)	1936	0.1 ~ 23.5	1.0	1.9	Bq L^{-1}
	Surface water (HTO)	863	< 0.1 ~ 11.1	0.7	1.3	Bq L^{-1}
	Shallow groundwater[a] (HTO)	378	< 0.1 ~ 8.4	0.9	1.2	Bq L^{-1}
	Deep groundwater [a] (HTO)	57	< 0.1 ~ 1.2	0.1	0.3	Bq L^{-1}
	Unspecified groundwater (HTO)	59	< 0.1 ~ 4.8	0.6	1.0	Bq L^{-1}
	Soil (LWT)	20	0.5 ~ 3.2	1.3	1.4	Bq L^{-1}
	Soil (OBT)	12	0.6 ~ 3.4	2.0	2.0	Bq L^{-1}
	Living organisms (TFWT)	85	0.5 ~ 10.6	2.0	2.3	Bq L^{-1}
	Living organisms (OBT)	104	0.4 ~ 61.4	2.1	4.4	Bq L^{-1}
Vicinity of nuclear facilities	Atmosphere (HTO)	38	0.3 ~ 5600	1.9	181	Bq L^{-1}
	Atmosphere (HTO)	435	1.3 ~ 13,710	27.0	3822	mBq m^{-3}
	Atmosphere (HT)	207	4.1 ~ 2763	8.4	30.3	mBq m^{-3}
	Atmosphere (CH_3T)	165	0.3 ~ 164	1.2	3.1	mBq m^{-3}
	Precipitation (HTO)	250	0.1 ~ 1554	1.1	66.2	Bq L^{-1}
	Surface water[a] (HTO)	241	< 0.1 ~ 422	1.4	8.3	Bq L^{-1}
	Shallow groundwater[a] (HTO)	209	< 0.1 ~ 790,000	0.4	5838.0	Bq L^{-1}
	Unspecified groundwater (HTO)	44	< 0.1 ~ 472	6.2	45.5	Bq L^{-1}
	Soil (LWT)	193	0.1 ~ 1222	29.1	62.8	Bq L^{-1}
	Soil (OBT)	29	5.8 ~ 248	46.2	70.2	Bq L^{-1}
	Living organisms (TFWT)	326	0.3 ~ 983	8.2	38.6	Bq L^{-1}
	Living organisms (OBT)	260	0.3 ~ 1050	14.3	34.4	Bq L^{-1}

[a]For groundwater, a sampling depth of 150 m was set as a threshold to divide the groundwater into shallow or deep groundwater

and long-term effects of anthropogenic tritium release on the environment, surveys on atmospheric HTO have been conducted in areas without nuclear facilities for many years in both China and Japan. Figure 1 plots the pooled concentrations of atmospheric tritium [27, 31–55] by latitude and distance from the proximal coastlines in China and Japan over the past three decades.

As indicated in Fig. 1a, the level of atmospheric HTO has significantly decreased in China since the 1990s; the same tendency can also be observed in Japan. The median value in China in the 1990s (10.80 Bq L^{-1}) is approximately 30 times greater than that (0.34 Bq L^{-1}) in the 2010s; the decrease in Japan is also more than four times (1.90 Bq L^{-1} in the1990s, 0.37 Bq L^{-1} in the 2000s). It is well known that tritium transporting from the stratosphere to the troposphere (so-called "tropopause input") is the major tritium source by nature in our living environment. In the case of atmospheric nuclear weapon tests, large amounts of anthropogenic tritium were introduced into the stratosphere and mixed with cosmogenic tritium; the level of tritium in the atmosphere near the earth's surface increased through the STEM process [27] after the tests. However, tritium released from atmospheric nuclear weapons tests naturally decays and spread globally. Therefore, it is reasonable to explain the different decreasing rates of atmospheric HTO between China and Japan after the end of atmospheric nuclear weapon tests in 1980. After the 2010s, the atmospheric HTO returned to approximately its natural level in East Asia, indicating that the impact of nuclear weapon tests on the atmospheric HTO in East Asia is negligible at present.

Regarding the level of atmospheric HTO in China, a clear change with latitude can also be observed in Fig. 1a. A Spearman correlation analysis demonstrated that the correlation coefficients between the level of atmospheric HTO and latitude in China were 0.59 and 0.84 for the 1990s and 2010s, respectively; the P values were all less than 0.01. As mentioned above, natural tritium is mainly

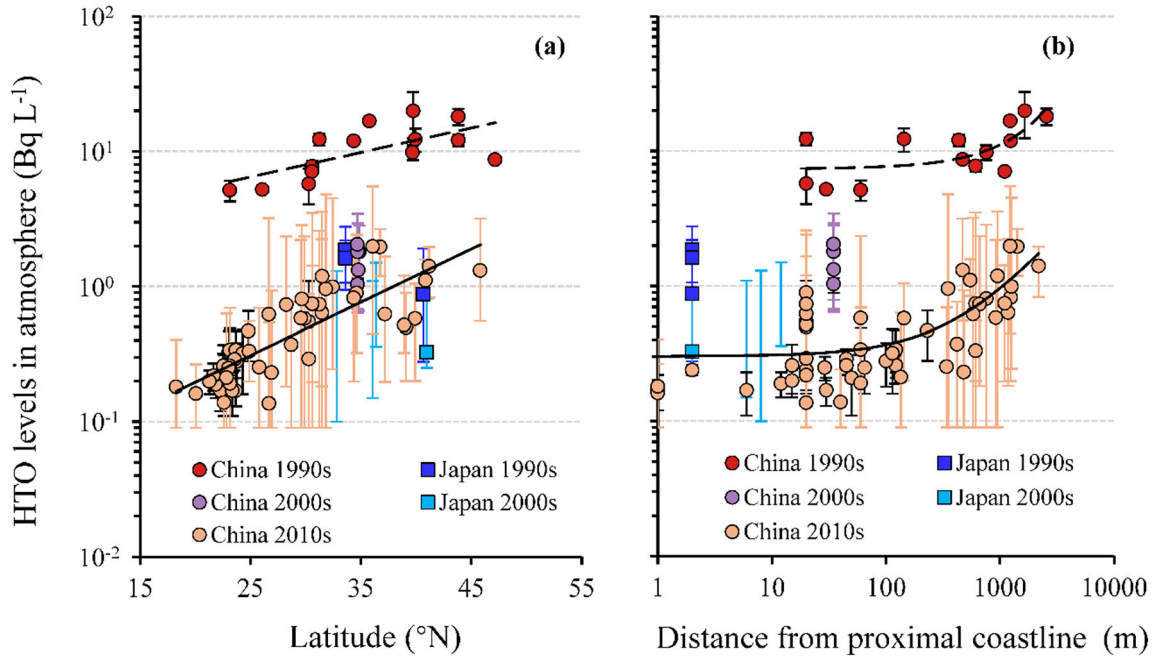

Fig. 1 (Color online) Levels of atmospheric HTO monitored in regions without nuclear facilities in China and Japan: **a** latitudinal profile, **b** continental profile

produced by the nuclear reactions of cosmic neutrons with ^{14}N in the upper atmosphere. Moreover, the intensity of cosmic rays is positively correlated with the geomagnetic latitude. Therefore, it is easy to explain the latitudinal distribution of atmospheric HTO levels. Despite the release of tritium from atmospheric nuclear weapon tests before 1980, the rapid homogenization of stratospheric material did not significantly change the latitudinal distribution of tritium sources from south to north. However, the net flux of STEM was reported to be considerably greater in the mid-latitude region where East Asia is located than in the low-latitude or high-latitude regions because of the occurrence of tropopause folds [2, 18, 27]. This natural phenomenon also supports the latitudinal distribution of atmospheric tritium. Furthermore, several studies have pointed out that the thickness of the tropopause typically decreases from the equator to the poles [27]. This means that anthropogenic tritium in the tropopause could experience a greater dilution in low-latitude regions. In summary, the significant latitudinal distribution of atmospheric HTO in China could be attributed to the above reasons.

Compared to China, the latitudinal distribution of atmospheric HTO levels in Japan is less apparent, as indicated in Fig. 1a. In general, the ocean is considered a sink for storing environmental tritium, and a large amount of ocean water can significantly dilute the level of atmospheric tritium, i.e., the level of tritium in oceans is extremely low [3, 18]. Therefore, an air mass originating from the ocean is often considered a tritium-depleted source. When the tritium input from the tropopause is vertically transported to the ground surface, the horizontal transport of the oceanic air mass with an extremely low level of tritium can dilute the level of atmospheric tritium. As the territory of Japan is completely surrounded by the Pacific Ocean, Sea of Okhotsk, Sea of Japan, and East China Sea, it is thought that the more significant impact of oceanic air masses on atmospheric tritium makes the latitudinal distribution of atmospheric HTO less obvious. To test the above explanation of the oceanic air mass effect, the atmospheric HTO levels were replotted against the distance from the proximal coastline in both China and Japan, as displayed in Fig. 1b. It is clear that the closer to the proximal coastline, the lower the level of atmospheric HTO, which can be generally observed in China, regardless of the monitoring period. Correlation analysis revealed that the correlation coefficients between the level of atmospheric HTO and distance from the proximal coastline in both China and Japan were 0.72 and 0.70 in the 1990s and 2010s, respectively; the *P* values were all less than 0.01. In fact, a significant dilution effect of oceanic air mass on the level of atmospheric HTO has been reported in previous studies [33, 34, 38].

3.2.2 *Atmospheric HTO in vicinity of nuclear facilities*

Table 3 summarizes the atmospheric HTO reported in the vicinity of the nuclear facilities in China, Japan, and South Korea over the past 30 years. The facilities included

nuclear power plants (NPPs) (Qinshan and other NPPs in China, Fukushima in Japan, and Wolsong NPPs in South Korea), a fission research reactor (Kyoto University in Japan), nuclear fuel reprocessing plant (Tokai in Japan), and fusion test device (National Institute for Fusion Science (NIFS) in Japan). As shown in Table 3, the concentrations of atmospheric HTO monitored in the vicinity of the nuclear facilities varied significantly at the different sampling sites. In particular, in the vicinity of heavy-water reactors (HWRs) (Wolsong NPPs and Qinshan NPPs), the differences between the minimum and median or maximum values were several orders of magnitude, and a clear difference was also observed in the vicinity of the research reactor at Kyoto University. The results indicate that the release of anthropogenic tritium from nuclear facilities can lead to hotspot concentrations, and attention should be given to tritium release from HWRS and even research reactors. This implies that more detailed monitoring of atmospheric tritium with higher spatial and temporal resolutions in the vicinity of nuclear facilities is desirable and necessary for more accurate assessments of the potential impacts of tritium release from nuclear facilities on the ecological environment.

3.2.3 Atmospheric HT and CH3T in vicinity of nuclear facilities

For measurements of atmospheric HT and CH_3T, the gaseous samples typically first require conversion into HTO under high-temperature oxidation conditions. Sample pretreatments are more time-consuming and difficult than atmospheric HTO measurements. However, monitoring of atmospheric HT and CH_3T in the vicinity of nuclear facilities are indispensable for the accurate evaluation of the release and operating status of nuclear facilities.

Table 4 displays the reported concentrations of atmospheric HT and CH_3T monitored near Qinshan NPPs in China, the nuclear fuel reprocessing plant in Tokai, and fusion test device in the NIFS in Japan. As indicated in Table 4, wide ranges of atmospheric HT and CH_3T were observed in the vicinity of the nuclear facilities; the concentrations depended on the type of facility. Overall, the concentrations of HT and CH_3T were relatively low near the NIFS, as they were in a test status. Extremely high concentrations of HT were observed in the vicinity of the heavy-water reactor at Qinshan and nuclear fuel reprocessing plant in Tokai. By comparing the median values of HT and HTO (see Table 3), it was found that the released HT accounted for at least 10% of the total tritium for the HWR and fuel reprocessing plant. In a more detailed survey, Koarashi et al. reported that 19–28% of the tritium released to the atmosphere from the reprocessing plant in Tokai presented the HT form [56]. The above results indicate that not only HTO, but also HT and CH_3T should be monitored in the vicinity of nuclear facilities. Moreover, the emission and environmental fate of HT and CH_3T should be further studied to assess their ecological effects.

3.3 Tritium levels in precipitation

For a more objective reflection of the spatial and temporal distributions of natural tritium in precipitation (also called rain tritium), the concentrations of rain tritium reported in areas without nuclear facilities in China [28, 36, 37, 41, 42, 45, 48, 49, 57–62], Japan [28, 35, 39, 51, 52, 63–70] and South Korea [28, 71–77] in the past three decades were extracted and plotted against the latitude in Fig. 2.

Compared with the atmospheric HTO concentrations displayed in Fig. 1, it can be observed that although the level of tritium in precipitation is slightly lower than that in the atmosphere, a similar latitudinal distribution of tritium concentrations can be observed in precipitation. Based on the biogeochemical cycle of hydrogen isotopes, tritium and its isotopes originating from the tropopause input are primarily presented as ice crystals before reaching the melting layer approximately 5 km above the earth's surface. When they are fully converted into tritiated water vapor, the dynamic isotope exchange of condensation–evaporation with the surrounding droplets occurs during the subsequent deposition process. Therefore, the level of tritium in raindrops largely depends on the level of tritium in the

Table 3 Atmospheric HTO concentrations reported near nuclear facilities in East Asia

Country	Sampling site	Concentration (mBq m^{-3})			
		Minimum	Median	Maximum	Reference
China	Qinshan	6.52	144	4363	[43, 44, 50, 148]
	Other NPPs	10.0	22.0	151	[45, 46, 48, 49]
Japan	Fukushima	2.59	14.8	143	[55]
	Tokai	12.0	20.0	1100	[51, 54]
	NIFS	1.25	6.07	26.3	[38, 53, 153]
	Kyoto University	9.03	96.4	9683.1	[52]
South Korea	Wolsong	6.32	1788	113,710	[71, 127, 72, 106]

Table 4 Concentrations of atmospheric HT and CH_3T reported near nuclear facilities in China and Japan

Country	Site	Tritium form	Concentration (mBq m^{-3})			Reference
			Minimum	Median	Maximum	
China	Qinshan	HT	1.06	15.4	2763	[43, 50]
		CH_3T	1.97	7.12	164	[43, 50]
Japan	NIFS	HT	4.07	8.02	11.3	[38, 53, 153]
		CH_3T	0.26	1.15	6.14	[38, 53, 153]
	Tokai	HT	8.5	17.0	374.1	[51, 54, 154]
		CH_3T	ND	6.6	No Data	[155]

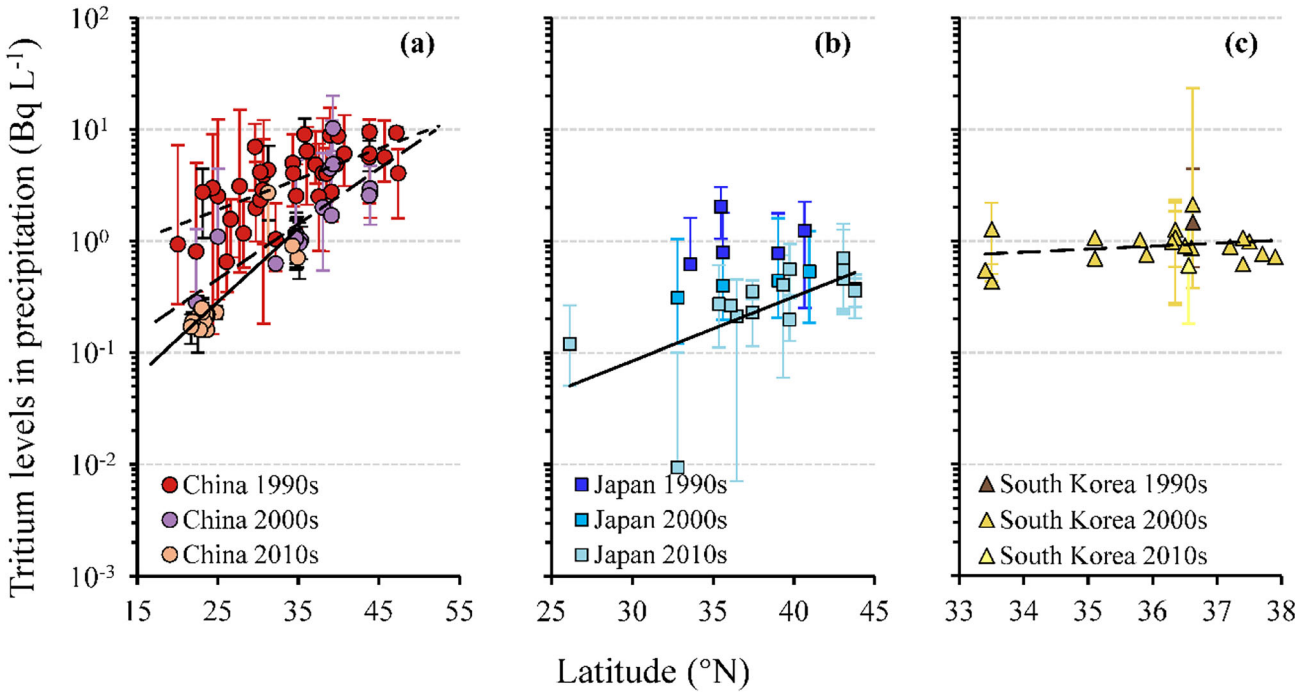

Fig. 2 (Color online) Levels of tritium (Bq L^{-1}) in precipitation: **a** in China, **b** Japan, and **c** South Korea

atmosphere and isotope exchange rate, which allows the latitudinal distribution of atmospheric tritium to be replicated in the rainfall. However, the dilution effect caused by the abundant number of hydrogen stable isotopes in rainfall and the challenge in establishing isotope exchange equilibrium together could lead to a slightly lower level of tritium in rainfall than in atmospheric water vapor.

As shown in Fig. 2a, both latitudinal and temporal variations in HTO concentrations in precipitation are apparent in China. The positive correlation between latitude and tritium level in precipitation was significant in all three periods (1990s: $r = 0.63$, $P < 0.01$; 2000s: $r = 0.78$, $P < 0.01$; 2010s: $r = 0.55$, $P < 0.01$). This confirms the influence of the tropopause input on tritium during precipitation. Considering the reported data in China in different periods covered a wide range of latitudes, for simplicity, only the data on rain tritium reported near 25°N were further extracted to study the temporal change in rain. In the region of 25°N, the median concentrations of tritium in precipitation were 1.27 and 0.21 Bq L^{-1} in the 1990s and 2010s, respectively. The decline was approximately six times over three decades, which was marginally faster than the decline (approximately 5.4 times) due to the physical decay of tritium. Conversely, the median concentrations of tritium in atmospheric water vapor were 4.62 and 0.20 Bq L^{-1} in the 1990s and 2010s, respectively, and the decline coefficient was approximately 23. The comparison result implies that there is a strong dilution effect of the tritium transfer process from the atmosphere to rainwater. Furthermore, rapid decreases in tritium concentrations in both the atmosphere and rainwater also suggest that oceanic air masses with extremely low levels of tritium can largely dilute the levels of tritium in both the atmosphere and rainfall, especially in low-latitude coastal regions.

As indicated in Fig. 2b, the latitudinal and temporal variations in tritium in precipitation in Japan were similar to those in China. The median tritium concentrations in precipitation in Japan were 0.69 and 0.27 Bq L^{-1} in the 1990s and 2010s, respectively. The decline coefficient was approximately three, which was smaller than that in the region of 25°N in China. The main reason for this difference could be that the concentration of tritium in precipitation in China in the 1990s was approximately one order of magnitude greater than that in Japan in the same period, and the majority of the monitoring sites in Japan were north of 25°N [33, 34, 38]. In general, the high concentration of tritium in precipitation in China in the 1990s is considered to be the residual effect of atmospheric nuclear weapon tests. However, both the dilution of long-distance transportation of air masses from China and the relatively considerable dilution effect of oceanic air masses typically reduce the concentration of tritium in precipitation in Japan. As indicated in Fig. 2c, the level of tritium in precipitation in South Korea was between those in China and Japan in the same latitudinal span. This can be explained by the reasons that the concentration of atmospheric tritium in South Korea is more likely to be influenced by continental monsoons from high-latitude regions [33, 34, 38], and the dilution effect of oceanic air masses is smaller than that in Japan. Based on the data from a national survey in South Korea [77], a significant correlation (correlation coefficient of 0.65) was also found between the concentration of tritium in precipitation and distance to the coastline.

It should be noted that the level of tritium in precipitation monitored at the majority of sites in Japan, South Korea, and coastal regions of China in the 2010s decreased to less than 1 Bq L^{-1}, which could raise a challenge to precisely determine the tritium concentrations in rainfall using conventional sampling and measuring methods. Therefore, further research and development of sampling and sample pretreatment methods as well as more advanced measurement devices are desired for accurate measurements of low-level environmental tritium in the future. Tritium levels with one to four orders of magnitude higher than the baseline were also observed in precipitation collected near nuclear facilities in South Korea, Japan, and China [37, 41, 45, 46, 48, 49, 63, 70, 72]. For example, tritium in precipitation was observed to be greater than 19 Bq L^{-1} in Tsukuba at the beginning of the Fukushima accident [66, 75]. In South Korea, tritium in precipitation around the Wolsong NPPs was reported [58] to be as high as 1554 Bq L^{-1}. These observations indicate that rapid isotope exchange can occur on the surface surrounding a nuclear facility, which allows the tritium concentration in precipitation to be an indicator of the tritium emission status in these facilities.

3.4 Tritium levels in surface water

Driven by the hydrologic cycle process, HTO in the atmosphere and precipitation can be delivered to surface water reservoirs through surface runoff. Figure 3 plots the reported concentrations of tritium in surface water and latitude of the monitoring sites in areas without nuclear facilities in China [31, 36, 41, 44–48, 57, 59, 61, 62, 78–87], Japan [88–96], and South Korea [97] over the past three decades. As indicated in Fig. 3, the overall level of tritium in the surface water was marginally lower than that of the atmospheric HTO at the same site and period; the level of tritium in river water was similar to that in lake, spring, and drinking waters, whereas the level of tritium in seawater was the lowest. The low concentration in seawater can be attributed to the dilution effect of the large amounts of stable hydrogen isotopes (i.e., ^{1}H and ^{2}H) in the ocean.

As in Figs. 1 and 2, similar latitudinal distributions of tritium concentration in surface water in China and Japan can also be seen in Fig. 3. Correlation analyses have indicated that the correlation coefficients between the tritium concentrations in surface water in the 1990s in China and the latitudes were 0.692, 0.766, and 0.741 for river, lake, and drinking water, respectively; the P values were all less than 0.01.

During the observed period, the median tritium concentrations in surface water in China were approximately 3.02 and 1.34 Bq L^{-1} in the 1990s and 2010s, and the decreasing rate was estimated to be approximately 55.6% in the past years, which was considerably less than that of atmospheric HTO (96.8%) or tritium in precipitation (95.6%). It is thought that surface water not only has high mobility, but also contains abundant stable isotopes of hydrogen, leading to a rapid dilution effect on the difference in tritium global fallout. For the median tritium concentration in the surface water in Japan, a decreasing trend was also observed. However, this decline (43.8%) was less than that in China. This difference could be explained by the fact that surface water remains subject to persistent differences in tritium supply for specific latitudinal characteristics due to their direct exposure to air. Similarly, as the continuous weakening of tritium has originated from nuclear weapon tests and the intrusion of oceanic air masses, the level of tritium in surface water has largely decreased in the past years. In particular, in coastal regions the level of tritium in surface water at present is approaching the level in seawater. Such a low tritium level presents a new challenge for precise measurement.

Conversely, greater than 1000 Bq L^{-1} of tritium in surface water was reported near the Wolsong NPPs [98]. After the accident at the Fukushima Daiichi NPPs, the level of tritium was observed to be 184 Bq L^{-1} in a small pool 1.5 km distant from the accident site [94]; the level of

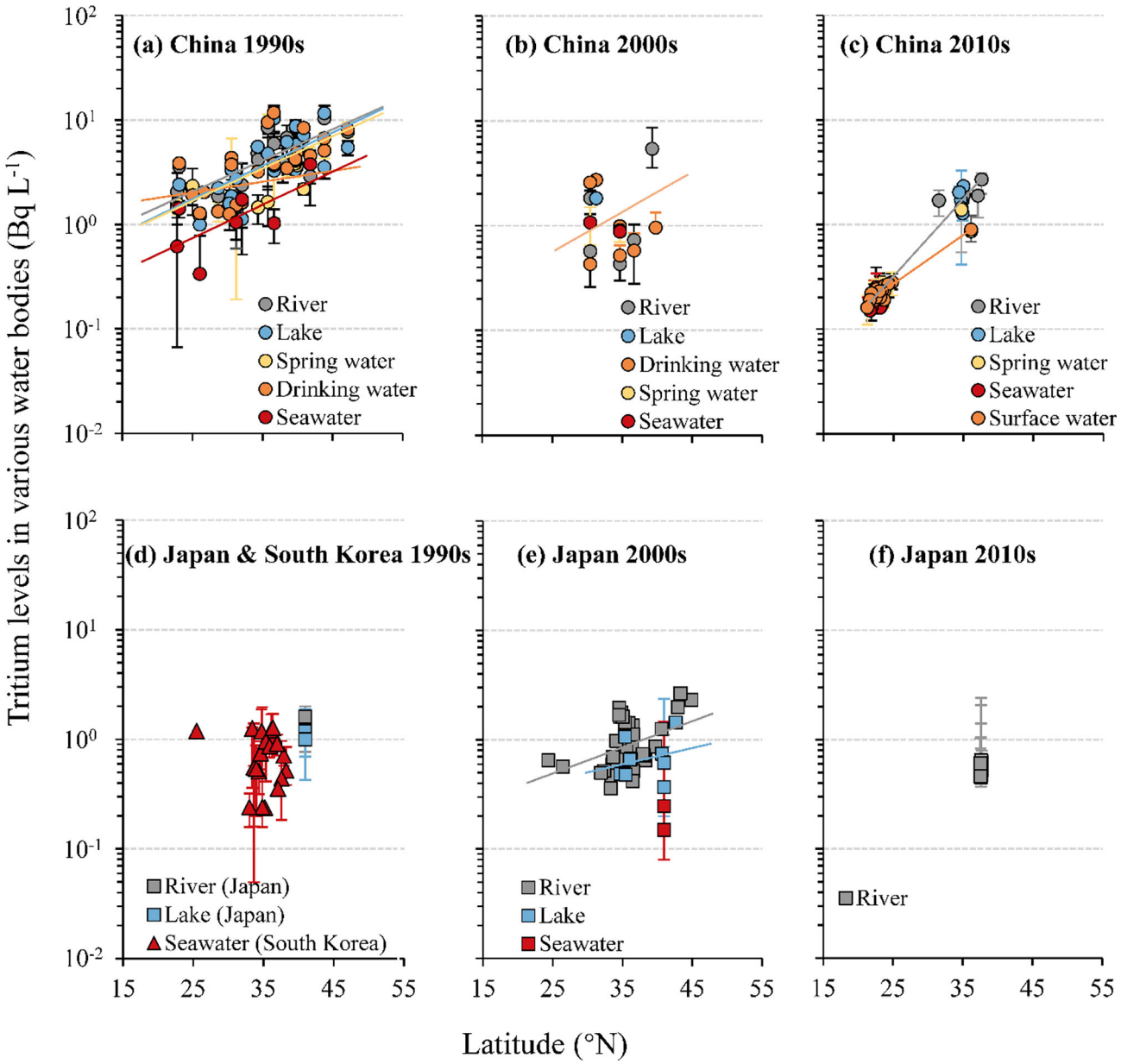

Fig. 3 (Color online) Levels of tritium (Bq L^{-1}) in different bodies of water in East Asia

tritium in the near seawater was also found to be three orders of magnitude greater than its original [99, 100]. These results indicate that the surrounding environments were significantly contaminated by tritium released from the HWRs and the accident. However, it should be noted that the high concentrations of tritium in surface water are still within the regulatory limit (6×10^4 Bq L^{-1}) for activity concentration in Japan [98]. In Japan, the annual discharges of the nuclear fuel reprocessing plant in Tokai were reported to be 1.9×10^{15} and 5.4×10^{14} Bq a^{-1} for gaseous and liquid tritium, respectively [101, 102], whereas the annual discharge limit has not been found for nuclear power plants. In China, both the annual activity and concentration limits have been set for tritium discharge. For a 3000 MW NPP in China, the annual activity limitations of gaseous tritium discharge are set to 1.5×10^{13} and 4.5×10^{14} Bq a^{-1} for pressured water reactors and HWRs, respectively; the annual limitations of liquid tritium discharge are set to 7.5×10^{13} and 3.5×10^{14} Bq a^{-1} for pressurized water reactors and HWRs, respectively [26]. Furthermore, the concentration limit of liquid tritium discharge is set to 100 Bq L^{-1} for an inland NPP in China [103]. In South Korea, it has been reported that the annual release limits for HWRs in gaseous and liquid effluents are controlled within 1.05×10^{17} and 3.48×10^{16} Bq a^{-1}, respectively [98, 104]. In summary, not all East Asian

countries have set both the annual activity and concentration limits for tritium release from nuclear facilities. Moreover, the existing limits are also inconsistent, even for the same reactor type.

3.5 Tritium levels in groundwater

Compared with surface water that can directly receive tritium supply from the atmosphere, the recharge of tritium in groundwater relies primarily on surface infiltration, making its concentrations and spatiotemporal profiles strongly relevant to the surface inventory of tritium and regional hydrogeological conditions. Before nuclear weapon tests, the level of natural tritium in groundwater was generally lower than 1 T.U. (one tritium unit is defined as one tritium atom per 10^{18} atoms of hydrogen), which is calculated to be approximately 0.118 Bq L^{-1} by setting Avogadro's number, the half-life of tritium, and molar mass of water to 6.02×10^{23}, 4500 days, and 18.0153×10^{-3} kg mol^{-1}, respectively. However, the level would increase after receiving recharge from "modern water" (i.e., water sources carrying anthropogenic tritium information) [105]. This feature makes tritium in groundwater an excellent indicator of the source and age of the recharged water. In addition, as water sustainability and quality are essential to highly populated regions, a number of investigations on tritium in groundwater have been conducted in East Asia over the past decades.

Figure 4 displays the level of tritium in shallow groundwater (here defined as a sampling depth < 150 m) investigated in areas without nuclear facilities in China, Japan, and South Korea over the past three decades [31, 32, 41, 58, 62, 69, 76, 81, 84, 88, 99, 106–126]. Compared with the level of tritium in surface water, the level of tritium in shallow groundwater is virtually the same or marginally lower than that in surface water at the same site and during the same period. Furthermore, the spatiotemporal distribution of tritium levels in shallow groundwater is similar to that in surface water.

In spatial terms, the latitude effect is also clearly present in shallow groundwater in areas without nuclear facilities in China and South Korea, as indicated in Fig. 4. In the 1990s, the correlation coefficients between the tritium concentration in shallow groundwater and latitude were found to be 0.60 and 0.80 in China and South Korea, respectively. The positive correlations were all significant ($P < 0.01$), suggesting that the level of tritium in groundwater in East Asia was also influenced by the latitude effect. In addition, several studies have indicated that a significant negative correlation exists between the concentration of tritium and depth of groundwater [107]. Combining the statistical data presented in Table 2, it can be observed that the median value of tritium in deep groundwater is approximately one order of magnitude lower than that in shallow groundwater. This concentration gradient related to groundwater depth confirms the downward migration of tritium from the surface to the ground layer.

In addition, a significant decrease in the tritium level of groundwater has been observed in regions without nuclear facilities in China, Japan, and South Korea over the past three decades. In the 2010s, the median tritium concentrations in China, Japan, and South Korea were 0.71, 0.37, and 0.72 Bq L^{-1}, respectively, which were approximately 27.0, 42.9, and 43.5% less than their original values in the 1990s. Therefore, the tritium level in shallow groundwater in areas without nuclear facilities is approaching its natural value in the majority of regions of East Asia.

Nevertheless, it has also been observed that the tritium concentration in shallow groundwater near two nuclear power plants in East Asia was approximately four orders of magnitude greater than the natural level [127, 128]. Moreover, after the severe accident at the Fukushima Daiichi NPPs, the concentrations of tritium in well water near the accident site ranged from 10 to 3000 Bq L^{-1} [99, 122], and the greatest concentration was reported to be as high as 7.9×10^5 Bq L^{-1} in well water near the damage site [99]. These results highlight the importance of strengthening tritium monitoring around nuclear facilities and accident sites.

3.6 Tritium levels in soil and living organisms

Surface organic-rich receptors such as those in soil and living organisms can also receive a continuous tritium supply from the atmosphere and precipitation. Because this is the main pathway for transferring tritium from inorganic to organic pools, the metabolic processes of tritium at all trophic levels of the food chain have attracted considerable attention in the field of radioecology research [129]. Depending on whether the environmental tritium is or is not tightly bound to the organic matter, the tritium in organisms can typically be classified into TFWT and OBT [130]. While in the soil environment, the concept of tissue-free water tritium is often referred to as LWT [131]. In this study, with the limitation of available data on OBT, the OBT was not further classified into exchangeable and non-exchangeable OBT for comparison, although this classification is vital from a radioecological point of view [132]. Moreover, it should be emphasized that the residence time of tritium in soil/living organisms depends significantly on the binding molecules. The data presented here provide only a snapshot of the dynamics of environmental tritium in a specific time window.

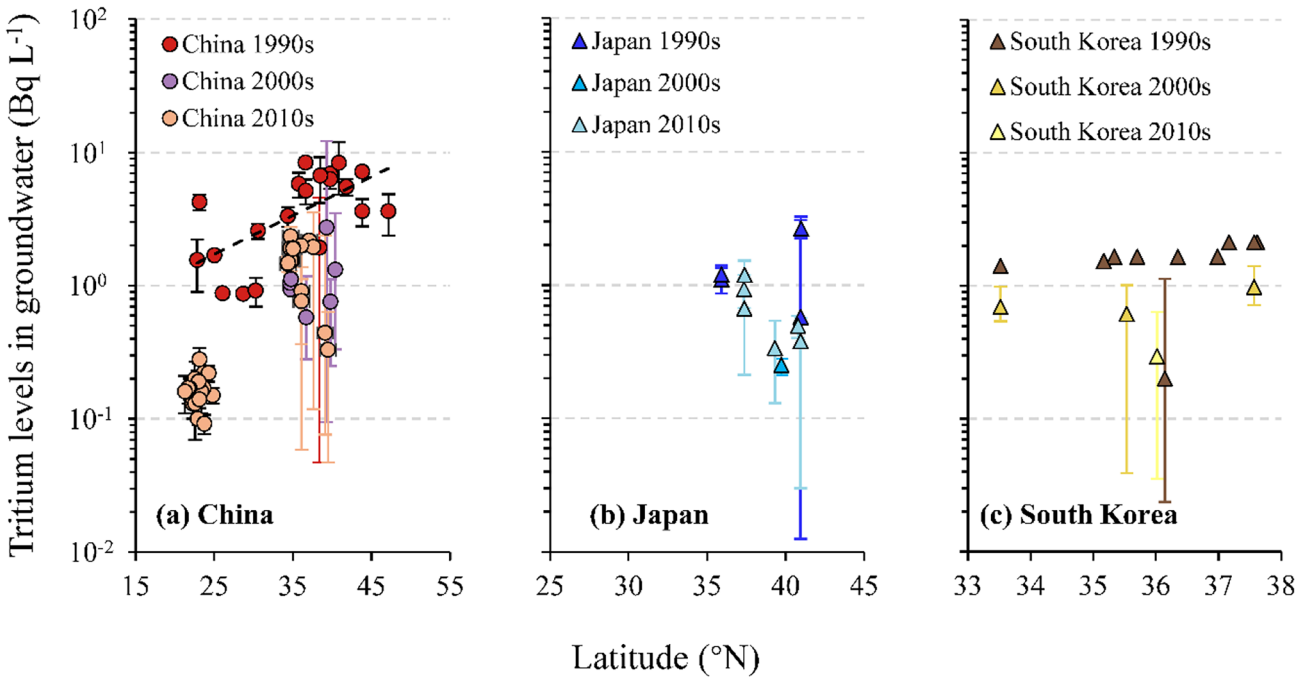

Fig. 4 (Color online) Level of tritium (Bq L^{-1}) in shallow groundwater in East Asia

3.6.1 Tritium levels in soil

Table 5 summarizes the tritium concentrations in soil reported in several places in China, Japan, and South Korea [54, 127, 133–138]. It can be observed that the mean LWT concentrations in soil in places without nuclear power plants (Akita and Rokkasho) are typically one to two orders of magnitude less than those in places with nuclear power plants (Qinshan, Tokai, and Wolsong).

Taking Akita in Japan as an example, the level of LWT in the area without nuclear facilities is slightly higher than the level of tritium in local precipitation (approximately 0.9–1.1 Bq L^{-1}) [138]. The reason for the higher level of tritium in soils is considered to be the additional HTO contribution from the oxidation of atmospheric HT in the soil [139]. However, because of the fast evaporation rate of soil water and limited depth (0–20 cm) of porous soil that can exchange with atmospheric water [130, 132, 139], the tritium concentration in soil typically remains at the same order of magnitude as that in the atmosphere and surface water. The rapid equilibrium of HTO in the atmosphere–soil–vegetation system was also verified in a field experiment performed in the vicinity of the Tokai reprocessing plant by Fujita et al. [54].

Table 5 Tritium concentrations in soil reported in sites in East Asia

Country	Specie	Site	Period	Depth (cm)	Concentration (Bq L^{-1})			Reference
					Minimum	Mean	Maximum	
China	LWT	Qinshan[a]	2010s	0–25	0.53	28.3	143	[133–137]
	OBT	Qinshan[a]	2010s	0–25	5.81	46.2	248	[133, 137]
Japan	LWT	Tokai[a]	2004	0–5	1.48	4.74	8.05	[54]
		Akita[b]	1989	0–20	1.24	2.05	3.16	[138]
		Rokkasho[b]	1989	0–20	0.52	0.63	1.38	[138]
	OBT	Akita[b]	1989	0–20	1.49	2.07	2.70	[138]
		Rokkasho[b]	1989	0–20	0.63	1.93	3.35	[138]
South Korea	LWT	Wolsong[a]	1992–1995	0–5	0.1	122	1223	[127]

[a]Regions with nuclear facilities

[b]Regions without nuclear facilities

However, as indicated in Table 5, the levels of LWT in the soil near the HWRs in both China and South Korea were more than one order of magnitude higher than those in areas without nuclear facilities. This implies that tritium released from heavy-water reactors can contaminate nearby soils. In general, the level of OBT in soil had a wider range and higher value than the level of LWT at the same depth. This demonstrates the persistence of tritium from historical release in the soil organic compartments.

3.6.2 *Tritium levels in living organisms*

Two indices are usually considered for tritium in living organisms (i.e., plants and animals). One is the difference between the TFWT value and level of tritium in the atmosphere or precipitation in the same region. The second is the persistence of tritium in the organic components of the environmental compartments, which is often expressed as the concentration ratio of OBT to TFWT.

Table 6 summarizes the concentrations of TFWT and OBT in living organisms in areas without nuclear facilities in China, Japan, and South Korea [79, 80, 140, 141]. As indicated in Table 6, both the concentrations of TFWT and OBT are typically in the same order of magnitude, and their medians are comparable to their local rain tritium levels at the same period (Shanghai: approximately 3.87 Bq L^{-1} [79, 142], Akita: 0.9–1.1 Bq L^{-1} [138], South Korea: approximately 1.5 Bq L^{-1} [23]). This suggests an isotope exchange equilibrium of HTO between precipitation and living organisms. However, it was also found that the concentrations of TFWT in pine needles and certain green vegetables were normally greater than those in other terrestrial organisms [140, 141]. These results indicate that different species have different tritium uptake and metabolism [129, 130, 139]. Therefore, species with relatively high uptake and fast metabolism of atmospheric tritium are often used as indicators to check for tritium release near nuclear facilities [44, 48, 71, 72, 136, 137, 143–147]. For example, numerous studies have used the TFWT concentration in pine needles as an indicator [51, 71, 72, 138, 143, 144, 148]. Recently, evergreen-tree leaves collected from the Fukushima region were also utilized to estimate the levels of environmental tritium in an accidental field at the beginning stage [145, 146].

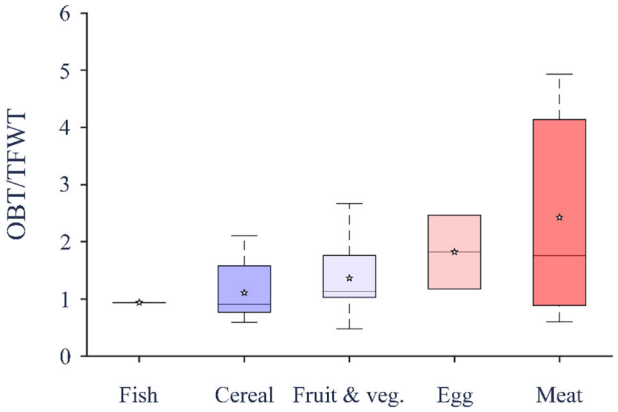

Fig. 5 (Color online) Concentration ratios of OBT/TFWT in different foods

Based on the data summarized in Table 6, the ratios of OBT to TFWT concentrations in food in the above areas are plotted in Fig. 5. As indicated in Fig. 5, the ratio of OBT/TFWT ranges from 0.4 to 5.1 among different species of food; the ratio also varies with sampling area, even for the same food. In a recent study, Fujita et al. reported that the concentration ratio of OBT/TFTW ranged from 0.55 to 1.47 in Chinese cabbage and cabbage collected near the Tokai reprocessing plant [149]. These results indicate the different persistence of tritium in different organic components and environmental compartments. Among the ratios, 66% of OBT/TFWT ratios were greater than one, which is similar to the value of 75% for terrestrial organisms reported in other regions [82].

4 Conclusions and perspectives

The level of environmental tritium in East Asia has attracted increasing attention owing to the rapid development of nuclear power plants and the coming release of tritium-contaminated wastewater from Fukushima into the Pacific Ocean [150, 151]. To better understand the environmental levels and behavior of both natural and anthropogenic tritium in East Asia, a relatively comprehensive review and analysis of environmental tritium was conducted in this work. The main findings are summarized in the following.

Table 6 Tritium concentrations in living organisms reported in East Asia

Area	TFWT (Bq L^{-1})			OBT (Bq L^{-1})			Reference
	Minimum	Mean	Maximum	Minimum	Mean	Maximum	
Shanghai, China	2.8	3.0	10.6	6.7	14.3	18.6	[79, 80]
Akita, Japan	0.8	1.6	3.3	0.7	1.4	2.1	[140]
South Korea	0.9	1.8	4.2	1.0	2.5	4.1	[141]

(1) The levels of environmental tritium vary significantly based on the existence or nonexistence of nuclear reactors, nuclear weapon tests, or the occurrence of nuclear accidents. In particular, in the vicinity of HWRs or accident sites, the level of environmental tritium can be several orders of magnitude higher than that in other areas without nuclear facilities.
(2) The impact of atmospheric nuclear weapon tests before the 1990s on environmental tritium has basically been eliminated as its physical decay, the global geo-ecological cycle, and the level of environmental tritium in the regions of East Asia without nuclear facilities have virtually returned to their natural background.
(3) In general environments, the level of tritium in the atmosphere is slightly higher than that in precipitation and surface water. The level of tritium in shallow groundwater is nearly of the same order of magnitude as that in surface water, and the level of seawater is typically the lowest among the above environmental media.
(4) In general environments, the levels of tritium in environmental media are positively related to the geomagnetic latitude and distance from the coastline. The level of environmental tritium in inland areas at higher latitudes is typically higher.
(5) The levels of TFWT and OBT and their ratios in certain living organisms are promising indicators for studying the impact of anthropogenic tritium release on environmental ecology.

This review established the largest database regarding the concentrations of environmental tritium to date. However, given the limited open literature and incomplete sampling information in the literature, the data summarized in this paper could possibly not fully reflect their representativeness in East Asia; in particular, the data on soil and living organisms remain far from sufficient. Seasonal variations in environmental tritium have also been reported [27]. Therefore, further surveys on environmental tritium are required to study the levels and behaviors of environmental tritium in East Asia.

Based on this review, it is clear that the level of environmental tritium in East Asia has returned virtually to its background value in areas without nuclear facilities. As it is usually not easy to accurately determine the background level of tritium at present [1, 152], more advanced sampling, pretreatment, and analytical methods need to be developed. Moreover, there is evidence that the environment near nuclear reactors and accident sites has been polluted by tritium release. Therefore, it is also necessary to continue studies on tritium behavior and its long-term ecological impacts.

Author Contributions All authors contributed to the study conception and design. Material preparation, data collection, and analysis were performed by Bin Feng and Wei-Hai Zhuo. The first draft of the manuscript was written by Bin Feng and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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