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Assessment of tritium content and radiation hazard in the water body of the Suifen river basin in China

Yan Shi[1,2,3] · ZhiQiang Liu[3] · Baiyao Ding[3] · Junfeng Zhao[4] · Yan Li[3] · Shan Wang[3] · Lei Wang[3] · Hongtao Zhao[1,2,3] · Yongjun Liu[1,2]

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Abstract
The tritium content in the surface water of the Suifen River in China was measured using an ultra low-level liquid scintillation spectrometer. In August and October, a total of 30 samples were collected (15 per month) The measured ^{3}H activity concentration ranged from 5.08 to 9.75 Bq L^{-1}, and the annual effective dose ranged from 0.07 to 0.13 μSv. These values are below WHO's recommendations, indicating no risk to human health. Although the ^{3}H activity concentration was higher in October than in August, statistical analysis did not show a significant difference between the two months.

Keywords Tritium · Surface water · Radioactivity · Annual effective dose · Seasonal

Introduction

Tritium (^{3}H) is a radioactive isotope of hydrogen, which decays into the stable element helium through beta decay, with a half-life of 12.3 years. The average energy of β ray emitted by the decay of tritium is 5.7 keV, with a maximum energy of 18.6 keV, Its maximum range in air is 5 mm, and about 0.56 μm in water. Natural tritium originates from the interaction between neutrons generated by cosmic rays in the upper atmosphere and nitrogen atoms, constituting the "stable" background of environmental tritium, accounting for 65% in the oceans and 27% on land and in the biosphere [1]. In nature, tritium primarily exists in the form of tritiated water (HTO), and participates in the natural water cycle through the transportation of rainwater and the exchange of water vapor between the air and the sea. Artificial tritium holds a significant place in the global tritium inventory, with its primary sources being nuclear tests and the operation of nuclear power reactors. Tritium emitted from nuclear facilities mainly exists in the form of tritiated water (HTO), tritiated hydrogen (HT), and a small amount of tritiated methane (CH_3T) [2, 3]. Natural tritium is present in the environment and food, existing in three chemical forms: HT, HTO and organically bound tritium (OBT), which enter the body through inhalation, skin absorption, or ingestion [4–6]. Moreover, nuclear power stations and nuclear explosions also release large amounts of tritium into the environment, far exceeding natural tritium levels, and transferring among natural ecological chains, such as aquatic food chains [7–10].

On August 24th, 2023, Japan began discharging Fukushima contaminated water into the ocean. The sources of Fukushima contaminated water mainly include: the original coolant in the reactors, the cooling water injected at the time of the accident, and the groundwater and rainwater that flows through the reactors [11]. The contaminated water directly contacts the damaged reactor cores and contains more than 60 fission products and transuranic elements found within the cores [12]. Despite Tokyo Electric Power Company (TEPCO) using multiple decontamination

✉ Hongtao Zhao
zhaohongtao2019@163.com

1 Key Laboratory of In-Fiber Integrated Optics of Ministry of Education, College of Physics and Optoelectronic Engineering, Harbin Engineering University, Harbin 150001, China

2 Key Laboratory of Photonic Materials and Devices Physics for Oceanic Applications, Ministry of Industry and Information Technology of China, College of Physics and Optoelectronic Engineering, Harbin Engineering University, Harbin 150001, China

3 Key Laboratory of Nuclear Technology Application, Heilongjiang Institute of Atomic Energy, Harbin 150086, China

4 College of Nuclear Science and Technology, Harbin Engineering University, Harbin 150001, China

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systems, including the Advanced Liquid Processing System (ALPS) and strontium and caesium adsorption devices, to treat the radioactive nuclides in the contaminated water, no decontamination system can remove all radioactive nuclides, especially tritium (^{3}H) [13]. In September 2023, TEPCO announced the latest measurement results of ten artificial radioactive nuclides in nearly 310 contaminated water storage tanks [14]. The results showed that the concentration of ^{3}H in the contaminated water storage tanks ranged from 1.31×10^5 to 2.50×10^6 Bq L^{-1}, which is significantly higher than the World Health Organization (WHO) recommended limit concentration of ^{3}H in drinking water (1×10^4 Bq L^{-1}), and also exceeds the maximum discharge concentration of ^{3}H permitted by Japanese law (6×10^4 Bq L^{-1}). As of January 2024, Japan has discharged 23,351 tons of contaminated water. Liu et al. established diffusion models of radioactive materials at both macroscopic and microscopic levels and achieved long-term simulations of the Fukushima contaminated water discharge plan. The simulation results found that the contaminated water would reach the coastal waters of China 240 days after discharge and the North American coast 1200 days later, covering almost the entire North Pacific [15].

Chum salmon (Oncorhynchus keta Walbaum) belong to the salmon family and the genus of large salmonids. They are typical anadromous fish species. As one of the Pacific salmon species, they are widely distributed along the coast of the North Pacific. Born in freshwater rivers, chum salmon migrate to the sea after hatching, where they grow and develop. Upon reaching maturity, they migrate upstream to their birthplaces to spawn and reproduce. After spawning, the parent fish gradually die [16]. Therefore, chum salmon can serve as indicator organisms to monitor the migration of radioactive nuclides from the ocean to freshwater rivers [17]. Every year, a large number of chum salmon from the North Pacific migrate to the Suifen River in China to spawn. With their migration, the radioactive nuclide tritium (^{3}H) from the ocean may be brought into and released into the Suifen River [7–9, 17, 18]. Suifenhe is located at the junction of the eastern segment of the North China Platform and the Jilin-Heilongjiang fold belt. It is also in the overlap zone between the ancient Asian tectonic domain and the Binhai-Pacific tectonic domain, specifically belonging to the Jiamusi-Xingkai micro-block and its southern margin. The geological strata from the Middle and Late Proterozoic eras to the Paleozoic era have undergone varying degrees of regional metamorphism and tectonic deformation. This has resulted in the formation of different types of folds and ductile deformation zones, accompanied by activities of magma intrusion. The concentration of ^{3}H in the geological genesis is very low and can be considered negligible [19]. Thus, by analysing the content of the radioactive nuclide ^{3}H in the water of the Suifen River, it is possible to determine whether the Suifen River is affected by the discharge of contaminated water into the sea, while also assessing radiation hazards.

Materials and methods

Study area

The study area is located in the Dongning section of the Suifen River in the eastern part of Heilongjiang Province, China. The river is approximately 20 km long, with no nuclear facilities nearby. The Suifen River, the main water source of Dongning, is one of the five major river systems in Heilongjiang Province. It has two sources: the southern source is the Dasuifen River, which originates from the Tumen Mountain northwest of Hunchun in Jilin Province; the northern source is the Xiaosuifen River, which originates from the south side of the Shendong Mountain, Taiping Ridge. The Dasuifen River and the Xiaosuifen River converge near Daohe Town, forming the Suifen River which flows northeast through Dongning into Russian Primorsky Krai, and then into the Sea of Japan near Vladivostok. Each year, a large number of chum salmon from the North Pacific migrate to the Suifen River to spawn.

Collection and preparation of samples

In this study, due to the narrow width of the Suifen River (less than 10 m), sampling was conducted at the center of the river. To obtain a comprehensive understanding of the distribution of tritium content in the Suifen River, a total of 15 sampling points were established at approximately 1.5 km intervals along the river. The sampling took place in August and October 2023, corresponding to the pre- and post-salmon migration period. The locations of the sampling sites are shown in Fig. 1. The sample numbers from right to left in Fig. 1 are sequentially labelled as R1–R15. Water samples were stored in brown glass bottles, which were pre-washed with hydrochloric acid and then rinsed with distilled water. Prior to sampling, the sampling equipment was thoroughly cleaned. During sampling, the equipment was washed three times with the water to be sampled before collection began. Then, about 1 L of surface water was collected from 0.5 m below the water surface into the polyethylene plastic barrel containers and sealed. When immersing the sampler into the water, orient the opening towards the upstream direction to prevent disturbance of the water and entry of debris. First, use the sampler to collect the water, and then transfer it into a container to prevent contamination of the outer surface of the container. After sample collection, record the location information of the sampling points.

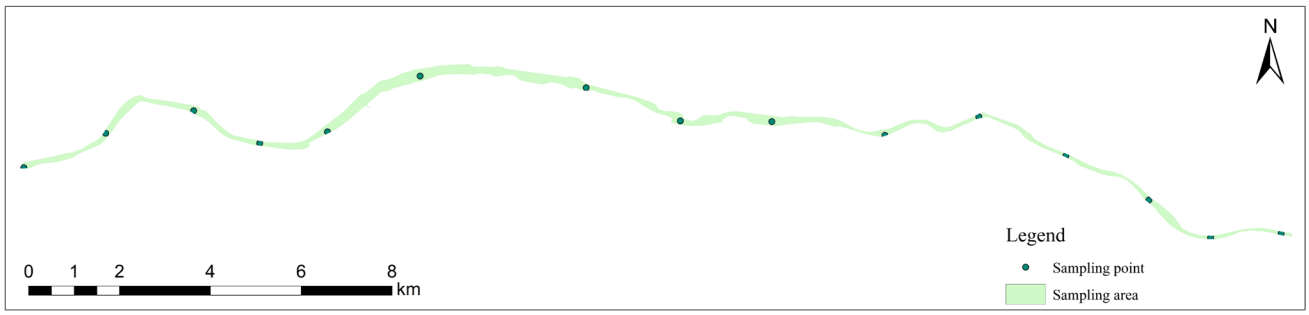

Fig. 1 Distribution of sampling points in surface water

Measurements of radioactivity and calculation of radiation hazard indicators

The analysis of tritium in water was based on the Chinese National Standard "HJ 1126–2020 Analysis method for tritium in water" [20]. A 300 mL water sample was placed into a round-bottom flask, with about 0.5 g of potassium permanganate added, and then subjected to atmospheric pressure distillation. Approximately 150–200 mL of distillate with an electrical conductivity lower than 10 μS cm^{-1} was collected, discarding the first 50 mL of distillate and collecting the middle portion. After distillation, 8 mL of the distillate and Ultima Gold™ LLT scintillator (Perkin Elmer, USA) were placed into a plastic liquid scintillation vial, with the sample-to-scintillator ratio being 8:12 mL. The prepared samples were then analysed for tritium concentration using a SIM-MAX LSA3000 (SIM, China) ultra low-level liquid scintillation spectrometer (LSS). A standard tritium solution (971 ± 27 Bq g^{-1}, reference date: October 10, 2022), calibrated by the National Institute of Metrology, China, was used to determine the counting efficiency. The prepared samples were placed in the ultralow-level liquid scintillation spectrometer for dark adaptation for over 12 h and measured for 1000 min.

The counting efficiency of the instrument for tritium is calculated based on Eq. (1) [20].

$$E = \frac{N_s - N_b}{0.06 \times m \times C_s} \quad (1)$$

where E is the counting efficiency of the liquid scintillation for tritium, N_s is the counting rate of the standard tritium solution (min^{-1}), N_b is the counting rate of the background solution (min^{-1}), m is the mass of the sample (g), C_s is the activity concentration of the standard tritium solution (Bq L^{-1}) and 0.06 is the conversion coefficient (0.001 L $g^{-1} \times 60$ min^{-1} Bq^{-1}).

The tritium activity concentration in the water samples is calculated according to Eq. (2) [20].

$$C = \frac{N_c - N_b}{0.06 \times m \times E} \quad (2)$$

where C is the activity concentration of tritium in the water sample (Bq L^{-1}), and N_c is the counting rate of the water sample (min^{-1}).

Estimation of internal dose of water

As the Suifen River serves as a source of drinking water for the local area, it is necessary to assess the internal dose after ingestion of the water. The annual effective dose (AED) for tritium was calculated according to Eq. (3) [21].

$$D = D_{W1} \times D_{CF} \times Y \times C \quad (3)$$

where D is the annual effective dose from the ingestion of tritium in the water (Sv), D_{W1} is the daily consumption of water (the value was estimated to be 2 L d^{-1}) [22], D_{CF} is the dose conversion factor of ^3H for adults (1.8×10^{-11} Sv Bq^{-1}) [12, 23], Y is the ingestion period (365 d), and C is the tritium activity concentration in the water sample (Bq L^{-1}).

Geo-spatial analysis

Using ArcGIS 10.6, the spatial distribution map of radionuclides within the water of the study zone was created through the application of the Inverse Distance Weighted (IDW) technique. This approach facilitated spatial analysis on the generated maps. The study evaluated the distribution zone for ^3H concentrations. IDW interpolation, a widely adopted method for variable mapping, is recognized for its exactness and ability to conform to continuous spatial variation models. It employs a weighted linear combination of sample points, leveraging statistical and mathematical methodologies to generate surfaces and forecast values at points where measurements are not available [24]. The IDW interpolation relies on a fundamental equation as follows:

$$Z(x_0) = \sum_{i=1}^{n} z(x_i) \cdot d_{ij}^{-p} / \sum_{i=1}^{n} d_{ij}^{-p} \quad (4)$$

where Z represents the interpolated value at a specific grid node, while z_i refers to the values of neighbouring data points and d_{ij0} denotes the distances between the grid node and these data points.

The IDW becomes feasible by moving data into the GIS context. Consequently, analyses can be carried out in regions lacking data. This makes IDW particularly suited for integration into a GIS environment, enabling analysis in areas where data might be sparse or completely absent, by utilizing the spatial relationships and distances between known data points to estimate values at unknown locations.

Statistical analysis

IBM SPSS Statistics, a robust statistical analysis software developed by IBM, caters to a wide range of functions including data management, advanced analytics, multivariate analysis, business intelligence and applications in criminal investigations. The analysis involving multiple variables was performed with IBM SPSS version 23, leading to the creation of box plots derived from experimental data.

Results and discussion

Activity concentration and annual effective dose

Table 1 displays the ^{3}H activity concentration and annual effective dose in the surface water of Suifenhe for August and October. Figures 2 and 3 illustrate the spatial distribution of ^{3}H in surface water for these months, respectively. The range of ^{3}H activity concentration in surface water was 5.08–9.75 Bq L^{-1}, with an average of 6.92 Bq L^{-1}, which is below the WHO recommended limit of 10,000 Bq L^{-1} for drinking water. The annual effective dose varied from 0.07 to 0.13 μSv, with an average of 0.09 μSv, remaining under the WHO recommended guideline of 0.1 mSv for drinking water [22].

In Table 2, the ^{3}H activity concentrations of surface water samples were compared with the those of other researchers in different regions around the world [25–40]. In this study, the ^{3}H activity concentration in surface water was notably higher than that in countries like France, Romania and Greece. However, it aligns with the findings of Ren et al. in China, falling within the normal range for northern China. The variance can be attributed to the study area's location in a high-latitude region, higher than that of France, Romania and Greece. The production of natural tritium in the atmosphere shows an increase with latitude. This latitudinal dependency on the production rate, coupled with the enhanced transfer of tritium from the stratosphere to the troposphere at higher latitudes, leads to a non-uniform distribution of natural tritium in precipitation [41]. Moreover, the release points of anthropogenic tritium into the atmosphere (nuclear test sites and nuclear power plants) are predominantly situated in the mid to high latitudes of the Northern Hemisphere [40, 42]. The intensity of cosmic ray neutrons also increases with latitude, thereby raising the levels of cosmogenic tritium. Additionally, within the same latitude, moving from the coast towards inland, the tritium content in precipitation increases with distance from the coastline, known as the continental effect. This effect is primarily due to two reasons: coastal areas experience high rainfall and moisture levels, leading to greater dilution of tritium by tritium-poor ocean water; and as water masses move from the coast inland, the longer distance allows for more extensive isotopic exchange opportunities than in coastal areas, gradually increasing tritium content inland. Therefore, the tritium content in the surface water within this study area is comparatively higher than in other countries and regions [43]. However, when compared to Russia [27] and Germany [37], the tritium content in the rivers is significantly lower. This is because the two rivers in Russia and Germany have nuclear power facilities in their vicinity, which leads to an increase in tritium content.

Table 1 ^{3}H activity concentration and annual effective dose in surface water

Sample	^{3}H activity concentration (Bq L^{-1})		Annual effective dose (μSv)	
	August	October	August	October
1	5.27 ± 0.21	7.11 ± 0.17	0.07	0.09
2	8.04 ± 0.15	6.19 ± 0.19	0.11	0.08
3	7.02 ± 0.15	7.94 ± 0.17	0.09	0.10
4	7.21 ± 0.19	6.19 ± 0.17	0.09	0.08
5	6.47 ± 0.18	6.65 ± 0.18	0.09	0.09
6	7.67 ± 0.16	7.39 ± 0.16	0.10	0.10
7	6.00 ± 0.16	7.48 ± 0.16	0.08	0.10
8	7.48 ± 0.17	7.85 ± 0.16	0.10	0.10
9	7.30 ± 0.18	6.28 ± 0.16	0.10	0.08
10	6.37 ± 0.16	7.67 ± 0.18	0.08	0.10
11	7.57 ± 0.16	5.91 ± 0.19	0.10	0.08
12	5.08 ± 0.19	6.74 ± 0.22	0.07	0.09
13	5.17 ± 0.18	6.56 ± 0.22	0.07	0.09
14	6.28 ± 0.17	6.84 ± 0.16	0.08	0.09
15	8.13 ± 0.15	9.75 ± 0.20	0.11	0.13
Min	5.08	5.91	0.07	0.08
Max	8.13	9.75	0.11	0.13
Ave	6.73	7.10	0.09	0.09

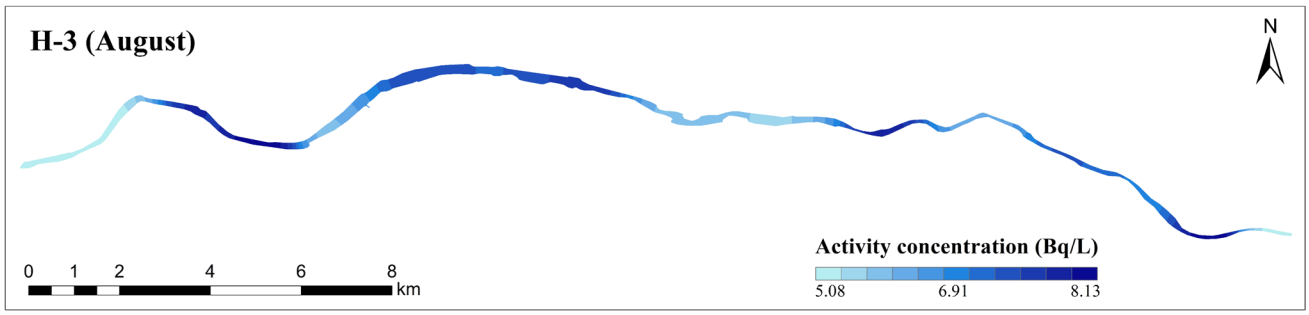

Fig. 2 Spatial distribution map of ^{3}H activity concentration in surface water for August

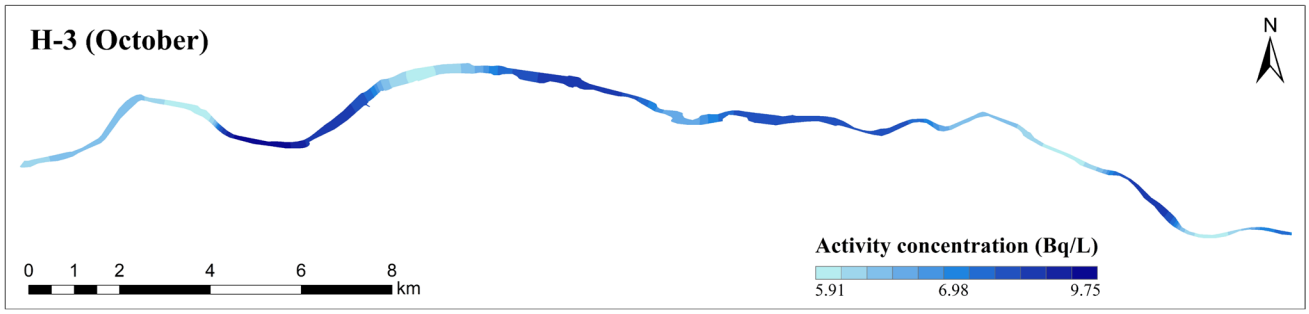

Fig. 3 Spatial distribution map of ^{3}H activity concentration in surface water for October

Table 2 Surface water activity concentration (Bq L^{-1}) in different regions around the world

Country	^{3}H (Bq L^{-1})		References
	Range	Average	
France	0.12–0.86	0.41	[25]
Russia	0.56–3.49	1.43	[26]
Russia	13–26	–	[27]
India	1.9–42.1	4.0	[28]
Turkey	1.606–4.417	2.493	[29]
Turkey	2.45–3.17	–	[30]
Japan	0.36–2.66	1.06	[31]
Romania	0.83–1.45	–	[32]
Greece	0.82–1.03	0.94	[33]
Spain	0.71–6.44	3.6	[34]
Iraq	–	0.562	[35]
Poland	0.43–0.94	0.74	[36]
Germany	–	32.4	[37]
China (Heilongjiang)	14.9–17.8	–	[38]
China (Liaoning)	–	14.4	[38]
China (Gansu)	–	7.55	[39]
China (Nei Mongol)	–	6.13	[39]
China (Nei Mongol)	1.57–2.62	2.03	[40]
China (Xinjiang)	–	9.25	[39]
China (Qinghai)	–	9.37	[39]
China	5.08–9.75	6.92	(Present work)

Seasonal variation of concentration in surface water

To investigate if the tritium content in the Suifen River is influenced by marine nuclear pollution, water samples were collected during two periods: before and after the migration of chum salmon, specifically in August and October. The variation of tritium content in various environmental waters across different regions also changes with the seasons. In China, the tritium content in precipitation is higher in winter and spring, and lower in summer and autumn [44, 45]. Figure 4 presents the box plot of ^{3}H activity concentration in surface water samples for August and October, showing that the concentration in August is lower than in October. This observation could be attributed to the increased precipitation in the Suifen River basin during this period, with the continuous influence of precipitation and monsoon providing a plausible explanation for the rapid dilution patterns of tritium [46, 47]. On the other hand, chum salmon migrate from the ocean to the Suifen River for spawning and die every year between September and October [48, 49]. During their 3–5 years in the ocean, chum salmon may absorb and transform tritium from nuclear-polluted water discharged into the ocean, which is then released into freshwater when they spawn and die in the Suifen River [7–9]. After the Fukushima Daiichi nuclear disaster, Arai conducted research on salmonids, which are migratory, and found

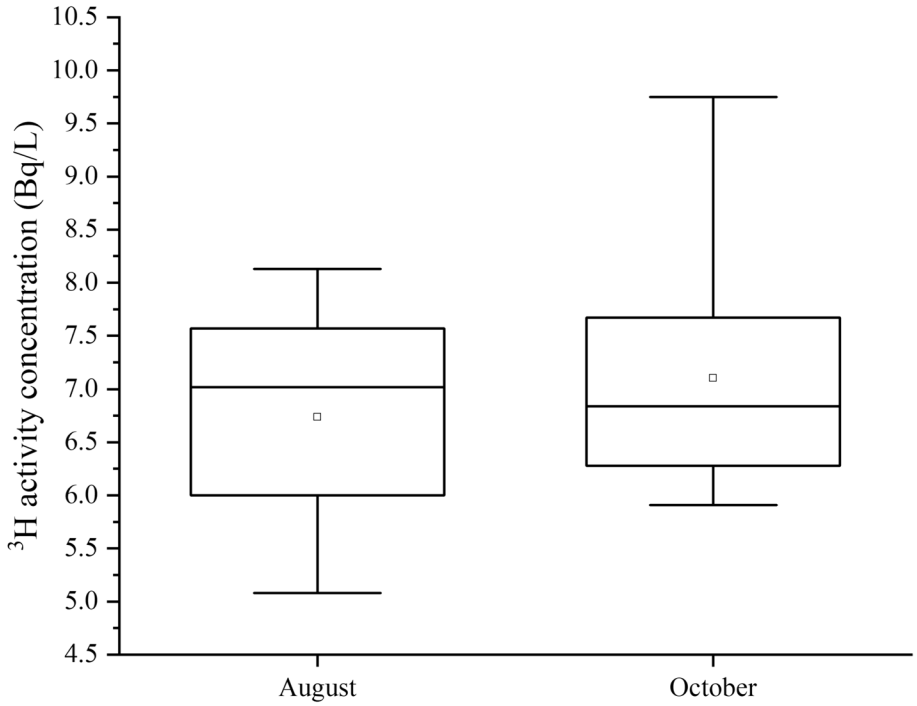

Fig. 4 Box plot of ^{3}H activity concentration in surface water for August and October

that they accumulate radioactive nuclides such as ^{137}Cs and ^{134}Cs, migrating these substances between freshwater and the ocean [17]. Similarly, Madigan et al. sampled and analysed Pacific Thunnus orientalis, revealing that these fish could rapidly transport radioactive nuclides from the Fukushima accident across the entire North Pacific [50]. Thus, the higher ^{3}H activity concentration in the Suifen River in October compared to August could also be related to the migration of chum salmon. However, statistical analysis (paired *t* test) of the ^{3}H activity concentration in surface water samples from August and October showed no significant difference between the two periods ($p > 0.05$, at the 95% significance level). Therefore, further research focusing on the Suifen River and chum salmon is required, involving the collection and analysis of a large number of water and fish samples for tritium content.

Conclusion

The analysis of ^{3}H activity concentration and annual effective dose in the Suifen River, China, indicates that both metrics are below the guidelines provided by the World Health Organization (WHO). The ^{3}H activity concentration falls within the normal range for northern China. It was observed that the ^{3}H activity concentration in the Suifen River before the migration of chum salmon is lower than after, meaning the concentration of ^{3}H in surface water samples is higher in October than in August. This variation could be attributed to climate factors and possibly the influence of nuclear pollution water discharge, although statistical analysis showed no significant difference between the two periods. Future research should thus focus on the mechanism of tritium migration and transformation by chum salmon. The findings from this study will serve as foundational data for evaluating the dose impact on the regions concerned in the event of an unforeseen tritium release.

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Data availability The data that support the findings of this study are available from the corresponding author, [Hongtao Zhao], upon reasonable request.

Declarations

Conflict of interest The author has no competing interests to declare that are relevant to the content of this article.

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