

## Determination and human health risk assessment of TFWT, OBT and carbon-14 in seafood around Qinshan Nuclear Power Plant

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### ABSTRACT

This work aims to evaluate the effects of the operation of Qinshan nuclear Power Plant (QNPP) on tritium (<sup>3</sup>H) and carbon-14 (<sup>14</sup>C) levels in seafood and assess the health risks caused by seafood consumption. Five kinds of seafood, including marine fish, prawn, razor clam, crabs, and seaweed, were collected from QNPP and the sea around Hangzhou Bay. The activity concentrations of tissue free water tritium (TFWT), organically bound tritium (OBT) and <sup>14</sup>C were determined, respectively, and the annual intake and annual effective dose (AED) were calculated. The results showed that the TFWT, OBT, and <sup>14</sup>C activity concentrations of the seafood in the surrounding area of QNPP ranged from 2.00 to 74.75 Bq/L, <1.04 to 19.68 Bq/L and 0.09 to 0.17 Bq/g-C, respectively. The TFWT, OBT, and <sup>14</sup>C activity concentrations of the seafood in Hangzhou Bay ranged from 1.36 to 10.55 Bq/L, 1.08 to 6.78 Bq/L and 0.07 to 0.13 Bq/g-C, respectively. The differences were not statistically significant. The total AED from <sup>3</sup>H and <sup>14</sup>C due to the seafood consumption for the residents in the surrounding of QNPP and Hangzhou Bay were  $1.96 \times 10^{-4}$  and  $1.61 \times 10^{-4}$  mSv/year, respectively. The results showed that the operation of QNPP had no obvious effect on <sup>3</sup>H and <sup>14</sup>C accumulation in seafood, and the dose burden of population was low.

### 1. Introduction

Tritium (<sup>3</sup>H) and carbon-14 (<sup>14</sup>C) are two low-energy beta-emitters with half-lives of 12.3 and 5730 years, respectively. These are two important radionuclides produced primarily through cosmic-ray neutron reaction in the atmosphere, nuclear weapon testing, nuclear accidents, and the operation of nuclear facilities (Yim et al., 2006; Eyrolle et al., 2018; Hamlat et al., 2018). Nuclear power plants worldwide produce approximately  $1.1 \times 10^{15}$  Bq/a <sup>14</sup>C, of which all operating nuclear power plants emit approximately  $1.1 \times 10^{14}$  Bq/a <sup>14</sup>C into the atmosphere in gaseous form, and reprocessing plants emit approximately  $3.7 \times 10^{14}$  Bq/a <sup>14</sup>C into the atmosphere in both gaseous and liquid forms (Charles, 2001). Given the development of nuclear energy, the amount of <sup>3</sup>H and <sup>14</sup>C released into the environment may increase (Guo et al., 2020; Wu et al., 2020).

<sup>3</sup>H is found all over the biosphere. It can exist as a gas (HT) or in the

form of tritium water (HTO). <sup>3</sup>H can be absorbed into human body via various routes, including inhalation, dermal penetration, and ingestion, and participate in the anabolism of organic matter in living organisms, forming organically bound tritium (OBT). People living near nuclear power plants may be exposed to internal irradiation hazards if they consume tritium-contaminated food. Tritium irradiation may cause genetic variation and carcinogenic effects (Hagger et al., 2005).

<sup>14</sup>C is primarily released as gases and liquids through wastewater pipelines and chimneys. While the <sup>14</sup>C concentrations in reactor water sampled during the operation were reported ranging within 0.1–2 MBq/L, those taken during shutdown and in groundwater near a waste management area ranged from –100 to 1800 Bq/L (Caron et al., 2000). <sup>14</sup>C, which exists as carbon dioxide (CO<sub>2</sub>), mixes with non-radioactive CO<sub>2</sub> in the air, participates in the photosynthesis of plants, enters into the food chain, and endangers the environment and human body. According to the Canadian experience, 99 % of the <sup>14</sup>C intake is by the

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human body. As the half-life of  $^{14}\text{C}$  is very long, its contribution to the global collective dose burden cannot be ignored.

The QNPP is 8 km southeast of Wuyuan Town in Haiyan County, Zhejiang Province, at the foot of the Qinshan Mountains. It is approximately 90 km, 30 km, and 80 km straight away from Shanghai, Jiaying, and Hangzhou, respectively. The QNPP is China's first nuclear power plant and commenced commercial operation in December 1991. QNPP is China's only nuclear power plant with a heavy water reactor. Notably, heavy water reactors produce more  $^3\text{H}$  and  $^{14}\text{C}$  when operating than other reactors.

The Japanese government began dumping contaminated radioactive wastewater from the Fukushima Daiichi Nuclear Power Plant (FDNPP) into the ocean in 2023. Although most radionuclides in the water have been treated by an Advanced Liquid Processing System (ALPS), the water still contains radionuclides whose half-lives are comparatively long, such as  $^3\text{H}$  and  $^{14}\text{C}$  (Zhao et al., 2021).  $^3\text{H}$  and  $^{14}\text{C}$  have been rapidly incorporated into marine organisms either by uptake from seawater or by food ingestion (Alava et al., 2016; Buesseler, 2012; de With et al., 2021; Madigan et al., 2012). The contaminants from FDNPP accident contaminated will reach China's southeastern coastal waters after a certain period of time, gradually spread to the East China Sea and the Bohai Sea, affecting the seafood in China's sea areas (Liu et al., 2022). Thus, it is crucial to carry out  $^3\text{H}$  and  $^{14}\text{C}$  monitoring for radiation protection.

The national standard (GB4792-1984) sets the public exposure limit at 1 mSv/a (Industrial Hygiene Laboratory). In this study, we measured the concentrations of different seafood species including marine fish, prawn, razor clams, crabs, and seaweed to determine the activity concentrations of tissue free water tritium (TFWT), OBT, and  $^{14}\text{C}$  at various sites in QNPP and Hangzhou Bay. Moreover, we performed an early assessment of the possible harm to human health associated with  $^3\text{H}$  and  $^{14}\text{C}$  exposure from seafood eating. The possible risk to human health from ingesting  $^3\text{H}$  and  $^{14}\text{C}$  in seafood was assessed by calculating the annual radiation dose and annual effective dose (AED).

## 2. Material and methods

### 2.1. Sample collection

In this study, we conducted two sampling campaigns in April and October 2022, by purchasing five dead seafood samples from the surrounding of QNPP and commercial trawlers around Hangzhou Bay. The samples were stored and transported in a frozen state. Specifically, samples purchased from QNPP surrounding include the mullet, prawn, razor clam, sea carb, and seaweed. Samples purchased around the Hangzhou Bay included scaly hairpin anchovy or bombay duck, prawn, razor clam, potunide, and seaweed. After collection, the marine fish, prawns and sea crabs were washed quickly with purified water. The razor clams were washed thoroughly with purified water, and the soft parts of the bodies were collected. Seaweeds were collected, cut with a knife to remove the inedible parts (roots and rotten parts), and washed with purified water to remove the residual sediments. The treated sample was dried at 50 °C for 10–15 min, then minced and mixed well. The edible part of the sample after pretreatment was more than 1 kg. A 3–8 g mixed sample was taken in the sample tray of the moisture analyzer, the temperature of the moisture analyzer was set at 105 °C, and the mass fraction of tissue free water in the fresh sample was measured. The detailed sampling information is shown in Table S1 and Fig. S1.

### 2.2. Sample treatment for the measurement of TFWT, OBT and $^{14}\text{C}$

Marine fish, prawn, razor clam, crabs, and seaweed were frozen at  $-20\text{ }^\circ\text{C}$  for 48 h, and then processed using a vacuum freeze dryer (Labconco, 4 L,  $-105\text{ }^\circ\text{C}$ ). To extract all of the tissue free water at vacuum pressures of  $< 10^{-2}$  mbar, the system was operated for 2–3 days.

The free water in the sample was separated by vacuum freeze-drying. The free water sample was distilled, and 20 mL of distillate was collected. For each sample, the  $^3\text{H}$  activity concentration of the free water was determined using liquid scintillation counter (LSC, Quantulus 1220, PerkinElmer).

To measure OBT and  $^{14}\text{C}$  activity concentrations of seafood, the sample treatment was undertaken by loading 50 g of freeze-dried sample into a tube furnace combustion system for oxidation combustion. Combustion products of water ( $\text{H}_2\text{O}$ ) and  $\text{CO}_2$  were collected using a cold trap ( $-110\text{ }^\circ\text{C}$ ) and 3 mol/L sodium hydroxide solution (NaOH), respectively. The  $\text{H}_2\text{O}$  was refluxed with potassium permanganate for 2 h, followed by distillation, and finally 20 mL of distillate was collected. The pH of NaOH solution for absorbing  $\text{CO}_2$  was adjusted to 10–11 with ammonium chloride ( $\text{NH}_4\text{Cl}$ ), and calcium chloride solution ( $\text{CaCl}_2$ ) was added to obtain white calcium carbonate ( $\text{CaCO}_3$ ) precipitate. The white precipitate was filtered and washed with deionized water and absolute ethanol, then dried at 105 °C in an oven.

### 2.3. Measurement of the activity concentration of OBT and $^{14}\text{C}$

#### 2.3.1. Tritium

In a 20 mL polyethylene vial, 8 g purified  $^3\text{H}$  fraction ( $^3\text{H}$  water) and 12 mL liquid scintillation cocktail (Ultima Gold LLT, PerkinElmer) were thoroughly mixed. After sitting for 12 h (Feng et al., 2020; Li et al., 2020; Nikolov et al., 2013) in the LSC under dark conditions,  $^3\text{H}$  in the samples was measured for 1000 min. The method detection limit (MDA) for  $^3\text{H}$  was 1.04 Bq/L.

#### 2.3.2. $^{14}\text{C}$

2 g of  $\text{CaCO}_3$  precipitates, 14 mL of scintillation cocktail (Optiphase Hisafe 3, PerkinElmer), and 4 mL of deionized water were transferred to a 20 mL polyethylene vial and thoroughly mixed. After sitting for 2 h in the LSC under dark conditions,  $^{14}\text{C}$  in the samples was measured for 300 min. The MDA of  $^{14}\text{C}$  was 0.09 Bq/g.C.

### 2.4. Activity concentration

The following equation was used to obtain the TFWT activity in each sample:

$$A_{TFWT} = \frac{(N_{TFWT} - N_b) \times \omega}{60 \times m_H \times E_H} \times 1000$$

The following equation was used to obtain the OBT activity in each sample:

$$A_{OBT} = \frac{(N_{OBT} - N_b) \times m_{OBT} \times (1 - \omega)}{60 \times m_H \times E_H \times Y_H \times M} \times 1000$$

where  $A_{TFWT}$  and  $A_{OBT}$  are the activity concentration of TFWT and OBT in the samples (Bq/kg-fresh),  $N_{TFWT}$  and  $N_{OBT}$  are the count rates of TFWT and OBT in the analyzed sample ( $\text{min}^{-1}$ ),  $N_b$  is the count rate of the background sample ( $\text{min}^{-1}$ ),  $\omega$  is the moisture content of the sample (%),  $m_{OBT}$  is the amount of water produced by the combustion of the dry sample (g),  $M$  is the amount of dry sample processed in the combustion device (g),  $m_H$  is the amount of the sample contained in the counting vial (g),  $E_H$  is the counting efficiency of LSC for  $^3\text{H}$  (%), and  $Y_H$  is the recovery of hydrogen during the combustion process (%).

The following equation was used to obtain the  $^{14}\text{C}$  activity in each sample:

$$A_C = \frac{(N_C - N_0) \times m_{c\text{CO}_3} \times (1 - \omega)}{60 \times m_C \times E_C \times Y_C \times M} \times 1000$$

where  $A_C$  is the activity concentration of  $^{14}\text{C}$  in the samples (Bq/kg-fresh),  $N_C$  is the count rate of  $^{14}\text{C}$  in the analyzed sample ( $\text{min}^{-1}$ ),  $N_0$  is the count rate of the background sample ( $\text{min}^{-1}$ ),  $m_C$  is the amount of dry sample processed in the combustion device (g),  $m_{c\text{CO}_3}$  is the amount

**Table 1**  
Comparisons of TFWT activities concentrations in seafood from QNPP and Hangzhou Bay.

QNPP	Activity concentration of $^3\text{H}$ in TFWT (Bq/L)	Activity concentration of TFWT in the original sample (Bq/kg-fresh)	Hangzhou Bay	Activity concentration of $^3\text{H}$ in TFWT (Bq/L)	Activity concentration of TFWT in the original sample (Bq/kg-fresh)	Mann-Whitney U test	
						Z	p
Mullets <sup>1</sup>	4.86	3.35	Scaly hairfin anchovies	3.87	2.45	-1.16	0.28
Mullets <sup>2</sup>	2.00	1.46	Bombay ducks	8.28	6.91		
Prawn <sup>1</sup>	2.32	1.74	Prawn <sup>1</sup>	1.36	1.01		
Prawn <sup>2</sup>	9.64	8.14	Prawn <sup>2</sup>	6.71	5.43		
Razor clam <sup>1</sup>	7.41	5.30	Razor clam <sup>1</sup>	1.62	1.19		
Razor clam <sup>2</sup>	4.50	3.90	Razor clam <sup>2</sup>	2.49	2.15		
Sea crab <sup>1</sup>	17.10	8.81	Portunid <sup>1</sup>	3.08	1.99		
Sea crab <sup>2</sup>	74.75	52.96	Portunid <sup>2</sup>	10.55	8.11		
Seaweeds	4.70	2.63	Seaweeds	3.56	1.99		

Note: superscripts 1 and 2 are the results of samples taken at two different times.

of  $\text{CaCO}_3$  produced by combustion of the dry sample (g),  $m_c$  is the amount of the sample contained in the counting vial (g),  $E_C$  is the counting efficiency of LSC for  $^{14}\text{C}$  (%),  $Y_C$  is the recovery of C during the combustion process (%).

## 2.5. Dose calculation

The following equation was used to obtain the annual intake:

$$I_i = C_i \times M_i$$

where  $I_i$  is the annual radioactive dose (Bq/year) (Yu et al., 2019),  $C_i$  is the activity concentrations of TFWT, OBt or  $^{14}\text{C}$  (Bq/kg), and  $M_i$  is the annual consumption of each type of seafood by the inhabitants (kg/year). The annual consumption is shown in Table S1.

For specific environmental transfer pathways (foodborne), the final effective dose can be estimated by multiplying the radionuclide conversion coefficient by human intake. The dose conversion coefficient is radionuclide-specific, and the work was conducted according to the dose factors given by the International Commission on Radiological Protection (ICRP).

The following equation was used to obtain the AED:

$$H_D = D \times I_i$$

where  $H_D$  is the AED (mSv/year),  $D$  is the dose conversion coefficient (General Administration of Quality Supervision, 2002) (mSv/Bq, HTO is  $1.8 \times 10^{-8}$ , OBt is  $4.2 \times 10^{-8}$ , and  $^{14}\text{C}$  is  $5.8 \times 10^{-7}$ ),  $I_i$  is the annual intake (Bq/year). The formula for calculating the ratio of AED to annual dose constraint is:

$$p = \sum (H_D) \div 0.25 \times 100\%$$

where  $p$  is the proportion to the annual dose constraint (%),  $H_D$  is the total AED to be accumulated in the year (mSv/year), 0.25 is the standard annual dose constraint (mSv) (Ministry of Environmental Protection, 2011).

The radiation-induced health risks to the population were evaluated with the AED from the intake of radionuclides in the seafood samples.

## 2.6. Statistics

Statistical analyses were carried out to describe and comprehend the statistical properties of the measured radioactive concentration levels. The t-test and Mann-Whitney U test were performed using SPSS 25.0 software. For all statistical evaluations, a two-tailed  $p < 0.05$  was considered significant. If the data is lower than the detection limit of the analysis method, the MDA value is substituted for the data. Use R language (4.2.2) for cluster analysis and draw heatmap.

## 3. Results and discussion

### 3.1. TFWT and OBt activity concentrations

The results are shown in Table 1. The activity concentration of TFWT in seafood around QNPP and Hangzhou Bay ranged from 2.00 to 74.75 Bq/L and 1.36 to 10.55 Bq/L, respectively. The activity concentration of fresh samples ranged from 1.46 to 52.96 Bq/kg-fresh and 1.01 to 8.11 Bq/kg-fresh, depending on moisture content. The activity concentration

**Table 2**  
Comparisons of OBt activities concentrations in seafood from QNPP and Hangzhou Bay.

QNPP	Activity concentration of $^3\text{H}$ in OBt (Bq/L)	Activity concentration of OBt in the original sample (Bq/kg-fresh)	Hangzhou Bay	Activity concentration of $^3\text{H}$ in OBt (Bq/L)	Activity concentration of OBt in the original sample (Bq/kg-fresh)	Mann-Whitney U test	
						Z	p
Mullets <sup>1</sup>	2.31	0.54	Scaly hairfin anchovies	1.08	0.23	-1.01	0.35
Mullets <sup>2</sup>	< 1.04	< 0.15	Bombay ducks	1.57	0.14		
Prawn <sup>1</sup>	3.77	0.46	Prawn <sup>1</sup>	2.35	0.35		
Prawn <sup>2</sup>	1.60	0.13	Prawn <sup>2</sup>	1.29	0.13		
Razor clam <sup>1</sup>	3.44	0.51	Razor clam <sup>1</sup>	1.43	0.22		
Razor clam <sup>2</sup>	5.54	0.45	Razor clam <sup>2</sup>	6.78	0.47		
Sea crab <sup>1</sup>	13.44	2.43	Portunid <sup>1</sup>	1.77	0.31		
Sea crab <sup>2</sup>	19.68	2.58	Portunid <sup>2</sup>	4.24	0.51		
Seaweeds	4.83	0.42	Seaweeds	4.32	0.40		

Note: superscripts 1 and 2 are the results of samples taken at different times.

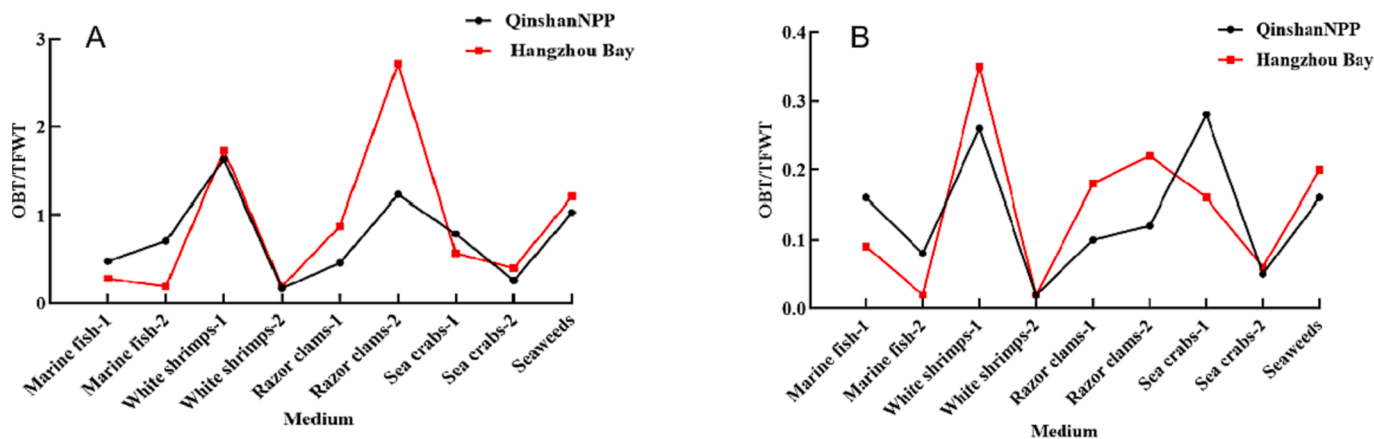


Fig. 1. A (Bq/L) and B (Bq/kg-fresh) of OB/T/TFWT. Note: The superscript 1 and 2 are the results of the samples taken at two different times.

of TFWT in seafood around QNPP is higher than in Hangzhou Bay. Despite these differences, no statistical difference in TFWT activity concentration was detected in seafood among sample sites ( $Z = -1.16$ ,  $p = 0.28$ ).

The activity concentration of OB/T in seafood around QNPP and Hangzhou Bay ranged from  $<1.04$  to  $19.68$  Bq/L and  $1.08$  to  $6.78$  Bq/L, respectively (Table 2). The activity concentration of fresh samples based on water content ranged from  $0.13$  to  $2.58$  Bq/kg-fresh and  $0.13$  to  $0.51$  Bq/kg-fresh. The activity concentration of OB/T in seafood around QNPP was higher than in Hangzhou Bay. However, by the Mann-Whitney U test, there was no evidence of a statistically significant difference in OB/T results ( $Z = -1.01$ ,  $p = 0.35$ ).

The activity concentrations of TFWT and OB/T of sea crabs from the QNPP were higher than those of other seafood ( $<10$  Bq/L). This may be explained by the distance between the sampling site of sea crabs and the QNPP being  $< 1$  km and sea crabs ingesting wastewater containing  $^3\text{H}$  discharged from the NPP.

The specific results of TFWT and OB/T activity concentrations in seafood around QNPP and Hangzhou Bay are shown in Tables 1 and 2. These values are significantly lower than the reference values ( $6.5 \times 10^5$  Bq/kg) recommended by the Ministry of Health of the People's Republic of China (Chinese Standards, 1994) (Ministry Of Health, 1992). During 2014–2016, the HTO concentration in mullet at the QNPP ranged from  $2.2$  to  $2.7$  Bq/kg (Yang et al., 2020). The radiation environment monitoring station of Zhejiang Province detected the activity concentrations of  $^3\text{H}$  in marine fish and jellyfish around QNPP and Hangzhou Bay, and the TFWT results were lower than the MDA of  $1.2$  Bq/kg (Liang et al., 2009). Moreover, the OB/T activity of marine biota captured near the Fangchenggang NPP in the Qingzhou Bay ranged from  $1.88$  to  $12.9$

Bq/L (Lin et al., 2020). Furthermore, the findings of this investigation and the data in the cited literatures are congruent, indicating that  $^3\text{H}$  activity concentrations around NPP are at background levels.

### 3.2. OB/T and TFWT ratios in seafood

OB/T is a bioaccumulative organic pollutant that is persistent (Jaeschke et al., 2011). It was calculated based on total OB/T and TFWT activity concentration. As shown in Fig. 1 (A), the mean OB/T/TFWT for the five types of seafood ranged from  $0.17$  to  $2.72$ . The average values for each food species followed the order of razor clam  $>$  seaweed  $>$  prawn  $>$  sea crab  $>$  marine fish. OB/T has a longer half-life in organisms than TFWT, and its metabolism in organisms is slow (Jaeschke et al., 2013). One reason is that marine fish are more mobile and can spend their lifetime in areas where the  $^3\text{H}$  levels may differ (Kim et al., 2019). The  $^3\text{H}$  levels in their diet at their habitat region may also affect their tissue OB/T contents (McCubbin et al., 2001).

### 3.3. $^{14}\text{C}$ activity concentrations

Table 3 shows the  $^{14}\text{C}$  measurement results. The concentration of  $^{14}\text{C}$  activity in seafood around QNPP and Hangzhou Bay ranged from  $0.09$  to  $0.17$  Bq/g-C and  $0.07$  to  $0.13$  Bq/g-C, respectively. The activity concentration of fresh samples ranged from  $6.84$  to  $15.95$  Bq/kg-fresh and  $4.60$  to  $17.77$  Bq/kg-fresh, with no significant difference between the two places ( $t = 1.02$ ,  $p = 0.81$ ). The results demonstrated that the wastewater containing  $^{14}\text{C}$  discharged from nuclear power plants has a minor effect on the activity concentration of  $^{14}\text{C}$  in seafood. The  $^{14}\text{C}$  activity concentrations observed in this study were found to be similar as

Table 3

Comparison of  $^{14}\text{C}$  activity concentration in seafood from QNPP and Hangzhou Bay.

QNPP	Activity concentration of $^{14}\text{C}$ (Bq/g-C)	Activity concentration of $^{14}\text{C}$ in the original sample (Bq/kg-fresh)	Hangzhou Bay	Activity concentration of $^{14}\text{C}$ (Bq/g-C)	Activity concentration of $^{14}\text{C}$ in the original sample (Bq/kg-fresh)	t-test	
						t	p
Mulletts <sup>1</sup>	0.10	14.11	Scaly hairfin anchovies	0.13	17.77	1.02	0.81
Mulletts <sup>2</sup>	0.13	15.95	Bombay ducks	0.09	6.30		
Prawn <sup>1</sup>	0.09	8.54	Prawn <sup>1</sup>	0.10	10.40		
Prawn <sup>2</sup>	0.14	7.44	Prawn <sup>2</sup>	0.11	8.17		
Razor clam <sup>1</sup>	0.09	11.07	Razor clam <sup>1</sup>	0.11	11.26		
Razor clam <sup>2</sup>	0.15	10.36	Razor clam <sup>2</sup>	0.11	4.76		
Sea crab <sup>1</sup>	0.09	9.58	Portunid <sup>1</sup>	0.10	11.07		
Sea crab <sup>2</sup>	0.17	15.47	Portunid <sup>2</sup>	0.11	8.76		
Seaweeds	0.10	6.84	Seaweeds	0.07	4.60		

Note: superscripts 1 and 2 are the results of samples taken at two different times.

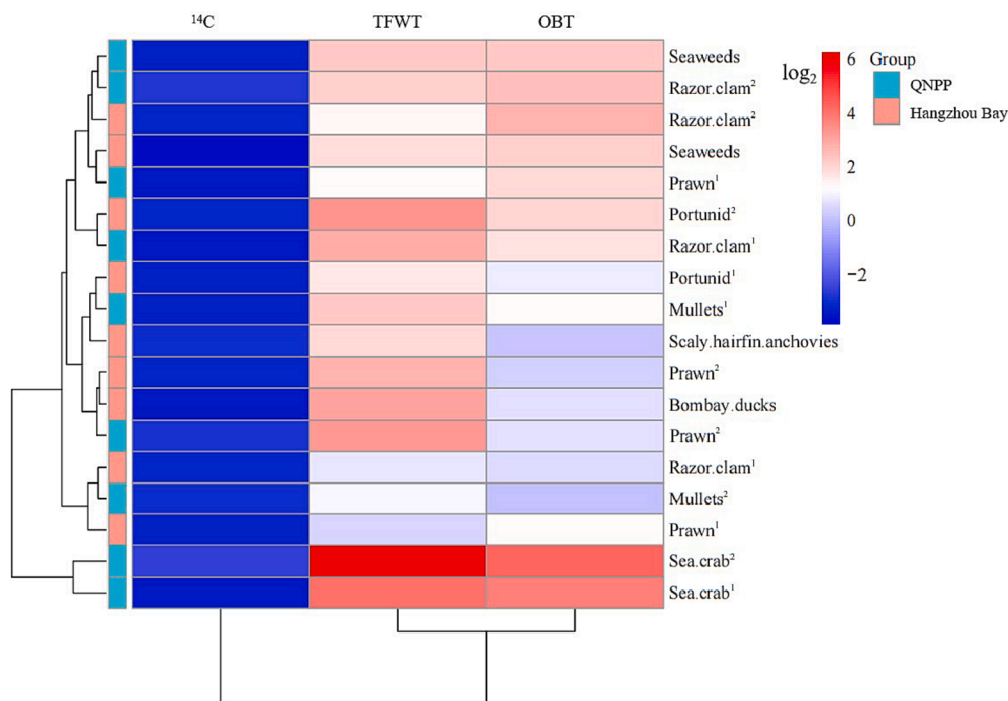


Fig. 2. Systematic clustering heatmap.

Table 4  
AED from <sup>3</sup>H and <sup>14</sup>C due to consumption of seafood.

Monitoring points	Sample type	Seafood annual radioactive dose (Bq)			AED (mSv/year)			Seafood AED(mSv/year)	Total AED (mSv/year)	Annual dose constraint (%)
		TFWT	OBT	<sup>14</sup> C	TFWT	OBT	<sup>14</sup> C			
QNPP	Marine fish	43.85	5.66	246.37	7.89 × 10 <sup>-7</sup>	2.38 × 10 <sup>-7</sup>	1.43 × 10 <sup>-4</sup>	1.44 × 10 <sup>-4</sup>	1.96 × 10 <sup>-4</sup>	0.08
	Marine prawns	20.51	1.22	33.17	3.69 × 10 <sup>-7</sup>	5.14 × 10 <sup>-8</sup>	1.92 × 10 <sup>-5</sup>	1.96 × 10 <sup>-5</sup>		
	Razor clam	3.58	0.37	8.35	6.45 × 10 <sup>-8</sup>	1.57 × 10 <sup>-8</sup>	4.84 × 10 <sup>-6</sup>	4.92 × 10 <sup>-6</sup>		
	Sea crabs	94.32	7.65	38.25	1.70 × 10 <sup>-6</sup>	3.21 × 10 <sup>-7</sup>	2.22 × 10 <sup>-5</sup>	2.42 × 10 <sup>-5</sup>		
	Seaweeds	1.67	0.27	4.34	3.00 × 10 <sup>-7</sup>	1.12 × 10 <sup>-8</sup>	2.52 × 10 <sup>-6</sup>	2.83 × 10 <sup>-6</sup>		
Hangzhou Bay	Marine fish	76.71	3.03	197.28	1.38 × 10 <sup>-6</sup>	1.27 × 10 <sup>-7</sup>	1.14 × 10 <sup>-4</sup>	1.15 × 10 <sup>-4</sup>	1.61 × 10 <sup>-4</sup>	0.06
	Marine prawns	13.37	1.00	38.55	2.41 × 10 <sup>-7</sup>	4.19 × 10 <sup>-8</sup>	2.23 × 10 <sup>-5</sup>	2.26 × 10 <sup>-5</sup>		
	Razor clam	1.30	0.27	6.24	2.34 × 10 <sup>-7</sup>	1.13 × 10 <sup>-8</sup>	3.62 × 10 <sup>-6</sup>	3.87 × 10 <sup>-6</sup>		
	Sea crabs	15.42	1.25	30.28	2.78 × 10 <sup>-7</sup>	5.26 × 10 <sup>-8</sup>	1.76 × 10 <sup>-5</sup>	1.79 × 10 <sup>-5</sup>		
	Seaweeds	1.26	0.25	2.92	2.27 × 10 <sup>-7</sup>	1.07 × 10 <sup>-8</sup>	1.69 × 10 <sup>-6</sup>	1.93 × 10 <sup>-6</sup>		

the results (22.2–26.2 Bq/kg fresh weight) (Huang et al., 2012) of marine fish in the seas around QNPP from 2007 to 2009 and the results (45.80 ± 9.75, 29.87 ± 6.03, 26.58 ± 6.10, 19.84 ± 4.03, and 10.68 ± 3.45 Bq/kg) (Cao et al., 2024) of seafood from Sanmen, and lower than those reported for samples collected around France (800 Bq/kg, fresh sample) (Le Guen and Siclet, 2009). There are very few reports on the radioactivity levels of <sup>14</sup>C in seafood, especially for prawn, crabs, shellfish and seaweeds, and data obtained in this study provide references for other relevant studies.

### 3.4. Systematic cluster analysis

Analysis containing all 18 seafood samples using clustering heatmap was adopted, as there was no statistically significant difference in the activity concentrations of TFWT, OBT, and <sup>14</sup>C in seafood from QNPP and Hangzhou Bay. As shown in the Fig. 2, the samples can be divided into two groups: two sea crab samples collected around QNPP were grouped together, while the other 16 samples were grouped together. This division may be attributed to the proximity of sea crab habitat to the nuclear power plant (<1 km), resulting in a higher intake of radionuclides.



### 3.5. Estimation of internal exposure doses to residents

The estimated AED values from  $^3\text{H}$  and  $^{14}\text{C}$  due to consumption of seafood are presented in Table 4. The annual intake of TFWT, OBT and  $^{14}\text{C}$  in the five seafood categories around QNPP ranged from 1.67 to 94.32 Bq, 0.27 to 7.65 Bq and 4.32 to 246.37 Bq, resulting in total AED of  $1.96 \times 10^{-4}$  mSv/year, accounting for 0.08 % of the annual dose constraint. The annual intake of TFWT, OBT and  $^{14}\text{C}$  in the five seafood categories in Hangzhou Bay ranged from 1.26 to 76.71 Bq, 0.25 to 3.03 Bq and 2.92 to 197.28 Bq, resulting in an AED of  $1.61 \times 10^{-4}$  mSv/year and accounting for 0.06 % of the annual dose constraint. Moreover, the total annual effective ingestion dose from the intake of detected radionuclides ( $^3\text{H}$  and  $^{14}\text{C}$ ) from the ingestion of seafood consumption is below the acceptable level of 1.0 mSv/year established by the ICRP for the Public.

### 4. Conclusions

During the operation of nuclear power plants, waste containing  $^3\text{H}$  and  $^{14}\text{C}$  will be discharged into the ocean, which can cause internal radiation hazards to the human body through ingestion of seafood. However, there are currently limited reports on monitoring  $^3\text{H}$  and  $^{14}\text{C}$  in seafood. In this study, we collect samples according to the dietary habits of residents, detect the  $^3\text{H}$  and  $^{14}\text{C}$  activity concentrations in five types of seafood around QNPP, and compare them with Hangzhou Bay. The results showed that the concentrations of TFWT, OBT, and  $^{14}\text{C}$  in five types of seafood around QNPP were higher than those in Hangzhou Bay. However, the differences were not statistically significant. These are significantly lower than the reference values (6.5105 Bq/kg). The calculation, results indicate that the annual intake and effective ingestion dose values were low, and the dose burden was low. We foresee the need for continuing to monitor  $^3\text{H}$  and  $^{14}\text{C}$  in the environment and seafood around nuclear power plants and estimate public health risks in the future works.

### CRedit authorship contribution statement

**Xiaoxiang Ma:** Writing – original draft, Investigation. **Yiyao Cao:** Methodology, Data curation. **Taotao Zheng:** Writing – review & editing, Validation. **Shunfei Yu:** Methodology. **Hua Zou:** Supervision, Software. **Xinyu Gong:** Data curation. **Yi Cao:** Data curation. **Hong Ren:** Writing – review & editing, Validation, Supervision.

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Data availability

Data will be made available on request.

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.fochx.2024.101243>.

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