



# Activity Levels of $^{210}\text{Po}$ , $^{210}\text{Pb}$ and Other Radionuclides ( $^{134}\text{Cs}$ , $^{137}\text{Cs}$ , $^{90}\text{Sr}$ , $^{110\text{m}}\text{Ag}$ , $^{238}\text{U}$ , $^{226}\text{Ra}$ and $^{40}\text{K}$ ) in Marine Organisms From Coastal Waters Adjacent to Fuqing and Ningde Nuclear Power Plants (China) and Radiation Dose Assessment

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With the rapid development of nuclear power, the radiation impacts on edible marine organisms, and the potential radiation risks to humans have become of considerable concern to public health. In this study, the activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  as well as those of other radionuclides in fishes (*Mugil cephalus*, *Konosirus punctatus*, *Largehead hairtail*, and *Larimichthys polyactis*), crustaceans (*Mantis shrimp*, *Parapenaeopsis hardwickii*, and *Portunus trituberculatus*), bivalves (*Crassostrea gigas*, *Sinonovacula conzcta*), and macroalgae (*Gracilaria*, *Porphyra*) collected in the coastal area adjacent to the Fuqing and Ningde nuclear power plants (NPPs) were determined. The activity range of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  was 0.60–48.09 and 0.07–2.76 Bq/kg<sub>freshweight</sub>, respectively, with  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratios of 1.1–189.7. The ranking of  $^{210}\text{Po}$  activity levels in marine organisms was bivalve mollusks > crustaceans > fishes > macroalgae. The calculated bioconcentration factors of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  were 636–44,944 and 3–1,226 L/kg, respectively. These values provide a new supplement to the IAEA reference database. The radiation dose rates for these marine organisms ranged from 0.037 to 1.531  $\mu\text{Sv/h}$ , which was much lower than the ERICA ecosystem screening benchmark of 10  $\mu\text{Gy/h}$ . The calculated committed effective dose received by humans from ingestion of these marine organisms was 0.06–2.99 mSv. Overall,  $^{210}\text{Po}$  was the dominant radiation dose contributor in marine organisms and humans, whereas the dose contributions from the artificial nuclides  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  were negligible.

**Keywords:** lead, polonium, marine biota, nuclear power plant, dose assessment

## INTRODUCTION

The polonium isotope  $^{210}\text{Po}$  (half-life,  $T_{1/2} = 138.4$  d) and its grandparent  $^{210}\text{Pb}$  ( $T_{1/2} = 22.26$  y) are nonconservative, naturally occurring radionuclides within the uranium  $^{238}\text{U}$  decay chain, which is ubiquitous in the environment of the earth. The isotopes  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the atmosphere mainly originated from the release of  $^{222}\text{Rn}$  from the ground and its subsequent decay. Due to their strong particle reactivity, they are firmly attached to the aerosol soon after they are produced. With the dry and wet depositions, they are subsequently discharged into the terrestrial and marine environment *via* dry and wet deposition (Seiler and Wiemels, 2012). Due to their unique geochemical properties,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are used as a tracer pair to study the dynamic processes of aerosols in the atmosphere and estimate the residence times of aerosols (Aba et al., 2020). They are also used to study particle scavenging processes in the sea, particularly in assessing the export of particulate organic carbon (POC) fluxes from the euphotic zone (Zhang et al., 2020; Bam and Maiti, 2021), as well as specific marine food chain processes (Strady et al., 2015). Indeed, beyond the oceanographic application of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , their accumulation in marine organisms and transfer to human consumers of seafood, and the resulting radiation doses to marine organisms or committed effective doses to humans are also issues of public concern. This is especially true for  $^{210}\text{Po}$ , as it is one of the most radiotoxic nuclides that emit high-energy ( $\sim 5.3$  MeV) alpha rays and is the main contributor of the radiation dose received by marine organisms and humans (UNSCEAR, 2000; Sivakumar, 2014; Men et al., 2020a,b).

Marine organisms usually concentrate  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  from the marine environment. Although the activity levels of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the marine environment are relatively low compared with those in the terrestrial environment, different marine organisms can concentrate these two radionuclides to relatively high levels with high concentration factors (CFs) ( $\sim 10^2$  to  $\sim 10^5$ ) (IAEA, 2004). Therefore,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  provide the main radiation source for marine organisms. In seawater, there are relatively higher levels of other naturally occurring nuclides, such as uranium  $^{238}\text{U}$  ( $12.2\text{--}215.4$  Bq/m $^3$ ), radium  $^{226}\text{Ra}$  ( $0.22\text{--}7.20$  Bq/m $^3$ ), and potassium  $^{40}\text{K}$  ( $\sim 12,000$  Bq/m $^3$ ), and artificial radionuclides, such as cesium  $^{137}\text{Cs}$  ( $< 3.2$  Bq/m $^3$ ) and strontium  $^{90}\text{Sr}$  ( $< 2.2$  Bq/m $^3$ ) (IAEA, 2005; Liu, 2010). Marine organisms also concentrate these nuclides in their body, which thus also produce self-radiation. Since the 1980s, the concept of human-centered environmental protection has gradually evolved into the concept of ecological protection in which the whole ecosystem is the protection target within the field of radiation protection. Many international organizations and government departments have been studying the effects of ionizing radiation on nonhuman species, including the International Commission on Radiation Protection (ICRP), the International Atomic Energy Agency (IAEA), the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), and the European Commission (EC). Additionally, after the 2011 Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident, the rapid development of nuclear power has raised increasing attention to the radiation impacts on marine organisms and the potential

radiation risks to public health (Yu et al., 2018; Men et al., 2020a,b).

At present, the Fuqing and Ningde Nuclear Power Plants (NPPs), located on the coast of Fujian province (Figure 1), are in operation. The marine organisms living in the area adjacent to these two NPPs provide ideal experimental test subjects to study the concentrations of radionuclides in the marine environment as well as to undertake radiation dose assessment. In this study, data are provided on the activity levels of naturally occurring and artificial radionuclides in marine organisms used as bio-monitors of nuclear power plant operations. Activity levels of  $^{210}\text{Po}$ ,  $^{210}\text{Pb}$ , and other naturally occurring or artificial radionuclides were investigated in fish, crustaceans, bivalve mollusks, and macroalgae in the areas surrounding Fuqing and Ningde NPPs, and the resulting radiation doses to both marine organisms and humans were assessed.

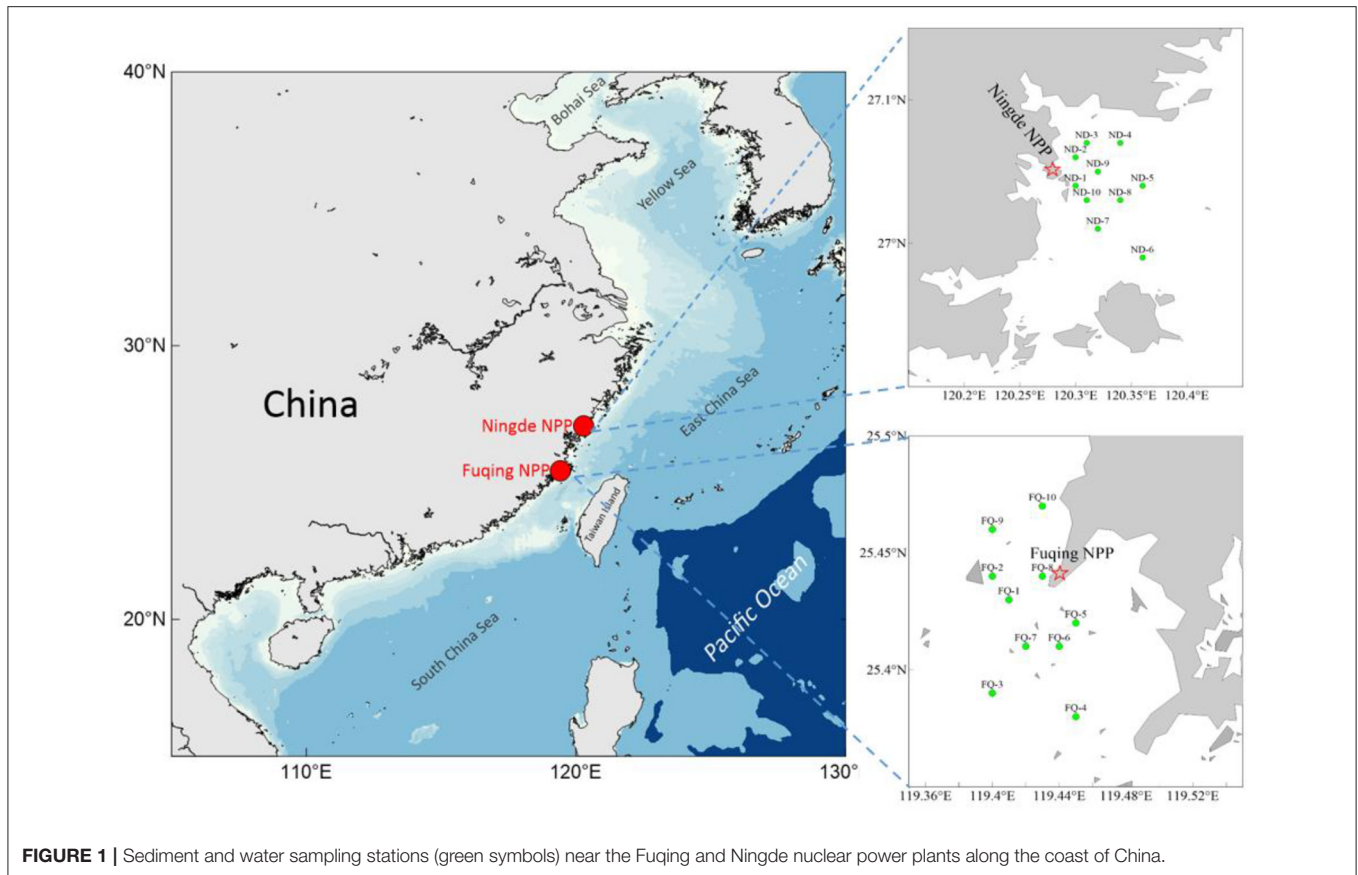
## MATERIALS AND METHODS

### Sample Collection

Samples of marine organisms were obtained by hired fishermen in areas adjacent to Fuqing and Ningde NPPs (i.e., within 10 km) in July 2020 (Figure 1). Twelve samples with a fresh weight of  $\sim 2.2\text{--}10.6$  kg each were collected. They were refrigerated and immediately sent to the laboratory (within 24 h). Marine organisms include fishes (the mullet *Mugil cephalus*, *Konosirus punctatus*, *Largehead hairtail*, and *Larimichthys polyactis*), crustaceans (*Mantis shrimp*, *Parapenaeopsis hardwickii*, and crab *Portunus trituberculatus*), bivalves (soft tissues of the Pacific oyster *Crassostrea gigas*, and razor clam *Sinonovacula conzcta*), macroalgae [the red algae *Gracilaria* spp. (*Gracilariaceae*) and *Porphyra* spp. (*Bangiaceae*)] (Figure 2). Seawater and sediment samples were also collected at each of 10 stations near Fuqing NPP and Ningde NPP (Figure 1).

### Sample Processing and Analysis

The weighed marine organism samples were dried to constant weight for 48–96 h at 60°C in a drum dryer. Dried samples were pulverized, using agate mortar and pestle sets in preparation for the radioactive analysis. About 1 g of these pulverized dry samples was used for the measurement of  $^{210}\text{Po}$ , using  $\alpha$  spectrometer (Canberra 7200) (Štok and Smodiš, 2011). The rest was transferred into crucibles and ashed in a muffle furnace at 450°C for 24–40 h. The ashes were ground and weighed at room temperature, stored in sealed boxes ( $\sim 100$  g per sample) for 20 days until analysis. Canberra BE6530 and GR4021 HPGe spectrometers were used to determine the activities of  $^{210}\text{Pb}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  $^{110m}\text{Ag}$ ,  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ , and  $^{40}\text{K}$  (Men et al., 2017). The di (2-ethylhexyl) phosphoric acid (HDEHP) extraction- $\beta$  counting method and the Ortec MPC-9604  $\alpha/\beta$  counter were employed for  $^{90}\text{Sr}$  analysis (Men et al., 2017), using  $\sim 10$  g of the ashes. Seawater and sediment samples were also analyzed according to the Technical Specification for Marine Radioactivity Monitoring (State Oceanic Administration of China, 2011). All marine organisms were analyzed whole, except for the bivalves whose shells were removed. Parallel sample analysis was



**FIGURE 1** | Sediment and water sampling stations (green symbols) near the Fuqing and Ningde nuclear power plants along the coast of China.

implemented for *Konosirus punctatus* and *Mantis shrimp*; the results were in good agreement within an error <3%.

Specifically, ca. 1 ml of 0.12848 Bq/ml  $^{209}\text{Po}$  was added to 1 g of a dry biological sample, and then the spiked sample was digested with a mixture of concentrated nitric acid and hydrogen peroxide. After steaming until nearly dry, 2 mL of concentrated hydrochloric acid (HCl) was added and steaming carried out again, and the residue was dissolved with 2-M HCl. After filtration, the filtrate was placed in an  $\alpha$  spectrometer for measurement over 24 h. The chemical yield for  $^{209}\text{Po}$  ranged from 52 to 89%, averaging  $72 \pm 12\%$  (SD,  $n = 14$ ) after adding 1 ml of 0.12848 Bq/ml of  $^{209}\text{Po}$  standard solution.

Seawater (5 L) was taken from each station for analysis. A known amount of  $^{209}\text{Po}$  ( $\sim 1$  g) was added to the seawater samples to determine the yield. The spiked samples were co-precipitated with ferric hydroxide by adding  $\sim 50$  mg of  $\text{Fe}^{3+}$  and adjusting the pH to  $\sim 8$ , with the addition of concentrated ammonium hydroxide ( $\text{NH}_4\text{OH}$ ). The precipitate was then dissolved in concentrated HCl, and auto-deposition was carried out. The analysis of other radionuclides in seawater is described in detail by Men et al. (2017).

## Radiation Dose for Marine Organisms

The ERICA assessment tool (version 1.3, Tier 2) was used to evaluate the dose rates for marine organisms (Beresford

et al., 2007; Men et al., 2020a,b). The average biological parameters of the specimens sampled, including length, width, and height, as well as weight, are listed in **Table 1**, and were used to calculate the radiation doses listed (the biological parameters were determined for all individuals of each species in the sample). The average nuclide activities in seawater and sediment were used to estimate the external dose rates. The activity levels of these nuclides in the marine organism were used to estimate the internal dose rates. The low beta, beta/gamma, and alpha weighing factors were taken to be 3, 1, and 10, respectively. The other parameters were set to their default values.

## Committed Effective Dose for Humans Consuming Various Marine Organisms

After ingestion or inhalation by humans, some radionuclides persist in the body and irradiate various tissues for many years. The resulting total effective dose over a lifetime (70 years or number of years up to reaching age, 70 for infants, 50 years for adults) is the committed effective dose (ICRP, 2007; Men et al., 2017). This dose received by a human per unit intake (1 Bq) of a given radionuclide is the radionuclide-specific dose coefficient (DC) for ingestion (Fisher et al., 2013), which converts the energy emitted from the ingested radionuclide into a radionuclide-specific, committed effective dose for human



**FIGURE 2** | Specimens of the marine organism sampled in this study.

adults (Sv). For calculation of the committed effective dose for ingestion of marine organisms in this study, the ingestion rate was assumed as exact ingestion rates were not available. Here, the mean *per capita* consumption rate of aquatic products in China (50.97 kg/year) in 2018 was used to estimate the committed effective dose (FAOSTAT, 2018). This was calculated by multiplying the radionuclide activity in the marine organism (Bq/kg *freshweight*) by the ingested mass (kg) and the DC (Sv/Bq) (ICRP, 2012).

## RESULTS AND DISCUSSION

### Activity Levels of $^{210}\text{Po}$ and $^{210}\text{Pb}$ and Other Radionuclides in Marine Organisms

The activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  as well as other radionuclides in marine organisms from the coastal area adjacent to Fuqing and Ningde NPPs are listed in **Table 2**;  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  activities ranged from 0.60 to 48.09 Bq/kg *freshweight* and 0.07 to 2.76 Bq/kg *freshweight*, respectively. These values are within

**TABLE 1** | Average biological parameters of the sampled marine organisms.

Sea area	Organism	Length (cm)	Width (cm)	Height (cm)	Mass (kg)
Fuqing NPP	<i>Mugil cephalus</i>	27.00	5.00	4.00	1.100
	<i>Gracilaria</i>	60.00	0.10	0.10	0.006
	<i>Portunus trituberculatus</i>	10.00	6.00	3.00	0.400
	<i>Konosirus punctatus</i>	18.00	4.50	4.00	0.110
	<i>Porphyra</i>	35.00	2.00	0.10	0.009
Ningde NPP	<i>Sinonovacula constrzcta</i>	5.00	2.00	2.00	0.012
	<i>Largehead hairtail</i>	60.00	5.00	2.00	1.000
	<i>Crassostrea gigas</i>	7.00	4.00	3.00	0.056
	<i>Parapenaeopsis hardwickii</i>	8.00	1.00	1.00	0.020
	<i>Mantis shrimp</i>	16.00	2.50	2.00	0.050
	<i>Larimichthys polyactis</i>	15.00	6.00	2.00	0.080
	<i>Porphyra</i>	20.00	1.50	0.10	0.007

**TABLE 2** | Activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  and other radionuclides in marine organisms sampled in this study.

Sea area	Organisms	$^{210}\text{Po}$	$^{210}\text{Pb}$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{238}\text{U}$	$^{226}\text{Ra}$	$^{40}\text{K}$	$^{210}\text{Po}/^{210}\text{Pb}$ <sub>A.R.</sub>
		Bq/kg fresh weight							
Fuqing NPP	<i>Mugil cephalus</i>	2.25 ± 0.24	1.33 ± 0.36	0.05 ± 0.01	0.37 ± 0.03	2.66 ± 0.08	1.61 ± 0.03	121.1 ± 3.3	1.7
	<i>Gracilaria</i>	3.06 ± 0.19	2.76 ± 0.73	0.01 ± 0.01	0.03 ± 0.01	0.50 ± 0.02	0.13 ± 0.01	106.6 ± 2.9	1.1
	<i>Portunus trituberculatus</i>	41.04 ± 0.67	/	ND	0.39 ± 0.04	0.69 ± 0.21	1.22 ± 0.03	81.9 ± 2.4	/
	<i>Konosirus punctatus</i>	2.07 ± 0.27	0.32 ± 0.09	ND	0.08 ± 0.03	0.25 ± 0.02	0.24 ± 0.01	107.7 ± 2.9	6.5
	<i>Porphyra</i>	0.60 ± 0.13	0.51 ± 0.14	ND	0.09 ± 0.01	0.13 ± 0.01	0.08 ± 0.01	93.4 ± 2.5	1.2
Ningde NPP	<i>Sinonovacula constrzcta</i>	33.09 ± 1.09	1.42 ± 0.38	ND	0.05 ± 0.01	1.03 ± 0.04	0.36 ± 0.01	174.9 ± 4.8	23.3
	<i>Largehead hairtail</i>	32.25 ± 0.74	0.17 ± 0.05	0.08 ± 0.01	0.44 ± 0.05	0.15 ± 0.01	0.09 ± 0.01	63.7 ± 1.7	189.7
	<i>Crassostrea gigas</i>	48.09 ± 1.06	0.65 ± 0.18	0.03 ± 0.01	0.50 ± 0.05	0.33 ± 0.01	0.04 ± 0.01	76.3 ± 2.1	74.0
	<i>Parapenaeopsis hardwickii</i>	13.29 ± 0.59	0.14 ± 0.05	0.03 ± 0.01	0.75 ± 0.08	0.71 ± 0.03	0.53 ± 0.01	56.9 ± 1.6	94.9
	<i>Mantis shrimp</i>	21.54 ± 0.7	0.30 ± 0.08	0.03 ± 0.01	0.10 ± 0.02	0.65 ± 0.03	0.35 ± 0.01	55.4 ± 1.5	71.8
	<i>Larimichthys polyactis</i>	15.53 ± 0.71	/	ND	0.03 ± 0.01	0.04 ± 0.01	0.11 ± 0.01	72.5 ± 2.0	/
	<i>Porphyra</i>	0.68 ± 0.13	0.07 ± 0.03	ND	0.09 ± 0.01	0.07 ± 0.01	0.02 ± 0.01	51.0 ± 1.4	9.7

ND, not detected.  $^{134}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$  were also undetectable. The MDA (minimum detectable activity) for  $^{137}\text{Cs}$ ,  $^{134}\text{Cs}$ , and  $^{110\text{m}}\text{Ag}$  was 0.0014 Bq/kg freshweight, (661.7 keV), 0.0014 (604.7 keV) Bq/kg freshweight, and 0.0012 Bq/kg freshweight (657.8 keV), respectively, during a counting time of 96,708 s and with 10 kg samples. The MDA for  $^{210}\text{Po}$  was 0.0022 Bq/kg freshweight (5,304.5 keV) during a counting time of 172,800 s and with 5 g samples. The blank for  $^{210}\text{Po}$  was 2.7641 Bq/kg freshweight. The blank for  $^{90}\text{Sr}$  was 0.215 cpm. The blank for  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ , and  $^{40}\text{K}$  was 0.016 cpm, 0.004 cpm, and 0.005 cpm, respectively. The CRM (certified reference material) was 100-g fish ash (standard values: 5.9815 Bq/kg fishash,  $^{134}\text{Cs}$ ; 34.2954 Bq/kg fishash  $^{137}\text{Cs}$ ; 0.7990 Bq/kg fishash  $^{110\text{m}}\text{Ag}$ ). The measured values for  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ , and  $^{110\text{m}}\text{Ag}$  in 100-g fish ash dry weight during the counting time of 176,619 s were 0.1405, 0.5023, and 0.0104 Bq/kg fishash. / indicates lack of data.

the reported ranges of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in marine organisms in China ( $^{210}\text{Po}$ : 0.117–65.8 Bq/kg freshweight;  $^{210}\text{Pb}$ : 0.02–6.88 Bq/kg freshweight) (Li et al., 2016, 2018; Lin et al., 2016; Dong et al., 2018; Lin, 2018). The limit of  $^{210}\text{Po}$  activity recommended in fish, meat, and shrimp by the Chinese National Standard on limited concentrations of radioactive materials in foods (GB 14882-94) is 15 Bq/kg freshweight (Ministry of Health of the People's Republic of China, 1994). About 50% of  $^{210}\text{Po}$  activities in marine organisms reported in the present study

exceeded this value. Most  $^{210}\text{Po}$  activities were higher than the UNSCEAR representative  $^{210}\text{Po}$  activities in marine fish, crustaceans, and mollusks (2.4, 6, and 15 Bq/kg freshweight) (UNSCEAR, 2000). The activity levels of  $^{210}\text{Po}$  varied greatly among the different marine species. For example, the highest and lowest  $^{210}\text{Pb}$  activities were measured in *Crassostrea gigas* and *Porphyra*, respectively. In general,  $^{210}\text{Po}$  activities in marine organisms ranked in the order bivalves > crustaceans > fishes > macroalgae.

**TABLE 3** | Average activities of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  and other radionuclides in seawater/sediment.

Sea area	$^{210}\text{Po}$	$^{210}\text{Pb}$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{238}\text{U}$	$^{226}\text{Ra}$	$^{40}\text{K}$
	Seawater (Bq/m <sup>3</sup> )/Sediment (Bq/kg)						
Fuqing NPP (n = 10)	2.24/87.8	2.51/82.5	1.31/1.13	0.71/0.17	33.6/41.5	3.36/31.2	11,550/687.9
Ningde NPP (n = 10)	1.07/108.6	1.28/104.7	1.46/2.08	0.74/0.20	33.5/51.8	2.75/31.2	11,510/647.4

$^{134}\text{Cs}$  and  $^{110m}\text{Ag}$  were undetectable in seawater and sediment.

**TABLE 4** | Bioconcentration factors of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  and other radionuclides in marine organisms sampled in this study.

Organisms	$^{210}\text{Po}$	$^{210}\text{Pb}$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{238}\text{U}$	$^{226}\text{Ra}$	$^{40}\text{K}$
<i>Mugil cephalus</i>	1,004	591	38	521	79	479	10
<i>Gracilaria</i>	1,366	1,226	8	42	15	39	9
<i>Portunus trituberculatus</i>	18,321	18	/	549	20	363	7
<i>Konosirus punctatus</i>	924	142	/	113	7	71	9
<i>Porphyra</i>	268	227	/	127	4	24	8
<i>Sinonovacula constrzcta</i>	30,925	1,109	/	68	29	131	15
<i>Largehead hairtail</i>	30,140	133	55	595	4	33	6
<i>Crassostrea gigas</i>	44,944	508	21	676	9	15	7
<i>Parapenaeopsis hardwickii</i>	12,421	109	21	1,014	20	193	5
<i>Mantis shrimp</i>	20,131	234	21	135	18	127	5
<i>Larimichthys polyactis</i>	14,514	3	/	41	1	40	6
<i>Porphyra</i>	636	55	/	122	2	7	4
Fish <sup>a</sup>	2,000	200	100	3	1	100	\
Macroalgae <sup>a</sup>	1,000	1,000	5	1	100	100	\
Crustaceas <sup>a</sup>	20,000	90,000	50	5	10	100	\
Molluscs <sup>a</sup>	20,000	50,000	60	10	30	100	\

/Indicates that the value was below the detection limit or was not determined; \ indicates lack of data in the database of IAEA recommended values. <sup>a</sup>IAEA recommended value (IAEA, 2004).

The accumulation of  $^{210}\text{Po}$  in marine organisms is related to food type, life cycle stage, trophic level, and body size (Carvalho, 2018). Firstly, suspension-feeding bivalves are primary consumers that mainly ingest phytoplankton and detrital particulate organic matter. Crustaceans are opportunistic secondary consumers that mainly ingest benthic organisms. Biomagnification can significantly enhance the  $^{210}\text{Po}$  activity level in bivalves (Fowler, 2011; Dong et al., 2018). Secondly, bivalves that usually live on the bottom showed higher  $^{210}\text{Po}$  activities due to rapid bottom deposition and biological adsorption. The higher  $^{210}\text{Po}$  level in their bodies has been attributed to bioconcentration (Sirelkhatim et al., 2008; Lin, 2018). Finally,  $^{210}\text{Po}$  is typically more concentrated in the digestive tract and hepatopancreas or in the gonads (Carvalho, 2018; Dong et al., 2018; Hurtado-Bermudez et al., 2019). The  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratios in the present study ranged from 1.1 to 189.7 (Table 2). It is reported that both  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  bind strongly to organisms, and that  $^{210}\text{Pb}$  is preferably associated with the mineral fractions of bones and shells. Compared with  $^{210}\text{Pb}$ ,  $^{210}\text{Po}$  is primarily associated with proteins in organisms and can penetrate the cell cytoplasm. Therefore,  $^{210}\text{Po}$  can be more effectively assimilated in marine organisms than  $^{210}\text{Pb}$ ,

resulting in  $^{210}\text{Po}/^{210}\text{Pb}$  activity ratios >1 in most marine organisms (Stewart et al., 2008).

As shown in Table 2, the activities of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ , and  $^{40}\text{K}$  ranged from undetectable to 0.08, 0.03–0.75, 0.04–2.66, 0.02–1.61, and 51.–174.9 Bq/kg<sub>freshweight</sub>, respectively. The activity levels ranked in the order  $^{40}\text{K} > ^{210}\text{Po} > ^{210}\text{Pb} > ^{238}\text{U} > ^{226}\text{Ra} > ^{90}\text{Sr} > ^{137}\text{Cs}$ . The activity levels of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in marine organisms were  $\sim 10^{-2}$  to  $\sim 10^{-1}$  Bq/kg<sub>freshweight</sub>, which is within background levels (Liu and Zhou, 2000; Chen et al., 2003; Zhang, 2015; Lou et al., 2018). Those of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  activities in fish, meat, and shrimp established by the Chinese National Standard on limited concentrations of radioactive materials in foods are 290 and 800 Bq/kg<sub>freshweight</sub>, respectively (Ministry of Health of the People's Republic of China, 1994). The radioisotope  $^{210}\text{Po}$  is the major natural decay product from the uranium series and provides the largest radiation dose to the human body via consumption of marine organisms (UNSCEAR, 2000; Carvalho, 2011; Khot et al., 2021; Kong et al., 2021). Indeed, the scavenging rate of  $^{210}\text{Po}$  is higher than that of other radionuclides in the atmospheric environment (Alam and Mohamed, 2011), resulting in high  $^{210}\text{Po}$  deposition in the marine environment. In turn, marine organisms show a stronger

affinity for <sup>210</sup>Po than for other radionuclides (Bogdan, 1997; Lin, 2018), resulting in a higher activity level of <sup>210</sup>Po than that of other radionuclides. The activity levels of <sup>90</sup>Sr and <sup>137</sup>Cs in marine organisms in the present study are far below these values. The average activities of <sup>210</sup>Po and <sup>210</sup>Pb as well as other radionuclides in seawater and sediment in the sea area adjacent to Fuqing and Ningde NPPs are listed in Table 3. The data in Tables 2, 3 were used to estimate the radiation doses for the corresponding marine organisms.

### Bioaccumulation of <sup>210</sup>Po and <sup>210</sup>Pb and Other Radionuclides in Marine Organisms

The bioconcentration factor is defined as the activity ratio of a radionuclide in the marine organism or biota to that in ambient seawater (L/kg) and is an indicator of the accumulation capacity of a given organism for a particular nuclide (Arnot and Gobas, 2006; Alava and Gobas, 2016; Ishii et al., 2020). Bioconcentration factors in different radionuclides vary widely due to their different biochemical properties, while bioconcentration factors (BCFs) in different marine organisms differ greatly due to their different bioaccumulation capacities. Even within the same species, BCFs vary among individuals due to differences in physiology, microhabitat, etc. For the sake of convenience and standardization, a set of values for different radionuclides and different kinds of marine organisms was recommended by the IAEA (Table 4) (IAEA, 2004). Using the data for seawater and marine organism samples in the present study, the BCFs of <sup>210</sup>Po and <sup>210</sup>Pb as well as those of other radionuclides can be estimated (Table 4). The BCFs for <sup>210</sup>Po and <sup>210</sup>Pb were in the ranges 636–44,944 and 3–1,226, respectively. BCFs of <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>238</sup>U, <sup>226</sup>Ra, and <sup>40</sup>K were in the range 5–55, 41–1,014, 1–79, 7–479, and 4–15 L/g<sub>freshweight</sub>, respectively. The BCF data reported in this study provide a useful supplement of information for the IAEA database.

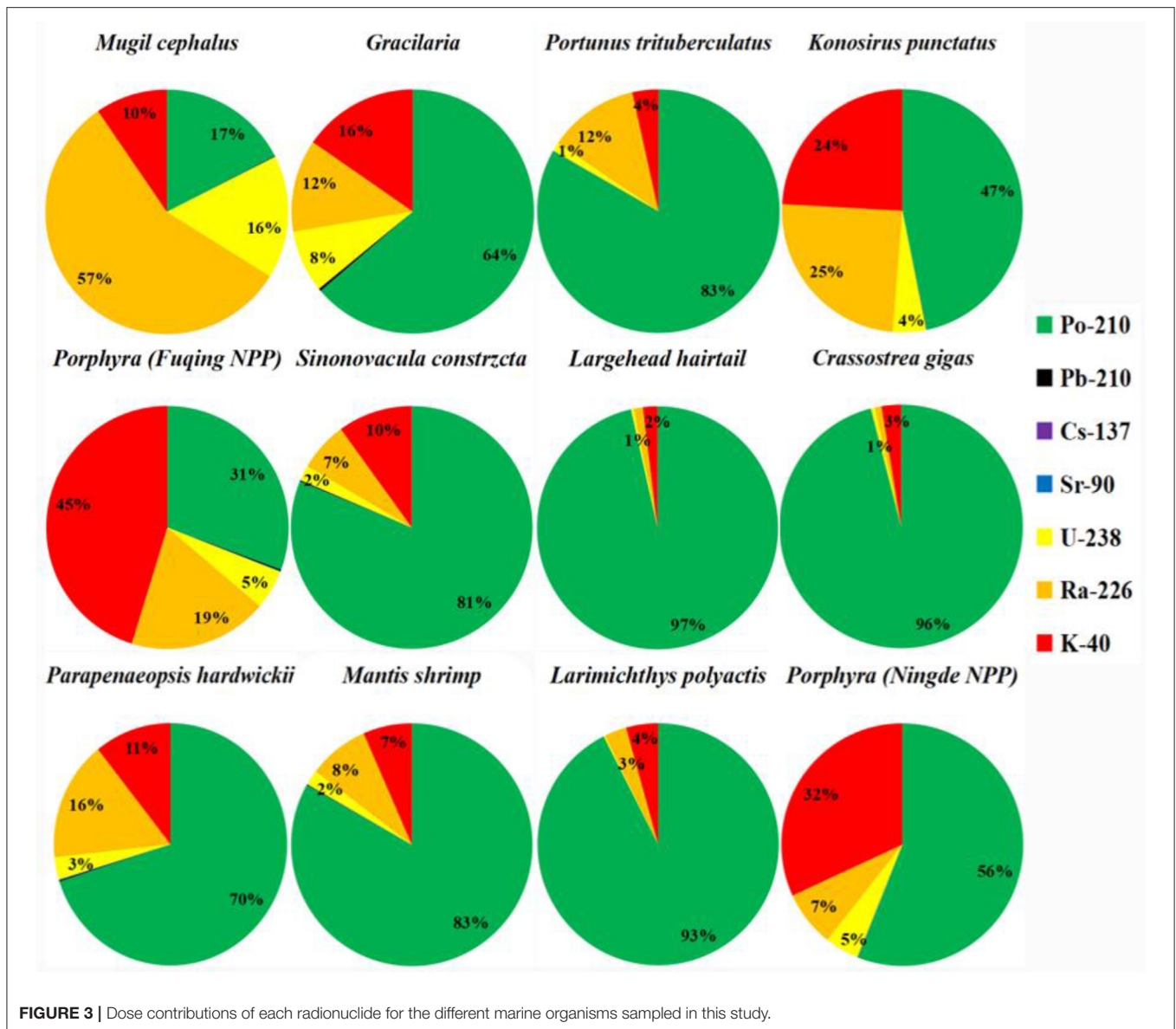
### Radiation Dose Assessment

The radiation doses for nonhuman species have become an issue of increasing public health concern. The ERICA tools downloaded freely from the internet are widely used for radiation assessment (Garnier-Laplace et al., 2011; Johansen et al., 2015; Men et al., 2017, 2020a,b). As shown in Tables 1–4, the radiation doses received by marine organisms in the studied area were assessed, using the ERICA tools. The internal and external dose rates derived for each radionuclide and the total radiation dose rates are listed in Table 5. The total dose rates ranged from 0.037 to 1.531 μSv/h. Around the Ningde NPP, the highest and lowest radiation doses were observed in *Crassostrea gigas* and *Porphyra*, respectively. Overall, these values are markedly lower than the ERICA ecosystem screening benchmark of 10 μGy/h (Beresford et al., 2007) and the most conservative safety benchmark, which is one to two orders of magnitude lower than the International Commission on Radiological Protection (ICRP)-derived reference levels for corresponding reference animals or plants (ICRP, 2008; Fisher et al., 2013; Men et al., 2017). This suggested that there are no irradiation effects on marine organisms in the area adjacent to Fuqing and Ningde NPPs.

TABLE 5 | Internal and external radiation dose rates derived for each radionuclide for the different marine species sampled in this study (in μSv/h).

Organisms	<sup>210</sup> Po		<sup>210</sup> Pb		<sup>137</sup> Cs		<sup>90</sup> Sr		<sup>238</sup> U		<sup>226</sup> Ra		<sup>40</sup> K		Total
	Internal	External	Internal	External	Internal	External	Internal	External	Internal	External	Internal	External	Internal	External	
<i>Mugil cephalus</i>	68.7	0.00000007	0.337	0.000007	0.009	0.0004	0.23	0.00002	64.3	0.000004	223.5	0.003	36.9	1.0	394.9
<i>Graclaria</i>	93.5	0.00000008	0.514	0.000121	0.001	0.0005	0.01	0.00020	12.1	0.000021	18.0	0.004	20.3	2.3	146.7
<i>Portunus trituberculatus</i>	1,253.9	0.000199604	0.010	0.240170	/	0.2235	0.24	0.00240	16.7	0.002909	169.3	13.283	24.6	27.8	1,506.3
<i>Konosirus punctatus</i>	63.2	0.00000007	0.080	0.000013	/	0.0004	0.05	0.00003	6.0	0.000007	33.3	0.003	31.5	1.1	135.4
<i>Porphyra (Fuqing NPP)</i>	18.3	0.00000007	0.123	0.000027	/	0.0004	0.05	0.00007	3.1	0.000012	11.1	0.003	25.7	1.3	59.8
<i>Sinonovacula costizcta</i>	1,011.0	0.000476112	0.345	1.459055	/	0.5396	0.03	0.01597	24.9	0.015291	49.9	32.585	48.6	75.5	1,244.9
<i>Largehead hairtail</i>	985.3	0.00000007	0.043	0.000010	0.014	0.0004	0.27	0.00003	3.6	0.000005	12.5	0.003	18.9	1.1	1,021.8
<i>Crassostrea gigas</i>	1,469.3	0.000116337	0.162	0.230209	0.005	0.1313	0.30	0.00246	8.0	0.002707	5.5	7.824	22.1	17.8	1,531.3
<i>Parapenaeopsis hardwickii</i>	406.0	0.000285720	0.034	0.903700	0.004	0.3241	0.43	0.00953	17.2	0.009096	73.5	19.541	15.8	46.1	579.8
<i>Mantis shrimp</i>	658.1	0.000235172	0.074	0.544626	0.005	0.2655	0.06	0.00588	15.7	0.006198	48.5	15.889	15.8	35.9	790.9
<i>Larimichthys polyactis</i>	474.5	0.00000007	0.001	0.000017	/	0.0004	0.02	0.00004	1.0	0.000008	15.3	0.003	20.8	1.2	512.8
<i>Porphyra (Ningde NPP)</i>	20.8	0.00000008	0.013	0.000126	/	0.0005	0.03	0.00021	1.7	0.000021	2.8	0.004	9.5	2.4	37.1

/Lack of data due to the fact that the activities of <sup>137</sup>Cs in marine organisms were below the MDA.



**FIGURE 3** | Dose contributions of each radionuclide for the different marine organisms sampled in this study.

The dose contributions of different nuclides in different species were plotted in **Figure 3** and show that  $^{210}\text{Po}$  was the dominant dose contributor except for *Mugil cephalus* and *Porphyra* (Fuqing NPP) (47–97%), while  $^{226}\text{Ra}$  and  $^{40}\text{K}$  were the main dose contributors for *Mugil cephalus* and *Porphyra* (Fuqing NPP), respectively. The contribution from external and internal doses for each nuclide (**Table 5**) suggests that the internal doses were much greater than the external doses. In general, the greatest internal dose should be from  $^{210}\text{Po}$  sources because of its alpha emissions. Additional main contributors should be  $^{226}\text{Ra}$  and  $^{238}\text{U}$ , which produced intermediate internal doses because of alpha emissions. Due to high-activity levels in seawater ( $\sim 11,500$  Bq/ $\text{m}^3$ ) and marine organisms (51–174.9 Bq/kg  $_{\text{freshweight}}$ ) as well as emitted high-energy  $\gamma$ -rays (1,460 keV),  $^{40}\text{K}$  generated much higher internal and external dose rates than  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ,

and  $^{90}\text{Sr}$  (EI-Arabi, 2007). Indeed, the dose contribution from  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  was  $<0.13\%$ , which was extremely low compared to that of naturally occurring radionuclides.

### Radiation Dose Assessment for Humans

The calculated committed effective dose for humans from ingestion of marine organisms in the area adjacent to Fuqing and Ningde NPPs was 60.74–2,990.41  $\mu\text{Sv}$  (**Table 6**). Results show that a maximum committed dose of 2.99 mSv will be received over the following 50 years based on assumed consumption of 50.97 kg of these marine organisms in 1 year. In terms of species, *Porphyra* had the lowest committed effective dose to humans ( $<100$   $\mu\text{Sv}$ ), while *Portunus trituberculatus*, *Sinonovacula constricta*, *Largehead hairtail*, *Crassostrea gigas*, and *Mantis shrimp* had committed effective doses exceeding



**TABLE 6** | Committed effective dose<sup>a</sup> to humans (in  $\mu$ Sv) from the ingestion of various marine organisms (annual consumption).

Radionuclide	DC <sup>b</sup> nSv/Bq	<i>Mugil cephalus</i>	<i>Gracilaria</i>	<i>Portunus trituberculatus</i>	<i>Konosirus punctatus</i>	<i>Porphyra</i>	<i>Sinonovacula constricta</i>	<i>Largehead hairtail</i>	<i>Crassostrea gigas</i>	<i>Parapenaeopsis hardwickii</i>	<i>Mantis shrimp</i>	<i>Larimichthys polyactis</i>	<i>Porphyra</i> (Ningde NPP)
<sup>210</sup> Po	1,200	137.62	187.16	2,510.17	126.61	36.70	2,023.92	1,972.54	2,941.38	812.87	1,317.47	949.88	41.59
<sup>210</sup> Pb	690	46.78	97.07	1.41	11.25	17.94	49.94	5.98	22.86	4.92	10.55	0.14	2.46
<sup>137</sup> Cs	13	/	0.01	/	/	/	/	0.05	0.02	0.02	0.02	/	/
<sup>90</sup> Sr	28	0.53	0.04	0.56	0.11	0.13	0.07	0.63	0.71	1.07	0.14	0.04	0.13
<sup>238</sup> U	45	6.10	1.15	1.58	0.57	0.30	2.36	0.34	0.76	1.63	1.49	0.09	0.16
<sup>226</sup> Ra	280	22.98	1.86	17.41	3.43	1.14	5.14	1.28	0.57	7.56	5.00	1.57	0.29
<sup>40</sup> K	6.2	38.27	33.69	25.88	34.03	29.52	55.27	20.13	24.11	17.98	17.51	22.91	16.12
Sum		252.30	320.97	2,557.01	176.01	85.72	2,136.70	2,000.96	2,990.41	846.06	1,352.18	974.63	60.74

<sup>a</sup>Indicates that the value was below the MDA. <sup>b</sup>The annual per capita consumption rates of the Chinese population (50.97 kg/year) are for all types of seafood combined, whereas the dose calculations in this study conservatively assumed that the entire consumption consisted solely of a given sample species. The assumed exposure time is 50 years for adults. <sup>c</sup>DC (dose coefficients) radionuclide-specific committed effective dose coefficients for adult human ingestion (ICRP, 2012).

1,000  $\mu$ Sv. *Crassostrea gigas* produced radiation doses close to 3,000 Sv. The contribution of <sup>210</sup>Po to these committed effective doses was 43–99%. Based on this assumed consumption, the committed effective dose in 1 year ( $\sim$ 0.06 mSv) was far below the 1-m Sv standard (Ministry of Environmental Protection of China, 2002). It is, therefore, safe to consume these marine organisms. The contributions of different nuclides ranked in the order <sup>210</sup>Po > <sup>210</sup>Pb/<sup>40</sup>K > <sup>226</sup>Ra > <sup>238</sup>U > <sup>90</sup>Sr > <sup>137</sup>Cs, where <sup>210</sup>Po was the dominant contributor to the committed effective dose at 43–99%. In contrast, the artificial nuclides <sup>90</sup>Sr and <sup>137</sup>Cs contributed only 0.03–2.2%, and thus made the lowest contribution. This suggests that the committed effective dose from anthropogenic nuclides <sup>90</sup>Sr and <sup>137</sup>Cs is negligible when the NPPs are in operation.

## CONCLUSIONS

The activity levels of <sup>210</sup>Po and <sup>210</sup>Pb in fishes (*Mugil cephalus*, *Konosirus punctatus*, *Largehead hairtail*, *Larimichthys polyactis*), crustaceans (*Mantis shrimp*, *Parapenaeopsis hardwickii*, *Portunus trituberculatus*), bivalve mollusks (*Crassostrea gigas*, *Sinonovacula conzcta*), and macroalgae (*Gracilaria*, *Porphyra*) collected in coastal waters adjacent to Fuqing and Ningde NPPs were in the range 0.60–48.09 Bq/kg<sub>freshweight</sub> and 0.07–2.76 Bq/kg<sub>freshweight</sub>, respectively. The activity ratios of <sup>210</sup>Po/<sup>210</sup>Pb were in the range 1.1–189.7; calculated BCFs of <sup>210</sup>Po and <sup>210</sup>Pb in marine organisms were 636–44,944 and 3–1,226 L/kg, respectively. The radiation dose rates in the studied marine organisms, ranging from 0.037 to 1.531  $\mu$ Sv/h, were markedly lower than the ERICA ecosystem screening benchmark of 10  $\mu$ Gy/h, suggesting that there were no detectable irradiation effects on the marine organisms studied. The committed effective dose to humans from ingestion of these marine organisms was in the range of 0.06–2.99 mSv. Overall, when the Fuqing and Ningde NPPs are in operation, <sup>210</sup>Po is the dominant radiation dose contributor to both marine organisms and humans, and the dose contributions from artificial nuclides <sup>90</sup>Sr and <sup>137</sup>Cs can be considered negligible.

## DATA AVAILABILITY STATEMENT

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding author/s.

## ETHICS STATEMENT

The animal study was reviewed and approved by Third Institute of Oceanography.

## AUTHOR CONTRIBUTIONS

WM designed this work and performed the data analysis. JS performed the sample analysis and radiation assessment. JS and WM wrote the manuscript together. FW and JW edited this manuscript. All authors contributed to the article and approved the submitted version.

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