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Polycyclic aromatic hydrocarbons in benthos of the northern Bering Sea Shelf and Chukchi Sea Shelf

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ABSTRACT

Eighteen polycyclic aromatic hydrocarbons (PAHs) were detected in benthos collected onboard the 'Snow Dragon' in the Northern Bering Sea Shelf and Chukchi Sea Shelf during the 6^{th} Chinese National Arctic Research Expedition (CHINARE 2014). Σ_{18}PAHs for all biota samples ranged from 34.2 to 128.1 ng/g dry weight (dw), with the highest concentration observed in fish muscle (*Boreogadus saida*) samples close to St. Lawrence Island. The PAH composition pattern was dominated by the presence of lighter 3 ring (57%) and 2 ring (28%) PAHs, indicating oil-related or petrogenic sources as important origins of PAH contamination. Concentrations of alkyl-PAHs (1-methylnaphthalene and 2-methylnaphthalene) were lower than their parent PAH (naphthalene) in all biological tissue, and their percentage also decreased significantly ($p<0.05$) compared with those in the corresponding sediment. There were no significant relationships between PAH concentrations and trophic levels, which is possibly due to the combined results of the complex benthic foodweb in the subarctic/Arctic shelf region, as well as a low assimilation/effective metabolism for PAHs. According to toxic potency evaluation results from TCDD toxic equivalents (TEQs) and BaP-equivalent (BaPE) values, whelk (*Neptunea heros*) and starfish (*Ctenodiscus crispatus*) are two macroinvertebrate species showing relatively higher dioxin-like toxicity and carcinogenic risk.

Introduction

Transfer to the ocean environment and burial in benthic sediments is deemed as one of the long-term global sinks for hydrophobic chemicals (Dachs et al., 2002; Jonsson et al., 2003; Ma et al., 2015). Polycyclic aromatic hydrocarbons (PAHs) are a class of hydrophobic compounds that mainly originate from incomplete combustion and pyrolysis of carbonaceous materials (Lohmann et al., 2007; Manzetti, 2013). These contaminants are of special concern because of their carcinogenic and mutagenic characteristics (Kim et al., 2013; Lemieux et al., 2015) and are listed as persistent organic pollutants (POPs) under the Aarhus Protocol (UNECE, 1998). PAHs are quite ubiquitous in the marine environment. This group of chemicals tends to adsorb rapidly on suspended materials, sediments and biological tissues once released into the marine environment (Dachs and Eisenreich, 2000), as they possess relatively high organic-carbon/water and octanol/water partitioning coefficients (e.g., $\log K_{OC} \geq 4.5$, $\log K_{OW} \geq 3.4$) (Ma et al., 2010). Therefore, the sedimentary PAHs possess toxic potential toward bottom-dwelling organisms, and continually transfer

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and accumulate in high-trophic-level animals (Harvey et al., 2014).

For the remote pelagic regions, such as the North Pacific, North Atlantic and Arctic Oceans, previous field observation and modeling results have demonstrated long range atmospheric transport (LRAT) from mid-latitude regions and subsequent atmospheric deposition as typical sources and environmental behavior for PAHs (Ding et al., 2007; Ma et al., 2013; Sofowote et al., 2011; Wang et al., 2010). Transport through surface ocean currents to the pristine high latitude oceans is also non-negligible (Ma et al., 2018). Meanwhile, PAHs in the pelagic system might be transferred to the benthic system by the formation of deep seawater (Lohmann et al., 2006), vertical settlement with attached organic-rich particles (Dachs et al., 2002), as well as gradual movement from coastal to continental shelf regions within sediment (Jonsson et al., 2003). Thus, these contaminants are further exposed to the benthos, especially those living in the shallow shelf regions.

The Bering and Chukchi Sea shelf regions represent important regional sinks for PAHs. Due to the inflow of nutrient-rich Pacific seawater through the Bering Strait to the Arctic, the Chukchi Sea Shelf region is known for high primary production and rich benthos resources (Grebmeier et al., 2006), and especially the diversity of both infaunal and epifaunal populations, during the open water period in summer (Dunton et al., 2014). Recent climate change has induced increasing coastal erosion and permafrost thawing, especially in the Arctic shelf region through turbidity currents or ice rafting of sediments (AMAP, 2012), which is deemed as a PAH source. In addition to continental sources, the Chukchi shelf is also threatened by commercial oil and natural gas extraction activities. In spite of the low percentage of PAHs (0.2% - 7%) in the components of crude oil, the resulting oil seep is considered as one of the natural sources of PAHs (Ma et al., 2017; Yunker et al., 2011). Several researches have discussed the biogeochemical processes of PAHs in this highly studied high-latitude region: such as the atmospheric deposition process from the North Pacific to the high Arctic (Ke et al., 2017; Ma et al., 2013); the use of PAHs as biomarkers to trace terrigenous organic carbon (Ma et al., 2017; Yunker et al., 2011); as well as their toxicological response in Arctic cod (Harvey et al., 2014). However, regarding their presence and potential toxic potency in benthos, only concentrations in whelk *(Neptunea heros)* have been reported (Harvey et al., 2014).

The aim of this study is (1) to determine the residue levels and composition of PAHs in representative benthic marine organisms in the Northern Bering Sea Shelf and Chukchi Sea Shelf; (2) to discuss the possible relationship between PAH contamination status and the trophic levels of benthos; (3) to evaluate the potential toxic potency, typical dioxin-like toxicity and carcinogenic risk of PAHs in benthos of the Arctic shelf region.

1. Materials and methods

1.1. Field sampling

A total of twelve benthic organism samples were collected during the 6th Chinese National Arctic Research Expedition (CHINARE 2014) onboard 'Snow Dragon' in the Northern Bering Sea Shelf and Chukchi Sea Shelf by a trawl net (Fig. 1). The benthos included: whelk (*Neptunea heros*), scallop (*Delectopecten randolphi*), starfish (*Ctenodiscus crispatus*), crab (*Hyas crab*), shrimp (*Lepidepecreum* sp.) and fish (*Boreogadus saida*). Sampling details are listed in **Appendix A Table S1**. These benthos were selected because they are widely distributed in the Bering and Chukchi Sea Shelf and thus are representative organisms for investigating the status of contamination.

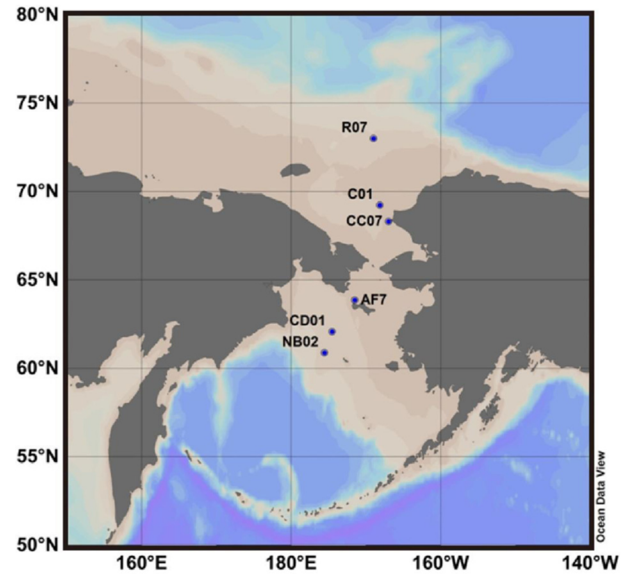

Fig. 1 – Sampling sites in the Bering Sea Shelf and Chukchi Sea Shelf.

1.2. Extraction, analysis, QC/QA

A total of 16 parent-PAHs (US EPA priority control) and 2 alkylated-PAHs, including naphthalene (Nap), 1-methylnaphthalene (1-MN), 2-methylnaphthalene (2-MN), acenaphthylene (Acl), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fluor), pyrene (Py), benzo[a]anthracene (BaA), chrysene (Chry), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (InP), dibenz[a,h]anthracene (DBahA) and benzo[ghi]perylene (BghiP) were analyzed in this research. The DCM and *n*-hexane (Picograde) we used were purchased from LGC Standards (Wesel, Germany). The surrogate and injection standards, ^{13}C-PCB-208 and PAH-Mix 9 deuterated, were from LGC Standards (Wesel, Germany).

All samples were immediately freeze-dried at -55°C for ≥ 48 hr, and then ground to fine powder. The extraction, fractionation and analysis procedure followed our previously published method (Zhong et al., 2012). Briefly, about 1 g samples were spiked with surrogate standards (PAH-Mix 9 deuterated, see Appendix A Table S2) and Soxhlet extracted with DCM for 24 hr. Then, the extracts were evaporated and further cleaned on a self-packed silica column. The first fraction, eluted with *n*-hexane, was collected and concentrated. Finally, ^{13}C–PCB 208 was added as an injection standard. The samples were analyzed with an Agilent 6890N gas chromatograph coupled to an Agilent 5975 mass spectrometer (GC-MS) (Agilent Technologies, Avondale, PA, USA), operating in electron impact and selective ion monitoring modes (SIM), and fitted with a HP-5MS capillary column (30 m × 250 μm i.d.; 0.25 μm film thickness, J&W Scientific) for chromatographic separation.

The PAH-Mix surrogate standards (including 9 deuterated compounds) were spiked in five procedural blanks to monitor matrix effects, with the injection standard ^{13}C–PCB 208. The recoveries of target PAHs ranged from (60 ± 3)% to (91 ± 7)% in the spiked samples. The method detection limits (MDLs) for each PAH were quantified as the mean level plus three times the standard deviation (3σ) of the procedural blanks. MDLs ranged from 0.005 ng/g for BkF to 0.25 ng/g for Nap. The major PAH contamination found in the procedural blanks comprised Nap, Phe and Py with concentrations of (0.14 $\pm$ 0.04), (0.95 $\pm$ 0.035) and (0.085 $\pm$ 0.035) ng/g, which accounted for $<$ 1.1% of those compounds in the organism samples.

1.3. *Stable isotope analysis and trophic level calculations*

The detailed protocol for stable C and N isotopes was based on our previously published method (Li et al., 2016). Briefly, subsamples of powdered muscle tissue were analyzed using an IsoPrime 100 isotope ratio mass spectrometer (IsoPrime Corporation; Cheadle, UK) and vario ISOTOPE cube elemental analyzer (Elementar Analysensysteme GmbH; Hanau, Germany). The reference standards for carbon and nitrogen were VPDB and AIR, respectively. The intra-lab standards used to normalize δ^{13}C and δ^{15}N values were USGS 24 (-16.1‰) and USGS 26 (53.7‰), respectively. The analytical errors of δ^{13}C and δ^{15}N values were <0.1‰.

For trophic level calculations, it was assumed that scallop collected in the Chukchi Sea Shelf, which showed the lowest δ^{15}N value of all isotope analysis results, occupied the secondary trophic position (trophic level (TL) =2.0). A recent northern Chukchi Sea benthic ecosystem report relating to the biogeochemical characteristics also confirmed that Bivalvia generally occupied the secondary trophic position (Dunton et al., 2014). The trophic levels of other organisms were calculated based on Eq. (1) (Fisk et al., 2001):

$$TL = 2.0 + \left(\delta^{15}N_{benthos} - \delta^{15}N_{scallop}\right)/3.8 \quad (1)$$

where, $\delta^{15}N_{benthos}$ is the δ^{15}N value in the specified benthos; $\delta^{15}N_{scallop}$ is the δ^{15}N value for scallop. For the Arctic marine food web, the widely used trophic enrichment factor for δ^{15}N is 3.8 (Hobson et al., 2002).

2. Results and discussion

2.1. *PAH concentrations*

PAHs were detected in all of the biota samples. The detailed concentration information of 18 detected PAHs (including 16 parent PAHs and 2 alkyl-PAHs) in biota samples is listed in **Table 1**. Generally, the total concentrations of 18 PAHs (Σ_{18}PAHs) for all biota samples ranged from 34.2 to 128.1 ng/g dry weight (dw), with a mean of (69.3 $\pm$ 31.2) ng/g dw. The two highest Σ_{18}PAHs were found in fish (128.1 ng/g dw) and starfish (118.4 ng/g dw) at Station AF7 close to St. Lawrence Island and Station R07 in the northern Chukchi Sea shelf, respectively; while the lowest total PAH concentrations of 34.2 ng/g dw were observed in scallop at Station CC07 in the Chukchi Sea Shelf. Generally, the detected PAH concentrations for macro-invertebrates in this study (34-118 ng/g dw) were just slightly lower than those in mussels from the Mediterranean Sea (25.1-390 ng/g dw) (Baumard et al., 1998a, 1998b), in macro-invertebrates from Bohai Sea, China (19.3-215.3 ng/g dw) (Wan et al., 2007) and shellfish from Guimaras Island, Philippines, which was seriously polluted by an oil spill (448 ng/g dw) (Uno et al., 2017). The observed fish concentrations in this research (36-128 ng/g dw) were comparable to those detected in the Mediterranean Sea (14.7-139 ng/g dw) (Baumard et al., 1998b), and even slightly higher than those from Bohai Sea, China (8.7-58.7 ng/g dw) (Wan et al., 2007), but lower than fishes from the South China Sea associated with oil and gas activities (199-606 ng/g dw) (Yu et al., 2019) (Appendix A Table S3). These comparison results further demonstrated that even in the high latitude sub-Arctic/Arctic shelf regions, the benthos were still threatened by PAH contamination. Increasing continental inputs induced by recent climate change and oil extraction activities would exacerbate the potential risk of PAH exposure to this vulnerable ecosystem.

It was noticeable and interesting that the total concentration of PAHs decreased in adult fishes. At the same sampling site (both CD01 and AF7), the adult fishes (including a pregnant one) showed lower total concentrations of PAHs compared to the larval ones (p<0.05), which indicated that the fishes were capable of depurating these PAHs. Previous studies also reported that for the invertebrate Northern Neptunea whelks collected at the Chukchi Sea Shelf, the larger ones showed lower concentrations of PAHs in foot muscles compared to smaller ones (Harvey et al., 2014).

Total concentrations of eight typically carcinogenic PAHs (BaA, Chry, BaP, BbF, BkF, DBahA, BghiP and IP) ranged from 0.25 to 19.2 ng/g dw, with an average of (5.7 $\pm$ 6.4) ng/g dw, and accounting for 8.2% on average of the observed Σ_{18}PAHs. It was notable that whelk and crab samples collected in Station NB02 of the Bering Sea Shelf displayed the highest total concentrations of eight carcinogens (19.2 and 15.2 ng/g dw), accounting for 26.3% and 37.4% of the eighteen total PAHs, respectively. BaP, which is reported as a highly possible carcinogen, displayed relatively high concentrations in two whelk samples (4.2 and 4.4 ng/g dw), about 10-1000 times higher than all other benthic organisms (ranging from 0.004 to 1.06 ng/g dw). Therefore, the whelks in the Northern Bering and Chukchi Sea Shelf may have relatively higher potential for carcinogenic risk.

2.2. *PAH composition*

Independent of the species, the PAH composition pattern was dominated by the presence of lighter 3 ring (57%) and 2 ring (28%) PAHs, followed by 4 ring (10%) PAHs, while the contributions of relatively heavier 5-6 ring PAHs were quite low (5.6%). Specifically, the 3-ring Phe, on average contributing 47% to Σ_{18}PAHs, was the most abundant PAH, followed by the 2-ring Nap (15%) and 4-ring Py (7.1%) (Appendix A Fig. S1). It was reported that the lower molecular weights (two and three ring PAHs) mainly originated from petrogenic sources, whereas the higher molecular weights, such as 4 and 5 ring PAH species, were predominantly of pyrogenic origin (Larsen and Baker, 2003). Therefore, to some extent, the high presence in all samples of a low-ring PAH profile indicated oil-related or petrogenic sources as important origins of PAH contamination.

During the 4th Chinese National Arctic Research Expedition (CHINARE 2010), surface sediment samples were also collected in the Bering Sea Shelf and Chukchi Sea Shelf and analyzed for PAHs. Both molecular diagnostic ratios (MDR) and the statistical source apportionment method indicated that petrogenic origin PAHs, such as oil seeps and coastally/terrestrially eroded material due to permafrost thawing, particularly affected the Bering/Chukchi Sea shelf sediment (Ma et al., 2017). Then during the feeding process, PAHs could be bioaccumulated in benthos through desorption from sediments. Thus, the benthos and sediments share the same PAH sources.

Compared with our previously reported sedimentary PAH profile, the composition pattern of PAHs in benthic organisms showed a different style behavior (Fig. 2). Specifically, concentrations of the two investigated alkyl-PAHs (1-MN and 2-MN) were less abundant than their parent PAH (Nap) in all biological tissue. Percentages of alkyl-PAHs also decreased significantly (p<0.05) compared with those in the corresponding sediment. Similarly, in the coast area of Guimaras Island (Philippines) associated with an oil spill, the parent PAHs

Table 1 – Concentrations (ng/g dw) of PAHs in benthos of the Bering and Chukchi Sea shelf.

Benthos	Station	Nap	2-MN	1-MN	Acl	Ace	Flu	Phe	Ant	Fluor
Whelk	NB02	1.76	1.22	0.52	1.23	1.38	3.82	12.67	1.08	1.66
Crab	NB02	10.81	5.80	2.91	0.04	1.01	2.81	25.09	4.92	0.30
Starfish	NB02	8.07	1.73	0.81	0.06	1.59	1.80	24.95	1.76	2.38
Fish-adult	CD01	8.03	1.45	0.91	0.10	0.23	0.75	12.66	0.95	2.10
Fish-larval	CD01	7.08	2.59	1.43	0.07	0.32	1.51	30.65	2.61	6.94
Fish-adult/pregnant	AF7	31.60	4.17	2.06	0.03	0.73	1.95	34.66	2.37	4.49
Fish-larval	AF7	22.11	3.91	2.10	0.90	0.77	2.39	66.80	5.29	9.02
Scallop-1	CC07	3.40	1.95	0.96	0.19	5.94	3.09	44.91	3.96	8.71
Scallop-2	CC07	1.74	1.12	0.52	0.10	2.30	1.31	18.25	1.14	2.21
Whelk	C01	17.64	3.26	1.74	0.05	0.45	1.86	17.55	1.40	1.54
Shrimp	C01	3.87	2.88	1.62	0.05	4.20	4.75	37.27	2.93	2.69
Starfish	R07	11.51	4.06	2.36	0.08	3.20	2.82	63.22	5.54	5.99

Benthos	Station	Py	Chry	BaA	BbF	BkF	BaP	InP	DBahA	BghiP	Sum
Whelk	NB02	0.12	0.02	0.11	n.d.	0.43	4.37	2.22	1.43	6.61	40.65
Crab	NB02	n.d.	7.43	8.67	0.05	0.36	0.10	0.97	1.26	0.36	72.88
Starfish	NB02	3.22	0.13	0.02	0.02	0.01	0.25	0.33	0.19	0.21	47.54
Fish-adult	CD01	2.82	0.01	0.01	0.01	0.02	0.08	0.44	5.05	0.49	36.10
Fish-larval	CD01	8.33	0.54	0.21	0.14	0.05	0.04	0.26	0.04	0.11	62.93
Fish-adult/pregnant	AF7	6.14	0.12	0.03	0.01	0.003	0.004	0.06	0.09	0.03	88.54
Fish-larval	AF7	12.97	n.d.	n.d.	0.06	0.01	0.14	0.87	0.40	0.33	128.07
Scallop-1	CC07	11.17	0.44	0.09	0.30	0.04	0.32	0.12	0.35	0.73	86.68
Scallop-2	CC07	3.02	0.60	0.07	0.06	0.27	0.35	0.24	0.17	0.71	34.18
Whelk	C01	2.33	0.06	0.01	0.002	0.004	0.08	0.04	0.02	0.04	48.09
Shrimp	C01	1.57	0.09	0.05	0.24	0.25	4.23	0.72	0.22	0.28	67.91

n.d.: not detected.

Table 2 – The calculated 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD, pg/g) toxic equivalents (TEQs) and BaP-equivalent (BaPE, ng/g dw) for all benthos

Benthos	Station	TEQs	BaPE
Whelk	NB02	0.087	5.449
Crab	NB02	0.071	1.476
Starfish	NB02	0.006	0.389
Fish-adult	CD01	0.028	3.149
Fish-larval	CD01	0.010	0.105
Fish-adult/pregnant	AF7	0.002	0.067
Fish-larval	AF7	0.014	0.457
Scallop-1	CC07	0.009	0.567
Scallop-2	CC07	0.034	0.501
Whelk	C01	0.045	4.460
Shrimp	C01	0.001	0.095
Starfish	R07	0.057	6.395

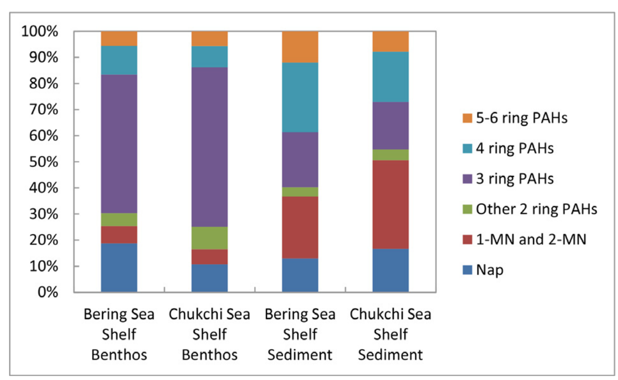

Fig. 2 – PAH composition profile of benthos and corresponding sediment in the Bering Sea Shelf and Chukchi Sea Shelf.

in shellfish also degraded more slowly than the alkyl-PAHs (Uno et al., 2017). This is likely attributable to the efficient metabolic transformation of alkyl-PAHs and persistent characteristics of parent PAHs.

2.3. *Relationship with trophic level status*

For all benthos samples collected, the stable $\delta^{13}C$ isotope signatures were in the range of -21.2‰ to -14.8‰, with an average of (-18.6 ± 1.7)‰ (Appendix A Table S4). Benthic food webs were quite complex in the Bering and Chukchi Sea Shelf region. A recent food web model of the northern Chukchi Sea included two possible/distinctive carbon source end-members preferred by benthic consumers, typically pelagic and benthic carbon (Dunton et al., 2014). For field measurement results in this study, the starfishes displayed the greatest ^{13}C enrichment characteristics (mean stable $\delta^{13}C$ isotope signatures of -15.8‰), followed by the crab and shrimp sample ($\delta^{13}C$ value of -17.9‰ and -18.3‰, respectively), which indicated their closer link to the ^{13}C-enriched carbon in the sediment; for example, the benthic microalgae, ice algae, or microbially degraded organic matter, etc. Some benthos showed relatively higher values of $\delta^{13}C$, such as some whelk and fish samples (with some $\delta^{13}C$ value of -21.2‰ and -20.1‰, respectively), which indicated

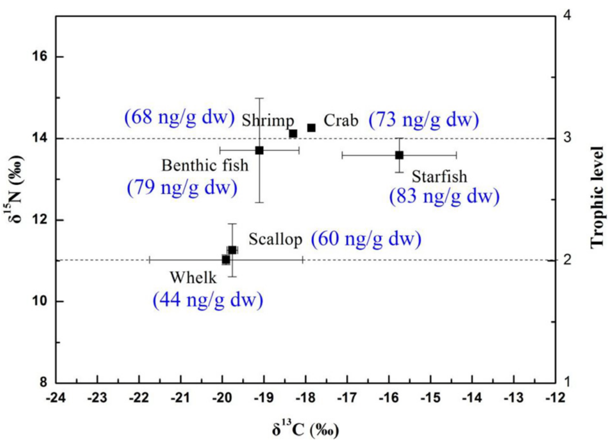

Fig. 3 – Mean (±S.E.) stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope values in benthos from the Bering and Chukchi Sea Shelf. Isotopically determined trophic level (TL) is plotted on the secondary y-axis. Whelk samples were assumed to occupy TL 2.0. Average values of PAH concentrations are displayed in blue. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

their possible/partial food source from the pelagic system - ^{13}C-depleted phytoplankton carbon.

Moreover, the range of $\delta^{15}N$ isotope values (from 10.8‰ to 14.8‰) was not as large (Appendix A Table S4). Thus, these calculation results indicated that the benthos generally occupied the second and third trophic levels. In detail, the derived general trophic level positions for fish, shrimp, crab and starfish were 2.8, 2.9, 2.9 and 2.7 respectively, while the mean placements for whelk and scallop were at the second trophic level (TL=2.1). Considering the relationship between the concentrations of total/different ringed PAHs and trophic levels, there were no significant Pearson/statistical correlations between PAH concentrations and trophic levels (Appendix A Table S5). This is partly/likely due to the complex benthic foodweb in this Arctic shelf region. In addition, PAHs could be efficiently metabolized because of the catalytic function of Cytochrome P450, which is commonly found in marine organisms (Valery et al., 2001). Previous research also demonstrated that the gut assimilation efficiencies of PAHs were lower than those of PCBs in the same K_{ow} (octanol-water partition coefficient) ranges (Thomann and Komlos, 1999). Unlike other typical POPs that showed discernible food web biomagnification, such as polychlorinated biphenyls (PCBs), trophic dilution of PAHs through the marine food web had previously been confirmed in Bohai Bay, North China (Wan et al., 2007), the coastal region of the Ariake Sea, Japan (Nakata et al., 2003), as well as the Baltic Sea (Broman et al., 1990).

2.4. *Toxic potency evaluation*

Several studies indicated that PAHs could elicit dioxin-like activity, with BkF showing the greatest toxic potential. In order to evaluate the potential risks of the observed PAHs, 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) toxic equivalents (TEQs) were calculated for all benthos. In this research, for the relative potencies (REP) of several priority PAHs, typically Chry, BaA, BbF, BkF, BaP, IP and DBahA, we used data from *in-vitro* H4IIE luciferase assays (Appendix A Table S6) (Villeneuve et al., 2002). Total TEQ concentrations ranged from 0.001-0.087 pg/g TEQs. These levels were comparable with those of tidal flat species of the Ariake Sea, Japan (Nakata et al., 2003). Whelk, crab and starfish showed relative higher TEQs concentrations (0.087, 0.071 and 0.057 pg/g TEQs, respectively), which indicated their higher potential for dioxin-like toxicity. On average, BkF accounted for 44% of the TEQs.

Moreover, in order to evaluate the carcinogenic risk of PAHs, the commonly used BaP-equivalent (BaPE) was also calculated. BaPE includes the risk assessment all carcinogenic PAHs (BaP, BaA, BkF, BbF, DBahA and InP). In this index, the weight of BAP is set as 1, and the relative carcinogenicity weights of other PAH are listed in Appendix A Table S6 (Liu et al., 2009). Results showed that BaPE ranged from 0.067-6.4 ng/g dw, with an average of (1.9 $\pm$ 2.3) ng/g dw. Whelk and starfish showed relative higher BaPE concentrations (6.4, and 5.0 ng/g dw), which indicated their higher potential carcinogenic risk. Considering both dioxin-like toxicity and carcinogenicity risk, whelk and starfish in the Bering and Chukchi shelf areas are benthos with relatively higher toxic potential.

Declaration of Competing Interest

None

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Appendix A Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2020.04.021.

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