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Polycyclic aromatic hydrocarbons (PAHs) in wild marine organisms from South China Sea: Occurrence, sources, and human health implications

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ABSTRACT

Concentrations of 16 US EPA priority polycyclic aromatic hydrocarbons (PAHs) were measured in 15 marine wild organism species from South China Sea. The concentration (dry weight) of 16 PAHs ranged from 94.88 to 557.87 ng/g, with a mean of 289.86 ng/g. The concentrations of BaP in marine species were no detectable. The composition of PAHs was characterized by the 2- and 3-ring PAHs in marine species, and NA, PHE and FA were the dominant constituents. PAHs isomeric ratios indicated PAHs mainly originated from grass, wood and coal combustion, and petroleum. The human health risk assessment based on the excess cancer risk (ECR) suggested the probability of PAHs posing carcinogenic risk to human beings with consumption of marine organisms were negligible (probability $< 1 \times 10^{-6}$).

Polycyclic aromatic hydrocarbons (PAHs) are a large group of about 10,000 different organic compounds with 2 to 7 fused aromatic rings with natural as well as anthropogenic sources (Wenzl et al., 2006; Haritash and Kaushik, 2009; Kim et al., 2013; Purcaro et al., 2013; Gu et al., 2016a). PAHs have gathered significant environmental concern because of their carcinogenicity and mutagenicity (Xia et al., 2012; Zhang et al., 2012; Purcaro et al., 2013; Gu et al., 2016a). PAHs are widespread environmental contaminants in the global biogeochemical cycles. So far >200 PAHs have been found and 16 PAHs are listed as priority pollutants by the European Union (EU) and the US Environmental Protection Agency (US EPA) (Sverdrup et al., 2002; Navarro et al., 2009; Gu et al., 2013).

Marine organisms comprising fish and cephalopods, is consumed by humans worldwide for their high protein and low saturated fat content and for omega fatty acids, which are known for their health benefits (Naylor et al., 2000; Gu et al., 2015a; Gu et al., 2015b; Gu et al., 2016b). The consumption wild marine organisms associated with adverse human health effects is an important problem. The accumulation of PAHs in aquatic organisms can pose a long-term burden on biogeochemical cycling in the ecosphere (Gu et al., 2015a). When PAHs enter the food chain, the may accumulate to dangerous levels and be harmful to human health (USEPA, 2000a; Almeida et al., 2012; Bandowe et al., 2014; Gu et al., 2015a). For this reason, analysis of chemical quality of aquatic organisms, particularly the contents of PAHs in wild marine organisms is extremely important. South China Sea is an important fishery zone based on our institute's numerous fishery resource surveys from 1960s to the present (Wang and Yuan, 2008; Yang et al., 2009; Zhang et al., 2013; Yan et al., 2014; Zhang et al., 2016). However, concentrations of PAHs in wild marine organism species of South China Sea are unknown. Therefore, the present study was designed to (1) determine concentrations of PAHs in marine organisms from South China Sea, and (2) identify potential source and evaluate human health risk of PAHs.

From March to April 2015, a total of 294 marine organism samples were collected at 19 sampling stations in South China Sea (Fig. 1). The samples were of fifteen species in total, made up of ten fish species and five cephalopod species, with the ranges in physical characteristics of samples summarized in Table 1. Once the organisms were on deck, the organisms were slaughtered or stunned at −20 °C refrigeration house, then transported to the laboratory and rinsed four times with distilled water. The edible tissues were removed from each organism, and dried using a vacuum freeze-drying instrument, then ground gently to get a homogeneous powder, and stored in a brown glass bottle at −20 °C until analysis.

Extraction and measurement methods for 16 PAHs in marine organisms have been described in more detail in our previous study (Sun et al., 2016). The 16 US EPA priority PAHs such as naphthalene (NA), acenaphthylene (ACL), acenaphthene (AC), fluorene (FL), phenanthrene

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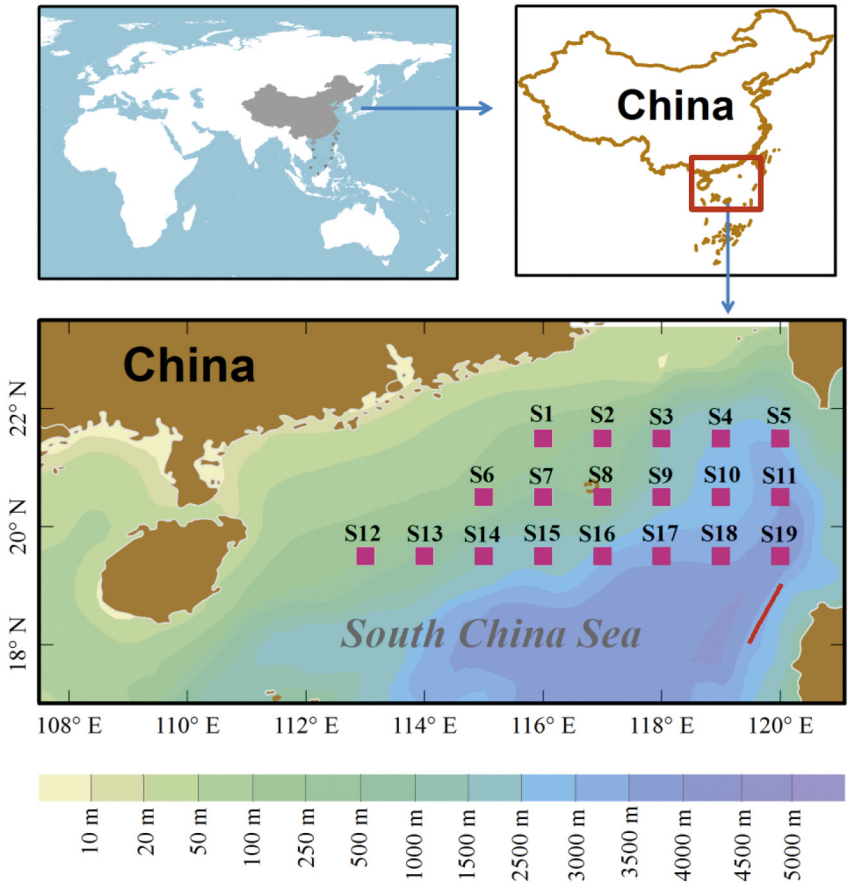

Fig. 1. Sampling stations in South China Sea.
Gridded bathymetric data are from http://www.bodc.ac.uk/data/online_delivery/gebco/.

(PHE), anthracene (AN), fluoranthene (FA), pyrene (PY), benzo[*a*]anthracene (BaA), chrysene (CHR), benzo[*b*]fluoranthene (BbFA), benzo[*k*]gluoranthene (BkFA), benzo[*a*]pyrene (BaP), Indeno[1,2,3-*c*,*d*]pyrene (IP), dibenzo[*a*,*h*]anthracene (DBahA), and benzo[*g*,*h*,*i*]perylene (BghiP) were analyzed by an Agilent 7890A gas chromatograph coupled with a 5975C mass spectrometer.

All analytical data were subject to strict quality assurance and quality control. Analyses of a reagent blank, a spiked blank, matrix spike, and sample duplicate were processed with every batch of 10 samples. The surrogate recoveries (mean ± standard deviation) were 68.7 ± 8.4%, 88.5 ± 7.2%, 95.4 ± 9.4%, and 87.7 ± 7.6% for acenaphthene-d_{10}, chrysene-d_{12}, perylene-d_{12} and phenanthrene-d_{10}, respectively. Recoveries of individual PAHs ranged from 79% for ACL to 117% for BbFA. The relative percent analysis difference between duplicate samples was <15%. The method detection limits (S/N = 3) varied from 0.01 to 0.16 ng/g.

The potency equivalent concentrations (PEC) of 16 total PAHs for marine species were calculated with the following Eq. (1):

$$PEC = \sum TEF_i \times C_i \quad (1)$$

Table 1
Characteristics of wild marine organism samples caught and analyzed as part of the study.

Groups of organisms	Species	Standard body length (mm)	Body weight (g)	N[a]	Feeding habit	Sampling stations
Fish	*Auxis tapeinosoma*	146–247	25.5–251.5	50	Piscivore	S2, S3, S4, S5, S6, S7, S9, S10, S11
	Brama japonica	118–179	37–137	30	Benthivore–piscivore	S7, S8, S9, S12, S13
	Decapterus macrosoma	179–215	68–145.5	25	Zooplankton	S9, S13, S15, S16
	Cubiceps squamiceps	116–152	26–39.5	30	Zooplankton	S7, S8, S9, S13
	Gemphylus serpens	446–830	128–918	20	Benthivore–piscivore	S3, S4, S7 S12
	Mene maculata	161–165	105–120	8	Benthivore–Zooplankton	S1
	Auxis thazard	231–242	179.5–218	7	Piscivore-invertebrate	S13
	Macrura reevesi	171–207	101.5–186	10	Planktivore	S9, S13
	Boesemanichthys firmamentum	244.5–367	245–260	5	Piscivore-invertebrate	S12
	Gastrophysus spadiceus	237–261	355–492	4	Benthivore	S18
Cephalopods	*Symplectoteuthis oualaniensis*	120–234	64–774.5	80	Benthivore–piscivore	S2, S3, S4, S5, S6, S7, S8, S9, S11, S12, S13, S14, S15, S16, S17, S19
	Liocranchia reinhardti	134–212	98–419	6	Benthivore–piscivore	S1
	Leachia pacifica	23.5–47.5	80.5–109	7	Benthivore–piscivore	S4
	Todarodes pacificus	14–17	81–91	8	Benthivore–zooplankton	S1
	Argonauta argo	136–226.5	110–130	4	Benthivore–zooplankton	S1

[a] No. of samples.

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