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RESEARCH ARTICLE

Occurrence, tissue distribution, and risk assessment of progestins, androgens, estrogens, and phenols in wild freshwater fish species

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Abstract

The presence of endocrine-disrupting chemicals (EDCs) in aquatic environments such as water, sediment, and sludge received more and more attention. However, the bioaccumulate properties of EDCs, particularly progestins and androgens, in various tissues of different wild freshwater fish species, as well as their effects on human health, have not been fully studied. The muscle, liver, and gills of three wild fish species obtained from the East Dongting Lake in southern China were examined for the presence of 19 EDCs (4 progestins, 5 androgens, 6 estrogens, and 4 phenols). Seventeen analytes were detected in all fish samples, and the concentrations of progestins, androgens, estrogens, and phenols ranged from ND–78.80 ng/g (wet weight, ww), ND–50.40 ng/g ww, ND–3573.82 ng/g ww, and ND–88.17 ng/g ww, respectively. The bioaccumulation of some EDCs in wild fish from East Dongting Lake was species-specific. Additionally, AND, EES, P4, and E2 were discovered in the liver at higher levels than in the muscle, suggesting that livers had a larger ability for enriching these EDCs than the muscle. Furthermore, the relationships between the fish sizes and the EDC concentrations indicated that total weight and length had a negligible impact on the bioaccumulation of EDCs in various fish species. Most importantly, the effects of EDCs on human health as a result of fish consumption were assessed. Although the estimated daily intakes (EDIs) of most EDCs were much lower compared with the corresponding acceptable daily intakes (ADIs) via consuming fish collected in this study, the EDI of EE2 in *Silurus asotus* was higher than the ADI of E2, indicating that *Silurus asotus* from East Dongting Lake should be eaten in moderation by local residents.

Keywords Endocrine-disrupting chemicals · Freshwater fish species · Tissue distribution · Progestogens · Androgens · Health risk assessment

Introduction

Since endocrine-disrupting chemicals (EDCs) can interfere with the endocrine systems of organisms, the occurrence of EDCs in the environment is of great concern (Liang et al. 2020; Šauer et al. 2020; Thrupp et al. 2018). Among these EDCs, progestins, androgens, and estrogens—natural and synthetic sex hormones—as well as phenols with estrogenic characteristics, are of particular interest, since they can modify gene expression, disrupt reproduction function, delay sexual development, and affect sexual differentiation in aquatic organisms even at nanograms per liter levels (Huang et al. 2019; Kidd et al. 2007; Liang et al. 2019, 2018; Purdom et al. 1994). Additionally, they also pose a risk to human health such as obesity, diabetes, breast cancer, and prostate cancer and lead to abnormal development of the reproductive organs (Diamanti-Kandarakis et al. 2009).

Fish are sensitive indicator organisms for the effects of exposure to contaminants in the aquatic environment (Ellestad et al. 2014). As consumers at high trophic levels, fish can accumulate a considerable amount of EDCs through the gills, oral intake, and surface skin (Jia et al. 2017). Therefore, selecting fish as the subject could indicate not only the direct effects of contaminants on biota, such as bioconcentration, but also the indirect trophic transmission

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along food chains, such as biomagnification (Fan et al. 2019). Moreover, due to their high nutritional value, fish are commonly consumed by human beings (Jia et al. 2017). According to the Food and Agriculture Organization of the United Nations (2019), the fish production and supply in Asia were the highest in the world owing to the enormous demands for fish as the daily diet of Asians (Cheung et al. 2008). Therefore, consumption of fish exposed to EDCs may pose a great threat to human health (Jia et al. 2016). In addition, a chemical's capacity for bioaccumulation could be utilized to predict long-term negative consequences that were not always addressed by acute toxicity and short-term exposure tests (Nallani et al. 2012). Thus, the studies on bioaccumulation of progestins, androgens, estrogens, and phenols in fish are significant because present regulatory efforts to identify the environmental and human risks of contaminants are based on bioaccumulation assessments (Arnot and Gobas 2006). However, research on EDCs in the environment has been predominantly focused on estrogens and phenols, and little focus has been placed on the bioaccumulation of progestins and androgens in fish despite their prevalence in pharmaceutical therapeutic applications and their existence in the aquatic environment.

Fish feeding habits are the primary determinants of the transmission of pollutants among food webs (Arnot and Gobas 2004). Accordingly, contaminant residues in different fish species are influenced by feeding behaviors. However, the species-specific distributions of EDCs, particularly progestogens and androgens, in wild freshwater fish with various feeding behaviors in the lake have been less reported. In addition, the toxic effects of chemicals on aquatic organisms typically correlate with the absorption and accumulation of these chemicals in specific tissues (Ismail et al. 2021). Therefore, determining the contamination of EDCs in multiple tissues instead of only muscle is more comprehensive.

Dongting Lake, located in northeastern Hunan province, China, is the first large lake in the downstream region of the Three Gorges Reservoir and exhibits a highly dynamic hydrological regime and high turbidity (Dai et al. 2005). Dongting Lake is an important international wetland with numerous ecological functions. However, Dongting Lake has recently experienced the accumulation of contaminants and serious deterioration of the lake ecosystem (Müller et al. 2008). Therefore, three wild edible freshwater fish species were obtained from the East Dongting Lake and used to analyze the distribution characteristics of the 19 EDCs (4 progestins, 5 androgens, 6 estrogens, and 4 phenols) in their muscle, gill, and liver tissues. As far as we know, the concentrations of progestins and androgens in wild freshwater fish have not yet been reported, despite that progestins and androgens were generally considered to disrupt the reproductive system of aquatic organisms. Additionally, the relationships between the measured EDCs and fish sizes among different fish species were investigated. Finally, the effects of target EDCs on human health by fish consumption in this area were estimated in order to reduce and control them.

Materials and methods

Sampling

In September 2017, 69 fish samples in total were obtained from an expert fisherman using fishing nets in the vicinity of Yueyang City in Dongting Lake. The fishes included 22 *Ctenopharyngodon idella* (*C. idella*, total length 37.3 ± 1.44 cm and total weight 0.96 ± 0.08 kg), 19 *Carassius auratus* (*C. auratus*, total length 21.4 ± 1.31 cm and total weight 0.34 ± 0.05 kg), and 28 *Silurus asotus* (*S. asotus*, total length 34.2 ± 6.14 cm and total weight 0.27 ± 0.15 kg). These three species, which are categorized as herbivorous, omnivore, and carnivorous fish, respectively, were the most prevalent edible species in the lake. The fish samples were promptly dissected after measuring the live total length and total weight. The muscle, liver, and gills were collected, cleaned with ultrapure water, dried and homogenized, and then stored at − 20 °C.

Chemical and sample preparation

In total, 19 EDCs, including 4 progestins (progesterone (P4), levonorgestrel (LNG), medroxyprogesterone (MP), and norethindrone (NET)), 5 androgens (testosterone (TES), androstenedione (AND), methyltestosterone (MT), 19-nortestosterone (19-NT), and stanozolol (ST)), 6 estrogens (estrone (E1), 17β-estradiol (E2), 17α-ethinylestradiol (EE2), diethylstilbestrol (DES), dienestrol (DIE), and hexestrol (HEX)), and 4 phenols (bisphenol A (BPA), 4-nonylphenol (4-NP), 4-*tert*-octylphenol (4-*t*-OP), and 4-octylphenol (4-OP)), were chosen for this study and determined simultaneously with 4 internal standards (progesterone-d_9; testosterone-d_3; 17β-estradiol-2,4-d_2; and bisphenol A-d_{16}). In the Text S1 and Text S2 of Supplementary materials, the specifics relating to chemical, material, and sample preparation were covered. The concentrations of EDCs were reported as nanograms per gram wet weight (ww) for fish samples.

LC–MS analysis and QA/QC

Nineteen target analytes were analyzed by the method described in our previous study based on Agilent 1260–6460 liquid chromatography-mass spectrometry (LC–MS, Agilent Technologies, USA) (Luo et al. 2019). More information on instrumental analysis was shown in Text S3 of the Supplementary materials.

To guarantee the effectiveness and reproducibility of the analytical method, quality assurance and quality control (QA/QC) were carried out. The method detection limit (MDL) varied from 0.14–1.70 ng/g ww. The method quantitation limit (MQL) varied from 0.43–5.67 ng/g ww. There were no target EDCs found in the procedure blanks. The recoveries ranged from 52.7 to 119%. The RSDs ranged for repeatability and reproducibility were 0.45–14.84% and 0.54–23.97%, respectively. Specific method performance parameters are displayed in Table S1.

Risk characterization

Only the fish muscle was investigated for its potential health risks because the muscle was the primary tissue ingested by the local population. The highest concentrations of EDCs detected in fish muscles (given in Table 1) were used to estimate the health risks to locals after consuming fish based on "a worst-case scenario."

Estimated daily intake (EDI) of each EDC by fish consumption was calculated by employing the following equation:

$$EDI_i = \frac{C_i \times DIR}{BW} \tag{1}$$

where C_i (ng/g) is the concentration of the compound i, DIR (g/day) is the daily ingestion rate of fish muscle for consumers, and BW (kg) is the body weight for residents. In this study, BW and DIR for people living in Hunan Province, China, are 58.7 ± 12.0 kg and 0.060 ± 0.046 kg/day, respectively (Jia et al. 2018).

Statistical analysis

Target EDC concentrations below the MDL were not considered while determining the detection frequency. For purposes of statistical analysis, the data below the MQL were set to zero (Lu et al. 2017). The significant differences ($p < 0.05$) between concentrations of different EDCs for each tissue were evaluated by the Mann–Whitney *U* test through SPSS software (IBM, USA). Moreover, the comparison of concentrations of each EDC between different species and tissues was also performed by the Mann–Whitney *U* test. The Spearman correlation analysis was developed to evaluate the correlation between concentrations of each EDC and fish sizes by SPSS software.

Results and discussion

Occurrence of EDCs in wild freshwater fish

Target EDC concentrations in the wild freshwater fish from East Dongting Lake are summarized in Table 1. Seventeen analytes (including 4 progestogens, 5 androgens, 5 estrogens, and 3 phenols) were detected in all fish samples containing the muscle, liver, and gills, but E1 and 4-*t*-OP were not found in any fish samples.

As for estrogens, interestingly, synthetic estrogen DES was detected with the highest detected frequency (DFs = 15.53%) among target estrogens despite being listed as a banned veterinary drug in China since 2011 (Yang et al. 2021). Our previous study also showed that the DES had the highest detection frequency in the muscle of wild fish from the Changsha section of the Xiangjiang River (Zhou et al. 2019). The Xiangjiang River flows into Dongting Lake eventually. The high detection frequencies of DES in wild fish in Xiangjiang River and Dongting Lake suspected that DES was still illegally used by residents for livestock production, and the supervision of relevant drugs should be further strengthened. Compared with previous studies, the E2 concentrations (ND–16.46 ng/g) in fish species from East Dongting Lake in the present study were lower than those in Taihu Lake, China (4.91–364 ng/g dw) (Wang et al. 2015). The concentrations of DES in fish in this study (ND–3.21 ng/g) were the same as those in Loma Lake, China (0.11 ng/g) (Dan et al. 2017). The concentration of EE2 in fish in the present study (ND–3574 ng/g) was much higher than that in Xiangjiang Lake, China (ND–27.3 ng/g) (Zhou et al. 2019) and in Loma Lake, China (1.18 ng/g,). To date, there is no existing data pertaining to the concentrations of DIE and HEX in fish.

Among the three phenols, the detection frequency of BPA (28.64%) and the average concentration of BPA (2.05 ng/g) were the highest. The BPA levels measured in fish samples from East Dongting Lake (ND–88.17 ng/g) were in accordance with the finding from Dianchi Lake (ND–83.5 ng/g) (Liu et al. 2011) and higher than those from the Netherlands (1–11 ng/g) (Belfroid et al. 2002). BPA is primarily used in the manufacture of epoxy resins and polycarbonate plastics (Gyllenhammar et al. 2012; Natalie et al. 2010), and its high detected concentration may be related to industrial development and sewage and wastewater discharge in lakeside cities. The concentration of 4-NP (ND–0.35) in fish in the current study was lower than that in Xiangjiang River (ND–148 ng/g) (Zhou et al. 2019).

In addition, the detection frequency of natural progestin P4 (24.27%) in this study was the highest among the target progestins, which might be related to its endogenous existence in the aquatic environment and fish. Natural progestogens, which served as sex pheromones in teleost fish, are essential for oocyte growth and maturation, spermatogenesis, and sperm maturation (Kime 1990; Kobayashi et al. 2002; Nagahama and Yamashita 2008; Scott et al. 2010). The concentrations of P4 in this study (ND–20.26 ng/g) were higher than those in the muscle of cultured fish (ND–5.4 ng/g) in three freshwater aquaculture farms in

Table 1 Concentrations and detection frequencies of EDCs in three different fish species

EDC		Muscle			Liver			Gills		
		C. idella (*n* = 22)	*C. auratus* (*n* = 19)	*S. asotus* (*n* = 28)	*C. idella* (*n* = 22)	*C. auratus* (*n* = 19)	*S. asotus* (*n* = 28)	*C. idella* (*n* = 22)	*C. auratus* (*n* = 19)	*S. asotus* (*n* = 28)
P4	Range (ng/g)	ND–6.96	ND	ND–2.38	ND–20.26	ND–3.41	ND–4.92	ND–11.90	ND–2.33	ND–7.61
	Mean (ng/g)	0.48 ± 1.50^{ab}	ND	0.20 ± 0.49^{ab}	1.09 ± 4.36^{ab}	0.87 ± 1.22	0.54 ± 1.22^{ab}	1.27 ± 2.75^{c}	0.35 ± 0.68	0.58 ± 1.59
	DF (%)	27.27	0	32.14	9.09	36.84	21.43	40.91	26.32	22.22
MP	Range (ng/g)	ND	ND–1.54	ND–0.65	ND–23.98	ND–78.80	ND–45.31	ND–0.66	ND–1.60	ND–14.36
	Mean (ng/g)	ND^{b}	0.08 ± 0.35	0.02 ± 0.12^{b}	1.43 ± 5.18^{ab}	7.89 ± 18.9	3.67 ± 10.3^{ab}	0.03 ± 0.14^{bc}	0.19 ± 0.48	1.04 ± 3.04
	DF (%)	0	5.26	3.57	13.64	31.58	17.86	4.55	15.79	18.52
LNG	Range (ng/g)	ND–5.21	ND–1.80	ND–23.35	ND–25.62	ND	ND	ND–5.08	ND–3.58	ND
	Mean (ng/g)	0.40 ± 1.18^{ab}	0.09 ± 0.41	1.01 ± 4.41^{ab}	1.16 ± 5.46^{ab}	ND	ND^{b}	0.44 ± 1.44^{ac}	0.19 ± 0.82	ND
	DF (%)	18.18	5.26	14.29	4.55	0	0	9.09	5.26	0
NET	Range (ng/g)	ND–5.00	ND–0.39	ND–3.35	ND–28.66	ND–61.02	ND	ND–24.69	ND–14.60	ND–28.49
	Mean (ng/g)	0.42 ± 1.14^{ab}	0.02 ± 0.09	0.16 ± 0.64^{ab}	1.43 ± 6.11^{ab}	6.43 ± 14.9	ND^{b}	**3.65 ± 7.43^{ac}**	1.87 ± 3.77	**1.30 ± 5.51**
	DF (%)	18.18	5.26	14.29	9.09	26.32	0	36.36	31.58	11.11
19-NT	Range (ng/g)	ND–1.18	ND–1.09	ND–3.90	ND	ND–1.59	ND–1.00	ND–3.41	ND–0.28	ND–1.41
	Mean (ng/g)	0.05 ± 0.25^{b}	0.07 ± 0.25	0.14 ± 0.74^{b}	ND^{b}	0.08 ± 0.37	0.04 ± 0.19^{b}	0.16 ± 0.73^{bc}	0.01 ± 0.06	0.11 ± 0.33
	DF (%)	4.55	10.53	3.57	0	5.26	3.57	4.55	5.26	11.11
AND	Range (ng/g)	ND–0.54	ND–0.57	ND–1.04	ND–45.07	ND–2.30	ND	ND–1.78	ND–1.76	ND–2.27
	Mean (ng/g)	0.05 ± 0.16^{ab}	0.14 ± 0.24	0.08 ± 0.26^{ab}	**3.47 ± 9.65^{a}**	**0.21 ± 0.63**	ND^{b}	0.21 ± 0.47^{ac}	0.41 ± 0.70	0.14 ± 0.53
	DF (%)	9.09	26.32	10.71	40.91	10.53	0	22.73	26.32	7.41
MT	Range (ng/g)	ND–5.58	ND–0.51	ND–1.12	ND–50.40	ND–3.53	ND–1.62	ND–2.32	ND	ND–2.07
	Mean (ng/g)	0.36 ± 1.20^{ab}	0.05 ± 0.15	0.06 ± 0.24^{ab}	3.07 ± 11.2^{ab}	0.19 ± 0.81	0.13 ± 0.42^{ab}	0.11 ± 0.49^{bc}	ND	0.22 ± 0.65
	DF (%)	18.18	10.53	7.14	9.09	5.26	10.71	4.55	0	11.11
TES	Range (ng/g)	ND–0.65	ND–0.50	ND–0.95	ND–17.52	ND–8.24	ND–2.95	ND–0.70	ND–1.32	ND–3.12
	Mean (ng/g)	0.06 ± 0.16^{ab}	0.09 ± 0.17	0.13 ± 0.26^{ab}	1.41 ± 3.93^{ab}	1.06 ± 2.19	0.18 ± 0.67^{b}	**0.05 ± 0.17^{ac}**	0.19 ± 0.46	**0.40 ± 0.86**
	DF (%)	13.64	21.05	21.43	22.73	26.32	7.14	9.09	15.79	22.22
ST	Range (ng/g)	ND	ND–0.53	ND	ND–4.94	ND–3.26	ND–3.61	ND–0.81	ND–2.12	ND–2.36
	Mean (ng/g)	ND^{b}	0.06 ± 0.12	ND^{b}	0.22 ± 1.05^{ab}	0.50 ± 1.08	0.17 ± 0.71^{b}	0.04 ± 0.17^{bc}	0.17 ± 0.54	0.09 ± 0.45
	DF (%)	0	5.26	0	4.55	21.05	7.14	4.55	10.53	3.70
4-NP	Range (ng/g)	ND–0.35	ND	ND	ND	ND	ND	ND	ND	ND
	Mean (ng/g)	0.02 ± 0.07^{b}	ND	ND^{b}	ND^{b}	ND	ND^{b}	ND^{b}	ND	ND
	DF (%)	4.55	0	0	0	0	0	0	0	0
4-*t*-OP	Range (ng/g)	ND–1.04	ND–2.02	ND–1.27	ND–17.24	ND	ND–5.54	ND–23.56	ND–8.82	ND–10.05
	Mean (ng/g)	0.05 ± 0.22^{b}	0.16 ± 0.51	0.19 ± 0.42^{ab}	1.04 ± 3.76^{ab}	ND	0.71 ± 1.60^{ab}	2.18 ± 6.48^{ac}	0.46 ± 2.02	1.14 ± 2.53
	DF (%)	4.55	10.53	17.86	13.64	0	17.86	13.64	5.26	22.22

Table 1 (continued)

EDC		Muscle			Liver			Gills		
		C. idella ($n = 22$)	*C. auratus* ($n = 19$)	*S. asotus* ($n = 28$)	*C. idella* ($n = 22$)	*C. auratus* ($n = 19$)	*S. asotus* ($n = 28$)	*C. idella* ($n = 22$)	*C. auratus* ($n = 19$)	*S. asotus* ($n = 28$)
BPA	Range (ng/g)	ND–1.94	ND–3.58	ND–1.81	ND–12.42	ND–27.55	ND–23.14	ND–88.17	ND–3.73	ND–30.57
	Mean (ng/g)	0.05 ± 0.72^{a}	0.61 ± 1.11	0.41 ± 0.64^{a}	1.80 ± 3.84^{ab}	1.90 ± 6.51	2.86 ± 5.13^{a}	**7.09 ± 18.9^{ac}**	**0.39 ± 1.16**	2.55 ± 6.61
	DF (%)	36.36	26.32	35.71	22.73	10.53	35.71	40.91	10.53	29.63
DES	Range (ng/g)	ND	ND–0.75	ND–0.71	ND–1.83	ND	ND–2.06	ND–2.34	ND–3.21	ND–3.02
	Mean (ng/g)	ND^{b}	0.08 ± 0.23	0.08 ± 0.22^{ab}	0.02 ± 0.44^{ab}	ND	0.18 ± 0.54^{ab}	0.38 ± 0.72^{ac}	0.27 ± 0.84	0.29 ± 0.65
	DF (%)	0	10.53	10.71	22.73	0	14.29	31.82	10.53	33.33
DIE	Range (ng/g)	ND–0.69	ND	ND–0.25	ND–12.54	ND–17.81	ND–3.64	ND–11.01	ND–2.78	ND–2.41
	Mean (ng/g)	0.03 ± 0.15^{b}	ND^{b}	0.01 ± 0.05^{ab}	0.57 ± 2.67^{ab}	1.97 ± 5.06	0.27 ± 0.88^{ab}	1.25 ± 3.25^{ac}	0.15 ± 0.64	0.09 ± 0.46
	DF (%)	4.55	0	3.57	4.55	15.79	10.71	18.18	5.26	3.70
E2	Range (ng/g)	ND	ND–1.37	ND	ND–3.40	ND–16.46	ND–14.31	ND	ND–1.92	ND
	Mean (ng/g)	ND^{b}	0.07 ± 0.31^{b}	ND	0.15 ± 0.73^{b}	0.87 ± 3.78	1.31 ± 3.31^{ab}	ND^{b}	0.17 ± 0.52	ND
	DF (%)	0	5.26	0	4.55	5.26	17.86	0	10.53	0
EE2	Range (ng/g)	ND	ND	ND–73.12	ND–40.90	ND–28.96	ND–6.00	ND–3573	ND–2.41	ND–36.48
	Mean (ng/g)	ND^{b}	ND	2.90 ± 13.85^{ab}	**6.96 ± 13.45^{ab}**	1.52 ± 6.64	**0.64 ± 1.67^{ab}**	163 ± 762^{ac}	0.13 ± 0.55	3.37 ± 9.92
	DF (%)	0	0	7.14	27.27	5.26	14.29	13.63	5.26	11.11
HEX	Range (ng/g)	ND	ND–0.75	ND	ND	ND	ND–0.48	ND	ND	ND–1.59
	Mean (ng/g)	ND^{b}	0.08 ± 0.23^{b}	ND^{ab}	ND^{b}	ND	0.02 ± 0.09^{b}	ND^{b}	ND	0.11 ± 0.39
	DF (%)	0	10.53	0	0	0	3.57	0	0	7.41

Different lowercase letters indicate the significant differences ($p < 0.05$) among the concentrations of different EDCs in each organ within the same fish species. The bolded concentrations indicate a significant difference ($p < 0.05$) of EDC concentrations between different fish species in the same tissue.

DF detection frequency, *ND* not detected, < *MQL* the concentration below the MQL

Guangzhou, China (Liu et al. 2017). The concentrations of MP, LNG, and NET in fish samples from East Dongting Lake ranged from ND–78.80 ng/g, ND–25.63 ng/g, and ND–61.02 ng/g, respectively. TES, like P4, also had the highest detection frequency among 5 targeted androgens. The concentrations of 19-NT, AND, MT, and ST in fish samples from East Dongting Lake exhibited a range of ND–3.90 ng/g, ND–45.07 ng/g, ND–50.40 ng/g, and ND–4.96 ng/g, respectively. However, the research on progestins and androgens in freshwater fish remains limited; to our knowledge, no other investigations have yet reported on the contamination characteristics of progesterone and androgen in fish.

Distributions of EDCs in different fish species and tissues

The concentrations of each EDC in different fish species are shown in Table 1. As shown in Table 1, for liver samples, the concentrations of AND in *C. idella* (ND–45.07 ng/g) were significantly higher than those of omnivorous *C. auratus* (ND–2.30 ng/g), and the concentrations of EE2 in *C. idella* (ND–40.90 ng/g) were significantly higher than those of *S. asotus* (ND–6.00 ng/g) ($p < 0.05$). For gill samples, the concentrations of BPA in *C. idella* (ND–88.17 ng/g) were significantly higher than those in *C. auratus* (ND–3.73 ng/g) ($p < 0.05$). The contents of NET (ND–24.69 ng/g) and TES (ND–0.70 ng/g) in *C. idella* were lower than those in *S. asotus* (ND–28.49 ng/g and ND–3.12 ng/g, respectively). The finding suggested that the bioaccumulation of some EDCs in fish was species-specific. However, for *C. idella*, *C. auratus*, and *S. asotus*, there was no significant difference in the EDC concentrations in the muscle among different species, indicating that the trophic role is not the only factor influencing the bioaccumulation of EDCs in the specific tissue; other factors such as ecological habits, growth dilution, and metabolic capability may also affect the bioaccumulation of EDCs in food chains (Sun et al. 2017; Yuan et al. 2012; Zhou et al. 2007).

The distributions of EDCs in different tissues of each fish species are shown in Fig. 1. In *C. idella*, the concentrations of androgen AND and estrogen EES in the muscle were significantly lower than those in the liver. Similarly, in the case of *C. auratus*, the P4 concentrations in the muscle were notably lower than those in the liver. In *S. asotus*, the levels of estrogen E2 were remarkably lower than those in the liver. Liu et al. (Liu et al. 2011) studied the distribution of E2 in wild *C. auratus* in Dianchi Lake and found that the enrichment ability of estrogens and phenols in *C. auratus* liver was significantly more potent than those in the muscle. At present, there was no research on the distribution of progestins and androgens in different fish tissues. The liver plays a crucial role in primary metabolism and is the major site for the accumulation, biotransformation, and excretion of pollutants (Moon et al. 1985). The concentrations of AND ($K_{ow} = 2.8$), EES ($K_{ow} = 3.7$), P4 ($K_{ow} = 3.9$), and E2 ($K_{ow} = 4$) in the liver exhibited higher values compared to those in the muscle, which showed higher hepatic accumulation. Interestingly, in *S. asotus*, the contents of P4 ($K_{ow} = 3.9$) in the liver were considerably lower than those in the muscle, and the LNG ($K_{ow} = 3.5$) levels in the muscle were also higher than those in both the liver and gills. *C. auratus* and *S. asotus* are herbivorous and carnivorous, respectively. These results underscore the distinct tissue-specific accumulation of EDCs with similar or even identical hydrophobicity across various fish species with disparate trophic roles. This observation further underscores the pivotal role of a trophic level of fish species in dictating the distribution of EDCs among various tissues. In *C. idella*, the levels of estrogen DES in the muscle were significantly lower than those in the gills, which may be due to the passive exchange of contaminants between fish and the aquatic environment through the gills. In addition, the gill is an important site of interaction with contaminants because it is the first organ to be in contact with water and resuspended sediment particles (Jia et al. 2016). The results suggested that water exposure might be the main exposure route of DES to *C. idella*.

Correlation analysis

Spearman correlation analysis was used to determine the relationship between the concentrations of EDCs in the analyzed tissues (muscle, liver, and gills) and the total lengths and weights of the three fishes. The correlation coefficients are shown in Table 2.

For *C. idella*, the concentrations of P4 in the muscle were negatively correlated with total lengths ($r = -0.45$, $p < 0.05$). The concentrations of BPA in the muscle were positively correlated with total weights ($r = 0.52$, $p < 0.05$). There was a positive correlation between the MT levels in the liver and total lengths ($r = 0.44$, $p < 0.05$). For *S. asotus*, the concentrations of HEX in the gills were positively correlated with total weights ($r = 0.48$, $p < 0.05$). These results indicated that P4, BPA, MT, and HEX gradually accumulated in the fish as the fish grew. However, there was no significant correlation between the total weights and lengths of the *C. auratus* and the concentrations of EDCs. The levels of pollutants in fish tissues depend on the combined effects of feeding behavior, habitat, geographic location, life stage, and other factors on the pollutant intake and elimination rate. Juvenile fish have higher

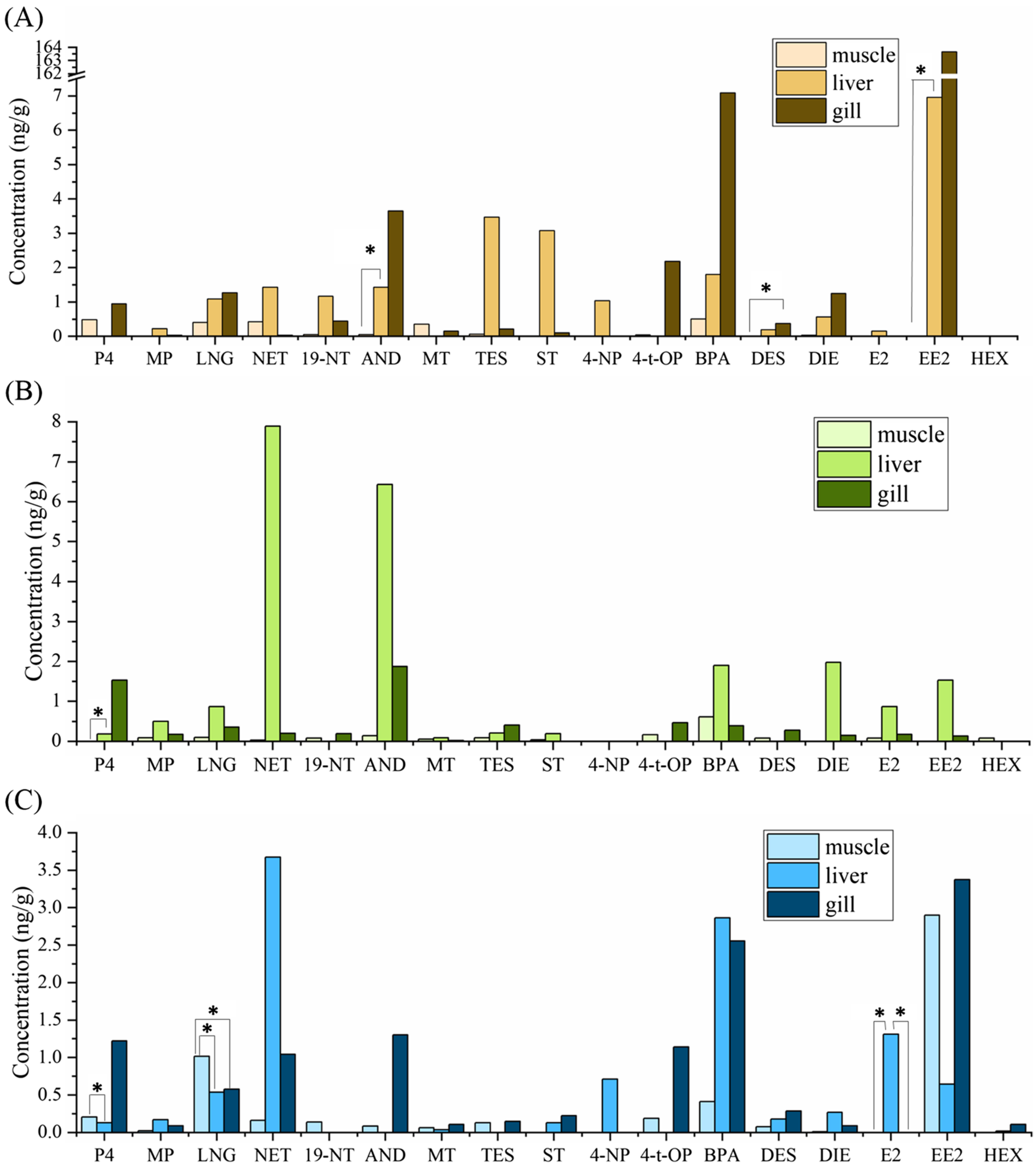

Fig. 1 Concentrations of EDCs in different tissues **A** *C. idella*, **B** *C. auratus*, and **C** *S. asotus*. *$p < 0.05$ indicate the significant differences between different tissues

metabolic capability and intake rate than adult fish, and adult fish accumulate more EDCs as they grow. Considering that fish have been exposed to EDCs for a long time, a balance between the rate of uptake and elimination of EDCs in the fish might account for the weak correlation (Jia et al. 2017). Therefore, the correlation between total weight and length and EDC concentration of different fish species was various, and the influence of body size on the bioaccumulation of EDCs in different fish species was relatively small.

Health risk assessment

Diet is a crucial pathway for humans taking EDCs. Previous clinical observations and epidemiological analysis indicated that EDCs might have harmful effects on the neurological and reproductive systems, leading to the onset of obesity and cancer (Diamanti-Kandarakis et al. 2009). Since all studied fish are traded species and valuable for fish exports, the EDIs of diverse EDCs of residents after consuming these three contaminated fish species were

Table 2 Spearman correlation coefficients between the EDCs and fish sizes

EDC	*C. idella*						*C. auratus*						*S. asotus*					
	Weight			Length			Weight			Length			Weight			Length		
	Muscle	Liver	Gills	Muscle	Liver	Gills	Muscle	Liver	Gills	Muscle	Liver	Gills	Muscle	Liver	Gills	Muscle	Liver	Gills
P4	−0.19	0.08	0.01	−0.45*	0.00	0.18	/	0.04	−0.02	/	0.03	0.00	0.21	0.07	0.31	−0.09	0.23	0.03
MP	/	−0.01	/	/	0.12	/	/	0.32	/	/	0.11	/	/	−0.10	0.36	/	−0.12	0.39
LNG	0.37	−0.05	−0.23	0.09	0.10	−0.10	/	/	0.00	/	/	0.15	−0.25	/	/	0.14	/	/
NET	−0.28	0.18	−0.03	0.14	0.25	−0.22	/	0.01	0.17	/	0.00	−0.19	−0.28	/	0.40	−0.25	/	0.45
19-NT	−0.28	/	−0.05	−0.26	/	0.10	0.24	0.24	/	0.15	0.15	/	0.03	0.18	0.06	−0.18	0.20	0.33
AND	/	0.01	−0.05	/	0.09	0.10	/	0.04	0.16	/	−0.27	0.28	/	/	−0.16	/	/	−0.40
MT	0.09	0.18	−0.05	0.00	0.44*	0.10	/	0.00	/	/	0.15	/	−0.13	0.27	/	/	0.33	/
TES	/	−0.40	/	/	−0.16	/	/	−0.27	0.05	/	−0.16	−0.18	/	−0.17	−0.11	−0.33	−0.12	−0.13
ST	/	0.02	−0.33	/	0.00	−0.33	/	−0.12	0.27	/	−0.32	0.22	0.06	0.08	0.02	/	0.18	0.05
CAF	/	/	−0.27	/	/	0.11	/	0.02	0.10	/	0.39	0.05	/	0.32	−0.17	0.00	0.32	−0.12
4-NP	/	/	/	/	/	/	/	/	/	/	/	/	/	/	/	/	/	/
4-*t*-OP	/	−0.07	−0.31	/	−0.24	−0.14	/	/	−0.22	/	/	−0.09	/	0.12	0.30	/	0.20	0.39
BPA	0.52*	0.05	0.28	0.20	0.02	0.23	0.35	−0.03	0.09	0.25	0.22	0.06	0.15	−0.20	−0.27	0.10	−0.12	−0.20
DES	/	0.14	−0.20	/	0.27	−0.12	0.34	/	0.04	0.21	/	0.00	−0.21	−0.05	0.20	−0.13	0.09	0.23
DIE	−0.33	0.02	0.12	−0.33	0.00	0.03	/	−0.07	−0.30	/	−0.19	−0.29	/	0.05	−0.02	/	0.08	0.12
E2	/	/	/	/	/	/	/	−0.13	/	/	−0.09	/	/	−0.10	/	/	−0.05	/
EE2	/	0.23	0.22	/	0.02	0.25	/	0.09	/	/	0.15	/	0.03	−0.13	−0.30	0.08	−0.32	−0.15
HEX	/	/	/	/	/	/	/	/	/	/	/	/	/	/	0.48*	/	/	−0.17

* $p < 0.05$

Table 3 EDIs and ADIs of the muscle in three different fish species from East Dongting Lake

EDC	EDI (μg/BWkg/day)			ADI (μg/BWkg/day)
	C. idella	*C. auratus*	*S. asotus*	
Progestins				
P4	0.0071	/	0.0024	30
MP	/	0.0016	0.0007	/
LNG	0.0053	0.0018	0.0239	/
NET	0.0051	0.0004	0.0034	/
Androgens				
TES	0.0007	0.0005	0.0010	2
AND	0.0006	0.0006	0.0011	/
MT	0.0057	0.0005	0.0011	/
19-NT	0.0012	0.0011	0.0040	/
ST	/	0.0005	/	/
Estrogens				
E2	/	0.0014	/	0.05
DES	/	0.0008	0.0007	/
DIE	0.0007	/	0.0003	/
EE2	/	/	0.0747	/
HEX	/	0.0008	/	/
Phenols				
BPA	0.0020	0.0037	0.0018	5
4-NP	0.0004	/	/	/
4-*t*-OP	0.0011	0.0021	0.0013	/
Total EDI (μg/BWkg/day)	0.0299	0.0158	0.1164	/

calculated according to Eq. (1). The EDIs of EDCs in wild *C. idella*, *C. auratus*, and *S. asotus* in East Dongting Lake are shown in Table 3.

ADI is the acceptable daily intake of a pollutant that does not cause obvious health risks in a person's life (Diogo et al. 2013). When EDI of EDC is higher than ADI, EDC will hurt human health. *C. idella*, *C. auratus*, and *S. asotus* are typical wild freshwater fish often eaten by residents. According to the Joint FAO/WHO Expert Committee on Food Additives (JECFA) and General Administration of Quality Supervision, Inspection, and Quarantine of the People's Republic of China, the ADI of P4, TES, E2, and BPA is 30 μg/BWkg/day, 2 μg/BWkg/day, 0.05 μg/BWkg/day, and 5 μg/BWkg/day, respectively. There was no ADI for other EDCs at present, so the ADIs of these four substances were used as the representatives of the four kinds of substances to be compared with EDIs of other EDCs. As shown in Table 3, the EDIs of most EDCs were much lower compared with the corresponding ADIs. However, the EDI of EE2 in *S. asotus* (0.0747 μg/BWkg/day) was higher than the ADI of E2 (0.05 μg/BWkg/day), indicating that *S. asotus* from East Dongting Lake should be eaten in moderation by local residents. Given the composite nature of EDCs in the fish, the total EDIs of three fish species were compared. As shown in Table 3, the total EDI of *S. asotus* was higher than those of *C. idella* and *C. auratus*, also revealing that consumption of carnivorous *S. asotus* posed higher health risks than the ingestion of *C. idella* and *C. auratus* from East Dongting Lake.

Conclusions

Seventeen analytes (including 4 progestogens, 5 androgens, 5 estrogens, and 3 phenols) were detected in wild freshwater fish from East Dongting Lake except E1 and 4-*t*-OP which were not found in any fish samples. The concentrations of EDCs in the muscle, liver, and gills were variable. The liver presented a higher affinity for the accumulation of AND, EES, P4, and E2 than the muscle. The trophic level of fish species played an important role in the distribution of EDCs in various tissues. According to the results of the Spearman correlation analysis, the influence of total weight and length on the bioaccumulation of EDCs in different fish species was relatively small. In addition, the risk assessment showed that *S. asotus* from East Dongting Lake should be eaten in moderation by local residents.

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Data availability The data will be available on request.

Declarations

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