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# 白洋淀优势鱼类中喹诺酮类抗生素的生物累积特征及其与环境因子相关性

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**摘要:** 喹诺酮类抗生素(quinolone antibiotics, QNs)易富集于水生生物中,近年来在我国湖泊中广泛检出,且其生物富集系数和营养传递行为具有明显的时空异质性. 本研究选取白洋淀9种优势鱼类为研究对象,分析14种QNs的生物累积特征及其与环境因子的相关性,评估了QNs健康风险. 结果表明,白洋淀水体中 $\sum$ QNs质量浓度范围为0.740 0~1 590 ng·L<sup>-1</sup>,其中氟甲喹(flumequine, FLU)、噁唑酸(oxolinic acid, OXO)和氧氟沙星(ofloxacin, OFL)检出率较高,FLU平均质量浓度最高;鱼类体内 $\sum$ QNs含量范围为17.1~146 ng·g<sup>-1</sup>,其中环丙沙星(ciprofloxacin, CIP)和FLU平均含量较高. 生物累积系数(bioaccumulation factors, BAF)范围(L·kg<sup>-1</sup>)为96.2(BAF<sub>MAR</sub>)~489(BAF<sub>CIP</sub>),表明QNs在鱼类中的生物累积能力较低. 5种检出率较高的QNs[恩诺沙星(enrofloxacin, ENR)、FLU、马波沙星(marbofloxacin, MAR)、诺氟沙星(norfloxacin, NOR)和OFL]的营养放大因子(trophic magnification factors, TMF)范围为0.714(TM<sub>MAR</sub>)~1.33(TM<sub>ENR</sub>),其中ENR呈营养放大,FLU、MAR和QNs呈营养稀释. 理化参数与BAF和TMF相关性分析结果表明,pH、T、SD、DO、COD、TP、TN、NH<sub>4</sub><sup>+</sup>-N、NO<sub>3</sub><sup>-</sup>-N和PO<sub>4</sub><sup>3-</sup>-P与BAF和TMF相关性较为显著. 健康风险评估结果表明,CIP的危害系数(hazard quotient, HQ, 0.004 0~0.026)显著高于其它QNs(≤0.005 0),危害指数(hazard index, HI)范围为0.001 0~0.035,具有高健康风险. 因此,应制定更为严格的抗生素排放标准,减少湖泊外源有机质和营养物质输入,以减少污染物在鱼类体内的生物累积,降低抗生素污染水平和健康风险.

**关键词:** 喹诺酮类抗生素(QNs); 白洋淀; 优势鱼类; 生物累积; 营养传递; 环境因子

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## Bioaccumulation Characteristics of Quinolones (QNs) in Dominant Fish Species and Their Correlation with Environmental Factors in Baiyangdian Lake

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**Abstract:** In recent years, quinolone antibiotics (QNs), which easily bioaccumulate in aquatic organisms, have been widely detected in lake ecosystems, and the bioaccumulation and trophic transfer behavior are obviously spatiotemporally different. In this study, the bioaccumulation and trophic transfer behavior of fourteen QNs in nine dominant fish species were studied, the correlation with environmental factors was analyzed, and the health risk of QNs was evaluated in Baiyangdian Lake. The results showed that the mass concentrations of  $\sum$  QNs in water varied from 0.740 0 to 1 590 ng·L<sup>-1</sup>. Furthermore, the detected frequencies of flumequine (FLU), oxolinic acid (OXO), and ofloxacin (OFL) were higher, and the average mass concentration of FLU was the highest. The content of  $\sum$  QNs in fish ranged from 17.1 to 146 ng·g<sup>-1</sup>, and the average contents of ciprofloxacin (CIP) and OFL were higher. The bioaccumulation factors (BAF) were in the range of 96.2 (BAF<sub>MAR</sub>)-489 (BAF<sub>CIP</sub>) L·kg<sup>-1</sup>, indicating the bioaccumulation of QNs was low in dominant fish species. The trophic magnification factors (TMF) of five QNs (enrofloxacin (ENR), FLU, marbofloxacin (MAR), norfloxacin (NOR), and OFL) varied from 0.714 (TM<sub>MAR</sub>) to 1.33 (TM<sub>ENR</sub>), indicating ENR exhibited trophic magnification, while FLU, MAR, and  $\sum$  QNs exhibited trophic dilution. The results of correlation analysis between environmental parameters and BAF/TMF showed that pH, T, SD, DO, COD, TP, TN, NH<sub>4</sub><sup>+</sup>-N, NO<sub>3</sub><sup>-</sup>-N, and PO<sub>4</sub><sup>3-</sup>-P were significantly related to the bioaccumulation of QNs in fish. The results of human health risk showed that the hazard quotient (HQ) of CIP (0.004 0-0.026) was significantly higher than that of other QNs (≤0.005 0), and the hazard indices (HI) ranged from 0.0010 to 0.035, indicating a high level of health risk. Therefore, to reduce the health risk, the standard and residue limits of QNs should be set in Baiyangdian Lake.

**Key words:** quinolone antibiotics (QNs); Baiyangdian Lake; dominant fish species; bioaccumulation; trophic transfer; environmental factors

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抗生素作为一种抑菌和杀菌化合物,被广泛用于人类或动物的疾病治疗,常用于畜牧业和鱼类养殖中以促进动物生长<sup>[1,2]</sup>。据第三届畜禽养殖污染防治与资源化国际会议数据统计,我国畜禽养殖抗生素年用量达 9.7 万 t,占全国抗生素年消耗量近 50%。抗生素因其分子较大,不能被机体完全吸收,约 90% 以母体化合物或代谢产物随粪便或尿液排出体外<sup>[3,4]</sup>,最终进入水环境<sup>[5]</sup>。研究表明,即使抗生素处于低浓度,对水生生物也会产生负面影响<sup>[6~8]</sup>。不同来源的抗生素长期输入,呈现假持久性,加强了抗生素在水环境中的生物累积和传递<sup>[9,10]</sup>。

按照化学结构和性质划分,抗生素包括磺胺类、喹诺酮类、四环素类、大环内酯类、聚醚类和  $\beta$ -内酰胺类等<sup>[11,12]</sup>,其中喹诺酮类抗生素(quinolone antibiotics, QNs)常用于人类和动物的疾病治疗<sup>[13]</sup>。QNs 的生物降解率较低,在地表水<sup>[14~17]</sup>、地下水<sup>[18,19]</sup>、饮用水<sup>[20,21]</sup>和海洋<sup>[22]</sup>中广泛检出。有研究表明,QNs 易富集于水生生物中<sup>[9,23,24]</sup>,海河中环丙沙星和诺氟沙星在鲫鱼体内具有生物累积作用,平均生物累积系数分别为  $3\,262\text{ L}\cdot\text{kg}^{-1}$  和  $284\text{ L}\cdot\text{kg}^{-1}$ 。Li 等<sup>[23]</sup>的研究结果表明,恩诺沙星和氟罗沙星在白洋淀鱼类中的生物累积系数分别为  $4\,490\text{ L}\cdot\text{kg}^{-1}$  和  $4\,130\text{ L}\cdot\text{kg}^{-1}$ 。莱州湾鱼体内环丙沙星、依诺沙星、恩诺沙星、诺氟沙星和氧氟沙星的平均生物累积系数分别为 159、200、6 310、501 和  $1\,000\text{ L}\cdot\text{kg}^{-1}$ <sup>[25]</sup>。珠江中环丙沙星和恩诺沙星在罗非鱼体内的生物累积系数范围分别为  $468\sim 525\text{ L}\cdot\text{kg}^{-1}$  和  $95.5\sim 1\,479\text{ L}\cdot\text{kg}^{-1}$ ,氧氟沙星在鲫鱼体内的生物累积系数范围为  $33.9\sim 1.38\times 10^4\text{ L}\cdot\text{kg}^{-1}$ <sup>[26]</sup>。不同水环境中抗生素污染情况不同,不同鱼类对抗生素的生物累积和传递作用也存在显著差异。然而此前的研究多集中于抗生素在生物体内的生物富集和营养传递行为,对抗生素生物富集和营养传递行为与环境因子的相关性研究较为缺乏。

白洋淀位于雄安新区核心区,为构建蓝绿交织的新区提供重要的生态支撑。然而,白洋淀长期遭受保定市城市污水、水产养殖废水以及周边居民生活污水排放的综合影响,抗生素污染处于较高水平<sup>[23,27,28]</sup>。喹诺酮类、四环素类和大环内酯类等多类抗生素在白洋淀水体、沉积物和水生生物中均被检出<sup>[5,14,23,28]</sup>。有研究表明,白洋淀的环境因子和 QNs 呈现显著空间差异性<sup>[27,29]</sup>,而生物体内的污染物含量与环境因素密切相关<sup>[30]</sup>。因此,为明晰抗生素生物富集和营养传递行为与环境因子的相关性,本文以白洋淀为研究区域,通过分析白洋淀优势鱼类的 QNs 含量,探讨 QNs 在优势鱼类中的生物累积

和营养传递特征,研究其与环境因子相关性,评估 QNs 的健康风险,以期为白洋淀抗生素污染排放和风险管理提供理论依据和数据支撑。

## 1 材料与方法

### 1.1 样品采集

2018 年 4 月和 8 月于白洋淀 32 个采样点采集水样和鱼类样品,采样点如图 1 所示。每个采样点采集 4 L 表层水,做好标记,保存于冷藏箱中,运回实验室于  $-20^{\circ}\text{C}$  冰柜内储存待用<sup>[27]</sup>;共采集 9 种优势鱼类样品,包括鲢鱼、鲫鱼、草鱼、鲤鱼、泥鳅、乌鳢、黄魮鱼、黄鳊和棒花鱼,每种鱼类样品不少于 10 条,共采集 197 条。两批次样品混合后进行测定分析。水样进行理化分析[水深(water depth, WD)、水温(temperature,  $T$ )、pH、溶解氧(dissolved oxygen, DO)、透明度(secchi depth, SD)、化学需氧量(chemical oxygen demand, COD)、总磷(total phosphorus, TP)、总氮(total nitrogen, TN)、氨氮(ammonia nitrogen,  $\text{NH}_4^+-\text{N}$ )、硝氮(nitrate nitrogen,  $\text{NO}_3^--\text{N}$ )和  $\text{PO}_4^{3-}-\text{P}$ 等]和 QNs 质量浓度测定;鱼类样品进行 QNs 含量测定。

依据白洋淀水体理化特征(表略),将采样点进行聚类分析(图 2),聚类结果显示 S1~S7 为生境 1, S8~S24 为生境 2, S25~S32 为生境 3。生境 1 主要受上游保定市城市污水影响,生境 2 主要受当地水产养殖废水和居民生活污水影响,生境 3 受人类活动干扰较少。

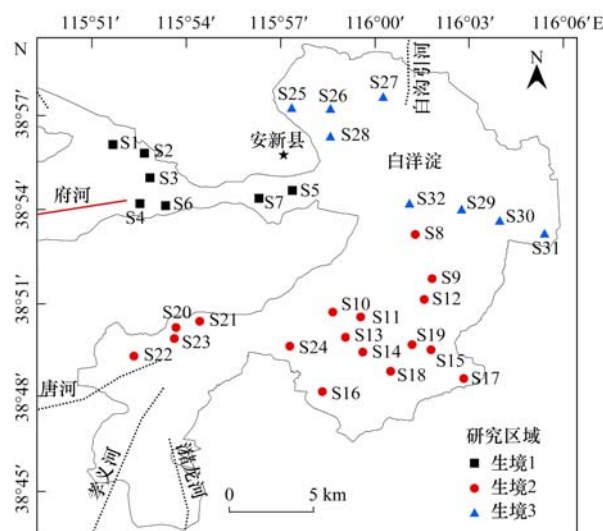


图 1 白洋淀采样点示意

Fig. 1 Sampling sites of Baiyangdian Lake

### 1.2 QNs 测定

水样预处理和 QNs 质量浓度测定方法参考文献<sup>[25]</sup>。

鱼类样品运回实验室后于  $-50^{\circ}\text{C}$  条件下冷冻干



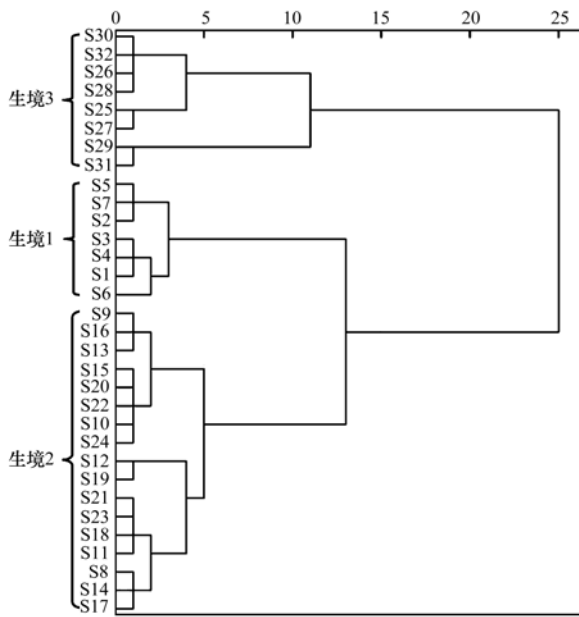


图2 依据水体理化参数的聚类分析

Fig. 2 Classification analysis results for sampling sites based on physical and chemical attributes in water

干燥,然后粉碎,过10目筛子,筛下物于 $-20^{\circ}\text{C}$ 条件下装袋标记,冷冻保存,以备分析<sup>[31]</sup>. 鱼类样品中QNs含量测定方法如下:称取0.5 g鱼粉末于暗处放置24 h后,倒入快速溶剂萃取仪(吉天APLE-3000多通道全自动快速溶剂萃取仪)的萃取池,加入适量硅藻土. 提取溶剂为丙酮/甲醇(1/1,体积比). 工作条件如下:萃取温度为 $70^{\circ}\text{C}$ ;萃取压力为1 500 psi(10 350 kPa);预热5 min;静态萃取5 min;最终萃取体积60 mL,冲洗体积为萃取池体积的60%;氮气吹扫90 s;萃取循环3次. 萃取液用旋转蒸发仪浓缩至2 mL,然后用蒸馏水稀释至500 mL. 剩余操作同水样分析.

本研究的目标抗生素为14种QNs:环丙沙星(ciprofloxacin, CIP)、达氟沙星(difloxacin, DIF)、依诺沙星(enoxacin, ENO)、恩诺沙星(enrofloxacin, ENR)、氟罗沙星(fleroxacin, FLE)、氟甲喹(flumequine, FLU)、马波沙星(marbofloxacin, MAR)、诺氟沙星(norfloxacin, NOR)、氧氟沙星(ofloxacin, OFL)、沙氟沙星(sarafloxacin, SAR)、司帕沙星(sparfloxacin, SPA)、奥比沙星(orbifloxacin, ORB)、噁唑酸(oxolinic acid, OXO)和吡哌酸(pipemidic acid, PIP). 本研究使用安捷伦1200超高效液相色谱-电喷雾串联质谱(high performance liquid chromatography-electrospray ionization tandem mass spectrometry, HPLC-ESI-MS-MS)和多反应监测(multiple reaction monitoring, MRM)分析样品中QNs含量.

### 1.3 QNs在鱼类中的富集和传递

鱼类对QNs的累积效应用生物累积系数

(bioaccumulation factors, BAF)表征<sup>[32]</sup>,计算公式如下:

$$\text{BAF} = (C_{\text{鱼}}/C_{\text{水}}) \times 1\,000 \quad (1)$$

式中, $C_{\text{鱼}}$ 为鱼体内QNs含量( $\text{ng}\cdot\text{g}^{-1}$ ), $C_{\text{水}}$ 为水体中QNs的质量浓度( $\text{ng}\cdot\text{L}^{-1}$ ).

QNs在鱼类中的营养传递水平用营养放大因子(trophic magnification factor, TMF)表征,计算公式如下:

$$\lg C_{\text{鱼}} = a + b \times \text{TL} \quad (2)$$

$$\text{TMF} = e^b \quad (3)$$

式中,TL为鱼类的营养级, $a$ 为 $y$ 轴的截距, $b$ 为斜率.

鱼类的营养级根据文献<sup>[33]</sup>计算:

$$\text{TL} = [(\delta^{15}\text{N}_{\text{鱼}} - \delta^{15}\text{N}_{\text{基准}})/2.98] + 2 \quad (4)$$

式中, $\delta^{15}\text{N}_{\text{鱼}}$ 是鱼类的同位素值, $\delta^{15}\text{N}_{\text{基准}}$ 是基准生物(中国圆田螺)的平均同位素值,其值选择之前研究<sup>[5]</sup>中的数值[10.36‰(生境1)、9.43‰(生境2)和8.74‰(生境3)]计算,2.98是每个营养级水平的预期 $\delta^{15}\text{N}$ 值<sup>[34]</sup>,2是基线生物(中国圆田螺)的理论营养级水平<sup>[33, 35]</sup>. 白洋淀鱼类同位素 $\delta^{15}\text{N}$ 分析和结果详见文献<sup>[5]</sup>.

### 1.4 健康风险评估方法

为评估食用QNs污染鱼类的潜在健康风险,根据鱼类体内QNs计算日估摄入量(estimated daily intake, EDI)<sup>[36]</sup>. 计算公式如下:

$$\text{EDI} = C_{\text{鱼}} \times \text{FR}/\text{BW} \quad (5)$$

式中, $C_{\text{鱼}}$ 为鱼体内QNs含量( $\mu\text{g}\cdot\text{g}^{-1}$ ),FR为中国成年人日估摄入鱼量( $\text{g}\cdot\text{d}^{-1}$ ),依据《中国食物与营养发展纲要(2014~2020年)》,FR取 $49.30\text{ g}\cdot\text{d}^{-1}$ ;BW为设定的成人平均体重(kg),按60 kg计算.

本研究用危害系数(hazard quotient, HQ)表征健康风险<sup>[37]</sup>,计算公式如下:

$$\text{HQ} = \text{EDI}/\text{ADI} \quad (6)$$

式中,ADI为日均可接受摄入量,依据联合国粮食农业组织,世界卫生组织和相关文献<sup>[38]</sup>可知,CIP、ENR、FLU、NOR和OFL的ADI分别为1.6、2.0、30.0、11.4和 $5.7\mu\text{g}\cdot(\text{kg}\cdot\text{d})^{-1}$ .

本研究中利用危害指数(hazard index, HI =  $\sum \text{HQ}$ )计算食用白洋淀鱼类的健康风险<sup>[9]</sup>. 当 $\text{HI} < 0.01$ 表示该健康风险可忽略;当 $\text{HI} \geq 0.01$ 表示健康风险较大<sup>[39]</sup>.

### 1.5 数据统计与分析

采用SPSS 22.0软件进行统计分析和单因素方差检验(One-way ANOVA), $P < 0.05$ 表明存在显著差异. 利用ArcGIS 10.7软件和Origin 2018软件制

图. BAF 和 TMF 与环境因子间的相关性采用 SPSS 22.0 软件进行 Spearman 秩相关分析.

2 结果与分析

2.1 白洋淀水体理化 和 QNs 的分布特征

白洋淀水体理化参数表明(表 1),大部分理化参数(COD、TP、TN、NH<sub>4</sub><sup>+</sup>-N、NO<sub>3</sub><sup>-</sup>-N 和 PO<sub>4</sub><sup>3-</sup>-P)呈现生境 1 > 生境 2 > 生境 3 的趋势,其中 DO、COD、TP 和 TN 超出Ⅲ类水水质标准

(GB 3838-2002). 白洋淀水体中共检出 11 种 QNs, FLU 检出率最高, 其次为 OXO 和 OFL; ∑ QNs 质量浓度范围为 0.740 0 ~ 1 590 ng·L<sup>-1</sup>, FLU 平均质量浓度最高(168.0 ng·L<sup>-1</sup>),除 PIP、OXO、FLU 和 ENO 外,其余 QNs 均呈现生境 1 > 生境 2 > 生境 3 的趋势(图 3). 单因素方差检验结果表明(表略),PIP、MAR、NOR 和 ENO 在 3 类生境中呈现显著差异(*P* < 0.05),其它 QNs 差异不显著.

表 1 水体理化特征

| 项目   | 水体   |      |      |      |                           |                            |                           |                           |                                                        |                                                        |                                                         |
|------|------|------|------|------|---------------------------|----------------------------|---------------------------|---------------------------|--------------------------------------------------------|--------------------------------------------------------|---------------------------------------------------------|
|      | WD/m | pH   | T/℃  | SD/m | DO<br>/mg·L <sup>-1</sup> | COD<br>/mg·L <sup>-1</sup> | TP<br>/mg·L <sup>-1</sup> | TN<br>/mg·L <sup>-1</sup> | NH <sub>4</sub> <sup>+</sup> -N<br>/mg·L <sup>-1</sup> | NO <sub>3</sub> <sup>-</sup> -N<br>/mg·L <sup>-1</sup> | PO <sub>4</sub> <sup>3-</sup> -P<br>/mg·L <sup>-1</sup> |
| 生境 1 | 2.04 | 7.97 | 8.38 | 0.48 | 4.43                      | 41.45                      | 0.49                      | 4.46                      | 0.85                                                   | 0.78                                                   | 0.09                                                    |
| 生境 2 | 2.64 | 8.62 | 9.95 | 0.65 | 5.33                      | 31.78                      | 0.23                      | 3.13                      | 0.62                                                   | 0.14                                                   | 0.06                                                    |
| 生境 3 | 2.94 | 8.10 | 11.1 | 0.97 | 8.75                      | 20.25                      | 0.11                      | 2.23                      | 0.31                                                   | 0.071                                                  | 0.04                                                    |

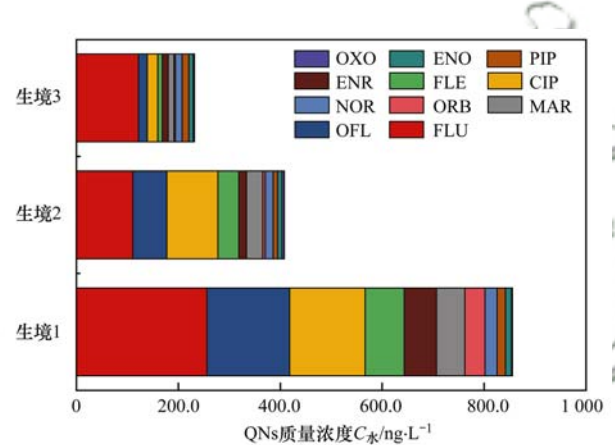


图 3 白洋淀水体 QNs 质量浓度

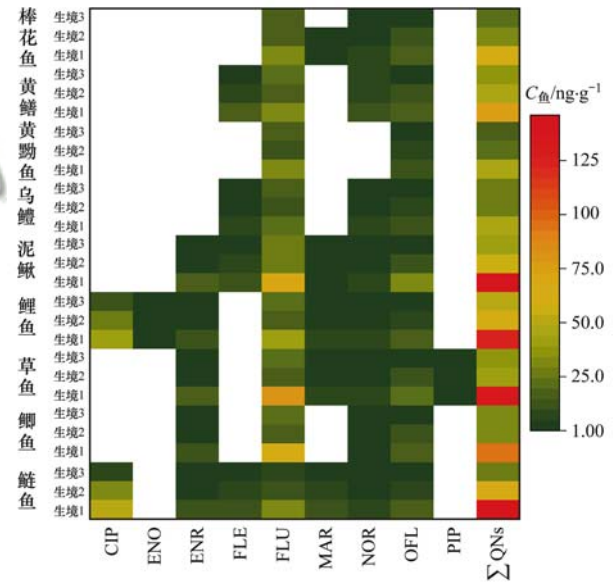
Fig. 3 Mass concentration of QNs in Baiyangdian Lake

2.2 白洋淀鱼体内的 QNs 含量

白洋淀鱼类样品中共检出 9 种 QNs,其中 FLU 和 OFL 检出率最高(100%),其次为 NOR、ENR 和 MAR(≥ 50.0%), QNs 平均含量(ng·g<sup>-1</sup>)范围为 1.85(ENO) ~ 28.1(CIP)(图 4). 鱼类体内 ∑ QNs 含量范围为 17.1 ~ 146 ng·g<sup>-1</sup>,其中泥鳅体内 ∑ QNs 平均含量最高(80.4 ng·g<sup>-1</sup>),黄魮鱼体内 ∑ QNs 平均含量最低(28.5 ng·g<sup>-1</sup>). CIP(28.1 ng·g<sup>-1</sup>)和 FLU(27.3 ng·g<sup>-1</sup>)在鱼类体内的平均含量高于其它 QNs(< 15.0 ng·g<sup>-1</sup>). 就空间差异而言,除 ENO、FLU 和 PIP 外,其余 QNs 均呈现生境 1 > 生境 2 > 生境 3,与水体中 QNs 质量浓度空间分布一致.

不同 QNs 在不同鱼类中的含量存在差异(图 5),鲢鱼、鲤鱼、草鱼和泥鳅中 QNs 检出种类较多(> 5 种). 就 QNs 种类而言,鱼类体内 FLU 平均相

对含量最高(52%),其次为 OFL(18%),与水体中各 QNs 相对质量浓度基本一致,表明鱼体内与其生活的水环境中 QNs 相对含量特征具有一定相似性.



空白部分表示未检出

图 4 鱼类体内 QNs 含量特征

Fig. 4 Characteristics of QNs concentrations in fish species

2.3 白洋淀 QNs 在优势鱼类中的 BAF 和 TMF

白洋淀优势鱼类中 QNs 的 BAF 结果见图 6 和表 2. 就优势鱼类的 BAF 差异而言(图 6),鲤鱼的 BAF 平均值最高(163 L·kg<sup>-1</sup>),黄魮鱼的 BAF 平均值最低(58.3 L·kg<sup>-1</sup>); 鲤鱼、乌鳢、黄鳊和棒花鱼的 BAF 呈现生境 3 > 生境 2 > 生境 1,鲫鱼、草鱼、泥鳅和黄魮鱼的 BAF 则在生境 2 值最小. 就 QNs 种类差异而言(表 2),BAF<sub>CIP</sub> 平均值最高(361 L·kg<sup>-1</sup>),BAF<sub>MAR</sub> 的平均值最低(122 L·kg<sup>-1</sup>); BAF



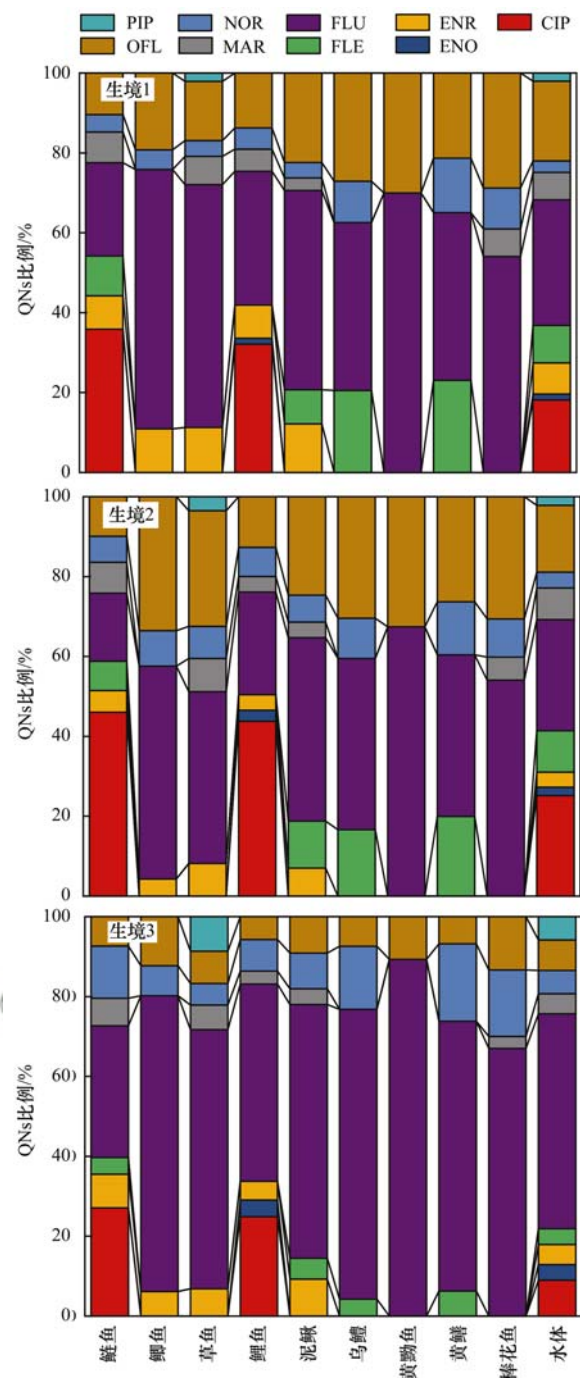


图5 白洋淀3个生境鱼类及水体中QNs丰度特征  
Fig. 5 Characteristics of QNs in fish species and water of three habitats in Baiyangdian Lake

在生境1、生境2和生境3的范围( $L \cdot kg^{-1}$ )分别为111( $BAF_{OFL}$ )~309( $BAF_{CIP}$ )、96.2( $BAF_{MAR}$ )~286( $BAF_{CIP}$ )和140( $BAF_{MAR}$ )~489( $BAF_{CIP}$ )。

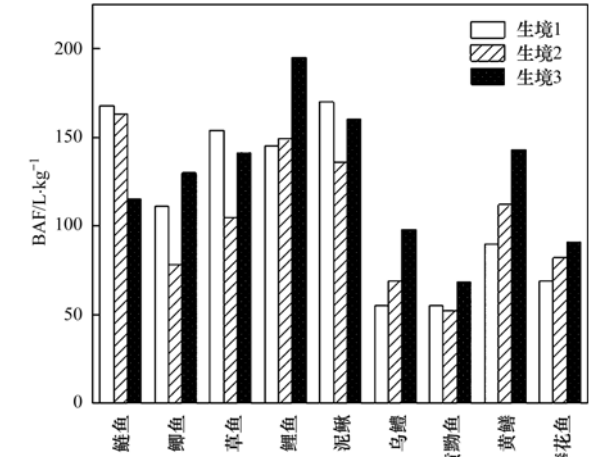


图6 鱼类QNs的BAF  
Fig. 6 BAF of QNs in fish

不同生境的鱼类 $\delta^{15}N$ 稳定同位素数据存在差异性(表3),生境1[11.36‰(鲤鱼)~15.34‰(乌鳢)]和生境2[10.62‰(鲤鱼)~14.80‰(乌鳢)]的 $\delta^{15}N$ 高于生境3[9.722‰(鲤鱼)~14.35‰(乌鳢)]。就鱼类而言,鲤鱼的 $\delta^{15}N$ 值最低,乌鳢的最高。3个生境中各鱼类营养级(TL)范围为2.33~3.88,TL值空间分布为生境3(2.33~3.88)>生境2(2.40~3.80)>生境1(2.33~3.67)。

本研究中,鱼类体内QNs检出率存在较大差异,因此,选择检出率>50%的QNs(ENR、FLU、MAR、NOR、OFL)和 $\sum$ QNs计算TMF值(图7),3个生境中各QNs的TMF范围为0.714( $TMF_{MAR,生境3}$ )~1.33( $TMF_{ENR,生境3}$ ),MAR和OFL的TMF值在生境2最大,ENR、FLU和NOR的TMF值在生境3最大。就QNs而言,TMF呈现出 $TMF_{ENR} > TMF_{NOR} > TMF_{OFL} > TMF_{FLU} > TMF_{MAR}$ 。

2.4 白洋淀QNs的BAF和TMF与环境因子的相关性

Spearman秩相关分析结果表明(表4),理化因

表2 白洋淀3个生境中鱼类体内QNs的BAF/ $L \cdot kg^{-1}$

Table 2 Observed BAF of QNs in fish from three habitats in Baiyangdian Lake/ $L \cdot kg^{-1}$

| 项目  | $BAF_{CIP}$ | $BAF_{ENO}$ | $BAF_{ENR}$ | $BAF_{FLE}$ | $BAF_{FLU}$ | $BAF_{MAR}$ | $BAF_{NOR}$ | $BAF_{OFL}$ | $BAF_{PIP}$ |
|-----|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|
| 生境1 | 309         | 151         | 207         | 177         | 176         | 129         | 266         | 111         | 167         |
| 生境2 | 286         | 202         | 198         | 153         | 151         | 96.2        | 243         | 148         | 175         |
| 生境3 | 489         | 237         | 225         | 182         | 163         | 140         | 285         | 158         | 230         |

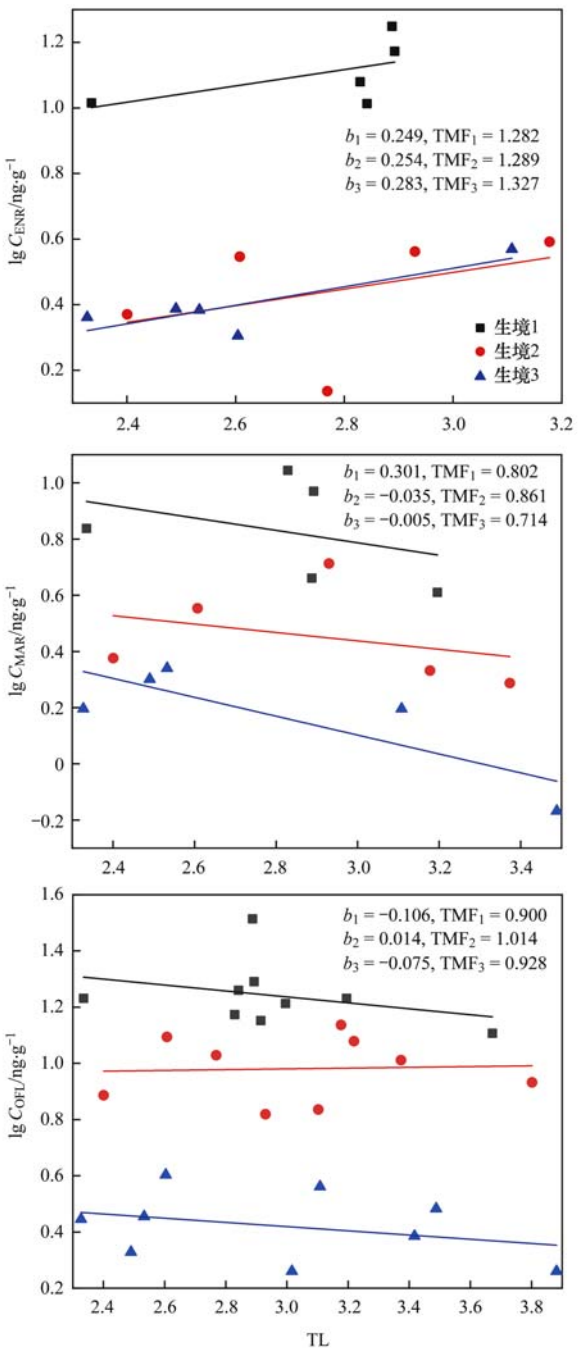
子与BAF/TMF存在相关性。除WD外,其余水体理化因子均与BAF和TMF显著相关( $P < 0.05$ 或 $P < 0.01$ ),其中pH与BAF呈负相关,与TMF呈正相关;

$BAF_{ENO}$ 、 $BAF_{OFL}$ 、 $BAF_{PIP}$ 、 $TMF_{ENR}$ 、 $TMF_{FLU}$ 、 $TMF_{NOR}$ 和 $TMF_{QNs}$ 与T、SD和DO显著正相关,与COD、TP、TN、 $NH_4^+-N$ 、 $NO_3^--N$ 和 $PO_4^{3-}-P$ 显著负相关。

表3 白洋淀鱼类  $\delta^{15}\text{N}$  稳定同位素和营养级 (TL) 空间差异

Table 3 Spatial variation in stable isotope diagram ( $\delta^{15}\text{N}$ ) and trophic level (TL) of fish in Baiyangdian Lake

| 鱼类  | 生境 1                           |      | 生境 2                           |      | 生境 3                           |      |
|-----|--------------------------------|------|--------------------------------|------|--------------------------------|------|
|     | $\delta^{15}\text{N}/\text{‰}$ | TL   | $\delta^{15}\text{N}/\text{‰}$ | TL   | $\delta^{15}\text{N}/\text{‰}$ | TL   |
| 鲢鱼  | 12.83                          | 2.83 | 12.20                          | 2.93 | 10.20                          | 2.49 |
| 鲫鱼  | 12.87                          | 2.84 | 11.72                          | 2.77 | 10.54                          | 2.60 |
| 草鱼  | 13.02                          | 2.89 | 11.24                          | 2.61 | 10.33                          | 2.53 |
| 鲤鱼  | 11.36                          | 2.33 | 10.62                          | 2.40 | 9.722                          | 2.33 |
| 乌鲤  | 15.34                          | 3.67 | 14.80                          | 3.80 | 14.35                          | 3.88 |
| 黄鲂鱼 | 13.09                          | 2.91 | 12.72                          | 3.10 | 11.77                          | 3.02 |
| 棒花鱼 | 13.92                          | 3.20 | 13.52                          | 3.37 | 13.18                          | 3.49 |
| 泥鳅  | 13.01                          | 2.89 | 12.94                          | 3.18 | 12.04                          | 3.11 |
| 黄鳝  | 13.33                          | 3.00 | 13.06                          | 3.22 | 12.96                          | 3.42 |



$b_1$ 、 $b_2$  和  $b_3$  分别为生境 1、生境 2 和生境 3 中对应的公式(2)中的  $b$  值；

$\text{TMF}_1$ 、 $\text{TMF}_2$  和  $\text{TMF}_3$  分别为生境 1、生境 2 和生境 3 中鱼类的营养放大因子 (TMF)

图7 3 个生境中鱼类体内 QNs 含量对数与 TL 的关系

Fig. 7 Relationships between log-transformed concentrations of QNs and trophic levels (TL) in fish from three habitats

2.5 白洋淀食用鱼的健康风险

各 QNs 的健康风险如图 8 所示. 就各 QNs 而言,  $\text{HQ}_{\text{CIP}}$  (0.0040 ~ 0.026) 显著高于其他 QNs 的  $\text{HQ}$  (<0.005 0). 就空间差异而言, 生境 1 的  $\text{HQ}$  值最高, 生境 3 最低, 与水体 QNs 质量浓度和鱼类中 QNs 含量趋势一致.  $\text{HI}$  范围为 0.001 ~ 0.035,  $\text{HI}_{\text{鲢鱼, 生境1}}$  (0.035)、 $\text{HI}_{\text{鲢鱼, 生境2}}$  (0.019)、 $\text{HI}_{\text{草鱼, 生境1}}$  (0.012)、 $\text{HI}_{\text{鲤鱼, 生境1}}$  (0.029)、 $\text{HI}_{\text{鲤鱼, 生境2}}$  (0.016) 和  $\text{HI}_{\text{泥鳅, 生境1}}$  (0.014) 均大于 0.01, 处于高健康风险水平, 表明食用白洋淀鱼类存在较大风险. 就各鱼

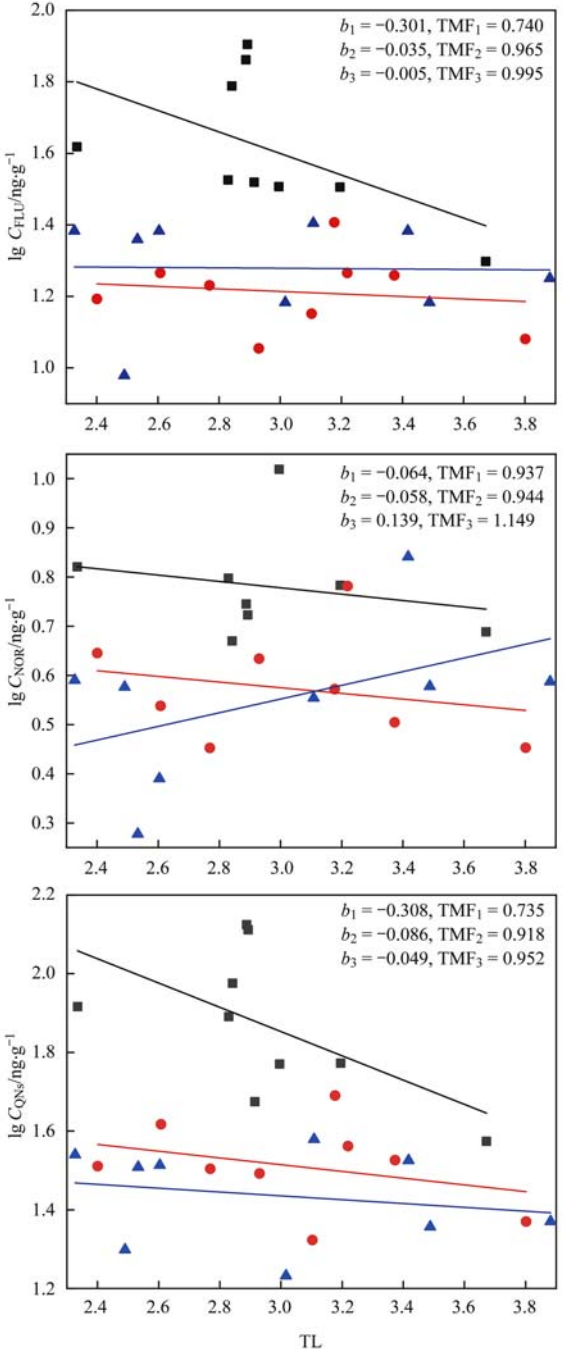


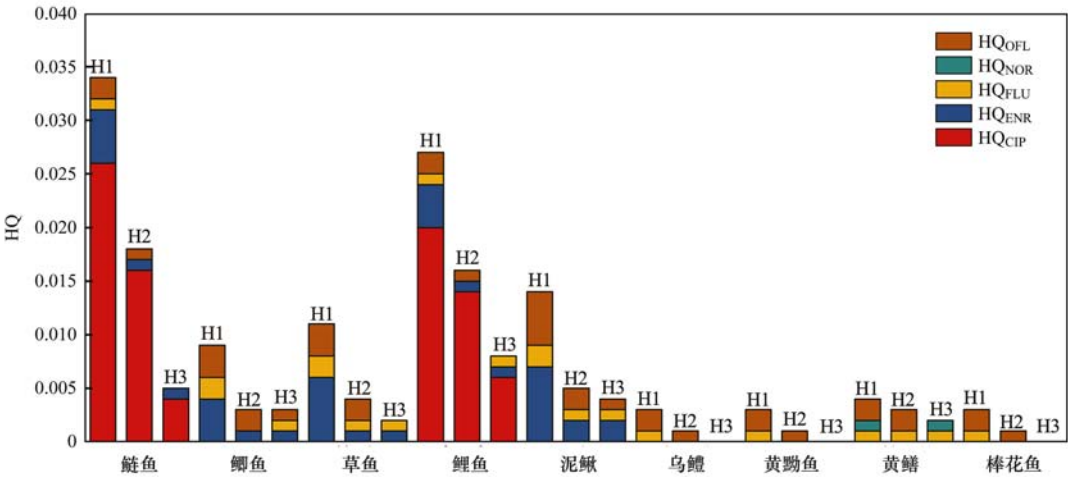


表 4 环境因子与 QNs 的 BAF/TMF 的 Spearman 相关性

Table 4 Spearman rank correlation between environmental factors and the BAF/TMF of QNs

|                    | WD     | pH        | T       | SD       | DO       | COD       | TP        | TN        | NH <sub>4</sub> <sup>+</sup> -N | NO <sub>3</sub> <sup>-</sup> -N | PO <sub>4</sub> <sup>3-</sup> -P |
|--------------------|--------|-----------|---------|----------|----------|-----------|-----------|-----------|---------------------------------|---------------------------------|----------------------------------|
| BAF <sub>CIP</sub> | 0.103  | -0.772 *  | 0.051   | 0.309    | 0.309    | -0.309    | -0.180    | -0.309    | -0.463                          | -0.309                          | -0.323                           |
| BAF <sub>ENO</sub> | 0.309  | 0.154     | 0.772 * | 0.926 ** | 0.926 ** | -0.926 ** | -0.849 ** | -0.926 ** | -0.772 *                        | -0.926 **                       | -0.815 *                         |
| BAF <sub>ENR</sub> | 0.103  | -0.772 *  | 0.051   | 0.309    | 0.309    | -0.309    | -0.18     | -0.309    | -0.463                          | -0.309                          | -0.323                           |
| BAF <sub>FLE</sub> | 0.103  | -0.772 *  | 0.051   | 0.309    | 0.309    | -0.309    | -0.18     | -0.309    | -0.463                          | -0.309                          | -0.323                           |
| BAF <sub>FLU</sub> | -0.103 | -0.874 ** | -0.463  | -0.309   | -0.309   | 0.309     | 0.386     | 0.309     | 0.051                           | 0.309                           | 0.220                            |
| BAF <sub>MAR</sub> | 0.103  | -0.772 *  | 0.051   | 0.309    | 0.309    | -0.309    | -0.180    | -0.309    | -0.463                          | -0.309                          | -0.323                           |
| BAF <sub>NOR</sub> | 0.103  | -0.772 *  | 0.051   | 0.309    | 0.309    | -0.309    | -0.18     | -0.309    | -0.463                          | -0.309                          | -0.323                           |
| BAF <sub>OFL</sub> | 0.309  | 0.154     | 0.772 * | 0.926 ** | 0.926 ** | -0.926 ** | -0.849 ** | -0.926 ** | -0.772 *                        | -0.926 **                       | -0.815 *                         |
| BAF <sub>PIP</sub> | 0.309  | 0.154     | 0.772 * | 0.926 ** | 0.926 ** | -0.926 ** | -0.849 ** | -0.926 ** | -0.772 *                        | -0.926 **                       | -0.815 *                         |
| BAF <sub>QNs</sub> | 0.103  | -0.772 *  | 0.051   | 0.309    | 0.309    | -0.309    | -0.18     | -0.309    | -0.463                          | -0.309                          | -0.323                           |
| TMF <sub>ENR</sub> | 0.309  | 0.154     | 0.772 * | 0.926 ** | 0.926 ** | -0.926 ** | -0.849 ** | -0.926 ** | -0.772 *                        | -0.926 **                       | -0.815 *                         |
| TMF <sub>FLU</sub> | 0.309  | 0.154     | 0.772 * | 0.926 ** | 0.926 ** | -0.926 ** | -0.849 ** | -0.926 ** | -0.772 *                        | -0.926 **                       | -0.815 *                         |
| TMF <sub>MAR</sub> | -0.103 | 0.772 *   | -0.051  | -0.309   | -0.309   | 0.309     | 0.180     | 0.309     | 0.463                           | 0.309                           | 0.323                            |
| TMF <sub>NOR</sub> | 0.309  | 0.154     | 0.772 * | 0.926 ** | 0.926 ** | -0.926 ** | -0.849 ** | -0.926 ** | -0.772 *                        | -0.926 **                       | -0.815 *                         |
| TMF <sub>OFL</sub> | 0.103  | 0.874 **  | 0.463   | 0.309    | 0.309    | -0.309    | -0.386    | -0.309    | -0.051                          | -0.309                          | -0.220                           |
| TMF <sub>QNs</sub> | 0.309  | 0.154     | 0.772 * | 0.926 ** | 0.926 ** | -0.926 ** | -0.849 ** | -0.926 ** | -0.772 *                        | -0.926 **                       | -0.815 *                         |

1) \* 表示  $P < 0.05$  水平显著相关; \*\* 表示  $P < 0.01$  水平显著相关



H1、H2 和 H3 分别代表生境 1、生境 2 和生境 3

图 8 白洋淀鱼类体内 QNs 的 HQ

Fig. 8 HQ for QNs in Baiyangdian Lake fish

类而言,鲢鱼、鲤鱼的食用风险显著高于其他鱼类.

3 讨论

3.1 白洋淀水体和鱼类体内 QNs 含量特征

QNs 在我国地表水环境中被广泛检出,其在水体中的质量浓度量级从 $\text{ng}\cdot\text{L}^{-1}$ 到 $\mu\text{g}\cdot\text{L}^{-1}$ . 本研究中白洋淀水体中  $\sum$  QNs 的质量浓度范围为 $0.7400 \sim 1590 \text{ ng}\cdot\text{L}^{-1}$ ,高于巢湖<sup>[40]</sup>、博斯腾湖<sup>[41]</sup>、洪湖<sup>[42]</sup>、鄱阳湖<sup>[43]</sup>和长江南段<sup>[44]</sup>等水域,低于太湖贡湖湾<sup>[45]</sup>等,这与水体周围环境密切相关. 而与之之前的白洋淀研究相比,本研究结果水体中的  $\sum$  QNs 质量浓度低于 Cheng 等<sup>[28]</sup>的研究结果,这可能是因为近些年国家政府加大了对畜禽业和渔业中的抗生素使用的管控<sup>[46]</sup>,降低了 QNs 的排放.

白洋淀鱼体内 QNs 水平( $91.8 \text{ ng}\cdot\text{g}^{-1}$ )高于莱州湾( $56.0 \text{ ng}\cdot\text{g}^{-1}$ )<sup>[9]</sup>,鱼体 CIP 含量( $\text{ND} \sim 51.5 \text{ ng}\cdot\text{g}^{-1}$ )高于珠江三角洲( $4.61 \sim 33.3 \text{ ng}\cdot\text{g}^{-1}$ )<sup>[26]</sup>和海河( $1.67 \sim 12.5 \text{ ng}\cdot\text{g}^{-1}$ )<sup>[6]</sup>,而鱼体 NOR 含量( $\text{ND} \sim 10.5 \text{ ng}\cdot\text{g}^{-1}$ )则低于珠江三角洲( $3.38 \sim 101 \text{ ng}\cdot\text{g}^{-1}$ )<sup>[26]</sup>和海河( $0.500 \sim 51.6 \text{ ng}\cdot\text{g}^{-1}$ )<sup>[6]</sup>,各地区抗生素使用水平不同,导致水体抗生素污染也存在差异.

3.2 白洋淀 QNs 在鱼体内的生物累积及其与环境因子相关性

白洋淀 3 个生境中鱼体内 9 种 QNs 的 BAF 范围为:  $96.2 \text{ L}\cdot\text{kg}^{-1}$  ( $\text{BAF}_{\text{MAR},\text{生境}2}$ )  $\sim 489 \text{ L}\cdot\text{kg}^{-1}$  ( $\text{BAF}_{\text{CIP},\text{生境}3}$ ),小于 $1000 \text{ L}\cdot\text{kg}^{-1}$ ,表明 QNs 在鱼类体内的生物富集相对较低<sup>[32]</sup>.  $\text{BAF}_{\text{OFL}}$ 、 $\text{BAF}_{\text{NOR}}$ 、

BAF<sub>CIP</sub>和BAF<sub>ENR</sub>的平均值分别为139、265、361和210 L·kg<sup>-1</sup>,低于太湖和之前的白洋淀研究结果[88.0 ~ 214 L·kg<sup>-1</sup> (BAF<sub>OFL</sub>)、251 ~ 512 L·kg<sup>-1</sup> (BAF<sub>NOR</sub>)、545 ~ 2 008 L·kg<sup>-1</sup> (BAF<sub>CIP</sub>)和4 490 L·kg<sup>-1</sup> (BAF<sub>ENR</sub>)]<sup>[23,24]</sup>. 环境因子对抗生素的归趋和残留起着重要的作用<sup>[47,48]</sup>,这种现象可能是环境因子在不同地区的时空差异所导致<sup>[24,49]</sup>.

本研究中的TMF<sub>OFL</sub>(0.900 ~ 1.01)和TMF<sub>ENR</sub>(1.28 ~ 1.33)高于之前的白洋淀研究中的值(0.380 ~ 0.520)<sup>[23]</sup>,TMF<sub>NOR</sub>(0.937 ~ 1.15)低于太湖<sup>[24]</sup>和莱州湾<sup>[9]</sup>中的值(1.24 ~ 1.25). 由文献[50]可知,白洋淀中ENR、生境3的NOR和生境2的OFL呈营养放大,其他QN<sub>s</sub>(FLU、MAR、生境1和生境2的NOR、生境1和生境3的OFL、 $\sum$ QN<sub>s</sub>)呈营养稀释,与之前的研究结果基本一致<sup>[24,51]</sup>. 而目前抗生素的生物稀释机制尚不明晰,但可推测其与环境因子密切相关.

白洋淀QN<sub>s</sub>的BAF/TMF与环境因子的相关性结果表明,pH、*T*、SD、DO、COD、TP、TN、NH<sub>4</sub><sup>+</sup>-N、NO<sub>3</sub><sup>-</sup>-N和PO<sub>4</sub><sup>3-</sup>-P与QN<sub>s</sub>在鱼类体内富集和传递显著相关. 有研究发现QN<sub>s</sub>具有较强的吸附能力(吸附系数高达260 ~ 5 012 L·kg<sup>-1</sup>)<sup>[52]</sup>,可与水环境中的有机质发生吸附作用,沉降于沉积物中,进而导致水环境中QN<sub>s</sub>浓度降低,最终鱼类对QN<sub>s</sub>的生物累积降低. 有研究表明<sup>[49]</sup>,黄海南部和长江口中pH对抗生素的富集存在影响. 另外当环境条件(pH、离子条件等)发生变化时,沉积物中的抗生素解吸附进入水环境中,最终被生物富集. 因此,理化参数是影响抗生素生物富集和传递的重要因素,且pH对其影响极大<sup>[53]</sup>. 有研究表明,pH可影响抗生素的解离,而非解离形态的抗生素易跨过细胞膜被生物吸附<sup>[54]</sup>. Bergsjø等<sup>[55]</sup>的研究发现pH可影响虹鳟鱼对磺胺二甲嘧啶的生物富集,在pH为7 ~ 8时,生物富集能力最强. 温度可以通过影响抗生素在水中和生物膜之间扩散过程、抗生素和蛋白质的结合速率和类脂物成分发生改变等,进而影响抗生素的生物吸附和释放. 另有研究表明,水体中其它污染物、盐度和离子强度等均可影响抗生素的形态和理化特性<sup>[56,57]</sup>,从而影响抗生素的生物富集和营养传递.

#### 4 结论

(1)白洋淀水体中QN<sub>s</sub>存在显著空间差异,鱼类体内QN<sub>s</sub>也存在空间和物种差异性.

(2)鱼类对QN<sub>s</sub>的生物富集能力在生境3最

高,其次为生境2和生境1;ENR呈营养放大,FLU、MAR和 $\sum$ QN<sub>s</sub>呈营养稀释.

(3)pH、*T*、SD、DO、COD、TP、TN、NH<sub>4</sub><sup>+</sup>-N、NO<sub>3</sub><sup>-</sup>-N和PO<sub>4</sub><sup>3-</sup>-P与QN<sub>s</sub>在鱼类体内的生物富集和传递显著相关.

(4)白洋淀鱼类整体存在较大的食用健康风险,因此,应制定更为严格的抗生素排放标准,减少湖泊外源有机质和营养物质输入,以降低抗生素污染水平和健康风险.

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