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大冶湖表层沉积物-水中多环芳烃的分布、来源及风 险评价

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摘要:于 2015年8月采集大冶湖表层沉积物8个及上覆水样8个,使用GC-MS分析16种EPA优控PAHs.结果表明在表层沉积物及水体中 \sum PAHs范围分别为:35.94~2032.73 ng·g⁻¹和27.94~242.95 ng·L⁻¹,平均值分别为940.61 ng·g⁻¹和107.77 ng·L⁻¹;表层沉积物中PAHs分布呈现湖中高于岸边趋势,水体则呈大致相反趋势,表层沉积物中以4~5环高环化合物为主要组分,在水体中主要以2环以及4环和5环PAHs为主,与国内外其他湖泊相比处于中度污染水平;来源解析表明大冶湖表层沉积物及水体中多环芳烃主要来自于高温燃烧源,沉积物中PAHs高环分子都占据绝大部分,反映出了沉积物受矿冶冶炼长期累积污染的效应;所检测沉积物中各单体PAH及 \sum PAHs含量均未超过ERM以及FEL,表明大冶湖表层沉积物中PAHs无潜在生态风险;终生致癌风险评价表明大冶湖水体中PAHs通过摄入和皮肤接触风险都处于USEPA推荐的可接受水平范围之内,但都高于瑞典环保局和英国皇家协会推荐的最大可接受风险水平,需要对7种致癌PAHs污染加以防治. 关键词:大冶湖;表层沉积物;水;多环芳烃;来源解析;健康风险

大庭园:大陆湖, 农丛600万岁, 水, 少百万元, 木杨州州, 庭水八雪

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Distribution, Sources and Risk Assessment of the PAHs in the Surface Sediments and Water from the Daye Lake

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Abstract: 8 surface sediments and 8 water samples were collected from the Daye Lake in August 2015. The 16 kinds of EPA control polycyclic aromatic hydrocarbons (PAHs) were analyzed by GC-MS. The results showed that the PAHs concentrations of surface sediments and water ranged from 35. 94 ng·g⁻¹ to 2 032. 73 ng·g⁻¹ and from 27. 94 ng·L⁻¹ to 242. 95 ng·L⁻¹, with average contents of 940. 61 ng·g⁻¹ and 107. 77 ng·L⁻¹, respectively. The distribution of PAHs in surface sediments indicated that the contents in the center samples were higher than those in the bank samples, but the water showed nearly the opposite tendency. The 4-5 rings high molecular weight PAHs were the main components in the surface sediments, and the 2, 4 and 5 rings PAHs were given priority in water. Compared with the other domestic and oversea lakes, the PAHs pollution of the Daye Lake was at a moderate level. Source apportionment showed that the PAHs in surface sediments and water from the Daye Lake came from the combustion source, HWM-PAHs were the dominant part of the PAHs in the sediment, reflecting the sediment PAHs pollution under the effects of mining and smelting over a long period; All monomer PAHs and total PAHs content in sediment did not exceed the ERM and FEL limiting values, showing that there was no particularly serious ecological risk caused by PAHs in the surface sediments from the Daye Lake; the incremental lifetime cancer risks assessment showed that the uptake risk of PAHs in Daye Lake water through the ingestion and dermal absorption were both in the acceptable range recommended by the USEPA, but all sites had higher risk than the acceptable risk level recommended by the Sweden environmental protection agency and Royal society. The pollution of seven carcinogenic PAHs needs prevention and control.

Key words: Daye Lake; surface sediments; water; PAHs; source apportionment; health risk

多环芳烃(PAHs)是中性、非极性广泛存在于环境中的一类持久性有机污染物(POPs),含有两个或两个以上芳香环,因其高熔点、高持久性及"三致"(致癌、致畸、致突变)特性^[1,2],多环芳烃通常会对水环境中生物体产生遗传毒性,并通过食物链累积对人体健康产生危害,PAHs 成为全球性研究热点问题,近年来关于 PAHs 破坏人体内分泌活动

报道十分常见^[3,4]. 环境中的多环芳烃污染物主要是人为来源,如石油的直接泄漏以及石油产品精炼、

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加工或者石油、木材及其他有机质的燃烧,少量来自于火山爆发,森林火灾以及岩石分化等自然源^[5-8].因此,了解多环芳烃地球化学行为包括他们的分布、来源对于环境污染防治工作开展具有非常重要的意义.

黄石市位于湖北省东南部,是我国矿冶历史名 城,大冶湖是黄石市面积最大的天然湖泊,为长江中 下游代表性中型浅水湖泊,湖体狭长,呈东西走向, 后流入长江. 大冶湖主要接纳地表径流以及周边排 污,湖泊面积约为63.4 km²,平均水位约16 m,最大 蓄水量可达2×108 m3. 大冶湖底和四周储藏有大 量的金属矿藏,至今流域内已被探明的矿产资源 多达43种,其中以铜、铁、金、煤、钼、硫等矿产 资源储藏量最为丰富,因而采矿、选矿、黑色和有 色金属冶炼业十分发达,是我国九大有色冶炼业 基地之一,这些矿产及工业大多分布于湖区上游 及西岸,重金属冶炼过程中产生的大量有机污染 物(如 PAHs),曾长期未经处理直接进入湖泊水体 及沉积物,存在较严重生态环境风险.而大冶湖流 域随着经济开发圈的建成,逐步由郊区扩展为城 区. 因此,查明大冶湖水体及沉积物中 PAHs 污染 水平、组成及分布,开展大冶湖 PAHs 来源与风险 评价,对于环大冶湖生态新区建设具有科学的指 导意义,可为大冶湖等浅水湖泊的有机污染物防 治提供科学依据.

1 材料与方法

1.1 样品采集

于2015年8月根据大冶湖的走向,尽量靠湖中线,按照等距离分布依次用抓斗式采样器共采集了8个表层沉积物样品,在采集沉积物样品之前先于水面20cm以下采集配套表层水体样品. 沉积物样品采集后置于聚乙烯密实袋中,水样采集后保存于事先用丙酮清洗、现场用待采集处的水体再润洗3次的棕色玻璃瓶中,采样现场使用冰块保存所采集的表层沉积物及水体样品,运回实验室后,在-20℃条件下保存样品待分析. 采样点分布如图1.

1.2 样品预处理

沉积物样品处理:表层沉积物经自然阴干,并去除动植物残体及杂质后,称取 10 g 样品及加入 5.0 μ L 回收率指示物氘代萘(Naphthalene- d_8)、氘代二氢苊(Acenaphthene- d_{10})、氘代菲(Phenanthrene- d_{10})、氘代**蕌**(Chrysene- d_{12}) 和氘代苝(Perylene- d_{12}),用 CH_2Cl_2 120 ~ 140 mL 水浴温度 45℃下,在

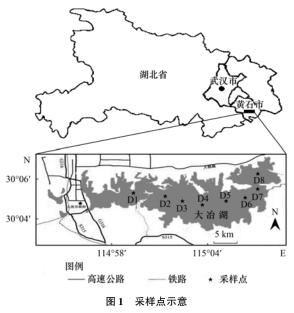


Fig. 1 Sampling sites

索氏提取器内抽提 24 h,并用铜片脱硫.

水样处理:取1 L 水样置于分液漏斗中,加入 $5.0~\mu$ L 回收率指示物,然后立即加入 25~mL CH_2Cl_2 进行萃取,重复萃取 $3~\chi$.

沉积物及水样处理后的抽提液经旋蒸仪(德国 Heidolph RE-52)40℃条件下浓缩至约5 mL 后,加入5~10 mL 正己烷,继续浓缩至约5 mL. 浓缩液通过装有去活化的硅胶和氧化铝(体积比2:1)的层析柱净化分离. 用二氯甲烷和正己烷(体积比2:3)混合液淋洗. 淋洗液浓缩至0.5 mL,转移至2 mL 细胞瓶,用柔和的氮气(纯度>99.99%)吹至0.2 mL,加入内标标准物,低温保存至上机分析. 其中,硅胶置于烘箱180℃烘12 h,氧化铝置于烘箱270℃烘12 h,作用是使其达到活化效果. 冷却后,分别加3%质量比的去离子水去活化.

1.3 仪器分析和试剂标准

采用 Agilent 公司气相色谱-质谱联用仪(GC-MS 7890A-5975C MSD)分析 16 种美国 EPA 优控 PAHs 污染物,色谱柱为 DB-5MS 熔融石英毛细柱 (30 m×0.25 mm×0.25 μm). 色谱柱程序升温:初始温度 85℃,持续 2 min; 以 4℃·min⁻¹ 升温至 290℃后,持续 25 min,直至所有组分从色谱柱中流出. 进样口温度为 280℃,载气为氦气(纯度 > 99.99%),流速为 1 mL·min⁻¹,不分流进样,进样量为 1.0 μL.

美国环保署(USEPA)列出的 16 种优控 PAHs 污染物: 2~3 环包括萘(Nap)、苊烯(Acy)、苊 (Ace)、芴(Flu)、菲(Phe)、蒽(Ant)这 6 种,4 环 包括芘(Pyr)、荧蒽(Fla)、苯并[a]蒽(BaA)、**菌** (Chr)这 4 种,5 环包括苯并[b] 荧蒽(BbF)、苯并 [k] 荧蒽(BkF)、苯并[a] 芘(BaP)、二苯并[a,h] 蒽(DBA)这 4 种,6 环包括茚并[1,2,3-cd] 芘 (IcdP)、苯并[ghi] 苝(BghiP)这 2 种. PAHs 混合标准液(美国 Supeco):含萘- d_8 、苊- d_{10} 、菲- d_{10} 、**茄**- d_{12} 和苝- d_{12} ,所使用有机试剂正己烷(色谱纯)购自美国 Tedia 公司,二氯甲烷(色谱纯)购自美国 J. T. Baker 公司,内标标准物:六甲基苯.

1.4 质量控制/质量保证(QA/QC)

沉积物和水样处理过程中都使用平行样、方法空白和程序空白进行质量控制和保证. 方法空白中无目标化合物检出,平行样分析中 PAHs 相对偏差均小于 10%,在误差允许范围内. Nap-D₈、Acy-D₁₀、Phe-D₁₀、Chr-D₁₂和 Pyr-D₁₂的平均回收率分别是 $58\% \pm 6\%$ 、 $82\% \pm 9\%$ 、 $108\% \pm 10\%$ 、 $112\% \pm 13\%$ 及 $118\% \pm 11\%$. 最终结果经回收率校正.

2 结果与讨论

2.1 表层沉积物及水体中 PAHs 污染水平

表层沉积物及其配套水体中 PAHs 含量见表 1 所示. 在表层沉积物中 \sum PAHs 含量范围为 35. 94 ~2 032. 73 ng·g⁻¹,均值为 940. 61 ng·g⁻¹; 7 种致

癌单体多环芳烃 ∑, PAHs 含量范围为 18.16 ~ 1 289.92 ng·g⁻¹,均值为 576.32 ng·g⁻¹. 而在水体 中 > PAHs 含量范围为,27.94~242.95 ng·L⁻¹,均 值为 107.77 ng·L-1; 7 种致癌单体多环芳烃 ∑₂PAHs 含量范围为 14.82 ~ 94.79 ng·L⁻¹,均值 为 62.60 ng·L-1. 我国尚无评判 PAHs 污染水平的 标准,根据 Long 等[9]的生态风险评价原则,没有最 低安全阈值的高环致癌性多环芳烃 BkF 和 BbF 只 要存在于环境中,就会对生物体健康产生威胁,在整 个大冶湖采样区这两种 PAH 单体检出率在沉积物 和水体中都为 100%, 且与美国 EPA 标准比较大冶 湖水体中7种致癌 PAHs 中 BaP、BkF、BbF 这3种 超标率均为100%,而我国地表水环境质量标准 (GB 3838-2002) 中规定 BaP 的限值为 2.8 ng·L⁻¹ 在大冶湖水体中 BaP 超标率达 100%, 表明研究区 PAHs 污染存在一定的风险.

大冶湖表层沉积物中PAHs处于中等污染水平与国内其他典型的河流、湖泊对比(表2),大冶湖表层沉积物中PAHs含量远高于柘林湾养殖区、浙江千岛湖和太湖、略高于巢湖及太湖沉积物中PAHs含量,远低于辽河表层沉积物中PAHs含量,略低于珠江表层沉积物中的PAHs,与安庆长江沿岸

表 1 大冶湖表层沉积物及表层水体中 PAHs 含量¹⁾

| /1 · A #ba | TT W. | mp.c | 沉积物/ng | g•g −1 | | 水样/ng·L-1 | |
|-----------------------|-------|-------|------------------|--------|-------------------|-----------|--------|
| 化合物 | 环数 | TEF | 范围 | 均值 | 范围 | 均值 | EPA 标准 |
| Nap | 2 环 | 0.001 | 0.21 ~29.12 | 14.61 | 9.05 ~ 50.83 | 22.68 | _ |
| Acy | 3 环 | 0.001 | 0.14 ~ 1.84 | 0.95 | N. D. ~1.97 | 0.47 | _ |
| Ace | 3 环 | 0.001 | 0.21 ~4.88 | 2.49 | N. D. ~5.03 | 1.63 | _ |
| Flu | 3 环 | 0.001 | 2.04 ~49.62 | 22.51 | N. D. ~ 2.01 | 0.73 | _ |
| Phe | 3 环 | 0.001 | 5.34 ~ 188.00 | 91.95 | N. D. ~28.68 | 7.34 | _ |
| Ant | 3 环 | 0.01 | 0.50 ~ 14.64 | 6.94 | 0.13~31.96 | 6.57 | _ |
| Fla | 4 环 | 0.001 | 4.98 ~ 238.69 | 113.54 | 0.23 ~ 7.84 | 2.15 | _ |
| Pyr | 4 环 | 0.001 | 3.48 ~ 178.87 | 86.04 | 0.12~6.56 | 1.52 | _ |
| BaA | 4 环 | 0.1 | 1.30 ~80.52 | 40.83 | N. D. ~5.53 | 1.47 | 4.4 |
| Chr | 4 环 | 0.01 | 1.46 ~ 156.06 | 62.12 | N. D. ~6.85 | 1.12 | 4.4 |
| BbF | 5 环 | 0.1 | 6.16 ~ 393.52 | 173.25 | 5.36 ~37.35 | 19.87 | 4.4 |
| BkF | 5 环 | 0.1 | 1.62 ~ 104.76 | 44.89 | 2.67 ~32.94 | 16.16 | 4.4 |
| BaP | 5 环 | 1 | 1.06 ~ 103.29 | 47.21 | 6.79 ~49.75 | 18.69 | 4.4 |
| DBA | 5 环 | 1 | 3.25 ~247.59 | 106.3 | N. D. ~ 10.33 | 4 | 4.4 |
| IcdP | 6 环 | 0.1 | 3.31 ~263.99 | 101.73 | N. D. ~31.06 | 1.29 | 4.4 |
| BghiP | 6 环 | 0.01 | 0.58 ~57.25 | 25.27 | N. D. ~16.05 | 2.09 | _ |
| LMW- | PAHs | | 8.75 ~267.00 | 139.45 | 10.44 ~62.16 | 39.41 | |
| HMW- | PAHs | | 23.30 ~1 444.49 | 669.6 | 15.44 ~ 145.80 | 62.28 | |
| \sum_{7} I | PAHs | | 18.16 ~1 289.92 | 576.32 | 14.82 ~94.79 | 62.6 | |
| $\sum P$ | AHs | | 35.94 ~ 2 032.73 | 940.61 | 27.94 ~ 242.95 | 107.77 | |

¹⁾ LMW-PAHs 表示低分子量(2~3 环) 多环芳烃化合物;HMW-PAHs 表示高分子量(4~5 环) 多环芳烃化合物;N. D. 表示未检出; \sum_7 PAHs 表示 7 种致癌多环芳烃(BaA、Chr、BbF、BkF、BaP、DBA、IcdP);TEF 表示毒性当量 $^{[10]}$

湖泊群以及厦门杏林湾表层沉积物中 PAHs 污染水平相当;大冶湖水体中 PAHs 含量远高于珠江近海口以及舟山青浜岛水体中 PAHs 的含量,略高于西江水体 PAHs 的含量,低于巢湖水体、湖南饮用水以及广东养殖鱼塘水体中 PAHs 含量,

远低于辽河大庆湖泊群以及钱塘江水体中 PAHs 含量.与国内相关研究对比可知大冶湖沉积物中 多环芳烃污染与国内湖泊、河流污染程度大致相同,而水体中多环芳烃较之于国内湖泊、河流污染程度较轻.

表 2 国内典型湖泊、河流中沉积物、水体 PAHs 含量1)

| Table 2 | Concentrations of PAHs | in the | sediment and | water from | domestic and | overseas typical lakes and rivers | |
|---------|-------------------------|--------|--------------|------------|--------------|-----------------------------------|---|
| rabie 2 | Concentiations of Latis | m me | sediment and | water mom | domesuc and | Overseas typical lakes and fivers | , |

| 项目 | 研究区域 | PAHs 种类 | 含量范围 | 均值 | 污染水平 | 文献 |
|------------|--------|---------|---------------------|----------|------|--------|
| | 柘林湾 | 16 | 71.03 ~ 148.64 | 107.87 | 中等 | [11] |
| | 千岛湖 | 16 | 258.00 ~906.00 | 558.00 | 中等 | [12] |
| | 巢湖 | 16 | 109.70 ~6 245.80 | 908.51 | 中等 | [13] |
| | 杏林湾 | 16 | 413.00 ~ 2748.81 | 949.56 | 中等 | [14] |
| 沉积物/ng·g-1 | 太湖 | 16 | 256.60 ~ 1 709.00 | 829.00 | 中等 | [15] |
| | 安庆湖泊群 | 16 | 5.5 ~ 3 608.8 | 941.50 | 中等 | [16] |
| | 珠江 | 16 | 217.00 ~ 2 680.00 | 1 028.00 | 高水平 | [6] |
| | 辽河 | 16 | 120.80 ~22 120.00 | 3 281.00 | 高水平 | [15] |
| | 大冶湖 | 16 | 35.94 ~ 2 032.73 | 940.60 | 中等 | 本研究 |
| | 珠江口 | 16 | 15.50 ~54.90 | _ | _ | [17] |
| | 舟山 | 16 | 45.96 ~ 101.08 | 66.45 | _ | [18] |
| | 西江 | 16 | 21.70 ~ 138.00 | _ | _ | [19] |
| | 巢湖 | 16 | 95.60 ~ 370.10 | 170.71 | _ | [13] |
| 水体/ng·L-1 | 湖南饮用水 | 16 | 70.22 ~673.80 | 253.13 | _ | [20] |
| | 广东养殖鱼塘 | 16 | 53.55 ~679.97 | 272.53 | _ | [21] |
| | 大庆湖泊群 | 16 | 200.00 ~ 1 210.00 | _ | _ | [22] |
| | 钱塘江 | 10 | 2 436.00 ~ 9 663.00 | 5 538.50 | _ | [23] |
| | 大冶湖 | 16 | 27.94 ~ 242.95 | 107.77 | _ | 本研究 |

¹⁾污染水平:低污染水平: $0 \sim 100 \text{ ng} \cdot \text{g}^{-1}$; 中等污染水平: $100 \sim 1000 \text{ ng} \cdot \text{g}^{-1}$; 高污染水平: $1000 \sim 5000 \text{ ng} \cdot \text{g}^{-1}$; 超高污染水平: $>5000 \text{ ng} \cdot \text{g}^{-1}$

2.2 表层沉积物及水体中 PAHs 分布及组成

2.2.1 表层沉积物及水体中 PAHs 分布

大冶湖表层沉积物及水体中 PAHs 分布及组成 如图 2 图所示. 由图 2(a) 可知, 沉积物中 PAHs 除 了 D8 处外其他采样处的含量都较高,因大冶湖周 边排污口很多,且尹家湖、红星湖、三里七湖这3个 内湖的污染物直接排放至大冶湖,对大冶湖污染影 响较大: 表层沉积物中 PAHs 分布呈现湖中心高于 岸边的趋势,可能与排污口冲刷,表层沉积物扰动岸 边较湖心处大相关,最高值点在 D5 处,而 D5 处水 体 PAHs 含量也较高,可能是排污口水体交换频繁、 水流将污染物更多携带到水流最低区域(湖心区 域),进而沉降到沉积物中,最小值出现在 D8 采样 点,主要是因为 D8 靠近金海煤业开发区,接近西塞 山开发区的南沿,新港物流园的西边界及韦源口排 污口,表层沉积物受水体扰动大,颗粒物很难沉积, 而表层沉积物中 PAHs 主要来自于水中悬浮颗粒 物[6],颗粒物所吸附的 PAHs 随颗粒物一起沉积至 沉积物中.

PAHs 在水体中的分布与在表层沉积物中的分

布大致呈相反趋势,库岸区高于湖心区, ∑PAHs含量峰值在人湖区 D1 处,最小值在湖中心区 D4处. D1、D2、D3 靠近尹家湖、红星湖、三里七湖这 3 个内湖入湖口,受内湖周边矿冶生产影响较大,其中 D1 处接近大港排污口; D4 为湖心位置,水体扰动较小,且受工矿企业等污染源影响较小,水体中多环芳烃含量较低;而 D6、D7、D8 处靠近金海煤业开发区,其中 D8 处水体污染较严重,PAHs含量较高;D5 处水体 PAHs含量也较高,一方面可能与 D5 处围网养鱼以及插网捕鱼有关,围网使得污染物集聚在网沿处,污染物难以扩散,从而引起水体 PAHs含量增加,柯盛等[25]证实养殖污染源对水体 PAHs 影响远大于生活等污染源的影响;另一方面可能与水体-沉积物处于吸附-解析动态平衡状态有关.

2.2.2 表层沉积物及水体中 PAHs 组成

有研究证实冶炼厂周边环境介质中 PAHs 组成以高环 PAHs 化合物为主^[26,27],大冶湖表层沉积物与水体中 PAHs 组成与其他冶炼厂周边环境中PAHs 组成相一致.由图 2(b)可知,大冶湖表层沉

积物中4~5 环 HMW-PAHs 明显高于2~3 环 LMW-PAHs 的含量,表层沉积物中4~5 环占 \sum PAHs 的65.7%~71.3%,平均71.2%;而在水体中主要以2 环以及4 环和5 环 PAHs 为主,占水体中 \sum PAHs 的68%~97.55%,其中 HMW-PAHs 含量占 \sum PAHs 的57.8%,3 环以及6 环 PAHs 含量基本可以忽略不计,特别是6环 PAHs 仅在 D1、D6、D8处有检出,说明大治湖水体及表层沉积物中 PAHs 受矿治冶炼影响较严重.水体中6环 PAHs 大多未检出而沉积物中6环 PAHs 含量也较低,但两者之

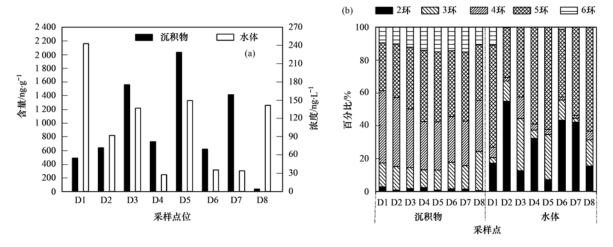


图 2 大冶湖表层沉积物及表层水体中 PAHs 分布及组成

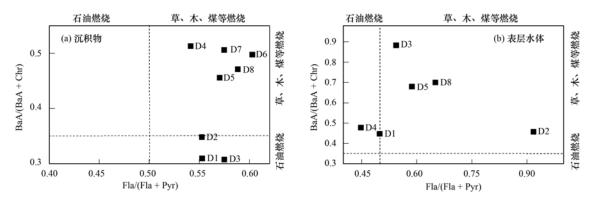
Fig. 2 Distribution and constitution of PAHs in the surface sediment and water from the Daye Lake

2.3 表层沉积物及水体中 PAHs 来源解析

在环境介质中 PAHs 来源主要是人为来源,自然来源贡献很低^[28,29]. 人为来源主要为高温燃烧过程排放和石油产品的挥发、泄漏、低温转化不完全燃烧等,HMW-PAHs 主要来自于生物质及化石燃料的高温燃烧所排放,而 2~3 环 LMW-PAHs 则主要来源于石油产品的低温转化、不完全燃烧以及石油及其精炼品的挥发、泄漏等^[28,29]. 本研究利用PAHs 组分分子比例诊断法来分析大冶湖表层沉积物及水体中PAHs 的来源(图 3).

运用同分异构体比值进一步判断 PAHs 的来源,在实际运用中,往往选择稳定性较好的 PAH 同系物作为源解析的标志物,基于 PAH 同系物之间比值来区分燃烧源以及石油源,包括 Ant/(Ant + Phe)、Fla/(Fla + Pyr)、BaA/(BaA + Chr)和 IcdP/(IcdP + BghiP)等.在本研究的水体中由于很多单体在水中未检出,则选取 Fla/(Fla + Pyr)、BaA/(BaA + Chr)来推断 PAHs 的来源.大量研究证实^[6,8,30,31],当 Fla/(Fla + Pyr) < 0.4 时,指示石油泄

漏源,介于0.4~0.5之间时,指示石油及其精炼品 的燃烧源,>0.5时,指示煤等化石燃料及生物质的 燃烧源; 当 BaA/(BaA + Chr) < 0.2 时, 表明主要是 石油泄漏污染源,>0.35 时表示煤等化石燃料及生 物质的燃烧源,介于两者之间则指示石油及其精炼 品的燃烧源. 在大冶湖水体及沉积物中 Fla/(Fla+ Pyr) 比值都大于 0.4, 表明大冶湖水体及沉积物中 PAHs 都源自于石油或者生物质、煤等的燃烧;在 大冶湖沉积物中 BaA/(BaA + Chr) 比值除了 D1、 D2、D3 介于 0.2~0.35 之间,其他点位比值都大于 0.35, 而在水体中由于 D6、D7 两个点位 BaA 及 Chr 未检出,其他点位 BaA/(BaA + Chr) 比值都大于 0.35,表明大冶湖水体及沉积物中 PAHs 主要受高 温燃烧来源的影响.来源解析显示大冶湖水体及沉 积物中 PAHs 主要来自于燃烧源,而黄石市是一个 矿冶之城,冶炼厂高温燃烧对于大冶湖水体沉积物 中 PAHs 有显著地贡献,在所有采样点沉积物中高 分子量 PAHs 都占据绝大部分,反映出了沉积物受 矿冶冶炼长期累积污染的效应,同时也表现出了



(a)沉积物;(b)表层水体

图 3 大冶湖表层沉积物及表层水体中 PAHs 来源解析

Fig. 3 Sources of the PAHs in the surface sediment and water from the Daye Lake

沉积物是 POPs 汇的特性.

2.4 沉积物生态风险评价

质量基准法和质量标准法是用来评估土壤及沉积物中 PAHs 潜在生态风险的常用方法,本研究利用 Long 等^[9]提出的沉积物质量基准以及加拿大于2006 年发布的多环芳烃沉积物质量标准^[12,32,33]对大治湖沉积物潜在生态风险进行表征,评价结果见表3.质量基准法分为效应低值(ERL)对生物体毒副

作用发生的风险几率 < 10% 和对生物体毒副作用发生的风险几率 > 50% 的效应中值(ERM),相对污染系数 RCF 是对沉积物中 PAHs 污染的定量表征,RCF 为沉积物中 PAHs 浓度与效应低值的比值;质量标准法分为 5 个阈值 REL、TEL、OEL、PEL、FEL,分别表示罕见效应、临界效应、偶然效应、可能效应以及频繁效应的浓度阈值^[12,32,33],将 16 种优控 PAHs 任一种超过最高限值的点位给标注出,见表 3.

表 3 沉积物质量基准法及质量标准法阈值及评价结果1)

| Table 3 Thre | shold and | evaluation | result of | sediment | quality | guidelines | and qualit | v standard |
|--------------|-----------|------------|-----------|----------|---------|------------|------------|------------|
|--------------|-----------|------------|-----------|----------|---------|------------|------------|------------|

| DAII | 质量基准 | 作法阈值 | 评价结果 | | 质 | 量标准法阈 | 值 | | 评价 | 结果 |
|----------------------|-------|-------------|--------------------|-------|-------|-------|-------|-------|----------------------------------|------------|
| PAHs | ERL | ERM | RCF > 1 点位 | REL | TEL | OEL | PEL | FEL | 区间 | 点位 |
| Nap | 160 | 2 100 | _ | 17 | 35 | 120 | 390 | 1 200 | > FEL | _ |
| Acy | 16 | 500 | _ | 3.3 | 5. 9 | 30 | 130 | 340 | | |
| Ace | 44 | 640 | _ | 3.7 | 6. 7 | 21 | 89 | 940 | $\mathrm{PEL} \sim \mathrm{FEL}$ | D3 、D5 、D7 |
| Flu | 19 | 540 | D3 , D6 , D7 | 10 | 21 | 61 | 140 | 1 200 | | |
| Phe | 240 | 1 500 | _ | 25 | 42 | 130 | 520 | 1 100 | | |
| Ant | 85. 3 | 1 100 | _ | 16 | 47 | 110 | 240 | 1 100 | $\mathrm{OEL} \sim \mathrm{PEL}$ | D2 、D4 、D6 |
| Fla | 600 | 5 100 | _ | 47 | 110 | 450 | 2 400 | 4 900 | | |
| Pyr | 665 | 2 600 | _ | 29 | 53 | 230 | 880 | 1 500 | | |
| BaA | 261 | 1 600 | _ | 14 | 32 | 120 | 390 | 760 | $\mathrm{TEL} \sim \mathrm{OEL}$ | D1 |
| Chr | 384 | 2 800 | _ | 26 | 57 | 240 | 860 | 1 600 | | |
| BaP | 430 | 1 600 | _ | 11 | 32 | 150 | 780 | 3 200 | | |
| DBA | 63.4 | 260 | D3 、D4 、D5 、D6 、D7 | 3.3 | 6. 2 | 43 | 140 | 200 | $\mathrm{REL} \sim \mathrm{TEL}$ | _ |
| BbF | N. A. | N. A. | | N. A. | | |
| BkF | N. A. | N. A. | | N. A. | < REL | D8 |

^{1) &}quot;N. A."表示无最低安全阈值,"一"表示没有相应点位

质量基准法评价结果显示:大冶湖表层沉积物中 Flu 及 DBA 两种单体 PAH 在 D3、D6、D7 都超出了 ERL,介于 ERL~ERM 之间,而 DBA 在 D4、D5也超出了 ERL,介于 ERL~ERM 之间,证明在大冶湖沉积物中 PAHs 潜在生态风险发生几率不大.但在 D5 处非常接近效应区间中值(ERM),且无最低

安全阈值的致癌 PAHs 单体 BbF、BkF 和 IcdP 在大 治湖沉积物中检出率均高达 100%, 无最低安全阈 值的 PAHs 只要存在于环境中就会对生物体产生毒 副作用.

质量标准法评价结果显示: D8 小于 REL, 低于 罕见效应浓度值, 对生物的不良影响概率可以忽略

不计,但 D8 处水体 PAHs 含量较高,尤其是 D8 处靠近两个工业园区,一个物流园区,需要对 D8 处水体以及排污口进行长期监测,防止 PAHs 污染进一步加重; D1 介于 TEL 和 OEL 之间,对生物的不良影响概率较低,但 D1 为大冶湖外湖入湖口,且水体中PAHs 含量最高,需加强检测,最好能够长期在线监测,控制污染物排放入湖; D2、D4、D6 这 3 个点位PAHs 浓度介于 OEL 与 PEL 之间,这 3 处表层沉积物中PAHs 对生物的潜在危害较大;表层沉积物中PAHs 对生物潜在危害最大出现在 D3、D5、D7 这 3 个点位,PAHs 浓度介于 PEL 与 FEL 之间,而 D5 处为淡水养殖边界处,且水体 PAHs 含量也较高,需要引起关注,并对沉积物毒性风险进行评估,对 PAHs 污染采取治理措施. 大冶湖所有采样点位中检出的PAHs 浓度效应值都低于 FEL 值,表明大冶湖沉积物

对生物体潜在风险不高. 但 D5、D6、D7 靠近大冶湖 渔业养殖区域,且生态风险概率也处于中度偏高水 准,因此该区域清淤工作需要重点清理,且清淤后的 底泥需要妥善处置,以免引起二次污染. 结合质量基 准法和质量标准法可以看出大冶湖表层沉积物中最 易对生物体造成危害的 PAHs 单体化合物是 Flu、 DBA 以及没有最低安全限值的 BbF、BkF 和 IcdP,对 生物的不良影响概率最高处是 D3、D5、D7.

2.5 水体健康风险评价

利用 USEPA 推荐的终生致癌风险(ILCRs)结合毒性当量浓度(TEQ_{BaP})^[10,34,35]对大冶湖水体中PAHs 对人体的潜在致癌风险进行定量表征,地表水及地下水体PAHs 主要经过摄入以及皮肤接触两个暴露方式会对人体产生潜在致癌风险,计算公式如下(其中各参数含义及取值见表4)^[35~37]:

$$\begin{split} \text{TEQ}_{\text{BaP}} &= \sum c_i \times \text{TEF} \\ \text{ILCRs}_{\frac{B}{A}} &= \frac{\text{TEQ}_{\text{BaP}} \times \text{IRW} \times \text{CSF}_i \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \\ \text{ILCRs}_{\frac{b}{b} \text{KE}} &= \frac{\text{TEQ}_{\text{BaP}} \times \text{SA} \times K_{\text{p}} \times \text{ABS} \times \text{ET} \times \text{EF} \times \text{ED} \times \text{CF} \times \text{CSF}_{\text{d}}}{\text{BW} \times \text{AT}} \end{split}$$

表 4 终生致癌风险评价参数^[35-37]
Table 4 Parameters used for estimation of the incremental lifetime cancer risks (ILCRs)

| 参数 | 含义 | 取值/单位 | 参数 | 含义 | 取值/单位 |
|---------------|--------|------------------------|------------|------------------|--------------------------------------|
| c_i | 化合物浓度 | 见表 1 | SA | 皮肤暴露面积 | $2~800~{\rm cm}^2$ |
| TEF | 毒性当量 | 见表 1 | $K_{ m p}$ | 皮肤渗透常数 | 0.001 |
| IRW | 直接摄人率 | 2. 0 L·d ⁻¹ | ABS | 皮肤吸收因素 | 0.001 |
| EF | 每年暴露天数 | 350 d | ET | 暴露时间 | 0.6 h·d ⁻¹ |
| ED | 持续暴露时间 | 70 a | CF | 转换系数 | 1 L•(1 000 cm ³) $^{-1}$ |
| \mathbf{BW} | 体重 | 70 kg | CSF_i | 摄人 BaP 的致癌斜率系数 | 7.3 kg·d·mg ⁻¹ |
| AT | 平均时间 | $ED \times EF d$ | CSF_d | 皮肤接触 BaP 的致癌斜率系数 | 25 kg·d·mg ⁻¹ |

由表 5 可知大冶湖水体中 PAHs 总致癌风险 (\sum ILCRs) 为 1.49×10⁻⁵ ~ 1.59×10⁻⁶ a⁻¹之间,均值为 5.08×10⁻⁶ a⁻¹;通过皮肤接触途径产生的致癌风险介于 4.57×10⁻⁹ ~ 4.26×10⁻⁸ a⁻¹,均值为 1.46×10⁻⁸ a⁻¹,通过直接摄入途径产生的致癌风险介于 1.56×10⁻⁵ ~ 1.48×10⁻⁶ a⁻¹之间,均值为 5.06×10⁻⁶ a⁻¹,通过摄入暴露途径产生的致癌风险是皮肤接触暴露途径的 10³ ~ 10⁴ 倍.大冶湖水体中 PAHs 总致癌风险(\sum ILCRs)都介于 USEPA所推荐的可接受致癌风险水平(10⁻⁶~10⁻⁴)之间,除 D1、D5、D8 这 3 处外都低于国际辐射防护委员会(ICRP)推荐的最大可接受风险水平 5.0×10⁻⁶ a⁻¹,D1 处致癌风险是 ICRP 最大可接受风险水平的 2.98 倍,但都高于瑞典环保局和英国皇家协会推荐

的最大可接受风险水平 1.0 × 10 ⁻⁶ a ⁻¹, D1 处甚至超出了该限值一个数量级. 致癌风险最高出现在D1 处,最低值出现在D6 处. 需要对入湖区加以防治,对 3 个内湖水体污染加以治理,防治内湖污染物对大冶湖水体的进一步污染. 大冶湖水体中ILCRs值分布与TEQ_{BaP}分布相一致,而TEQ_{BaP}值与7种致癌物质浓度有关,因此ILCRs值与7种致癌物质浓度有关,因此ILCRs值与7种致癌物质浓度全量高于D6处。最低值出现在D6处而不是 PAHs最低值D4处,因为D4处7种致癌PAHs浓度含量高于D6处也高于D7处,因此D4处健康风险反而比D6、D7处要高. 虽然大冶湖不是饮用水源地,但多环芳烃亲脂疏水特性使其可以在生物体内大量富集,通过食物链最终进入人体,而大冶湖为淡水渔业养殖场,因此实际致癌风险还大

| 表 5 水体中 PAHs 毒性当量浓度及终生致摄风险(| 表 5 | 水体中 PAHs 毒性当量浓度及终生致癌风险 | 命(ILC | R) |
|-----------------------------|-----|------------------------|-------|----|
|-----------------------------|-----|------------------------|-------|----|

| Table 5 | TEO _{PoP} and incremental | lifetime cancer risks of | f the PAHs in water | from the Dave Lake |
|----------|------------------------------------|--------------------------|----------------------|--------------------|
| 1 abic 3 | 1 LOD DOD and incremental | menne cancer risks o | a the rairs in water | Hom the Daye Lake |

| 项目 | D1 | D2 | D3 | D4 | D5 | D6 | D7 | D8 | 均值 |
|--|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|-----------|
| TEQ _{BaP} /ng·L ⁻¹ | 71. 03 | 10. 31 | 14. 71 | 9. 60 | 30. 32 | 7. 62 | 9. 51 | 41.02 | 24. 26 |
| 皮肤接触/a-1 | 4. 26E-08 | 6. 18E-09 | 8. 83E-09 | 5. 76E-09 | 1.82E-08 | 4. 57E-09 | 5. 70E-09 | 2.46E-08 | 1.46E-08 |
| 摄入/a ⁻¹ | 1.48E-05 | 2. 15E-06 | 3. 07E-06 | 2. 00E-06 | 6. 32E-06 | 1. 59E-06 | 1. 98E-06 | 8. 56E-06 | 5.06E-06 |
| \sum ILCRs /a $^{-1}$ | 1. 49E-05 | 2. 16E-06 | 3. 08E-06 | 2. 01E-06 | 6. 34E-06 | 1. 59E-06 | 1. 99E-06 | 8. 58E-06 | 5. 08E-06 |

于计算值.因此需要对大冶湖多环芳烃污染开展防治工作,特别是人湖区,需要对3个内湖排进大冶湖的水体进行监控,并对3个内湖水体加以治理.

3 结论

- (1)在大冶湖表层沉积物及水体中 16 种 EPA 优控 PAHs, \sum PAHs 分别为 35.94~2032.73 ng·g⁻¹、27.94~242.95 ng·L⁻¹,平均值含量分别为 940.61 ng·g⁻¹、107.77 ng·L⁻¹,表层沉积物污染呈现湖中高于岸边趋势,水体则呈相反趋势,表层沉积物中以 4~5 环高环化合物为主要组分,在水体中主要以 2 环以及 4 环和 5 环 PAHs 为主,大冶湖与国内其他湖泊污染水平相当,需对清淤后的污泥进行妥善处置.
- (2)来源解析表明大冶湖表层沉积物及水体中 多环芳烃主要来自于高温燃烧源,沉积物中 PAHs 高环分子都占据绝大部分,反映出了沉积物受矿冶 冶炼长期累积污染的效应,同时也表现出了沉积物 是 POPs 汇的特性.
- (3)大冶湖沉积物中各 PAHs 化合物的浓度均未超过 ERM 以及 FEL,说明大冶湖表层沉积物 PAHs 潜在生态风险发生概率不高,但是有3个点位已经超过了质量标准法的可能效应浓度值,且这3个点位靠近渔业养殖区,需要对环境及人体风险进行评价.
- (4)健康风险评价表明,大冶湖水体中 PAHs 终生致癌风险水平处于美国 EPA 推荐的可接受水平范围内,但都高于瑞典环保局和英国皇家协会推荐的最大可接受风险水平 1.0×10⁻⁶ a⁻¹,存在一定的潜在危害,7 种致癌 PAHs 的污染需要加强治理,需要对入湖区域污染加以治理,控制污染物入湖.

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