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高爽、白霉、白岩、雷团团、土刚、李时海、陆朝阳、七娜、郝明亮、黄同峰(1575) 2015~2017年北京及近周边平房燃煤散烃及其污染排放遥感测算 起文意、李令军、鹿海峰、姜磊、张立坤、王新辉、邱昀(1594) 基于地基遥感的杭州地区气溶胶光学特性 齐冰、车慧正、徐婷婷、杜荣光、胡德云、梁卓然、马千里、姚杰(1604) 四川省人为源挥发性有机物组分清单及其臭氧生成潜势 周子航、邓也、谭钦文、吴柯颖、宋丹林、黄凤霞、周小玲(1613) 餐饮源挥发性有机物组成及排放特征 高雅琴、王红丽、许睿哲、景盛翱、刘跃辉、彭亚荣(1627) 广州番禺大气成分站一次典型光化学污染过程 PAN 和 O3 分析 邹宇、邓雪娇、李菲、殷长秦(1634) 北京市典型道路扬尘化学组分特征及年际变化 胡月琪、李萌、颜起、张超(1645) 南昌市扬尘 PM、中多环芳烃的来源解析及健康风险评价 于瑞莲、郑权、刘贤荣、王珊珊、敖旭、张超(1646) 现实工况下挖掘机尾气排放特征分析 马帅、张凯山、王帆、庞凯莉、朱怡静、李臻、毛红梅、胡宝梅、杨锦锦、王斌(1670) 雾。罐天人体平均呼吸高度处不同粒径气溶胶的微生物特性 杨唐、韩云平、李珠、《敬(1688) 支持向量机回归在臭氧预报中的应用 苏筱倩、安俊琳、张玉欣、梁静舒、刘静达、王鑫(1697) 基于中国电网结构及一线典型城市车辆出行特征的 PHEV 二氧化碳排放分析 郝旭、王贺武、李伟峰、欧阳明高(1705) 岩溶槽谷区地下河硝酸盐来源及其环境效应:以重庆龙风槽谷地下河系统为例 标准,生工工建、吴韦、彭学义、刘九维(1715) 股州湾表层水体中邻苯二甲酸酯的污染特征和生态风险 刘成、孙翠竹、张哿、唐缭、邹亚丹、徐擎擎、李锋民(1726) 湛江湾沉积物中六六六(HCHs)、滴滴涕(DDTs)有机氯农药的分布特征与风险评估 张哿、唐缭、邹亚丹、徐擎擎、李锋民(1726) 湛江湾沉积物中六六六(HCHs)、滴滴涕(DDTs)有机氯农药的分布特征与风险评估 张哿、唐缭、邹亚丹、徐擎擎、李锋民(1726) 港位系化系统中,DOM 米偿特性及影响用表位任意、以为该准系表光,视镜明、陈法锦、于赤灵、李嘉诚、梁字钊、宋建中(1734)
内蒙古河套濯区不同盐碱程度土壤 CH。收収现律 物义柱,焦燕,物铭德,温息片(1950)水稻光合碳在植株-土壤系统中分配与稳定对施磷的响应 王莹莹,肖谋良,张昀,袁红朝,祝贞科,葛体达,吴金水,张广才,高晓丹(1957)土壤水分和温度对西南喀斯特棕色石灰土无机碳释放的影响 徐学池,黄媛,何寻阳,王桂红,苏以荣(1965)黄土丘陵区侵蚀坡面土壤微生物量碳时空动态及影响因素 覃乾,朱世硕,夏彬,赵允格,许明祥(1973)农用地土壤抗生素组成特征与积累规律 孔泉 是,张世文,爰起甲,胡青贵(1981)
  生物发酵制药 VOCs 与嗅味治理技术研究与发展 ··· 王东升,朱新梦,杨晓芳,焦茹媛,赵珊,宋荣娜,吕明晗,杨敏(1990)《环境科学》征订启事(1612) 《环境科学》征稿简则(1787) 信息(1663,1796,1833)
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胶州湾表层水体中邻苯二甲酸酯的污染特征和生态 风险

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摘要: 以胶州湾为研究区域,选取 15 种常用邻苯二甲酸酯(phthalate esters, PAEs)作为检测目标,采用搅拌棒吸附萃取-气相色谱质谱联用的方法,于 2015 年 8 月和 11 月以及 2016 年 1 月对胶州湾开展 3 次大面积调查,检测分析了胶州湾表层水体中 PAEs 的含量、组成、空间分布和季节变化,同时对胶州湾水体中 PAEs 进行了风险评价.结果表明:①2015 年 8 月和 11 月、2016 年 1 月胶州湾表层海水中 PAEs 的总浓度范围分别为 3.63~21.20、2.24~12.60 和 0.01~4.15 μg·L⁻¹,平均浓度分别为 11.10、5.26 和 0.80 μg·L⁻¹.②受入海径流和洋流影响,胶州湾表层水体中 PAEs 浓度表现出近岸高,远岸低,且总体东岸浓度高于西岸.与国内外其他研究相比,胶州湾表层水体中 PAEs 含量处于中等水平,但在海洋中属于污染较严重的海域.③3 个季节 PAEs 浓度分布差异较大,受降雨量等因素影响整体呈现夏季 > 秋季 > 冬季的趋势,3 个季节主要检出种类均为 DBP、BBP 和 DEHP.④生态风险评估结果表明,胶州湾各站位 DBP 的风险商值(RQ)均大于 1,即生态风险较大,其他 PAEs(RQ < 1)风险较小.PAEs 已成为胶州湾中一类具有潜在威胁的有机污染物,其在环境中的行为和生态危害仍需进一步研究.

关键词:胶州湾;邻苯二甲酸酯(PAEs);空间分布;季节变化;生态风险评价 中图分类号: X55 文献标识码: A 文章编号: 0250-3301(2019)04-1726-08 **DOI**: 10.13227/j. hjkx. 201808217

Pollution Characteristics and Ecological Risk Assessment of Phthalate Esters (PAEs) in the Surface Water of Jiaozhou Bay

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Abstract: In order to better understand phthalate esters (PAEs) pollution in Jiaozhou Bay, surface water samples were collected for content analysis in August and November 2015 and January 2016. Fifteen kinds of PAEs were detected by stir bar sorptive extraction and gas chromatography-mass spectrometry. The content, composition, spatial distribution, seasonal variation, and ecological risk assessment of the PAEs in the surface waters were examined and analyzed. The results showed that: ① The total concentrations of PAEs in the surface seawater in August and November 2015 and January 2016 were 3.63-21.20 μg·L⁻¹, 2.24-12.60 μg·L⁻¹, and 0.01-4.15 μg·L⁻¹, respectively, and the average concentrations were 11.10 μg·L⁻¹, 5.26 μg·L⁻¹, and 0.80 μg·L⁻¹, respectively. ② Influenced by runoff and ocean currents, the concentration of PAEs in the surface water of Jiaozhou Bay is higher near the shore and lower in the middle, and the total concentration of the east coast is higher than that of the west coast. Compared with surveys of other rivers and oceans, the PAEs content in the surface water of Jiaozhou Bay is moderate, but it is heavily polluted compared with other oceans. ③ Considering the rainfall and other factors, the distribution of PAEs in the three seasons followed the order summer > autumn > winter. The main PAEs detected were DBP, BBP, and DEHP. ④ The results of the ecological risk assessment showed that the risk quotient (RQ) of DBP at each station in Jiaozhou Bay was greater than 1; that is, the ecological risk was relatively large, while the risk of the other PAEs (RQ < 1) was small. PAEs have become a potentially threatening organic pollutant in Jiaozhou Bay, phthalate esters (PAEs); spatial distribution; seasonal variation; ecological risk assessment

邻苯二甲酸酯(PAEs)是海洋中最常检测到的内分泌干扰物(EDCs)之一,被广泛添加到玩具,化妆品,食品包装和医疗器械等产品中以增强塑料制品的可塑性和强度^[1,2].全球每年的 PAEs 产量从1975年的180万t迅速增加到2011年的800多万t^[3].中国每年生产的PAEs超过4.50×10⁶t,消费量高达2.20×10⁶t^[4].由于PAEs与塑料制品(例如聚氯乙烯,聚丙烯)之间是以范德华力或氢键链接,因而PAEs易脱离塑料制品释放到周围环境中,

并在大气,水,土壤中广泛存在^[1]. 目前在世界各海域中均检测出不同浓度的 PAEs^[3,5,6]. 作为一种环境激素, PAEs 对水生生物具有致癌、致畸、致突变效应,并能通过食物链传递对人类的健康造成威

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胁^[1,3].因此,美国环保局(US EPA)已将6种PAEs列入129种优先控制的污染物名单中,包括邻苯二甲酸二甲酯(DMP)、邻苯二甲酸二乙酯(DEP)、邻苯二甲酸二正丁酯(DBP)、邻苯二甲酸二正辛酯(DNOP)、邻苯二甲酸丁苄酯(BBP)和邻苯二甲酸二(2-乙基)己酯(DEHP).同时,我国也将DMP、DBP和DNOP列为优先控制污染物.PAEs污染的广泛性、严重性已经引发了国内外研究学者和管理者的广泛关注.

胶州湾是位于青岛的一个半封闭性的浅水海 湾,与海泊河、李村河和大沽河等多条河流相 连[7], 湾内水体自净能力较差. 山东半岛蓝色经济 区的快速发展, 使环胶州湾地区的工农业生产、港 口运营、水产养殖、城市旅游和居民生活等过程中 产生的生活污水更多地随入海河流进入胶州湾, 使 胶州湾成为城市污水的排放沟渠, 对胶州湾产生了 严重的环境压力. 目前已有许多研究对胶州湾内的 重金属[7]、多环芳烃[8,9]、多溴联苯醚[10]、壬基 酚[11]等的污染现状进行了调查, 但对于胶州湾表 层水体中 PAEs 的污染现状和分布特征却鲜见报 道. 如张道来等[12] 仅研究了青岛近岸表层沉积物 中 PAEs 的分布特征, 发现数个样品中 PAEs 污染 严重且未对水体中 PAEs 的浓度情况进行检测, 缺 乏对胶州湾整个海区水环境中 PAEs 的分布特征及 季节差异的研究,因此本研究以2015年8月、2015 年11月和2016年1月采集的胶州湾21个站点内 表层水体样品为研究对象, 重点分析了 PAEs 的含 量组成、空间分布、季节变化和生态风险, 以期为 胶州湾水环境保护提供基础数据.

1 材料与方法

1.1 胶州湾表层水体采样点空间分布

为了解胶州湾表层水体中 PAEs 浓度分布的空间特征和季节变化,本研究分3个航次分别于2015年8月(夏季,共计19个站位)、2015年11月(秋季,共计14个站位)和2016年1月(冬季,共计14个站位)对胶州湾表层水体进行采样,各采样点的经纬度与其他信息如图1.

水样采集方法:用 5 L 的不锈钢铁桶经润洗后 采集表层水样(水深 0~1 m 之间),装入润洗后的 1 L 的玻璃烧杯中.用孔径 0.7 μ m,直径 47 mm 的 GF/F 玻璃纤维膜(450 $^{\circ}$ C,灼烧 5 h)过滤,去除水中的颗粒物,取 250 mL 抽滤后的水样装入棕色玻璃样品瓶中,加入 0.5 mL 50 g·L⁻¹的叠氮化钠固定,4 $^{\circ}$ C 冰箱内避光保存.记录采样时间、采样地点、样品站位号与编号.

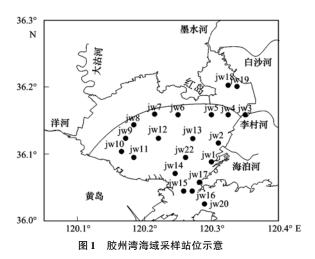


Fig. 1 Sampling stations of Jiaozhou Bay

1.2 供试试剂与仪器

- (1)标准品 15 种 PAEs 的混合标准品(DMP、DEP、DIBP、DBP、DMEP、DMPP、DEEP、DPP、DHXP、BBP、DBEP、DCHP、DEHP、DPhP 和DNOP)和内标物苯甲酸苄酯(benzyl benzoate,BBZ)浓度均为1 mg·L⁻¹,纯度 99.5%,均购自德国 Dr. Ehrenstorfer.
- (2)实验试剂 甲醇(德国 Merck 公司), 乙腈 (天津科密欧公司), 正己烷(意大利 Sigma 公司), 二氯甲烷(天津科密欧公司); 氯化钠(NaCl), 购自 国药集团化学试剂有限公司.
- (3)主要仪器 安捷伦 7890-5975C 气相色谱质谱联用仪(美国,安捷伦), HB-5MS 毛细管柱色谱柱(30 m×0.25 μm×0.25 μm),多点加热磁力搅拌器(德国, IKA),超声波清洗器 KQ-3200B(昆山),微量进样器(上海高鸽),吸附磁力搅拌棒,聚二甲基硅氧烷(PDMS)涂层厚度 1 mm,长度 10 mm

1.3 胶州湾表层海水中 PAEs 检测方法[13]

量取 100 mL 超纯水,加入 10 mL 的甲醇和 100 μL 内标 BBZ 使水样中内标物的浓度为 1 μg·L⁻¹.混匀后,移取 30 mL 水样于 30 mL 的玻璃瓶中,加入 0.57 g NaCl(调节样品海水盐度至 5%),加入吸附萃取搅拌棒,立即拧紧盖子,每组样品 3 个平行.在室温条件下,以 360 r·min⁻¹的转速进行搅拌萃取,萃取时间 2 h. 萃取完成后,用镊子夹出搅拌棒放入内插管中,加入 200 μL 甲醇和 50 μL 乙腈作解析溶剂,在超声条件下进行解析 50 min,解析溶液供 GC-MS 分析.将萃取搅拌棒置于色谱进样瓶中,加满甲醇超声清洗,单次超声 30 min 后更换新的甲醇,重复 3 次,以备下次使用.

1.4 色谱条件

本实验测定 PAEs 采用 GC-MS 进行分析, 色

谱柱类型为 DB-5MS(30 m×0.25 mm×0.25 μm) 毛细管柱,色谱条件是:进样口温度 300℃;柱箱起始温度 70℃,保持 2 min;以 25 ℃·min⁻¹升至 150℃;以 3 ℃·min⁻¹升至 170℃;以 30 ℃·min⁻¹升至 185℃;以 3 ℃·min⁻¹升至 195℃;以 60 ℃·min⁻¹升至 225℃;以 8 ℃·min⁻¹升至 228℃,保持 10 min. 氦气作为运载气体,流速为 1 mL·min⁻¹. 传输线温度为 280℃,离子源温度为 230℃. 选择离子扫描(SIM)模式,不分流进样 1 μL,溶剂延迟 6 min.

1.5 质量控制与质量保证

实验过程中全部使用玻璃或不锈钢金属器材,所有玻璃器皿需先 20% 硫酸酸缸中浸泡 24 h,冲洗烘干后,450°C于马弗炉中灼烧 5 h. 玻璃纤维膜和铝箔使用前均 450°C于马弗炉中灼烧 5 h,贮存在不锈钢铁盒中,每组样品均设 3 个平行样. 水体中 15种 PAEs 检出限为 0.25 ng·L $^{-1}$ (DBP) \sim 489 ng·L $^{-1}$

(DMEP), 回收率为80%~113%.

1.6 数据分析

采用软件 Excel 2010、Spss Statistics 19.0、Surfer 2012 和 Origin 9.1 对实验数据进行处理及分析.

2 结果与讨论

2.1 胶州湾表层水体中 PAEs 的含量、组成

3 个航次各站点检出的 13 种 PAEs 浓度的分析结果如表 1. 2015 年 8 月胶州湾所有站位的水样中检出 12 种 PAEs, 其总浓度为 3.63 ~ 21.20 $\mu g \cdot L^{-1}$, 平均值为 11.10 $\mu g \cdot L^{-1}$. 2015 年 11 月共检出 8 种 PAEs, 总浓度为 2.24 ~ 12.60 $\mu g \cdot L^{-1}$, 平均值为 5.26 $\mu g \cdot L^{-1}$. 2016 年 1 月胶州湾共检出 8 种 PAEs, 总浓度为 0.01 ~ 4.15 $\mu g \cdot L^{-1}$, 平均值为 0.80 $\mu g \cdot L^{-1}$. 其中 DBP、BBP 和 DEHP 的检出率在 3 个航次中均达到 100%.

表 1 胶州湾表层海水中 13 种 PAEs 的浓度分布 1 / $\mu g \cdot L^{-1}$

2015-11 2015-08 2016-01 项目 浓度范围 浓度范围 平均值 平均值 浓度范围 平均值 n.d. 0.36 ± 0.47 n. d. ~8.34 0.97 ± 2.07 n. d. n. d. ~1.56 DMP n. d. ~14.80 2.46 ± 4.99 n. d. n. d. ~0.18 DEP n. d. // 0.08 ± 0.08 n. d. ~1.81 0.35 ± 0.52 0.24 ± 0.22 DIBP n. d. ~0.71 n. d. ~ 0.81 0.19 ± 0.22 0.04 ~ 11.20 DBP 2.67 ± 2.31 0.65 ~ 1.38 0.98 ± 0.22 $0.01 \sim 0.09$ 0.03 ± 0.03 BBP 1.11 ~ 5.69 1.09 ~ 1.10 2.24 ± 1.24 1.09 ± 0.00 0.28 ~ 0.09 0.01 ± 0.02 n. d. ~1.87 DMPP 0.37 ± 0.55 $0.00 \sim 0.02$ n. d. ~1.16 1.28 ± 0.36 0.01 ± 0.01 DPP n. d. ~1.01 0.42 ± 0.50 n. d. n. d. n. d. ~0.01 0.00 ± 0.00 DHXP n. d. ~1.14 0.54 ± 0.31 n. d. n. d. n. d. n. d. DEHP $0.44 \sim 1.06$ 0.61 ± 0.18 $0.22 \sim 0.33$ 0.28 ± 0.04 $0.00 \sim 1.58$ 0.35 ± 0.45 DEEP n. d. ~1.69 0.18 ± 0.53 n. d. ~2.16 1.42 ± 0.49 n. d. n. d. DNOP n. d. n. d. n. d. ~2.41 1.16 ± 1.17 n. d. n. d. DBEP n. d. ~1.22 0.06 ± 0.28 n. d. ~3.38 2.83 ± 0.32 n. d. n.d. DCHP n. d. ~1.23 0.32 ± 0.56 n. d. n. d. n. d. n. d. > PAEs $3.63 \sim 21.20$ 11.10 ± 5.44 2.24 ~ 12.60 5.26 ± 2.73 0.01 ~4.15 0.80 ± 1.10

Table 1 Concentration of the 13 PAEs in the surface water in Jiaozhou Bay/µg·1

1)n.d. 表示该项未检出,下同

10 种主要 PAEs 浓度分布情况如图 2,分析发现 2015 年 8 月胶州湾中 PAEs 主要由 DBP 和 BBP 贡献,2015 年 11 月 DMPP、DEEP、DBP 和 BBP 是 PAEs 的主要贡献者,2016 年 1 月各类 PAEs 的含量相当,其中 DMP、DBP 和 DEHP 贡献量较大.其中 DEP 和 DBP 被广泛应用于化妆品中[1,2,14]. DEHP 主要来源于塑料和重化工产业[15].此外,DMP、DEP、DBP 和 DEHP 也是生活垃圾(例如玩具、塑料包装等)中的主要 PAEs^[3].可见,胶州湾表层水体中 PAEs 主要来源于塑料和重化工工业,以及生活垃圾.不同季节 PAEs 组成的差别主要源自 PAEs

自身性质的差异和降雨量的影响. 研究发现由于冬季降水量小, 烷基链较长的 DBP、BBP、DEHP, 辛醇-水分配系数($\lg K_{ow}$)较大, 更易吸附在颗粒物或者死亡藻细胞表面沉降到沉积物中^[3], 因而烷基链较短的 DMP 在冬季贡献量偏高.

与已报道的其他河流、湖泊和海洋相比(表2), 胶州湾表层水体中 PAEs 含量处于中等水平, 但在海洋中属于污染较严重的海域. 导致胶州湾地区 PAEs 污染严重的原因一是由于胶州湾属于典型的半封闭的富营养化高危型海湾[16], 水交换能力弱, 水交换时间为 80 d, 而湾顶部及西部部分区域

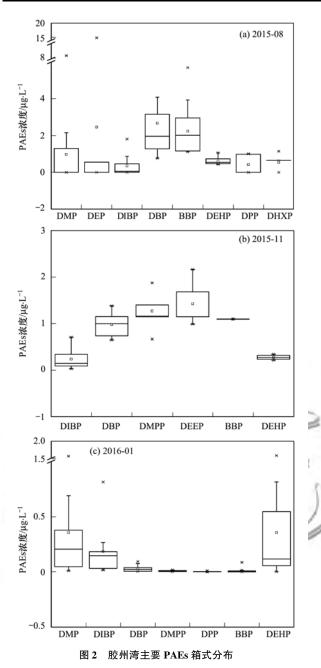


Fig. 2 Box plot of the main PAEs in Jiaozhou Bay

水交换时间可达 120 d^[17],沿岸山东半岛蓝色经济区发展迅速,众多入海河流携带大量污染物进入胶州湾,沿岸污水处理厂污水的排放以及海上航运、跨海大桥等污染源连续输入都增加了胶州湾中PAEs 污染. 此外,沿岸污水处理厂废水即使均达标排放,由于很多有机污染物如 PAEs 并没有列入污水处理标准中,污水处理厂的处理装置对 PAEs 的去除率仅能达到 18%^[18,19],进一步加剧了胶州湾中 PAEs 污染.

2.2 胶州湾表层海水中 PAEs 的空间分布特征

胶州湾表层水体中 PAEs 的空间分布如图 3、4 所示. 2015 年 8 月各个站位 PAEs 浓度普遍较高,其中胶州湾东北部的李村河、娄山河、墨水河和白沙河入海口附近的 jw3、jw4、jw6、jw19 和湾口的 jw15、jw16 和 jw17 站位的 PAEs 总浓度均高于 10 μg·L $^{-1}$. 2015 年 11 月东北部李村河入海口附近的 jw4 和洋河、大沽河入海口附近的 jw9、jw10 和 jw11 站位的 PAEs 含量均高于6 μg·L $^{-1}$,而 2016 年 1 月 除海泊河人海口附近的 jw1 外,含量均低于 2 μg·L $^{-1}$.

总体来看,胶州湾表层水体中 PAEs 的空间分布表现为近岸浓度较高,中部浓度较低,且总体东岸浓度高于西岸.夏季东北部高浓度 PAEs 产生是受胶州湾入海径流输入的影响,青岛市工业区主要分布在东岸,且李村河,海泊河等入海河流基本无自身径流^[7],携带大量有机污染物进入胶州湾,另有团岛污水处理厂和海泊河污水处理厂的污水均直接排入胶州湾,给胶州湾东岸海水带来巨大的环境压力,此外,鱼虾贝类的养殖场很多分布在东北部的红岛、李沧附近海域,加重了该地区的 PAEs 污染^[10].夏季湾口处(站位 jw20)浓度也很高,湾口处是胶州湾与外海交换的关键点,夏季该地区主要

表 2 世界不同水域中邻苯二甲酸酯类浓度比较/ $\mu g \cdot L^{-1}$

	Table 2 Compar	ison of PAEs concentr	ations in different bas	ins of the world∕ μg∙	Γ_{-}	
地点	DMP	DEP	DBP	BBP	DEHP	文献
Marseille 湾,法国	0.00 ~ 0.01	0.00 ~ 0.03	0.01 ~0.32	0.00 ~4.90	0.04 ~ 0.80	[6]
Orge 河,法国	n. d. ~0.09	n. d. ~1.43	$0.04 \sim 0.20$	n. d. ~0.14	n. d. ~2.08	[20]
Rhone River,法国	0.01	0.03	0.04	0.01	0.41	[21]
Mediterranean Sea,法国	0.00	0.01	0.09	0.00	0.10	
Dutch 河,荷兰	$0.00 \sim 0.19$	$0.07 \sim 2.30$	$0.07 \sim 3.10$	n. d.	$0.90 \sim 5.00$	[22]
Pretoria,南非	n. d.	n. d.	$0.18 \sim 0.34$	n. d.	0.06 ~ 0.24	[23]
Cape Town,南非	n. d.	n. d.	$0.23 \sim 0.63$	n. d.	$0.06 \sim 3.42$	[23]
地中海	$0.00 \sim 0.14$	$0.02 \sim 0.05$	n. d.	n. d.	$0.03 \sim 0.62$	[24]
北京什刹海,中国	$0.05 \sim 0.14$	$0.01 \sim 0.01$	$0.01 \sim 0.16$	n. d. ~0.51	$0.14 \sim 0.52$	[25]
长江武汉段,中国	n. d. ~0.30	n. d. ~0.37	n. d. ~35.60	n. d.	$0.01 \sim 54.73$	[26]
长江重庆段,中国	$0.13 \sim 6.44$	0.21 ~4.16	$0.21 \sim 60.70$	n. d.	$0.70 \sim 26.30$	[27]
台湾,中国	n. d.	n. d. ~2.50	1.00 ~ 13.50	n. d.	1.00 ~ 18.50	[28]
珠江口,中国	$0.04 \sim 0.14$	n. d. ~0.95	$0.06 \sim 2.04$	0.79 ~ 5.32	1.08 ~8.84	[29]
胶州湾,中国	n. d. ~2.15	n. d. ~14.90	0.04 ~ 11.20	1.11 ~ 5.70	0.44 ~ 1.07	本研究

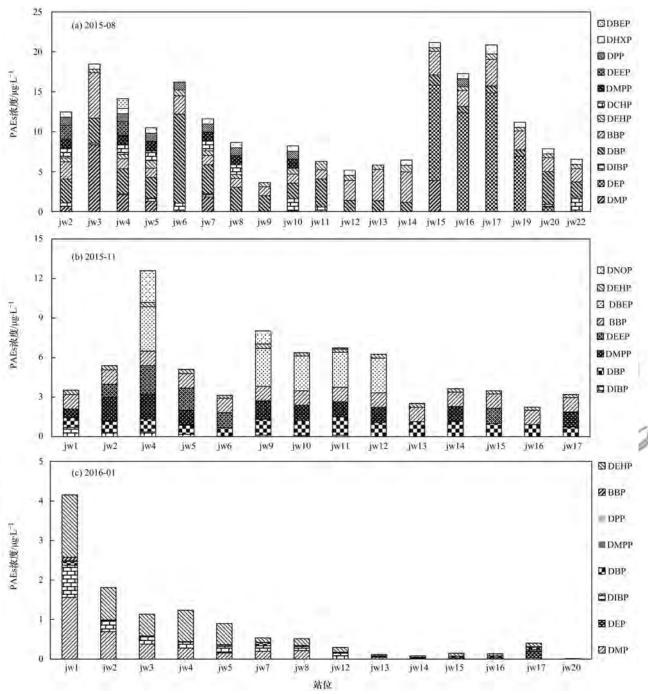


图 3 胶州湾各站位表层水体中 PAEs 含量组成

Fig. 3 Concentration and distribution of PAEs in the surface water of Jiaozhou Bay

是东南季风,影响了胶州湾与黄海的水交换,导致了湾口高浓度的出现.另外,沿跨海大桥分布的jw4~jw7这4个站位含量偏高可能是受到跨海大桥交通污染物排放的影响.秋季胶州湾西北部jw9~jw12的高浓度 PAEs 主要源自大沽河和洋河的入海径流,大沽河是青岛最大的河流,沿岸常住人口达241.9万人[16],对大沽河水质产生很大压力.

2.3 胶州湾表层海水中 PAEs 的季节性变化

如表 1 和图 4 所示, 胶州湾夏季、秋季和冬季 PAEs 的总含量均值分别为 11.10、5.26 和 0.80 μ g·L⁻¹. 结果表明 PAEs 的总含量呈现夏季(2015

年8月)>秋季(2015年11月)>冬季(2016年1月)的趋势,且主要检出种类相似,DBP、BBP、DEHP含量最高,但秋季和冬季检出的PAEs种类(8种)和单种PAE的浓度较夏季(12种)均有所减少.

季风气候、水文条件和 PAEs 自身的理化性质是半封闭的胶州湾 PAEs 含量季节差异最重要的因素^[16,30]. 胶州湾地区为典型的温带季风气候,夏季受东南季风影响产生的风生流会使污染物向北部堆积^[31],而冬季西北季风又会加速湾内污染物向湾外运输,导致夏季胶州湾内 PAEs 含量普遍偏高,

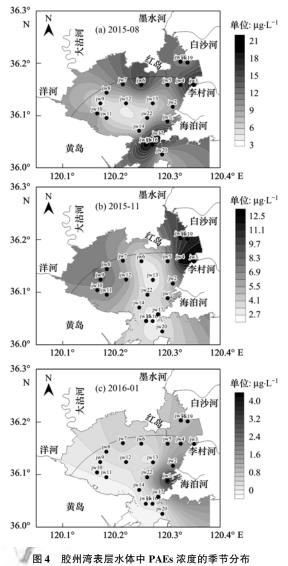


Fig. 4 Seasonal variation of the PAEs content in the surface water of Jiaozhou Bay

且最高浓度出现在东北部, 秋冬季节 PAEs 浓度下降显著. 胶州湾人海径流多为季风雨源型河流, 受季节降雨量影响很大, 如大沽河 6~9 月汛期径流量是全年总量的 78.3% [16]. 夏季降雨量大, 雨水将大量污水和固体废物带入河流, 通过地表径流再

2.4 胶州湾表层海水中 PAEs 的生态风险评估

为了解胶州湾表层水体中 PAEs 的生态风险情况,对 3 个季节各采样站位中 4 种主要 PAEs 的生态风险进行了评估,采用了欧盟适用于现有化学物质与新化学物质的风险评价技术指南(technical guidance document, TGD)中的商值风险评估方法,对 4 种 PAEs 的风险商值(risk quotient, RQ)进行分析:

$$RQ = \frac{MEC}{PNEC}$$

式中, MEC 为环境中 PAEs 的测量浓度, PNEC 为预测的无效应浓度, DMP、DEP、DBP 和 DEHP 的PNEC 值分别为 96、96、0.50 和 1.54^[30,34], 结果如表 3.2015 年 8 月各个站位 DMP、DEP、DBP 和 DEHP 的 RQ 平均值为 0.03、0.08、5.35 和 0.40,即 4 种 PAEs 的生态风险大小为: DBP > DEHP > DEP > DMP. 2015 年 11 月 DBP 和 DEHP 的 RQ 平均值为 1.95 和 0.18,生态风险大小为: DBP > DEHP. 2016 年 1 月 DMP、DEP、DBP 和 DEHP 的 RQ 平均值为 0.00、0.00、0.06 和 0.23,4种PAEs的生态风险大小为: DEHP > DBP

表 3 胶州湾表层水体中主要 PAEs 生态风险评价结果

Table 3 Ecological risk assessment results for the PAEs in the surface water of Jiaozhou Bay

日期(年-月)	PAEs	水体浓度/μg·L ⁻¹	RQ	RQ 平均值
	DMP	0 ~ 2. 15	0 ~ 0. 09	0.03
2015-08	DEP	0 ~ 14. 80	0 ~ 0. 16	0.08
2013-08	DBP	0.04 ~11.20	1.53 ~22.30	5. 35
	DEHP	0.46 ~ 1.06	0. 28 ~ 0. 69	0.40
2015-11	DBP	0. 65 ~ 1. 38	1. 30 ~ 2. 70	1. 95
2013-11	DEHP	0. 22 ~ 0. 33	0. 14 ~ 0. 20	0. 18
	DMP	0 ~ 1.56	0 ~ 0. 02	0.00
2016-1	DEP	0 ~ 0. 18	0 ~ 0.00	0.00
2010-1	DBP	0.01 ~ 0.09	0. 01 ~ 0. 19	0.06
	DEHP	0.00 ~ 1.58	0.00 ~ 1.02	0. 23

> DMP > DEP. 胶州湾表层水体中只有8月和11月的所有站位中 DBP的 RQ > 1,表明夏季和秋季胶州湾表层水体中 DBP存在较大的生态风险,但在冬季1月的结果中 DBP生态风险较小.此外,3个季节中各个站位的 DMP、DEP、DEHP计算所得RQ值均小于1,表明 DMP、DEP、DEHP的生态风险在可接受范围.此外,4种 PAEs在3个不同季节的RQ值均表现为夏季 > 秋季 > 冬季,与 PAEs总体的季节分布规律一致.

3 结论

胶州湾表层水体中 PAEs 的污染已相当普遍,15 种被监测的 PAEs 中有 13 种被检出,其中 DBP、BBP、DEHP 含量较高. 总体来说,胶州湾表层水体中 PAEs 浓度表现出近岸高,中部低,且总体东岸浓度高于西岸. 受降雨量和 PAEs 自身理化性质等因素影响,水体中 PAEs 浓度表现出夏季 > 秋季 > 冬季的趋势. 与其他河流海洋相比,本研究中胶州湾表层水体中 PAEs 含量处于中等水平,但在海洋中属于污染较严重的海域. 夏季和秋季胶州湾表层水体中 DBP 具有潜在的生态风险,DMP、DEP和DEHP的生态风险在可接受范围内. 因此需加强胶州湾中 PAEs 尤其是 DBP 的监控.

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