

目次

基于机器学习的珠三角秋季臭氧浓度预测	陈镇, 刘润, 罗征, 薛鑫, 汪瑶, 赵志军 (1)
粤港澳大湾区大气PM _{2.5} 浓度的遥感估算模型	代园园, 龚绍琦, 张存杰, 闵爱莲, 王海君 (8)
典型输送通道城市冬季PM _{2.5} 污染与传输变化特征	代武俊, 周颖, 王晓琦, 齐鹏 (23)
郑州市夏季PM _{2.5} 中二次无机组分污染特征及其影响因素	和兵, 杨洁茹, 徐艺斐, 袁明浩, 翟诗婷, 赵长民, 王申博, 张瑞芹 (36)
重庆典型城区冬季碳质气溶胶的污染特征及来源解析	彭超, 李振亮, 向英, 王晓宸, 汪凌韬, 张晟, 翟崇治, 陈阳, 杨复沐, 翟天宇 (48)
2022年8月成渝两地臭氧污染差异影响因素分析	陈木兰, 李振亮, 彭超, 邓也, 宋丹林, 谭钦文 (61)
2020年“三连击”台风对我国东部地区O ₃ 污染的影响分析	花丛, 尤媛, 王峰, 张碧辉 (71)
北京城区夏季VOCs初始体积分数特征及来源解析	张博韬, 景宽, 王琴, 安欣欣, 鹿海峰, 王陈婧, 王友峰, 刘保献 (81)
机动车减排降碳综合评价体系综述	范朝阳, 佟惠, 梁晓宇, 彭剑飞 (93)
基于LEAP模型的长三角某市碳达峰情景	杨峰, 张贵驰, 孙佑, 谢放尖, 揣小伟, 孙瑞玲 (104)
广东省船舶二氧化碳排放驱动因素与减排潜力	翁淑娟, 刘颖颖, 唐凤, 沙青娥, 彭勃, 王辉嘉, 陈诚, 张雪驰, 李京洁, 陈豪琪, 郑君瑜, 宋献中 (115)
给水厂典型工艺碳排放特征与影响因素	张翔宇, 胡建坤, 马凯, 高欣慰, 魏月华, 韩宏大, 李克勋 (123)
中国饮用水中砷的分布特征及基于伤残调整寿命年的健康风险评估	窦殿程, 齐媛, 肖淑敏, 苏高新, 郭宇新 (131)
太湖水体和沉积物中有机磷酸酯的时空分布和风险评估	张成诺, 钟琴, 栾博文, 周涛, 顾帆, 李伟飞, 邹华 (140)
水产养殖环境中农兽药物的污染暴露水平及其风险影响评价	张楷文, 张海燕, 孔聪, 顾润润, 田良良, 杨光昕, 王媛, 陈凤, 沈晓盛 (151)
长江朱沱断面磷浓度与通量变化及来源解析	姜保峰, 谢卫民, 黄波, 刘旻璇 (159)
珠江河口地表水锰氧化物对磷的“载-卸”作用	李睿, 梁作兵, 伍祺瑞, 杨晨晨, 田帝, 高磊, 陈建耀 (173)
富春江水库浮游植物功能群变化的成因	张萍, 王炜, 朱梦圆, 国超旋, 邹伟, 许海, 朱广伟 (181)
合浦盆地西部地区地下水水化学特征及形成机制	陈雯, 吴亚, 张宏鑫, 刘怀庆 (194)
新疆车尔臣河流域绿洲带地下水咸化与污染主控因素	李军, 欧阳宏涛, 周金龙 (207)
京津冀地区生态系统健康时空演变及其影响因素	李魁明, 王婉燕, 姚罗兰 (218)
近30年辽河三角洲生态系统服务价值时空演变及影响因素分析	王耕, 张芙蓉 (228)
光伏电站建设对陆地生态环境的影响:研究进展与展望	田政卿, 张勇, 刘向, 陈生云, 柳本立, 吴纪华 (239)
大兴安岭林草交错带植被NDVI时空演变及定量归因	石淞, 李文, 曲琛, 杨子仪 (248)
西南地区不同类型植被NPP时空演变及影响因素探究	徐勇, 郑志威, 孟禹弛, 盘钰春, 郭振东, 张炎 (262)
不同海拔梯度下极端气候事件对松花江流域植被NPP的影响	崔嵩, 贾朝阳, 郭亮, 付强, 刘东 (275)
基于InVEST与CA-Markov模型的昆明市碳储量时空演变与预测	帕茹克·吾斯曼江, 艾东, 方一舒, 张益宾, 李牧, 郝晋琨 (287)
基于PLUS-InVEST模型的酒泉市生态系统碳储量时空演变与预测	石晶, 石培基, 王梓洋, 程番苑 (300)
长江下游沿江平原土壤发育过程中碳库分配动态	胡丹阳, 张欢, 宿宝巍, 张娅璐, 王永宏, 纪佳辰, 杨洁, 高超 (314)
漓江流域喀斯特森林土壤有机碳空间分布格局及其驱动因子	申楷慧, 魏识广, 李林, 储小雪, 钟建军, 周景钢, 赵毅 (323)
不同土地利用方式对岩溶区土壤有机碳组分稳定性的影响	陈坚淇, 贾亚男, 贺秋芳, 江可, 陈畅, 叶凯 (335)
紫色土丘陵区坡地柑橘园土壤碳氮的空间分布特征	李子阳, 陈露, 赵鹏, 周明华, 郑静, 朱波 (343)
氮添加与凋落物处理对橡胶林砖红壤有机碳组分及酶活性的影响	薛欣欣, 任常琦, 罗雪华, 王文斌, 赵春梅, 张永发 (354)
重庆化肥投入驱动因素、减量潜力及环境效应分析	梁涛, 赵敬坤, 李红梅, 王妍, 曹中华, 张务帅, 王孝忠, 郭超仪, 石孝均, 陈新平 (364)
中国土壤中全氟和多氟烷基物质的分布、迁移及管控研究进展	刘浩然, 邢静怡, 任文杰 (376)
基于多源辅助变量和随机森林模型的耕地土壤重金属含量空间分布预测	解雪峰, 郭炜炜, 濮励杰, 缪源卿, 蒋国俊, 张建珍, 徐飞, 吴涛 (386)
基于源导向的农用地土壤重金属健康风险评估及优先控制因子分析	马杰, 葛森, 王胜蓝, 邓力, 孙静, 蒋月, 周林 (396)
铜陵某废弃硫铁矿矿区土壤重金属污染特征及来源解析	李如忠, 刘宇昊, 黄言欢, 吴鸿飞 (407)
天水市主城区公交站地表灰尘重金属来源解析及污染评价	李春艳, 王新民, 王海, 吕晓斌 (417)
基于大田试验的土壤-水稻镉对不同调理剂的响应	唐乐斌, 刘新彩, 宋波, 马丽钧, 黄凤艳 (429)
腐殖质活性组分对土壤镉有效性的调控效应与水稻安全临界阈值	胡秀芝, 宋毅, 王天雨, 蒋珍芬, 魏世强 (439)
生物炭与铁钙材料对镉砷复合污染农田土壤的修复	吴秋产, 吴骥子, 赵科理, 连斌, 袁峰, 孙洪, 田欣 (450)
人体微塑料污染特征及健康风险研究进展	马敏东, 赵洋尘, 朱龙, 王伟平, 康玉麟, 安立会 (459)
聚苯乙烯微塑料联合镉污染对土壤理化性质和生菜(<i>Lactuca sativa</i>)生理生态的影响	牛佳瑞, 邹勇军, 简敏菲, 黄楚红, 李金燕, 穆霆, 刘淑丽 (470)
转录组分析植物促生细菌缓解高粱微塑料和重金属复合污染胁迫机制	刘泳岐, 赵超禹, 任学敏, 李玉英, 张英君, 张浩, 韩辉, 陈兆进 (480)
微塑料对土壤中养分和镉淋失的影响	赵群芳, 褚龙威, 丁原红, 王发园 (489)
微塑料和非对土壤化学性质、酶活性及微生物群落的影响	刘沙沙, 秦建桥, 吴贤格 (496)
民勤荒漠绿洲过渡带人工梭梭林土壤细菌群落结构及功能预测	王安林, 马瑞, 马彦军, 吕彦勋 (508)
不同灌溉水盐度下土壤真菌群落对生物炭施用的响应	郑志玉, 郭晓雯, 闵伟 (520)
厨余垃圾有机肥对土壤微生物活性及功能的影响	刘美灵, 汪益民, 金文豪, 王永冉, 王嘉和, 柴一博, 彭丽媛, 秦华 (530)
土壤真菌群落结构对辣椒长期连作的响应特征	陈芬, 余高, 王谢丰, 李廷亮, 孙约兵 (543)
叶面喷施硅肥对再生水灌溉水稻叶际细菌群落结构及功能基因的影响	梁胜贤, 刘春成, 胡超, 崔二苹, 李中阳, 樊向阳, 崔丙健 (555)
昌黎县海域细菌群落和抗生素抗性基因分析	王秋水, 程波, 刘悦, 邓婕, 徐岩, 孙朝徽, 袁立艳, 左嘉, 司飞, 高丽娟 (567)
基于高通量定量PCR与高通量测序技术研究城市湿地公园抗生素抗性基因污染特征	黄福义, 周曙仡, 潘婷, 周昕原, 苏建强, 张炯 (576)
城区第四系沉积柱中抗生素的垂向分布特征及环境影响因素	刘可, 童蕾, 甘翠, 王逸文, 张嘉越, 何军 (584)
氢氧化钾改性玉米秸秆生物炭对水中土霉素的吸附特性及机制	刘总堂, 孙玉凤, 费正皓, 沙新龙, 温小菊, 钱彬彬, 陈建, 谷成刚 (594)
CO ₂ 气氛热解与硝酸改性的生物炭Pb ²⁺ 吸附性能对比	江豪, 陈瑞芝, 朱自洋, 王琳, 段文斌, 陈芳媛 (606)

太湖水体和沉积物中有机磷酸酯的时空分布和风险 评估

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摘要: 为探究有机磷酸酯(OPEs)在太湖水环境中的时空分布特征和生态风险水平,利用固相萃取和超高效液相色谱-串联质谱方法测定太湖水体和沉积物中的单体和低聚OPEs,其中单体OPEs包括烷基OPEs、氯代烷基OPEs和芳香基OPEs。结果发现,OPEs在太湖水体和沉积物的空间分布差异不显著;水体和沉积物赋存的OPEs皆有明显的季节差异性,夏季OPEs的赋存含量平均值(水体中为752.7 ng·L⁻¹,沉积物中为124.0 ng·g⁻¹)显著高于冬季(水体中为498.5 ng·L⁻¹,沉积物中为54.5 ng·g⁻¹),其中单体OPEs的季节差异性尤为明显;不同类型OPEs在水体和沉积物中的赋存浓度排序均为:氯代烷基OPEs > 烷基OPEs > 芳香基OPEs ≈ 低聚OPEs;OPEs在水体和沉积物间的伪分配系数(K_d)在夏季(0.05 ~ 4.17 L·g⁻¹)高于冬季(0.02 ~ 3.47 L·g⁻¹),且与其辛醇水分配系数显著正相关。基于OPEs在太湖水体中的中位浓度,采用风险商值(RQ)法评估太湖水环境OPEs的生态风险,发现水体中的OPEs整体上呈现中等生态风险(RQ在冬、夏两季分别约为0.34和0.35);冬夏两季不同类型OPEs的生态风险排序与OPEs的赋存浓度相对应;单体OPEs在夏季的生态风险高于冬季;虽然评估结果表明低聚OPEs的生态风险较低,并且低于单体OPEs,但目前有关低聚OPEs的毒理研究还鲜见报道,亟需开展深入研究以提供数据支持。

关键词: 有机磷酸酯(OPEs); 太湖; 地表水; 沉积物; 生态风险

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Spatiotemporal Occurrence of Organophosphate Esters in the Surface Water and Sediment of Taihu Lake and Relevant Risk Assessment

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Abstract: Aiming to explore the spatiotemporal occurrence of organophosphate esters (OPEs) in the aquatic environment of Taihu Lake and to assess the relevant ecological risk, monomeric and oligomeric OPEs in the surface water and sediment of Taihu Lake were determined using solid phase extraction and ultra-high performance liquid chromatography-tandem mass spectrometry. The target monomeric OPEs included chlorinated OPEs, alkyl OPEs, and aryl OPEs. There was no significant difference in the spatial distribution of OPEs in water and sediment between the pollutant-impacted and less-impacted zones. The average concentrations of OPEs in summer and winter were, respectively, 752.7 and 498.5 ng·L⁻¹ in water and 124.0 and 54.5 ng·g⁻¹ in sediment, indicating an obvious seasonal difference, especially for the monomeric OPEs. The OPEs levels in both the water and sediment were ranked in the order of chlorinated OPEs > alkyl OPEs > aryl OPEs ≈ oligomeric OPEs. The pseudo-partitioning coefficients (K_d) of OPEs between the sediment and surface water of Taihu Lake were much higher in summer (0.05-4.17 L·g⁻¹) compared to those in winter (0.02-3.47 L·g⁻¹) and were significantly positively correlated with the lg K_{ow} values of OPEs. Based on the median concentrations of OPEs in the water of Taihu Lake, the ecological risk of OPEs was assessed by risk quotient (RQ) values, which indicated a medium risk at an RQ level of 0.34 during winter and 0.35 during summer. The assessment results showed that the risk ranking of OPEs was consistent with that of their concentrations, and the monomeric OPEs posed a higher ecological risk in summer compared to that in winter. Although the ecological risk of oligomeric OPEs in this study was not serious and was lower than that of monomeric OPEs, it is an urgent requirement to conduct ecotoxicology studies on oligomeric OPEs in the future since available data is highly limited at present.

Key words: organophosphate esters (OPEs); Taihu Lake; surface water; sediment; ecological risk

有机磷系阻燃剂(organophosphate flame retardants, OPFRs)作为溴代阻燃剂的替代品,在全球的需求增长迅速,近些年已跻身于第一大类阻燃剂行列^[1,2]。中国是OPFRs生产大国,拥有201家生产厂,约占全球55%的份额^[3]。广泛应用的OPFRs主要为有机磷酸酯(organophosphate esters, OPEs),OPEs按取代基的不同可划分为烷基OPEs、氯代烷基OPEs和芳香基OPEs。该阻燃剂通过物理混合而非化学结合的方式添加到电子设备、纺织品、塑料、新能源汽车、电缆和家具等产品中^[4],因而容易经挥发和磨损释放到环境中^[2]。截至目前,OPEs在大气、室

内、地表水、污水、土壤等环境中皆有检出^[5-9]。然而,有关OPEs在环境中赋存特征的研究仍然存在很大空白。例如:大多数研究仅关注单体OPEs而忽略了新兴低聚OPEs在环境中的赋存状况^[10];不同季节条件下环境中OPEs的赋存差异还鲜有报道。鉴于OPEs的潜在危害,如引发斑马鱼甲状腺功能障碍^[11]、抑制鲢鱼生长发育^[12]、影响孩童骨骼健康^[13]、

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增强直肠癌细胞活性^[14]等,非常有必要针对 OPEs 的环境赋存特征开展更广泛、更深入的研究,进而为 OPEs 生态环境风险的评估以及相关治理技术的探索提供支持。

地表水环境与人类息息相关,具有生物多样性保护、饮用水供给、农业灌溉、水产养殖和休闲娱乐等重要功能^[15,16]。近几年,OPEs 在地表水环境中的赋存情况日益引起关注。曹渺等^[17]、李栋等^[18]和 Lv 等^[19]分别对黄河、长江和鄱阳湖中赋存的单体 OPEs 开展了相关研究,发现氯代 OPEs 在水体中的浓度显著高于其他 OPEs,并且此类 OPEs 的潜在风险高。除单体 OPEs 外,地表水环境中的低聚 OPEs 也不容忽视,黄河^[20]、长江^[21]和莱州湾^[21]等地皆检出低聚 OPEs,并且有研究发现太湖沉积物中双酚 A-双(二苯基磷酸酯)(BDP)的浓度与单体 OPEs 的浓度相当^[10]。在地表水环境的水体和沉积物中,Zhao 等^[10]和 Liu 等^[22]的研究发现 OPEs 的赋存状况存在明显差异,这一现象一方面可能归因于水体是污染物的载体而沉积物是污染物的“汇”和“源”,另一方面可能与 OPEs 的物化特性(如 $\lg K_{ow}$ 值)有关^[23]。此外,有学者针对不同季节下 OPEs 的赋存情况,以黄河^[20]和长江^[24]为模型开展了研究,发现 OPEs 的赋存有显著性

季节差异。因而,针对典型地表水环境,同时调研水体和沉积物中单体和低聚 OPEs 的赋存特征,并分析季节的影响有重要意义。

太湖是中国第三大淡水湖泊,是重要的饮用水水源地和水产养殖基地^[25],毗邻苏州、无锡和湖州等经济发达地区,是中国污染较为严重的大型湖泊之一^[26,27]。本研究选择太湖作为地表水环境模型,分析了 12 种单体 OPEs 和 3 种低聚 OPEs 在冬夏两季太湖水体和沉积物中的污染水平和分布特征,并采用风险商值法评估了不同类型 OPEs 在冬夏两季间的风险差异。本研究既可以补充不同季节太湖水环境中 OPEs 赋存差异的研究空白,也可以为太湖流域的 OPEs 防控和环境管理提供依据,还可以为地表水环境中 OPEs 风险评估模型的建立提供数据支持。

1 材料与方法

1.1 标样与试剂

15 种目标 OPEs 的标准品(纯度 $\geq 99\%$)购自德国 Dr. Ehrenstorfer 公司和加拿大 Toronto Research Chemicals 公司,其名称及理化性质如表 1 所示。实验分析所用的乙腈(色谱级)、二氯甲烷(色谱级)和甲醇(色谱级)购自国药集团化学试剂公司(北京)。

表 1 15 种目标 OPEs 的名称及理化性质
Table 1 Names and physicochemical properties of the 15 target OPEs

分类	名称	缩写	CAS 号	分子式	相对分子质量	$\lg K_{ow}$	25℃时溶解度 / $\text{mg} \cdot \text{L}^{-1}$
烷基 OPEs	磷酸三丁酯	TBP	126-73-8	$\text{C}_{12}\text{H}_{27}\text{O}_4\text{P}$	266.3	4	2.8×10^2
	磷酸三乙酯	TEP	78-40-0	$\text{C}_6\text{H}_{15}\text{O}_4\text{P}$	182.1	0.8	4.9×10^5
	磷酸三丙酯	TPrP	513-08-6	$\text{C}_9\text{H}_{21}\text{O}_4\text{P}$	224.1	2.3	4.0×10^5
	磷酸三(丁氧基乙基)酯	TBEP	78-51-3	$\text{C}_{18}\text{H}_{39}\text{O}_7\text{P}$	398.5	3.8	1.1×10^3
	磷酸三辛酯	TEHP	78-42-2	$\text{C}_{24}\text{H}_{51}\text{O}_4\text{P}$	434.6	4.2	2.0×10^{-6}
氯代烷基 OPEs	磷酸三(2-氯乙基)酯	TCEP	115-96-8	$\text{C}_6\text{H}_{12}\text{Cl}_3\text{O}_4\text{P}$	285.5	1.8	7.8×10^4
	三(2-氯异丙基)磷酸酯	TCPP	13674-84-5	$\text{C}_9\text{H}_{18}\text{Cl}_3\text{O}_4\text{P}$	327.6	2.6	1.6×10^3
	磷酸三(1,3-二氯异丙基)酯	TDCP	13674-87-8	$\text{C}_9\text{H}_{15}\text{Cl}_6\text{O}_4\text{P}$	430.9	3.7	7.0
芳香基 OPEs	磷酸三甲苯酯	TCrP	1330-78-5	$\text{C}_{21}\text{H}_{21}\text{O}_4$	368.4	6.3	0.1
	磷酸三间甲苯酯	TMPP	563-04-2	$\text{C}_{21}\text{H}_{21}\text{O}_4\text{P}$	368.4	5.4	8.1×10^{-7}
	磷酸三苯酯	TPhP	115-86-6	$(\text{C}_6\text{H}_5)_3\text{PO}_4$	326.3	4.6	1.9
	2-乙基己基二苯基磷酸酯	EHDPP	1241-94-7	$\text{C}_{20}\text{H}_{27}\text{O}_4\text{P}$	362.4	5.7	1.9
低聚 OPEs	双酚 A-双(二苯基磷酸酯)	BDP	5945-33-5	$\text{C}_{39}\text{H}_{34}\text{O}_8\text{P}_2$	692.6	8.2	1.9×10^{-1}
	间苯二酚四苯基二磷酸酯	RDP	57583-54-7	$\text{C}_{30}\text{H}_{24}\text{O}_8\text{P}_2$	574.5	6.4	7.4×10^{-2}
	2,2-双氯甲基-三亚甲基-双[双(2-氯乙基)磷酸酯]	V6	38051-10-4	$\text{C}_{13}\text{H}_{24}\text{Cl}_6\text{O}_8\text{P}_2$	583.0	1.9	58.0

1.2 研究区域和样品采集

太湖坐落在经济发达、人口密集的中国长三角地区,其水域面积为 $2\,338\text{ km}^2$,水深为 1.9 m ^[28]。太湖具有明显的进水和出水区域,其北部及西北部为主要的进水区,出水主要通过东南部的太浦河流出^[29,30]。太湖周边的常住人口及企业在北部及西北部尤为密集,因而有更多的污染物从这些区域入

湖^[31~33]。基于太湖进出水方向及污染物入湖特点,本研究将太湖划分为污染物高影响区和低影响区,采样点分布如图 1 所示。

样品采集工作于 2022 年 1 月(冬季)和 8 月(夏季)开展,采样点共计 17 个,依次用 L1 ~ L17 表示,具体位置如图 1 和表 2 所示。在每个采样点,表层水样使用不锈钢采水器采集约 $1\,200\text{ mL}$ (水面下 $0 \sim 0.5$

m),避光保存于棕色玻璃瓶中;表层沉积物样品使用不锈钢抓泥器采集约 500 g(深度 0 ~ 10 cm),放入装有铝箔的塑封袋内.水样和沉积物样品皆冷藏运回实验室,水样在 24 h 内完成预处理,沉积物样品放入 -80℃冰箱内以备冷冻干燥.

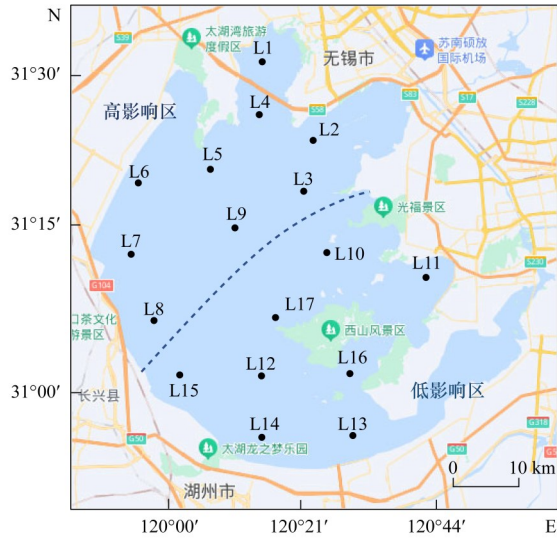


图 1 太湖采样点示意

Fig. 1 Sampling locations in Taihu Lake

表 2 太湖采样点的地理坐标信息

Table 2 Geographic coordinates of sampling locations in Taihu Lake

采样点	经度(E)/(°)	纬度(N)/(°)
L1	120.172 767	31.468 611
L2	120.246 161	31.378 889
L3	120.229 464	31.310 278
L4	120.162 222	31.396 619
L5	120.096 944	31.333 333
L6	119.963 889	31.311 111
L7	119.958 363	31.216 667
L8	119.984 127	31.139 818
L9	120.103 333	31.225 833
L10	120.269 722	31.232 778
L11	120.439 255	31.189 404
L12	120.150 575	31.062 778
L13	120.299 982	30.977 494
L14	120.134 667	30.993 056
L15	120.028 696	31.060 364
L16	120.311 624	31.053 876
L17	120.189 722	31.134 722

1.3 样品预处理

水样先后用 1.2 μm 和 0.7 μm 的玻璃纤维滤膜(直径 47 mm,Whatman,英国)过滤,过滤后的水样量取 1 L,用盐酸调 pH 为 3.然后,采用固相萃取法对水样进行净化和浓缩,所用的固相萃取小柱为 ENVI-18(6 mL,500 mg,Supelclean,美国),固相萃取步骤如下:用 15 mL 二氯甲烷、15 mL 乙腈和 15 mL 超纯水(pH = 3)对小柱进行活化;以每秒 1 ~ 2 滴的流速上

样;上样完毕后用 10 mL 含 10% 甲醇的超纯水淋洗;淋洗完成后负压抽干 30 min;干燥结束后,使用 12 mL 的二氯甲烷和乙腈混合溶剂(体积比为 1:3)进行洗脱;收集洗脱液氮吹至干,最后用乙腈定容至 500 μL.

沉积物样品经冷冻干燥后,研磨并过 100 目筛,取 1 g 样品置于 30 mL 玻璃离心管内,采用超声离心法提取样品中的目标 OPEs,具体步骤如下:在样品中加入 10 mL 乙腈,涡旋振荡 1 min,超声 30 min,离心 20 min(3 000 r·min⁻¹),上述步骤重复 3 次,合并上清液至棕色玻璃瓶内,加入超纯水(pH = 3)定容至 500 mL.然后,参考水样固相萃取的步骤对定容后的样品进行固相萃取,氮吹至干后用乙腈定容 1 mL.

1.4 目标 OPEs 分析

目标 OPEs 分析所用的仪器为超高效液相色谱-三重四级杆串联质谱仪(UPLC-MS/MS, Waters ACQUITY UPLC Xevo TQ,美国),配置的色谱柱型号为 Waters ACQUITY BEH C18(2.1 mm×100 mm,1.7 μm).色谱分离的柱温为 40℃,进样量为 5 μL,以 5 mmol·L⁻¹乙酸铵为流动相 A,以乙腈为流动相 B,流动相流速为 0.4 mL·min⁻¹,相关的梯度洗脱程序如下:0 ~ 0.25 min,10% B;0.25 ~ 3 min,10% ~ 90% B;3 ~ 4 min,90% B;4 ~ 4.01 min,90% ~ 10% B;4.01 ~ 5 min,10% B.质谱分析的条件为:离子源温度 150℃,脱溶剂温度 500℃,锥孔反吹气流量 50 L·h⁻¹.脱溶剂气流量 900 L·h⁻¹,碰撞气流量 0.15 mL·min⁻¹.所有目标 OPEs 在电喷雾正离子模式下分析,相关的离子对、驻留时间和碰撞能信息如表 3 所示.

表 3 目标 OPEs 的质谱信息

Table 3 Mass spectrometry conditions for the target OPEs

化合物名称	母离子(<i>m/z</i>)	子离子 ¹⁾ (<i>m/z</i>)	驻留时间/min	碰撞能/eV
TEP	182.9	98.79	0.027	18
TPrP	225.28	98.85	0.022	16
TBP	267.32	98.84	0.22	18
TCEP	285.18	98.90	0.027	24
TCPP	329.00	99.00	0.022	30
TPhP	327.29	215.05	0.022	28
EHDPP	362.90	250.86	0.058	20
TMPP	368.90	164.96	0.029	48
TCrP	368.97	164.99	0.029	50
TBEP	399.40	299.33	0.029	12
TDCP	430.83	98.82	0.022	24
TEHP	435.17	98.78	0.022	14
RDP	574.84	151.74	0.029	66
V6	582.65	360.77	0.022	20
BDP	693.40	367.19	0.078	34

1)子离子为定量离子

1.5 质量控制与保障(QA/QC)

OPEs的定量分析方法为外标法,为有效实现质量控制与保障,采用自来水和空白沉积物样品制备目标OPEs的标准溶液以避免基质效应.具体方法为:首先按照样品的预处理方法(1.3节)对自来水和空白沉积物进行预处理,然后在氮吹至干复溶后的乙腈溶液中加入15种目标OPEs的标准品,配制浓度为1、5、10、50、100和200 $\mu\text{g}\cdot\text{L}^{-1}$ 的标准溶液.空白样品中皆未检出OPEs,15种OPEs的标准曲线线性相关系数 R^2 均大于0.99.水样OPEs检出限(LOD, $S/N=3$)为0.01~0.34 $\text{ng}\cdot\text{L}^{-1}$,方法定量限(LOQ, $S/N=10$)

$$RQ_{\text{MEC/PNEC}} = \sum_{i=1}^n \frac{\text{MEC}_i}{\min(\text{EC50}_{i, \text{algae}}, \text{EC50}_{i, \text{crustacean}}, \text{EC50}_{i, \text{fish}})} \times \text{AF}$$

式中,MEC表示太湖水体中目标OPEs的实际浓度($\mu\text{g}\cdot\text{L}^{-1}$);PNEC为预测无效应浓度($\mu\text{g}\cdot\text{L}^{-1}$),通过查找最为敏感的水生生物的半数致死浓度,再按照计算公式得到;MEC_{*i*}表示检测的 n 类混合物中的第 i 个组分的实际最大环境浓度;EC50_{*i*}是指 n 类混合物中第 i 个组分的半致死浓度(包括藻类、甲壳类、鱼类的半致死浓度); $RQ_{\text{MEC/PNEC}}$ 表示基于MEC/PNEC总和的风险商;AF为评价因子,采用欧盟水框架指令的推荐值10~1 000.当 $RQ < 0.1$ 时,生态风险低;当 $0.1 \leq RQ < 1.0$ 时,生态风险中等;当 $RQ \geq 1.0$ 时,生态风险高.

1.7 数据处理

采用Origin 2021和Excel 2020软件处理数据并作图,采用IBM SPSS Statistics 25分析不同类型OPEs的季节差异性以及高、低影响区间的差异性(独立样本 T 检验).

2 结果与讨论

2.1 太湖水体中OPEs的时空分布特征

在冬夏两季,太湖水体中OPEs在各采样点的浓度分布如图2所示.由图2可知,15种目标OPEs除RDP在部分采样点检出外(冬季检出率为35.3%,夏季检出率为41.2%),其余OPEs在冬夏两季的检出率均为100%.与2014年^[10]和2019年^[25]太湖水体中OPEs的检出率相比,本研究发现有6种OPEs(TBEP、TEHP、EHDPP、TMPP、BDP和RDP)的检出率有明显增加,预示着太湖周边OPEs的使用及排放正在逐年增多.

从空间分布来看,在高影响区, $\rho(\sum_{15} \text{OPEs})$ 范围为390.5~624.4 $\text{ng}\cdot\text{L}^{-1}$ (冬)和662.81~1 002.20 $\text{ng}\cdot\text{L}^{-1}$ (夏);在低影响区, $\rho(\sum_{15} \text{OPEs})$ 范围为388.10~679.18 $\text{ng}\cdot\text{L}^{-1}$ (冬)和315.79~1 161.39 $\text{ng}\cdot\text{L}^{-1}$ (夏).

为0.03~1.14 $\text{ng}\cdot\text{L}^{-1}$;沉积物OPEs检出限(LOD, $S/N=3$)为0.09~0.25 $\text{ng}\cdot\text{g}^{-1}$,方法定量限(LOQ, $S/N=10$)为0.12~0.68 $\text{ng}\cdot\text{g}^{-1}$.在每批样品分析时,同时检测一个加标样品(质量控制样品),用于分析回收率,以保障检测质量.水样的加标回收率为64.8%~100.3%,沉积物的加标回收率为61.0%~99.7%.实验过程用到的所有玻璃容器均经过自来水和超纯水清洗3遍,最后超声烘干以避免环境污染.

1.6 生态风险评估

采用风险商值(risk quotient, RQ)法对太湖水体中OPEs的风险进行评估,计算公式如下所示^[21,24]:

$$\text{MEC}_i$$

通过独立样本 T 检验分析发现,在冬夏两季, $\sum_{15} \text{OPEs}$ 在两个区域之间皆没有显著性差异($P > 0.05$).张文萍等^[25]研究发现,太湖西北部OPEs的浓度水平较高,但从整个太湖来看OPEs在水体中的浓度无明显差异,与本研究的发现基本一致.庄园^[34]的研究表明,太湖水体中的OPEs同样在西北部污染较为严重,并将太湖水体中OPEs的来源归因于入湖河流的携带.

基于太湖水体中OPEs空间分布差异性小的特点,本研究从整个太湖的角度分析了不同类型OPEs在冬夏两季的赋存浓度(图3).在冬季,太湖水体中氯代烷基OPEs的浓度最高,尤其是TCEP(99.71~181.00 $\text{ng}\cdot\text{L}^{-1}$)和TCPP(161.42~231.02 $\text{ng}\cdot\text{L}^{-1}$);烷基OPEs中的TBP和TEP的浓度也较高,其浓度范围分别为28.85~124.29 $\text{ng}\cdot\text{L}^{-1}$ 和5.49~138.95 $\text{ng}\cdot\text{L}^{-1}$;芳香基OPEs和低聚OPEs的浓度相对较低,最高浓度皆低于50 $\text{ng}\cdot\text{L}^{-1}$,其中浓度较高的芳香基OPEs有EHDPP(0.70~33.40 $\text{ng}\cdot\text{L}^{-1}$)和TPhP(1.76~24.05 $\text{ng}\cdot\text{L}^{-1}$),浓度较高的低聚OPEs为BDP(0.30~19.44 $\text{ng}\cdot\text{L}^{-1}$).在夏季,不同类型OPEs的浓度排序和冬季基本一致:氯代烷基OPEs>烷基OPEs>芳香基OPEs≈低聚OPEs.但是,值得注意的是夏季 $\sum_{15} \text{OPEs}$ 的浓度明显高于冬季,其中单体OPEs中除了芳香基OPEs之外,其余单体OPEs(氯代烷基OPEs和烷基OPEs)的浓度在夏季显著高于冬季,另外,虽然低聚OPEs的浓度在两个季节之间不存在显著差异,但在夏季的浓度更高(表4).夏季浓度较高的OPEs包括TCEP(66.33~436.00 $\text{ng}\cdot\text{L}^{-1}$)、TCPP(102.98~496.75 $\text{ng}\cdot\text{L}^{-1}$)、TBP(84.13~207.71 $\text{ng}\cdot\text{L}^{-1}$)、TEP(31.45~229.25 $\text{ng}\cdot\text{L}^{-1}$)、和BDP(0.60~21.92 $\text{ng}\cdot\text{L}^{-1}$).

张文萍等^[25]调研了太湖流域中13种单体OPEs,

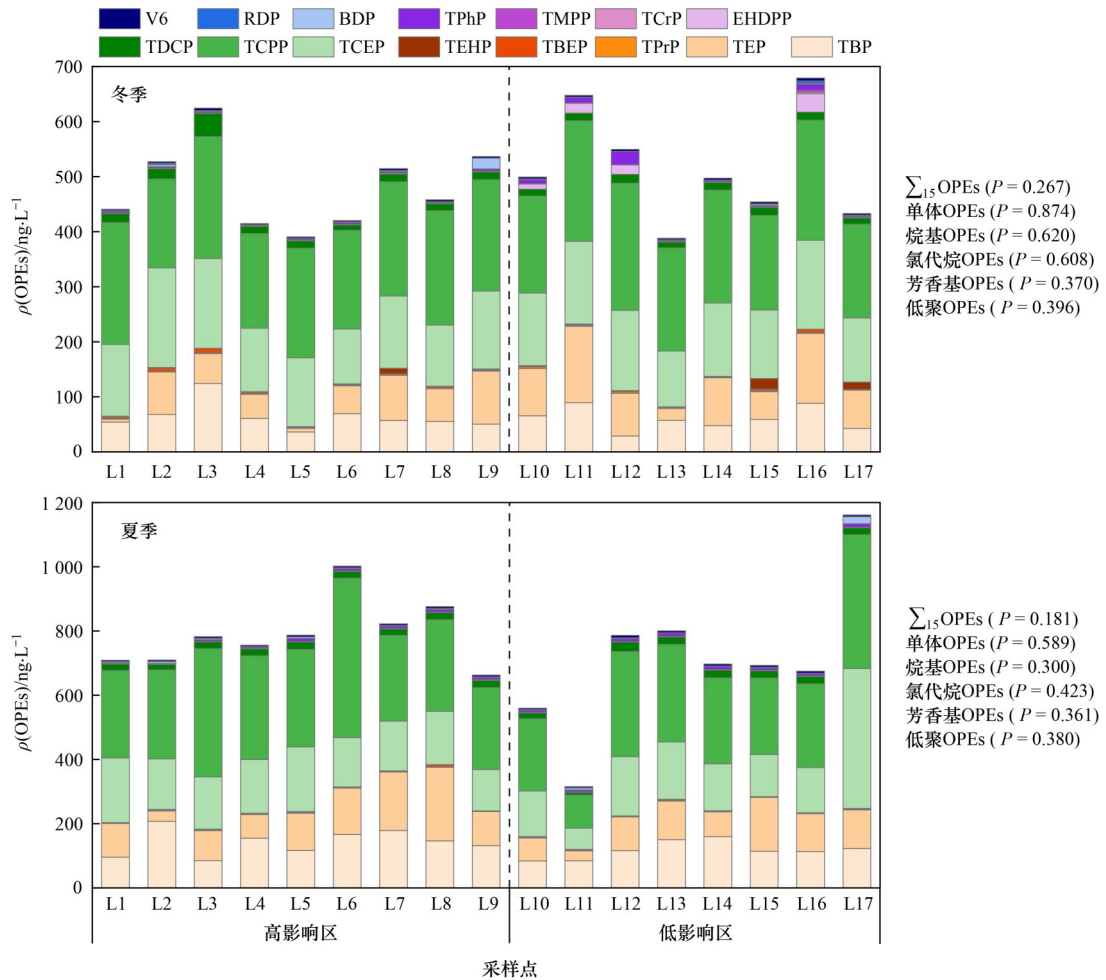


图2 太湖水体冬季和夏季各采样点 OPEs 的浓度

Fig. 2 Concentrations of OPEs in the water of Taihu Lake at each sampling location in winter and summer

研究结果发现氯代烷基 OPEs 的含量最高,与本研究的发现一致.曹渺等^[17]针对黄河流域 14 种单体 OPEs 开展研究,其中赋存浓度最高的也为氯代烷基 OPEs.在中国胶州湾^[7]和日本东京湾^[35]同样发现氯代烷基 OPEs 的浓度最高.上述现象表明,氯代烷基 OPEs 可能是目前地表水体中占比最大的 OPEs,这可能与氯代烷基的产量大和应用广泛相关,加之氯代烷基在环境中的半衰期较长(8.6 ~ 21.3 d),且不易被降解^[36],从而导致了氯代烷基 OPEs 在水体中的高赋存现象. OPEs 在夏季太湖水体中的浓度显著高于冬季,这一现象在李栋等^[18]和 Shi 等^[37]的研究中也有类似发现,该现象可能与气候条件有密切关系.有学者发现强光照和高温更易使 OPEs 扩散至大气中,进而在降雨和大气沉降的作用下汇集至河流湖泊中^[38].太湖处于亚热带地区,夏季光照强度大、温度高, OPEs 较容易从环境中被释放至大气和灰尘中,加之夏季雨量充沛, OPEs 易通过降雨作用及地表径流(或入湖河流)进入湖泊中富集,因此可能导致了夏季太湖水体中 OPEs 的高赋存.

表4 太湖水体冬夏两季不同类型 OPEs 的显著性差异分析¹⁾

Table 4 Significant difference analysis for the concentrations of OPE in the water of Taihu Lake between the winter and summer

类型	冬季浓度平均值/ $\text{ng}\cdot\text{L}^{-1}$	夏季浓度平均值/ $\text{ng}\cdot\text{L}^{-1}$	P 值
$\sum_{15}\text{OPEs}$	498.54	752.74	0.000
单体 OPEs	492.61	743.51	0.000
烷基 OPEs	135.86	246.54	0.000
氯代烷基 OPEs	345.49	488.06	0.001
芳香基 OPEs	11.27	8.91	0.525
低聚 OPEs	5.93	9.23	0.570

1) P 值 < 0.05 表示存在显著性差异

2.2 太湖沉积物中 OPEs 时空分布特征

不同季节太湖沉积物中 OPEs 在各采样点的含量分布如图 4 所示.冬季除 V6 未检出外,其余 14 种 OPEs 均有检出,其中 TPrP、TEHP 和 RDP 的检出率低于 100%,分别为 17.7%、76.5% 和 52.9%,其余 11 种 OPEs 的检出率皆为 100%.夏季沉积物中 OPEs 均被检出,除了 TPrP、RDP 和 V6 之外(检出率分别为 23.5%、94.1% 和 5.9%),其余 12 种 OPEs 的检出率皆为 100%.与文献报道数据相比较,沉积物中 OPEs

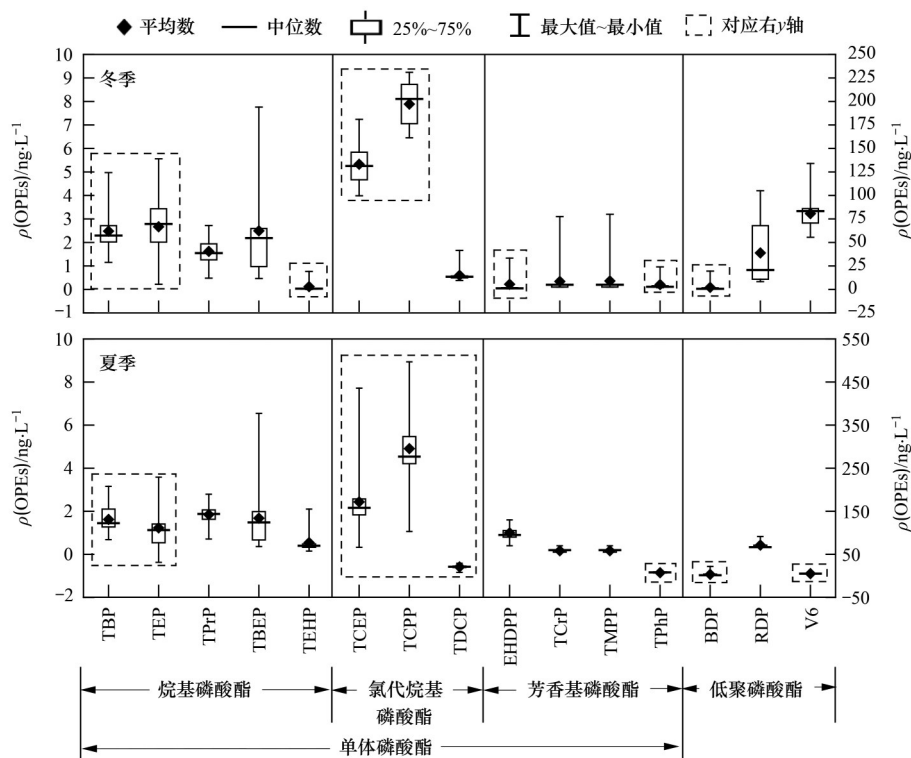


图3 太湖水体中不同类型OPEs在冬季和夏季的赋存浓度

Fig. 3 Concentrations of OPEs in the water of Taihu Lake in winter and summer

的检出率较2014年^[10]和2019年^[39]均有提高。

在空间分布上, $\omega(\sum_{15} \text{OPEs})$ 在高影响区的范围(以dry weight, dw计)为45.30 ~ 117.00 ng·g⁻¹(冬)和69.70 ~ 227.40 ng·g⁻¹(夏),在低影响区为47.80 ~ 104.80 ng·g⁻¹(冬)和80.50 ~ 176.40 ng·g⁻¹(夏)。对不同区域的OPEs进行显著性差异分析发现,所有OPEs在区域间不存在明显差异($P > 0.05$),并且不同类型OPEs在高、低影响区之间也没有差异性($P > 0.05$),这与水体的规律基本一致。严小菊^[40]和张文萍^[41]的研究结果表明太湖沉积物中OPEs不存在显著区域分布特征,与本研究的结果一致。

太湖沉积物中OPEs的赋存规律与水体基本一致,即:氯代烷基OPEs > 烷基OPEs > 芳香基OPEs ≈ 低聚OPEs,具体情况如图5所示。在冬季,氯代烷基OPEs中TCEP和TCPP含量较高,范围分别为5.10 ~ 46.40 ng·g⁻¹和19.20 ~ 36.50 ng·g⁻¹;烷基OPEs中TBP含量较高,为7.10 ~ 18.30 ng·g⁻¹;芳香基OPEs和低聚OPEs虽然整体含量均较低,但值得注意的是 $\omega(\text{BDP})$ 的最大值可达50.10 ng·g⁻¹。在夏季,太湖沉积物中 $\sum_{15} \text{OPEs}$ 含量显著高于冬季,其中单体OPEs尤为明显。另外,虽然低聚OPEs在沉积物中的含量不存在明显的季节差异($P = 0.074$)但其在夏季的含量更高(表5)。夏季含量较高的OPEs包括TBP(2.30 ~ 49.30 ng·g⁻¹)、TCEP(1.80 ~ 119.10 ng·g⁻¹)、TCPP

(19.80 ~ 84.80 ng·g⁻¹)、TPhP(7.00 ~ 16.70 ng·g⁻¹)和BDP(1.10 ~ 75.20 ng·g⁻¹)。地表水环境的沉积物和水体之间存在污染物的吸附和解吸过程^[38],有研究报道沉积物对污染物的吸附能力与水体的pH和水温呈正相关关系^[42]。本研究现场监测的水体pH在冬夏两季分别为7.90和9.03,水温在冬夏两季分别为10.38℃和30.64℃因此推测太湖沉积物中OPEs在夏季的高赋存可能与水体的pH以及水温有关。

基于太湖沉积物中各OPEs的中位含量可知,TBP、TCPP和TCEP是太湖沉积物赋存的主要OPEs,与长江^[20]和胶州湾^[7]沉积物中的主要OPEs一致。TCEP和TCPP是典型的两种氯代烷基OPEs,其产量大、应用广泛、半衰期长和不易降解,因而较容易富集在地表水环境中^[36]。TBP作为一种添加型阻燃剂,除常用于阻燃剂和增塑剂外,同时还被用在润滑油和液压油中^[43],较容易通过挥发作用释放至环境中^[44]。此外,虽然低聚OPEs中BDP的含量低于上述3种单体OPEs,但其在沉积物中的含量不容忽视(含量最大值可达75.20 ng·g⁻¹),Zhao等^[10]也有类似的发现。鉴于BDP的高产量(全国OPEs产量第4)^[3]及其潜在代谢产物双酚A^[45]的风险,该污染物同样需要引起高度重视。

2.3 OPEs在水体和沉积物中的分配

为了解OPEs在水体和沉积物中的分配情况,本研究对冬夏两季OPEs的伪分配系数[pseudo-

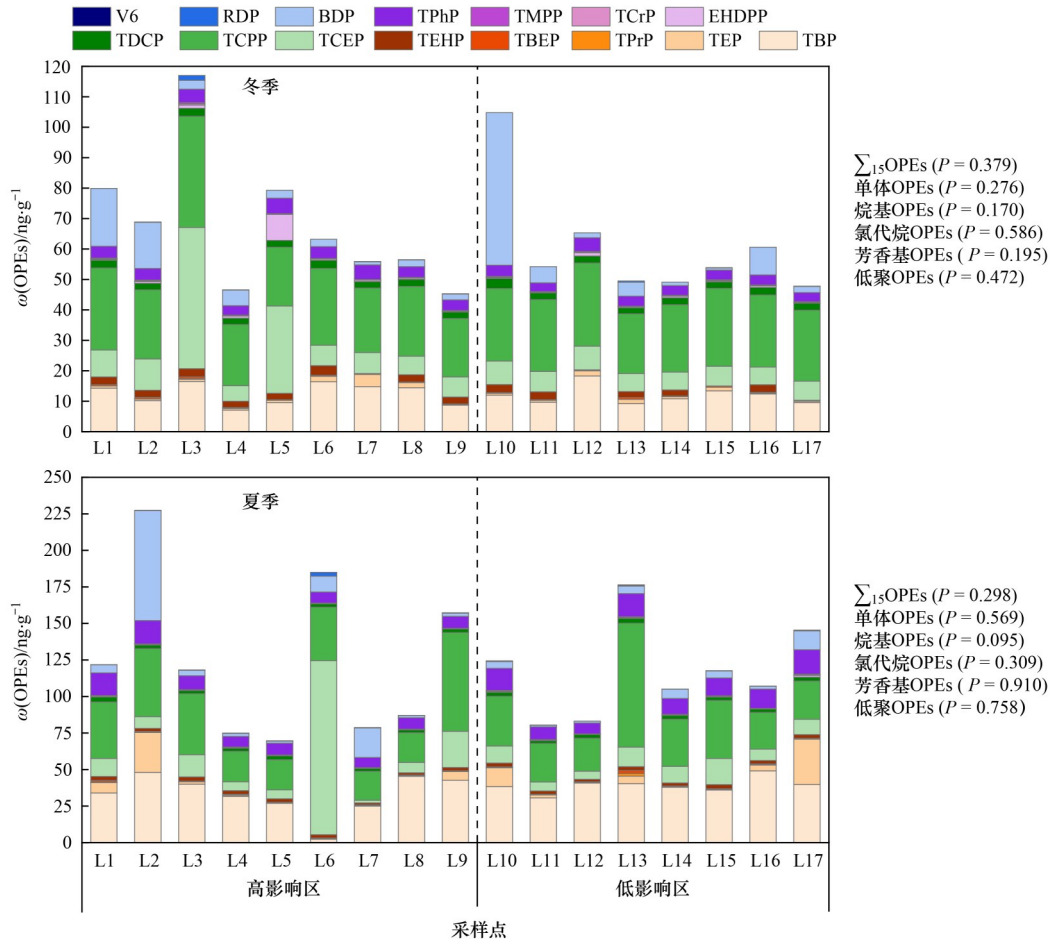


图4 太湖沉积物冬季和夏季各采样点OPEs的含量

Fig. 4 Concentrations of OPEs in the sediment of Taihu Lake at each sampling location in winter and summer

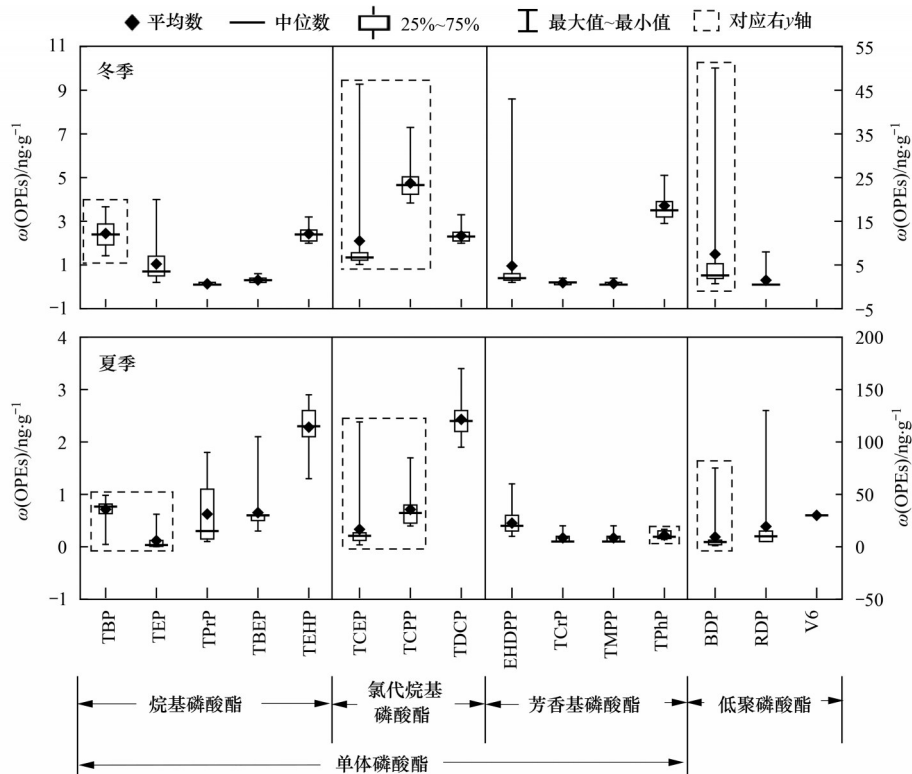


图5 太湖沉积物中不同类型OPEs在冬季和夏季的赋存浓度

Fig. 5 Concentrations of OPEs in the sediment of Taihu Lake in winter and summer

表5 太湖沉积物冬夏两季不同类型OPEs的显著性差异分析¹⁾
Table 5 Significant difference analysis for the concentrations of OPE in the sediment of Taihu Lake between the winter and summer

类型	冬季含量平均值/ $\text{ng}\cdot\text{g}^{-1}$	夏季含量平均值/ $\text{ng}\cdot\text{g}^{-1}$	P 值
$\sum_{15}$ OPEs	54.53	124.01	0.000
单体 OPEs	49.23	110.26	0.000
烷基 OPEs	15.42	48.71	0.000
氯代烷基 OPEs	29.86	46.50	0.000
芳香基 OPEs	3.95	15.04	0.000
低聚 OPEs	5.30	13.76	0.074

1) P 值 < 0.05 表示存在显著性差异

partitioning coefficient, K_d = 沉积物中污染物含量($\text{ng}\cdot\text{g}^{-1}$)/水体中污染物浓度($\text{ng}\cdot\text{L}^{-1}$)]^[20]进行了分析(表6). 由表6可知,OPEs在冬季的 K_d 值($\text{L}\cdot\text{g}^{-1}$,下同)范围为0.02(TEP)~3.47(BDP),在夏季的 K_d 值范围为0.05(TEP)~4.17(TEHP),BDP和TEHP的 K_d 值分别在冬夏两季的所有OPEs中最高,表明这两种OPEs较容易被沉积物吸附,这与其在沉积物中的赋存情况相符(图5). 在冬夏两季,15种目标OPEs的 K_d 值和 $\lg K_{ow}$ 皆呈现显著的正相关关系(表6),表明 $\lg K_{ow}$ 越高的OPEs较容易被沉积物吸附,因而亲脂性高的BDP和TEHP在沉积物中呈现出较高的含量. 由此可知,OPEs的物化性质可能是影响其在水体和沉积物之间分配的主要因素之一.

此外, $\sum_{15}$ OPEs在夏季的 K_d 值($0.87\text{ L}\cdot\text{g}^{-1}$)是冬季的1.74倍,表明OPEs在夏季更容易富集在沉积物中. 虽然在夏季沉积物更容易吸附水体中的OPEs,但是水体中的OPEs在夏季明显高于冬季,主要原因可能是在这个季节水体污染物的输入量更大,沉积物吸附的量无法抵消输入量的影响,因而虽然沉积物吸附能力在夏季增强,但整体上水体中的OPEs浓度还是呈现增大趋势.

表6 OPEs在太湖沉积物和水体间的伪分配系数(K_d)及与 $\lg K_{ow}$ 的相关性¹⁾

Table 6 Pseudo-partitioning coefficients of OPEs between the sediment and surface water of Taihu Lake and their correlations with $\lg K_{ow}$

类型	$K_d/\text{L}\cdot\text{g}^{-1}$		与 $\lg K_{ow}$ 的相关性	
	冬季	夏季	冬季	夏季
TBP	0.20	0.27		
TEP	0.02	0.05		
TPrP	0.06	0.34		
TBEP	0.13	0.39		
TEHP	0.80	4.17		
TCEP	0.08	0.10		
TCPP	0.12	0.12		
TDCP	0.16	0.12	$P = 0.000$	$P = 0.000$
EHDPP	0.18	0.45	$R^2 = 0.36$	$R^2 = 0.72$
TCrP	0.53	1.04		
TMPP	0.41	1.00		
TPhP	0.72	1.45		
BDP	3.47	2.60		
RDP	0.19	0.90		
V6	—	0.11		
$\sum_{15}$ OPEs	0.50	0.87		

1) P 值 < 0.05 表示存在显著性差异,“—”表示无有效数据

季($0.50\text{ L}\cdot\text{g}^{-1}$)的1.74倍,表明OPEs在夏季更容易富集在沉积物中. 虽然在夏季沉积物更容易吸附水体中的OPEs,但是水体中的OPEs在夏季明显高于冬季,主要原因可能是在这个季节水体污染物的输入量更大,沉积物吸附的量无法抵消输入量的影响,因而虽然沉积物吸附能力在夏季增强,但整体上水体中的OPEs浓度还是呈现增大趋势.

2.4 太湖水环境OPEs的生态风险评估

鉴于太湖水体具有水产养殖、饮用水供给和农业灌溉等重要功能,本研究重点针对水体中OPEs的赋存开展太湖水环境生态风险评估,但这并不表明太湖沉积物中的OPEs不存在生态风险,有关内容需要进一步开展研究. 根据太湖水体中OPEs的浓度计算所得的风险商值如图6所示,整体上太湖水体中的 $\sum_{15}$ OPEs具有中等生态风险(冬季 $RQ = 0.34$;夏季 $RQ = 0.35$),其中单体OPEs的生态风险中等,低聚OPEs的生态风险较低(冬季 $RQ = 0.08$;夏季 $RQ = 0.04$). 在单体OPEs中,氯代烷基OPEs的生态风险最高,为中等程度,而烷基OPEs和芳香基OPEs为低生态风险. 张文萍等^[25]和吕佳佩等^[46]也对太湖水体中的OPEs进行了生态风险评估,发现氯代烷基OPEs为低生态风险,低于本研究的评估结果,预示着OPEs的风险可能正在逐年加剧.

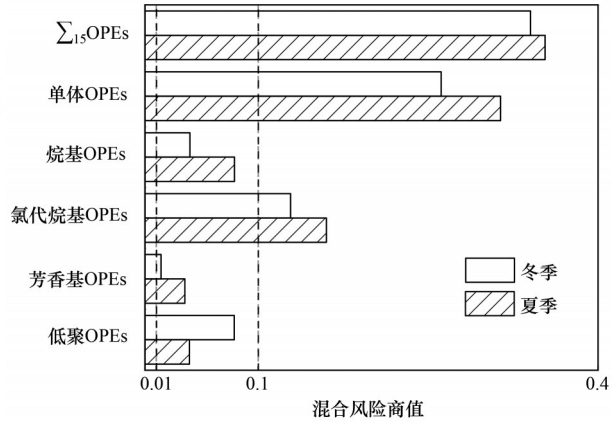


图6 不同季节太湖水环境OPEs的风险商值
Fig. 6 Risk quotient values of OPEs in the water environment of Taihu Lake in different seasons

除低聚OPEs之外,OPEs在夏季的生态风险皆高于冬季,并且冬夏两季OPEs的生态风险排序整体上皆呈现为:氯代烷基OPEs > 烷基OPEs > 芳香基OPEs ≈ 低聚OPEs,这与OPEs在水体中的赋存规律基本一致,表明OPEs的浓度是影响OPEs生态风险的一个重要因素. 单体OPEs中的氯代烷基OPEs是目前开展毒理研究较多的一类OPEs^[47],已有研究发现其会对鱼类的生长发育产生危害^[48],并且还具有神经毒性^[49]、基因和细胞毒性^[50],因而地表水体中的氯代

烷基 OPEs 亟需探索相关的控制措施. 低聚 OPEs 在本研究中的生态风险较低,但并不表明此类 OPEs 不需要引起重视,目前与其有关的毒理研究非常有限,需要深入开展相关研究以为生态风险评估提供更多的数据支持.

3 结论

(1)太湖水体中的 $\sum_{15}$ OPEs 在高、低影响区之间的赋存浓度皆不存在显著性差异. 水体中不同类型的 OPEs 在夏季的赋存浓度均高于冬季,其中单体 OPEs 的季节差异性显著. OPEs 在太湖水体中的赋存浓度排序为:氯代烷基 OPEs > 烷基 OPEs > 芳香基 OPEs ≈ 低聚 OPEs.

(2)太湖沉积物中的 $\sum_{15}$ OPEs 在高、低影响区之间的赋存含量无明显差异. 沉积物中的单体 OPEs 在冬夏两季差异明显且在夏季含量更高,虽然低聚 OPEs 不存在显著性季节差异,但在夏季的赋存含量更高. OPEs 在太湖沉积物中的赋存含量排序与水体基本一致.

(3)OPEs 在水体和沉积物中的伪分配系数(K_d) 在夏季更高,表明 OPEs 在夏季更容易被沉积物吸附,但不能忽略夏季污染物排放通量的影响. OPEs 的 K_d 值与其 $\lg K_{ow}$ 显著正相关,表明亲脂性较高的 OPEs 更容易被沉积物吸附.

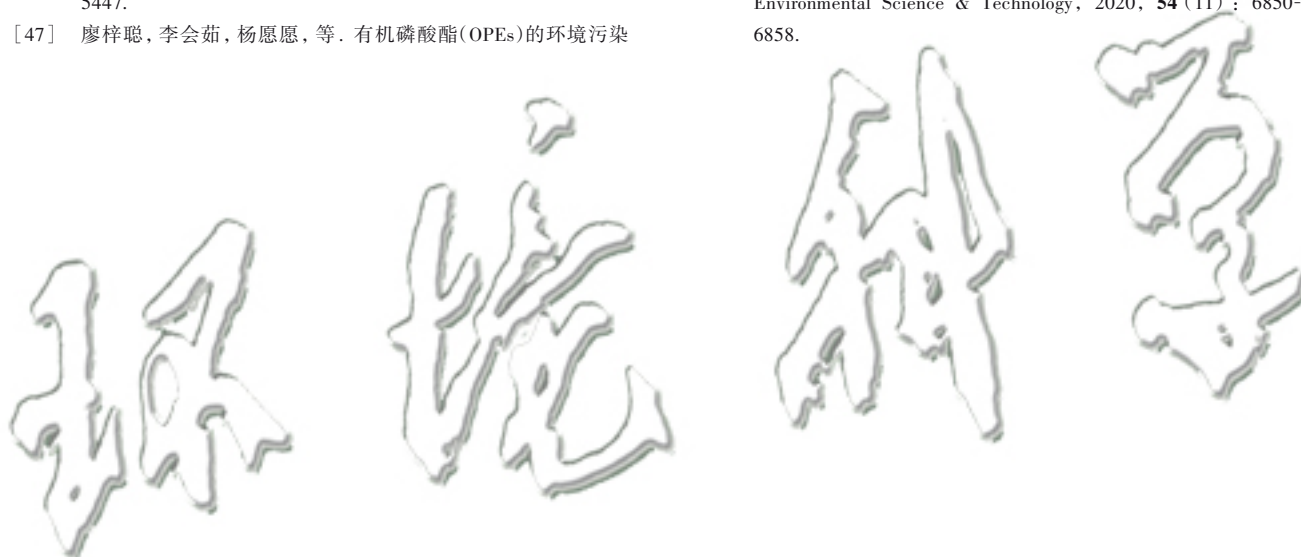
(4)风险商值结果表明 $\sum_{15}$ OPEs 具有中等生态风险,不同类型的单体 OPEs 在夏季的生态风险皆高于冬季. 整体上冬夏两季 OPEs 的生态风险排序为:氯代烷基 OPEs > 烷基 OPEs > 芳香基 OPEs ≈ 低聚 OPEs. 低聚 OPEs 的生态风险虽然较低,但因其相关毒理数据的匮乏亟需开展深入研究.

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CONTENTS

Prediction of Autumn Ozone Concentration in the Pearl River Delta Based on Machine Learning	CHEN Zhen, LIU Run, LUO Zheng, <i>et al.</i> (1)
Remote Sensing Model for Estimating Atmospheric PM _{2.5} Concentration in the Guangdong-Hong Kong-Macao Greater Bay Area	DAI Yuan-yuan, GONG Shao-qi, ZHANG Cun-jie, <i>et al.</i> (8)
Variation Characteristics of PM _{2.5} Pollution and Transport in Typical Transport Channel Cities in Winter	DAI Wu-jun, ZHOU Ying, WANG Xiao-qi, <i>et al.</i> (23)
Characteristics of Secondary Inorganic Ions in PM _{2.5} and Its Influencing Factors in Summer in Zhengzhou	HE Bing, YANG Jie-ru, XU Yi-fei, <i>et al.</i> (36)
Characteristics and Source Apportionment of Carbonaceous Aerosols in the Typical Urban Areas in Chongqing During Winter	PENG Chao, LI Zhen-liang, XIANG Ying, <i>et al.</i> (48)
Analysis of Influencing Factors of Ozone Pollution Difference Between Chengdu and Chongqing in August 2022	CHEN Mu-lan, LI Zhen-liang, PENG Chao, <i>et al.</i> (61)
Analysis of O ₃ Pollution Affected by a Succession of Three Landfall Typhoons in 2020 in Eastern China	HUA Cong, YOU Yuan, WANG Qian, <i>et al.</i> (71)
Characteristics and Source Apportionment of VOCs Initial Mixing Ratio in Beijing During Summer	ZHANG Bo-tao, JING Kuan, WANG Qin, <i>et al.</i> (81)
Review of Comprehensive Evaluation System of Vehicle Pollution and Carbon Synergistic Reduction	FAN Zhao-yang, TONG Hui, LIANG Xiao-yu, <i>et al.</i> (93)
Study of Peak Carbon Emission of a City in Yangtze River Delta Based on LEAP Model	YANG Feng, ZHANG Gui-chi, SUN Ji, <i>et al.</i> (104)
Driving Forces and Mitigation Potential of CO ₂ Emissions for Ship Transportation in Guangdong Province, China	WENG Shu-juan, LIU Ying-ying, TANG Feng, <i>et al.</i> (115)
Carbon Emission Characteristics and Influencing Factors of Typical Processes in Drinking Water Treatment Plant	ZHANG Xiang-yu, HU Jian-kun, MA Kai, <i>et al.</i> (123)
Distribution Characteristics of Arsenic in Drinking Water in China and Its Health Risk Based on Disability-adjusted Life Years	DOU Dian-cheng, QI Rong, XIAO Shu-min, <i>et al.</i> (131)
Spatiotemporal Occurrence of Organophosphate Esters in the Surface Water and Sediment of Taihu Lake and Relevant Risk Assessment	ZHANG Cheng-nuo, ZHONG Qin, LUAN Bo-wen, <i>et al.</i> (140)
Exposure Level and Risk Impact Assessment of Pesticides and Veterinary Drugs in Aquaculture Environment	ZHANG Kai-wen, ZHANG Hai-yan, KONG Cong, <i>et al.</i> (151)
Variation in Phosphorus Concentration and Flux at Zhutuo Section in the Yangtze River and Source Apportionment	LOU Bao-feng, XIE Wei-min, HUANG Bo, <i>et al.</i> (159)
“Load-Unload” Effect of Manganese Oxides on Phosphorus in Surface Water of the Pearl River Estuary	LI Rui, LIANG Zuo-bing, WU Qi-rui, <i>et al.</i> (173)
Factors Influencing the Variation in Phytoplankton Functional Groups in Fuchunjiang Reservoir	ZHANG Ping, WANG Wei, ZHU Meng-yuan, <i>et al.</i> (181)
Hydrochemical Characteristics and Formation Mechanism of Groundwater in the Western Region of Hepu Basin, Beihai City	CHEN Wen, WU Ya, ZHANG Hong-xin, <i>et al.</i> (194)
Controlling Factors of Groundwater Salinization and Pollution in the Oasis Zone of the Cherchen River Basin of Xinjiang	LI Jun, OUYANG Hong-tao, ZHOU Jin-long (207)
Spatial-temporal Evolution of Ecosystem Health and Its Influencing Factors in Beijing-Tianjin-Hebei Region	LI Kui-ming, WANG Xiao-yan, YAO Luo-lan (218)
Spatial and Temporal Evolution and Impact Factors Analysis of Ecosystem Service Value in the Liaohe River Delta over the Past 30 Years	WANG Geng, ZHANG Fu-rong (228)
Effects of Photovoltaic Power Station Construction on Terrestrial Environment: Retrospect and Prospect	TIAN Zheng-qing, ZHANG Yong, LIU Xiang, <i>et al.</i> (239)
Spatiotemporal Evolution and Quantitative Attribution Analysis of Vegetation NDVI in Greater Khingan Mountains Forest-Steppe Ecotone	SHI Song, LI Wen, QU Chen, <i>et al.</i> (248)
Spatio-temporal Variation in Net Primary Productivity of Different Vegetation Types and Its Influencing Factors Exploration in Southwest China	XU Yong, ZHENG Zhi-wei, MENG Yu-chi, <i>et al.</i> (262)
Impacts of Extreme Climate Events at Different Altitudinal Gradients on Vegetation NPP in Songhua River Basin	CUI Song, JIA Zhao-yang, GUO Liang, <i>et al.</i> (275)
Spatial and Temporal Evolution and Prediction of Carbon Storage in Kunming City Based on InVEST and CA-Markov Model	Paruke Wusimanjiang, AI Dong, FANG Yi-shu, <i>et al.</i> (287)
Spatial-Temporal Evolution and Prediction of Carbon Storage in Jiuquan City Ecosystem Based on PLUS-InVEST Model	SHI Jing, SHI Pei-ji, WANG Zi-yang, <i>et al.</i> (300)
Soil Carbon Pool Allocation Dynamics During Soil Development in the Lower Yangtze River Alluvial Plain	HU Dan-yang, ZHANG Huan, SU Bao-wei, <i>et al.</i> (314)
Spatial Distribution Patterns of Soil Organic Carbon in Karst Forests of the Lijiang River Basin and Its Driving Factors	SHEN Kai-hui, WEI Shi-guang, LI Lin, <i>et al.</i> (323)
Effect of Land Use on the Stability of Soil Organic Carbon in a Karst Region	CHEN Jian-qi, JIA Ya-nan, HE Qiu-fang, <i>et al.</i> (335)
Spatial Distribution Characteristics of Soil Carbon and Nitrogen in Citrus Orchards on the Slope of Purple Soil Hilly Area	LI Zi-yang, CHEN Lu, ZHAO Peng, <i>et al.</i> (343)
Effects of Experimental Nitrogen Deposition and Litter Manipulation on Soil Organic Components and Enzyme Activity of Latosol in Tropical Rubber Plantations	XUE Xin-xin, REN Chang-qi, LUO Xue-hua, <i>et al.</i> (354)
Analysis on Driving Factors, Reduction Potential, and Environmental Effect of Inorganic Fertilizer Input in Chongqing	LIANG Tao, ZHAO Jing-kun, LI Hong-mei, <i>et al.</i> (364)
Research Progress on Distribution, Transportation, and Control of Per- and Polyfluoroalkyl Substances in Chinese Soils	LIU Hao-ran, XING Jing-yi, REN Wen-jie (376)
Prediction of Spatial Distribution of Heavy Metals in Cultivated Soil Based on Multi-source Auxiliary Variables and Random Forest Model	XIE Xue-feng, GUO Wei-wei, PU Li-jie, <i>et al.</i> (386)
Health Risk Assessment and Priority Control Factors Analysis of Heavy Metals in Agricultural Soils Based on Source-oriented	MA Jie, GE Miao, WANG Sheng-lan, <i>et al.</i> (396)
Contamination Characteristics and Source Apportionment of Soil Heavy Metals in an Abandoned Pyrite Mining Area of Tongling City, China	LI Ru-zhong, LIU Yu-hao, HUANG Yan-huan, <i>et al.</i> (407)
Source Appointment and Assessment of Heavy Metal Pollution in Surface Dust in the Main District Bus Stops of Tianshui City	LI Chun-yan, WANG Xin-min, WANG Hai, <i>et al.</i> (417)
Response of Cadmium in Soil-rice to Different Conditioners Based on Field Trials	TANG Le-bin, LIU Xin-cai, SONG Bo, <i>et al.</i> (429)
Regulation Effects of Humus Active Components on Soil Cadmium Availability and Critical Threshold for Rice Safety	HU Xiu-zhi, SONG Yi, WANG Tian-yu, <i>et al.</i> (439)
Using Biochar and Iron-calcium Material to Remediate Paddy Soil Contaminated by Cadmium and Arsenic	WU Qiu-chan, WU Ji-zi, ZHAO Ke-li, <i>et al.</i> (450)
Research Progress on Characteristics of Human Microplastic Pollution and Health Risks	MA Min-dong, ZHAO Yang-chen, ZHU Long, <i>et al.</i> (459)
Effects of Polystyrene Microplastics Combined with Cadmium Contamination on Soil Physicochemical Properties and Physiological Ecology of <i>Lactuca sativa</i>	NIU Jia-rui, ZOU Yong-jun, JIAN Min-fei, <i>et al.</i> (470)
Transcriptome Analysis of Plant Growth-promoting Bacteria Alleviating Microplastic and Heavy Metal Combined Pollution Stress in Sorghum	LIU Yong-qi, ZHAO Si-yu, REN Xue-min, <i>et al.</i> (480)
Effects of Microplastics on the Leaching of Nutrients and Cadmium from Soil	ZHAO Qun-fang, CHU Long-wei, DING Yuan-hong, <i>et al.</i> (489)
Effect of Microplastics and Phenanthrene on Soil Chemical Properties, Enzymatic Activities, and Microbial Communities	LIU Sha-sha, QIN Jian-qiao, WU Xian-ge (496)
Prediction of Soil Bacterial Community Structure and Function in Minqin Desert-oasis Ecotone Artificial <i>Haloxylon ammodendron</i> Forest	WANG An-lin, MA Rui, MA Yan-jun, <i>et al.</i> (508)
Response of Soil Fungal Community to Biochar Application Under Different Irrigation Water Salinity	ZHENG Zhi-yu, GUO Xiao-wen, MIN Wei (520)
Effects of Organic Fertilizer of Kitchen Waste on Soil Microbial Activity and Function	LIU Mei-ling, WANG Yi-min, JIN Wen-hao, <i>et al.</i> (530)
Response Characteristics of Soil Fungal Community Structure to Long-term Continuous Cropping of Pepper	CHEN Fen, YU Gao, WANG Xie-feng, <i>et al.</i> (543)
Effects of Foliar Application of Silicon Fertilizers on Phyllosphere Bacterial Community and Functional Genes of Paddy Irrigated with Reclaimed Water	LIANG Sheng-xian, LIU Chun-cheng, HU Chao, <i>et al.</i> (555)
Analysis of Bacterial Communities and Antibiotic Resistance Genes in the Aquaculture Area of Changli County	WANG Qiu-shui, CHENG Bo, LIU Yue, <i>et al.</i> (567)
High-throughput qPCR and Amplicon Sequencing as Complementary Methods for Profiling Antibiotic Resistance Genes in Urban Wetland Parks	HUANG Fu-yi, ZHOU Shu-yi-dan, PAN Ting, <i>et al.</i> (576)
Characteristics of Vertical Distribution and Environmental Factors of Antibiotics in Quaternary Sedimentary Column in Urban Areas	LIU Ke, TONG Lei, GAN Cui, <i>et al.</i> (584)
Adsorption Performance and Mechanism of Oxytetracycline in Water by KOH Modified Biochar Derived from Corn Straw	LIU Zong-tang, SUN Yu-feng, FEI Zheng-hao, <i>et al.</i> (594)
Comparison of Pb ²⁺ Adsorption Properties of Biochars Modified Through CO ₂ Atmosphere Pyrolysis and Nitric Acid	JIANG Hao, CHEN Rui-zhi, ZHU Zi-yang, <i>et al.</i> (606)