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Song Z H, Ding Y, Li J, et al. Sedimentary records of emerging contaminants in alpine lakes worldwide: A meta-analysis [J]. China Environmental Science, 2025,45(9):5190−5203.

全球高山湖泊中新污染物的历史沉积记录研究

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摘要：为探索全球高山湖泊沉积物中新污染物的沉积历史、来源机制与生态风险,系统整合 42 篇文献中 2026 条新污染物监测数据,采用统一单位与浓度归一化的处理方法,以 5 年时间步长构建年代序列进行趋势分析.结果表明,全球高山湖泊沉积物中持久性有机污染物(POPs)总体处于较低水平,其中多环芳烃(PAHs)、滴滴涕(DDTs)、六六六(HCHs)、多氯联苯(PCBs)和多溴二苯醚(PBDEs)浓度范围分别为:0.160~2140ng/g、0.001~12.9ng/g、1.30~372ng/g、0.001~12.9ng/g 和 0.119~2.50ng/g.PAHs 呈持续上升趋势,DDTs 和 HCHs 呈现 20 世纪 80 年代和 21 世纪初两个沉积峰值,反映了人类活动产生的“一次源”排放及全球变暖驱动下的“二次源”释放.微塑料丰度(19~2640 个/kg)整体较低但呈持续增长趋势,也与人类活动及气候变化相关.区域人类活动与全球气候变暖是影响高山地区新污染物沉积的主要因素.PAHs 的毒性当量持续上升,生态风险不断增加;微塑料丰度较低但高危聚合物比例高,生态风险不容忽视.然而,当前与微塑料相关的沉积年代学研究较为缺乏,高山湖泊沉积物中新污染物的不可提取态残留物(NER)研究相对较少,内分泌干扰物和抗生素相关报道极为有限.未来亟待推进非洲高原等数据匮乏区域新污染物调查、加强共迁移物质等多种类新污染物协同研究.

关键词：高山湖泊；青藏高原；沉积物；新污染物；持久性有机污染物；微塑料；来源；风险
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Sedimentary records of emerging contaminants in alpine lakes worldwide: A meta-analysis. SONG Zi-hao[1], DING Yang[1,2*], LI Jun[3], TANG Xiao-yan[1], LIU Wei-jie[2], BAO Qian[1], SUN Zhe[1], XING Xin-li[2], QI Shi-hua[2] (1.Key Laboratory of Land Resources Evaluation and Monitoring in Southwest, Ministry of Education, College of Geography and Resources Science, Sichuan Normal University, Chengdu 610066；2.State Key Laboratory of Biogeology and Environmental Geology, School of Environmental Studies, China University of Geosciences, Wuhan 430078；3.School of Earth Sciences and Resources, China University of Geosciences, Beijing 100083). *China Environmental Science*, 2025,45(9)：5190~5203

Abstract：To explore the sedimentary history, sources, and ecological risks of emerging contaminants in global alpine lake sediments, 2,026 monitoring data points from 42 published studies were systematically integrated. Unit standardization and concentration normalization were applied, and temporal sequences with 5-year intervals were constructed for trend analysis. The results showed that persistent organic pollutants (POPs) in global alpine lake sediments stayed at generally low levels, with concentrations of 0.160~2140, 0.001~12.9, 1.30~372, 0.001~12.9, and 0.119~2.50 ng/g for PAHs, DDTs, HCHs, PCBs, and PBDEs, respectively. Concentrations of PAHs showed a continuous rising trend, whereas concentrations of DDTs and HCHs exhibited two peaks in the 1980s and early 2000s, reflecting "primary source" emissions from human activities and "secondary source" releases driven by global warming. Microplastic abundance (19~2640 items/kg) remained relatively low overall but displayed an increasing trend, also related to human activities and climate change. The toxic equivalents of PAHs continues to rise, leading to increasing ecological risks; while microplastics exhibit relatively low abundance but high proportion of hazardous polymers, indicating non-negligible ecological threats. However, current research on microplastic chronological sequence in alpine lake sediments remains scarce; studies of non-extractable residues (NER) of emerging contaminants in alpine lake sediments are relatively limited; and reports on endocrine-disrupting compounds and antibiotics are extremely rare. Investigations on emerging contaminants in data-scarce regions such as the African plateau and research on multiple categories of emerging contaminants especially co-migrating substances are both needed in the future.

Key words：alpine lakes；Tibetan Plateau；sediments；new pollutants/emerging contaminants；persistent organic pollutants (POPs)；microplastics；source；risk

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新污染物是指新发现的、合成或天然存在的,能在环境中检测到并对人类和生态系统具有潜在危害,但尚未纳入管理或现有管理措施不足的有毒有害化学物质,主要包括持久性有机污染物(POPs)、微塑料、内分泌干扰物和抗生素等[1].新污染物来源广泛、种类繁多,其在环境中的赋存与迁移转化规律复杂多变,给环境监测、风险评估和污染治理带来了前所未有的挑战[2].目前新污染物污染已成为全球及我国亟需应对的重大环境问题之一.

高山湖泊生态环境脆弱,受人类活动干扰少,成为污染物的重要储存库和环境变化信息的重要档案库[3].“高山冷阱效应”(Mountain cold-trapping)使得高山地区成为全球 POPs 循环的主要聚集地之一[4],并最终通过长距离迁移、干湿沉降及河流输送等方式进入高山湖泊环境中[5];大气和河流输送也是微塑料在偏远高山湖泊地区富集的重要途径[6].高山湖泊因而成为研究新污染物污染历史及其对环境变化响应的理想场所[5].未受扰动的高山湖泊沉积物岩心较好记录了历史上的新污染物污染过程及排放差异[3,7].因此研究全球高山湖泊沉积物中新污染物的沉积历史对于理解不同地区新污染物的排放差异、演变规律等方面具有重要意义.

本文系统整合了全球各高山湖泊沉积物中新污染物的沉积记录,提取并解析了不同新污染物的浓度数据,探讨了浓度归一化趋势和主要来源,总结了影响高山湖泊新污染物沉积的因素,并对相关研究趋势进行展望.研究结果可为高山生态环境保护、新污染物风险防控和污染历史重建等领域提供科学依据与决策支持.

1　研究方法

1.1　文献检索及数据收集

本研究的数据主要来源于中国知网 CNKI 和 Web of Science 两个数据库,以持久性有机污染物、微塑料、内分泌干扰物、抗生素为检索对象.中文文献检索关键词:“高山湖泊”(或“偏远高山湖泊”、“高原湖泊”)、“新污染物”(或“持久性有机污染物”、“全氟化合物”、“氯化石蜡”等;“微塑料”;“抗生素”;“内分泌干扰物”、“邻苯二甲酸酯”、“双酚类”等)和“沉积物”(或“沉积柱”、“岩心”、“沉积记录”).英文文献检索关键词:“Mountain lake”或(“Remote alpine mountain lake”、“Plateau lake”)、“Emerging contaminants”(或“Persistent organic pollutants”、“Perfluorinated compounds”、“Chlorinated paraffins”等;“Microplastics”;“Antibiotics”;“Endocrine disruptors”、“Phthalates”、“Bisphenol”等)和“Sediment”(或“Sedimentation column”、“Sediment core”、“Sedimentary record”).检索年限不限.

表 1　全球高山湖泊沉积柱采样点信息及新污染物类型

Table 1　Basic information on sampling sites for alpine lake sediment cores worldwide and types of emerging contaminants investigated

采样区域	所在地区	湖泊数量(个)	海拔(m)	污染物类型	沉积柱年份	文献来源
青藏高原	亚洲	19	1500~5010	PAHs/DDTs/PCBs/HCHs/氯化石蜡/多形态 PAHs/微塑料	1860~2019	[3, 8-28]
云贵高原	亚洲	4	1720~1970	PAHs/微塑料	1910~2017	[29-31]
天山山脉	亚洲	1	1050	PAHs	1941~2010	[16]
阿尔卑斯山脉	欧洲	9	1230~2750	PAHs/DDTs/PCBs	1945~2010	[32-33]
塔特拉山脉	欧洲	3	1850~2300	DDTs/HCHs	1940~2000	[34]
比利牛斯山脉	欧洲	3	1520~1680	PCBs/DDTs	1945~2010	[35-36]
落基山脉	北美洲	3	1850~2200	PAHs/PCBs/PBDEs/全氟化合物	1863~2011	[37-38]
墨西哥高原	北美洲	3	2040~2090	PAHs/PCBs/PBDEs	1910~2010	[36, 39]
安第斯山脉	南美洲	3	1150~4320	PAHs	1886~2000	[40]
东非高原	非洲	3	1130~3000	PCBs/DDTs	1945~2005	[41-42]
南非高原	非洲	1	2400	PAHs/PCBs	1920~2017	[43]

遵循以下原则对文献进行筛选,以确保分析数据的可获得性和准确性:采样湖泊必须为高山湖泊(海拔大于 1000m);研究对象为持久性有机污染物、微塑料、内分泌干扰物、抗生素四类新污染物中的一种或多种;采集的沉积柱已进行定年处理或标有明确的采样时间.

根据内容剔除部分不相关文献后,最终筛选出有效文献 42 篇,数据 2026 条.若文献浓度数据以图件形式呈现,则利用 GetData Graph Digitizer 2.24 软件(http://getdata-graph-digitizer.com)提取相关数据.湖泊沉积柱污染物数据以 POPs 为主,主要包含多环芳烃(PAHs)、滴滴涕及其同系物(DDTs)、六六六及其异构体(HCHs)、多氯联苯(PCBs)和多溴二苯醚(PBDEs).采样点分布在五个大洲共 52 个高原湖泊,涵盖了全球大部分高山地区,所在地区及污染物类型如表 1 所示,采样点名称及具体位置如图 1 所示.微塑料、内分泌干扰物和抗生素在历史沉积记录研究中缺乏相关数据,无法详细探讨其浓度变化趋势;文中仅针对部分湖泊的表层沉积物微塑料污染浓度数据,单独讨论其变化趋势.此外,对文献中提及的沉积记录影响因素及定年手段等要素频次进行统计.

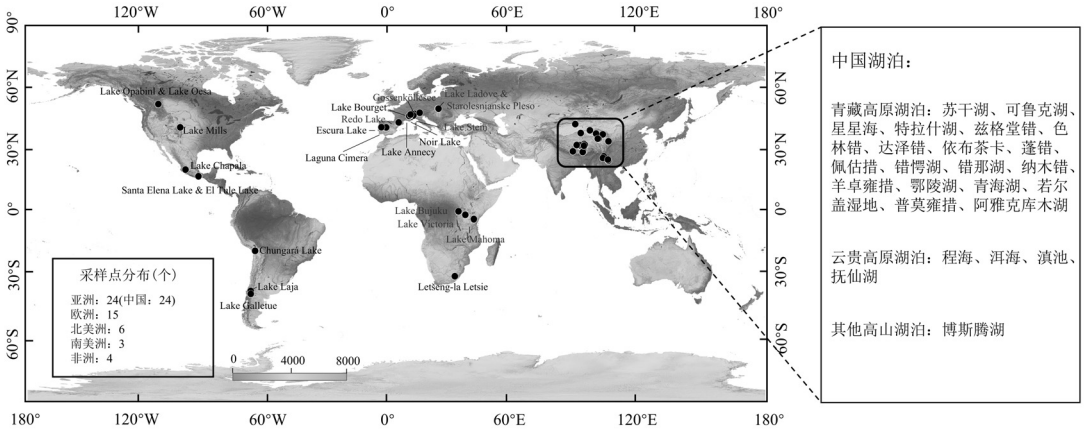


图 1 全球高山湖泊沉积柱采样点分布

Fig.1 Geographical distribution of sampling sites for alpine lake sediment cores worldwide

1.2 数据处理

将统计的 POPs 数据进行分类,浓度单位统一转换为 ng/g,“未检出”或“低于检测限”的浓度记为零值.微塑料的丰度单位统一为个/kg.为确保数据可比性,去除沉积柱 1920 年以下、2020 年以上数据,年代范围统一为 1920~2020 年.为更清晰显示其年际变化趋势,对所有 POPs 浓度数据进行归一化,将各年代浓度分别除以峰值浓度,以 5 年为时间步长分别计算全球和中国范围内的平均值和标准差[44].使用 R 4.3.3(https://cloud.r-project.org/) 进行图件绘制.

1.3 POPs 与微塑料来源解析方法

为分析污染物来源,对沉积物中 PAHs、DDTs 和 HCHs 的不同单体、异构体或同系物数据进行二次处理,分别计算苯并[a]蒽与□(BaA/(BaA+Chr))、茚并[1,2,3-c,d]芘与苯并[g,h,i]苝(IcdP/(IcdP+BghiP))、荧蒽与芘(Fla/(Fla+Pyr))、轻环与中重环多环芳烃(LMW-/HMW-PAHs)、甲体与丙体六六六(α-HCH/γ-HCH)以及滴滴涕与其好氧降解产物滴滴伊(DDT/DDE)比值,同样以 5 年为步长进行平均值和标准差的计算.微塑料的来源则依据 Wang 等[45]提出的水环境系统微塑料特定来源分类,基于聚合物的类型对沉积物中微塑料来源进行定量分析.

1.4 POPs 与微塑料生态风险评估方法

采用毒性当量(TEQ)法评价沉积物中 PAHs 的生态风险,使用相对于 BaP 的毒性当量因子(TEF)来量化沉积物 PAHs 混合物的毒性当量(TEQ_{BaP})浓度[46],并以 5 年为步长进行平均值和标准差的计算.

根据聚合物毒性和微塑料丰度两个指标,采用风险指数(H)和污染物负荷指数(PLI)对高山湖泊中微塑料的潜在风险进行评估[47].

风险指数法参考聚合物危险评分法,利用塑料聚合物危害等级模型,将不同聚合物类型的毒性作为评估微塑料污染风险的指标[28].

污染负荷指数法可反映区域内的微塑料污染状况,与微塑料丰度直接相关,可用于评估高山湖泊沉积物中微塑料的总体污染水平[48].

2　结果分析

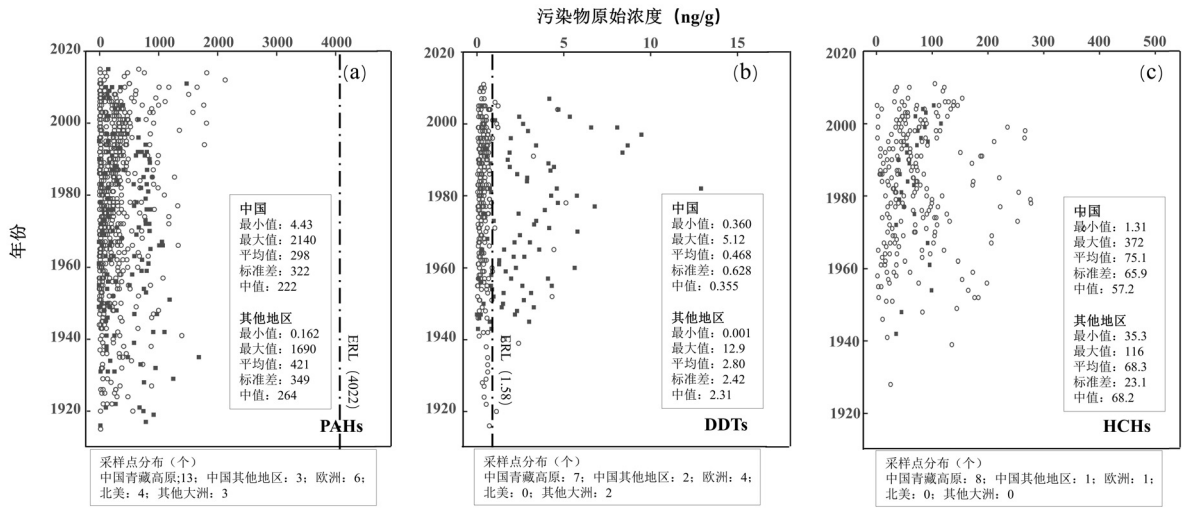
2.1　POPs 的沉积记录及归一化特征

图 2 为 1920~2020 年全球高山湖泊沉积物中 POPs 原始浓度与归一化浓度垂向分布.由于中国样点占比高,中国与世界高山湖泊沉积物中 POPs 变化趋势差异可能未完全反映中国与其他地区的污染物排放差异.本文仅根据已有研究和数据进行解析.

由图2(a)可得,PAHs浓度为0.16~2140ng/g,均值(324 ± 331ng/g)低于广西宣武八仙湖(1330~8770ng/g)[49]和南京玄武湖(796~10500ng/g)[50]等平原丘陵地区湖泊沉积物,也远低于效应区间低值(ERL)[51],潜在风险较低.除极少的自然来源外[52],PAHs 与人类经济发展息息相关,是工业化与城市化的指示剂[15-16].图 2(g)显示 PAHs 自 1920 年起在中国和全球范围内逐渐上升,反映了全球经济持续发展.中国趋势与世界趋势大致相似,但中国的增长趋势在 80 年代后期增速更快,与中国在 20 世纪 80 年代后的快速工业化相对应[53-54].该趋势也与 Shen 等[55]对 1995 年全球 PAHs 排放及发达国家与发展中国家的 PAHs 排放对比结果相符.

DDTs 浓度整体相对较低(0.001~12.9ng/g,均值1.13±1.96ng/g),仅有部分国外湖泊浓度高于 ERL 风险数值.其在 20 世纪 70 年代达到峰值,与全球广泛应用时期相对应[56].在20世纪80年代被全球禁用后,DDTs 的浓度随之明显下降.中国 DDTs 浓度下降更为显著,且高山湖泊浓度水平和峰值普遍低于全球其他地区,这可能由于中国相对落后的工农业发展所致[57].同为有机氯农药(OCPs)的 HCHs 浓度为1.3~372ng/g,均值(74.4±62.8)ng/g.中国 HCHs 浓度高于世界其他地区,可能与中国曾为全球最大工业 HCH 生产国有关[58].图 2(h)与图 2(i)显示 DDTs 和 HCHs 的垂向分布呈现两次峰值,分别为 20 世纪 80 年代和 21 世纪初.HCHs 的中国趋势与全球趋势基本相同.DDTs 趋势中国与全球存在轻微差异.60 至 80 年代的峰值与该时期 OCPs 大量使用有关[56],80 年代后降低趋势也与全球禁用相吻合.但 21 世纪初出现二次峰值,这说明 DDTs 和 HCHs 可能仍通过一次或二次源排放到环境中[59].

PCBs 浓度为 0.001~12.9ng/g,均值(1.13 ± 1.96)ng/g,趋势变化与其在工业生产中的使用高峰期一致.80 年代后,受全球生产限制导致 PCBs 浓度略微下降,但浓度增长趋势仍在继续.这可能与添加 PCBs 的相关产品(如电子设备等)在使用和废弃过程中仍持续释放 PCBs 到环境中有关[60].与其他地区相比,中国的数值变化略有滞后,且整体水平明显偏低.这可能是由于中国并非 PCBs 的主要生产国,且其相关排放量较低[61].图 2(j)显示 PCBs 自下而上呈“之”字形趋势,出现多次峰值,中国的归一化趋势增速远超世界平均水平,可能归因于中国高山湖泊沉积物中 PCBs 浓度较低导致的较高不确定性[62].与 OCPs 趋势相同的是,尽管 PCBs 在 20 世纪 80 年代受到生产限制,其垂向分布趋势仍在不断上升.具体来源及影响因素将在下一节中详细讨论.



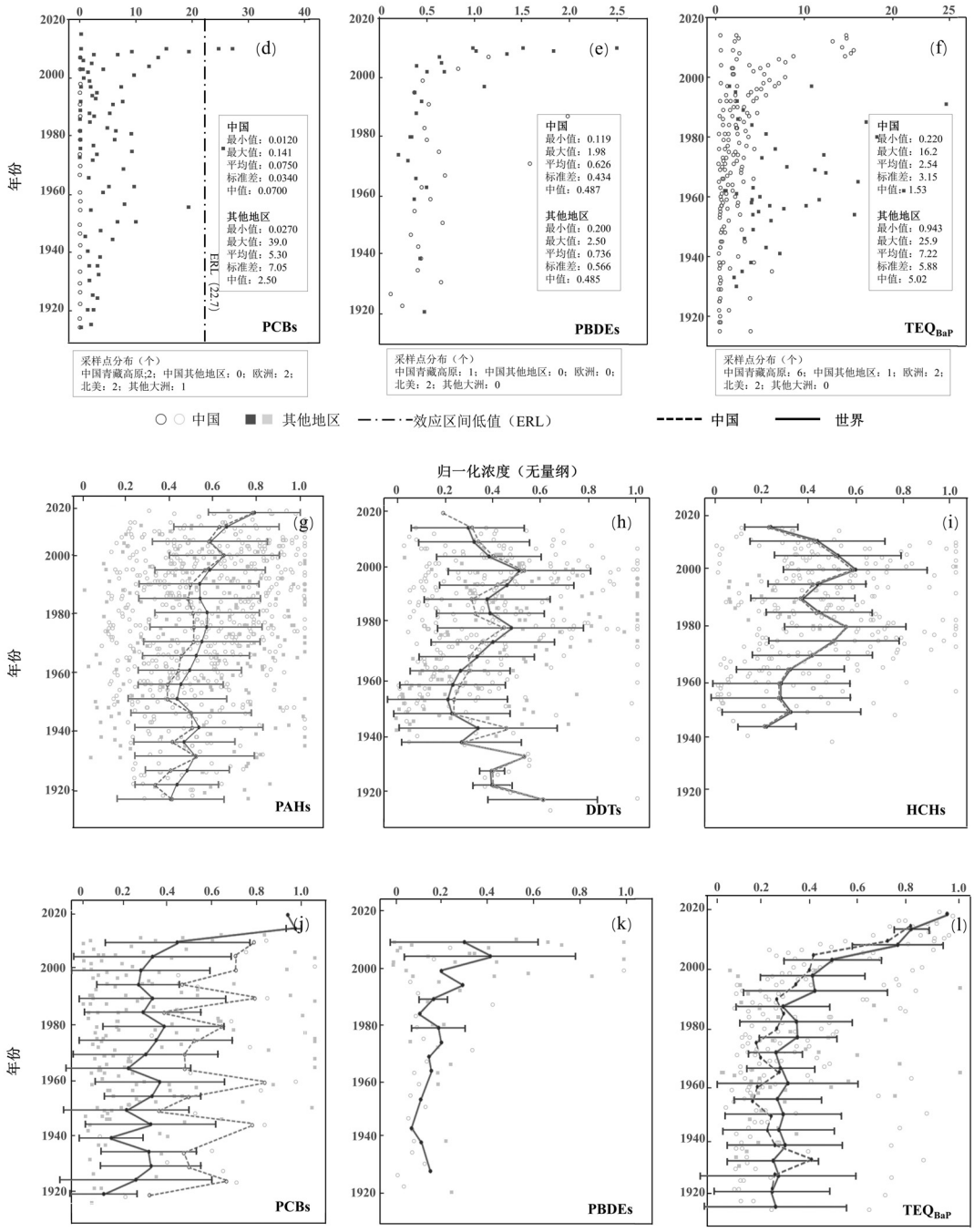


图 2 全球高山湖泊沉积物中 POPs 原始浓度与归一化浓度垂向分布

Fig.2 Vertical distribution of original and normalized concentrations of POPs in alpine lake sediments worldwide

原始浓度图中,空心圆代表中国,实心方块代表其他地区;归一化图中,空心圆表示归一化数据,实心圆和实线表示以五年为步长计算的全球平均值及标准差,虚线表示中国平均值

PBDEs 数值点较少,整体含量较低(0.12~2.5ng/g,均值 0.68±0.504ng/g),且在 20 世纪 70 年代才开始上升,与其作为新型阻燃剂在塑料制品等消费品中的应用有关.中国 PBDEs 含量在 20 世纪 80 年代才出现上升趋势,这与其在中国的使用时间较晚有关(自 80 年代逐渐开始使用[63]).由于近十年来 PBDEs 被大量用作阻燃剂,其增长趋势可能还会继续上升[64-65].

对于近年新纳入《斯德哥尔摩公约》的 POPs,其在高山湖泊沉积记录研究相对较少.近期研究仅讨论了高山湖泊沉积物中氯化石蜡[22]和全氟化合物[38]的赋存特征及来源,得克隆、UV-328 等新增列 POPs 尚未见报道.

2.2 高山湖泊中微塑料沉积记录

当前高山湖泊微塑料研究主要集中于表层水

体或水生生物,高山湖泊沉积物相关报道较少[6].目前统计到的高山湖泊研究集中在青藏高原地区,采样时间均在 2015 年后,且相关沉积物研究未对其进行测年分析,因此本文根据采样年份对相关数据进行归纳(图 3).据图 3 可知,高山湖泊沉积物中微塑料丰度(19~2640 个/kg,均值 624±889 个/kg)较低,数值均小于武汉汤逊湖(18100 个/kg)[66],呈现出逐年递增趋势.虽有研究表明,微塑料污染与人口密度和城市化程度相关[67],高山湖泊地区受人类活动影响较小,微塑料丰度应远小于人类活动密集湖泊区域.但已统计数据中多数丰度值高于美国安大略湖(488 个/kg)[68],依布茶卡丰度也与其他高山湖泊存在较大差异,这说明高山湖泊微塑料沉积状况仍在一定程度上受到人为或其他因素的影响.

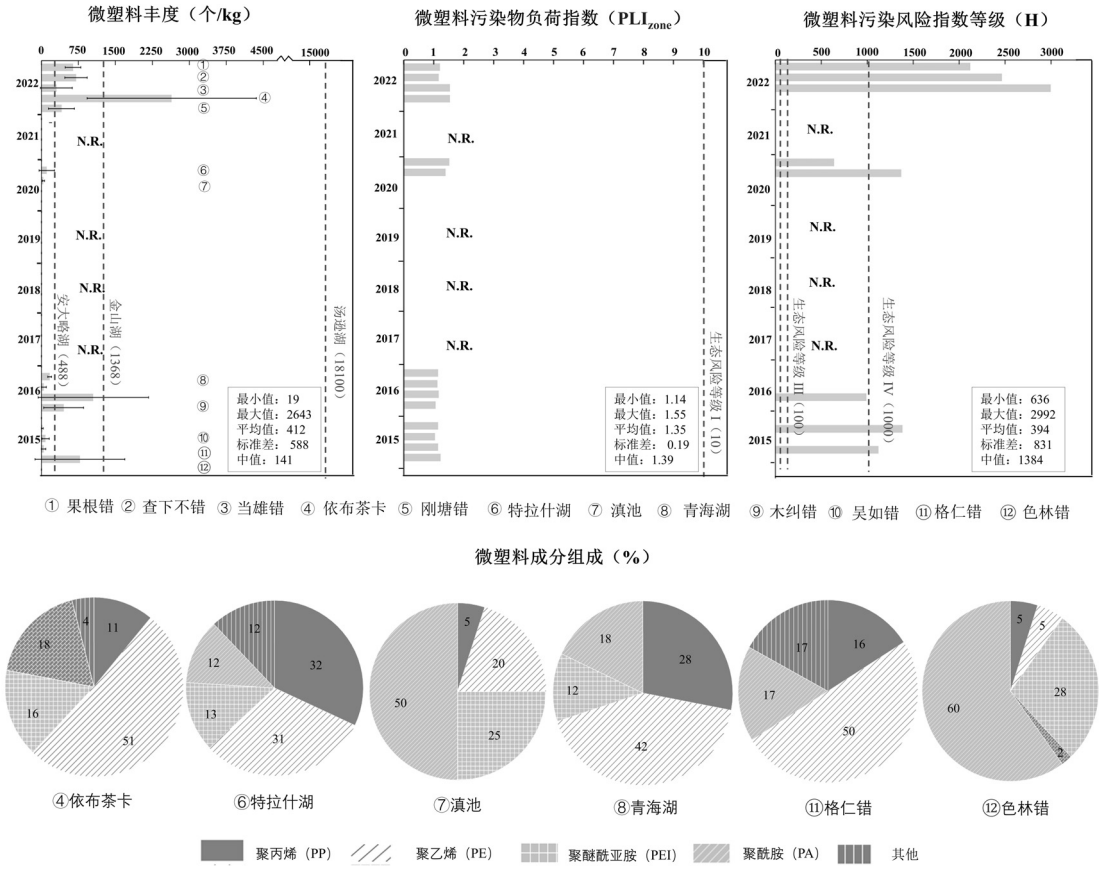


图 3　全球高山湖泊表层沉积物中微塑料丰度及类型

Fig.3　Abundance and types of microplastics in surface sediments of alpine lakes worldwide

当前已发表高山湖泊沉积物微塑料相关论文地点均位于中国

3　讨论

3.1　POPs 与微塑料的来源解析

PAHs 同分异构体之间的比值可判断其来源[16].通常 BaA/(BaA+Chr)<0.2、IcdP/(IcdP+BghiP)<0.2、Fla/(Fla+Pyr)<0.4 指示石油源;0.2<BaA/(BaA+Chr)<0.35、0.2<IcdP/(IcdP+BghiP)<0.5、0.4<Fla/(Fla+Pyr)<0.5 代表石油燃烧;而 BaA/(BaA+Chr)>0.35、IcdP/(IcdP+BghiP)>0.5 和 Fla/(Fla+Pyr)>0.5 指示煤和木材燃烧[69-70].此外,轻环 PAHs(2~3 环)与中重环 PAHs(4~6 环)比值可用于判断 PAHs 来源,因比值差异较大,对其取常用对数进行分析.比值小于 0 表明以高温燃烧(如机动车汽油燃烧)过程为主,大于 0 表明以低温燃烧(如生物质和煤炭燃烧)过程为主[20].图 4 显示了 PAHs 异构体比值的历史变化趋势.BaA/(BaA+Chr)比值呈出曲折下降趋势;IcdP/(IcdP+BghiP)比值波动起伏较大;Fla/(Fla+Pyr)比值趋势则较为稳定;轻环 PAHs 与中重环 PAHs 比值呈逐年下降趋势.由比值变化大致可得,高山地区历史上的 PAHs 污染来源以燃烧源为主,1980 年之前以煤

和木材的燃烧为主,21 世纪后则逐渐转变为以石油燃烧为主,高温燃烧过程产生的 PAHs 呈现不断增加趋势,这可能由周边地区汽车保有量以及湖泊上的船舶增多导致[16].但由于本文统计数据的复杂性,且存在部分单体未检出的情况,因此各比值间的历史趋势存在一定差异.

环境中 HCHs 主要来源于工业 HCH 和林丹的使用.工业 HCH 以 α-HCH(60%~70%)为主、γ-HCH 占比 10%~12%,而林丹几乎为纯的 γ-HCH.因此,可用 α-HCH/γ-HCH 比值指示 HCHs 的来源[71]:该比值在 5~7 时,主要来源于工业 HCH,小于 3 一般代表林丹和工业 HCH 的混合输入[72].如图 4 所示,α-HCH/γ-HCH 的历史趋势与 HCHs 浓度趋势相似,1970 年后的下降趋势与工业 HCH 的广泛禁用有关,二次上升趋势则与环境中 HCH 的二次释放相关[57].DDTs 在好氧条件下的主要降解产物为 DDE、陆地风化过程有利于 DDE 的形成和保存[73],用 DDT/DDE 比值趋势可指示 DDTs 降解情况[71].该值小于 1 可指示 DDTs 已经历充分降解,大于 1 则代表 DDTs 以母体化合物为主,可能为新近使用.由图 4 可知,DDT/DDE 的比值呈现从大于 1 到小于 1 的波动下降趋势,反映了 20 世纪 80 年代全球禁用后沉积的 DDTs 更可能是来自湖泊集水区和大气沉降的“风化”DDTs,而不是继续使用的“新鲜”DDTs.

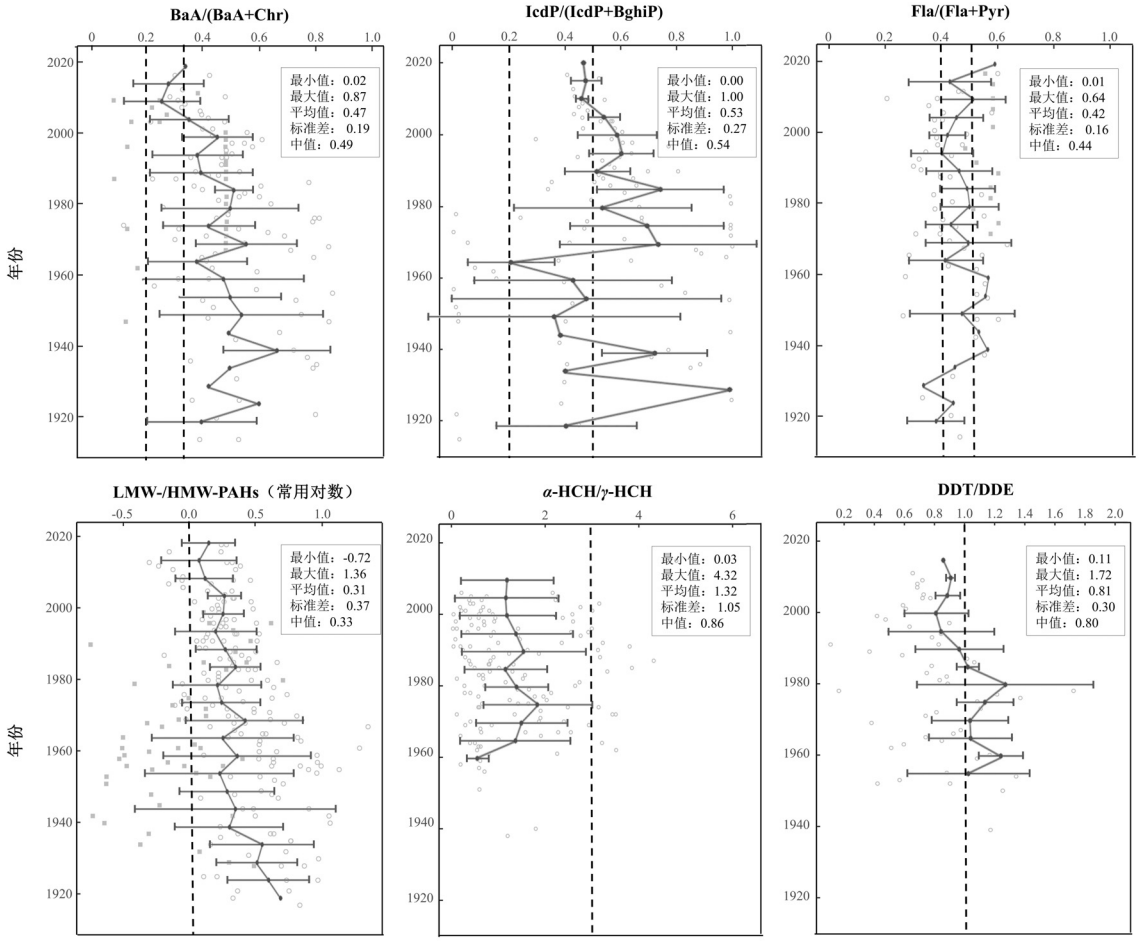


图 4 全球高山湖泊沉积物中 POPs 指纹化合物比值垂向分布

Fig.4 Vertical distribution of the ratios of fingerprint compounds for POPs in alpine lake sediments worldwide

形态特征、聚合物类型在一定程度上可反映微塑料的来源[28],本文选取 6 个数据详实的高山湖泊,通过微塑料组分对其来源进行探讨.由图 3 可知,聚乙烯和聚酰胺为高山湖泊表层沉积物中微塑料的主要成分.聚乙烯被广泛应用于工业和日用品,如纺织物、食品包装等,是环境中最常见的塑料[74],会随日用品磨损等途径进入环境,并通过河流汇集进入湖泊沉积物中[75].聚酰胺是塑料帐篷及青藏高原特

有经幡的主要材料,多放置于户外,受青藏高原多风、强太阳辐射等条件影响脱落进入环境[29].随着旅游业和宗教文化的不断发展,该来源成为影响青藏高原湖泊微塑料沉积的重要因素[76].

3.2　POPs 与微塑料生态风险评估

由图 2(l)可知,TEQ_{BaP} 数值不断增长,且近年来增速加快,已超过 PAHs 浓度的增长趋势,表明近年来新增 PAHs 以毒性较大的中重环 PAHs 为主.图 4 中的指纹化合物比值趋势也佐证了这一点.

各高山湖泊微塑料风险数值如图 3 所示.PLI_{zone} 数值较低,生态风险等级均为Ⅰ类且远低于标准值,表明高山湖泊沉积物中污染负荷风险较低;但 H 值较高,大多数高山湖泊风险等级为Ⅳ类,说明沉积物中微塑料的聚合物风险较高.由于微塑料可吸附携带其他污染物[77],其真实风险可能被低估.

3.3　不同地区 POPs 垂向分布变化及环境指示意义

图 5 对比了东亚、欧洲和北美高山湖泊沉积物中 PAHs 归一化浓度趋势,这三个地区是全球人口最密集、经济最发达地区.东亚 PAHs 浓度(4.43~1830 ng/g)随时间持续增加,北美浓度(49.4~1490 ng/g)在 1950 年左右达到最大值,欧洲浓度(17.5~1690 ng/g)则在 1960 年达到最大峰值,随后浓度开始下降.对比 Shen 等[78]统计中、美、德三国机动车 PAHs 排放量可得出,由于引入节能减排技术,欧美发达国家在 20 世纪 70 年代后道路机动车 PAHs 排放量大幅降低.虽然中国在九十年代后也开始逐步推广该技术,但其增长的机动车数量抵消了这一下降趋势[55].

高山湖泊沉积物中 DDTs 与 HCHs 的变化趋势与周边排放强度及全球 OCPs 使用管控密切相关[72].整体上中国 DDTs 浓度(0.03~5.12ng/g)低于欧洲(0.001~12.9ng/g),数值变化也较为平稳,与上文提及该时期中国较落后的工农业有关.与 OCPs 全球排放量的单次峰值不同的是[79],中国、欧洲的 DDTs 归一化浓度及中国的 HCHs 归一化浓度均为双峰趋势(图 5),可归因于气候变化下冰冻圈融化带来的污染物“二次”释放[62,80].

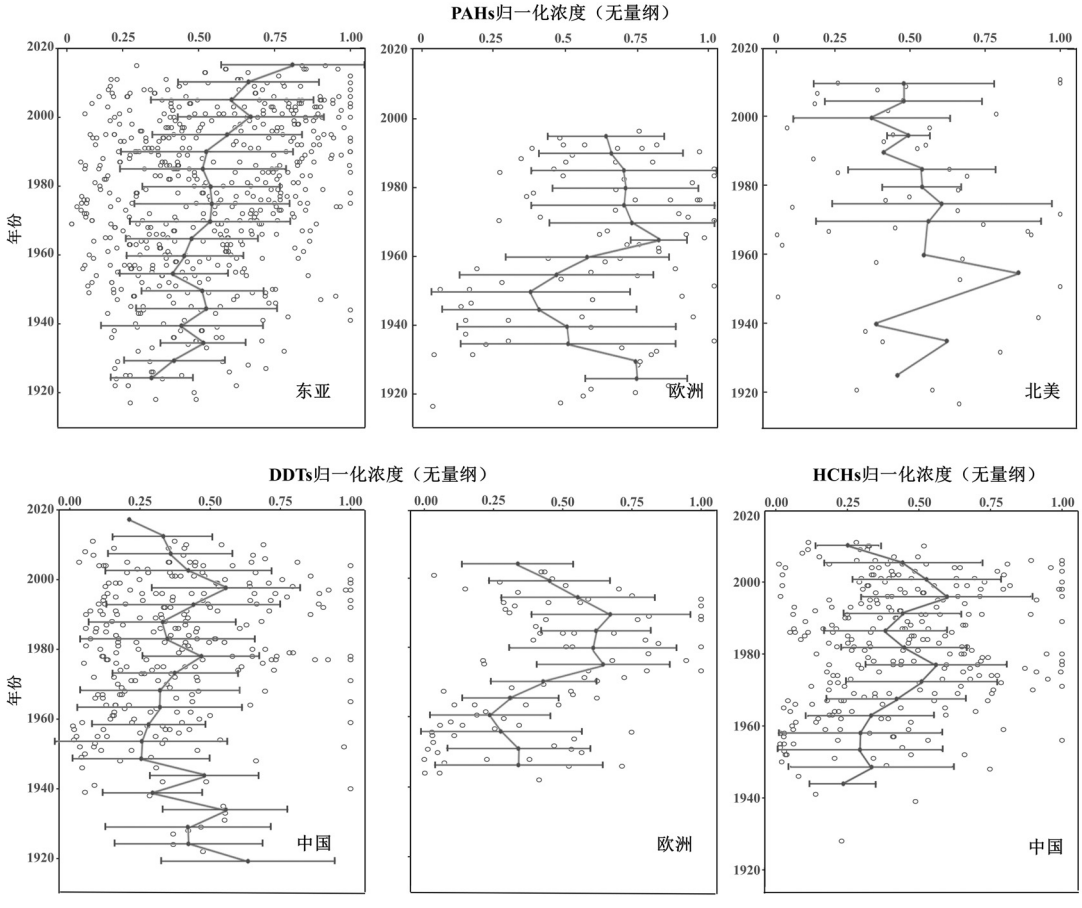


图 5　不同地区高山湖泊沉积物中 POPs 浓度垂向分布

Fig.5　Vertical distribution of POPs concentrations in alpine lake sediments across regions

3.4 高山湖泊 POPs 与微塑料沉积记录影响因素

3.4.1 区域人类活动与气候变化 本文统计文献中,共提及四类影响沉积物中新污染物含量的因素,包括人为活动、环境变化、技术原因等.42 篇文献中,有 20 次提及区域人类活动对沉积物中污染物含量和趋势的影响,成为被提及最多的影响因素.影响 POPs 沉积的人类活动包括了当地生物质燃烧[20]、能源结构的转变[15-16]、汽车轮船尾气排放[16]、旅游登山活动[14]、OCPs 农药过量使用[81]等.影响微塑料丰度的人类活动主要为日常塑料制品使用的磨损[27]、当地旅游和登山活动导致的纤维磨损[28]以及青藏高原独有的经幡磨损脱落产生[29].此外,施用地膜等农业活动也是影响青藏高原河谷地区微塑料丰度的重要人类活动因素[82].对于距离人口密集区较近的高山湖泊,人类活动排放的新污染物总量可能远高于大气长距离迁移带来的输入量.

高山湖泊作为气候敏感区,受到全球气候变暖的显著影响.文献中提及气候变化影响的有 13 次,提及次数仅次于区域人类活动.局地气象异常和全球气候变暖影响高山湖泊沉积环境[83].比如,厄尔尼诺现象使 POPs 的沉积发生变化[84];降水异常使得一些高山湖泊(尤其较低海拔、无冰川分布的高原热带湖泊)产生集水区扰动,影响新污染物在湖泊中的沉积[85].冰川加速融化使原本封存在冰川中的新污染物产生“二次释放”[80],全球 DDTs 和 HCHs 的浓度双峰趋势(图 2)、部分高山湖泊 PCBs 的近年浓度增长趋势[86]等均佐证了这一点[9].冰川融化也导致封存在冰雪中的微塑料进入湖泊[87].气候变暖持续加剧,对高山湖泊环境的影响将逐步加深.

3.4.2 定年技术手段及采样方法 百年尺度上污染物沉积历史重建须以精准的定年技术手段和标准的采样方法为基础.目前最为常见的测年方法是放射性同位素定年法,主要为放射性核素 ^{210}Pb(半衰期 22.3a)和 ^{137}Cs(半衰期 30.2a),以及用 ^{226}Ra、^{241}Am、^{239,240}Pu、^{129}I 等放射性核素进行佐证[88].由于 Cs 为碱金属元素,化学性质活跃、容易在沉积物中移动;Pb 同位素会受到半衰期的限制,定年的精确性和测量范围受到一定影响[89-90].为保证定年准确性,可通过如纹泥测年、火山灰校准、球状碳颗粒定年等多样化的定年手段进行多重验证[91-92].此外,沉积物采样方法的标准化对于准确重建历史污染物沉积记录也至关重要.比如,在微塑料研究中,不同采样网的网孔大小会显著影响到微塑料颗粒的回收率,进而导致不同研究间数据不具可比性[93].制定并普及快速且标准的微塑料采样方法尤为必要.

3.5 不可提取态新污染物:风险与示踪潜力

有机污染物进入沉积物、土壤等多孔介质后,可与基质紧密结合,形成不可提取态残留物(non-extractable residue,NER;又称结合态残留)[94].常规分析方法无法提取到 NER,使得沉积物中新污染物 NER 研究较为滞后.

部分新污染物可能在沉积物中形成较高比例的 NER.比如,渤海海湾沉积物中[95],壬基酚形成 NER 比例高达 80%~90%.新污染物的降解代谢产物也可能形成一定比例的 NER,使得其生态风险更具复杂性和隐蔽性.美国加州近海沉积物[96]和德国 Teltow 运河沉积物[97]中均发现,多种 DDTs 降解产物(如 DDA)可形成 NER,两地沉积物中 DDTs 及其降解产物形成 NER 比例分别为 5%和 52%.无论对于新污染物母体化合物还是其降解代谢产物,形成的 NER 均可能在一定条件下再活化,产生风险效应[98].

NER 可保留污染及环境变化信息,有助于污染物历史重建和来源识别[99].Li 等[23]对青藏高原湖泊沉积物中的多形态 PAHs 研究发现,PAHs-NER 的浓度与组成变化,可以作为人类世地层标志物.德国 Teltow 运河沉积物[97]中 DDTs-NER 研究表明,污染源识别不应仅依赖可提取态,NER 同样具有指示意义.

尽管目前高山湖泊沉积物中新污染物 NER 研究较为匮乏,但河流与海洋沉积物以及土壤中新污染物 NER 的研究框架可供参考.

3.6 当前研究不足的新污染物:抗生素与内分泌干扰物

高持久性和长距离迁移能力是化学品产生较高环境危害性的两个关键特征[100].与 POPs 相比,抗生素大气半衰期较短,难以通过大气远距离传输.比如,γ-HCH 和 PCB153 的大气半衰期分别为 14 和 50 d,磺胺甲恶唑和土霉素大气半衰期仅为 0.04 和 0.001 d(基于 OH 自由基反应速率常数进行计算[101]).高海拔区域紫外强度和自由基浓度较高,抗生素通常会在大气和表层水体中快速光降解,极少量物质可能由于进入颗粒物微孔[102]或与载体形成 NER[103]

的方式免于被光降解和生物降解.相比之下,抗生素抗性基因(ARGs)更易随大气传播和长距离迁移扩散[104],给高山地区带来未知生态环境风险.内分泌干扰物种类繁多,一部分与抗生素类似,大气半衰期相对较短;另一部分可能具备与 POPs 类似的长距离迁移及沉积途径,并进入高山湖泊沉积物中.

然而,近年来工农业、旅游等本地人类活动持续增加(见 3.4.1 部分),抗生素和内分泌干扰物可能通过地表径流等方式[105]直接进入高山湖泊.近期研究表明[106],高寒畜牧业发展及抗生素使用,可能使得高山湖泊成为抗生素的汇集地.

目前抗生素及内分泌干扰物研究集中于工业发达、人口稠密地区[107],偏远地区研究较少,高山湖泊沉积物中新污染物残留特征鲜见报道.高山水环境中仅有少量篇次关注抗生素和内分泌干扰物,如阿尔卑斯山河流中抗生素[108]及乌干达维多利亚湖水中有机磷酸酯[109]研究.

3.7　研究展望

当前有相关报道的高山湖泊主要分布在亚洲的青藏高原和欧洲的阿尔卑斯山.美洲的沉积物新污染物研究集中在五大湖地区,高山湖泊新污染物报道较少.对于有“高原大陆”之称的非洲,湖泊沉积物中新污染物历史沉积资料极为有限.非洲高原主要位于热带地区,区域气候机制复杂,工业活动集中度低,研究非洲高山湖泊新污染物的沉积历史对于理解热带地区人类活动和气候变化的影响具有重要意义.

大多数研究仅聚焦单类新污染物,较少同时关注产生途径类似的多种类新污染物.比如,多环芳烃相关报道较多,但燃烧伴生产物多氯萘、二噁英等未见报道;少量研究关注了微塑料对外源污染物的吸附携带行为,但微塑料本身也含有各类添加剂.加强对伴生或共迁移物质的研究,有助于更好开展高山湖泊中新污染物的源解析工作,为全面评估高山湖泊新污染物污染状况提供有力支撑.

4　结论

4.1　全球高山湖泊沉积物中 POPs 浓度整体较低.PAHs 浓度持续上升;DDTs 和 HCHs 在 20 世纪 70 年代前后达到峰值,21 世纪初出现二次峰值;PCBs 持续增加,中国地区低于其他地区;PBDEs 浓度较低,近年来呈现增长趋势.微塑料丰度相对较低,但整体呈增长趋势.PAHs 毒性当量不断上升,且近年增长趋势显著;微塑料污染负荷风险较低,但聚合物毒性风险较高.

4.2　高山湖泊沉积物中 POPs 主要源于燃烧和工农业活动.PAHs 来源逐渐由煤和木材燃烧转向石油燃烧;HCHs 来源于工业 HCH 和林丹的使用;20 世纪 80 年代禁用后,DDTs 主要来自“风化”DDTs 沉积.微塑料主要来源于日用品磨损及受区域旅游文化活动的影响.

4.3　东亚高山湖泊沉积物中 PAHs 归一化浓度持续上升,北美和欧洲在 20 世纪中后期出现下降现象.中国和欧洲 DDTs、中国 HCHs 趋势呈现一次排放和二次释放导致的双峰趋势.

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