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水体中短链氯化石蜡污染研究进展*

Review on Short-chain Chlorinated Paraffins Pollution in Water

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摘要:短链氯化石蜡(SCCPs)拟列入"斯德哥尔摩公约"禁止使用的一类持久性有机污染物,对生态环境和人体健康具有极大的潜在危害,已成为国际环境科学领域的一个研究热点。水体是SCCPs重要的"汇",本文着重综述了水生态系统中沉积物、水、生物体等不同环境介质中SCCPs的含量分布及对水生生物的危害,并对SCCPs的研究重点和前景进行展望。

关键词:短链氯化石蜡　水体　污染状况
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Abstract: Short-chain chlorinated paraffins (SCCPs), as a candidate of persistent organic pollutants in "Stockholm Convention", have potential hazard to ecological environment and people health, which becomes a new hotspot in field of environmental science worldwide. The water is an important destination of SCCPs. The distribution of SCCPs in different environmental media and the potential threaten of SCCPs to aquatic animals were critically reviewed in this paper. Meanwhile, the research progress and trend of SCCPs were also reviewed.

Key words: short-chain chlorinated paraffins, water, contamination level

0　引言

氯化石蜡(CPs)是直链正构烷烃的氯代衍生物,工业上用作阻燃剂、增塑剂、金属加工油和皮革处理剂等,其中碳链长度为10～13个碳原子的氯化石蜡被定义为短链氯化石蜡(SCCPs)。相对于中链氯化石蜡(MCCPs,碳链长度为14～17)和长链氯化石蜡(LCCPs,碳链长度为18～30),短链氯化石蜡因其较强的生物累积特性和更高的毒性而受到广泛关注。自20世纪50年代以来,我国已经开始生产氯化石蜡,随着国内塑料制品工业的迅速发展,氯化石蜡行业发展迅速。目前,我国已是氯化石蜡第一生产大国,截至2015年,国内氯化石蜡总产能达160万t。氯化石蜡产品中SCCPs含量普遍在6%以上[1-2]。这一方面对我国的生态环境和人体健康带来极大的潜在危害,另一方面面临着国际社会要求淘汰SCCPs消费的巨大压力。

2002年,欧盟水框架指令已禁止使用SCCPs,并将其列为水中的首要危险化学品之一。2008年"关于持久性有机污染物(POPs)斯德哥尔摩公约"将其列入POPs备选清单,我国也于同年启动对SCCPs的评估工作。2011年的斯德哥尔摩大会上SCCPs仅以一票之差最终未被列入POPs受控名

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单[3]。2015 年 3 月欧盟对现行的塑料制品法规(EU)No10/2011 进行升级,法规中进一步要求减少 SCCPs 的使用量[4]。2015 年 11 月欧盟又在《持久性有机污染物法规》中修订了对 SCCPs 的限制[5]。现有研究表明,SCCPs 普遍存在于各种环境介质中,尤其在水体中广泛存在。本文综述了 SCCPs 在水体中的环境行为及毒性研究进展,期望对 SCCPs 的深入研究提供参考。

1 SCCPs 的理化性质

SCCPs 是一类组成复杂的混合物,常温下为淡黄色或无色粘稠液体。SCCPs 亨利定律常数是 0.7～18 Pa·m^3·mol^{-1},蒸汽压为 3.2×10^{-4}～0.066 Pa(文献[6]),这表明 SCCPs 可以从大气中沉降到土壤、底泥和水中,也可以从上述介质中再活化到空气中。SCCPs 的辛醇水分配系数($LogK_{ow}$)的值为4.39～8.69(文献[7]),具有较高的脂溶性,因此在水生生物中有较强的生物富集及放大能力。SCCPs 也具有较高的辛醇空气分配系数($logK_{OA}$),表明 SCCPs 具有吸附在大气的有机相中进行长距离迁移的能力[8]。多数 SCCPs 混合物的溶解度是 22.4～994 μg·L^{-1},C_{10} 和 C_{13} 混合物的溶解度为 6.4～2370 μg·L^{-1}(文献[6])。随着碳链的增加,SCCPs 的水溶性和蒸气压均有下降的趋势,但与一些氯化芳烃不同,在 5 个氯原子以内,随着氯原子个数的增加,水溶性增大[9]。

2 水体中 SCCPs 的污染现状

目前为止还未发现自然来源的 SCCPs。水体中 SCCPs 主要来自 SCCPs 及其他链长 CPs 生产、储存、运输和使用过程中的排放;设备冲洗和使用后的金属加工/切削液也都可能成为 SCCPs 进入水生生态系统的源头。

2.1 SCCPs 在沉积物中的分布

沉积物是 SCCPs 重要的"汇",是国内外 SCCPs 研究的热点领域。在欧洲工业发达地区,巴塞罗那河流底泥中的含量为 0.25～3.04 μg·g^{-1}(干重)[10];挪威河流沉积物中检测出很高的 SCCPs(19400 ng·g^{-1})(湿重)[11]。捷克工业区附近河流沉积物中 SCCPs 浓度介于无检测值至 347.4 ng·g^{-1}(干重)之间[12]。北美五大湖地区,在安大略湖汉米尔顿港附近的沉积物中检测到最高浓度 SCCPs,五大湖其他区域 SCCPs 浓度为 5.9～410 ng·g^{-1}(干重)[13-14]。亚洲地区,日本 2 条河流沉积物中 SCCPs 水平为 4.9～484.4 ng·g^{-1}(干重)[15]。上述研究表明人类工业活动的强弱是决定该地区沉积物中 SCCPs 含量的主要原因。Tomy 等[16-17]检测到北极地区偏远湖泊沉积物中的 SCCPs 浓度为 1.6～257 ng·g^{-1}(干重),发现 SCCPs 在包括极地区域在内的广泛地理范围内普遍存在,表明 SCCPs 可以通过长距离大气传输在全球范围内分布。我国的 SCCPs 研究起步较晚,但进展很快。陈满英等[18]报道珠江三角洲地区沉积物(池塘、河流和河口及柱芯)SCCPs 的浓度为 320～6600 ng·g^{-1}(干重),该研究发现 SCCPs 沉积埋藏过程中可能存在脱氯的降解过程。陈茹等[19]报道了黄渤海地区沉积物样品中 SCCPs 浓度在 14.5～85.2 ng·g^{-1}(干重),陈晨等[20]报道辽河口海域沉积物中 SCCPs 的浓度为 64.9～1683.4 ng·g^{-1}(干重)。上述结果暗示河流排放是海洋中 SCCPs 的主要来源。李慧娟等[21]报道 SCCPs 在长江三角洲和闽浙沿岸的表层沉积物含量水平为 9.0～37.2 ng·g^{-1}(干重),该研究同时发现 SCCPs 在沉积物中的迁移很大程度上受总有机碳(TOC)影响。

2.2 SCCPs 在水中的分布

虽然 SCCPs 在水中的溶解度不高,但广泛存在于各种水环境中。西班牙 Llobregat 河和英国 Dawen 河的 SCCPs 浓度分别为 300～2100 ng·L^{-1}(文献[22-23])和 200～1700 ng·L^{-1}(文献[24]),高于已报道的日本几条河流(7.6～220 ng·L^{-1})[25-26]及加拿大劳伦斯河(15.74～59.57 ng·L^{-1})[27]的 SCCPs 含量。2000—2004 年对安大略湖的持续监测显示,湖中 SCCPs 的平均浓度为 1.194 ng·L^{-1},处于较低水平[28]。Castells 等[29]对西班牙污水处理厂进出水样品的检测发现:进水口 SCCPs 浓度为 310～620 ng·L^{-1};出水口 SCCPs 未检出。Zeng 等[30]测得北京高碑店污水处理厂进水口的 SCCPs 浓度(4200～4700 ng·L^{-1})显著高于出水口浓度(364～416 ng·L^{-1}),但活性污泥中 SCCPs 含量为(15.6±1.4)μg·g^{-1}(干重),结果暗示虽然污水处理厂能够有效净化水中 SCCPs,但沉积物容易造成二次污染。有关海水中 SCCPs 浓度的报道较少,于国龙[31]对辽宁普兰店湾的海水进行调查,测得 SCCPs 的浓度为 493.87～1490 ng·L^{-1};马新东等[32]研究辽东湾海域海水中 SCCPs 的浓度在 4.1～13.1 ng·L^{-1},结果表明海水中 SCCPs 的污染浓度与人类生产活动正相关。

2.3 SCCPs在水生生物中的分布

SCCPs的物理性质决定其在水生生物中有较强的生物富集及放大能力。目前已报道的各种水生生物体内SCCPs数据较多。英国河流中,底栖生物、蚯蚓和鱼类体内SCCPs的含量为0.05～0.8 $\mu g \cdot g^{-1}$(湿重)[24]。北美五大湖地区,安大略湖鱼类体内的SCCPs浓度为7.01～2630 $ng \cdot g^{-1}$(湿重)[33];密歇根湖和安大略湖糠虾体内SCCPs浓度的平均值为2.1～7.5 $ng \cdot g^{-1}$(脂重)[34];五大湖的鱼类和无脊椎生物中SCCPs浓度的平均值为130～500 $ng \cdot g^{-1}$(脂重)[35]。海洋中,挪威境内鲑鱼、鳕鱼和北极点红鲑等海洋鱼类体内SCCPs的浓度在108～3700 $ng \cdot g^{-1}$(脂重)[11]。Tomy等[8]测得北极地区白鲸、环斑海豹和海象等海洋哺乳动物体内的SCCPs浓度为95～626 $ng \cdot g^{-1}$(湿重)。上述水生生物样本体内SCCPs的主要成份是碳链长度较短和低氯代组分,这些组分的挥发性较高,更易被生物体累积。马新东等[36]测得辽东湾不同生物体内SCCPs的浓度为86～4400 $ng \cdot g^{-1}$(湿重),且不同物种间的含量差异比较显著,其中浓度最低的生物为浮游动物,浓度最高的为扇贝。于国龙等[31]测得辽河入海口海域14种生物体内的SCCPs含量为758.62～17027.26 $ng \cdot g^{-1}$(干重)。姜国等[37]测得长三角和珠三角地区不同种鱼类体内的SCCPs含量分别为8～130 $ng \cdot g^{-1}$和9～118 $ng \cdot g^{-1}$(湿重)。Yuan等[38]调查了中国环渤海区域9个沿海城市软体动物样品中的SCCPs浓度水平,发现SCCPs在我国海产品中可普遍被检出。现有研究表明,我国水生生物体内SCCPs的质量分数普遍位于目前世界报道水生动物体内SCCPs质量分数范围的中高端,赋存程度不容乐观。

2.4 SCCPs对水生生物的毒性效应

目前,国外以大鼠为对象对SCCPs的生物毒性做了一些的研究。不同剂量水平的SCCPs会导致大鼠出现各种临床症状,如脊椎突出,呼吸异常,活动减少,直至死亡[39]。Cooley等[40]发现幼年虹鳟鱼在含有SCCPs的环境中暴露21 d即出现严重的肝脏组织病理学反应,纤维损害和肝脏细胞坏死严重。Madeley和Maddock[41]通过SCCPs对虹鳟鱼的毒性研究,认为SCCPs对水生生物的毒性可能需要很长一段时间才能表达出来。另有研究表明,大型蚤(*Daphnia magna*)、糠虾(*Mysidopsis bahia*)和贻贝(*Mytilus edilus*)等水生无脊椎动物对SCCPs相对比较敏感[42-43]。现有研究认为SCCPs对陆生脊椎动物毒性较低,对鱼类的急性毒性阈值超过其在水中的溶解度,但一些水生无脊椎动物对SCCPs则相对比较敏感[9,44]。另外,由于SCCPs是一类亲脂性化合物,储存于脂肪中的SCCPs可通过脂肪代谢再进入血液而威胁健康[45]。总的来说,虽然SCCPs对多种生物的毒性作用已有相关报道,但研究广度和深度仍然不足。

3 展望

我国是CPs生产和消费大国,也是受SCCPs污染较重的国家之一。由于SCCPs具有持久性、长距离迁移性、生物富集性和生物毒性等特性,必须关注其对生态环境及人体健康的潜在风险;加大SCCPs的研究力度,对控制污染、保护环境具有深远的意义。目前,国内外对SCCPs的研究还集中在实验室阶段,不同实验室对同一种SCCPs样品分析结果差异巨大,开展相对成熟的、标准化的定量方法研究以及国际实验室间分析比对研究是必要的。SCCPs毒性效应和致毒机制的研究仍比较粗浅,需加强对其致毒效应的机制和分子机理的研究。另外海洋作为SCCPs重要的汇,应加强SCCPs在海洋中环境行为的研究力度。

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