

中国近海持久性毒害污染物研究进展

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摘要:持久性毒害污染物具有三致效应和遗传毒性,对近海生态环境危害极大。持久性毒害污染物主要通过入海河流和沿岸直排输入大海,主要赋存于近海沉积体系。持久性毒害污染物在沉积物分布累积主要受到沉积环境的影响,包括入海河流径流量、输沙量、水动力、河口海湾冲淤演变等,以及其自身物理化学性质和沉积物性质如颗粒物大小和有机质含量等。不同海区的研究表明,重金属污染整体上较轻微,胶州湾沉积物 Cu、As 污染较其它地区严重,珠江口盐沼 Cd、Zn 污染最为严重;近海沉积物有机污染主要集中在工业活动密集的珠江三角洲及邻近海域、长江口及闽江口海区,多氯联苯(PCBs)污染以珠江三角洲最为严重,有机氯农药(OCPs)污染在东南部河口及邻近海区较为严重。在明确持久性毒害污染物在不同沉积环境下差异和共性的基础上,提出若干今后需加强的研究,包括污染物迁移转化规律、重金属化学形态分析、微生物降解机制和污染物相互作用,等。

关键词:持久性毒害污染物;重金属;持久性有机污染物;分布特征

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Persistent toxic substances in offshore zone of China: a review

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Abstract: Persistent toxic substances (PTS), which have carcinogenesis, teratogenesis and mutagenesis and ecotoxicity, can cause heavy damages to coastal ecological environments and systems. PTS are mainly transported into coast and adjacent sea by riverine input and/or industrial wastewaters discharge. Most of PTS, especial for particle-active components, are deposition in coastal sedimentary environment. The distribution and accumulation of PTS in coastal surface sediments are determined not only by the properties of PTS and surface sediment but by sedimentary environments such as river runoffs, fluvial sediment discharge, alternation between erosion and deposition in estuary areas. Based on the collected data, the heavy metal contamination levels in coastal sediments of China are generally low, exception of contents of Cu and As in Jiaozhou Bay and Cd and Zn in Pearl River Estuary. Persistent Organic Pollutants (POPs) are mostly concentrated in highly urbanized and industrialized offshore zones, such as the Pearl River Delta, the Changjiang estuary and Minjiang River estuary. Polychlorinated biphenyls(PCBs) pollution is more heavier in the Pearl River Delta than in other areas. Much heavy pollution of organochlorine pesticides (OCPs) was observed in estuarine and coastal zones in southeastern China. Based on the various behaviors of PTS in different coastal environments several research issues, such as quantitative research

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on transport and transformation of pollutants, chemical speciation analysis of heavy metals, microbial action, and the interaction among various pollutants, are elucidated.

Key Words: PTS; Heavy metal; POPs; offshore zone; distribution

据2006年中国海洋环境质量公报,近年来我国海域总体污染形势严峻,生态系统健康状况持续恶化趋势仍未得到有效缓解,污染源主要为无机氮和活性磷酸盐等。此类污染物生物降解性较强,如果治理措施恰当可以一定程度修复生态环境。持久性毒害污染物是一类具有强毒性、环境难降解性,并通过食物链在生物体内累积放大的污染物,其危害比常规污染物更为严重。近海是我国最为重要的鱼类产地,持久性毒害污染物对于近海环境的危害极大,影响沿海地区的可持续发展和国民健康。2006年我国首次开展部分入海排污口的持久性毒害污染物监测,我国近海环境研究热点多集中持久性毒害污染物总体含量^[1~4]、空间分布特征^[5~9]以及对海洋生态影响评价^[10~15]等方面。

持久性毒害污染物包括有毒重金属(Pb、Hg、Cd、Cr、As,等)、持久性有机污染物多氯联苯(polychlorinated biphenyls,PCBs)、多环芳烃(polycyclic aromatic hydrocarbons,PAHs)、有机氯农药(organochlorine pesticides,OCPs),以及一些有机金属化合物,等。这类污染物持久性和远距离迁移能力很强,其环境行为往往具有一定的相似性。因此,有必要总结我国近海持久性毒害污染研究的进展,归纳此类污染物来源,在近海环境中的分布特征及控制因素、污染水平与生态影响评价,分析中国近海不同地球化学环境下持久性毒害污染物的差异和共性,在此基础上提出若干今后需要进一步研究的问题。这将有助于全面认识我国近海污染的现状与趋势,对于制订合理的治理对策具有重要意义。

1 持久性毒害污染物来源

我国近海持久性毒害污染物主要通过入海河流或沿岸直接排放输送入海,还包括船舶、海上采矿、海洋倾倒废弃物^[16]等海上活动以及大气输送^[6,17]等方式,其中河流携带重金属3.0万t,砷0.6万t,入海排污口排海的重金属4.6万t,挥发酚、氰化物、苯胺、硝基苯、铬、硫化物等8.4万t^[18]。可见,重金属污染物以沿岸直排污污染源为主。多环芳烃、有机氯农药、多氯联苯类等持久性有机污染物在排海污水中也普遍检出^[18]。大气输送对重金属向近海输送的贡献已纳入常规海洋环境监测范围,长期监测结果表明我国重点海域大气气溶胶的Cr、Cu、Pb沉降通量呈上升趋势^[18];对有机污染物的海-气交换通量目前还未进行长期大范围监测。我国海洋倾倒废弃物主要为清洁疏浚物,90%以上倾倒区的环境状况未发生显著变化^[18],以东海倾倒区为例,重金属生态危害程度非常轻微^[16]。

沉积物中重金属来源复杂多样,我国有关重金属来源判断的研究方法大多采用主成分分析(PCA)^[6,19,20]、元素富集因子法(EF)^[21~23]以及²⁰⁶Pb/²⁰⁷Pb^[1,6]等方法。持久性有机污染物来源判断多采用生物标志物等方式判断,如PAHs的来源判断多采用低分子量与高分子量的比率(LMW/HMW)^[24,25],或荧蒽/(荧蒽+芘)、菲/蒽、荧蒽/芘等组分之间比值方法^[14,24,26,27]。

2 持久性毒害污染物分布特征的控制因素

近海环境持久性毒害污染物主要源于工业废水排放,进入水体的污染物主要吸附在悬浮颗粒物上,随颗粒物沉降进入沉积体系。持久性毒害污染物在沉积物中的含量与分布受到多种因素控制,一方面与其自身物理化学性质相关,另一方面也与所处沉积环境密切相关。不同海区之间污染状况既存在一定相似性,在污染物来源、污染状况、影响因素等方面又各具特色。掌握不同海区持久性污染物分布特征形成的主导因素,对于海洋环境监测与治理具有重要意义。

2.1 持久性毒害污染物物理化学性质

对珠江口的研究^[28]发现,Zn比TiO₂、V、Cr等污染范围大得多,这主要是由于Zn总是以溶解态存在,迁移能力强,只有在最大混浊带附近才会因胶体吸附作用而沉淀下来。重金属在近海环境主要地球化学过程包

括沉淀作用、络合作用、吸附作用,等。长江口最大混浊带附近水体重金属以悬浮颗粒表面有机物为媒介吸附在悬浮颗粒表面,导致重金属浓度在最大混浊带附近形成峰值^[29]。PCBs 也会发生一系列挥发、溶解、吸附和脱附、化学分解以及微生物降解过程,使得低氯原子数同系物含量减少而高氯原子数升高,明显改变沉积物中 PCBs 组分^[30]。PAHs 的环境行为与 PCBs 类似,环数越高对悬浮颗粒吸附能力就越强,使得赋存到沉积物中的 PAHs 往往分子量较高^[31]。OCPs 研究以六六六类有机氯农药 (hexachlorocyclohexanes, HCHs) 和滴滴涕类有机氯农药 (dichlorodiphenyltrichloroethane, DDT) 为热点。DDT 在不同自然条件下降解产物不同,厌氧条件下代谢产物多为 DDD,好氧条件下代谢产物多为 DDE,因此很多研究^[32~34]利用 DDT 与其代谢产物 (DDD + DDE) 的比值、p,p'-DDE 与 DDT 比值^[35],或 p,p'-DDT 与 DDT 比值^[36]指示研究区是否存在 DDT 的新输入源。可见,持久性毒害污染物自身物理化学性质和环境行为是影响它们在沉积物和生物体中的含量和分布的重要因素。

2.2 排放源和沉积环境

持久性毒害污染物在沉积物中的含量和分布直接受到污染源位置影响。排海重金属污染物分布峰值往往位于排污口一定范围内的沉积物中。胶州湾底质沉积物 Cd、Cr、Cu 和 Pb 的高含量区直接分布于排污口^[3]。长江口潮滩^[37]、珠江三角洲^[38]、大连湾^[8]、厦门西港及闽江口^[39]等海区也都表现出研究站点距排放源越近,有机污染物含量越高的特点。

近海沉积环境,如入海河流径流量、携带泥沙量、河口海湾冲淤演变、水动力作用、沉积物颗粒大小及有机质含量等,多种控制因素决定了持久性毒害污染物在沉积物中的分布特征。长江入海引起的水体盐度变化对悬浮物质絮凝沉降产生影响,同时改变水、颗粒相吸附解吸,使得溶解态重金属在迁移过程中表现出非保守特征^[40]。珠江携带颗粒物主要沉积在珠江口西部,造成河口表层沉积物中重金属分布呈西高东低特点,同时珠江口洪淤枯冲,使得重金属含量出现季节性差异^[41]。长江口系丰水多沙型河口,表层沉积物的重金属分布形成东西纵向两侧低、中间高,南北横向南高北低的格局^[42];大量泥沙产生独特的稀释机制减轻长江口重金属污染,导致其含量明显低于沿岸潮滩^[43]。在胶州湾^[3],外海潮波基本沿沧口水道、中央水道和西侧水道进入湾内,这 3 条水道支配了重金属分布,特别是沧口水道阻止胶州湾东岸污染物往西扩散,使水道以西地区基本保持重金属含量较低状态。杭州湾^[7]的细颗粒悬沙分布格局东高西低、北高南低,颗粒态重金属 Cu、Pb 也显示出类似分布特征,这主要是由于细颗粒物质比表面大吸附力强,成为持久性毒害污染物主要载体。深圳大亚湾红树林湿地^[27]也研究发现,PAHs 和粘土、总有机碳之间存在显著正相关关系。由此可见,只有了解不同海区沉积动力过程和沉积物组成,才能正确分析持久性毒害污染物分布特征。

3 我国主要海区持久性毒害污染物污染水平及评价

沉积物作为持久性毒害污染物的巨大储库,其污染程度一方面影响底栖生物生存环境,另一方面也会在沉积地球化学环境,如水体温度、盐度、pH、氧化还原电位、络合容量等物理化学条件发生变化时,将累积的污染物重新释放出来,形成 2 次污染。总结持久性毒害污染物在不同海区的污染水平和生态影响评价,明确我国近海不同地球化学环境中持久性毒害污染物的差异和共性,对于海洋环境监测与治理具有重要意义。表 1、表 2 分别归纳了我国主要海区表层沉积物的重金属、有机物污染水平。

我国近海沉积物重金属污染整体上比较轻微,海洋环境公报也表明近岸海域沉积物质量总体上良好,但个别海域污染已达到需要重视的程度。从表 1 可以发现,胶州湾沉积物 Cu、As 污染较其它地区严重,富集因子分析结果^[45]表明 Cu 达到中度污染,As 在部分海区,如大沽河口,污染较为严重。珠江口、上海滨岸潮滩、西厦门湾、胶州湾、大亚湾等近海环境 Pb 污染水平比较接近,均没有超过沉积物质量一类标准,但根据富集因子法和地质积累指数的计算结果^[47],西厦门湾普遍存在 Pb 污染。Cr 在胶州湾、上海滨岸潮滩、西厦门湾和珠江口盐沼的污染水平也较为接近。Cd、Zn 污染,以珠江口盐沼最为严重,特别是 Cd 污染已经超过海洋沉积物质量二类标准,甚至是农业土壤环境质量标准 12 倍,表明珠江口盐沼湿地重金属污染已经对当前农业开垦造成严重生态危害^[49]。天津海域、胶州湾部分海区 Hg 污染程度较为接近,富集因子分析结果^[45]表明胶州湾

部分海区存在 Hg 污染。某些对于环境敏感的生物可用于记录或指示环境中重金属污染状况, 目前多采用蚌类^[52,53]、鱼类、虾蟹^[54]等作为海洋环境重金属污染的生物指示。渤海蚌类 Cd 含量超过国家生物质量标准, 特别是莱州湾受黄河排污影响, 其蚌类 As、Hg、Pb 含量均高于渤海其它地区^[53]。东海沿岸蚌类重金属含量研究发现, 崇明岛、杭州湾蚌类体内重金属含量高于东海沿岸其它地区^[52]。珠江口鱼类 Pb 污染严重, 总体上珠江口水生生物痕量金属平均浓度高于中国其它地区, 且最近几年仍有增加趋势^[54]。

表 1 我国主要海区表层沉积物重金属污染水平(mg/kg)

Table 1 Concentrations of heavy metals in surface sediments in main coastal areas of China

海区 Area	Cu	Pb	Zn	Cd	Hg	As	Cr
天津海域 Tianjin's Sea ^[44]	24.70 ~ 32.04	15.21 ~ 19.41	74.63 ~ 147.64	0.07 ~ 0.20	0.25 ~ 0.97	5.46 ~ 7.52	—
胶州湾 Jiaozhou Bay ^[45]	9.74 ~ 499	14.19 ~ 91.21	47.64 ~ 170	0.072 ~ 1.94	0.018 ~ 2.11	21.02 ~ 37.84	42.7 ~ 430
苏北浅滩 North Jiangsu Shoal ^[46]	—	—	—	0.059 ~ 0.183	0.007 ~ 0.0369	5.3 ~ 19.2	—
上海滨岸潮滩 Tidal Flat of Shanghai Coastal Zone ^[2]	91	47	189	0.03	—	—	78
崇明东滩 Chongming wetland ^[4]	42	38	118	70	—	—	—
西厦门湾 Western Xiamen Bay ^[47]	19 ~ 97	45 ~ 60	65 ~ 223	0.11 ~ 1.01	—	—	37 ~ 134
厦门同安湾/大嶝海域 Tong'An Bay/Dadeng ^[48]	20.4/25.2	31.8/26.6	—	0.057/0.096	0.060/0.047	7.27/6.65	—
珠江口 Pearl River Estuary ^[41]	22.5 ~ 66.7	28.1 ~ 85.3	68.5 ~ 255.5	0.02 ~ 4.10	—	—	—
珠江口盐沼 Coastal wetland of Pearl River Estuary ^[49]	14.7 ~ 141.1	28.5 ~ 39.4	239.4 ~ 345.7	2.79 ~ 4.65	—	—	40.2 ~ 110.9
深圳湾 Shenzhen Bay ^[50]	50	40	137	—	—	—	—
大亚湾 Daya Bay ^[51]	24	32	89	0.042	0.162	8.0	—

表 2 我国主要海区表层沉积物有机污染水平(ng/g)

Table 2 Concentrations of priority control organic pollutants in surface sediments in main coastal areas of China

海区 Area	PCBs	PAHs	HCHs	DDT	OCPs
东海 East China Sea	—	—	0.05 ~ 1.45 ^[58]	<0.06 ~ 6.04 ^[58]	—
长江口 Yangtze Estuary	0.19 ~ 18.85 ^[59]	22 ~ 182 ^[60]	ND ~ 30.40 ^[37]	ND ~ 0.57 ^[37]	1.25 ~ 36.01 ^[59]
珠江三角洲海岸 coast of Pearl River Delta	6.0 ~ 290 ^[10]	94 ~ 4300 ^[10]	11.95 ~ 352.62 ^[10]	1.4 ~ 600 ^[10]	—
珠江三角洲 Pearl River Delta	11.5 ~ 485.45 ^[30]	217 ~ 2680 ^[13]	0.14 ~ 17.04 ^[38]	2.6 ~ 1628.8 ^[38]	—
大亚湾 Daya Bay	0.85 ~ 27.37 ^[34]	115 ~ 1134 ^[61]	0.32 ~ 4.16 ^[34]	0.14 ~ 20.27 ^[34]	2.43 ~ 86.25 ^[34]
渤海 Bohai Sea	ND ~ 2.1 ^[9]	28 ~ 1081.9 ^[11]	—	0.4 ~ 2.0 ^[9]	—
大连湾 Dalian Bay	1.0 ~ 153.13 ^[8]	32.7 ~ 3559 ^[62]	7.535 ~ 92.30 ^[8]	2.118 ~ 72.3 ^[8]	—
锦州湾 Jinzhou Bay	0.6 ~ 32.56 ^[8]	—	5.77 ~ 323.07 ^[8]	0.97 ~ 154.9 ^[8]	—
厦门海港 Xiamen Harbour	<0.01 ~ 0.32 ^[63]	247 ~ 480 ^[63]	<0.01 ~ 0.14 ^[63]	<0.01 ~ 0.06 ^[63]	<0.01 ~ 0.58 ^[63]
厦门西港 Xiamen Western Harbour	9.72 ~ 33.72 ^[39]	425.3 ~ 1522 ^[39]	—	8.61 ~ 73.7 ^[39]	—
闽江河口 Minjiang River Estuary	15.1 ~ 57.9 ^[64]	112 ~ 877 ^[65]	2.99 ~ 16.21 ^[64]	1.57 ~ 13.06 ^[64]	28.8 ~ 52.07 ^[64]

—: 未检出

研究区域重金属的天然来源构成重金属的背景值, 在污染程度与生态影响的评价中是必须考虑的因素。重金属背景值的确定最好采用具体研究区域沉积物的背景值^[20], 如果缺乏这方面的数据, 还可以采用全球页

岩平均组成^[55]、一类海洋沉积物质量标准^[41]、工业化前沉积物中重金属的全球最高背景值^[16]。此外,也可以利用参考元素(Al^[21,43,56]、Fe^[47]、Fe₂O₃^[28]等)与重金属元素之间的相关关系确定背景值。目前多采用潜在生态危害指数法分析沉积物中重金属含量^[12,16,20],很多研究还结合地质积累指数法^[57]、富集因子法^[21]等,多种评价方法的综合运用有助于全面反映研究区域重金属污染程度以及其生态危害程度。

对比我国主要海区沉积物有机污染物的含量(表2),PCBs污染,以珠江三角洲最为严重,多由电子垃圾的不正确处置造成。珠江河口蚌类等生物体有机污染物含量也表明PCBs污染已经到了可能危害食用大量海产品人群健康的水平^[66];珠江三角洲沉积物毒害有机物风险评价结果^[14]表明,某些地区(如广州河段和澳门河口)为高生态风险区,可能对生态环境产生严重影响。对于PAHs污染,各海区的测定方法、PAHs总量确定以及数据分析手段多有不同,增加了地区之间对比的难度和不确定性。高度城市化、工业化的沿海地区,PAHs污染也较为严重。长江口作为商业、运输枢纽,PAHs含量虽处于低—中等水平,但个别PAHs化合物(蒽和芴)含量已超过基于生物毒性试验的沉积物质量标准^[67]。各海区PAHs污染来源不尽相同,长江口近岸潮滩^[67]、珠江三角洲大部分地区^[14]、大连湾近岸^[62]、厦门西港^[39]等海区主要来源于石油类污染,而鸭绿江^[26]、渤海大部分海区^[11]、北黄海^[68]、闽江口^[39]等海区主要来源于矿物燃料燃烧。

表2大致反映出我国OCPs污染以东南部河口及邻近海区较为严重,闽江口沉积物中某些OCPs含量已接近我国国家水质标准临界值^[64],长江口潮滩沉积物中OCPs含量尚未对生物产生显著影响^[37],珠江三角洲沉积物OCPs污染已经达到可能影响当地渔民健康的程度^[10]。整体上,有机污染水平与经济发展水平直接相关,珠江三角洲及邻近海域,长江及厦门闽江口工业活动密集,经济发展速度较快,持久性有机污染较其它地区严重。

4 研究展望

近年来,由于人类活动对近海海洋环境的影响日益加剧,进入海洋的持久性毒害污染物正在改变海洋生物地球化学环境,对海洋生态系统危害程度加大。针对我国近海持久性毒害污染物的研究已有大量报道,但尚存在许多需要进一步关注的问题,主要包括以下几方面:

(1) 我国目前尚缺乏对近海环境长期连续的多学科综合性调查,多数研究尚停留在调查局部海区污染物含量、平面分布特征以及对海洋生态环境影响的评价上,对于污染物迁移转化规律多为定性描述,针对其过程与积累演化机制的定量研究明显不足。

(2) 研究多利用重金属污染物总量评价其生态危害程度,较少涉及重金属化学形态分析,无法体现不同形态重金属生物有效性和生物毒性差异。沉积物重金属化学形态的研究可加深对水体污染物迁移转化机理的认识,但尚存在很多不足,如提取方法不同会导致不同研究者的数据缺乏可比性,等。

(3) 对于近海持久性毒害污染物来源的判断,无论重金属还是有机污染物,都是分析自身或其组分含量,得到近似结论。应结合同位素示踪技术明确其污染源和迁移途径,加深对研究区域环境污染历史的认识。

(4) 近海水体和沉积物中存在大量微生物,这些微生物对排入近海的持久性毒害污染物具有吸收、转化、降解作用。在微生物影响下,这些污染物地球化学行为发生一系列变化,如重金属能够与微生物形成稳定的螯合物而降低其毒性,目前对于这些变化及微生物降解机制的研究还不够深入。

(5) 已有的研究多根据某种单一污染物在近海环境中的生物地球化学行为进行评价和预测,很少考虑不同污染物之间的相互作用。因此,研究近海持久性毒害污染物之间的相互作用对于评价其共同导致的生态环境影响是十分必要的。

为了更为有效地监测、预测我国近海环境的质量状况,今后需要注意以下问题:

- (1) 继续已有观测,注重定量研究持久性毒害污染物的迁移、转化过程,取得定量的参数化成果;
- (2) 加强沉积物重金属形态分析,今后在样品收集、测试、分析手段上应统一,另一方面也需要发展新型环境监测技术和化学分析方法,以获得准确、可靠、可比性强的数据;
- (3) 同位素示踪技术作为一种研究环境生态问题的有效手段,应加大在海洋环境污染方面的应用力度;

(4) 近海环境中微生物作用的研究,对于发展近海环境重金属和有机物污染的生物处理技术,以及污染海区的生态修复都具有重要指导作用;

(5) 开展海洋环境、海洋生物与环境污染物之间的耦合关系研究,提出立足中国近海的区域特点,又具有一定普适意义的模型与理论;

(6) 加强数据分析技术和数学模型等手段的运用,建立海域污染状况数据库,并适当运用遥感与地理信息技术,这对于建立污染物影响下的生态系统动力学模型具有重要意义。

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