

我国海洋环境典型新污染物的分布及生态效应

揭晓蒙¹, 王文涛¹, 咎飞翔², 沈国锋³, 王菊英⁴

(1.中国 21 世纪议程管理中心, 北京 100038; 2.华中科技大学 环境科学与工程学院, 湖北 武汉 430074;
3.北京大学 城市与环境学院, 北京 100871; 4.国家海洋环境监测中心, 辽宁 大连 116023)

摘 要:国内外广泛关注的新污染物包括持久性有机污染物、内分泌干扰物、抗生素、微塑料等, 近年来在海洋环境中被频繁检出, 新污染物的分析检测方法、环境行为、毒性识别与风险评估等问题已成为海洋生态环境领域的研究热点。本文重点分析了全氟/多氟烷基化合物、阻燃剂、内分泌干扰物、抗生素和微塑料等新污染物在我国海洋环境的污染现状和生态效应, 并与其他国家及地区海洋环境中的新污染物分布进行了比较, 以期为全面开展我国海洋新污染物的陆海统筹协同治理提供技术支撑。

关键词:全氟及多氟烷基化合物; 阻燃剂; 内分泌干扰物; 抗生素; 微塑料; 海洋环境生态效应

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Distribution and ecological effects of typical emerging pollutants in marine environment of China

JIE Xiaomeng¹, WANG Wentao¹, ZAN Feixiang², SHEN Guofeng³, WANG Juying⁴

(1.Administrative Center for China's Agenda 21, Beijing 100038, China; 2.School of Environmental Science & Engineering, Huazhong University of Science and Technology, Wuhan 430074, China; 3.College of Urban and Environmental Sciences, Beijing University, Beijing 100871, China; 4.National Marine Environmental Monitoring Center, Dalian 116023, China)

Abstract: The emerging pollutants, including persistent organic pollutants (POPs), endocrine disruptors, antibiotics and microplastics, as one of global environmental main concerns, have been frequently detected in the marine ecosystem. As an important natural “sink” for pollutants, studies on the emerging pollutants detection, environmental behavior, toxicity identification, and risk assessment in the marine environment has become a hot topic. This paper comprehensively analyzed the pollution condition and ecological effects of typical emerging pollutants, i.e., per- and polyfluoroalkyl substances, flame retardants, endocrine disrupting compounds, antibiotics and microplastics, on marine environment in China, and compared the domestic condition with the relevant countries and regions. The results could provide technical support for comprehensive land-sea environmental protection of emerging pollutants in China.

Key words: per- and polyfluoroalkyl substances (PFAS); flame retardants; endocrine disrupting compounds; antibiotics; microplastics; ecological effects in the marine environment

近年来, 随着经济社会的高速发展, 新污染物引发的环境问题日益严峻, 不仅对全球生态环

境和人类健康造成严重威胁, 也在一定程度上制约着联合国可持续发展目标的实现^[1]。新污染

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作者简介: 揭晓蒙 (1988—), 女, 北京人, 副研究员, 硕士, 主要从事海洋科技政策、可持续发展与能源环境经济研究, E-mail: jiexm@acca21.org.cn

通信作者: 王文涛 (1982—), 男, 河南焦作人, 研究员, 博士, 主要从事海洋和极地生态环境保护、气候变化与可持续发展研究, E-mail: wangwt@acca21.org.cn

物是指具有潜在生物毒性、环境持久性、生物累积性等特征,对生态环境和人体健康可能存在较大风险,但目前尚未纳入管理或现有管理措施不足的有毒有害物质。目前,国内外广泛关注的新的污染物主要包括国际公约管控的持久性有机污染物、内分泌干扰物、抗生素等,已在全球多个国家和地区的水、沉积物、大气、生物等样品中频繁被检出。我国《重点管控新污染物清单(2023年版)》已将抗生素、环境内分泌干扰物、壬基酚等14类新污染物列入重点管控范围,实施禁止、限制、限排等环境风险管控措施。

海洋作为全球水循环的主要起点和归宿,不可避免地成为各类污染物的“汇”。目前,海洋生态环境严重衰退,加之新污染物沿陆海多介质迁移,持久性有机污染物、微塑料、抗生素及其耐药基因、放射性核素等新污染物的生态风险持续加重,长期累积可能加速海洋生态环境受损。因此,开展新污染物的分析检测、环境行为、毒性识别与风险评估等研究,是加强海洋领域新污染物治理的前提条件,也是深入了解化学物质环境风险的具体特征和关键环节的重要基础,对于建立健全“禁、减、治”环境风险防控体系具有重要意义。本文重点针对全氟及多氟烷基化合物、阻燃剂、内分泌干扰物、抗生素和微塑料等新污染物在我国海洋环境的污染现状和生态效应进行分析,并与其他国家和地区的相关情况进行比较,旨在为全面开展我国海洋新污染物的陆海统筹协同治理提供技术支撑。

1 海洋环境中新污染物的污染状况

1.1 全氟及多氟烷基化合物

全氟及多氟烷基化合物(PFAS)在全球海洋环境中普遍存在,甚至在受人类活动影响较小的两极地区也被检出。Muir等^[2]总结了2010—2019年全球各海域全氟辛酸(PFOA)的分布,其中我国渤海和黄海浓度较高,中值浓度为9.05 ng/L,高于印度洋(5.79 ng/L)、北海(1.40 ng/L)、南大西洋(0.208 ng/L)、北冰洋(0.054 ng/L)、日本海(0.05 ng/L)、地中海(0.0014 ng/L)与波罗的海(0.28 ng/L),也高于我国东海(0.975 ng/L)。通

过比较不同海域水样中 $\Sigma C4 - C10$ 全氟羧酸(PFCAs)和 $\Sigma C4 - C10$ 全氟磺酸(PFSAs)的分布特征,发现污染严重区域主要集中在靠近大型工业城市的沿岸和半封闭海湾。

沉积物中PFAS的分布受到海水-沉积物界面吸附、解吸过程的影响,PFAS自身的理化性质直接影响其吸附行为和分配能力^[3]。一般而言,随着氟化碳链长度的增加或氟化程度的提高,其疏水性增强,也更易吸附到沉积物表面。因此,长链PFAS在沉积物中的检出率和含量普遍较高。Zhong等^[4]分析了我国东海和黄海沉积物中的31种PFAS,发现在检出率高于10%的19种PFAS中,全氟辛磺酸(PFOS)、PFOA等10种长链PFAS的含量显著高于全氟丁酸(PFBA)等4种短链PFAS,另外,以六氟环氧丙烷二聚体羧酸(HFPO-DA)为代表的5种新型PFAS被检出。海洋沉积物中PFAS的空间分布特征与海水相似,大致呈现近岸河口含量高、远洋浓度低的特点。例如,我国渤海的小清河口、东南沿岸和长江口的PFAS含量较高,19种PFAS的总含量($\Sigma_{19}PFAS$)分别为4.67 ng/g、4.74 ng/g和2.56 ng/g,且从河口到开阔海域的PFAS浓度呈现明显下降趋势^[4-5]。

在我国近岸海域,PFOS和PFOA等传统PFAS呈较高浓度。近年来随着PFAS生产逐渐转向短链同系物和新型替代品,新型PFAS的检出率与浓度水平逐年上升,甚至达到与传统PFAS相当的水平。全氟烷基次膦酸(包括6:6和6:8 PFPIAs)、全氟丁基磺酰胺(FBSA)和HFPO-DA等新型PFAS的检出与当地PFAS的生产和使用密切相关。Zhong等^[4]在我国东海和黄海沉积物中首次检出FBSA和HFPO-DA,其中HFPO-DA检出率达到100%,含量(0.0180~0.450 ng/g)略高于传统PFOS(0.00200~0.558 ng/g)。不同海域的污染程度与当地PFAS生产使用情况密切相关,山东作为我国主要氟化工产业的所在地,其近岸海域中新型PFAS的污染较为显著。研究表明,渤海海域HFPO-DA的检出率为100%,海水平均浓度为2.00 ng/L^[6],莱州湾海水中6:2氟调羧酸(6:2

FTCA)的平均浓度高达 5.55 ng/L,超过全氟癸酸(PFDA)等部分传统 PFAS^[7]。值得关注的是,F-53B 作为我国自主研发的铬雾抑制剂,已替代 PFOS 在电镀行业广泛使用了 40 多年,其主要成分氯代多氟烷基醚磺酸盐(6:2 Cl-PFESA)目前仅在我国生产和使用,在莱州湾海水中 6:2 Cl-PFESA 的浓度为 1~10 ng/L,在其他国家和地区的海水中则很少或仅痕量检出^[4]。

1.2 阻燃剂

对阻燃剂类新污染物的研究表明,使用较多的多溴联苯醚(PBDEs)及新型非多溴联苯醚(non-PBDEs)类阻燃剂,如六溴环十二烷(HBCDs)、1,2-二(2,4,6-三溴苯氧基)乙烷(BTBPE)、十溴二苯乙烷(DBDPE)等均在海洋环境中被检出。海水中 HBCD 浓度一般较低,变化范围为 0.002~0.03 mg/L,这与入海河流及河口水体的浓度基本一致。吴限等^[8]调查了黄海北部近岸水体中 HCBd 分布,浓度范围为 0.12~2.23 ng/L,其非对映异构体组成特征表明,海水中 HBCDs 可能来源于沿岸工业用 HBCDs 商品。Möller 等^[9]调查了 33.23°N—84.5°N 范围内海水中得克隆(DP)的分布,其浓度范围为 0.006~0.4 pg/L,且随纬度增高而明显降低,这与其在低纬度沿海附近的排放有关,如朝鲜半岛海域及珠江口浓度远高于极地区域。海水中短链氯化石蜡(SCCPs)的分布也易受陆源输入影响,我国东海近岸海域 SCCPs 的浓度远高于辽东湾^[10]。

多数阻燃剂类新污染物疏水性较强,易于与颗粒物结合而沉降于海底。在东海表层沉积物中,SCCPs 的含量为 5.8~64.8 ng/g(干重),随着离岸距离的增加其含量逐渐下降,SCCPs 的空间分布特征表明,SCCPs 可通过大气或洋流进行区域或全球远距离传输^[11]。SCCPs 在珠江流域水体、沉积物和生物体内分布受辛醇-水分配系数(K_{ow})影响,沉积物中 SCCPs 以 Cl 8—10 同族体为主,水体中主要是 Cl 5—7 同族体^[12]。

1.3 内分泌干扰物

全球近岸海域均有检出 EDCs,如类固醇激素、双酚 A、烷基酚、个人护理品等。我国近岸

海域水体中以酚类 EDCs 为主,占 EDCs 总量的 78.94%~96.24%,双酚 A(BPA)检出率为 100%。表 1 列出了不同国家和地区近岸海域重点关注的 EDCs 浓度分布。我国近岸海域普遍受到 EDCs 的污染,尤其是类固醇激素如雌酮(E1)、雌三醇(E3)的最高浓度分别达到 204.4 ng/L 和 278.4 ng/L^[13]。烷基酚类呈现季节变化,即壬基酚(NP)和叔辛基酚(OP)的检出率在冬季约为 90.0%,而夏季仅为 59.4%和 3.1%。同时,我国沿海水域的 EDCs 呈现显著的空间变化。南海水域中 EDCs 总浓度显著高于其他地区,冬季 EDCs 平均总浓度以南海最高,其次为东海、渤海与黄海。南海与东海 NP 浓度高于其他海域^[14],渤海海域 OP 浓度显著高于其他海域^[15]。E1 存在于渤海和黄海近岸海域,E1 平均浓度也以南海最高,其次为黄海、渤海与东海^[15-16]。E3 和 EE2 仅在黄海、东海、南海海域水体中检出^[13]。

在我国海洋沉积物中 EDCs 以酚类为主,包括壬基酚、辛基酚和双酚类化合物,其中 NP 占比最高,含量范围为 n.d.~1642.8 ng/g dw,低于美国一个数量级(13700 ng/g)。3PA 在所有沉积物样品中被检出。其次是个护理品类,其中二苯甲酮-3(BP-3)和对羟基苯甲酸甲酯(MeP)含量较高。雌激素类化合物在沉积物中含量较低,以雌酮(E1)和己烯雌酚(DES)为主。除了需关注母体 EDCs 外,其环境转化产物在海洋环境中也具有一定污染水平,如珠江口沉积物中对羟基苯甲酸酯代谢物的含量明显高于其母体对羟基苯甲酸酯^[14]。

1.4 抗生素

近 10 年来,海水中大多数抗生素浓度为 ng/L 量级,但在某些海域如海水养殖密集区,其浓度已攀升至 $\mu\text{g/L}$ 量级^[33-36],尤以磺胺类和大环内酯类抗生素最为普遍,特别是位于渤海莱州湾的海水养殖区周边,甲氧苄啶的浓度高达 332.44 $\mu\text{g/L}$,同时,磺胺地索辛、磺胺二甲嘧啶、恩诺沙星和土霉素等抗生素的浓度也达到了 $\mu\text{g/L}$ 量级,其中兽用抗生素占主导地位,这主要归咎于养殖废水的排放^[33]。值得注意的是,即使是南极地区同样存在抗生素污染,其中环丙沙

表 1 不同国家和地区近岸海域水体中 EDCs 的浓度水平(单位:ng/L)

Tab.1 Concentration of EDCs in coastal waters of different countries (Unit: ng/L)

国家/ 地区	类固醇				酚类				药品及个人护理品								参考文献	
	E1	E2	E3	EE2	NP	OP	BPA	BP-3	BP-1	BP-4	MeP	EtP	PrP	BuP	BzP	TCS		TCC
中国	n.d. ~ 204.4	n.d.	n.d. ~ 278.4	n.d. ~ 120.7	n.d. ~ 41.6	n.d. ~ 68.1	20.9 ~ 1350.0	2.21 ~ 36.65	0.58 ~ 2727	2.91 ~ 27.80	1.41 ~ 53.4	n.d.	0.44 ~ 69.9	NR	n.d.	2.64 ~ 96.5	n.d. ~ 1.95	[13, 17-18]
希腊	n.d.	n.d.	n.d.	n.d.	n.d. ~ 346	n.d. ~ 18.2	10.6~52.3	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	[19-20]
日本	n.d. ~ 12	n.d. ~ 1.2	NR	NR	37 ~ 1100	4.2 ~ 290	31 ~ 2400	NR	NR	NR	2.59 ~ 17.8	n.d. ~ 0.132	n.d. ~ 2.84	n.d. ~ 34.0	n.d.	NR	NR	[13-14]
新西兰	0.02 ~ 103.9	0.4 ~ 175	n.d.	0.1 ~ 106.3	0.04 ~ 1240	<0.04 ~ 834.8	<0.08 ~ 2470	<0.01 ~ 603	2.11 ~ 280	NR	1.1 ~ 62	<0.3 ~ 15	<0.5 ~ 128	<0.2 ~ 7.1	NR	0.008 ~ 99.3	NR	[15]
南极洲	<7.0	NR	n.d. ~ <2.0	n.d. ~ <1.4	NR	0.3 ~ 1.8	<1.3 ~ 29.5	12.0 ~ 88.4	<0.8~10.3	NR	2.1 ~ 62	<0.3 ~ 15	<0.5 ~ 7.9	<0.2 ~ 7.1	NR	n.d. ~ 1.7	NR	[16, 21]
意大利	<1.2 ~ 10	<1.0 ~ 175	NR	<0.8 ~ 34	<0.5 ~ 211	NR	<1.0 ~ 145	54.5 ~ 5607	5.2 ~ 600	NR	NR	NR	NR	NR	NR	NR	NR	[22-23]
葡萄牙	2.2 ~ 13.7	8.1 ~ 21.7	NR	9.6 ~ 33.2	17.2 ~ 3166	14.5 ~ 617	33.5 ~ 129	NR	NR	NR	NR	NR	NR	NR	NR	NR	NR	[17]
美国	n.d. ~ 112	n.d. ~ 93	n.d. ~ 51	n.d. ~ 831	n.d. ~ 40000	NR	n.d. ~ 12000	n.d. ~ 9500	NR	NR	0.312 ~ 45.5	n.d. ~ 0.459	n.d. ~ 3.52	n.d. ~ 0.755	n.d.	NR	NR	[13, 17-20]
西班牙	<LOD	<LOD ~ 120	<LOD ~ 480	<LOD ~ 280	<29 ~ 60	<1.2 ~ 1.5	<1.1	NR	NR	NR	5.1 ~ 21	<0.3 ~ 1.6	<0.5 ~ 1.6	<0.2 ~ 0.7	<0.2	NR	NR	[24-25]
德国	0.1 ~ 4.1	0.15 ~ 3.6	NR	0.1 ~ 5.1	0.3 ~ 84	0.02 ~ 18	n.d. ~ 249	NR	NR	NR	NR	NR	NR	NR	NR	0.001 ~ 6.87	NR	[24, 26-28]
新加坡	<1 ~ 304	NR	<3 ~ 451	NR	20 ~ 2760	n.d.	n.d. ~ 2470	NR	NR	NR	NR	NR	NR	NR	NR	<0.55 ~ 10.5	NR	[24, 29]
巴西	<0.07 ~ 279.5	NR	<0.08 ~ 67.4	<0.39 ~ 4390	NR	NR	NR	NR	NR	NR	0.11 ~ 0.98	0.38 ~ 9.7	0.70 ~ 7.9	1.90 ~ 11	NR	<0.70 ~ 66	NR	[30-31]
突尼斯	NR	NR	NR	NR	NR	NR	NR	16.4 ~ 66.9	n.d.	n.d.	17.6 ~ 31.9	7.2 ~ 37.7	n.d. ~ 18.8	n.d. ~ 17.1	NR	NR	NR	[32]

注: n.d. 为未检出; NR 为未提及

星浓度范围为 4~218 ng/L^[37], 表明抗生素具有在海洋中远距离迁移的能力。

我国渤海、黄海、东海和南海海水中均有抗生素检出, 其中, 东海海域检出的抗生素种类最多, 共检出 43 种, 涵盖了磺胺类、氟喹诺酮类、大环内酯类、四环素类、 β -内酰胺类等多种抗生素^[38]。莱州湾不仅抗生素检出浓度在全国最高, 部分抗生素如甲氧苄啶(322440 ng/L)和磺胺地索辛(42550 ng/L)等的检出浓度更是高出其他海域 2~5 个数量级, 这可能与其紧邻海水养殖区的地理位置有关^[33]。南海海域的抗生素检出浓度也较高, 其中土霉素的最高浓度达到了 15163 ng/L^[36]。

海洋沉积物中抗生素的含量大多为 ng/g 量级, 最常见的为氟喹诺酮类和四环素类抗生素, 如氧氟沙星、诺氟沙星、土霉素和四环素等。波罗的海南部由于邻近河流集水区, 农业用地占比较多且旅游业发达, 导致抗生素含量较高, 其中土霉素含量高达 1515 ng/g, 是目前报道的沉积物中最高的抗生素含量^[39]。在北极海域沉积物中检出了较高浓度的环丙沙星(685 ng/g)及在挪威使用最多的氟喹诺酮类抗生素(92%)^[40], 显示出了抗生素污染分布的广泛性。人口密集区域、海水养殖场和污水处理厂邻近的海洋沉积物中抗生素含量普遍较高^[38-39, 41]。此外, 抗生素的结构也可以影响抗生素与沉积物间的相互作用, 疏水抗生素($\log K_{ow} > 2$)通常更易被沉积物吸附^[42], 如我国东南沿海海湾沉积物中检出含量最高的抗生素就是疏水的磺胺甲恶唑, 其含量范围为 38.3~1260 ng/g, 主要来源于养殖废水^[41]。

1.5 微塑料

全球海洋环境中的微塑料污染普遍存在(表 2), 包括南北极海域和全球各大洋沿岸^[43]。太平洋、大西洋和印度洋的副热带环流为微塑料主要聚集区, 北极海域的微塑料污染也较为严重^[44]。此外, 全球海洋微塑料主要以颗粒、碎片、纤维、薄膜、微珠和泡沫等形态存在^[45]。全球海洋微塑料的主要颜色为透明、黑色、白色、红色、黄色、蓝色、绿色和其他(包括灰色、棕色、紫色、粉色、半透明和橙色等)^[46], 其中白色、透明最为常见^[47]。在全球海洋中, 聚乙烯

(PE)、聚丙烯(PP)、聚苯乙烯(PS)和聚酯(PET)是最常见、最丰富的微塑料聚合物类型, 其次是聚氯乙烯(PVC)、尼龙(PA)等^[48]。

表 2 部分海域海水中微塑料丰度概况

Tab.2 Overview of the abundance of microplastics in global and domestic seawater

地点	微塑料丰度/items·m ⁻³	参考文献
西太平洋	(201 ± 134)	[49]
地中海(亚得里亚海)	147500 items·km ⁻²	[50]
黄海	(279 ± 140.12)	[51]
桑沟湾	(20060 ± 4730)	[52]
渤海	(114.61 ± 34.69)	[51]
渤海湾	2000 ~ 4600	[53]
南海	(103.4 ± 98.3)	[54]
中沙环礁表层	n.d. ~ 160	[54]
东海	(71.15 ± 27.92)	[51]
长江口	(2984.7 ± 2219.3)	[48, 55]
杭州湾	0.77 ~ 9.6	[56, 57]

注: n.d.为未检出

我国近岸海域和陆架边缘海均检测到微塑料^[58], 与全球海域的微塑料含量相比处于中低水平(表 2)。近岸海域微塑料污染水平通常高于外海^[52], 例如, 渤海近岸和近海均存在较高浓度微塑料污染^[51]。尽管目前已有大量研究, 但仍有较多不足之处, 例如, 超过 40% 的记录未提供微塑料聚合物类型的光谱学信息^[59]。此外, 统一且规范化的采样和样品预处理技术的缺失, 制约着污染状况的准确评估。研究表明, 采样和预处理过程中使用不同尺寸的筛网会显著影响微塑料丰度数据。例如, 使用 0.33 mm 而非 0.05 mm 的筛网采集水样时, 可能导致近 10⁴ items/m³ 的微塑料被低估^[53]。

海洋中的微塑料通过扩散、悬浮、再悬浮、沉积和掩埋等途径在海水和沉积物间迁移, 其中沉积物也被认为是微塑料重要的赋存“汇”。基于全球 186 个海洋保护区的调查发现, 海洋沉积物中微塑料丰度范围为 n.d. ~ 9187.5 items/kg^[60]。在与全球海域沉积物中微塑料的丰度比较时, 我国沉积物中的微塑料丰度大致处于中等水平(表 3)。

表 3 海洋沉积物中微塑料丰度

Tab.3 The abundance of microplastics in marine sediments

地点	微塑料丰度/ items·kg ⁻¹	参考文献
全球海域	北冰洋	854 ~ 1369 [61]
	大西洋	(67 ± 76) [62]
	南太平洋	333 ~ 33300 [60]
	印度西海岸	(220 ± 50) [62]
我国海域	东海	(663 ± 582) [51]
	黄海	(646 ± 806) [63]
	渤海	(273 ± 131) [51]
	南海	(73 ± 21) [51]

2 新污染物的海洋生态环境效应

2.1 海洋生物体中新污染物的蓄积

海洋环境中的新污染物可通过摄食、体外暴露等途径进入海洋生物体内,并持久存在和蓄积,进一步随食物链和食物网产生生物放大效应。大量监测研究表明,海洋生物体内已存在不同程度的新污染物,其浓度水平取决于化合物种类、环境污染程度、生物类型和组织器官等多种因素。同时,新污染物还能在营养级之间、亲代和子代之间产生传递,导致其环境污染与影响范围从时间和空间上进一步扩大。

(1)全氟及多氟烷基化合物

以 PFAS 为例,其在生物个体层面的检出水平一般被认为由化合物的蓄积能力和生物暴露水平共同决定。长链 PFAS 的蓄积能力一般大于短链同系物,例如, Martín 等^[64]使用棘皮动物海参(*Holothuria tubulosa*, Gmelin, 1791)评估了 PFBA、PFHxA、PFOS 等 6 种 PFAS 的生物蓄积性,发现 PFCAs 的生物蓄积性随全氟烷基链长的增加而增加。此外,PFAS 的官能团结构也影响其生物蓄积能力,例如,含有磺酸基团的 PFOS 生物蓄积因子(bioaccumulation factor, BAF)为 4.39,显著高于相同碳原子数的羧酸基 PFOA (BAF=3.59)。另外,生物个体的暴露水平主要取决于生物的摄食习性和生存环境。底栖生物主要通过摄入沉积物颗粒暴露于长链 PFAS,而浮游生物则主要暴露于周边海水环境中较高浓度的短链 PFAS。Lee 等^[65]研究了以贻贝(*Mytilus edulis*)和牡蛎(*Crassostrea gigas*)为代表的底栖

生物,其体内检测出的 PFAS 主要为全氟十一烷酸(PFUnDA)、全氟十二烷酸(PFTrDA)和全氟十四烷酸(PFTeDA)等长链 PFCAs,与所处环境沉积物中的检出结果基本一致,PFTrDA 含量最高,约为 3.13 ng/g dw。Khan 等^[66]在浮游生物体内则主要检测到 PFBA、全氟戊酯(PFPeA)、全氟庚酸(PFHpA)等短链 PFCAs,碳原子数大于等于 9 的长链 PFCAs 仅占 10%,这可能是由于短链 PFAS 倾向于进入水相环境,从而在浮游生物体内造成高富集量。

同一生物体内,不同组织的 PFAS 浓度水平可能存在显著差异。由于 PFAS 具有亲水和疏水的两性特征,且与蛋白质结合能力较强,因此在肝脏、血液等蛋白质含量高的组织中蓄积浓度较高。Zhang 等^[67]分析了搁浅东亚江豚(*Neophocaena asiaeorientalis sunameri*)的各种组织器官,检测到 PFOA 等 17 种 PFAS,其中 10 种在肝脏中浓度最高,尤其是 PFUnDA、PFOS、6:2 Cl-PFESA 含量分别为 48.2 ng/g ww、121 ng/g ww、69.3 ng/g ww,其次在心脏、肠道、肾脏中含量较高,而在皮肤、脾脏、胃、肺和肌肉中最少。Falk 等^[68-69]发现在全氟壬酸(PFNA)、PFOA、PFBS、PFOS 和全氟己烷磺酸(PFHxS)共 5 种 PFAS 的摄食暴露实验中,虹鳟鱼(*Oncorhynchus mykiss*)肝脏的污染物浓度最高(平均约为 404 ng/g),其次是血液(平均约为 263 ng/g)(图 1)。

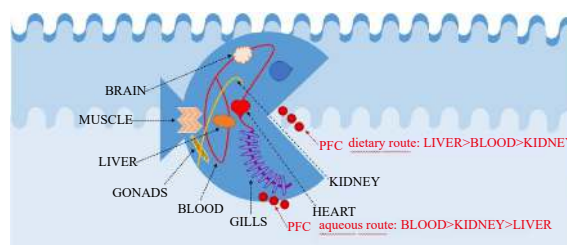
图 1 鱼类摄取 PFAS 后的典型组织分布特征^[69]

Fig. 1 Scheme of preferred tissues for the biodistribution of PFCs in fish^[69]

除传统 PFAS 以外,海洋生物体内同样能检测到各种新型 PFAS。例如,Chen 等^[70]对我国渤海的海湾扇贝(*Argopecten irradians*)、章鱼(*Octopoda*)等 6 种底栖生物进行检测,发现 PFOS(0.238 ng/g ww)与其替代品 6:2 Cl-

PFESA的平均含量(0.157 ng/g ww)相近。Li等^[71]在莱州湾小清河口的浮游生物、甲壳类、鱼类等5类海洋生物体内检测到以六氟环氧丙烷三聚体羧酸(HFPO-TA)和全氟-2-甲氧基乙酸(PFMOAA)为主的PFOA替代品, HFPO-TA含量(37.3 ng/g dw)和PFMOAA含量(3412.14 ng/g dw)远高于PFOA含量(6.064 ng/g dw)。F-53B、HFPO-TA等替代品为我国独有,国外相关数据较少,需要重点关注这些物质在我国海域海洋生物内的蓄积情况。

(2)阻燃剂

海水养殖中大量使用聚苯乙烯塑料导致养殖水域及养殖海产品面临较高的阻燃剂暴露风险。Kakimoto等^[72]分析了日本市面采购的鱼类和海洋哺乳动物膳食鱼油中HBCD的含量,检出率达到68%,浓度为<0.9~67 ng/g(脂干重);对于直接以聚苯乙烯泡沫为附着点生长的贻贝,其聚苯乙烯内部的HBCD通过迁移大量地富集在贻贝体内,其含量高达5160 ng/g(脂干重),比附着在金属和岩石的贻贝体内约高出2个数量级^[73]。同时,受日本周边海域表层和深层海水污染等影响,该水域鱼类提取的沙丁鱼油和鲨鱼鱼肝油产品中也检出较高含量的HBCD。此外,其他阻燃剂类新污染物,如DBDPE、DPs、BTBPE、六溴苯(HBB)等在我国西沙永兴岛海域鱼体中也被检出,其中DBDPE和DPs是鱼体内最主要的卤代阻燃剂,两者的平均贡献值均大于80%^[74]。

部分阻燃剂具有生物富集能力,研究表明 α -六溴环十二烷(α -HBCD)在Gal Island附近海湾贻贝和牡蛎中的生物浓缩系数最高,分别达到 1.03×10^6 和 1.08×10^6 ^[75]。Tomy等^[76]通过用DPs投喂虹鳟鱼,研究了其在生物体内的富集情况,结果表明,syn-DP在除肝脏以外的鱼体组织内易产生富集,这与前文讨论的PFAS更易富集于肝脏不同。此外,anti-DP在水生动物中的降解速率高于syn-DP,且这种现象随着营养级的增大而更加显著,这说明高级生物对syn-DP具有更好的富集性。在大型海藻体内,DPs的平均含量为(4.84±1.25) ng/g(湿重),显著高于温尼伯湖的白眼鱼[(0.73±0.12) ng/g 湿重]、金目鱼[(0.76±0.17) ng/g 湿重]、浮游动物[(0.55±0.04) ng/g 湿

重]^[77]。海藻对DPs有超强的富集能力,富集系数达到21200,主要来源于表层海水以及底泥的综合作用^[78]。SCCPs和MCCPs具有生物蓄积性,辽东湾近海海洋动物中SCCPs的浓度为86~4400 ng/g(干重),以C10Cl5和C11Cl5等低氯代低碳原子数组分为主,其对数生物蓄积因子(log BAFs)为4.1~6.7,生物体-沉积物累积系数(BSAFs)为0.1~7.3^[79]。Huang等^[12]分析珠江口海域海洋生物中SCCPs的浓度为376.3~8596 ng/g(脂重),也存在生物累积和沿食物链放大现象。Gustafsson等^[80]在波罗的海采集的蓝贻贝中BDE-47、BDE-99和BDE-153的生物累积因子BAF分别为1300000 L/kg、1400000 L/kg和220000 L/kg。在荷兰沿海及Schelde河口采集的蓝贻贝中BDE-99和BDE-100的log BAF为9.0, BDE-28的log BCF为7.4, BDE-47的log BCF为8.4, BDE-153的log BCF为8.2。在我国华南电子废弃物回收厂采集的螺、对虾、鱼和水蛇中PBDEs的log BAF范围为2.9~5.3^[81];在珠江附近某电子垃圾回收厂采集的鱼类中BTBPE、PBEB和HBBZ的BAF分别为2090~1200000 L/kg、2040~347000 L/kg和525~12300 L/kg。另一项研究发现,珠江鱼类体内DBDPE的脂肪当量BAFs为5010000~12600000 L/kg^[82]。

(3)其他新污染物

EDCs在全球沿海生物中也普遍被检出,如鱼类、贝类、甲壳类、海洋哺乳动物等。我国北部湾海域的生物样品中均检出防腐剂对羟基苯甲酸甲酯^[21],其总浓度为0.21~14.04 ng/g ww,尤其在蓝圆鲈(*Decapterus maruadsi*)体内含量最高;除母体外,其代谢产物在南海贝类生物体内呈现出较高水平,总含量高达8190 ng/g^[22]。我国渤海沿海海域软体动物中也检出双酚类和二苯甲酮类EDCs,其中BPA和BPF超过总浓度的90%;胶州湾EDCs对软体动物、甲壳类动物和鱼类构成高风险($RQ > 1$),对藻类构成低至中度风险($RQ < 1$),同时针对EDCs的浓度和检出频率筛查研究结果表明,17 α -炔雌醇、雌酮、三氯卡班、三氯生和17 β -雌二醇是该海域优先管控的高风险EDCs^[23]。

对于抗生素而言,磺胺甲恶唑、环丙沙星和恩诺沙星是海洋生物体中最常检测到的抗生素,特别是氟喹诺酮类,由于具有较高的生物蓄积性,其含量普遍高于其他类别的抗生素^[33]。韩国海水养殖水产品中的恶唑酸(1~3341 ng/g)是检出含量最高的抗生素,其他抗生素含量也较高,达到了 $\mu\text{g/g}$ 水平^[34]。珠江口海域水产品中也发现了较高含量的氟喹诺酮类和大环内酯类抗生素,尤其是诺氟沙星(256 ng/g)和阿奇霉素(366 ng/g)^[36],莱州湾水产品中的恩诺沙星高达270 ng/g^[35]。研究表明抗生素在生物体中的积累存在组织特异性,在野生海鱼中,氟喹诺酮类抗生素主要积累在肌肉中,而大环内酯类抗生素则在肝脏中蓄积量较高^[35]。例如,根据Liu等^[83]的研究,野生海鱼通过接触和摄入抗生素,甲氧苄啶、磺胺类和氟喹诺酮类抗生素主要积聚在肌肉中,大环内酯类抗生素主要积聚在肝脏中。Zhang等^[84]研究表明在部分近海鱼类样本中,抗生素诺氟沙星、脱水红霉素和罗红霉素的生物积累因子(BAF)高于5000 L/kg,表明它们具有生物积累性,在珊瑚礁鱼类食物网中,营养级放大因子(TMFs)计算结果表明依诺沙星经历了显著的营养放大。总体而言,抗生素在海洋环境中会通过食物链和食物网传递,可能导致高营养级生物体内积累抗生素,并威胁它们的健康和繁殖能力。

对于微塑料而言,研究发现不论海洋生物体的地理来源如何,其体内的微塑料水平在营养级或分类群体之间差异较大^[85]。例如,Hermabessiere等^[86]对法国近岸海域的贻贝(*Mytilus edulis*)和角皮藻(*Cerastoderma edule*)研究发现,其体内的微塑料数量浓度分别为(0.76±0.40) items/ind和(2.46±1.16) items/ind。较大型的脊椎动物体内呈现更高的微塑料生物累积量,如我国杭州湾的研究发现,赤鼻棱鳀(*Thryssa kammalensis*)体内微塑料丰度最高(22.21±1.70 items/ind),远高于黄海的小型鱼(2.6 items/ind)^[48,87]。另外,浮游动物体内微塑料累积水平通常较低,主要与其个体、口器大小等有关。另外值得注意的是,海洋生物主要摄入纤维状微塑料,微塑料纤维的长径比越大,越易滞留在生物体内^[88]。主要摄入微塑

料的颜色为蓝色、黑色、透明或白色等,尺寸多为<500 μm ^[89],且无论微塑料的形状和尺寸如何,PS和PP是最常见的聚合物类型,其次是PET和PA^[85]。

2.2 新污染物的海洋生态效应及环境风险

新污染物造成的海洋污染已经成为全球性问题,较高的稳定性和生物蓄积性使得新污染物极易在生物体内蓄积,可能引起神经毒性、免疫毒性、发育毒性和致死效应等多种负面影响,对海洋生态安全构成威胁。

现有毒理学研究表明,PFAS暴露会对水生生物个体产生肝脏、免疫、代谢、内分泌、生殖、生长发育等多种毒性作用,极端情况下还可导致生物死亡。针对弓背青鳉(*Oryzias latipes*)的研究发现^[90],F-53B和聚苯乙烯纳米塑料(PS-NPs)的联合暴露引起该生物体内超氧化物歧化酶(SOD)和过氧化氢酶(CAT)活性增加。PFOS通过抑制编码转铁蛋白和VI类乙醇脱氢酶相关基因表达,表现出比PFOA更强的肝脏毒性^[91]。Wood等^[92]在北太平洋海域的观测结果显示,PFUnA、PFNA、PFBS和PFDA等10种PFAS会诱发绿海龟(*Chelonia mydas*)和印太玳瑁(*Eretmochelys imbricata*)的免疫抑制,主要表现为免疫球蛋白M(IgM)抗体水平下降。PFOA可以通过上调半滑舌鳎(*Cynoglossus semilaevis*)肝脏、肌肉、肠等组织中热休克蛋白HSP70的表达产生免疫毒性^[93],PFOS可诱导宽吻海豚(*Tursiops truncatus*)CD4+和CD8+两种T细胞增殖,并促进 γ -干扰素等促炎细胞因子的产生,增强宽吻海豚的免疫反应,造成免疫系统失调^[94]。Ishibashi等^[95]发现6:2 FTOH和8:2 FTOH能显著增加成年雄性日本青鳉(*Oryzias latipes*)的卵黄原蛋白(vtg)基因遗传表达和雌激素水平,导致性激素代谢紊乱。PFOS可上调大西洋鳕鱼(*Gadus morhua*)体内脂肪酸代谢相关基因的表达,从而提高脂质代谢水平^[96];Li等^[97]发现PFOA暴露严重影响蓝贻贝(*Mytilus edulis*)体内相关降解通路,从而阻碍了PFOA自身的代谢和清除,引起PFOA的生物蓄积。PFOS和PFOA可干扰日本青鳉的性腺功能^[98],显著降低其繁殖能力^[99],PFOS暴露可能在青鳉孵化早期

诱导孵化酶(HCE和LCE)活性和水平提高,导致孵化时间缩短、孵化率增加,但孵化后幼鱼的存活率下降^[100],PFTTrDA暴露也可诱导雌性青鳉幼鱼的肝脏过度生长,产生氧化应激损伤,增加肿瘤抑制基因P53的表达,导致细胞凋亡和F1胚胎畸形^[101]。

已有研究表明,多种阻燃剂(如TBBPA、SCCPs等)均具有胚胎毒性、肝脏毒性、神经毒性、内分泌毒性、多代及遗传毒性等多种效应,主要表现为胚胎的致畸、生长周期延长以及胚胎心率的下降等;对肝脏主要表现为对酶活性、肝脏的组织结构形态的影响以及诱导肝脏中相关基因的表达等;对神经细胞主要表现为突触变短、数量减少,最终影响受体结合,阻碍神经递质的传导等。对于海洋无脊椎动物而言,HBCD可能引起细胞损伤以及内分泌系统的紊乱和能量代谢的改变,从而影响其发育和繁殖^[102]。从代谢水平上看,不同物种不同组织器官对HBCDs的异构体具有不同的富集能力和净化速率, γ -HBCD和 β -HBCD在黄姑鱼(*Nibea albiflora*)体内具有较高的净化速率和代谢能力^[103]。肝脏中HBCDs的代谢能力最强,肌肉中最弱,这将会导致不同生物对HBCDs的毒性耐受程度不同^[104]。Koh等^[105]对微生物的毒性研究表明,氯含量为62%的SCCPs对生物发光细菌的最低可见效应浓度(EC_{20})为0.05 mg/L;SCCPs对青鳉晶胚的毒性机理为麻醉,其 $LOEC$ 为55~460 μ g/L。

鱼类作为海洋EDCs生态毒理效应和机制研究的模式生物,发生在鱼类发育关键时期的EDCs暴露,可引起鱼类性腺发育异常、生长畸形、后代繁殖数量减少以及性别比例失衡,进而导致鱼类种群数量严重下降,广泛影响生物的生长、发育、免疫等适应能力,引起进化潜能的丢失,最终导致生物多样性丧失^[106]。此外,EDCs引起的鱼类亲代不利影响和生理功能异常还会跨世代传递给子代^[107]。这种跨世代毒性效应会影响子代的存活能力、生长发育水平和生理功能,改变种群的性别比例,损害子代捕食、求偶和躲避敌害等行为能力,从而对种群发展和生态系统健康产生深远影响^[108-109]。除了EDCs自身对海洋生物的负面影响之外,还需引起重视的

是,EDCs的环境转化产物与代谢物可能具有同样甚至更为严重的危害^[110-111]。例如,对羟基苯甲酸酯在环境转化过程形成的二聚物具有比原型更强的雌激素效应^[112]。除了通常关注的游离态EDCs外,结合态EDCs的生态效应也不容忽视,例如,酶水解生物样品中代谢物的比例(38%~79%)高于非酶水解生物样品中的代谢物比例(2.9%~65%)。EDCs的毒性作用过程和机制极为复杂,具有生物发育阶段的特异性(胚胎阶段更为敏感)、毒性效应的非经典性(低剂量暴露具有更高毒性)和协同/拮抗性(不同于单个EDC所产生的效应)等特点^[113],目前EDCs毒性作用机制大多针对哺乳动物,通常认为其通过受体-介导途径与直接影响激素合成酶等途径发挥内分泌干扰作用。然而,EDCs对海洋生物特别是海洋无脊椎动物的毒性作用机制仍有待进一步阐明,在无脊椎动物食物网中,大多数合成类固醇类EDCs具有生物富集性和随食物链的传递性。例如,17 α -甲睾酮沿食物链的传递呈现生物放大作用,17 β -勃地酮则呈现生物稀释现象,而这两种EDCs的结合态代谢物却在整个食物网中均呈现生物稀释现象^[114]。海洋软体动物如贻贝往往具有对酚类(双酚A、壬基酚、辛基酚)等EDCs的高生物累积性,因此很可能将EDCs沿食物链传递至更高营养等级的生物^[115],从而对生物体内EDCs污染水平产生放大作用。有研究表明,海洋环境中EDCs暴露不仅能够对物种、种群和生态系统水平对生物产生危害,而且通过食用受EDCs污染的海产品给人群带来的暴露风险也不容忽视。

抗生素进入海洋环境,对水生生物具有毒理效应,如在一定程度上抑制和干扰藻类生长、光合作用、大型蚤生长和繁殖能力等^[116]。Lin等^[117]研究表明,海洋环境中左氧氟沙星和诺氟沙星通过影响藻类细胞的生长、光活性和酶活性来抑制微藻的生长。Kong等^[118]研究发现,渤海中外源抗生素和抗性基因的输入可显著影响群落功能,改变海洋沉积物中的细菌群落组成。此外,海洋环境中的多种抗生素污染可与海洋酸化对扇贝造成拮抗作用,导致扇贝出现神经系统疾病^[119]。抗生素一定程度上会在基因水平或幼体

发育阶段对水生生物产生毒性影响。Pereiro 等^[120]报道了斑马鱼(*Danio rerio*)暴露在磺胺甲恶唑和克拉霉素的混合物(10 $\mu\text{g/L}$)中两周后,其免疫系统受到明显损伤,且其对病毒的易感染程度也明显增加。Lei 等^[121]研究发现,4种抗生素(磺胺二甲基嘧啶、恩诺沙星、强力霉素和氟苯尼考)通过不同的机制对斑马鱼幼体的脂质代谢产生破坏作用。Zhang 等^[122]研究表明,抗生素和海洋酸化协同通过内在和外途径可诱导扇贝(*Argopecten irradians irradians*)细胞凋亡。值得注意的是,海洋酸化和高盐度有时会增大药物(如抗生素)对海洋生物的毒性作用^[123]。海洋环境中微生物大量存在,抗生素的存在可促进 ARB 的产生,并加速 ARGs 的传播,造成全球范围内的公共卫生问题,使得原本可以治疗的感染疾病变得难以控制,增加了医疗成本并对公共健康构成威胁。耐药性的增加还可能导致新抗生素的研发速度赶不上耐药性的发展速度,He 等^[124]研究发现, β -内酰胺类、四环素类和多重药物类抗性基因在海水养殖场中经常被检测到,20%左右检测到的 ARGs 被列为高风险、当前风险和未来风险。Xu 等^[125]通过宏基因组学和宏转录组学研究表明,人类活动可能通过改变硝酸盐和磷酸盐浓度以及海洋温度来影响 ARGs 的表达。

关于微塑料对浮游植物影响的研究,大多关注微藻暴露于微塑料后的生长(如光合速率)及后续对生态系统能量流动的影响等方面^[126]。微塑料可通过摄食、呼吸和皮肤接触等途径进入生物体,并在体内转运直至排出。研究发现,微塑料能在生物体内滞留,可能对生物的生长发育、行为特征、生殖水平、免疫系统及相关基因表达产生毒性效应^[127]。此外,微塑料还会对海洋生态系统和物种群落造成危害,包括生物多样性降低、食物链中断、栖息地破坏和生态服务损失等方面。除了微塑料本身的危害外,其制造过程中的添加剂及吸附的污染物也会对生物体造成多样化的损害^[128-129]。由于许多微塑料上的共存污染物具有持久性、生物蓄积性和潜在毒性,微塑料的载体效应也被认为是亟须优先关注的生态风险之一。

3 海洋环境中新污染物治理研究展望

新污染物在海洋环境中无处不在,对海洋生态系统乃至人类健康产生深远且未知的影响,因此,迫切需要加大投入,在当前加大监测、调查力度的基础上,建议深入开展以下研究工作:

(1)流域-河口-近海主要污染物的产汇机制及智能筛查技术。研究人为影响和气候变化综合作用下的入海污染物“源-径-汇”产汇机制,辨识流域-河口-近海污染物的迁移转化过程,揭示典型流域水文特征及其污染物变化的响应机制;开发海洋环境暴露下新污染物风险源识别参数的智能筛查技术,研究适用于流域-河口-近海环境下新污染物计算毒理学效应的预测预警技术,为新污染物的海洋环境风险评估提供数据参考和科学依据。

(2)海洋生态环境多要素智能监测技术。围绕海洋污染物在线实时监测、高通量自动化、高灵敏精准监测与质控技术,突破以新污染物为主的非靶向高通量筛查、靶向精准定量与效应导向等先进智能分析技术,实现海洋环境监测技术方法和设备的规范化、标准化、系列化;构建数据“获取-传输-质控-融合-挖掘-评价”全链条数据感知与数据挖掘评价技术体系,建设完善海洋新污染物的智慧监测与评价大数据平台。

(3)陆海统筹的海洋环境质量基准和标准物质体系。围绕适用于我国流域-河口-近海污染物环境风险精准识别与科学评估的海洋环境基准制定技术,开发适用于我国海洋生物区系特征的新污染物水质基准推导与制定技术规范,构建具有河口、近岸海域环境特征的新污染物生态风险评估数据库与技术框架体系;研制海水、海洋沉积物和海洋生物体环境标准样品或标准参考物质,实现我国不同海域和海洋环境特征下关键新污染物的环境风险精准识别与确定。

(4)流域-河口-近海污染物精准溯源与全过程协同管控治理技术。建立针对流域-河口-近海的陆海联动入海路径下目标污染物的入海通量评估方法,确定污染物来源类型的定量溯源技术,区分流域、河口、海湾、近岸等不同类型海(流)域的污染物环境风险管控评价技术体系;构

建流域-河口-近海不同尺度下污染物水动力输移-环境归趋耦合的数值模型,提出流域-海域过渡区生境改善与生态修复技术方案;围绕“一湾一策”“一河一策”管理需求,基于监测、评价和风险评估,研究适用于入海河流、河口、海湾特点的标准统筹、空间统筹、机制统筹的陆海统筹差异化管控策略和实施机制,建立针对各类污染源的入海污染治理技术,制定入海污染治理主体责任清单和成效巡查评估清单,以有效支撑陆海统筹环境保护和协同治理。

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