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环境介质中多溴联苯醚(PBDEs)分布特征的研究进展*

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摘 要 由多溴联苯醚(PBDEs)所引起的环境问题越来越受到社会各界的关注,了解环境介质中PBDEs的分布特征是对其污染进行有效控制的前提.本文从非生物介质(大气、水体、沉积物、污泥及土壤)和生物体内(植物、动物、人体及食物链)两方面入手,对PBDEs在世界各地不同环境介质的分布特征进行综述.高溴代PBDEs因其较低的挥发性和水溶性,极易吸附于土壤、底泥和空气悬浮颗粒等固体介质,而低溴代PBDEs在各环境介质中都有检出;生物体中PBDEs的分布特征与生物种类、污染源、营养级以及个体组织的特异性有一定关系,人体血液、母乳、头发等也检出了PBDEs.随着北美、欧盟等地区从2004年开始逐渐对五溴、八溴和十溴PBDEs实施禁用,其含量在北美和欧盟部分地区有所下降,但是由于十溴PBDEs仍在全球很多地区使用,又因其具有持久性和长距离迁移性,要实现环境介质中PBDEs含量的控制仍需要一定时间,因此未来研究仍需对PBDEs生产和使用区域开展污染水平及趋势相关研究,并对PBDEs的环境行为深入探讨.

关键词 多溴联苯醚(PBDEs), 环境介质, 生物体, 分布特征.

Distribution of polybrominated diphenyl ethers (PBDEs) in the environment: A review

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Abstract: Environmental problems induced by polybrominated diphenyl ethers (PBDEs) are receiving more and more attention. The knowledge about the distribution of PBDEs is the premise for the effective pollution control. This paper reviewed the distribution of PBDEs in the atmosphere, water, sediments, sludge, soil, plants, animals, human body and food chain all over the world. Owing to the low volatility and water solubility, the higher brominated PBDEs can be easily adsorbed to solid particles like soil, sediment and airborne particles. However, the lower brominated PBDEs are detected in all the environmental media. In living organisms, the distribution characteristics of

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PBDEs are depended on biological species, pollution sources, trophic levels and tissue specificity, and PBDEs are also found in human such as blood, breast milk and hair. With the prohibition of penta-, octa-, and deca-BDEs in North American and some regions of European Union, the concertrations of PBDEs in the environment have declined to some degree. However, deca-BDEs is still used in other regions around the world. Due to its persistence and long-distance transport, the control of PBDEs in the environment is still a long-term work. Thus, future research needs to focus on the level, trend, and behavior of PBDEs in the regions where they are produced and used.

Keywords:PBDEs, environmental matrices, living organisms, distribution characteristics.

多溴联苯醚(polybrominated diphenyl ethers, PBDEs)是一种添加型溴代阻燃剂,被广泛应用于塑料、纺织品、电子产品、油漆等领域,因其缺乏化学键的束缚,很容易逸散到大气中,通过大气干湿沉降进入水体、土壤和生物体中,又因其具有难降解性和易于积累的特点,对生态环境构成了巨大危害[1-2].早在 1979 年,DeCarlo 等报道了美国某 PBDEs 生产厂家周围的土壤和污泥中 BDE-209 的存在[3],两年后在瑞典维斯坎河纺织厂下游鱼体内发现多种 PBDEs[4].Haglund 等在波罗的海的斑海豹、灰海豹和鲱鱼体内,健康瑞典男性的脂肪组织,以及商用鱼油等多种样品中检出四、五、六溴代 PBDEs[5].近年来,在偏远地区,比如南极地衣和苔藓样品中也检出 12 种 PBDEs[6].2003 年杨永亮等[7]在中国青岛近岸沉积物和贻贝中检出 21 种 PBDEs 单体,这是国内最早关于 PBDEs 的报道,随后 PBDEs 逐渐被报道出现在中国香港海水[8]、广州大气[9]等中国其它地区的多种环境介质中.目前 PBDEs 已在大气、水体、底泥、土壤以及多种生物介质中检出,并广泛分布于世界各地.已有研究证明 PBDEs 能在动物体内富集,并产生一定程度的健康危害,比如干扰性激素和甲状腺激素[10-12].由于低剂量的五溴联苯醚(penta-BDEs)和八溴联苯醚(octa-BDEs)就可以引起毒性[13],从 2004 年开始欧盟和北美地区逐渐禁止了 penta-BDEs、octa-BDEs 产品的生产和使用,2006 年中国限制了 PBDEs 的使用,并于 2014 年开始禁止生产、流通、使用和进出口商用 penta、octa-BDEs[14-15].尽管 2008 年欧盟和美国的一些州相继采取了禁用十溴联苯醚(deca-BDEs)的措施,但 deca-BDEs 仍在世界上许多地区使用,同时之前产品中的 PBDEs 不断释放进入环境,由此所引发的环境问题仍然值得关注[16].

由于 PBDEs 的持久性、长距离迁移性及其对生物的毒性,有必要全面认识其在全球范围内各种环境介质中的分布特征.本文总结了近些年来国内外大气、水体(包括沉积物和污泥)、土壤、植物、动物和人体等多种环境介质中 PBDEs 的分布特征,并对未来的研究方向进行展望,旨在促进 PBDEs 的控制及管理.

1　环境介质中 PBDEs 的分布特征(Distribution characteristics of PBDEs in the environmental media)

目前市场上使用最多的商品是 deca-BDEs,由于其具有低的挥发性和水溶性,极易吸附于土壤等固体介质上,大部分高溴代 PBDEs 都沉积在土壤、底泥和空气悬浮颗粒中;而低溴代 PBDEs 具有相对较高的挥发性和水溶性,在各种环境介质中都有检出.

1.1　大气中 PBDEs 的分布特征

室温下 PBDEs 的蒸汽压较低,容易挥发到大气中,并随大气长距离迁移.世界各地大气中都有不同程度 PBDEs 检出,甚至在北极、青藏高原等背景偏远的地区都有发现[17-42].PBDEs 的蒸汽压随溴代数的增加而降低,因此高溴代 PBDEs(如 BDE-209)更易与颗粒物结合,而低溴代 PBDEs(如 BDE-28、47、99)更易在大气中长距离传输[17].

在背景和偏远地区,长距离传输是其主要来源,PBDEs 单体均处于很低的浓度水平,以 BDE-47、99 为主.在北极,夏季低溴代 PBDEs 通过挥发进入,冬季 PBDEs 通过气溶胶从中纬度地区输送到北极[19],Wang 等[20]研究发现北极 ∑PBDEs(包含 BDE-209)的平均浓度(17.3 $pg \cdot m^{-3}$)高于北太平洋(12.8 $pg \cdot m^{-3}$)及其它背景地区(1.5—8.5 $pg \cdot m^{-3}$),可能是受北美大量使用 penta-BDEs 及温度效应的影响;且由于大气环流和强烈的空气混合作用,PBDEs 在北极地区并无明显的地理分异;而北太平洋地区呈现从中纬度向高纬度地区的递减趋势,可能是由于 PBDEs 在空气团输送过程中发生了稀释、沉积和

分解等作用.在南极,科学考察站的人类活动是 PBDEs 的潜在排放源,以 BDE-17、28 为主,∑PBDEs(包含 BDE-209)(0.7—3.0 pg·m^{-3})低于北极[21].大气中 PBDEs 浓度存在季节差异,夏季气温升高使 PBDEs 易于挥发和散逸,冬季降温则使其易于沉降和吸附[22],如加拿大阿勒特大气中∑PBDEs呈现夏季高于冬季的趋势,尤其是低溴代 PBDEs 的相对百分比[23].Hung 等研究表明北极大气中不同 PBDEs 单体的倍增时间差异较大(从 BDE-209 的 3.5 年到 BDE-153 的 28 年),这可能是因为 penta-BDEs 及 octa-BDEs 在北美及欧洲一些国家逐步淘汰或禁用,但是由于 deca-BDEs 产品的持续使用,BDE-209 的含量持续上升[24].

就全球而言,亚洲大气中 PBDEs 浓度最高,尤其是中国 PBDEs(包含 BDE-209)的浓度为 2.0—787 pg·m^{-3})[25,27-30]普遍高于 10 年前美国和欧洲(nd—85 pg·m^{-3})[18,31-34].Deng 等[25]研究发现广州、中国香港的 PBDEs 浓度分别为 204—372 pg·m^{-3}和 33.8—358 pg·m^{-3}.对于同一区域,大气中 PBDEs 浓度呈现:城市>乡村[29]、工业区>非工业区[26-27]的趋势.Strandberg 等研究发现美国芝加哥∑PBDEs(包含 BDE-209)(33—77pg·m^{-3})远高于乡村(4.4—21pg·m^{-3})[31].吴辉等[30]采集了中国某溴代阻燃剂生产地的大气样品,其∑PBDEs(包含 BDE-209)(16000—240000 pg·m^{-3})比广西南宁(31.4—787 pg·m^{-3})高 2—3 个数量级.电子垃圾拆解活动也是大气中 PBDEs 的重要来源[35-36],如中国最主要的电子垃圾拆解地之一广东贵屿大气中∑PBDEs(5379—47187 pg·m^{-3})是广州和中国香港城市区域(33.8—372 pg·m^{-3})的 58—691 倍[25].

室内空气中 PBDEs 含量远高于室外,也是室外空气中 PBDEs 的一个重要来源[37].室内空气中 PBDEs 主要来自电器的使用,与室内家具数量、装修程度的关系不明显;由于办公室内电器产品(如电脑、空调等)相对密集和频繁的使用,使其 PBDEs 浓度通常大于家居环境[39,43];且无论是办公还是家居环境,同一区域内不同采样点 PBDEs 分布差异均较大,这可能与电器数量、使用频率以及释放源与采样点距离等因素有关[37-41,43].Zhu 等[42]研究发现中国东部室内灰尘中∑PBDEs(2120 ng·g^{-1})高于西部(847 ng·g^{-1}),城市高于乡村,表明社会经济发展对其浓度水平影响显著.

1.2 水体、沉积物及污泥中 PBDEs 的分布特征

大气中的 PBDEs 可以通过干湿沉降迁移到水体、土壤和生物体等环境介质中,土壤中的 PBDEs 通过淋溶、径流进入水体,造成水体中 PBDEs 污染.由于 PBDEs 具有较高的辛醇-水分配系数($\lg K_{ow}$)和较低的水溶性,水体中 PBDEs 的含量相对较低[44],低溴代 PBDEs(如 BDE-28、47、99)在水中流动性更强,而高溴代 PBDEs 在污染源附近底泥中的残留量更高[45].

相关研究表明,仅包含水相的水体中 PBDEs 以 BDE-47 和 BDE-99 为主[8,46-48],而包含悬浮颗粒相的水域[49-51]以及河流沉积物和污水处理厂的污泥[46-74]中,BDE-209 含量则相对较高(表 1).中国香港、加拿大和瑞士的水体中∑PBDEs(不含 BDE-209)(nd—100.64 pg·L^{-1})和 BDE-209 浓度(nd—23.2 pg·L^{-1})均低于美国旧金山湾(15.5—337 pg·L^{-1},12.2—533 pg·L^{-1}),处于较低污染水平[8,47,51-52];而珠江口水体中 BDE-209 浓度极高(76—5693 pg·L^{-1}),主要来自河流沿岸城市中 deca-BDEs 的大量使用和排放[50].由于电子垃圾拆解活动广东清远电子垃圾拆解地水库(23.8—25.0 ng·L^{-1})和浙江丰江(22.4 ng·L^{-1})的表层水体中 PBDEs(包含 BDE-209)浓度远高于其他地区水体[53-54].也有研究表明水中 PBDEs 含量具有明显的季节变化特征,如中国东海表层海水∑PBDEs含量在冬季(2.9—5.5 ng·L^{-1})高于夏季(0.7—1.5 ng·L^{-1})[55].

BDE-209 为沉积物中最主要的单体,比如美国五大湖的沉积物中 BDE-209 占∑PBDEs的 83%—98%[56-58],中国香港红树林的底泥样品中,BDE-209 浓度(1.5—75.9 ng·g^{-1} dw)比其他单体高 1—2 个数量级[59].∑PBDEs(不含 BDE-209)在美国五大湖、韩国、加拿大、瑞士、中国长三角、中国南海、中国东海、中国香港、中国台湾等地水平相近(nd—14.4 ng·g^{-1} dw),而荷兰和珠江三角洲(0.1—114 ng·g^{-1} dw)则比它们高 1—2 个数量级,珠江三角洲(0.7—7340 ng·g^{-1} dw)和松花江(2.8—9862 ng·g^{-1} dw)的 BDE-209 浓度远高于其他地区[56-68].Marvin 等研究表明,加拿大尼亚加拉河∑PBDEs在 1980—1988 年间呈持续上升趋势,在 1988 年之后增速加快并于 1995 年达到最大值 35 ng·g^{-1}dw[69].天津大沽排污河河口 PBDEs 和 BDE-209 浓度均显示上层底泥中含量高于下层底泥,表明近几年我国环境介质中 PBDEs 含量呈上升趋势[65].

表 1　PBDEs 在表层水、沉积物和污泥中的分布

Table 1　Distribution of PBDEs in the surface of water, sediments and sludge

国家/地区 Country/Area	年份 Year	PBDEs 单体 PBDEs congeners	∑PBDEs (均值)^{b} ∑PBDEs (mean)^{b}	BDE-209(均值) BDE-209(mean)	主要单体 Main congeners	文献 Reference
表层水(pg·L^{-1})^{b}						
中国香港沿海	2005	BDE-28,47,99,100,153,156,183	nd—97.8	nd	BDE-28,47,100,183	[8]
美国密歇根湖	2004	BDE-47,66,99,100,153,154	18	na	BDE-47,99	[46]
加拿大温尼伯河	2004	BDE-47,85,99,100,153,154	9.6—100.6	nd	BDE-47,99	[47]
南极表层雪	2011—2012	BDE-17,28,47,66,71,85,99,100,153,154,183	130—340	nd	BDE-47,99	[48]
荷兰	1999	BDE-28,47,99,100,153,209	1.6	0.1—4	BDE-47,99,209	[49]
中国珠江江口	2005	BDE-28, 47, 99, 100, 153, 154, 183,209	26.1—156.9	76—5693	BDE-47,99,209	[50]
瑞士图恩湖	2007	BDE-28,47,99,100,153,183,209	17—78^{a}	12.5—23.2(15.6)	BDE-47,99,209	[51]
中国浙江丰江	2009—2010	BDE-47,99,100,153,154,183,209	22400^{a}	na	BDE-47,153,183,209	[53]
中国清远电子垃圾拆解地	2006	BDE-28,47,66,85,99,100,138,153,154, 183, 196, 197, 203, 205, 206, 207,208,209	23800—25000 (24400)^{a}	400—410	BDE-28,47,99	[54]
沉积物和污泥(ng·g^{-1} dw)^{b}						
加拿大温尼伯河	2004	BDE-47,85,99,100,153,154,209	1.2—1.6(1.3)^{a}	0.5—0.8(0.6)	BDE-47,99,209	[47]
瑞士图恩湖	2005	BDE-28,47,99,100,153,183,209	0.2—5.4^{a}	0.1—5.1(1.0)	BDE-47,99,209	[51]
美国五大湖	2001—2002	BDE-28,47,66,85,99,100,153,154,183,209	0.5—6.3	4—242.0	BDE-47,99,209	[56-58]
中国香港红树林	na	BDE-28, 47, 99, 100, 153, 154, 183,209	0.6—14.4	1.5—75.9	BDE-153,183,209	[59]
中国珠江三角洲	2002—2004	BDE-28, 47, 66, 99, 100, 138, 153, 154,183,209	0.1—94.7	0.7—7340	BDE-47,99,209	[63]
中国天津大沽排污河河口	2005	BDE-17,28,47,66,71,85,99,100,138,153,154,183,209	0.1—0.5	nd—14.9	BDE-209	[65]
中国台湾西南部河口与沿海	2005	BDE-17,28,47,66,99,100,138,153,154,183,190,205,206,209	nd—1.8	nd—6.3	BDE-209	[66]
中国松花江	2008	BDE-28, 47, 99, 100, 153, 154, 183,209	2.9—9871(397)^{a}	2.8—9862	BDE-47,183,209	[67]
中国东海	2013	BDE-15, 28, 47, 99, 100, 153, 154, 183, 209	0.03—1.3(0.3)	0.2—3.2(0.6)	BDE-47,99,209	[68]
美国加州污水处理厂	na	BDE-47,99,100,153,154,209	1918—2086	1010—1440(1183)	BDE-47,99,209	[70]
德国污水处理厂	2002—2003	BDE-28, 47, 99, 100, 153, 154, 183,209	12.5—288(126)	97.1—2217(429)	BDE-47,99,209	[71]
中国污水处理厂	2005	BDE-17,28,47,66,71,85,99,100,138,153,154,183,209	6.2—57(25.5)	<1—1108.7(68.5)	BDE-47,99,183,209	[72]
科威特污水处理厂	2005—2006	BDE-47,99,100,153,154,183,209	0.9—27.8(9.3)	4.8—1595(181.8)	BDE-47,99,209	[74]
中国珠江三角洲污水处理厂	2006—2007	BDE-28,47,66,85,99,100,153,154,183, 196, 197, 203, 205, 206, 207,208,209	158—23750^{a}	150—22894(6586)	BDE-47,99,209	[76]
捷克污水处理厂	2007	BDE-28,47,49,66,85,99,100,153,154,183,209	205—605	685—1403	BDE-47,99,209	[77]

注:a. ∑PBDEs(含 BDE-209);b.∑PBDEs(不含 BDE-209);c.中位数.

Note:a. ∑PBDEs(including BDE-209);b. ∑PBDEs(not including BDE-209);c median.

污水处理厂污泥中也存在大量 PBDEs,以 BDE-47、99、209 为主[70].美国加州污水处理厂的∑PBDEs(不含 BDE-209)(1918—2086 ng·g^{-1} dw)比其他污水处理厂(0.9—605 ng·g^{-1} dw)高 1—2 个数量级,中国珠三角污水处理厂的 BDE-209 浓度(150—22894 ng·g^{-1} dw)远高于其他污水处理厂(<1—2217 ng·g^{-1} dw)[70-77].Wang 等在中国 26 个城市 31 个污水处理厂采样,结果表明其 PBDEs 浓度(不含 BDE-209)(6.2—57 ng·g^{-1} dw)低于欧洲和北美国家(12.5—2086 ng·g^{-1} dw),BDE-209 平均占 PBDEs 总量的 55%,低于其在沉积物和自然水体中所占比例[72].

1.3 土壤中 PBDEs 的分布特征

大气和水体中 PBDEs 可迁移到土壤,其中 PBDEs 的生产、运输及其产品的使用、电子垃圾拆解活动、污水灌溉以及污泥土地利用都是土壤中 PBDEs 的重要来源,同时,土壤中 PBDEs 特别是高溴代 PBDEs 通常被认为来自于大气的干湿沉降[41],目前世界各地土壤中已检出多种 PBDEs 同系物(表 2)[26,34,73,78-88].Harrad 等在英国横跨西米兰德的城市乡村带土壤中检出的∑PBDEs(0.07—3.9 ng·g^{-1} dw)[34]与 Hassanin 等在英国和挪威(欧洲背景)林地和草地土壤中的结果(0.07—12 ng·g^{-1} dw)较为一致[78];而青藏高原的∑PBDEs(0.004—0.3 ng·g^{-1} dw)则远低于其他背景地区土壤[80,83-85].Cetin 等研究表明土耳其市区(8.7—18.6 ng·g^{-1} dw)和工业区(0.5—2840 ng·g^{-1} dw)土壤中∑PBDEs高于郊区(0.8—6.8 ng·g^{-1} dw)[26].广东清远电子垃圾拆解地 PBDEs 浓度(包含 BDE-209)(296 ng·g^{-1} dw)比农田(15.6—34.1 ng·g^{-1} dw)高 1 个数量级以上[82].污泥施用增加了土壤中 PBDEs 浓度,最显著的是 BDE-209;西班牙和美国弗吉尼亚的研究表明污泥施用土壤∑PBDEs(30—689 ng·g^{-1} dw, 0.5—140.5 ng·g^{-1} dw)远高于未施污泥的土壤(20.7 ng·g^{-1} dw, nd—11.0 ng·g^{-1} dw)[73,75].同时,土壤有机质、黏土矿物含量以及海拔高度和土壤深度对 PBDEs 在土壤中的组成和分布有一定影响,如随着土壤深度的增加逐渐降低,低溴代较高溴代 PBDEs 有更强的迁移能力[85-86];对青藏高原的研究发现,随着海拔的升高(1439—3940 m),PBDEs 含量逐渐降低,但上升到 4479 m 时其含量明显增加(4500—4900 m)[83-84].

表 2 PBDEs 在土壤中的分布(ng·g^{-1}dw)

Table 2 Distribution of PBDEs in soil(ng·g^{-1}dw)

国家/地区 Country/Area	采样地 Sampling sites	年份 Year	PBDEs 单体 PBDEs Congeners	∑PBDEs (均值)^a ∑PBDEs (mean)^a	BDE-209(均值) BDE-209(mean)	主要单体 Main congeners	文献 Reference
英国和挪威(欧洲背景)	林地和草地	1998	BDE-17,28,32,35,47,49,71,75,77,85,99,100,119,138,153,154,166,181,183,190	0.07—12(1.4) b	na	BDE-47,99	[78]
英国	城市乡村带	2003—2004	BDE-28,47,99,100,153,154	0.07—3.9 b	na	BDE-47,99	[34]
西班牙	污泥施用	2005	BDE-47,99,100,153,154,183,196,206,207,208,209	30—689	24.6—655	BDE-183,209	[73]
	对照			20.7	14.6	BDE-183,209	
美国弗吉尼亚	污泥施用	2006	BDE-28,47,99,100,153,154,183,209	0.5—140.5	nd—78.7	BDE-47,99,209	[75]
	对照			nd—11.0	nd—10.0	BDE-47,99,209	
中国广东贵屿	酸解地	2004	BDE-3,7,15,28,33,47,49,66,71,77,85,99,100,119,126,138,153,154,183,190,197,203,207,209	2720—4250(3570)	1270	BDE-209	[79]
	打印机垃圾场			893—2890(1440)	510	BDE-99,209	
	池塘			263—604(399)	328	BDE-209	
	稻田			34.7—70.9(48.2)	37.3	BDE-209	
	水库			2.0—6.2(3.8)	2.8	BDE-209	
中国青藏高原南部	背景	2005	BDE-17,28,47,66,71,85,99,100,138,153,154,183,190,209	0.004—0.03(0.01)	nd	BDE-28,47,99	[80]
中国东部	电子垃圾拆解地	2007	BDE-28,47,66,85,99,100,138,153,154,209	71.6—5710(1910)	69.9—5530(1800)	BDE-47,99,209	[81]
	对照城市土壤			0.2—0.4(0.3)	nd—0.3(0.2)	BDE-47,99,209	
	农田			0.08—0.3(0.2)	0.09	BDE-47,99,209	
	化工园区			2.0—269(40.6)	1.9—268(40.3)	BDE-47,99,209	
	对照乡村土壤			0.3—0.8(0.6)	nd—0.8(0.5)	BDE-47,99,209	

注:a.∑PBDEs(含 BDE-209);b.∑PBDEs(不含 BDE-209).

Note:a. ∑PBDEs(including BDE-209);b. ∑PBDEs(not including BDE-209).

2　生物体内 PBDEs 的分布特征(Distribution characteristics of PBDEs in living organism)

环境介质中的 PBDEs 可通过扩散、呼吸、主动和被动吸收等作用进入生物体,并沿着食物链在生物体间迁移富集,最终进入人体,给生态环境和人体健康带来潜在的暴露风险.生物体内 PBDEs 的分布特征与生物种类、所处环境的污染状况以及个体组织的特异性具有一定关系.

2.1　植物中 PBDEs 的分布

目前 PBDEs 已在世界各地的植物体内检出(表 3),包括喜马拉雅山和南极等背景点和偏远地区[6,89],植物体内的 PBDEs 含量一般在 $pg\cdot g^{-1}$—$ng\cdot g^{-1}$级[82,87,89-102].偏远地区的植物体以低溴代 PBDEs 为主,高永飞等[89]研究发现喜马拉雅山地区沿珠穆朗玛峰北坡海拔 5000 m 以上的苿籽草和棘豆样品中∑PBDEs在 2.1—3.6 $ng\cdot g^{-1}$ lw 之间,属全球背景水平,其主要来源于大气的长距离传输.而在电子垃圾拆解地、PBDEs 生产厂、人口稠密混杂工业区的大都市 PBDEs 含量较高,且大多以高溴代 PBDEs 尤其是 BDE-209 为主[82,87,91-99].Hu 等[99]研究发现北京 6 种树树皮中 PBDEs 的均值(包含 BDE-209)(780 $ng\cdot g^{-1}$ lw)低于美国阿肯色州(5700 $ng\cdot g^{-1}$lw)和中国东部电子垃圾拆解地(1400 $ng\cdot g^{-1}$ lw),高于欧洲(德国比利时,72 $ng\cdot g^{-1}$ lw;意大利里雅斯特,13 $ng\cdot g^{-1}$ lw)和韩国(140 $ng\cdot g^{-1}$lw).除乔木外,草本植物的叶片及根系也检出多种 PBDEs,以低溴代 PBDEs 尤其是 BDE-47、99 为主[100,102].Wang 等[100]通过分析多种植物根系及叶片中的∑PBDEs(3.3—94.3 $ng\cdot g^{-1}$dw),发现植物体低溴代 PBDEs 比例高于土壤.此外,谷物、蔬菜、水果等食物中也检测出了 PBDEs[90,101].

PBDEs 在不同植物及组织中的分布存在一定差异,同时其含量也随季节变化发生改变.Yogui 等[6]研究发现 PBDEs 在苔藓体内的含量高于地衣,其差异可能由于植物从大气中吸收 PBDEs 的机制.Hu 等[99]发现 PBDEs 在不同树种间有着不同的浓度分布特征,以柳树为最高,大的叶表面积使其能更好地与空气中 PBDEs 作用,又因其低脂肪含量,是理想的大气中 PBDEs 被动采样器.St-Amand[103]发现加拿大渥太华某垃圾填埋场附近云杉针叶中 PBDEs 含量在夏季保持持续上升,而之后到春天呈现下降趋势,春季萌芽期最低,并认为下降是因为冬春交替之际针叶蜡质层的退化和磨损所致.

表 3　PBDEs 在植物体内的分布($ng\cdot g^{-1}$lw)

Table 3　Distribution of PBDEs in plants($ng\cdot g^{-1}$lw)

国家/地区 Country/Area	种类 Species	年份 Year	PBDEs 单体 PBDEs congeners	∑PBDEs(均值)[b] ∑PBDEs(mean)[b]	BDE-209(均值) BDE-209(mean)	主要单体 Main congeners	文献 Reference
南极乔治王岛	地衣、苔藓	2005—2006	BDE-15,28,33,47,49,66,85,99,100,153,154,183	0.1—1.2[c]		BDE-47,99,100	[6]
中国喜马拉雅山	苿籽草、镰型棘豆	2005	BDE-17,28,47,66,71,99,100,153,183	2.1—3.6(2.9)		BDE-28	[89]
巴基斯坦	小麦、水稻	2013	BDE-28, 35, 47, 100, 99, 154, 153,183	0.07—46[c]		BDE-99	[90]
中国莱州湾 BFRs 生产厂	草本植物、超市蔬菜	2006	BDE-28, 47, 99, 100, 153, 154, 183,206,207,208,209	70—5900	52—3100	BDE-209	[91]
中国 68 个城市	马尾松、杨树等树皮	2004—2007	BDE-17,28,47,49,66,71,85, 99, 100, 138, 153, 154, 183, 190,209	0.02—48.3(2.8)[ac]	37.7—88.4[e]	BDE-209	[95]
北美	硬木及针叶树树皮	2000—2001	BDE-17,28,47,49,66,85,99, 100, 138, 153, 154, 166, 183, 196,203,206,207,208,209	2.3—5700[a]	nd—5570	BDE-47,99,209	[97]
中国北京	松、杨树等树皮	2009—2010	BDE-28, 47, 99, 100, 153, 154, 183,206,209	99—3700(780)[a]	95—3600(760)	BDE-209	[99]
西班牙	蔬菜、水果	2006	BDE-47,99,100,153,154,183	0.01—0.05[d]		BDE-47,99	[101]
纽约和伦敦岛,北极斯瓦尔巴特群岛	苔藓、丛生虎耳草、仙女木、莎草等	2011—2012	BDE-17,28,47,66,71,85,99,100,138,153,154,183,190	0.04—0.5[c]		BDE-47,99,183	[102]

注:a.∑PBDEs(含 BDE-209);b.∑PBDEs(不含 BDE-209);c.dw:干重;d.ww:湿重;e. 单位为百分比,表示占∑PBDEs比例.

Note:a. ∑PBDEs(including BDE-209);b. ∑PBDEs(not including BDE-209);c. dw:Dry weight;d. ww:Wet weight;e. The percentage of the PBDEs content against the ∑PBDEs.

2.2 动物中 PBDEs 的分布特征

对于陆生和水生动物体内 PBDEs 的分布均有大量研究(表 4).陆生动物体内 PBDEs 的分布与其种类、性别及器官的特异性有关,又因其可与污染源直接接触,很多陆生生物都有很高的 PBDEs 含量,且比水生生物更易积累高溴代 PBDEs,以 BDE-209 为最主要单体[88,104-112].刘娟等[104]研究发现,公鸡鸡肉和鸡肝组织∑PBDEs(42 ng·g^{-1}lw、39 ng·g^{-1}lw)高于母鸡(20 ng·g^{-1}lw、9.3 ng·g^{-1}lw),且鸡肉中 PBDEs 含量显著高于鸡肝.Qin 等[106]研究发现浙江台州电子垃圾拆解区附近的鸡脑中 PBDEs 相比其他组织器官低,血脑屏障保护脑免受 PBDEs 的生物积累,且高溴代 PBDEs 较低溴代更有效.Daso 等[109]发现南非的地犀鸟和肉垂鹤的蛋壳样品中 PBDEs 分布存在明显的差异,可能是由于这两种鸟类具有不同的食性、栖息地选择、迁移行为以及代谢能力等.Yu 等[112]研究一个陆生食物网发现 BDE-209 在高营养级生物体内存在脱溴代谢,红隼、麻雀和老鼠体内九溴代 PBDEs(nona-BDEs)/deca-BDEs 的值(1、1.4、1.2)明显大于土壤和草(0.4、0.5).

表 4 PBDEs 在陆生动物体内的分布 (ng·g^{-1}lw)

Table 4 Distribution of PBDEs in terrestrial animals (ng·g^{-1}lw)

国家/地区 Country/Sites	种类 Species	年份 Year	肌肉 Muscle	肝脏 Liver	脑 Brain	脂肪 Fat	∑PBDEs^{a}	主要单体 Main congengers	文献 Reference
中国广东清远电子垃圾回收场	鸡	2005	4.4—4382.9^{e} 3.2—178.6^{f}	3.6—7916.5^{e} 0.7—395.8^{f}			0.7—7916.5	BDE-209	[104]
中国河北白洋淀	鸭子	2005—2006	2.4 0.06^{d}	4.3 0.4^{d}	5.3 0.4^{d}		0.03—0.7^{bd}	BDE-47,99	[105]
中国浙江台州	三黄鸡	2000—2001	1092.4	1077.8	15.2	3138.1	15.2—3138.1	BDE-209	[106]
南非林波波省东部草原	地犀鸟、肉垂鹤蛋壳	2006					nd—264000^{i} 5980—7146000^{j}	BDE-47,100	[109]
中国喜马拉雅山横断山	大熊猫和红熊猫	2013	25.3—2045^{h}	38.4—742^{g} 52.6—581^{h}	83.0^{g}	16.4—279^{h}	16.4—2158	BDE-209	[110]
阿拉斯加北部和西部	北极熊	2004—2007		0.2—2.7			0.2—2.7	BDE-47	[111]

注:a.∑PBDEs(含 BDE-209);b.∑PBDEs(不含 BDE-209);c.dw:干重;d.ww:湿重;e.公鸡;f.母鸡;g.大熊猫;h.红熊猫;i.地犀鸟;j.肉垂鹤.

Note:a ∑PBDEs(including BDE-209);b. ∑PBDEs(not including BDE-209);c. dw:dry weight;d. ww:wet weight;e. Cock;f. Hen;g. Panda;h. Red panda;i. Ground hornbill;j. Wattled Crane.

虽然整体而言水生生物较陆生生物的 PBDEs 含量偏低,但 PBDEs 已广泛分布于包括偏远地区在内的水生生物体内,并在人类活动较为密集的地区达到较严重污染水平.Yang 等在中国青藏高原高山湖泊的鱼体内检出较低水平的∑PBDEs(0.09—4.3 ng·g^{-1} dw)[113],而法国罗纳河 4 种鱼体内∑PBDEs(4.5—182.5 ng·g^{-1} dw)高于欧盟国家的环境质量标准(42.5×10^{-3} ng·g^{-1} dw)4000 倍[114],泰晤士河等 4 条英国河流中 3 种鱼体内 PBDEs 含量(2—44 ng·g^{-1} lw)也远高于该标准[115].不同于陆生生物更容易积累高溴代 PBDEs,除了一些污染源(BDE-209)附近的地区外,大部分地区的水生生物体内以 BDE-47 为主[91,113-115].水生生物体内 PBDEs 单体的分布特征也受水生生物种类、组织器官、食物来源和代谢能力等影响[116-119].Sun 等[116]研究发现珠江三角洲地区的 3 种鱼体内 PBDEs 同系物分布差别很大,鲮鱼和罗非鱼以 BDE-47 为主,而琵琶鱼以 BDE-209 为主.田胜艳等[120]研究发现底层鱼比表层鱼更易富集 PBDEs,杂食性鱼类比肉食性鱼类富集的 PBDEs 更高;鱼体内 BDE-47/99 的值高于无脊椎动物,而 BDE-153/154 的值低于无脊椎动物,并认为可能是由于鱼类对 PBDEs 的代谢转化能力更强.

自 1960 年 PBDEs 产品生产以来,PBDEs 逐渐在世界各地的动物体内检出,并随时间基本呈现逐步增加的趋势[121-125].旧金山湾港海豹体内的 PBDEs 含量在 1989—1998 年间增大了两个数量级(从 88 ng·g^{-1} lw到 8325 ng·g^{-1} lw)[121],中国南海雄性江豚体内 PBDEs 含量从 1990 年的 84 ng·g^{-1} lw 升高到 2001 年的 980 ng·g^{-1}lw[122],北极及北大西洋地区鲸、海豹等海洋哺乳动物中 PBDEs 含量在 19 世纪末 20 世纪初达到最大值,反映出全球 PBDEs 工业产品产量和使用量的增加[123].随着各国相继对 PBDEs 商品的生产和使用采取了一系列限制措施,生物体 PBDEs 含量的增速变缓并开始呈现下降趋

势.北美萨利什海的海豹体内 PBDEs 在 1984—2003 年间呈指数增长，但在 2009 年出现下降，这是因为 2004 年开始禁用 penta-BDEs 和 octa-BDEs[124].Houde 等研究发现加拿大北极海豹体内 PBDEs 含量在 1998—2008 年间呈明显增长趋势，随后到 2011 年则持续下降，可能由于 2009 年 penta-BDEs 和 octa-BDEs 的全球监管[125].

2.3　人体中 PBDEs 的分布特征

关于人体内 PBDEs 分布，主要以母乳、血液和头发为研究对象，除此之外，也有研究在人体其他组织中检出 PBDEs.母乳中 PBDEs 的分布随地区的不同而变化，北美地区远高于其他地区；母乳中 PBDEs 单体以 BDE-47、99、153 为主，且日本、印度尼西亚、韩国、加纳、斯洛伐克、英国、瑞典、希腊、澳大利亚以及美国加州等地的研究都表明 BDE-47 含量最高，然而也有一些亚洲和欧洲国家由于 deca-BDEs 的大量使用，BDE-209 在母乳中有较高含量，甚至成为最主要单体[126-135].Park 等研究发现美国加州母乳中 PBDEs 含量（包含 BDE-209）（mean，134 ng·g^{-1} lw）超出越南、中国、韩国和日本（mean，0.4—3.7 ng·g^{-1} lw））以及欧洲（median，2—3 ng·g^{-1} lw）1 个数量级以上，也高于太平洋西北地区（mean、93.9、49.3 ng·g^{-1} lw）、德克萨斯州（6.2—580 ng·g^{-1} lw）等北美的其他地区[127].Meironyté 等发现 1972—1997 年间瑞典母乳中 PBDEs 含量从 0.07 ng·g^{-1}上升到 4.0 ng·g^{-1}，所检测到的单体种类也由最初的两种（BDE-47，153）变化为 1996/1997 年的 8 种[136].

血液中 PBDEs 的分布与人群的饮食习惯、生活习惯、所处区域环境状况及从事的职业有关，个体内也因组织的特异性而发生变化[137-143].Fromme 等研究表明 BDE-153 在吸烟者血液中多于不吸烟者[137].Uemura 等发现日本普通人群血液中的 PBDEs（2.7—4.8 ng·g^{-1} lw）以 BDE-209 为主，并认为鱼类的摄入是六溴代以下的 PBDEs 积累的主要因素[139].Shen 等研究发现浙江路桥、龙游、天台的儿童血液中 PBDEs 含量和单体组成差异显著，拥有电子垃圾拆解地的路桥污染最严重（32.1 ± 17.5 ng·g^{-1} lw），以 BDE-99、47、153 为主，龙游和天台污染较轻（12.1 ± 7.6、8.4 ± 4.0 ng·g^{-1} lw），分别以 BDE-153、156、147 和 BDE-47、116、15 为主[140].Zhu 等研究办公室清洁工、大学生、警察 3 类人群的血清 PBDEs 含量，发现办公室清洁工 ∑PBDEs（1.7—1980（140）ng·g^{-1} lw）远高于大学生（2.1—210（15）ng·g^{-1} lw）和警察（0.5—150（18）ng·g^{-1} lw），且以高溴代 PBDEs 为主，这表明清洁工可能通过摄入尘粒而富集更多高溴代 PBDEs[141].屈伟月等研究发现由于胎盘屏障作用婴儿脐血液 ∑PBDEs（1.5—12（3.9）ng·g^{-1} lw）低于母体血液（1.6—17（4.4）ng·g^{-1} lw），尤其是高溴代 PBDEs 更难透过胎盘到达胎儿[142].Ma 等研究表明美国纽约新生儿血液中 PBDEs 含量在 1997—2002 年间变化不大，随后尤其是 2004—2011 年间出现明显下降[144].

除母乳和血液外，头发也被用来指示人体内 PBDEs 的分布状况[96,145-146].Zheng 等研究表明，PBDEs 在垃圾拆解地周围居民头发内平均含量（43.2 ng·g^{-1} dw）为普通城市、乡村居民（16.6、10.0 ng·g^{-1} dw）的 3—4 倍[146].此外，在胎盘组织、精液中也检出 PBDEs[147-148]，中国浙江台州门诊男性患者血液中 ∑PBDEs约为精液中的两倍，其中血液中 BDE-153、209 含量远高于精液，而 BDE-28、47、99 差异不大，表明低溴代 PBDEs 较高溴代更易迁移到精液[147].

2.4　食物链生物富集放大作用

大量研究结果表明，陆生和水生食物网均存在 PBDEs 的生物放大，且放大程度依据不同单体种类而变化，BDE-153、154、155 是大多数陆生和水生食物网中生物放大因子（Trophic magnification factors，BMFs）较高的单体[62,87,149-155].Nie 等对中国电子垃圾拆解地陆生食物链的研究表明，总 PBDEs 的 BMFs 值为 1.6—3.6，即在所有食物链上存在生物放大作用；而各 PBDEs 单体的 BMFs 值为<1—11.6，这说明 PBDEs 单体具有不同的生物转化和累积能力[87].Shaw 等研究发现大西洋海豹体内的 PBDEs 的 BMFs 值为 17.1—76.5，以 BDE-153 和 BDE-155 最高（148—677，12—236）[153].生物放大作用还受食物链长度、类型以及生物组织器官的影响.Ma 等研究食物链浮游动物-无脊椎动物-鱼发现 PBDEs 单体的 BMFs 值为 0.61—3.21，而将海鸟纳入该食物链，则该值变为 0.8—4.4[152].Zheng 等发现，渤海湾包括浮游动物、无脊椎动物和鱼类在内的 11 种海洋生物体内 6 种 PBDEs 单体的 BMFs 变化范围是 0.2—78.6，最高 BMFs 值为食物链（矛尾复虾虎鱼到脉红螺）中 BDE-154（78.6），各 PBDEs 单体 BMFs 在肌肉和肝脏中也不同[156].在渤海湾食物网的研究发现，随着 PBDEs 单体 lg $K_{\rm OW}$的增加，其 TMFs 先增大后减少，当 lg $K_{\rm OW}$较

低时 PBDEs 同系物可迅速排出体外，而当 lg $K_{OW} > 6.87$ 时，生物可利用性降低[157]. 吴江平等[158]等研究发现，大部分生物的 BMFs 值都在 1—30 之间，BDE-154、183、209 的 BMFs 值较其他单体偏低，鸟类的 BMFs 值高于鱼类，哺乳动物 BMFs 值变化较大.

然而也有一些研究发现，随营养级增加生物体内的 PBDEs 含量发生了减少. 法国卢瓦尔河和塞纳河食物网的研究表明，营养级越高，PBDEs 含量在一些脂肪含量较高的动物体内（如鳗鱼、尸食性螃蟹）降低，发生"生物稀释"[159]. Kelly 等在研究加拿大北极海洋食物网时发现，PBDEs 的 TMFs 变化范围为 0.7—1.6，只有 BDE-47 超过了 1，BDE-28、66、99、100、118、153、154 均小于 1，可能是由于生物对 PBDEs 的代谢转化和快速排出所致[93].

3 结论与展望（Conclusion and prospects）

PBDEs 可以随大气、水体和生物长距离迁移，通过大气干湿沉降进入水体、土壤和生物体，并在不同环境介质间发生迁移，目前在世界各地多种环境介质中均有检出. 关于环境介质中 PBDEs 的时空分布特征的研究全面而广泛，不同环境介质的研究现状为：大气的相关研究多于水体和土壤，而水体又以沉积物居多；对于生物的研究，陆生植物多于水生植物，水生动物多于陆生动物；人体则以直接暴露者或是污染源附近的居民等特殊暴露人群作为主要研究对象，而普通人群的研究较少，其中血液、母乳及头发是研究最多的介质. 全球各区域间研究现状也存在较大差异：北美和欧洲的研究起步较早，而非洲、南美洲、亚洲的部分地区研究较少，不利于从全球尺度把握 PBDEs 的污染现状，我国虽然起步较晚，但近些年进展迅速. PBDEs 的分布特征表现为：偏远地区的大气中主要以低溴代 PBDEs 为主，同时低溴代 PBDEs 也是水体的主要单体，而高溴代 PBDEs 则在大气悬浮颗粒物、污染源附近的底泥、土壤以及污水处理厂的污泥中有较高的百分含量. 生物体中 PBDEs 的分布特征与生物种类、污染源、营养级以及个体组织的特异性有一定关系，如陆生生物含量普遍高于水生生物，高营养级生物大都高于低营养级生物；生物种属间和组织间的差异可能是由于其吸收、转化能力不同. 全球各区域对商品化 PBDEs 生产和使用的不同带来其全球时空分布的差异：北美以 penta-、octa-BDEs 产品为主，其环境中主要单体多为 penta-、octa-BDEs；而亚洲和欧洲更多使用 deca-BDEs，通常 deca-BDEs 是最主要单体. 总体来说，20 世纪之前环境中 PBDEs 含量呈上升趋势，2004 年开始各国陆续对 penta-BDEs、octa-BDEs 产品的生产和使用采取一些措施后，其增长率开始变小，部分区域的含量呈现下降趋势，尤其是在北美和欧洲. 然而在亚洲等很多地区 deca-BDEs 仍在使用，导致其含量仍持续增加. 通过对不同环境介质中 PBDEs 时空分布特征的分析，能够进一步了解全球各地各圈层的 PBDEs 污染源、分布特征，为对比分析国内外污染现状和水平提供数据基础，为 PBDEs 污染控制法律法规政策的制定提供参考.

由于各地 PBDEs 商品的流通以及 PBDEs 的持久性和长距离迁移性，实现环境中 PBDEs 的控制仍需很长时间，因此未来需重点关注仍在使用 deca-BDEs 产品的区域，以及 PBDEs 的生产加工地和电子垃圾拆解地等典型污染区域；并增加对于现有研究较少的地区及研究对象的研究；比如从特殊人群扩大到普通大众，农作物和加工食品中的分布也需更加详细地了解；PBDEs 在各环境介质间会发生一定的迁移和转化，未来需深入探讨其在大气、土壤、水体和生物体中的迁移转化机制，以进一步分析其归趋. 这些研究工作将有利于全球尺度把握 PBDEs 的污染现状和趋势，了解其在不同地区的时空分布特点，从而更加有针对性地对其污染进行控制.

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