

环境科学

(HUANJING KEXUE)

ENVIRONMENTAL SCIENCE

第34卷 第10期

Vol.34 No.10

2013

中国科学院生态环境研究中心 主办
科学出版社 出版



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中国东北主要河流沉积物中多溴二苯醚的含量状况及生态风险分析

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摘要: 为揭示多溴二苯醚(PBDEs)在东北主要河流流域内的污染现状,通过采集该地区流域内表层沉积物样品,采用GC-NCI-MS技术对沉积物中41种PBDEs同类物进行分析. 结果发现沉积物中BDE209含量低于检出限,其它40种PBDEs同类物总含量(不包含BDE209)范围(干重)为0.91~17.67 ng·g⁻¹. 其中第二松花江吉林市上游和下游沉积物样品中PBDEs的检出含量最高,分别为15.86 ng·g⁻¹、17.67 ng·g⁻¹,以BDE207和BDE47为主,分别占PBDEs总量的86.5%和76.6%;其它河流沉积物中各同族体含量差异并不明显. 实验结果与国内外最近的文献报道值相比较,再结合生态风险分析显示,东北主要河流沉积物中PBDEs的含量处于低污染水平,目前不存在生态风险.

关键词: 多溴二苯醚; 沉积物; 生态风险; 暴露水平; 污染; 东北主要河流

中图分类号: X522; X820.4 文献标识码: A 文章编号: 0250-3301(2013)10-3825-07

Analysis of Ecological Risk and the Content Situation of Polybrominated Diphenyl Ethers in Sediments from Northeast China River Basin

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Abstract: To investigate the polybrominated diphenyl ethers (PBDEs) pollution status, sediment samples were collected from major rivers in northeast China. Contents of 41 PBDEs congeners in sediments were measured using GC-NCI-MS. BDE209 was not detected, measured level of total PBDEs (excluding BDE209) ranged from 0.91 to 17.67 ng·g⁻¹ dry weight, the highest concentrations of PBDEs were detected in the sediment samples from upstream and downstream of Jilin City in the Second Songhua River Basin, with 15.86 and 17.67 ng·g⁻¹. BDE207 and BDE47 were the predominant PBDE congeners, with their concentrations accounting for 86.5% and 76.6% of the total PBDEs concentration measured in the samples. Each congener content difference was not obvious in other river sediments. PBDEs levels monitored in the present study were compared to those reported recently for districts located at home and abroad, and with ecological risk analysis. PBDEs content is at a low level in sediments of Northeast China River Basin and there is no ecological risk.

Key words: PBDEs; sediment; ecological risk; exposure level; pollution; Northeast China River Basin

东北地区是我国重工业、农业地区. 随着经济的迅速发展,城市人口密度加大,人类活动加剧,能源消耗量增大,环境污染问题也随之而来. 多溴二苯醚(PBDEs)属于溴代阻燃剂的一种,其热稳定性好,阻燃效率高,因此被当作添加型阻燃剂应用于包括纺织、家具、建材和电子等产品中,由于本身没有化学键的束缚,以致易从其应用产品(如电子产品)向环境中释放. 河流传输是环境污染物特别是POPs的主要迁移途径之一,PBDEs在河流沉积物中主要以颗粒相式存在且长期停留在水环境中,这使得PBDEs易通过食物链由水生物转移到人体当中. 又由于PBDEs与多氯联苯、二噁英具有相似的化学

性质和毒性、生物富集性,故过量的PBDEs对人体内分泌调节、神经和生殖等会产生严重的负面影响^[1~3].

就东北地区主要河流沉积物中PBDEs的研究,之前有林忠胜等^[4]、史鑫源^[5]学者做过相关研究,但未把松花江、嫩江、辽河、大凌河、辽河大辽河、浑河等重要支流纳入一个整体的研究范围,而且随着人们生产活动及城镇工业化的变化发展,环境生

收稿日期: 2012-08-11; 修订日期: 2013-04-18

基金项目: 全国土壤现状调查及污染防治专项项目(1210823804); 国土资源大调查项目(1212011220055)

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持 1 min, 然后以 $10^{\circ}\text{C}\cdot\text{min}^{-1}$ 的速率升至 220°C , 保持 3 min, 再以 $20^{\circ}\text{C}\cdot\text{min}^{-1}$ 的速率升至 300°C , 保持 10 min.

质谱条件: 负化学电离源 (NCI); 接口温度 280°C , 离子源温度 250°C , 四极杆温度 150°C , 反应气为甲烷; 数据采集采用选择离子模式 (SIM). 一~九溴 BDEs 特征离子 m/z 为 79 和 81; BDE209 特征离子 m/z 为 79、81、486.7 和 488.7; $^{13}\text{C}_{12}$ -BDE209 特征离子 m/z 为 415.6 和 494.6.

定性与定量分析: 定性分析方法采用与标样保留时间对照与 MS 确证的方法进行定性分析; 定量分析采用 5 点校准曲线法.

1.5 质量控制

QA/QC 控制方法包括采样现场空白、方法空白、加标空白、基质加标平行和样品平行样, 所有的空白和样品等都进行相同的处理过程, 包括萃取、净化、分析等. 以 BDE50 和 $^{13}\text{C}_{12}$ -BDE209 作为回收率内标, PCB209 作为进样内标. 目标化合物的基质加标回收率为 $72.5\% \sim 115.7\%$ ($n=5$). 每 12 个样品增加一个实验室空白和一个平行样品. 平行样品的相对标准偏差 $<15\%$. 以 3 倍信噪比作为方法的检测限 (LODs), 二~九溴二苯醚的方法检测限为 $0.001 \sim 0.05 \text{ ng}\cdot\text{g}^{-1}$, BDE209 为 $0.18 \text{ ng}\cdot\text{g}^{-1}$. 样品中回收率内标 BDE50 和 $^{13}\text{C}_{12}$ -BDE209 的回收率 $80\% \sim 115\%$, 所有分析结果均以干重表示.

2 结果与讨论

2.1 沉积物中 PBDEs 的含量

在东北地区主要河流沉积物中, 共检测出 40 种 PBDEs 同类物. 在 40 种同类物中, 辽河流域检测到 25 种, 松花江流域检测到 24 种, 其中第二松花江流域检测到 23 种; 嫩江检测到 8 种; 从嫩江和第二松花江交汇后到巴彦县以西的松花江干流及相应的支流 (拉林河、呼兰河、阿什河) 检测到 16 种同类物. 就单个采样点来看, 各采样点的检测率较低, 最低的是辽河铁岭上游 (S-02)、双台子河上游 (S-04)、嫩江齐齐哈尔下游 (S-16)、松花江哈尔滨上游 (S-22) 均为 10%, 最高的是浑河下游 (S-08) 仅为 40%, 检测率相对较高的是浑河下游 (S-08) 和阿什河出口处 (S-24), 而在 \sum PBDEs 的含量相对较高的第二松花江的吉林市上游 (S-11 含量为 $15.84 \text{ ng}\cdot\text{g}^{-1}$) 和下游 (S-12 含量为 $17.67 \text{ ng}\cdot\text{g}^{-1}$), 其检测率也仅为 20% 和 30% (图 2). BDE-209 在各研究点位均未

检出, 主要由于: ①研究区各采样点 \sum PBDEs 含量普遍较低, 而且 BDE-209 的检出限 ($0.18 \text{ ng}\cdot\text{g}^{-1}$) 远比其他化合物检出限 ($0.001 \sim 0.05 \text{ ng}\cdot\text{g}^{-1}$) 高, 所以 BDE-209 未被检出; ②BDE209 可通过化学降解、光降解、微生物降解、生物降解等途径脱溴为低溴 BDEs 同族体^[6].

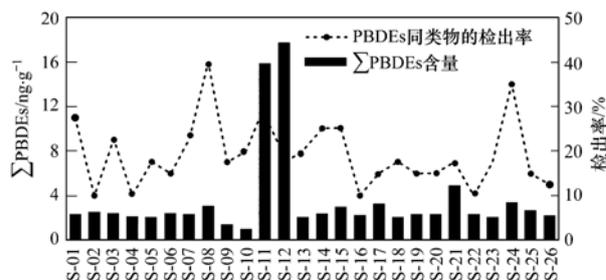


图 2 东北主要河流沉积物中 PBDEs 各同类物的检测率及总 PBDEs 的含量

Fig. 2 Detection rate and concentration of PBDEs congeners from sediments in Northeast China River Basin

东北地区主要河流沉积物中被检出的 40 种 PBDE 同类物的总含量 ($\sum_{40} \text{PBDEs}$) 为 $0.91 \sim 17.67 \text{ ng}\cdot\text{g}^{-1}$, 中位含量值为 $2.24 \text{ ng}\cdot\text{g}^{-1}$. 除第二松花江吉林市上游 (S-11)、第二松花江吉林市下游 (S-12)、松花江支流拉林河 (S-21) 之外, 其它河流沉积物中 \sum PBDEs 含量皆小于 $4.00 \text{ ng}\cdot\text{g}^{-1}$. 就局部流域而言, 辽河流域 (该流域范围点位编号是 S-01 ~ S-10) \sum PBDEs 含量为 $0.91 \sim 2.98 \text{ ng}\cdot\text{g}^{-1}$, 松花江流域 (该流域范围点位编号是 S-11 ~ S-26) \sum PBDEs 含量为 $1.94 \sim 17.67 \text{ ng}\cdot\text{g}^{-1}$. 在松花江流域中, 第二松花江流域 (S-11、S-12、S-13、S-14、S-15) \sum PBDEs 含量为 $1.94 \sim 17.67 \text{ ng}\cdot\text{g}^{-1}$, 嫩江流域 (S-16、S-17、S-18、S-19、S-20) \sum PBDEs 含量为 $1.99 \sim 3.15 \text{ ng}\cdot\text{g}^{-1}$, 巴彦县以西松花江干流及相应支流 (拉林河、呼兰河、阿什河) \sum PBDEs 含量为 $2.00 \sim 4.82 \text{ ng}\cdot\text{g}^{-1}$. 另外, \sum PBDEs 含量最高的河段是第二松花江吉林市上游及吉林市下游, 所检出的同类物主要以四溴 BDEs 中 BDE-47 化合物和九溴 BDEs 中 BDE-207 化合物分布为主, 其质量分数分别占 PBDEs 总量的 86.5% 和 76.6%. 在其它持久性有机污染物的研究中, 第二松花江吉林市上游、下游的多环芳烃相含量相对整个松花江流域也处于较高水平^[7]. 这表明, 第二

松花江 \sum PBDEs 含量与东北地区其它河流的含量存在明显的差异. 极高值与极低值的含量差近 20 倍. 除第二松花江之外, 其它河流流域沉积物中 \sum PBDEs 含量仅在 $0.91 \sim 4.82 \text{ ng} \cdot \text{g}^{-1}$ 之间. 纵观研究区 PBDEs 的分布特征, 除了第二松花江吉林市上游和下游外相对较高外, 其它地区含量差异并不明显.

与国内外其它地区报道值的比较表明, 虽然第二松花江吉林市上游和下游, 其沉积物中 PBDEs 含

量比研究区其它河流(段)处高出几倍甚至 20 倍, 但也远不及美国、英国、德国、西班牙等国家的部分河流的含量高. 如美国旧金山湾沉积物中 PBDEs 的含量是辽河流域的 71 倍、是嫩江流域 67 倍、是松江流域的 12 倍, 也远低于中国的珠江、莱州湾、香港地区的最高值, 如中国的莱州湾沉积物中 PBDEs 的含量是辽河流域的 604 倍、是嫩江流域 571 倍、是松江流域的 102 倍(表 1). 由此可见, 东北地区主要河流沉积物中 PBDEs 的含量和国内外相比处于较低水平.

表 1 不同地区沉积物中 PBDEs 含量比较(干重)

Table 1 Comparison of PBDEs concentrations in sediments from different areas (dry weight)

地点	$N^{1)}$	含量/ $\text{ng} \cdot \text{g}^{-1}$	文献	地点	N	含量/ $\text{ng} \cdot \text{g}^{-1}$	文献
Lake DV09, Canada	— ²⁾	$0.17^{4)}$	[8]	Pearl River Estuary, China	—	$0.11 \sim 13.03$	[25]
Taku sewage Estuary, China	13	$0.1 \sim 0.5$	[9]	Guiyu pollution River, China	14	$51.3 \sim 16.1$	[26]
Yangtze River Delta, China	40	ND ³⁾	[10]	Scheldt Estuary, Netherlands	—	$0.6 \sim 17.6$	[27]
Ny-Ålesund Lakes, Norway	14	$0.06 \sim 0.60$	[11]	Netherland	5	$0.6 \sim 17.6$	[28]
Lake Ellison, Norway	10	0.73	[12]	松花江流域, 中国	24	$1.94 \sim 17.67$	本研究
Beijing, China	10	$1.3 \sim 1.8$	[13]	Rive and Offshore, Portugal	—	$0.5 \sim 18.0$	[29]
Lake Huron, USA	17	$1.02 \sim 1.87$	[14]	River and coastal sediments, Portugal	40	$0.5 \sim 21.0$	[29]
Haihe River Basin, China	—	$0.06 \sim 2.10$	[15]	Pearl River Estuary, China	—	$0.33 \sim 21.83$	[30]
Industrial Port, Korea	20	$2.03 \sim 2.25$	[16]	Denmark	—	$0.06 \sim 25.20$	[30]
Bohai, China	10	2.45	[17]	Sundarban mangrove wetland, India	12	$0.08 \sim 29.03$	[31]
Lake Michigan, USA	7	2.60	[18]	Ocean, Korea	—	$1.1 \sim 33.8$	[32]
Lake Ontario, Canada	20	2.80	[19]	Cinca River, Spain	8	$0.3 \sim 34.1$	[33]
辽河流域, 中国	25	$0.91 \sim 2.98$	本研究	Hadley Lake, USA	—	$5.2 \sim 37.6$	[20]
Lake Superior, USA	9	$0.49 \sim 3.14$	[20]	Pearl River Estuary, China	10	$9.88 \sim 39.0$	[34]
嫩江流域, 中国	8	$1.99 \sim 3.15$	本研究	Niagara River, USA	9	$0.72 \sim 48$	[35]
South China Sea Northern part, China	—	$0.04 \sim 4.48$	[21]	Pearl, China	—	$0.78 \sim 49.28$	[21]
Qingdao inshore, China	21	$0.12 \sim 5.51$	[22]	Viskan Rive, Sweden	—	$8 \sim 50$	[36]
Busan Bay, Korea	20	$0.38 \sim 5.86$	[14]	Hong Kong Offshore, China	14	$1.7 \sim 52.1$	[37]
Jinhae Bay, Korea	20	$0.03 \sim 6.02$	[14]	East Pearl River Estuary, China	9	$0.04 \sim 94.7$	[38]
Great Lakes, USA	9	$0.5 \sim 6.33$	[23]	San Francisco Bay, USA	22	ND ~ 212	[39]
Ocean, Korea	—	$0.05 \sim 6.37$	[24]	Tees Estuary, Britain	—	$1.3 \sim 1271$	[40]
Lake DV09, Canada	—	$0.12 \sim 6.87$	[16]	Laizhou Bay, China	11	$1.3 \sim 1800$	[41]
Westem Scheldt, Netherlands	—	$0.30 \sim 12.58$	[25]	Coastal marine sediments, Kuwait	7	$80 \sim 3800$	[42]

1) “ N ”表示检测到的目标化合物数目; 2) “—”表示无相关数据; 3) “ND”表示未检出; 4) \sum PBDEs 含量按最大值或平均值升序排列

2.2 沉积物中 PBDEs 的生态风险分析

仅仅与国内外不同地区的含量对比, 不足以反映研究区多溴二苯醚污染的生态风险, 故有必要对多溴二苯醚的污染进行生态风险分析. 毒害污染物的暴露评价一般有 2 种途径: 一种是根据使用量推算环境浓度, 此浓度一般以预测环境浓度 PEC (predicted environmental concentration) 表示; 另一种是定点采样仪器分析测定浓度, 此浓度一般以测定环境浓度 MEC

(measured environmental concentration) 表示^[43]. 本研究以样品沉积物样品中 PBDEs 测定的环境浓度 MEC 作为暴露评价途径来分析 PBDEs 的生态风险.

2.2.1 暴露水平分析

常见的多溴二苯醚阻燃剂工业品^[44]是五溴二苯醚 (PeBDE)、八溴二苯醚 (OBDE) 和十溴二苯醚 (DBDE), 均为商业混合物. 据美国环境保护署关于 PBDEs 分类标准假设^[44]三溴、四溴 BDE 来源于于

PeBDE,五~九溴 OBDE, BDE209 来源 DBDE,则 PeBDE 暴露水平为三溴、四溴 BDE 的总含量, OBDE 的暴露水平为五~九溴 BDE 的总含量,

DBDE 的暴露水平为 BDE209 含量^[45,46],故可得出东北主要河流沉积物中 PeBDE、OBDE、DBDE 的暴露水平 MEC(表 2).

表 2 PBDEs 工业品在东北主要河流沉积物中暴露水平

Table 2 Exposure level industrial products of PBDEs form sediments in Northeast China River Basin

PBDEs 工业品	辽河流域的 MEC/ng·g ⁻¹									
	S-01	S-02	S-03	S-04	S-05	S-06	S-07	S-08	S-09	S-10
PeBDE	0.344	0.017	0.095	0.009	0.022	0.032	0.067	0.314	0.027	0.011
OBDE	0.248	0.004	0.093	0.004	0.296	0.031	0.090	0.863	0.023	0.034
PBDEs 工业品	第二松花江流域的 MEC/ng·g ⁻¹					嫩江流域的 MEC/ng·g ⁻¹				
	S-11	S-12	S-13	S-14	S-15	S-16	S-17	S-18	S-19	S-20
PeBDE	0.315	13.937	0.066	0.151	0.082	0.002	0.052	0.010	0.032	0.024
OBDE	13.953	ND ¹⁾	0.070	0.051	0.692	0.011	0.049	0.040	0.077	0.065
PBDEs 工业品	松花江干流及相应支流的 MEC/ng·g ⁻¹									
	S-21	S-22	S-23	S-24	S-25	S-26				
PeBDE	0.026	0.007	0.017	0.313	0.037	0.019				
OBDE	2.279	0.008	0.040	0.823	0.200	0.013				

1)“ND”表示未检出

2.2.2 生态风险表征

由上述图 2 研究区的含量分布情况与国内外其它地区比较得知,东北地区主要河流沉积物中 PBDEs 的含量处于较低水平. 而对 PBDEs 生态风险评价首先确定其有风险还是无风险,若含量较低,无生态风险,就此作罢,若有生态风险,则需进一步深入研究. 故暂先采用较为保守的^[47]、低水平^[48]的生态风险分析方法即商值法^[49]来表征研究区生态风险水平. 商值法原理是:当生态风险商值 >1 时,存在生态风险,且数值越大,风险水平越高;当生态风险商值 ≤1 时,无生态风险.

该方法是采用研究区的暴露水平与来自于

PeBDE、OBDE、DBDE 产品针对淡水或海水沉积相环境中潜居生物(成年红虫)的多物种无显著影响浓度(multi-species no observable effect concentration, MSNOEC)的比值求得生态风险商值^[50]. PeBDE、OBDE、DBDE 产品的 MSNOEC 分别为 310、1 340、4 536 ng·g⁻¹^[51]. 运用公式:

$$Q = \text{MEC}/\text{MSNOEC} \quad (1)$$

式中, Q 为生态风险商值, MEC 为暴露水平. 结合研究区暴露水平(表 2)换算可得出,在东北主要河流沉积物中,来自于工业品 PeBDE、OBDE 的 PBDEs, 其风险商值均在 0.05 之下,远低于 1(图 3),故不存在生态风险.

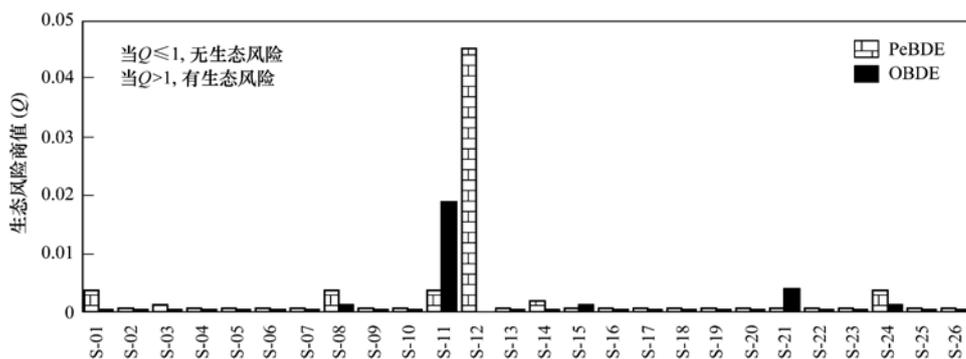


图 3 东北主要河流沉积物中 PBDEs 生态风险评价结果

Fig. 3 Evaluate results of ecological risk of PBDEs from sediments in Northeast China River Basin

3 结论

(1) 在东北主要河流 26 个监测点中,检测到 \sum_{40} PBDEs (不包括 BDE209) 的含量为 0.91 ~

17.67 ng·g⁻¹,第二松花江吉林市上游及下游中含量相对较高,分别为 15.84 ng·g⁻¹、17.67 ng·g⁻¹,其它流域中如辽河流域、嫩江流域、松花江干流及相应支流中含量差异较低,含量介于 0.91 ~ 4.82

ng·g⁻¹之间.除了第二松花江吉林市上游和下游外,研究区不同采样点含量差异并不明显,PBDEs分布较为均衡.

(2)与国内外其它研究值相比,研究区域内沉积物中PBDEs处于中低水平,但位于吉林市下游采样点受点源污染的影响较为明显,沉积物中PBDEs含量高出研究区中位值的7.8倍,值得关注.

(3)商值法进行的生态风险分析结果表明,研究区虽然存在PBDEs污染,但其生态风险商值极低,就目前而言,沉积物中PBDEs不会对该地区造成生态危害.

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环 境 科 学

(HUANJING KEXUE)

(月刊 1976年8月创刊)

2013年10月15日 34卷 第10期

ENVIRONMENTAL SCIENCE

(Monthly Started in 1976)

Vol. 34 No. 10 Oct. 15, 2013

主 管	中国科学院	Superintended	by	Chinese Academy of Sciences
主 办	中国科学院生态环境研究中心	Sponsored	by	Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences
协 办	(以参加先后为序) 北京市环境保护科学研究院 清华大学环境学院	Co-Sponsored	by	Beijing Municipal Research Institute of Environmental Protection School of Environment, Tsinghua University
主 编	欧阳自远	Editor-in -Chief		OUYANG Zi-yuan
编 辑	《环境科学》编辑委员会 北京市2871信箱(海淀区双清路 18号, 邮政编码:100085) 电话:010-62941102, 010-62849343 传真:010-62849343 E-mail: hjkx@rcees. ac. cn http://www. hjkx. ac. cn	Edited	by	The Editorial Board of Environmental Science (HUANJING KEXUE) P. O. Box 2871, Beijing 100085, China Tel:010-62941102, 010-62849343; Fax:010-62849343 E-mail: hjkx@rcees. ac. cn http://www. hjkx. ac. cn
出 版	科 学 出 版 社 北京东黄城根北街16号 邮政编码:100717	Published	by	Science Press 16 Donghuangchenggen North Street, Beijing 100717, China
印 刷 装 订	北京北林印刷厂	Printed	by	Beijing Bei Lin Printing House
发 行	科 学 出 版 社 电话:010-64017032 E-mail: journal@ mail. sciencep. com	Distributed	by	Science Press Tel:010-64017032 E-mail: journal@ mail. sciencep. com
订 购 处	全国各地邮电局	Domestic		All Local Post Offices in China
国外总发行	中国国际图书贸易总公司 (北京399信箱)	Foreign		China International Book Trading Corporation (Guoji Shudian), P. O. Box 399, Beijing 100044, China

中国标准刊号: ISSN 0250-3301
CN 11-1895/X

国内邮发代号: 2-821

国内定价: 90.00元

国外发行代号: M 205

国内外公开发行