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基于碳减排目标与排放标准约束情景的火电大气污染物减排潜力 李辉,孙雪丽,庞博,朱法华,王圣,晏培



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武汉典型饮用水水源中典型 POPs 污染特征与健康风 险评估

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摘要: 为了揭示武汉典型饮用水水源中典型持久性有机污染物(POPs)的污染特征与风险水平,采用固相萃取-气相色谱-质谱 定性定量分析法,对武汉长江及其支流上18个典型集中式饮用水水源地,共26个采样点水体中多环芳烃(PAHs)、有机氯农 药(OCPs)和多氯联苯(PCBs)浓度进行了检测,分析了 POPs 的污染水平,并开展健康风险评估. 结果表明, 26 个采样点均有 PAHs 检出,除苯并[k]荧蒽检出率为 88. 46% 外,其他 15 种单体检出率均为 100. 00%,多环芳烃累积 $hoig(\sum {\sf PAHs}ig)$ 检出范围 为 57. 04~475. 79 ng·L⁻¹, 平均值为 173. 86 ng·L⁻¹. PAHs 污染程度总体较低, PAHs 主要以中低环芳烃为主,来源于以石油 源为主的混合源. 共有 8 种 OCPs 被检出, (\(\sum \) OCPs) 范围为 ND ~ 4. 57 ng·L⁻¹, 平均值为 0. 78 ng·L⁻¹, OCPs 浓度水平相对 较低. 共有 24 种 PCBs 被检出, $\rho\left(\sum PCBs\right)$ 范围为 ND ~77. 49 $\operatorname{ng} \cdot L^{-1}$, 平均值为 9. 88 $\operatorname{ng} \cdot L^{-1}$, PCBs 主要以不易降解的高 氯联苯为主,部分点位 PCBs 浓度超过我国地表水环境质量标准限值, HeptaCBs-180 物质需要引起持续关注. 健康风险评估结 果显示,研究区域内 PAHs 和 PCBs 的致癌风险指数均处于 10 -6 ~ 10 -4 ,对人体可能产生潜在的致癌风险; OCPs 和 PCBs 的非 致癌风险指数均小于1,不会对人体产生非致癌风险.

关键词:武汉;长江;饮用水水源地;持久性有机污染物(POPs);健康风险

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Pollution Characteristics and Risk Assessment of Typical POPs in Typical Drinking Water Sources in Wuhan

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Abstract: To identify the pollution characteristics and risk level of common persistent organic pollutants (POPs) in drinking water sources in Wuhan, solid-phase extractiongas chromatography-mass spectrometry (SPE-GC-MS) was used to analyze 26 water samples from 18 drinking water sources located in the mainstream and tributaries of the Changjiang River, Wuhan. The concentrations of polycyclic aromatic hydrocarbons (PAHs), organochloride pesticides (OCPs), and polychlorinated biphenyls (PCBs) were measured, the pollution status of the common POPs was analyzed, and their health risks were assessed. The results showed the presence of PAHs in all the 26 samples. Except for benzo(k) fluoranthene, which had a detection rate of 88.46%, the detection rates of the other 15 PAHs were 100.00%. The detection range of ρ > PAHs was 57. 04-475. 79 ng·L⁻¹, with an average of 173. 86 ng·L⁻¹. The pollution levels of PAHs were overall low and the PAHs mainly comprised low and medium cyclic aromatic hydrocarbons derived from a mixed source, and dominated by petroleum derivatives. A total of eight OCPs were detected, and the range of (> OCPs) was ND-4.57 ng·L⁻¹ with a mean concentration of 0.78 ng·L⁻¹, which indicated that the concentration levels of OCPs were relatively low. A total of 24 types of PCBs were detected, and the range of ρ (\sum PCBs) was ND-77. 49 ng·L⁻¹ with a mean of 9. 88 ng·L⁻¹. The PCBs mainly comprised refractory high-chlorinated biphenyls, and the concentrations of PCBs in some samples exceeded the limit of the surface water standard in China. Our results suggest that the concentration of HeptaCBs-180 should be continuously monitored in the future. The results of the health risk assessment showed that the carcinogenic risk indexes of PAHs and PCBs in our study area were in the range of 10⁻⁶ to 10⁻⁴, which may pose a potential carcinogenic risk to humans. The non-carcinogenic risk indexes of OCPs and PCBs were less than 1, and did not pose a non-carcinogenic

Key words: Wuhan; Changjiang River; drinking water sources; persistent organic pollutants (POPs); health risk

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持久性有机污染物 (persistent organic pollutants, POPs)是一类由天然源或人为源产生的、 对人体和生态环境产生严重危害的污染物质. 具有 毒性大和难以生物降解的特点,能够在环境中长期 存在并通过食物链富集,进而通过各类环境介质大 范围传播[1]. 其中,多环芳烃(polycyclic aromatic hydrocarbons, PAHs)是一类具有较强溶解性、疏水 性和挥发性的碳氢化合物,主要来自于有机物的不 完全燃烧; 多氯联苯 (polychlorinated biphenyls, PCBs)^[2]和有机氯农药(organochloride pesticides, OCPs)[3]广泛应用于工业生产及动植物病虫害防 治.目前已有研究表明,在我国的长江[4]、太湖[5]、 巢湖[6]、白洋淀[7]和三峡水库[8]等水体中均检出了 POPs,这其中包括很多重要的饮用水水源地. POPs 具有很强的致癌、致畸、致突变性[9],可能对内分泌 产生干扰效应[10]. 联合国环境规划署早在 2001 年 就签署通过了《关于持久性有机污染物的斯德哥尔 摩公约》,该公约致力于对 POPs 在生产、流通、使用 和处理的全过程进行限制,截至目前已有30种物质 被纳入其中[11].

保障饮用水安全已经成为世界共识并受到公 众及相关部门的重视,确保饮用水安全的前提是 必须首先保障饮用水水源地水体的安全. 长江流 域作为我国最大的流域,分布着众多的饮用水水 源地,其中有53个水源地被列入国家重要饮用水 水源地,占国家重要饮用水源地的30%[12~14].作 为长江中游城市群的核心,武汉地区的饮用水水 源地主要分布在长江、汉江及其支流. 随着长江中

游流域经济带的快速发展,流域及周边农业、工业 和交通运输服务势必会对武汉饮用水水源地水体 造成污染[15]. 近年来,有关饮用水水源地 POPs 污 染的研究逐渐增多,而有关武汉典型饮用水水源 地 POPs 污染现状的调查还未开展. 本研究针对武 汉典型饮用水水源地水体中 POPs 的污染特征进 行分析,并对其健康风险进行评估和研究,以期为 保护长江流域生态环境和饮用水安全提供数据支 持和管理支撑.

1 材料与方法

1.1 试剂与仪器

对水样中16种优控PAHs、29种OCPs和28种 PCBs 进行检测(见表 1),各物质标准品购自美国 Wellington 公司. 采用固相萃取-气相色谱-质谱 (SPE-GC-MS)内标分析法进行定性定量测定, HLB (6 mL, 500 mg) SPE 小柱购自美国 Waters 公司; 二 氯甲烷(农残级)和甲醇(色谱纯)购自美国 Merck 二萘嵌苯-d12,测定 OCPs 的替代物为三苯基磷酸 酯,测定 PCBs 的替代物为 2,4,5,6-四氯间二甲苯; 测定 PAHs 和 PCBs 选用的内标物均为十氯联苯,测 定 OCPs 的内标物为五氯硝基苯; 所有替代物和内 标物均购自美国 Supelco 公司. 主要仪器如下: 24 孔固相萃取装置(美国 Mediwax 公司)、RE-52B 型 旋转蒸发仪(上海亚荣生化仪器厂)、KL512 型氮吹 仪(北京康林科技有限公司)和 7890A/5975C 型 GC-MS 联用仪(美国 Agilent 公司).

表 1 武汉典型饮用水水源地水体中检测的 POPs 物质

Table 1 POPs detected in typical drinking water sources in Wuhan

多环芳烃(PAHs)

萘(Nap)、苊烯(Acy)、二氢苊(Ace)、芴(Flu)、菲(Phe)、蒽(Ant)、荧蒽(Fla)、芘(Pyr)、苯并「a]蒽(BaA)、**肅**(Chry)、 苯并[b]荧蒽(BbF)、苯并[k]荧蒽(BkF)、苯并[a]芘(BaP)、茚并[1,2,3-cd]芘(InP)、二苯并[a,h]蒽(DahA)和苯并 「ghi] 花(二萘嵌苯)(BghiP)

五氯苯、α-HCH、六氯苯、β-HCH、γ-HCH、HCH、七氯、艾氏剂、氧化氯丹、环氧七氯 B、环氧七氯 A、γ-氯丹、ο,p'-DDE、 有机氯农药(OCPs) α-氯丹、α-硫丹、反式九氯、p,p'-DDE、狄氏剂、o,p-DDD、异狄氏剂、β-硫丹、顺式九氯、o,p'-DDT、p,p'-DDD、异狄氏 醛、硫酸盐硫丹、p,p'-DDT、异狄氏酮和甲氧滴滴涕

2,4'-二氯联苯(DiCBs-8)、2,2',5-三氯联苯(TriCBs-18)、2,4,4'-三氯联苯(TriCBs-28)、2,2',3,5'-四氯联苯

多氯联苯(PCBs)

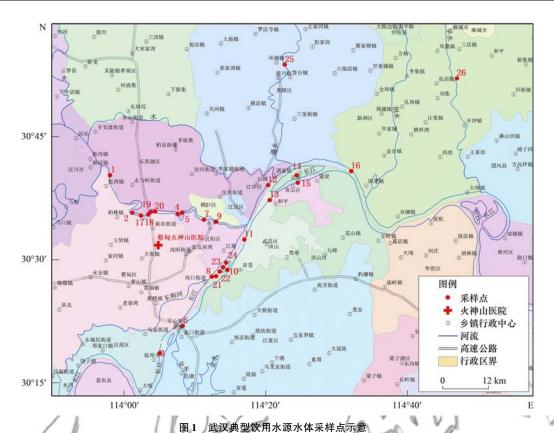
(TetraCBs-44)、2,2',5,5'-四氯联苯(TetraCBs-52)、2,3',4,4'-四氯联苯(TetraCBs-66)、3,3',4,4'-四氯联苯(TetraCBs-77)、3,4,4',5-四氯联苯(TetraCBs-81)、2,2',4,5,5'-五氯联苯(PentaCBs-101)、2,3,3',4,4'-五氯联苯(PentaCBs-105)、2,3,4,4',5-五氯联苯(PentaCBs-114)、2,3',4,4',5-五氯联苯(PentaCBs-118)、2',3,4,4',5-五氯联苯(PentaCBs-123)、3,3',4,4',5-五氯联苯(PentaCBs-126)、2,2',3,3',4,4'-六氯联苯(PentaCBs-128)、2,2',3,4,4',5'-六氯联苯 (HexaCBs-138)、2,2',4,4',5,5'-六氯联苯(HexaCBs-153)、2,3,3',4,4',5-六氯联苯(HexaCBs-156)、2,3,3',4,4',5'-六氯联苯(HexaCBs-157)、2,3′,4,4′,5,5′-六氯联苯(HexaCBs-167)、3,3′,4,4′,5,5′-六氯联苯(HexaCBs-169)、 2,2',3,3',4,4',5-七氯联苯(HeptaCBs-170)、2,2',3,4,4',5,5'-七氯联苯(HeptaCBs-180)、2,2',3,4',5,5',6-七氯联 苯(HeptaCBs-187)、2,3,3′,4,4′,5,5′-七氯联苯(HeptaCBs-189)、2,2′,3,3′,4,4′,5,6-八氯联苯(OctaCBs-195)、 2,2',3,3',4,4',5,5',6-九氯联苯(NonaCBs-206)和2,2',3,3',4,4',5,5',6,6'-十氯联苯(DecaCB-209)

1.2 样品采集

本研究于2020年9月在湖北省武汉市,选取了 18 个集中式饮用水水源地,共计26 个采样点位进 行了表层水样采集, 26 个采样点详细点位分布见

图 1.

其中1~24号点位位于长江干流及汉江干流, 25 号点位位于长江支流滠水河, 26 号点位位于长 江支流举水河. 所有水样经 0.45 μm 纤维滤膜过滤



1 Sampling sites of typical drinking water sources in Wuhan

后装入棕色玻璃瓶中,并在4℃下冷藏保存,24 h 内运送到实验室进行处理分析.

1.3 样品的预处理和分析

水样预处理: ①HLB 小柱与固相萃取装置连接后,用6 mL 二氯甲烷以及甲醇,依次对 HLB 小柱进行预先淋洗,期间 HLB 小柱不要流干,用6 mL 超纯水对 HLB 小柱进行活化,活化后再加入6 mL 超纯水,准备上样. ②水样中加入10 ng 替代物,开始固相萃取,处理后的水样通过活化后的 HLB 小柱,使用真空泵抽滤,控制溶液流速为5 mL·min⁻¹,水样滴完后,继续抽滤0.5 h. ③用10 mL 二氯甲烷洗脱后,用旋转蒸发仪和氮吹仪浓缩至0.5 mL,加入内标物后,置于4℃冰箱冷藏至开始 GC-MS 分析.分析 POPs 时所用 GC-MS 仪器分析条件参考文献[16].

1.4 样品的质量控制

本研究采用了方法空白、回收率实验和精密度对 PAHs 和 OCPs 进行质量控制和保证. 在实验过程中,在每组 10 个样品中增加 1 个方法空白,相对标准偏差(RSD) < 20% (n=6). 水样中 PAHs 和 OCPs 的加标回收率分别为 77.00% ~ 101.00% 和 68.00% ~ 84.00%. PAHs 和 OCPs 的方法检测限范围分别为 0.09 ~ 1.08 $\operatorname{ng\cdot L^{-1}}$ 和 0.07 ~ 0.16 $\operatorname{ng\cdot L^{-1}}$.对 PAHs 样品分别添加萘-d8、**福**-d12 和二

萘嵌苯-d12 作为回收率指示物,结果显示 PAHs 的回收率范围分别为 78.30%~105.60%、74.80%~101.70%和 70.60%~109.75%,16 种 PAHs 的检出限为 0.12~0.96 ng·L⁻¹,方法空白中有少量苊烯(0.12 ng·L⁻¹)被检出.最后的检测结果经回收率校正的空白扣除.对 PCBs 进行质量控制和保证,在分析过程中采用样品平行样、内标法定量、空白加标和方法空白.每 10 个样品为 1 组,分别设置一个空白加标、基质加标、方法空白和溶剂空白.每个样品都有替代标准物,相对标准偏差(RSD)<20%(n=6),说明检测数据可靠.每个样品均进行 3 次平行测定,取平均值为最终结果,方法空白没有物质检出.空白加标和基质加标回收率分别为 67.30%~116.70%和 71.90%~118.20%,检出限为 0.10~4.30 ng·L⁻¹,定量限为 1.80~5.70 ng·L⁻¹.

1.5 健康风险评价

本研究采用美国环保署(EPA)推荐的健康风险评价模型^[17]对武汉典型饮用水水源中检出的 POPs 进行健康风险评估,包括致癌风险和非致癌风险,具体公式如下:

致癌风险指数(R):

$$R = CDI_{oral} \times SF_{oral}$$
 (1)

 $\mathrm{CDI}_{\mathrm{oral}} = (\mathrm{CW} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}) / (\mathrm{BW} \times \mathrm{AT})$

(2)

式中,R 为终生致癌风险指数;CDI_{oral}为人体每天饮水摄入量[mg·(kg·d)⁻¹];SF_{oral}为物质的致癌斜率参数(kg·d·mg⁻¹),具体数值由相关研究及 EPA 综合风险信息系统(IRIS)确定;CW 为物质在水中检出浓度(mg·L⁻¹);IR 为日饮水量(L·d⁻¹),查阅文献[18,19],成人为 1.85 L·d⁻¹,儿童为 1.00 L·d⁻¹;EF 为暴露频率 365 d·a⁻¹;ED 为暴露持续时间,参考中国统计数据,成年男性为 74 a,成年女性为 78 a,儿童推荐为 12 a;BW 为体重,成年男性为 67.7 kg,成年女性平均为 59.6 kg,儿童为 26.8 kg^[20];AT 代表暴露时间,计算可知,成年男性为 27 010 d、成年女性为28 470 d和儿童为4 380 d.

非致癌风险指数(HI):

$$HI = CDI_{oral}/RfD$$
 (3)

式中,RfD 为非致癌参考量(mg·kg⁻¹·d⁻¹),取值根据 EPA 综合风险信息系统确定.

当致癌风险指数小于 10⁻⁶时,不会对人体造成 致癌危害; 致癌风险指数在 10⁻⁶~10⁻⁴时,人体暴 露可能具有潜在致癌风险,应该重视其健康问题; 当致癌风险指数大于 10⁻⁴时,将对人体造成致癌危 害. 当非致癌风险指数小于 1 时,不会产生非致癌健 康风险; 当非致癌风险指数大于 1 时,将产生非致 癌健康风险.

对于PAHs,终生致癌风险模型已经广泛应用在PAHs 对各类人群的健康风险评估^[21],由于优控的系列物质并没有完整的毒性参数,但其作用机制相似,为了确定各单体PAHs的毒性大小,本研究引入了毒性当量因子TEF^[22],其标准设定BaP为1,其他单体PAHs的TEF值通过与等量BaP毒性比较大小

而得出,PAHs 致癌风险指数(R)公式如下:

$$R = \frac{\text{TEQ}_{\text{BaP}} \times \text{IR} \times \text{CSF} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}}$$
 (4)

$$TEQ_{BaP} = \sum_{i=1}^{n} c_i \times TEF_i$$
 (5)

式中, TEQ_{BaP} 为 PAHs 基于 BaP 的毒性当量 $(mg \cdot L^{-1})$; CSF 为致癌斜率因子,参考文献[23], 取值为7.3 kg·d·mg⁻¹; c_i 为单体 PAHs 在水中检出浓度 $(mg \cdot L^{-1})$,其余指标均与公式(2)中相关评价参数一致.

2 结果与讨论

2.1 PAHs 检出结果

PAHs 检测结果如表 2 所示. 从检出率方面来看,除苯并[k] 荧蒽检出率为 88. 46% (在 4、5 和 22 号点位没有检出)外,其他 15 种物质检出率均为 100%. 检 出 浓 度 方 面, ρ (萘) 最 大,达 37. 89 ng·L⁻¹,范围为 16. 58 ~ 66. 21 ng·L⁻¹;其次是菲,平均检出浓度达 27. 53 ng·L⁻¹;检出二氢苊最小,平均值为 1. 17 ng·L⁻¹,范围为 0. 47 ~ 2. 13 ng·L⁻¹、从空间分布方面来看,26 个水体采样点位中,22 号采 样 点 累 积 ρ (\sum PAHs) 最 大,为 475. 79 ng·L⁻¹,其次是 26 号点位的 401. 47 ng·L⁻¹,5 号点位 ρ (\sum PAHs) 最 大,为 475. 79 ng·L⁻¹,其次是 26 号点位的 401. 47 ng·L⁻¹,5 号点位 ρ (\sum PAHs) 为 173. 86 ng·L⁻¹,具体来看,其中 17 ~ 24 号点位于污水处理厂排口下游 500 ~ 4 000 m 范围 内,平均 ρ (\sum PAHs) 为 232. 93 表2 武汉典型饮用水水源中 PAHs 检出情况¹⁾

Table 2 Detection of PAHs in typical drinking water sources in Wuhan

PAHs 组分	最大值/ng·L-1	最小值/ng·L-1	平均值/ng·L-1	检出率/%
萘(Nap)	66. 21	16.58	37. 89	100. 00
苊稀(Acy)	2. 73	0. 54	1. 29	100.00
二氢苊(Ace)	2. 13	0.47	1. 17	100.00
芴(Flu)	9. 81	1.37	4. 49	100.00
菲(Phe)	62. 77	9.72	27. 53	100.00
蔥(Ant)	3. 31	0.67	1.61	100.00
荧蒽(Fla)	17. 51	2.68	8. 20	100.00
芘(Pyr)	9. 75	1.48	4. 55	100.00
苯并[a]蒽(BaA)	15. 90	0.79	3. 26	100.00
苗 (Chry)	25. 33	1.09	5. 45	100.00
苯并[b]荧蒽(BbF)	270. 24	2.52	25. 20	100.00
苯并[k]荧蒽(BkF)	23. 22	ND	5. 53	88. 46
苯并[a]芘(BaP)	15. 28	1.49	5. 82	100.00
茚并[1,2,3-cd]芘(InP)	67. 54	1.46	16. 94	100.00
二苯并[a,h]蒽(DahA)	16. 01	0.49	4. 68	100.00
苯并[ghi]菲(二萘嵌苯)(BghiP)	91. 95	1.88	20. 24	100.00
∑ PAHs	475. 79	57. 04	173. 86	100. 00

表 3 国内外各地区湖泊和河流水体中 PAHs 对比 $^{1)}/ng \cdot L^{-1}$

Table 3	Comparison	of PAHs in	lake and	river waters	in different	regions of	China and	abroad/ng·L ⁻¹
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湖泊河流	采样年份	PAHs 种类	范围	平均值	文献
武汉长江饮用水源地	2020	16	57.04 ~ 475.79	173.86	本研究
北江	2020	16	41.20 ~413.80	90.80	[23]
长江下游支流水体	2019	19	37.30 ~ 285.90	78.31	[24]
莫斯科河(俄罗斯)	2013	15	50.60 ~ 120.10	_	[25]
三峡水库	2008	16	13.80 ~ 97.20	30.11	[26]
晋江	2011	16	42.00 ~63.00	53.23	[27]
钱塘江	2005	15	70.30 ~ 1 844.40	283.30	[28]
考卡河(哥伦比亚)	2010 ~ 2011	12	52.10 ~12 888.20	2 344. 50	[29]

^{1)&}quot;一"表示文献中无此数据

ng·L⁻¹; 剩余 18 个点位平均 ρ (\sum PAHs)为 147.61 ng·L⁻¹,空间上表现出污水处理厂尾水排口处的 PAHs 污染水平要高于其他饮用水水源地点位. 由表 3 可以看出,武汉典型饮用水源水体中检出 ρ (PAHs)与北江枯水期(41.20~413.80 ng·L⁻¹)和长江下游支流水体(37.30~285.90 ng·L⁻¹)持平、略高于俄罗斯的莫斯科河(50.60~120.10 ng·L⁻¹)、三峡水库(13.80~97.20 ng·L⁻¹)和晋江(42.00~63.00 ng·L⁻¹),明显低于钱塘江(70.30~1844.40 ng·L⁻¹)和考卡河(52.10~12888.20 ng·L⁻¹),总体上武汉典型饮用水水源 PAHs 污染程度较低,但随着长江航运的快速发展,大量船只的尾气排放及燃油泄漏均会加重长江与汉江流域的

PAHs 污染状况,为保障饮用水安全,需要持续关注水源地水质状况.

将武汉典型饮用水水源中检出的 16 种 PAHs 按其含有的苯环数量分为低分子 PAHs(2~3环)、中分子 PAHs(4环)和高分子 PAHs(5~6环)^[30],具体各环组成比例见图 2. 武汉典型饮用水水源中 PAHs 主要以 3、4 和 5 环为主,其中 3 环占 PAHs 比例为 31%~33%, 4 环占 PAHs 比例为 25%~27%、5 环占 PAHs 比例为 20%~25%,各采样点在环数组成上基本一致. 总体来看,武汉典型饮用水源水体中 PAHs 主要以中低环为主,这是因为低环数 PAHs 亲水性更强,环数越低越容易向水体中聚集^[31].

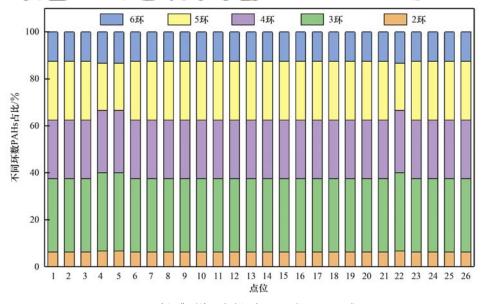


图 2 武汉典型饮用水水源中不同环数 PAHs 组成

Fig. 2 Composition of PAHs with different ring numbers in typical drinking water sources in Wuhan

不同来源的 PAHs 在单体组成上具有一定的差异,为了更好地研究武汉典型饮用水水源中 PAHs 的来源,本研究采用分子比值特征法^[32]对几种同分异构单体的浓度比值进行了分析,包括 Ant/(Ant + Phe)、BaA/(BaA + Chry)、Fla/(Fla + Pyr)和 InP/

(InP + BghiP). 不同来源 PAHs 的源解析比值组成见表 4.

由图 3 可知,武汉典型饮用水水源中 PAHs 的来源主要是石油源和石油燃烧源,部分点位存在煤及生物质燃烧源.通过分子比值特征法计算可得,

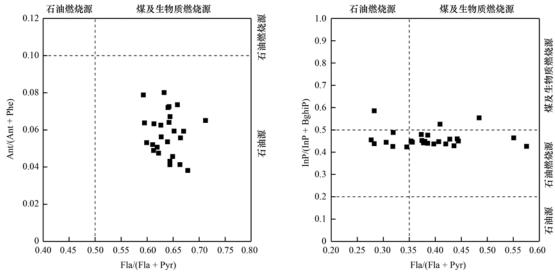


图 3 武汉典型饮用水水源中 PAHs 来源分析

Fig. 3 Source analysis of PAHs in typical drinking water sources in Wuhan

表 4 PAHs 的源解析比值

Table 4 Source resolution discrimination of PAHs

			1/0/
项目	石油源	石油燃烧源	煤及生物质燃烧源
Ant/(Ant + Phe)	< 0.10	>0.10	18 (5 -
BaA/(BaA + Chry)	< 0.20	0.20 ~ 0.35	>0.35
Fla/(Fla + Pyr)	< 0.40	0.40 ~ 0.50	>0.50
InP/(InP + BghiP)	< 0.20	0.20 ~ 0.50	>0.50

26 个点位均有石油源,部分点位还有石油燃烧源或 煤及生物质燃烧源.由此可知,武汉典型饮用水水源中 PAHs 的来源受到了石油源、石油燃烧源和煤及 生物质燃烧源共同的影响,作为长江航道的重点城市,船舶航运石油燃烧及泄漏和机动车尾气排放都 会对水源地水质产生影响.

2.2 OCPs 检出结果

武汉典型饮用水水源中 OCPs 检出情况如表 5 所示. 从检出物质种类来看,在 29 种 OCPs 中,共有五氯苯、六氯苯、 γ -氯丹、 α -氯丹、2,4'-DDE、4,4'-DDE、顺式九氯和反式九氯这 8 种 OCPs 被检出. 从检出率方面来看,六氯苯检出率最高,达到了88.46%;其次是五氯苯,检出率为 50%;2,4'-DDE和4,4'-DDE这 2 种物质检出率最低,仅为 3.85%,均只在 9 号采样点检出. 从检出物质浓度来看, ρ (六氯苯)最高,检出范围为 ND ~ 0.69 ng·L⁻¹,平均值为 0.24 ng·L⁻¹, ρ (4,4'-DDE)最低,平均值仅为 0.02 ng·L⁻¹.

各采样点位 8 种 OCPs 的累积浓度如图 4 所示. 9 号点位的 $\left(\sum OCPs\right)$ 最大,为 3. 32 $ng \cdot L^{-1}$;其次是 21 号和 1 号点位, $\left(\sum OCPs\right)$ 分别为 2. 61 $ng \cdot L^{-1}$ 和 2. 53 $ng \cdot L^{-1}$; 17 和 20 号点位的 OCPs 均

表 5 武汉典型饮用水水源中 OCPs 检出情况

Table 5 Detection of OCPs in typical drinking water sources in Wuhan

Table 3	Detection of OCI	s in typicar din	iking water sot	nces in wunan
OCPs	最大值 /ng·L ⁻¹	最小值 /ng·L ⁻¹	平均值 /ng·L ⁻¹	检出率/%
五氯苯	0.49	ND	0.14	50.00
六氯苯	0.69	ND	0. 24	88. 46
γ -氯丹	0. 48	ND	0.08	30. 77
α -氯丹	0.58	ND	0.09	34. 62
2,4'-DDH	E 0. 67	ND	0.03	3. 85
4,4'-DDI	E 0. 60	ND	0.02	3. 85
顺式九象	0.58	ND	0.10	26. 92
反式九象	0.48	ND	0.07	15. 38

1) ND 表示未检出

未检出. 26 个点位检出的 (∑OCPs) 范围为 ND~ 3.32 ng·L⁻¹,平均值为 0.75 ng·L⁻¹. 由表 6 可以看 出,武汉典型饮用水水源中 OCPs 的检出浓度与白 洋淀表层水体中(0.69~4.50 ng·L-1)、钱塘江杭州 段干流(1.32~6.68 ng·L-1)和千岛湖库区(1.90~ 7.60 ng·L⁻¹)相当,略低于固城湖水体(26.74~ 46. 12 ng·L⁻¹). 由此可知, 武汉典型饮用水水源 OCPs 的污染程度相对较轻. 检出物质为六氯苯、五 氯苯、九氯、氯丹和 DDE 类,其中六氯苯在我国已有 50 多年的生产历史,是我国 POPs 污染场地的主要 污染源之一,六氯苯的主要来源可以分成历史残 留污染源和新引入的污染源两类. 虽然六氯苯在 我国已经被禁止作为农药直接使用,但由于六氯 苯的持久性,使其可以在水体、土壤和沉积物等环 境介质中存留数年甚至数十年或更长的时间,另 外杀虫剂、杀菌剂、合成橡胶助剂、木材防腐剂和 有机合成化工原料的生产等成为六氯苯新的污染 来源. 氯丹是一种广谱有机氯杀虫剂,广泛用于控

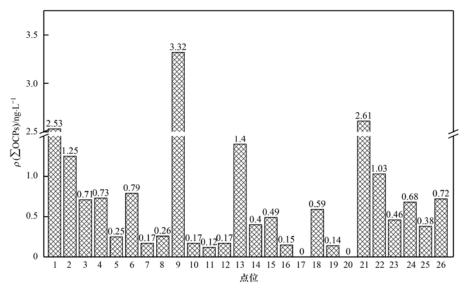


图 4 武汉典型饮用水水源中 $ho\left(\begin{array}{c} \sum \mathrm{OCPs} \end{array}\right)$

Fig. 4 Distribution of ρ (\sum OCPs) in typical drinking water sources in Wuhan

表 6 国内各地区湖泊和河流水体中 OCPs 对比 $^{1)}/ng \cdot L^{-1}$

Table 6 Comparison of OCPs in lake and river waters in different regions of China/ng·L-1

湖泊河流	采样年份	OCPs 种类 范围	平均值	主要检出单体	文献
武汉长江饮用水源地	2020	8 ND ~ 3. 32	0. 75	六氯苯	本研究
白洋淀	2015	6 0. 69 ~ 4. 50	1.77	HCHs	[33]
钱塘江杭州段干流	2013	5 1. 32 ~ 6. 68	1 1 100	α -HCH	[34]
千岛湖库区	2011	8 1.90 ~ 7.60	15+1	p , p' -DDE	[35]
固城湖	2015	19 26. 74 ~ 46. 12	33. 43	HCHs	[36]

1)"一"表示文献中无此数据; ND 表示未检出

制农作物和森林的病虫害,也用于家庭和工业等的白蚁防治.工业合成的 DDT 杀虫剂进入到自然环境中以后,在好氧条件下, DDT 代谢成为 DDE^[16],由此表明检出的 DDE 可能来源于 DDT 在长期环境中的降解.

2.3 PCBs 检出结果

武汉典型饮用水水源 PCBs 检出结果如表 7 所示. 除 TetraCBs- 44、TetraCBs- 52、NonaCBs- 206 和 DecaCB-209 未被检出外,共有 24 种物质被检出. 检出率方面, PentaCBs-105 检出率最高,达到65. 38%; 其次是 PentaCBs-114 和 PentaCBs-118,分别为61. 54%和50. 00%;检出率最低的物质为 DiCBs-8、TriCBs-18、TetraCBs-77、TetraCBs-81、PentaCBs- 101和 OctaCBs- 195,均只在 1 个点位被检出. 检出浓度方面,检出 HeptaCBs- 180浓度最大,范围为 ND~11. 79 ng·L $^{-1}$,平均浓度为 1. 24 ng·L $^{-1}$;其次是 HeptaCBs- 189,检出范围为 ND~8. 70 ng·L $^{-1}$,平均浓度为 0. 39 ng·L $^{-1}$; TriCBs- 18 浓度最小,平均浓度为 0. 01 ng·L $^{-1}$.

各点位 PCBs 组分的累积浓度如图 5 所示. 检

出最高的组分为 HeptaCBs-180, 达 11. 79 ng·L⁻¹, 在 4号点位出现,远高于我国的滴水湖(0.16~0.45 ng·L⁻¹)^[37],略高于南山老龙洞岩溶地下水(1.6~ 5.4)^[38]检出浓度. 其余 PCBs 组分的浓度较低,由表 8 所示,武汉典型饮用水源检出 PCBs 与上海市地表 水中(ND~34.84 ng·L-1)、北京市地表水中(2.99 ~ 32.7 $\text{ng} \cdot \text{L}^{-1}$) 和太湖上游(4.24 ~ 14.46 ng·L-1)污染程度相当,略高于意大利台伯河 (0.19~6.82 ng·L⁻¹),远低于埃及尼罗河三角洲 水体(14 400.00~20 200.00 ng·L⁻¹). 根据我国 《地表水环境质量标准》(GB 3838-2002),允许在 水中的 $\rho\left(\sum PCBs\right)$ 限值为 20 ng·L⁻¹, 而 EPA 规 定的水中允许 $\rho(\sum PCBs)$ 限值为 14 ng·L⁻¹, 4 号和9号点位 PCBs 显著超标,其余点位 PCBs 均 满足我国地表水环境质量标准的限值.4号和9号 点位位于汉江下游,这2个点位可能受到了城市 点源的污染. 但是本研究只进行了单次采样检测, 不能反映 PCBs 的全年丰度水平,后续还将开展跟 踪监测,充分了解 PCBs 污染状况. 水体中 PCBs 的

表 7 武汉典型饮用水水源 PCBs 检出情况1)

Table 7	Detection	of PCB	s in	typical	drinking	water	sources in	Wuhan
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PCBs 同系物	PCBs 物质	最大值/ng·L-1	最小值/ng·L	-1 平均值/ng·L ⁻¹	检出率/%
Di-CBs 系列	DiCBs-8	0. 57	ND	0. 02	3. 85
Ti CD。 玄利	TriCBs-18	0. 25	ND	0.01	3. 85
Tri-CBs 系列	TriCBs-28	1. 34	ND	0.07	7. 69
	TetraCBs-66	2. 04	ND	0. 22	23. 08
Tetra-CBs 系列	TetraCBs-77	4. 72	ND	0. 18	3. 85
	TetraCBs-81	3. 13	ND	0. 12	3. 85
	PentaCBs-101	2. 76	ND	0. 11	3. 85
	PentaCBs-105	3. 86	ND	0. 87	65. 38
	PentaCBs-114	4. 22	ND	0. 74	61. 54
Penta-CBs 系列	PentaCBs-118	3. 99	ND	0. 58	50.00
	PentaCBs-123	4. 43	ND	0. 43	26. 92
	PentaCBs-126	3. 85	ND	0.49	30. 77
	PentaCBs-128	4. 60	ND	0. 30	11. 54
	HexaCBs-138	4. 20	ND	0. 65	30. 77
	HexaCBs-153	4. 78	ND	0. 22	7. 69
Hexa-CBs 系列	HexaCBs-156	3. 83	ND	0.81	46. 15
Hexa-CDs 35.94	HexaCBs-157	2. 78	ND	0. 54	30. 77
	HexaCBs-167	4. 16	ND	0. 51	26. 92
	HexaCBs-169	4. 10	ND	0. 67	30.77
	HeptaCBs-170	3. 40	ND	0. 20	7. 69
Hepta-CBs 系列	HeptaCBs-180	11.79	ND	1. 24	30.77
Tiepta-CDs 2079	HeptaCBs-187	2. 73	ND	0. 21	11.54
/	HeptaCBs-189	8.70	ND	0.39	26. 92
Octa-CBs 系列	OctaCBs-195	7.98	ND	0.31	3. 85
) ND 表示未检出	8	11/10		/ 1 / 2	(. 6

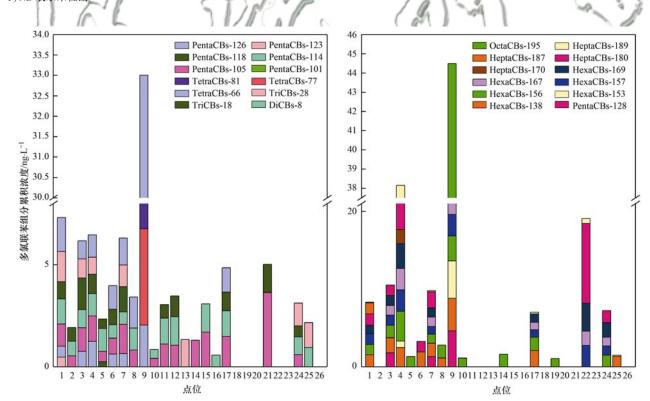


图 5 武汉典型饮用水水源中 PCBs 累积浓度

Fig. 5 Cumulative concentrations of PCBs in typical drinking water sources in Wuhan

赋存特征通常与它们的分子结构和迁移转化有关. 由表7可知,武汉典型饮用水水源中 PCBs 同系物的累积浓度排序为:七氯联苯>五氯联苯>六氯联 苯>四氯联苯.由此可知,难降解的高氯联苯同系物 在水体中浓度相对更高,这些物质更容易在水体中 残留,这与程加德等^[40]的研究结果相一致.

表 8	国内外各地区湖泊和河流水体中 PCBs	对比1)/ng·L-1
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m 11 o	Comparison of PCBs in	1.1 1		1 · cc .		c cl ·	1 1 1/ 1	- 1
Table 8	Comparison of PCBs in	Take and	river waters	ın differeni	regions	of China	and abroad/ng·L	•

湖泊河流	采样年份	PCBs 种类	范围	平均值	文献
武汉长江饮用水源地	2020	24	ND ~77.49	9. 88	本研究
上海地表水	2013	14	ND ~34.84	4. 42	[37]
北京地表水	2015 ~ 2016	8	2. 99 ~ 32. 70	(10.90 ± 10.40)	[39]
太湖上游	2019	18	4. 24 ~ 14. 46	7. 28	[40]
台伯河(意大利)	2014 ~ 2015	32	0. 19 ~ 6. 82	2. 03	[41]
尼罗河三角洲(埃及)	2013	10	14 400. 00 ~ 20 200. 00	_	[42]

1)"一"表示文献中无此数据,下同; ND 表示未检出

3 健康风险评估

为了评估人体暴露在低剂量 POPs 下的健康风险,本研究对所检出的 POPs 物质均纳人风险评价范围,在评价时只考虑了通过饮水途径所造成的健康风险. PAHs 各物质的毒性当量因子和致癌斜率因子参考文献[22,23], OCPs 和 PCBs 各物质的致癌斜率因子与非致癌参考剂量见表 9.

3.1 PAHs 健康风险评估

对武汉典型饮用水水源 26 个水样中 PAHs 进行健康风险评估,结果如图 6 所示. 采样点对不同人群的致癌风险指数范围分别为 $1.17 \times 10^{-6} \sim 2.22 \times 10^{-5}$ 、 $1.33 \times 10^{-6} \sim 2.52 \times 10^{-5}$ 和 $1.60 \times 10^{-6} \sim 3.03 \times 10^{-5}$. 武汉典型水源地 26 个水样的致癌健康风险指数均处于 $10^{-6} \sim 10^{-4}$ 范围,对人

表 9 OCPs 和 PCBs 物质致癌斜率因子与非致癌参考剂量参数

Table 9 Carcinogenic slope factor and non-carcinogenic reference dose parameters of OCPs and PCBs

dose parameters of OCI's and I CDs				
化合物	物质	RfD /mg•kg ⁻¹ •d ⁻¹	SF _{oral} /kg·d·mg ⁻¹	文献
OCPs	五氯苯	_	_	_
	六氯苯	8×10^{-4}	1.60	[43]
	γ -氯丹	5×10^{-4}	0. 35	[44]
	α-氯丹	5×10^{-4}	0. 35	[44]
	2,4'-DDE	10+	0. 34	[44]
	4,4'-DDE	1 1/4	0. 34	[44]
	顺式九氯	~ 14	+ %	///
	反式九氯	i d i	<u> </u>	05
PCBs	(D) di	2×10^{-5}	2. 0	[45,46]
	7/5	70. 6		

体可能产生潜在的致癌风险,特别是对儿童的健康风险相对更大,这与刘佳等^[47]和 Shi 等^[48]的研究结果相一致.

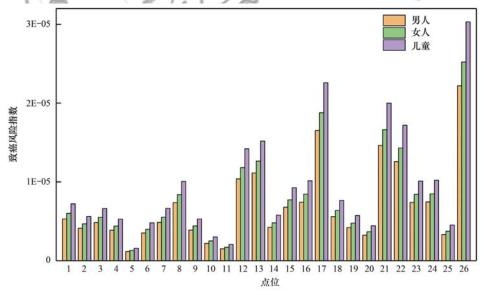


图 6 武汉典型饮用水水源中 PAHs 致癌风险指数

Fig. 6 Carcinogenic risk index of PAHs in typical drinking water sources in Wuhan

3.2 OCPs 健康风险评估

对武汉典型饮用水水源中 OCPs 进行健康风险评估,结果如图 7 所示. 研究区域不同人群 OCPs 暴露致癌风险指数范围分别为 $0 \sim 2.72 \times 10^{-7}$ 、 $0 \sim 3.09 \times 10^{-7}$ 和 $0 \sim 3.71 \times 10^{-7}$,非致癌风险指数范围

分别为 0~4.67×10⁻⁴、0~5.31×10⁻⁴和 0~6.38×10⁻⁴.由此可知,研究区域致癌风险指数均低于10⁻⁶,表明不会对人体产生致癌风险;儿童和成人的非致癌风险指数均远小于1,表明水体中的 OCPs不会对人体产生非致癌风险.

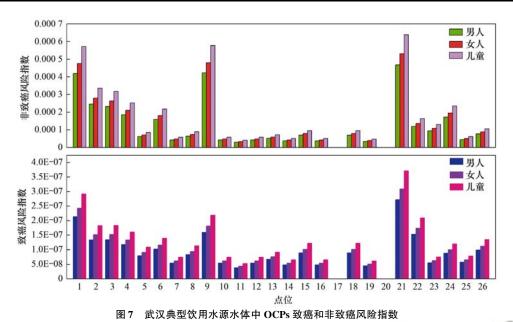


Fig. 7 Carcinogenic and non-carcinogenic risk indexes of OCPs in typical drinking water sources in Wuhan

3.3 PCBs 健康风险评估

对武汉典型饮用水水源中 PCBs 进行健康风险评估,结果如图 8 所示. 成年男性、成年女性和儿童这 3 种不同人群 PCBs 暴露致癌风险指数范围分别为 $0 \sim 4.83 \times 10^{-5}$ 、 $0 \sim 5.51 \times 10^{-5}$ 和 $0 \sim 6.12 \times 10^{-5}$,非致癌风险指数范围分别为 $0 \sim 0.11$ 、 $0 \sim 10.11$

0.12 和 0 ~ 0.14. 致癌风险指数在 10 ⁻⁶ ~ 10 ⁻⁴, 对人体可能产生潜在的健康危害; 非致癌风险指数均小于 1, 不会对人体产生非致癌风险. 因此, 建议重视武汉典型饮用水水源中 PCBs 的污染, 特别是 HeptaCBs-180 和 HeptaCBs-189 物质,以保障饮用水安全.

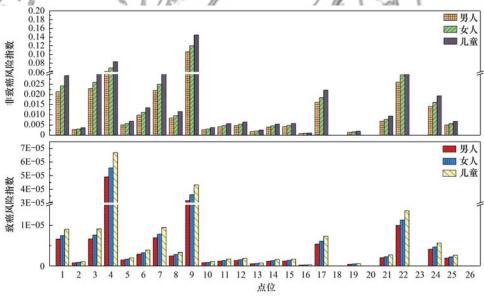


图 8 武汉典型饮用水水源中 PCBs 致癌和非致癌风险指数

Fig. 8 Carcinogenic and non-carcinogenic risk indexes of PCBs in typical drinking water sources in Wuhan

4 结论

(1)武汉典型饮用水水源地 26 个采样点均有 PAHs 检出, 萘检出浓度最大, 二氢苊检出浓度最小, PAHs 主要以中低环芳烃为主, 来源于以石油源 为主的混合源. 武汉典型饮用水水源 PAHs 污染程 度总体较低, 污水处理厂尾水排口处的 PAHs 污染 水平相对较高.

- (2) 武汉典型饮用水水源地共有 8 种 OCPs 被检出, OCPs 浓度水平相对较低. 共有 24 种 PCBs 被检出, PCBs 主要以不易降解的高氯联苯为主,部分点位 PCBs 浓度超过我国地表水环境质量标准限值, HeptaCBs-180 浓度相对较高,需持续关注和跟踪监测.
- (3)健康风险评估结果显示,武汉典型饮用水水源 PAHs 和 PCBs 的致癌风险指数均处于 10⁻⁶~

10⁻⁴范围,对人体可能产生潜在的致癌风险;OCPs 和 PCBs 的非致癌风险指数均小于1,不会对人体产生非致癌风险.

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