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我国邻苯二甲酸酯暴露水平与健康效应研究进展^{*}

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摘要 随着工业的迅速发展, 邻苯二甲酸酯(phthalate acid esters, PAEs)类增塑剂被广泛用于日常生产和生活。作为公认的内分泌干扰物质(endocrine disrupting chemicals, EDCs), PAEs对生态环境和人类健康构成的潜在威胁已经得到了多方面的研究证实。此类物质可以通过饮食摄入、呼吸吸入和皮肤接触等途径从环境介质中进入人体, 与体内的激素受体结合, 干预内源性激素与其受体之间的相互作用, 扰乱内分泌系统的各个环节, 从而影响人体健康。近年来, 我国科研人员开展了多项关于PAEs的人群暴露及健康效应研究, 研究对象涵盖儿童、青少年、成人和孕妇等多个不同群体。研究发现, 我国居民普遍暴露于PAEs, 并且这种暴露已经对人体健康造成了负面影响。本文系统综述了我国PAEs的环境污染现状、人群内暴露水平以及PAEs对居民健康的负面影响, 旨在为预测PAEs的暴露趋势和量化其健康风险提供数据支持。文章进一步对PAEs的复合暴露研究和环境流行病学调查进行展望, 以期为EDCs的规范管理和政策制定提供科学参考依据。

关键词 邻苯二甲酸酯, 内分泌干扰物, 暴露, 健康效应。

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Research progress on phthalate acid esters exposure levels and health effects in China

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Abstract With the rapid development of industry, phthalate acid esters (PAEs) have become increasingly prevalent in daily production and life. Recognized as endocrine-disrupting chemicals (EDCs), the potential threats posed by PAEs to the ecological environment and human health have been substantiated by numerous studies. Multiple exposure pathways, including dietary intake, respiratory inhalation, and dermal contact with environmental media, facilitate the entry of PAEs into the human body. These compounds bind to hormone receptors, disrupting the natural interplay

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between endogenous hormones and receptors, unsettling the functions of the endocrine system, and ultimately affecting human health. In recent years, Chinese researchers have incrementally embarked on studies examining the exposure levels and health effects of PAE among different populations—children, adolescents, adults, and pregnant women included. Studies have found that Chinese residents are commonly exposed to PAEs, leading to negative impacts on human health. This paper comprehensively reviews the current environmental exposure situation of PAEs in China, the internal exposure levels in different population groups, and the adverse health effects of PAEs on Chinese residents. The findings provide valuable data to support the prediction of PAEs exposure trends and the quantification of associated health risks. Furthermore, the review offers prospects for combined exposure assessment and environmental epidemiological investigations concerning PAEs, aiming to provide a scientific reference for the regulation and policy-making concerning endocrine-disrupting chemicals.

Keywords phthalates acid esters, endocrine disrupting chemicals, exposure, health effects.

邻苯二甲酸酯(phthalate acid esters, PAEs)是一类用于改善高分子材料性能的化学品,常作为增塑剂而被广泛添加到儿童玩具、食品包装、医疗耗材、室内装潢材料、农用薄膜及个人护理用品等消费品中^[1-3]。据统计,PAEs的全球年产量约为600—800万吨^[4]。中国作为全球最大的增塑剂生产国和消费国,增塑剂的产量从2014年的385.3万吨增长到2021年的426.2万吨^[5]。常见的PAEs如表1所示,包括邻苯二甲酸二甲酯、邻苯二甲酸二乙酯、邻苯二甲酸二丁酯、邻苯二甲酸苄基丁基酯、邻苯二甲酸二正丁酯、邻苯二甲酸二异丁酯和邻苯二甲酸二(2-乙基己基)酯。当前的主要发展方向是研发无毒环保型增塑剂,但是尚未发现经济实惠且具有优良增塑性能的替代品能够完全取代PAEs。因此,我国市场中PAEs的生产量和使用量仍呈上升趋势^[6]。

表1 常见的几种PAEs及其代谢物的基本信息

Table 1 Basic information of several common PAEs and their metabolites

PAEs(中文) Chinese name	PAEs(英文) English name	缩写 Abbr.	CAS号	mPAEs(中文) Chinese name	mPAEs(英文) English name	缩写 Abbr.	CAS号
邻苯二甲酸二甲酯	Dimethyl phthalate	DMP	131-11-3	邻苯二甲酸单甲酯	Monomethyl Phthalate	MMP	4376-18-5
邻苯二甲酸二乙酯	Diethyl phthalate	DEP	84-66-2	邻苯二甲酸单乙酯	Monoethyl phthalate	MEP	2306-33-4
邻苯二甲酸二丁酯	Dibutyl phthalate	DBP	84-74-2	邻苯二甲酸单丁酯	Monobutyl phthalate	MBP	131-70-4
邻苯二甲酸苄基丁基酯	Benzyl butyl phthalate	BBzP	85-68-7	邻苯二甲酸单苄酯	Mono-Benzyl phthalate	MBzP	2528-16-7
邻苯二甲酸二正丁酯	Di- <i>n</i> -butyl phthalate	DnBP	84-74-2	邻苯二甲酸单正丁酯	Mono- <i>n</i> -Butyl phthalate	MnBP	131-70-4
邻苯二甲酸二异丁酯	Di-isobutyl phthalate	DiBP	84-69-5	邻苯二甲酸单异丁酯	Mono-isobutyl phthalate	MiBP	30833-53-5
邻苯二甲酸二(2-乙基己基)酯	Di-(2-ethylhexyl) phthalate	DEHP	117-81-7	邻苯二甲酸单(2-乙基己基)酯	Mono-(2-ethylhexyl) phthalate	MEHP	4376-20-9

注:邻苯二甲酸酯代谢物mPAEs(phthalate metabolites, mPAEs)。

PAEs的理化性质相对稳定,但作为添加型增塑剂,其与塑料基质之间仅以氢键或范德华力相结合。这种结合力较为薄弱,因此在塑料制品的生产、使用和废弃过程中,PAEs容易释放进入环境^[7-8]。此外,PAEs可通过呼吸暴露、皮肤接触和消化道摄食等途径进入人体,对人类健康造成潜在不良影响^[9-10]。人体生物监测可以反映人体内PAEs的暴露水平,并可用于比较普通人群和特殊人群的暴露情况^[11]。据报道,美国、德国和韩国等发达国家已经开展了包括PAEs在内的多项人体生物监测项目^[12],但我国居民的PAEs暴露水平和总体暴露趋势仍需要进一步梳理。本研究对近10年来Web of Science、PubMed、CNKI等数据库中关于PAEs的文献进行系统回顾,探讨了我国PAEs内、外暴露水平的最新研究进展,并总结了PAEs暴露对我国居民健康产生的不良影响。本研究有助于全面了解我国PAEs的环境污染状况和人体负荷水平,同时也为国家和地方政府制定更有效的管理政策、防控措施以及健康指导提供了科学参考依据。

1 PAEs 的环境污染现状(Environmental pollution status of PAEs)

PAEs 的挥发和浸出特性使其成为环境中的主要污染物。大部分 PAEs 源于人工合成, 少部分由微生物或植物产生^[13]。在消费品中, PAEs 的用途多样且构成比例较高。大量研究表明, 我国的灰尘、水体、土壤和食品中普遍存在 PAEs 污染。城市化、工业化和农业生产等人类活动是导致环境中 PAEs 浓度升高的主要原因^[14]。欧盟的 REACH 法规、RoHS 指令, 美国的《消费品安全改进法案》、第 65 号提案, 加拿大的消费品安全法案等都对特定 PAEs 的使用提出了限制要求^[15]。这些法规直接导致欧洲和北美地区 PAEs 的生产受到限制, 但在中国、巴西和印度等发展中国家, PAEs 仍然被广泛生产和使用。

1.1 灰尘

PAEs 属于半挥发性有机污染物, 易于吸附在降尘或颗粒物上, 普遍存在于室内空气和环境空气中。据文献报道, 室内空气中 PAEs 的污染程度主要与 PAEs 单体的理化性质、家居建材类型和家庭用品相关^[16~18]。Wang 等^[19]检测了北京市 4 种室内灰尘样本中 15 种 PAEs 的浓度, 发现不同室内环境中 DEHP 的中值浓度最高。类似的结果也在上海市^[20](中值浓度: $428.00 \mu\text{g}\cdot\text{g}^{-1}$)、天津市^[21](中值浓度: $127.11 \mu\text{g}\cdot\text{g}^{-1}$)、成都市^[22](中值浓度: $151.00 \mu\text{g}\cdot\text{g}^{-1}$) 和广州市^[23](中值浓度: $355.00 \mu\text{g}\cdot\text{g}^{-1}$) 的居民住宅灰尘样本中被检测到, 可能是室内装潢材料和家居用品释放 PAEs 所致。杨其帆等^[24]调查了上海市不同场所室内灰尘中 PAEs 的分布特征, 同样发现室内环境中 DEHP 的含量最高, 这可能与我国较高的 DEHP 的使用率有关。

环境空气中的 PAEs 主要源于室内空气扩散、工业排放和废弃物释放, 且不同地区和不同季节的污染水平存在明显差异^[25]。在杭州市的大学城内, 室外空气中的 PAEs 主要来自室内空气扩散、建筑施工和工业排放, 14 种 PAEs 的总浓度在气相中为 $1573.00 \text{ ng}\cdot\text{m}^{-3}$, 在颗粒相中为 $126.00 \text{ ng}\cdot\text{m}^{-3}$ ^[26]。在北京, 室外空气细颗粒物($\text{PM}_{2.5}$)中的 PAEs 主要来自塑料工业和其他类型的燃料排放^[27], 冬季和春季的平均浓度分别为 $815.22 \text{ ng}\cdot\text{m}^{-3}$ 和 $455.82 \text{ ng}\cdot\text{m}^{-3}$ 。作为中国重要的工业中心, 天津市空气颗粒物中检测到的 6 种 PAEs 的平均浓度在工业区最高($135.90 \text{ ng}\cdot\text{m}^{-3}$), 并且在冬季超过了春季和夏季^[28]。西安市开展的研究中也观察到了类似的季节变化特征^[18], 可能是寒冷季节 PAEs 的挥发减少, 大部分附着在颗粒物上所致。在我国的大部分地区, 室内空气和环境空气中检测到的 PAEs 主要是 DEHP、DEP 和 DBP, 这与我国塑料制品中使用的主要 PAEs 类型相一致^[5,29~30]。

1.2 水体

PAEs 通过大气颗粒物沉降、地表径流、生活废水和工业污水排放等途径进入水环境, 其中 DEHP 和 DBP 的含量最高, 且在许多采样点中, 这两种化合物的浓度已经超过了《地表水环境质量标准》(GB 3838—2002) 中规定的浓度限值(DEHP 为 $8.00 \mu\text{g}\cdot\text{L}^{-1}$, DBP 为 $3.00 \mu\text{g}\cdot\text{L}^{-1}$)^[31]。我国是一个工业大国, 工业生产密集的地区面临着严峻的 PAEs 污染问题, 尤其是一些特定的 PAEs 单体。研究表明, DBP、DiBP、DMP 和 DEHP 是重庆市不同行业废水中主要的 PAEs 污染物, 它们的总量占比超过 80%。在这些样本中, PAEs 的平均浓度在进水中为 $10.49 \mu\text{g}\cdot\text{L}^{-1}$, 而在处理后的出水中降至 $3.34 \mu\text{g}\cdot\text{L}^{-1}$ ^[32]。污水处理厂是处理城市生活废水的关键设施。Sun 等^[33]研究发现, 哈尔滨市最大的污水处理厂中 19 种 PAEs 的平均浓度在进水和出水样本中分别为 $30.10 \mu\text{g}\cdot\text{L}^{-1}$ 和 $11.70 \mu\text{g}\cdot\text{L}^{-1}$, DEHP、DnBP 和 DiBP 是其中含量最高的 PAEs 同系物。类似的研究发现, 在青岛市的 3 家污水处理厂中, 16 种 PAEs 的平均浓度在进水中为 $0.13\text{--}0.19 \mu\text{g}\cdot\text{L}^{-1}$, 在出水中降至 $0.04\text{--}0.06 \mu\text{g}\cdot\text{L}^{-1}$ ^[34]。上述研究结果表明, 虽然污水处理过程可以显著降低出水中 PAEs 的含量, 但仍然难以完全去除, 这可能会导致地表和地下水源的二次污染, 并对饮用水源构成威胁^[35]。

湖泊属于缓流水体, 有利于 PAEs 的累积。研究表明, 我国湖泊中 PAEs 的浓度分布呈现出明显的季节差异。梁子湖^[36]、千岛湖^[37]和太湖^[38]中 PAEs 的平均浓度在枯水期分别为 0.69 、 2.63 、 $0.92 \mu\text{g}\cdot\text{L}^{-1}$, 而在丰水期上升至 1.52 、 7.99 、 $4.31 \mu\text{g}\cdot\text{L}^{-1}$, DiPB 和 DEHP 是其中主要的 PAEs。这种浓度差异可能与季节变化有关, 不同季节的降水量和人类活动频率存在显著差异。此外, 我国的南海地区^[39]、东海海域^[40]、汉江^[41]、松花江^[42]、长江^[43~44]、黄河^[45]和珠江流域^[46~47]也检测到了不同程度的 PAEs 污染, 尤其是 DBP、DiBP 和 DEHP。综上所述, PAEs 在我国水环境中普遍存在, 其分布和浓度受到季节变化、地理位置和人类活动等多种因素的影响。

1.3 土壤

随着我国农业现代化的持续推进, PAEs 被广泛添加到肥料、农药和塑料薄膜中。这种做法在提高农业生产效率的同时, 不可避免地加剧了土壤中 PAEs 的污染。研究指出, 我国不同地区土壤中 PAEs 的浓度普遍处于全球较高水平, 且在多数地区, PAEs 的总量超过了《土壤环境质量标准》(GB 15618—1995) 中规定的浓度限值^[48–50]。一项研究调查了我国茶园土壤中 PAEs 的污染现状, 发现 5 种 PAEs 的平均浓度为 $1.04 \mu\text{g}\cdot\text{g}^{-1}$, 且北方地区的污染水平明显高于南方, 这一现象可能与当地的施肥量与地膜的使用情况有关^[51]。在贵州省的烟草种植基地, 土壤中 6 种 PAEs 的平均浓度高达 $5.40 \mu\text{g}\cdot\text{g}^{-1}$, 主要污染来源是地膜、肥料和农药^[52]。青岛市^[53] 和黄河三角洲地区的农田土壤^[54] 中也检测到了 PAEs 的存在, 其中 DEHP 和 DBP 是研究区域土壤中最主要的 PAEs 单体。上述研究表明我国农业土壤中 PAEs 的污染分布广泛, 且形势较为严峻。据统计, 我国是农用地膜产量和使用量最高的国家之一^[55], 地膜的应用虽然提高了土壤的温度、湿度和养分利用效率, 但同时也增加了土壤中 PAEs 的残留^[56–58]。例如, 在甘肃省开展的一项研究中发现, 温室土壤($1.04 \mu\text{g}\cdot\text{g}^{-1}$) 和露地农田($0.44 \mu\text{g}\cdot\text{g}^{-1}$) 中 PAEs 的平均含量明显高于森林($0.14 \mu\text{g}\cdot\text{g}^{-1}$) 和草原($0.06 \mu\text{g}\cdot\text{g}^{-1}$)^[59]。在南京市的辣椒种植区^[60], 地膜覆盖区域土壤中 PAEs 的总浓度($0.94 \mu\text{g}\cdot\text{g}^{-1}$) 是未覆盖地膜区域($0.34 \mu\text{g}\cdot\text{g}^{-1}$) 的近 3 倍。吉林省人参种植基地也显示出类似的趋势, 塑料大棚内土壤中 PAEs 的残留水平($56.80 \mu\text{g}\cdot\text{g}^{-1}$) 远高于未覆盖地膜的土壤($1.65 \mu\text{g}\cdot\text{g}^{-1}$)^[61]。这些研究结果表明, 地膜的使用是导致农田土壤中 PAEs 积累的主要原因。

与农田土壤的污染特征不同, 城市土壤中 PAEs 的含量受到多种因素的综合影响。首先, 城市功能分区会影响 PAEs 的污染水平。例如, 在重庆市, 城市公园和商业区土壤中 PAEs 的含量最高($0.31 \mu\text{g}\cdot\text{g}^{-1}$), 其次是工业区($0.30 \mu\text{g}\cdot\text{g}^{-1}$), 而住宅区($0.23 \mu\text{g}\cdot\text{g}^{-1}$) 相对较低^[62]。这一发现表明不同城市区域的功能差异导致了土壤中 PAEs 污染水平的差异。其次, 人类活动对城市土壤中 PAEs 的污染程度也有重要影响, 尤其是在高人流和高物流的地区, 如杭州市的西湖风景区, 游客活动和交通运输明显加剧了该地区 PAEs 的污染水平^[63]。此外, 塑料产品市场周边、电子制造业周边、污水灌溉区、电子垃圾拆解厂及回收区等^[64–68] 特定场所也存在较高水平的 PAEs 污染。这些研究结果反映了城市土壤中 PAEs 污染的复杂性和多样性, 应当引起有关部门的重视。

1.4 食品

PAEs 是一类脂溶性塑化剂。在我国, DBP 和 DEHP 是食品中最常见的 PAEs 同系物^[69]。根据《食品安全国家标准》(GB 9685—2016) 规定, 食品接触材料及制品用添加剂中 DBP 和 DEHP 的含量不得超过 $0.30 \mu\text{g}\cdot\text{g}^{-1}$ 和 $1.50 \mu\text{g}\cdot\text{g}^{-1}$ ^[70]。然而, 在实际监测中, 这两种化合物的残留量通常超过了上述的标准限值。例如, 食品接触纸及制品中 DBP 和 DEHP 的检出范围分别在 $2.34\text{--}2.65 \mu\text{g}\cdot\text{g}^{-1}$ 和 $1.19\text{--}18.0 \mu\text{g}\cdot\text{g}^{-1}$ 之间^[71]。德阳市塑料包装食品中 DBP 的最大残留浓度为 $1.46 \mu\text{g}\cdot\text{g}^{-1}$ ^[72], 湖州市塑料包装黄酒中 DBP 的平均浓度为 $0.37 \mu\text{g}\cdot\text{g}^{-1}$ ^[73], 河南省市售的食用植物油中 DBP 和 DEHP 的超标率分别为 21.6% 和 3.6%^[74]。此外, 冯艳红等研究发现, 农产品中也普遍存在 DBP 和 DEHP 污染^[75]。在江苏省邳州市的大蒜产区^[76], 大蒜蒜瓣中这两种化合物的检出率最高, 平均浓度分别为 $0.61 \mu\text{g}\cdot\text{g}^{-1}$ 和 $0.17 \mu\text{g}\cdot\text{g}^{-1}$ 。在北京市的温室蔬菜生产基地^[77], DEHP、DiBP 和 DnBP 合计占 PAEs 总量的 90% 以上。上述研究结果表明, 各类食品中普遍存在 PAEs 污染, 且多数地区食品接触材料中 DBP 和 DEHP 的残留浓度超过了安全标准限值, 提示我们应采取有效措施减少食品及食品接触材料中 PAEs 的含量。

2 PAEs 的人体内暴露水平(Human exposure levels of PAEs)

环境中的 PAEs 可通过呼吸道、消化道和皮肤接触等途径进入人体, 并在体内发生代谢反应。其中, 短链 PAEs 会被迅速水解, 转化为邻苯二甲酸单酯, 即初级代谢物; 而长链 PAEs 在转化为单酯后, 还会进一步被氧化成更亲水的次级代谢物^[78–79]。这些代谢物在人体内分布, 并可通过尿液或粪便排出。目前, 研究人员已在尿液、血液、母乳和精液等生物样本中检测出多种 mPAEs, 并以此反映体内 PAEs 的实际负荷水平。

2.1 尿液

尿液是研究人体内 PAEs 暴露水平最常用的生物样本, 具有收集简单、样本量大且无侵入性伤害

等优势。通过对尿液样本中的 mPAEs 进行筛选和定量，可以反映出 PAEs 在体内的实际吸收水平，即内暴露水平^[80]。我国不同地区居民体内 PAEs 的负荷水平如表 2 所示。研究发现，儿童、孕妇以及生活在工业化程度较高地区的人群更容易接触到 PAEs。Huang 等^[81]从中国的 26 个省会城市收集了千余份晨尿样本，观察到不同年龄段的人群中 mPAEs 的浓度存在显著差异，儿童和青少年尿液中 mPAEs 的浓度最高。Wang 等^[82]分析了我国 8 岁至 11 岁小学生尿液样本中 9 种 mPAEs 的含量，发现上海市、南通市和台州市的小学生普遍暴露于 PAEs，其中，南通市和台州市等制造业密集地区儿童尿液中 mPAEs 的含量更高。同样，Yao 等^[83]的研究结果显示，深圳市 6 岁至 8 岁儿童也普遍暴露于 PAEs，并且所调查区域儿童尿液中 MEP、MiBP 和 DEHP 代谢物的浓度与家庭收入呈正相关性。Dong 等^[84]研究发现，上海市 2 岁到 19 岁儿童在成长过程中面临的 PAEs 暴露水平较高，这可能与其在此阶段频繁接触塑料制品有关。此外，儿童的个人行为（如地面爬行、手口接触、吮吸塑料玩具等）和生活方式（室内活动时间长）也是其体内 PAEs 水平较高的主要原因^[85-86]。

表 2 我国居民尿液中 mPAEs 的中值浓度

Table 2 The median concentration of mPAEs in urine of Chinese residents

地理地区 Region	人群 Population	单位 Unit	MBP	MMP	MEP	MiBP	MnBP	MBzP	MEHP	MEOHP	MEHHP	MECPP	文献 References	
华东	上海市	儿童	$\mu\text{g}\cdot\text{L}^{-1}$	—	11.40	10.10	46.20	58.10	0.20	4.50	14.50	24.70	39.00	[86]
	上海市	婴儿	$\mu\text{g}\cdot\text{g}^{-1}$	—	54.61	234.79	465.79	344.01	9.38	207.03	29.68	42.50	77.40	[108]
	上海市	乳母	$\mu\text{g}\cdot\text{g}^{-1}$	—	28.48	44.12	123.76	105.17	1.07	24.71	9.83	20.19	16.05	
	上海市	成人	$\mu\text{g}\cdot\text{g}^{-1}$	—	2.93	8.82	8.42	12.92	2.07	8.84	4.67	11.64	13.95	[109]
	上海市	成人	$\mu\text{g}\cdot\text{g}^{-1}$	—	2.69	8.33	7.61	11.50	1.99	8.72	4.38	10.96	13.15	[110]
	厦门市	儿童	$\mu\text{g}\cdot\text{L}^{-1}$	172.6	32.78	8.07	23.16	—	—	8.10	3.52	17.72	—	[111]
	厦门市	新生儿	$\mu\text{g}\cdot\text{L}^{-1}$	17.5	0.00	3.31	—	—	0.00	0.22	0.48	0.00	—	[112]
	舟山市	婴幼儿	$\mu\text{g}\cdot\text{L}^{-1}$	7.91	3.39	3.31	13.64	—	—	3.06	1.32	1.51	9.10	[113]
	台湾省	儿童	$\mu\text{g}\cdot\text{g}^{-1}$	—	9.70	7.40	16.20	30.20	1.20	80.00	15.50	24.10	—	[114]
	马鞍山市	儿童	$\mu\text{g}\cdot\text{g}^{-1}$	257.73	29.65	26.65	—	—	0.12	6.27	17.94	24.80	—	[115]
华北	杭州市	妇女	$\mu\text{g}\cdot\text{g}^{-1}$	—	13.37	4.18	26.17	21.25	0.01	2.01	2.09	5.38	—	[116]
	六安市	老年人	$\mu\text{g}\cdot\text{g}^{-1}$	42.76	3.36	2.26	—	—	0.65	0.47	2.17	2.35	—	[117]
	六安市	老年人	$\mu\text{g}\cdot\text{L}^{-1}$	43.64	3.51	2.30	—	—	0.80	0.54	2.25	2.59	—	[118]
	马鞍山市	孕妇	$\mu\text{g}\cdot\text{g}^{-1}$	47.65	11.97	7.97	—	—	0.40	0.08	2.50	6.64	—	[119]
	马鞍山市	孕妇	$\mu\text{g}\cdot\text{g}^{-1}$	47.27	11.99	7.94	—	—	0.08	2.50	6.61	4.79	—	[120]
	天津市	孕妇	$\mu\text{g}\cdot\text{g}^{-1}$	37.10	46.50	15.20	10.60	—	7.85	20.90	0.46	0.89	4.82	[121]
	天津市	大学生	$\mu\text{g}\cdot\text{L}^{-1}$	17.30	6.34	11.40	7.65	—	—	0.82	1.36	3.21	3.96	[122]
	天津市	儿童	$\mu\text{g}\cdot\text{L}^{-1}$	—	—	18.36	31.60	26.24	0.09	7.50	9.61	16.71	46.12	[123]
	北京市	孕妇	$\mu\text{g}\cdot\text{g}^{-1}$	—	4.22	17.60	20.90	51.00	—	5.03	—	—	—	[124]
	武汉市	孕妇	$\mu\text{g}\cdot\text{g}^{-1}$	206.00	20.30	15.00	—	—	—	3.14	13.80	15.80	17.60	[89]
华中	武汉市	孕妇	$\mu\text{g}\cdot\text{L}^{-1}$	87.20	5.78	8.55	—	—	0.07	2.26	5.96	7.08	9.72	[125]
	武汉市	妇女	$\mu\text{g}\cdot\text{L}^{-1}$	—	—	11.32	18.75	62.08	0.10	2.45	4.74	6.33	9.57	[126]
	襄阳市	儿童	$\mu\text{g}\cdot\text{L}^{-1}$	61.90	32.00	18.00	—	—	2.77	6.11	8.27	15.40	—	[127]
	十堰市	儿童	$\mu\text{g}\cdot\text{g}^{-1}$	—	17.90	13.50	10.70	33.60	2.80	4.20	6.80	15.80	10.80	[128]
	鄂州市	孕妇	$\mu\text{g}\cdot\text{L}^{-1}$	—	31.92	40.80	19.50	56.60	2.15	8.64	66.10	62.60	—	[129]
	鄂州市	孕妇	$\mu\text{g}\cdot\text{L}^{-1}$	30.6	17.08	—	—	—	3.80	8.26	—	—	—	[130]
	深圳市	儿童	$\mu\text{g}\cdot\text{g}^{-1}$	—	22.60	13.80	36.80	212.00	—	7.30	12.80	22.20	—	[83]
	深圳市	初产妇	$\mu\text{g}\cdot\text{L}^{-1}$	139.00	4.59	4.87	—	—	0.24	2.86	4.31	5.45	—	[88]

续表 2

地理地区 Region	人群 Population	单位 Unit	MBP	MMP	MEP	MiBP	MnBP	MBzP	MEHP	MEOHP	MEHHP	MECPP	文献 References	
华中	深圳市	儿童	$\mu\text{g}\cdot\text{L}^{-1}$	63.60	24.70	0.22	25.30	—	0.46	11.10	38.40	38.90	—	[131]
	广州市	孕妇	$\mu\text{g}\cdot\text{g}^{-1}$	51.80	16.70	8.17	33.80	—	—	2.35	3.34	4.19	7.36	[132]
	广州市	成人	$\mu\text{g}\cdot\text{L}^{-1}$	240.12	6.15	9.95	37.63	—	—	13.67	12.39	16.99	—	[133]
西南	遵义市	孕妇	$\mu\text{g}\cdot\text{g}^{-1}$	102.00	2.28	12.50	29.20	—	0.06	7.05	7.48	9.56	71.40	[134]
东北	吉林省	大学生	$\mu\text{g}\cdot\text{L}^{-1}$	33.11	—	21.56	—	—	—	2.55	4.79	3.40	9.97	[135]

注: “ $\mu\text{g}\cdot\text{L}^{-1}$ ”代表使用尿比重校正法校正的质量浓度; “ $\mu\text{g}\cdot\text{g}^{-1}$ ”代表使用尿肌酐校正法校正的质量浓度; “—”代表未检验。

Note: “ $\mu\text{g}\cdot\text{L}^{-1}$ ” represents the mass concentration corrected using the urine specific gravity correction method; “ $\mu\text{g}\cdot\text{g}^{-1}$ ” represents the mass concentration corrected using the urine creatinine correction method; “—” indicates not tested.

孕妇尿液中 mPAEs 的暴露水平也是一个值得关注的问题, 因为此类化合物可能会通过母亲传递给胎儿, 进而影响婴儿的生长发育。通常情况下, 女性在怀孕后会减少个人护理用品的使用频率, 因此, 其尿液中 mPAEs 的含量和类型主要受 PAEs 的环境污染特征(以 DEHP 和 DBP 为主)所影响。例如, He 等^[87] 收集了上海市孕妇的尿液样本, 发现在所有检测到的 mPAEs 中, MBP 的浓度最高(中值浓度: $25.29 \mu\text{g}\cdot\text{L}^{-1}$)。这些发现与在深圳^[88]、武汉市^[89]、遵义市^[90-91]和马鞍山市^[92]进行的出生队列研究结果一致, 表明 MBP(DBP 的代谢物)是孕妇体内主要的 PAEs 化合物。另一项研究在广州市区^[93]进行, 同样发现孕妇尿液中检出的 PAEs 以 MBP 为主, 中值浓度为 $47.20 \mu\text{g}\cdot\text{L}^{-1}$, 该研究还指出, 与新型的 mPAEs 相比, 传统的 mPAEs 在孕妇尿液中占比更高。

工作类型和办公环境也是人体内 PAEs 暴露水平的重要影响因素。Li 等^[94] 检测了天津市某电子垃圾回收厂工人体内 PAEs 的含量, 发现园区工人尿液中 mPAEs 的浓度(中值浓度: $389.00 \mu\text{g}\cdot\text{L}^{-1}$)明显高于附近居民(中值浓度: $285.00 \mu\text{g}\cdot\text{L}^{-1}$)。这一趋势在深圳市某垃圾焚烧厂^[95]和广东省某电子垃圾收集区^[96]也有所体现。表明与非直接暴露的居民相比, 电子垃圾回收和处理场所的工作人员更容易受到 PAEs 的暴露。Wang 等^[97] 研究发现, 从事废塑料回收工作也会提高工人尿液中 mPAEs 的水平, 这可能与他们在手动分离、清洗、粉碎和收集废塑料的过程中频繁接触 PAEs 有关。此外, 由于 PAEs 在个人护理产品中的普遍应用, 某百货商店化妆品和香水柜台销售人员在换岗后尿液中 mPAEs 的浓度显著升高^[98]。综上所述, PAEs 在不同人群中的暴露水平存在差异, 通常与环境中 PAEs 的污染特征密切相关。

尿液中 mPAEs 的浓度也可用于比较不同国家人群中 PAEs 的暴露水平。在韩国育龄妇女中, MEP 是最主要的 PAEs 代谢物, 其在尿液样本中的几何平均值为 $7.72 \mu\text{g}\cdot\text{L}^{-1}$ ^[99]。在瑞典人群的尿液样本中, 浓度最高的 mPAEs 同样是 MEP, 几何平均值为 $70 \mu\text{g}\cdot\text{g}^{-1}$ ^[100]。澳大利亚昆士兰州的相关研究也发现了这一趋势, 但该地区居民尿液中 MEP 的平均浓度($150 \pm 100 \mu\text{g}\cdot\text{L}^{-1}$)明显高于前两个国家^[101]。由于 MEP 的母体化合物 DEP 主要存在于化妆品和个人护理用品中^[102], 因此, 发达国家人群对于化妆品和个人护理产品的高使用频率可能是其尿液中 MEP 浓度较高的主要原因。相比之下, 发展中国家的人群(如中国居民^[103]、印度妇女^[104]、俄罗斯^[105]和巴西^[106]的学龄儿童)则更容易接触到 DBP 和 DEHP^[107], 这可能与上述地区居民对含有 PAEs 的塑料产品的高需求量有关。这些发现可以帮助我们了解不同人群中 PAEs 内暴露水平的差异性, 并进一步揭示了可能的暴露途径。

2.2 血液

与尿液相比, 血液中 mPAEs 的含量能够更准确地反映人体内 PAEs 的暴露水平。然而, 科研用途下采集血液样本的条件较为复杂, 目前关于血液中 PAEs 暴露水平的研究相对较少。重庆市交警血清中 PAEs 的含量会随着空气污染程度的加剧而升高, 这表明环境中 PAEs 的污染分布可能是影响人体血液中 PAEs 含量的主要因素^[136]。在浙江省衢州市的研究中发现, 由于频繁使用个人护理产品和化妆品, 女性血清中 mPAEs 的平均浓度通常高于男性^[137]。此外, 一些研究报道了孕妇血清和胎儿脐带血中 PAEs 的暴露水平。在天津市, 孕妇血清中主要的 PAEs 代谢物是 MBP 和 MiBP, 其中值浓度分别为 $1.30 \mu\text{g}\cdot\text{L}^{-1}$ 和 $5.58 \mu\text{g}\cdot\text{L}^{-1}$ ^[121]。而在杭州市, 孕妇血清中浓度最高的 PAEs 是 DEHP(中值浓度: $35.46 \mu\text{g}\cdot\text{L}^{-1}$) 和 DBP(中值浓度: $17.82 \mu\text{g}\cdot\text{L}^{-1}$), 这一结果与该地区环境中 PAEs 的主要污染特征相符(以 DEHP 和

DBP 为主)^[138]. 另一些研究进一步发现母亲与其胎儿之间的 PAEs 暴露水平存在显著相关性. 例如, 重庆市胎儿脐带血中 PAEs 的浓度增加可能与孕妇接受静脉输注治疗有关^[139]. 在广州市花都区, 胎儿脐带血中 4 种 mPAEs 的浓度与其母亲血液中 mPAEs 的浓度呈正相关, 表明胎儿在子宫内已经暴露于 PAEs^[140]. 在哈尔滨市, 新生儿脐带血中检出了 11 种不同的 PAEs, 这可能与其母亲在怀孕期间使用化妆品和接触塑料产品有关^[141]. 这些研究结果强调了在孕期接触 PAEs 可能会增加胎儿的暴露风险. 因此, 识别和减少孕妇 PAEs 的暴露途径对于保护胎儿健康具有重要意义.

2.3 母乳及羊水

母乳是婴儿生长发育的重要营养来源. 它不仅富含丰富的蛋白质、碳水化合物和脂肪, 同时也可能含有一些环境化学物质. Deng 等^[142]首次在人类母乳样本中同时检测到 11 种 PAEs、24 种 mPAEs 和 14 种 PAEs 的替代品, 在这些检出物中, mPAEs 的中值浓度($11.20 \mu\text{g}\cdot\text{L}^{-1}$)显著高于其对应的母体化合物($6.99 \mu\text{g}\cdot\text{L}^{-1}$). 李海玲等^[143]调查了哈尔滨市母乳样本中 PAEs 的暴露水平, 发现 PAEs 和 mPAEs 的中值浓度分别为 $87.30 \mu\text{g}\cdot\text{L}^{-1}$ 和 $21.80 \mu\text{g}\cdot\text{L}^{-1}$, 并指出母乳可能是婴儿 PAEs 暴露的重要途径. 此外, 天津市的一项研究发现, 母亲血清中 mPAEs 的种类与胎儿羊水中的相似, 表明 PAEs 及其代谢物能够从母体血液转移到羊水中^[121]. 这些研究结果揭示了母婴之间存在潜在的 PAEs 传递途径, 但仍需开展进一步的研究来探索 PAEs 的母婴传递机制及其健康影响.

2.4 精液

睾丸是 PAEs 的靶器官之一, 精液中 PAEs 和 mPAEs 的含量被认为能间接反映其对生殖系统的潜在毒性影响. 徐廷云等^[144]研究发现, 苏州地区男性精液中 PAEs 的主要组分是 DiBP 和 DEHP, 其中, 不育男性精液中 PAEs 的单体含量和总量均高于正常生育组的男性. Wang 等^[145]同样观察到不育男性精液中 PAEs 含量较高这一现象, 并进一步指出 PAEs 的累积含量可随年龄增长而显著增加. 上述研究表明 PAEs 可能与男性不育之间存在一定关联. 此外, 在武汉市的男性精液样本中, mPAEs 的浓度主要与生活方式有关, 例如吸烟、静脉输液、使用洗面奶和面霜等^[146], 这些日常生活习惯可能会导致精液中特定 PAEs 的浓度增加, 进一步影响男性的生殖健康.

3 PAEs 的健康效应(Health effects of PAEs)

PAEs 是最常见的环境内分泌干扰物之一, 其与类固醇激素结构相似, 可以作为体内激素受体的配体, 或干扰激素的合成、释放及代谢过程, 进而对激素的平衡产生影响, 导致内分泌系统的功能紊乱, 产生多种生理和代谢问题^[147–149]. 此外, 研究报告显示, 频繁接触 PAEs 可能会增加哮喘及过敏性疾病的风险^[150–151]. 考虑到不同国家与地区居民之间存在地理和种族差异, 本研究重点关注我国居民接触 PAEs 的潜在健康风险.

3.1 胎儿不良妊娠结局

PAEs 及其代谢物呈脂溶性, 可以通过母体血液进入胎儿体内, 进而影响胎儿的生长发育. 大量流行病学研究表明, 妊娠期接触 PAEs 与多种不良妊娠结局有关, 包括流产、早产、胎儿宫内窘迫、死胎、畸胎和低出生体重等^[152]. 在我国一些地区开展的大规模病例对照研究中发现, 流产妇女的 PAEs 暴露水平显著高于正常妊娠妇女, 例如北京^[124]、南京^[153]、台湾^[154–155]、上海和浙江^[156]等地. 在广西的出生队列中, 产前 PAEs 暴露可能会导致胎儿生长受限^[157–158]. 此外, 研究发现妊娠期频繁接触 PAEs 还可能会增加胎儿畸形的风险^[159].

3.2 儿童生长发育异常

儿童期接触 PAEs 可能会对儿童的生长发育产生负面影响. 研究表明, PAEs 与神经发育损伤以及青春发动时相改变有一定关联^[148,160–161]. 广西柳州市儿童尿液中监测到的 DEHP 代谢物(MEHHP、MEOHP、MEHP)与儿童注意力缺陷多动障碍有关^[162]. 马鞍山市的一项队列研究发现, 儿童暴露于 PAEs 会阻碍其执行能力的发展^[115]. 另一项在天津市开展的研究中发现, 儿童频繁接触 PAEs 会加重自闭症谱系障碍的症状^[163], 这进一步支持了 PAEs 对神经发育有损伤作用. 此外, 另一些研究表明, 出生后暴露于 PAEs 可能会干扰儿童的青春发动时相, 从而影响他们的性征发育和身体变化^[164–165]. 邓敏等^[166]观察到性早熟女童血清中 DEHP 的浓度明显升高, 这可能与 PAEs 的内分泌干扰作用有关. Zhang 等^[167]

研究发现,女童乳房发育加快和月经初潮提前与其尿液中高浓度的 MEHP 有关。综上所述,出生前(妊娠期)和出生后(儿童期)暴露于 PAEs 都可能会影响儿童的正常生长发育。因此,不仅需要进行早期的筛查和监测,还应该采取有效的预防措施来减少孕妇和儿童接触 PAEs 的风险。

3.3 成人生殖功能障碍

多项研究表明,PAEs 的暴露可能会影响成人的生殖健康,主要表现为男性的精液质量下降、精子数量减少、睾丸发育不良,以及女性的卵巢功能不全、卵巢早衰、异位妊娠,甚至是生殖系统肿瘤等^[168–170]。PAEs 是个人护理用品中常见的添加剂,频繁使用指甲油、香水和化妆品会增加女性 PAEs 的内暴露水平。卵巢在女性的生殖内分泌系统中起着关键作用。已有研究证实,长期接触 PAEs 可能会增加卵巢早衰的风险,导致卵巢的储备功能下降,进而影响女性的生殖健康^[171]。Zhang 等的研究表明,我国育龄妇女接触 PAEs 会促进多囊卵巢综合征的发展^[172]。流行病学研究发现,子宫内膜异位症患者血清中 PAEs 的浓度显著升高^[173–174],提示子宫内膜异位症的发病可能与 PAEs 暴露有关。此外,PAEs 暴露也被认为可能会增加生殖系统肿瘤的发生风险,如子宫肌瘤、卵巢癌、乳腺癌和子宫内膜癌等^[175–178]。

睾丸是男性最主要的生殖器官,大量的动物研究已经证实了 PAEs 暴露与男性生殖功能障碍之间的直接关联^[179]。流行病学研究发现,胎儿期暴露于 PAEs 与男婴睾丸发育不良综合征有关^[180],这表明母亲在怀孕期间接触 PAEs 可能会导致男婴的生殖器官发育异常。此外,PAEs 暴露会对男性的精液质量产生显著不良影响,成年男性接触 PAEs 可能会破坏精子 DNA 的完整性、诱导生殖细胞凋亡,进而导致精子活力下降^[181–183]。综上所述,PAEs 对男性生殖健康的负面影响主要表现在生殖器官发育异常以及精液质量下降等方面。这些研究发现强调了减少接触 PAEs 以维护成人生殖健康的重要性。

3.4 代谢系统疾病

近年来的流行病学研究显示,暴露于 PAEs 可能会增加人体发生代谢紊乱的风险。在我国,居民接触 PAEs 与超重、肥胖、心血管疾病、高血压和糖尿病之间存在密切关联。徐缙等^[184]研究发现,PAEs 可能会促进人群肥胖的发生。另一项研究支持这一观点,即男童在接触 PAEs 后体重、体质指数和体脂率升高^[185]。Dong 等^[111] 和 Gao 等^[186] 分别探讨了我国儿童 PAEs 暴露与肥胖和血脂异常之间的关联。研究结果显示,发育早期接触 PAEs 会显著增加儿童超重、肥胖和血脂异常的风险。由于 PAEs 具有亲脂性,因此,从环境介质转移到人体组织中的 PAEs 及其代谢物容易在脂肪组织中累积。这种累积可能会影响脂肪细胞的能量代谢,进而导致血脂和血糖水平异常^[187–188]。马鞍山市的一项队列研究发现,妊娠早期接触单一或多种 PAEs 可能会导致孕妇的血糖和血压升高,并增加其罹患代谢综合征的风险^[189]。另一些研究发现,PAEs 的内暴露水平与糖尿病的发病率相关。例如,上海市中老年糖尿病患者尿液中 mPAEs 的水平明显高于健康人群^[190]。同样,天津市志愿者尿液中 mPAEs 的浓度与 II 型糖尿病之间存在显著的正相关关系^[191]。此外,PAEs 暴露还可能会导致儿童或成人的血压升高,甚至可能进一步诱发心脑血管疾病^[192–194]。总之,当前的多数研究都支持 PAEs 在肥胖、高血压和糖尿病的发生和发展中起到的促进作用。因此,公众应增强意识,减少 PAEs 暴露,并以此作为预防代谢性疾病发生和进展的一种策略。

3.5 其它

PAEs 暴露可能会触发人体的免疫应答,导致炎症反应、过敏症状、鼻炎和哮喘等健康问题。马鞍山市的一项队列研究发现,孕妇在妊娠期接触 PAEs 可能会促进婴儿哮喘的发展^[195]。在上海市的一项研究中,高浓度高分子量的 PAEs 与女孩的哮喘^[196] 和男孩的鼻炎诊断^[197] 显著相关。由于 PAEs 被用于多种产品中,尤其是儿童玩具和食品包装,这使得儿童更容易接触到此类物质,从而增加了他们呼吸道疾病的患病风险。此外,PAEs 暴露还可能会对肺功能产生不良影响。研究表明,长期接触 PAEs 可导致儿童^[198] 和成人^[199] 的肺功能损伤。值得注意的是,目前关于 PAEs 暴露与疾病发生发展之间关联的研究大多数是基于病例对照研究结果所示。虽然这种研究方法可以揭示它们的相关性,但却无法确立因果关系。因此,未来需要开展更多的队列研究,以进一步证实 PAEs 暴露对人体健康的具体影响。

4 结论及展望(Conclusions and prospects)

本文系统总结了我国 PAEs 的环境污染现状,不同人群的 PAEs 内暴露水平,并探讨了 PAEs 对我

国居民健康的负面影响。研究结果表明, PAEs 在我国环境介质中普遍存在, 以 DEHP 和 DBP 为主。同时, 有报告指出, 我国居民体内一些特定 mPAEs 的负荷水平相对较高, 如 MBP、MiBP、MnBP 和 MEHP, 这与个人的生活方式、居住环境和职业类别等多种因素相关。此外, 作为环境污染物的敏感人群, 儿童和孕妇呈现出较高的 PAEs 暴露风险, 具体而言, PAEs 暴露与胎儿不良妊娠结局、儿童生长发育异常、成人生殖功能障碍、肥胖症、高血压、糖尿病和过敏性炎症等健康问题存在潜在关联。

环境化学物质的人体内暴露研究是近年来环境领域的研究热点。当前, 人们对于 PAEs 的接触已从单体向多种物质的混合物进行转变。因此, 在未来的科学的研究中, 应当将 PAEs 的风险评估重点从单一化学物质向多源、多类别及多作用途径的综合评估转变。同时, 应当鼓励开展流行病学与环境科学等多学科的交叉研究, 推动基于内、外暴露的综合研究发展, 以期更全面地了解环境化学物质对人体健康的潜在风险。

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