

人体有机磷酸酯内暴露水平及相关健康风险的研究进展

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摘要

有机磷酸酯(organophosphate esters, OPEs)作为新型阻燃剂与增塑剂, 凭借其广泛的工业与商业应用, 在全球环境中广泛分布。鉴于其易释放和持久存在的特性, OPEs已成为环境及人体中普遍检出的新兴污染物。现有研究显示, OPEs可通过吸入、摄入以及皮肤接触等多途径进入人体, 已在尿液、血液、母乳、指甲及头发等多种生物基质中被检测到。此外, 流行病学研究提示OPEs暴露与多种不良健康结局相关。因此, 本文对不同地区、性别、年龄及职业人群的暴露水平展开综述, 剖析人体内OPEs的暴露特征, 旨在为系统评估OPEs的暴露风险以及制定环境防控与健康管理策略提供科学依据。

关键词

有机磷酸酯, 人群暴露, 生物监测, 健康风险, 环境流行病学

Human Internal Exposure to Organophosphate Esters and Associated Health Risks: A Review

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Abstract

Organophosphate esters (OPEs), widely used as novel flame retardants and plasticizers, have become globally distributed in the environment due to their extensive industrial and commercial

applications. Owing to their tendency to leach and their persistence, OPEs have emerged as ubiquitous environmental contaminants detected in both environmental media and the human body. Current studies indicate that OPEs can enter the human body through multiple pathways, including inhalation, ingestion, and dermal absorption, and can be detected in various biological matrices such as urine, blood, breast milk, nails, and hair. Epidemiological evidence further suggests that OPE exposure is associated with multiple adverse health outcomes. This review summarizes the exposure levels of OPEs among populations of different regions, sexes, ages, and occupations, aiming to elucidate the characteristics of internal exposure and provide scientific evidence for comprehensive risk assessment, environmental control, and health management strategies regarding OPEs.

Keywords

Organophosphate Esters, Human Exposure, Biomonitoring, Health Risks, Environmental Epidemiology

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1. 前言

有机磷酸酯(organophosphate esters, OPEs)作为磷酸的衍生物,其结构由中心磷酸盐分子和异相取代基构成[1]。鉴于 OPEs 具备优异的阻燃性与增塑性能,在工业和商业领域有着广泛用途,已被应用于电子、塑料、汽车部件、建筑材料、家具、石油、纺织品等各类产品中,主要用作阻燃剂和增塑剂[2]。随着部分溴化阻燃剂被《斯德哥尔摩公约》列为持久性有机污染物并逐步淘汰,OPEs 作为合适的替代品,其需求和产量稳步增长。全球 OPEs 销量从 2011 年的 50 万吨增至 2018 年的 1 亿吨以上[3]。中国是全球 OPEs 产量和消费量最高的国家之一,2020 年 OPEs 年产量约为 36 万吨[4]。

OPEs 的环境赋存问题日益受到关注。研究显示,OPEs 已广泛存在于水体、沉积物、土壤、空气以及室内外灰尘等多种环境介质中[5]。且部分氯代 OPEs 具有环境持久性,能够长期存在于生态系统中[6]。而环境中的 OPEs 可通过多种途径进入人体,引发潜在的健康风险。近年来,有关 OPEs 人群暴露和健康效应的研究不断增多。系统了解人群的内暴露水平对于评估 OPEs 对人群的健康影响具有重要的意义。因此,本文对目前人体内 OPEs 的暴露特征以及人群的健康风险进行综述,以期今后的研究提供参考。

2. 暴露方式

环境中的 OPEs 主要通过三种途径进入人体,即摄入、吸入和皮肤吸收。摄入途径包括食物和饮料的摄入以及灰尘的摄入等;吸入主要来源于空气中的 OPEs;皮肤吸收则与皮肤接触含有此类化学物的产品有关。由于不同 OPEs 的理化性质以及人群中个体生活方式差异,各暴露途径对人体总量暴露的贡献比例是不同的。尽管每种暴露途径都不应被低估,但多项研究表明膳食摄入是人群 OPEs 暴露的主要来源[7][8]。例如,比利时的一项研究报道成年人经膳食暴露 OPEs 的每日摄入量为 $103 \text{ ng} \cdot \text{kg} \cdot \text{bw}^{-1} \cdot \text{day}^{-1}$ [9]。

3. 人体内 OPEs 的暴露特征

目前针对人体内 OPEs 暴露水平的研究日益增多,OPEs 已广泛在人体内检出。由于大多数 OPEs 在人体内代谢较快,因此尿液样本中的检出物主要为 OPEs 代谢物。其中,磷酸二(2-氯乙基)酯(Bis(2-chloroethyl)phosphate, BCEP)、磷酸二(1,3-二氯-2-丙基)酯(Bis(1,3-dichloro-2-propyl)phosphate, BDCIPP)和

磷酸二苯酯(Diphenyl phosphate, DPHP)是最常被报道的 OPEs 代谢物,其母体化合物分别为磷酸三(2-氯乙基)酯(Tris(2-chloroethyl)phosphate, TCEP)、磷酸三苯酯(Triphenyl phosphate, TPHP)和磷酸三(1,3-二氯异丙基)酯(Tris(1,3-dichloro-2-propyl)phosphate, TDCIPP)。不同人群在代谢功能、生活方式以及职业工作等方面具有差异,因此 OPEs 在不同人群中的暴露水平也呈现出不同的特征。

3.1. 时间分布

截至目前,关于人体内 OPEs 暴露水平随时间变化趋势的研究仍较有限。中国山东[10]的一项研究对这一趋势进行了初步探索。结果显示,2011 年血清样品中 Σ OPEs 浓度为 340~791 ng/g lw (平均 536 ng/g lw),2015 年的血清样品中 Σ OPEs 浓度为 415~778 ng/g lw (平均 605 ng/g lw),呈总体上升趋势。尤其是 TCEP,其 2015 年的浓度较 2011 年增加了约 13%。此外,瑞典的一项研究也观察到类似的变化,母乳中的 TCEP 浓度从 1998 年的 2.6 ng/g lw 升高至 2006 年的 8.1 ng/g lw [11]。因此人体内的 OPEs 水平可能随时间呈上升趋势。不过,鉴于相关研究数量有限,仍需开展进一步的长期监测与验证研究。

3.2. 地区分布

不同国家或地区人群体内 OPEs 的暴露水平差异很大。一项荟萃分析[12]汇总了来自五个国家动态样本(尿液、血清、母乳)中的 OPEs 水平,美国、中国、加拿大、日本和挪威的人群 OPEs 平均暴露水平分别为 0.60、1.28、0.51、0.68 和 0.40 ng/g,其中中国水平最高,约为挪威的三倍。然而,从单体化合物的暴露水平来看结果并不一致。例如,中国上海[13]人群尿液样本中 DPHP 中位浓度为 66 pg/mL,北京[14]尿液样本中为 0.18 ng/mL,而澳大利亚[15]、美国[16]和加拿大[17]等发达国家尿液中位浓度分别为 37、1.74 和 4.71 ng/mL,高出中国约 1~2 个数量级。以积累样本指甲为检测基质的两项研究的结果与上述一致。美国[18]普通人群指甲中 Σ OPEs 浓度(90~59,800 ng/g)远高于中国[19]普通人群(58.0~590 ng/g)。此外,美国普通人群指甲中的主要 OPEs 同系物是 TPHP,而中国普通人群则以磷酸三(1-氯异丙基)酯(Tri-(1-chloroisopropyl)phosphate, TCIPP)为主。这些结果均表明 OPEs 暴露水平具有显著的地域差异。而这些结果的差异不仅与样本量和检测方法有关,更与不同国家阻燃剂法规、OPEs 的生产和消费模式密切相关。

相对于全球背景水平,特定地区的 OPEs 暴露更为突出。例如,中国广东[20]人群尿液中 BCEP 的中位浓度为 1.04 μ g/L,远高于德国[21] (0.2 μ g/L)、加拿大[17] (0.46 μ g/L)和美国[22] (0.63 μ g/L)。主要原因是广东省为 TCEP 的主要生产和使用地区[23]。除地区差异外,人体 OPEs 内暴露还存在城乡差异。中国重庆[24]的一项研究显示,农村人群头发中 Σ OPEs 浓度范围为 42.0~4230 ng/g,而城市人群头发中的 Σ OPEs 为 820~6870 ng/g,平均值分别为 415 和 2380 ng/g。城市人群毛发中的 OPEs 浓度均显著高于农村($P < 0.05$)。相关研究认为,城市地区 OPEs 含量升高可能与工业、商业活动的强度和人口密度有关[25]。

3.3. 人群分布

3.3.1. 年龄分布

现有研究普遍表明,儿童体内的 OPEs 水平通常高于成年人。Ospina 等人[26]对美国国家健康与营养检查调查参与者尿液数据的分析显示,DPHP 在不同年龄段人群体内的浓度水平顺序为:6~11 岁 > 12~19 岁 > 20~59 岁。6~11 岁儿童和 12~19 岁青少年尿液中 BCEP 水平均显著高于 20~59 岁的成年人($P < 0.01$)。法国[27]的出生队列研究显示儿童头发中 TDCIPP 中位浓度(110 ng/g)约为母亲样本中浓度值(47 ng/g)的两倍。除特殊职业人群外,大多数研究显示 OPEs 在成人体内的水平与年龄也存在负相关关系。美国波士顿[28]一项针对 31 名成年上班族的研究发现,随着年龄的增长尿液中的 BDCIPP 水平逐渐降低(≤ 37 岁: 611 pg/mL; 38~56 岁: 376 pg/mL; ≥ 57 岁: 286 pg/mL)。类似地,针对挪威[29] 61 名 20~65 岁成年人的

研究发现,晨尿中 DPHP 和磷酸二(2-丁氧基乙基)羟乙酯(Bis(2-butoxyethyl)hydroxyethyl phosphate, BBOHEP)的浓度与参与者的年龄显著负相关($P = 0.02$)。

OPEs 水平与年龄之间的负相关可能是由于以下原因。首先,不同年龄阶段 OPEs 的酶活性和毒代动力学是不同的[30]。其次,不同年龄群体之间的新陈代谢率影响通过吸入和皮肤吸收的暴露量[31]。最后, OPEs 化合物对特定人群可能具有特定的暴露途径。例如,儿童特有的行为,如爬行和手口行为[32]可能导致室内灰尘及婴儿用品中的 OPEs 摄入体内而增加该年龄段的 OPEs 暴露。

3.3.2. 性别分布

多数研究表明女性体内的 OPEs 浓度普遍高于男性,这一趋势已在指甲、头发和尿液等不同生物样本基质中均有报道。例如,中国的一项研究[33]发现女性指甲中 Σ OPEs 浓度(193 ng/g)显著高于男性(123 ng/g) ($P < 0.05$),且在 TCEP 水平上结果一致;美国[18]的研究同样在头发和指甲样本中均发现女性 TPHP 水平高于男性且具有统计学意义。越南[34]以尿液样本为基质的研究显示女性尿液中 BDCIPP 和 DPHP 的平均水平(分别为 4.64 和 8.56 ng/mL)略高于男性(分别为 0.92 和 7.37 ng/mL)。在儿童青少年人群中也观察到类似的性别差异。中国东部[30]的一项研究显示,12~15 岁青少年女孩尿液中的 TCEP、磷酸二(1-氯-2-丙基)酯(Bis(1-chloro-2-propyl)phosphate, BCIPP)和 OH-DPHP 水平均显著高于男孩($P < 0.05$),并且女孩尿液中 OH-DPHP 的中位数浓度是男孩的 3 倍($P < 0.001$)。

OPEs 暴露的性别差异可能源于多种因素。首先,部分 OPEs 已被应用于个人护理品和化妆品中(例如指甲油),女性通常比男性的使用频率更高[33]。其次,女性可能更长期暴露于室内微环境中,有研究表明室内灰尘中的 OPEs 含量与尿液中其代谢物的浓度有关,频繁家务劳动(如拖地)可能导致暴露水平增加[35]。然而,也有个别研究结果不一致。例如,Wang 等[36]在中国东部的研究发现女性尿液中磷酸二(2-丁氧基乙基)酯(Bis(2-butoxyethyl)phosphate, BBOEP)和 BDCIPP 的含量水平均低于男性。这可能是因为各研究中存在着不同的混杂因素,这同时说明 OPEs 的性别分布差异尚无定论,仍需大规模的队列研究进行深入探讨。

3.3.3. 职业分布

从事涉及 OPEs 生产使用或处理的职业人群,其体内的 OPEs 水平普遍高于普通人群,不同职业类型间的 OPEs 暴露水平及分布也存在显著差异。目前已经发现的 OPEs 暴露的常见职业包括消防员、建筑材料生产与施工、美甲沙龙、电子垃圾回收和汽车拆解等。消防员与一般人群尿中多种 OPEs 代谢物的暴露情况为 BCEP (中位数: 0.84 vs. <0.1 ng/mL)、BCIPP (0.24 vs. 0.11 ng/mL)、BDCIPP (3.3 vs. 0.30 ng/mL)和 DPHP (4.0 vs. 0.26 ng/mL) [37]。电子垃圾拆解工人[38]尿液中 Σ OPEs 的中位浓度(21.9 ng/mL, 范围: 4.24~164 ng/mL)和汽车拆解工人尿中位浓度(15.1 ng/mL, 范围: 4.00~89.2 ng/mL)也显著高于参考样本(2.49 ng/mL, 范围: 0.79~16.5 ng/mL)。同时,研究已表明从事相关职业活动会显著增加瞬时 OPEs 的暴露水平从而风险增加。美甲师工作前后尿液中的 Σ OPEs 浓度范围分别为 1.54~18.6 ng/mL 和 4.48~19.9 ng/mL,工作后尿液中 TPHP 和 DPHP 浓度是工作前的 5.2 倍和 1.8 倍[39]。同样,从事使用含有有机磷阻燃剂材料的建筑工人[40]在工作后尿液样本中的 OPEs 浓度明显上升。以上证据均提示从事与 OPEs 密切接触的职业人群存在更高的内暴露水平。未来应针对高风险职业群体开展系统性监测与健康风险评估,为职业防护与暴露控制策略提供科学依据。

4. 不同生物基质的应用

目前的研究已采用了多种人体生物基质对 OPEs 进行检测,包括血液、尿液、母乳、胎盘、头发和指甲等[12]。此外,还有少量研究使用了脑脊液、痰液样本[33] [41]。尿液因其无创易采集的优点是最常使

用测定 OPEs 代谢物的基质, 并且可反映多个来源和人体暴露途径的贡献, 但 OPEs 在体内半衰期较短[42], 其结果难以反映长期暴露水平。血液也是目前的重点检测基质, 可反映人体内部较长时期接触污染物的情况。由于人类对代谢的敏感性不同, 关于人体血液中 OPEs 的研究远少于对持久性有机污染物的研究。其它生物基质如头发和指甲以非侵入性方式取样, 样本易于收集、运输和储存且可以反映体内的累积暴露。但由于样本前处理过程相对复杂且采样部位对结果一致性影响较大, 此类研究相对较少。

OPEs 水平和组成模式在采样基质(如血液、头发、指甲和胎盘)之间存在差异, 即使同样是累积样本(如指甲和头发)的检测结果也不尽相同。本质上是因为人体不同部位代谢周期的变化导致 OPEs 的处理方式存在显著差异。因此不同基质间的结果对比存在显著复杂性。此外, 个体代谢酶多态性会导致即使相同暴露水平下不同个体在同一生物基质中的浓度差异也很大。上述均表明, 单一基质的检测结果给暴露水平对比带来挑战且难以全面反映个体的真实暴露水平, 需要结合多种基质数据进行同时检测, 交叉验证。

5. 人群相关健康风险

近年来, 越来越多在人群中开展的流行病学研究探讨了 OPEs 对人群健康的潜在影响。现有证据表明, OPEs 暴露与多种健康损害相关, 包括内分泌干扰作用[43]、肝肾毒性[44][45]、肺功能下降[46]、甲状腺疾病[47]、生殖发育毒性[48]等。例如, Luo 等人[43]在青少年中的研究发现尿液中某些 OPEs 代谢物与糖尿病前期和葡萄糖稳态指数相关, 且该关联具有性别依赖性, 在女性中, BDCIPP 暴露量每增加一个单位, 糖尿病前期风险增加 2.51 倍。中国武汉一项基于 1981 名成年人的研究[49]也发现 BDCIPP 与高脂血症存在正相关关系, 且 BDCIPP、DPHP、以及 4-OH-DPHP 等多种代谢物与高密度脂蛋白胆固醇呈负相关。这些证据提示 OPEs 可能对人体有内分泌干扰效应并且会导致人体代谢紊乱。此外还有流行病学研究显示体内 OPEs 水平与抑郁症存在关联, BCEP 暴露每增加一个单位, 抑郁症风险增加 1.2 倍[50]。然而, 现有的研究结论仍存在一定不一致性, 例如一项基于 NHANES 尿液数据的研究显示 DPHP 与肝功能指标碱性磷酸酶呈正相关[44], 但是这种关联在广州人群[51]的研究中未得到重复验证。总体而言, 现有流行病学证据支持 OPEs 对人群健康的潜在危害, 但其因果关系及剂量-反应模式尚不明确。未来需结合大样本前瞻性队列研究与实验毒理学研究, 以阐明 OPEs 暴露的健康效应机制, 为风险评估与公共卫生干预提供更加可靠的科学依据。

6. 总结与展望

人体内 OPEs 的暴露水平已不容忽视, 并且受到地区因素、人群因素(性别、年龄、职业)等多种因素的影响。现有流行病学证据已表明 OPEs 暴露与多种健康风险相关。然而, 目前人体 OPEs 的内暴露研究仍然存在短板: (1) 依赖单一生物基质; (2) 缺少大样本队列研究。因此未来研究应加强对多种生物基质(如血液、毛发、指甲等)的综合监测, 并结合多区域、多人群的大样本系统调查进行动态监测, 建立内暴露数据库以揭示更全面稳定的 OPEs 暴露谱。此外, 还应进一步展开 OPEs 暴露的溯源研究, 利用多种模型(如源解析模型)定量识别人体暴露途径的贡献占比, 以及明确区域 OPEs 污染的核心来源(如特定产业产生、特定高风险产品等)。这将有利于得到更加具体的 OPEs 暴露特征, 为展开有针对性的人群监测和制定防治策略打下坚实的基础, 实现从“广谱防控”到“精准干预”的转变。

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