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全氟与多氟烷基化合物的生态风险现状与分析技术研究进展

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摘要: 全氟与多氟烷基物质 (PFASs) 广泛存在于土壤、水体和大气中，其持久性、迁移性和生物累积性导致其治理难度较大，其中地下水污染问题尤为严重，对生态环境安全和人类健康构成威胁。本文从 PFASs 的污染来源、迁移转化途径、污染现状及分析检测技术等方面，对环境介质中 PFASs 的研究现状进行总结。全氟辛酸 (PFOA) 和全氟磺酸 (PFOS) 是当前全球重点管控的 PFASs 物质。美国饮用水标准将 PFOA 和 PFOS 的限值均设定为 4ng/L，而中国《生活饮用水卫生标准》对两者的限值分别为 80ng/L 和 40ng/L。目前 PFASs 在地下水中的浓度范围从 ng/L 到 μg/L 不等，其中短链 PFASs 因较高的水溶性和迁移性，检出频率显著更高。在检测技术方面，气相色谱-质谱联用 (GC-MS) 和液相色谱-质谱联用 (LC-MS) 等高灵敏度、高分辨率的质谱分析方法仍是 PFASs 检测的主流手段。然而，上述方法依赖复杂的样品前处理和实验室条件，难以满足实时监测和应急响应的需求。随着监管力度的不断增强，现有技术面临着超低浓度检测灵敏度不足、复杂基质干扰以及缺乏统一分析标准等挑战。未来研究应重点关注新型 PFASs 的识别与毒性评估，以及检测技术的优化与创新；同时，结合人工智能技术优化数据处理和非靶向筛查的方法，提高检测效率和准确性。

关键词: 全氟与多氟烷基化合物；生态风险；污染水平；地下水；分析检测技术

要点:

(1) 地下水 PFASs 含量及种类易受土壤、地表水和大气等多种环境介质的影响。

(2) LC-MS 与 GC-MS 作为 PFASs 主流检测方法，在实时监测等方面仍存在技术局限。

(3) 机器学习等新方法的引入为新型 PFASs 的分析提供了潜在解决方案。

中图分类号: O656　　**文献标识码:** A

全氟与多氟烷基物质 (Per- and Polyfluoroalkyl Substances, PFASs) 是一类人工合成的有机氟化合物，其特点是至少含有一个完全氟化的甲基或亚甲基碳原子 (不含任何 H、Cl、Br、I 原子)，同时因其结构中大量 C—F 键的存在而具有极高的热稳定性和难降解性[1]。PFASs 常用于工业制造、消费品、防护材料等领域，例如纺织品防水剂、食品包装材料和消防泡沫等[2-5]。然而，PFASs 的化学惰性和高度稳定性也导致其在环境中难以降解，被称为“永久化学品”[6]，对生态环境和人类健康构成潜在威胁。

近年来研究发现，PFASs 已在大气[7]、水体[8]、土壤[9]与沉积物[10]中广泛存在。其中，地下水作为重要的饮用水源，其面临的 PFASs 污染风险尤为突出[11]。PFASs 通过地表水渗透和土壤淋溶等[12]进入地下水系统。由于 PFASs 具有极高的化学稳定性和难降解性，一旦进入地下水，将在地下水中长期存在。在受到 PFASs 污染的场所，即使生产或处置活动已停止，其仍可持续地成为地下水

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污染的重要源头, 并能长时间、远距离地迁移至周围地下水系统[13]。目前由于常规污水处理工艺对 PFASs 的去除效率较低, 某些情况下甚至可能呈现为负值。例如, Liu 等[14]发现在采用厌氧-缺氧-好氧 (A^2/O) 工艺的某污水处理厂中, 全氟丁酸 (PFBA) 与全氟辛酸 (PFOA) 等传统 PFASs 的平均去除率为−5.5%±15%, 氢化全氟羧酸 (H-PFCAs)、全氟烷基磺酰胺 (FASAs) 与氟调聚羧酸类 (FTCA) 等新型 PFASs 平均去除率为−62%±12%。去除率出现负值可能与污水处理过程中 PFASs 的代谢转化有关[15], 这意味着 PFASs 能通过水系统对人类健康和生态环境构成持续而潜在的长期威胁。

当前, PFASs 的分析方法主要包括液相色谱-质谱联用法 (LC-MS) 与气相色谱-质谱联用法 (GC-MS) 等[16]。尽管这些技术在灵敏度和选择性方面取得了显著进展, 但仍存在一些局限性。上述基于质谱的分析检测方法通常需要较为复杂的样品前处理和实验室条件, 无法实现现场快速检测, 难以满足实时监测和应急响应的需求[17]。近年来, 一些新兴方法, 如电化学传感技术[18], 因其快速、便携和低成本的特点, 逐渐成为 PFASs 快速分析检测研究的热点。同时, 机器学习等人工智能技术的引入为日益增长的 PFASs 种类的非靶向筛查和数据分析提供了新的可能性[19]。本文系统评述了 PFASs 在不同环境介质中的污染现状, 重点关注其在地下水中的迁移富集机制和影响因素; 同时, 深入探讨了目前 PFASs 的分析检测技术研究进展, 评估其在实际应用中的适用性和局限性, 提出了未来应重点关注的研究方向, 包括: 开发更高灵敏度和更高选择性的检测技术; 加强对新型 PFASs 化合物的研究和检测方法的开发; 建立统一的分析标准和方法; 开发基于人工智能的 PFASs 数据分析平台。

1　环境中 PFASs 的来源与迁移转化特征

氟化工业是环境中 PFASs 的主要来源之一[20], 在 PFASs 的生产过程中, 企业会产生大量的废水和固体废物, 这些废弃物中可能含有未反应的 PFASs 及其副产物。此外, PFASs 用于 200 多种消费品, 包括纺织品、消防泡沫, 甚至攀岩绳和吉他弦等物品[21], 在使用过程中, 这些产品会释放 PFASs, 导致其进入环境。

污水处理厂的排放也是 PFASs 进入环境的另一重要途径[15]。由于传统污水处理工艺对 PFASs 的去除效率较低, 许多 PFASs 在处理后仍然存在于出水和污泥中。根据研究[22], 在美国密歇根州的 10 家污水处理厂中, 出水 PFASs 浓度显著高于进水浓度, 最高达进水浓度的 19 倍。这是由于进水中的前体物质在处理过程中转化为 PFASs 所致, 加剧了水体污染。污水处理厂中的 PFASs 主要来自含 PFASs 产品的直接排放 (如工业制造[23]和纺织品[24]等消费品) 以及污水中 PFASs 前体物质的原位转化。在好氧处理过程中, 微生物和酶可将全氟烷基醇 (FTOH) 等前体转化为全氟羧酸 (PFCA)[25], N-乙基全氟辛磺酰胺 (N-EtFOSA) 和 N-乙基全氟辛基磺酰胺乙醇 (N-EtFOSE) 等前体也可转化为全氟辛烷磺酸 (PFOS)[26]。这些 PFASs 前体的转化是污水处理厂中 PFASs 种类和浓度增加的重要原因。

PFASs 在环境中的迁移途径主要包括大气干湿沉降、水体迁移、土壤淋洗和地下水下渗等[1]。PFASs 通过地表水体的流动进入河流、湖泊和海洋等水域。由于 PFASs 不同分子结构特征, 其在水中的分布行为存在差异[27]。这种分布不均的现象主要源于 PFASs 分子结构中疏水和亲水部分与水相、颗粒物及沉积物等的相互作用, 使得不同类型的 PFASs 在水中表现出不同的行为。长链 PFASs 倾向于与有机物结合, 而短链 PFASs 则更易在水中分散。水体中 PFASs 的浓度和分布还受温度、盐度和化学组成等多重环境因素调控。研究发现[28], PFASs 在土壤-水界面处的滞留和迁移行为受到土壤类型、pH 值和有机质含量等因素的影响。

目前 PFASs 污染已在水体、土壤和大气中被广泛检出。因此, 有效地控制和治理不同环境介质中的 PFASs 污染至关重要, 其迁移转化途径如图 1 所示。

2　环境中 PFASs 的生态风险与污染水平

2.1　PFASs 的生态环境风险

PFASs 的生态环境风险主要体现在对生态系统和人类健康的潜在威胁。在生态环境方面, PFASs 的存在会干扰生态系统的物质循环过程, 改变微生物群落结构, 降低生物多样性。研究表明[29-30], PFASs 对水生生物存在毒性影响, 可能导致鱼类和其他水生生物的生长和繁殖受到抑制。此外, PFASs 的生物累积性使其在食物链中逐渐富集, 最终可能对顶级捕食者 (包括人类) 造成更大的风险[31]。在人类健康方面, PFASs 的潜在危害已引起全球范围的广泛关注。以美国为例, 美国环境保护署 (US EPA) 在 2016 年就对 PFOS 和 PFOA 制定了 70ng/L 的饮用水终生健康建议浓度限值[32];

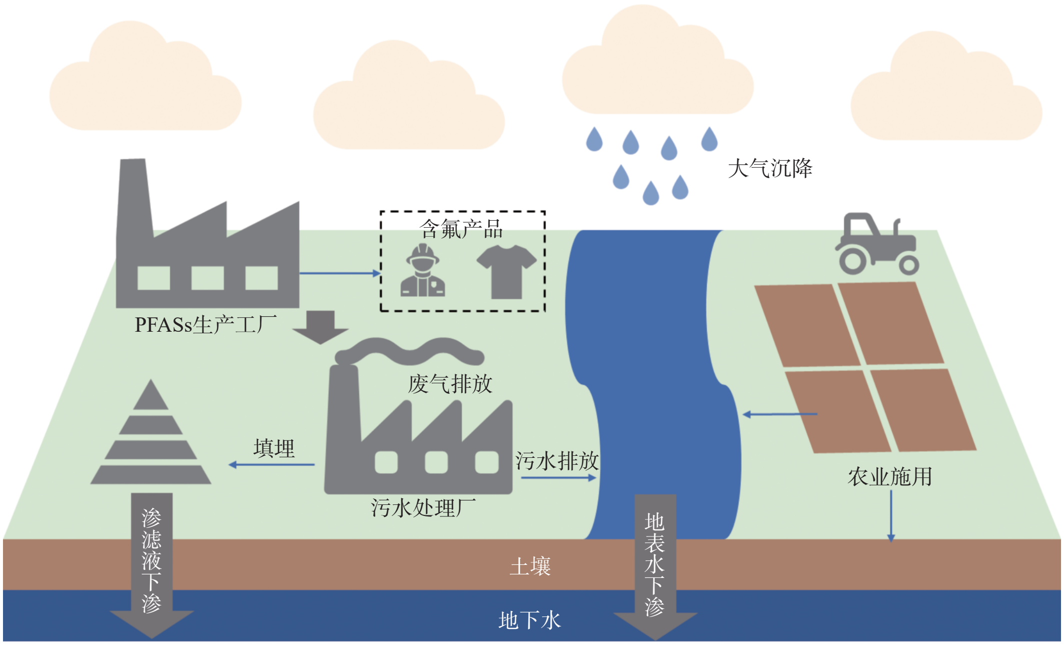

图1　PFASs 在不同环境介质的迁移示意图

Fig. 1　Schematic diagram of PFASs migration in different environmental media.

2022 年 US EPA 将 PFOA 限值降至 0.004ng/L, PFOS 限值调整为 0.02ng/L; 2024 年 4 月美国颁布了首个具有法律效力的国家饮用水标准《National Primary Drinking Water Regulations》, 对饮用水中的 6 种 PFASs 物质建立了最大污染物水平 (MCL), 其中 PFOA 与 PFOS 的 MCL 值均为 4ng/L。其他国家和地区也发布了针对饮用水中 PFASs 的限制标准。例如, 中国《生活饮用水卫生标准》(GB 5749—2022) 规定 PFOA 和 PFOS 的限值分别为 80ng/L 和 40ng/L; 欧盟饮用水水质指令 [DIRECTIVE (EU) 2020/2184] 则规定总 PFASs 含量在饮用水中的限值为 100ng/L。

随着 PFASs 的使用和排放问题日益突出, 公众对其潜在危害的关注也在不断增加。 近年来, 越来越多的研究聚焦于 PFASs 对人类健康的影响。研究已证实 PFASs 可经胎盘转移至胎儿体内, 对胎儿构成健康风险[33]。更令人担忧的是, 高浓度 PFASs 暴露与多种健康问题如影响生殖系统[34]、肾功能下降[35]、心血管疾病[36]和影响血脂稳态[37]等存在潜在关联。

2.2　环境中 PFASs 的污染现状

2.2.1　水环境中 PFASs 的污染水平

PFASs 作为一类持久性有机污染物, 广泛存在于各种水环境中, 包括地表水[38]、地下水[39]和饮用水[40]。PFASs 在水环境中的污染具有显著的区域差异性。靠近工业排放源和军事基地的水体通常显示出更高的污染浓度。例如, 美国北卡罗莱纳州一家 PFASs 生产工厂周边地区的地下水总 PFASs 浓度平均值高达 1863ng/L, 附近河流的总 PFASs 平均浓度为 1717ng/L[41]。欧洲多国的研究[42-44]同样显示水环境中总 PFASs 普遍超过欧盟设立的饮用水限值 (100ng/L), 且即使在北极等[45]偏远地区也能检测到这类物质, 这意味着 PFASs 具有显著的长距离传输能力。

由于地下水污染的隐蔽性和长期性, 使得地下水中 PFASs 的治理难度更大。PFASs 的持久性与难降解性, 使其能够长期污染地下水, 对饮用水安全和生态环境造成严重威胁。根据 EPA 数据显示, 在美国来自 4920 个饮用水供应系统的近 37000 个 PFAS 样本中, 72% 的 PFASs 检出发生在地下水中, 且检测的地下水的平均总 PFASs 浓度 (210ng/L) 均高于地表水 (90ng/L)[46]。此外, 地下水中短链 PFASs 的检出率高于长链 PFASs, 这可能与其较低的辛醇-水分配系数和较高的水溶性有关[12]。一项针对中国华北地区农业区水源调查也支持了该结论[47]: 随着 PFASs 碳链长度的增加, 其在地下水中的平均浓度呈下降趋势, 该研究主要在地下水中检出短链全氟丁烷磺酸盐 (PFBS) 和 PFOA 的主要同系物。地下水环境中 PFASs 浓度与国家 (地区) 发达程度和地下水类型显著相关, 不同国家和地区地下水环境的典型 PFASs 浓度及其种类具有较大差异 (表 1)。

表 1　部分国家和地区地下水环境中典型 PFASs 浓度及其种类

Table 1　Concentrations and types of PFASs in groundwater environments of some countries and regions.

地下水样品来源	地下水环境类型	典型 PFASs 化合物及其浓度 (ng/L)					ΣPFASs (ng/L)	参考文献
		PFBA	PFPeA	PFOA	PFBS	PFOS		
中国广州	垃圾填埋场地下水	n.d ~ 119	n.d ~ 16.4	n.d ~ 27.4	n.d ~ 544	n.d ~ 9.42	3.61 ~ 784	[48]
中国北京	再生水灌区地下水	2.94	0.94	2.88	1.15	＜0.1	1.07 ~ 24.19	[39]
中国青岛	饮用水源地下水	5.26	0.35	3.32	4.44	4.84	19.5	[49]
美国北卡罗来纳州	氟化工厂附近地下水	7.3 ~ 35.9	6.2 ~ 23.5	5.3 ~ 7.5	5.1 ~ 15.1	10.7 ~ 143.4	20.3 ~ 4773	[41]
美国东部地区	饮用水源地下水	n.d ~ 42	n.d ~ 20	n.d ~ 1500	n.d ~ 24	n.d ~ 98	n.d ~ 1645.4	[50]
瑞典	消防场地地下水	n.d	n.d	＜5 ~ 390	＜1 ~ 390	5.3 ~ 16000	16 ~ 22000	[51]
韩国	城市地下水	n.d	0.84 ~ 6.16	3.06 ~ 6.72	1.04 ~ 3.18	n.d	n.d ~ 36.9	[52]
法国	城市地下水	n.d ~ 63	n.d ~ 213	0.06 ~ 14	0.04 ~ 38	0.07 ~ 80	0.56 ~ 638	[53]
捷克共和国	城市地下水	0.277 ~ 7.03	0.021 ~ 7.77	0.021 ~ 4.16	0.021 ~ 1.06	0.021 ~ 8.09	n.d ~ 23.9	[54]
巴西南里奥格兰德州	饮用水源地下水	n.d ~ 56.1	n.d ~ 165	n.d ~ 249	n.d ~ 5.1	n.d ~ 15.8	718	[55]

注:“n.d”表示该数据未检出。一些国家的地下水样品在文献中未提供具体地区信息。

尽管目前许多国家已开始逐步淘汰 PFOA 和 PFOS 等 PFASs 并对其出台相关限制值, 但由于其持久性, 上述物质仍然普遍存在于水环境中[56]。目前, 国际社会对 PFASs 的监管仍处于发展阶段, 现有监测数据存在较大不确定性, 亟需建立更为全面和统一的监测标准。

2.2.2　土壤和沉积物中 PFASs 的污染水平

土壤和沉积物是 PFASs 环境行为研究的关键介质, 它们不仅是全球重要的 PFASs 储库[57], 更是 PFASs 向地下水迁移的关键传输介质[9]。氟化工业区周边土壤和沉积物中 PFASs 浓度通常较高, 四川成都市某工业园区周边表层土壤中检出 12 种 PFASs, 总浓度平均值为 104.84ng/g (干重)。其中, 全氟庚酸 (PFHpA, 72.99ng/g, 占比 70%) 和 PFOA (29.25ng/g, 占比 28%) 为主要污染物。此外, 消防活动是土壤 PFASs 污染的重要来源之一。Dauchy 等[58] 发现在消防训练场地中 PFOS、6∶2 含氟调聚物磺酸 (6∶2 FTSA) 和 6∶2 含氟调聚物磺酰胺烷基甜菜碱 (6∶2 FTAB) 是表层土壤中最主要的 PFASs, 最高浓度可达 357000μg/g。尽管存在黏土层, PFASs 仍可渗透至地表以下 15m。该训练场的地下水 PFASs 浓度高达 8300ng/L, 6∶2 FTAB 作为短链氟化物质甚至可深入至地表以下 20m。

不同土壤和沉积物的理化特性 (如: 有机质含量、黏土矿物组成、pH 值和离子类型及浓度等) 显著地影响 PFASs 的吸附、保留和迁移能力[59-61]。高有机质的土壤对 PFASs 具有更强的吸附能力, 这可能导致其在土壤中累积, 但同时也增加了 PFASs 在土壤中的长期滞留, 并可能继续缓慢释放到地下水中[62]。

2.2.3　大气环境中 PFASs 的污染水平

作为持久性有机污染物, PFASs 除了通过水体和土壤传播, 还可以通过大气远距离传播至偏远地区。自 2009 年起, 北极监测点的监测结果显示, 6∶2、8∶2 与 10∶2 FTOH 等是最常检出的氟化物, 浓度范围为＜0.4pg/m^3 至 9.7pg/m^3[63]。

Wu 等[64] 分析了中国东南沿海和山区城市 (包括福建省福州、厦门、漳州和南平) 的大气中 PFASs 特征, 发现大气中 PFASs 浓度变化范围为 7.8pg/m^3 至 290.0pg/m^3 不等, 其中南平市的 PFASs 浓度最为突出, 并指出全氟己酸 (PFHxA) 是该地区大气中最主要的 PFASs, 这很可能与当地工业设施的排放密切相关。Li 等[65] 针对河北省石家庄市的空气和颗粒物 PFASs 含量进行了分析, 结果显示, 空气中 PFASs 浓度 (颗粒和气相总和) 介于 31.7206pg/m^3 至 206pg/m^3 之间, 平均值为 77.9pg/m^3。值得注意的是, PFBA 和 PFOA 是当地大气中占比最高的 PFASs(占比近 60%)。研究推测, 这两种物质高浓度的原因可能源于当地生物医药、纺织和化学合成等行业的排放。此外, 在降水过程中, 研究观察到大气中 PFASs 浓度出现暂时性增加。通过分析 PFASs 在雨水和气相中的逸度比, 发现 PFASs 存在着从雨水扩散到气相的显著趋势。这一现象可以解释为什么即便在降水结束后, 气相中 PFASs 浓度仍在持续增加, 为理解大气环境中 PFASs 的动态变化提供了重要线索。

城市固废焚烧排放的含氟气体也值得关注。在天津市开展的一项研究表明[66], 生活垃圾焚烧与填埋场空气中的中性 PFASs 总浓度范围为 393 ~

19000pg/m^3。其中，FTOH 浓度最高，范围为 187～19000pg/m^3，占主导地位；8∶2 FTOH 尤其突出，贡献率达 50%～75%；其次为 6∶2 FTOH(11%～48%) 和 10∶2 FTOH(1.8%～26%)。与天津市区一般大气水平相比，这些区域 PFAA 前体物浓度明显更高 (中位数∑FTOHs：463pg/m^3 *vs.* 248pg/m^3)。这一现象表明生活垃圾焚烧与填埋厂可能是大气中 PFASs 的重要排放源之一。

综上所述，大气环境中的 PFASs 污染是一个复杂而重要的环境问题，未来的研究应集中于大气中 PFASs 的监测、来源解析、沉降机制以及对生态系统的潜在影响，以便更好地理解 PFASs 在环境中的行为和风险。这将为制定有效的污染控制策略提供科学依据，帮助保护生态环境和人类健康。

3　环境介质中 PFASs 分析检测技术

3.1　样品提取与浓缩方法

在 PFASs 检测分析中，不同介质中样品的特性存在显著差异。因此，样品制备、萃取和预浓缩已成为 PFASs 分析中不可或缺的关键步骤，其目的在于提高检测灵敏度，消除背景干扰，确保分析结果的准确性。目前，对于气体、固体和液体中 PFASs 的样品前处理方法主要分为索氏提取、固相萃取 (SPE) 和液液萃取 (LLE)[67] 等。对于不同环境介质中 PFASs 的分析，样品预处理方法列于表 2。

3.1.1　水样预处理方法

通常水样中的 PFASs 浓度较低，目前 SPE 方法较为成熟与广泛，SPE 能够更有效地将目标分析物与干扰物质分离，缩短萃取时间，并使用更少的有机溶剂[72]，因此被广泛应用于水样中 PFASs 的分析。SPE 通常使用聚合物基的萃取柱，如 Oasis-WAX、Oasis-HLB 和 Strata X-AW 等市售的萃取柱，均对 PFASs 有良好的富集和纯化效果[73]，能够有效地分离目标化合物与干扰物质。对于中性 PFASs，通常使用甲醇作为洗脱溶剂；而对于离子型 PFASs，如全氟羧酸 (PFCAs) 和全氟磺酸 (PFSAs)，则常用含氨水的甲醇进行洗脱。

影响 PFASs 回收率的影响因素很多。例如，Brumovsky 等[74] 评估了吸附剂类型、pH 值、盐度和洗脱液对 PFASs 回收率的影响。该研究使用了三种不同的 SPE 柱：Oasis-HLB、Strata™-X 和 Oasis-WAX，制备了三种不同盐度的基质，包括自来水、人工海水和两者的 1∶1 混合物。在 pH=8 且海水含量为 50%(苦咸水) 或 100%(海水) 的条件下，使用 Oasis-HLB 吸附剂和甲醇作为洗脱液时，PFASs 的回收率达到了最高，分别为 82.8% 和 78.3%。相比之下，在相同条件下 Strata™-X 吸附剂的回收率略低，分别为 75.6% 和 71.3%。而 Oasis-WAX 吸附剂的回收率则相对较低，但其在去除干扰有机物方面具有优势，因为在这些条件下，PFCAs 和 PFSAs 不易被洗脱。

3.1.2　固体样品预处理方法

固体样品 (如污泥、沉积物和土壤) 的预处理通常比水样更复杂，需经过干燥、筛分、均质、提取和浓缩等步骤。根据《土壤和沉积物 全氟辛基磺酸和全氟辛酸及其盐类的测定 同位素稀释/液相色谱-三重四极杆质谱法》(HJ 1334—2023)，称取样品后加入内标和提取溶液，振荡提取，离心收集上清液，过滤后通过弱阴离子交换 SPE 柱净化。此外，也有研究[75] 表明，羽毛样品进行 SPE 净化并未改善分析结果，甚至回收率和精密度可能降低。因此，需根据具体样品类型来选择合适的预处理方法。

3.1.3　气体样品预处理方法

气体样品中 PFASs 主要为挥发性、半挥发性、中性化合物 (如 FTOH、FASE、FASA)[76]。目前，

表 2　不同环境介质中 PFASs 分析样品预处理方法

Table 2　Sample pre-treatment methods for PFAS analysis in different environmental media.

样品类型	PFASs 来源	样品预处理方法	分析仪器	参考文献
气体	城市气体颗粒物	超声辅助溶剂萃取＋SPE	HPLC-QqQ; HPLC-QTOF-MS	[68]
	污水处理厂 (主动采样：进水口和曝气池；被动采样：进水口、曝气池、二沉池、高效沉淀池、深床滤池、臭氧接触池和出水口)	气相：索氏萃取 颗粒相：甲醇超声萃取	GC-QTOF-MS	[69]
液体	地下水	SPE	HPLC-MS/MS	[49]
	海水	SPE	TSQ LC/MS	[70]
	污水	玻璃纤维过滤＋串联 SPE	UPLC-Orbitrap MS	[69]
固体	土壤	冻干＋超声辅助溶剂萃取＋SPE	UPLC-MS	[58]
	污泥	冻干＋超声辅助溶剂萃取＋SPE	UPLC-HRMS	[71]

空气中 PFASs 的采样方法主要包括主动空气采样方法和被动采样方法。主动空气采样方法, 是通过抽气装置将空气经过滤膜, 有效地收集颗粒物中的 PFASs, 该方法可同时采集气相和颗粒相 PFASs, 具有较高的采样效率。被动采样方法由于其噪音小、成本低, 目前也被广泛应用于气体中 PFASs 的采集。此外, 挥发性 PFASs 易从常用吸附剂 (如聚氨酯泡沫) 中损失[77], 而玻璃纤维滤膜和石英纤维滤膜则可能不可逆地吸附气相中的 PFASs[78]。为提高采样和测试的可靠性, 可通过选择不同吸附剂等方式以应对不同种类的 PFASs。样品萃取时, 常用加压溶剂萃取 (PLE), 溶剂为乙酸乙酯和丙酮等, 其优点在于溶剂用量少、提取时间短, 可减少污染并提高前处理效率。对于小体积室内空气样品, 常采用 SPE, 洗脱溶剂的选择取决于 PFASs 的类型 (中性 PFASs 用甲醇[79], 离子型 PFASs 用含甲醇的铵盐溶液[80])。

目前, 针对不同环境介质中 PFASs 的分析, 尚无通用的提取和净化方法。随着 PFASs 种类不断增加, 需要持续地探索和优化提取技术, 提高其选择性和灵敏度。

3.2　分析检测技术

3.2.1　色谱-串联质谱法

气相/液相色谱-串联质谱法因其高灵敏度、选择性和定量能力, 可检测痕量 PFASs, 是分析 PFASs 的首选方法。该方法需要指定的内标物和标准品, 可同时分析传统和新型 PFASs, 具有广泛的适用性。例如, 中国《生活饮用水标准检验方法 第 8 部分: 有机物指标》(GB/T 5750.8—2023) 规定了超高效液相色谱串联质谱法 (UPLC-MS/MS) 为检测水样中 11 种 PFASs 的标准方法; 美国 EPA 用于饮用水中 PFASs 分析的方法定量了 14 种至 25 种 PFASs, 其中不到 50% 的目标被认为是基于链长和醚取代的新出现的 PFASs[81]。虽然这些靶向分析方法在短期内提供了重要信息, 但无法完全覆盖不断涌现的新型 PFASs。

尽管色谱-串联质谱技术具有在单次分析中检测多种 PFASs 的优势, 但在处理不同样品基质和进行样品预处理时难度较大。在整个取样和样品制备过程中, 必须严格遵循质量保证/质量控制 (QA/QC) 程序, 以尽可能地减少背景污染对测定结果的潜在影响, 防止 PFASs 检测结果出现假阳性或假阴性。例如, 选择合适的容器材料进行 PFASs 分析是一项重大挑战, 因为 PFASs 会吸附在容器上 (如玻璃容器), 导致分析浓度降低; 或从含氟聚合物材料容器 (如聚四氟乙烯, PTFE) 中浸出, 从而增加了样品中的污染水平[82-83]。

3.2.2　非靶向筛查法

高分辨质谱 (HRMS) 技术的进步及其与液相色谱和离子迁移谱等前端分离技术的结合, 实现了高分辨率、高精度和高灵敏度的测量[81]。这一能力在 PFASs 的分析中非常重要, 尤其是在缺乏标准物质的情况下, HRMS 能够鉴定环境中未知 PFASs。最近的研究是利用 HRMS 技术, 通过可疑物质筛查和非靶向筛查工作流程, 已鉴定出数千种 PFASs, 极大地拓展了 PFASs 分析的深度和广度[84]。Casey 等[85]探讨了气相色谱-高分辨率质谱法 (GC-HRMS) 在筛查和非靶向分析 PFASs 中的应用, 以建立的 141 种 PFASs 化合物的数据库为基础, 开发了针对 GC-HRMS 的工作流程, 并在实际环境样品中进行了验证。

3.2.3　新兴检测技术

除了传统的 LC-MS、HRMS 和 GC-MS 技术外, 一些新兴的检测技术也逐渐被应用于 PFASs 的分析, 以达到快速分析的目的[86]。例如, 电化学传感器[87]、免疫分析[88]、表面增强拉曼光谱 (SERS)[89] 和离子迁移谱 (IMS)[90] 等技术, 这些技术具有快速、简便、成本低的优点, 在现场快速监测和应急检测中具有很大的应用潜力。

电化学传感器具有实时监测和快速响应的特点, 可以实现对 PFASs 的原位检测。Khan 等[91]开发了一种低成本传感器, 可在 5min 内实现对 pg/L 水平 PFOS 的快速检测。该传感器利用银纳米颗粒 (AgNPs) 嵌入富氟 Ti_3C_2 基多层 MXene 材料中, 通过静电作用和 F-F 相互作用, 实现了对 PFOS 和其他长链 PFASs 化合物的特异性结合。结果表明, 该传感器在 50pg/L 至 1.6ng/L 范围内表现出良好的线性响应, 检测限 (LOD) 为 33pg/L, 定量限 (LOQ) 为 99pg/L。此外, 该传感器对干扰物无响应, 具有极高的灵敏度和选择性, 适用于水样中 PFASs 的低成本快速筛查。

然而, 这些新兴技术也存在一些局限性。例如, 其灵敏度和选择性可能不如传统色谱-质谱联用仪器; 此外, 电化学检测方法目前主要集中于传统 PFASs (如 PFOA 和 PFOS) 的检测, 而对于其他种类 PFASs 的研究仍较为有限[92]。因此, 未来的研究应致力于提高这些新兴技术的性能, 拓展其在实际环境样品中的应用范围, 以实现对更多种类 PFASs 的高灵敏度和高选择性检测。

3.3 分析技术的挑战

PFASs 的环境分析技术尽管取得了显著进展，但仍面临诸多相互关联的挑战，这些挑战共同制约着 PFASs 污染监测的准确性和全面性。

环境样品的复杂性是其分析检测的首要难题。水体、土壤和气体等样品基质的成分多样，常常含有大量干扰物质，这些物质与 PFASs 间的相互作用会显著影响检测的灵敏度和准确性。因此，开发高效、选择性强的样品前处理技术至关重要。加强 SPE、固相微萃取 (SPME) 以及新型吸附材料的研发，旨在提高 PFASs 的回收率并有效地降低基质干扰。

PFASs 的结构多样性进一步增加了分析的复杂性。众多同系物和结构相似的化合物给定性定量分析带来巨大挑战。许多新型 PFASs 的理化性质和环境行为尚未完全阐明，缺乏相应的标准物质和分析方法，严重限制了对 PFASs 污染的全面评估。HRMS 技术凭借其高分辨率和精确质量数测定能力，在鉴定未知 PFASs 和区分结构相似同系物方面发挥着关键作用。然而，HRMS 数据处理的复杂性仍需进一步改进，有待开发智能化更高的数据处理算法和软件，以提高数据分析的效率和准确性。

环境样品中 PFASs 的极低浓度与复杂多样性对检测技术提出了苛刻要求，尤其对于地下水环境而言。目前，机器学习等人工智能技术为解决这些分析挑战提供了创新路径。George 等[93]将机器学习成功地应用于地下水 PFASs 检测分析，结果表明即使在数据高度缺失 (超过 95%) 的情况下，机器学习模型仍能通过深入挖掘共污染物之间的潜在关联性，准确预测 PFASs 浓度。特别值得注意的是，该模型在地理区域随机交叉验证中表现卓越，显著提高了对未经检测地下水水源的 PFASs 浓度预测能力，为地下水环境污染的精准识别提供了新的技术范式。

机器学习在 PFASs 分析中的应用前景广阔，不仅可提高分析效率，还有助于发现新型 PFASs 化合物，深化对其环境行为和毒性机制的理解。然而，模型的有效性高度依赖于高质量的训练数据，因此需要收集大量具有代表性的实验数据。未来，将机器学习与高通量筛选、自动化分析等先进技术相结合，有望进一步推动 PFASs 分析技术的发展，为环境风险评估和污染控制提供更可靠的科学支撑。

4 总结与展望

PFASs 广泛存在于土壤、水体、大气中，由于不同环境介质间的迁移转化，土壤、大气和地表水中的 PFASs 通过多种途径不断地向地下水系统渗透和迁移，如渗滤和径流等过程。PFASs 的持久性、迁移性和生物累积性等特性使其治理难度极大，对地下水安全和人类健康构成重大威胁。地下水环境的复杂性和 PFASs 的多样性使得地下水 PFASs 污染的监测和修复面临挑战。

在 PFASs 检测技术方面，近年来取得了显著进展，HRMS 技术的应用极大地提高了检测的灵敏度、准确性和分辨率，能够有效地识别和监测各种 PFASs，尤其是非靶向筛查技术能够发现新型和未知的 PFASs，同时一些新兴检测技术的发展也为 PFASs 的原位检测与应急响应提供了潜在可用的方法。然而，当前技术仍存在一些不足，例如对于超低浓度 PFASs 的检测灵敏度仍需提高，对于新型 PFASs 的检测方法有待完善，缺乏统一的分析标准和方法也限制了数据的可比性和可靠性。

建议未来研究重点关注以下方向。

(1) 开发更高灵敏度、更高选择性的检测技术。亟待发展基于新型传感器的快速检测技术，以及提高 HRMS 技术的灵敏度和选择性，满足日益增长的超低浓度 PFASs 检测需求。

(2) 加强对新型 PFASs 化合物的研究和检测方法的开发。随着新型 PFASs 的不断出现，需要持续开发针对这些新型化合物的检测方法，并深入研究其环境行为和毒理学效应。

(3) 建立统一的分析标准和方法。制定统一的采样、前处理和分析标准，以确保数据的可比性和可靠性，为 PFASs 污染的生态风险评估和管理提供科学依据。

(4) 开发基于人工智能的 PFASs 数据分析平台。利用机器学习和深度学习算法，建立能够自动识别、分析和解释 HRMS 等高通量检测技术产生的海量数据的智能化平台，提高数据处理效率，降低人为误差，并辅助发现新型 PFASs 和预测其环境行为。

Research Progress on the Ecological Risk Status and Analytical Techniques of Per- and Polyfluoroalkyl Substances

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HIGHLIGHTS

(1) The concentration and categories of PFASs in groundwater are susceptible to influences from various environmental media such as soil, surface water, and the atmosphere.
(2) Despite being the predominant detection methods for PFASs, LC-MS and GC-MS still exhibit technical limitations in areas such as real-time monitoring.
(3) Emerging methods like machine learning offer promising solutions for analyzing novel PFASs.

ABSTRACT: This paper is a review of the current state of pollution and analytical techniques for per- and polyfluoroalkyl substances (PFASs) in various environmental media. PFASs are widely present in soil, water, and the atmosphere, with groundwater pollution being particularly severe. The persistence, mobility, and bioaccumulation of PFASs pose significant challenges for remediation efforts, threatening ecological safety and human health. The complexity of groundwater environments and the diversity of PFASs present challenges for monitoring and remediation. In terms of PFASs detection technologies, significant progress has been made in recent years; the application of high-resolution mass spectrometry (HRMS) has greatly enhanced the sensitivity, accuracy, and resolution of detections, especially with non-targeted screening techniques that can identify new and unknown PFASs. However, current technologies still have limitations, such as the need for improved sensitivity in detecting ultra-low concentrations of PFASs, and the lack of standardized analytical methods restricts the reliability and comparability of data. Future research is recommended to focus on enhancing the sensitivity of PFAS detection technologies, establishing unified analytical standards, and incorporating tools such as artificial intelligence to assist in analysis, in order to effectively address the environmental and health challenges posed by PFAS pollution. The BRIEF REPORT is available for this paper at http://www.ykcs.ac.cn/en/article/doi/10.15898/j.ykcs.202412080252.

KEY WORDS: per- and polyfluoroalkyl substances (PFASs); ecological risk; pollution level ; groundwater; analytical detection technologies

BRIEF REPORT

Per- and Polyfluoroalkyl Substances (PFASs), characterized by their exceptional chemical stability and environmental persistence, pose severe contamination risks across global ecosystems[1]. These synthetic compounds, featuring fully fluorinated carbon chains resistant to degradation, accumulate in air[7], water[8], soil[9], and sediments[10]. Groundwater systems are particularly vulnerable due to leaching and long-range transport dynamics[11-13]. Alarmingly, conventional wastewater treatments like anaerobic-anoxic-oxic (A^2/O) processes exhibit negative PFAS removal efficiencies (e.g., −62% for emerging H-PFCAs) due to precursor transformations, exacerbating contamination risks[14-15].

Current analytical methods for PFASs, while sensitive, face critical operational limitations that hinder real-world monitoring. Although LC-MS and GC-MS remains the gold standard for laboratory-based detection, its reliance on complex pretreatment and specialized facilities renders it impractical for on-site applications. Emerging technologies, such as electrochemical sensors[18] and AI-enhanced non-targeted HRMS screening[19], offer rapid and portable alternatives. However, most field-deployable tools focus narrowly on legacy PFASs (PFOA/PFOS) and lack the sensitivity of traditional methods.

The ecological risk status of PFASs and analytical advancements are systematically evaluated in this review, to guide future mitigation strategies. By synthesizing evidence on environmental migration mechanisms, groundwater exposure pathways, and limitations of current detection technologies, we identify critical research priorities: standardizing protocols for emerging PFASs, integrating AI-driven predictive models, and deploying real-time sensors to monitor contamination hotspots. These insights aim to accelerate the development of scalable solutions for safeguarding environmental security against persistent PFAS threats.

1. Environmental sources, transport, and ecological risks of PFASs

PFASs primarily enter the environment through fluorochemical manufacturing[20], consumer product emissions [21], and wastewater treatment systems. Research has revealed that PFAS concentration in treated wastewater can exceed influent levels by 19-fold as a consequence of precursor transformation, such as microbial conversion of FTOHs to PFCAs[25] and N-EtFOSA to PFOS[26] during aerobic processes[22]. These persistent contaminants migrate through atmospheric deposition, hydrodynamic processes, and soil-groundwater interactions. Their distribution patterns are significantly influenced by molecular structure: long-chain PFASs tend to bind with organic matter, while their short-chain counterparts exhibit enhanced aqueous mobility[27]. Soil characteristics, particularly organic content and pH, further modulate their migration potential, as evidenced by significant PFAS penetration (15–20m depth) observed at fire training facilities[58].

The ecological implications of PFAS contamination are severe, stemming from their bio-accumulative properties and toxicity. These compounds disrupt microbial communities[29], impair aquatic organism reproduction[30], and pose human health risks through placental transfer[33] and associations with renal and cardiovascular disorders[35-37]. Regulatory frameworks remain inconsistent globally, with varying threshold limits: the United States has established a 2024 MCL of 4ng/L for PFOA/PFOS[32], China maintains standards of 80/40ng/L for PFOA/PFOS (GB 5749—2022), and the European Union enforces a limit of 100ng/L for total PFASs [DIRECTIVE (EU) 2020/2184]. Despite phase-out initiatives, legacy PFAS persist due to their remarkable environmental stability[56], as demonstrated by extreme soil contamination levels near firefighting sites (357000μg/g PFOS)[58] and their detection in remote Arctic regions (FTOHs $\leqslant$9.7pg/m^3)[63].

Urban atmospheric studies reveal hotspot variability in Fujian Nanping (290pg/m^3, PFAA[64]) and Shijiazhuang (77.9pg/m^3, PFBA/PFOA 60%[65]) with municipal waste incineration identified as a critical emission source (187–19000pg/m^3 FTOHs in Tianjin landfills[66]).

Current challenges include the lack of standardized global monitoring protocols and incomplete understanding of precursor transformation risks. Addressing this multifaceted environmental challenge requires integrated mitigation strategies encompassing source control, advanced wastewater treatment technologies, and harmonized regulatory frameworks.

2. Analytical and detection techniques for PFASs in environmental media

Variations in sample properties across environmental matrices demand rigorous pre-concentration and extraction procedures to ensure accurate PFAS analysis. Established methodologies, including Soxhlet extraction, solid-phase extraction (SPE), and liquid-liquid extraction (LLE)[67], effectively address matrix interference challenges while enhancing analytical sensitivity for gaseous, solid, and aqueous samples.

2.1 Sample extraction and enrichment of PFASs in environmental media

Water sample analysis: The detection of PFASs in aqueous environments primarily relies on SPE due to its effectiveness in isolating target compounds from complex matrices. Commercial SPE cartridges, such as Oasis-

WAX, Oasis-HLB, and Strata X-AW, efficiently concentrate both neutral PFASs (using methanol elution) and ionic species like PFCAs/PFSAs (using methanol with ammonium additives)[73]. Research[74] has demonstrated that Oasis-HLB cartridges achieve superior recoveries under pH 8 conditions (82.8% in brackish water, 78.3% in seawater), surpassing the performance of Strata™-X (75.6%–71.3%) and Oasis-WAX (which shows lower recovery but better organic interference elimination). The significant impact of salinity and pH variations emphasizes the importance of optimizing protocols for specific matrices.

Solid sample analysis: The analysis of soil, sediment, and sludge samples requires thorough pretreatment steps, including drying, sieving, and homogenization before extraction. While China's HJ 1334—2023 standard recommends isotope dilution with anion-exchange SPE purification, research indicates that universal SPE applications may not be suitable for all matrices. For instance, feather sample analysis revealed diminished recovery and precision following SPE purification[75], highlighting the necessity of developing matrix-specific methodologies.

Gas sample analysis: Airborne PFASs, particularly FTOHs and FASEs, require specialized sampling approaches due to their volatility and phase partitioning characteristics. While active sampling with filters effectively captures particle-bound PFASs, passive samplers provide a cost-effective alternative. Adsorbent selection is crucial: polyurethane foam risks losing volatile PFASs[77], and glass fiber filters can irreversibly adsorb PFASs[78]. Pressurized liquid extraction (PLE) using ethyl acetate or acetone offers advantages in reduced solvent consumption and improved throughput. For small indoor air samples, SPE remains effective, utilizing methanol for neutral PFASs[79] and ammonium-methanol combinations for ionic species[80].

The diverse structural characteristics of PFASs preclude the development of universal extraction protocols. Future methodological innovations must focus on addressing current gaps in selectivity and sensitivity, particularly for emerging precursors and ultra-short-chain homologs.

2.2 Analytical and detection techniques

Liquid/gas chromatography coupled with tandem mass spectrometry (LC/GC-MS/MS) maintains its position as the gold standard for PFAS analysis, offering ultrahigh sensitivity for trace-level detection, superior selectivity, and precise quantification. Global regulatory frameworks have widely adopted these techniques, as exemplified by China's GB/T 5750.8—2023 standard, which targets 11 PFASs in drinking water using UPLC-MS/MS, and the US EPA Method 533/537.1, which quantifies 14–25 PFASs, including emerging congeners with varied chain lengths or ether substitutions comprising less than 50% of the total[81]. Despite their proven reliability, significant challenges remain: (1) limited detection capability for novel PFASs lacking reference standards, and (2) matrix-specific interferences requiring rigorous quality control measures (QA/QC) to prevent false positives and negatives. Notable analytical concerns include PFAS adsorption to glass containers, leading to concentration underestimation, and contaminant leaching from fluoropolymer-based materials (e.g., PTFE), resulting in elevated background levels [82-83].

High-resolution mass spectrometry (HRMS), combined with chromatographic or ion mobility separation, enables comprehensive identification of unknown PFASs through exact mass determination and fragmentation pattern analysis. Modern HRMS workflows incorporate suspect screening databases (such as the 141 PFASs compounds in GC-HRMS[85]) and advanced computational tools, successfully identifying thousands of previously unmonitored PFASs in environmental samples[84]. While this approach proves invaluable for detecting emerging contaminants, it faces limitations due to incomplete spectral libraries and requirements for high technical expertise.

Novel onsite monitoring tools, including immunoassays, electrochemical sensors, surface-enhanced Raman spectroscopy (SERS), and ion mobility spectrometry (IMS), provide cost-effective options for field deployment. Khan[91] developed an MXene-based electrochemical sensor achieving PFOS detection at 33pg/L (LOD) within 5min through AgNPs-fluorinated MXene interactions. However, these emerging methods currently show lower sensitivity compared to LC-MS/MS (μg/L *versus* ng/L) and limited scope, primarily focusing on legacy compounds like PFOA/PFOS[92]. While SERS and IMS demonstrate potential for multiplexed detection, they require further optimization for complex environmental matrices.

3. Concluding remarks

PFASs exhibit remarkable environmental persistence and mobility, alongside documented ecological toxicity, which collectively contribute to their global environmental concerns. These characteristics highlight the need for interdisciplinary approaches to address the issue. Although detection technologies have advanced significantly, challenges remain. Current laboratory-based methods such as LC/GC-MS/MS are often incompatible with real-time field monitoring, and standardized analytical protocols struggle to detect emerging congeners. These limitations are further compounded by regulatory inconsistencies and the inefficiency of conventional treatment methods, which may inadvertently exacerbate contamination through precursor transformations. This review aims to highlight potential limitations in existing frameworks for addressing PFASs' complex contamination pathways and emphasizes the value of continued innovation in detection, risk assessment, and global governance strategies.

4. Future perspectives

In response to the mounting PFAS issue, future research can concentrate on the following four areas. (1) High-sensitivity detection technologies: develop field-deployable sensors and enhance HRMS workflows to detect ultratrace PFASs (≤pg/L) with focus on multiplexed detection of legacy and emerging congeners. (2) Emerging PFAS characterization: systematically investigate novel compounds' environmental behavior and toxicology while establishing validated detection methods. (3) Standardization of protocols: harmonize global standards for sampling, pretreatment, and analysis to ensure data reliability and comparability in ecological risk assessment. (4) AI-driven data analytics: build machine learning platforms to automate HRMS data interpretation, predict contamination patterns, and accelerate discovery of unknown PFASs. By integrating state-of-the-art analytical advancements, stringent standardization, and intelligent data systems, these measures will enable proactive PFASs management and protect ecosystems.

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