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工业过程二恶英的排放特征及其控制技术^{*}

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摘要 工业排放是环境中二恶英 (PCDD/Fs) 最主要的人为排放源。2010 年我国九部委联合发布 PCDD/Fs 污染防治指导意见, 之后出台多项政策要求对主要行业持久性有机污染物 (POPs) 开展污染防治。在一系列防治措施下, PCDD/Fs 的工业排放水平有所下降, 整体取得良好成效。本文针对固体废弃物焚烧、钢铁生产、有色金属生产和水泥窑协同处置四类主要行业的 PCDD/Fs 排放研究进展进行综述, 阐述了不同行业 PCDD/Fs 排放量、排放特征及其变化趋势, 比较分析了目前四类主要行业针对 PCDD/Fs 排放的控制技术及其效果, 并对烟气中 PCDD/Fs 污染控制技术的发展方向进行了展望。本文可为更加深入地了解工业排放 PCDD/Fs 的研究现状以及污染控制技术提供参考。

关键词 工业排放, 二恶英, 排放特征, 控制技术。

Dioxin emission characteristics and control technologies in industrial processes

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Abstract Industrial emissions are the main sources of anthropogenic polychlorinated dibenzo-p-dioxin and dibenzofurans (PCDD/Fs) in the environment. In 2010, nine ministries and commissions of China jointly issued the Guidance on the Strengthening of Dioxin Pollution Prevention, and then issued many policy documents for pollution prevention and control of persistent organic pollutants (POPs) in major industries. Under a series of control measures, the industrial emission of PCDD/Fs has been reduced in recent years. In this review, we summarized the research progress on PCDD/Fs emission in solid waste incineration, iron and steel smelting, non-ferrous metal smelting and coordinated disposal of cement kiln. The emission levels, emission characteristics and temporal

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trend of PCDD/Fs from these different industries were discussed, the control technologies and their effects to PCDD/Fs emission in four major industries were also evaluated. The development about PCDD/Fs control measures in flue gas was finally prospected. This review may enhance understanding the industrial emissions of PCDD/Fs and their control effects.

Keywords industrial emissions, dioxins, emission characteristics, control technology.

二恶英(dioxins)是一类具有相似结构和性质的氯代芳香烃族杂环化合物的统称,包括多氯代二苯并-对-二恶英 (polychlorinated dibenzo-p-dioxins, PCDDs) 和 多 氯代 二 苯 并 呋 喹 (polychlorinated dibenzofurans, PCDFs), 共有 210 个同族体. PCDD/Fs 作为典型的非故意产生的持久性有机污染物(UP-POPs), 其来源包括自然源和人为源两大类, 前者包括火山爆发、森林火灾等一些自然过程, 后者包括固体废弃物焚烧、钢铁生产、有色金属生产、含氯化学品生产和纸浆漂白等工业过程. 由于环境中的 PCDD/Fs 主要来源于人类活动, 自然排放的 PCDD/Fs 极少, 故针对 PCDD/Fs 的研究主要围绕人为源展开^[1-2].

研究显示, 2004 年我国 PCDD/Fs 大气排放量为 5042 g 毒性当量(TEQ), 之后一段时间未见官方统计数据, 但有研究指出 2016 年我国 PCDD/Fs 大气排放量为 10366 g TEQ^[3-4], 尽管不同研究中对排放因子和生产强度的选择差异较大, 导致 PCDD/Fs 大气排放量的计算存在一定差异^[5], 但金属生产、固体废弃物焚烧等工业污染源导致的 PCDD/Fs 排放量占大气总排放量的 90% 以上^[4]. 因此积极削减工业源排放的 PCDD/Fs 是其污染防治的关键, 也是我国履行《关于持久性有机污染物的斯德哥尔摩公约》, 推动国内经济高质量发展和生态文明建设的必然选择.

2010 年我国加强大气污染物防治, 工业废气治理投资快速增长, 并于 2014 年达到峰值^[6]. 但截止 2018 年, 我国除固体废弃物焚烧行业外的其他行业专门针对 UP-POPs 控制的措施十分有限, 且相关工作多停留在实验室研究阶段^[7], 与 2010 年的研究状况几近相似^[8]. 减少工业污染源 UP-POPs 的排放仍然是我国 POPs 污染控制面临的最大挑战^[7].

尽管目前对 PCDD/Fs 的工业排放源已有大量报道, 但对于不同工业的 PCDD/Fs 排放特征、污染控制措施及其成效评估的文献综述仍然相对较少, 且近年来随着工业的发展, 不同工业 PCDD/Fs 排放特征和排放量也发生了一定的变化, 因此, 有必要进一步对比以前和近年来 PCDD/Fs 排放特征、控制措施变化. 根据联合国环境规划署(UNEP)在 2013 年提出的《鉴别及量化 PCDD/Fs 类排放标准工具包》以及其他研究对不同行业 PCDD/Fs 的排放因子及排放量的核算结果^[9-12], 本文选取固体废弃物焚烧、钢铁生产、有色金属生产和水泥窑协同处置固体废弃物这四类排放因子较大、生产强度较高的行业为主要研究对象, 系统总结了固体废弃物焚烧、钢铁生产、有色金属生产和水泥窑协同处置四类重要工业源 PCDD/Fs 排放的相关研究进展, 阐述了不同行业 PCDD/Fs 排放特征及变化趋势, 比较分析了这四类重要行业针对 PCDD/Fs 排放采取的控制技术及其效果, 在此基础上对工业生产过程中 PCDD/Fs 污染控制技术的发展方向进行了展望. 本文可为更加深入了解工业排放 PCDD/Fs 的研究现状及其污染控制技术提供参考.

1 不同工业烟气中 PCDD/Fs 排放特征(Emission characteristics of PCDD/Fs in flue gas of different industries)

焚烧等工业热过程中的 PCDD/Fs 生成机理包括高温气相合成、低温异相催化前驱体反应和低温异相催化从头合成等. 异相反应被认为是热过程 PCDD/Fs 的主要生成机理, 可通过分析样品中 PCDFs/PCDDs 比值是否大于 1 来判断某排放源的 PCDD/Fs 生成途径是从头合成还是前驱体反应占主导地位^[13].

1.1 固体废弃物焚烧

固体废弃物焚烧主要指生活垃圾、危险废物、医疗废弃物等固体废弃物的焚烧^[3, 7]. 焚烧能减少 70%—80% 的质量以及 90% 的体积^[14], 且焚烧产生的热能不仅能有效杀灭病原体, 还可以用来发电^[15-16], 因此焚烧逐渐成为固体废弃物集中处置的首选方法^[17-18]. 2010—2020 年, 我国城市生活垃圾

焚烧处理量从 2317 万 t 增长到 14608 万 t(年增长率为 53.0%), 处理量和增长率均超过传统的填埋处理(9598 万 t 下降为 7772 万 t, 年增长率为-19.0%); 危险废物产生量从 1587 万 t 增长到 7282 万 t, 年增长率为 35.9%^[6, 19].

从 PCDD/Fs 指纹分布看, 大部分固体废弃物焚烧产生的烟气中 PCDD/Fs 以 7—8 氯代同族体为主, 少部分以 4—5 氯代同族体为主, 且 PCDFs/PCDDs 比值通常显著大于 1, 其生成机理主要为从头合成^[7, 20—22]. 从 PCDD/Fs 排放量, 2004 年我国固体废物焚烧大气 PCDD/Fs 排放量为 610 g TEQ(占大气 PCDD/Fs 排放量 12.1%), 2016 年为 2469 g TEQ(占大气 PCDD/Fs 排放量 23.8%)^[3—4], 同 2004 年相比, 2016 年我国固体废弃物焚烧 PCDD/Fs 排放量增加 1859 g TEQ(304.8%), 排放占比升高 11.7%. 在焚烧量相同的情况下, 焚烧医疗废弃物和危险废弃物产生的 PCDD/Fs 要远高于生活垃圾焚烧的排放量^[23].

固体废弃物焚烧厂因规模、工艺和操作控制等差异较大, PCDD/Fs 的排放水平有很大差别($0.5\text{--}3500 \mu\text{g}\cdot\text{t}^{-1}$ TEQ)^[11]. Ni 等^[24]在 2009 年的研究中指出, 我国生活垃圾焚烧过程中 PCDD/Fs 的平均排放因子为 $1728 \text{ ng}\cdot\text{t}^{-1}$ TEQ, 这与 2013 年 UNEP 提供的排放因子参考范围相一致^[11], 2018 年 Zhu 等^[25]的研究结果显示排放因子有所下降($27\text{--}225 \text{ ng}\cdot\text{t}^{-1}$ I-TEQ), 其均值为 $170 \text{ ng}\cdot\text{t}^{-1}$ I-TEQ, 这可能与后来的焚烧厂采取更加完善的控制措施有关. 若以 2020 年我国生活垃圾焚烧量 14608 万 t^[6] 和 Zhu 等的排放因子^[25]进行推测, 我国 2020 年生活垃圾焚烧 PCDD/Fs 排放量达 $3.9\text{--}36.7 \text{ g TEQ}$. 对于医疗废弃物, Cao 等^[26]2009 年的研究指出我国此类焚烧炉烟气中 PCDD/Fs 排放因子为 $0.78\text{--}474 \mu\text{g}\cdot\text{t}^{-1}$ I-TEQ, 据此估算的当年医疗废弃物焚烧产生的 PCDD/Fs 为 4.87 g TEQ ; 若以 2019 年我国医疗废弃物产量(226 万 t)^[27]进行推测, PCDD/Fs 年排放量可达 $1.76\text{--}1071 \text{ g I-TEQ}$.

总体相比于 2004 年, 2016 年我国固体废弃物焚烧行业大气 PCDD/Fs 总排放量增加 1859 g TEQ(304.8%), 排放占比升高 11.7%^[3—4]. 同时, 由于焚烧技术的推广, 新冠疫情后医疗废弃物的产量急剧增加(增幅可达 24.7%)^[28], 危险废弃物处置量于 2020 年首次超过产生量^[6, 19], 这可能直接导致固体废弃物焚烧 PCDD/Fs 排放量的增加, 然而相关研究报道比较欠缺, 相关工作有待进一步开展.

1.2 钢铁生产

钢铁生产流程可分为长流程和短流程两种, 其中长流程是指以铁矿石为原料, 以烧结、球团、炼焦、高炉炼铁、转炉炼钢和轧钢等工序为整套流程的生产工艺; 短流程则是以废钢和直接还原铁为原料, 直接从电炉炼钢开始的生产工艺^[29]. 我国长流程炼钢约占 90% 左右^[30], 但因电炉炼钢过程中废钢原料中的塑料和油漆等有机物对该过程 PCDD/Fs 的产生有重要影响^[31], 故本文中的钢铁生产主要是指长流程生产工艺和电炉炼钢.

炼焦、烧结、电弧炉炼钢等钢铁生产过程中生成的 PCDD/Fs 均以 7-8 氯代同族体为主, 且 PCDFs/PCDDs 比值大于 1, 其主要生成途径为从头合成^[11, 32—34]. 从排放量来看, 2004 年我国钢铁行业大气 PCDD/Fs 排放量为 1923 g TEQ, 而针对 2016 年的研究则估算为 5333 g TEQ, 同 2004 年相比 PCDD/Fs 排放量增加 177.3%^[3—4]. 我国钢铁生产行业大气 PCDD/Fs 排放的 90% 以上集中在 3 个环节: 铁矿石烧结(60% 以上)、电弧炉炼钢(20%—30%)和炼焦(5%—10%)^[35—36], 因此后续研究控制应重点关注这些主要过程.

汤铃等^[30]对我国 966 家钢铁企业(占我国粗钢产量 96.4%)进行研究表明, 2018 年我国钢铁行业烧结和电炉工序的 PCDD/Fs 排放因子分别为 $1583\text{--}1246 \text{ ng I-TEQ}\cdot\text{t}^{-1}$, 而炼焦等其它工序的排放因子小于 $300 \text{ ng I-TEQ}\cdot\text{t}^{-1}$; 关于烧结和电炉排放 PCDD/Fs 的研究结果与 Wang 等^[37—38]的结果基本一致($1330\text{--}7610 \text{ ng}\cdot\text{t}^{-1}$ I-TEQ 和 $177\text{--}869 \text{ ng}\cdot\text{t}^{-1}$ I-TEQ), 但远高于 2020 年杨艳艳等^[36]的研究结果($(180\pm220) \text{ ng}\cdot\text{t}^{-1}$ I-TEQ 和 $(270\pm230) \text{ ng}\cdot\text{t}^{-1}$ I-TEQ), 这可能与后者所涉及的研究企业数量较少、生产工艺和污染控制措施较为先进等因素有关. 根据汤铃^[30]等获得的排放因子和高炉炼铁物料平衡关系(每 t 生铁需要 1.6 t 铁矿石和 0.4 t 焦炭)^[39—41], 结合我国 2020 年生铁和粗钢产量(分别为 88898、106477 万 t)^[6], 2020 年我国烧结和电弧炉炼钢大气 PCDD/Fs 排放量分别为 2252 g I-TEQ 和 133 g I-TEQ(焦炭和转炉炼钢分别为 57 g I-TEQ、266 g I-TEQ), 明显高于固体废弃物焚烧的 PCDD/Fs 估算值.

总体来看, 相比于 2004 年, 2016 年我国钢铁行业大气 PCDD/Fs 排放量增加 3410 g TEQ(117.3%), 排放占比升高 13.3%^[3—4]; 同时结合现有数据对我国钢铁行业大气 PCDD/Fs 的排放进行计算, 结果表明

目前钢铁行业仍具有较高的大气 PCDD/Fs 排放水平,因此,针对该行业 PCDD/Fs 排放及其控制的研究仍需持续加强。

1.3 有色金属生产

有色金属生产包括有色金属生产和再生有色金属生产,其中再生有色金属生产因原料中含废弃导线、电子部件和废旧塑料等,为 PCDD/Fs 的产生提供了丰富的氯源,经物料中的铜、铁等金属的催化后可生成大量 PCDD/Fs(与有色金属生产相比可增加 1—3 个数量级)^[37, 42—43]。有色金属种类丰富,原料和生产工艺的不同对 PCDD/Fs 的排放特征和排放量有较大影响,但多数研究表明有色金属行业排放的 PCDD/Fs 主要源于铝、铜、铅生产过程^[42, 44]。

从指纹分布来看,铜生产过程中产生的 PCDD/Fs 多以 7—8 氯代同族体为主,且高氯代单体比例同原料中废铜含量成正比;而铝、铅、镁生产过程中多以 4—7 氯代同族体为主,主要生成途径为从头合成^[45—48]。从排放量来看,2004 年我国有色金属行业大气 PCDD/Fs 排放量为 563 g I-TEQ^[3];近年来关于有色金属行业 PCDD/Fs 排放的研究数据较少,文献报道 2013 年再生铝生产过程中 PCDD/Fs 排放量为 609 g I-TEQ^[42],高于 2004 年有色金属行业的总排放量,由此推测 2004 至 2013 年有色金属生产行业 PCDD/Fs 排放量可能呈现出一定的增加趋势。

聂志强^[49]对铜、镁冶炼以及废旧导线焚烧回收过程的研究表明,PCDD/Fs 排放因子范围为 38.5—5569 ng·t⁻¹ TEQ;这与 Yu 等^[38, 50]的研究结果基本一致(14.2—24451 ng·t⁻¹ I-TEQ),但远低于 Zou 等^[37]的研究结果(0.24—1.7 g·t⁻¹ I-TEQ,其中二次铅生产的排放因子为 4297 ng·t⁻¹ I-TEQ)。排放因子范围变化较大的原因可能与有色金属类型、生产原料、生产工艺和控制措施等有关。目前有色金属产量以精炼铜、电解铝以及十种有色金属总产量来核算,因此难以对有色金属行业排放的 PCDD/Fs 进行相对精细的计算,但 2004 至 2020 年我国十类有色金属总产量从 1430 万 t 增加到 6188 万 t^[6, 51],相关生产过程排放的 PCDD/Fs 总量可能出现相应增加。

1.4 水泥窑协同处置

因固体废弃物中含有水泥生产所需的部分原料,同时水泥窑的工作温度较高(1600 °C 以上)、物料停留时间长(30 min 以上),因此水泥窑常被开发用于固体废弃物的协同处置^[52—53]。但固体废弃物中的大量氯源和金属催化剂在高温过程中可能导致 PCDD/Fs 的产生^[3],因此水泥窑协同处置也是 PCDD/Fs 的排放源。水泥窑协同处置过程中废弃物的类型、添加量、处理工艺等均会影响 PCDD/Fs 的排放特征和排放量^[54]。

从指纹分布特征看,除少数样品中 PCDD/Fs 以 7—8 氯代同族体为主外,大部分水泥窑协同处置过程产生的 PCDD/Fs 以 4—6 氯代同族体为主^[53—56],主要生成途径为从头合成。从排放量看,2004 年我国水泥生产过程 PCDD/Fs 排放量为 365.3 g TEQ^[3]。张婧等^[57]研究指出,不同炉型的水泥窑 PCDD/Fs 排放因子差别可达 100 倍,而我国主要采用的水泥立窑生产工艺,PCDD/Fs 排放因子为 5.0 μg·t⁻¹ TEQ,远高于干法旋窑。Aykan^[58]对协同处置危险废物和医疗废弃物的水泥窑进行研究,结果表明烟气中 PCDD/Fs 排放量为每年 0.02 g。2018 年 Zou 等^[55]研究指出,我国水泥窑协同处置过程 PCDD/Fs 排放因子为 0.01—1.35 mg·t⁻¹ I-TEQ。尽管 2020 年我国水泥生产高达 339736 万 t^[6],但其中协同处置固体废弃物生产的水泥比例并不清晰,无法对该过程 PCDD/Fs 排放量进行计算^[59]。以水泥工业计划中提出的 2015 年建成 10% 的协同处置水泥厂的目标来推算^[59],水泥窑 PCDD/Fs 排放量将达到 2397 g I-TEQ,这与钢铁生产行业的排放量几乎相当。由于我国水泥窑协同处置固体废弃物的生产线投产较晚,相关研究的基础数据仍然较少,因此加强水泥窑协调处置固体废弃物过程中 PCDD/Fs 的排放监测研究十分必要,可为准确评估该行业 PCDD/Fs 排放量提供重要科学依据。

1.5 不同工业过程 PCDD/Fs 排放特征比较

基于以上排放特征分析,固体废弃物焚烧、钢铁生产和铜生产排放的 PCDD/Fs 多以 7—8 氯代同族体为主,而水泥窑协同处置和铅、铝等有色金属生产过程中多以 4—6/7 氯代同族体为主;尽管不同行业的 PCDD/Fs 指纹分布特征有所不同,但均以呋喃类为主要同族体,表明其来源主要为从头合成机理^[20, 34, 45, 54]。

从排放量分析(表1,图1),PCDD/Fs排放量依次为钢铁生产>固体废弃物焚烧>有色金属生产>水泥窑协同处置;依据现有文献数据进行估算,2020年PCDD/Fs排放量依次为钢铁生产>水泥窑协同处置>有色金属生产>固体废弃物焚烧,但水泥窑协同处置的排放量存在较大不确定性,仍需要更多的研究结果进行支撑。

表1 我国PCDD/Fs的主要排放源及其排放量

Table 1 Main emission sources of dioxins and their emissions in China

排放源 Emission source	排放因子/(ng·t ⁻¹ I-TEQ) Emission factor	年排放量/(g TEQ) Annual emission		参考文献 References
		大气 Atmosphere	总量 Total	
生活垃圾	—	125.8	338	
危险废物	—	57.27	243.27	[3]
医疗废物	—	427.4	1176.3	
总计(2004)	—	610.47	1757.57	
				[24]
	1728	—	—	[25]
生活垃圾	27—225	—	—	[23]
	12200	217	—	[60]
固体废弃物焚烧	56—607	—	—	[26]
危险废物	70—3270	—	—	[23]
工业废物	302500	103	—	[20]
	97800	272	—	[61]
医疗废物	780—473930	—	—	[4]
	1923.6	0.466	—	[30]
总计(2013)	—	1280	—	[37]
总计(2016)	—	2469	—	[38]
铁矿石烧结	—	1522.5	1523.4	
钢铁冶炼	—	150.9	1125.4	[3]
铸铁生产	—	10.7	97	
炼焦		239.2	252.6	[36]
总计(2004)	—	1923.31	2998.4	
	1582.95	—	—	[30]
铁矿石烧结	772.2—827.9	—	—	[37]
	1330—7610			[38]
	180±220			[36]
钢铁生产	1245.85	—	—	[30]
	270±23			[36]
电弧炉	3160	—	—	[37]
	177—869	—	—	[38]
炼焦	160.09	—	—	[30]
	28.9(WHO)	—	—	[62]
总计(2011)	—	6817	—	[63]
总计(2012)	—	618	—	[64]
总计(2015)	—	1216.83	—	[35]
总计(2016)	—	5333	—	[4]
总计(2018)	—	2240	—	[30]

续表1

排放源 Emission source	排放因子/(ng·t ⁻¹ I-TEQ) Emission factor	年排放量/(g TEQ) Annual emission		参考文献 References
		大气Atmosphere	总量Total	
铜生产	—	403	1133.8	
铝生产	—	133.5	365.5	
铅生产	—	13.4	17.4	[3]
其他	—	12.99	51.85	
总计(2004)	—	562.89	1568.55	
铜生产	38.5、651(WHO) 14.2	— —	— —	[49]
铝生产	1240.2	—	—	
有色金属生产	铅生产 锌生产	3140.0 166.0	— —	[50]
	再生铜	241719—1707200 14802 24451.3	— 37.5 —	[37] [65—66] [50]
	再生铝	147819—434840 84.8—2720	— —	[37] [38]
	再生铅	4297	—	[37]
	镁生产	412(WHO)	—	[49]
	废旧导线回收	5569(WHO)	—	[49]
水泥窑(2004)	—	365.3	365.3	[3]
水泥窑协同处置	水泥窑 水泥窑	5000 0.01—1.35 mg	0.02g —	[57—58] [55]
上述四类总计(2004)	—	3461.97	6437.22	[3]
所有污染源总计(2004)	—	5042.4	10236.8	[3]

注:“—”:表示未提及;其他:包锌、黄铜和青铜、镁等未提及的有色金属生产;

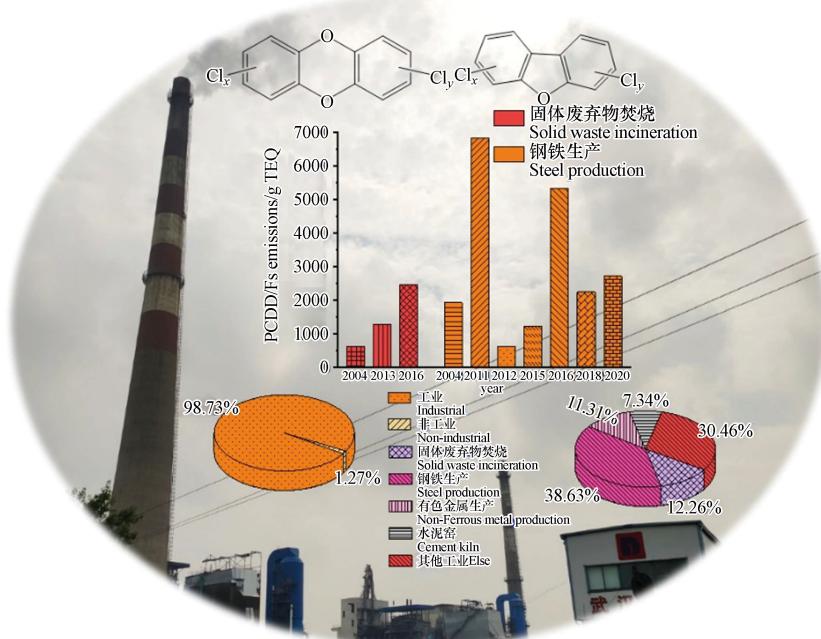


图1 四类工业大气PCDD/Fs排放量示意图^[3—4,30,35,61,63—65]

Fig.1 Atmosphere PCDD/Fs emission form four types of industrial^[3—4,30,35,61,63—65]

2 不同工业烟气 PCDD/Fs 污染控制技术 (Control technology of PCDD/Fs in different industrial processes)

相比于发达国家, 我国 PCDD/Fs 污染控制工作起步较晚^[67]。根据 PCDD/Fs 的生成机理及其来源, PCDD/Fs 的控制主要针对生成前、生成中和生成后三个过程开展相关工作^[44]。原料中的 PCDD/Fs 大多在高温下可直接分解, 因此高温再生成是 PCDD/Fs 排放量的主要来源, 故 PCDD/Fs 的控制减排主要通过控制运行的工作参数、添加抑制剂或增加末端空气污染控制装置(APCDs)等措施^[68](详见表 2), 本文主要针对工业烟气末端处理装置及其控制效果进行综述介绍。

表 2 PCDD/Fs 的全过程控制方法

Table 2 Whole process control method of PCDD/Fs

过程 Process	方法 Method	参考文献 References
生成前	物料预处理、添加辅助燃料、配制垃圾衍生燃料等	[69–70]
生成中	改进炉膛结构、调整工作参数(含氧量、气体湍流度、温度区间及停留时间、多段燃烧等)、含硫含氮抑制剂(硫酸、氨、含硫煤等)、烟气循环、烟气急冷等	[69, 71–74]
生成后	活性炭吸附、除尘器拦截(袋式、静电等除尘器等)、选择性催化还原、光降解、等离子体降解、高级氧化等	[72, 75–80]

由于不同行业烟气中 PCDD/Fs 的排放特征和浓度有所差别, 烟气温度、烟气量、烟气中粉尘和氮氧化物等常规污染物的种类和数量相差较大, 因此不同行业 APCDs 存在一定差异, 而 PCDD/Fs 多以协同净化为主^[81], 且不同行业排放控制标准不尽相同(见表 3), 因此本文针对不同行业的措施效果分别进行综述。

表 3 不同行业烟气 PCDD/Fs 排放控制标准

Table 3 PCDD/Fs emission control standard for different industries

行业 Industry	限值/(ng·m ⁻³ TEQ) Limiting value	开始时间 Time	参考文献 References
生活垃圾焚烧	0.1	2014	[82]
危险废物焚烧	0.5	2001	[83]
火葬场	0.5	2015	[84]
炼钢工业(电炉、烧结、球团)	0.5	2012	[85–86]
再生铜、铝、铅、锌	0.5	2015	[87]
水泥窑协同处置固体废物	0.1	2013	[88]

2.1 固体废弃物焚烧

固体废弃物焚烧作为 PCDD/Fs 主要的排放源, 相关控制技术比较完善^[7]。Wei 等^[89]研究发现, 经过垃圾发酵等预处理措施和焚烧参数控制后, 采用半干洗涤器(SDS)+干洗涤器(DS)+活性炭喷射(AC)+袋式除尘器(BF)+选择性催化还原(SCR)技术组成的 APCDs 对烟气中 PCDD/Fs 进行脱除, 最终的排放水平可达 $0.0028 \text{ ng} \cdot \text{m}^{-3}$ I-TEQ, 远低于 $0.1 \text{ ng} \cdot \text{m}^{-3}$ I-TEQ 的控制标准; 许多研究也表明, 通过良好的过程和末端控制, 固体废弃物焚烧厂烟气中 PCDD/Fs 的排放基本都能满足相关标准要求^[25, 90–91]。值得注意的是, 一些研究也报道了焚烧厂由于控制技术不达标或不稳定, 造成存在 PCDD/Fs 超标排放的现象(排放水平最高可达 $8.12 \text{ ng} \cdot \text{m}^{-3}$ I-TEQ, 均值为 $0.423 \text{ ng} \cdot \text{m}^{-3}$ I-TEQ)^[20, 24–26, 92–95]。基于已有文献报道(表 4), 目前固体废弃物焚烧行业的 PCDD/Fs 末端控制技术基本以 AC+BF 为主, 配以 SDS、DS、WS、SCR、SCNR 等不同技术组成 APCDs, 可有效降低烟气中 PCDD/Fs 浓度^[96–98]。

2.2 钢铁生产

钢铁生产流程较长, 不同工序烟气理化性质差异较大, 其中烧结因烟气温度高、含尘量大等原因不适合使用 BF, 而静电除尘器(ESP)使用较为普遍^[99]。2018 年, 我国烧结和炼钢的 PCDD/Fs 达标率仅为 33.3% 和 66.7%^[100–101]。近期研究表明^[36, 102], 截止 2021 年, 我国钢铁行业排放烟气中 PCDD/Fs 的浓度范围为 $0.05\text{--}2.93 \text{ ng} \cdot \text{m}^{-3}$ I-TEQ, 均值为 $0.42 \text{ ng} \cdot \text{m}^{-3}$ I-TEQ, 同 2005—2019 年相比下降 1—2 个数量级, 能够满足 $0.5 \text{ ng} \cdot \text{m}^{-3}$ I-TEQ 的排放要求。钢铁生产行业的 PCDD/Fs 末端控制技术以 ESP 或 BF 为

主,配备SCR、湿法脱硫等脱硫脱硝技术组成的APCDs(表4),可对PCDD/Fs等污染物进行协同控制。尽管排放烟气中PCDD/Fs的浓度能够达到 $0.5\text{ ng}\cdot\text{m}^{-3}$ I-TEQ的排放要求,但其排放浓度仍普遍高于固体废弃物焚烧行业。因此钢铁行业尤其是烧结、电炉炼钢等工序的PCDD/Fs排放形势仍较为严峻,相关污染控制研究工作需进一步加强。

表4 工业烟气PCDD/Fs控制技术

Table 4 Collaborative dioxin control technology for industrial flue gas

工业类型 Industrial Type	空气污染控制装置 Air pollution control devices(APCDs)	进口 Before	出口 After	效率 Efficiency	参考文献 Reference
生活垃圾焚烧	SDS+DS+AC+BF+SCR	0.2253	0.0028	98.76%	[89]
		—	0.0365	—	[98]
	SNCR+SDS+AC+BF	—	0.076—0.153	—	[107]
		—	0.007—0.095	—	[25]
	SDS+AC+BF+SCR	—	0.41	—	[108]
		—	0.06	—	[108]
	热交换+SDS+AC+BF	2.58	0.0246	99%	[109]
	急冷+SDS+AC+BF	—	0.45	—	[94]
		—	0.078	—	[110]
	SDS+AC+BF	—	0.008—0.12	91.7%—99.3%	[93]
固体废弃物焚烧		—	0.026	—	[94]
		—	0.099	—	[98]
	DS+AC+BF	—	0.0844	—	[38]
	WDS+AC+BF	—	0.082	—	[98]
	AC+BF	—	0.239	—	[38]
	CY+SDS+BF	—	0.54	—	[111]
	WDS+BF	—	0.50	—	[94]
	SDS+BF	—	1.33	—	[94]
	CY+ESP	16.137	0.946	94.14%	[112]
	CY+ESP+BF	0.23	1.948	-747%	[112]
		0.436	5.018	-1051%	[112]
危险废物焚烧	VS+CY+AC+BF	113	0.054(WHO)	99.95%	[90]
	SDS+AC+BF	—	0.01—11.91	—	[92]
	AC+BF	—	0.225	—	[38]
医疗废弃物焚烧	SDS+AC+BF+WDS	5.32	0.07	98.68%	[91]
	DS+AC+BF	—	1.64	—	[38]
	SDS+AC+BF	—	0.07—12.21	—	[92]
	SDS+BF	—	0.07	—	
电弧炉炼钢	BF	—	0.17	—	[113]
		—	0.148—0.757	—	[38]
		—	0.34	—	[37]
钢铁生产	ESP+脱硫	—	0.003—0.557	—	[36]
	ESP+WFGD	2.3 ± 0.56	0.99 ± 0.53	—	[99]
	烧结	ESP+SFGD	0.32—0.69	0.022—0.2	[99]
		WFGD+WESP	—	0.15	[103]

续表 4

工业类型 Industrial Type	空气污染控制装置 Air pollution control devices(APCDs)	进口 Before	出口 After	效率 Efficiency	参考文献 Reference
钢铁生产	烧结	ESP+SCR	—	0.137—0.657	— [38]
		ESP	—	0.233	— [38]
		BF	—	0.005—0.48	— [37]
	炼焦	BF	—	0.006—0.057	— [36]
		—	(4.9—89.3)×10 ⁻³ (WHO)	—	— [33]
		BF	—	0.00870	— [38]
有色金属生产	再生铜	—	(0.0039—0.03)×10 ⁻³	—	[114]
		—	0.310	—	[38]
		—	0.84	—	[115]
		BF	—	0.004—0.37	— [46]
	再生锌	—	0.009—1.29	—	[47]
		GS或ESP+BF	—	0.48	—
		BF	—	0.05	— [103, 115]
	再生铅	ESP+GS+BF	—	0.037	—
		BF+WDS+DS	—	—	[37]
		AC+BF	—	0.1	— [45]
水泥窑	再生铝	BF	—	(5.68—44)×10 ⁻³	— [38]
		—	2.05	—	[37]
		WDS	—	0.88	— [37]
	水泥窑协同处置	—	5.9×10 ⁻³	—	[115, 103]
		ESP	—	(9.3—49.3)×10 ⁻³	— [116]
		—	0.01—0.19	—	[55]
	WDS	—	0.076	—	[106]
		BF	—	(17.8—90.8)×10 ⁻³	— [116]
		—	0.01—0.46	—	[55]

单位: ng·m⁻³ I-TEQ; “—”: 未提及; WDS: 湿法除尘器; CY: 旋风除尘器; VS: 文丘里洗涤器; WFGD: 湿法脱硫; SFGD: 半干法脱硫; WESP: 湿法静电除尘; GS: 重力沉降

unit: ng·m⁻³ I-TEQ; “—”: Not Reported; WDS: Wet dust collector; CY: Cyclone dust collector; VS: Venturi scrubber; WFGD: Wet flue gas desulfurization; SFGD: Semi-dry desulphurization; WESP: Wet electrostatic precipitator; GS: Gravity settling

2.3 有色金属生产

有色金属生产通常采用的烟气 PCDD/Fs 控制技术见表 4。目前我国对再生有色金属生产行业烟气 PCDD/Fs 的排放限值为 0.5 ng·m⁻³ TEQ^[87]。研究表明^[47, 103], 有色金属生产厂采用以 BF 或 ESP 为主要控制技术时, 烟气中 PCDD/Fs 排放水平为 0.009—0.13 ng·m⁻³ I-TEQ, 能够持续满足 0.5 ng·m⁻³ TEQ 的限值要求。但部分工厂排放 PCDD/Fs 的水平接近甚至超过限值要求(表 4), 且有色金属生产厂 PCDD/Fs 排放超标率可达 22.2%^[101]。这表明能否有效利用现有控制技术(如 BF 或 ESP 为主的烟气污染控制系统)对有色金属行业的 PCDD/Fs 污染控制具有重要影响。

2.4 水泥窑协同处置

水泥窑协同处置行业针对烟气污染采取的控制技术和手段见表 4。水泥工业本身对排放的烟气中 PCDD/Fs 浓度水平并无明确限值, 目前协同处置固体废物的水泥窑烟气中 PCDD/Fs 排放限值为 0.1 ng·m⁻³ TEQ^[88, 104]。尽管水泥窑协同处置固体废物时原料中 PCDD/Fs 浓度较高, 但经过高温分解处理后, 烟气采用 BF、BF+SNCR(选择性非还原催化)或 ESP 为主的 APCDs 进行净化, PCDD/Fs 排放水平可达 0.011—0.076 ng·m⁻³ I-TEQ^[55, 103, 105—106], 均可使 PCDD/Fs 以较低浓度排放。

2.5 不同工业过程 PCDD/Fs 污染控制效果比较

从表 4 中可以看出, 目前固体废弃物焚烧行业多以 BF 或 AC+BF 为主要技术, 配备 SDS、DS 等非 ESP 技术组成的 APCDs 对烟气进行深度净化; 钢铁生产、有色金属生产、水泥窑协同处置行业则以 BF 或 ESP 为主要技术, 配备 SDS、脱硫脱硝等技术组成的 APCDs 对烟气进行处理。当过程控制和末端控制均能得到有效保障时, 烟气中 PCDD/Fs 的排放水平多处于较低水平, 但钢铁生产和有色金属生产的 PCDD/Fs 排放强度仍然明显高于其他行业。

3 结果与展望(Results and perspectives)

本文系统的总结了固体废弃物焚烧、钢铁生产、有色金属生产和水泥窑协同处置等四类主要行业烟气中 PCDD/Fs 排放特征及污染控制的研究进展。从排放特征来看, 水泥窑协同处置行业排放的 PCDD/Fs 以 4—6/7 氯代同族体为主要单体, 而固体废弃物焚烧、钢铁生产、有色金属生产等行业排放的 PCDD/Fs 以 7—8 氯代同族体为主, 四类工业源的主要生成机理均为从头合成; 从现有排放量数据来看, 2004—2016 年我国大气 PCDD/Fs 总排放量上升明显, PCDD/Fs 排放量依次为: 钢铁生产>固体废弃物焚烧>有色金属生产>水泥窑协同处置; 依据现有文献数据进行估算, 2020 年 PCDD/Fs 排放量依次为钢铁生产>水泥窑协同处置>有色金属生产>固体废弃物焚烧, 但水泥窑协同处置固体废弃物产生 PCDD/Fs 的研究仍比较有限, 相关工作亟需加强。

从烟气中 PCDD/Fs 的控制技术来看, 在传统污染控制装置基础上增加活性炭吸附、催化剂、抑制剂等可以有效降低 PCDD/Fs 的大气排放^[117–119]。但目前各类工业源的 PCDD/Fs 末端控制多以除尘器结合吸附脱除装置为主, 并未实现 PCDD/Fs 的总量削减; 此外由于记忆效应导致的 PCDD/Fs 排放水平变化以及飞灰中高浓度 PCDD/Fs 带来的固废处置问题也给现有技术升级带来较大难度^[46, 89, 120–123], 因此, 如何有效控制 PCDD/Fs 的排放总量仍然面临极大挑战。末端控制技术方面, AC+BF 吸附技术存在活性炭使用量大、价格高、活性炭吸附效率低、存在记忆效应、产生高毒性粉煤灰等缺点^[108, 124–127], 近年来, 生物质制备活性炭、活性炭改性处理、双袋式除尘器、喷射聚丙烯胺、协同处置粉煤灰、热等离子体、紫外光降解等技术逐渐得到开发, 能够有效改善或避免吸附法存在的问题^[76–80, 128–136], 然而诸多新技术尚停留在实验室阶段, 且 PCDD/Fs 降解技术还存在反应时间长、去除效率不稳定等问题^[137–140]。因此尚无法在企业层面上推广应用。

一些针对工业过程中 PCDD/Fs 生成的研究发现, 传统硫脲、硫酸铵等含 N 或含 S 的抑制剂存在氨溢出、额外成本等问题, 研发使用无氨溢出风险的氧化钙, 或采用含 N 或 P 的污泥等作为抑制剂可降低污染控制成本, 且具有良好的抑制 PCDD/Fs 生成的控制效果^[141–145], 可能是未来控制某些工业过程中 PCDD/Fs 排放的重要技术手段。

基于以上研究现状, 本文对典型工业过程中 PCDD/Fs 排放特征及其污染控制研究做以下两方面展望:

(1) 工业过程是 PCDD/Fs 人为排放的主要来源, 尽管我国已经制定了相关行业的排放标准, 排放总量也有所下降, 但是部分行业仍然存在 PCDD/Fs 排放量增加的趋势, 因此及时更新典型行业 PCDD/Fs 排放因子并完善排放清单, 对于我国履行《斯德哥尔摩公约》和降低 PCDD/Fs 暴露风险具有重要意义。

(2) 对工业排放 PCDD/Fs 的控制应遵循“源头-过程-末端”的全过程控制原则。开发新技术新材料, 通过对原料预处理等措施从源头上弱化 PCDD/Fs 生成条件; 通过添加抑制剂等措施从过程中减少 PCDD/Fs 的生成; 积极研发 PCDD/Fs 的催化降解技术, 结合活性炭吸附等末端控制技术可实现 PCDD/Fs 排放总量的有效削减。

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