



我国PM_{2.5}和臭氧污染前体物排放源清单的现状与质量评估

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摘要 大气污染源排放清单是研究细颗粒物(PM_{2.5})和臭氧污染成因、制定PM_{2.5}和臭氧污染精准协同防控策略的重要基础数据。依托国家科技项目以及各级管理部门的强力推进, 近年来我国PM_{2.5}和臭氧污染前体物排放清单研究与编制工作得到了迅速发展, 积累了相对完善的本土前体物排放因子及PM_{2.5}和VOCs成分谱数据集, 建立了较为系统的前体物排放清单表征方法和编制指南; 交通、卫星等大数据和多种校验评估方法逐渐得到重视与应用, 污染源和污染物覆盖逐渐精细化, VOCs组分排放清单逐渐受到关注, 各种尺度前体物排放清单的时空分辨率、时效性和可靠性有了显著提升, 基本满足区域和城市开展PM_{2.5}与臭氧污染防控对排放源清单的需求。尽管如此, 我国前体物排放清单依然存在不确定性较大、排放因子和成分谱数据建立缺乏规范化评估、组分清单校验薄弱、排放源清单编制质量评估方法缺失等不足。未来工作需要在排放因子和成分谱数据集规范化、排放清单校验与质量评估方法指南编制、近实时和短临预测排放清单方法学与业务化、不同类型排放清单建立方法与评估等方面继续深入。最后, 提出了前体物排放清单编制质量评估方法的思路, 为清单使用者评估前体物排放源清单编制质量、进一步规范前体物排放清单编制工作提供借鉴和参考。

关键词 PM_{2.5}和臭氧, 前体物排放清单, 挥发性有机物组分清单, 校验与评估, 不确定性分析

自2013年《大气污染防治行动计划》(简称“大气十条”)发布以来, 我国开展了以细颗粒物(PM_{2.5})为首要目标的大气污染防治工作, PM_{2.5}污染改善成效显著, 2015~2019年期间337个城市PM_{2.5}年平均浓度大幅度下降了22.0%^[1]。尽管如此, 许多城市PM_{2.5}浓度年平均值尚未达标, 其中京津冀及周边地区、长江三角洲(简称“长三角”)和汾渭平原地区以PM_{2.5}为首要污染物的超

标天数依然分别占总超标天数的48.0%、45.1%和56.4%^[2]。随着一次排放PM_{2.5}大力减排, PM_{2.5}中较难控制的二次污染组分, 如硝酸盐和二次有机气溶胶(secondary organic aerosol, SOA)等, 占比呈现上升趋势^[3]。与此同时, 我国臭氧污染问题逐渐凸显, 呈现污染程度加重、影响范围扩大和污染时段延长的演变态势^[4,5]。相比于2015年, 2019年337个城市臭氧年平均浓度的平

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Huang Z J, Sha Q E, Zhu M N, et al. Status and quality evaluation of precursor emission inventories for PM_{2.5} and ozone in China (in Chinese). Chin Sci Bull, 2022, 67: 1978–1994, doi: 10.1360/TB-2021-0783

均值上升了22.1%^[1],显示我国大气污染格局发生了深刻变化,进入了PM_{2.5}和臭氧污染协同防控的新阶段。

前体物排放清单是研究PM_{2.5}和臭氧污染成因、制定PM_{2.5}和臭氧污染协同防控策略的重要基础数据。臭氧和PM_{2.5}在大气复合污染过程中大量产生,同根同源,且通过多种途径相互影响^[6]。因此,不同于一般的大气污染源排放清单,PM_{2.5}和臭氧污染前体物排放清单更加关注能转化为二次污染或者参与大气自由基循环的二氧化硫(SO₂)、氮氧化物(NO_x)、氨(NH₃)、挥发性有机物(VOCs)及组分、氯(Cl)、气态亚硝酸(HONO)等活性组分^[7,8]。PM_{2.5}一次排放是大气PM_{2.5}的重要来源,其排放的一次有机气溶胶(primary organic aerosol, POA)也能通过老化转化为SOA,因此也是编制PM_{2.5}和臭氧污染前体物排放清单、开展二次污染模拟不可或缺的污染物^[9]。我国大气污染物排放清单研究主要起始于21世纪初,经过20余年的发展和积累,已经构建了较为完整和实际可行的排放清单表征方法体系^[10,11],积累的排放因子和成分谱数据集基本能够支撑PM_{2.5}和臭氧污染前体物排放清单的编制与建立实践,形成了覆盖多种前体物、多种污染源、不同时空尺度的前体物排放清单数据集^[12]。随着我国大气环境管理迈向PM_{2.5}和臭氧污染精细化协同防控阶段,以及大数据时代的到来,管理和研究人员对前体物排放清单的需求正在从清单建立到清单精细化和质量提升的过渡,对前体物排放清单的组分覆盖、时空精度、时效性和可靠性等方面提出了更高的要求。

本文从排放清单表征技术方法体系、基础数据、物种和污染源种类覆盖、不同尺度前体物排放清单建立实践、校验和评估以及大数据应用等方面分析总结了现阶段我国PM_{2.5}和臭氧污染前体物排放源清单的发展现状,同时也分析了我国前体物排放源清单在排放因子数据集、成分谱数据集、VOCs组分清单种类以及质量评估和校验等方面的不足。鉴于质量评估对前体物排放源清单改进、业务化应用与政策制定和规划管理至关重要,本文也尝试性地提出了前体物排放源清单编制的质量评估方法思路,为清单使用者评估排放清单质量、进一步规范前体物排放清单编制流程提供借鉴和参考。

1 PM_{2.5}和臭氧污染前体物排放源清单的发展现状

我国对排放清单的研究起步相对较晚,许多工作

是随着我国区域大气污染问题的出现及认识的深入而展开的。自20世纪80年代末至90年代,我国大气污染主要以煤烟型污染和酸雨污染为主,因此大部分排放清单主要关注电厂、民用和重点行业等能源燃烧部门的SO₂、NO_x、可吸入颗粒物(PM₁₀)和粉尘排放以及农业部门的NH₃排放^[13,14],少数涉及扬尘和生物质燃烧等^[15]。这些排放清单多数利用国家、部门及各省市统计年鉴公布的统计数据,以及各行业部门的统计信息,采用国外的排放因子(如AP-42排放因子库),建立“自上而下”的国家尺度排放清单,空间分辨率往往较为粗糙,一般为100 km左右,清单结果也主要服务于国家污染物总量控制,甚少用于空气质量模型的相关研究^[10]。随着我国大气复合污染问题的日渐突出,排放清单逐步引起了研究者和管理部门的重视,大气污染源排放清单研究得到迅速发展。如图1所示,自2000年以来,我国发表的排放清单文献迅速增加。在表征方法、污染源和污染物覆盖、基础数据质量、校验与评估等方面均有长足的提升,能够支撑不同尺度PM_{2.5}和臭氧污染前体物排放清单的建立,基本满足不同区域和尺度的二次污染防治的需求。本文从以下几个方面总结我国前体物排放清单的发展与研究现状。

1.1 排放源清单表征方法体系

以国家“十五”科技攻关计划、“十一五”国家高技术研究发展计划(“863”计划)重大项目课题、大气重污染成因与治理攻关、国家重点研发计划等项目为依托,我国学者在大气排放源分类、排放清单定量表征、排放因子和成分谱建立、前体物组分清单构建、时间与空间分配、不确定性分析、校验与评估等方面开展了较为系统的方法学研究,初步发展形成了区域和城市高分辨率大气污染物排放清单建立的方法体系^[10,11]。2013年,我国按照“规范统一、科学实用、轻重缓急、循序渐进”的原则,提出了中国排放清单编制技术指南体系构建的基本思路,并于2014~2016年间,在清华大学等单位的努力下,先后发布了专门针对PM₁₀、PM_{2.5}、VOCs、NH₃以及道路机动车、非道路移动源、生物质燃烧源、扬尘颗粒物和民用煤燃烧的大气污染源排放清单编制技术指南^[16,17],初步形成了我国大气污染物排放清单编制技术支撑体系。排放清单编制逐步向规范化和业务化迈进。针对日益增长的城市空气质量精细化管理与区域联防联控需求,环境保护部于2017年组织相关研究机构进一步编制了《城市

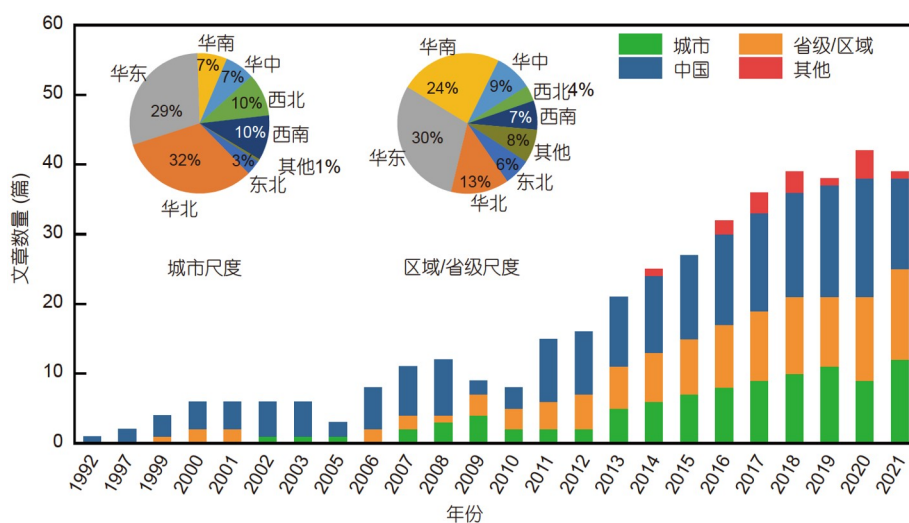


图1 中国不同年份大气污染源排放清单文献统计, 及城市和区域/省级尺度的不同区域文献占比. 中文文献为304篇, 来自中国知网数据库的北大中文核心期刊要目总览和EI来源期刊的检索结果, 检索关键词为排放清单和排放源清单; 英文文献335篇, 来自Web of Science数据库期刊的检索结果, 检索关键词为emission/emissions+China和emission inventory/inventories+China. 所有结果均剔除温室气体和非排放清单编制的文献

Figure 1 The annual number of papers associated with emission development in China and the percentage of papers from different regions at city and regional/provincial scales. With the keywords emission/emissions+China and emission inventory/inventories+China, 304 papers from Chinese core journal criterion of PKU (Peking University) or EI journal were obtained from the database of China National Knowledge Infrastructure (CNKI) and 335 SCI papers were obtained from the Web of Science database. Papers on greenhouse gases or without emission developments were excluded

大气污染物排放清单编制技术手册》和《“2+26”城市大气污染防治跟踪研究工作手册》^[10,11], 初步形成了规范化、涵盖全行业和多污染物、具备时空变化信息的高分辨率网格化城市和区域人为源大气污染物综合排放清单编制技术体系. 然而, 在实践应用中发现了这些指南中存在一些问题. 例如, 化工、建材、溶剂等行业的产品和原辅料种类缺失较多, VOCs无法细分到组分, 指南中部分的排放因子与实际情况差异较大, 特别是VOCs、PM_{2.5}、NH₃等污染物. 针对这些问题, 2020年国家大气污染防治攻关联合中心设立了臭氧和PM_{2.5}复合污染协同防控科技攻关项目, 部署了数个排放清单编制指南更新课题, 正在从污染源覆盖完整性、排放因子本土化与更新、排放源成分谱数据集本土化、VOCs组分清单、臭氧生成潜势(ozone formation potential, OFP)、时空分辨率等方面推进臭氧和PM_{2.5}污染前体物排放清单编制技术体系的进一步完善.

1.2 本土排放因子和成分谱数据建立

在过去的20年, 我国学者针对机动车、电厂、水泥、钢铁、船舶、生物质燃烧、有机溶剂使用等重点排放部门和行业开展了大量的排放因子本土实测和优化工作, 建立了大量基于生产技术、末端处理技术、

环境与气象条件等精细分类的污染物排放因子^[12]. 例如, 环境保护部颁发的《道路机动车大气污染物排放清单编制技术指南》基于本土实测数据将排放因子精细分类至燃料类型、车型、排放标准四级源分类, 并基于本土机动车行驶特征与气象参数对排放因子进行了修正(https://www.mee.gov.cn/gkml/hbb/bgg/201501/t20150107_293955.htm). 我国在2007和2017年开展的两次污染源普查也给出了基本覆盖主要污染源的SO₂、NO_x、PM、VOCs等前体物排放系数^[18]. Li等人^[12]详细总结了主要污染源本地实测排放因子, 指出我国电厂PM_{2.5}排放、工业锅炉排放、工业溶剂排放、秸秆燃烧以及农牧源的排放因子依然匮乏或者存在较高的不确定性. 因此近年来, 一些学者也针对本土化不足、更新滞后或缺失排放因子的超低燃煤电厂^[19,20]、生物质燃烧锅炉^[21]、机动车及其NH₃排放^[22,23]、溶剂使用源^[24,25]和船舶源^[26]等开展了排放因子实测工作. 近年来, 我国学者也开始关注天然源异戊二烯^[27]、土壤HONO^[28]、人为源HONO^[28-30]、燃烧源Cl^[31]以及机动车HCHO^[32]等关键前体物组分的排放测试, 其中土壤^[33]、机动车^[30]、船舶^[34]、生物质燃烧^[35]等重点源HONO排放因子已经基本实现本土化. 通过大量测试和调研数据的积累, 我国已初步建立了可反映本土排

放特征、覆盖重点行业的排放因子,并逐步精细到主要子行业 and 关键组分,逐步实现重点行业关键前体物本土排放因子的全行业覆盖,基本摆脱了对国外排放因子数据库的依赖。

除了排放因子,我国在VOCs与颗粒物(particulate matter, PM)成分谱方面也开展了大量的本土测试,污染源逐步覆盖了工业源、交通运输源、溶剂使用源、生物质燃烧源、扬尘源、餐饮源等主要VOCs与PM污染源^[36],建立了可反映中国本土VOCs和PM重点源排放特征的成分谱数据集。在PM成分谱数据集方面, Liu等人^[37]建立的CSPSS(China Source Profile Shared Service)成分谱数据库收录了超过200条PM成分谱。曹军骥等人(<http://www.sourceprofile.org.cn/home/article/index/id/24.html>)建立的KLACP成分谱数据库覆盖了工业、燃煤、机动车、扬尘、生物质燃烧、开放源六大类大源,收录了元素、无机离子、碳组分、有机组分,以及同位素等组分。在VOCs成分谱方面, Mo等人^[38]构建的VOCs成分谱数据库收录了85条中国本土源成分能谱,覆盖了五大类排放源和75种常见组分。随着测试手段和分析技术的升级,VOCs成分谱覆盖的组分数量显著提升。例如,2020年,Zhou等人^[39]建立的四川省VOCs成分谱数据集已经覆盖了519种组分。Sha等人^[40]在广泛收集我国现有本土成分谱研究成果的基础上,结合现场测量弥补了机动车、溶剂使用源等关键排放源的甲醛等含氧VOCs(oxygenated volatile organic compounds, OVOCs)组分,并对收集和实测的成分谱进行了质量评估、筛选和融合,构建了覆盖近400种组分,8类大源、101类子源、包含350多条中国本土测试的综合VOCs成分谱数据集。这些数据覆盖的污染源和物种不断完善,本土化程度逐渐提高,基本上可支撑我国PM_{2.5}和臭氧污染前体物组分清单建立、二次污染溯源、空气质量模拟、PM_{2.5}与臭氧污染防控及管控决策等工作。

1.3 排放源清单涵盖的污染源和组分

前体物排放清单的污染源分类精细程度也逐步提升。能源部门的源分类从火力发电为主,逐步扩展到火力发电、供热、炼焦生产、工业燃烧、民用燃烧等,燃料类型从以煤为主逐步扩展到燃煤(进一步细分煤型)、焦炭、原油、汽油、柴油、燃料油、天然气、成型生物质燃料等多种类型^[41]。水泥、钢铁等一些重点行业发展了基于技术水平和设备信息的排放清

单^[36,42,43]。此外,前期关注较少的溶剂使用行业、机动车蒸发排放、餐饮等污染源也纳入前体物排放清单表征^[43-45]。总体上,现阶段研究已经将污染源分类精细到4级源,涵盖超过上百种污染源。例如,清华大学建立的MEIC(the Multi-resolution Emission Inventory for China)排放清单涵盖700余种污染源^[46];Huang等人^[43]建立的广东省排放清单涵盖了约800种污染源。

我国排放清单研究工作关注的污染物种类逐渐从SO₂、NO_x和CO扩展至SO₂、NO_x、CO、PM₁₀、PM_{2.5}、BC、OC、VOC和NH₃。同时,为了开展大气氧化性研究以及提高二次污染过程模拟的准确性,国家和区域尺度的PM_{2.5}和VOCs组分排放清单也相继建立。其中区域尺度组分排放清单研究主要集中在珠三角和长三角地区^[43,47-51]。随着本土源排放测试研究的推进,组分排放清单建立过程中采用的本土成分谱数据占比逐渐提升,涉及的VOCs组分数量也大幅度增加。例如,Streets等人^[47]最早采用美国SPECIATE数据库建立了覆盖19种VOCs组分的中国排放清单。Zhang等人^[48]与Wu和Xie^[52]在SPECIATE数据库的基础上,进一步融合了我国本土实测的VOCs成分谱,并弥补了部分以往研究缺失的OVOCs组分,完善了中国VOCs物种排放清单。Wu和Xie^[52]采用相同的方法建立了覆盖153类VOCs组分的中国组分排放清单。不断完善的组分排放清单部分改善了VOCs排放活性低估的不足。例如,相比Zheng等人^[53]在2009年建立的珠三角组分排放清单,Huang等人^[43]在2021年建立的广东省VOCs组分排放清单新增了近280种VOCs组分,其中120多种组分为OVOCs,这些新增的VOCs组分贡献了近30%的珠三角OFP(图2)。涉及PM_{2.5}组分的排放清单研究虽然较少,但排放清单中考虑的PM_{2.5}组分数量在近年来同样呈现增加的趋势。例如,长三角组分排放清单中的PM_{2.5}组分数量从2013年的18种^[54]增加到了2021年的43种^[55]。除了VOCs和PM_{2.5}组分清单,我国学者也开始关注S/IVOCs(semi-volatile and intermediate VOCs)、氯和气态亚硝酸(HONO)等排放清单^[43,56-59],但目前相关研究还相对较少。Wu等人^[57,58]先后采用了POA比值法建立了珠三角及中国的IVOCs和SVOCs排放清单,但由于缺乏直接观测的IVOCs和SVOCs排放因子,IVOC和SVOCs排放清单依然存在较大的不确定性;我国现阶段建立的氯排放清单大部分为国家尺度^[7,60],主要关注气态Cl₂、HCl和二氯甲烷等有机氯以及颗粒氯^[61];HONO排放清单主要为船舶^[34]和机动车^[30]等重点源排放清

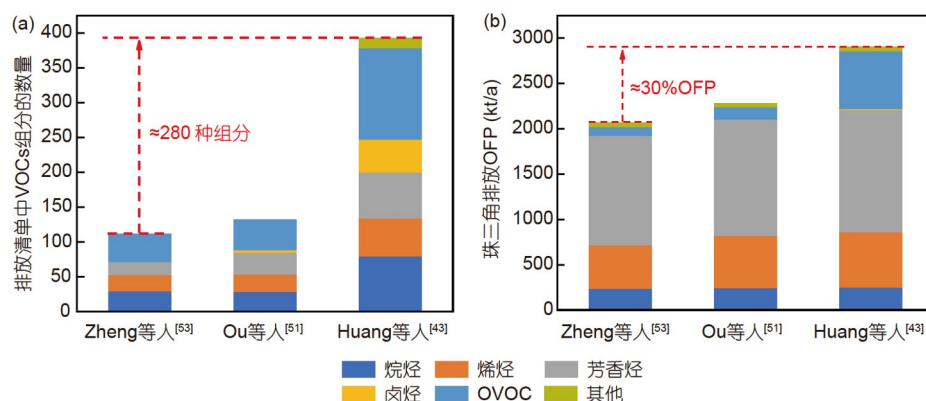


图2 2009、2015和2021年建立的VOCs组分清单覆盖的物种数量(a)和珠三角OFP排放量(b)。其中各研究的OFP排放量均采用2017年珠三角VOCs排放总量进行重新计算

Figure 2 The number of VOC species (a) and OFP emissions in the Pearl River Delta (PRD) region (b) developed in 2009, 2015 and 2021. The OFP emissions for each study were recalculated using the total VOCs emissions in PRD in 2017

单, 还缺乏综合HONO排放清单。

1.4 前体物排放清单编制与建立实践

随着PM_{2.5}和臭氧污染研究与管控的深入, 我国已经建立了“国家-区域-城市”尺度前体物排放清单数据集。在国家以及更大尺度方面, 我国学者建立了INTEX-B(the Intercontinental Chemical Transport Experiment Phase B)排放清单^[48]、中国多尺度排放清单模型MEIC(<http://meicmodel.org/>)^[62]、亚洲人为源排放清单MIX^[46]、北京大学全球尺度等综合排放清单^[63]以及生物质燃烧、机动车、电厂、工业源、非道路移动、天然源、NH₃排放、NO_x排放、VOCs等单源或者单污染物排放清单^[49,64~71]。相比TRACE-P(the Transport and Chemical Evolution over the Pacific)^[47]和REAS(Regional Emission Inventory in Asia)等东亚排放清单^[72], 我国学者建立的国家排放清单采用了更为详细的活动水平数据和更多的本地排放因子, 空间分辨率最高达到1 km^[73], 能更好地反映我国污染源的排放特征。在区域和省级尺度方面, 前体物排放清单研究主要集中在华东地区(以长三角为主)^[45,55]、华南地区(以珠三角为主)^[43,50]、华北地区(以京津冀为主)^[74~76]以及西南地区(以川渝为主)^[77]等(图1)。并且相比国家尺度排放清单, 通常具有更高的空间分辨率, 普遍为3~5 km(图3), 部分能够达到1 km, 物种和污染源覆盖也更加精细与全面。国家尺度和区域尺度排放清单为研究区域二次污染传输与形成、空气质量预报预警、制定区域联防联控等工作提供了数据支撑。而利用实地调研的信息, 城

市尺度排放清单往往能更好地表征城市污染源排放的空间特征和行业结构特征, 是我国城市开展PM_{2.5}和臭氧污染防控的基础数据。在环境保护部的推动下, 2014年北京、上海、广州、深圳、南京、天津、武汉、济南、沈阳、石家庄、成都、乌鲁木齐、福州和长沙作为试点城市率先探索了城市排放清单编制工作。随后, 京津冀大气污染传输通道上的“2+26”城市也开始编制城市排放清单^[78]。为此, 2014年之后的城市尺度排放清单研究迅速增加, 并且集中在我国华北地区(以京津冀为主)和华东地区(以长三角为主)(图1)。超过50%的城市尺度网格化排放清单的空间分辨率能达到1 km以上(图3)。

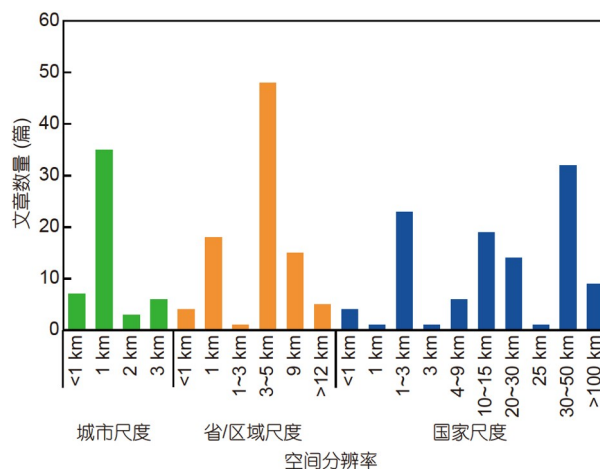


图3 中国前体物排放清单的空间分辨率分布

Figure 3 Spatial resolution distributions of precursor emission inventories in China

总体上,我国PM_{2.5}和臭氧污染前体物排放清单时空精度不断提升,编制尺度逐渐向城市甚至更小尺度发展。然而,当前的前体物排放清单还存在较大的不确定性。图4统计了中国50多篇文献(见补充材料)的综合排放清单不确定性的量化结果。总体上,SO₂和NO_x排放的不确定性相对较小,其不确定性范围上限均值分别为26%(上限的95%置信区间为9%~66%)和28%(15%~56%);而PM_{2.5}、VOCs和NH₃排放的不确定性较大,分别为55%(13%~111%)、54%(11%~122%)和46%(16%~107%)。由于前期研究缺乏本土化的组分排放因子,基于成分谱建立的VOCs组分、OC和BC排放清单不确定性均值超过100%;在所有的污染源中,扬尘、生物质燃烧、溶剂使用等污染源的不确定性较为突出,其中扬尘源PM_{2.5}和PM₁₀排放的不确定性上限平均分别为135%(21%~265%)和82%(20%~160%),生物质燃烧源的不确定性上限均值处于85%~140%,溶剂使用源VOCs排放的不确定性上限均值为127%(75%~161%)(表1)。这些污染物和排放源是未来前体物排放清单改进的重点之一。

1.5 前体物排放清单校验与评估

校验与评估是提升前体物排放清单质量的重要手段。SO₂、NO_x和BC等一次污染物由于其相对较低的反应活性和较为充足的观测数据,校验手段相对较多。现有研究多采用横向对比法、趋势分析法、反演校验、

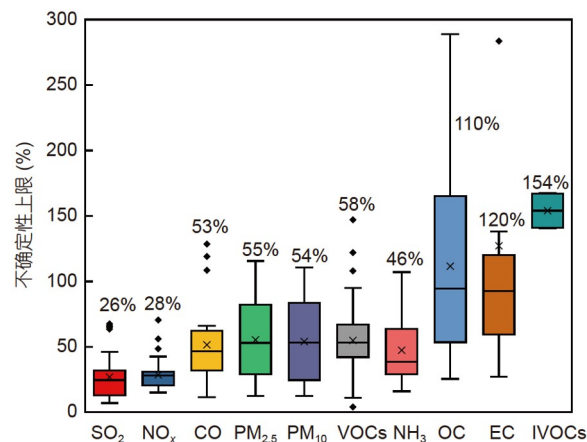


图4 中国不同污染物的不确定性范围上限,统计数据来自本文收集的有量化不确定性结果的综合源文献(见补充材料)

Figure 4 Upper limits of uncertainty ranges of various pollutants in China, based on the collected papers with quantified uncertainty (see Supplementary materials)

直接与地基或卫星遥感观测对比或者利用模式模拟与观测间接对比等方法评估排放清单在总量、时间、空间等方面的合理性^[50,79,80],并进行优化^[81,82]。近年来,也有研究通过同位素源解析校验BC排放的源结构^[83]。总体上,SO₂排放清单与观测数据或者同类型清单具有较好的一致性,而NO_x排放清单则存在一定差异性,可能与电厂、工业锅炉和移动源排放的不确定性较大有关^[84-87]。此外,反演前后的BC排放清单也存在较大差异,表明BC排放清单还需要进一步改进^[88-91]。

表1 中国不同污染源的不确定性范围上限^{a)}

Table 1 Upper limits of uncertainty ranges of different sources in China

污染源	SO ₂	NO _x	CO	PM _{2.5}	PM ₁₀	VOCs	NH ₃	OC	EC
电厂	31%	20%	45%	54%	58%	43%	32%	26%	48%
固定燃烧	95%	83%	101%	83%	84%	114%	52%	110%	210%
工业过程源	54%	44%	68%	63%	60%	70%	57%	114%	119%
溶剂使用源						127%			
存储运输源						97%			
道路移动源	68%	43%	63%	41%	77%	64%	81%	97%	108%
非道路	50%	38%	66%	56%	42%	68%		100%	191%
生物质燃烧	106%	93%	99%	121%	125%	129%	85%	131%	126%
农牧源							57%		
扬尘源				135%	82%			251%	276%
天然源						71%	18%		
餐饮						79%		301%	337%

a) 统计数据来自本文收集的、有量化不确定性的单源或综合源前体物排放清单文献(见补充材料)

相比于 SO_2 、 NO_x 和BC, 大气 NH_3 容易受到大气反应和干湿沉降影响, 其排放清单校验与评估也更为困难, 相关研究还较为匮乏. 现有研究主要利用反演校验和基于稳定氮同位素溯源方法校验评估 NH_3 排放. 例如, Kong等人^[92]利用我国的大气氨观测网络数据和卡尔曼滤波反演模型优化了中国 NH_3 月排放特征. Pan等人^[93]利用稳定氮同位素溯源技术发现, 非农业源对大气环境 NH_3 的贡献可能超过农业源, 机动车尾气和工业氨气排放的强度有可能被低估.

不同研究建立的VOCs及其组分排放清单存在较大差异^[94], 但VOCs组分复杂且大部分组分活性高, 排放后容易发生大气二次转化, 导致VOCs及组分排放清单的校验极具挑战. 现有研究多采用基于观测数据的“自上而下”方法对VOCs排放清单进行验证, 主要方法有: (1) 通过盒子模型或采用惰性污染物作为参比物的排放比法校验VOCs组分排放量; (2) 利用排放清单驱动模式, 然后通过对VOCs或臭氧观测值和模拟值校验排放清单; (3) 利用受体模型源解析校验排放清单源结构. 校验结果表明, 烯烃、烷烃和苯等组分排放基本可靠, 但OVOCs排放普遍低估, 需要进一步改进^[95-97]. 然而, 这些校验方法本身也具有较大的不确定性, 通常需要综合多种手段进行评估^[97]. 特别是化学活性较高的VOCs组分, 其排放清单校验需要考虑光化学反应的影响. 有研究指出, 由于光化学反应的VOCs损耗, 基于大气环境VOCs组分浓度的受体模型解析难以直接与VOCs排放清单解析进行对比^[98]. 为此, 我国学者发展了OH光化学年龄、受体模型解析-线性拟合等方法降低化学损耗对VOCs排放校验的影响, 提高利用观测数据校验VOCs及组分排放的可行性^[99]. 例如, Yuan等人^[100]根据观测浓度计算VOCs化学损耗的新算法, 构建了基于活性VOCs化学消耗的源解析新模型, 提高VOCs活性组分来源解析的可靠性. 总体上, VOCs及其组分清单校验依赖于高质量的VOCs组分观测, 但现阶段我国VOCs组分观测站点少, 而已有的组分观测数据通常缺乏醛酮类等高活性化学组分, 导致VOCs排放的校验研究还非常薄弱.

不确定性分析是对编制过程可能导致排放清单的不确定性来源及影响进行诊断分析, 也是改进排放清单质量的重要手段. 不确定性分析可分为定性、半定量和定量分析^[101]. 其中常见的定性分析方法有数据质量评级方法; 半定量分析方法有数据质量指数(data quality indicator, DQI)^[102]和NUSAP(numeral unit spread

assessment pedigree)方法^[103]; 定量分析方法能够量化清单的不确定性, 识别关键不确定性来源, 指导排放清单改进, 是排放清单不确定性分析的优选方法, 其常见的不确定性传递方法有基于蒙特卡罗或拉丁超立方抽样的随机模拟法、自展模拟、模糊分析和误差传递方法等^[102]. 总体上, 我国前体物排放清单编制对不确定性分析越来越重视, 但在不确定性分析与应用方面还存在较大不足, 具体表现在: (1) 应用不确定性分析的研究还相对较少, 在本文统计的600多篇文献(见补充材料), 仅有近40%的排放清单研究进行了定量不确定性分析, 20%左右的研究定性讨论了排放清单的不确定性(图5); (2) 部分研究人员对可变性、测量精确度、数据代表性等引起排放清单输入参数不确定性的来源缺乏认识, 导致输入参数不确定性量化过程容易受主观性影响, 不确定性分析结果缺乏可靠性; (3) 排放模型参数、活动水平和排放因子等排放清单重要输入参数的不确定性通常被低估或忽略, 导致排放清单量化的不确定性整体偏低; (4) 当前大部分研究以排放清单不确定性范围量化作为主要目标, 但很少研究利用不确定性信息指导排放清单改进, 部分研究也会错误地将不确定性量化结果作为评估排放清单准确性的指标.

1.6 大数据在排放清单中的应用

随着物联网和计算机的发展, 大数据在近年来也逐渐应用于我国电厂、工业源、机动车、扬尘源、生物质开放燃烧、船舶等人为源排放表征, 提升排放清单的时空精度和时效性. 例如: 利用浮动车GPS数

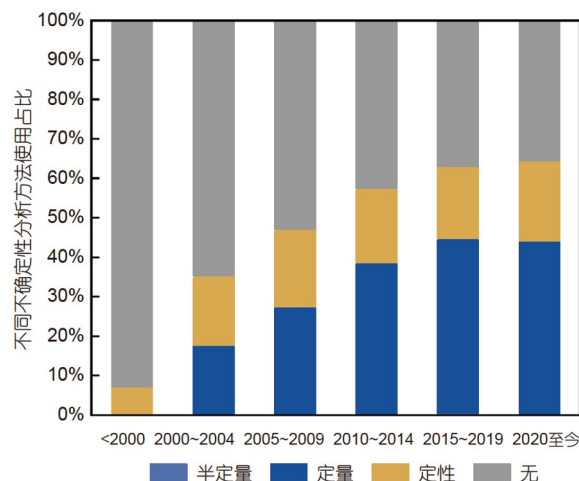


图5 不确定性分析在我国排放清单研究的应用情况
Figure 5 Applications of uncertainty analysis in China's emission inventories

据^[104,105]、商业地图导航数据^[106]、交通拥堵系数^[107]、卡口流量^[108]、地磁断面^[104]、射频识别^[109]和视频识别数据^[110]等交通大数据解析的路网车速、流量和车队信息,我国学者发展了多种机动车排放表征方法,建立了北京、上海、广州、南京等城市机动车排放清单^[104~106,111]以及区域尺度柴油车排放清单^[112],空间分辨率最高达到街区尺度^[111],部分方法已经初步实现机动车排放的近实时表征^[105]。基于AIS(automatic identification system)的动力法,建立了长三角、珠三角、渤海湾等沿海区域以及中国近海船舶动态排放清单^[68,113~115],时间分辨率最高可精细到分钟级,空间分辨率最高可达100 m左右,能够反映船舶排放在不同尺度的时空分布特征^[116]。利用MODIS(Moderate Resolution Imaging Spectroradiometer)、VIIRS(Visible Infrared Imaging Radiometer)、GOES(Geostationary Operational Environmental Satellite)等卫星遥感火点产品、辐射能量或过火面积产品等卫星数据,建立了多个覆盖中国地区的生物质开放燃烧动态排放清单产品,包括GFED(Global Fire Emissions Database)、FINN(Fire INventory from NCAR)、GFAS(Global Fire Assimilation System)、QFED(Quick Fire Emissions Dataset)等全球尺度产品^[117~120],以及AMMABB(African Monsoon Multidisciplinary Analysis Biomass Burning)全球区域尺度产品^[121],时效性上最高可实现仅滞后一天的排放动态更新。卫星遥感数据提供的大范围遥感影像数据空间分辨率可达1 m甚至更高,有研究也利用卫星遥感数据识别的分散、无规则的建筑、裸露地面和铺装道路信息,提高我国区域和城市尺度的扬尘排放表征^[122,123],其空间分辨率最高可达30 m;进一步结合谷歌地球平台(Google Earth engine)提供的每月卫星影像产品,扬尘排放清单还能实现月动态更新^[124]。NH₃排放受到气象因子、土壤条件、管理方式等因素的影响,并且在土壤植被和大气之间存在双向交换。为了提高NH₃排放表征的时空精度,有学者利用高时间分辨率的气象数据作为驱动,通过耦合NH₃双向交换模型、生态模型和空气质量模型等,实现农业和畜牧养殖NH₃排放的动态表征,时间精度最高可达1 h^[125,126]。固定源在线监测(continuous emission monitoring system, CEMS)获取的高时间分辨率浓度数据能够准确体现烟囱口污染物的小时排放变化,为此国内也有研究建立了基于CEMS数据的电厂和工业源的小时排放清单^[127~129]。互联网获取的POI(point of interest)数据被用

于提升餐饮、农牧源、工业溶剂和加油站蒸发等面源排放的空间特征表征^[130,131]。以上研究大多关注单种污染源的排放表征,为了实现所有污染源排放的动态表征,也有研究尝试利用地面观测数据或者卫星观测数据,基于排放反演技术建立综合排放清单的近实时动态更新方法。然而,这种方法目前只能动态更新NO_x、CO等活性较低的污染物排放,并且难以表征排放清单的源结构变化^[132,133]。

现阶段利用大数据开展前体物排放表征的探索方向主要集中在三方面。(1)多源大数据融合方法学建立,进一步提升表征精度。例如TransCAD、Underwood等交通流模型^[105,134]以及多源交通来源数据^[104]融合以获取更高时空分辨率或覆盖度的交通流信息;多卫星火点融合方法提升生物质开放燃烧动态表征的时间分辨率^[135]。(2)交通源排放的近实时或更大尺度的排放表征方法研究。海量的交通流数据处理量导致基于大数据的机动车排放表征研究多集中在城市尺度,为了实现区域尺度甚至更大尺度的机动车排放动态表征,需要进一步提升排放表征方法的效率和可行性。例如,Wu等人^[112]利用机器学习模型,结合历史道路车流量变化谱和近实时路网监测数据,建立了京津冀区域小时的尺度交通源排放清单;Huang等人^[43]运用Apache Spark Streaming技术提高单船排放的表征效率,实现船舶排放的多尺度近实时表征。(3)结合机器学习技术预测污染源短临排放,但目前研究还相对较少,主要集中在机动车和船舶等较容易预测的污染源^[37,136]。例如,Zhang等人^[137]利用小波变换和长短期记忆网络的时间序列模型,结合路网机动车排放监测数据和气象数据预测未来1~3 h的机动车排放等。

2 当前PM_{2.5}和臭氧污染前体物排放源清单存在的问题

2.1 排放因子数据集还存在较大的不确定性

近年来,我国学者建立的本土排放因子以及编制指南或手册中推荐的排放因子已经广泛应用于各尺度前体物排放清单编制。然而,(1)由于缺乏权威的排放测试规范与方法流程,本土建立的排放因子不确定性仍然较大;(2)由于排放清单编制指南或手册的撰写与发布较为仓促,对其推荐的排放因子并没有进行严格的论证与评估,出现部分排放因子偏离实际污染源排放特征、与指南中相同污染源排放因子存在较大差异

等问题; (3) 我国目前缺乏官方权威的、并且经过严格审核论证的排放因子数据集, 这是我国排放清单质量进一步改进的瓶颈。

2.2 尚无规范化的官方PM和VOCs成分谱数据库

尽管我国已经整理建立了一些本土PM和VOCs成分谱数据集, 但这些数据集尚存在一些不足: (1) 本土数据集收录的源成分谱数量不足1000条, 组分数不足600种, 并且不少行业的源成分谱缺失关键行业或关键活性组分, 相较之下, 美国SPECIATE数据库有近6000条成分谱和近3000种组分^[138]; (2) 缺乏统一的数据质控标准和源成分谱建立方法, 导致不同研究建立的源成分谱差异大, 难以融合汇总; (3) 目前, 源成分谱测试工作多集中于长三角、珠三角、京津冀等地区, 考虑到不同地区之间的生产活动水平差异显著, 区域建立的源成分谱能否适用于其他地区, 还未经过评估、讨论; (4) 温度等气象条件对VOCs源成分谱具有较大影响, 我国乃至全球还尚未建立不同气象条件的VOCs源成分谱数据集。

2.3 前体物排放源清单编制质量评估方法缺失

排放清单的校验与评估的不足主要体现在: (1) 当前的前体物排放清单校验多关注于国家和区域尺度的基准年排放清单校验, 而对于城市尺度排放清单以及趋势排放清单的校验与评估研究还很少; (2) 不确定性分析是评估排放清单的重要方法, 也是排放清单建立的必要环节, 但我国尚未编制适合前体物排放清单特征的业务化不确定性分析指南, 以规范化前体物排放清单的不确定性分析的流程与方法; (3) 缺乏全面评估排放清单编制质量的方法体系。目前, 排放清单的校验和评估方法都是从单一角度对排放清单的合理性或可靠性进行评估, 但忽略了从应用角度对排放清单的精细程度和规范性进行评估; (4) NH_3 、VOCs及重要组分以及Cl、HONO等高活性污染物的排放表征仍存在较大不确定性, 其校验评估方法也较为薄弱。

2.4 类型单一的排放清单不足以支撑 $\text{PM}_{2.5}$ 和臭氧污染精细化管理

我国有必要构建针对不同应用场景或类型的前体物清单, 以支撑不同尺度 $\text{PM}_{2.5}$ 和臭氧精细化日常与应急管控需求。根据覆盖的空间尺度分类, 排放清单可分为国家、区域、城市、区县、企业/工艺过程等排放清

单; 根据时间尺度可分为基准年排放清单、历史趋势排放清单、未来预测排放清单等; 根据用途差异可分为模式输入清单、应急管控清单、规划情景清单、减排潜力清单等。然而, 我国目前的前体物清单研究大多数为国家、区域和部分城市尺度的基准年清单, 基于过程环节的企业级精细化排放清单和减排潜力清单、应急管控清单较为匮乏, 用于各种规划的未来预测清单和情景清单规范化建立的方法学还较欠缺等。例如, 在城市大气污染防治实践中, 通常根据城市基准年排放清单识别排放较大的企业作为管控对象。但在要求企业落实减排、应急等任务时, 不少企业不清楚排放产生在哪些环节, 应该如何减排, 措施实施是否可以完成减排任务等。

2.5 前体物排放清单的时效性难以支撑污染过程精细化管理

$\text{PM}_{2.5}$ 和臭氧污染精细化管理依赖于对污染过程的精准预报、污染成因的精深分析与管控方案的精细制定。这就要求前体物排放清单的时效性能够准确反映污染源的动态排放特征。然而, 传统以年为单位的清单动态更新机制难以及时表征污染源的近期排放变化, 导致污染预报、成因分析和管控方案制订等可能存在偏差。例如, 由于新型冠状病毒肺炎疫情的影响, 2020年2月全国的 SO_2 、 NO_x 和VOCs的排放比2019年同期分别下降了29%、31%和26%, 而全国各地预报系统采用的排放清单无法及时更新, 导致该时段的预报容易发生偏差^[139]。城市不利气象条件应对根据基准排放清单确定重点管控企业名单和减排方案, 但由于基准排放清单通常滞后1~2年, 部分重点企业可能已经迁移到其他地区或者完成技术改造, 导致排放清单的结果严重偏离实际情况。为了更好地支撑污染过程应急和精细化管理, 需要发展更为高效的前体物排放清单动态更新方法, 提升前体物排放清单的时效性。

3 前体物排放清单编制质量评估

随着我国 $\text{PM}_{2.5}$ 和臭氧污染协同防控的不断推进, 前体物排放清单编制已经陆续成为各城市大气环境管理部门的日常工作。建立业务化的排放清单编制工作机制和流程能够保证前体物排放清单编制的制度化、程序化、规范化^[78]。然而, 受认知和技术水平以及数据质量差异的影响, 不同科研和管理人员建立的前体物排放清单的质量会存在差异。如何对前体物排放清单

编制质量进行评估是规范化排放清单编制、提高排放清单质量、推动排放清单业务化应用的重要前提,也是当前管理部门使用排放清单的需求。目前,国外仅有美国国家环境保护局在2016年发布了《生命周期排放清单数据质量评估指南》^[140]以及少数研究初步探讨了排放清单质量评估的内容^[141],国内外还尚未建成立体系的排放清单质量评估方法。为此,本文借鉴排放清单半定量不确定性分析和层次分析法,提出了我国开展前体物排放清单编制的质量评估思路(图6)。排放清单质量评估方法的建立需要经过大量的审核、讨论和优化。本文只是作初步探索,具体方法和案例参见本课题组2022年出版的《排放源清单与大气化学传输模型的不确定性分析》一书。

前体物排放清单的质量体现为多个方面,不同类型的排放清单质量均可从基础数据质量、清单精细程度、清单结果合理性和排放清单编制规范性这4个维度进行评估。(1)基础数据质量决定了排放清单的精细程度、不确定性以及表征结果的合理性,是影响排放清单质量的决定性因素。基础数据质量分析重点在评估数据来源的权威性、可靠性、完整性以及在时空和技术层面上的代表性;(2)精细程度量化是评估排放清单是否满足应用的需求,其重点评估内容包括源分类和污染物的精细程度、表征方法适合度、时空分辨率、点源化率、时效性和动态更新频率等评估;(3)排放清单合理性是指排放清单对实际排放情况的反映程度,需要结合多种清单校验手段从排放总量、时空特征、源结构和不确定性分析等角度进行评估;(4)排放清单编制的规范性是保证排放清单质量的基础,重点

评估内容包括排放清单编制流程是否完整、排放清单编制过程中涉及的资料是否存档且可追溯、清单报告内容是否记录完整等。

考虑到排放清单类型和应用场景多样、编制工作涉及多方面的数据和内容,排放清单质量评估方法必须具备完整性、客观性、可行性和灵活性的特点。完整性要求评估指标能较为全面地体现排放清单质量的多个维度;客观性要求评估过程尽可能降低人为主观因素,选取的评估指标和评估标准容易量化或分级;可行性是指在众多影响排放清单编制质量的因素中选取具有代表性、容易分级的评估指标对排放清单质量进行综合评估;灵活性要求评估方法能够胜任不同类型和不同应用需求的排放清单质量评估。因此,如何定义各维度的评估指标、量化评估指标得分以及综合分析,是开展排放清单质量评估的关键。

为了保证质量评估方法的可行性和完整性,需要采用具有代表性、容易评级的影响因素作为评估指标对排放清单质量的各项维度进行综合评估。例如,精细程度可以采用源分类的精细程度、污染物的覆盖面、表征方法的适合度、时空分辨率、点源化率、时效性和动态更新频率作为评估指标。这些指标相对容易量化和划分评级。另外,为了提高各项维度评估的量化程度,评估指标可以是多层次的。例如,基础数据来源质量的一级评估指标可考虑活动水平、排放因子及相关参数、时空分配因子和成分谱,二级评估指标可采用数据来源、数据完整性、地域代表性、年份代表性、技术代表性等。

为了保证质量评估的客观性,可借鉴排放清单半定量不确定性分析方法,需要对每个维度的评估指标建立起相对应的评分矩阵。根据评分矩阵,采用专家判断方法对各个维度每一层级评估指标进行打分评定。评分矩阵的建立可参考数据质量指数DQI、评分谱系NUSAP等半定量不确定性分析方法^[103,142]。

最后,为了保证质量评估的灵活性,可采用层次分析法,由下到上逐层计算各层次评估指标的得分,形成排放清单质量的综合评分。层次分析法是由Saaty^[143]提出的一种定量与定性相结合的多目标决策分析与评估方法,其最主要的特点是能根据不同应用场景对排放清单质量要求,通过两两对比同一层次的所有评估指标,建立各层级评估指标的判断矩阵,赋予各层级评估指标的权重系数,使质量评估方法能够根据应用场景进行差异化计算。此外,层次分析方法也能根据评

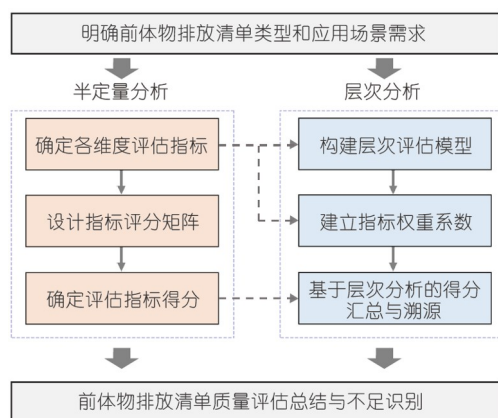


图6 (网络版彩色)排放清单编制质量评估思路

Figure 6 (Color online) The framework for the quality assessment of emission inventories

估指标的层次关系, 识别影响最终质量评分的关键指标.

4 展望

前体物排放清单是研究PM_{2.5}和臭氧污染成因、制定PM_{2.5}与臭氧污染协同防控策略的重要基础数据. 近年来, 我国PM_{2.5}和臭氧污染前体物排放清单在方法学研究清单编制实践方面都有迅速发展. 为了进一步提升前体物排放清单质量, 更好地支撑不同尺度的PM_{2.5}和臭氧污染协同防控, 本研究认为我国PM_{2.5}和臭氧污染前体物排放清单的未来工作应该在以下四方面加强: (1) 为了降低前体物排放清单的不确定性, 建议我国尽早建立经过科学评估论证的、规范、开放、统

一的排放因子数据库与成分谱数据库; (2) 建立不同类型的前体物排放清单, 并根据前体物清单的作用、内容、特点、数据需求等要求, 建立不同类型前体物排放清单的质量评估和不确定性分析规范与流程, 编制发布相关指南, 提高排放清单质量; (3) 开展近实时排放清单和短临排放预测清单研究, 探索利用大数据、卫星和地基观测数据等近实时和短临预测清单构建的方法学, 提升前体物排放清单的时效性, 支撑短期污染应急管控和空气质量预报; (4) 发展基于大数据建立的排放清单不确定性分析和质量评估方法, 尽早建立相关标准和指南, 推动我国建立高质量、满足PM_{2.5}和臭氧污染精细防控需求的前体物近实时排放清单.

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图4文献列表和表1统计采用的文献列表等

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Summary for “我国PM_{2.5}和臭氧污染前体物排放源清单的现状与质量评估”

Status and quality evaluation of precursor emission inventories for PM_{2.5} and ozone in China

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Developing precursor emission inventories is fundamental to deepen our understanding of air pollution and formulate refined co-control strategies for PM_{2.5} and ozone. Thanks to numerous research projects and the strong support by local governments, precursor emissions for PM_{2.5} and ozone have been rapidly advanced in China over the past two decades, achieving the following key progresses: (1) Feasible methodologies and guidelines or manuals for precursor emission developments have been established. These include nine technical guidelines for the development of emission inventories of air pollutant sources for PM₁₀, PM_{2.5}, volatile organic compounds (VOCs), and NH₃ emissions as well as on-road mobile sources, non-road mobile sources, biomass combustion sources, particulate matter (PM) from dust fugitive sources, and residential coal combustion sources published in 2014–2016; the technical manual for *Urban Air Pollutant Emission Inventory Development* and workbook for *the Study on Air Pollution Prevention and Control in “2+26” Cities* released in 2017. (2) Comprehensive datasets of local emission factors and speciated profiles of PM and VOCs have been established to cover major anthropogenic emission sources and reduce the uncertainty in emission estimation. In recent years, emission factors for isoprene, nitrous acid (HONO), chlorine (Cl), and other key precursor species of PM_{2.5} and ozone formation have also been developed. The most recent VOC profile datasets contain 400 to 500 VOC species from nearly 100 different sources. The less-studied oxygenated VOC (OVOC) species were also included. (3) With the progress in local emission factors, speciated profiles, and activity data, emission sources and species involved in emission inventories are more comprehensive and inclusive. For instance, the number of VOC species has increased by about 140 in China and by about 280 in the Pearl River Delta (PRD) region. The inclusion of more VOC species boosts the ozone formation potential in PRD by 30%. Emissions of semi-volatile and intermediate VOCs (S/IVOCs), Cl and HONO have also been developed. (4) A large number of precursor emissions at national, regional/provincial and city scales have been established to meet the requirements of PM_{2.5} and ozone control in different levels. These regional or provincial emissions mainly focused in East China (30%), South China (24%), and North China (13%), whereas city-level emissions were concentrated in North China (32%), East China (30%), and Southwest China (10%). (5) To improve the spatial and temporal accuracy and timeliness, big data have been applied to improve the anthropogenic emission inventories from power plants, industrial sources, motor vehicles, dust sources, biomass open burning, ship emissions, etc. in China. (6) Novel techniques, including source apportionments based on isotope analysis and emission inversion, have been applied to calibrate emissions. Overall, the temporal-spatial resolution, timeliness, and reliability of precursor emissions for PM_{2.5} and ozone in China have been significantly improved in recent years. Nonetheless, uncertainty in precursor emission inventories remain significant, particularly for emissions of speciated VOCs, organic carbon (OC), and black carbon (BC), as well as dust fugitive sources, biomass burning, industrial combustion sources, and solvent-use sources, whose upper limits of uncertainty range typically exceed 100%. Meanwhile, official emission factors and speciated profiles are unavailable, and the capacity of speciated emission validation is not well developed. Methods for assessing inventory quality remain largely vacuum. Future research should focus on expanding datasets for standardized emission factors and speciated profiles, investigating methods and compiling guidebooks for emission validation and quality assessment, developing and commercializing near real-time and short-term prediction inventories, and establishing and evaluating multi-type precursor emission inventories. This study proposes a conceptual framework to assess the quality of precursor emission inventories, which could serve as a reference to assess the quality of precursor emission inventories for inventory users and help standardize the development of precursor emission inventories.

PM_{2.5} and ozone, precursor emissions, speciated VOCs (volatile organic compounds) emission inventories, validation and evaluation, uncertainty analysis

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