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长三角区域人为源活性挥发性有机物高分辨率排放清单

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摘要: 基于长三角区域41个城市本地实测,结合美国EPA的SPECIATE 4.4数据库,建立了长三角区域人为源活性挥发性有机物(VOCs)高分辨率排放清单,分析了区域内VOCs的排放特征和组分构成; 计算了VOCs的臭氧生成潜势(OFP)和二次有机气溶胶生成潜势(SOAP). 结果表明,2017年,长三角区域人为源VOCs排放总量为 4.9×10^6 t,其中工艺过程源、工业溶剂使用源、移动源、生活源、储运源、农业源和废弃物处理源排放贡献分别为: 34.3%、27.1%、19.5%、9.7%、6.1%、2.5%和0.4%. 芳香烃和烷烃是VOCs的主要种类,均各占长三角VOCs排放总量的25%. 工艺过程源、工业溶剂使用源、移动源和生活源OFP贡献率分别为38.3%、21.5%、16.4%和13.2%,SOAP贡献率分别为26.2%、34.1%、18.1%和17.9%,与VOCs排放量的主要贡献源基本一致. 各城市VOCs重点排放行业存在较大差异,重点城市群以石化化工和装备制造为主,区域北部则以木材家具等涂装行业为主. 计算表明,丙烯、间/对-二甲苯和乙烯是臭氧主要贡献源; 甲苯、1,2,4-三甲苯和间/对-二甲苯是二次有机气溶胶主要贡献源. 下阶段VOCs的精细化治理可向基于化学反应活性的主控行业转变,可将化工、石化、汽车制造、纺织和木材家具等关键行业治理放在优先位置,同时根据城市特征,制定差异化的VOCs治理路径.

关键词: 挥发性有机物(VOCs); 活性组分; 排放清单; 大气污染; 长三角区域

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High-resolution Emission Inventory of Reactive Volatile Organic Compounds from Anthropogenic Sources in the Yangtze River Delta Region

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Abstract: A high-resolution emission inventory of anthropogenic active volatile organic compounds (VOCs) for the Yangtze River Delta (YRD) region was developed based on the local measurement of 41 cities in the region and the specific 4.4 database of EPA. The emission characteristics and composition of VOCs were analyzed. The ozone formation potential (OFP) and secondary organic aerosol formation potential (SOAP) of VOCs were calculated. The results showed that the total emission of anthropogenic VOCs in the YRD in 2017 was 4.9×10^6 t. The emission contributions of process sources, industrial solvent sources, mobile sources, domestic sources, storage and transportation sources, agricultural sources, and waste treatment sources were 34.3%, 27.1%, 19.5%, 9.7%, 6.1%, 2.5%, and 0.4%, respectively. Aromatic hydrocarbons and alkanes were the main components of VOCs, accounting for 25% of the total VOCs emissions in the region. The contribution rates of OFP from process sources, industrial solvent sources, mobile sources, and domestic sources were 38.3%, 21.5%, 16.4%, and 13.2%, respectively, and the contribution rates of SOAP were 26.2%, 34.1%, 18.1%, and 17.9%, respectively, which was basically consistent with the main contribution sources of VOCs emissions. The emission characteristics of the key industries in each city were obviously different. The key urban agglomeration of VOCs emission was mainly petrochemical industries and equipment manufacturing, whereas the northern part of the region was mainly wood furniture and other coating industries. The results showed that propylene, *m*-xylene, *p*-xylene, and ethylene were the main contribution sources of ozone, whereas toluene, 1,2,4-trimethylene, *m*-xylene, and *p*-xylene were the main contribution sources of secondary organic aerosols. In the next stage, the fine management of VOCs can be transformed into the main industries based on chemical reaction activity, which can give priority to the governance of key industries such as the chemical industry, petrochemical, automobile manufacturing, textile, wood, and furniture and can formulate different governance paths according to urban characteristics.

Key words: volatile organic compounds (VOCs); reactive compounds; emission inventory; air pollution; the Yangtze River Delta region

挥发性有机物(volatile organic compounds, VOCs)是近地面臭氧(O_3)和二次有机气溶胶(SOA)生成的重要前体物[1~4]. VOCs化学组分复杂,不同来源和组分的VOCs组分反应活性差异较大,对O_3和SOA生成的潜势也不尽相同[5~7]. 因此,建立基于化学反应活性的人为源VOCs精细化排放清单是有效制定$PM_{2.5}$和O_3污染协同控制措施的重要基础,对摸清各城市各行业排放现状、满足空气质量模型模拟需求、细化实施减排政策等具有重要的指导意义. 近年来,大量研究已在全国和区域尺度建立了VOCs排放清单. 全国尺度方面,清华大学构建的多尺度排放清单模型(multi-resolution emission inventory for China, MEIC)建立了1990~2017年人为源VOCs分组分排放清单,并取得了广泛应用[8]; 北京大学谢绍东团队也建立了详细的

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VOCs 组分清单,并评估了其对 O_3 和 SOA 生成的潜在贡献[9,10]. 此外,各重点区域和省份也先后利用本地化污染源数据建立了高分辨率的 VOCs 排放清单[11~16],源类和空间分辨率的精细度得到了大幅提升. 为了进一步摸清各类人为源 VOCs 排放的成分谱特征,各区域通过本地化实测结合国内外参考源谱相继形成了各源类 VOCs 成分谱库[7,17~20]. 将高分辨率清单与详细源类成分谱资料相结合,是科学评估区域活性 VOCs 组分来源及其对 O_3 和 SOA 生成潜在贡献的重要手段.

长三角是我国经济活动最为频繁和工业体量最大的区域之一,且区域行业门类十分齐全,VOCs 排放强度位居全国前列[21]. 为掌握区域大气污染物排放特征,Huang 等[22]、Fu 等[23]和 An 等[24]先后基于本地污染源调查建立并更新了长三角区域高分辨率人为源排放清单,并获得了 VOCs 分组分排放特征. 在此期间,Zhao 等[25]通过实测不断丰富并改进了江苏等省市的 VOCs 分组分排放清单[26,27],有效地提升了 O_3 的模拟效果. 但是,针对长三角具体城市和详细源类的活性 VOCs 组分排放特征研究及其对 O_3 和 SOA 贡献的综合评估仍相对较少. 因此,本研究在建立长三角区域高分辨率人为源 VOCs 排放清单的基础上,结合以本地化实测为主的 VOCs 成分谱数据库,以 2017 年为基准年,建立精细化的基于化学反应活性的人为源活性 VOCs 组分排放清单,旨在为有效制定 $PM_{2.5}$ 和 O_3 污染协同控制措施提供科学支撑.

1　材料与方法

1.1　人为源 VOCs 排放清单构建

长三角地区包括了江苏、浙江、安徽和上海三省一市共 41 个城市. 研究区域西起 118.25°E,东至 122.42°E,南抵 28.90°N,北达 33.30°N,面积约 350 400 km^2. 本研究将研究区域划分为11 979个 4 km×4 km 的网格.

VOCs 排放源类分为固定燃烧源、移动源、储运源、生活源、农业源和废弃物处理源,以及大部分工业过程和溶剂使用来源,如石油炼制、焦炭生产、化工制造、纺织、家具制造、包装印刷、汽车制造、造船和建筑涂料等,基本覆盖涉 VOCs 排放的所有人为源.

VOCs 排放估算方法主要采用排放因子法,溶剂使用源则采用物料平衡法. 活动水平数据主要来自 2017 年长三角工业源调查结果及各城市统计年鉴,其中,工业源包括了超过 3 万家企业(如固定燃烧源、工业过程和溶剂使用源,对于点源调查不够全面的行业以最小行政单元做面源补充),用于计算的数据包括能源消费量、工业产品产量和溶剂使用量等调查结果,以及本地化调研获得的各行业 VOCs 治理技术及其效率等信息. 具体的计算方法可以参见文献[24].

1.2　VOCs 成分谱数据

本研究的 VOCs 成分谱数据主要来自长三角本地实测,包括汽油车和柴油车[28,29]、船舶[30,31]、油气挥发[32]、餐饮[33]、生物质燃烧[34]和大部分的工艺过程源和溶剂使用源[32,35,36],如石化、炼焦、化工、纺织、家具制造、包装印刷、汽车制造、船舶制造和建筑涂装等. 对于无本地化实测结果的源类以及已有研究未识别的 VOCs 物种,采用美国 EPA 的 SPECIATE 4.4 数据库[37]进行了补充. 最终本研究共确定 108 个涉 VOCs 排放源类的 424 种组分,含烷烃、烯烃、芳香烃、卤代烷烃和含氧有机物(OVOCs)等. 图 1 所示为长三角区域主要源类的 VOCs 物种大类构成.

1.3　活性 VOCs 排放估算

根据 VOCs 基础排放清单,结合各源类 VOCs 成分谱,可计算获得 VOCs 分物种排放清单,计算方法如式(1)所示[9].

$$E_i = \sum_j (E_j \times f_{ij}) \tag{1}$$

式中, E_i 为 i 物种的排放总量,t; E_j 为 j 类污染源的排放总量,t; f_{ij}为 i 物种在 j 类污染源的 VOCs 总排放量中所占的质量分数,无量纲.

为进一步评估各类污染源 VOCs 排放对 O_3 和 SOA 生成的潜势贡献,本研究分别利用各类 VOCs 物种的最大增量反应活性因子(MIR)和 SOA 产率(Y)计算臭氧生成潜势(OFP)和 SOA 生成潜势(SOAP),具体如式(2)和式(3)所示.

$$\mathrm{OFP}_j = \sum_i (E_{ij} \times \mathrm{MIR}_i) \tag{2}$$

$$\mathrm{SOAP}_j = \sum_i (E_{ij} \times Y_i) \tag{3}$$

式中,OFP_j 为 j 类污染源的臭氧生成潜势,t(以 O_3 计); SOAP_j 为 j 类污染源的 SOA 生成潜势,t(以 SOA 计); E_{ij}为 j 类污染源中 i 物种的排放总量, t; MIR_i 为 i 物种的最大增量反应活性因子(O_3/VOCs), $t \cdot t^{-1}$; Y_i 为 i 物种的 SOA 产率,%.

1.4　不确定性分析

根据区域各源类活性 VOCs 活动水平数据和排放因子,可计算各排放源活性 VOCs 排放水平的不确定性,计算方法如式(4)和式(5)所示[22].

$$\mathrm{CV} = \frac{U}{E} = 1.96 \times \sqrt{(1 + C_{\mathrm{a}}^2) \times (1 + C_{\mathrm{f}}^2) - 1} \tag{4}$$

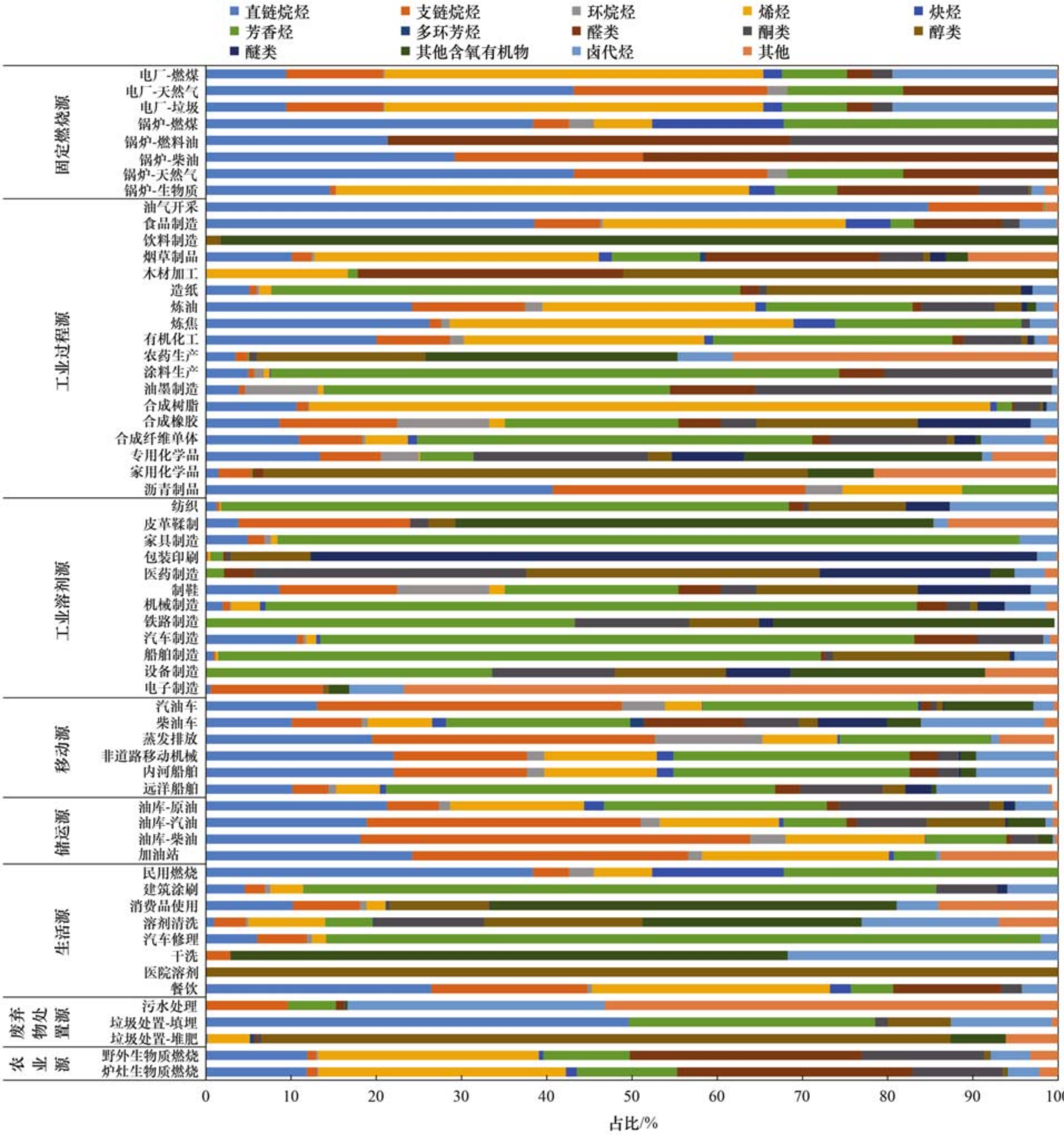

图 1 长三角区域主要源类 VOCs 物种大类构成

Fig. 1 VOC species in major categories from the main sources in the Yangtze River Delta region

$$U = \sqrt{\sum_j \sum_k U_{j,k}^2} \tag{5}$$

式中,CV 为变异系数; U 为各排放源不确定性; E 为排放率; C_a 为活动水平数据的不确定性, C_f 为排放因子的不确定性; j 与 k 分别为物质类别和排放源类,均无量纲.

2 结果与讨论

2.1 长三角区域人为源 VOCs 排放特征

2017 年长三角区域人为源 VOCs 排放总量为 487.5 万$t\cdot a^{-1}$,排放分担率如图 2 所示. 工业源(含工业过程源和工业溶剂源)共计 299.2 万$t\cdot a^{-1}$,是长三角 VOCs 主要排放来源,占区域 VOCs 总排放量的 60% 左右. 其中,工业过程源和工业溶剂源分别占 34.3% 和 27.1%. 工业过程源中的主要排放行业包括化工、橡塑、石化和非金属制品等,工业溶剂源中的主要行业包括木材家具、应用设备、设备制造、电子制造、医药和纺织等. 石化和化工行业是该地区重工业的支柱产业,其产量与产值均居全国前列,VOCs 排放分担率分别达到 3.2% 和 16.4%. 而该地区高速发展的房地产业,也拉大了木材家具行业的需求,因此其在工业溶剂源中 VOCs 排放占比最高. 移动源、生活源、储运源、农业源和废弃物处理源的 VOCs 排放占比依次为 19.5%、9.7%、6.1%、2.5% 和 0.4%. 移动源、储运源、生活源和

农业等源类排放均较为集中，分别以机动车排放、储油库排放、民用溶剂排放和家用秸秆燃烧排放为主.

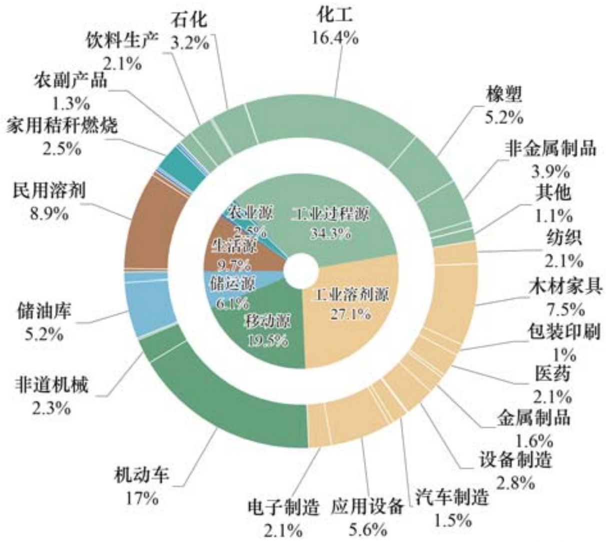

图 2　2017 年长三角区域各类 VOC 人为源的排放分担率

Fig. 2　VOC emission contributions from each source in the Yangtze River Delta region for the year 2017

2.2　主要源类 VOCs 组分构成

图 3 所示为长三角主要源类的 VOCs 化学组分构成. 芳香烃和烷烃是长三角区域 VOCs 排放的主要物种，均占 VOCs 排放总量的 25%. OVOCs 在 VOCs 排放中的占比也达到了 21.9%，其中醛类、酮类、醇类和酯类分别占 5.0%、4.4%、9.0% 和 3.5%. 卤代烃排放占比较小，仅为 3.1%. 机动车排放是该区域烷烃(包括直链烷烃、支链烷烃和环烷

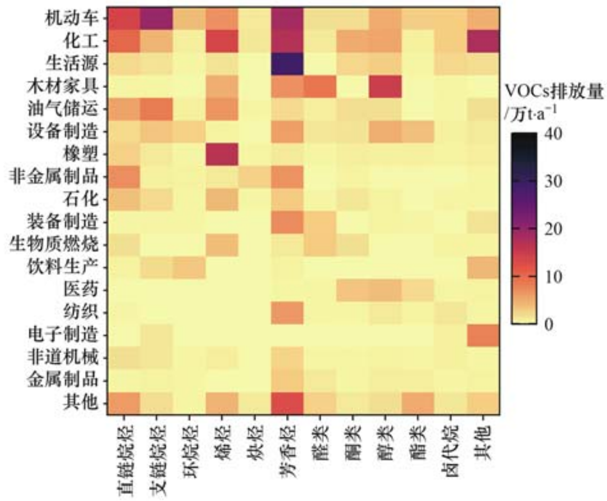

图 3　长三角区域主要源类的 VOCs 大类组分排放分布

Fig. 3　Emission distribution of VOC species in major categories from the main sources in the Yangtze River Delta region

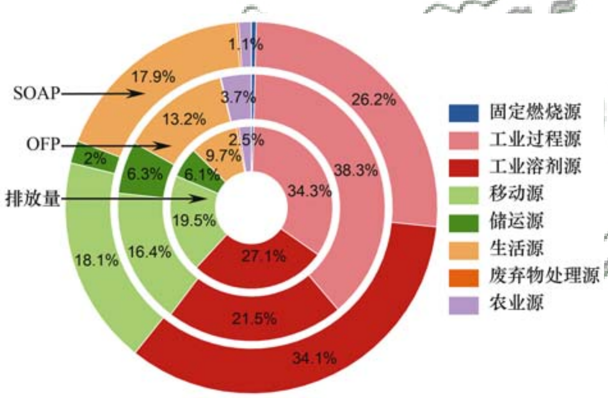

图 4　长三角区域基于排放量、OFP 和 SOAP 的人为源贡献占比

Fig. 4　VOC emissions, their OFP, and SOA contributions from different sources in the Yangtze River Delta region

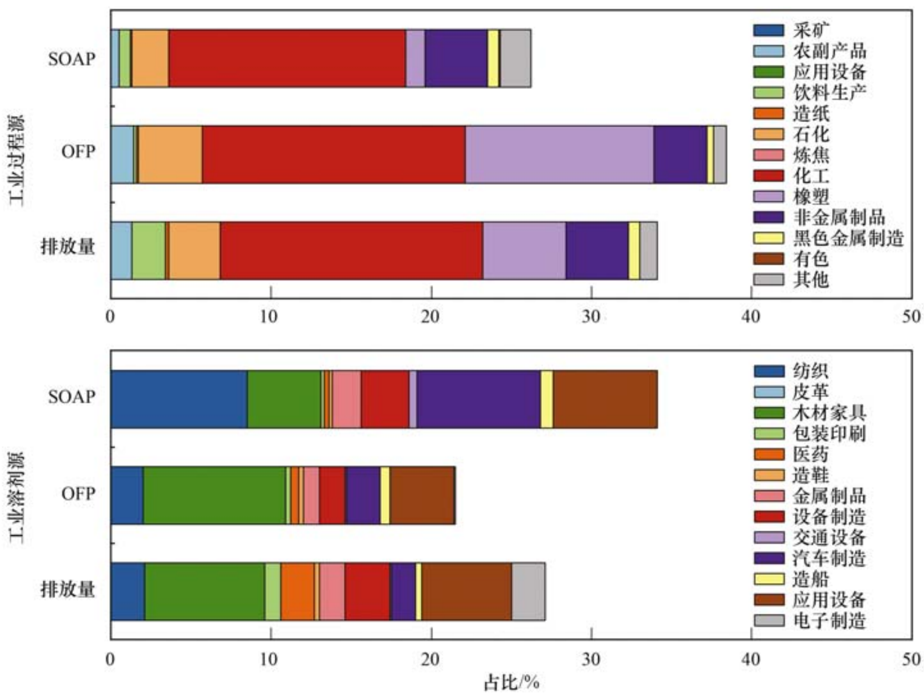

图 5　长三角区域关键工业行业对 VOCs 排放、OFP 和 SOAP 的贡献

Fig. 5　VOC emissions, their OFP, and SOA contributions from key industrial sectors in the Yangtze River Delta region

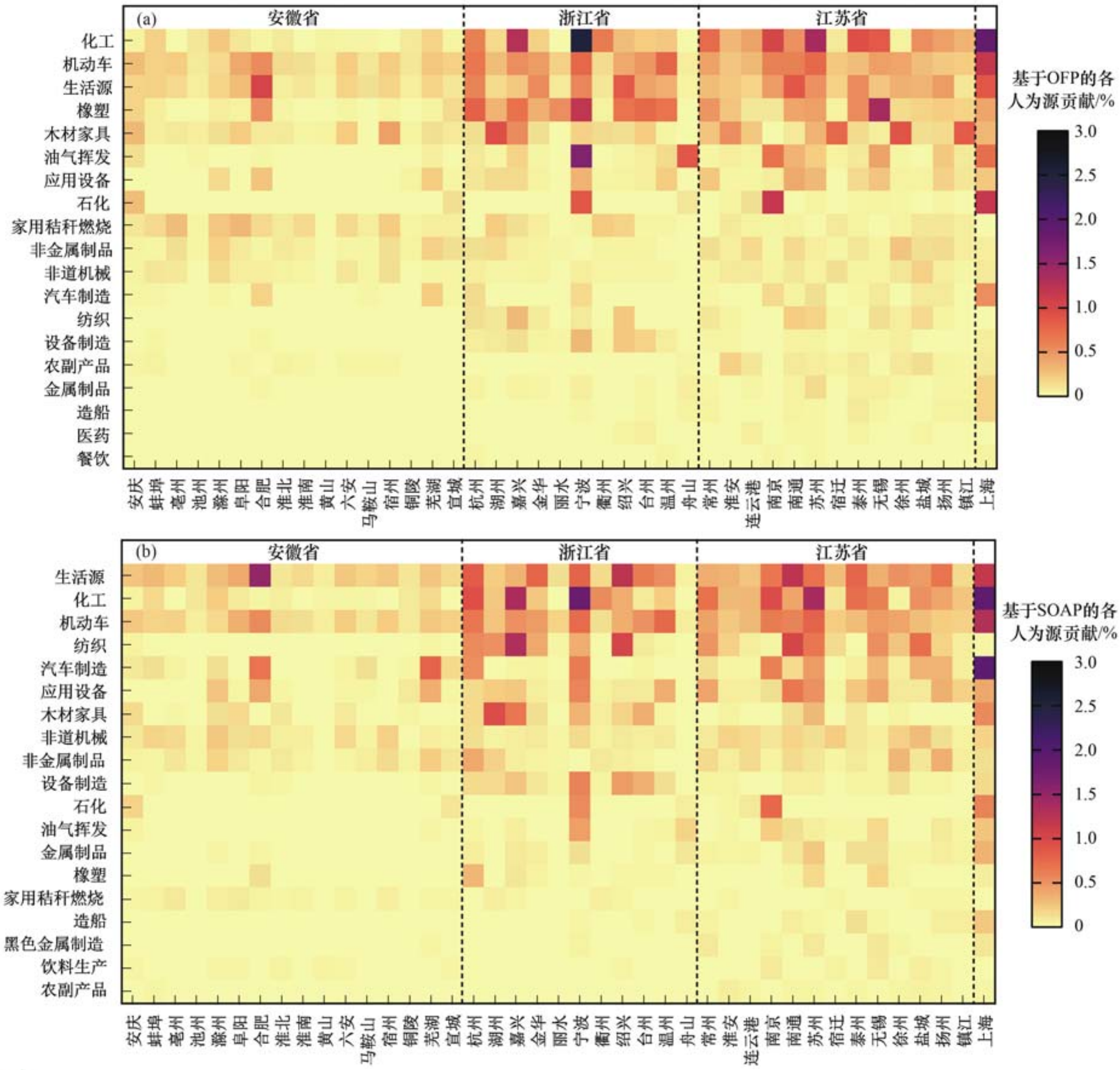

图 6 长三角各城市排放源对 OFP 和 SOAP 生成潜势贡献

Fig. 6 Contributions of VOC emissions from different sources in each city to their OFP and SOA formation potentials

烃)和芳香烃的主要来源,分别占各物种排放总量的 31.2% 和 15.1%. 化工行业则以烯烃和芳香烃排放为主. 此外,橡塑行业烯烃排放也相对突出. 生活源的芳香烃排放明显高于其他源类,占芳香烃排放总量的 23.7%. 相比于生活源,工业溶剂源门类较多,芳香烃和 OVOCs 排放占比均相对较高,分别占各自总排放量的 29.3% 和 33.3%,尤其是木材家具行业涉及醛、醇类 OVOCs 排放占比最高.

2.3 各源类 VOCs 排放及其 O_3 与 SOA 生成潜势比较

图 4 所示为基于排放量、OFP 和 SOAP 的长三角主要人为源排放占比比较. 可见,基于 OFP 和 SOAP 识别的人为源排放贡献与其原始排放占比接近,工业溶剂源、工业过程源、移动源和生活源均为贡献最高的四大源类. 从 OFP 贡献来看,工业溶剂源和移动源所占的贡献略低于其排放占比,工业过程源和生活源的 OFP 贡献则高于其排放占比; 从 SOAP 贡献来看,工业过程源所占的贡献较其排放占比下降了 20%,工业溶剂源的 SOAP 贡献则超过工业过程源,成为 SOAP 贡献最高的源类. 图 5 进一步给出了长三角地区工业过程源和工业溶剂源分行业的 OFP 和 SOAP 排放贡献. 化工行业是对工业过程源 OFP 和 SOAP 贡献最大的部门,分别占 16.4% 和 14.8%; 其次为橡塑行业,其大量的烯烃排放贡献了 11.8% 的 OFP. 汽车制造和纺织行业是对工业溶剂源 SOAP 贡献最大的部门,分别 7.7% 和 8.5%; 木材家具则贡献了工业溶剂源约 8.9% 的 OFP 潜势.

2.4 各城市分源类 VOCs 排放对 O_3 与 SOA 生成潜势的贡献

图 6 进一步列出了长三角各城市中对 OFP 和 SOAP 潜势贡献最高的前 19 个二级源类. 从中可

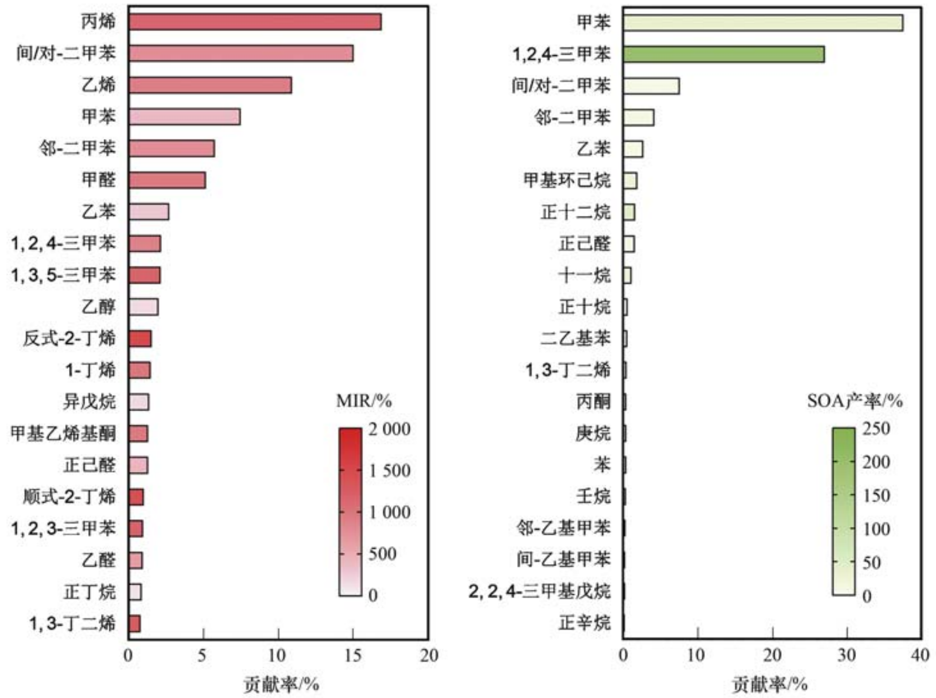

图 7　基于 OFP 和 SOAP 贡献的长三角区域关键人为源 VOCs 组分

Fig. 7　Key anthropogenic VOC species in the Yangtze River Delta region based on their OFP and SOAP contributions

知，宁波和上海的化工行业是在长三角分城市和分源类中的 OFP 贡献最高，分别占长三角人为源 OFP 的 2.5% 和 1.9%. 其次为无锡的橡塑行业和苏州的化工行业，均贡献了区域 OFP 的 1.4%. 上海和南京的石化行业、宁波的橡塑行业、合肥的生活源和上海的机动车对区域 OFP 的贡献均大于 1.0%. 按城市划分的排放源中，对 SOFP 贡献最高的 5 个源类分别为：上海的汽车制造行业(2.0%) > 上海的化工行业(1.9%) > 宁波的化工行业(1.8%) > 合肥的生活源(1.5%)；其次为上海、南通和绍兴的生活源，嘉兴、绍兴和南通的纺织行业也超过了 1.0%.

2.5　区域主要 O_3 和 SOA 生成潜势物种排序

图 7 所示为对长三角区域 OFP 和 SOAP 贡献最高的前 20 种 VOCs 组分. 对 OFP 贡献最高的物种依次为丙烯(16.8%)、间/对-二甲苯(15.0%)、乙烯(10.9%)、甲苯(7.5%)和邻-二甲苯(5.7%)等，丙烯的主要排放行业依次为橡塑、化工和炼油，间/对-二甲苯、邻-二甲苯为民用溶剂、机动车和化工，乙烯为化工、橡塑和油气挥发，甲苯为纺织、化工和机动车. 甲苯、1,2,4-三甲苯、间/对-二甲苯、邻-二甲苯和乙苯是对 SOAP 贡献最高的物种，分别贡献了人为源 SOAP 的 37.5%、27.0%、7.6%、4.2% 和 2.7%，其中，甲苯 SOAP 贡献较高的主要行业依次为纺织、化工和机动车，1,2,4-三甲苯为化工、家具制造和炼油，间/对-二甲苯、邻-二甲苯和乙苯为民用溶剂、机动车和化工. 由此可见，上述组分和对应的主要行业是控制长三角区域 O_3 和 SOA 污染的关键.

2.6　不确定性评估

基于本地排放因子和区域活动数据进行不确定性分析，评估表明，长三角区域 VOCs 排放的平均不确定性为 −44% ~ 68%，电厂、锅炉、炼油、化工制造、黑色金属制造、有色金属制造、机动车和非道路移动机械排放不确定性分别为 −28% ~ 22%、−19% ~ 23%、−40% ~ 57%、−71% ~ 167%、−41% ~ 61%、−44% ~ 70%、−46% ~ 69%、−50% ~ 86%. 其中，电厂和锅炉等固定燃烧源的排放基于详细的活动数据和当地实测，不确定更低；黑色金属制造和有色金属制造等工业行业因为对各工艺环节相对准确的排放估算，不确定性相对较低，但化工行业仍存在大量未分类的工艺流程，排放仍存在较大的不确定性；移动源的不确定性则主要来源于活动水平的变化.

3　结论

(1) 长三角区域人为源 VOCs 排放主要来自工业过程源和工业溶剂源，共占区域排放总量的 60% 以上. 烷烃(25%)、芳香烃(25%)和 OVOCs(22%)是长三角区域人为源 VOCs 排放的主要组分.

(2) 基于 OFP 和 SOAP 贡献识别的主要人为源

与 VOCs 主要排放贡献源接近,工业溶剂源、工业过程源、移动源和生活源均为贡献最高的四大来源.

(3)长三角城市 VOCs 重点排放行业存在较大差异,长三角东部重点城市群以石化化工和装备制造为主,区域北部城市则以木材家具等涂装行业为主,下阶段 VOCs 治理需根据城市特征,制定差异化治理目标和政策措施.

(4)对长三角区域 OFP 贡献占比最高的组分依次为丙烯、间/对-二甲苯、乙烯、甲苯和邻-二甲苯,对区域 SOAP 贡献最高的组分依次为甲苯、1,2,4-三甲苯和间/对-二甲苯、邻-二甲苯和乙苯. 综合来看,化工、橡塑、机动车等行业对区域 OFP 和 SOAP 的贡献相对较高. 基于本研究获得的各城市详细源类活性 VOCs 组分 OFP 和 SOAP 贡献,可细化区域 O_3 和 SOA 污染防治的主控行业,推动下一阶段 VOCs 精细化治理向基于化学反应活性的主控行业转变.

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