

(HUANJING KEXUE)

ENVIRONMENTAL SCIENCE

第38卷 第5期

Vol.38 No.5

2017

中国科学院生态环境研究中心 主办



新 能 静 尊 (HUANJING KEXUE)

ENVIRONMENTAL SCIENCE

第38卷 第5期 2017年5月15日

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南京工业区夏冬季节二次有机气溶胶浓度估算及来源解析

刘静达1,安俊琳1*,张玉欣1,师远哲1,林旭2

(1. 南京信息工程大学, 气象灾害教育部重点实验室, 气候与环境变化国际合作联合实验室, 气象灾害预报预警与评估协同创新中心, 中国气象局气溶胶与云降水重点开放实验室, 南京 210044; 2. 杭州市环境监测中心站, 杭州 310007)

摘要: 采用 2015 年 6 月 15 日 ~ 7 月 15 日及 2015 年 12 月 16 日 ~ 2016 年 1 月 15 日期间 GC5000 在线气相色谱仪得到的挥发性有机物(volatile organic compounds,VOCs)数据、DRI-2001A 热/光碳分析仪对膜采样分析得到的 EC(elemental carbon)、OC (organic carbon)数据,使用气溶胶生成系数法(fractional aerosol coefficient,FAC)、EC 示踪法及正矩阵因子分析(positive matrix factorization,PMF)对南京工业区二次有机气溶胶(secondary organic aerosol,SOA)浓度进行估算及来源解析。研究发现南京工业区 SOA 污染主要来源于芳香烃类物质,其对夏、冬季节 SOA 贡献率分别为 80. 39%、94. 63%,主要贡献者为苯、甲苯、乙苯、二甲苯(benzene、toluene、ethylbenzene、xylene,BTEX);对南京工业区 SOA 浓度进行估算,得到夏季 SOA 浓度值为 5. 84 ~ 20. 88 μ g·m⁻³,平均浓度为 12. 15 μ g·m⁻³,冬季为 2. 17 ~ 17. 73 μ g·m⁻³,平均浓度为 6. 91 μ g·m⁻³,冬季 SOA 浓度平均水平明显低于夏季。SOA 浓度值随风速及降水量的增大而减小;使用 PMF 受体模型对 VOCs 进行源解析分析得到夏季 SOA 污染主要来源于涂料使用、石油加工及石油化工源,SOA 贡献值分别为 0. 65、0. 21、0. 18 μ g·m⁻³. 冬季 SOA 污染主要来自于涂料使用,SOA 贡献值为 0. 94 μ g·m⁻³.

关键词: VOCs; SOA 浓度; FAC 系数法; EC 示踪法; PMF 源解析

中图分类号: X513 文献标识码: A 文章编号: 0250-3301(2017)05-1733-10 DOI: 10.13227/j. hjkx. 201610167

Estimating the Secondary Organic Aerosol Concentration and Source Apportionment During the Summer and Winter in the Nanjing Industrial District

LIU Jing-da¹, AN Jun-lin^{1*}, ZHANG Yu-xin¹, SHI Yuan-zhe¹, LIN Xu²

(1. Key Laboratory of Meterological Disaster, Ministry of Education, Joint International Research Laboratory of Climate and Environment Change, Collaborative Innovation Center on Forecast and Evaluation of Meterological Disasters, Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science and Technology, Nanjing 210044, China; 2. Hangzhou Environmental Monitoring Center, Hangzhou 310007, China)

Abstract: Volatile organic compounds (VOCs) were determined by GC5000, an automatic on-line Gas Chromatography-Flame Ionization Detector. Elemental carbon (EC) and organic carbon (OC) were determined by the thermal/optical method using DRI-2001A during the periods of June 15th-July 15th 2015 and December 16th 2015 - January 15th 2016. The concentration of secondary organic aerosol(SOA) was estimated by fractional aerosol coefficients (FAC) and EC tracer method. The source apportionment relied on the positive matrix factorization model (PMF). There were several conclusions: First, aromatic hydrocarbon was the main substance causing the SOA pollution in the Nanjing Industrial district, the contributions of aromatic hydrocarbon to SOA during summer and winter were 80.39% and 94.63%, respectively. The main contributers were benzene, toluene, ethylbenzene, *m*, *p*-xylene and *o*-xylene (BTEX). In the summer, SOA concentration ranged from 5.84-20.88 μg·m⁻³ with an average of 12.15 μg·m⁻³ and in the winter ranged from 2.17-17.73 μg·m⁻³ in which the average concentration was 6.91 μg·m⁻³. Secondly, SOA concentration decreased when wind and precipitation increased. By using the PMF model, a total of 7sources of SOA were determined in summer and 6 were determined in winter. There were 3 main sources in summer, including painting, petroleum processing and petrochemical industry, and the contributions to SOA were 0.65 μg·m⁻³, 0.21 μg·m⁻³, 0.18 μg·m⁻³, respectively. In winter, the most important SOA pollution was from painting, in which the contribution was 0.94 μg·m⁻³.

Key words: volatile organic compounds (VOCs); concentration of secondary organic aerosol; fractional aerosol coefficients (FAC); EC tracer method; positive matrix factorization model and source apportionment

收稿日期: 2016-10-24; 修订日期: 2016-11-23

基金项目:中国科学院战略性先导科技专项(B类)(XDB05020206);国家自然科学基金项目(91544229,41305135);江苏省高校"青蓝工程"项目;江苏省江苏高校优势学科建设工程项目(PAPD)

作者简介: 刘静达(1993~),女,硕士研究生,主要研究方向为大气环境,E-mail: nuistljd@163.com

^{*} 通信作者,E-mail:junlinan@nuist.edu.cn

大气气溶胶(particulate matter, PM)是悬浮在大气中的微小的固体或液体微粒的总称. 其中,细颗粒物的环境效应远比粗颗粒物大,它可以危害人体健康、降低能见度、影响气候变化等 $^{[1^{-3}]}$. 有机气溶胶(organic aerosol, OA)包括污染源直接排放的一次有机气溶胶(primary organic aerosl, POA)以及二次转化形成的 SOA. 一般认为在 PM $_{2.5}$ 中,OA约占到总质量浓度的 20% ~60%,而 SOA 平均可以占到 OA的 20% ~70%,在光化学烟雾的条件下甚至可以占到 70%以上 $^{[4]}$. 在中国,颗粒物中 SOA约占OA的 30% ~95% $^{[5^{-7}]}$.

SOA 是挥发性有机物(volatile organic compounds, VOCs)大气氧化的产物,是大气细颗粒物的重要组成部分^[8]. VOCs 通过各种天然源和人为源排入大气. 在城市地区大气中的主要是由各种人为活动如交通运输、工业生产等引起的排放,进入大气中经过复杂的化学变化和物理过程产生SOA. 这些SOA(包括一些脂类、酸类等)对大气环境质量和人体健康产生更大的危害,其引起的环境污染问题日益凸显. 研究 SOA 的浓度值及其来源特征对揭示复合型大气污染的形成机制和人们健康影响都具有重要意义.

近30年来,对SOA的估算工作得到了国内外学者的重视,其中Grosjean等^[9,10]提出的FAC法与EC示踪法为估算SOA浓度中较为常用的方法.FAC方法的优势在于可以用来估算各种VOC对SOA生成潜势所占比率.对北京^[11]、美国南加州地区^[12]、加拿大英属哥伦比亚地区^[13]等地进行分析,一致认为SOA的主要来源为芳香烃类VOCs氧化生成的.EC示踪法被广泛地应用于SOA生成潜势的

估算,有研究利用 EC 示踪法对全国 14 个城市站点进行冬、夏季同步观测,并估算中国城市大气环境中 SOC(secondary organic carbon)浓度^[14].

由 Paatero 等^[15]提出运用 PMF 模型解决源解析问题. PMF 模型是综合了数据中的误差估计来解决一个受限制加权最小二乘线性模型的矩阵分解法. 由于 VOCs 是 SOA 的前体物,因此通过 PMF 方法分析 VOCs 来源对 SOA 的贡献有重要意义. Sun 等^[16]分析出北京大气中 VOCs 来源为车辆排放、溶剂使用、生物源、燃烧、化工产业及 CFC(氟利昂)这 6个排放源. 林旭等^[17]对南京北郊四季的 VOCs 进行源解析发现不同季节的源解析结果显示工业排放和汽车尾气是南京北郊最主要的 VOCs 污染源.

南京工业区集中了较多的发电、钢铁和石化等重工企业,是典型的工业区. 当地石油化工企业的生产、存储和运输等过程是大气中 VOCs 的重要来源,其中高活性高浓度的 VOCs 也容易造成南京工业区 SOA 的浓度升高. 国内目前对 SOA 的研究主要集中于北京、上海、深圳等地,对南京工业区 SOA 的研究较少. 本研究将利用 FAC 法估算南京工业区夏、冬季节的 VOCs 对 SOA 的生成贡献情况,利用 EC 法估算南京工业区 SOA 浓度情况并使用 PMF 受体模型法对 VOCs 进行源解析,结合 FAC 法分析出对 SOA 浓度组成主要因子,以期为防治 SOA 污染提供建议.

1 材料与方法

1.1 观测时间及观测站点

本研究观测时间为 2015-06-15~07-15 及 2015-12-16~2016-01-15. 如图 1 所示,观测站点位



图 1 观测点地理位置及周围环境情况示意

Fig. 1 Geographical location and the surrounding environment of the observation station

于南京市浦口区南京信息工程大学气象楼楼顶(32°12′N,118°43′E)海拔高度为62 m. 观测点东边500 m 处为南北走向的宁六线干路、双向六车道、江北快速路及地铁S8号线(高架). 观测点正东方向及东北方向3~6 km 处有石油化工厂,钢铁厂及化工厂等,观测点正南方为南京高新区,西南部为龙王山风景区. 具体情况如图1所示. 观测点位置符合中国气象局发布的《大气成分站选址要求》^[18],采样点附近排放源稳定,观测资料能够反映典型城市工业区大气污染特点.

1.2 仪器及分析方法

1.2.1 仪器介绍

VOCs 观测采用由德国 AMA 公司提供的集自 动采样、分析、富集于一体的 GC5000 在线气相色 谱仪. 该系统主要包括 GC5000VOC(单级富集)和 GC5000BTX(两级富集)两台分析色谱仪,可分别测 量 C2~C6 的低沸点物种和 C4~C12 的高沸点物 种,检测器均为氢火焰离子化检测器(FID). 其测量 原理:环境空气样品通过采样经干燥后直接进入分 析系统,有机物在富集管的特殊材料中吸附解析后, 进入毛细管柱进行分离,而后由 FID 检测有机物含 量. 仪器可检测 C2~C12 共计 56 种挥发性有机物, 其中包括29种烷烃、10种烯烃、16种芳香烃和乙 炔. 系统时间分辨率为 1 h. 仪器详细技术参数见 文献[19]. 同时为了保证数据的有效性和可靠性, 观测期间利用 DIM200 校准模块(稀释 100 倍)进行 4周左右一次的校正,其中校正气体采用美国环保 署认可的 Linde Gas North America LLC 提供的混合 标气.

使用石英滤膜采集样品,使用前在马弗炉中以800℃焙烧5h,冷却后放入恒温恒湿箱中平衡24h,用1×10⁻⁵的精密电子天平称重后放入冰箱中低温保存.膜采样使用天虹TH-150C(TSP)中流量空气总悬浮微粒采样器每天采集样品两次,每个样品连续采集11.5h,自08:00~19:30及20:00~07:30.采样结束称重后将样品低温保存在冰箱中直至分析.

DRI-2001A 热/光碳分析仪是一款应用热光法测量原理、设计精良、非常成熟的分析仪,可适用于美国 IMPROVE_A, NIOSH, 加拿大 MSC1 和中国香港 EPD-TOT 等方法,使用膜采样工作后得到的石英膜进行分析,测量大气颗粒物样品中的有机碳(OC)和元素碳(EC)含量. 该型仪器是目前世界上最先进的测量大气样品中 OC/EC 的分析仪器. 样品分析开始、结束和每15 个样品间都采用 He/CH4

标注气体对一起进行校准,确保初始和最后 FID 信号漂移在±3以内、校准峰面积相对偏差在5%以内.实验开始前,对仪器检漏,5 s 内样品炉的压力不变即为不漏气.分析样品前,高温烘烤并做仪器空白确保已除去所残留杂质.为确保数据精确可靠,对每套样品的空白膜进行平行分析,前后误差在10%内可再进行后续样品分析.

气象资料观测:依托南京信息工程大学气象综合观测场,自动气象站记录的全天候连续观测的主要气象要素资料(风速、降水量),其采样间隔为1h,剔除明显的异常值.

1.2.2 分析方法

(1)FAC 方法

本研究基于 Grosjean 的烟雾箱实验,采用 FAC 方法来估算环境大气中 VOCs 的 SOA 生成潜势 $^{[9,10]}$:

$$SOA_p = VOCs_0 \times FAC$$
 (1)

式中, SOA_p 是 SOA 生成潜势; $VOCs_0$ 是排放源排出的初始浓度;FAC 是 SOA 的生成系数. 考虑到受体点测得的 VOCs 往往是经过氧化后的浓度 $VOCs_0$,它与排放源排出的初始度 $VOCs_0$ 之间的关系可通过下式表示:

VOCs₁ = VOCs₀ × (1 - FVOCr) (2) 式中, FVOCr 为 VOCs 物种参与反应的分数(%). Grosjean 假设 SOA 只在白天生成(08:00~17:00), 且 VOCs 只与·OH 发生反应生成 SOA.

公式(1)和(2)中用到的 FAC 和 FVOCr 由烟雾箱实验获得.

(2)EC 示踪法

元素碳 EC 来自化石燃料和生物质等含碳燃料不完全燃烧的直接排放;一次燃烧排放 EC 的同时也伴随产生有机碳 OC(organic carbon, OC). 因此, EC 可作为指示燃烧排放 OC 的示踪物种^[20,21]. 根据 EC 可估算一次有机碳和二次有机碳.

$$SOA = SOC \times Coef_{SOA/SOC}$$

 $SOC = TOC - EC \times (OC/EC)_{min}$

式中,Coef_{SOA/SOC}为 SOC 向 SOA 的转化系数;(OC/EC)_{min}为 OC/EC 比值的最小值. 详细方法见文献 [22,23].

(3)PMF 受体模型

PMF 由芬兰赫尔辛基大学的 Dr. Paatero 在 20 世纪 90 年代中期开发^[24],详细方法见文献[20].

其原理为:假设X为 $n \times m$ 矩阵,n为样品数,m为化学成分的数目,那么X可以分解为X = GF + E,

其中 G 为 $n \times p$ 的矩阵, F 为 $p \times m$ 的矩阵, p 为主要污染源的数目, E 为残数矩阵. 定义:

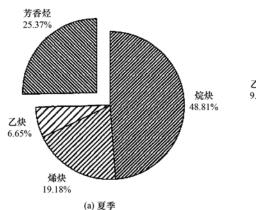
$$e_{ij} = X_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}$$

$$(i = 1, \dots, n; j = 1, \dots, m; k = 1, \dots, p)$$

$$Q(E) = \sum_{i=1}^{n} \sum_{j=1}^{n} (e_{ij}/s_{ij})^{2}$$

式中, X_{ij} 为第 i 个样本中第 j 种物质的浓度; g_{ik} 为第 k 个因子对第 i 个样本的贡献; f_{kj} 为第 j 种物质在第 k 个因子中的百分数; e_{ij} 为第 i 个样本中第 j 种物质的残余; s_{ij} 为第 i 个样本中第 j 种物质的不确定值估计.

本文的 PMF 源解析使用 EPA PMF5.0 模型.将信噪比在 2.0~3.0 之间的物质定义为弱(week),不确定(UNC)扩大 3 倍,降低计算权重.信噪比小于 2.0 的物质定为差(bad),模式计算不采用.大于



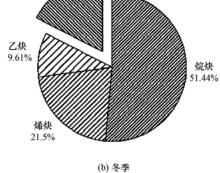


图 2 夏、冬季节各类 VOC 占比

Fig. 2 VOC percentages during summer and winter

2.2 夏、冬季节 VOCs 对 SOA 生成贡献

FAC 估算的 SOA 生成潜势虽然偏低,但是仍能给出 SOA 生成的大致数量级,并指明各 SOA 前体物的相对贡献[11].对 SOA 生成潜势有贡献的 VOCs 主要包括 12 种非芳香烃类物质及 13 种芳香烃类物质.通过 FAC 法计算得出表 1,其中给出 VOCs 对 SOA 的生成贡献潜势值,其中芳香烃类在夏、冬季分别占 TVOCs 的 25.37%、17.44%,对 SOA 的贡献高达到 80.39%、94.63%,非芳香烃对 SOA 的贡献率仅占 19.61%、5.37%.说明芳香烃类物质是 SOA 的主要前体物. 吕子峰等[11]利用 FAC 法对北京市夏季 SOA 生成潜势进行估算,结果芳香烃对 SOA 生成潜势为 76%;希腊雅典环境大气中 60%~90%的 SOA 来自芳香烃[21];在加拿大英属哥伦比亚地区,这个比例约为 80% [13].与本文结论基本一致,因此认为城市中 SOA 污染的主要来源为芳香烃

3.0 的定为强(strong),模式直接使用.

2 结果与讨论

芳香烃

2.1 夏、冬季节 VOCs 的体积分数

物质. 通过与这些城市进行对比可以发现南京工业区芳香烃物质在 VOCs 中所占比率较其他城市高. 这是由于芳香烃物质化学活性较强且浓度高,造成对 SOA 的生成贡献较其他城市高.

表1为夏、冬季节 VOCs 对 SOA 生成潜势情况,可发现 BTEX 为夏、冬季节主要贡献物,但也存在夏、冬贡献差异. 其中夏季 BTEX 对 SOA 生成潜势为 1.43 μg·m⁻³,贡献率为 65.96%,非 BTEX 的 芳香烃对 SOA 生成潜势为 0.31 μg·m⁻³,占比 14.43%. 冬季 BTEX 对 SOA 生成潜势为 0.82 μg·m⁻³,贡献率高达 81.97%,非 BTEX 对 SOA 生成潜势为 0.12 μg·m⁻³,贡献率 12.66%. 说明南京工业区 SOA 污染的主要物质为 BTEX,BTEX 是与石油化工产品、产业相关的常见单环芳烃类物质^[21]. 其中夏季正十一烷对 SOA 的贡献率达至 15.04%,但在冬季贡献率仅为 1.71%. 正十一烷用

表 1	夏	冬季	VOCs	对 SOA	的生	成港垫

Table 1	Potential	formation	of	VOCs to	SOA	during summer and w	inter

				夏季浓度	夏季 SOA	夏季 SOA	冬季浓度	冬季 SOA	冬季 SOA
类别	物种名称	FAC/%	Fvoc _r /%	平均值	× 10 ⁻²	贡献率	平均值	×10 ⁻²	贡献率
				×10 ⁻⁹	/μg·m ⁻³	/%	×10 ⁻⁹	$/\mu g \cdot m^{-3}$	/%
	甲基环戊烷	0. 17	10	0.13	0. 10	0.04	0. 15	0. 10	0.06
	环己烷	0. 17	14	0.47	0.41	0.16	0. 20	0. 15	0.09
	正庚烷	0.06	14	0.14	0.05	0.02	0. 17	0.05	0.03
	甲基环己烷	2. 70	20	0.09	1. 41	0.60	0. 10	1. 50	0.90
	2-甲基庚烷	0.50	10	0.30	0. 72	0.39	0.09	0. 25	0. 15
非芳香烃类	3-甲基庚烷	0.50	10	0.04	0. 08	0.05	0. 03	0.08	0.05
非力育定矢	正辛烷	0.06	17	0.09	0. 04	0.02	0. 11	0.04	0.02
	正壬烷	1.50	20	0.03	0. 22	0.16	0. 01	0. 11	0.06
	正癸烷	2.00	22	0.06	0. 90	0.45	0. 12	1. 91	1. 15
	正十一烷	2.50	25	1.41	32. 93	15.04	0. 12	2. 85	1.71
	正十二烷	3.00	26	0.13	4. 04	1.87	0.05	1. 69	1.01
	异戊二烯	2.00		0.89	1. 78	0.82	0. 11	0. 22	0. 13
总计				3.78	42. 58	19.61	1. 26	8. 96	5. 37
	苯	2.00	10	2.49	19. 28	8.88	2. 18	16. 85	10.09
	甲苯	5.40	12	2.39	60. 27	27.75	2. 88	72. 61	43.48
	乙苯	5.40	15	1.28	38. 54	17.75	1. 25	37. 66	22. 55
	间/对-二甲苯	4. 70	34	0.44	14. 96	6.89	0. 27	9. 09	5. 45
	邻二甲苯	5.00	26	0.32	10. 18	4. 69	0.06	1. 98	1. 18
	异丙苯	4. 00	13	0.06	1. 49	0.69	0.04	1. 02	0.61
芳香烃类	正丙苯	1.60	12	0.04	0. 42	0. 19	0.16	1. 54	0.92
	对-乙基甲苯	2. 50	21	0.22	3. 67	1.69	0. 11	1. 88	1. 13
	间-乙基甲苯	6. 30	31	0.06	2. 88	1.33	0.04	2. 18	1.30
	1,3,5-三甲基苯	2. 90	74	0.05	2. 88	1.33	0. 03	1. 68	1.01
	邻-乙基甲苯	5. 60	23	0.11	4. 47	2.06	0. 12	4. 79	2. 87
	1,2,4-三甲基苯	2.00	58	0.54	13. 85	6.38	0. 18	4. 56	2.73
	1,2,3-三甲基苯	3.60	51	0.04	1. 69	0.77	0.06	2. 18	1.30
总计				8.06	174. 59	80.39	7. 38	158. 01	94. 63

于制造制冷剂、杀虫剂、润滑剂、油漆等^[22],这与夏、冬季节使用的溶剂种类不同有关.

2.3 气象要素变化对 SOA 浓度的影响

利用 EC 示踪法对南京工业区 SOA 浓度进行估算,分析风速与降水量与 SOA 浓度的关系(如图3),研究发现夏季 SOA 浓度为 5.84 ~ 20.88 $\mu g \cdot m^{-3}$,平均浓度为 12.15 $\mu g \cdot m^{-3}$;冬季 SOA 浓度为 2.17~17.73 $\mu g \cdot m^{-3}$,平均浓度为 6.91 $\mu g \cdot m^{-3}$.

气象条件是影响 SOA 浓度值大小的重要因素. 静稳条件下往往造成气态污染物的累积从而导致二次颗粒物的生成. 夏季中 SOA 浓度最小值出现在 6 月 27 日,当日平均风速达 2.33 m·s⁻¹,降水量达 5.88 mm,而 7 月 2 日 SOA 浓度 高 达 20.88 μg·m⁻³,平均风速为 0.98 m·s⁻¹,无降水. 发现 SOA 浓度值随降雨量和风速的增大而减小. 此现象在冬季中也有良好体现,其中 SOA 浓度最小值出现在 1 月 5 日,其中 1 月 4 日 20:00 至 1 月 5 日 07:00 有强降水过程,累计雨量达 2.52mm,当日平均风速

为 2.61 m·s⁻¹.

Xu 等[23] 利用 EC 示踪法对北京市 2009 年 8 月 至 2011 年 8 月的数据进行分析,对 SOC 估算得到 夏季 SOC 浓度值为 1~3 μg·m⁻³,冬季 SOC 浓度值 为2~6 μg·m⁻³. 由于北京冬季供暖需求大,污染 物排放增多,造成冬季 SOC 污染较夏季严重. Khan 等[24]估算出意大利东北部地区 Veneto 的 SOC 浓度 平均值为 3.8 μg·m⁻³. Lonati 等^[25]对 Milan 的 SOC 进行估算,估算结果为 4.36 μg·m⁻³, Harrison 等^[26] 对 Birmingham 的 SOC 进行估算,估算出夏季 SOC 浓度值为 0.6 µg·m⁻³,冬季为 1.2 µg·m⁻³,由 SOC 向 SOA 转化过程中的转化系数一般为 1.2~1.6,在 城市中一般为 1.6^[27],将各地区 SOC 与转化系数相 乘,得出各地区 SOA 浓度值. 与南京工业区 SOA 浓 度值进行对比发现,南京冬季 SOA 浓度明显低于夏 季,这是由于冬季光照强度较夏季弱、光化学反应 时间较夏季短[28,29]及风速大等一系列原因综合导 致的. 对比分析其他城市冬季 SOA 浓度值明显高

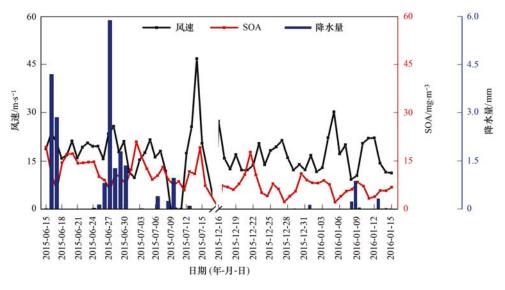


图 3 风速与降水量对 SOA 浓度的影响

Fig. 3 Effect of wind speed and precipitation on SOA concentration

于夏季,这与冬季供暖,污染源排放的 VOCs 浓度增大有关.

2.4 PMF 受体模型源解析

本文最终对夏季中 42 种 VOCs, 冬季 32 种 VOCs 进行源解析.解析结果的残数值大部分在 -3.0 ~3.0 之间,随着因子数的调整,计算结果趋于稳定.本文的选取原则详见文献[30].

2.4.1 夏季 PMF 源解析结果分析

PMF 经过多次运行,最终确定为 7 个因子,其中 6 个为人为源,1 个为生物源. 从图 4 各排放源对 VOCs 的贡献可以看出,因子(a)中乙苯、二甲苯、苯乙烯等苯系物贡献值较大,这些物质为涂料使用的主要成分^[31]. 异戊二烯是生物排放的主要示踪物,由于观测点紧邻龙王山,且夏季树木茂盛造成异

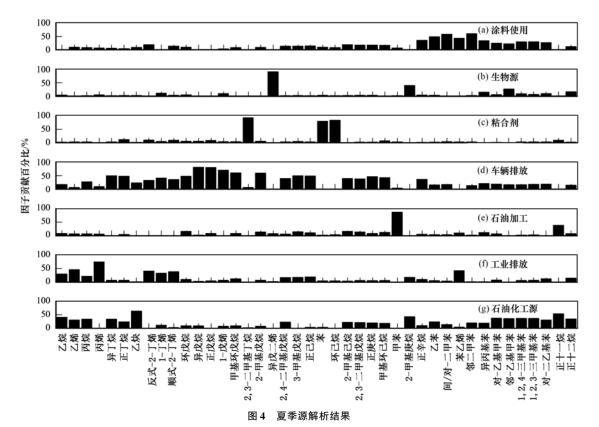


Fig. 4 Result of source apportionment in summer

戊二烯排放量较大,因此将因子(b)定为生物 源[32]. 苯、正己烷对因子(c)贡献较大,这两种物 质主要来源于粘合剂的使用[33],2,3-二甲基丁烷主 要来源于有机合成过程[34]. 异戊烷、正戊烷主要来 源于因子(d)的贡献,这两种物质为汽油未完全燃 烧的产物,正丁烷及异丁烷为汽油挥发产物[35,36], 因此将其归纳为车辆排放源. 因子(e)中主要贡献 物为甲苯,甲苯可从石油加工中的芳构化过程制得, 由粗苯精制可以得到甲苯[37];正十一烷主要用于石 油研究和有机合成,因此将其定义为石油加工. 丙 烯、反式-2-丁烯、1-丁烯及顺式-2-丁烯在因子(f) 中占比较大,为工业源的排放的典型 VOCs^[31]. 因 子(g)中乙烯、乙炔、丙烯及苯系物含量较高,这些 物种均为石油工业源排放的 VOCs 特征组分,常来 源于制鞋、家具、羽毛类产品、印刷、家居和其他 一些工业生产过程[38],将其定义为石油化工源.

2.4.2 冬季 PMF 源解析结果分析

图 5 为对冬季 PMF 源解析的结果,共解析出 6 种人为源. 其中将因子(a)定义为涂料使用,是因为苯系物及正辛烷对其贡献较大,这些物质均为涂料使用的主要来源^[39]. 因子(b)中,主要贡献物为环己烷及间/对-二甲苯. 环己烷主要用于制备尼龙 6 及尼龙 66,主要用作纤维素醚、树脂、蜡、油脂、沥

青、橡胶等物质的优良溶剂^[40],间/对-二甲苯为进一步生产不饱和聚酯、塑料、树脂固化剂、纤维、树脂的原料^[41],因此将其定义为溶剂使用. 苯来源于石油加工的芳构化过程或通过焦化工业中的粗苯精制得到^[42],因此将因子(c)定义为石油加工,苯为其产物. 其余3种人为源与夏季工业排放、车辆排放及石油化工源吻合性强,因此认为是相同排放源.

2.4.3 夏、冬季源解析结果对比分析

在 PMF 源解析过程中,可以发现,夏季与冬季的排放源情况基本稳定,但在 SOA 贡献比率上存在 差异.

在涂料使用一项中,冬季与夏季的涂料使用组成成分及对 SOA 生成潜势的贡献存在差异. 甲苯的化学活性较强、对 SOA 生成潜势贡献大,而冬季涂料使用源中排放的甲苯含量较高,造成其对 SOA 生成潜势贡献明显增大. 冬季并未解析出生物源,这是由于冬季龙王山植被减少. 冬季并未解析出生物源及粘合剂源,前者是由于冬季龙王山植被减少,后者是因为其在冬季排放出的 VOCs 类物质浓度较低及冬季风速较大等综合原因. 冬季解析出的溶剂使用源主要来源于轮胎加工厂、皮革制造厂等. 其他源并未存在明显的冬夏差异.

利用 FAC 法估算的冬季排放的 SOA 生成潜势

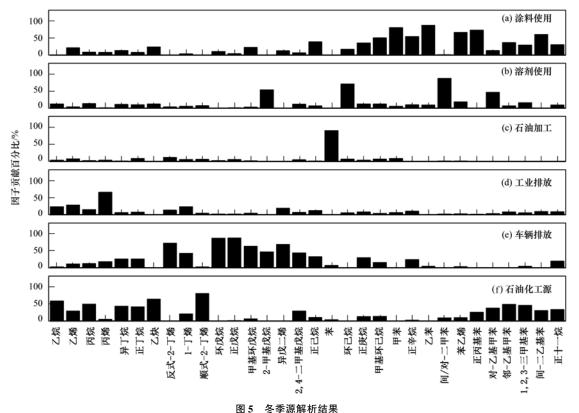


Fig. 5 Result of source apportionment in winter

远小于夏季,这与 EC 示踪法估算结果趋势相一致. **2.5** 夏、冬季节各源对 SOA 生成潜势结果估算

综合 FAC 法结合 PMF 源解析方法,得到各因子中 VOCs 对 SOA 的生成潜势及各因子对 SOA 的贡献值,如图 6. 从中可以得到,夏季 SOA 生成潜势值为 2.00 μg·m⁻³,其中 BTEX 及 1,2,4-三甲基苯为南京工业区夏季 SOA 污染的主要贡献者,其中石油加工、涂料使用及石油化工源为三大污染源,造成这种现象的原因,是因为这 3 种源主要排放的为

VOCs 活性较强的芳香烃类物质. 三者对 SOA 生成潜势分别为 0.65、0.50 及 0.30 $\mu g \cdot m^{-3}$,分别占总 SOA 浓度的 32.46%、25.24% 及 15.09%. 冬季 SOA 生成潜势值为 1.48 $\mu g \cdot m^{-3}$,冬季 SOA 生成潜势间最大的物质仍主要为 BTEX,主要贡献源为涂料使用、石油加工及溶剂使用,生成潜势分别为 0.94、0.21 和 0.18 $\mu g \cdot m^{-3}$,分别占总 SOA 浓度的 63.57%、14.45% 及 12.16%. 其中涂料使用中仅甲苯的 SOA 生成潜势就达 0.56 $\mu g \cdot m^{-3}$.

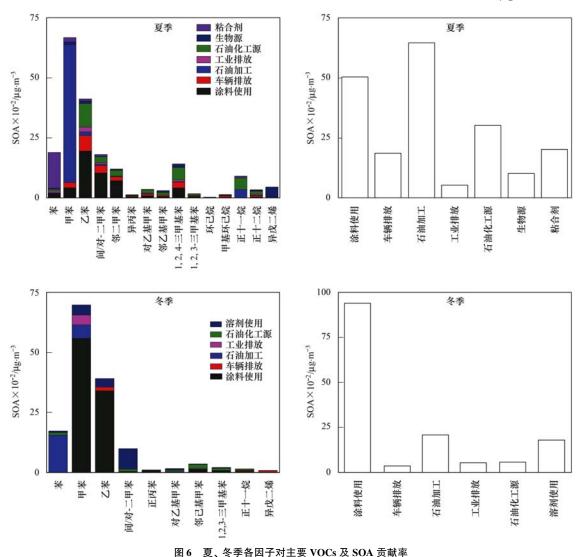


Fig. 6 Main VOCs factors and SOA contribution percentages during summer and winter

3 结论

(1)南京工业区夏季 VOCs 的体积分数明显小于冬季,但夏季芳香烃体积分数为冬季的 1.14 倍. SOA 污染主要来源于芳香烃类物质,造成南京工业区夏季 SOA 浓度值高于冬季. 芳香烃物质对夏、冬季节 SOA 的贡献率分别为 80.39%、94.63%,主要

贡献者为 BTEX.

(2)对南京工业区进行 SOA 估算,发现夏季 SOA 浓度值为 5.84 ~ 20.88 $\mu g \cdot m^{-3}$,平均浓度为 12.15 $\mu g \cdot m^{-3}$;冬季为 2.17 ~ 17.73 $\mu g \cdot m^{-3}$,平均浓度为 6.91 $\mu g \cdot m^{-3}$.冬季 SOA 浓度平均水平明显低于夏季. SOA 浓度值随风速及降水量的增加而减小.

(3)使用 PMF 受体模型对 VOCs 进行源解析分析得到夏季共有 7 个 SOA 污染源,其中主要污染源为涂料使用、石油加工及石油化工源,SOA 浓度值分别为 0.94、0.21、0.18 μg·m⁻³.冬季共有 6 个污染源,主要污染源为涂料使用,SOA 贡献值为 0.94 μg·m⁻³.冬季与夏季的源解析结果存在差异,但主要贡献源排放情况稳定.

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