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贵阳市花溪城区大气PM_{2.5}中碳质气溶胶的变化特征及来源解析

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摘要: 碳质气溶胶作为大气气溶胶的重要组成部分, 对大气环境质量、人类健康及全球气候变化有着重要的影响。为探究贵阳市花溪城区大气细颗粒物(PM_{2.5})中碳质气溶胶的变化特征及来源, 于2020年不同季节开展大气PM_{2.5}原位观测研究, 利用热/光学碳分析仪(DRI Model 2015)测定大气PM_{2.5}的碳质组分。结果表明, 观测期间大气 $\rho(\text{PM}_{2.5})$ 、 ρ [总碳质气溶胶(TCA)]、 ρ [有机碳(OC)]、 ρ [二次有机碳(SOC)]和 ρ [元素碳(EC)]的平均值分别为:(39.7±22.3)、(14.1±7.2)、(7.6±3.9)、(4.4±2.6)和(2.0±1.0) $\mu\text{g}\cdot\text{m}^{-3}$, OC/EC的平均值为(3.9±0.8)。 $\rho(\text{PM}_{2.5})$ 、 $\rho(\text{TCA})$ 、 $\rho(\text{OC})$ 、 $\rho(\text{SOC})$ 和 $\rho(\text{EC})$ 呈现冬季最高[(52.6±28.6)、(17.0±9.6)、(9.1±5.2)、(6.1±3.9)和(2.4±1.2) $\mu\text{g}\cdot\text{m}^{-3}$], 夏季最低[(25.1±7.1)、(11.6±3.6)、(6.3±1.9)、(3.7±1.2)和(1.6±0.6) $\mu\text{g}\cdot\text{m}^{-3}$]的季节变化特征。OC/EC季节变化呈现:夏季(4.2±0.8)>冬季(3.8±0.9)>秋季(3.8±0.5)>春季(3.7±0.9), 表明花溪城区各季节均存在SOC生成。SOC与OC呈现显著相关($R^2=0.9$), 且随着大气氧化性增强, SOC浓度呈增加趋势。OC与EC各季节均呈现较好相关性, 其中秋季最高($R^2=0.9$), 其他3个季节偏低(R^2 为0.74~0.75), 表明二者具有共同来源。通过OC/EC值范围初步判断碳质气溶胶来源于机动车尾气排放、燃煤排放和生物质燃烧排放。为了进一步定量解析主要排放源对碳质气溶胶的贡献, 利用PMF模型对碳质气溶胶来源解析, 结果表明贵阳市花溪城区碳质气溶胶主要来源为燃煤源(29.3%)、机动车排放源(21.5%)和生物质燃烧源(49.2%)。

关键词: PM_{2.5}; 碳质气溶胶; 二次有机碳; 来源解析; 花溪

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Characteristics and Source Analysis of Carbonaceous Aerosols in PM_{2.5} in Huaxi District, Guiyang

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Abstract: Carbonaceous aerosol, as an important component of atmospheric aerosol, has a significant impact on atmospheric environmental quality, human health, and global climate change. To investigate the characteristics and sources of carbonaceous aerosol in atmospheric fine particulate matter (PM_{2.5}) in Huaxi District of Guiyang, an in-situ observational study was conducted during different seasons in 2020, and the carbonaceous components of PM_{2.5} were measured using a thermal-optical carbon analyzer (DRI Model 2015). The results of the study showed that the average concentrations of PM_{2.5}, total carbonaceous aerosol (TCA), organic carbon (OC), secondary organic carbon (SOC), and elemental carbon (EC) concentrations during the observation period were (39.7±22.3), (14.1±7.2), (7.6±3.9), (4.4±2.6), and (2.0±1.0) $\mu\text{g}\cdot\text{m}^{-3}$, respectively, and the mean value of OC/EC was (3.9±0.8). $\rho(\text{PM}_{2.5})$, $\rho(\text{TCA})$, $\rho(\text{OC})$, $\rho(\text{SOC})$, and $\rho(\text{EC})$ showed a seasonal variation pattern with the highest in winter [(52.6±28.6), (17.0±9.6), (9.1±5.2), (6.1±3.9), and (2.4±1.2) $\mu\text{g}\cdot\text{m}^{-3}$], and the lowest in summer [(25.1±7.1), (11.6±3.6), (6.3±1.9), (3.7±1.2), and (1.6±0.6) $\mu\text{g}\cdot\text{m}^{-3}$, respectively]. The seasonal variation in OC/EC showed summer (4.2±0.8) > winter (3.8±0.9) > autumn (3.8±0.5) > spring (3.7±0.9), indicating the presence of SOC generation in all seasons in Huaxi District. SOC showed a significant correlation with OC ($R^2=0.9$), and the SOC concentration tended to increase with the increase in atmospheric oxidation. OC showed a good correlation with EC in all seasons, with the highest in autumn ($R^2=0.9$) and lower correlations in the other three seasons (R^2 ranged from 0.74 to 0.75), indicating a common source. According to OC/EC ratio range, it was preliminarily determined that carbonaceous aerosol came from vehicle exhaust emissions, coal burning emissions, and biomass combustion emissions. In order to further quantify the contribution of major emission sources to carbonaceous aerosol, the results of this study using PMF to analyze the sources of carbonaceous aerosol showed that the main sources of carbonaceous aerosol in Huaxi District of Guiyang were coal combustion sources (29.3%), motor vehicle emission sources (21.5%), and biomass combustion sources (49.2%).

Key words: PM_{2.5}; carbonaceous aerosol; secondary organic carbon; source apportionment; Huaxi

当前,我国大气PM_{2.5}污染已成为政府、研究者以及公众关注的重大环境问题。大气PM_{2.5}化学成

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分复杂,主要由碳质组分、水溶性离子和金属元素等组成,其中碳质气溶胶是大气 $\text{PM}_{2.5}$ 的重要组成部分,主要由有机碳(organic carbon, OC)和元素碳(elemental carbon, EC)组成^[1]. OC来源较复杂,除了工业生产活动排放、燃料燃烧和自然源等主要污染源排放的一次有机碳(primary organic carbon, POC),此外,大气中的挥发性有机污染物经过光化学氧化反应、凝结及成核后从气相转化到颗粒相的二次有机碳(secondary organic carbon, SOC)^[2]. EC主要来自化石燃料和生物质的不完全燃烧过程,属于一次排放过程,在大气中存在状态相对稳定^[3]. OC和EC组分对城市大气 $\text{PM}_{2.5}$ 的贡献占比变化幅度较大,一般约为20%~70%^[4]. 碳质气溶胶在气候变化过程中扮演着重要角色,对地球辐射平衡产生直接和间接辐射强迫影响,OC和EC分别通过散射和吸收作用对大气造成负和正的辐射强迫,其光学散射和吸收特性会降低大气能见度,并且可携带大量有毒有害物质进入人体,对健康造成危害^[5-7]. 多种源解析方法被应用于追踪大气中碳质气溶胶的潜在来源^[8-10]. 受体模型方法中,相比于化学质量平衡(chemical mass balance, CMB)和主成分分析模型(principal component analysis, PCA),正交矩阵因子分解模型(positive matrix factorization, PMF)解析矩阵中每个因子的分担率均为非负值且不需要当地颗粒物组分的源成分谱,可以对每一个单独的样品点进行解析,是大气颗粒物及其化学组分源解析研究中应用较为广泛的受体模型之一^[11].

目前对大气 $\text{PM}_{2.5}$ 中碳质气溶胶的研究主要集中在碳组分的浓度水平、时空演化规律、辐射效应、粒径分布、转化机制及来源解析方面,且研究区域主要集中于京津冀、长三角、珠三角、关中地区及成渝地区. 张哲^[12]对2018年长三角典型城市大气 $\text{PM}_{2.5}$ 中碳质组分时空变化特征研究表明,上海、杭州和南京碳质气溶胶组分在 $\text{PM}_{2.5}$ 中的占比为29%~34%, $\rho(\text{OC})$ 年均值分别为 (8.0 ± 5.0) 、 (12.4 ± 6.6) 和 $(9.3 \pm 4.5) \mu\text{g} \cdot \text{m}^{-3}$, 且是EC浓度的两倍,表明长三角地区主要城市的含碳气溶胶是以有机碳为主. Wang等^[13]对青藏高原地区大气中碳质气溶胶中黑碳组分研究发现,黑碳气溶胶的来源、传输及辐射效应很可能与大气条件的复杂变化相关. Huang等^[14]利用PMF模型对广州市大气 $\text{PM}_{2.5}$ 中碳质组分来源解析结果表明,燃煤、交通排放、土壤扬尘和船舶排放是碳质气溶胶的主要排放源.

贵阳市作为贵州省的省会城市、西南陆海新通道重要节点城市、国家内陆开放型经济试验区核心区和全国生态文明示范城市,对贵阳市生态环境保

护的力度和压力都在日益增加. 随着工业化进程不断加快,能源结构和产业结构发生了很大变化,随之大气 $\text{PM}_{2.5}$ 、 SO_2 、 NO_2 和 O_3 等污染物也相应发生变化,贵阳市污染源由以工业源为主,向工业源、生活源和交通源等并存转变,不断出现新型复合污染. 有学者针对近年贵阳市大气碳质气溶胶进行了研究,如敖娅等^[15]对贵阳市秋冬季 $\text{PM}_{2.5}$ 与 PM_{10} 中黑碳气溶胶进行了分析;王珍等^[16]分析了贵阳市秋冬季 $\text{PM}_{2.5}$ 中OC和EC的浓度变化特征. 但是贵阳市大气颗粒物中含碳气溶胶的连续性、系统性观测数据仍比较有限,对含碳气溶胶来源及形成机制还不清楚. 因此,本研究于2020年对花溪城区大气 $\text{PM}_{2.5}$ 中碳质组分进行观测研究,并通过理论计算SOC和POC组分,分析不同季节OC、EC、SOC和POC的演化规律,并对OC和EC的来源进行解析,确定贵阳市花溪城区OC和EC的主要排放源,以期有效控制及治理贵阳市花溪城区大气 $\text{PM}_{2.5}$ 中的碳质气溶胶污染提供基础数据和理论依据.

1 材料与方法

1.1 采样点和采样方法

采样点设置在贵州大学资源与环境工程学院楼顶(106.65°E , 26.44°N),距地面约20 m,观测站所处地形平坦,受局地环流影响较小,周围500 m内多为文教、行政和居民区,周围无明显的建筑物遮挡,视野开阔,无明显的局地排放源,采样点南面800 m处有一条交通主干道甲秀南路.

在2020年春季(4月)、夏季(7月)、秋季(10月)和冬季(12月)利用青岛金仕达智能颗粒物中流量采样器(KB-120F)采集大气 $\text{PM}_{2.5}$ 样品,采样流量为 $100 \text{ L} \cdot \text{min}^{-1}$. 使用石英纤维膜(直径90 mm, Whatman QMA, 英国)富集 $\text{PM}_{2.5}$, 滤膜使用前将石英膜在马弗炉中以 450°C 焙烧3 h,以消除可能残留滤膜表面的有机物,冷却后放入恒温恒湿箱中平衡24 h(温度为 25°C , 湿度为50%). 每张滤膜连续采集23.5 h(即09:00至次日08:30),采样时将提前准备好的滤膜放入 $\text{PM}_{2.5}$ 切割器,采样开始时和采样结束后分别记录大气环境温度、天气情况、气压、工况体积、标况体积和异常情况. 观测期共采集大气 $\text{PM}_{2.5}$ 样品123个,每天样品采集完立即称量后用膜盒装好再用锡箔纸封存装入自封袋,放入冰箱低温保存以备后续化学组分分析使用.

1.2 样品分析

OC和EC的质量浓度采用热/光学碳分析仪进行分析. 为了消除实验误差,测样前进行1~2次空白膜测定,将分析仪残留的气体充分燃烧. 测样时,在无氧

纯 He 的环境中,分别在 140℃(OC1)、280℃(OC2)、480℃(OC3)和 580℃(OC4)温度下热解有机碳,部分 OC 碳化形成 OPC. 然后样品在含 2% 氧气的氦气环境下,于 580℃(EC1)、740℃(EC2)和 840℃(EC3)逐步加热氧化元素碳. 上述各个温度梯度下挥发出的含碳化合物经 MnO₂催化氧化转化成 CO₂,再经 Ni 催化还原转化为 CH₄,通过火焰离子化检测器(FID)定量检测. 最后定义 OC 总量为 OC1+OC2+OC3+OC4+OPC, EC 总量为 EC1+EC2+EC3-OPC,关于热光碳分析仪详细分析原理见文献[17].

1.3 数据分析

TCA 通常用 OC 转化成的总有机物(OM)与 EC 之和来估算,城区颗粒物 OM 为 OC 的 1.6 倍则^[18]:

$$TCA = 1.6 \times OC + EC$$

SOC 浓度计算公式为:

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

式中,OC 为有机碳浓度(μg·m⁻³),EC 为元素碳浓度(μg·m⁻³), (OC/EC)_{min} 为各季节研究时段两者最小值^[19].

PMF 是一种不需要源谱数据和气象数据的多变量因子分析受体模型,PMF 基于最小二乘法对原始数据以及不确定度数据进行分解,确定目标函数 Q 最小化的解,从而确定污染源的贡献率和源成分谱,再根据源成分谱来对源进行识别,具体详细内容见文献[20,21].

2 结果与讨论

2.1 PM_{2.5} 和 TCA 总体特征

观测期间,ρ(PM_{2.5})均值为(39.7±22.3) μg·m⁻³,

稍高于国家环境空气质量二级标准年均限值 35 μg·m⁻³(GB 3095-2012),日均最大值出现在 12 月 25 日,为 123 μg·m⁻³,高于国家二级标准(75 μg·m⁻³). 与国内其他城市观测期 ρ(PM_{2.5})相比较,本研究其平均值明显低于成都(118.0 μg·m⁻³)^[22]、重庆(56.2 μg·m⁻³)^[23]和长沙(52.3 μg·m⁻³)^[24]等周边城市,高于张家口(32.3 μg·m⁻³)^[25]、承德市(31.3 μg·m⁻³)^[26]和舟山(26.3 μg·m⁻³)^[27]等地区. ρ(PM_{2.5})季节变化呈现出冬季[(52.6±28.6) μg·m⁻³] > 春季[(46.4±18.5) μg·m⁻³] > 秋季[(35.0±19.2) μg·m⁻³] > 夏季[(25.1±7.1) μg·m⁻³] 的变化特征,且冬季浓度平均值是夏季的约 2 倍. 冬季 PM_{2.5} 容易出现高值,可能是由于冬季不利的气象条件和源排放叠加造成 PM_{2.5} 浓度的累积^[28]. ρ(TCA)平均值为(14±7.2) μg·m⁻³,对 PM_{2.5} 平均贡献率为 42.7%,表明花溪城区碳质气溶胶是 PM_{2.5} 的重要组成部分,控制碳质气溶胶对防控大气 PM_{2.5} 的污染具有重要意义. ρ(TCA)春、夏、秋和冬季均值分别为:(15.2±5.4)、(11.6±3.6)、(12.6±7.5)和(17.0±9.6) μg·m⁻³,呈现与 PM_{2.5} 一致的季节变化特征. TCA 对 PM_{2.5} 平均贡献呈现夏季(46.3%)高于其他 3 个季节(见表 1),表明清洁天大气 PM_{2.5} 中碳质气溶胶占主导^[19,29]. 这可能是由于夏季强烈的阳光和高温会促进二次碳质气溶胶的生成,而频繁的降雨清洁,导致 PM_{2.5} 浓度整体降低^[30]. 本研究 TCA 与国内其他城市研究结果比较,低于厦门市郊区[(28.0±15.7) μg·m⁻³]^[31]、乌鲁木齐[(24.2±9.8) μg·m⁻³]^[32]和承德市[(21.4±13.7) μg·m⁻³]^[26]等国内城市,表明贵阳市花溪区碳质气溶胶污染较低,但在研究期间冬季气溶胶污染的增加,仍是防控碳质气溶胶污染需要关注的焦点.

表1 贵阳市花溪城区不同季节大气 PM_{2.5}、各碳质组分的浓度平均值以及对 PM_{2.5} 的贡献率
Table 1 Average concentration of atmospheric PM_{2.5}, carbon chemical components, and their contribution to PM_{2.5} during different seasons in Huaxi District, Guiyang

名称	春季	夏季	秋季	冬季	平均值
ρ(PM _{2.5})/μg·m ⁻³	46.4±18.5	25.1±7.1	35.0±19.2	52.6±28.6	39.7±22.3
ρ(TCA)/μg·m ⁻³	15.2±5.4	11.6±3.6	12.6±7.5	17.0±9.6	14.1±7.2
ρ(OC)/μg·m ⁻³	8.1±2.7	6.3±1.9	6.7±4.0	9.1±5.2	7.6±3.9
ρ(EC)/μg·m ⁻³	2.3±1.0	1.6±0.6	1.8±1.0	2.4±1.2	2.0±1.0
ρ(POC)/μg·m ⁻³	3.9±1.7	2.6±1.0	3.0±1.8	3.1±1.6	3.2±1.6
ρ(SOC)/μg·m ⁻³	4.1±1.5	3.7±1.2	3.7±2.4	6.1±3.9	4.4±2.6
OC/EC	3.7±0.9	4.2±0.8	3.8±0.5	3.8±0.9	3.9±0.8
TCA/PM _{2.5} /%	33.5	46.3	35.3	32.3	36.9
OC/PM _{2.5} /%	17.8	25.1	18.9	17.2	19.8
EC/PM _{2.5} /%	5.0	6.2	4.1	4.8	5.3
POC/PM _{2.5} /%	8.5	10.5	8.7	6.2	8.5
SOC/PM _{2.5} /%	9.3	14.5	10.2	11.1	11.3
POC/OC/%	48.7	42.0	45.2	33.6	41.9
SOC/OC/%	51.3	58.0	54.8	66.4	58.1

2.2 $PM_{2.5}$ 中碳质组分特征

图1展示了贵阳市花溪城区不同季节大气 $PM_{2.5}$ 中OC、EC、EC/ $PM_{2.5}$ 、OC/ $PM_{2.5}$ 和OC/EC的逐日变化特征。观测期间, $\rho(OC)$ 和 $\rho(EC)$ 变化分别在 $2.0 \sim 21.0 \mu g \cdot m^{-3}$ 和 $0.5 \sim 5.6 \mu g \cdot m^{-3}$ 之间,平均值分别为 $(7.6 \pm 3.9) \mu g \cdot m^{-3}$ 和 $(2.0 \pm 1.0) \mu g \cdot m^{-3}$ 。表2是本研究含碳气溶胶浓度水平与国内其他城市比较,含碳气溶胶浓度水平在中国北部、西南地区及沿海城市存在着区域差异^[33],主要归因于不同能源类型的排放源不同^[30]。花溪城区OC和EC浓度平均值与天津相近,OC低于北京和郑州,EC则略高。与上海、杭州、深圳和珠海等沿海城市相比,OC和EC较低。但与南京相比,贵阳市花溪城区碳质气溶胶浓度较高。

$\rho(OC)$ 和 $\rho(EC)$ 在春、夏、秋、冬季的逐日变化范围($\mu g \cdot m^{-3}$)分别为:2.6~13.5和0.5~4.4、2.0~12.3和0.5~2.8、1.9~17.2和0.5~4.6、2.1~21.0和1.1~5.6,其中12月25日出现峰值,浓度分别为 $21.0 \mu g \cdot m^{-3}$ 和 $5.6 \mu g \cdot m^{-3}$,季节变化呈现冬季高于其他3个季节,结合气象要素可知,该日高湿低温(日均温度为 $7.7^{\circ}C$,相对湿度为86%)静风条件增强了碳质气溶胶的累积,导致污染物不能扩散,该研究结果与徐雪梅等对成都市主城区 $PM_{2.5}$ 中碳质组分的

研究结果一致^[34]。冬季出现类似的变化情况较多,这主要的原因可能为:①冬季燃煤和生物质燃烧,尤其农村散煤和生物质燃烧增加是该季节大气污染的重要来源,对OC的排放贡献较大。有研究发现武汉市冬季居民燃煤供暖以及散煤燃烧使得OC浓度显著增加,为SOC的生成提供了大量前体物^[35];②冬季低温有利于半挥发有机物在颗粒物表面凝结,导致碳质组分浓度增加^[36],此外,冬季汽油和柴油车冷启动会增加OC的排放,温度低会导致燃料燃烧不完全,导致污染物的排放高于其他季节^[37,38];③冬季容易逆温,混合层高度低,不利于污染物扩散,这与广州市冬季碳质组分研究结果相同^[39]。

OC/ $PM_{2.5}$ 和EC/ $PM_{2.5}$ 在春、夏、秋、冬季平均值分别为:(17.8 ± 1.8)%和(5.0 ± 1.2)%、(25.1 ± 2.9)%和(6.2 ± 1.4)%、(18.9 ± 3.1)%和(5.1 ± 1.1)%、(17.2 ± 3.6)%和(4.8 ± 1.2)%。观测期间,EC对 $PM_{2.5}$ 贡献的季节差异较小,贡献率变化范围为2.6%~8.3%,平均值为(5.3 ± 1.3)%,OC对 $PM_{2.5}$ 的贡献率为(19.8 ± 4.3)%,与王珍等^[46]对贵阳市秋冬季研究结果相比,花溪城区的碳组分浓度明显降低,随着国家实施《大气污染防治行动计划》以来对大气污染源排放的控制,花溪城区碳质气溶胶的污染明显减轻。

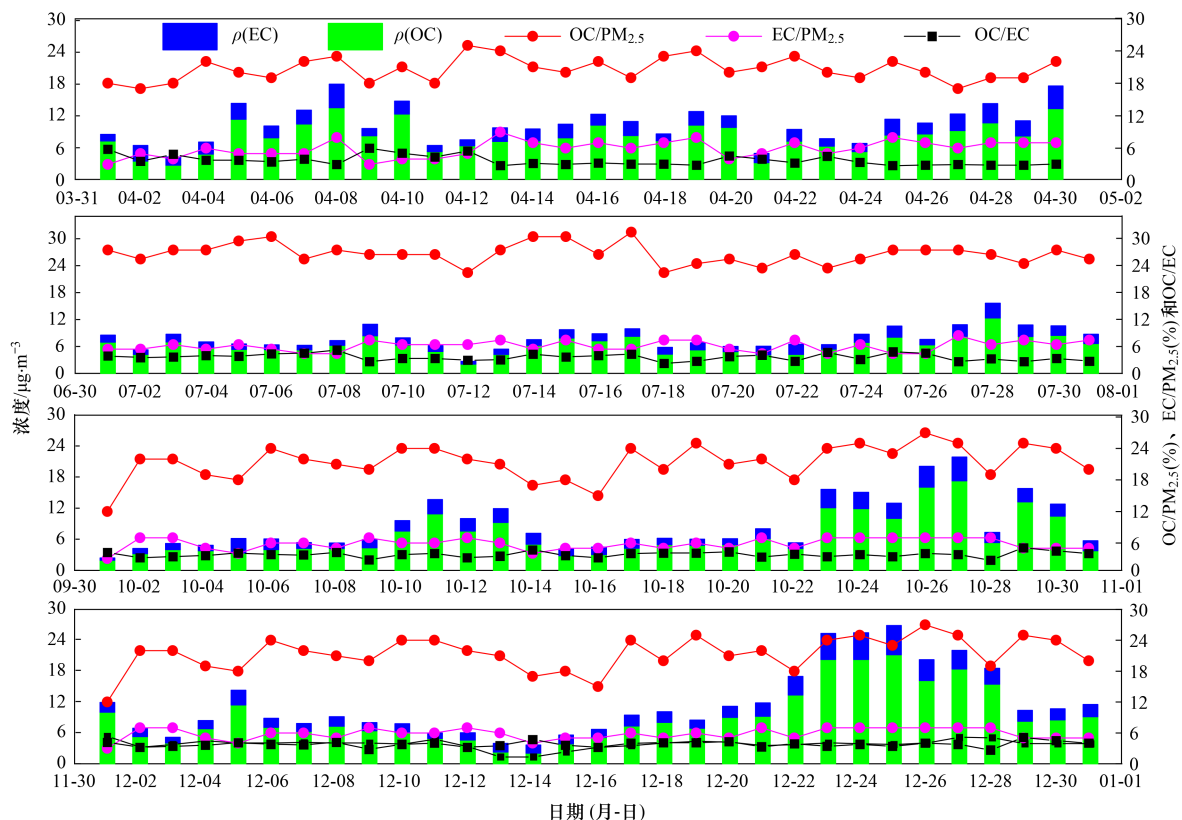


图1 贵阳市花溪城区不同季节 $PM_{2.5}$ 中OC、EC、EC/ $PM_{2.5}$ 、OC/ $PM_{2.5}$ 和OC/EC的逐日变化

Fig. 1 Daily variation in OC, EC, EC/ $PM_{2.5}$, OC/ $PM_{2.5}$, and OC/EC in $PM_{2.5}$ during different seasons in Huaxi District, Guiyang

表 2 国内主要城市大气PM_{2.5}中OC和EC的浓度平均值及OC/EC比较¹⁾

Table 2 Comparison of the OC and EC average concentration and ratios of OC/EC in PM _{2.5} in Huaxi District of Guiyang and some cities in China					
城市	采样时间(年-月)	$\rho(\text{OC})/\mu\text{g}\cdot\text{m}^{-3}$	$\rho(\text{EC})/\mu\text{g}\cdot\text{m}^{-3}$	OC/EC	文献
北京	2017-12~2018-12	11.2±7.8	1.2±0.8	9.7	[19]
天津	2017-01~2019-12	7.7	2.1	—	[40]
郑州	2019-04~2020-01	9.4	1.5	—	[41]
上海	2018-03~2018-12	8.0±5.0	3.8±2.4	—	[12]
杭州	2019-10~2019-12	10.0	2.4	4.2	[27]
南京	2017-01~2017-12	5.9±3.4	2.9±1.5	2.0±0.7	[42]
广州	2015-01~2015-11	8.3±4.5	3.6±2.7	2.8±1.0	[43]
深圳	2015	12.0	2.7	2.5	[44]
珠海	2014-04~2015-01	9.4±4.6	2.6±1.6	4.3	[45]
重庆	2019-11~2020-10	9.0±5.7	2.5±1.5	3.8±1.4	[46]
武汉	2018-01~2018-12	9.2±1.1	3.8±0.5	—	[47]
百色	2015-12~2016-12	11.7	4.7	2.4	[48]
成都	2020-01~2021-05	6.4±3.2	3.2±1.1	2.2±0.5	[49]
贵阳	2013-10~2013-12	20.5	3.6	—	[16]
贵阳	2020-04~2020-12	7.6±3.9	2.0±1.0	3.9±0.8	本研究

1)“—”表示文章中无相关数据

2.3 PM_{2.5}中二次有机气溶胶变化特征

碳质气溶胶主要由一次排放(EC和POC)和二次生成(SOC)组成,因常用OC/EC值初步判断排放源和化学转化的老化特性,OC/EC常被用作估算SOC和示踪碳质气溶胶排放源,当OC/EC值大于2时,可认为存在SOC污染^[50].本研究OC/EC季节变化呈现夏季(4.2±0.8)>冬季(3.8±0.9)>秋季(3.8±0.5)>春季(3.7±0.9)的变化趋势,说明花溪城区各

季节中都存在SOC,即使冬季的光化学反应较弱也均存在SOC.O₃浓度的增加可能会导致大气氧化能力的增强^[51],从而影响SOC浓度水平^[52].由图2可见,SOC与OC显著相关($R^2=0.9$)且在总氧化剂($\text{O}_x=\text{O}_3+\text{NO}_2$)浓度增加的情况下,SOC浓度总体上也随之增加,表明大气氧化能力增强,OC部分参与二次转化,并且O₃作为前体物参与碳组分的形成和转化^[53].

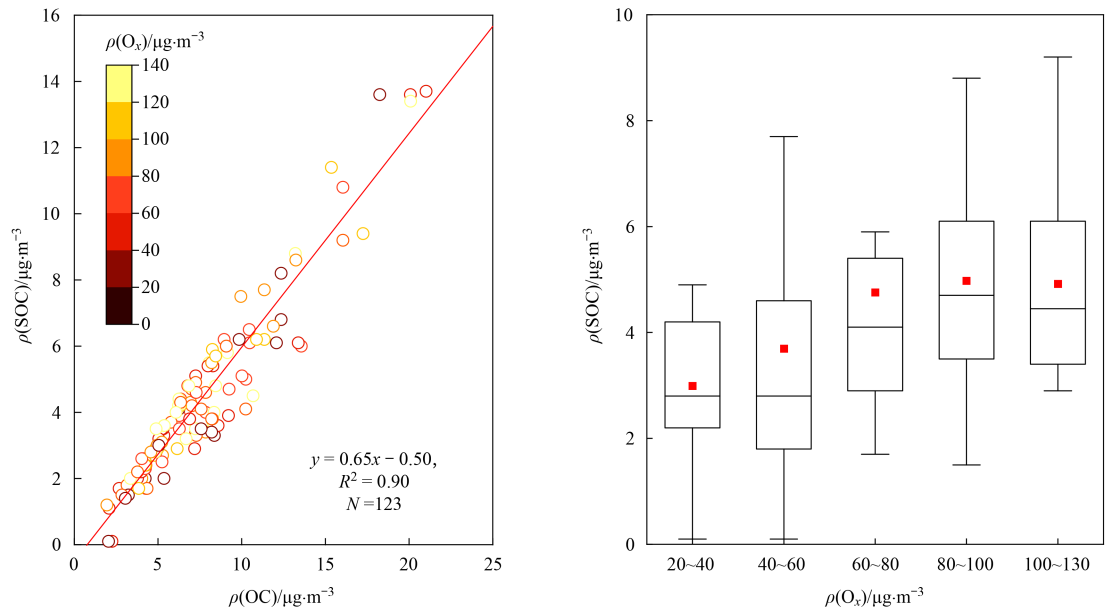


图 2 SOC与OC相关性和SOC随O_x浓度水平的变化

Fig. 2 Correlation between SOC and OC, and variation in SOC with different O_x concentration levels

OC是POC和SOC二者之和^[54],当PM_{2.5}中SOC二次转化增加,SOC/PM_{2.5}增大,POC/PM_{2.5}随之降低.本研究中, $\rho(\text{SOC})$ 平均值为(4.4±2.6) μg·m⁻³,低于乌鲁木齐(8.1 μg·m⁻³)^[32]、郑州(6.0 μg·m⁻³)^[55]、北京

[(5.4±5.8) μg·m⁻³]^[19]和重庆(11.7 μg·m⁻³)^[56]等国内重点城市,高于匈牙利布达佩斯郊区(1.51 μg·m⁻³)、印度昌迪加尔地区(4.0 μg·m⁻³)和泰国清迈(2.88 μg·m⁻³)等地区^[57-59].由图3可知, $\rho(\text{SOC})$ 在春、夏、

秋和冬季的平均值分别为: (4.1 ± 1.5) 、 (3.7 ± 1.2) 、 (3.7 ± 2.4) 和 $(6.1 \pm 3.9) \mu\text{g} \cdot \text{m}^{-3}$, 对 $\text{PM}_{2.5}$ 的贡献率分别为: $(9.3 \pm 1.9)\%$ 、 $(14.5 \pm 2.6)\%$ 、 $(10.2 \pm 2.1)\%$ 和 $(11.1 \pm 3.8)\%$. 冬季 SOC 浓度平均值明显高于其他 3 个季节, 主要原因可能是由于冬季增加的生物质燃烧和煤炭燃烧排放导致 SOC 前体物积累^[60], 且低温

高湿的气象条件导致了大气污染物质的长久停留, 促进了 SOC 的形成, 进一步导致 SOC 浓度升高. 尽管夏季 SOC 浓度最低, 但其在 $\text{PM}_{2.5}$ 中的占比却最高, 这可能是夏季 $\text{PM}_{2.5}$ 质量浓度低, 且较高的温度和较强的光化学作用促进了 VOCs 向 SOC 的化学生成^[61-63].

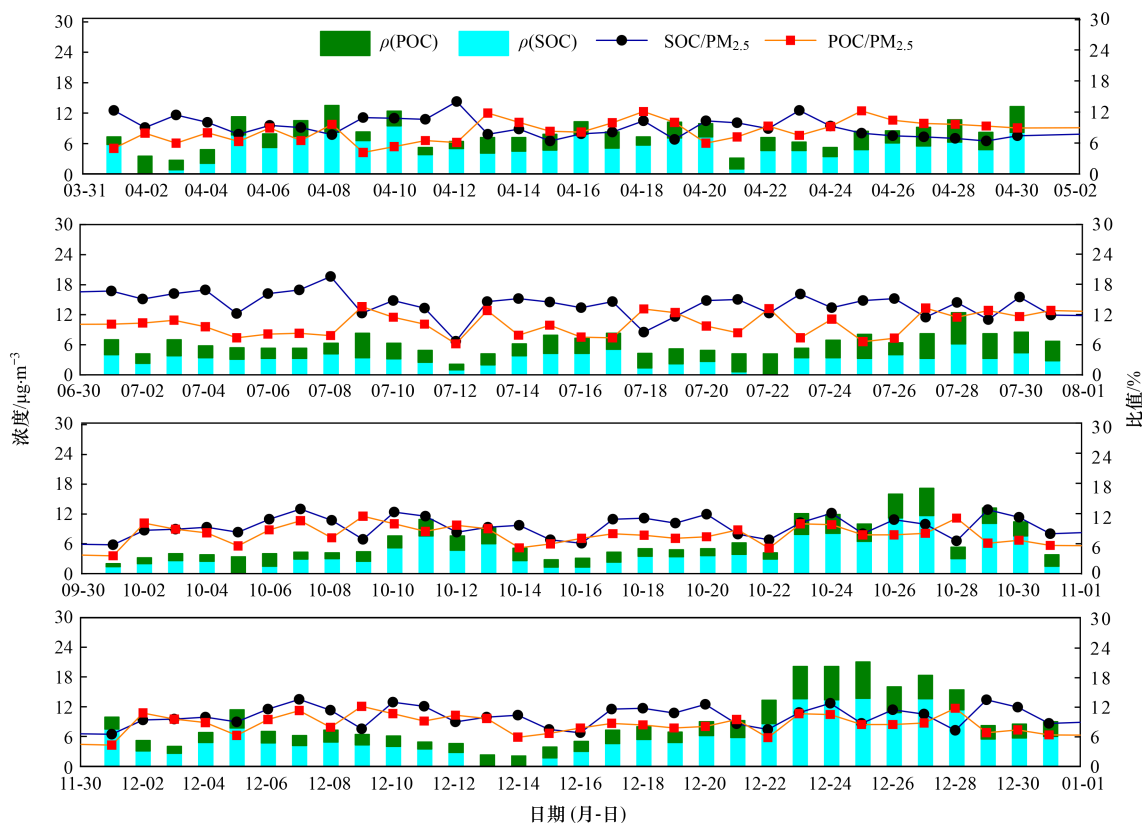


图3 贵阳市花溪城区不同季节 POC、SOC、POC/ $\text{PM}_{2.5}$ 和 SOC/ $\text{PM}_{2.5}$ 的逐日变化

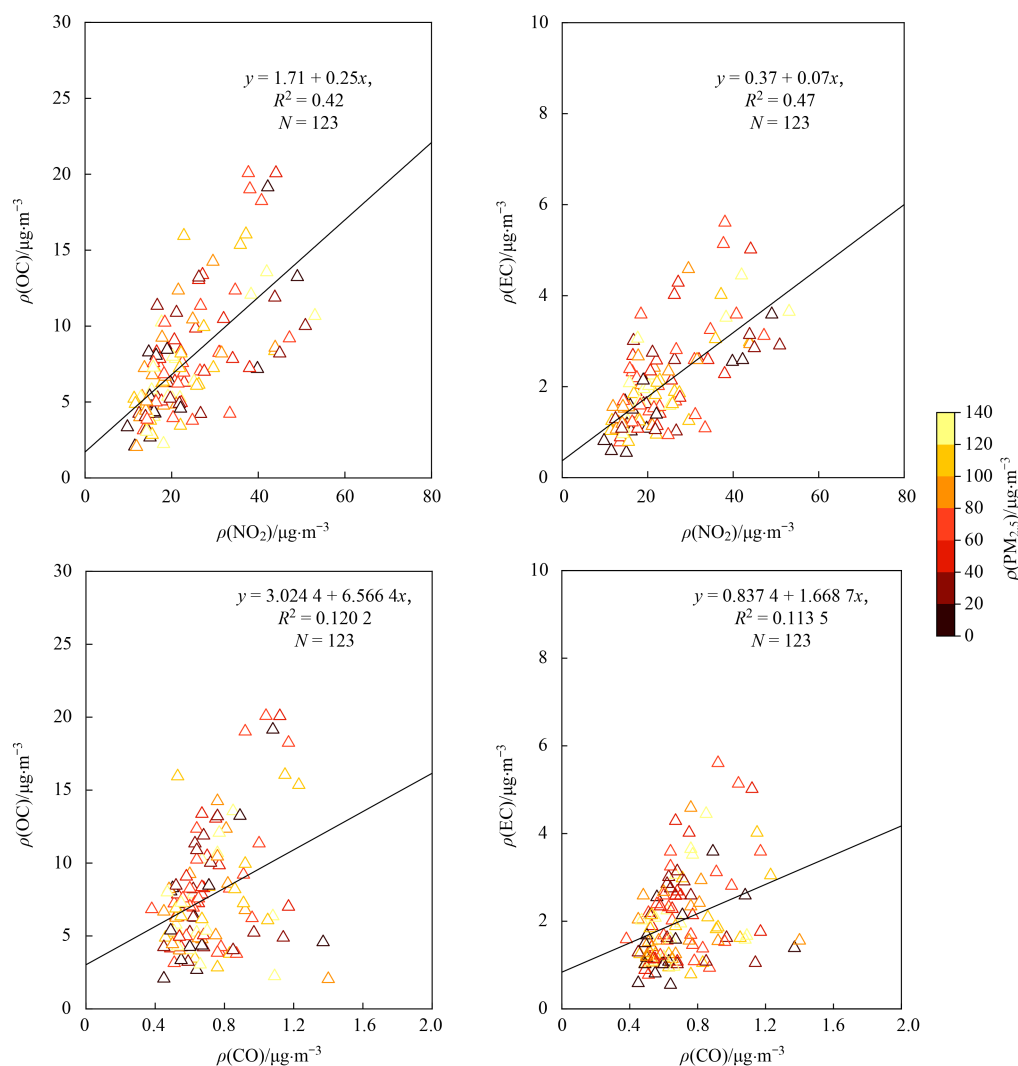
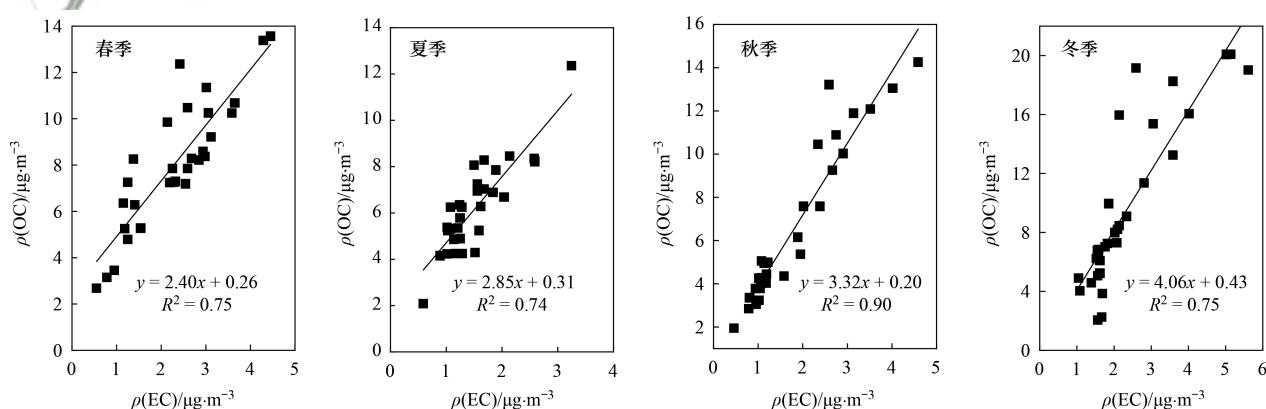
Fig. 3 Daily variation in POC, SOC, POC/ $\text{PM}_{2.5}$, and SOC/ $\text{PM}_{2.5}$ during different seasons in Huaxi District, Guiyang

2.4 碳质气溶胶来源分析

OC/EC 值通常作为燃烧相关源的指标, 由此可定性判断碳质气溶胶的来源. 当 OC/EC 的值在 1.0 ~ 4.2 时, 表明有柴油和汽油车的尾气排放; 在 2.5 ~ 10.5 时, 表明有燃煤排放^[64]; 比值为 4.1 ~ 14.5 或 16.8 ~ 40.0 时, 表明有生物质燃烧^[65,66]. 由图 1 可知, 本研究 OC/EC 值在 1.3 ~ 6.0 之间, 最高值出现在春季 4 月 9 日 (6.0), 最低值出现在冬季 12 月 14 日 (1.3), 各季节 OC/EC 差异显著, 呈现夏季最大, 春季最小的趋势. 结合花溪城区不同季节 OC/EC 的变化范围及均值为: 2.8 ~ 6.0 (3.7) (春季)、2.8 ~ 5.8 (4.2) (夏季)、2.7 ~ 5.1 (3.8) (秋季) 和 1.3 ~ 5.3 (3.8) (冬季), 通过 OC/EC 值范围初步判断碳质气溶胶来源于机动车尾气排放、燃煤排放和生物质燃烧排放.

OC 和 EC 的相关性在一定程度上可以评价二者的同源性, 如果 OC 和 EC 相关性好, 说明 OC 和 EC 大

部分可能来自于相似或一致的污染源, OC 主要为 POC; 反之, 两者的来源差异很大或具有二次污染^[67,68]. 一般而言, 汽油车排放的 CO 多于 NO_x , 而柴油车排放的 NO_x 多于 CO^[69]. 在观测期间, OC 或 EC 与 CO 和 NO_2 之间存在一定的相关性, 表明碳质气溶胶来源与车辆尾气排放有关. 由图 4 可知, OC 或 EC 与 NO_2 之间存在中度相关性 ($R^2 > 0.42$), 进一步表明 OC 和 EC 的来源可能与柴油车辆有关. 花溪城区不同季节 $\text{PM}_{2.5}$ 中 OC 和 EC 相关性表现为秋季较高, 而春夏冬季相似的特点 (见图 5), 与殷丽娜对南京市的研究结果类似^[29]. 秋季 ($R^2 = 0.9$) $\text{PM}_{2.5}$ 中 OC 与 EC 相关性较好, SOC 对 OC 的贡献相对较低, 说明 OC 与 EC 有共同的污染来源, 这与秋季 OC 主要为 POC 的结论相一致^[70], 主要来源于机动车排放. 春夏季 ($R^2 > 0.74$) 的 OC 与 EC 之间的线性拟合斜率明显低于秋季, 表明花溪区在春夏季存在着多种排放源对碳质气溶胶的贡献, 除机动车尾气外, 燃煤燃气和生物质燃烧的排放

图4 OC和EC与NO₂和CO的相关性Fig. 4 Correlation of OC and EC with NO₂ and CO图5 PM_{2.5}中OC与EC的相关性Fig. 5 Correlation between OC and EC in PM_{2.5}

也会对碳质气溶胶贡献,而夏季相关性较差主要是受光化学反应的影响,挥发性有机物形成SOC所导致的^[39]。

采集的样品利用热/光学碳分析仪和 IMPROVE-A(TOR)分析方法对大气 PM_{2.5} 中的碳组分进行分析,最后获得 8 种碳组分(OC1、OC2、OC3、OC4、EC1、

EC2、EC3 和 OPC)。有研究表明^[61,71~73],生物质燃烧后各组分中 OC1 含量最多,燃煤排放各碳组分中 OC2 含量最多,OC2、OC3 和 OC4 也代表煤燃烧排放,EC1 和 OPC 为汽油车尾气的特征组分,EC2 和 EC3 是柴油车尾气排放组分,可由各碳组分的含量占比对碳质气溶胶的来源进行初步分析判断。图 6 展示了贵阳

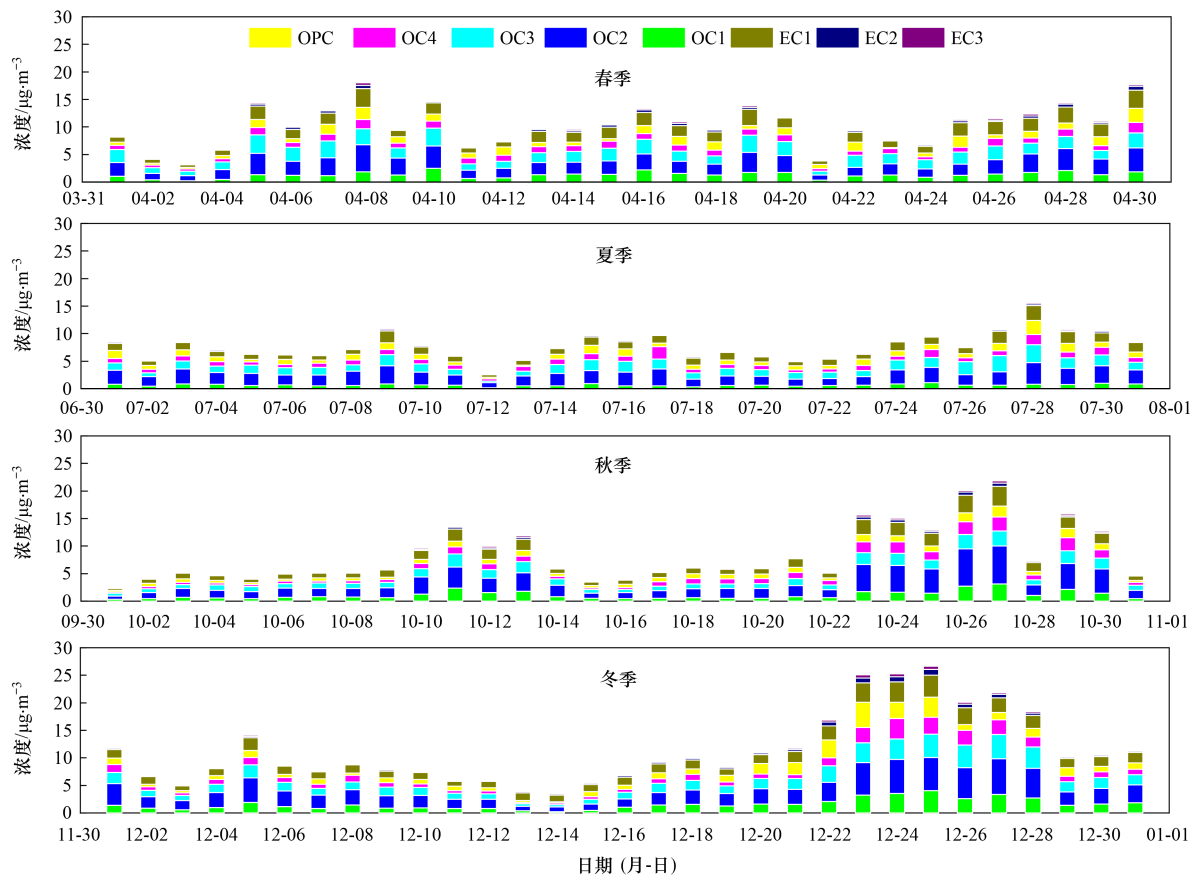


图6 贵阳市花溪城区不同季节碳质气溶胶中各组分的逐日浓度变化

Fig. 6 Daily concentration variation of each component in carbon aerosols during different seasons in Huaxi District, Guiyang

市花溪城区不同季节8种碳组分的逐日浓度变化。观测期间内,各碳质组分浓度平均值分别为:OC2 ($2.62 \mu\text{g}\cdot\text{m}^{-3}$) > OC3 ($1.72 \mu\text{g}\cdot\text{m}^{-3}$) > EC1 ($1.60 \mu\text{g}\cdot\text{m}^{-3}$) > OC1 ($1.15 \mu\text{g}\cdot\text{m}^{-3}$) > OPC ($1.04 \mu\text{g}\cdot\text{m}^{-3}$) > OC4 ($1.02 \mu\text{g}\cdot\text{m}^{-3}$) > EC2 ($0.25 \mu\text{g}\cdot\text{m}^{-3}$) > EC3 ($0.15 \mu\text{g}\cdot\text{m}^{-3}$),其中OC2浓度最高,表明与燃煤源排放相关。但每种组分在不同的季节表现出的浓度水平相差各异,其中春季各组分逐日变化平稳,而秋冬季变化幅度较大,冬季12月1~12日各组分变化保持平稳,而后呈现各组分逐渐增加,与静风、高相对湿度和较低的温度有关。

从不同季节8种碳组分在(OC+EC)中的贡献率可知(图7),不同季节各组分贡献较高的分别为OC2、OC3和EC1,三者总和对春、夏、秋和冬季的贡献率分别为62.4%、64.5%、62.3%和60.1%,表明碳质气溶胶来源主要为机动车汽油排放和燃煤排放。其余5种碳组分OC1、OC4、OPC、EC2和EC3的含量较小,其中(EC1+OPC)和(EC2+EC3)变化范围分别为26.1%~28.4%和3.4%~4.6%,故受机动车排放源(尤其是汽油车尾气排放)影响相对较大。从图7中发现贵阳市花溪城区OC1在夏季的含量较其他3个季节低,这说明夏季生物质燃

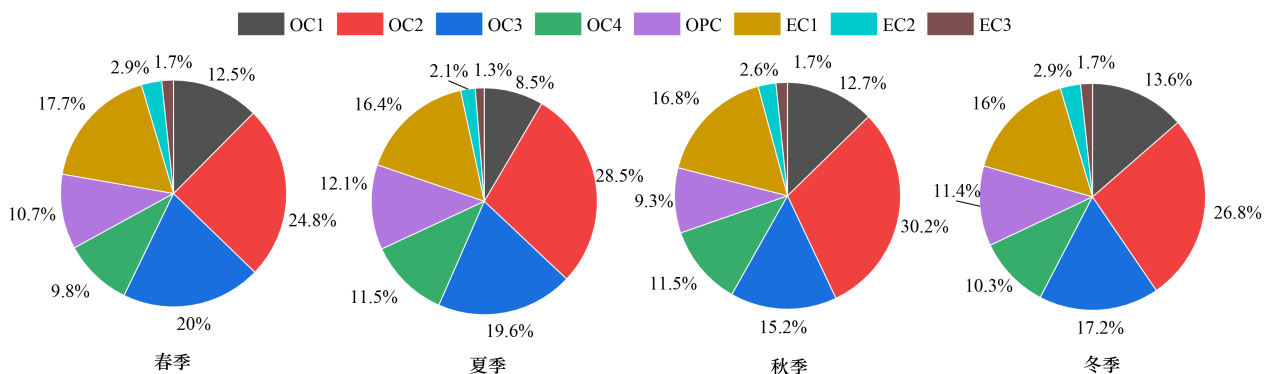


图7 贵阳市花溪城区不同季节不同碳组分对OC+EC的贡献率

Fig. 7 Contribution rates of different carbon components to OC+EC in different seasons in Huaxi District, Guiyang

烧较少. 综上所述, 通过对各季节大气 PM_{2.5} 中 8 种碳组分的对比分析, 可以得知: OC1、OC2、OC3 和 EC1 的含量在大气 PM_{2.5} 中较高, 表明贵阳市花溪城区碳质气溶胶主要来源于燃煤、机动车尾气和生物质燃烧, 这与 OC/EC 比值法初步判断污染源一致.

为了进一步定量解析主要排放源对碳质气溶胶的贡献, 本研究利用 EPA PMF 5.0 模型对采样期间贵阳市花溪城区大气 PM_{2.5} 中的 8 种碳组分进行来源解析, 根据各类碳组分在不同污染源排放中的含量以及物种贡献率, 以期判断来源和贡献. 通过来源解析

结果发现(见图 8), 因子 1 中 EC2 和 EC3 是主要贡献组分, 贡献率为 50.8% 和 71.9%, 为机动车尾气排放. 因子 2 中高载荷组分为 OC2、OC3、OC4 和 OPC, 贡献率分别为 31.6%、33.9%、40.2% 和 63.4%, 是煤燃烧的标志性组分^[74], 故因子 2 为燃煤源. 因子 3 中 OC1 是生物质燃烧样品中丰富的碳组分, 其贡献率为 64.0%, 该特征表征生物质源占主导的组分, 因此因子 3 为生物质燃烧源. PMF 源解析表明, 机动车源、燃煤源和生物质燃烧源是贵阳市花溪城区大气 PM_{2.5} 中碳质气溶胶的主要来源, 其贡献率分别为 21.5%、29.3% 和 49.2%.

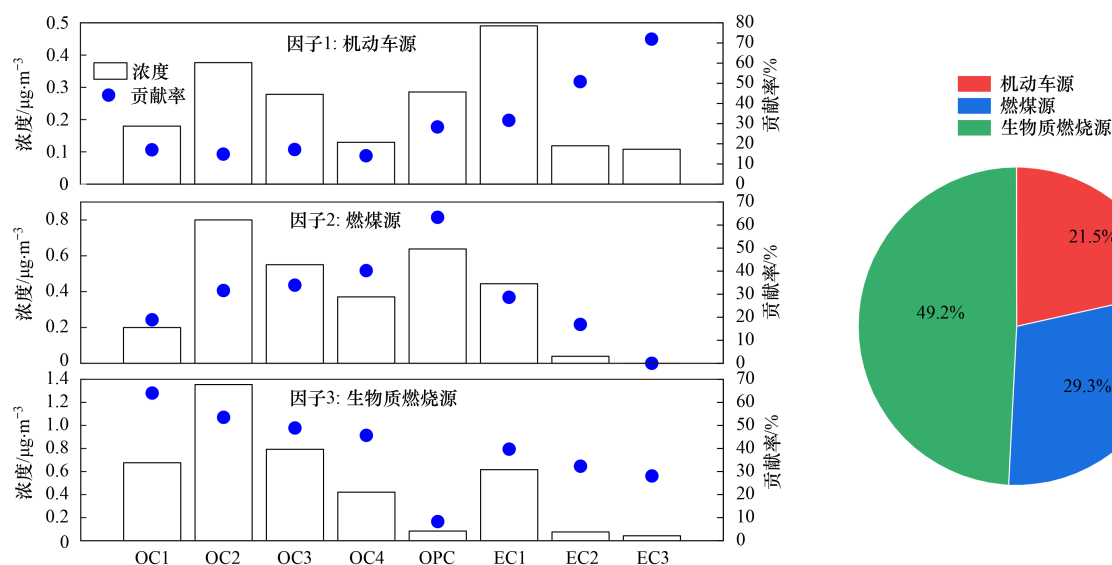


图 8 贵阳市花溪城区观测期间 PM_{2.5} 中 OC 和 EC 组分的 PMF 因子的浓度和贡献率

Fig. 8 PMF source factor profiles for the OC and EC components of PM_{2.5} samples throughout the observation period in Huaxi District of Guiyang in terms of concentrations and percentages

3 结论

(1) 观测期间贵阳市花溪城区 ρ (PM_{2.5})、 ρ (TCA)、 ρ (OC)、 ρ (SOC) 和 ρ (EC) 分别为: (39.7±22.3)、(14.1±7.2)、(7.6±3.9)、(4.4±2.6) 和 (2.0±1.0) $\mu\text{g}\cdot\text{m}^{-3}$, OC/EC 的平均值为 3.9±0.8. PM_{2.5}、TCA、OC、SOC 和 EC 浓度呈现冬季最高、春秋季节次之、夏季最低的季节变化特征.

(2) OC/EC 季节变化呈现夏季 (4.2±0.8) > 冬季 (3.8±0.9) > 秋季 (3.8±0.5) > 春季 (3.7±0.9) 的变化特征, 表明花溪城区各季节均存在 SOC 生成. SOC 与 OC 呈现显著相关 ($R^2=0.9$), 且随着大气氧化性增强, SOC 浓度呈增加趋势. ρ (SOC) 季节变化呈现冬季 [(6.1±3.8) $\mu\text{g}\cdot\text{m}^{-3}$] > 春季 [(4.1±1.5) $\mu\text{g}\cdot\text{m}^{-3}$] > 秋季 [(3.7±2.4) $\mu\text{g}\cdot\text{m}^{-3}$] ≈ 夏季 [(3.7±1.2) $\mu\text{g}\cdot\text{m}^{-3}$] 的变化特征, 但 SOC/PM_{2.5} 呈现夏季显著高于其他 3 个季节.

(3) OC 与 EC 各季节均呈现较好相关性, 其中秋季最高 ($R^2=0.9$), 其他 3 个季节偏低 (R^2 为 0.74 ~

0.75), 表明二者具有共同来源. 利用 PMF 模型对碳质气溶胶来源的定量解析表明, 贵阳市花溪城区碳质气溶胶主要来源为燃煤源 (29.3%)、机动车排放源 (21.5%) 和生物质燃烧源 (49.2%).

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