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重庆典型城区冬季碳质气溶胶的污染特征及来源解析

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摘要:于2021年1~2月在重庆典型城区万州区(WZ)、渝北区(YB)和双桥区(SQ)同步采集 PM2.5样品,分析冬季碳质气溶胶的污染特征、来源及潜在源区.结果表明,观测期间 SQ的 $\rho(PM_{2.5})$ 、 $\rho(OC)$ 和 $\rho(EC)$ 均值分别为(72.6±33.3)、(18.2±8.2)和(4.4±1.7) μ g·m⁻³,高于 WZ[(67.2±30.3)、(17.2±7.4)和(5.1±2.4) μ g·m⁻³]和 YB[(63.4±25.7)、(15.4±6.3)和(4.2±1.9) μ g·m⁻³].与清洁日相比,WZ 污染日 EC 浓度及其对总碳的贡献率均涨幅最大(103.0% 和 8.1%),但 OC/EC 值下降最明显(-10.5%),表明 WZ 污染日碳质气溶胶的一次排放明显增强.观测期间 SQ和 YB的 $\rho(SOC)$ 均值分别为(7.7±4.8) μ g·m⁻³和(6.9±2.8) μ g·m⁻³,明显高于 WZ[(4.5±1.9) μ g·m⁻³],表明二次转化对 SQ和 YB碳质气溶胶的影响相对较大. 此外,与 WZ 不同,SQ和 YB的 SOC/OC 值整体随 PM2.5浓度上升而增大,且 SOC 浓度与气溶胶液态水含量(AWC)、NO2浓度和 NOR 值等均显著线性相关(P<0.01),表明通过液相反应生成含一NO2官能团的 SOC可能是 SQ和 YB碳质气溶胶浓度持续上升的主要因素。正定矩阵因子(PMF)解析结果表明,WZ 的生物质/煤炭燃烧混合源贡献率(47.4%)明显高于 YB(34.2%)和 SQ(38.1%),而 YB 受汽油车排放和二次转化的影响较为突出.浓度权重轨迹分析(GWT)结果表明,各城区污染日碳质气溶胶主要受本地及其东北方向相邻城区(如长寿区)的影响.

关键词:碳质气溶胶;污染过程;二次有机碳;来源解析;重庆

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Characteristics and Source Apportionment of Carbonaceous Aerosols in the Typical Urban Areas in Chongqing During Winter

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Abstract: To investigate the characteristics, source apportionment, and potential source areas of carbonaceous aerosols in Chongqing during winter, $PM_{2.5}$ samples were collected from January 2021 to February 2021 in the urban areas of Wanzhou (WZ), Yubei (YB), and Shuangqiao (SQ). The results showed that the average mass concentrations of $PM_{2.5}$, OC, and EC in SQ were (72.6 ± 33.3) , (18.2 ± 8.2) , and (4.4 ± 1.7) $\mu g \cdot m^{-3}$, respectively, higher than those in WZ [(67.2 ± 30.3) , (17.2 ± 7.4) , and (5.1 ± 2.4) $\mu g \cdot m^{-3}$] and YB [(63.4 ± 25.7) , (15.4 ± 6.3) , and (4.2 ± 1.9) $\mu g \cdot m^{-3}$]. Compared with that during the clear period, the concentration and fraction of EC in total carbon increased by 103.0% and 8.1%, respectively, in WZ compared to that in other areas during pollution period, whereas the OC/EC ratio was decreased significantly (-10.5%), indicating that the primary emission of carbonaceous aerosols increased significantly during the pollution period. The average mass concentrations of secondary organic carbon (SOC) in SQ and YB were (7.7 ± 4.8) $\mu g \cdot m^{-3}$ and (6.9 ± 2.8) $\mu g \cdot m^{-3}$ significantly higher, respectively, than that in WZ [(4.5 ± 1.9) $\mu g \cdot m^{-3}$] during the campaign. This indicated that the secondary transformation had a greater influence on the carbonaceous aerosols in SQ and YB than that in WZ. Furthermore, in contrast to that in WZ, the ratios of SOC/OC were increased with the increase in $PM_{2.5}$ concentrations, and significant correlations between SOC concentrations might be mainly driven by the SOC with $-NO_2$ groups produced by aqueous chemical reactions during winter in SQ and YB. The positive definite matrix factor (PMF) results in these urban areas showed that the contribution of biomass/coal combustion source in WZ (47.4%) was significantly higher than that in YB (34.2%) and SQ (38.1%), whereas the gasoline motor vehicle emission and secondary transformation impacts were more significant in YB. The results of the c

Key words: carbonaceous aerosols; pollution period; secondary organic carbon; source apportionment; Chongqing

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碳质气溶胶是环境大气中PM2.5的重要组成部 分,约占PM_{2.5}浓度的20%~50%^[1,2],主要包括有机碳 (organic carbon, OC)、元素碳(elemental carbon, EC) 和碳酸盐碳(carbonate carbon, CC),其中CC占比通常 小于5%,在研究中一般不予考虑[3,4]. OC由上百种有 机物组成,既包括燃烧源直接排放的一次有机碳 (primary organic carbon, POC),也包括挥发性有机物 (volatile organic compounds, VOCs)等气态前体物通过 复杂化学反应或 POC 经老化反应等生成的二次有机 碳(secondary organic carbon, SOC)[5,6]. EC(通常也称 blank carbon, BC)是以单质状态存在的碳,来源于化 石燃料和生物质燃料的不完全燃烧^[7]. OC 具有散射 和吸收光的作用,EC则是具有强吸光性的物质,两者 直接或间接影响全球和区域气候变化[8,9]. 鉴于碳质 气溶胶对空气质量、气候变化和人体健康的重要影 响,且具有复杂的污染来源,环境大气中碳质气溶胶 的污染特征及来源一直是国内外研究的热点[10,11].

目前,国内外关于碳质气溶胶的研究方向主要 聚焦于污染特征、来源解析和光学特性等方面[5.12~14]. OC和EC是我国环境大气气溶胶的主要污染物,但国 内的相关研究起步较晚,主要集中在京津冀、珠三 角、长三角和汾渭平原等地区,且各地区的污染特征 差异明显[15-18]. Yang 等[15]研究了北京市 2005~2008年 OC和EC的污染特征,发现秋冬季较高的OC和EC浓 度主要受生物质燃烧排放影响. Yao 等 17 研究发现上 海市 2015~2017年冬季 OC 和 EC 浓度逐年下降,主要 受机动车排放的影响. 张颖龙等[18]基于嘉兴市冬季 碳质气溶胶的污染特征研究发现,轻度污染时段POC 对PM,、贡献较大,而中度以上污染时段PM,、主要受 SOC 影响. 此外,早期研究指出我国城市大气霾污染 期间二次有机气溶胶约占有机气溶胶的44%~ 71%[19,20],其中二次有机气溶胶可能主要由液相反应 生成,但相关研究成果主要来源于实验室模拟.

重庆位于我国西南地区,是代表四川盆地大地形的典型山地城市,其大气污染物的排放源复杂、排放强度大;同时,该地区还是我国传统的雾多发区,冬季高湿静稳天气频发.早期研究发现,四川盆地PM_{2.5}污染源贡献及其组分特征与国内其他地区存在较大差异,且重庆PM_{2.5}中碳质组分占比高于其他城市^[21],这可能与山地城市特殊的地形地貌、气象条件和污染源分布等有关.近年来,重庆碳质气溶胶的污染问题引起了研究学者的广泛关注^[22-26].例如,Wang等^[22]研究发现冬季重庆碳质气溶胶浓度以及OC/EC值均明显高于盆地内的成都.文献[23,24]研究了重庆城区夏冬季PM_{2.5}中碳质组分污染特征,发现冬季OC和EC浓度约为夏季的2~3倍,主要来源于冬季

生物质燃烧和机动车尾气排放. Ding 等^[25]研究发现冬季碳质气溶胶浓度高于其他季节,除了受生物质燃烧影响外,还可能与冬季较强的非均相反应有关. Chen 等^[26]基于单颗粒气溶胶质谱仪研究了重庆城区冬季颗粒物污染特征,发现二次老化是颗粒物中OC的重要来源. 以上研究表明,重庆冬季碳质气溶胶的污染来源复杂,污染程度在盆地内相对较重,对PM_{2.5}污染形成和区域气候变化影响明显. 然而,目前关于重庆碳质气溶胶的研究主要集中在单个观测点OC、EC浓度水平和污染来源的季节变化特征等方面,关于山地城市不同碳质组分污染的空间差异性及其对PM_{2.5}污染和二次有机气溶胶形成的研究较少.

本研究以重庆典型城区(渝北区、万州区和双桥区)为研究区域,其中渝北区、万州区和双桥区分别位于重庆中心城区、渝东北地区和渝西地区,同步采集2021年1~2月(2021年冬季)PM2.5样品,分析盆地大地形下典型山地城市冬季不同城区不同碳质组分的污染特征及其对PM2.5污染贡献,初步探讨SOC的形成途径,并基于PMF受体模型、浓度权重轨迹分析等方法解析碳质气溶胶的污染源贡献和污染潜在源区,以期为重庆PM2.5污染的防控治理和深入研究提供宝贵的经验和科学依据.

1 材料与方法

1.1 样品采集

本研究采样地点分别设置在渝北区、万州区和 双桥区 3 个主要城区,采样点位于空气质量监测站附近,具有较强的城区代表性(图1). 位于重庆中心城区的渝北区采样点(YB: 29.60°N,106.37°E)和渝东北地区的万州区采样点(WZ: 30.84°N,106.50°E)周边均以居住区和办公区为主,点位距离城市主干道约50 m. 其中,YB点位地势较为平坦,周边商业写字楼和高层住宅小区相对较多;WZ点位三面环山,周边以老旧小区为主,两个点位的局地污染源排放和

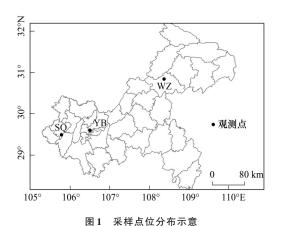


Fig. 1 Distribution of the sampling sites

传输情况存在较大的差异. 位于渝西地区的双桥区 采样点(SQ: 29.49°N, 105.78°E)周边以居住区和工 业区为主,附近汽车装备制造等行业企业较多.各点 位同步采样时间为2021年1月21日至2月20日,采 样频率为逐日(每天采样23h),采样仪器为Comde-Derenda PNS 颗粒物采样器,采样流量为 16.7 L·min⁻¹, 采样滤膜为石英纤维滤膜和 Teflon 滤膜 (whatman). 采样前将石英纤维滤膜置于600℃的马 弗炉中焙烧6h,去除残留的碳质组分;采样前后将 Teflon 滤膜置于恒温恒湿条件下(20℃,相对湿度 50%)48 h后称重.此外,本研究在采样期间每10天 随机采集一组全程空白样品,空白样品的采集工作 在当日PM,5样品采集开始之前完成,采集时间为 0 h,其前处理、采样操作和运输保存等环节与 PM,5 样品一致.同期的SO₂、NO₂等常规污染物浓度数据 来自附近空气质量监测站,温度(T)和相对湿度(RH)等气象数据来自重庆市气象局每日发布.

1.2 样品分析

本研究根据 IMPROVE 协议规定的热/光反射法 (thermal/optical reflection, TOR), 采用 DRI Model 2015 多波段热/光碳分析仪分析 PM,5样品中各碳质组分 浓度. 具体分析过程:截取 0.526 cm²的样品滤膜放 入仪器样品舟中进行加热分析,首先在无 O,的纯 He 环境中,依次在140、280、480和580℃下进行加热将 OC1、OC2、OC3和OC4转化为CO;;然后在含2%O,的 He/O₂环境中,分别在580、740和840℃下逐步加热, 将EC1、EC2和EC3转化为CO,,上述CO,均是由各环 节加热释放的碳质组分经 MnO,氧化炉转化生成,最 终由 NDIR 非分散红外检测器定量检测. IMPROVE 协议将 OC 定义为 OC1+OC2+OC3+OC4+OP, EC 定义 为EC1+EC2+EC3-OP, 总碳(TC)定义为OC+EC[27],本 研究采用635 nm 波段的测量数据进行分析,当OP值 为负值时记为 0. 该仪器检出限为 0. 2 μg·m⁻³,精密度 要求为±5%以内,每天测试样品前对仪器进行烘炉 [ρ(TC)≤0.3 μg·m⁻³]、CH₄/He标准气校准(OC3、EC1 和 CH₄三者峰面积的相对标准偏差≤5%). 此外,每10 个样品随机开展1次平行样品分析(相对误差≤5%), 同时测量全程空白并在结果中扣除.PM,5中水溶性 无机离子浓度采用 Dionex-1100 型离子色谱分析仪测 定,分析方法及质控信息见文献[28].

1.3 数据分析

1.3.1 ISORROPIA-II模型

ISORROPIA-II模型可以模拟 Na*-K*-NH₄*-Mg^{2*}-Ca^{2*}-SO₄^{2*}-NO₃*-Cl*-H₂O 气溶胶系统的热力学平衡,并通过"Forward Problem"和"Reverse Problem"两种模式推算处于稳态模式下的颗粒态水溶性离子浓度和气

态酸碱性气体浓度,进而模拟计算气溶胶液态水含量(aerosol water content, AWC)^[29]. 其中,"forward problem"模式以气温、相对湿度以及 NH₃、H₂SO₄、Na、HCl、HNO₃、K、Ca和Mg在气态和颗粒态中的总浓度为输入参数进行模拟计算;而"reverse problem"模式则是以气温、相对湿度和上述各组分在颗粒态中的浓度为输入参数进行模拟计算.鉴于观测期间未获取气态 NH₃、HNO₃和 HCl浓度,本研究以颗粒态水溶性离子浓度、气温和相对湿度为输入参数,采用"Reverse Problem"模式计算获得 AWC值。早期研究发现 AWC值与 RH 正相关,其中 AWC值的模拟误差随 RH 的增大而减小^[30,31],考虑到观测期间各采样点的 RH 日均值较高(>60%),因此本研究模拟计算的 AWC值较为可靠.

1.3.2 SOC的计算

EC来自污染源的一次排放,因此EC示踪法常用于估算一次和二次有机碳含量,本研究基于OC/EC最小值法估算SOC的浓度:

别为有机碳和元素碳的浓度,μg·m⁻³;(OC/EC)_{min}为观测期间OC/EC最小值.

1.3.3 正定矩阵因子分解(PMF)

正定矩阵因子分解(positive matrix factorization, PMF)是一种基于化学组分并结合源特征定量解析污染源贡献的源解析方法,不依赖污染源的结构和数目,避免了因污染源成分谱缺失或不确定性导致的结果偏差^[32]. PMF方法采用最小二乘法解析出非负、非正交的因子:

$$\boldsymbol{X}_{ij} = \sum_{k=1}^{p} \boldsymbol{G}_{ik} \boldsymbol{F}_{kj} + \boldsymbol{E}_{ij}$$
 (2)

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} (E_{ij}/\sigma_{ij})^{2}$$
 (3)

式中,X为 $n \times m$ 的化学组分浓度矩阵,G为 $n \times p$ 的源贡献矩阵,F为 $p \times m$ 源廓线矩阵,E为 $n \times m$ 残差矩阵; X_{ij} 为样品i中组分j的浓度, G_{ik} 为因子k对样品i的贡献($G_{ik} \ge 0$), F_{ij} 为因子k中组分j的浓度($F_{ij} \ge 0$), F_{ij} 为样品i中组分i的浓度与其解析值的残差, G_{ij} 为数据 X_{ij} 的不确定度,其计算方法见文献[33]. 当获得的污染源数量较为合理时,本研究选择Q值最小的拟合结果作为最优解.

1.3.4 浓度权重轨迹分析(CWT)

利用TrajStat软件分析观测期间碳质气溶胶的气团来源及潜在源区^[34]. 后向轨迹分析(HYSPLIT)所用的气象数据由美国国家环境预报中心全球资料同化系统(global data assimilation system)提供. 由于采样开始时间为北京时间23:00,本研究以世界标准时间

(UTC)15:00 为起始时间(间隔 1 h),以地面以上 100 m为起始高度开始往后推算 48 h气团轨迹,并将所有后向轨迹进行聚类分析^[35].浓度权重轨迹分析(CWT)基于污染物浓度和轨迹停留时间,通过引入权重函数,模拟潜在源区污染物的权重浓度数值,进而反映潜在源区的污染程度^[36].本研究将轨迹覆盖区域划分为 $i \times j$ 个网格,网格分辨率为 $0.1^{\circ} \times 0.1^{\circ}$,具体计算公式:

$$CWT_{ij} = \sum_{k=1}^{M} C_{k} \tau_{ijk} / \sum_{k=1}^{M} \tau_{ijk}$$
 (4)

$$WCWT_{ij} = CWT_{ij} \times W_{ij} \tag{5}$$

$$W_{ij} = \begin{cases} 1.00, & 80 > n_{ij}, \\ 0.70, & 20 < n_{ij} \le 80, \\ 0.42, & 10 < n_{ij} \le 20, \\ 0.05, & n_{ii} \le 10. \end{cases}$$
 (6)

式中, C_k 为轨迹 k 经过网格(i,j)时对应的污染物浓度, τ_{ik} 为轨迹 k 在网格(i,j)停留的时间,M 为轨迹总数, $WCWT_{ij}$ 为网格(i,j)上的平均污染权重浓度, n_{ij} 为经过网格(i,j)的所有轨迹数, W_{ij} 为权重函数,用于降低 $WCWT_{ii}$ 的不确定性.

2 结果与讨论

2.1 PM_{2.5}、OC和EC浓度变化特征 观测期间,SQ、WZ和YB的PM_{2.5}及其碳质组分

浓度日变化特征如图 2 所示 . 2021 年冬季 SQ、WZ 和 YB 的 ρ (PM, 5) 均 值 分 别 为 (72.6 ± 33.3)、(67.2 ± 30. 3) 和 (63. 4 ± 25. 7) μg·m⁻³; ρ(OC) 均值分别为 (18.2 ± 8.2) 、 (17.2 ± 7.4) 和 (15.4 ± 6.3) μg·m⁻³,占 PM, ₅浓度的 25.6%、25.3% 和 27.1%; ρ(EC)均值分 别为 (4.4 ± 1.7) 、 (5.1 ± 2.4) 和 (4.2 ± 1.9) µg·m⁻³, 在PM,5中占比为6.6%、6.1%和7.6%(图2和表1). 从空间分布上看,观测期间SQ和WZ的PM,。及其OC 和EC浓度明显高于中心城区YB,表明重庆主城周边 城区的碳质气溶胶污染程度相对较高.此外,YB的 OC和EC浓度变化幅度相对较大(图2),两者最高浓 度与最低浓度的比值分别为7.0和9.0,高于SO(6.8 和 6.0)和 WZ(5.8 和 8.6),可能与各城区排放源和气 象条件差异有关[23-26]. 值得注意的是,YB的 $\rho(OC)$ 和 ρ(EC)均值明显低于 2015 年冬季[(24.2 ± 13.6) $\mu g \cdot m^{-3} \pi (5.9 \pm 3.2) \mu g \cdot m^{-3}]^{[22]} (表 1); 但 WZ 的 \rho (OC)$ 均值略高于2013年冬季[(16.82 ± 6.87)µg·m⁻³][24], 表明在长期治理下重庆中心城区碳质气溶胶浓度明 显下降,但渝东北地区的效果不明显. 与国内主要城 市相比(表1),重庆典型城区冬季OC和EC浓度均高 、上海^[38]、南京^[39]和广州^[40],低于北京^[41]、承 德[42]、西安[43]和宝鸡[44];与国外主要城市相比,高于发 达国家城市,如韩国光州[45]、美国洛杉矶[46]和意大利

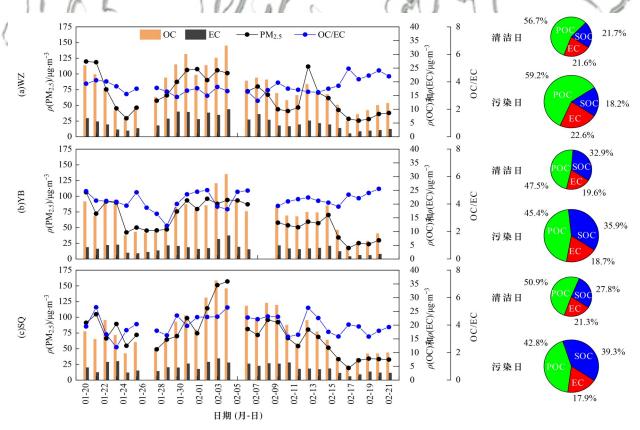


图 2 2021年重庆典型城区冬季 PM_{2.5}、OC 和 EC 浓度时间变化

Fig. 2 $\,$ Temporal variations in $PM_{2.5}$, OC, and EC concentrations during winter in the typical urban areas in Chongqing in 2021

米兰^[47],但明显低于一些发展中国家城市,如印度新德里^[48],这可能主要与各地不同的能源结构有关.

按照我国环境空气质量标准(GB 3095-2012)规定,本研究将污染日定义为 ρ (PM_{2.5})日均值 > 75 μ g·m⁻³,清洁日 ρ (PM_{2.5})日均值 < 75 μ g·m⁻³. 如图 2所示,观测期间 WZ、YB 和 SQ 污染天数分别为 11、11

和 12 d,污染天数占比均超过 38%,各采样点 OC 和 EC浓度变化趋势与 $PM_{2.5}$ 一致 . WZ、YB 和 SQ 污染日 ρ (OC) 日均值分别为(24.9 ± 4.7)、(21.5 ± 4.1)和(24.5 ± 6.9) μ g·m⁻³,约为清洁日的 1.9、1.8 和 1.8 倍; ρ (EC) 均值分别为(7.4 ± 1.8)、(5.0 ± 1.6)和(5.6 ± 1.4) μ g·m⁻³,约为清洁日的 2.0、1.6 和 1.5

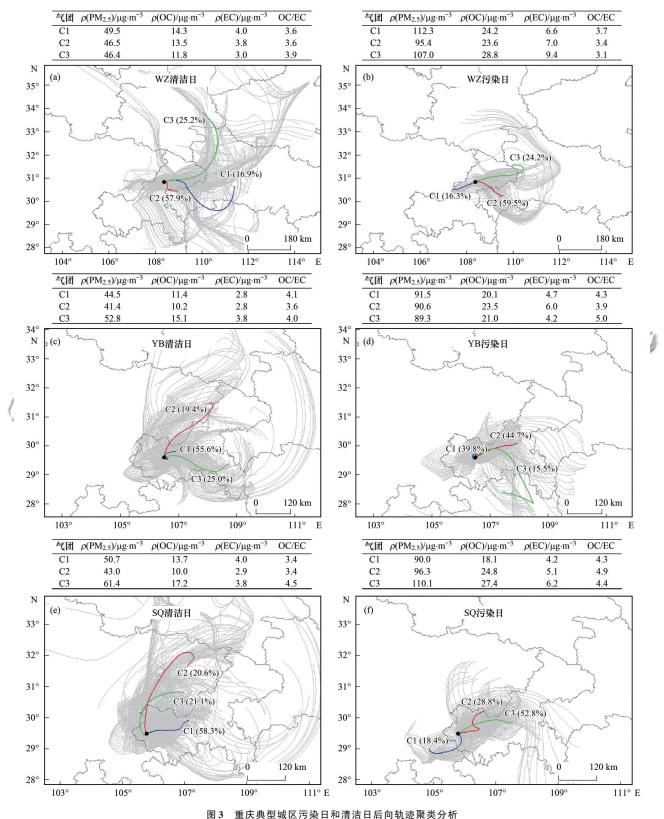


Fig. 3 Cluster analysis of the backward trajectory during the pollution period and clear period in the typical urban areas in Chongqing

倍.与其他城区相比,WZ污染日EC浓度和EC/TC值较清洁日均涨幅最大,但EC/PM_{2.5}降幅最小;此外,与SQ(4.5、3.7)和YB(4.5、4.1)相反,WZ污染日OC/EC值仅为3.4,明显低于清洁日(3.8),表明WZ污染日碳质气溶胶的一次排放明显增强,且排放强度高于其他城区.值得注意的是,各采样点污染日来自东北方向的短距离气团均携带较高的PM_{2.5}、OC和

EC浓度,且 OC/EC 值低于其他气团(图 3),表明污染日较高的 PM_{2.5}及其碳质组分浓度可能主要受点位东北方向的局地排放影响. 其中,WZ 污染日来自东北方向的气团轨迹 3 对应图 3 所有气团轨迹中最高的 $\rho(OC)$ 值和 $\rho(EC)$ 值(28.8 $\mu g \cdot m^{-3}$ 和 9.4 $\mu g \cdot m^{-3}$),以及最低的 OC/EC 值(3.1),表明 WZ 东北方向碳质气溶胶的一次排放强度较大,对站点影响较明显.

表 1 国内外主要城市冬季 $PM_{2.5}$ 、OC 和 EC 浓度对比 $^{1)}/\mu g \cdot m^{-3}$

	Table 1	Comparison of PM _{2.5} , OC, an	d EC concentrations du	ıring winter in dome	stic and foreign c	ities /µg∙m ⁻³	
位置	城市	采样时间(年-月)	$\rho(\mathrm{PM}_{2.5})$	$\rho(OC)$	ho(EC)	方法	文献
	万州		67.2 ± 30.3	17.6 ± 7.4	5.1 ± 2.4		
重庆城区	渝北	2021-01 ~ 2021-02	63.4 ± 25.7	15.4 ± 6.3	4.2 ± 1.9	TOR/IMPROVE-A	本研究
	双桥		72.6 ± 33.3	18.2 ± 8.2	4.4 ± 1.7		
	万州	2013-11 ~ 2013-12	_	16.82 ± 6.87	6.21 ± 2.06	TOR/IMPROVE-A	[24]
成渝地区	渝北	2015-01 ~ 2015-02	115.1 ± 53.9	24.2 ± 13.6	5.9 ± 3.2	TOR/IMPROVE-A	[22]
	成都	2020-12 ~ 2021-02	70.1 ± 36.5	11.1 ± 5.8	4.6 ± 2.5	热燃烧/光吸收法	[37]
	上海	2014-12	_	10.13 ± 6.22	3.06 ± 2.14	TOT/NIOSH	[38]
长三角地区	南京	2017-01	_	8.61 ± 3.46	2.42 ± 1.11	TOT/NIOSH	[39]
珠三角地区	广州	2014-12 ~ 2015-01	63 ± 28	11.6 ± 5.5	5.0 ± 2.5	TOR/IMPROVE-A	[40]
立冲影响反	北京	2016-12	10 -	19.0	5.5	TOT/NIOSH	[41]
京津冀地区	承德	2019-01	47.68 ± 30.37	23.48 ± 11.44	5.06 ± 1.93	TOR/IMPROVE-A	[42]
沙油五臣	西安	2017-01 ~ 2017-02	145.43 ± 96.28	29.02 ± 14.01	6.96 ± 4.16	TOR/IMPROVE-A	[43]
汾渭平原	宝鸡	2018-11 ~ 2019-01	2	18.2 ± 10.4	4.6 ± 3.0	TOR/IMPROVE-A	[44]
	韩国光州	2015-12 ~ 2016-01	35 ± 18	5.0 ± 2.7	1.0 ± 0.4	TOT/NIOSH	[45]
E 44 14 E	印度新德里	里 2015-12 ~ 2016-01	223 ± 69	31.6 ± 12.2	10.5 ± 4.0	TOR/IMPROVE-A	[46]
国外城区	美国洛杉矶	Л 2013-01	10.36 ± 1.09	3.97 ± 0.68	0.51 ± 0.05	TOT/NIOSH	[47]
P 30	意大利米	± 2014-01 ~ 2014-02	10/1	4.62 ± 1.76	0.86 ± 0.36	TOT/NIOSH	[48]

1)"一"表示相关信息未报道

2.2 SOC的污染特征

EC主要来源于一次排放,在大气中相对稳定,因此通常被用来分析OC的一次排放和二次转化^[49].如图4所示,各采样点观测期间OC和EC浓度均显著线性相关(P < 0.01),WZ的相关系数($R^2 = 0.93$)明显高于YB和SQ(R^2 为0.87和0.77),表明OC和EC主要的一次来源相近,其中WZ两者一次污染源的同源性可能强于其他地区.早期研究表明,OC/EC>2则存在二次反应生成的SOC^[50],观测期间SQ、WZ和YB的OC/EC值范围分别为2.4~5.3、2.6~5.0和2.4~5.1,均值分别为3.6±0.5、4.0±0.7和4.3±0.6,表明各采样点每天都有SOC生成.

由表 2 可知,观测期间 WZ、YB 和 SQ 的 ρ (SOC) 均值分别为(4.5 ± 1.9)、(6.9 ± 2.8)和(7.7 ± 4.8) μ g·m⁻³,对 OC的平均贡献率分别为 27.5%、43.4%和 40.0%. 其中,YB 的 SOC/OC 均值高于其他城区,且 SOC浓度仅次于 SQ,表明中心城区冬季二次污染对碳质气溶胶的影响较为显著.污染日 WZ、YB和 SQ

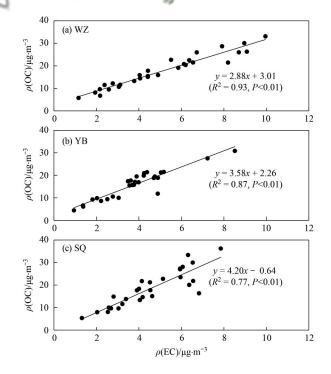


图 4 典型城区冬季 OC和 EC浓度的相关性

Fig. 4 Correlations between OC and EC during winter in the typical urban areas

表 2 典型城区冬季 POC、SOC 浓度以及 SOC/POC、SOC/OC 均值

Table 2 Averages of POC, and SOC concentrations and the ratios of SOC/POC, and SOC/OC during winter in the typical urban areas

75 D		WZ			YB			SQ	
项目	清洁日	污染日	平均值	清洁日	污染日	平均值	清洁日	污染日	平均值
$\rho(SOC)/\mu g \cdot m^{-3}$	3.7 ± 1.1	6.0 ± 2.1	4.5 ± 1.9	5. 1 ± 2. 1	9.5 ± 1.0	6.9 ± 2.8	4.8 ± 2.6	12. 2 ± 3. 7	7.7 ± 4.8
$\rho(POC)/\mu g \cdot m^{-3}$	9. 6 ± 4. 1	19. 4 ± 4.6	13.3 ± 6.4	7.3 ± 3.1	12. 0 ± 3.8	9.2 ± 4.0	8.8 ± 3.4	13.3 ± 3.4	10. 7 ± 4.0
SOC/POC/%	44. 2	33.7	40. 4	75.4	84. 6	79. 1	55.5	94. 9	71.0
SOC/OC/%	29. 3	24. 2	27. 5	42.3	45. 1	43.4	34.7	48. 2	40. 0

的 ρ (SOC)均值分别为(6.0±2.1)、(9.5±1.0)和(12.2±3.7) μ g·m⁻³,较清洁日分别增加0.6倍、0.9倍和1.5倍(表2).此外,本研究还对比分析了3个站点碳质组分对1月28日至2月9日PM_{2.5}污染生消的影响,发现各站点差异明显.如图5所示,WZ的 ρ (PM_{2.5})仅与 ρ (POC)显著正相关(P<0.01);YB的 ρ (PM_{2.5})值则分别随OC/EC、 ρ (SOC)和SOC/OC值的增大而增大;而SQ的 ρ (PM_{2.5})值与一次和二次碳质组分浓度均显著正相关(P<0.01).此外,YB轻度污染日[75 μ g·m⁻³< ρ (PM_{2.5})≤115 μ g·m⁻³]的PM_{2.5}浓度、SOC浓度和SOC/TC值较清洁日分别上涨69.3%、91.1%和118.5%,涨幅显著高于SQ(58.2%、66.6%和22.1%)和WZ(45.5%、23.8%和-31.5%);SQ中度及以上污染日[ρ (PM_{2.5})>115 μ g·m⁻³]的SOC浓度和SOC/TC值较轻度污染日分别

上涨 48.9% 和 12.6%,涨幅低于 $PM_{2.5}$ 浓度(66.2%). 上述结果表明,冬季 SOC 对 YB 的 $PM_{2.5}$ 及其碳质气溶胶浓度上升的影响大于 SQ 和 WZ,尤其是在轻度污染日.

早期研究发现,SOC是由挥发性有机物(VOCs) 在大气中化学转化生成,其中以液态水作为介质发生的液相反应是城市大气气溶胶中SOC生成的重要途径,而气溶胶液态水含量 AWC是影响液相反应速率的关键因素[51.52]. 观测期间,WZ、YB和SQ的AWC值范围分别为 $3.4 \sim 72.1$ 、 $4.6 \sim 76.6$ 和 $3.9 \sim 70.9$ µg·m⁻³,均值分别为 (33.4 ± 21.3) 、 (38.3 ± 18.0) 和(24.7 ± 16.8) µg·m⁻³. 图 6展示了冬季WZ、YB和SQ的SOC浓度与AWC、氮氧化率(NOR)、硫氧化率(SOR)和NO。浓度的相关性,其中NOR和SOR的计算公式见文献[22]. 与WZ不同(P > 0.1),

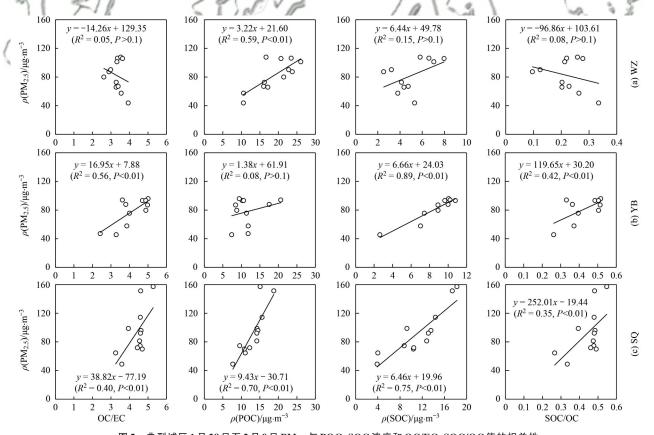


图 5 典型城区 1月 28 日至 2月 9日 PM_{2.5}与 POC、SOC 浓度和 OC/EC、SOC/OC 值的相关性

Fig. 5 Correlations between PM_{2.5} and the concentrations of POC and SOC and the values of OC/EC and SOC/OC from January 28 to February 9 in the typical urban areas

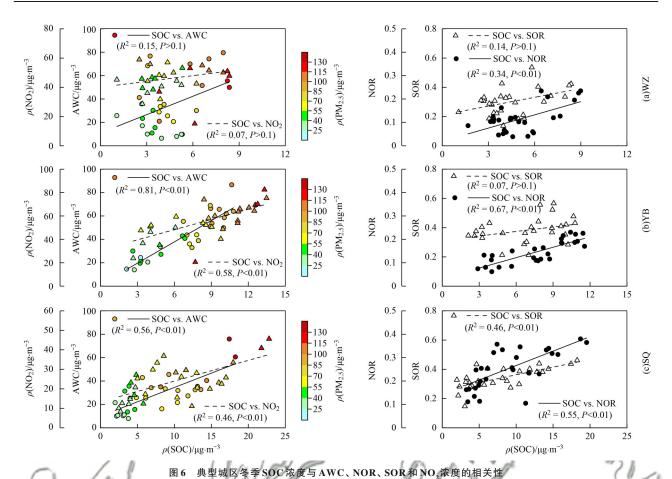


Fig. 6 Correlations between SOC concentrations and the values of AWC, NOR, SOR, and NO_2 concentrations during winter in the typical urban areas

观测期间 SQ和 YB的 SOC浓度与AWC显著线性相 关(P < 0.01),两者的相关系数 R^2 分别为 0.54 和 0.77,表明 SQ 和 YB 气溶胶中 SOC 的主要生成途径 可能是液相反应,而不是光化学反应,这与Bao等[53] 在四川德阳发现的当AWC < 200 μg·m⁻³时液相反 应主导夜间二次有机气溶胶形成的结果相吻合. 值 得注意的是,SQ和YB的SOC浓度与NO2浓度和 NOR 值均显著线性相关(P < 0.01),其中 YB 的相关 系数 $(R^2 = 0.58, 0.67)$ 高于 $SQ(R^2 = 0.46, 0.55)$;此 外,SOC、NO。、AWC浓度和NOR值均随PM。。浓度 的上升而增大(图6). 上述结果表明,在工业和交通 较为发达的SQ和YB,SOC是导致PM25浓度上升的 主要因素,其中通过液相反应生成含一NO。官能团 的有机物可能是SOC的重要组分,且高NO,浓度可 能有利于这类有机物的生成,因此亟需加大工业源 和移动源 NO,排放的管控力度.

2.3 碳质组分含量与来源解析

本研究基于IMPROVE协议规定的热/光反射法测得PM_{2.5}中碳质组分OC1、OC2、OC3、OC4、EC1、EC2和EC3的浓度.早期研究表明,OC1是生物质燃烧排放的特征组分,OC2和OC4常用于示踪燃煤

排放,OC3和EC1在汽油车尾气中含量丰富,EC2和 EC3 则是柴油车排放的标识组分[50]. 此外,本研究 还使用 K+示踪生物质燃烧排放,CF示踪燃煤排 放^[54]. 如图 7 所示, WZ、YB 和 SQ 污染日 OC1 在 TC 中占比分别为12.8%、11.7%和10.2%,均高于清 洁日(11.3%、11.1%和8.8%).与YB和SQ不同, WZ 污染日(OC2+OC4)/TC 值同比清洁日上升 24.5%, 而(OC3+EC1)/TC值同比下降16.7%(图7). 此外,虽然各城区污染日和清洁日(EC2+EC3)/TC 值差异较小(1.1%~1.8%),但WZ污染日EC2和 EC3 总浓度值[(0.54 ± 21.3)µg·m⁻³]显著高于YB 和 SQ(P < 0.01). 值得注意的是, WZ 污染日 K^{+} 和 CI 浓度均值同比清洁日分别上升 62.5% 和 231.2%, 涨幅高于 YB (36.4%和192.9%)和 SQ (31.1%和153.3%);同时,与YB和SQ不同(P> 0.1), WZ 污染日 CI 浓度与 SO, 和 OC2+OC4 浓度均 显著线性相关(P < 0.01),相关系数 R^2 分别为 0.44 和 0.34. 上述结果表明,与 YB 和 SQ 相比,污染日 WZ碳质气溶胶受生物质燃烧、燃煤和柴油车排放 的影响可能相对较大,而受汽油车排放的影响 较小.

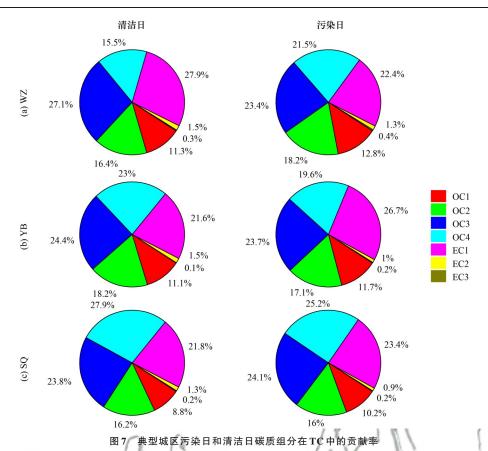


Fig. 7 Mass fraction of carbon components in TC during the pollution period and clear period in the typical urban areas

为进一步识别冬季各典型城区碳质 气溶胶的污 染来源,本研究采用受体源解析模型PMF 5.0基于碳 质组分和水溶性离子组分浓度定量判定污染源排放 因子,定量解析气溶胶中TC的污染源贡献,源解析结 果如图 8 所示. 因子 1 中 OC2、OC4、Na⁺、K⁺和 Cl 的 载荷较大,可代表生物质/煤炭燃烧混合源;因子2 中EC2、EC3和Ca2+的载荷较大,可代表柴油车排放 源;因子3的主要组分为OC3和EC1,可代表汽油车 排放源;因子4中二次组分NH,*、NO,*和SO,42的载 荷较大,可代表二次转化.如图8所示,WZ的TC污 染源贡献大小依次为生物质/煤炭燃烧混合源 (47.4%)>汽油车排放源(21.6%)>柴油车排放源 (18.1%)>二次转化源(12.9%);YB依次为生物质/ 煤炭燃烧混合源(34.2%)>汽油车排放源 (26.9%) > 二次转化源(23.6%) > 柴油车排放源 (15.3%); SQ 依次为生物质/煤炭燃烧混合源 (38.1%) > 柴油车排放源(23.1%) > 二次转化源 (19.7%) > 汽油车排放源(19.1%). 从空间分布上 看,WZ的生物质/煤炭燃烧混合源贡献率明显高于 YB和SQ;YB的汽油车排放源和二次转化源贡献率 高于WZ和SQ相对较高;而SQ的柴油车排放源贡 献率相对较高. 鉴于PMF源解析方法和OC/EC最小 值法的差异,两种方法获取二次转化对TC的贡献 率有所不同,但该污染源贡献率的空间分布较为一

致(图 2 和图 8). 综上,生物质/煤炭燃烧混合源是 冬季气溶胶中TC的主要污染源,尤其是在渝东北 地区;而中心城区受汽油车排放和二次转化的影响 相对较大,这与上述各城区能源结构的实际情况 相符.

鉴于大气污染物通过气团传输对受体点气溶胶 浓度影响较大,本研究使用CWT方法分析冬季各典 型城区污染日和清洁日的 OC 和 EC 潜在源区,其 WCWT的空间分布如图9所示.与清洁日不同,各典 型城区污染日OC和EC主要潜在源区均分布在城区 本地及东北方向的局部地区. 其中,WZ污染日EC主 要潜在源区为万州区东北部本地区域,贡献浓度在 8.5 μg·m⁻³左右,而 OC 主要潜在源区为梁平区西南 部地区、万州区、云阳县和巫溪县南部地区(> 18 μg·m⁻³);YB污染日EC主要潜在源区为渝北区本地 东北部地区和长寿区,贡献浓度在4.5 μg·m⁻³以上, OC和EC潜在源区的空间分布基本一致,在渝北区与 长寿区交界处、渝北区与涪陵区交界处和长寿区本 地西南部地区的潜在贡献浓度最高(> 20 μg·m⁻³). 与YB相似,SQ污染日OC和EC主要潜在源区一致, 分布在双桥区东北部地区、中心城区南部的大渡口 区、渝北区与长寿区交界处,对OC和EC贡献的浓度 分别在 24 μg·m⁻³以上和 5 μg·m⁻³以上.整体上看,各 城区冬季污染日OC、EC主要潜在源区基本一致,主

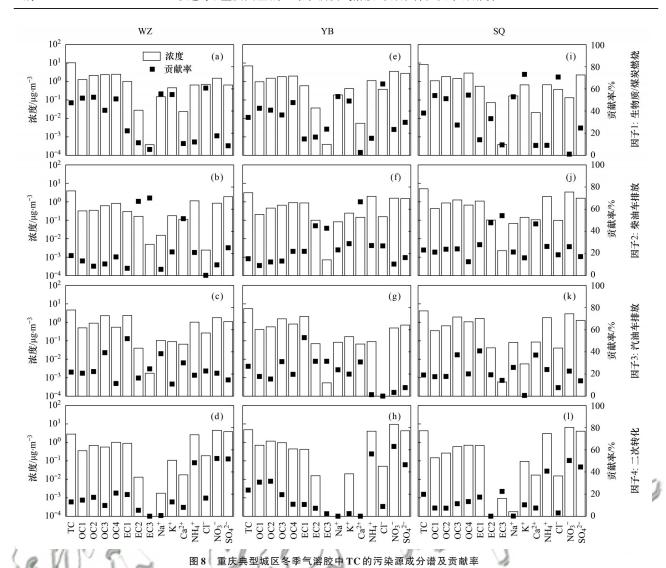


Fig. 8 Source profiles and source contributions of TC in aerosols during winter in the typical urban areas in Chongqing

要分布在本地及其东北方向的相邻城区,表明污染 日碳质气溶胶仍以本地排放为主,同时来自长寿区 的污染传输也需要高度关注.

3 结论

(1)2021年冬季重庆典型城区 WZ、YB和 SQ的 ρ (OC)均值分别为(17.2±7.4)、(15.4±6.3)和(18.2±8.2) μ g·m⁻³, ρ (EC)均值分别为(5.1±2.4)、(4.2±1.9)和(4.4±1.7) μ g·m⁻³,呈重庆主城周边城区高于中心城区的分布趋势.污染日 WZ、YB、SQ的OC和 EC 均值浓度约为清洁日的1.9倍和2.0倍、1.8倍和1.6倍、1.8倍和1.5倍;同比 YB和 SQ,污染日 WZ碳质气溶胶受本地一次排放的影响较为明显.

(2) WZ、YB和SQ冬季 ρ (SOC)均值分别为(4.5±1.9)、(6.9±2.8)和(7.7±4.8)μg·m⁻³,占 OC 的27.5%、43.4%和40.0%.观测期间,SOC是导致YB和SQ的PM_{2.5}及其碳质组分浓度持续上升的主要因素,通过液相反应生成含—NO₂官能团的有机物可能

是SOC的重要组分,高NO₂浓度有利于这类有机物的 生成

(3)碳质气溶胶主要受生物质/煤炭燃烧排放影响,其次是汽油车排放、柴油车排放和二次转化.生物质/煤炭燃烧混合源对WZ碳质气溶胶的贡献率为47.4%,高于YB和SQ;中心城区YB的汽油车排放和二次转化贡献相对突出(26.9%和23.6%);而SQ受柴油车排放影响相对较大(38.1%和23.1%).污染日各城区碳质气溶胶受本地及其东北方向相邻城区的影响,其中长寿区大气污染物的排放对YB和SQ影响较大.

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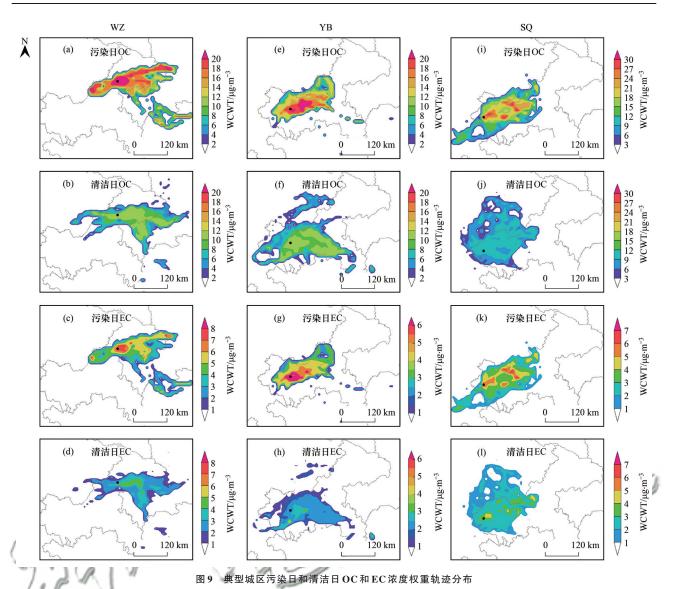


Fig. 9 Weight trajectory distribution of OC and EC concentrations during the pollution period and clear period in the typical urban areas

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