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基于碳减排目标与排放标准约束情景的火电大气污染物减排潜力 李辉,孙雪丽,庞博,朱法华,王圣,晏培



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洛阳市大气细颗粒物化学组分特征及溯源分析

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摘要:为研究洛阳市大气细颗粒物($PM_{2.5}$)的化学组分及来源的时空分布特征,对汾渭平原地区较为欠缺的 $PM_{2.5}$ 相关研究进行补充,在 2018 年 4 月至 2019 年 1 月在洛阳市高新和林校 2 个点位进行了样品采集,对 $\rho(PM_{2.5})$ 、化学组分(水溶性离子、碳质组分、元素)和来源进行分析. 2 个点位的年均 $\rho(PM_{2.5})$ 分别为(76.6 ± 37.9) $\mu_g \cdot m^{-3}$ 和(83.2 ± 38.9) $\mu_g \cdot m^{-3}$,季节变化由高到低均为:冬季、春季、秋季和夏季. 高新和林校的 9 种水溶性离子浓度分别占 $PM_{2.5}$ 的 55.1% 和 54.2%,林校的二次离子(NO_3^- 、 SO_4^{2-} 和 NH_4^+)年均浓度之和高于高新. 高新和林校的 ρ [有机碳(OC)]、 ρ [元素碳(EC)]分别为(12.4 ± 7.7) $\mu_g \cdot m^{-3}$ 、(1.2 ± 0.5) $\mu_g \cdot m^{-3}$ 和(13.4 ± 7.7) $\mu_g \cdot m^{-3}$ 、(1.3 ± 0.5) $\mu_g \cdot m^{-3}$,林校的含碳组分在各季节均高于高新;高新和林校冬季的二次有机碳(SOC)在 OC中质量分数分别为 67.8% 和 77.3%,远高于其他季节.化学质量平衡结果表明,高新和林校的主要贡献源均为二次硝酸盐(26.9% 和 27.1%)、二次硫酸盐(14.5% 和 14.8%)、燃煤(12.6% 和 11.6%)、SOA(10.8% 和 12.2%),高新的生物质源贡献较高,而林校的扬尘源和机动车源贡献较高。后向轨迹和潜在源贡献因子分析表明,洛阳市春季不仅受到来自西北方向的传输,来自西南地区的污染传输也不能忽略;夏季既受到正东方向的季风影响,又有来自正南方向的潜在污染;秋季污染物主要来自东南方向,同时也存在西北方向的潜在来源;冬季受到的传输影响则主要来自周边区域,污染来源较为集中.

关键词: $PM_{2.5}$; 来源解析; 化学质量平衡(CMB)模型; 后向轨迹(HYSPLIT)模型; 聚类分析; 汾渭平原中图分类号: X513 文献标识码: A 文章编号: 0250-3301(2021)12-5624-09 **DOI**: 10.13227/j. hjkx. 202104256

Characteristics of Chemical Composition and Source Apportionment of Atmospheric Fine Particulate Matter in Luoyang

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Abstract: To study the temporal and spatial distribution characteristics of the chemical components and sources of atmospheric fine particulate matter ($PM_{2.5}$) in Luoyang and to supplement the relative lack of $PM_{2.5}$ -related research in the Fen-Wei Plain, samples were collected at two sites (Gaoxin and Linxiao) in Luoyang from April 2018 to January 2019, and the mass concentration, chemical composition (water-soluble ions, carbonaceous components, and elements), and sources of $PM_{2.5}$ were analyzed. The annual average $\rho(PM_{2.5})$ at the two sites were $(76.6 \pm 37.9) \ \mu g \cdot m^{-3}$ and $(83.2 \pm 38.9) \ \mu g \cdot m^{-3}$, respectively. $PM_{2.5}$ showed the highest average concentration in winter, followed by spring and autumn, and the lowest in summer. The concentrations of nine water-soluble ions of Gaoxin and Linxiao accounted for 55. 1% and 54. 2% of $PM_{2.5}$, of which secondary ions $(NO_3^-, SO_4^{2-}, \text{ and }NH_4^+)$ in Linxiao were higher than those in Gaoxin. The annual average concentrations of organic carbon (OC) and elemental carbon (EC) were $(12.4 \pm 7.7) \ \mu g \cdot m^{-3}$ and $(1.2 \pm 0.5) \ \mu g \cdot m^{-3}$ in Gaoxin and $(13.4 \pm 7.7) \ \mu g \cdot m^{-3}$ and $(1.3 \pm 0.5) \ \mu g \cdot m^{-3}$ in Linxiao, respectively, and the average concentrations of carbonaceous constituents during the four seasons in Linxiao were higher than those in Gaoxin. The secondary organic carbon (SOC) of Gaoxin and Linxiao in winter accounted for 67.8% and 77.3% of OC, respectively, which was much higher than that in the other seasons. The results of the chemical mass balance model suggested that the main contribution of $PM_{2.5}$ in the two sites were secondary nitrate $(26.9\% \ and 27.1\%)$, secondary sulfate $(14.5\% \ and 14.8\%)$, coal combustion $(12.6\% \ and 11.6\%)$, and secondary organic aerosol $(10.8\% \ and 12.2\%)$. The contribution of biomass burning was higher in Gaoxin than that in Linxiao, whereas fugitive dust and vehicle emissions contributed more to the $PM_{2.5}$ in Linxiao. The analysis of backward traject

Key words: PM_{2.5}; source apportionment; chemical mass balance (CMB) model; hybrid single-particle Lagrangian integrated trajectory (HYSPLIT) model; cluster analysis; Fen-Wei Plain

近年来,随着社会高速发展,我国频繁发生空气污染现象,大气颗粒物中的细颗粒物(PM_{2.5},即环境空气中空气动力学当量直径小于或等于 2.5 μm 的悬浮颗粒物^[1])已经成为我国大多数城市的首要污染物^[2].有研究发现,PM_{2.5}主要来源于人为及机动车排放、煤炭燃烧、工业生产、建筑及道路扬尘和生

物质燃烧等直接排放源^[3],以及与 SO₂、NO_x、VOCs 和 NH,等气态污染物发生复杂化学反应形成的二

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次排放源[4]. PM,5不仅会造成能见度降低[5]和引起 雾-霾等复合型污染天气的发生,还会对人体健康造 成不利影响,国内外学者发现呼吸道疾病[6]、心血 管疾病[7]和大脑健康[8]等均与人体直接暴露于颗 粒物环境有直接关系. PM25的主要成分包括水溶 性离子、有机碳(organic carbon, OC)、元素碳 (elemental carbon, EC)和无机元素^[9]. 其中,水溶性 离子具有明显的吸湿性,可以直接影响降水的酸 度[10],在PM,5中占有很大的比重,李欢等[11]的研 究证实水溶性离子组分约占 PM25的 30%~80%; 碳质组分对 PM25的贡献通常可以达到 10%~ 50% [12],并且 OC 和 EC 的来源存在异同[13],史芳 天等[14]和张俊峰等[15]的研究根据这一特征,利用 其比值定性分析了成都、北京和石家庄市含碳组分 的来源特征. PM,5的来源分析可以通过受体模型 来实现, Anu 等[16]的研究为探究大气细颗粒物的 源头,利用化学质量平衡模型(chemical mass balance model, CMB) 实现了不同环境受体点的 PM,,源解析: 另外后向轨迹模型(hybrid singleparticle Lagrangian integrated trajectory model, HYSPLIT)可以实现气团输送的轨迹模拟,从而判断 PM_{2.5}的传输来源,段时光等[17]的研究为探究郑州 市的污染物传输来源,利用 HYSPLIT 模拟了郑州市 的气流贡献轨迹,并结合潜在源贡献因子(potential source analysis function, PSCF) 方法得到了 PM2.5污 染的潜在来源. 随着珠三角和长三角地区空气质量 的整体好转,2017年汾渭平原PM25浓度已成为我 国仅次于京津冀地区的第二高区域[18], 2018 年汾 渭平原首次被列为大气污染重点区域[19],污染防治 要求非常紧迫.

目前对于汾渭平原的研究主要集中在西安^[20]、太原^[21]和平顶山^[22]等城市,本文选取洛阳市进行研究,为汾渭平原东部地区的研究补充数据.洛阳市位于河南省西部,整体地势西高东低,境内山川丘陵交错,地形复杂,加上盆地地形不利于污染物的扩散,PM_{2.5}污染严重^[19].目前已有少数对洛阳市的大气污染特征的研究,张攀等^[23]研究了洛阳市 2014年冬季大风天气下 PM₁₀和 PM_{2.5}的变化情况;王群^[24]、周兵利等^[25]和靳朝喜^[26]研究了 2015年洛阳市细颗粒物的变化趋势及含碳组分、重金属等,并进行来源解析和风险评估;齐静文等^[27]研究了 2018年秋冬季 PM_{2.5}中多环芳烃的污染特征和来源.然而,以上研究主要针对细颗粒物中的某一组分或单一季节中的变化,缺乏长期、全面和系统地研究分析.

为深入研究洛阳市大气细颗粒物的化学组分和

污染特征,本文综合探究大气 PM_{2.5}的季节变化,以及水溶性离子和碳质组分的污染特征;使用 CMB模型分析各季节 PM_{2.5}的主要来源,为 PM_{2.5}源解析提供现实依据;并利用 HYSPLIT模型和 PSCF 方法分析各季节的污染传输途径和潜在来源.本研究得到的结果有利于科学认知洛阳市大气颗粒物的组分分布特征和来源,以期为洛阳市大气污染防控决策提供参考.

1 材料与方法

1.1 样品采集

本研究选用了河南省洛阳市华夏路社区服务站 (高新, 112°04′E, 34°06′N, 市区)和河南林业职业 学院(林校, 112°05′E, 34°07′N, 市区, 国控点). 高 新地处居民生活区,周围有周山森林公园、河南科技 大学附属中学和居民小区; 林校位于洛阳市中心地 带,人口密集,车流量较大,集居住、商业、交通和餐 饮于一体,属于城区居住区.2个采样地点相距约 12.5 km,均为典型的城市站点,且周围无工厂排放, 能较好反映城市整体环境空气水平. 采样仪器为武 汉天虹大气颗粒物智能采样仪(TH-16A,流量为 16.7 L·min⁻¹), 4 个通道平行采集 2 张直径 47 mm 的石英滤膜(Whatman 1851-865)和2张直径47 mm 的 Teflon 滤膜(Whatman 7592-104). 采样时间分别 为2018年3月31日至2018年5月2日(春季)、 2018年7月1日至2018年8月6日(夏季)、2018 年10月5日至2018年11月30日(秋季)和2018 年12月1日至2019年1月29日(冬季),每次采样 连续采集23 h. 其中高新共得163组有效数据,林校 共得161组有效数据.

1.2 分析方法

实验通过美国戴安公司的 ICS-900 型(阴离子) 离子色谱仪和 ICS-90 型(阳离子) 离子色谱仪分析 膜样品中的水溶性无机离子(Na^+ 、 NH_4^+ 、 K^+ 、 Mg^{2+} 、 Ca^{2+} 、 F^- 、 Cl^- 、 NO_3^- 和 SO_4^{2-}),测定方法参考文献[28]. 采用碳分析仪(Sunset Lab Inc Model, 美国) 和热光法测定膜样品中 OC 和 EC 组分和含量,并辅助激光透射法校正 OC 和 EC 的切割点[29].

根据洛阳市的实际情况,采用 USEPA CMB 8.2 模型在源类、受体成分谱确定的前提下,通过建立线性方程计算出不同源类贡献值浓度大小^[16],对洛阳市城区各个季节的细颗粒物污染来源进行分析.

使用 TrajStat 进行轨迹聚类,将林校设置为起始点,起始高度为 500 m,起始时间为 00:00、06:00、12:00 和 18:00,计算 48 h 后向轨迹. 将轨迹覆盖区域进行 $0.5^{\circ} \times 0.5^{\circ}$ 网格化,结合日均 $PM_{2.5}$ 浓度进行

分析. 潜在源贡献函数(PSCF)是网格内轨迹超过阈值的个数与总轨迹个数的比值,本研究使用的阈值为环境空气质量标准(GB 3095-2012)中的二级日均标准限值75 μg·m⁻³,为减少 PSCF 的不确定性,引入权重因子得到 WPSCF 值,PSCF 和 WPSCF 的计算参考段时光等^[17]的研究.

1.3 质量控制

本实验所用滤膜无毛刺无破损,采样前后称重均需在平衡室(温度在 $20 \sim 25$ °C之间,相对湿度为 $50\% \pm 5\%$)内平衡 24 h,同一张滤膜的两次称重质量之差小于 0.04 mg. 采样时每 15 d 一个全过程空白滤膜,每 7 d 一个过程空白滤膜,仪器的质量控制参考孙有昌等 $[^{28}]$ 的研究.

2 结果与讨论

2.1 PM2.5及组分特征

洛阳市气象要素和 2 个点位 $PM_{2.5}$ 浓度时空分布如图 1 所示,气象数据来自全球气象网站(www.wunderground.com). 洛阳市高新年均 $\rho(PM_{2.5})$ 为 (76.6±37.9) $\mu g \cdot m^{-3}$,林校年均 $\rho(PM_{2.5})$ 为(83.2±38.9) $\mu g \cdot m^{-3}$,超过环境空气质量标准(GB 3095-2012)二级标准年均限值(35 $\mu g \cdot m^{-3}$)的 1.2倍和 1.4倍. 高新日均 $\rho(PM_{2.5})$ 第 95 百分位数为

140. 5 μ g·m⁻³, 林校日均 ρ (PM_{2.5})第95百分位数为154. 0 μ g·m⁻³, 分别超出环境空气质量标准(GB 3095-2012)二级标准日均限值(75 μ g·m⁻³)的0.9倍和1.1倍. 采样期内, 高新和林校的 ρ (PM_{2.5})变化趋势基本一致, 超标天数分别占总采样天数的44.2%和52.2%, 较2015年细颗粒物超标率(占比60.0%以上)^[24]相对减少, 但该区域大气污染仍较为严重.

2个站点间的 $\rho(PM_{2.5})$ 差异较小,可能由于人口密度相当,所受人类活动的影响相似. 高新和林校的 $\rho(PM_{2.5})$ ($\mu g \cdot m^{-3}$)季节均值变化特征为:冬季(88.1 ± 43.1 和 91.5 ± 41.7) >春季(81.4 ± 34.8 和 91.3 ± 40.6) > 秋季(75.5 ± 36.2 和 83.1 ± 36.8) >夏季(53.2 ± 20.1 和 57.2 ± 21.8),洛阳市PM_{2.5}的季节变化趋势与深圳^[30]和郑州^[31]等相似.春季受沙尘天气影响, $\rho(PM_{2.5})$ 较高,同时该季节为旅游热季,大量游客涌人洛阳市观赏牡丹,导致机动车尾气排放升高,污染加剧.夏季高温多雨,空气质量较好. 秋冬季因采暖期燃煤量增加,导致污染加重,风速低,降水减少,逆温现象频繁,不利于颗粒物扩散.

洛阳市高新和林校的 9 种水溶性离子浓度变化 范围分别为 $6.9 \sim 121.8~\mu g \cdot m^{-3}$ 和 $8.2 \sim 117.5$

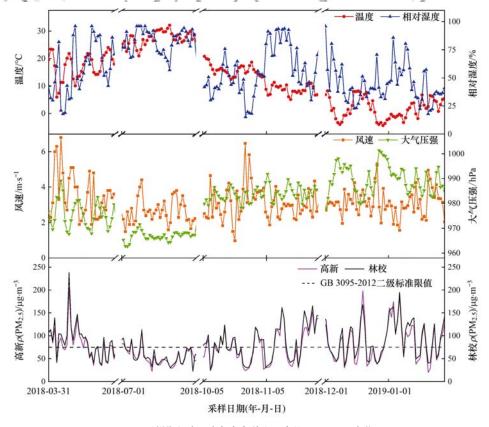


图 1 采样期间洛阳市气象条件和 2 点位 $\rho(PM_{2.5})$ 变化

Fig. 1 Variation in meteorological conditions and $\rho(PM_{2.5})$ at Gaoxin and Linxiao during the sampling period

μg·m⁻³,各占 PM_{2.5}的质量分数为 55.1% 和 54.2%, 该比值高于郑州(52.2%)[31]和武汉(42.0%)[32], 低于石家庄(60.7%)[33]. 从表1可知,高新和林校 的9种水溶性离子组分浓度由大到小均为:NO, > $SO_4^{2-} > NH_4^+ > Cl^- > Ca^{2+} > K^+ > Na^+ > F^- > Mg^{2+}$.

二次离子 NO; 、SO; 和 NH; 是水溶性离子中 的主要成分. 秋季 $\rho(NO_3^-)$ 明显提高且林校高于高 新,表明林校受到交通排放的影响较大[34]. 夏季 $\rho(SO_4^{2-})$ 最高且林校高于高新,可能由于温度高促 进光化学反应生成二次硫酸盐[35];冬季由于采暖 期燃煤和工业燃煤排放大量气态 SO,,导致 $\rho(SO_4^{2-})$ 升高,仅次于夏季. NH_4^+ 是 $PM_{2.5}$ 中重要的 碱性离子,在高温条件下颗粒态 NH,NO。易分解成

气态的硝酸和氨[36],导致 $\rho(NH_4^+)$ 和 $\rho(NO_3^-)$ 在夏 季低; 冬季气温低、大气层结构稳定和 NO, 的排放 增多均有利于 NH4NO, 的稳定存在,增加了 $\rho(NH_4^+)$ 和 $\rho(NO_3^-)$ 的季节差异. 此外, NO_3^-/SO_4^{2-} 在春夏均小于1,说明春夏季大气氨和硫主要贡献 源以固定源[37]为主,秋冬季比值大于1,则以移动 源[37] 为主. 洛阳市的 NO₃ /SO₄ 总比值为 1.67,表 明本地交通源对大气污染的贡献更加突出. 春季地 面尘土易被卷至空中产生沙尘天气导致 $\rho(Ca^{2+})$ 在 该季节最高. K+和 Cl-通常作为生物质燃烧和燃煤 的示踪物[11],浓度季节变化相似,秋季浓度升高可 能来自生物质燃烧,冬季则可能与采暖期燃烧排放 增多有关.

表 1 高新和林校 PM_{2.5}中水溶性离子浓度/µg·m⁻³

Table I	Water-solul	ole ions charac	cteristics of PM ₂	$_{1.5}$ at the sites o	f Gaoxin and I	.inxiao/ μg∙m	_
Na +)	$\rho(\operatorname{NH_4^+})$	$\rho(K^+)$	$\rho(\mathrm{Mg^{2+}})$	$\rho(\operatorname{Ca^{2+}})$	$ ho({ m F}^-)$	ρ(Cl ⁻)	

点位	季节	$\rho(\mathrm{Na^{+}})$	$\rho(\operatorname{NH_4^+})$	$\rho(\mathrm{K^{+}})$	$\rho(\mathrm{Mg^{2+}})$	ho (Ca ²⁺)	$ ho$ (F^-)	ρ(Cl ⁻)	$\rho(\mathrm{NO_3^-}) - \rho(\mathrm{SO_4^{2-}})$
'	春季	0.5 ± 0.5	5.4 ± 3.1	0.5 ± 0.2	0.2 ± 0.2	2. 4 ± 2. 1	0.4 ± 0.3	1.0 ± 0.6	8.8 ± 5.6 9.6 ± 3.2
	夏季	0.3 ± 0.1	8.7 ± 3.6	0.4 ± 0.2	0.0 ± 0.0	0.2 ± 0.1	0.0 ± 0.0	0.4 ± 0.4	8. 7 ± 7 . 6 15. 7 ± 5 . 7
高新	秋季	0.4 ± 0.1	10. 3 ± 6.2	0.8 ± 0.3	0.1 ± 0.0	0.6 ± 0.6	0.2 ± 0.1	1.3 ± 1.0	21.9 ± 14.3 8.3 ± 4.9
	冬季	0.5 ± 0.1	11.7 ± 6.1	1.4 ± 0.9	0.1 ± 0.0	0.8 ± 0.4	0.2 ± 0.2	3.4 ± 1.8	23. 1 \pm 14. 2 10. 5 \pm 5. 9
	全年	0.4 ± 0.2	9.6 ± 5.7	0.9 ± 0.7	0.1 ± 0.1	0.9 ± 1.2	0.2 ± 0.2	1.8/±1.7	17. $7 \pm 13. 8$ 10. $6 \pm 5. 7$
	春季	0.5 ± 0.5	6. 3 ± 3. 6	0.5 ± 0.2	0.2 ± 0.2	2.5 ± 1.9	0.3 ± 0.3	0.9 ± 0.4	10. $2 \pm 6. 8$ 10. $5 \pm 3. 9$
(0)	夏季	0.3 ± 0.1	9.0 ± 3.6	0.4 ± 0.1	0.0 ± 0.0	0.3 ± 0.1	0.0 ± 0.0	0.5 ± 0.3	9.0 ± 6.6 17.2 ± 5.8
林校	秋季	0.4 ± 0.2	11. 1 ± 5. 9	0.9 ± 0.3	0.1 ± 0.1	0.8 ± 0.6	0.2 ± 0.1	1.8 ± 1.2	23.6 ± 13.9 9.0 ± 5.0
	冬季	0.5 ± 0.4	11.6 ± 6.3	1.4 ± 0.9	0.1 ± 0.1	0.9 ± 0.6	0.2 ± 0.1	3.9 ± 1.9	$22.9 \pm 13.9 10.8 \pm 5.9$
	全年	0.4 ± 0.3	10. 1 ± 5. 7	0.9 ± 0.7	0. 1 ± 0. 1	1.0 ± 1.1	0.2 ± 0.2	2. 1 ± 1. 9	18.7 ± 13.5 11.2 ± 5.9

OC 和 EC 是碳在大气 PM,5中的主要存在形 式, EC 主要来自含碳燃料的一次燃烧排放, OC 则 包括直接排放的一次有机碳(primary organic carbon, POC) 和二次反应等途径形成二次有机碳 (secondary organic carbon, SOC) [12]. 洛阳市 PM_{2.5}中 的年均 ρ (OC) 和 ρ (EC) 分别为 (12.9 ± 7.7) $\mu g \cdot m^{-3}$ 和 (1.3 ± 0.5) $\mu g \cdot m^{-3}$, 占 PM, 5 的 16.2% 和 1.6%. 高新和林校的 $\rho(OC)(\mu g \cdot m^{-3})$ 季节变化 较为明显,呈现:冬季(18.7±8.4 和18.9±8.0)> 秋季(11.8±5.6 和 13.4±6.8) > 春季(7.8±2.6 和8.8±3.2) > 夏季(6.1±1.3 和6.9±2.1) 的变 化趋势. 林校的含碳组分在各季节均高于高新,表明 该点位受到机动车尾气排放影响以及污染程度较重 有关.

OC/EC 比值是局地碳质气溶胶排放和区域 SOC 转化的综合结果,常被用来识别碳质气溶胶的 来源[14]. 有研究认为 OC/EC 比值为 2.5~5.0 表示 来自汽车尾气排放[34], 5.0~10.5 表示来自燃煤排 放[34], 16.8~40.0表示来自生物质燃烧排放[38]. 根据 OC/EC 比值范围得到洛阳市春夏季 PM,5中的 碳质组分主要来自汽车尾气排放,而秋冬季的 OC

和 EC 主要来自煤炭燃烧和生物质燃烧. 同时, OC/ EC 比值也可以鉴别颗粒物的二次来源[15],OC/EC 比值大于2时,说明有SOC的生成,比值越大,二次 污染越严重. 高新和林校的 OC/EC 年均值分别为 11.3 和10.7(图2),说明洛阳市的二次污染严重, 高新冬季 OC/EC 比值高于林校,表明高新还受到生 物质燃烧的影响.

由于 SOC 形成机制比较复杂,常用 OC/EC 最

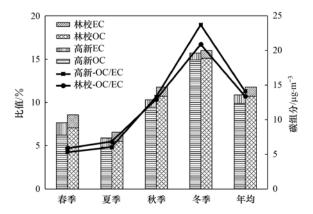


图 2 高新和林校 PM_{2.5} 中碳质组分浓度比较

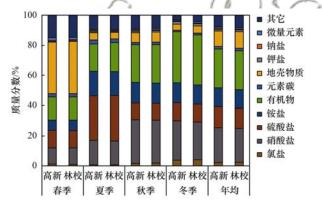
Fig. 2 Carbon component concentrations of PM_{2.5} at Gaoxin and Linxiao

小比值法对 SOC 进行估算,计算公式如下[15]:

SOC = OC - EC × (OC/EC)_{min} (1) 式中,(OC/EC)_{min}为样品中 OC/EC 的最小值,SOC、OC 和 EC 的单位均为 μ g·m⁻³. 计算结果显示,高新和林校年均 ρ (SOC)为7.4 μ g·m⁻³和8.6 μ g·m⁻³,高于郑州(6.0 μ g·m⁻³)[³⁸]和舟山(3.5 μ g·m⁻³)[^{39]};且2点位年均 ρ (SOC)分别占 ρ (OC)的50.0%和55.1%,高于德阳、成都和眉山(47.0%、46.0%和38.0%)[^{14]};冬季高新和林校的 ρ (SOC)/ ρ (OC)高达67.8%和77.3%,高于同季节北京(49.3%)[^{12]}和石家庄(45.0%)[^{15]}. 以上数据表明2个点位的SOC污染严重,其中林校的二次污染更加严重.

2.2 PM, 质量重构及来源解析

化学质量浓度重构可以得到大气颗粒物中的组分污染特征和贡献度,通常用来和 $PM_{2.5}$ 实测值进行比较^[40]. 本研究将 $PM_{2.5}$ 的化学组分分为: 硫酸盐(SO_4^{2-})、硝酸盐(NO_3^{-})、氯盐(Cl^{-})、铵盐(NH_4^{+})、钾盐(K^{+})、元素碳(EC)、有机物($OM=1.4\times$



[OC])、地壳物质 $(1.89 \times [Al] + 2.14 \times [Si] + 1.4 \times [Ca] + 1.43 \times [Fe]$)、微量元素和其他.

图 3 为高新和林校四季的化学组分重构结果, 可以看出 2 点位化学组分变化趋势相似. 春季地壳 物质重构质量浓度和质量分数(29.7 µg·m⁻³, 29.6%)远高于其他季节,说明扬尘源是洛阳市春 季大气污染的主要来源; 夏季硫酸盐的重构质量浓 度和质量分数(16.5 μg·m⁻³, 30.3%)明显增大,可 能是由于高温条件促进了硫酸盐的转化; 秋冬季有 机物和硝酸盐的重构质量浓度和质量分数(48.0 $\mu g \cdot m^{-3}$, 56.3%) 高于春夏季(21.0 $\mu g \cdot m^{-3}$, 31.8%),供暖期燃烧大量的煤炭导致大气颗粒物 排放升高,并且由于大气结构层稳定、气温低,进而 二次转化增多,加剧了大气污染,增加了有机物和硝 酸盐的浓度. 综上所述,二次无机盐(高新36.5%,林 校 36.0%) 和有机物(高新 26.7%, 林校 26.7%) 是 洛阳市大气颗粒物中主要污染物. 因此,洛阳市需要 加强控制硫酸盐和硝酸盐前体物的排放,加快新能 源转型,减少空气污染.

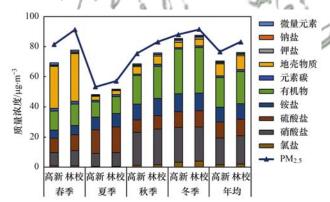


图 3 高新和林校四季的平均重构特征

Fig. 3 Average reconstruction characteristics of four seasons at the sites of Gaoxin and Linxiao

由于 SOC 的源成分谱难以获得,无法通过模型直接解析. 为降低不确定性,采用 OC/EC 最小比值法估算 SOC 浓度,二次有机气溶胶(SOA)以 1.4 × [SOC]计算^[38],将 SOC 和 SOA 从受体成分谱中去除,重新构建受体成分谱. 将采样期间 PM_{2.5}各组分的平均浓度作为 CMB 模型的受体输入数据,将减去SOA 的 PM_{2.5}浓度作为 CMB 模型的 PM_{2.5}浓度输入,得到洛阳市 2018 年采样期间各污染源对环境 PM_{2.5}的贡献率,结果如图 4,可以看出 2 点位源贡献的季节变化趋势相似. 春季由于沙尘天气导致扬尘源贡献最高,其中林校受到扬尘影响(23.8%)高于高新(22.9%);夏季高新(34.2%)和林校(37.1%)均为二次硫酸盐的贡献最高,这可能是由于 NH₄NO₃在高温天气影响下易受热分解,因此二次硝酸盐贡献在夏季最低;秋季主要贡献源相似,均为二次硝

酸盐、SOA和燃煤(高新61.5%,林校63.0%);冬季受供暖影响,高新和林校的燃煤贡献最高,分别为14.4%和13.3%.从不同点位结果来看,林校的二次盐类和SOA污染(54.1%)较高新(52.2%)更为严重,高新的燃煤和工艺过程污染(12.6%和7.2%)则高于林校(11.6%和6.9%),且各季节机动车源在林校贡献均高于高新,该结果与组分特征得出的结论一致.从全年结果来看,洛阳市的二次污染较重,二次硝酸盐和二次硫酸盐的总和在各季节的贡献率均居于首位,SOA在秋冬季的贡献率较高;燃煤、机动车和扬尘的贡献率相当,均在10.0%以上;工艺过程和生物源的贡献相对较低.综合全年污染源占比来看,洛阳市PM,5污染主要为二次污染.

2.3 气团输送路径及潜在源区

以林校(112°05′E, 34°07′N)为起始点,分析洛

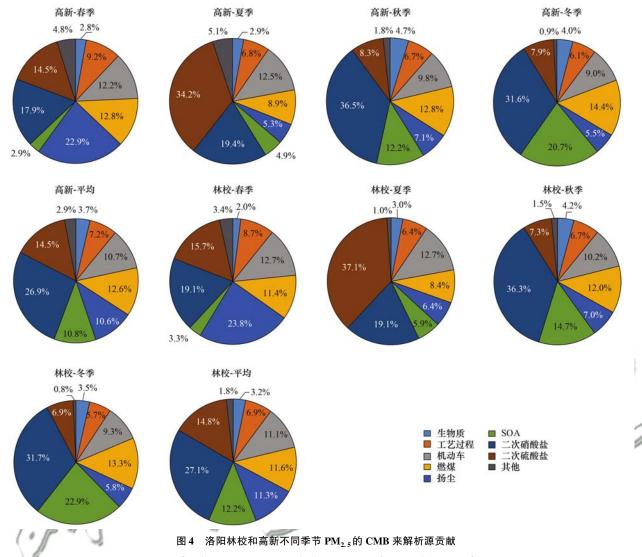


Fig. 4 Results of source apportionment for four seasons at the sites of Gaoxin and Linxiao

阳市 2018 年 4 月至 2019 年 1 月不同气团的污染轨 迹,聚类结果如图 5. 春季有 4 条主要传输途径:轨 迹 1(28.3%) 和轨迹 3(22.5%) 均来自西北方向, 轨迹1起源于陕西省东部,途经山西省西南部到达 洛阳,气团轨迹短、移动速度慢;轨迹3起源于蒙古 国南部,途经内蒙古自治区、陕西省和山西省,传输 距离长且气团移动速度较快. 轨迹 2(27.5%)来自 西南方向; 轨迹 4(21.7%)来自渤海西南部. 夏季 有3条主要传输途径且均为短距离传输,气团移动 缓慢: 轨迹 1 (37.1%) 来自山东南部: 轨迹 2 (32.3%)来自湖南省北部; 轨迹 3(30.7%)来自安 徽省西北部. 秋季有 4 条主要传输途径:轨迹 1 (48.4%)来自东南方向安徽省和河南省交界处,主 要为省内短距离传输. 轨迹 2(25.0%)和轨迹 3 (17.7%)均来自西北方向,轨迹2为长距离传输, 气团移动速度最快;轨迹3为短距离传输,由内蒙 古南部经山西省和陕西省到达河南省. 轨迹 4 (8.9%)来自山东省北部,由河南省东部进入洛阳.

冬季仍主要受到西北方向气团的影响,轨迹 1 (41.9%)和轨迹 3(22.6%)分别为短距离和长距离传输;轨迹 2(26.6%)来自河南南部南阳盆地,气团移动缓慢且途中颗粒物持续积累;轨迹 4 (8.9%)来自北京东南方向,气团移动较快,说明洛阳地区也受到京津冀地区污染物的传输扩散影响.

为了确定各季节污染物的潜在来源,使用PSCF 计算得到 WPSCF 值. 春季 WPSCF 高值区主要为河南省西部、山西省南部、陕西省东部、湖北省西北部和重庆市北部,即洛阳市周边省市地区,其WPSCF 值为 0.4~0.6 之间(图 6),污染物主要来自周边地域及本地源排放. 夏季来源较为分散,主要受到山东省南部和安徽省北部的区域传输影响,其WPSCF值>0.4,显示出夏季主要受季风气候影响,此外湘鄂交界区域也有一定程度的贡献. 秋季主要为河南省中部区域各城市之间的相互影响,以及安徽省西南部和河南省东南部的传输影响,其WPSCF值为 0.5~0.6 之间,这些地区对洛阳市的大气污染

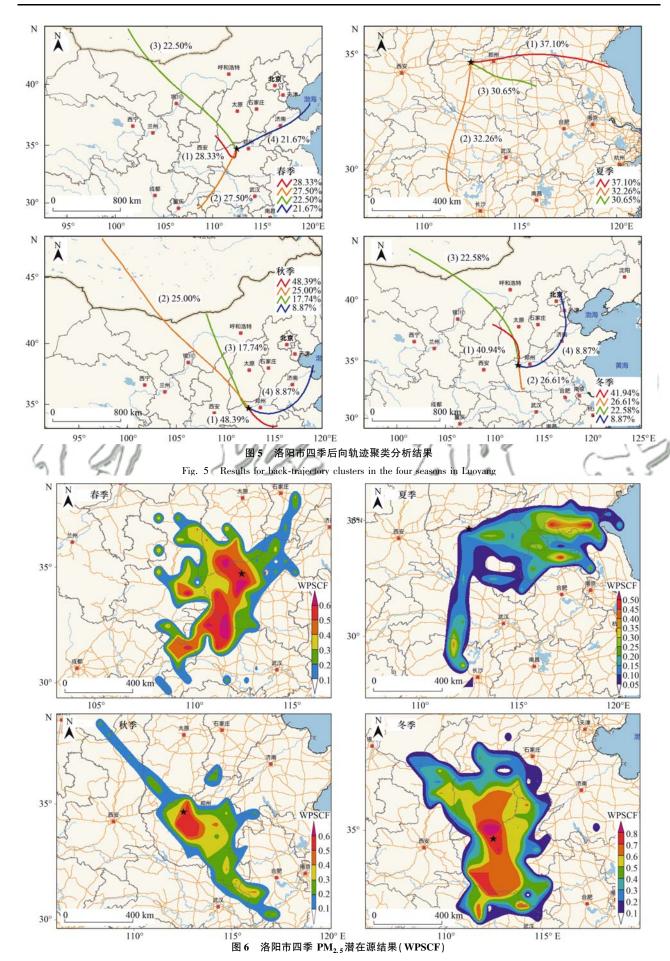


Fig. 6 Results of potential sources (WPSCF) of $\ensuremath{\text{PM}}_{2.5}$ in the four seasons in Luoyang

有较大影响. 冬季 WPSCF 高值区明显增大,主要为洛阳市周边城市,其 WPSCF 值为 0.6~0.8,污染来源集中,冬季不利地形和气象条件使污染物更加不易扩散.

3 结论

- (1)高新和林校年均 $\rho(PM_{2.5})$ 分别为(76.6±37.9) $\mu g \cdot m^{-3}$ 和(83.2±38.9) $\mu g \cdot m^{-3}$,是环境空气质量标准(GB 3095-2012)二级标准年均限值(35 $\mu g \cdot m^{-3}$)的2.2倍和2.4倍.2点位季节平均浓度变化一致,由高到低为:冬季、春季、秋季和夏季.
- (2)高新和林校 9 种水溶性离子浓度变化范围分别为 $6.9 \sim 121.8~\mu g \cdot m^{-3}$ 和 $8.2 \sim 117.5~\mu g \cdot m^{-3}$,各占 $PM_{2.5}$ 的 55.1% 和 54.2%. 林校的含碳组分在各季节均高于高新,且 2 个点位 OC/EC 比值全年均远大于 2,SOC 污染严重.
- (3)基于 CMB 源解析结果,全年林校的机动车源贡献较高,高新的生物质源贡献较高,而秋冬季林校的燃煤源贡献明显高于高新.高新和林校全年主要来源均为二次硫酸盐、二次硝酸盐、燃煤和 SOA.
- (4)根据后向轨迹分析,洛阳市大气 PM₂, 污染在夏季受到正东方向的季风影响显著,其他 3 个季节更多表现为受到周边省市的短距离传输影响. WPSCF 结果表明,除了周围地区的相互影响,PM_{2.5}的主要潜在来源还有陕豫交界、晋豫交界、豫鄂交界、鲁苏交界、皖鄂交界和湘鄂交界.

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