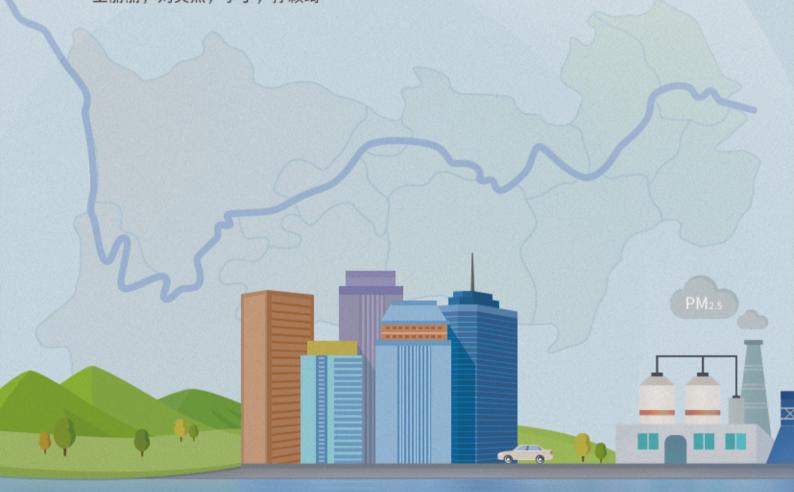




ENVIRONMENTAL SCIENCE

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长江经济带PM2.5空间异质性和驱动因素的地理探测 王丽丽, 刘笑杰, 李丁, 孙颖琦



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	地膜覆盖对农田土壤养分和生态酶计量学特征的影响



华北南部重污染城市周边区域二次气溶胶的化学特征 及来源解析

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摘要:为探究华北南部地区重污染城市邯郸市北部地区冬季大气颗粒物的化学组分及来源,于 2020 年 11 月 23 日至 12 月 12 日采集了 PM_1 和 $PM_{2.5}$ 样品并进行了分析. 观测期间日平均 $\rho(PM_1)$ 和 $\rho(PM_{2.5})$ 分别为 114. 53 $\mu g \cdot m^{-3}$ 和 124. 25 $\mu g \cdot m^{-3}$, $PM_1/PM_{2.5}$ 比值的变化范围为 83. 3% ~ 95. 3%,明显高于京津冀其他城市,表明邯郸地区细颗粒物尤其是亚微米颗粒物污染严重. 与清洁天相比,重污染期间 PM_1 中 $SNA(SO_4^2 \cdot NO_3^-$ 和 NH_4^+)增加 14. 5%, $PM_{2.5}$ 中 SNA 增加 15. 2%,尤其氦氧化率 (NOR) 在重污染天增长 3 倍;随着污染程度的加深, PM_1 和 $PM_{2.5}$ 中二次有机碳(SOC) 占比增加 22. 0% 和 12. 5%,SOC 易在 粒径小的颗粒物中聚集,而 PM_1 中一次有机碳(POC) 和元素碳(EC) 占比下降 15. 4% 和 6. 6%, $PM_{2.5}$ 中 POC 和 EC 占比下降 8. 2% 和 4. 3%.上述结果表明二次形成对颗粒物重污染具有重要贡献. 随着污染程度的加重,颗粒物中液态含水量增加,硫氧 转化率(SOR) 和氦氧转化率(NOR) 均升高,表明液相化学反应对二次无机盐的生成具有重要贡献. 随着污染程度的加深,无机元素呈上升趋势;无机元素中 Se、As、Pb 和 Zn 富集程度较高. 根据主成分分析法(PCA) 源解析结果,二次源、工业源、机动车源和生物质燃烧源是 PM_1 和 $PM_{2.5}$ 主要的来源。潜在源贡献因子分析(PSCF) 结果表明, $SO_4^2 \cdot NO_3^-$ 、EC、OC 和无机元素的高值区域都主要来自观测区域的北方向和西南方向.

关键词:重污染; PM1; PM2.5; 来源解析; 后向轨迹; 潜在源区

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Chemical Characteristics and Sources of Atmospheric Aerosols in the Surrounding District of a Heavily Polluted City in the Southern Part of North China

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Abstract: In order to explore the chemical composition and source profiles of atmospheric particulate matter in winter in the northern area of Handan, a heavily polluted city in the southern part of North China, PM₁ and PM_{2.5} samples were collected and analyzed from November 23 to December 12, 2020. During the observation period, the daily average ρ(PM₁) and ρ(PM_{2.5}) were 114.53 μg·m⁻³ and 124.25 μg·m⁻³, respectively, and the ratio of PM₁/PM_{2.5} was 83.3%-95.3%, which was significantly higher than those of other cities in the Beijing-Tianjin-Hebei region, indicating that air pollution of fine particulate matter, especially sub-micron particulate matter, was more serious in Handan. Compared with that during clean days, SNA (SO₄² , NO₃⁻ , and NH₄⁺) in PM₁ increased by 14.5% during heavy pollution, and SNA in PM_{2.5} increased by 15.2%; the nitrogen oxidation rate (NOR) in particular increased by three times on heavy pollution days. With the deepening of pollution, the proportion of secondary organic carbon (SOC) in PM₁ and PM_{2.5} increased by 22.0% and 12.5%, respectively. SOC tended to accumulate in small particles, whereas the proportion of primary organic carbon (POC) and elemental carbon (EC) in PM₁ decreased by 15.4% and 6.6%, and the POC and EC in PM_{2.5} decreased by 8.2% and 4.3%, respectively. The above results indicated that secondary formation played an important role in the heavy pollution of particulate matter. With the aggravation of air pollution, the liquid water content of the particles increased, and both the sulfur oxidation ratio (SOR) and nitrogen oxidation ratio (NOR) increased, indicating that the aqueous phase chemical reaction made an important contribution to the formation of secondary inorganics. With the deepening of pollution, inorganic elements were on the rise; Se, As, Pb, and Zn were highly enriched in inorganic elements. The results of principal component analysis (PCA) showed that secondary formation, industrial emissions, vehicle exhaust, and b

Key words: heavy air pollution; PM1; PM2.5; source apportionment; backward trajectory; potential source

随着社会经济的发展、人们生活水平的提高, 大气环境污染防治及其影响备受关注. 近年来大气

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颗粒物污染事件已经成为京津冀地区的焦点, PM, 5(空气动力学当量直径小于 2.5 μm 的颗粒 物)已经对大气能见度[1]、人类健康[2]和气候[3]产 生了严重影响. 尽管邯郸市及区县在"打赢蓝天保 卫战"期间,采取了一系列大气防治的管控措施, 并且在一定程度上减轻了大气颗粒物污染,特别 在冬季,由于气象不利条件及人为活动造成的高 强度排放,雾-霾等大气细颗粒物污染事件屡见不 鲜. 2020年11~12月邯郸总共经历了31 d污染 天,占比50.8%.由此表明邯郸正在遭受细颗粒物 的污染,尤其是亚微米颗粒物 PM₁,有研究表明, PM, 在长距离传输上扮演者重要的角色[4]. 此外 邯郸位于京津冀南部,西临太行山脉,与晋、豫、 鲁三省接壤,温带大陆性气候和地理位置会使污 染物的区域传输和空气质量产生重要的影响. 因 此研究华北南部地区细颗粒物变化及迁移特征有 重要意义.

自 2013 年以来,针对京津冀地区大气污染,相关学者也做了大量研究. Gao 等^[5] 和 Xu 等^[6] 的研究对北京颗粒物来源进行了分析,二次气溶胶占比最高(35.4%~42.4%).在 1999~2018 年北京市冬季 PM_{2.5}及其组分的对比研究中发现,2018 年冬季的 PM_{2.5}浓度及碳质组分都达到近 20 年较低水平,一次燃烧源对重污染的贡献较大,且在重污染期间 SO²₄ 和 NO₅ 受相对湿度和 NH₃ 的影响较大^[7,8].在 2018 年秋冬季典型霾污染的研究中, PM_{2.5}中水溶性离子和碳组分是此次霾污染形成的主要原因,同时 SNA 的暴发式增长抬高了颗粒物的浓度,高湿对 SO₂ 的液相氧化过程有着促进作用;源解析发现燃煤源为石家庄市首要污染源^[9].

针对邯郸市 $PM_{2.5}$ 的化学组分特征、来源和区域贡献相应学者也做了大量的研究. 有研究表明,2015 年邯郸市 $PM_{2.5}$ 浓度为 $122~\mu g \cdot m^{-3}$;当低温及相对湿度大于 60% 时,二次转化更为明显,同时风速小于 $1.0~m \cdot s^{-1}$ 且风向为南风和西南风时,污染物更容易积累 $^{[10]}$; Chen 等 $^{[11]}$ 的研究对邯郸市冬季 $PM_{2.5}$ 做了源解析,发现二次源、生物质燃烧和工业排放是污染物主要的来源,通过潜在源解析(PSCF),表明邯郸受南部区域的传输影响较为突出. 2017 年邯郸市 $PM_{2.5}$ 中 ρ [有机碳(OC)]和 ρ [元素碳(EC)]均值分别为 $17.09~\mu g \cdot m^{-3}$ 和 $4.11~\mu g \cdot m^{-3}$,道路扬尘、燃煤和机动车尾气是主要的污染来源 $^{[12]}$. 但针对 PM_1 的研究不多,相比城市关注度,重污染城市周边区县的研究也就更为空缺.

本文以华北南部地区邯郸市鸡泽县为研究对 象,该地位于邯郸市东北部,地势低,有大量铸造企 业,毗邻邯郸市永年区,与邢台市交界,污染严重, 2020年在全省 167 区县中 PM_{2.5}浓度排名倒数第三. 因此研究该区域对污染物化学组分特征及来源将有更深入地了解,可为大气污染区域联防联控提供科学的理论指导.

1 材料与方法

学

1.1 样品采集

采样点位于邯郸市鸡泽县第一中学楼顶(114.51°E,36.93°N),采样高度距地面约15 m.周围是居住区与教学区,无明显的污染源,基本能够代表所在区域的大气环境状况.2020年11月23日至12月12日,使用滤膜采样器采集 PM_{2.5}和 PM₁样品各20个,采集时间段为当日20:00~次日19:30,共计23.5h,采样滤膜均为石英膜.

1.2 质量控制

使用中流量采样器(KB-120F)采集 $PM_{2.5}$ 颗粒物,流量为 16.67 L·min^{-1} ,采样器切割头的直径为 90 mm, PM_1 采用(TE-Wilbur 型, TISCH, USA)采样器,流量为 16.67 L·min^{-1} ,切割头直径为 47 mm. 在采样过程中,仪器记录了平均温度、标况体积、采样时间等参数.根据环保部发布的环境空气 $PM_{2.5}$ 的测定重量法(GB/T15432)对颗粒物质量浓度进行分析,采样前将石荚膜在 550° 马弗炉中焙烧 5.5 h,称重前将石荚膜放在恒温箱(温度 25° $\pm 0.5^{\circ}$ 和湿度 $30\% \pm 5\%$)中恒温 24 h 以上,并在电子天平(瑞士,XS205dualrange,0.01 mg)上进行多次称量,保证其中两次的偏差控制在 $\pm 20 \text{ µg}$ 以内,取这两次称重质量均值,通过重量差来计算 $PM_{2.5}$ 和 PM_1 的浓度.

1.3 样品分析

水溶性离子分析时,加入 50 mL 去离子水,其电阻率为 18.2 M Ω ·cm,在超声波内超声 1 h,静置后进行过滤,最后利用离子色谱仪(DionexICS-1100)测得 Ca²+、NH $_4^+$ 、Mg²+、Na $_4^+$ 、K $_4^+$ 、SO $_4^2$ -、NO $_3^-$ 和 Cl $_4^-$ 以 种无机离子成分。OC/EC 的分析:用元素碳与有机碳分析仪(DRI Model 2001A,美国沙漠所)对样品中的碳组分进行分析.无机元素分析:使用ICP/MS(Agilent7500a)进行元素含量分析,从而获得 Pb、Zn、Cd、As、Se、K、Na、Ca、Mg、Al、Mn 和 Fe 等元素的浓度^[13].

1.4 基于 ISORROPIA Ⅱ模型气溶胶液态含水量估算

气溶胶热力学模型 ISORROPIA II 常用来计算 $Na^+-K^+-Ca^{2+}-Mg^{2+}-NH_4^+-Cl^--NO_3^--SO_4^{2-}-H_2O$ 的平 衡组分,目前已经广泛应用到区域及全球大气模型

中,且现有的众多研究中用于大气和气溶胶的物理状态和组成中[14,15]. 此模型分为正向和反向两种模式,正向模式是在给定气体和气体的总浓度时,计算平衡库分配的气溶胶种类,反向模式则基于温度、湿度和气溶胶浓度来计算,本研究采用反向模式计算. 此外 ISORROPIA II 模型提供"亚稳态"和"固态+液态"两种解决方案,考虑到采样期间相对湿度较高,于是选择前者[16]. 由此计算出热力学平衡状态下 H⁺和 ALWC 浓度.

1.5 后向轨迹模型

通过使用 HYSPLIT-4 模型观测气团 24 h 的后向轨迹,该模型适用计算和分析大气污染物输送和扩散轨迹,计算模式欧拉和拉格朗日,起始高度设置为 500m,每天计算 24 条轨迹,基于后向轨迹结果,模拟污染物输送的路径并进行聚类分析.

基于 HYSPLIT, 潜在源解析(PSCF) 可用于识别 受体周围污染物的可能源区域 $^{[1^{-3}]}$, 在本研究中, 它被用来识别采样点的 $PM_{2.5}$ 及其物种的潜在来源区域. PSCF 分析是基于 GIS 软件中的 TrsjStat 进行的. PSCF 的中心域是 36. 92°N, 114. 87°E, 水平分辨率为 0.5° × 0.5° × 0.5° . PSCF 值计算公式如式(1):

$$PSCF_{ij} = \frac{M_{ij}}{N_{ij}} \tag{1}$$

式中, N_{ij} 为落入ij单元内的端点数量, M_{ij} 为同一单元内源贡献值高于任意设置的阈值的端点数量.为了减少有限点数的网格单元 N_{ij} 的不确定性,PSCF值是通过其与 W_{ij} 相乘得到的. WPSCF 计算见式(2), W_{ii} 的计算见式(3):

$$WPSCF_{ij} = PSCF_{ij} \times W_{ij}$$
(2)
$$W_{ij} = \begin{cases} 1.00, & 80 < N_{ij}, \\ 0.70, & 20 < N_{ij} \le 80, \\ 0.42, & 10 < N_{ij} \le 20, \\ 0.05, & N_{ii} \le 10. \end{cases}$$
(3)

2 结果与讨论

2.1 颗粒物浓度及水溶性离子特征

采样期间 ρ (PM, 5)均值为 124. 3 μg·m⁻³,是我 国《环境空气质量标准》中 PM,5二级标准限值(35 μg·m⁻³)的 3.54 倍,说明此期间的 PM。5污染较为 严重. 与国内其他城市相比, 高于 2019 年驻马店市 采暖季 (117.0 $\,\mu g \cdot m^{-3}\,)^{[17]}$ 及郑州市 (69.6 μg·m⁻³)^[18]; 2018 年同期 ρ (PM₁) 均值为 77.6 μg·m⁻³, 2020 年 PM, 浓度变化范围为 62.4 ~ 189.9 μg·m⁻³,平均值为114.5 μg·m⁻³,是2018年 浓度的 1.5 倍. PM₁/PM_{2.5}的比值变化范围 83.3% ~95.3%,高于秋季长江流域临安的66.0%[19]和北 京的79.4%^[20]. 深入研究发现,表1中 PM₁/PM_{2.5} 比值与湿度相关性较好(R=0.618),但与风速的相 关性较差(R = 0.183),表明高湿度、低风速条件 下,促进二次离子形成,且在 PM, 中较为突出,因此 导致 PM₁/PM_{2.5}的比值升高. 颗粒物及气象因素的 变化趋势如图 1 所示,其中平均风速为 1.2 m·s⁻¹, 相对湿度均值为84%,平均温度为1.6℃,NO2均值 为 50 μg·m⁻³, CO 均值为 1.4 μg·m⁻³, 表明存在生 物质和散煤燃烧.

表 1 气态污染物、温湿度、风速和 PM₁/PM₂₅比值的 Pearson's 相关性¹⁾

	SO ₂	NO ₂	CO	湿度	温度	风速	PM ₁ /PM _{2.5}
	502	1102	CO	业 及	血及	八还	1 M ₁ /1 M _{2.5}
SO_2	1						
NO_2	0. 550 *	1					
CO	-0.155	0. 575 *	1				
湿度	-0.702 **	-0.055	0. 617 *	1			
温度	-0.259	-0.186	-0.254	0. 245	1		
风速	-0.343	0.076	-0.020	0. 169	0. 217	1	
$PM_1/PM_{2.5}$	- 0. 397	0. 227	0. 540 *	0.618*	0. 501	0. 183	1

1)*表示在 0.05 水平(双侧)上显著相关, **表示在 0.01 水平(双侧)上显著相关

根据《环境空气质量标准》(GB3095-2012),结合研究区大气污染水平,本研究中将日均 ρ (PM_{2.5}) 小于等于75 μ g·m⁻³为清洁阶段(C);PM_{2.5}的日均浓度在76~150 μ g·m⁻³为轻/中度污染阶段(S);PM_{2.5}的日均浓度在151~250 μ g·m⁻³为重污染阶段(H). 由图 2 可知,PM_{2.5}中 NO₃⁻和 SO₄²一从清洁天、轻/中度污染天到重污染天浓度呈上升趋势,NO₃⁻和 SO₄²-在轻/中度污染天和重度污染天占比分别

为 43. 4% 和 46. 6%、27. 2% 和 27. 5%;PM₁ 中 NO₃⁻ 和 SO₄² 在轻/中度污染天和重度污染天占比分别为 40. 3% 和 43. 4%、24. 9% 和 25. 7%,占比较高,NH₄⁺ 占比在轻/中度污染天和重污染天均高于清洁天,且 3 种离子占比较 2018 年同期均有所下降,可能与此阶段不利的气象条件以及气态前体物较高的二次转化率密切相关. 另外相对较高的 SO₄² 也可能与燃煤锅炉湿法脱硫过程中的直接排放有关,因

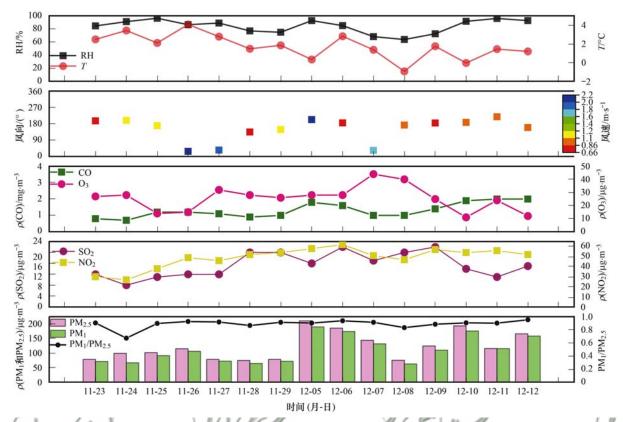


图 1 大气颗粒物和气态污染物浓度及气象条件变化趋势

Fig. 1 Time series of atmospheric particulate matter, gaseous pollutants and meteorological conditions

为环境空气中硫酸盐的前体物 SO_2 浓度较低(均值为 $16 \mu g \cdot m^{-3}$).

 $SNA(SO_4^{2-} \ NO_3^- \ an NH_4^+)$ 是 $PM_{2.5}$ 中重要的组分,通常对 $PM_{2.5}$ 的贡献率在 77.5% 左右 $[^{21}]$. 本研究,在轻/中度污染天和重污染天贡献分别为 44.2% 和 49.9%,明显高于清洁天(34.7%);对于 PM_1 而言,SNA 占比从清洁天的 34.2% 增加到污染天的 48.7%,由于 SO_2 和 NO_x 主要来自煤燃烧和机动车尾气的排放,因此用 NO_3^-/SO_4^{2-} 能反映移动源和固定源的相对影响大小 $[^{22}]$,本研究轻/中度污染时段和重污染时段 $PM_{2.5}$ 中 NO_3^-/SO_4^{2-} 值分别为 1.5 和 2.1, PM_1 中 NO_3^-/SO_4^{2-} 值分别为 1.7 和 1.8,二者均远大于 0.5,此外污染时段 $PM_{2.5}$ 和 PM_1 中 NO_3^- 和 NH_4^+ 较清洁天显著上升,分别是其 3.3倍和 2.6 倍、3.3 倍和 2.9 倍,说明污染时段移动源对水溶性离子的贡献大于固定源,因此应严控机动车尾气排放.

2.2 碳组分分析

碳组分通常包括有机碳(OC)和元素碳(EC). OC由一次有机碳(POC)以及二次有机碳(SOC)组成.本文采用 Lim 等^[23]提出的元素碳示踪法计算SOC的浓度,计算公式为:

$$POC = EC \times (OC/EC)_{min}$$
 (4)

$$SOC = OC - POC \tag{5}$$

式中,(OC/EC)min为OC/EC浓度比值的最小值.

本研究期间 PM, 中碳质组分 $\rho(OC)$ 和 $\rho(EC)$ 均值分别为 17.6 µg·m⁻³和 5.3 µg·m⁻³, PM₂₅中 碳质组分 $\rho(OC)$ 和 $\rho(EC)$ 均值分别为 15.8 $\mu g \cdot m^{-3}$ 和 4.7 μg·m⁻³, 高于郑州重污染期间 PM₂₅中 ρ (OC) 和 ρ (EC) 为 13.4 μg·m⁻³ 和 2.3 μg·m^{-3 [17]}, 低于山地型城市阳泉市 PM25 中的 23.9 μg·m⁻³和 6.5 μg·m^{-3 [24]}. 计算得到 ρ [总碳 (TC)] 分别为 22. 8 μg·m⁻³ 和 20. 5 μg·m⁻³, 占 PM₁ 和 PM_{2.5}的 20% 和 17%; PM₁ 和 PM_{2.5} 中 ρ(SOC) 均值分别为 5.6 μg·m⁻³和 7.4 μg·m⁻³, 占 OC 的 32% 和 47%, 说明 SOC 对 OC 的贡献较大. OC/EC 比值作为碳组 分来源的依据,本研究中PM,和PM,5中OC/EC比 值分别为 3.3 和 3.2,大于 2,同时也证明了 SOC 的 污染较为严重;另外机动车尾气和燃煤源的OC/EC 比值通常为 1.0~4.2 和 2.5~10.5, 由此说明汽车 尾气和煤燃烧是冬季重污染过程中颗粒物的重要 来源.

通过进一步分析,从图 3 可知, $PM_{2.5}$ 及 PM_1 中 $\rho(EC)$ 、 $\rho(POC)$ 和 $\rho(SOC)$ 浓度在清洁天分别为 4.3、8.2 和 5.3 $\mu g \cdot m^{-3}$ 及 4.9、11.7 和 1.5 $\mu g \cdot m^{-3}$; 重度污染天则分别达到 6.2、11.8 和 13.2

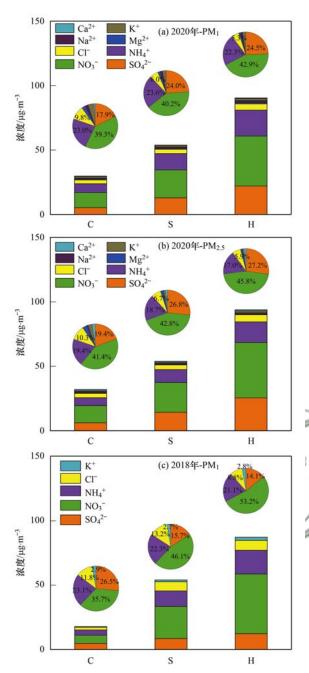


图 2 清洁天(C)、轻/中度污染天(S)、重度污染天(H) $PM_{2.5}$ 和 PM_1 中水溶性离子浓度及占比变化特征

Fig. 2 Water-soluble ion concentration and proportion in $PM_{2.5}$ and PM_1 on clean (C), slight/medium pollution (S), and heavy pollution days (H)

μg·m⁻³及7.1、16.6 和 10.4 μg·m⁻³,由此说明随着污染程度的增加,一次排放和二次转化是重污染过程中颗粒物的重要组分.但从占比情况分析,与水溶性离子浓度占比随污染程度的加剧的趋势不同, $PM_{2.5}$ 中 EC 和 POC 占比分别从清洁天的 24.1% 和 46.0%下降到重度污染过程的 19.8% 和 37.8%, PM_1 中 EC 和 POC 占比分别从清洁天的 27.4% 和 64.2%下降到重度污染过程的 20.8% 和 48.8%;相反, PM_1 和 $PM_{2.5}$ 中 SOC 则从 24.1% 和 8.4% 上

升至 42.4% 和 30.4%,进一步侧面印证了重污染过程中二次污染是重要的来源.

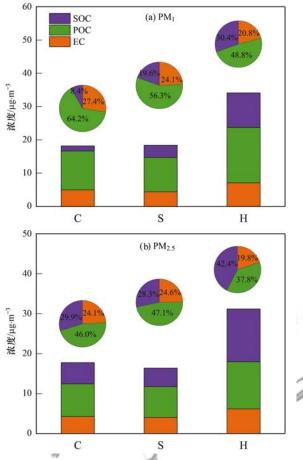


图 3 清洁天(C)、轻/中度污染天(S)、重度污染天(H) 颗粒物中碳组分浓度及占比变化特征

Fig. 3 Concentration and proportion of carbonaceous components in particulate matter on clean (C), slight/medium pollution (S), and heavy pollution days (H)

2.3 硫酸盐和硝酸盐的二次转化机制

硫酸盐和硝酸盐分别由 SO₂ 和 NO₂ 通过非均相反应生成. 因此本研究利用硫氧转化率(SOR)和 氮氧转化率(NOR)评价气态污染物二次转化程度^[25-27]. 计算公式如下:

$$SOR = c(SO_4^{2-})/[c(SO_4^{2-}) + c(SO_2)]$$
 (6)

NOR =
$$c(NO_3^-)/[c(NO_3^-) + c(NO_2)]$$
 (7) 式中, $c(X)$ 为物种的浓度.

 SO_4^{2-} 的形成主要有两种形式:① SO_2 通过气相反应被 OH 自由基氧化;②Fe 和 Mn 催化反应与溶解的 H_2O_2 或 O_2 反应,形成硫酸盐^[28].此外,煤炭燃烧也能直接生成 SO_4^{2-} .

 NO_3 的形成主要通过两个均相反应, $①NO_2$ 与 OH 反应生成 HNO_3 ,随后 HNO_3 与 NH_3 反应形成硝酸盐,该反应在白天占主导地位;②在夜间, NO_2 首先与硝酸根自由基(主要有 O_3 产生)反应,然后与 N_2O_5 反应,最后 N_2O_5 在遇水生成 HNO_3 ,进一步产

生 NO_3^- . 前人研究表明, NO_3^- 的形成对白天的 NO_2 和 O_3 浓度较为敏感, 此外, NH_3 可能会促使 HNO_3 分解并改变气溶胶酸度, 从而影响 N_2O_5 的吸收 $^{[29-31]}$.

图 4 结果显示,在不同污染阶段,PM₁ 和 PM_{2.5} 中 ρ (EC)与 ρ (SO₄²⁻)、 ρ (NO₃⁻)、SOR 和 NOR 有着 明显的不同. 随着污染程度的加剧,PM 中 ρ (SO₄²⁻)从清洁 阶段到重度污染过程由 6.2 μ g·m⁻³ 增加至 25.5 μ g·m⁻³, ρ (NO₃⁻)由 13.3 μ g·m⁻³ 增加至 42.9 μ g·m⁻³;PM₁中 ρ (SO₄²⁻)从清洁阶段到重度污染过程由 5.3 μ g·m⁻³ 增加至 22.2 μ g·m⁻³, ρ (NO₃⁻)由

11. 7 μg·m⁻³增加至 38. 7 μg·m⁻³,由此说明重污染天气下 NO_3 变化较为明显,且 NO_3 在 $PM_{2.5}$ 和 PM_1 中的占比与 SO_4^{2-} 相比较高,同时 $PM_{2.5}$ 中 NOR 由清洁天的 0. 2 增加至重度污染过程的 0. 4, PM_1 中 NOR 由清洁天的 0. 2 增加至重度污染过程的 0. 7,说明重污染期间氮氧化物的转化率较高,且在 PM_1 中尤为突出. 从清洁天到重污染天,随着 NO_3^- 浓度的增加, NO_3^- /EC 的比值同样显著增加,其中 $PM_{2.5}$ 和 PM_1 中的 NO_3^- /EC 在重度污染期间均值分别为 7. 1 和 5. 4,分别是清洁天的 2. 2 倍和 2. 3 倍. 在重污染期间,气溶胶液态含水量(ALWC)和相对湿度较高,对 EC 浓度的影响较大[11].

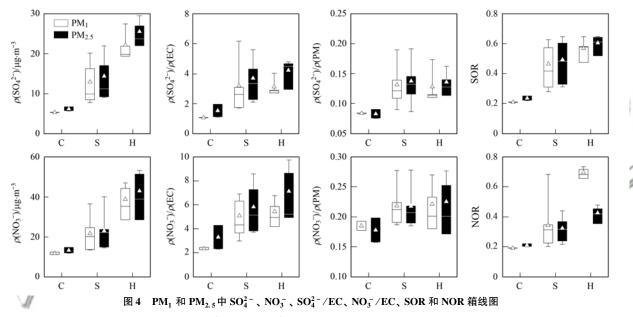


Fig. 4 Box plots of SO_4^{2-} , NO_3^- , SO_4^{2-}/EC , NO_3^-/EC , SOR and NOR in PM_1 and $PM_{2.5}$

从图 5 和图 6 可知,随着污染程度的加深, PM_{2.5}和 PM₁ 中 ALWC 随之增加,其中 PM_{2.5}-ALWC 均值为 107.5 μg·m⁻³, PM₁-ALWC 均值为 118.5 μg·m⁻³. 随着 ALWC 和湿度的增加, SO₂ 的转化速率要快于 NO₂. 因此, ALWC 的增加会促进气态污染物的二次转化. 有研究表明,当湿度较高时,促进 SO₂ 向 SO₄² 转化, SOR 会明显地增加^[32];然而对于 NO₃⁻ 而言,高湿度条件下,水在颗粒物上的反应一定程度上会抑制硝酸盐的生成^[33]. 除此之外,冬季 NO₂ 与·OH在白天的反应是生成 NO₃⁻ 的主要途径,因此在高 ALWC 的情况下, NO₃⁻ 的生成也因光照条件不足从而受到抑制^[11]. 总之, ALWC 会提高气态污染物的二次转化速率,尤其是 SO₂ 向 SO₄² 的转化,表明液相反应对硫酸盐的生成起着重要的作用.

2.4 无机元素分析

本研究中,无机元素的浓度从高到低顺序依次

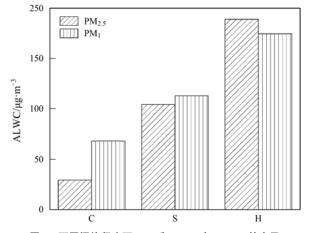


图 5 不同污染程度下 PM₁ 和 PM_{2.5} 中 ALWC 的水平

Fig. 5 Content of ALWC in PM₁ and PM_{2.5} at different pollution levels

为:Al>Ca>Fe>Zn>Pb>Ti>Ni>Mn>As>Cr>Se,发现Al、Ca和Fe等地壳元素浓度较高. 富集因子法可以衡量颗粒物中无机元素的来源, 富集因子

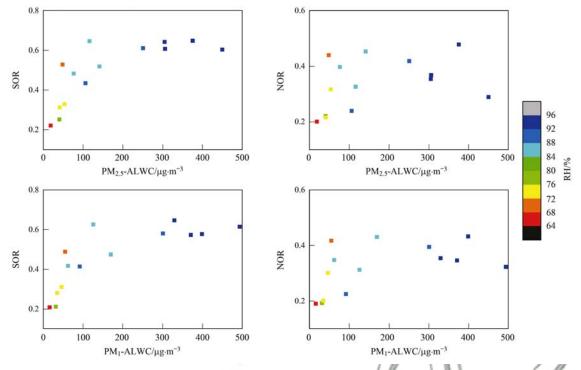


图 6 PM_{2.5}、PM₁ 中 ALWC 与 SOR 和 NOR 的散点图

Fig. 6 Scattering plots of ALWC and SOR, NOR in PM_{2.5} and PM₁ colored by relative humidity

(EF)计算公式如下

 $EF = (c_i/c_o)_{\text{#B}}/(c_i/c_o)_{\text{FF}}$ 式中, c_i 表示元素 i 的浓度, c_o 表示参比元素的浓 度. EF 中参比元素选择 Al,研究显示,当 EF≤1 时, 微量富集; 1 < EF ≤ 10, 为轻度富集; 10 < EF ≤ 100 为中度富集; 100 < EF ≤ 1000, 为高度富集; EF > 1000,为超富集[34].表2为不同污染程度下无机元 素的 EF 值,由表可知,随着污染程度的加深,10 种 无机元素均呈上升趋势. Cr、Ni、Pb、Zn 和 As 通常 来自机动车刹车片和汽车尾气, Zn、As 和 Pb 为中 度富集. Ca 为轻度富集,主要来自于扬尘. Ni 和 Cr 为化石燃料的标志物,均为轻度富集. Se 为超富集, 主要与工业生产有关.

表 2 无机元素在不同污染程度下的富集因子变化

Table 2 Enrichment factor of inorganic elements

under different pollution levels 无机元素 C Н 0.06 Ti 0.03 0.05 Cr1.03 1.50 2.06 Mn 0.10 0.13 0.23 Fe 0.09 0.11 0.15 2.08 3.55 Ni 3.01 1.28 2.01 Ca 1.89 Zn 17.82 24.27 38. 79 As 51.85 67.74 86.00 1 199. 29 1 898. 25 Se 678.07 Pb 23.41 33.84 61.18

2.5 源解析

本研究利用主成分分析法(PCA)进行源解析

(表3),主要包括水溶性离子、碳组分和无机元素 共计23种,特征值大于1的因子共2个.对于 PM_{2.5},因子1中NH₄、SO₄-、NO₃、OC、K+、Cl-、 Zn、Cd 和 Pb 载荷较高,NH₄ 、SO₄ - 和 NO₃ 主要来

表 3 主成分旋转矩阵

Table 3	Rotated con	•	atrix of princi	ipal compone	nt analysis
<i>2</i> п	Л	PI	M _{2.5}	P	M ₁
组织	<i>T</i> –	因子1	因子2	因子1	因子2
0	С	0. 779	0.020	0.805	0.006
E	C	0.744	-0.117	0.849	0.012
Na	+	0.497	-0.229	0.305	-0.268
NH	4	0. 934	0. 258	0. 926	0. 275
K	+	0.899	0.040	0. 788	0.056
Mg^2	2 +	0.388	-0.185	0. 243	-0.019
Ca ²	!+ .	-0.072	-0.242	-0.005	-0.231
Cl	-	0.878	0.043	0. 798	0.057
NO	3	0.889	0. 250	0.859	0.361
SO_2^2	2 -	0.881	0.334	0.871	0.387
A	1 .	-0.017	0.893	-0.024	0.784
T	i	0. 126	0. 525	0. 242	0.556
C	r	0. 168	0. 935	0. 197	0.977
M	n	0. 262	0. 598	0. 296	0.426
F	е	0.456	0.834	0.499	0.802
N	i ·	-0.016	0.956	-0.014	0.961
C	a ·	-0.162	0.761	-0.127	0.754
Zı	n	0.851	0.372	0.863	0. 252
A	s	0. 152	0.044	0. 143	0.036
Se	е	0.424	0.685	0.502	0.740
Co	d	0.880	-0.055	0.877	-0.096
Ba	a	0.431	0.676	0. 341	0.514
Pl	b	0.842	0. 262	0.890	0. 163
方差贡	献/%	37. 06	28. 76	35. 07	30. 59

自二次转化,K⁺、Cl⁻和 OC 主要被认为是生物质和 煤炭燃烧示踪物^[11],Zn、Cd 和 Pb 与工业生产有 关^[18],方差贡献 37.06%,因此因子 1 被识别为二次 源、燃烧源和工业源的综合源;因子 2 中 Cr 和 Ni 载荷较高,Cr 和 Ni 主要与机动车尾气有关^[35],方差 贡献 28.76%,因此被识别为机动车源.对于 PM₁,因子 2 机动车源贡献率高于 PM_{2.5},说明粒径越小,对 机动车尾气的贡献越大. 综上所述,主要受二次源、工业生产、燃烧源和机动车源的影响.

2.6 后向轨迹分析

污染物的来源通常有两部分组成,一是本地排放;二是来自于区域传输污染,识别了该地区污染物的潜在源区,可以为大气污染联防联控提供科学依据.有研究表明,京津冀地区的 PM_{2.5}受区域传输污染较为严重,占比在 30%~40% 之间^[36,37].本研究将 24 h 后向轨迹总共聚类为 6 类,结果如表 4 和图 7 所示. Cluster 1 (C1) 为短距离传输,主要来自山

西省、河北省和河南省,如晋城市、安阳市和邯郸 市. C1 占比为 34.2%, 且 ρ(PM, 5) 均值为 94.0 μg·m⁻³, 无机元素在该轨迹浓度最高为 1.8 μg·m⁻³; Cluster 2(C2)、Cluster 4(C4)和 Cluster 6 (C6)主要来自于西北部,均为长距离传输,占比分 别为 20.6%、9.7% 和 1.9%, ρ(PM_{2.5})均值分别为 123. 4、129. 8 和 116. 8 μg·m⁻³,且 SO₄²⁻、NO₃、EC 和 OC 在 C2、C4 和 C6 中均值较高,表明与生物质 燃烧有直接的联系[38]; SO₄ 和 NO₃ 浓度较高可能 归因于矿物气溶胶的较大表面积,为 NO,和 SO,提 供了非均相反应的条件,同时有可能来自于西北地 区的大气流动,导致区域传输污染,从而抬高了污染 物的浓度均值. 总体来说,此区域受西北方向的传输 影响较为明显. Cluster 3(C3)和 Cluster 5(C5)主要 来自东北地区,如北京市、天津市、沧州市和衡水 市,占比分别为 15.8% 和 17.8%,此时 $\rho(PM_{25})$ 均 值分别为 82. 5 μg·m⁻³和 60. 2 μg·m⁻³

表 4 不同轨迹下大气颗粒物及主要组分的平均浓度/ $\mu g \cdot m$

TC 1.1 4	A	c (Elinia	/ 6	1 .	/	1	1:cc .	A	
Table 4	Average concentration of	of atmospheric	particulate matter	and main	components	under	amerent	trajectories/	$\mu g \cdot m$

轨迹	$\rho(\mathrm{PM}_{2.5})$	$ ho(\mathrm{SO_4^{2-}})$ $ ho(\mathrm{NO_3^-})$	ho(EC) $ ho(OC)$	ρ(无机元素)
CI	94. 0	15. 9 27. 9	4. 3 16. 6	1.8
C2	123. 4	18. 4 32. 9	5. 1 17. 8	1.5
C3	82. 5	16. 2 23. 2	3. 9	1.6
C4	129. 8	18. 6 26. 7	5. 5	1.3
C5/	60. 2	10. 8 18. 6	3. 9 11. 7	1.7
C6	116. 8	26. 9 28. 5	5. 8 21. 8	1. 1

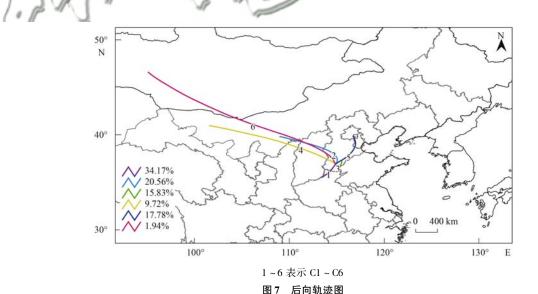


Fig. 7 Backward trajectory diagram

图 8 显示了 PM_{2.5}、SO₄²、NO₃、EC、OC 和无机元素的 WPSCF 分布. 总体来看,污染物的潜在源分布主要集中在中国内部,且集中在邯郸西南部、东北部和西北部. 其中邯郸南部地区 WPSCF 值高,原因可能是南部地区空气质量较为严重,且很大程

度上受重工业排放的污染物影响. 其它高值区域还有东北部(石家庄市、邢台市和北京市)和西北地区(如山西省太原市等). SO_4^2 、 NO_3 、EC、OC 和无机元素的 WPSCF 高值区域都主要来自观测区域的东北方向和西南方向,这与 Zhang [39] 的研究结果类似.

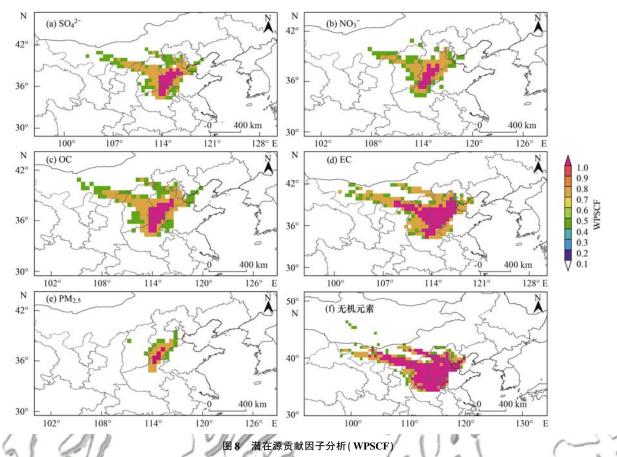


Fig. 8 Potential source contribution factor analysis results

3 结论

(1)ρ (PM_1) 和ρ $(PM_{2.5})$ 均值分别为 114. 53 μg·m⁻³ 和 124. 25 μg·m⁻³, 高于 2018 年冬季ρ (PM_1) 均值 77. 6 μg·m⁻³; $PM_1/PM_{2.5}$ 的比值变化范围 83%~95%, 占比较高. 另外 SNA 是 $PM_{2.5}$ 和 PM_1 中主要的水溶性离子,且 PM_1 中 SNA、OC、EC 占比较高,表明污染物更容易在细小颗粒物中积累.与清洁天相比, NOR 在重污染期间增长 3 倍,表明 NO₂ 二次转化明显.

- (2) PM_{2.5}中 SOC 占比随污染程度的加深上升 12.5%, PM₁中 SOC 上升 22%, 而 EC 和 POC 下降, 表明二次污染是重污染过程中颗粒物的重要组成部分; ALWC 对 SOR 和 NOR 的转化速率有促进作用.
- (3)随着污染程度的加深,无机元素浓度增加. 富集因子显示,Se 为超富集,Zn、As 和 Pb 为中度 富集.
- (4)主成分分析显示主要受二次源、机动车源、燃烧源和工业源影响,且粒径越小对机动车源的贡献越大. HYSPLIT 结果显示 6 类轨迹对颗粒物和化学组分有显著的影响. SO₄²、NO₃、EC、OC 和无机元素的 WPSCF 主要高值区域都主要来自观测区域的东北方向和西南方向,如河南(安阳和郑州)和河

北(石家庄和邢台)等.

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