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北京南郊区 PM。,中水溶性无机盐季节变化及来源分析

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摘要:为研究北京偏南地区细颗粒物 $(PM_{2.5})$ 中水溶性无机离子的变化特征,利用大气细颗粒物快速捕集系统及化学成分分析系统 RCFP-IC,于 2016 年对北京南郊区大兴 $PM_{2.5}$ 中 9 种水溶性无机离子 $(Cl^- NO_2^- NO_3^- SO_4^{2-} Na^+ NH_4^+ K^+ NH_4^- NH_4$

关键词:PM_{2.5}; 水溶性无机盐; 季节变化; 日变化; 来源

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Seasonal Variation and Source Analysis of Water-soluble Inorganic Salts in PM_{2.5} in the Southern Suburbs of Beijing

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Abstract: In order to study the variation of water-soluble inorganic ions in the four suburbs of Beijing using the atmospheric fine particulate matter rapid trapping system and chemical composition analysis system (RCFP-IC), we carried out measurements for nine water-soluble inorganic ions (Cl-, NO₂-, NO₃-, SO₄--, Na+, NH₄+, K+, Mg²⁺, Ca²⁺) in PM_{2.5} with continuous on-line observations for one year in Beijing's southern suburbs in 2016. The transport process of pollutants and the potential sources of pollutants were evaluated by combining a trajectory clustering method and potential source contribution factor analysis method (PSCF). During the observation period, the total concentration of the nine water-soluble inorganic ions was 38.6 µg·m⁻³, and results showed that the concentration in winter and spring was high and in summer and autumn was low. The order of the concentration from high to low was $SO_4^{2-} > NO_7^{-} > NH_4^{+} > Ca^{2+} > NO_7^{-} > Cl^{-} > Na^{+} > K^{+} > Mg^{2+}$. In winter, the SO_4^{2-} , NO_7^{-} and NH_4^{+} accounted for 75.7% of the total measured water-soluble ions, followed by 72.8% in spring and 60.2% in summer. With an increase in air pollution, the concentrations of SO_4^{2-} , NO_3^{-} , and NH_4^{+} increased significantly, indicating that SO_4^{2-} , NO_3^{-} , and NH_4^{+} were closely related to the deterioration of air quality. SO₄² was dominant in the formation of secondary ions compared to NO₃ and NH₄⁺; and SO₄², NO₃⁻, and NH₄ had significant diurnal variations. The diurnal variation of the SO₄²⁻ statistic (hours) was bimodal, and the peak values were at about 10:00 and 18:00. The diurnal variation of NO₃ and NH₄ had single peaks, with the peak appearing at 10:00. The trend of the diurnal variation for these two ions was similar. Finally, the sources of pollution in the southern suburbs of Beijing mainly included secondary sources, coal-fired sources, and mixed sources of dust and dust. The main potential source of pollution in the southern suburbs was in the southeastern part of the observation site, while the northeastern airflow was favorable for the diffusion and dilution of

Key words: PM25; water-soluble inorganic ions; seasonal variation; diurnal variation; source

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大气细颗粒物 ($PM_{2.5}$ 空气动力学直径 $\leq 2.5 \mu m$ 的颗粒物),是多数城市首要空气污染物之一^[1]. 当前,我国大气霾污染频发与高浓度 $PM_{2.5}$ 密切相关^[2], $PM_{2.5}$ 由于比表面积大、粒径小,容易在空气中滞留等特点,容易成为其他污染物的反应体和载体,从而富集更多有害的化学组分^[3]. $PM_{2.5}$ 的化学组分包括有机碳、元素碳、水溶性离子、地壳元素和各种微量元素等,其中水溶性无机离子是 $PM_{2.5}$ 的一类重要组分,其对大气气溶胶酸碱度产生直接贡献,并对成核和云滴增长起到了重要的作用^[4].

当前,国内外学者针对北京地区 PM_{2.5}长期变化研究主要集中在 PM_{2.5}质量浓度时空分布变化特征,化学组成(水溶性离子、无机元素、碳质组分与单体有机物等)和来源等方面^[5-7],然而上述研究主要集中在北京城市区域,缺少对北京郊区的观测.根据环境保护部公布的《2016 年空气质量公报》显示^[8],北京从北到南 PM_{2.5}质量浓度呈现递增趋势.因此对北京南郊 PM_{2.5}质量浓度呈现递增趋势.因此对北京南郊 PM_{2.5},后量改善等.当前研究有助于污染成分分析和空气质量改善等.当前研究表明北京深受 PM_{2.5}污染影响并且 SO²₄ 和NO³₃ 是能见度降低的主要贡献者,然而,关于时间变化(尤其是日变化)和南郊区 PM_{2.5}水溶性离子来源分配的分析缺乏,而且对于水溶性无机离子的研究多采用传统的滤膜采样技术,缺少高时间分辨率在线观测技术.

本研究基于对北京南郊区代表站点大兴站 $PM_{2.5}$ 中水溶性无机离子(Cl^- 、 NO_2^- 、 NO_3^- 、 SO_4^{2-} 、 Na^+ 、 NH_4^+ 、 K^+ 、 Mg^{2+} 、 Ca^{2+})展开为期 1 a 的观测,同时结合 $PM_{2.5}$ 质量浓度,对南郊区水溶性无机离子浓度水平、季节变化、 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 日变化及水溶性无机离子来源进行分析,探讨了不同空气质量级别下主要离子的变化规律,并对冬季进行气团轨迹聚类的基础上采用 PSCF(潜在源贡献因子分析法)识别了 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 的潜在源区,以期为掌握北京南郊区 $PM_{2.5}$ 中水溶性无机离子污染特征提供科学依据.

1 材料与方法

1.1 采样地点和采样时间

采样地点(116.336°E,39.493°N)位于北京大 兴区榆垡南8 km² 范围内,国家野生动物园南侧站 点附近1 km 区域内无污染源,站点有较好的空间 代表性.站点所在区域年平均气温12.0℃,年平均 降雨量为398 mm,降雨主要集中在夏季.采样时间 为2016年1月1日至2016年12月31日.

1.2 方法和仪器

利用 RCFP-IC 对 PM_{2.5}中水溶性盐浓度展开连 续观测. RCFP-IC 实验原理及基本性能参数见文献 [9]. 大气细颗粒物进入系统前通过串联的扩散管 去除酸性和碱性气体, 热的过饱和水蒸气与冷的气 溶胶流正交混合,绝热膨胀,使细粒子长大成为能 被惯性撞击器捕集的大粒子;长大的粒子在增长器 末端与气体垂直分离,在惯性力的作用下沿轴线方 向撞入带有内标的收集液,在蠕动泵推动下收集液 被送入离子色谱进行在线的分离检测. 系统为在线 连续自动采样,采样分析频率设定为30 min. 水溶 性无机离子分析仪器使用的是美国 Dionex 公司的 ICS-900 离子色谱, 阴离子分析柱为 Ionpac AS14, 4 mm×250 mm, 淋洗液为 3.5 mmol·L⁻¹ Na₂CO₃ + 1.0 mmol·L⁻¹ NaHCO₃, 阳离子分析柱为 Ionpac CS12A, 4 mm×250 mm, 淋洗液为20 mmol·L⁻¹甲 烷磺酸.

颗粒物质量浓度观测采用美国赛默飞世尔公司生产的双通道 1405DF 型振荡天平法的 $PM_{2.5}$ 和 PM_{10} 分析仪. 观测粒径为 2.5 μ m 以下颗粒物. 该仪器最低检测限 0.06 μ g·(m^3 ·h) $^{-1}$, 质量分辨率为 0.10 μ g· m^{-3} . 数据质控和保证方法详见 $PM_{2.5}$ 质控手册^[10].

1.3 RCFP-IC 质量控制

RCFP-IC 质量控制选用外标法和内标法结合. 内标物示踪法是为了校正蠕动泵和抽气泵产生的气体流量和液体流量,控制系统的稳定性,添加大气中不存在的 Li⁺,内标物为 100 × 10⁻⁶ (体积比,1 mL LiF 和 1 mL 去离子水)LiF 溶液,以保证数据准确. 外标法是对大气颗粒物中的水溶性离子的定量分析,用不同浓度的标准溶液以每月一次的频率对仪器进行标定. 要获得水溶性离子的检出限,将1 g·L⁻¹的离子标准溶液稀释至 1 mg·L⁻¹,然后逐渐进行稀释直至 IC 检测出的离子峰值高出噪声峰值的 3 倍,取一个稀释浓度,重复 6 次,并测量得到标准偏差. 3 倍标准差对应正常运转下的 RCFP得到的各离子浓度为 RCFP-IC 的检测限[11].

RCFP-IC 系统的背景空白主要是进样口膜上的 痕量离子,实验数据需要扣除系统空白. 空白试验 是在更换进样口膜以及添加实验用去离子水的情况 下进行的. 空白实验采用电阻率为 $18.2~M\Omega \cdot cm$ 的 去离子水1~mL, 重复 $3~\chi$, 实验期间, 系统空白均 低于 $1~\mu g \cdot m^{-3}$.

1.4 数据分析方法

- (1)实验中选用的源解析技术:因子分析法 (PCA)是根据相关性及方差分析,将颗粒物的化学成分数据中的多个变量归结为真正起作用的最少数目的独立因子,以期获得主要的污染来源,该模型在本次研究中选用 SPSS19.0 软件实现.
- (2)在本实验中,使用 TrajStat 软件^[12],该软件利用 NOAA Air Resources Laboratory 研发的 HYSPLIT 模式的计算模块,并结合了美国环境预报中心和国家大气研究中心联合执行的全球再分析资料,对应有效的小时样本数据,计算观测期间到达南郊区的1 435条 48 h 后向轨迹,轨迹计算的起始点高度为距地高度 500 m AGL (Above Ground Level),并对计算出的后向轨迹进行聚类分析.
- (3)在得到的后向轨迹基础上使用 PSCF(潜在源贡献因子分析法)估计污染物的潜在源区.该方法原理及具体的操作步骤详见文献[13]. PSCF的值越大,表明该网格点对观测点的离子浓度贡献越大.高 PSCF值所对应的网格组成的区域就是南郊区硫酸盐、硝酸盐及铵盐浓度的潜在源区,经过该区域的轨迹就是对离子浓度有影响的输送路径.

2 结果与讨论

2.1 2016 年南郊区水溶性无机离子浓度水平分析

图 1 为南郊区 PM,5中主要的水溶性无机离子 浓度分布特征. 从中可知, 南郊区总水溶性无机离 子年平均浓度为 38.6 μg·m⁻³, 并呈现冬春高, 夏 秋低的特征. 春夏秋冬水溶性无机离子的平均浓度 分别为 45.1、31.9、33.4 和 54.9 μg·m⁻³, 春季水 溶性无机离子质量浓度较高,整个夏季质量浓度较 低且变化范围不大, 秋季水溶性无机离子质量浓度 开始回升,冬季达到最高,这一变化规律与杨懂艳 等[14] 对北京市 PM,5 中水溶性无机离子变化的研究 具有很好的一致性. 各离子质量浓度高低顺序为 $SO_4^{2-} > NO_3^{-} > NH_4^{+} > Ca^{2+} > NO_2^{-} > Cl^{-} > Na^{+} > K^{+}$ > Mg²⁺, 其中 SO₄²⁻、NO₃⁻ 和 NH₄⁺ 平均质量浓度分 别为 14.5、9.9 和 4.7 μg·m⁻³, 其次是 Ca²⁺ 和 Cl⁻, 两种离子的平均质量浓度分别为 4.1 μg·m⁻³ 和 1.9 μg·m⁻³, 而 Na⁺ 和 K⁺ 仅为 1.2 μg·m⁻³ 和 0.9 μg·m⁻³. SO₄²⁻、NO₃⁻ 和 NH₄⁺ 质量浓度之和占 所测水溶性离子浓度之和的70.5%,占PM,5的质 量分数高达31%,其中,3种离子在冬季浓度占比 最高为75.7%;春季次之,为72.8%;夏季最低, 仅为60.2%,说明二次无机离子是PM,5的主要组

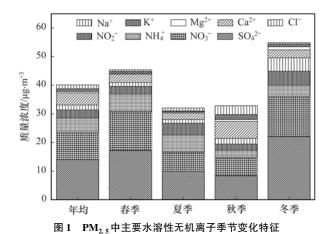


Fig. 1 Seasonal variation of water-soluble inorganic ions in $PM_{2.5}$ 成部分.

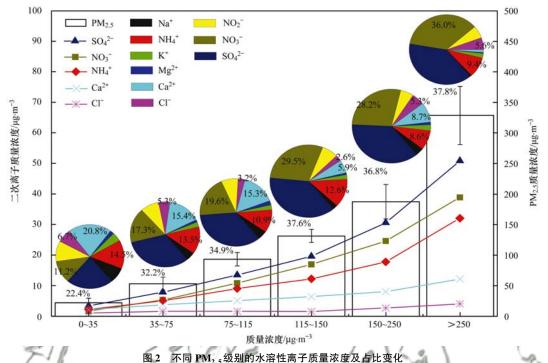
根据 PM,5质量浓度将环境空气质量划分为 6 种级别, 分别为优(0~35 μg·m⁻³)、良(35~75 μg·m⁻³)、轻度污染(75~115 μg·m⁻³)、中度污染 (115 ~ 150 μg·m⁻³)、重度污染(150 ~ 250 μg·m⁻³)、严重污染(250 μg·m⁻³以上). PM_{2.5}中各 水溶性无机离子的平均质量浓度随污染程度增强而 增大. 观测期间, 除二次离子 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 外, Ca2+和 Cl-这2种离子浓度也较高,是 PM, 中 水溶性无机离子的主要组分. 图 2 是不同 PM25级 别的水溶性无机离子质量浓度及各离子浓度占水溶 性无机离子浓度的百分比变化特征. 如图所示, 在 轻度污染、中度污染、重度污染和严重污染程度 下, 这 5 种水溶性无机离子的质量浓度(WSIIs)分 别为 41.9、52.5、61.2 和 92.1 μg·m⁻³; 在这 4 种 级别下, SO₄²、NO₅ 和 NH₄ 分别占 WSIIs 的 77.9%、82.0%、84.9%和87.9%,说明随污染程 度增加, SO₄²⁻、NO₃⁻ 和 NH₄⁺ 这 3 种水溶性无机离 子的贡献增加.

另外,本文对不同 $PM_{2.5}$ 级别中各离子浓度占水溶性无机离子浓度的百分比变化特征进行分析,随着 $PM_{2.5}$ 质量浓度的增加, Ca^{2+} 、 Cl^- 占 $PM_{2.5}$ 的比例降低,而 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 这 3 种离子质量浓度占 $PM_{2.5}$ 质量浓度的比值急剧上升,说明 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 是 $PM_{2.5}$ 质量浓度增加的主导离子。在严重污染程度中 SO_4^{2-} 的累积速率最明显,说明 SO_4^{2-} 的快速增长对 $PM_{2.5}$ 的形成有较大贡献。观测期间, SO_4^{2-} 浓度水平明显高于以往北京市的研究结果[15,16],也明显高于背景站上甸子[17]及南方城市上海及香港的水平[18,19]。南郊区代表站点大兴站与同是郊区站点的韩国光州科技学院站点相比,

SO₄- 比光州科技学院站点显著高出 73. 36% [20].

燃煤、生物质燃烧和海盐都是 Cl^- 的排放源,但北京属于内陆城市,基本不受海盐离子的影响,而北京冬季采暖会燃烧大量化石燃料,排放较高浓度的 Cl^- 和 K^+ . K^+ 作为生物质燃烧的示踪物^[21],本观测结果中未见冬季 K^+ 浓度显著升高,因此推

测南郊区冬季 Cl^- 主要来源于供暖燃煤,且 Cl^- 浓度在冬季最高(4.67 $\mu g \cdot m^{-3}$),其在总离子中的比例也上升到 9.3%,说明 Cl^- 的浓度变化受冬季燃煤影响最大. 受到春季沙尘的影响,主要来源于矿尘的 $Ca^{2+[22]}$ 在春季最高,平均浓度为 2.67 $\mu g \cdot m^{-3}$,其对总离子的贡献比例达到 6.3%.



11 11 12 12.5 WARNING EIG 1 X EIG XX 11 10 X 10

Fig. 2 Variation of concentrations of water-soluble ions at different PM_{2.5} levels

2.2 SO_4^{2-} NO_3^- 和 NH_4^+ 的日变化特征分析

 SO_4^2 、 NO_3^- 和 NH_4^+ 这 3 种水溶性无机离子是水溶性离子的主要组分,且这 3 种水溶性无机离子的日变化会受到人为源的干预(如汽车排放和化石燃料燃烧)及其产生机制的显著影响^[23]. 因此本文将探讨 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 这 3 种水溶性无机离子日均浓度的变化规律,如图 3 所示,其浓度范围分别是 31.85 ~ 39.40、12.06 ~ 29.41 和 21.64 ~ 37.28 $\mu g \cdot m^{-3}$.

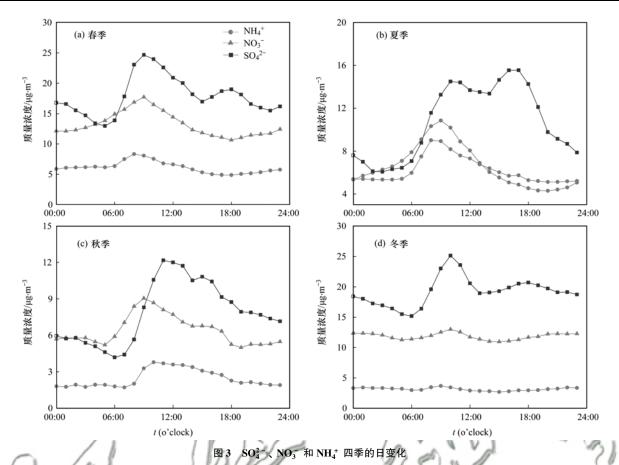
受不同排放源的影响,不同离子的日变化特征也不同. SO²⁻ 在四季均出现两个浓度高峰,随着太阳的升高浓度增加并在 10:00 左右完成浓度累积并达到峰值,随着时间的推移浓度降低在 18:00 附近出现第二个峰值,而且白天浓度明显高于夜间浓度. 这一规律可以解释为早峰浓度增加,由于光化学作用增强;傍晚峰值,由于边界层高度降低的污染物累积的结果,这与济南、珠三角观测结果类似^[24,25]. 白天浓度高可以解释为白天太阳辐射较强,光化学或非均相化学反应会促进 SO²⁻ 的产

生[26]

观测期间,春、夏和秋季, NO_3^- 峰值出现在早上 $07:00 \sim 09:00$,下午 $16:00 \sim 18:00$ 达到 NO_3^- 浓度最低值. 上午的峰值与早高峰机动车污染排放有关,下午边界层高度增加导致低浓度 NO_3^- 出现. 冬季, NO_3^- 日变化不明显,可能由于低温下热力学平衡的微小影响^[27]. 在四季, NH_4^+ 与 NO_3^- 的日变化特征相似,表明存在 $NH_4^+NO_3$.

2.3 水溶性无机离子的来源

为了解析南郊区 $PM_{2.5}$ 中水溶性无机离子的来源,利用主成分分析方法对全年水溶性无机离子进行解析,获得表 1 的 PCA 结果,结果显示对水溶性无机离子提取了 2 个主成分因子,共解释了63.64%的污染源.因子 1 中载荷值较高的为 SO_4^{2-} 、 NO_3^- 、 NH_4^+ 和 Cl^- ,相关系数依次为 0.879、0.890、0.717和 0.604,解释了总方差的 37.38%. SO_4^{2-} 、 NO_3^- 和 NH_4^+ 是二次源的典型代表物种 [28], Cl^- 的来源比较复杂多样,海盐、燃煤等均有可



Diurnal profiles of SO₄² NO_3^- , and NH_4^+ in the four seasons

方羊

能^[29],但北京离海有 150 km 以上^[30], 所以忽略 Cl⁻来自海上气溶胶的输送可能, 因此因子1 定义 为二次源和燃煤源. 因子2解释了总方差的 26.26%, 其中 Ca2+和 Mg2+载荷值较高, 分别是 0.92和0.90,这两种离子主要来源于建筑扬尘或 土壤风沙尘[31],因此将因子2定义为土壤风沙尘 和建筑扬尘的混合源.

南郊区 PCA 结果

}	因子1	因子2	公因子:
-	0.879	0.07	0.77
-	0.89	-0.028	0. 79
+	0.717	0. 181	0. 54
	0.604	0.012	0.26

Results of PCA in the Daxing district

211/1	四 1 1	□ 1 Z	公囚 1 万左
SO ₄ -	0. 879	0. 07	0. 777
NO_3^-	0.89	-0.028	0. 794
$\mathrm{NH_4}^+$	0.717	0. 181	0. 546
Cl -	0.604	0.013	0. 365
K +	0. 421	0.486	0. 413
Na +	0.11	0. 721	0. 532
Mg^{2+}	-0.002	0.903	0. 815
Ca ^{2 +}	-0.021	0. 921	0. 849
方差贡献/%	37. 4	26. 3	63. 6
Cl^{-} K^{+} Na^{+} Mg^{2+} Ca^{2+}	0. 604 0. 421 0. 11 -0. 002 -0. 021	0. 013 0. 486 0. 721 0. 903 0. 921	0. 365 0. 413 0. 532 0. 815 0. 849

2.4 潜在污染源区分析

组分

PM,5中水溶性无机离子变化特征不仅受本地 污染源的影响, 外来气团的输送也会对其造成影

响, 而冬季是四季中污染最严重的季节, 离子浓度 均有明显升高,尤其SO₄-、NO₃、NH₄ 这3种离子 的质量浓度增加明显.

为了进一步探究南郊区冬季 PM,5中水溶性无 机离子的污染来源,利用后向轨迹聚类分析方法对 到达南郊区的气团进行聚类,获得图 4(a) 所示结 果,再利用潜在源贡献因子分析方法(PSCF)对 SO_4^{2-} 、 NO_3^- 、 NH_4^+ 这 3 种离子可能的污染源区进行 解析, 获得图 4(b)~4(d)的结果显示, A 轨迹主 要来自东南方向, 该轨迹较短, 途经河北及天津, 可以发现, SO₄²、NO₅ 、NH₄ 的高 PSCF 值(PSCF 大于0.6)区域均主要集中在南郊区东南部, 重污 染源区从青岛、淄博、衡水、沧州延伸到保定天津 廊坊等地大于 0.6 的 PSCF 值逐渐增大, 颜色逐渐 加深,对大兴的离子浓度影响逐渐增强. B 轨迹主 要来自西部,对应 SO_4^{2-} 、 NO_3^{-} 和 NH_4^{+} 的中等 PSCF 值,主要污染源来自西部草原、沙漠与黄土地 区等. C 轨迹主要来自于西伯利亚, 经蒙古高原到 达观测站点, 且气流轨迹长、风速大, 有利于污染 物扩散和稀释, 由图 4(b)~4(d)对比可看到当 C 类轨迹带来的气团增多时红色区域和绿色区域的覆

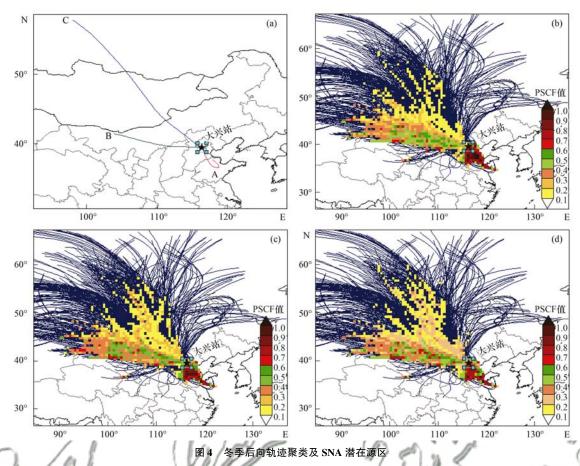


Fig. 4 Backward trajectory clustering in winter and the potential source areas for SO_4^{2-} , NO_3^{-} , and NH_4^{+}

盖面会明显减少,该类情况在对北京市区的研究中也有出现^[32],对应的低 PSCF 值,甚至有些网格区域的 PSCF 值为 0,因此 C 类气团会使南郊区污染浓度降低.

3 结论

- (1) 北京南郊区 9 种水溶性离子总浓度达到 38.6 μg·m⁻³, 其中 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 是水溶性无 机离子的主要组分; 随着 $PM_{2.5}$ 级别的增加, Ca^{2+} 、 Cl^- 、 SO_4^{2-} 、 NO_3^- 和 NH_4^+ 均有不同程度地增大, SO_4^{2-} 、 NO_3^- 和 NH_4^+ 浓度占总离子质量浓度比例明显增加,相反 Ca^{2+} 、 Cl^- 所占比例呈现降低趋势.
- (2)SO₄²⁻ 统计日变化为双峰型,峰值分别出现在 10:00 和 18:00 左右, NO₃⁻ 和 NH₄⁺ 呈单峰型,峰值出现在 10:00 左右,两种离子的日变化趋势相似.
- (3)主成分分析结果表明,南郊区主要污染物为 SO_4^2 、 NO_3^- 和 NH_4^+ ,来源为二次源. Ca^{2+} 和 Mg^{2+} 也有贡献,来源为燃煤源和土壤风沙尘及建筑扬尘的混合源.

(4)南郊区冬季 SO_4^{2-} 、 NO_3^- 及 NH_4^+ 的主要潜在污染源区位于观测点东南部,而偏北气流有利于污染物扩散和稀释.

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