



ISSN 0250-3301 CODEN HCKHDV
HUANJING KEXUE

- 主办 中国科学院生态环境研究中心
- ■出版科学出版社



2019

Vol.40 No.3 第40卷 第3期

#### ENVIRONMENTAL SCIENCE

第40卷 第3期 2019年3月15日

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# 京津冀郊区站点秋冬季大气 PM2.5 来源解析

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摘要:为了增进对京津冀地区大气  $PM_{2.5}$ 来源情况的认识,于  $2014\sim2015$  年秋冬季在京津冀地区 4 个郊区站点进行了  $PM_{2.5}$  的采样,并用化学质量平衡模型 (chemical mass balance model,CMB)进行了  $PM_{2.5}$  源解析工作。结果表明:二次颗粒物(36% ~58%)、交通(8% ~26%)、民用燃煤(8% ~16%)和生物质燃烧(5% ~16%)是京津冀郊区站点秋冬季  $PM_{2.5}$ 的主要贡献源。其中,二次硝酸盐是大部分站点秋冬季  $PM_{2.5}$ 的首要贡献源(11% ~27%)。不同污染程度的源解析显示,冬季各站点各污染源在重污染天的贡献变化趋势的同步性不如秋季明显,且秋季二次源在重污染天的贡献增加值( $47.2\sim115.7~\mu g \cdot m^{-3}$ )明显高于一次源( $29.5\sim43.4~\mu g \cdot m^{-3}$ ),但此现象在冬季不显著。对比北京市城区源解析结果,发现郊区燃煤总贡献率较为相似,但郊区燃煤源中多以民用燃煤为主,这说明对于京津冀城郊地区,控制民用燃煤源对  $PM_{2.5}$ 污染控制有重要意义。

关键词:源解析; 化学质量平衡模型(CMB); 京津冀; PM,5重污染; 民用燃煤

中图分类号: X513 文献标识码: A 文章编号: 0250-3301(2019)03-1035-08 DOI: 10.13227/j. hjkx. 201808007

# Source Apportionment of PM<sub>2.5</sub> in Suburban Area of Beijing-Tianjin-Hebei Region in Autumn and Winter

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Abstract: To identify the main sources of  $PM_{2.5}$  in Beijing-Tianjin-Hebei (BTH) region,  $PM_{2.5}$  samples were collected at four suburban sites in BTH region during autumn and winter in 2014-2015. Source apportionment of  $PM_{2.5}$  was conducted using the chemical mass balance model (CMB). It shows that the main sources of  $PM_{2.5}$  in autumn and winter were secondary aerosols (36%-58%), traffic (8%-26%), residential coal combustion (8%-16%), and biomass burning (5%-16%). Secondary nitrate was the most important source of  $PM_{2.5}$  at most sites during autumn and winter (11%-27%). The source apportionment at different pollution levels indicates that the coherence of the increasing trend of different sources among the four sites were much more obvious in autumn than in winter. Also, the increasing contribution of secondary sources (47.2-115.7  $\mu$ g·m<sup>-3</sup>) was much higher than that of primary sources (29.5-43.4  $\mu$ g·m<sup>-3</sup>) in autumn, but such trend was not significant in winter. The total contribution of coal combustion at suburban sites was quite similar to that in urban sites, but in suburban areas residential coal combustion dominates the contribution from coal combustion. Thus, it is very necessary for suburban areas of the BTH region to control emissions from residential coal combustion.

Key words: source apportionment; chemical mass balance model (CMB); Beijing-Tianjin-Hebei region; heavy PM<sub>2.5</sub> pollution; residential coal combustion

京津冀地区细颗粒物  $(PM_{2.5})$  污染频发,污染程度重. 2015 年北京市、天津市和河北省环境质量公报显示 $[1^{-3}]$ ,京津冀  $PM_{2.5}$  年均浓度值  $(81、70和77 \mu g \cdot m^{-3})$  严重超过国家环境空气质量二级标准的年均限值  $(35 \mu g \cdot m^{-3})$ ,污染形势严峻。京津冀地区  $PM_{2.5}$  重污染过程多发生在秋冬两季。据统计,京津冀地区在 2013~2014 年期间长时间大范围的重污染过程共发生 31次,其中 23次均发生在秋冬两季[4].

细颗粒物来源解析对 PM<sub>2.5</sub>减排和污染控制具有重要意义. 受体模型作为主要的源解析方法在京津冀地区源解析工作中被广泛使用<sup>[5~14]</sup>. 受体模型解析结果表明, 燃煤、交通、扬尘以及二次气溶胶

是京津冀地区  $PM_{2.5}$ 的主要来源,且近年来京津冀地区  $PM_{2.5}$ 污染从燃煤主导[5.6]的煤烟型污染发展为燃煤和机动车 $[7^{-14}]$ 复合型污染. 污染源构成的复杂性与变化性,要求大气  $PM_{2.5}$ 源解析工作不断更新,以反映污染源的实际变化.

现有京津冀地区的源解析研究多集中在城市地区<sup>[6~12]</sup>,针对郊区或者农村地区的源解析<sup>[5,13,14]</sup>工作较少. 然而因为生产、生活方式的不同,城镇和

收稿日期: 2018-08-01; 修订日期: 2018-09-05

基金项目: 国家自然科学基金项目(21625701); 大气重污染成因与治理攻关项目(DQGG0301)

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郊区的大气  $PM_{2.5}$ 来源可能有较大差异 $^{[5,13]}$ . 为识别京津冀郊区大气  $PM_{2.5}$ 的来源,本研究在京津冀地区布置了 4 个郊区站点,开展了秋冬季大气  $PM_{2.5}$ 的样品采集工作,并利用化学质量平衡模型 (chemical mass balance, CMB)进行了分季节以及分污染程度的源解析,旨在为京津冀郊区地区大气  $PM_{2.5}$ 的控制提供科学依据.

#### 1 材料与方法

#### 1.1 样品采集

本研究在北京、天津、廊坊设置了 4 个站点(如图 1),其中北京市包括琉璃河和永乐店站.琉璃河站位于北京市西南部房山区的琉璃河镇,与河北省涿州市接壤,交通发达.永乐店站位于北京市东南部通州区的永乐店镇,交通上临近首都环线高速(1.6 km).天津市的站点位于宁河区岳龙镇(宁河站),是天津市与河北东部交界处.河北省的站点则位于廊坊市大城县(廊坊站),地处河北省与天津市东南部交界处.以上 4 个站点均位于郊区,周围村庄较为密集.



图 1 京津冀地区 4 个站点分布情况

Fig. 1 Site location in Beijing-Tianjin-Hebei region

本研究于 2 个采样时间段进行  $PM_{2.5}$  的采集 (2014年10月10日~11月14日和2015年1月4日~2月4日),分别代表京津冀地区秋冬季  $PM_{2.5}$  的污染情况. 采样仪器为 Thermo RP2300(琉璃河、永乐店、宁河)和 Thermo RP2000(廊坊). 采样器设两通道,其中一通道置 Teflon 膜(47 mm),采样流量16.7  $L \cdot min^{-1}$ ;二通道置石英膜(47 mm),采样流量10  $L \cdot min^{-1}$ . 采样时间为每天上午10:00至次日上午9:30,时长共计23.5 h. 采样前,将铝箔纸包覆的石英膜放入马弗炉里在550℃的条件下加热6h以上,去除膜上的残留的杂质. 将 Teflon 膜在采样前后分别使用精度为0.01 mg 或0.001 mg 的电子天平在恒温、恒湿控制实验室中进行称重,用于测定  $PM_{2.5}$ 质量浓度. 在离线采样的同时,采用美国热电公司生产的 Thermo 42/43/48/49 I(琉璃河、永乐店、宁河)和

美国 Teledyne 公司生产的 API T100/200/300/400(廊坊)进行常规六项气体污染物的监测( $SO_2$ 、 $NO_2$ 、 $O_3$ 、 $PM_{2.5}$ 、 $PM_{10}$ 和 CO),监测分辨率分别为 5 min(琉璃河、永乐店)和 1 h(宁河、廊坊).

**1.2** PM<sub>2.5</sub>样品中碳组分、无机离子组分和无机元素的化学分析

 $PM_{2.5}$ 样品中的碳组分分析选用美国沙漠研究所(desert research institute, DRI)开发的Model2001A型热/光碳分析仪,并按照IMPROVE-A分析程序和反射修正法(TOR)进行炭化校正.取石英膜超声 1 h 后的上清液,采用美国 DIONEX 公司的 Dionex ICS-3000 离子色谱仪进行无机离子的分析,共分析了包括  $F^-$ 、 $Cl^-$ 、 $NO_3^-$ 、 $SO_4^{2-}$ 、 $NH_4^+$ 、 $K^+$ 、 $Na^+$ 、 $Ca^{2+}$ 、 $Mg^{2+}$ 在内的 9 种离子. 无机元素的测定采用 X 射线荧光法(X-ray fluorescence spectrometer,XRF),分析仪器为日本 Rigaku 公司生产的 RIX3000,测定了包括 Mg、Al、Si、P、S、K、等 27 种元素.

### 1.3 PM<sub>2.5</sub>样品质量控制

将各站点秋冬季离线样品称重结果和颗粒物在线监测质量浓度进行对比,发现各站点二者相关性均较好,秋季  $R^2$  在 0.79 以上,冬季永乐店  $R^2$  略低为 0.66,其他站点  $R^2$  均在 0.89 以上,将  $PM_2$ 。离线质量和各组分质量浓度加和相比较,发现各站点秋冬季两者相关性均良好,秋季  $R^2$  在  $0.79 \sim 0.98$ ,冬季  $R^2$  在  $0.86 \sim 0.97$ . 阴阳离子平衡分析显示,各站点秋冬两季阴阳离子相关性较好, $R^2$  在  $0.90 \sim 0.99$ . 基于以上分析,剔除离群点,得到秋季各站点有效样品 109 个,冬季有效样品 112 个.

#### 1.4 来源解析模型

CMB 模型由一组线性方程组的最小二乘解组成,它表示受体颗粒物中某种化学组分的浓度等于所有污染源中该化学组分的质量分数和该污染源对受体的贡献浓度值的乘积的线性和<sup>[15]</sup>,其数学表达式为:

$$c_i = \sum_{j=1}^J F_{ij} \times S_j$$

式中, $c_i$  为大气颗粒物中化学组分 i 的质量浓度, $\mu g \cdot m^{-3}$ ;  $F_{ij}$  为污染源 j 排放的颗粒物中化学组分 i 的质量分数, $g \cdot g^{-1}$ ;  $S_j$  为颗粒物污染源 j 贡献的质量浓度, $\mu g \cdot m^{-3}$ . 将颗粒物污染源中化学成分的质量分数及其不确定度与大气颗粒物样品中化学成分的浓度及其不确定度输入到 CMB 模型中,采用有效方差最小二乘法[15] 进行计算,可以得到各污染源对大气颗粒物中不同化学组分的贡献浓度值与总贡献率以及不确定度.

#### 1.5 颗粒物源成分谱

根据 2012 年京津冀地区 PM<sub>2.5</sub> 排放清单<sup>[16]</sup> 可知,京津冀地区 PM<sub>2.5</sub> 的主要一次污染源有工业、机动车、扬尘等. 其中工业部门以工业燃煤为主,同时考虑钢铁和水泥生产. 由于郊区存在秋季秸秆焚烧现象以及冬季散煤采暖现象,污染源也考虑了民用燃煤和生物质燃烧源. 基于以上,本研究考虑的一次污染源有城市扬尘、工业燃煤、钢铁生产、民用燃煤、汽油车、柴油车以及生物质燃烧. 对于二次气溶胶没有直接对应的源,则考虑二次硫酸盐以及二次硝酸盐的虚拟源谱. 对于二次有机物,采用最小 OC/EC 比值法<sup>[12]</sup>进行估算.

通过调研<sup>[11,16,18-29]</sup>,收集以上污染源源谱,结果见表 1. 其中城市扬尘、民用燃煤、生物质燃烧直接采用文献 [11,22,23] 中的结果. 由于汽油车 PM<sub>2.5</sub>排放较低,很少有本地 PM<sub>2.5</sub>全组分研究,所以参考美国环保署 Speciate 4. 4 源谱库中汽油车排放的综合源谱 [<sup>24]</sup>. 在搜集已有相关研究结果 [<sup>25~28]</sup> 的基础上,根据京津冀地区清单结果中不同排放标准柴油车对柴油车总 PM<sub>2.5</sub>排放量的贡献比例 [<sup>16]</sup>,对不同排放标准的柴油车源谱进行加权平均,得到了柴油车综合源谱. 针对钢铁生产,根据各个生产环节(烧结、高炉、炼钢以及配料、整粒等) PM<sub>2.5</sub>的排放量 [<sup>16]</sup> 对各环节排放颗粒物的源谱 [<sup>20,21]</sup> 进行了加权平均.

表 1 京津冀地区 PM25主要污染源源谱/%

			Table 1			I <sub>2.5</sub> of Beijing	-Tianjin-Hebei r	egion/%	10	~ 1
项目	城市払小[11]	工业燃煤[18,19]		失生产[16,20		民用燃煤[22]	生物质燃烧[23]	汽油车[24]	柴油车[26,25~28]	水泥生产[18,22,29]
- 火口	姚巾100王。	工业涨床	北京	天津	河北	氏用 燃 殊 。	生物灰然死。	17個十二	未佃干	八化生)
OC	$6.9 \pm 2.3$	$11 \pm 3.8$	$3.2\pm0.9$	6. $5 \pm 3.2$	$5.7 \pm 2.7$	$18 \pm 28$	34 ± 14	$55 \pm 6.3$	23 ± 4. 1	13 ± 13
EC	$1.7 \pm 1.7$	$2.0 \pm 1.6$	0. $7 \pm 0.7$	$1.7 \pm 1.3$	1. $3 \pm 1$ . 1	$0.2 \pm 0.1$	3. $0 \pm 0.7$	19 ± 14	$55 \pm 9.0$	$2.4 \pm 3.0$
$NH_4^+$	$0.7 \pm 0.1$	$2.0 \pm 1.7$	$0.~0\pm0.~0$	$0.\ 2\pm0.\ 2$	$0.2 \pm 0.2$	$0.3 \pm 0.3$	$3.0 \pm 0.8$	$1.7 \pm 1.2$	$0.8 \pm 1.0$	$0.2 \pm 0.1$
$NO_3^-$	$0.6 \pm 0.9$	$0.4 \pm 0.4$	$0.3 \pm 0.2$	$0.6 \pm 0.5$	$0.7 \pm 0.5$	$0.4 \pm 0.6$	$0.6 \pm 0.2$	$0.2 \pm 0.1$	$1.3 \pm 0.9$	$0.2 \pm 0.1$
$SO_4^2$	$6.5 \pm 9.0$	$46 \pm 9.3$	9. $5 \pm 4.3$	$9.8 \pm 4.6$	11 ±4.2	$15 \pm 21$	1. 9 ± 1. 1	$0.9 \pm 3.6$	$3.4 \pm 3.4$	1.7 ± 1.1
Cl 7	$0.7 \pm 0.5$	$0.8 \pm 0.3$	$2.9 \pm 0.7$	$3.7 \pm 1.5$	$3.3 \pm 1.2$	$2.7 \pm 3.8$	$23 \pm 7.1$	$0.1 \pm 0.3$	$0.5 \pm 0.0$	2. 9 ± 1. 8
Al	$5.5 \pm 0.3$	$0.4 \pm 0.2$	$0.5 \pm 0.1$	$2.8 \pm 2.2$	2. 8 ± 2. 4	$0.1 \pm 0.1$	$0.0 \pm 0.0$	$0.2 \pm 0.1$	$0.1 \pm 0.0$	$0.4 \pm 0.3$
Si	16 ± 1.8	$2.3 \pm 1.2$	$2.7 \pm 0.2$	$6.\ 1\pm2.\ 8$	$5.9 \pm 3.0$	$0.4 \pm 0.2$	0. $1 \pm 0.0$	$0.4 \pm 0.1$	$0.1 \pm 0.0$	$3.7 \pm 2.9$
Na	$1.5 \pm 0.2$	$9.3 \pm 2.3$	$2.8 \pm 0.8$	3. $1 \pm 0.9$	$3.4 \pm 0.8$	$0.9 \pm 1.4$	$0.2 \pm 0.1$	$0.0 \pm 0.1$	$0.7 \pm 0.0$	$5.5 \pm 4.9$
Mg	$1.2 \pm 0.6$	$0.1 \pm 0.1$	$0.9 \pm 0.7$	$1.3 \pm 0.4$	1. $3 \pm 0.4$	$0.0 \pm 0.0$	$0.1 \pm 0.0$	$0.1 \pm 0.1$	$0.0 \pm 0.0$	$2.2 \pm 2.2$
K	$0.9 \pm 0.7$	$2.1 \pm 1.6$	$6.0 \pm 2.2$	$6.\ 7\pm2.\ 4$	$5.8 \pm 2.2$	$5.0 \pm 7.1$	$6.9 \pm 3.5$	$0.1 \pm 0.0$	$0.3 \pm 0.0$	$0.7 \pm 0.2$
Ca	$8.0 \pm 1.3$	$0.6 \pm 0.1$	6. $5 \pm 0.1$	6. $6 \pm 0$ . 1	6. $5 \pm 0.1$	$0.2 \pm 0.2$	$0.1 \pm 0.0$	$0.4 \pm 0.6$	0. $8 \pm 0.1$	$0.4 \pm 0.5$
Ti	$0.6 \pm 0.3$	$0.0 \pm 0.0$	$0.\ 5\pm0.\ 2$	$0.5 \pm 0.3$	$0.6 \pm 0.2$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	0. $1 \pm 0.0$	6. $1 \pm 1.4$
Cr	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.~0\pm0.~0$	$0.\ 1\pm0.\ 1$	$0.1 \pm 0.1$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.1 \pm 0.0$	$0.6 \pm 0.7$
Mn	$0.0 \pm 0.0$	$0.0 \pm 0.0$	2. $1 \pm 1.3$	$1.0 \pm 0.9$	$1.1 \pm 0.9$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	0. $1 \pm 0.0$	$0.3 \pm 0.4$
Fe	$2.7 \pm 0.2$	$1.4 \pm 0.8$	$34 \pm 26$	$24 \pm 9.4$	$25 \pm 8.7$	$0.2 \pm 0.1$	$0.0 \pm 0.0$	$0.4 \pm 0.9$	$0.5 \pm 0.1$	$0.4 \pm 0.5$
Ni	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.\ 1\pm0.\ 0$	$0.1 \pm 0.0$	$0.1 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.3 \pm 0.1$	$1.1 \pm 0.9$
Cu	$0.0 \pm 0.1$	$0.0 \pm 0.0$	$0.~0~\pm 0.~0$	$0.\ 2\pm0.\ 1$	$0.2 \pm 0.1$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	0. $1 \pm 0.1$	$0.0 \pm 0.0$	$0.0 \pm 0.0$
Zn	$0.1 \pm 0.0$	$1.7 \pm 0.7$	4. $1 \pm 0.3$	$1.~8~\pm1.~8$	$2.1 \pm 1.8$	$0.4 \pm 0.5$	$0.0 \pm 0.0$	$0.3 \pm 0.3$	$0.1 \pm 0.1$	$0.0 \pm 0.0$
Pb	$0.0 \pm 0.0$	$0.8 \pm 0.4$	$0.2 \pm 0.1$	1. $8 \pm 2$ . 1	$1.2 \pm 1.8$	$0.7 \pm 0.7$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.0 \pm 0.0$	$0.2 \pm 0.2$

#### 2 结果与讨论

**2.1** NO<sub>2</sub> 与 PM<sub>2.5</sub> 中 NO<sub>3</sub> 、SO<sub>2</sub> 与 PM<sub>2.5</sub> 中 SO<sub>4</sub> 相 关性分析

秋冬季各站点  $NO_2$  与  $NO_3^-$ 、 $SO_2$  与  $SO_4^{2-}$  均显示出一定的时间同步性(图 2,以琉璃河站秋季样品为例). 相关性分析发现(表 2), $NO_2$  与  $NO_3^-$  相关性较强,除宁河站点秋季受样品数量影响外,其他站点秋冬两季二者的相关性系数均在 0.628~0.869(P<0.05),且冬季相关性较秋季更为明显.此相关性与下文提到的二次硝酸盐对大气  $PM_{2.5}$ 的贡献较为突出相吻合.  $SO_2$  与  $SO_4^{2-}$  之间的相关性较  $NO_2$  与  $NO_3^-$  弱,除宁河、廊坊站点秋季,琉璃河

冬季样品外, 弱或中等相关性显著( $R^2$ 为 0.394 ~ 0.636, P < 0.05).

#### 2.2 京津冀地区各站点秋冬季源解析结果

秋季各站点源解析结果如图 3(a) 所示. 二次源对  $PM_{2.5}$  的贡献明显  $(38.2 \sim 59.1 \ \mu g \cdot m^{-3})$ ,占  $PM_{2.5}$  总质量的  $36\% \sim 56\%$ . 其中,二次硝酸盐在各站点均为首要的污染源  $(19.4 \sim 31.3 \ \mu g \cdot m^{-3})$ ,其贡献率在  $19\% \sim 27\%$ ;其次是二次有机物  $(14.9 \sim 19.4 \ \mu g \cdot m^{-3})$ ,贡献率在各站点较为相似  $(14\% \sim 17\%)$ . 对于琉璃河和永乐店站点,汽油车的贡献十分显著  $(15\% \ n \ 16\%)$ . 这可能是由于琉璃河站点和永乐店站点位于省间边界,临近省间运输要道(琉璃河站点距 G107 国道约  $2 \ km$ ,永乐店站点距

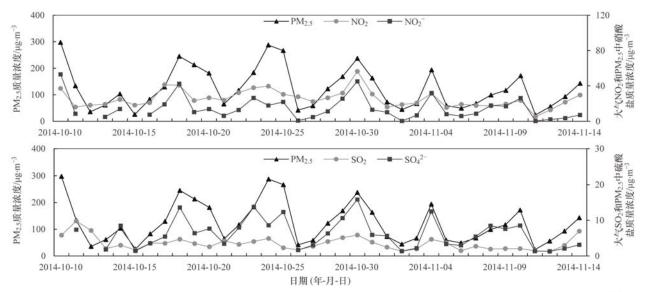


图 2 秋季琉璃河站点大气  $SO_2$ 、 $NO_2$  和  $PM_{2.5}$ 中硫酸根、硝酸根日均浓度

Fig. 2 Daily average concentration of SO<sub>2</sub>, NO<sub>2</sub>, sulfate and nitrate in PM<sub>2.5</sub> at the Liulihe site in autumn

表 2 秋冬季各站点  $SO_2$ ,  $NO_2$  与  $PM_{2.5}$ 中硫酸根、硝酸根相关性分析

Table 2 Correlation analysis of SO<sub>2</sub>, NO<sub>2</sub> and sulfate, and nitrate in PM<sub>2.5</sub> at all sites

		SO, 与	SO <sub>4</sub> -		10	NO <sub>2</sub> 与	NO <sub>3</sub>	
站点	秋	李 /	~ / 《 冬季	È	利	(季		冬季
	$R^2$	P	$R^2$	P	$R^2$	RU	$R^2$	B
琉璃河	0. 421	0. 013 2	0.295	0. 102	0. 739	$6.02 \times 10^{-7}$	0. 869	1. 10E – 10
永乐店	0. 594	0.000132	0.513	0.00318	0.713	$1.05 \times 10^{-6}$	0.755	9. 33E – 07
宁河	0.101	0.608	0. 394	0. 0311	0. 435	0. 020 7	0.641	0. 000 137
廊坊	0.348	0. 0647	0.636	9. $29 \times 10^{-5}$	0.628	0. 000 269	0. 838	2. 19E – 09

离首都环线高速 1.5 km),省际运输排放强度较高<sup>[30]</sup>导致的.对于宁河和廊坊站点,生物质燃烧是主要的一次贡献源,其贡献值和贡献率分别为 14.5~17.8 μg·m<sup>-3</sup>和 15%~16%.参考美国国家航空航天局公布的卫星火点图可知[图 4(a)],较高的生物质燃烧贡献可能与京津冀地区秋季秸秆开放燃烧现象有关.同时廊坊站点的工业燃煤贡献(13%)明显高于其他站点(3%~6%),这与河北省高污染重工业较为密集相一致.

冬季,二次气溶胶对  $PM_{2.5}$ 的贡献仍旧明显[图 3(b)],贡献值为  $50.1 \sim 82.8 \ \mu g \cdot m^{-3}$ ,分担率为  $41\% \sim 58\%$ . 其中,永乐店和宁河站的二次硝酸盐 贡献仍最为显著(23% 和 18%),但琉璃河和廊坊站,二次有机物的贡献(24% 和 17%)凸显. 相比秋季,宁河和廊坊站冬季二次气溶胶的贡献无明显变化,但北京两站点冬季二次气溶胶的贡献明显增加,增加值分别为  $19.3 \ \mu g \cdot m^{-3}$  和  $19.2 \ \mu g \cdot m^{-3}$ . 冬季  $PM_{2.5}$ 的主要一次污染源仍是汽油车、民用燃煤和生物质燃烧,其贡献值分别为  $15.4 \sim 32.7$ 、 $14.4 \sim 18.7 \ 和 7.2 \sim 12.8 \ \mu g \cdot m^{-3}$ . 相比秋季,冬季汽油车和民用燃煤对  $PM_{2.5}$ 的贡献值增加明显(5~17  $\mu g \cdot m^{-3}$ 和  $5 \sim 11 \ \mu g \cdot m^{-3}$ ). 这可能是因为冬季

气温较低,机动车排放因子有所增加<sup>[31]</sup>,民用散煤燃烧量增加,且因为冬季静稳性天气形势频繁出现,扩散条件较差<sup>[32]</sup>,导致近地面污染源对 PM<sub>2.5</sub> 贡献较显著。由于冬季采暖,民用燃煤源冬季贡献理应增加更显著,但可能是因为秋季采样时段临近采暖季,散煤供暖的居民提前供暖导致秋季的结果也包含了采暖的影响,所以秋冬季结果差异不明显。北京郊区 PM<sub>2.5</sub> 排放清单结果<sup>[33]</sup>也表明,11 月和1 月民用燃煤对 PM<sub>2.5</sub> 排放量的贡献仅差 10%,但和其他月相差 50% 以上。冬季生物质燃烧源的贡献整体较秋季低(5%~9%),这与图 4(b)中火点密度下降相一致。这说明进入冬季,仍有一定的生物质开放燃烧现象,但强度明显下降。

#### 2.3 区分污染程度的京津冀郊区源解析结果

根据国家"环境空气质量指数 (air quality index, AQI)技术规定(试行)"中  $PM_{2.5}$ 在良和重度污染级别下的标准浓度 (75  $\mu g \cdot m^{-3}$  和 150  $\mu g \cdot m^{-3}$ ),将  $PM_{2.5}$ 样品分为三类: $PM_{2.5}$ 质量浓度为0~75、75~150 和大于150  $\mu g \cdot m^{-3}$ .分别对这三类样品做源解析,得到区分污染程度的秋冬季源解析结果 (如图 5 和图 6).

秋季,对比重污染天和清洁天,各站点二次污

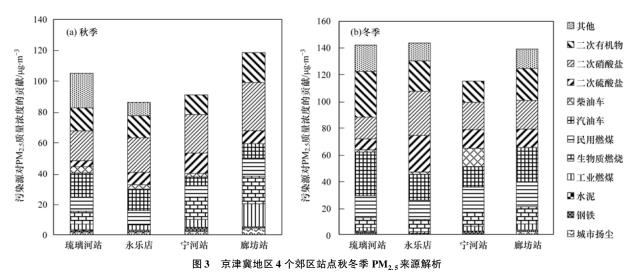


Fig. 3 Source apportionment of PM<sub>2.5</sub> at four suburban sites of Beijing-Tianjin-Hebei region in autumn and winter

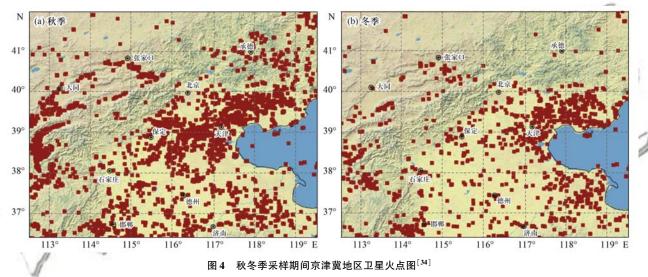


Fig. 4 Firemap of Beijing-Tianjin-Hebei region during the sampling period in autumn and winter

染源的贡献值增加幅度(47.2~115.7 μg·m<sup>-3</sup>)明显高于一次源(29.5~43.4 μg·m<sup>-3</sup>),相应地重污染天的二次源  $PM_{2.5}$ 总质量分担率(38% ~71%)也

明显高于清洁天(33%~47%). 这表明了  $SO_2$  和  $NO_x$  等气态前体物的二次转化是重污染天气  $PM_{2.5}$  的重要成因. 4 个站点二次硝酸盐的贡献在重污染

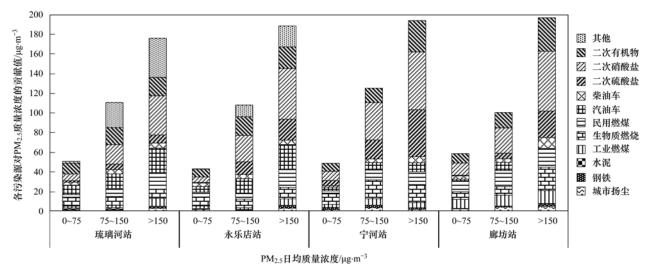


图 5 秋季京津冀地区 4 个郊区站点不同污染水平大气 PM<sub>2.5</sub>来源解析

Fig. 5 Source apportionment of PM<sub>2.5</sub> in four suburban areas of the Beijing-Tianjin-Hebei region in autumn based on different pollution levels

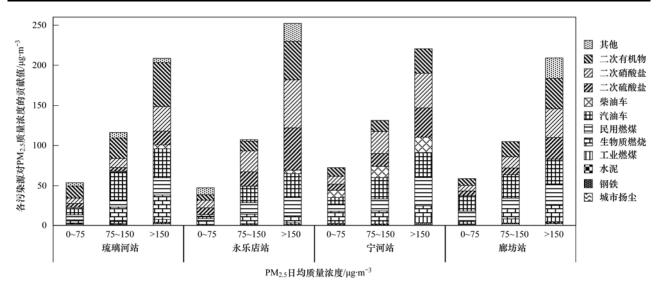


图 6 冬季京津冀地区 4 个郊区站点不同污染水平下的大气 PM<sub>2.5</sub>来源解析

Fig. 6 Source apportionment of PM2.5 in four suburban areas of the Beijing-Tianjin-Hebei region in winter based on different pollution levels

时均有明显增加(清洁 13% ~ 21%, 重污染 23% ~ 31%), 二次硫酸盐的贡献除琉璃河站点外,均有明显增加(清洁 1% ~ 11%, 重污染 11% ~ 25%). 琉璃河站点的二次硫酸盐贡献无论是清洁天(3%)还是重污染天(4%)均不明显,这与琉璃河站点大气  $SO_2$ 浓度(3.6  $\mu g \cdot m^{-3}$ )较低有关. 一次污染源中,琉璃河和永乐店站民用燃煤和汽油车贡献在重污染天增加明显(13.1 ~ 13.8  $\mu g \cdot m^{-3}$  和 15.2 ~ 20.1  $\mu g \cdot m^{-3}$ ), 但宁河和廊坊站则是生物质燃烧和民用燃煤在重污染天增加明显(7.8 ~ 16.2  $\mu g \cdot m^{-3}$  和 7.8 ~ 9.7  $\mu g \cdot m^{-3}$ ).

冬季重污染期间各污染源的贡献情况明显较秋季复杂. 重污染期间,除永乐店站,二次源的贡献值增加量(45.3~79.8 μg·m<sup>-3</sup>)仍高于一次源(76.1~82.0 μg·m<sup>-3</sup>),但二者间的差异明显小于秋季. 对于永乐店站,重污染天二次源增加值明显(135.6 μg·m<sup>-3</sup>),显著高于其他站点,从增长率来看,其中二次有机物增长比例最高(约为6倍),这可能是与永乐店地区冬季 VOCs 污染显著有关[35].一次污染源中,所有站点民用燃煤在重污染天的贡献值增加明显(15.3~26.1 μg·m<sup>-3</sup>),其增加值也明显高于秋季.

整体来看,冬季各站点间重污染天各污染源贡献的变化趋势较秋季同步性下降.这可能是因为冬季气温低,二次气溶胶转化率下降,导致冬季重污染过程中二次源贡献增加值不如秋季突出,与此同时本地一次源的贡献较秋季显著.另一方面,也可能是因为冬季风速较低,区域传输影响较秋季小<sup>[36]</sup>,导致本地源主导.因此,秋季重污染应急应在区域层面上调控,而冬季则更应加强本地污染源控制.

#### 2.4 城区与郊区来源解析结果对比

将本研究北京两郊区站的源解析结果与相似采样时间的研究中北京市城区源解析结果[8.13] 相对比,发现本研究得到的燃煤总贡献率(12%~15%)与文献中的结果(12%~23%)较为相似. 但本研究区分了民用燃煤和工业燃煤的贡献,发现琉璃河站和永乐店站民用燃煤贡献率均高于工业燃煤,说明北京市燃煤排放控制的重点是民用散煤. 相比已有研究中二次源的贡献率(16%~47%),本研究二次源贡献(37%~58%)略高,这可能是因为本研究两站点分别位于北京西南和东南传输通道上,区域传输的影响比城区站点更明显. 另外,本研究交通源的贡献(15%~24%)略高于文献结果(9%~18%),这可能是因为本文两郊区站均临近省间交通要道,省际运输对站点周围大气 PM2.5的影响较为明显.

#### 3 结论

- (1)4 个郊区站点秋冬季源解析结果显示:二次颗粒物 $(36\% \sim 58\%)$ 、交通 $(8\% \sim 26\%)$ 、民用燃煤 $(8\% \sim 16\%)$ 和生物质燃烧 $(5\% \sim 16\%)$ 是京津冀郊区秋冬季  $PM_{2.5}$ 的主要来源.
- (2)针对重污染天的源解析发现,二次气溶胶在重污染期间贡献率增加明显,其中秋季二次源对重污染的贡献比冬季更为显著,且秋季各站点的污染来源变化呈现出更强的同步性.
- (3)尽管琉璃河和永乐店两个郊区站点燃煤源对 PM<sub>2.5</sub>的总贡献率与北京城区较为接近,但郊区站点的燃煤源以民用燃煤为主,说明通过清洁能源替代民用散煤对京津冀大气 PM<sub>2.5</sub>控制有重要意义.

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《环境科学》2019年全年12期.

国内统一连续出版物号: CN 11-1895/X

国际标准连续出版物号: ISSN 0250-3301

国外发行代号:M 205

国内邮发代号:2-821

编辑部地址:北京市海淀区双清路 18 号(2871 信箱) 邮编:100085

电话:010-62941102;传真:010-62849343;E-mail:hjkx@rcees. ac. cn;网址:www.hjkx. ac. cn

# **HUANJING KEXUE**

Environmental Science (monthly)

Vol. 40 No. 3 Mar. 15, 2019

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