

(HUANJING KEXUE)

ENVIRONMENTAL SCIENCE

第36卷 第8期

Vol.36 No.8

2015

中国科学院生态环境研究中心 主办

斜学出版社出版



ENVIRONMENTAL SCIENCE

第36卷 第8期 2015年8月15日

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夏季珠江三角洲地区 $PM_{2.5}$ 化学组分特征及其对大气能见度的影响

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摘要:于2010年夏季在珠三角主要城市广州、佛山、东莞、深圳、珠海以及广州郊区从化同步采集 $PM_{2.5}$ 样品,利用热光反射碳分析仪和离子色谱分别分析样品中有机碳/元素碳和水溶性离子浓度,并同步收集能见度和气象数据.在此基础上对珠三角主要城市大气 $PM_{2.5}$ 中主要化学成分的浓度水平和空间分布特征进行分析,并利用 IMPROVE 方程重建大气消光系数,探讨 $PM_{2.5}$ 的主要化学组分对大气能见度的影响.结果发现,观测期间珠三角地区 $PM_{2.5}$ 中的主要化学成分空间分布特征明显,广州、佛山和东莞浓度较高,珠海和深圳浓度较低. $(NH_4)_2SO_4$ 、有机物(OM)、EC 和 NH_4NO_3 对夏季珠三角大气消光系数贡献率分别为 39%、31%、12% 和 13%.

关键词:霾;空间分布; PM,5; 化学组分; 大气能见度

中图分类号: X513 文献标识码: A 文章编号: 0250-3301(2015)08-2758-10 DOI: 10.13227/j. hjkx. 2015.08.005

Chemical Compositions in $PM_{2.5}$ and Its Impact on Visibility in Summer in Pearl River Delta, China

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Abstract: Aerosol samples of $PM_{2.5}$ were collected simultaneously at 6 sites from five cities (Guangzhou urban, Conghua (suburban of Guangzhou), Foshan, Dongguan, Shenzhen and Zhuhai) in Pearl River Delta region during the summer of 2010. The concentrations of organic carbon (OC), elemental carbon (EC) and water-soluble ions were determined by thermal/optical carbon analyzer and ion chromatography, respectively. The characteristics of $PM_{2.5}$, OC, EC and ions, spatial distribution were discussed. Moreover, ambient light extinction coefficients were reconstructed by IMPROVE formula. The results showed that spatial distribution characteristics of $PM_{2.5}$ and its chemical compositions were obviously different. The $PM_{2.5}$ in Guangzhou, Foshan and Dongguan were higher than those in Zhuhai and Shenzhen. The contributions of $(NH_4)_2SO_4$, OM, EC and NH_4NO_3 to ambient light extinction coefficient were 39%, 31%, 12% and 13%, respectively.

Key words: haze; spatial distribution; PM2.5; chemical composition; visibility

由于我国经济的快速发展,化石燃料的大量消耗,使得我国东部沿海大部分地区霾天气频繁发生^[1-3].现有的研究表明,霾天气的发生与大气环境中 PM_{2.5}(指空气动力学直径 < 2.5 μm 的大气颗粒物)密切相关^[4-7]. PM_{2.5}的主要化学成分如硫酸盐、硝酸盐、有机物和元素碳等均具有较强的消光能力^[8,9].因此,了解区域内 PM_{2.5}中主要化学成分及其浓度特征对于进一步了解区域大气能见度下降或霾天气的成因具有重要意义.

广东省位于我国南部,面积42 794 km²,人口近3 870万,是全国经济最发达的地区之一,同时也是我国大气污染重点控制区之一.近几十年来,珠三

角地区大气能见度呈明显下降趋势^[2]. 目前,对珠三角有关大气能见度下降问题的研究主要集中于广州,对珠三角其它城市的研究报道很少^[6,10~22]. 因此,同步开展该区域主要城市 PM_{2.5}中主要化学成分特征研究,对于较为全面地了解珠三角区域性大气能见度下降的原因,制定切实可行的治理政策具有重要的指导作用.

考虑到夏季南海季风的影响,珠三角地区大气 PM_{2.5}浓度在夏季较低,其主要化学成分一定程度上

收稿日期: 2014-11-24; 修订日期: 2015-03-21

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可以代表珠三角本地污染背景,对于进一步研究其它季节 PM_{2.5}区域性输送和能见度下降研究具有重要参考价值.为此,本研究于 2010 年 8 月 7 ~ 21 日连续 15 d 在深圳、东莞、广州、从化、佛山、珠海等珠三角这 6 个地点同步采集 24 h PM_{2.5}样品,并获取 PM_{2.5}中主要化学成分 OC、EC 和水溶性离子等浓度数据,同步还收集了能见度、风速、温湿度、降雨等气象资料.在此基础上,分析了珠三角主要城市和郊区 PM_{2.5}中主要化学成分与大气能见度之间的关系.

1 材料与方法

1.1 采样地点

珠江三角洲是西江、北江和东江流入南海冲积 沉淀而形成的三角洲,位于广东省中南部,珠江下 游,南海北部,毗邻香港和澳门,是我国重要的加工制造和高新技术产业基地,也是我国经济发展最迅速、最发达的区域之一. 其地处热带和亚热带,受南亚热带季风气候影响,冬季少雨多吹北风,夏季多雨多吹南风.

据地理位置本研究选取珠江三角洲地区深圳、东莞、广州、从化、佛山和珠海这6个采样地点同步采集 PM_{2.5}样品. 具体采样位置的分布见表1和图1.

1.2 样品采集

于 2010 年 8 月 7 ~ 21 日利用美国 MiniVol 颗粒物采样器 (air metrics corporation) 每天采集一个 $PM_{2.5}$ 样品. 采样时间为当日的 10:00 到次日的 10:00. 整个观测期间每个点位采集 15 个样品 (其中深圳为 13 个样品), 2 个空白样品. 采样器流量为5.0 $L\cdot min^{-1}$, 使用的滤膜为直径47 mm的石英

表 1 珠三角观测点位概况

	Table 1	Surrounding environment of the	sampling sites in Pearl River Delta		
城市	方 采样点位	采样高度/m	经纬度	备注	
深圳	川 洪湖公园	8	22°34′55″N, 114°07′43″E	城市	
东莞	· 东莞第二水厂	20	23°03′37″N, 113°44′55″E	城市	
广州	性南环境科学研究所	50	23°07′26″N, 113°21′17″E	城市	
佛山	山 南海政协	25	23°01′59″N, 113°08′15″E	城市	
从作	と 英豪学校	20	23°41′47″N, 113°42′08″E	郊区	
珠洋	中山大学珠海分校	50	22°20′49″N 113°35′05″F	郊区	

24.0° ○ 采样地点 23.89 23.69 23.4° 23.2 23.0° 东莞 22.89 22.69 22.4° 22.29 22.09 112.89 114.4° E 112.49

图 1 珠江三角洲六采样点位置分布示意

Fig. 1 Geographical locations of 6 sampling sites in Pearl River Delta

膜(Whatman, England). 滤膜在采样前用 800℃ 高温灼烧 4 h,冷却后放在恒温恒湿箱(温度 20~

23℃,相对湿度 35%~45%)平衡 24 h,然后再进行称重. 称重方法依据美国 EPA 规定方法 用精度为 1 μg 的电子天平进行称量,且前后两次称量的误差小于 $10~\mu g^{[23]}$.

1.3 样品化学成分分析

所有样品的 OC(organics carbon)和 EC(element carbon)分析采用美国沙漠研究所研制 DRI Model 2001 热光碳分析仪(Thermal/Optical Carbon Analyzer).应用IMPROVE 热光反射的实验方法,该方法的主要测试原理见文献[24].

剪取四分之一的石英膜加入 10 mL 去离子水 $(R>18.2 \text{ M}\Omega\cdot\text{cm})$,超声萃取 1 h,采用 $0.45 \text{ }\mu\text{m}$ 的过滤器过滤定容,用中国科学院地球环境研究所气溶胶实验室的 Dionex-600 型离子色谱仪对水溶性离子进行分析.具体分析方法见文献[25].

1.4 其它数据收集

气象数据来源于各城市当地气象站.具体数据包括能见度、风速、风向、温度、相对湿度、大气压和降雨量等气象因子资料.采样期间,气象数据汇总见表 2.

表 2	采样期间的气象统	≥件
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Table 2 Selected meteorological parameters during the sampling period	Table 2	Selected	meteorological	parameters	during t	the sampling	period
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采样点	平均气压 /kPa	日均气温 /℃	相对湿度/%	平均风速 /m·s ⁻¹	降水量/mm
从化	1 00. 40	27. 9	80. 8	1. 31	60. 3
广州	1 00. 40	29. 3	72. 3	1. 37	135. 8
佛山	1 00. 80	29. 8	75. 0	1. 97	118. 9
东莞	1)	28. 5	75. 6	2. 12	34. 7
深圳	_	28. 8	75. 9	1.79	64. 3
珠海	_	28. 8	81. 3	2. 28	79. 9
总平均	1 00. 53	28. 9	76. 8	1. 81	82. 7

1)"一"表示无数据

日均能见度由每天 4 个时次的能见度计算得到. 计算方法是先将每天 4 个时次的能见度换算成大气消光系数求其几何均值,再将消光系数的均值换算成能见度^[3]. 消光系数 b_{ext} 根据公式(1)得到^[26].

$$b_{\rm ext} = 3.912 \times 10^6 / \nu \tag{1}$$

式中, v 为能见度, 消光系数单位为 Mm⁻¹.

1.5 数据分析方法

1.5.1 PM, 质量重建

根据 IMPROVE $PM_{2.5}$ 质量浓度重建公式 $^{[27]}$,利用 $PM_{3.6}$ 的化学组分重建 $PM_{3.5}$ 质量浓度. 具体公式如下:

$$PM_{2.5} = (NH_4)_2SO_4 + NH_4NO_3 + OM + EC + [FS] + [SS]$$
 (2)

式中, $[(NH_4)_2SO_4] = 1.375 \times [SO_4^2]$; $[NH_4NO_3] = 1.29 \times [NO_3^-]$; [OM] (organic matter) $= 1.8 \times [OC]$; [FS] (fine soil) $= 2.2 \times Al + 2.49 \times Si + 1.63 \times Ca + 2.42 \times Fe + 1.94 \times Ti$; [SS] (sea salt) $= 1.8 \times [Cl^-]$. 由于缺少同步测量的气溶胶重金属元素分析数据,细土壤尘成分被归入未测组分当中. 未测组分还包括水分、不可溶金属元素及其它未知成分.

1.5.2 消光系数重建

采用美国 IMPROVE 研究计划得到的颗粒物消光系数(b_{ext})公式,计算珠三角 6 个采样点大气颗粒物的消光系数. 具体 IMPROVE 方程如下:

$$b'_{\text{ext}} = b'_{\text{ap}} + b'_{\text{sp}} \tag{3}$$

$$b'_{ab} = 10 \times [EC] \tag{4}$$

 $b'_{\text{\tiny sp}} = 2.2 \times f_{\text{\tiny S}}(\text{RH}) \times \left[\text{Small}(\text{NH}_4)_2 \text{SO}_4 \right] +$

 $4.8 \times f_{L}(RH) \times [Large(NH_4)_2SO_4] +$

 $2.4 \times f_{\rm S}(\rm RH) \times [\rm Small \, NH_4 NO_3] +$

 $5.1 \times f_{L}(RH) \times [Large NH_4NO_3] +$

 $2.8 \times \lceil \text{Small OM} \rceil + 6.1 \times \lceil \text{Large OM} \rceil +$

 $1.7 \times f_{ss}(RH) \cdot [SS] +$

$$1 \times \lceil FS \rceil + 0.6 \lceil CM \rceil \tag{5}$$

式中,当 [Total X] < 20 μ g·m⁻³, [Large X] = [Total X]²/(20 μ g·m⁻³); 当 [Total X] > 20 μ g·m⁻³, [Large X] = [Total X]; [Small X] = [Total X] - [Large X]. 式中 X分别为(NH₄)₂ SO₄、NH₄NO₃ 和 OM. 考虑到 PM_{2.5}中未测量的化学成分(主要是金属元素和非水溶性物质)也会导致散射消光,其消光效率与 FS 相类似,也有研究在消光重建中考虑这一部分,取得比较理想的效果^[20]. 因此本研究增加这部分(other)对消光系数的贡献,消光效率为1 m²·g⁻¹. 因此原 IMPROVE 消光系数方程(5)进行如下修正:

$$b'_{\rm sp} = 2.2 \times f_{\rm S}({\rm RH}) \times [{\rm Small}({\rm NH_4})_2 {\rm SO_4}] + 4.8 \times f_{\rm L}({\rm RH}) \times [{\rm Large}({\rm NH_4})_2 {\rm SO_4}] + 2.4 \times f_{\rm S}({\rm RH}) \times [{\rm Small}\ {\rm NH_4NO_3}] + 5.1 \times f_{\rm L}({\rm RH}) \times [{\rm Large}\ {\rm NH_4NO_3}] + 2.8 \times [{\rm Small}\ {\rm OM}] + 6.1 \times [{\rm Large}\ {\rm OM}] + 1.7 \times f_{\rm SS}({\rm RH}) \cdot [{\rm SS}] + 1 \times [{\rm FS}] + 0.6 [{\rm CM}] + [{\rm others}] \qquad (6)$$
 式中, $[({\rm NH_4})_2\ {\rm SO_4}]$ 、 $[{\rm NH_4NO_3}]$ 、 $[{\rm OM}]$ 、 $[{\rm SS}]$ 计 算同公式(2). $[{\rm FS}] = [{\rm fine}\ {\rm soil}]$, ${\rm CM}({\rm coarse}\ {\rm mass}) = [{\rm PM}_{10}] - [{\rm PM}_{2.5}]$, $[{\rm others}] = [{\rm PM}_{2.5}] - 1.375$ $[{\rm SO_4^{2-}}] - 1.29 [{\rm NO_3^{-}}] - 1.8 [{\rm OC}] - [{\rm FS}] - [{\rm EC}]$. $b'_{\rm ap}$ 、 $b'_{\rm sp}$ 单位为 ${\rm Mm}^{-1}$,化学组分 $[{\rm SO_4^{2-}}]$ 、 $[{\rm NO_3^{-}}]$ 、 $[{\rm CI}^{-}]$ 、 $[{\rm Ca}^{2+}]$ 、 $[{\rm PM}_{10}]$ 、 $[{\rm PM}_{2.5}]$ 、 $[{\rm OC}]$ 、 $[{\rm EC}]$ 单位为 ${\rm \mu g}$ ·m⁻³. RH为相对湿度, $f_{\rm S}({\rm RH})$ 、 $f_{\rm L}({\rm RH})$ 和 $f_{\rm SS}({\rm RH})$ 为相对湿度增长系数,其取值参见文献 $[{\rm 28}]$.由于粗粒子 CM 对 $b_{\rm sp}$ 的贡献很小 $[{\rm 20}]$,且本研究未同步测量 ${\rm PM}_{10}$ 浓度,因此忽略 CM 对 $b_{\rm sp}$ 的贡献.

2 结果与讨论

2.1 珠三角地区 PM25浓度空间分布特征

夏季珠三角地区 $PM_{2.5}$ 总平均浓度为(36.0 ± 17.5) μ g·m⁻³,小于我国空气质量标准(GB 3095-

2012) 中 $PM_{2.5}$ 的日均值二级标准 75 $\mu g \cdot m^{-3}$ (见表 3). 其浓度水平与我国南部北部湾地区夏季 $PM_{2.5}$ 浓度相当(38.4 ± 17.7) $\mu g \cdot m^{-3}$ [29], 高于沿海城市 青岛(30.1 $\mu g \cdot m^{-3}$) 和厦门(25.2 $\mu g \cdot m^{-3}$) [24], 但显著低于北京(117.2 $\mu g \cdot m^{-3}$)、长春(59.6

 $μg \cdot m^{-3}$)、天津 (103.2 $μg \cdot m^{-3}$)、重庆 (116.3 $μg \cdot m^{-3}$)、杭州 (90.6 $μg \cdot m^{-3}$)、上海 (52.2 $μg \cdot m^{-3}$)、武汉(70.8 $μg \cdot m^{-3}$)等城市^[24]. 总体来讲,珠三角主要城市夏季 $PM_{2.5}$ 浓度相对较低,接近于国家 $PM_{5.5}$ 的日均值一级标准.

表 3 夏季珠三角地区 $PM_{2.5}$ 中主要化学成分浓度 $^{1)}/\mu g \cdot m^{-3}$

Table 3 Cl	hemical component	concentrations i	n PMa.	during summer	in Pearl	River Delta/u	.g • m -3
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		_		_		1.6	
项目	深圳 (n=13)	东莞 (n=15)	广州 (n = 15)	佛山 (n = 15)	从化 (n=15)	珠海 (n=15)	总平均 (n=88)
PM _{2.5}	22. 7 ± 5. 8	46. 5 ± 19. 2	45. 3 ± 12. 0	49. 4 ± 17. 8	30.6 ± 6.2	19.6 ± 9.5	36. 0 ± 17. 5
OC	4.2 ± 1.4	7.9 ± 3.1	8.2 ± 2.3	9.4 ± 2.7	5.9 ± 1.5	3.3 ± 1.8	6. 5 ± 3.1
EC	1.8 \pm 0.9	3.5 ± 1.4	3.3 ± 1.3	3.4 ± 1.2	1. 6 ± 0.5	0.9 ± 0.8	2.4 ± 1.5
F -	0.00 ± 0.00	0.07 ± 0.08	0.0 ± 0.0	0.0 ± 0.0	0.0 ± 0.0	0.01 ± 0.04	0.01 ± 0.04
Cl -	0.1 ± 0.1	0.5 ± 0.8	0.2 ± 0.2	0.4 ± 0.5	0.1 ± 0.2	0.0 ± 0.0	0.2 ± 0.4
NO_3^-	1. 3 \pm 0. 8	4.0 ± 2.5	3.7 ± 1.6	3.6 ± 2.3	1.0 ± 0.4	1.6 ± 0.9	2.6 ± 2.0
SO ₄ -	4. 1 ± 1.7	7.4 ± 3.4	7. 7 ± 2.6	8.8 ± 3.8	6.9 ± 1.5	4.1 ± 2.0	6.6 ± 3.1
nss-SO ₄ -	3.9 ± 1.7	7.2 ± 3.3	7. 5 ± 2.6	8.5 ± 3.7	6. 7 ± 1.4	4.0 ± 1.9	6. 3 ± 3 . 1
Na ⁺	0.7 ± 0.2	0.7 ± 0.2	1. 2 ± 0.4	1.3 ± 0.4	0.9 ± 0.2	0.4 ± 0.2	0.8 ± 0.4
NH ₄ ⁺	0.1 ± 0.1	0.8 ± 1.2	0.9 ± 1.0	1.2 ± 1.3	0.5 ± 0.4	0.0 ± 0.1	0.6 ± 1.0
K ⁺	0. $1 \pm 0. 1$	0.3 ± 0.3	0.3 ± 0.3	0.4 ± 0.2	0.2 ± 0.1	0.0 ± 0.1	0.2 ± 0.2
nss-K +	0.0 ± 0.1	0.3 ± 0.3	0.2 ± 0.1	0.3 ± 0.2	0. 1 \pm 0. 1	0.0 ± 0.1	0.2 ± 0.2
Mg^{2} +	0.0 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0.1 ± 0.0	0. 1 ± 0.1
Ca ^{2 +}	0.2 ± 0.2	0.5 ± 0.3	1.0 ± 0.4	1.0 ± 0.4	0.7 ± 0.2	0.4 ± 0.4	0.6 ± 0.4

1) n 为样品数, $nss-SO_4^{2-} = SO_4^{2-} - 0.252 Na^+$, $nss-K^+ = K^+ - 0.037 Na^+$

从 $PM_{2.5}$ 的空间分布上看,不同城市之间浓度差异较大.海滨城市深圳和珠海的浓度最低(分别为22.7 μ g·m⁻³和19.6 μ g·m⁻³),主要受到南海季风的影响,清洁的海洋气流有利于污染物的稀释,此外较少的工业排放也是导致上述两个城市 $PM_{2.5}$ 浓度较低的主要原因.佛山大气 $PM_{2.5}$ 浓度最高,达到(49.4 ± 17.8) μ g·m⁻³. 其次为东莞和广州城区,其 $PM_{2.5}$ 分别为(46.5 ± 19.2) μ g·m⁻³和(45.3 ± 12.0) μ g·m⁻³,主要与当地高强度的污染物排放有关[30].相对沿海点位珠海和深圳而言,位于珠三角夏季下风向的从化郊区点位 $PM_{2.5}$ 浓度为(30.6 ± 6.2) μ g·m⁻³,略高于深圳和珠海,可能是受到上风向污染物的输送影响.

2.2 主要化学成分空间分布特征

2.2.1 碳组分空间分布特征

观测期间,珠三角地区城市大气 $PM_{2.5}$ 中 OC 和 EC 的总平均浓度分别为(6.5±3.1) μ g·m⁻³和(2.4±1.5) μ g·m⁻³(见表 4),分别占 $PM_{2.5}$ 质量浓度的 18.2%和6.5%. 总碳(TC = OC + EC)占 $PM_{2.5}$ 质量浓度为 33.2%. 与 2002 年和 2006 年珠三角城市夏季 $PM_{2.5}$ 中 OC 和 EC 观测结果对比(见表 4),本研究观测期间 OC 和 EC 浓度较 2002 年观测结果有明显下降,而与 2006 年 OC 观测结果比较接近,EC 有

明显下降. 可见近几年珠三角的大气污染控制措施对碳组分的削减效果比较明显.

与毗邻的北部湾地区^[29]夏季 OC 和 EC 观测结果对比,珠三角 OC 浓度相对较低,而 EC 浓度相对较高.与京津冀、长三角及中部地区的大城市如北京、天津、上海、重庆、武汉等比较(见表 4),珠三角夏季 OC 和 EC 浓度相对较低.总体来看,珠三角地区夏季 PM_{2.5}中 OC 和 EC 浓度处于相对较低的水平.

从 OC 和 EC 空间分布上看,从化、广州、佛山、东莞、深圳、佛山、珠海等地大气 OC、EC 浓度空间分布特征明显(见图 2),其中 OC 浓度顺序为:佛山 > 广州 > 东莞 > 从化 > 深圳 > 珠海,EC 浓度顺序为:东莞 > 佛山 > 广州 > 深圳 > 从化 > 珠海.整体上看,佛山、东莞、广州的 OC 和 EC 浓度明显高于从化、深圳、珠海,其中前 3 个城市大气 OC、EC 平均浓度分别为后 3 个城市的 1.9 和 2.3 倍.

通常 OC 和 EC 之间的相关性关系可以用来判断碳气溶胶的主要来源^[34]. 由图 3 可见,观测期间珠三角地区不同城市 OC 和 EC 的相关性较好(R^2 : 0.66~0.85),可见观测期间珠三角各采样点的 OC 和 EC 有一定的共源性. 其中,东莞的相关系数最高(R=0.92,P<0.001),珠海相关系数相对最低

表 4 国内部分地区和城市 OC、EC 浓度比较/ $\mu g \cdot m^{-3}$

Table 4	Comparisons	of OC a	nd EC of	other areas	and cities	in C	hina/ug•m	- 3

地区	城市/类型	类型	采样时间	OC	EC	文献
珠三角地区	地区总平均	城区	2010 年夏季	6. 5	2. 4	本研究
	地区总平均	混合	2006 年夏季	5. 3	5. 7	[31]
		混合	2006 年冬季	8. 4	6. 5	[31]
	地区总平均	混合	2002 年夏季	9. 2	4. 1	[32]
	地区总平均	混合	2001 年冬季	14. 7	6. 1	[33]
	广州	城区	2004 年夏季	17. 5	5. 7	[22]
	香港	城区	2004 年夏季	11.3	4. 3	[22]
		背景地区	2004 年夏季	5. 6	1. 4	[22]
北部湾地区	地区总平均	城区	2009 年夏季	9. 4	2. 2	[29]
		郊区	2009 年夏季	8.8	1. 4	[29]
长三角地区	上海	城区	2003 年夏季	13. 3	2. 9	[24]
	杭州	城区	2003 年夏季	17. 1	3. 6	[24]
京津冀地区	北京	城区	2003 年夏季	17. 2	4. 6	[24]
	天津	城区	2003 年夏季	16. 5	3. 7	[24]
中部地区	重庆	城区	2003 年夏季	25. 1	8. 0	[24]
	武汉	城区	2003 年夏季	14. 2	3. 0	「24

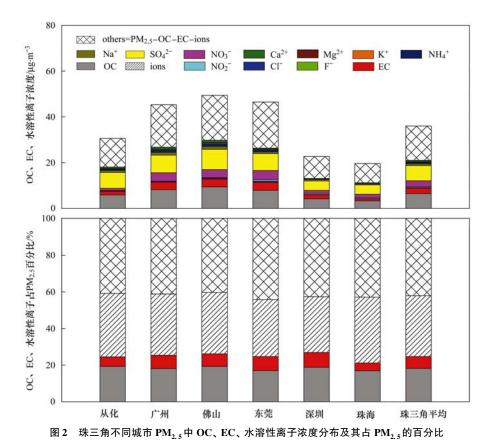


Fig. 2 Distribution of OC, EC and ions concentrations, and its percentage of $PM_{2.5}$ in Pearl River Delta cities

(R=0.81, P<0.001). 6 个采样点 OC 和 EC 回归方程的斜率介于 $1.31\sim2.69$, 其中深圳最低, 从化最高. 根据燃煤、机动车排放和生物质燃烧等不同类型排放源的 OC/EC 平均比值(分别为 2.7、1.1 和 $9.0^{[35]}$), 可以判断珠三角夏季 OC 和 EC 主要来

源是机动车.

特别值得注意的是,珠三角地区 OC/EC 平均值为3.1,变化范围为1.7~7.9,空间分布特征也很明显:珠海(4.6) > 从化(3.8) > 佛山(2.9) > 深圳(2.6) = 广州(2.6) > 东莞(2.3)(见表5). 各个采

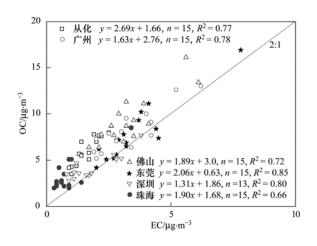


图 3 OC 和 EC 的相关关系

Fig. 3 Correlations between organic carbon and elemental carbon

样点 OC/EC 算术比值明显高于 OC 与 EC 回归斜率,可见夏季珠三角地区二次反应生成的有机碳(SOC)对 OC 的贡献也比较明显.

为进一步了解 SOC 对各观测点 OC 的贡献,利用最小 OC/EC 值方法间接估算珠三角各采样点 SOC 的形成量,其计算方程式为 SOC = OC_{tot} - EC × (OC/EC)_{min},式中,OC_{tot} 为总有机碳,(OC/EC)_{min} 为观测最小比值^[36].估算结果见表 5.从中可知,从化与珠海 SOC 占 OC 比例高达 45%以上,广州、佛山、东莞、深圳 SOC 占 OC 比例在 20% ~ 30% 之间.已有的研究表明,珠三角地区二次有机气溶胶以植被排放源形成为主^[37].从化与珠海相对植被覆盖率较高,主要来源于植被排放的挥发性有机物(VOCs)在高温和高辐射强度的夏季更易于转化成 SOC^[38].

表 5 珠三角地区 OC/EC、SOC、SOC/OC 结果统计 Table 5 Statistics of OC/EC, SOC, SOC/OC

		in Pearl Riv	ver Delta	
位置	样本数	OC/EC	SOC /µg·m ⁻³	SOC/OC /%
从化	15	3.8	2. 8	46. 7
广州	15	2.6	2. 1	26. 1
佛山	15	2.9	2. 1	22. 7
东莞	15	2.3	2. 1	25. 4
深圳	13	2.6	1. 1	27. 7
珠海	15	4.6	1.8	53. 9
总平均	88	3. 1	2. 0	33. 9

2.2.2 水溶性离子空间分布特征

2010 年夏季珠三角地区大气 $PM_{2.5}$ 中总水溶性 离子总平均值为(12.1 ± 6.8) μ g·m⁻³,占 $PM_{2.5}$ 的 平均质量分数为 33.2% ± 5.5% (见图 2),表明水溶性组分是珠三角地区大气细粒子的主要组分之一.

与国内其他城市夏季水溶性离子占 $PM_{2.5}$ 的比例相比,比西安^[28](55%)和北京^[29](46%)低,与上海^[30](32%)相当. 珠三角夏季总水溶性离子浓度空间分布特征为:佛山[(17.0 ± 8.4) μ g·m⁻³] > 广州[(15.3 ± 5.2) μ g·m⁻³] > 东莞[(14.8 ± 7.9) μ g·m⁻³] > 从化[(10.6 ± 2.2) μ g·m⁻³] > 珠海[(7.1 ± 3.7) μ g·m⁻³] > 深圳[(7.0 ± 3.2) μ g·m⁻³].

观测期间珠三角地区各城市大气 $PM_{2.5}$ 中阴离子浓度变化趋势基本一致(见表 3): $SO_4^{2^-} > NO_3^- > NO_2^- > Cl^- > F^-$. 阳离子浓度变化趋势不同城市间存在明显差异,其中从化、广州、深圳为: $Na^+ > Ca^{2^+} > NH_4^+ > K^+ > Mg^{2^+}; 东莞为: NH_4^+ > Na^+ > Ca^{2^+} > K^+ > Mg^{2^+}; 佛山为: Na^+ > NH_4^+ > Ca^{2^+} > K^+ > Mg^{2^+}; 珠海为: Na^+ = Ca^{2^+} > Mg^{2^+} > NH_4^+ = K^+$. 总体上, $SO_4^{2^-}$ 和 NO_3^- 是夏季珠三角地区最主要的两种水溶性离子,占总离子浓度百分比为 (76. 3 ± 7. 2)%.

通常城市地区大气硫酸盐主要来源于大气中 SO, 的氧化[39]. 已有源谱研究表明,珠三角地区 90%以上的 SO,来源于电厂和工业排放[30]. 佛山、 广州、东莞等观测点 nss-SO₄ 浓度较高(分别为 8.5、7.5、7.2 μg·m⁻³),从化、珠海和深圳 nss- SO_4^{2-} 浓度相对较低(分别为 6.7、4.0、3.9 μg·m⁻³),与已有的珠三角 SO,排放清单结果基本 一致[40]. NO; 主要由大气中 NO; 的转化形成, 而 NO, 主要来源于人为源,如机动车排放、工业排放 等. 在珠三角地区,87%的 NO,来源于电厂和机 动车排放[30]. 本次观测期间,东莞(4.0 μg·m⁻³)、广州(3.7 μg·m⁻³)和佛山(3.6 $\mu g \cdot m^{-3}$) 的 NO, 浓度明显高于深圳(1.3 μg·m⁻³)、从化(1.0 μg·m⁻³)和珠海(1.6 μg·m⁻³),表明前3个城市NO_x排放量更大.这一 结果与珠三角地区 NO. 排放清单结果也基本一 致[40]. NH4 主要来源于人类活动排放,其浓度排 序为佛山 > 广州 > 东莞 > 从化 > 深圳 > 珠海. 城 市地区 Ca2+离子常来源于道路交通和建筑工地活 动[41],珠三角地区 Ca2+浓度均排在阳离子浓度的 前3位,可能与亚运前期大规模基建活动有关.

为了了解珠三角地区气溶胶的酸碱性,分别将阴阳离子的质量浓度转换为当量浓度. 具体转换公式如下.

阳离子当量 = Na⁺/23 + NH₄⁺/18 +

$$K^{+}/39 + Mg^{2+}/12 + Ca^{2+}/20$$
 (7)
阴离子当量 = $F^{-}/19 + Cl^{-}/35.5 + NO_{3}^{-}/62 + SO_{4}^{2-}/48$ (8)

由图 4 可见,阴阳离子之间呈极显著的相关关系,反映了本研究所观测的 9 种主要离子成分为 PM_{2.5}的主要离子. 所有样品均位于阴阳离子当量比值 1:1线之上,表明珠三角地区夏季大气 PM_{2.5}均呈酸性^[42]. 回归方程的斜率表明,不同珠三角城市 PM_{2.5}的酸性程度不同,其中深圳和珠海 PM_{2.5}明显偏酸性,下风向从化 PM_{2.5}接近中性.

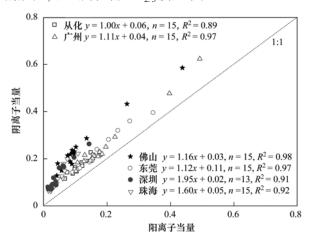


图 4 阴阳离子相关关系

Fig. 4 Total anions versus total cations

2.2.3 PM_{2.5}质量重建

为进一步了解测量的化学成分能否体现 $PM_{2.5}$ 浓度特征,根据 IMPROVE 质量浓度重建公式,利用 $PM_{2.5}$ 的化学组分重建 $PM_{2.5}$ 质量浓度. 由图 5 可见,重建 $PM_{2.5}$ 浓度与实测值之间的 Pearson 相关系数较高,均大于 0.90,斜率范围为 0.70~0.75,约 26% 的质量未能重建,这部分主要包括水分、无机元素以及未知成分. 总体来讲,含碳气溶胶(OM+EC)、硫

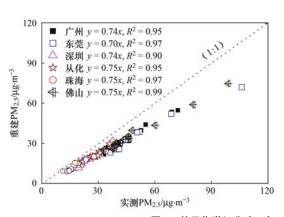
酸盐和硝酸盐是 $PM_{2.5}$ 的主要成分,其所占比例分别为 40.0%、24.7% 和 8.8%.上述主要化学组分能较好地代表 $PM_{2.5}$ 质量浓度.

36 卷

2.3 大气消光系数重建及其主要贡献因子

利用 IMPROVE 方程对珠三角主要城市的大气消光系数进行重建,从化、广州、佛山、东莞、深圳和珠海观测期间实测消光系数与重建消光系数相关性较好(R^2 分别为 0.54、0.62、0.70、0.57、0.52 和 0.76). 除了广州外,其它各点位的大气消光系数重建值均小于实测值(见图 6). 导致上述重建效果相对较差的原因可能是实测消光系数由每日 4 次观测的能见度换算而来,而消光系数重建值 b'_{ext} 是由 24 h PM_2 ,样品化学成分换算而来,因此两者之间必然受到能见度瞬时变化的影响. 总体来讲,重建值 b'_{ext} 基本能反映珠三角大气消光系数的实际变化趋势. 因此 IMPROVE 方程可以用于初步判别观测期间珠三角各城市大气消光系数的贡献因子及其贡献率.

夏季珠三角地区各个点位大气消光系数重建值 b'_{ext} 见表 6. 从中可知,珠三角夏季大气消光系数重建值 b'_{ext} 为 (193 ± 115) Mm^{-1} ,范围为 58 ~ 699 Mm^{-1} . b'_{ext} 最大值出现在东莞,次高值出现在佛山,这与上述两个城市较高的 $PM_{2.5}$ 浓度趋势一致. 尽管珠三角夏季 $PM_{2.5}$ 浓度较低,但重建值 b'_{ext} 还是存在大于 391 Mm^{-1} (对应的能见度为 10 km) 的情况,即还是发生了霾天气. 综合来看,观测期间珠三角大气消光系数最主要的贡献因子是 (NH_4) $_2SO_4$ 和 OM,其贡献率分别达到 39% 和 31%,两者贡献之和高达 70%;其次为 EC 和 NH_4NO_3 ,其贡献率分为 12% 和 13%;海盐及其它成分贡献较小,仅有 5% (见图 6).



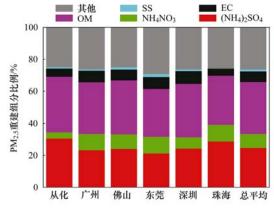


图 5 基于化学组分珠三角 6 个地区 $PM_{2.5}$ 质量浓度重建

Fig. 5 Reconstruction of PM2 5 mass concentrations by chemical compositions in Pearl River Delta

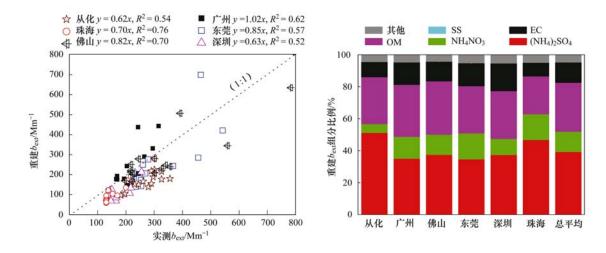


图 6 珠三角夏季重建 b'_{ext} 与 b_{ext} 相关关系、重建消光系数主要贡献因子的贡献率

Fig. 6 Measured versus reconstructed light extinction coefficients and contribution factors in summer in Pearl River Delta

表 6 珠三角地区大气重建消光系数 $b'_{\rm ext}$ 结果统计/ ${
m Mm}^{-1}$

Table 6 Statistics of reconstructed light extinction coefficients in Pearl River Delta/Mm $^{-1}$

位置	样本数	平均值	标准差	最小值	最大值
从化	15	170	39	97	225
广州	15	240	95	143	443
佛山	15	280	129	167	635
东莞	15	249	144	140	699
深圳	13	107	38	68	213
珠海	15	103	58	58	280
总平均	88	193	115	58	699

各观测点重建值 b'ext 的空间分布为:佛山>东莞>广州>从化>深圳>珠海,与其大气 PM2.5 的变化趋势基本一致. 由图 6 可知,在珠三角地区,(NH4)2SO4 是能见度下降最主要的贡献者,其贡献率在 34% ~52%之间;其次是 OM,其贡献率范围为 23% ~34%; EC 和 NH4NO3 贡献率均在 5% ~17%之间,SS 和其它成分贡献之和均小于 6%. 其中,从化和珠海(NH4)2SO4 贡献率均超过 46%,可见在污染较轻的地区,硫酸盐对能见度的下降的贡献率可高达一半以上;珠海的 OM 贡献率相对较低为 24%,广州、佛山、东莞、深圳的 OM 贡献率均超过 29%,可见在大城市地区机动车和 SOC 对能见度的影响不容忽视. 总体来讲,进一步的加强对燃煤和机动车的控制才能有效地缓解大气能见度下降的问题.

3 结论

(1) 2010 年夏季珠三角地区 $PM_{2.5}$ 日均质量浓度为(36.0 ± 17.5) μ g·m⁻³,接近于国家 $PM_{2.5}$ 的日

均值一级标准. $PM_{2.5}$ 中主要化学成分 OC、EC 和总水溶性离子浓度分别为(6.5 ± 3.1)、(2.4 ± 1.5) 和(12.1 ± 6.8) μ g·m⁻³.

(2)珠三角地区 PM_{2.5}中主要化学成分空间分布特征明显,佛山、东莞和广州 PM_{2.5}中主要化学成分(OC、EC 和水溶性离子)明显高于从化、深圳和珠海.

 $(3)(NH_4)_2SO_4$ 是珠三角地区能见度下降最主要的贡献者,其贡献率在 $34\% \sim 52\%$ 之间;其次是 OM,其贡献率范围为 $23\% \sim 34\%$.

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HUANJING KEXUE

Environmental Science (monthly)

Vol. 36 No. 8 Aug. 15, 2015

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(HUANJING KEXUE)

(月刊 1976年8月创刊)

2015年8月15日 第36卷 第8期

ENVIRONMENTAL SCIENCE

(Monthly Started in 1976)

Vol. 36 No. 8 Aug. 15, 2015

	2013	107119 11 30 15 31 0 33			
主	管	中国科学院	Superintended	by	Chinese Academy of Sciences
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		18号,邮政编码:100085)			KEXUE)
		电话:010-62941102,010-62849343			P. O. Box 2871, Beijing 100085, China
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		E-mail; hjkx@ rcees. ac. cn			E-mail; hjkx@ rcees. ac. cn
		http://www.hjkx.ac.cn			http://www.hjkx.ac.en
出	版	44 42 42 KG 24	Published	by	Science Press
_	7400	北京东黄城根北街 16 号			16 Donghuangchenggen North Street,
		邮政编码:100717			Beijing 100717, China
印刷装	专订	北京北林印刷厂	Printed	by	Beijing Bei Lin Printing House
发	行	斜华出版社	Distributed	by	Science Press
		电话:010-64017032			Tel:010-64017032
		E-mail:journal@mail.sciencep.com			E-mail:journal@mail.sciencep.com
订 购	处	全国各地邮电局	Domestic		All Local Post Offices in China
国外总统	发行	中国国际图书贸易总公司	Foreign		China International Book Trading Corporation (Guoji
		(北京 399 信箱)			Shudian), P. O. Box 399, Beijing 100044, China

中国标准刊号: ISSN 0250-3301 CN 11-1895/X

国内邮发代号: 2-821

国内定价:120.00元

国外发行代号: M 205

国内外公开发行