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上海市含碳大气颗粒物的粒径分布

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摘要:使用 STAPLEX 大流量六级采样器(<0.49 μ m、0.49 ~0.95 μ m、0.95 ~1.5 μ m、1.5 ~3.0 μ m、3.0 ~7.2 μ m 和 >7.2 μ m),结合 DRI Model 2001 热光碳分析仪(TOR),分析了 2010 年 5 月 ~2011 年 5 月期间上海市嘉定区(市郊)、徐汇区(市区)的大气颗粒物样品中有机碳(OC)和元素碳(EC)的粒径分布.结果表明,不同粒径的 OC 和 EC 质量浓度均呈双峰分布,较高峰出现于 <0.49 μ m 粒径段,次高峰则出现于 >3.0 μ m 的两个粒径段.嘉定区(JD)和徐汇区(XH)PM3.0 中 OC 的质量浓度分别为 16.35 μ g·m⁻³和 11.85 μ g·m⁻³,EC 质量浓度分别为 2.22 μ g·m⁻³和 1.91 μ g·m⁻³,市郊大气颗粒物中碳组分质量浓度高于市区,说明市郊碳污染更为严重.在 <1.5 μ m 的粒径段,嘉定区 OC 与 EC 的同源性较好,表明其中大部分 OC 来自于燃烧源.两地区不同粒径 OC/EC 值与不同排放源特征值的对比,可以说明徐汇区更多受到机动车尾气排放和道路扬尘的影响.通过 EC 示踪法计算二次有机碳(SOC)质量浓度可知:上海市 SOC 质量浓度较高,PM3.0 中达到 6.76 μ g·m⁻³,占 OC 的质量分数为 69%,粒径分布呈双峰分布,峰值位于 0.49 ~0.95 μ m 和 3.0 ~7.2 μ m 粒径段.

关键词:颗粒物; OC; EC; 粒径分布; 上海

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Size Distribution of Carbonaceous Particulate Matter in Atmosphere of Shanghai, China

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Abstract: Using STAPLEX pumping system with a six-stage cascade impactor (size range; < 0.49 μm, 0.49-0.95 μm, 0.95-1.5 μm, 1.5-3.0 μm, 3.0-7.2 μm, > 7.2 μm), the atmospheric particulate samples at suburb (Jiading District) and urban (Xuhui District) sites in Shanghai, China were collected from May 2010 to May 2011. The organic carbon (OC) and the element carbon (EC) of those samples were analyzed by a DRI Model 2001 thermal/optical carbon analyzer with the reflectance (TOR) method. The size distributions of OC and EC in atmospheric particles both showed bimodal distributions at Jiading and Xuhui, with the peaks at size range of <0.49 μm. Meanwhile, the average annual concentrations of OC and EC in PM_{3.0} were 16.35 μg·m⁻³ and 2.22 μg·m⁻³ at Jiading, 11.85 μg·m⁻³ and 1.91 μg·m⁻³ at Xuhui, respectively. The higher concentrations of particulate matters together with their carbonaceous species at the suburb site indicated that the particulate and carbonaceous aerosol pollution was more serious at suburb than at urban in Shanghai. Compared with Xuhui, the OC shared a higher homology with EC at < 1.5 μm sizes at Jiading, which suggested that OC was mostly derived from combustion sources at urban site in Shanghai. Furthermore, the OC/EC mass ratios at various particulate sizes and those ratios of different sources were discussed. The result suggested that more vehicle emission and more road dust were present at Xuhui District. Moreover, the second organic carbon (SOC) were estimated using EC as a tracer of primary organic carbon, the SOC mass concentration in PM_{3.0} was 6.76 μg·m⁻³ and had a proportion of 69% of OC in Shanghai, which also showed a bimodal distribution with peaks at size of 0.49-0.95 μm and 3.0-7.2 μm, respectively.

Key words: aerosol particle; organic carbon; element carbon; size distribution; Shanghai

大气颗粒物是大气中主要污染成分之一,是导致大气环境能见度降低和影响气候变迁的重要因素^[1],可吸入颗粒物更是危及人类健康的环境"杀手"之一. 碳是大气颗粒物中几种主要富含元素之一,主要以有机碳(organic carbon, OC)和元素碳(element carbon, EC)形式存在. 不同的碳质组分,其物理化学性质存在差异,EC是大气中主要的吸光成分,是大气中能见度降低的重要原因,同时影响大气热量平衡^[2]. OC含有大量的有毒成分,如多环芳

烃(PAHs)^[3]、二噁英^[4]等,严重危害人类健康.不同粒径的颗粒物中的碳组分对人体健康、大气能见度和大气成云降雨的影响也有不同^[5].目前国内外对大气颗粒物中碳的研究主要集中在特定粒径区

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间,如 $PM_{2.5}$ 、 $PM_{10}^{[6,7]}$,并局限在北京、广州等地^[8~10],针对上海市大气颗粒物中的碳污染,特别是碳组分粒径分布特征的研究相对较少.

上海市是中国的经济中心,至 2011 年末,人口已经突破2 300万人,机动车则超过 310 万辆^[11],煤、石油、矿石和水泥的消耗,造成大量的烟尘排放和污染,直接导致雾霾天气的增加,带来严重的大气污染问题.本研究选择上海市嘉定区和徐汇区分别作为上海市郊和市区的典型代表,通过对两地区大气颗粒物中不同粒径的碳组分进行对比分析,得到了上海市大气含碳颗粒物的粒径分布特征,初步判断含碳污染物的来源,以期为大气颗粒物污染的有效治理提供基础资料和科学依据.

1 样品采集与分析

大气颗粒物采样器布置在上海市郊区——嘉定、市区——徐汇区两个采样点. 嘉定采样点 (31.40°N,121.29°E)放置在上海市嘉定区中国科学院上海应用物理研究所 102 楼楼顶,高 18 m,位于上海市中心城区西北约 30 km,宝山工业区以西约 20 km. 嘉定采样点周围无工业厂房、高大建筑,分布有 3 个自然村,地势开阔,植被茂盛,东 300 m处是一条二级公路,主要碳排放源有 3 个木柴小锅炉,一个燃煤锅炉以及生物源排放的有机碳. 徐汇区采样点(31.19°N,121.44°E)位于上海市徐汇区环境检测中心三楼楼顶,距地面 12 m,紧邻漕溪路与南丹路交汇路口. 该地区是典型的商业区代表,人口密度高、车流量大、高层建筑多,采样点可能受到机动车尾气、道路扬尘、商业餐饮油烟等排放源影响. 两地相距约 30 km,地理位置如图 1 所示.

采样仪器选用美国 STAPLEX 公司的 M235 大流量六级采样器,流速 1. 13 $\text{m}^3 \cdot \text{min}^{-1}$,1 ~ 6 级粒径范围依次是: < 0. 49 μm 、0. 49 ~ 0. 95 μm 、0. 95 ~ 1. 5 μm 、1. 5 ~ 3. 0 μm 、3. 0 ~ 7. 2 μm 和 > 7. 2 μm . 为了消除周末效应带来的影响 $^{[12,13]}$,采样时间从周二上午 09:00 开始至周四上午 09:00 结束,连续采样 48 h. 采样前石英膜在 850℃下烘烤 6 h,以消除膜上可能存在的碳. 所有待检测样品置于 -20℃冰柜中保存,采样前后恒温恒湿 48 h,用 Mettler Toledo微量天平(精度 10^{-5} g)称量石英膜,由差值法测定颗粒物质量,采用美国沙漠研究所研制的 DRI Model 2001 热光碳分析仪(TOR 热光反射法)进行样品分析 $^{[14]}$. 样品采集从 2010 年 5 月持续至 2011年 5 月,选取两地区相同采样日期的各 12 套(共 24



图 1 嘉定区与徐汇区采样点示意

Fig. 1 Sampling sites in Jiading and Xuhui

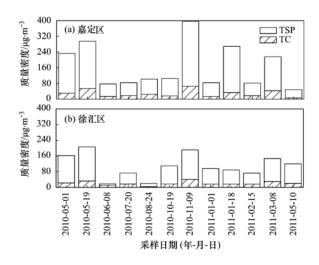
套 144 个)有效样本进行对比分析,另添加嘉定 22 套(共 34 套 204 个)样品对嘉定区进行 OC、EC 相关性和二次有机碳(secondary organic carbon, SOC)的分析.

2 结果与讨论

2.1 大气总悬浮颗粒物质量浓度与碳组分分析

采样期间大气总悬浮颗粒物(total suspend particle, TSP)与碳组分(total carbon, TC)质量浓度分布如图 2 所示,其中碳组分 TC = OC + EC. 嘉定区 TSP 平均质量浓度(137.59 μg·m⁻³)和 TC 平均质量浓度(27.74 μg·m⁻³)较徐汇区(108.86 μg·m⁻³、20.68 μg·m⁻³)偏高,说明嘉定区空气污染比徐汇区更为严重. 而从时间分布来看,两采样点的 TSP 浓度变化趋势较为一致,例如起始日期为2010-05-01、2010-05-19、2010-11-09、2011-03-08的样品,嘉定区和徐汇区的 TSP 质量浓度均比较高,说明两者的大气颗粒物污染随时间变化呈现一致性,意味着上海市市区与市郊的大气颗粒物可能存在着整体性的污染特征. 为进一步分析上海市大气颗粒物中碳组分的污染特征,图 3 给出了颗粒物中 TC 质量浓度和 TSP 中 TC 的粒径分布.

从图 3 中可看出,嘉定区与徐汇区的 TC 粒径分布均为双峰分布,最高峰值均位于 $< 0.49 \mu m$,次高峰值则分别位于 $> 7.2 \mu m$ (JD)和 $3.0 \sim 7.2 \mu m$



图中标注日期为样品采集的起始日期, 从当日上午09:00起,持续48h后结束

图 2 上海市大气总悬浮颗粒物与碳组分质量浓度分布

Fig. 2 Concentration distribution of TSP and TC in Shanghai

(XH)的粒径段. 其中,在 < 3.0 μm 的 4 个粒径段, 嘉定区和徐汇区的 TC 质量浓度均随粒径的增大而 减小,说明在这4个粒径段的颗粒物中总碳含量存 在明显的粒径分布特征,且这种分布与采样点无关. 细颗粒物中的含碳物质主要来自于燃烧生成的一次 碳(EC和POC)和由气态前体物通过气-粒转换生 成的二次碳(SOC).一方面,由燃烧产生的一次碳 主要分布在小粒径颗粒中(本研究中为 < 0.49 μm 的颗粒物),并在大气滞留过程中不断吸湿生长,而 该过程受采样点分布影响小. 另一方面,二次生成 的碳与大气中气态前体物浓度以及生成环境相关, 同样受区域性采样点分布影响较小. 而对于>3.0 μm 的两个粒径段,TC 的质量浓度相差不大,嘉定 区分别为 4.52 µg·m⁻³、4.65 µg·m⁻³,徐汇区分别 为 3. 71 μg·m⁻³、3. 21 μg·m⁻³. 由于两地区采样点 的 > 3.0 µm 颗粒物的来源存在明显的差异:嘉定区 采样点周边绿化率高,粗颗粒中可能更多地来自于 植物碎片和花粉等自然源: 徐汇区采样点周边为道 路和高楼,粗颗粒中可能更多受到道路扬尘和机动 车刹车磨损以及轮胎摩擦等人为来源的影响. 来源 上的差异决定了两采样点 TC 浓度在这两个粒径段 上的分布差异,嘉定区 > 7.2 μm 颗粒物中 TC 的浓 度大于 3.0 ~ 7.2 μm 颗粒物中 TC 的浓度,而徐汇 区则相反. 这种差异可能是花粉颗粒和植物碎片等 产生的影响,因为花粉和植物碎片的粒径一般处于 >7.2 µm 的粒径段.

对于碳组分占颗粒物的质量分数,从图 3 可看出,其百分数随着粒径的增大而有减小的趋势,在

<0.49 μm和 0.49 ~ 0.95 μm 粒径段约为 25%,而在 > 7.2 μm 粒径段,则降低到 15% 左右.可见,含碳物质更容易在细颗粒物中富集,这与唐小玲等[15]对广州大气颗粒物粒径分析的研究结果是一致的.另外,在细粒径颗粒物中,碳所占质量分数随粒径增大而减小,说明细颗粒在增长过程中,含碳物质的增长并不是同步的.

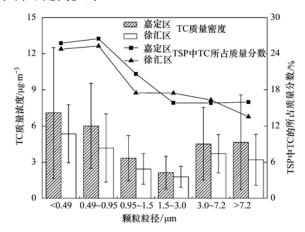


图 3 TC 质量浓度及 TC/TSP 的粒径分布

Fig. 3 Size distribution of TC concentrations and the percentages of TC in TSP

2.2 OC、EC 粒径分布

上海市嘉定区与徐汇区大气颗粒物中 OC、EC 质量浓度的粒径分布如图 4 所示,两地区不同粒径 OC、EC 的质量浓度均呈双峰分布,但 OC 的分布特征与 EC 存在着明显的差异. OC 质量浓度较高峰出现于 <0.49 μm 粒径段,次高峰则出现于 >3.0 μm 的两个粒径段:在 <0.49 μm 粒径段,嘉定区为6.00 μg·m⁻³,徐汇区为4.44 μg·m⁻³,均占 OC 总量

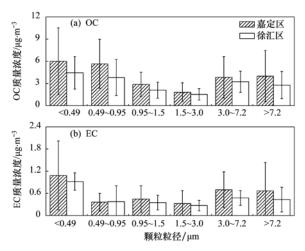


图 4 上海市大气颗粒物 OC、EC 质量浓度粒径分布

Fig. 4 Mass concentration for size distribution of OC and EC in Shanghai aerosol particles

的 25%;而在 > 3.0 μ m 的两个粒径段,嘉定区为 7.81 μ g·m⁻³,徐汇区为 6.00 μ g·m⁻³,分别占 OC 总量的 32% (JD) 和 34% (XH). 而在 < 0.49 μ m 粒径区间, EC 占 EC 总量的 30% (JD) 和 33% (XH), 在 > 3.0 μ m 的两个粒径段,分别占 EC 总量的 38% (JD) 和 32% (XH). 相对于 OC 而言,EC 更多地集中在 < 0.49 μ m 的颗粒物中,这可能是因为汽车尾气、煤炭燃烧等 EC 的主要排放源,其产生的一次颗粒粒径集中在 < 0.2 μ m 的粒径范围内^[16].

两地区 PM_{3.0} 中 OC 质量浓度分别为 16.35 μg·m⁻³(JD)、11.85 μg·m⁻³(XH),EC 质量浓度分 别为 2. 22 μg·m⁻³(JD)、1. 91 μg·m⁻³(XH),嘉定 区碳组分质量浓度偏高,在各粒径区间内,嘉定区的 OC、EC 质量浓度分别是徐汇区的 1.2~1.5、1~ 1.5倍,充分说明市郊空气污染比市区更为严重,在 王杨君等[17]对上海市大气颗粒物观测也呈现出市 郊大气颗粒物污染明显高于市区的特点. 但在国内 其他城市的研究中,如陈刚才等[18]对重庆观音桥 (市区)和缙云山(市郊)和迟旭光等[19]对北京天坛 公园(市区)和十三陵(市郊)监测时发现,市区 OC 和 EC 的质量浓度远远高于市郊. 存在这样的差异, 与郊区采样点的大气环境有密切关系,但值得注意 的是,上海市从20世纪90年代开始进行城市空间 布局调整,将工业生产主体从中心城区向周边地区 迁移,对改善市区空气质量起到明显作用,通过王杨 君等[17]2007~2008年对上海市大气颗粒物采样结 果与往年对比发现,中心城区的空气质量已经有了 比较大的改善,不过这也同时导致市郊的空气质量 的恶化.

从城市大气环境监测的角度来说,由于不同城市功能区之间的污染水平存在差异,郊区的污染水平可能更高,需要设置更多的郊区监测点来保证对整个上海市大气颗粒物污染水平进行综合全面的评价. 从环境保护治理措施来讲,上海市的工业布局决定了未来上海市的大气污染呈现为中心城区污染低,周围郊区污染相对严重的特点,但是由于大气扩散与运动,中心城区也不可避免受到周围污染严重地区的影响. 因此,严格控制郊区一次污染源的排放,才能够更有效地改善上海市大气环境状况.

2.3 OC、EC 的相关性分析

大气颗粒物中的 EC 仅来自于燃烧源,可以作为一次有机碳(primary organic carbon, POC)的示踪物.而 OC 中不仅含有与 EC 一起产生的 POC,还有经大气光化学反应生成的二次有机碳(secondary

organic carbon, SOC),以及其他生物颗粒,如孢子、细菌、病毒等,除 POC 以外的其他 OC 成分会使 OC 与 EC 的相关性变差. Turpin 等 $^{[20]}$ 认为, OC 和 EC 的关系可以被用来区分碳颗粒物的来源,若 OC 与 EC 的相关性好,表明 OC 和 EC 来自相同污染源. 因此利用 OC 和 EC 的相关性可在一定程度上对大气碳颗粒物的来源进行定性分析. 由于徐汇区采样数目较少,本研究仅对嘉定区 34 套(共计 204 个)样品的不同粒径区间按照公式 ρ (OC) = $a+b \times \rho$ (EC)进行回归计算 $^{[8]}$. 其中截距 a 代表非燃烧源的有机碳部分,如生物颗粒等,斜率 b 表示燃烧源排放的有机碳与元素碳的比值, R^2 为相关系数,结果如图 5 所示.

在 < 0. 49 μ m、0. 49 ~ 0. 95 μ m 和 0. 95 ~ 1. 5 μ m 粒径段, R^2 分别为 0. 79、0. 82 和 0. 69,相关性较好, 说明 OC 与 EC 的同源性较好. 在 < 0. 49 μ m 和 0. 95 ~ 1. 5 μ m 粒径段 a 值分别为 - 0. 32 和 0. 11,接近于 0,说明 OC 主要来自燃烧源;对于 0. 49 ~ 0. 95 μ m 粒径段, OC、EC 相关性好但 a 值较大,说明该粒径区间内 OC 来源比较稳定,但有部分 OC 来自非燃烧源,有研究表明,在 0. 7 ~ 0. 8 μ m 粒径段,小颗粒在生长过程中吸附了大量的二次反应产物 [21]. 而在 1. 5 ~ 3.0 μ m、3.0 ~ 7.2 μ m 和 > 7.2 μ m 粒径段, R^2 分别为 0. 06、0. 05 和 0. 11, OC 与 EC 的相关性较差,a 值也较大,表明大颗粒中的 OC 来源较为复杂,有更多来自非燃烧源的有机碳 [15],嘉定区粗颗粒中的生物气溶胶如花粉、孢子、细菌、病毒等会对实验结果造成较大影响.

Michael 等[21] 对美国加州洛杉矶市市区的大气 颗粒物观测中认为,市区污染源主要受到机动车尾 气、道路扬尘、烹饪和燃煤等因素的影响,并分析 各排放源的粒径分布特征,峰值在 0.2~0.3 µm 粒 径段的颗粒物主要源自机动车尾气、烹饪和燃煤, 道路扬尘的影响集中在 > 2.5 μm 的粒径段. Chow 等[22]研究表明不同的污染源所排放颗粒物的 OC/EC值存在明显差异,如在 PM。中柴油和汽油车 尾气排放的 OC/EC 值为 1.0~4.2, 燃煤排放为 2.5 ~10.5[23],生物质燃烧排放为16.8~40.0[24],烹调 排放为 32.9~81.6^[25],在 2.5~10 μm 粒径段道路 扬尘的 OC/EC 值为 8.8^[22]. 在 < 1.5 μm 的粒径区 间内 OC 与 EC 的同源性较好,本研究试用 OC/EC 值来推测大气颗粒物中碳成分的来源,图6列出了 上述碳排放源的 OC/EC 特征值与本次测量值的对 应关系.

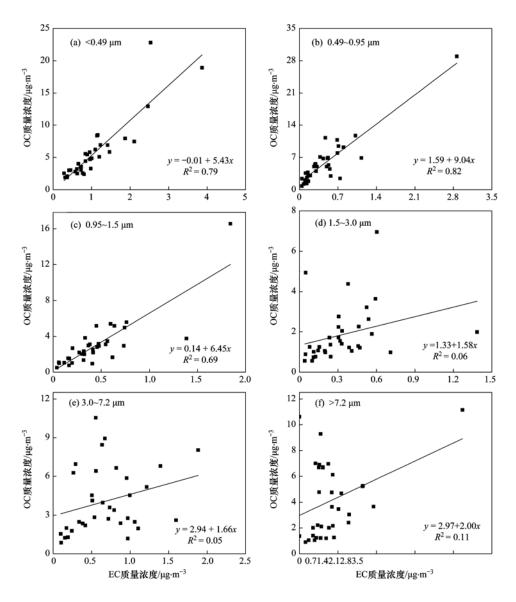


图 5 嘉定区不同粒径 OC、EC 的相关性分析

Fig. 5 Correlation of OC and EC with different diameters in Jiading

通过各粒径区间 OC/EC 值与排放源 OC/EC 特征值比较可以发现:在 < 0. 49 μ m 和 0. 95 ~ 1. 5 μ m 粒径区间内,两地主要受到燃煤和机动车尾气的影响;在 0. 49 ~ 0. 95 μ m 粒径段,两地 OC/EC 值偏高,这可能是受到机动车尾气、生物质燃烧等多方面因素的综合影响,前文进行 OC、EC 相关性分析时,也认为此粒径段除燃烧源产生的 OC 外,还含有部分稳定的非燃烧源产生的 OC. 在 < 3. 0 μ m 粒径段,徐汇区 OC/EC 值分别为 4. 6 (< 0. 49 μ m)、13. 9 (0. 49 ~ 0. 95 μ m)、6. 3 (0. 95 ~ 1. 5 μ m)、6. 1 (1. 5 ~ 3. 0 μ m),均小于嘉定区对应粒径的 OC/EC 值(5. 8、16. 8、8. 3、7. 6),可以说明该地区受到机动车尾气的影响更加明显。在 3. 0 ~ 7. 2 μ m 和 > 7. 2 μ m 粒径段,徐汇区 OC/EC 值分别为 7. 0、6. 9

高于嘉定区 4.5、5.3,表明徐汇区更多受到道路扬尘的影响,这与采样点周围的环境一致. 另外,本实验采用光热法^[26]进行碳组分分析时,大气颗粒物中土壤来源的碳酸盐^[27,28]在高温下分解生成 CO₂,会造成 EC 测量值偏大,从而使 OC/EC 值变小,而嘉定区位于市郊,大气中有更多土壤来源的颗粒物,这也会给实验结果造成一定影响.

2.4 二次有机碳(SOC)分析

大气颗粒物中的 SOC 来自于燃烧源和植物挥发而产生的气态有机物,如苯并芘、异戊二烯、萜烯等,被大气中的 O₃、OH 自由基等氧化,经过气-粒转化效应而进入颗粒相^[29]. EC 在大气中具有良好的稳定性,在大气中滞留时间较长,不易发生化学转化,所以 EC 可以作为一次排放的示踪物.由于一次

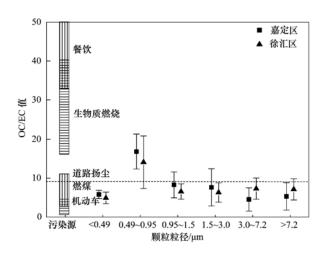


图 6 嘉定区与徐汇区不同粒径 OC/EC 值

Fig. 6 Values of OC/EC with different diameters in Jiading and Xuhui

排放源的 OC 与 EC 存在一个相对稳定的比值,一般 认为当大气颗粒物中 OC/EC 值高于某个临界值 (通常取 2. 0)时,大气颗粒物中含有 SOC^[30],采用 EC 示踪法对 SOC 进行估算^[31]:

$$\rho(\text{SOC}) = \rho(\text{OC}) - \rho(\text{EC}) \times \left[\frac{\text{OC}}{\text{EC}}\right]_{\text{min}}$$
 (1)

式中, $\rho(SOC)$ 为二次有机碳质量浓度; $\rho(OC)$ 和 $\rho(EC)$ 分别代表样品中有机碳和元素碳实测浓度; [OC/EC] min 表示测定周期中 OC/EC 的最小值. 由 于样本数过少造成统计信息误差偏大,导致采用的 [OC/EC]_{min}有可能高于[OC/EC]_{primary},使计算结果 SOC 比实际值偏小,本研究只针对嘉定区 34 套(共 计 204 个) 样品进行 SOC 粒径分析. 在[OC/EC] min 取值上,Turpin等[32]对洛杉矶空气样品进行分析发 现,特定天气(低温、阴雨、臭氧浓度低以及气团不 稳定)的大气光化学活性低,不利于 SOC 的生成, OC/EC 值较低, 因此可以将一定时期内的[OC/ EC] min 近似等同于[OC/EC] primary. 但是 OC 中含有 大量的水溶性有机物,降雨会导致测得的 OC/EC 值 偏小,取值应当首先排除降雨条件下的测定 值[33,34],本研究中[OC/EC]min的选取也参考了采样 期间的气象条件,从各个粒径区间内,选取晴朗天气 下各个粒径的最小 OC/EC 值作为[OC/EC]min,将 上述取值代入公式(1),得到结果如图7所示.

图 7 表示上海市嘉定区大气颗粒物中 SOC 质量浓度以及 OC 中 SOC 所占质量分数的粒径分布情况,其中,SOC 粒径分布呈现双峰分布,峰值位于 $0.49 \sim 0.95~\mu m$ 和 $3.0 \sim 7.2~\mu m$ 粒径段,质量浓度达到 $2.19~\mu g \cdot m^{-3}$ 和 $3.26~\mu g \cdot m^{-3}$,各粒径区间的

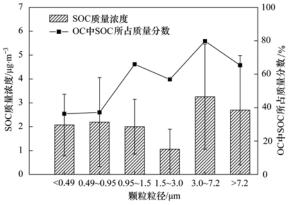


图 7 嘉定区 SOC 质量浓度及 SOC/OC 值的粒径分布

Fig. 7 Size distribution of SOC mass concentration and the percentage of SOC in OC in Jiading

SOC/OC值最高为 80% (3.0 ~ 7.2 μm),最低值为 36% (<0.49 μm),颗粒物中平均值为 54%. 在唐小玲等[15]对广州市大气颗粒物的研究中也得到类似结论: SOC 峰值出现在 0.49 ~ 0.95 μm (3.87 μg·m⁻³)和 3.0 ~ 7.2 μm (3.10 μg·m⁻³)粒径区间, 3.0 ~ 7.2 μm 粒径上的 SOC/OC 值最高 (74%), <0.49 μm 粒径上的值最低 (16%),平均值为 47%. 有研究认为,由二次光化学反应直接生成的颗粒浓度峰值集中在 0.2 μm^[35],而本次测量中 SOC 的粒径分布并没有表现出这样的特点. 这可能是因为一方面大颗粒物多来自于土壤扬尘,其中碳酸盐等碱性物质吸收硫化物,氮化物等酸性气体,生成盐类和水分,更容易吸收可溶性有机物^[36];另一方面,大颗粒较大的体积为气-粒转化提供了大的反应表面,更有利于 SOC 向颗粒态转化^[36].

Cao 等^[37]研究珠三角地区冬夏季 PM_{2.5}中 SOC 质量浓度均值为 4.9 μg·m⁻³,占 OC 质量分数为 51.3%.本次观测得到上海地区大气 PM_{3.0}中 SOC 的年平均质量浓度为 6.76 μg·m⁻³,占 OC 质量分数 为 69%.这表明上海可能比广州地区提供了更有利于 SOC 形成的大气环境与气候条件,如较高浓度的气态前体物、充足的阳光照时间、较低的温度等^[38].上海市的年平均气温为 15.6℃,光照时间为 1 874 h;而广州的年平均温度为 22.6℃,光照时间为 1 628 h.较低的气温有利于气态向颗粒态的转变,充足的阳光则保证大气中 OH 自由基、O₃等氧化剂的浓度,使得气态活性气体更容易被氧化为半挥发性产物附着在大气颗粒上^[39].

3 结论

(1)2010年5月~2011年5月采样期间,嘉定

区(市郊)和徐汇区(市区) $PM_{3.0}$ 中 OC 质量浓度分别为 16.35 $\mu g \cdot m^{-3}$ (JD)、11.85 $\mu g \cdot m^{-3}$ (XH),EC 质量浓度分别为 2.22 $\mu g \cdot m^{-3}$ (JD)、1.91 $\mu g \cdot m^{-3}$ (XH),市郊碳组分质量浓度明显偏高,说明由于上海市工业布局调整的影响,市郊大气中碳污染比市区更为严重.

- (2)上海市大气颗粒物中碳组分主要集中在 <3.0 μm 粒径段,其中 EC 更多集中在 <0.49 μm 粒径段,占全部 EC 的 30% 以上. OC 与 EC 的相关性分析表明,嘉定区在 <1.5 μm 的粒径段,OC 与 EC 的同源性较好,细颗粒中大部分 OC 来自于燃烧源,在 0.49 ~0.95 μm 粒径段存在部分稳定的非燃烧源,而在粗颗粒(>3.0 μm)中存在较多的非燃烧源的有机碳.
- (3)在 < 3.0 μm 的粒径区间内,徐汇区OC/EC 值分别为 4.6 (< 0.49 μm)、13.9 (0.49 ~ 0.95 μm)、6.3 (0.95 ~ 1.5 μm)、6.1 (1.5 ~ 3.0 μm),均小于嘉定区对应粒径的 OC/EC 值(5.8、16.8、8.3、7.6),在 3.0 ~ 7.2 μm 和 > 7.2 μm 粒径段,徐汇区 OC/EC 值(7.0、6.9)高于嘉定区(4.5、5.3). 通过不同粒径 OC/EC 值与不同排放源特征值的对比,可以认为在 < 3.0 μm 粒径区间内,徐汇区更多受到机动车尾气的影响,而在 > 3.0 μm 粒径区间内,徐汇区更多
- (4) 通过 EC 示踪法计算嘉定区 SOC 含量, $PM_{3.0}$ 中 SOC 的年平均质量浓度为 6.76 $\mu g \cdot m^{-3}$, 占 OC 的 69%, 不同粒径 SOC 质量浓度呈现双峰分布, 峰值位于 0.49 ~ 0.95 $\mu m (2.19 \ \mu g \cdot m^{-3})$ 和 3.0 ~ 7.2 $\mu m (3.26 \ \mu g \cdot m^{-3})$ 粒径段. 与珠三角地区相比,上海市 SOC 浓度较高,与该地区较高浓度的气态前体物,充足的阳光,较低的温度有关.

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