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《环境科学》征订启事(4061) 《环境科学》征稿简则(4132) 信息(4233, 4293, 4304)

长三角地区气溶胶光学性质与新粒子生成观测

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摘要: 2019年5月27日~6月27日对江苏省常州市的气溶胶光学性质参数、颗粒物数浓度和PM_{2.5}组分进行观测, 联用扫描电迁移率粒径谱仪(SMPS)、黑碳仪(AE33)、腔衰减相移式单次反照率监测仪(CAPS)、在线离子色谱分析仪(MARGA)和RT-4型有机碳/元素碳(OC/EC)分析仪分析: ①新粒子生成期间化学组分与光学参数的变化; ②IMPROVE、MIE理论重建消光系数与实测值的闭合性对比。观测期间共有两次明显的新粒子生成事件, 粒子粒径从4 nm持续增长到64 nm, 在新粒子生成初期硫酸盐贡献较大, 生成过程中实测平均消光系数为95.40 Mm⁻¹, IMPROVE模型重建平均消光系数为140.20 Mm⁻¹, MIE理论模型计算平均消光系数为93.54 Mm⁻¹, 低于我国城市气溶胶消光系数均值300 Mm⁻¹。本次观测采用多仪器联用的方式从颗粒物数浓度粒径谱、化学组分谱等不同的方面更好地对气溶胶理化性质进行表征。

关键词: 新粒子生成; 化学组分; 光学参数; MIE理论; IMPROVE方法

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Observation of Aerosol Optical Properties and New Particle Formation in the Yangtze River Delta

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Abstract: In a recent field campaign focused on air quality study, aerosol optical properties, particle number concentration, and PM_{2.5} components were monitored in Changzhou, Jiangsu Province, from May 27 to June 27, 2019. An array of instruments were deployed that included scanning mobility particle size spectrometer (SMPS), aethalometer (AE33), cavity attenuation phase shift single albedo monitor (CAPS-ALB), monitor for aerosols and gases in ambient air (MARGA) and RT-4 organic carbon/elemental carbon (OC/EC) carbon analyzer to study the ① changes in chemical composition and optical parameters of the new particles generated during the campaign period. ② comparison of the aerosol extinction coefficient recorded by these instruments and measured value in the reconstruction of IMPROVE (interagency monitoring of protected visual environment) and the calculated coefficient using MIE theory model were carried out. During the entire campaign, two new particle generation events were observed and also found that the particle size continued to increase from 4 nm to 64 nm. It was monitored that in the initial stage of new particle generation, sulfate contributed greatly. The measured average aerosol extinction coefficient during the period of particle generation, using these instruments was 95.40 Mm⁻¹, while the average aerosol extinction reconstruction using the IMPROVE model was observed to be 140.20 Mm⁻¹. The theoretical calculations based on Mie theory model yielded an average extinction coefficient of 93.54 Mm⁻¹. It was found that the average aerosol extinction in Changzhou is lower than the average value of the urban aerosol extinction coefficient, which is measured to be 300 Mm⁻¹ in China, during this period. The deployment of multiple instruments in a single campaign is more desirable because the combination of all observations helped in better characterization of the physicochemical properties of ambient aerosols from various aspects, including particle size spectrum and chemical composition.

Key words: new particle generation; chemical constituent; optical coefficient; MIE theory; IMPROVE method

气溶胶光学性质在很大程度上取决于气溶胶的粒径分布、化学成分、混合状态和湿度^[1-3]。目前, 气溶胶光学性质研究上主要有: ①基于颗粒物数浓度粒径谱观测采用 MIE 理论模型计算全散射(消光)

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系数^[4]; ②基于气溶胶的化学组分特征, 采用 IMPROVE (interagency monitoring of protected visual environment) 方法重建消光系数^[5]. 将采用光学仪器(光腔^[6~8]、开放光程^[9]和积分浊度计^[10])进行实际测量的气溶胶光学性质参数与颗粒物粒径谱(MIE 模型)和化学组分谱(IMPROVE 方法)计算得到的理论值进行比对分析, 成为目前较为通用的气溶胶光学闭合实验研究方法^[11,12].

基于 MIE 理论模型的气溶胶数浓度粒径分布谱计算消光系数, 其中气溶胶粒径观测尤为重要. 上世纪末, 国外学者就开始了针对不同环境下气溶胶粒径数谱进行观测, 包括城市^[13]、森林^[14]、乡村^[15]、极地^[16]和沿海^[17,18]等典型区域, 在观测过程中新粒子这一概念被提出, 即核膜态气溶胶(3~20 nm)的数目浓度急剧增加并持续增长, 在粒径谱图中呈香蕉状特征^[19]. 新粒子经由大气物理和化学过程进一步长大至艾根模态, 可影响空气质量、人群健康、直接或间接影响天气和气候^[20]. 我国的气溶胶数浓度粒径谱观测多集中于华北(主要为北京)^[21,22]、西北^[23]和珠江三角洲^[24~26], 在长江三角洲地区, 气溶胶粒径观测集中于南京^[19,27]和上海^[28]等地, 主要侧重研究新粒子生成机制、成核速率和成核方式等. 刘新民等^[29]利用 MIE 理论模型重建北京市大气气溶胶散射系数, 与实际测量值吻合较好; 谢银海^[30]利用 MIE 激光雷达反演分析了 2012 年沙尘天和 2013 年的雾-霾天的消光系数, 与 CALIPSO 卫星反演结果吻合; 刘斌等^[31]于 2019 年对北京地区气溶胶利用总量双峰体积谱和化学组分体积谱结合 MIE 理论方法进行重建消光系数, 得到的闭合相关系数为 0.9686 和 0.9734. 因此普遍认为 MIE 理论消光可以代表实测消光.

美国 IMPROVE 检测网络利用气溶胶化学组分质量浓度及其消光效率重构消光系数, 最早于 1994

年被提出^[32], Pitchford 等^[33]于 2007 年对原有 IMPROVE 方法进行修订. 目前国内现有研究表明, 上海(0.83)^[34]、北京(0.79)^[35]、广州(0.99; 0.97)^[5]、南京(0.89)^[36]、厦门(0.91)^[3]、黄山(0.84)^[37]和杭州(0.81)^[38]等地区利用 IMPROVE 方法对气溶胶化学组分重建消光系数, 与实测值也有较好的相关闭合性, 硝酸盐、硫酸盐和有机物是消光的主要贡献来源, 且随位置和季节变化. 而长江三角洲的观测主要集中在南京和上海等人口较为密集的城区, 关于新粒子生成期间郊区化学组分变化引起的消光参数变化报道尚不多见. 常州位于长江三角洲中心地带, 气溶胶研究主要基于碳质气溶胶、颗粒物来源解析和 PM_{2.5} 组分解析, 对于气溶胶光学性质的研究偏少.

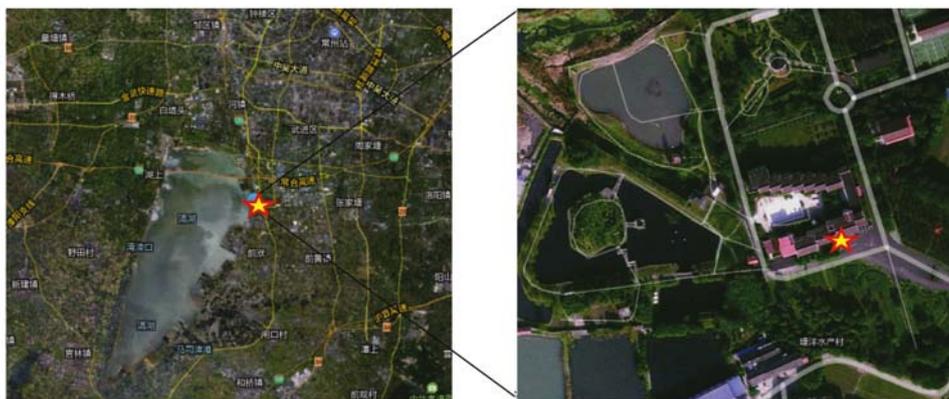
本研究在常州郊区进行为期一个月的综合观测, 以光学消光性质闭合研究为基点, 采用颗粒物 MIE 散射模型和 IMPROVE 方法对观测期间新粒子生成过程中光学性质以及 PM_{2.5} 化学组分特征进行分析, 探讨长三角背景地区新粒子生成期间气溶胶消光性质的粒径谱和组分谱两种闭合方法研究.

1 材料与方法

1.1 观测地点与时间

本次观测站点位于江苏省常州市溧湖地区(31°48'N, 119°58'E). 如图 1 所示, 时间为 2019 年梅雨季前的 5 月 27 日~6 月 27 日. 常州市地处长三角中心地带, 属于亚热带季风气候. 溧湖位于常州市西南方向, 周边 60% 是生态湿地、绿化园林, 20% 是水域. 周边无明显污染源排放及高层建筑群, 能够较好地代表长三角梅雨季前的区域大气状况.

采样地点位于疗养院南侧 4 楼房间. 楼高约 12 m, 采样管高约 15 m, 所有采样管均连接 2.5 μm 切割头.



五角星位置为采样点

图 1 观测点位置示意

Fig. 1 Location of monitoring site

1.2 观测仪器与参数

1.2.1 颗粒物数浓度粒径谱分布

颗粒物数浓度监测主要用两台扫描电迁移率粒径谱仪(厂商:美国 TSI, 型号分别为 SMPS3080 和 SMPS3082), 两台仪器分别使用 NANO DMA (3085) 和 LONG DMA (3081); 粒子计数器均为醇类 CPC (型号分别为 3776 和 3750). 气溶胶样品先通过放射性中和器, 然后进入 DMA. DMA 通过改变电压来筛选不同粒径的颗粒物, 被选出的颗粒物进入 CPC 吸湿长大后被计数.

NANO DMA 筛选的粒径范围为 2.02 ~ 63.8 nm, 样气流量为 1.5 L·min⁻¹, 鞘气流量为 15 L·min⁻¹; LONG DMA 筛选的粒径范围为 14.1 ~ 736.5 nm, 样气流量为 0.3 L·min⁻¹, 鞘气流量为 3 L·min⁻¹, 两台仪器使用一根三通分离的进样管, 两台仪器有部分颗粒物粒径段重合 (14.1 ~ 63.8 nm), 2019 年 6 月 3 ~ 16 日期间由于断电原因造成部分数据缺失. 图 2 所示为 LONG DMA 经过 Particle Loss Calculator 校正后与 NANO DMA 各个粒径下的平均数浓度的正态分布曲线, 两者结合可以代表大气颗粒物的分布状况.

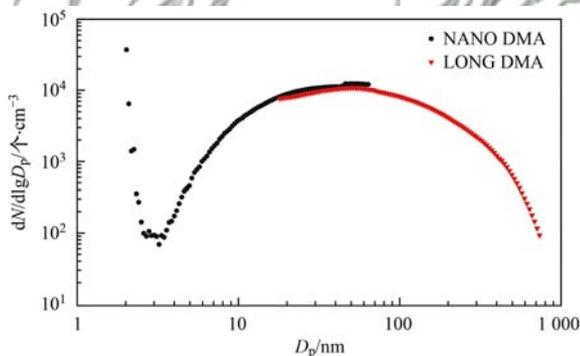


图 2 颗粒物数浓度粒径分布谱图

Fig. 2 Particle number concentration and particle size distribution spectrum

1.2.2 气溶胶化学组分

气溶胶水溶性离子组分用在线离子色谱仪 (MARGA ADI 2080) 分析, 时间分辨率为 1 h. MARGA 采用水相分离的方法, 样气以 1 m³·h⁻¹ 的速度进入采样箱, 在采样箱内通过旋转式液膜气蚀器对可溶气体经过双氧水定量吸收, 随后被蒸汽气溶胶喷射器捕获, 脱气后的样品与溴化锂混合, 再由两套离子色谱分析系统同时对阴离子和阳离子进行检测^[39].

有机碳/元素碳 (OC/EC) 使用美国 Sunset Lab 研制和生产的 1 h 分辨率的 RT-4 型有机碳/元素碳 (OC/EC) 分析仪分析定量. 样气进入分析仪沉积在石英膜上并在氦气 (He) 的非氧化环境中逐级升温,

致使 OC 被加热挥发 (该过程中也有部分 OC 被碳化); 此后样品又在氦气/氧气混合气 (He/O₂) 环境中逐级升温, 该过程中 EC 被氧化分解为气态氧化物. 这两个步骤中所产生的分解产物都随着通过分析室的载气 (同时也是环境气及反应气, 亦即 He 或 He/O₂) 经过二氧化锰 (MnO₂) 氧化炉被转化为 CO₂ 后由离子火焰法 (FID) 或是由非红外色散法 (NDIR) 定量检验^[40].

MARGA 和 OC/EC 分析仪由于仪器故障, 2019 年 6 月 14 日之后停测, 数据缺失.

1.2.3 光学性质参数

光学性质参数用腔衰减相移式单次反照率监测仪 (cavity attenuated phase shift-albedo monitor, CAPS-ALB) 和七波段 (370、470、520、660、880、950 nm) 黑碳仪 (aethelometer, AE-33) 测得.

CAPS-ALB 由美国 Aerodyne 公司生产, 可监测颗粒物的消光系数 b_{ext} , 进样流量为 0.85 L·min⁻¹. 主要是通过检测由光源调频引起的散射积分信号来测量消光系数, 利用真空光电二极管检测光源方形波调频外差检波相移量测量总消光系数^[41].

AE-33 由美国 Magee Scientific 公司生产, 可根据光透过沉积黑碳的纸带时的衰减量得到黑碳浓度信息, 进样流量为 5 L·min⁻¹. 通过式 (1) 可进一步得到气溶胶吸收系数:

$$b_{\text{abs}} = \text{BC} \times \sigma_{\text{air}} \quad (1)$$

式中, BC 为黑碳浓度 ($\mu\text{g} \cdot \text{m}^{-3}$), b_{abs} 为气溶胶吸收系数 (Mm^{-1}), 根据 AE-33 手册 (2018 年 10 月版) 提供的黑碳仪第三通道 (520 nm) 气溶胶吸收截面 $\sigma_{\text{air}} = 13.14 \text{ m}^2 \cdot \text{g}^{-1}$, 此外 2019 年 6 月 16 ~ 17 日由于更换纸带造成部分数据缺失.

由于两台仪器的测量参数不同, 可以得到观测期间气溶胶的散射系数 b_{sca} ($b_{\text{sca}} = b_{\text{ext}} - b_{\text{abs}}$). 在气溶胶光学性质的表征中, 单次散射反照率 (single scattering albedo, SSA) ω 在辐射传输中起着非常重要的作用, 它是指气溶胶散射系数与消光系数的比值 [式 (2)]:

$$\omega = \frac{b_{\text{sca}}}{b_{\text{ext}}} = \frac{b_{\text{sca}}}{b_{\text{sca}} + b_{\text{abs}}} \quad (2)$$

式中, b_{abs} 、 b_{sca} 和 b_{ext} 分别为气溶胶吸收、散射和消光系数, 单位均为 Mm^{-1} .

2 结果与讨论

2.1 颗粒物数浓度粒径谱测量

采用两台扫描电迁移率粒径谱仪 (SMPS) 测量颗粒物数浓度粒径谱. 一台测量颗粒物粒径范围为 2.02 ~ 63.8 nm, 另一台测量颗粒物粒径范围为

14.1 ~ 736.5 nm. 图 3(a) 为整个观测期间 2019 年 5 月 27 日 ~ 6 月 27 日的颗粒物数浓度粒径谱. 5 月 28 日和 29 日可以看到明显新粒子生成过程, 颗粒物粒径随时间显著增长, 粒子粒径从 4 nm 开始持续增长到 64 nm, 在这段时间内颗粒物数浓度急剧增长, 最大可达到 $54\,400 \text{ 个} \cdot \text{cm}^{-3}$, 其余时间段颗粒物数浓度相比于 5 月 28 日和 29 日较低, 最大为 $20\,000 \text{ 个} \cdot \text{cm}^{-3}$.

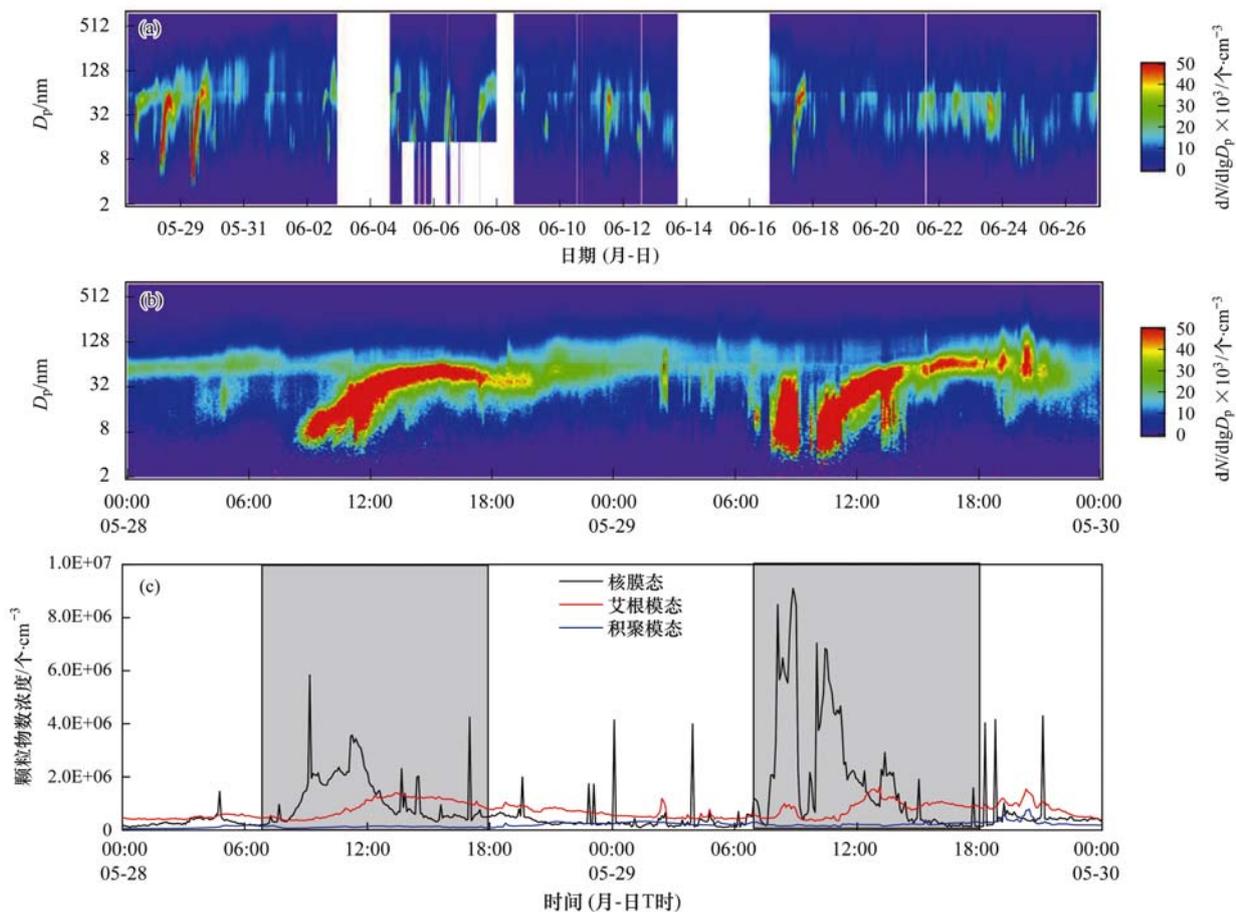
5 月 28 日和 29 日的颗粒物数谱浓度图如图 3(b) 所示, 可以看到从 07:00 开始新粒子开始生成, 18:00 该现象停止, 07:00 ~ 18:00 时之间颗粒物粒径和数目浓度急剧增长, 新粒子粒径达到 64 nm 停止增长, 19:00 ~ 06:00 时之间颗粒物数浓度减小. 将两日的颗粒物浓度按 3 个模态理论划分: 核模态 ($D_p < 30 \text{ nm}$)、艾根模态 ($30 \text{ nm} < D_p < 100 \text{ nm}$) 和积聚模态 ($100 \text{ nm} < D_p < 800 \text{ nm}$) 如图 3(c) 中所示, 在新粒子生成期间(灰色方框标注)核模态变化最明显, 出现多个显著峰值; 在新粒子生成观测期间艾根模态较核模态变化不明显; 积聚模态粒径范围

较大, 且新粒子生成过程中数目偏少, 无明显变化. 新粒子生成期间颗粒物浓度的模态变化与其他观测结果较为一致^[23].

2.2 $\text{PM}_{2.5}$ 化学组分测量

在整个观测期间, 去除相对湿度大于 90% 的小时数据样本^[41], $\text{PM}_{2.5}$ 的日均质量浓度为 $48.19 \mu\text{g} \cdot \text{m}^{-3}$, 浓度范围为 $26.84 \sim 48.19 \mu\text{g} \cdot \text{m}^{-3}$; 碳组分 OC 和 EC 的日均浓度分别为 $9.09 \mu\text{g} \cdot \text{m}^{-3}$ 和 $3.47 \mu\text{g} \cdot \text{m}^{-3}$, 分别占比 18.86% 和 7.20%; 水溶性离子 NO_3^- 、 SO_4^{2-} 和 NH_4^+ 日均浓度分别为 8.82、5.72 和 $4.54 \mu\text{g} \cdot \text{m}^{-3}$, 分别占比 18.29%、11.88% 和 9.42%; 其他水溶性离子 (K^+ 、 Ca^{2+} 、 Mg^{2+} 、 Na^+ 和 Cl^-) 总量占比 5.20%. 图 4(a) 和 4(b) 分别为 $\text{PM}_{2.5}$ 组分日均浓度及占比情况, 灰色线框部分为新粒子生成时段.

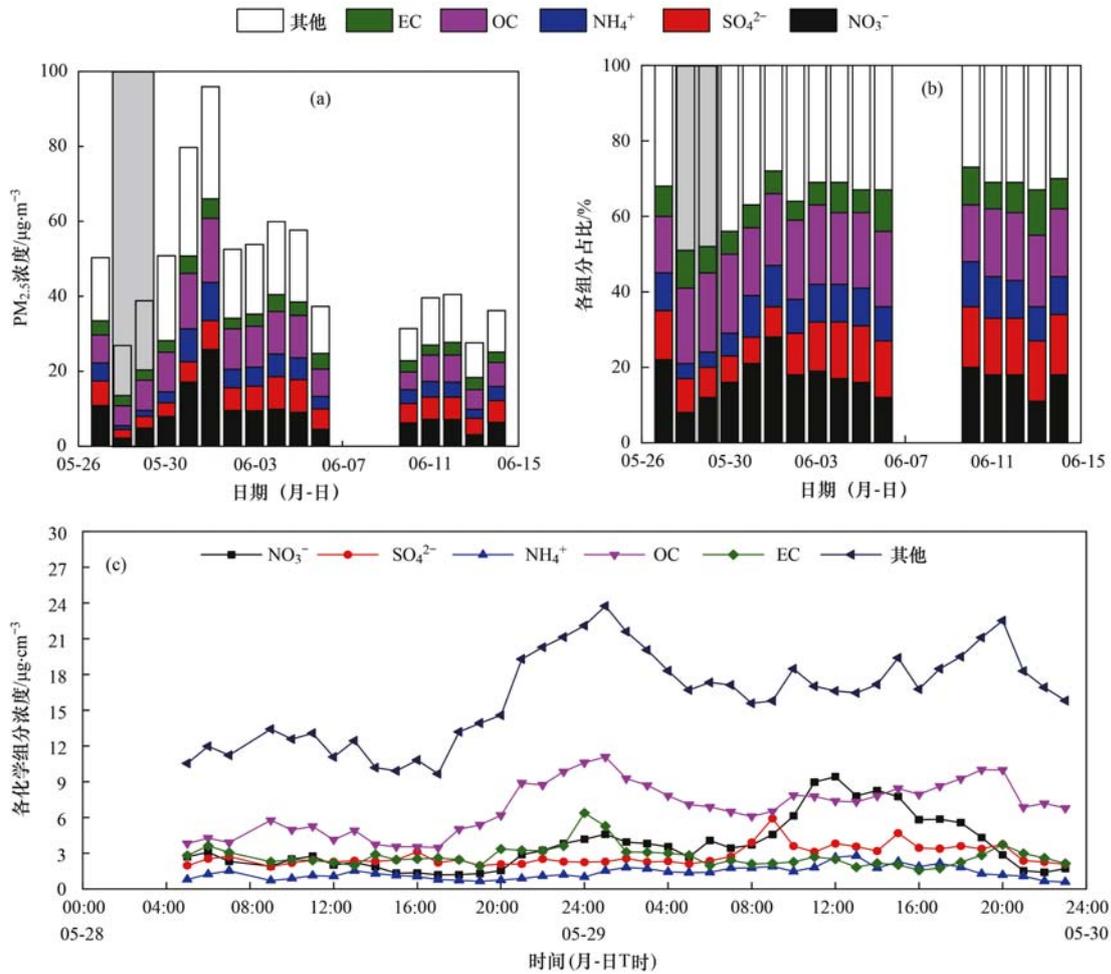
5 月 28 日和 29 日的各组分浓度时序图如图 4(c) 所示, 5 月 28 日 3 种主要水溶性离子浓度无明显变化, 5 月 29 日在新粒子生成前期及初期, 硫酸盐浓度增加明显, 这一结果与霍俊涛等^[42] 和胡敏等^[43] 的研究结果一致.



(a) 观测期间整体数目粒径谱; (b) 新粒子生成期间数目粒径谱; (c) 新粒子生成期间颗粒物三模态数目浓度变化时序图(灰色线框代表新粒子生成时段)

图 3 颗粒物数目粒径谱及 3 个模态数浓度

Fig. 3 Particle size spectrum and 3 modal concentration



(a) 观测期间 $PM_{2.5}$ 各组分浓度; (b) 观测期间各组分平均浓度占比;
(c) 新粒子生成期间组分浓度变化时序图; (a) 和 (b) 中灰色线框为新粒子生成时段
图 4 观测及新粒子生成期间 $PM_{2.5}$ 各组分浓度变化趋势

Fig. 4 Changes in the concentration of $PM_{2.5}$ during the observation period and the period of new particle generation

2.3 气溶胶光学性质测量

2.3.1 实测参数

图 5(a) 和 5(b) 为整个观测期间仪器实时在线测量得到的光学系数(消光、吸收和散射系数)和单次散射反照率(SSA)的变化趋势, 由于 AE33 数据缺失, 删除对应的光学性质参数, 造成图 5(a) 和 5(b) 中部分数据缺失. 消光、吸收和散射系数的平均值为 195.38 、 39.87 和 155.51 Mm^{-1} ; SSA 的平均值为 0.77 , 说明常州地区在观测期间气溶胶散射作用大于吸收作用.

5月28日和29日气溶胶光学性质参数如图5(c)所示, 单次散射反照率如图5(d)所示. 5月28日消光、吸收和散射系数的平均值分别为 62.37 、 27.47 和 34.90 Mm^{-1} , SSA 的平均值为 0.57 ; 5月29日消光、吸收和散射系数的平均值分别为 128.43 、 52.46 和 75.98 Mm^{-1} , SSA 的平均值为 0.60 , 与5月28日相比, 光学性质参数整体偏高. 两日的光学性质参数从新粒子开始生成时间 07:00 开

始降低, 一直持续到 18:00 都处于峰谷, 18:00 之后光学性质参数又开始达到一个高峰, SSA 是在 07:00 时开始升高, 07:00 ~ 18:00 处于一个峰值状态, 18:00 之后开始降低, 与光学性质参数的变化趋势相反, 5月29日碳质气溶胶的平均含量相对于5月28日降低.

2.3.2 组分散射系数重建

应用 IMPROVE 方程研究气溶胶化学组分对光全散射(消光)的贡献, 选定波长为 532 nm . 简化后的 IMPROVE 方程^[5]一般可表述为:

$$b_{sp} = 2.2 \times f_s(\text{RH}) \times [\text{Small}(\text{NH}_4)_2\text{SO}_4] + 4.8 \times f_L(\text{RH}) \times [\text{Large}(\text{NH}_4)_2\text{SO}_4] + 2.4 \times f_s(\text{RH}) \times [\text{Small NH}_4\text{NO}_3] + 5.1 \times f_L(\text{RH}) \times [\text{Large NH}_4\text{NO}_3] + 2.8 \times [\text{Small OM}] + 6.1 \times [\text{Large OM}] + 1.7 \times f_{ss} \times [\text{SS}] + 1.0 \times [\text{FS}] + 0.6 \times [\text{CM}]$$

$$[\text{Large X}] = [\text{Total X}]^2/20, \quad [\text{Total X}] < 20$$

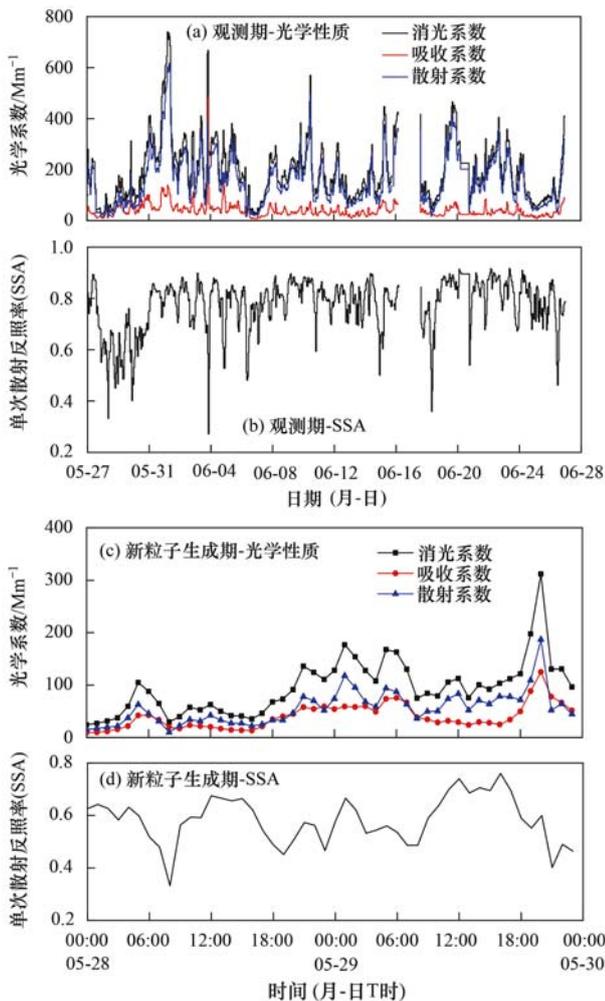


图5 光学性质参数和单次散射反照比变化趋势

Fig. 5 Optical properties and single-scattering albedo trends

$$[\text{Large X}] = [\text{Total X}], [\text{Total X}] \geq 20$$

$$[\text{Small X}] = [\text{Total X}] - [\text{Large X}] \quad (3)$$

式中, $[\]$ 表示气溶胶各化学组分质量浓度 ($\mu\text{g}\cdot\text{m}^{-3}$), 消光系数 b_{sp} 的单位为 Mm^{-1} . $[(\text{NH}_4)_2\text{SO}_4] = 1.37 [\text{SO}_4^{2-}]$; $[\text{NH}_4\text{NO}_3] = 1.29 [\text{NO}_3^-]$; $[\text{OM}] = 1.4 [\text{OC}]$; $[\text{FS}] = 2.2 [\text{Al}]$

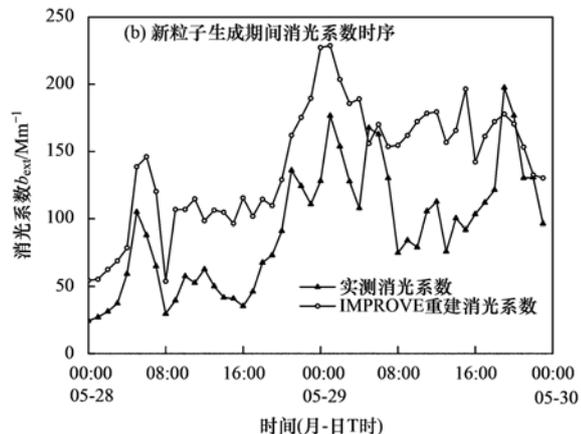
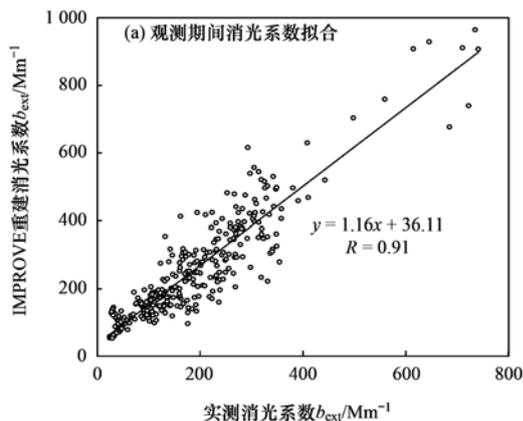


图6 实测与 IMPROVE 方程估算消光系数线性拟合和时序

Fig. 6 Measured and IMPROVE-calculated linear fitting and timing diagram of the extinction coefficient

$+ 2.49 [\text{Si}] + 1.62 [\text{Ca}] + 2.42 [\text{Fe}] + 1.94 [\text{Ti}]$; $[\text{CM}] = [\text{PM}_{10} - \text{PM}_{2.5}]$; 由于缺乏土壤元素的测量, 假定 Ca^{2+} 占细土壤质量浓度的 5%, 即 $[\text{FS}] = 20 [\text{Ca}^{2+}]$; 吸湿性增长曲线参考 Jung 等^[44] 的研究.

图 6(a) 为整个观测期间的实测消光系数与组分重建消光系数的线性拟合, 相关系数 $R^2 = 0.83$, 相较于南京 (0.89) 偏低, 高于杭州 (0.81), 与上海 (0.83) 数值一致, 说明相关性吻合较好. 图 6(b) 为新粒子生成期间实测消光系数与重建消光系数的时序对比图, 实测平均消光系数为 95.40 Mm^{-1} , 重建平均消光系数为 140.20 Mm^{-1} , 重建消光系数较实测值高 44.80 Mm^{-1} .

2.3.3 数浓度粒径谱重建消光系数

MIE 散射理论计算气溶胶全散射 (消光) 系数根据迭代算法确定颗粒物复折射率, 即颗粒物相对于周围空气环境的复折射率, 用 $m = n - k \times i$ 表示, 折射率实部 n 主要与光散射有关, 折射率虚部 k 主要与光吸收有关^[45]. 由于新粒子生成时颗粒物的混合状态会发生改变, 因此假设新粒子生成期间颗粒物的平均复折射率为 $m_1 = 1.233 - 0.002 \times i$, 其余时间段颗粒物平均复折射率为 $m_2 = 1.24 - 0.002 \times i$, 结合颗粒物数谱得到整个观测期间消光系数的最大值为 395.44 Mm^{-1} , 最小值为 21.12 Mm^{-1} , 平均值为 126.52 Mm^{-1} .

图 7 分别为实际测量与 MIE 理论计算的消光系数线性回归分析和新粒子生成期间消光系数时序图. 在整个观测期间, 实测与理论值相关系数 $R^2 = 0.83$, 与 IMPROVE 重建相关系数相等. 在新粒子生成期间理论消光系数平均值 93.54 Mm^{-1} , 且新粒子生成期间 MIE 理论重建时序值与实测值有一致的变化趋势, 平均理论消光系数要比实测值低 1.86 Mm^{-1} .

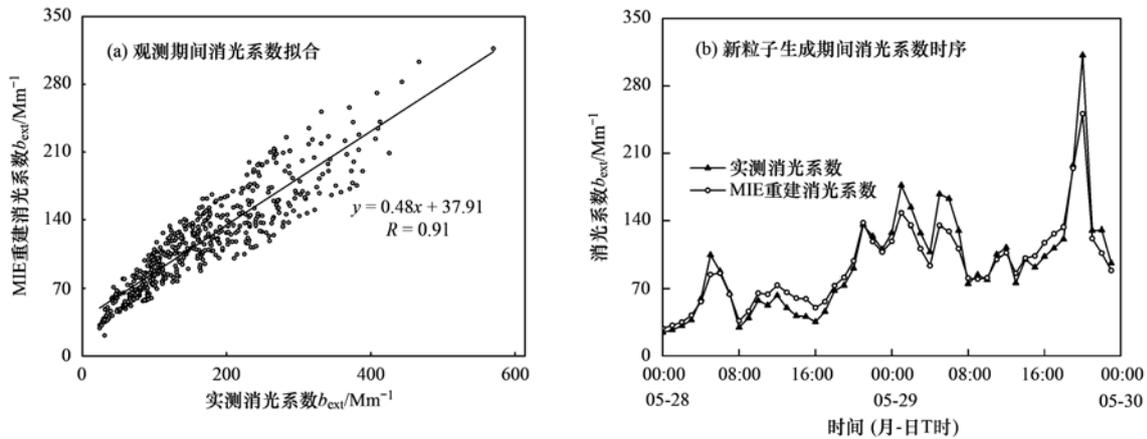


图7 实测与MIE理论计算消光系数线性拟合和时序

Fig. 7 Measured and MIE-calculated linear fitting and timing diagram of the extinction coefficient

3 结论

(1) 颗粒物数谱浓度表明: 新粒子集中在日间生成, 最大粒径可达到 64 nm, 主要影响艾根模态以及核膜态。

(2) 整个观测期间颗粒物化学组分有机碳占比最高, 其次是硝酸盐; 根据时序变化图推测在新粒子生成初期硫酸盐贡献较大, 其次是硝酸盐和铵盐。

(3) 对颗粒物单次散射反照比 (SSA) 进行分析发现新粒子生成影响颗粒物散射系数较明显。

(4) 新粒子生成期间 IMPROVE 重建消光系数较实测均值高 44.80 Mm^{-1} , 相关系数为 0.83, 该方程在不同地区的适用性有待进一步研究。

(5) 新粒子生成期间用 MIE 理论结合颗粒物数目粒径谱计算消光系数较实测均值低 1.86 Mm^{-1} , 相关系数为 0.83, 但是趋势一致, 推测由于联用仪器的进口流量差异, 造成不同粒径的颗粒物损失。

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