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Q1 Insights into the formation of secondary organic 2 carbon in the summertime in urban Shanghai

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A B S T R A C T

To investigate formation mechanisms of secondary organic carbon (SOC) in Eastern China, ambient measurements were conducted in an urban site in Shanghai in the summer of 2015. A period of high O₃ concentrations (daily peak >120 ppb) was observed, during which daily maximum SOC concentrations exceeding 9.0 μg/(C·m³) continuously for a week. Diurnal variations of SOC concentration and SOC/OC ratio exhibited both daytime and nighttime peaks. The SOC concentrations correlated well with O_x (=O₃ + NO₂) and relative humidity in the daytime and the nighttime, respectively, suggesting that secondary organic aerosol formation in Shanghai is driven by both photochemical production and aqueous phase reactions. Single particle mass spectrometry was used to examine the formation pathways of SOC. Along with the daytime increase of SOC, the number fraction of elemental carbon (EC) particles coated with organic carbon (OC) quickly increased from 38.1% to 61.9% in the size range of 250–2000 nm, which was likely due to gas-to-particle partitioning of photochemically generated semi-volatile organic compounds onto EC particles. In the nighttime, particles rich in OC components were highly hygroscopic, and the number fraction of these particles correlated well with relative humidity and SOC/OC nocturnal peaks. Meanwhile, as an aqueous-phase SOC tracer, particles that contained oxalate-Fe(III) complex also peaked at night. These observations suggested that aqueous-phase processes had an important contribution to the SOC nighttime formation. The influence of aerosol acidity on SOC formation was studied by both bulk and single particle level measurements, suggesting that aqueous-phase formation of SOC was enhanced by particle acidity.

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54 Introduction

56 Secondary organic aerosol (SOA) constitutes a large proportion of
 57 fine particle mass in the atmosphere (Hayes et al., 2013; Jimenez

et al., 2009; Kanakidou et al., 2005; Zhang et al., 2007a) and plays 58
 an important role in health impairment, visibility reduction and 59
 climate change (Hand et al., 2012; Mauderly and Chow, 2008). SOA 60
 is a major source of brown carbon and has been estimated to 61
 account for 15%–50% of total light absorption in the atmosphere 62

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63 and snow (Chung et al., 2012; Liu et al., 2014). Compared
64 to primary organic aerosol (POA), SOA generally owns higher
65 hygroscopicity that will influence particle optical properties and
66 cloud condensation nuclei formation (Chen et al., 2012; Pöschl
67 et al., 2010; Pilinis et al., 1995).

68 Due to the importance of SOA, formation pathways of SOA
69 have been a focus in aerosol research in the past 3 decades.
70 A major formation pathway of SOA is the photo-oxidation
71 of volatile organic compounds (VOCs) in gas phase followed
72 by gas-to-particle partitioning of low volatility products (Kroll
73 and Seinfeld, 2008; Odum et al., 1996). Both anthropogenic VOCs
74 (e.g., aromatic hydrocarbons) and biogenic VOCs such as
75 isoprene can serve as precursors for SOA through photo-
76 oxidation reactions (Ding et al., 2012; Henze et al., 2008).
77 Recently, aqueous-phase enhanced gas-to-particle partitioning
78 processes and reactions draw more attentions. Kinetic models
79 suggested that aqueous-phase processes might have a sub-
80 stantial contribution to the total SOA mass, which was
81 underestimated before (Lim et al., 2010; Volkamer et al., 2009).
82 Field and laboratory studies based on aerosol mass spectrom-
83 etry (AMS) indicated that ambient SOA presented a higher
84 oxygen-to-carbon (O/C) ratio than SOA formed in dry smog
85 chamber experiments (Aiken et al., 2008; Chen et al., 2015;
86 Ng et al., 2010). Aqueous reactions can produce highly oxidized
87 SOA with O/C ratios comparable to the most oxidized SOA
88 components observed in ambient air (Sun et al., 2010; Yu et al.,
89 2014). Furthermore, SOA tracers showed a better correlation
90 with aerosol liquid water than organic aerosol mass (Hennigan
91 et al., 2009; Zhang et al., 2012). These observations revealed
92 the aqueous-phase processes are important in the formation
93 of SOA.

94 Despite much progress has been achieved, many aspects of
95 SOA formation pathways remain unclear (Ervens et al., 2011;
96 Hallquist et al., 2009). In particular, the complexity of SOA
97 formation is a major uncertain factor for predicting the
98 concentration, property, and environmental effects of organic
99 aerosol (Hodzic and Jimenez, 2011; Kanakidou et al., 2005;
100 Shrivastava et al., 2017). However, so far there have been
101 relatively few ambient studies that examined both daytime and
102 nighttime SOA formation during one experiment. Moreover,
103 few studies have reported the impact that aerosol compositions
104 have on the SOA formation in ambient conditions and most of
105 them were based on measurements of ensemble particles, thus
106 lacking of detailed information on mixing state of SOA at single
107 particle level. Single particle mass spectrometry has been used
108 to characterize certain SOA species (e.g., amine, oxalate, and
109 organosulfate) (Hatch et al., 2011; Huang et al., 2012; Rehbein
110 et al., 2011; Yang et al., 2009), but few of these works offered a
111 whole picture on the mixing state of SOA.

112 To investigate the mixing state and formation pathways of
113 SOA in Eastern China, we conducted a two-month field mea-
114 surement study in summer at an urban site in Shanghai —
115 a megacity with a population of 24 million. The summer in
116 Shanghai is characterized by high temperature, high humidity,
117 intense solar radiation, and abundant anthropogenic and bio-
118 genic gaseous precursors (C. Huang et al., 2011; Ma et al., 2014),
119 supplying a favorable environment for SOA formation. We
120 combined quantitative measurement of secondary organic
121 carbon (SOC) concentration with online single particle analysis
122 to study the diurnal variation of SOC production. The ambient

oxidative capacity, particle mixing states and aerosol liquid
123 water content were analyzed. As an indicator of SOC, oxalate
124 particles were selected to verify the formation pathways of
125 SOC. The impacts of the aerosol acidity on the observed SOC
126 formation were also examined. 127

1. Methodology 128

1.1. Sampling location and measurements 130

This field campaign was conducted at the Shanghai Air Supersite
131 at Pudong (31.22838°N, 121.53301°E) from July to September, 2015.
132 As a typical station for studying aerosol characteristics in the
133 megacity, this site is located in the downtown of Shanghai, close
134 to the Century Avenue, a very busy street with continuous high
135 traffic volume in the daytime, and only about 3 km away from
136 the central business district Lujiazui (Fig. S1). Experimental
137 instruments were deployed on the roof of a six-story building
138 (more than 20 m above the ground), with little influence from
139 road dust. 140

PM_{2.5} mass concentration was measured by a Tapered
141 Element Oscillating Microbalance combined with Filter Dynamic
142 Measurement System (TEOM-FDMS, TEOM 1405-F, ThermoFisher
143 Scientific Inc., USA.), in which FDMS can complement the lost
144 volatile components to PM_{2.5} after TEOM sampling. The concen-
145 trations of gas phase NO₂, SO₂, CO, O₃ were measured by Thermo
146 Environmental Instruments (Model 42i, 43i, 48i, 49i respectively,
147 ThermoFisher Scientific Inc., USA.). Meteorological parameters
148 (relative humidity, wind speed, wind direction, temperature
149 and rain event) were acquired from a meteorological station
150 (Vaisala Corp., Finland). The solar radiation was measured by
151 a pyranometer (Kipp & Zonen, CMP6, Netherlands). Backward
152 trajectories were calculated using the Hybrid Single Particle
153 Lagrangian Integrated Trajectory (HYSPPLIT) model (Stein et al.,
154 2015). 155

Single particle size and chemical information were obtained
156 from the Single Particle Aerosol Mass Spectrometer (SPAMS,
157 Hexin Analytical Instrument Co., Ltd. China.). Individual particles
158 were firstly sized and then desorbed/ionized to generate positive
159 and negative mass-spectra. The effective sampling range of
160 SPAMS in term of particle size is from 250 to 2000 nm (Li et al.,
161 2011). Detailed description of the SPAMS is available in the
162 supplementary material and the previous publications (Gong
163 et al., 2016; Li et al., 2011). 164

Hourly mass concentrations of elemental carbon (EC) and
165 organic carbon (OC) were measured by a semi-continuous OC/EC
166 analyzer (Model 4, Sunset Laboratory Inc., Portland, USA.)
167 based on the National Institute of Occupational Safety and
168 Health thermal/optical transmittance measurement protocol
169 (NIOSH 5040), with a PM_{2.5} impactor inlet. A charcoal denuder
170 was installed in the sampling line to prevent the adsorption
171 of organic vapors on the filter, and the sucrose solution was
172 used for the external calibration to assure the measurement
173 accuracy. 174

A Monitor for Aerosols and Gases in Air (MARGA,
175 Applikon Analytical B. B. Corp., ADI 2080, Netherlands), with
176 a PM_{2.5} cyclone impactor, was deployed to measure the gases
177 and particulate water-soluble inorganic species in hourly
178 resolution. Detailed description about the MARGA is available
179

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