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Seasonal variation and size distribution of biogenic secondary organic aerosols at urban and continental background sites of China

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ABSTRACT

Size-resolved biogenic secondary organic aerosols (BSOA) derived from isoprene and monoterpene photooxidation in Qinghai Lake, Tibetan Plateau (a continental background site) and five cities of China were measured using gas chromatography/mass spectrometry (GC/MS). Concentrations of the determined BSOA are higher in the cities than in the background and are also higher in summer than in winter. Moreover, strong positive correlations ($R^2 = 0.44\text{--}0.90$) between BSOA and sulfate were found at the six sites, suggesting that anthropogenic pollution (i.e., sulfate) could enhance SOA formation, because sulfate provides a surface favorable for acid-catalyzed formation of BSOA. Size distribution measurements showed that most of the determined SOA tracers are enriched in the fine mode ($<3.3\ \mu\text{m}$) except for cis-pinic and cis-pinonic acids, both presented a comparable mass in the fine and coarse ($>3.3\ \mu\text{m}$) modes, respectively. Mass ratio of oxidation products derived from isoprene to those from monoterpene in the five urban regions during summer are much less than those in Qinghai Lake region. In addition, in the five urban regions relative abundances of monoterpene oxidation products to SOA are much higher than those of isoprene. Such phenomena suggest that BSOA derived from monoterpenes are more abundant than those from isoprene in Chinese urban areas.

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Introduction

Secondary organic aerosols (SOA) are important constituents of particulate matters (PMs) in the atmosphere (Wang et al., 2015),

and affect weather and climate directly by absorbing and scattering sun light and indirectly by acting as cloud condensation nuclei (Surratt et al., 2016; Turpin and Huntzicker, 1995). On the global scale SOA derived from biogenic origins is much

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more abundant than that from anthropogenic sources, because biogenic volatile organic compounds (BVOCs) are predominant in the atmosphere (Griffin et al., 1999; Hoffmann et al., 1997). Isoprene and monoterpenes are the major BVOCs, accounting for 44% and 11% of the global BVOCs, respectively (Guenther et al., 1995). Globally, around two-thirds of BVOCs are subject to photochemical oxidation with anthropogenic pollutants (Paulot et al., 2009), and NO_x and SO₂ take important roles during the oxidation processes (Ng et al., 2016; Wang et al., 2016; Xu et al., 2015). A recent modeling study predicts that globally up to 70% of isoprene is photochemically oxidized under high-NO_x conditions (urban or industrial regions) (Paulot et al., 2009). NO_x can alter SOA formation by influencing peroxy radical chemistry in BVOCs oxidation process (Claeys et al., 2013; Surratt et al., 2016). Moreover, NO₃ radical, which is formed through the reaction of NO₂ with O₃, can oxidize BVOCs and form condensable products that have high SOA yields (Ng et al., 2016; Xu et al., 2015). Laboratory chamber simulations have shown that SOA mass yield from isoprene oxidation in the presence of NO_x could increase dramatically with pre-existing acidified sulfate seeds (1%–3% and 3%–30% under high- and low-NO_x conditions, respectively) (Chan et al., 2010; Chu et al., 2014; Surratt et al., 2006, 2010), suggesting that both SO₂ and NO_x may have a synergetic effect on BSOA formation.

Due to rapid economy expansion and urbanization atmospheric NO_x level in China has been increasing (Zhang et al., 2015). The abundant NO_x, together with high level of SO₂, could promote the photochemical oxidation of volatile organic compounds including BVOCs, leading to abundant SOA in the country (Huang, 2014; Huang et al., 2017; Wang et al., 2017). In the current work, size-segregated aerosols were collected in five megacities of China and Qinghai Lake, a Tibetan Plateau background region, and determined for biogenic secondary organic aerosols (BSOA) derived from isoprene and monoterpenes, respectively. The purposes of this study were to investigate the seasonal and spatial variations in abundances and compositions of BSOA in the six regions and explore the role of sulfate in BSOA formation process.

1. Experimental section

1.1. Aerosol sampling

Aerosol samples were collected in five cities and Qinghai Lake of China, i.e., Urumqi (43.9°N and 87.5°E), Xi'an (34.3°N and 108.9°E), Shanghai (31.4°N and 121.5°E), Chengdu (30.7°N and 104.0°E), Guangzhou (23.4°N and 113.3°E) (Fig. 1). In the background site, the sampling was conducted at Bird Island, which is located at the northwestern rim of Qinghai Lake. In Urumqi, samples were collected on the rooftop of a three-story building (about 10 m above the ground) on the campus of the Institute of Desert Meteorology at the urban center. In Xi'an, aerosol samples were collected on the rooftop of a three-story building (about 10 m above the ground) on the campus of the Institute of Earth Environment of CAS, which is located in the downtown area of the city. Samples from Shanghai were collected on the rooftop of Shaw House on the campus of Fudan University in the city. In Chengdu, samples were collected on the rooftop of a three-story building (about

10 m above the ground) on the campus of Chengdu Institute of plateau Meteorological at the urban center. In Guangzhou, aerosol samples were collected on the rooftop (50 m above the ground) of a building on the campus of the South China Institute of Environmental Science, which is located in the urban center of Guangzhou.

The sampling time and meteorological parameters during each of the sampling periods are given in Table 1. All the size-segregated samples were collected using Anderson 8-stage air samplers (PSW-8, Thermo-electronic Company, USA) with the cutoff sizes of 9.0, 5.8, 4.7, 3.3, 2.1, 1.1, 0.7 and 0.4 μm at an airflow rate of 28.3 L/min during the summer and winter of 2012. At the five urban sites the size-segregated aerosol sampling were performed for 3 days in each set, while the aerosol collection in Qinghai Lake was conducted for 6 days in each set in order to obtain enough materials. A set of field blank samples were also collected at each site in each season by mounting the filters onto the sampler for about 10 min without pumping air. A total of 58 sets were collected with 25 sets for the summer and 33 sets for the winter, respectively. All the samples were collected onto pre-baked (450°C for 8 hr) quartz fiber filter. After sampling, the filters were individually sealed in an aluminum foil bag and stored at –18°C prior to analysis.

1.2. Sample extraction, derivatization and GC/MS analysis

The details of sample extraction and derivatization were documented elsewhere (Ren et al., 2017a, 2017b; Wang and Kawamura, 2005). Here we only give a brief introduction. One fourth of the filter was extracted with a mixture of dichloromethane/methanol (2:1, V/V) three times each for 15 min at room temperature. Then the extracts were concentrated to dryness and derivatized with *N*, *O*-bis-(trimethylsilyl) trifluoroacetamide at 70°C for 3 hr. Finally the derivatives were analyzed using an Agilent GC-MS system (HP7890 plus GC-5975N MSD, Agilent Co., USA).

A total of 12 target compounds were quantified (see Table 2), which are isoprene SOA tracers (3-MeTHF-3,4-diols, 2-methylglyceric acid, C5-alkene triols and 2-methyltetros) and α-pinene SOA tracers (cis-pinonic acid, cis-pinonic acid, 3-hydroxyglutaric acid and 3-methyl-1,2,3-butanetricarboxylic acid). These α-pinene SOA tracers can also be generated by β-pinene and *d*-limonene oxidation (Jaoui et al., 2005); thus, here we use monoterpenes instead of α-pinene when discussing the precursors.

1.3. Quality assurance/quality control

Airflow rate of the sampler was calibrated for twice at each site in each season before and after the sampling by using the mass flowmeter (4100 Series Flowmeters, TSI, USA.). Field blank filters were also handled by the procedure described above. The results showed that no serious contamination was found (less than 5% of the mass in the real samples). During the GC-MS analysis, the response factors of cis-pinonic acid and cis-pinonic acid (PA) were determined using authentic standards, while owing to the commercial unavailability, GC/MS response factors of 3-MeTHF-3,4-diols (*trans*-3-methyltetrahydrofuran-3,4-diol and *cis*-3-methyltetrahydrofuran-3,4-diol), 2-methylglyceric

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