



Short communication

Missing ozone-induced potential aerosol formation in a suburban deciduous forest



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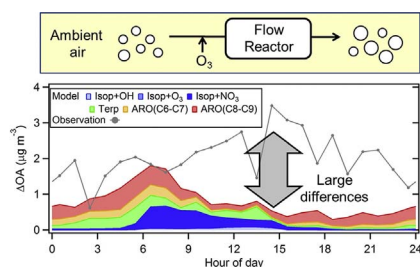
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GRAPHICAL ABSTRACT



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ABSTRACT

As a new approach to investigating formation processes of secondary organic aerosol (SOA) in the atmosphere, ozone-induced potential aerosol formation was measured in summer at a suburban forest site surrounded by deciduous trees, near Tokyo, Japan. After passage through a reactor containing high concentrations of ozone, increases in total particle volume (average of $1.4 \times 10^9 \text{ nm}^3/\text{cm}^3$, which corresponds to 17% that of pre-existing particles) were observed, especially during daytime. The observed aerosol formations were compared with the results of box model simulations using simultaneously measured concentrations of gaseous and particulate species. According to the model, the relative contributions of isoprene, monoterpene, and aromatic hydrocarbon oxidation to SOA formation in the reactor were 24, 21, and 55%, respectively. However, the model could explain, on average, only ~40% of the observed particle formation, and large discrepancies between the observations and model were found, especially around noon and in the afternoon when the concentrations of isoprene and oxygenated volatile organic compounds were high. The results suggest a significant contribution of

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missing (unaccounted-for) SOA formation processes from identified and/or unidentified volatile organic compounds, especially those emitted during daytime. Further efforts should be made to explore and parameterize this missing SOA formation to assist in the improvement of atmospheric chemistry and climate models.

1. Introduction

Aerosol particles in the atmosphere are known to have significant impacts on climate, the atmospheric environment, ecosystems, and human health (Boucher, 2015; Lelieveld et al., 2015). Secondary organic aerosol (SOA) particles, which are generated during the oxidation of volatile organic compounds (VOCs) emitted from both biogenic and anthropogenic sources, constitute a large fraction of submicron particles in the atmosphere (Jimenez et al., 2009). However, the formation processes of SOA in the atmosphere remain largely uncertain (e.g. Hallquist et al., 2009; Chen et al., 2015; Glasius and Goldstein, 2016).

Formation yields of SOA (defined as the ratio of generated SOA mass to the amount of reacted VOC) from various VOCs have been determined based on experiments using smog chambers or oxidation flow reactors (Hallquist et al., 2009; Lambe et al., 2015; Bruns et al., 2015; and references therein). Although the dependency of the formation yields on a number of parameters, such as temperature, relative humidity (RH), seed particles, co-existing gases, and species and exposures of oxidants, has been investigated, it is almost impossible to investigate all SOA formation processes in the real atmosphere, including the possible interactions between different oxidation processes and the various parameters.

A number of observational studies have been conducted to estimate SOA formation processes in the atmosphere by measuring tracer compounds such as *cis*-2-methyl-1,3,4-trihydroxy-1-butene, 2-methylthreitol, and 2-methylerythritol for isoprene-derived SOA; 3-isopropylglutaric acid, *cis*-pinonic acid, and pinic acid for monoterpene-derived SOA; and 2,3-dihydroxy-4-oxopentanoic acid for aromatic hydrocarbon-derived SOA (e.g. Kleindienst et al., 2007a, 2010; Hu et al., 2008; Ding et al., 2012). However, there are potentially large uncertainties in such estimations because the yields of tracer molecules in the atmosphere may not necessarily be the same as those obtained in laboratory experiments owing to the complexity of the SOA formation process and possible chemical transformation of the tracer molecules in the real atmosphere.

As an alternative approach to obtaining information on SOA formation processes, the monitoring of potential SOA formation after inducing ambient air perturbations may be useful. Very recently, Jimenez and coworkers reported potential aerosols formation in a Potential Aerosol Mass (PAM) flow reactor, with perturbations caused by oxidant additions (Ortega et al., 2016; Palm et al., 2016, 2017). For an urban area in the Los Angeles region of the USA in late spring, Ortega et al. (2016) reported that reactor SOA formation under irradiation by ultraviolet (UV) light, for which VOCs were estimated to be mainly oxidized by hydroxyl radicals (OH), peaked at night and correlated with trimethylbenzene concentrations. At a pine forest in Colorado, USA, in summer, Palm et al. (2016, 2017) also reported greater reactor SOA formation at night for all cases of UV irradiation, ozone (O₃) addition, and nitrate radical (NO₃) addition; this was mainly due to the oxidation of monoterpenes. Whereas the observed reactor SOA formations for O₃ and NO₃ additions were consistent with the predicted SOA formations based on measured monoterpene and sesquiterpene concentrations, the observed reactor SOA formation under UV irradiation (OH oxidation) was reported to be 3.4 times larger than that of the model prediction (Palm et al., 2016, 2017).

In the present study, potential aerosol formation with the addition of O₃ was investigated by comparing total particle volumes with and without passing the particles through an oxidation flow reactor. The study was carried out in summer at a suburban forest site surrounded by deciduous trees, near Tokyo, Japan. By comparing the observed SOA

formation in the reactor with that predicted by the model using the reported SOA formation yields from chamber experiments, the contribution of unidentified SOA formation processes is highlighted. Because small forests with deciduous trees are ubiquitous in urban, suburban, and rural areas across the world, the results obtained in the present study will be widely applicable to regions having similar environments, where both biogenic and anthropogenic emissions are expected to influence SOA formation. This study aimed to demonstrate that observing potential aerosol formation to reveal unidentified SOA formation processes is useful in improving model predictions for evaluating impact of aerosol particles.

2. Material and methods

2.1. Observations

Observations were conducted from July 31 to August 8, 2015 as part of the AQUAS-TAMA (Air Quality Study at FM Tama) 2015 field campaign at the Field Museum Tama Hill (FM-Tama, 35.6385°N, 139.3781°E) measurement station of Tokyo University of Agriculture and Technology (Matsuda et al., 2015). This station is located in a suburban forest (~0.115 km²) and is surrounded by a residential area ~30 km west of the center of Tokyo (Fig. S1). The dominant tree species in the forest are *Quercus serrata* (Japanese konara oak), *Quercus acutissima* (sawtooths oak), and *Cryptomeria japonica* (Japanese cedar).

A schematic diagram of the experimental apparatus used to measure aerosol formation potential is shown in Fig. S2. Aerosol particles were sampled through an inlet placed 4.6 m above the ground surface. The size distributions of particles between 17.5 and 982.2 nm, which were either passed or not passed through the reactor, were simultaneously measured every 5 min using two scanning mobility particle sizers (SMPSs) (TSI, model 3936L72), SMPS-1 and SMPS-2, the sampling and sheath flow rates of which were fixed at 0.2 and 2.0 L per minute (lpm), respectively. These size distributions were measured under dry conditions by passing the particles through diffusion dryers with silica gel before introduction to the SMPSs. A 33L stainless steel cylindrical cell was used as the flow reactor. The O₃ was generated by irradiating filtered oxygen gas (Japan Fine Products, > 99.999%) with ultraviolet light at 184.9 nm from two mercury (Hg) lamps (Hamamatsu, L937-02). The O₃ was continuously added to the reactor at a flow rate of 0.020 lpm. Note that no particle formation was observed when O₃ was added after passing ambient air thorough an activated carbon denuder, suggesting that the contribution of impurities in the oxygen cylinder to aerosol formation in the reactor was negligible. The concentration of O₃ in the reactor was monitored using a UV absorption analyzer (Thermo Scientific, model 49ij). The total flow rate and average residence time in the reactor were 0.6 lpm and 55 min, respectively. The total particle volume generated in the reactor was obtained by subtracting the total particle volume of aerosol that was passed through the reactor from that measured without passage through the reactor, after taking account of the delay and dull effects of residence time in the reactor. The effects of particle loss and differences in the sensitivity of the two SMPSs were corrected using the particle size distributions obtained during periods when the Hg lamps were turned off (July 31, August 3-4, and August 8). Size dependent particle dry deposition was used for the corrections. Typical particle losses were between 28% and 48% for particles with diameters between 100 nm and 500 nm.

During the campaign, ambient concentrations of gaseous nitrogen oxides (NO and NO₂), carbon monoxide (CO), and O₃ were monitored using commercial instruments. The concentrations of VOCs, such as

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