



Effects of inorganic seeds on secondary organic aerosol formation from photochemical oxidation of acetone in a chamber



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HIGHLIGHTS

- Acetone is confirmed to be one of the sources of SOA.
- Participation of inorganic seeds into acetone SOA is determined.
- Chemical compositions and formation mechanism of acetone SOA are proposed.

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ABSTRACT

Photochemical oxidations of acetone were studied under different inorganic seed (NaCl, (NH₄)₂SO₄ and NaNO₃) conditions in a self-made chamber. The results show that no secondary organic aerosol (SOA) can be formed in the experiments either in the absence of artificially added seed particles or in the presence of solid status of the added particles. Liquid water content is the key factor for the formation of SOA in the experiments with seeds. The amount of SOA was only about 4–7 μg m⁻³ in the experiments with the initial acetone of ~15 ppm under different seed conditions. The analysis of SOA compositions by Exactive-Orbitrap mass spectrometer equipped with electro-spray interface (ESI-MS) shows that chlorine-containing and sulfur-containing compounds were detected in SOA formed from the experiments with NaCl and (NH₄)₂SO₄ seeds, respectively, which were not identified in SOA from those with NaNO₃. The compositions of SOA were mainly esters, organonitrates, hydroperoxides, etc. It is concluded that inorganic seed particles participated into the formation of SOA. Acetone SOA was mainly formed in the aqueous phase in which dissolved SOA precursors underwent further oxidation reactions, esterification reactions and/or radical-radical reactions. Our experiments further demonstrate that low-molecular-weight VOCs, such as acetone, can form SOA under certain conditions in the atmosphere, although their contributions to SOA may not be large.

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1. Introduction

Acetone (CH₃COCH₃) is the simplest ketone, which contains 3 carbons and is one of the important atmospheric carbonyl-containing volatile organic compounds (VOCs). Acetone accounts

for 22.5% of the total concentration of carbonyl-containing VOCs in the atmosphere of Chinese megacities like Beijing, Shanghai and Guangzhou (Wang et al., 2014). The biomass burning, direct biogenic emissions, primary anthropogenic emissions, and secondary formation from atmospheric oxidations of acetone precursor VOCs are the main sources of atmospheric acetone (Singh et al., 1994). The global source of acetone was estimated to be 95 Tg yr⁻¹, and the concentration of acetone in the troposphere was 0.2–3 ppb (Jacob et al., 2002). Moreover, the concentration of acetone can reach up to 6 ppb in summer in Nanjing urban district, China (Yang

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et al., 2016). Acetone is reactive and can be removed through photolysis, reacting with OH radicals and surface deposition. Since photolysis of acetone mainly occurs at the wavelength of 290–320 nm in the troposphere, the reaction with OH radicals is more efficient in the lower troposphere (Arnold and Chipperfield, 2004). The peroxy radicals formed from acetone have a great influence on the oxidative capacity of the troposphere (Orlando and Tyndall, 2012). Thus, it is necessary to understand the process of oxidation reactions of acetone and to study whether gas-particle partitioning would occur under some conditions.

Secondary organic aerosol (SOA) widely exists in the troposphere as an important component of fine particulate matter (Pandis et al., 1992; Hallquist et al., 2009; Spracklen et al., 2011; Huang et al., 2014), which has a great influence on the air quality, oxidative capacity of the troposphere and the health of human being (Jacobson et al., 2000; Zhang et al., 2014; Bai, 2014). Previous studies have demonstrated that some low-molecular-weight (LMW) VOCs, such as ethylene, acetylene and propylene, which contain 2–3 carbons, can form SOA under some certain conditions (Volkamer et al., 2009; Jia and Xu, 2016; Ge et al., 2016, 2017a, 2017b). In these work, it has been shown that seed particles and liquid water content (LWC) are the key factors for the formation of relatively large amount of SOA, and LWC can generally enhance the formation of SOA. Ethylene ozonolysis can also form SOA without seeds at dry conditions (<1% RH), although the amount of SOA is less than $1 \mu\text{g m}^{-3}$ (Sakamoto et al., 2013). However, whether acetone can form SOA or not under some certain conditions has not been studied before.

There are different kinds of preexisted particles in the atmosphere. Inorganic particles are the most common particles (Wang et al., 2011). Different types of particles have different effects on the formation of SOA. Volkamer et al. (2009) found that the yield of C_2H_2 SOA, which did not correlate with the organic mass portion of different inorganic and/or organic seeds, increased with the liquid water of the seed. However, Volkamer et al. did not analyze the influence of different seed particles on SOA compositions. Liggio et al. (2005) firstly pointed out that organic sulfates can be formed through the heterogeneous reactions of glyoxal in the presence of $(\text{NH}_4)_2\text{SO}_4$ seeds, which has been further demonstrated in later studies (Surratt et al., 2007; Minerath et al., 2008; Schindelka et al., 2013; Worton et al., 2013; Beardsley and Jang, 2016). Ge et al. (2017a) found that chlorine-containing organic compounds in propylene SOA can be identified in the presence of NaCl seeds, but they did not study the role of different seeds in the formation of propylene SOA. The researches about the influence of seed aerosols on SOA compositions from LMW VOCs are still scanty.

Huang et al. (2014) indicated that during the severe haze pollution in four cities of China in 2013, sulfate concentrations in the atmosphere were $11\text{--}26 \mu\text{g m}^{-3}$, accounting for 8–18% of the total amount of $\text{PM}_{2.5}$. Similarly, nitrate and chloride concentrations were $7.1\text{--}13.6 \mu\text{g m}^{-3}$ and $1.5\text{--}3.9 \mu\text{g m}^{-3}$, accounting for 7–14% and 2–4% of the total amount of $\text{PM}_{2.5}$, respectively. Thus, these three species of inorganic compounds of sulfate, nitrate and chloride took up about one third of $\text{PM}_{2.5}$ mass concentrations. Certainly, a high SOA yield can occur in the coastal atmosphere because of the strong hygroscopic properties of sea-salt particles (Beardsley et al., 2013).

In this work, we studied the SOA formation from acetone photolysis in a self-made smog chamber, and further analyzed the influences of different types of seeds (NaCl, $(\text{NH}_4)_2\text{SO}_4$, NaNO_3) on the chemical compositions of SOA. The formation mechanism of acetone SOA was proposed. To our knowledge, this is the first time to study the process of SOA formation from photochemical oxidation of acetone, and identify the chemical compositions of acetone SOA under different seed particle conditions.

2. Experimental

2.1. Material, monitoring and analysis

The experiments of acetone photolysis were conducted in a 1.3 m^3 Teflon bag in a $2 \times 1.2 \times 2\text{-m}$ self-made indoor smog chamber equipped with 42 lamps (F40BLB, GE; UVA-340, Q-Lab, USA). The thickness of Teflon bag reactor was 0.08 mm with the minimum surface/volume ratio of 5.8 m^{-1} . The NO_2 photolysis rate constant was used to reflect the light intensity in the reactor, which was measured to be 0.44 min^{-1} . Experimental details have been provided in our previous work (Jia and Xu, 2014, 2016; Ge et al., 2016, 2017a, 2017b), so a brief summary is given here.

The background gas was zero air. The zero air was produced by Zero Air Supply (Model 111 and Model 1150, Thermo Scientific, USA). Two additional hydrocarbon traps were used to get further purified zero air. The chamber was filled with dry zero air, and then followed by humid vapor by bubbling zero air through ultrapure water (Milli Q, 18M Ω , Millipore Ltd., USA) to the desired RH. A hygrometer (Model 645, Testo AG, Germany) was used to measure the RH in the experiments.

Acetone was from J&K Scientific Ltd. with the purity of 99.8%, which was introduced into the reactor by a syringe slowly and allowed to vaporize prior to initiating the reaction. The NO_2 gas (500 ppm) with N_2 as the background gas was from Beijing Huayuan Gas Chemical industry Co. Ltd., China, which was injected into the reactor directly by a syringe. To increase the concentration of OH radicals during the experiments, 30% hydrogen peroxide solution was added into the reactor, which was estimated to be about 10 ppm in the chamber according to the injection volume. Seed particles were prepared with a constant-output atomizer (Model 3076, TSI, USA) from 150 mg L^{-1} aqueous solutions of different salts. The purity of NaCl, NaNO_3 and $(\text{NH}_4)_2\text{SO}_4$ was 99.99%, 99.99% and 99.5%, respectively. After all the reactant gases and seeds were added into the chamber, the reactor would be maintained without any activities for one hour to make the reactants completely mixed before turning on the black-lights to initiate the reaction.

The concentration of acetone was measured by GC/MS (7890A/5975C, Agilent, USA). The concentrations of NO_x and O_3 were measured by Model 42C- NO_x analyzer and Model 49C- O_3 analyzer. The particle number and mass concentrations were measured by the Scanning Mobility Particle Sizer (SMPS, Model 3936, TSI), which was consisted of a differential mobility analyzer (DMA, Model 3081, TSI) and a condensation particle counter (CPC, Model 3776, TSI). The sampling flow rate of SMPS was 0.3 L min^{-1} and the sheath flow rate was 3 L min^{-1} . The density of solid NaCl, NaNO_3 and $(\text{NH}_4)_2\text{SO}_4$ at 5% RH is 2.2 g cm^{-3} , 2.3 g cm^{-3} and 1.8 g cm^{-3} , respectively; and their density at the liquid phase at ~80% RH is 1.2 g cm^{-3} , 1.3 g cm^{-3} and 1.2 g cm^{-3} , respectively. It should be noted that the background air is clean enough that the concentration of particles was below the instrumental limitations of the SMPS.

The wall loss constants of NO_2 and O_3 were measured under low and high RH conditions, and the results were similar with our previous work (Ge et al., 2017a). The wall loss constant of acetone at 5% RH and 87% RH was measured to be $3.3 \times 10^{-6} \text{ s}^{-1}$ and $5.0 \times 10^{-6} \text{ s}^{-1}$, respectively. The wall loss constant of particles was measured to be $3.0 \times 10^{-5} \text{ s}^{-1}$ under 5% RH and $6.0 \times 10^{-5} \text{ s}^{-1}$ under 90% RH conditions. The static electricity of Teflon bag was removed before the start of all the experiments (Ge et al., 2017a, 2017b).

At the end of the experiments that were conducted for 3 h, the particles, including the seed particles and SOA formed in the seed droplets, were collected on a ZnSe disk with a Dekati low-pressure

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