Dependence of Cloud Condensation Nuclei Activity of Secondary Organic Aerosol (SOA) on Particle Size

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Secondary organic aerosol (SOA) contributes significantly to the cloud condensation nuclei (CCN) in the atmosphere. The CCN activity of SOA is usually regarded as an ensemble property of the whole aerosol population, whereas aerosol consists of particles of different sizes, which may have different chemical composition and thus varying CCN activity.

In this study, we investigated the size resolved CCN activity of SOA formed in the SAPHIR chamber from various precursors including biogenic volatile organic compounds (VOCs) and anthropogenic VOCs: α-pinene, monoterpene mixture, boreal tree emissions, toluene, xylene, and biogenic-anthropogenic VOC mixtures. The CCN activity of SOA parameterized as κ was found to depend on supersaturation corresponding to particle size. κ decreases with increasing size, indicating smaller particles contain more hygroscopic compounds.

AMS analysis shows that the oxidation level represented by f_{44} (fractional contribution of m/z 44 to total organics signal) decreases significantly and f_{43} (fractional contribution of m/z 43 to total organics signal) increases slightly with increasing particle size. This indicates that smaller particles have higher oxidation level and contain more oxygenated compounds. Since oxidation level (represented by f_{44} or O/C ratio) correlates with CCN activity here as well as in many other previous studies (Massoli et al., 2010; Lambe et al., 2011; Alfarra et al., 2013), the size dependence of CCN activity can be attributed to size dependence of chemical composition: smaller particles have higher oxidation level. The dependence of CCN activity and particle composition on particle size is likely due to the Kelvin effect resulting in more volatile and less oxygenated compounds condensing mainly on larger particles and/or heterogeneous oxidation leading to more oxygenated compounds on smaller particles because of their higher surface to volume ratio.

Other reasons which may contribute to the size dependence of CCN activity including the inherent change of κ with solute concentration and co-condensation of organic vapor during the activation are also discussed.

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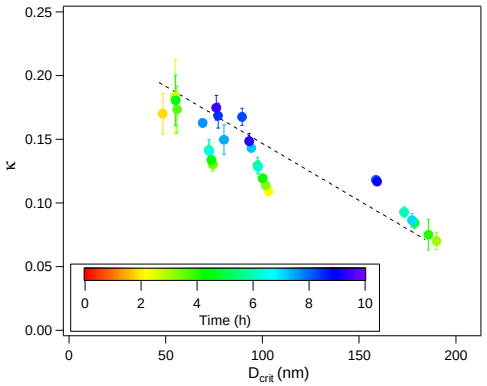

Figure 1. CCN activity of SOA (κ) from α-pinene photooxidation as function of critical diameter. The color indicates the time since the beginning of the reaction. The dashed line is used to guide the eyes.

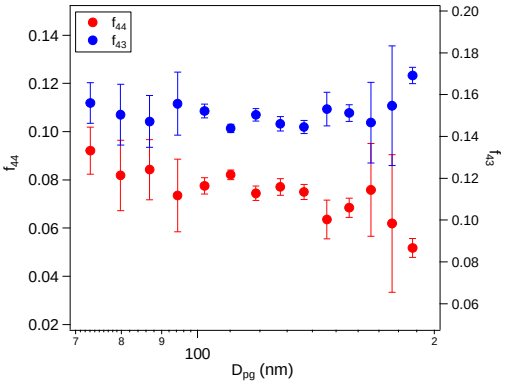

Figure 2. Chemical composition represented by f_{44} and f_{43} of SOA from α-pinene photooxidation as a function of particle size.

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