



## Cloud forming potential of secondary organic aerosol under near atmospheric conditions

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[1] Cloud droplets form by nucleation on atmospheric aerosol particles. Populations of such particles invariably contain organic material, a major source of which is thought to be condensation of photo-oxidation products of biogenic volatile organic compounds (VOCs). We demonstrate that smog chamber studies of the formation of such biogenic secondary organic aerosol (SOA) formed during photo-oxidation must be conducted at near atmospheric concentrations to yield atmospherically representative particle composition, hygroscopicity and cloud-forming potential. Under these conditions, the hygroscopicity measured at 95% relative humidity can be used reliably to predict the CCN activity of the SOA particles by assuming droplet surface tension of pure water. We also show that the supersaturation required to activate a given size of particle decreases with age. **Citation:** Duplissy, J., et al. (2008), Cloud forming potential of secondary organic aerosol under near atmospheric conditions, *Geophys. Res. Lett.*, 35, L03818, doi:10.1029/2007GL031075.

[2] Aerosol particles partially counteract the warming effect of greenhouse gases by modifying cloud radiative properties. Depending on their size and chemical composition, natural and anthropogenic aerosol particles may act as cloud condensation nuclei (CCN) and thereby change the cloud radiative properties and lifetime. The magnitude of this effect is the single biggest unknown in modeling radiative forcing (<http://www.ipcc.ch>). Atmospheric aerosols often consist of (internal) mixtures of organic and inorganic substances. The cloud forming potential (i.e. the hygroscopic properties) of the inorganic constituents are generally well known, whereas not much is known on the hygroscopic properties associated with the organic matter, which typically contributes 20–50% to the mass of fine aerosol particles [Kanakidou et al., 2005]. Secondary organic aerosol (SOA), formed by photo-oxidation of volatile organic compounds (VOCs), is a major contribution to this organic fraction [Robinson et al., 2007]. In this study, we conducted smog chamber experiments on the SOA's hygroscopic properties and ability to act as CCN, using precursor

concentrations approaching atmospheric levels. Hygroscopicity measurements in sub- and supersaturated conditions were compared in order to investigate potential surface tension reduction effects of SOA solution droplets. The surface tension may be important for models inferring the CCN potential from measured hygroscopic properties at sub-saturated conditions. It has been shown that organic material collected from Po Valley fog samples may lower the surface tension in bulk solution and it was inferred that they may similarly affect surface tension in growing cloud droplets [Facchini et al., 1999].

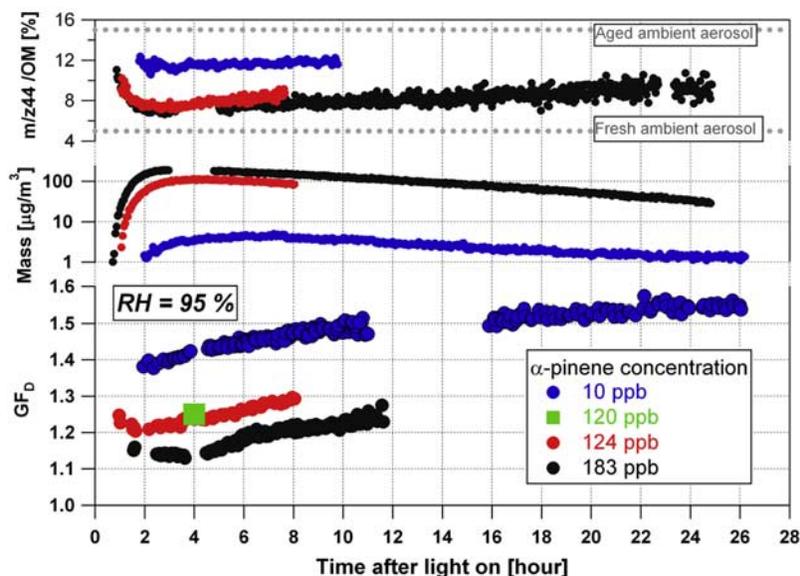
[3] Experiments were performed at the PSI chamber [Paulsen et al., 2005] using varying initial concentrations of 183 to 10 ppbv  $\alpha$ -pinene, a frequently used aerosol precursor representative of biogenic emissions (comparable results were found when using the anthropogenic organic aerosol precursor 1,3,5-trimethylbenzene). The chemical nature of the SOA particles was investigated using the Aerodyne aerosol mass spectrometer (AMS) [Jayne et al., 2000] (see Text S1 of the auxiliary material).<sup>1</sup> The signal ratio of the mass fragment ( $m/z$ ) 44 to total organic was used to gauge broad chemical functionality. A higher ratio represents a higher degree of oxygenation, and this ratio in atmospheric organic aerosols has been shown to increase with increasing photochemical age [McFiggans et al., 2005]. It can be seen from Figure 1 that the  $m/z$  44 to total organic signal ratio approaches the range observed for aged organics in the atmosphere - values between 15 and 20 % are typical for rural and remote locations [Alfarra et al., 2006] - as the  $\alpha$ -pinene concentration is reduced towards an initial chamber mixing ratio of 10 ppbv. The precursor concentration also has a pronounced effect on the physical properties of the SOA particles. A hygroscopicity tandem differential mobility analyzer (HTDMA, see SI) [Baltensperger et al., 2005] was used to determine the diameter growth factor,  $GF_D$ , at 95% relative humidity (RH) compared to particle dry size. Figure 1 (bottom), presents a systematic trend of decreasing  $GF_D$  with increasing  $\alpha$ -pinene concentration. At low precursor concentrations, both  $GF_D$  and  $m/z$  44 to total organic ratio show a continuous increase with illumination time. AMS  $m/z$  44 corresponds mostly to the  $\text{CO}_2^+$  fragment. It has been shown to be formed by decarboxylation of the volatilizing organic compound on the vaporizer in the instrument and is mainly produced by dicarboxylic and multifunctional acids [Alfarra, 2004]. The water uptake of an organic aerosol particle in the presence of a given water vapor loading is dependent on polarity [Topping et al.,

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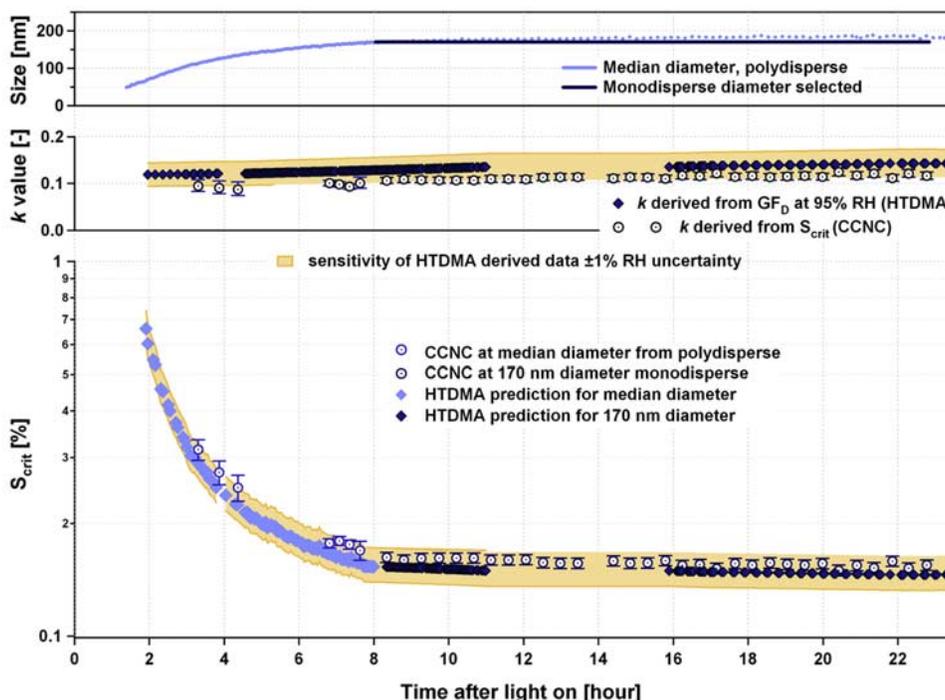
**Figure 1.** Dependence of mass concentration, degree of oxidation, and hygroscopic growth factor of SOA on initial precursor concentration for photo-oxidation experiments of  $\alpha$ -pinene and  $\text{NO}_x$ . (top) The ratio of  $m/z$  44 to total organic mass (OM) as determined by an Aerodyne AMS including values representative of ambient organic aerosols. (middle) Mass of SOA produced during the experiment as measured with the SMPS assuming a density of  $1.3 \text{ g/cm}^3$ . Note the ordinate scale is logarithmic. (bottom) Hygroscopic growth factor  $GF_D$ , defined as the diameter at 95% RH divided by the dry diameter,  $D_0$ , as measured by the HTDMA.  $GF_D$  values originate from different  $D_0$ , but they are all shown with equal symbols due to a lack of size dependence at any time. The value for 120 ppb (green square) relates to *Varutbangkul et al.* [2006], who reported a  $GF_D(85\%) = 1.09$ , when 108 ppbv of  $\alpha$ -pinene had reacted and the SOA mass peak was reached. This corresponds to a  $GF_D(95\%) \approx 1.25$ .

2005]. We therefore contend that the increase in equilibrium water content results from an increase in the oxygenation of components in the SOA with the attendant increase in the SOA average polarity. At higher precursor concentrations, both  $GF_D$  and  $m/z$  44 to total organic ratio show a dip at around 2 to 4 hours of illumination. We hypothesize that this results from a higher degree of oxygenation of the species taking part in particle nucleation and initial growth than of all higher vapor pressure species that partition into the particle phase when the concentrations of gaseous reaction products and SOA mass are highest.

[4] Figure 1 (middle) shows that higher precursor concentration increases the total aerosol mass concentration. The yield (SOA mass produced per reacted organic precursor mass unit) [Dommen *et al.*, 2006; Odum *et al.*, 1996] is also higher as the SOA mass increases by a factor of  $\sim 40$  while the reacted precursor mass increases only by a factor of 24 (at the maximum aerosol mass). The reaction yields of gaseous oxidation products depend only weakly on the initial VOC concentration but more strongly on the initial VOC to  $\text{NO}_x$  ratio [Kroll *et al.*, 2006]. Therefore, the increase in the SOA yield is further evidence for the partitioning of more volatile, less oxygenated compounds towards the particle phase, as shown by the decrease of  $GF_D$  and the ratio  $m/z$  44 to total organic with increasing precursor concentration. Altogether, the results shown in Figure 1 make it clear that the drive to improve process description by increasing the formed aerosol mass makes the experiments less representative of the atmosphere.

[5] The activation of the SOA particles in supersaturated water vapor was investigated at an atmospherically reasonable range of water supersaturations using a cloud condensation nucleus (CCN) counter (DMT, see SI). A particle may form a cloud droplet above its critical supersaturation,  $S_{crit}$ , experiencing unlimited growth as long as the water vapor supply is not depleted to levels below this value. Figure 2 shows measurements of the size evolution of the SOA particles throughout a 24-hour illuminated experiment (Figure 2, top), alongside their corresponding CCN behavior (Figure 2, bottom). During the period of rapid transient growth in the first 8 hours of the experiment, the CCN counter measured the full polydisperse number size distribution of SOA particles (with geometric standard deviation  $\sigma_g$  decreasing from 1.70 to 1.35 during the experiment). A substantial decrease of the critical supersaturation was seen, corresponding to the increase in median particle diameter. From hour 8 to 23, the CCN counter was used to track the evolution of the critical supersaturation for a monodisperse size cut ( $\sigma_g \sim 1.027$ ) of 170-nm diameter particles. The critical supersaturation decreased slowly from 0.16 % to 0.15 %, which is clearly beyond instrumental error (dark blue circles of Figure 2, also Figure S3). The pronounced decrease of  $S_{crit}$  during the first 8 hours can be explained by the increasing particle size, whereas the small decreasing trend thereafter must be solely caused by changing hygroscopicity and/or solution surface tension (see also Figure S5).

[6] Recently the semi-empirical ‘ $k$ -Köhler theory’ has been introduced [Petters and Kreidenweis, 2007, and refer-

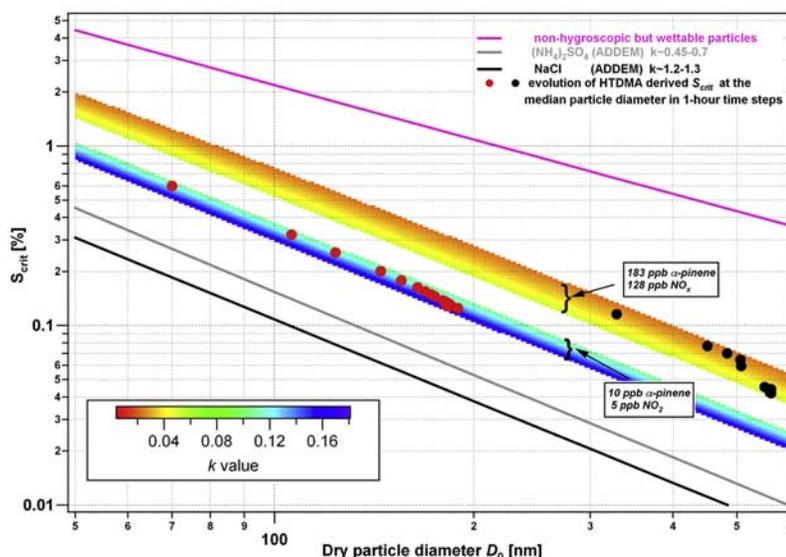


**Figure 2.** (bottom) Critical supersaturation  $S_{crit}$  measured with the CCN counter and predicted from HTDMA measurements of SOA produced by photo-oxidation of 10 ppbv  $\alpha$ -pinene in the presence of 3.8 ppbv  $\text{NO}_x$ . (top) The  $S_{crit}$  shown correspond to the median diameter of the polydisperse aerosol at times <8 h (light blue color) and to the monodisperse diameter selected with a Differential Mobility Analyzer (DMA) at times >8 h (dark blue color). The change of  $S_{crit}$  at  $t < 8$  h is mostly driven by change in size, whereas the slight trend (see also Figure S3) at  $t > 8$  h is solely due to the change in SOA hygroscopic properties. (middle) (Size independent)  $k$  values, derived from both HTDMA  $GF_D$  and CCN  $S_{crit}$ , again illustrating the change in SOA hygroscopic properties. The sensitivity of HTDMA derived data to an RH error of  $\pm 1\%$  at 95% RH is indicated by the yellow shaded areas. CCN data until 3 hours after lights on are not shown because of lack of suitable calibration data above  $S = 0.4\%$ .

ences therein], in order to predict critical supersaturations based on  $GF_D$  measurements from an HTDMA at water subsaturated conditions or vice versa.  $k$  is the only free parameter of the ‘ $k$ -Köhler theory’, capturing all compound properties as well as the degree of dissociation and deviations from ideal behavior as a function of water activity, thus fully determining the hygroscopic growth. Here  $S_{crit}$ -derived  $k$  values (as detailed in the supporting online text) have been used to distinguish trends in hygroscopicity and surface tension from the pure size effects on measured  $S_{crit}$  (Figure 2, middle). From hour 3 onwards the  $S_{crit}$ -derived  $k$  values show a weak but consistently increasing trend in time (from 0.09 to 0.12), which indicates increasing particle hygroscopicity or potentially decreasing solution surface tension. Hygroscopicity effects will be at least partially responsible as there is a corresponding increase in HTDMA  $GF_D$ . This is in contrast to the trend of decreasing CCN activity reported by VanReken *et al.* [2005] for ozonolysis of  $\alpha$ -pinene.

[7] For a number of specific inorganic and organic compounds, Petters and Kreidenweis [2007] have shown that  $S_{crit}$ -derived  $k$  values typically agree within uncertainty with  $GF_D$ -derived  $k$  values, indicating that the ‘ $k$ -Köhler theory’ is applicable for many compounds. Furthermore, they have shown that the  $k$  value of a mixed particle of known composition can also be computed from the constituent’s  $k$  values with a simple formula equivalent to the

Zdanovskii-Stokes-Robinson (ZSR) mixing rule [Stokes and Robinson, 1966] for water uptake of mixed solutions. Thus the ‘ $k$ -Köhler theory’ is an efficient tool for modeling studies to investigate the sensitivity of CCN activity and cloud properties to the composition of atmospheric particles. Our data show that the hygroscopic growth and CCN activity of  $\alpha$ -pinene and 1,3,5-TMB SOA produced at atmospherically relevant precursor concentrations also obey the ‘ $k$ -Köhler theory’. Agreement within experimental uncertainty can be seen between  $GF_D$ -derived and  $S_{crit}$ -derived  $k$  values (Figures 2, middle, and S5, middle) as well as between  $GF_D$ -derived and directly measured  $S_{crit}$  (Figures 2, bottom, and S5, bottom). For the HTDMA  $GF_D$ -derived predictions, the surface tension was assumed to be that of pure water and  $k$  was assumed to be constant for  $\text{RH} > 95\%$ . By measurement of growth factors across a wide range of RH (Figure S2), it was found that the  $k$  value of  $\alpha$ -pinene SOA shows little variation between 70% and 95% RH. There is no reason for this to be generally expected, but we can conclude from it that the  $k$  value at  $\sim 100\%$  RH is likely to deviate less than 25% from the  $k$  value measured at 95% RH. Smooth growth approximately along  $k$ -Köhler curves with constant  $k$  indicates an absence of pronounced solubility limitations. As predictions of  $S_{crit}$  are only moderately sensitive to  $k$  (Figure S4), the surface tension becomes the only highly sensitive parameter in the predictions. We can conclude from the above reconciliation of HTDMA and



**Figure 3.** Sensitivity of the calculated critical supersaturation  $S_{crit}$  to the  $k$ -value and particle dry size, assuming a surface tension of pure water at  $T = 293$  K. Theoretical lines for pure NaCl,  $(\text{NH}_4)_2\text{SO}_4$ , and non-hygroscopic, but wettable particles are also shown. Ranges are given for  $k$ -values of  $(\text{NH}_4)_2\text{SO}_4$  and NaCl as the  $k$ -values depend on the dry size of the particles. Red and black circles indicate the evolution in 1-hour time steps of the measured median particle diameter and the corresponding  $S_{crit}$  derived from HTDMA growth factors assuming RH-independent  $k$  (the first points start after 1.5 and 2 hours of lights on for the 183 ppbv and 10 ppbv  $\alpha$ -pinene experiments, respectively). The upper and lower colored bands indicate the hygroscopicity range covered during the course of the 183-ppbv and 10-ppbv  $\alpha$ -pinene experiments, respectively. Each color of the rainbow shows the dependence of  $S_{crit}$  on particle size whereas at a constant particle size the color change describes the dependence of  $S_{crit}$  on the  $k$ -value. In the low concentration case only  $\text{NO}_2$  was added to speed up the initial reactions.

CCNC measurements that the  $\alpha$ -pinene SOA behavior can be described with ‘ $k$ -Köhler theory’ and that  $\alpha$ -pinene SOA is not likely to alter the surface tension of a solution droplet more than 10% (Figure S4). These appear to be general features of SOA formed by photo-oxidation, as equally good agreement between the HTDMA and CCN derived  $k$  values was found for the photo-oxidation of 1,3,5-TMB in the presence of  $\text{NO}_x$  (Figure S5). SOA was not covered in the study by *Petters and Kreidenweis* [2007]. However, *Prenni et al.* [2007] reported significant disagreement between HTDMA and CCN derived  $k$  values for SOA produced by ozonolysis of  $\alpha$ -pinene and other precursors. They found CCN activation behavior similar to the results in our study, whereas they observed substantially lower hygroscopic growth factors. This discrepancy at subsaturated conditions may be due to different systems investigated (ozonolysis versus photo-oxidation of  $\alpha$ -pinene), or higher SOA mass loadings in the study by *Prenni et al.* [2007], but clearly further work is needed to clarify this issue.

[8] *Ervens et al.* [2005] showed that surface-active compounds have the greatest potential to increase the predicted cloud number concentration and thus radiative forcing in cloud simulations, when compared with simplified simulations assuming the properties of ammonium sulfate for all particles. The presence of surface active compounds has been reported in aerosol and cloud water samples [*Dinar et al.*, 2006; *Facchini et al.*, 1999]. In order to study the possibility that the organics are surface active, we tried to reproduce the observed  $\text{GF}_D$  as a function of RH and  $S_{crit}$  using a model organic substance with adjustable molecular volume, activity coefficients, and surface tension (see Text S1

for details). In contrast to the bulk surface active behavior of organic compounds extracted from atmospheric aerosol samples, we conclude from our study that the surface tension reduction at critical supersaturation is minimal, which also contributes to the success of the  $k$ -Köhler theory for the SOA generated in the oxidation of  $\alpha$ -pinene or 1,3,5-TMB as simulated in the chamber. This is in agreement with an investigation of humic-like organics extracted from ambient particles, indicating that these compounds hardly reduce the droplets surface tension at its critical size for activation despite their surface active behavior in concentrated bulk solutions [*Wex et al.*, 2007]. Thus, surface active compounds must stem from other, possibly primary sources [*Mochida et al.*, 2006]. Another topic of recent debate is the potential of organic compounds to form surface coatings hindering water uptake, thus reducing the growth and CCN activity of otherwise hygroscopic material [*McFiggans et al.*, 2006]. The SOA formed through photo-oxidation of  $\alpha$ -pinene or 1,3,5-TMB is unlikely to exhibit such behavior, as SOA formed in such systems is readily water-soluble (Figure S2) and obeys standard equilibrium Köhler theory (Figure 2), which would not be the case if the equilibrium was not reached due to kinetic limitation of the water uptake on the timescale of the measurement.

[9] Figure 3 quantifies the influence of size and composition on CCN activity for different aerosol particles including the range covered by the SOA in this study. The upper and lower colored bands indicate the hygroscopicity range covered during the course of the 183-ppbv and 10-ppbv  $\alpha$ -pinene experiments, respectively. The top and bottom borders of each band correspond to the minimum and

maximum hygroscopicity, respectively, observed at the beginning and the end of each experiment, while the red and black circles illustrate the actual growth of median particle size and corresponding evolution of  $S_{crit}$  in 1-hour steps. The clear separation of the bands for the different precursor concentrations shows the magnitude of the effect of the chemical composition -  $S_{crit}$  follows a different function of particle diameter in each case. The smaller effect of illumination time is shown by the path of the circles within the bands. Under all conditions, the SOA is clearly more CCN active than non-hygroscopic but wettable particles, but less CCN active than inorganic salts such as ammonium sulfate or NaCl. It has recently been proposed that the effect of particle size swamps the composition effects on CCN activity of atmospheric aerosols [Dusek et al., 2006], but it is seen here that particle composition in general and simulating SOA with inappropriate precursor concentration also has a considerable effect on cut-off diameters at a given supersaturation. For example, at 0.5% supersaturation, the critical diameter  $D_{crit}$  is 75 nm and 121 nm for the 10-ppbv and 183-ppbv  $\alpha$ -pinene experiments respectively (compared to 433 nm for non-hygroscopic but wettable particles and 46.9 nm for ammonium sulfate).

[10] A comprehensive thermodynamic description of the relationship between particle composition, hygroscopic growth and CCN activity requires substantial computational resources and properties of every compound such as density, volume of mixing, molecular mass, dissociation constants, interaction parameters, etc. Semi-empirical ‘ $k$ -Köhler theory’ is an efficient alternative, which has been shown to be applicable for the SOA produced in this study, thus providing a tool for future studies looking into the relative importance of particle size and composition effects for CCN activity and cloud properties.

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## References

- Alfarra, M. R. (2004), Insights into atmospheric organic aerosols using an aerosol mass spectrometer, Ph.D. thesis, Inst. of Sci. and Technol., Univ. of Manchester, Manchester, U. K.
- Alfarra, M. R., et al. (2006), A mass spectrometric study of secondary organic aerosols formed from the photooxidation of anthropogenic and biogenic precursors in a reaction chamber, *Atmos. Chem. Phys.*, *6*, 5279–5293.
- Baltensperger, U., et al. (2005), Secondary organic aerosols from anthropogenic and biogenic precursors, *Faraday Discuss.*, *130*, 265–278.
- Dinar, E., et al. (2006), Cloud condensation nuclei properties of model and atmospheric HULIS, *Atmos. Chem. Phys.*, *6*, 2465–2481.
- Dommen, J., A. Metzger, J. Duplissy, M. Kalberer, M. R. Alfarra, A. Gascho, E. Weingartner, A. S. H. Prevot, B. Verheggen, and U. Baltensperger (2006), Laboratory observation of oligomers in the aerosol from isoprene/NO<sub>x</sub> photooxidation, *Geophys. Res. Lett.*, *33*, L13805, doi:10.1029/2006GL026523.
- Dusek, U., et al. (2006), Size matters more than chemistry for cloud-nucleating ability of aerosol particles, *Science*, *312*(5778), 1375–1378.
- Ervens, B., G. Feingold, and S. M. Kreidenweis (2005), Influence of water-soluble organic carbon on cloud drop number concentration, *J. Geophys. Res.*, *110*, D18211, doi:10.1029/2004JD005634.
- Facchini, M. C., et al. (1999), Cloud albedo enhancement by surface-active organic solutes in growing droplets, *Nature*, *401*(6750), 257–259.
- Jayne, J. T., et al. (2000), Development of an aerosol mass spectrometer for size and composition analysis of submicron particles, *Aerosol Sci. Technol.*, *33*(1–2), 49–70.
- Kanakidou, M., et al. (2005), Organic aerosol and global climate modelling: A review, *Atmos. Chem. Phys.*, *5*, 1053–1123.
- Kroll, J. H., et al. (2006), Secondary organic aerosol formation from isoprene photooxidation, *Environ. Sci. Technol.*, *40*(6), 1869–1877.
- McFiggans, G., et al. (2005), Simplification of the representation of the organic component of atmospheric particulates, *Faraday Discuss.*, *130*, 341–362.
- McFiggans, G., et al. (2006), The effect of physical and chemical aerosol properties on warm cloud droplet activation, *Atmos. Chem. Phys.*, *6*, 2593–2649.
- Mochida, M., M. Kuwata, T. Miyakawa, N. Takegawa, K. Kawamura, and Y. Kondo (2006), Relationship between hygroscopicity and cloud condensation nuclei activity for urban aerosols in Tokyo, *J. Geophys. Res.*, *111*, D23204, doi:10.1029/2005JD006980.
- Odum, J. R., et al. (1996), Gas/particle partitioning and secondary organic aerosol yields, *Environ. Sci. Technol.*, *30*(8), 2580–2585.
- Paulsen, D., et al. (2005), Secondary organic aerosol formation by irradiation of 1,3,5-trimethylbenzene-NO<sub>x</sub>-H<sub>2</sub>O in a new reaction chamber for atmospheric chemistry and physics, *Environ. Sci. Technol.*, *39*(8), 2668–2678.
- Petters, M. D., and S. M. Kreidenweis (2007), A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, *7*, 1961–1971.
- Prenni, A., M. D. Petters, S. M. Kreidenweis, P. J. DeMott, and P. J. Ziemann (2007), Cloud droplet activation of secondary organic aerosol, *J. Geophys. Res.*, *112*, D10223, doi:10.1029/2006JD007963.
- Robinson, A. L., et al. (2007), Rethinking organic aerosols: Semivolatile emissions and photochemical aging, *Science*, *315*(5816), 1259–1262.
- Stokes, R. H., and R. A. Robinson (1966), Interactions in aqueous nonelectrolyte solutions: part I: Solute-solvent equilibria, *J. Phys. Chem.*, *70*(7), 2126–2131.
- Topping, D. O., et al. (2005), A curved multi-component aerosol hygroscopicity model framework. Part 2: Including organic compounds, *Atmos. Chem. Phys.*, *5*, 1223–1242.
- VanReken, T. M., N. L. Ng, R. C. Flagan, and J. H. Seinfeld (2005), Cloud condensation nucleus activation properties of biogenic secondary organic aerosol, *J. Geophys. Res.*, *110*, D07206, doi:10.1029/2004JD005465.
- Varutbangkul, V., et al. (2006), Hygroscopicity of secondary organic aerosols formed by oxidation of cycloalkenes, monoterpenes, sesquiterpenes, and related compounds, *Atmos. Chem. Phys.*, *6*, 2367–2388.
- Wex, H., T. Hennig, I. Salma, R. Ocskay, A. Kiselev, S. Henning, A. Massling, A. Wiedensohler, and F. Stratmann (2007), Hygroscopic growth and measured and modeled critical super-saturations of an atmospheric HULIS sample, *Geophys. Res. Lett.*, *34*, L02818, doi:10.1029/2006GL028260.
- M. R. Alfarra, U. Baltensperger, J. Dommen, J. Duplissy, M. Gysel, A. Metzger, A. S. H. Prevot, and E. Weingartner, Laboratory of Atmospheric Chemistry, Paul Scherrer Institut, CH-5232 Villigen, Switzerland. (martin.gysel@psi.ch)
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