Adsorptive uptake of water by semisolid secondary organic aerosols

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Key Points:

- The low solubility of slightly oxygenated SOA limits its water uptake at RH < 100%
- Slightly soluble SOA takes up water mainly by adsorption at RH < 100%
- Discrepancy in k for RH < 100% and RH > 100% produces uncertainty in modeled ARI

Supporting Information:

- Text S1, Figures S1–S9, and Table S1

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Adsorptive uptake of water by semisolid secondary organic aerosols

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Abstract Aerosol climate effects are intimately tied to interactions with water. Here we combine hygroscopicity measurements with direct observations about the phase of secondary organic aerosol (SOA) particles to show that water uptake by slightly oxygenated SOA is an adsorption-dominated process under subsaturated conditions, where low solubility inhibits water uptake until the humidity is high enough for dissolution to occur. This reconciles reported discrepancies in previous hygroscopicity closure studies. We demonstrate that the difference in SOA hygroscopic behavior in subsaturated and supersaturated conditions can lead to an effect up to about 30% in the direct aerosol forcing—highlighting the need to implement correct descriptions of these processes in atmospheric models. Obtaining closure across the water saturation point is therefore a critical issue for accurate climate modeling.

Aerosols affect climate in two ways: aerosol-radiation interactions (ARI) (the direct effect) and aerosol-cloud interactions (ACI) (the indirect effect). While the indirect effect has received far more attention recently, the current Intergovernmental Panel on Climate Change Assessment Report 5 suggests nearly equal magnitudes and uncertainties for both effects (roughly $0.5 \pm 0.5\,\mathrm{W\,m^{-2}}$ cooling in each case) [*Boucher et al.*, 2013]. Secondary Organic Aerosol (SOA) often dominates aerosol mass in remote areas [*Jimenez et al.*, 2009; *Hallquist et al.*, 2009] and consists of a highly complex mixture of sometimes very viscous and sparingly soluble compounds [*Petters et al.*, 2009; *Renbaum-Wolff et al.*, 2013; *Virtanen et al.*, 2010]. It is very likely that SOA was even more dominant in preindustrial times when water-soluble sulfates and inorganic nitrates were much less abundant. Understanding the direct and indirect effects of SOA is critical to narrowing the uncertainties in aerosol-climate interactions, which remain stubbornly the largest sources of uncertainty in climate forcing.

ARI is due to light scattering and absorption by particles: Scattering depends on particle size, which increases dramatically when particles swell with water on a humid day or as they approach cloud base. ACI is due to light scattering by cloud droplets: Cloud-droplet number is controlled by the number of particles that activate as air rises through the condensation level and relative humidity (RH) exceeds 100%. Both subsaturated swelling and supersaturated activation are controlled by particle hygroscopicity, but the thermodynamic regimes and measurement methods are very different.

Here we focus on the critical transition region linking these two effects, with important implications for pristine environments dominated by organic particulate matter and especially highly uncertain preindustrial conditions. That transition is the swelling of particles via water uptake as RH rises toward 100% and the subsequent growth to cloud droplets of a subset of those particles (the cloud condensation nuclei (CCN)). Especially below 100% RH, relevant to ARI, highly nonideal behavior by different SOA types strongly influences particle swelling (hygroscopic growth), with substantial implications for climate forcing.

If solutions were ideal (with water activity equal to its mole fraction in the particle phase and also its vapor saturation ratio), subsaturated and supersaturated water uptake would be fairly straightforward, though the unknown composition of especially SOA complicates matters. The dependence of hygroscopicity on particle composition can be represented with a single parameter, κ, which is commonly used in global models to describe the hygroscopic properties of atmospheric aerosol particles. For ideal solutes, κ would remain constant across both the subsaturated and supersaturated regimes [*Petters and Kreidenweis*, 2007]. It is typically calculated from subsaturated hygroscopicity tandem differential mobility analyzer (HTDMA) measurements and supersaturated cloud condensation nuclei counter (CCNc) measurements (i.e., κ_{HGF} and κ_{CCN}; see equations (7) and (8) in the supporting information). However, a number of recent studies conducted on SOA indicate that the two measurements yield different κ values, namely, that $\kappa_{HGF} < \kappa_{CCN}$ [*Duplissy et al.*, 2008; *Hersey et al.*, 2013; *Massoli et al.*, 2010; *Prenni et al.*, 2007]. There are several studies discussing the observed behavior [*Petters et al.*, 2009; *Prenni et al.*, 2007; *Wex et al.*, 2009], but the underlying reasons are unresolved.

1. Results and Discussion

To investigate this issue, we conducted CCNc measurements in parallel with the HTDMA measurements to determine the effects of chemical composition, oxidation level, the phase state, and RH on the associated water uptake properties of biogenic SOA particles formed from isoprene (C_5H_8), α-pinene ($C_{10}H_{16}$), and longifolene ($C_{15}H_{24}$) precursors. We formed pure SOA particles by OH and/or O_3 oxidation of those gas-phase precursors in a flow reactor followed by homogeneous nucleation [*Lambe et al.*, 2011; *Kang et al.*, 2007]. We then simultaneously measured: (i) subsaturated water uptake (hygroscopic growth factor (HGF)) using HTDMA, (ii) supersaturated cloud-droplet formation using a CCNc [*Roberts and Nenes*, 2005; *Lance et al.*, 2006], (iii) particle bounced fraction (BF) using an Aerosol Bounce Instrument (ABI, see Figure S3), and (iv) SOA oxidation state using a compact time of flight aerosol mass spectrometer (c-ToF-AMS) [*Drewnick et al.*, 2005]. We measured the particles hygroscopic growth factor (HGF) as the ratio of wet to dry particle diameter at a controlled RH (see supporting information). ABI measurements are qualitatively related to particle phase (see supporting information); we deduce that the particles with $BF > 0$ are solid or semisolid and that the particles with $BF = 0$ behave mechanically as liquids [*Virtanen et al.*, 2010]. The c-ToF-AMS constrains the SOA oxygen-to-carbon ratio (O:C), a measurable surrogate of the overall oxygenation that ranged from 0.39 to 0.86 (see Figures S1 and S2). Figures 1a and 1b show HGF and BF measurements for the three SOA types at different O:C. The HGF at RH ~90% ranged from 1.07 to 1.37 depending on the SOA type and O:C. Further, the HGF for each SOA type and RH increased with O:C, consistent with previous observations [*Jimenez et al.*, 2009; *Duplissy et al.*, 2008; *Massoli et al.*, 2010]. To relate HGF to phase state, symbols in Figures 1a and 1b are colored by the RH at which BF approached 0. As can be seen, SOA particles that attain $BF = 0$ at $RH > 90\%$ have the smallest HGF values. On the other hand, SOA particles that attain $BF = 0$ at $RH = 61–72\%$ have the highest HGF values.

To investigate the hygroscopic behavior in more detail, we calculated the hygroscopicity parameter κ under subsaturated and supersaturated conditions (Figure 1c). Because κ implicitly assumes a completely soluble phase, which may not be the case, we note that our calculated κ values are instead "effective" κ values (κ_{eff}) that relax this solubility assumption (see supporting information). As can be seen (Figure 1c), for less oxidized, semisolid α-pinene SOA ($O:C = 0.45$) and longifolene SOA ($O:C = 0.39$ to 0.56), $\kappa_{HGF,eff}$ decreases with increasing RH. Furthermore, BF does not reach zero until at $RH > 90\%$ for these particles, implying a solid or semisolid phase at relatively high RH. To compare these observations for SOA to the well-known adsorptive water uptake behavior of solid insoluble particles, we measured the HGFs for SiO_2 particles (white stars in Figures 1a and 1c), which display similar trends (Figure 1c) [*Keskinen et al.*, 2011]. Because SiO_2 particles can take up water only via surface adsorption, this suggests that a similar mechanism occurs for semisolid or sparingly soluble SOA particles, where the apparent hygroscopic growth is due to surface adsorption of water rather than bulk water uptake. As a result, solubility and possibly diffusion limitations in the particle bulk inhibit water uptake until dissolution occurs at very high RH when the water activity is high enough to dissolve even sparingly soluble compounds [*Mikhailov et al.*, 2009]; recently *Lienhard et al.* [2014] observed a similar transition for aqueous citric acid droplets near glass transition humidity.

The adsorption behavior can be quantified in the subsaturated regime using Frenkel-Halsey-Hill (FHH) adsorption theory [*Adamson and Gast*, 1997; *Sorjamaa and Laaksonen*, 2007] (see supporting information). Figure 1c shows that the FHH adsorption model reproduces the decrease in $\kappa_{HGF,eff}$ with increasing RH for

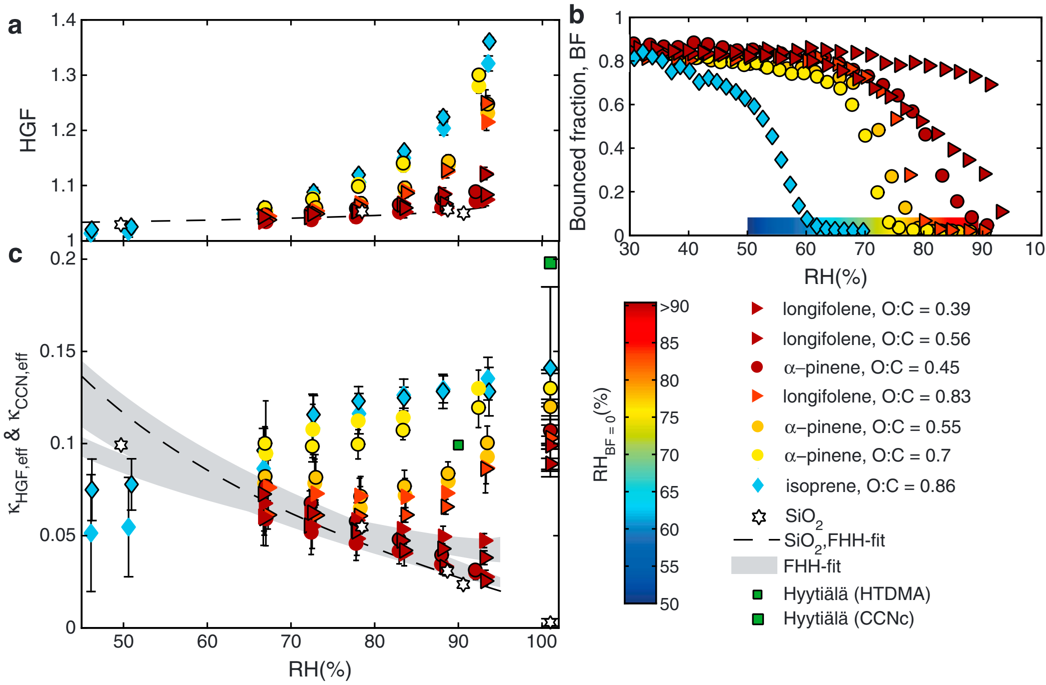

Figure 1. Hygroscopic behavior of laboratory-generated SOA particles described using results from HTDMA, CCNc, and ABI measurements. Spheres represent α-pinene, triangles represent longifolene, and diamonds represent isoprene SOA particles. (a) The measured HGFs as a function of RH. Mobility diameters, d_p, of the SOA particles, 50 nm (without edge color) and 100 nm (black edges), and 50 nm for SiO_2 (white stars) are plotted. (b) The colors of the data points represent the RH values where the bounced fractions falls to zero as shown (i.e., for isoprene the zero point is RH ~ 62%, whereas for α-pinene with lowest O/C the zero point is RH ~ 92%). (c) The hygroscopicity parameters calculated from HTDMA and CCNc data, the gray area represents Frenkel-Halsey-Hill (FHH) isotherms fit to the data (see supporting information), and the green rectangles represent ambient data (d_p = 100 nm) measured in spring 2012 at the SMEAR II station in Hyytiälä, Finland (see supporting information). Error estimates for ambient $\kappa_{HGF,eff}$ and $\kappa_{CCN,eff}$ are ±0.03 and ±0.04, respectively.

SiO_2. A similar trend is observed for low O:C α-pinene and longifolene SOA: the water uptake under low RH conditions is clearly enhanced when compared to higher water activities suggesting that surface adsorption dominates the water uptake at low RH (Figure S5). In addition to surface adsorption of water, small quantities of SOA may also dissolve into the adsorption layer of the particles as RH increases. *Kumar et al.* [2009] presented a model describing insoluble particles having a fraction of soluble material on their surface. This theory combines the FHH adsorption term and a Raoult's law term. However, based on our calculations (Figure S6) 0.5 nm layer of soluble material on 50 nm particle reproduces already significantly different κ behavior compared to the observed indicating that the fraction of the soluble material needs to be minor. In contrast to the less oxygenated, high carbon number SOA, for isoprene SOA and highly oxidized α-pinene SOA (O:C = 0.7), $\kappa_{HGF,eff}$ increases monotonically with increasing humidity. In conjunction with the decrease in BF for these particles in the lower range of RH (61–72%), these observations indicate that isoprene SOA and highly oxidized α-pinene SOA liquefy at lower water activities because they consist of compounds having higher solubility. It is also highly possible that there is a range in solubility of the compounds comprising SOA formed in each experiment. In that case with increasing RH larger and larger fraction of compounds dissolves, driving an increasing $\kappa_{HGF,eff}$ with increasing RH. For α-pinene SOA with O:C = 0.55 and longifolene SOA with O:C = 0.83, an intermediate trend is observed: $\kappa_{HGF,eff}$ decreases with increasing humidity up to RH ≈ 80%. At higher RH, $\kappa_{HGF,eff}$ increases with increasing RH suggesting a transition from an adsorption- to a dissolution-dominated processes.

Comparing the measured κ_{HGF} at RH = 90% and κ_{CCN} values for different SOA systems it appears that differences in κ_{HGF} and κ_{CCN} are smallest for particles that dissolve at lower RH (i.e., isoprene SOA, α-pinene SOA with O:C > 0.7, longifolene SOA with O:C > 0.83) and largest for particles exhibiting adsorption-dominated water uptake over the entire HTDMA measurement range (α-pinene SOA with O:C = 0.45, longifolene SOA with O:C < 0.56). The discrepancy diminishes with increasing oxygenation of

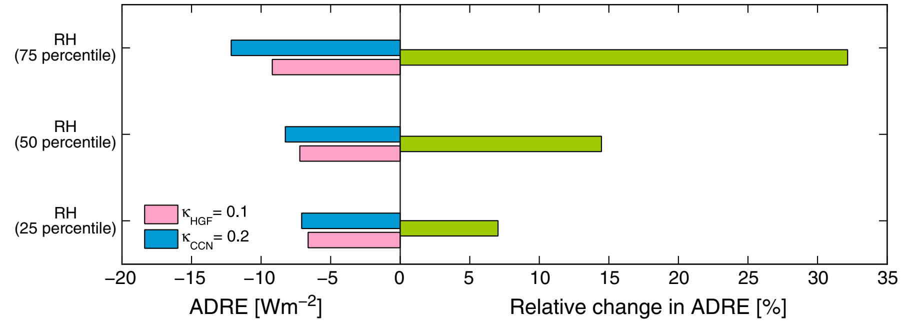

Figure 2. Comparison of simulated Aerosol Direct Radiative Effect (ADRE) predicted from measured hygroscopicity κ values in Hyytiälä forest. (left) ADREs ($W\,m^{-2}$) calculated by the SBDART model [*Ricchiazzi et al.*, 1998] are shown for two different total κ values measured in the campaign ($\kappa_{HGF} = 0.1$ and $\kappa_{CCN} = 0.2$, respectively). (right) Relative change in ADRE (%) between the two simulations calculated by equation (20) in Text S1 is shown (see supporting information).

the particles (see Figure S7). Previous work [*Petters et al.*, 2009] suggests that the observed $\kappa_{HGF} < \kappa_{CCN}$ discrepancy is caused either by a strongly composition-dependent activity coefficient of water or by a mixture of infinitely water-soluble compounds and sparingly soluble compounds that dissolve in solution at RH > 90%. Our present work suggests that $\kappa_{HGF} < \kappa_{CCN}$ because of adsorption-dominated water uptake by amorphous SOA in the subsaturated regime when the SOA is partially oxygenated. The particles evidently contain two phases—an organic core with a quite thin coating of almost pure water.

The low water uptake we observe is presumably due to either diffusion or solubility limitations. Laboratory studies of amorphous sucrose particles show that for highly soluble sucrose particles at room temperature, dissolution of the viscous glassy core proceeds rapidly [*Tong et al.*, 2011]. This suggests that the diffusion of water in the viscous particles may not be the kinetic parameter limiting particle size change once water is able to adsorb to the particle surface and dissolution of the glassy core can proceed toward an equilibrium solution state. To further investigate this possibility, we performed HGF measurements of 50 and 100 nm α-pinene SOA particles (O:C = 0.4) at 90% RH and room temperature using humidification times of 8 s and 20 min (otherwise, the humidification times in HTDMA and ABI was 8 s, see supporting information). Within measurement accuracy, the measured HGF values were the same at both humidification times, suggesting that dissolution under subsaturation conditions is governed by the solubility characteristics of the particles rather than by diffusion limitations. With increasing oxidation level, the fraction of water-soluble compounds at a given RH increases, resulting in increasing κ_{HGF} as a function of RH. The behavior for less oxidized α-pinene SOA and more oxidized longifolene SOA shown in Figure 1c is consistent with this interpretation; these particles show a transition from adsorptive behavior (decreasing κ with increasing RH) to absorptive behavior near 80% RH, suggesting that the solubility limit for these SOA particles is near this water activity.

Finally, we compare our laboratory observations to ambient measurements obtained in a boreal forest environment (Hyytiälä Forestry Field Station, SMEAR II [*Hari and Kulmala*, 2005]) where monoterpene SOA typically comprises a major fraction of the atmospheric aerosol [*Finessi et al.*, 2012; *Raatikainen et al.*, 2010]. Paired ambient $\kappa_{HGF,eff}$ (at RH = 90%) and $\kappa_{CCN,eff}$ measurements obtained for ambient organic-containing particles during spring 2012 at SMEAR II (see supporting information) are shown in Figure 1c. These particles have an organic mass fraction of 0.81 with O:C = 0.49 and ammonium sulfate mass fraction of 0.19. The average $\kappa_{CCN,eff} = 0.195$ value is 95% higher than the corresponding $\kappa_{HGF,eff} = 0.10$ value, which is in the range of our laboratory measurements. Thus, similar inhibited hygroscopic growth is observed for laboratory and ambient biogenic SOA particles of similar source type and oxidation levels. Inhibited hygroscopic growth for particles dominated by anthropogenic SOA has also been reported [*Hersey et al.*, 2013].

The ambiguity about the κ values has important implications for quantifying the climate effects of SOA in atmospheric models. In some models organics are treated as completely water soluble and in other models as water insoluble when calculating CCN activation. This can lead to more than ±0.4 $W\,m^{-2}$ uncertainty in the aerosol indirect forcing between preindustrial and present-day conditions [*Liu and*

Wang, 2010]. Besides affecting modeled ACI, hygroscopic growth (i.e., aerosol water content) has significant implications for the predicted optical properties of aerosol and thus their direct effect on climate. Most atmospheric models use only one κ value, i.e., either κ_{CCN} or κ_{HGF} is used to describe the aerosol interaction with water vapor in both subsaturation and supersaturation regimes. Figure 2 shows how the calculated aerosol direct radiative effect (ADRE, W m^{-2}) changes if κ_{CCN} is employed in the calculation instead of κ_{HGF} for a case study representing the conditions in boreal forest during spring (see Figure S8), where aerosol composition is dominated by SOA (organic mass fraction = 0.81, O:C = 0.49). Here the ADRE is defined as a perturbation in net (downward minus upward) radiant energy by total aerosol on the top of the atmosphere (i.e., an effective radiative forcing due to ARI). The median relative change in the ADRE is of the order of 15% (the quartiles being at about 7% and 32%) thus clearly affecting the estimates of direct radiative effects of the SOA particles (see supporting information).

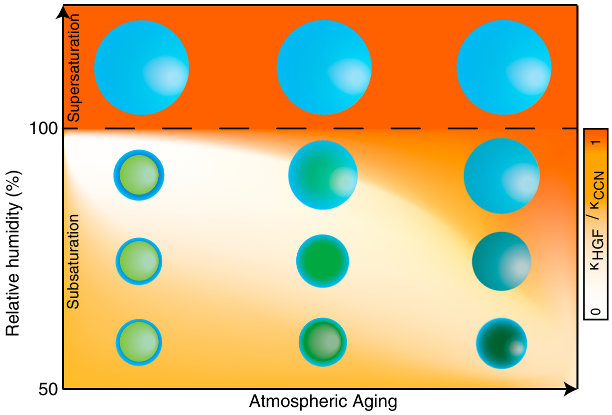

Figure 3. Schematics of the water uptake processes of SOA particles in the atmosphere. Subsaturated swelling (κ_{HGF}) can vary dramatically with minor differences in supersaturated droplet activation (κ_{CCN}); consequently particles can have very different direct and indirect climate effects. The background color scale indicates the ratio of these parameters at given subsaturated and supersaturated conditions, whereas the darkness of the green color in particles denotes their atmospheric age. The contrast is largest when adsorption is the dominant water uptake mechanism even at high RH. This is the case for low O:C SOA particles on the left. With increasing oxidation, i.e., increasing atmospheric age of the particles, the solubility increases and the dissolution RH decreases, decreasing the discrepancy between the κ_{HGF} and κ_{CCN} values.

Our results show that for subsaturated conditions, the water uptake of SOA with relatively low O:C is an adsorption-dominated process. Low solubility hinders the miscibility and hence water uptake until the RH is high enough for dissolution to occur. With increasing oxidation, i.e., increasing atmospheric age of the particles, the solubility increases, the dissolution RH decreases, and the κ_{HGF} approaches the values of κ_{CCN} (see Figure 3). These findings significantly increase our understanding of one of the most relevant atmospheric processes involving organic aerosols. They also reconcile previously described differences between the hygroscopicity of SOA measured in subsaturated and supersaturated conditions. When calculating the ADRE or the CCN activation of SOA to estimate its climate effects, special attention should be paid to κ values used in the calculation to obtain accurate results.

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