

Response to Reviewer 1:

We thank the referee for their comments and suggestions. Our responses to the comments (along with the original comments in italic) are given below

Major comments:

1. This is a well organized and well written paper discussing the relationship of hygroscopicity of the organic component of aerosol particles with the relative level of oxygenation of those particles as defined by the ratio of m/z_{44} to the total organic mass concentration based on AMS measurements. The observations include both chamber data and ambient data. The approaches to the analyses are clearly presented. Overall, I find it to be a strong paper with the exceptions of the apparent motivation and conclusions and as per comment 2 below. The abstract and particularly the conclusions do not describe the results of the paper well and tout the concept of using these data in models as a parameterization for the hygroscopicity of the organic component of the atmospheric aerosol. This concept could be an important application, but it's unclear that models are currently able to adequately predict the mass concentration of the organic aerosol let alone the level of oxygenation. The paper simply and nicely discusses processes and chemistry, and some of that should show up in the conclusions. The most important contribution is the summary of the KappaOrg with f_{44} for a variety of conditions.

2. The authors compare their results with those of Chang et al. (2009):

- The authors acknowledge that their data are for sub-saturated conditions whereas the Chang et al data were for supersaturated conditions. However, they do not attribute any of the differences (primarily slope) to this fundamental difference. For example, could not solute-solute interactions, which the authors state are neglected in their model (P19318, lines 5-6), be of some consequence? The droplets for the sub-saturated conditions will obviously be more concentrated than for the supersaturated conditions. Previous comparisons for chamber SOA have found Kappa estimated from HTDMA to be different than Kappa estimated from CCN (Duplissy et al., 2007; Prenni et al., 2007; Wex et al., 2009). It is important to discuss these discrepancies as well as provide readers with a more complete perspective on the issue. - It appears that the authors refer to the discussion paper of Chang et al. rather than the ACP version (Volume 10, pp. 5047-5064, 2010). In their ACP paper, Chang et al. state that the range for their KappaOrg to O/C relationship is only valid for 0.3 to 0.6. At the bottom of page 19323, you refer to the relationship from Chang et al as $\text{KappaOrg} = 0.30 \text{ O/C}$ whereas in Chang et al. it is given as $\text{KappaOrg} = 0.29 \text{ O/C}$.

We agree, with the reviewers and we remove the data from Chang et al. from figure 7 and table 3, which bring confusion between supersaturated and sub-saturated regime. We have replaced the text from page 19323, line 27 onwards

“In addition, Chang et al. (2009) reported a similar relationship for κ_{org} derived from measurements done at supersaturated conditions ($\kappa_{org} = (0.30 \pm 0.05) \times (\text{O/C})$) which is also shown in Figure 7. This result is in agreement with our TMB data in the range f_{44} 0.06-0.10 and with our field and chamber data around f_{44} 0.10-0.14. Chang et al. (2009) extrapolated their data to $\kappa_{org}=0$, which seems not to be justified for that study. At higher f_{44} (greater than 0.14) the data of Chang et al. (2009) and Raatikainen et al. (2010) tend to be lower than our extrapolations. Further experiments are needed to constrain the relationship above this value. “

with a more comprehensive discussion of previous literature data for sub- and supersaturated RH conditions.:

“In this study we investigate the relationship between the oxidation level of the SOA and its hygroscopic growth factor at subsaturated RH. Chang et al. (2010) reported a similar relationship for K_{org} ($K_{org} = (0.29 \pm 0.05) \times (O/C)$) derived from ambient CCN measurements at supersaturated RH. This slope differs from our results, which can likely be attributed to decreased importance of non-ideal interactions in dilute solutions at the point of CCN activation (Petters et al., 2009). Several laboratory studies reported differences between HTDMA- and CCNC-derived κ_{org} of SOA (e.g. Prenni et al., 2007; Wex et al., 2009; Juranyi et al. 2009). Massoli et al. (2010) showed for different proxies of anthropogenic and biogenic SOA that the relationship between O/C ratio and CCN-derived kappa value is non-linear.”

References added:

Petters, M. D., Wex, H., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol - Part 2: Theoretical approaches. *Atmos. Chem. Phys.*, 9, 3999-4009, 2009.

Prenni, A. J., Petters, M. D., Kreidenweis, S. M., DeMott, P. J., and Ziemann, P. J.: Cloud droplet activation of secondary organic aerosol. *J. Geophys. Res.*, 112, D10223, doi:10.1029/2006JD007963, 2007.

Wex, H., Petters, M. D., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol: Part 1-Evidence from measurements. *Atmos. Chem. Phys.*, 9, 3987-3997, 2009.

Jurányi, Z., Gysel, M., Duplissy, J., Weingartner, E., Tritscher, T., Dommen, J., Henning, S., Ziese, M., Kiselev, A., Stratmann, F., George, I., and Baltensperger, U.: Influence of gas-to-particle partitioning on the hygroscopic and droplet activation behaviour of α -pinene secondary organic aerosol. *Phys. Chem. Chem. Phys.*, 11, 8091-8097, 2009.

Massoli, P., Lambe, A. T., Ahern, A. T., Williams, L. R., Ehn, M., Mikkilä, J., Canagaratna, M. R., Brune, W. H., Onasch, T. B., Jayne, J. T., Petäjä, T., Kulmala, M., Laaksonen, A., Kolb, C. E., Davidovits, P., and Worsnop, D. R.: Relationship between aerosol oxidation level and hygroscopic properties of laboratory generated secondary organic aerosol (SOA) particles. *Geophys. Res. Lett.*, 37, L24801, 2010.

Minor comments:

P 19312, line 19 – define UNIFAC

This has been added to the text: UNiversal Functional Activity Coefficient

P 19313, line 1 – Hegg et al. (Hegg, D. A., S. Gao, W. Hoppel, G. Frick, P. Caffrey, W. R. Leitch, N. Shantz, J. Ambrusko, and T. Albrechtinski, 2001: Laboratory studies of the efficiency of selected organic aerosols as CCN. Atmos. Res., 155-166.2001) and Shantz et al. (Shantz, N.C., W.R. Leitch, Peter F. Caffrey, 2003: Effect of organics of low solubility on the growth rate of cloud droplets. J.

Geophys. Res., 108, 4168- 4177.2003) looked at the hygroscopicity of α -pinene oxidation products in a chamber.

The two references have been added.

P 19313, line 4 – clarification of this statement is needed. Was the higher yield due to increased SOA mass concentration or some other factor?

The text: “Duplissy et al. (2008) showed that at higher precursor concentration (resulting in a higher SOA yield) the hygroscopicity of the SOA is lower. They proposed that at higher concentration more volatile (and less oxidized) products partition into the particle phase, leading to a decrease of the hygroscopicity of the particle.” has been replaced by “Duplissy et al. (2008) showed that at higher precursor mixing ratios, which result in higher SOA concentrations, the hygroscopicity of the SOA is lower.”

P 19315, section 2.2 – Consistent with the Mexico discussion, a brief outline here of the instrumentation at the Jungfraujoch would be helpful.

The following sentence has been added in the section 2.2

“An Aerodyne quadrupole AMS and an HTDMA were deployed at the Jungfraujoch research station.”

P 19316, lines 4-8 – have these three HTDMAs been compared?

No, these three HTDMAs have not been compared. The main technical comparisons between these HTDMAs are given in the next line: “The residence times of the aerosol in the humidifier, the chosen RH and the operating temperature were (15 s, 95%, 20 °C), (10 s, 85%, 40 °C) and (20 s, 85%, 25 °C) correspondingly.”

P 19317, line 6 – as follows

It has been corrected.

P 19319, line 11 – lower

It has been corrected.

P 19320, lines 10-11 – SOA from α -pinene being discussed here. The reference needs to be clarified.

We added: (see Figure 1 in Duplissy et al. 2008).

P 19340, Figure 7 – “sup” should be super.

It has been corrected.