Anonymous Referee #1

Received and published: 15 October 2009

General comments: The present manuscript discusses how relative humidity affects hygroscopicity and SOA chemical composition. The authors found that hygroscopicity (k value) of SOA particles increases linearly with the increase in water mixing ratio.
The change in k values was related with AMS signals. This result demonstrates that hygroscopicity of SOA particles can be related with SOA chemical compositions using currently available techniques. I think that this result stimulates further investigations on the topic. The manuscript seems to be well written. I have some comments.

Author response: We thank the reviewer for his/her comments on our manuscript. In the following, the original reviewer comments are in italic and our responses to his/her comments in regular type.

Specific comments P16687 Section 2.1 I could not find the descriptions on chamber temperature and VOC concentration in the chamber. As these values may affect the chemical composition and hygroscopicty of SOA particles, please mention them.

Author response: Experiments took place at room temperature (typically 24°C). The real concentration of α-pinene inside the chamber was not specifically measured during this work. Indeed, as describe in our previous paper (Wex et al., 2009), "the α-pinene vapor was generated by injecting a liquid flow of α-pinene, controlled by a microliter peristaltic pump into a stainless Swagelok tube, fitting reducer (SS-200-R-2) where it evaporated into 5-10L min-1 of dry hydrocarbon free synthetic air. (…) In the absence of chemical reactions, α-pinene concentrations were in excess of that of ozone, i.e. < 1 ppmv. So the amount of VOC that reacted was controlled with the ozone concentration". Therefore, the SOA only depended on the ozone concentration, which was measured during all experiments. This was added in the text.

P16688L2: According to the text, it is likely that 2-butanol was used as OH scavenger. However, Figure 1 indicates that 1-butanol was employed. Which compound was used in the experiment?

Author response: Only 2-butanol was used as OH-scavenger during this study. We corrected it on Fig. 1.

P16689L15: Is it confirmed that TD-Bypassed data are identical to the data obtained at thermodenuder operated at the room temperature? Gas/particle partitioning of semivolatile compounds may change in the thermodenuder because charcoal absorbs volatile gas compounds.

Author response: Your remark can indeed be confirmed. The heated part of the TD is always followed by a section containing active coal in order to avoid recondensation of the volatile fraction on the particles (Huffman et al., 2008; Wehner et al., 2002; Wu et al., 2009) and references within). In our work, the Mass Fraction Remaining (MFR) is calculated as the ratio of the SOA mass concentration measure at a define temperature to the concentration in the by-pass line (*i.e.* at room temperature). Each time, the MFR measuring with a TD setting at room temperature was close to 100% (according to sampling error measurements). For this reason, we can conclude that the active charcoal did not affect the SOA composition.

P16690L16 Does this mean that the DMA for LACIS was scanned as a DMPS? It is worth to mention which diameter(s) was selected.

Author response: Mentioning the DMPS here was a typo. The DMA at the top of LACIS selecting particles for LACIS was set to a fixed diameter for each experiment. It was set to 350 nm for experiment 7, to 275 nm during experiment 9, and it was set to 300 nm for experiment 10 at a RH of 99% to derived κ; as already mention in the text. (Detail of the selected LACIS diameter particles was added on table 1 of the manuscript). The DMA in front of the CCNc, however, was used as a DMPS, so that the critical diameter for activation could be derived while at the same time number size distribution were obtained. The size range that was scanned went from 30 nm to 500 nm in 30 steps, with a logarithmic equidistant spacing between the steps.
The text in part 2.3 "Determination of the hygroscopic properties of the SOA" was modified to: "The hygroscopic growth factor was obtained from the measured diameter at the LACIS outlet divided by the corresponding dry diameter selected at the DMA (see table 1 for detail of the selected particle diameter used for each experiment)."

P16690L21 Is Kelvin effect considered in this calculation? Is it possible to describe briefly how water activity is calculated?

Author response: Absolutely, the Kelvin effect has to be (and was) accounted. As precise in our previous paper (Petters et al., 2009), water activity was calculated according to the following equation:

$$a_w = \frac{S}{K}$$

Where S is the saturation ratio (S=RH/100) and K is the Kelvin term.

$$K = \exp\left(\frac{4\sigma_{s/a} M_w}{RT\rho_w D}\right)$$

Where ρ_w is the density of the water, Mw is the molecular weight if water, $\sigma_{s/a}$ is the surface tension of the solution/air interface, R is the universal gas constant, T is the temperature, and D is the diameter of the droplet. It now is mentioned in the manuscript, that the curvature of the particles was accounted for.

P16712 (Figure 7) It seems that data of experiment 9 are different from that of experiment 10. It is pronounced for CO2+ and CHO. Can the authors give the possible explanations for this difference? Although, the authors are trying to relate the oxidation levels with k value in this figure, this point is not discussed in detail. k value is affected by various factors (e.g., Molecular weight, soluble fraction, non-ideality of solution, density). It is not obvious which factor is influenced by the fraction of CO2+ or CHO+. Can the authors add some discussion regarding this point?

Author response: It is true that the results for CO_2^+ and CHO for experiment number 9 appear to be slightly higher than the other points. Data from experiment 9 and 10 are, however, still in agreement within the measurement uncertainty.

It is true that molecular weight, chemical composition and soluble fraction are directly influencing κ. However, ozonolysis of α-pinene lead to formation of C10 and C9 compounds with one or more aldehydes and organic acids functions (pinonaldehyde, norpinonaldehyde, pinonic acid, pinic acid, norpinoninc acid …) as well as smaller organic compounds. Induced modifications of the SOA chemical composition by the presence of water during the ozonolysis procedure correspond to changes between different oxidation pathways which will directly contribute to the changes on the volatility and hygroscopic properties of the particles. With the setup used during this work, we can not determine the exact chemical composition of the generated SOA; AMS provided information of the bulk of the SOA. Moreover, the ionization source of the AMS induced a high fragmentation level of the organic compounds. For this reason, modification of the molecular weight of the molecule by example can not be measured. However, modification of the bulk composition can easily be measured using the oxidation level of the SOA as we did using ratio CO_2^+/CH and ratio CHO/CH. This is the reason that we correlated κ with the oxidation level parameters. In the last part of the manuscript (3.2.2), we made some hypothesis to correlate our results with the state-of-the-art of the α-pinene ozonolysis mechanism and concluded by hypothesis that observed modification on the chemical composition and physical properties could be partially explained by increased of formation of pinonaldehyde and pinonic acid.

Technical comments Abbreviations should be shown after the word is shown in full form. e.g., Leipzig Aerosol Cloud Interaction Simulator (LACIS) (P16685L6)

Author response: Corrected.

P16695L9 "Fig 6" in the text likely corresponds to Figure 7.

Author response: It was corrected in the text.

Figures In some figures (Figure 2,4,6,and 7), markers for data are too small, and I could not see them well. Would you make them larger?

Author response: Data markers were increased in all figures.

Figure5-7(y-axis) It seems that "ratio mz44/organic" and "contribution of m/z44 to organic signal" are the same. If so, please use only one notation.

Author response: It was changed in figure 5 to "ratio m/z 44 to organic signal".

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