Answers to Referee #1

The authors appreciate the time the reviewer have spent in assisting us to produce a high quality, understandable publication. All the requested corrections and suggestions are addressed and introduced to the revised version of the manuscript.

Major comments:

The gap of knowledge, which this study strives to fill, could be written out more explicitly both in the introduction and conclusions. According to this, the main goals of the study, given in the end of the introduction, should be revised. At least the goals (1) and (2) are not discussed in the manuscript at all. Also, more discussion about how the results obtained in this study would contribute to, e.g., estimating the radiative forcing, should be added, because this is given as a motivation for this work in the introduction. Also, the literature could be rechecked for the last 2-3 years.

Reply: We agree that the introduction was not properly connected to the main text and thus, the goals of this study were revised: "Our main goals were 1) to characterize the size- dependency of aerosol hygroscopicity and volatility in a boreal forest environment and 2) to get new insight into the relation between aerosol chemical composition and its water uptake properties via the performed measurements. Such information is essential for both light scattering properties and cloud condensation nuclei activity of atmospheric aerosols".

The recent literatures on aerosol hygroscopicity were rechecked and the following papers were discussed in the introduction: Henning et al., 2012 and Saarikoski et al., 2008.

The conclusions are more like a summary: Please consider making the section more compact with only the conclusions in it, or rename it "Summary and conclusions".

Reply: We agree. The section was renamed as "Summary and conclusions" according to your suggestion.

Minor comments:

P29100, L11-13: Please add appropriate references.

Reply: The references Reutter et al. (2009) and Zhang et al. (2008) were added.

P29100, L25-27: The introduction of different measurement techniques can be limited.

Reply: The sentence was shortened to the following form: “These techniques include e.g. an electrodynamic balance (EB, Tang and Munkelwitz, 1994) and hygroscopicity tandem differential mobility analyzer (H-TDMA, Liu et al., 1978; Swietlicki et al., 2008).”

P29100, L28-29: Repetition of P29100, L23-24.

Reply: L23-24 discusses different techniques or methods; while L28-29 discusses different compounds. According to your suggestion we modified the text as: “Measurements of aerosol hygroscopic properties have been performed extensively around the world both in laboratory and field conditions using different techniques, e.g. an electrodynamic balance (EB, Tang and Munkelwitz, 1994) and hygroscopicity tandem differential mobility analyzer (H-TDMA, Liu et al., 1978; Swietlicki et al., 2008). With these techniques hygroscopic properties of many inorganic and some organic compounds have been investigated extensively (Hämeri et al., 2001; Wise et al., 2003; Massoli et al., 2010)”.

P29101, L11-14: Five references for thermo denuder; are all these necessary?

Reply: We agree that not all of these references are needed. We removed the first two references from the list.

P29101, L22-25: Some evaporate below 300 °C – some not, this is trivial. Please rephrase.

Reply: Different compounds evaporate at different temperatures according to their volatility properties (Turpin et al., 2001; Raatikainen et al., 2010). Most compounds present in atmospheric aerosol will fully evaporate at temperatures below 300 ˚C which is also the maximum temperature (approximately) used in this study for thermodenuder experiments. However, there are compounds that are non-volatile even at such high temperatures. These include black carbon (BC), sea salt and crustal material. For submicron particles in inland areas sea salt and crustal material are not important. According to previous studies (Backmann et al., 2010; Häkkinen et al., 2012), in addition to BC there may be also other essentially non-volatile compounds, possibly organic polymers or salts, present in submicron atmospheric aerosol. We added discussion about this to the revised manuscript.

P29103, L22: Does the 45 min scan time include all particle sizes?

Reply: Yes, within the 45 minutes, all particle size scans are covered.

P29103, L23: Do Villani et al. and Johnson et al. refer to the accuracy of your instrument?

Reply: 2% error was obtained from the calibration of this technique. The sentence in the manuscript was modified as: “The relative humidity of the aerosol after the humidifier was set to 90% within 2% error. There are also other studies using a VH-TDMA setup (Villani et al., 2008; Jonhson et al., 2005)”.

Subsection 2.2.2: Please give the supersaturation(s) that were used in the CCNc analysis.

Reply: The text was modified as: “In this study, five levels of supersaturation were applied: 0.09, 0.22, 0.48, 0.74 and 1.26 %.”

Subsection 2.2.3: The DMPS and VDMPS have different size ranges; any comments on this?

Reply: The VDMPS measured particles up to 1 μm in diameter alike the DMPS. However, because no PM1.0 cyclone was used in front of the VDMPS, the VDMPS was occasionally detecting evaporated supermicron particles, which caused noise in the data at the larger end of particle sizes. The noise was removed by selecting size range of 20-500 nm for further analysis. For the calculation of aerosol volume fraction remaining (VFR), the same size range (20-500 nm) was selected from the DMPS data. More detailed information on the size range selection for the VDMPS-DMPS analysis can be found in Häkkinen et al. (2012). We modified the section (Sect. 2.2.3) concerning the size ranges of the DMPS and VDMPS accordingly.

Subsection 2.2.3: The particle losses in the thermodenuder are given but how about the particle losses in the other instruments?

Reply: Compared with the particle losses in the thermodenuder, the particle losses in the other instruments can be neglected, since particle losses increase with increasing temperatures. In the thermodenuder, high temperatures were applied, so particle losses were significant and had to be taken into account.

P29105, L1: Are there other refractory compounds than BC and sea salt that should be taken into account?

Reply: Besides BC and sea salt, there are some other refractory compounds that AMS cannot measure, e.g. crustal materials and metals. However, at our measurement site in Hyytiälä, the concentrations of crustal materials and metals are typically quite low (Saarikoski et al., 2005), especially in submicron aerosol, and therefore these species were not included in our analysis.

P29105, L3: The aethalometer measured PM2.5 but the AMS only PM1.0, could you comment on this?

Reply: Based on limited literature data on BC size distributions (e.g. Hitzenberger and Tohno (2001); Jaffrezo et al., 2005; Saarikoski et al., 2008), only a relatively small fraction of BC is expected to be in the size ranges 1.0 – 2.5 μm. Thus, the different cut off sizes should not be a major problem here. We added a mention of this issue to the revised manuscript.

Subsection 2.2.5: The trace gases were measured at several heights. Is there variation in the concentrations between the different heights? Which of these was used when the aerosol (at which height was the aerosol measured?) and trace gases were compared in Figure 3?

Reply: Yes, there is some variation in the concentration of these trace gases between different height levels. In this study, we used the concentrations measured at the 4.2 m height, as the aerosol measurements were also performed at the same height. Therefore, the comparison between the aerosol properties and the concentrations of trace gases is reasonable.

P29106, L12: The median temperatures could be listed here.

Reply: The following sentence was added to the text: "The median temperatures obtained in this study were 35, 65, 95, 125, 155, 185, 215, 245 and 268 °C."

Equation 1: GF(90% RH) can be omitted.

Equation 2: GF(T) can be omitted.

Equation 3: GF(90% RH, T) can be omitted.

Equation 4: GF(90% RH, T, aerosol residual) can be omitted.

Reply: We agree that these equations are simple and already known, but since there are so many parameters and variables in the manuscript, we prefer to keep them there to make everything easy to follow.

Also, could this be written as GFH,residual = GFV H /GFV , and further the GFV H and GFV could be substituted into the equation.

Reply: Yes, the equation was changed accordingly: $GF_{H,residual} = \frac{GF_{VH}}{GF_V} = \frac{Dp\ (90\%\ RH,T)}{Dp\ (T,RH_{dry})}$

Equation 5: The substitution of D = DpGFH could be written out for clarity. Should S(D) be now S(Dp, GFH), and can you substitute S(Dp, GFH) = 0.9?

Reply: That is correct. The text and equation (Eq. 5) were changed as suggested.

Subsection 2.3.4: Can the same ϰ value be used for subsaturation (VTDMA) and supersaturation (CCNc)?

Reply: This issue is addressed below (see answer to comment P29111).

P29110, L10: There is fluctuation in the BC mass fraction (Figure 2) during the measurement period, could you give, e.g., the average and standard deviation here?

Reply: The standard deviation and average values were added to the text (mean BC mass fraction of 5.88% with standard deviation of 2%).

P29110, L12: Please give justification for the assumption that the BC mass was solely in submicron particles. Was this true also during the biomass burning period?

Reply: Unfortunately we do not have information about the BC size distribution during our measurements. However, based on previous studies (e.g. Jaffrezo et al., 2005; Saarikoski et al., 2008; see also our response to the comment P29105 above) the assumption that BC is mainly in submicron particles is reasonable. Hitzenberger and Tohno (2001) observed that only a small fraction (around 15% on average) of BC mass is in coarse mode particle in urban areas. This information can be related to e.g. biomass burning aerosols. However, the BC mass fractions given in our study should be taken as upper estimates.

P29110, L21-27: What is your interpretation of the positive correlation between the GFH and sulphate?

Reply: GF_H and sulphate concentrations were correlated positively. This is because aerosol particles containing more sulphate are more hygroscopic. We mentioned this in the revised manuscript.

P29111, L17-19: This might be the answer to my previous comment for subsection 2.3.4. Could one reason for the failure of the comparison be that Equation (5) is for a droplet and Equation (6) is for a wet particle, so the sizes of these and therefore the amount of water on them and their composition are quite different?

Reply: The text was revised by adding new reasons as: "Several factors may contribute to these observations. First, it is know that organic compounds have different degrees of dissolution at sub- and supersaturated conditions, and different components with varying solubilities in the same particle may exhibit discontinuous hygroscopicity (Prenni et al., 2007). Second, the particle mixing state and potential phase separation (liquid or solid phases) could make aerosol hygroscopicity to depend on RH (Zardini et al., 2008; Henning et al., 2012). The observed differences in hygroscopicities between the sub- and supersaturated conditions could partially be due to the different characteristic designs of the instruments. For example, the HTDMA and CCNc have different residence times for the aerosol to reach equilibrium at the desired RH or supersaturation (Juranyi et al., 2009). The study by Good et al. (2010) found a close agreement between κ_{CCN} and κ_{HTDMA} when sulfate and ammonium comprised the majority of the aerosol mass. This suggests that the aerosol composition, and especially the presence of organic compounds, play a key role in the differences in the aerosol hygroscopicity between sub- and supersaturated conditions".

P29112, L8-10: An unclear sentence, please clarify.

Reply: This issue was addressed above (Comment P29111).

P29112, L14-15: The CCNc data in Figure 5 shows the opposite (larger particles have lower hygroscopicity).

Reply: You are correct. We modified the text accordingly: "General patterns of κ as a function of particle diameter were quite similar from both HTDMA and CCNc measurements: the aerosol hygroscopicity increased with increasing particle diameter up to 100-200 nm (see Fig. 5)". However, there was a decrease in κ_{CCN} for 200 nm particles compared to particles of 100 nm. It should be also noted that the variation in aerosol hygroscopicity for the CCNc data (200 nm particles) is the greatest.

P29112, L23-27: This paragraph belongs to the Methods.

Reply: The paragraph was moved to subsection 2.3.3 (Materials and Methods, Volume Fraction Remaining).

P29113, L3-6: Can you use the AMS results to discuss the differences in the particle chemical composition?

Reply: Unfortunately our AMS data were not size resolved and thus, we could not use AMS results directly to investigate the differences in aerosol volatility as a function of particle size.

P29113, L12: Can you give estimation, how much more the aerosols would evaporate during the extra 9 seconds in the VTDMA?

Reply: Since the VFR from VDMPS data was obtained for polydisperse aerosol volume distribution (20-500 nm), the obtained VFR describes the volatility of relatively big particles rather than small ones. VFR from VTDMA data was, instead, obtained for monodisperse aerosol and thus, the largest difference in the VFRs (VDMPS vs. VTDMA) is observed between VDMPS and VTDMA-50 nm. It should be noted that the difference between VFRs of VDMPS and VTDMA-150 nm is not very significant indicating that the difference in the residence times (1 s vs. 10 s) is not that important when studying Hyytiälä aerosol. For comparison the MFR (mass fraction remaining) of laboratory generated aerosol, α-pinene, (Riipinen et al., 2010) at 200 ˚C was around 50% lower when 10 s residence time was used compared to 1 s residence time. We added discussion about this to the revised manuscript.

Subsection 3.3: This subsection could be moved to the Methods section.

Reply: We agree this calculation is a method, yet it also gives results that can be discussed here rather than elsewhere in the text. Therefore we think the proper place for the subsection about theoretical aerosol hygroscopicity is in Results and Discussion.

P29116, L5: Talking about evaporation, should this be GFV,BC = 1?

Reply: That is correct. The text was changed accordingly as $GF_{V,BC}=1$.

P29116, After Equation (10): The values of the parameters could be inserted here instead of Table 2.

Reply: We prefer to keep Table 2, since we think it helps the reader to follow the discussion easier than if the same information were part of the text.

P29118, L6: An unclear sentence, please clarify.

Reply: The sentence was changed into the following form: “Since black carbon is hydrophobic, this result supports recent studies that have found very low volatile, yet hygroscopic material that is not BC, in submicron ambient aerosol particles.”

P29118, L20-27: This discussion could be in the Results section, not in Conclusions.

Reply: We agree. The discussion concerning the hygroscopicity of low-volatile organics was moved subsection 3.3 as suggested.

Table 1: The structure of the table is unclear. Please consider separating the three last rows.

Reply: These rows in Table 1 were separated as suggested.

Table 2: This table is unnecessary, if the values are given in the text after Equation (10).

Reply: We prefer to keep Table 2 (see our response above).

Figure 4: By writing “too few data points for 50 nm and 110 nm”, should these plots be removed?

Reply: We will leave the data points but do not make deep conclusions about the size dependency. Even though there were not too many data points for this period for these two sizes, the plots are correct in that they represent the available data.

Figure 5: Please consider changing either the red or magenta; they are easily mixed.

Reply: The color (magenta) was changed to black.

Figure 6: Is Hakkinen et al. data from VTDMA or VDMPS measurements? Please indicate in the legend or the caption.

Reply: Häkkinen et al. (2012) data is from VDMPS measurements. This information was added to the figure caption.

Reference:

Henning, S., Ziese, M., Kiselev, A., Saathoff, H., Möhler, O., Mentel, T. F., Buchholz, A., Spindler, C., Michaud, V., Monier, M., Sellegri, K., Stratmann, F.: Hygroscopic growth and droplet activation of soot particles: uncoated, succinic or sulfuric acid coated, Atmos. Chem. Phys., 12, 4525-4537,2012.

Hitzenberger, R. and Tohno, S.: Comparison of black carbon (BC) aerosols in two urban areas – concentrations and size distributions, Atmos. Environ., 35, 2153–2167, 2001.

Jaffrezo, J. -L., Aymoz, G., Cozic, J.: Size distribution of EC and OC in the aerosol of Alpine valleys during summer and winter, Atmos. Chem. Phys., 5, 2915-2925, 2005.

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Saarikoski, S., Mäkelä, T., Hillamo, R., Aalto, P. P., Kerminen,V.-M., and Kulmala, M.: Physico-chemical characterization and mass closure of size-segregated atmospheric aerosols in Hyytiälä, Finland, Boreal Env. Res., 10, 385–400, 2005.

Saarikoski, S., Frey, A., Mäkelä, T., Hillamo, R.: Size distribution measurement of Carbonaceous particle matter using a low pressure impactor with quartz substrates, Aerosol Science and Technology., 42, 603–612, 2008.

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