Answers to Referee #2

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:

There are some parts in the manuscript that could be improved considerably. In Page 29110 the correlations between GF and different concentrations is not done as well as it should be. In Figure 3 linear fit is presented, although it is obvious that there is no linear correlation between the GF and organics to sulfate ratio. I would prefer presenting correlations so that the mass fraction of different particulate phase compounds could be shown in x-axis. Also if it is stated that correlations between other variables are tested then those could be presented. If there is correlation between GF and ozone it would be nice to see how good the correlation is.

Reply: We agree that the fits in Fig. 3 do not represent the data as well as it should. Thus, Fig. 3 was modified as suggested (see Fig. 1A) and new fittings were made based on the logarithm of the variables in the x-axis (aerosol mass fraction of different compounds).

We prepared a table (see Table 1A) presenting correlations between the values of GF_H of different sizes and the concentration of chemical species both in particle phase and in gaseous phase: SO_2, O_3, H_2SO_4-proxy and SO_4^{2-}, BC and organics with correlation coefficient (R) and confident interval values (in square brackets). The effective data number vs. the total data number is shown in brackets. Among these six species, hygroscopic growth factor correlated the best with gas phase O_3 and sulfate (from AMS data) with relatively high data coverage. The positive correlation between GF_H and ozone could be linked to the fact that high ozone concentration in the air means more oxidizing agents and likely more oxidized aerosols. However, this reasoning is too speculative and the correlation is probably more to do with anthropogenic emissions. The more sulfates there were in the particles, the particles were observed more hygroscopic. For other species, hygroscopic growth factor correlated better with smaller particles (50 nm, 75 nm) compared with larger particles (110 nm). However, it should be noted that the chemical composition information from AMS and aethelometer is not size resolved, and therefore the size dependent correlations cannot be done.

It would be interesting to see how particle volatility is affected by biomass burning. Similarly it would be nice to see if the HTDMA and CCNc derived hygroscopicity has similar difference with different air masses. Plotting for example time series of CCNc derived hygroscopicities in a similar way, as growth factors would make this possible.

Reply: VFR as a function of temperature for the whole period and the whole period without biomass burning and biomass burning period are plotted in Fig. 2A. The main difference is observed at temperatures >100 °C but <150 °C, where particles evaporated more during the biomass burning period than during other times. This agrees with the results illustrated in Fig. 4 (in the manuscript) that the mass fraction of organic material is higher than other species during biomass burning period. Particles from biomass burning period have lower hygroscopicity in general (see the upper panel in Fig. 4), thus have a lower O:C value, indicating higher volatility that allows the particles to evaporate more. However, the hygroscopicity and volatility of organic materials may vary a lot during the whole measurement period. Hence, here we can only speculate where the observed differences in volatilities and hygroscopicity between different air masses may be resulting from different groups of organic materials with different origins.

The time series of CCNc-derived and HTDMA-derived hygroscopicity parameter are both plotted in Figure 3A. In general, CCNc-derived hygroscopicity parameter is higher than HTDMA-derived hygroscopicity parameter for all sizes for both periods (biomass burning vs. no biomass burning). The discrepancy between CCNc-derived and HTDMA-derived kappa is even larger for biomass burning period. This could be explained by the fact that the fraction of organic materials in the aerosol is higher than during the background period. Meanwhile, the properties of the organics, including hygroscopicity and volatility, may vary a lot between both periods. Both the fact that there is more organics in biomass burning aerosol and the fact that the organic properties may be very different in different air masses would enhance the effect of organic compounds on the difference between two instruments' results. This is discussed in more detail in the answer to the last major comment.

The discussion about the possible composition of nonvolatile aerosol remaining after heating is weak. There is no discussion about the role of sodium chloride or potassium salts for example. The aethelometer measurement for black carbon concentration has high uncertainty and thus the hygroscopicity of unknown nonvolatile component must be quite uncertain.

Reply: We agree. We put more emphasis on discussing the issues you raised in the revised manuscript. At the measurement site in Hyytiälä, concentration of sodium chloride and potassium salts are typically quite low (Saarikoski et al., 2005), and their importance in the chemical composition of aerosol (especially submicron size) is minor. Raw data obtained from Aethalometer has high uncertainty that is mainly related to the non-linearity in the relationship between BC concentrations and light transmission through the filter (Reid et al., 1998; Bond et al., 1999). However, in this study the raw data was corrected using an approach by Weingartner et al. (2003). Also, by using high wavelength data (880 nm) the uncertainty in BC concentrations caused by brown carbon can be minimized. The assumption made for BC mass being solely in submicron aerosol is also a good assumption, at least based on previous size-resolved BC data showing that only around 15 % of BC mass is in coarse mode particles in urban areas (Hitzenberger and Tohno, 2001). In Hyytiälä the percentage may be similar or possibly lower. Therefore, we believe the results presented in this study e.g. concerning the hygroscopicity of the non-volatile aerosol fraction are reliable. However, we also acknowledge that alike other experimental data, the BC data has its uncertainties.

In page 29114 I do not completely agree with the authors in the analysis of small particle composition. If the composition is more homogeneous, why the VFR is decreasing almost linearly as function of temperature? What do you actually mean by homogeneous here? What does it actually mean for chemical composition if smaller particles have higher hygroscopic growth factor after volatilization than the bigger particles but the VFR is smaller?

Reply: We agree that “homogeneous” was an incorrect term here. What we actually mean is well mixed. One potential reason for the observed differences is that the smallest particles (50 nm here) have so small sulfate fraction that its evaporation does not influence the temperature dependence of this curve in the lower part of Fig. 7 (in the manuscript). The gradual drop in GF for these particles in the upper part of Fig. 7 is most likely due to presence of organic compounds of different saturation vapor pressures (i.e. volatility) in these particles. This part of the text was rewritten to reflect these points in a clear way.

For small particles hygroscopicities from sub- and supersaturations are closer to each other than for large particles, although the mass fraction of organics is probably largest for small particles. At the same time the difference between hygroscopicities is said to be caused by dissolution of organics. How do you explain this? It would be interesting to see time series instead of mean values. Also it should be stated if only the temporally overlapping data is presented in Figure 5 or is it mean of all data available from the campaign.

Reply: In the literature, dissolution of organics is often used to explain the differences in particle hygroscopicity between sub- and supersaturated conditions. We agree that observed size dependency is not consistent with this idea indicating there is more to that than organic dissolution. This part of the text was rewritten 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”.

Minor comments:

Page 29099, line 11-12: I would not say it is a fact that particulate organics have a different degree of dissolution in sub- and supersaturated conditions. Not at least all of them, so please rewrite the sentence.

Reply: We removed this claim from the abstract (see our response above).

Page 29100, line 12-14: Hygroscopicity affects the CCN number concentration but it will not determine it alone as aerosol particle number concentration is quite important factor also. More than that, it will not determine the lifetime of cloud, but might in some cases affect it. Please rewrite.

Reply: The sentence was changed to: “The ability of particles to take up water, called hygroscopicity, is one of the important factors that determine the number concentration of cloud condensation nuclei and also affect the lifetime of the clouds (Reutter et al., 2009; Zhang et al., 2008).”

Page 29113, lines 3-5: How important Kelvin effect is when volatility is measured? I think it is quite minor compared to composition.

Reply: We agree that the role of the Kelvin effect is probably minor here.

The sentence was modified as: “Small particles were observed to evaporate more at lower temperatures than bigger particles, probably due to differences in the particle chemical composition.”

Page 29113, lines 23-25: What about sodium chloride and potassium salts, or other inorganic components that have high temperature for volatilization?

Reply: Refractory compounds such as sea salt and crustal material are not important in inland areas for the chemical composition of submicron aerosol (see more above). We have addressed this issue in the revised manuscript.

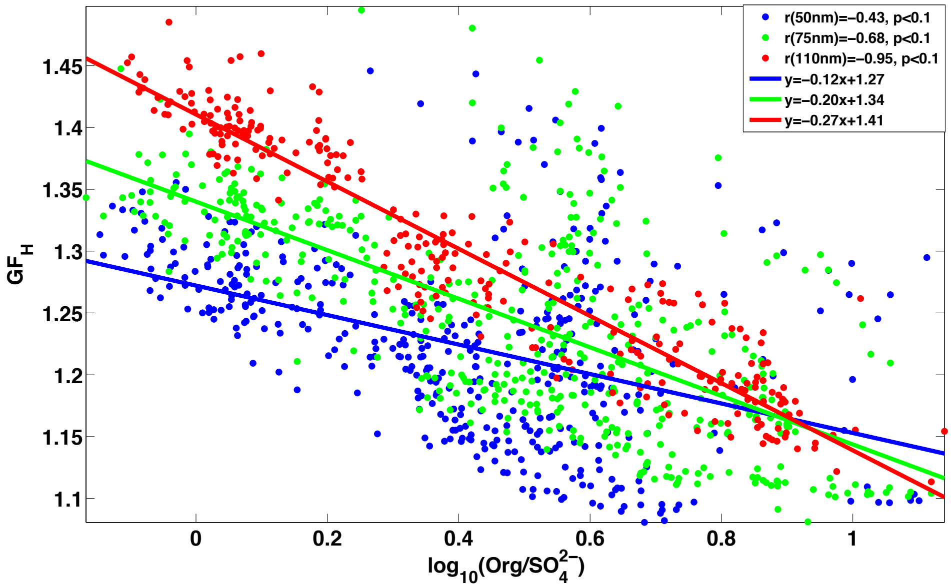

Figure 1A: Correlations between the averaged hygroscopic growth factor and the ratio between the concentrations of particulate organics and sulfates from AMS data for three different particle sizes. The observed correlations were negative and the best correlation was for the particle diameter of 110 nm.

Table 1A: Correlation (with p-value within 5%) between growth factor (GF_H) and the concentrations of chemical species both in particle phase and in gaseous phase: SO_2, O_3, H_2SO_4-proxy and SO_4^{2-}, BC and organics. The numbers in the bracket are the effective data number to the total data number. The values in square brackets are the confidence intervals.

Size **Species**	**50 nm**	**75 nm**	**110 nm**
SO_2	R=0.5211 [0.394 0.628] (107/493)	R=0.45572 [0.314 0.578] (100/535)	R=-0.14772 [-0.412 0.139] (35/340)
O_3	R=0.58 [0.461 0.679] (103/493)	R=0.6110 [0.481 0.715] (81/535)	R=0.4547 [0.217 0.642] (40/324)
H_2SO_4-proxy	R=0.5538 [0.404 0.675] (73/226)	R=0.4786 [0.324 0.608] (81/255)	R=-0.1792 [-0.37 0.025] (66/161)
BC	R=0.31714 [0.148 0.468] (87/484)	R=0.20789 [0.015 0.386] (73/513)	R=-0.26151 [-0.492 0.003] (40/323)
SO_4^{2-}	R=0.61528 [0.472 0.727] (67/434)	R=0.72907 [0.607 0.818] (57/468)	R=0.6287 [0.293 0.827] (17/292)
Org	R=0.2632 [0.089 0.422] (86/434)	R=0.0845 [-0.117 0.279] (70/468)	R=-0.4106 [-0.657 -0.086] (25/292)

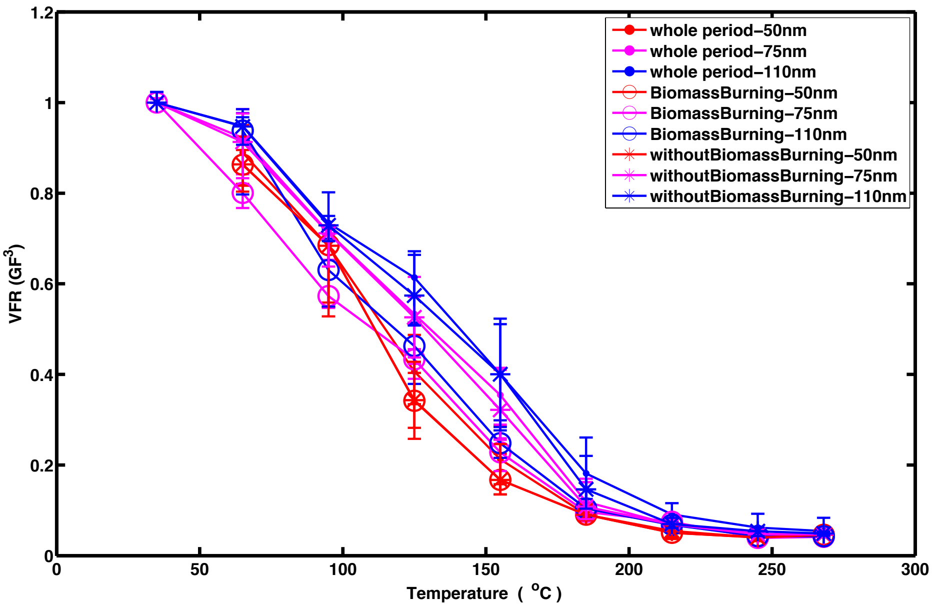

Figure 2A: Volume fraction remaining as a function of temperature obtained from VTDMA for different periods.

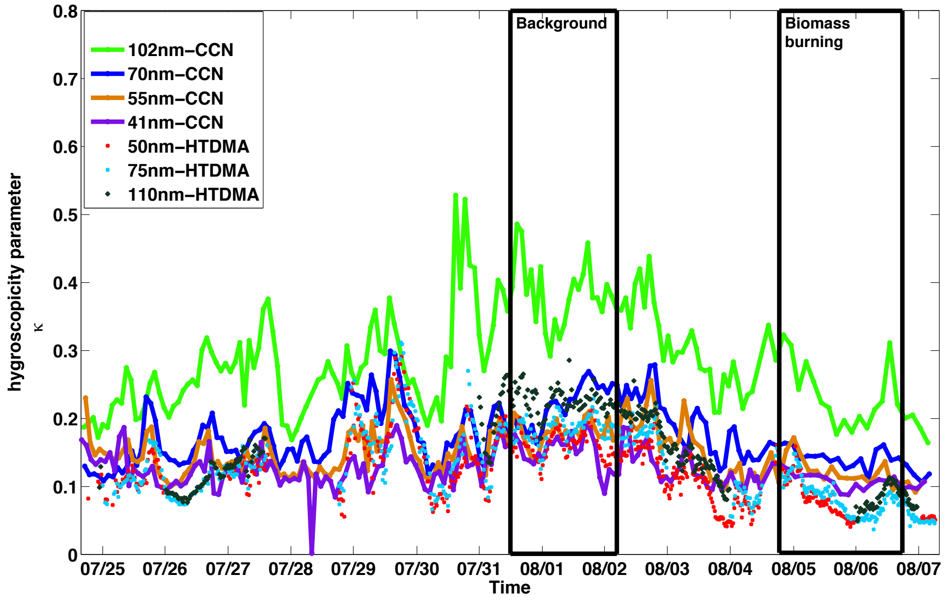

Figure 3A: Time series of hygroscopicity parameter obtained from both CCNc and HTDMA for different sizes.

Reference:

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