

Response to Referee #1

The authors would like to thank Referee #1 for his/her detailed, extensive and valuable comments to further improve and clarify the MS. We have considered all recommendations, and made the appropriate alterations. The changes can be explicitly tracked in the annotated version of the MS. Our specific responses to the comments are as follows.

Major comments

1. My major concern with this work is that some speculative conclusions are drawn from the measurements that are currently not supported (some instances are listed in the specific comments). The author should provide further evidences (or data) to support their conclusions or more extensive discussion, otherwise they should be removed.

We revised thoroughly and improved the Results and discussion section at several places and from several aspects with more detailed arguments. We also modified the body text to make our intentions and statements more detailed, specific, and further literature sources were also included to support our conclusions.

2. The presentation of data should be improved. There were several parameters used in the article to describe particles hygroscopicity and volatility. In order to avoid any confusion, I suggest that the authors use HGF and VGF to present particles hygroscopic and volatile growth factor in the body text and figures. The subscript like κ_{LH} is also suggested.

We adopted all these suggestions at several places throughout the abstract, body text and figures.

3. It would be advisable to discuss the impact of anthropogenic activities on aerosol hygroscopicity and volatility in detail. The comparison between workday and weekend is quite interesting. However, it still lacks some deep investigation. For example, it would be very interesting to know how the relationship between the number fraction and growth factor of NH and LV would change in workday and weekend. The comparison of the diurnal variation of particle number size distribution was also an interesting way in this section.

We completed new investigations in the requested sense, and their results were included in the discussions.

4. Although authors showed the data at Budapest, it was not very clear to me how the results of this study could be compared with previous studies at other urban areas.

We added new aspects and reformulated several sentences to place our results in an international framework in a more extensive manner.

Specific comments

1 P4 Line 12: Please add a citation about the earlier study here.

Several new reference was added.

2 P4 Line 34: Please add the unity of each parameter.

The actual values utilised in the calculations were explicitly given. The units of the parameters comply with SI units according to the Mathematical notation and terminology guidelines of the journal.

3 P4 Line 36: It would be better to use Kelvin temperature instead of Celsius temperature here.

The data was converted to Kelvin unit.

4 P4 Line 39: Is the depression of surface tension only controller by HULIS?

Atmospheric humic-like substances are reported to be the most abundant and important surface active aerosol component (e.g. Facchini et al., 1999; Fuzzi et al., 2001). They can decrease the surface tension of water substantially (Decesari et al., 2001; Salma et al., 2006), and this is ordinarily considered to be the most important contribution of the depression. The related sentence was reformulated to be more precise, and to include jointly the new aspects raised in Comments 4–6.

5 P5 Line 1-4: Is there other organics matter can reach the thermodynamic equilibrium fast enough to depress the surface tension in the humidifier tube.

A number of empirical relationships were reported to relate surface tension to concentration of OC or WSOC in droplets (e.g. Facchini et al., 1999). These were, however, suggested mainly for predicting the effect of organics on cloud activation. A full consideration of the effect in multicomponent aerosol systems has not been reported yet. It is expected, however, that such effects can be important in H-TDMA studies on nucleation mode particles (≈ 25 nm; Swietlicki et al., 2008). Therefore, the effects of organic compounds other than HULIS were not

considered in the present study. The related sentence was reformulated to include jointly the new aspects raised in Comments 4–6.

6 P5 Line 8: Please explain why the influence of surface tension is smaller under subsaturated condition.

The sensitivity of hygroscopic growth to the surface tension becomes more important with decreasing dry particle diameter and increasing RH since these are the conditions under which HGF is most sensitive to the Kelvin factor. The change in the Kelvin term caused by the altered surface tension in sub-saturated conditions is smaller when compared to its dependency in cloud activation. The related sentence was reformulated to include jointly the new aspects raised in Comments 4–6.

7 P5 Line 17: Why the parameter “volume fraction remaining” was used here to describe the particle volatility? The parameter “volatile growth factor” is quite enough in discussing particle volatility.

The two terms, namely the volatility growth factor and the volume fraction remaining have related meanings. The former quantity shows the diameter change, while the latter property represents a very expressive picture on the physical appearance of the coating and core of particles, and on their volume ratio. Therefore, we would like to keep this in the MS as well as an auxiliary property.

8 P5 Line 16: Please specify the actual value of RH when measuring the GF.

The actual RH corresponding to the HGF was added.

9 P5 Line 28: What is the meaning of “N” here? Do you mean total particles? It was a little bit confused. Please change the expression.

The abbreviation *N* was explained in page 5 line 23 of the original MS (and in the table/figure captions as well). We added further textual formulation now to increase its visibility.

10 P5 Line 37: It may be not necessary to mention the reason of missing data.

The part of the sentence was removed as requested.

11 P6 Line 3-4: According to Fig. 1, these two modes did not seem such distinct on January 16th, and the particle number concentration decreased on the same time. It would be interesting to analyze the data during this period.

The HGFs for the NH and LH modes were the closest to each other in the very beginning of 16 January during the time interval in which the atmospheric concentrations were low. Nevertheless, they could be well resolved. The effect was related to data fluctuations and possible effects of local meteorology.

12 P6 Line 30: According to former section, the RH in the humidifier was set to 85% at the beginning of measurement and then set to 90%. Please specific the GF-PDF was measured under which RH value. Please use HGF-PDF and VGF-PDF in this article.

The actual RH corresponding to the HGF was explicitly given in the text. The suggested notifications were adopted.

13 P6 Line 34: In sentence "... the median ambient concentration of these particles ...". Please add the actual value of the number concentration in this sentences.

The median concentration was expressed numerically as well as (in addition to Fig. 2a) as requested.

14 P7 Line 30: The author should note the equation used in calculating mean growth factor and number fraction.

Short descriptions with the equations were added (Eqs. 2 and 3.)

15 P7 Line 31: In order to avoid any confusion, please use "number fraction" instead of "relative concentration" in the discussion.

The requested change was adopted at several places in the body text and table/figure captions.

16 P7 Line 37: "The slight change in the hygroscopic growth behaviour cannot be reliably interpreted without measurements at high (>95%) RHs". Please give a more specific discussion on this sentence. Why the higher RH measurement was needed here?

The differences in the HGF are expected to become larger with RH in the sub-saturated interval, so the increased RH to 95% or even higher are expected to yield more evident or more reliable differences. The related sentence was reformulated.

17 P8 Line 17-19: I don't think that the GF for pure NH_4NO_3 and $(\text{NH}_4)_2\text{SO}_4$ higher than the measured value indicates the LH species become coated with inorganics species. It is not strong evidence. Please modify this sentence.

The sentence was removed.

18 P8 Line 21: Please use the full name instead of "TED/EDS".

The abbreviation TEM/EDS was extended and resolved as transmission electron microscopy with electron energy-loss spectroscopy (TEM/EELS).

19 P8 Line 34: "Our data are slightly below the average GFs". Please list the value of average GFs.

The sentence was extended to clarify our intension, and a comprehensive reference was added.

20 P9 Line 7: Please use standard deviation instead of "SD".

The abbreviation SD was accepted or even promoted in our recent ACP papers in the copy-edition phase as it conforms the English guidelines and house standards of the journal.

21 P9 Line 21-23: The correlation coefficients less than zero suggests that the number fraction of NH and LV modes were negative correlated with the total particle number concentration. Please explain the relationship between these negative correlations with the traffic emissions.

The negative signs were simply a typing mistakes, and they were removed.

22 Figure 4: Please use κ_{mean} , κ_{NH} and κ_{LH} in the label of y-axis.

The subscripts were added to the figures as requested.

23 P9 Line 41-42: The κ_{LH} was larger during daytime could result from the development of boundary layer at noon, which would be able to bring aged aerosol from upper atmosphere layer to the ground.

This effect was also added to the discussion as an important possibility, and a new reference was given.

24 P10 L1-2: Because 50 nm particles were more originated from primary emission. It would be better to be mentioned in the discussion.

We extended the text to include this information explicitly.

25 P10 L4-5: The changes in chemical composition including mass distribution for a certain particle size would lead to the change of number fraction in each mode and mean κ . The sentence should be modified.

The sentence was extended accordingly.

26 Figure 5: Please use VGF in the label of y-axis.

The axis label was modified as requested.

27 P11 Line 36-37: "... and since the atmospheric residence time of the particles with the selected diameters is estimated to be several hours." What is relationship between the particles residence time and separating the data? This sentence was not necessary. Please delete it or try to explain more.

Due to the limited residence time of particles with the investigated diameters, they remain in the air for several hours only, and therefore, their concentration levels on workdays and holidays usually differ substantially because of their different sources intensities during these time intervals. It is this basic property that makes the comparison of the workdays and holidays sensible. The statement was separated from the first part of the sentence, and largely reformulated to avoid the misunderstanding.

28 P12 Line10-11: "... but without substantial variation during the daylight period". It might be wrong. According to fig. 6 (c), the κ values increased during daytime and became much higher than in nighttime. Please explain that.

The sentence dealt with the daylight period only. Nevertheless, we reformulated and extended it to finish its misleading character.

29 Section 4.5: There is not essentially difference between VGF and VFR. Please delete the part of VFR and focus on the relationship between VGF and HGF.

The two terms, namely the volatility growth factor and the volume fraction remaining have related meanings. The former quantity shows the diameter change, while the latter property represents a very expressive picture on the physical appearance of the coating and core of particles, and on their volume ratio. Therefore, we would like to keep this in the MS as well as an auxiliary property. Nevertheless, we shortened its discussion as requested.

30 Section 4.5: It would be interesting to discuss the relationship between VGF and HGF for each mode.

New aspects of the comparison of VGF and HGF separately for the modes was added to the body text.

31 P13 Line 34-35: “The smallest particles (with dry diameters of 50 and 75 nm) appeared to be dominated by vehicle emissions since their size range, diurnal variability and timing matched the traffic intensity and 35 emissions.” It doesn’t consist with the former discussion in page 11 line 2-3 “Particles with diameters of 50 and 70 nm seemed more volatile during the daylight time period than the 110- and 145-nm particles. The latter two sizes are in the typical size range of fresh diesel emissions (Charron and Harrison, 2003)”. Please explain that.

It was not evident from the text that the first statement referred to LV particles, while the second sentence dealt with total particles. We reformulated one of the sentences and deleted the other sentence to clarify this situation.

Additional references

- Decesari, S., Facchini, M. C., Matta, E., Lettini, F., Mircea, M., Fuzzi, S., Tagliavini, E., and Putaud, J.-P.: Chemical features and seasonal variation of fine aerosol water-soluble organic compounds in the Po Valley, Italy, *Atmos. Environ.*, 35, 3691–3699, 2001.
- Facchini, M. C., Mircea, M., Fuzzi, S., and Charlson, R. J.: Cloud albedo enhancement by surface-active organic solutes in growing droplets, *Nature*, 401, 257–259, 1999.
- Fuzzi, S., Decesari, S., Facchini, M. C., Matta, E., Mircea, M., and Tagliavini, E.: A simplified model of the water soluble organic component of atmospheric aerosol, *Geophys. Res. Lett.*, 20, 4079–4082, 2001.
- Salma, I., Ocskay, R., Varga, I., and Maenhaut, W.: Surface tension of atmospheric humic-like substances in connection with relaxation, dilution and solution pH, *J. Geophys. Res.*, 111, D23205, doi:10.1029/2005JD007015, 2006.

Imre Salma
14–02–2018