

Interactive comment on “Hygroscopic growth of urban aerosol particles in Beijing (China) during wintertime: a comparison of three experimental methods” by J. Meier et al.

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Thank you very much for your comments to the manuscript. In the following, your statements appear in italic face, our comments in standard face.

The H-TDMA data is given very little room and is not nearly used to its full potential in the paper. This is the biggest limitation of the paper.

We decided to give more information about the hygroscopic growth factors of the H-TDMA measurements. This includes an additional figure with 'nearly hydrophobic', 'less hygroscopic' and 'more hygroscopic' growth factors and number fractions for the three air masses A1, A2 and A3. We also extended Figure 07 with H-TDMA growth

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factors for the sampling periods I1, I2 and I3. According to these changes we will discuss in more detail the hygroscopic growth factors of the H-TDMA and their potential information. Please note that there is a separate paper published by Massling et al. (2009) discussing hygroscopic properties of the Beijing aerosol in detail.

The general comment concerning the figures is to use more colours instead of line and marker styles that are hard to distinguish.

We changed some figures and most of them are more coloured now to distinguish individual parameters.

Having a native English speaker read through and reformulate some of the text would benefit the paper as a whole.

Thank you for this hint, our experienced scientists will read the manuscript again.

Page 6895, line 20: "the deliquescence point of common inorganic salts."

The chosen RH ensures the exceedance of the deliquescence points of most common inorganic salts with atmospheric relevance.

Section 3.1: When calculating the DHGF, the authors state "We are aware that the assumption of homogeneously mixed particles is not true in atmospheric aerosols, since they tend to be externally mixed (Swietlicki et al., 2009). However, the procedure is assumed to yield trustworthy results as long as no dramatic changes occur in terms of external particle mixture with particle diameter." The H-TDMA is the best instrument to measure the mixing state of the aerosol. Yet the authors do not use, or even mention, this information. If they really want to make a good comparison, and try to explain differences between the three methods, this is certainly of great importance.

According to the additional figure (as mentioned before) with information about the 'nearly hydrophobic', 'less hygroscopic' and 'more hygroscopic' growth factors and corresponding number fractions measured by the H-TDMA, the mixing state of the particles will be explained and discussed in detail.

Section 3.2 If the data for the different hygroscopic groups are not presented in the

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paper, there is no need to first divide the data into three groups and then average them back. This is a completely redundant step and should be removed, both from the analysis itself, and from the paper since only serves to lengthen and complicate the text. After the change of some figures (especially Fig. 05 and 07) and the inclusion of a figure with 'nearly hydrophobic', 'less hygroscopic' and 'more hygroscopic' growth factors and number fractions of the H-TDMA measurements this section in the manuscript will find its authentication.

Page 6899, line 18: I assume that it is NH_4 and $[\text{2}^*\text{SO}_4 + \text{NO}_3]$ that correlate. Yes, you're right, thank you for the hint.

Page 6900, line 3: Add some motivation to why you select 1.7 as the average density of the particles. On line 22 you say this is "based on chemical data". Which chemical data? To me 1.7 sounds high for the overall density. How sensitive are your results to this number? And how much could you improve the result by varying the assumed $\text{GF}=1.0$ for the "non-soluble" fraction? It is known that oxygenated organics have a GF greater than 1.0

In general, it is difficult to make a decision concerning the particle density. In recent publications dealing with measurements in Beijing (Wehner et al. (2004, 2008), Wu et al. (2008)) a particle density of 1.7 g cm⁻³ was used.

Additionally, a study published by Pitz et al. (2008) presents the particle density in urban air in a German city. The particle density was studied in a one-year-period. A strong day-to-day-variation of the density for PM_{2.5} was found (1.05 to 2.36 g cm⁻³). The annual means for the years 2005 and 2006 were 1.6 and 1.7 g cm⁻³, respectively. Based on your comment concerning the density (1.7 g cm⁻³) and the growth factor for the insoluble fraction of the particles ($\text{GF}_{\text{insol}}=1.0$) a small sensitivity study was performed. A lower particle density (1.5 g cm⁻³) was assumed and a higher growth factor for the insoluble fraction (1.2) was used. As we wrote in the manuscript the calculated hygroscopic growth factors are strongly dependent on the chemical substances and the order of these substances. This is also the case for other particle densities and a

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higher growth factor for the insoluble fraction. For the main cases the chosen particle density of 1.5 g cm⁻³ ($\text{GF}_{\text{insol}}=1.0$) generates too low hygroscopic growth factors and a particle density of 1.5 g cm⁻³ and a GF_{insol} of 1.2 generates too high hygroscopic growth factors compared to our results in the current paper version. It should be noted, that oxygenated organics were only observed to have very small GFs at 90% RH and rather more start their growing process above 90% RH. The decision of using a growth factor of 1.0 for the nearly hydrophobic group is based on previous measurements of different authors (Weingartner et al., 1997): Massling et al., 2005). Recent H-TDMA measurements had shown a very low growth factor of particles with a high fraction of organic matter (Petters and Kreidenweis, 2007).

In general, there are many unknown parameters, which will hopefully be clarified in future studies. The solubility model which we used is - as we mentioned - based on a very simple approach to determine hygroscopic growth factors when only bulk chemistry is available.

Page 6901, line 2 and page 6902, line 4: I realize that the measurements were conducted in one of the most polluted places on the planet, but I suggest to reformulate "relatively clean" describing the air masses, since 200 $\mu\text{g m}^{-3}$ can not be considered clean, even though it is the least polluted period of your measurements

You are absolutely right, when you notice, that 200 $\mu\text{g m}^{-3}$ is not clean, but we think it is obvious that the notation "relatively clean" in that case just means, that it is cleaner than during the other periods. But we will rename it, to make it clearer.

Page 6901, line 12: Was the max. wind really as low as 0.12 m s⁻¹? This is not the case according to figure 1.

Thank you very much for that hint, the maximum wind speed for the whole period A1 was 7.7 m s⁻¹, during the days (6:00-18:00) a maximum wind speed of 7.7 m s⁻¹ (mean: 3.8 m s⁻¹) was reached and during the nights (18:00-6:00) the maximum wind speed was 5.5 m s⁻¹ (mean: 2.7 m s⁻¹).

I changed it in the text at follows:

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During daytime, the wind speed was higher as compared to nighttimes (maximum day: 7.7 m s⁻¹; maximum night: 5.5 m s⁻¹). Due to calm winds, the accumulation of urban aerosol was thus greatest during nighttimes. The particle mass concentration was highest during the last two nights.

Page 6902, line 14: remove "events"

Ok. This is done in the future version of the manuscript.

Page 6902, line 24: I only count nine. In figure 7 I2, there are two data points below 100 nm and in figure 3 you have the lowest range at 100 nm. Please explain/correct.

Thank you very much; you are right there are only nine stages. We corrected the data points in figure 7 (I2).

Table 2. All the information in this table is found in figure 6, and thus table 2 can be removed.

Yes, you're right. The correlation coefficient R can be read from Figure 6.

Figures 1, 5 and 7. The plots are hard to read. Use more colors to make it easier to distinguish between data.

We changed Figure 1 and 7 and did it more colored. According to Figure 5, we think the lines and symbols are easy to distinguish.

Fig. 1. Also add the times of the impactor samples.

The impactor sampling periods are already illustrated in Figure 1 (at the top, together with the three air masses (A1, A2, A3)).

Fig. 5. In A3 the DHGF below 40 nm is higher for 55% than for 77%. This should be noted in the next, and explained. If nothing else, it is a good marker for the uncertainty in the summation technique or the error in RH. The last sentence says that "error bars indicate the measurement errors." In the text these are explained as the uncertainty based on RH variability. These are not the same thing and should be clarified.

A study was performed and published describing the uncertainty of the summation

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method (Birmili et al. (2009)). Here, the sensitivity of the summation method (SM) was tested and a typical uncertainty of 0.05 for particles of $D_p = 70$ nm in diameter was found. It was found that the uncertainty increases with decreasing particle diameter (diameter: 30 nm \rightarrow uncertainty: 0.1) and that it is strongly connected with the experimental uncertainty. There will be more detailed information on this issue in the future manuscript.

Page 6906, line 6-8. The authors have presented no data on GF of less or more hygroscopic particles, and thus this statement can not be verified. Only if all your particles were classified as "more hygroscopic" would this statement be correct. If there is some mix of less or non-hygroscopic material in the particles, the GF might even seem high. The most reliable measurements are still the H-TDMA data, and those are around 1.4.

According to changes of additional information on the H-TDMA part in this manuscript we will make this section more clear.

Page 6909, line 20: Also spell out the C in CHGF.

Ok, its done in the new manuscript version.

As mentioned, to improve the paper, have a native English speaker read through it. Currently the language is not very good, whether it be due to the authors' ability to write in English or only due to careless finishing. Below are some grammatical corrections.

Page 6891, line 18: I expect you mean "rise".

The industrial rise in China has led to air pollution that has been acknowledged to greatly influence regional climate, and influences the worldwide level of air pollutants as well (Mage et al., 1996; Fenger, 1999).

Page 6891, line 27: "regionally emitted combustion particles"?

Bergin et al. (2001) suggested that the poor visibility may be due mainly to emissions from local and regional sources of combustion particles.

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Page 6900, line 22: of -> a

Figure 1 presents the particle mass size distribution and the total particle mass (PM1: $D_{aero} \leq 1 \mu\text{m}$) calculated by means of the particle number size distribution of dry particles (measured with TDMPs) and assuming a mean particle density of 1.7 g cm^{-3} based on chemical data.

Page 6905, line 10: "probably due to because of" should be corrected

For all three relative humidities, the growth factors of particles larger than 200nm are smaller, which is probably due to a lower fraction of soluble compounds in this size range.

Page 6906, line 24: "show a partly agreement" should be corrected

Generally, the DHGFs and the CHGFs only agree within a limited size range for the individual cases.

Page 6910, line 2: Remove "That".

The methods agree with respect to their trend in CHGF as a function of particle size, manifesting a maximum CHGF in the lower accumulation mode (100-300 nm), even if the absolute values are not matched.