

Anonymous Referee #2

Comment:

The work published by Pan et al. reports observational results from the hygroscopic behaviors of atmospheric aerosol in the vicinity of Beijing mega-city. The dependence of the aerosol scattering coefficient on relative humidity is investigated, and potential links to the aerosol source region and to aerosol chemical composition are studied. The impact of aerosol chemistry and the resulting hygroscopic behaviors and particle optical properties still requires a broadening of the experimental data base although research started in the early 1970s and 1980s (Covert et al., 1972; Fenn et al., 1981).

The paper reports results from multi-stage impactor analyses on particle chemical composition restricted to a particle size range PM 2.1. Aerosol scattering properties are reported from a humidified nephelometer operated at RH = 80%. The nephelometer sampled from the total aerosol (PM 11). The authors used the term aerosol hygroscopic growth factors although they analyzed modifications of the aerosol scattering coefficient with humidity. A more precise terminology is required here. Furthermore, a more detailed description of the experimental procedure is needed.

Aerosol humidification function values for the aerosol scattering coefficient vary from values reported in the literature. The authors identified this deviation without giving potential explanations. In particular, high values of the order of 2.2 were found for two episodes which are assumed to have a strong urban aerosol impact, however without showing higher ratios of organic matter to ammonium sulfate. Such high humidity growth function values at RH = 80% are neither found in laboratory and field studies on aerosol hygroscopic growth (McFiggans et al., 2006; Gysel et al., 2007; Massling et al., 2007), nor in a recent numerical analysis of humidification effects on aerosol optical properties in the Pearl River delta in China (Cheng et al., 2008). The authors should revisit this discrepancy.

A general difficulty concerning this publication is the fact, that chemical composition and aerosol scattering coefficients are reported for completely different size ranges which makes a consistent data interpretation almost impossible. Following Table 1, mass concentrations between PM 2.1 and PM 11 varied by a

factor of 2.6 on average, and by a factor of 3.9 at maximum.

In Table 5, the authors expand the relationship between chemical composition and aerosol light scattering. This table requires further explanation because in particular the column on PM 2.1 / PM 11 is found confusing. In general, no significant correlation is found between composition and optical properties modification by humidity growth.

In its current form, the results presented in the paper are not very valuable. The major limitation arises from the fact that chemical composition and aerosol optical properties are reported for different particle size ranges. Furthermore, no particle size distribution information is reported although it should be available from the impactor data. Having at least the mass size distribution available for different chemical species should allow performing model calculations on the expected variation of aerosol scattering with relative humidity based on the chemical composition. The authors are encouraged to conduct this step of theoretical interpretation of observational data in order to make their case stronger.

The authors thank you for your precious contribution for the paper improvement. Your comments could be divided into four parts, As follows:

Part 1:

A more precise terminology is required here, and a more detailed description of the experimental procedure is needed.

Replay:

Great thanks for your precious advises. As you suggested, more precise terminological expression of aerosol hygroscopic growth should be used, for instance, the “hygroscopic growth factor of aerosol scattering coefficient $f(RH)$ ” will be utilized instead of hygroscopic growth factor in order to avoid confusing $f(RH)$ and $g(RH)$. In some previous papers, the light wavelength information was also involved in the hygroscopic growth factor of scattering, as $f(80\%, 525nm)$, this will be corrected in the manuscript. And detailed descriptions of experimental procedures are added in the manuscript.

Part 2:

Aerosol humidification function values for the aerosol scattering coefficient vary from values reported in the literature. The authors identified this deviation without giving potential explanations.

Replay:

Yes, you are right. In the revised manuscript, we have compared our findings with many previous works in order to make insights into aerosols scattering humidification function variations.

Part 3:

In particular, high values of the order of 2.2 were found for two episodes which are assumed to have a strong urban aerosol impact, however without showing higher ratios of organic matter to ammonium sulfate. Such high humidity growth function values at RH = 80% are neither found in laboratory and field studies on aerosol hygroscopic growth (McFiggans et al., 2006; Gysel et al., 2007; Massling et al., 2007), nor in a recent numerical analysis of humidification effects on aerosol optical properties in the Pearl River delta in China (Cheng et al., 2008). The authors should revisit this discrepancy.

Replay:

Thank you so much for this remark. During the experiment, we have applied strictly the quality control procedure (SOP) to the operation of the instrument. The zero check was done automatically by pumping in particle-free air once each day, and weekly span check was performed manually using pure HFC-134a gas. The results of the zero/span check indicated that the bias for zero check was less than 2 Mm⁻¹, and less than 5% for span check. There was no evidence indicating that the nephelometer doesn't work well on this day (May 15th). In addition, the mean $f(\text{RH}<40\%)$ for these days was around 0.99 ~ 1.01, and the constructed scattering coefficient based on aerosol compositions could account for about 88% of measured ones for May 15th. All indicated the two nephelometers were in good conditions and the results were reliable.

Therefore, we agree that it's difficult to explain such high hygroscopic properties of aerosol in terms of the fraction of aged organic matters, as mentioned in the

previous manuscript. Nevertheless, such a phenomenon was also found in previous works (Day et al., 2000; Kotchenruther et al., 1999), and seemed to be due to mixing aerosols process (Choi et al., 2002; Cruz et al., 2000; Hameri et al., 2002; Huang et al., 2005; Saxena et al., 1995), further discussions about this strong hygroscopicity will be conducted in the revised manuscript.

Part 4:

Chemical composition and aerosol scattering coefficients are reported for completely different size ranges which makes a consistent data interpretation almost impossible. The major limitation arises from the fact that chemical composition and aerosol optical properties are reported for different particle size ranges. Furthermore, no particle size distribution information is reported although it should be available from the impactor data. Having at least the mass size distribution available for different chemical species should allow performing model calculations on the expected variation of aerosol scattering with relative humidity based on the chemical composition.

Replay:

We agree with you. In the preview manuscript, we did not provide complete information concerning aerosols chemical composition and scattering coefficients for data consistent interpretation. This has been corrected in the revised paper, and we provide information about particles size distribution and model results. We performed modeling experiment by using **IMPROVE equation and Mie theory**.

Reference:

- Choi M Y and Chan C K 2002. The effects of organic species on the hygroscopic behaviors of inorganic aerosols. *Environ. Sci. Technol.*[J] 36(11): 2422-2428.
- Cruz C N and Pandis S N 2000. Deliquescence and Hygroscopic Growth of Mixed Inorganic? Organic Atmospheric Aerosol. *Environ. Sci. Technol.*[J] 34(20): 4313-4319.
- Day D E, Malm W C and Kreidenweis S M 2000. Aerosol light scattering measurements as a function of relative humidity. *J Air Waste Manag Assoc.*[J] 50(5): 710-6.

- Hameri K, Charlson R and Hansson H C 2002. Hygroscopic properties of mixed ammonium sulfate and carboxylic acids particles. *AIChE Journal*. [J] 48(6).
- Huang X F, Hu M, He L Y, et al. 2005. Chemical characterization of water-soluble organic acids in PM_{2.5} in Beijing, China. *Atmospheric Environment*. [J] 39(16): 2819-2827.
- Kotchenruther R A, Hobbs P V and Hegg D A 1999. Humidification factors for atmospheric aerosols off the mid-Atlantic coast of the United States. *Journal of Geophysical Research*. [J] 104(D2): 2239-2252.
- Saxena P, Hildemann L M, McMurry P H, et al. 1995. Organics alter hygroscopic behavior of atmospheric particles. *Journal of Geophysical Research-Atmospheres*. [J] 100(D9).