Anonymous Referee #1

Comment:

This paper examines the relationship between f(RH) determined from RH-controlled nephelometer measurements and attempts to relate these to aerosol chemical composition and transport pathways.

"Hygroscopic growth factors" are referred to in the title and various places in the paper. Hygroscopic growth factors [D(rh)/(D(dry)] were not measured. What was measured is f(RH) - [Bsp(rh)/Bsp(dry)]. References to hygroscopic growth factors should be removed and replaced with f(RH), which is enhancement of Bsp due to hygroscopic growth.

There is also a reference to deliquescence RH on p. 5095. There is no clear evidence for deliquescence in Fig. 4 and the conclusions state that f(RH) varied smoothly and monotonically.

There is a fundamental deficiency in the experimental design in that the nephelometers measured TSP while the chemistry presented represents PM2.1. Table 5 shows that the PM2.1/PM11 ratio was 50% or less. Thus, the f(RH) must be depressed by the presence of a significant mass of large particles. The results are thus inconsistent with those of Day and Malm, for example, who used nephelometers preceded by size selective inlets.

It is difficult for the authors to obtain quantitative light scattering closure based on measured f(RH) and PM2.1 chemical composition. Because of the large coarse composition, the relationship between f(RH) and chemistry is more qualitative than quantitative. I suggest that the authors attempt to do do some closure calculations to make the results more quantitative. They could used dry scattering efficiencies used in the IMPROVE equation, i.e., 3 m2/g for ammonium sulfate and ammonium nitrate, 4 for organic carbon mass, 1 m2/g for fine dust, and 0.6 m2/g for coarse material > 2.1 um. The authors routinely use a factor of 1.4 to convert OC to OMC. This may be appropriate for fresh urban emissions but aged OMC should display a higher ratio, e.g., 1.8 (Pitchford et al., 2007, AWMA, 57, 1326-1336). Closure could be obtained if particles larger than 2.1 um are composed of non-hygroscopic dust with f(RH) = 1. The case would be difficult for periods where there was a high proportion of coarse sea salt, which is very hygroscopic. The authors would have had a much better opportunity to explain f(RH) in terms of chemical composition had they used a size-selective inlet with their nephelometers.

The authors attempt to relate some cases of high f(RH)>2 with high proportions of aged OMC. While authors such as Dinar et al. and Gysel et al. have shown that ambient organics are

hygroscopic, the measured growth factors were never high enough to produce an f(RH)>2.

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

Part 1:

"Hygroscopic growth factors" are referred to in the title and various places in the paper. Hygroscopic growth factors [D(rh)/(D(dry)] were not measured. What was measured is f(RH) - [Bsp(rh)/Bsp(dry)]. References to hygroscopic growth factors should be removed and replaced with f(RH), which is enhancement of Bsp due to hygroscopic growth.

Reply:

We greatly thank you for this important remark. We recognized the difference between the "Hygroscopic growth factors" g(RH)= [D(rh)/(D(dry)] and f(RH)= [Bsp(rh)/Bsp(dry)], and have replaced the term "hygroscopic growth factors" by hygroscopic growth factor of aerosol scattering coefficient f(RH) in the latest revised manuscript accordingly.

Part 2:

There is also a reference to deliquescence RH on p.5095. There is no clear evidence for deliquescence in Fig. 4 and the conclusions state that f(RH) varied smoothly and monotonically.

Reply:

As the referee pointed out, it was inappropriate to use the word "deliquescent RH" because the fitting f(RH) curves did not show clear "leap feature" behavior during aerosols growth, this has been corrected in the revised manuscript.

Part 3:

There is a fundamental deficiency in the experimental design in that the nephelometers measured TSP while the chemistry presented represents PM2.1. Table 5 shows that the PM2.1/PM11 ratio was 50% or less. Thus, the f(RH) must be depressed by the presence of a signifiant mass of large particles. The results are thus inconsistent with those of Day and Malm. I suggest that the authors attempt to do some closure calculations to make the results more quantitative. Closure could be obtained if particles larger than 2.1 um are composed of non-hygroscopic dust with f(RH) = 1.The case would be difficult for periods where there was a

high proportion of coarse sea salt, which is very hygroscopic.

Reply:

We agree to the referee that relationship between f(RH) of TSP and PM2.1 chemical composition is more qualitative than quantitative in the present of large portion of coarse composition. According to your suggestion, we performed scattering closure calculations using both IMPROVE equation and Mie scattering theory coupled with size-segregated aerosol chemical compositions. Furthermore, according to calculations, we found that the influence of sea salt was weak. This confirmed your viewpoint. The results will be added into the manuscript.

Part 4:

The authors attempt to relate some cases of high f(RH)>2 with high proportions of aged OMC. While authors such as Dinar et al. and Gysel et al. have shown that ambient organics are hygroscopic, the measured growth factors were never high enough to produce an f(RH)>2.

Reply:

Thank you so much for this remark. For proper design control 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(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. 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 conducted in the revised manuscript.

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 PM2. 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).