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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of relative humidity and particles size distribution on aerosol light extinction in urban area of Guangzhou" *by* Z. J. Lin et al.

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First of all, we would thank all anonymous referees for their valuable opinions and suggestions for the manuscript. With a month effort, some modifications and corrections have been made in the revised manuscript. Main features are: a. ISORROPIA II (Fountoukis, C. and Nenes, A., 2007) has been applied to determine the associations among ions and the mass of water uptake at a specific RH. Furthermore, EORI and EGF required by the Mie Model have been re-calculated accordingly. b. The particles number concentration measured by APS and the light scattering coefficient measured by Nephelometer have been averaged into daily values in order to be compatible with

PM2.5 sample. c. The aerosol optical properties have been simulated at the same RH as the one recorded inside Nephelometer. The comparison between modeled result and Nephelometer measurement serves as an indicator of the extent the practical method "captures" the aerosol light scattering coefficient of the real atmosphere. d. The size distribution and hygroscopicity of modeled optical properties as well as the relevant seasonal variation are discussed. The uncertainties of the measurement and modeling are stated.

Reply to Anonymous Referee #1

1. In this study, PM2.5 was sampled by quartz filter, which might react with water soluble ions of PM. Furthermore, all quartz filters have artifacts and the blank might also be high. How artifacts are handled in this study and what are the blank for major water soluble ions in the quartz filter?

Reply: We concur that there are artifacts when using quartz filter for PM2.5 sampling. However, the high loading of PM2.5 in Guangzhou can block the Teflon filter easily, which can affect the flow rate of samplers and increase the sampling errors. Moreover, there are still some previous studies (Shen, et al., 2009; Wang et al., 2011) in China using the quartz filter for the similar reasons. In this study, the field blanks were determined and the average values of 12 blank filters of Na+, NH4+, K+, Mg2+, Ca2+, F-, Cl-, NO3-, and SO42- were 0.671$\pm$0.091, 0.002$\pm$0.002, 0.005$\pm$0.006, 0.006$\pm$0.007, 0.052$\pm$0.064, 0.168$\pm$0.036, 0.425$\pm$0.094, 0.077$\pm$0.096 and 0.362$\pm$0.082 mg L-1, respectively. Although the blank value of Na+, F-, Cl- and Ca2+ were slightly higher than other species, blank filter was collected every 10 samples and the blank values were quite stable. All results in this study were blank subtracted. Moreover, the values of ambient samples were significantly higher than the blank value, which can reduce the errors.

2. The uncertainties of all measurements were not reported in the manuscript although it is very important for modeling aerosol optical properties based on measurements. It

would be nice for the authors to add one table to list all uncertainties of individual measurement techniques. The authors are also encouraged to discuss the uncertainties of model results as consequence of all measurement uncertainties.

Reply: We agree that the uncertainties of measurements are important for modeling. Further, in current study, the uncertainties of PM2.5 sampling, Ion & OC/EC analysis, Nephelometer, APS, Ambient RH detection are 1.08%, 2.49%, 2.30%, 4.46% and 3.87%, respectively. Accordingly, the modeled aerosol optical property has a total uncertainty of 8.74% which are calculated following the method of merging uncertainties recommended by IPCC guideline.

3. The RH inside the Nephelometer was monitored. What is the measured result? In line 22 of page 15650, it seems that the authors considered the aerosol to be completely dried out, probably this is rarely the case. Typically, the RH inside the Nephelometer will be smaller than the ambient RH. How do the authors convert the RH inside the Nephelometer to the ambient RH under which bsp, bap, bep, and bw0 were discussed?

Reply: The RH inside the Nephelometer was recorded and was between 17% and 73%. The modeled bsp, bap, bep and bω0 have been re-calculated at that RH condition. Furthermore, the model result is compared to the total scattering coefficient measured by the Nephelometer.

4. The fg,j values for each species is from the literature. Figure 4 showed that some sorts of assumption had to be made concerning a smoothing of the hysteresis curves. What decisions are made concerning using the deliquescent or crystallization branch of the hysteresis curves?

Reply: This point will no longer affect the article as the previous method has been substituted by ISORROPIA II in estimating the hygroscopic growth of chemical species for the revised manuscript.

5. Line 3 in page 15647, "Since POM, EC, and other unidentiïňĄed components were considered to have no hygroscopic growth. . .", what is the literature for reference of this statement ? Generally, water soluble organic carbon accounts for 50% of the total organic carbon although its hygroscopic ability is smaller than inorganic salt. It would be nice to take the hygroscopic growth of POM into consideration in the model.

Reply: We agree that this statement had not been properly addressed in the last manuscript. Although we continue to follow the assumption (the hygroscopicity of POM is not considered in the model), we provide supporting reasons for the revised manuscript, which are: (1) As a whole, the hygroscopic growth of SOA (Secondary Organic Aerosol) was around 1.2 at 90% RH (Gysel et al., 2007; Stock et al., 2011); (2) The hygroscopicity of some extracts from WSOC was recognized (Gysel et al., 2004). However, there was no WSOC speciation in present study. (3) The water uptake by the aged organic aerosol only accounted for a few percent of total water uptake (Bougiatioti et al., 2009; Engelhart et al., 2011). (4) Not like those inorganic salts, a more accurate RH dependence of POM has not been well established. (5) The POM has not been included in the ISORROPIA II model yet.

6. The mass of uptake water was estimated by Eq.(1). Please give the reference literature for Eq.(1).

Reply: The mass of water uptake was estimated by ISORROPIA II instead of the Eq.1.

7. Is water included in the j-th component in Eq.(2) and (3) ? If so, what is the value of fg,j for water component? How does the number concentration of water determined in Eq.(5)?

Reply: (1) Water is included in Eq.2 and 3. The relevant values for water were listed in Table 2, which included density, real and imaginary part of optical refractive index. (2) “Internal mixture” was assumed, where it was considered all chemical components mixed into every single particle whose number concentration was measured by APS.

8. Line 6 in page 15650, "it is able to estimate bsp, pm0.5-2.5 by the Mie model based on the EORI, EGF of PM2.5 and Nj,pm0.5-2.5 from APS measurement." This statement is based on the assumption that the chemical components and their mass fraction of PM0.5-2.5 are entirely consistent with that of PM2.5. But, the size distribution of chemical components varies greatly, especially for ultra-fine particles and fine particles. Furthermore, the chemical species was measured with low resolution (23.5h). But, the modeling is with high resolution of 1 hour. How do the authors handle the dataset from low resolution to high resolution? The uncertainties of the assumption in this study should be evaluated.

Reply: We agree that the size distribution of chemical component varies for ultra-fine and fine particles. However, in the current study, it was a trial to work out a practical method to simulate the optical properties of PM0.5-2.5 just with the APS measurement and PM2.5 sampling. Moreover, in the revised manuscript, the particles number concentration measured by APS and the scattering coefficient measured by Nephelometer have been averaged to daily values in order to be compatible with PM2.5 sample.

9. Line 18 in page 15651, "Regardless of the difference in chemical composition. . .". As the question (7), the estimation of bap,pm2.5-20 (bsp,pm2.5-20) should take the size distribution of elemental carbon (sulfate, nitrate, ammonium, . . .) into consideration.

Reply: “Internal mixture” was assumed, where it was considered all chemical components mixed into every single particle whose number concentration was measured by APS.

10. Line 24 in page 15653, the number of 0.87 should be 0.86 according to Figure 5.

Reply: The correlation coefficient has been corrected for the revised manuscript.

11. Line 9 in page 15654, "hbap,pm0.5-2.5 fluctuated around 1 when RH increased from 37% to 66%, and then began to drop."When the aerosol is assumed to be internal mixing, the “focus effect” of light absorption increases with the RH increasing and the

hbap,pm0.5-2.5 should also increase, but the value of hbap,pm0.5-2.5 began to drop with the RH increasing in this study.

Reply: The RH dependence of bap was once discussed in some previous studies (Bohren and Huffman, 1998; Redemann et al., 2001; Nessler et al., 2005; Cheng et al., 2008). On one hand, the hygroscopic growth of aerosol enhances the “focusing effect” and tends to amplify the bap. On the other hand, the EORI being lowered due to the increasing amount of water uptake tends to lower bap. Since ISORROPIA II was used to calculate the mass of water uptake in the revised manuscript, the RH dependence of the absorption coefficient has been assessed again, the result of which shares some similarity with a previous literature (Cheng et al., 2008.)

12. It would be nice to add a figure illustrating the temporal series of mass concentrations for each species and each size bins based on PM2.5 data.

Reply: Temporal variations of mass concentrations of chemical components of PM2.5 are illustrated in the revised manuscript.

13. In figure 5, it should illustrate the value of the measured and modeled bsp, not the standardized bsp.

Reply: It has been modified for the revised manuscript

14. In figure 7, the title of y axis should be "fraction of scattering coefficient" and "fraction of absorption coefficient".

Reply: It has been modified for the revised manuscript

15. How to estimate the optical properties of PM1.0? The method could not be found in the manuscript, but the result was illustrated in figure 7.

Reply: This point has been clarified in the revised manuscript. Further, the size distribution of optical properties of PM0.5-20 is illustrated instead.

16. The optical parameters of possible chemical components were listed in table 2,

but, which components are used in the Mie model? For example, what is the chemical form of sulfate in the calculation of bsp. If the chemical form of sulfate is (NH4)2SO4, the reason for this argument should be clarified.

Reply: Since ISORROPIA II model is being applied in the revised manuscript, the chemical form of every identified component in PM2.5 will be clarified with examples: NH4HSO4, (NH4)2SO4, NaHSO4, Na2SO4, NH4NO3, NaNO3, NH4Cl, NaCl, K2SO4, MgSO4, CaSO4 and H2SO4.

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Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/12/C7539/2012/acpd-12-C7539-2012-supplement.pdf

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