

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.

Anonymous Referee #3

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The manuscript discussed the impact of relative humidity on aerosol optical properties, including light scattering and absorption etc through Mie simulations based on the measured aerosol chemical composition and number size distributions. The contribution to light extinction by different aerosol compositions and different size ranges were analyzed for multiple seasons, which could be interesting to readers of ACP. However, the overall data analyses, discussion and arguments are not strong. I would suggest it for publication only if the major concerns from reviewers have been sufficiently addressed.

Major Comments:

1. Uncertainties

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Besides the measurement uncertainties, there are several assumptions made in this study which will impose uncertainties to the simulation of optical properties. These uncertainties should be quantified or at least clearly stated/described in the manuscript.

If I understand right, for each PM_{2.5} sampling period, a constant aerosol chemical composition (derived from the filter analysis) has been assumed for the entire particle size range (0.5–20 micrometer). The refractive index, hygroscopic growth and density etc were all calculated based on this assumption. Uncertainties due to this assumption should be discussed, e.g., in the calculation of the light absorption (as discussed in major comment #2).

Besides, to derive bsp,PM_{0.5} and bap,PM_{0.5}, the author basically used a subtraction method which is subject to uncertainties in nephelometer measurements, angular correction, Mie simulation of aerosol optical properties in the size range of 0.5 to 2.5 micrometer, as well as in the simple empirical parameterization of light absorption by PM_{2.5} mass. Given the fact that mass fraction and scattering contribution of PM_{0.5} are both dominate components in the upper panels of Figure 7, the uncertainties of derived scattering and absorption, as well as the mass of PM_{0.5} need to be carefully discussed.

2. Effect of particle size distribution

Beside relative humidity, the manuscript is also titled by the impact of particle size distribution on aerosol light extinction. However, the discussions about the size effects are limited. I would suggest the authors to show at least the monthly averaged particle number size distributions in different seasons to assist the discussion. Or the authors could try higher size-resolution. For example, by using the number size distribution measured by APS, the authors may also calculate and show the size distribution of esp and eap. In addition, Fig. 8 (p15655, l6) is missing, or do the authors mean the lower two panels of Figure 7?

As discussed above, for each PM_{2.5} sampling period, a constant ratio of EC to to-

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tal particles (+water, according to the chemical composition analysis from the PM2.5 samples) has been assumed for the entire size distribution in this study. This is rarely the case especially for the large particles. I do not think the authors have adequate or enough solid evidence to support the statement that "large particles were more efficient in light absorption" (line 6-7 in page 15655).

3. Comparison between Mie simulation and nephelometer observation

The scattering coefficient measured by nephelometer is the sum of the scattering coefficients of particles in the whole size range. The calculated scattering coefficients (by Mie model) in the range of 0.5 to 2.5 micrometer is a subset of total particle scattering (superset). A comparison between the superset and the subset (as in Fig 5) can NOT serve as a validation of the Mie model simulation. Especially when looking at the upper panels in Figure 7, the extinction of particles smaller than 0.5 (a subtraction between nephelometer observation and Mie simulation) dominate the total extinction most of the time. And the contribution by particles between 0.5 to 2.5 micrometers is in general small (according to Figure 7), except in April 09.

Comparison of the standardized values (in Figure 5) shows if the scattering coefficients of particles from 0.5 to 2.5 micrometer are in the same trend as the total particles scattering. Although good correlation does not mean the Mie calculation is validated, a rather low correlation coefficient as for Oct 09 also does not mean the simulation is poor. Actually, it might be interesting if the author could try to explore why they correlated well to each other sometimes but sometimes not. For example, different trends may due to a change in particle size distribution. The typical number size distribution from 3nm in Oct in Guangzhou may be found in literature. Also, one could also see that there is a significant change (increase) in the mass scattering coefficient (esp, lower panel in Figure7) in the late Oct 09 and early Jan 10 when the correlation is poor. More discussion is needed in this regard.

4. Mixing state of EC

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The authors discussed about the influence of aged air mass on the mass light absorption coefficient around line 10-20 in page 15655. From my point of view, for a certain size distribution, the light absorption is determined by the refractive index when using Mie model. In the current study, the refractive index is calculated under the assumption of internal mixed aerosol particles. Under such assumption, a lower single scattering albedo either means that the EC mass fraction is high, or the mass fraction of chemical compositions with higher real part of refractive index (e.g., Na₂SO₄, MgSO₄ etc) is relatively low, or the particle number size distribution is different. But it can NOT imply any information about the mixing state of EC, since the basic assumption of the entire calculate (optical, refractive index, density etc) is the aerosol particles are completely internally mixed.

Specific comments:

1. The introduction needs to be re-organized and be more specific and focusing.
2. What is the sampling relative humidity of PM2.5? How did you control the sampling and chemical analysis relative humidity at 40% as in Table 1?
3. Quartz filter is usually used for thermal EC/OC analyses. To determine the PM2.5 mass and aerosol chemical composition, Teflon filter would be much better than Quartz filter. How much uncertainties would be induced by using the Quartz filter to serve this purpose?
4. The relative humidities at ambient, dry or measurement conditions have not been clearly described in this manuscript. For example, what is the sampling relative humidity in nephelometer (more specific than lower than 70%)? Did the author treat the aerosols inside nephelometer as dry particles, or also calculated the scattering coefficient according to the instrument-recorded relative humidity inside the nephelometer when using Eq. 14? Are data in Figure 3 at ambient conditions?
5. Seasonality is the highlight of this manuscript. I suggest the authors discuss more

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about the seasonal variations in chemical composition, hygroscopicity, and optical properties.

6. After getting the reconstructed compositions, the authors could use the AIM model (<http://www.aim.env.uea.ac.uk/aim/aim.php>) to calculate the corresponding ions (ionsAIM). Comparison between ionsAIM and measured ions can be used to validate the reconstructed chemical compositions.

7. Most of the data reported in the manuscript are either standardized or as a proportion ratio (fraction). No real measurement data were presented (neither absolute concentration of different chemical species nor nephelometer measured scattering and back scattering coefficients). I suggest presenting these data in tables or figures, which would be interesting and valuable for the community.

Technical comments:

1. The English in the manuscript is not satisfying and need to be carefully revised.
2. Please rephrase the sentence "Since socioeconomic developed in recent years, . . ." (Line 23 in page 15641).
3. "Seinfeld and Pandis" instead of "Seinfeld and Spyros" (e.g., Line 3 in page 15641 and several other places).
4. It is hard for me to distinguish the colors of Mg²⁺ and NO₃⁻ in figure 2.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 15639, 2012.