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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol optical properties in a rural environment near the mega-city Guangzhou, China: implications for regional air pollution and radiative forcing" *by* R. M. Garland et al.

Anonymous Referee #1

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Title: Aerosol optical properties in a rural environment near the mega-city Guangzhou, China: implications for regional air pollution and radiative forcing

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Atmospheric Chemistry and Physics Discussion

General Comments:

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Interactive Discussion
Discussion Paper

This manuscript is well written and presents a wealth of valuable data combined with insightful analysis. I found the analysis of the diurnal variation of aerosol scattering coefficients and absorption coefficients (Figure 9) to be particularly interesting and important for understanding the total atmospheric column radiative effects from ground-based particle sampling measurements. However, I am somewhat surprised (given the comprehensive suite of measurements and the rigorous analysis) that there was no data or analysis presented on the aerosol humidification effects on aerosol optical properties. There is extensive analysis of the dry single scattering albedo, but what is really needed for radiative forcing calculations are the ambient aerosol properties at the high relative humidity which occurs in southern China in July. It is expected that there would be a significant sulfate component to the aerosol in China (due to sulfur dioxide emissions from coal combustion) that would result in significant particle humidification growth, which would yield increases in both scattering and single scattering albedo. On another issue, I am not completely convinced that the heavy pollution episode of July 23-25 is due to biomass burning. I did not find compelling evidence to determine whether meteorology (stagnation) or additional source strength (fires) were more important in elevating the aerosol concentrations and additionally there was no mention of the role of coagulation in increasing fine mode particle size which would also increase the scattering coefficient and single scattering albedo. The rate of coagulation increases as particle concentration increases, and therefore must have played some role in the heavy pollution event. The manuscript would be improved with the addition of some discussion of the dynamics of the particle size distributions (especially the fine mode) and their effects on optical properties. On page 7 you state: -a detailed discussion of the particle size distributions will be presented elsewhere.- However some of this information needs to be summarized in the current paper and applied to the analysis.

I think this paper will make an important contribution to the literature, however I urge the authors to address the issues raised above in addition to responding to the specific comments listed below.

ACPD

8, S1842–S1847, 2008

Interactive Comment

Specific Comments:

Page 2: -The daytime average single scattering albedo of 0.87 appears- should be -The daytime average mid-visible single scattering albedo of 0.87 appears-

Page 9: -The overall accuracy of the PAS calibration in this study was within 10%, thus the overall accuracy of the measured PAS absorption coefficients are also ś10% (Schnaiter, 2005).- Please give an estimate of the uncertainty in the computed single scattering albedo based on all measurement uncertainties.

Page 13: Can you present some additional data/ analysis to strengthen your claim that biomass burning was the principal reason for the heavy aerosol loading on July 23-25, possibly satellite hot spot images?

Page 15: -The single scattering albedo increased during this time to 0.88, which is common in smoldering fires (Reid, 1998). The parameters determined during the smoky period can be considered to represent the optical properties of emissions from the burning of plant farming waste.- While it is true that the aerosol from smoldering combustion have less black carbon and therefore higher single scattering albedo (SSA), coagulation of fine mode particles under high concentrations (such as this pollution event) will also increase scattering and result in higher SSA. Some discussion of fine mode size distribution dynamics should be included here.

Page 16: -In order to convert to atmospheric relevant conditions, the hygroscopic growth of these aerosol particles and its impact on their light scattering and absorption would need to be quantified, which is beyond the scope of this manuscript.- However this is what is needed for accurate radiative forcing studies, which you address in Section 3.4. If measurements of hygroscopic growth were not made during PRIDE, then I would argue that you should not attempt to make the radiative forcing calculations since the analysis would be biases towards low values of SSA that are representative of dry aerosol only, not ambient atmospheric conditions.

ACPD

8, S1842–S1847, 2008

Page 21: -Figure 10 shows the correlation of SIGMAs,550 and SSA532 to local wind direction and local wind speed.- Should be: -Figure 10 shows the relationship of SIGMAs,550 and SSA532 to local wind direction and local wind speed.-

-No strong correlation was seen between SIGMAs,550 (and SIGMAa,532, though not shown) and local wind direction.- Please give the r2 value here to quantify the level of correlation.

Page 22: -Average SSA532 values (0.8-0.9) are not correlated with wind direction.- Again please give the r2 value. Also the following 2 sentences after this suggest that there is some correlation, making this paragraph confusing.

Page 22, Figure 11A: It is not typical to compute 24-hour back trajectories. Usually 5 to 7 day back trajectories are presented since aerosol residence time in the troposphere averages about 1 week. Please explain why you chose to compute only 24-hour trajectories.

Page 24: -In contrast, the highest SSA532 values were measured on 12 and 20 July when the air masses were slowly moving and hence residing longer over local continental sources, which resulted in elevated aerosol concentration (high extensive properties) and photochemical aging (high single scattering albedo).- Again some discussion of coagulation causing larger size fine mode particles, and thus increased scattering (thereby increasing SSA), is needed here.

Page 26: -Many studies have used the wavelength dependence of alpha to gain information on the size distribution of the particles (e.g., Eck et al., 1999; 2001; Schuster et al., 2006; Gobbi et al., 2007).- Note these studies have computed curvature from extinction not scattering AOD. Also, please add the ONeill et al. (2001) references.

ONeill, N. T., T. F. Eck, B. N. Holben, A. Smirnov, O. Dubovik, and A. Royer (2001), Bimodal size distribution influences on the variation of Angstrom derivatives in spectral and optical depth space, J. Geophys. Res., 106, 9787-9806.

ACPD

8, S1842–S1847, 2008

Page 27: -To our knowledge, the second derivative observed on 25 July is the highest reported for atmospheric aerosols. It is substantially larger than the second derivative values reported for biomass burning aerosol in Bolivia (alphae 2.09; Eck et al., 1999) and in Zambia (alphae$<$2.2; Eck et al., 2001).- Be careful, the 2nd derivative for scattering coefficient would be expected to be greater than for the extinction coefficient (similarly as derived from extinction AOD). See Eck et al. (2001) Figure 13a that shows curvature decreasing with increasing absorption. Also, you should compute the theoretical limits of curvature with Mie code, since it seems that your high value of 3.44 may exceed what is physically possible (likely $<$2.5). Measurement uncertainty (bias) perhaps contributed to the very high values you have inferred from data acquired during PRIDE.

-This suggests that the driving force for the curvature is not only the pollution level.- Eck et al. (2001) Figure 13b shows that the driving force behind the curvature is the radius of the fine mode size particles.

-Using other ground based techniques (i.e., sun photometers) it has been suggested that the main driving force of the curvature is the fine/coarse ratio of the size distribution (e.g., Eck et al., 1999; Schuster et al., 2006; Gobbi et al., 2007).- Although the fine/coarse ratio is important in determining the curvature, a factor that is even more important is the size of the fine mode particles.

Page 31: -Figure 17 displays SSA532 versus b550 for this campaign (including 23-25 July) with the color scale as local time.- Should be: -Figure 17 displays SSA532 versus b550 for this campaign (including 23-25 July) with the color scale as local date.-

Page 32: -Since SSA532 and b550 are negatively correlated (Fig. 17), they partially offset their respective radiative forcing as an increasing b550 indicates increased cooling while a decreasing SSA532 indicates increased warming.- Please be more specific: decreasing SSA results in increased warming of the aerosol layer, but increased cooling of the surface (due to decreased surface irradiance).

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8, S1842–S1847, 2008

Page 33: -Alternatively, as SSA532 decreases, at a fixed b550, the forcing efficiency increases, indicating increasing warming of the surface.- As SSA decreases the solar flux decreases rapidly, therefore resulting in surface cooling. Do you mean the aerosol layer warms as the SSA decreases?

Page 35: -Around periods of intense pollution there was curvature in the dependence of SIGMAs on wavelength, which caused a wavelength dependence of the Angstrom exponent. This curvature can be explained by the high proportion of aerosol volume in the fine fraction.- The large curvature is influenced even more by the size of the fine mode particle radius; the larger the submicron particle radius the larger the curvature.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 6845, 2008.