Response to comments of referee #1

General Comments:

Reviewer: *This work describes a new approach to estimate the mixing state of light absorbing carbonaceous (LAC) species based on optical and number size distribution data. The new algorithm was applied to the optical (hemispheric backscattering fractions) and number size distribution data obtained from a recent field campaign in the North China Plain. The model was carefully analyzed for possible error sources and the uncertainties in all the parameters were evaluated. A diurnal variation trend in the mixing state of LAC was observed and attributed to the evolution in the mixing layer. The method presented appears to be convincing and has the merit to determine the LAC mixing state with limited instruments and data, which may provide a better understanding on the impacts of LAC on global radiation budget. The manuscript is reasonably well written, addressed an important subject in atmospheric sciences, and can be considered for publication in ACP, provided that following issues are adequately addressed.*

Response: Thanks for the comments.

Major Comments:

Reviewer:

Atmospheric nanoparticles with diameter less than ~40 nm appear to contribute much less on the aerosol optical properties. As a result, when the fraction of these nanoparticles in number concentration is high, it may affect the cross section calculation. Is this factor considered in the model presented?

Response:

We thank the reviewer for this comment. For several reasons, we did not treat the nanoparticles separately in the model.

As mentioned by the reviewer, when the particle is much smaller than the wavelength of incident light, it is in the Rayleigh scattering regime. In this regime, the pattern of scattered light intensity is symmetrical in the forward and backward direction and more or less independent of particle shape (Seinfeld and Pandis, 1998). In the current study, the BHMie code and the BHCOAT code (Bohren and Huffman, 1983) was applied for all particles, including the particles from 3 nm to 40 nm. We did not treat Mie scattering and Rayleigh scattering regime separately, because the Mie theory can serve as the basis of a computational procedure to calculate the scattering and absorption of any sphere (Bohren and Huffman, 1983) and there is no large difference between treating them separately or not.

The assumed mixing states of small particles could also bring uncertainties. The fractions of core-shell structured particles were assumed to be the same for all particle size. Also the relative size of the core (the ratio of the core size to the particle size) was assumed to be the same. Under such condition, the LAC core in core-shell mixed nanoparticles might be very small (several nanometers or even less), which might be far from the real condition. However, as indicated by the referee, the contribution of those nanoparticles on aerosol optical properties would be negligible. Fig. 1 (upper panel) shows the scattering and absorption cross sections for particles with different diameters. For particles with diameter below 100 nm, scattering cross sections increase dramatically (from 1e-12 to 1e-2) with increasing diameters. For particles with diameter below 40 nm, scattering cross sections are exclusively smaller than 1e-5, in contrast to a magnitude of

1e-2 for particles in the accumulation mode. Fig. 1 (lower panel) displays the time series of number fractions for particles with diameter smaller than 40 nm during the HaChi summer campaign. The fraction of particles with diameter smaller than 40 nm varies over a range of 0.02 to 0.7. Generally, the scattering cross section of particles smaller than 40nm is several magnitudes smaller than that of the particles larger than 40nm. To reach an optical contribution of the same magnitude, the number concentration of particles smaller than 40nm has to be magnitudes larger, which was not found in the observations.

As stated above, the influence of particles with diameter below 40 nm on total optical properties could thereby be neglected, and has not been separately considered in this study.

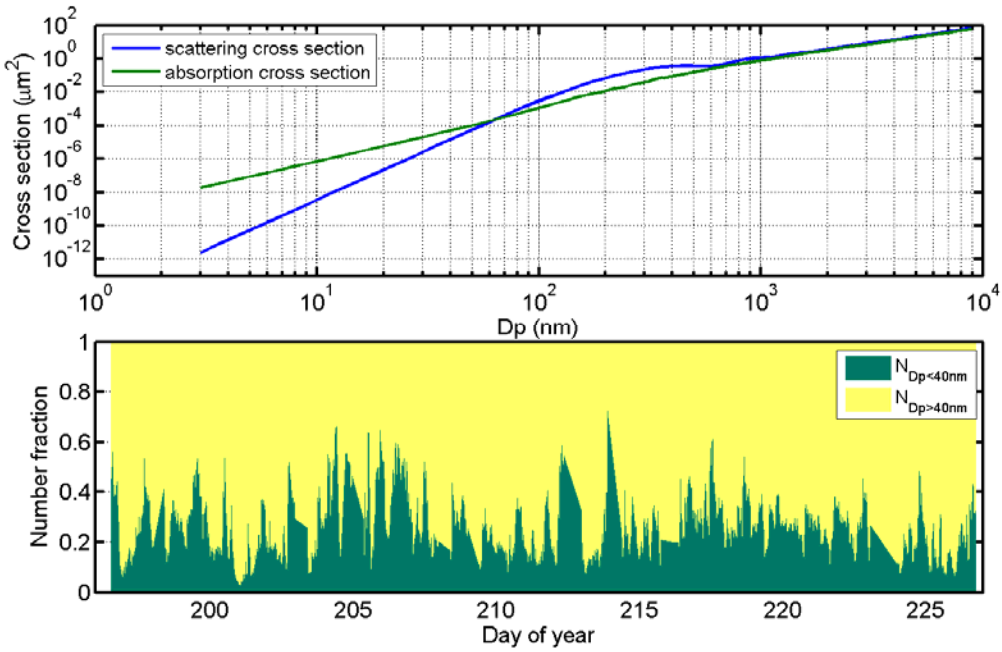

Fig. 1. Upper panel: The scattering and absorption cross section calculated from Mie theory for particles from 3 nm to 10 μm. The refractive index was assumed as 1.54-0.04i in the calculation. Lower panel: the time series of number fraction for particles with Dp<40 nm for the HaChi summer campaign.

Reviewer:

Several recent papers on the subject (Pagels et al., Aerosol Sci. Tech. 43, 629, 2009; Xue et al., Phys. Chem. Chem. Phys. 11, 7865, DOI:10.1039/b700001a, 2009; Xue et al., Environ. Sci. Technol. 43, 2787, 2009; Khalizov et al., J. Geophys. Res. 114, D05208, doi:10.1029/2008JD010595, 2009; Khalizov et al., J. Phys. Chem. A 2009, 113, 1066; Zhang et al., Proc. Natl. Acad. Sci. USA 105, 10291, 2008) and books (e.g., Mishchenko, M. I.; Hovenier, J. W.; Travis, J. W. Light Scattering by Nonspherical Particles: Theory, Measurements, and Applications. Academic Press, 2000) will be helpful for the analysis in section 4.4 related to the LAC shape and cross sections.

Response:

We do appreciate the valuable information the reviewer kindly provided. We have read them through. They are very helpful for our analysis, such as "And the optical properties of LAC are very sensitive to the mixing state (Khalizov et al., 2009)" in sect. 1 paragraph 3 and "Freshly generated soot exists in the form of chain-like aggregates. However, during aging processing, a

marked change in morphology of soot particles occurs. It was found that a transformation of the soot aggregates to spherical particles occurs as sulphuric acid and water condense onto fresh soot. Also, the soot core was restructured to more compact form (Zhang et al., 2008; Pagels et al., 2009)" in sect. 4.4.1 paragraph 2.

Other Comments:

Reviewer:

Page 27482 line 16-page 27483 line 5. It may be more appropriate to discuss the possible mixing state of LAC in the introduction part.

Response:

Thanks for this suggestion. We have merged the 3rd paragraph of sect. 3.1 with corresponding paragraphs in the introduction, and rephrased the 3rd paragraph of the introduction.

Reviewer:

Figure 6. What do the black dash lines stand for?

Response:

The black dashed lines stand for the corresponding uncertainty of the MAE (3σ). The description for figure 6 "The black line corresponds with MAE=6.6m^2g^{-1}. The error bar of each data…" has been revised as "The black line corresponds with MAE=6.6m^2 g^{-1}. The black dashed lines stand for the estimated uncertainty of the MAE (±3σ). The error bar of each data…"

References:

Bohren, C. F. and Huffman, D. R.: Absorption and Scattering of Light by Small Particles, John Wiley, Wiley, New York, 1983.

Seinfeld, J. and Pandis, S.: Atmospheric chemistry and physics: from air pollution to climate change, Wiley, Inc., New York, 1998.