

***Interactive comment on* “Quantification of black carbon mixing state from traffic: implications for aerosol optical properties” by M. D. Willis et al.**

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Response to Anonymous Referee # 2

The authors would like to thank Referee #2 for their comments on this manuscript, which have helped to improve the clarity of the paper. Our responses to specific comments and the corresponding changes to the manuscript are detailed below.

1. I like the style of this paper which is short and straightforward. But sometimes it is also inconvenient to always try to find things in supplement. Maybe the author can consider moving some important contents back to the main text.

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Authors' response: Sections 2, 3, 4 and 7 of the Supplement have been added to the main paper, and Figures 1, 2 and 3 have been modified to include data from both the roadside and non-roadside sites. The discussion and figure relating to particle coating thickness estimation have also been added to the main text of the paper.

2. Fig. 5: It seems the SSA derived with PASS-3 measurement is with very large uncertainty.

Authors' response: We acknowledge that the measurements of SSA at 405 nm have a large amount of scatter, especially for the HOA-rich plume example shown in the paper (part (b) of the relevant figure). Uncertainty in the SSA measurement is impacted by uncertainty in both the absorption and scattering measurements. In particular, the SSA measurement is associated with a larger degree of scatter when the black carbon (and total aerosol) loading is low. This relatively large degree of scatter will arise when the black carbon loading is low and the instrument is approaching its detection limit. Since the SSA measurements are used as a qualitative illustration only, we do not believe that this uncertainty impacts any of the conclusions made in this work.

3. In the box model simulation a constant mixing height was assumed. There are studies based on PartMC-MOSAIC simulation suggest the diurnal evolution of mixing layer plays a very important role in the variation of aerosol mixing state (e.g. Liu et al., 2011). I am wondering if you will have largely different result if the variation of mixing layer is switched on.

Authors' response: We thank the reviewer for raising this point. We changed the text in the paper (i.e., text that was previously in Section 7 of the Supplement, and has been moved to the main paper) as follows: "The initial gas phase concentrations are the same as in Zaveri et al. (2010). In contrast to Zaveri et al. (2010), we prescribe a constant mixing height (400 m), relative humidity (70%) and temperature (298.15 K) to simplify the interpretation of the results. Since the entrainment of background aerosol

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can modify the aerosol mixing state substantially (Liu et al., 2011), we include constant dilution with the background at a rate of $1.5 \times 10^{-5} \text{ s}^{-1}$. The background aerosol is non-absorbing and consists of ammonium sulfate mixed with biogenic secondary organic aerosol. This dilution rate corresponds to about 75% of the aerosol being replaced in a 24 h period, comparable to a diurnal mixing height increase of 500 m to 2000 m (e.g., Liu et al., 2011), although imposed uniformly over the day for simplicity."

4. Fig. 6: Are the calculated aerosol optical properties at dry state or ambient state (70% RH in your model)? I think it would be more interesting to see the results for ambient condition.

Authors' response: The aerosol optical properties are calculated for ambient conditions, i.e. we include the water content of the aerosol. To make this clear, we now address this explicitly in the Methods section as follows: "We calculate the optical properties for ambient conditions (including aerosol water content) at a wavelength of 550 nm and the critical supersaturations of each particle in a post-processing step according to Zaveri et al. (2010)."

5. P33559 L18-20: Overestimations in absorption efficiency would result in underestimates of SSA.

Authors' response: This error in wording has been corrected. The sentence has been revised to: "Previous work has clearly demonstrated that calculations of aerosol optical properties depend upon assumptions about particle mixing state, with assumptions of uniform internal mixing producing overestimates of absorption efficiency and underestimates of single scattering albedo."

6. P33560 L7: please define "rBC" here

Authors' response: Done. This sentence has been revised to: "...to assess the mixing state of refractory black carbon (rBC) containing particles derived from vehicle

emissions.”

7. P33561 L10: were used
Authors’ response: Done.

8. P33566 L3: Fig. 4e and f
Authors’ response: This error has been corrected in the revised manuscript.

9. P33582 L1: measurement-constrained
Authors’ response: This error has been corrected.

10. Figures: both m^{-3} and $/\text{cm}^3$ are used in axis label. Please use the same style throughout the paper.
Authors’ response: This inconsistency has been fixed, and cm^{-3} or m^{-3} is used in all figures.

References

- Liu, P. F., Zhao, C. S., Göbel, T., Hallbauer, E., Nowak, A., Ran, L., Xu, W. Y., Deng, Z. Z., Ma, N., Mildenberger, K., Henning, S., Stratmann, F., and Wiedensohler, A.: Hygroscopic properties of aerosol particles at high relative humidity and their diurnal variations in the North China Plain, *Atmospheric Chemistry and Physics*, 11, 3479–3494, doi:10.5194/acp-11-3479-2011, <http://www.atmos-chem-phys.net/11/3479/2011/>, 2011.
- Zaveri, R., Barnard, J., Easter, R., Riemer, N., and West, M.: Particle-resolved simulation of aerosol size, composition, mixing state, and the associated optical and cloud condensation nuclei activation properties in an evolving urban plume, *Journal of Geophysical Research*, 115, doi:10.1029/2009JD013616, 2010.

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