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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Mass concentrations of black carbon measured by four instruments in the middle of Central East China in June 2006" *by* Y. Kanaya et al.

**Y. Kanaya et al.**

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Reply to the Referee 3:

We thank the reviewer very much for reading our paper carefully and giving us valuable comments. Detailed responses to the comments are given below.

Comment 1. I like the authors' statement, that large uncertainty is identified in the measurement, but that measurement is still useful to constrain the emission rate. However, I think that statement should be qualified. This single location doesn't constrain the emissions from all of China. Furthermore a comparison between measurement and model would also contain some model errors. I suggest that authors re-write that statement to more clearly identify what can be understood.

Reply 1. We calculated backward trajectories from the top of Mt. Tai every 6 hours for one month (June 2006) and studied their spatial coverage. The hourly locations of the trajectories were given scores of exp(-t/t0), where t is the time for the air mass to take to travel from that location to Mt. Tai and t0 is 5 days (a typical lifetime of BC), and then the spatial distribution of the scores was calculated with a resolution of 1x1 degree. Only the trajectory points below 2500 m altitude were considered. The scores were then multiplied by the gridded emission rates of BC (Streets et al., 2003) to estimate the likely area from which the observed BC originated. Considering that the grids with values exceeding 0.7 percent of total (corresponding to the ratio of 5 hours to 720 hours) are significant, the origin area is estimated to be around 5x5 degrees over Central East China. Although the main part of Central East China is covered, the emissions from whole China are not fully constrained by the measurement, as suggested by the referee. In the revised manuscript, we will mention "emission rate of BC from Central East China" instead. Regarding the model errors, the statement will be qualified in the revised manuscript by adding "even when the model results are associated with an uncertainty of a factor of 1.5."

Comment 2. Some of the figures, especially the time series, are difficult to see. I am not sure what can be done about this. Possibly in Fig 3, the legends could be removed from inside the panels, and the high points could be cut off. Same comment for Fig 9.

Reply 2. We do not think it is a good idea to remove the high points artificially. The size information (PM1 and PM2.5) will be put into parentheses for better readability in Fig. 3. For Fig. 9 (a), the size information duplicated for MAAP and EC (or heated PSAP and EC) will be omitted (e.g., MAAP(PM1)/EC(PM1, NIOSH) in the original manuscript will be rewritten as MAAP/EC(NIOSH) (PM1))

Comment 3. There is a large number of acronyms in this paper. Although these are very well explained by the authors, this reader found it difficult to remember what each one stood for and had to search for the explanatory paragraphs many times. I suggest a table that contains the acronym, the instrument used to measure the quantity, and



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the principle of measurement.

Reply 3. A Table for this purpose will be included as Table A1 of Appendix A in the revised manuscript.

Comment 4. Page 14958, line 19: suggest "explanations" instead of "possibilities"

Reply 4. Correction is made accordingly.

Comment 5. Page 14959, line 19: "most significant region"- for what? What fraction of emissions occurs here?

Reply 5. We will rewrite it into "an outstanding region with high emission rates". The emission rate of BC from CEC (defined as 110-123 degE, 30-40 degN according to Richter et al., 2005) is 32 percents of that from all of China, on the basis of an emission inventory study (Streets et al.2003).

Comment 6. Page 14961, line 2: "measurements with less uncertainty"- I suggest that this approach doesn't reduce uncertainty, rather just helps understand it

Reply 6. We agree with the referee and replace the phrase "one approach to achieve BC measurements with less uncertainty" with "one approach toward better quantification of BC concentrations". We also remove the phrase "with less uncertainty" found in the original manuscript in page 14961, line 11.

Comment 7. Page 14962, line 28, spell out SUS

Reply 7. We will mention "stainless steel" instead.

Comment 8. Page 14963, description of heating PSAP sample. I understand the purpose of this heating-removing substance that could be coated on to the particles. But I do not understand it, physically. Certainly the PSAP sample is not made at 400 C, so a recooling occurs. Are we certain that all the volatile material evaporates or decomposes and never returns to the particle phase?



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Reply 8. It is an assumption with the PSAP instrument that all the volatile material evaporates or decomposes with heating and never returns to the particle phase, although it is verified with an Aerodyne AMS instrument as mentioned in the manuscript. This will be emphasized after revision.

Comment 9. Page 14963, line 14, and 14964, line 28. Mass absorption cross section is given for PSAP and aethalometer. Authors should discuss how uncertain this factor is: it might affect whether comparison with other instruments looks high or low.

Reply 9. We agree with the referee. We might be able to attribute the systematic deviation of the slopes from unity in Figs. 7 and 8 to the wrong cross sections at least partly. In the revised manuscript, we will include the following sentences in section 2:

It should be noted that the assumed mass absorption cross sections for the optical instruments are the ones empirically determined for particles under certain conditions. The conditions are not necessarily identical for all of the instruments. Therefore, it is not obvious that they are readily applicable for the black carbon particles at the top of Mt. Tai. The cross sections, depending on size, shape, and mixing state of the particles theoretically, should have uncertainties of a factor of 2. In this study, we use the cross sections as recommended by the manufacturers first for simplicity and then discuss systematic differences in the BC concentrations derived from the instruments.

Additionally, we will insert one relevant paragraph in the end of section 3.4 that may smoothly connect the results in section 3.4 with the discussion in section 3.5:

The deviations of the slope values in Figs.7 and 8 from unity could be attributable to the wrongly assumed absorption cross sections for the optical instruments at least partially. But it is not possible to re-determine the cross sections because none of the instruments can be regarded as standard. Nonetheless, it should be noted that there is a systematic tendency that all of the optical instruments need to assume larger cross sections than originally assumed to reduce the BC values down to the levels of EC (or that the EC values are consistently underestimated by some reasons). This tendency



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is studied in more detail in the next section.

Comment 10. Page 14963, line 26. "IMPROVE program." IMPROVE program uses reflectance rather than transmittance for optical correction. Some literature suggests that this difference is important. Authors should make clear that the use of transmittance is unlike IMPROVE and hence, the comparison may not be exactly like the full IMPROVE method. The comparison given here is valuable, however, because it shows the effect of 550C temperature without the confounding factor of the different optical correction.

Reply 10. The usage of transmittance in this study as opposed to reflectance in the normal IMPROVE protocol will be mentioned in section 2 of the revised manuscript.

Comment 11. Page 14964, line 9, "change in transmittance... used to determine BC concentrations optically." What cross section was used and how did this compare with the other cross sections?

Reply 11. The BC mass loading (in microgram cm-2) on the filter was empirically determined by 2.25ABS + 0.75(ABS)2, where ABS (absorbance) is determined from the temporal decrease in the laser transmittance during sample collection. The mass loading was then converted to concentration using the filter area and sample volume. We will add this information in the revised text. It is not meaningful to compare cross sections among the instruments because the filter material is different.

Comment 12. Section 3.2. NIOSH vs IMPROVE. This is a nice comparison, and I appreciate the contribution. However, it seems that there could be some confounding factors. The period when the NIOSH program operated has much different concentrations than the later period, when the IMPROVE operated. If the response depends on loading or concentration, this comparison is invalid. Further, the slope of the NIOSH line could be very much affected by the high points. I suggest that the authors also include regression statistics for the NIOSH and Opt-EC when limiting the Opt-EC concentration to 3 ug/m3.



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Reply 12. We had noticed this and included the analysis limiting the data set with Opt-EC with <2.64 ug/m3 in text of section 3.2.

Comment 13. Page 14968, lines 17-21. There is a relatively better agreement between MAAP and IMPROVE. However the disagreement ( 50 percents) is quite high and I don't think the 10 percent better agreement with the IMPROVE is important in light of this large disagreement.

Reply 13. We will weaken the sentence as IMPROVE* temperature protocol tends to give higher concentrations than does NIOSH, showing a "slightly" better agreement with the MAAP data. We do not strongly mean that IMPROVE* is better, but just wanted to clarify the magnitude relationship.

Comment 14. Section 3.5, differences (starting on line 12). These are really three explanations: the lensing, the cancellation by MAAP, and the split point. The MAAP instrument is supposed to cancel scattering but it does not account for the lensing effect, which is actually an increase of absorption.

Reply 14. We will remove the phrase "the cancellation (of scattering in MAAP) might not have worked well enough for the type of OC present in the studied atmosphere", because it was too speculative. Now we do not question the cancellation of scattering by MAAP and thus we mention two explanations (lensing effect and split point) from the beginning of discussion.

Comment 15. Page 14970, line 15. Interesting observation of volatility observed with low BC/EC ratio.

Reply 15. We thank the referee for this comment. We feel that more systematic understanding of the differences among the instruments is necessary to achieve reliable observations of BC in the future.

Comment 16. Page 14970, line 23-26. This explanation is puzzling. Authors have suggested that thermal EC measured by IMPROVE and by NIOSH are different. This



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suggests that OC/EC ratio for these two should be slightly different. Then, how could MAAP BC to EC ratio be the same for the two different measurements of EC? Also, I do not understand how this observation could be used to determine that the optical instruments are overestimating. Some clarification is needed.

Reply 16. As suggested by the referee, the averaged level of the OC/EC ratio tends to be lower for the IMPROVE* protocol than NIOSH. Here we wanted to describe if the "relative" temporal variations of the OC/EC ratio are coincident with the relative variations of the BC/EC ratios with the IMPROVE* protocol, as they were with NIOSH. To make this point clearer, we will mention that "qualitative" relationship between the MAAP BC/EC ratio and the OC/EC ratio was unaltered by the selection of the temperature program in the revised manuscript.

Comment 17. Page 14971, lines 10-11. "Aging does not affect the determination of the split point between OC and EC." How can you be sure of that statement? I agree that an aging effect is a reasonable explanation, but I don't think that the other explanation is eliminated.

Reply 17. We will change this phrase into "Considering that the tendency of underestimation of EC is not straightforwardly explained in terms of aging"

Comment 18. Page 14971, last paragraph (continues to 14972). This is a nice collection of studies, but I think the discussion should be sharpened. Also, not discussed, is the possible change in conversion of optical signal to mass (absorption cross-section in m2/gram). This is mentioned in the end of conclusions but should be discussed earlier. I think possibly this discussion would be more readable if the studies were summarized in a table showing instruments (for both BC and EC), ratio observed, and hypothesis advanced by the authors.

Reply 18. The possible change in the absorption cross section that the referee points out here is just the same issue as we have discussed in terms of lensing effect in section 3.5, which increases the absorption cross sections. As written earlier, we will



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include several sentences in section 2 and one paragraph in the end of section 3.4, to let the readers be aware that the used cross sections are those only empirically determined and thus uncertain. We do not suppose an additional table for the comparison to other studies is helpful, because they mostly discuss differences between Aethalometer and EC, while we discuss differences between MAAP (or heated PSAP) and EC here.

References

Richter, A., Burrows, J. P., Nuess, H., Granier, C, Niemeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437, 129-132, doi: 10.1038/nature04092, 2005.

Streets, D. G., Bond, T. C., Carmichael, G. R., Fernandes, S. D., Fu, Q., He, D., Klimont, Z., Nelson, S. M., Tsai, N. Y., Wang, M. Q., Woo, J.-H., and Yarber, K. F.: An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, J. Geophys. Res., 108(D21), 8809, doi:10.1029/2002JD003093, 2003.

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 14957, 2008.