

Lee et al. investigated the mixing state of BC and the chemical characteristics of its coatings near vehicle emissions. They found that substantial formation of SOA coatings on BC particles was due to photochemistry in the afternoon, whereas POA was strongly associated with BC from fresh vehicular emissions in the morning rush hours. The paper potentially has significant implications to the formation of SOA coatings on BC in many urban environments, so is well within the scope for ACP. The paper is very interesting although some clarifications regarding the data analysis are required. The paper can be recommended for publication after the following questions are addressed.

General comments:

In Line 8-11, Page 5, the authors mentioned that CE for rBC particles increased (or rBC particle beam width decreases) with coating thickness. Although the variation of CE will not impact calculations of R_{BC} , it will certainly influence the calculation of $NR-PM_{rBC}/NR-PM$, which refers to the proportion of internally mixed aerosols. The authors should evaluate the impact of variable CEs on the calculation of $NR-PM_{rBC}/NR-PM$. In addition, gas-phase CO_2 will contribute to the CO_2^+ signal and thus influence the chemical composition of OA (Aiken et al., 2007; Aiken et al., 2008). Was the contribution of gas-phase CO_2 to CO_2^+ signal corrected in this study? This information was missing in the manuscript.

Specific comments:

line 19-20, page 5: The data for SP-AMS on July 21 are not available, please give some clarifications.

line 26, page 5: What are the uncertainties of these items in Figure 2?

line 26-31, page 5: During the daytime from July 21 to 22, mass loadings of rBC and nitrate peaked in the afternoon on July 21 and in the morning on July 22 while the NO_x mixing ratios kept at relatively low levels. Does this indicate that other sources of rBC existed? Should the data on these two days be excluded from the calculation? Also, the poor correlation between NO_x and nitrate on these days may indicate formation mechanisms of particulate NH_4NO_3 other than the OH radicals oxidation of NO_2 .

line 37-39, page 5: Although sulfate followed the diurnal patterns of nitrate in the morning, it does not really mean its significant formation from photochemistry. Is it possible that regional transportation contributed to this pattern? Is there any other evidence such as the pattern of the precursor gas SO_2 to draw this statement? Figure 1 shows that there are quite some changes in wind directions/speed during the hot periods.

Page 6. The authors have indicated that C_x^+ signals can be derived from standard rBC. However, in the PMF analysis of the $NR-PM_{rBC}$, C_x^+ were used to identify an rBC rich

factor. I am somewhat confused here. Are data on Figure 3 and discussions on Page 6 (first paragraph) are for the whole particle or the coating? If they are just for the coating, why do these C_x^+ exist and how would we know if they are not from rBC?

Line 20-30, page 6. What is the major source of OOA-2? It does not seem to be the result of further reactions of OOA-1. The authors also discard the possibility of regional transport although it was mentioned in the conclusion later.

Line 33, Line 6. It is a bit awkward to me that R_{BC} is called “thickness”, given that it is a mass ratio.

line 6-7, page 7: The term POA may confuse the readers that COA is also included.

line 4-11, page 8: As shown in Figure S6, COA exhibited two peaks, one was around 17:00-18:00 similar to that of VOOA and the other larger peak was at midnight. This trend is largely different from typical diurnal patterns of COA that show a strong peak during dinner time. Also, large fractions of oxidized fragments were observed for m/z 28, 42 and 55 in COA mass spectra. Could COA be already oxidized and not primary in origin?

Line 15, page 8. Can organic sulfur compounds be formed in relation to VOOA? Huang et al. (2015) estimated OS lower bounds based on AMS measurements in HK.

At times, I am confused what exactly rBC means. Is it the refractory BC without OA or including the OA (coating) on the BC? For example, line 8 on page 7, it says “OOA-1 contributed up to ~60 wt% of rBC mass...” This may be related to my earlier question on the nature of C_x^+ . On the other hand, terminology like rBC rich OA factor seems to imply a different definition of rBC. Figure 4 also uses “total rBC”.

A conclusion is that the OOA in coatings of rBC are less oxygenated than those in VOOA. Just a speculation, would it be possible that the OOA in rBC condensed onto HOA coated rBC which promote partitioning of less oxygenated species while VOOA condensed on more hydrophilic particles?

Technical comments:

line 18, page 1: “missions” should be “emissions”.

Reference:

Aiken, A. C., DeCarlo, P. F., and Jimenez, J. L.: Elemental Analysis of Organic Species with Electron Ionization High-Resolution Mass Spectrometry, *Analytical Chemistry*, 79, 8350-8358, 10.1021/ac071150w, 2007.

Aiken, A. C., DeCarlo, P. F., Kroll, J. H., Worsnop, D. R., Huffman, J. A., Docherty, K. S., Ulbrich, I. M., Mohr, C., Kimmel, J. R., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M., Ziemann, P. J., Canagaratna, M. R., Onasch, T. B., Alfarra, M. R., Prevot, A. S. H., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U., and Jimenez, J. L.: O/C and OM/OC Ratios of Primary, Secondary, and Ambient Organic Aerosols with High-Resolution Time-of-Flight Aerosol Mass Spectrometry, *Environ Sci Technol*, 42, 4478-4485, 10.1021/es703009q, 2008.