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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Diurnal and day-to-day characteristics of ambient particle mass size distributions from HR-ToF-AMS measurements at an urban site and a suburban site in Hong Kong" *by* Berto P. Lee et al.

Anonymous Referee #1

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This manuscript presented a detailed analysis of a large data set of size-resolved particle composition measured by HR-AMS in Hong Kong. Both long-term trends and diurnal variations of the mass size distributions of submicron organic material, sulfate, and nitrate are discussed on the basis of previous understanding about the sources. Variations in the particle mixing state are also evaluated. This is perhaps the first study that looked at long-term AMS mass size distributions systematically, which potentially may serve as a good example of utilizing such data to derive better understanding of the sources and the atmospheric processing of submicron particles. The current

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Discussion paper

manuscript has however not yet arrived there. My main suggestions are (1) to justify the possible bias of the deconvolution of the mass size distributions ((Bian et al., 2014)maybe exclude some data; see my comment #1) and (2) to make a clear difference on which results are novel and which ones have already been published from previous analysis. Also, some figures contain too much information and hence are difficult to read. I therefore think a major revision or a resubmission is needed before this paper being accepted as a publication on ACP.

Specific comments:

(1) Regarding the analysis method (Section 2.2):

a) As shown in Figure D1, Aitken-mode peak often occurred in the left tail of the Accumulation-mode peak. If the heights of the two peak differ a lot (for example, one as only a few percent of the other one), it is easy to overfit the small peak, which may cause large uncertainties in quantifying the small peak. It is unclear to me how such overfitting is controlled in this study. Are any of the data points in Figures 1, 2, and 5 subject to this possibility?

b) I disagree that the transmission efficiency of the AMS lens unlikely affects the presented analysis (Line 122-126). Because the particle velocity calibration only spans for a certain range, extrapolation of fit may lead overestimation of the mass size distributions in small sizes. Slow vaporization and bounce may lead overestimation at Dva > 1 μm (Ref.: http://cires1.colorado.edu/jimenez-group/wiki/index.php/AMSUsrMtgs, Best Practices: IE and Velocity Calibrations - Ed Fortner & John Jayne). More importantly, the transmission efficiency for standard AMS lens drops at $\sim$ Dva> 400 nm or < 100 nm, and is below 20% for Dva > 1 μm or < 60 nm (Liu et al., 2007). The transmission therefore skews the size distributions. Zhang et al. (2005) showed that in Pittsburgh when the AMS suggests an accumulation mode at 500 nm, MOUDI shows a peak at 900 nm Dva. In this case, the fitting to AMS distributions might miss the main mode. Bian et al. (2014) showed that sulfate and nitrate etc. indeed occurred in mode size

much greater than 500 nm Dva in Hong Kong. Given all the reasons, for urban area that has larger accumulation mode, I think the parameters (GSD, MMD, and integrated area) from fitting to the right peak (e.g., in Figure D1) cannot represent the actual accumulation-mode distributions. Ideally, the size distributions can be corrected for transmission efficiency (at least for the right side). But it is very difficult to obtain the transmission efficiency for a specific AMS with standard vaporizer. I suggest the authors to justify their accumulation-mode analysis by additional data (e.g., from SMPS or MOUDI) or improved algorithm. Otherwise, it may not be meaningful to discuss the accumulation-mode changes.

(2) Figures 1 and 2: The discussions in page 5-11 are difficult to follow by reading those figures. For example, diurnal profiles for four gaseous pollutants have no size dependence. Showing them twice with the two particle modes is very confusing. Similarly, the shaded diurnal profiles for total submicron mass of different species made the figures difficult to read. I suggest to move those into a separate figure.

(3) The numbering of section 3.1, 3.2, and 3.2.1 seems wrong.

(4) Line 157: “median values” - it is better to clarify in the captions of Figures 1 and 2 what are the medians.

(5) Line 161: What is “residual traffic”?

(6) Line 168: The abbreviations only need to be defined when the full terms first appear. Same in figure captions.

(7) Line 191: What is “residual organic particle mass”?

(8) Line 186 and Line 215: Figures do not appear in order.

(9) Line 217-219: The smaller fraction of Aitken-mode to the total increase may be caused by a greater accumulation mode contribution. In the summer, we expect to have more SOA in general (stronger emissions of the precursors and stronger oxidation), which also may lead increased organic submicron particle mass.

(10) Line 244-247: The matching of ozone and sulfate is not enough to prove that the nighttime sulfate peak is contributed by heterogeneous SO2 oxidation by ozone. Are there any other evidence to support (PMF aqueous-processing factor, results from other studies, and so on)?

(11) Line 273-274: While the median MMD seem showing little change, the mean and 25th\75th percentiles show significant diurnal variations (Figure D2). Why? Also, although in Line 103-105, there is a bit information about the diurnal distributions. Figure D2 would confuse readers a lot by the ranking of the values (meaning that medians were not located between 25th and 75th). It is important to clarify what the median, mean, and 25th\75th stand for? I mean not the median values of MMD values but the MMD from a reconstructed distribution, right?

(12) Overall the discussion in Section 3 only focused on what were seen from this study. Do the interpretations agree or disagree with what are known from other studies (other than AMS). For example, for mixing state, are the findings here consistent with the understanding from single particle analysis? The paper needs to show which results are novel and which ones have already been published from previous analysis in terms of understanding the sources and atmospheric processing of submicron particles.

Technical remarks: Line 107: Extra period after “the world”. Line 157: Add hyphen between “Aitken” and “mode” when used as adjective. Similarly for “accumulation-mode particle concentrations” and so on.

Reference:

Bian, Q., Huang, X. H. H., and Yu, J. Z.: One-year observations of size distribution characteristics of major aerosol constituents at a coastal receptor site in Hong Kong - Part 1: Inorganic ions and oxalate, Atmos. Chem. Phys., 14, 9013-9027, 10.5194/acp-14-9013-2014, 2014.

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Zhang, Q., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L.: Time- and size-resolved chemical composition of submicron particles in Pittsburgh: Implications for aerosol sources and processes, J. Geophys. Res., 110, D07s09, 10.1029/2004jd004649, 2005.

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