Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-162-RC2, 2017

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 #2

Received and published: 20 June 2017

This manuscript reports a systematic study on the long term chemically-resolved size distribution data measured by a high-resolution AMS from an urban and a suburban location in Hong Kong. Measured size distributions of individual species were fitted using a bimodal lognormal model and the derived mode sizes and submode concentrations were analyzed for seasonal and diurnal variations. Based on these results, the authors discussed the influences of different sources on aerosol sizes, differences between urban and suburban aerosols, and variations in aerosol mixing states.

The work reported in this ms is technically sound and interesting and the synthesis of

long-term AMS size distribution data is a novel undertaking. However, the assumption that all aerosol size distributions are bimodal appears to be overly simplified and somewhat arbitrary. Urban particles, in particular, are contributed by various primary and secondary sources and particle from different sources tend to have different size distributions. Although I could see the benefit of simplifying the complexity by using a bimodal assumption, it would be helpful that the authors elaborate a bit more on the justification for this treatment and provide more details on how well the biomodal log-normal model perform in fitting the observation data. Maybe a more systematic evaluation of the quality of fit for the size distribution data is more appropriate than one example (Fig. D1). Also, I would like mention that more sophisticated methods, such as the 3-dimentional factor analysis reported in Ulbrich et al. (2012), maybe useful to explore the number of modes. I also notice that the naming of the size modes in this work is a bit confusing. Aitken mode refers to particles smaller than 100 nm in diameter. However, according to Fig. 1 and 2, the mode diameters for the so-called "Aitken mode" determined through bimodal log-normal fitting are all above 100 nm, some even reaching 200 nm. Additionally, the discussions on diurnal variations of aerosol size mode focus very much on the impacts of emissions sources and physical and chemical processes. However, changes of air masses due to wind shifts or upwind impacts could also be important and should be evaluated.

Following are detailed comments:

The numbering of the sections does not seem logic. For example, according to content, 3.2.1 is parallel to 3.2.

Line 11 - 12: this sentence is difficult to understand, consider to revise.

Line 63, the AMS lens transmission is close to zero for particles smaller than 30-35 nm, so it is not precise to say the Aitken mode particles (10-100 nm) are covered by AMS.

Line 65 states that the AMS particle size data from ambient measurements are rarely

investigated in depth. This is not true. A number of studies, including a few from more than 10 years ago, analyzed the size-resolved composition data from AMS quite extensively and utilized the information to elucidate aerosol sources, new particle formation and growth mechanisms, and other atmospheric processes. Several references (not the complete list) are provided at the end of this comment in the reference section. In addition, Ulbrich et al. (2014) reported a comprehensive study on the size-resolved mass spectral data from an ambient study using 3-D factorization models. Considering that this manuscript focuses on AMS size distribution data, I'd like to recommend that the authors provide a background review on previous works in the introduction. Additionally, I notice that citations are sometime missing when findings from the authors' own research group are mentioned. This could cause confusion when the results from this work alone are sufficient to support the claim. A thorough check for in-text citations is recommended.

Line 91, what's the RH at the exit of the dryer?

Line 130 - 132, the sentence "Utilizing ..." is vague, consider to revise.

Line 160, "sweep-out" by what, rain?

Line 161, what's residual traffic?

Line 167 – 169, clarify what the decreases correspond to.

Line 170- 173, are there SMPS measurements to support the increase of particle number concentrations?

Line 172, where does the cutoff size of 50 nm come from?

Line 174 – 190, how important was COA in Aitken mode around noon time? What about contributions from secondary aerosol formation and other primary sources such as HOA? Summertime SOA and SIA formation tend to be higher and can influence particles in all size modes. I am not sure change of cooking behavior was the only reason for the different diurnal shapes between spring and summer.

Line 189 – 190, this sentence is somewhat confusing

Line 213-215, this sentence is confusing. Please clarify.

Line 215-216, what does "nucleation of gas-phase emissions" mean?

Line 222-223, "reduce nucleation... of more volatile exhaust component on fresher, smaller particles..."? Did nucleation ever occur with the volatile component in the atmosphere?

Line 244-247, this discussion seems somewhat speculative. Are there data to support the nighttime heterogeneous oxidation of SO2 by O3 in Hongkong during spring time? Has this issue been investigated in previous publication(s)? Did wind direction or air mass origin play a role in the observed size mode change?

Line 437, "particles containing different species were similar in size" is confusing.

Line 449, small particles are not just processed by condensational growth and coagulation. In the presence of high humidity, they can also go through aqueous-phase processing.

Fig. D6, can wind data be provided as well?

References:

Allan et al. (2003), Quantitative sampling using an Aerodyne Aerosol Mass Spectrometer. Part 2: Measurements of fine particulate chemical composition in two UK Cities, Journal of Geophysical Research-Atmospheres, 108(D3), 4091

Alfarra et al. (2004), Characterization of urban and regional organic aerosols in the lower Fraser Valley using two Aerodyne Aerosol Mass Spectrometers, Atmospheric Environment, 38, 5745–5758 Drewnick et al. (2004), Measurement of ambient aerosol composition during the PMTACS-NY 2001 using an Aerosol Mass Spectrometer. Part II: Chemically speciated mass distributions, Aerosol Science & Technology, 38(S1), 104-117.

Zhang et al. (2004), Insights into the chemistry of new particle formation and growth events in Pittsburgh based on Aerosol Mass Spectrometry, Environmental Science & Technology, 38(18), 4797-4809

Ulbrich et al. (2012), Three-dimensional factorization of size-resolved organic aerosol mass spectra from Mexico City, Atmospheric Measurement Techniques, 5, 195-224, doi:10.5194/amt-5-195-2012.

Setyan et al. (2014), Chemistry of new particle growth in mixed urban and biogenic emissions: insights from CARES, Atmos. Chem. Phys., 14, 6477-6494

Sun, Y et al. (2016), Primary and secondary aerosols in Beijing in winter: sources, variations and processes, Atmos. Chem. Phys., 2016(16), 8309-8329, doi:10.5194/acp-16-8309-2016.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-162, 2017.