Summary

Cheung et al. conducted a set of ambient measurements from which they calculated sizedependent volatility shrinkage factors (VSF) of aerosols in Guangzhou after heating to 300°C in a tandem differential mobility analyzer. Size-selected particles ranging from D_m = 40 to 300 nm were examined. Mass concentrations of OC and EC were also measured. Particles were classified as "completely volatile" (CV; VSF \sim 0), "high volatility" (HV; VSF< 0.4), "medium volatility" (MV; 0.4<VSF< 0.9) and "low volatility" (LV; VSF > 0.9). Three primary results are reported: (1) the number and volume fraction of CV particles decreases with increasing particle size, while the LV particle number and volume fractions increase with increasing diameter (2) size-resolved measurements combined with average diurnal patterns suggest that 40 nm CV and LV particles represent local, fresh emissions, whereas >80 nm HV and MV particles represent aged emissions. (3) A closure analysis of VHTDMA and OC/EC analyzer measurements suggests that organics comprise a significant fraction of the measured MV and LV. Overall, the results are interesting, but I suggest additional analysis of the data before I would support publication in ACP. In particular, I think it would be useful to present more of the OC/EC results to assist with, and expand on, the interpretation of the VHTDMA measurements.

Main Comments

- 1. In my opinion, the closure analysis -- which currently focuses on a comparison of EC + OC2 + OC3 + OC4 versus LV + MV is incomplete. The volatility-resolved VHTMDA and OC/EC analyzer measurements should in principle allow for a more comprehensive closure/intercomparison study. Because the volatility fractions in both instruments are affected by the specific operation conditions, I think expanding on this subject in Section 3.3 would be interesting and possibly help with the interpretation of the VHTDMA measurements. I suggest that this subject be a major focus of a revised manuscript. For example:
 - a. CV versus OC1
 - b. HV versus OC1 and/or OC2
 - c. MV and OC2 and/or OC3
- 2. I think the authors should plot and discuss campaign-average mass fractions of OC1, OC2, OC3, OC4 and EC to accompany the volume fractions of VM, CV, HV, MV and LV that are presented in Figure 6 and related discussion.
- 3. Similarly, the authors could plot time series and diurnal patterns of OC1, OC2, OC3, OC4 and EC mass fractions as is done in Figure 7 and related discussion of the volume fractions of VM, CV, HV, MV and LV.

Minor/Technical Comments

- 4. It is not clear to me how understand the difference between "Volatile Materials" (VM) are defined. I assumed that "VM" becomes "CV" after heating to 300°C, but this does not seem to be the case because separate volume fractions of "VM" and "CV" are presented in Figures 6 and 7. Please clarify the definition of VM.
- 5. OC2, OC3 and OC4 are never defined in the manuscript.

- 6. What is the residence time in the heated section of the VTMDA, and how sensitive are the HV/MV/LV classifications to the residence time?
- 7. P25275, L8-10: The authors state: "Upon heating at 100°C and beyond, volatile components of the particle such as sulfate, nitrate and volatile organics vaporize". Please plot VSF (at 300°C) of ammonium sulfate, perhaps as a supplemental figure, over a few sizes ranging from 40 nm to 300 nm. I would not have thought that ammonium sulfate completely vaporizes at only 300°C.