

***Interactive comment on “Vertical distribution of atmospheric particulate matters within urban boundary layer in southern China: size-segregated chemical composition and secondary formation through cloud processing and heterogeneous reactions” by Shengzhen Zhou et al.***

**Anonymous Referee #2**

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This manuscript presents measurements of the vertical distribution of aerosol composition in Guangzhou, China and, based on those data, provides insight into the sources and formation mechanisms of the different chemical components. The manuscript is certainly understandable, but would require editing prior to publication. As is noted in the paper, datasets such as these can be very useful and yet there are very few avail-

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able. Because of that utility I feel that this dataset should be published. However, I feel that the use of the data to infer aerosol sources is weak and can probably not be refined to the level that would make ACP the appropriate journal for publication.

Simultaneous and semi-continuous measurements such as those described here are challenging to make and are more amenable to collection and off-line analysis techniques such as those used here. Nevertheless, interpretation of the data is constrained by the resulting low time (24 h) and size (7 bins) resolution. For example, some of the central conclusions of the manuscript are based on the relative concentrations of species in the droplet mode, but that mode is contained in a very broad 0.44 – 1.0 micron bin. The time resolution also complicates the interpretation, especially when attempting to connect the measurements with meteorology in the case studies.

I have questions/concerns about the impactors, in part because I have never used these and am unfamiliar with aspects of their operation. i) How were they calibrated? There is no information provided and the calibrated (?) cut sizes just happen to be exactly the same as those reported by the manufacturer. ii) How is the flow rate controlled? I ask because any pressure and/or temperature dependence would influence the recorded concentration height dependence. And connected to that, are the reported concentrations at local temperature and pressure or are they adjusted to standard (or other) conditions? iii) Is the air dried upstream of the impactors? For such a large flow rate I suspect the answer is no. And if not, this could have important impacts on the relative size distributions of the different chemical species and of the same species at different heights. The size distributions of hygroscopic species such as sulfate and nitrate would be shifted, while those of less- or non-hygroscopic species such as OC and EC would not. I appreciate that such shifts are not solely responsible for the differing size distributions, but they could be a contributor. The size distribution of those hygroscopic species would also vary with height due to variation in RH. This might partly explain the observation that the sulfate size distribution was shifted more into the droplet mode at 488 m, which was argued to be evidence of cloud process-

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ing in the manuscript (page 9, line 10). The average RH of between 78% and 80% suggests the bias could be significant.

The authors interpret the relatively flat vertical profile of sulfate compared with EC and OC as an indication of the importance of cloud processing. But there is no explanation provided about how that contrast would differ for local emissions of EC and OC and simply regional production of sulfate (gas or aqueous phase). I'm not so sure the difference would be easily discerned.

I agree that using meteorology to interpret the aerosol data and to constrain the origin and formation of the different species is logical. But I don't agree that almost exclusively relying on WRF model output is reasonable. Why not at least validate those elements of the model predictions for which surface and/or satellite observations are available. Cloud cover and cloud base height are two that come to mind.

For much of the discussion I believe it would be better to describe variations in absolute rather than relative concentrations. I recognize that for some explanations it is appropriate to describe differences in percent contribution of one or more species to the total concentration. But for other descriptions varying concentrations of other species unnecessarily complicates the results. One example is the conclusion on page 8, line 12 of favorable formation of the inorganic ion species based on relative changes in composition with height. The percentages would of course change in the same way if the concentrations of those species increased or those of other species decreased.

Minor issues in the order in which they appear in the manuscript:

Page 3, Line 8: What are irregular sizes? And dust is usually used instead of sand.

Page 7, Line 24: SNA spelled out only later.

Figure 2: Some explanation should be provided for the mismatch between the real-time and offline estimates of PM<sub>2.5</sub>.

Figure 5: Some explanation should be provided for why the concentration of NH<sub>4</sub> is

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highest at 488 m while that of SO<sub>4</sub> and NO<sub>3</sub> are not.

Figures 5 and 6: I believe the 0.1 on all of the x-axes is supposed to be 0.01 and 0.1 is for some reason not shown.

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