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Atmospheric Chemistry and Physics Discussions

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 #1

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The study by Zhou et al. is an interesting one considering the fact that very high towers are not readily accessible to researchers. Having said that, similar studies have been performed around the world and despite claiming the uniqueness of the study it is not entirely unique, except for utilising perhaps the highest tower. However, it is not the height of the tower which makes any study unique, but instead scientific insights about

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Discussion paper

the processes drawn from it. The study is not without significant drawbacks and needs significant improvement to warrant publication in the respected journal of Atmospheric Chemistry and Physics. Last but not least, English of the manuscript needs significant improvement as many sentences are unclear or dubious.

Major comments

The introduction needs significant improvement as overall interpretation of PM sources and processes is rather outdated, or straightforward or not consistent with the most recent fundamental papers. Very often the authors choose to reference either old papers missing out on recent ones, or choose to reference very recent, neglecting pioneering earlier papers. It is unclear what exactly were the goal and aims of the study other than utilising a very high tower. Those goals should normally arise from the earlier papers by identifying scientific knowledge gaps and which the authors choose to advance upon.

The paper currently stands more like a report rather than a scientific paper. It presents data, but lacks coherent view. More often than not the authors seek consistency with other studies or providing references which support or fit their data. Taken altogether, the paper is currently a collection of interpretations which are not always consistent with each other and most importantly lacking conclusive findings which would advance rather than confirm already known processes or phenomena.

The study is lacking an overview of all the profiles, splitting into certain categories and introducing the scope and variability of the data set first. There is a complete lack of uncertainty and error analysis. Analytical and sampling uncertainty would propagate into vertical profile uncertainties which would then make profiles or concentrations at different heights significantly different or not. The authors choose to select specific episodes or profiles which are most obvious or interpretable and neglecting which are not. Selected profiles are certainly insightful, but only when put into overall context.

Considering the challenges in organising such a study it is pity that high frequency mi-

crometeorological measurements (including an important vertical wind speed) were not undertaken making it impossible to derive fluxes (refer to papers by (Valiulis, Ceburnis et al. 2002, Ceburnis, O'Dowd et al. 2008).

Minor comments

Page 2, line 27. Outdated literature overview of the processes involved and oversimplifying the system.

Page 3, line 5. Old literature support. There is plenty of hard evidence that the first sub-mode can also be formed by cloud processing, e.g. (Ovadnevaite, Zuend et al. 2017)

Line 6. Unclear sentence - what was exactly demonstrated worldwide?

Line 9. Sea salt can also be submicron down to 20nm, e.g. (Ovadnevaite, Manders et al. 2014, Cravigan, Ristovski et al. 2015)

Line 19. One of the earliest papers published by (Valiulis, Ceburnis et al. 2002) which also estimated emissions from the observed gradients.

Page 4, line 13. Introduction should only present goals and objectives of the study and not the description of measurements performed unless nobody measured them before which is not the case here.

Line 26. delete "the other three levels".

Page 5, line 5. Why 168m level is missing? 168m is often missing in the results section and is not clearly explained why.

Page 6, line 17. Why the study is focused only in several pollution episodes when an overview of gradient should be presented first including error bars and uncertainties. Selected gradients discussed later become suspicious whether they are representative or just being random.

Line 23. It is not the consistency with other studies that makes the measurements reliable trusted. Instead, decreasing concentration with height points at the ground/surface sources as opposed to increasing concentration with height pointing at sources aloft (Ceburnis, O'Dowd et al. 2008)

Page 7, line 1. This needs to be investigated if not occurring due to temperature inversions impeding mixing. That can be especially true in winter, but temperature inversions readily forming under clear sky condition due to radiative cooling.

Line 6. "Concentration gap" is unclear and unsuitable term.

Line 10. Composition cannot be vertically distributed - chemical components are distributed instead.

Line 16. Repetition.

Line 21. It does not need to be associated to EC as many other species are emitted by sources at the ground.

Line 24. Please spell acronyms used for the first time.

Line 27. No established vertical gradients...

Page 8, line 12. Secondary WSOC formation is the scientific fact - why is it missing from interpretation?

Line 21. If distributions were averaged they must be presented with errors bars or ranges.

Line 27. Sulfate having similar formation mechanism to what?

Page 9, line 4. Coagulation is negligible at typical ambient number concentrations (refer to e.g. W.C. Hinds Aerosol Technology Textbook). In-cloud coalescence of droplets is more likely or multiple cloud cycles could explain production of several modes.

Line 20. Formation of nitrate is not exclusive to sea salt or dust particles, any surface

would promote heterogeneous reactions.

Page 11, line 5. Chloride particles do not exist and chloride cannot be considered separately from sodium or other balancing ion like ammonium.

Line 13. Incorrect suggestion. That finding is only demonstrating aged aerosol which undergone significant chemical processing during advection to the sampling location.

Line 22. Unclear sentence. Similar origin of OC at three heights? Its unlikely as similar concentrations can be produced by proportional contribution of ground sources versus in-cloud processing.

Line 27. That is not a possibility, but rather the only plausible explanation. However, the fact that nearby chimneys may have affected the profiles diminishes the value of this study making the interpretation of profiles very speculative and simply fitting the observations.

Page 12, line 21. Observed, not found.

Line 28. Why the authors suggest what was already pointed out as contribution from nearby chimneys and stacks?

Page 13, line 10. ...temperature inversion extending from 118 to 488 meters...

Line 16-23. Schematics is haphazardly constructed and needs much better discussion and reasoning based on observations. 8 lines are absolutely insufficient. This schematics should be significantly improved or removed altogether. Figure 9. Why an upward convective transport missing? Schematics is lacking sampling heights to validate the processes.

Line 26. "Utilizing the 610m Canton Tower in Guangzhou" has to be moved to the sentence end.

Line 28. Small or smaller? Shallower is perhaps the better word.

Page 14, line 6. OC missing

Line 15. In order for the results of the study helping understanding formation mechanisms, the data interpretation needs considerable improvement.

Ceburnis, D., C. D. O'Dowd, G. S. Jennings, M. C. Facchini, L. Emblico, S. Decesari, S. Fuzzi and J. Sakalys (2008). "Marine aerosol chemistry gradients: Elucidating primary and secondary processes and fluxes." Geophysical Research Letters 35(7): L07804.

Cravigan, L. T., Z. Ristovski, R. L. Modini, M. D. Keywood and J. L. Gras (2015). "Observation of sea-salt fraction in sub-100âĂŁnm diameter particles at Cape Grim." Journal of Geophysical Research: Atmospheres 120(5): 1848-1864.

Ovadnevaite, J., A. Manders, G. de Leeuw, D. Ceburnis, C. Monahan, A. I. Partanen, H. Korhonen and C. D. O'Dowd (2014). "A sea spray aerosol flux parameterization encapsulating wave state." Atmospheric Chemistry and Physics 14(4): 1837-1852.

Ovadnevaite, J., A. Zuend, A. Laaksonen, K. J. Sanchez, G. Roberts, D. Ceburnis, S. Decesari, M. Rinaldi, N. Hodas, M. C. Facchini, J. H. Seinfeld and C. O. Dowd (2017). "Surface tension prevails over solute effect in organic-influenced cloud droplet activation." Nature 546(7660): 637-641.

Valiulis, D., D. Ceburnis, J. Sakalys and K. Kvietkus (2002). "Estimation of atmospheric trace metal emissions in Vilnius City, Lithuania, using vertical concentration gradient and road tunnel measurement data." Atmospheric Environment 36(39-40): 6001-6014.

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