Dear Professor Barbara Ervens,

Thank you for your letter and for the reviewer's comments concerning our manuscript entitled "*Vertical distribution of atmospheric particulate matter within the urban boundary layer in southern China: Size-segregated chemical composition and secondary formation through cloud processing and heterogeneous reactions*". Those comments are all valuable and very helpful for revising our paper, as well as the important guidance to our work. Accordingly, we made some changes in the revision to improve the manuscript.

We want to take this opportunity to explain the main motivation for this study. Though many measurements of atmospheric aerosols have been made at ground level, very few was focused on vertical measurements of size-resolved particulate matter within the urban boundary layer. We are fortunate to carry out long term measurements based on ~ 610 m tall Canton tower. It is innovation to reveal aerosol vertical distribution and formation in subtropical urban areas. It was found that cloud processing and heterogeneous reaction could be important formation pathways for the major aerosol compositions, contributing largely to air pollution in this urban region. In addition, we provided case studies to illustrate that cloud processing and heterogeneous reactions play important roles in the haze pollution in this area.

To broaden the scientific scope, we adopted the reviewer's suggestion to perform WRF-Chem simulation to see how well the model performance at reproducing the measurement results. Our results showed that model can simulate some components well such as ammonium and some components, such as sulfate, nitrate and OC, are poorly simulated. It indicated that the measurement data can be used to improve model in the future work. However, the comments were on the model improvement in this reviewer's suggestion. We agree that it is important for model development, but it is beyond the scope of this study. Hence, we still think this paper should focus primarily on measurement data analysis.

Once again, thank you very much for helping us with our manuscript. Major changes in the revision and point-to-point responds to the reviewer's comments are listed below. We hope that all the concerns raised by the reviewer shall be resolved.

Sincerely yours,

Shengzhen Zhou, PhD/Associate Professor
Xuemei Wang, PhD/Professor

Response to referee

The revised paper has improved, and it offers a useful look at the potentially more important processes in the evolution of the components of aerosol particles in this region. The authors enhanced the discussion of OC and EC, and they now include some comparisons with the results of WRF-Chem simulations. However, I find the comparisons and their discussion to be a little disappointing for the following reasons:
[A]: We would like to express our sincere appreciation for the reviewer's careful reading and invaluable comments to improve the paper. We tried our best to improve the manuscript and made some changes in the manuscript. Please kindly find our itemized responses to the specific comments below. The reviewer's comments are in black, and authors' responses are in blue. The changes in the manuscript are highlighted in red.

• The comparisons are not shown in the main text.
• There are no comparisons with OC, EC or NaCl.
• Other than to say that "it might not be well simulated", there is no attempt to assess the model's simulation of cloud with cloud observations (e.g. Figure S11), which is fundamental to their argument for cloud processing.
• Overall, the discussion seems hurried. It does not carefully consider all the major issues that may be associated with the comparison to their measurements and the use of the model to develop the conceptual picture they present in Figure 9.

The authors state that model evaluation "is beyond the scope of this study", yet they use the model results (e.g. Figure 7) to build their concept of the processes affecting haze formation in this region (Figure 9). This is not evaluation for the model. It should be evaluation to support your results, in other words, "does the model support their portrayal of the processes?".
[A]: Thank you for the comments. We have made lots of testing to WRF-Chem model, and some of the results are put in the text and supplementary.

"The performance statistics for meteorological elements such as pressure, air temperature, relative humility and wind speed of three vertical layers on the Canton Tower and chemical pollutants such as $PM_{2.5}$, NO_x, O_3 and SO_2 are shown in Table S5 and Table S6. Here, the statistical measures such as 5 Observation Mean, Simulation Mean, the Mean Bias (MB), the Normalized Mean Bias (NMB), the Normalized Mean Error (NME), the Mean Relative Bias (MRB), the Mean Relative Error (MRE), the Root Mean Squared Error (RMSE) and the correlation coefficient (CORR) are used for modeling validation.

Table S5. Comparison of Simulated Hourly Meteorological Elements with Observation Data

			Mean						
Meteorology (Unit)	**Height**	**number[a]**	**Obs.**	**Sim.**	**MB**	**NMB[b]**	**NME[b]**	**RMSE**	**CORR**
Autumn									
PRES (hPa)	GND	925	1015.4	1013.7	-1.7	-0.2	0.2	1.8	0.98
	121m	950	1002.0	1002.0	0.0	0.0	0.1	0.7	0.98
	454m	952	959.9	959.2	-0.7	-0.1	0.1	1.3	0.94
TA (°C)	GND	952	24.8	23.7	-1.2	-4.7	6.8	2.1	0.92
	121m	950	23.4	22.9	-0.6	-2.4	5.0	1.5	0.94
	454m	952	20.6	20.8	0.2	0.9	3.9	1.1	0.95
RH (%)	GND	952	62.5	65.6	3.1	5.0	12.4	9.8	0.82
	121m	950	64.9	67.1	2.2	3.4	11.2	9.1	0.85
	454m	952	72.4	73.0	0.6	0.9	9.6	8.8	0.86
WS (m/s)	GND	952	0.7	2.3	1.6	227.0	234.8	2.1	0.62
	121m	753	2.1	5.6	3.6	170.6	177.1	4.7	0.30
	454m	936	4.1	6.6	2.5	60.2	74.6	4.0	0.55
Winter									
PRES (hPa)	GND	758	1021.2	1019.5	-1.6	-0.2	0.2	1.8	0.99
	121m	770	1006.8	1007.8	1.1	0.1	0.1	1.3	0.99
	454m	776	962.9	964.9	2.0	0.2	0.2	2.4	0.97
TA (°C)	GND	765	14.8	12.7	-2.2	-14.6	16.2	2.9	0.91
	121m	525	14.7	13.5	-1.1	-7.7	10.5	1.8	0.94
	454m	648	10.7	11.4	0.8	7.4	41.2	5.1	0.84
RH (%)	GND	765	67.0	68.4	1.4	2.1	14.4	11.9	0.84
	121m	522	82.2	71.0	-11.2	-13.6	13.9	15.1	0.85
	454m	245	73.9	54.2	-19.7	-26.7	28.1	25.1	0.85
WS (m/s)	GND	765	0.9	2.4	1.5	175.7	179.1	1.9	0.71
	121m	526	2.0	6.4	4.4	215.5	220.6	5.7	0.23
	454m	751	4.8	8.2	3.3	68.9	76.3	4.6	0.66

[a] the number of observed data

[b] the unit of NMB and NME is in %, other statistical variables are same as the meteorological element

Table S6. Comparison of Simulated Hourly Chemical Pollutants with Observation Data (unit: μg/m^3)

			Mean						
Meteorology (Unit)	**Height**	**number[a]**	**Obs.**	**Sim.**	**MB**	**NMB[b]**	**NME[b]**	**RMSE**	**CORR**
Autumn									
$PM_{2.5}$	GND	958	43.6	42.6	-1.0	-2.3	59.9	35.9	0.24
	168m	954	35.5	20.8	-14.7	-41.5	54.1	23.6	0.09
	488m	947	27.5	13.0	-14.5	-52.8	59.9	19.9	0.22
NO_x	GND	949	101.7	90.7	-10.9	-10.7	62.0	93.4	0.41
	168m	922	75.4	28.0	-47.4	-62.9	68.9	69.3	0.13

	488m	950	27.1	7.7	-19.3	-71.5	79.7	28.9	0.24
O_3	GND	939	36.1	39.6	3.5	9.6	58.5	31.1	0.72
	168m	947	58.3	72.0	13.8	23.6	62.9	46.6	0.64
	488m	947	103.8	95.7	-8.2	-7.9	37.8	51.9	0.55
SO_2	GND	953	13.8	11.7	-2.1	-15.3	61.5	12.0	0.10
	168m	949	17.8	5.1	-12.7	-71.4	72.4	16.7	0.03
	488m	951	12.8	2.9	-9.9	-77.6	78.2	13.8	0.16
Winter									
$PM_{2.5}$	GND	775	40.8	47.5	6.8	16.6	50.1	27.9	0.43
	168m	756	33.0	28.7	-4.3	-12.9	42.0	20.5	0.35
	488m	779	21.7	18.3	-3.4	-15.6	45.6	13.9	0.35
NO_x	GND	755	115.4	167.1	51.7	44.8	83.4	147.7	0.45
	168m	785	75.7	29.4	-46.3	-61.1	66.6	68.9	0.33
	488m	705	25.6	5.7	-20.0	-77.9	85.5	29.5	0.28
O_3	GND	761	19.8	19.6	-0.2	-1.0	77.3	22.2	0.65
	168m	786	26.6	61.3	34.7	130.1	137.5	43.3	0.49
	488m	784	63.4	88.5	25.0	39.5	44.0	33.7	0.38
SO_2	GND	784	9.4	22.0	12.5	132.4	150.7	22.2	0.23
	168m	786	11.8	7.8	-4.1	-34.5	48.2	8.8	0.05
	488m	783	9.7	5.4	-4.3	-44.0	53.7	7.4	0.05

[a] the number of observed data

[b] the unit of NMB and NME is in %, other statistical variables are same as the meteorological variable

We have done our best to simulate the vertical distributions of major components of $PM_{2.5}$, and found some potential problems to WRF-Chem model.

I suggest addressing that broader question by addressing the following more specific questions:

• **1.** Is it reasonable to expect that these data represent the grid box(es) that are being compared?

[A]: Thank you for the suggestions. In order to evaluate the general performance of the WFR-Chem as suggested, average data with uncertainties were plotted for the comparison between simulation and observation. We think this is a reasonable method for the comparison.

• **2.** How do the WRF-Chem simulations of OC, EC and NaCl compare to the observations? This will help to further identify model limitations.

[A]: We have compared concentration profiles of OC, EC and NaCl from the WRF-Chem simulations with those from our observations. The results show that, to some extent, the model still underestimate concentration of these species (Figure i). The WRF-Chem model setup and verification were given in the *supplementary.*

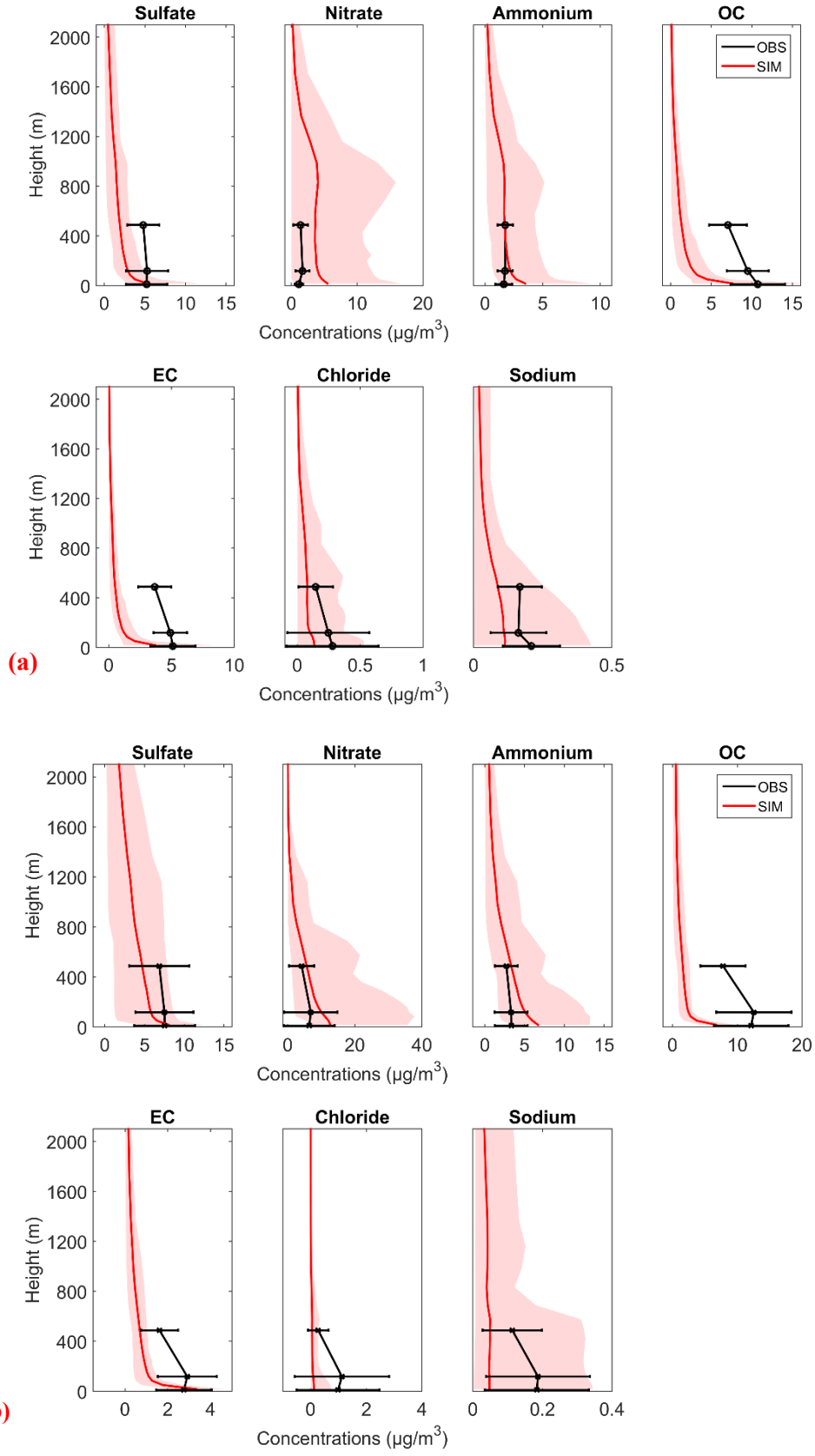


Figure i. The vertical concentration profiles of sulfate, nitrate, ammonium, OC, EC, sodium and chloride in $PM_{2.5}$ during (a) autumn and (b) winter field study (The red solid lines are the average modeled concentrations and the shaded regions indicate the

minimum and maximum values of the simulation; the average measurement data were in black with horizontal error bars).

• **3.** If WRF-Chem does not simulate sulphate in a reasonable way, then might it be because the temperature structure and winds of the lower levels are incorrect (as suggested by their statement that SO_2 was underestimated "possibly due to the insufficient upward transport of SO_2"), because SO_2 emissions are underestimated or because of a deficiency in the aqueous phase conversion of SO_2 to sulphate (e.g. representation of cloud; underestimation of oxidant concentrations; etc.)?

[A]: We totally agree that the underestimation of sulfate was possibly attributed to the underestimation of SO_2 precursor or a deficiency in the aqueous phase conversion of SO_2 to sulphate. These discussions were included in the last revision in response to the reviewer's insightful comments (page 17, lines 22-27):

"Sulfate was generally underestimated in WRF-Chem model at the upper level, while was in relatively good agreement with observation at ground level. Possible reasons for the underestimations of sulfate are: (1) SO_2 precursors were underestimated at the upper levels (by about 45% to 77%, Table S6), possibly due to the insufficient upward transport of SO_2 in the current model, especially in urban area where the urban canopy is low in resolution; (2) heterogeneous/multiphase formations of sulfate in droplets or aerosol water have not been fully considered in current model (Chen et al., 2016; Cheng et al. 2016)."

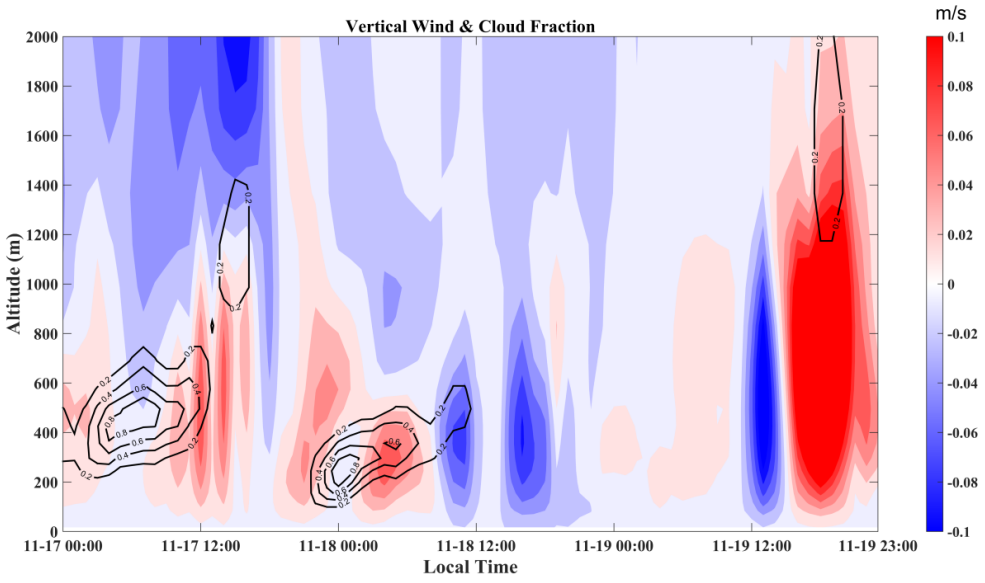
Our results present a unique vertical distribution dataset which should be useful for further model improvement. As suggested by the reviewer, many factors may contribute to inconsistency between simulation results and measurements, including meteorological parameters, cloud representation, underestimation of oxidant concentrations. We admitted that it is difficult to fully simulate the aerosol compositions using model such as WRF-Chem, especially in the vertical direction, which should still be an open question to the scientific community. This information was added in the manuscript (page 18, lines 7-16):

"The large discrepancies between observation and simulation on sulfate and nitrate suggested that physical and chemical mechanisms in current WRF-Chem model still need to be improved to better predict aerosol mass and composition. Based on our observation, in-cloud aqueous phase reactions and heterogeneous reactions should play important roles in sulfate and nitrate formation, which need to be refined in the model. Evaluation of WRF-Chem model incorporating the above-mentioned mechanisms is beyond the scope of this study and in-depth investigation needs to be done in future. Hence, more studies, such as long-term aerosols and high frequency micrometeorological measurements (Valiulis et al., 2002; Ceburnis et al., 2008; Ervens, 2015), are needed to identify the key aerosol sources and formation pathways, and to further improve the air quality models."

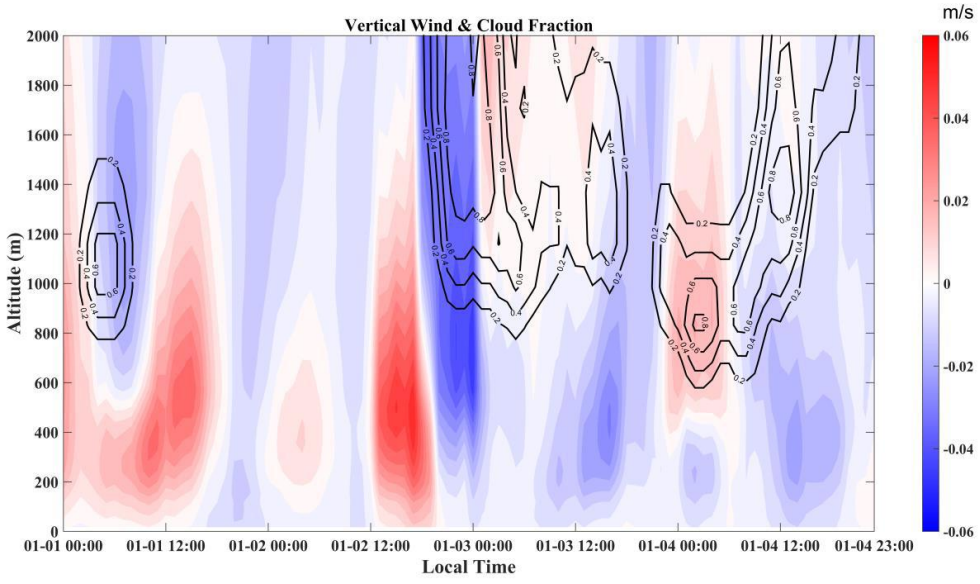
• **4.** Based on RH, it would seem that the model gets the clouds about right in the winter. Is that correct? What about autumn?

[A]: We detected and modeled low clouds both in autumn and in winter. However, the

density of the clouds in autumn was different from that in winter and much more clouds were observed in winter. We have provided some additional evidences in the manuscript: (1) The meteorological output from WRF-Chem simulations (Figure 8 in the manuscript); (2) The satellite images and ceilometer measurement (only in winter) (Figures S10 and S11 in the supplementary); (3) Records of in-situ observation (Figure ii).

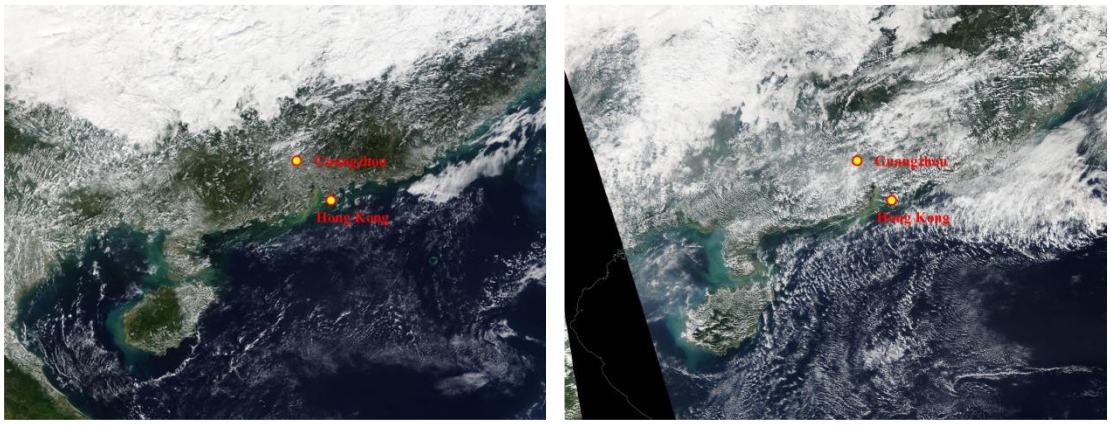


(a)



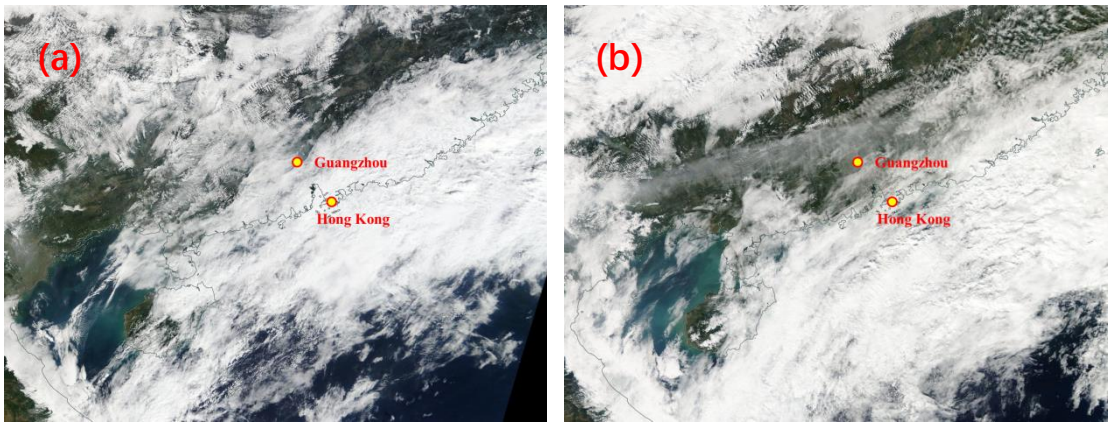
(b)

Figure 8. Distribution of vertical wind (color scale, red: upward; blue: downward) and cloud fraction (black contour line) simulated by the WRF model during (a) autumn and (b) winter.

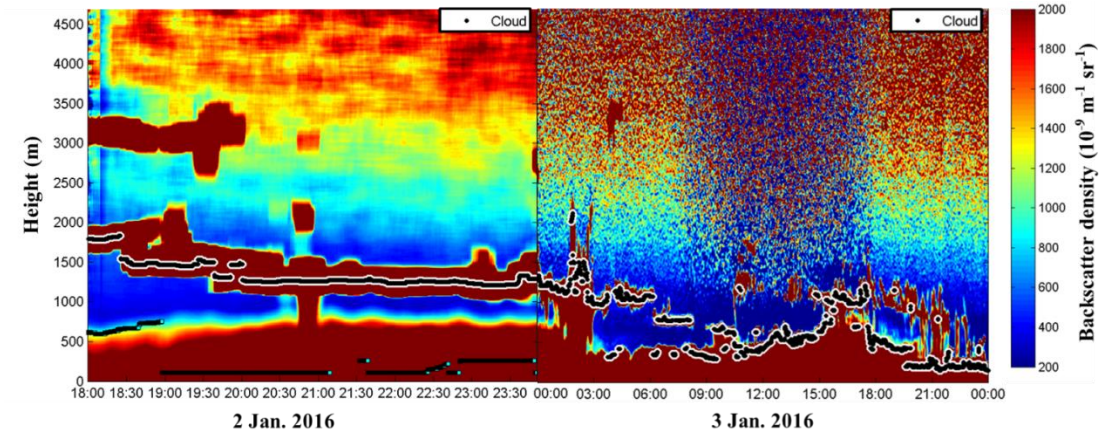


(a) November 18, 2015 (b) November 19, 2015

Figure S10. MODIS images show the cloud covers over the PRD region during the autumn pollution episode (https://earthdata.nasa.gov/earth-observation-data/near-real-time/rapid-response).



(a) January 02, 2016 (b) January 03, 2016



(c) Aerosol backscatter densities measured by ceilometer in Jan. 2 and Jan. 3, 2016.

Figure S11. Cloud cover from MODIS satellite remote sensing and cloud heights measured by ceilometer (Model CL-31, Vaisala Corp.) during the winter pollution episode.

(a)

(b)

Figure ii. Clouds in the (a) autumn haze episode; (b) winter haze episode (photos were taken at the 488 m platform of Canton tower at local time ~ 11:30 am).

• **5.** If upward transport is the problem, then is it valid to use the profiles in Figure 7 to develop the concept shown in Figure 9?
[A]: According to model simulations, airshed convection could induce upward transport, although it is still subjected to large uncertainties regarding the magnitude. Hence, we believe that the conceptual scheme in Figure 9 we propose is valid.

Other comments:
1) Page 1, line 21 – I find the statement starting with "Great progress has recently been made" needs to be referenced. I don't understand how great progress can be made at

ground level, without making it aloft. Ground level and the air above are connected. If you match things well at the ground, but not above, then it is not progress. I suggest the authors think more about their opening statement.

[A]: Thank you and we have rephased this sentence. Some references are in the introduction section.

"Many studies have recently been made on understanding the sources and formation mechanisms of atmospheric aerosols at ground level."

2) Page 2, line 4 - "droplet mode" is a less commonly used term. It has basis, but you need to discuss it first before using it. Also, it presumes the process that you are trying to establish. Use it in your discussion, but I suggest removing 'droplet' here and on line 7 as in my next comment.

[A]: We have deleted "droplet" in line 4.

3) Page 2, lines 7-10 – I suggest "Our results suggest that much of the sulfate and nitrate are formed from aqueous-phase reactions, and we attribute coarse mode nitrate formation at the measurement site to the heterogeneous reactions of gaseous nitric acid on existing sea-derived coarse particles in autumn.

[A]: We have changed the sentence (page 2, lines 7-9):

"Our results suggest that the majority of the sulfate and nitrate is formed from aqueous-phase reactions, and we attribute coarse mode nitrate formation at the measurement site to the heterogeneous reactions of gaseous nitric acid on existing sea-derived coarse particles in autumn."

4) Page 2, lines 10-11 - This sentence is repetitious. Reduce to "Case studies show that in combination with stagnant weather conditions, sulfate and nitrate from aqueous-phase and heterogeneous reactions contribute to haze formation during autumn and winter in the PRD region." Also, define 'PRD'.

[A]: We have changed accordingly.

"Case studies show that in combination with stagnant weather conditions, sulfate and nitrate from aqueous-phase and heterogeneous reactions contribute to haze formation during autumn and winter in the Pearl River Delta (PRD) region."

5) Page 3, lines 8-12 – I believe that 'droplet mode' resulted from "fog" studies. Often, there are significant differences between fog and cloud, and those differences are reflected in residual size distributions. It seems that you are knowledgeable about this, but you need to make it clear in this paragraph.

[A]: We agree that fog processing also leads to droplet mode residual particles. We have changed in the revision:

"The condensation submode particles originate from primary emissions and growth of smaller particles by coagulation and condensation, while droplet submode ones mainly result from cloud/fog processing or coagulation of smaller particles (Seinfeld and Pandis, 2006).

6) Page 4, line 15 – Change "the vertical size-resolved chemical composition" to "size-resolved chemical composition in the vertical"

[A]: It has been changed.

"However, measurements of size-resolved chemical composition in the vertical within the urban boundary layer are still lacking."

7) Page 4-5, lines 28-2 - "Additionally, more consistent evidences of aerosol formation through heterogeneous reactions are needed from field measurements, laboratory experiments and model simulations." Is awkward. Perhaps "Additionally, more studies of aerosol formation resulting from heterogeneous reactions are needed from field measurements, laboratory experiments and model simulations."

[A]: We have rephased in the revision.

"Additionally, more studies of aerosol formation resulting from heterogeneous reactions are needed from field measurements, laboratory experiments and model simulations."

8) Page 5, lines 17-20 – There is no mention of modelling here. Evaluation of the model is not one of your objectives, but the use of modelling in evaluating your observations is part of your work, and I think it should be mentioned.

[A]: We added the model evaluation in the objectives of this study (page 5, lines 20-22).

(3) Evaluating the simulation performance of WRF-Chem model in the vertical based on the measurement data.

9) Page 8, line 25 – page 9, line 4 – I don't see the need for this classification. It adds unnecessary words, and you never use it beyond here. You only have 3 measurement altitudes, and you do not use "Type…" in any of the figures. Just start with ay something like, "We classify the ... components into those peaking at the ground, those peaking at 118 m and those peaking at 488 m", and modify the remainder of the paragraph accordingly. Also, I think these statistics should be in the main text, if you are going to discuss them here.

[A]: Thank you for the suggestion. We have changed as suggested. The present manuscript is very long. We think it's better to put the statistical tables in the supplementary.

10) Page 9, line 15 – "likely"; line 18 – "cleanER"

[A]: Changed.

11) Page 12, lines 15-18 – The proposition you refer to here was put forward decades ago. Re-write as "The coarse-mode nitrate was likely formed ... coarse particles (e.g. Anlauf ...)."

[A]: We agree and have changed in the manuscript (page 12, lines 17-20).

"The coarse-mode nitrate was likely formed the heterogeneous reactions of gaseous nitric acid with pre-existing sea- and soil-derived coarse particles (Anlauf et al., 2006; Harrison and Pio, 1983; Harrison and Kitto, 1990; Pakkanen, 1996; Wall et al., 1988;

Wu and Okada, 1994; Zhuang et al., 1999a)."

12) Page 12, line 20 - 6 - This equation isn't necessary. I suggest removing it and adding the references to the list beginning with Anlauf et al.
[A]: We have removed this equation and relevant sentences.

13) Page 12, lines 21-22 – This sentence repeats what is said four lines above. Remove it, and add the references to the "(Anlauf et al….)" list.
[A]: We have deleted the sentence and added the reference to the list as suggested.

14) Page 12-13, lines 28-1 - At best, "activation" is misleading in this context. Substantial water uptake by coarse NaCl particles can, and more likely occurs without true 'activation'. Replace with something like "Sea salt particles can grow by water uptake in fogs and clouds." I think this entire discussion could be better integrated and shortened, since the preference of HNO3 for NaCl over Calcium compounds that you mention is likely related to water solubility. I would make this point on line 25, and then refer to deliquesced particles, fog droplets and cloud droplets. As it stands now, it is as if the aqueous-phase processes are in addition to whatever drives your finding, stated on lines 22-24, and as further demonstrated by the repetitious statement on line 5 of page 13.
[A]: Thank the reviewer for the valuable suggestions. We rephased this part in the revision (Pages 12-13, lines 20-3).
"We found that coarse-mode Na^+, Cl^-, and NO_3^- were at almost the same particle size, while Ca^{2+} peaked at a particle size larger than NO_3^- (Fig. S3). It is thus reasonable to conclude that coarse-mode NO_3^- is probably associated with sea salt rather than Ca^{2+}, which is consistent with the previous work in Hong Kong (Zhuang et al., 1999b). Sea salt particles can grow by water uptake in fogs and clouds. A previous study showed that a substantial amount of nitrates forms when HNO_3 reacts with deliquesced sea-salt as compared to the dry NaCl particles (Brink, 1998). Hence, we speculated that nitrates were formed from the reactive uptake of HNO_3 in the deliquesced sea salt droplets rather than dry particles in Guangzhou. The back-trajectory cluster analysis showed that the sampled air masses were predominantly from the South China Sea and moved toward Guangzhou in autumn (Fig. S4), bringing high concentrations of sea salt particles available for heterogeneous reactions. Moreover, high relative humidity, fog, and low clouds which were observed during the observation, could facilitate the heterogeneous formation of coarse-mode nitrates."

15) Page 14, line 2 – "were slightly higher"
[A]: We have changed the phrase.

16) Page 14, line 11 – "demonstrating more aged aerosol" - Are you saying this simply because the aerosol was sampled at a higher elevation?
[A]: Our results showed that higher chloride depletion in the higher elevation, suggesting the aerosols were more aged.

17) Page 14, lines 12-15 – Might some of the winter salt particles be attributed to dry salt lakes in the interior of China?

[A]: It is possible that salt particles can be originated from remote dry salt lakes, such as the lakes in Qinghai province in northwestern China. The salt particles from lakes may be carried along with the dust storms, which usually occur in Spring. We think dry salt lakes were not the sources of winter salt particles in our measurement.

18) Page 14, line 18 – "smallER"

[A]: It has been changed.

19) Page 14, lines 21-22 – But the profiles of SO4=and OC differ. Secondary contributions to SO4= appear to be more elevated than OC, at least in autumn.

[A]: We agree that SO_4^{2-} could be more elevated at the higher levels due to the aqueous phase reaction.

Organic aerosols can originate from direct emission (primary organic carbon, POC) or atmospheric secondary formation (secondary organic carbon, SOC). The ground OC sources would impact the vertical distributions of OC. However, sulfate is produced mainly from atmospheric reactions. That could be the reason why SO_4^{2-} and OC had different vertical profiles.

20) Page 14, line 24 - The ratio is determined by 2 quantities. A change in the ratio doesn't necessarily result from a change in just one of those quantities. Maybe the ratio at the source differs, or maybe precipitation near the source remove some of the particles, including EC, after which the sulphate, nitrate and OC are replenished by secondary reactions. The statement, "indicating the presence of secondary organic aerosols", needs more justification.

[A]: Besides the OC/EC ratios, we estimated the secondary organic carbon (SOC) concentrations using the EC tracer method in the revised manuscript. We found that SOC accounted for a large fraction of OC (Figure S6). We added this information in the text, and the EC-tracer method was included in the supplementary.

"The high OC/EC ratios were found in particles with sizes larger than 0.25 μm, especially for droplet mode particles, indicating the enhancement of the secondary formation of OC in this mode. We further evaluated the contributions of secondary organic carbon (SOC) to OC using EC-tracer method (Castro et al., 1999; Zhou et al. 2014). The results showed that SOC accounted for a large fraction of OC in our study (Figure S6)."

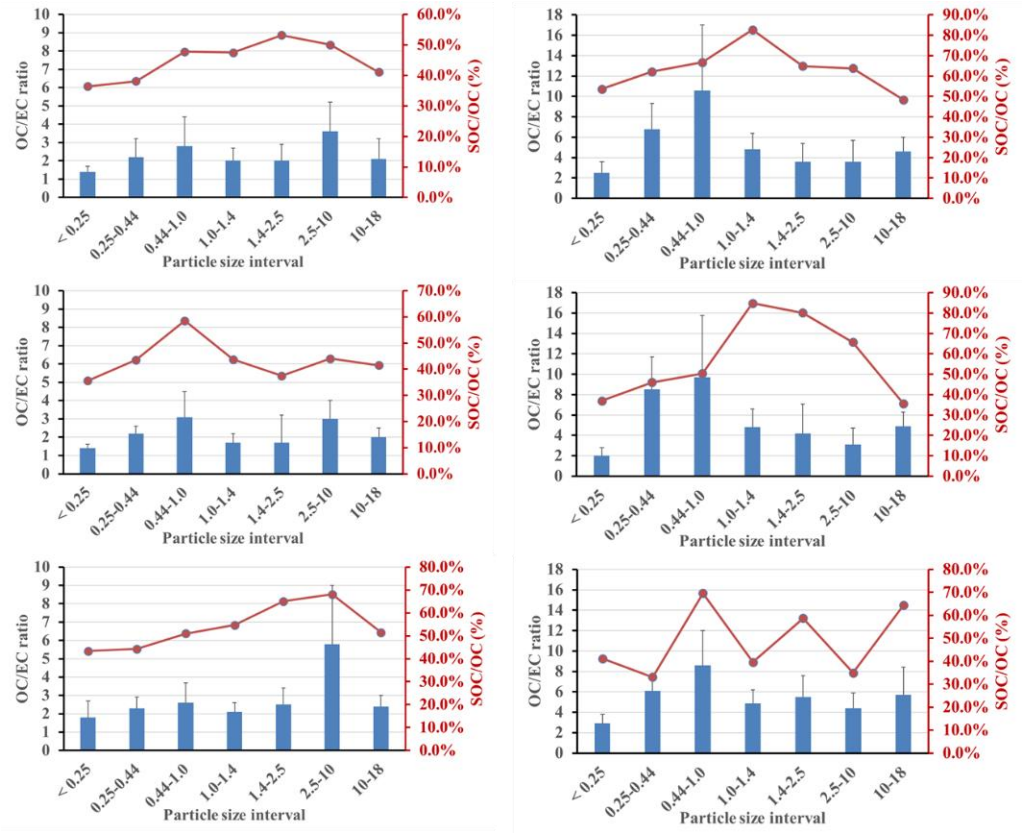


Figure S6. Average OC/EC ratios and percentages of SOC in OC at different sizes during autumn and winter.

3. Estimation of secondary organic carbon concentration

It is difficult to separate primary organic carbon (POC) from secondary organic carbon (SOC). Currently, T no simple, direct analytical technique is available for the separation. Here we applied an indirect method (the minimum OC/EC ratio method) to estimate the secondary organic carbon (OC_{sec}) formation with the following equation (Cao et al., 2004; Castro et al., 1999; Zhou et al., 2014):

$$OC_{sec} = OC_{tot} - EC \times (OC/EC)_{min}$$

where OC_{sec} is secondary organic carbon, OC_{tot} is total organic carbon, EC is elemental carbon, and $(OC/EC)_{min}$ is the minimum OC/EC ratio. The minimum ratio of OC/EC may be affected by many factors such as meteorological conditions, the variation of the emission source, and the transport of aged aerosols. Thus, SOC concentration derived by the EC tracer method could be underestimated in this study. In order to reduce this uncertainty, the $(OC/EC)_{min}$ is defined as the minimum OC/EC ratio of each size interval at each height in autumn and winter respectively.

References:

Cao, J. J., Lee, S. C., Ho, K. F., Zou, S. C., Fung, K., Li, Y., Watson, J. G., and Chow, J. C.: Spatial and seasonal variations of 25 atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China, Atmos. Environ., 38, 4447-4456, https://doi.org/10.1016/j.atmosenv.2004.05.016, 2004.

Castro, L.M., Pio, C.A., Harrison, R.M., Smith, D.J.T., 1999. Carbonaceous aerosol in urban and rural European atmospheres: estimation of secondary organic carbon concentrations. Atmos. Environ. 33, 2771e2781. http://dx.doi.org/10.1016/s1352-2310(98)00331-8.

Zhou, S. Z., Wang, T., Wang, Z., Li, W. J., Xu, Z., Wang, X. F., Yuan, C., Poon, C. N., Louie, Peter K.K., Luk, Connie W.Y., and Wang, W. X: Photochemical evolution of organic aerosols observed in urban plumes from Hong Kong and the Pearl River Delta of China, Atmos. Environ., 88, 219–229, http://dx.doi.org/10.1016/j.atmosenv.2014.01.032, 2014.

21) Page 15, lines 24-25 – What about radiative cooling?

[A]: Radiative cooling usually occurs under the circumstances of cloud free. As showed in *Figure S10 and S11*, there were many clouds during these periods. And, wind directions were changed from south to north (*Figure S8*). Therefore, we thought that temperature inversion in our study should be caused by the convergence of two different air streams.

22) Page 16, line 1 – Should be Fig. 7a?

[A]: It has been changed.

23) Page 16, lines 14-15 – "Our results suggest that aqueous-phase and heterogeneous reactions contributed significantly to the sulfate and nitrate in the PRD region during this episode.

[A]: The sentence has been changed to (page 16, lines 12-13):

"Our results suggest that aqueous-phase and heterogeneous reactions contributed significantly to the sulfate and nitrate in the PRD region during this episode."

24) Page 16, line 2 – As in my first review, this is NOT "strong convection". I suggest "Low-level cloud was observed during this period, associated with weak convection simulated by WRF."

[A]: We have changed in the revised manuscript (page 16, lines 20-21).

"Low-level cloud was observed during this period, associated with weak convection simulated by WRF."

25) Page 17, line 3 – What is meant by "aggravated"?

[A]: We changed to "facilitated".

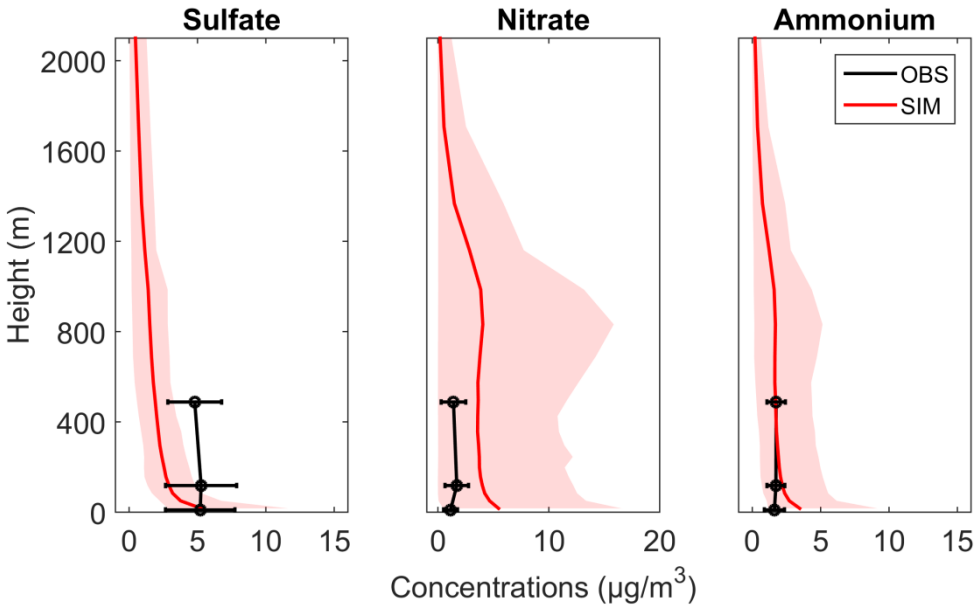
26) Page 17, discussion at end of Section 3.3 – There is no discussion of precipitation in the paper. Was there precipitation? If so, might it have played a role?

[A]: There was no rain during two aerosols pollution events we discussed.

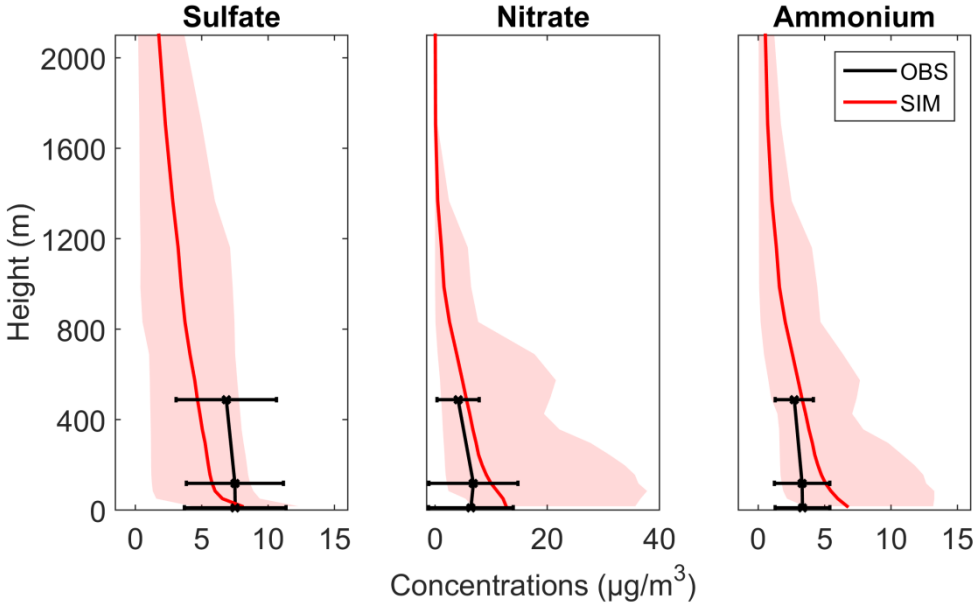
Page 15, lines 13-14: "There was no rain during the pollution episodes."

27) Page 17, line 23 – Figures S13 and S14 should be in the main text.

[A]: We moved these figures to the main text as Figure 10.



(a)



(b)

Figure 10. The vertical concentration profiles of sulfate, nitrate, and ammonium in $PM_{2.5}$ during (a) autumn and (b) winter (The red solid lines are the average modeled concentrations and the shaded regions indicate the minimum and maximum values of the simulation; the average measurement data were in black with horizontal error bars).

28) Page 18, line 21 – "Vertical characteristics and potential formation processes of…"
[A]: This sentence has been changed accordingly.
"Vertical characteristics and potential formation processes of size-resolved aerosols were studied during autumn and winter seasons utilizing the 610 m Canton Tower in Guangzhou."