

Major comments

1) The formation processes of sulfate and nitrate are well known. Thus, the current study does not contribute much to the understanding of the underlying processes that lead to aerosol formation.

In order to make the current study a substantial contribution to our understanding of general implications for atmospheric science rather than being focused on the investigation of local air pollution, the manuscript's focus needs to be expanded.

Referee #4 suggests to apply WRF-Chem not only to predict the meteorological parameters but also to simulate formation of sulfate and nitrate for comparison to measurements. This would provide useful information about model performance and it would broaden the scope of the manuscript as general conclusions on model performance could be drawn using your unique data set of size-segregated vertical aerosol profiles.

2) The formation processes and sources of organic aerosol are much more uncertain than those of sulfate and nitrate. Given that a large mass fraction of the aerosol in the study region is composed of organics, some discussion should be dedicated to them. For example, Referee #4 points out that the different vertical gradients of organics compared to inorganics may give evidence of different sources. This idea should be elaborated on and the discussion extended accordingly.

3) Referee #3 suggests improving the structure of your discussion and to more clearly differentiating between hypotheses and clear evidence of aerosol processing based on your observations. Your statements should be stronger and more specific; at many places they seem vague, e.g.

p. 1, l. 10: The results from pollution case studies further showed that atmospheric aqueous-phase and 10 heterogeneous reactions together with adverse weather conditions, such as temperature inversion and calm wind, resulted in the autumn and winter haze pollution in the PRD region

p. 9, l. 21: However, back trajectory analysis of air masses showed that regional transport was unlikely the important source during the sampling time (Fig. S4) and then local chemical formation was likably the source that led to high SNA mass concentrations.

p. 10, l. 6: indicating the favorable secondary formation or regional transport of aerosols at the higher altitude

p. 17, l. 9: suggesting different nitrate formation mechanisms

Minor comments

- Referee #1 appreciates the detailed uncertainty analysis that you provided in the response to the referee report. It should be included in the manuscript.

- Both Referee #2 and #3 pointed out that the hygroscopic growth of particles might lead to a significant shift in diameters which might affect conclusions about aerosol size distributions at different heights. Please take into account the numbers for growth factors etc as suggested by Referee #3 and discuss possible implications for your conclusions on aerosol mass formation vs aerosol growth by water uptake.

- At several places in the manuscript, you use 'heterogeneous aqueous phase reaction' or seem to use 'heterogeneous' and 'multiphase' reactions equivalently. Please check carefully for the correct use of terminology: 'Heterogeneous processes' are processes that occur on surfaces (droplet, particles) where the

reactants are in two different phases (gas and condensed phase). ‘Multiphase processes’ are processes that occur in the bulk of the aqueous phase into which the reactants might be taken up from or products released to the gas phase (Ravishankara, A. R.: Heterogeneous and Multiphase Chemistry in the Troposphere, *Science*, 276, 1058–1065, 1997.)

- Numbers should be rounded to significant digits throughout the manuscript, e.g. 44 ± 14 instead of 44.1 ± 14.9

- p. 8, l.11: What are abnormal days, as opposed to ‘normal days’?

- Nitrate formation from hydrolysis of N_2O_5 is well known. In the atmosphere, likely no completely dry particles exist as mixed particles have a very low efflorescence relative humidity and continuously undergo efflorescence/deliquescence cycles. The discussion on op. 12 can be shortened.

- p. 14, l. 10-14: These lines seem out of place in the ‘OC and EC’ section.

- p. 15, l. 10-12: Was the increase in SO_2 sufficient to lead to the observed decrease in SOR? In other words, does this imply that no sulfate formation occurred?