

## ***Interactive comment on “Modeling of aerosol property evolution during winter haze episodes over a megacity cluster in northern China: Roles of regional transport and heterogeneous reactions” by Huiyun Du et al.***

**Anonymous Referee #2**

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The authors present results from a model study exploring the mass concentration levels of aerosol, in particular sulfate, over Northern China. They conclude that large fractions of secondary inorganic aerosol (SIA) are formed from SO<sub>2</sub> that has been emitted from Beijing and that is oxidized in other regions and then transported back to the study region. Heterogeneous chemistry contributes a large fraction of this sulfate. In addition to SIA levels, they also look at the aging degree of aerosols, i.e. by the coating of black carbon particles (BC). In general, measurements and model results agree well. The topic of this manuscript is of high interest since sources of aerosol that leads to strong

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haze periods in China are not fully understood yet. However, the study is in large parts obscure and details of the model are not explained which makes it difficult to appreciate the potentially new findings. The manuscript may be acceptable for publication after my comments below will be addressed.

Major comments

1) Only the levels and formation of sulfate are discussed whereas aerosol mass is composed of many more inorganic and organic compounds. While a comprehensive analysis of all aerosol constituents might exceed the scope of the paper, the limitation to sulfate should be made clear in the title, abstract and throughout the manuscript.

2) Several parts of the paper seem disconnected from each other and/or available information is not sufficiently used in the discussion:

a) The transport of SO<sub>2</sub> away from Beijing and its subsequent oxidation followed by transport back to Beijing is an interesting thought. However, it would be much more convincing if HYSPLIT trajectories were included in the discussion showing this recirculation of air masses.

b) Can the different source regions during the various episodes be connected to back trajectories and different emissions in the various source regions?

3) It is not clear what is exactly meant by ‘aqueous’ and ‘heterogeneous’ chemistry. Does aqueous phase chemistry only include cloud chemistry? Which oxidants are considered? Is metal-assisted oxidation included? Is heterogeneous chemistry the oxidation of S(IV) by NO<sub>2</sub> or are other processes included as well? Which parameters are included in the model parameterization? How well are they constrained?

4) Most of the figures need to be improved. Contrasts are hard to see and often they are overloaded with information in much too small font.

Figure 2: What are the horizontal green lines?

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Figure 4: Why is only the range up to 600 nm considered here even though the measurements and model bins extended further?

Figure 5: The solid circles are too small. Are they supposed to be colored as the caption suggests?

Figure 6: The pie charts and numbers are too small. Also the legends should be increased for better readability.

Figure 7: The black numbers on the dark grey pie charts are hard to read.

Figure 9: The text says that the figure shows regional sources. However, here all sources LC, LTC, RTC and RLC are shown.

Figure 11a): I suggest removing the lines between the symbols as they are meaningless and imply a non-existing trend.

Figure 12: - This figure contains way too much information. The numbers in the pie charts cannot be read. At the very least, this figure needs to be increased in size. However, it might be easier to include some of the information in an additional table.

- In the caption, it is not clear what 'black lines' are referred to here.

5) I am confused about the treatment of organics in the discussion of measured and modeled aerosol. It is well known that also organics can be directly emitted from various sources. However, for example, in the caption of Figure 7, OM and PA are separately listed. Please explain somewhere what PA (primary aerosol) includes and how the proportions of primary vs secondary organic aerosol are tracked within the model.

6) Many of the results seem trivial. They should be either presented as such or their novelty should be better highlighted if they indeed are surprising for the particular conditions in the current study.

a) l. 344 ff: It is well known that secondary aerosol exceeds primary aerosol after a short period of aging.

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b) l. 340/341: This sentence is trivial. What other sources could SIA have if not chemical production?

c) It is mentioned that particles are aged within  $\sim 2$  hours. Thus, is it surprising that in most cities a large fraction of PA originated from local emissions (l. 331)?

d) R(BC) for fresh particles should be  $\sim 0$ . Thus, the sentence in l. 457 is trivial.

e) l. 565: 'heterogeneous chemistry played the most crucial role under high pollution levels': It is not clear how you arrive at this conclusion. It is obvious that under high pollution levels (i.e. high SO<sub>2</sub> levels) the contribution of PA might be small. However, it is not evident to me why the absolute contributions of gas and aqueous phase chemistry should not be enhanced equally.

Minor comments

l. 158 ff: It is not clear what it here means. Do you mean 'emission from region i'?

l. 171: Is 'n' the number of all regions. Please specify.

l. 279: Figure S2 only shows SO<sub>2</sub>, not NO<sub>2</sub>.

l. 364/365: I do not understand this sentence.

l. 368 – 374: This text sounds awkward and should be reworded. As it is written it implies that the clean or polluted conditions, respectively, determined the various source regions. However, it would be more reasonable to say that the wind direction from the various source regions led to the transport of the respective air masses into the study region. Because of the transport distance and/or pollution level in the source region, the resulting pollution level in the study region was high or low, respectively.

l. 478: '... this would affect radiation and climate change' should be removed.

l. 488: What does such a high R(BC) in a source region mean? Aerosol transported from that region will always appear aged.

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l. 545: 'the major form of SO<sub>4</sub><sup>2-</sup>' should be replaced by 'the major source of SO<sub>4</sub><sup>2-</sup>'

Table S1: The caption should include more details.

Technical comments

l. 45: 'experiencing' should be 'experienced'

l. 71: 'physicochemical' misspelled

l. 204: Draxler misspelled

l. 235: were obtained

l. 296: 'respectively' misspelled

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Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-1189>, 2018.