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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Heterogeneous sulfate aerosol formation mechanisms during wintertime Chinese haze events: Air quality model assessment using observations of sulfate oxygen isotopes in Beijing" *by* Jingyuan Shao et al.

Anonymous Referee #2

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It has been a long question regarding the origin(s) of the non-zero Δ17O (but also Δ33S and Δ36S values) observed in tropospheric aerosols. This study implements the four majors SO2 oxidation pathways into the GEOS-Chem model in clouds and on aerosols and compared with the sulfates measured in China (coupled with the O-isotopes). This study also discussed the contribution of the different oxidation pathways by estimating the pH in urban aerosols. While current models underestimate the sulfate formation in aerosols, results in this study significantly reduced the difference between observed and modeled in both sulfates concentrations and oxygen isotopes.

However uncertainties remain but it has been identified that O2+TMI oxidation pathway is dominant when pH < 5. Another oxidation pathways on alkaline dust has also been hypothetized (very speculative) to explain the high Δ 17O-values.

General comments:

1) Input of stratospheric sulfates and sulfates resulting from Criegee radicals oxidation pathway are nowadays largely discussed when taking into account the S-isotopes. It is suggested that those processes are responsible for high Δ 33S-values which cannot be explained yet by current knowledge. How would those processes affect the results ? Is it possible to determine their contribution ? I suggest these processes being discussed.

2) The std_run model can account for the seasonality observed in the aerosols. Does the implementation of the four oxidation pathways change the results ? Is it more accurate ? If not, what would be the drivers ?

3) The RHs seem to be an important factor when it comes to understand the sulfate formation. However, as described in the paper, whywould the Sichuan province be the only one to exhibit the largest enhancement of sulfate formation ? Many places in China might be characterized by high humidity and air stagnation ?

4) The model supposes a constant contribution of primary aerosols emitted by the human. However the contribution of each sources might change depending of the seasons which should also affect the O-isotopes (towards 0 ‰). Thus how would this sources variations change the results ?

5) Recent studies suggested that dust particles would be responsible for pollution in China during winter, this certainly due to photooxidation by mineral dust. Does the new models takes into account input of dust mineral from desert etc ? It has also been shown that others reactions occurred on the dust particles besides NO2 and ozone (OH for example). How would it affect the O-isotopes ? What would be the contribution

of this oxidation pathway during the winter ? Would it change the contribution value (9%) ? I suggested this being discussed.

6) Although this study is very interesting and promising, results show that many uncertainties remain when it comes to understand the different oxidation pathways undertaken by SO2. I suggest that the conclusion should be more contrasted as O2+TMI cannot alone account for all the O-isotopes signatures.

7) With all the pollution in China, none of the models predict high pollution in Beijing which is surprising. How could it be explained ?

8) In Figure 1, why the SOR is not correlated to SO4 during the 11/5 ?

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