

***Interactive comment on* “Possible heterogeneous hydroxymethanesulfonate (HMS) chemistry in northern China winter haze and implications for rapid sulfate formation” by Shaojie Song et al.**

Anonymous Referee #2

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This manuscript presents very interesting results for the organic sulfate production during haze periods in northern China. Results from this manuscript clearly showed that nearly all the sulfate measured can be attributed to inorganic sulfate during dry and clean periods while up to one third of total sulfate is attributed to organic sulfate (OS). Among them, hydroxymethanesulfonate (HMS) is likely the major OS species. The results are very intriguing and worthy of being further explored. However, several major issues need to be resolved before the manuscript can be publishable.

1. It seems the title is misleading. The major idea of this paper is to conclude that HMS is likely the major OS species as the results and discussion section clearly followed this

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logic. In addition, no clear conclusion can be made for rapid sulfate production or even oxidation of HMS which leads to the sulfate formation is still speculative. The chemistry itself is not new and all the reactions in the text were cited from literature. Based on this reason, I would suggest the authors to change the title of this paper to something like “Major contribution of Hydroxymethanesulfonate (HMS) to organic sulfate in northern China winter haze”.

2. The authors mentioned that HMS may serve as a reservoir for sulfate, if oxidation of HMS is rapid in the presence of aqueous OH radical, HMS will be quickly converted to sulfate which means that the formation of this intermediate is not important in term of the interpretation of sulfate from the AMS measurements, that is, sulfate from either this pathway or SO₂ oxidation is measured as inorganic sulfate. Only when HMS is present in a significant concentration, it becomes important as a major contributor to OS. The authors need to clarify this point.

3. In addition to HMS, the authors list several categories of organic sulfate including methanesulfonate, sulfones, and organosulfates etc. Are there still any other types of OS which might not be considered because of the limitation of the current measurement techniques? In addition, the authors mentioned that several common organosulfates were not detected by the SPAMS; however, that does not mean that they are not significantly present. It is possible that the SPAMS was not capable of detecting them due to its limitation measurement scheme.

4. According to the thermodynamic rules, ammonia will be titrated before it can be taken up by nitrate aerosols. So why it bothers that these fragment ratios should be related to the ammonium nitrate? (line 25 on p5)

Some minor comments

1. Fig. 2b doesn't show any information on the RH. Do you mean Fig. 2a-b?

2. Line 4 on p6 here miss “to be” between “considered” and “the concentration”

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3. Line 30 on p9 I don't think you can make cloud related statements since the periods are all covered by cloud

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-1015>, 2018.

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