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Song et al. 2020/10/16

The authors thank the valuable comments from the referee #3 very much. We have addressed the comments made corresponding changes to the manuscript.

Comments are in black and responses are in blue.

Response to Anonymous Referee #3

This manuscript presents a global modeling study of heterogeneous chemistry of hydroxymethanesulfonate (HMS) using the GEOS-Chem model. Recently HMS has been detected in a few measurement studies and its concentrations could reach several micron grams per cubic meters. The current study incorporates HMS chemistry into a three-dimensional chemical transport model and shows the spatial and seasonal variations of the modeled HMS. It discusses the major factors, including emission, decomposition, oxidation and pH calculation, leading to the modeled patterns of HMS, which can be referred to the parameterization of other liquid phase reactions. Overall, this manuscript is very well written and should be published in this journal after addressing a few minor revisions as listed below.

We thank the referee for the positive evaluation and very helpful comments. Our responses to the specific comments are provided below.

1. There is a measurement paper by Wei et al. recently published (doi:10.1021/acs.estlett.0c00528). They measured HMS in Beijing using two analytical methods (ion chromatography and UHPLC-LTQ-Orbitrap mass spectrometry). This paper should be mentioned in the introduction and comparison should also be made with the model results.

We have mentioned this paper in the Introduction: "*high mass concentrations of hydroxymethanesulfonate (HMS), a hydroxyalkylsulfonate species, have been detected in winter Beijing, China using an aerosol mass spectrometer by Song et al. (2019a), and using an improved ion chromatography method by Ma et al. (2020**), and using a UHPLC-LTQ-Orbitrap mass spectrometry by Wei et al. (2020).***" In fact, the measurement periods in Wei et al. (2020) and Ma et al. (2020) are overlapped and their results are also consistent. Therefore, the same comparison results are expected between the model and these two measurement studies.

2. The authors reported the simulated pH in cloud water. What is the simulated pH in aerosol water?

The GEOS-Chem modeling analysis uses the ISORROPIA-II thermodynamic equilibrium module to estimate the aerosol pH values. The current analysis has found that HMS is mainly formed and processed in cloud water. The sensitivity simulation that adds the relevant processed in aerosol water does not seem to change the global results significantly. For the sake of simplicity, we do not show the modeled values and distributions of aerosol water pH in this current paper.

3. P2L6: What is the major source of dimethyl sulfide DMS?

We have provided the major source of DMS in this sentence: "*MSA is produced primarily by the oxidation of biogenic dimethyl sulfide (DMS,* ***mainly from marine phytoplankton****) and is likely the major organosulfur species in many regions over the oceans"*.

4. P2L10: Can the authors comment on the relative importance of other hydroxyalkylsulfonate compounds?

The current understanding is that HMS is the most abundant hydroxyalkylsulfonate compounds in the atmosphere. We have made this statement clear in the revised manuscript: "*hydroxymethanesulfonate (HMS****), the most abundant hydroxyalkylsulfonate species commonly found in the atmosphere****, have been detected in winter Beijing, China*". More discussions can be found in the paper Song et al. (2019a) cited in the manuscript. The relative abundance of different hydroxyalkylsulfonate HAS species depends on several factors including the level of carbonyl precursors, aqueous pH values, etc.

5. P9 R27, R28: There is a recent paper by Wang et al. (www.nature.com/articles/s41467-020-16683-x) discussing the fast oxidation of SO2 by NO2 and HONO. Are the reaction rate constants comparable to this study?

Thanks for the question. As shown in Table 1 of the manuscript, we use the same reaction rate constants between SO_2 and NO_2 and HONO with those used in Wang et al. (2020). In fact, both are obtained from the laboratory studies conducted in 1980s. We also cite this paper by Wang et al. (2020) in the revised manuscript.