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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China" *by* B. Zheng et al.

Anonymous Referee #1

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General comments

The authors used WRF and CMAQ model to simulate air quality in North China for a winter month and have described the inability of the current CMAQ model in predicting high observed inorganic aerosol concentrations. They added several heterogeneous reactions into the CMAQ model which then improved the model performance for inorganic aerosol concentrations. They also examined the impact of anomalous meteorological conditions on model predictions. While the article is well-prepared, model

predictions and subsequent conclusions are direct results of the selected uptake coefficients for the heterogeneous reactions. If different uptake coefficients are selected, then model predictions and conclusions will be different.

Specific comments

Page 16733 - Abstract

Abstract should be revised to clearly indicate that selection of uptake coefficients for the heterogeneous reactions is arbitrary and the use of other values over-predicts sulfate compared to observed data.

Page 16736, line 18 (section 2)

Not clear about the meaning of "offline-coupled Weather Research and Forecasting (WRF) model and CMAQ 5.0.1". If the authors used WRF model to generate meteorological fields which were then subsequently used to drive the CMAQ model, then the word "coupled" is misleading. Please clearly describe how they were used.

Page 16738-16741 (section 2.2)

I agree with the comment made by J. F. Muller and also think that the selection of the uptake coefficients is arbitrary and has subsequently resulted in improvement of model performance. The uptake coefficients in the referenced articles deal with dust not sulfate or nitrate. The article should clearly indicate that the selection of the uptake coefficients for the heterogeneous reactions is arbitrary.

Page 16745-16749 (section 4.2-4.4)

While the current model cannot capture the observed sulfate concentrations, the improvement of model predictions is a coincident due to the selection of the uptake coefficients.

The authors have completed model simulations with higher uptake coefficients from Wang et al., 2012. The results of such simulation can be presented so that readers ob-

tain a complete picture of the impacts of heterogeneous reactions on model predictions in China.

The metal catalysis pathway can be important for enhancing wintertime sulfate concentrations (Alexander et al., 2009). The authors have not presented any comparison of predicted Fe and Mn concentrations to observed data in North China. The under-prediction of Fe and Mn can also contribute to the under-prediction of sulfate in China. The heterogeneous nitryl chloride production can also enhance winter hydroxyl level which can subsequently enhance winter sulfate (Sarwar et al., 2014). The impact of such chemistry on winter sulfate in January 2013 in North China is unknown. Chemistry and aerosols can affect meteorological conditions which can subsequently affect pollutant levels (Grell et al., 2005; Wang et al., 2012). Such effects can be especially important in highly polluted conditions. The authors have not examined the impacts of chemistry and aerosols feedback on meteorological conditions and their subsequent impact on pollutants. While the heterogeneous reactions with arbitrarily selected uptake coefficients enhance and improve model performance for inorganic aerosols, these additional factors are likely to further enhance model predictions.

Page 16751-16752 (section 5)

Summary and conclusions need to be qualified to reflect that while the heterogeneous reaction can reproduce the observed data, the uptake coefficients used here are highly uncertain and the use of other available uptake coefficients leads to model over-predictions. The uptake coefficients used in this study were developed for dust particles and have been arbitrarily adopted for this study. Future studies should focus on improving the uptake coefficients for particles relevant to North China. Other potential chemical reactions and feedback of chemistry and aerosols on meteorology can also affect the model predictions which have not been examined in this study. Future studies need to explore impacts of such additional factors on model predictions.

Technical corrections

Page 16734, line 29 and other pages

Should the citation of Y. S. Wang et al., 2014 be written as Wang et al., 2014?

Page 16740, line 23 and other pages

Should the citation of K. Wang et al., 2012 be written as Wang et al., 2012?

Page 16744, 25 and other pages

Should the citation of L. Wang et al., 2010 be written as Wang et al., 2010?

Page 16747, 22 and other pages

Should the citation of X., J. Zhao et al., 2013 be written as Zhao et al., 2013?

References:

Alexander et al., transition metal-catalyzed oxidation of atmospheric sulfur: global implications for the sulfur budget, JGR, 114, D02309, 2009.

Grell et al., fully coupled “online” chemistry within the WRF model. Atmospheric Environment, 39(37), 6957-6975, 2005.

Sarwar et al., importance of tropospheric ClNO2 chemistry across the Northern Hemisphere, Geophysical Research Letters, 41, 4050-4058, 2014.

Wong et al., WRF-CMAQ two-way coupled system with aerosol feedback: software development and preliminary results, Geosci. Model Dev., 5, 299-312, 2012.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 16731, 2014.