

Dear Anonymous Referee,

Thank you for your thorough review of the manuscript. We have read the editor's and the reviewer's comments carefully, taken all of the reviewer's comments into consideration and revised the manuscript accordingly. All the changes have been highlighted in the revised manuscript. Our detailed responses, including a point-by-point response to the reviews and a list of all relevant changes, are as follows:

**Replies to the comments from Anonymous Reviewer 1:**

**Q1: This manuscript presents the assess of the contributions of cloud chemistry to the SO<sub>2</sub> and sulfate levels in typical regions in China using a WRF/CUACE model. Evaluating the cloud chemistry scheme in WRF/CUACE by the in-situ cloud chemistry observations at Mount Tai in summers of 2015 and 2018 in case-1; Quantifying the contributions of cloud chemistry to the SO<sub>2</sub> and sulfate changes in a typical winter pollution month of December 2016 in case-2. This manuscript presents a study of cloud-based SO<sub>2</sub> processing for sulfate formation over China using a regional coupled chemistry-weather model. The chemistry the study is based on is at least 7 years out of date.**

A: Thanks for your comments. The development of cloud chemistry scheme has a long history (J. Seinfeld, Atmospheric Chemistry and Physics, 1997) and is still under active researches as Tilgner et al. ACP 2021 shows.

There are many ways to understand cloud chemistry: by laboratory experiments to disclose the kinetic and mechanize of cloud chemistry in chamber, by field measurements in the similar situation of cloud which is mostly done at high mountain sites, and finally to combine the results of the former two in numerical models to present the mechanism of cloud chemistry simulation and predict the cloud chemistry and their contribution to the atmospheric chemistry and their feedbacks to weather and climates. The scheme used in this paper, i.e. in-cloud oxidant mechanisms by O<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> has been based on a thorough evaluation and used in several regional and global models such as MOZART, NARCM, CMAQ, WRF-CUACE, WRF-Chem, and MC3. Though it is more than 7 years old, the fundamental scheme is still valid and useful.

We agree with the reviewer that some new developments on cloud chemistry have been evolved. A few of them such as GEOS-Chem, ESM2, and CHIMERE have added N-chemical mechanisms, Fe and Mn for TMIs pathway, which we intended to add to our model as well.

**It has been known for many years that cloud water processing is insufficient to explain sulfate formation in China, particularly during heavy pollution events, and aerosol chemistry is crucial for understanding this phenomenon (there is extensive literature on this, summarized, for example, in the reviews by Tilgner et al. ACP 2021 and Liu et al. ES&T 2021). This is not acknowledged or discussed in the manuscript.**

A: Thanks very much for your comments and the information of the two very important references. The two papers by Tilgner et al. ACP 2021 (we have already cited in the manuscript) and Liu et al. ES&T 2021 have presented a very detailed valuable view on acidity in cloud chemistry and the different pathways for cloud chemistry. We agree with the reviewer's point that **“cloud water processing is insufficient to explain sulfate formation in China, particularly during heavy pollution events”**. However, this paper is not intended to demonstrate the dominant contributions of cloud chemistry to the heavy pollution events, but to quantify the relative contributions of cloud chemistry to the SO<sub>2</sub> depletion and sulfate formation in different regions of China as compared to other processes.

We have carefully looked for and read the literatures relative to this content and added these valuable points into our paper as the following:

“Multiphase oxidation of sulfate aerosols from SO<sub>2</sub> in aerosol particles in high humidity environment is one of the main causes of explosive growth of particulate matter in East Asia haze (Guo et al., 2014; Cheng et al., 2016; Song et al., 2019). From observations and laboratory works, there are four main pathways for this kind of oxidation of SO<sub>2</sub> which are H<sub>2</sub>O<sub>2</sub>, O<sub>3</sub>, NO<sub>2</sub>, and transition metal ions (TMIs) (Iibusuki and Takeuchi, 1987; Martin et al., 1991; Alexander et al., 2009; Harris et al., 2013; Cheng et al., 2016; Wang et al., 2016 ). Additional pathways of organic peroxides (ROOH) (Yao et al., 2019; Wang et al., 2019; Ye et al., 2018; Dovrou et al., 2019, photolysis products of nitrate (pNO<sub>3</sub><sup>-</sup>) (Gen et al., 2019a; 2019b), and excited triplet states of photosensitizer molecules (T\*) (Wang et al., 2020) have also been found recently to be important for multiphase oxidation of sulfur dioxide during very heavy hazy days. Unfortunately there are still much uncertainties and gaps to put all of those pathways into model applications from observational and laboratory studies (Pye et al., 2020; Ravishankara, 1997; Liu et al., 2021). Several regional and global models have tried to include only two, O<sub>3</sub> and H<sub>2</sub>O<sub>2</sub>, in-cloud oxidant in cloud chemistry mechanisms, (Park et al., 2004; Tie et al., 2005; Salzen et al., 2000; Chapman et al., 2009; Leighton and Ivanova, 2008). A very few models can simulate the pathway of NO<sub>2</sub>, TMIs of Fe or Mn ions (Ge et al., 2021; Binkowski and Roselle, 2003; Chang et al., 1987; Terrenoire et al., 2015; Menut et al., 2013).”

Together with 22 more literatures added into the reference list:

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aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources, *Atmos. Chem. Phys.*, 9, 945 – 964, <https://doi.org/10.5194/acp-9-945-2009>, 2009.

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**An intercomparison of the simulation results with field data is shown which, unsurprisingly, does not show very good agreement.**

A: Even though previous studies have studied the cloud chemistry and its impacts on sulfate formation in polluted regions in China and pointed out the impacts were insufficient to explain the sulfate pollution. There are still many gaps related with the new pathways discovered by observations and laboratory studies, and the quantitatively evaluation of cloud chemistry and the pathways in numerical models to represent the cloud chemistry in China, especially in different pollution zones which is affected with different emissions, terrains and climates. This is exactly what we try to investigate with our model.

This is true as we all realized the complex formation scheme of sulfate aerosol by many factors, i.e., emission, nucleation, heterogeneous chemistry, cloud chemistry, meteorology etc., which all contributes to the sulfate formation. As the ambient conditions vary greatly from region to region in China, the relative importance of each factor may change a lot depending on the geographic locations of the concerns. This paper is intended to quantify the relative contributions of cloud chemistry to the SO<sub>2</sub> depletion and sulfate formation in different regions of China as compared to other processes. We fully agree with the reviewer's point that the cloud chemistry was not fully responsible for the heavy pollution of sulfate.

From our study we found that:

“The assessment of cloud chemistry contributions to the changes of SO<sub>2</sub> and sulfate in NCP, YRD, PRD and SCB regions by WRF/CUACE revealed that except for PRD, all other three regions are significantly affected by the cloud chemistry, of which the SCB is the most obvious.....

In addition, surface sulfate increases by 40-90% in Beijing, Nanjing, Chengdu and Guangzhou in December during heavy pollution, similar to that of previous studies (Turnock et al., 2019). Above all, the average contribution of cloud chemistry during the pollution period was significantly greater than that for all December. Vertically, the results of the pollution process for the 2016 winter heavy pollution episode showed that the cloud chemistry influence was mainly in the middle and lower troposphere below 5 km for 4 representative cities.

Generally, the cloud chemistry improved the model performance by reducing the SO<sub>2</sub> overestimates and enhancing the correlations with observations for both SO<sub>2</sub> and sulfate.”

**The presentation quality is also low, with multiple typographical errors.**

A: Thanks for your comments. We have reviewed the whole manuscript and corrected a lot of typographical errors and polished a lot of incorrect sentences in the abstract and the main context together with the figure and equations to make the manuscript more friendly to readers. Here are some screen shots of the corrected parts:

For the abstract:

20 A regional online chemical weather model WRF/ CUACE (China Meteorological Administration Unified Atmospheric  
Chemistry Environment) was used to assess the contributions of cloud chemistry to the SO<sub>2</sub> and sulfate levels in typical  
regions in China. By comparing with several time series of in-situ cloud chemical observations on Mount Tai in Shandong  
Province of China, the CUACE cloud chemistry scheme was found to well reproduce the cloud processing the consumptions  
of H<sub>2</sub>O<sub>2</sub>, O<sub>3</sub> and SO<sub>2</sub>, and consequently was used in the regional assessment for a heavy pollution episode and monthly  
25 average in December 2016. During cloud availability in heavy pollution episode, the sulfate production increases 40-80% and  
SO<sub>2</sub> reduces over 80%. ~~During the heavy pollution episode, it is found that~~And the cloud chemistry mainly affects the  
middle and lower troposphere below 5 km as well as within the boundary layer, and contributes significantly to SO<sub>2</sub> reduction  
and sulfate increase in east-central China. Among ~~the the four typical polluted zones in China~~regions of North China Plain  
(NCP), Yangtze River Delta (YRD) and Sichuan Basin (SCB), the ~~Sichuan Basin (SCB)~~SCB is mostly affected by the cloud  
30 chemistry, with the average SO<sub>2</sub> abatement up to about 1-15 ppb and sulfate increase up to more than 50  $\mu\text{g}/\text{m}^3$ , followed by  
~~Yangtze River Delta (YRD)~~YRD where the contribution of cloud chemistry is still significant, averaging up to 1-3 ppb for  
SO<sub>2</sub> abatement and 5-20  $\mu\text{g}/\text{m}^3$  for sulfate increase. The cloud chemistry contribution to ~~Pearl River Delta (PRD)~~ PRD and

## For the introduction:

There has been very serious air pollution in central-east China where four heavy pollution ~~regions-zones~~ of North China  
Plain (NCP), Yangtze River Delta (YRD), Sichuan Basin (SCB) and Pearl River Delta (PRD) are located (Yao et al., 2021;  
10 Zhang et al., 2012). Although many global and regional models ~~have contained~~include sulfate formation mechanisms in cloud  
chemistry, few models have assessed its contribution, especially the lack of detailed assessment of regional cloud chemistry  
on sulfate and SO<sub>2</sub> in China and those ~~four typical~~ pollution regions. ~~Many~~regional chemical ~~transport~~ models have  
reported the ~~over-estimate~~results of SO<sub>2</sub> ~~over-estimate~~ problems (Buchard et al., 2014; He et al., 2015; Wei et al., 2019; Sha  
et al., 2019; Georgiou et al., 2018). ~~T~~The inadequate inclusion or lack of cloud chemistry of SO<sub>2</sub> consumption simulations is  
15 one of the main ~~problems-causes~~ (Ge et al., 2021). Therefore, there is a very important need to ~~accurately~~ quantify the  
contribution of cloud chemistry in these ~~typical regions~~ and ~~thein~~ central-east China when it comes to heavily polluted  
weather to get a better understand of multi-dimensional pollution ~~interactions~~distributions, especially ~~in-between~~ the upper  
layer ~~besides and~~ the surface. <sup>↵</sup>

This study is intended to use an on-line coupled chemical weather platform of CMA<sup>+</sup>, WRF/CUACE, to analyze and  
10 evaluate the SO<sub>2</sub> in-cloud oxidation process in the four ~~pollution~~ regions ~~in China~~, with two objectives: (1) evaluating the  
cloud chemistry scheme in WRF/CUACE by the in-situ cloud chemistry observations at Mount Tai in summers of 2015 and  
2018; and (2) quantifying the contributions of cloud chemistry to the SO<sub>2</sub> and sulfate changes in a typical winter pollution  
month of December 2016 ~~with a very long lasting heavy pollution episode~~. It is aimed to establish a system to assess the  
relative contribution of cloud chemistry to SO<sub>2</sub> oxidation pathways and sulfate productions to other clear-sky processes. <sup>↵</sup>

And added the star sign to figure:

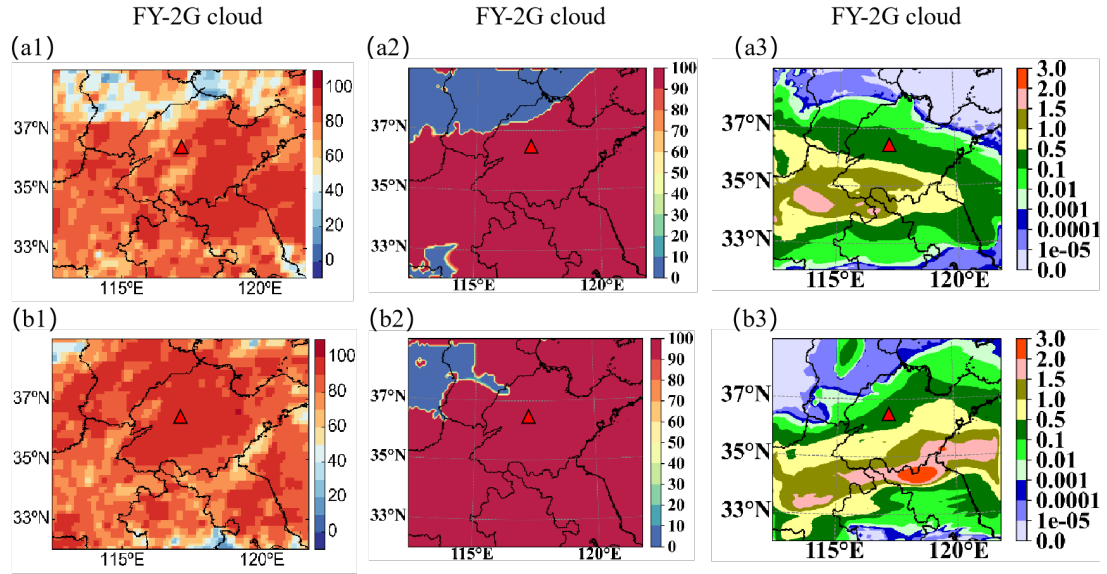


Figure 1. (a1, b1): the cloud image of FY2G, (a2, b2): the cloud fraction by WRF/CUACE. (a3, b3): the liquid water content by WRF/CUACE (Units: g m<sup>-2</sup>). (a) is for 8:00 LST on 24 June, (b) is for 8:00 LST on 25 June.

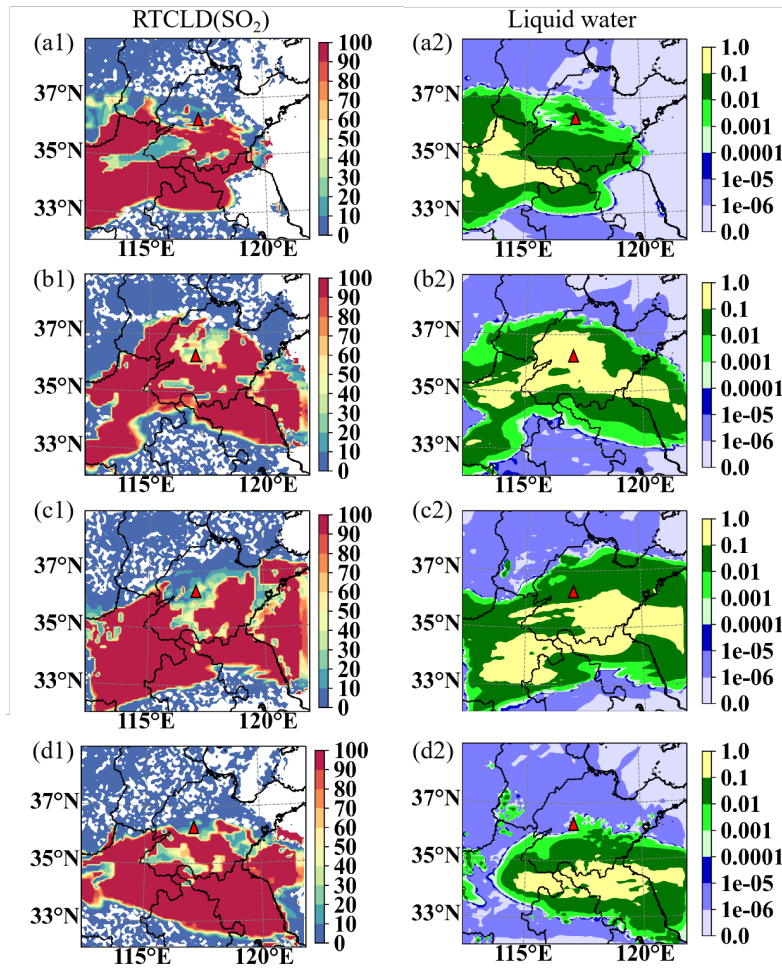


Figure 4. The percentage of SO<sub>2</sub> oxidized (a1, b1, c1 and d1) (%) and the liquid water content (a2, b2, c2 and d2) (g/kg) by WRF/CUACE, where (a) is for 2:00 LST on 24 June, (b) is for 8:00 LST on 24 June, (c) is for 2:00 LST on 25 June and (d) is for 8:00 LST on 25 June.