

Response to the Comments of Referee #3

Revealing the sulfur dioxide emission reductions in China by assimilating surface observations in WRF-Chem

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We would like to thank to the reviewer for giving constructive criticisms, which are very helpful in improving the quality of the manuscript. We have made minor revision based on the critical comments and suggestions of the referee. The referee's comments are reproduced (black) along with our replies (blue) and changes made to the text (red) in the revised manuscript. All the authors have read the revised manuscript and agreed with submission in its revised form.

Anonymous Referee #3

Comment NO.1: *The manuscript used the Four-Dimensional Local Ensemble Transform Kalman Filter (4D-LETKF) and WRF-Chem to dynamically update the SO₂ emission grid by grid over China by assimilating the ground-based hourly SO₂ observations. The topic is relevant and useful, and the results help reduce the uncertainty of emission inventory and improving the forecasting of SO₂. I recommend this paper for publication after the following points are addressed.*

Response: We thank the referee for this very positive assessment of our manuscript.

Comment NO.2: *Since the implementation of strict emission mitigation strategies in 2013, there is a large reduction of SO₂. These reductions are primarily caused by the relocation and/or phased out of power plants and high-emitting industrial factories. In Fig. 6, the SO₂ both with MIX and the inverted emissions were underestimated around Gansu. It is not clear that the system works well when the prior emissions were underestimated. And if the locations of emission sources have been relocated, such as the factories or power plants are built/abandoned, does the assimilation method works well?*

Response: Agree. The underestimation of the surface SO₂ concentration with the original MIX emission over northwestern China such as the Gansu province is potentially attributable to the increasing SO₂ emissions due to energy industry expansion and relocation over northwestern

China. The SO₂ emissions and surface concentrations over the Gansu province are increased to reduce the negative biases in the assimilation experiments as shown in Figs. S4 and S6 in the Supplement, indicating our emission inversion system also works well when the prior emissions are underestimated. However, the simulated surface SO₂ concentrations with the inverted emissions are still underestimated over the Gansu province. The reason for the underestimation is twofold: (1) there are limited observations to be assimilated over northwestern China because the observation sites are sparse; (2) the initial priori MIX SO₂ emission over northwestern China is small and underestimated, inducing the model uncertainty is small relative to the observation one. This translates to a reduced impact of the observation on the priori emission.

Changes in Manuscript: Please refer to the revised manuscript, Page 12 Lines 396-405.

Comment NO.3: *In fig. 10, FR_CM with inverted emission and H50kmT1h10Ps recalculation were similar. And the results show that the simulated SO₂ with inverted emission were always less than observation for all sites. Could that be explained?*

Response: Yes, it could be explained. The simulated SO₂ surface concentrations in all sites with the inverted emission in both the FR_CM and assimilation recalculation are generally underestimated. This is due to the inverted emission is sufficient to reduce the overestimations of SO₂ concentration over the priori SO₂ emission hotspot regions but insufficient to eliminate the underestimations over northwestern China.

Changes in Manuscript: Please refer to the revised manuscript, Page 14 Lines 444-447.

Comment NO.4: *Please add a) b) c) : : : etc. in figure 5, 8 and 10. And the legend of Fig.11 NCP (red line) was an error.*

Response: Done. We have corrected the legend of Fig. 11.

Changes in Manuscript: Please refer to the revised manuscript, Figure 5, 8, 10, and 11.

Comment NO.5: *P9L265 Please add the last access date.*

Response: Done.

Changes in Manuscript: Please refer to the revised manuscript, Page 10 Line 297.