Response to the Comments of Referee #2

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 #2

Comment NO.1: *The "top-down" emission inventories of air pollutants such as sulfur dioxide are crucial to the studies of air quality prediction and emission control policy. The authors develop an emission inversion system based on the WRF-Chem model and 4D-LETKF assimilation method. This system is tested by inverting SO_2 emissions with the surface observations. It takes the advantages of considering the nonlinear sulfur chemistry by ensemble forecasts with perturbed emissions, generating the flow-dependent model errors, and localizing the observation impacts. To optimize the assimilation system, the authors also make a lot of efforts to tune the inversion system parameters. The performances of this system are evaluated by comparing to the independently updated "bottom-up" emissions. Results show that the spatial distribution and magnitude of the SO_2 reductions over China are both well revealed by this system. This emission inversion system and its application are sound, and the results are convincing. I would like to recommend accepting this study after some minor revisions.*

Response: We thank the referee for this very positive evaluation.

Comment NO.2: *In ensemble data assimilation, the inflation of background covariance or the analysis covariance is generally required to avoid filter divergence. Do you use any inflation in your assimilation system? Please clarify this.*

Response: Yes, we use the inflation of the analysis covariance in our assimilation system. We

have added the multiplicative inflation factor ρ in formula (4), and the inflation factor ρ is fixed at 1.1 to inflate the analysis covariance as same as our previous studies.

Changes in Manuscript: Please refer to the revised manuscript, Page 6 Lines 173-174.

Comment NO.3: *P5L155: As this paper employs the 4D-LETKF method, it would be helpful to clarify the '4D' /temporal features and 'L'/ spatial localization in the formulas of this method.*

Response: Done. We have clarified the '4D' /temporal features and 'L'/ spatial localization in the formulas (3) and (4).

Changes in Manuscript: Please refer to the revised manuscript, Page 6 Lines 174-177.

Comment NO.4: *P4L132: Do you also nudge the meteorological fields in the PBL?*

Response: The meteorological fields in the Planetary Boundary layer (PBL) are not nudged.

Changes in Manuscript: Please refer to the revised manuscript, Page 5 Lines 138-139.

Comment NO.5: P*5L155: Does the I in formula (4) represent the identity matrix?*

Response: Yes, the *I* in formula (4) represents the identity matrix.

Changes in Manuscript: Please refer to the revised manuscript, Page 6 Line 171.