Response to the Comments of Referee #1

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

Tie Dai, Yueming Cheng, Daisuke Goto, Yingruo Li, Xiao Tang, Guangyu Shi, and Teruyuki Nakajima

We would like to thank 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 #1

Comment NO.1: This manuscript developed a new emission inversion system based on 4D-LETKF and WRF-Chem to update the SO$_2$ emission by assimilating the ground-based hourly SO$_2$ observations. The inverted SO$_2$ emission over China in November 2016 is well in agreement with the “bottom-up” estimation, indicating that the newly developed emission inversion system can efficiently update the SO$_2$ emissions based on the routine surface SO$_2$ observations. Their investigation is interesting and valuable. The manuscript is well written and structured. I recommend publication after addressing the following concerns.

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


Response: Accept. We have added the references in the revised manuscript.

Changes in Manuscript: Please refer to the revised manuscript, Page 2 Lines 61-64.


Response: Accept. We have added the references in the revised manuscript.

Changes in Manuscript: Please refer to the revised manuscript, Page 3 Lines 80-83.

Comment NO.4: Line 142: How do you decide the locations of the super-observations?

Response: The locations of the super-observations are assumed as the locations of the covered model grid cells.

Changes in Manuscript: Please refer to the revised manuscript, Page 5 Lines 149-150.

Comment NO.5: Line 143: How do you decide the assimilated and independent verification observation sites?

Response: The assimilated and independent verification observation sites are randomly decided.

Changes in Manuscript: Please refer to the revised manuscript, Page 5 Lines 151-152.

Comment NO.6: Line 180: How does the emission model forecast the emissions $E^f_{t_{n+1}}$ for 12 hours? How the temporal and spatial distribution of the ensemble spread of the emissions $E^f_{t_{n+1}}$ ranged? Could you please show time series of hourly ensemble spread of the emissions $E^f_{t_{n+1}}$ from 00:00 UTC 8 November to 00:00 UTC 18 November 2016 and their spatial distributions at typical time. Please discuss the forecast model first since the DA depends on the details of the forecast model.

Response: The optimized SO$_2$ emission ensemble $E^a_{t_n}$ has SO$_2$ emissions at 12 hourly
timeslots, which are used to calculate the first guess SO$_2$ emission ensemble $E_{t_{n+1}}^f$ in sequence for the next assimilation cycle.

Time series of the hourly ensemble spreads of the forecast SO$_2$ emissions averaged over China from 00:00 UTC 8 November to 23:00 UTC 17 November 2016 are shown in Fig. S1 in the Supplement. Spatial distributions of the ensemble spreads of the forecast SO$_2$ emissions at 00:00 UTC November 13 are shown in Fig. S2 in the Supplement.

As shown in Figs. S1 and S2 in the Supplement, the temporal and spatial distributions of the ensemble spread of the forecast emissions $E_{t_{n+1}}^f$ are significantly sensitive to the assimilation system parameters. The SO$_2$ emission inversion depends on the forecast model, therefore, sensitivity experiments for various different emission forecasts are conducted to tune the assimilation system as given in Table 1.

**Changes in Manuscript:** Please refer to the revised manuscript, Page 6 Lines 194-199.

**Comment NO.7:** Line 181-183: Please write a bit more about the generation of the initial prior ensemble of SO$_2$ emissions. And also a bit more about the spatial distribution of the ensemble spread of the prior emissions $E_{t_0}$.

**Response:** Done. The initial prior ensemble of SO$_2$ emission is generated by perturbing the freely public available MIX Asian inventory $S$ for November 2010. For example, the SO$_2$ emission for ensemble member $i$ at a given location $(x, y)$ is calculated as $f_i(x, y)S(x, y)$, and the perturbation $f_i(x, y), \{i = 1, 2, ..., k\}$, follows a lognormal distribution in the $k$-dimensional space. The mean and the variance of the perturbations $f(x, y)$ are equal to 1 and the MIX SO$_2$ uncertainty (i.e., 35%). The horizontal perfect correlated and random uncorrelated perturbations are both created to generate the initial prior ensemble $E_{t_0}$ and the associated first guess SO$_2$ emission ensemble $E_{t_{n+1}}^f$. For the horizontal perfect correlated perturbations, same random perturbation factor $f_i(x, y)$ throughout the whole domain emission grids including vertical and temporal spaces per member is applied. For the horizontal random uncorrelated perturbations, the perturbation factor $f_i(x, y)$ is generated independently in horizontal space but dependently in vertical and temporal spaces. The spatial distribution of the ensemble spread of the $E_{t_0}$ with either horizontal perfect correlated or random uncorrelated perturbations has the similar pattern as the MIX Asian inventory $S$, which is generally equal to 35% multiplying
Changes in Manuscript: Please refer to the revised manuscript, Page 7 Lines 199-207.

Comment NO.8: Line 197-200: The SO\textsubscript{2} concentrations are updated “by recalculation of the WRF-Chem ensemble with the optimized emissions”: so the uncertainties of the forecast SO\textsubscript{2} concentrations could still be large. This will influence the assimilation results. Please discuss a bit more about them.

Response: Done. Theoretically, the uncertainties of the forecast SO\textsubscript{2} concentrations by recalculation of the WRF-Chem ensemble are dependent on the optimized emissions. Lower uncertainties of the initial SO\textsubscript{2} conditions for the next assimilation cycle should be found with higher accurate optimized SO\textsubscript{2} emissions, which in turn makes the SO\textsubscript{2} emission inversion more reasonable. Sensitivity experiments for the SO\textsubscript{2} emission inversions as described in section 3 are performed to choose the best assimilation system parameters.

Changes in Manuscript: Please refer to the revised manuscript, Page 7 Lines 224-229.

Comment NO.9: Line 225-230: The spatial correlations among the grid points of the forecast emissions are not clear, so are the spatial correlations among the initial prior ensemble of SO\textsubscript{2} emissions. Figure 3: Which data are used to obtain the averaged SO\textsubscript{2} emissions? Could you please show the difference between the analysis and MEIC 2016, or the ratio?

Response: The spatial correlation coefficients among the initial prior ensemble of SO\textsubscript{2} emissions over every two model grids are equal to one, and this makes the spatial correlations among the grids points of the forecast emissions are also equal to one.

In Figure 3, the inverted SO\textsubscript{2} emissions of each assimilation experiment are obtained by averaging the ones over the ensemble members. The spatial distributions of the mean differences of the MIX and inverted SO\textsubscript{2} emissions minus the MEIC ones are shown in Fig. S3 in the Supplement, and the spatial distributions of the mean ratios between the inverted SO\textsubscript{2} emissions and the MIX ones are shown in Fig. S4 in the Supplement.

Changes in Manuscript: Please refer to the revised manuscript, Page 8 Lines 257-259 and Page 10 Lines 307-310.

Comment NO.10: Line 250: Are the initial and lateral boundary chemical fields perturbed?

Response: Since we don’t know the uncertainties of the global model MOZART-4/GEOS-5, the initial and lateral boundary chemical fields are not perturbed in this study.
**Changes in Manuscript:** Please refer to the revised manuscript, Page 9 Lines 282-284.

**Comment NO.11:** Line 272: Could you please show time series of hourly SO$_2$ emissions of the prior, the forecast and the analysis of the assimilation experiments from 00:00 UTC 8 November to 00:00 UTC 18 November 2016, not only the mean spatial distribution in Figure 3. These will make the reader to understand a priori value and the adjustment SO$_2$ emissions easily. Figure 6 and 7: I guess the SO$_2$ concentrations are obtained from the DA experiments. But I am not sure if they are the updated results by recalculation the WRF-Chem ensemble with the optimized emissions. Could you please show the difference between the updated concentrations and the original?

**Response:** Done. The time series of the hourly SO$_2$ emissions averaged over China of the initial MIX prior, the forecast and the analysis of the assimilation experiment H50kmT1hE10Ps from 00:00 UTC 8 November to 23:00 UTC 17 November 2016 are shown in Fig. S5 in the Supplement, which illustrates the adjustment of SO$_2$ emissions with data assimilation. The SO$_2$ concentrations in each assimilation experiment are obtained by averaging the ones over the WRF-Chem ensemble recalculations with the optimized emissions. The spatial distributions of the mean SO$_2$ concentrations simulated with the original MIX emissions and the updates of the simulated SO$_2$ concentrations with the inverted SO$_2$ emissions are shown in Fig. S6 in the Supplement.

**Changes in Manuscript:** Please refer to the revised manuscript, Page 10 Lines 310-313 and Page 11 Lines 360-365.

**Comment NO.12:** L391: Could you please show the diurnal variations of the inverted SO$_2$ emissions of the DA experiments?

**Response:** Done. The diurnal variations of the inverted SO$_2$ emissions over China and the NCP subregion are also shown in Fig. 11c.

**Changes in Manuscript:** Please refer to the revised manuscript, Page 14 Lines 449-450.