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# Inverse modeling of $SO_2$ and $NO_x$ emissions over China using multisensor satellite data – Part 1: Formulation and sensitivity analysis

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## Averaging kernel impacts on data assimilation

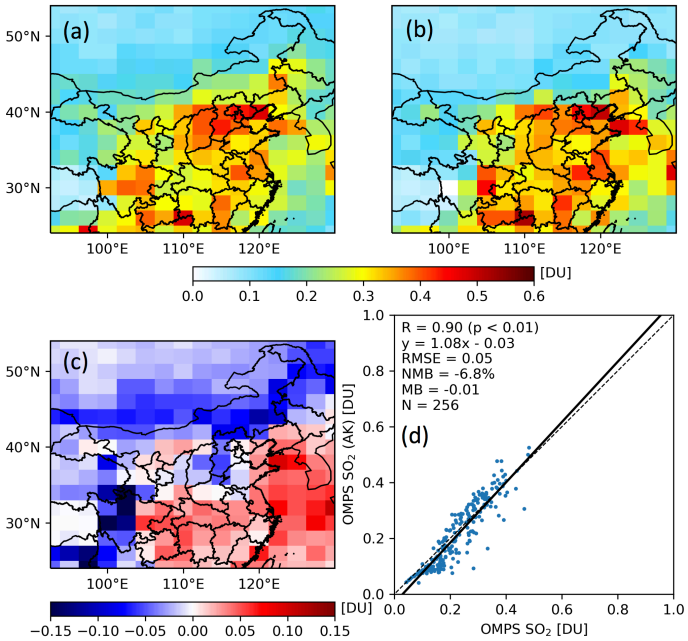


**Figure S1. OMPS $SO_2$ Vertical Column Density (VCD) retrievals in Arpil 2018. (a) and (b) are operational VCDs and the VCDs that are modified through averaging kernel according to formula S1, respectively. (c) is the differences between the modified and operational VCDs. (d) is scatter plot of modified VCDs versus operational VCDs. Linear correlation coefficient (R), linear regression equation, root mean squared error (RMSE), normalized mean bias (NMB), mean bias (MB), and number of observations (N) are shown over the scatter plot.**

$$\Omega_v^{new} = \Omega_v^{opl}(1 + (\boldsymbol{A} - \boldsymbol{1}) \cdot (\boldsymbol{S}^{opl} - \boldsymbol{S}^{new})) \qquad \text{(S1)}$$

Equation S1 is used to calculate modified $SO_2$ ($NO_2$) vertical column density $\Omega_v^{new}$. In equation S1, $\Omega_v^{opl}$ is OMPS operational $SO_2$ ($NO_2$) VCD, $\boldsymbol{A}$ is averaging kernel vector, $\boldsymbol{1}$ is a vector with all elements of 1, $\boldsymbol{S}^{opl}$ is $SO_2$ ($NO_2$) is the shape factor that is used in OMPS trace gas retrieving process, and $\boldsymbol{S}^{opl}$ is $SO_2$ ($NO_2$) is shape factor from GEOS-Chem simulation of this paper.

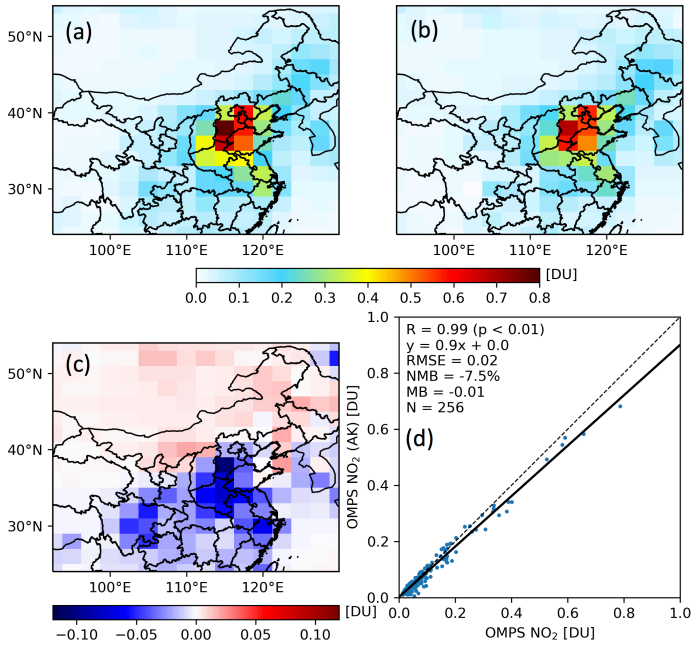


**Figure S2. OMPS $NO_2$ Vertical Column Density (VCD) retrievals in October 2013. (a) and (b) are operational VCDs and the VCDs that are modified through averaging kernel according to formula S1, respectively. (c) is the differences between the modified and operational VCDs. (d) is scatter plot of modified VCDs versus operational VCDs. Linear correlation coefficient (R), linear regression equation, root mean squared error (RMSE), normalized mean bias (NMB), mean bias (MB), and number of observations (N) are shown over the scatter plot.**

## The iteration halt criterion

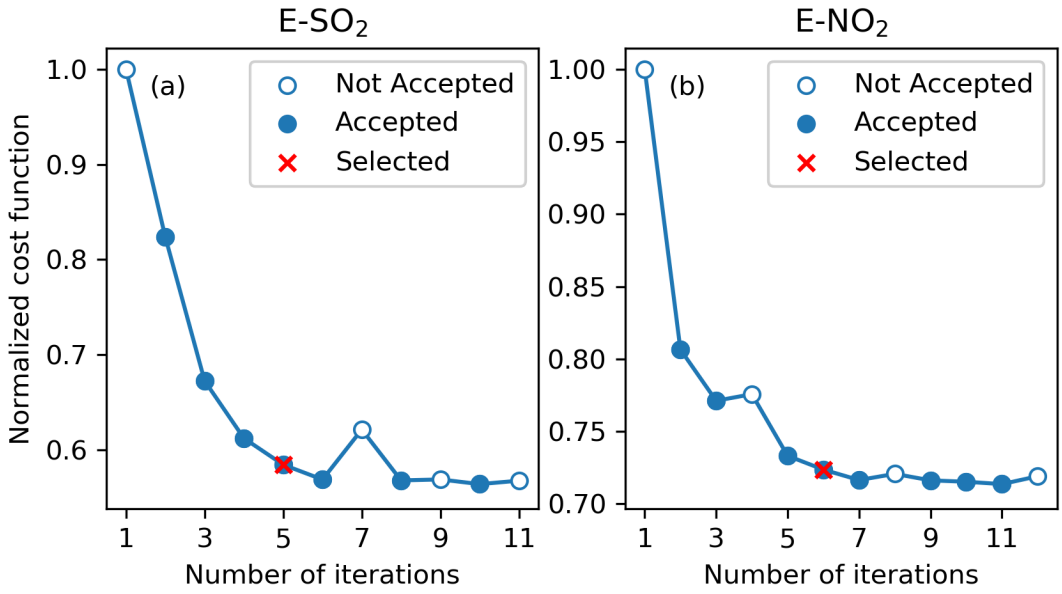


**Figure S3. Normalized cost function (the ratio of the cost function at a iteration to that at the 1st iteration) for E-$SO_2$ (a) and E-$NO_2$ (b). A iteration is accepted (solid circle) if its cost function value is smaller than that of any previous iterations, otherwise not accepted (empty circle). The 1st iteration (prior) is defined as not accepted. The iterations that are selected based on the halt criterion are marked with red cross.**

We choose less than 3% reduction in the cost function between two successful iterations as a criterion to halt the minimization. This selection is to expediate the computation while still maintain the similar accuracy for the optimization; further tests show that the more iterations (after <3% reduction of cost function) doesn't yield discernable difference in the cost function values (Fig. S3) and optimization results (Table S1 and S2).

**Table S1. Anthropogenic $SO_2$ emissions for October 2013 from E-$SO_2$ at each iteration.**

| Iteration | 1 (prior) | 5 (selected) | 6 | 7 | 8 | 9 | 10 | 11 |
|---|---|---|---|---|---|---|---|---|
| $SO_2$ [Gg S] | 1166 | 748 | 744 | Not accepted[a] | 746 | Not accepted | 749 | Not accepted |

[a]Posterior emission total amount at the iteration that is not accepted (cost function value is not smaller than that of any previous iterations) is not shown.

**Table S2. Anthropogenic $NO_x$ emissions for October 2013 from E-$NO_2$ at each iteration.**

| Iteration | 1 (prior) | 6 (selected) | 7 | 8 | 9 | 10 | 11 | 12 |
|---|---|---|---|---|---|---|---|---|
| $NO_x$ [Gg N] | 714 | 672 | 667 | Not accepted[a] | 666 | 666 | 666 | Not accepted |

[a]Posterior emission total amount at the iteration that is not accepted (cost function value is not smaller than that of any previous iterations) is not shown.

## Averaging kernel (scattering weight) impacts on evaluation

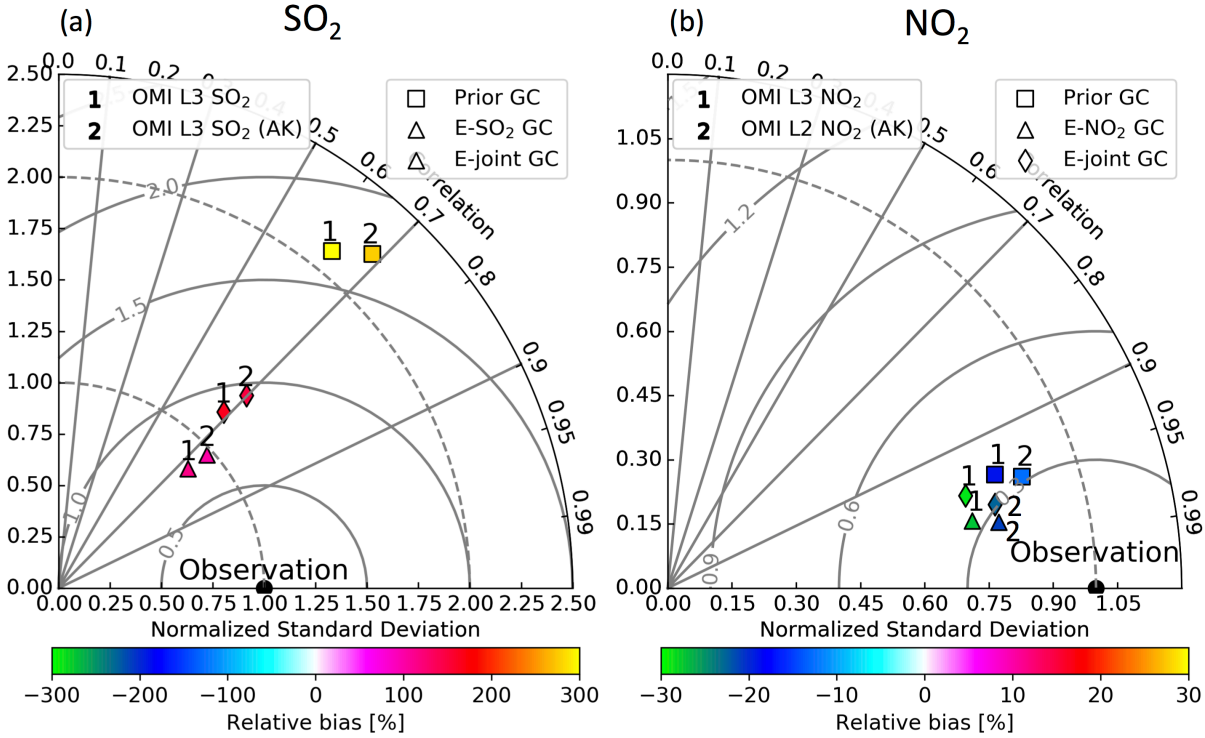


**Figure S4. Taylor diagrams for comparing of VCDs of $SO_2$ (a) and $NO_2$ (b) from the GEOS-Chem simulations (squares for prior, triangles for posterior E-$SO_2$ (a) or E-$NO_2$ (b), and diamonds for E-joint) with that from the OMI (label 1 for operational level 3 $SO_2$ (a) or level 3 $NO_2$ (b) and label 2 for the level 3 $SO_2$ (a) that are modified by considering the vertical profiles from the GEOS-Chem simulation with which is to be compared or the level 2 $NO_2$ (b) that are modified by considering the vertical profiles from the GEOS-Chem simulation with which is to be compared) in October 2013 over China.**

$$\Omega_s^{opl} = \Omega_v^{opl} \cdot M^{opl} \quad \text{(S2)}$$

$$M^{new} = \boldsymbol{W} \cdot \boldsymbol{S}^{new} \quad \text{(S3)}$$

$$\Omega_v^{opl} = \frac{\Omega_s^{opl}}{M^{new}} \quad \text{(S4)}$$

Equation S2 is used to convert OMI $SO_2$ ($NO_2$) vertical column density $\Omega_v^{opl}$ to $SO_2$ ($NO_2$) slant column density $\Omega_s^{opl}$ by multiplying $SO_2$ ($NO_2$) air mass factor $M^{opl}$ from OMI product. Equation S3 is used to calculate new $SO_2$

($NO_2$) air mass factor $M^{new}$, where $\boldsymbol{W}$ is $SO_2$ ($NO_2$) scattering weight, and $\boldsymbol{S}^{new}$ is $SO_2$ ($NO_2$) shape factor that from the GEOS-Chem simulation with which is to be compared. Equation S4 is used to calculate new OMI $SO_2$ ($NO_2$) vertical column density $\Omega_v^{opl}$.

## $NO_2$ lifetime

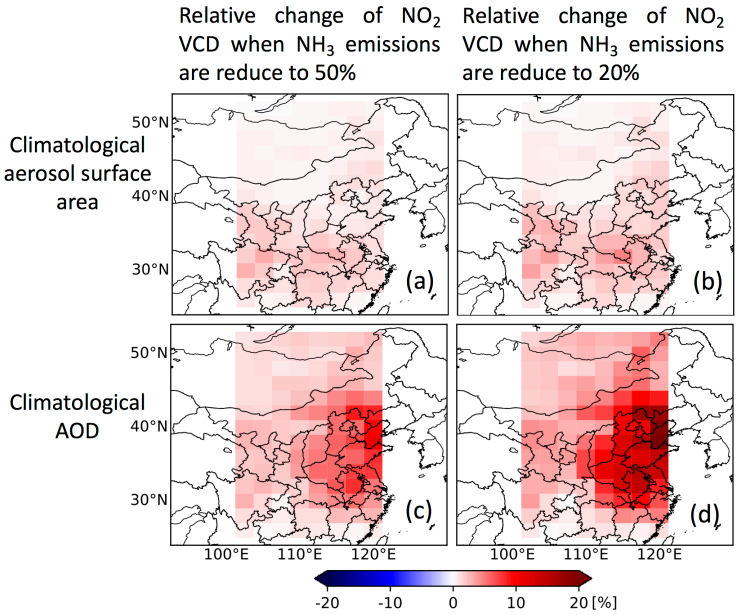


**Figure S5. Relative change of GEOS-Chem $NO_2$ VCDs when $NH_3$ emissions reduce to 50% (a) and 20% (b), respectively at OMPS overpassing time, and a dataset of climatological aerosol surface area is used, regardless of scenarios of $NH_3$ emissions. (c) and (d) are similar to (a) and (b), respectively, but a dataset of climatological AOD rather than climatological aerosol surface area is used.**

Figure S5 a and b show the relative change GEOS-Chem $NO_2$ VCDs with $NH_3$ emissions reducing to 50% and 20%, respectively, in the situation that a dataset of climatological aerosol surface area is used, regardless of scenarios of $NH_3$ emissions, hence reflecting the impact of photolysis of $O_3$ and $NO_2$ on $NO_2$ lifetime when $NH_3$ emissions are reduced. The situation that a dataset of climatological AOD is used is shown in Fig. S4 c and d ; thus they are the impact of $N_2O_5$ chemistry on $NO_2$ lifetime. Apparently, the impact of photolysis of $O_3$ and $NO_2$ on $NO_2$ lifetime caused by the reduction of $NH_3$ emissions is negligible compared to that of $N_2O_5$ chemistry.