

Reply to reviewers and editors:

We thank all reviewers for their careful reading of the manuscript, and for their many constructive feedbacks. The original comments by reviewers are in black font, our replies are in blue.

Reviewer #2

This manuscript presents joint inversion results of SO₂ and NO_x emissions over China using the GEOS-Chem adjoint model and OMPS satellite observations for October 2013. The inversion results were compared against assimilated OMPS observations and independent OMI observations. Several sensitivity calculations were conducted to optimize the joint inversion framework. The joint inversion approach is unique, while the comparison against the OMI observations is interesting. I would, however, advise the authors to revise the manuscript. These revisions should be made before the manuscript can be considered for publication in ACP.

Thanks for the positive comments. We did our best to address them in the revision.

[Major comments]

The model horizontal resolution (2°x2.5° resolution) is clearly too coarse for current regional (not global) emission research, which could lead to serious problems for many applications (e.g., systematic biases in the downscaling analysis (Part 2)). In the previous study by the author's group (Qu et al, 2019), regional Chinese regional emissions were estimated at 0.5°x0.667° resolution using a hybrid 4D-Var/Mass balance approach to save computational resources for the multiple-year calculations, while conducting a one-month adjoint calculation at 0.5°x0.667° resolution using the same adjoint model with a nested domain for East Asia. In the same way, one-month inversion calculation at 0.5°x0.667° resolution using OMPS observations must be doable and should be tested in the present study. This is essential for evaluating the joint inversion performance using in-situ observations (please see my comment below), as already performed by Qu et al. (2019) for OMI assimilation results. It could also provide improved information (e.g., reduced systematic errors for each grid point, considering the non-linear chemistry) for down-scaling analysis (Part 2). For long-term emission estimations, the authors could still use the hybrid inversion framework at 0.5°x0.667° resolution (together with the

downscaling approaches, if resolutions higher than at $0.5^\circ \times 0.667^\circ$ resolution are needed). Thus, I don't think the coarse resolution regional joint inversion will be needed for any applications. At the very least, $0.5^\circ \times 0.667^\circ$ resolution joint inversion calculations should be performed for key experiments.

Thanks for the good suggestions. We acknowledge that it is better to optimize emissions directly at fine resolution (such as $0.5^\circ \times 0.667^\circ$, and $0.25^\circ \times 0.3125^\circ$) rather than coarse resolution (such as $2^\circ \times 2.5^\circ$), and that it is doable for one-month inversion at fine resolution. After careful consideration, we think it is both practical and reasonable to assimilate OMPS retrievals at the resolution of $2^\circ \times 2.5^\circ$, as OMPS pixel size could be much larger than fine-resolution grid boxes (and OMI whose pixel size is 13 km x 24 km at nadir and 26 km x 128km at edge). OMPS pixel size is 50 km x 50 km at nadir, and becomes 190 km x 50 km at edges. Thus, OMPS pixel size is comparable to (at nadir) or much larger than (at edges) $0.5^\circ \times 0.667^\circ$ grid box. An OMPS pixel may cover several $0.5^\circ \times 0.667^\circ$ grid boxes and cannot resolve variations of concentrations owing to variations emissions at that fine resolution. Currently, the nested GOES-Chem adjoint model only supports the $0.5^\circ \times 0.667^\circ$ GEOS-5 meteorological field and the $0.25^\circ \times 0.3125^\circ$ GEOS-FP meteorological field. And the GEOS-5 meteorological field has a temporal coverage from 2004 to mid-2013; data after mid-2013, e.g., our study time period, is unavailable. Although $0.25^\circ \times 0.3125^\circ$ GEOS-FP meteorological field is up to date, apparently its resolution is too fine to compare with OMPS. We have added this explanation in Sect. 3.2.

The joint inversion results, including those from the sensitivity calculations, need to be evaluated against independent in-situ measurements, in order to obtain the optimized system. For this, the authors need to use their $0.5^\circ \times 0.667^\circ$ resolution joint inversion system. Resolutions higher than $0.5^\circ \times 0.667^\circ$ would be required for reducing representation gaps, as discussed in Part 2.

Nevertheless, Qu et al (2019) already demonstrated that joint inversions at $0.5^\circ \times 0.667^\circ$ resolution can be evaluated using in- situ surface observations. This is also essential for evaluating possible biases in both OMPS and OMI satellite observations, which can be one of the most important results from the present study.

As answered the previous question, due to large pixel size of OMPS, we didn't attempt to constrain the emissions at the resolution finer than the satellite instrument on the monthly basis. In addition, for those retrievals at high resolution (such as TROPOMI), the retrieval uncertainty is expected to

be alleviated after aggregating pixel-level retrieval into the coarser resolution. Furthermore, even if the inversion were conducted at the resolution of $0.5^\circ \times 0.667^\circ$ or $0.25^\circ \times 0.3125^\circ$, it is still very challenging to evaluating possible bias in both OMPS and OMI satellite observations, as representation gaps still exists in the two resolutions. Zheng et al. (2017) showed that surface SO_2 (NO_2) concentration simulations from WRF-CMAQ, when evaluating with in situ observations, have a NMB of -23% (0%), 7% (32%), and 41% (45%) at the resolutions of 36 km ($\sim 0.36^\circ$), 12 km ($\sim 0.12^\circ$), and 4 km ($\sim 0.04^\circ$), respectively; this shows that representation gaps still exist at 12 km ($\sim 0.12^\circ$), which is already finer than 0.5° and 0.25° . Thus, perhaps it is not surprising that large negative bias exists when evaluating posterior GEOS-Chem $0.5^\circ \times 0.667^\circ$ simulations with in situ SO_2 (Fig. 10 in Qu et al. (2019)) and NO_2 (Fig. 11 in Qu et al. (2019)) in Qu et al. (2019), and it is somewhat assertive to conclude that the negative bias imply the negative bias of OMI SO_2 and NO_2 retrievals. In Qu et al. (2019), the improvements of posterior simulations when evaluating with in situ SO_2 and NO_2 surface concentrations are mainly represented by Normalized Mean Square Error (NMSE) rather than bias.

The goal of this paper is NOT to replicate the method by Qu et al. (2019). Rather, the goal of this paper is to illustrate how OMPS data could be used to improve an air quality *forecast* through monthly update of emissions (possibly in near real time manner) at a resolution much finer than OMPS. Hence, if implemented, our method of using $2^\circ \times 2.5^\circ$ resolution for performing the optimization can save considerable computational time (and is much more feasible for a research group such us in the university), and then using the downscaling method (part II developed by this study), the finer resolution forecast can be made in a practical manner (suitable for a regional modeling group for air quality forecast). In contrast, the focus of Qu et al (2019) is the re-analysis of emissions, as opposed to forecasting of air quality at the finer scales. In contrast, optimization at $0.5^\circ \times 0.67^\circ$ will still require downscaling method for air quality forecast (normally at ~ 10 km resolution). We have added the elaborations above in section 3.2.

Although the joint inversion reduced the total computational cost, its scientific benefits (required for ACP, not for GMD) are not very clear. The discussions in Sections 4.4 and 4.5 are

interesting. Adding evaluations using any AOD, NH₃, and relevant observations would be helpful to demonstrate the scientific value of the joint inversion.

We have expanded discussion of the scientific benefits of the approach in response to the first comment from the reviewer 1, see above. Further, ACP's scope is very broad and developing data assimilation techniques for using new satellite data has been published in ACP, such as Chen et. al (2018). Please look at this part I and part II paper as a whole – they effectively showcase an approach that is economic in computation to use OMPS data to improve air quality forecast at fine scale. In part II, we did many independent evaluations. Furthermore, we also studied the results of the sensitivity to NH₃ – a topic that has not been studied before in data assimilation. We consider they have good scientific merits.

[A few more specific comments]

L203 “In this study, OMPS SO₂ and NO₂ tropospheric VCDs are retrieved using the shape of NO₂ vertical profiles from GEOS-Chem simulations (Yang et al., 2013; Yang et al., 2014), although differences of model version, simulation year, and emission inventory still exists” These profiles can be largely different. The lack of averaging kernel in the observation operator can lead to serious problems. Please justify and demonstrate its impacts. Otherwise, data assimilation adjustments can be meaningless.

Thanks for pointing out this. We totally agree that differences of model version, simulation year, and emission inventory could lead to profile differences. Thus, we compare operational OMPS retrievals with VCDs modified through averaging kernel to investigate how much VCD differences are caused by profile differences. We have added the discussion below to Sect. 4.1.1 and figures below to supplement.

For SO₂

The SO₂ NMB (106.5%) between GOES-Chem prior simulation and OMPS is much larger than the NMB (-6.8%, Fig S1) caused by the difference of SO₂ vertical profiles between OMPS SO₂ retrieval algorithm and current prior simulation; thus averaging kernel is not considered in the OMPS SO₂ observation operator.

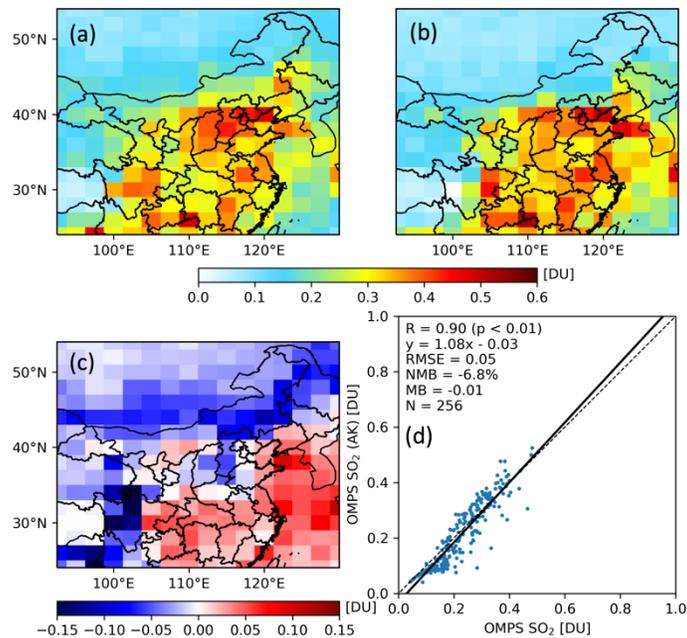


Figure S1. OMPS SO₂ Vertical Column Density (VCD) retrievals in April 2018. (a) and (b) are operational VCDs and the VCDs that are modified through averaging kernel according to formula S1, respectively. (c) is the differences between the modified and operational VCDs. (d) is scatter plot of modified 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.

For NO₂

Similarly, the averaging kernel is not considered in the OMPS NO₂ observation operator for optimization for the following reasons. First, the OMPS NO₂ retrieval differences due to the profile differences can lead to a NMB of -7.5% (Fig S2), which is still smaller than the prior GEOS-Chem simulation NMB (10.9%, Fig. 31). Second, a NMB of 10.9% for model NO₂ VCD simulation is not a very large value, as the difference between satellite NO₂ VCD retrievals and ground-based measurements could be comparable to this value. For example, Krotkov et al. (2017) shows that OMI NO₂ VCD retrievals, on average, are ~10% larger than ground-based FTIR spectrometer. Thus, current research should mainly focus on the change of the spatial distribution (such as linear correlation coefficient) rather than bias of prior and posterior GEOS-Chem NO₂ VCD simulation. Finally, given that linear correlation coefficient between OMPS retrievals and that are modified through integration of averaging kernel and NO₂ vertical profile from this study is as large as 0.99, averaging kernel is not treated in the OMPS NO₂ observation operator.

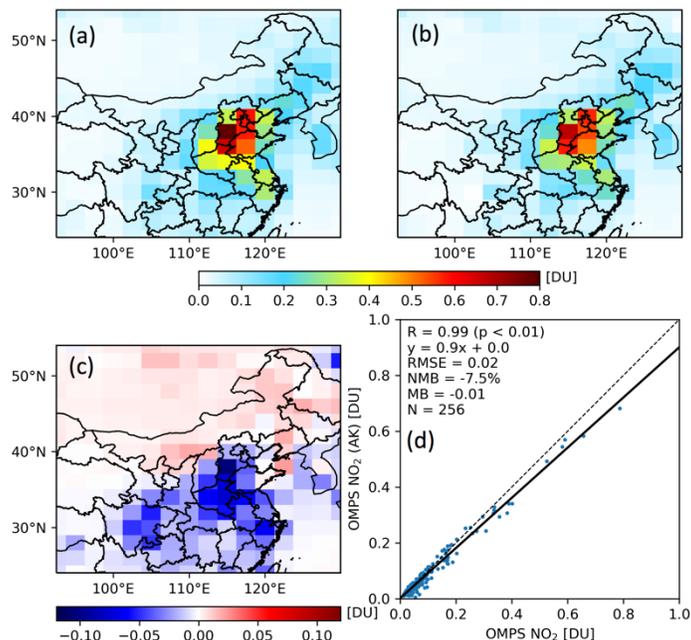


Figure S2. OMPS NO₂ Vertical Column Density (VCD) retrievals in October 2013. (a) and (b) are operational VCDs and the VCDs that are modified through averaging kernel according to formula S1, respectively. (c) is the differences between the modified and operational VCDs. (d) is scatter plot of modified 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.

L204 “Hence, the difference between the GEOS-Chem simulations and the OMPS retrievals is mostly ascribed to the uncertainty of the emissions.” This may not be true and requires further investigation.

We acknowledge that when averaging kernel is not considered in the observation operator, the profile differences can contribute to the difference between the GEOS-Chem simulations and the OMPS retrievals. Additionally, GEOS-Chem model uncertainty can also contribute to the difference between the GEOS-Chem simulations and the OMPS retrievals, and it is difficult to estimate model uncertainty. Thus, we delete the sentence in the revised manuscript; we added the discussion in Sect. 4.1.1 to show that difference between the GEOS-Chem simulations and the operational OMPS retrievals is larger than difference between operational OMPS retrievals and retrievals modified through consideration of averaging kernel. Therefore, the inverse modeling results are statistically significant. We also acknowledge that GEOS-Chem model uncertainty affects inverse modeling results, thus we apply optimized emission inventory to another model (different version of the GEOS-Chem model with much finer resolution) to show that improvement of air quality simulation and forecasts is obtained though the uncertainty of

models. In addition, we also follow the suggestion of evaluating the optimized emission inventory in a consistent framework for part II manuscript; please see details in our reply for part II manuscript.

OMI L3 data is used for validation. Without applying the averaging kernels, comparisons may not provide meaningful information. This needs to be investigated.

Thanks for the suggestion. We acknowledge there should be differences between apply and do not apply averaging kernels (or scattering weights) in the comparisons. Following the suggestion, we investigate how this affect evaluation. In the manuscript, OMI L3 SO₂ and OMI L3 NO₂ are used for evaluation. In OMI L3 SO₂ dataset, only the best pixel in a 0.25°x0.25° grid cell is retained, and the observational geometry information for the pixel is also available. Thus, we can still apply scattering weights to OMI L3 SO₂. In OMI L3 NO₂ dataset, observational geometry information it not available, thus we can only apply scattering weights to OMI L2 NO₂. The text and figures below are added to supplement. We have emphasized in the main text that these conclusions do not change in Sect. 4.1.3.

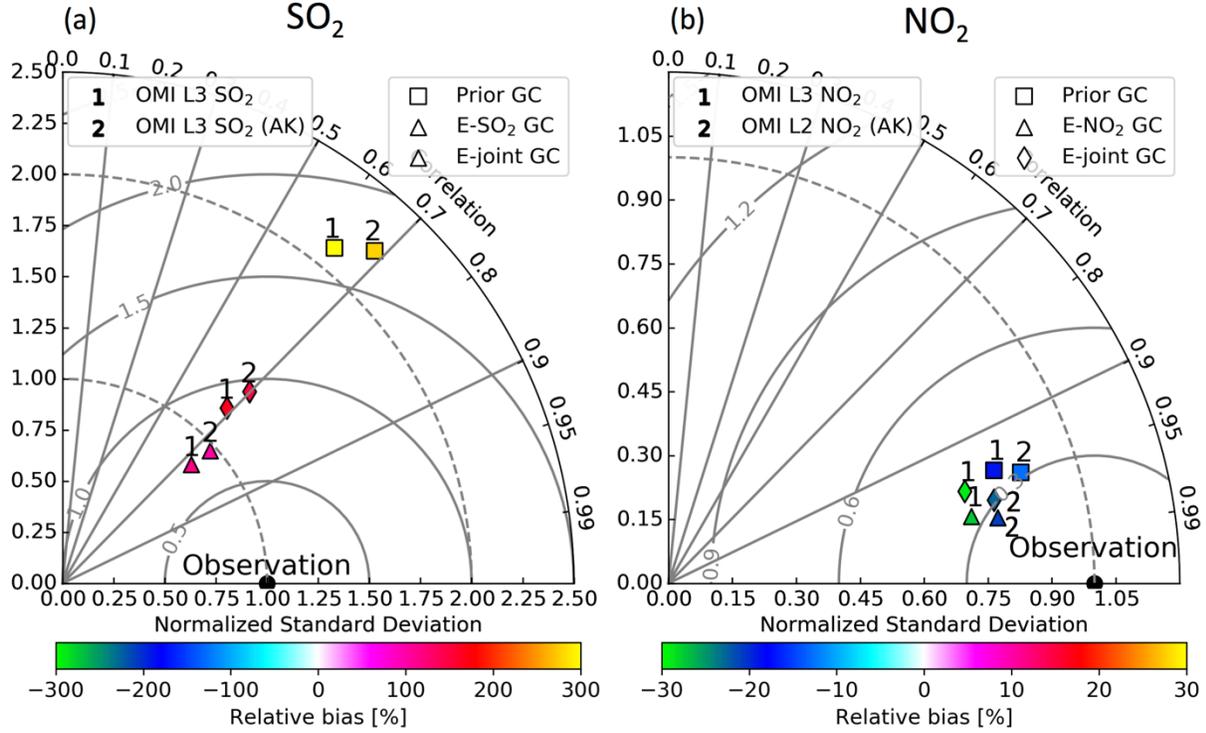


Figure S4. Taylor diagrams for comparing of VCDs of SO₂ (a) and NO₂ (b) from the GEOS-Chem simulations (squares for prior, triangles for posterior E-SO₂ (a) or E-NO₂ (b), and diamonds for E-joint) with that from the OMI (label 1 for operational level 3 SO₂ (a) or level 3 NO₂ (b) and label 2 for the level 3 SO₂ (a) that are modified by considering the vertical profiles from the GEOS-Chem simulation with which is to be compared or the level 2 NO₂ (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.

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

$$M^{new} = \frac{S^{new}}{W} \quad (S3)$$

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

Equation S2 is used to convert OMI SO₂ (NO₂) vertical column density Ω_v^{opl} to SO₂ (NO₂) slant column density Ω_s^{opl} by multiplying SO₂ (NO₂) air mass factor M^{opl} from OMI product. Equation S3 is used to calculate new SO₂ (NO₂) air mass factor M^{new} , where W is SO₂ (NO₂) scattering weight, and S^{new} is SO₂ (NO₂) shape factor that from the GEOS-Chem simulation with which is to be compared. Equation S4 is used to calculate new OMI SO₂ (NO₂) vertical column density Ω_v^{opl} .

Whether use OMI data without applying scattering weight (Label 1 in Fig. S4) or OMI data with applying scattering weight (Label 2 in Fig. S4), the main conclusions in Sect. 4.1.3 does not change. These conclusions are:

- (1) For SO₂, posterior GEOS-Chem simulations (E-SO₂ and E-joint) show smaller NMB and better spatial distribution (in terms of NCRMSE) than prior GEOS-Chem simulations when evaluating with OMI SO₂ (apply or not apply scattering weight).
- (2) For NO₂, the improvements for E-NO₂ and E-joint are reflected in terms of R when evaluating with OMI tropospheric VCDs (apply or not apply scattering weight), although the two experiments show larger negative NMB than the prior simulation.

L325 and some other places, “Our finding of a large reduction. . .“ The discussion about trends between the 2013 October inversion and the 2010 inventories does not make any sense.

Thanks for pointing out this. The MIX 2010 inventory was derived through bottom-up approach, while the 2013 October inversion inventory is derived through integration of GEOS-Chem adjoint model and OMPS SO₂ and NO₂ vertical column density retrievals. We have to acknowledge that systematic bias exists in both of the inventories, and so, the difference between the two emission inventories should not be considered as trend. To investigate trends, emission inventories should be derived from the same approach. In the revision, we therefore have removed the discussions and emphasized that the differences should not be considered as trends.

L330 “in some model grid cells”: Please discuss the spatial pattern.

Sure. We have added “Although the relative difference between E-joint and E-NO₂ proved to be less than 2% in terms of total anthropogenic NO_x emissions over China (Fig. 4k), it is up to 40% over Shanxi province, and both grids with large positive differences and grids with large negative differences exist over North China Plain (Fig. 4 l).” in section 4.1.2.

Section 4.2 does not provide very useful information and can be removed or shortened.

Thank you for the comment. This section shows the impacts of data quality control and spatial balance. Although it does not include much scientific information, it helps to understand how observational errors are assumed. Thus, we hope to keep it.

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