

First of all, we would like to warmly thank the reviewer for her/his time in improving our work through helpful and suggestive comments.

This paper updates existing SO<sub>2</sub> emission inventories over China by using OMI observations and CTM. New source areas missing from the bottom-up inventories are identified and SO<sub>2</sub> emission trends are interpreted. However, it is not very easy for readers to follow the contents, in particular the methodology part. I strongly suggest the authors spend some time on improving this part.

General comments:

1. The introduction section needs to be improved. I suggest focusing on literatures related to authors' own work, instead of a very general introduction. The relationship between the previous studies and this work needs to be clarified. More recent work, e.g., Krotkov et al., 2016, van der A et al., 2017, needs to be included.

Introduction expanded as requested.

2. The method developed by Martin et al., 2003 works very well for NO<sub>x</sub>, because NO<sub>x</sub> lifetime is relatively short and it does not bring significant uncertainties by ignoring transport between grid cells. However, this is not the case for SO<sub>2</sub>. A further analysis is necessary to convince the method is still solid for SO<sub>2</sub>.

The issue is known to the authors and we have long discussed it also with esteemed colleagues in the field. In contrast to the equivalent recent NO<sub>x</sub> emission estimates by the Martin technique [see for e.g. Zyrichidou et al., 2015<sup>1</sup>], we are working on a coarser 0.25x0.25 degree grid. However, since both the a priori emissions as well as the modelling inputs are on a monthly scale, we were unable to configure a way to quantify any smearing effect due to transport [daily effect]. Hains et al., 2008<sup>2</sup>, provide a global scale estimate for the SO<sub>2</sub> lifetime to be 19 ± 7 h, while Fioletov et al, 2015<sup>3</sup>, provide a range of 4h to 12h for the lifetime for SO<sub>2</sub>. Other studies (Lee et al, 2011<sup>4</sup>) show even larger variability for the lifetime of SO<sub>2</sub>, between 16 and 40h. Considering this large range of estimates for the lifetime of SO<sub>2</sub> we can only claim that our estimates should be valid for the lower lifetime estimates of SO<sub>2</sub> and of course this range of uncertainty in the SO<sub>2</sub> lifetime would be a main source of uncertainty in our a posteriori estimates. We have added an explanatory section at the end of section 2.2 on the matter.

3. In section 4.2, the authors tabulate the significant differences between inventories, but without any explanations for the reasons. I suggest a similar analysis as conducted in your recent work (Ding et al., 2017) to explore the possible reasons.

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<sup>1</sup> Zyrichidou, I., M.E. Koukouli, D. Balis, K. Markakis, A. Poupkou, E. Katragkou, I. Kioutsioukis, D. Melas, K.F. Boersma, M. van Roozendaal, Identification of surface NO emission sources on a regional scale using OMI NO, Atmospheric Environment, <http://dx.doi.org/10.1016/j.atmosenv.2014.11.023>.

<sup>2</sup> Hains, J. C., B. F. Tabumann, A. M. Thompson, J. W. Stehr, L. T. Marufu, B. G. Doddridge, and R. R. Dickerson (2008), Origins of chemical pollution derived from mid-Atlantic aircraft profiles using a clustering technique, Atmos. Environ., 42, 1727–1741, doi:10.1016/j.atmosenv.2007.11.052.

<sup>3</sup> Fioletov, V. E., C. A. McLinden, N. Krotkov, and C. Li (2015), Lifetimes and emissions of SO<sub>2</sub> from point sources estimated from OMI. Geophys. Res. Lett., doi: 10.1002/2015GL063148, 2015.

<sup>4</sup> Lee, C., R. V. Martin, A. van Donkelaar, H. Lee, R. R. Dickerson, J. C. Hains, N. Krotkov, A. Richter, K. Vinnikov, and J. J. Schwab, SO<sub>2</sub> emissions and lifetimes: Estimates from inverse modeling using in situ and global, space-based (SCIAMACHY and OMI) observations, J. Geophys. Res., doi:10.1029/2010JD014758, 2011.

This is indeed the next logical step in this work, one which we are already undertaking. First results were presented to the scientific community during the 18<sup>th</sup> GEIA conference in Hamburg in September 2017 [presentations online here: <http://www.geiacenter.org/community/geia-conferences/2017-conference>] and we are actively working on a comparison paper, following the logic of the work performed for NO<sub>x</sub> in Ding et al., 2017. However, we feel that adding this material to this paper would render it rather long and beyond the scope which is to introduce the new emission inventory.

Specific comments:

1. Page 2, line 14, the meaning of “usable manner” is confusing. Please consider rephrasing it.

You are correct, line simplified.

2. Page 2, line 38, please consider rephrasing “emission fields”.

Line rephrased.

3. Page 3, line 16, please state the reason for the given error of 50%.

The MEIC inventory does not have an associated error estimate included and we were forced to assume one. In our new work, where the bottom-up and the top-down inventories are inter-compared in detail, we have performed sensitivity studies on the methodology by altering this value from a small estimate of 10% to a large estimate of 90% and will present the effect this has on the final updated emission inventory.

4. Page 3, line 27, please clarify the reason why the emissions in “great Beijing areas” is best represented.

Line added in the text.

5. Figure 1. It is not easy to distinguish the differences between graphs using the current legend.

We have altered the colour bars accordingly.

6. Page 8, line 3, please clarify the sources of the uncertainty of the CHIMERE SO<sub>2</sub> columns.

In the work of Beekmann and Derognat, 2003<sup>5</sup>, and subsequently in Deguillaume, et al., 2007<sup>6</sup>, a Bayesian Monte Carlo analysis was applied to the CHIMERE model over Paris in order to estimate the overall uncertainty with respect to the following CHIMERE model input parameters: anthropogenic and biogenic emissions, meteorological parameters such as wind speed and mixing layer height, actinic fluxes, quantum yields, and chemical rate coefficients. However, they only report assessments for tropospheric ozone, and then on secondary NO<sub>x</sub> and VOC formation, and not on SO<sub>2</sub>. CHIMERE runs were also used to assess SCIAMACHY observations [Blond et al., 2007<sup>7</sup>]

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<sup>5</sup> Beekmann, M., and C. Derognat (2003), Monte Carlo uncertainty analysis of a regional-scale transport chemistry model constrained by measurements from the Atmospheric Pollution Over the Paris Area (ESQUIF) campaign, *J. Geophys. Res.*, doi:10.1029/2003JD003391.

<sup>6</sup> Deguillaume, L., M. Beekmann, and L. Menut (2007), Bayesian Monte Carlo analysis applied to regional-scale inverse emission modeling for reactive trace gases, *J. Geophys. Res.*, 112, D02307, doi:10.1029/2006JD007518.

<sup>7</sup> Blond, N., K. F. Boersma, H. J. Eskes, R. J. van der A, M. Van Roozendaal, I. De Smedt, G. Bergametti, and R. Vautard (2007), Intercomparison of SCIAMACHY nitrogen dioxide observations, in situ measurements and air quality modeling results over Western Europe, *J. Geophys. Res.*, 112,

which includes error estimates but again for NO<sub>2</sub> only.

Within the framework of the EU FP7 MarcoPolo project, <http://www.marcopolo-panda.eu/>, an ensemble of modelled SO<sub>2</sub> estimates were inter-compared with in-situ observations and Figure 1 shows the relative percentage error of each model. During the OMI/Aura overpass time, CHIMERE has about 20-40% uncertainty SO<sub>2</sub> on surface concentration.

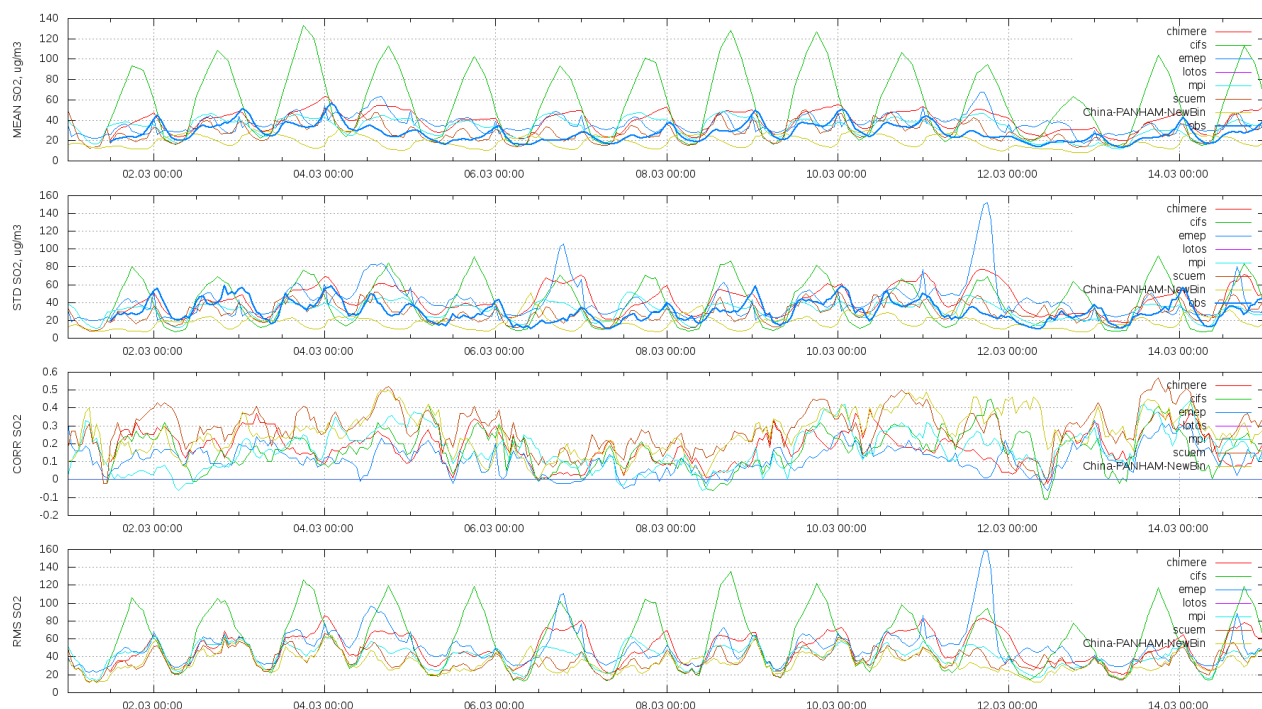


Figure 1. Inter-comparison of SO<sub>2</sub> estimates by different model runs [in different colours] to the CHIMERE estimate [red line]. From top to bottom: mean SO<sub>2</sub>, STD SO<sub>2</sub>, CORR SO<sub>2</sub> and RMS SO<sub>2</sub>. **Unpublished results.**

7. Page 9, line 21, it is not accurate to say “the OMI observations are point daily measurements”. The OMI observation cannot be treated as a “point”.

You are of course correct, line re-phrased.

8. Page 9, line 29. How many levels of CHMIERE output are used in this study? It says 8 here, but 7 before.

Apologies, small typo error mixing up the words layers and levels. The entire text was checked and amended accordingly.

9. Page 9, line 30. What is the “OMI 58 AK levels”?

Phrase added.

10. Page 17, line 19. What is the definition of “SO<sub>2</sub> emission fields”?

Wording rephrased throughout the text. We simply meant that we are producing an actual spatial domain, in lat/lon, of emissions and not total SO<sub>2</sub> emitted masses over specific source locations.