Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2018-661-AC1, 2018

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatial and temporal changes of SO_2 regimes over China in recent decade and the driving mechanism" *by* Ting Wang et al.

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Received and published: 24 October 2018

We are grateful to reviewer #1 for the valuable and constructive comments that are helpful to improve the manuscript. We have tried to follow all of the suggestions, and make changes accordingly in the main text with red color.

This paper uses OMI SO2 retrievals to study the effects of emissions and meteorology on SO2 loading over eastern China during 2005-2016. Monthly OMI SO2 from BIRA DOAS retrievals are compared with estimated SO2 emissions from the China

Statistical Yearbook. The authors show that OMI-observed SO2 has decreased significantly over eastern China, particularly for areas with the strongest emissions. They use an EOF analysis to demonstrate that the change is not monotonic and has four phases, with SO2 increasing during 2005-2007, decreasing sharply during 2007-2008 and 2014-2016, and only slightly increasing or decreasing during 2008-2013. They also show that the emissions and OMI SO2 are highly correlated over northern part of the country, but less so for southern China. They propose that abnormally dry and stagnant conditions over southern China during 2008-2013 may have caused a slight increase in SO2 loading, despite continued reduction in emissions. While several studies have examined the recent changes in SO2 pollution over China using satellite data, this study attempts to provide a somewhat different perspective. The conclusion that meteorology may play a fairly prominent role in the inter-annual changes in SO2 over southern China is interesting. The paper is well-organized and figures are mostly clear. However, I am not completely convinced that the emission data used can fully support the conclusions drawn in the study. I'd recommend that major changes be made before the paper can be accepted for publication in Atmos. Chem. Phys.

Reply: First of all, we would like to again acknowledge reviewer for the positive comments on our paper. We agree that it is essential to assess the reliability of the emission data used in the study. We have addressed this issue through (i) verification of the sulphur emission statistics released in the official yearbook (OYB), and (ii) repetition of the analyses using another independent inventory.

To verify the OYB inventory and corroborate findings, the Multi-resolution Emission Inventory for China (MEIC) developed by Tsinghua University is adopted. In addition, two other estimates of national annual totals (REASv2 and Zhao) are also used. The references about MEIC, REASv2 and Zhao are listed in the main text.

Overall, the results based on MEIC confirm and reinforce the conclusions obtained

by OYB. Firstly, the comparison between the four databases show that, despite the spread in their magnitude, the reported temporal variations are characterized by a similar behavior. We conclude that this justifies the use of OYB as a main source of information. Please see details in Section 2.2 and Figure 1. Secondly, the main conclusions drawn from OYB are not modified when the MEIC dataset is used. This can be judged from the pairwise comparisons listed below and the associated interpretations in the main text.

There certainly exists non-trivial uncertainties related to the currently available emission inventories, especially when considering small spatial or temporal scales or when specific sectors are targeted. Accordingly, we have added a comment in the new Section 6 to highlight current issues and challenges that need to be addressed in the future.

Table pairwise comparison figures between OYB and MEIC

OYB	Figure 2c	Figure 9a	Figure 10	Figure 11
MEIC	Figure 2d	Figure 9b	Figure S3	Figure S4

Specific comments: The authors indicate that the emission data used in this study have very strong seasonal changes in SO2 emissions from China (almost half of emissions in winter, and only 10% in summer). But this is quite different from many previously published emission inventories which generally suggest a much smaller seasonal change (such as HTAP). Also according to a number of previous studies, the residential sector is in general estimated to contribute roughly 10% of all SO2 emissions. This is quite different from what is shown in Figure 9 of this study. The authors may consider using

a different, more widely recognized emission inventory for their analysis and check if their conclusion still stands.

Reply: In the sentence "nearly half of the annual totals is released in winter when intensive heating takes place", the wordings "nearly half" and "released" are not appropriate leading to some confusion. In fact, Figure 3 actually depicts the seasonal variation of SO2 VCDs rather than SO2 emissions. In the revised version of the manuscript, we have reformulated the statement as follows: about 35% of the annual totals is taken up by winter, while SO2 in summer only accounts for 15%; the remaining 50 percent is almost equally divided in between spring and autumn. Please see Lines 228-231.

To explain the pronounced seasonal cycles in SO2 concentration, an additional figure (Figure 4) has been introduced which correlates the annual cycle of SO2 VCDs with sulphur emission, precipitable water and temperature at the four hotspots. Although intensive heating during winter in North China raises sulphur release, the variability of the SO2 emissions alone is not sufficient to drive the pronounced seasonality of SO2. The remaining variation is associated to seasonal changes in the meteorological conditions. The observed seasonality of atmospheric SO2 loadings is therefore resulting from variations of both emissions and meteorology. Further details are given in Section 3.2.

In Figure 9 (Figure 11 in the revised version), the Y-axis in a positive direction does not start at zero. We do this because our aim is relative change rather the absolute magnitude. If the origin of Y-Axis is set to zero, the blue bar denoting industrial emission is too tall to recognize the delta change.

It also appears that the emission data used here are on a provincial level (and not

gridded) and the authors calculate the emission strength based on the area of each province. Can the authors confirm that? If so, how do the authors calculate total emissions (for example those in Figure 8) for a domain that partially covers several different provinces? Also note that the emissions and SO2 loading can be quite inhomogeneous even within the same province.

Reply: We confirm that emission data used here are on a provincial level. Meanwhile, the terms "emission" or "emission amount" always refers to "emission strength", defined as emitted SO2 per unit area. To make it clear, we emphasize this in Lines 206-209.

Regional averaged quantities for North or South China are estimated as a weighted average by assigning the district area as a weight. Such explanation is added in Lines 334-336. In fact, the North China and South China are delimited by tracing provinces' boundaries, so that the North China and South China domains completely cover several different provinces. The demarcation line between the two portions are added in Figure 9.

We understand that emission and SO2 loading can be quite inhomogeneous within a same region, but this is not considered in this study due to the limitation that only continuous emission data on provinces are gathered at hand. Meanwhile, to match emission data, the gridded SO2 VCD is aggregated to provincial level. Consequently, the analysis given in Section 4 is constrained to provincial or multi-provincial levels. In Section 6, we particularly point out that future studies should use both gridded SO2 VCDs and gridded SO2 emission inventories. Please see Lines 458-462.

It is not clear how the "north" and "south" are defined in this study. One would assume that Cheng-Yu, PRD, and YRD are all part of the "South". But the SO2 time series

in Figure 6 indicates that they have different trends during 2008-2013. How would the authors explain these different trends when Figure 10 appears to show generally similar meteorological conditions for the three regions?

Reply: Thanks for your suggestion. We added a demarcation line in Figure 9 that separates South China and North China.

We also realize that Figure 6 may not be clear enough to convey the major idea. This is because the monthly anomalies mask the primary signal, and trend is also not a suitable indicator for rebound phenomenon. Therefore we designed a new figure (Figure 8) which presents the temporal evolution of annual SO2 from 2005 to 2016 in each province of eastern China, with the segment over 2009-2013 highlighted by red color. It confirms that the SO2 does not evolve in a monotonic way but shows a striking rebound during 2009 to 2013. This pattern is observed throughout most of the region, with only two exceptions: the Guizhou and Guangdong provinces that experienced a monotonic decrease since 2005. See Figure 8 and detailed explanation in Lines 305-313.

The "sudden downward shift of household emissions" in the south is quite surprising. Do the authors have an explanation for this? Or is this simply indicative of methodology change in the emission inventory?

Reply: We have double checked and can confirm that the OYB inventory shows a sudden downward shift of household emissions for South China. Unfortunately, the official year book doesn't provide any clue whether this shift results from a change in methodology or reflects real changes in emission inventory. As a result, we decided to avoid any speculation in the main text.

We also note that the MEIC inventory does not shown evidences for such an "abrupt-shift" behavior. Nevertheless, both inventories do report a reduction of household emissions in South China, irrespective of the exact manner. The phrase "a sudden downward shift of household emissions" has been removed from the abstract and the conclusions. Instead we refer to "the coordinated cuts of industrial and household emissions". Please see Lines 39-40 and 438.

Figure 4: there seem to be some negative SO2 values in the figure? Can the authors confirm that?

Reply: In this study, all negative SO2 values are eliminated prior to the research. Thus, there are no negative values in Figure 4 (Figure 6 in the revised version). The label bar includes 'less than 0' bin, because we expect it to be symmetric. However, no patch on the shaded plot indicates less-than-0 value.

In addition, the YRD and PRD regions are slightly enlarged in the revised manuscript, to mitigate possible uncertainties.

Figure 8: What is the unit for emissions? What does each data point represent in the scatter plot?

Reply: The unit for emission is ton/km2, and each data point corresponds to one year and one province. Such key notes are inserted into the figure caption. Please see Figure 10.

Figure 10: which area is the vertical profile in (d) for?

Reply: The area is 23-31°N, 105-122°E rectangle. In addition, we find that the result is not sensitive to the chosen area.

Writing: the authors should also make an attempt to improve the writing. Short, simple sentences in some cases may make the paper easier to follow.

Reply: Thank you very much for your suggestion. Further polishing has been applied in the revised version.

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