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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.

Ting Wang et al.

wangting@mail.iap.ac.cn

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We thank reviewer #2 for his suggestions to improve our manuscript. We have done our best to address each of them. The revisions in the manuscript are highlighted by red color.

General comments: In this paper, Ting Wang et al. analyzed the spatial distribution and temporal variation trends of SO2 VCD and emissions in different regions of China in the last decade based on the OMI observation and emission inventory. Further they

discussed effects of meteorological conditions on the SO2 variations based on the differences of emissions and SO2 VCDs in South China. In general the scientific topic is meaningful, and the perspective of understanding effects of meteorology on SO2 depositions and dispersions is novel. However I have two major concerns below:

Reply: We would like to again acknowledge the reviewer for his positive comments on our manuscript.

1) A credible emission inventory is quite a foundation of the study. However the authors do not give a peer-reviewed publication of the emission inventory in Section 2. The authors should cite some papers to introduce the methodology and validation of the inventory. Meanwhile it could be more convincing if the authors do the same analysis based on another available peer-reviewed emission inventory

Reply: Thanks for this valuable suggestion. The official estimate of SO2 emission inventory is published every year, and it is certain that peer review processes are undertaken before publication although these are not open to the public. The yearbook also briefly introduces the methodology to build the inventory, which has been added in Section 2.2. Unfortunately, no further detailed information is provided in the yearbook.

We understand the crucial concern regarding the reliability of the emission data used. To guarantee the robustness of this study, we have sought to address the concern in the revised version as you suggested. A similar comment was raised by the first referee and we refer to our corresponding reply.

2) The author did not consider any effects of regional transports in the discussion of the discrepancy of SO2 VCD and emissions. SO2 life time could be long and has a big variability. SO2 could be transported by on an order of 100 km, especially during night.

Reply: Thank you for this comment. The effect of regional transport is of importance, and climate-chemical coupled models are usually applied for its evaluation. Alternatively, we have used an Effective Emission Index (EEI) that accounts for the impact from both local and remote sources. Adopting results published by Zhang et al. (2015), we get: for North China, within-region SO2 emission contribute 68% followed by 19% from South China and 13% from other regions; for South China, within-region emissions provide 66%, while transport from North China and other regions amounts to 17% and 17% respectively. Based on these statistics and assuming that the EEI is linearly dependent on N and S and that external contributions remain fixed, the EEI index is formulated. For comparison purpose, we also define an Emission Index (EI) that considers single effect from within-in region emission. The detailed procedure to construct the indices are presented in Lines 339-365.

Note that we only consider the inter-regional transport between North China and South China. Because the spatial scale is large, we find that integrating the role of inter-regional transport does not alter the overall pattern and result. However, the best way to unveil fine scale details of transport is using climate-chemical coupled models. Due to the limited resources available for this study, this could unfortunately not be attempted. Future directions are highlighted in Section 6.

Specific Comments:
1) Line 116-117: how significant is the improvement of the new product on the study

of variation trends? Do the authors compare the variation trends based on the new product with those based on the previous product?

Reply: Compared to the BRD OMI NASA SO2 product, the BIRA retrievals proved to be better both in terms of noise level and accuracy. This product also includes a full characterization (errors, averaging kernels, etc.). The improved OMI PCA SO2 product of NASA show similar performance and long-term trends as the BIRA product. The BIRA SO2 product has also been validated in China using long-term MAX-DOAS data (Theys et al., 2015; Wang et al., 2017). As these comparisons have been done before this study, we added a short remark in Lines 125-130.

2) Line 120-121: What kind of background correction is applied? Can the correction cause artifacts of some weak signals of SO2 in some regions which are dominated by the natural sources as discussed in Line 151-155?

Reply: The correction we use here is based on a parameterization of the background values that are then subtracted from the measurements. The scheme first removes pixels with high SZA (>70°) and SCDs larger than 1.5 DU (measurements with presumably real SO2) and then calculates the offset correction by averaging the SO2 data on an ozone slant column grid. This is done independently for each across-track position and hemisphere, and the correction makes use of measurements averaged over a time period of two weeks around the measurement of interest. The details of background correction can be found in Theys et al. (2015).

Yes, the low level SO2 columns are subject to large uncertainties and the background correction is an important source of error. However, the regions with weak SO2 signals/background SO2 are not the subject of the present paper.

3) Fig. 3a: The author should explain the line around 40 N latitude with high values in winter

Reply: There is a belt of large positive values extending along 40°N in winter. However, it is a known artefact due retrieval limitations at large solar zenith angles. This does not incur any barrier to subsequent investigations, since our focus is eastern China that does not include this belt. Please see details in Lines 273-275.

4) Line 161 and Fig. 2a: snow could cover the surface in the western and northern part of China in the seasons, except summer. The snow covered surface could impact the retrievals of SO2 VCD. This could be the reason of the missing values of satellite SO2 VCDs in the two regions, especially in winter. Do the authors consider the point in the discussion?

Reply: Following your suggestion, we added Figure S2 and a paragraph to elaborate on this point. Figure S2 is designed to evaluate the availability of monthly SO2 data relative to the entire period. As mapped in Figure S2, there appears to be a substantial fraction of data gaps in western and northeastern China, especially in the winter half year. This can be attributed to snow cover surfaces and high solar zenith angles, which invalidates the measurability. However, the missing data issues in northeast and western China have virtually no impact on our study, because we mainly focus on the highly polluted eastern China.

Please see Figure S2 and relevant interpretations in Lines 135-145.

Meanwhile in the lines of 161-163, the authors attribute the higher SO2 amounts in summer than other seasons to the natural emissions. However the snow coverage effect could also play a role.

Reply: In the light of the considerable data gaps in western China as shown in Figure S2, it is impossible to draw firm conclusion. Therefore, we remove this statement in the revised version.

5) Line 159: The authors conclude that "nearly half of the annual totals is released in winter" because of the significant higher SO2 VCD in winter than in other seasons. However SO2 lifetime could be also longer in winter. The larger SO2 VCD values could be also related to longer lifetime of SO2 due to its easy accumulations in winter.

Reply: Thank you for this suggestion. The lifetime does take an important role in shaping the seasonality of SO2 VCD. To better explain the pronounced seasonal cycles in SO2 concentration, an additional figure (Figure 4) has been included to illustrate the annual cycle of SO2 VCDs and its relation to sulphur emission, precipitable water and temperature at the four hotspots. On the one hand, intensive heating during winter in North China raises sulphur release. However, emission alone is not adequate to explain the pronounced seasonality of SO2. Temperature and humidity are cold and dry in winter due to the influence of winter monsoon, which jointly weakens the rate of oxidation and wet deposition. Accordingly, it is expected that SO2 molecules will have a longer lifetime and will thus accumulate easier, as you suggested. The opposite is true for summer, when chemical reaction is active and wet removal is effective. Please see Figure 4 and relevant interpretations in Lines 233-243.

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