Interactive comment on "Multi-source SO₂ emissions retrievals and consistency of satellite and surface measurements with reported emissions" by Vitali Fioletov et al.

Anonymous Referee #2

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The study links SO2 emissions as well as surface measurements to column measurements from satellite by a simple dispersion model. While previous studies on this topic focus on individual point sources, this study uses a generalized model function which allows to derive emission estimates for a list of sources (even close to each other) at once. By establishing the link between emissions and columns, even "reconstructed" SO2 columns were generated for the time before actual satellite measurements are available. The paper is well written. Results are impressive and convincing, and the method is innovative. It should be published in ACP after dealing with the following issues:

We would like to thank the reviewer for the evaluation and comments that helped us improve the manuscript.

General comments:

1. OMI SO2 Bias

The good results are only reached after removing a somehow mysterious "retrieval bias". When reading Page 7, Line 3, I was thinking about some constant, or weakly latitude dependent bias. But in fact, the bias has systematic spatial structure and considerable spatial gradients. The authors argue that the enhanced OMI signal at the East coast is not reflecting true SO2, and in particular the comparison to OMPS is convincing. However, the reasons for this OMI "bias" remain unclear. I don't find the given reasons (O3 interference, stray light) convincing at all.

I see the need for the high degree of Polynomials fitted to remove the unexplained spatial features. However, I would not call it a "bias" (which I would associate with something like a constant offset). In addition, the authors should - extend the description of the characteristics of the bias in the paper and point out the spatial pattern (US Eastcoast) in the main text,

- extend the discussion of possible reasons (in paper or supplement),

- be aware that the high degree of the fitted polynomial actually removes any unexpected signal (by adding degrees of freedom, anything can be fitted), thus the good fit results are not that surprising,

- discuss how far the bias removal might affect the emission estimate, in particular for the study on wind dependency (see next point).

Perhaps the importance of the polynomial-based bias is somewhat overstated in the paper. While we believe the bias is real, its magnitude is typically within +/-0.1 DU and its impact on emission estimates is rather small (unless we are dealing with area emissions from large areas). We have added about a page of text and three figures to the Supplement that illustrate how the degree of the polynomials affects the bias, the fitting results, and the emissions themselves.

One of the factors that contributes to the bias is surface reflectivity and we have added some discussion about it to the Supplement.

Degrees of freedom is not a big issue here since we are dealing with hundreds of thousands of satellite pixels.

We prefer to use the term "bias" as we used it in our previous work related to point sources where it was indeed a constant offset. To address the reviewer's concern, we have highlighted in a few places that we are dealing with a local bias that that changes relatively slowly with latitude and longitude (compared to signal from emission sources).

2. Dependency on wind speed

The application of the model fit for different wind speed bins is quite interesting. However, the authors do not provide the resulting emission estimates. The authors claim that VCDs are not good proxies for emissions as they depend on wind speed (Page 10, Line 17). But from Figure 4, I have the impression that not only the local, but also the integrated VCD depends on wind speed, which should not be the case according to the model function. Is this the case? Please provide the emission estimates for the 3 wind speed bins. If they are different, discuss possible reasons. Could the difference be related to the fitted Polynomial? Please provide maps of the fitted bias for each wind speed bin in the supplement.

There is some confusion here. We have not estimated the emissions for three wind speed bins. The emission estimates were done using the entire data set. The purpose of Figure 4 is to show that the signal from the same sources would appear differently in OMI data for different wind speeds. OMI SO₂ values over the same sources would be higher if the wind speed is low and lower if the wind speed is high. Furthermore, the right column of Figure 4 is not related to actual OMI measurements. It is a reconstruction of VCD distribution based on emission inventories, the plume model, and the actual wind data. We show it to illustrate that we are able to capture the dependence of the OMI SO2 "signal" on the wind speed.

We modified the text to make this clear. As we believe that the bias is related to the retrieval procedure, we assumed that it did not depend on the wind speed. So, the bias is the same for all three wind speed bins.

Estimates for different wind speed bins were discussed in the Supplement (section 5) to our previous paper (Fioletov et al., GRL, 2015)

Detailed comments:

Page 4 Line 29: "...do not vary much" - have you checked this? How would the reconstructed VCDs look like if e.g. the wind data from 2006 would be used instead?

We added such a figure to the Supplement.

Page 6 Line 17: "prescribed SO2 decay time" - please provide details here and give the numbers used for tau for the different seasons.

Details are given in the Appendix. We have changed the text to "The detailed formulas *and prescribed seasonal decay times* are given in the Appendix" to highlight that.

Page 7 Line 3: "change slowly": this would apply for a polynomial of degree 2, but not for degree 6!

We have added the clarification "change slowly (compared to signal from emission sources)". Yes, degree 6 is high, but the analyzed area is huge.

Page 7 Line 11: "artifact from the retrieval": please extend the discussion of the artifact and possible reasons (here or in the supplement).

We added some discussion to the Supplement

Page 8: I understand the reason for the structure of Figures 1&2, but the order of the text is a bit confusing: it first refers to Fig. 2b, then Fig. 1, then Fig. 2a, and in the following to particular columns of Fig. 1. Please try to make the text plus references to Figures more smooth. It would also help a lot to have the columns of Fig. 1 labelled (a to e or I to V) to avoid references like 'Figure 1 (the "VCD from emissions" column)'.

Corrected as suggested.

Page 8 Line 26: "Figure 1" -> Figure 1 (e)" (or 1 V).

Corrected

Page 9 Line 32: For the correlation of reconstructed VCDs with OMI (bias removed and emission-related signal extracted), the same model is assumed for both datasets, and any non-matching measurement is removed from the OMI data (by bias removal). Thus, the good correlation is not that surprising.

Page 11 Line 15: "reached *until* 2014".

Corrected

Page 12 Line 26 end of sentence: dot missing.

Corrected

Page 13 Line 6: 0.91 is reached after bias removal, as stated in brackets, but these are NOT the "actual OMI measurements" any more!

Changed to "the reconstructed VCDs and the OMI-based values"

Page 13 Line 18: I agree in general, but the requirements on spatial resolution and quality of emission inventories would be high, and sources from power stations, industry and traffic are often close to each other. The authors should add a statement that emission inventories with good spatial resolution would be required.

Corrected as suggested

Page 15 Equation A1: The division by wind actually converts the decay rate from time to space reference system. It would be helpful to indicate this by adding a subscript "t" to lambda, and replace "lambda_1" by "lambda_x"

This may create some confusion: we use "y" as the up/downwind coordinate, so it is more logical to use lambda_y. We also prefer to use the same symbols as in the previous publications.

Page 16 Line 4: For y>0, sigma_1 is just sigma, so how far does "sigma_1 increased with the distance from the source"?

In chosen coordinate system, "y" is negative downwind. We have added a reminder of this point in the text.

Page 16 Line 12: "calculates" -> "calculated"

Corrected

Figures 1 and 5: - add lat/lon coordinates.

- add column numbers (a to e or I to V)

We added the column numbers (I to V) and specified the lat/lon coordinates of the area in the caption

Figure 2:

- shift a, b, c to top left corner of panel or even above the panel

- "d" is missing

Corrected

Figure 8: Why is 1980-1982 included when there have been no measurements?

Corrected. The 1980-1982 panel has been removed.

Figure 9: place labels a-d above panels.

Corrected as suggested