Review of "SO2 emissions and lifetimes derived from TROPOMI observations over India using a flux-divergence method" by Chen et al.

This paper uses TROPOMI SO2 VCDs (the COBRA data product) with a variant of the flux-divergence method as pioneered by Beirle et al. (2019) to derive SO2 emissions on a 0.1 x 0.1 deg grid over India. The authors use a calculated effective lifetime assuming first order loss due to chemistry and physical removal processes that varies with season and location, with a representative value of roughly 15 hours. Their derived emissions were found to be roughly a factor of two smaller than existing bottom-up inventories and another satellite-derived SO2 emissions database.

This is a well written paper, and easy to follow. But it seems odd to me that conclusion (anthropogenic SO2 emissions in India are only half what we thought) is really just mentioned in passing. Also, there is no validation of the results… comparisons are made with databases that differ by this factor of two, but that is it. The modifications to the flux-divergence methodology as previously employed are such that they require validation in their own right. Furthermore, when one obtains results that contradict previous studies it behooves the authors to provide some rationale as to why this is, and what could be behind the bias.

While I am not convinced that the flux-divergence method is the best approach for SO2 emissions as its strength is for extended area sources (e.g., urban NOx emissions), and SO2 comes from a collection of point sources (albeit occasionally in fairly close proximity), it is worthwhile to explore its effectiveness. Retrieving emissions on a grid smears out the emissions which usually can be geolocated to within a couple km, and makes getting the total for a facility sometimes challenging since one must figure out which grid boxes need to be summed over.

Aside from the larger issue of validating this rather extra-ordinary result, my main issue, and one I see as sufficiently serious as to warrant a major rethink of this work, lay in how the lifetime is derived. The authors attempt to calculate the lifetime considering simple physical (dry deposition) and first order chemical (via SO2 + OH) loss, and these individual lifetime are combined. As an aside, why is only dry deposition included? Adding it would not impact my argument below, but it seems like wet deposition should be considered to be consistent with the approach chosen by the authors. Dry depsotion lifetime were quite long, >60 hours, and thus do not have a huge impact on the combined lifetime. Chemical lifetime were calculated using an OH field ($\tau = 1/[k\,[OH]]$) from the ECMWF CAMS forecast model, and are roughly 20 hours, varying in space and time. The combined lifetime is something like 15 hours.

My concerns regarding this lifetime issue are laid out here:

1. Using the OH field from ECMWF CAMS is not appropriate as the spatial resolution is 0.4 x 0.4 degrees (~40 km), and thus represent an average OH value, with half or more of the averaging area (on average; including the upwind portion) representing background values and chemistry. What is relevant is OH in the plume where the bulk of the SO2 is, and OH at the plume core, plume edge, and background will all be much different. Using different models with comparable resolution to look at differences does not help in this regard. If one wants to use model OH then something like a plume-following chemical box model is the most appropriate choice.

2. In previous applications of the flux-divergence method, the lifetime was derived using the data itself (including and especially in the original formulation by Beirle et al. (2019)… and this seems

like the obvious method to employ here. Complications such as non-linear chemistry will then be accounted for. There is sufficient signal to tease out some spatial and seasonal differences from the TROPOMI data itself. At the very least the authors should have validated their calculated lifetimes using the data itself!

3. A simple analysis of the plumes themselves in the satellite data suggests that the effective lifetime is shorter than 15 hours. The authors do not ever show the actual TROPOMI SO2 data in their paper (there is one panel in the supplement), which seems strange considering it is the basis for the emissions calculations. Shown below in Figure 1 is an OMI SO2 VCD averaged over 2014-2017 (this is all I had handy; TROPOMI will look similar, but hot spots will appear sharper due to its higher spatial resolution). The dots are the larger SO2 sources. These are unpublished, diagnostic figures from the same EMG (exponentially modified Gaussian) method as published in Fioletov et al. (2016, 2018), McLinden et al. (2016) and later papers by that group. The left panel is the mean VCD (minus a slowly varying background bias, an artefact of the method, as discussed in the references above). The right is the reconstruction of the satellite data assuming a 6 hour effective lifetime.

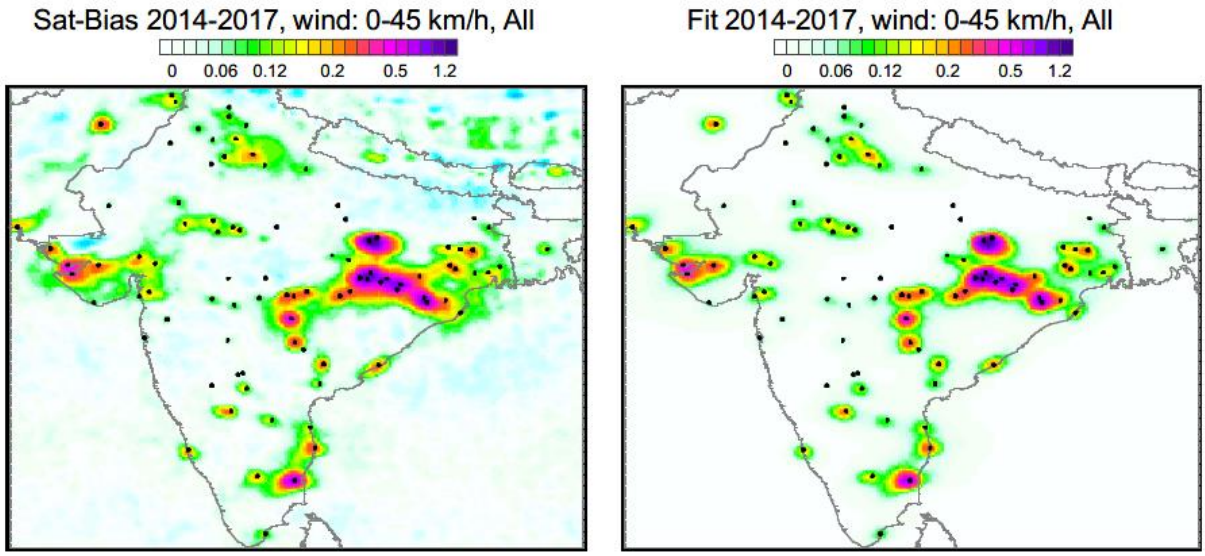

Figure 1: SO2 VCDs over India. Left: Observed OMI VCDs, with a large scale bias removed. Right: Reconstructed (and accounting for the resolution of the satellite) using the Fioletov et al. method assuming a six hour lifetime.

The maps above, with the hotspots so closely linked to their emissions location, suggest a 6 hour lifetime is about right. Increasing this to 15 hours would smear the SO2 out to a point where it would look like a smoothed version of the observations.

4. For EMG methods similar to that employed in my point 3, de Foy et al. (2015) argued that there is a dispersion component to the effective lifetime, and it reduces the overall lifetime relative to

the chemical+physical lifetime. In my understanding of the flux divergence method, diffusion is not accounted for and so it seems reasonable that the effective lifetime appropriate for this method may also have a dispersive component. Further, in the case of an isolated point source, as it the case for many here, the two methods are virtually the same with the main difference being that the EMG method of Fioletov et al. assumes a plume shape, and the flux divergence method does not constrain the plume, but derives emissions on a grid. If this case, the physical interpretation and value of the effective lifetime should be very similar, and I would argue that my Figure 1 above is the most compelling argument for a shorter lifetime. See Vindhyachal in particular (the most northerly isolated hotspot). If the physical interpretation of the lifetime is not the same, then one would need to look at how the different methods perform when emissions are well know, such as from CEMS or other direct measurements. The EMG method of Fioletov et al has been extensively validated against such measurements in the US, Europe, and for volcanic sources as well.

5. Application to India is more difficult since there is little reference data to validate against. In such cases it is useful to apply the method to other locations where such data is available. For example, in the US, there is CEMS data to compare against. The authors even mention it can applied anywhere. If one wants their results to be received with confidence, this is an essential step.

Additional comments:

Presumably there is nothing that unusual about Indian OH levels (the lifetime as derived here, to first order only depends on k and [OH]), and thus had the authors preformed a global analysis they would have found similar results (ie, ~a factor of two smaller emissions as compared to current, best emissions values) everywhere. Even just for India this has large implications, but for the globe this would lead us to rethink the SO2 budget.

Line 382: "But we see a noticeable smoothing effect and an overall positive bias on emissions estimated with a fixed 6-hour lifetime compared to the emissions estimated with a local, variable lifetime, especially around the source location"

But given the colour-scale, and comparing Figure S8 with Figure 5, this is exactly what one would expect when you ~double the emissions… there should be more red because all values are larger for the 6 hour lifetime, and thus it would appear to be smoother. The only way this line of reasoning could be reliably used is to compare an isolated source, and then normalize them their peak value. Eyeballing Vindhyachal, the large isolated point source in my Figure 2, below, the effect, even with the 2x emissions, appears marginal. Bigger picture, this type of analysis is indirect at best… again, why not use the actual data to deduce the lifetime?

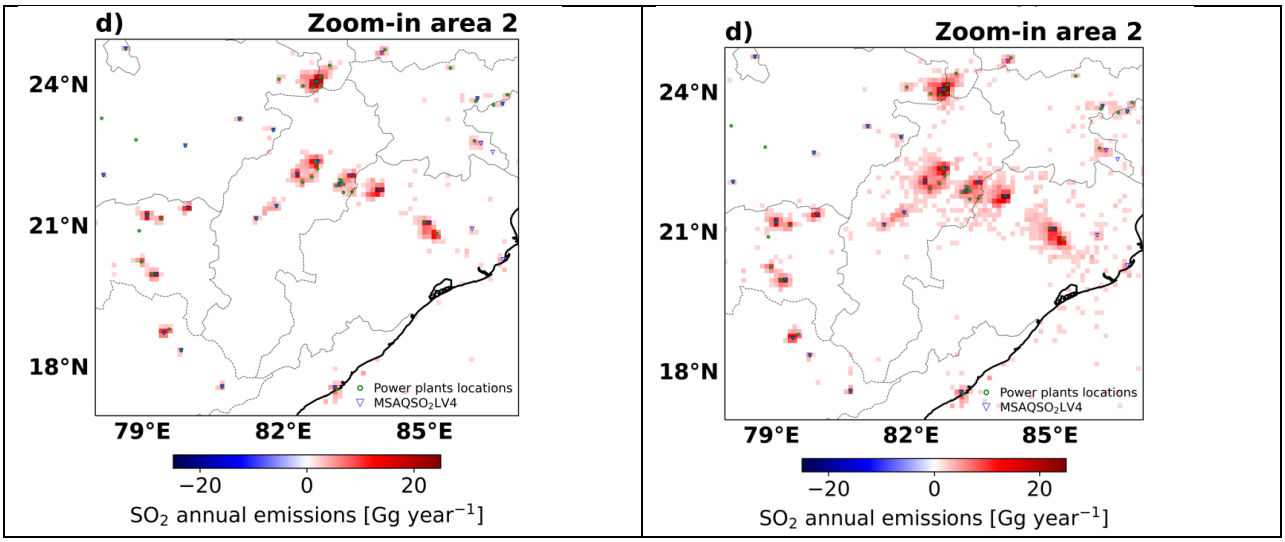

Figure 2: left Figure 5d (longer, variable lifetime); right: Figure S8d, 6 hour lifetime.

In closing, I would like to say that this study has merit. However, the approach used to derive the lifetime is quite concerning, and it has very large implications for the SO2 budget. Therefore as it currently stands I feel this paper should be rejected until the points above are addressed in a comprehensive manner.

References:

De Foy (2015) https://www.sciencedirect.com/science/article/pii/S1352231015301291

Fioletov et al (2016) www.atmos-chem-phys.net/16/11497/2016/

Fioletov et al (2018) https://doi.org/10.5194/acp-17-12597-2017

McLinden et al. (2016) https://www.nature.com/articles/ngeo2724