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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A global catalogue of large SO_2 sources and emissions derived from the Ozone Monitoring Instrument" *by* Vitali E. Fioletov et al.

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A uniform census of large global SO2 sources is a significant advancement in monitoring the inputs to the atmosphere for air quality and climate change assessment. This well written paper makes use of data from the second generation of space-borne UV mapping spectrometers to detect and quantify global ground level sources of sulfur dioxide. While large volcanic eruption SO2 cloud masses have been measured for over 35 years starting with discrete wavelength TOMS instruments, detection of industrial sources in the boundary layer need the full spectrum of backscattered sunlight available through new technology in the OMI and GOME instruments, among others. This paper also takes advantage of the OMI Principle Component SO2 retrieval algorithm

for PBL sources with its doubled sensitivity and reduced regional biases. New source detection and analysis tools from Fioletov et al., 2015 are then used in calculation of source strengths. The wind rotation and upwind - downwind difference methods are clever ways to locate and measure sources for inclusion in the census.

The scope of the paper is broad and a challenge to review in depth. My review focuses on technical details rather than environmental and industrial issues. However the significance of this dataset for identifying "missing" sources is great and should lead to more accountability in the bottom-up emissions estimates. For example, it's easy to see in the Appendix that the Ilo smelter in Peru reduced its high emissions down to zero by 2009 and kept it there through 2013, but emissions returned again in 2014 almost to 2007 levels. The failures of bottom up emission inventories, such as in Mexico for whatever reason, will become obvious.

The error analysis shown in Table 1 seems to be conservative but the large size of some error sources like the 35% uncertainty from lifetime and width assumptions suggests that these errors could be reduced through refinement of SO2 decay rates at individual sites.

This work can resolve questions about the relative contributions of volcanoes and anthropogenic sources. A global inventory of SO2 from degassing volcanoes is significant contribution.

This research study is another success for the NASA MEaSUREs program. I recommend publication after relatively minor revisions.

General Comments

This paper depends on information in Fioletov et al., 2015 to be understood. For example, "Fitting Uncertainty" is not defined except as a standard deviation of the estimated emissions. It would be good to point the reader to the 2015 paper in the Introduction. A brief summary of the techniques presented in that paper would resolve questions that

arise in Section 3 and the Appendix.

P 5, Section 3.1 A discussion of sample sizes is missing. How many samples are needed to detect a source? How many samples are collected to compute the annual emission value? What is the time resolution in trends?

P 7. It is a concern that an average lifetime of 6 hours is used for all sources although it must depend on variable factors, like humidity. Is it practical to fine-tune the lifetime if meteorological data are available?

Figure 12 shows a time series of total annual SO2 emissions for the four primary source types while Figure13 shows the total SO2 emissions for separate countries or regions and the ratio to reported emissions. Both figures show the results for three different AMF choices. The main points of the figures are lost when AMF information is included. It has already been made clear that the fixed value of 0.36 in the production dataset does not apply globally or to all individual stations. Unless the AMF choice influences the time series why include it here redundantly? If the 3rd case is the best why not just show that and perhaps condense the effects of AMF choice into a few sentences or a summary chart. Then the real info about the trends and ratio of reported to satellite annual emissions will become the focus of the figures.

The catalogue in the Appendix contains a single value for the AMF for each site. Is this the average of the AMF's for each of the observations at that site or the result of prescribed conditions? If so how are the effects of changing conditions on the AMF accounted for?

The range of AMF's in the source dataset is unexpectedly large (< 0.3 - > 1.8). I can understand the value of 1.87 for Erebus volcano due to permanent snow/ice cover and altitude. But the substantial variability implies that the PCA dataset is not representative of global conditions. However, it is probably unreasonable to compute global values of AMF for each OMI pixel. It seems that the current approach of computing AMF's just the source locations is probably the best.

One piece of information missed in the Appendix is the sample size for each source. How many observations go into each annual emission data point? You might add a new page in the workbook to include this information.

Specific Comments:

P 6, L 18ff. The choice of the letter, a, to represent SO2 mass is awkward and leads to confusion with the article, a (P 7, L 6). I realize that a was used previously for this variable but I would consider changing to a less ambiguous symbol.

P 7, L 6: The phrase "depends on a linearly." should be "depends linearly on a." This would not be as clumsy if "a" were replaced by "m" or some other letter or symbol.

P 10, L 11. If the use of SO2 cross sections for a stratospheric temperature (203K) in DOAS boundary layer retrievals creates a 19 % difference from PCA (283K) retrieved values how can the emission estimates be the same within 5%. Please elaborate on how the emission algorithm could decrease the bias.

P 11. The term "emission-factor" is not defined. Please correct this.

P 16, Sec. 5.4 Volcanoes: In the context of this paper on the detection and evaluation of SO2 sources in the PBL I think the limitations of measuring volcanic emissions is overstated. McCormick et al and the other cited authors have rightfully addressed the possible sources of error in volcanic plume retrievals in their papers. However, the majority of these plumes are released into the free troposphere above the PBL where the AMF is readily computed. In addition any ash falls out rapidly near the volcano. The "numerous issues such as limited instrument sensitivity to volcanic plumes at low altitudes" seem simple when compared with the large uncertainties in the measurement of industrial emissions in the PBL.

Snow covered volcanic cones might affect a single OMI pixel and the plume drifts free of the volcano peak over the lower altitude terrain with its seasonal changes of reflectivity. Thus I don't believe "Albedo effects from snow-covered volcanic cones" is a significant

problem (except for Erebus, where constant snow cover seems to be accounted for in the Catalogue). For passively degassing volcanoes, the vent height is generally known and the SO2 plume is usually released above the PBL, so detection is better than industrial sources. The NASA OMI BRD dataset provides solutions for different altitude regimes to allow the user to choose the appropriate altitude for the plume. The PCA dataset as implemented by GSFC focused on air quality and PBL emissions so the data need to be corrected for other altitude regimes, as the authors have done.

P 19, L 30. The phrase 'sand covered areas with high albedos" seems misleading. The UV albedo of sand ($\sim$10%) is not large relative to the visible wavelengths although it is larger than for vegetation.

Figure 2. The term "Nobs" is not in my vocabulary. Can you define it in the text or replace it with a common term?

Technical corrections:

P 4,L 7. The exponent in 10ˆ26 is mislocated.

P 6, L 4 and P 7, L 18. Typo; "centered" instead of "cantered".

P 13, L 5. Use "ground" instead of "grounded".

P 31, Table 1. Note for Lifetime and Width: delete "Used" in "Used prescribed. . ..."

P 35, Figure 3. Subplot identifier, d, has escaped to the first line of the caption. All the identifiers could be made more prominent.

Throughout the text, in-line references are fully enclosed in parentheses. e.g. on p 11, 'the value reported by (Kaldellis et al., 2004)'. In other journals this would be 'the value reported by Kaldellis et al.,(2004)'.

Figure 13 caption: line 5. (b and e), not (b and d)

References:

The Beirle et al paper is referenced twice; both the discussion and the published paper are included. I assume the latter replaces the discussion paper and the former should be deleted.

Attachment:

ReadMe page;

Line 4 Col B: "Longitude" misspelled

Line 6, Col B: "Source" misspelled.

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