Atmos. Chem. Phys. Discuss.,
doi:10.5194/acp-2016-1142-RC2, 2017

Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Particulate sulfur in the upper troposphere and lowermost stratosphere – sources and climate forcing" *by* Bengt G. Martinsson et al.

Anonymous Referee #2

Received and published: 22 March 2017

This manuscript reports on measurements of aerosol sulfur in aerosol samples collected from the IAGOS-CARIBIC platform over a 16 year period. Analysis focuses on a new regression technique that the authors suggest can be used to infer both the gradient of sulfur concentration and the integrated burden in the lowermost stratosphere (LMS), starting at the dynamical tropopause and extending to 3 km above it. It is also suggested that this analysis provides an estimate of the relative contribution of stratospheric sulfate mixed downward and tropospheric sources on the sulfur concentration in the upper troposphere (UT), and how these contributions vary seasonally.

My biggest problem with the paper is that the authors do not show and discuss enough raw data to demonstrate that the regression approach is reasonable. Figure 1 does

give the reader a useful impression of both the range and seasonality of sulfur observed in the UT, and supports the authors impression that there is little correlation with distance below the tropopause. However, the LMS concentration data are never shown so the reader has no idea if fitting linear regressions is a remotely logical approach. This is compounded by the fact that the analysis apparently required multiple steps which are not well explained in section 2.3.

I realize that this group has written a number of previous papers on this data set, and perhaps some of these have already presented spatial and seasonal distributions of sulfur in the LMS in ways that set the stage for this new analysis. However, I did not, and readers in general should not have to, read these earlier works to understand this one.

I could provide a fairly long list of specific sentences and paragraphs that I found to be confusing or misleading. However, I just noticed that reviewer 1 has suggested major revision, starting with fundamentally changing the approach to analysis, which will clearly require rewriting most of the text. Therefore it seems that specific editorial suggestions to improve clarity are premature

I agree with the concerns reviewer 1 raised regarding the use of concentrations rather than mixing ratios, and relying on distance from the tropopause as the independent indicator of degree of stratospheric character captured by a given sample. (I also note that simply defining this distance for sample intervals approaching 2 hours in length would often seem ambiguous, even without double tropopauses or crossings of the tropopause.). I cannot comment on the suggestion to extend the analysis used by Friberg et al., 2014, since, as noted above, I am not familiar with this paper.

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1142, 2017.