

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

### **Anonymous Referee #1**

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This manuscript analyzes measurements of particulate sulfur taken from commercial aircraft altitudes in the IAGOS and CARIBIC programs. It attempts to derive the fraction of upper tropospheric sulfate that is from the stratosphere. This is an excellent data set but I find the analysis inadequate for several reasons. A previous paper by many of the same authors on a subset of these data (Friberg et al., 2014 but it only uses data through 2008) is a much better analysis. I would recommend resubmission with completely new analysis based on extending the Friberg et al. techniques to include the newer data and more emphasis than the Friberg paper on non-volcanic periods.

1) The manuscript (line 135) states that the analysis is based on concentrations per unit volume rather than mixing ratio. This is a mistake; straightforward analysis of fits

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of concentration versus a parameter like distance from the tropopause requires the use of mixing ratio. For an idealized example, suppose the aircraft ascends 1 km as it goes through a perfectly uniform air mass. That ascent would change both the x-coordinate (distance from the tropopause) and the y-coordinate (concentration not corrected to mixing ratio). This coupling of the independent and dependent variables makes it impossible to interpret the slopes in a simple fashion. The stated reason for using a volume concentration (integration over an altitude range) can always be done later in the analysis.

2) The distance from a tropopause defined by potential vorticity (PV) is not a very good choice for the independent variable (Figures 1 and 2 and subsequent analysis). First, the PV values come from a meteorological analysis with substantial uncertainty. A colleague I spoke to estimated +/- 500 meters. There are also ambiguities with multiple tropopauses. Consider what Figure 2a would look like with uncertainties of +/- 500 m in the horizontal for most points, and more for a few points in the neighborhood of multiple tropopauses. Note that using an independent variable with significant uncertainty not only introduces noise into line fits but also biases the results to smaller slopes and, for positive data, larger intercepts. (This is worse than uncertainty in the dependent variable, which introduces noise but not bias.) Second, there is no reason why the gradient in PV has to be uniform with distance above the tropopause, so deeply stratospheric air could be close to or far above the tropopause. No tracer is perfect, but ozone, as used in Friberg et al., would be a far better choice than distance from a PV tropopause.

3) The analysis of the stratospheric influence fraction is very convoluted with no propagation of uncertainty shown. There are three successive line fits to data, as shown in Figure 2a to 2c. After reading through the manuscript several times, and having worked extensively with similar data, I still do not understand how the measurement uncertainties and atmospheric noise propagate into the results.

4) Putting aside the choice of independent and dependent variables and the propagation of uncertainty, there is a conceptual problem with defining the stratospheric influ-

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ence from a corrected intercept derived from the line fits, as is done in this manuscript. Such an analysis of the slope and intercept of two variables in the lowermost mid-latitude stratosphere generally assumes that both variables are conserved quantities controlled largely by transport and mixing (e.g. Plumb 1996 JGR tropical pipe paper). But sulfate mass in the lowermost stratosphere is mostly controlled by sedimentation (Wilson et al., Steady state aerosol distributions..., ACP, 2008). In the presence of sedimentation, it is not obvious what the slope and intercept of a correlation plot mean. Indeed, it is clear from Figure 5 in Wilson et al. that a line fit over an altitude range that goes deep into the stratosphere could easily produce an intercept unrelated to the tropopause value.

Lesser concerns are: (a) Distance from the tropopause is strongly correlated with latitude, since commercial flights generally only get well above the tropopause at high latitudes. This makes it difficult to separate latitude and altitude as causes of a correlation. (b) The introduction is too broad.

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