

Review of Paper: **Particulate sulfur in the upper troposphere and lowermost stratosphere – sources and climate forcing** by Martinsson et al. 2017, submitted to *Atmospheric Chemistry and Physics*

Martinsson et al. studied sources regions of upper tropospheric sulfur using long-term IAGOS-CARIBIC data. The authors show that the relative contributions (either from transport below or mixed down from the stratosphere) in UT sulfur are dependent on seasons, volcanic activities. The paper has gone through the first round of review with substantial changes based on previous reviewers especially reviewer #1. In general the paper is in good shape, but some concerns (especially on tropospheric contributions) need to be addressed before publication.

- a. In the abstract, please give more definition on UT and LMS, i.e. LMS is ~3 km above dynamic tropopause; while UT is how many km below?
- b. Line 25 *“We find that tropospheric sources dominate during summer and the fall as a result of downward transport of the Asian tropopause aerosol layer (ATAL) formed in the Asian monsoon”*
Line 576 *“The ATAL is transported downwards, and affects the extratropical tropopause region in August to December.”*
These arguments are unclear to me, what does “downwards transport” mean? A recent paper “efficient transport of tropospheric aerosol into the stratosphere via the Asian summer monsoon anticyclone” shows that ATAL is mainly lifted above locally to the stratosphere and further transport to high latitudes. See their Figure 4.
- c. The same paper above argues that in summer/fall seasons, convections transport tracer gases (SO₂) and aerosol to UT. Do you think it maybe more likely a reason for higher tropospheric contribution of UT sulfur rather than downward transport of ATAL?
- d. *“SO₂ measurements by the satellite-based instrument MIPAS indicate a UT seasonal variation in the NH midlatitudes with low concentrations in December to March and the highest concentrations in June to September (Höpfner et al., 2015).”*
In my impression MIPAS SO₂ product is of large uncertainties, and may not be meaningful in UT. Shown in Rollins et al. (2017, GRL), MIPAS overestimates the SO₂ concentration in TP by a factor of ~3 or so.
- e. I found the discussions on SO₂, CO, OH are difficult to understand. Seems authors claim that direct transport of SO₂ and sulfate aerosol from lower troposphere is not going to explain the seasonal variation on tropospheric contribution to UT sulfur, because they will be washed out or cloud-processed. And the authors suggest the concentrations of OH and SO₂ are responsible. Well, are we certain that new TP, SO₂ oxidation is OH-limited? Any know seasonal cycle on SO₂ emissions can explain the seasonal variation? I think some surface SO₂ can survive in UT especially in deep convection.