

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2022-274

Anonymous Referee #1

Referee comment on "Enhanced sulfur in the upper troposphere and lower stratosphere in spring 2020" by Laura Tomsche et al., Atmos. Chem. Phys. Discuss.,
<https://doi.org/10.5194/acp-2022-274-RC1>, 2022

This paper by Tomsche et al. presents new SO₂ and SO₄²⁻ (along with other trace gas species) measurements obtained during the BLUESKY aircraft research campaign in May/June 2020, during the European Covid-lockdown which offered an unprecedented opportunity to measure air masses characterised by drastically decreased air pollution. Interestingly, the authors find strongly enhanced SO₂ and SO₄²⁻ concentrations in the upper troposphere and lower stratosphere, respectively, despite overall reduced air pollutant emissions. While these observations are of great value and offer a benchmark against which the chemistry in air quality and other chemistry transport models can be tested, I find that the conclusions provided in the abstract not to be supported by enough evidence within the paper. The explanations for the enhanced sulfur-species concentrations comprise a range of different possibilities, which the authors fail to meaningfully constrain by their evaluations. I therefore cannot recommend this paper for publication in ACP in its current form.

Specific comments:

L72, Section 2.1 BLUESKY mission:

- It would be interesting for the reader to have a short summary of the mission goals added here. It seems obvious but it should be made more explicit as you have done nicely in the abstract.
- Figure 1. Could the paths of the different flights be represented in different linestyles or shades of blue/red to help emphasize that they were carried out on different days? The current figure may show the coverage, is otherwise though not very informative.
- Also, you talk about coordination of the flights between Falcon and HALO, but the coverage is rather different. What was the main aim of the coordination?

L96 Sensitivity of measurement to moisture: It seems somewhat arbitrary to use a specific altitude as cut-off since you could find samples with high/low moisture content even below/above 8 km depending on the meteorological situation you're flying in. What is the range of H₂O mixing ratios you can/cannot easily perform this correction for? Did you measure H₂O and if yes, with which instruments?

L138 Could be written more clearly. I suggest to replace 'along the same longitudes and vice versa' with 'and vice versa along the longitudes outside this range'

L142 Related comment. I suggest to explicitly say that O₃ and HNO₃ are positively correlated as expected and repeat the longitude range here, since 'in the mentioned longitude range' may not be clear to readers given that you talk about two in L138.

L148 I would rewrite this sentence here to point towards the more in-depth analysis and discussion in Section 5 and without claiming it is 'just' from long-range transport. As it currently stands here, I cannot judge from Figure 5 whether the evidence you provide is good enough to underpin this result. For example, I see one trajectory rising from rather low altitude starting around the Eastern coast of North America ending at the measurement location.

Figure 3: I suggest adding the tropopause height onto the figure.

L163 It would seem important to indicate the average altitude of the dynamical and/or thermal tropopauses over the region during this time period as well, not to give the impression of choosing what fits best your lower bound of the mixing layer. I would expect the 2PVU tropopause being close to the 330K isentrope, so it would confirm your choice of ozone value for defining the chemical tropopause.

L203 Is this an expected concentration range for the mixing layer to be found in at these latitude bands and season?

Figure 4 and discussion section 3.3: It is really hard to follow the discussion of this figure. The only really outstanding feature I can detect when looking at these panels is that there are some high concentrations in SO₄²⁻ in the troposphere (at ozone values below 100 ppbv and CO values between 100 and 125 ppbv). It would be nice to have this feature explained by the backward trajectories. Otherwise, it is expected that SO₂ decreases and SO₄²⁻ increases as one goes into the stratosphere due to the aging of air, which is reflected both in the very strong anticorrelation between the two sulfur species and also in the SO₄²⁻ / (SO₄²⁻ + SO₂) ratio visible in Figure 2 and 3. Maybe you could circle the points you are referring to in the figure in case I have missed what you are really referring to?

L215/Figure 5. This evaluation would be more effective and convincing if you would identify the exact times/altitude/geographical location of when unusual trace gas observations are made and then calculate your backward trajectories from locations on a finer grid point around these. I then would suggest not only to show lat-lon plots, but also time-altitude evolution of the trajectories and also PV along with it, so that the reader can see where/when tropospheric influence or strong uplift within the Asian monsoon may have happened.

Figure 5 Why is there an empty panel (j)?

L253-262 I cannot fully follow your argumentation here. Don't you miss to account for the 72% reduction in air traffic due to Covid when you calculate your 50% increase in aviation SO₂ emission increases?

L291-295 This conclusion is not very satisfying. Much more should have been done to better disentangle all the different possible explanations of the enhanced SO₂ throughout the upper troposphere. The reader knows in principle as much as before reading your paper about the potential sources of the enhanced sulfur species. If you could do the backward trajectories in a more targeted way, that is identifying the origin of high-SO₂ events for different flights more meaningfully, this could be a much better step to answer the question you wish to answer.

Section 5: It would be interesting to see whether the clear anti-correlation in SO₄²⁻ with ozone seen for the flight in Figure 2 is also found for other flights. This could then be used to identify truly anomalous correlations for which again more targeted backward trajectories could potentially yield much more insight than is provided here. As the discussion is presented here, multiple explanations from existing literature but no additional insights are provided, leaving the reader once more unsatisfied about having learned anything new.

L360 I do not think that you have provided real evidence that you have measured enhanced SO₄²⁻ concentrations in the stratosphere, since we do not know what the 'normal' background would be. The difference seen in Figure 6 could simply be due to measuring within the troposphere at altitudes 11-15 km for the CAFE-Africa campaign data, which to my understanding was performed within the tropics.

Minor corrections/typos:

L31 geo engineering --> geoengineering

L90 in flight --> in-flight

L146 delete 'to' in front of '266 ppb...'

L179 write either 'with stratospheric sulfate aerosol' or 'with the stratospheric sulfate aerosol layer'