

Review of Toon et al: *Atmospheric Carbonyl Sulphide (OCS) measured remotely by FTIR solar absorption spectrometry* for
Atmospheric Chemistry and Physics

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The article by Toon et al is part of a special issue celebrating twenty-five years of the Network for Detection of Atmospheric Composition Change (NDACC) between ACP, AMT and ESSD. The paper describes the retrieval and resulting timeseries of carbonyl sulfide (OCS) from solar absorption FTIR measurements using the MkIV spectrometer, covering that 25 year period.

Traditionally, interest in atmospheric abundances of OCS has centred around the fact that it is the most abundant sulfur gas in the atmosphere and its role as a precursor for the stratospheric sulfate aerosol layer. More recently, studies have explored the potential of using OCS as a tracer for photosynthetic uptake, which would allow some additional understanding of the partitioning of net ecosystem exchange of CO₂ between respiration and photosynthesis components, which are co-located and difficult to differentiate between via direct CO₂ flux measurements or inverse modelling from atmospheric concentration measurements. OCS measurements, however, are relatively rare in comparison to those of CO₂, so it is therefore encouraging to see publication of long timeseries of OCS amounts.

One interesting aspect is the removal of stratospheric variability using the relationship with N₂O, something that has also previously been utilised to look at tropospheric methane columns.

In general, the paper is scientifically sound and worthy of publication. I would suggest that it perhaps more suited for AMT than ACP, but given

this is a shared special issue and it is not clear cut, I won't insist on that!

I have two major comments, though I hesitate to use the word major.

1. throughout the paper, references to S are spelt with 'ph'. While this is acceptable in (British) English, the IUPAC spelling uses an 'f'. I believe that is what should be used here.
2. the paper could benefit from some careful reading and editing. It currently reads a lot like it was written in a rush to meet a deadline. There are lots of places where the language could be tighter or more formal. There are also places where it is a bit repetitive and/or long-winded. I will try to point out these, but no doubt I will miss some so I'd encourage a careful re-reading during the revision process.

After addressing these and other minor comments the manuscript will be suitable for publication.

1 General Comments

- There are a number of sentences beginning with “And” or “So” that could be rephrased.
- Figures using colour scales should have a legend/key with the colour scale on them rather than having to search for a description in the caption

2 Technical Comments

- line 32 result → results
- lines 38-42 - what about the relative affinity of plants towards CO₂ and OCS? How does this affect the seasonal cycles?
- line 44 - ATMOS has not been introduced yet, but is used as an acronym several times before it is defined (currently about line 97-98)
- line 57 - decrease → decreased
- line 59 - repeat of “OCS column”

- line 100 - the section could maybe be split somewhere or renamed, as it covers not only the observations but also details on the fitting.
- line 108/Table 1 - millions of cubic feet? Is this actually relevant? If so, should it not be in SI units? Otherwise omit.
- Figure 1 - I realise it would clutter the figure, but it would be nice to see the interfering gases plotted in the spectrum as well. Maybe offset vertically from the OCS.
- Figure 2 and elsewhere - it would be good to have a key for the colour scale in the figure somewhere, rather than just a description in the caption
- line 152 - exact same → same
- line 160 - maybe relate the green points to what this means w.r.t. time.
- line 160, 167 - “it is clear” - maybe tighten the language here - this is somewhat subjective.
- line 180 - incomplete sentence here.
- line 186 - “fairly” linear - what is the uncertainty in this fit? e.g. what is the residual around the linear fit in this range?
- line 216 then → than
- line 221 - Max → maximum
- line 222 - e.g. clouds - clarify?
- line 223 - why the different number of coadds at lower airmasses? How were these numbers chosen (i.e. whether to average four or six spectra)?
- line 224 - maybe “averaged” instead of “average”
- line 238 - given you have used different selections of windows between the balloon- and ground-based spectra, can you be certain of the consistency between the analyses?

- line 244 - I appreciate this is going into an NDACC special issue, but there is no prior context in this article for NDACC or how it operates within the infrared working group.
- line 255 - you could maybe refer to previous work where stratospheric relationships between species have been used to remove stratospheric variability or infer tropospheric abundances.
- line 257-259 - was the consistency of the AKs the major criterion used in the selection? It would be nice to be clear here.
- line 262 - the same? or just similar?
- Figure 4 - legend for colours would be nice
- line 281 - how are you judging the precision and accuracy of your OCS retrievals?
- line 292 - the interfering species could be shown offset to the OCS. It would be good to see these, particularly to observe if there is any relationship with the patterns in the residuals.
- line 300 - remove “of course”
- line 302 - how are the surface pressure and H₂O column used to infer the dry air column?
- line 307 - delete “So” and maybe add “therefore after N₂O
- line 308 - is this really noise, or rather variability?
- line 309 - can you not quantify what the spread of values is?
- line 325 - season → seasonal
- line 329-330 - presumably, however, the fitting improves going from 2 harmonics to 3. Maybe clarify
- line 370 - was this the only time measuring at Fairbanks?
- line 372 - it is probably true that the enhanced seasonal cycle relative to other locations is due to the proximity of the boreal forest, but maybe you could explain why this is likely to increase the seasonal cycle amplitude

- line 407 - maybe briefly comment on how site-to-site biases could be assessed (or not, given the relative feasibility)
- Table 4 and elsewhere - what do you actual mean by precision? Precision is a term that can refer to a number of things, so a definition of how you are calculating/determining precision here would be good. Presumably it is something like the standard deviation in the retrievals over a period of time that you expect the atmospheric state to not change.
- line 428-429 - the large seasonal cycle reported in Griffith et al came from one year's data. Can the consistency of this with later years be inferred from any other studies (e.g. Kremser et al)?
- line 443 - wording at the end of the sentence
- line 451 - how was xCO₂ retrieved?
- line 458-461 - maybe tighten the language and reporting of the analysis here
- line 472-473 - maybe reword to make this less subjective
- paragraph from line 485 - this seems like it warrants a separate section, or that it should be condensed.
- line 518 - base → based
- line 521-523 - this could maybe be mentioned earlier
- line 526 - rms is an acronym, so capitalise
- line 705 - 120 ppt → 120 ppb. Also consistency - is it linear above 100 or 120 ppb?
- line 752 and thereafter - these are not complete sentences, reword please.
- line 824 - it would be interesting to see what average residuals look like, but admittedly most likely going into too much detail for this manuscript

- Table B.2. - is colour necessary here? You could separate with bolded horizontal lines or alternating shaded and non-shaded
- line 875-876 - to play devil's advocate - if these windows are systematically lower, could you not just scale them to match the other windows? In fact, I guess it is not inconceivable that these are correct (though unlikely, I will concede), because we do not know the "truth" here.
- line 880 - the values do not exceed 1 by very much...
- line 888 - subjectivity here
- line 895 - the worst? Or just having the worst agreement with the other windows?
- line 899 - maybe formalise the language a little
- line 934 - spectral → spectra
- line 939-940 - maybe explicitly state why this requires favouring the weaker N₂O lines (presumably because the OCS lines are weak).
- line 945 and thereafter - repetitive from Appendix B, so you could refer to that instead of explaining them again
- Table C.2. - units for center and width
- line 964 - you didn't define line mixing as LM
- line 969 - again - not necessarily the worst, but in the worst agreement with the other windows
- line 970 - formatting with E" etc.
- line 974 - why is this surprising? Unless I have misunderstood, you have argued extensively that the narrow windows should have better fits/RMS.

Apologies for the delayed response.

3 References