

Global Optimal Estimation Retrievals of Atmospheric Carbonyl Sulfide Over Water from IASI Measurement Spectra for 2018

Response to Reviewers' Comments

We thank the reviewers for taking the time to evaluate our manuscript and for their positive and helpful comments. These comments are reproduced below in *italics*, followed by '>>' and our responses.

Reviewer 1

The study addresses the problem of estimating atmospheric OCS from IASI radiances over the ocean and inland water bodies. The analysis is global and considers the year 2018. The retrieval methodology utilizes a suitable forward model and employs Optimal Estimation to address the inverse problems. The study is well-organized, featuring a detailed analysis that compares IASI retrievals with other OCS products, as well as in situ and remote sensing platforms.

In general, I enjoyed reading the paper, which addresses the issue of OCS retrievals from IASI with a high degree of maturity. Therefore, my remarks are minor, although they address some technical points that I think need clarification to strengthen the good work done by the authors.

General comments

*Correlation between DOF and surface temperature. The correlation is explained by the supposed high thermal contrast, which, according to the authors, is higher in the tropical region. I do not like this explanation. In general, the thermal contrast is vital for day soundings over land and the ocean, especially in tropical areas where it is almost zero. Also, remember that for the sea, the emissivity is approximately 1 everywhere in the spectral domain of interest. Therefore, the thermal contrast is irrelevant. In effect, from Fig. 4 we see that the AK close to the surface is nearly zero. **The correlation is primarily an effect of the larger signal present in the tropics; therefore, the correlation with surface temperature is trivial and does not add significantly to the problem.** I would spend only a few words about this trivial effect, trying not to invoke thermal contrast, which is again irrelevant.*

>> Thank you for this detailed interpretation of our statements around DOFS and thermal contrast. The authors agree that explaining higher DOFS using thermal contrast in the tropics is not fully accurate. Upon revisiting this discussion, plots S6 and S7 show that thermal contrast does not necessarily correlate with higher DOFS. However, there is some correlation between thermal contrast and OCS total columns, which is somewhat to be expected due to the nature of interpreting radiative transfer and atmospheric absorption in the infrared, i.e., improves how well we are able to distinguish a signal of atmospheric absorption of trace gases – which you mentioned in your comment. In any case, we have toned down the comparison between DOFS and SST.

The discussion about OCS columns and thermal contrast at the start of Section 3.2 remains, as that relationship is seen in the supplementary figures.

*A more interesting point on which I would like to draw the author's attention is the possible **seasonality in the DOF**, which is **driven by the normal dependence of weather conditions on the season rather than biological effects**. The problem is addressed extensively in the paper doi: 10.1117/12.2599761, which the authors should consider and **use to demonstrate that the OCS retrieved is not affected by spurious behavior driven by the background modulation with the season**, rather than by biogenic activity leading to the natural sources and sinks of OCS. This modulation effect is expected to be negligible within the tropical belt, where there are no seasons, but could have an impact on mid- and high-latitude areas and stations, as shown in Fig. 12.*

>> Thank you for this comment. We read this manuscript thoroughly and thought it very insightful in emphasising the challenge in differentiating between source and sink acquisition locally to peaks and troughs in columnar OCS, and meteorology/airmass advection. While airmass transport is highlighted as a major consideration, the authors do also highlight that the DOF variability is dependent on surface temperature and emissivity, mainly over land. So, we don't think it's necessary to go into extensive analysis and discussion in this work. However, it is something that will be important to consider in our future work on retrievals over land.

A reference to this work has been included at the end of Section 4.2. With a brief highlight to how our work may be subject to these effects.

A further comment on this referenced work; it was very interesting to read at the end of Section 3.2 that there are believed to be sources of OCS in Northern Africa. These hotspots of OCS have appeared in our preliminary work on land retrievals, and an immediate assumption was poor handling of high albedo over desert regions. So, this is an exceptionally important distinction highlighted by the authors.

Reviewer 2

The paper presents a comprehensive analysis of OCS retrievals with IASI and a detailed comparison with modeling and ground-based datasets. I think that the paper is generally well-written and contains a lot of information which is of interest to a broad audience. The role of OCS in the atmosphere has been emerging as key to many aspects of global interest lately, therefore any effort devoted to obtaining long-term, global records of OCS abundances is relevant and timely. This manuscript makes no exception.

Specific comments

1) The introduction section is very detailed and includes a lot of information. For the purpose of showing sources and sinks of OCS, I think that the manuscript should include an infographics with fluxes and sources/sinks and related processes. It certainly would take some time, but it would be a great addition to the paper, and would bolster the readability of the introduction.

>> Thank you for the suggestion. We agree it would be a valuable addition, however, there are follow-up publications in discussion amongst the co-authors where an infographic of this style would be better suited. Specifically using this satellite data in flux inversions and source/sink attribution. We hope this is ok and look for an infographic in one of those publications!

2) Lines 202-205: Rather than talking about the fact that the signal due to background OCS is above noise level, it would be more useful to say how big of an OCS variation would yield a detectable signal by IASI.

>> Thank you for this suggestion, it's certainly worth clarifying. Additional tests have been performed to investigate this. Please see supplementary figures S1 and S2. These show a detectable OCS signal in profiles with a tropospheric concentration to about 100 ppt. Additional text has been included after the lines highlighted above.

To summarise, we used the RFM to model TOA spectra using different OCS profiles, scaled from a tropical a priori, using meteorology used in the ULIRS, for a pixel at 3.6°S and 90°E.

3) Lines 225-226: Is this because of specific contamination from some gases? In any case, contamination is always present, and this sentence probably can be removed.

>> This sentence has been moved to later in the paragraph, after we have specified the P branch is of most interest due to less overlap with H₂O absorption features, when compared to the R branch.

4) Line 226: Please note that IASI apodized resolution is 0.5 cm⁻¹, while the sampling is 0.25 cm⁻¹. There is no ambiguity about that.

>> This is in reference to the modelled resolution of the absorption spectra shown, i.e. spectra was modelled at intervals of 0.25 cm^{-1} wavenumbers using the RFM, to match that of the sampled IASI spectra.

5) Line 264: *I was wondering if the authors considered using a covariance for temperature profiles built using a representative dataset of profiles (such as ERA5) instead of a Markovian matrix with a uniform variance across all altitudes.*

>> As we were using the IASI L2 temperature profiles we use only a nominal temperature covariance, to allow a small amount of flexibility. The assumption is that the profiles used are of high quality for the scene being viewed as they are already a Level 2 product. Furthermore, while T is in the state vector, like the covariance, that is mainly there for fitting purposes in the forward modelling, rather than expecting significant change.

6) Line 275: *Do you use any inflation factor for the IASI noise to account e.g. for eventual spectroscopic biases? If so, please specify. In any case, I would comment on any choice made as this is typically a relevant point in any retrieval scheme based on Optimal Estimation.*

>> The instrument noise data was delivered to us from EUMETSAT via CNES in 2020, via an ftp server, rather than a public website. The instrument noise is quantified and applied to each pixel in the IASI 2x2 IFOV. No scaling is applied to account for biases. A further line and clarification has been added in the text at Line 204.

7) *Caption of Figure 4: the coordinates of point (b) as specified here are not in the indian ocean, please check the coordinates (perhaps 64.1 E?)*

>> Thank you. This was indeed meant to be 64.1 E.

8) Lines 332-333: *I would probably expand a little bit explaining why, as the concept that a correlation between chi square and surface temperature does not translate into a bias in the retrieved column is not obvious.*

>> Thank you for this comment. This has been addressed with further elaboration on Line 326-331.

9) Line 494: *that altitude range (> 5 km) is above the airmass that contains the large majority of OCS total column. Given that, it would be very beneficial if the manuscript would comment on the differences in light of the IASI retrievals presented.*

>> This is a good point. We also compare TOMCAT with ground-based observations in the referenced publication (doi.org/10.5194/acp-23-10035-2023), so there is some evaluation of the model done at the surface. However, We understand that the comparison of total columns includes a region of poor sensitivity in the measurements and of minimal evaluation in the model. Additionally, the need to validate the model further is emphasised in the proceeding paragraph.

Reviewer 3

This manuscript by Cartwright et al. (2025) presents a study entitled "Global Optimal Estimation Retrievals of Atmospheric Carbonyl Sulfide Over Water from IASI Measurement Spectra for 2018". The manuscript describes scientific details about the IASI OCS retrieval algorithm, the comparison between IASI OCS retrievals and TOMCAT simulations, as well as NOAA surface measurements, and outlines further development and research plans. This is an important dataset for advancing satellite-based OCS measurements and is highly valuable for OCS-related studies. The manuscript is well-written, and the figures are presented clearly. I recommend the publication of this manuscript after the authors address the general and specific comments listed below.

General Comments

1) *Why the land OCS are not retrieved? I think the land OCS retrieval are quite useful.*

>> In preliminary testing we saw questionable results from using this same retrieval setup over land. It was suspected this was associated with the characterisation of emissivity in the forward modelling. A follow-up piece of work can be found in the lead author's PhD thesis on addressing this and attempting to resolve issues using a spectrally emissivity product – CAMEL: <https://doi.org/10.25392/leicester.data.23920251.v1>. Co-authors David Moore and Jeremy Harrison are leading on-going development to the retrieval to incorporate land OCS retrievals.

2) *Why the COS radiance is very smooth compared with other species (CO₂, O₃, H₂O, CO), in Figure 1?*

>> The spectral features of OCS are very fine spectrally, which means when modelled at a resolution of 0.25 cm⁻¹ (the same as sampled MetOp measurement spectra) they are not resolved, and it takes an average over the spectral intervals considered. This results in a smooth absorption spectra compared to one with more visible features. See Figure 3.2 in the linked thesis above or Figure 8 in <http://dx.doi.org/10.1016/j.iqsrt.2017.07.006> for reference.

3) *ACE-FTS profiles are used in the algorithm as a priori profiles. Will other OCS satellite observations be considered to provide prior information?*

>> ACE-FTS provides very good representation of OCS down to its observing limit (around 6 km) and effectively has 1 DOF per measurement altitude level. That being said, when compared with other satellite products, i.e. MIPAS, ACE-FTS does differ slightly, but as we use fairly loose constraint on the OCS covariance, the choice of one such product over another should not significantly impact retrieved IASI OCS. Data more commonly used for *a priori* profiles in optimal estimation approaches, tend to be modelled. However, as we wanted to compare the retrieved total columns with modelled OCS from TOMCAT, we opted to utilise satellite observations and a tropospheric constant, rather than using TOMCAT profiles, so not to create a bias towards TOMCAT.

4) *The abstract includes technical details (e.g., MetOp-A/B, ULIRS) that may obscure readability for broader audiences. I recommend simplifying technical terminology and prioritizing the study's scientific significance. Specifically, emphasize the research motivation, e.g., the paucity of OCS observations (both ground-based and satellite-derived), to clarify why this work is needed.*

>> Thank you. This is a fair assessment. Part of the reason for the use of technical terminology and abbreviations is the rigid word limit of 250 for abstracts within EGU publications. The scope of the abstract could be focused more on the importance and usage case of this dataset. We have completed a re-write.

Specific Comments

Line 23: *"This study demonstrates the ability of the IASI instrument to accurately measure OCS in the troposphere". Satellite instrument is never accurate.*

>> Amended accurately measure to 'detect'

Line 176: *Table 1 missed a bottom line.*

>> Thank you. Amended

Line 213: *"(OCS, CO₂, H₂O, temperature profiles and surface temperature)". CO₂, H₂O should be CO₂, H₂O.*

>> Thank you. Amended.

Line 216: *"i.e. in b, but their contributions were not adjusted." b should be bold b.*

>> Thank you. Amended.

Line 287: "and χ^2 is the cost function, J (Rodgers, 2000), referred to as the chi-squared test." χ^2 is not the cost function. Please rewrite the sentence.

>> We have condensed the sentences down to: "A good quality convergence is considered to have been reached if $\chi^2 \approx m$, where we consider m is the first part of the right-hand-side of Eq. **Error! Reference source not found.**, normalised by the number of measurement channels (see Fig. 2a), referred to as the chi-squared test.". And removed other mentions of the cost function.

Line 298: "Averaging kernels, denoted $\mathbf{A}$ in". "denoted $\mathbf{A}$ " to "denoted as $\mathbf{A}$ ".

>> Thank you. Amended.

Line 313: "profile provides approximately one piece of information for the OCS column." What does it mean of one piece of information for the OCS column?

>> This is in reference to how much information we are extracting from employing satellite observations to resolve the forward problem. To have one degree of freedom for signal, or one piece of information, across a range of atmospheric layers would suggest we are confident in the OCS quantity in that partial column. Where we have distinguished information of OCS from the signal, rather than the contaminating instrument noise or other species.

Line 346: "where T corresponds to the final iteration of Eq. 7," T should be $\mathbf{T}$.

>> Thank you. Amended.

Line 349: "As shown in Fig. 6, S_s is the major", S_s should be S_s .

>> Thank you. Amended.

Line 352: "This is also the case for S_m , the". S_m should be S_m .

>> Thank you. Amended.

Line 460: Figure 9 is generally in red color at a first glance. Can the figure be improved for a better visualization?

>> Thank you. We agree and have updated the figure to start the colourbar at 5%.

Line 563: "are between 18 and 38%," 18 to 18% to avoid misunderstanding.

>> Thank you. Amended.

Line 569: "and can be linked to surface fluxes." More specially, this is linked to the biosphere uptake, which can lead to smaller COS mixing ratios in the atmosphere.

>> Included: "in this case biospheric uptake, which leads to smaller atmospheric OCS mixing ratios."