Response to Reviewer #1

Many thanks for the helpful work of Referee #1 on our manuscript. Below you can find our specific answers to the comments in blue. We have also revised the manuscript according to the suggestions by both Reviewers. The changes are marked with red in the word document. For extended changes we have indicated the line-number of the revised manuscript. We have acknowledged the two anonymous reviewers in the acknowledgements.

General Comments

The authors present a retrieval for the measurement of total column ozone based on the "QASUME" reference spectroradiometer, based on absolute measurements of the spectral irradiance which are traceable to SI. This general concept is a break from the traditional notion of "traceability" in the ozone community which has historically always meant traceability to an artefact, namely Dobson 83 or the Toronto Brewer triad.

The manuscript is very relevant and suitable for publication in AMT with minor revisions. Most of my specific comments below are essentially just requests for a bit more guidance for the reader.

Due to this comment and due to the comments of Reviewer 2, we have re-worded some sentence of the manuscript, for better guidance for the reader.

During the revision of the manuscript, we have added two corrigenda:

1. *For the final standard retrieval, we have used the SO2 cross section HITRANS from Hermans (2019) (Line 203)*
2. *The uncertainty from solar spectra is corrected to be 0.196 % since the factor 0.5 was not included in the calculation (line 285, Table 1).*

My one overall comment is that I would appreciate some more introductory motivation and discussion for the concept of a "traceable" retrieval, as I suspect this idea will not be very familiar to many of the readers of AMT (in contrast to the idea of a traceable measurement in a laboratory setting, or the concept of an uncertainty budget for an atmospheric retrieval, both of which are widely understood).

What is the motivation for the approach taken here in the manuscript, compared to the traditional Dobson and Brewer concept of traceability? (You do already give two advantages in lines 16-17 I note).

We appreciate this positive review of our manuscript. Regarding "Traceability" we have introduced the motivation in the abstract and now included some more introduction of the motivation of the work in the revised manuscript (e.g. Lines 66 – 73, Abstract and Conclusion).

Some specific points I would like to see discussed are as follows:

Is the definition of "traceability" of a retrieval you give (line 73), in common use?

We clarify that we have used our own definition of "traceability" for retrieval with a model. To our knowledge there is not a common definition of traceability regarding retrieval models. We have clarified in the revised manuscript. (Lines 73).

Additional data are required in the retrieval, for example profiles of ozone and temperature measured by ozonesondes. However these measurements are not traceable, unlike those from QASUME – what is the impact of that?

Many thanks for this hint. Indeed, some input parameters are not traceable. In order to take this into account, we have used a very conservative value for the estimated uncertainty of these components. The impact of the uncertainty of the effective ozone temperature from sondes and reanalysis data is investigated and stated in more detail in the revised manuscript (lines 304 - 308).

The uncertainty is based specifically on conditions in Davos. However, the text states the instrument has been used at 33 different locations around the world. Does this mean the traceability of the ozone retrieval does not apply at these locations?

We have highlighted that the results of his study are based on the specific location at Davos. We also clarify, that QASUME is used at the mentioned 33 station for global UV irradiance and not for direct spectral solar irradiance measurements to retrieve total column ozone. At two specific locations (Teneriffe and El Arenosillo, Spain) QASUME measured direct solar irradiance and retrieved TCO. The results will be published in a WMO report in 2022. Preliminary results compared to Brewer showed similar results as in this study. We now mention the two other sites in the revised manuscript (also according to the suggestion of Reviewer 2 (lines 413 - 417).

Even though you have an established climatology it is always possible for the atmosphere at any one moment to be in a very unusual state, or even in an unprecedented state – how does your approach cope with that? Can you still say "accounting for all possible uncertainties"? (Line 19)

Indeed, this statement is a bit exaggerated. We agree with the Reviewer that might be some uncertainties which are not considered in the study. We have removed "for all possible uncertainties".

Specific Comments

Line 28 The website is useful but you need to also give a formal citation, for example to the most recent assessment.

Done

Line 32 "Variations of the solar constant" isn't the main point here.

Done – we have removed this part

Line 45 perhaps "with the then state-of-the-art"

Done

Line 55 What do you mean by "fundamental" here? You then go on to say most of the difference can be accounted for if well-understood issues are taken into account.

We have removed "fundamental" and added "biases" instead of "uncertainties"

Lines 58-60 I think it would be better to list a wider range of instruments here that have also used a similar spectral range even if not in the same geometry. For example, NDACC UV spectroradiometers have been making similar measurements for decades (although global, not direct) from which it is possible to retrieve total ozone. Similarly there are the DOAS and MAX-DOAS instruments which are nowadays very widely used (using zenith rather than direct-sun measurements of course).

Many thanks for your suggestion. However, we in this study we would like to focus on TCO instruments measuring direct irradiance only.

Line 64 Actually, I would have "expected" the opposite. Measuring the full spectrum gives you many more data points but, each point does not necessarily contain any additional independent information and, speaking very generally, a ratio is usually able to be measured much more precisely than an absolute quantity.

We agree that our statement is rather speculative. We have removed the sentence.

Line 72 This definition of "traceability" (for a retrieval) is not the same as the definition of traceability for an instrument. You need to explain to the reader whether this definition is your own or is generally used.

Indeed, we have used our own definition of "traceability" for retrieval with a model. We have clarified in the revised manuscript (lines 73).

Line 111 Is the airmass for ozone?

Yes. Done

Line 119 Why is "traceable" in quotation marks here – is the implication that you are using the word in a particular way?

Since we have defined "traceable" earlier we have removed the quotation mark.

Lines 147-151 It seems a bit odd to me that you would show the plot of residuals before giving any details of what you are fitting. Normally in a paper this would be the other way around. Perhaps you could let the reader know the details are coming.

We have clarified this issue.

Line 152 Saying the residuals are "flat" seems a bit optimistic to me, there is some possible structure there, apart from the high frequency variation

We have formulated the sentence more conservatively (line 151)

Lines 152-154 Is it possible to explain this more clearly?

We have reformulated the sentence (line 153 - 154)

Line 162 Using afglus can only be a starting approximation for a specific location and season etc

Recalculation TCO with other standard atmospheres than afglus revealed neglectable changes of TCO in Davos. We have revised the manuscript accordingly (line 191).

Line 174 Using values for that particular day, or an average, or something else? Would this introduce an additional uncertainty?

As in Gröbner et al 2021 and specified in the manuscript we have used interpolated values from soundings every 2 or 3 days (lines 175 - 178).

Lines 171-174 As mentioned in my general comments, the use of outside information like the Payerne ozonesondes provokes a number of questions. Ozonesondes and radiosondes have their own issues of course and are not traceable to SI. Do you know how representative Payerne is for the vertical structure of ozone over Davos?

Gröbner et al. 2021 compared the effective temperature from Payerne with ECMWF reanalysis data from Davos. The differences between the two datasets was defined as the uncertainty of effective temperature in the manuscript in section 3.4. We have added that the effective temperature is linearly interpolated for the missing days and smoothed with a 10 days tunning mean as in Gröbner et al. 2021 (lines 174 - 177).

Lines 175-181 I think you should mention the limitations of this assumption for the AOD.

A more appropriate parametrisation would use the Angstroem approximation (AOD=beta . lambda^-alpha, with lambda the wavelength in micrometer. The tests we have performed have shown that the retrieval of TOC is not affected by using either approach (linear parametrisation of AOD or using the Ansgtroem law). This is essentially due to the short spectral range of 45 nm that is used for the retrieval algorithm, between 305 nm and 350 nm, where a linear or Angstroem AOD fitting function does not show any significant deviations. Clearly, for a larger spectral interval, the use of the Angstroem fitting function would be more appropriate (lines 184 – 185).

Lines 185 You should give at least some details of your assumptions for SO2, and how good you expect them to be, particularly as SO2 is likely to be highly variable in time and space. Lines 190-199 How do you know there will not be other non-negligible absorbers within this wavelength range?

Also in Gröbner et al. 2021 it can be found that: "the main atmospheric absorber in the measured wavelength band is ozone. Even though sulfur dioxide and nitrogen dioxide also absorb in this wavelength range, their amount in the atmosphere above Arosa and Davos is so small that it can be neglected here". We have added this information in the revised manuscript (lines 188 – 189).

Line 206 Is the "overall uncertainty of 0.91%" for every wavelength?

The uncertainty is not given for every wavelength in this region of 305 – 345 nm. at wavelengths shorter than 305 nm the uncertainty is slightly larger. Therefore, we use 0.91% for the entire wavelength band.

Line 213 Shouldn't you assume the more conservative limit of 0.72% ? Is there a physical consideration here?

Effects of substantial spectral correlations can be detected by the differences of QASUMEFTS (which is derived from QASUME) and an independent solar spectrum (TSIS). The impact of potential spectral correlations is included and discussed in the uncertainty assessment from two different solar reference spectra. (lines 191 - 193). Therefore "unfavorable correlation" from the measurement uncertainty is a even more conservative assumption than "no correlation". Finally, there are no correlations stated in

the uncertainty assessment of QASUME, the cross section or the solar spectrum. We have clarified in the revised manuscript. (e.g. lines 214, 218 – 219, 289 – 291).

Lines 215-216 Wouldn't the better approach here be to consider the uncertainties of the laboratory-measured ozone cross-section, (which I understand was one of the original motivations for the ATMOZ project) and propagate them through to the resulting total ozone value?

Lines 245-248 I don't follow the reasoning here. The ozone cross section at a specific temperature and wavelength has a true value which can in principle be measured, and we hope is getting more accurately measured as laboratory techniques improve. Why even consider the older Bass & Paur values at all?

This is a good point. Indeed, we have calculated the impact of the uncertainty given by the cross section as random (uncorrelated) noise to the cross section. However, due to the convolution of the cross section the noise was reduced, and the resulting uncertainty was less than 0.06%. To obtain a more realistic value we decided to estimate the uncertainty by comparing TCO from different cross section, assuming that all cross sections may be measured with best possible technology. Since Bass & Paur is still in use for brewer and Dobson we have also included this cross section.

We have clarified this in the revised manuscript (lines 226 - 231).

Line 240 This is quite confusing. Is there a difference between IUP and IUP_A and IUPA?

We have clarified this in the revised manuscript. We now use IUPA instead of IUP_A through the entire manuscript.

Lines 250-266 Again I am struggling with the reasoning. Doesn't the TSIS come with an uncertainty, which you could propagate through to the total ozone value?

According to the same argument as when considering the noise to the cross-section (uncertainty less than 0.06% due to convolution) we have chosen a second ETS to assess the overall uncertainty. We have added this argument in the revised manuscript (lines 272 – 277).

Lines 269-285 Is the standard deviation really enough, do you know whether extreme values are properly represented in the uncertainty? Also do you need to take into account the uncertainty in the mean value of 225.2 K caused by radiosonde errors or bias?

We assume that the measurement uncertainty of the radiosondes is far below 2.5K. However, the 2.5K from the comparison of soundings and reanalysis data seems to cover realistically the overall uncertainty (lines 305 – 308).

Lines 309-310 It is not clear to me what happened to the aerosol and SO2 and their effect on the uncertainty?

Aerosols and SO2 are used as fitting parameters. Their effect is covered by the computational uncertainty from the least square fit algorithm.

Line 323 Do you use a normal distribution for the random values?

The type of the distribution is indicated in Table 1 second column. We have indicated this in the revised manuscript (line 356).

Technical comments

Line 29 Remove the apostrophe

Done

Line 32 "surface of the Earth" or "Earth's surface"

Done

Line 40 "have" should be "has"

Done

Line 41 "to [form] a global network"

Done

Line 88 "has been" operated

Done

Line 91 Replace "chapter" with "section"

Done

Line 99 "enables" rather than "ensures", or perhaps "ensures outdoor measurements are able to be made"

Done

Line 103 Replace "timeout" with "missing "

Done

Line 109 Insert "to" after the word "equal"

Done

Line 109 "a maximum of 4.5 minutes"

Done

Line 112 "described" not "describes"

Done

Line 137 "et al."

Done

Line 149 "surface of the earth"

Done

Line 151 You don't mean "exemplarily" here

Done

Line 182 You need to re-word this, at present it reads as if Lord Rayleigh is personally scattering the photons around!!

Done

Line 203 Delete "well", otherwise it sounds like the authors are complimenting themselves!

Done

Line 207-209 I can't quite follow the meaning of this sentence, please re-word to make it clearer

We have reworded the sentence

Line 324 Apostrophe should be a comma.

Done

Line 330 Apostrophe should be a comma.

Done

Line 334 "negligibly" instead of "neglectable"

Done

Line 336 "proves" not "proofs"

This is removed due to the comment of Reviewer 2.