

We thank the reviewer for the comments and suggestion, and apologize for the misprints.

Review of the paper "Tropospheric ozone retrieval by a combination of TROPOMI/S5P measurements with BASCOE assimilated data" by Klaus-Peter Heue et al.

The manuscript presents a new way to retrieve global tropospheric ozone by mainly using TROPOMI NRTI total ozone and BASCOE assimilated stratospheric ozone profiles. A bias correction climatology was used to reduce BASCOE deficits in the upper stratosphere. The retrieved ozone column is mapped on the TROPOMI measurement spatial resolution by interpolating BASCOE sub columns in space and time to TROPOMI. Results are validated with ozone sondes, TROPOMI CCD tropical ozone columns, and OMPS-MERRA2 data. A short section discusses the results for different regions of the Earth. I recommend publication after addressing the comments given below.

Does the paper address relevant scientific questions within the scope of AMT?

Yes

Does the paper present novel concepts, ideas, tools, or data?

Yes

Are substantial conclusions reached?

Yes

Are the scientific methods and assumptions valid and clearly outlined?

This needs to be improved. See comments below.

Are the results sufficient to support the interpretations and conclusions?

Yes

Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?

No formula was given in the paper.

Do the authors give proper credit to related work and clearly indicate their own new/original contribution?

Yes

Does the title clearly reflect the contents of the paper?

Yes

Does the abstract provide a concise and complete summary?

Yes but it should be upgraded a bit: Some typos (TROMOMI,..) and missing information (tropopause level?)

Is the overall presentation well structured and clear?

The presentation is OK but can be improved (see comments).

Is the language fluent and precise?

Quite some errors throughout the paper that need to be taken care of.

Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

No formulae, symbols/units must be streamlined (see comments)

Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

The subsection in section 5 are rather short, could be compressed into one? Otherwise see comments below.

A table/overview of bias/dispersion of BF vs OS, CCD, OM2 might help to get a clear view on accuracy/precision.

Are the number and quality of references appropriate?

Yes

Is the amount and quality of supplementary material appropriate?

-

Questions/Comments:

Lines:

6: OMPS-MERRA-2 data (globally? time span?)

globally for the time period where S5P-BASCOE is available -updated in the manuscript

11: exceptional larger positive deviation (where?)

mostly in the northern polar regions - updated in the manuscript

12: expected spatial and temporal pattern (what is expected?)

in the tropics a wave one structures is expected and in the mid latitudes an annual cycle with a summer maximum - updated in the manuscript

24: beginning of the mission in 2018 = wrong, 2018 op. phase E2

Updated

33: MERRA-2 = citation?

Gelaro et al., 2017 , updated

65: It makes use = the dataset? sounds strange

replaced by "The algorithm makes use of"

72: ozone sonde will be explained briefly = data?

updated

77: total ozone column is retrieved = use of op. data? which one?

NRTI operational data as described in section 2.2 data - updated in the manuscript

75: maybe reorder chapter 2.1 and 2.2

We fully understand the referee's suggestion, but want to keep the current order as we first give a brief overview, before going into details. This was emphasized by adding references to the respective subsections.

94: TROPOMI data were reprocessed internally at DLR = full mission reprocessing to 2.1?

Yes - that is the advantage when total ozone processor was developed at the same institute, however this data set is not public available

update to "the DLR internal full TROPOMI mission reprocessing"

101: to prepare the EU Copernicus Atmospheric Monitoring Service = for what?

replaced by :

This implementation of BASCOE was developed to improve the representation of stratospheric composition in the EU Copernicus Atmospheric Monitoring Service (CAMS), by providing independent analyses of ozone and five other species which are also observed by MLS (HCl, ClO, HNO₃, N₂O, H₂O). These analyses are used to evaluate the analyses and forecasts of stratospheric ozone which are delivered operationally by CAMS (e.g. Ramonet et al., 2019) and also to verify research versions of the CAMS system where the stratospheric chemistry module from the BASCOE system is implemented into the CAMS system (Huijnen et al., 2016).

102: reduced, as expected for the austral spring = few words of explanation can help

The observed reduction in the stratospheric ozone is caused by the well known ozone hole.

updated in the manuscript.

112: Is there a change in BASCOE bias over time or is the early version not used anymore?

The early version is not used anymore; the current version is used since 2016 and the bias reported here is stable. Here is the revised paragraph:

“An early version (q2.4) of BASCOE-FD has been evaluated against total ozone ground-based measurements, ozonesonde profiles and satellite profiles over the period 2009-2012 (Lefever et al., 2015). The agreement was usually within +/- 10 % but degraded to +/- 40 % in the tropical tropopause layer (TTL). The version used here (5.7) runs operationally since March 2016. It is evaluated every three months for the validation of the CAMS operational analyses, indicating stable biases which are usually smaller than 5 % in the middle stratosphere and 15 % in the TTL (e.g. Sudarchikova et al., 2021).”

118: climatology: explain (e.g. resolution in time/space)

The original resolution is 5° latitude and 1 day. the climatology was interpolated to 2.5° latitude and smoothed in time. The information was added to the text.

Fig1: add resolution to caption (mixingratio in title)

updated

we added the total and tropospheric column as suggested by the reviewer and combined the figures 1 and 3 together with these new ones in one overview plot.

120: Where does the PV/PT data comes from?

The PV and PT data are imported from ECMWF

121: Outside the tropics = which latitude? Is there a sharp cut or a mixed region?

The tropics extend to 30° N and S. In this region the maximum pressure level $\text{Max}(P(\text{PV}=3.5), P(\text{PT}=380\text{K}))$ is used. Outside the tropics only the $P(\text{PV}=3.5)$ is used. There is no mixed region, we clarified it:

"The calculation of the tropopause pressure is done in two independent steps, first the PV, PT tropopause is calculated. Outside the tropics (outside 30° South to North) the tropopause is defined as the Potential Vorticity isosurface at 3.5 PVU and inside the tropics as the isentropic isosurface with a potential temperature of 380 K or 3.5 PVU, whatever is lower.

The second steps is based on the WMO (World Meteorological Organisation) definition, lowest level with dT/dz is ≤ 2 K/km and remains ≤ 2 K/km in the next 2 km. The potential vorticity and temperature are extracted from ECMWF operational analyses at a reduced spectral resolution (T31) corresponding to the coarse grid of BASCOE-FD.

In the final step the two definitions are combined by choosing the lower altitude / higher pressure level. For practical reasons the centre pressure level of the respective grid cell is given. "

Fig.2: what is the bias/dispersion of MLS stratospheric columns? Can longitudinal variations be neglected?

As stated by the MLS Quality and Description document (https://mls.jpl.nasa.gov/data/v4-2_data_quality_document.pdf , May 2022) the accuracy of MLS O₃ column values is around 4 % and the average biases between MLS and latitudinally-binned data from lidar and ozonesonde sites across the globe is typically within 5 % or better, with poorer behaviour at low latitudes in the UTLS. Added to the manuscript.

We have to admit we never looked into the longitudinal variations in the difference between BASCOE and MLS. Due to the fact that ozone concentration above 4 hPa is low we assume the longitudinal change to be of secondary interest.

127: BASCOE ozone deficit above 4 hPa = citation available?

Yes, this deficit is described in Errera et al., 2019, already cited in this context (l 117), more details are given in Skachko et al 2016. (referenced therein)

131: 5.5 x 3.5 km² = not in 2018! pixel size switch at 2019/08/06
corrected

132: observation time = UTC time changing for every pixel?

The unit of time interpolation is not relevant here. Of course a consistent time unit has to be used between TROPOMI and BASCOE and other time-dependent data (e.g. external tropopause).

The time of the S5P observation does not change for every pixel but for every scanline, however, during the interpolation the fact that all pixels in a scanline are taken simultaneously is not used, thereby the algorithm can be easily transferred to other sensors e.g. GOME-2.

no change in the manuscript

Fig3: BASCOE, right? add resolution to caption, corrected for bias?

Fig 3 shows the stratospheric column (BASCOE including bias correction), interpolated to the S5P pixels, so it has the S5P resolution of 5.5 x 3.5 km. Clarified in the figure caption.

we added the total and tropospheric column and combined the figures 1 and 3 together with these new ones in one overview plot.

where does the jump in column near date line (~170°) come from?

The data shown are interpolated to S5P resolution in space and time. At 170°W you can see both the first and the last orbit of this day. The jump is caused by the time difference of ~23 hour between the measurements in this area. Clarified in the figure caption

Why only show stratospheric column and not tropospheric in section 2.4?

The referee is certainly right it would be interesting to show the tropospheric data as well. On the other hand, we already show many figures and examples for tropospheric columns are shown in section 5. Therefore we introduced the overview plot for the figure 1 and 3. together with total and tropospheric column.

142: Dec.2021: inspection date?

According to the AMT guidelines the access date has to be given with the webpage, clarified.

150: to correct what? what climatology was used and how?

151: Does the correction depend on difference between reference and cloud height?

To normalize the above cloud ozone column for the varying cloud altitudes to the reference level, the partial ozone column between the cloud optical altitude and the reference level was added. This correction column is based on the climatology by McPeters and Labow (2012).

152: Tropical clouds are not on average at 10km, especially convective clouds.

The S5P cloud retrieval underestimates the height in comparison to e.g. CALIPSO.

The S5P cloud retrieval might underestimate the cloud top height, especially for ice clouds, but what is more important for the tropospheric ozone calculation is to use the same cloud properties as in the total column ozone retrieval. Therefore, we use the cloud data given in the total ozone data file.

155: what is the defined correction height? 10km or 280hPa.

The operational S5P product is adjusted to 270hPa?

This was typo in the manuscript, at the beginning of the S5P CCD project the reference level was at 280hPa but we changed it to 270hPa, somehow I kept the 280 here. Corrected

157: why certain bands?

corrected: "It is assumed that for each latitude band the stratospheric ozone column is constant along the longitude and varies only slowly in time." see also answer to review comments #2.

168: climatology the same as used for cloud height correction?

yes, added

170: who is responsible for the product?

corrected to S5P-BASCOE

177: what version 2.1 algorithm means?

The algorithm is similar to TOMS v8, more details are described by McPeters et al 2019. we added this information

191: Reference for sonde comparisons?

Elshorbany, Y. Y, H. C. Kapper, J. R. Ziemke, S. A. Parr, The Status of Air Quality in the United States during the COVID-19 Pandemic: A Remote Sensing Perspective, Rem. Sens., 13(3), 369, <https://doi.org/10.3390/rs13030369>. 2021. added

213: why the two step approach and not grid directly to the reference grid?

The S5P-BASCOE data are compared to two different products with different resolution and to reduce the effort of the gridding we used a smaller pixel size. Moreover, the gridded data were used for the monthly or weekly means shown in section 5. But the referee is right, 0.5° by 1° would be sufficient for our comparison approach

217: S5P ATBD says 270hPa?

corrected see comment l 155

Fig4: Latitudes missing.

updated

220: looks like there is a land-sea contrast as positive bias is mostly over land

There is indeed a larger positive bias over northern Africa but there is also a positive bias over the Pacific Ocean and negative bias over South America. If there was a land-sea contrast it would be most obvious in the long term mean. The figure below shows the mean deviation S5P-BASCOE - S5P_CCD for the complete comparison period (April 2018 to October 2020). We still see a positive bias over the Saharan desert, but also over the Pacific Ocean. On the other hand, negative deviations show up over the Indian Ocean and the adjacent landmasses in Indonesia, but also over Africa.

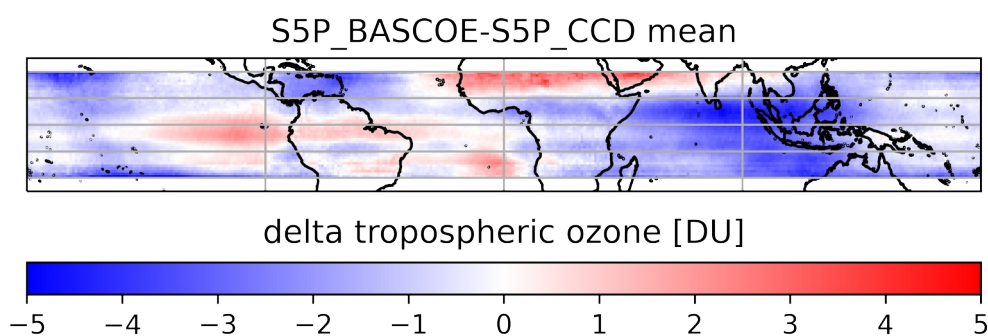


Figure 1: Mean difference between S5P-Bascoe and the S5P_CCD data for the complete comparison period

221: where is it documented?
S5P Validation reports, added

224: negative bias and variability of about 4 DU. => bias = 4DU?
Clarified exact numbers for bias and deviation are given -0.91 ± 5.76 relative to CCD

224: three peaks are found = and that means?
Clarified,

The time series of the tropical averaged difference between S5P-BASCOE and S5P_CCD is illustrated in Figure 6. The figure also includes a comparison to the OMPS-MERRA-2 tropospheric column which will be discussed in more detail in section 3.2.2. The daily averaged tropical differences to the CCD also show a negative bias -0.91 DU and a standard variation of 5.76 DU. The time series shows a high variability of about 10 DU peak to peak. Three smaller peaks are observed in June / July 2018 and large one in May / June 2019. Some of the peaks also occur in the comparison with OMPS-MERRA-2. The large peak in May / June 2019 results from a decrease in the S5P_CCD data in this period, the cause is not yet fully understood. Also for the first peak 10th June 2018 a deviation in the CCD data (not shown in the paper) contributes to the increase in the differences. For the next two peaks it seems to be an overestimation of the tropical ozone by S5P-BASCOE.

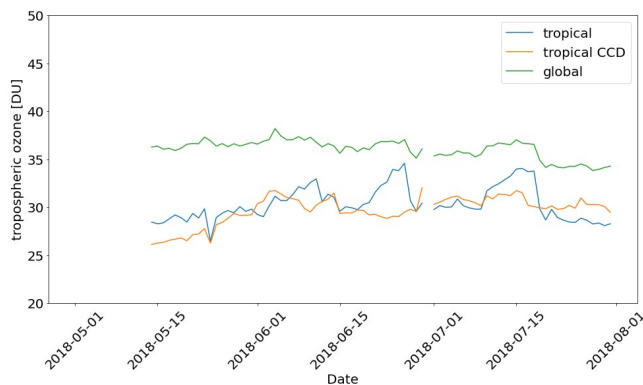


Figure 2: Absolute values of tropospheric ozone columns from CCD (orange) and S5P-BASCOE (blue for the tropics) for 2018-05-15 until 2018-08-01

Fig5: checked how subset of OMPS-M2 in tropics compared?
included, shows a few small peaks and no annual cycle
annual pattern vs CCD: are there explanations for it?

I am not sure this is really the case, in the absolute values the annual cycle agrees well between S5P-BASCOE, OMPS-MERRA2 and CCD, however the amplitude is in the range of 2 DU this might not be obvious in the absolute values. According to validation report of the S5P_CCD there are indications of an annual cycle in the deviation relative to some sounding stations and to other CCD data from OMI and GOME-2B.

Daily values? Global mean?

Yes, the global / tropical daily mean values are shown in the figure, clarified in the caption.

"Time series of the daily mean differences between S5P-BASCOE and S5P_CCD (green, tropical) and OMPS-MERRA-2 (blue, global or orange, tropical)"

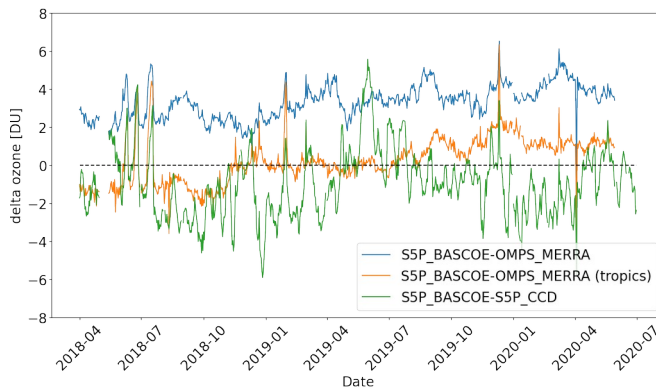


Figure 3: Time series of the daily mean differences between S5P-BASCOE and S5P_CCD (green, tropical) and OMPS-MERRA-2 (blue, global or orange, tropical)

235: mean difference 3-4DU: why no exact value?
changed: 3.34 ± 7.64 DU for OMPS

244: 3.5PVU means BASCOE will have higher columns, right?
Yes, if the tropopause is set to 3.5 PVU instead of 2.5 PVU this means the tropopause is higher and hence the tropospheric column is higher, added to the manuscript

246: which year
August 2019 (see section 2.3) added

Fig6: figures too small, hard to read
Changed to a vertical arrangement - increase in size

Fig6: difference in North Atlantic can be up to 15DU.
Large BASCOE trop. column mostly over oceans, any explanations?
According to figure 8 (updated, tropopause pressure difference and related ozone difference for May 2020) the difference due to the tropopause pressure might reach up to 5 DU. The total column might also contribute a few DU (~2). This explains already half of the observed differences. Also cloud filtering might have an influence, especially as the cloud filter might favour high ozone columns over the Atlantic Ocean.

Fig7: Date? difference 3.5-2.5?
The difference $3.5 - 2.5 = 1$
Do you mean whether the figure contains the difference in the O_3 subcolumns upto $P(3.5 \text{ PVU})$ and $P(2.5 \text{ PVU})$?

Yes, it contains $P_{\text{tropopause}}(3.5 \text{ PVU}) - P_{\text{tropopause}}(2.5 \text{ PVU})$ and the corresponding ozone subcolumns

$$\int_{P_{\text{Surface}}}^{P(3.5 \text{ PVU})} O_3(p) dp - \int_{P_{\text{Surface}}}^{P(2.5 \text{ PVU})} O_3(p) dp \text{ respectively}$$

Following the recommendation of reviewer #2 the date was changed to monthly mean for May 2020, as in figure 6

252: that means the 3.5 PVU is too high and adds a lot of stratospheric ozone into the BASCOE tropospheric column?
No, it just means that different definitions of the tropopause, cause different tropospheric ozone columns.

On the other hand, the difference between S5P-BASCOE and OMPS-MERRA-2 can not be fully explained by the difference in the tropopause. Parts of the difference is also caused by difference in the total column (here 1-2 DU is less than 1%) or by other difference in the stratospheric columns (beside the tropopause) - updated

258: which tropopause definition here? what does mean pressure mean?

(clarified) "The tropopause pressure (3.5 PVU) is read from the collocated S5P-BASCOE files and the mean tropopause pressure is used as upper limit for the sonde integration."

264: where is the sonde station? I wouldn't call 25DU "slight".

The sonde station is at Hohenpeißenberg as mentioned in the figure caption. clarified in the text. The word "slight" is removed. The respective sentence concerns total columns, where 25DU is about 6% which is within the acceptable range for outliers in the total columns.

270: Reference?

No, W. Steinbrecht explained it as chat-comment to my presentation.

Fig8: which satellite sample a 100 km circle? S5P has a higher resolution.

The reviewer is right the resolution of S5P is much higher. On the other hand parts of the area around the sounding stations might be hidden by clouds, and the data are affected by statistical noise. We averaged over a respective area to reduce the influence of noise and clouds around the sounding stations.

We study the influence of the radius in more detail, depending on the radius the number of comparisons is reduced, and the global mean deviation changes slightly. Based on this finding we reduced the radius to 25 km.

The table shows the dependency of the global mean deviation and the number of comparisons on the radius around the sounding stations.

radius	100 km	50 km	25 km
number of comparisons	3112	2687	2254
mean difference [%]	15.03	14.99	12.81
standard deviation [%]	30.18	30.26	29.11
mean difference [DU]	3.33	3.15	2.59
standard deviation [DU]	9.53	9.50	9.32

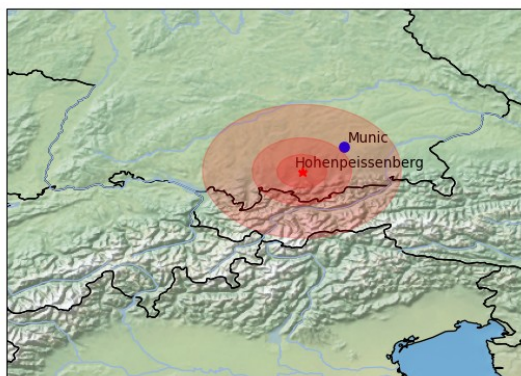


Figure 4 same as figure 8 (old) in the manuscript with the different averaging radii mentioned in the table

271: how and why is the sonde data scaled?

The scaling of the sonde data is performed by the data providers.

First they integrate the sonde data to the total column and add a climatology based correction for the difference between burst altitude and top of atmosphere to get the total column. The total column is scaled to agree with spectrometer's observation. The same scale factor is applied to the mixing ratio at each altitude. According to Logan et al. (2012) the scaling is not applied to all sonde data in Europe and the scaling factor varied between 0.8 and 1.2.

Clarified:

"At some sonde stations the data providers integrate the data up to the top of atmosphere, assuming a climatology above the burst altitude, and compare it with nearby total column observations e.g. from Dobson spectrometers. The measured mixing ratios are scaled according to the ratio of the total columns. This scaling is quite common though not used in general (e.g. Logan et al., 12). It helps harmonizing the data for long term time series it also corrects for short term variations and artificial trends. The scaling factors vary between 0.8 and 1.2.

However, if the ozone effective temperature is not considered in the Dobson spectrometer data retrieval, this might result in a smaller ozone column, especially in the winter month. In this case also the scaled sonde data is underestimated."

273: how large are those biases? ~8DU is not that small. relative differences are used in S5P to describe biases. what is the overall bias/dispersion? is there a time dependency?

If the observable is close to zero the relative error, might be misleading, or not defined. Therefore for some trace gases e.g. SO₂ the absolute deviation might be a better choice. I am not sure if this is really the case for tropospheric ozone, here I think both the relative and the absolute deviations might be justified. With respect to the comment we partly changed to relative values for the sonde based comparison. Figure 10 (time series at Hohenpeißenberg) still uses absolute difference for both tropospheric and total ozone column, Figure 11 (box-whisker-plot) shows percental deviation. The table lists both, absolute and relative mean deviation per latitude.

Up to now we focused on roughly two years of data, this period is from my point of view not long enough to retrieve solid statistics about time dependencies, especially with respect to the sparse sampling for some of the sounding locations. For Hohenpeißenberg we see an overestimation in the winter period relative to the sondes.

278: end of sentence is missing.

Corrected:

"A small positive bias (~5 DU) relative to the sonde data is found for tropical to mid-latitudes, details are shown in table 1. The global mean deviation equals 2.8 DU or 13.5 % with a standard deviation of 9.4 DU or 29.9 % for the 2191 comparisons used in this study."

285: typical enhancements are e.g. Mediterranean: rephrase. why are they typical?

Typical in that sense, that they are observed regularly (each year) in the specific region and have been studied for many years now, seen comment by Owen Cooper for the enhancement over the US or Lelieveld et al. 2002 for the eastern Mediterranean.

clarified:

"Figure 12 shows the global tropospheric mean ozone distribution for the four seasons. All data from March, April and May and the years 2018 to 2020 were average for the first subplot, and respectively for the other subplots. During the northern hemispheric spring the tropospheric ozone column is enhanced over the northern oceans. During the northern hemispheric summer three major enhancements can be seen: South Eastern US (section, 4.4), Eastern Mediterranean (section 4.3) and North Eastern China (not discussed here)."

290: columns correlated with other parameter e.g. cloud height?

Besides transport other explanations like cloud structures might be possible as well, compare also to the difference to OMPS-M2 over the Atlantic Ocean.

Clarified

"Also cloud properties influence the observed tropospheric ozone pattern, as already discussed in section 3.2.2."

Fig12: same map borders would be good
updated, See also answer to referee #2

302: how much can be attributed to the positive bias?

The bias is not known everywhere and at any time. According to the update table (suggested by the reviewer) for this latitude band (30°-40°N) the mean bias is about 6.3% or 1.2 DU.

The extreme values of 80 DU are not the summer mean maximum, which is just above 60 DU. The respective bias can hence be estimated to less than 5 DU. We will leave this estimate to reader.

304: The Berlin 20d mean has a lower dispersion than Athens from my point of view.

clarified: the seasonal amplitude in Athens is larger as in Berlin, the short-term day to day variations however are higher in Berlin. See also comment to referee #2.

Fig14: Monthly mean?

yes, the figure for the US shows monthly mean values. clarified

320: small bias (~2-4DU = 5-10%, max~15DU)? Total columns or tropopause definition?

There are several different sources for deviations between the two data sets, the total column and the tropopause height are the most obvious ones. For S5P-BASCOE a cloud filter is applied, the data are gridded and interpolated between different resolutions for the final product as well as within the retrieval, this might also cause part of the observed structures.

Clarified in chapter "3.2.2 Comparison to OMPS-MERRA-2"

CCD similar bias ~4DU

323: what does variability in difference means?

this sentence refers to figure 5 (time series of S5P-BASCOE compared to CCD and OMPS-MERRA2). While the difference to OMPS-MERRA-2 changes only slightly in time the difference to the CCD data shows a much higher variability as a function of time.

325-327: doubling of sentences. slight bias ~5DU (fig10)?

- Conclusions should be carefully revised

updated

GENERAL ISSUES

Errors: quite a few typos, misspellings. Text needs to be heavily redacted.

units = sometimes no space between number and unit

corrected

nouns = capital first letter e.g. Tropopause?

deg or ° = unify

corrected

reference = needs to be checked thoroughly (e.g. German verbs not shown correct, spacing at wrong places)

corrected (French, German and Spanish special characters)

ERRORS (only subset, thorough inspection necessary):

5: extend these data record into the future = ?

deleted.

8: ozone sonde data = sub columns?

corrected, "integrated sonde data"

20: IPCC, 2013(@)

corrected

72: next section = number?

added

77: The second step includes = rephrase, hard to understand

changed: "In the second step the ozone profiles from BASCOE (section 2.3) are integrated between the tropopause pressure and the top of the atmosphere."

80: place = space?

changed

88: (Clouds as Layers Loyola et al., 2018).

removed "Clouds as Layers"

104: the changes * 3, rephrase

changed: "Since it is an operational service, the BASCOE-FD system has evolved with time due to the changes in the ECMWF operational system. Moreover BASCOE-FD was adapted to the updates in MLS retrieval algorithm and those in the BASCOE system (see the changelog here: http://www.copernicus-stratosphere.eu/4_NRT_products/3_Models_changelogs/BASCOE.php, Dec. 2021).

145: OFL -> OFFL

changed, also NRT to NRTI

166: to the 380 K level 80 to 130 hPa for

clarified

225: 2* periods

deleted one of them

228: Tropospheric ozone columns from OMPS-MERRA-2 are described = misleading, retrieval is described

clarified

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J., Scheeren, H. A., Sciare, J., Schlager, H., Schultz, M., Siegmund, P., Steil, B., Stephanou, E. G., Stier, P., Traub, M., Warneke, C., Williams, J., and Ziereis, H.: Global air pollution crossroads over the Mediterranean, *Science*, 298, 794–799, 2002.

Logan, J. A., Staehelin, J., Megretskaia, I. A., Cammas, J.-P., Thouret, V., Claude, H., De Backer, H., Steinbacher, M., Scheel, H.-E., Stübi, R., Fröhlich, M., Derwent, R., Changes in ozone over Europe: Analysis of ozone measurements from sondes, regular aircraft (MOZAIC) and alpine surface sites, *J. Geophys. Res.*, 117, D09301, doi:10.1029/2011JD016952, 2012.

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