# Author's Response

Here I've listed the referee's notes and major concerns/comments, followed by my own response in blue. Minor comments have been handled and I'm thankful to the referee's for going through my manuscript with such attention, as it improves my work greatly.

First the referee responses are listed then the major comments and my responses for each referee are shown.

# Referee Notes:

### Anonymous Referee #1

Received and published: 6 February 2017

The current study presents a method to identify stratosphere-to-troposphere transport (STT) events and estimate the associated ozone flux to the troposphere, based on ozonesonde profiles from three sites located in the Southern Hemisphere extratropics. Subsequently, the seasonality of STT events is determined, as well as the favorable synoptic conditions. Based on the stratospheric contribution to tropospheric ozone column estimated from the ozonesondes, the GEOS-Chem simulated tropospheric ozone columns are extrapolated to assess the stratospheric contribution over the Southern Ocean region. As the STT is of great importance for the tropospheric ozone budget and variability, and the number of relevant studies (both observational and modeling) for the examined region is limited, I find the topic of the paper within the scope of ACP. On the other side, there are several issues that need to be addressed before consideration for publication in ACP.

### Anonymous Referee #2

The authors present an observation-based method to estimate the total stratospheric ozone flux in the Southern Ocean. I think the approach is interesting and complement some model-based methods, and is also of interest to the readership of ACP. However, the method comes with some major uncertainties and I wonder whether an extrapolation to the whole Southern Ocean from only three measurement sites is reasonable. My major concerns are listed below, and based on them I only recommend the manuscript ready for publication in ACP if a carefully revised manuscript is provided.

### Anonymous Referee #3

Received and published: 23 February 2017

The paper by Greenslade and coauthors presents an analysis of ozone soundings from three locations between -31 ◦ S and -69 ◦ S over several years. The authors analyse the profiles for ozone enhancements in the troposphere, which they link to stratosphere-to-troposhpere exchange associated with cut-off lows and cyclones. Based on these enhancements they estimate the fraction of excess ozone from the stratosphere as percent of the tropospheric ozone column. They calculate a flux of ozone for the southern hemisphere by using tropospheric columns from GEOS-Chem times this fraction times the frequency of occurrence of STT events. This flux estimate is more than an order of magnitude lower than estimates from literature using various techniques. The authors conclude that these differences are due to conservative estimates of several thresholds and assumption regarding their method.

Observationally based estimates of ozone transport especially in the southern hemisphere are sparse and therefore valuable. However, the authors provide a flux estimate which is far from other studies, probably due to the sparse spatial and temporal coverage. If this however is the case, the method is simply not applicable in this case and the deduced flux does not mean anything. If the method is valid for the given data set I miss a careful analysis of the reasons for the discrepancy. Therefore I don’t see the paper as an ACP paper in the current form.

# Anonymous Referee 1

### Major Comments:

1) Calculating the 99th percentile from the perturbation profiles over that layer (2 to

1 Km below the tropopause) is a fairly strict criterion. Wouldn’t this threshold choice

avoid the selection of deeper stratospheric intrusion events as “STT events”?

This should read as 2 km above the surface to 1 Km below the tropopause since, as you point out, the one kilometre range would miss deeper intrusions. I've now changed the text to reflect this.

Have you consider modifying this criterion, and include others (e.g. significant negative O3 relative humidity correlation values above a threshold) to minimize false STT detection?

2) The seasonality of STT events presented in Fig. 7 is not in line with the findings of

Škerlak et al. (2015) for the examined region. How are your results (STT seasonality)

compared with other modeling studies (Elbern et al., 1998; Sprenger et al., 2003)?

I have added comparisons between the seasonalities at my three sites and results from Wauben et al., 1998: Comparison of modeled ozone distributions with sonde and satellite observations, and Sprenger et al., 2003.

The measurement sites are not in the regions which have a clear winter maximum seen in figure 1 of Sprenger et al., 2003, and the large scale winter maximum shown by all three studies seems to be dominated by the system in that region – this has now been noted in the text.

Is there any evidence from other studies that STT frequency over the examined region

exhibits a maximum during the austral summer (DJF) and not during the austral winter

(JJA) when the jet stream is strongest over the broader region?

I have not found any other literature which suggests a Summer peak over the Southern Ocean or Australia, however the prior work has not looked at the sites examined here (for instance see my previous response).

Have you tried to detect STT events from the model results? I guess this is strongly depended to the vertical resolution of the model, but it would be very interesting to see how the observed and

modeled STT seasonalities are compared.

Not only the vertical resolution but also the low horizontal resolution of GEOS-Chem output means an analysis would only perceive very large scale STT events, as they would need to span a large portion of 2 degrees latitude by 2.5 degrees longitude.

3) To my understanding, using the seasonality of STT events from the three sites to

extrapolate model results over the Southern Ocean region is a quite simplified and

coarse approach, especially when considering the previous comment.

This is correct, after seeing the reviews the SO extrapolation has been moved to a supplementary pdf as an example of one possible utility of the ozonesonde event detection. It is too simplified and uncertain to add any real substance to the paper.

TODO: Instead an estimate of STT ozone flux near the three sites has been performed and compared to Skerlak et al. 2014, Sprenger et al. 2003, and Olsen et al. .

4) Overall, the presentation of the results can be further improved (please check my

suggestions further below), as well as the writing of the manuscript.

### Comments:

Škerlak et al. (2014) presented an STE climatology using the ERA-Interim data. This

study is important not only for the introduction, as it describes the STT climatology

for the SH, but for intercomparison of the results also. Similar climatologies can be

found in the modeling studies of Roelofs and Lelieveld (1997) and James et al. (2003).

Recently, Akritidis et al. (2016) explored the impact of stratospheric intrusions on tropospheric ozone and the associated stratospheric contribution over the eastern Mediterranean and the Middle East region, a task that is relevant with some of the purposes of this study.

Page 4, lines 3-4: Since the study is based on the ozonesondes launched from the three sites, it is important to present the location of the sites.

A very good idea, a brief description of the sites has been added: “... Melbourne, a major city in Victoria, Australia is in the far south eastern section of the Australian mainland, actual releases are north of the central business district in the Broadmeadows suburb. Macquarie island is isolated from the Australian mainland, situated in the remote southern ocean and unlikely to be affected by any local pollution events. Davis is on the coast of Antarctica and also unlikely to experience the effects of anthropogenic pollution.”

Page 4, line 22: “Figure 1 shows the monthly mean tropopause altitudes at ..”, while in Fig. 1 caption is stated “Multi-year monthly median tropopause altitude ..”. Is it the mean or the median? Please modify accordingly.

It is the median, text updated

Page 5, Figure 1: a) The shadings used to describe the 10th and 90th percentiles are

rather confusing. I suggest you replace the shadings with dashed lines (same color as

the solid lines). b) Increase the range of the vertical axis to show the 10th percentile

value for February. c) Is it the case that tropopause drops below 4 km (10th percentile)

over Davis? What is the minimum tropopause height value over Davis during February?

Page 5, lines 5-6: “This seasonality at the high latitude sites is driven by a decrease in

photochemical destruction under the reduced radiation conditions around polar night.”

Please include a reference or information about the NOx levels at these sites (if avail-

able) to justify this statement.

Page 6, line 14: It is important to know the vertical resolution of the GEOS-Chem model

near the tropopause (although it can partially be seen from Fig. 13), as it is important

for the tropopause height detection and the tropospheric ozone column calculations

from the model results.

Page 7, lines 22-23: “The interpolated profiles … high frequency perturbations).” This is a rather brief description of the procedure. A more detailed description including a reference (if available) for the FT application would be necessary.

Page 7, lines 27-28: “We next use all the perturbation profiles at each site to calculate the 99th percentile perturbation value for the site”. How exactly is this cut-off threshold calculated? In Section 2.5, Page 9, the authors state that is calculated “between 2 km and 1 km below the tropopause”. This information should be provided earlier in the manuscript, at the point that the 99th percentile threshold is initially mentioned (Section 2.3).

Page 8, Figure 3: Why the two panels have different units? Are the ozone units of

the left panel “1e+12 molecules cm-3”? Please change accordingly the Figure and the

Figure caption. mixing ratio -> number density

Page 9, lines 1-2: “For this reason all detected STT events found near smoke plumes

are flagged”. How is “near” defined?

In my opinion, Figures 4, 5 and 6 are more supportive-descriptive without adding any-

thing new. Therefore, I suggest including them as a supplement. Moreover, Figures 5

and 6 can be merged into one.

Page 11, line 17: “We use the ERA-I 500 hPa data to subjectively classify the events

based on their likely meteorological cause.” Do the authors classify the events by visual

inspection of the 500 hPa maps for every STT event date?

Page 11, lines 20-21: “The stratospheric polar vortex may create ozone folds without

other sources of upper tropospheric turbulence”. Please include a reference for the

above statement.

Page 14, lines 16-20: “The seasonal distributions . . . first half of the year”. To my

understanding Fig. 7 and Fig. 8 are not quite similar. Moreover, comparing Fig. 8

with Fig. 7 where fire influences are also included is somehow unfair. The fact that

ozonesondes are launched monthly at Davis from December to June is also the case

for Fig. 7, where high STT frequencies are found for the respective period.

Page 16: How is the modeled tropospheric column ozone calculated? How is the

tropopause defined in the GEOS-Chem results?

That is a good question! GEOS-Chem uses the tropopause height provided by GEOS-5 met fields which are calculated using a lapse-rate definition using the first minimum above the surface in the function 0.03\*T(p) – log(p), with p in hPa.

This has now been added to the text

Page 17, lines 3-4: “Over Melbourne, ozone in the lower troposphere is well represented, but the model overestimates ozone from around 4 km to the tropopause”. This

is also seen for Macquarie and should be added to the discussion.

Page 19: “Figure 14 shows the mean fraction of total tropospheric column ozone (cal-

culated from ozonesonde profiles) attributed to stratospheric ozone intrusions at each

site, averaged over days when an STT event occurred.” Please explain in more detail

how is this fraction calculated.

Page 19: “to the entire Southern Ocean region, defined here as 35\_ S-75\_ S to en-

compass”. What is the longitudinal range?

Page 20: Fig. 14 and Fig.15 can be merged into one.

Page 22: “If we we assume a fractional ozone impact due to each event STT event of

I=35% based on their results”. The 30-40% stratospheric contribution found by Terao

et al. (2008) is seen only during spring and at 500 hPa. Therefore, assuming a 35%

stratospheric contribution to the tropospheric column ozone seems a bit arbitrary.

# Anonymous Referee 2

### Major Concerns:

1. **Extrapolation to Southern Ocean**: The authors look at three measurement sites (Davis,

Macqaurie, and Melbourne) in the Southern Ocean (SO), and then extrapolate their results to the

whole SO. I don’t think that this is valid. I think there ia quite a lot of spatial and temporal

variability that gets neglected in doing so. To make my point more clearly, I copy a figure (Fig. 16)

from Skerlak et al. (2014) here:

It shows the seasonally averaged STT ozone flux for the period 1979-2011. Evidently, there is a lot

of spatial and temporal variability. The next figure (Fig. 17) from Skerlak et al. (2014) shows the

estimated ozone flux into the PBL, which exhibits a still stronger variability. Hence, I think the

authors must be rather hesitating in extrapolating their results. I suggest to restrict the conclusionsabout the STT flux more to the regions around the three measurement sites. It will still be possible

to compare the values, e.g., with the values in Skerlak et al. (2014).

2. **Transport aspect**: An aspect that is not sufficiently discussed in the manuscript is the transport

of the ozone-rich air from its crossing to the measurement site. For instance, in Figure 5 the authors

show an STT event and the geopotential height field at 500 hPa. A nice cut-off low pressure system

is discernible in the geopotential. But it is not clear whether the STT event really occurred below

this cut-off. In fact, it could have happened quite a distance away from it and the be advected to this

place. I would argue that the transport aspect become more important if an STT event is detected at

middle or lower-tropospheric levels, i.e., when it is rather ‘detached’ from the tropopause above. As

an example, the following study shows that the crossing of the tropopause takes place in the western

North Atlantic but an ozone signal is discernible in the profile over western Europe:

Trickl, T. et al. "How stratospheric are deep stratospheric intrusions? LUAMI 2008." Atmospheric

Chemistry and Physics 16.14 (2016): 8791-8815.

I think the authors should more carefully discuss this aspect of STT event. Possibly, the do a short

literature review dealing with ozone transport and the long-range character of stratospheric

intrusions. It would also be interesting, and relevant to this manuscript, how long signals in

stratospheric ozone remain discernible in an atmospheric column after the air parcels have crossed

the tropopause.

3. **Uncertainty**: The method comes with quite a few uncertainties! I list some of them:

- P7,L30: “STT events at altitudes below 4 km are removed to avoid surface pollution, and

events within 0.5 km of the tropopause are removed to avoid false positives induced by the

sharp transition to stratospheric air.” → I see the problem with the near-surface STT events.

But still, even at this low altitude it could be due to a stratospheric intrusion.

This is one possible false negative which could occur, I have added a note at PX “...TODO...”.

Further, I expect quite some ozone flux to be across the tropopause without a very clear peak-like

structure in the profile. This could, e.g., be the case if the ozone flux is more related to a

continuous ‘diffusion’ of ozone across the tropopause in contrast to an ozone flux going

along with a coherent cross-tropopause air streams in distinct weather systems.

While this is true, the focus of this work is on discrete influx events, or folds, rather than continuous enhancement or reduction due to cross tropopause transport. This is due to the difficulty of detecting continuous influx with weekly ozonesondes. I do appreciate the detailed comments and have noted that we are likely ignoring this source of ozone enhancement at PX “...TODO...”.

- P7,L9-12: “This estimate is conservative because it does not take into account any ozone

enhancements outside of the detected peak that may have been caused by the STT, and also

ignores any enhanced ozone background amounts from synoptic-scale stratospheric mixing

into the troposphere.” → The ozone background is also enhanced in mixing across the

troposphere, or the background at any of the stations is enhanced by STT events taking place

outside its ‘range’.

This is true, thanks for pointing it out. I've added this possibility to the text: “... TODO...”.

- In section 5 (P19,L9) the overall ozone flux is determined as the product of the monthly

likelihoods of STT (f), the monthly mean fraction of an ozone column attributed to

stratospheric ozone (I) and the mean tropospheric ozone column (Omega). All these factors

come with a lot of uncertainty! Be it due to the method applied, or the spatial and temporal

variability.

- P9,L16: “While ozone production occurs in some biomass burning plumes, this is not

always the case; therefore ozone perturbations detected during transported smoke events

may or may not be caused by the plume. For this reason all detected STT events found near

smoke plumes are flagged.” → These events are not included in the calculation of the ozone

flux, but still they could be of relevance!- P9,L7-9: “ We use the 99th percentile because at this point the filter locates clear events

with no obvious false positives. Event detection is highly sensitive to this choice; for

example, using the 98.5th percentile instead increased detected events by 10 (22%) at Davis,

19 (40%) at Macquarie Island, and 24 (33%) at Melbourne.” → Does this mean that with a

98.5 th percentile, some of the events are clear false positives? Wo do you decide that this is

the case? I am not sure whether this is obvious. In short, an additional uncertainty of the

method.

Given all these uncertainties, the estimate of the total STT flux based on the ozone profiles must be

rather conservative and going along with a big overall uncertainty! This is already discussed by the

authors, i.e., they are fully aware of it. What I would, however, suggest is a separate section (or

extended paragraph) where all uncertainties are presented and, if possible, quantified.

# Anonymous Referee 3

### Major comments:

1) Overall the manuscript leaves me a bit puzzled, since I’m not sure what to take out of this work. The authors state that their value of the ozone enhancement fraction might be largely underestimated by a factor of ten. If this is the case it is difficult to get the benefit of the study. Though the approach is reasonable, maybe the statistics and spatial coverage is to small to cover the full variability and frequency of occurrence of STT events for a quantitative flux calculation for the southern hemisphere. If the difference between observations and models is really between factors 30-200 depending on the reference (p.21, l.8-12), this needs more clarification than simply replacing observation with model results to find agreement with other studies. I found this approach at least very questionable.

2) Further: I missed a quantification of the uncertainties. This is partly done in section 2.5, but it is e.g. not clear why a threshold of 99% is the best choice nor which factor specifically leads to the very low flux estimate. I suggest the authors use a bunch of northern hemispheric sondes with higher spatial and temporal density to gauge their approach before applying it to the southern hemisphere.