Review for

Stratospheric ozone intrusion events and their impacts on tropospheric ozone

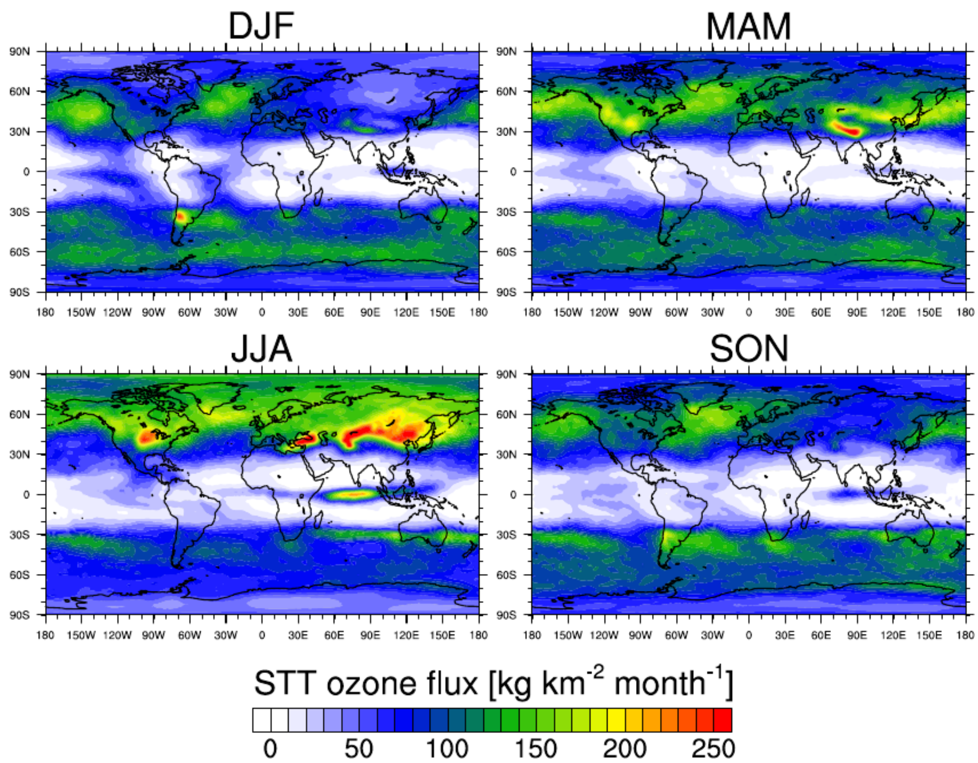
by Greenslade et al.

Synopsis:

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.

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 conclusions about 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. 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.

- 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'.

- 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.

Minor Comments:

- P2,L13-15: "While models show decreasing tropospheric ozone due to stratospheric ozone depletion propagated to the upper troposphere through vertical mixing (Stevenson et al., 2013), recent work based on the Southern Hemisphere ADditional OZonesonde (SHADOZ) network suggests increasing upper tropospheric ozone near southern Africa, most likely due to stratospheric mixing (Liu et al., 2015; Thompson et al., 2014)" → Simplify sentence structure! Rephrase.

- P2,L24: " excedes" → "exceeds"

- P2,L29: "STT is responsible" → "STT to be responsible"

- P3, L4: "mixing across the tropopause mainly caused by the jet streams" → a little strange formulation. Mixing is not caused by the jet streams; maybe you can write that it is associated by the jet streams.

- P3,L10-11: "A big influence on the high surface ozone concentrations over the eastern Mediterranean is stratospheric mixing and anticyclonic subsidence (Zanis et al., 2014)" → "A big influence on the high surface ozone concentrations over the eastern Mediterranean can be attributed stratospheric mixing and anticyclonic subsidence (Zanis et al., 2014)"

- P3,L11-12: The authors might want to consider the following studies dealing with STT and ozone fluxes over the eastern Mediterranean:

Tyrlis, E., B. Škerlak, M. Sprenger, H. Wernli, G. Zittis, and J. Lelieveld (2014), On the linkage between the Asian summer monsoon and tropopause fold activity over the eastern Mediterranean and the Middle East, J. Geophys. Res. Atmos., 119, 3202–3221, doi:10.1002/2013JD021113.

Akritidis, D. et al. "On the role of tropopause folds in summertime tropospheric ozone over the eastern Mediterranean and the Middle East." Atmospheric Chemistry and Physics 16.21 (2016): 14025-14039.

- P3,L14-15: "The strength (ozone enhancement above background levels), horizontal scale, vertical depth, and longevity of these intruding ozone tongues vary with weather, topography, and season." → This is a rather general statement. What do you mean with weather?

- P3,L30-33: How relevant is it for the reader to know how the ozone mixing ratio is quantified? If not relevant, I would remove this sentence. It sounds rather technical!

- P4,L8-9: "Characterisation of STT events requires a clear definition of the tropopause. The two most common tropopause height definitions are the standard lapse rate tropopause (WMO, 1957) and the ozone tropopause (Bethan et al., 1996)." → I would mention already at this place the dynamical tropopause which is defined by means of a potential vorticity iso-surface. I would guess it to be rather similar to the ozone tropopause, but to differ from the WMO one.

- P4,L17: "The ozone tropopause can be less robust during stratosphere-troposphere exchange;" → What does 'robust' mean? What defines whether a tropopause is robust or not? The ozone tropopause certainly allows for much more details (and a much more complicated structure) than the WMO tropopause. But I would not say that it is less robust because of this!

- P4,L19-21: "In this work, the lower of these two tropopause altitudes is used. This choice avoids occasional unrealistically high tropopause heights due to perturbed ozone or temperature measurements in the ozonesonde data." → I feel a little uncomfortable by this definition! The two definitions of the tropopause are rather different, and by simply taking the lower one seems 'dangerous'. The authors should motivate this approach more clearly. At least, I would like to know how often the ozone tropopause 'wins' and how often the WMO one. I would expect the ozone tropopause most often to be at lower heights than the WMO one! Correct?

- P7,L22-23: "The interpolated profiles are then bandpass-filtered using a Fourier transform to retain perturbations with vertical scales between 0.5 km and 5 km (removing low and high frequency perturbations)" → I see the 0.5-km threshold. What exactly is the aim of the low-pass filtering threshold (5 km). A more clear description would be helpful.

- P7,33-34: "The STT event is confirmed if the perturbation profile drops below zero between the ozone peak and the tropopause" → Why does have to drop below zero?

- P8,Figure 3: Just for curiosity: In the ozone profile the Ozone mixing ratio (OMR) is rather low right above the identified STT event. The OMR is higher than immediately below the STT event. Is their a simple reason why the OMR is so low right above the STT peak?

- **P9,L16:** "all detected STT events found near smoke plumes are flagged." → What does 'near' mean?

- P 10,L15-16: "Data from the European Centre for Medium-range Weather Forecasts (ECMWF) Interim Reanalysis (ERA-I) (Dee et al., 2011) product are used for synoptic-scale examination of weather patterns over our three sites on dates matching detected STT events" → Please rephrase! For instance: "Synoptic-scale weather patterns are examined based on the ERA-Interim dataset (Dee et al., 2011). More specifically, the ERA-I products over the three sites are used on dates matching detected STT events.

- P11,L17+26: Here, the STT event is subjectively linked to a meteorological feature, a cut-off low-pressure system. The argument is not very 'strong'. I don't think that a lowering of the tropopause itself can explain the flux of stratospheric ozone. It would be interesting to see a vertical cross section the cut-off low, with tropopause height included. Is the cut-off low eroded away from below, or how does the flux across the tropopause in the cut-off low really takes place? Some further thoughts on this might be helpful. The following paper might be a starting point:

Stohl, A., et al. "Stratosphere‐troposphere exchange: A review, and what we have learned from STACCATO." Journal of Geophysical Research: Atmospheres 108.D12 (2003).

- P12, Figure 6: In this Figure 6 relative humidity is shown in addition to ozone. I wonder whether this signal is available for all soundings, if if so, whether it would be worthwhile to include it into the identification method of STT events. Of course, this would be based on the dryness of stratospheric air.

- P12,L10: “This summertime peak is due to a prevalence of summer low-pressure storms and fronts” → Please add a reference which supports this statement. Reutter et al. (2015) doesn't deal with that, as far as I see. Further, I don't see why the summer low-pressure storms and fronts increase turbulence. Here too, a reference might be appropriate.

- P12,L11-P13,L1: “At Davis, there is increased Antarctic winter activity, which may be due to the polar vortex and it’s associated lowered tropopause and increased turbulence” → Again, I think that this statement must be supported by a reference. Why is Antarctic winter activity increased (by the way, what is meant with 'activity'?)? Why is this related to the polar vortex? And, why is there enhanced turbulence in this case? As a starting point, the authors might want to look at the SH climatologies of extratropical cyclones:

Jones, David A., and Ian Simmonds. "A climatology of Southern Hemisphere extratropical cyclones." Climate Dynamics 9.3 (1993): 131-145.

Simmonds, Ian, and Kevin Keay. "Mean Southern Hemisphere extratropical cyclone behavior in the 40-year NCEP–NCAR reanalysis." Journal of Climate 13.5 (2000): 873-885.

- P13,L3-5: “This seasonality is not seen in the recent ERA-Interim tropopause fold analysis performed by Škerlak et al. (2015), where a winter maximum over Australia can be seen (although in the subtropics only - from around 20 S to 40 S)” → This comparison is somewhat misleading, because Davis is located at 69 S, but the authors mention that the fold maximum in Skerlak et al. (2015) occur between 20-40 S. I wonder whether the authors would benefit from the following data source that provides global monthly climatologies of several features (among them tropopause folds):

Sprenger, M. et al. "Global climatologies of Eulerian and Lagrangian flow features based on ERA-Interim reanalyses." Bulletin of the American Meteorological Society 2017 (2017).

- P13, Figure 7: At Davis no STT events are associated with fronts, quite in contrast to the two neighboring months. I wonder whether this is correct!

- P14,L10-11: “and assume that the vertical temperature gradients within the intrusion respond most rapidly to transported heat, which is an additional characteristic of stratospheric air” → I do not understand this statement! Which transported heat? Why do the vertical temperature gradients respond most rapidly to it? Please explain!

-P15,L24-28: “There is no clear relationship between meteorological conditions and event altitude.” → Could this be related to the fact that the transport (and time) between the tropopause crossing and the STT event in the ozone profile is neglected? The same question can be asked whether for the link to meteorology at the the other two sites.

- P15,L29: “Simulation of southern mid-latitude ozone columns” → While reading this section I wondered what it has to do with the topic of the paper, i.e., with STT events. Of course, later (in section 5) it becomes perfectly clear. But, maybe its better to add one or two sentences that make it clear to the reader from the beginning. One or two introductory sentences to section 4 might be OK.

- P16,L12: “the r2 values decrease to .07, .11, .30 respectively” → As far as I can see these de-seasonlized values are particularly relevant. Right? But the explained variance (equal to r^2) seems to be rather low?

- P17,L1-12: At first reading I wondered whether I should worry about GEOS-Chem's inability to capture STT events. I also wondered whether the underestimation of ozone in the lower troposphere (up to 6 km) has something to do with the model's deficiency in capturing the STT events. Only in the next section 5 I learned what the GEOS-Chem's ozone columns are used for. I guess that the reading of section would have been more 'rewarding' if I had knew in advance why we are now looking at these GEOS-Chem simulations. In short, as before I would suggest to guide the reader a little more clearly and help him/her not to be mislead... Some short introductory sentences to section 4 might be sufficient to do so.

- P20,L1-7: Here some aspects of limitations of the method are discussed, or the rather strong assumptions are discussed and what could be done in a next step. As suggested in my major concerns, I would appreciate if all these discussions are concisely, but in sufficient depth, discussed at one place in the manuscript.

- P21, Figure 16: The ozone flux in the lower panel essentially has the same shape as the f part in the upper panel. Is it, hence, correct to say that the estimated ozone flux is essentially determined by this factor (the monthly frequency of STT events)? This might be relevant if a more detailed analysis of the uncertainties of the method is provided.

- P21,L7-13: Here the estimates from the three sites are extrapolated to the Southern Ocean. As stated in my major concern, this most likely is too 'strong'. I would prefer if the fluxes at the three sites are taken to estimate the STT ozone flux in the regions around them, and then compared to the fluxes from other studies (Stevenson et al., 2006; Sprenger et al., 2003). In addition to the studies listed, I would also look at the following study:

Skerlak, B., M. Sprenger, and H. Wernli. "A global climatology of stratosphere-troposphere exchange using the ERA-Interim data set from 1979 to 2011." Atmospheric Chemistry and Physics 14.2 (2014): 913.

Possibly, from these global climatologies some regional estimates of ozone flux can be extracted and compared to the fluxes at the three sites.

- P22,L5-6: “Comparison with ERA-Interim reanalysis data suggested the majority of events were caused by turbulent weather in the upper troposphere due to low pressure fronts, followed by cut-off low pressure systems” → The term 'turbulent weather' is not very meaningful! I think the link to turbulence could simply removed from the manuscript. It is not really shown in the paper, and ERA-Interim might not be the best dataset to address it due to its coarse resolution. I think just listing the weather systems would be OK, although the link to them could be more deeply discussed in the paper.