

***Interactive comment on* “The underestimated role of stratosphere-to-troposphere transport on tropospheric ozone” by Thomas Trickl et al.**

Anonymous Referee #3

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This paper begins with the surprising title, “The underestimated role of stratosphere-to-troposphere transport on tropospheric ozone” which implies that all previous estimates of the global STE budget over the past 20-30 years are far too low. If supported by solid evidence, this would be a landmark paper that would force a major reassessment of the global models that quantify the tropospheric ozone budget. This is an extraordinary claim and extraordinary claims require extraordinary evidence. This paper provides no such evidence as it merely reports measurements from a single location followed by speculation, incorrect assumptions and absolutely no convincing arguments that previous STE budgets are too low. In addition, the study’s major conclusions were discovered and reported by other well-known papers 18-19 years earlier using more extensive datasets. I firmly recommend that this paper be rejected.

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This review is framed around the following question: Which studies, specifically, have underestimated the role of stratosphere-to-troposphere transport on tropospheric ozone?

1) The global role of STE on the tropospheric ozone budget can only be estimated by models as the current observational network is far too limited to provide global coverage. There are many well-known studies in the literature on this topic. Stevenson et al. (2006) and Young et al. (2013) provide estimates from model ensembles, with Stevenson et al. estimating the STE global ozone flux to be approximately 550 Tg, with a range of 400-800 Tg, and Young et al. giving a slightly narrower range of 400-660 Tg. Recently, three new studies (Olsen et al., 2013; Yang et al., 2016; Jaeglé et al., 2017) have received a lot of attention from the modelling community because they estimate the STE ozone flux using the latest NASA global reanalyses that are available at much higher resolution than the chemistry-climate model simulations reported by Stevenson and Young. These reanalyses also have improved dynamics with excellent coupling between the stratosphere and the troposphere, and they assimilate satellite observations to produce accurate estimates of the stratospheric ozone burden. For example a recent paper by Knowland et al. (2017) demonstrates that the NASA MERRA-2 reanalysis accurately simulates deep stratospheric intrusions above North America due to its very high horizontal resolution of 50 km and 72 vertical layers. The average STE net ozone fluxes of the three budget studies mentioned above are 489 (Olsen), 493 (Jaeglé) and 448 Tg (Yang), similar to previous estimates reported by Stevenson and Young and similar to other estimates made in the 1990s. Essentially, the STE net ozone flux has been estimated to be roughly 500 Tg for the past 20 years, regardless of the methodology. What evidence do the authors have that all of these studies have underestimated the STE net ozone flux?

2) Despite the wide availability of publications that provide estimates of the STE net ozone flux, this paper only focuses on a single estimate of the ozone flux, a very old publication by Roelofs and Lelieveld (1997) that uses a relatively primitive global model

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that was developed in 1995, with very coarse resolution of 3.75 degrees. The authors seem to approve of this model because it estimates that 40% of the tropospheric ozone burden is from the stratosphere, which agrees with the authors' estimate of stratospheric ozone above southern Germany. But the estimate by Roelofs and Lelieveld is averaged over the entire globe and across all seasons and cannot be applied to any particular location. They state that the quantity of stratospheric ozone in the troposphere varies with altitude, latitude and season. The fact that this 40% value matches the value above southern Germany is merely coincidental and does not mean that the observations from a single location can provide meaningful evaluation of an average global estimate. Even if the authors could evaluate the Roelofs and Lelieveld estimate at multiple locations and across all seasons and conclude that their estimate is correct, how would this prove that previous estimates of STE are too low? Roelofs and Lelieveld's estimate of the net global flux of stratospheric ozone into the troposphere is 459 Tg per year, and their estimate of gross photochemical ozone production is 3425 Tg per year, a factor of 7.5 greater than the net flux from the stratosphere. These numbers are entirely consistent with the ozone budget estimates described above by Stevenson, Young, Olsen, Yang and Jaegle. By accepting the results of Roelofs and Lelieveld, which agree with all the other studies, the authors are essentially agreeing with the consensus view that STE contributes roughly 450-500 TG of ozone per year to the troposphere. Again: Which studies, specifically, have underestimated the role of stratosphere-to-troposphere transport on tropospheric ozone?

3) The authors highlight the fact that their lidar detects many dry layers above southern Germany especially in summer, and claim that this is a new result. But this is not a new finding. Nearly 20 years ago the late Reginald Newell at MIT published a well-known paper in Nature in 1999 (cited 126 times according to Google Scholar) with the title: "Ubiquity of quasi-horizontal layers in the troposphere". They found, using the extensive MOZAIC database of ozone and water vapor profiles that fine layers in the atmosphere are found everywhere, and that dry layers with enhanced ozone are the most common, indicating a pervasive influence from STE. A year later V. Thouret

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published an even more extensive analysis based on thousands of MOZAIC profiles around the world (half above western Europe), and here are some of their important findings:

“At northern midlatitudes we find 4 times more layers in summer than in winter, while in tropical Asia we observe a spring maximum in the occurrence of the layers. The most abundant layer type everywhere is O₃+H₂O– and corresponds to the signature of stratospheric intrusions or continental pollution.” “The most surprising feature in this study is the lack of any seasonal variations. The only seasonal cycle and regional difference observed concern the average of the number of layers per profile (strong maximum in summer at midlatitudes and in spring in tropical Asia).” “This new finding reveals that stratospheric intrusions are not negligible in summer at midlatitudes or in the tropics, as previously thought”

Therefore, the finding by Trickl et al. regarding the high frequency of ozone-rich layers in the mid-troposphere above Europe during summer is nothing new. Thouret et al. reached the same conclusion 18 years earlier using a far more extensive data set and a much more thorough and clear analysis. The excellent and ground-breaking papers by Newell et al. and Thouret et al. demonstrate that since the publication date of these papers, STE has not been underestimated above western Europe or other regions of the northern hemisphere (where MOZAIC observations were available). Their findings were not mentioned by Trickl et al.

4) HYSPLIT is a very useful Lagrangian tool for a first look at prevailing transport patterns, but for quantitative analysis such as quantifying the amount of air or ozone transported from the stratosphere to the troposphere, it is an extremely poor choice, especially when far better models such as LAGRANTO or FLEXPART are available. The authors know how to use LAGRANTO and apply it to short-term transport analysis (<= 5 days) but for some reason, they don't use this superior tool for longer time periods. Instead they simply run HYSPLIT 15-day single back trajectories that don't give any information as to when, or if, the air parcel crossed the tropopause. They also don't give

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any information as to the various source regions of the air parcel. As has been shown over and over again by the many FLEXPART studies, single back trajectories are inferior to so-called retroplumes (based on 10,000-20,000 or more trajectories), and for 15-day periods single trajectories are essentially useless. Just because a 15-day single back trajectory from a dry air layer nearly circles the globe, doesn't mean that the dry filament persisted for 15 days. The HYSPLIT analysis in this paper did nothing to further my understanding of STE processes.

5) On page 2 line 35 the authors speculate that global climate change is responsible for a doubling of stratospheric ozone at Zugspitze (increasing from 11.3 ppb to 23.4 ppb) for the 26-year time period from 1978 to 2004, but provide no analysis to back this up. They even state that this is a reasonable explanation. To the contrary, this is not a reasonable hypothesis at all. A brief check of the latest global temperature trend provided by NASA, NOAA or the EU's Copernicus shows that the global temperature over that time period increased by 0.4 degrees C. So do the authors believe that for this relatively small increase in temperature the net global STE flux increased by 100%? What would be the mechanism? A doubling of the overturning of the Brewer-Dobson circulation? Wouldn't such an enormous change in the global circulation be clearly evident in other components of the general circulation, such as a dramatic widening of the tropics? Surely such a major change in the general circulation over such a short time period would have been extensively described in the peer-reviewed literature and would be evident in the ECMWF ERA-Interim reanalysis wind fields, the same ones used by LAGRANTO. Such an analysis has already been conducted by Skerlak et al (2014) for the period 1979-2011. They find no global increase in stratosphere-to-troposphere transport. This published finding immediately disproves the speculation of the authors in the Introduction. Furthermore, an excellent paper in Nature Geoscience by Jessica Neu (NASA JPL) shows that natural fluctuations of the stratospheric circulation due to ENSO and the Quasi-biennial Oscillation (on the order of 40%) produce tropospheric ozone changes of only 2%. Perhaps there was a shift in the regional transport patterns over Europe that increased STT over 1978-2004? Based on the results of Skerlak et

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al. this doesn't seem to be the case either. But given that the authors of this paper know how to use LAGRANTO, and given that the LAGRANTO analysis of Skerlak et al. must be archived somewhere, why don't they simply take the Skerlak et al. model output and explore this possibility?

6) What is the purpose of Section 3.4 Trans-Atlantic Transport? The paper is supposed to be about the underestimated role of stratosphere-to-troposphere transport on tropospheric ozone. Yet in this section they address a completely different topic which is the transport of surface ozone from North America to Europe. This is nothing new and the peer-reviewed literature is full of studies that explore this topic, studies which are far more thorough and convincing. Here the authors use a few HYSPLIT back trajectories to speculate that ozone was transported from the US boundary layer to the free troposphere above Germany. The authors speculate that the rising trajectories could be associated with a warm conveyor belt. But they provided no analysis to confirm this transport mechanism. This isn't difficult to confirm. The authors could check archived satellite imagery. The authors could also use the RAQMS model output that is shown in Figure 14. Brad Pierce has more than 20 years of experience running model analyses for aircraft campaigns all around the world. It's easy for him to quantify the contribution of the USA or STT to ozone above Germany. Instead the authors rely on just a few single HYSPLIT back trajectories. I learned nothing new from this section, and if I want to understand how ozone is transported from the USA to Europe I will consult other studies in the literature which are for more thorough and quantitative.

7) In addition to the serious problems described above, in general the manuscript is poorly written and disorganized, and tends to jump from one idea to another in a very confusing manner.

8) Finally, Hans-Eckhart Scheel is listed as a co-author but he died nearly five years ago. While his measurements may have been included in this paper, he was not able to contribute to the writing of the paper and therefore he should not be listed as a co-author as he is unable to approve of the data interpretation or the conclusions.

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