

Interactive comment on “The governing processes and timescales of stratosphere-to-troposphere transport and its contribution to ozone in the Arctic troposphere” by Q. Liang et al.

Q. Liang et al.

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We thank the anonymous reviewer for the helpful comments.

1. Definition of O3ST and O3TR, O3 chemistry due to NOx Even if these (artificial) species allow some deep insights into the undergoing processes (e.g. very important Fig. 7), they do not sufficiently explain the impact of stratospheric NOx on the tropospheric ozone chemistry. The question that still arises is what is the impact of the stratosphere (i.e. via direct transport versus via NOx) on the seasonality of ozone in the troposphere. In section 5.2, there is some discussion on this point but the mentioned separation is not quantified (in contrast to the contribution of transport from the stratosphere that was convincingly analyzed in the section 5.1)

We agree with the reviewer in the importance of the quantification of the impact of stratospheric NO_x on the tropospheric ozone chemistry. Due to the complex NO_x-O₃ chemistry and recycling between NO_x and NO_y species, a credible analysis requires a comprehensive evaluation of O₃, NO_x, and HNO₃, PAN concentrations and their photochemistry in the Arctic troposphere which is limited by the sparse observations available. The main focus of this paper is to serve as a pre-analysis for the NASA ARCTAS mission in aiming to understand the dynamics of stratosphere-troposphere exchange and how that affects the seasonality of trace gases in the Arctic troposphere, thus quantification of this impact is beyond the scope of this paper. As a continuing study, we will use aircraft measurements obtained during ARCTAS to evaluate the O₃-NO_x chemistry in the GMI model and to conduct a more comprehensive model study to quantify the impact of stratospheric NO_x on the tropospheric ozone.

2. Definition of the DT-tracer family In Fig. 4 there are significant differences between the CFC-12S anomaly (that is black in the figure and gray in the legend) and the dynamical tracers (color lines). In particular, I would expect a better agreement in LS. A part of these differences probably results from the initialization of the dynamical tracers. Why you do not use CFC-12S above the tropopause to define the DT-family tracer? (the use of an average of 100 ppbv and of peak-to-trough amplitude is not very convincing).

We set the DT-tracer to a uniform seasonal cycle that is normalized based on the seasonal cycle of CFC-12S averaged between 70-90N at 150 hPa. This seasonal cycle differs from that of CFC-12S, particularly in the mid-latitudes, because both the amplitude and phase of the seasonal cycle of CFC-12S vary with latitude (smaller amplitude and earlier seasonal minimum in the mid-latitudes than the polar region). It is difficult to use CFC-12S alone to infer transport timescale because the dependence of the amplitude and phase of the seasonal cycle together with the rapid exchange of air between the mid- and high- latitudes complicates the interpretation of the delay in seasonal maximum. This is explained in detail in section 4.3. The use of a uniform seasonal cycle for all latitudes eliminates the interference of the latitudinal-varying seasonal cycle of

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the tracers.

3. *NO_x-driven ozone chemistry. There is a gap in explaining the impact of NO_x on the ozone cycles (Fig. 9 and section 5.2). I still do not understand how NO_x coming from the stratosphere (where in summer the NO_x-driven catalytic ozone destruction dominates, e.g. Crutzen et al, 1970, see high ozone destruction rate above 200 hPa in Fig. 9) changes its “behavior” from strong destruction in LS to production (around 300 hPa in summer) and then destruction around 700 hPa. I know that there are different regimes of NO_x with respect to P(O₃) but the paper does not clarify this point. Even if the discussion up to the section 5.2 is a very comprehensive analysis, the discussion of NO_x-related chemistry is significantly weaker (maybe an additional figure between fig. 9 and 10 explaining different regimes of NO_x with respect to P(O₃) would help).*

The reason why NO_x contributes to ozone destruction in the lower stratosphere, production in the upper troposphere, and destruction again around 700 hPa, is tied to the increasing importance of the reaction between HO₂ and NO (HO₂ + NO → NO₂ + OH) as concentrations of HO_x increase significantly from the lower stratosphere to the troposphere. In the lower stratosphere, reactions of odd oxygen with HO_x and NO_x are important loss processes. In the troposphere, HO₂ + NO → NO₂ + OH produces odd oxygen (NO₂ is a form of odd oxygen), and net production is largely determined by the relative magnitudes of HO_x and NO_x. At very low NO_x levels (<20 ppbv), HO_x reacts with O₃. This leads to net destruction of O₃. As NO_x increases, the production of ozone from HO₂ + NO → NO₂ + OH increases and exceeds destruction. Thus net production changes from negative values to positive ones. We follow the reviewer's suggestion and add a figure (figure 10) showing how P(O₃) changes with NO_x in the Arctic lower stratosphere and troposphere. In this figure, we can clearly see the regime shift from net ozone destruction to net production at NO_x about 20 ppbv in the troposphere. This is different from the impact of NO_x in the lower stratosphere, which acts monotonically to destroy ozone.

Minor points:

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1. *p 19379/par 20 ...whether they have stratospheric sources or not ... CO, CO₂, N₂O and CH₄ you mentioned have mainly tropospheric sources and no stratospheric origin (please clarify)*

We have changed this to "whether they are related to STE or not".

2. *p 19380/par 15 if slower downward transport, why short-lived species may have little dependence on STE?*

Because of the long transport time, the amplitude and phase of the seasonal cycle of short-lived species that is associated with STE diminish significantly due to chemical destruction in the troposphere. We have clarified this in the text.

3. *p 19384/par 5 what are typical lifetimes of O₃ and HCl (please give numbers for your examples)*

We have added the typical lifetimes of O₃ (lifetime from a few days to 100 days depending on altitude, latitude and season) and HCl (a few weeks) in the text.

4. *p 19385/par 10 Definition of CFC-12S. What is the lower boundary (=0 ?)*

CFC-12S is used to track aged emissions that accumulates in the atmosphere and has at some time been in contact with the stratosphere, until it is removed by photolysis in the upper atmosphere. Its lower boundary does not equal zero and changes with time, depending on the net change in its atmospheric burden due to surface emissions and photolytic destruction. The deviation from its annual mean concentration provides information on stratosphere-to-troposphere transport. We have clarified this in the manuscript.

p5. 19387/par 15 why active springtime chemistry ? Is the "wintertime downwelling" not sufficient to explain CFC-12, HNO₃ and O₃?

We have changed it to "wintertime downwelling".

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