Interactive comment on “Passenger aircraft project CARIBIC 1997-2002, Part II: the ventilation of the lowermost stratosphere” by A. Zahn et al.

Anonymous Referee #2

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This second paper based upon measurements from the CARIBIC project presents results on different aspects of stratosphere-troposphere exchange. According to the abstract, the results can be grouped into three topics: (i) chemical composition of the extratropical tropopause mixing layer, (ii) dynamical processes associated with STE and (iii) estimation of the cross-tropopause flux of ozone in the extra-tropics. These are all very important issues for our chemical and dynamical understanding of the transport processes near the extratropical tropopause. However, I have problems with important aspects of this study (as I will try to explain in more detail below): results on topic (i) are not all clear and consistent, the interpretations on topic (ii) are (too) speculative and not in agreement with other recent publications and the estimation of the O3 flux (topic iii) is not described well enough. I recommend to do a major revision before the paper can be reconsidered for publication in ACP.
Major comments:

A) The discussion in sections 3-5 is not always clear and consistent. For instance: a) section 3 para "A similar ...": I do not fully understand the aim of the slope analysis (dO3/dPV). First it is said that one should not look at O3/PV because of "possibly varying contributions from tropospheric O3". But in the end the conclusion is that the slopes dO3/dPV can only be explained with "inmixing of tropospheric air". Does the slope not vary mainly due to the differing diabatic downward transport of ozone from the overworld? b) section 4 para "Therefore, the stratospheric ...": why is the "intensification of the seasonal O3 variation" emphasized so strongly? Plotted in Fig. 1d/e are relative variations. A Figure with absolute variations would look differently and not show this maximum in the lowermost stratosphere. c) section 4 para "The only remaining ...": I do not agree with the hypothesis "this inflow should reach its maximum impact in autumn in order to explain ...". Before, it was mentioned that the stratospheric impact on TP-level O3 maximizes in April and that the observed max. of O3 at the TP is in June. I can't see how a max. tropospheric influence in autumn can shift the O3 max. from April to June(?). Also, Figure 3 shows max. CO transport into the stratosphere in spring and a minimum in autumn - in contrast to the argumentations in the paper for the role of TST on O3. d) section 5 para "The mixing line ...": Are the end points of the mixing lines (Fig. 4) taken to define CO(UT)? Why these points and not the rectangles shown also in Fig. 4? It is not mentioned/ discussed that for some flights the rectangles differ strongly from the squares. How, in such a case, can a representative value for CO(UT) be determined? Also it seems to me, that 94+/−10 ppbv (Table 1) is not a very accurate estimate for the values shown in Fig. 3 (e.g. <70 ppbv in autumn). e) section 5 para "O3(UT) is by ...": Here it is stated that "the seasonal variation of CO(LMS) is apparently small". This is in disagreement with the description of Fig. 3 in the last para of section 4 ("CO maximizes in April in the UT ..."). It is confusing for the reader that two different analyses of the same data set lead to not fully consistent results. A similar problem occurs in the same para in section 5: The formulation "This apparent discrepancy ..." (between the CARIBIC and Hohenpeissenberg data) is in
contrast to the previous discussion of Fig. 1 (first para in section 2: "... close agreement is manifest").

B) The estimation of the cross-tropopause flux of ozone in section 6 needs further explanations: a) Are the terms "downward", "net" and "net downward" O3 flux properly used? I think that only the downward O3 flux is considered in this section. To get numbers for the net flux, consideration of the upward O3 flux would be necessary. b) I do not understand equation (1): Why is the O3 value taken at the top of the mixing layer and not at the tropopause level? Both O3(LMS) and the mass flux across 380K have a distinct annual cycle: is the simple product of their annual mean a good approximation for the annual mean ozone flux? c) In part I section 1 para "Although mostly ..." it is written that "... there are several applications for which these two physical tropopauses (thermal, PV) are not suitable ... when the transport of trace constituents across the TP is to be studied". Nevertheless, in part II mass flux values from Appenzeller et al. across the 2-pvu TP are used. d) para "Assuming that ...": I can’t follow the discussion here: What is meant by "do not alter much"? and what is the "‘6" peak-to-peak value?

C) I do not agree with several interpretations given in sections 7 and 8: a) section 7 para "(ii) The O3-CO ...": From the compactness of the tracer-tracer correlations it is inferred that for instance deep convective injections across the TP are of minor importance. I think that a such a conclusion can not be made based upon tracer-tracer correlations because a very recent "injection" of tropospheric air into the LMS (whether through deep convection or any other dynamical process) can not be identified in a O3-CO correlation plot. The "injected" tropospheric air still has low O3 and high CO values and can not be distinguished from "real" tropospheric air. In my opinion, O3-CO correlation plots are excellent to study what happens in the stratosphere AFTER the TST event, but they contain very limited information about the dynamics of the TST event itself. It is important to distinguish between TST/STT (transport) and subsequent mixing in the LMS. b) section 7 para "(iii) Another constraint ...": What exactly is meant by "surprisingly small short-term scatter"? (or in section 8 para "Importantly, the observed
"surprisingly weak short-term variability"?). It is in contrast to the formulation in the abstract (maybe a typo?) "The observed surprisingly weak spatial homogeneity"!

Reference is made to Figs. 5 in both articles. I would not call the variability "surprisingly weak": The plus/minus 1-sigma variation at a given time is almost half of the amplitude of the interpolated seasonal cycle. c) Furthermore, I can not see how the qualitative assessment of the variability shown in Fig. 5 can lead to the conclusion that "STE is not controlled by the local instantaneous meteorological situation". In order to see the impact of the local dynamics on the chemical composition of the LMS it would be necessary to look at the detailed time-resolved measurements from CARIBIC. If I understand Fig. 5 correctly it does contain only one value per flight and does can not resolve mesoscale structures. d) Sprenger et al. (2003) (not in list of references) have estimated the cross-TP mass fluxes in the vicinity of TP folds and found significantly higher values than in non-folded locations. These estimates might be inaccurate but they are in disagreement with the statement in the paper that "... regulate only to a minor degree the amount of air transferred". e) section 7 para "In summary ...": What is meant by "this exchange (below 320K) is presumably shallow and will not result in significant net tracer exchange"? Why should the (according to Sprenger and Wernli) much weaker exchange above 320K lead to a more significant tracer exchange? Also, only the exchange below 320K is sometimes "deep" (terminology used in Wernli and Bourqui, Sprenger and Wernli, Stohl et al.) that is, it leads to a rapid and direct transport of air between the (polluted) lower troposphere and the stratosphere.

Futher comments:

1) section 1, para "(i) Stratospheric air ...": I don’t know about the study by Junge, but Ehhalt and Haumacher investigated strontium 90 in rain, i.e. the wet scavenging of fallout debris. I am not sure that their results can be compared directly to the Lagrangian studies mentioned in this paragraph. Stohl et al. investigated the seasonal cycle of direct transport of stratospheric air down to the boundary layer which is different from wet scavenging. Typo: "Wernli et al." should read "Wernli and Bourqui", also below.
2) section 1, para "(iii) After studies ...": What is meant by "no consistent, quantitative, and 3-dim illustration of ... is available to date. The studies by Sprenger and Wernli, and James et al. (2003, JGR) are quantitative, they consider 3-dim aspects (deep vs. shallow exchange) and they are consistent as far as the utilized ECMWF data is consistent. Please be more specific (maybe it is meant that quantitative estimates of chemical species are missing?).

3) section 1, para "The O3 and CO data ...": It is mentioned that in Part I "details can be found about the meteorological conditions along the CARIBIC flight routes". Also later in section 5 there are references to section 2 in part I "CARIBIC aircraft sampled stratospheric air almost exclusively around tropopause fold structures". However, I can’t find neither a detailed meteorological description nor mentioning of tropopause folds in part I.

4) section 4, para "The only remaining ..." (and generally throughout the paper): many model calculations (e.g. Chen 1995) looked at selected isentropes and found a distinct seasonal cycle for these isentropes. Sprenger and Wernli (2003) showed STT and TST mass fluxes for all isentropes from 290-350K and indicated, that inferring the seasonal cycle of STT or TST from selected isentropes is not representative and can be misleading.

5) section 8 para "In agreement ...": Why is the O3 max. at the tropopause in Fig. 1 in June and in Fig. 5 in April/May?

6) General: the term "tropopause transition layer" is mainly used in the tropics. Here it is used as kind of a synonym for the extratropical mixing layer. I suggest to only use the latter term.

7) Typos: "Ovarlez et al." (section 1); Solomon et al (not italic, section 4); "If this growing ... was only due ..." (section 4); "Ovarlez et al." (section 5); "constraints" (section 6); Seo and Bowman (2001 or 2002?).