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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evidence of tropospheric layering: interleaved stratospheric and planetary boundary layer intrusions" *by* J. Brioude et al.

J. Brioude et al.

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We thank the referee for her/his helpful comments, and we would like to follow her/his suggestion to motivate this paper as a scoping study for a future analysis of a much larger MOZAIC dataset. In this response we intend to show that our Lagrangian technique can motivate the study better in terms of a climatology of stratosphere-troposphere exchanges. Preliminary results we have got with the Lagrangian technique processed over a few hundreds MOZAIC profiles in winter and in summer are very encouraging. They show that the work is in the pipeline. In writing a revised version of the paper, we propose to add a short section on perspectives based on these preliminary results. It seems to us that the paper is relatively short and to the point, and that in adding a short section for the perspectives of the climatology, the paper will be worth publishing.

Below, we answer to the general and specific issues raised by the referee.

Referee: "Examples of tropospheric profiles with air masses of differing origins have been in the literature many times in the past and I am not convinced that ‘another’ example of a stratospheric intrusion in the troposphere is needed. As the authors state this particular profile has already been discussed in the Nedelec [2003] paper. Thus this case in itself brings nothing new to the literature."

With regard to the particular MOZAIC profile discussed here, only the highest tropopause fold at 4.7km altitude was commented in the Nedelec et al. (2003) paper. Furthermore, the latter paper had the main technical objective to validate the MOZAIC carbon monoxide instrumentation. Case studies are one way of learning about the processes, and with the development of new techniques, the level of insight also increases. The Lagrangian technique we are proposing indeed has reduced the uncertainties inherent to stratosphere to troposphere transport on that particular profile. In addition to detect the 4.7km altitude tropopause fold, the technique demonstrates the existence of a second tropopause fold just above the boundary layer at 2.2km of altitude, with an ozone signature as small as 8 ppbv in excess of the tropospheric background. With regards to the tropospheric ozone budget that all global chemistry transport models still do not perform in an accurate manner, and given the high degree of precision of this technique, we think that the case study exemplifies a way of improving our quantitative assessment of the stratospheric flux.

"The Lagrangian analysis performed on this intrusion is again not in itself novel. This form of analysis has been performed extensively by Andrea Stohl, Co-worker and other groups. Although I don’t think it has been applied to this particular example, the conclusions drawn from this example are no different from those conclusions drawn from other examples using this technique."

It is true that Lagrangian analyses being accompanied by synoptic analyses have been extensively applied to document stratospheric intrusions. However, the reviewer’s

ACPD

7, S801–S809, 2007

Interactive Comment

statement is not valid with regards to the application of the RDF technique with a dispersive Lagrangian model. To the knowledge of the authors, this form of analysis has only been applied by Legras et al. (2003), and furthermore the goal was to reconstruct stratospheric ozone profiles. This is a novelty to apply this technique to reconstruct tropospheric ozone profiles. Another novelty of our technique is to couple forward and backward runs of FLEXPART to reconstruct the stratospheric-origin contribution in a profile. Results on this particular profile show that the technique allows the detection of stratospheric intrusions as old as 6.5 days. Finally, the possibility to systematically apply the technique in a blindfold mode on large datasets, i.e. without accompanying synoptic analysis, gives it a much more general and meaningful character. It will allow to better investigate at seasonal and climatic scales the stratospheric intrusion term of the tropospheric ozone budget because the framework in which to apply the Lagrangian technique will be controlled by a large observational dataset.

"The last paragraph of the conclusions sections suggests that a systematic analysis of all the MOZAIC data will be conducted in this way and thus this paper basically acts as a scoping study for this future wider and more interesting paper. I would suggest therefore that this paper is motivated in that way - as a scoping study for this future work. I'm not sure that we need another example of a stratospheric intrusion in the literature - enough exist. However, a systematic Lagrangian analysis of the MOZAIC dataset would I think provide a unique insight into the processes controlling the concentration of O3 in the troposphere and this paper could be reformulated to describe the methods for this study. If this paper can be re-written in this way I am will be probably happy for it to be published with the corrections suggested below. Otherwise I am not minded to have another paper describing another example of a stratospheric intrusion in the troposphere."

We are willing to follow the reviewer's suggestion in motivating this paper as a scoping study for a future analysis of a much larger MOZAIC dataset. In re-writing the paper,

ACPD

7, S801–S809, 2007

and in addition to the demonstration of the performances of the Lagrangian technique on the particular MOZAIC profile, we propose to add a short section on perspectives. Preliminary results we have got in processing a few hundreds MOZAIC profiles with the Lagrangian technique will be shown. These preliminary results are described below.

MOZAIC data for 2003 over Frankfurt have been used. This choice is motivated by the fact that 3 MOZAIC aircraft (over a total of 5) were operating at that period of time on that airport. Data processed so far are representative of two seasons, the winter from January 15 to February 27 (representing 110 profiles), and the summer, July and August (representing 129 and 140 profiles, respectively).

Compared to summer profiles, the monthly winter profile has the lowest ozone mixing ratios and the largest CO mixing ratios in the troposphere. Conversely, July and August have larger ozone mixing ratio and lower carbon monoxide mixing ratio in the troposphere. Although of interest, variations in boundary layer profiles below 2km altitude are out of the scope of this study. Synoptic conditions throughout August 2003 involve a strong heat wave, anticyclone blocking conditions and a higher tropopause compared to July (Trigo et al., 2005). Ozone differences between July and August start from zero at 6.5km altitude and increase up to 35 ppbv at 10km altitude in the vertical ozone gradient characteristic of the tropopause layer.

Monthly reconstructed RDF-ozone profiles are also used. Coherent and significant monthly-scale contributions of stratospheric flux start above 2.5km altitude (4.5km) in winter (summer) and increase with altitude. The winter profile gets the highest contribution of the stratospheric flux in the entire troposphere. About 33% of the observed ozone at 3km altitude is explained by a stratospheric contribution. This finding agrees with the winter seasonal occurrence maximum of deep stratosphere-troposphere transport events (Wernli and Bourqui, 2002). Such a monthly-scale signature of the stratospheric-origin contribution into the lower troposphere in winter further gives a rationale to the case study presented in the paper. The stratospheric contribution comes to about 50% at 5km altitude and increases up to finally justify 100%

ACPD

7, S801–S809, 2007

Interactive Comment

EGU

of the observed ozone profile at and above the tropopause. For July and August the stratospheric contribution is lesser, although being quite significant in the middle troposphere (about 25% at 6km altitude) and increasing in altitude (up to 50% in August at 10km altitude). Interestingly, the increasing difference of ozone seen with altitude between monthly observed profiles of July and August is faithfully reproduced by the monthly RDF-ozone profiles. A preliminary conclusion then is that the ozone decrease in the upper troposphere across the summer is associated with the decreasing of the stratospheric-origin ozone contribution, which agrees with the reinforcement of anticyclone blocking conditions during August.

Winter and summer monthly RDF-ozone profiles are computed in different ways with regards to the time frame of backward trajectories. The winter profile is an average of the ensemble of reconstructed ozone profiles available every three hours from day D-5 to day D-10 relatively to the date D of every MOZAIC profile. Sensitivity tests show that this is the latter period of time during backward trajectories that optimizes the stratospheric contribution. If the averaging is done with available profiles from day D to day D-4, the winterly contribution of stratospheric-origin ozone at about 3km altitude strongly decreases. In agreement with previous studies (Wernli and Bourqui, 2002 ; Sprenger and Wernli, 2003, James et al., 2003), it shows that winter deep stratosphere to troposphere transport events that have a significant importance on the tropospheric ozone budget have quite long residence times, i.e. larger than 4 days on average.

Summer RDF-ozone profiles are constructed using the ensemble of reconstructed profiles available for the 10-days period of time for backward trajectories. Sensitivity tests show that monthly summer RDF-ozone profiles based on the average of three-hourly values picked up within periods of two consecutive days chosen at any time across the 10-days period of time for backward trajectories strongly resembles the ones shown on Figure 1. The meaning of such a coherence of the stratospheric-origin contribution on short time periods is that summer stratospheric intrusions have on the average quite short residence times (less than 2 days).

ACPD

7, S801–S809, 2007

26 MOZAIC ozone profiles over Cairo (Egypt) for the winter period (January, 15 - February, 28, 2003) are also used. The vertical profile shows a mean tropospheric ozone mixing ratio of 40 ppbv with a maximum of about 55 ppbv between 700 and 800 hPa. The RDF-ozone profiles computed as an average of the ensemble of reconstructed ozone profiles available every three hours from day D to D-4 and from D-5 to day D-10 capture the latter ozone maximum. According to the results of the Lagrangian technique from D-5 to D-10 and from between 700 and 800 hPa, up to 60% of the ozone (33ppbv / 55ppbv) may be associated with stratosphere to troposphere transport. Lagrangian studies have shown that tropopause folds are the key feature for cross-tropopause exchange in the subtropics (Sprenger et al., 2003 ; James et al., 2003 ; Sprenger and Wernli, 2003 ; Wernli and Bourqui, 2002), either directly related with the subtropical jet activity or indirectly related with the polar jet activity (followed by the southward transport of tropopause folds). The preliminary result shown here is encouraging and shows that the Lagrangian technique we propose will allow further quantitative assessment of the impact of tropopause folds on the tropospheric ozone budget.

comments: Why are the NOy observations not used for the analysis? Was it not the NOy equipped aircraft that made these observations? If this is the case it should be stated: MOZAIC NOy observations are performed by only one MOZAIC aircraft. It turns out that it was not that aircraft which performs this specific ascent (February 10, 2002).

graphs are often missing axis labels. This is unacceptable. The graphs are often hard to read and interpret. The authors should spend some time simplifying graphs so that they are easier to read and understand. Figure 3 is especially difficult. I'm not convinced that the 3 plots need to be in one figure. The graphs will be improved in the revised version. But, we might already try to clarify legend of fig3.

Fig. 3. Stratospheric-origin indications of FOLD2 on (a) horizontal Flexpart map, (b) GOES-EAST satellite image and (c) Flexpart vertical cross section using the February

3, 18:00 UTC 64257;elds (i.e. 6.75 days prior to the time of observation see fig 1, 10 February 12:00UTC). (a) : stratospheric ozone tracer at 5km altitude as derived from Flexpart simulation in the forward mode. Contours represent the percentages (0.05%, 0.1%, 0.25%,0.5%, and 1.5%) of air parcels associated with the retro-plume of FOLD2. Green, red and black contours are for particles with end-of-trajectory altitudes in the layers 0.5-3 km, 3-5 km, and 5-10 km, respectively. The red cross indicates the position of the cluster of particles described in the text. (b) GOES-EAST water vapor channel radiances (in K). Black contours represent the contribution of the stratospheric ozone tracer (0.01, 0.015, 0.02, 0.025, and 0.03 ppbv contours) related to the RDF-ozone of FOLD2. (c) Vertical-cross section of the stratospheric-ozone tracer derived from the FLEXPART run in forward mode. The trace of the cross-section at the surface is indicated on (a) and the red cross represents the position of the cluster described in the text.

description of the lagrangian model products is very confused. This should be worked on to make it clearer and more explicit as to when the forwards and backwards approaches are used. We will re-write the text more precisely in the reviewed version.

The authors use the term ‘residence time’ frequently within the paper. This has a specific scientific meaning but I don’t think that is what is being used here. The authors should define what they mean by ’residence time’ - I think they mean the time since the feature was formed in the atmosphere but they should be more specific in this regard. The reviewer is right. We employ the term "residence time" as the time since a feature was formed in the atmosphere. We will define it more precisely in the revised paper.

authors use the word ‘particles’ when referring the movement of airmasses. This is confusing the atmosphere contains particles in the form of aerosols, the Lagrangian model uses the concept of particles in the transport of air and then the authors use it to mean ’air mass.’ The authors should attempt not to refer to particles unless they are referring to the Lagrangian calculations rather than the general transport of airmasses.

ACPD

7, S801–S809, 2007

Interactive Comment

We will make clearer the use of particles in the concept of air transport representation by Lagrangian models and will avoid to use this word when referring the movement of air masses.

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ACPD
7, S801–S809, 2007