

Reply to the reviewers of manuscript ACPD-2010-0554:

High-ozone Layers in the Middle and Upper Troposphere above Central Europe: Potential Import from the Stratosphere along the Subtropical Jet Stream,

by Thomas Trickl, Nathalie Bäertsch-Ritter, Holger Eisele, Markus Furger, Robert Mücke, Michael Sprenger and Andreas Stohl

Thomas Trickl, May 16, 2011

General Remarks

I am glad to see a somewhat positive general impression in both reports, but I am surprised by the amount of concern listed in the reviews since many ideas of many critical readers (including the co-authors and myself) had already been adopted. The main criticism concerns the amount of detail and some style issues. Assisted and stimulated by the very constructive reports further shortening has become possible. I also rearranged some of the text and added some sentences on processes extracted from the literature provided. I also appreciate the suggestion to transfer some of the figures to an electronic appendix ("supplementary material") as a solution and prepared such a file.

The reviewers ask for more information on the relevant processes and less details on the analysis. The bad message is: There are no experimental data or model results available for visualizing these processes (as mentioned in the Discussion). Thus, we cannot get beyond the information the models used in the study are delivering, and modelling has limitations. Because of these limitations we have examined such a high number of cases which adds more confidence to our conclusions. The material finally presented is a small fraction of what has been analysed.

I added more statements indicating what is known and what is new, but tried to avoid expressions such as "novel". The Introduction was modified to relate our results clearer to the jet-stream mechanism.

Reply to Review 1

The texts from the two reports are written in italics.

This paper describes analysis of high ozone episodes observed in layers in the mid-troposphere over Europe using trajectory modeling techniques, and attributes some of this ozone to stratospheric sources following influx associated with the subtropical jet stream. It describes a number of case studies when mid-tropospheric layers containing high ozone were observed but could not previously be explained, and it resolves these by attributing them to stratospheric influence over longer timescales than had previously been considered. It demonstrates that this mechanism is important, but does not extend as far as quantifying the impacts.

A quantification of the impacts is clearly beyond the scope of this effort. A refined study of the kind of (Sprenger et al., 2003) could be a solution. It would certainly also be interesting to estimate the fate of all the ozone further downstream. As we have been frequently seen an erosion of free-tropospheric layers can be awfully slow (I just submitted a new manuscript on this issue to ACP). I added a few sentences to the Discussion.

I found the manuscript very interesting but somewhat difficult to follow. It is heavily descriptive, containing a high level of detail and revealing a very thorough analysis of some high-quality atmospheric measurements. However, the scientific contributions are not immediately clear, and are in part obscured by the level of detail in the description. The authors need to extract the novel

scientific aspects of this study (the focus on long range transport of ozone from shallow intrusions, and the importance of influx along the subtropical jet stream) and stress these more strongly.

Major adjustments were made. Nevertheless, the analysis of the observations is a primary focus and must be described in sufficient detail. I do not want to emphasize too much on the progress in modelling required for this effort. This should be obvious to the specialists I am not aware about a similar thorough study analysing transport once around the globe for such a comprehensive data set.

At present there is too much emphasis on explaining observational details and too little on providing valuable insight into the processes involved.

I cannot provide insight in processes that occur on the other side of the world (see also reply to Reviewer 2). Some more literature on what is known on processes, kindly made available by Reviewer 2, is now briefly discussed in the Introduction. The amount of detail of the analysis is a really small fraction of what has been done. It is important to present a minimum amount of the work to convince the reader about its quality and to show that the models quite nicely reproduce the detailed findings and, therefore, for the transport along the jet stream should be much more reliable than known from assessments in the lower-troposphere.

Considerable effort has gone in to the high-quality analysis described, but the resulting paper needs some reorganization if it is to make a substantial scientific contribution. It needs to state explicitly how it advances the state of knowledge beyond the earlier published analyses that are cited, and how it might be extended to provide insight into the mechanisms involved or into the magnitude of their impacts (even if this is not followed up here).

This suggestion is adopted in the revised version. Here, some of the mechanisms mentioned in the literature are now available and a paper mentioning that STT along the subtropical jet stream had for a long time been clearly underestimated is cited. It was also tried to emphasize more what is new. There is no way to identify the detailed processes involved in our case because of the very long travel (see also reply to second review). A few statements about the potential impact are added to the Discussion.

The topic of the paper is clearly appropriate for publication in ACP and is likely to be of interest to readers. However I would recommend revision and believe that considerable clarification is needed before it is ready for publication. Specifically, I recommend that the case studies section is rearranged, and provide further suggestions below.

Rearranging and shortening has been done.

General Comments

The paper would be substantially clearer if the authors explained their hypothesis clearly and in detail to start with, and then used the case studies to support their assertions.

This had already been done, but was perhaps obscured by the paragraph on the progress in modelling in the Introduction which is shifted and shortened now. A part of Introduction was rewritten to relate our work better to what is known. However, an empirical approach is, still, preferred. We do not want to define a hypothesis and see if there are data to support it!

I would suggest that these three case studies are combined so that the situations can be compared and contrasted right from the start, and that the subsections then introduce the different analysis approaches.

I have problems with such an approach. Perhaps I do not understand this comment correctly, but I have been sure that a joint description of somewhat different cases results in a considerable chaos. Instead, the approach chosen shows three almost identical sequences that can be easily compared because of the returning structures. I added an overview on the general, returning structure of the three chapters on the cases studies. The problem had been the complex second case study, but I decided last year to throw away the full discussion of three of the four examples which were too difficult to describe in a coherent way. Now, I removed even more material. I also shifted misplaced information on what is already known into the "Background" section. This makes the description of the observational material much shorter.

Currently the case studies are described sequentially (one per subsection) and the analysis approaches are applied to each (one per sub-subsection), and the thread of the paper therefore jumps about too much. While there is much detail of interest in each case study, it is easy for the reader to lose their way because the level of detail distracts from the main thread of the argument.

As mentioned, I do not fully agree. However, further improvements have been achieved. An introduction to the general structure is added.

Many aspects of the case studies have been described in earlier publications, so the authors need to be more selective about what is shown here.

This has been explicitly attempted. However, on request I had added short summaries on the earlier findings to the "Background" sections. The four-day ozone time series for two cases are not shown because they are already published. Nevertheless, I now add that for the first case to the electronic appendix.

I appreciate that some attempt has already been made at this, e.g., by omitting the lidar time series figures of the first two case studies, which are published elsewhere, but this omission actually makes the case study descriptions more difficult to follow, as it requires extensive reference to previous papers to follow the arguments.

I appreciate this sentence since this exactly what I have been thinking! However, since I prefer to have all material available when reading a paper I changed my mind and added the first time series to the electronic appendix (see above).

This paper should focus on the aspects of the study supporting the current arguments alone (although the lidar profiles are required to provide context). The written style of the paper is formal and educated, but the phrasing is awkward and long-winded in many places, and there are elements of narrative (e.g., at the start of section 3.1.4) that are unnecessary.

I tried to make further improvements, based on both reviews. The start of Sec. 3.1.4 was modified, and one narrative sentence in Sec. 3.2.1 was removed (this information is already given in the "Measurements" subsection).

In many places the ideas could be expressed more clearly and concisely without loss of information, and I have identified some issues below. There are too many cross-references, both forward references to explanations occurring later in the text and to previous studies. While this is useful for the expert searching for specific details, it is not helpful for the general reader who is looking for a coherent scientific story running through the paper. If 20-day simulations are available, it would be good to focus only on these.

I removed some of the cross references.

It is explicitly stated that the crucial 12-km panels are only available in the 15-d simulations. The 15-d plots are also important for comparisons with the LAGRANTO results because they indicate where the plume was next to -15 d.. The colleague who created this special option, quit before the end of the project, and the option (or are more refined one based on PV) was never introduced into the new operational package of FLEXPART. There is no doubt that I would have preferred to stay with a single modelling approach!

Sections 3.1.3 and 3.3.3 currently introduce the 15-day simulations first and then describe the 20-day simulations; it is not clear why this is necessary or what this adds, and it makes the text more difficult to follow. There is no discussion of the reliability of the trajectories used here. Given the uncertainty in the meteorological data and the propagation of errors over the long timescales considered, it would be useful to present some assessment of reliability, or to cite previous studies that have assessed this.

This is now done more explicitly in the Discussion. Some statements had been already available!

Very little use is made of the Lagranto analysis except in Case 1. Can this be tied in better for the other case studies? Again, it would be clearer if this analysis was discussed for all case studies at once.

It does not make sense to repeat all the rather similar findings. Thus, just brief description of what agrees with the first case and what not is given for the other two. The figure for the second case was moved into the supplementary material, following the suggestion of the second reviewer to consider this possibility for some of the figures. The discussion of the results for Case 3 was extended: Because of its clarity this is the key case in this paper and Reviewer 2 asks for more details.

To discuss all cases (in less detail) at once is a matter of the Discussion. In the Results section such an approach would be enormously confusing.

Specific Comments

The abstract needs to be rearranged. The scientific contributions of this paper are left until the last two sentences of the abstract, and the statements made here are vague. The new results of this study need to be stated clearly here.

The abstract is changed. However, the observational material and the model development are important parts of this work and I hesitate not to name these contributions a less important result.

30475, l.18: The first sentence of the introduction should be reversed so that the subject (dry air masses with high ozone) comes first.

This is obvious, thank you!

30479, Section 2 describing the methods should be condensed, particularly the measurement details which are covered in previous papers. The section should focus on the new aspects of the analysis used in this paper.

The description of the instrumentation is already rather condensed, and references are given for more detail. I just shortened the sentence on the ozone sondes. In the case of the models we have been asked for this kind of explicit description earlier. The models are crucial for the understanding of the progress made and an explicit description had been demanded at an earlier stage. Nevertheless, I found several sentences that could be removed.

30478 1.19 and 30491, 1.13: *It seems a little irregular to refer the reader to the earlier, non-peer reviewed ACPD version of this same manuscript for description of the cases not described here!*

The fact that that paper was rejected does not mean that its content was wrong. The past reviewers have mostly complained about the number of case studies, the complexity of the descriptions and the lack of more details on processes. This is now done. It is fully adequate to cite this material since it is available in ACPD and this offers the unique chance to make available some of the missing details for interested readers. Nevertheless, I moved this referencing to Table 1 where it does not interrupt the text.

30488 1.24: *the estimation of stratospheric influence here is crude. It would be helpful to provide some justification for this approach to provide the reader with a better understanding of the associated uncertainties.*

It is obvious that just a crude estimate of the upper limit that can be expected is attempted. Typical LS lidar results usually are 200-500 ppb, and the numerous MOZAIC data for flights across the North Atlantic examined for (Trickl et al., 2003) also show ozone values of about 500 ppb at cruising altitude. The calculation is done in the following way:

$$500 \text{ ppb} * 0.12 + x * 0.88.$$

For $x = 40, 50,$ and 60 ppb one obtains an enhancement by 55 ppb, 54 ppb and 53 ppb, respectively. Numerical particles that exhibit STT at later times do not matter in this simplified calculation since they were tropospheric on the previous day, very likely too short for becoming fully stratospheric. I slightly modified this paragraph.

Figures

There are a lot of figures in the paper, and combining some (e.g., Figs 2 and 3) would allow them to be compared and contrasted much more easily. If the three case studies are considered together, there would be much more scope for this, e.g., combining the retroplume, Lagranto and vertical profile figures so that the case studies can be compared under the same analysis approaches.

Combining Figs. 2 and 3 is not possible since they are too large. Omitting one of them is also not possible because the difference in speed over the Atlantic for the two layers is too high for the 15-d simulation and, thus, the full pathway is not visible here. The full pathway is only seen for the faster upper layer. This is the first case study of this kind in the paper and, therefore, a more explicit description is adequate. In other situations I now moved figures to the supplementary material following the suggestion of Reviewer 2.

The retroplume summary figures (Figs 5 and 11) are not easy to interpret. it would be clearer to present the clustering as a probability density with the magnitude represented by the color or shading. The current method using circles is difficult to make out, and neighboring circles overlap each other. The figure would be more legible if the ABL and STR panels from the three layers are each combined into one so that there are three lines on each panel.

These figures will be printed in full-page layout in order to make the details better discernible. A full identification is really not crucial since we are only interested in the uppermost cluster. Otherwise, I would have used the horizontal map of the clusters that includes colour-coded altitude information. Merging the ABL and STR panels do not make sense in my view. For us, the STR panel is most important.

The contribution plots (Figs 6 and 20) need to be replotted so that the axis labels are coherent (not "nearest integer").

I had thought about this issue when improving the quality of these figures since I also had problems with this rounding. We now agreed on using least-squares-fitting approach to solve this problem since the raw data are not available.

Figs 7, 14 and 21: note in the caption that the variable shown is wind speed.

Thank you!

Other issues

admixed -> mixed into or mixed together (throughout text)

The word "rather" is overused, and the meaning is ambiguous in many places. Please replace it with "very" or "relatively" depending on context, and remove it where unnecessary. (throughout text)

30476 l.4: remove "e.g." and "on"

30476 l.6: remove "by us" (and cite study if needed)

30476 l.27/29: remove "even"

30477 l.9: "Quite differently" -> "In contrast" or "However"

30477 l.12: remove "on"

30477 l.19: "exhibits some similarity with" -> "is similar to"

30478 l.9: remove "also" and preceding comma

30478 l.16: remove "fifteen and now even"

30478 l.23: "We focus on presenting just three" -> "We present three"

30478 l.26: "as well as" -> "and"

30485 l.24: "representing an inverted atmosphere" not needed, remove.

30490 l.11: "not easily possible" -> "not easy" or "not possible"?

30491 l.25: The meaning of the sentence starting "The principal..." is unclear, please rewrite this

30499 l.15: "(in some cases: most)" confusing, please remove.

30502 l.15: hibernal -> winter

30502 l.24: rephrase "material" here

Changes were made with just two exceptions.

Reply to Review 2

This work by Trickl et al., presents and describes three case studies of ozone-rich air-masses to Central Europe, with the aim of investigating the possibility that STE occurring along the subtropical jet stream (SJS) can influence tropospheric ozone budget. The paper is a newer and improved version in respect to the paper already published on ACPD by the same authors in 2009 ("High-ozone layers in the middle and upper troposphere above Central Europe: strong import from the stratosphere over the pacific Ocean"). In respect to this previous version, the paper goal is more clear, new analyses were added and the paper structure was notably improved. The role of

SJS in triggering STE leading to stratospheric air-mass transport over Europe has been recognized in this work. This permitted to clarify some high-ozone events already presented in previous work (e.g. Trickl et al., 2003) but not well understood.

In general, I think that the authors should work a little bit more in making the reading more linear and, possibly, omitting some of the huge amount of detailed information provided in the paper. To this aim the authors should consider the possibility to move some of the figures to the electronic auxiliary materials.

This is a really good suggestion! I have not been aware of this possibility and I decided to move one LAGRANTO figure and other material to an electronic appendix.

On the contrary, also basing on the long experience of these authors in studying STE, I would like to see in the "Discussion" section more sentences (even speculative) about the dynamical processes (CAT? Tropopause foldings? Differential advection?) favoring the STE in connection with the SJS: this would represent a valuable add-on for other scientists working in the field.

It is impossible in the current study to differentiate between these different processes, especially since they are not acting separately from each other, but often in sequence. For instance, differential advection creates strong (horizontal) tracer gradients and tropopause folding brings stratospheric air masses low down in the atmosphere.

However, neither one of these processes causes actual mixing of stratospheric and tropospheric air. On the other hand, CAT causes mixing, but is most effective subsequent to differential advection and tropopause folding enhancing tracer gradients. Convection and radiative processes are also important for mixing and/or transformation of stratospheric into tropospheric air masses. In a numerical model study, some of the mentioned processes can be controlled (e.g., by switching off certain parameterizations), allowing to derive quantitative information on the importance of individual processes. In our study, however, which is based on observations and modelling using analyzed meteorological data, such a quantification is not possible (also considering the enormous distance and time difference) and we only see the total effect of all of these processes. Even in numerical model simulations, derived numbers for the individual processes are not always very meaningful, as the net effect depends non-linearly on the various processes. Thus, an accurate answer as to the relative importance of the various processes, is not even possible or meaningful. It is their total effect, which counts in the end.

One additional related sentence is included in both the Introduction and the Discussion now.

Concerning the analyses, my principal concern is related to the fact that (for all the presented case studies) the authors made a direct comparison of the in-situ LIDAR profiles with profiles of modeled quantities (i.e. the stratospheric air-mass fraction) at 10 - 20 days upstream to the measurement site. As I will explain in the specific comments, I'm not convinced about the reliability of this comparison ...

I am more and more convinced. It seems that long-range transport along coherent air streams can be more accurately calculated than lower-tropospheric trajectories that exhibit chaos and trajectories are horizontally only accurate to within 10 % of their length at best. If the backward trajectory propagates within a coherent air stream the chaotic behaviour quite obviously becomes much smaller. For the FLEXPART plumes the full spread is obtained and not single guesses. This gives a good idea of the probability of passing over a certain area. Of course the results do not necessarily

represent the absolute truth. However, both the FLEXPART and the LAGRANTO results were obtained based on the same ECMWF input data!

After some discussion with Dr. Stohl I added a few sentences related to reliability to the Discussion.

Basing on that, I recommend publication on ACP after revisions (mainly related with the paper structure) because in my opinion the paper is scientific-sounding and covers a potentially important and relatively not already extensively investigated topic (i.e. influence of sub-tropical jet stream on STE and O3 budget). The manuscript focus is within the ACP scope clearly demonstrate the effectiveness of merging high-quality measurements with model analyses.

SPECIFIC COMMENTS

-Introduction:

Pag 30475, line 1: basing on the number here provided (six 4-day episode from 1996 to 2001) it seems that these events can only play a minor role in determining the yearly ozone variability over central Europe: please comment.

This issue is beyond the scope of the current study! I added some statement to the Discussion. What is more important is that the subtropical jet stream over an extended period of the year could yield a significant contribution to the overall STT budget. We have not attempted to estimate how frequently this kind of high-ozone stream overpasses Central Europe. In favourable years this could be up to once per month during some part of the year (seasonal dependence of jet occurrence: see Koch et al.). A clarification requires a modelling study which could also tackle the interesting question about the further fate of these air streams.

A few adjustments were made.

Pag 30476, line 3: please indicate a reference (Trickl et al., 2003)

This citation was already made in the preceding sentence.

Pag 30476, line 4: also the works by Langford (1999) and Zachariasse et al. (2000) should be cited:

Langford, A. O. (1999), Stratosphere-troposphere exchange at the subtropical jet: Contribution to the tropospheric ozone budget at midlatitudes, Geophysical Research Letters, 26, 2449-2452

Zachariasse, M., van Velthoven, P.F.J., Smit, H,G,J, Lelieveld, J., Mandal, T.K., Kelder H., Influence of stratosphere-troposphere exchange on tropospheric ozone over the tropical Indian Ocean during the winter monsoon, J. Geophys. Res., 105, D12, 15403- 15416, 2000.

Thank you for this information which is now included!

Pag 30478, line 7: please add a reference.

The reference is this sentence itself. There are quite a few more observations of upper-tropospheric advection from beyond North America than those presented here. I removed "also in other investigations" since this is difficult to understand and just a brief hint on this exists in a conference paper on our long-term free-tropospheric aerosol observations (I hope that the analysis can be completed some time).

-2.1 Measurements:

Page 30479, line 10: please can you specify “lower” and “upper” troposphere?

This not ultimately important since the uncertainties also depend on the ozone distribution and sometimes on the residual solar background. Nevertheless, I introduced a number (< 4 km) to give some more concrete validity range for the specification $\pm 3\%$. "in the upper troposphere" could be converted to "next to the tropopause", but what would one really gain from this?

Page 30480, line 14: please can you add a reference where details on in-situ measurements (e.g. experimental set-up) were presented before?

I added (Trickl et al., 2010) which was co-authored by H. E. Scheel who operates the stations. There, a more explicit description is found.

Page 30480, line 15: the description of the experimental set-up is very detailed for the ozone sounding. Maybe you can shorten it and referring to earlier work for more detailed description.

This part is already considerably shortened with respect to the information I had received. I shortened the sentence about the ozone sondes.

-2.2 Models

Pag 30482, line 6-19: From these sentences is it not clear what the “Column” plates presented in Fig.2, 3, 9 and 18 are representing. I suppose these plates indicated the “emission sensitivity” described from line 6 to 11. Certainly, one can read the paper by Seibert and Frank (2004) and Stohl et al. (2003), but using the same nomenclature between the FLEXPART outputs and the figures presented in this work (as you did for the “footprint” product) will help!

I added the following sentences to Sec. 2.2: "Integrating the emission sensitivity over the entire atmospheric column, is an effective way of displaying the overall horizontal transport pathway of a sampled air mass. Showing the emission sensitivity for a certain altitude level (e.g., 12 km), illustrates the pathway at this altitude level." I also extended the caption of Fig. 2.

Pag 30482, line 20: For simplifying the reading, you can provide here the list of cases (or simulations) for which the “Frost et al” inventory have been used. Did you note some differences by using the two inventories?

The inventory of Frost et al. (2006) was used for all the twenty-day simulations, but not for the fifteen-day simulations which were done prior to 2006. I clarified this.

Page 30482, line 27 – pag 30483 line 2. If correctly understood, the fraction of stratospheric particle was only calculated for the 20-day simulation. Thus I suggest to move this sentence to page 30483 at line 25.

I do not like this sequence either: Changed!

Pag 30483, line 2 to 10: I suggest to move this paragraph (which guide the reader to the interpretation of retroplume cluster plots) to page 38488 (line 15) where this output is presented for the first time: this probably would further help the reader.

I do not agree. There, it would interrupt the flow. The key word "cluster" is mentioned in the description of Fig. 5 and should stimulate the reader to have a look at the section on the model description. The most difficult kind of FLEXPART panels, the "retroplume cluster plots", which show a confusing multitude of coloured circles and numbers, is no longer used in the new version of the paper. Thus, the understanding of the analysis sections is much easier.

Pag 30483, line 25: with the purpose of better helping the reader, you should indicate in the FLEXPART figures (by simply adding a code on the figure top) if these are referred to the 15-day or the 20-day simulations.

Good idea! I added phrases wherever this information was missing.

Pag 30483, line 11-25: I would shorten this paragraph. I would only explain the “hard” differences between 15-day and 20-day runs.

I removed a few less important sentences.

-3. Results:

Pag 30485, line 3-5: I would skip these sentences.

The introduction of the results section was changed anyway for more clearness. In this context this paragraph disappeared.

-3.1 Case 1:

Pag 30486, line 11: basing on table 1, there was 2 layers from 4.5 to 7.5 km and from 7.5 to 11 km. Please update.

The layer of main interest is the lower one because there is no indication of a contact with a remote PBL in the model results. I changed the phrase to "4.3 to 7.5 km" and also changed the number in the table. "7.5 km" in the table is a compromise since the upper end moved from 7 to 8 km.

Pag 30846, line 11: please substitute “in our 2003 study” to “Trickl et al (2003)”

Done

Pag 30487, line 17-22: this sentence can be skipped as the FLEXPART outputs have been already described in section 2.2.

To use all these different options of FLEXPART makes the entire description of the results difficult to understand. It is mostly the 12-km option that has led to the use the 15-day figures rather than those for 20 days. Thus, I had decided to add this sentence for clearness. I modified the sentence in order not to repeat what is written in Sec. 2.2.

Pag 30488, line 6: For increase the reading simplicity I would specify that this is referred to the 7 – 10 km layer. Moreover, how did you diagnose STT by trajectories? Did you analyze PV along the trajectories? Did you use FLEXTRA or LAGRANTO? You should be a little bit more specific Moreover, from this paragraph it seems that this STE (between Kamchakta and 40N, 180E) was the leading processes determining the observed ozone increase. However on the following (line 27) it was reported that: “there is little influence from the intrusions over Kamchatka”. Please, clarify.

To speed up the procedure I used HYSPLIT which I can operate myself without asking for support. Given the numerous details on models and modelling I did not want to confuse the readers. I added a short statement including a reference.

Pag 30488 line 10- 12: I would skip this sentence.

This is quite necessary with the changes made. Thank you!

Pag 30488, line 14. As already noted in the general comments I’m quite skeptical about the possibility of directly compare in-situ results with simulation profiles from 15-day earlier!!!

As mentioned, I added statements on the reliability to the Discussion. The backward plume covers the relevant regions. To my opinion its spread to a good approximation includes the uncertainties, relative to the ECMWF "truth". There are uncertainties regarding the spread which is influenced by the parameterization and numerical noise.

*Pag 30488, line 24: I'm not completely sure about these numbers...I don't think that you can extrapolate the amount of ozone transported over the measurement site simply multiplying the stratospheric air-mass fraction (calculated for air-mass 15 days before the arriving at the measurement site) for the average amount of ozone in the lower stratosphere (0.12 * 500 ppb) ! Surely, dilution and mixing processes can act in decreasing the concentration of stratospheric ozone.*

The direct comparison of tropospheric O₃ measurements and the stratospheric contribution derived from a combination of model results and "typical" O₃ values in the lower stratosphere is certainly not fully quantitative, but serves to show the order of magnitude of a possible stratospheric contribution. The reviewer warns that dilution and mixing processes could dilute the stratospheric ozone. However, these processes are accounted for by FLEXPART and are the ultimate reason why the fraction is not either zero or one. It is the dilution and mixing that generates a fraction f , $0 < f < 1$ at the measurement location and time.

Some changes were made (see also the reply to Review 1).

I'm wondering why you don't show the profile of the stratospheric fraction at the time of air-mass arrival over the measurement site?

20-day simulations were made for two times, 10 CET and 19 CET, the later one being the time of the measurement shown in Fig. 1. The vertical range in which enhanced STT fractions are seen agrees quite well. What would one gain from looking at the situation at the very beginning?

Pag 30489, line 24: The map showing the jet streams (Fig. 7) show interesting hints, nevertheless I had some difficult to link the different maps with the backward movement of air-masses. You may add a number (from 1 to 4) to each jet stream plate and then superimpose these number on the retroplume maps.

This is, in principle, an excellent idea, and I also had been thinking about this. However, the FLEXPART plume is not a set of trajectories with clearly defined positions and times. For Case 1 this is a particular problem since a lot of different clusters travelling with different air speed must be considered. However, in this case the latitudinal band is rather constant with time. What is known from the 15-d simulation is where the plume for the upper layer was approximately located at: above Northern Morocco close to the end of the backward tongue as mentioned!

The situation is much different for Case 5 (July 2001). Here, some assignments are possible and I decided to use the average position of the uppermost cluster in the 20-d runs for marking a cross on the respective jet-stream panel wherever such an assignment looked reasonable.

How the authors decided that the important STE were those over the Strait of Gibraltar, Caucaso, China, Japan and not (for instance) the green dots over West USA on May ,27?

This is easy to understand: the FLEXPART tracer plume does not suggest important STT there. A closer look reveals: time and altitude do not fit.

Pag 30490, line 5. probably the work by Langford et al. (1999) can be profitably cited in this context.

The full discussion is in Sec. 4. There, earlier work on this issue is cited. The paper by A. Langford is now cited in the Introduction.

Pag 30490: line 14: this sentence is not clear to me.

A travel time of less than 15 days means an average speed of the order of 80 km/h (or more when considering that the advection path is not straight)!

-3.2 Case 2

Pag 30490, line 16: Please add the starting day (as you did for the previous paragraph title)

Added

Pag 30491, line 14: please remove the word “final”.

Removed

Pag 30491, line 15: in my opinion these last sentences can be removed. They did not add nothing of vital to the paper.

They do! I added these sentences on purpose to make visible the astonishing reproducibility of the observations for such a meteorological situation for all these cases (I had missed this aspect in the 2003 paper because I did not have a look at the days before the beginning of the measurements). The measurement stated two days after the clearing and immediately showed that high-ozone layer!

Pag 30492, line 10: I supposed that the 60-80 ppb of ozone here reported, were referred to air-quality measurement in the US. In this case 80 ppbv of ozone would imply not-negligible photochemical production that in case of sufficient precursor emissions could have sustain photochemical production also during the air-mass travel!

In principle this could be the case, but so far we have never seen any evidence in any of our case studies and in the results of a number of modelling studies. The classical example is the case studied by Stohl and Trickl (1999) showing about 90 ppb on both sides of the Atlantic and at the cruising altitude of a MOZAIK Airbus. This finding was hardened by a EURAD CTM calculation within ATMOFAST: NO_x was removed from the air mass in the early phase.

As explicitly stated in the Discussion, there is also the possibility of lightning-induced ozone production which to my opinion creates higher uncertainties because there NO_x is produced next to the air streams of interest. Since the topic of the present study very-long-range transport I would like to stay with the present version. This part is now moved to the "Background" section because this was already known.

Pag 30492, line 25: did you rule out any influence from NO_x thunderstorm production?

Not fully! But this kind of contribution is not very likely soon after the thunderstorm due to the ongoing westerly advection. The measurements, unfortunately, were not resumed immediately. I have been looking for some opportunity like this one to look for ozone before and after a thunderstorm for many years, this requiring rapid clearing. However, a less complex background situation would be more favourable!

Pag 30493, line 1. I suggest to eliminate the sentences starting from “Most trajectories...”. A more deep analysis will be shown just in the following.

I agree, thank you!

Pag 30493, line 25: please rephrase: “We, thus, show results only for days from -20 to -15”

Done

Pag 30493, line 28 – page 30494, line 6: Again, I'm not convinced about the direct comparison of LIDAR data with simulated profile referring to 20-15 days earlier! I suggest to remove these sentences and, if not used for other purposes, Fig 10 (the same can be applied to Fig. 4 and Fog. 19).

I have problems in understanding this concern. It is important to stress this agreement, this is a key result. Should I tell the public that this result was not obtained? The big spike is where it should be and confirms the idea from the 12-km panels in the 15-day results. To me this is a great result, despite obvious uncertainties. I have been criticized to present that many cases in the earlier version (now even six cases have been analysed). This had been done on purpose to show that this kind of agreement is seen again and again and that these observations are astonishingly reproducible. Further shortening would give rise to more and more questions.

Our arguments concerning the reliability of 20-day modelling of coherent air streams have been discussed above.

Pag 30494, line 8: please substitute "in Figs. 8c and 10b" with "on 27 May at 11:00 (Fig. 8c)"

Done

Pag 30494, line 9: please add "(7000 – 7250 m)" after "in the aerosol profiles"

Done

Pag 30494, line 13: from Fig. 9 (plate 12000 m) I was not able to see any retroplume over Canada. Probably did you mean Alaska?

I had removed the corresponding figure for the twenty-day run. It is not seen in the time-integrated plot. Thank you! This detail can be omitted.

Paragraph 3.2.4: in the paragraph 3.1.4 you was very specific in identifying the location where STE could have been occurred. I suggest to be a little more specific also in this case.

The amount of detail in Sec. 3 has been criticized and, therefore, I am not happy with this suggestion. I think after the detailed description of the jet-stream results in first case study the reader should accept that I have done this job very carefully. I removed the LAGRANTO figures from the second and third case study and added them to the "supplementary material" for interested readers.

Pag 30496, line 11 to 21: I will skip or significantly reduce this very detailed paragraph.

I hope you really mean the humidity paragraph of Case 5! I have always been aware of this complexity, but in view of the importance of the humidity data I could not convince myself to remove all. I now moved much this part to the "supplementary material", and added the sonde profiles there.

-3.3 Case 5

Paragraph 3.3.2. It was quite difficult for me to follow this paragraph that appears a little bit confused. Contribution to layer L1 and L2 are explicitly described, but not for L3.

It is interesting to read this judgement. As mentioned, I had fears for the description of humidity data, but not for the rest. L3 is mentioned (Fig. 17)! I now added information on the vertical positions.

Moreover, it is almost not possible to follow the single back-trajectories on figures 16 and 17. Is it possible to show only back-trajectories for layers L1 and L3? Please check also the figure numbers!

Unfortunately, this is no longer possible, I had asked myself. Indeed, a close look is necessary to follow some of the trajectories. The plot will be printed large enough.

Pag 30497, line 13: "In addition, for L1. . .", this sentence is not clear. I suggest to remove it also considering that in the following only L3 layer will be analyzed.

I simplified this sentence and added it to the previous one.

Pag 30497, line 19: please substitute "It follows. . ." with "The "12000 m" modeled backward plume is very similar to the. . .".

I do not want to repeat the phrase "12000 m" which was used in the preceding sentence. My suggestion is "This slice of the backward plume follows the maxima of the column sensitivity even over Asia".

Pag 30498: once again, I'm skeptical about the effectiveness about the comparison of model results concerning vertical profile five to twenty days before the observations!

I think that this case study is the most convincing one, also due to the absence of emissions in layer L3. The conclusions clearly differ from speculation.

The sentence about EURAD results: this not adds important information for the paper.

The sentence about the EURAD results is a relict from the earliest version when EURAD co-authored this effort. It is a nice aspect, but there many nice aspects. I followed the recommendation and removed this sentence.

I think that the time-altitude plot you didn't show can provide more indications about the evolutions of the observed layers: I suggest to show it in place of Figure 19!

Fig. 19 is one of the most important figures in the entire paper.

Paragraph 3.3.4: you should add some details on this key paragraph. For instance it is not clear when the STE over East Asia and South of Alaska has been occurred. This information is important to understand the aging of air-mass observed at the measurement site.

This even is the key case! I added marks as described in the reply to Review 1 and added more explanations. LAGRANTO, in general, seems to underestimate STT because of the section criteria.

-4. Discussion:

Sometimes the case studies are referred following the numeration in table 1 and sometimes by using the date of occurrence, please homogenize.

Within the Discussion section I could not find much inhomogeneity. Thus, I examined the entire manuscript.

Pag. 30500, line 7: the paragraph citing EURAD simulation and discussing the complexity of modelling ozone input seems a little bit outside of the paper focus.

Here, the EURAD result makes sense since this one-year run should have captured more than FLEXPART. EURAD was, again, removed later in the Discussion section.

Pag 30501, line 14: STE can occur also at the exit region of sub-tropical jet stream (see e.g. Langford, 1999).

No doubt! But where is the exit in our case? What we write here is also related to the sum of the outflow from many partial exits of partial streaks. However, the assignment of STT events in the jet-stream figures is awfully difficult and would require the inspection of individual trajectories creating the respective dots.

-Figures:

Fig.2,3, 9,18: Please rename the third plate: “NOx PBL source contribution”

Done

Figures 4, 10, 19: Is it not clear to me if this stratospheric fraction is the average or the highest values over the considered time periods (e.g.,. from -20 to -15 days in fig. 4)

I used the highest values because according the rule of thumb used for Case 1 this corresponds to the guess of the ozone mixing ratio over our site. This is explicitly mentioned for two cases, and I now added this information also to Case 1.

Figures 5,11: what do the bubble diameters represent? (I suppose the number of particles). Please, specify in figure captions.

It is the number of particles as specified in Sec. 2.2. I added this information to the caption of Fig. 5.

Figure 15: please substitute the x-axis labels with dates and hours.

Done

Figure 16,17: is it possible to show only the back-trajectories related to layers L1, L2, L3?

This would be great, but is not crucial. But currently there is no chance to get support for FLEXTRA. I am going to ensure that the plot will be printed sufficiently large which usually is not a problem with ACP.

TECHNICAL COMMENTS

There are several typesetting errors (some are listed below). I suggest the authors to careful check the manuscript.

Pag 30476, line 7: please, delete one of the right brackets.

I do not see an error (accepted by ACPD here and in the other cases)

Pag 30484, line 26: please, delete one of the right brackets.

The bracket is correct, but this part was completely replaced anyway.

Pag 30486, line 11: please, substitute “our 2003 study” with “Trickl et al. (2003)”.

Done

Pag 30490, line 27: please, check the brackets!

I do not see an error

Pag 30491, line 13: please, modify in “Trickl et al. (2009)”

I am referring to the paper, not to the people!

Pag 30497, line 1: substitute “Fig. 15” by “Fig. 16”

This had been done in my WORD version, but not in the TEX document. Done!

Pag 30497, line 6: substitute “Fig. 16” by “Fig. 17”

Done

Pag 30504, line 4: please, delete one of the right brackets.

No error found

Thank you for this tremendous reviewing effort!