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Atmospheric Chemistry and Physics Discussions

Interactive comment on "High-ozone layers in the middle and upper troposphere above Central Europe: strong import from the stratosphere over the Pacific Ocean" *by* T. Trickl et al.

T. Trickl et al.

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General Remarks

After all the efforts of our team to optimize this manuscript we are somewhat shocked by the responses we received. When looking closely at the reports, I realized that it is frequently asked for what we explicitly tried to accomplish. From this I conclude that the main task is to improve the guidance through the text, and we made the necessary adjustments. The paper was thoroughly revised. Most importantly, a substantially more explicit interpretation is now given: I had been too careful in the first version.

In addition to elucidating the principal transport pathways it has been our principal purpose to demonstrate the reproducibilty of these observations (and their differences:

length, layer thickness, PBL contribution present or almost pure STT). Thus, we present as many as five cases, i.e., all relevant longer lidar series obtained during the warm seasons between 1996 and 2001 (it is exciting to realize that all these cases exhibit these specific layers!). The call to shorten the number of case studies to a maximum of two is in strong conflict with this goal. We accept shortening some of the case studies.

It had been planned to discuss the first two cases in most detail that show situations without and with Asian contributions, respectively. However, the description of the particularly nice measurements of Case 4 continues to urge me to present some more details also there. In fact, a closer look during the revision showed that this example gives the best hint on the importance of the branch along the subtropical jet on the modelled STT fractions, and the text was modified accordingly. Reviewer 2 suggested dropping the September-2000 case. This case is, indeed, the least important one, and I decided to remove the figures and to shorten the description quite considerably, referring to the material shown in 2003. The description of the very impressive fifth case is short anyway. This case is important because of the high ozone values lasting over a rather extended period of time and because of the absence of significant Asian contributions.

It is important to note that the length of the text itself is not excessive. It would not exceed six or seven pages in a printed version. It is the number of figures that needs to be addressed. The material was already strongly cut before the submission, starting from initially more than thirty figures. Even an aerosol profile prepared for Case 1 was removed that is now explicitly called for by Reviewer 2. We removed the September-2000 figures, but added some other useful material to address some of questions in the second report.

There are sometimes quite different views in the two reports, in particular concerning the material on emissions. Not in all cases I have been able to find a compromise.

Specific replies:

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Review 1:

It is absolutely not true that we just stress the ubiquity of layers. We clearly go beyond a paper such as [Newell et al., 1999] where just statistical aspects are discussed. We do address a number the questions of importance, such as:

- Where does the excessive ozone come from?
- What are the potential underlying mechanisms?
- Why is this STT mechanism so much different from the excessively studied deep intrusions?

It is true that the interpretation was insufficient and I apologize for just alluding to some of the relevant literature. A much more explicit discussion is now added, based on additional information I received from M. Sprenger. However, I could not answer the question concerning the reason of the reproducibility of these air streams. This should be the subject of a separate meteorological study. The main goal in our present study has been to analyse the observations, present some evidence of the underlying mechanisms discussed and to show the similarities and the differences. The reproducibility cannot be proved on the basis of just a single case. I am aware that reading all that material in detail may be tough, but its extent does not exceed that found in numerous publications in this field. However, I considerably shortened the description of the analysis of the least important, the September-2000 case. In summary, there are now three detailed and two rather short cases studies (see General Remarks above).

The demanded detailed analysis of the processes responsible for the elevated ozone would be rather difficult. I had strongly advocated forward simulations from selected “source regions” in order to visualize how the transfer occurs. I learnt that this requires considerable improvements of the existing modelling tools. Also, given the time spread of the plumes 15 to 20 days backward in time any reviewer would ask serious questions.

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Any major effort was made impossible anyway by the early end of the TUM group even before the end of ATMOFAST. Thus, I made a few attempts with forward trajectories that looked rather reasonable (mentioned for Case 1, but also carried out for other cases). Some field studies with air-borne instrumentation would be helpful.

I also do not think that an omission of the discussion of potential Asian contributions is a good idea. If we had dropped this topic one or the other reviewer (and at the end the community of the readers) would have complained for sure! As a matter fact, Reviewer 2 wants to see even more emission panels. The mixing of PBL and STT contributions in the vicinity of frontal systems is well known (see introduction). Thus, at least some attempt of estimating the Asian influence must be made. In fact, there are cases with and cases without co-existence of these different air masses.

Reply to specific questions:

(1) The paper says “midlatitude North America”. Cooper et al. (2006) conclude that lightning-induced ozone prevails in the the southern and eastern U.S. For stations further to the north an enhancement above 8 km due to STT is determined. For Trinidad Head this contribution is 27 %.

(2) Shallow does not mean thin! Sprenger et al. distinguish deep, medium and shallow intrusions. The shallow intrusions seem to be the most important ones accounting for about three times more STT than the deep ones. I added a few more sentences.

(3) Thank you for pointing this out! My statement on this issue in the Discussion is too short although I exactly alluded to it. This is now changed.

(4, minor comment) A few degrees are missing in these plots (as visible from the contour of East Siberia); this is the way I received them.

Review 2

General remarks:

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Second paragraph on p. S677: Some of the measurement series and a partial interpretation have, indeed, already been published before. However, as clearly follows from our text, the part of the observations we analyse here had not been understood at the time of the earlier publication. Nevertheless, this statement and the corresponding one in the third paragraph of the report urged us to make this difference even clearer. In particular, we rewrote the relevant parts of the introduction.

Third paragraph on p. 677:

- The number of case studies is adequate, as pointed out in the introduction. However, it is not necessary to describe the results for Cases 3 and 5 in all detail. Thus, we shortened these parts. In particular, Case 5 is highly important because of the high ozone mixing ratio, its length and the missing PBL contribution! The focus of Sec. 3 is now on Cases 1 (historically important series!), 2 and 4 which we believe to be essential. For Case 1 we did not find Asian contributions, for Case 2 we have strong indications of Asian dust, and for Case 4 there are layers with and without Asian contributions (and the highest modelled STT fraction). In order to reduce the confusion we added several sentences for guidance.

- As to the models, the FLEXTRA results shown for Case 4 are the best choice for demonstrating that there is a single coherent air stream at constant high altitude back to the Gulf of Alaska since the FLEXPART analysis gives just mean altitudes of a given cluster. Two trajectory figures are given to visualize the origin of layers L1-L3. The step-by-step extension to 15 and 20 days denonstrates that there is no significant upper-tropospheric branching, which is extremely important for relating the STT fractions to the subtropical jet. In general, the 15-day simulations are only presented because the important 12000-m option, unfortunately, is not available in the 20-day version of the program. I regret this degree of complexity!

- I have difficulties in understanding the comments “Figures from single cases are also not presented in a chronological order......” and “..discuss the observations

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and simulations in a separate section....": This kind of structure was almost strictly followed!!! We stay in a chronological order first for the data, then for the three types of models. In my view this is better than jumping from model to model for the individual examples. A guideline is now available at the beginning of Sec. 3. Quite a few improvements were introduced. For the complex description of Case 2 I introduced a few additional titles to separate the descriptions of the 20-day simulations.

First paragraph on p. S678: Considering our considerable effort in reducing the number of figures we hesitate to add meteorological maps. Four of meteorogically similar cases have been characterized before, including Cases 1-3 (Stohl and Trickl, 1999; Trickl et al, 2003). The focus has been on the reproducibility of the findings rather than on a full meteorological analysis. I added a few lines on the typical meteorological situation and the above references in Sec. 4.

Second paragraph on p. S678: We now add more information on and some discussion of the shallow transfer (mostly in Sec. 4, but also elsewhere). This was, indeed, insufficient!

Third paragraph on p. S678: The FLEXPART model has been described in numerous publications. We, therefore, decided to focus on what is needed for the analyses in the paper. However, Dr. Stohl re-arranged the FLEXPART description for more fluent reading. Since we have completely lost contact to Dr. James a clearer explanation of the 12000-m simulations is not possible. Since he prepared the FLEXPART version for the 15-d simulations they cannot be repeated and, thus, NO_x must remain the species of our choice in this case. Of course, from an experimental point of view, CO is the better tracer. However, the model runs do not include chemical ageing and the vertical distributions for NO_x and CO do not differ much in structure. I eliminated the NO_2 panels in the 20-day simulations. However, SO_2 is an ideal tracer for Chinese air. There are obvious differences with respect to CO and I think it is better to retain these

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panels. A remark concerning this was added.

A table containing, e.g., layer thickness and length of the individual episode was added as suggested. Quantitative numbers of the model results cannot be given since the model does not yield absolute concentrations except for the vertical distributions of emission tracers.

First paragraph on p. S679: I examined the manuscript concerning missing citations of figures and was unsuccessful. The demand to show more figures is in some conflict with Reviewer 1. Before the first submission I had tried very hard to shorten the number of figures. However, I added two more profiles of STT fractions since this facilitates the understanding.

Specific comments:

Second paragraph on p. S679: I added the requested information.

Third paragraph on p. S679:

- As stated two separate layers are seen, not only one. Just the lower one descended. It can no longer be seen in Fig.1. I added a few explanations. Shading the upper layer is unnecessary, all is now clearly described and and one aerosol profile was added to Fig. 1.
- The 12000-m chart is never named “reliable”. It just visualizes areas were STT might have taken place (as clearly stated). In quite a few situations a proof comes from the STT fractions with the more recent modelling tools of the 20-day simulations. We regret that not all tools have been available at all times. This would have helped us in simplifying the manuscript.
- The altitude interval of 5.5 to 5.75 km was chosen because the highest stratospheric fraction is seen here. We added a figure showing the vertical distribution of the maximum STT fraction that visualizes the much more extended range

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with stratospheric contributions, but revealing a rather spiky structure (modelling noise?). This figure clarifies that the 5.5-5.75-km bin belongs to the layer under consideration.

- Neither TOMS nor MODIS images are available for that period. From my experience I would expect elevated aerosol in the presence of smoke!
- I added an aerosol measurement to Fig. 1 as requested. Unfortunately, I had to select one for an earlier time because the background in the 19:00-CET data is noisier. The altitude of the aerosol layer did not change much during the second half of the day.
- As stated the model results did not suggest any major North American influence below 8 km

First paragraph on p. S680: As stated I verified that there is an intrusion for coordinates in the Kamtchatka region. I did not calculate trajectories for the entire area exhibiting read colour. This was a tedious job and I had to limit my efforts. I had called for a more complete forward approach using FLEXPART, but this could not be accomplished due to the end of the TUM group.

Second paragraph on p. S680: This high-pressure zone, indeed, extended that far! I would love to present material on this. But this paper does not only cover a single case and there are certain limitations.

Third paragraph on p. S680: Here, just the observations are described. The analysis is given further below. There is no indication of major forest fires in the satellite images (I added a sentence).

Fourth paragraph on p. S680: The suggestions made here are partly in conflict with the first review that recommends shortening of the PBL material. I am not happy with

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the considerable amount of detail demanded which would be even more confusing for the reader.

- At the time when the 15-day simulations were made (2004) we decided to go for the whole layer. The model was not yet set up for an analysis in the 250-m bins as available in the more recent operational version. As mentioned, these simulations are used in our paper mostly to visualize the intersection with 12 km, which looks remarkably coherent despite the large time-altitude box chosen. This option is not available in the operational 20-day version of FLEXPART.
- The source contribution plot for the example given in Fig. 8 does not differ much from that in Fig. 7. The vertical distribution is given in Fig. 9 anyway!
- It is, indeed, difficult to determine the regions where STT took place. The high number of days with elevated STT fractions suggests that the source regions spread considerably along the path of the air mass. Also Siberia may be involved. However, there is no reason to become nervous about this source region not being part of the Pacific. This just verifies that the atmosphere is rather variable. It is astonishing enough to see all the similarities in quite a number of cases. As a matter of fact, I made a few adjustments in the abstract to clarify that we are not only discussing the Pacific. Furthermore, I added “and Asia” to the title since, after some more findings (Case 4) and some discussions with the Zürich group, I feel much more confident now about my interpretation of the branch leading back to North Africa.
- The peak at -4 d very likely corresponds to the input from eastern Canada and the North Atlantic. I added a short statement.
- I do not understand the remark “...it would be very important to divide the thick layer ....into more layers....”: This is is exactly what we have done with the new

tools used in the 20-day simulations! And it is reasonable to assume that I had carefully looked at the result for almost all individual 250-m bins, a rather time-consuming job! However, it is impossible to present all this information in a scientific paper without creating major chaos. The most important action during the revision has been to add more profiles of the STT fractions.

First paragraph on p. S681: We cannot distinguish between all these aerosol types. As state earlier we did make (and inspected) simulations for **all** altitudes and, therefore, also for 7.0 to 7.5 km! This has been the main instrument of our interpreation! I added a remark concerning the absence of fires in the vicinity of the tracer plume.

Second paragraph on p. S681: It is an excellent suggestion to skip the September-2000 case! The information content is not that high. Rather than eliminating this case completely I strongly shortened the text and removed the figures. It has been the focus of this paper to present results for a large number of cases to document the importance of these findings. In my view it is a rather special experience to find this kind of beviour in all relevant longer time series under similar conditions so far studied! Just a few sentences pointing out the most important findings are left. By the way, the Pacific is not the exclusive focus of this paper. “Pacific” originates from our finding that there is some air-mass import from the west coast of North America and beyond that needs clarification and obviously also covers adjacent regions. The title was, nevertheless, changed to include also Asia.

Third paragraph on p. S681: Thank you for this compliment! I think that this the most beautiful of the four-day series ever accomplished at our site. The interpretation is rather easy and I decided not to shorten much here. I even realised that this case is the best one to verify the importance of the branch leading back to North Africa (subtropical jet). This is hardened by the trajectories that demonstrate the absence of branching (and subsidence) all the way backward to the large red area in Fig. 17 (12-km panel) south of Alaska and subsequently by the retroplume (text added!). Although

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the STT results are so convincing I added the emissions figure I had removed earlier. Fig. 17 contains sufficient information to tell the reader that the transport pathways look similar to those in the other examples.

Fourth paragraph on p. S681: Here, the FLEXTRA results are clearly superior since they yield the exact altitudes of the air mass, not just an average altitude of a cluster. As just mentioned this (together with the FLEXPART results) also demonstrates the absence of a local intrusion south of Alaska. This is hardened by PV < 1 in the third panels (I added a statement).

Fifth paragraph on p. S681: Changed!

Sixth paragraph on p. S681: These figures were eliminated when I shortened the manuscript. Given the fact that I would like to stay with three main case studies I hesitate to add this information again.

Seventh paragraph on p. S681: I eliminated the emission figures during shortening. STT is clearly dominating in this case. Although Reviewer 1 suggested to drop all the emission panels I re-insert the figure containing the vertical distribution of emissions. This gives some idea on their importance. I hesitate to name any of these numbers "quantitative" because of both the limited accuray of the model and the uncertainties in the emission inventories (see "Discussion"). I also do not want to give STT/PBL branching ratios since the exact source regions of the respective fractions are not known.

First paragraph on p. S682: See above!

Second paragraph on p. S682: This really short case study shows that not all transport patterns are the same! But the differences are not really severe. STT clearly takes place over the Pacific, but also over the western U.S. The backward plume obviously does not travel that quickly and, therefore, does not fully reach the Mediterranean Sea. It is important to present the lidar data because the results are particularly spectacular (long duration of that episode and persistent high ozone values). Nevertheless,

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I slightly shortened this subsection further. In summary, three major case studies remain.

Minor comments and technical corrections:

I am astonished to see that, indeed, both latitudes and longitudes are missing and changed this! The backward times depend on the altitude and, therefore, are not unique. Thus, in the retroplume analysis the backward times are displayed in the panels titled “Retroplume Analysis”, inside the circles. I admit that the circles are somewhat small in the ACPD version, but these graphics were prepared for full-page printing.

2nd comment: I added β_P to the axis titles.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 3113, 2009.