

## ***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.***

### **Anonymous Referee #2**

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#### General comments:

This paper describes a number of cases where dry layers with elevated ozone mixing ratios were observed in the middle and upper troposphere over Garmisch-Partenkirchen with lidar and other measurements. The authors claim that these layers are related to a strong import of ozone-rich airmasses from the stratosphere mainly from a region over the Pacific by using FLEXTRA and FLEXPART simulations. Five case studies within the time period 1996–2001 are presented. However, several of these cases have already been presented in other publications by the authors Trickl et al. (2003, 2009a), at least to some extent.

The focus of the paper is well within the scope of ACP and interesting for the community. However, unfortunately the structure of the paper (especially the presentation of the results) is very chaotic. The paper needs to be rewritten and improved substantially before it can be published. The method used to analyse the ozone features (FLEXPART) is appropriate, however the description and interpretation of the results cannot be published in the present way. A number of suggestions to improve the paper are given below.

Already at the beginning of the introduction the reader gets the feeling that you already described everything in your 2003 paper. Point out more precisely what the reader can gain by reading the present paper compared to your 2003 paper, except that more cases have been added. What is new in your study compared to the other studies you mention in the introduction focusing on dry air streams with elevated ozone?

Several figures and results have already been presented in the previous 2003 publication. At the same time, many important features are mentioned and briefly described but not shown in any figures. Far too many case studies are presented (five) and the reader gets more confused than convinced about the message of the authors. Figures from single cases are also not presented in a chronological order and same manner from case to case. I would suggest to reduce the number of described cases to maximum two (e.g. at least 21-24 July 2001), present the same kind of FLEXPART-products for these cases (not FLEXTRA) in a more systematic way, discuss the observations and simulations in a separate section and try to understand the mechanism that causes these observations more in detail. Try to concentrate more on cases including Asian anthropogenic pollution, since these are more rare compared to North American pollution events in the literature. It is also very confusing since both 10, 15, and 20 days backward simulations are presented. Present only the most useful simulations (20 days) and also only simulations from FLEXPART which includes much more information than FLEXTRA simulations. Point out the importance of using 20 days simulations to understand the observations more precisely.

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It is mentioned that all the presented cases starts one or two days after the onset of high-pressure periods during the warm season. Add also some meteorological maps showing the development of a typical pressure situation over Central Europe causing these events.

In the Abstract you write "shallow transfer from the stratosphere" and then "suggest an important mechanism for stratosphere-to-troposphere transport": The reader of the paper expects to find out more about this "new" mechanism. However, an explanation for this shallow transfer is missing. Perhaps you are looking for mechanisms described by Sprenger et al. (2003), Cooper et al. (2004a-b) or by Liang et al. (2007), as you mention briefly in the introduction? It would be worth to describe this mechanism in more detail in your paper and try to understand what is happening over the Pacific. What can your observations and simulations contribute to understand this mechanism better?

In Section 2.2 you describe the FLEXPART model. This section needs improvement. Add a brief general model description. Describe all (!) the different FLEXPART products more systematically and in detail. The chart "12000 m" (no FLEXPART "standard" product) also needs some explanation here. Use also the same names as given on the plots shown. Describe the products in the same order you present them later. It would perhaps be more useful to use the product "footprint emissions sensitivity with CO" (instead of NO<sub>x</sub>) if you look for anthropogenic emission. Also reduce the vertical FLEXPART distributions to CO only (cut SO<sub>2</sub> and NO<sub>2</sub>, since no additional useful information is given).

I would also suggest to add a table where you summarise the characteristics of your high-ozone layers: altitude range and its change with time, vertical (horizontal) extension, ozone mixing ratio, relative humidity, aerosol backscatter coefficient, duration over Garmisch-Partenkirchen, source contribution (% strat., Asia, NA, EU, specific regions), air mass age since emission and so on.

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When you describe your observations also mention at the same time the figure that is relevant to what you describe. Show more figures related to the final selected cases (maximum two).

Specific comments:

Page 3118, lines 17-28: Add range, uncertainty and resolution of the aerosol backscatter measurements.

Pages 3122-3123: Here a period is described (29 May to 1 June 1996) where a stratospheric intrusion was observed together with a high-ozone layer transported from the BL of North America (studies already published). The latter layer was mainly observed above 8 km but then gradually descended down to 4 km. Are these layers also visible in Fig. 1? It would be more useful if you also shade all the interesting layers in Fig. 1. Then a third type of dry high-ozone layers was observed on 31 May, between 5-7 km, which could not be attributed to transport from the NA-BL. It is mentioned that this layer must have "a potential stratospheric air admixture from beyond North America". Fig. 2 gives some hints in this direction, however I am not sure how reliable the "12000 m" chart for this stratospheric source is, if the tropopause reaches 12 km which occurs occasionally in summer at 40°N. What about the small maximum in the "12000 m" chart over SE United States? In Fig. 4 you then present retroplumes, but for the interesting layer you choose an altitude 5.5-5.75 km which is more at the lower edge of the plume (ozone 70-80 ppb) and almost too low. Around 10-12 days backward you also have an enhanced contribution from higher "stratospheric"(?) altitudes (green dots), is that also already beyond the United States? What about an aged high-ozone layer from forest fires, which are also known to be very dry? Can you exclude this source (look at MODIS hot spots globally)? Why not also show your aerosol backscatter measurements in Fig. 1? How can you be sure that it is not an uplifted polluted layer from Mexico City (up to 270 ppb!, page 3124, line 20), since Fig. 2 gives some hints in this direction also for the 5-7 km layer?

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Page 3123, line 29: "big intrusion is seen in Fig. 3 between Kamchatka and about 40° N, 180° E": I see more an "extended area" with a large number of trajectories ending at 12 km. Is this a stratospheric intrusion extending from 40 to 20°N?

Page 3125, lines 18-19: "a long high-pressure zone formed that extended from the region south of Newfoundland to 30° E": Can a high pressure ridge really reach this extension? It would be very useful to show maps of the meteorological development (e.g. geopotential at 500 hPa).

Page 3126, lines 20-21: "rather remarkable for the free troposphere and corresponds to a visual range of less than 10 km": How can you be sure that this is not due to emissions from forest fires (westcoast U.S. border to Canada or Asia, footprint Fig. 7b)? Again you should check with MODIS hot spots. Later you speculate that it might be dust, why not already mention it here?

Pages 3127-3128: In Fig. 7 you show FLEXPART simulations for the whole layer 5-9 km on 27 May, which is really deep. Here you really get a very wide spectrum of contributing ozone sources (NA, Asia, Pacific). Would it not be better to show simulations for a narrower layer (e.g. 7-8 km) and also to divide the whole layer into regions with high and low backscatter (anthropogenic/stratospheric sources)? In contrast to Fig. 7, in Fig. 8 you show FLEXPART simulations for a very thin layer (5.75-6.0 km) on 26 May which also coincides with an enhancement in the aerosol backscatter coefficient (Fig. 6a). More information on the BL source is needed and the source contribution plot must be added to Fig. 8. It is also not clear from Fig. 8 where the stratospheric source is located. In Siberia? But this is very different from the Pacific focused on up to now. What about the importance of a BL source (mainly Asia?)? In Fig. 10 (lowest panel 7.0-7.25 km) from 27 May a very recent and large contribution from the stratosphere is seen (max. 4 days back). Was this intrusion located over Canada? Again it would be very important to divide the thick layer (5-9 km) in Fig. 7 into more layers to find out more about the different ozone sources and mechanisms.

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Page 3128, lines 27-28: "big aerosol peak in Fig. 6b and c between 7 and 8 km is caused by dust from the Asian deserts". Can you distinguish in your aerosol backscatter measurements between anthropogenic, forest fire and dust aerosols in general? If you speculate that this layer comes from the Takla Makan and/or Gobi desert, it would also be important to make FLEXPART simulations for this specific layer (around 7.0-7.5 km on 27 May).

Page 3129: "3.3 Case 3: September 2000": I would skip this case. The focus of the paper "on the import from the stratosphere over the Pacific" is not clearly seen in this case compared to the other two cases presented before.

Page 3130: "Case 4: 21 to 24 July 2001": The ozone lidar measurements shown in Fig. 14 are very nice and interesting. If you try to concentrate more on this case and explain it more in detail and try to understand the dynamics that causes the observed ozone pattern, you would have a very interesting case study to present.

Page 3131, lines 9-21: What is the benefit of showing FLEXTRA trajectories (Fig. 15-16) compared to the FLEXPART products shown later (Fig. 17)? I would concentrate on FLEXPART products and show these for the 3 different ozone layers (L1-3) in Fig. 14.

Page 3131, line 20: "most trajectories above 5 km stay at high altitudes and end over the Pacific (Fig. 16).": For the calculated time period the trajectories end over the Pacific, but it does not mean that they end there if you calculate further backward it time.

Page 3131, lines 22-29 and page 3132, lines 1-3: Humidity measurements from other stations and platforms are mentioned, however not shown. Perhaps you can also show some of these measurements when you reduce the number of your case studies?

Page 3132, lines 11-12: "PBL contributions from both North America and East Asia are seen": Quantify these emissions and add numbers like this to the suggested table.

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Page 3133, lines 12-13: "The vertical distribution of emissions for layer L3 (not shown) show only half the mixing ratios calculated for L1 and L2. But they include a much higher Asian fraction": Again show this simulation and quantify the emissions and add numbers like this to the suggested table.

Page 3133: "3.5 Case 5: August 2001": I would also skip this case. It is also not a very clear case since the majority of the trajectories ends over the United States.

Minor comments and technical corrections:

- Add longitude and latitude to the FLEXPART maps. It would also be very useful if the days backward in time are highlighted in the FLEXPART column plots.
- Fig. 6: add the axis for the aerosol backscatter coefficient.

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

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