Reply to the Reviewers´ Comments on

Elevated ozone in boreal fire plumes – the 2013 smoke season

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Thomas Trickl, July 30, 2015

Thank you very much for the valuable comments that show that several details must be clarified. The items of the reviews are printed in italics, the answers in normal text.

Anonymous Referee #1

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General comments: This paper by Trickl et al, presents and discusses four case studies concerning the appearance of atmospheric layers with elevated aerosol contents as detected by the aerosol Lidar working at the Garmisch-Partenkirchen (Germany).

Since several years, the lidar systems working at IFU provide a set of high-quality observations concerning aerosol, ozone and water vapor, very useful to investigate the transport of these atmospheric tracers in the free troposphere and the atmospheric processes affecting their variability.

Concerning the paper significance, the presented research is well within the scope of Atmospheric Chemistry and Physics. The most original findings is related to the possibility that the ozone increases observed during some of the aerosol plumes are related to Stratosphere-to-troposphere exchange and not to photochemical production by ozone precursor emitted by boreal fires. However, the authors relies too much to the "large-scale" HYSPLIT back-trajectories for interpreting and attributing the occurrence of ozone and aerosol layers. I think this represents the major weakness of this work that should be addressed or better discussed before publication to ACP.

I do not fully agree. HYSPLIT is used as just a tool to make sure that the smoke is really coming from Canada. No further claims are made. I admit that the description of the analyses was kept short since as many as four cases were selected to illustrate the considerable variability of the observations. In addition to a large number of trajectory calculations we also used satellite images and sonde data, and we add few words about this.

Moreover, more discussion should be deserved in commenting the implication of the achieved results to scientific progress. E.g., as stated from the authors at pag 13266, events like those presented in this work are not frequent (at least as deduced by Garmisch Lidar measurements): thus, why we should be concerned about that?

The paper contains a number of conclusions:

(1) The hypothesis of low- or moderate loss smoke transport from the boreal regions of North America to Europe is hardened

(2) There is no pronounced formation of ozone during the transport in the case of the July 2013 fires. This perhaps the first study of long-range transport of smoke with rather dense measurements of ozone in the free troposphere over an extended period of time.

(3) The role of the stratospheric air tongues slowly descending from remote regions studied in recent years is touched. Their role in propagating some of the smoke during that period is pointed out.

(4) A very special layer was presented with extremely low humidity that had presumably descended from the stratosphere over roughly two weeks, picking up a significant amount of smoke without a clear sign of mixing. This even extends the time span of our recent paper (2014).

We do not draw any conclusions on the frequency of such events. The lidar measurements have not be dense in time, before 2007 they had been even interrupted for many years. Since 2013 we kept a closer eye on events like this one and found another, less spectacular period in 2014. Our paper confirms that these smoke phases can last rather long, as seen previously. It is also not only important what we observe here. The impact of these fires to mid- and high latitudes in general can an important factor of the air quality.

The presentation quality is fair. However, too many figures are presented and they should be significantly re-arranged before publication. I recommend to make use of the supplementary material to show "ancillary" information and to improve the manuscript readability.

I have no problem with the number of figures. 15-20 figures are normal in full-size atmospheric papers (in many publications even multi-panel figures are shown) and have been routinely accepted in my case. There should not be a problem in an electronically published journal. Shifting information to an external appendix is not that favourable since this means an extra effort for the reader. I suggest to let the editor decide.

I am not sure what "re-arranged" means. If this refers to arranging the aerosol and ozone profiles for better comparison: This has been intended and explicitly asked for below the figure captions.

Finally, if I can make a personal comment, it is really a shame that the very valuable continuous in-situ measurements at Zugspitze peak have been interrupted.

I truly appreciate this statement. After 2005 the ozone trend changed, and continuing the measurements would be very exciting. Maybe there is a chance to resume the Zugspitze series, but I am sceptical about the Wank series.

Major comments: To attributing the origin of the aerosol/ozone/water vapor layers, the authors made a massive use of the HYSPLIT back-trajectories. However, the authors did not provide many details about simulations set-up (i.e. which meteorological dataset is used for input, at which spatial resolution?) or comments/quantification of trajectory uncertainities.

I am sorry about this: we have mentioned the uncertainties so many times that this point was forgotten here! However, the meteorological data set ("re-analysis") and the three-dimensional mode of calculation ("model vertical velocity") was mentioned! A summary on our HYSPLIT efforts is now available a new subsection "Analysis Tools". I have made almost 200 calculations with HYSPLIT for the smoke period in July 2013 with different approaches. Our experience with the "re-analysis" mode of operation has been excellent. The new chapter also gives information on the other tools used, i.e., satellite images and radiosonde profiles.

The figures showing HYSPLIT back-trajectory plots reported CDC1 meteorological file (e.g. Fig. 18-19). If the NCEP re-analysis data provided by the READY laboratory is used, they are characterized by 2.5 degree latitude-longitude on a global grid. I'm rather skeptical about the possibility that single back-trajectories initialized by this coarse meteorological data set can be able to diagnose the origin of the thin atmospheric structures like those presented by the lidar vertical profiles.

I am shocked about the coarse resolution of the re-analysis data mentioned here. Indeed, a closer look at the HYSPLIT web site yielded some confirmation. My experience HYSPLIT run with "re-analysis" has been excellent, in agreement with what is known from other re-analysis data sets. The results for the free troposphere have looked more reasonable than for better-resolved GDAS data that I use for preliminary analyses. For example, with few exceptions, STT layers could be related to air descending from 8-12 km.

I recommend that the authors re-calculated (at least) ensembles of back-trajectories for evaluating the robustness of the identified transport patterns and discuss the uncertainty related with layer attribution (as partially done for instance for the event on 13 and 16 July). In the case, the authors already done this "sensitivity" study, the results should be more clearly discussed in the paper.

As mentioned above very detailed sensitivity analyses had already been done in great detail before submitting the manuscript, including calculating ensemble trajectories. At least the fine-scale layer search was mentioned in the text! I added some more statements.

By Table 1, you listed 14 events. But, you described just 4 of them. The information from this table is used only sporadically in the paper. I would suggest to move it to Supplementary Material or to better discuss the table information (as an instance in the Section 5).

The table gives an overview of the findings. Do you want us to discuss a total of 14 cases? Four cases are almost too much, but necessary to show the considerable difference between the measurement days. I added a sentence on the dry layers listed in the table in the Discussion.

Specific comments Please use the nomenclature" equivalent black carbon" (eqBC) along the paper, because you used an optical method (MAAP 5012) for determining its concentrations.

The responsible co-author agreed: Changed!

Pag 13273: I'm not sure if "personal communication" can be used as a reference. A published result can be preferable.

A published result should be preferred indeed. But at the time of our analysis an exchange of information by e mail was the only choice. I do not want to hide my source, and this is the first time in 33 years of publishing that I get a comment like this. I contacted M. Fromm again, and he sent me a rather comprehensive unpublished table that can very likely be obtained from him by interested readers on request.

Pag 13274 (figure 4): it is difficult to clearly see the "specific broad hump" in the aerosol "structure". Several features are visible in the CO and aerosol variations. Probably, you should superimpose colored areas to clearly identify the periods possibly affected by the fire events and the dust outbreaks. Please consider the possibility to move this plot to the Supplementary Material.

I explicitly mention that the hump shows up during the entire smoke period. This period is defined. I now add the peak mixing ratio and its position for more clearness.

Pag 13275: please join in a single figure (e.g. plate A and B), Figs 5 and 6. This will allow the reader to directly compare the two profile. Line 10: "moderate ozone": looking at the whole profile, it looks that a minimum in ozone appeared between 5.5 and 8 km, thus not so "moderate": : : Line 15: please describe how behaves the radiosonde RH profile (maybe adding it to the ozone profile reported in Fig. 6). Moreover, a map with location of experimental sites would help the reader to better evaluate the spatial representativeness of radio-sounding stations. Line 18: I do not see the need to cite STE forecast (not used) in this context. Line 20: you should show these backtrajectories outputs (even in the Supplementary Material).

As mentioned a two-column arrangement of the ozone and aerosol figures was asked for when submitting the manuscript. 50 ppb is moderate (not low) at our site in summer! Lower values are only found for subtropical marine air. In winter we see 40-45 ppb.

The reason for citing the STE forecast is that this tool would be better than HYSPLIT as it is based on PV (now explained). Quite e few people know about this and could ask.

A map would be nice, but would lead to an additional figure. In the new section “Analysis Tools” the stations used, as well as their directions and distances are given. The position of Garmisch-Partenkirchen is now marked in Fig 1.

Line 20: I could make the related plots available, but I want to avoid to avoid more than 20 figures as well as supplementary material. The aerosol layer is more important. Thus, Fig. 7 was chosen.

Pag: 13276, line 1-10: here the authors admit that a slight change of back-trajectories initialization (time or altitude), change the analysis result (no fire influence). This is a hint for uncertainty in the attribution of air-masses path and origin. A more sophisticated analysis (e.g. calculation of back-trajectories ensemble) is required to better assess this point). Section 4.2.2.: Again, merge Fig 8 and 9 and show the RH profile. Also show back-trajectory analysis. This can be valid for all the case studies.

Ensemble trajectory calculations had been done and did not change the interpretation. As said I have made almost 200 trajectory calculations with three and more trajectories, forward, backward and ensemble. This now mentioned in the new section on the analysis tools. We just give a few examples of trajectory images since the source regions Canada and Alaska are reproduced again and again: And there the fires were really burning! The general flow pattern is confirmed by satellite images.

RH profiles: This is in principle an excellent suggestion, we have done this in the past. I tried, but found that the two RH profiles add confusion, even when shifting them by +100 % RH. An explanation would require a lot of words.

Merging figures: see above!

Section 4.2.3: what do you mean by “HYSPLIT detects the layer exactly at 3.2 km, but locates the import from the zone of fires slightly above this”? HYSPLIT do not provide information about aerosol contents. lease explain better in the text. Line 21- 26: are HYSPLIT transport simulation enough reliable? Also these sentences, point out the necessity of better discussing and quantify the uncertainties related with these back-trajectory results: : : Line 30-31: please quantify the RH values seen by radiosoundings.

We changed this part, also preferring a more careful statement about the absence of smoke above 3 km at 11:00 CET. The new text makes clear that there are, indeed, limitations: The trajectory material is very clear for the early morning, but becomes very complex at 11 a.m., too complex for conclusions (especially the ensemble calculations that I made a few days ago to see the full spread).

Pag 13278: the pictures by Figure 12 and 13 are really impressive but I would move them to Supplementary Material. Line 21-23: please provide a reference where it is possible to see a typical stratospheric intrusion layer development. Figure 14. I’m not a Lidar expert but there are no possibility to separate fire from PBL fine particles? This would help in assessing the possible intrusion of the BB plume to the PBL: : :

The two figures infer some idea about the impression of the plume that cannot be easily described by text. One does not see too many photographs in scientific papers, but they exist. If the editor votes for including less figures in the paper this suggestion will be adopted. As said: I try to avoid supplementary material.

It is possible to distinguish particle sizes with lidar systems. This requires using multiple wavelengths. Our three-wavelength aerosol lidar (high-spectral-resolution lidar) has been out of operation due to the lack of funding.

Pag 13279, line 4-5: please provide a reference. Line 14-24. It is rather complicated for the reader to match the different layers in aerosol, ozone and water vapor profiles. E.g. you mentioned low humidity in "two partial plumes". What do you mean for "partial"? To provide the exact altitude of the layers can help the reader in identifying what feature are you discussing: : :

Reference for our observations of orographic lifting (lines 4-5): Unfortunately, I have just given a single example in a conference paper, although we have seen this many times.

Two partial plumes: We now provide the exact altitudes.

Pag 13280. Please, merge Fig 18-19 in one figure (plate A, B). Did you try to calculate forward trajectories from the end-point of the backward trajectory calculated on 9 July 2013 to evaluate if you return to the starting location of the back-trajectory?

Merging: See above!

We had calculated both forward and backward trajectories as well as ensemble trajectories to make sure that our statements are robust. The result was very convincing indeed. I add a few statements, also including information derived from CALIOP.

Pag 13281, line 13, I would change "correlation" with "link or relationship". Line 14. This sentence is rather "strong". I would specify that your results can be valid for the presented case studies.

Changed to "strongly related"! Lined 14: changed!

Discussion and conclusions: I think that to refer to intrusion "Type X", do not really add important information to the paper. Line 7 -12: this sentence seems a little bit out of place: you did not investigate STE seasonality in this paper.

"Type 6" just refers to our earlier study and, therefore, makes sense: I detected this type that looks somewhat different from the better known intrusions rapidly descending from the stratosphere above Greenland as it descends over many more days. The role of the rather frequent Type-6 intrusions in exporting boreal fires is an important result of our paper. I added one sentence emphasizing this high frequency that was particularly high during the first half of 2015.

Anonymous Referee #2

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The authors present a set of very interesting measurements showing the influence of North American boreal fires on ozone and aerosol in the free troposphere above a high elevation site in Germany. The title gives the impression that the elevated ozone they observed is the result of biomass burning, but the thrust of the paper is quite the opposite. A more appropriate title would be something like "Elevated ozone in boreal fire plumes-influence of descending stratospheric air"

The title looks neutral, and it looks interesting because of the possibility that an influence of biomass burning was found! The only change I would accept would be to replace "Elevated" by "Stratospheric", but this does not fit well to the second part of the title that indicates the extended period of time.

The measurements are of high quality, but the interpretation is weakened by the tendency of the authors to expect too much from HYSPLIT back trajectory calculations.

This is not true: The trajectories are used just for roughly verifying the source regions. In one case we go beyond this, but we explicitly state that we would not be convinced about our conclusions without the H_2O measurements! We now clarify our view in more detail. We also add statements about some of our satellite analyses.

I don't question the conclusions, but the validity of any individual 315-hour (13+ days) back trajectory is highly questionable, particularly if it is calculated using the low resolution (2.5_ x 2.5_ horizontal) Reanalysis data. Extending the HYSPLIT trajectories by running a second set as in Figure 19 seems particularly dangerous. The "astonishingly coherent" trajectories in Figure 18 are most likely due to the low vertical resolution of the Reanalysis model, which is confined to standard pressure levels (i.e. 1000,925,850,700,600,500,: : :) that are spaced much greater than the 100 m differences used in the calculations. The interesting profiles from July 13 are indeed consistent with differential transport that shifts from polar air with a stratospheric influence, to marine-influenced air from the Caribbean boundary layer, but is unlikely that HYSPLIT can resolve these narrow layers. It is unfortunate that the water vapour lidar wasn't operational, since any marine boundary layer associated with the "ozone hole" should also have had high water vapour.

As pointed out in the first reply we have not been aware about the coarse resolution of the re-analysis data field. In contrast to GDAS the specifications are difficult to find. Over the years I have analysed more than more than one thousand measurements with HYSPLIT in the re-analysis mode (Trickl et al., 2010; 2011; 2013; Fromm et al., 2010). With few exceptions specific layers in the free troposphere could be reasonably explained and distinguished from adjacent layers. In particular, STT layers (verified by strong descent) could be rather be rather routinely resolved, as expected from trajectories based on other re-analysis models. Some details were added, and the general aspects summarized in a new section "Analysis Tools".

The statements about the Caribbean influence was revised: the behaviour of the trajectories for that layer at 11 CET was found to be too chaotic. The missing of H_2O data from our lidar is, indeed, unfortunate, but the results for two sonde stations around the flow direction shows RH values around 50 %. We added a sentence on this fact.

More robust (and convincing) results might be obtained using HYSPLIT with the higher resolution GDAS1 model (1_x1_ horizontal and more vertical levels) run in ensemble or matrix mode. The coherence (or lack thereof) seen in ensemble runs will provide a good idea of how many hours back the trajectories can be trusted. It would be much better to compare the measurements with FLEXPART. Satellite water vapour imagery might also be useful.

I have done comparisons with calculations based on GDAS1, and also found some coherence. However, despite the higher resolution, the results looked less convincing. I think this could be due to a different way of generating the meteorological data, just with short delay. In the July 16 case, the GDAS1 trajectories start to deviate from the re-analysis ones south of Greenland and pass north of mainland Canada. They are too short for identifying source regions further to the west. However, I traced the plume with images from the CALIOP space-borne lidar. The best co-incidence in space and time was found over the Québec province incidence. The corresponding image shows two thin aerosol layers right where the re-analysis trajectories pass. I added a few statements.

Style suggestion: It is common to read of "surprising" or "unexpected" results in the literature, but rare to come across the words "frightening" and "astonishing". The latter is used no less than three times in this manuscript. The authors might consider substituting more restrained adjectives (e.g. "unusual") in their text.

Thank you for this suggestion, changed!

A few more specific points.

P13266, L27: Increased NOx in transported fire plumes has been attributed to thermal decomposition of PAN, not photo-decomposition.

Thank you, changed!

P13268, L 26: A visual range of 1500 km is a meaningless quantity since humans can’t even see 313 nm radiation.

Changed to the correct expression “standard visual range” that is defined by a drop in radiation intensity to 2 %.

P13282, L15: Wild fire plumes often contain significant quantities of water vapour volatilized from the fuel source.

Thank for this remark! However, this does not make the interpretation easier.

P13281, L1: There is no obvious increase in the in situ ozone in Figure 20.

There is a short 8-10-ppb hump starting at the broken line. The increase in ozone in the lidar data is also not very impressive. Ozone is not always a strong indicator of STT, but H_2O is (Trickl et al., 2014). I explicitly give this number in the text now.

Figures.

It would be useful to have the surface ozone data added to Figure 4 as it is in Figure 20. Also, the different multiplicative factors make the plot hard to read. It could be improved by adding a right-hand scale for CO, and arrows or vertical lines to mark the times of the ozone and aerosol profiles plotted in the other figures. Perhaps a second panel could be added here that expands the July period discussed in the text?

I added the ozone curve to Fig. 4 and found normal summertime values during the smoke phase and also other periods, but lower ones in the Saharan dust period since this implies air from lower latitudes. The ozone curve is deceiving and and reduces the quality of the figure. We prefer to omit it, but add a sentence.

Indeed, a second scale for CO would be helpful for those readers who know typical mixing ratios. On the other hand also a new scale adds complexity and the panel for the graphs becomes narrower. I think that a multiplication by a factor of five is not too difficult!

The information in Figures 5/6, 8/9, 10/11, and 15/16 would be easier to digest if each ozone profile was plotted together with the corresponding backscatter profile on a different horizontal axis. Each Figure could show four narrower plots. This would make it easier to see the relationships between aerosol and ozone. The ozone scale could be truncated since the stratospheric part above 12 km is not germane to the discussions.

This is explicitly asked for in the submitted version of the manuscript. Therefore, we will keep an eye on a two-column arrangement in the proofs.