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Transport of Biomass Burning Aerosol into the Extratropical Tropopause Region over Europe via Warm Conveyor Belt Uplift

P. Joppe et al.

Author comments to Reviewer #3

The reviewer comments are written in this font style and color.

Our answers are written in this font style and color.

Changes in the revised version of the manuscript are written in red.

The above manuscript describes aircraft-borne aerosol particle measurements conducted in the upper troposphere and lowermost stratosphere over Europa. The analysis focuses on a short time period of one of the TPEx campaign flights, when the outflow region of a warm conveyor belt (WCB) was probed and influence from biomass burning was found. Overall, the manuscript is well written, the set-up with towed sensor shuttle is relative unique, and the supporting meteorological data are very useful. In contrast to previous studies, the detected biomass signature stems from smaller wildfire events which reach the tropopause region due to the WCB and not by their own dynamics (i.e. as pyroconvection). As this is somehow new and could be relevant to the UT/LS chemistry or UT/LS influence on the radiation budget, the manuscript should be published. However, there are a few questions concerning the data processing and the interpretation of the results. Moreover, the prove of the relevance of such kind of events is missing, which diminishes the value of the results (are they relevant or not?)

Thank you very much for the detailed reading of the manuscript and the positive consideration of the topic for publication. Thanks also for the detailed questions and comments which help to improve the manuscript.

Specific remarks:

p. 1, l. 9: I can see the "800 particles per cm3" in Fig. 3, but for me the background is about 200 particles per cm3, hence at least a factor of four lower, not two.

We checked the exact values in the time series. The pollution shows an enhancement of 2.5 compared to the UTLS background and up to 4 compared to the tropospheric background. For clarification we changed the phrasing to:

This is higher by a factor of more than two compared to the UTLS background and up to a factor of 4 higher than the tropospheric background.

p. 2, l. 24: the sulfate aerosol and the BC you refer to, which one is it? The average total atmospheric amount? Please specify.

Thank you for the careful reading. Yes, we refer to the average total atmospheric amount. We added this information to the revised version of the manuscript.

For example, at the top of the atmosphere (TOA), the global radiative effect of sulfate aerosol is a cooling effect up to about -1.3 Wm^{-2} whereas the global radiative effect of black carbon (BC) shows a strong heating effect of up to 0.9 Wm^{-2} (Masson-Delmotte et al., 2023; Kalisoras et al., 2024; Ramanathan and Carmichael, 2008; Räisänen et al., 2022).

p. 2, l. 35: Please cite not only references from your group, there are other publications which have shown which are related to your topic, for instance Brioude et al, Atmos. Chem. Phys., 7, 4229–4235, 2007 or Zahn et al., J. Geophys. Res. 105, 1527-1535, 2000.

We agree with the reviewer that the references should be neutral and not only from the own group. Thank you to the reviewer for suggesting us these publications, which were new for us. We added these references.

p. 3, l. 57: The sentence on the BC lifetime, this hold true for other particles as well. And after modification, please move it somewhere else (or delete it), as it does not fit to the rest of the paragraph.

We deleted this sentence in order to stay within the focus of this paragraph, which deals with OA.

p. 3, l. 74: You focus on a dedicated, short section of one flight. But looking at the flight pattern and having in mind that there was a second flight on the same day, did you check your data for the chance of having probed the airmass a second time, later (or earlier) in the campaign? On page 15 you suggest that partly probing the same airmass again have been the case.

Unfortunately, we were not able to fly the days before F07 to probe the air mass during uplift and WCB transport, due to airport closure over the weekend. On the following day, we conducted a flight, but with focus on PBL characterization and convective uplift over central Germany. Thus, this flight probed completely different air masses. Therefore, the only possibility of probing partly the same air mass is the already written suggestion.

p. 3, l. 78: The statement of the "highly variable tropopause altitude", well, it looks variable, but not highly variable, e.g. there is no tropopause fold. Please modify the statement or justify why it is valid.

We changed the statement to only a variable tropopause in the measurement region.

The goal of research flight F07 was to probe a region with a variable tropopause altitude (see Fig. 1a). As consequence, we expected enhanced cross-tropopause mixing as a consequence of a

low-pressure system over the North Sea west of Norway and predicted low Richardson numbers in the restricted air space (not shown).

p. 5, l. 97: Even if the particle size range which is most interesting for you is less prone to particle losses in your inlet system and sampling line, you must at least provide an estimate or an upper limit on the respective particle losses.

We added a corresponding sentence regarding the particle losses at the end of the inlet description.

In the measured size range of the UHSAS we calculated transmission efficiencies of 86 % at the boundaries and 95 % at diameters around 300 nm. These calculations were performed for an ambient pressure of 300 hPa and 240 K using the Particle Loss Calculator by von der Weiden et al. (2009).

p. 5, l. 105: Related to the point above, how about an in-flight intercomparison between the TOSS and the Learjet instruments? There must be a flight condition, where this should have been possible and this would highly increase confidence in the data quality. Similar to the radiation calculations later on, it is not sufficient to refer to a potential future paper.

We have an in-flight intercomparison between the TOSS and the Learjet instruments. This intercomparison as well as the complete characterization of instruments are described in Bozem et al. (2025) which is now available as preprint. Therefore, we updated the reference to the preprint.

p. 6, l. 108: Again, the statement on the upper inlet cut, which analysis is it based on?

We performed particle loss calculations by the Particle Loss Calculator by von der Weiden et al., 2009. For this, we transferred the inlet metrics into the software for calculation. For clarification we added the following sentence:

The particle loss calculation was done using the Particle Loss Calculator described in von der Weiden et al., 2009.

p. 6, l. 115: You state that you operated 3 CPCs, but you provide only two lower threshold diameters. Why?

To cross-check the data quality of the aerosol number concentration during the flights, two of the three mc-CPC channels were operated at the same dT and thus at the same cutoffs. For reasons of clarity, however, we only show one channel here.

p. 6, l. 120: If I´m not totally wrong, 5 km in 30 s would result in a TAS of 167 m/s, which seems to be very slow for a jet aircraft in the upper troposphere. Are you sure the number is correct?

Yes, this is right. The TAS is lower than for typical jet aircraft in the UTLS. This comparatively low air speed is forced by technical constraints when the TOSS is deployed and towed below the Learjet. The reported TAS is taken from the Learjet avionic data system.

p. 7, l. 154: I did not find the time resolution of the filter sampling, please provide this information.

The filter sampling has no fixed time resolution. The sampling period depends on the meteorological and expected conditions, such as troposphere, tropopause region, stratosphere or expected polluted regions from the CAMS forecast with adjustments based on the in-situ measurements. As we moved the SOAP analysis to the appendix, referring to Reviewer#4 a detailed overview over the filter sampling periods is provided in Table D1.

p. 7, last paragraph: I´m not an expert in this, but can you exclude particle changes during the storage times? Either way, could you please write a sentence, if these can be excluded (and if yes, why) or if they are of minor importance or …

Thank you for raising your concerns about artifacts that might occur due to the storage. It indeed can happen. However, here we want to refer to Resch et al. 2023 (https://doi.org/10.5194/acp-23-9161-2023,), who did a thorough study on storage conditions. Based on this we stored our filters below 0 °C in order to prevent reactions occurring on the filter or losses. More details can also be found by the added reference of Breuninger et al., 2025 which describes the filter sampling in more detail.

p. 9, l. 214: Same statement as in the first comment, I do not see this factor two, it is at least a factor of four.

We reformulated the sentence to the following, also referring to the reply to the first comment.

[…] because the time series shows a very small-scale pollution event with an increase in particle number concentration by more than a factor of two compared to the UTLS background.

p. 9, l. 225: If I´m not totally wrong a H2O mixing ratio of 100 ppmv is rather typical for the midlatitude summertime UT and not an indicator for stratospheric air, or?

Yes, this is correct and 100 ppmv is more upper tropospheric than lower stratospheric. We rephrased the paragraph to show the mixing of chemically stratospheric air (N2O and O3) with UT air masses (H2O).

The interpretation of the mixed air masses into chemically stratospheric air is also supported by the O3 mixing ratios above 150 ppbv and H2O values near 100 ppmv H2O.

p. 11, l. 250: Why do you provide the information that the flight pattern was flown after the TOSS deployment? Do you want to say, that the TOSS was not applied during that flight pattern? If so, please state it in that way. Same in the figure caption of Fig. 4.

This information is provided, because the TOSS can only be detached from the aircraft and operated in the restricted air space. Further, we want to express by this sentence that we analyzed the part of the flight during which the TOSS was already running and measuring. The additional information from the TOSS is needed to calculate vertical gradients of potential temperature over the whole pattern shown in Fig. 4.

p. 11, Fig. 5.: This figure puzzles me a lot. First of all, why is the first a delay in the AMS data and in the next peak the AMS is ahead? Secondly the strange looking UT/LS background volume size distribution, why are there jumps of 50% in relative narrow size bins? Is there an issue with the assumed refractive index of the particles? And the error bars are misleading, the measurement period is short, hence it would be much better to indicate the measurement uncertainty here, which should be some ten percent, I guess. Volume size distributions derived from OPC measurements are highly uncertain! The data behind this figure need a deeper analysis. The different colors in Fig. 5b are not explained in the legend.

Thank you for the careful reading of the figures in the manuscript. We hope we can deliver some helpful explanations to this figure in the following:

The delay in the AMS data or the shift of the data has two reasons which partly overlap. First, we averaged the AMS signal over 3.5 minutes with a running mean to reduce the noise and obtain signals above the detection limits. This is the largest factor for the shift.

Regarding the size distribution in Figure 5(b), the jumps are due to an adjusted bin scheme. We merged the measured 99 size channels to 9 channels, to account for the different refractive indices of the particles used during calibration (Bozem et al., 2025). This rearrangement produces jumps in the size distribution as there is no smooth transition from smaller to larger particles as in the high resolved measurements with 99 channels. The range of refractive indices used during calibration (1,34 - 1,39) and the new bin scheme allows for a better representation of real atmospheric aerosol.

Thank you for the consideration of taking the measurement uncertainty as error bars. We corrected the figure with the measurement uncertainties as error bars.

Possible sources for the observed high CO values in the stratosphere are advected biomass burning residues or local uplifted pollution from the surface. We analyzed the aerosol size distributions measured by the UHSAS at the Learjet in order to find some hints for possible biomass burning in these size distributions. Therefore, we calculated the aerosol volume distribution over 10 s, which is sufficient enough to average over the full peak of aerosol number concentration. Furthermore, we also calculated volume distributions during the local UTLS background between the two consecutive pollution events and the tropospheric background in the middle of the pattern without observed pollution. The detailed times are also given in Table C1. These volume distributions (Fig. 5b) show significant differences between the UTLS

background and the polluted air masses. While the UTLS background shows a constant distribution up to 500 nm followed by a decrease in larger aerosol particles, we observe a modal distribution with a mode between 200 and 400 nm during the pollution events. This observed mode is robust against the measurement uncertainty, with only small overlaps larger than 300 nm there the uncertainty is highest due to instrumental issues of gain stitching. Such a mode in the volume distribution has previously been reported for observed aged biomass burning aerosol (Alonso-Blanco et al., 2014; Ditas et al., 2018; Brock et al., 2021; Schill et al., 2022; Holanda et al.,2023).

p. 12, last paragraph: You argue here and in the following that you have at least up to the LMS a non-negligible amount of soot in your particles (see also Fig. 7). And in the troposphere it seems to be (for me) unrealistically high. This will definitely affect your optical particle measurements, was this considered in your data processing? Otherwise you cannot trust the distributions in Fig. 5b.

We agree that the amount of soot is very high. But we also want to emphasize that this is only a rough estimation of soot and a best guess with high uncertainties. Unfortunately, there have been no soot measurements during the TPEx mission. In the processing of the UHSAS data there is no chance of reprocessing with different refractive indices, but we calibrated the UHSAS with a large spread of refractive indices in order to adjust the bin scheme and cover many types of atmospheric aerosol. This calibration is done to better trust the measured size distributions. We reformulated the paragraph of BC calculation to emphasize the uncertainties of this method and discuss the likely too high concentrations.

The estimation of the rBC mass concentration is only possible, because both instruments measure in the same size range. We also have to emphasize that this estimation is only a best guess estimation with high uncertainties, based on several assumptions, and the estimated mass concentration has to be regarded as upper limit. The measurement uncertainties of the CARBIC-AMS are already in the range of 30 % (Canagaratna et al., 2007; Bahreini et al., 2009). The conversion of the number size distribution measured by the UHSAS to a volume distribution add a further considerable uncertainty. Since we used a difference between two measurements the calculated rBC mass concentration can in principle include other components, such as sea salt or mineral dust in addition to rBC. In the remote atmosphere, mineral dust and sea spray are typically found in the coarse mode above 1 μm diameter, although some contribution to the submicron aerosol mass has also been observed (Brock et al., 2021). Furthermore, BB aerosol may also contain non-refractory salts such as KCl (Dang et al., 2022). The gained amount of rBC is too high for real atmospheric values in the order of less than 5 % (Yu et al., 2019). Therefore, this approximation can be regarded only as upper limit, but is consistent with the observed enhanced CO and the SEM particle analysis. The information on black carbon may serve as an indicator of particle origin, especially regarding biomass burning. Furthermore, the estimated rBC fraction is influenced by an instrumental drift of the CARIBIC-AMS in the beginning of the flight due to short preparation times with respect to reducing the background in the vacuum system. For the estimation we assume that all refractory aerosol that is not detected by CARIBIC-AMS is composed of black carbon.

p. 16, Fig. 8: I understand the "normalized by sample volume" on the y-axis (please remove the empty parenthesis), but I do not understand the "normalized to the flight blank and sampling time" in the figure caption. And these are two different statements, right?

Thank you for the careful reading, here was a small error in the formulation. We reformulated it to the following and adjusted the figure.

…signal of the flight blank was subtracted and the measurements were normalized according to the sampling volume…

p. 16, l. 319: The interpretation of the particulate BB tracers, what do the results indicate? That there have always been BB influence during the sampling period?

The results indicate that the tracers were confirmed as well by UHPLC-HRMS, by using authentic standards. Here, the tracers were identified using authentic standards. The results therefore indicate a slight increase of BB-tracers for F07 Filter2, though it is correct that the sampling time was not sufficient to get extremely good signals and moreover significant differences. The main issue here is the time resolution of the filters, which is not precisely able to catch a 2 min BB-event. However, we wanted to highlight the broad variety of confirming instrumentation that we had onboard, strengthening our analysis.

p. 16, l. 323: The short lifetime of levoglucosan is how long? Please provide this number in the text.

We added the lifetime of levoglucosan to the main text, which is between 0.5 and up to 4 days according to Hoffmann et al. 2009 (https://doi.org/10.1021/es902476f).

p. 19, l. 374: The whole paragraph. Either you show results of the radiative impact here or you give an estimate on how frequent such events might occur. Otherwise the value of your results is hard to estimate, i.e. are they relevant.

In this paragraph we describe the possible radiative impact based on the measurements of the vertical gradient of potential temperature.

Furthermore, during the time of the review process another publication by was submitted (Khaykin et al., 2025) which also describes the uplift of BB pollution, showing this relative new pathway of biomass burning aerosol uplift. In contrast to our observation. Khaykin et al. show the local uplift close to the fires, but they also show that this process is relevant, especially for smaller biomass burning events with not sufficient energy for self-lofting of the pollution. A radiation simulation for our observation is still planned, but this is still work in progress.

We added the discussion of the new publication in this section to show the relevance of this process:

Referring to the observations by Ditas et al. (2018) typical heating rates as consequence of rBC are in the magnitude of 0.07 $K \cdot d^{-1}$ up to 0.44 $K \cdot d^{-1}$ in extreme cases, which shows a

significant contribution to the radiative feedback on lower stratospheric dynamics. Combining the uncertainties of the rBC approximation and the gradient calculation of potential temperature (31 %, see Bozem et al. (2025)) we end up in the same regime of expected heating rates of roughly 0.1 $K \cdot d^{-1}$ of the tropopause region after the WCB uplift.

[…] As the transport of rBC and BB plumes was mostly studied in the presence of pyroconvection and fast uplift of pollutants to the tropopause region, current studies like this and that of Khaykin et al. (2025) show an additional transport pathway towards the UTLS. This uplift can occur close to the fire locations like in Khaykin et al. (2025) or after low-level long range transport far away from the pollution source as shown in this study.

[…] Finally, we were able to show an additional pathway of BB pollution into the extratropical tropopause region by WCB uplift. In contrast to other studies (Khaykin et al., 2025), we observe this uplift mechanism after low-level long-range transport.

p. 20, Fig. 11: Again I doubt the BC fraction of 40% at the lowest flight level. I assume that there must have been other particle material like dust.

We agree that the fraction of BC is much too high. As mentioned earlier we reformulated the part of the BC estimation to emphasize the uncertainties of this method and that this fraction is the highest possible fraction which is possible without any other particle types.

p. 28, Fig. D1: The first too low AMS mass concentrations, could these be caused by the AMS warm- up time?

Yes, this is correct. Unfortunately, the warm-up time during this mission was rather short, compared to other measurement campaigns. Therefore, the first part of the flight the mass concentrations are lower which also increases the uncertainties in the soot calculation but can not be responsible for the whole difference. This discussion is also added to the main part of the manuscript in the paragraph where we introduce the soot estimation.

Technical corrections:

p. 3, l. 76: Please change to "The TOSS and the aircraft were equipped …"

We rephrased this sentence as suggested.

p. 3, l. 81: Please specify what is meant with "stratospheric intrusions which increase during the flight", are they reaching deeper into the troposphere or are they covering a larger area or do they occur more frequently?

[…] which are growing in spatial extent during the flight (green patches in Fig. 1b).

p. 4, Fig. 1: The flight path is displayed in red, not in black, as stated in the figure caption.

Thank you for this note, as it was a change during submission, we corrected it in the caption.

p. 6, l. 132: Please provide a reference publication for the instrument and the uncertainties.

We added the requested reference, which is Müller et al. (2012) and Kunkel et al. (2019).

p. 7, l. 149: The first two sentences of this paragraph provide partly the same information. Please remove this doubling.

Removed as suggested.

p. 8, l. 208: The information that you use N2O for defining the chemical tropopause is already given in line 203. Please remove one of the two sentences.

Removed as suggested.

p. 9, l. 211: This subsection is quite long, do you see any chance to split it? This would make it easier for the reader to follow your line of arguments.

We divided this subsection into 2 subsections to make the reading easier.

p. 9, l. 235: The "recent particle formation event" could it be an aircraft plume encounter? Did you check for instance flightradar24 for such a possibility?

We did not check this event in detail, because it is not focus of this study. But we agree that this is an interesting feature and we can not exclude that this might be an aircraft plume encounter.

p. 10, l. 237: "the chemical stratosphere" is probably not the correct term (what would this be?), you mean "chemically stratospheric air".

You are right, we mean chemically stratospheric air and adjusted this formulation in the revised version.

p. 11, Fig. 4: Please provide the particle size range information in the legend of Fig. 4a.

We added the size information to the axis label of Fig. 4a.

p. 13, l. 285: You refer to Fig. D1 and discuss it here in the main text. Consequently, the figure should be shown here. Same for figure F2 later on.

As suggested, we moved both figures into the main part of the manuscript.

p. 17, l. 346: The half sentence "and the biomass burning pollution trajectories" does not fit here and seems to me remnant from a former test version.

Thank you for the careful reading, as you are right, we corrected this term.

p. 18, l. 359: Please change "aerosol number concentration" to "aerosol particle number concentrations".

Changed as requested.

p. 21, l. 407: "the chemical ... stratosphere" is wrong again, you mean "in chemically stratospheric air".

As written above, we rephrased this formulation.

p. 21, l. 429: Please exchange "into" with "on", otherwise the sentence does not make sense.

We changed the phrasing to the requested formulation.

p. 28, l. 477: there is a space missing in-between "gcm" and also in ".5min" in the next line.

We added the missing space in the unit.

p. 31, Fig. F1: the red line indicating the back trajectories are hard to see over the orange/brown background. Please use a different color, e.g. bright green.

We changed the color to a bright yellow.

In addition to the requested revisions, we reformulated the following paragraphs in the manuscript in order to make some statements clearer and strengthen the analysis on request of one co-author (line numbers according to the track changes document):

- In the main part of the manuscript, we replaced TOSS by TPC-TOSS to be consistent with the now included manuscript by Bozem et al. (2025).
- Line 33: lowermost stratosphere (LMS)
- Line 35: There are several additional processes which influence the chemical composition and other properties of the aerosol on shorter timescales and more locally, such as convective events, planetary and synoptic scale waves, associated with baroclinic instabilities and vertical transport from the PBL to the UT ahead the surface cold fronts by warm conveyor belts (WCBs). These processes often generate strong shear, thus favorable conditions for turbulence and mixing (Zahn et al., 2000; Brioude et al., 2007; Kaluza et al., 2021, 2022; Lachnitt et al., 2023)
- Line 137: For the simultaneous measurement of the trace gases nitrous oxide (N2O) and carbon monoxide (CO) the Quantum Cascade Laser based spectrometer University Mainz airborne QCL Spectrometer (UMAQS) is used (Müller et al., 2015; Kunkel et al., 2019)
- Line 215: The tropopause height is highly variable in time and space and depends further on the used definition. During summer months the dynamical 2 PVU tropopause tends to be lower than thermal WMO tropopause or the PV-gradient tropopause (Kunz et al., 2011; Turhal et al., 2024).
- Line 384: In contrast to the pollution trajectories, those trajectories, which indicate pristine UTLS background, are also crossing Canada and regions with active wildfires, but in higher altitudes (Fig. 9b and Fig. 10).
- Line 390: In addition to differences of the altitude of the trajectories, we observed differences between upwind velocity during the uplift process into the UTLS.

Additional References:

Bozem, H., Joppe, P., Li, Y., Emig, N., Afchine, A., Breuninger, A., Curtius, J., Hofmann, S., Ismayil, S., Kandler, K., Kunkel, D., Kutschka, A., Lachnitt, H.-C., Petzold, A., Richter, S., Röschenthaler, T., Rolf, C., Schneider, L., Schneider, J., Vogel, A., and Hoor, P.: The TropoPause Composition TOwed Sensor Shuttle (TPC-TOSS): A new airborne dual platform approach for atmospheric composition measurements at the tropopause, EGUsphere [preprint], https://doi.org/10.5194/egusphere-2025-3175, 2025.

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