

ACP manuscript acp-2025-1346

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.

Review of the revised ACP manuscript acp-2025-1346

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

By Philipp Joppe, Johannes Schneider, Jonas Wilsch, Heiko Bozem, Anna Breuninger, Joachim Curtius, Martin Ebert, Nicolas Emig, Peter Hoor, Sadath Ismayil, Konrad Kandler, Daniel Kunkel, Isabel Kurth, Hans-Christoph Lachnitt, Yun Li, Annette Miltenberger, Sarah Richter, Christian Rolf, Lisa Schneider, Cornelis Schwenk, Nicole Spelten, Alexander L. Vogel, Yafang Cheng, and Stephan Borrmann.

The revised manuscript improved, but there is still a small number of issues which are not well addressed. One of them is major and request some bigger change.

Specific remarks:

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.

You state that you added the Brioude et al. reference, but it does not show up in your list of additional references. As I could not find the revised manuscript, I could not check if it is listed there.

We are sorry that you did not had access to the revised version. We checked it again and it is included in the revised version. Further, we include it in the additional reference list below this author comment.

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

I fully understand your answer and your reasons, my request is to let the reader know your arguments (add it to the text), otherwise the reader will puzzle as I did.

We added it accordingly to the new revised version.

Here, we decided to operate two of the three mc-CPC channels at the same cutoffs, to cross-check the data quality of the aerosol number concentration during the flights.

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 ...

Again, your answer is good, but please provide this information to the reader and change your text accordingly. (As I said, I did not have the revised manuscript, so sorry, if you already did this)

It was added to the revised version, which was unfortunately not available to you.

After sampling, the filters were sealed in aluminium foil and stored in a freezing box to minimize artefacts and losses of the collected aerosol particles (Resch et al., 2023).

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.

I do not understand your averaging argument concerning the shift. Right averaging does not lead to a shift. Moreover, it would not explain why one peak is delayed and one is ahead, because it would affect both peaks equally.

We checked this data again carefully. These shifts seem to come from reduced instrument sensitivity due to short preparation time and ongoing background reduction. Therefore, the first peak is delayed. The second peak in our opinion is not ahead and is going more or less in parallel with the UHSAS increase. Nevertheless, we removed the AMS concentration from this figure to reduce the complexity and avoid possible misinterpretation.

Concerning Fig. 5b my comment was probably not specific enough. Of course there are jumps when you merge bins, what I meant I that there is no physical reason, why there should be such very narrow particle modes in your particle volume size distribution. I have seen this before in OPC data and they were caused by the wrong refractive index/particle shape assumptions. Hence I do not trust the shape of your volume size distributions, but admit that the relative amplitudes for the different regions could be meaningful (limitation: see below). As I assume that you do not like to carry out a more detailed optical analysis of your data, my suggestion would be to state this in the text. The shape of the size distribution (the ups and downs) are probably errors of the analysis procedure (wrong refractive index and/or shape), but the amplitudes show (somehow, see below) the relative differences between the different regions.

We thank you for the clarification about your comment and are sorry for the not satisfying answer to it. You are right that there is a huge impact of the refractive index on the size distribution measurements. We tried our best with calibrations and adjusted bin schemes to reduce these uncertainties. However, there are still remaining limitations and uncertainties. To account for those, we changed this Figure to show the uncertainty of the distributions due to undersizing as consequence of absorption processes of the aerosol particles. For this, we used the results published by Moore et al., 2021. They provide sizing uncertainties of the UHSAS with respect to different refractive indices and potential absorption by black carbon. We added these uncertainties to our measured size distributions and show the uncertainty range. As a result, the regions can still be differentiated from each other.

As optical particle detection reveals some uncertainties due to absorbing aerosol particles, especially when calculating volume distributions, we additionally provide an uncertainty range. For this, we use the sizing uncertainties for a UHSAS according to Moore et al. (2021). Based on in-situ observations and laboratory studies, they provide sizing errors for different particle types, especially for wildfire biomass burning aerosol. Following Moore et al. (2021), we calculated the uncertainty range for potential 3 % oversizing up to 20 % undersizing as consequence of light absorption by wildfire biomass burning aerosol particles. The inferred volume distributions (Fig. 5b) show significant differences between the UTLS background and the polluted air masses. Even though the uncertainty range is high, we observe a modal distribution with a mode between 200 and 400 nm during the pollution events. This mode is robust against the measurement uncertainty and can be differentiated from the UTLS and tropospheric background.

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.

The problem with the very high potential BC content was also raised by the second reviewer. I also still believe that even the statement of a “rough estimate” in your reply is wrong, these are

unrealistically high fractions and if they are not caused by the uncertainties in the two methods there was likely another aerosol material there. But this was not the focus of my question. What does not get together is that you state on the one hand a very high BC content in your particles, but on the other hand work with OPC response functions for non-absorbing particles only (as far as I could see in the Bozem et al, 2025 manuscript). Moreover, this BC contents changes with the region under consideration, hence even the relative ratios of the magnitudes in Fig. 5b are questionable.

Sorry for saying this, but this is a big, for me unacceptable inconsistency in you data and makes the absolute values questionable. This issue must be addressed somehow.

We understand your concerns. As mentioned above, we now included the uncertainty range of the size distribution measurements due to a possible undersizing of absorbing particles. Since we can not exclude other aerosol types than black carbon completely, we changed the terms “rough estimate” or “upper limit” of BC to “refractory aerosol, potentially BC”.

After these indications for potential BB influence within the tropopause region, we use the CARIBIC-AMS measurements of non-refractory compounds to estimate the amount of refractory aerosol (rA) which can also consists partly of black carbon (BC).

Additional References:

Brioude, J., Cooper, O. R., Trainer, M., Ryerson, T. B., Holloway, J. S., Baynard, T., Peischl, J., Warneke, C., Neuman, J. A., De Gouw, J., Stohl, A., Eckhardt, S., Frost, G. J., McKeen, S. A., Hsie, E.-Y., Fehsenfeld, F. C., and Nédélec, P.: Mixing between a stratospheric intrusion and a biomass burning plume, *Atmospheric Chemistry and Physics*, 7, 4229–4235, <https://doi.org/10.5194/acp-7-4229-2007>, 2007.

Moore, R. H., Wiggins, E. B., Ahern, A. T., Zimmerman, S., Montgomery, L., Campuzano Jost, P., Robinson, C. E., Ziemba, L. D., Winstead, E. L., Anderson, B. E., Brock, C. A., Brown, M. D., Chen, G., Crosbie, E. C., Guo, H., Jimenez, J. L., Jordan, C. E., Lyu, M., Nault, B. A., Rothfuss, N. E., Sanchez, K. J., Schueneman, M., Shingler, T. J., Shook, M. A., Thornhill, K. L., Wagner, N. L., and Wang, J.: Sizing response of the Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) and Laser Aerosol Spectrometer (LAS) to changes in submicron aerosol composition and refractive index, *Atmospheric Measurement Techniques*, 14, 4517–4542, <https://doi.org/10.5194/amt-14-4517-2021>, 2021.

Zahn, A., Brenninkmeijer, C. A. M., Maiss, M., Scharffe, D. H., Crutzen, P. J., Hermann, M., Heintzenberg, J., Wiedensohler, A., Güsten, H., Heinrich, G., Fischer, H., Cuijpers, J. W. M., and van Velthoven, P. F. J.: Identification of extratropical two-way troposphere-stratosphere mixing based on CARIBIC measurements of O₃, CO, and ultrafine particles, *Journal of Geophysical Research: Atmospheres*, 105, 1527–1535, <https://doi.org/10.1029/1999jd900759>, 2000.