Title: Organic aerosols mixing across the tropopause and its implication for anthropogenic pollution of the UTLS

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Response to reviewers

To begin with we want to thank all the reviewers as well as the editor for the time and effort, reading and evaluating our manuscript. We took all comments into account and tried to implement them in the best way possible (lines mentioned refer to the **revised, clean version**). In general, we want to emphasize that the main message of this paper is to highlight the possibilities that come with our measurement technique and the motivation to do more research on that. The literature of organic aerosol in the stratosphere measured via UHPLC-HRMS is quite limited and therefore there are a lot of things we can show but cannot explain without speculating too much. The vast molecular diversity, and highly complex processes need to be studied further, and more measurements are needed to find clearer interpretation of our findings. We understand and feel the urge to explain things, but of course we want to be certain when interpreting results. Here, we present a comparison between compounds found uniquely in stratosphere and troposphere, which, to now, is mostly an observation to be shared with the community. We hope that we have deeper insights and certainty in the future based on more measurements.

Referee #1 – response:

This manuscript presents results from aerosol filter samples taken at various altitudes that were then analyzed for organic compounds using chromatography followed by orbitrap MS. The mass spectrometer had sufficient resolution to identify atomic formulas. There was both a broad analysis for unknown compounds and a targeted analysis for a few compounds of interest, such as PFAs. These are some of the first measurements with this level of analytical detail of organic aerosol at high altitudes.

The manuscript is interesting and well written. I suggest some revisions:

*1) The science issue that I see is the discussion **near line 330** of the low oxidation state in the stratosphere. This is surprising, considering the long residence time in the stratosphere and that most of the organic aerosols there came through the upper troposphere, so in order to be less oxidized in the stratosphere they would need to be react, in an oxidizing environment, to reduce the O:C ratio. It is not impossible but*

surprising and I think unlikely. If true it is important. As correctly cited, Benoit et al. saw the same thing with a similar technique to this manuscript. However, Appel et al. 2022 (cited, but only for sulfate) found the exact opposite result with an aerosol mass spectrometer (AMS). They measured very high O:C ratios in the stratosphere, higher than the troposphere. An "f44" of about 0.28 (Appel et al, Figure 11) corresponds to an O:C atomic ratio greater than 1 (Aiken et al., EST, 2008). There is a similar discrepancy for H:C (Appel et al. and Ng et al., ACP, 2011). The stratospheric Appel et al. data on figure 5 in this manuscript would be almost at the embedded pie charts. Conference presentations of AMS data from the ATom mission agree with Appel et al. (https://espoarchive.nasa.gov/archive/browse/atom/DC8/AMS-60s). Either the AMS is drastically overestimating O:C or the UHPLC/electrospray is drastically underestimating it. Do highly oxidized compounds make it through your column? You don't have to solve the comparison to AMS in this manuscript, but it should be acknowledged better.

Thank you for pointing out the indeed surprising results of lower OSc in the stratosphere. We have two explanations for the contradictory results. First, we are aware that our offline technique might lead to losses of HOM for instance, which is not the case for online AMS measurements. Therefore, we agree that it is probably due to the measurement technique. Additionally, we expect that some freshly formed or transported aerosol might contribute to low OSc. In Appel et al. (2022) the assumption is made that the SOA is formed in the UT, getting slowly transported into the LS. We on the other hand investigated convective systems (especially F09), where aerosol is transported into the stratosphere quite fast without long residence times. The differences are not as distinct in F05 as in F09, supporting this hypothesis. To acknowledge this discrepancy and limitations of our technique better, we added the following in **line 362 and following**:

et al., 2023). ~~Since the stratosphere contains less water vapor than the troposphere and thus a decreased production of OH, the oxidative capacity differs for each region within the UTLS and is lower~~ Comparing our findings with Appel et al. (2022), who found higher oxidized compounds in the stratosphere than in the troposphere~~(Esler et al., 2001). The differences~~ , we want to highlight that the differences in $\overline{OS}_C$ are more pronounced for the convection hunting flight~~than for the~~ . In that case we expect the aerosols to have lower residence times in the stratosphere, compared to stable conditions and longer residence times leading to possibly more oxidation. For the fold, where the overlap of both compound groups is larger, ~~but~~ stratospheric compounds are still showing the tendency of ~~stratospheric compounds~~ appearing in the lower $\overline{OS}_C$~~.~~, indicating as well new formation in the stratosphere or mixing at the tropopause. As a limitation of our technique it has to be mentioned that uncertainties in the detection of highly oxidized molecules (HOM) derive from the offline filter treatment that may lead to HOM decomposition and therefore an under representation in our dataset (Bianchi et al., 2019).

There is some confusion about the Esler et al. reference. It is mostly about high OH concentrations in mixed tropospheric and stratospheric air rather than the concentrations in one or the other. More generally, relative (mixing ratio) OH concentrations are higher in the stratosphere than in the troposphere; absolute (cm-3) concentrations are lower in the stratosphere just because of air density. The diffusion

coefficient of OH is higher in the stratosphere than the troposphere, again because of air density. The overall impact on oxidation rate is complex, but the long residence time in the stratosphere implies a lot of opportunity for oxidation.

Thank you for this clarification. We removed this statement and rewrote the section in **line 363**.

2) I would like to see a little more discussion of the analytical methods for those with moderate expertise. What would you tell somebody who was an expert in a related field, maybe something like gas-phase atmospheric mass spectrometry, about the strengths and weaknesses of your technique? For example, are there certain classes of compounds where the blanks are significant and other classes where they are insignificant? What classes of compounds might be missed by the ionization method? Another example is my question above about how well highly oxidized compounds make it through the chromatography column. This doesn't have to have a huge amount of detail, just some major points.

We added some discussion about the strengths and weaknesses of our technique in **line 204 and 215**, as well comparison to AMS for a better understanding. Regarding highly oxidized molecules, a review paper by Bianchi et at. (2019) (https://pubs.acs.org/doi/10.1021/acs.chemrev.8b00395) describes the difficulties when measuring HOM due to offline filter sampling. In principle, our set up is able to measure stable HOMs and we do see compounds such as C9 H14 O7. However, the filter treatment may lead to losses such as of highly reactive peroxides, which are prone to occur in HOMs. To incorporate this limitation, we added the following in **line 367**.

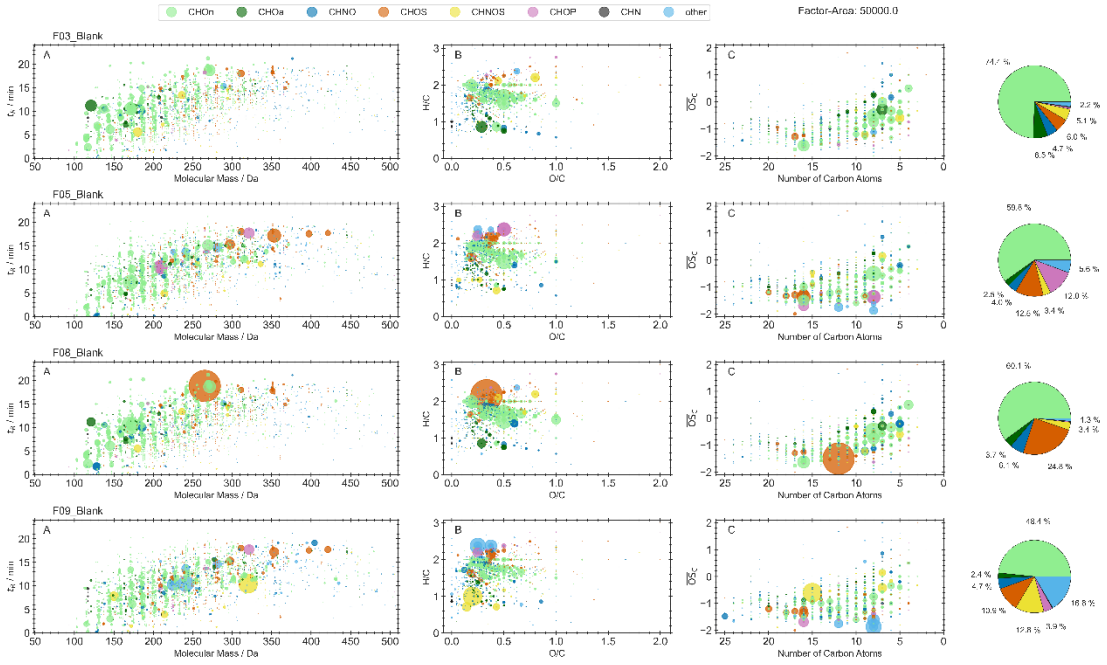
other). The limitations of this setup are derived by the solubility of compounds and the ionization efficiency. Using HESI, only compounds that carry a basic or acidic functional group can be ionized, neglecting compounds such as polycyclic aromatic hydrocarbons (Venter, 2024). The solvent used for extraction and the mobile phase determines detectable compounds as well. The C_{18}-column limits how well compounds can be separated, leading to a poor separation of very polar compounds, which therefore might not be detected as a feature.

different groups do not directly represent concentrations of the respective compound. Especially the compound group CHN is barley detectable in negative mode. Still, the occurrence of the compound groups can be compared relatively between the samples, indicating changes in the composition of organic aerosol. Since our method is an offline method, the detectability of compounds is dependent on the sampling time. In comparison to online techniques, longer sampling times lead to the detection of more compounds by enriching them on the filter. Additionally, the chromatographic separation through a column provides the retention time as a valuable information, which is used to identify and distinguish compounds. In comparison to AMS, HESI as a soft-ionization technique minimizes fragmentation, allowing for the assignment of molecular formulas based on the molecular ion. As a consequence of filter sampling, artifacts and losses cannot be excluded (Resch et al., 2023) and the time resolution is lower compared to online techniques. Therefore we suggest using online and offline techniques in parallel to retrieve to most information.

In general, each region and each flight shows its own unique chemical composition as it will be discussed in the following sections.

the stratosphere or mixing at the tropopause. As a limitation of our technique it has to be mentioned that uncertainties in the
370 detection of highly oxidized molecules (HOM) derive from the offline filter treatment that may lead to HOM decomposition and therefore an under representation in our dataset (Bianchi et al., 2019).

Regarding compound classes that are found on the blanks, I added a figure below as an example, plotting the compounds that are detected on the different flight filter blanks. First, I want to note that all of our samples are blank corrected by the respective flight filter blank. Of course, as you mentioned, it is important to do so since especially the CHOn and CHOa compounds are background signals, as you can see in the figure below. The compounds in the background are dependent on the environment and thus blanks are crucial for each measurement. Additionally, contamination by solvents used can be enriched during our evaporation step and thus appear in the filter blank as well. Here we want to mention **line 132** where we highlighted that only signals with a sample-to-blank ratio greater than 5 are detected as a feature.



3) Could you add a table or other information on a handful of the most abundant compounds? I agree we don't want massive lists. But for example, is there one organosulfate (glycolic acid sulfate, IEPOX sulfate, or ?) that is sometimes more abundant, and what it is its concentration? Oxalic acid has been identified as a small but significant (a few %) of aerosol organics in some remote environments. Is it measurable? Maybe a table of the two or three most abundant compounds in each major category (like CHOn, CHOS, not necessarily every category) and how they varied between samples.

We agree that this is useful information and added a table in the SI, mentioned in **line 285, Table 3**. Here, we want to mention that we have mostly no information on concentrations, since that would require calibration measurements and thus the

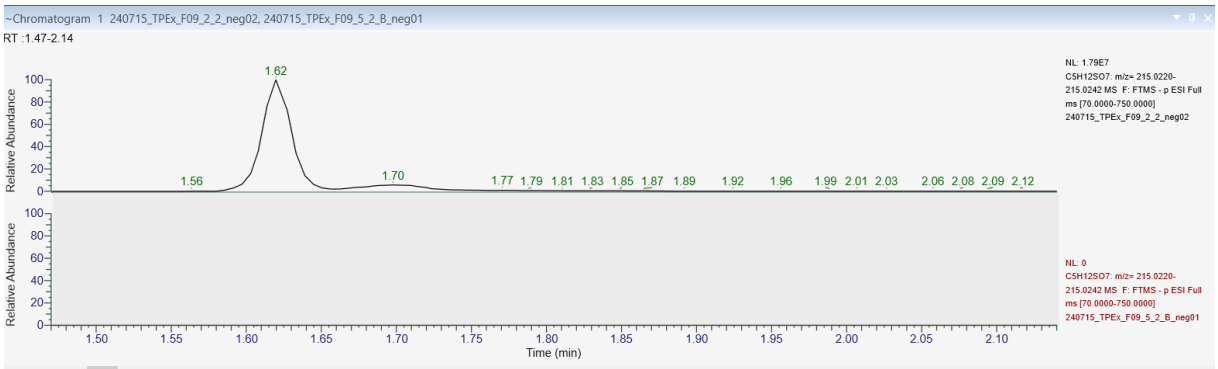
availability of authentic standards. The signal intensity therefore indicates the variability between samples but is not necessarily intercomparable between different compounds. We attached the following table to the SI:

illustrates different transport processes. Our results particularly highlight the role of altitude and geographical location in relation to the chemical composition of the organic aerosols. The most abundant compounds for each elemental composition group
285 and their variability is described in Table. S3.

Table S3. Measured intensity of the most abundant moelcule found in the respective elemental composition groups CHOn, CHNO, CHOS, CHNOS, CHOP and *other*, measured with Orbitrap mass spectrometry in negative ionization mode. The Intensity is displayed for each sample, indicated by the Flight number (F05 and F09) and the sample number (1–4). Intensity threshold (i.t.) was set to 1E+05.

Molecular Formula	F05-1	F05-2	F05-3	F05-4	F09-1	F09-2	F09-3	F094
CHOn								
C9 H16 O4	1.37E+09	3.34E+07	1.81E+07	2.14E+07	1.30E+07	7.84E+07	1.34E+10	5.43E+09
C9 H14 O4	< i. t.	3.55E+05	5.38E+06	< i. t.	2.97E+06	2.13E+09	1.49E+08	3.66E+07
CHNO								
C9 H17 N O3	2.25E+05	4.21E+07	3.28E+09	9.31E+07	< i. t.	1.91E+06	1.95E+06	9.88E+06
C17 H35 N O5	9.04E+05	< i. t.	2.91E+05	< i. t.	1.95E+06	9.06E+08	1.47E+08	< i. t.
CHOS								
C17 H28 O3 S	8.46E+08	< i. t.	< i. t.	< i. t.	1.14E+08	3.06E+07	5.08E+05	4.71E+09
C16 H34 O6 S	4.06E+05	4.27E+05	1.09E+06	1.19E+06	9.84E+08	1.59E+09	9.94E+08	1.59E+09
CHNOS								
C15 H15 N O3 S2	1.45E+05	< i. t.	< i. t.	1.11E+05	1.12E+10	2.05E+05	2.70E+05	< i. t.
C5 H11 N O4 S	1.75E+07	6.84E+08	< i. t.	3.35E+08	8.86E+07	3.00E+06	2.64E+05	1.99E+08
CHOP								
C8 H19 O4 P	7.07E+09	< i. t.	1.08E+05	< i. t.	5.80E+07	2.01E+07	4.82E+07	3.46E+09
C12 H27 O4 P	4.14E+05	< i. t.	< i. t.	< i. t.	2.24E+07	1.25E+09	4.05E+08	< i. t.
other								
C8 H19 O3 P S	2.63E+05	< i. t.	< i. t.	< i. t.	< i. t.	1.29E+05	2.01E+10	< i. t.
C12 H27 O3 P S	< i. t.	< i. t.	3.17E+05	< i. t.	1.30E+07	7.84E+07	1.34E+10	< i. t.

Regarding the specifically asked compounds, I checked our data for oxalic acid (C2H2O4) and could not find a signal attributed to this compound. I am afraid, that it is too polar to be properly separated on our C18-column. Regarding glycolic acid sulfate (C2H4O6S), we couldn't find a signal either. IEPOX sulfate on the other hand, or at least the respective mass trace (C5H12SO7) was detected (see chromatogram below, top: sample; bottom: blank), though we did not check with a reference standard and thus can only verify that this compound has the same sum formula as IEPOX-sulfate.



4) I found some of the colors on Figure 4 and subsequent figure impossible to associate with the legend. I cannot tell for sure which one is CHOa, CHN, or CHNO. CHOP and CHOS can be distinguished when the bars or slices are big but not when they are small. I'd suggest using hatching on some of the categories to help distinguish them. It would also help if the legend were in the same order as the bars: why is CHN right above CHOn in the bars but at the other side of the legend? Or am I mixing up the colors?

For better visibility, we changed the following for figures 3-6:

Figure 3 – **line 315**: We changed the color of CHN to grey. We added in the caption the order in which the groups are displayed in the bars (the legend reads in columns from left to right). Since the smaller slices are hard to see, we decided against hatching, since we wanted to keep the color coding consistent throughout all the figures and latest figure 5 would suffer from hatched bubbles. For enhanced clarity in figure 3, we added table 1 in **line 199**, where the information displayed can be retrieved as well.

Figure 3. Relative abundance in % of the elemental composition groups CHOn, CHOa, CHNO, CHOS, CHNOS, CHOP, CHN and *other* (the order mentioned applies to the stacked bars from bottom to top), measured with Orbitrap mass spectrometry in negative ionization mode (also given in Table 2). Displayed are the filters that were sampled throughout the flights F05 and F09. In panel F09, the filter sampled at the ground station Taunus Observatory is added.

Figure 4: We refined the caption to clarify the legend **(line 338)** and changed the color code for CHN to grey.

Figure 4. Five-day backward trajectories of the sampled period for F05: filter 3 (stratospheric) and 4 (tropospheric) and F09: filter 2 (tropospheric) and filter 4 (stratospheric). The altitude of the air mass is displayed in km and color coded by the color bar displayed. The origin of the respective air mass is marked by a blue dot. The pie charts show the abundance of the respective elemental composition group.

Figure 5 – **line 346**: We changed the color code for CHN to grey. We agree that this plot is hard to read and very busy, which unfortunately lies in the nature of fingerprint plots.

Figure 6 – **line 484**: We changed the whole color code and decreased the transparency, so that the coding is not mistaken with colors used before and better readable. Additionally, we changed the symbol for the stratospheric samples to a diamond to be better distinguishable.

Figure 6. The colored dots display the peak area (y-axis) of the target compounds PFOA, PFNA, PEE and TCPP for the respective filter. The boxplot next to it displays all filter blanks (n $\geq$ 11) that have been collected throughout the whole campaign. The stratospheric filters are marked with a star (*) and displayed as a diamond in the plot. Colorless circles mark ~~ouliers~~ outliers that are defined as $x < Q1 - 1.5 \cdot IQR$ or $x > Q3 + 1.5 \cdot IQR$ with $IQR = Q3 - Q1$, with IQR as the interquantile range.

*5) Very minor: **Line 349** and following "Level 2", etc. is not defined. Figure 5, please specify atomic or mass O:C. **Line 231** about sulfate versus altitude a better reference would be Wilson et al., 2008, www.atmos-chem-phys.net/8/6617/2008/. **Line 29** the 50% organic fraction only applies just above the tropopause.*

In **line 139** we mentioned the confidence levels according to Schymanski et al.. For clarification we added the following in **line 139 and following:**

for our compound identification, we used the confidence levels established by Schymanski et al. (2014). According to this
140 classification, level 1 corresponds to unambiguously identified compounds (reference standard, MS^2 spectrum), level 2 to a probable structure (library match and/ or diagnostic evidence), level 3 tentative candidate (structure, substituent , class), level 4 unequivocal molecular formula and level 5 exact mass. We used the mzCloud database (HighChem LLC, 2013-2021) for comparing MS^2 spectra of commercial chemicals. correspond to confidence level 1.

To clarify whether its atomic or mass O/C in Figure 5, we added 'atomic' in **line 350.**

sphere. All compounds are displayed in a Van Krevelen diagram, which shows the atomic O/C against H/C ratio, indicating
350 degree of saturation and oxidation state. The pie charts indicate the relative number contribution of each elemental composition

Thank you for the suggestion, we changed the reference in **line 257** to Wilson et al. 2008.

255 compared to on average 0.15 $\mu g\ m^{-3}$. An elevated sulfate abundance is generally expected according to earlier research by Crutzen (1976) and Junge et al. (1961), observing a stratospheric sulfate layer. Moreover, previous work studying the UTLS region shows high sulfate concentrations with increasing altitude ~~(Appel et al., 2022; Duncan et al., 2007; Tomsche et al., 2022)~~ (Wilson et al., 2008; Duncan et al., 2007; Tomsche et al., 2022). The same relative abundance of organosulfates is observed

We changed the statement in **line 31** about the occurrence of stratospheric aerosol in the following way:

30 in the stratosphere (Solomon et al., 2023).
Organic matter makes up to 50 % of ~~stratospheric aerosol~~ aerosol in the lower stratosphere (Murphy et al., 2013; Froyd et al., 2009) and is either being formed by gas-to-particle conversion (Hallquist et al., 2009) or transported to high altitudes from

Referee #2 – response:

This manuscript presents interesting and valuable datasets on organic aerosol composition in the UTLS, an important and understudied region. The authors describe the collection of UTLS aerosol samples from an aircraft platform and subsequent detailed offline molecular-level analysis. The study is within the scope of ACP and should be of broad interest to the atmospheric science community. However, there are areas where the discussion of chemical composition results could be improved, especially in Section 3.5 ("Non-target analysis of organic compounds within different parts of the UTLS"). More clarity in interpretation would strengthen the manuscript.

Here we added additional statements, to contextualize our findings better in **line 344 and 465**.

troposphere and stratosphere, and hence there are no characteristics of aerosols studied so far that explain the different occurrences. Here, we present the possibilities of our technique, encouraging more research in this field to gain a better understanding
345 of the findings presented below.

3.6 Identified anthropogenic influence in the UTLS

Especially the occurrence of organothiophosphates, which were found in the stratosphere, suspects anthropogenic influence that should be of further concern. To highlight the possibilities of a targeted approach and focus more on investigating the

A major scientific concern I have is the results and discussions of the oxidation state in Figure 5 and the section "Comparison of chemical characteristics." It is surprising to me that the stratospheric organic samples appear less oxidized than the tropospheric samples, given the longer aerosol residence times in the stratosphere and the generally oxidizing environment there. Actually, the reported O/C values (< 0.5 for most data) in Figure 5 for both stratospheric and tropospheric samples are also low compared with ambient aerosol summaries (e.g., Chen et al., 2015: https://agupubs.onlinelibrary.wiley.com/doi/10.1002/2015GL063693). This raises the possibility that the offline analysis might be missing more oxygenated, highly polar species.

Have the authors compared the offline O/C and H/C results with online AMS measurements for validation? This would be important, given that AMS data are available for these flights and could reveal whether highly oxidized material is underestimated by the offline approach. The authors should discuss this explicitly, as it has implications for the broader interpretation of UTLS organic aerosol properties.

Thank you for pointing out this issue. We have discussed parts of why the O/C might be relatively low already, referring to **referee #1**, but want to extend on that. First of all, the sensitivity of CARIBIC-AMS was unfortunately not sufficient, which was due to the conditions during the campaign (too short of preparation-time before the flights and short flights in general). As you can see in the plot below, the error bars are too large to confidentially interpret the oxidation state of the organic aerosols.

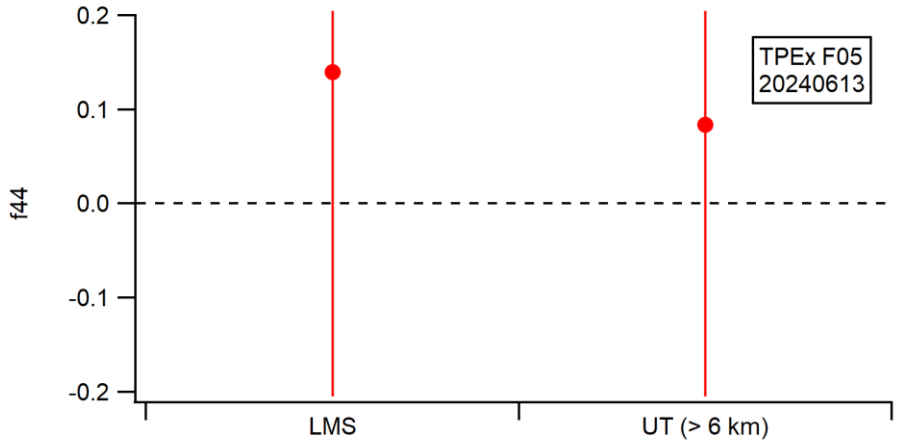
The plots you see below show the averaged f44=org44/org over the time period for the respective atmospheric layer which is defined as follows:

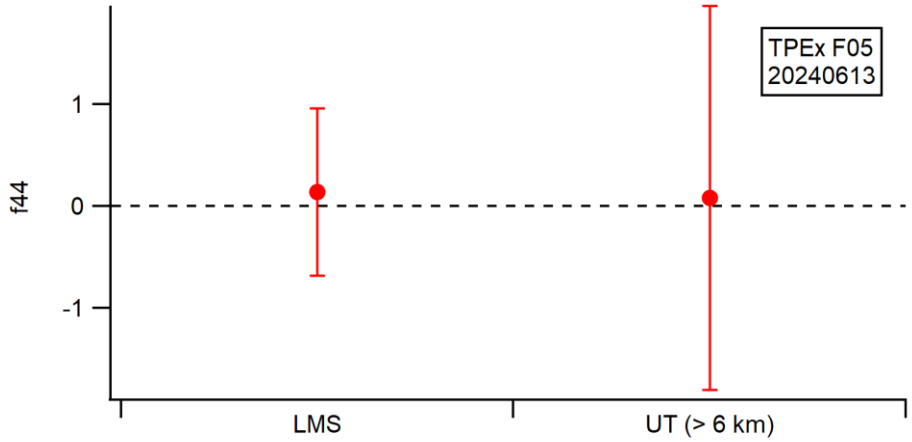
LMS: O_3>100 ppbv, N_2O<330 ppbv

UT: O_3<100 ppbv, N_2O>330 ppbv, > 6 km (F05) and > 8 km (F09).

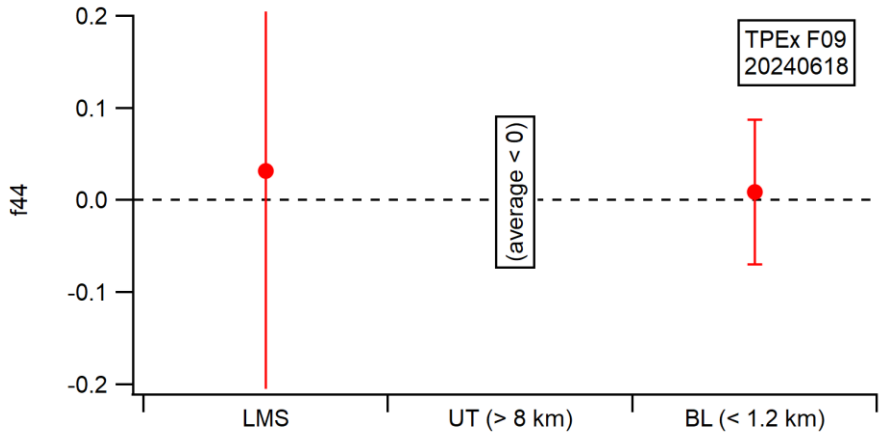
BL: < 1200 m, only F09, without time right before and after launching/ landing. The error bars are derived from the standard deviation of the mean and the gaussian deviation.

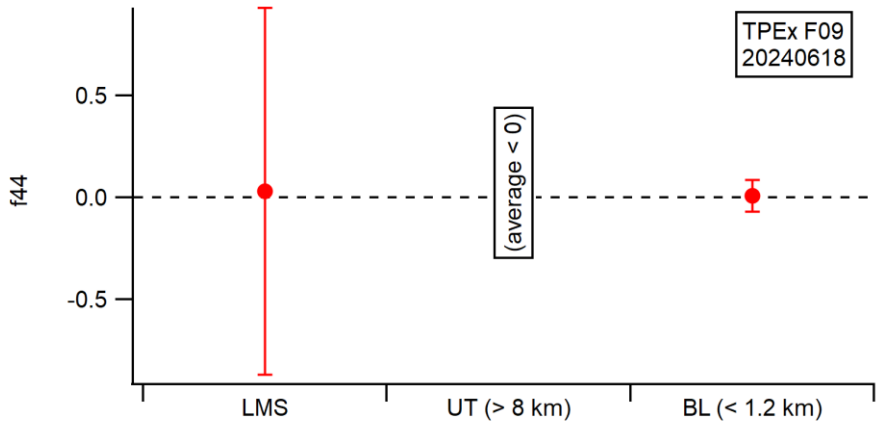
F05:


0.2
0.1
0.0
-0.1
-0.2
f44
TPEx F05
20240613
LMS
UT (> 6 km)


1
0
-1
f44
TPEx F05
20240613
LMS
UT (> 6 km)

F09:


0.2
0.1
0.0
-0.1
-0.2
f44
TPEx F09
20240618
(average < 0)
LMS
UT (> 8 km)
BL (< 1.2 km)



As already discussed in a comment to **referee #1**, there are some limitations when it comes to measuring HOM with an offline approach (Bianchi et al. 2019), and highly polar compounds are difficult to separate chromatographically with our C_{18}-column as well. We incorporated these limitations in **line 204 and 215**. Moreover, I would emphasize that online techniques such as HR-AMS result in different O/C than offline techniques as mentioned above, due to losses and decomposition that occur during the sample handling and the filter sampling itself. We added on that in **line 367**. When comparing O/C to other measurements done by UHPLC-HRMS, e.g. Pereira et al. 2025 (https://acp.copernicus.org/articles/25/4885/2025/) or Thoma et al. 2025 (https://pubs.rsc.org/en/content/articlehtml/2025/ea/d4ea00163j), we see the same range for O/C.

I also have a few specific questions and comments.

Specific comments:

1. *It would be helpful to include additional references on the radiative and chemical importance of UTLS organic aerosols, such as:*

- *Murphy et al., 2021: https://acp.copernicus.org/articles/21/8915/2021/*
- *Li et al., 2021: https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2021GL094427*

These citations were included in **line 38** along with the following text:

processing and deposition, although detailed processes are still unknown (Jimenez et al., 2009; Pöschl, 2005). Especially in the context of radiative effects on climate, the difference between aerosol produced in the stratosphere or transported into the
40 stratosphere is important to understand to assess the radiative effect (Murphy et al., 2021). The significant organic content in stratospheric aerosol hereby contributes to a substantial impact on the aerosol optical depth, which broadly ranges for different complexes, highlighting the need to understand the composition of stratospheric aerosol (Li et al., 2021).

- *Solomon et al., 2023: https://www.nature.com/articles/s41586-022-05683-0*

We added this reference in **line 28**:

son et al., 2019; Andreae et al., 2018; Benoit et al., 2023; Dumelié et al., 2024). Wildfires and the related aerosols are moreover suspected to contribute to enhancing ozone loss rates and show the importance of understanding aerosols and their composition
30 in the stratosphere (Solomon et al., 2023).

*1. **Line 79-80**: Please clarify which aerosol loss processes are included in the sampling efficiency calculation. Does this account for non-isokinetic sampling bias at the inlet tip?*

The losses are calculated according to Weiden et al. 2009 (https://amt.copernicus.org/articles/2/479/2009/) with a program called 'Particle Loss Calculator', which is described in the paper mentioned. To clarify the loss-mechanisms included we added the following in **line 86**:

85 Based on calculations according to von der Weiden et al. (2009), we estimated the particle losses, caused by the design of the inlet system, to be less than 10 % in the size ranges from 10 nm to 2000 nm (Fig. S2 and S3). The loss-mechanisms included in the loss-calculations are non-isoaxial sampling, non-isokinetic sampling, diffusion, sedimentation, turbulent inertial deposition, inertial deposition in bend and inertial deposition in contraction. SOAP therefore captures particles in the Aitken

2. For all reported concentrations in µg m $^{-3}$, please state whether they are given at STP or ambient conditions. This is important for comparison with other studies.

The concentrations are given at STP. To clarify we added the following in **line 210**:

Figure 3 shows the relative abundance of each compound group for the respective region (exact numbers can be found
210 in Table 2). Additionally, AMS measurements of submicron sulfate (SO_4^{2-}, concentration given at standard pressure and temperature) as well as the size distribution measured by UHSAS are compared to our observations (Fig. S4–S6).

3. In the section "CHOS, troposphere vs. stratosphere," you describe a correlation between organosulfates and sulfate concentrations measured by the online AMS. Could you clarify how the offline method distinguishes organosulfates from inorganic sulfate? In addition, can the online AMS measurements provide any direct information on organosulfates beyond the bulk sulfate concentration?

Sulfate and organosulfates are separated chromatographically, whereas the sulfate elutes very early, and has a *m/z* of 97. The CHOS compounds on the other hand elute later and have a higher *m/z*. Here, the soft ionization retains the molecular ion and therefore we can assign organic compounds that contain a sulfate group.

In theory yes, unfortunately the performance of the AMS was not as we hoped, therefore the noise of our data is too high to say anything about that.

4. Table 2 and Figure 3: The fraction/trend of CHOS (organosulfates) in the upper troposphere and lower stratosphere appear to be opposite for F05 and F09. Could this be

related to differences in tropopause fold influence versus convective injection between the two flights, or are there other possible explanations?

That is a very good observation you made. We would suspect as well that the reason is the difference between tropopause fold and convective phenomena, but we have no clear interpretation for that. Here we would emphasize again that we would need more flights and more data to have solid interpretation. As an outlook one could think about assessing whether its biogenic or anthropogenic CHOS, since F09 is a flight over Germany and F05 on the other hand over sea. We added this in **line 261**.

both observations that we made with our filter samples. The opposite trend of CHOS in the stratosphere for F05 and F09 could be due to the difference in a flight over land and a flight over sea, which needs to be investigated in further studies.

5. Figure 6: The colors for different samples within a single flight are difficult to distinguish, and the stratospheric samples are not easily identifiable. Consider using a more distinct color palette and clearer legends.

We changed the colors as well as the markers for the stratospheric samples. The stratospheric samples are now marked in a diamond shape for enhanced clarity.