

**Responses to the reviewer's comments on
"A one-year ACSM source analysis of organic aerosol particle contributions from anthropogenic sources after long-range transport at the TROPOS research station Melpitz" by Atabakhsh et al.**

Please find below the response to reviewer #1:

Reviewer #1 (R1):

We thank the reviewer for his insightful review and constructive comments. We highly appreciate your time in reviewing the manuscript. All the comments/suggestions were taken into consideration and incorporated in the revised manuscript, which has improved the quality of the revised manuscript. The point by point response to all the comments and suggestions of reviewer #1 (R1) is provided in the following sections. For clarity, the reviewer's comments are provided in blue (RC), the author's comment (AC) is in black, and the revised parts of the manuscript are shown in red.

Atabakhsh et al. have reported a nice study of one-year ACSM/MAAP measurement. Using rolling PMF and a multilinear regression model, they have conducted detailed source appointments for non-refractory aerosol components and eBC. In addition, to better identify the origin of aerosol sources, clustering analysis was also applied using the back-trajectory cluster method. Based on these approaches, the variations of chemical composition, aerosol mass concentrations, and diurnal cycles among the meteorological seasons were carefully examined and discussed, especially the comparison was performed between the cold and warm seasons. I have a couple of comments on the determination of the emission sources of sulfate, nitrate and org. The comments are not serious but need to be clarified and justified. Overall, the manuscript is well written and easy to read. I would recommend its publication after my comments are addressed.

AR: Thank you for your generally positive view on our submission. We have revised in our manuscript, see below at the individual points. We think this way the discussion of the results as well as their interpretation was much improved.

General comments:

R1C1: The interpretation of sources and diurnal cycles of aerosol components relies heavily on the wind rose patterns and NWR analysis in Sections 3.1-3.3. If I understand correctly, both techniques can only determine local emission sources. However, you have claimed the significance of long-range transported aerosol sources for different measured aerosol species in this study. The statement appears bit abrupt and needs more clarification. Since you have conducted nice back-trajectory and cluster analysis later, I would suggest you already cite those results to support your conclusions in Sections 3.1-3.3.

AC1: We thank the reviewer for the comment. The non-parametric wind regressions model (NWR) can be used to investigate not only the local but also the transported emission sources as it was already done in for example (Marin, et al., 2019). With this regard, high chemical species mass concentrations observed at low wind speeds, can be recognized as local sources, whereas high mass concentrations associated to higher wind speeds correspond to more transported ones coming from this specific wind sectors. Consequently, the NWR analysis not only provides information on local emissions but also gives some details on the transported sources associated with a main prevailing wind at high wind speed. Nevertheless, we have to recognize that this approach did not provide any information on the distance as well as the location of the emissions' areas. In order to clarify the statement, the following lines have been added to the NWR definition:

- **Line 214-216 (revised version):** Non-parametric wind regressions (NWR) were used to approximate the OA source concentrations at a given wind direction and speed (Henry et al., 2009) in order to investigate not only the local but also the prevalent wind sector associated with transported emission sources (Marin, et al., 2019).

Since the wind direction did not provide any information on the air mass origin, cluster analysis on air mass back-trajectories was used to identify the long-range transported emissions and it was discussed in Sect. 3.4. However, we briefly included them in Sect. 3.1-3.3, we decided to keep Sect. 3.4 (Seasonal air mass clustering) as an overview of this study which comprehensively discussed the origins of chemical species and PMF sources. Therefore, some parts of Sect. 3.1-3.3 have been revised as follows:

- **Line 286-291 (revised version):** Furthermore, the NWR plots (Fig. S3) show that during the winter time, sulphate mostly comes from the north and east sectors with wind speeds above 5 m/s which can be associated with dominant transported sulphate sources. Although the eastern wind sector remains visible for the sulphate in the summer time, the high concentrations of sulphate can be observed during periods with low wind speed and without a specific wind sector; which corresponds to local sulphate formation. Sect. 3.4 will go into detail about the long-range transported emissions later on.
- **Line 307-311 (revised version):** Nitrate profiles from NWR plots (Fig. S3) present two different wind directions for the whole period which might be associated with transported nitrate from Leipzig and Torgau (50 km in the south-west and 7 km in the north-east of Melpitz, respectively) with higher wind speed. Since the reaction pathway of OH and NO₂ can result in nitrate formation (Yang et al., 2022), this mechanism can be linked to traffic emissions in residential areas. These long-range transported sources together with locally formed emissions could describe higher mass concentrations for nitrate and ammonium due to e.g., meteorological conditions and abundant precursors in winter time.
- **Line 339-343 (revised version):** Furthermore, considering measured eBC-PM₁ in regard to wind speed and wind direction from NWR plots (Fig. S3), eBC-PM₁ presented transported and local emissions. The highest mass concentrations for fall, winter, and spring seasons could be linked to north-easterly and south-westerly winds with higher wind speed (above 10 m/s). While in summer time it is mostly linked to the surrounding emissions regardless of wind direction with lower wind speed (Fig. S3).
- **Line 403-412 (revised version):** Analyzing the NWR plots demonstrates the highest HOA mass concentration was observed at low wind speed during the warm period (Fig. 7) indicating, rather local emission sources. While during the cold period a clear increase of the mass concentration can be associated with the highest wind speed (> 10 m/s) mostly coming from the North to East sector. During periods with wind speeds below 10 m/s, the two dominant wind sectors (NE and SW) can be observed. The first one might be associated with emission plumes coming from either the surrounding traffic emissions (the federal street B 87), as well as the domestic emissions are associated not with house heating in summer but with hot water production (van Pinxteren et al., 2020), as well as the city of Torgau (with approx. 20,000 inhabitants, distance from 7 km). Although the SW sector shows a lower HOA mass concentration in comparison to the NE one, it corresponds to the direction of the city of Leipzig (above 600 000 inhabitants, approx. 50 km). Therefore, it might be associated with the influence of the pollution plume of the city of Leipzig.
- **Line 436-439 (revised version):** By analyzing the NWR model, the high mass concentration of BBOA in cold months, regardless of wind speed can be observed with two wind sectors coming from north-east and south-west directions. These BBOA emissions are mainly attributed to residential heating in Melpitz village and also indicate the effect of transported biomass burning emissions to the sampling site with higher wind speed (> 10 m/s, Fig. 7).
- **Line 467-470 (revised version):** CCOA showed the highest mass concentration and mass fraction during the winter (1.58 µg/m³, 23 %, respectively; Fig. 5a; Table. 1). By analyzing the NWR plots, this high mass concentration during winter time can be related to the surrounding emissions and long-range transported air masses coming from two different directions, north-easterly and south-westerly (Fig. 7).
- **Line 514-516 (revised version):** This high mass concentration in cold seasons can be seen from the NWR plot (Fig. 7) presenting local emissions with low wind speed (> 5 m/s) and transported emissions from east, north-east, and south-west directions with high wind speed (< 5 m/s).
- **Line 540-542 (revised version):** Furthermore, since nitrate could be originated locally or arrived from a long distance to Melpitz (Sect. 3.1.1), with a good correlation between LO-OOA and nitrate (R²= 0.59) during winter, the long-range transported LO-OOA from different directions reaching to measuring site could be explained (Fig. 7).

Specific comments:

RIC2: L16, How about nucleation? Should it be a source of secondary aerosol?

AC2: We thank the reviewer for the comment. This is true that one of the mechanisms for the formation of SOA could be organic nucleation (Zhu and Penner, 2019), since the condensation of organic species, plays an important role in the growth of newly formed particles into larger ones. Moreover, small particles do not play a significant role in the total particle mass. Only when the newly formed particles reach a significant diameter (approx. 100 nm), they can significantly influence the overall mass. At this point, the competition between the contribution of condensing organic species to the newly formed particle growth and the pre-existing particles is hard to distinguish. Therefore, we preferred to use the SOA, which indirectly includes all the different processes of SOA formation.

RIC3: L48-49, PM₁ also has an effect on air quality, right?

AC3: PM₁, as one of the air pollutants itself, it is not necessary to address it actually affects the air quality. To clarify the reason why PM₁ is important, the text has been modified as follows:

Line 45-49 (revised version): The submicronic particles known as PM₁ (particles with an aerodynamic diameter less than 1 μm), not only have a negative impact on human health (Pop and Dockery, 2016; Daellenbach et al., 2020) but also have a significant effect on visibility (Shi et al., 2014) and climate (Shrivastava et al., 2017). **It is ability to penetrate to respiratory system make it more dangerous, therefore more relevant to mitigate adverse health impact.**

RIC4: Sec. 2.2 How did the temperature vary at the measurement station where ACSM was located, since the ambient temperature may affect the instrument sensitivity from season to season?

AC4: Since all instruments were situated in a container with air conditioning, the temperature of the instruments have been exposed to fairly constant over seasons. Therefore, we consider the influences by ambient temperature to the instrument sensitivities are negligible in our case.

RIC5: Sec.3.1 Since ACSM is associated with a couple of measurement uncertainties, such as the determinations of collection efficiency and response factor, it would make a lot of sense to compare ACSM derived concentration to other instruments in the parallel measurements.

AC5: The intercomparing of the ACSM results with collocated instruments including SMPS, off-line PM₁, and PM_{2.5} for the total particulate mass, water-soluble ions and OC/EC was already discussed in deep detail in our previous work (Poulain et al., 2020), it appears to be out of the scope of this paper to repeat this work and it was only referred to it on the preprint manuscript in line 117-118 preprint version (“Details on the QA/QC for this dataset can be found in Poulain, et al., (2020)”). In order to better refer to our previous work, the sentence line was changed as follows:

Line 122-125 (revised version): **The quality assurance of the ACSM measurements was performed by comparing them with collocated measurements including MPSS, and high-volume filter samples (PM₁ and PM_{2.5}) for the total particle mass concentration, water-soluble ions (nitrate, sulphate, and ammonium), as well as OC/EC. Details on the QA/QC and instrumental uncertainties can be found in Poulain et al., (2020).**

RIC6: L228-231, As you have mentioned in the nitrate results in the next paragraph, nitrate concentration dropped in the afternoon due to the vertical mixing. Similar phenomenon is expected for the sulfate species too. However, a flat diurnal cycle was observed in winter when photochemistry was weak. Did it indicate that, except for long-range transport, there were additional sources for sulfate around noon in wintertime? Can you comment on it? In addition, you observed a high peak at noon in other three seasons, and you interpreted it to be the photochemistry of SO₂. I just wonder if enough OH are available for SO₂ photochemistry to explain such a diurnal peak given the effect of boundary layer mixing? A rough estimation on this point would not go amiss.

AC6: It is true that vertical mixing can influence the ambient mass concentration, leading to a decrease of the mass concentration by a “dilution effect”. However, it is not the only process that can influence the ambient concentration of chemical species. For example, it is well known that ammonium nitrate is a semi-volatile compound in equilibrium between the particle phase (NH_4NO_3) and the gas phase (NH_3 and HNO_3), which is affected by the gas phase concentration of NH_3 and HNO_3 , temperature, and hygroscopicity. Therefore, the decrease of the nitrate mass concentration during day time is not only influenced by the vertical mixing but can also be affected by its gas-to-particle equilibrium. Thermodynamics of the ammonium nitrate at Melpitz was already discussed in Poulain et al., (2011).

On the opposite, sulfuric acid is a less volatile species than HNO_3 and will condense on the particle phase, leading to ammonium sulphate, which will be mostly affected by the dilution effect due to the change of the mixing layer height. Consequently, the mentioned drop of the ammonium nitrate in the afternoon can be associated with a change in the ammonium nitrate partitioning, while ammonium sulphate stays in the particle phase and is constant during the winter. Moreover, the higher sulphur dioxide (SO_2) concentration in winter, can also lead to photochemically formed H_2SO_4 and finally in particulate sulphate.

Furthermore, Größ et al., (2018) presented the relationship between SO_2 , the hydroxyl radical, OH, and sulfuric acid (H_2SO_4) in the boundary layer for different cases at the Melpitz station (June 2010, May 2008, June 2010, and August 2008). They highlighted that high sulfuric acid concentrations were caused primarily by the high level of SO_2 and hydroxyl radical $\bullet\text{OH}$ with significant solar radiation around noon. This could simply explain the high peak at noon not only in the winter time but also the in the other season as well.

RIC7: L231-232, You should make it clear that it is valid only for the same season that a high sulfate concentration was shown at low wind speed, but does not hold when you compare it among different seasons.

AC7: This comment was already answered in our answer to the general comment of the reviewer. The mentioned text has been revised as follows:

Line 286-291 (revised version): Furthermore, the NWR plots (Fig. S3) show that during the winter time, sulphate mostly comes from the north and east sectors with wind speeds above 5 m/s which can be associated with dominant transported sulphate sources. Although the eastern wind sector remains visible for the sulphate in the summer time, the high concentrations of sulphate can be observed during periods with low wind speed and without a specific wind sector; which corresponds to local sulphate formation. Sect. 3.4 will go into detail about the long-range transported emissions later on.

RIC8: L248-250, You had very good discussion on the local contributions to eBC and org in section 3.1.2, did the local source contribute to nitrate during winter season? As far as I understand, NWR only determines local sources, nor the long-range transported ones (Fig. S2). If you argue the dominant sources of nitrate from long-range transport, you might need to justify it. Furthermore, your cluster analysis shows a high local nitrate mass concentration (CS-ST), should this suggest a local source of nitrate?

AC8: We thank the reviewer for the comment. The discussed lines are: “In winter, ammonium-nitrate remains mainly in the particle phase (Seinfeld and Pandis, 2006) and, like sulphate, arrived at the measurement site due to the long-range transported emissions which not only came from the north-eastern but also south-western flow, describing higher mass concentrations for nitrate and ammonium (Fig. S2).”, pre-print version, which we are going to answer to the comment in two parts:

- Since the statement is talking about ammonium nitrate during winter time, the word ‘mainly’ just emphasizes that ammonium nitrate mostly exists in particle phase at low temperature (Spindler et al., 2010) due to its volatility, and we did not claim about the major source of nitrate here. However, in the lines 307-312 (revised version), the long-range transported emissions of nitrate have been mentioned, since the study of Spindler et al. (2010) confirms the arriving long-range transported particles (including sulphate, nitrate, ammonium, and carbonaceous materials) from the other side toward Melpitz site.

- As we already discussed in response to ‘General comments’, the NWR model works with local wind measurements but can determine the local and transported emissions. However, besides the local source for nitrate, it shows two different directions with a higher wind speed during winter time rather than other seasons (Fig. S8). This high wind speed can bring the particles from pollution sources toward the measuring site. Furthermore, to confirm the long-range transported emissions, cluster and back-trajectory analysis are used which is widely explained in Sect. 3.4. Nevertheless, in the winter time, all results from NWR, cluster, and back-trajectory analysis present two sources:

- local source as a result of both the NWR plot (Fig. S3) and cluster analysis with CS-ST air mass (Fig. 9).

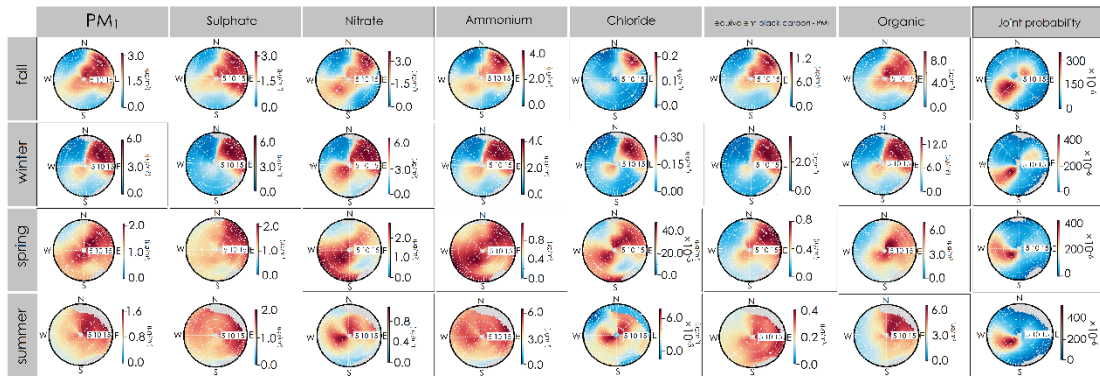


Fig. S3: Seasonal NWR plots for the different chemical compositions (in $\mu\text{g}/\text{m}^3$). PM₁ is the average of all the compositions.

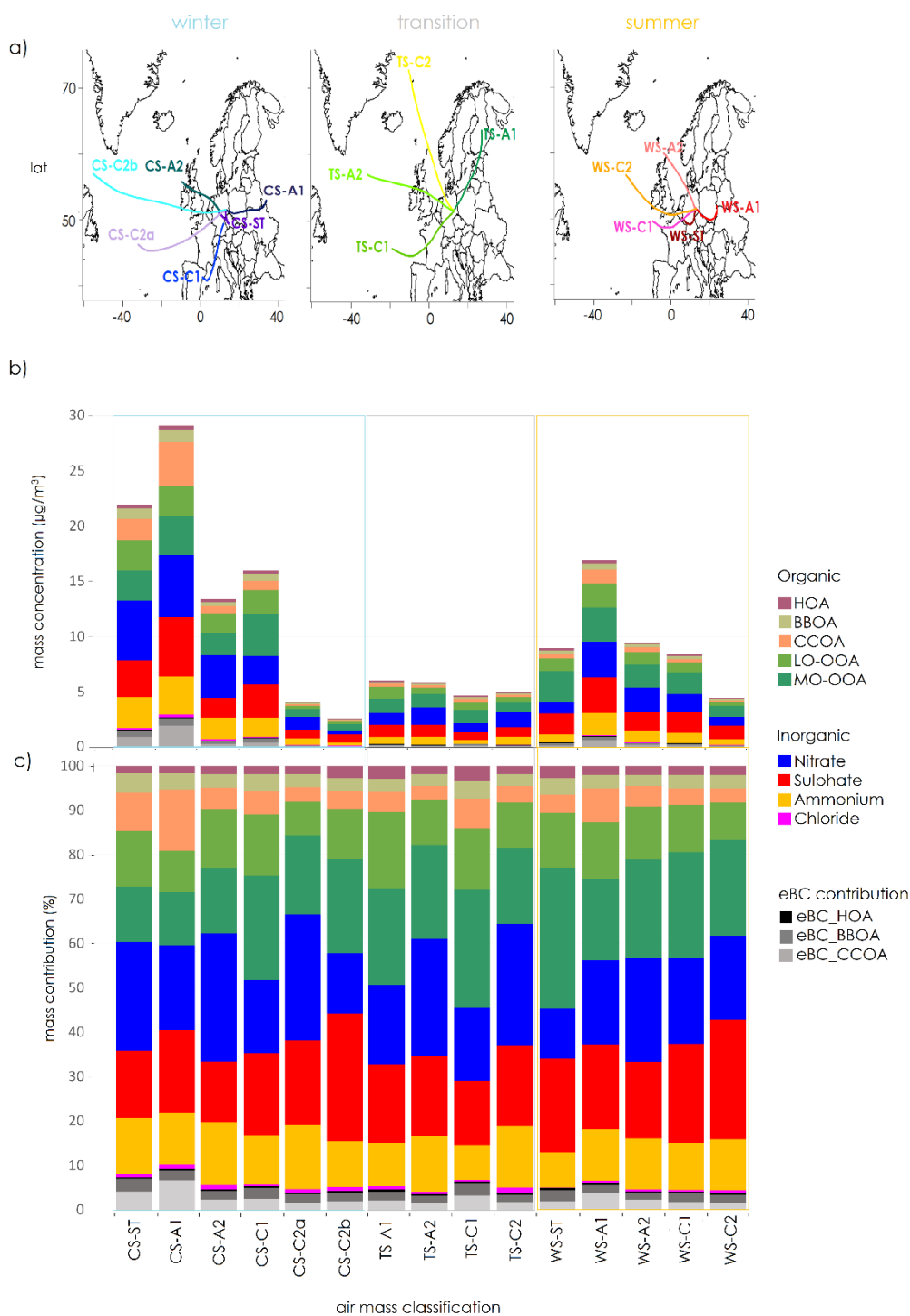


Fig. 9: a) air mass classification based on one-year backward trajectories cluster analysis at 12:00 UTC, b) influence of air mass to the PM₁ data and PMF factors, and c) contribution of them which averaged from 10:00 to 14:00 UTC.

b) long-range transported emission which not only can be seen from the NWR plot (Fig. S3) with higher wind speed in two different directions but also cluster CS-A1 as eastern air mass and CS-A2 north-western air mass (Fig. 9) confirm a high mass concentration during winter time, representing the aging effect due to the long-time transfer over the continents.

To avoid misunderstanding, the statement has been revised as following:

Line 307-315 (revised version): Nitrate profiles from NWR plots (Fig. S3) present two different wind directions for the whole period which might be associated with transported nitrate from Leipzig and Torgau (50 km in the south-west and 7 km in the north-east of Melpitz, respectively) with higher wind speed. Since the reaction pathway of OH and NO₂ can result in nitrate formation (Yang et al., 2022), this mechanism can be linked to traffic emissions in residential areas. These long-range transported sources together with locally formed emissions could describe higher mass concentrations for nitrate and ammonium due to e.g., meteorological conditions and abundant precursors in winter time. However, in winter, ammonium-nitrate remains mainly in the particle phase (Seinfeld and Pandis, 2006) since it can totally be changed from gas to particle phase at lower temperature (Spindler et al., 2010). High values of nitrate and ammonium in spring time are linked to agricultural fertilization (Stieger et al., 2018).

RIC9: L254-256, Based on my rough estimation of chemical composition during winter (Fig. 2), nitrate and sulfate were fully neutralized by ammonium as ammonium nitrate and sulfate. It sounds like the contribution of organic nitrate was negligible in this study.

AC9: Thank you for the comment. We agree with the reviewer about the neutralization process, since the previous work (Poulain et al., 2020) estimated the neutralization state of the particles assuming a full neutralization by nitrate and sulphate, and confirmed that particles can be considered as fully neutralized during the measurement for Melpitz site, which also agrees with previous AMS measurements made at Melpitz (Poulain et al., 2011). Furthermore, Kiendler-Scharr et al. (2016) have already shown that organo-nitrate can play a significant role in the nitrate signal. However, it was possible to quantify them only within high resolution AMS. Since Q-ACSM is working at a unit mass resolution (UMR), it is not possible to distinguish nitrate from organic signals at m/z 30 (CH₂O⁺ and/or C₂H₆⁺) and m/z 46 (CH₂O₂⁺, C₂H₆O⁺) ratios. Therefore, estimating the organo-nitrate would only introduce uncertainties to measurements, therefore, we did not consider to conduct this analysis in this study.

To make the statement clearer, we added the following lines to the ACSM section of manuscripts:

Line 126-132 (revised version): The ACSM ammonium mass concentration mainly corresponds to ammonium nitrate and ammonium sulphate salts. Previously by Poulain et al. (2020), the neutralization state of the particles was estimated for datasets assuming complete neutralization by nitrate, sulphate, and chloride. Therefore, the particles are neutralized when considering nitrate, ammonium, and sulphate in this study. Furthermore, the significant role of organo-nitrate and organo-sulphate on signals of nitrate and sulphate is not negligible (Kiendler-Scharr et al. (2016)). Since the Q-ACSM is working at a unit mass resolution (UMR), it is not possible to distinguish nitrate and sulphate from organic. Therefore, estimating the organo-nitrate would only introduce uncertainties to measurements, therefore, we did not consider to conduct this analysis in this study.

RIC10: L281-283, Traffic is a good source of nitrate, how does it affect your observed nitrate in this study?

AC10: Thank you for the comment. The statement in these lines (317-320 preprint version), mainly explains the two peaks of eBC-PM₁ in the morning and evening based on possible traffic emissions from federal street (B 87) which is located approximately 1.5 km away from the station in the north direction. This statement concluded mostly based on the similarity of diurnal profiles of eBC-PM₁ and nitrogen oxides which is known as a tracer for traffic emissions (Fig. S4) and not referred to as particulate nitrate.

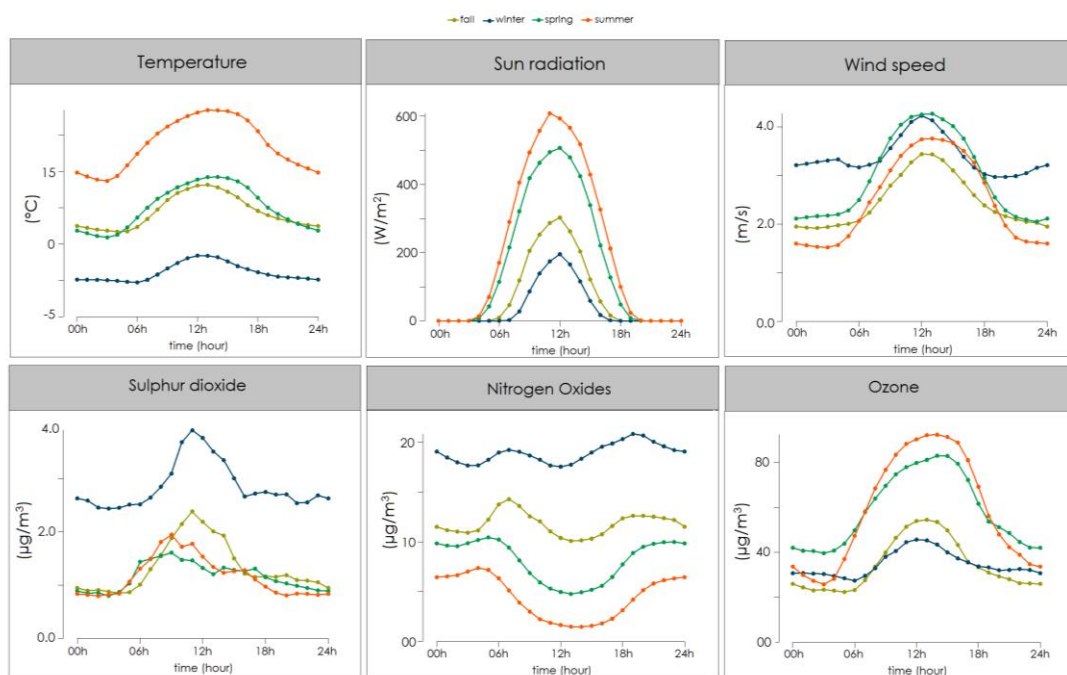


Fig. S4: Seasonal diurnal cycle of Temperature, sun radiation, Sulphur dioxide, Nitrogen Oxides, and Ozone.

Regarding nitrate source, nitrate formation from the reaction of OH and NO₂ pathway is well-known (e.g. Yang et al., 2022). The observed nitrate in our study (Fig. S3) is related to two main wind directions for all the seasons, which can be associated to transport from Leipzig and Torgau (50 km and 7 km distance from Melpitz, respectively). Therefore, we added the following statement to the nitrate discussion in Sect. 3.1:

Line 307-312 (revised version): Nitrate profiles from NWR plots (Fig. S3) present two different wind directions for the whole period which might be associated with transported nitrate from Leipzig and Torgau (50 km in the south-west and 7 km in the north-east of Melpitz, respectively) with higher wind speed. Since the reaction pathway of OH and NO₂ can result in nitrate formation (Yang et al., 2022), this mechanism can be linked to traffic emissions in residential areas. These long-range transported sources together with locally formed emissions could describe higher mass concentrations for nitrate and ammonium due to e.g., meteorological conditions and abundant precursors in winter time.

RIC11: L304-306, I agree with you that boundary layer mixing played a significant role to determine the measured org diurnal, but I don't think you can neglect the fact of evaporation of semi-volatile organic compounds in the middle day, especially in spring and summer seasons.

AC11: Thank you for the comment. We agree with the reviewer that the evaporation of semi-volatile particles is one of the reasons for the reduction of total OA during the day time (Schaap et al., 2004; Keck and Wittmaack, 2005). Therefore, we have added the following line to this section:

Line 370-371 (revised version): During warm days, evaporation of semi-volatile organics from the particle phase cannot be completely excluded (Schaap et al., 2004; Keck and Wittmaack, 2005).

RIC12: L331-332, Since you have observed very similar nighttime HOA concentrations in all seasons (Fig. 6), could the slightly different HOA concentration between summer and winter be explained by evaporation of HOA or photochemical conversion to LO/MO-OOA in summer?

AC12: Thank you for the comment. In general, low temperature in winter results in condensation of POA, which has been observed in all 22 sites in Chen et al. (2022). However, the temperature might play a role in the slightly lower HOA concentration during the night in summer time by emphasizing the

evaporation and oxidation processes of the emitted particles (Saha, et al., 2018). The difference between the HOA concentration during the night is very small, and most certainly can be covered by the uncertainties of the PMF results ($\pm 32.5\%$, Fig. S2).

Moreover, we will not expect any photochemical aging during the night but the highest nighttime ozone concentration might also be considered as a potential aging mechanism (Kodros, et al., 2020).

The following sentences have been added to the HOA paragraph:

Line 422-425 (revised version): Nevertheless, the differences between HOA mass concentration during the night time from summer to winter season (Fig. 6) are small and can be covered by the uncertainties of PMF result ($\pm 32.5\%$, Fig. S2), however, it can be explained by different emission sources, condensation of POA (Chen et al., 2022), evaporation, oxidation processes (Saha, et al., 2018), and potential night time aging process by high ozone concentration (Kodros, et al., 2020).

RIC13: L335-337, HOA concentrations observed in summer and winter were very similar, (Fig. 6), sounding more like the characteristics of local emission sources. This is also in line with the usual view of HOA as a local emission factor. The city of Leipzig is located in the sector of HOA wind rose (Fig. 7). Considering that Leipzig is 50km away from to the measurement station, we assume that the average wind speed is 4m/s, and it took only 3 hours to transport Leipzig city aerosol particles to the measurement station. As you have already stated that HOA is mainly emitted by household heating and a minor source of traffic, is it possible that the city of Leipzig, as a local/or regional source, is a HOA contributor? Moreover, here you have attributed HOA to be a long-range transport one in winter. You have shown two peaks in the morning and evening in the diurnal cycle, which sounds they were more likely related to traffic rush hours, do you think if the long-range transported HOA can explain this typical diurnal cycle?

AC13: Thank you for highlighting these points. The NWR plot of HOA (Fig. 7) presents the surrounding/local and transported emissions and also two directions which can be associated with Leipzig (with approx. 600,000 inhabitants, in 50 km south-westerly sector of the measuring site) and Torgau (with approx. 20,000 inhabitants, in 7 km north-easterly sector) cities. Therefore:

- Surrounding/locally form HOA, is mostly related to household heating (van Pinxteren, et al., 2020) for all year, and partly related to traffic emission coming from Melpitz village itself and the main street (B 87, approx. 1.5 km north of the station), which could explain the two peaks linked to the traffic rush hours.
- However, these two sectors together with cluster and back-trajectory analysis (Fig. 9), can explain the long-range transported emissions arriving at Melpitz. Clusters CS-A1, CS-A2, and CS-C1 in the winter time, and cluster WS-A1 in summer (Fig. 9), present three different air masses coming to Melpitz which include part of the HOA.

Regarding the points mentioned above, the following sentences have been modified:

Line 403-415 (revised version): Analyzing the NWR plots demonstrates the highest HOA mass concentration was observed at low wind speed during the warm period (Fig. 7) indicating, rather local emission sources. While during the cold period a clear increase of the mass concentration can be associated with the highest wind speed (> 10 m/s) mostly coming from the North to East sector. During periods with wind speeds below 10 m/s, the two dominant wind sectors (NE and SW) can be observed. The first one might be associated with emission plumes coming from either the surrounding traffic emissions (the federal street B 87), as well as the domestic emissions are associated not with house heating in summer but with hot water production (van Pinxteren et al., 2020), as well as the city of Torgau (with approx. 20,000 inhabitants, distance from 7 km). Although the SW sector shows a lower HOA mass concentration in comparison to the NE one, it corresponds to the direction of the city of Leipzig (above 600 000 inhabitants, approx. 50 km). Therefore, it might be associated with the influence of the pollution plume of the city of Leipzig.

The diurnal patterns of HOA reproduced two peaks in the morning and evening for all seasons (Fig. 6), which is related to traffic rush hours and linked to surrounding emissions from the main street (B 87,

approx. 1.5 km north of the station), Melpitz village itself, and emissions coming from Leipzig and Torgau residential areas.

R1C14: L441-443, Though I agree to that night chemistry may play a role, how about the effect of boundary layer mixing on the diurnal cycle during the daytime?

AC14: Thank you for the comment. We agree with the reviewer that the boundary layer has an effect on the particles such as MO-OOA and the following sentence has been added to this part in the revised manuscript:

Line 523-526 (revised version): Meanwhile, MO-OOA diurnal cycles presented a seasonal variation as well, with a remarkable enhancement in the evening and night time during winter (Fig. 6), indicating a potential regional formation mechanism containing night time chemistry (Tiitta et al., 2016), and descending pattern from night time to day time due to planetary boundary layer effect.

R1C15: L445-447, Sulfate is also considered as a regional or long-range transported chemical species (e.g. Zhang et al., 2005). Does a better correlation between MO-OOA and sulfate indicate the nature of long-range transported MO-OOA in this study? (Zhang, Q., et al., Hydrocarbon-like and oxygenated organic aerosols in Pittsburgh: insights into sources and processes of organic aerosols, Atmos. Chem. Phys., 5, 3289-3311, 2005.)

AC15: We thank the reviewer for the comment. We agree with the reviewer that sulphate could both originate locally and be transferred from other sites to the measuring site, which we previously discussed sulphate in Sect. 3.1. Moreover, we also mentioned a good correlation between MO-OOA with sulphate has been found (Table. 1). Nevertheless, it is pretty unspecific to state the nature of MO-OOA, since MO-OOA (same as LO-OOA) could hardly be associated with a single chemical process nor aging of a specific source (like transported or locally formed SOA) because the exact chemical composition of OOA are unknown and the aging of the primary sources could also lead to OOA profiles (Jimenez, et al., 2009). Consequently, the correlation of MO-OOA and sulphate only indicates that both are secondary compounds without a clear indication of their primary sources (anthropogenic or biogenic).

R1C16: L454-456, are you saying nitrate was formed locally or via long-range transport?

AC16: Thank you for the comment. We are going to reply to this comment based on the discussion we have had on R1C1 and R1C8. This is true that nitrate has regional background sources in Melpitz (such as night time production, agricultural fertilization especially in spring, and traffic emission), but from the NWR plot together with cluster and back-trajectory analysis (Fig. 9) the long-range transported nitrate can be seen as well which are coming from different directions to measuring station.

R1C17: L458-459, As you have stated in lines 248-251 that nitrate is a long-range transported species, does a good correlation between LO-OOA and nitrate indicate that LO-OOA is also mainly from long-range transport?

AC17: We thank the reviewer for the comment. Regarding the discussion in R1C8 and R1C16, which are explaining the two different sources for nitrate as regional and transported are recognized in our study. The correlation of nitrate and LO-OOA is also discussed in lines 458-459 (preprint version) mostly for winter time, which can confirm the long-range transported origin of LO-OOA. Therefore, the following section has been revised.

Line 540-542 (revised version): Furthermore, since nitrate could be originated locally or arrived from a long distance to Melpitz (Sect. 3.1.1), with a good correlation between LO-OOA and nitrate ($R^2=0.59$) during winter, the long-range transported LO-OOA from different directions reaching to measuring site could be explained (Fig. 7).

Technical corrections:

R1C18: L23, the sentence “Melpitz represents due to its location the Central European aerosol” reads odd. Please reword it.

AC18: The text was revised as follows:

Line 19-24 (revised version): Here, the chemical composition and organic aerosol sources of submicron aerosol particles measured by an aerosol chemical speciation monitor (ACSM) and a multi-angle absorption photometer (MAAP) were investigated at Melpitz from September 2016 to August 2017. The location of the station at the frontier between Western and Eastern Europe makes it the ideal place to investigate the impact of long-range transport over Europe. Indeed, the station is under the influence of less polluted air masses from westerly directions and more polluted continental air masses from Eastern Europe.

R1C19: L123, please give a full name of the acronym when it appears for the first time.

AC19: We corrected the text and wrote the full name of PNSD as ‘particle number size distribution’ in line 139 of the revised version.

R1C20: L248, Fig. S3→Fig. 3?

AC20: Corrected. It is ‘Fig. S8’, line 307 revised version.

R1C21: L301, ...in night time, ...

AC21: Corrected: ‘in night time’, line 365 revised version.

R1C22: Fig.S2, the pixel resolution in Fig. S2 is rather poor. What does the radius axis in each rose plot represent?

AC22: Fig. S2 from pre-print version was replaced with high resolution Fig. S3 in the supplementary. The radius axis represents the wind speed in each plot.

R1C23: L372, Fig. 11?

AC23: We have removed ‘Fig. 11’ in the text since it was a mistake and there is no Fig. 11.

R1C24: L375, Fig. 4b→Fig.4c

AC24: Thank you for the point, however there is no Fig. 4c in the manuscript.

R1C25: L510, Fig. S3 and S4→Fig. S4 and S5?

AC25: The numbers of tables have been corrected: ‘Tables S4 and S5’, line 574 revised version.

R1C26: L579, Fig. 11?

AC26: The number of figures has been corrected as ‘Fig. S4’, line 648 revised version.

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