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 #2:

Reviewer #2 (R2):

The authors thank the reviewer for providing constructive comments and insightful suggestions on the manuscript. We highly appreciate your time in reviewing the manuscript. The point-by-point response to all the comments and suggestions of reviewer #2 (R2) is provided in the following sections. For easy visualization, the reviewer's comments (R2 C) are provided in blue and the author's response (AR) is given in black color below the reviewer's comment. All the comments/suggestions were taken into consideration and incorporated in the revised manuscript which has improved the quality of the revised manuscript. The revised parts of the manuscript along with the references are shown in red.

Review on article "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.

Article is based on year-long dataset where chemical composition is broadly characterized using ACSM and MAAP and source apportionment used to characterize the sources of PM. Article is easy to follow and understandable. However, the major issue seems to be that most of the data is already published by previous publications, at least Chen et al., 2022 (source apportionment at least for ACSM) and Poulain et al., 2020 (composition, source areas). If this is the case, it would be utmost important to firstly clarify and explain in experimental section explain the differences between these articles and in more detail give the novelty value of this dataset throughout the manuscript. It is clear that all results from 22 datasets are not covered by Chen et al, but at the moment without reading both side-by-side it is not clear what is. Also, the numbers (e.g. average mass and contributions) seem to be slightly different between this and Chen et al. article, which to me is not clear why. Was the data re-analyzed? In general, I think there are some nice results from the BC source apportionment and cluster analysis, but it is not exactly clear what is covered by previous articles, especially Poulain et al., 2020.

I propose re-writing/re-focusing this article so that it firstly identifies the novelty value and focuses on that more clearly. In this large and interesting dataset, this should not be a large issue and may not be that large of a work. Finally, the language should be checked thoroughly before resubmitting.

AC: We thank the reviewers' comments, however, we must emphasize some misunderstanding regarding the citing papers mentioned above. We agree that the source apportionment developed in this paper was already integrated into the European ACSM/AMS overview paper from Chen et al (2022). The main part of the Chen et al (2022) methodological paper was to establish the best practice for rolling windows PMF analysis. For this purpose, the results obtained on 22 different ACSMs over Europe were compared in a phenomenology discussion to first emphasize the robustness of the approach and second discuss the differences between the type of station (urban and rural-background) over Europe. However, Chen et al., (2022) did not discuss how the different sources were identified, nor depict the change of the entire chemical composition of the particles over the season (organic, inorganic, black carbon), nor the influence of the meteorological conditions at any station (especially the impact of air mass trajectory), nor the link between the organic aerosol and the black carbon to discuss the different source of black carbon, which all of them are the main focus of the present manuscript. Therefore, the present work provides valuable additional information compared to Chen et al., 2022, which is essential to better understand the complexity of the air mass dependency on air quality in central Europe.

Moreover, we think that the reviewer mixed up two of our previous papers. The cited paper Poulain et al. (2020, published in AMT) was focused on the quality assurance of the ACSM measurements made at Melpitz. In that paper, the ACSM data were systematically compared with collocated measurements including MPSS, off-line PM_1, and $PM_{2.5}$ high-volume filter samples for water-soluble ions (sulphate,

nitrate, and ammonium) as well as OC/EC. In Poulain et al. (2020) paper, neither source apportionment nor wind direction or air mass trajectory analysis was performed. In another paper from Poulain et al. (2021, ACP special issue HCCT-2010), source apportionment analysis on AMS data was published as well as some air mass cluster analysis. However, the measurements were not performed at Melpitz station. It is possible that the reviewer confuses the three following papers:

- Poulain et al., 2020: https://doi.org/10.5194/amt-13-4973-2020, The ACSM robustness, quality assurance, and impact of upper size cutoff diameter have been discussed at the same station, Melpitz.
- Poulain et al., 2021: https://doi.org/10.5194/acp-21-3667-2021, The sources, and impact of long-range transport on carbonaceous aerosol have been discussed in this study for Central Germany during HCCT-2010.

We appreciate reviewer spotted the difference, between the current study and Chen et al., (2022), The BC data were accidentally applied CDCE in Chen et al. (2022), which made it around 2 factors higher than current paper. Therefore, it causes the differences in fractions of PM_1 compositions. The co-authors of this paper (also the first author and corresponding author of Chen et al. (2022)) will try to correct this mistake soon.

Detailed comments:

R2C1: Abstract: lines 16-25, maybe consider condensing this text. It repeats some things several times.

AC1: Thank you for asking to condense this text. Lines 16-25 of the preprint version are rewritten as follows:

Line 16-21 (revised version): To better understand their sources, investigations have been focused on urban areas in the past, while rural-background stations are normally less impacted by surrounding anthropogenic sources. Therefore, they are predisposed for studying the impact of long-range transport of anthropogenic aerosols. 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.

R2C2: Line 47: PMs -> PM fractions

AC2: Since we are using 'fractions' as 'contribution' during the whole manuscript, we decided to use 'PMs' which means all the particulate matters, to prevent confusion later on. However, the following part of manuscript have been revised:

Line 45-49 (revised version): The submicronic particles known as PM_1 (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.

R2C3: Line 48: please consider adding another reference to epidemiological study that links PM and health more broadly. Several articles exist (e.g. Pope, C. A. and Dockery, D. W.: Health Effects of Fine Particulate Air Pollution: Lines that Connect, J. Air & Waste Manage. Assoc 56, 35, 2006.)

AC3: Thank you for the comment and suggestion. We agree with the reviewer which PM health effect is not negligible. Therefore, we added the suggested reference to this statement: 'Pop and Dockery, 2016', line 47 revised version.

R2C4: Line 63: was -> has been

AC4: Thank you for the correction. The verb 'has been', line 63 revised version.

R2C5: Lines 85-90: As the source apportionment is published already by Chen et al., article, I would recommend re-defining this scope. As Chen et al. had 22 datasets, it is likely that not many details were given for each site, but the basic results of source apportionment e.g. timeseries of factors were given there. The novelty value is not clear.

AC5: Thank you for the comment and recommendation. The overview paper by Chen et al., (2022) focused on, first: the determination of a unified rolling PMF analysis approach and its validation at different sampling sites; and second: the comparison of the absolute sources at the different European sites. In the present work, we considered the entire aerosol chemical composition not only the organic fraction, with a specific focus on the influence of the wind direction and air mass on the chemical composition. Furthermore, Chen et al., (2022) only focused on the identification of the organic sources at each sampling site, while our study also investigated the sources of eBC as well with three anthropogenic sources related to eBC. This last approach is important since the currently used aethalometer model is limited to splitting the eBC in contributions of fossil fuel (eBC_{ff}) and wood burning (eBC_{wb}). Taking it all together, we agree that the final results of the organic PMF analysis were already presented in Chen et al (2022). But the present manuscript provides a significant additional and detailed discussion on the organic sources as well as new results to make it a stand-alone work without repeating the result discussed in the previous work.

The following text is revised in the introduction section for the mentioned paragraph in order to clarify the novelty of this study in comparison to Chen et al., 2022 and other publications as well:

Line 84-95 (revised version): The current study comprehensively investigates the PM_1 aerosol particle chemical compositions and the various OA sources for Melpitz as a rural-background station, based on ACSM and multi-angle absorption photometer (MAAP) measurements from September 2016 to August 2017, using the most advanced rolling PMF with ME-2 implemented in the SoFi Pro package (Datalystica Ltd., Villigen, Switzerland) (Parworth et al., 2015; Canonaco et al., 2013; Canonaco et al., 2020). Although previous papers already considered this dataset, they were focused on quality assurance (Poulain et al., 2020) to depict the European aerosol chemical composition (Bressi et al., 2021 and Chen et al., 2022) or the relationship between the CCN properties (Wang et al., 2022, Schmale et al., 2017), none of these papers were focused on carbonaceous source identification (OA and eBC) nor discussed the strong dependency of the aerosol chemical composition to the air mass origin. Therefore, a multi-linear regression model was used to estimate the contribution of equivalent black carbon (eBC) to the various primary organic PMF factors such as hydrocarbon-like organic aerosol, biomass burning organic aerosol, and coal combustion organic aerosol. Meanwhile, to better understand the emission area of PM_1 chemical composition and PMF factors, the influence of air mass origin was investigated based on self-developed back-trajectory cluster methods (BCLM).

R2C6: Chapter 2. experimental. Clearly explain what is previously published and what is done first time in this article so reader can understand the novelty value of this article.

AC6: Thank you for the comment. The previous paper (Chen et al., 2022) presented the developed rolling PMF model by validating it on the various data from different stations and also discussed the absolute sources of the organic and inorganic stations with no regard for geographical origin. While the scope of current study is to provide detailed descriptions of chemical composition and OA sources with additional evidences of long-range transportation airmasses, also the eBC-PM_1 fraction was discussed.

The following text is revised in the introduction section for the mentioned paragraph in order to clarify the novelty of this study in comparison to Chen et al., 2022 and other publications as well:

Line 84-95 (revised version): The current study comprehensively investigates the PM_1 aerosol particle chemical compositions and the various OA sources for Melpitz as a rural-background station, based on ACSM and multi-angle absorption photometer (MAAP) measurements from September 2016 to August 2017, using the most advanced rolling PMF with ME-2 implemented in the SoFi Pro package (Datalystica Ltd., Villigen, Switzerland) (Parworth et al., 2015; Canonaco et al., 2013; Canonaco et al., 2020). Although previous papers already considered this dataset, they were focused on quality assurance (Poulain et al., 2020) to depict the European aerosol chemical composition (Bressi et al., 2021 and Chen et al., 2022) or the relationship between the CCN properties (Wang et al., 2022, Schmale et al., 2017), none of these papers were focused on carbonaceous source identification (OA and eBC) nor discussed

the strong dependency of the aerosol chemical composition to the air mass origin. Therefore, a multi-linear regression model was used to estimate the contribution of equivalent black carbon (eBC) to the various primary organic PMF factors such as hydrocarbon-like organic aerosol, biomass burning organic aerosol, and coal combustion organic aerosol. Meanwhile, to better understand the emission area of PM_1 chemical composition and PMF factors, the influence of air mass origin was investigated based on self-developed back-trajectory cluster methods (BCLM).

R2C7: Line 121: Please explain the conversion, not everybody have time to check the reference "Conversion of the eBC mass concentration from the PM10 inlet to the ACSM PM1 cut-off was made by applying a correction factor of 0.9 following Poulain et al (2011)." and add discussion why the eBC measured for PM10 is shown in plots with ACSM that measures mainly submicron particles. Also, to the ACSM chapter would be good to add the size range of particles that ACSM measures.

AC7: We apologized for the confusion. It is true that the MAAP is measuring the PM_{10} eBC, while the ACSM has a near PM_1 cut-off. Therefore, soot concentration for PM_1 is required to perform a proper PM_1 mass closure. To better understand the split of the eBC between PM_1 and PM_{10}, temporary but parallel measurements using two MAAPs, one connected to the PM_{10} inlet and a second one to a PM_1 inlet were performed at Melpitz. This comparison demonstrated that the soot concentration in PM_1 is around 90 % of that in PM_{10}, with this ratio being only weakly time-dependent. Consequently, for this study, we estimated soot in the PM_1 by multiplying the soot concentration on PM_{10} by a constant factor of 0.90. In the entire manuscript, the reported eBC corresponds to the estimated eBC in the PM_1 range. To avoid further confusion the text was changed as follows:

Line 135-138 (revised version): The eBC mass concentration from the PM_{10} data was multiplied by a constant factor of 0.9 following Poulain et al (2011) to estimate the eBC mass concentration in the PM_1 fraction. Consequently, all the eBC mass concentrations reported and discussed here correspond to the eBC in the PM_1 fraction and are referred to as eBC-PM_1.

R2C8: Line 129: very long sentence. also, the sentence speaks both about levoglucosan and monosaccharide anhydrides in plural. maybe clarify if you measured only levo or also galactosan and mannosan.

AC8: Thank you for the comment. All the other sugars such as galactosan and mannosan are measured by HPAEC-PAD, but since we only used levoglucosan in our study, we just kept levoglucosan in this part.

Line 146-148 (revised version): Levoglucosan as a tracer for wood burning combustion was measured following Iinuma et al., (2009) using high performance anion exchange chromatography coupled with an electrochemical detector (HPAEC-PAD).

R2C9: Line 139-140: please give the version numbers for both SOFI-pro and igor. in future, it usually helps to know which version was used, because they tend to improve and usually some bugs are fixed.

AC9: Thank you for the suggestion and points. We have included the number of versions for both SoFi and Igor Pro in the sentences as follows:

Line 154-156 (revised version): The PMF method was used to allocate the source of the OA (Paatero and Tappert, 1994) through the Source Finder professional (SoFi Pro, version 8.0.3.1, Canonaco et al., 2021) software package (Datalystica Ltd., Villigen, Switzerland), within the Igor Pro software environment (Igor Pro, version 8.04, Wavemetrics, Inc., Lake Oswego, OR, USA).

R2C10: Line 206: maybe compare also to long-term dataset presented by Poulain et al., 2020?

AC10: Thank you for the comment. As the referee suggested, we made a comparison between our study and Poulain et al., (2020) with data period from June 2012 to November 2017 and similar values for mean mass concentration and seasonal trend could be observed. The following line has been added to the mentioned section:

Line 248-253 (revised version): Compared to previous ACSM long-term measurements of Poulain et al., (2021) at the same station, a similar mean mass concentration of PM_1 was observed in the period from June 2012 to November 2017 (Poulain et al., 2021: 10.23 µg/m^3 and this study: 10.49 µg/m^3; respectively), and presented same seasonal trends for all the chemical species (Table. S2) with a highest mass concentration in the winter and lowest mass concentration in the summer time (13.15 µg/m^3 and 7.64 µg/m^3, respectively; Table. S2). Consequently, the results obtained from the current study can be considered as a representative ACSM study for Melpitz station.

We also added the Table. S2 to the Supplementary file:

Table. S2: PM_1 seasonal mass concentration (µg/m^3) of Poulain et al, (2021), and average from the current study.

Species	Fall	Winter	Summer	Spring	Average	the current study
Org	3.83	4.58	4.41	4.28	4.27	4.84
SO_4^{2-}	1.53	1.86	1.37	1.41	1.54	1.67
NO3-	2.24	3.79	0.90	3.07	2.50	2.16
NH_4^+	1.10	1.63	0.65	1.35	1.18	1.11
Cl-	0.04	0.07	0.01	0.05	0.04	0.05
eBC-PM_1	0.69	1.22	0.30	0.56	0.69	0.66
Tot	9.43	13.15	7.64	10.72	10.23	10.49

R2C11: Line 224-227. Please clarify what this means and why these measurement periods are important?

AC11: Thank you for the comment. The discussed lines are: "Moreover, with enhanced irradiations in summer, sulphate formation from photochemistry could be enhanced as well. This result is consistent with the mean PM_1 mass concentration measured by AMS for the three periods during fall (16. September.2008 to 03. November.2008), winter (24. February.2009 to 25. March.2009), and summer (23. May.2009 to 09. June.2009) campaigns reported by Poulain et al., (2011)." in the preprint version. We are going to answer this comment in two parts:

- Part one: Since the photochemical oxidation process of sulphur dioxide is direct cause of the increment of sulphate in the atmosphere, therefore sulphate can increase during the summer due to the high solar radiation.
- Part two: To make a comparison of the Melpitz study between the present study with an AMS study from Poulain et al., (2011), the three periods have been discussed since the measurement campaign of Poulain et al., (2011) took place on these three limited periods which covered different parts of the seasonal cycle, we also took the same period of our data, and compared them. It showed that the sulphate contribution to total PM_1 is higher in the summer time rather than in winter time due to the photochemical oxidation process.

Furthermore, we made a comparison between our study and Poulain et al., (2020) with a long-term ACSM dataset (from June 2012 to November 2017) and similar values for sulphate mean mass concentration could be observed, (15 % in Poulain et al., and 16 % in the current study).

To avoid the confusion, we have revised the discussed section and the following line has been added to the mentioned section:

Line 274-282 (revised version): Moreover, the sulphate contribution to the total PM_1 was higher during the summer than winter time, since with enhanced irradiations in summer, sulphate formation from photochemistry could be enhanced as well. This sulphate higher contribution in summer over winter is consistent with the mean PM_1 mass concentration measured by AMS for the three periods during fall (16. September.2008 to 03. November.2008), winter (24. February.2009 to 25. March.2009), and summer (23. May.2009 to 09. June.2009) campaigns reported by Poulain et al., (2011). In comparison with previous ACSM long-term measurements of Poulain et al., (2021) at Melpitz station, a similar mean mass concentration of sulphate was observed in the period from June 2012 to November 2017 (Poulain et al., 2021: 1.54 µg/m^3 and this study: 1.67 µg/m^3; respectively; Table. S2). This comparison indicates the current study as a case study of ACSM for Melpitz station within 5-year ACSM data, with the best data coverage of time in a year.

R2C12: Line 209-211: "Fig. S2 presents the coming high polluted air masses for total PM_1 to the measurement site in the current study; the polluted Eastern Europe flow with high mass concentration and south-west with low mass concentration was more clearly found in winter time rather than in other seasons, which will be comprehensively discussed in the Sect. 3.4." should it be e.g. "lower mass concentration for SW", as the lowest were the for north west and south.. I am struggling to understand where this is compared.

AC12: Thank you for the comment. We compared the result of the Melpitz station with other rural-background stations from two other publications (Bressi et al., 2021 and Chen et al., 2022) throughout Europe, lines 257 and 261 of the revised version. From the comparison, we found that there is a similarity in annual PM_1 mean mass concentration between Melpitz and other rural-background stations in mid-latitude Europe.

We also used 'lower' instead of 'low' in line 254 revised version.

R2C13: Line 212: maybe here also comparison to long-term concentrations shown by Poulain et al., 2020? this would give reader an idea whether this chosen year was typical or an anomaly.

AC13: Thank you for the comment. As the referee suggested, we made a comparison between our study and Poulain et al., (2020) with data period from June 2012 to November 2017 and similar values for mean mass concentration and seasonal trend could be observed. The following lines have been added to the mentioned section:

Line 248-253 (revised version): Compared to previous ACSM long-term measurements of Poulain et al., (2021) at the same station, a similar mean mass concentration of PM_1 was observed in the period from June 2012 to November 2017 (Poulain et al., 2021: 10.23 µg/m^3 and this study: 10.49 µg/m^3; respectively), and presented same seasonal trends for all the chemical species (Table. S2) with a highest mass concentration in the winter and lowest mass concentration in the summer time (13.15 µg/m^3 and 7.64 µg/m^3, respectively; Table. S2). Consequently, the results obtained from the current study can be considered as a representative ACSM study for Melpitz station.

We also added the Table. S2 to the Supplementary file:

Table. S2: PM_1 seasonal mass concentration (µg/m^3) of Poulain et al, (2021), and average from the current study.

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NO3-	2.24	3.79	0.90	3.07	2.50	2.16
NH_4^+	1.10	1.63	0.65	1.35	1.18	1.11
Cl-	0.04	0.07	0.01	0.05	0.04	0.05
eBC-PM_1	0.69	1.22	0.30	0.56	0.69	0.66
Tot	9.43	13.15	7.64	10.72	10.23	10.49

R2C14: Line 232. clarify how do you separate locally formed sulphate from any other source?

AC14: We thank the reviewer for the comment. The non-parametric wind regressions model (NWR) had been used to investigate not only the local but also the transported emission sources (Marin, et al., 2019) in Sect. 3.1-3.3. With this regard, if high concentrations are observed at low wind speeds, they are recognized as local sources, whereas high concentrations at high wind speeds are more transported ones; which means: when there is almost no wind, the concentration is local and spread throughout the station, whereas when there is more wind, the concentration is systematically associated with a specific wind sector, which may correspond to the transport process. 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.

R2C15: Line 258: what is the detection limit for chloride in ACSM? is 0.05 µg m-3 above dl?

AC15: The mean mass concentration for chloride in this study is 0.05 µg/m^3, which is above the chloride detection limit, < 0.011 µg/m^3 is the detection limit for chloride (Ng et al., 2011).

R2 C16: Chapter 3. should the name be results and discussion?

AC16: Thank you for the suggestion. The title of Sect. 3 has been changed to 'Results and discussion' as suggested, in line 236 (revised version).

R2C17: For chapters 3.1.1 and 3.1.2. I am bit struggling to see the novelty value. Please highlight the new results and their meaning/impact for science renewal.

AC17: Thank you for the comment. The mentioned Sect. (3.1.1 and 3.1.2) are discussions around ACSM and MAAP data analysis. The seasonality of chemical compositions in individual sites were not really covered in Chen et al. (2022). Also, the long-range transportation influence to this specific site was not investigated by Chen et al. (2022), therefore, giving the special location of Melpitz, the discussion in 3.1.1 and 3.1.2 are important to address these questions. In order to highlight the novelty of this manuscript, we have revised the following lines in the introduction section:

Line 84-95 (revised version): The current study comprehensively investigates the PM_1 aerosol particle chemical compositions and the various OA sources for Melpitz as a rural-background station, based on ACSM and multi-angle absorption photometer (MAAP) measurements from September 2016 to August 2017, using the most advanced rolling PMF with ME-2 implemented in the SoFi Pro package (Datalystica Ltd., Villigen, Switzerland) (Parworth et al., 2015; Canonaco et al., 2013; Canonaco et al., 2020). Although previous papers already considered this dataset, they were focused on quality assurance (Poulain et al., 2020) to depict the European aerosol chemical composition (Bressi et al., 2021 and Chen et al., 2022) or the relationship between the CCN properties (Wang et al., 2022, Schmale et al., 2017), none of these papers were focused on carbonaceous source identification (OA and eBC) nor discussed the strong dependency of the aerosol chemical composition to the air mass origin. Therefore, a multi-linear regression model was used to estimate the contribution of equivalent black carbon (eBC) to the various primary organic PMF factors such as hydrocarbon-like organic aerosol, biomass burning organic aerosol, and coal combustion organic aerosol. Meanwhile, to better understand the emission area of PM_1 chemical composition and PMF factors, the influence of air mass origin was investigated based on self-developed back-trajectory cluster methods (BCLM).

R2C18: For the chapter 3.1.1.: please add other references also, outside Melpitz.

AC18: Thank you for the comment and suggestion. As the referee suggested, we compared the present study with other rural-background stations from Bressi et al., (2021) which only presented the winter and summer results. From the comparison of diurnal profiles between two studies for Melpitz station (Fig. S4 from Bressi et al., 2021 and Fig. 3 from our study) with different time coverage, all the main chemical compositions (named below) showing the similar patterns:

- Sulphate presents an increase during the day and decreased in the night time for summer the season, with no significant changes in the winter season.
- Nitrate and ammonium present a decreasing pattern during the day, and an increasing pattern during the night time in both summer and winter time.
- Organic also shows similar patterns, decreasing during the day and increasing during the night in both summer and winter time.

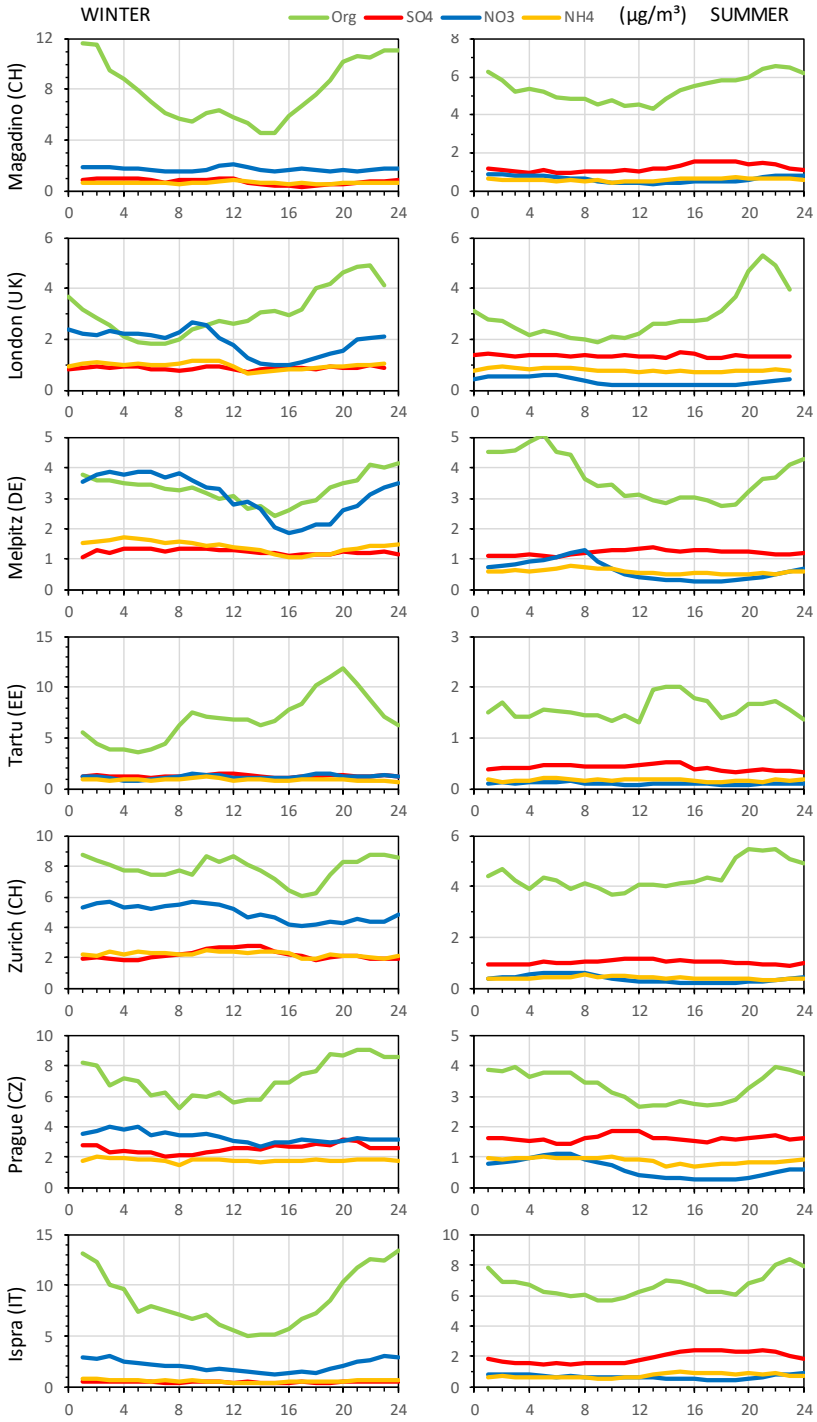

Figure S4 from Bressi et al., (2021): daily cycles (median values) in the 4 main components of NR-PM$_1$ concentrations (µg/m^3) measured at the 21 selected sites in winter (DJF) and summer (JJA).

Fig. 3: Seasonal diurnal cycle of PM_1 for ACSM organic and inorganic species (Time is in UTC).

In comparison with other rural-background stations, there is no similarity of the main chemical composition in each diurnal profile of the different stations, because every station has particular conditions such as geographical location, different types of sources, and various meteorological conditions.

We have added other stations to make a better comparison between rural-background stations in Europe as follow in Sect. 3.1.1 and 3.1.2:

Line 265-270 (revised version): Moreover, the comparison between Bressi et al., (2021) and current study (Fig. S4 from Bressi et al 2021, Fig. 3 from the current study) for Melpitz station with different time coverage shows that the daily variation of ACSM sulphate, nitrate, and ammonium are similar in both winter and summer seasons. In comparison with other ACSM/AMS rural-background stations in Europe (Fig. S4, Bressi et al., 2021), the mean daily cycle of the PM_1 chemical components (sulphate, nitrate, and ammonium) does not show a similar pattern to the other stations (Bressi et al., 2021) due to the different geographical location and meteorological conditions.

In addition to Sect. 3.1.1, we also added a similar comparison for the organic part in Sect. 3.1.2 as follows:

Line 371-374 (revised version): In comparison between Bressi et al., (2021) and the current study for Melpitz station, the daily variation of organic are similar in both winter and summer seasons, while there are differences between Melpitz with other rural-background stations due to the different geographical location and meteorological conditions (Bressi et al., 2021).

R2C19: Chapter 3.2. Source apportionment. I think quite many of the things discussed here (like factor identification) should go to experimental or supplementary material. These are fairly well known and are not results. I am struggling to find the novelty value of this chapter. The source apportionment is extensively discussed in literature, and results seem to be in-line with previous research results. Please, try to highlight the novelty further.

AC19: Thank you for the comment. Sect. 3.2 which is named 'source apportionment of OA', explains each of the organic aerosol sources by starting with a 2-3 lines definition of them. This is true that factor identification is already known and discussed in previous literatures, but we are explaining every PMF

factor as a result of this study by a definition of them which are not only strongly linked to the following statements, such as correlation with specific tracer, but in a way that it can also make it understandable for other readers outside of PMF/AMS-ACSM field as well. Furthermore, for some of them like OOAs, it is important to define the *m/z* 44 and 43, since the discussion cannot be completed without mentioning the *m/z* at the beginning of result discussion, and triangle plots could be discussed without these definitions. With respect to the reviewers' comment, we think that with 2-3 sentences about the factor identification, we can explain the PMF factors results with such details in a more reliable way.

R2C20: Line 359:" The high value of BBOA" I would rephrase to be "high concentration. "

AC20: Thank you for the suggestion. As suggested, the term 'high value' is changed to 'high mass concentration', line 436 (revised version).

R2C21: Chapter 3.3. Very interesting approach. I suggest moving the method description and formulas to the experimental and leaving here results and discussion. Please, try to discuss the value of results here also.

AC21: We thank the reviewer for the comment and suggestion. We moved the method description and formulas of eBC-PM_1 source apportionment to the experimental section (lines 195-212 revised version) under the title of '2.5 eBC-PM_1 source apportionment', and we left only its result in the 'Result and discussion' as suggested by reviewer: 3.3 Source apportionment of eBC-PM_1, lines 550-566 revised version.

Here by the multilinear regression model, we identified the contribution of primary organic aerosols named HOA, BBOA, and CCOA, on eBC-PM_1 for two reasons:

- First, previous literatures were not investigating the sources and emission area of carbonaceous aerosol based on OA-PMF sources,
- Second, eBC can be split in eBC_{Cff} and eBC_{wb} based on aethalometer model which means it can only consider there two sources, and no more.

To this order, we added the following paragraph to the manuscript.

Line 91-94 (revised version): Therefore, a multi-linear regression model was used to estimate the contribution of equivalent black carbon (eBC-PM_1) to the various primary organic PMF factors such as hydrocarbon-like organic aerosol, biomass burning organic aerosol, and coal combustion organic aerosol.

R2C22: Line 483: "CCOA appeared to have the largest source of eBC," I would suggest rephrasing to be CCOA appeared **to be** the largest source of eBC",

AC22: Thank you for the suggestion. As suggested, the term 'to have' is changed to 'to be' in the text, line 552 (revised version).

R2C23: Chapter 3.4 the titles could be more informative.

AC23: Thank you for the comment. The title of Sect. 3.4 has been changed from 'Impact of air mass origin and trajectory analysis' to 'Seasonal air mass clustering', Line 567 (revised version).

R2C24: Chapter 3.4. also, the technical description of cluster analysis and identified clusters would be more suitable for experimental than results. Also, please give details of the analysis, based on what the clusters were identified and how long data was used. figure legend says 13-years, text is speaking of a year.

AC24: Thank you for the comment. As the reviewer suggested, the technical description of cluster analysis has been moved to the experimental section (lines 229-235, 2.6 Air mass trajectory analysis) which is included detailed information on this technique. We apologize for the confusion about the Fig. 9 legend and manuscript text, which is corrected as follows:

Line 1198 (revised version): a) air mass classification based on one-year backward trajectories cluster analysis at 12:00 UTC.

Now in Sect. 2.6, line 229-235 (revised version): In this method, the different clusters can be divided according to the different seasons (CS: cold season; TS: transition season; and WS: warm season), and meteorological synoptic patterns (ST: stagnant; A1: anticyclonic with air mass coming from Eastern Europe; A2: anticyclonic with air mass coming from the west; C1: cyclonic with air mass coming from relatively south; C2: cyclonic with air mass coming from the west and south west). However, the clustering approach did not consider spring and fall separately, and therefore the transition clusters correspond to both spring and fall. Finally, a total of fifteen clusters were identified, corresponding to different meteorological conditions over the course of the year. Descriptive analysis, cluster processing, and data processes and products are all described in detail by Sun et al., (2020) and Ma et al., (2014).

Now in Sect.3.4, line 568-574 (revised version): As mentioned before, the geographical origin of the PM_1 chemical species and also PMF components are not only emitted from the surrounding area but also transported. Therefore, to better identify the origin of their sources, trajectory analysis, and their clustering analysis were applied using the self-developed back-trajectory cluster method (BCLM) (Sun et al., 2020; Ma et al., 2014; Hussein et al., 2006). Regarding this cluster approach, six air masses were identified at Melpitz station for the winter season, four air masses for the transition seasons, and five air masses for the summer season (Fig. 9a). The number of clusters with their corresponding mean mass concentration of PM_1 chemical species and PMF factors of organics are summarized in Table. 2 and with more details in Tables S4 and S5.

R2C25: Line 519: clarify what means "surrounding emission origin". is it like very local, regional or even LRT?

AC25: Thank you for the comment. 'Surrounding' corresponds to the local emissions, which means emissions from Melpitz village itself, around the measuring station as well as short-distance transport (that can be emissions from Leipzig and Torgau cities).

Furthermore, during winter time, one of the identified air masses from the BCLM model for Melpitz station which is recognized based on the synoptic patterns is CS-ST (which means cold season-stagnant), presenting the stable air condition around the station. In this situation, air masses can be known as surrounding emissions, and when we want to point to their origin, it has been called surrounding emission origin. It is similar to WC-ST air mass which means warm season-stagnant. To better understated and avoid the complexity and confusion in this part, we have revised the following statement:

Line 579-580 (revised version): These surrounding emissions refer to the emissions from Melpitz station directly, Melpitz village, and short distance transported particles like particles from Leipzig and Torgau.

R2C26: Line 516-517: "This cluster with the highest mass concentration of LO-OOA to the PM mass (2.73 µg/m3) could confirm the role of freshly formed SOA originating around the station from primary biomass burning and coal combustion emissions (mass concentration of 0.97 µg/m3 and 1.89 µg/m3, respectively)." please clarify this conclusion. Also, it is surprising that this happens in winter when UV is lowest. Did you see same in summer?

AC26: Thank you for the comment. SOA is considered to be formed by biomass burning and coal combustion as well, especially during the winter time when biogenic emissions are minimal (Lanz et al., 2010). In fact, the Aqueous-phase processing of biomass-burning emissions contributes to SOA formation, which this aqueous SOA absorbs both UV and visible light more efficiently that other OA components (Gilardoni et al., 2016). Furthermore, under dark conditions, biomass burning emissions age rapidly in the presence of NO_2 and O_3, producing a similar amount of SOA as under photochemical conditions (Kodros, et al., 2020). Therefore, with a high value of ozone during night time (Fig. S4), the dark aging chemistry can cause SOA formation in the winter season.

In order to clarify the mentioned statement, we have revised this section as follows:

Line 580-585 (revised version): This cluster presented the highest mass concentration of LO-OOA to the PM mass (2.73 μg/m^3). In fact, SOA is considered to be formed by biomass burning as well as coal combustion, particularly during the winter when biogenic emissions and UV radiation are low (Lanz et al., 2010; Kodros, et al., 2020). In this condition and in the presence of NO_2 and O_3, the biomass burning emissions could age rapidly and produce SOA. In conclusion, this cluster could confirm the role of freshly formed SOA which originated from the primary biomass burning and coal combustion emission (mass concentrations of 0.97 μg/m^3 and 1.89 μg/m^3, respectively).

R2C27: Line 617: please add better compared to what? also, add how you observed the improvement

AC27: Thank you for the comment. The mentioned statement is pointing to the advantage of the rolling approach compare to the regular PMF, which is performed over the entire dataset with the assumption that the OA profiles are static which can result in high errors for a long-term dataset because OA chemical fingerprints are expected to change over the time. Instead, in the rolling technique a small-time window moves in daily steps across the whole dataset instead of running PMF on the entire dataset. Therefore, the model can slowly adjust the factor profiles over various periods, which provides well-separate OA factors.

Line 687-690 (revised version): For OA source apportionment, PMF in a rolling fashion has been applied using the SoFi Pro, which provided the decomposition of time-dependent factor profiles that were able to better capture the variability of OA sources across seasons in comparison with the conventional seasonal PMF.

R2C28: Conclusions: please try to highlight the novelty of the results

AC28: We thank the reviewer for asking to highlight the novelty of the results, the new findings of the study are summarized as follows.

i. In addition to OA source apportionment, the eBC-PM_1 source apportionment has been studied using a multilinear regression model based on primary organic aerosol sources; HOA, BBOA, CCOA. While it is common to determine the contribution of eBC_{Cff} and eBC_{wb} based on aethalometer model which means it can only consider these two sources, and no more.
ii. There have been numerous PMF-based studies in the subject area of OA source apportionment. However, data coverage for most of them is for one year or less and also not for a one-year long continuous including four seasons.
iii. Since most of the previous source apportionment publications studied the urban and urban-background sites, there is still a lack of information about the sources in rural and rural-background sites which can better present the impact of long-range transport of anthropogenic aerosols.
iv. Chen et al. (2022) provided a large overview of the European geographical OA sources but did not discuss the factors influence/control the OA sources at each site, while in the present study, the geographical and influence factor of OA sources has been investigated with NWR model, cluster and back-trajectory analysis.
v. The study expresses the geographical origin of the PM_1 in terms of the origin of chemical composition, and the origin of PMF sources for a rural site. Using the cluster and back-trajectory analysis, the origin of sources results in terms of the different seasons and meteorology conditions.
vi. Various source apportionment analysis has been done in the previous studies over the OA with the PMF approach. From Chen et al. (2022), only two stations reported CCOA and the second one is an urban station with coal combustion sources nearby. While this study investigated the existence of a coal combustion source over the year on Melpitz as a rural site. The cluster and back trajectory analysis are providing the geographical origin of this source for various seasons.

Conclusion: The following lines/paragraphs have been added/modified/rewritten in Sect. 4 “Conclusion” of the revised manuscript to incorporate the suggestions of the reviewer:

Line 682-725 (revised version): The chemical compositions of non-refractory fine aerosol (NR-PM_1) at the German rural-background observatory Melpitz were investigated in this study over a one-year period

between September 2016 and August 2017. Overall, the averaged total PM_1 mass concentration is 10.49 $\mu g/m^3$ and follows a clear seasonal pattern, with the highest mass concentration during winter (15.95 $\mu g/m^3$) and the lowest mass concentration during summer time (6.24 $\mu g/m^3$). The organic aerosol was the most significant component, accounting for 46% of total PM_1 and showing significant seasonal dependency (39 % in winter to 58 % in summer). It was followed by sulphate (15 % and 20 %) and nitrate (24 % and 11 %). For OA source apportionment, PMF in a rolling fashion has been applied using the SoFi Pro, which provided the decomposition of time-dependent factor profiles that were able to better capture the variability of OA sources across seasons in comparison with the conventional seasonal PMF. The final solution enabled the identification of five factors throughout the one-year measurements of OA; HOA, BBOA, CCOA, LO-OOA, and MO-OOA. Using the correlation between HOA, BBOA, and CCOA with eBC-PM_1, a multilinear regression approach was applied to perform the source apportionment of eBC-PM_1.

Generally, in Melpitz, HOA as a minor source of OA (6 % of the contribution of total organic mass) and eBC-PM_1 (8 % of the total eBC-PM_1) was associated with: a) low traffic emissions, b) household heating in winter, and c) the central heating for hot water production for all the seasons which uses multiple fuel types in the Melpitz area. BBOA representing 7.9 % of the contribution of total organic mass and 37 % of the total eBC-PM_1, showed a seasonal effect, emphasizing the impact of house heating during winter. Similar to HOA, the presence of BBOA during summer was due to central heating which uses multiple fuel types in the Melpitz area. The most dominant anthropogenic source was associated with CCOA with a 15.4 % contribution of total organic mass and 55 % of the total eBC-PM_1 with the highest mass concentration and contribution of PM during winter rather than summer. Although a certain fraction of CCOA could be linked to surrounding domestic heating (van Pinxteren et al., 2020), it is rather associated with power plant emissions and long-range transport all year round which is supported by cluster and back-trajectory analysis. LO-OOA and MO-OOA referred to oxidized oxygenated organic aerosol (32.4 % and 38.4 % of the contribution of total organic mass, respectively), were identified as a secondary organic aerosol with the highest mass concentration during the cold months and the lowest mass concentration during the warm months. LO-OOA mass concentration decreased during the day due to dilution, and the evaporation process resulted in aging into MO-OOA.

A combination of the NWR model and cluster analysis was used to better understand the origin of the aerosol reaching the station. Overall, Melpitz is influenced by fifteen types of air masses, such as long-range continental, marine, and surrounding emissions. During winter and summer time, easterly continental air masses, CS-A1 and WS-A1 with an anticyclonic pattern come from Eastern Europe and showed a significant particle mass concentration, especially high POA (and CCOA) mass concentration at the measurement site. Marine clusters, mostly coming from the south/west/north side with aged marine air masses including nitrate and sulphate, also have important roles in the PM mass concentration at the Melpitz site over the entire period (winter: CS-A2, CS-C2b, and CS-C2a, transition: TS-C, TS-A2 and TS-C2, and summer: WS-Ca, WS-C2, and WS-A2). However, the surrounding emissions are recognized as another important source of emissions which include high organic and inorganic components during winter and summer (CS-ST and WS-ST, respectively).

Our results emphasize the importance of the long-range transported emissions of coal combustion related aerosol particles regardless of the season, which supports that the main CCOA source is related to coal power plants emissions. However, coal power plants emissions not only affect the surrounding air quality but can also be transported over long distances. It is important to note that the overall coal combustion mass concentration presented here can certainly be underestimated since the identified CCOA factor is associated with freshly emitted organic aerosol and no factor associated with potential aged coal combustion was identified. Because coal still is an important energy source in the European energy mix (68.4 % of all energy in the EU was produced from coal, crude oil, and natural gas, Energy Statistics - an Overview - Statistics Explained, 2022) as well as on a global scale and also that it still will be in use for the coming decades (until 2040, Europe's Coal Exit - Europe Beyond Coal : Europe Beyond Coal, 2022), further researches should be done on the identification of coal emissions across Europe in order to better understand its atmospheric aging processes.

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