RC1

This study investigates the seasonal composition, sources, and formation mechanisms of submicron aerosols in the Jülich region of Germany based on the JULIAC observation campaign, with a focus on the secondary formation processes of organic aerosols, the influence of regional transport, and the impact of heatwave events on aerosol properties. This study provides important insights into the seasonal variations of aerosols and their environmental effects in the study area. However, some aspects require further clarification or improvement to enhance the scientific rigor and readability of the paper.

Specific comments

1. Line 78-83: The first sentence of this paragraph mentions that "long-term regional transport affects atmospheric conditions as well as aerosol properties", but the subsequent text only focuses on MSA without directly explaining how "regional transport" specifically influences MSA or aerosol characteristics. It is recommended to add one or two sentences clarifying the direct relationship between regional transport and the distribution or concentration of MSA.

   Thanks for this valuable comment. As suggested, we added the following explanatory paragraph in the manuscript line 79-84.

   Draft line 79-84: In addition to local primary emissions and secondary formation, long-term regional transport affects atmospheric conditions as well as aerosol properties. For example, methanesulfonic acid (MSA, CH_3SO_3H) is widely regarded as a tracer for marine air transport because its precursor, dimethyl sulfide (Zorn et al., 2008; Ge et al., 2012), is primarily produced by phytoplankton and anaerobic bacteria in the ocean (Charlson et al., 1987). Regional transport carries MSA inland or to higher altitudes, shaping its atmospheric concentration and distribution, which in turn can influence CCN activity and climate (Yan et al., 2019).

2. This study employs both 24-hour and 72-hour backward trajectories. Please explain the rationale behind using trajectories of different durations.

   In this study, both 24-hour and 72-hour backward trajectory analyses were conducted for all seasons. Generally, the 72-hour trajectories better captured the observed changes in OA properties identified by the PMF analysis. However, an exception is illustrated in Fig. 9c, where the 24-hour trajectories aligned more closely with the MSA–OA source. By contrast, the corresponding 72-hour trajectories in Fig. 11 suggest that the "blue" cluster extended from a purely inland pathway to briefly cross a marine region, which was not reflected in the MSA–OA results. This brief marine influence in the 72-hour cluster may be an artifact of the trajectory clustering. To provide the most consistent interpretation with the observed OA source, the 24-hour trajectories were therefore used in Fig. 9c.

3. Line 199-205: The methodology for estimating liquid water content using the ISORROPIA-II model requires further refinement. Please clarify whether the model operates in forward or backward mode, and elaborate on the impact of missing gaseous pollutants (such as HCl, HNO_3, HNO_2, SO_2, and NH_3) on the model's performance, along with an assessment of the associated uncertainties. For example, this may affect the conclusions in line 286-288.

Thanks for these comments, we added more discussion details in Section 2.6 of the manuscript (see section right below). Since both forward and reverse modes of ISORROPIA-II appear to yield similar predictions for aerosol liquid water content, there is no need to change lines 286–288.

Draft line 221-231: The aerosol liquid water content (ALWC) was estimated using the reverse mode of the ISORROPIA-II thermodynamic model (Fountoukis and Nenes, 2007). It uses measured ambient RH, temperature, and aerosol-phase concentrations to infer gas-phase species and predict thermodynamic equilibrium. This approach is advantageous when gas-phase precursors are unavailable, as in our dataset. By contrast, the forward mode of ISORROPIA-II requires both gas- and aerosol-phase inputs to simulate aerosol composition. However, previous evaluations have demonstrated strong consistency in the ALWC between the two modes for major aerosol components. (Fountoukis and Nenes, 2007) reported normalized mean errors of 3.4 ± 1.1% for aqueous sulfate and 2.5 ± 1.3% for aqueous nitrate, which are the main drivers of ALWC. Since ALWC primarily scales with total aerosol mass, it is relatively insensitive to gas-phase constraints; accordingly, (Guo et al., 2015) observed nearly identical ALWC predictions in both modes (slope = 0.993, intercept = $-0.005\ \mu g\ m^{-3}$, $R^2 = 0.99$). Consequently, relying on the reverse mode ALWC is a solid approach.

4. Section 2.2 “Instrumentation” and Table 1 describe auxiliary instruments (e.g., SMPS, CPC) and report total particle volume/number concentrations, but these data are scarcely discussed in later sections. Please either incorporate relevant analysis or consider removing redundant information for better focus.

   The reviewer’s suggestion is very helpful. We have removed the total number and total volume data from SMPS in Table 1 to place better focus on aerosol composition. Since the SMPS data are still used in the discussion and in Table S6 regarding the possible heterogeneous formation pathways of organosulfate compounds, the instrumentation description of SMPS has been retained.

5. Line 298-309: Although the author mentioned the proportion falling under condition II when pH <0, it should be noted that a non-negligible portion of winter, summer, and autumn data still falls within condition I. The authors pointed out that the organic sulfur calculation method has limited reliability under low pH or high ammonium nitrate conditions (Figure 4), but they do not discuss the potential impact of the resulting errors on the conclusions. It is recommended to supplement the uncertainty analysis or include comparative validation results with other methods.

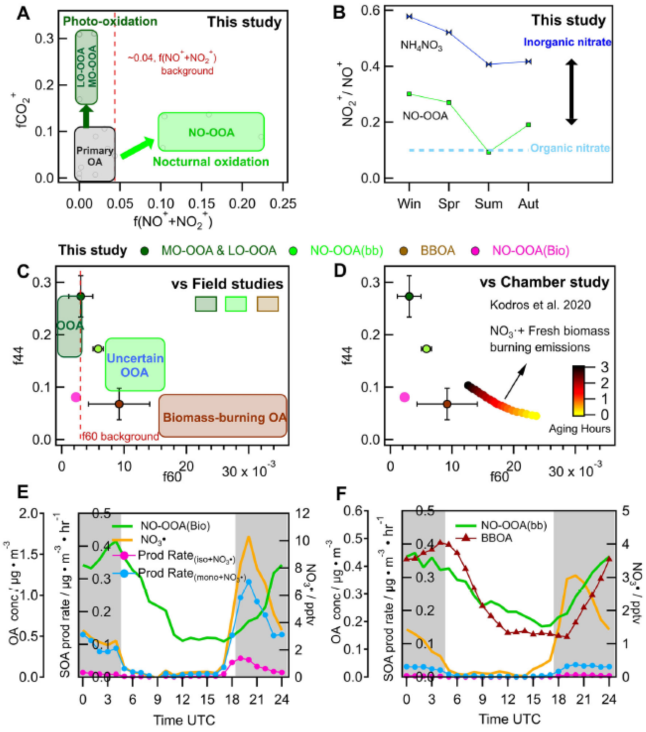
   The bias of the OS calculation method outside condition II was indeed overlooked in the previous discussion and has now been added, as shown below. Unfortunately, it is still difficult to quantify a clear uncertainty range, since the ambient quantification of OS remains an unsolved problem. The fragment method utilized here offers a potential approach, using the fragmentation patterns of standard AS and MSA to roughly estimate OS concentrations from AMS measurements. Schueneman et al. (2021) demonstrated in laboratory studies that the fragmentation pattern of AS changes significantly with increasing acidity and the mass fraction of NH_4NO_3. That study shows if a fixed neutralized AS fragmentation pattern from lab was applied to calculate ambient OS out of range “condition II”, then it would lead to a systematic **underestimation** of OS concentrations.

   Draft line 324-332: The feasibility of the method used here to calculate organosulfate concentrations (Section 2.3) has been previously evaluated (Schueneman et al., 2021). The results indicate that this method can reliably determine ambient organosulfate concentrations when aerosol properties fall within a specific range: aerosol acidity pH > 0 and an ammonium nitrate fraction below 0.3 (corresponding to condition II, marked by red dashed box in Figure 4). Outside of this range, there is a high likelihood that organosulfate concentrations are underestimated. In this study, high concentrations of aerosol organosulfate were found in summer and autumn based on this method, with high reliability indicated by 84–99% of the periods falling within condition II (Figure 4). In contrast, the greatest uncertainty occurs in winter, when only 43% of the measurements meet condition II, suggesting an underestimation of organosulfate concentrations during that season.

6. Line 330-332: LO-OOA and MO-OOA are defined by oxidation degree, while NO-OOA is categorized by time. Could there be potential overlaps between NO-OOA and the other two subtypes? Additionally, is NO-OOA also resolved during daytime? It would be better to provide diurnal variation plot to clarify day-night differences, and supplement the discussion with a comparison of their mass spectral signatures to better distinguish these components.

   Thank you for this comment. The identification and distinct properties of the NO-OOA factor were thoroughly discussed in our previous paper (Liu et al., 2024, DOI: 10.1038/s41612-024-00747-6), where NO-OOA was first introduced as a nighttime SOA formation source. As shown below, in Figures (A) and (C) of that study, NO-OOA displays clear differences compared to LO-OOA and MO-OOA. The diurnal pattern is indeed a key feature for distinguishing daytime (LO-OOA + MO-OOA) from nighttime (NO-OOA) OOA formation, as further illustrated in Supplementary Figures S3–S6, as well as mass spectra of factor. Specifically, LO-OOA and MO-OOA both peak in the late afternoon, whereas NO-OOA peaks at night. For better guidance, the sentence below is added in draft:

   Draft Line 349-351, For the OA concentrations, the contributions of the identified sources are shown, with the complete mass spectra and diurnal patterns of seasonal OA sources provided in Fig. S4–S7



7. Line 352-354: A comparative analysis of NO-OOA and biomass burning tracers across different seasons would help clarify their potential sources and interactions.

Same as above, this is also the critical discussion addressed in our previous paper (Liu et al., 2024, DOI: 10.1038/s41612-024-00747-6). So here we directly cite the conclusion. To make this clear, the corresponding sentence is modified as below.

Draft Line 376-378, The distinct oxidation level of summer NO-OOA, as discussed in the prior study (Liu et al., 2024), is attributed to biogenic precursors driving NO_3-initiated oxidation in summer, in contrast to biomass-burning emissions being the precursors in the other seasons.

8. Figure 7 demonstrates good correlations between NO-OOA and both $C_2H_4O_2$ and CO, it would be valuable to examine whether these relationships vary seasonally.

   Thanks for this comment. However, as described above, the seasonal correlation of NO-OOA is already discussed in our prior paper. The content suggested by the reviewer here is shown in Supplement Table 6 in (Liu et al., 2024, DOI: 10.1038/s41612-024-00747-6).

9. Line 390: The word “for” should be deleted.

   Done.

10. Line 402-405: It would be better to incorporate meteorological data (particularly wind direction/speed) to better identify the sugar factory’s contribution. Additionally, could the relative contributions of the two distinct BBOA sources be further differentiated and quantified?

    Thank you for your comment. A polar plot of the seasonal source contributions has been added to the Supplement (see below), with autumn BBOA shown in row 4, column 2, and BBOA2 in row 4, column 3. Corresponding discussion has also been incorporated into the draft, as outlined below.

    Draft Line 426-430: The most abundant OA contribution from biomass-burning is found in autumn, where two distinct BBOA sources are identified: one dominated by local emissions and the other associated with regional transport (Fig. S2). The local BBOA source is likely related to residential heating and nearby emissions from the sugar factory, which have active production of sugar only during the autumn phase.

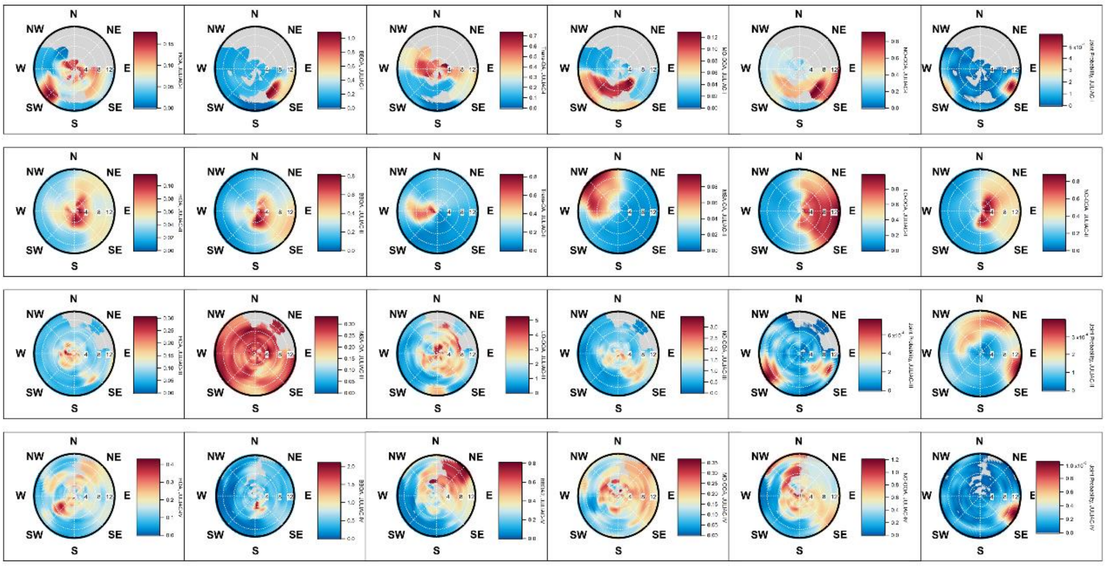


Figure S1: Polar diagrams using non-parametric wind regressions are shown for all JULIAC-intensive phases, illustrating source factors contribution as well as the joint probability, which captures the combined influence of wind direction and speed on the presence of all these compounds. The polar graph is created based on the wind direction and wind speed measured in 50-m height. The OA contribution from all OA sources are utilized as color code in the polar graph to illustrate the potential regional source.

11. Line 422: Please clarify why O_x wasn't employed for this particular analysis.

Ox (O_3 + NO_2) was used prior to source apportionment analysis to provide a rough indicator of seasonal variations in atmospheric oxidation capacity, or say the overall intensity of secondary oxidation. After source apportionment, however, SOA sources are further resolved, and their formation pathways from daytime and nighttime chemistry can be distinguished. Therefore, when correlating daytime OOA with oxidants, O_3 alone is a more appropriate choice, as it avoids potential interference from the diurnal pattern of NO_x emissions.

Line 427: The preceding dot before (Liu et al., 2024) should be removed.

Done.

12. Line 437-441: Quantitative comparison of diurnal and seasonal variations in LO-OOA, MO-OOA, NO-OOA contributions would strengthen the analysis.

    To address this comment, we added additional sentences in the manuscript. Please find the quantitative comparison summarized here:

Draft Line 445-446: LO-OOA and MO-OOA are the only source factors showing distinct diurnal peaks around noon (UTC 11:00-14:00, Fig. S4–S7), a pattern characteristic of SOA formed through photochemical processes.

Draft Line 450-451: The NO-OOA factor resolved in this study is attributed to nocturnal OOA formation dominated by NO3 chemistry, with a diurnal peak around midnight (UTC 20:00–04:00, Fig. S4–S7).

Draft Line 464-468: Seasonal variations in the relative contributions of daytime (LO-OOA, MO-OOA) and nighttime (NO-OOA) oxidation pathways indicate that OOA formation is dominated by daytime oxidation of biogenic VOCs in spring and summer, accounting for 64–66% of OOA. In contrast, during autumn and winter, nocturnal oxidation (NO-OOA) exceeds daytime oxidation and dominates OOA formation, contributing 75–80%.

13. Line 504: Would this methodological bias occur across other seasons?

    The clear MSA-OA source was only resolved during spring and summer, which are carefully discussed here. From this study, we can only conclude that in summer there is a higher chance of overestimating the marine influence on OA if absolute MSA or MSA-OA concentrations are used as tracers.

14. Figure 10 suggests that both Mean Cluster 1 and Mean Cluster 2 exhibit marine influences, yet no marine source contribution is identified. Please clarify why marine sources are not reflected in the apportionment results? And additionally, what about the percentage distribution of air masses from different trajectory directions

    In Figure 10, we highlight the potential bias introduced when using absolute MSA or MSA-OA concentrations as tracers for marine transport. The dark-red shaded period represents a typical local-emission-dominated episode with strong formation of OS and sulfate. During this period, the absolute concentrations of MSA and MSA-OA (Fig. 10a) also appear elevated. This could indicate either unidentified terrestrial sources of aerosol MSA, as suggested by previous studies (Zhou et al., 2017; Young et al., 2016; Ge et al., 2012), or methodological deviations. To reduce this bias, we propose using the mass fraction of MSA or MSA-OA as tracers. As shown in Fig. 10b, this approach reveals the marine influence more clearly in cluster 1 (blue background) and cluster 2 (yellow background). The fraction of time periods assigned to each cluster during the AMS measurement period is also illustrated in the time series. Without considering the AMS measurement constraint, the corresponding fractions for clusters 1–3 are 48%, 14%, and 38%, respectively (Fig. 12).

15. Line 519-520: The fire spots and trajectory plots suggest influences from local European fires, yet the analysis primarily emphasizes Russian contributions. Could you clarify why other regional fire impacts (e.g., from Europe) were not addressed

    The hotspot plots in Fig. 11 show accumulated fire detections during spring. These hotspots are detected based on thermal anomalies in satellite imagery, and therefore single and isolated dots are often due to noise but not real fire. In contrast, clusters of persistent hotspots are much more reliable indicators of wildfire activity. As shown in Fig. 11, such clusters were primarily located in Russia during the campaign period.

16. Line 525: How does the mass spectral signature of BBOA at this time differ from other periods? There should be distinguishable features between relatively fresh and aged BBOA

Yes, you are right. As shown in the fCO_2 vs. $fC_2H_4O_2$ space below, the orange diamond markers represent transported wildfire BBOA. The mass fraction of the tracer $C_2H_4O_2$ is clearly lower than that observed in BBOA from other seasons. This decrease in $C_2H_4O_2$ is reported to be consistent with the aging time in chamber experiments (Kodros et al., 2020), as illustrated in the inset panel. This discussion was added in the draft and the figure was now included in the supplement.

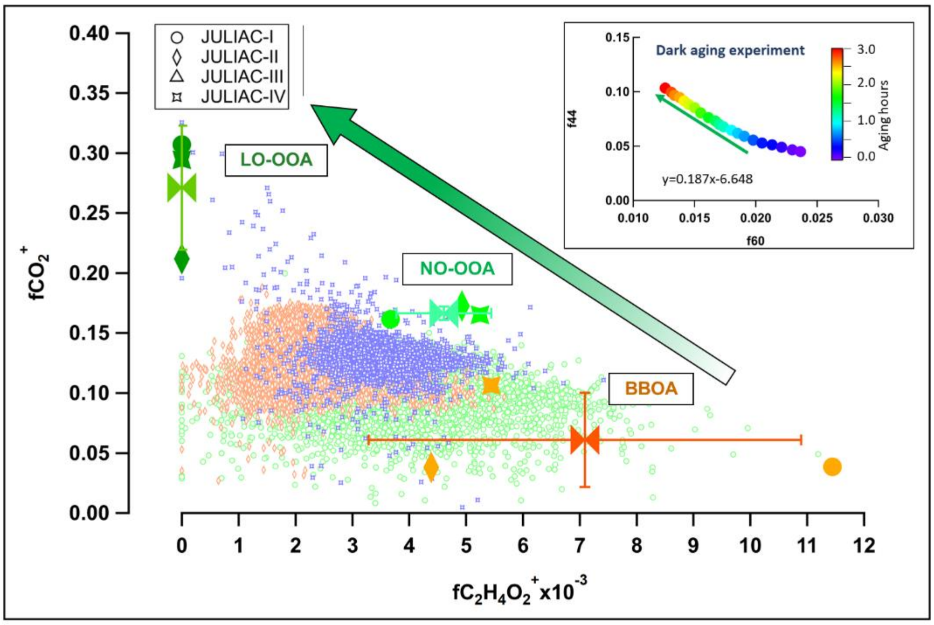


Figure S9 : fCO2+ (fraction of fragment CO2+ in OA) vs fC2H4O2+ (fraction of fragment C2H4O2+ in OA) for raw organic aerosols, and MO-OOA, NO-OOA, and BBOA factors' profile during the JULIAC-I, JULIAC-II, and JULIAC-IV are given. Means value and stand derivation error bar for these three types of source factors are also displaye. A small window at the upper right corner displays the evolution of HR-ToF-AMS measured OA through f60 vs f44 space for 3 hours' biomass burning dark aging laboratory experiments (Kodros et al., 2020).

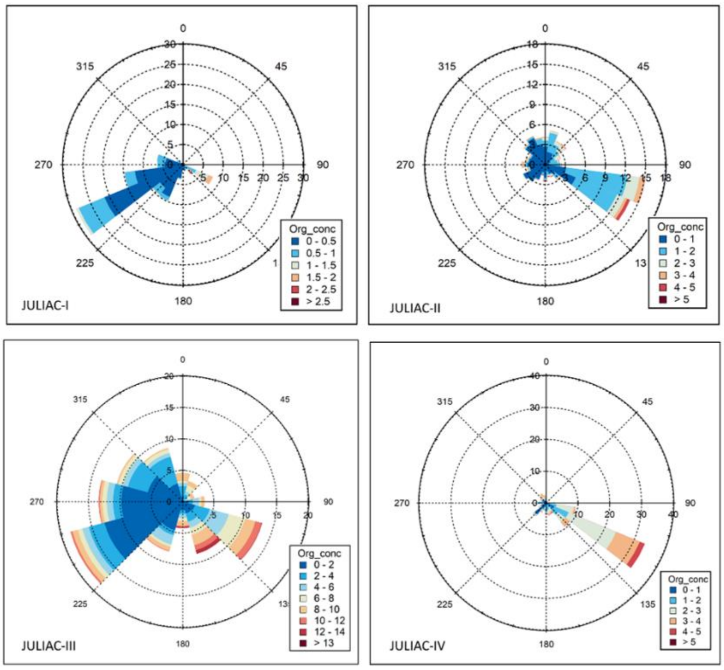
17. Figure 12: The air mass trajectories indicate marine influences across all seasons, yet MSA-OA contributions are only identified under marine air masses in spring/summer source apportionment results, not in autumn/winter. Additionally, please clarify whether PMF was run separately for each season or as a consolidated dataset in "PMF analysis" section in the main text.

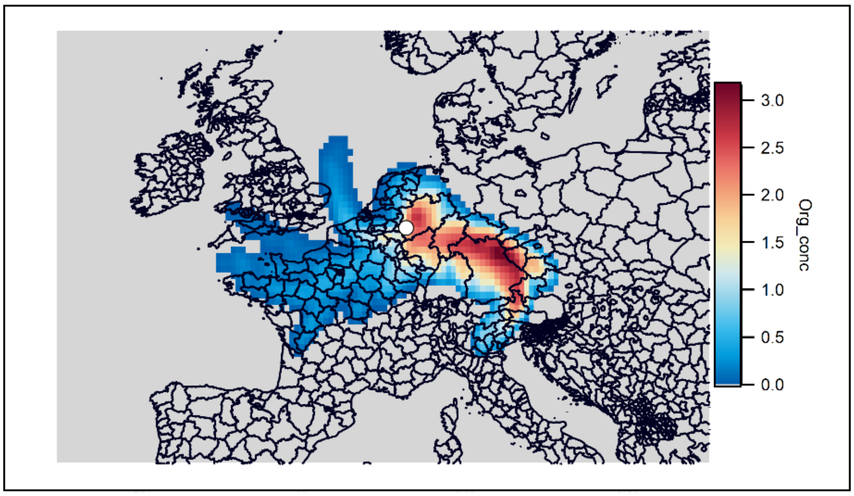
Yes, MSA-OA was observed only during the warm seasons, which is a key conclusion we highlight regarding seasonal variations in the effects of regional transport. This finding suggests that the seasonal activity of phytoplankton and anaerobic bacteria is an important factor modulating the marine influence. Corresponding modification in the PMF analysis section was made as below in the draft:

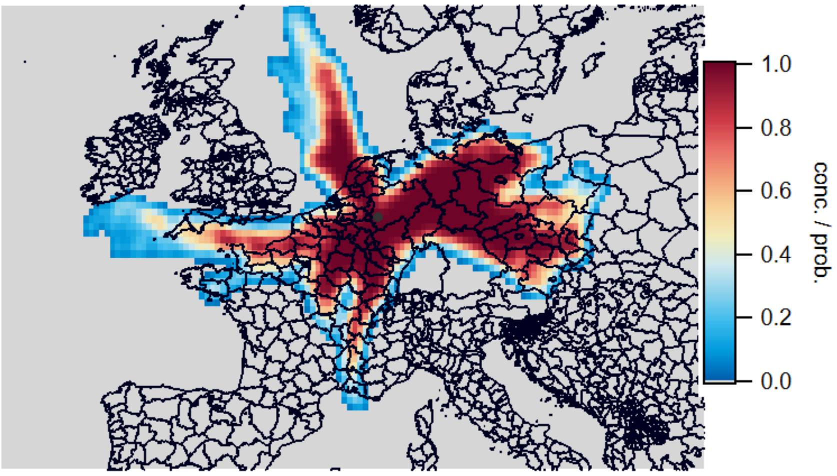
Draft Line 194-196: To investigate seasonal variations in the sources and contributions of organic aerosols, PMF analysis was performed separately for each season on organic aerosol concentrations (m/z 12–160) measured by HR-ToF-AMS during the JULIAC campaign.

18. Section 4.1.3 provides a relatively weak analysis of the regional transport factor. It is recommended to incorporate other statistical methods (such as Potential Source Contribution Function, PSCF) for supplementary validation.

    Thanks for this comment. Several statistical methods, such as wind rose analysis and concentration field (CF) analysis, PSCF…… were applied using the Zefir toolkit to identify potential regional transport sources. For detailed insights, please see the figures below. However, we ultimately found that trajectory clustering provides the most informative representation. Thus,we keep on using it in the main draft.







19. Line 603-608: The relationship between increased ALWC and organic sulfur formation during heatwaves requires more direct data support (e.g., SO_2 concentration, aqueous-phase reaction rates).

    Thank you for raising this point. The pity is that we do not have concurrent measurements of SO_2 or IEPOX for this ambient campaign, and ALWC was estimated with ISORROPIA using particle-phase inputs only. Consequently, we cannot unambiguously identify a dominant pathway for OS increase during a heatwave. We have revised the discussion to frame our interpretation as plausible rather than definitive and to note the key uncertainties. But to address this gap, we are planning targeted chamber experiments to repeat heatwave-like conditions, like the changes in BVOC emissions, aerosol acidity, ALWC…. to understand the gas-phase/heterogeneous chemistry (including IEPOX uptake) more straightforward.

20. The terms “organosulfur” and “organosulfates” are used interchangeably in the text and should be standardized (note that “organosulfur” should be a broader term encompassing organic sulfur compounds and organosulfates).

    Thank you for this comment. We agree with the reviewer that organosulfur is a broader term, while organosulfates represent a specific subset. Since our AMS analysis based on sulfate fragmentation patterns specifically targets organosulfates, we have revised the manuscript to consistently use the term “organosulfates”, which is also the more common terminology in the aerosol research community.

21. In lines 283 and 323, "ALWC" can be used directly since it has already been defined earlier.

    Done.

RC2

This manuscript provides an analysis of the seasonal variability, sources, and formation mechanisms of submicron aerosols in the Jülich region of Germany, based on measurements from the JULIAC campaign. Particular attention is given to secondary organic aerosol formation and its influence from characteristic events during the measurement period, such as regional transport episodes and heatwave conditions. The paper is generally well written and could be considered for publication once the following concerns are addressed.

Specific comments:

1. Line 41. The direct and indirect effects are not described. Please add a brief description of radiation and cloud interactions.

   Thanks for this valuable comment. As suggested, we added the following sentence in the manuscript

   Draft Line 43-44: Hence, their climate impact varies through direct scattering and absorption of radiation and indirect modification of cloud properties as cloud condensation nuclei

2. Line 45. The statement "playing a key role in altering the environmental impact of aerosols". Please specify how these impacts are altered.

   Thanks for this suggestion. The modification in draft is attached as below

   Draft Line 45-46: Therefore, understanding aerosol emissions and secondary formation is essential, as these processes determine aerosol properties and how they influence radiation and clouds, altering climate impacts.

3. Line 71. Correct the reference "Liu et al. (Liu et al., 2024)"

   Thanks. We corrected the reference.

4. Section 2.2. "Instrumentation": The manuscript does not specify the instrument models used and does not describe the calibration protocols, for example, those applied to the PTR-ToF-MS during the field campaign. Please include them.

   The instruments performance, data quality, and data processing procedures for AMS have been thoroughly documented in previous JULIAC campaign studies. To avoid repetition, we have added clearer citations and brief introduction as shown below. We thank the reviewer for pointing out the missing details on PTR calibration protocols, which has been specified in section 2.2 as shown below.

   Comprehensive descriptions of the maintenance and data processing of HR-ToF-AMS in this study are provided in prior studies (Liu et al., 2022, 2024). In brief, ionization calibrations were conducted biweekly throughout the campaign using an ammonium nitrate standard following Drewnick et al. (2005). The relative ionization efficiency (RIE) of species was also determined every two weeks using ammonium sulfate, following Jimenez et al. (2003), and the RIE values applied for each campaign phase are summarized in Table S*. The AMS collection efficiency (CE) was constrained by comparison with concurrent SMPS measurements, assuming an average aerosol density of 1.4 g cm^{-3} (Cross et al., 2007). A constant CE of 0.96 was applied for JULIAC-I and II, whereas a composition-dependent CE (Middlebrook et al., 2012) with a value of 0.57 was used for JULIAC-III and IV. One flow rate correction was performed once during JULIAC-II by comparing the logged AMS flow (derived from lens pressure) with measurements from a flow rate Gilibrator.

   For the PTR-ToF-MS, the drift tube was operated at an E/N of 126 Td during JULIAC I and II and adjusted to 120 Td during JULIAC III and IV. The PTR was calibrated on a weekly basis whenever possible. For calibration, the instrument was connected to our Liquid Calibration Unit (LCU), and a multi-step calibration was performed using a gas standard at ~60% relative humidity. We used four concentration steps and one zero step (five steps in total), with the standard diluted in the range of 1–15 ppb. The list of VOCs included in the calibration standard, along with their mixing ratios and uncertainties, is provided in Table S2. Further details on data quality and instrument performance during the JULIAC campaign are provided in Cho et al. (2023).

5. Section 2.5 “Trajectory model”: Specify the altitude at which the back trajectories were calculated.

   The back trajectories were calculated with the trajectory endpoints set at 50 m above ground level. No specific trajectory starting altitude was prescribed, so the paths represent near-surface air mass transport. The sentence below has been added into the main draft.

   Draft Line 214-217: The trajectory arrival times match the detection times of instruments at the JULIAC campaign, with the trajectory endpoints set at 50 m above ground level. This altitude corresponds with the height of the sampling tube inlet of ambient air to the SAPHIR chamber.

6. Line 245: Please ensure consistent formatting of “O_x”.

   done

7. Line 261: Space between “America” and “(Kiendler-Scharr”

   done

8. Line 263-266: The authors report high organic-nitrate fractions during summer (62%), yet the cited comparative studies are from spring, and the authors’ springtime results are considerably lower (14%). Please provide comparable references from summer or justify why springtime observations are directly comparable; otherwise, consider rephrasing the comparison.

   This is not the first time that a high mass fraction of organic nitrate has been observed in the European area. For instance, the fraction of organic nitrate to total aerosol nitrate was found to be 67% at Cabauw, Netherlands in March 2008, 73% at Melpitz, Germany in March 2009, 63% at Harwell, UK in October 2008, and 67% at Vavihill, Sweden in October 2008 (Kiendler-Scharr et al., 2016).

9. Line 274: The reported organosulfur concentrations (summer up to 6 µg/m^3; autumn up to 2 µg/m^3) do not match the values shown in Figure 3. Please verify and correct the text or the figure to ensure consistency.

   Thanks for pointing out this mistake.

   The concentration of organosulfate is the highest in summer (JULIAC-III, up to 2.5 µg/m3) (Figure 3), followed by autumn (JULIAC-IV, up to 0.85 µg/m3).

10. Line 273: Space between “heating” and “(Liu et al., 2024)”.

    Done

11. Lines 304, 313, and 315: The citation of Schueneman et al. (2021) should be corrected.

    done

12. Line 388: Space between “of” and “R^2”.

    Done

13. Line 399 – 409: The attribution of BBOA emissions to the sugar factory is unclear. Is there a specific reason why the factory operates only in autumn? It would be useful to also assess other potential influences, such as meteorological parameters (e.g., wind direction and speed).

    The attribution of BBOA emissions to the sugar factory in Jülich is consistent with its seasonal “Rübenkampagne” (sugar beet campaign), when the factory operates around the clock from autumn to early winter (typically October–December) to process the beet harvest. This short but intensive production period leads to strong, localized emissions, while outside the campaign the factory remains largely idle—minimizing odors and addressing complaints from nearby residents.

    And yes, it’s truly helpful to combine with a meteorological analysis, as added in supplement Fig. S2. The relevant discussion is now modified and given as below:

    The most abundant OA contribution from biomass-burning is found in autumn, where two distinct BBOA sources are identified: one dominated by local emissions and the other associated with regional transport (Fig. S2). The local BBOA source is likely related to residual heating and nearby emissions from the sugar factory, which have active production of sugar only during the autumn phase.

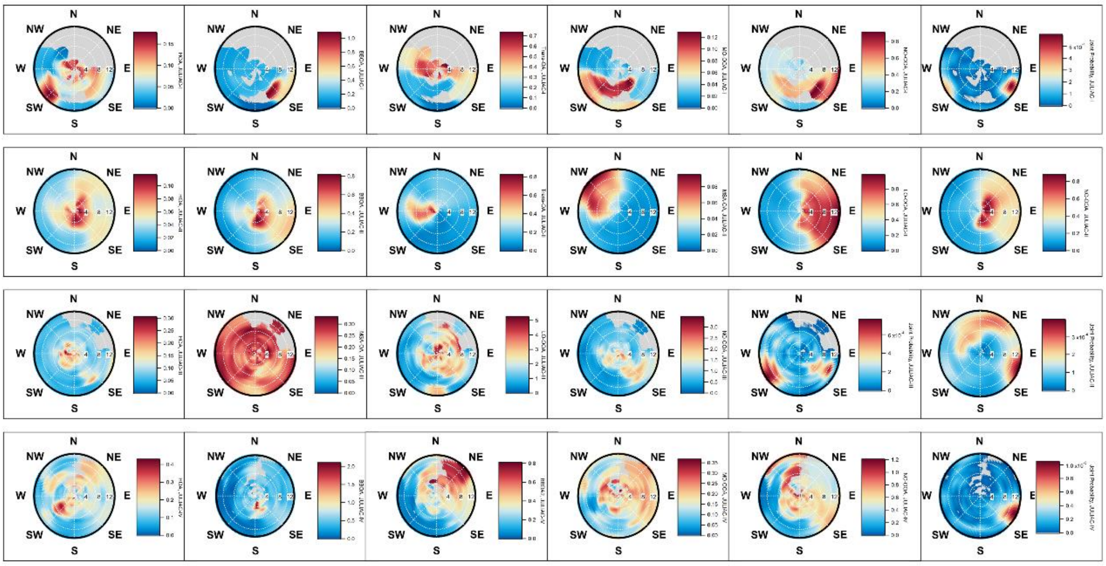


Figure S2: Polar diagrams using non-parametric wind regressions are shown for all JULIAC-intensive phases, illustrating source factors contribution as well as the joint probability, which captures the combined influence of wind direction and speed on the presence of all these compounds. The polar graph is created based on the wind direction and wind speed measured in 50-m height. The OA contribution from all OA sources are utilized as color code in the polar graph to illustrate the potential regional source.

14. Please ensure consistent use of the term “VOC” or “VOCs” throughout the manuscript.

    Done

15. Line 515-528: Please consider whether wildfire plumes from other European countries (e.g., Poland, where numerous hotspots were detected) may have contributed to the aerosol population.

    A fun fact is that many hotspots that appear to be in Poland are actually in Kaliningrad, a province of Russia. The wildfire events clearly located in the Russian region are influenced by Russian policy. In 2019, Russia's wildfire control policy was primarily based on the 2006 Forest Code, which allowed authorities to ignore remote fires in designated "control zones" if firefighting was deemed not economically viable. More detailed discussion are available in: https://papers.ssrn.com/sol3/papers.cfm?abstract_id=4007978

    These hotspots are detected based on thermal anomalies in satellite imagery, and therefore, single and isolated dots we saw in the European area are often due to noise, but not real fire(Schroeder et al., 2014).

16. Is the O:C ratio different between autumn BBOA sources (residential heating, sugar factory) and aged BBOA from regional wildfire plumes (e.g., Russia region)? This comparison could reveal important differences in oxidation state between local and transported BBOA in spring and autumn.

    Unfortunately, this study did not reveal a clear difference in O:C between local and regionally transported biomass burning plumes. However, a distinct decrease in the mass fraction of $C_2H_4O_2$ was observed in the regional wildfire plume compared to other BBOA. The related discussion is provided below.

    Draft Line 551-556, In addition, the mass fraction of the biomass burning tracer, $fC_2H_4O_2^+$, in BBOA from spring is clearly lower than that observed in other seasons (Fig. S9). This observation is consistent with chamber experiments of biomass burning dark aging, where $fC_2H_4O_2^+$ decreases with increasing aging time (Kodros et al., 2020).

17. Figures S3-S6: The x-axis labels in the diurnal variability plot appear overcrowded. please adjust the spacing or format to improve them.

    Done

18. Table S6: “Sulfate” is repeated in the caption, and the volume/surface columns are somewhat confusing. Please clarify

    Thanks for pointing that out. This table shows the aerosol volume concentration measured by SMPS… The unclear header has been improved in the table.

RC3

This work describes the aerosol composition during one-year measurements at a semi-rural area in Germany. The results provide interesting insights of the contribution of secondary organic aerosol sources to the organic aerosol and highlighted the presence of organic nitrate species. However, some modifications and technical corrections should be considered before publishing this work. Additionally, I consider the number of figures in the main text should be reduced, however I let it to the editor consideration.

1. Section 2.2 introduce the different instrumentation deployed and later the agreement time between the trajectory arrival and the detection time of the instruments is highlighted. However, details of data acquisition resolution and other technical details for the mass spectrometry techniques are not provided (e.g., collection efficiency for organics in AMS, conditions in the drift tube and E/N for PTRMS). I suggest the authors add a table in the supplement with the technical information.

   We agree with the reviewer that additional technical details of the deployed mass spectrometers improve clarity and reproducibility. The instruments performance, data quality, and data processing procedures for AMS have been thoroughly documented in previous JULIAC campaign studies. To avoid repetition, we have added clearer citations and brief introduction as shown below.

   Draft Line 121-152: Comprehensive descriptions of the maintenance and data processing of HR-ToF-AMS in this study are provided in prior studies (Liu et al., 2022, 2024). In brief, ionization calibrations were conducted biweekly throughout the campaign using an ammonium nitrate standard following Drewnick et al. (2005). The relative ionization efficiency (RIE) of species was also determined every two weeks using ammonium sulfate, following Jimenez et al. (2003), and the RIE values applied for each campaign phase are summarized in Table S*. The AMS collection efficiency (CE) was constrained by comparison with concurrent SMPS measurements, assuming an average aerosol density of 1.4 g cm^{-3} (Cross et al., 2007). A constant CE of 0.96 was applied for JULIAC-I and II, whereas a composition-dependent CE (Middlebrook et al., 2012) with a value of 0.57 was used for JULIAC-III and IV. One flow rate correction was performed once during JULIAC-II by comparing the logged AMS flow (derived from lens pressure) with measurements from a flow rate Gilibrator.

   For the PTR-ToF-MS, the drift tube was operated at an E/N of 126 Td during JULIAC I and II and adjusted to 120 Td during JULIAC III and IV. The PTR was calibrated on a weekly basis whenever possible. For calibration, the instrument was connected to our Liquid Calibration Unit (LCU), and a multi-step calibration was performed using a gas standard at ~60% relative humidity. We used four concentration steps and one zero step (five steps in total), with the standard diluted in the range of 1–15 ppb. The list of VOCs included in the calibration standard, along with their mixing ratios and uncertainties, is provided in Table S2.

   Further details on data quality and instrument performance during the JULIAC campaign are provided in Cho et al. (2023).

2. In lines 248 to 250, the correlations of higher O/C ratios with increases Ox concentrations are highlighted only during summer. However, in Figure 2 a similar observation is reported during the spring period. This observation requires further discussion.

   The previous phrasing ‘only during summer’ was indeed ambiguous. We have revised the corresponding discussion to clearly emphasize the summer period, thereby highlighting that this performance is consistent with strong SOA formation.

   Line 251: During summer, the strongest correlation between the O:C ratios of OA and the O_x concentrations is observed, suggesting the significant contribution of secondary OA formation to total OA. This assumption is further discussed and supported by the results of the source apportionment analysis presented in Section 4.1.

3. In Lines 267 and 268 the role of NO3 chemistry is highlighted to contribute to OA production during all the seasons, specially during the night. What is the role of other parameters measured (e.g., temperature and RH) into this observation as seasonal variations are also reported in Figure 3.

   Thanks for this excellent question. We addressed this point in detail in our previous NO-OOA study (https://doi.org/10.1038/s41612-024-00747-6). In that work, we discussed not only the volatility shifts induced by temperature changes, but also the potential role of heterogeneous and aqueous-phase chemistry at higher RH conditions, as well as the influence of boundary layer development. Our conclusion was that all these factors slightly affect, but none of them can be considered the driving factor behind the observed enhancement of nighttime SOA.

4. In lines 301 to 303, the authors highlight that organosulfur calculations can be affected by environmental parameters such as acidity. During the one-year calculations, the fragmentation method seems to be feasibly. I wonder about the applicability of this methodology during the heatwave period, where environmental conditions and chemistry is enhanced. Could the authors provide information on this? Could that have an impact into the organic and inorganic nitrate?

   Thank you for this comment. Schueneman et al. (2021) showed that the fragmentation pattern of ammonium sulfate varies with aerosol acidity and NH_4NO_3 fraction, which can lead to underestimation of OS when conditions deviate from the recommended “condition II.” In our study, aerosol acidity was only approximated from ion balance (without gas-phase measurements). Since aerosol pH is highly sensitive to temperature and humidity, we would expect acidity to increase during heatwaves (Pratap et al., 2025). However, this change in aerosol pH was not evident from our ion-balance estimates based solely on particle-phase data. Considering the general tendency for aerosol acidity to increase with temperature (Pratap et al., 2025; Lv et al., 2024), we acknowledge that OS formation during heatwaves may still be underestimated in our analysis. Nevertheless, the deviation during the heatwave remains uncertain due to measurement limitations and methodological uncertainties.

   For nitrate, as discussed in draft, the clear decrease of nitrate mass fraction during summer reflects its high volatile nature. That effect also exists during a heatwave event and might to some degree enlarge the fraction of organic nitrate to inorganic nitrate. Heatwaves are also accompanied by stronger radiation, hence clearly increasing the atmospheric oxidation capability, like the increase in the concentration of OH, O3, and NO3, as illustrated in figure 13. Higher concentrations of NO3 speed up the formation of organo nitrate.

Technical details:

5. Line 19: Define the acronym JULIAC.

   Done

6. Lines 73 and 74 should be merged with the main goals introduced in lines 94 to 97.

   In this study, comprehensive measurements and analyses are conducted to investigate the seasonal variations in aerosol composition, with a particular focus on organic aerosol contributions from local primary emissions, secondary formation, and long-range regional transport. Building upon the previous findings by(Liu et al., 2024), this study provides a more detailed analysis of the seasonal variations of nocturnal OOA formation from the perspective of its precursors. Special attention is given to the summer period, when a significant heatwave event occurred.

7. Lines 101-102: “aimed to explore annual variations of and source contributions to oxidant concentrations”. This sentence is not clear.

   Thanks for this notification. We rewrote the sentence to:

   aimed to understand annual variations of atmospheric oxidant concentrations (Cho et al., 2022), the composition of gas-phase species (Tan et al., 2022), and submicron aerosols (Liu et al., 2024)

8. Line 111: delete the and before submicron aerosol.

   done

9. Line 143: replace made by reported.

   done

10. Line 175: replace aerosol organics concentrations by organic aerosol concentrations.

    done

11. In line 180, the use of prior factors from literature were used to constrain PMF analysis was mentioned. Can the authors provide further details of the additional constrains considered.

    Done. We added Table S3-4 here, which contains the prior factor citation information.

12. Line 193: What is the resolution of the GDAS used for the HYSPLIT model.

    We used GDAS with 1° horizontal resolution. The line was adopted accordingly.

13. Line 202: The acronyms for RH and T are defined here but used since before. Introduce the abbreviations before and homogenize the text accordingly.

    done

14. Modify the colors in right panel of Figure 6, as differences between the seasons are not clear.

    Done

15. Line 421: replace secondary organic aerosol by SOA

    Done

16. Line 525: replace primary organic aerosol and secondary organic aerosol by POA and SOA.

    Done

17. 526: delete organic aerosol, the acronym was defined before.

    done

18. Lines 569: “In this study, a decrease in monoterpene concentrations is observed during a heatwave event, with the average levels dropping to 60% of the pre-heatwave values (Figure 13).” The decrease of monoterpene concentrations is not clear in Figure 13 as values are lower than 0.3 ppb. The authors should consider displaying this profile on the right axes.

    Done

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