General Comments:

This manuscript (MS-ID: egusphere-2024-3015) presents a comprehensive study on the molecular characterization of organic aerosols in urban and forested areas of the Paris region using high-resolution mass spectrometry (HRMS). The authors provide valuable insights into the chemical composition, sources, and potential interactions between anthropogenic and biogenic emissions that influence the organic aerosol characteristics in this mixed environment. However, the current version of manuscript should be strengthened by providing more detailed explanations and discussions on the observed patterns, the potential drivers of the urban-rural differences, and the implications of the findings for regional air quality and atmospheric chemistry. Additionally, the authors need to consider addressing the specific comments and technical suggestions provided to improve the overall clarity, coherence, and presentation of the manuscript. Overall, this manuscript can be published after addressing these major concerns and issue listed below. But, I would let editor to decide.

Major Concerns:

1. On page 3, lines 65-70, the authors claim that the Paris area is surrounded by low urbanized areas mostly composed of intensive agriculture fields and forest. But it is not clear how this spatial distribution of urban and rural areas may influence the mixing of anthropogenic and biogenic emissions. The authors should provide more details on the potential transport and interactions between these different air masses. (Page 3, lines 65-70)

2. Line 150, the authors applied methanol to extract filter sample, what about other solvents like water or CH_3CN. How can authors ensure that methanol can extract all major OAs in the filter samples. Additional experimental evidence needs to be supported.

3. What's the extraction efficiency of methanol?

4. Line 168, The authors state that CHN and CHS families were not considered here due to limitation of test mode. But I would like to know the abundance of these compounds and how they compete with CHO, CHON, CHOS, CHONS, and CHNS?

5. Line 180, what's the uncertainties or biases for the assumption of using carboxylic acids (R-COOH) instead of other likely compounds? This would eventually have the impact on the classification of aromatics types.

6. The authors mention the detection of C5H12SO7 and C10H17NSO7 compounds, which they attribute to isoprene and α-pinene oxidation products, respectively. The authors should provide more details on the potential formation mechanisms and the role of anthropogenic pollutants (NOx and SO2) in the formation of these organosulfate compounds.

Otherwise, it would be great challenge to allow readers to follow the interpretation. (Page 11, lines 277-283)

7. In section 3.2.1, the authors discuss the similarities and differences in the Van Krevelen diagrams between the urban and forested sites. While they provide some interpretations of the different compound domains, more detailed explanations are needed on the potential sources and formation processes associated with the observed patterns. (Page 14, section 3.2.1)

8. In section 3.1, the authors discuss the temporal variability of OC and EC concentrations at the two sites. However, they do not provide a clear explanation for the observed similarities and differences between the urban and forested sites. More discussion is needed on the potential drivers of these patterns, such as the influence of meteorology, air mass transport, and local versus regional sources. (Page 7, section 3.1)

9. The authors mention the detection of $C_5H_{12}SO_7$ and $C_{10}H_{17}NSO_7$ compounds, which they attribute to isoprene and α-pinene oxidation products, respectively. More details on the potential formation mechanisms and the role of anthropogenic pollutants (NO_x and SO_2) in the formation of these organosulfate compounds are required. (Page 10, lines 277-283)

10. The authors perform a correlation analysis to investigate the influence of meteorological parameters and anthropogenic pollutants on the chemical composition of the organic aerosols. The discussion of these results could be strengthened by providing more mechanistic explanations for the observed correlations, particularly for the differences between the urban and forested sites. (Page 16, section 3.3)

11. The authors conclude that the forested area of Rambouillet is affected by anthropogenic inputs, influencing the atmospheric chemical composition. A clear quantification or assessment of the relative contributions of anthropogenic versus biogenic sources to the organic aerosol composition at the two sites would help to better understand the extent of the urban influence on the forested area. (Page 19, lines 464-467)

12. The authors state that the HRMS analysis showed similar patterns of the contributions of anthropogenic and biogenic emissions on both sites for periods of pollution. However, a clear definition or criteria for what constitutes a "period of pollution" is missing. More information is needed on how these periods were identified and how they differ from the background conditions. (Page 19, lines 469-471)

13. The similarities in the chemical composition between the urban and forested sites during the pollution periods have been discussed and the authors are encouraged to supply the potential implications of these findings for the regional air quality and the impact of urban emissions on the surrounding environment. (Page 19, lines 469-471)

14. The authors state that the detection of compounds associated with biogenic and anthropogenic oxidation products highlights the importance of understanding urban and rural chemistries at the molecular level. However,

they do not provide a clear discussion on how this molecular-level information can be used to improve our understanding of the complex interactions between different emission sources and their impact on the regional atmospheric composition. (Page 19, lines 449-457)
15. The authors mention the use of the ACROSS campaign data, but they do not provide any details on the specific objectives, experimental design, or other relevant information about this campaign. (Page 4, lines 110-115)
16. A big picture or schematic is strongly recommended to summarize the finding in this work.

Technique Issue:

1. In Figure 2, the y-axis labels for the mass spectra are not clearly legible. Please consider increasing the font size or adjusting the layout to improve the readability of the figure.
2. Figure S3 are not clearly legible and unreadable.