The manuscript by Yang and co-workers investigates the role of organic carbon complexed with metals as a potentially important pathway for sulfate transformation. The study employs a broad range of analytical and computational techniques to characterize the investigated systems. Overall, the conclusions are generally supported by the presented data, and I therefore believe that publication may be warranted after revision.
My main concern is that several of the analytical techniques are not sufficiently discussed in terms of their limitations and interpretation. As a result, some conclusions appear to rely more on correlations than on direct evidence of causation. For example, changes in absorption spectra are attributed to the formation or absence of metal–ligand complexes, yet alternative explanations, such as contributions from contaminant metals or other matrix effects, do not appear to have been adequately evaluated.
I believe the manuscript would benefit from a more detailed discussion of the analytical capabilities, limitations, and quality assurance aspects of each technique used. In addition, the following points should be addressed:

1. Can the authors provide more context regarding the distinction between "weak" and "strong" complexation? Ideally, this should be related to stability constants (log K values) and placed into a broader chemical complexation context.
2. It is not entirely clear what is meant by the terms "OC@Me" and "OC + Me". These terms are used frequently throughout the manuscript, but the distinction between them is not adequately explained.
3. The samples appear to be prepared under relatively simple laboratory conditions. How representative are these systems of real atmospheric environments? Under which atmospheric conditions and in which environments would such complexes realistically form?
4. The ROS experiments and the subsequent analyses appear to have been conducted on different subsamples. Could this introduce variability that affects the interpretation of the results? Please discuss the implications for data consistency and reproducibility.
5. The DFT calculations are interesting and potentially valuable. However, it is not entirely clear whether the computational models adequately represent the real systems studied experimentally. Consequently, the reported correlations may not necessarily support the processes occurring in atmospheric environments. Furthermore, the methodological description of the DFT calculations is too brief to properly assess the quality and reliability of the computational work.
6. A more comprehensive discussion of quality assurance and quality control (QA/QC) procedures is needed for most, if not all, analytical techniques. For example, what were the detection limits of the ion chromatography (IC) measurements, and how close were these limits to the lowest concentrations reported in the study?
7. I am concerned that insufficient attention has been paid to potential artifacts arising from sample preparation, storage, and analytical procedures. For example:

- For the Cu measurements obtained by XPS, how can the authors exclude changes induced during sample storage?
- Were possible beam-induced effects assessed during XPS analysis?
- The UV–Vis measurements were performed offline. Why were in situ measurements not considered, for example using a fiber-optic probe? Please discuss how sample handling may have affected the observed results and provide evidence that the reported trends are not analytical artifacts.

8. The Conclusions and Atmospheric Implications sections appear somewhat simplistic given the complexity of the studied system. The associated uncertainties and limitations should be acknowledged more explicitly. Inclusion of uncertainty estimates, error propagation, and sensitivity analyses would substantially strengthen the atmospheric relevance of the findings and provide a more realistic assessment of the robustness of the conclusions.