

Reviewer 1

This is a well-written and comprehensive study investigating the complexation of glutamic acid (Glu) with Fe(II)/Fe(III) and its implications for atmospheric chemistry, particularly in cloud water. The experiments are well-designed, and the results provide valuable insights into the role of amino acids in modifying Fenton and photo-Fenton reactions. The findings are novel and contribute significantly to our understanding of atmospheric oxidative processes.

- The atmospheric relevance could be further emphasized by discussing how variations in cloud pH, light intensity, or iron/ligand ratios might influence the observed processes.

Response

We appreciate the reviewer's insightful suggestion to strengthen the discussion of the atmospheric relevance of our findings. In the revised manuscript, we have added a paragraph discussing how typical variations in cloud water pH (usually ranging from 3 to 7), diurnal changes in solar radiation, and fluctuating iron-to-ligand ratios can modulate the reaction kinetics and pathways of the studied system. This addition highlights the broader implications of our results for cloud and aerosol chemistry under realistic atmospheric conditions. The new discussion can be found on **page 26-27, lines 510-518 and lines 522-526**.

- It would be better to explicitly state in the first paragraph of 'Introduction' that the role of amino acids in modifying iron redox chemistry and OH production remains poorly understood.

Response

We have revised the introduction, this sentence has been added in the first paragraph. The modification can be found at **lines 50-54**.

- Were the pH conditions (3.8–5.6) chosen to represent specific atmospheric scenarios (e.g., polluted vs. remote clouds)? A brief justification would be useful.

Response

The pH values of 3.8 and 5.6 were chosen based on both experimental needs and atmospheric relevance. pH 5.6 was used in kinetic studies to promote Fe(II)-Glu complex formation, while pH 3.8 was used in ESR experiments where •OH detection is more effective under acidic conditions. These values fall within the typical pH range of cloud water and represent different environments—pH 3.8 for polluted regions and pH 5.6 for remote areas. This allows us to study the system under realistic atmospheric conditions. A clarification has been added in the revised manuscript at **lines 133–139**.

- For the photolysis experiments, was the light spectrum adjusted to match real solar conditions?

Response

The light spectrum was not specifically adjusted, but the spectral distribution is similar to natural solar radiation, which is important for the photoreactions of Fe(III) and Fe(III)-Glu, As shown in Fig. SM2 and our previous publication. The modification is provided at **lines 169-173**.

- The reported rate constant ($1.54 \times 10^4 \text{ M}^{-1} \text{ s}^{-1}$) is a key finding. However, how does this compare with other Fe(II)-organic complexes (e.g., oxalate, citrate)? A brief discussion would be useful.

Response

We did the comparison with our recently published value of Fe(II)-Oxalate, a brief discussion was added as well. The modifications were provided at **lines 271-276**.

- The detection of formate/acetate as primary LMCT products is interesting. Could these compounds further complex Fe(III) and influence subsequent reactions?

Response

Yes, you are right. As we wrote in the part of atmospheric implications at **lines 537-539**, those generated compounds can further complex with iron and participate in the consequent photoreactions.

- The discussion of Fe-Glu fractions in cloud water is insightful, but how might these vary in highly polluted vs. marine environments?

Response

The Fe-Glu fraction in cloud water is expected to vary depending on the environmental context. In highly polluted environments, elevated levels of iron and amino acid (such as Glu from biomass burning or urban sources) may promote the formation of Fe-Glu complexes. In contrast, marine clouds often contain lower concentrations of both Fe and amino acid (like Glu is around 33 pM collected in Venice on the Sacca San Biagio Island), leading to a smaller Fe-Glu fraction. The more discussion was provided in atmospheric implications part at **lines 510-518**.

- How might these findings affect our understanding of SOA formation?

Response

Thank you for the insightful question. Our findings suggest that Fe(III)-Glu complexes can influence aqueous-phase SOA formation in two key ways. First, the photooxidation of glutamic acid generates small organic acids (e.g., formate, acetate) and ammonium, which are known precursors of SOA in cloud water. Second, the presence of Fe-Glu alters the generation of •OH radicals and introduces additional LMCT-driven pathways, potentially modifying the oxidation chemistry of water-soluble organic compounds. These results highlight a previously underappreciated mechanism by which metal–amino acid complexes may affect the oxidative capacity of cloud water and contribute to SOA formation, particularly under polluted or organic-rich atmospheric conditions. The more discussion was provided in atmospheric implications part at **lines 528-535**.