Comments on “Boosting aerosol surface effects: Strongly Enhanced Cooperative Surface Propensity of Atmospherically Relevant Organic Molecular Ions in Aqueous Solution” by Harmanjot Kaur et al.

The paper investigates the surface propensity of organic surfactant molecular ions in aqueous solutions, which is crucial for understanding atmospheric aerosol behaviour. It focuses on carboxylic acids and alkyl amines, common organics surfactants in aerosols, and measures their surface enrichment using liquid-jet photoelectron spectroscopy. The study reveals an exponential increase in surface propensity with chain length and a significant boost in surface enrichment due to cooperative ion-ion interactions in mixed solutions. Results show surface compositions can vastly differ from bulk, impacting atmospheric processes like droplet formation and chemical aging. This research enhances the understanding of aerosol surface phenomena needed for climate model. The topic fits scope of ACP. The paper can be considered for publication after revisions that address the following concerns.

Major comments:

(1) As the authors described that the experiments were all performed under 10 ℃ (solution), 2×10^{-4} mbar (environment), So how do the authors think that the experiments could provide valuable information for atmospheric implication. I suggest the author give more explanations on temperature setting and improved the atmospheric implication part.

(2) As reported in the literature (for example, Figure 1 in Noziere, Baduel, and Jaffrezo (2014), surfactant delayed surface tension equilibrium, indicating that bulk-to-surface partitioning could undergo an equilibrium process in several seconds or minutes. I did not find description the duration time of signal collection process (but I guess it may in a very short time). So how do the authors consider the equilibrium process in bringing uncertainty to the current experimental results?

(3) The authors only selected 1:1 molar ratio for carboxylic acids (C1-C6, vertical) and alkyl amines (A1-A6, horizontal) mixed solutions in this manuscript. Can the authors explain why chose 1:1 molar ratio for experiments as a represent? since I personally think only one ratio of the mixtures may not be sufficient. And I am not sure that when the molar ratio alters (e.g., 1:3 (i.e., carboxylic acids dominated) and 3:1 (i.e., alkyl amines dominated), the factor results may change (Line 51-52: …a factor of several hundred).

Minor comments:

(4) Abstract: I personally think that the abstract is too long; please condense it to clearly state the main conclusions and their atmospheric significance.

(5) Tables: The format of most tables (1, 3, 4, 5, 6, 7) should be adjusted since they are not fit the specific guidelines.

(6) Line 51-52: Please add specific molar ratio, since the authors only have 1:1 molar ratio mixtures' result and it is unclear for other ratios.

(7) Line 64: "(Mccormick and Ludwig, 1967)" is not appropriated based on your reference part. It should be "McCormick".

(8) Figure 1 and Figure 4: If there are multiple subfigures, they need to be numbered (e.g., A, B, …). Do not used top, middle, and bottom.

(9) Figure 1: How to explain that all peak positions for single organic (p1 and p2 in the first two subfigures) slightly moved to the right in the third subfigure (spectrum signals for mixed solutions). In addition, why peak 2 in subfigure (A) and subfigure (B) were different as they both represent the C atom in CH_3 group.

(10) Table 1: The number of decimal places retained should be consistent (at least for same row). In addition, the values in the 4th row (carbon-normalized I_{norm}= I_{norm} /(x,y)) seems to be wrong, for example, for C4 column, 11.5/3=3.83, your value is 2.87), please check the whole calculation and carefully revised the corresponding Figures and Tables.

(11) Line 430-431: The author should give some explanations on what caused the different molecular orientations for alkyl ammonium and carboxylate anions.

(12) Line 516-517: Please add references.

(13) References section: The format of the references is not consistent (e.g., some journal names are full but others are abbreviations)**.** Please carefully check the format throughout the whole references based on the ACP guideline or other published paper.

Noziere, B., Baduel, C., & Jaffrezo, J. L. (2014). The dynamic surface tension of atmospheric aerosol surfactants reveals new aspects of cloud activation. *Nature Communications, 5*(1), 3335. https://doi.org/10.1038/ncomms4335