Responses to Referee #2's comments

We are grateful to the reviewer for your valuable and helpful comments on our manuscript "Reaction of SO_3 with H_2SO_4 and Its Implication for Aerosol Particle Formation in the Gas Phase and at the Air-Water Interface" (MS No.: **egusphere-2023-2009**). We have revised the manuscript carefully according to reviewer's comments. The point-to-point responses to the Referee #2's comments are summarized below:

Referee Comments:

The authors have addressed most of my comments appropriately, though some issues stil need some (quite easily implemented) revisions. Going through the issues using the numbering in my original review:

Response: We would like to thank the reviewer for the positive and valuable comments, and we have revised our manuscript accordingly.

Major issues

Comment 1.

Irrelevance of surface processes to NPF/nucleation: here unfortunately the terminology (and associated implications) is STILL in places incorrect: processes occurring on already-existing surfaces by definition have nothing to do with "nucleation". So please rephrase concepts like "nucleation potential" or "nucleation ability" - if the particle is already formed it is NOT nucleation! Also the comparison of various ion-molecule binding energies to neutral H_2SO_4-NH_3 is not really meaningful, and is not saying much about how the presence of $H_2S_2O_7$ on a surface might affect the adsorption/absorption of species such as $(COOH)_2$. Rather the comparison should be between for example SO_4^{2-} and/or HSO_4^- (present in H_2SO_4-containing aqueous droplets) and $S_2O_7^{2-}$.

Response: Thanks for your valuable comments. We agree with the reviewer's statement and according to the reviewer's suggestion, the sentence of "Moreover, we evaluated the nucleation potential of $S_2O_7^{2-}$ on SA-A cluster by considering geometrical structure and the formation free energies of the $(SA)_1(A)_1(S_2O_7^{2-})_1$ clusters." has been changed as "Moreover, we evaluated whether $S_2O_7^{2-}$ could lead to increased particle growth on SA-A cluster by considering geometrical structure and the formation free energies of the $(SA)_1(A)_1(S_2O_7^{2-})_1$ clusters." in Lines 14-16 Page 17 of the

revised manuscript. The sentence of "we predict that $S_2O_7^{2-}$ at the air-water interface has stronger nucleation potential" has been changed as "we predict that $S_2O_7^{2-}$ at the air-water interface would lead to increased particle growth" in Lines 25-26 Page 17 of the revised manuscript.

Comment 2.

Enhancement factor: I don't completely buy the authors justification for not using a constant SO_3 (e.g., of course there are other sinks, but this is easy enough to model as well - just like "other" sinks of e.g. H_2SO_4 can be modelled within ACDC if needed), but I can accept this: no further changes needed. Though I do have a slight terminological suggestion: in the new text on page 13, please don't use terms such as "more favorable" when what is really meant is that one reaction is more competitive (faster) due to higher reactant concentrations. (Both are quite favourable, but as most things in the atmosphere, the system is under kinetic, not thermodynamic, control.)

Response: Thanks for your valuable comments. According to the reviewer's suggestion, the sentence of "the hydrolysis reaction of SO_3 with $(H_2O)_2$ was more favorable than the $SO_3 + H_2SO_4$ reaction" has been changed as "the hydrolysis reaction of SO_3 with $(H_2O)_2$ dominates over the $SO_3 + H_2SO_4$ reaction" in Line 26 Page 14 of the revised manuscript.

Comment 3.

$H_2S_2O_7$ decomposition by hydration: it's unfortunate that the authors did not try to quantify this, but the caveat of likely H_2SO_7 hydration is now mentioned appropriately. However, I would suggest mentioning this major caveat once also in the abstract, as many people unfortunately only read the abstracts of studies. Other than that, no further changes needed. (As a side note, I don't think the [DSA] estimating using equilibrium constants for the SO_3 + SA reaction is particularly meaningful - rather a steady-state treatment between that formation reaction and hydrolysis would be needed - but in the absence of data for the hydrolysis channel I accept that it's a decent way of getting an upper bound, as long as it's properly recognised as such).

Response: Thanks for your valuable comments. According to the reviewer's suggestion, the sentence of "although $H_2S_2O_7$ is easily hydrolyzed with water to form H_2SO_4, it can directly participate in H_2SO_4-NH_3-based cluster formation and can present a more obvious enhancement effect on SA-A-based cluster formation" has been added in the abstract.