Dear Editor,

Thank you very much for your handling our manuscript "**Molecular-level evidence for marine aerosol nucleation of iodic acid and methanesulfonic acid**" (MS No.: acp-2021-595). We have revised the manuscript carefully according to reviewers' valuable comments. The point-to-point responses to the reviewers' comments are summarized below:

Referee #1:

Particle nucleation events have been repeatedly observed in marine environments and are associated with large increases in the concentration of particles smaller than 20 nm. While atmospheric observations provide the definitive evidence on which compounds are essential for this process, computational methods have the advantage of studying simple binary or ternary systems and revealing important interactions. Ning et al. investigated the nucleation mechanisms of iodic acid (IA) and methane sulfonic acid (MSA) using high level quantum chemical calculations combined with the Atmospheric Clusters Dynamic Code (ACDC). They proved that MSA can participate in the early nucleation steps with HIO_3 molecules, at least from a molecular dynamic point of view. They further show that the MSA enhancement over the HIO_3 system is dependent on the HIO_3 concentration and the temperature. The paper is well written and presents new insights into the marine nucleation mechanism. Therefore, I recommend the publication of this study in ACP after considering the comments listed below.

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

Comment 1: The authors have put a big emphasis on comparing their results to atmospheric observations, which is invalid in some cases and has weakened this study. For example, Figure 5b assumes that MSA concentration is equal to 1×10^7 molecules/cm^3 in all presented sites, clearly overestimating the MSA concentration in many locations. Additionally, the comparison to Beck et al. (2020) shown in Figure 6 does not give additional merit to the proposed MSA-IA mechanism, especially that the authors are aware that sulfuric acid (SA) and ammonia seem to play a significant role at this site and that IA and SA could have a synergetic role (Rong et al., 2020). It is recommended to put less emphasis on this comparison and instead focus on the results of the simulations, for example, moving figure S5 or S6 from the supplementary to the main text.

Response: We appreciate this constructive suggestion. As suggested by the reviewer (comment

1 and comment 19), the original Figure 5b has been moved to the supplement in order to weaken the comparison with the field observation. In its place, the redrawn Figure 5b in the revised manuscript presents the contribution of the IA-MSA clustering pathway at different [MSA] (10^6 – 10^7 molecules cm^{-3}) and [IA] (10^6 – 10^8 molecules cm^{-3}) in a similar form to Rong et al. (2020)'s Figure 3b. To further put less emphasis on comparison to Beck et al. (2020), Figure 6 has been replaced with Figure S5 in the main text according to the reviewer's suggestion.

Comment 2: The authors are encouraged to discuss the reasons behind the discrepancy in the formation rates presented here and in a previous study. The same group have reported that the formation rates of the pure IA system at [IA] of 1×10^8 cm^{-3} with a temperature of 278K and 2×10^{-3} s^{-1} CS is below 1×10^{-5} cm^{-3} s^{-1} (Rong et al., 2020), while the formation rates presented in Figure 3 of this study at similar conditions is higher than 1×10^{-2} cm^{-3} s^{-1}.

Response: Thanks for the reviewer's suggestions. The discrepancy in the formation rates is attributed to the fact that the cluster structure and thermodynamic properties were calculated at a different level of theory in the present work. In the previous study (Rong et al., 2020), the double-ζ basis set (aug-cc-pVDZ-PP) is employed for iodine atom. To get more accurate results, while the larger triple-ζ basis set (aug-cc-pVTZ-PP) is used for iodine in the present manuscript. In that case, the simulated J by ACDC based on the different ΔGs of the clusters obtained will in turn differ because of the sensitivity of cluster evaporation to ΔG.

Comment 3: The authors should also further discuss the limits of this study, causing 'discrepancies' with results reported in the literature. A very brief explanation is currently given in lines 273-274, but it is not sufficient. Optimally, the reader would understand the limits of this study compared to chamber or atmospheric measurements at an early stage of the manuscript. For example, the authors should discuss the difference between this study and that of He et al. (2021), resulting in different formation rates for the pure IA system, or that MSA is never present in the atmosphere without SA or that the MSA clusters are expected to be stabilized by water in the atmosphere (Chen et al., 2020).

Response: This is a very pertinent point – thank you for bringing it up. According to the reviewer's suggestion, we have added a description of the limitations of the IA-MSA nucleation mechanism in lines 268-273 of the revised manuscript as follows:

"In addition to IA and MSA, which are the focus of this study, other iodine-containing components such as HIO_2 and iodine oxides (I_2O_4 and I_2O_5) may also participate in the clustering processes. Moreover, considering the complexity of the marine atmosphere, other non-iodine nucleation precursors, such as SA, NH_3, amines, etc., may also affect the nucleation process. Particularly with SA, because MSA and SA coexist in the air and both are formed during the oxidation of DMS in the marine atmosphere. Therefore, in future studies, the influence of the above factors on the nucleation mechanism of marine aerosols will also be considered."

Specific comments:

Comment 4.

Line 42: Please add here the corrections He et al. (2021) made on the Sipila et al. (2016) proposed IA self-nucleation mechanism.

Response: According to the reviewer's helpful suggestion, the corrections He et al. (2021) made on the Sipila et al. (2016) was added in lines 40 – 42 of the revised manuscript as follows:

"…the coastal NPF is primarily driven by subsequential addition of IA and involves the participation of I_2O_5. More recently, He et al. (2021) demonstrated experimentally that, in addition to IA and I_2O_5, iodous acid (HIO_2) and I_2O_4 are also involved in the cluster formation process, with HIO_2 playing a key role in the stabilization of neutral IA clusters."

Comment 5.

Line 45: Beck et al. (2020) did not measure MSA and IA in the particle phase but in clusters using a CI-API-TOF (which could be gaseous). Thus, the sentence in its current form is misleading.

Response: Thanks for the reviewer's reminding. "…in the particle phase" has been corrected as "…in the smallest clusters" according to the study of Beck et al. (2021).

Comment 6.

Line 83: There is no footnote for the electronic supplementary information (ESI). Please remove the symbol after 'ESI'. (Also in lines 127 and 133).

Response: According to the reviewer's suggestion, all the symbols after 'ESI' have been removed in the revised manuscript.

Comment 7.

Line 84: Please add more information on the ACDC simulations. For example, that the simulations do not include the effect of water or charge.

Response: As suggested by the reviewer, more information on ACDC simulations has been added in lines 104 - 108 of the revised manuscript as follows:

“In the present study, the ACDC simulations only modelled the neutral cluster formation process and did not consider the charge, nor the effect of water. Since IA is weakly bound to water, it is less inclined to exist as hydration of IA in tropospheric conditions (Khanniche et al., 2016). Meanwhile, the nucleation efficiency of MSA and water is low (Arquero et al., 2017). Thus, the effect of water on the conclusion in this study is limited.”

Comment 8.

Line 88: What does the J in equation (2) stand for? It is misleading to have J here because the reader would think that it refers to formation rate, and the formation rate is not equal to dc/dt.

Response: Thanks for the reviewer’s professional suggestion. J does cause some misleading in equation (2) and has been removed from that equation.

Comment 9.

Line 99: Please refer to the ACDC boundary conditions presented in Table S5 in this section or somewhere else in the text.

Response: According to the reviewer’s suggestion, the ACDC boundary condition has been referred to the Table S5 in lines 107 - 108 of the revised manuscript. The added content is: “The settings of the boundary conditions of the ACDC simulations are discussed in Section S1 (ESI) and summarized in Table S5.”

Comment 10.

Line 113: Please replace ‘the’ by ‘a’ in the sentence: The similar situation...

Response: As suggested by the reviewer, ‘the’ has been replaced with ‘a’ in the similar situation of the manuscript.

Comment 11.

Line 144: Table S2 contains information about the Gibbs formation free energy only and does not include evaporation rates. Evaporation rates are presented in Table S4 and only at one temperature. This should be clarified.

Response: Thanks for the reviewer's suggestion. The contents in Table S4 were clarified in line 157 of the revised manuscript.

Comment 12.

Line 149: Refer to Table S4 after referring to Fig. 2b.

Response: According to the reviewer's suggestion, Table S4 has been referred after referring to Fig. 2b in the revised manuscript.

Comment 13.

Line 155: The supplement also shows similar figures to Fig. 2 but at 298 K (Fig. S2) and 258 K (Fig. S3). Please refer to these figures in the main text or delete them.

Response: The Fig. S2 and Fig. S3 has been referred in line 154 of the revised manuscript.

Comment 14.

Line 171: Should this be referring to the coagulation sink instead?

Response: Thanks for the reviewer's valuable suggestion. Coagulation sink is indeed an important treatment. Considering that a cluster size dependent coagulation sink coefficient has no important effect on steady-state cluster concentrations (McGrath et al., 2012), the constant condensation sink coefficients were chosen in the ACDC simulations of the present study.

Comment 15.

Line 191: Please adjust the caption of Fig. 4 to include the MSA concentration in the purple cones, the IA concentration in the red cones and the IA and MSA concentration in the blue cones.

Response: According to the reviewer's suggestion, the description of the color of cones in the caption of Figure 4 has been added to the revised manuscript (lines 204 - 205).

Comment 16.

Line 193: Also refer to Table S6 here.

Response: Thanks for your carefulness. The reference to Table 6 has been added in lines 208 - 209 of the revised manuscript as follows: “The specific *R* values were summarized in Table S6.”

Comment 17.

Line 193: Please refer to and discuss Figure S5 while presenting the temperature effect.

Response: According to the reviewer’s suggestion, Figure S5 has been referred and the corresponding discussion to were added in the lines 249 - 251 of revised manuscript.

Comment 18.

Line 224: Beck et al. (2020) did not show MSA-IA clusters and did not measure these exclusively in the particle phase (see comment on Line 45), so this reference cannot be used here to support your conclusion here.

Response: According to the reviewer’s suggestion, the citation of Beck et al. (2020) has been removed from the conclusion section of revised manuscript.

Comment 19.

Line 225-255: As the authors mention, the analysis shown in this section is highly dependent on the chosen MSA concentration for the simulations. An average MSA concentration of 1×10^7 molecules cm^{-3} is an overestimate for MSA measured in most of the cites sites. Thus, I suggest that the analysis is repeated with a more reasonable concentration or the reference to locations is omitted, and a figure similar to Rong et al. (2020)’s Figure 3b is presented instead (it could also be presented as a stacked bar graph with different temperatures listed next to each other). Otherwise, Figure 5b can be moved to the supplement, and less emphasis on it is given in the main text.

Response: According to the reviewer’s valuable suggestion, the mentioned analysis has been repeated at a more reasonable concentration of MSA (2.5×10^6 molecules cm^{-3}) (Bork et al., 2014) and the resulting Figure 5b has been moved to the supplement (Fig. S5). In the revised manuscript, the modified Figure 5 presents the contribution of the IA-MSA clustering pathway at varying [MSA] ($10^6 - 10^7$ molecules cm^{-3}) and [IA] ($10^6 - 10^8$ molecules cm^{-3}), which is like Rong et al. (2020)’s Figure 3b. For the reviewer’s convenience, the modified Figure 5 in the revised manuscript is presented as following:

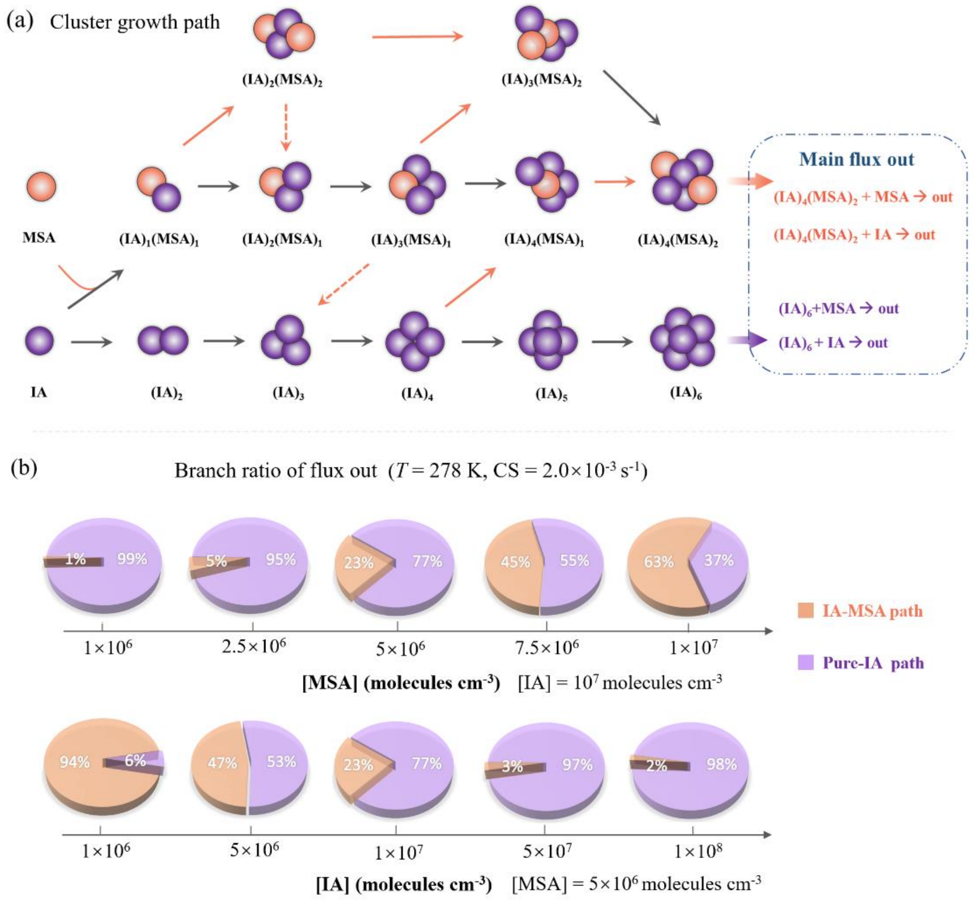


Figure 5. (a) Main cluster growth pathway of IA-MSA nucleating system at T = 278 K, CS = 2.0×10^{-3} s^{-1}, [IA] = 10^7 and [MSA] = 5×10^6 molecules cm^{-3}. The black and orange arrows refer to the pathways of colliding with IA and MSA, respectively, where the dashed arrows indicate the evaporation of MSA. **(b)** Branch ratio of IA-MSA (orange pie) and pure-IA (purple pie) growth pathway under varying [MSA] (10^6 – 10^7 molecules cm^{-3}) and [IA] (10^6 – 10^8 molecules cm^{-3}).

The corresponding statements of Figure 5b were added as follows:

"In the atmosphere, the distribution of IA and MSA varies by regions, affecting the contribution of IA-MSA clustering pathways accordingly. Hence, the branch ratios of flux out through the IA-MSA path (orange pie) and pure-IA path (purple pie) at varying [MSA] (10^6 – 10^7 molecules cm^{-3}) and [IA] (10^6 – 10^8 molecules cm^{-3}) are presented in Fig. 5b to access the impact of IA-MSA mechanism. As shown in Fig. 5b, the branch ratio of IA-MSA and pure-IA path is highly dependent on [MSA] and [IA]. At the condition of T = 278 K, CS = 2.0×10^{-3} s^{-1} and [IA] = 10^7 molecules cm^{-3}, the contribution of IA-MSA path increases from 1% to 63% with the increasing of [MSA]. Additionally, given the uneven distribution of IA, the analysis was further carried out within the atmospherically relevant range of [IA] (10^6 – 10^8 molecules cm^{-3}). The results show that the contribution of IA-MSA path decreases from 94% to 2% with the increasing of

[IA] (10^6 –10^8 molecules cm^{-3}). These findings indicate that the IA-MSA mechanism contributes more in regions with higher [MSA] and lower [IA]. Furthermore, the branch ratio was calculated based on field conditions (temperatures and [IA]) reported by He et al. (2021) and presented in Fig. S5. The results indicate that the IA-MSA mechanism does have stronger effects in polar regions than in mid-latitude coastal regions due to lower temperatures, which is also consistent with the above findings.”

To further put less emphasis on comparison to Beck et al. (2021), Figure 6 in the revised manuscript has been replaced with Figure S5 according to the reviewer’s suggestion.

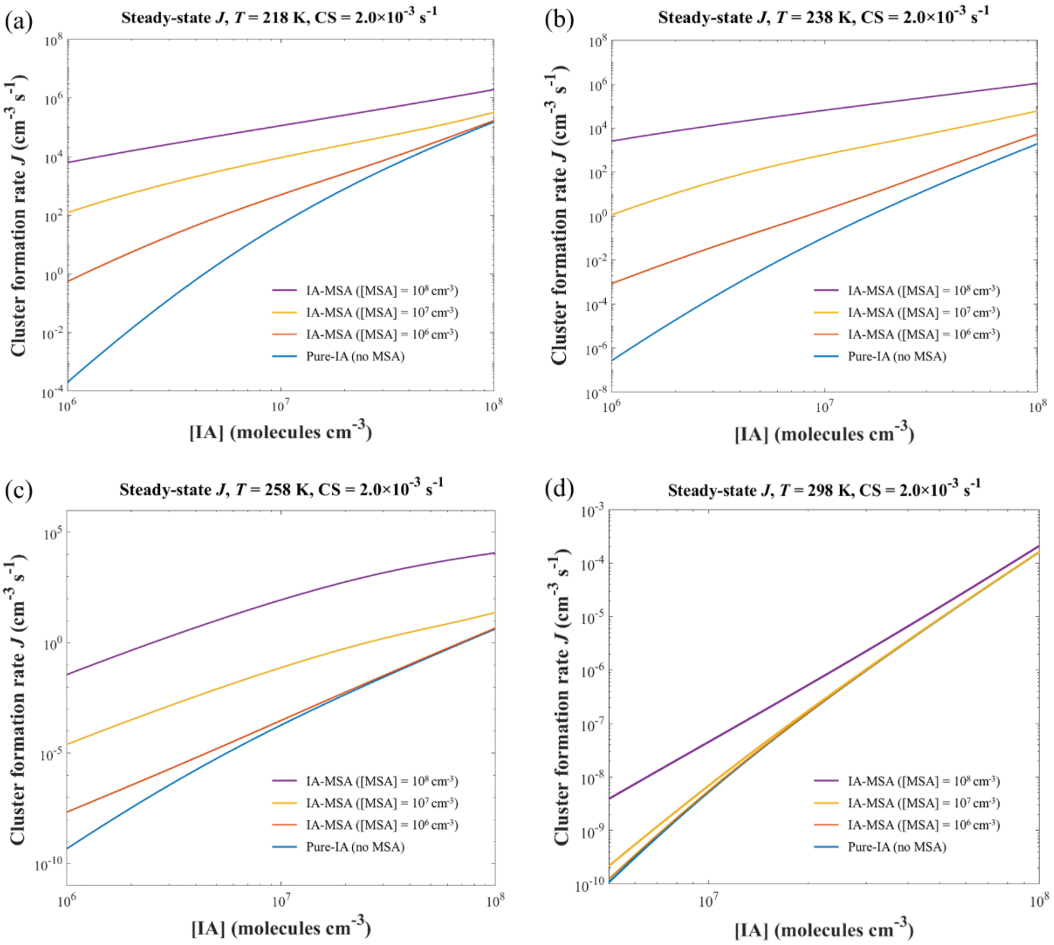


Figure 6. The simulated cluster formation rate J (cm^{-3} s^{-1}) of the IA-MSA system at different temperatures (a) 218, (b) 238, (c) 258, (d) 298 K, [IA] = 10^6 – 10^8 molecules cm^{-3}, [MSA] = 0, 10^6, 10^7, 10^8 molecules cm^{-3}, and CS = 2.0×10^{-3} s^{-1}.

The corresponding statements of Figure 6 were added in the revised manuscript as follows: “Most of the analysis above in the text was performed at 278 K. To further probe the impact of temperature on J systematically, Figure. 6 presents the simulated J at additional temperatures (218, 238, 258 and 298 K), [IA] = 10^6 – 10^8 molecules cm^{-3}, [MSA] = 10^6 (red line), 10^7 (yellow line), and 10^8 molecules cm^{-3} (purple line). At a relatively high T = 298 K (Fig. 6d), the improvement on J by the addition of MSA was not significant compared to the pure-IA system, except at higher [MSA] = 10^8 molecules cm^{-3} and relatively lower [IA]. At lower T = 258 K (Fig. 6c), the enhancement on J by MSA is stronger in all cases except at lowest [MSA] = 10^6 molecules cm^{-3}. Moreover, such boost on J was further enhanced at 238 K (Fig. 6b). Lower concentrations of MSA (10^6 molecules cm^{-3}) also significantly promote the formation of IA clusters, mainly because the low temperature weakens the cluster evaporation."

Comment 20.

Line 256-276: This section is dedicated for ACDC simulations at conditions of MSA, IA, temperature, and CS identical to those reported in Beck et al. (2020). However, the comparison to the measurements at Ny- Ålesund is not straightforward, as mentioned in the 1st general comment. Please discuss more the limitations or give less emphasis on this comparison.

Response: According to the reviewer's value suggestion, the comparison to the measurements at Ny-Ålesund (the original Figure 6) and the corresponding statement have been removed from the main text.

Comment 21.

Line 284-286: This sentence must be rephrased to have a less strong statement because the analysis performed depends highly on the chosen MSA concentration.

Response: Thanks for the reviewer's constructive suggestion. The statement about the contribution of IA-MSA clustering pathways has been rephrased to a less strong form in lines 281 - 283 of the revised manuscript as follows: "Moreover, the IA-MSA clustering pathway potentially contributes more in the colder polar regions, especially with higher [MSA] and lower [IA]. The IA-MSA mechanism is highly dependent on the distribution of MSA and IA in the marine atmosphere."

Comment 22.

Line 293: It is essential to mention here the other important players. For example, MSA is never present in the atmosphere without SA as both are important DMS oxidation products.

Response: According to the reviewer's pertinent suggestion, the statement about other important players for marine NPF was added in lines 287 - 289 of the revised manuscript as follows: "… multi-component nucleation model. For example, both SA and MSA originate from the oxidation of DMS, so their coexistence in the atmosphere may synergistically promote the formation of IA clusters, which is worthy of future studies."

Comment 23.

Line 307: Please review the reference list:

- There are references with missing journal names or abbreviated journal names in the author list. For example, Bates et al. (2020), Elm and Kristensen et al. (2017), Hatakeyama et al. (1982), Takegawa et al. (2020).
- There are some references that do not have the complete author list. For example, Beck et al. (2020) and He et al (2021).
- The Seinfeld and Pandis citation is incorrect and refers to Jeffrey Steinfeld's review of the book.
- Provide a URL for Stewart (2016).

Response: Thanks for the reviewer's carefulness review, the above references have been completed and all references have been double-checked.

Comment 24.

Figure S1: The caption of this figure could be misleading because the word 'stable' could be interpreted from the view of having a ratio of collision frequency to total evaporation that is higher than 1 (Fig. 2c). So please replace the word 'stable' with the 'lowest free energy'. Please also include the temperature in the caption.

Response: Thanks for the reviewer's valuable suggestion. "…identified stable configurations" has been corrected to "…identified configurations with lowest free energy" in the revised supplement. The temperature has been added in the caption of Figure S1.

Referee #2:

The revision of this manuscript is satisfactory and recommended to be accepted.

Response: We would like to thank the reviewer for taking the time to review our manuscript and for the positive comments.

Referee #3:

This manuscript investigates the nucleation mechanism of iodic acid and methanesulfonic acid, two important species for marine particle formation and growth processes. While the topic is interesting and the presentation of this work (text, figure) is good, there are substantial flaws associated with the content.

The conclusion of this manuscript is in clear conflict with several important field observations cited here (Sipilä et al. 2016, Baccarini et al. 2020 and Beck et al. 2020). Despite the authors tried very hard to somehow use these studies to validate this manuscript, the presented results in this manuscript really show the opposite – IA-MSA nucleating is probably not very important in these regions, as shown by not matching results compared to field observations (if we have to assume that they are calculating the right cluster formation rates).

I'm surprised by comparing an earlier but recent work from the same group (Rong et al. 2020; Xiuhui Zhang is the corresponding author for both studies) and found that there are substantial differences (3 to 4 orders of difference) in the calculated cluster formation rates for the same system (pure IA nucleation) using similar methods. My investigation on this suggests that the authors are using slightly different methods (double zeta basis set in Rong et al. 2020 and triple zeta basis set in this manuscript) in these two manuscripts. However, this is unlikely the primary reason for such a substantial difference. Additionally, the authors used different geometries for the iodine clusters presented in these two studies. E.g., in Rong et al. 2020 the iodic acid dimer is connected by two halogen bonds while in this study they are connected by two hydrogen bonds. I'm astonished by the fact that there is not a single word mentioning this in this manuscript.

Additionally, a fundamental criterion in all QC+ACDC studies is that the cluster formation of the larger clusters (out of box clusters) needs to be at the kinetic rate (McGrath et al. 2012), so the accommodation coefficient can be assumed unity. This is not the case as shown by Fig.2C and Fig. S2C. The largest calculated growth to evaporation ratio (thus somehow a growth potential) is around 7 at 278 K and is around 0.2 at 298 K. In my opinion, these are too small numbers to be considered stable and the authors need to continue their calculations to find the stable clusters. As this is a fundamental problem, the whole manuscript would need to be substantially modified to accommodate this change. Finally, the clusters formation rates for pure iodic acid provided in this study are roughly 6 orders of magnitude lower than recent results published by the CLOUD experiments (He et al. 2021). While I understand that quantum chemical calculations can have uncertainties and experiments also have errors, such a substantial difference is not acceptable since this potentially points out substantial problems in the calculations. Note, addressing my last point will increase the discrepancy, not reducing it.

Given the significant problems noted above, I would recommend rejecting this manuscript from ACP. However, if the authors are given chance to respond, the authors are urged to address all the raised points and to interpret their results properly even if they get negative results which are still much better than overstatements, especially considering most ACP audiences do not have strong backgrounds in quantum calculations and will adopt wrong numbers from this study.

Response: Thanks for the reviewer's professional and helpful comments. And we have carefully revised the manuscript accordingly.

Major comments:

Comment 1: The authors deployed identical QC methods in this work and their earlier work (Rong et al. 2020; Xiuhui Zhang is the corresponding author for both studies). The authors calculated formation rates for pure IA nucleation in both studies, but a 1000 to 10,000 times difference can be found by comparing Fig. 3 of this study and Fig. 2 in Rong et al. 2020. A further check on Table S12 in Rong et al. 2020 and Table S2 in this study shows substantially different Gibbs free energy values for the same IA clusters at the same temperature. E.g., at 278 K, Rong et al. 2020 gives a ΔG of -5.92 kcal mol^{-1} while this study gives -9.51 kcal mol^{-1}. The difference seems small but as it goes in the exponential part of the evaporation rate equation, the resulted evaporation rates can be significantly different. My simplified calculation suggests that a -9.51 kcal mol-1 value easily results in a 500 times lower evaporation rate of iodic acid dimer compared to a value of -5.92.

A further investigation on this matter comparing these two papers suggest at least two major differences. Rong et al. 2020 uses a double zeta basis set (aug-cc-pVDZ-PP) while this study uses a triple-zeta basis set (aug-cc-pVTZ-PP). However, I highly doubt that this is the primary reason for such a significant difference. I further tried to compare the geometries of the IA dimer in these two papers. As the authors provided wrong coordinates for IA dimer in Supp-Section 6 in Rong et al. 2020, I have to infer from their Fig. S3 (cited here as Figure R1). The IA dimer is connected by two halogen bonds. However, in this study, the IA dimer is connected by two hydrogen bonds from my reproduced results based on coordinates in Table S7 of this manuscript. Additionally, the geometries of larger IA clusters in this study are also significantly different from Rong et al. 2020. The authors should visualize the IA clusters and any other clusters which are not yet visualized besides IA-MSA clusters in Fig S1.

Because of these significant differences in these two papers, the authors are urged to do at least as follows: 1) they should calculate all the commonly used clusters (at least all of the IA clusters) in Rong et al. 2020 and this study by both the double zeta and triple zeta basis sets for two sets of geometries provided in both papers (so 2x2 matrix for every cluster). 2) the authors should discuss the results coming from item 1) in the Main Text and give reasons for their updated results and potential errors associated with the Rong et al. 2020 or this study. 3) The authors are encouraged also to discuss in the main text why the geometries provided in their current study should represent global minima and why the geometries are significantly different in Rong et al. 2020 and this study.

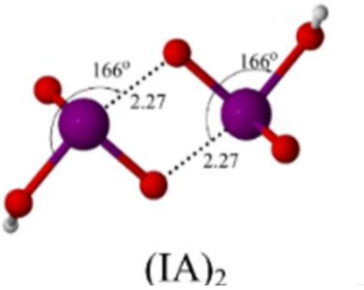


Figure R1. Screenshot of Figure S3 in Rong et al. 2020.

Response: Thanks for your professional and valuable comment. The responses to each of the reviewer's suggestions are presented below.

Item 1) from the reviewer: "they should calculate all the commonly used clusters (at least all of the IA clusters) in Rong et al. 2020 and this study by both the double zeta and triple zeta basis sets for two sets of geometries provided in both papers (so 2x2 matrix for every cluster)."

Response: As suggested by the reviewer, all of the studied IA clusters in Rong et al. 2020 and this study have been calculated at both double zeta and triple zeta basis sets. The resulting ΔGs at 278K of IA clusters are collected in the following Table A1, which has also been added in the revised supporting file (Table S2).

Table A1. The Gibbs formation free energies ΔG_{278K} (kcal mol^{-1}) of the studied IA clusters in Rong et al. 2020 and this study calculated at the RI-CC2/aug-cc-pVTZ(-PP)//ωB97X-D/6-311++G(3df,3pd) + aug-cc-pVDZ-PP (for I) (DZ) and RI-CC2/aug-cc-pV(T+d)Z(-PP)//ωB97X-D/6-311++G(3df,3pd) + aug-cc-pVTZ-PP (for I) (TZ), respectively.

Cluster	ΔG_{278K}-Rong (DZ)	ΔG_{278K}-Rong (TZ)	ΔG_{278K}-this study (DZ)	ΔG_{278K}-this study (TZ)
(IA)$_2$	-5.92	-8.07	-7.96	-9.51
(IA)$_3$	-15.73	-18.47	-16.05	-18.79
(IA)$_4$	-34.41	-38.48	-34.41	-38.48
(IA)$_5$	-52.37	-56.28	-52.37	-56.28
(IA)$_6$	-70.67	-74.41	-70.67	-74.41

It is worth noting that the $(IA)_2$ and $(IA)_3$ clusters in Rong et al. 2020 and this study are different. Specifically, the $(IA)_2$ in Rong is halogen-bonded, while in this study it is hydrogen-bonded. And this issue will be further discussed in the response to item 2) below.

Item 2) from the reviewer: "the authors should discuss the results coming from item 1) in the Main Text and give reasons for their updated results and potential errors associated with the Rong et al. 2020 or this study."

Response: Thanks for the reviewer's valuable suggestion. The following are **A)** the causes of the differences in values, **B)** the reasons for updating the data , and **C)** the potential errors in the Rong et al. 2020 or this study.

A) In the Rong et al. 2020 and this study, the levels of theory employed in cluster structure optimization and frequency calculations are different. The larger triple-ζ basis set (aug-cc-pVTZ-PP (for iodine)) was used in this study compared to the double-ζ basis set (aug-cc-pVDZ-PP (for iodine)) of Rong et al., (2020). As shown in the above Table A1 (second and third columns), the different basis sets lead to differences in the calculated ΔGs of IA clusters ($(IA)_{2\text{-}6}$) in the range of 2.15 ~ 4.06 kcal mol^{-1}. This is one reason for the difference in the calculated ΔGs values.

   In addition, as mentioned above and shown in Fig. A1, the geometries of IA clusters (IA dimer and IA trimer) employed in the Rong et al. 2020 and the present manuscript are different. This is another reason. It further leads to a difference of 0.32 ~ 1.44 kcal mol^{-1} of ΔGs values , at the same level of theory (third and fifth columns in Table A1).

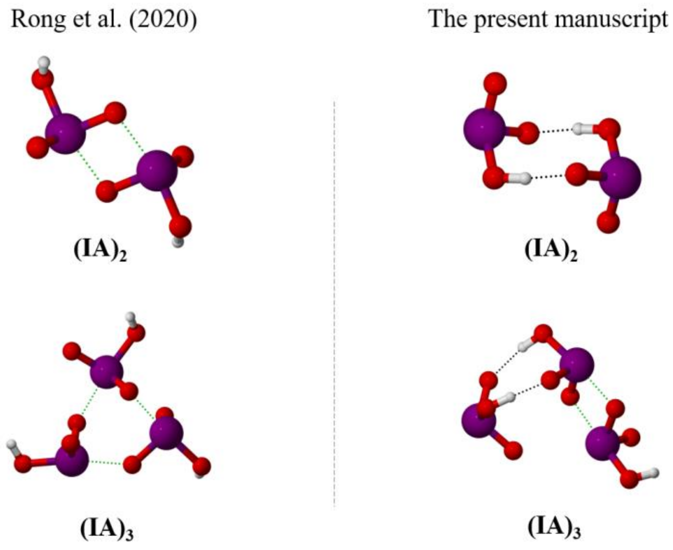


Figure A1. The different IA cluster structures employed in Rong et al. (2020) and the present manuscript (including IA dimer and trimer).

B) The following are the reasons for the choice of the double-ζ basis set in the Rong et al. 2020, and the subsequent update of the results in the present paper:

Due to large number of electrons in iodine atom, the QC calculations of clusters involving IA are expensive. Considering the variety of clusters calculated in the Rong et al. 2020, coupled with the limited computational resources available at that time, the double-ζ basis set was chosen as a compromise between computational accuracy (Benchmark, Table S1-S6 in Rong et al. 2020) and resource consumption to present the reasonable trends and corresponding mechanisms of sulfuric acid and NH_3 promoting IA cluster formation. After updating the computational resources, in the present manuscript, the larger triple-ζ basis set was herein used to reduce errors in the subsequent dynamic simulations because higher level of theory usually implies a better calculation accuracy.

As suggested by the reviewer, we have declared the reason for updating the results in the lines 66-69 of the revised manuscript.

C) The potential errors in the Rong et al. 2020 or this study are as follows:

Thanks for the reviewer's professional comments. After careful comparison (Table A1) we found that with the same structure, the ΔG calculated in Rong et al. 2020 at the double-ζ basis set (aug-cc-pVDZ-PP for iodine) is higher than those under triple-ζ basis set (aug-cc-pVTZ-PP for iodine), which will lead to a lower cluster formation rate.

Item 3) from the reviewer: "The authors are encouraged also to discuss in the main text why the geometries provided in their current study should represent global minima and why the geometries are significantly different in Rong et al. 2020 and this study."

Response: Thanks for the reviewer's helpful suggestion. The reasons for the selection of the different IA cluster conformations (IA dimer and trimer) in the Rong et al. 2020 and this work are summarized below:

The sampling process of cluster isomers is computationally demanding and subject to uncertainties (Elm et al., 2020). The employed IA dimer and trimer in this manuscript are not the lowest-energy isomers due to our incomplete consideration. Thanks for the reviewer's careful review. The IA dimer and trimer with lowest energy obtained after recalculation are consistent with those of Rong et al. 2020. In addition, we have also checked the other employed IA clusters and confirmed that these clusters were selected with the lowest energies.

Thanks again for the reviewer's professional comment. We have corrected the structure of IA dimer and trimer to be consistent with that of Rong et al. 2020. Accordingly, we have also recalculated all relevant data and updated graphs and tables in the revised manuscript and revised supporting file.

Item 4) from the reviewer: "The authors should visualize the IA clusters and any other clusters which are not yet visualized besides IA-MSA clusters in Fig. S1."

As suggested by the reviewer, all the clusters studied in the present manuscript, containing IA clusters as well as other previously unpresented clusters, have been visualized in the revised Fig. S1 of the supplement. In addition, by a comparison of with the original data, we find that the mentioned Cartesian coordinates of IA dimer and trimer provided in SI of Rong et al. (2020) lost the corresponding negative sign due to typos. And the corrected coordinates are added in the Appendix in this response. Further, we are contacting the corresponding publisher to correct this issue in these days.

Comment 2: Another fundamental problem is that neither this study nor Rong et al. 2020 seem to even remotely repeat CLOUD measurements on pure iodic acid nucleation (He et al. 2021). For example, this study calculates 6 orders of magnitude lower cluster formation rates compared to He et al. 2021 and the difference goes to 9 orders when comparing Rong et al. 2020 with He et al. 2021. While it is understandable that the large number of electrons in iodine atom cause substantial challenges in QC calculations, such a substantial difference must be explained, as it could potentially suggest that either there are some fundamental errors in the QC + ACDC calculations related to coding, methods and basis sets employed or because the authors are calculating based on wrong assumptions.

Response: Thanks for the reviewer's constructive suggestions. We realized that there is a significant discrepancy between the simulated rates in the present manuscript and the CLOUD measurements (He et al. 2021). The reasons for this phenomenon are as follows (A and B):

A) The nucleation components involved in the ACDC simulation (only IA) of the present manuscript and CLOUD experiment (He et al., 2021) are different. If we understand correctly, He et al., (2021) presented a high dependence of the nucleation rate on the concentration of IA (Fig. 1, He et al., 2021), but in fact, not only IA is involved in nucleation, but also other iodine components, such as I_2O_4, I_2O_5, and HIO_2, play a non-negligible role in forming IA particles (Fig. 2 and Fig. 3, He et al., 2021). Therefore, using the simulated rate containing only IA to compare the experimental nucleation rates of multiple iodine components such as I_2O_4, I_2O_5, and HIO_2, will inevitably lead to large differences.

B) In addition to the differences in the components mentioned in item 1), as the reviewer

> mentioned, there are uncertainties in both experiments and simulations. The superposition of errors may further amplify the difference between experiments and simulations. In addition, there are also differences in the way nucleation rates are calculated between the experiments and simulations. All of these would lead to the fact that we cannot simply assess the cause of the discrepancy and attribute it to one point.

Therefore, there is uncertainty in comparing the specific results of experiments and simulations in this case. In fact, theoretical calculations prefer to show that the IA-MSA system have higher cluster formation rates than the pure-IA system, indicating the enhancement of MSA on IA cluster formation, which is particularly evident in marine regions with rich MSA and sparse IA.

Comment 3: I'm not convinced that there are sufficient IA-MSA clusters calculated in this study which would allow the authors to consider larger clusters than IA4MSA2 and IA6 as nucleated clusters. The largest ratios of growth to evaporation in Fig. 2c are 2 (IA4MSA2) and 7 (IA6) which barely provide growth potential for these clusters. How can these clusters be considered stable enough? This even more true when looking at the Fig. S2C in which the largest value is 0.2. This is an essential assumption for this study and all other QC + ACDC studies and many of the numbers in this study will make no sense if this is not varied. The author should extend their calculations until finding stable clusters.

Additionally, the authors mistakenly conclude in lines 171-172 that "condensation sink" has a minor impact on the cluster formation rate. First, likely the authors are not talking about condensation sink but coagulation loss or a combination of other losses. This should be clarified, and they should describe clearly that whether they applied the "CS" uniformly for all clusters. Typical in ACDC models one would set monomer species as constants and therefore condensation sink does not affect the monomer condensation. If their calculations correctly find stable clusters with low enough evaporation rates (and thus high growth potential from condensation), they will likely find the "CS" as an important factor influencing their calculated cluster formation rates. It's possible that they find such odd results because they have not found the clusters with low enough evaporation rates as mentioned above. This is evident from the fact that if stable clusters are found, condensational growth is likely dominating the growth and its value is comparable to their "CS" values of 1e-4 to 2.6e-3 s^{-1} when acid is ranging from 1e6 to 1e8 cm^{-3}. The lowest evaporations rates given in this study (Fig. 2B) are not far from the "CS".

Response: Thanks for the reviewer's professional comment. The specific response to each item of the reviewer are as follows:

Item 1) from the reviewer: "The largest ratios of growth to evaporation in Fig. 2c are 2 (IA4MSA2) and 7 (IA6) which barely provide growth potential for these clusters. How can these clusters be considered stable enough?"

Response: In ACDC simulations (McGrath et al., 2012), relatively stable clusters are those in which collisions with molecules can be assumed to dominate over cluster evaporation (Oona and Tinja, 2020). Specifically, it is to calculate the ratio of the rate at which the studied cluster collides with the IA or MSA monomer to its total evaporation rate. The following is an example of the IA4MSA2 cluster.

$$\frac{\beta_{\mathrm{I}} C_{\mathrm{I}}}{\sum \gamma} = \frac{\beta_{\mathrm{I}} C_{\mathrm{I}} \cdot C_{\mathrm{IA4MSA2}}}{\sum \gamma_{\mathrm{IA4MSA2}} \cdot C_{\mathrm{IA4MSA2}}} = 2 > 1$$

where β_{I} is the rate coefficient of cluster collision with IA monomer, C_{I} is the concentration of IA monomer, and $\sum \gamma$ is the total evaporation rate coefficient of the studied cluster.

If $\beta_{\mathrm{I}} C_{\mathrm{I}} / \sum \gamma > 1$, the corresponding cluster would be considered to be relatively stable against evaporation, and has the "growth potential". Once these stable clusters on the boundary form and grow further out of the simulated system, these formed clusters out of system are unlikely evaporate back into the system (Oona and Tinja, 2020; McGrath et al., 2012). Thus, as suggested by the reviewer, the controversial statement "stable enough" has been changed to "relatively stable" in the revised manuscript. In addition, the above explanation has been added in lines 162 – 167 (main text) and Section S1 (supplement) for the clarity of the reader.

Item 2) from the reviewer: This even more true when looking at the Fig. S2C in which the largest value is 0.2. This is an essential assumption for this study and all other QC + ACDC studies and many of the numbers in this study will make no sense if this is not varied.

Response: This is a very helpful point – thank you for bringing it up. In fact, the present ratios of growth to evaporation ($\beta_{\mathrm{I}} C_{\mathrm{I}} / \sum \gamma$) in Fig. 2C, Fig. S2C and Fig. S3C are the lowest values, since the chosen C_{I} at this point is the lowest concentration of IA monomer (1.0×10^6 molecules cm^{-3}). In fact, the $\beta_{\mathrm{I}} C_{\mathrm{I}} / \sum \gamma$ of the mentioned cluster is in the range of 0.2 - 20 under the studied range of IA concentration (10^6 - 10^8 molecules cm^{-3}). In this case, the clusters (value of 0.2 at [IA] = 1.0×10^6 molecules cm^{-3}) can grow out of the system at most of studied [IA] (5×10^6 ~ 10^8 molecules cm^{-3}.

Thanks to the professional advice of the reviewer. In order to let the reader know that the presented $\beta_{\mathrm{I}} C_{\mathrm{I}} / \sum \gamma$ is the minimum value, we have added notes in lines 163 – 165 (main text) and Section S1 (supplement) to remind the reader.

Item 3) from the reviewer: "First, likely the authors are not talking about condensation sink but coagulation loss or a combination of other losses. This should be clarified, and they should describe clearly that whether they applied the "CS" uniformly for all clusters.",

Response: As pertinently suggested by the reviewer, the statement about "CS" uniformly for all clusters has been added in line 184 of the revised manuscript. This treatment is also commonly used in other theoretical simulation studies (Bork et al., 2014; Shen et al., 2019; Xu et al., 2020).

Item 4) from the reviewer: "Additionally, the authors mistakenly conclude in lines 171-172 that "condensation sink" has a minor impact on the cluster formation rate… .

Response: In this study, before getting the corresponding conclusions ("the different CS ($1.0\times10^{-4} \sim 2.6\times10^{-3}$ s^{-1}) have a minor impact on the cluster formation rate"), we have tested the effect of different CS values on cluster formation rate J (cm^{-3} s^{-1}) in Fig. S4 of supporting file. For the reviewer's convenience, we have copied and presented it as follows.

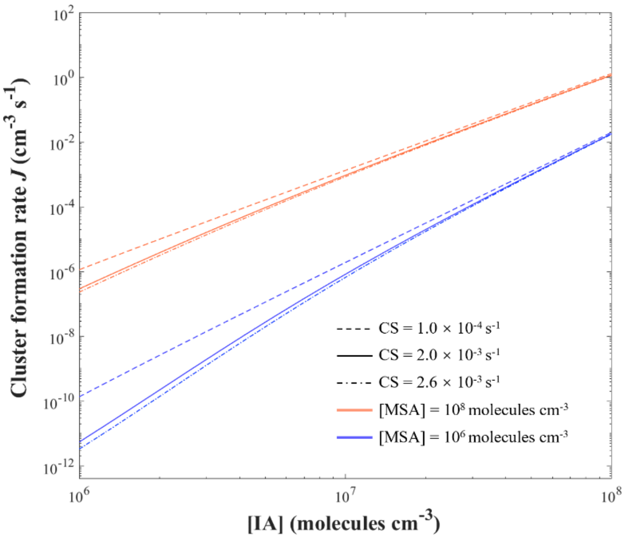


Figure S4. The simulated cluster formation rate J (cm^{-3} s^{-1}) of the IA-MSA system at different condensention sink (CS) coefficients (CS = $1.0\times10^{-4} \sim 2.6\times10^{-3}$ s^{-1}), T = 278 K, [IA] = $10^{6} \sim 10^{8}$ molecules cm^{-3}, and [MSA] = 10^{6} (blue lines), 10^{8} (orange lines) molecules cm^{-3}.

As the reviewer expertly suggested, as shown in Fig. 4, different CS values indeed have an impact on J. In the case of higher [IA] and [MSA], the J is relatively high and the effect of CS is relatively little. However, the effects of different CS values are more pronounced in the case of low J because the order of magnitude of J at this moment is comparable to or lower than the employed CS values. To ensure the rigor of the statement, we have changed the "the different CS have a minor impact on the cluster formation rate" to the "the different CS have an impact on the simulated J, especially in the case of low J (Fig. S4), but less on presenting the promotion of MSA on IA cluster formation and the main conclusions of this study." in lines 182-184 of the revised manuscript. Thanks again for the reviewer's kind reminder.

Specific comments:

Comment 4. The authors mention three important studies (Sipilä et al. 2016, Baccarini et al. 2020 and Beck et al. 2020) as the backbone of this study. However, none of these studies support their calculations. Sipilä et al. 2016 and Baccarini et al. 2020 indicate that IA NPF dominate the NPF events they observed at Mace Head and the central Arctic, respectively, while Beck et al. 2020 suggests SA-NH3 is the dominating nucleation mechanism in Ny Alesund by comparing SA with nucleation rates. They do, however, suggest MSA contributes to particle growth. These studies go against what the authors suggest.
Lines 273-274: this is not enough to respond my comments in the initial screening. Agreeing with the importance of other molecules goes against their title, and thus the main theme of this manuscript. If I understand correctly from the literature, other iodine species can be formed at the same time as iodic acid. Similarly, both MSA and SA are formed from DMS in marine environments. These species are very likely to co-exist in marine environments at different levels. Picking up two species from the list and claim it to be a marine aerosol nucleation mechanism is not acceptable unless it is either supported by their calculations or by field observations. However, the mentioned three studies clearly disagree with the IA-MSA nucleation mechanisms. Additionally, the cluster formation rates derived in this study are too low to explain field observations (details below).

Response: Thanks for your pertinent feedback and suggestions. The reviewer's comments and corresponding responses are summarized below.

Item 1) from the reviewer: "The authors mention three important studies (Sipilä et al. 2016, Baccarini et al. 2020 and Beck et al. 2020) as the backbone of this study. However, none of these studies support their calculations."

Response: We agree with you that the three mentioned references (Sipilä et al., 2016; Baccarini et al., 2020; Beck et al., 2021) do not support the importance of the IA-MSA mechanism. However, the findings of these studies were based on observations obtained in specific areas and do not inherently conflict with the results presented in this manuscript. The reasons are as follows (A, B, and C):

A) For Mace Head (Sipilä et al., 2016), the simulations were only compared with the observations at this site in Fig. 5b of original manuscript. The simulation results show that the IA-MSA mechanism plays a much smaller role of 1% in Mace Head where IA concentrations can be as high as 10^8 molecules cm^{-3}. In the lines 238 - 239 of preprint manuscript, we have also emphasized that for regions with high IA concentrations like Mace Head, the contribution of the IA-MSA mechanism is minor and the NPF process remains dominated by IA. This conclusion does not seem to conflict with Sipilä et al. (2016).

B) During the Arctic Ocean 2018 expedition (Baccarini et al., 2020), the simulation results based on the field conditions ($T = 268$ K, CS = 1×10^{-4} s^{-1}, [IA] = 10^5~10^6 and [MSA] = 10^5 molecules cm^{-3}) in the present manuscript shows a contribution of 1% - 16% to cluster formation from the IA-MSA mechanism. Most of the contribution is attributed to nucleation of IA (declared in lines 250 - 254 of the preprint manuscript). This seems not to be in conflict with the conclusion that the NPF observed by Baccarini et al. (2020) is mainly driven by IA.

C) As to Ny-Ålesund (Beck et al., 2020), the present results in Fig. 6 suggest that the simulated rate of IA-MSA mechanism only partially matches a small fraction of the observed rates, and most of the unmatched rate might come from the contribution of SA-NH_3 ion-induced nucleation (declared in lines 270 - 272 of the preprint manuscript). This result seems also not conflicting.

In addition to the mentioned specific regions with higher [IA] and relatively lower [MSA], considering the vastness of the ocean, the simulations presented in this manuscript suggest that the IA-MSA mechanism could potentially play an important role in regions with sparse IA and rich MSA.

Thanks again for the reviewer's careful review and valuable comment. To add, the original intention of citing the work of Sipilä et al. (2016) and Baccarini et al. (2020) in the background was only to show that IA is a key NPF driver in coastal and polar regions, not to support the importance of IA-MSA mechanisms in these regions. And in the section of rate analysis, the citation of Sipilä et al. (2016) aims to support the reasonableness of the IA atmospheric concentration range employed in the simulations, rather than compare with the corresponding field observation.

Item 2) from the reviewer: "Similarly, both MSA and SA are formed from DMS in marine environments. These species are very likely to co-exist in marine environments at different levels. Picking up two species from the list and claim it to be a marine aerosol nucleation mechanism is not acceptable unless it is either supported by their calculations or by field observations. However, the mentioned three studies clearly disagree with the IA-MSA nucleation mechanisms. Additionally, the cluster formation rates derived in this study are too low to explain field observations (details below)".

Response: Indeed, as the reviewer expertly suggested, there are many components in the real atmosphere besides IA and MSA, such as SA or other iodine components that may participate in the nucleation process together. However, this study focuses more on the impact of MSA, a representative oceanic sulfur-containing acid. And other components will be taken into account in future research.

Comment 5. Fig 6: this figure is misleading. The authors overlooked the conclusions from Beck et al. 2020 which suggested SA-NH3 to be the nucleating mechanism in Ny-Alesund, supported by measurement data. The authors adopted the cluster formation rates from Beck et al. 2020 and presented it as a rectangle in the plot which falsely overlaps with their data. If iodic acid is nucleating with MSA in Ny Alesund, one would expect that the cluster formation rates go up with increasing iodic acid. If the data are correctly presented, the authors will find that their data are very far away from the field observation they presented. All the discussions following this figure are therefore problematic. If I have to assume that the authors do calculate the IA-MSA cluster formation rates correctly (which I doubt as presented below), the results suggest that the IA-MSA mechanism does not play a role in Ny Alesund thus agreeing with Beck et al. 2020.

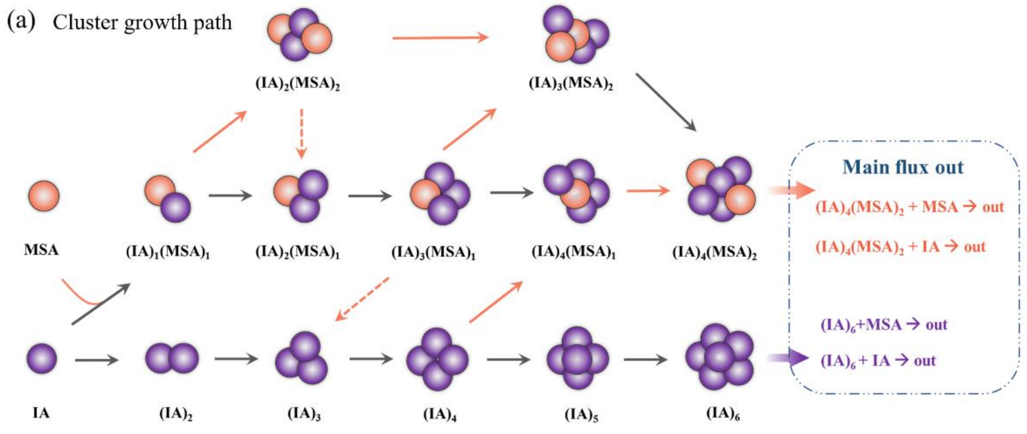
Response: Thanks for the helpful comments. As professionally suggested by the reviewer, we realized that the cluster formation rate of the IA-MSA mechanism simulated in Fig. 6 is at variance with the reported rate (Beck et al. 2020). Thus, the controversial Fig. 6 has been removed from the revised manuscript.

In addition, we have rewritten the corresponding results section (lines 240 – 273 in the revised manuscript) to focus more on the simulation results rather than any comparison with the field observation.

Comment 6. 5B: the authors cited iodic acid data from another study and assumed an MSA value of 1e7 cm-3. While I agree with the authors that there are not enough MSA measurements around the world, the authors should not overclaim their quantitative results because of their huge uncertainties. Additionally, many other species are not considered in the current mechanism which further deepens their discrepancy. The numbers in this plot are repeated in the Discussions and are presented as the key results in the Results part. This will be very misleading for future readers and should be all removed since there is no way for this study to get any remotely correct estimations.

Response: As professionally suggested by the reviewer, the simulations in Fig. 5B would indeed overestimate the impact of the IA-MSA mechanism on regions with MSA concentrations below 10^7 molecules cm^{-3}. We agree with you and have therefore removed Fig. 5B from the revised manuscript. Moreover, to present the results that the contribution of the IA-MSA mechanism is highly dependent on the MSA and IA concentration, the contribution of the different growth pathway varying with [MSA] (10^6 –10^7 molecules cm^{-3}) and [IA] (10^6 –10^8 molecules cm^{-3}) has been presented in the redrawn Fig. 5b, as follows:

(a) Cluster growth path



(b) Branch ratio of flux out (T = 278 K, CS = 2.0×10^{-3} s^{-1})

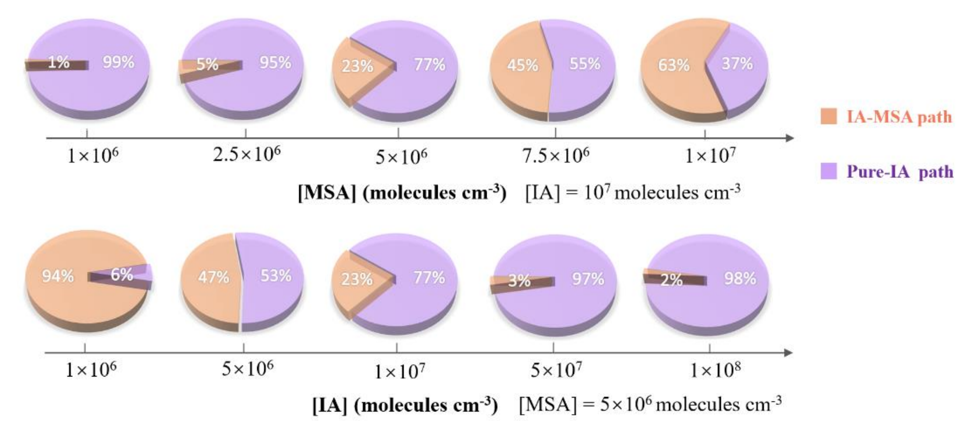


Figure 5. (a) Main cluster growth pathway of IA-MSA nucleating system at T = 278 K, CS = 2.0×10^{-3} s^{-1}, [IA] = 10^7 and [MSA] = 5×10^6 molecules cm^{-3}. The black and orange arrows refer to the pathways of colliding with IA and MSA, respectively, where the dashed arrows indicate the evaporation of MSA. **(b)** Branch ratio of IA-MSA (orange pie) and pure-IA (purple pie) growth pathway under varying [MSA] (10^6 – 10^7 molecules cm^{-3}) and [IA] (10^6 – 10^8 molecules cm^{-3}).

In addition, given that the results at [MSA] = 10^7 molecules cm^{-3} would overestimate the impact of the IA-MSA mechanism on some sites, the relevant statement has been removed from the revised manuscript.

Minor comments:

Line13-16: repeating message.

Response: Thanks for the reviewer's carefulness reading. The repeating message has been removed and integrated in lines 13 – 15 of revised manuscript as follows: "Moreover, MSA can promote IA clusters formation, particularly in cold marine regions with sparse IA and rich MSA. For the IA-MSA nucleation mechanism, in addition to self-nucleation of IA, the IA-MSA-involved clusters can also directly participate in the nucleation process."

Line24: ground based open ocean nucleation was not frequently measured but relatively rarely

Response: Thanks for the reviewer's suggestion, the "frequent" has been removed in the revised manuscript.

Line29: Marine NPF, particularly in remote areas, is more …

Response: Accordingly, the corresponding sentence has been changed to "Marine NPF, particularly in remote areas, is more affected by biological emissions compared to …" in line 26 of the revised manuscript.

Line40: The coastal NPF and low tide correlation was already established back to the early 2000s, e.g., O'Dowd, 2002.

Response: According to the reviewer's professional suggestion, the corresponding reference for O'Dowd, 2002 has been added in line 38 of the revised manuscript.

Line45: I do not find particle composition measurements of MSA and IA in Beck et al. 2020?

Response: In fact, in the study of Beck et al. 2020, the original statement is "At Ny-Ålesund, MSA, IA and nitrate/nitric acid are detected in some of the smallest clusters…" in the caption of Fig. 3 (Beck et al. 2020). Herein, according to the study (Beck et al., 2020), the "MSA and IA found in particles" has been corrected to "MSA and IA were detected in the smallest clusters" in line 45 of the revised manuscript.

Line47: mechanism should either be singular to plural.

Response: Thanks for the reviewer's suggestion. For consistency, the "mechanisms" has been changed to "mechanism".

Line53: in marine environments / in marine regions

Response: According to the reviewer's suggestion, the "at marine regions" has been changed to "in marine environments".

Line92: "t" to "the"

Response: "t" has been corrected to "the".

Line143: boundary layer to troposphere? Tropopause? Stratosphere? Clarify.

Fig. 2 and throughout this manuscript. Either use molecules cm-3 or pptv throughout this manuscript. Don't mix these two units.

Response: Thanks for the reviewer's professional comment. According to the previous study (Williamson et al., 2019), it is "boundary layer to free troposphere". As suggested by the reviewer, the unit (pptv) used in Fig. 2 has been changed to "molecules cm^{-3}" throughout this manuscript to ensure uniformity of units.

Line 152: explain beta and C. What are the subscriptions for beta and C in Fig 2C?

Fig.4: write the exact conditions (fixed values) for the three sets of simulations explicitly in the caption.

Response: Thanks for the reviewer's helpful suggestion. In Fig. 2C, the β and C represent the collision rate coefficient and monomer concentration, respectively. The subscription 'I' for $\beta_{\mathrm{I}}C_{\mathrm{I}}$ represents IA. Specially, β_{I} is the rate coefficient of cluster collision with IA monomer, and C_{I} is the concentration of IA monomer. These explanations have been added in the caption of Fig. 2 (lines 173 – 175 of the revised manuscript).

In addition, according to the reviewer's suggestion, the exact conditions (fixed values) for the three sets of simulations in Fig. 4 has been added in the corresponding caption (lines 204 - 205).

Line 204: it is not clear to me why reduced collision rates result in reduced *R* since all collision rates are reduced not just collisions between MSA and IA.

Response: Thanks! This is a valuable suggestion from the reader's perspective. I am sorry for our unclear presentation. Indeed, as the reviewer expertly analyzed, the decrease in temperature would reduce the intermolecular collision between IA and MSA, as well as IA and IA. The cause of *R* decay needs to be analyzed in terms of its definition as follows:

$$R = \frac{J_{\text{IA-MSA}}}{J_{\text{pure-IA}}} = \frac{J\,(\text{IA-MSA cluster + pure-IA cluster})}{J\,(\text{pure-IA cluster})}$$

The numerator term $J_{\text{IA-MSA}}$ is affected by the formation of both pure-IA clusters and IA-MSA clusters. While the denominator term $J_{\text{pure-IA}}$ is only affected by the formation of pure-IA clusters. Therefore, when the overall collision rate between IA and IA, as well as IA and MSA, is reduced due to the decrease in temperature, the numerator would be affected more than the denominator, which in turn leads to a reduced R. For the avoidance of confusion, the corresponding explanation has been added in lines 218 - 221 of the revised manuscript.

Line 221: these findings. I don't find MSA and IA detected in the particle phase in Beck et al. 2020. Could the authors specify where I can find the information there?

Response: Thanks for the reviewer's helpful suggestion. The restatement in the present manuscript is not accurate and should be "smallest clusters" rather than "particles". The corresponding statement in the work of Beck et al. 2020 is in the caption of Fig. 3 of "At Ny-Ålesund, MSA, IA and nitrate/nitric acid are detected in some of the smallest clusters…".

Line 233: the MSA concentrations are likely not always this high in these sites. Could the authors give a bit more reasoning in their choice of 1e7 cm-3?

Response: Thanks for your valuable comment. The reason is that according to the global distribution of MSA simulated by GEOS-Chem model (Chen et al., 2018), MSA concentrations can reach ppt level in most marine areas (Fig. 2(d) in Chen et al. 2018) including our studied regions. Hence, the MSA concentration employed in the simulations was chosen at the order of 10^7 cm^{-3}.

As professionally suggested by the reviewer, the simulations in Fig. 5b ([MSA] = 10^7 molecules cm^{-3}) would overestimate the impact of the IA-MSA mechanism on regions with concentrations below 10^7 molecules cm^{-3}. Considering the fact that the MSA concentration of 10^7 molecules cm^{-3} is not applicable for some marine regions, Fig. 5b has been removed from the main text.

Thanks again for the reviewers' professional and carefulness review. Accordingly, we have tried our best to improve the manuscript.

Sincerely Yours,

Prof. Xiuhui Zhang

Reference

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Appendix

Table 1. Cartesian coordinates of the IA dimer and trimer in the study of Rong et al., (2020) at the ωB97X-D/6-311++G(3df,3pd) + aug-cc-pVDZ-PP with ECP28MDF (for I) level of theory.

$(IA)_2$

Atoms	X	Y	Z
I	1.605061	-0.109352	-0.267819
O	2.025006	-1.477616	0.807848
O	0.441189	0.976342	0.730524
O	3.150010	1.046146	0.139201
H	3.315691	1.034733	1.088579
I	-1.605061	0.109352	0.267819
O	-0.441189	-0.976343	-0.730522
O	-2.025004	1.477616	-0.807848
O	-3.150011	-1.046145	-0.139204
H	-3.315691	-1.034732	-1.088582

$(IA)_3$

Atoms	X	Y	Z
I	1.545974	-1.434287	0.250626
O	2.033030	-0.476331	-1.207134
O	-0.211853	-1.929524	-0.055720
O	2.265224	-3.140916	-0.427111
H	2.210512	-3.143192	-1.389052
I	0.660386	1.874886	-0.216130
O	0.871637	0.519496	1.066009
O	-0.317464	3.088235	0.662250
O	2.415626	2.687111	0.162019
H	2.319948	3.214632	0.964029
I	-2.180521	-0.650525	-0.103596
O	-1.227504	0.715410	-0.917840
O	-2.135176	-0.387443	1.665104
O	-3.929096	0.185073	-0.450347
H	-4.015338	0.965703	0.109455