Dear Editor,

Thank you very much for your handling our manuscript "**Rapid Iodine Oxoacids Nucleation Enhanced by Dimethylamine in Broad Marine Regions**" (Ms No.: egusphere-2023-1774). These comments are all valuable and very helpful for revising and improving our paper. We have revised the manuscript carefully according to the reviewers' comments and the point-to-point responses are summarized below:

Reviewer #2:

The paper investigates the nucleation mechanism involving iodine oxoacids and dimethylamine (DMA). The authors suggest that DMA can enhance iodine oxoacid nucleation in ambient conditions and compare their results with ambient measurements. The mechanism explored by the authors is new; therefore, it may be worthy of publication in ACP. However, the authors fail to support their statement that their mechanism is important in broad marine regions. None of the three field observations supports their statements. In fact, two out of three observations strongly oppose the idea that HIO_3-HIO_2-DMA is an important mechanism, while the last one shows great ambiguity. A major revision is clearly needed. The authors should revise their manuscript so that ambient observations are correctly interpreted, and the implications of their results should be constrained properly before this manuscript can be accepted.

Response: We sincerely thank for the reviewer's careful review of our manuscript. The detailed responses are listed as follows.

Major Comment 1. While the subject is of great interest considering recent ambient and laboratory studies of iodine nucleation, I am not convinced by the authors that DMA is essential for pristine marine aerosol nucleation processes. The authors mention two field measurements from Ireland and Antarctica sites. However, my brief glance over these studies suggests that the evidence from these sites is clearly against their hypothesis, i.e., DMA is not important in the nucleation events observed there. In the Mace Head case (Sipila 2016), nucleation from iodine species was proposed by the authors to be the dominant mechanism there, and other nucleating species, e.g., SA, A, DMA, played a minor role there. In fact, the APi-TOF data did not even show the presence of A and DMA. Therefore, the authors' conclusion that iodine nucleation could not explain the observation is more likely a problem with the authors' calculations. Additionally, the authors' way of saying HIO3-HIO2 is not sufficient to explain the nucleation in Mace Head is rather flawed. The authors use an example case from Figure 1 of Sipila 2016. They adopted the corresponding nucleation rate as the bottom line of their range in Figure 5A. They further introduced an independent study (O'Dowd 2002) which reported an extraordinary 1e6 cm^{-3} s^{-1} nucleation rate as the upper limit in Figure 5A. However, it is rather clear from Sipila 2016, Figure 3, that the example case represented the upper limit of the field observation. Therefore, the nucleation rate range is much lower than what the authors presented in their Figure 5A. In conclusion, their conclusion that HIO_3-HIO_2-DMA is important in the Mace Head station is not reasonable. In the Aboa station, the authors (Jokinen 2018) also clearly suggested that the nucleation mechanism there was dominated by the SA-A mechanism instead of iodine-dominated mechanisms. The authors measured little to no DMA-containing clusters in the APi-TOF, and therefore, HIO_3-HIO_2-DMA is not an important mechanism there.

The authors propose that the reason why DMA was not measured in Mace Head and Aboa was that

the Nitrate-CIMS in these campaigns was not sensitive enough to measure DMA. However, they ignored the fact that the APi-TOF instrument was used in these campaigns and is extremely sensitive to DMA. The low (or no) DMA signals measured in these campaigns simply suggest that DMA was not important there, and the title and content of this paper exaggerate the implication of this manuscript. The proposed mechanism, at maximum, could be important for polluted coastal environments where clean marine air masses meet DMA-containing polluted air masses. The authors also used an observation from a Chinese coastal site to imply the HIO_3-HIO_2-DMA nucleation mechanism is important. However, there appear to be no iodine measurements in the mentioned study (Yu 2019). Therefore, their statement that HIO3-HIO2 nucleation mechanism is not sufficient while HIO_3-HIO_2-DMA is, is not reasonable since we do not know the exact nucleation mechanism there in the first place. I strongly suggest the authors restrict the scope of their study and try to avoid overstatements.

Response: Thanks for the professional suggestions. We agree with the reviewer's opinions that our previous statements in original Section 3.3 may have overstated the environmental significance of the HIO_3-HIO_2-DMA mechanism due to not considering the impact of other potential mechanisms. Hence, in the revised discussion of Section 3.3 (lines 268-325 of the revised Manuscript), we will discuss the environmental significance of the HIO_3-HIO_2-DMA system through the comparison with other potential mechanisms.

The reviewer raised a concern regarding the use of the API-TOF instrument at Mace Head and the absence of DMA in the detection. We agree with the reviewer's opinions that DMA may have insignificant contributions according to the field measurements. Therefore, in the revised discussion, we first proposed that HIO_3-HIO_2 nucleation may be the dominant mechanism of Mace Head region. Subsequently, based on model simulations or atmospheric observations, we assessed the potential influence of DMA and other nucleation precursors (H_2SO_4, MSA) on HIO_3-HIO_2 nucleation solely from a theoretical simulation perspective. Ultimately, we suggested that in pristine areas with abundant iodine sources, even if DMA and other precursors can reach high concentrations simulated in this study, their corresponding enhancements are limited.

The revised discussions are presented in lines 283-293 of the revised Manuscript as follows:

"Previous studies have demonstrated that local nucleation is primarily driven by high concentrations of HIO_3 (Sipilä et al. 2016). The simulated results showed that the J of HIO_3-HIO_2 system can reach up to levels of 10^4 cm^{-3} s^{-1}, which is consistent with the upper limit of formation rates reported in the field observation (Sipilä et al. 2016). Furthermore, we evaluated the potential impact of DMA and other precursors on the J of HIO_3-HIO_2 system based on the concentrations from model simulations or gas-phase measurements reported at Mace Head (Yu and Luo 2014; Sipilä et al. 2016). The results indicate that DMA can promote the J significantly only at lower iodine concentrations. As the increases of iodine oxoacids, the J of HIO_3-HIO_2-DMA system gradually approaches that of HIO_3-HIO_2 system, indicating a less significant enhancement by DMA. Similar patterns about the enhancement by H_2SO_4 and MSA can also be shown. This indicates that in primitive regions with abundant iodine sources, even if the precursors (DMA, H_2SO_4, and MSA) can reach the high concentrations used in the simulation in this study, their corresponding enhancement is limited."

Moreover, the reviewer raised a concern regarding the rapid nucleation rate between 10^4 and 10^6 cm^{-3} s^{-1} reported by O'Dowd et al (O'Dowd et al. 2002). We found that this research did not explicitly mention the contribution of iodine species. This implies that we may overestimate the iodine-driven nucleation rate of Mace Head when using the nucleation rate between 10^4 and 10^6 cm^{-3} s^{-1}. Hence, we

have removed the data of observational nucleation rates that are higher than 10^4 cm^{-3} s^{-1} in the revised Section 3.3.

The reviewer mentioned that the nucleation mechanism in Aboa region was dominated by the H_2SO_4-A mechanism instead of iodine-dominated mechanisms. We agree with the reviewer's viewpoint. In the Aboa region, the influence of HIO_3-HIO_2-DMA is relatively weaker compared with H_2SO_4-A nucleation. This has been explicitly addressed in our revised discussion, thus avoiding the exaggeration issues.

The revised discussions are as follows:

"Moreover, the simulated J of HIO_3-HIO_2-DMA system is also slightly lower than the formation rates (0.05 - 0.12 cm^{-3} s^{-1}) of ion-induced H_2SO_4-A system reported by field observations (Jokinen et al. 2018). Hence, due to the overall lower concentrations of iodine and amine components, the nucleation process is predominantly driven by H_2SO_4 molecules. This suggests that in regions with scarce iodine and amine sources, the contribution of the DMA-enhanced HIO_3-HIO_2 mechanism to the particle formation is limited."

Additionally, the reviewer raised a question that "The authors also used an observation from a Chinese coastal site to imply the HIO_3-HIO_2-DMA nucleation mechanism is important. However, there appear to be no iodine measurements in the mentioned study (Yu 2019)". We apologize for not comprehensive citing the reference, resulting in the misunderstanding of the iodine measurements in Zhejiang. In Zhejiang, Yu et al. observed NPF events driven by iodine species and detected HIO_3 and HIO_2 in the particle phase. The subsequent work collaborated by Xia et al. with Yu et al. (Xia et al, 2020) reported the nucleation rates observed in three different days of Zhejiang ($J_1 = 4.2 \times 10^2$, $J_2 = 5.3 \times 10^2$, $J_3 = 1.0 \times 10^3$ cm^{-3} s^{-1}), which serves as the data source for us to derive the conclusion. We have provided comprehensive references to the relevant studies in the revised Manuscript.

The revised discussions are presented in lines 268-271 of the revised Manuscript as follows:

"The Zhejiang region experiences the frequent NPF events, closely associated with a high-intensity iodine-driven nucleation process (Yu et al. 2019). However, our simulation results under the conditions of Zhejiang (Xia et al., 2020; Yu et al., 2019) indicate that relying solely on HIO_3-HIO_2 nucleation (red curve in Figure 1(a)) appears insufficient to explain the rapid formation rates of ambient environment (gray shaded area)."
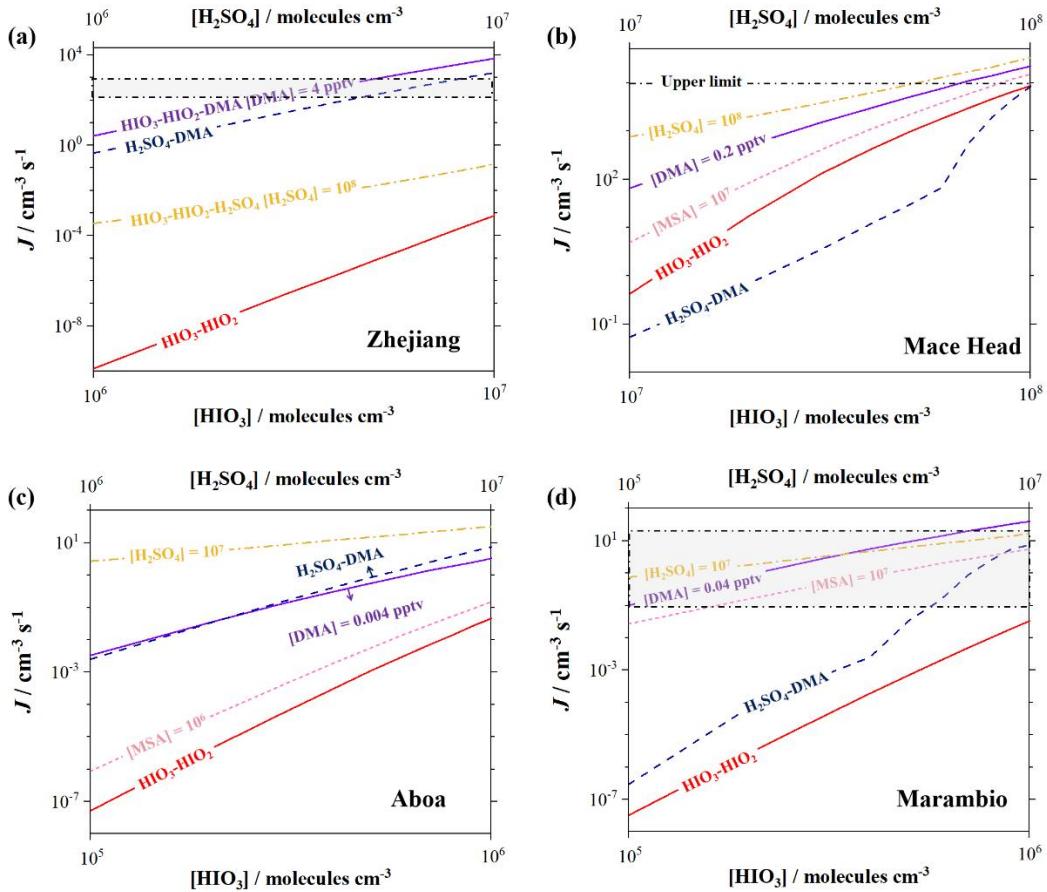
Figure 1. The cluster formation rates (J, cm^{-3} s^{-1}) of HIO$_3$-HIO$_2$ (red), HIO$_3$-HIO$_2$-DMA (purple), HIO$_3$-HIO$_2$-H$_2$SO$_4$(yellow), HIO$_3$-HIO$_2$-MSA (pink), and H$_2$SO$_4$-DMA (blue) systems under the simulated conditions of (a) Zhejiang: [HIO$_3$] = 10^6 - 10^7, [HIO$_2$] = 2×10^4 - 2×10^5, [H$_2$SO$_4$] = 10^6 - 10^8 molecules cm^{-3}, and [DMA] = 4 pptv, (b) Mace Head: [HIO$_3$] = 10^7 - 10^8, [HIO$_2$] = 2×10^5 - 2×10^6, [H$_2$SO$_4$] = 10^7 - 10^8, [MSA] = 10^7 molecules cm^{-3}, and [DMA] = 0.2 pptv, (c) Aboa: [HIO$_3$] = 10^5 - 10^6, [HIO$_2$] = 2×10^3 - 2×10^4, [H$_2$SO$_4$] = 10^6 - 10^7, [MSA] = 10^6 molecules cm^{-3}, and [DMA] = 0.004 pptv, and (d) Marambio: [HIO$_3$] = 10^5 - 10^6, [HIO$_2$] = 2×10^3 - 2×10^4, [H$_2$SO$_4$] = 10^5 - 10^7, [MSA] = 10^7 molecules cm^{-3}, and [DMA] = 0.04 pptv. The shaded area (grey) represents the actual nucleation rates observed locally.

Major Comment 2. Additionally, the scientific quality of this study is not high compared to the authors' previous studies and other similar studies. Most such studies will calculate the clusters with up to 8 monomers, while this study only calculated clusters with up to 5 monomers. How large are these clusters? Have they reached the critical size? I can understand the difficulty in getting clusters with 8 monomers for the three-component system, but 5 monomers are clearly not sufficient, and the authors are urged to discuss whether this can be improved.

Response: Thanks for the professional suggestions. According to the reviewer's suggestion, we expanded the entire system, including the ternary, binary, and unary parts, to six molecules through the same theoretical workflow to improve the scientific quality of this study. On the one hand, the collision rates of six-molecule clusters exceed the evaporation rates, tending to grow to seven or eight-molecule clusters. On the other hand, the size of six-molecule clusters reaches 1.2-1.3 nm. The sizes of these clusters are close to or have reached the size of critical nucleus (Zhang et al, 2012). Hence, the six-molecule system

can, to some extent, mitigate the issue of overestimating the system's formation rate due to the size of clusters being small. We have provided the structures of the newly added six-molecule clusters (Figure 2, which is the Figure S4 of the Supporting Information) below, along with the corresponding dynamic diameters of these clusters (Table 1). This information has also been added in Section S3 of the revised Supporting Information.
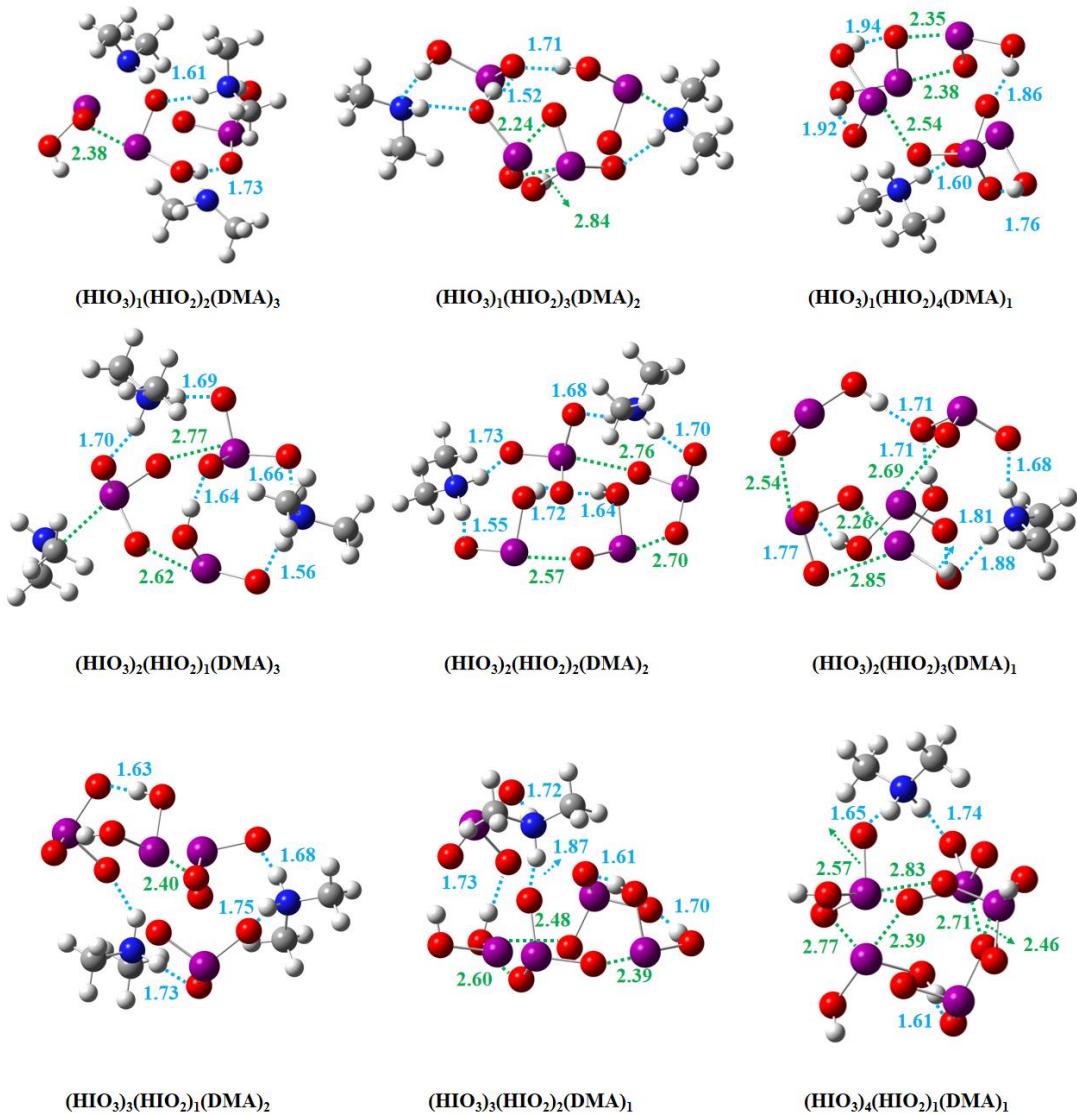
Figure 2. The most stable structures of six-molecules HIO_3-HIO_2-DMA clusters identified at the ωB97X-D/6-311++G(3df,3pd) (for H, C, N, and O atoms) + aug-cc-pVTZ-PP with ECP28MDF (for I atom) level of theory. The white, grey, blue, red, and purple balls represent the H, C, N, O, and I atoms, respectively. The hydrogen bonds and halogen bonds are shown in blue and green dashed lines, respectively. The values of bond lengths are given in Å.

Table 1. The mobility diameters (nm) of HIO_3-HIO_2-DMA clusters.

Clusters	Mobile diameters (nm)
$(HIO_3)_3(HIO_2)_1(DMA)_2$	1.28
$(HIO_3)_1(HIO_2)_3(DMA)_2$	1.26
$(HIO_3)_2(HIO_2)_2(DMA)_2$	1.27

$(HIO_3)_1(HIO_2)_2(DMA)_3$	1.27
$(HIO_3)_2(HIO_2)_1(DMA)_3$	1.28
$(HIO_3)_4(HIO_2)_1(DMA)_1$	1.27
$(HIO_3)_1(HIO_2)_4(DMA)_1$	1.26
$(HIO_3)_3(HIO_2)_2(DMA)_1$	1.27
$(HIO_3)_2(HIO_2)_3(DMA)_1$	1.27

Minor Comments.

Line 11: Journal name should not be presented. The reference style should be consistent throughout the manuscript.

Response: Thanks for the reviewer's careful check. We have changed the reference style in the first line of abstract.

Line 12-13: Is there clear evidence supporting the author's claim that the mentioned mechanism cannot explain NPF in broad marine regions? Or is it just that it may be insufficient to explain all cases? It sounds like quite a strong statement the authors are trying to make.

Response: Thanks for the professional suggestions. As suggested by the reviewer, only in certain coastal areas influenced by urban air masses, such as the Zhejiang region, former studies indicated that iodine species could drive nucleation processes, while the HIO_3-HIO_2 nucleation mechanism may not sufficiently explain the field observation results (Yu et al, 2019; Xia et al, 2020; Ma et al, 2023; Zu et al, 2024), prompting us to further consider the involvement of DMA. Consequently, the statements made in the abstract were exaggerated. Hence, we have adjusted the statement in lines 12-13 of the revised Manuscript as follow:

"However, HIO_3-HIO_2 nucleation may not effectively drive the observed rapid new particle formation (NPF) in certain coastal regions influenced by urban air masses."

Line 32: "are originated" → "originate"

Line 33: are thought

Line 34: coastal areas

Response: Thanks for the reviewer's suggestions. We have modified the above corresponding mistakes in the revised manuscript.

Line 40: How did Yu 2019 measure HIO_2? Could the authors confirm this?

Response: Yu and co-workers used a nano-microorifice uniform deposit impactor (nano-MOUDI; MSP Corp, Shoreview, MN) to collect the sample (10-18 nm) of aerosols or PM2.5 during NPF events. Subsequently, they detected IO_2^- signal, which may be the corresponding result of HIO_2 entering the mass spectrometry, in nano-MOUDI sample through an integrated mass spectrum of molecular ions (Yu et al, 2019).

Line 43-45: The statement seems different than the definitive statement in the abstract. Could the authors clarify: is iodine nucleation not explaining broad marine NPF or is it sometimes insufficient? Do we know where iodine is important and when they are not? Is there field evidence?

Response: Thanks for the reviewer's suggestions. We have realized the exaggeration issue raised in the abstract. To the best of our knowledge, iodine oxoacids nucleation is significant in clean marine

environments, supported by field evidence from such as the Mace Head region (Sipila et al, 2016). However, it may be sometimes insufficient in polluted coastal regions (Ma et al, 2023), such as Zhejiang regions (Yu et al, 2019; Zu et al, 2024). In order to provide an accurate and reasonable introduction to the background of iodine oxoacids nucleation, we have adjusted the statement in lines 62-66 of the revised Manuscript as follow:

"In short, the iodine oxoacids (HIO_x, $x = 2, 3$) can drive rapid particle formation, and they may play an important role in marine and polar NPF process. However, in certain coastal areas influenced by urban air masses, such as the Zhejiang region, former studies indicated that iodine species could drive nucleation processes, while the HIO_3-HIO_2 nucleation mechanism may not sufficiently explain the field observation results (Yu et al. 2019; Ma et al. 2023; Zu et al. 2024; Xia et al. 2020), which indicates that other nucleation precursors may be involved."

Line 136: The references to tables S1-S4 appear to be missing. Should they be added somewhere? Or otherwise, reorder the tables to make S5 the S1.

Response: Thanks for the reviewer's suggestions. We listed the detailed collision and total evaporation frequencies of the HIO_3-HIO_2-DMA system at all simulated temperatures and condensation sinks in Tables S1-S4. The references to Tables S1-S4 are in lines 151-152 of the revised manuscript.

Lines 150-159: Interesting observations here related to the proton transfer. Since iodine is a halogen, could the authors also comment on the role of halogen bonding in cluster formation? Is the halogen bonding important? Previous studies that the authors have cited (e.g., Zhang 2022) seem to point to a strong involvement of halogen bonds. Therefore, I encourage the authors to have a detailed session discussing the halogen bonds and a further session comparing the halogen bonds and hydrogen bonds, which are more important? Additionally, the authors suggest that DMA competes for a proton from HIO2, giving the impression that they are separate things while in reality, it appears to be synergistic effects (Figure 4), instead of competition (i.e., adding DMA would never reduce the nucleation rates).

Response: Thanks for the professional suggestions. We have added the discussion about the influence of proton transfer, halogen bonds, and hydrogen bonds on the stability of clusters.

"It is worth noting that, in terms of the stability (evaluated by ΔG) of clusters, the proton transfer plays a crucial role. Taking two 3-molecule clusters, $(HIO_3)_2(HIO_2)_1$ (ΔG = -30.05 kcal mol^{-1}) and $(HIO_3)_1(HIO_2)_1(DMA)_1$ (ΔG = -35.19 kcal mol^{-1}), as an example, we observed that the $(HIO_3)_2(HIO_2)_1$ cluster with higher energy is formed through three XBs and one HB, while $(HIO_3)_1(HIO_2)_1(DMA)_1$ cluster with lower energy is formed through three HBs after proton transfer. Additionally, when the number of proton transfers is the same, clusters with more halogen bonds formed by HIO_2 generally have lower ΔG, which can be seen from $(HIO_3)_1(HIO_2)_2(DMA)_1$ (ΔG = -50.07 kcal mol^{-1}, formed by three XBs and two HBs) and $(HIO_2)_4$ (ΔG = -58.08 kcal mol^{-1}, formed by four XBs) clusters. Similar results can also be observed in other ternary clusters."

Additionally, the reviewer pointed out that the relationship between DMA and HIO_2 appears to be to synergistic effects rather than competition. This is a valuable suggestion. Due to HIO_2 molecules being amphiprotic rather than basic, we have removed the original statement of the competition between two bases (DMA and HIO_2) during nucleation. As suggested by the reviewer, we have added the statements in lines 274-277 of the revised Manuscript as follows:

"Notably, the increase of the concentrations of HIO_2 and DMA can both enhance the J to match the field observations, indicating that HIO_2 and DMA molecules exhibit a synergistic effect on HIO_3

nucleation, which may have significant contributions to NPF events in the polluted coastal areas where marine iodine species intersect with high concentrations of DMA."

Lines 161-176: What is the nucleation rate simulated by this study for Figure 3? Does it agree with the field observation they refer to? Analyzing the branching ratio has to be based on a reasonable agreement between the field observation and their simulation.

Response: The simulated cluster formation rate of original Figure 3 is 2.1×10^4 cm^{-3} s^{-1}, which is consistent with the reported results of 10^4 cm^{-3} s^{-1} in the corresponding field observations (Sipila et al, 2016).

Lines 173-176: The discussion here about DMA detection makes no sense. Even if the nitrate-CIMS did not measure DMA, the APi-TOF deployed in the mentioned paper should have captured DMA. Have Sipila et al. (2016) measured DMA with their APi-TOF? Note, APi-TOF would capture DMA if there were over 5e5 molec. cm-3 levels of DMA.

Response: Thanks for the valuable suggestions. Our previous discussion was not comprehensive enough and appeared to be contradictory to the field observation results. Therefore, we have removed the discussion about DMA detection and modified the statements of the role of DMA in the revised Section 3.3 to avoid exaggerating the implication of this manuscript.

Lines 255-257: Are there iodine measurements at the mentioned site? Where did the author get the concentrations? If there were no measurements of iodine species, how did the author derive the conclusion that the nucleation rates are too high to be explained?

Response: In Zhejiang, Yu et al. observed NPF events driven by iodine species and detected HIO$_3$ and HIO$_2$ in the particle phase. The subsequent work collaborated by Xia et al. with Yu et al. (Xia et al, 2020) reported the nucleation rates observed in three different days of Zhejiang ($J_1 = 4.2 \times 10^2$, $J_2 = 5.3 \times 10^2$, $J_3 = 1.0 \times 10^3$ cm^{-3} s^{-1}), which serves as the data source for us to derive the conclusion. The gas-phase concentration of HIO$_3$ is set based on the data provided by Xia et al (Xia et al, 2020). Due to the lack of the data of HIO$_2$, we estimated the gas-phase concentration of HIO$_2$ based on the ratio of [HIO$_3$] to [HIO$_2$] in the field observations (Sipila et al. 2016) and CLOUD experiment (He et al. 2021), which showed that the ratio of [HIO$_3$] to [HIO$_2$] is about 20 to 100 depending on the concentration of iodine vapor. The ratio presented in the main text was 50 according to the former field observations in Mace Head (Sipila et al. 2016) and the other results obtained from two different ratios (20 and 100) of [HIO$_3$] and [HIO$_2$] can be seen from the Section S5 of the Supporting Information.

Figure 5: When the authors calculate the HIO$_3$-HIO$_2$ rates, which they suggest citing from another study, did the author use the same size of clusters as the HIO$_3$-HIO$_2$-DMA system (5 monomers in this system)? This will also influence the nucleation rates of these systems.

Response: Yes, we used the same size of HIO$_3$-HIO$_2$ clusters (5 monomers before, 6 monomers now) with the HIO$_3$-HIO$_2$-DMA clusters, aiming to eliminate errors caused by the size of clusters when calculating cluster formation rates.

Figure 5A: The field observation range is very misleading. How is it possible that the nucleation rate in the ambient reaches 1e6? Did the O'Dowd 2002 paper measure iodine species? If not, what is the acid and nucleation rate range in the mentioned Sipila 2016 paper? The authors should use a dataset that has

both acid and nucleation rate measurements instead of assembling different datasets to prove their points. The authors mentioned one case from the Sipila 2016 with a 1e4 nucleation rate and 1e8 HIO_3; it appears it agrees with their high-end HIO_3-HIO_2 simulation and low-end of HIO_3-HIO_2-DMA simulation. How can they conclude from this?

Response: Thanks for the professional suggestions. O'Dowd et al (O'Dowd et al. 2002) measured a rapid nucleation rate between 10^4 and 10^6 cm^{-3} s^{-1}, but did not explicitly mention the contribution of iodine species. This implies that we may overestimate the iodine-driven nucleation rate of Mace Head. Hence, we have removed the data of observational nucleation rates that are higher than 10^4 cm^{-3} s^{-1}.

As the reviewer mentioned, our simulation showed that the observational nucleation rates agree with the upper limit of HIO_3-HIO_2 system and the lower limit of HIO_3-HIO_2-DMA system. This indicates that the enhancing effect of DMA in such scenarios may be overestimated due to the overestimation of [DMA] in the simulation compared to the actual situation. Moreover, in pristine areas with abundant iodine sources, even if DMA and other precursors can reach high concentrations simulated in this study, their corresponding enhancements are limited. We have added the discussion in lines 291-294 of the revised Manuscript.

Figure 5: What are the shades in the figure? It is very confusing since I do not find an explanation for this. Please clarify. Additionally, the authors should compare their HIO_3-HIO_2 nucleation rate with the experimental work (He 2021?) to see whether their rates are reasonable before deriving any meaningful conclusions.

Response: Thanks for the professional suggestions. The shades in original Figure 5 represent the range of nucleation rates covered by the ternary HIO_3-HIO_2-DMA and binary HIO_3-HIO_2 systems. The upper limit corresponds to the scenario where the precursor concentrations are simultaneously at their maximum values, while the lower limit corresponds to the scenario where the precursor concentrations are simultaneously at their minimum values. To avoid the difficulty for readers caused by excessive information in the figure, we have eliminated the original presentation method that combined upper and lower limits with shaded areas.

As suggested by the reviewer, we have conducted the comparison between experimental results and the simulated formation rates of the expanded six-molecule HIO_3-HIO_2 system, and the corresponding results are presented in Figure 3. The simulation was performed under the same conditions with CLOUD experiment (He et al, 2021) ([HIO_3] = 10^6 - 10^8 molecules cm^{-3}, [HIO_2] = 2×10^4 - 2×10^6 molecules cm^{-3}, T = 283 (red), 263 (blue) K, and CS = 2.2×10^{-3} s^{-1}). The circles in Figure 3 represent the experimental results, while the squares represent the theoretical results. As shown in Figure 3, at lower T, the simulated formation rates are in good agreement with the experimental results. At higher T, the simulated results exhibit a similar growth trend to the experimental results, although the simulated rates are generally about one order of magnitude higher. This indicates that the theoretical method we employed exhibits a high level of accuracy, particularly at lower T. As a result, the predictive results obtained through these simulations can be reliable. We have added the discussion in Section S2 of the revised Supporting Information.
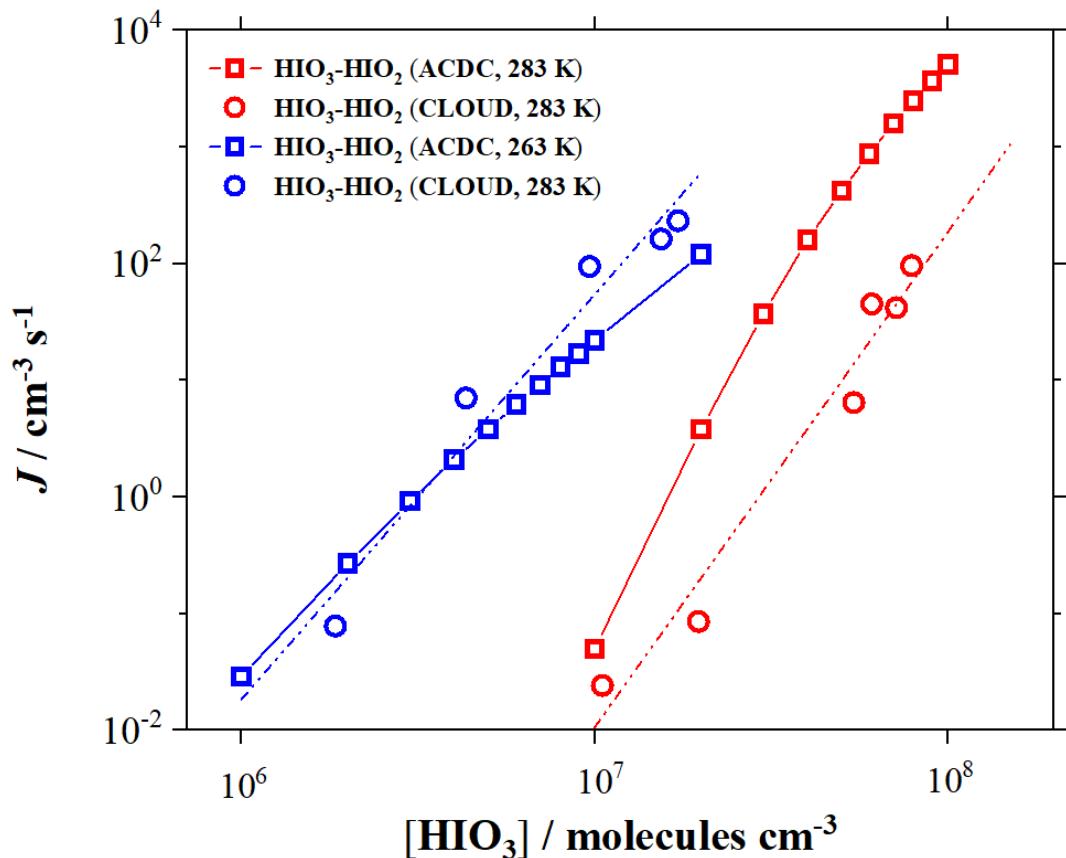
Figure 3. The cluster formation rates (J, cm^{-3} s^{-1}) of HIO$_3$-HIO$_2$ system obtained from experiment (CLOUD) and theoretical (ACDC) results. The simulation was performed under the conditions of [HIO$_3$] = 10^6 - 10^8, [HIO$_2$] = 2×10^4 - 2×10^6, T = 283 (red), 263 (blue) K, and CS = 2.2×10^{-3} s^{-1}. The circles represent the experimental results, while the squares represent the theoretical results.

Lines 264-272: The original paper from Jokinen et al. 2018 (10.1126/sciadv.aat9744) clearly suggests that SA-NH$_3$ was the nucleation mechanism there and did not measure DMA in the APi-TOF (Fig 2 of that study). This study's calculation has no relevance to the conditions there. Also, please cite the mentioned paper for the discussion here.

Response: Thanks for the professional suggestions. According to the reviewer's suggestions, we are aware that our statement may overlook the contributions of some more significant mechanisms. The simulated J of HIO$_3$-HIO$_2$-DMA system is lower than the formation rates (0.05 - 0.12 cm^{-3} s^{-1}) of ion-induced H$_2$SO$_4$-A system reported by field observations (Jokinen et al. 2018). Hence, due to the overall lower concentrations of iodine and amine components, the nucleation process is predominantly driven by H$_2$SO$_4$-A system clearly. This suggests that in regions with scarce iodine and amine sources, the contribution of the DMA-enhanced HIO$_3$-HIO$_2$ mechanism to the particle formation is limited. We have added the discussion in Section 3.3 of the revised manuscript and cited the mentioned paper for the discussion.

We have tried our best to improve the manuscript and the main changes have been highlighted in yellow in the revised manuscript.

Sincerely,

Xiuhui Zhang

Key Laboratory of Cluster Science Ministry of Education of China

School of Chemistry and Chemical Engineering

Beijing Institute of Technology

Beijing 100081, P.R. China

Email: zhangxiuhui@bit.edu.cn

Reference:

Almeida, J., S. Schobesberger, A. Kurten, I. K. Ortega, O. Kupiainen-Maatta, A. P. Praplan, A. Adamov, A. Amorim, F. Bianchi, M. Breitenlechner, A. David, J. Dommen, N. M. Donahue, A. Downard, E. Dunne, J. Duplissy, S. Ehrhart, R. C. Flagan, A. Franchin, R. Guida, J. Hakala, A. Hansel, M. Heinritzi, H. Henschel, T. Jokinen, H. Junninen, M. Kajos, J. Kangasluoma, H. Keskinen, A. Kupc, T. Kurten, A. N. Kvashin, A. Laaksonen, K. Lehtipalo, M. Leiminger, J. Leppa, V. Loukonen, V. Makhmutov, S. Mathot, M. J. McGrath, T. Nieminen, T. Olenius, A. Onnela, T. Petaja, F. Riccobono, I. Riipinen, M. Rissanen, L. Rondo, T. Ruuskanen, F. D. Santos, N. Sarnela, S. Schallhart, R. Schnitzhofer, J. H. Seinfeld, M. Simon, M. Sipila, Y. Stozhkov, F. Stratmann, A. Tome, J. Trostl, G. Tsagkogeorgas, P. Vaattovaara, Y. Viisanen, A. Virtanen, A. Vrtala, P. E. Wagner, E. Weingartner, H. Wex, C. Williamson, D. Wimmer, P. L. Ye, T. Yli-Juuti, K. S. Carslaw, M. Kulmala, J. Curtius, U. Baltensperger, D. R. Worsnop, H. Vehkamaki, and J. Kirkby. 2013. 'Molecular understanding of sulphuric acid-amine particle nucleation in the atmosphere', *Nature*, 502: 359-+.

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