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 #1:

Zu et al. investigate the influence of dimethylamine (DMA) on HIO_3-HIO_2 cluster formation using quantum chemical methods and atmospheric cluster dynamics simulations. This is an excellent and natural extension of the previous studies on iodine oxoacids by the same group. A funneling approach is used to identify the cluster configurations lowest in free energy. The final cluster structures are calculated using density functional theory (ωB97X-D/6-311++G (3df,3pd)) and the single point energy is calculated using RI-CC2/aug-cc-pVTZ calculations. The calculated thermochemistry is applied as input to the atmospheric cluster dynamics code (ACDC) to simulate new particle formation rates in various marine regions (Mace Head, Zhejiang and Aboa). The main finding is that the HIO_3-HIO_2 cluster formation rates does not correspond to the NPF observations, but DMA enhance the cluster formation rate by several orders of magnitude, thereby increasing the agreement between the modelling and the observations. I only have some minor quarrels with the applied methodology. The cluster formation simulations are very sensitive to the quantum chemical data, so some sensitivity runs should be performed to see how robust the conclusions are to the applied level of theory. In addition, the influence of other nucleation precursors (SA, MSA, multiple bases, etc.) should be further discussed in the manuscript to emphasize that the HIO_3-HIO_2-DMA mechanism is not the only explanation for the gap between theory and measurements. However, the authors do not need to carry out the actual calculations, just discuss the potential importance of other species. Overall, I believe the chosen systems and current study at hand is an interesting addition to the literature. The manuscript is easy to follow and the topic falls within the scope of ACP.

Response: Thanks for the reviewer's valuable comments, and we have revised our manuscript accordingly.

Major Comment 1. Introduction: I am missing some introduction to what we, in general, know about cluster formation from previous quantum chemical studies. Please put the current study into context of the whole field and not just iodine studies. What vapours have previously been studied and found important and what are the main findings of previous work?

Response: Thanks for the reviewer's professional and valuable suggestions. According to the reviewer's suggestion, we have added the introduction about the potential nucleation vapours and the relevant main findings that have been previously found important in the Introduction of the revised manuscript. The revised statements highlighted in yellow in the revised manuscript are as follows:

"Atmospheric aerosols, the intricate suspension formed by fine particles in the atmosphere, exert far-reaching influences on global climate (Haywood and Boucher 2000; Murphy and Ravishankara 2018; Lee et al. 2019), radiation balance (Haywood and Boucher 2000), and human health (Pope and Dockery 2006; Gong et al. 2014). A significant source of atmospheric aerosols on the world-wide scale is the new particle formation (NPF), encompassing nucleation and subsequent growth (Zhang 2010). The nucleation

is identified as the key process of NPF events. Therefore, understanding the nucleation mechanism is vital for comprehending the behavior of aerosols (Zhang et al. 2012; Kalivitis et al. 2015).

Sulfuric acid (H_2SO_4) is considered a crucial precursor for the nucleation in continental regions. However, under actual atmospheric conditions, the H_2SO_4-Water (W) binary nucleation is far from sufficient to explain the observed strong NPF events (Elm 2021a). Therefore, additional components are essential for nucleation. Specifically, abundant atmospheric bases, such as ammonia (A) and alkylamines [methylamine (MA), dimethylamine (DMA), trimethylamine (TMA), and ethylenediamine (EDA)] (Elm et al., 2017; Kirkby et al., 2011; Kurtén et al., 2008; Weber et al., 1996; Xie and Elm, 2021), are recognized as important stabilizers for H_2SO_4-driven nucleation. Computational work by Kurtén et al. (Kurtén et al. 2008) and experimental studies by Almeida et al. (Almeida et al. 2013) indicated that, despite the lower atmospheric concentration of DMA (few pptv.), the promoting effect of DMA on the H_2SO_4-driven nucleation rate is several orders of magnitude higher than that of A. Moreover, nitric acid (NA) is also a potential precursor in the nucleation process (Knattrup and Elm, 2022). Liu et al. (Liu et al. 2018; Liu et al. 2021) showed the significant promoting effect of NA on the classical H_2SO_4-A and H_2SO_4-DMA nucleation mechanisms by theoretical methods. Additionally, Wang et al. (Wang et al. 2020) studied the nucleation process of mixed vapor of NA and A under atmospheric conditions in the CLOUD chamber at the European Organization for Nuclear Research, and subsequently considered the promoting effect of H_2SO_4 on NA-A system (Wang et al. 2022).

Given the vast expanses of the ocean, marine aerosols play an indispensable role in the global aerosol system (O'Dowd and de Leeuw 2007). Over the oceans, H_2SO_4 and methanesulfonic acid (MSA), the oxidation products of dimethylsulfide (DMS), is considered as important nucleation precursors over the oceans (Elm 2021b). Theoretical calculations (Chen, Li, Wang, Liu, et al. 2020; Chen, Li, Wang, Luo, et al. 2020; Shen et al. 2020) have indicated that basic precursors such as A, MA, and DMA can promote MSA-based nucleation processes. However, a recent study on aerosol acidity showed that global models substantially overestimate the concentrations of A, especially in polar regions (Nault et al. 2021). This may slightly weaken the influence of H_2SO_4-based and MSA-based nucleation in marine atmosphere (He et al. 2023)."

Major Comment 2. Line 38: How high are the HIO_3 and HIO_2 concentrations measured at Mace Head? Please state the concentrations here as well.

Response: According to the measurement in Mace Head (Sipila 2016), the concentrations of HIO_3 and HIO_2 are up to10^8 and 2×10^6 molecules cm^{-3}, respectively. We have added the concentrations of HIO_3 and HIO_2 in the revised *Introduction* section.

Major Comment 3. Line 73-74: "Firstly, the ABCluster program (Zhang and Dolg, 2015) was performed to generate up to 120000 initial isomer structures using the artificial bee algorithm." Where does the number 120000 come from? Is this the ABCluster population value (SN) times the number of generations? Some more information on the ABCluster parameters would be a useful addition.

Response: Thanks for the valuable suggestion. We have listed the detailed information on the ABCluster parameters in Section S1 of the revised Supporting Information. As the reviewer pointed out, the number of initial isomer structures (120000) is obtained by multiplying the population size (SN = 300) by the maximal generations (g_{max} = 400).

Information	Parameters
Population size (*SN*)	300
Maximal cycle number (g_{max})	400
Scout limit (g_{limit})	3
Estimated cluster size (Å)	3 - 13
Number of local minima (LMs) to be saved	1000

Major Comment 4. Line 75-91: I am missing some comments on the accuracy of the applied configurational sampling methodology and the applied quantum chemical methods.

Only saving 1000 local minima from the ABCluster search sounds a bit low. How certain are the authors that they have located the global minimum? As the UFF forcefield cannot handle bond breaking a more diverse pool of clusters is usually desirable. This is usually done by performing ABCluster runs with ionic monomers as well (see Kubečka et al., https://doi.org/10.1021/acs.jpca.9b03853). Only selecting the lowest 100 cluster configurations based on PM7 could lead to the global minimum cluster being missed (see Kurfman et al., https://doi.org/10.1021/acs.jpca.1c00872). Could the authors comment on this aspect?

Response: Thanks for the reviewer's professional comments. We apologize for missing to show some information about the configurational sampling methodology. We had utilized ion monomers for sampling during the cluster configuration search and had manually constructed some potential stable clusters with multiple hydrogen/halogen bonding sites based on the chemical intuition. The relevant statements have been added in lines 97-102 and 111-112 of the revised Manuscript as follows:

"Due to the inability of the UFF force field to effectively handle bond breaking issues, we utilized ion monomers for sampling during the configuration search of ternary clusters (Kubecka et al., 2019). From the most stable configuration of binary HIO_2-DMA clusters (Figure S4), it showed that the proton transfer from HIO_2 to DMA is forbidden in all cluster except for $(HIO_2)_1(DMA)_1$, indicating that this process is difficult to occur spontaneously. Hence, we only considered the ion monomers where HIO_3 donates protons or HIO_2/DMA accept protons."

"Additionally, we manually constructed some potential stable clusters with multiple hydrogen/halogen bonding sites based on the chemical intuition."

Considering the computational cost, we saved 1000 local minima from the ABCluster search and selected the lowest 100 cluster configurations based on PM7, which may lead to the global minimum cluster being missed. The relevant statements have been added in lines 108-112 of the revised Manuscript as follows:

"Notably, even though we try our best to search for the global minimum configurations of clusters considering the computational cost, saving 1000 local minima from the ABCluster search and selecting the lowest 100 cluster configurations based on PM7 may lead to the global minimum cluster being missed (Kurfman, Odbadrakh and Shields, 2021)."

How accurate are the RI-CC2/aug-cc-pVTZ calculations? The leading terms in the CC2 equations are MP2-like, at the cost of N5. Hence, you could get accurate DLPNO-CCSD(T)/aug-cc-pVTZ calculations at essentially the same computational cost. The authors admit that previous agreement with experiments is caused by random cancellation of errors. Our previous work has shown (Schmitz et al., https://doi.org/10.1021/acsomega.0c00436) that RI-CC2/aug-cc-pVTZ is severely overbinding, i.e. yielding too negative binding energies, thus leading to too stable clusters. Where is the remaining error cancellation coming from? All missing effects in the simulations (hydration, ionic effects, anharmonicity, potential inadequate sampling, ect …) would make the clusters more stable and hence make the current results agree less with experiments.

Response: Thanks for the reviewer's professional comments. We agree with the reviewer's opinions that using RI-CC2/aug-cc-pVTZ method may lead to too stable clusters and added the statement in Section S2 of the revised Supporting Information as follows:

"Compared to DLPNO-CCSD(T) results, the RI-CC2 results overestimate the ΔG of clusters to some extent, leading to higher cluster formation rates in ACDC simulations (Schmitz and Elm, 2020)."

We have added the discussion on the cancellation of errors in Section S2 of the revised Supporting Information as follows:

"The cancellation is coming from two types of error: the overestimation of binding energy obtained at RI-CC2 method compared with other methods (Schmitz and Elm, 2020) and the underestimation of cluster formation rates simulated by ACDC compared with the experimental results, possibly due to the lack of consideration of hydration, ionic effects, and inadequate sampling, etc."

ACDC simulations are extremely sensitive to the applied QC methods, and we can essentially get whatever we want by tweaking the level of theory. Hence, some further information on how we can trust the results is warranted. How robust are the conclusions to the applied level of theory? I suggest the authors test if the ωB97X-D/6-311++G(3df,3pd) calculations without RI-CC2 are yielding the same conclusions. Hence, this would not require additional calculations, but vastly improve the reliability of the study.

Response: We conducted additional calculations for the entire ternary system (n = 6) at the DLPNO-CCSD(T)/aug-cc-pVTZ level of theory. Furthermore, we compared the simulated cluster formation rates obtained at ωB97X-D/6-311++G(3df,3pd), RI-CC2/aug-cc-pVTZ, and DLPNO-CCSD(T)/aug-cc-pVTZ level of theory with the CLOUD experiment results (Figure 1, which is the Figure S1 of the Supporting Information) to assess the reliability of results obtained at RI-CC2/aug-cc-pVTZ method. As shown in Figure 1, without performing single-point energy correction, the cluster formation rates simulated at ωB97X-D/6-311++G(3df,3pd) level of theory (the diamonds) are significantly lower than the experimental results (the circles). Therefore, we chose to perform the single-point correction to obtain simulated cluster formation rates that agree more with the experimental results. Compared to the results obtained at the DLPNO-CCSD(T)/aug-cc-pVTZ level of theory, employing the single-point energy at RI-CC2/aug-cc-pVTZ level of theory leads to a slight overestimation of the simulated cluster formation rates (Schmitz and Elm, 2020). This overestimation is more pronounced at lower concentrations of iodine oxoacids, reaching approximately two orders of magnitude. It is worth noting that, compared to the results obtained at the DLPNO-CCSD(T)/aug-cc-pVTZ level of theory, the simulated cluster formation rates at the RI-CC2/aug-cc-pVTZ level of theory appear to be more in line with the CLOUD experiment, especially at lower temperatures. The simulation results in Figure 1 demonstrate that ACDC simulations are highly sensitive to the quantum chemical methods. Considering that the RI-CC2 method agree the

most with experimental results while saving computational resources, we chose the RI-CC2 method for single-point correction. We have added these discussions in lines 117-122 of the revised Manuscript and Section S2 of the revised Supporting Information.

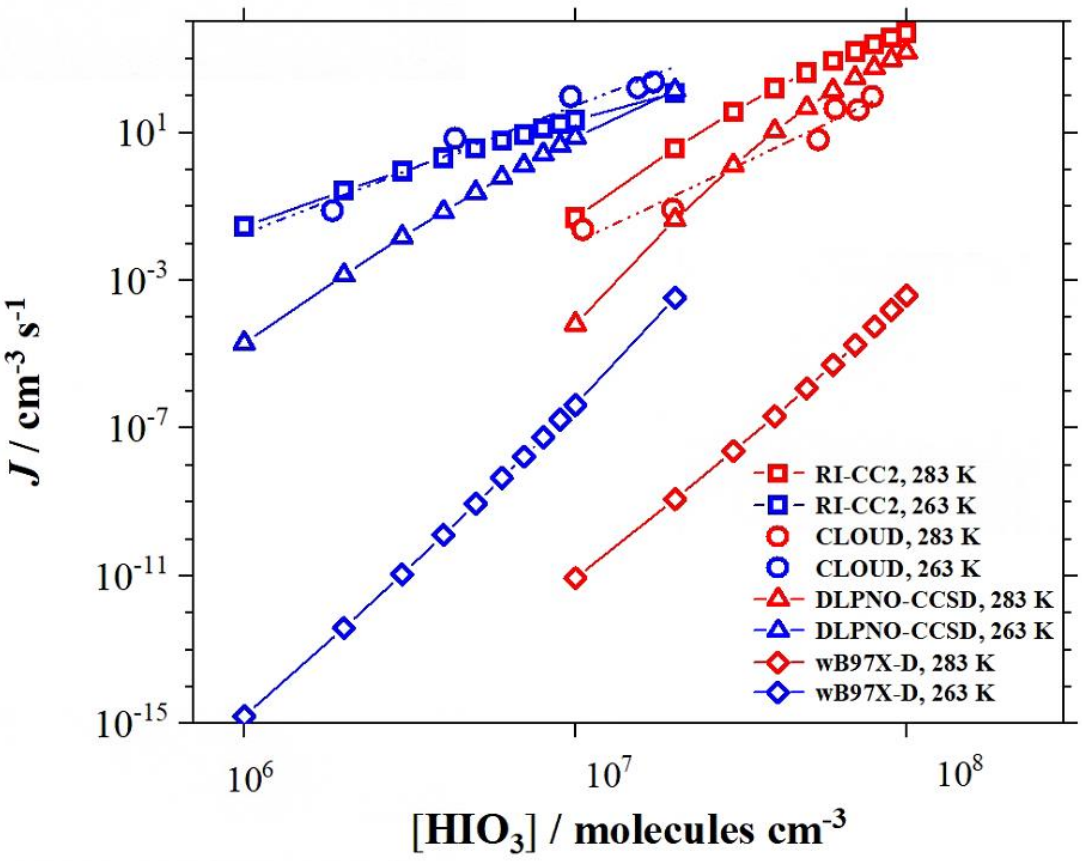

Figure 1. The cluster formation rates (J, cm^{-3} s^{-1}) of HIO_3-HIO_2 system obtained from experiment (CLOUD) and theoretical results at RI-CC2 and DLPNO-CCSD level of theory. The simulation was performed under the same conditions of [HIO_3] = 10^6 - 10^8, [HIO_2] = 2×10^4 - 2×10^6, T = 283 (red) / 263 (blue) K, and CS = 2.2×10^{-3} s^{-1}. The circles represent the experimental results, the squares represent the theoretical results at RI-CC2/aug-cc-pVTZ (for H, C, N and O atoms) + aug-cc-pVTZ-PP with ECP28MDF (for I atom) level of theory, the triangles represent the theoretical results at DLPNO-CCSD(T)/aug-cc-pVTZ (for H, C, N and O atoms) + aug-cc-pVTZ-PP with ECP28MDF (for I atom) level of theory, and the diamonds represent the theoretical results at 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.

Major Comment 5. Line 113-115: Please also mention the boundary conditions here in the main text. Setting the boundary clusters as clusters consisting of only six molecules could lead to artefacts in the ACDC simulations, thereby yielding too high cluster formation rates (see Besel et al., https://doi.org/10.1021/acs.jpca.0c03984). For some acid-base systems the "critical cluster" is already found within the initial 2x2 cluster system, however this depends on the given base (https://doi.org/10.1021/acs.jpca.3c00068). Overall, I am not entirely convinced that the boundary conditions are adequate in the current study and might yield too high cluster formation rates. Please elaborate on this aspect.

Response: Thanks for the reviewer's valuable comments. We have expanded the size of the entire nucleation system to six molecules, and accordingly, the conditions of boundary clusters have also been altered. The new boundary clusters in this work were set to be the clusters formed by the combination of kinetically stable clusters ($\beta c/\Sigma\gamma > 1$) with six molecules ($n = 6$) and the corresponding monomers at the concentration c. In addition, for acid-base clusters such as HIO_3-DMA, "4×4" clusters were also set to be boundary clusters. As shown in Figure 2 (Figure S2 of the Supporting Information), the small size of boundary clusters may result in an overestimation of the cluster formation rates by 1-2 orders of magnitude. The larger simulated systems and boundary clusters can address this issue, providing simulation results that align more closely with CLOUD experiment results (Figure 1). We have added the new settings of boundary clusters in Section S3 of the revised Supporting Information.

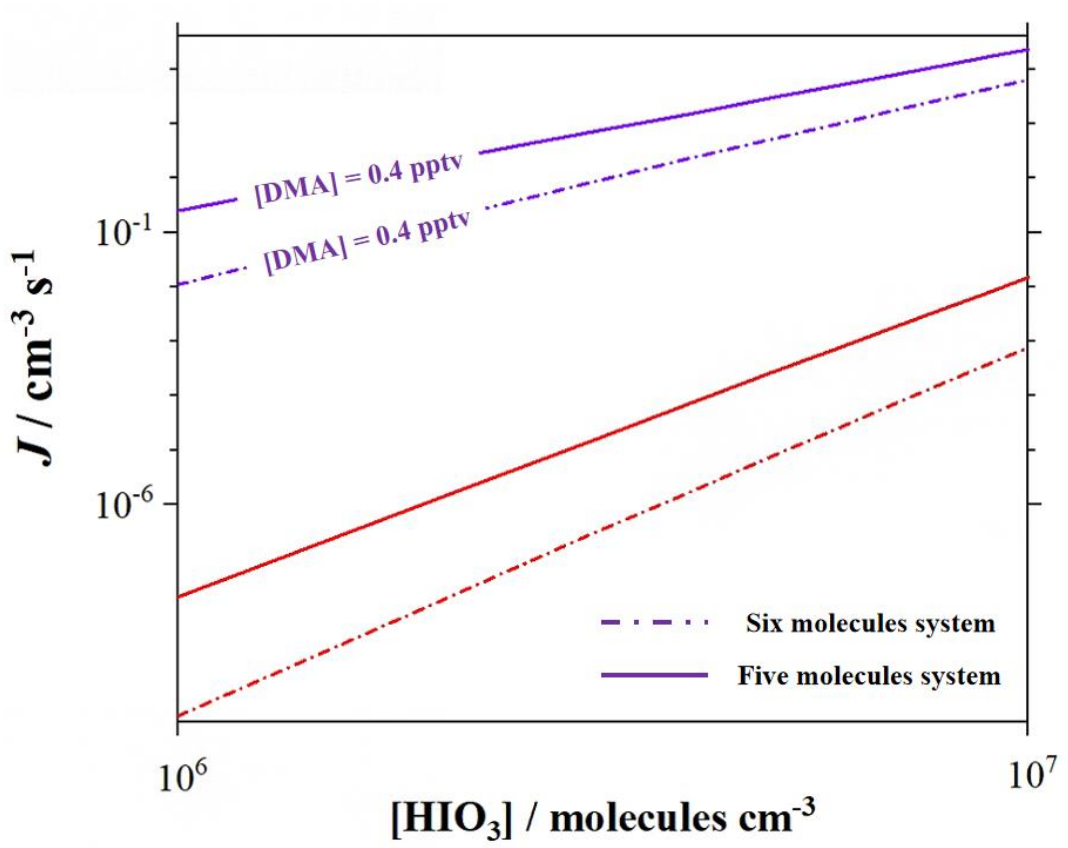

Figure 2. The simulated cluster formation rates (J, cm^{-3} s^{-1}) of HIO_3-HIO_2 (red) and HIO_3-HIO_2-DMA (purple) systems under the simulated conditions of $[HIO_3] = 10^6$ - 10^7, $[HIO_2] = 2\times10^4$ - 2×10^5, [DMA] = 0.4 pptv, $T = 293$ K, and CS = 10^{-2} s^{-1}. The solid curves represent the simulated J for the five-molecule system, while the dashed curves represent the simulated J for the six-molecule system.

Major Comment 6. Line 118-132: I do not see what Figure 1 is contributing with to the present study. It is simple chemistry to identify the donor/acceptor groups in molecules. No need for electrostatic potential maps for doing this. Please remove this part.

Response: Thanks for the reviewer's valuable comments. According to the reviewer's suggestion that ESP analysis and original Figure 1 contributes quite less to the present study, we have removed the discussion of electrostatic potential prediction and Figure 1 in Section 3.1 to Section S4 of the revised Supporting Information.

Major Comment 7. Line 137: "… , which proves the prediction of electrostatic potential analysis" Please remove.

Response: We have removed the statement that "… , which proves the prediction of electrostatic potential analysis" in Section 3.1.

Major Comment 8. Line 143: "… and HIO2 can also act as a stabilizing base in the neutral nucleation process of HIO3-HIO2" and "Hence, the participation of DMA may potentially lead to a competition between two basic molecules for proton transfer reaction." I am not completely comfortable calling iodous acid a base (in the Brøndsted-Lowry acid-base formalism). I agree that the HIO2 show peculiar proton transfer dynamics, but I would refrain from calling it a base. I guess it is technically amphoteric.

Response: Thanks for the reviewer's professional comments. Calling HIO_2 as "basic molecule" or "base" may lead readers to misinterpret the property of HIO_2. Considering the fact that HIO_2 can accept the proton from iodic acid (HIO_3), we have adjusted the description of HIO_2 from "basic molecule" to "amphoteric molecule exhibiting base-like behavior in the system". Additionally, we have removed the related statements of "multiple bases" and "competition of bases" in Sections 3.2 and 3.3.

Major Comment 9. Line 153-154: "… which possesses relatively stronger basicity than HIO2 in the process of proton transfer." What is the gas-phase basicity and pKa values of DMA and HIO2 respectively?

Response: According to previous research, the gas-phase basicity of DMA is 896.5 kJ mol^{-1} (Yang et al. *J. Phys. Chem. Lett.* 2018, 9, 18, 5647–5652) and the acid dissociation constant (pK_a) of HIO_2 is 6.0 (Schmitz, G. *Int. J. Chem. Kinet.* 2008, 40, 647−652.). We have added this information in lines 167-169 of the revised manuscript.

Major Comment 10. Line 180: What is the absolute cluster formation rate for the conditions given in Figure 3? How does this compare to the conventional SA-DMA system?

Response: Thanks for the professional suggestions. We have added the absolute cluster formation rate (J, cm^{-3} s^{-1}) of HIO_3-HIO_2-DMA system in Table 1 under the conditions of the original Figure 3. However, considering that the original Figure 3 was not representative enough to highlight the environmental significance of the HIO_3-HIO_2-DMA system, we adjusted the simulation conditions in Figure 3 to Zhejiang, the eastern coast of China, which serves as an intersection of high [DMA] pollution air masses and marine iodine air masses (Zhang et al., 2023; Zhu et al., 2019).

Former research (Zhang et al. 2022) has made some discussion about the nucleation abilities of HIO_3-HIO_2, H_2SO_4-A, and H_2SO_4-DMA system. The results show that the J of HIO_3-HIO_2 system is at least an order of magnitude higher than that of the H_2SO_4-DMA system, and significantly higher than that of the H_2SO_4-A system. Although the concentration distribution of HIO_3 and particularly HIO_2, is not as widespread as the concentration distribution of H_2SO_4 and DMA molecules, especially in urban areas, this is still sufficient to demonstrate the strong nucleation capability of the HIO_3-HIO_2 system. Moreover, our study revealed that in a polluted environment, the cluster formation rate of HIO_3-HIO_2 system can be further enhanced by air pollutant DMA, especially at high [DMA] derived from urban pollution. Therefore, through indirect comparison with H_2SO_4-A and H_2SO_4-DMA systems, we think the J of the HIO_3-HIO_2-DMA system might be higher than conventional H_2SO_4-DMA system in coastal areas with high [DMA] pollution. We have added the discussion in the revised Supporting Information.

Table 1. The absolute cluster formation rate (J, cm^{-3} s^{-1}) of HIO_3-HIO_2-DMA system under the conditions corresponding to Figure 3.

Conditions	J (HIO_3-HIO_2-DMA)
$[HIO_3] = 10^8$ molecules cm^{-3} $[HIO_2] = 2\times10^6$ molecules cm^{-3} [DMA] = 0.02 pptv T = 287 K, CS = 2×10^{-3} s^{-1}	2.1×10^4 cm^{-3} s^{-1}

Major Comment 11. Line 201-202: "… and the HIO3-HIO2-DMA ternary nucleation is critical in explaining the missing sources of new particles especially in the place where the concentrations of HIO2 and DMA are similar." I would be careful stating that the HIO3-HIO2-DMA mechanism is the "critical" missing link. It might contribute, but other mechanisms might also be important. Some discussion on the potential other species (SA, MSA, multibases, water, etc.) that might contribute to cluster formation in marine environments would be a welcome addition to the manuscript.

Response: Thanks for the professional suggestions. According to the reviewer's suggestion, we have added a comparison with other potential nucleation mechanism (HIO_3-HIO_2-H_2SO_4, HIO_3-HIO_2-MSA, and H_2SO_4-DMA) in Section 3.3 of the revised Manuscript as follows:

"To evaluate the influence of DMA on HIO_3-HIO_2 system and the environmental significance of HIO_3-HIO_2-DMA system, we compared the J of HIO_3-HIO_2 (red), HIO_3-HIO_2-DMA (purple), HIO_3-HIO_2-H_2SO_4 (yellow), HIO_3-HIO_2-MSA (pink), and H_2SO_4-DMA (blue) systems under the simulated conditions of typical marine and polar regions. The results of HIO_3-HIO_2-H_2SO_4 (Zu et al. 2024) and HIO_3-HIO_2-MSA systems (https://doi.org/10.5194/egusphere-2023-2084) were obtained from former studies at the same level of theory with HIO_3-HIO_2-DMA system. The configurations of H_2SO_4-DMA clusters were obtained from the Atmospheric Cluster Database (ACDB) (Elm 2019). Subsequently, geometric optimizations and frequency calculations were performed at the same (ωB97X-D/6-311++G(3df,3pd)) level of theory with HIO_3-HIO_2-DMA system. Notably, we believe that the influence of other mechanisms (HIO_3-HIO_2-A, etc.) is widespread and significant. However, due to the lack of available data, other mechanisms are not discussed in this study and will be considered in further studies."

The detailed comparison with other potential mechanisms is presented in lines 268-325 of the 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 4(a)) appears insufficient to explain the rapid formation rates of ambient environment (gray shaded area). It is noteworthy that the NPF events in the local area is found to be influenced not only by marine components but also by urban pollutants (Zhu et al. 2019; Liu et al. 2022). During polluted periods, the emission capacity of gas-phase DMA is exceptionally strong, and high concentrations of DMA can further enhance the J of HIO_3-HIO_2, resulting in a significant enhancement of up to 10^8 times at [DMA] = 4 pptv. 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. Moreover, the urban pollution also leads to an abundant concentration of gas-phase H_2SO_4. This renders the impact of the HIO_3-HIO_2-H_2SO_4 and H_2SO_4-DMA mechanisms non-negligible. As shown in Figure 3(a), the simulated *J* of H_2SO_4-DMA and HIO_3-HIO_2-H_2SO_4 systems (blue dashed line) is about one or four orders of magnitude lower than that of HIO_3-HIO_2-DMA system, respectively. This indirectly indicates that the HIO_3-HIO_2 system promoted by DMA possesses remarkable nucleation ability and might make unexpected contributions in specific regions, thereby providing an explanation for some missing fluxes of particles in the atmosphere.

In contrast, the results from the Mace Head region present a different situation. 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. The primary nucleation mechanism is likely to be the HIO_3-HIO_2 mechanism, which is supported by the on-site measurements of the components of nanoparticles (Sipilä et al. 2016).

Recent research has shown that the ice-influenced ocean may also be important sources of DMA (Dall'Osto et al. 2017; Dall'Osto et al. 2019). Hence, we also evaluate the environmental significance of HIO_3-HIO_2-DMA system in the ice-covered polar regions. As shown in Figure 3(c), we performed a simulation under the conditions of Aboa station. The simulation results indicate that HIO_3-HIO_2-H_2SO_4 and H_2SO_4-DMA are more efficient nucleation mechanisms than HIO_3-HIO_2-DMA system in the local area. 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. In contrast, in the Marambio region with relatively abundant DMA and scarce iodine oxoacids, HIO_3-HIO_2-DMA system may also have significant contributions [Figure 3(d)]. The [HIO_3] used in the simulation is still about an order of magnitude lower than [H_2SO_4] and [MSA]. However, [DMA] is about an order of magnitude higher than that in the Aboa region. In this case, the *J* of HIO_3-HIO_2 system is significantly enhanced by the relatively abundant DMA. Compared to the acidic components such as H_2SO_4 and MSA, DMA elevates the *J* of the HIO_3-HIO_2 system to the range of 10^{-1} to 10^1 cm^{-3} s^{-1}, matching the field observation results, while [DMA] is only one-tenth of [H_2SO_4] and [MSA]. This indicates that, considering nucleation ability, the enhancement effect of DMA on the HIO_3-HIO_2 system may be superior to H_2SO_4 and MSA, which we speculate that is likely related to the base stabilization effect of DMA within acidic clusters. Additionally, our results demonstrate that the

nucleation ability of HIO_3-HIO_2-DMA is stronger than that of H_2SO_4-DMA. This means that the HIO_3-HIO_2-DMA ternary mechanism may be an important contributor to iodine-containing particles, especially in regions where there are sufficient iodine and amine sources.

In summary, the results show that HIO_3-HIO_2-DMA ternary nucleation mechanism may have significant contributions to the formation of nanoparticles, especially in those regions with abundant iodine and amine sources. This previously overlooked mechanism may provide an explanation for some missing fluxes of atmospheric iodine particles. Moreover, the observed formation rates in the field can result from multiple rapid nucleation systems or may be solely attributed to a specific system, depending significantly on the variations of precursor concentrations in different regions. Hence, the simulation of scenarios where various components synergistically participate is needed in the future study to accurately assess the roles of different components such as HIO_3, HIO_2, H_2SO_4, MSA, A, and DMA. This may contribute to a fundamental understanding of atmospheric particle formation, providing a comprehensive insight into the entire evolution process of atmospheric aerosols."

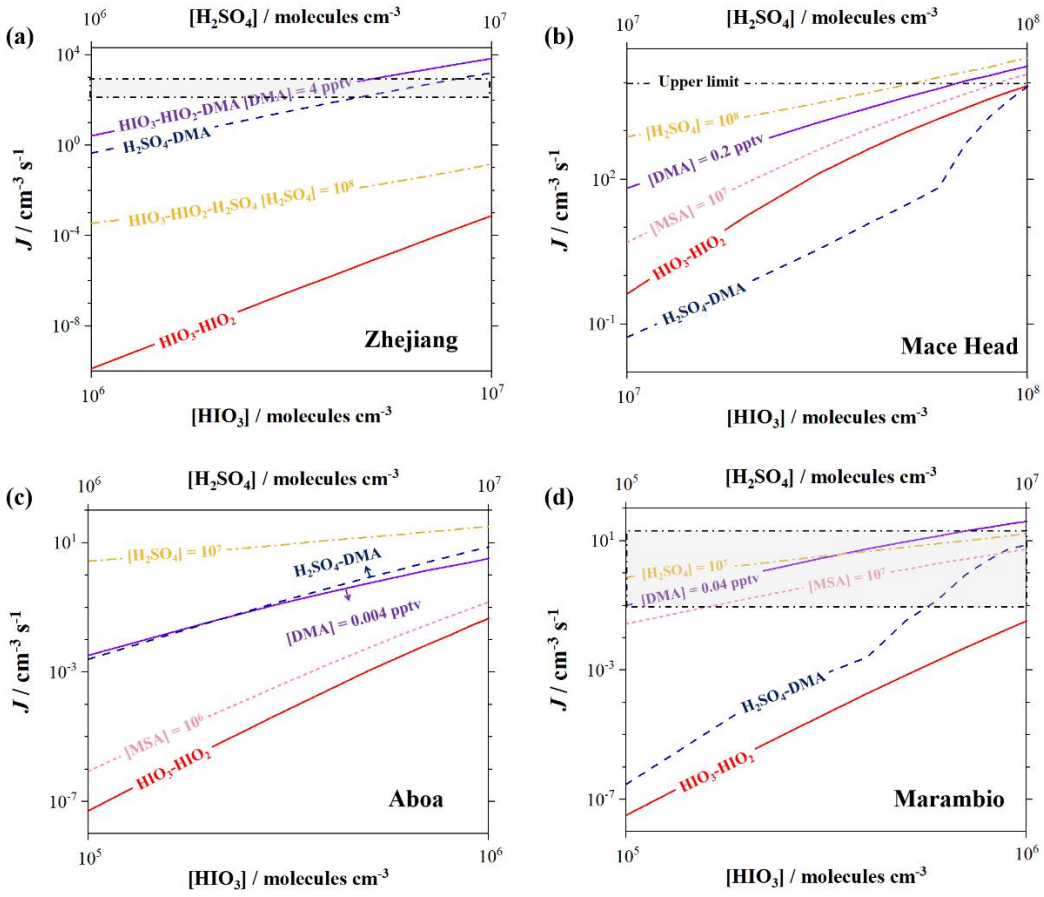

Figure 3. The cluster formation rates (J, cm^{-3} s^{-1}) of HIO_3-HIO_2 (red), HIO_3-HIO_2-DMA (purple), HIO_3-HIO_2-H_2SO_4(yellow), HIO_3-HIO_2-MSA (pink), and H_2SO_4-DMA (blue) systems under the simulated conditions of (a) Zhejiang: $[HIO_3] = 10^6$ - 10^7, $[HIO_2] = 2\times10^4$ - 2×10^5, $[H_2SO_4] = 10^6$ - 10^8 molecules cm^{-3}, and [DMA] = 4 pptv, (b) Mace Head: $[HIO_3] = 10^7$ - 10^8, $[HIO_2] = 2\times10^5$ - 2×10^6, $[H_2SO_4] = 10^7$ - 10^8, [MSA] = 10^7 molecules cm^{-3}, and [DMA] = 0.2 pptv, (c) Aboa: $[HIO_3] = 10^5$ - 10^6, $[HIO_2] = 2\times10^3$ - 2×10^4, $[H_2SO_4] = 10^6$ - 10^7, [MSA] = 10^6 molecules cm^{-3}, and [DMA] = 0.004 pptv, and (d) Marambio: $[HIO_3] = 10^5$ - 10^6, $[HIO_2] = 2\times10^3$ - 2×10^4, $[H_2SO_4] = 10^5$ - 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 12. Line 204-205: "This is the first time that a combined influence of multiple bases has been discovered in the nucleation process driven by HIO_3, …" Again, I am not comfortable calling HIO_3-HIO_2-DMA a "two"-base systems. Please remove this sentence.

Response: According to the reviewer's valuable suggestion, we have removed the related statements of "multiple bases" and "competition of bases" in Section 3.2.

Major Comment 13. Line 231: "The *J* of HIO_3-HIO_2-DMA and HIO_3-HIO_2 in Mace Head are shown …" To avoid misinterpreting this as actual measurements at Mace Head, please specify that these are simulations of conditions corresponding to Mace Head.

Response: Thanks for the reviewer's suggestions. We have revised the corresponding statement to "The simulated *J* of HIO_3-HIO_2-DMA …" in Section 3.3 of the revised manuscript.

Major Comment 14. Section 3.3 – cluster formation rates: I understand the rationale behind Figure 5. However, I believe it would be worth to more clearly state in the text that this is just ***a mechanism***, potentially one out of many, that increases the rates such that they match the observations. The measurements are essentially the sum of all possible nucleation pathways. This means that all possible nucleating precursor vapours contribute to the measured *J*-value. For instance, how would the results be influenced if your simulations included water, sulfuric acid or base synergy such as having both ammonia and DMA present? All these factors would yield clusters lower in free energy, increasing the cluster formation rates, and hence push the agreement further away from the observations.

Response: Thanks for the professional suggestions. Considering that other mechanisms might also be important, we realize that labeling the ternary mechanism of HIO_3-HIO_2-DMA system as "critical" may overestimate its environmental significance. As suggested by the reviewer, we have introduced other potential nucleation mechanisms (HIO_3-HIO_2-H_2SO_4, HIO_3-HIO_2-MSA, and H_2SO_4-DMA) for comparison to comprehensively evaluate the environmental significance of the HIO_3-HIO_2-DMA system in Section 3.3 of the revised Manuscript. The results showed that the observed formation rates in the field can result from multiple rapid nucleation systems or may be solely attributed to a specific system, depending significantly on the variations of precursor concentrations in different regions. We have added this statement in lines 317-319 of the revised Manuscript.

Previous studies have indicated that H_2O contributes less significantly to strong acid-base systems dominated by proton transfer, such as the H_2SO_4-DMA system. The HIO_3-HIO_2-DMA system exhibits nucleation ability comparable to the H_2SO_4-DMA system, with proton transfer processes in almost all ternary clusters. Hence, we speculate that the contribution of H_2O to the HIO_3-HIO_2-DMA system is relatively weak similar with the contribution of H_2O to the H_2SO_4-DMA system. We have added the discussion in lines 184-191 of the revised Manuscript.

As for the influence of H_2SO_4 and multibases, these components could exhibit synergistic or competitive effects on atmospheric nucleation. The nucleation process in real atmosphere is complex. Hence, the simulation of scenarios where various components participate simultaneously is needed in the future study to accurately assess the roles of different components such as H_2SO_4, MSA, A, and DMA in the iodine oxoacids nucleation. We have added these statements in lines 319-322 of the revised Manuscript.

Major Comment 15. Line 293-295: "However, considering the conditions of humidity in oceanic atmosphere and the complexity of marine NPF events, future research should investigate the role of water molecules and other crucial precursors to establish a comprehensive multi-component nucleation mechanism in the marine atmosphere." I believe this is a very important point, that should be mentioned and discussed much earlier and not just as an outline.

Response: Thanks for the professional suggestions. Based on the existing data, we have considered the impacts of other potential components (H_2SO_4, MSA) on the HIO_3-HIO_2 system in the revised Section 3.3, along with a comparison between the HIO_3-HIO_2-DMA and classical H_2SO_4-DMA nucleation mechanisms. Moreover, we have discussed the potential impact of H_2O molecules on HIO_3-HIO_2-DMA system in the revised Section 3.1. Through the discussion in Sections 3.1 and 3.3, we emphasize the urgent need for an atmospheric model that can simultaneously consider various rapid nucleation mechanisms and we intend to address this issue in our future research.

Minor Comments. Line 12: derive -> drive

Line 13: broad marine regions -> various? marine regions

Line 51: Quelever -> Quéléver

Line 86: Kuerten should be Kürten.

Line 176: Kurten -> Kurtén. Please check the spelling of all Finnish authors in the references as many umlauts are missing.

Response: Thanks for the reviewer's careful check. We have revised the corresponding mistakes and apologized for not spelling the name of the Finnish authors in the correct ways.

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)."

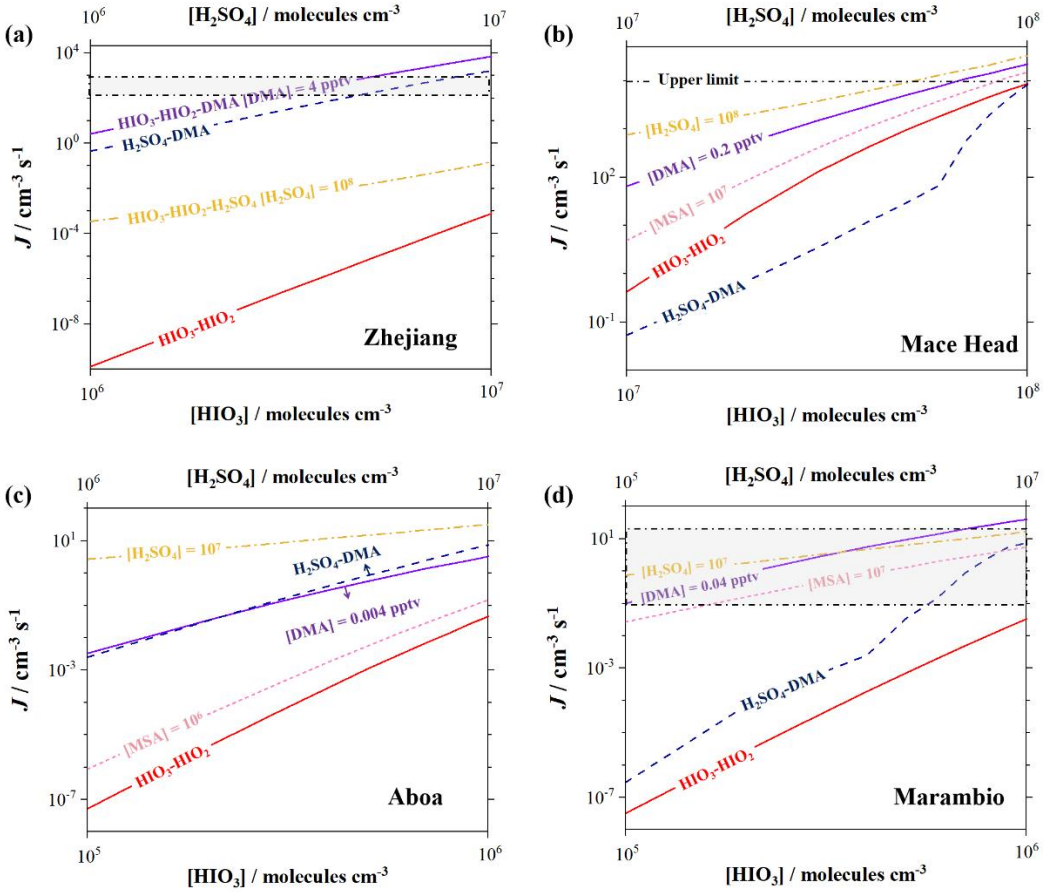

Figure 1. The cluster formation rates (J, cm^{-3} s^{-1}) of HIO_3-HIO_2 (red), HIO_3-HIO_2-DMA (purple), HIO_3-HIO_2-H_2SO_4(yellow), HIO_3-HIO_2-MSA (pink), and H_2SO_4-DMA (blue) systems under the simulated conditions of (a) Zhejiang: $[HIO_3] = 10^6$ - 10^7, $[HIO_2] = 2\times10^4$ - 2×10^5, $[H_2SO_4] = 10^6$ - 10^8 molecules cm^{-3}, and [DMA] = 4 pptv, (b) Mace Head: $[HIO_3] = 10^7$ - 10^8, $[HIO_2] = 2\times10^5$ - 2×10^6, $[H_2SO_4] = 10^7$ - 10^8, $[MSA] = 10^7$ molecules cm^{-3}, and [DMA] = 0.2 pptv, (c) Aboa: $[HIO_3] = 10^5$ - 10^6, $[HIO_2] = 2\times10^3$ - 2×10^4, $[H_2SO_4] = 10^6$ - 10^7, $[MSA] = 10^6$ molecules cm^{-3}, and [DMA] = 0.004 pptv, and (d) Marambio: $[HIO_3] = 10^5$ - 10^6, $[HIO_2] = 2\times10^3$ - 2×10^4, $[H_2SO_4] = 10^5$ - 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.

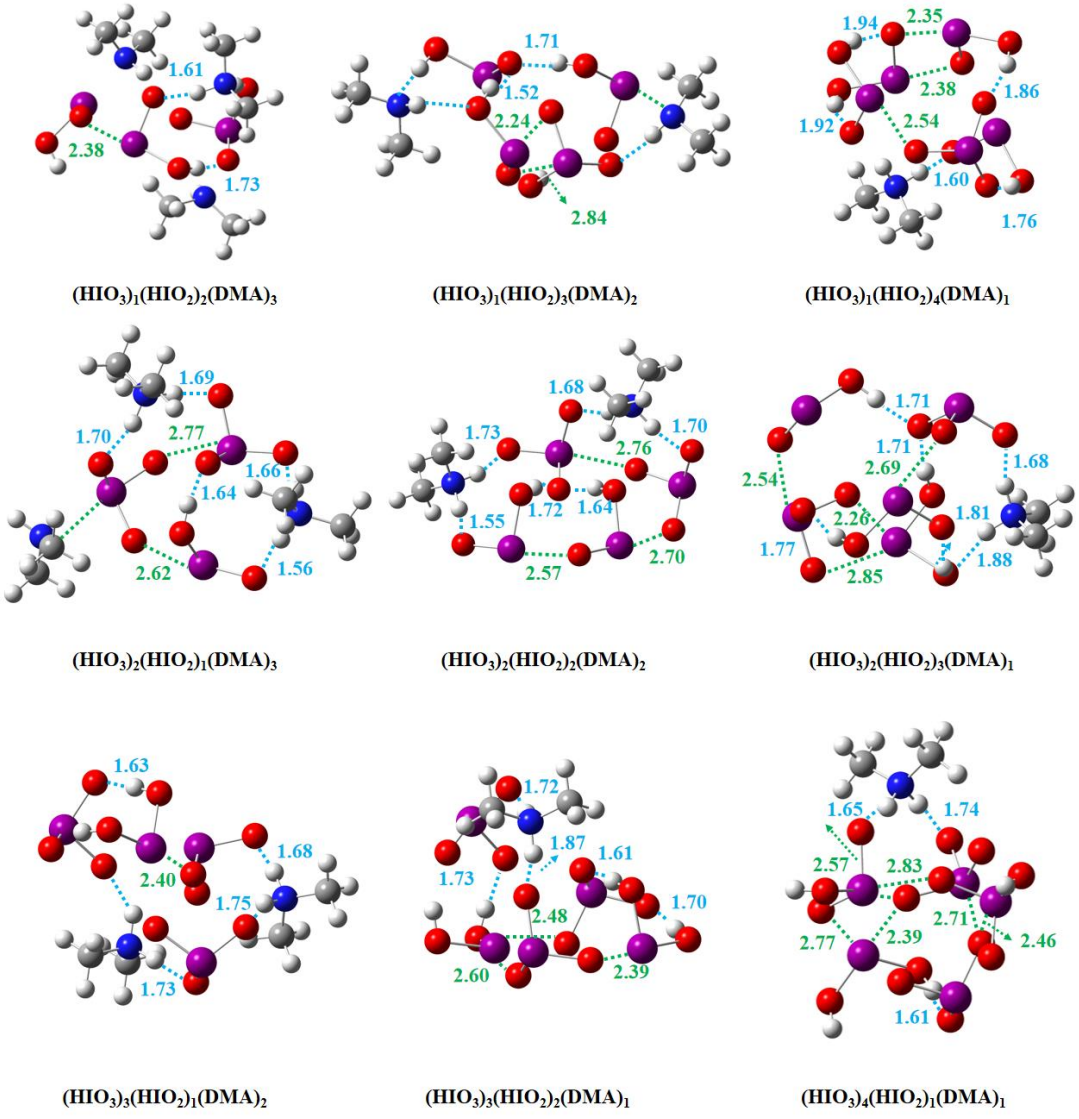

Figure 2. The most stable structures of six-molecules HIO_3-HIO_2-DMA clusters identified at the ωB97X-D/6-311++G(3df,3pd) (for H, C, N, and O atoms) + aug-cc-pVTZ-PP with ECP28MDF (for I atom) level of theory. The white, grey, blue, red, and purple balls represent the H, C, N, O, and I atoms, respectively. 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.

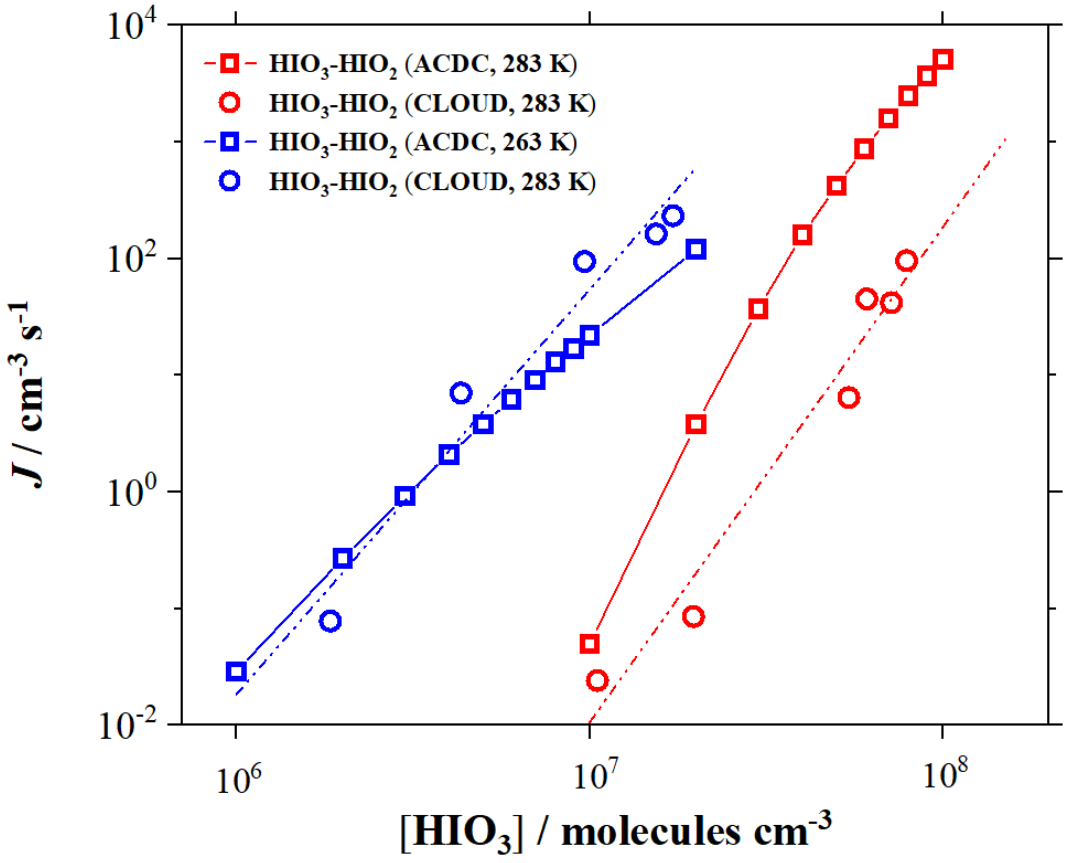

Figure 3. The cluster formation rates (J, cm^{-3} s^{-1}) of HIO_3-HIO_2 system obtained from experiment (CLOUD) and theoretical (ACDC) results. The simulation was performed under the conditions of [HIO_3] = 10^6 - 10^8, [HIO_2] = 2×10^4 - 2×10^6, T = 283 (red), 263 (blue) K, and CS = 2.2×10^{-3} s^{-1}. 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_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-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,
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