Responses to Referee #1's comments

We are grateful to the reviewers for their valuable and helpful comments on our manuscript "**Influence of atmospheric conditions on the role of trifluoroacetic acid in atmospheric sulfuric acid-dimethylamine nucleation**" (MS No.: acp-2020-1186). We have revised the manuscript carefully according to reviewers' comments. The point-to-point responses to the Referee #1's comments are summarized below:

General Comments:

Liu and co-workers investigated the influence of different atmospheric conditions on the role of trifluoroacetic acid (TFA) in sulfuric acid (SA) - dimethylamine (DMA) nucleation process using the Density Functional Theory combined with the Atmospheric Cluster Dynamics Code, which is based the previous study on that TFA can participate in SA-DMA-based nucleation under the local atmospheric temperature and nucleation precursor concentrations of Shanghai, China. This study reports the enhancement of particle formation rate by TFA and the contributions of SA-DMA-TFA cluster to the cluster formation pathways can be especially significant in cold and polluted areas, which can not only clarify the different roles of TFA in SA-DMA-based nucleation under broad atmospheric conditions, but also can reveal the potential implications of the usages of Freon alternatives on NPF under a wide range of atmospheric conditions. In general, the manuscript is well written and is of broad interest to the readership of Atmospheric Chemistry and Physics. I can recommend publication in Atmospheric Chemistry and Physics after the following comments have been addressed.

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

Specific Comments:

Comment 1.

Lines 22-24: "Although sulfuric acid (SA) - dimethylamine (DMA) nucleation mechanism has been identified in the urban city of Shanghai, China (Yao et al., 2018), the nucleation mechanisms are not fully understood and species contributing to NPF under different environments remain to be studied" Please further elaborate why the nucleation mechanisms are still not fully understood even though the SA-DMA nucleation mechanism has been identified in Shanghai.

Response: According to the reviewer's suggestion, we now state in Lines 22-25 of the revised manuscript that "Although sulfuric acid (SA) - dimethylamine (DMA) nucleation mechanism has been observed in various places around the world (Yao et al., 2018; Deng et al., 2020; Brean et al., 2020), there were still a lot of species observed in the atmosphere but not be fully assigned molecular formulas because of their chemical complexity.", and in Lines 27-28 of the revised manuscript that "However, other possible species that may potentially enhance the NPF rates and the

corresponding nucleation mechanism still should be further explored."

Comment 2.

Lines 29-30: "…, PFCAs are generally believed to be an important class of environmental contaminants present in various environments" Please provide relevant references for the atmospheric importance of PFCAs.

Response: According to the reviewer's suggestion, the relevant references for the atmospheric importance of PFCAs has been added in Lines 30-31.

Comment 3.

Section 3.1: From Figure 1, it seems that the $(SA)_3 \cdot (DMA)_4 \cdot (TFA)_1$ clusters can be set as boundary clusters. Are there other clusters that can be set as boundary? The detailed boundary clusters that can grow out of the simulated system by Atmospheric Cluster Dynamics Code and the reasons should be illustrated.

Response: We now state in Lines 113-116 of the revised manuscript that "The boundary of ACDC simulation should be set as the smallest clusters that are stable enough to grow outside of the simulated system (McGrath et al., 2012). Based on the stability of the studied clusters, $(SA)_4 \cdot (DMA)_3$ and $(SA)_3 \cdot (DMA)_4 \cdot (TFA)_1$ clusters are set to be boundary, which is consistent with that in the previous study (Lu et al., 2020)."

Comment 4.

Line 137: The corresponding temperature and DMA concentration for the 13 times enhancement by TFA should be presented.

Response: The corresponding temperature and DMA concentration for the 13 times enhancement by TFA have been added in Lines 148-149 of the revised manuscript that "… more than 13 times in January at [DMA] of 4.69×10^8 molecules cm^{-3} and 265 K."

Comment 5.

Line 195 and line 196: The detailed growing way of clusters in SA-DMA-TFA cluster formation pathway should be further illustrated. The reasons for that some clusters involving TFA does not present in the main cluster formation pathway should be elaborated.

Response: We now state in Lines 209-212 of the revised manuscript that "The clusters involving TFA can be formed initially by the addition of one TFA molecule to $(SA)_1 \cdot (DMA)_1$ cluster, and continue growing by the addition of one DMA molecule and then the addition of one $(SA)_1 \cdot (DMA)_1$ cluster. Other studied clusters involving TFA can't present in the main cluster formation pathway because of relatively low stability."

Comment 6.

Supplement, Table S5: Are these simulated results based on the thermodynamic parameters, such as Gibbs free formation energies (ΔG), at the corresponding

temperatures shown in Table S5? If so, the ΔG of studied clusters at different temperatures of the studied cities in different months should be presented in the Supplement.

Response: We now list the Gibbs free formation energies (ΔG) in Table S10 of the revised Supplement at the relevant temperatures of the studied cities in different months shown in Table S9.

Technical corrections:

Comment 7.

Line 73: "The collision rate coefficients $\beta_{i,j}$ between clusters *i* and *j* were ..." should be "The collision rate coefficients, $\beta_{i,j}$, between clusters *i* and *j* were ..."

Response: "The collision rate coefficients $\beta_{i,j}$ between clusters *i* and *j* were ..." has been corrected as "The collision rate coefficients, $\beta_{i,j}$, between clusters *i* and *j* were ..." in Line 76 of the revised manuscript.

Comment 8.

Line 83: "where P_{ref} is the reference pressure (in this case 1 atm) where the formation free energies..." should be "where P_{ref} is the reference pressure (in this case 1 atm), at which the formation free energies..."

Response: "where P_{ref} is the reference pressure (in this case 1 atm) where the formation free energies..." has been corrected as "where P_{ref} is the reference pressure (in this case 1 atm), at which the formation free energies..." in Line 85 of the revised manuscript.

Comment 9.

Line 86: "3.1 In Influence of temperature and nucleation precursor concentrations on cluster stability and growth trend …" should be "3.1 In Influence of temperature and nucleation precursor concentrations on the stability and growth trend …"

Response: According to the reviewer's suggestion and based on the addition of the discussion on influence of humidity in Section 3.1, "3.1 In Influence of temperature and nucleation precursor concentrations on cluster stability and growth trend …" has been corrected as "3.1 Influence of atmospheric conditions on the stability and growth trend …" in Line 88 of the revised manuscript.

Comment 10.

Line 135: "… temperature in spring and winter is relatively lower than other time all the year-round, respectively." should be "… temperatures in spring and winter are all relatively lower than other time all the year-round."

Response: "… temperature in spring and winter is relatively lower than other time all the year-round, respectively." has been corrected as "… temperature in spring and in winter is relatively lower than in other seasons all the year round." in Lines 146-147 of the revised manuscript.

Comment 11.

Supplement, Line 1 and Line 3: The "ΔG" should be in italic, such as "Δ*G*".
Response: The "ΔG" has been corrected in italic as "Δ*G*" in Line 29 and Line 31 of the revised Supplement.