

Responses to Referee #1's comments

We are grateful to the reviewers for their valuable and helpful comments on our manuscript “A novel formation mechanism of sulfamic acid and its enhancing effect on methanesulfonic acid-methylamine aerosol particle formation in agriculture-developed and coastal industrial areas” (Manuscript ID: EGUSPHERE-2024-2638). We have revised the manuscript carefully according to reviewers' comments. The point-to-point responses to the Referee #1's comments are summarized below:

Referee Comments

The manuscript egusphere-2024-2638, “A novel formation mechanism of $\text{NH}_2\text{SO}_3\text{H}$ and its enhancing effect on methanesulfonic acid-methylamine aerosol particle formation in agriculture-developed and coastal industrial areas”. The work studied the formation of sulfamic acid via HNSO_2 hydrolysis in the gas phase and at the air-water interface by using theoretical methods. Then, the author investigated the new particle formation for the role of sulfamic acid in $\text{CH}_3\text{SO}_3\text{H}-\text{CH}_3\text{NH}_2$ system. The work is very interesting for understanding the chemical processes of sulfamic acid in the atmosphere. However, there are some issues that should be addressed before publication.

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

Major issues

Comment 1.

In Line 39, “the concentration of $\text{NH}_2\text{SO}_3\text{H}$ was expected to reach up to $10^8 \text{ molecules} \cdot \text{cm}^{-3}$ ”, the concentration of sulfamic acid was only estimated by theoretical method, not measured by field observations. Therefore, it is required to elucidate this point.

Response: Thanks for the suggestion of the reviewer. As the suggestion of the reviewer, in Lines 38-40 Page 2 of the revised manuscript, the sentence of “the atmospheric concentration of $\text{NH}_2\text{SO}_3\text{H}$ was expected to reach up to $10^8 \text{ molecules} \cdot \text{cm}^{-3}$ (Li et al., 2018).” has been changed as “the atmospheric concentration of SFA estimated by theoretical method of CCSD(T)-F12/cc-pVDZ-F12//M06-2X/6-311++G(3df,3pd) (Li et al., 2018) was expected to reach up to $10^8 \text{ molecules} \cdot \text{cm}^{-3}$ ”.

Comment 2.

Lines 41-42, "the sources of $\text{NH}_2\text{SO}_3\text{H}$ in the atmosphere have been well investigated (Lovejoy and Hanson, 1996; Pszona et al., 2015; Li et al., 2018; Larson and Tao, 2001; Manonmani et al., 2020; Zhang et al., 2022)." In fact, sulfamic acid has been not investigated by field measurements. Therefore, it is not well investigated in the atmosphere.

Response: Thanks for the suggestion of the reviewer. Indeed, it is true that SFA has not been measured in the field. Therefore, atmospheric sulfamic acid has not been well studied. In Lines 41-43 Page 2 of the revised manuscript, "So, the sources of $\text{NH}_2\text{SO}_3\text{H}$ in the atmosphere have been well investigated (Lovejoy and Hanson, 1996; Pszona et al., 2015; Li et al., 2018; Larson and Tao, 2001; Manonmani et al., 2020; Zhang et al., 2022)." has been changed as "So, the sources of SFA in the atmosphere has been focused by several groups (Lovejoy and Hanson, 1996; Pszona et al., 2015; Li et al., 2018; Larson and Tao, 2001; Manonmani et al., 2020; Zhang et al., 2022).".

Comment 3.

Lines 46-47, "for the hydrolysis of SO_3 assisted by water molecule (10^{-11} - 10^{-10} $\text{cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$) (Kim et al., 1998; Hirota et al., 1996; Shi et al., 1994)." Some important references are missing such as J. Am. Chem. Soc. 2023, 145, 19866-19876. and J. Am. Chem. Soc. 1994, 116, 10314-10315.

Response: Thanks for the suggestion of the reviewer. We apologize for missing some important references. As the suggestion of the reviewer, some important references have been added in Lines 46-49 Page 2 of the revised manuscript, which has been organized as "which was close to the value for the hydrolysis of SO_3 assisted by water molecule (10^{-11} - 10^{-10} $\text{cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$) (Kim et al., 1998; Hirota et al., 1996; Shi et al., 1994; Kolb et al., 1994; Long et al., 2013; Long et al., 2023; Ding et al., 2023; Cheng et al., 2023; Wang et al., 2024).".

Comment 4.

What is the concertation of HNSO_2 in the atmosphere? This is very necessary for determining the importance of HNSO_2 in the atmosphere.

Response: Thanks for the suggestion of the reviewer. As the suggestion of the reviewer, we have conducted an extensive review of the relevant literature. However, the concentrations of HNSO_2 in the atmosphere have not been reported. As the absence of the concentration of HNSO_2 , the

competitiveness between MSA-assisted HNSO₂ hydrolysis and the NH₃-assisted ammonolysis of SO₃ (the traditional source of SFA) cannot be further confirmed. The related discussion has been found in Line 236 Page 8 to Line 237 Page 9 of the revised manuscript, which has been organized as “However, due to the absence of the concentration of HNSO₂, the competitiveness of these two reactions cannot be further confirmed.” Although the concentration of HNSO₂ has not been reported, it is still important to study HNSO₂ hydrolysis with MSA in the gas phase and at the air-water interface. The detailed importance of HNSO₂ hydrolysis with MSA has been presented as follows.

In the gas phase, with the significant decrease in atmospheric water molecules with increasing altitude, MSA has a significantly greater catalytic ability than H₂O in accelerating the rate of HNSO₂ hydrolysis within 5-15 km. At the air-water interface, two types of reactions, the ions forming mechanism and the proton exchange mechanism to form NH₂SO₃⁻···H₃O⁺ ion pair were observed on the timescale of picosecond, which is at least two orders of magnitude faster than the corresponding gas-phase reaction. Nobly, considering the overall environment of sulfuric acid emission reduction, the present findings suggest that SFA may play a significant role in NPF and the growth of aerosol particles as *i*) SFA can directly participate in the formation of MSA-CH₃NH₂-based cluster and enhance the rate of NPF from these clusters by approximately 10³ times at 278.15 K; and *ii*) the NH₂SO₃⁻ species at the air-water interface can attract gaseous molecules to the aqueous surface, and thus promote particle growth.

Comment 5.

The reliability of the chosen methods should be clarified in the HNSO₂ + CH₃SO₃H reaction. Although the traditional method CCSD(T)/M06-2X has been widely used for atmospheric reactions, it should be noted that there are quite large uncertainties for estimating barrier height. This should clearly tell the potential readers.

Response: Thanks for the suggestion of the reviewer. As the suggestion of the reviewer, the reliability of CCSD(T)/aug-cc-pVDZ//M06-2X/6-311+G(2df,2pd)-based calculation method has been verified as follows. Firstly, the geometry and frequency calculation involved in the HNSO₂ hydrolysis were verified (Fig. S2) at three different theoretical levels of M06-2X/6-311++G(3df,2pd), M06-2X/6-311++G(3df,3pd) and M06-2X/aug-cc-pVTZ and experimental values. Then, based on the M06-2X/6-311++G(2df,2pd) optimized geometries, the corresponding

single point energy calculations (Table S1) were performed at the CCSD(T)-F12/cc-pVDZ-F12, CCSD(T)-F12/cc-pVTZ-F12, CCSD(T)/CBS and CCSD(T)/aug-cc-pVTZ levels, respectively. The main revision has been made as follows.

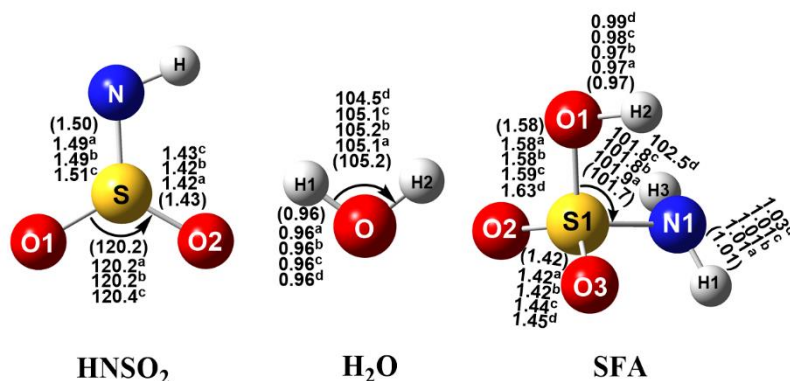


Fig. S2 The optimized geometrical structures for the species involved in the HNSO₂ hydrolysis at several different levels of theory.

a, b and, c respectively represents the values obtained at the M06-2X/6-311++G(3df,2pd), M062X/6-311++G(3df,3pd) and M06-2X/aug-cc-pVTZ level of theory, ^d represents the experimental values (The values in parentheses were obtained at the M06-2X/6-311++G(2df,2pd) level of theory; bond length is in angstrom and angle is in degree.).

(a) The geometric parameters of the reactants of HNSO₂, H₂O and NH₂SO₃H (SFA) have been displayed in Fig. S2. As seen in Fig. S2, the mean absolute deviation of calculated bond distances and bond angles between the M06-2X/6-311++G(2df,2pd) level and the experimental reports were 0.02 Å and 0.57°, respectively. This reveals that the calculated bond distances and bond angles at the M06-2X/6-311++G(2df,2pd) level agree well with the available experimental values (From the pubchem database, <https://pubchem.ncbi.nlm.nih.gov/#opennewwindow>). In addition, we have re-optimized all equilibrium structures of HNSO₂, H₂O and NH₂SO₃H at three different theoretical levels of M06-2X/6-311++G(3df,2pd), M062X/6-311++G(3df,3pd) and M06-2X/aug-cc-pVTZ levels. For the calculated geometrical parameters of these species, the mean absolute deviation of calculated bond distances and bond angles between the M06-2X/6-311++G(2df,2pd) level and the other levels were within 0.02 Å and 0.2°, respectively. Therefore, due to its efficiency, the M06-2X/6-311++G(2df,2pd) was adopted to optimize the geometries of all stationary points involved in the HNSO₂ hydrolysis. Based on this, in Lines 121 to 124 Page 5 of the revised manuscript, the sentence of “It is noted that the calculated bond distances and bond angles at the M06-2X/6-311++G(2df,2pd) level (Fig. S2) agree well with the available values (Fig. S2) from the experiment and three different theoretical levels of M06-2X/6-311++G(3df,2pd), M062X/6-311++G(3df,3pd) and M06-2X/aug-cc-pVTZ levels.” has been added.

Table S1 The Energy barriers (ΔE) and unsigned error (UE) ($\text{kcal}\cdot\text{mol}^{-1}$) for the HNSO_2 hydrolysis at different theoretical the potential energy profile (ΔG) correction

Methods	ΔE^a	ΔE^b	ΔE^c	UE
CCSD(T)/CBS//M06-2X/ 6-311++G(2df,2pd)	3.4	29.7	-23.0	0.00
CCSD(T)-F12/cc-pVDZ-F12//M06-2X/ 6-311++G(2df,2pd)	3.6	30.6	-22.0	0.71

^a, ^b and ^c respectively denote the species of pre-reactive complexes, transition states and products involved in the HNSO_2 hydrolysis.

(b) To further confirm the reliability of the CCSD(T)-F12/cc-pVDZ-F12//M06-2X/6-311++G(2df,2pd) level of theory, single-point energy calculations for the HNSO_2 hydrolysis in the gas phase have been performed at two different levels of CCSD(T)/CBS and CCSD(T)-F12/cc-pVDZ-F12 based on the optimized geometries at the M06-2X/6-311++G(2df,2pd) level. Notably, the complete basis set (CBS) obtained by basis set extrapolation is used as the reference basis set. As presented in Table S1, compared with unsigned error calculated at the CCSD(T)/CBS//M06-2X/6-311++G(2df,2pd) level, unsigned errors calculated at CCSD(T)-F12/cc-pVDZ-F12//M06-2X/6-311++G(2df,2pd) was $0.71 \text{ kcal}\cdot\text{mol}^{-1}$. This suggests that the relative energies obtained at the CCSD(T)/aug-cc-pVDZ//M06-2X/6-311++G(2df,2pd) level was reasonable. Considering the computational accuracy and cost, the CCSD(T)/aug-cc-pVDZ//M06-2X/6-311++G(2df,2pd) method was chosen to calculate the single point energies of all the species involved in the HNSO_2 hydrolysis. Thus, in Lines 130 to 133 Page 5 of the revised manuscript, the sentence of “The CCSD(T)/aug-cc-pVDZ method was chosen to calculate the relative energies as the fact that, compared with unsigned error (Table S1) calculated at the CCSD(T)/CBS//M06-2X/6-311++G(2df,2pd) level, unsigned errors calculated at CCSD(T)-F12/cc-pVDZ-F12//M06-2X/6-311++G(2df,2pd) was $0.71 \text{ kcal}\cdot\text{mol}^{-1}$.” has been added.

Comment 6.

In kinetics calculations, it is unclear. There are lots of issues that must be addressed. Provide the details of VRC calculations. For example, how to set pivot points and what is the electronic structure method for VRC-TST calculations? The author should provide the input files for VRC-TST and MESMER calculations in Supporting information to help the potential readers to understand the computational details.

Response: Thanks for the suggestion of the reviewer. The pivot point setting method and the electronic structure method for VRC-TST calculation are provided in detail (shown in Part S1 in the Supplement). Meanwhile, the input files for VRC-TST and MESMER calculations have been provided in Supporting information. The main revision has been made as follows.

(a) Herein, we describe the implementation details of the VRC-TST calculation in Part S1 in the Supplement. Specifically, there are two assumptions in VRC-VTST calculation: (1) the contribution of the vibrational modes of reactants to the partition function is canceled by the corresponding contribution of transition states to the partition function; (2) the internal geometries of reactants are fixed along the reaction coordinate. The reaction coordinate in VRC-VTST is different from that in RP-VTST and determined by the pivot points of each reactant fragment. For the HNSO₂ hydrolysis reaction, the pivot points of HNSO₂ (points 1 and 2) are located at a distance $\pm d$ along its S axis. Meanwhile, the pivots of H₂O (points 3 and 4) are located at a distance $\pm d$ perpendicular to H₂O molecule lane. As shown in Fig. S6, the Multiwfn package combined with the VMD software is adopted to visualize the reaction system and help determine the location of pivot points. The reaction coordinate value (s) is defined as the minimum of the distance (r_{ij}) between the pivot point i ($=1$ or 2) and pivot point j ($=3$ or 4), where i and j represent the pivot points of HNSO₂ and H₂O molecules, respectively. Hence, each of the four dividing surfaces is obtained by symmetrically placing two pivot points of each radical fragment (1-3, 1-4, 2-3, and 2-4). For example, if the reaction coordinate s is equal to r_{23} , one of the four dividing surfaces (2-3), is determined by the locations of pivot points 2, 3 and the reaction coordinate s . There are total four pair of pivot points, the other three dividing surfaces (1-3, 1-4, 2-4) are defined by their corresponding pivot points and reaction coordinates s . Note that the locations of pivot points are critical to the rate constant calculation. Considering the difference between HNSO₂ and H₂O molecules, the distance s between pivot points varies from 2.5 to 6 Å for HNSO₂ and H₂O in each case with a 0.5 Å grid increment. So, in Lines 139 to 141 Page 5 of the revised manuscript, the sentence of the “Meanwhile, two pivot points (Bao et al., 2016; Long et al., 2021; Georgievskii and Klippenstein, 2003; Meana-Pañeda et al., 2024) were selected to calculate the high-pressure limiting rate for the HNSO₂ hydrolysis (shown in Part S1 in the Supplement).” has been added. Also, the computational details of VRC-VTST calculations have been added in Line 159 Page S15 to 180 Page S16 of the revised Supplement.

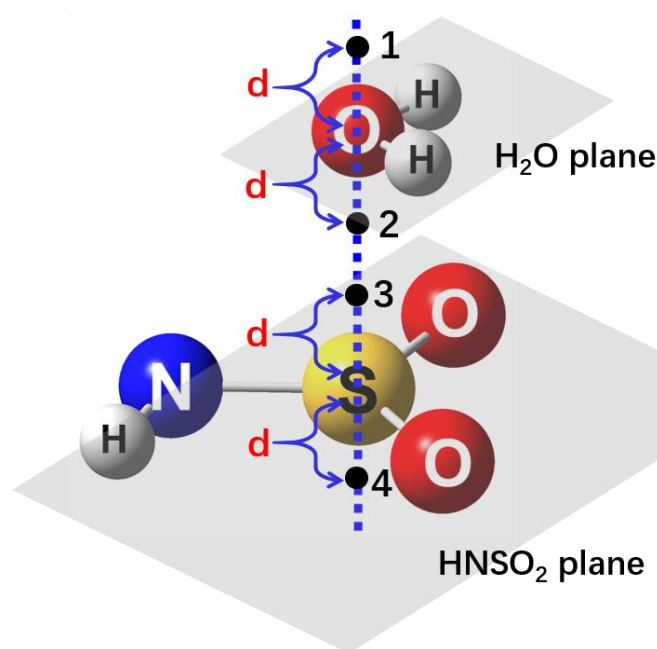


Fig. S6 The placements of the pivot points for the HNSO₂ hydrolysis

(b) The electronic structure method for VRC-TST calculations is based on Gaussian 09 program using the M06-2X/6-311++G(2df,2pd). So, in Lines 137-139 Page 5 of the revised manuscript, the sentence of the “It's worth noting that the electronic structure method for VRC-TST calculations is based on Gaussian 09 program using the M06-2X/6-311++G(2df,2pd).” has been added.

(c) The input files for VRC-TST and MESMER calculations have been provided in Supplement.

Comment 7.

According to the authors' previous research (Phys. Chem. Chem. Phys., 2022, 24, 4966-4977), the reaction of HNSO₂ with nH₂O also has a sufficiently low free energy barrier, which implies that HNSO₂ can undergo hydrolysis or decomposition directly at the gas-liquid interface or in the bulk phase. This seems to contradict the explanation on line 228 (page 8), given that the concentration of water is sufficiently high.

Response: Thanks for the suggestion of the reviewer. According to the previous work (Phys. Chem. Chem. Phys., 2022, 24, 4966-4977), the hydrolysis of HNSO₂ assisted by H₂O, (H₂O)₂ and (H₂O)₃ involved a loop structure mechanism. These reactions were known to occur via the initial formation of ring hydrogen bonding complex HNSO₂⋯(H₂O)_n (n = 1-3) with the calculated relative free energy of 0.2-3.6 kcal·mol⁻¹ followed by their rearrangement to form NH₂SO₃H. As the higher entropy effect, hydrogen bonding complex HNSO₂⋯(H₂O)_n (n = 1-3) were formed hardly under

actual atmospheric conditions, and thus the loop structure mechanism for the hydrolysis of HNSO₂ assisted by H₂O, (H₂O)₂ and (H₂O)₃ is not easy to occur in the gas phase. This is similar with CH₃SO₃H-assisted gaseous hydrolysis of HNSO₂ which does not occur within the 100 ps.

At the air-water interface, the HNSO₂ molecule is stable and does not dissociate within 10 ps, where the loop structure of hydrogen bonding complex HNSO₂⋯(H₂O)_n (*n* = 1-3) has not been observed. This is proved by the BOMD simulation illustrated in Fig. S8 where the hydrated form of HNSO₂ was not conducive to HNSO₂ hydrolysis at the air-water interface. So, even if the concentration of water molecules at the air-water interface is sufficiently high, the probability that HNSO₂ can be hydrolyzed or decomposed either directly at the air-water interface or in the bulk phase is small. This is agreed with the simulation results. Based on this analysis above, in Lines 272-274 Page 10 of the revised manuscript, the sentence of the “Meanwhile, although HNSO₂ remains stable at the air-water interface (seen in Fig. S8) and does not dissociate within 10 ps, the hydrated form of HNSO₂ illustrated in Fig. S8 was not conducive to HNSO₂ hydrolysis at the air-water interface.” has been added to prove that the hydrated form of HNSO₂ was not conducive to HNSO₂ hydrolysis at the air-water interface.

Comment 8.

Why did the authors not consider a third access channel in the gas phase, that is, the reaction pathway of HNSO₂⋯CH₃SO₃H + H₂O? Considering the reactions at the gas-liquid interface, it seems more plausible that HNSO₂⋯CH₃SO₃H would first form a complex before reacting with water molecules. Considering the reactions at the gas-liquid interface, it seems more plausible that HNSO₂⋯CH₃SO₃H would first form a complex before reacting with water molecules.

Response: Thanks for the suggestion of the reviewer. Indeed, the reaction pathway of HNSO₂⋯MSA + H₂O is feasible. However, the concentration of water molecules in the atmosphere is about 10¹⁸ molecules·cm⁻³, which is much higher than those of HNSO₂ and MSA (10⁵-10⁹ molecules·cm⁻³). Considering the harsh conditions for the initial formation of dimers between HNSO₂ and MSA (i.e., HNSO₂ and MSA are sufficiently concentrated in the atmosphere.), we predict that the primary preliminary dimers will continue to be dominated by HNSO₂⋯H₂O and MSA⋯H₂O complexes. So, in Line 203 Page 7 to Line 207 Page 8 of the revised manuscript, the sentence of the “As the concentration of water molecule (10¹⁸ molecules·cm⁻³ (Anglada et al., 2013)) in the atmosphere is

much higher than those of HNSO₂ and MSA (10⁵-10⁹ molecules·cm⁻³ (Shen et al., 2020)), the reaction pathway of HNSO₂···MSA + H₂O is hard to occur in actual atmospheric conditions. So, Channel MSA proceeds through the initial formation of dimers (HNSO₂···H₂O and MSA···H₂O) via collisions between HNSO₂ (or MSA) and H₂O.” has been added.

Comment 9.

In Section 3.3, the authors examined the impact of MSA-MA-SFA clusters on nucleation. Interestingly, DMA, which has a stronger nucleation capability, and NH₃, which has a higher concentration, were excluded. I would like the authors to provide some appropriate justifications for this.

Response: Thanks for the suggestion of the reviewer. Previous studies have demonstrated that MSA-driven new particle formation (NPF) has attracted growing attention, as MSA significantly contributes to NPF in scenarios with only natural sources of SO₂ were present. Currently, atmospheric bases, including methylamine (MA), monoethanolamide, and dimethylamine (DMA), have a key role in MSA-driven aerosol particle generation and growth, where MA exhibits the strongest enhancing capability (*Environ. Sci. Technol.*, 2017, 51, 243-252; *J. Phys. Chem. B*, 2016, 120, 1526-1536; *Environ. Sci. Technol.*, 53, 14387-14397, 2019; *Atmos. Environ.*, 2023, 311, 120001). So, we choose MA over DMA and NH₃. This choice is similar to that previously reported in the relevant references (*Atmos. Chem. Phys.*, 22, 2639-2650; 2022*Atmos. Environ.*, 2023, 311, 120001). Based on this analysis above, in Lines 94-96 Page 4 of the revised manuscript, the sentence of the “Initially, the binary nucleation of MSA with inorganic ammonia and organic amines in the atmosphere has been reported, where MA exhibits the strongest enhancing capability (Chen et al., 2016; Chen and Finlayson-Pitts, 2017; Shen et al., 2019; Hu et al., 2023).” has been reorganized to prove that the MSA-MA system was chosen over MSA-DMA.

Comment 10.

Since the ammonolysis of SO₃ is the primary pathway for SFA formation, the authors could have compared it with the current pathway, which would be necessary for accurately assessing the atmospheric significance of the current reaction.

Response: Thanks for the suggestion of the reviewer. As the suggestion of reviewer, we compared the NH_3 -assisted ammonolysis of SO_3 with the MSA-assisted HNSO_2 hydrolysis. In Line 233 Page 8 to Line 237 Page 9 of the revised manuscript, “Besides, MSA-assisted HNSO_2 hydrolysis is reduced by $4.9 \text{ kcal}\cdot\text{mol}^{-1}$ in energy barrier than the NH_3 -assisted ammonolysis of SO_3 with its rate constant at 298 K ($2.85 \times 10^{-11} \text{ cm}^3\cdot\text{molecule}^{-1}\cdot\text{s}^{-1}$) close to the value of ammonolysis of SO_3 with NH_3 ($4.35 \times 10^{-10} \text{ cm}^3\cdot\text{molecule}^{-1}\cdot\text{s}^{-1}$) (Li et al., 2018). However, due to the absence of the concentration of HNSO_2 , the competitiveness of these two reactions cannot be further confirmed.” has been added.

Comment 11.

In Fig. 6b and Fig. 7b, it is necessary for the authors to carefully examine whether the significant abrupt changes caused by the concentrations of SFA and MA are reasonable.

Response: Thanks for the suggestion of the reviewer. It is noted that in Fig. 6(b), due to the competitive relationship between MSA and SFA, at low concentrations of SFA, the binding capacity of MSA with MA is stronger than that of SFA with MA, resulting in only a small amount of SFA participating in cluster formation. However, as the concentration of SFA increases, the number of $(\text{MSA})_x\cdot(\text{MA})_y\cdot(\text{SFA})_z$ (where $y \leq x + z \leq 3$) ternary clusters increases, leading to the formation of more hydrogen bonds and a significant increase in R_{SFA} . Similarly, in Fig. 7(b), at a certain concentration of SFA and MA, as the concentration of MSA increases, the hydrogen bonds between SFA and MA are disrupted, leading to more binding of MA and MSA rather than SFA, resulting in a sharp decrease in R_{SFA} . In Lines 364-369 Page 12 of the revised manuscript, “It is noted that in Fig. 6(b), due to the competitive relationship between MSA and SFA, at low concentrations of SFA, the binding capacity of MSA with MA is stronger than that of SFA with MA, resulting in only a small amount of SFA participating in cluster formation. However, as the concentration of SFA increases, the number of $(\text{MSA})_x\cdot(\text{MA})_y\cdot(\text{SFA})_z$ (where $y \leq x + z \leq 3$) ternary clusters increase, leading to the formation of more hydrogen bonds and a significant increase in R_{SFA} .” has been added.

Comment 12.

In the introduction, the authors mention that the pK_a may affect the transfer of protons, thereby affecting the catalytic ability. Whether similar trends will also directly affect the nucleation

271 capability should be considered, such as in the cases of MSA-MA-SFA, MSA-MA-SA, and SA/FA-
272 MA-SFA.

273 **Response:** Thanks for the suggestion of the reviewer. We apologize for the misunderstanding about
274 pK_a in Lines 63-65 Pages 2-3. Indeed, our aim is to illustrate the importance of MSA as a catalyst
275 from pK_a perspective. In order not to create ambiguity, as for the discussions of pK_a , the sentence
276 of the “It was noted that as the acidity of $\text{CH}_3\text{SO}_3\text{H}$ ($pK_a = -1.92$) was significantly stronger than
277 that of water ($pK_a = 15.0$) and formic acid ($pK_a = 3.74$), it may be predicted that the proton transfer
278 reaction for the hydrolysis of HNSO_2 with $\text{CH}_3\text{SO}_3\text{H}$ was much easier than those with water and
279 formic acid. It was also noted that although $\text{CH}_3\text{SO}_3\text{H}$ was less acidic than H_2SO_4 ($pK_a = -3.00$),
280 with the global reduction in the concentration of H_2SO_4 resulting from SO_2 emission restrictions,
281 the contribution of $\text{CH}_3\text{SO}_3\text{H}$ to aerosol nucleation has received the widespread attention of
282 scientists.” had been deleted. Meanwhile, the importance of MSA as a catalyst in HNSO_2 hydrolysis
283 has been organized as “It was noted that, with the global reduction in the concentration of H_2SO_4
284 resulting from SO_2 emission restrictions, the contribution of MSA to aerosol nucleation has received
285 the widespread attention of scientists.” in Lines 63-66 Page 3 of the revised manuscript.