Supplementary Materials: Enhancing Potential of Trimethylamine Oxide on Atmospheric Particle Formation

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1. Enthalpies, Entropies and Gibbs Free Energies

Table S1 presents binding enthalpies ΔH, entropies ΔS and Gibbs free energies ΔG of all sulfuric acid clusters with trimethylamine oxide (TMAO), dimethylamine (DMA) and guanidine (GUA). The corresponding TMAO-containing cluster structures in xyz format are found in the supplementary zip folder.

Table S1. Binding enthalpies (ΔH, kcal/mol), entropies (ΔS, kcal/(mol K)) and Gibbs free energies (ΔG, kcal/mol, calculated at 298.15 K).

Cluster	ΔH	ΔS	ΔG
1sa1tmao	−32.18322673	−34.920	−21.77
1sa2tmao	−56.06440660	−73.704	−34.09
1sa3tmao	−73.37817629	−109.196	−40.82
1sa4tmao	−88.28659373	−139.939	−46.56
2sa1tmao	−60.03857843	−76.472	−37.24
2sa2tmao	−90.56865434	−110.843	−57.52
2sa3tmao	−119.4695584	−157.075	−72.64
2sa4tmao	−141.0910972	−190.579	−84.27
3sa1tmao	−85.45125574	−123.634	−48.59
3sa2tmao	−121.9005101	−154.788	−75.75
3sa3tmao	−155.9899463	−197.338	−97.15
3sa4tmao	−183.9793932	−236.825	−113.37
4sa1tmao	−106.3854315	−164.245	−57.42
4sa2tmao	−148.7282098	−194.019	−90.88
4sa3tmao	−190.1134950	−242.604	−117.78
4sa4tmao	−221.9682212	−284.864	−137.04
2tmao	−13.81391411	−34.505	−3.53
3tmao	−24.91821751	−70.085	−4.02
4tmao	−37.93158466	−106.902	−6.06
1sa1dma	−22.17176522	−30.259	−13.15
1sa2dma	−36.53433065	−66.044	−16.84
1sa3dma	−51.82356685	−105.073	−20.50
1sa4dma	−64.44927049	−145.991	−20.92
2sa1dma	−54.46582365	−75.803	−31.87
2sa2dma	−82.46719797	−116.287	−47.80
2sa3dma	−102.1051307	−157.467	−55.16
2sa4dma	−116.5346491	−191.740	−59.37
3sa1dma	−78.94652149	−120.447	−43.04
3sa2dma	−110.9413881	−159.104	−63.51
3sa3dma	−142.3888000	−202.227	−82.10
3sa4dma	−163.9221030	−241.571	−91.90
4sa1dma	−97.45230567	−160.934	−49.47
4sa2dma	−135.1529839	−206.896	−73.47
4sa3dma	−166.0450292	−240.667	−94.29
4sa4dma	−196.1323563	−275.835	−113.89
2dma	−3.654761626	−26.762	4.32
3dma	−11.51196982	−60.408	6.50
4dma	−19.50276342	−84.997	5.84
1sa1gua	−29.38971226	−30.365	−20.34
1sa2gua	−49.50901342	−70.716	−28.43
1sa3gua	−71.41231126	−111.597	−38.14
1sa4gua	−90.94789974	−152.269	−45.55
2sa1gua	−62.17903499	−82.402	−37.61
2sa2gua	−100.8614343	−111.970	−67.48
2sa3gua	−126.5774611	−167.702	−76.58
2sa4gua	−151.0306295	−207.854	−89.06
3sa1gua	−86.19536698	−120.908	−50.15
3sa2gua	−131.4567435	−166.061	−81.95
3sa3gua	−170.3301352	−200.817	−110.46
3sa4gua	−203.9377305	−240.451	−132.25
4sa1gua	−107.4876551	−161.589	−59.31
4sa2gua	−156.9157684	−209.605	−94.42
4sa3gua	−202.8881849	−247.559	−129.08
4sa4gua	−255.8136593	−293.435	−168.33
2gua	−12.42103630	−34.152	−2.24
3gua	−24.88994819	−73.183	−3.07
4gua	−44.92701155	−115.500	−10.49
2sa	−17.76423906	−36.844	−6.78
3sa	−36.70924364	−80.667	−12.66
4sa	−52.62105419	−107.963	−20.43

2. Dimethylamine and Guanidine Containing Clusters

Figure S1 presents the main cluster growth pathways of SA–GUA and SA–DMA clusters at [acid]=[base]=10^6 and [acid]=[base]=10^7 cm^{-3}, at 298.15 K.

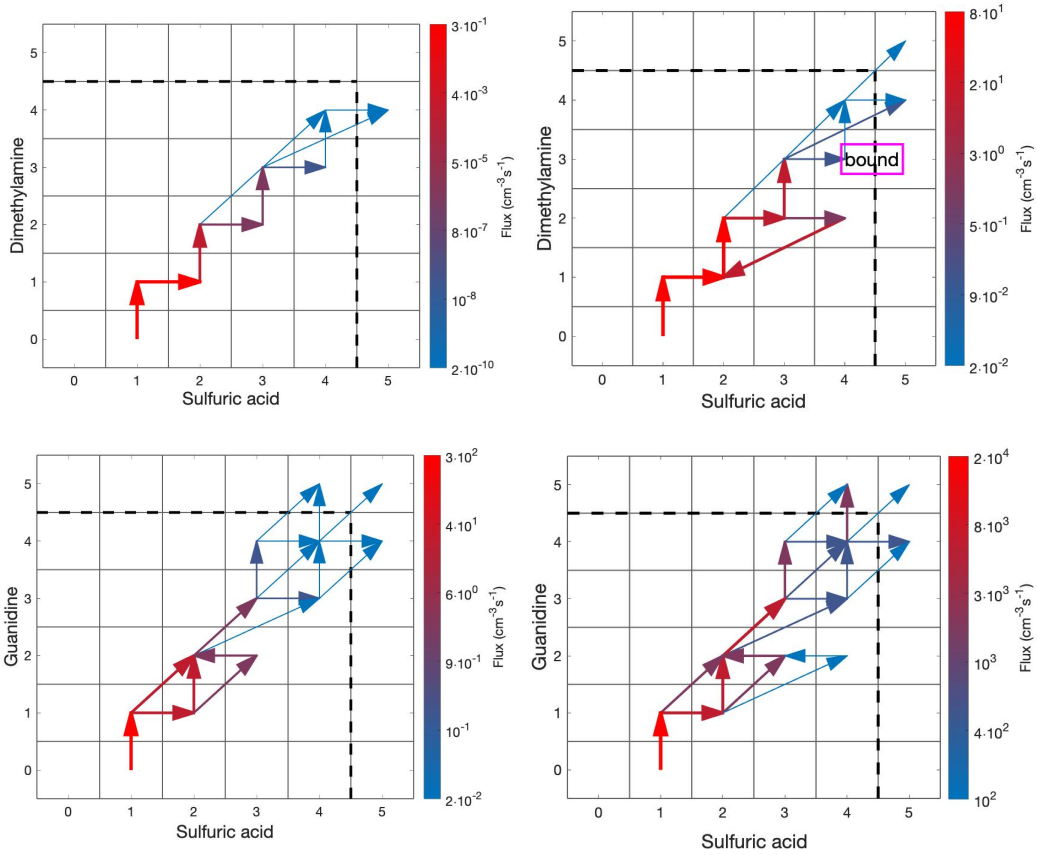

Figure S1. Main cluster growth pathways for SA–DMA (top) and SA–GUA clusters (bottom) at 298.15 K and vapor concentrations of [acid]=10^6 cm^{-3} and [base]=10^6 cm^{-3} (left) and [acid]=10^7 cm^{-3} and [base]=10^7 cm^{-3} (right). The x- and y-axes give the numbers of acid and base molecules in the cluster, respectively.

Figure S2 shows the actual vapor concentration-dependent Gibbs free energies of SA–GUA and SA–DMA clusters at [acid]=[base]=10^6 and [acid]=[base]=10^7 cm^{-3}, at 298.15 K.

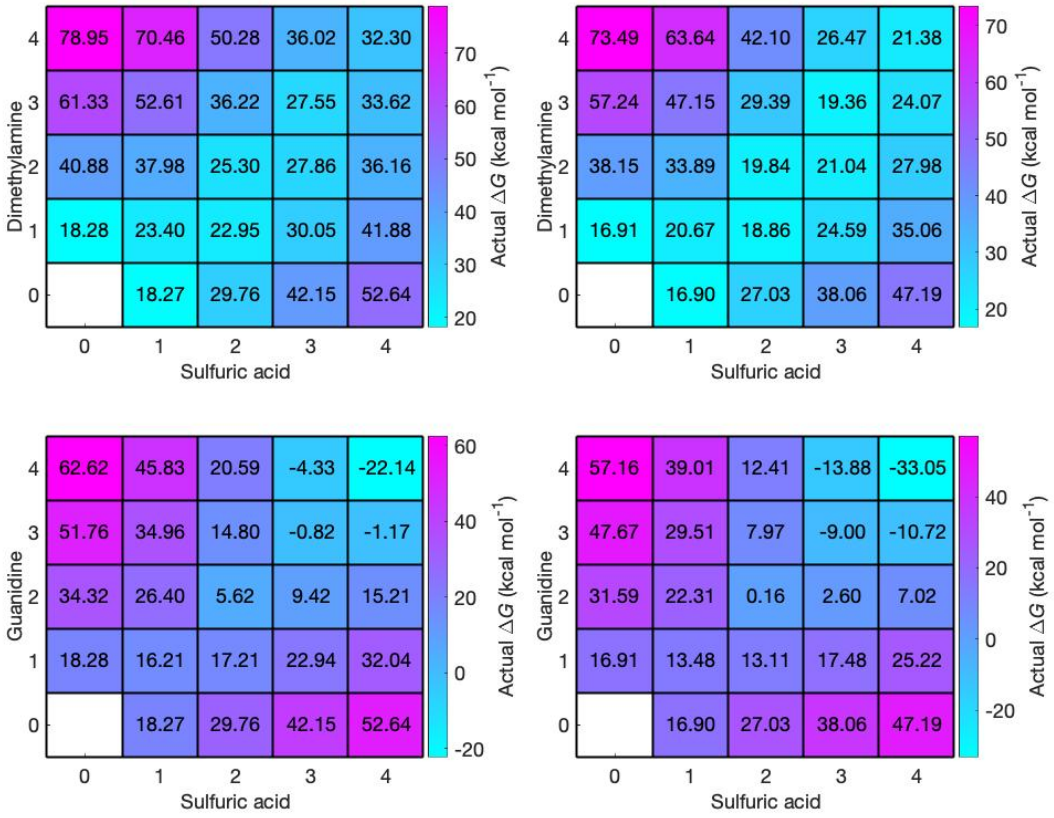

Figure S2. Vapor concentration-dependent Gibbs free energies for SA–DMA (top) and SA–GUA clusters (bottom) at 298.15 K and vapor concentrations of [acid]=10^6 cm^{-3} and [base]=10^6 cm^{-3} (left) and [acid]=10^7 cm^{-3} and [base]=10^7 cm^{-3} (right). The x- and y-axes give the numbers of acid and base molecules in the cluster, respectively.

Figure S3 shows the steady-state cluster distributions of SA–GUA and SA–DMA clusters at [acid]=[base]=10^6 and [acid]=[base]=10^7 cm^{-3}, at 298.15 K.

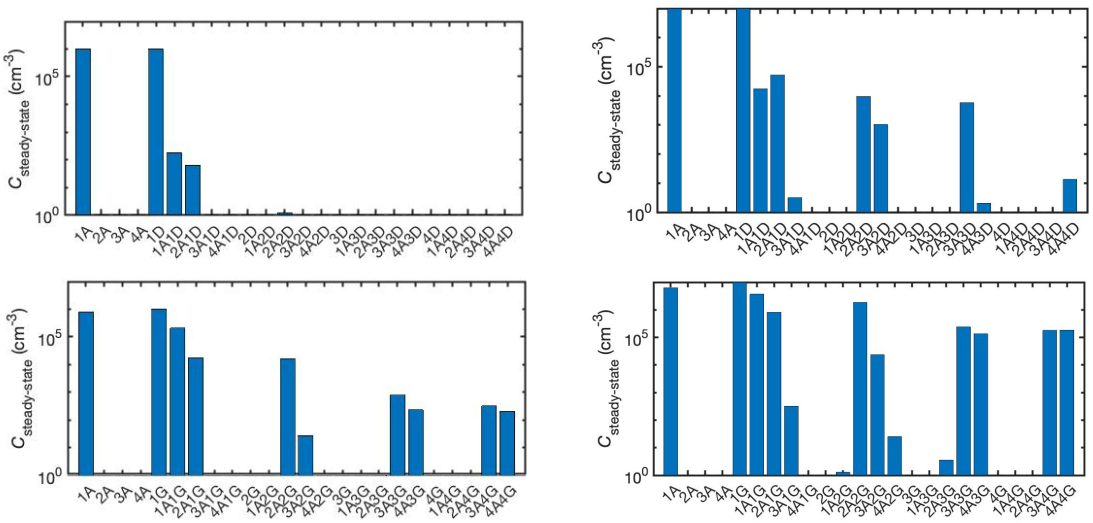

Figure S3. Steady-state cluster distributions for SA–DMA (top) and SA–GUA clusters (bottom) at 298.15 K and vapor concentrations of [acid]=10^6 cm^{-3} and [base]=10^6 cm^{-3} (left) and [acid]=10^7 cm^{-3} and [base]=10^7 cm^{-3} (right). Here A refers to sulfuric acid, D to dimethylamine and G to guanidine.