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Supplement of

Influence of atmospheric conditions on the role of trifluoroacetic acid in atmospheric sulfuric acid–dimethylamine nucleation

Ling Liu et al.

Correspondence to: Xiuhui Zhang (zhangxiuhui@bit.edu.cn)

Section 1

From the ratios of $\beta \cdot C/\Sigma\gamma$ for the studied SA-DMA-TFA clusters (Fig. 1), it can be seen that the $(SA)_2(DMA)_3(TFA)_1$ and $(SA)_1(DMA)_2(TFA)_1$ clusters are relatively stable against evaporation and can be able to grow into larger clusters. The $(SA)_1(DMA)_1(TFA)_1$ cluster is the initial and key cluster to form $(SA)_2(DMA)_3(TFA)_1$ and $(SA)_1(DMA)_2(TFA)_1$ clusters as shown in Fig. 4, and this cluster formation pathway through $(SA)_1(DMA)_1(TFA)_1$ involves a modest thermodynamic barrier shown by the previous study (Lu et al. 2020). Hence, the formation of $(SA)_1(DMA)_1(TFA)_1$ cluster is the limiting step in SA-DMA-TFA new particle formation, which is similar to the analysis of the limiting step of $(SA)_n(Base)_n$ system (Elm, 2017). Therefore, in order to understand the influence of relative humidity (RH) on the SA-DMA-TFA system, the evaluation of the influence of RH on the formation kinetics of $(SA)_1(DMA)_1(TFA)_1$ cluster is of significance. Herein, the kinetic property of hydrated clusters relevant to the formation of the hydrated $(SA)_1(DMA)_1(TFA)_1$ clusters, which involves collisions of smaller clusters with monomers and evaporation of monomers from larger clusters, has been studied. The relative hydrate distributions of the clusters (Tables S5-S6) and the effective collision coefficients and evaporation coefficients (Table S7) weighted average over the hydrate distributions (Paasonen et al., 2012) are calculated at 280 K and 260 K. The Cartesian coordinates of these studied hydrated clusters are listed in Table S8. The relative distributions of the unhydrated $(SA)_1(DMA)_1(TFA)_1$ and $(SA)_1(DMA)_2(TFA)_1$ clusters are more than 50%, which are higher than those of the corresponding hydrated clusters at different RHs (20%, 40%, 60%, 80%, and 100%) and different temperatures (280 K and 260 K). Hence, most of $(SA)_1(DMA)_1(TFA)_1$ and $(SA)_1(DMA)_2(TFA)_1$ clusters are unhydrated in the atmosphere. Furthermore, the ratios of effective collision frequencies with nucleation monomers versus total effective evaporation frequencies ($\beta_{eff}C/\Sigma\gamma_{eff}$) of these two key clusters almost vary slightly within one order of magnitude at different RHs and different temperatures (Table S7). Therefore, the studied SA-DMA-TFA system is insensitive to the variation of humidity, which is similar to the SA-DMA system (Olenius et al., 2017).

Section 2

Table S1. Gibbs free formation energies (Δ*G*, kcal/mol) of the studied clusters at different temperatures at the RI-CC2/aug-cc-pV(T + d)Z//M06-2X/6-311++G(3df,3pd) level of theory.

Clusters	Δ*G* (kcal/mol)	
	280 K (Lu et al., 2020)	260 K
$(TFA)_2$	-5.76	-6.51
$(TFA)_3$	-3.79	-5.24
$(SA)_1$ $(TFA)_1$	-7.83	-8.53
$(SA)_1$ $(TFA)_2$	-13.49	-15.03
$(SA)_2$ $(TFA)_1$	-14.68	-16.27
$(DMA)_1$ $(TFA)_1$	-8.65	-9.31
$(DMA)_1$ $(TFA)_2$	-19.06	-20.57
$(DMA)_1$ $(TFA)_3$	-23.56	-26.06
$(DMA)_2$ $(TFA)_2$	-34.55	-36.88
$(SA)_1$ $(DMA)_1$ $(TFA)_1$	-27.21	-28.67
$(SA)_2$ $(DMA)_1$ $(TFA)_1$	-42.13	-44.48
$(SA)_1$ $(DMA)_2$ $(TFA)_1$	-44.92	-47.27
$(SA)_1$ $(DMA)_2$ $(TFA)_2$	-52.41	-55.74
$(SA)_2$ $(DMA)_2$ $(TFA)_1$	-61.91	-65.25
$(SA)_1$ $(DMA)_1$ $(TFA)_2$	-36.32	-38.57
$(SA)_2$	-9.04	-9.72
$(SA)_3$	-15.33	-16.87
$(SA)_2$ $(DMA)_1$	-36.30	-37.85
$(SA)_2$ $(DMA)_2$	-56.95	-59.26
$(SA)_3$ $(DMA)_1$	-50.06	-52.44
$(SA)_3$ $(DMA)_2$	-73.77	-76.90
$(SA)_3$ $(DMA)_3$	-94.24	-98.18
$(SA)_1$ $(DMA)_1$	-15.28	-15.98
$(SA)_2$ $(DMA)_3$ $(TFA)_1$	-79.52	-83.64
$(SA)_1$ $(DMA)_3$ $(TFA)_2$	-68.39	-72.51
$(DMA)_3$ $(TFA)_3$	-54.18	-58.31
$(DMA)_2$ $(TFA)_3$	-43.13	-46.42

Table S2. Collision coefficients (β, cm^3 s^{-1}) for each cluster in the present study.

Collisions	β (cm^3 s^{-1})	
	280 K (Lu et al., 2020)	260 K
DMA+SA	2.45×10^{-10}	2.36×10^{-10}
$(SA)_1$ $(DMA)_1$+SA	2.67×10^{-10}	2.57×10^{-10}
$(SA)_2$ $(DMA)_1$+SA	2.61×10^{-10}	2.52×10^{-10}
$(SA)_2$ $(DMA)_1$+DMA	3.91×10^{-10}	3.77×10^{-10}
$(SA)_2$ $(DMA)_2$+SA	3.05×10^{-10}	2.94×10^{-10}
$(SA)_3$ $(DMA)_2$+DMA	5.82×10^{-10}	5.61×10^{-10}
DMA+TFA	2.85×10^{-10}	2.75×10^{-10}
$(SA)_1$ $(DMA)_1$+TFA	2.95×10^{-10}	2.84×10^{-10}
$(SA)_2$ $(DMA)_1$+TFA	2.84×10^{-10}	2.74×10^{-10}
DMA+$(SA)_1$ $(DMA)_1$ $(TFA)_1$	4.39×10^{-10}	4.23×10^{-10}
$(SA)_2$ $(DMA)_2$+TFA	3.28×10^{-10}	3.16×10^{-10}
DMA+$(SA)_2$ $(DMA)_2$ $(TFA)_1$	4.90×10^{-10}	4.72×10^{-10}
$(DMA)_1$ $(TFA)_1$+TFA	3.26×10^{-10}	3.15×10^{-10}
TFA+$(SA)_1$ $(DMA)_1$ $(TFA)_1$	3.17×10^{-10}	3.05×10^{-10}
$(DMA)_1$ $(TFA)_2$+DMA	4.47×10^{-10}	4.31×10^{-10}
TFA+$(SA)_1$ $(DMA)_2$ $(TFA)_1$	3.52×10^{-10}	3.39×10^{-10}
DMA+$(SA)_1$ $(DMA)_2$ $(TFA)_2$	4.79×10^{-10}	4.62×10^{-10}
$(DMA)_1$ $(TFA)_2$+TFA	3.21×10^{-10}	3.10×10^{-10}
$(DMA)_2$ $(TFA)_2$+TFA	3.50×10^{-10}	3.38×10^{-10}
$(DMA)_2$ $(TFA)_3$+DMA	4.69×10^{-10}	4.52×10^{-10}
TFA+TFA	2.25×10^{-10}	2.17×10^{-10}
$(TFA)_2$+TFA	3.35×10^{-10}	3.23×10^{-10}
SA+TFA	1.97×10^{-10}	1.90×10^{-10}
$(SA)_1$ $(TFA)_1$+TFA	3.04×10^{-10}	2.93×10^{-10}
$(SA)_2$+TFA	2.80×10^{-10}	2.69×10^{-10}
SA+SA	1.70×10^{-10}	1.64×10^{-10}
$(SA)_2$+SA	2.55×10^{-10}	2.46×10^{-10}
$(TFA)_2$+SA	3.11×10^{-10}	3.00×10^{-10}
$(TFA)_3$+SA	3.38×10^{-10}	3.25×10^{-10}
$(SA)_1$ $(TFA)_1$+SA	2.79×10^{-10}	2.69×10^{-10}
$(SA)_1$ $(TFA)_2$+SA	4.54×10^{-10}	4.37×10^{-10}
$(SA)_2$ $(TFA)_1$+SA	3.86×10^{-10}	3.72×10^{-10}
$(DMA)_1$ $(TFA)_1$+SA	2.99×10^{-10}	2.88×10^{-10}
$(DMA)_1$ $(TFA)_2$+SA	2.99×10^{-10}	2.87×10^{-10}
$(DMA)_1$ $(TFA)_3$+SA	3.11×10^{-10}	3.00×10^{-10}
$(DMA)_2$ $(TFA)_2$+SA	3.28×10^{-10}	3.16×10^{-10}
$(DMA)_3$ $(TFA)_3$+SA	3.32×10^{-10}	3.20×10^{-10}
$(DMA)_2$ $(TFA)_3$+SA	3.06×10^{-10}	2.95×10^{-10}
$(SA)_1$ $(DMA)_1$ $(TFA)_1$+SA	2.93×10^{-10}	2.83×10^{-10}
$(SA)_2$ $(DMA)_1$ $(TFA)_1$+SA	3.95×10^{-10}	3.80×10^{-10}
$(SA)_1$ $(DMA)_2$ $(TFA)_1$+SA	3.29×10^{-10}	3.17×10^{-10}

$(SA)_1(DMA)_2(TFA)_2$+SA	3.14×10^{-10}	3.02×10^{-10}
$(SA)_2(DMA)_2(TFA)_1$+SA	3.22×10^{-10}	3.10×10^{-10}
$(SA)_1(DMA)_1(TFA)_2$+SA	4.31×10^{-10}	4.15×10^{-10}
$(SA)_2(DMA)_3(TFA)_1$+SA	3.41×10^{-10}	3.29×10^{-10}
$(SA)_1(DMA)_3(TFA)_2$+SA	3.41×10^{-10}	3.28×10^{-10}
$(TFA)_2$+DMA	4.60×10^{-10}	4.43×10^{-10}
$(TFA)_3$+DMA	5.10×10^{-10}	4.91×10^{-10}
$(SA)_1(TFA)_1$+DMA	4.14×10^{-10}	3.99×10^{-10}
$(SA)_1(TFA)_2$+DMA	6.75×10^{-10}	6.51×10^{-10}
$(SA)_2(TFA)_1$+DMA	5.76×10^{-10}	5.55×10^{-10}
$(DMA)_1(TFA)_3$+DMA	4.74×10^{-10}	4.56×10^{-10}
$(SA)_2(DMA)_1(TFA)_1$+DMA	5.93×10^{-10}	5.71×10^{-10}
$(SA)_1(DMA)_1(TFA)_2$+DMA	6.46×10^{-10}	6.23×10^{-10}
$(SA)_1(DMA)_1(TFA)_1$+$(SA)_1(DMA)_1$	3.80×10^{-10}	3.66×10^{-10}
$(SA)_1(DMA)_2(TFA)_1$+$(SA)_1(DMA)_1$	4.14×10^{-10}	3.99×10^{-10}

Table S3. Total evaporation coefficients ($\Sigma\gamma$, s^{-1}) for each cluster in the present study.

Clusters	$\Sigma\gamma$ (s^{-1})	
	280 K (Lu et al., 2020)	260 K
$(TFA)_2$	9.40×10^{4}	1.03×10^{4}
$(TFA)_3$	3.03×10^{11}	1.06×10^{11}
$(SA)_1$ $(TFA)_1$	4.00×10^{3}	3.63×10^{2}
$(SA)_1$ $(TFA)_2$	7.54×10^{3}	5.82×10^{2}
$(SA)_2$ $(TFA)_1$	2.90×10^{5}	2.37×10^{4}
$(DMA)_1$ $(TFA)_1$	1.32×10^{3}	1.16×10^{2}
$(DMA)_1$ $(TFA)_2$	6.41×10^{1}	3.05
$(DMA)_1$ $(TFA)_3$	2.59×10^{6}	2.12×10^{5}
$(DMA)_2$ $(TFA)_2$	9.53×10^{-3}	2.38×10^{-4}
$(SA)_1$ $(DMA)_1$ $(TFA)_1$	3.77	1.73×10^{-1}
$(SA)_2$ $(DMA)_1$ $(TFA)_1$	2.10×10^{5}	2.07×10^{4}
$(SA)_1$ $(DMA)_2$ $(TFA)_1$	1.73×10^{-4}	2.77×10^{-6}
$(SA)_1$ $(DMA)_2$ $(TFA)_2$	1.31×10^{4}	7.27×10^{2}
$(SA)_2$ $(DMA)_2$ $(TFA)_1$	1.16×10^{6}	8.23×10^{4}
$(SA)_1$ $(DMA)_1$ $(TFA)_2$	6.44×10^{2}	4.11×10^{1}
$(SA)_2$	1.96×10^{2}	1.56×10^{1}
$(SA)_3$	8.23×10^{4}	6.77×10^{3}
$(SA)_2$ $(DMA)_1$	2.75×10^{-7}	3.01×10^{-9}
$(SA)_2$ $(DMA)_2$	7.82×10^{-7}	1.07×10^{-8}
$(SA)_3$ $(DMA)_1$	1.25×10^{-1}	3.87×10^{-3}
$(SA)_3$ $(DMA)_2$	5.95×10^{-4}	1.24×10^{-5}
$(SA)_3$ $(DMA)_3$	1.61×10^{-6}	2.05×10^{-8}
$(SA)_1$ $(DMA)_1$	7.62×10^{-3}	2.47×10^{-4}
$(SA)_2$ $(DMA)_3$ $(TFA)_1$	2.31×10^{-4}	4.64×10^{-6}
$(SA)_1$ $(DMA)_3$ $(TFA)_2$	4.23×10^{-3}	1.04×10^{-4}
$(DMA)_3$ $(TFA)_3$	2.92×10^{1}	1.29
$(DMA)_2$ $(TFA)_3$	1.85×10^{3}	9.12×10^{1}

Table S4. Ratios ($\beta C/\Sigma\gamma$) between monomer molecule collisions and evaporation coefficients for each cluster involving TFA in the present study. [SA] = 1.0×10^{6} molecules cm^{-3}, [DMA] = 1.0×10^{7} molecules cm^{-3}, [TFA] = 1.0×10^{6} molecules cm^{-3}.

Clusters	$\beta C/\Sigma\gamma$	
	280 K	260 K
Collision with SA monomer: C = [SA]		
$(TFA)_2$	3.30×10^{-9}	2.90×10^{-8}
$(TFA)_3$	1.11×10^{-15}	3.06×10^{-15}
$(SA)_1(TFA)_1$	6.98×10^{-8}	7.41×10^{-7}
$(SA)_1(TFA)_2$	1.49×10^{-9}	1.54×10^{-8}
$(SA)_2(TFA)_1$	1.33×10^{-9}	1.57×10^{-8}
$(DMA)_1(TFA)_1$	2.26×10^{-7}	2.48×10^{-6}
$(DMA)_1(TFA)_2$	4.65×10^{-6}	9.44×10^{-5}
$(DMA)_1(TFA)_3$	1.20×10^{-10}	1.41×10^{-9}
$(DMA)_2(TFA)_2$	3.45×10^{-2}	1.33
$(DMA)_3(TFA)_3$	1.14×10^{-5}	2.48×10^{-4}
$(DMA)_2(TFA)_3$	1.66×10^{-7}	3.23×10^{-6}
$(SA)_1(DMA)_1(TFA)_1$	7.77×10^{-5}	1.64×10^{-3}
$(SA)_2(DMA)_1(TFA)_1$	1.88×10^{-9}	1.84×10^{-8}
$(SA)_1(DMA)_2(TFA)_1$	1.91	1.15×10^{2}
$(SA)_1(DMA)_2(TFA)_2$	2.39×10^{-8}	4.16×10^{-7}
$(SA)_2(DMA)_2(TFA)_1$	2.78×10^{-10}	3.77×10^{-9}
$(SA)_1(DMA)_1(TFA)_2$	6.69×10^{-7}	1.01×10^{-5}
$(SA)_2(DMA)_3(TFA)_1$	1.48	7.08×10^{1}
$(SA)_1(DMA)_3(TFA)_2$	8.05×10^{-2}	3.14
Collision with DMA monomer: C = [DMA]		
$(TFA)_2$	4.89×10^{-8}	4.30×10^{-7}
$(TFA)_3$	1.68×10^{-14}	4.61×10^{-14}
$(SA)_1(TFA)_1$	1.03×10^{-6}	1.10×10^{-5}
$(SA)_1(TFA)_2$	2.22×10^{-8}	2.29×10^{-7}
$(SA)_2(TFA)_1$	1.98×10^{-8}	2.34×10^{-7}
$(DMA)_1(TFA)_2$	6.98×10^{-5}	1.42×10^{-3}
$(DMA)_1(TFA)_3$	1.83×10^{-9}	2.15×10^{-8}
$(DMA)_2(TFA)_3$	2.54×10^{-6}	4.95×10^{-5}
$(SA)_1(DMA)_1(TFA)_1$	1.16×10^{-3}	2.45×10^{-2}
$(SA)_2(DMA)_1(TFA)_1$	2.82×10^{-8}	2.76×10^{-7}
$(SA)_1(DMA)_2(TFA)_2$	3.65×10^{-7}	6.35×10^{-6}
$(SA)_2(DMA)_2(TFA)_1$	4.24×10^{-9}	5.74×10^{-8}
$(SA)_1(DMA)_1(TFA)_2$	1.00×10^{-5}	1.52×10^{-4}
Collision with TFA monomer: C = [TFA]		
$(TFA)_2$	3.56×10^{-9}	3.13×10^{-8}
$(SA)_1(TFA)_1$	7.59×10^{-8}	8.07×10^{-7}
$(DMA)_1(TFA)_1$	2.46×10^{-7}	2.71×10^{-6}

$(DMA)_1(TFA)_2$	5.01×10^{-6}	1.02×10^{-4}
$(DMA)_2(TFA)_2$	3.68×10^{-2}	1.42
$(SA)_1(DMA)_1(TFA)_1$	8.39×10^{-5}	1.77×10^{-3}

Table S5. Relative hydrate distributions of the clusters at varying relative humidities (RHs) at 280 K.

Clusters	Relative hydrate distributions				
	RH=20%	RH=40%	RH=60%	RH=80%	RH=100%
SA	100%	99%	97%	94%	89%
$(SA)_1 (H_2O)_1$	0%	0%	0%	0%	0%
$(SA)_1 (H_2O)_2$	0%	0%	0%	0%	0%
$(SA)_1 (H_2O)_3$	0%	1%	3%	6%	11%
TFA	90%	80%	72%	65%	59%
$(TFA)_1 (H_2O)_1$	10%	18%	24%	29%	33%
$(TFA)_1 (H_2O)_2$	0%	2%	4%	6%	8%
$(TFA)_1 (H_2O)_3$	0%	0%	0%	0%	0%
$(SA)_1 (TFA)_1$	72%	52%	39%	30%	23%
$(SA)_1 (TFA)_1 (H_2O)_1$	23%	34%	39%	39%	38%
$(SA)_1 (TFA)_1 (H_2O)_2$	5%	14%	22%	30%	37%
$(SA)_1 (TFA)_1 (H_2O)_3$	0%	0%	0%	1%	2%
$(DMA)_1 (TFA)_1$	12%	6%	4%	3%	2%
$(DMA)_1 (TFA)_1 (H_2O)_1$	87%	92%	92%	92%	92%
$(DMA)_1 (TFA)_1 (H_2O)_2$	1%	2%	4%	5%	6%
$(SA)_1 (DMA)_1 (TFA)_1$	87%	76%	68%	61%	56%
$(SA)_1 (DMA)_1 (TFA)_1 (H_2O)_1$	13%	23%	31%	37%	42%
$(SA)_1 (DMA)_1 (TFA)_1 (H_2O)_2$	0%	1%	1%	2%	2%
$(SA)_1 (DMA)_2 (TFA)_1$	95%	90%	85%	81%	77%
$(SA)_1 (DMA)_2 (TFA)_1 (H_2O)_1$	5%	10%	14%	17%	20%
$(SA)_1 (DMA)_2 (TFA)_1 (H_2O)_2$	0%	0%	1%	2%	3%

Table S6. Relative hydrate distributions of the clusters at varying relative humidities (RHs) at 260 K.

Clusters	Relative hydrate distributions				
	RH=20%	RH=40%	RH=60%	RH=80%	RH=100%
SA	100%	99%	96%	92%	85%
$(SA)_1\,(H_2O)_1$	0%	0%	0%	0%	0%
$(SA)_1\,(H_2O)_2$	0%	0%	0%	0%	0%
$(SA)_1\,(H_2O)_3$	0%	1%	4%	8%	15%
TFA	90%	80%	72%	65%	59%
$(TFA)_1\,(H_2O)_1$	10%	18%	24%	29%	33%
$(TFA)_1\,(H_2O)_2$	0%	2%	4%	6%	8%
$(TFA)_1\,(H_2O)_3$	0%	0%	0%	0%	0%
$(SA)_1\,(TFA)_1$	68%	47%	34%	25%	19%
$(SA)_1\,(TFA)_1\,(H_2O)_1$	26%	36%	39%	39%	37%
$(SA)_1\,(TFA)_1\,(H_2O)_2$	6%	17%	27%	35%	42%
$(SA)_1\,(TFA)_1\,(H_2O)_3$	0%	0%	0%	1%	1%
$(DMA)_1\,(TFA)_1$	8%	4%	3%	2%	1%
$(DMA)_1\,(TFA)_1\,(H_2O)_1$	91%	94%	94%	94%	94%
$(DMA)_1\,(TFA)_1\,(H_2O)_2$	1%	2%	3%	4%	5%
$(SA)_1\,(DMA)_1\,(TFA)_1$	85%	74%	65%	58%	53%
$(SA)_1\,(DMA)_1\,(TFA)_1\,(H_2O)_1$	15%	25%	34%	40%	45%
$(SA)_1\,(DMA)_1\,(TFA)_1\,(H_2O)_2$	0%	1%	1%	2%	2%
$(SA)_1\,(DMA)_2\,(TFA)_1$	95%	90%	85%	80%	76%
$(SA)_1\,(DMA)_2\,(TFA)_1\,(H_2O)_1$	5%	10%	14%	18%	21%
$(SA)_1\,(DMA)_2\,(TFA)_1\,(H_2O)_2$	0%	0%	1%	2%	3%

Table S7. Ratios ($\beta C/\Sigma\gamma$) between monomer molecule collisions and evaporation coefficients for the key clusters involving TFA in the present study at different relative humidities (RHs) and different temperatures. C = 1.0×10^{8} molecules cm^{-3}.

Clusters	$\beta C/\Sigma\gamma$					
	RH=0%	RH=20%	RH=40%	RH=60%	RH=80%	RH=100%
	280 K					
$(SA)_1(TFA)_1$	1.03×10^{-5}	8.75×10^{-6}	7.35×10^{-6}	6.28×10^{-6}	5.48×10^{-6}	4.86×10^{-6}
$(DMA)_1(TFA)_1$	2.26×10^{-5}	1.77×10^{-4}	3.08×10^{-4}	4.16×10^{-4}	5.01×10^{-4}	5.67×10^{-4}
$(SA)_1(DMA)_1(TFA)_1$	1.16×10^{-2}	1.22×10^{-2}	1.25×10^{-2}	1.27×10^{-2}	1.27×10^{-2}	1.27×10^{-2}
$(SA)_1(DMA)_2(TFA)_1$	1.91×10^{2}	1.77×10^{2}	1.66×10^{2}	1.57×10^{2}	1.49×10^{2}	1.43×10^{2}
	260 K					
$(SA)_1(TFA)_1$	1.10×10^{-4}	9.41×10^{-5}	8.02×10^{-5}	6.94×10^{-5}	6.11×10^{-5}	5.47×10^{-5}
$(DMA)_1(TFA)_1$	2.48×10^{-4}	2.86×10^{-3}	5.08×10^{-3}	6.87×10^{-3}	8.28×10^{-3}	9.36×10^{-3}
$(SA)_1(DMA)_1(TFA)_1$	2.45×10^{-1}	2.62×10^{-1}	2.73×10^{-1}	2.79×10^{-1}	2.83×10^{-1}	2.84×10^{-1}
$(SA)_1(DMA)_2(TFA)_1$	1.15×10^{4}	1.05×10^{4}	9.77×10^{3}	9.14×10^{3}	8.61×10^{3}	8.18×10^{3}

Table S8. Cartesian coordinates of the most stable hydrated clusters in the present study at the M06-2X/6-311++G(3df,3pd) level of theory.

$(SA)_1 (H_2O)_1$

Atoms	X	Y	Z
S	0.574277	-0.075955	0.121591
O	-0.215605	0.288808	1.248460
O	1.737700	-0.870444	0.247437
O	-0.338533	-0.732296	-0.941762
O	0.979121	1.291265	-0.552739
H	-1.289890	-0.469017	-0.773800
H	1.736148	1.150356	-1.135650
O	-2.661959	0.105985	-0.109758
H	-2.259274	0.361340	0.731812
H	-3.381199	-0.493949	0.099086

$(SA)_1 (H_2O)_2$

Atoms	X	Y	Z
S	0.986992	-0.150388	-0.099481
O	0.432583	1.170078	-0.640315
O	1.134685	0.126299	1.452466
O	-0.005244	-1.171702	-0.181044
O	2.277203	-0.349376	-0.644188
H	-0.562404	1.301754	-0.403772
H	1.971276	0.580502	1.615265
H	-2.423091	0.553179	-0.021326
O	-2.007743	1.446196	-0.076036
H	-2.204764	1.896542	0.747917
H	-1.846181	-1.395479	0.029141
O	-2.764155	-1.093281	0.112361
H	-3.265331	-1.555999	-0.561485

$(SA)_1 (H_2O)_3$

Atoms	X	Y	Z
O	-1.506990	-1.371735	-1.163177
H	-1.773950	-0.567008	-1.621186
H	-1.971831	-1.328300	-0.314030
S	1.244190	0.029228	0.161933
O	2.620912	0.184108	0.414141
O	1.056128	-1.232150	-0.708744

O	0.784181	1.192212	-0.765548
O	0.322056	-0.028098	1.266286
H	0.082261	-1.372103	-0.911487
H	-0.192657	1.341878	-0.695079
O	-1.876525	1.466547	-0.654087
H	-2.225041	1.049168	0.154071
H	-2.243910	2.351570	-0.702420
O	-2.325355	-0.349927	1.336061
H	-2.804143	-0.522403	2.148080
H	-1.373038	-0.308106	1.551677

$(TFA)_1\ (H_2O)_1$

Atoms	X	Y	Z
O	3.362699	-0.069069	-0.051778
H	3.011637	0.832051	0.014031
H	4.125166	-0.141963	0.530676
C	0.333949	0.138464	-0.025218
O	0.874293	1.217382	-0.027412
O	0.909025	-1.041694	-0.032221
H	1.895390	-0.910166	-0.028620
C	-1.202835	-0.003071	0.004431
F	-1.583079	-0.615283	1.132693
F	-1.629468	-0.733550	-1.030289
F	-1.786010	1.188251	-0.046857

$(TFA)_1\ (H_2O)_2$

Atoms	X	Y	Z
H	-2.363187	1.452463	-0.054670
O	-3.313308	1.240393	-0.060936
H	-3.743089	1.802363	0.590710
H	-3.183163	-0.447357	0.032091
H	-3.222819	-1.881344	-0.632125
O	-2.797748	-1.356898	0.054197
C	0.139960	0.005991	0.000751
O	-0.523776	1.019907	-0.038579
O	-0.269253	-1.224491	0.058303
H	-1.296106	-1.289418	0.057130
C	1.683582	0.076656	-0.004439
F	2.182969	-0.659920	-1.002578
F	2.171107	-0.398651	1.148131
F	2.101456	1.329252	-0.153874

$(TFA)_1\ (H_2O)_3$

Atoms	X	Y	Z
H	-1.745831	1.668112	-0.275550
O	-2.651031	2.006936	-0.351570
H	-2.625124	2.915354	-0.048644
H	-2.845443	-1.289027	0.040340
H	-2.419424	-2.329993	-1.044649
O	-2.135400	-1.924108	-0.223709
C	0.620692	-0.087283	-0.028386
O	-0.122011	0.858538	-0.042894
O	0.324986	-1.343761	-0.079687
H	-0.681481	-1.514816	-0.141124
C	2.152349	0.113896	0.056531
F	2.738731	-0.370895	-1.034687
F	2.647816	-0.523804	1.112156
F	2.456020	1.394967	0.160798
O	-4.028624	-0.181646	0.404797
H	-4.346195	-0.139677	1.307992
H	-3.661203	0.700298	0.192867

$(SA)_1\ (TFA)_1\ (H_2O)_1$

Atoms	X	Y	Z
C	1.740147	-0.060491	-0.076518
O	1.029422	0.615663	-0.779728
O	1.411654	-1.090282	0.633207
H	0.439288	-1.292383	0.540432
C	3.247734	0.254390	0.054113
F	3.959442	-0.755137	-0.433482
F	3.573254	0.421464	1.329264
F	3.553181	1.350586	-0.610669
S	-2.110935	-0.516021	-0.126415
O	-3.414932	-0.924559	-0.511050
O	-1.154261	-1.505286	0.277396
O	-2.196242	0.497585	1.037383
O	-1.524253	0.298837	-1.299437
H	-3.050500	1.016221	0.952365
H	-0.549434	0.458204	-1.153322
O	-4.558925	1.458392	0.547073
H	-4.776658	0.705609	-0.018806
H	-4.727621	2.250282	0.031612

$(SA)_1 (TFA)_1 (H_2O)_2$

Atoms	X	Y	Z
S	1.665172	-0.721455	-0.012451
O	2.985776	-1.190736	0.234451
O	0.675877	-1.682138	-0.413534
O	1.653962	0.382581	-1.068227
O	1.195849	-0.022349	1.286405
H	2.396454	1.092043	-0.894464
H	0.223414	0.185551	1.218072
O	3.483939	2.024975	-0.618749
H	4.255516	1.521016	-0.262914
H	3.279295	2.716191	0.015075
O	5.286734	0.378848	0.403672
H	4.662400	-0.361192	0.455270
H	6.060791	0.053152	-0.058947
C	-2.113785	-0.102309	0.120697
O	-1.369774	0.442463	0.899504
O	-1.842888	-1.058407	-0.705776
H	-0.882766	-1.331250	-0.638928
C	-3.594951	0.326537	0.016461
F	-4.388484	-0.712102	0.246093
F	-3.846442	0.785124	-1.203932
F	-3.869655	1.273699	0.891300

$(SA)_1 (TFA)_1 (H_2O)_3$

Atoms	X	Y	Z
S	-1.835354	0.418045	-0.544635
O	-3.017216	-0.040423	-1.196473
O	-0.659448	0.626414	-1.346608
O	-1.416089	-0.556034	0.564642
O	-2.186072	1.724279	0.173105
H	-2.229160	-0.935004	1.079103
H	-1.328371	2.207843	0.473082
O	-3.424194	-1.495170	1.746353
H	-4.174023	-1.531702	1.104608
H	-3.739818	-1.024952	2.520786
O	-5.242764	-1.348471	-0.177002
H	-4.641108	-0.853171	-0.754547
H	-5.547167	-2.103922	-0.683027
C	2.356454	0.089545	-0.161069
O	2.085541	1.109932	0.415460
O	1.629739	-0.574137	-1.002407

H	0.742770	-0.136287	-1.139376
C	3.710573	-0.616245	0.081107
F	4.276822	-0.964516	-1.066093
F	3.513580	-1.716825	0.802414
F	4.538499	0.174187	0.738743
O	-0.012733	2.816246	0.823086
H	0.138894	3.087922	1.730675
H	0.727274	2.224052	0.585800

$(DMA)_1$ $(TFA)_1$ $(H_2O)_1$

Atoms	X	Y	Z
H	-1.361042	-0.630567	-0.329075
O	-2.098890	2.282775	-0.404316
H	-1.169200	2.035559	-0.205440
H	-2.071265	3.108162	-0.888350
C	0.708817	0.107761	-0.099332
O	0.296568	1.228835	0.197598
O	0.058897	-0.905533	-0.447083
C	2.238137	-0.139235	-0.001052
F	2.700021	-0.747844	-1.091943
F	2.499831	-0.930899	1.044698
F	2.925415	0.982628	0.160662
N	-2.422381	-0.416110	-0.086683
H	-2.619900	0.509038	-0.488809
C	-2.485844	-0.295752	1.382678
H	-2.176460	-1.241482	1.820915
H	-1.800466	0.491317	1.684853
H	-3.498498	-0.050868	1.693989
O	-3.296421	-1.460613	-0.639700
H	-3.191837	-1.478234	-1.720381
H	-2.990406	-2.422005	-0.235096
H	-4.332398	-1.264697	-0.371729

$(DMA)_1$ $(TFA)_1$ $(H_2O)_2$

Atoms	X	Y	Z
H	-1.006079	-0.898345	-0.191929
O	-3.573128	1.093086	-1.049159
H	-2.983335	1.749976	-0.620828
H	-3.684941	1.378299	-1.956978
C	1.036992	0.066932	-0.023389
O	0.549472	1.130317	0.359707

O	0.462761	-0.986588	-0.373269
C	2.586245	-0.021886	-0.083020
F	2.992559	-0.316115	-1.319563
F	3.029174	-0.987062	0.726992
F	3.175201	1.109824	0.277494
N	-2.071647	-0.998853	0.040769
H	-2.610941	-0.322880	-0.524342
C	-2.254449	-0.669697	1.467865
H	-1.600418	-1.308057	2.056940
H	-1.988958	0.372115	1.621581
H	-3.291689	-0.834944	1.750581
C	-2.462569	-2.378903	-0.293507
H	-2.280379	-2.553158	-1.349675
H	-1.852580	-3.062791	0.291183
H	-3.514698	-2.533295	-0.066404
O	-1.747047	2.440993	0.353316
H	-1.551746	3.369527	0.483594
H	-0.878897	1.974563	0.324126

$(SA)_1$ $(DMA)_1$ $(TFA)_1$ $(H_2O)_1$

Atoms	X	Y	Z
S	1.034353	-0.956744	0.107501
O	2.336821	-1.554530	0.274425
O	0.044751	-1.830524	-0.500491
O	1.090386	0.344758	-0.562713
O	0.558809	-0.674037	1.576180
H	2.553330	0.931518	-0.430724
H	-0.375006	-0.361564	1.537260
C	-2.608037	-0.122899	0.088614
O	-1.975618	0.136444	1.083232
O	-2.275143	-0.876389	-0.903212
H	-1.341506	-1.269827	-0.774286
C	-4.020699	0.472166	-0.112886
F	-4.927411	-0.500535	-0.131919
F	-4.090005	1.126855	-1.267606
F	-4.325594	1.309474	0.861730
N	3.541588	1.248997	-0.200254
H	4.161830	0.512597	-0.579043
C	3.662081	1.246204	1.275086
H	2.933885	1.941513	1.684081
H	3.449698	0.241897	1.629588
H	4.668870	1.544548	1.554988
C	3.813993	2.558426	-0.818454
H	3.695366	2.476022	-1.894451

H	3.104814	3.283870	-0.429339
H	4.827229	2.870544	-0.578944
O	4.823467	-1.137414	-0.709143
H	5.174213	-1.672463	-1.422028
H	3.981803	-1.545702	-0.435565

$(SA)_1 (DMA)_1 (TFA)_1 (H_2O)_2$

Atoms	X	Y	Z
H	-0.080007	-0.419317	1.240587
S	-2.015823	-0.670788	-0.135703
O	-3.090703	0.293975	-0.159460
O	-1.421949	-0.796430	1.219289
O	-0.963591	-0.410056	-1.108430
O	-2.659618	-2.036520	-0.458734
H	-0.219115	1.243525	-0.650434
H	-1.944966	-2.744951	-0.409905
N	-0.431023	2.220308	-0.389006
H	-0.985086	2.154117	0.497300
O	-2.211616	2.118407	1.703286
H	-2.777708	1.503098	1.199760
H	-1.962752	1.618067	2.485390
O	-0.657324	-3.667654	-0.274406
H	-0.337722	-3.983011	-1.123021
H	0.060250	-3.109994	0.072353
C	1.680442	-0.836609	0.511218
O	1.525004	-2.008004	0.298224
O	0.895257	-0.010079	1.122044
C	2.934497	-0.107072	-0.020026
F	3.475713	0.678497	0.907292
F	2.566594	0.679169	-1.044104
F	3.851497	-0.945849	-0.451329
C	0.793544	3.010180	-0.152221
H	1.381867	2.517530	0.615449
H	1.365009	3.068380	-1.073802
H	0.505829	4.006536	0.171968
C	-1.324994	2.775882	-1.431283
H	-0.776761	2.840975	-2.366849
H	-2.171916	2.103519	-1.534325
H	-1.655435	3.762230	-1.117548

$(SA)_1 (DMA)_2 (TFA)_1 (H_2O)_1$

Atoms	X	Y	Z
S	1.317575	-1.578670	0.034019
O	0.602600	-2.827653	-0.209331
O	2.729985	-1.753850	0.289467
O	0.631230	-0.747733	1.013227
O	1.279068	-0.754265	-1.316709
H	-0.981677	-2.508475	0.023221
C	-1.119567	1.291251	-0.575722
O	-0.178033	2.021477	-0.301340
O	-1.141006	0.215848	-1.225427
H	0.362202	-0.351713	-1.411671
C	-2.527188	1.723191	-0.076991
F	-3.188461	0.662345	0.431858
F	-3.263536	2.191470	-1.083601
F	-2.484914	2.648185	0.866889
N	-1.927133	-2.059672	0.164883
H	-1.782659	-1.102421	-0.207388
C	-2.228207	-2.015950	1.609083
H	-2.284308	-3.034702	1.984076
H	-1.421423	-1.480650	2.099856
H	-3.176448	-1.508831	1.762474
C	-2.955880	-2.730413	-0.648409
H	-2.647289	-2.707314	-1.689073
H	-3.061008	-3.759431	-0.315227
H	-3.900081	-2.204393	-0.532557
H	1.449604	1.270086	0.280764
N	2.448932	1.511785	0.378972
H	2.983863	0.914422	-0.283666
C	2.916511	1.223035	1.751289
H	2.329844	1.812998	2.450027
H	2.777870	0.164372	1.941559
H	3.967707	1.490841	1.820894
C	2.610701	2.923039	-0.022036
H	2.211958	3.043982	-1.023493
H	2.044866	3.548317	0.662101
H	3.666637	3.177723	0.005951
O	4.220637	-0.000046	-1.160360
H	3.948293	-0.103261	-2.075381
H	3.876384	-0.800751	-0.719571

$(SA)_1 (DMA)_2 (TFA)_1 (H_2O)_2$

Atoms	X	Y	Z
S	1.446453	1.638159	-0.268362
O	1.240698	0.838327	0.940887
O	2.769707	2.205594	-0.357425
O	0.378835	2.609877	-0.469197
O	1.365517	0.620200	-1.463670
H	-1.139022	2.152579	-0.190917
H	0.422298	0.238972	-1.480437
C	-1.033881	-1.281006	-0.512056
O	-1.026730	-0.268888	-1.241654
O	-0.095892	-1.946558	-0.075005
H	1.630107	-1.214036	0.049678
C	-2.455999	-1.777552	-0.123916
F	-2.467174	-2.405864	1.051152
F	-2.921362	-2.625047	-1.040387
F	-3.332415	-0.765977	-0.037499
N	2.586399	-1.590610	0.130913
H	3.243170	-0.802590	0.308761
C	2.924339	-2.241782	-1.148507
H	2.187122	-3.016133	-1.342339
H	2.887426	-1.488354	-1.929165
H	3.921695	-2.667879	-1.079425
C	2.594309	-2.504002	1.287419
H	2.330340	-1.933920	2.173611
H	1.854380	-3.280720	1.118133
H	3.586184	-2.934221	1.399321
N	-2.124595	2.001937	0.156991
H	-2.113395	1.059611	0.567517
C	-3.076734	2.083548	-0.965793
H	-3.002113	3.071775	-1.411934
H	-2.811448	1.317578	-1.686267
H	-4.084370	1.917131	-0.593462
C	-2.363030	2.982652	1.232989
H	-1.617803	2.825923	2.006326
H	-2.266097	3.983181	0.820703
H	-3.359606	2.837601	1.641675
O	4.517788	0.356294	0.466714
H	4.834071	0.600201	1.338336
H	3.997135	1.131068	0.154912
O	-1.201170	0.114132	1.971966
H	-1.221325	-0.822975	2.180295
H	-0.290100	0.304388	1.672010

Table S9. Predicted concentrations of DMA (molecules cm^{-3}) by the global chemistry-transport model, temperatures (T, K), particle formation rates of SA-DMA-TFA system ($J_{SA\text{-}DMA\text{-}TFA}$, cm^{-3} s^{-1}) and SA-DMA system ($J_{SA\text{-}DMA}$, cm^{-3} s^{-1}) and the enhancement ($J_{SA\text{-}DMA\text{-}TFA}/J_{SA\text{-}DMA}$) on particle formation rates by TFA.

Cities	Month	[DMA] (molecules cm^{-3})	T (K)	$J_{SA\text{-}DMA\text{-}TFA}$ (cm^{-3} s^{-1})	$J_{SA\text{-}DMA}$ (cm^{-3} s^{-1})	Enhancement
Beijing	1	9.37×10^{7}	265	5.10×10^{2}	3.84×10^{1}	13.27
	2	8.63×10^{7}	269	2.99×10^{2}	3.85×10^{1}	7.75
	3	7.49×10^{7}	275	1.28×10^{2}	3.79×10^{1}	3.39
	4	6.26×10^{7}	284	5.01×10^{1}	3.39×10^{1}	1.48
	5	4.25×10^{7}	291	2.29×10^{1}	2.11×10^{1}	1.08
	6	4.45×10^{7}	296	1.56×10^{1}	1.51×10^{1}	1.03
	7	3.09×10^{7}	298	8.30	8.19	1.01
	8	5.12×10^{7}	296	1.78×10^{1}	1.71×10^{1}	1.04
	9	6.01×10^{7}	291	3.04×10^{1}	2.67×10^{1}	1.14
	10	7.26×10^{7}	283	6.10×10^{1}	3.62×10^{1}	1.69
	11	1.52×10^{8}	274	3.22×10^{2}	4.28×10^{1}	7.53
	12	1.04×10^{8}	267	4.67×10^{2}	3.95×10^{1}	11.85
Shanghai	1	5.21×10^{7}	281	5.29×10^{1}	3.29×10^{1}	1.61
	2	5.28×10^{7}	282	4.96×10^{1}	3.27×10^{1}	1.52
	3	2.56×10^{7}	284	2.30×10^{1}	2.02×10^{1}	1.14
	4	2.13×10^{7}	287	1.56×10^{1}	1.47×10^{1}	1.06
	5	2.10×10^{7}	291	1.10×10^{1}	1.06×10^{1}	1.03
	6	2.51×10^{7}	295	9.14	8.98	1.02
	7	2.16×10^{7}	299	4.48	4.45	1.01
	8	2.49×10^{7}	299	5.51	5.47	1.01
	9	2.89×10^{7}	296	9.66	9.49	1.02
	10	3.58×10^{7}	290	2.12×10^{1}	1.97×10^{1}	1.08
	11	6.07×10^{7}	285	4.58×10^{1}	3.28×10^{1}	1.40
	12	6.30×10^{7}	280	6.82×10^{1}	3.55×10^{1}	1.92
Los Angeles	1	2.87×10^{7}	275	4.33×10^{1}	2.61×10^{1}	1.66
	2	2.99×10^{7}	277	4.00×10^{1}	2.63×10^{1}	1.52
	3	2.66×10^{7}	282	2.67×10^{1}	2.23×10^{1}	1.20
	4	4.70×10^{7}	288	3.07×10^{1}	2.64×10^{1}	1.16
	5	2.49×10^{7}	294	1.00×10^{1}	9.83	1.02
	6	6.42×10^{7}	298	1.82×10^{1}	1.76×10^{1}	1.04
	7	2.81×10^{7}	301	4.98	4.95	1.01
	8	4.36×10^{7}	301	8.81	8.72	1.01
	9	2.80×10^{7}	296	9.31	9.15	1.02
	10	4.16×10^{7}	291	2.25×10^{1}	2.08×10^{1}	1.08
	11	3.56×10^{7}	284	3.12×10^{1}	2.56×10^{1}	1.22
	12	7.14×10^{7}	278	9.23×10^{1}	3.73×10^{1}	2.48
New Delhi	1	1.42×10^{8}	287	7.70×10^{1}	4.12×10^{1}	1.87
	2	1.36×10^{8}	291	5.43×10^{1}	3.82×10^{1}	1.42
	3	8.49×10^{7}	297	2.52×10^{1}	2.36×10^{1}	1.07

4	9.66×10^{7}	304	1.46×10^{1}	1.44×10^{1}	1.02
5	8.40×10^{7}	308	7.85	7.81	1.01
6	6.04×10^{7}	309	4.56	4.54	1.00
7	6.15×10^{7}	306	7.05	7.01	1.01
8	6.08×10^{7}	303	1.01×10^{1}	1.00×10^{1}	1.01
9	5.75×10^{7}	301	1.20×10^{1}	1.19×10^{1}	1.02
10	9.52×10^{7}	298	2.52×10^{1}	2.37×10^{1}	1.07
11	1.15×10^{8}	293	4.18×10^{1}	3.40×10^{1}	1.23
12	1.24×10^{8}	289	5.94×10^{1}	3.87×10^{1}	1.53

Table S10. Gibbs free formation energies (ΔG, kcal/mol) of clusters at different temperatures in different months of the studied cities shown in Table S9.

Clusters	ΔG (kcal/mol)				
	265 K	267 K	269 K	270 K	274 K
$(SA)_1 (DMA)_1$	-15.80	-15.73	-15.66	-15.63	-15.49
$(SA)_1 (DMA)_1 (TFA)_1$	-28.30	-28.16	-28.01	-27.94	-27.65
$(SA)_1 (DMA)_2 (TFA)_1$	-38.01	-46.45	-46.21	-46.09	-45.62
$(SA)_1 (TFA)_1$	-8.35	-8.28	-8.21	-8.18	-8.04
$(SA)_1 (DMA)_1 (TFA)_2$	-38.01	-37.78	-37.56	-37.44	-36.99
$(SA)_1 (DMA)_2 (TFA)_2$	-54.91	-54.57	-54.24	-54.07	-53.41
$(SA)_1 (DMA)_3 (TFA)_2$	-71.48	-71.07	-70.65	-70.45	-69.62
$(SA)_1 (TFA)_2$	-14.65	-14.49	-14.34	-14.26	-13.95
$(DMA)_1 (TFA)_1$	-9.15	-9.08	-9.02	-8.98	-8.85
$(SA)_2 (DMA)_1$	-37.47	-37.31	-37.16	-37.08	-36.77
$(SA)_2 (DMA)_1 (TFA)_1$	-43.89	-43.65	-43.42	-43.30	-42.83
$(SA)_2 (DMA)_2 (TFA)_1$	-64.41	-64.08	-63.74	-63.58	-62.91
$(SA)_2 (DMA)_3 (TFA)_1$	-82.61	-82.19	-81.78	-81.57	-80.75
$(SA)_2 (TFA)_1$	-15.87	-15.71	-15.55	-15.47	-15.15
$(SA)_2 (DMA)_2$	-58.68	-58.45	-58.22	-58.11	-57.64
$(SA)_2$	-9.55	-9.48	-9.41	-9.38	-9.24
$(DMA)_1 (TFA)_2$	-20.19	-20.04	-19.89	-19.82	-19.52
$(DMA)_2 (TFA)_2$	-36.29	-36.06	-35.83	-35.71	-35.25
$(TFA)_2$	-6.32	-6.25	-6.17	-6.14	-5.99
$(SA)_3 (DMA)_1$	-51.84	-51.61	-51.37	-51.25	-50.77
$(SA)_3 (DMA)_2$	-76.12	-75.81	-75.49	-75.34	-74.71
$(SA)_3 (DMA)_3$	-97.19	-96.80	-96.40	-96.20	-95.42
$(SA)_3$	-16.48	-16.33	-16.17	-16.09	-15.79
$(DMA)_1 (TFA)_3$	-24.38	-24.12	-23.85	-23.72	-23.19
$(DMA)_2 (TFA)_3$	-45.59	-45.27	-44.93	-44.77	-44.11
$(DMA)_3 (TFA)_3$	-57.27	-56.86	-56.45	-56.24	-55.41
$(TFA)_3$	-4.88	-4.73	-4.59	-4.52	-4.23

Table S10. Continued: Gibbs free formation energies (Δ*G*, kcal/mol) of clusters at different temperatures in different months of the studied cities shown in Table S9.

Clusters	Δ*G* (kcal/mol)				
	275 K	277 K	278 K	280 K	281 K
$(SA)_1 (DMA)_1$	-15.46	-15.39	-15.35	-15.28	-15.25
$(SA)_1 (DMA)_1 (TFA)_1$	-27.57	-27.43	-27.36	-27.21	-27.14
$(SA)_1 (DMA)_2 (TFA)_1$	-45.51	-45.27	-45.15	-44.92	-44.80
$(SA)_1 (TFA)_1$	-8.00	-7.93	-7.90	-7.83	-7.79
$(SA)_1 (DMA)_1 (TFA)_2$	-36.88	-36.66	-36.54	-36.32	-36.21
$(SA)_1 (DMA)_2 (TFA)_2$	-53.24	-52.91	-52.74	-52.41	-52.24
$(SA)_1 (DMA)_3 (TFA)_2$	-69.42	-69.01	-68.80	-68.39	-68.18
$(SA)_1 (TFA)_2$	-13.88	-13.72	-13.65	-13.49	-13.41
$(DMA)_1 (TFA)_1$	-8.82	-8.75	-8.72	-8.65	-8.62
$(SA)_2 (DMA)_1$	-36.69	-36.54	-36.46	-36.30	-36.22
$(SA)_2 (DMA)_1 (TFA)_1$	-42.72	-42.48	-42.37	-42.13	-42.01
$(SA)_2 (DMA)_2 (TFA)_1$	-62.74	-62.41	-62.24	-61.91	-61.74
$(SA)_2 (DMA)_3 (TFA)_1$	-80.55	-80.13	-79.93	-79.52	-79.31
$(SA)_2 (TFA)_1$	-15.08	-14.92	-14.84	-14.68	-14.60
$(SA)_2 (DMA)_2$	-57.53	-57.30	-57.18	-56.95	-56.84
$(SA)_2$	-9.21	-9.14	-9.11	-9.04	-9.01
$(DMA)_1 (TFA)_2$	-19.44	-19.29	-19.21	-19.06	-18.99
$(DMA)_2 (TFA)_2$	-35.13	-34.90	-34.79	-34.55	-34.44
$(TFA)_2$	-5.95	-5.87	-5.84	-5.76	-5.72
$(SA)_3 (DMA)_1$	-50.65	-50.41	-50.30	-50.06	-49.94
$(SA)_3 (DMA)_2$	-74.56	-74.25	-74.09	-73.78	-73.62
$(SA)_3 (DMA)_3$	-95.22	-94.83	-94.63	-94.24	-94.04
$(SA)_3$	-15.71	-15.56	-15.48	-15.33	-15.25
$(DMA)_1 (TFA)_3$	-23.05	-22.79	-22.65	-22.39	-22.25
$(DMA)_2 (TFA)_3$	-43.95	-43.62	-43.46	-43.13	-42.96
$(DMA)_3 (TFA)_3$	-55.21	-54.80	-54.59	-54.18	-53.97
$(TFA)_3$	-4.15	-4.01	-3.93	-3.79	-3.72

Table S10. Continued: Gibbs free formation energies (ΔG, kcal/mol) of clusters at different temperatures in different months of the studied cities shown in Table S9.

Clusters	ΔG (kcal/mol)				
	282 K	283 K	284 K	285 K	286 K
$(SA)_1 (DMA)_1$	-15.22	-15.18	-15.15	-15.11	-15.08
$(SA)_1 (DMA)_1 (TFA)_1$	-27.06	-26.99	-26.92	-26.85	-26.77
$(SA)_1 (DMA)_2 (TFA)_1$	-44.68	-44.57	-44.45	-44.33	-44.22
$(SA)_1 (TFA)_1$	-7.76	-7.72	-7.69	-7.65	-7.62
$(SA)_1 (DMA)_1 (TFA)_2$	-36.09	-35.98	-35.87	-35.76	-35.64
$(SA)_1 (DMA)_2 (TFA)_2$	-52.07	-51.91	-51.74	-51.57	-51.41
$(SA)_1 (DMA)_3 (TFA)_2$	-67.98	-67.77	-67.57	-67.36	-67.15
$(SA)_1 (TFA)_2$	-13.34	-13.26	-13.18	-13.11	-13.03
$(DMA)_1 (TFA)_1$	-8.59	-8.56	-8.52	-8.49	-8.46
$(SA)_2 (DMA)_1$	-36.15	-36.07	-35.99	-35.91	-35.84
$(SA)_2 (DMA)_1 (TFA)_1$	-41.90	-41.78	-41.66	-41.55	-41.43
$(SA)_2 (DMA)_2 (TFA)_1$	-61.57	-61.41	-61.24	-61.07	-60.91
$(SA)_2 (DMA)_3 (TFA)_1$	-79.10	-78.90	-78.69	-78.48	-78.28
$(SA)_2 (TFA)_1$	-14.52	-14.44	-14.36	-14.28	-14.20
$(SA)_2 (DMA)_2$	-56.72	-56.61	-56.49	-56.37	-56.26
$(SA)_2$	-8.98	-8.94	-8.91	-8.87	-8.84
$(DMA)_1 (TFA)_2$	-18.91	-18.84	-18.76	-18.69	-18.61
$(DMA)_2 (TFA)_2$	-34.32	-34.21	-34.09	-33.97	-33.86
$(TFA)_2$	-5.69	-5.65	-5.61	-5.57	-5.54
$(SA)_3 (DMA)_1$	-49.82	-49.70	-49.58	-49.46	-49.34
$(SA)_3 (DMA)_2$	-73.47	-73.31	-73.15	-73.00	-72.84
$(SA)_3 (DMA)_3$	-93.84	-93.65	-93.45	-93.25	-93.06
$(SA)_3$	-15.17	-15.09	-15.02	-14.94	-14.86
$(DMA)_1 (TFA)_3$	-22.12	-21.99	-21.86	-21.72	-21.59
$(DMA)_2 (TFA)_3$	-42.80	-42.64	-42.47	-42.31	-42.14
$(DMA)_3 (TFA)_3$	-53.76	-53.56	-53.35	-53.15	-52.94
$(TFA)_3$	-3.64	-3.57	-3.50	-3.43	-3.35

Table S10. Continued: Gibbs free formation energies (ΔG, kcal/mol) of clusters at different temperatures in different months of the studied cities shown in Table S9.

Clusters	ΔG (kcal/mol)				
	287 K	288 K	289 K	290 K	291 K
$(SA)_1 (DMA)_1$	-15.04	-15.01	-14.97	-14.94	-14.90
$(SA)_1 (DMA)_1 (TFA)_1$	-26.70	-26.63	-26.56	-26.48	-26.41
$(SA)_1 (DMA)_2 (TFA)_1$	-44.10	-43.98	-43.86	-43.75	-43.63
$(SA)_1 (TFA)_1$	-7.58	-7.55	-7.51	-7.48	-7.44
$(SA)_1 (DMA)_1 (TFA)_2$	-35.53	-35.42	-35.31	-35.19	-35.08
$(SA)_1 (DMA)_2 (TFA)_2$	-51.24	-51.07	-50.91	-50.74	-50.57
$(SA)_1 (DMA)_3 (TFA)_2$	-66.95	-66.74	-66.54	-66.33	-66.13
$(SA)_1 (TFA)_2$	-12.95	-12.88	-12.80	-12.72	-12.64
$(DMA)_1 (TFA)_1$	-8.42	-8.39	-8.36	-8.33	-8.29
$(SA)_2 (DMA)_1$	-35.76	-35.68	-35.60	-35.53	-35.45
$(SA)_2 (DMA)_1 (TFA)_1$	-41.31	-41.20	-41.08	-40.96	-40.84
$(SA)_2 (DMA)_2 (TFA)_1$	-60.74	-60.57	-60.41	-60.24	-60.07
$(SA)_2 (DMA)_3 (TFA)_1$	-78.07	-77.87	-77.66	-77.46	-77.25
$(SA)_2 (TFA)_1$	-14.13	-14.05	-13.97	-13.89	-13.81
$(SA)_2 (DMA)_2$	-56.14	-56.03	-55.91	-55.80	-55.68
$(SA)_2$	-8.81	-8.77	-8.74	-8.71	-8.67
$(DMA)_1 (TFA)_2$	-18.54	-18.46	-18.39	-18.31	-18.24
$(DMA)_2 (TFA)_2$	-33.74	-33.63	-33.51	-33.40	-33.28
$(TFA)_2$	-5.50	-5.46	-5.42	-5.39	-5.35
$(SA)_3 (DMA)_1$	-49.22	-49.10	-48.99	-48.87	-48.75
$(SA)_3 (DMA)_2$	-72.69	-72.53	-72.37	-72.22	-72.06
$(SA)_3 (DMA)_3$	-92.86	-92.66	-92.47	-92.27	-92.07
$(SA)_3$	-14.79	-14.71	-14.63	-14.56	-14.48
$(DMA)_1 (TFA)_3$	-21.46	-21.32	-21.19	-21.06	-20.93
$(DMA)_2 (TFA)_3$	-41.98	-41.81	-41.65	-41.49	-41.32
$(DMA)_3 (TFA)_3$	-52.73	-52.53	-52.32	-52.12	-51.91
$(TFA)_3$	-3.28	-3.21	-3.14	-3.06	-2.99

Table S10. Continued: Gibbs free formation energies (ΔG, kcal/mol) of clusters at different temperatures in different months of the studied cities shown in Table S9.

Clusters	ΔG (kcal/mol)				
	292 K	293 K	294 K	295 K	296 K
$(SA)_1$ $(DMA)_1$	-14.87	-14.84	-14.80	-14.77	-14.73
$(SA)_1$ $(DMA)_1$ $(TFA)_1$	-26.34	-26.26	-26.19	-26.12	-26.05
$(SA)_1$ $(DMA)_2$ $(TFA)_1$	-43.51	-43.40	-43.28	-43.16	-43.05
$(SA)_1$ $(TFA)_1$	-7.41	-7.37	-7.34	-7.30	-7.27
$(SA)_1$ $(DMA)_1$ $(TFA)_2$	-34.97	-34.86	-34.74	-34.63	-34.52
$(SA)_1$ $(DMA)_2$ $(TFA)_2$	-50.41	-50.24	-50.08	-49.91	-49.74
$(SA)_1$ $(DMA)_3$ $(TFA)_2$	-65.92	-65.72	-65.51	-65.31	-65.10
$(SA)_1$ $(TFA)_2$	-12.57	-12.49	-12.41	-12.34	-12.26
$(DMA)_1$ $(TFA)_1$	-8.26	-8.23	-8.19	-8.16	-8.13
$(SA)_2$ $(DMA)_1$	-35.37	-35.29	-35.22	-35.14	-35.06
$(SA)_2$ $(DMA)_1$ $(TFA)_1$	-40.73	-40.61	-40.49	-40.38	-40.26
$(SA)_2$ $(DMA)_2$ $(TFA)_1$	-59.91	-59.74	-59.57	-59.41	-59.24
$(SA)_2$ $(DMA)_3$ $(TFA)_1$	-77.05	-76.84	-76.64	-76.43	-76.23
$(SA)_2$ $(TFA)_1$	-13.73	-13.65	-13.57	-13.49	-13.41
$(SA)_2$ $(DMA)_2$	-55.57	-55.45	-55.34	-55.22	-55.11
$(SA)_2$	-8.64	-8.60	-8.57	-8.54	-8.50
$(DMA)_1$ $(TFA)_2$	-18.16	-18.09	-18.01	-17.94	-17.86
$(DMA)_2$ $(TFA)_2$	-33.16	-33.05	-32.93	-32.82	-32.70
$(TFA)_2$	-5.31	-5.27	-5.24	-5.20	-5.16
$(SA)_3$ $(DMA)_1$	-48.63	-48.51	-48.39	-48.27	-48.16
$(SA)_3$ $(DMA)_2$	-71.91	-71.75	-71.60	-71.44	-71.29
$(SA)_3$ $(DMA)_3$	-91.88	-91.68	-91.49	-91.29	-91.09
$(SA)_3$	-14.40	-14.32	-14.25	-14.17	-14.10
$(DMA)_1$ $(TFA)_3$	-20.80	-20.66	-20.53	-20.40	-20.26
$(DMA)_2$ $(TFA)_3$	-41.16	-41.00	-40.83	-40.67	-40.51
$(DMA)_3$ $(TFA)_3$	-51.71	-51.50	-51.29	-51.09	-50.88
$(TFA)_3$	-2.92	-2.85	-2.78	-2.70	-2.63

Table S10. Continued: Gibbs free formation energies (ΔG, kcal/mol) of clusters at different temperatures in different months of the studied cities shown in Table S9.

Clusters	ΔG (kcal/mol)				
	297 K	298 K	299 K	301 K	303 K
$(SA)_1$ $(DMA)_1$	-14.70	-14.66	-14.63	-14.56	-14.49
$(SA)_1$ $(DMA)_1$ $(TFA)_1$	-25.97	-25.90	-25.83	-25.68	-25.54
$(SA)_1$ $(DMA)_2$ $(TFA)_1$	-42.93	-42.81	-42.69	-42.46	-42.23
$(SA)_1$ $(TFA)_1$	-7.23	-7.20	-7.16	-7.09	-7.02
$(SA)_1$ $(DMA)_1$ $(TFA)_2$	-34.41	-34.29	-34.18	-33.96	-33.73
$(SA)_1$ $(DMA)_2$ $(TFA)_2$	-49.58	-49.41	-49.25	-48.91	-48.58
$(SA)_1$ $(DMA)_3$ $(TFA)_2$	-64.90	-64.69	-64.49	-64.08	-63.67
$(SA)_1$ $(TFA)_2$	-12.18	-12.11	-12.03	-11.88	-11.72
$(DMA)_1$ $(TFA)_1$	-8.10	-8.06	-8.03	-7.96	-7.90
$(SA)_2$ $(DMA)_1$	-34.98	-34.91	-34.83	-34.68	-34.52
$(SA)_2$ $(DMA)_1$ $(TFA)_1$	-40.14	-40.03	-39.91	-39.68	-39.44
$(SA)_2$ $(DMA)_2$ $(TFA)_1$	-59.07	-58.91	-58.74	-58.41	-58.07
$(SA)_2$ $(DMA)_3$ $(TFA)_1$	-76.02	-75.81	-75.61	-75.20	-74.79
$(SA)_2$ $(TFA)_1$	-13.33	-13.25	-13.18	-13.02	-12.86
$(SA)_2$ $(DMA)_2$	-54.99	-54.88	-54.76	-54.53	-54.30
$(SA)_2$	-8.47	-8.44	-8.40	-8.34	-8.27
$(DMA)_1$ $(TFA)_2$	-17.79	-17.71	-17.64	-17.49	-17.34
$(DMA)_2$ $(TFA)_2$	-32.59	-32.47	-32.36	-32.13	-31.89
$(TFA)_2$	-5.13	-5.09	-5.05	-4.97	-4.90
$(SA)_3$ $(DMA)_1$	-48.04	-47.92	-47.80	-47.56	-47.32
$(SA)_3$ $(DMA)_2$	-71.13	-70.97	-70.82	-70.51	-70.19
$(SA)_3$ $(DMA)_3$	-90.90	-90.70	-90.50	-90.11	-89.72
$(SA)_3$	-14.02	-13.94	-13.86	-13.71	-13.56
$(DMA)_1$ $(TFA)_3$	-20.13	-20.00	-19.87	-19.60	-19.34
$(DMA)_2$ $(TFA)_3$	-40.34	-40.18	-40.01	-39.69	-39.36
$(DMA)_3$ $(TFA)_3$	-50.68	-50.47	-50.27	-49.86	-49.44
$(TFA)_3$	-2.56	-2.49	-2.41	-2.27	-2.13

Table S10. Continued: Gibbs free formation energies (Δ*G*, kcal/mol) of clusters at different temperatures in different months of the studied cities shown in Table S9.

Clusters	Δ*G* (kcal/mol)				
	304 K	306 K	307 K	308 K	309 K
$(SA)_1$ $(DMA)_1$	-14.46	-14.39	-14.35	-14.32	-14.29
$(SA)_1$ $(DMA)_1$ $(TFA)_1$	-25.47	-25.32	-25.25	-25.18	-25.10
$(SA)_1$ $(DMA)_2$ $(TFA)_1$	-42.11	-41.88	-41.76	-41.64	-41.53
$(SA)_1$ $(TFA)_1$	-6.99	-6.92	-6.88	-6.85	-6.81
$(SA)_1$ $(DMA)_1$ $(TFA)_2$	-33.62	-33.40	-33.28	-33.17	-33.06
$(SA)_1$ $(DMA)_2$ $(TFA)_2$	-48.42	-48.08	-47.92	-47.75	-47.59
$(SA)_1$ $(DMA)_3$ $(TFA)_2$	-63.46	-63.05	-62.85	-62.64	-62.44
$(SA)_1$ $(TFA)_2$	-11.65	-11.49	-11.42	-11.34	-11.26
$(DMA)_1$ $(TFA)_1$	-7.87	-7.80	-7.77	-7.74	-7.70
$(SA)_2$ $(DMA)_1$	-34.44	-34.29	-34.21	-34.13	-34.06
$(SA)_2$ $(DMA)_1$ $(TFA)_1$	-39.33	-39.09	-38.98	-38.86	-38.74
$(SA)_2$ $(DMA)_2$ $(TFA)_1$	-57.91	-57.58	-57.41	-57.24	-57.08
$(SA)_2$ $(DMA)_3$ $(TFA)_1$	-74.58	-74.17	-73.97	-73.76	-73.56
$(SA)_2$ $(TFA)_1$	-12.78	-12.62	-12.54	-12.46	-12.39
$(SA)_2$ $(DMA)_2$	-54.19	-53.96	-53.84	-53.73	-53.61
$(SA)_2$	-8.24	-8.17	-8.13	-8.10	-8.07
$(DMA)_1$ $(TFA)_2$	-17.27	-17.12	-17.04	-16.97	-16.89
$(DMA)_2$ $(TFA)_2$	-31.78	-31.55	-31.43	-31.32	-31.20
$(TFA)_2$	-4.86	-4.79	-4.75	-4.71	-4.68
$(SA)_3$ $(DMA)_1$	-47.21	-46.97	-46.85	-46.73	-46.61
$(SA)_3$ $(DMA)_2$	-70.04	-69.73	-69.57	-69.42	-69.26
$(SA)_3$ $(DMA)_3$	-89.52	-89.13	-88.94	-88.74	-88.54
$(SA)_3$	-13.48	-13.33	-13.25	-13.17	-13.10
$(DMA)_1$ $(TFA)_3$	-19.21	-18.94	-18.81	-18.68	-18.55
$(DMA)_2$ $(TFA)_3$	-39.20	-38.87	-38.71	-38.54	-38.38
$(DMA)_3$ $(TFA)_3$	-49.24	-48.83	-48.62	-48.42	-48.21
$(TFA)_3$	-2.05	-1.91	-1.84	-1.76	-1.69

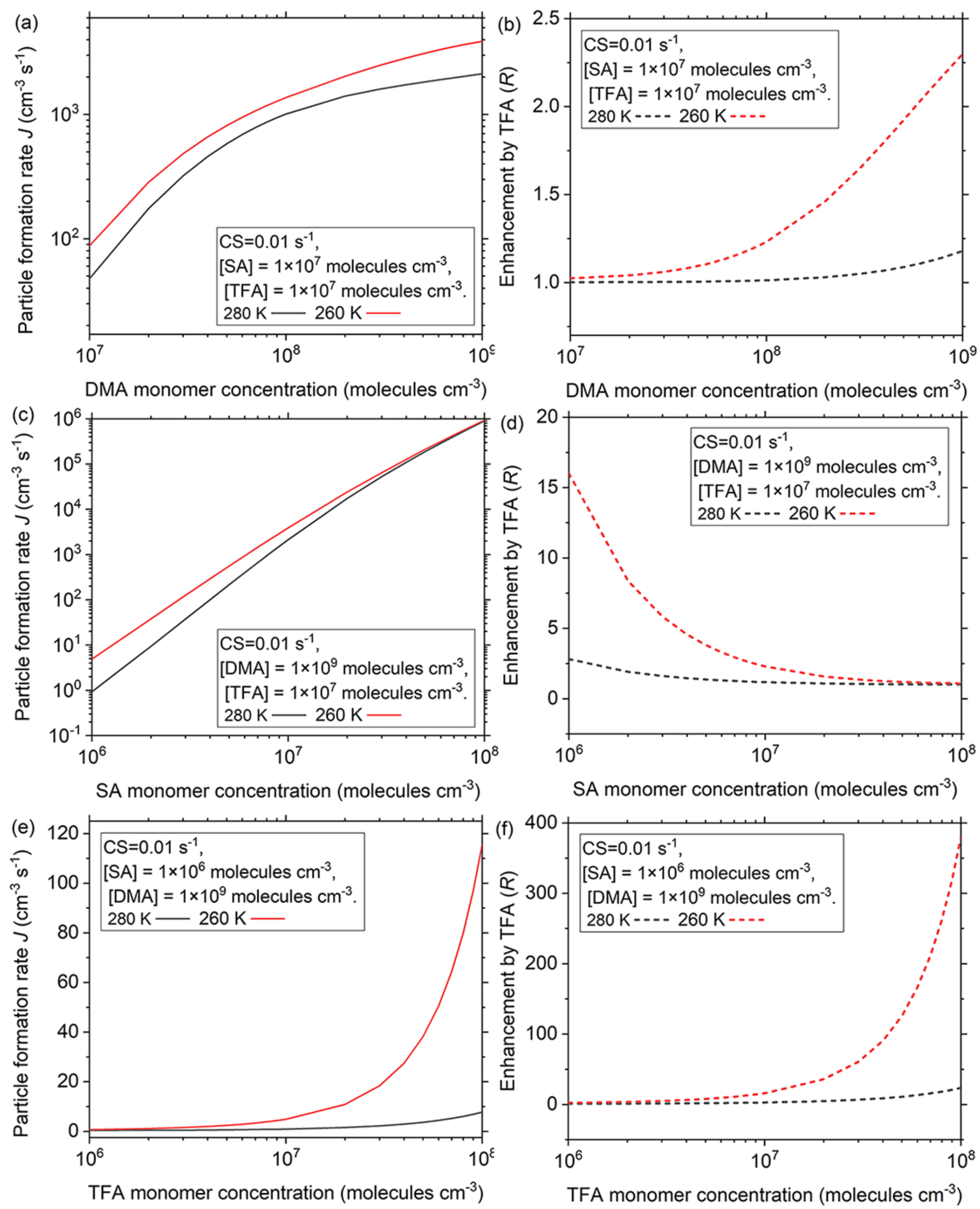

Figure S1. Particle formation rates (J, cm^{-3} s^{-1}) at different temperatures (280 K and 260 K) as a function of (a) DMA monomer concentrations, (c) SA monomer concentrations and (e) TFA monomer concentrations. Enhancement of particle formation rate by TFA (R, $R = J_{\text{SA-DMA-TFA}}/J_{\text{SA-DMA}}$) at different temperatures as a function of (b) DMA monomer concentrations, (d) SA monomer concentrations and (f) TFA monomer concentrations. Black and red lines are corresponding to 280 K and 260 K, respectively. CS = 0.01 s^{-1}.

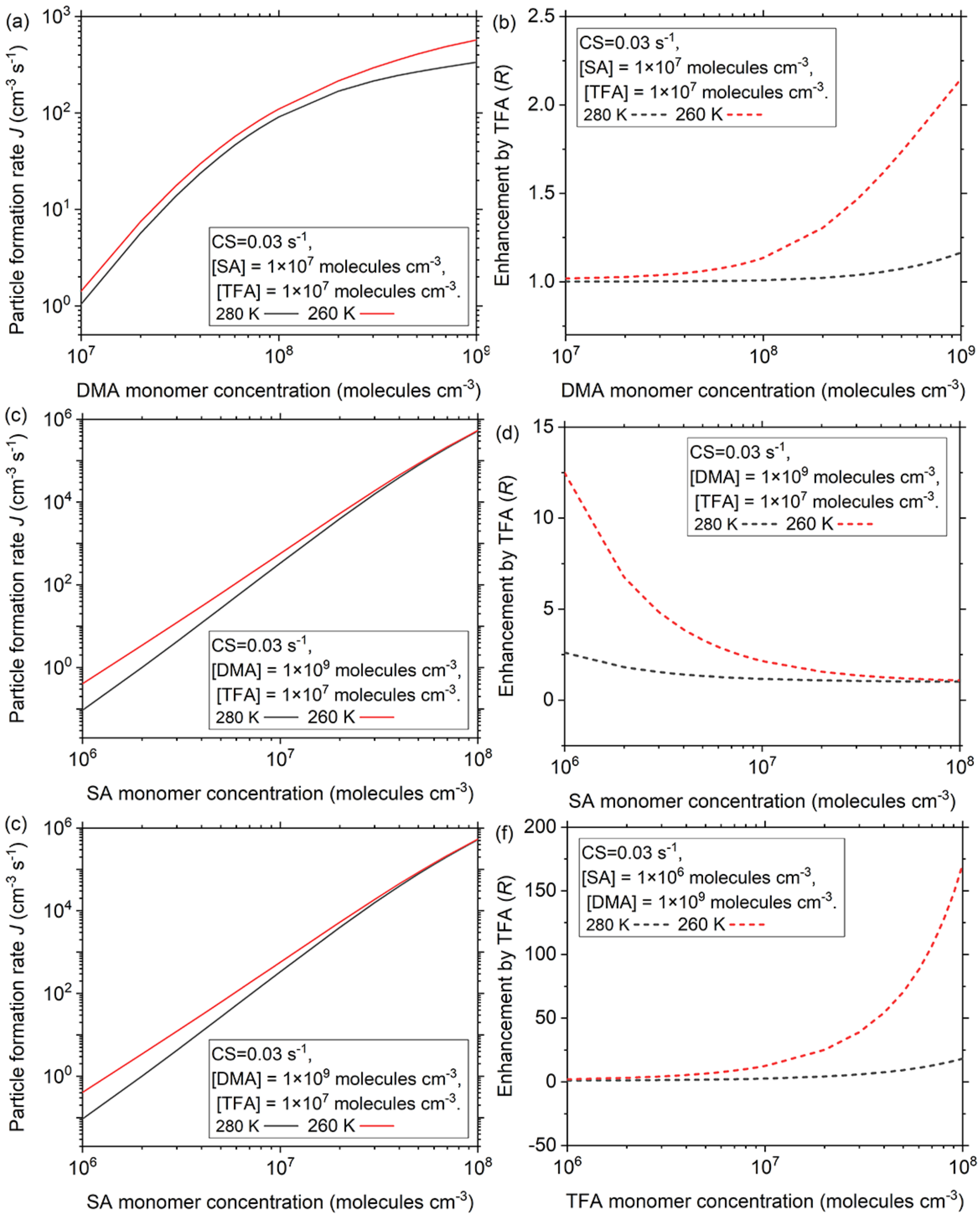

Figure S2. Particle formation rates (J, cm^{-3} s^{-1}) at different temperatures (280 K and 260 K) as a function of (a) DMA monomer concentrations, (c) SA monomer concentrations and (e) TFA monomer concentrations. Enhancement of particle formation rate by TFA (R, $R = J_{SA\text{-}DMA\text{-}TFA}/J_{SA\text{-}DMA}$) at different temperatures as a function of (b) DMA monomer concentrations, (d) SA monomer concentrations and (f) TFA monomer concentrations. Black and red lines are corresponding to 280 K and 260 K, respectively. CS = 0.03 s^{-1}.

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