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Supplemental Information

New Particle Formation and Growth from Methanesulfonic Acid, Trimethylamine and Water

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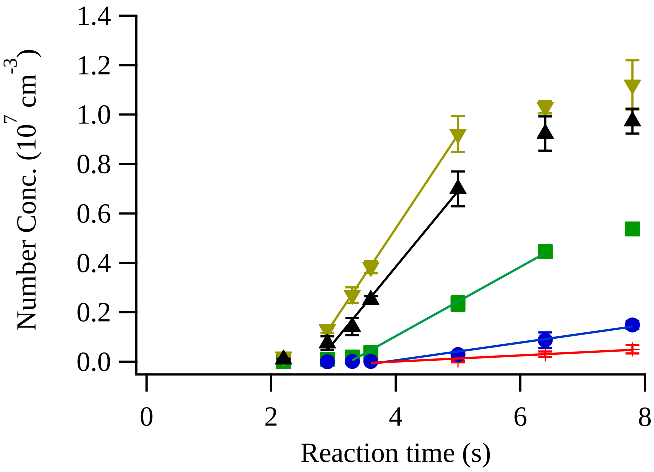

Figure S1. Number concentrations of particles measured by SMPS in Exp. 8 from the reaction of 1.8 ppb MSA with 2.0 ppb TMA as a function of reaction time at RH 7% (red plus), 15% (blue filled circle), 31% (green filled square), 48% (black filled triangle) and 56% (brown filled upside-down triangle). Data points (four for each RH) and the corresponding linear fittings to estimate particle formation rates, $J_{exp, >2.5\ nm}$ are shown.

Model Description

The model was written in C, and has been compiled with a solver, 'gsl_odeiv2_step_msbdf', provided in the GNU Scientific Library for the integration of ordinary differential equations. The model has 94 species and 157 reactions as listed in Table S1. Particle formation was simulated as a series of reactions representing the formation of small clusters from MSA, TMA and H_2O as well as the subsequent growth of the small clusters to detectable particles by addition of MSA, TMA, H_2O and small clusters. Gas precursors and clusters can also be lost due to a combination of uptake on the wall and condensation on particles. As described in the text, clusters can grow from different pathways depending on the RH and the relative initial concentrations of MSA and TMA in the system. Clusters that were treated as particles contain ~20-30 molecules as shown in bold in Table S1.

Inputs to the model include the precursor species (MSA, TMA, H_2O) and their initial concentrations based on the experimental values as described in the text. The initial concentrations of clusters and particles were set to be zero. The individual reactions and rate constants used are listed in Table S1. Reactions S1, S3, S5 and S7 in Table S1 were assumed to be diffusion-controlled, and rate constants were calculated using the theory of hard-sphere collisions with activation energies of zero. Rate constants for their reverse reactions (reactions S2, S4, S6 and S8 in Table S1) were calculated using the rate constants for the forward reactions and ΔG for the reactions that were determined previously using quantum chemical calculations.[1] Rate constants for the subsequent steps were adjusted to fit the experimentally determined time profiles of particle number concentrations. In order to decrease the number of independent parameters in the model, rate constants for the addition of each type of molecule/cluster to initial clusters in each pathway were set to be the same. The rate constants for some reactions are much smaller than those assumed for other steps. The reason is that these represent the overall reaction in the forward direction and do not take into account fast back-reactions. For example, the free energies for reactions S12/(5), S9/(7), S11/(9), S14/(10) and S17 are very small (0.5-1 kcal mol^{-1}), so that if the forward reactions are diffusion-controlled, the back reactions have rate constants of the order of 10^8 s^{-1}, and the net reactions in the forward direction are slow. The losses of gases and clusters, except MSA, TMA and MSA•H_2O, were assumed be first order with rate constants of 0.05 s^{-1}. The first order rate constants for the losses of MSA, TMA and MSA•H_2O were assumed to be larger, given their high diffusion rates. The outputs of the model were concentrations of each species at each time step. The time step used in the model was set to be 0.1 s. Time steps of 0.02 and 0.01 s were also tested for some experiments and shown not to affect the predicted concentrations.

Table S1. Reactions and corresponding rate constants used in the box model for the MSA-TMA-H_2O system

No.[a]	Reaction	Rate Constant[b]
S1 (2)	$MSA + H_2O \rightarrow MSA \bullet H_2O$	6.58E-10[c]
S2 (-2)	$MSA \bullet H_2O \rightarrow MSA + H_2O$	5.53E+08[d]
S3 (3)	$MSA \bullet H_2O + TMA \rightarrow MSA \bullet TMA \bullet H_2O$	1.32E-09[c]
S4 (-3)	$MSA \bullet TMA \bullet H_2O \rightarrow MSA \bullet H_2O + TMA$	3.09E+01[d]
S5 (1)	$MSA + TMA \rightarrow MSA \bullet TMA$	4.25E-10[c]
S6 (-1)	$MSA \bullet TMA \rightarrow MSA + TMA$	1.54E+01[d]
S7 (4)	$MSA \bullet TMA + H_2O \rightarrow MSA \bullet TMA \bullet H_2O$	1.66E-09[c]
S8 (-4)	$MSA \bullet TMA \bullet H_2O \rightarrow MSA \bullet TMA + H_2O$	9.96E+08[d]
S9 (7)	$MSA \bullet TMA \bullet H_2O + H_2O \rightarrow MSA \bullet TMA \bullet (H_2O)_2$	1.00E-19
S10 (8)	$MSA \bullet TMA \bullet (H_2O)_2 + MSA \rightarrow (MSA)_2 \bullet TMA \bullet (H_2O)_2$	5.00E-10
S11 (9)	$(MSA)_2 \bullet TMA \bullet (H_2O)_2 + H_2O \rightarrow (MSA)_2 \bullet TMA \bullet (H_2O)_3$	4.00E-18
S12 (5)	$MSA \bullet H_2O + H_2O \rightarrow MSA \bullet (H_2O)_2$	5.00E-20
S13 (6)	$MSA \bullet (H_2O)_2 + TMA \rightarrow MSA \bullet TMA \bullet (H_2O)_2$	1.00E-09
S14 (10)	$MSA \bullet TMA \bullet (H_2O)_2 + H_2O \rightarrow MSA \bullet TMA \bullet (H_2O)_3$	4.00E-18
S15	$MSA \bullet TMA \bullet (H_2O)_3 + MSA \rightarrow (MSA)_2 \bullet TMA \bullet (H_2O)_3$	1.00E-09
S16	$MSA \bullet TMA \bullet H_2O + MSA \rightarrow (MSA)_2 \bullet TMA \bullet H_2O$	3.00E-11
S17	$(MSA)_2 \bullet TMA \bullet H_2O + H_2O \rightarrow (MSA)_2 \bullet TMA \bullet (H_2O)_2$	1.00E-19
S18	$MSA \bullet TMA \bullet H_2O + MSA \bullet H_2O \rightarrow (MSA)_2 \bullet TMA \bullet (H_2O)_2$	4.00E-19
S19	$(MSA)_2 \bullet TMA \bullet (H_2O)_3 + TMA \rightarrow (MSA)_2 \bullet (TMA)_2 \bullet (H_2O)_3$	5.00E-10
S20	$(MSA)_2 \bullet (TMA)_2 \bullet (H_2O)_3 + MSA \rightarrow (MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_3$	4.00E-10
S21	$(MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_3 + H_2O \rightarrow (MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_4$	5.00E-10
S22	$(MSA)_2 \bullet (TMA)_2 \bullet (H_2O)_3 + MSA \bullet H_2O \rightarrow (MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_4$	2.00E-10
S23	$(MSA)_2 \bullet TMA \bullet (H_2O)_3 + MSA \bullet TMA \rightarrow (MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_3$	1.00E-11
S24	$(MSA)_2 \bullet TMA \bullet (H_2O)_3 + MSA \bullet TMA \bullet H_2O \rightarrow (MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_4$	8.00E-12
S25	$(MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_4 + TMA \rightarrow (MSA)_3 \bullet (TMA)_3 \bullet (H_2O)_4$	1.40E-10
S26	$(MSA)_3 \bullet (TMA)_3 \bullet (H_2O)_4 + MSA \rightarrow (MSA)_4 \bullet (TMA)_3 \bullet (H_2O)_4$	4.00E-10
S27	$(MSA)_4 \bullet (TMA)_3 \bullet (H_2O)_4 + H_2O \rightarrow (MSA)_4 \bullet (TMA)_3 \bullet (H_2O)_5$	5.00E-10
S28	$(MSA)_3 \bullet (TMA)_3 \bullet (H_2O)_4 + MSA \bullet H_2O \rightarrow (MSA)_4 \bullet (TMA)_3 \bullet (H_2O)_5$	2.00E-10
S29	$(MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_4 + MSA \bullet TMA \rightarrow (MSA)_4 \bullet (TMA)_3 \bullet (H_2O)_4$	1.00E-11
S30	$(MSA)_3 \bullet (TMA)_2 \bullet (H_2O)_4 + MSA \bullet TMA \bullet H_2O \rightarrow (MSA)_4 \bullet (TMA)_3 \bullet (H_2O)_5$	8.00E-12
S31	$(MSA)_4 \bullet (TMA)_3 \bullet (H_2O)_5 + TMA \rightarrow (MSA)_4 \bullet (TMA)_4 \bullet (H_2O)_5$	1.40E-10
S32	$(MSA)_4 \bullet (TMA)_4 \bullet (H_2O)_5 + MSA \rightarrow (MSA)_5 \bullet (TMA)_4 \bullet (H_2O)_5$	4.00E-10

S33	$(MSA)_5{\bullet}(TMA)_4{\bullet}(H_2O)_5 + H_2O \rightarrow (MSA)_5{\bullet}(TMA)_4{\bullet}(H_2O)_6$	5.00E-10
S34	$(MSA)_4{\bullet}(TMA)_4{\bullet}(H_2O)_5 + MSA{\bullet}H_2O \rightarrow (MSA)_5{\bullet}(TMA)_4{\bullet}(H_2O)_6$	2.00E-10
S35	$(MSA)_4{\bullet}(TMA)_3{\bullet}(H_2O)_5 + MSA{\bullet}TMA \rightarrow (MSA)_5{\bullet}(TMA)_4{\bullet}(H_2O)_5$	1.00E-11
S36	$(MSA)_4{\bullet}(TMA)_3{\bullet}(H_2O)_5 + MSA{\bullet}TMA{\bullet}H_2O \rightarrow (MSA)_5{\bullet}(TMA)_4{\bullet}(H_2O)_6$	8.00E-12
S37	$(MSA)_5{\bullet}(TMA)_4{\bullet}(H_2O)_6 + TMA \rightarrow (MSA)_5{\bullet}(TMA)_5{\bullet}(H_2O)_6$	1.40E-10
S38	$(MSA)_5{\bullet}(TMA)_5{\bullet}(H_2O)_6 + MSA \rightarrow (MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_6$	4.00E-10
S39	$(MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_6 + H_2O \rightarrow (MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_7$	5.00E-10
S40	$(MSA)_5{\bullet}(TMA)_5{\bullet}(H_2O)_6 + MSA{\bullet}H_2O \rightarrow (MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_7$	2.00E-10
S41	$(MSA)_5{\bullet}(TMA)_4{\bullet}(H_2O)_6 + MSA{\bullet}TMA \rightarrow (MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_6$	1.00E-11
S42	$(MSA)_5{\bullet}(TMA)_4{\bullet}(H_2O)_6 + MSA{\bullet}TMA{\bullet}H_2O \rightarrow (MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_7$	8.00E-12
S43	$(MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_7 + TMA \rightarrow (MSA)_6{\bullet}(TMA)_6{\bullet}(H_2O)_7$	1.40E-10
S44	$(MSA)_6{\bullet}(TMA)_6{\bullet}(H_2O)_7 + MSA \rightarrow (MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_7$	4.00E-10
S45	$(MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_7 + H_2O \rightarrow (MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_8$	5.00E-10
S46	$(MSA)_6{\bullet}(TMA)_6{\bullet}(H_2O)_7 + MSA{\bullet}H_2O \rightarrow (MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_8$	2.00E-10
S47	$(MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_7 + MSA{\bullet}TMA \rightarrow (MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_7$	1.00E-11
S48	$(MSA)_6{\bullet}(TMA)_5{\bullet}(H_2O)_7 + MSA{\bullet}TMA{\bullet}H_2O \rightarrow (MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_8$	8.00E-12
S49	$(MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_8 + TMA \rightarrow (MSA)_7{\bullet}(TMA)_7{\bullet}(H_2O)_8$	1.40E-10
S50	$(MSA)_7{\bullet}(TMA)_7{\bullet}(H_2O)_8 + MSA \rightarrow (MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_8$	4.00E-10
S51	$(MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_8 + H_2O \rightarrow (MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_9$	5.00E-10
S52	$(MSA)_7{\bullet}(TMA)_7{\bullet}(H_2O)_8 + MSA{\bullet}H_2O \rightarrow (MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_9$	2.00E-10
S53	$(MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_8 + MSA{\bullet}TMA \rightarrow (MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_8$	1.00E-11
S54	$(MSA)_7{\bullet}(TMA)_6{\bullet}(H_2O)_8 + MSA{\bullet}TMA{\bullet}H_2O \rightarrow (MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_9$	8.00E-12
S55	$(MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_9 + TMA \rightarrow (MSA)_8{\bullet}(TMA)_8{\bullet}(H_2O)_9$	1.40E-10
S56	$(MSA)_8{\bullet}(TMA)_8{\bullet}(H_2O)_9 + MSA \rightarrow (MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_9$	4.00E-10
S57	$(MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_9 + H_2O \rightarrow (MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_{10}$	5.00E-10
S58	$(MSA)_8{\bullet}(TMA)_8{\bullet}(H_2O)_9 + MSA{\bullet}H_2O \rightarrow (MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_{10}$	2.00E-10
S59	$(MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_9 + MSA{\bullet}TMA \rightarrow (MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_9$	1.00E-11
S60	$(MSA)_8{\bullet}(TMA)_7{\bullet}(H_2O)_9 + MSA{\bullet}TMA{\bullet}H_2O \rightarrow (MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_{10}$	8.00E-12
S61	$(MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_{10} + TMA \rightarrow (MSA)_9{\bullet}(TMA)_9{\bullet}(H_2O)_{10}$	1.40E-10
S62	$(MSA)_9{\bullet}(TMA)_9{\bullet}(H_2O)_{10} + MSA \rightarrow (MSA)_{10}{\bullet}(TMA)_9{\bullet}(H_2O)_{10}$	4.00E-10
S63	$(MSA)_{10}{\bullet}(TMA)_9{\bullet}(H_2O)_{10} + H_2O \rightarrow (MSA)_{10}{\bullet}(TMA)_9{\bullet}(H_2O)_{11}$	5.00E-10
S64	$(MSA)_9{\bullet}(TMA)_9{\bullet}(H_2O)_{10} + MSA{\bullet}H_2O \rightarrow (MSA)_{10}{\bullet}(TMA)_9(H_2O)_{11}$	2.00E-10
S65	$(MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_{10} + MSA{\bullet}TMA \rightarrow (MSA)_{10}{\bullet}(TMA)_9{\bullet}(H_2O)_{10}$	1.00E-11
S66	$(MSA)_9{\bullet}(TMA)_8{\bullet}(H_2O)_{10} + MSA{\bullet}TMA{\bullet}H_2O \rightarrow (MSA)_{10}{\bullet}(TMA)_9{\bullet}(H_2O)_{11}$	8.00E-12

S67	$(MSA)_{10}\bullet(TMA)_9\bullet(H_2O)_{11}$ + TMA → $\mathbf{(MSA)_{10}\bullet(TMA)_{10}\bullet(H_2O)_{11}}$ **(Particle)**[e]	1.40E-10
S68	$(MSA)_2\bullet TMA\bullet(H_2O)_3$ + MSA → $(MSA)_3\bullet TMA\bullet(H_2O)_3$	4.00E-12
S69	$(MSA)_3\bullet TMA\bullet(H_2O)_3$ + H_2O → $(MSA)_3\bullet TMA\bullet(H_2O)_4$	2.00E-12
S70	$(MSA)_2\bullet TMA\bullet(H_2O)_3$ + $MSA\bullet H_2O$ → $(MSA)_3\bullet TMA\bullet(H_2O)_4$	1.00E-12
S71	$(MSA)_3\bullet TMA\bullet(H_2O)_4$ + MSA → $(MSA)_4\bullet TMA\bullet(H_2O)_4$	5.00E-11
S72	$(MSA)_4\bullet TMA\bullet(H_2O)_4$ + TMA → $(MSA)_4\bullet(TMA)_2\bullet(H_2O)_4$	1.00E-09
S73	$(MSA)_4\bullet(TMA)_2\bullet(H_2O)_4$ + H_2O → $(MSA)_4\bullet(TMA)_2\bullet(H_2O)_5$	5.00E-10
S74	$(MSA)_3\bullet TMA\bullet(H_2O)_4$ + MSA•TMA → $(MSA)_4\bullet(TMA)_2\bullet(H_2O)_4$	5.00E-11
S75	$(MSA)_3\bullet TMA\bullet(H_2O)_4$ + $MSA\bullet H_2O$ → $(MSA)_4\bullet TMA\bullet(H_2O)_5$	4.00E-11
S76	$(MSA)_4\bullet TMA\bullet(H_2O)_5$ + TMA → $(MSA)_4\bullet(TMA)_2\bullet(H_2O)_5$	1.00E-09
S77	$(MSA)_3\bullet TMA\bullet(H_2O)_4$ + $MSA\bullet TMA\bullet H_2O$ → $(MSA)_4\bullet(TMA)_2\bullet(H_2O)_5$	3.00E-11
S78	$(MSA)_4\bullet(TMA)_2\bullet(H_2O)_5$ + MSA → $(MSA)_5\bullet(TMA)_2\bullet(H_2O)_5$	5.00E-11
S79	$(MSA)_5\bullet(TMA)_2\bullet(H_2O)_5$ +TMA → $(MSA)_5\bullet(TMA)_3\bullet(H_2O)_5$	1.00E-09
S80	$(MSA)_5\bullet(TMA)_3\bullet(H_2O)_5$ + H_2O → $(MSA)_5\bullet(TMA)_3\bullet(H_2O)_6$	5.00E-10
S81	$(MSA)_4\bullet(TMA)_2\bullet(H_2O)_5$ + MSA•TMA → $(MSA)_5\bullet(TMA)_3\bullet(H_2O)_5$	5.00E-11
S82	$(MSA)_4\bullet(TMA)_2\bullet(H_2O)_5$ + $MSA\bullet H_2O$ → $(MSA)_5\bullet(TMA)_2\bullet(H_2O)_6$	4.00E-11
S83	$(MSA)_5\bullet(TMA)_2\bullet(H_2O)_6$ + TMA → $(MSA)_5\bullet(TMA)_3\bullet(H_2O)_6$	1.00E-09
S84	$(MSA)_4\bullet(TMA)_2\bullet(H_2O)_5$ + $MSA\bullet TMA\bullet H_2O$ → $(MSA)_5\bullet(TMA)_3\bullet(H_2O)_6$	3.00E-11
S85	$(MSA)_5\bullet(TMA)_3\bullet(H_2O)_6$ + H_2O → $(MSA)_5\bullet(TMA)_3\bullet(H_2O)_7$	5.00E-10
S86	$(MSA)_5\bullet(TMA)_3\bullet(H_2O)_7$ + MSA → $(MSA)_6\bullet(TMA)_3\bullet(H_2O)_7$	5.00E-10
S87	$(MSA)_5\bullet(TMA)_3\bullet(H_2O)_6$ + $MSA\bullet H_2O$ → $(MSA)_6\bullet(TMA)_3\bullet(H_2O)_7$	5.00E-10
S88	$(MSA)_6\bullet(TMA)_3\bullet(H_2O)_7$ + H_2O → $(MSA)_6\bullet(TMA)_3\bullet(H_2O)_8$	5.00E-10
S89	$(MSA)_6\bullet(TMA)_3\bullet(H_2O)_8$ + MSA → $(MSA)_7\bullet(TMA)_3\bullet(H_2O)_8$	5.00E-10
S90	$(MSA)_6\bullet(TMA)_3\bullet(H_2O)_7$ + $MSA\bullet H_2O$ → $(MSA)_7\bullet(TMA)_3\bullet(H_2O)_8$	5.00E-10
S91	$(MSA)_7\bullet(TMA)_3\bullet(H_2O)_8$ + H_2O → $(MSA)_7\bullet(TMA)_3\bullet(H_2O)_9$	5.00E-10
S92	$(MSA)_7\bullet(TMA)_3\bullet(H_2O)_9$ + MSA → $(MSA)_8\bullet(TMA)_3\bullet(H_2O)_9$	5.00E-10
S93	$(MSA)_7\bullet(TMA)_3\bullet(H_2O)_8$ + $MSA\bullet H_2O$ → $(MSA)_8\bullet(TMA)_3\bullet(H_2O)_9$	5.00E-10
S94	$(MSA)_8\bullet(TMA)_3\bullet(H_2O)_9$ + H_2O → $(MSA)_8\bullet(TMA)_3\bullet(H_2O)_{10}$	5.00E-10
S95	$(MSA)_8\bullet(TMA)_3\bullet(H_2O)_{10}$ + MSA → $(MSA)_9\bullet(TMA)_3\bullet(H_2O)_{10}$	5.00E-10
S96	$(MSA)_8\bullet(TMA)_3\bullet(H_2O)_9$ + $MSA\bullet H_2O$ → $(MSA)_9\bullet(TMA)_3\bullet(H_2O)_{10}$	5.00E-10
S97	$(MSA)_9\bullet(TMA)_3\bullet(H_2O)_{10}$ + H_2O → $(MSA)_9\bullet(TMA)_3\bullet(H_2O)_{11}$	5.00E-10
S98	$(MSA)_9\bullet(TMA)_3\bullet(H_2O)_{11}$ + MSA → $(MSA)_{10}\bullet(TMA)_3\bullet(H_2O)_{11}$	5.00E-10
S99	$(MSA)_9\bullet(TMA)_3\bullet(H_2O)_{10}$ + $MSA\bullet H_2O$ → $(MSA)_{10}\bullet(TMA)_3\bullet(H_2O)_{11}$	5.00E-10
S100	$(MSA)_{10}\bullet(TMA)_3\bullet(H_2O)_{11}$ + H_2O → $(MSA)_{10}\bullet(TMA)_3\bullet(H_2O)_{12}$	5.00E-10
S101	$(MSA)_{10}\bullet(TMA)_3\bullet(H_2O)_{12}$ + MSA → $\mathbf{(MSA)_{11}\bullet(TMA)_3\bullet(H_2O)_{12}}$ **(Particle)**[e]	5.00E-10

S102	$(MSA)_{10}$•$(TMA)_3$•$(H_2O)_{11}$ + MSA•H_2O → **$(MSA)_{11}$•$(TMA)_3$•$(H_2O)_{12}$ (Particle)**[e]	5.00E-10
S103	MSA•TMA•H_2O + TMA → MSA•$(TMA)_2$•H_2O	1.40E-12
S104	MSA•$(TMA)_2$•H_2O + H_2O → MSA•$(TMA)_2$•$(H_2O)_2$	1.40E-10
S105	MSA•$(TMA)_2$•$(H_2O)_2$ + TMA → MSA•$(TMA)_3$•$(H_2O)_2$	7.50E-13
S106	MSA•$(TMA)_3$•$(H_2O)_2$ + MSA → $(MSA)_2$•$(TMA)_3$•$(H_2O)_2$	6.00E-11
S107	$(MSA)_2$•$(TMA)_3$•$(H_2O)_2$ + H_2O → $(MSA)_2$•$(TMA)_3$•$(H_2O)_3$	7.50E-11
S108	MSA•$(TMA)_2$•$(H_2O)_2$ + MSA•TMA → $(MSA)_2$•$(TMA)_3$•$(H_2O)_2$	8.17E-14
S109	MSA•$(TMA)_3$•$(H_2O)_2$ + MSA•H_2O → $(MSA)_2$•$(TMA)_3$•$(H_2O)_3$	3.17E-11
S110	MSA•$(TMA)_2$•$(H_2O)_2$ + MSA•TMA•H_2O → $(MSA)_2$•$(TMA)_3$•$(H_2O)_3$	3.17E-11
S111	$(MSA)_2$•$(TMA)_3$•$(H_2O)_3$ + TMA → $(MSA)_2$•$(TMA)_4$•$(H_2O)_3$	7.50E-13
S112	$(MSA)_2$•$(TMA)_4$•$(H_2O)_3$ + MSA → $(MSA)_3$•$(TMA)_4$•$(H_2O)_3$	6.00E-11
S113	$(MSA)_3$•$(TMA)_4$•$(H_2O)_3$ + H_2O → $(MSA)_3$•$(TMA)_4$•$(H_2O)_4$	7.50E-11
S114	$(MSA)_2$•$(TMA)_3$•$(H_2O)_3$ + MSA•TMA → $(MSA)_3$•$(TMA)_4$•$(H_2O)_3$	8.17E-14
S115	$(MSA)_2$•$(TMA)_4$•$(H_2O)_3$ + MSA•H_2O → $(MSA)_3$•$(TMA)_4$•$(H_2O)_4$	3.17E-11
S116	$(MSA)_2$•$(TMA)_3$•$(H_2O)_3$ + MSA•TMA•H_2O → $(MSA)_3$•$(TMA)_4$•$(H_2O)_4$	3.17E-11
S117	$(MSA)_3$•$(TMA)_4$•$(H_2O)_4$ + TMA → $(MSA)_3$•$(TMA)_5$•$(H_2O)_4$	6.00E-12
S118	$(MSA)_3$•$(TMA)_5$•$(H_2O)_4$ + H_2O → $(MSA)_3$•$(TMA)_5$•$(H_2O)_5$	5.00E-10
S119	$(MSA)_3$•$(TMA)_5$•$(H_2O)_5$ + TMA → $(MSA)_3$•$(TMA)_6$•$(H_2O)_5$	6.00E-12
S120	$(MSA)_3$•$(TMA)_6$•$(H_2O)_5$ + H_2O → $(MSA)_3$•$(TMA)_6$•$(H_2O)_6$	5.00E-10
S121	$(MSA)_3$•$(TMA)_6$•$(H_2O)_6$ + TMA → $(MSA)_3$•$(TMA)_7$•$(H_2O)_6$	6.00E-12
S122	$(MSA)_3$•$(TMA)_7$•$(H_2O)_6$ + H_2O → $(MSA)_3$•$(TMA)_7$•$(H_2O)_7$	5.00E-10
S123	$(MSA)_3$•$(TMA)_7$•$(H_2O)_7$ + TMA → **$(MSA)_3$•$(TMA)_8$•$(H_2O)_7$ (Particle)**[e]	6.00E-12
S124	MSA•TMA + MSA → $(MSA)_2$•TMA	3.00E-12
S125	$(MSA)_2$•TMA + TMA → $(MSA)_2$•$(TMA)_2$	1.00E-13
S126	MSA•TMA + MSA•TMA → $(MSA)_2$•$(TMA)_2$	3.00E-12
S127	$(MSA)_2$•$(TMA)_2$ + MSA → $(MSA)_3$•$(TMA)_2$	2.00E-11
S128	$(MSA)_3$•$(TMA)_2$ + TMA → $(MSA)_3$•$(TMA)_3$	2.00E-12
S129	$(MSA)_2$•$(TMA)_2$ + MSA•TMA → $(MSA)_3$•$(TMA)_3$	2.00E-12
S130	$(MSA)_3$•$(TMA)_3$ + MSA → $(MSA)_4$•$(TMA)_3$	1.00E-09
S131	$(MSA)_4$•$(TMA)_3$ + TMA → $(MSA)_4$•$(TMA)_4$	6.00E-11
S132	$(MSA)_3$•$(TMA)_3$ + MSA•TMA → $(MSA)_4$•$(TMA)_4$	2.00E-10
S133	$(MSA)_4$•$(TMA)_4$ + MSA → $(MSA)_5$•$(TMA)_4$	1.00E-09
S134	$(MSA)_5$•$(TMA)_4$ + TMA → $(MSA)_5$•$(TMA)_5$	6.00E-11
S135	$(MSA)_4$•$(TMA)_4$ + MSA•TMA → $(MSA)_5$•$(TMA)_5$	2.00E-10
S136	$(MSA)_5$•$(TMA)_5$ + MSA → $(MSA)_6$•$(TMA)_5$	1.00E-09

S137	$(MSA)_6$•$(TMA)_5$ + TMA → $(MSA)_6$•$(TMA)_6$	6.00E-11
S138	$(MSA)_5$•$(TMA)_5$ + MSA•TMA → $(MSA)_6$•$(TMA)_6$	2.00E-10
S139	$(MSA)_6$•$(TMA)_6$ + MSA → $(MSA)_7$•$(TMA)_6$	1.00E-09
S140	$(MSA)_7$•$(TMA)_6$ + TMA → $(MSA)_7$•$(TMA)_7$	6.00E-11
S141	$(MSA)_6$•$(TMA)_6$ + MSA•TMA → $(MSA)_7$•$(TMA)_7$	2.00E-10
S142	$(MSA)_7$•$(TMA)_7$ + MSA → $(MSA)_8$•$(TMA)_7$	1.00E-09
S143	$(MSA)_8$•$(TMA)_7$ + TMA → $(MSA)_8$•$(TMA)_8$	6.00E-11
S144	$(MSA)_7$•$(TMA)_7$ + MSA•TMA → $(MSA)_8$•$(TMA)_8$	2.00E-10
S145	$(MSA)_8$•$(TMA)_8$ + MSA → $(MSA)_9$•$(TMA)_8$	1.00E-09
S146	$(MSA)_9$•$(TMA)_8$ + TMA → $(MSA)_9$•$(TMA)_9$	6.00E-11
S147	$(MSA)_8$•$(TMA)_8$ + MSA•TMA → $(MSA)_9$•$(TMA)_9$	2.00E-10
S148	$(MSA)_9$•$(TMA)_9$ + MSA → $(MSA)_{10}$•$(TMA)_9$	1.00E-09
S149	$(MSA)_{10}$•$(TMA)_9$ + TMA → $(MSA)_{10}$•$(TMA)_{10}$	6.00E-11
S150	$(MSA)_9$•$(TMA)_9$ + MSA•TMA → $(MSA)_{10}$•$(TMA)_{10}$	2.00E-10
S151	$(MSA)_{10}$•$(TMA)_{10}$ + MSA → **$(MSA)_{11}$•$(TMA)_{10}$ (Particle)**[e]	2.00E-11
S152	MSA → Loss	7.00E-01
S153	TMA → Loss	2.00E-01
S154	H_2O → Loss	5.00E-02
S155	MSA•H_2O → Loss	5.00E-01
S156	MSA•TMA → Loss	5.00E-02
S157	$(MSA)_x$•$(TMA)_y$•$(H_2O)_z$ → Loss ((x+y+z) > 2)	5.00E-02

[a]Reaction numbers in parentheses correspond to those in the text, and numbers with minus signs represent reverse reactions;
[b]First-order rate constants in units of s^{-1} and second-order rate constants in units of cm^3 $molecules^{-1}$ s^{-1};
[c]Assumed to be diffusion-controlled and calculated using hard-sphere collision theory without activation energies;
[d]Calculated using forward rate constants and ΔG of reactions involving clusters;
[e]Species in bold are treated as particles in the model.

References

1. M. L. Dawson, M. E. Varner, V. Perraud, M. J. Ezell, R. B. Gerber and B. J. Finlayson-Pitts, *Proc. Natl. Acad. Sci. U. S. A.*, 2012, **109**, 18719-18724.