Supplement of

Molecular-level study on the role of methanesulfonic acid in iodine oxoacids nucleation

Jing Li et al.

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Table of Contents

energy density $H(r)$ at the corresponding bond critical points (BCPs) in the studied HIO_3-HIO_2-MSA-based clusters. The orange balls represent BCPs in the AIM theory analysis. HIO_3, HIO_2, and MSA are the shorthand for iodic acid, iodous acid and methanesulfonic acid, respectively. HB (hydrogen bond), XB (halogen bond).

Table S3. The boundary conditions of ACDC simulations at 268, 278, 288, and 298 K, respectively.

Table S4. The ratios of HIO_3 monomer collision frequencies versus total evaporation rate coefficients ($\beta C/\Sigma\gamma$) at T = 268, 278, 288, and 298 K. β is the rate coefficient of cluster collision with HIO_3 monomer, and C is the concentration of HIO_3 monomer (1.0×10^6 molec. cm^{-3}).

Table S5. The evaporation rate coefficients (γ, s^{-1}) of the studied clusters at 278 K.

Table S6. Cartesian coordinates of all clusters in the present study at the ωB97X-D/6-311++G(3df,3pd) + aug-cc-pVTZ-PP with ECP28MDF (for I) level of theory.

Supplementary Methods

Multi-step cluster conformational searching method.

First, the artificial bee colony algorithm combined with Universal force field (UFF) (Rappé et al., 1992) was employed to yield more than 7000000 initial configurations for each cluster by ABCluster software (Zhang and Dolg, 2015). Then up to 1000 relatively stable structures were selected to pre-optimized at the PM7 semiempirical method (Stewart, 2013) by MOPAC 2016 (Stewart, 2016). Next, about 100 structures with the lowest energy were selected to further optimized at the ωB97X-D/6-31+G* (for C, H, O and S atoms) + Lanl2DZ (for I atom) level of theory (Elm and Kristensen, 2017) by Gaussian 09 software (Frisch et al., 2009). Finally, the lower-lying 10 isomers were selected and reoptimized at the ωB97X-D function and 6-311++G (3df, 3pd) (for C, H, O and S atoms) + aug-cc-pVTZ-PP with ECP28MDF (for I atom) level of theory to obtain the global minimum one (Francl et al., 1982; Peterson et al., 2003).

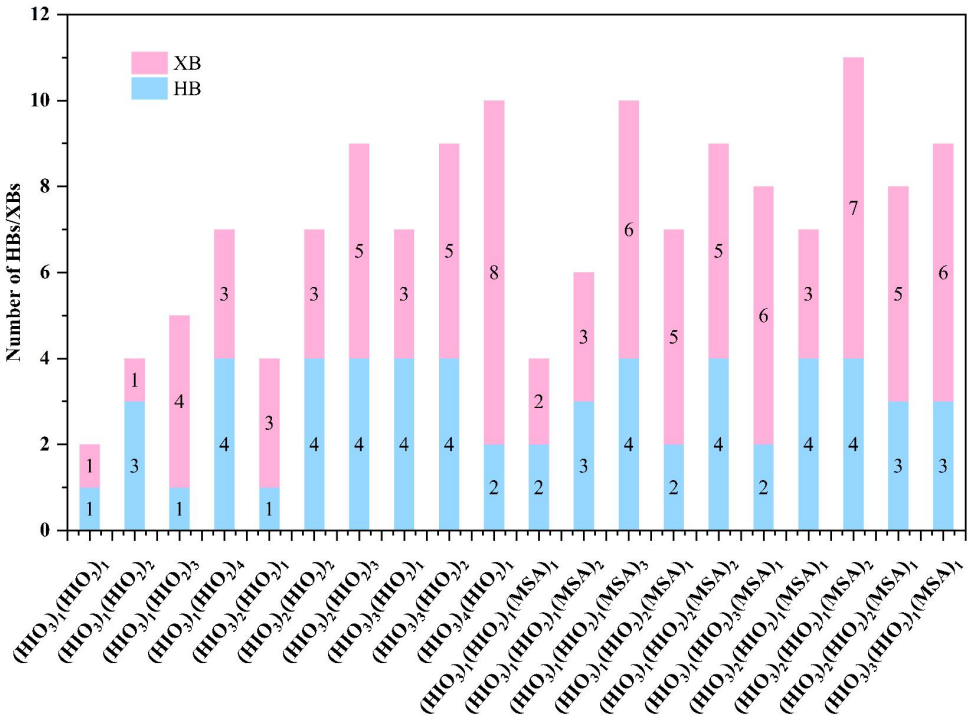


Figure S1. Number of hydrogen bonds (HBs) and halogen bonds (XBs) in the HIO_3-HIO_2 clusters and HIO_3-HIO_2-MSA clusters.

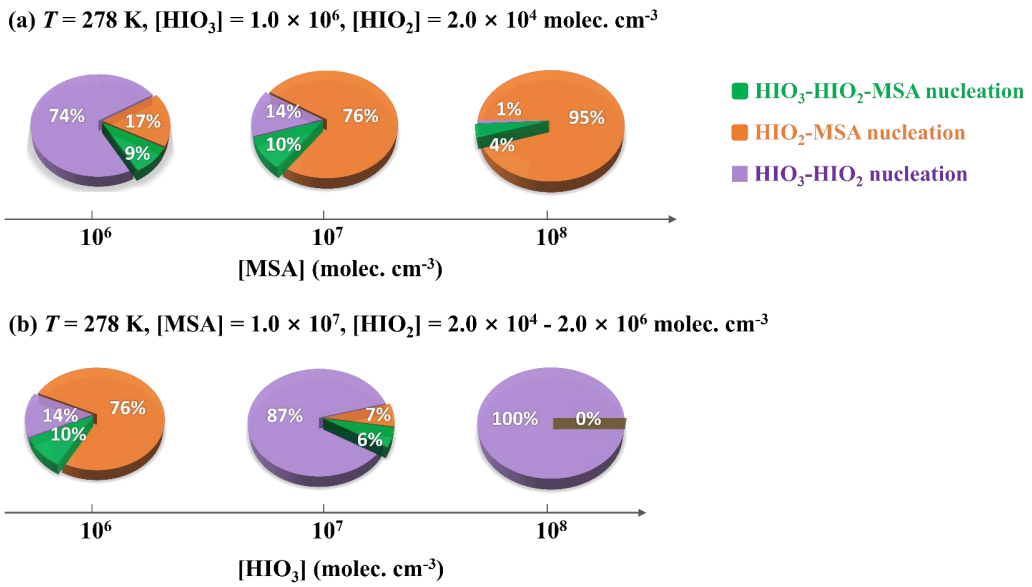


Figure S2. The branch ratio of different flux out of HIO_3-HIO_2-MSA-based system at (a) T = 278 K, [HIO_3] = 1.0 × 10^6, [HIO_2] = 2.0 × 10^4, [MSA] = 10^6 – 10^8 molec. cm^{-3}, and (b) T = 278 K, [HIO_3] = 10^6 – 10^8, [HIO_2] = 2.0 × 10^4 – 2.0 × 10^6, [MSA] = 1.0 × 10^7 molec. cm^{-3}.

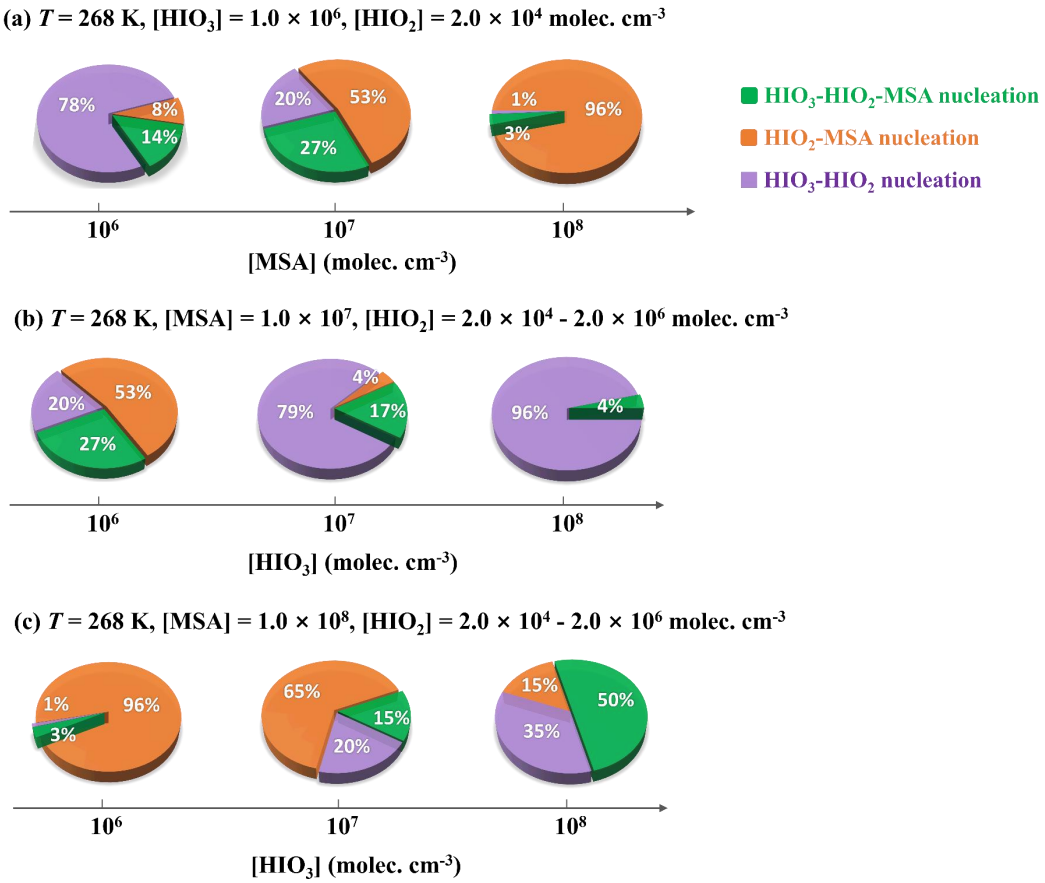


Figure S3. The branch ratio of different flux out of HIO_3-HIO_2-MSA-based system at (a) T = 268 K, $[HIO_3]$ = 1.0 × 10^6, $[HIO_2]$ = 2.0 × 10^4, [MSA] = 10^6 – 10^8 molec. cm^{-3}, (b) T = 268 K, $[HIO_3]$ = 10^6 – 10^8, $[HIO_2]$ = 2.0 × 10^4 – 2.0 × 10^6, [MSA] = 1.0 × 10^7 molec. cm^{-3}, and (c) T = 268 K, $[HIO_3]$ = 10^6 – 10^8, $[HIO_2]$ = 2.0 × 10^4 – 2.0 × 10^6, [MSA] = 1.0 × 10^8 molec. cm^{-3}.

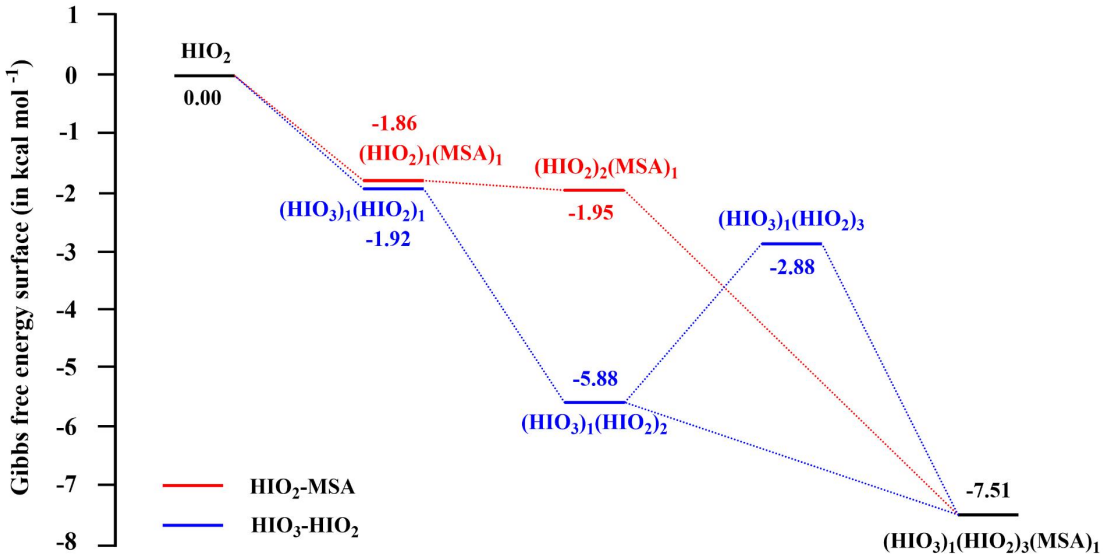


Figure S4. The Gibbs free energies of formation (ΔG, kcal mol^{-3}) of the growing clusters as a function of growth step in HIO_3-HIO_2-MSA nucleation system at T = 268 K, [HIO_3] = 1.0 × 10^6, [HIO_2] = 2.0 × 10^4, and [MSA] = 5.0 × 10^6 molec. cm^{-3}.

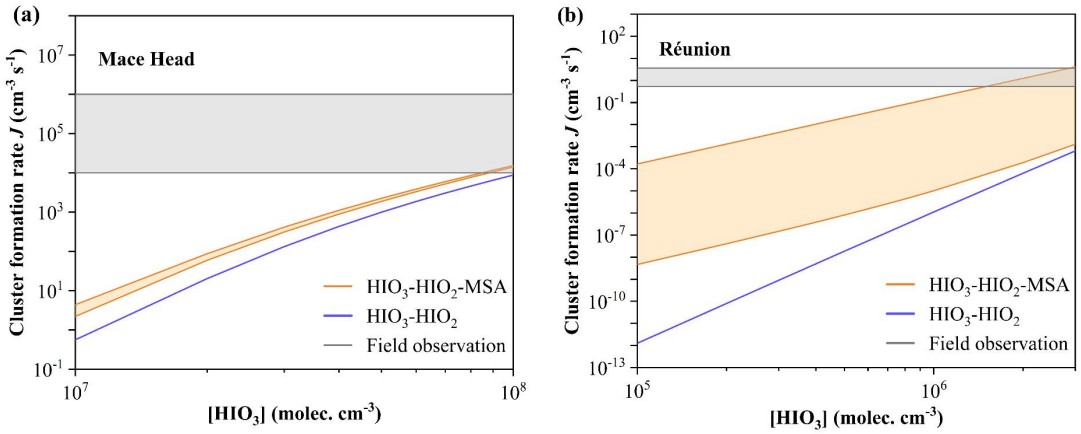


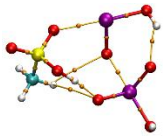
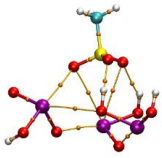
Figure S5. Comparison with the simulated cluster formation rates (J, cm^{-3} s^{-1}) and field observations at the ambient conditions of (a) Mace Head (T = 287 K, CS = 2.0×10^{-3} s^{-1}, $[HIO_3]$ = $10^7 - 10^8$, $[HIO_2]$ = $2.0 \times 10^5 - 2.0 \times 10^6$, and [MSA] = $10^6 - 10^7$ molec. cm^{-3}), (b) Réunion (T = 288 K, CS = 2.0×10^{-3} s^{-1}, $[HIO_3]$ = $10^5 - 3.0 \times 10^6$, $[HIO_2]$ = $2.0 \times 10^3 - 6.0 \times 10^4$, and [MSA] = $10^6 - 10^8$ molec. cm^{-3}). The orange area, purple line and gray area represent J(HIO_3-HIO_2-MSA), J(HIO_3-HIO_2), and J(Field observation).

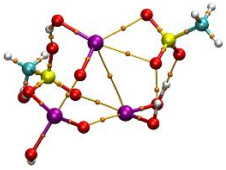
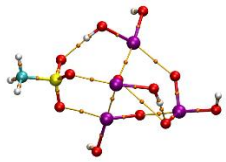
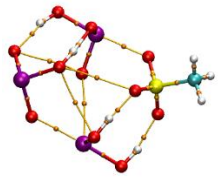
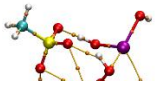
Table S1. The Gibbs formation free energies ΔG_{ref} (kcal mol^{-1}) of the studied HIO_3-HIO_2-MSA clusters at the RICC2/aug-cc-pV(T+d)Z(-PP)//ωB97X-D/6-311++G(3df,3pd) + aug-cc-pVTZ-PP with ECP28MDF (for I) level of theory, p = 1 atm and T = 268, 278, 288, and 298 K.

Clusters	ΔG_{ref} (kcal mol^{-1})			
	268K	278K	288K	298K
$(HIO_3)_2$	-8.45	-8.07	-7.70	-7.33
$(HIO_3)_3$	-19.26	-18.46	-17.68	-16.89
$(HIO_3)_4$	-39.70	-38.46	-37.26	-36.03
$(HIO_3)_5$	-57.93	-56.26	-54.64	-52.99
$(HIO_2)_2$	-18.05	-17.63	-17.22	-16.80
$(HIO_2)_3$	-39.72	-39.06	-38.02	-37.38
$(HIO_2)_4$	-59.98	-58.69	-57.45	-56.17
$(HIO_2)_5$	-77.63	-75.92	-74.26	-72.57
$(MSA)_2$	-11.37	-10.97	-10.57	-10.16
$(MSA)_3$	-16.88	-16.10	-15.33	-14.55
$(HIO_3)_1(HIO_2)_1$	-18.40	-17.99	-17.59	-17.18
$(HIO_3)_1(HIO_2)_2$	-40.62	-39.77	-38.94	-38.10
$(HIO_3)_1(HIO_2)_3$	-56.48	-55.20	-53.96	-52.70
$(HIO_3)_1(HIO_2)_4$	-82.51	-80.75	-79.05	-77.30
$(HIO_3)_2(HIO_2)_1$	-31.31	-30.46	-29.64	-28.79
$(HIO_3)_2(HIO_2)_2$	-56.42	-55.18	-53.97	-52.73
$(HIO_3)_2(HIO_2)_3$	-80.23	-78.52	-76.86	-75.16
$(HIO_3)_3(HIO_2)_1$	-52.92	-51.67	-50.45	-49.21
$(HIO_3)_3(HIO_2)_2$	-74.52	-72.80	-71.14	-69.44
$(HIO_3)_4(HIO_2)_1$	-66.16	-64.48	-62.84	-61.18
$(HIO_3)_1(MSA)_1$	-11.71	-11.31	-10.93	-10.54
$(HIO_3)_1(MSA)_2$	-20.56	-19.77	-19.00	-18.22
$(HIO_3)_1(MSA)_3$	-36.06	-34.81	-33.59	-32.34
$(HIO_3)_2(MSA)_1$	-21.80	-21.00	-20.22	-19.42
$(HIO_3)_2(MSA)_2$	-33.13	-31.89	-30.69	-29.46

$(HIO_3)_2(MSA)_3$	-48.73	-47.12	-45.55	-43.94
$(HIO_3)_3(MSA)_1$	-33.39	-32.23	-31.10	-29.94
$(HIO_3)_3(MSA)_2$	-53.59	-51.98	-50.41	-48.81
$(HIO_3)_4(MSA)_1$	-55.82	-54.12	-52.47	-50.78
$(HIO_2)_1(MSA)_1$	-17.48	-17.06	-16.66	-16.25
$(HIO_2)_1(MSA)_2$	-30.59	-29.78	-28.99	-28.18
$(HIO_2)_1(MSA)_3$	-41.85	-40.63	-39.44	-38.22
$(HIO_2)_2(MSA)_1$	-36.13	-35.30	-34.48	-33.65
$(HIO_2)_2(MSA)_2$	-58.70	-57.40	-56.14	-55.25
$(HIO_2)_2(MSA)_3$	-74.57	-72.87	-71.22	-69.52
$(HIO_2)_3(MSA)_1$	-59.74	-58.47	-57.24	-55.98
$(HIO_2)_3(MSA)_2$	-79.31	-77.63	-75.99	-74.31
$(HIO_2)_4(MSA)_1$	-77.90	-76.20	-74.54	-72.84
$(HIO_3)_1(HIO_2)_1(MSA)_1$	-30.42	-29.58	-28.77	-27.93
$(HIO_3)_1(HIO_2)_1(MSA)_2$	-45.90	-44.67	-43.48	-42.26
$(HIO_3)_1(HIO_2)_1(MSA)_3$	-60.92	-59.25	-57.62	-55.95
$(HIO_3)_1(HIO_2)_2(MSA)_1$	-50.60	-49.39	-48.21	-47.01
$(HIO_3)_1(HIO_2)_2(MSA)_2$	-71.42	-69.75	-68.13	-66.47
$(HIO_3)_1(HIO_2)_3(MSA)_1$	-76.73	-75.02	-73.35	-71.65
$(HIO_3)_2(HIO_2)_1(MSA)_1$	-46.33	-45.06	-43.82	-42.56
$(HIO_3)_2(HIO_2)_1(MSA)_2$	-62.32	-60.66	-59.04	-57.39
$(HIO_3)_2(HIO_2)_2(MSA)_1$	-65.80	-64.09	-62.43	-60.73
$(HIO_3)_3(HIO_2)_1(MSA)_1$	-65.83	-64.10	-62.42	-60.71

Table S2. The bond type, electron density $\rho(r)$ (a.u), Laplacian electron density $\nabla^2\rho(r)$ (a.u.), energy density $H(r)$ at the corresponding bond critical points (BCPs) in the studied HIO_3-HIO_2-MSA-based clusters. The orange balls represent BCPs in the AIM theory analysis. HIO_3, HIO_2, and MSA are the shorthand for iodic acid, iodous acid and methanesulfonic acid, respectively. HB (hydrogen bond), XB (halogen bond).

Cluster	Bond	Bond type	$\rho(r)$(a.u.)	$\nabla^2\rho(r)$(a.u.)	$H(r)$ (a.u.)
 $(HIO_3)_1(HIO_2)_1(MSA)_1$	O–H···O	HB	0.0691	0.1092	-0.0248
	O–H···O	HB	0.0224	0.0823	0.0018
	O–I···O	XB	0.0786	0.1509	-0.0213
	O–I···O	XB	0.0405	0.1239	-0.0016
 $(HIO_3)_1(HIO_2)_1(MSA)_2$	O–H···O	HB	0.0348	0.1086	-0.0021
	O–H···O	HB	0.0090	0.0310	0.0009
	O–H···O	HB	0.0582	0.1156	-0.0017
	O–I···O	XB	0.0429	0.1257	-0.0023
	O–I···O	XB	0.0476	0.1348	-0.0034
	O–I···O	XB	0.0299	0.0919	0.0003
 $(HIO_3)_1(HIO_2)_1(MSA)_3$	O–H···O	HB	0.0154	0.0620	0.0024
	O–H···O	HB	0.0521	0.1159	-0.0128
	O–H···O	HB	0.0495	0.1110	-0.0116
	O–H···O	HB	0.0783	0.0943	-0.0322
	O–I···O	XB	0.0114	0.0341	0.0006
	O–I···O	XB	0.0102	0.0288	0.0005
	O–I···O	XB	0.0190	0.0675	0.0019
	O–I···O	XB	0.0431	0.1244	-0.0025
	O–I···O	XB	0.0255	0.0873	0.0018
	O–I···O	XB	0.0381	0.1035	-0.0019
 $(HIO_3)_1(HIO_2)_2(MSA)_1$	O–H···O	HB	0.0644	0.1159	-0.0206
	O–H···O	HB	0.0208	0.0803	-0.0023
	O–I···O	XB	0.0449	0.1262	-0.0031
	O–I···O	XB	0.0119	0.0358	0.0009

	O–I···O	XB	0.0326	0.0952	0.0001
	O–I···O	XB	0.0116	0.0321	0.0004
	O–I···O	XB	0.0136	0.0391	0.0005
	O–H···O	HB	0.0242	0.0916	0.0018
	O–H···O	HB	0.0824	0.0964	-0.0349
	O–H···O	HB	0.0383	0.1096	-0.0041
	O–H···O	HB	0.0366	0.1111	-0.0031
	O–I···O	XB	0.0927	0.1744	-0.0307
$(HIO_3)_1(HIO_2)_2(MSA)_2$	O–I···O	XB	0.0250	0.0745	0.0003
	O–I···O	XB	0.0275	0.0784	-0.0002
	O–I···O	XB	0.0384	0.1231	-0.0006
	O–I···O	XB	0.0220	0.0724	0.0015
	O–H···O	HB	0.0453	0.1116	-0.0085
	O–H···O	HB	0.0429	0.1091	-0.0074
	O–I···O	XB	0.0247	0.0767	0.0009
	O–I···O	XB	0.0406	0.1109	-0.0023
	O–I···O	XB	0.0499	0.1345	-0.0051
$(HIO_3)_1(HIO_2)_3(MSA)_1$	O–I···O	XB	0.0794	0.1637	-0.0209
	O–I···O	XB	0.0648	0.1555	-0.0120
	O–I···O	XB	0.0283	0.0906	0.0011
	O–H···O	HB	0.0487	0.1180	-0.0103
	O–H···O	HB	0.0405	0.1131	-0.0054
	O–H···O	HB	0.0869	0.0835	-0.0394
	O–H···O	HB	0.0221	0.0793	0.0016
	O–I···O	XB	0.0386	0.1171	-0.0010
$(HIO_3)_2(HIO_2)_1(MSA)_1$	O–I···O	XB	0.0135	0.0422	0.0012
	O–I···O	XB	0.1999	0.4607	-0.1372
	O–H···O	HB	0.0482	0.1163	-0.0104
	O–H···O	HB	0.0268	0.1079	0.0023
	O–H···O	HB	0.0627	0.1092	-0.0204

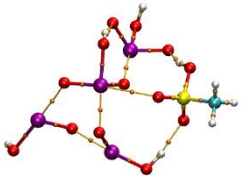
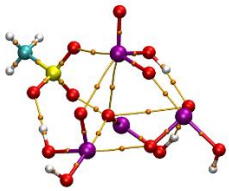
	O–H···O	HB	0.0586	0.1154	-0.0170
	O–I···O	XB	0.0122	0.0376	0.0009
	O–I···O	XB	0.0712	0.1462	-0.0165
	O–I···O	XB	0.0178	0.0554	0.0007
	O–I···O	XB	0.0215	0.0710	0.0008
	O–I···O	XB	0.0248	0.0802	0.0015
	O–I···O	XB	0.0607	0.1554	-0.0097
	O–I···O	XB	0.0068	0.0205	0.0007
 $(HIO_3)_2(HIO_2)_2(MSA)_1$	O–H···O	HB	0.0309	0.1010	-0.0010
	O–H···O	HB	0.0696	0.1038	-0.0254
	O–H···O	HB	0.0351	0.1040	-0.0029
	O–I···O	XB	0.0212	0.0675	0.0014
	O–I···O	XB	0.0911	0.1693	-0.0297
	O–I···O	XB	0.0644	0.1487	-0.0122
	O–I···O	XB	0.0548	0.1427	-0.0070
	O–I···O	XB	0.0621	0.1511	-0.0104
 $(HIO_3)_3(HIO_2)_1(MSA)_1$	O–H···O	HB	0.0356	0.1017	-0.0032
	O–H···O	HB	0.0336	0.1016	-0.0020
	O–H···O	HB	0.0542	0.1108	-0.0147
	O–I···O	XB	0.0258	0.0780	0.0010
	O–I···O	XB	0.0756	0.1668	-0.0181
	O–I···O	XB	0.0152	0.0451	0.0008
	O–I···O	XB	0.0230	0.0709	0.0010
	O–I···O	XB	0.0566	0.1467	-0.0079
	O–I···O	XB	0.0811	0.1587	-0.0225

Table S3. Boundary conditions at 268, 278, 288, and 298 K, respectively.

Temperature (K)	Boundary cluster
268 K	$(HIO_3)_6$
	$(HIO_3)_5(MSA)_1$
	$(HIO_3)_1(HIO_2)_5$
	$(HIO_2)_5(MSA)_1$
	$(HIO_3)_2(HIO_2)_4$
	$(HIO_3)_3(HIO_2)_3$
	$(HIO_2)_4(MSA)_2$
	$(HIO_2)_3(MSA)_3$
	$(HIO_3)_4(HIO_2)_2$
	$(HIO_3)_4(MSA)_2$
	$(HIO_3)_3(MSA)_3$
	$(HIO_3)_1(HIO_2)_3(MSA)_2$
	$(HIO_3)_3(HIO_2)_2(MSA)_1$
	$(HIO_3)_3(HIO_2)_1(MSA)_2$
	$(HIO_3)_1(HIO_2)_4(MSA)_1$
	$(HIO_3)_2(HIO_2)_3(MSA)_1$
278 K	$(HIO_3)_6$
	$(HIO_3)_5(MSA)_1$
	$(HIO_3)_2(HIO_2)_4$
	$(HIO_3)_1(HIO_2)_5$
	$(HIO_3)_3(HIO_2)_3$
	$(HIO_3)_4(HIO_2)_2$
	$(HIO_3)_4(MSA)_2$
	$(HIO_3)_3(MSA)_3$
	$(HIO_2)_4(MSA)_2$
	$(HIO_2)_5(MSA)_1$
	$(HIO_2)_3(MSA)_3$

	$(HIO_3)_1(HIO_2)_3(MSA)_2$
	$(HIO_3)_3(HIO_2)_1(MSA)_2$
	$(HIO_3)_3(HIO_2)_2(MSA)_1$
	$(HIO_3)_2(HIO_2)_3(MSA)_1$
	$(HIO_3)_1(HIO_2)_4(MSA)_1$
288 K	$(HIO_3)_2(HIO_2)_4$
	$(HIO_3)_1(HIO_2)_5$
	$(HIO_3)_3(HIO_2)_3$
	$(HIO_3)_4(MSA)_2$
	$(HIO_3)_3(MSA)_3$
	$(HIO_2)_3(MSA)_3$
	$(HIO_3)_1(HIO_2)_4(MSA)_1$
	$(HIO_3)_2(HIO_2)_3(MSA)_1$
	$(HIO_3)_1(HIO_2)_3(MSA)_2$
298 K	$(HIO_3)_2(HIO_2)_4$
	$(HIO_3)_1(HIO_2)_5$
	$(HIO_3)_3(HIO_2)_3$
	$(HIO_3)_4(MSA)_2$
	$(HIO_3)_3(MSA)_3$
	$(HIO_2)_3(MSA)_3$
	$(HIO_3)_1(HIO_2)_4(MSA)_1$
	$(HIO_3)_2(HIO_2)_3(MSA)_1$

Table S4. The ratios of HIO_3 monomer collision frequencies versus total evaporation rate coefficients ($\beta C/\Sigma\gamma$) at T = 268, 278, 288, and 298 K. β is the rate coefficient of cluster collision with HIO_3 monomer, and C is the concentration of HIO_3 monomer (1.0×10^6 molecules cm^{-3}).

Clusters	$\beta C/\Sigma\gamma$			
	268K	278K	288K	298K
$(HIO_3)_2$	6.08×10^{-7}	1.79×10^{-7}	5.86×10^{-8}	2.10×10^{-8}
$(HIO_3)_3$	2.61×10^{-5}	6.09×10^{-6}	1.60×10^{-6}	4.55×10^{-7}
$(HIO_3)_4$	1.83×10^{3}	2.15×10^{2}	3.05×10^{1}	4.84×10^{0}
$(HIO_3)_5$	2.86×10^{1}	3.97×10^{0}	6.46×10^{-1}	1.22×10^{-1}
$(HIO_2)_2$	4.10×10^{1}	5.87×10^{0}	9.81×10^{-1}	1.85×10^{-1}
$(HIO_2)_3$	1.86×10^{4}	2.89×10^{3}	2.59×10^{2}	3.77×10^{1}
$(HIO_2)_4$	1.30×10^{1}	1.10×10^{2}	2.33×10^{1}	3.80×10^{0}
$(HIO_2)_5$	9.54×10^{0}	1.41×10^{0}	2.37×10^{-1}	6.16×10^{-2}
$(MSA)_2$	1.21×10^{-4}	2.83×10^{-5}	7.31×10^{-6}	2.07×10^{-6}
$(MSA)_3$	9.92×10^{-10}	3.57×10^{-10}	1.40×10^{-10}	5.87×10^{-11}
$(HIO_3)_1(HIO_2)_1$	1.98×10^{1}	2.83×10^{0}	4.70×10^{-1}	8.68×10^{-2}
$(HIO_3)_1(HIO_2)_2$	3.45×10^{4}	3.59×10^{3}	4.45×10^{2}	6.44×10^{1}
$(HIO_3)_1(HIO_2)_3$	2.82×10^{-1}	4.27×10^{-2}	8.71×10^{-3}	1.83×10^{-3}
$(HIO_3)_1(HIO_2)_4$	9.20×10^{4}	8.90×10^{3}	1.03×10^{3}	1.40×10^{2}
$(HIO_3)_2(HIO_2)_1$	1.33×10^{-3}	2.61×10^{-4}	5.92×10^{-5}	1.46×10^{-5}
$(HIO_3)_2(HIO_2)_2$	3.03×10^{-1}	5.34×10^{-2}	1.08×10^{-2}	2.36×10^{-3}
$(HIO_3)_2(HIO_2)_3$	4.78×10^{5}	4.41×10^{4}	4.93×10^{3}	6.34×10^{2}
$(HIO_3)_3(HIO_2)_1$	1.67×10^{4}	1.95×10^{3}	2.65×10^{2}	4.19×10^{1}
$(HIO_3)_3(HIO_2)_2$	2.24×10^{1}	2.87×10^{0}	4.47×10^{-1}	8.00×10^{-2}
$(HIO_3)_4(HIO_2)_1$	2.19×10^{-3}	4.73×10^{-4}	1.06×10^{-4}	2.67×10^{-5}
$(HIO_3)_1(MSA)_1$	6.08×10^{-5}	1.39×10^{-5}	3.63×10^{-6}	1.04×10^{-6}
$(HIO_3)_1(MSA)_2$	3.63×10^{-7}	1.01×10^{-7}	3.14×10^{-8}	1.05×10^{-8}
$(HIO_3)_1(MSA)_3$	1.33×10^{-1}	2.11×10^{-2}	3.87×10^{-3}	7.83×10^{-4}
$(HIO_3)_2(MSA)_1$	6.50×10^{-6}	1.65×10^{-6}	4.63×10^{-7}	4.24×10^{-10}
$(HIO_3)_2(MSA)_2$	4.98×10^{-5}	1.07×10^{-5}	2.67×10^{-6}	7.33×10^{-6}

$(HIO_3)_2(MSA)_3$	8.22×10^{-4}	1.89×10^{-4}	4.88×10^{-5}	1.40×10^{-5}
$(HIO_3)_3(MSA)_1$	1.09×10^{-4}	2.70×10^{-5}	7.48×10^{-6}	1.12×10^{-4}
$(HIO_3)_3(MSA)_2$	6.03×10^{2}	7.36×10^{1}	1.05×10^{1}	1.71×10^{0}
$(HIO_3)_4(MSA)_1$	4.20×10^{-1}	6.46×10^{-2}	1.13×10^{-2}	2.22×10^{-3}
$(HIO_2)_1(MSA)_1$	3.11×10^{0}	4.63×10^{-1}	8.17×10^{-2}	1.59×10^{-2}
$(HIO_2)_1(MSA)_2$	1.59×10^{-3}	3.36×10^{-4}	7.91×10^{-5}	2.06×10^{-5}
$(HIO_2)_1(MSA)_3$	4.75×10^{-5}	1.10×10^{-5}	2.86×10^{-6}	8.15×10^{-7}
$(HIO_2)_2(MSA)_1$	1.41×10^{1}	2.03×10^{0}	3.35×10^{-1}	6.25×10^{-2}
$(HIO_2)_2(MSA)_2$	7.94×10^{4}	7.66×10^{3}	9.17×10^{2}	1.23×10^{2}
$(HIO_2)_2(MSA)_3$	2.68×10^{-1}	4.56×10^{-2}	9.13×10^{-3}	1.82×10^{-3}
$(HIO_2)_3(MSA)_1$	6.65×10^{2}	5.91×10^{1}	1.30×10^{1}	2.25×10^{0}
$(HIO_2)_3(MSA)_2$	2.49×10^{2}	3.27×10^{1}	4.95×10^{0}	1.11×10^{0}
$(HIO_2)_4(MSA)_1$	8.33×10^{0}	1.20×10^{0}	1.97×10^{-1}	7.22×10^{-2}
$(HIO_3)_1(HIO_2)_1(MSA)_1$	1.79×10^{-4}	3.76×10^{-5}	9.10×10^{-6}	2.39×10^{-6}
$(HIO_3)_1(HIO_2)_1(MSA)_2$	6.18×10^{-2}	1.09×10^{-2}	2.22×10^{-3}	5.08×10^{-4}
$(HIO_3)_1(HIO_2)_1(MSA)_3$	5.43×10^{-2}	9.21×10^{-3}	1.77×10^{-3}	3.80×10^{-4}
$(HIO_3)_1(HIO_2)_2(MSA)_1$	4.30×10^{-6}	1.19×10^{-6}	3.64×10^{-7}	1.21×10^{-7}
$(HIO_3)_1(HIO_2)_2(MSA)_2$	9.18×10^{-4}	2.06×10^{-4}	5.25×10^{-5}	1.46×10^{-5}
$(HIO_3)_1(HIO_2)_3(MSA)_1$	2.77×10^{0}	4.12×10^{-1}	6.98×10^{-2}	1.34×10^{-2}
$(HIO_3)_2(HIO_2)_1(MSA)_1$	4.83×10^{-2}	8.40×10^{-3}	1.65×10^{-3}	3.75×10^{-4}
$(HIO_3)_2(HIO_2)_1(MSA)_2$	2.46×10^{-1}	4.16×10^{-2}	8.05×10^{-3}	1.74×10^{-3}
$(HIO_3)_2(HIO_2)_2(MSA)_1$	1.35×10^{-6}	3.20×10^{-7}	8.53×10^{-8}	2.59×10^{-8}
$(HIO_3)_3(HIO_2)_1(MSA)_1$	1.02×10^{-3}	1.86×10^{-4}	3.94×10^{-5}	9.29×10^{-6}

Table S5. The evaporation rate coefficients (γ, s^{-1}) for all evaporation pathways of clusters at 278 K.

Evaporation pathways	Evaporation rate coefficients (γ, s^{-1})
$(HIO_3)_2 \rightarrow HIO_3 + HIO_3$	1.48×10^{3}
$(HIO_3)_3 \rightarrow (HIO_3)_2 + HIO_3$	4.76×10^{1}
$(HIO_3)_4 \rightarrow (HIO_3)_3 + HIO_3$	1.45×10^{-6}
$(HIO_3)_5 \rightarrow (HIO_3)_4 + HIO_3$	8.37×10^{-5}
$(HIO_2)_2 \rightarrow HIO_2 + HIO_2$	4.40×10^{-5}
$(HIO_2)_3 \rightarrow (HIO_2)_2 + HIO_2$	9.68×10^{-8}
$(HIO_2)_4 \rightarrow (HIO_2)_3 + HIO_2$	2.75×10^{-6}
$(HIO_2)_5 \rightarrow (HIO_2)_4 + HIO_2$	2.29×10^{-4}
$(MSA)_2 \rightarrow MSA + MSA$	1.11×10^{1}
$(MSA)_3 \rightarrow (MSA)_2 + MSA$	9.46×10^{5}
$(HIO_3)_1(HIO_2)_1 \rightarrow HIO_3 + HIO_2$	9.30×10^{-5}
$(HIO_3)_1(HIO_2)_2 \rightarrow HIO_3 + (HIO_2)_2$	2.68×10^{-8}
$(HIO_3)_1(HIO_2)_2 \rightarrow (HIO_3)_1(HIO_2)_1 + HIO_2$	5.24×10^{-8}
$(HIO_3)_1(HIO_2)_3 \rightarrow HIO_3 + (HIO_2)_3$	1.52×10^{-3}
$(HIO_3)_1(HIO_2)_3 \rightarrow (HIO_3)_1(HIO_2)_2 + HIO_2$	5.59×10^{-3}
$(HIO_3)_1(HIO_2)_4 \rightarrow HIO_3 + (HIO_2)_4$	3.62×10^{-8}
$(HIO_3)_1(HIO_2)_4 \rightarrow (HIO_3)_1(HIO_2)_3 + HIO_2$	6.64×10^{-11}
$(HIO_3)_2(HIO_2)_1 \rightarrow HIO_3 + (HIO_3)_1(HIO_2)_1$	1.09×10^{0}
$(HIO_3)_2(HIO_2)_1 \rightarrow (HIO_3)_2 + HIO_2$	1.75×10^{-8}
$(HIO_3)_2(HIO_2)_2 \rightarrow HIO_3 + (HIO_3)_1(HIO_2)_2$	5.77×10^{-3}
$(HIO_3)_2(HIO_2)_2 \rightarrow (HIO_3)_2(HIO_2)_1 + HIO_2$	2.79×10^{-10}
$(HIO_3)_2(HIO_2)_3 \rightarrow HIO_3 + (HIO_3)_1(HIO_2)_3$	3.73×10^{-9}
$(HIO_3)_2(HIO_2)_3 \rightarrow (HIO_3)_2(HIO_2)_2 + HIO_2$	3.68×10^{-9}
$(HIO_3)_3(HIO_2)_1 \rightarrow HIO_3 + (HIO_3)_2(HIO_2)_1$	1.59×10^{-7}
$(HIO_3)_3(HIO_2)_1 \rightarrow (HIO_3)_3 + HIO_2$	6.01×10^{-17}
$(HIO_3)_3(HIO_2)_2 \rightarrow HIO_3 + (HIO_3)_2(HIO_2)_2$	1.14×10^{-4}
$(HIO_3)_3(HIO_2)_2 \rightarrow (HIO_3)_3(HIO_2)_1 + HIO_2$	2.03×10^{-7}

$(HIO_3)_4(HIO_2)_1 \rightarrow HIO_3 + (HIO_3)_3(HIO_2)_1$	6.98×10^{-1}
$(HIO_3)_4(HIO_2)_1 \rightarrow (HIO_3)_4 + HIO_2$	2.92×10^{-11}
$(HIO_3)_1(MSA)_1 \rightarrow HIO_3 + MSA$	1.03×10^{1}
$(HIO_3)_1(MSA)_2 \rightarrow HIO_3 + (MSA)_2$	1.00×10^{3}
$(HIO_3)_1(MSA)_2 \rightarrow (HIO_3)_1(MSA)_1 + MSA$	2.12×10^{3}
$(HIO_3)_1(MSA)_3 \rightarrow HIO_3 + (MSA)_3$	1.74×10^{-5}
$(HIO_3)_1(MSA)_3 \rightarrow (HIO_3)_1(MSA)_2 + MSA$	1.61×10^{-2}
$(HIO_3)_2(MSA)_1 \rightarrow HIO_3 + (HIO_3)_1(MSA)_1$	1.82×10^{2}
$(HIO_3)_2(MSA)_1 \rightarrow (HIO_3)_2 + MSA$	6.13×10^{-1}
$(HIO_3)_2(MSA)_2 \rightarrow HIO_3 + (HIO_3)_1(MSA)_2$	2.48×10^{0}
$(HIO_3)_2(MSA)_2 \rightarrow (HIO_3)_2(MSA)_1 + MSA$	2.83×10^{1}
$(HIO_3)_2(MSA)_3 \rightarrow HIO_3 + (HIO_3)_1(MSA)_3$	1.88×10^{0}
$(HIO_3)_2(MSA)_3 \rightarrow (HIO_3)_2(MSA)_2 + MSA$	1.21×10^{-2}
$(HIO_3)_3(MSA)_1 \rightarrow HIO_3 + (HIO_3)_2(MSA)_1$	1.18×10^{1}
$(HIO_3)_3(MSA)_1 \rightarrow (HIO_3)_3 + MSA$	1.49×10^{-1}
$(HIO_3)_3(MSA)_2 \rightarrow HIO_3 + (HIO_3)_2(MSA)_2$	1.40×10^{-6}
$(HIO_3)_3(MSA)_2 \rightarrow (HIO_3)_3(MSA)_1 + MSA$	3.33×10^{-6}
$(HIO_3)_4(MSA)_1 \rightarrow HIO_3 + (HIO_3)_3(MSA)_1$	5.28×10^{-8}
$(HIO_3)_4(MSA)_1 \rightarrow (HIO_3)_4 + MSA$	5.32×10^{-3}
$(HIO_2)_1(MSA)_1 \rightarrow HIO_2 + MSA$	3.05×10^{-4}
$(HIO_2)_1(MSA)_2 \rightarrow HIO_2 + (MSA)_2$	1.35×10^{-5}
$(HIO_2)_1(MSA)_2 \rightarrow (HIO_2)_1(MSA)_1 + MSA$	9.35×10^{-1}
$(HIO_2)_1(MSA)_3 \rightarrow HIO_2 + (MSA)_3$	4.63×10^{-10}
$(HIO_2)_1(MSA)_3 \rightarrow (HIO_2)_1(MSA)_2 + MSA$	3.13×10^{1}
$(HIO_3)_2(MSA)_1 \rightarrow HIO_2 + (HIO_2)_1(MSA)_1$	3.40×10^{-5}
$(HIO_2)_2(MSA)_1 \rightarrow (HIO_2)_2 + MSA$	1.12×10^{-4}
$(HIO_2)_2(MSA)_2 \rightarrow HIO_2 + (HIO_2)_1(MSA)_2$	1.61×10^{-12}
$(HIO_2)_2(MSA)_2 \rightarrow (HIO_2)_2(MSA)_1 + MSA$	4.26×10^{-8}
$(HIO_2)_2(MSA)_3 \rightarrow HIO_2 + (HIO_2)_1(MSA)_3$	4.12×10^{-16}
$(HIO_2)_2(MSA)_3 \rightarrow (HIO_2)_2(MSA)_2 + MSA$	7.73×10^{-3}

$(HIO_2)_3(MSA)_1 \rightarrow HIO_2 + (HIO_2)_2(MSA)_1$	4.78×10^{-9}
$(HIO_2)_3(MSA)_1 \rightarrow (HIO_2)_3 + MSA$	5.30×10^{-6}
$(HIO_2)_3(MSA)_2 \rightarrow HIO_2 + (HIO_2)_2(MSA)_2$	1.08×10^{-6}
$(HIO_2)_3(MSA)_2 \rightarrow (HIO_2)_3(MSA)_1 + MSA$	9.38×10^{-6}
$(HIO_2)_4(MSA)_1 \rightarrow HIO_2 + (HIO_2)_3(MSA)_1$	9.62×10^{-5}
$(HIO_2)_4(MSA)_1 \rightarrow (HIO_2)_4 + MSA$	1.80×10^{-4}
$(HIO_3)_1(HIO_2)_1(MSA)_1 \rightarrow HIO_3 + (HIO_2)_1\ (MSA)_1$	1.07×10^{0}
$(HIO_3)_1(HIO_2)_1(MSA)_1 \rightarrow HIO_2 + (HIO_3)_1(MSA)_1$	3.26×10^{-5}
$(HIO_3)_1(HIO_2)_1(MSA)_1 \rightarrow MSA + (HIO_3)_1(HIO_2)_1$	6.86×10^{0}
$(HIO_3)_1(HIO_2)_1(MSA)_2 \rightarrow HIO_3 + (HIO_2)_1(MSA)_2$	1.63×10^{-2}
$(HIO_3)_1(HIO_2)_1(MSA)_2 \rightarrow HIO_2 + (HIO_3)_1(MSA)_2$	2.23×10^{-10}
$(HIO_3)_1(HIO_2)_1(MSA)_2 \rightarrow MSA + (HIO_3)_1(HIO_2)_1(MSA)_1$	1.39×10^{-2}
$(HIO_3)_1(HIO_2)_1(MSA)_3 \rightarrow HIO_3 + (HIO_2)_1(MSA)_3$	2.08×10^{-5}
$(HIO_3)_1(HIO_2)_1(MSA)_3 \rightarrow HIO_2 + (HIO_3)_1(MSA)_3$	5.52×10^{-10}
$(HIO_3)_1(HIO_2)_1(MSA)_3 \rightarrow MSA + (HIO_3)_1(HIO_2)_1(MSA)_2$	3.91×10^{-2}
$(HIO_3)_1(HIO_2)_2(MSA)_1 \rightarrow HIO_3 + (HIO_2)_2(MSA)_1$	6.55×10^{-2}
$(HIO_3)_1(HIO_2)_2(MSA)_1 \rightarrow HIO_2 + (HIO_3)_1(HIO_2)_1(MSA)_1$	2.11×10^{-6}
$(HIO_3)_1(HIO_2)_2(MSA)_1 \rightarrow MSA + (HIO_3)_1(HIO_2)_2$	2.67×10^{2}
$(HIO_3)_1(HIO_2)_2(MSA)_2 \rightarrow HIO_3 + (HIO_2)_2(MSA)_2$	1.68×10^{0}
$(HIO_3)_1(HIO_2)_2(MSA)_2 \rightarrow HIO_2 + (HIO_3)_1(HIO_2)_1(MSA)_2$	1.68×10^{-10}
$(HIO_3)_1(HIO_2)_2(MSA)_2 \rightarrow MSA + (HIO_3)_1(HIO_2)_2(MSA)_1$	1.08×10^{-6}
$(HIO_3)_1(HIO_2)_3(MSA)_1 \rightarrow HIO_3 + (HIO_2)_3(MSA)_1$	8.08×10^{-4}
$(HIO_3)_1(HIO_2)_3(MSA)_1 \rightarrow HIO_2 + (HIO_3)_1(HIO_2)_2(MSA)_1$	5.99×10^{-11}
$(HIO_3)_1(HIO_2)_3(MSA)_1 \rightarrow MSA + (HIO_3)_1(HIO_2)_3$	2.77×10^{-6}
$(HIO_3)_2(HIO_2)_1(MSA)_1 \rightarrow HIO_3 + (HIO_3)_1(HIO_2)_1(MSA)_1$	5.32×10^{-3}
$(HIO_3)_2(HIO_2)_1(MSA)_1 \rightarrow HIO_2 + (HIO_3)_2(MSA)_1$	9.73×10^{-10}
$(HIO_3)_2(HIO_2)_1(MSA)_1 \rightarrow MSA + (HIO_3)_2(HIO_2)_1$	3.26×10^{-2}
$(HIO_3)_2(HIO_2)_1(MSA)_2 \rightarrow HIO_3 + (HIO_3)_1(HIO_2)_1(MSA)_2$	2.34×10^{-3}
$(HIO_3)_2(HIO_2)_1(MSA)_2 \rightarrow HIO_2 + (HIO_3)_2(MSA)_2$	2.12×10^{-13}
$(HIO_3)_2(HIO_2)_1(MSA)_2 \rightarrow MSA + (HIO_3)_2(HIO_2)_1(MSA)_1$	6.02×10^{-3}

$(HIO_3)_2(HIO_2)_2(MSA)_1 \rightarrow HIO_3 + (HIO_3)_1(HIO_2)_2(MSA)_1$	2.33×10^{-2}
$(HIO_3)_2(HIO_2)_2(MSA)_1 \rightarrow HIO_2 + (HIO_3)_2(HIO_2)_1(MSA)_1$	9.31×10^{-6}
$(HIO_3)_2(HIO_2)_2(MSA)_1 \rightarrow MSA + (HIO_3)_2(HIO_2)_2$	1.06×10^{3}
$(HIO_3)_3(HIO_2)_1(MSA)_1 \rightarrow HIO_3 + (HIO_3)_2(HIO_2)_1(MSA)_1$	9.06×10^{-6}
$(HIO_3)_3(HIO_2)_1(MSA)_1 \rightarrow HIO_2 + (HIO_3)_3(MSA)_1$	7.06×10^{-16}
$(HIO_3)_3(HIO_2)_1(MSA)_1 \rightarrow MSA + (HIO_3)_3(HIO_2)_1$	1.83×10^{0}

Table S6. Cartesian coordinates of all clusters in the present study at the ωB97X-D/6-311++G(3df,3pd) + aug-cc-pVTZ-PP with ECP28MDF (for I) level of theory.

$(HIO_3)_1(HIO_2)_1(MSA)_1$:

Atoms	X	Y	Z
I	-1.877771	-0.752509	0.232491
O	-0.595483	-1.823796	-0.441557
O	-2.129309	0.529646	-0.990503
O	-3.347446	-1.887376	-0.363133
H	-3.316493	-2.009666	-1.318151
I	0.538518	1.786822	-0.007683
O	-0.280369	0.538764	1.127138
O	-0.982028	2.977162	-0.164915
H	-1.650294	2.452812	-0.639218
C	2.381400	-2.002110	1.293918
H	1.349560	-1.779093	1.553529
H	3.055380	-1.690334	2.087356
H	2.513617	-3.055293	1.064019
S	2.803335	-1.068186	-0.135331
O	4.109943	-1.391523	-0.576780
O	2.557737	0.329273	0.190156
O	1.799377	-1.531930	-1.201161
H	0.827546	-1.555140	-0.894535

$(HIO_3)_1(HIO_2)_1(MSA)_2$:

Atoms	X	Y	Z
I	2.409779	0.774129	-0.040206
O	1.385502	0.576253	1.430168
O	1.373911	1.601378	-1.214087
O	3.392857	2.316656	0.600986
H	2.789785	3.054133	0.751657
I	-0.952983	-0.240914	1.639934
O	-0.192173	-1.842568	2.353516
O	-2.791548	-0.834672	1.828123
H	-3.101196	-1.123232	0.961065
C	-1.635090	1.941809	-2.395478
H	-1.580410	3.023401	-2.301298
H	-2.078867	1.657620	-3.345129
H	-0.648400	1.503914	-2.254014
S	-2.689253	1.381700	-1.107970
O	-4.018312	1.835580	-1.303795

O	-2.052011	1.693605	0.151464
O	-2.703961	-0.170078	-1.294340
H	-1.806699	-0.574127	-1.495560
C	1.026057	-3.296909	-2.467723
H	1.757678	-3.951230	-2.002419
H	1.462361	-2.757735	-3.303648
H	0.154792	-3.862035	-2.787581
S	0.493560	-2.124785	-1.266745
O	-0.450874	-1.245829	-1.942264
O	1.721432	-1.408995	-0.868687
O	-0.082162	-2.871797	-0.166188
H	-0.035276	-2.407404	1.566781

$(HIO_3)_1(HIO_2)_1(MSA)_3$:

Atoms	X	Y	Z
I	-0.704502	1.880683	-0.087693
O	-0.262147	0.962564	-1.568210
O	0.769896	2.880001	0.140275
O	-1.718617	3.284402	-0.904918
H	-2.597548	2.909382	-1.066314
I	0.237573	-1.482650	-1.552860
O	-1.493460	-1.862291	-2.239184
O	0.793149	-3.313365	-1.239210
H	0.691830	-3.449932	-0.285722
C	-5.131422	-0.202325	0.675597
H	-5.602298	-0.293691	-0.299047
H	-5.397670	0.738529	1.148982
H	-5.404847	-1.040327	1.311180
S	-3.380752	-0.239309	0.457515
O	-2.769644	-0.081762	1.758671
O	-3.073136	0.902025	-0.429969
O	-3.081940	-1.512628	-0.184028
H	-2.145756	-1.747538	-1.495636
C	-0.352326	-1.151292	3.638131
H	-0.161837	-2.038803	4.234685
H	-1.363857	-1.142807	3.232807
H	-0.156843	-0.242376	4.199162
S	0.707021	-1.179657	2.247421
O	0.510522	0.019993	1.474837
O	0.561143	-2.434754	1.578032
O	2.110374	-1.088098	2.898449
H	2.710461	-0.516583	2.343792
C	5.133324	0.642192	-0.731776

H	5.048734	0.835176	-1.797054
H	5.572150	-0.335132	-0.549498
H	5.703289	1.421137	-0.234095
S	3.517878	0.610806	-0.048159
O	3.642807	0.423119	1.378359
O	2.734434	-0.371353	-0.743488
O	3.070689	2.029833	-0.374786
H	2.095029	2.304867	-0.152704

$(HIO_3)_1(HIO_2)_2(MSA)_1$:

Atoms	X	Y	Z
I	2.348350	-0.769952	-0.370994
O	1.654492	-0.933432	1.291176
O	3.963063	-0.109532	-0.099351
O	2.813854	-2.636535	-0.606809
H	3.520811	-2.864712	0.009545
I	-2.435446	-1.030080	-0.532754
O	-0.675580	-1.017821	-1.293881
O	-3.255475	-1.247275	-2.322592
H	-3.259906	-0.405511	-2.787043
I	-0.311703	0.399257	2.003701
O	-1.453783	-0.979816	1.421811
O	-1.672202	1.741859	2.352406
H	-1.643425	2.304136	1.558194
C	0.742195	3.687612	-2.070912
H	1.452733	3.361092	-2.824708
H	1.213050	4.370788	-1.369626
H	-0.127171	4.148493	-2.531313
S	0.197366	2.274013	-1.170936
O	-0.746074	2.744114	-0.175752
O	1.404151	1.671612	-0.591398
O	-0.416216	1.381600	-2.162983
H	-0.560623	-0.101299	-1.693138

$(HIO_3)_1(HIO_2)_2(MSA)_2$:

Atoms	X	Y	Z
I	-2.274708	-1.314077	-1.069938
O	-1.280980	0.083594	-1.611998
O	-2.834872	-0.822265	0.570168
O	-3.863092	-0.664896	-2.016261
H	-3.926249	0.294889	-1.977898

I	0.632696	-1.618253	1.095082
O	-0.580722	-2.309479	-0.180893
O	-0.456420	-1.721741	2.682021
H	-0.891987	-0.855016	2.762242
I	0.688431	1.614509	-1.302574
O	2.141989	2.852309	-1.109255
O	1.534733	0.503037	-2.608748
H	2.035517	-0.157807	-2.078329
C	-2.210521	3.345329	2.878658
H	-2.770544	4.077303	2.303823
H	-1.320249	3.795666	3.309752
H	-2.828191	2.892116	3.648119
S	-1.664285	2.084478	1.787508
O	-1.022339	1.045238	2.543856
O	-0.886171	2.707518	0.750861
O	-2.999114	1.580531	1.225491
H	-2.947752	0.587627	0.928655
C	5.145879	-1.203340	0.025882
H	5.462625	-1.041750	-1.000725
H	5.004780	-2.261776	0.225378
H	5.862700	-0.773971	0.720608
S	3.604091	-0.383395	0.266973
O	3.190244	-0.626641	1.636474
O	2.649949	-1.030216	-0.671988
O	3.827983	1.010367	-0.061212
H	2.840580	2.359109	-0.624881

$(HIO_3)_1(HIO_2)_3(MSA)_1$:

Atoms	X	Y	Z
I	3.100541	-0.494953	0.054224
O	2.179606	-1.070771	1.483798
O	2.472547	1.147459	-0.315090
O	4.647763	0.187051	1.046983
H	4.402718	0.972847	1.546784
I	0.062420	2.172851	-0.950176
O	-1.809613	2.599934	-0.820501
O	0.419481	3.698110	-2.140759
H	0.228934	3.455910	-3.050887
I	-1.082332	0.306963	2.092020
O	-0.525627	0.313681	0.284389
O	0.668997	0.672503	2.819187
H	1.295934	-0.000699	2.454745
I	-0.185243	-1.901950	-0.896517

O	1.558417	-1.340313	-1.301383
O	0.003276	-3.716490	-1.607974
H	-0.125092	-3.696140	-2.560272
C	-5.285320	-0.812393	-0.216000
H	-5.786708	0.143010	-0.091180
H	-5.534282	-1.491748	0.594069
H	-5.532742	-1.254333	-1.177365
S	-3.545561	-0.530966	-0.191771
O	-2.903926	-1.828621	-0.308546
O	-3.290245	0.092168	1.128168
O	-3.248350	0.375639	-1.288410
H	-2.341954	1.803875	-1.066678

$(HIO_3)_2(HIO_2)_1(MSA)_1$:

Atoms	X	Y	Z
I	-2.773171	-0.569794	0.577742
O	-3.115255	0.810216	-0.492635
O	-1.860841	-1.790462	-0.379870
O	-1.500766	0.059435	1.789981
H	-1.001840	0.929412	1.478961
I	0.168997	2.377382	-0.567292
O	-0.382966	2.188189	1.138075
O	0.041412	0.752066	-1.270487
O	-1.481935	3.081061	-1.270734
H	-2.168376	2.402925	-1.129338
I	0.590524	-2.309932	-0.694268
O	0.649959	-2.383007	1.206637
O	2.421517	-2.794143	-1.001080
H	2.934821	-1.967440	-0.840170
C	4.280227	0.941448	1.692637
H	3.934820	1.617441	2.469557
H	5.024361	1.422211	1.063883
H	4.676566	0.026750	2.125015
S	2.901359	0.510132	0.682594
O	3.401261	-0.377603	-0.351589
O	2.430470	1.796956	0.137610
O	1.915552	-0.104451	1.559334
H	1.072665	-1.523873	1.475161

$(HIO_3)_2(HIO_2)_1(MSA)_2$:

Atoms	X	Y	Z

I	1.093782	-1.924547	-1.130100
O	0.788916	-2.106659	0.671617
O	2.174892	-3.259322	-1.508651
O	-0.532941	-2.693983	-1.742588
H	-1.290898	-2.408139	-1.167747
I	-0.260065	-0.431970	1.808869
O	-0.040004	0.373751	0.174644
O	1.270762	-0.102861	2.655767
O	-1.103125	1.122009	2.601535
H	-2.038169	1.063428	2.341988
I	-0.816885	2.364153	-0.728820
O	-1.345194	1.183577	-2.120771
O	-1.576969	3.955428	-1.584113
H	-0.997674	4.261083	-2.287831
C	-4.943314	-1.579588	0.211887
H	-5.685012	-0.789147	0.284178
H	-4.933230	-2.183615	1.114711
H	-5.126029	-2.197329	-0.662966
S	-3.359337	-0.831759	0.033773
O	-2.382689	-1.918557	-0.030515
O	-3.139910	-0.018473	1.224822
O	-3.424062	-0.045957	-1.192370
H	-2.173130	0.707090	-1.814150
C	5.156706	1.385513	-0.097012
H	5.394987	2.292011	0.451764
H	5.308485	1.532078	-1.163093
H	5.735971	0.540665	0.262797
S	3.451272	1.032543	0.125257
O	3.167615	-0.230034	-0.510885
O	2.670833	2.156196	-0.287817
O	3.422602	0.866210	1.656187
H	2.555533	0.478486	2.004433

$(HIO_3)_2(HIO_2)_2(MSA)_1$:

Atoms	X	Y	Z
I	1.638876	2.217202	-0.876994
O	3.203167	1.345169	-1.008169
O	1.645011	2.991477	0.731909
O	2.207906	3.789233	-1.861349
H	2.927213	4.227571	-1.392465
I	0.047108	-0.140936	1.660468
O	-1.424765	0.926820	1.618775

O	0.843791	0.258916	0.072686
O	1.095476	1.117886	2.666750
H	1.312649	1.900316	2.115168
I	-3.401952	0.620074	0.341882
O	-2.335655	0.232535	-1.137462
O	-5.131298	0.380619	-0.578905
H	-5.300014	1.133343	-1.151360
I	-1.168116	-1.759971	-1.326957
O	-1.069978	-1.635326	0.555583
O	-0.063041	-3.365138	-1.418258
H	0.866112	-3.089567	-1.318258
C	4.335450	-3.200392	0.618297
H	5.030533	-3.207465	-0.215828
H	3.842428	-4.163437	0.721809
H	4.833440	-2.924774	1.543321
S	3.077614	-2.020085	0.288754
O	2.191544	-1.993737	1.421264
O	2.487423	-2.307678	-0.990669
O	3.922077	-0.745176	0.218596
H	3.516296	0.055401	-0.273325

$(HIO_3)_3(HIO_2)_1(MSA)_1$:

Atoms	X	Y	Z
I	-3.106823	-0.370436	-0.186573
O	-2.145238	-0.846587	-1.639348
O	-2.367302	1.168568	0.358986
O	-4.533365	0.501505	-1.186345
H	-4.209031	1.297354	-1.621863
I	0.061077	-2.029060	0.951439
O	-1.738609	-1.620778	1.064687
O	0.112706	-3.645227	1.641637
O	-0.024593	-2.465220	-0.911670
H	-0.820554	-2.007643	-1.267731
I	0.035066	2.050355	1.086344
O	1.860668	2.529156	1.051454
O	0.028319	0.731457	2.264618
O	-0.336960	3.459422	2.364098
H	0.015508	3.230682	3.232029
I	1.107430	0.609985	-2.134574
O	0.571602	0.435722	-0.333269
O	-0.645515	1.197801	-2.746282
H	-1.277691	0.480487	-2.534567
C	5.189397	-1.069024	-0.041490

H	5.790078	-0.188883	-0.251452
H	5.293215	-1.811758	-0.827119
H	5.449669	-1.494011	0.924183
S	3.499037	-0.583334	0.037939
O	2.724920	-1.783507	0.274350
O	3.229755	0.011272	-1.299116
O	3.397067	0.414591	1.089765
H	2.442469	1.711174	1.058207

Supplementary Reference

Elm, J. and Kristensen, K.: Basis set convergence of the binding energies of strongly hydrogen-bonded atmospheric clusters, Phys. Chem. Chem. Phys., 19, 1122–1133, http://doi.org/10.1039/c6cp06851k, 2017.

Francl, M. M., Pietro, W. J., Hehre, W. J., Binkley, J. S., Gordon, M. S., DeFrees, D. J. and Pople, J. A.: Self-consistent molecular orbital methods. XXIII. A polarization-type basis set for second-row elements, J. Chem. Phys. , 77, 3654–3665, http://doi.org/10.1063/1.444267, 1982.

Frisch, M. J., Trucks, G. W., Schlegel, H. B., Scuseria, G. E., Robb, M. A., Cheeseman, J. R., Scalmani, G., Barone, V., Mennucci, B., Petersson, G. A., Nakatsuji, H., Caricato, M., Li, X., Hratchian, H. P., Izmaylov, A. F., Bloino, J., Zheng, G., Sonnenberg, J. L., Hada, M., Ehara, M., Toyota, K., Fukuda, R., Hasegawa, J., Ishida, M., Nakajima, T., Honda, Y., Kitao, O., Nakai, H., Vreven, T., Montgomery, J. A., Peralta, J. E., Ogliaro, F., Bearpark, M., Heyd, J. J., Brothers, E., Kudin, K. N., Staroverov, V. N., Kobayashi, R., Normand, J., Raghavachari, K., Rendell, A., Burant, J. C., Iyengar, S. S., Tomasi, J., Cossi, M., Rega, N., Millam, J. M., Klene, M., Knox, J. E., Cross, J. B., Bakken, V., Adamo, C., Jaramillo, J., Gomperts, R., Stratmann, R. E., Yazyev, O., Austin, A. J., Cammi, R., Pomelli, C., Ochterski, J. W., Martin, R. L., Morokuma, K., Zakrzewski, V. G., Voth, G. A., Salvador, P., Dannenberg, J. J., Dapprich, S., Daniels, A. D., Farkas, O., Foresman, J. B., Ortiz, J. V., Cioslowski, J. and Fox, D. J.: Gaussian 09, Revision A.02, Gaussian Inc, Wallingford CT, https://gaussian.com/g09citation/ (last access: 07 May 2022), 2009.

Peterson, K. A., Figgen, D., Goll, E., Stoll, H. and Dolg, M.: Systematically convergent basis sets with relativistic pseudopotentials. II. Small-core pseudopotentials and correlation consistent basis sets for the post-d group 16–18 elements, J. Chem. Phys. , 119, 11113–11123, http://doi.org/10.1063/1.1622924, 2003.

Rappé, A. K., Casewit, C. J., Colwell, K. S., Goddard, W. A. and Skif, W. M.: UFF, a Full Periodic Table Force Field for Molecular Mechanics and Molecular Dynamics Simulations, J. Am. Chem. Soc., 114, 10024–10035, http://doi.org/10.1021/ja00051a040, 1992.

Stewart, J. J.: Optimization of parameters for semiempirical methods VI: more modifications to the NDDO approximations and re-optimization of parameters, J. Mol. Model., 19, 1–32, http://doi.org/10.1007/s00894-012-1667-x, 2013.

Stewart, J. J. P.: MOPAC 2016, Colorado Springs, CO (USA), http://openmopac.net/MOPAC2016.html, 2016.

Zhang, J. and Dolg, M.: ABCluster: the artificial bee colony algorithm for cluster global optimization, Phys. Chem. Chem. Phys., 17, 24173–24181, http://doi.org/10.1039/c5cp04060d, 2015.