Supporting information

-for-

A Possible Atmospheric Source of NH_2SO_3H: The Hydrolysis of $HNSO_2$ in the Presence of Neutral, Basic and Acidic Catalysts

Tianlei Zhang [a,*], Yongqi Zhang [a,#], Shiyu Tian [a,#], Mi Zhou [a,#], Dong Liu [a,#], Ling Lin [a,#], Qiang Zhang [a], Rui Wang [a,*], Balaganesh Muthiah [b,*]

[a] *Institute of Theoretical and Computational Chemistry, Shaanxi Key Laboratory of Catalysis, School of Chemical & Environment Science, Shaanxi University of Technology, Hanzhong, Shaanxi 723001, P. R. China*
[b] *Department of Chemistry, National Taiwan University, Taipei 106, Taiwan*

S. NO	Caption
S1	**Fig. S1** The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots(H_2O)_2$, $HNSO_2\cdots(H_2O)_3$ and $HNSO_2\cdots(H_2O)_4$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory
S2	**Fig. S2** The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots NH_3$ and $HNSO_2\cdots H_2O\cdots CH_3NH_2$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory
S3	**Fig. S3** The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots$HCOOH at the M06-2X/6-311+G(2*df*,2*pd*) level of theory
S4	**Fig. S4** The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots H_2SO_4$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory
S5	**Fig. S5** The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots H_2SO_4\cdots H_2O$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory
S6	**Fig. S6** The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots(H_2SO_4)_2$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory
S7	**Fig. S7** Hindrance potentials for $HNSO_2$, IM ($HNSO_2\cdots H_2O$), TS and IMF (NH_2SO_3H) calculated at the M06-2X/6-311+G(2*df*,2*pd*) level of theory
S8-S10	**Table S1** Zero point energy (ZPE/(kcal·mol^{-1})), entropies (S/(cal·mol^{-1}·K^{-1})), relative energies (ΔE and $\Delta(E$ + ZPE)/(kcal·mol^{-1})), enthalpies ($\Delta H(298)$/(kcal·mol^{-1})), and free energies ($\Delta G(298)$/(kcal·mol^{-1})) for the hydrolysis reaction of $HNSO_2$ without and with catalyst *X* (*X* = H_2O, $(H_2O)_2$, $(H_2O)_3$, NH_3, CH_3NH_2, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$)
S11	**Table S2** Equilibrium constants (cm^3·molecule^{-1}) for $HNSO_2\cdots H_2O$, $(H_2O)_2$, $HNSO_2\cdots(H_2O)_2$, $(H_2O)_3$, $(H_2O)_4$, $NH_3\cdots H_2O$, $CH_3NH_2\cdots H_2O$, $HCOOH\cdots H_2O$, *t*-

* Corresponding authors. Tel: +86-0916-2641083, Fax: +86-0916-2641083.
e-mail: ztianlei88@l63.com (T. L. Zhang); wangrui830413@163.com (R. Wang); bala.msc09@gmail.com (M. Balaganesh)
# Yongqi Zhang, Shiyu Tian, Mi Zhou, Dong Liu and Ling Lin are contributed equally to this work.

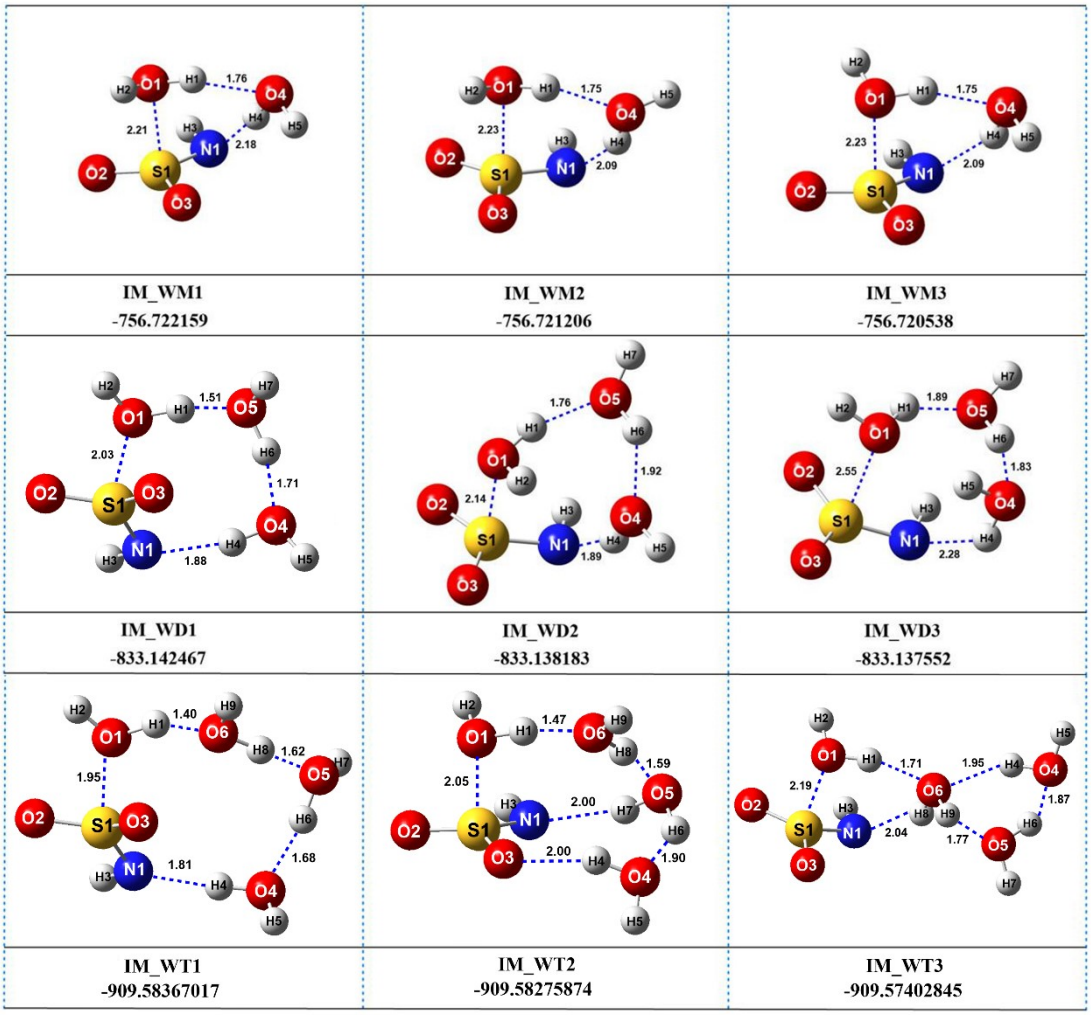


Fig. S1 The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots(H_2O)_2$, $HNSO_2\cdots(H_2O)_3$ and $HNSO_2\cdots(H_2O)_4$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory

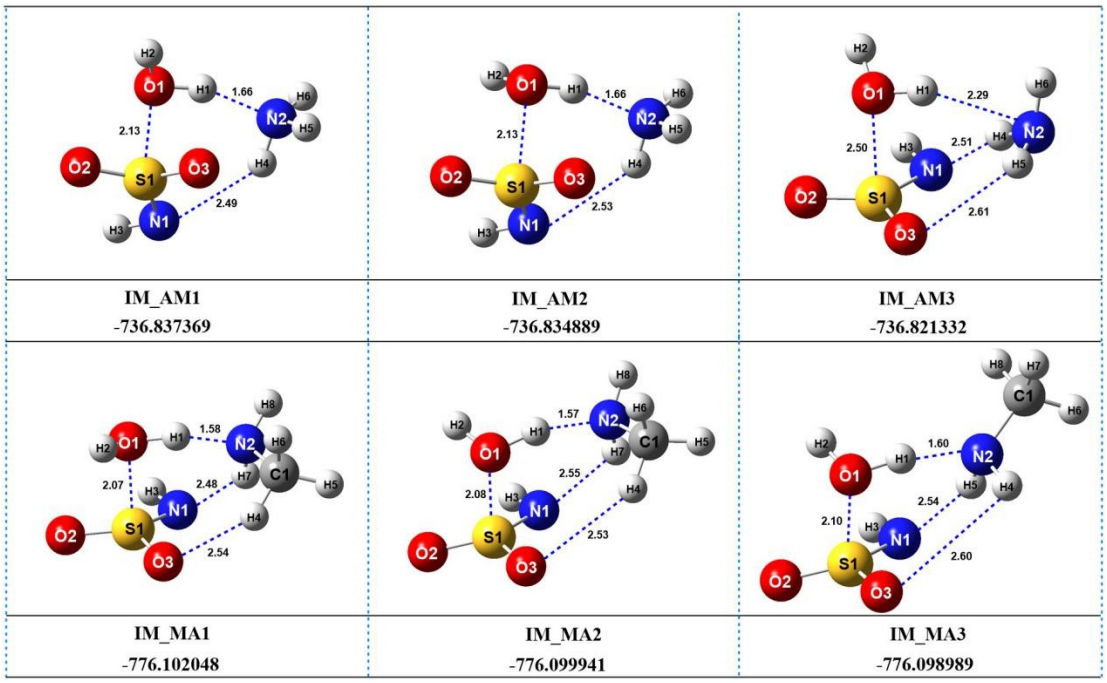


Fig. S2 The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots NH_3$ and $HNSO_2\cdots H_2O\cdots CH_3NH_2$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory

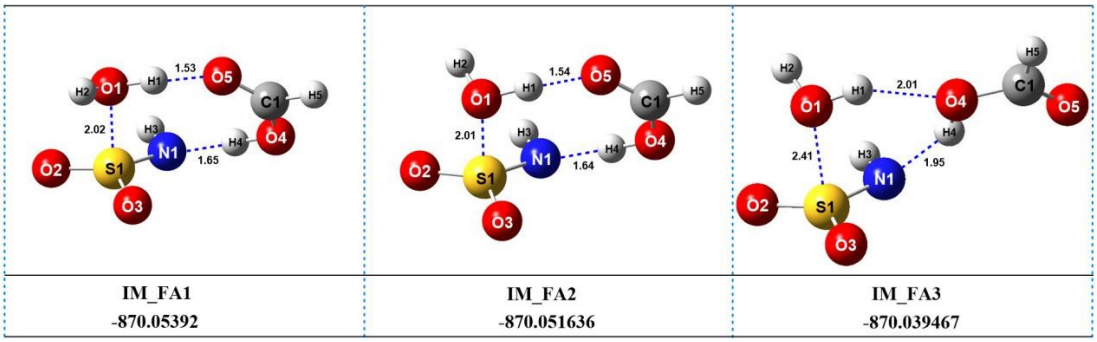


Fig. S3 The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots HCOOH$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory

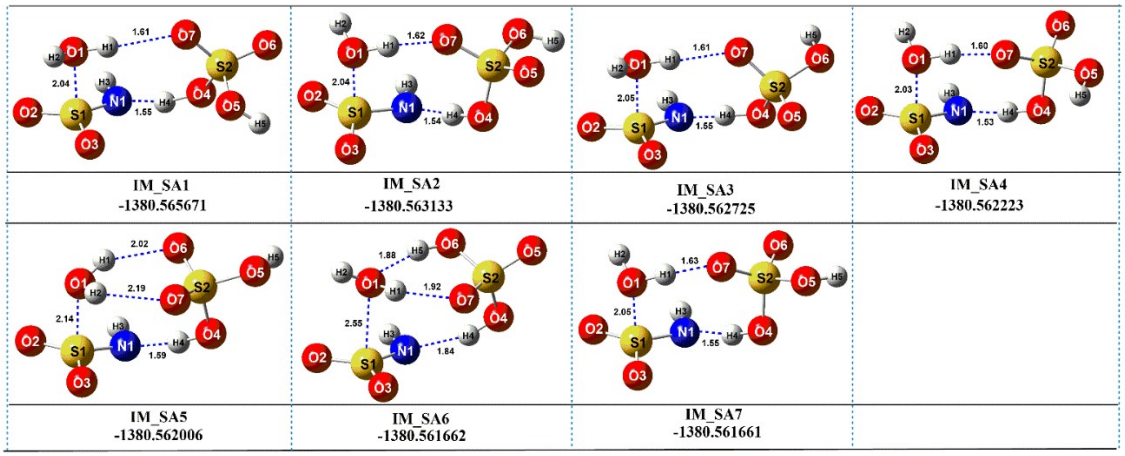


Fig. S4 The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots H_2SO_4$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory

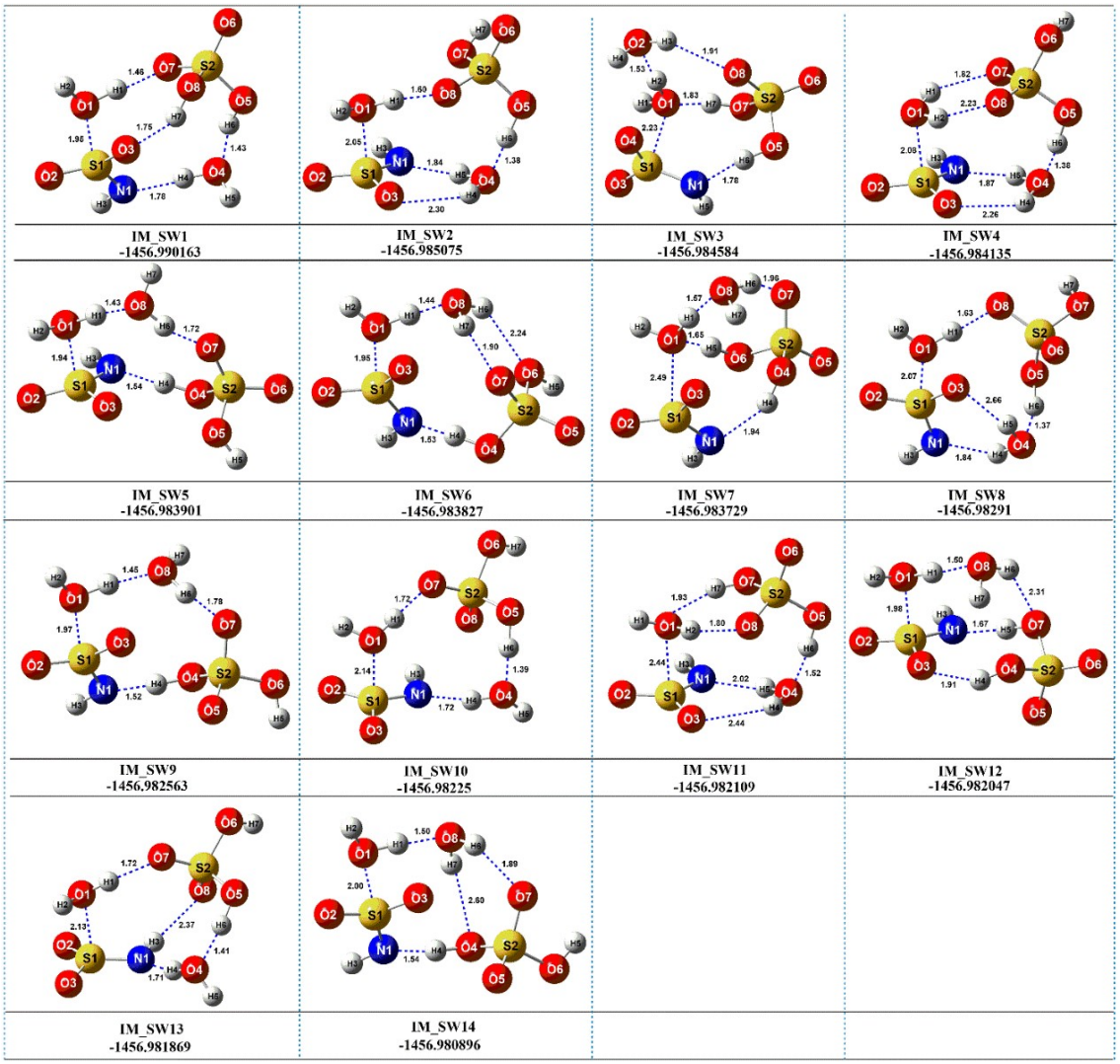


Fig. S5 The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots H_2SO_4\cdots H_2O$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory

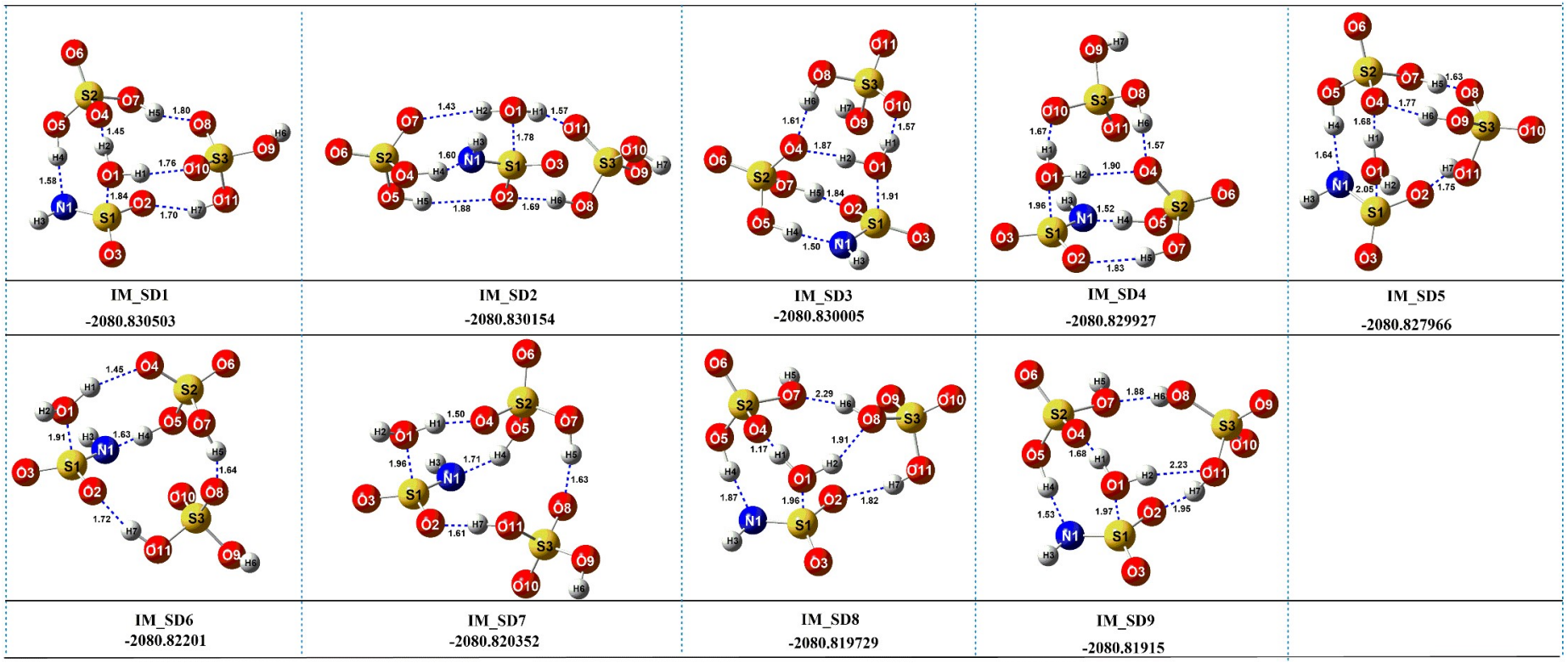


Fig. S6 The optimized geometries and electric energies (in Hartree-Fock) for the complexes of $HNSO_2\cdots H_2O\cdots(H_2SO_4)_2$ at the M06-2X/6-311+G(2*df*,2*pd*) level of theory

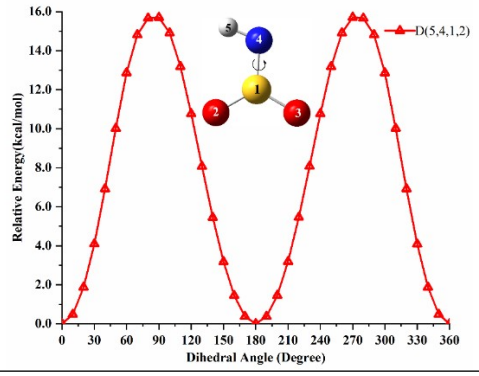
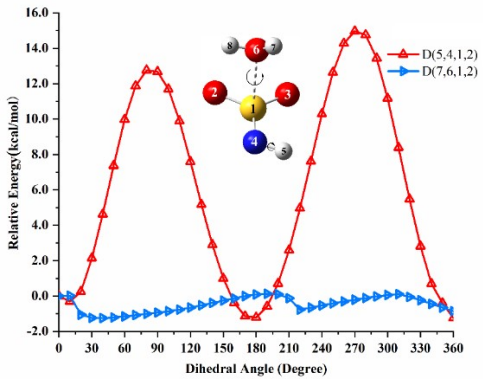
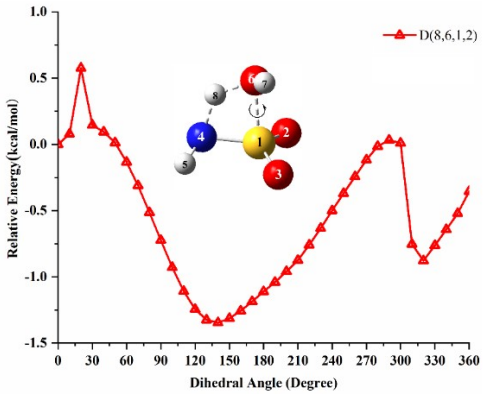
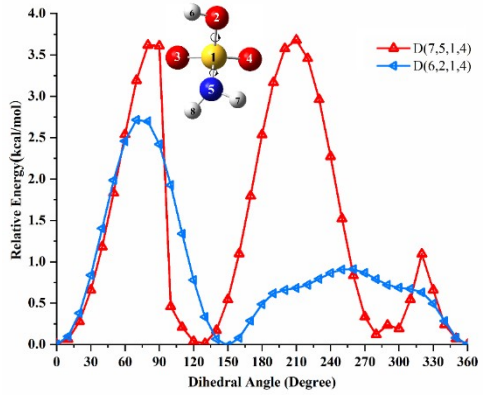
Species	Potential energy surfaces for the internal rotations
$HNSO_2$	
IM ($HNSO_2\cdots H_2O$)	
TS	
IMF (NH_2SO_3H)	

Fig. S7 Hindrance potentials for $HNSO_2$, IM ($HNSO_2\cdots H_2O$), TS and IMF (NH_2SO_3H) calculated at the M06-2X/6-311+G(2*df*,2*pd*) level of theory

Table S1 Zero point energy (ZPE/(kcal·mol^{-1})), entropies (S/(cal·mol^{-1}·K^{-1})), relative energies (ΔE and $\Delta(E + \text{ZPE})$/(kcal·mol^{-1})), enthalpies ($\Delta H(298)$/(kcal·mol^{-1})), and free energies ($\Delta G(298)$/(kcal·mol^{-1})) for the hydrolysis reaction of $HNSO_2$ without and with catalyst X (X = H_2O, $(H_2O)_2$, $(H_2O)_3$, NH_3, CH_3NH_2, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$)

Species	ZPE	ΔE	S	ΔG	$\Delta(E + \text{ZPE})$	ΔH
$HNSO_2 + H_2O$	28.7	0.0	110.6	0.0	0.0	0.0
IM	30.4	-6.0	83.2	3.8	-4.3	-4.5
TS	29.2	20.2	71.9	30.6	20.6	19.1
NH_2SO_3H	32.8	-35.9	73.2	-22.0	-31.8	-33.2
$HNSO_2 + 2H_2O$	42.3	0.0	155.7	0.0	0.0	0.0
$HNSO_2 + (H_2O)_2$	44.8	-5.0	133.2	3.5	-2.5	-3.2
$HNSO_2\cdots H_2O + H_2O$	44.0	-6.0	127.7	3.8	-4.3	-4.5
IM_WM1	47.4	-19.6 (-20.3)[a]	89.1	3.6	-14.5	-16.2
TS_WM1	45.8	-11.2 (-12.1)[a]	79.8	11.8	-7.7	-10.8
IMF_WM1	48.3	-44.5 (-42.9)[a]	87.1	-20.5	-38.6	-40.3
$NH_2SO_3H + H_2O$	46.4	-35.9	118.3	-22.0	-31.8	-33.2
$HNSO_2 + H_2O + (H_2O)_2$	58.4	0.0	178.3	0.0	0.0	0.0
$HNSO_2 + (H_2O)_3$	61.5	-10.7	144.9	1.2	-7.6	-8.7
$HNSO_2\cdots H_2O + (H_2O)_2$	60.1	-6.0	150.3	3.8	-4.3	-4.5
IM_WD′	62.8	-20.0	140.9	4.8	-15.6	-16.7
TS_WD′	62.1	-17.1	105.9	10.4	-13.3	-14.2
IM_WD1	63.5	-26.1 (-26.3)[a]	89.1	0.2	-20.9	-23.0
TS_WD1	62.1	-22.6 (-23.5)[a]	91.9	3.7	-18.9	-22.0
IMF_WD1	64.1	-51.6 (-52.5)[a]	103.5	-25.3	-45.9	-47.6
$NH_2SO_3H + (H_2O)_2$	62.5	-35.9	140.9	-22.0	-31.8	-33.2
$HNSO_2 + 2(H_2O)_2$	74.5	0.0	200.9	0.0	0.0	0.0
$HNSO_2 + (H_2O)_4$	77.4	-16.4	158.9	-2.3	-13.4	-16.4
$HNSO_2\cdots(H_2O)_2 + (H_2O)_2$	77.9	-14.6	156.7	0.1	-12.0	-13.0
IM_WT′	78.5	-22.4	126.9	2.5	-18.4	-22.4
TS_WT′	79.6	-22.0	121.4	3.8	-16.9	-22.0
IM_WT1	79.1	-27.6 (-28.7)[a]	114.4	0.5	-23.0	-27.6
TS_WT1	78.3	-26.3 (-28.7)[a]	107.5	2.3	-22.5	-26.3
IMF_WT1	79.7	-53.3 (-54.6)[a]	119.1	-25.2	-48.0	-53.3

$NH_2SO_3H + (H_2O)_3$	76.1	-30.9	208.4	-25.5	-29.3	-30.9
$HNSO_2 + H_2O + NH_3$	50.4	0.0	156.5	0.0	0.0	0.0
$NH_3\cdots H_2O + HNSO_2$	52.9	-6.4	133.8	2.2	-3.9 (-4.2)[b]	-4.6
$HNSO_2\cdots H_2O + NH_3$	52.1	-6.0	128.6	3.8	-4.3	-4.5
IM_AM1	55.0	-19.6	90.1	3.2	-15.0	-16.6
TS_AM1	54.4	-15.7	82.5	7.8	-11.7	-14.3
IMF_AM1	56.0	-45.3	92.4	-22.1	-39.8	-41.3
$NH_2SO_3H + NH_3$	54.5	-35.9	119.2	-22.0	-31.8	-33.2
$HNSO_2 + H_2O + CH_3NH_2$	69.1	0.0	168.1	0.0	0.0	0.0
$HNSO_2\cdots H_2O + CH_3NH_2$	70.8	-6.0	140.2	3.8	-4.3	-4.5
$CH_3NH_2\cdots H_2O + HNSO_2$	71.3	-7.2 (-7.73)[c]	143.1	2.1	-4.9	-5.4
IM_MA1	73.7	-20.4	97.1	4.1	-15.8	-17.1
TS_MA1	72.7	-19.3	91.1	5.2	-15.7	-17.7
IMF_MA1	74.3	-46.4	98.9	-21.7	-41.1	-42.4
$NH_2SO_3H + CH_3NH_2$	73.2	-35.9	130.7	-22.0	-31.8	-33.2
$HNSO_2 + H_2O + HCOOH$	50.3	0.0	169.9	0.0	0.0	0.0
$HNSO_2\cdots H_2O + HCOOH$	52.0	-6.0	141.9	3.8	-4.3	-4.5
$HCOOH\cdots H_2O + HNSO_2$	52.7	-10.2	139.2	0.7	-7.7 (-8.06)[d]	-8.5
IM_FA′	54.0	-16.2	101.9	7.2	-12.5	-13.1
TS_FA′	52.5	-13.7	100.9	8.8	-11.4	-11.8
IM_FA1	54.8	-22.0	95.2	3.3	-17.5	-19.0
TS_FA1	52.4	-19.4	89.9	4.4	-17.2	-19.4
IMF_FA1	55.4	-49.4	99.3	-24.5	-44.3	-45.5
$NH_2SO_3H + HCOOH$	54.4	-35.9	132.5	-22.0	-31.8	-33.2
$HNSO_2 + H_2O + t\text{-}H_2SO_4$	54.0	0.0	182.2	0.0	0.0	0.0
$HNSO_2\cdots H_2O + t\text{-}H_2SO_4$	55.7	-6.0	154.8	3.8	-4.3	-4.5
$t\text{-}H_2SO_4\cdots H_2O + HNSO_2$	55.9	-12.3	152.6	-2.1	-10.4 (-10.1)[e]	-11.0
IM_SA′	56.8	-18.6	144.8	1.8	-15.8	-15.9
TS_SA′	57.9	-18.3	122.7	4.5	-14.4	-16.6
IM_SA1	57.5	-23.8	108.8	0.4	-20.4	-21.5
TS_SA1	55.7	-22.2	103.8	1.2	-20.5	-22.2
IMF_SA1	58.7	-49.5	109.9	-24.3	-44.8	-45.9
$NH_2SO_3H + t\text{-}H_2SO_4$	58.1	-35.9	144.8	-22.0	-31.8	-33.2
$HNSO_2 + H_2O + c\text{-}H_2SO_4\cdots H_2O$	70.2	0.0	194.2	0.0	0.0	0.0
$c\text{-}H_2SO_4\cdots H_2O + HNSO_2\cdots H_2O$	71.9	-6.0	166.2	3.8	-4.3	-4.5
$c\text{-}H_2SO_4\cdots (H_2O)_2 + HNSO_2$	72.6	-12.5	163.0	-1.6	-10.1	-10.9
IM_SW′	71.9	-17.8	156.8	2.8	-16.1	-15.1

TS_SW′	73.3	-17.8	134.4	4.2	-14.7	-15.7
IM_SW1	73.5	-25.8	120.5	-0.9	-22.5	-24.0
TS_SW1	71.4	-23.1	111.7	0.9	-21.9	-24.3
IMF_SW1	74.7	-50.3	122.7	-26.0	-45.9	-46.8
NH_2SO_3H + *c*-H_2SO_4···H_2O	74.3	-35.9	156.8	-22.0	-31.8	-33.2
$HNSO_2$ + H_2O + $(H_2SO_4)_2$	80.4	0.0	213.9	0.0	0.0	0.0
$HNSO_2$···H_2O + $(H_2SO_4)_2$	82.0	-6.0	185.9	3.8	-4.3	-4.5
$(H_2SO_4)_2$···H_2O + $HNSO_2$	81.8	-12.6	183.0	-2.7	-11.2 (-11.6)[e]	-11.9
IM_SD′	82.3	-19.1	146.3	2.4	-17.2	-17.8
TS_SD′	81.6	-17.1	143.7	4.6	-15.9	-16.3
IM_SD1	83.5	-24.1	136.2	0.9	-21.0	-22.3
TS_SD1	81.4	-22.4	132.2	1.1	-21.3	-23.2
IMF_SD1	84.7	-49.6	134.9	-23.0	-45.2	-46.5
NH_2SO_3H + $(H_2SO_4)_2$	84.5	-35.9	176.5	-22.0	-31.8	-33.2

[a] The value was taken from reference (*Int. J. Quantum Chem.*, 2020, **120**, e26182)

[b] The value was taken from reference (*Int. J. Quantum Chem.*, 2021, **121**, e26432)

[c] The value was taken from reference (*Entropy*, 2015, **17**, 2764-2780)

[d] The value was taken from reference (*ChemPhysChem*, 2012, **13**, 323-329)

[e] The value was taken from reference (*Atmos. Environ.*, 2020, **243**, 117876)

Table S2 Equilibrium constants ($cm^3 \cdot molecule^{-1}$) for $HNSO_2 \cdots H_2O$, $(H_2O)_2$, $HNSO_2 \cdots (H_2O)_2$, $(H_2O)_3$, $(H_2O)_4$, $NH_3 \cdots H_2O$, $CH_3NH_2 \cdots H_2O$, HCOOH$\cdots H_2O$, *t*-$H_2SO_4 \cdots H_2O$, *c*-$H_2SO_4 \cdots H_2O$, *c*-$H_2SO_4 \cdots (H_2O)_2$, $(H_2SO_4)_2$ and $(H_2SO_4)_2 \cdots H_2O$ within the temperature range of 280-320 K

T/K	$HNSO_2 \cdots H_2O$	$(H_2O)_2$	$HNSO_2 \cdots (H_2O)_2$	$(H_2O)_3$	$(H_2O)_4$
280	1.11E-22	3.56E-21	1.29E-21	1.98E-20	6.68E-18
290	8.75E-23	3.02E-21	7.60E-22	1.21E-20	3.38E-18
298	7.32E-23	2.68E-21 (2.34E-21)[a]	5.11E-22	8.40E-21 (4.55E-20)[a]	2.03E-18 (6.23E-18)[a]
300	7.01E-23	2.60E-21	4.64E-22	7.69E-21	1.79E-18
310	5.71E-23	2.26E-21	2.93E-22	5.03E-21	9.89E-19
320	4.73E-23	1.99E-21	1.91E-22	3.38E-21	5.68E-19

T/K	$NH_3 \cdots H_2O$	$CH_3NH_2 \cdots H_2O$	HCOOH$\cdots H_2O$	*t*-$H_2SO_4 \cdots H_2O$	*c*-$H_2SO_4 \cdots H_2O$
280	5.23E-22	1.92E-22	4.98E-20	2.65E-18	1.55E-19
290	4.08E-22	1.43E-22	3.02E-20	1.36E-18	6.59E-20
298	3.79E-22 (3.91E-22)[b]	1.14E-22	2.07E-20 (1.10E-20)c	8.29E-19 (3.88E-19)[d]	4.10E-20 (5.16E-20)[d]
300	3.23E-22	1.08E-22	1.13E-20	7.35E-19	3.69E-20
310	2.61E-22	8.38E-23	1.23E-20	4.13E-19	2.21E-20
320	2.14E-22	6.60E-23	8.18E-21	2.41E-19	1.37E-20

T/K	*c*-$H_2SO_4 \cdots (H_2O)_2$	$(H_2SO_4)_2$	$(H_2SO_4)_2 \cdots H_2O$
280	8.91E-16	6.54E-15	1.47E-17
290	3.30E-16	4.68E-15	7.22E-18
298	1.57E-16 (1.54E-15)[d]	2.01E-15 (1.02E-16)[d]	4.24E-18 (4.06E-17)[d]
300	1.34E-16	1.63E-15	3.72E-18
310	5.51E-17	6.11E-16	2.01E-18
320	2.50E-17	2.43E-16	1.13E-18

The equilibrium constants ($cm^3 \cdot molecule^{-1}$) for the complexes involved in Table S2 were performed by ILT method in Mesmer code;

[a] The value was taken from reference (*J. Am. Chem. Soc.*, 2012, **134**, 20632-20644)

[b] The value was taken from reference (*Comput. Theor. Chem.*, 2020, **1176**, 112747)

[c] The value was taken from reference (*ChemPhysChem*., 2012, **13**, 323-329)

[d] The value was taken from reference (*J. Phys. Chem. A*, 2021, **125**, 2642-2652)

Table S3 Concentrations (molecules·cm^{-3}) of $(H_2O)_2$, $(H_2O)_3$, $NH_3\cdots H_2O$, $CH_3NH_2\cdots H_2O$, $HCOOH\cdots H_2O$, *t*-$H_2SO_4\cdots H_2O$, *c*-$H_2SO_4\cdots H_2O$, *c*-$H_2SO_4\cdots(H_2O)_2$ and $(H_2SO_4)_2$ within the temperature range of 280-320 K[a]

Catalysts	RH		280 K	290 K	298 K	300 K	310 K	320 K
$(H_2O)_2$	20%RH		9.48E+12	2.78E+13	6.03E+13 (5.44E+13)[b]	7.69E+13	1.93E+14	4.40E+14
	40%RH		3.78E+13	1.10E+14	2.58E+14 (2.18E+14)[b]	3.06E+14	7.71E+14	1.76E+15
	60%RH		8.55E+13	2.49E+14	5.43E+14 (4.90E+14)[b]	6.90E+14	1.74E+15	3.96E+15
	80%RH		1.53E+14	4.41E+14	1.03E+15 (8.70E+14)[b]	1.22E+15	3.09E+15	7.03E+15
	100%RH		2.37E+14	6.90E+14	1.59E+15 (1.36E+15)[b] (1.72E+15)[g]	1.91E+15	4.82E+15	1.10E+16
$(H_2O)_3$	20%RH		9.68E+09	3.23E+10	7.60E+10 (3.83E+11)[b]	1.02E+11	2.83E+11	6.98E+11
	40%RH		7.70E+10	2.55E+11	6.71E+11 (3.06E+12)[b]	8.07E+11	2.26E+12	5.59E+12
	60%RH		2.62E+11	8.64E+11	2.05E+12 (1.03E+13)[b]	2.73E+12	7.67E+12	1.89E+13
	80%RH		6.25E+11	2.04E+12	5.37E+12 (2.45E+13)[b]	6.45E+12	1.82E+13	4.47E+13
	100%RH		1.21E+12	3.99E+12	1.03E+13 (4.78E+13)[b]	1.26E+13	3.54E+13	8.73E+13
$NH_3\cdots H_2O$	20%RH	0.1 ppbv	1.42E+05	1.98E+05	2.80E+05	2.71E+05	3.60E+05	4.61E+05
		10 ppbv	1.42E+07	1.98E+07	2.80E+07	2.71E+07	3.60E+07	4.61E+07
		2900ppbv	4.12E+09	5.72E+09	8.07E+09	7.89E+09	1.05E+10	1.35E+10
	100%RH	0.1 ppbv	7.10E+05	9.87E+05	1.44E+06	1.35E+06	1.80E+06	2.30E+06
		10 ppbv	7.10E+07	9.87E+07	1.44E+08 (3.59E+08)[c]	1.35E+08	1.80E+08	2.30E+08
		2900pbv	2.06E+10	2.85E+10	4.14E+10	3.94E+10	5.26E+10	6.74E+10
$CH_3NH_2\cdots H_2O$	20%RH	80 ppbv	7.14E+05	9.90E+05	1.23E+06	1.34E+06	1.76E+06	2.24E+06
		180 ppbv	1.60E+06	2.22E+06	2.77E+06	3.01E+06	3.96E+06	5.03E+06
	100%RH	80 ppbv	3.57E+06	4.93E+06	6.33E+06	6.68E+06	8.82E+06	1.12E+07
		180 ppbv	8.02E+06	1.11E+07	1.42E+07	1.50E+07	1.98E+07	2.51E+07
$HCOOH\cdots H_2O$	20%RH	0.01ppbv	6.74E+05	7.33E+05	7.65E+05	7.95E+05	8.46E+05	8.81E+05
		10 ppbv	6.74E+08	7.33E+08	7.65E+08	7.95E+08	8.46E+08	8.81E+08
	100%RH	0.01ppbv	3.37E+06	3.65E+06	3.93E+06	3.97E+06	4.23E+06	4.40E+06
		10 ppbv	3.37E+09	3.65E+09	3.93E+09 (3.16E+09)[c]	3.97E+09	4.23E+09	4.40E+09
t-$H_2SO_4\cdots H_2O$	20%RH	[SA][f]=10^8	5.33E+07	4.96E+07	4.60E+07	4.68E+07	4.34E+07	3.85E+07
		[SA][f]=10^6	5.33E+05	4.96E+05	4.60E+05	4.55E+05	4.22E+05	3.85E+05

		[SA][f]=10^4	5.33E+03	4.96E+03	4.60E+03	4.55E+03	4.22E+03	3.85E+03
	100%RH	[SA][f]=10^8	2.67E+08	2.47E+08	2.36E+08 (2.40E+07)[d] (1.11E+08)[e]	2.27E+08	2.11E+08	1.93E+08
		[SA][f]=10^6	2.67E+06	2.47E+06	2.36E+06	2.27E+06	2.11E+06	1.93E+06
		[SA][f]=10^4	2.67E+04	2.47E+04	2.36E+04	2.27E+04	2.11E+04	1.93E+04
c-$H_2SO_4\cdots H_2O$	20%RH	[SA][f]=10^8	3.12E+06	2.40E+06	2.28E+06	2.28E+06	2.26E+06	2.19E+06
		[SA][f]=10^6	3.12E+04	2.40E+04	2.28E+04	2.28E+04	2.26E+04	2.19E+04
		[SA][f]=10^4	3.12E+02	2.40E+02	2.28E+02	2.28E+02	2.26E+02	2.19E+02
	100%RH	[SA][f]=10^8	1.56E+07	1.20E+07	1.17E+07 (1.47E+07)[e]	1.14E+07	1.13E+07	1.09E+07
		[SA][f]=10^6	1.56E+05	1.20E+05	1.17E+05	1.14E+05	1.13E+05	1.09E+05
		[SA][f]=10^4	1.56E+03	1.20E+03	1.17E+03	1.14E+03	1.13E+03	1.09E+03
c-$H_2SO_4\cdots(H_2O)_2$	20%RH	[SA][f]=10^8	2.53E+06	2.75E+06	2.83E+06	3.09E+06	3.18E+06	3.29E+06
		[SA][f]=10^6	2.53E+04	2.75E+04	2.83E+04	3.09E+04	3.18E+04	3.29E+04
		[SA][f]=10^4	2.53E+02	2.75E+02	2.83E+02	3.09E+02	3.18E+02	3.29E+02
	100%RH	[SA][f]=10^8	6.33E+07	6.83E+07	7.46E+07 (6.27E+07)[e]	7.68E+07	7.96E+07	8.23E+07
		[SA][f]=10^6	6.33E+05	6.83E+05	7.46E+05	7.68E+05	7.96E+05	8.23E+05
		[SA][f]=10^4	6.33E+03	6.83E+03	7.46E+03	7.68E+03	7.96E+03	8.23E+03
$(H_2SO_4)_2$	[SA][f]=10^8		9.95E+02	6.76E+02	2.75E+02 (1.40E+01)[e]	2.11E+02	7.48E+01	2.81E+01
	[SA][f]=10^6		9.95E-02	6.76E-02	2.75E-02	2.11E-02	7.48E-03	2.81E-03
	[SA][f]=10^4		9.95E-06	6.76E-06	2.75E-06	2.11E-06	7.48E-07	2.81E-07

[a] All of the energies in the calculated equilibrium constant in Table S3 were computed at the CCSD(T)-F12/cc-pVDZ-F12 level, while the partition functions were obtained at the M06-2X/6-311+G(2*df*,2*pd*) level.

[b] The values was reported from reference (*J. Phys. Chem. A*, 2013, **117**, 10381-10396)

[c] The values was taken from reference (*J. Phys. Chem. A*, 2010, **114**, 5796-580)

[d] The values was taken from reference (*Environ. Sci. Technol.*, 2015, **49**, 13112-13120)

[e] The values was taken from reference (*J. Phys. Chem. A*, 2021, **125**, 2642-2652)

[f] The values was taken from reference (*Proc. Natl. Acad. Sci. U. S. A*., 2019, **116**, 24966-24971)

Table S4 Rate coefficients ($cm^3 \cdot molecules^{-1} \cdot s^{-1}$) for the $HNSO_2 + H_2O$ reaction without and with X calculated by canonical variational transition (CVT) state theory with small curvature tunneling (SCT) correction within the temperature range of 280-320 K

Reaction	T (K)	280	290	298	300	310	320
Channel R1	K_{eq}	6.94E-23	5.41E-23	4.50E-23	4.31E-23	3.49E-23	2.87E-23
	k_{uni}	5.39E-06	1.19E-05	2.25E-05	2.64E-05	5.89E-05	1.31E-04
	k_b	3.74E-28	6.43E-28	1.01E-27	1.14E-27	2.05E-27	3.75E-27
Channel WM	K_{eq}	7.29E-22	4.33E-22	2.92E-22	2.66E-22	1.69E-22	1.10E-22
	k_{uni}	5.64E+06	7.77E+06	9.89E+06	1.05E+07	1.39E+07	1.81E+07
	k_b	4.11E-15	3.36E-15	2.89E-15	2.79E-15	2.35E-15	2.00E-15
Channel WD (one-step route)	K_{eq}	6.70E-19	2.66E-19	1.33E-19	1.12E-19	5.02E-20	2.35E-20
	k_{uni}	3.53E+10	3.83E+10	4.07E+10	4.13E+10	4.43E+10	4.72E+10
	k_{WD_o}	2.36E-08	1.02E-08	5.41E-09	4.64E-09	2.22E-09	1.11E-09
Channel WT (one-step route)	K_{eq}	9.74E-18	4.01E-18	2.06E-18	1.75E-18	8.05E-19	3.89E-19
	k_{uni}	3.65E+11	3.69E+11	3.72E+11	3.73E+11	3.77E+11	3.80E+11
	k_{WD_o}	3.55E-06	1.48E-06	7.66E-07	6.53E-07	3.03E-07	1.48E-07
Channel AM	K_{eq}	2.66E-21	1.41E-21	8.80E-22	7.85E-22	4.53E-22	2.71E-22
	k_{uni}	2.96E+10	3.21E+10	3.41E+10	3.46E+10	3.72E+10	3.97E+10
	k_b	7.88E-11	4.54E-11	3.00E-11	2.72E-11	1.68E-11	1.08E-11
Channel MA	K_{eq}	4.07E-21	2.05E-21	1.23E-21	1.08E-21	5.98E-22	3.42E-22
	k_{uni}	9.87E+11	9.80E+11	9.76E+11	9.74E+11	9.68E+11	9.63E+11
	k_b	4.02E-09	2.01E-09	1.20E-09	1.06E-09	5.79E-10	3.29E-10
Channel FA (one-step route)	K_{eq}	4.60E-19	1.95E-19	1.02E-19	8.72E-20	4.11E-20	2.04E-20
	k_{uni}	1.13E+12	1.13E+12	1.13E+12	1.13E+12	1.14E+12	1.14E+12
	k_{FA_o}	5.19E-07	2.20E-07	1.15E-07	9.89E-08	4.67E-08	2.31E-08
Channel SA (one-step route)	K_{eq}	8.16E-17	2.95E-17	1.38E-17	1.15E-17	4.72E-18	2.06E-18
	k_{uni}	7.36E+11	7.79E+11	8.13E+11	8.21E+11	8.62E+11	9.02E+11
	k_{SA_o}	6.01E-05	2.30E-05	1.12E-05	9.40E-06	4.07E-06	1.86E-06
Channel SW (one-step route)	K_{eq}	7.04E-18	2.58E-18	1.21E-18	1.01E-18	4.21E-19	1.85E-19
	k_{uni}	1.45E+12	1.47E+12	1.48E+12	1.48E+12	1.50E+12	1.51E+12
	k_{SW_o}	1.02E-05	3.79E-06	1.80E-06	1.50E-06	6.30E-07	2.79E-07
Channel SD (one-step route)	K_{eq}	3.16E-18	1.30E-18	6.69E-19	5.69E-19	2.63E-19	1.28E-19
	k_{uni}	6.54E+12	6.61E+12	6.66E+12	6.67E+12	6.73E+12	6.78E+12
	k_{SD_o}	2.07E-05	8.60E-06	4.45E-06	3.80E-06	1.77E-06	8.65E-07

k_{WD_o}, k_{WT_o}, k_{FA_o}, k_{SA_o}, k_{SW_o} and k_{SD_o} are respectively the bimolecular rate coefficients for the one-step routes of the hydrolysis reaction of $HNSO_2$ with $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4 \cdots H_2O$ and $(H_2SO_4)_2$.

Table S5 Rate coefficients ($cm^3 \cdot molecules^{-1} \cdot s^{-1}$) for the hydrolysis reaction of $HNSO_2$ without and with *X* and the available calculated rate ratio (v_s/v_o) between stepwise route and the corresponding one-step process calculated by master equation within the temperature range of 280-320 K

Reaction		*T* (K)	280	290	298	300	310	320
Channel R1		$K_{eq(IM1)}$	1.11E-22	8.75E-23	7.32E-23	7.01E-23	5.71E-23	4.73E-23
		$k_{uni(TS1)}$	3.98E-06	8.46E-06	1.56E-05	1.82E-05	3.96E-05	8.57E-05
		k_{R1}	4.42E-28	7.40E-28	1.14E-27	1.28E-27	2.26E-27	4.05E-27
Channel WM		$K_{eq(IM\_WM1)}$	1.29E-21	7.60E-22	5.11E-22	4.64E-22	2.93E-22	1.91E-22
		$k_{uni(TS\_WM1)}$	5.13E+06	7.00E+06	8.85E+06	9.37E+06	1.23E+07	1.60E+07
		k_{WM}	6.61E-15	5.32E-15	4.52E-15	4.35E-15	3.62E-15	3.06E-15
Channel WD	One-step	$K_{eq(IM\_WD1)}$	3.52E-20	1.98E-20	1.27E-20	1.10E-20	5.52E-21	2.89E-21
		$k_{uni\ (TS\_WD1)}$	2.03E+10	2.18E+10	2.30E+10	2.33E+10	2.48E+10	2.63E+10
		k_{WD_o}	7.14E-10	4.32E-10	2.92E-10	2.57E-10	1.37E-10	7.61E-11
	Stepwise	$K_{eq(IM\_WD')}$	1.30E-21	8.15E-22	5.77E-22	5.31E-22	3.56E-22	2.46E-22
		$k_{uni(TS\_WD')}$	2.11E+11	2.50E+11	2.84E+11	2.93E+11	3.39E+11	3.90E+11
		$k_{uni(TS\_WD1)}$	2.03E+10	2.18E+10	2.30E+10	2.33E+10	2.48E+10	2.63E+10
		$k_{uni(WD)}$	1.85E+10	2.01E+10	2.13E+10	2.16E+10	2.31E+10	2.46E+10
		k_{WD_s}	2.40E-11	1.64E-11	1.23E-11	1.15E-11	8.24E-12	6.06E-12
		v_{WD_s}/v_{WD_o}	5.98E+00	5.25E+00	4.83E+00	4.91E+00	5.30E+00	5.69E+00
Channel WT	One-step	$K_{eq(IM\_WT1)}$	1.15E-21	6.81E-22	4.60E-22	4.19E-22	2.66E-22	1.73E-22
		$k_{uni(TS\_WT1)}$	1.46E+11	1.52E+11	1.57E+11	1.58E+11	1.64E+11	1.69E+11
		k_{WT_o}	1.67E-10	1.04E-10	7.22E-11	6.62E-11	4.35E-11	2.93E-11
	Stepwise	$K_{eq(IM\_WT')}$	8.81E-23	6.75E-23	5.54E-23	5.28E-23	4.21E-23	3.41E-23
		$k_{uni(TS\_WT')}$	4.58E+10	4.90E+10	5.13E+10	5.31E+10	5.64E+10	6.06E+10
		$k_{uni(TS\_WT1)}$	1.46E+11	1.52E+11	1.57E+11	1.58E+11	1.64E+11	1.69E+11
		$k_{uni(WT)}$	3.49E+10	3.70E+10	3.86E+10	3.98E+10	4.19E+10	4.46E+10
		k_{WT_s}	3.07E-12	2.50E-12	2.14E-12	2.10E-12	1.76E-12	1.52E-12
		v_{WT_s}/v_{WT_o}	9.52E+01	1.07E+02	1.18E+02	1.22E+02	1.37E+02	1.54E+02
Channel AM		$K_{eq(IM\_AM1)}$	1.58E-20	8.35E-21	5.16E-21	4.60E-21	2.63E-21	1.56E-21
		$k_{uni(TS\_AM1)}$	2.67E+10	2.89E+10	3.06E+10	3.10E+10	3.32E+10	3.54E+10
		k_{AM}	4.22E-10	2.41E-10	1.58E-10	1.43E-10	8.74E-11	5.53E-11
Channel MA		$K_{eq(IM\_MA1)}$	3.50E-21	1.82E-21	1.11E-21	9.87E-22	5.58E-22	3.27E-22
		$k_{uni(TS\_MA1)}$	4.68E+11	4.76E+11	4.81E+11	4.82E+11	4.89E+11	4.95E+11
		k_{AM}	1.64E-09	8.66E-10	5.34E-10	4.76E-10	2.73E-10	1.62E-10
Channel FA	One-step	$K_{eq(IM\_FA1)}$	2.52E-21	1.26E-21	7.51E-22	6.62E-22	3.62E-22	2.06E-22
		$k_{uni(TS\_FA1)}$	1.90E+11	2.02E+11	2.10E+11	2.13E+11	2.24E+11	2.34E+11
		k_{FA_o}	4.80E-10	2.54E-10	1.58E-10	1.41E-10	8.10E-11	4.82E-11
	Stepwise	$K_{eq(IM\_FA')}$	1.44E-22	1.15E-22	9.79E-23	9.41E-23	7.79E-23	6.54E-23
		$k_{uni(TS\_FA')}$	1.01E+10	1.08E+10	1.13E+10	1.14E+10	1.21E+10	1.27E+10
		$k_{uni(TS\_FA1)}$	1.90E+11	2.02E+11	2.10E+11	2.13E+11	2.24E+11	2.34E+11
		$k_{uni(FA)}$	9.61E+09	1.02E+10	1.07E+10	1.08E+10	1.15E+10	1.21E+10
		k_{FA_s}	1.39E-12	1.18E-12	1.05E-12	1.02E-12	8.93E-13	7.90E-13

		v_{FA_s}/v_{FA_o}	1.43E+00	1.60E+00	1.88E+00	1.96E+00	2.37E+00	2.83E+00
Channel SA	One-step	$K_{eq(IM\_SA1)}$	7.18E-21	3.41E-21	1.95E-21	1.71E-21	8.92E-22	4.86E-22
		$k_{uni(TS\_SA1)}$	6.74E+11	6.88E+11	6.98E+11	7.01E+11	7.13E+11	7.25E+11
		k_{SA_o}	4.84E-09	2.35E-09	1.36E-09	1.20E-09	6.36E-10	3.52E-10
	Stepwise	$K_{eq(IM\_SA')}$	1.87E-22	1.41E-22	1.15E-22	1.09E-22	8.63E-23	6.92E-23
		$k_{uni(TS\_SA')}$	1.05E+10	1.11E+10	1.15E+10	1.16E+10	1.22E+10	1.27E+10
		$k_{uni(TS\_SA1)}$	6.74E+11	6.88E+11	6.98E+11	7.01E+11	7.13E+11	7.25E+11
		$k_{uni(SA)}$	1.03E+10	1.09E+10	1.14E+10	1.15E+10	1.20E+10	1.26E+10
		k_{SA_s}	1.93E-12	1.54E-12	1.31E-12	1.25E-12	1.04E-12	8.69E-13
		v_{SA_s}/v_{SA_o}	9.49E+00	1.02E+01	1.09E+01	1.09E+01	1.18E+01	1.26E+01
Channel SW	One-step	$K_{eq(IM\_SW1)}$	6.31E-20	2.70E-20	1.42E-20	1.22E-20	5.79E-21	2.88E-21
		$k_{uni(TS\_SW1)}$	1.30E+11	1.36E+11	1.40E+11	1.41E+11	1.46E+11	1.51E+11
		k_{SW_o}	8.20E-09	3.66E-09	1.99E-09	1.72E-09	8.48E-10	4.36E-10
	Stepwise	$K_{eq(IM\_SW')}$	3.82E-22	2.95E-22	2.44E-22	2.33E-22	1.87E-22	1.52E-22
		$k_{uni(TS\_SW')}$	6.04E+10	6.50E+10	6.87E+10	6.96E+10	7.42E+10	7.87E+10
		$k_{uni(TS\_SW1)}$	1.30E+11	1.36E+11	1.40E+11	1.41E+11	1.47E+11	1.52E+11
		$k_{uni(SW)}$	4.12E+10	4.40E+10	4.61E+10	4.66E+10	4.93E+10	5.18E+10
		k_{SW_s}	1.58E-11	1.30E-11	1.12E-11	1.08E-11	9.19E-12	7.89E-12
		v_{SW_s}/v_{SW_o}	1.54E+04	1.34E+04	1.21E+04	1.20E+04	1.05E+04	9.57E+03
Channel SD	One-step	$K_{eq(IM\_SD1)}$	4.40E-21	2.06E-21	1.16E-21	1.01E-21	5.20E-22	2.79E-22
		$k_{uni(TS\_SD1)}$	9.83E+11	1.01E+12	1.03E+12	1.03E+12	1.05E+12	1.07E+12
		k_{SD_o}	4.33E-09	2.07E-09	1.19E-09	1.04E-09	5.47E-10	2.99E-10
	Stepwise	$K_{eq(IM\_SD')}$	1.34E-23	9.67E-24	7.59E-24	7.16E-24	5.42E-24	4.19E-24
		$k_{uni(TS\_SD')}$	3.06E+11	3.36E+11	3.60E+11	3.66E+11	3.98E+11	4.29E+11
		$k_{uni(TS\_SD1)}$	9.84E+11	1.01E+12	1.03E+12	1.03E+12	1.05E+12	1.08E+12
		$k_{uni(SD)}$	2.34E+11	2.52E+11	2.67E+11	2.70E+11	2.89E+11	3.07E+11
		k_{SD_s}	3.12E-12	2.44E-12	2.02E-12	1.94E-12	1.57E-12	1.29E-12
		v_{SD_s}/v_{SD_o}	9.51E+01	9.72E+01	9.83E+01	9.90E+01	1.01E+02	1.03E+02

For the stepwise route, the unimolecular rate constants (k_{uni}) have been calculated according to the RRKM model described as $\frac{1}{k_{uni(WD)}} = \frac{1}{k_{uni(TS\_WD')}} + \frac{1}{k_{uni(TS\_WD1)}}$, $\frac{1}{k_{uni(WT)}} = \frac{1}{k_{uni(TS\_WT')}} + \frac{1}{k_{uni(TS\_WT1)}}$, $\frac{1}{k_{uni(FA)}} = \frac{1}{k_{uni(TS\_FA')}} + \frac{1}{k_{uni(TS\_FA1)}}$, $\frac{1}{k_{uni(SA)}} = \frac{1}{k_{uni(TS\_SA')}} + \frac{1}{k_{uni(TS\_SA1)}}$, $\frac{1}{k_{uni(SW)}} = \frac{1}{k_{uni(TS\_SW')}} + \frac{1}{k_{uni(TS\_SW1)}}$ and $\frac{1}{k_{uni(SD)}} = \frac{1}{k_{uni(TS\_SD')}} + \frac{1}{k_{uni(TS\_SD1)}}$; v_s/v_o is the rate ratio between stepwise route and the corresponding one-step process in the hydrolysis reaction of $HNSO_2$ with $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$.

To check whether stepwise routes are favorable than the corresponding one-step process or not

in hydrolysis of $HNSO_2$ reaction with $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$, it is necessary to calculate the rate ratio (v_s/v_o) between stepwise route and the corresponding one-step process. The corresponding rate ratio (v_s/v_o) of the hydrolysis of $HNSO_2$ in the presence of $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$ can be respectively expressed in Eq. (S1) to Eq. (S6):

$$\frac{v_{WD\_s}}{v_{WD\_o}} = \frac{K_{eq(H_2O)_3} \times k_{WD\_s} \times [HNSO_2] \times [H_2O] \times [(H_2O)_2]}{K_{eq(HNSO_2\cdots H_2O)} \times k_{WD\_o} \times [HNSO_2] \times [H_2O] \times [(H_2O)_2]} = \frac{K_{eq(H_2O)_3} \times k_{WD\_s}}{K_{eq(HNSO_2\cdots H_2O)} \times k_{WD\_o}} \quad (S1)$$

$$\frac{v_{WT\_s}}{v_{WT\_o}} = \frac{K_{eq(H_2O)_4} \times k_{WT\_s} \times [HNSO_2] \times [(H_2O)_2] \times [(H_2O)_2]}{K_{eq(HNSO_2\cdots(H_2O)_2)} \times k_{WT\_o} \times [HNSO_2] \times [(H_2O)_2] \times [(H_2O)_2]} = \frac{K_{eq(H_2O)_4} \times k_{WT\_s}}{K_{eq(HNSO_2\cdots(H_2O)_2)} \times k_{WT\_o}} \quad (S2)$$

$$\frac{v_{FA\_s}}{v_{FA\_o}} = \frac{K_{eq(HCOOH\cdots H_2O)} \times k_{FA\_s} \times [HNSO_2] \times [H_2O] \times [HCOOH]}{K_{eq(HNSO_2\cdots H_2O)} \times k_{FA\_o} \times [HNSO_2] \times [H_2O] \times [HCOOH]} = \frac{K_{eq(HCOOH\cdots H_2O)} \times k_{FA\_s}}{K_{eq(HNSO_2\cdots H_2O)} \times k_{FA\_o}} \quad (S3)$$

$$\frac{v_{SA\_s}}{v_{SA\_o}} = \frac{K_{eq(H_2SO_4\cdots H_2O)} \times k_{SA\_s} \times [HNSO_2] \times [H_2O] \times [H_2SO_4]}{K_{eq(HNSO_2\cdots H_2O)} \times k_{SA\_o} \times [HNSO_2] \times [H_2O] \times [H_2SO_4]} = \frac{K_{eq(H_2SO_4\cdots H_2O)} \times k_{SA\_s}}{K_{eq(HNSO_2\cdots H_2O)} \times k_{SA\_o}} \quad (S4)$$

$$\frac{v_{SW\_s}}{v_{SW\_o}} = \frac{K_{eq(H_2SO_4\cdots(H_2O)_2)} \times k_{SW\_s} \times [HNSO_2] \times [H_2O] \times [H_2SO_4\bullet\bullet\bullet H_2O]}{K_{eq(HNSO_2\cdots H_2O)} \times k_{SW\_o} \times [HNSO_2] \times [H_2O] \times [H_2SO_4\bullet\bullet\bullet H_2O]} = \frac{K_{eq(H_2SO_4\cdots(H_2O)_2)} \times k_{SW\_s}}{K_{eq(HNSO_2\cdots H_2O)} \times k_{SW\_o}} \quad (S5)$$

$$\frac{v_{SD\_s}}{v_{SD\_o}} = \frac{K_{eq((H_2SO_4)_2\cdots H_2O)} \times k_{SD\_s} \times [HNSO_2] \times [H_2O] \times [(H_2SO_4)_2]}{K_{eq(HNSO_2\cdots H_2O)} \times k_{SD\_o} \times [HNSO_2] \times [H_2O] \times [(H_2SO_4)_2]} = \frac{K_{eq((H_2SO_4)_2\cdots H_2O)} \times k_{SD\_s}}{K_{eq(HNSO_2\cdots H_2O)} \times k_{SD\_o}} \quad (S6)$$

In Eq. (S1) to Eq. (S6), $K_{eq(HNSO_2\cdots H_2O)}$, $K_{eq((H_2O)_3)}$, $K_{eq((H_2O)_4)}$, $K_{eq(HNSO_2\cdots(H_2O)_2)}$, $K_{eq(HCOOH\cdots H_2O)}$, $K_{eq(H_2SO_4\cdots H_2O)}$, $K_{eq(H_2SO_4\cdots(H_2O)_2)}$ and $K_{eq((H_2SO_4)_2\cdots H_2O)}$ are respectively the equilibrium constant for the formation of $HNSO_2\cdots H_2O$, $(H_2O)_3$, $(H_2O)_4$, $HNSO_2\cdots(H_2O)_2$, $HCOOH\cdots H_2O$, $H_2SO_4\cdots H_2O$, $H_2SO_4\cdots(H_2O)_2$ and $(H_2SO_4)_2\cdots H_2O$. k_{WD_s}, k_{WT_s}, k_{FA_s}, k_{SA_s}, k_{SW_s} and k_{SD_s} in Eq. (S1) to Eq. (S6) are respectively the bimolecular rate coefficients for the stepwise routes of the hydrolysis reaction of $HNSO_2$ with $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$. k_{WD_o}, k_{WT_o}, k_{FA_o}, k_{SA_o}, k_{SW_o} and k_{SD_o} in Eq. (S1) to Eq. (S6) are respectively the bimolecular rate constant for the corresponding one-step process of the hydrolysis reaction of $HNSO_2$ with $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$. v_{WD_s}/v_{WD_o}, v_{WT_s}/v_{WT_o}, v_{FA_s}/v_{FA_o}, v_{SA_s}/v_{SA_o}, v_{SW_s}/v_{SW_o} and v_{SD_s}/v_{SD_o} are respectively the corresponding rate ratio (v_s/v_o) between the stepwise route and one-step process involved in the hydrolysis of $HNSO_2$ with $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$. It can be seen in Table S5 that the calculated rate ratio (v_s/v_o) for the hydrolysis of $HNSO_2$ with $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$ are

respectively 5.98×10^{0}-4.83×10^{0}, 9.52×10^{1}-1.54×10^{2}, 1.43×10^{0}-2.83×10^{0}, 9.49×10^{0}-1.26×10^{1}, 9.57×10^{3}-1.54×10^{4} and 9.51×10^{1}-1.03×10^{2} within the temperature range of 280-320 K. This reveal that stepwise routes in Channels WD, WT, FA, SA, SW and SD are more favorable than the corresponding one-step processes.

Table S6 Pseudo-first-order rate constants (k') in s^{-1} for the favorable routes in the hydrolysis reaction of $HNSO_2$ with catalyst *X* within the temperature range of 280-320 K

Reaction			T (K)	280	290	298	300	310	320
Channel WM	20% RH		k'_{WM}	3.78E-20	4.47E-20	4.97E-20	5.24E-20	6.04E-20	6.78E-20
	40% RH			7.55E-20	8.89E-20	1.03E-19	1.05E-19	1.21E-19	1.36E-19
	60% RH			1.14E-19	1.34E-19	1.49E-19	1.57E-19	1.81E-19	2.03E-19
	80% RH			1.52E-19	1.78E-19	2.06E-19	2.09E-19	2.42E-19	2.71E-19
	100% RH			1.89E-19	2.23E-19	2.55E-19	2.62E-19	3.02E-19	3.39E-19
Channel WD_s	20% RH		k'_{WD_s}	4.50E-18	5.52E-18	6.23E-18	6.80E-18	7.99E-18	9.00E-18
	40% RH			1.79E-17	2.19E-17	2.66E-17	2.71E-17	3.19E-17	3.60E-17
	60% RH			4.06E-17	4.94E-17	5.61E-17	6.10E-17	7.20E-17	8.10E-17
	80% RH			7.25E-17	8.75E-17	1.06E-16	1.08E-16	1.28E-16	1.44E-16
	100% RH			1.13E-16	1.37E-16	1.64E-16	1.69E-16	2.00E-16	2.25E-16
Channel WT_s	20% RH		k'_{WT_s}	1.99E-19	2.73E-19	3.30E-19	3.82E-19	4.93E-19	6.03E-19
	40% RH			1.58E-18	2.15E-18	2.91E-18	3.03E-18	3.94E-18	4.82E-18
	60% RH			5.38E-18	7.30E-18	8.91E-18	1.03E-17	1.33E-17	1.63E-17
	80% RH			1.28E-17	1.72E-17	2.33E-17	2.43E-17	3.17E-17	3.86E-17
	100% RH			2.48E-17	3.37E-17	4.46E-17	4.75E-17	6.16E-17	7.54E-17
Channel AM	0.1 ppbv		k'_{AM}	1.23E-22	5.33E-23	2.84E-23	2.44E-23	1.18E-23	5.98E-24
	10 ppbv			1.23E-20	5.33E-21	2.84E-21	2.44E-21	1.18E-21	5.98E-22
	2900 ppbv			3.58E-18	1.54E-18	8.21E-19	7.10E-19	3.45E-19	1.75E-19
Channel MA	80 ppbv		k'_{MA}	2.27E-21	8.92E-22	4.39E-22	3.71E-22	1.65E-22	7.71E-23
	180 ppbv			5.10E-21	2.00E-21	9.86E-22	8.33E-22	3.71E-22	1.73E-22
Channel FA_s	0.01 ppbv		k'_{FA_s}	1.81E-23	9.02E-24	5.36E-24	4.72E-24	2.59E-24	1.48E-24
	2 ppbv			1.38E-21	6.78E-22	4.14E-22	3.67E-22	1.97E-22	1.16E-22
	10 ppbv			1.81E-20	9.02E-21	5.36E-21	4.72E-21	2.59E-21	1.48E-21
Channel SA_s	$[SA]=10^8$		k'_{SA_s}	1.99E-21	7.96E-22	4.02E-22	3.31E-22	1.50E-22	7.12E-23
	$[SA]=10^6$			1.99E-23	7.96E-24	4.02E-24	3.31E-24	1.50E-24	7.12E-25
	$[SA]=10^4$			1.99E-25	7.96E-26	4.02E-26	3.31E-26	1.50E-26	7.12E-27
Channel SW_s	$[SA]=10^8$	100%RH	k'_{SW_s}	2.20E-19	5.14E-20	2.05E-20	1.65E-20	5.72E-21	2.16E-21
		20%RH		4.39E-20	1.03E-20	4.00E-21	3.31E-21	1.14E-21	4.32E-22
	$[SA]=10^6$	100%RH		2.20E-21	5.14E-22	2.05E-22	1.65E-22	5.72E-23	2.16E-23
		20%RH		4.39E-22	1.03E-22	4.00E-23	3.31E-23	1.14E-23	4.32E-24
	$[SA]=10^4$	100%RH		2.20E-23	5.14E-24	2.05E-24	1.65E-24	5.72E-25	2.16E-25
		20%RH		4.39E-24	1.03E-24	4.00E-25	3.31E-25	1.14E-25	4.32E-26
Channel SD_s	$[SA]=10^8$		k'_{SD_s}	4.56E-26	1.19E-26	2.36E-27	1.52E-27	2.36E-28	4.09E-29
	$[SA]=10^6$			4.56E-30	1.19E-30	2.36E-31	1.52E-31	2.36E-32	4.09E-33
	$[SA]=10^4$			4.56E-28	1.19E-28	2.36E-29	1.52E-29	2.36E-30	4.09E-31

k'_{WD_s}, k'_{WT_s}, k'_{FA_s}, k'_{SA_s}, k'_{SW_s} and k'_{SD_s} are respectively the pseudo-first-order rate constants (k') for the stepwise routes in the hydrolysis reaction of $HNSO_2$ with $(H_2O)_2$, $(H_2O)_3$, HCOOH, H_2SO_4, $H_2SO_4\cdots H_2O$ and $(H_2SO_4)_2$.

Table S7 Equilibrium constants ($cm^3 \cdot molecule^{-1}$), concentrations ($molecules \cdot cm^{-3}$) of catalysts, and rate coefficient ($cm^3 \cdot molecules^{-1} \cdot s^{-1}$) involved in the hydrolysis reaction of $HNSO_2$ with catalysts of $(H_2O)_2$, $(H_2O)_3$, NH_3, HCOOH and $H_2SO_4 \bullet\bullet\bullet H_2O$ at different altitudes in troposphere

Altitude	T(K)	P(atm)	$K_{eq((H_2O)_3)}$	$[(H_2O)_2]$	k_{WD_s}	$K_{eq((H_2O)_4)}$	$[(H_2O)_3]$	k_{WT_s}
5 km	259.3	0.535	6.32E-20	2.70E+12	5.91E-11	3.40E-19	2.32E+10	4.84E-12
10 km	229.7	0.266	4.52E-19	2.30E+11	2.73E-10	2.87E-18	3.25E+09	1.06E-11
15 km	212.6	0.120	1.85E-18	6.30E+06	8.19E-10	1.32E-17	1.52E+03	1.86E-11
Altitude	T(K)	P(atm)	$K_{eq(HNSO_2 \bullet\bullet\bullet H_2O)}$	$[NH_3]$	k_{AM}	$K_{eq(HCOOH \bullet\bullet\bullet H_2O)}$	[HCOOH]	k_{FA_s}
5 km	259.3	0.535	1.99E-22	7.60E+09	7.38E-10	9.21E-20	2.00E+10	1.15E-11
10 km	229.7	0.266	5.41E-22	8.50E+08	8.19E-10	6.55E-19	8.30E+09	2.33E-11
15 km	212.6	0.120	1.12E-21	1.20E+08	5.57E-10	2.68E-18	3.20E+09	3.94E-11
Altitude	T(K)	P(atm)	$K_{eq(H_2SO_4 \bullet\bullet\bullet (H_2O)_2)}$	$[H_2SO_4 \bullet\bullet\bullet H_2O]$	k_{SW_s}			
5 km	259.3	0.535	8.15E-18	1.82E+07	2.52E-11			
10 km	229.7	0.266	1.04E-16	7.23E+06	5.68E-11			
15 km	212.6	0.120	6.38E-16	5.65E+03	1.03E-10			

[redacted], [redacted], [redacted], [redacted], and [redacted] are the equilibrium constants for the formation of $(H_2O)_3$, $(H_2O)_4$, $HNSO_2 \cdots H_2O$, $HCOOH \cdots H_2O$ and $H_2SO_4 \cdots (H_2O)_2$, respectively. $[(H_2O)_2]$, $[(H_2O)_3]$, $[NH_3]$, [HCOOH] and $[H_2SO_4 \cdots H_2O]$ are the concentrations of $(H_2O)_2$, $(H_2O)_3$, HCOOH and $H_2SO_4 \cdots H_2O$, respectively. k_{WD_s}, k_{WT_s}, k_{AM}, k_{FA_s} and k_{SW_s} are the bimolecular rate coefficient for the favorable routes in the hydrolysis of $HNSO_2$ with $(H_2O)_2$, $(H_2O)_3$, NH_3, HCOOH and $H_2SO_4 \cdots H_2O$, respectively.

Previous reports have shown that atmospheric species concentrations various with increasing altitude. The available concentrations of $(H_2O)_2$ at 5, 10 and 15 km altitudes were respectively known to 2.70×10^{12}, 2.30×10^{11} and 6.30×10^{6} $molecules \cdot cm^{-3}$, whereas the concentration of $(H_2O)_3$ at the same altitudes were 2.32×10^{10}, 3.25×10^{9} and 1.52×10^{3} $molecules \cdot cm^{-3}$. Besides, NH_3 concentration has considered the highest mixing ratios of 5, 10 and 15 km altitudes, the values were respectively 7.6×10^{9}, 8.5×10^{8} and 1.2×10^{8} $molecules \cdot cm^{-3}$. Similarly, within the altitudes range of 5-15 km, HCOOH was found at 2.00×10^{10}, 8.30×10^{9} and 3.20×10^{9} $molecules \cdot cm^{-3}$. Using the reported concentrations of H_2O and H_2SO_4 along with the equilibrium constants for formation of $H_2SO_4 \cdots H_2O$, the concentrations of $H_2SO_4 \cdots H_2O$ at 5, 10, and 15 km altitudes shown in Table S7 were calculated to be 1.82×10^{7}, 7.23×10^{6} and 5.65×10^{3} $molecules \cdot cm^{-3}$, respectively.

Table S8 Coordinates stationary points for the hydrolysis reaction of $HNSO_2$ without and with catalyst *X* at the M06-2X/6-311+G(2*df*,2*pd*) level of theory

H_2O

O	0.00000000	0.00000000	0.11649900
H	0.00000000	0.76129500	-0.46599700
H	0.00000000	-0.76129500	-0.46599700

$(H_2O)_2$

O	1.50569100	-0.00009600	-0.12131200
H	1.93076400	0.00059800	0.73673000
H	0.55803900	0.00008800	0.06152900
O	-1.38593700	0.00008100	0.11160400
H	-1.72349600	0.76397800	-0.36083000
H	-1.72334100	-0.76454300	-0.35976300

CH_3NH_2

N	-0.74729300	0.00000200	-0.12044700
H	-1.15029600	-0.81204300	0.32716200
H	-1.15029800	0.81204300	0.32717400
C	0.70594000	0.00000100	0.01769600
H	1.11270000	-0.87684700	-0.48363600
H	1.11273700	0.87670500	-0.48387100
H	1.07056900	0.00012300	1.05012000

$(H_2O)_3$

O	-1.25089000	-1.00492400	-0.08799700
H	-1.22161500	-0.03734600	-0.01017700
O	-0.24942000	1.58467300	0.10860000
H	0.57945100	1.08120700	0.06108700
O	1.50506700	-0.57081700	-0.08165000
H	0.66496900	-1.05559600	-0.05303700
H	-1.92418000	-1.30794500	0.52199900
H	-0.21325000	2.22741900	-0.60112200
H	2.07657400	-0.97920100	0.56962600

HCOOH

C	-0.13522200	0.39793800	-0.00003200
H	-0.11135800	1.49276800	-0.00007700
O	1.11006500	-0.08857900	0.00012000
O	-1.12630400	-0.26471500	-0.00010800
H	1.05259500	-1.05404500	0.00017500

$HNSO_2$

S	0.02668400	-0.00171700	-0.00001100
O	1.09556900	-0.93859100	0.00000200
O	0.30048000	1.40011400	0.00001100
N	-1.36507300	-0.55342000	0.00002300
H	-2.03982900	0.20922800	-0.00008900

NH_3

N	0.00000000	0.00000000	0.11194200
H	0.00000000	0.94034000	-0.26119900
H	-0.81435800	-0.47017000	-0.26119900
H	0.81435800	-0.47017000	-0.26119900

$NH_3{\bullet\bullet\bullet}H_2O$

N	1.37891100	0.02294000	-0.00004000
H	1.57739400	-0.95680000	0.16129600
H	1.85294100	0.54844600	0.72331000
H	1.80151900	0.27356200	-0.88473400
O	-1.54449200	-0.10468200	0.00008500
H	-1.94306300	0.76568100	-0.00034500
H	-0.58523000	0.04599200	0.00007100

$CH_3NH_2{\cdots}H_2O$

N	-0.78569800	-0.75244000	-0.00001100
H	-1.14243200	-1.23642200	0.81340400
H	-1.14297700	-1.23658200	-0.81309100
C	-1.21125500	0.65103300	-0.00000900
H	-0.79295100	1.14424800	0.87510100
H	-0.79393300	1.14394600	-0.87575700
H	-2.29488300	0.78928800	0.00056100
O	1.90412500	0.19150900	0.00005000
H	1.08225800	-0.33083200	-0.00013500
H	2.61933300	-0.44484000	-0.00035400

$HCOOH{\cdots}H_2O$

C	1.19839900	-0.04039600	-0.00665800
H	2.28553200	0.09174000	-0.00943900
O	0.57785300	1.12411100	-0.00750800
O	0.66615500	-1.11987900	-0.00409900
H	-0.39345200	0.96661100	-0.00327300
O	-1.88444300	0.00399200	0.08362500
H	-1.35133000	-0.80116700	0.01618400
H	-2.60766900	-0.08060200	-0.53966500

***trans*-H_2SO_4**

S	-0.00000200	0.00004200	-0.15523500
O	-1.02361600	-0.67898100	0.83994100
O	1.02364600	0.67853700	0.84026200
O	0.65945300	-1.06393300	-0.82215200
O	-0.65948000	1.06436600	-0.82157100
H	-1.69533400	-0.03319500	1.09597500
H	1.69535400	0.03260600	1.09595600

***trans*-$H_2SO_4\cdots H_2O$**

S	-0.57805100	-0.09385500	0.10579100
O	-0.91547600	1.39868600	-0.31193300
O	0.33458500	-0.57611800	-1.05461000
O	0.20499100	0.02533900	1.29568400
O	-1.78038400	-0.84768500	0.06286100
H	-1.67611700	1.39465400	-0.90731700
H	1.28939700	-0.37045800	-0.82912900
O	2.67486900	-0.04464300	-0.05621500
H	2.28840400	0.09965900	0.81818500
H	3.19844900	0.73318900	-0.26069700

$(H_2SO_4)_2$

S	-1.91516900	-0.01102300	0.09807800
O	-1.56158400	-1.19150800	-0.86237300
O	-3.30777500	-0.03591000	0.31541500
O	-0.99566100	-0.05718400	1.21042300
O	-1.58895900	1.26459900	-0.74196200
H	-0.59782100	-1.36092400	-0.85854800
H	-0.62690900	1.44143400	-0.74301700
S	1.74259400	0.02132600	-0.07210100
O	3.29058300	0.07407600	-0.24145700
O	1.64756800	-0.02957800	1.47635400
O	1.22816000	1.25649300	-0.57022300
O	1.24108700	-1.20280200	-0.63083500
H	3.67198300	-0.81176200	-0.15971800
H	0.68659000	-0.05908700	1.70290100

***cis*-$H_2SO_4\cdots(H_2O)_2$**

H	-2.21657000	2.18361600	0.03481700
O	-1.66336700	1.40174800	-0.01351100
H	-2.25396600	0.61329300	-0.01717300
H	-3.34217500	-1.45059500	-0.57006900
O	-2.81262200	-0.98754500	0.08107500
H	-1.93200000	-1.39217700	0.06385700
S	0.98122800	-0.20239700	-0.00334500
O	-0.04448400	-1.20812100	-0.01351200
O	0.73610100	0.76507200	-1.21514100
O	2.34619900	-0.56438000	-0.00983500
O	0.73943300	0.73457100	1.23370600
H	-0.18182400	1.09914700	-1.15791400
H	-0.18319400	1.05431400	1.19774600

$(H_2SO_4)_2\cdots H_2O$

S	-2.01249900	-0.34617900	0.08218800
O	-1.39969600	-1.09105000	-1.14589500
O	-3.24331700	-0.96974300	0.37846300
O	-0.98121500	-0.27668300	1.11221200
O	-2.25277900	1.08856900	-0.40784900
H	-0.47596900	-0.79842200	-1.30967300
H	-1.45551100	1.70227800	-0.22271600
S	1.93439900	-0.13489700	-0.01508200
O	3.37591900	-0.70004700	-0.21793500
O	1.37626500	-1.12537400	1.01944700
O	2.09346200	1.17402900	0.53069800
O	1.19567600	-0.26115000	-1.24798500
H	3.35006600	-1.53721100	-0.70216200
H	0.38438100	-0.90623700	1.15198900
O	-0.29280900	2.63475400	0.02610600
H	-0.06027800	3.22420500	-0.69522100
H	0.53487700	2.22617600	0.32598700

IM

S	-0.51069900	-0.05265500	0.03102800
O	-0.57652300	-1.12475800	-0.89652200
O	-0.28746500	-0.30989200	1.41861100
N	-0.80105300	1.32091400	-0.49000100
H	-0.74728300	1.98936400	0.27500300
O	2.05517400	0.04392900	-0.18461200
H	2.48181900	0.03544600	0.67502400
H	2.51453400	0.69704300	-0.71627700

TS			
S	-0.29104700	0.04659800	-0.06342200
O	-0.98267900	-1.12045500	-0.46596700
O	-0.79079600	0.87494800	0.98552500
N	0.64636300	0.70707200	-1.14129100
H	0.59698700	1.71880200	-1.06797100
O	1.32895400	-0.65510300	0.59532900
H	1.52687200	-0.27557500	1.46139100
H	1.56452000	0.06657600	-0.30872500

H_2NSO_3H			
S	-0.04656200	0.04510000	-0.14196200
O	-1.07645800	-0.22804800	1.02994500
O	-0.12329900	-1.02590600	-1.08266100
O	-0.26977400	1.40317200	-0.49916500
N	1.39634800	-0.12922600	0.63201400
H	-1.07825000	-1.16989600	1.24451100
H	1.74263100	0.75183900	0.98921600
H	2.06241600	-0.59271100	0.02862400

IM_WM1			
S	-0.73400800	-0.24372400	0.08662400
O	-0.19133600	-0.33565400	1.40243800
O	-1.96937700	0.42839400	-0.14014200
N	-0.13188700	-1.18272500	-0.92219100
H	-0.62008200	-1.09679800	-1.80968100
H	1.97134100	-0.86842300	-0.43134700
H	2.56832000	-0.44130700	0.90500400
O	2.53429600	-0.19469300	-0.02330700
H	1.38962400	1.12134500	-0.25170500
O	0.50282600	1.50885600	-0.43293600
H	0.34687200	2.20863000	0.20866500

TS_WM1			
S	0.65229500	0.12209100	0.10088200
O	0.51705800	-0.31799800	1.45508800
O	1.87711600	-0.05669000	-0.60527100
N	-0.19218200	1.37486500	-0.20445600
H	0.11025200	1.78543200	-1.08185100
H	-1.64158500	0.85548500	-0.04980800
H	-2.67906400	-0.04606000	0.84404600
O	-2.36324900	0.08069400	-0.05658900
H	-1.52810400	-0.71642900	-0.35460500
O	-0.42673000	-1.13747400	-0.69289400
H	-0.18651300	-2.00419700	-0.34338100

IMF_WM1			
S	0.65858400	-0.06754000	0.07775300
O	0.47564300	-0.13905500	1.49074600
O	1.94508200	-0.21855100	-0.51421600
N	-0.03978800	1.38948100	-0.37239600
H	0.24029700	2.08934300	0.30653800
H	0.28574900	1.65492800	-1.29589700
H	-3.46073300	-0.12107500	-0.38339900
O	-2.62042500	-0.09178300	0.07791200
H	-2.26478600	0.79871900	-0.03105500
O	-0.28062800	-1.06906600	-0.66217000
H	-1.21673700	-0.92000400	-0.37163800

IM_WD′			
O	3.46909400	-1.29459900	0.02015900
H	3.60537200	-0.33535800	-0.06918400
O	3.00325300	1.50868200	-0.13507000
H	2.13478300	1.07843700	0.00403100
O	0.95389700	-0.15831900	0.27747600
H	1.39121700	-0.95886300	-0.07155700
H	4.00973300	-1.71003900	-0.65931100
H	3.07213100	2.19491800	0.54516100
H	0.73306200	-0.35469600	1.20127800
S	-1.77359200	-0.05339200	-0.03690400
O	-1.84100500	-1.13363200	-0.96104000
O	-2.10754100	-0.21312400	1.34862000
N	-1.79255800	1.31605100	-0.68292400
H	-1.84249400	2.05544600	0.01937000

TS_WD′			
O	-3.28736600	-1.06334400	0.09396400
H	-3.24594000	-0.08970200	0.08190600
O	-2.37099300	1.58010400	-0.01784000
H	-1.52470600	1.13543400	-0.20038500
O	-0.45591000	-0.92660100	-0.53250800
H	-1.32577400	-1.19509700	-0.15451100
H	-3.86291700	-1.30144000	0.82443200

IM_WD1			
S	-1.01465700	-0.25121600	0.18056200
O	-0.41942000	0.22862200	1.38790100
O	-2.37950300	0.04243900	-0.11487900
N	-0.33311300	-1.45850800	-0.41374900
H	-0.85833600	-1.79728200	-1.21423900
H	1.53636500	-1.45746900	-0.23924800
H	2.89826100	-1.74734200	0.45115800

H	-2.39033500	2.35063200	-0.59446300	O	2.47214300	-1.17909300	-0.19232200
H	-0.24685600	-1.54303600	-1.24064600	H	2.32569900	0.51647200	0.01674100
S	1.46417000	0.02919700	0.10089300	O	1.97806700	1.43288000	0.12239400
O	1.54136700	-0.75587700	1.28492600	H	1.71363900	1.48022800	1.04702100
O	2.13792400	-0.35165400	-1.09222400	H	0.66436300	1.40116700	-0.61790600
N	1.07342200	1.47232800	0.33008600	O	-0.26264200	1.20617700	-1.01688500
H	1.13567200	2.00873400	-0.53177200	H	-0.82285000	1.98503700	-0.92593900
		TS_WD1				**IMF_WD1**	
S	0.95430500	0.19970500	0.15126100				
O	0.36784200	-0.27125800	1.37650900	O	-1.56774000	-1.11912400	-0.08889800
O	2.34904900	-0.00375300	-0.09057700	H	-1.53496900	-0.14236300	-0.02909400
N	0.27498900	1.45926200	-0.36652700	O	-1.12948500	1.55923900	0.08600600
H	0.79243500	1.83905300	-1.15208700	H	-0.15142100	1.55581800	0.07585100
H	-1.43816800	1.39177400	-0.22582800	O	1.12885800	-1.55873600	-0.09717100
H	-2.79089200	1.57731900	0.56267000	H	0.15079300	-1.55435100	-0.08090500
O	-2.38747700	1.08712400	-0.15649600	H	-2.18941500	-1.40841600	0.58006800
H	-2.20741400	-0.40163000	-0.01841300	H	-1.40596200	2.14707300	-0.61819900
O	-1.86696400	-1.37190300	0.03944300	H	1.40878200	-2.15120800	0.60177100
H	-1.61032300	-1.48046300	0.96539000	O	1.56629700	1.11824700	0.09910900
H	-0.80843800	-1.32883500	-0.52367300	H	2.20371600	1.41434800	-0.55177200
O	0.28376900	-1.08731800	-0.97935400	H	1.53504500	0.14209000	0.02991400
H	0.89924600	-1.83046100	-0.97875200				
		IM_AM1				**TS_AM1**	
S	-0.74850500	-0.22015500	0.09229600	S	0.67053700	0.14950000	0.10149100
O	-0.11759000	-0.36652100	1.36653500	O	0.09928000	0.07120200	1.42057400
O	-1.96641700	0.51643300	-0.01705700	O	1.96203600	-0.41940100	-0.13485400
N	-0.31253900	-1.21078900	-0.95035300	N	0.19934400	1.37006800	-0.66651400
H	-0.86752600	-1.08396800	-1.79219300	H	0.74200700	1.46367500	-1.51909800
H	1.38982900	0.98595400	-0.31061300	H	-1.97844500	0.97579300	-0.23021000
O	0.48679900	1.40752100	-0.50123600	H	-1.48765800	-0.71651800	-0.36353900
H	0.34212100	2.12218000	0.12729300	O	-0.39235500	-1.17850100	-0.68880600
N	2.55569100	-0.15575000	0.01266500	H	-0.16215600	-2.01427000	-0.26637000
H	3.51199800	-0.09979700	-0.31202500	N	-2.39988500	0.07333400	0.00768100
H	2.11276300	-0.96156000	-0.42146700	H	-2.48121800	0.03096600	1.01817800
H	2.56249500	-0.33401900	1.01014500	H	-3.30901700	-0.02186200	-0.42630400
		IMF_AM1				**IM_MA1**	
S	0.63746200	0.03076000	0.14188100	S	1.11635500	-0.23673300	-0.10219600
O	-0.07736000	-0.52163000	1.24825600	O	0.27258500	-0.62209300	-1.19210700
O	2.00378400	0.42851100	0.26600100	O	2.34472000	0.44296200	-0.36376700
N	-0.24487300	1.23891600	-0.44935100	N	0.87794200	-0.99694100	1.17409600
H	0.14957400	1.65649000	-1.28025900	H	1.57848000	-0.73452900	1.86160000
H	-1.24422100	0.99044500	-0.49536700	H	-0.90458700	1.04592900	0.45752500
H	-3.07196900	-0.80223900	-0.67309400	O	0.03487400	1.47570700	0.34049200
O	0.59653700	-1.05394300	-1.04637100	H	0.03841900	2.02454600	-0.45099900

H	1.44348100	-1.51732600	-1.05755600
N	-2.83545000	-0.03802100	-0.05281200
H	-2.39562100	-0.44330000	0.76772600
H	-3.70206000	0.39400300	0.24050500

N	-2.11145800	0.05270800	0.66751400
H	-2.69864400	0.27023500	1.46350700
H	-1.57861100	-0.77961600	0.90956900
C	-2.92080300	-0.18380900	-0.53604900
H	-3.45778200	0.72731200	-0.79289900
H	-2.25186900	-0.43689000	-1.35492600
H	-3.64508700	-0.98938000	-0.41015200

TS_MA1

S	1.06848200	0.02654100	-0.21751100
O	0.24478000	0.91942900	-0.98506000
O	2.31784400	0.49590500	0.29637700
N	0.85087000	-1.42939100	-0.56015800
H	1.55100300	-1.99642000	-0.09231800
H	-0.98901600	-0.15190700	0.99251700
O	0.04654300	0.14398900	1.41341200
H	0.08747800	1.06029400	1.71021100
N	-2.01937600	-0.61613200	0.25994600
H	-2.57212600	-1.30647200	0.75472500
H	-1.50675100	-1.10408100	-0.47537200
C	-2.86199500	0.45443300	-0.30028500
H	-3.37054900	0.97082300	0.51051400
H	-2.21256100	1.15357900	-0.81968000
H	-3.60502500	0.06700300	-0.99506300

IMF_MA1

S	-0.99948900	-0.15902600	0.05381200
O	-0.15159200	-0.48797200	1.15618500
O	-2.18345800	-0.91125200	-0.21786400
N	-0.07012800	-0.07196300	-1.25228600
H	-0.55474000	0.24181800	-2.08070900
H	1.94329700	1.00188200	0.82459400
O	-1.47886100	1.36445300	0.25073100
H	-2.36779800	1.35298700	0.62724100
N	2.38017000	0.74419500	-0.05315500
H	3.07430400	1.44883800	-0.26495100
H	0.84711000	0.36819600	-1.04392700
C	2.99524300	-0.58201900	0.07195300
H	3.76623900	-0.63950200	0.84321900
H	2.21449600	-1.30195100	0.30932300
H	3.43845600	-0.86318500	-0.88183100

IM_FA′

C	3.60036600	0.15244300	0.02694900
H	4.62759400	0.53461200	0.04980100
O	2.72054400	1.14068500	-0.04128300
O	3.33332100	-1.02507000	0.07038800
H	1.81728300	0.75064100	-0.04667200
O	0.54055100	-0.66770700	-0.14386800
H	1.13333600	-1.38225300	0.14449600
H	0.44177400	-0.76865100	-1.10193200
S	-1.98232700	0.03153500	0.03486700
O	-2.59628600	-0.71849200	-1.00842100
O	-2.14226200	-0.32145300	1.40252500
N	-1.63808900	1.45880100	-0.35051200
H	-1.28527600	1.97113500	0.45359100

TS_FA′

C	3.35932000	0.14850300	0.10268900
H	4.39324400	0.50730200	0.21450200
O	2.53780000	1.14836800	-0.22858300
O	3.02666200	-1.00834300	0.25585000
H	1.62676500	0.77925500	-0.30971300
O	0.21876400	-1.04348900	-0.21765800
H	1.00187200	-1.53750400	0.07449300
H	0.04897600	-1.29260500	-1.13720900
S	-1.77284200	0.10102600	0.05356800
O	-2.38327600	-0.47235300	-1.10658800
O	-2.23524700	-0.21183600	1.36342300
N	-1.06097400	1.41512700	-0.21474600
H	-0.75211700	1.83144800	0.65636400

IM_FA1

S	-1.23741000	-0.25059500	0.10837600
O	-0.80352600	-0.14237100	1.45984100
O	-2.58210900	0.02787600	-0.26327700
N	-0.38216800	-1.16589200	-0.74548200
H	-0.79072200	-1.29099300	-1.66596400
H	0.53242900	1.38081600	-0.34500400

TS_FA1

S	-1.19758900	-0.18221700	0.10433100
O	-0.85086100	-0.06391600	1.48180600
O	-2.53033400	0.02664100	-0.34891900
N	-0.33864800	-1.21375700	-0.63581600
H	-0.69784700	-1.40029900	-1.56474200
H	0.69997200	1.23498500	-0.32532600

O	-0.46153200	1.50446500	-0.50956600
H	-0.78031000	2.21114600	0.06434600
C	2.63259000	0.05545000	0.10605400
H	3.68432200	0.08020100	0.40408400
O	2.19013500	-1.15046400	-0.04465000
O	2.00931000	1.09017400	-0.05360000
H	1.21425400	-1.18053100	-0.31941400

IMF_FA1

S	-1.22136100	0.08201000	0.11051000
O	-0.27046200	1.04631800	0.59006300
O	-2.59552500	0.19429000	0.46883500
N	-0.65284900	-1.36957700	0.48010900
H	-1.22658500	-2.13231100	0.14917500
H	1.45261200	1.12168500	0.28927400
O	-1.15136400	0.10488700	-1.48782100
H	-1.84585900	0.68875000	-1.82070500
C	2.83677400	-0.11078900	-0.09762600
H	3.91202900	-0.13777200	-0.30088600
O	2.15335000	-1.10435100	-0.07545900
O	2.41973900	1.11568700	0.10808300
H	0.35298400	-1.45538700	0.31037200

TS_SA′

S	2.47630100	0.02602100	0.13452400
O	3.00563200	-0.50208600	-1.26771700
O	1.39114400	1.08185800	-0.28522100
O	1.81620500	-1.10876800	0.72931200
O	3.55525100	0.70932900	0.78155400
H	3.77532500	0.05698800	-1.53589800
H	0.51264900	0.60369100	-0.35371400
O	-0.93358400	-1.18890400	0.07390300
H	-0.14400200	-1.50154000	0.56393300
H	-1.19771900	-1.90264100	-0.52540200
S	-2.78313700	0.17581600	-0.00791100
O	-3.50839200	-0.67792100	-0.88604100
O	-2.98582000	0.15910700	1.40752900
N	-2.14558600	1.38836800	-0.66172500
H	-1.74126000	2.01462500	0.03080700

TS_SA1

S	1.93604300	-0.00462200	-0.10280600
O	1.82264500	0.48066600	1.40030700
O	1.25709500	-1.34847000	-0.12368500
O	1.12887900	0.97822200	-0.81355400

O	-0.40074700	1.35996000	-0.52768100
H	-0.75513600	2.11926900	-0.04621300
C	2.56475700	0.03871800	0.09037900
H	3.62307300	0.09270600	0.35578400
O	2.10327600	-1.13739000	-0.01147100
O	1.95682900	1.10620000	-0.08079600
H	1.04805100	-1.19916800	-0.28386500

IM_SA′

S	2.54741200	0.08068100	0.13751000
O	3.06130600	-0.73806000	-1.12198700
O	1.27266200	0.78560800	-0.40561300
O	2.15743000	-0.91268700	1.08560700
O	3.51149000	1.08376300	0.43143600
H	3.61594700	-0.16094000	-1.66624700
H	0.48656700	0.17916600	-0.25965700
O	-0.59962500	-1.01058800	0.10293800
H	0.03341300	-1.52405900	0.62377800
H	-0.87709600	-1.58628700	-0.62721100
S	-2.93291500	0.13539200	-0.00451800
O	-3.52768800	-0.91660800	-0.75539800
O	-3.12732800	0.27224400	1.39933900
N	-2.42795900	1.29633900	-0.83498300
H	-2.08106800	2.05119500	-0.24421900

IM_SA1

S	1.95879900	0.04474800	-0.09470100
O	1.79434100	0.12769500	1.47360700
O	1.33439700	-1.30379400	-0.46443400
O	1.13177500	1.12030200	-0.58362800
O	3.33983900	0.01949600	-0.41640700
H	2.47141700	-0.41401600	1.90025800
H	0.30456700	-1.25442900	-0.59831600
S	-2.02277200	-0.26623800	0.12683500
O	-1.44057400	-0.14407800	1.41753500
O	-3.40983100	-0.05884000	-0.09744400
N	-1.22087600	-1.12048800	-0.83575700
H	-1.70945800	-1.25630000	-1.71442600
O	-1.42423100	1.56195400	-0.55189700
H	-0.42971700	1.49191400	-0.55965700
H	-1.67282400	2.23820800	0.08962900

IMF_SA1

S	2.05232900	-0.01135200	0.14249200
O	2.57767500	0.89082000	-1.04666800
O	1.09637800	0.80892300	0.82057000
O	1.31831600	-1.16459100	-0.58616200

O	3.30650200	-0.11017400	-0.45527600	O	3.16796400	-0.56931600	0.81914800
H	2.45397100	-0.00419600	1.94781800	H	3.39233500	0.51087500	-1.40129800
H	0.13711000	-1.31831800	-0.32909300	H	-0.70431400	1.53524700	0.63940900
S	-2.01088900	-0.19507700	0.14231700	S	-2.08535200	-0.00170700	-0.13337700
O	-1.54730500	0.05473300	1.46127600	O	-1.10840100	-0.37220900	-1.12732000
O	-3.37093100	-0.04392100	-0.23165900	O	-3.46873200	-0.05781500	-0.45905400
N	-1.17564100	-1.23481600	-0.60650800	N	-1.69365800	1.45462400	0.40892800
H	-1.58226300	-1.49771500	-1.49649100	H	-2.33369100	1.81098200	1.10542200
O	-1.31741100	1.39222700	-0.69165900	O	-1.83653400	-0.93401900	1.13554400
H	-0.28755100	1.26936000	-0.72550500	H	0.39147800	-0.88833200	-0.83404200
H	-1.53002600	2.16349600	-0.14935200	H	-2.33516300	-1.75652900	1.03369200
		IM_SW′				**TS_SW′**	
H	0.49027800	2.93713900	0.48365900	H	-0.29491200	2.93572400	-0.00267900
O	0.76332100	2.06304300	0.20313700	O	-0.60659400	2.03736800	0.12899900
H	0.01064400	1.65361700	-0.28412400	H	0.11587200	1.52996800	0.56364800
H	-1.27567200	0.53824400	-2.03512500	H	1.46549300	-0.55047500	2.31305500
O	-0.98558000	0.44495100	-1.12544900	O	1.23708400	-0.38870100	1.34843100
H	-0.36217500	-0.29800700	-1.10279400	H	0.45696400	-0.93231800	1.15849000
S	2.60940400	-0.43013200	-0.01173800	S	-2.51127600	-0.38397900	-0.03980900
O	1.40641700	-0.87508600	-0.66001500	O	-1.36951500	-0.97187100	0.60729500
O	3.09350600	0.88800200	-0.71289800	O	-2.97005600	0.85701600	0.80360300
O	3.72346300	-1.29135000	0.10338000	O	-3.65969000	-1.16022800	-0.32325800
O	2.23668200	0.06883900	1.43040400	O	-2.03258800	0.27439000	-1.38330000
H	2.35434500	1.53390000	-0.69861000	H	-2.19375200	1.44235300	0.92440100
H	1.51791700	0.72771300	1.34701800	H	-1.28782300	0.87412000	-1.17883800
S	-3.10134300	-0.26688100	0.14723300	S	2.88669500	-0.14871100	-0.24643000
O	-3.93089000	0.03349500	-0.96830500	O	3.91815000	-0.14857900	0.73424000
O	-2.70927700	-1.60498000	0.44958000	O	2.56764000	-1.31081700	-0.99888500
N	-3.02064500	0.81971700	1.19330800	N	2.46773800	1.22165000	-0.72356700
H	-2.50092600	0.50627000	2.01022600	H	1.78184300	1.16349600	-1.47030000
		IM_SW1				**TS_SW1**	
S	-2.02309600	-0.30371100	-0.06031200	S	-1.98616200	-0.27915900	-0.04450800
O	-1.38630000	-1.10216300	1.12624900	O	-1.32633000	-1.13064300	1.07976600
O	-2.15617100	1.12532000	0.45487200	O	-2.01017200	1.13660800	0.46467400
O	-1.04104900	-0.28619100	-1.14374300	O	-1.04835000	-0.31372800	-1.19689200
O	-3.29038600	-0.87560400	-0.30809600	O	-3.27766700	-0.79621600	-0.28102100
H	-0.48801900	-0.76077700	1.32626700	H	-0.42462700	-0.77737100	1.29562200
H	-1.34552600	1.73660200	0.19275900	H	-1.14574300	1.77663800	0.18716000
S	2.13107500	-0.09209900	0.18362100	S	2.04912100	-0.13993100	0.14910600
O	1.15043700	-0.15350500	1.23542500	O	1.09703900	-0.12589300	1.23969000
O	3.32299300	-0.86078800	0.29298600	O	3.24219200	-0.90560800	0.28602800
N	2.08561100	1.20697000	-0.57748800	N	2.05123300	1.18625700	-0.58213000
H	2.84668000	1.27240700	-1.24592400	H	2.79716800	1.23971300	-1.26672100
H	0.58247400	2.13925100	-0.36546700	H	0.69281600	2.10084100	-0.33989000

H	-0.04537700	3.24558100	0.54028100
O	-0.25800300	2.60617300	-0.14385500
H	0.25581600	-0.95748300	-1.14246800
O	1.23327900	-1.27817800	-1.07304500
H	1.26861200	-2.19192900	-0.76232000

H	0.03710200	3.18234000	0.61381800
O	-0.16177200	2.58375800	-0.11150000
H	0.07605100	-0.84737700	-1.10869500
O	1.15576300	-1.21494200	-1.01064800
H	1.23564400	-2.13983200	-0.74072600

IMF_SW1

S	-2.14577700	-0.13563200	-0.04130100
O	-1.77594300	-0.32226700	1.46477800
O	-1.41415900	0.98622700	-0.56462900
O	-1.55622900	-1.42294700	-0.71823000
O	-3.55348000	-0.19882100	-0.13460700
H	-0.80304600	-0.30544100	1.56431300
H	-0.31796900	2.43660700	-0.31498000
S	1.99036600	-0.42266300	0.15164400
O	0.94437300	-0.37251500	1.12781500
O	3.10525500	-1.30113400	0.26672100
N	2.49850600	1.06387100	-0.08583700
H	3.29949300	1.15475900	-0.69233400
H	1.77836600	1.80646700	-0.11247100
H	0.29170100	3.57362100	0.53288000
O	0.48426300	2.96435700	-0.18193300
H	-0.62518800	-1.26511000	-0.96021100
O	1.21369600	-0.88760500	-1.19547400
H	1.79147200	-1.47763100	-1.69736300

IM_SD′

S	2.48351700	-1.48703500	-0.02752200
O	2.95524300	-2.40828700	1.14823800
O	3.04275000	-2.29338400	-1.22711000
O	1.04826700	-1.50887200	-0.03192600
O	3.15218800	-0.20936800	0.08546300
H	3.84530800	-2.14158700	1.43772100
H	2.67768900	-1.88261600	-2.06040800
S	0.68194900	2.40739400	0.05901900
O	1.90324200	1.85637700	-0.75338900
O	-0.52478000	2.13637200	-0.89267300
O	0.54926200	1.55512700	1.33909300
O	0.84288800	3.81039400	0.24146000
H	2.34732400	1.09482000	-0.26010000
H	-0.97540100	1.28615000	-0.64978700
S	-3.64068800	-0.82551500	-0.02014300
O	-3.29357200	-2.07573500	-0.65940400
O	-4.25863800	-0.85647500	1.26713500
N	-3.96218900	0.24632700	-1.03374400
H	-4.53130000	0.99170600	-0.63869500
O	-1.23840700	-0.20996300	0.46948800
H	-0.91209700	-1.13636300	0.29052900
H	-0.54018200	0.17661200	1.04428800

TS_SD′

S	2.36537300	-1.60510500	-0.02566200
O	2.86732100	-2.45539800	1.18601800
O	2.95216500	-2.43351500	-1.19794000
O	0.93227500	-1.67956100	-0.02514000
O	2.98505700	-0.30028900	0.02698400
H	3.76040900	-2.17388500	1.44509800
H	2.60429200	-2.03610400	-2.04251000
S	0.71638800	2.50475200	0.05147000
O	1.96551600	1.93766800	-0.70798500
O	-0.46115100	2.19450200	-0.91349500
O	0.56829800	1.67829000	1.34917100
O	0.87583300	3.90454800	0.19382600

IM_SD1

S	-2.40800500	-0.81905400	0.05428600
O	-3.75256300	-1.59727900	0.16329000
O	-2.66289200	-0.01854700	-1.24736200
O	-2.35986800	0.06154700	1.18129500
O	-1.33415000	-1.74900200	-0.13670700
H	-3.72995500	-2.39397300	-0.38621000
H	-1.86442700	0.55085100	-1.39930200
S	2.33905500	-1.19681200	0.07178300
O	1.31002700	-1.74831800	-0.96376700
O	3.00430100	-0.03295300	-0.66630000
O	1.56643200	-0.67914600	1.19884500
O	3.30698400	-2.19020300	0.33962100

H	2.33581600	1.13799000	-0.24561000
H	-0.91436600	1.34927100	-0.64927900
S	-3.56402500	-0.83064100	-0.01214600
O	-3.25407800	-2.11395200	-0.61181000
O	-4.14544800	-0.79290100	1.29089500
N	-3.88713400	0.20256600	-1.06482300
H	-4.43642400	0.97288600	-0.69606100
O	-1.16798700	-0.07628600	0.42010800
H	-0.82261000	-0.99097000	0.25604700
H	-0.54334400	0.31388900	1.08243600

H	0.38948500	-1.66135300	-0.64538900
H	2.50458400	0.86222900	-0.56284500
S	0.22761100	2.11606300	-0.18059200
O	-0.28328500	1.05589700	-1.02347600
O	-0.57459800	3.26295000	0.06015400
N	1.73352200	2.21650600	-0.29555400
H	2.09854600	3.03737400	0.17526100
O	0.04486100	1.27220000	1.44768200
H	-0.87629700	0.93211000	1.53574500
H	0.68285500	0.45690300	1.39777300

TS_SD1

S	-2.41449600	-0.78359200	0.05585100
O	-3.77412000	-1.53285000	0.17412000
O	-2.64823100	0.00626100	-1.25370400
O	-2.34733000	0.10892800	1.17365300
O	-1.35889000	-1.73800900	-0.11904800
H	-3.76868500	-2.33658400	-0.36557800
H	-1.84069100	0.56938700	-1.40056800
S	2.31776000	-1.19849600	0.05418800
O	1.27523900	-1.78544500	-0.94697800
O	2.93153500	-0.03341300	-0.68111500
O	1.54008600	-0.70135500	1.20948500
O	3.28607300	-2.18378600	0.34945900
H	0.35646200	-1.70242200	-0.62174000
H	2.42911000	0.93513200	-0.54876800
S	0.25322900	2.08128700	-0.15092800
O	-0.28306400	1.07854100	-1.04839300
O	-0.48810800	3.26482700	0.10345000
N	1.76956600	2.15752600	-0.30888400
H	2.17409100	2.94775400	0.17953700
O	0.09564900	1.22203700	1.38455200
H	-0.83674600	0.91479600	1.49618300
H	0.76489200	0.33617300	1.31348900

IMF_SD1

S	-1.63734200	1.55384900	0.02470800
O	-2.32045500	2.94507900	0.24964000
O	-0.83689300	1.76134600	-1.25726700
O	-0.75646900	1.36788400	1.14277100
O	-2.69020600	0.60313500	-0.19515300
H	-3.18976000	2.96094800	-0.17398500
H	0.16021700	1.60402500	-1.11379200
S	-0.70608200	-2.19904800	-0.08004900
O	-2.24586500	-2.06693200	-0.01378800
O	-0.16037100	-1.08336900	-0.81564800
O	-0.31330300	-1.98859100	1.43053500
O	-0.39101000	-3.52237800	-0.47058700
H	-2.50335500	-1.11560100	-0.04522500
H	1.86326200	-1.20678400	-1.17581000
S	2.47382600	0.59573100	-0.01883000
O	1.64213800	1.47283500	-0.81585800
O	3.72911200	1.05252600	0.45358300
N	2.71435200	-0.75116700	-0.86208100
H	3.44184600	-1.34517000	-0.49114000
O	1.58583400	0.18717200	1.23001600
H	0.74577800	0.71606900	1.22941900
H	0.45502200	-1.39347800	1.48589300