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Corrigendum to

"From quantum chemical formation free energies to evaporation rates" published in Atmos. Chem. Phys., 12, 225–235, 2012

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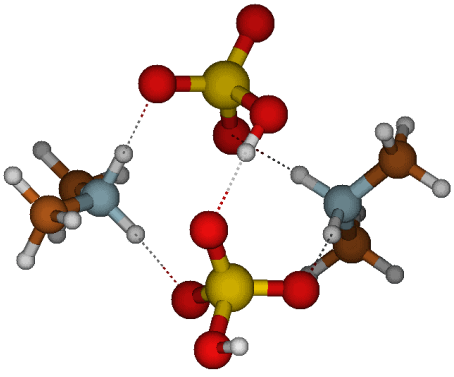

Fig. 1a. Most stable $(H_2OS_4)_2((CH_3)_2NH)_2$ cluster structure, Yellow, red, white, blue, and brown spheres represent sulfur, oxygen, hydrogen, nitrogen and carbon atoms, respectively.

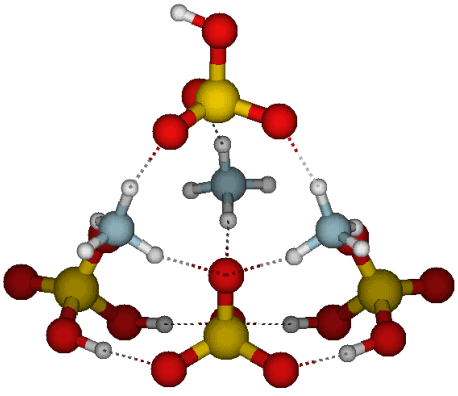

Fig. 1b. Most stable $(H_2OS_4)_4(NH_3)_3$ cluster structure, Yellow, red, white, and blue spheres represent sulfur, oxygen, hydrogen and nitrogen atoms, respectively.

We have found an error in our paper "From quantum chemical formation free energies to evaporation rates" by I. K. Ortega et al. (2012). We have discovered that the single point energy calculation for the $(H_2OS_4)_2((CH_3)_2NH)_2$ cluster was wrong. While the electronic energy used in the paper was $-1047222.51\,\mathrm{kcal\,mol^{-1}}$, the correct value is $-1047218.44\,\mathrm{kcal\,mol^{-1}}$, and thus the stability of the cluster was overestimated by $4.07\,\mathrm{kcal\,mol^{-1}}$. The conformer that was reported to be the most stable is not actually the most stable. Figure 1a shows the structure of the most stable conformer after correcting the error.

The formation free energy from monomers for this cluster is $-54.26\,\mathrm{kcal\,mol^{-1}}$, while the one reported in the manuscript was $-56.17\,\mathrm{kcal\,mol^{-1}}$. In addition, we have performed further conformational sampling for some other clusters included in the study and have found a better conformer for the $(H_2SO_4)_4(NH_3)_3$ cluster (Fig. 1b). This new conformer is $1.20\,\mathrm{kcal\,mol^{-1}}$ more stable than the one reported previously.

The overall conclusions of the article are not affected by these errors. According to the corrected results, $(H_2OS_4)_2((CH_3)_2NH)_1$ is the most stable two sulfuric acid cluster containing DMA, although the evaporation rate of this clusters is very close to the evaporation rate of $(H_2OS_4)_2((CH_3)_2NH)_2$ (Fig. 4, right panel). The local minimum of the evaporation rate for sulfuric acid–DMA clusters

Table 3. Gibbs free energy for the reaction $((H_2SO_4)_{n-1}\ (NH_3/\ (CH_3)_2NH)_m + H_2\ SO_4 \rightarrow (H_2SO_4)_n\ (NH_3/\ (CH_3)\ _2NH)_m)$ in kcal mol^{-1} at 298.15 K.

	(m) Ammonia			
Sulfuric acid (n)	1	2	3	4
1	−7.61	−16.63	−21.70	−27.06
2	−16.21	−20.89	−25.25	−26.11
3	−10.12	−16.08	−25.28	−28.27
4	−8.37	−11.07	**−15.35**	−20.08
	(m) DMA			
Sulfuric acid (n)	1	2	3	4
1	−15.40	−23.83	−30.67	−33.85
2	−19.48	**−33.97**	−37.62	−40.26
3	−12.92	**−13.00**	−27.24	−36.99
4	−7.62	−15.53	−15.15	−18.34

Table 4. Gibbs free energy for the reaction $((H_2SO_4)_n(NH_3/(CH_3)_2NH)_{m-1} + NH_3/(CH_3)_2NH \rightarrow (H_2SO_4)_n(NH_3/(CH_3)_2NH)_m)$ in kcal mol^{-1} at 298.15 K.

	(m) Ammonia			
Sulfuric acid (n)	1	2	3	4
1	−7.61	−4.82	−0.57	−2.05
2	−15.93	−9.50	−4.93	−2.91
3	−19.64	−15.46	−14.13	−5.90
4	−19.53	−18.16	**−18.41**	−11.83
	(m) DMA			
Sulfuric acid (n)	1	2	3	4
1	−15.40	−4.89	−4.79	−3.38
2	−26.99	**−19.38**	**−8.43**	−6.02
3	−33.50	−19.46	−22.68	−15.77
4	−32.64	−27.37	−22.30	−18.96

is now the cluster $(H_2OS_4)_3((CH_3)_2NH)_3$, but the evaporation rate of this cluster is relatively close to the evaporation rate of $(H_2OS_4)_2((CH_3)_2NH)_1$ (Fig. 4, right panel).

The following tables and figures are the corrected versions accounting for the errors (new values indicated in bold):

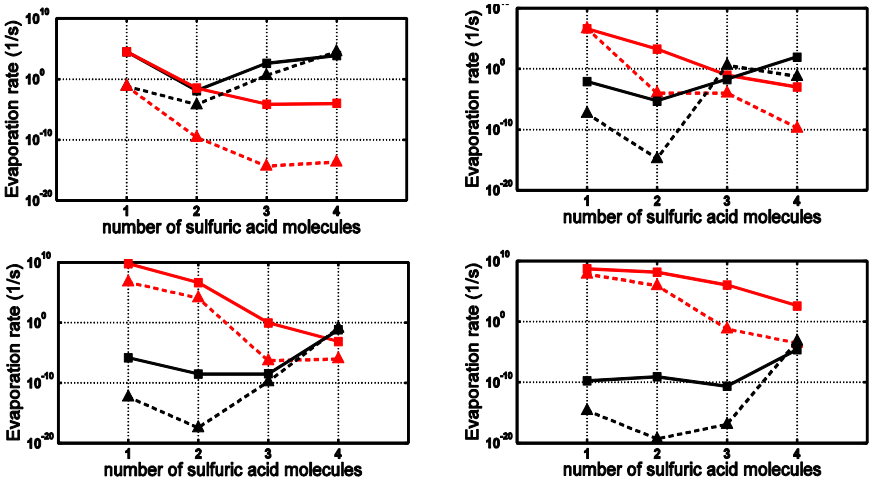

Fig. 2. Evaporation rates of acid (black lines) and base molecules (red lines) from ammonia (solid lines) and DMA (dashed lines) containing clusters. The top left, top right, bottom left, and bottom right panels give the results for 1, 2, 3, and 4 base containing clusters, respectively.

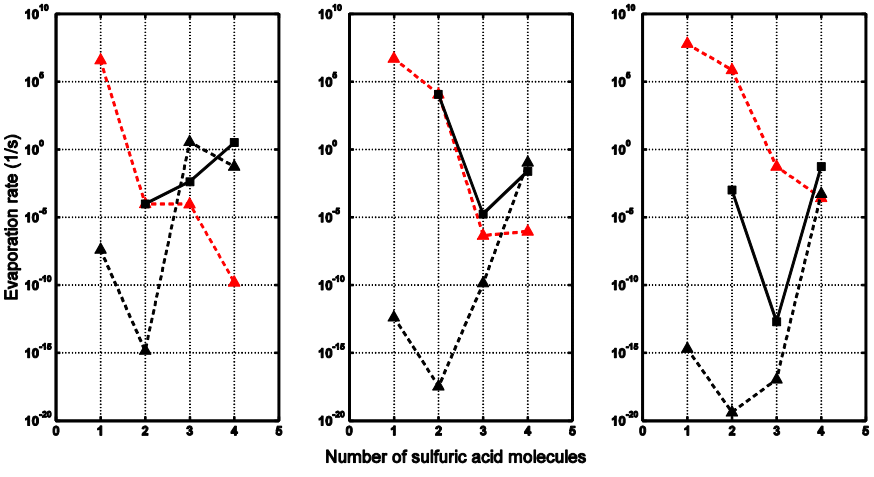

Fig. 3. Evaporation and fission (non-monomer evaporation) rates from DMA–H_2SO_4 clusters. Left panel: clusters with two DMA molecules. Middle panel: clusters with three DMA molecules. Right panel: clusters with four DMA molecules. Red dashed lines are the evaporation of DMA from the cluster, black dashed lines are the evaporation of sulfuric acid from the cluster, and solid black lines are the cluster fission reactions leading to the formation of the most stable daughter cluster ($(H_2SO_4)2 \times (CH_3)2NH$ for the fission of two DMA clusters and $(H_2SO_4)2 \times ((CH3)2NH)2$ for the fission of three and four DMA clusters).

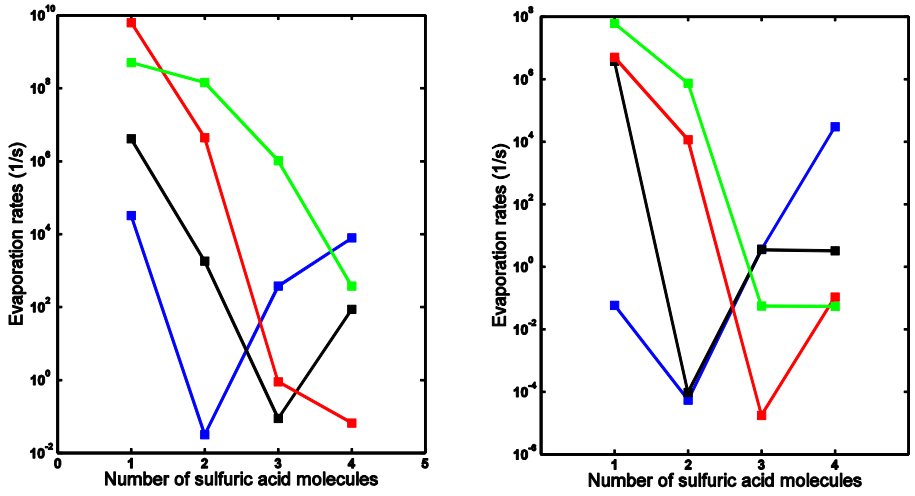

Fig. 4. Evaporation rates of different clusters versus number of sulfuric acid molecules in the cluster. The blue, black, red, and green lines represent clusters containing one, two, three, and four base molecules, respectively. The left panel is for ammonia clusters, while the right panel is for those containing DMA.

Table 5. Evaporation rates ($1\,s^{-1}$) for different clusters included in this work. (* typographic error corrected).

	Sulfuric acid evaporation rate	Ammonia evaporation rate	Sulfuric acid evaporation rate	DMA evaporation rate
Sulfuric acid (n)		1 Ammonia		1 DMA
1	3.24×10^{4}	3.24×10^{4}	5.94×10^{-2}	5.94×10^{-2}
2	1.22×10^{-2}	3.27×10^{-2}	5.47×10^{-5}	2.20×10^{-10}
3	3.83×10^{2}	7.30×10^{-5}	3.7	4.20×10^{-15}
4	7.97×10^{3}	9.91×10^{-5}	3.04×10^{4}	1.97×10^{-14}
		2 Ammonia		2 DMA
1	7.43×10^{-3}	4.11×10^{6}	4.10×10^{-8}	3.76×10^{6}
2	4.86×10^{-6}	1.83×10^{3}	$\mathbf{1.44 \times 10^{-15}}$	$\mathbf{9.66 \times 10^{-5}}$
3	1.73×10^{-2}	8.99×10^{-2}	**3.57**	9.31×10^{-5}
4	87.4	1.05×10^{-3}	5.32×10^{-2}	1.59×10^{-10}
		3 Ammonia		3 DMA
1	1.42×10^{-6}	5.93×10^{9}	4.24×10^{-13}	5.10×10^{6}
2	3.27×10^{-9}	4.40×10^{6}	3.41×10^{-18}	$\mathbf{1.15 \times 10^{4}}$
3	3.27×10^{-9}	0.9	1.44×10^{-10}	4.48×10^{-7}
4	$\mathbf{6.62 \times 10^{-2}}$	$\mathbf{7.12 \times 10^{-4}}$	0.1	9.03×10^{-7}
		4 Ammonia		4 DMA
1	1.70×10^{-10}	5.30×10^{8}	2.11×10^{-15}	6.10×10^{7}
2	8.03×10^{-10}	1.42×10^{8}	4.27×10^{-20}	7.49×10^{5}
3	2.20×10^{-11}	1.01×10^{6}	7.17×10^{-15}	36.9
4	2.35×10^{-5}	49.8	$^{*}\mathbf{5.26 \times 10^{-4}}$	2.72×10^{-4}

Table S3.1.

	(m) Ammonia				
Sulfuric acid (n)	0	1	2	3	4
0	–	–	4.20	8.70	12.01
1	–	−7.61	−12.43	−13.00	−15.05
2	−7.89	−23.82	−33.32	−38.25	−41.16
3	−14.30	−33.94	−49.40	−63.53	−69.43
4	−22.78	−42.31	−60.47	−**78.88**	−89.51
	(m) DMA				
Sulfuric acid (n)	1	2	3	4	
0	–	3.54	5.59	5.39	
1	−15.40	−20.29	−25.08	−28.46	
2	−34.88	−**54.26**	−62.70	−68.72	
3	−47.80	−67.26	−89.94	−105.71	
4	−55.42	−82.79	−105.09	−124.05	

Gibbs free energies of formation from monomers at 298.15 K in kcal mol^{-1}.

	(m) Ammonia				
Sulfuric acid (n)	0	1	2	3	4
0	–	–	−1.62	−7.45	−12.30
1	–	−16.00	−29.70	−40.45	−51.30
2	−17.85	−45.00	−64.46	−79.53	−91.64
3	−35.82	−66.06	−92.09	−117.57	−132.80
4	−52.60	−87.40	−115.13	−**145.17**	−164.35
	(m) DMA				
Sulfuric acid (n)	1	2	3	4	
0	–	−3.17	−12.68	−21.30	
1	−24.65	−40.08	−56.99	−72.24	
2	−57.06	−**87.57**	−108.24	−125.73	
3	−81.46	−113.58	−146.59	−177.69	
4	−101.00	−140.76	−173.65	−204.29	

Enthalpies of formation from monomers in kcal mol^{-1}.

	(m) Ammonia				
Sulfuric acid (n)	0	1	2	3	4
0	–	–	−19.52	−55.15	−81.56
1	–	−28.14	−57.92	−92.07	−121.57
2	−33.42	−71.02	−104.45	−138.44	−169.30
3	−72.19	−107.72	−143.17	−184.21	−212.56
4	−100.03	−151.24	−183.34	−**222.33**	−251.02
	(m) DMA				
Sulfuric acid (n)	1	2	3	4	
0	–	−22.52	−61.30	−89.52	
1	−31.01	−66.37	−107.02	−146.83	
2	−74.40	−**111.73**	−152.75	−191.22	
3	−112.90	−155.37	−190.01	−231.38	
4	−152.88	−194.43	−229.95	−269.13	

Entropies of formation from monomers in cal K^{-1} mol.

Table S4.2. Non-monomer evaporations.

$(H_2SO_4)_n$ $(NH_3)_m$		Evaporation rate (1/s)		
n	m	$(H_2SO_4)_1 \times (NH_3)_1$	$(H_2SO_4)_2 \times (NH_3)_1$	$(H_2SO_4)_2 \times (NH_3)_2$
2	1	1.22×10^{-2}		
3	1	3.05×10^{-4}	3.83×10^{2}	
4	1	1.21×10^{-5}	1.71×10^{2}	
1	2	4.11×10^{6}		
2	2	5.45×10^{-4}	1.83×10^{3}	
3	2	7.15×10^{-4}	7.15×10^{-4}	1.73×10^{-2}
4	2	1.54×10^{-4}	4.14	7.98×10^{-5}
1	3	1.20×10^{3}		
2	3	4.81×10^{-4}	3.27×10^{-4}	4.40×10^{6}
3	3	3.03×10^{-7}	1.12×10^{-10}	3.03×10^{-7}
4	3	$\mathbf{1.09 \times 10^{-6}}$	$\mathbf{1.25 \times 10^{-6}}$	$\mathbf{1.25 \times 10^{-6}}$
1	4	1.86×10^{-2}		
2	4	9.72×10^{-6}	1.15×10^{-9}	23.53
3	4	6.16×10^{-8}	1.43×10^{-14}	5.09×10^{-8}
4	4	4.21×10^{-4}	8.70×10^{-11}	1.95×10^{-7}
$(H_2SO_4)_n((CH_3)_2NH)_m$		Evaporation rate (1/s)		
n	m	$(H_2SO_4)_1((CH_3)_2NH)_1$	$(H_2SO_4)_2((CH_3)_2NH)_1$	$(H_2SO_4)_2((CH_3)_2NH)_2$
2	1	5.47×10^{-5}		
3	1	1.17×10^{-8}	3.71	
4	1	1.61×10^{-9}	5.74	
1	2	3.76×10^{6}		
2	2	$\mathbf{7.62 \times 10^{-8}}$	$\mathbf{9.66 \times 10^{-5}}$	
3	2	4.37×10^{-3}	4.37×10^{-3}	**3.75**
4	2	5.56×10^{-5}	3.34	$\mathbf{8.48 \times 10^{-6}}$
1	3	2.89		
2	3	2.09×10^{-10}	1.52×10^{-13}	$\mathbf{1.15 \times 10^{4}}$
3	3	$\mathbf{1.78 \times 10^{-5}}$	4.24×10^{-16}	$\mathbf{1.78 \times 10^{-5}}$
4	3	4.98×10^{-7}	$\mathbf{2.52 \times 10^{-2}}$	$\mathbf{2.52 \times 10^{-2}}$
1	4	3.11×10^{-4}		
2	4	2.81×10^{-11}	1.84×10^{-19}	$\mathbf{1.02 \times 10^{-3}}$
3	4	8.40×10^{-11}	3.82×10^{-24}	$\mathbf{1.95 \times 10^{-13}}$
4	4	2.83×10^{-4}	5.32×10^{-10}	$\mathbf{5.44 \times 10^{-2}}$

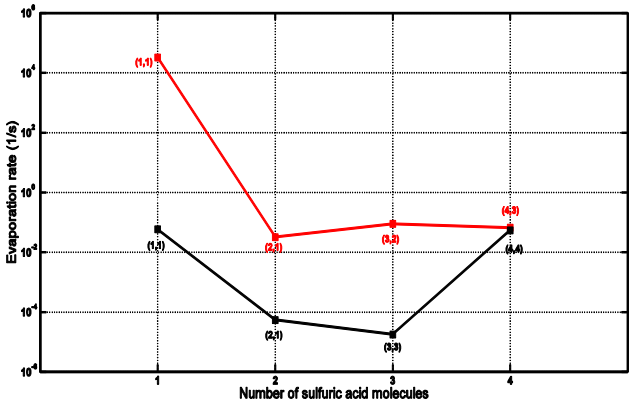

Fig. 5. Evaporation rates for the most stable clusters independent of the number of bases in it including non-monomer evaporation; composition of each cluster is indicated in parenthesis (acid : base); the black line corresponds to DMA containing clusters and the red line to ammonia containing clusters.