Impact of Quantum Chemistry Parameter Choices and Cluster Distribution Model Settings on Modeled Atmospheric Particle Formation Rates

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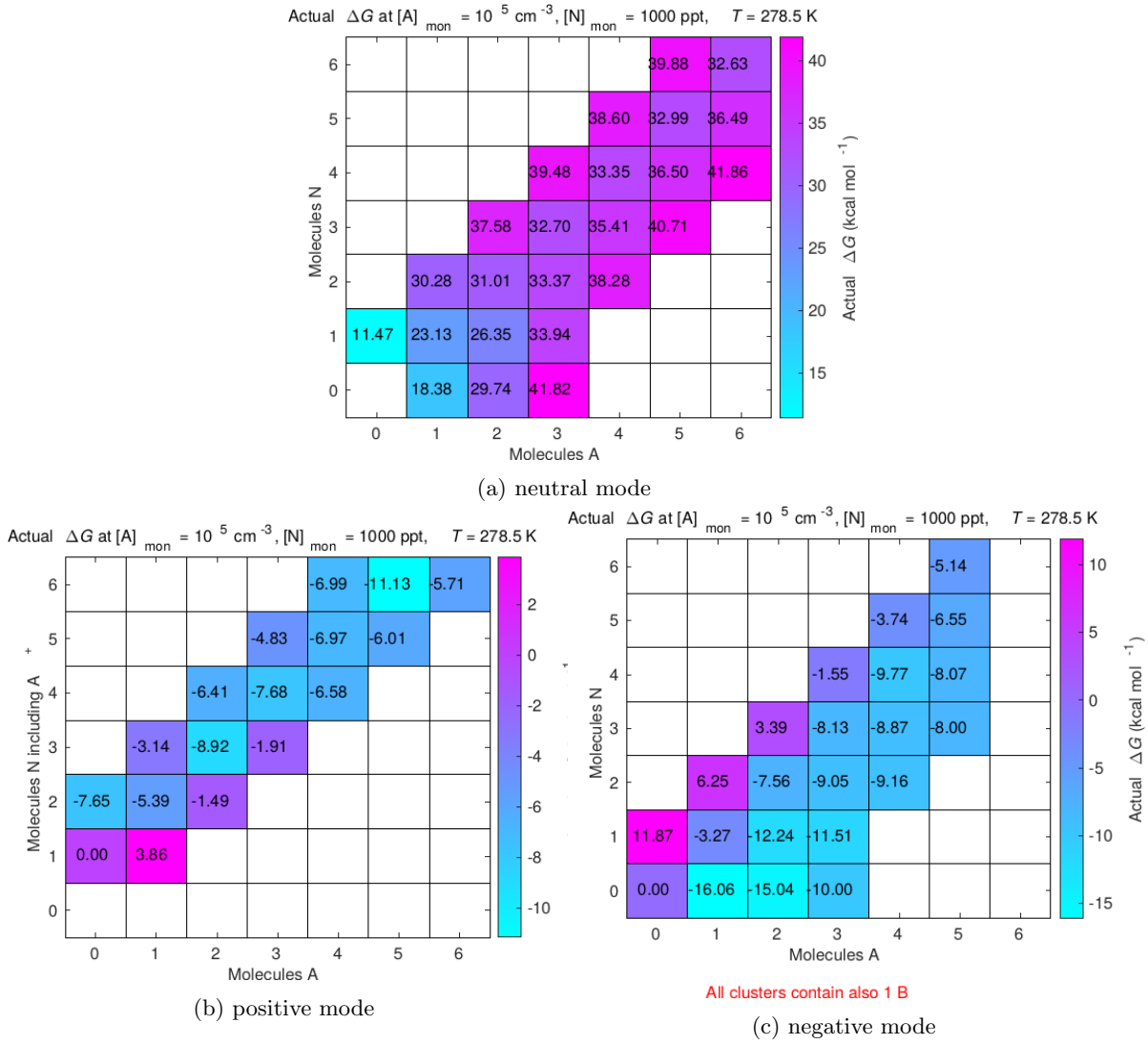
Supporting Information

The SI contains this document with two actual ΔG surfaces (*cf.* Fig. S1 and S2), the global minima that change for lower temperature (*cf.* Fig. S3) and Fig. S4 with the outgrowing cluster choice if it was solely made based on Eq. 7. Accompanying files contain a table with the DLPNO-CCSD(T) single point electronic energies, the zero-point vibrational correction, the thermal correction to the Gibbs energy, the dipoles and the polarizabilities. Additionally all Orca .out files, .xyz files and a reduced version of the Gaussian .log files are given. In filenames as well as in cluster names in the table the ending "_qh" shows that the specified cluster is only the global minimum after the application of the quasi-harmonic correction. The files are put into three folders: "neutral", "negative" and "positive". The "ACDC" folder contains the general "INPUT.inp" file and the "INPUT_ACDB.inp" prepared for the comparison with the ACDB data. The subdirectory "HS_files" contains all other files used for ACDC named as shown in Tab. S1.

Table S1: File naming for the ACDC input files.

file name	content
dipoles_YesQuasiHCorr.txt	Contains the dipoles and polarizabilities for all global minima calculated with quasi-harmonic correction.
dipoles_NoQuasiHCorr.txt	Contains the dipoles and polarizabilities for all global minima calculated without quasi-harmonic correction.
dipoles_ACDB.txt	Contains the dipoles and polarizabilities for all global minima from the ACDB.
HS_YesSymYesQuasiHCorr.txt	Enthalpies [kcal/mol] and entropies [cal/mol*K] for global minima considering monomer symmetry and quasi-harmonic correction.
HS_YesSymNoQuasiHCorr.txt	Enthalpies [kcal/mol] and entropies [cal/mol*K] for global minima considering monomer symmetry, but not quasi-harmonic correction.
HS_NoSymYesQuasiHCorr.txt	Enthalpies [kcal/mol] and entropies [cal/mol*K] for global minima considering no monomer symmetry, but considering quasi-harmonic correction.
HS_CompareToACDB.txt	Enthalpies [kcal/mol] and entropies [cal/mol*K] for global minima to be compared to ACDB.
HS_ACDB.txt	Enthalpies [kcal/mol] and entropies [cal/mol*K] for recalculated global minima taken from ACDB.
HS_YesSymYesQuasiHCorr_CLUSTERSYM_T.txt	Enthalpies [kcal/mol] and entropies [cal/mol*K] for global minima considering monomer symmetry, cluster symmetry and quasi-harmonic correction. T is the temperature times 10.

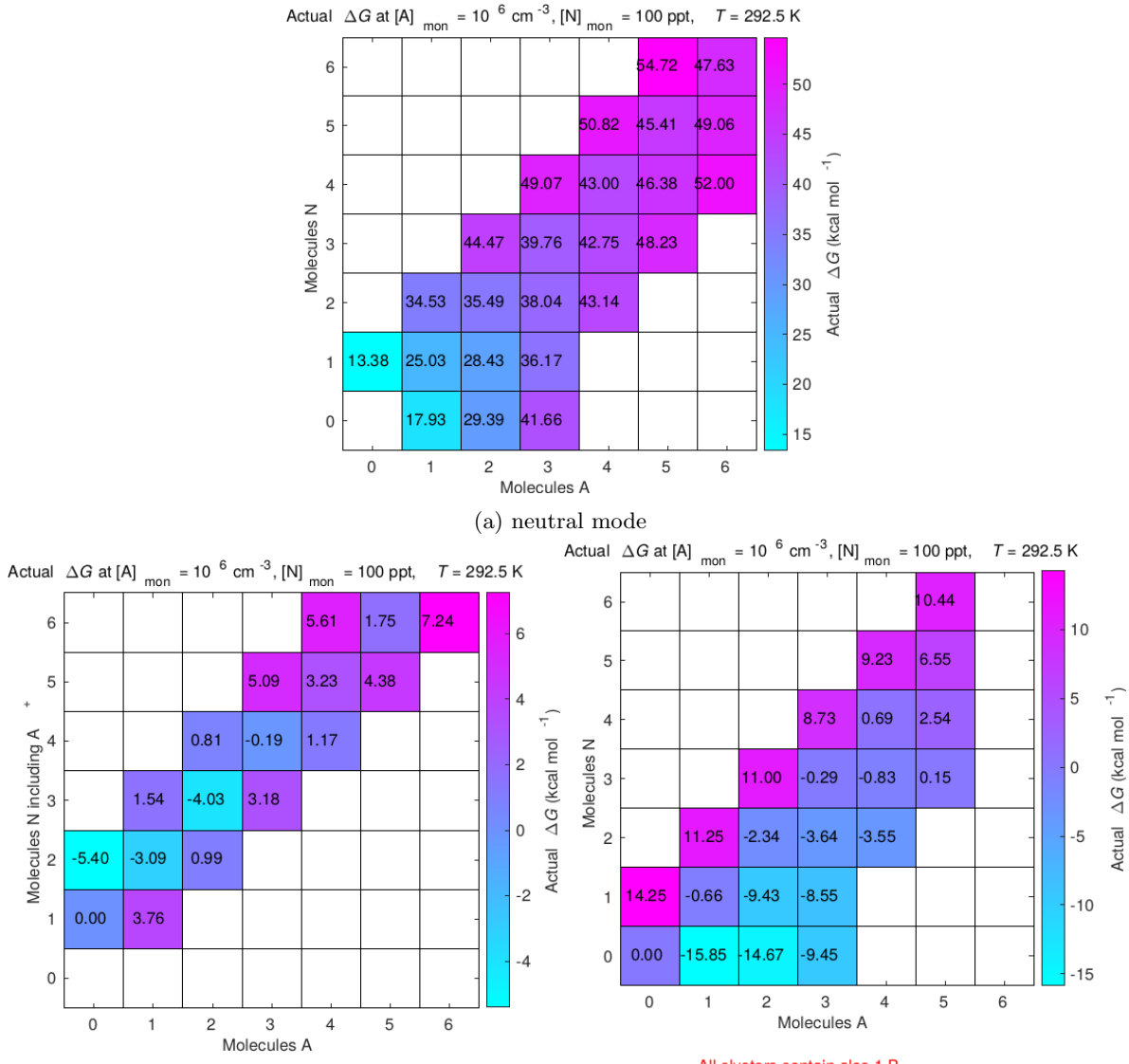


(a) neutral mode

(b) positive mode

(c) negative mode

Figure S1: A typical actual ΔG surface. $[\mathrm{A}]_{\mathrm{mon}}$ is the concentration of sulfuric acid monomers, $[\mathrm{N}]_{\mathrm{mon}}$ the concentration of ammonia monomers and B is bisulfate. Neutral mode depicts the saddle point shape. The critical cluster is the 4 sulfuric acid - 4 ammonia cluster. In charged modes most actual ΔGs are negative.

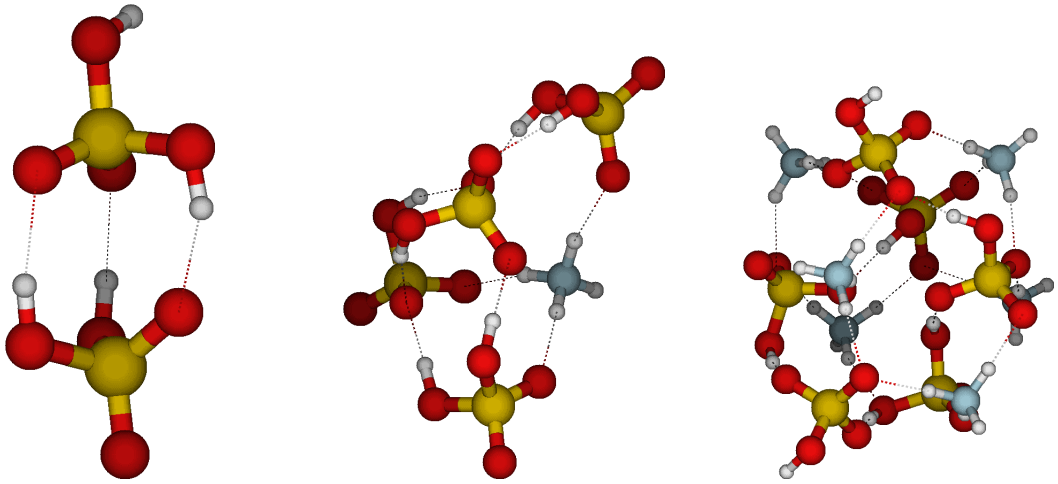


(a) neutral mode

(b) positive mode

(c) negative mode

Figure S2: The actual ΔG surface for high temperature and low monomer concentrations. $[\mathrm{A}]_{\mathrm{mon}}$ is the concentration of sulfuric acid monomers, $[\mathrm{N}]_{\mathrm{mon}}$ the concentration of ammonia monomers and B is bisulfate. In neutral mode the actual ΔG continuously rises, there is no critical cluster. Even in charged mode actual ΔGs rise and even reach positive values.



(a) (b) (c)

Figure S3: The three molecular clusters that are the actual global minimum once temperature is 248.4 K or lower. The energy differences relative to the used cluster structures at 248.4 K are -0.02 kcal/mol for cluster a, $(H_2SO_4)_2$, -0.06 kcal/mol for cluster b,$(H_2SO_4)_3H_2SO_4^-(NH_3)$, and, -0.05 kcal/mol for molecule c, $(H_2SO_4)_6(NH_3)_5NH_4^+$.

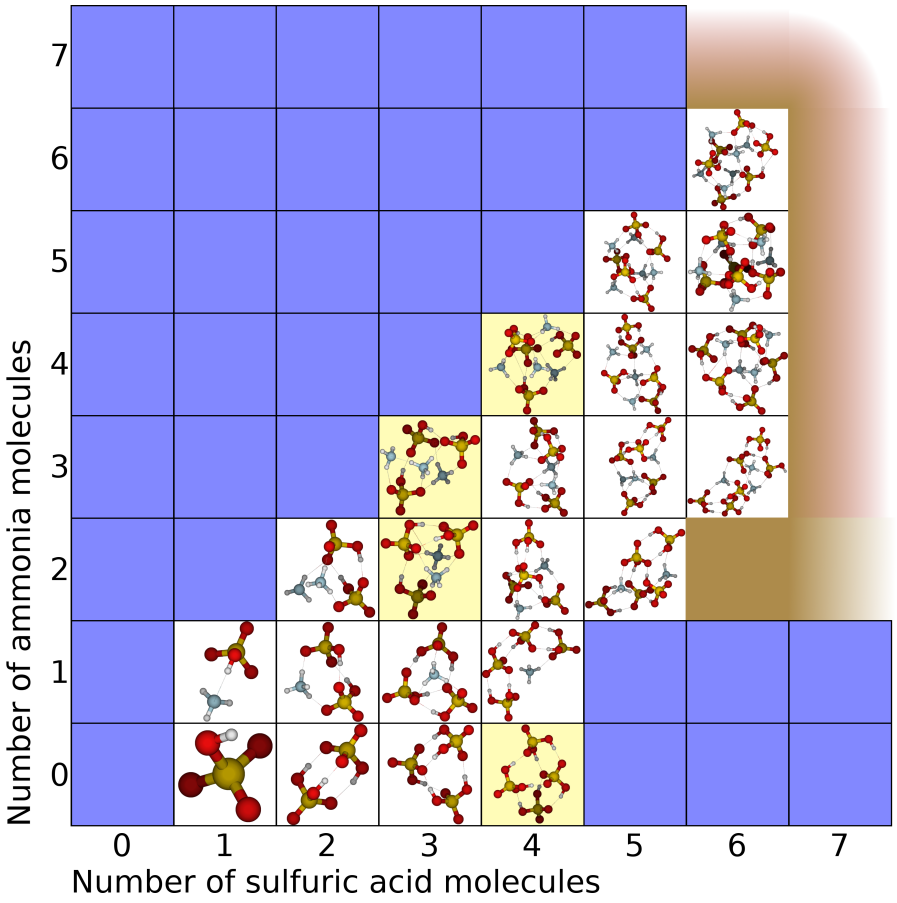


Figure S4: The outgrowing area (brown) of the negative mode clusters if the "pocket" was considered. Considering it lead to a maximum increase of 2.3% at T = 223 K, [ammonia] = 10 ppt and [sulfuric acid] = $1.6{\cdot}10^6$ cm^{-3} and 0.1% at T = 292 K, [ammonia] = 100 ppt and [sulfuric acid] = $4.0{\cdot}10^8$ cm^{-3}. For most sulfuric acid concentrations the change is well below 0.01%.