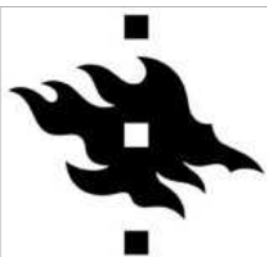


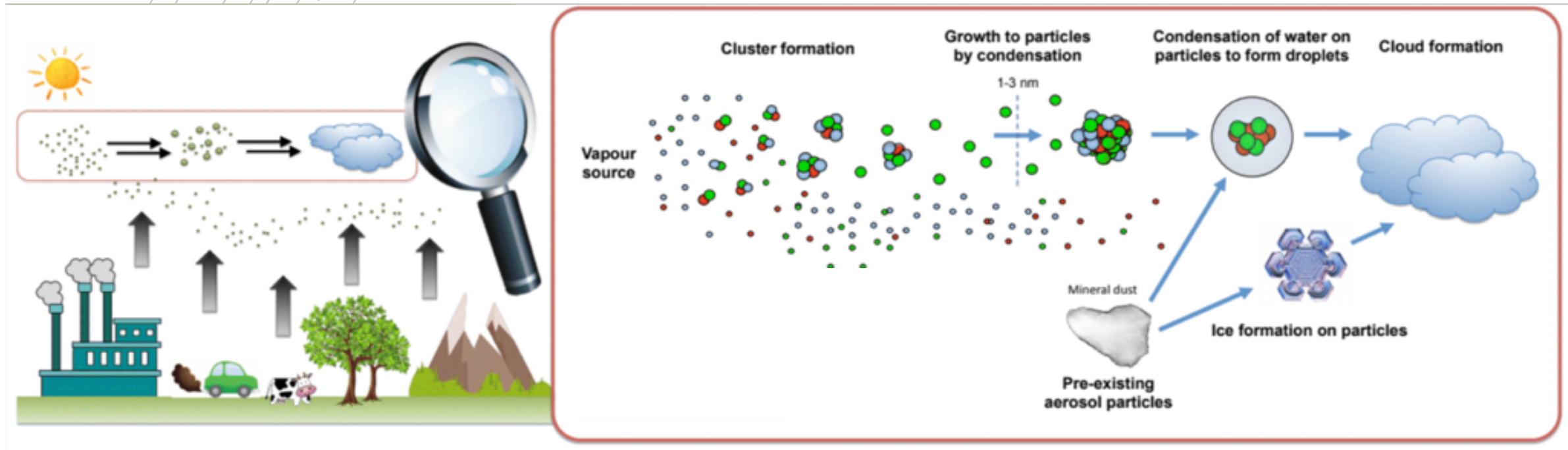
Molecular clusters: configurational sampling and global minimum search

J. Kubečka, V. Besel, T. Kurtén, H. Vehkamäki

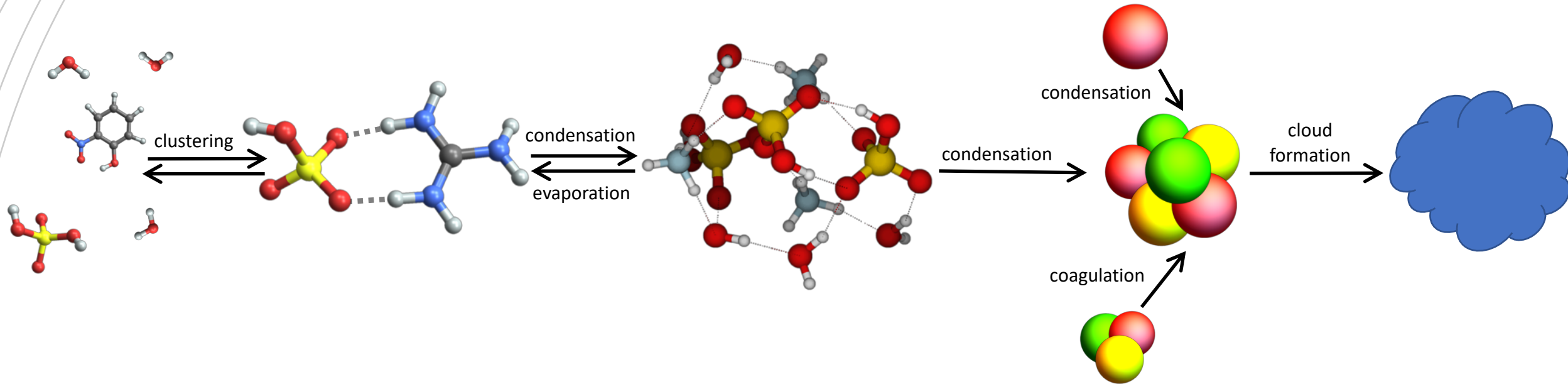


Jakub Kubečka

What we do?

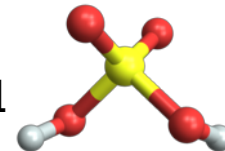


Intro & Motivation

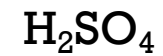


Content: - new-particle formation

- application: clusters of sulphuric acid



& base

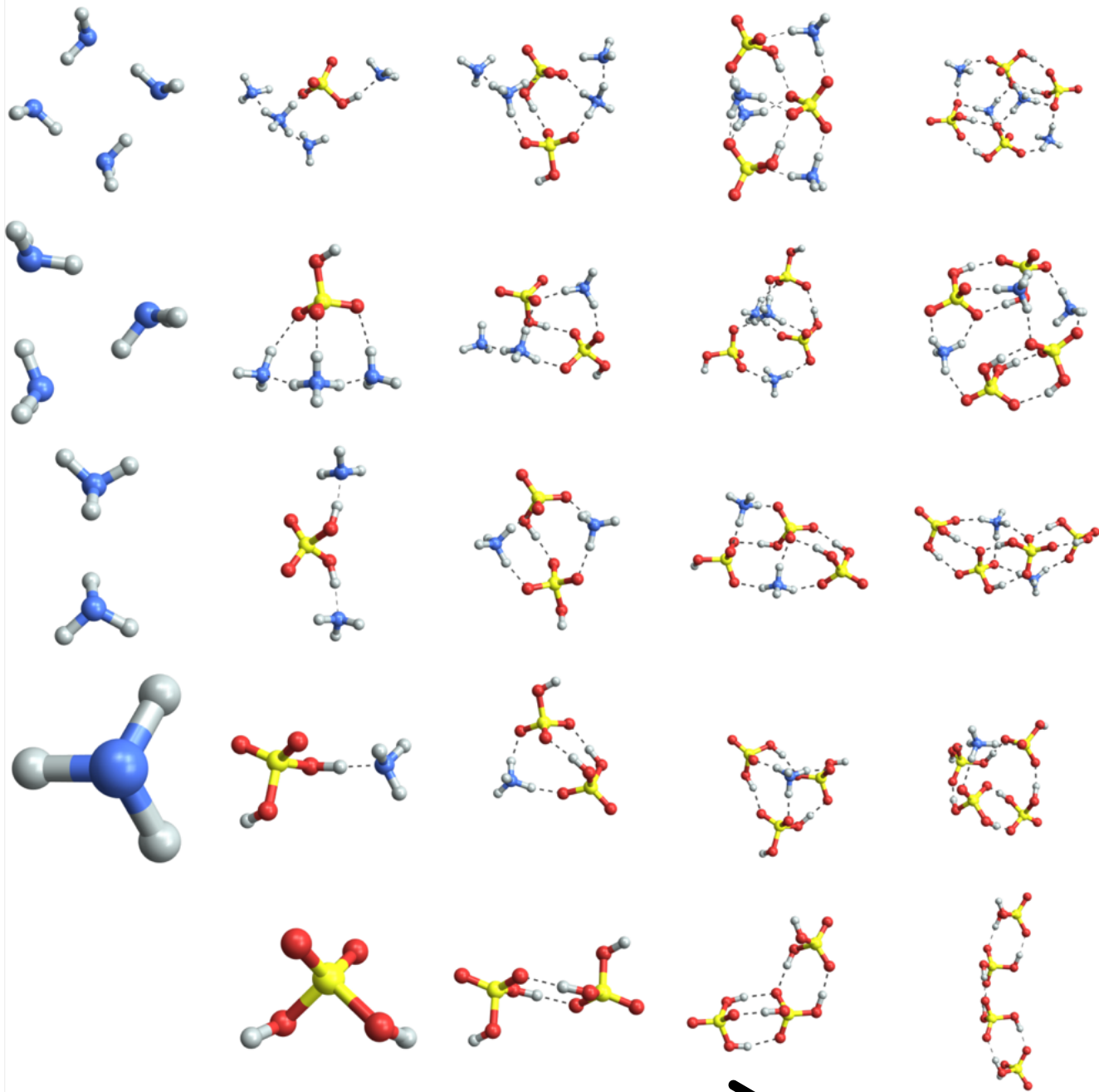


- configurational sampling

Example of clusters:

***sulphuric acid
and
ammonia***

ammonia



sulphuric acid

Model:

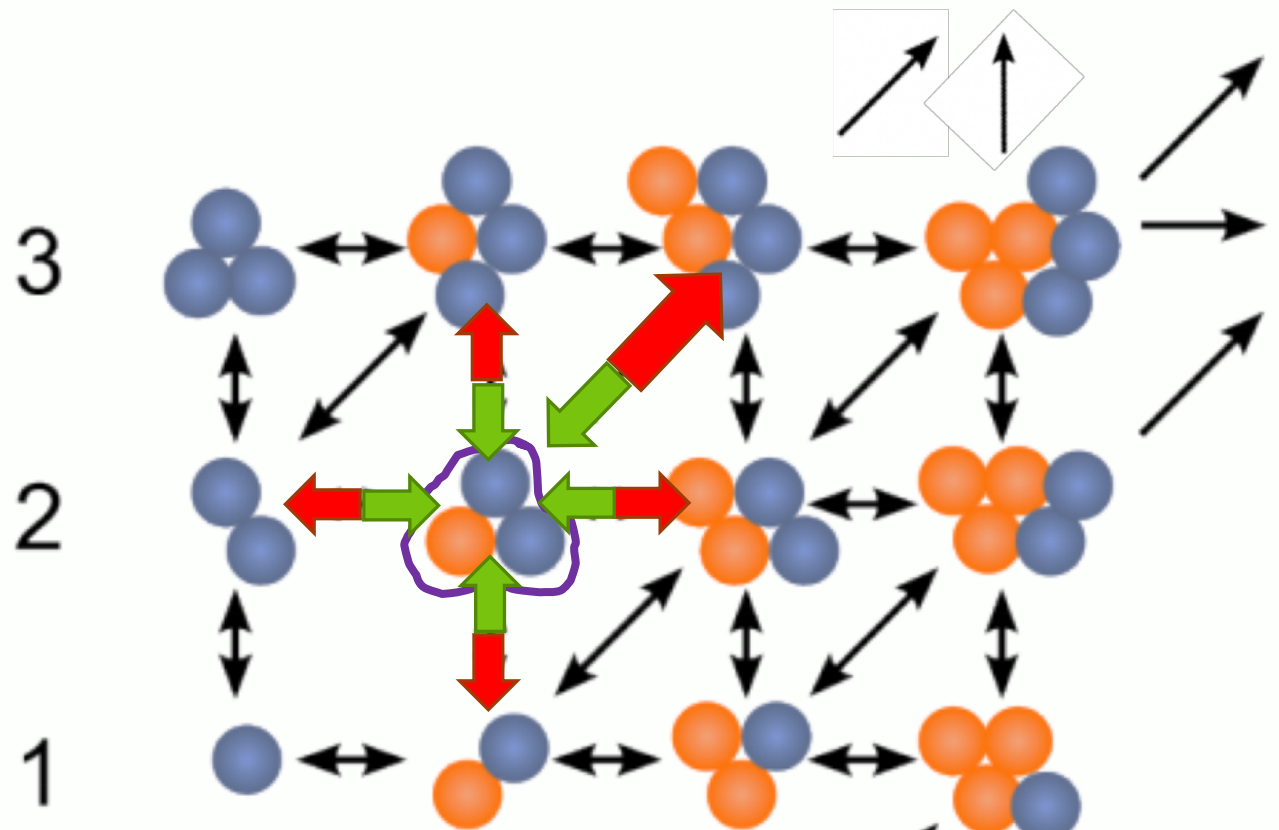
conc. change
growth (in)

growth (out)

evaporation (in)

evaporation (out)

$$\begin{aligned} \dot{c}_{21} = & +\beta_{20,01}c_{20}c_{01} \\ & +\beta_{11,10}c_{11}c_{10} \\ & -\beta_{21,10}c_{21}c_{10} \\ & -\beta_{21,11}c_{21}c_{11} \\ & -\beta_{21,01}c_{21}c_{01} \\ & +\gamma_{31\rightarrow 21}c_{31} \\ & +\gamma_{32\rightarrow 21}c_{32} \\ & +\gamma_{22\rightarrow 21}c_{22} \\ & -\gamma_{21\rightarrow 20}c_{21} \\ & -\gamma_{21\rightarrow 11}c_{21} \end{aligned}$$



Atmospheric Cluster Dynamics Code: a flexible method for solution of the birth-death equations

M. J. McGrath^{1,2}, T. Olenius¹, I. K. Ortega¹, V. Loukonen¹, P. Paasonen¹, T. Kurtén¹, M. Kulmala¹, and H. Vehkamäki¹

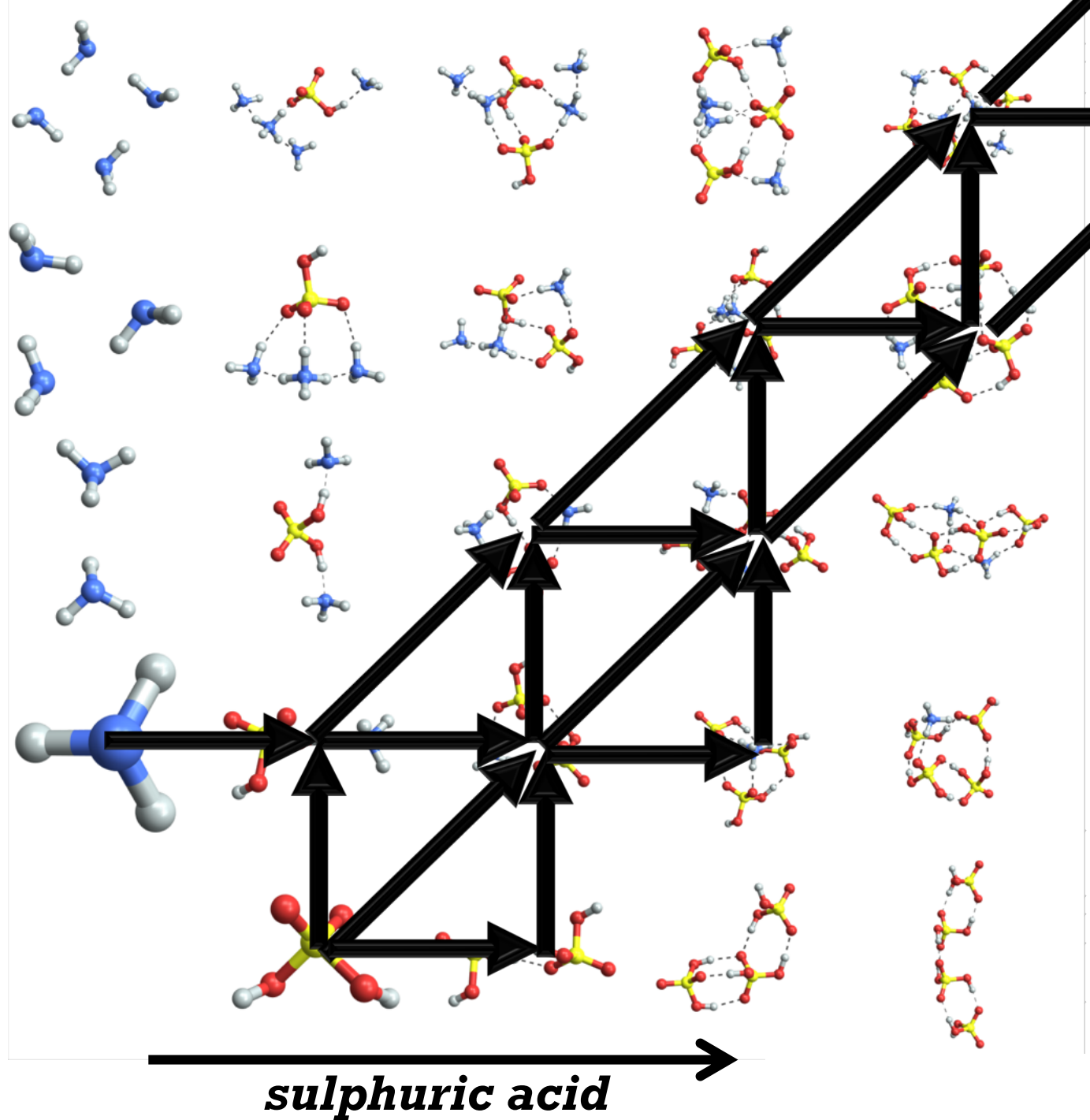
¹Department of Physics, P.O. Box 64, University of Helsinki, 00014, Helsinki, Finland

²Department of Biophysics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan



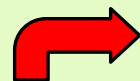
***Example of clusters:
sulphuric acid
and
ammonia***

ammonia




Configurational sampling

Potential Energy Surfaces (PES)

 GLOBAL MINIMUM




Quantum Mechanics (QM)

 Schrödinger equation

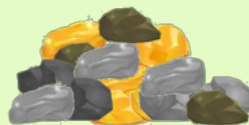
WFT or DFT



Semi-empirical

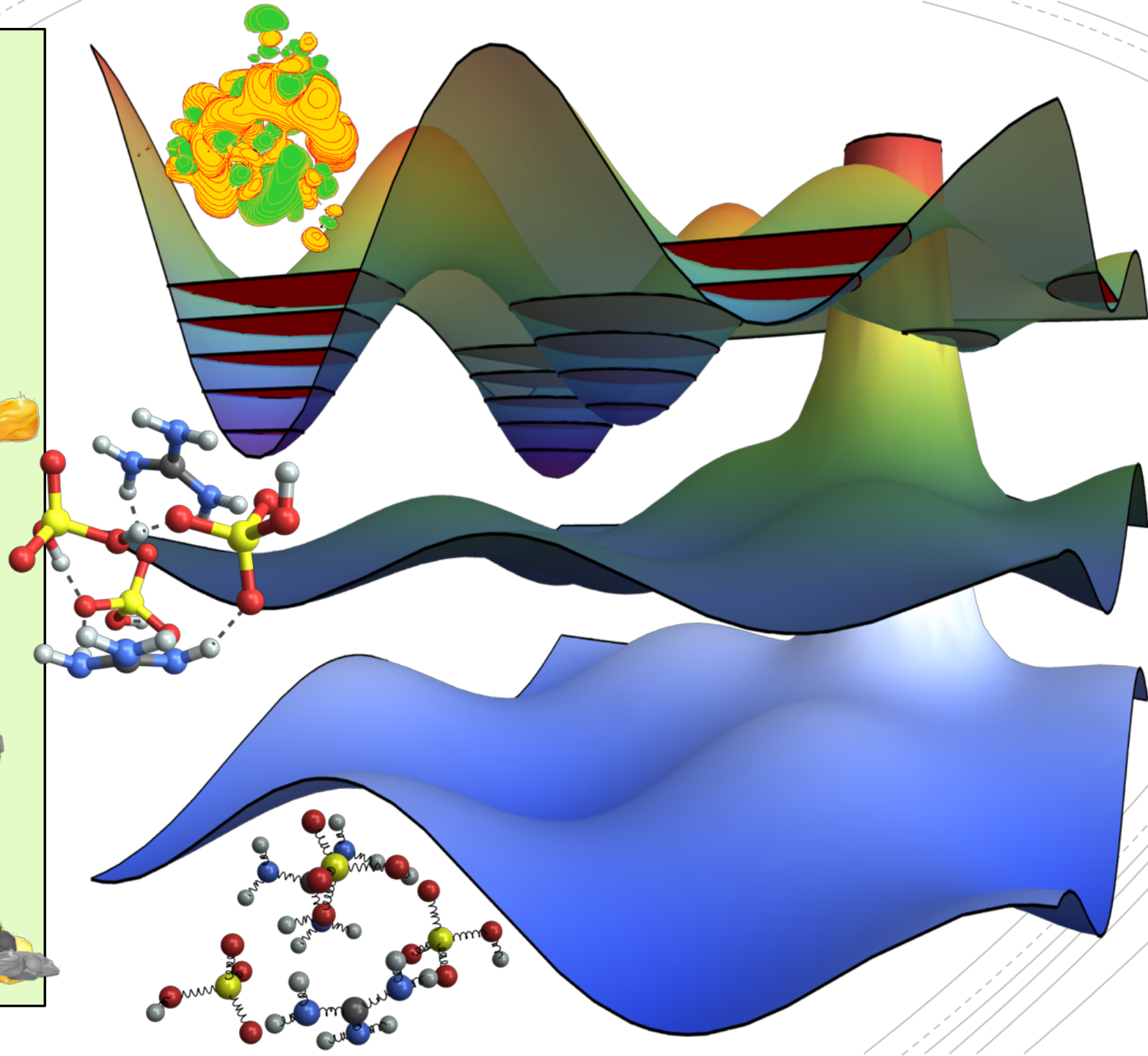
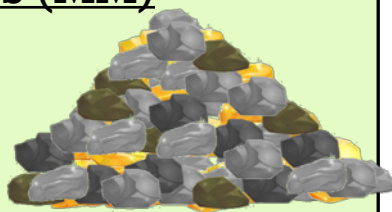
 Tight-binding (TB) methods,
semi-empirical methods

E.g.: PM7 or GNF-*x*TB



Molecular Mechanics (MM)

Force fields (FF)



Molecular Mechanics (MM)

- Force Field (FF) description

- Lennard-Jones interactions
- Coulomb interactions

- RIGID MOLECULES

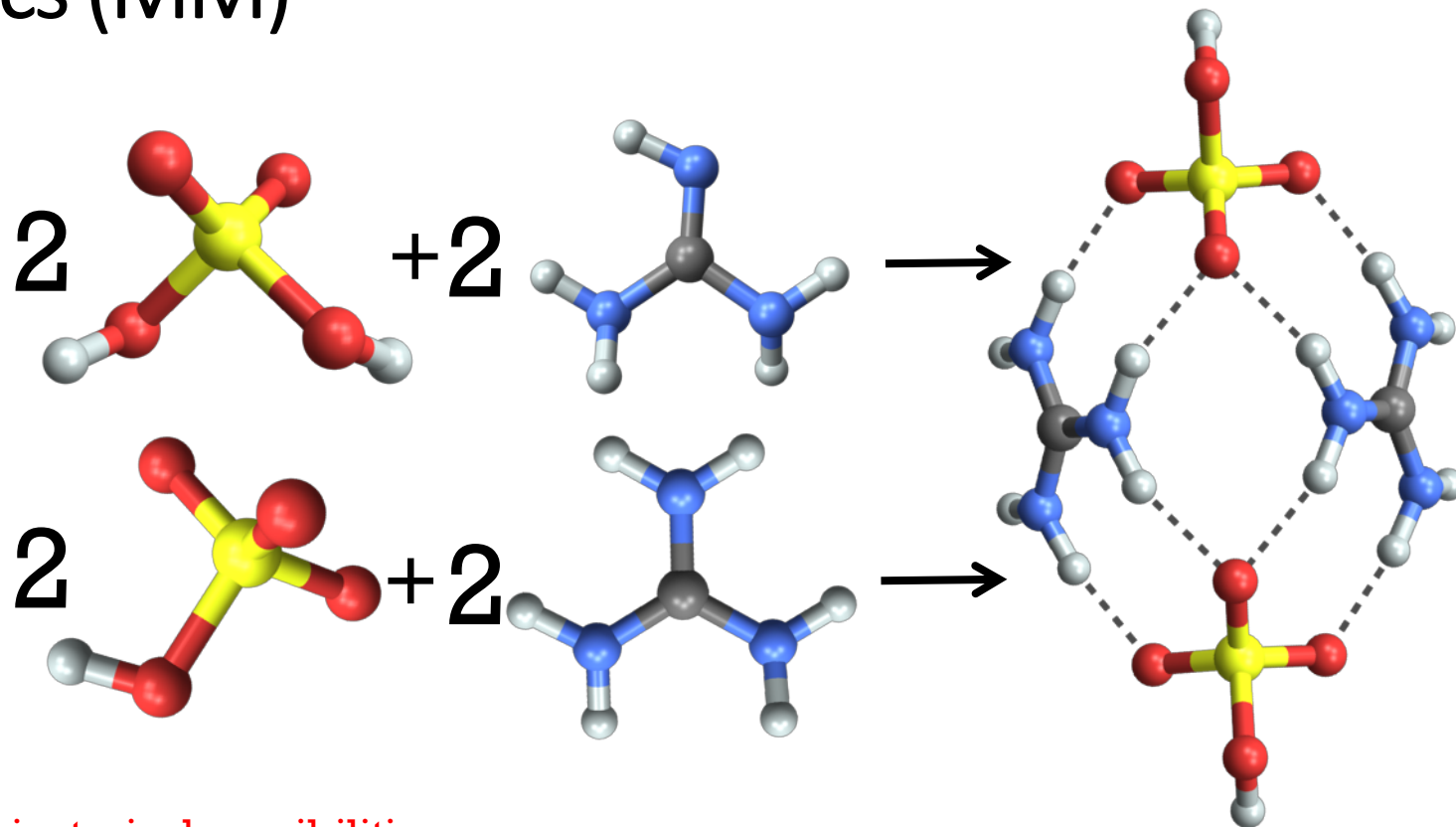
- no bond formation/breaking

- Protonation states

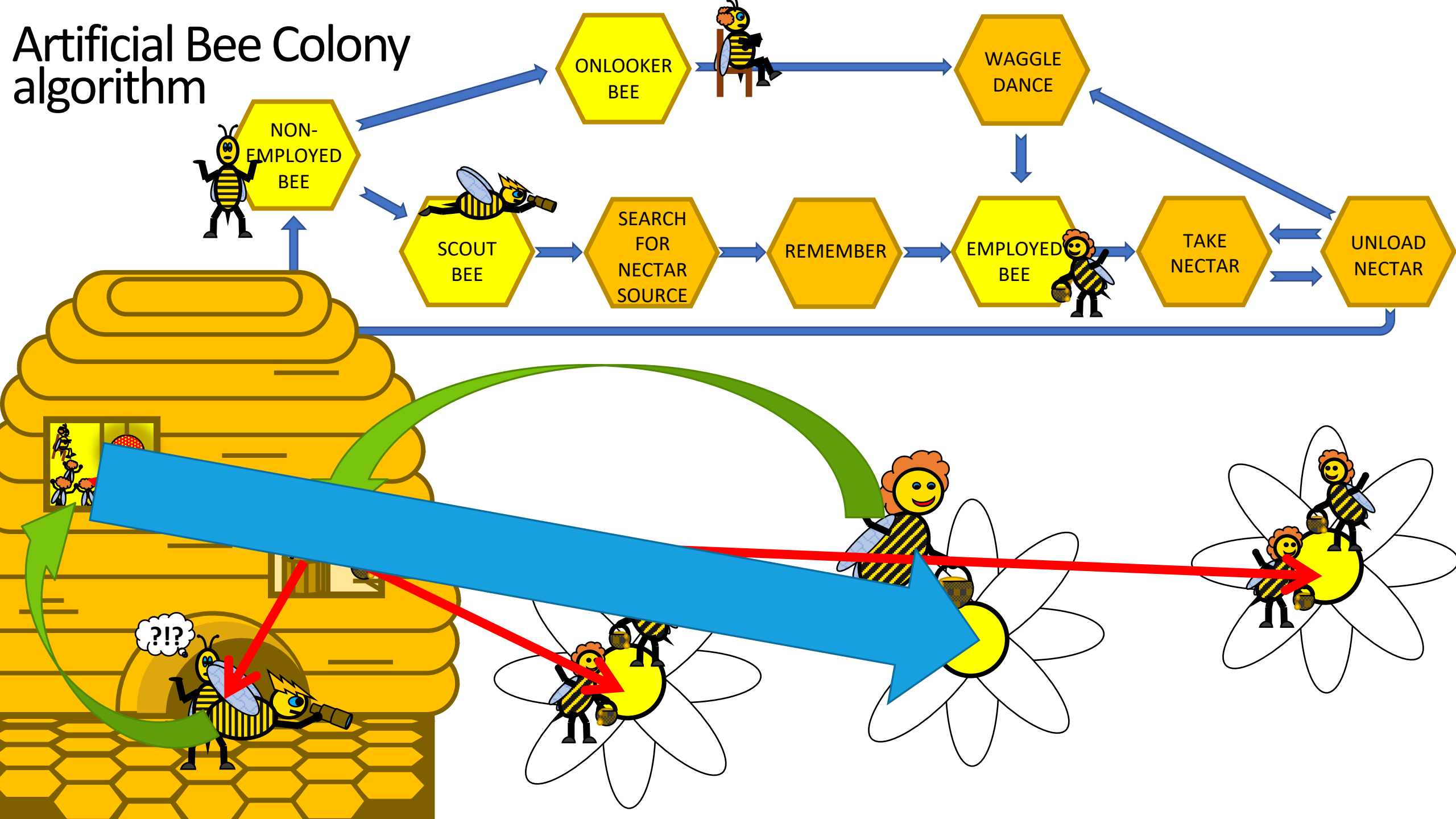
- **Configurational search:**

- sampling on grid // too much combinatorial possibilities
- Molecular Dynamics (MD) or Monte Carlo simulations // getting stuck in some minima
- Simulated annealing techniques for long time
- ...
- Genetic algorithms → Artificial Bee Colony (ABC) algorithm [Karaboga *et al.* (2008)]

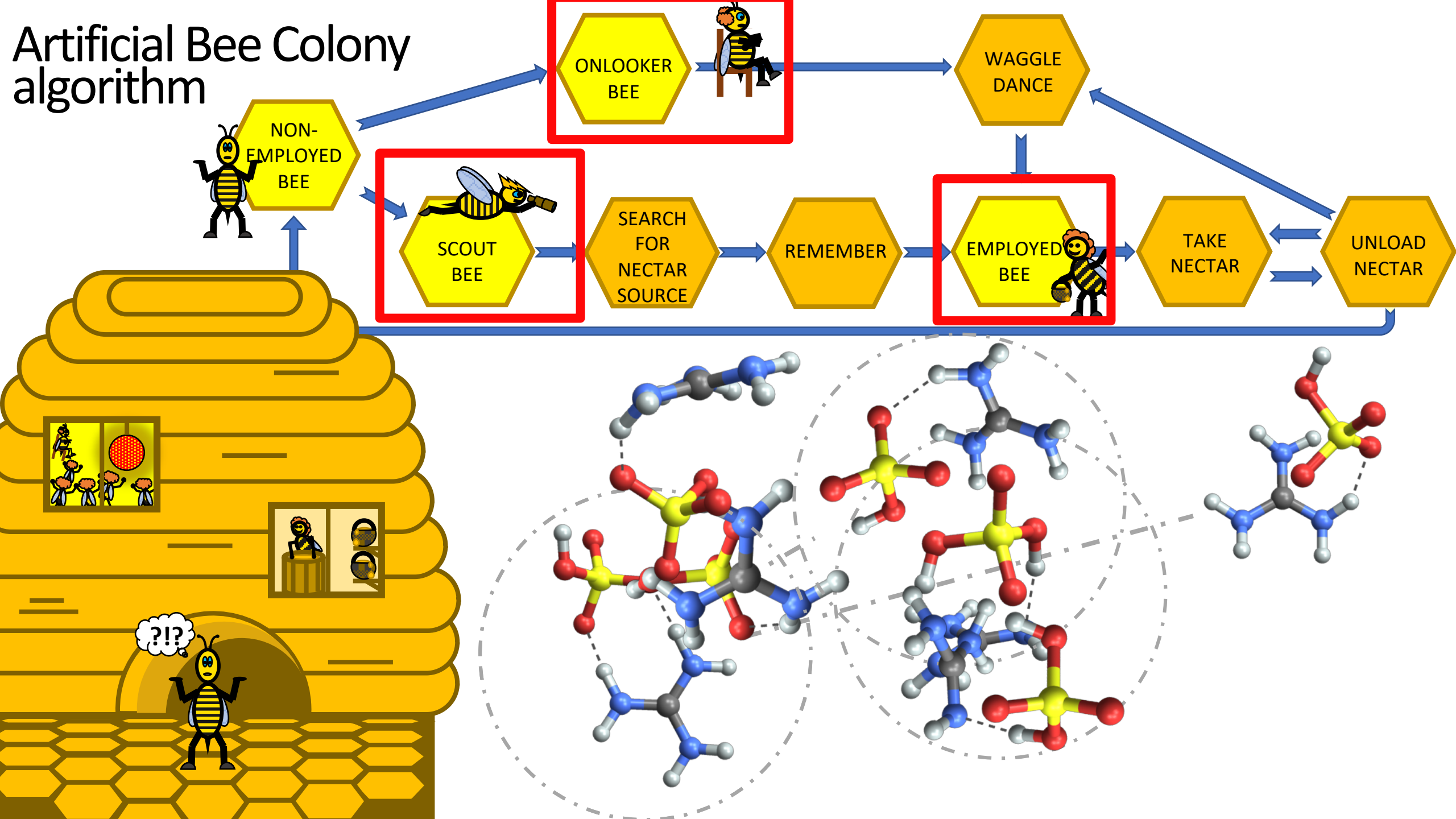
- **ABCluster** (program) [Zhang *et al.* (2015), Zhang *et al.* 2016)]



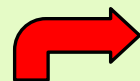
Artificial Bee Colony algorithm



Artificial Bee Colony algorithm




Potential Energy Surfaces (PES)

 GLOBAL MINIMUM




Quantum Mechanics (QM)

 Schrödinger equation

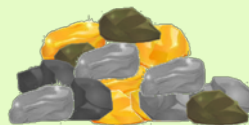
WFT or DFT



Semi-empirical

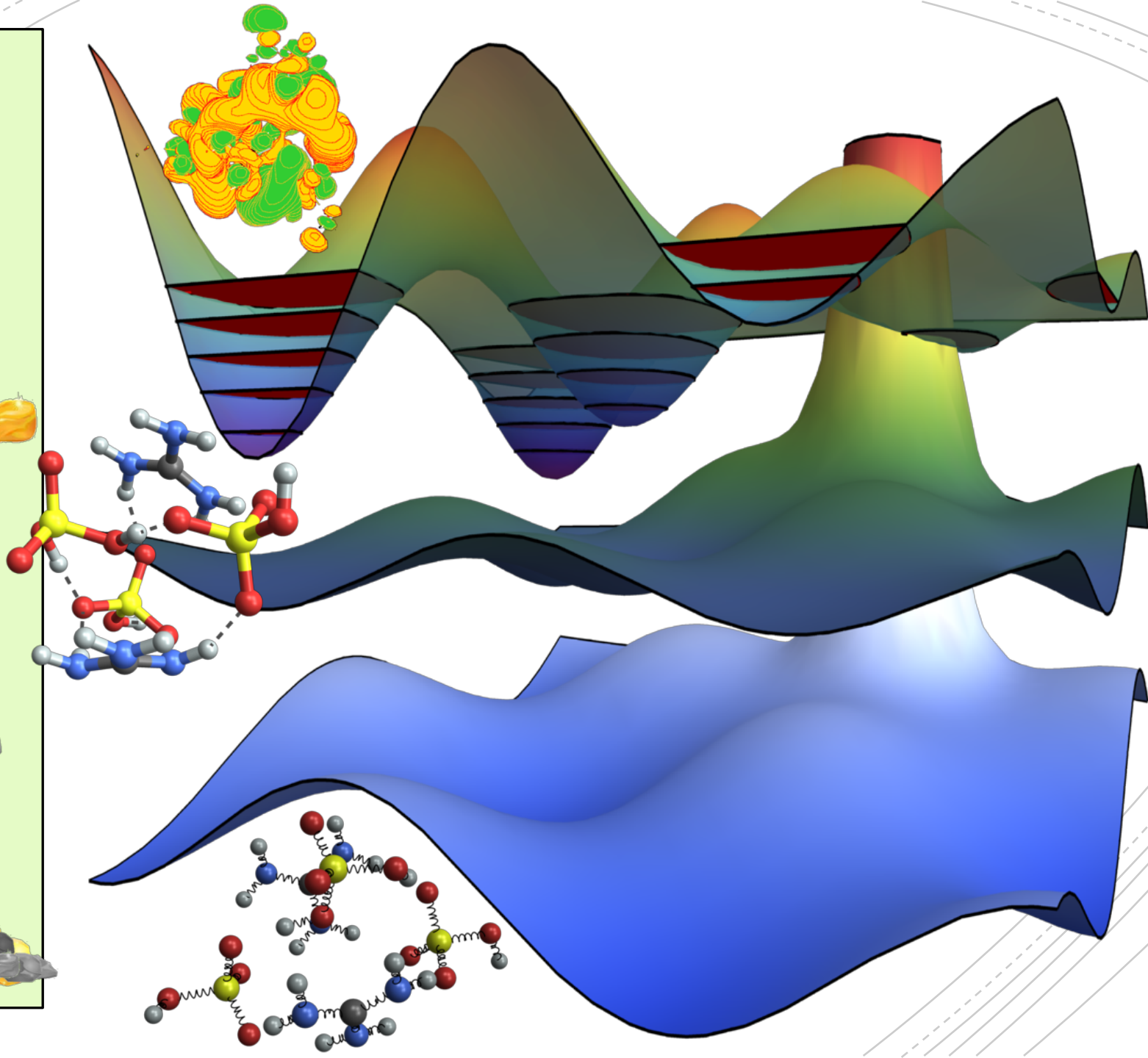
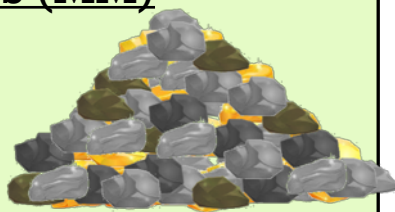
 Tight-binding (TB) methods,
semi-empirical methods

E.g.: PM7 or GNF-*x*TB



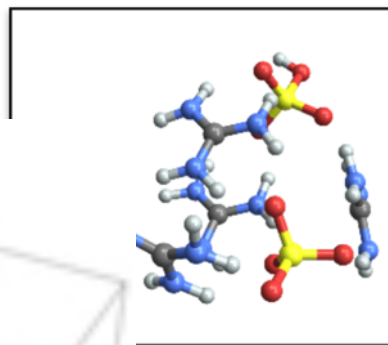
Molecular Mechanics (MM)

Force fields (FF)

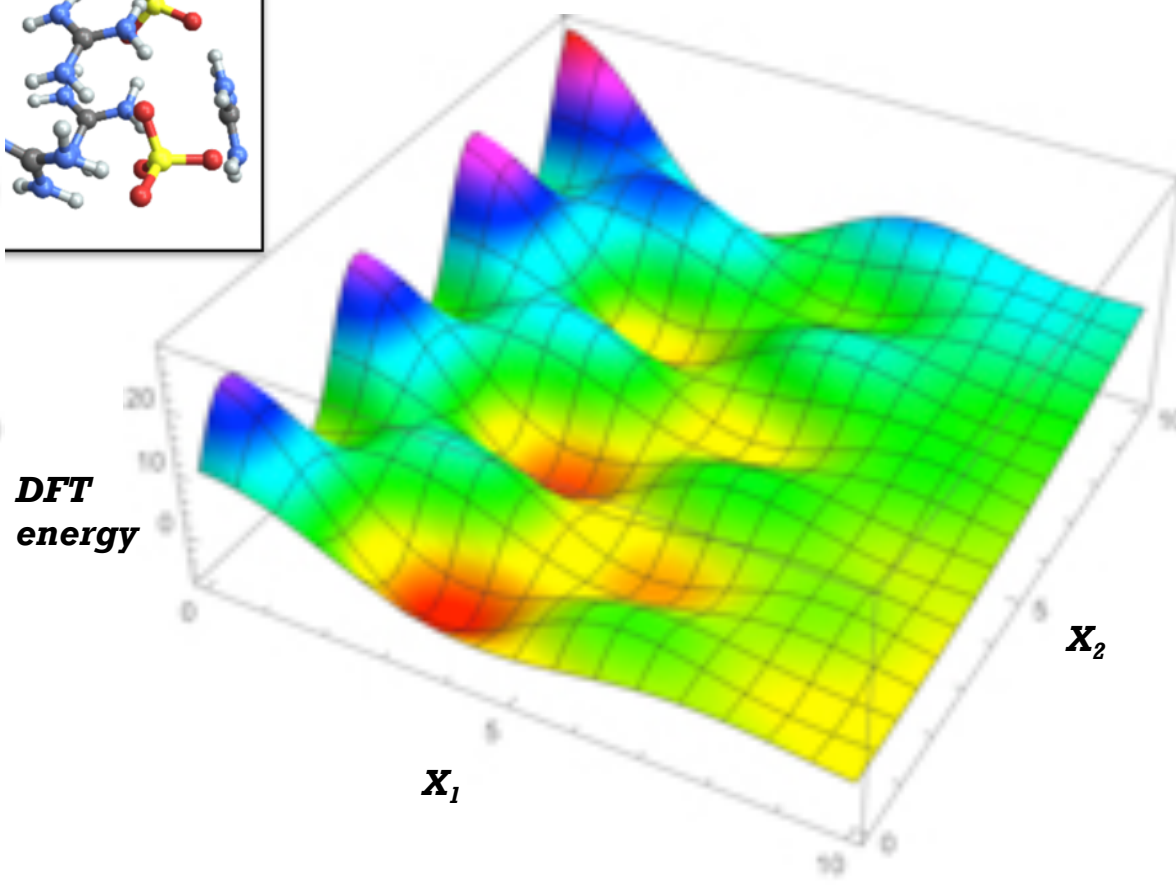
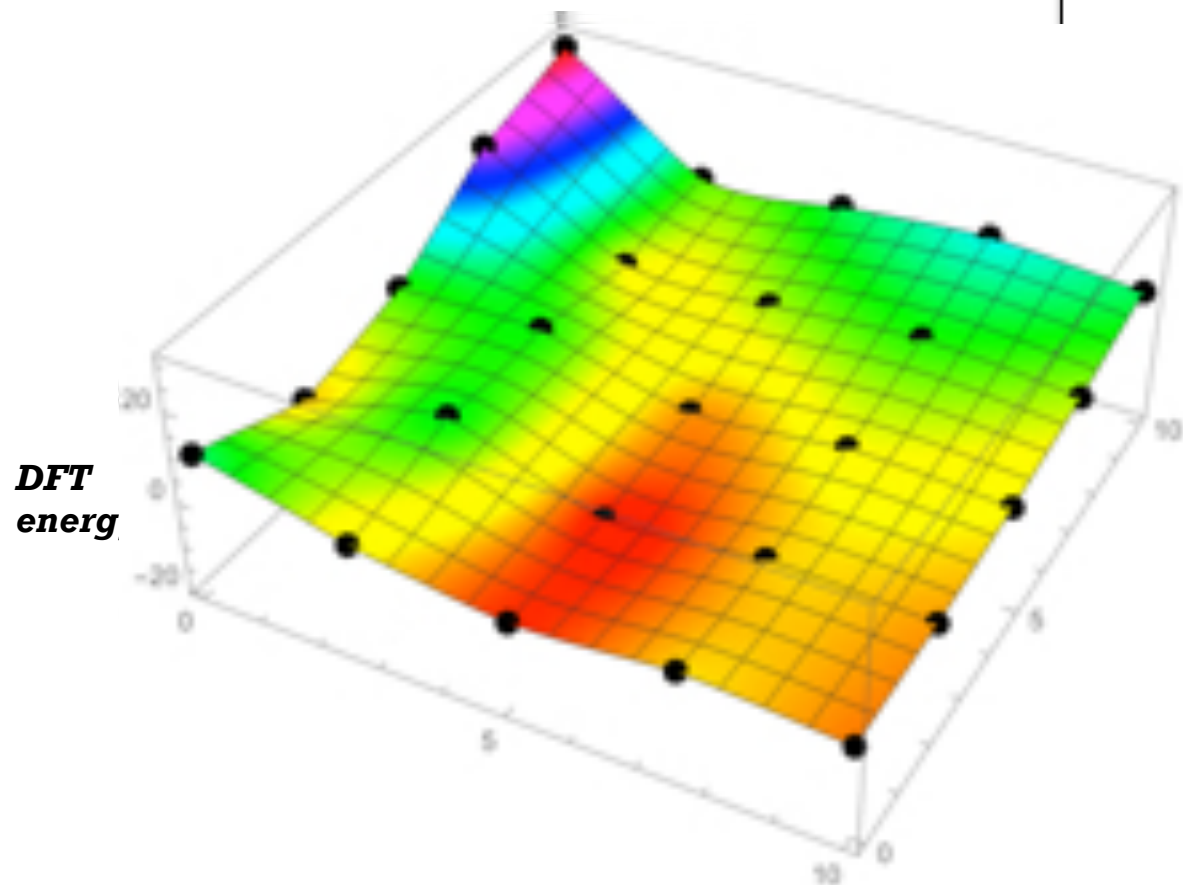


Towards Quantum Chemistry

- PCA→ $[X_1, X_2]$: semi-empirical energy, gyration radius, dipole moment, H-bond count ...



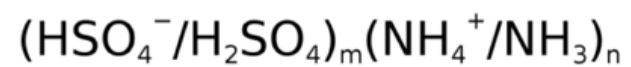
The real “surface” of minima
(which we do not know)



85 CALCULATIONS
CALCULATIONS

Applications

H_2SO_4 -ammonia clusters



$$m = n - 1$$

$$m = n$$

$$m = n - 1$$

$$m = n$$

$$m = n + 1$$

$$m = n$$

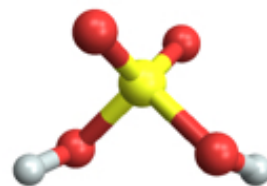
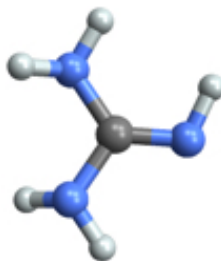
$$m = n + 1$$

positive

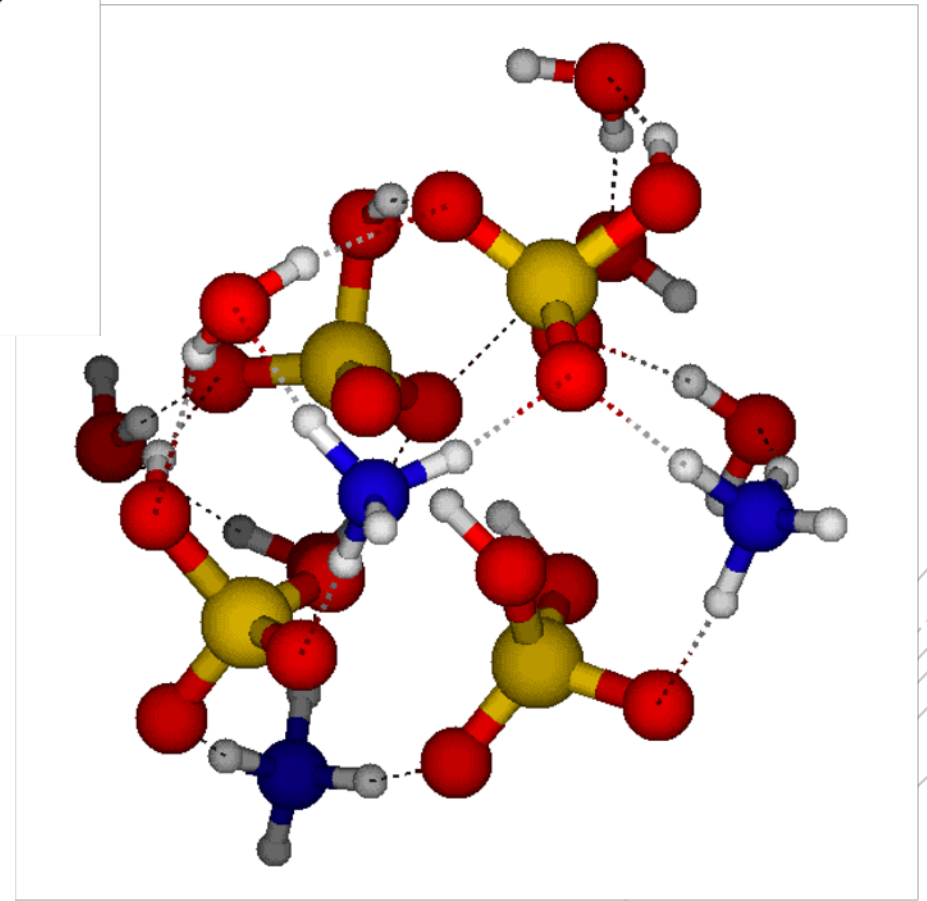
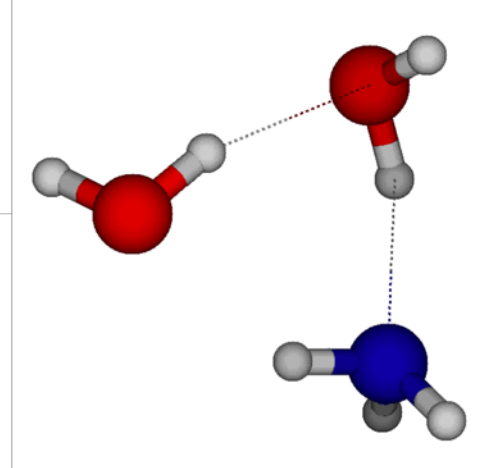
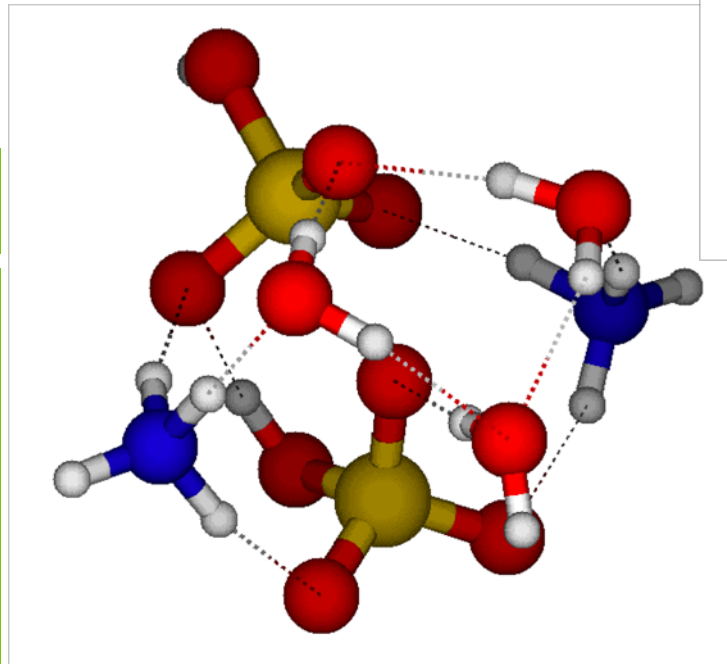
neutral

negative

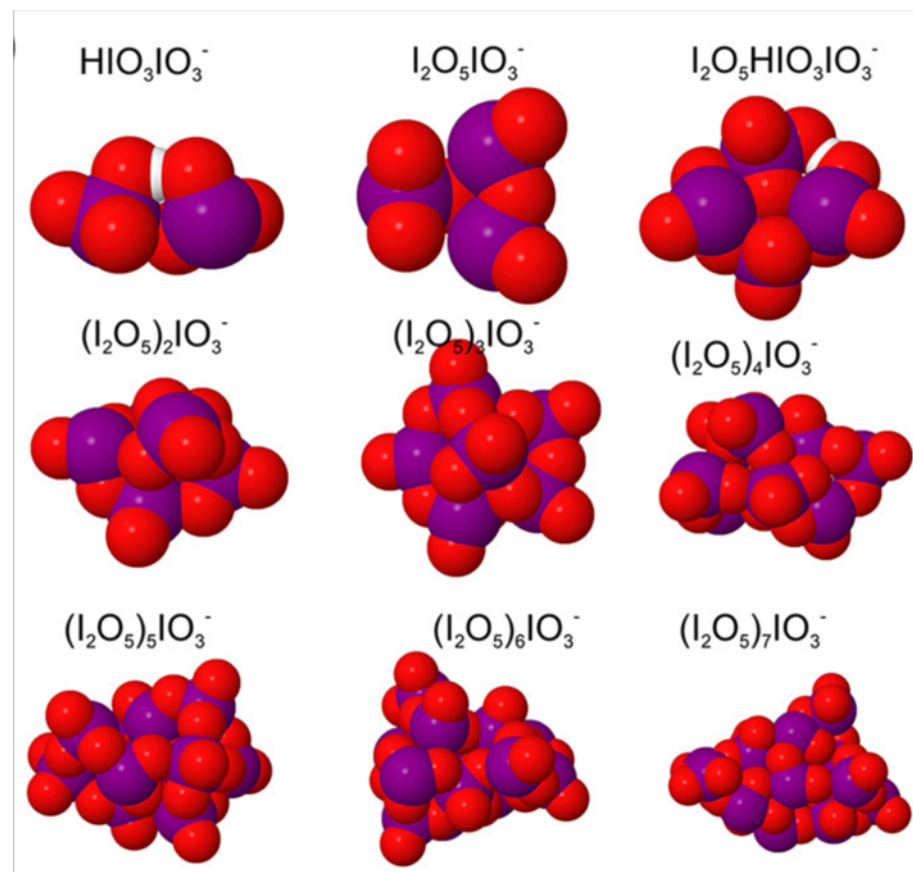
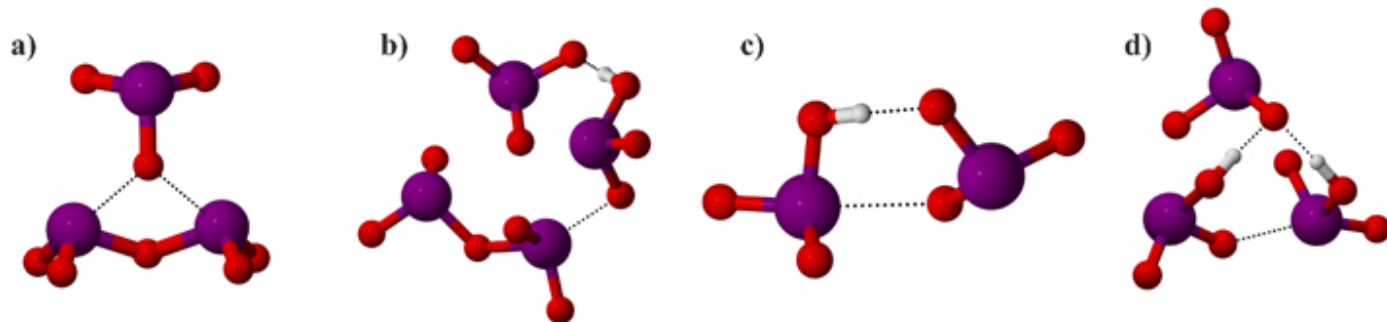
H_2SO_4 -guanidine clusters



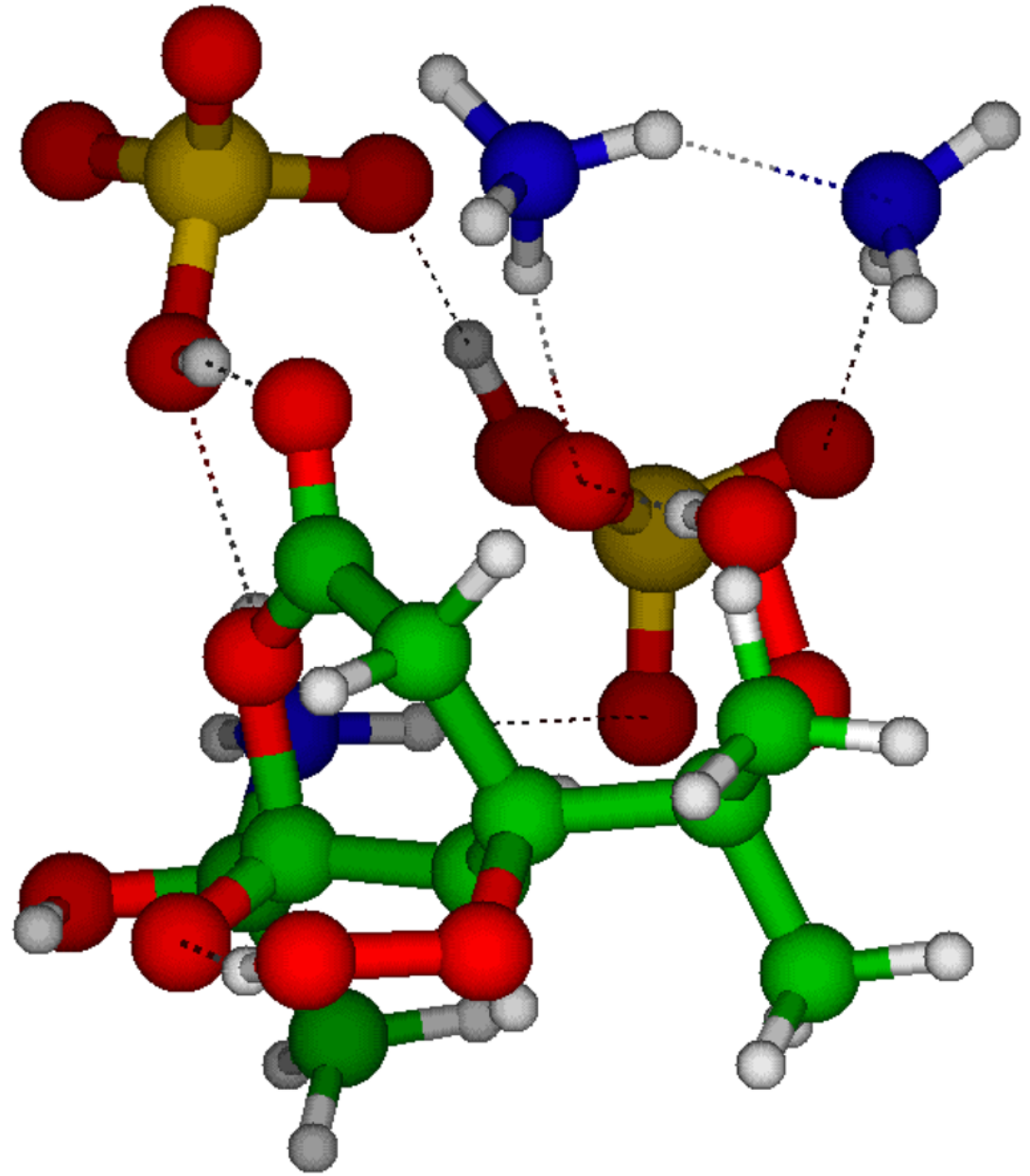
Hydrated clusters

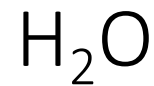
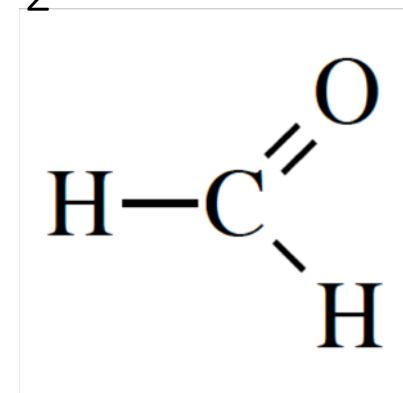
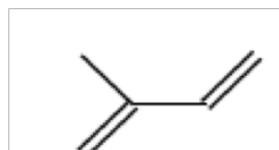
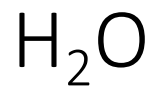
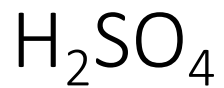
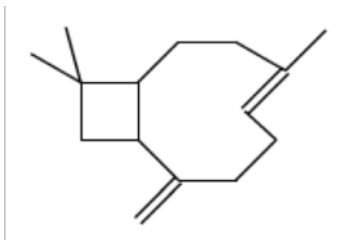


Iodine clusters

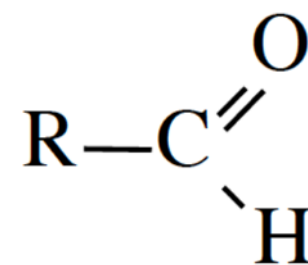
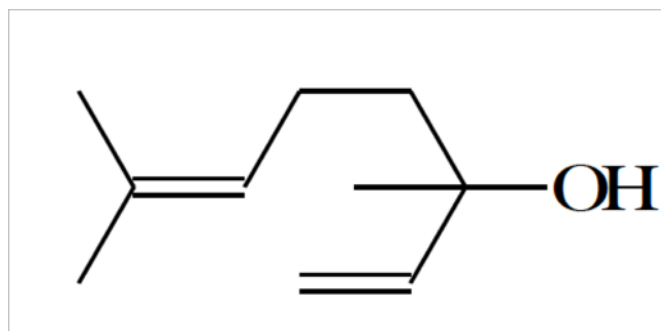
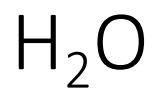
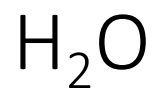
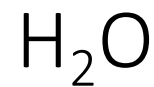
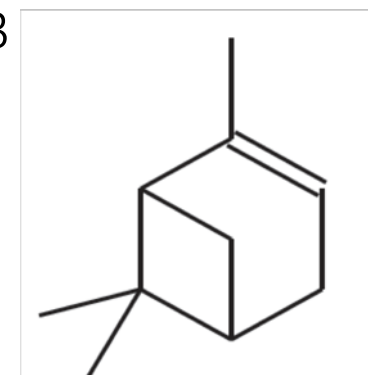
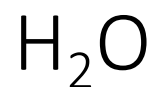
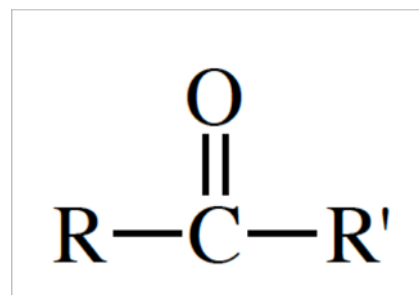
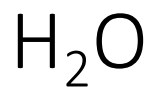


HOM
(highly oxygenated molecules)
clusters



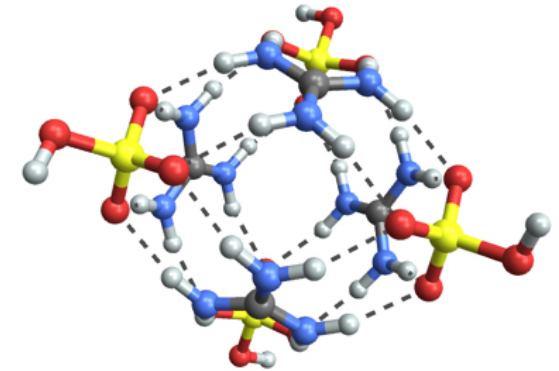


MACHINE LEARNING



Summary & Conclusion & Outlook

- Atmospheric nucleation!
- Configurational sampling of molecular clusters.



LOW LEVEL OF THEORY -> MEDIUM LEVEL OF THEORY -> SELECTION -> HIGH LEVEL OF THEORY
(genetic algorithm) (PCA, Gauss.reg.)





**Thank you
for your
attention.**



**SIMU
GROUP**

