

Growth of ice nanoparticles via uptake of individual molecules: pickup cross sections

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Synopsis We present cross sections for pickup of several atmospherically relevant molecules on ice nanoparticles with the 0.5-3 nm diameter range. The experimental values are supported by molecular dynamics simulations and analytical calculations based on long-range cluster-molecule potentials. The cross sections are all considerably larger than the geometrical cross section of nanoparticle and vary significantly for different guest molecules.

A recent investigation of atmospheric aerosol nucleation [1] has shown that the key steps for the aerosol formation occur in the sub-2-nanometer size range and that the nucleation-driving species are neutral clusters with small admixtures of organic compounds. We have investigated the growth process of such clusters in a laboratory experiment where the cluster beam with well-defined size distributions passes through a pickup cell filled with a gas of guest molecules. The quantity extracted from the experiment is the pickup cross section. It is deduced from the velocity decrease of the cluster beam due to momentum transfer during the pickup events. We have previously shown that this method provides reliable pickup cross sections for argon clusters [2], in contrary to other commonly used method for their determination (Poisson distribution of pickup fragments).

The present experiments focus on water clusters (ice nanoparticles) and pickup of atmospherically relevant molecules (H₂O, methane, NO_x, VOCs) on them at the COM collision energies up to 0.5 eV. The experimental cross sections vary considerably for individual guest molecules and are all significantly larger than the geometrical cross section of the nanoparticle [3]. For the pickup of water molecules, the experimental results were supported by the molecular dynamics (MD) simulations using the TIP3P water model. The MD cross section was obtained by simulating trajectories with varying impact parameters of the guest molecule. The MD results agree very well with the experimental data.

We have additionally developed analytical approach for calculating the cross section (by approximating the cluster by a sphere of evenly distributed van der Waals centers). The analytical model also agrees very well with the experimental

results (figure 1). This model was additionally used to calculate the size- and velocity dependence of the pickup cross sections - informations that are necessary in modeling the formation of atmospheric nanoparticles.

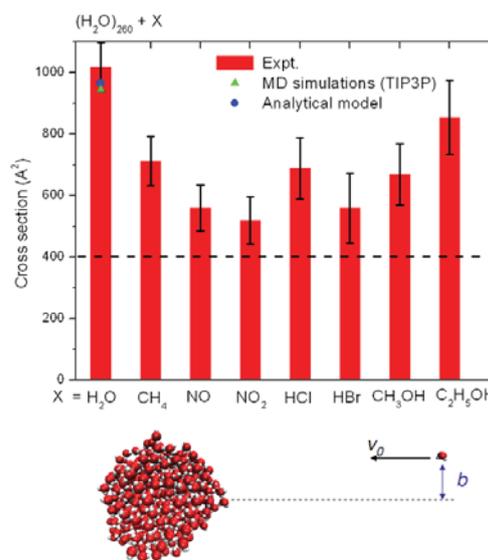


Figure 1. Top: Cross sections for pickup of various molecules on water clusters with the mean size of 260. The horizontal dashed line indicates the geometrical cross section of such clusters. **Bottom:** Illustration of cross section calculation with molecular dynamics simulations.

References

- [1] M. Kulmala *et al* 2013 *Science* **339** 943
- [2] J. Fedor *et al* 2011 *J. Chem. Phys.* **135** 104305
- [3] J. Lengyel *et al* 2012 *J. Chem. Phys.* **137** 034304 104305

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