

Global atmospheric particle formation from CERN CLOUD measurements

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New particle formation (or nucleation) is acknowledged as a significant source of climate-relevant aerosol throughout the atmosphere. However, performing atmospherically relevant nucleation experiments in a laboratory setting is extremely challenging. As a result, until now, the parameterisations used to represent new particle formation in global aerosol models were largely based on in-situ observations or theoretical nucleation models, and usually only represented the binary $\text{H}_2\text{SO}_4\text{-H}_2\text{O}$ system.

Several different chemicals can affect particle formation rates, even at extremely low trace concentrations, which are technically challenging to measure directly. Nucleation rates also respond to environmental changes in e.g. temperature in a highly non-linear fashion. The CERN CLOUD experiment was designed to provide the most controlled and accurate nucleation rate measurements to date, over the full range of free tropospheric temperatures and down to sulphuric acid concentrations of the order of 10^5 cm^{-3} . We will present a parameterisation of inorganic nucleation rates for use in global models, based on these measurements, which includes four separate nucleation pathways: binary neutral, binary ion-induced, ternary neutral, and ternary ion-induced.

Both inorganic and organic nucleation parameterisations derived from CLOUD measurements have been implemented in the GLOMAP global aerosol model. The parameterisations depend on temperature and on concentrations of sulphuric acid, ammonia, organic vapours, and ions. One of CLOUD's main original goals was to determine the sensitivity of atmospheric aerosol to changes in the nucleation rate over a solar cycle. We will show that, in a present-day atmosphere, the changes in climate-relevant aerosol (in the form of cloud-level cloud condensation nuclei) over a solar cycle are on average about 0.1%, with local changes of less than 1%. In contrast, anthropogenic changes in ammonia since pre-industrial times were estimated to have a much greater influence, resulting in a radiative forcing of between -0.62 and -0.66 W m^{-2} .

Including ternary inorganic pathways in GLOMAP improved the model's agreement with free tropospheric observations, especially aircraft measurements. The further inclusion of an organic parameterisation, which increased nucleation in the summertime boundary layer, brought our results more in line with observations made at surface stations. We therefore believe that, while the addition of other nucleation pathways (such as amine-induced nucleation) will doubtless improve agreement with local in-situ measurements, this model set-up provides a good representation of the global atmosphere as a whole.

By presenting this novel parameterisation at EGU, we hope to encourage its uptake among the aerosol modelling community.