

Supporting information to press briefing on Nature publications by the CLOUD collaboration:

Kirkby, J. et al. *Ion-induced nucleation of pure biogenic particles*. *Nature*, doi 10.1038/nature17953 (2016).

Tröstl, J. et al. *The role of low-volatility organic compounds in initial particle growth in the atmosphere*. *Nature*, doi 10.1038/nature18271 (2016).

The background to the CERN CLOUD experiment. CLOUD is studying how new aerosol particles form or “nucleate” in the atmosphere and grow to sizes where they modify clouds and climate. Using a particle beam from the CERN Proton Synchrotron, CLOUD is also investigating whether these processes are affected by ionisation from galactic cosmic rays. Atmospheric aerosol particles cool the climate by reflecting sunlight and by forming more numerous but smaller cloud droplets, which makes clouds brighter and extends their lifetimes. Cooling due to increased aerosol particles from human activities has offset part of the warming caused by increased greenhouse gases. To determine the amount of cooling requires knowledge of the aerosol state of the pre-industrial atmosphere. Unfortunately we cannot directly measure this since there are almost no regions of today’s atmosphere that are perfectly free of pollution. So the pre-industrial atmosphere must be simulated with climate models based on sound measurements of the underlying microphysical processes obtained by laboratory experiments. CLOUD brings together fundamental experiments with climate modeling in a single international collaborative effort.

What has CLOUD studied? CLOUD has studied the formation of new atmospheric particles in a specially designed chamber under extremely well controlled laboratory conditions of temperature, humidity and concentrations of nucleating and condensing vapours. In the present experiments we measured the formation and growth of particles purely from organic vapours emitted by trees (so-called biogenic vapours). The particular vapour studied was alpha-pinene, which gives pine forests their characteristic pleasant smell. Alpha-pinene is rapidly oxidised on exposure to ozone, creating vapours with extremely low volatilities but only tiny concentrations of around one molecule per trillion (10^{12}) air molecules.

What’s special about the CLOUD experiment? Using CERN know-how, the CLOUD chamber has achieved much lower concentrations of contaminants than all previous experiments, allowing us to measure particle nucleation and growth from biogenic vapours in the complete absence of contaminant vapours such as sulphuric acid. The collaboration has developed state-of-the-art instruments to measure the vapours, ions and aerosol particles at ultra low concentrations in the air sampled from the CLOUD chamber. We measure how these vapours and ions form molecular clusters and which vapours control the subsequent particle growth. A special feature of CLOUD is its capability to measure nucleation enhanced by cosmic-ray ionisation generated by a CERN pion beam - or with all the effects of ionisation completely suppressed by an internal electric field.

What has CLOUD discovered? CLOUD has found that oxidised biogenic vapours produce abundant particles in the atmosphere in the absence of sulphuric acid. Previously it was thought that sulphuric acid – which largely arises from sulphur dioxide emitted by fossil fuels – was essential to initiate particle formation. We found that ions from galactic cosmic rays strongly enhance the production rate of pure biogenic particles – by a factor 10-100 compared with particles without ions, when concentrations are low. We also show that oxidised biogenic vapours dominate particle growth in unpolluted environments, starting just after the first few molecules have stuck together and continuing all the way up to sizes above 50-100 nm where the particles can seed cloud droplets. The growth rate accelerates as the particles increase in size, as progressively higher-volatility biogenic vapours are able to participate. We quantitatively explain this with a model of organic condensation.

Why is it important for our understanding of climate? Ion-induced nucleation of pure biogenic particles may have important consequences for pristine climates since it provides a hitherto-unknown mechanism by which nature produces particles without pollution. And, once embryonic particles have formed, related but more abundant oxidised biogenic vapours cause the particle growth to accelerate. Rapid growth of the new particles while they are still small and highly mobile implies a larger fraction will avoid coagulation with pre-existing larger particles and eventually reach sizes where they can seed cloud droplets and influence climate. Pure biogenic nucleation and growth may raise the baseline aerosol state of the pristine pre-industrial atmosphere and so may reduce the estimated anthropogenic radiative forcing from increased aerosol-cloud albedo over the industrial period. Ion-induced pure biogenic nucleation may also shed new light on the long-standing question of a physical mechanism for solar-climate variability in the pristine pre-industrial climate.

A paper published simultaneously in *Science* (Bianchi, F. et al. *Science*, doi 10.1126/science.aad5456, 2016) reports observations made at the Jungfraujoch of pure organic nucleation in the free troposphere, confirming the relevance of the CLOUD measurements to the atmosphere.