Supporting information to press briefing on Nature publication, **Kirkby et al.,** ***"Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation", DOI 10.1038/nature10343***

The background to the CERN CLOUD experiment. CLOUD is tackling one of the most challenging and long-standing problems in atmospheric science – to understand how new aerosol particles are formed in the atmosphere and the effect these particles have on climate. Increases in atmospheric aerosol particles cool the climate by reflecting more sunlight and by forming additional cloud drops, thereby making clouds brighter. The increased amount of aerosol in the atmosphere caused by human activities is thought to have offset a large fraction of the warming caused by greenhouse gases. By current estimates, about half of all cloud drops are formed on aerosol particles that were "nucleated" (that is, produced from the clustering of trace atmospheric molecules rather than being emitted directly into the atmosphere, like sea spray particles). Nucleation is therefore likely to be important for climate. However, the physical mechanisms of nucleation are not understood, so global models have been based on theoretical calculations or have been adjusted to match observations. CLOUD aims to understand the nucleation process and therefore provide reliable aerosol physics to reduce the uncertainty in climate forcings and projections.

What exactly has CLOUD studied? CLOUD has studied the nucleation of new particles in a specially designed chamber under extremely well controlled conditions of temperature, humidity, ionisation and concentrations of nucleating vapours. We measured the creation of new particles caused by sulphuric acid and ammonia vapours, which have long been thought to account for nucleation in the real atmosphere. CLOUD also measured nucleation of new particles caused by ions that are generated in the air by cosmic rays. Carefully controlled laboratory experiments like CLOUD provide the best way of understanding whether cosmic rays could affect Earth's clouds and climate, as has been proposed.

What is special about the CLOUD experiment? The CLOUD chamber has much lower concentrations of contaminants than all previous experiments, allowing us to measure the nucleation due to controlled amounts of selected trace gases without the complicating effect of undetected gases. CLOUD used state of the art instruments to measure very low concentrations of atmospheric vapours and, with a unique new instrument, has measured the chemistry and growth of newly formed charged molecular clusters from single molecules up to full particles. Another unique aspect is the capability to measure nucleation due to ionising natural cosmic rays, or due to enhanced ionisation provided by the CERN pion beam - or with the effects of all ionisation completely suppressed.

What has CLOUD discovered and why is it important for our understanding of climate? There are several important discoveries from CLOUD. Firstly, we have shown that the most likely nucleating vapours, sulphuric acid and ammonia, cannot account for nucleation that is observed in the lower atmosphere. The nucleation observed in the chamber occurs at only one-tenth to one-thousandth of the rate observed in the lower atmosphere. Based on the first results from CLOUD, it is clear that the treatment of aerosol formation in climate models will need to be substantially revised, since all models assume that nucleation is caused by these vapours and water alone. It is now urgent to identify the additional nucleating vapours, and whether their sources are mainly natural or from human activities.

Secondly, we have found that natural rates of atmospheric ionisation caused by cosmic rays can substantially enhance nucleation under the conditions we studied – by up to a factor of 10. Ion-enhancement is particularly pronounced in the cool temperatures of the mid-troposphere and above, where CLOUD has found that sulphuric acid and water vapour can nucleate without the need for additional vapours. This result leaves open the possibility that cosmic rays could also influence climate. However, it is premature to conclude that cosmic rays have a significant influence on climate until the additional nucleating vapours have been identified, their ion enhancement measured, and the ultimate effects on clouds have been confirmed.