

Supporting information to press briefing on Nature publication, **Almeida et al. (CLOUD collaboration), "Molecular understanding of sulphuric acid-amine particle nucleation in the atmosphere", DOI 10.1038/nature12663.**

**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. Atmospheric aerosol particles cool the climate by reflecting sunlight and by forming smaller but more numerous cloud droplets, which makes clouds brighter and extends their lifetimes. The increased amount of aerosol in the atmosphere caused by human activities has compensated for 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 tiny concentrations of atmospheric molecules rather than being emitted directly into the atmosphere, like sea spray particles). Nucleation is therefore likely to be a key process in climate regulation. However, the physical mechanisms of nucleation are not understood. We also do not understand which molecules participate in nucleation, and whether they derive from natural sources or are emitted by human activities.

**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 vapours. We measured the creation of particles caused by sulphuric acid and tiny concentrations of dimethylamine near one molecule per trillion ( $10^{12}$ ) air molecules. Amines are atmospheric vapours closely related to ammonia, largely derived from anthropogenic activities (mainly animal husbandry), but they are also emitted by the oceans, the soil and from biomass burning. We studied particle formation involving amines because they are known to form strong chemical bonds with sulphuric acid and their presence might explain why nucleation is very often observed in the lower atmosphere.

**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 nucleation due to controlled amounts of selected trace gases without the complicating effect of unwanted gases. State-of-the-art instruments connected to the CLOUD chamber measure extremely low concentrations of atmospheric vapours and the molecular makeup and growth of newly-formed molecular clusters from single molecules up to stable aerosol particles. Another unique feature of CLOUD is the capability to measure nucleation enhanced by cosmic rays at intensities between ground level and (using a CERN pion beam) the top of the atmosphere - or with the effects of all ionisation completely suppressed by an internal electric field.

**What has CLOUD discovered and why is it important for our understanding of climate?** We have shown that amines at typical atmospheric concentrations of only a few molecules per trillion air molecules combine with sulphuric acid to form highly stable aerosol particles at rates similar to those observed in the lower atmosphere. This is the first time – in either the laboratory or the atmosphere - that the formation rates of atmospheric aerosol particles have ever been reproduced or identified with clusters of precisely-known molecular composition. The highly detailed measurements have enabled us to develop a fundamental understanding of the nucleation process at the molecular level, by showing that the experimental measurements can be reproduced by quantum chemical calculations of molecular clustering. The results suggest that natural and anthropogenic sources of amines could influence climate. In particular, amine scrubbing is likely to become the dominant technology for CO<sub>2</sub> capture from fossil fuel power plants, so anthropogenic amine emissions are expected to increase in the future. The spread of these amines into otherwise pristine regions could create new particles in the atmosphere and contribute to the cooling effect of particles on climate. We have also shown that ionisation by cosmic rays has only a small effect on the formation rate of amine-sulphuric acid particles, suggesting that cosmic rays are unimportant for the generation of these particular aerosol particles in the atmosphere. Our measurements leave open the possibility that nucleation of sulphuric acid particles in the lower atmosphere may also proceed with other vapours, for which the effect of cosmic ray ionisation may be different.