Neutral and ion-induced H_2SO_4 - H_2O particle formation 2: CLOUD experimental data spanning all atmospheric conditions

J. Duplissy[1], J. Merikanto[2], A. Määttänen[3], H. Henschel[1], N. M. Donahue[4], D. Brus[2], S. Schobesberger[1], M. Kulmala[1], H. Vehkamäki[1], and the CLOUD collaboration

[1] University of Helsinki, Helsinki, Finland.

[2] Finnish Meteorological Institute, Helsinki, Finland

[3] LATMOS/IPSL, UVSQ Université Paris-Saclay, UPMC Univ. Paris 06, CNRS, Guyancourt, France.

[4] Carnegie Mellon University, Pittsburgh, Pennsylvania, USA.

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Presenting author email: joonas.merikanto@fmi.fi

We present a data set of binary particle formation of sulfuric acid and water, measured in the CLOUD chamber at CERN (Figure 1) during the CLOUD3 and CLOUD5 campaigns (Duplissy *et al*, 2016). Four parameters have been varied to cover neutral and ion-induced binary particle formation processes: Sulfuric acid concentration (1e5 to 1e8 molecules per cm^{-3}), relative humidity (10% to 80%), temperature (208-293K) and ion concentration (0-4000 ions per cm^{-3}). In addition, we have derived a new version of Classical Nucleation Theory (CNT) to treat particle formation in neutral and ion-induced binary sulfuric acid-water systems (*Merikanto et al.*, 2016). The new model is also presented in a companion EAC2016 abstract by Merikanto *et al*., and parameterized for the use in large scale atmospheric models as presented in a companion abstract by Määttänen *et al*. The CLOUD data and the model are compared in this presentation (an example shown in Figure 1). We find a good agreement between theoretical and measured particle formation rates throughout the full range of measurements.

The CLOUD chamber at CERN (fully described in Duplissy *et al* (2016)) is a 3m-diameter electro-polished stainless-steel cylinder (26.1 m^3). The set-up is shown in Figure 1. A field cage is installed inside the chamber to allow the removal of ions, when required. The contents of the chamber is irradiated by UV light in the range 250-400 nm and mixed by two fans. Experimental runs can be performed at stable temperatures between 40°C and -65°C. The chamber is exposed to a 3.5 GeV/c secondary pion beam from the CERN PS, corresponding to the characteristic energies and ionization densities of cosmic ray muons in the lower troposphere. The beam intensity can be adjusted to cover the natural ion range concentration from ground level to the stratosphere. Ultra-pure air is obtained from the evaporation of cryogenic liquid N_2 and liquid O_2. The air is humidified with a Nafion humidifier. Trace gases such as SO_2 are added from gas cylinders containing pressurised N_2 as the carrier. The chamber instrumentation includes PTRMS, CIMS, Nano-SMPS, CPC battery, PSM, APi-ToF, NAIS, Gerdien, LOPAP, dew point sensor, SO_2 and O_3 analyser, as well as T, P and UV sensors. Sulfuric acid was generated by means of UV light, SO2 and water. From the APi-tof data, pure binary particle formation of sulfuric acid and water can be clearly identified, and particle formation processes including other species than water and sulfuric acid have been excluded from this study.

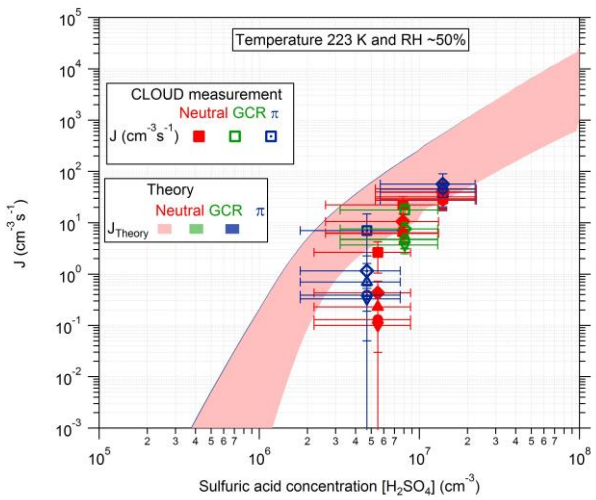

Figure 1: A comparison between CLOUD data taken at 223 K, 50% RH for neutral, GCR and GCR+beam (π) runs, and theoretical total formation rates under same conditions. Here, neutral formation dominates both theoretical and measured rates.

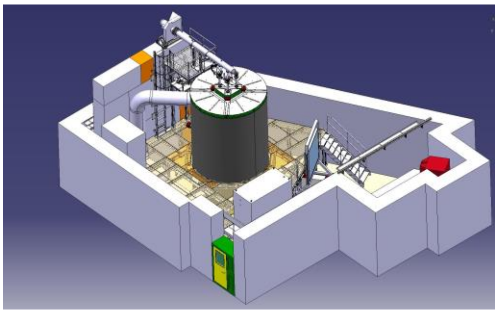

Figure 2. An illustration of CLOUD chamber in the T11 experimental zone at the CERN PS. The de-focused particle beam exits a dipole magnet (right), crosses the hodoscope counter (middle) and then traverses the 3m-diameter CLOUD chamber

Duplissy, J., et al. (2016). *J. Geophys. Res. Atmos.* **121**, doi:10.1002/2015JD023539.

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