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CLOUD Collaboration

Aerodyne Research Inc., Billerica, Massachusetts 01821, USA
California Institute of Technology, Div. of Chemistry & Chemical Engineering, Pasadena, California 91125, USA
Carnegie Mellon University, Center for Atmospheric Particle Studies, Pittsburgh PA 15213-3890, USA
CERN, CH-1211 Geneva, Switzerland
Finnish Meteorological Institute, FI-00101 Helsinki, Finland
Goethe-University of Frankfurt, Institute for Atmospheric and Environmental Sciences, 60438 Frankfurt am Main, Germany
Helsinki Institute of Physics, University of Helsinki, FI-00014 Helsinki, Finland
Karlsruhe Institute of Technology, Institute for Meteorology and Climate Research, 76344 Eggenstein-Leopoldshafen, Germany
Lebedev Physical Institute, Solar and Cosmic Ray Research Laboratory, 119991 Moscow, Russia
Leibniz Institute for Tropospheric Research, 04318 Leipzig, Germany
Paul Scherrer Institute, Laboratory of Atmospheric Chemistry, CH-5232 Villigen, Switzerland
TOFWERK AG, CH-3600 Thun, Switzerland
University of Eastern Finland, Department of Applied Physics, FI-70211 Kuopio, Finland
University of Helsinki, Department of Physics, FI-00014 Helsinki, Finland
University of Innsbruck, Institute for Ion and Applied Physics, 6020 Innsbruck, Austria
University of Leeds, School of Earth and Environment, LS2-9JT Leeds, UK
University of Lisbon and University of Beira Interior, 1749-016 Lisbon, Portugal
University of Manchester, School of Earth, Atmospheric and Environmental Sciences, Manchester M13 9PL, UK
University of Stockholm, Department of Applied Environmental Science, 10691 Stockholm, Sweden
University of Vienna, Faculty of Physics, 1090 Vienna, Austria

1 INTRODUCTION

Cloud droplets form on aerosol particles—tiny liquid or solid particles suspended in the atmosphere. Aerosol particles therefore affect cloud droplet concentrations and, in turn, influence a host of cloud properties including reflectivity, precipitation, lifetime, dynamics and electrification. Aerosol-cloud interactions have a major influence on climate. Increases of aerosol particles in the 20^{th} century due to anthropogenic activities are thought to have offset a large fraction of the warming due to rising atmospheric concentrations of greenhouse gases [2]. Aerosol-cloud processes constitute the largest present uncertainty in radiative forcing of climate, and limit our ability to make accurate projections of future climate [3].

CLOUD aims to resolve the influence of galactic cosmic rays on clouds and climate by carrying out a series of controlled laboratory measurements of aerosol-cloud processes. The experiment involves a large chamber exposed to a beam from the CERN PS, which provides an adjustable source of 'cosmic rays' [1]. Many of the fundamental physical and chemical processes involved in the condensation of trace atmospheric vapours to form aerosol particles, their growth to cloud-active sizes and their interaction with haze and clouds are poorly understood. CLOUD is therefore contributing important new results

on a wide range of aerosol-cloud processes—even before considering the possible influence of galactic cosmic rays.

In addition to studying aerosol nucleation and growth, CLOUD is also investigating possible direct effects of cosmic rays on cloud microphysics and aerosol-cloud processing. These experiments require the formation of liquid and ice clouds inside the CLOUD chamber (termed *cloudy* experiments).

CLOUD controls the ion concentrations inside the chamber from effectively zero up to atmospheric values between ground level and the upper troposphere (corresponding to full T11 beam intensity). The zero-ion conditions are simulated with an internal electric clearing field of about 20 kV/m, which sweeps ions out of the CLOUD chamber in about 1 second. Figure 1 shows an example run from the CLOUD7 campaign (1 Oct – 2 Dec 2012) during which changes of ionisation conditions directly lead to changes of the formation rate of aerosol particles (ion-induced nucleation).

2 CLOUD RUNS IN 2012

2.1 CLOUD6 (4 Jun – 2 Jul 12): technical run for cloud formation

During 2012 the first technical tests were made of a new system for controlled adiabatic pressure reductions (or, equivalently, volume expansions) inside the CLOUD chamber to create liquid and ice clouds (Fig. 2). The chamber normally operates at +5 mbar relative to atmospheric pressure (the small positive pressure ensures no back-flow of ambient air into the chamber). However the chamber and gas system are designed to operate at up to +200 mbar relative pressure and to make controlled adiabatic pressure reductions (volume expansions) down to +5 mbar. In this way, starting from relative humidities near 100%, the chamber can be operated as a classical Wilson cloud chamber—although operating at atmospheric conditions, which are far removed from those of an ionising particle detector.

In addition to the expansion system, several new components of the CLOUD facility were introduced for CLOUD6, including a fast-response horizontal thermocouple string for measurement of air temperature, two cyclone samplers for measuring interstitial aerosol particles, a second-generation UV sabre for increased UV intensity, and a CIGAR prototype (corona ion generator for aerosol research) to generate unipolar charges inside the chamber.

The expansion system operated very successfully. Figure 3 shows an example of the formation in the chamber of a supercooled liquid cloud and subsequent homogeneous freezing near 237 K.

2.2 CLOUD7 (1 Oct – 13 Dec 12): aerosol particle nucleation and growth with organic species

The CLOUD7 run involved the most complex series of experiments carried out so far by CLOUD. Around 25 sampling instruments were attached to the CLOUD chamber, including 9 state-of-art mass spectrometers to provide molecular analysis of vapours and nucleating clusters (Fig. 6 and Table 1). Several of these instruments were completely new and had only been in operation for a few weeks.

The primary goal of CLOUD7 was to investigate aerosol particle nucleation and growth involving α-pinene, which is an important biogenic monoterpene emitted by forests and frequently found in the boundary layer at mixing ratios of a few 100 pptv or more. Monoterpenes are oxidised in the atmosphere, producing low volatility oxidised organic compounds that play an important role in atmospheric aerosols. In order to carry out the CLOUD7 experiments, several new gases were developed for CLOUD:

- α-pinene ($C_{10}H_{16}$, 0–1200 pptv): provided from a custom-designed stainless steel evaporator.
- Nitrous acid (HNO_2, 0–1000 pptv): provided from a custom-designed stainless steel continuous-flow reaction vessel mixing H_2SO_4 and $NaNO_2$, and bubbling ultrapure N_2 through the mixture as carrier gas.
- Hydrogen (0.1%): provided from a battery of $12 \times 50\ l \times 200$ bar bottles containing electrolytically-produced H_2.

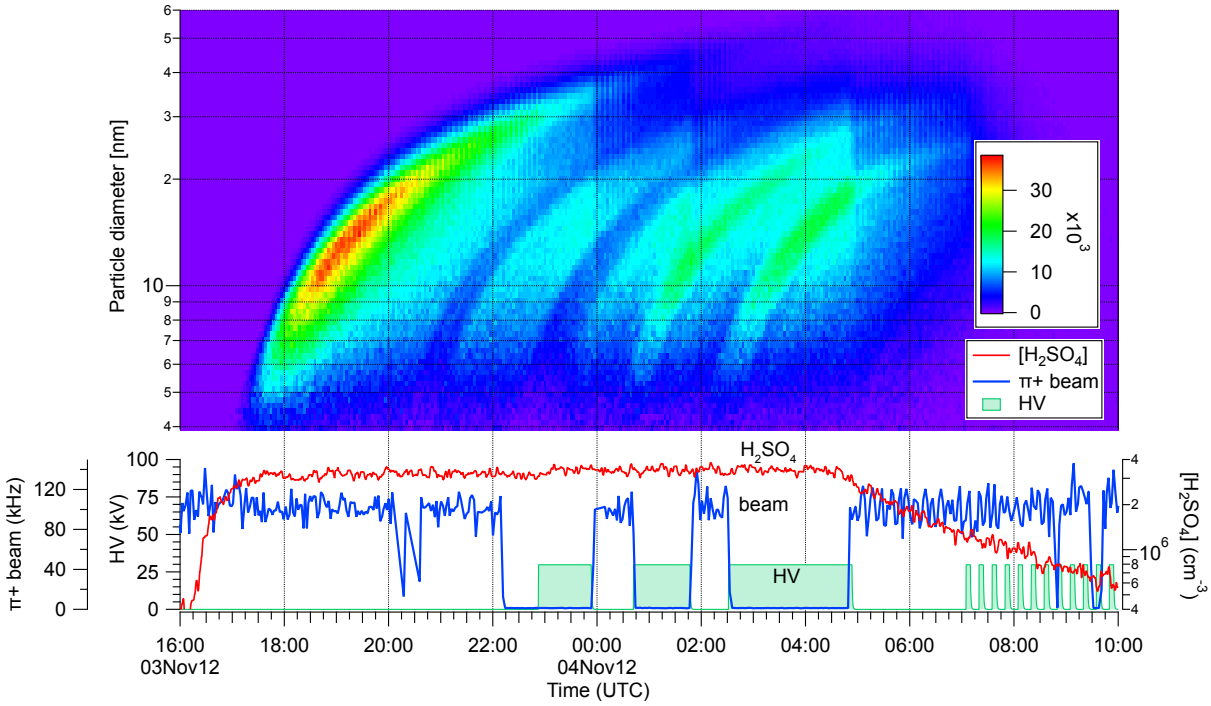

Fig. 1: Example of ion-induced particle formation during CLOUD7. a) The run started at 16:11, 3 November 2012, when the fibre-optic UV system was turned on to establish $4 \cdot 10^6$ cm^{-3} [H_2SO_4] in the chamber. The UV was turned off at 04:49, 4 November 2012, and the chamber cleared of aerosol particles in preparation for the next run. During the run, only the ionisation conditions were varied in the CLOUD chamber, by adjustments of the beam intensity and/or high voltage clearing field as indicated. b) The variations of ionisation produced a strong response in the rate of nucleation of new particles, as shown by nucleation bursts (banana-shaped events) recorded in the scanning mobility particle sizer (SMPS).

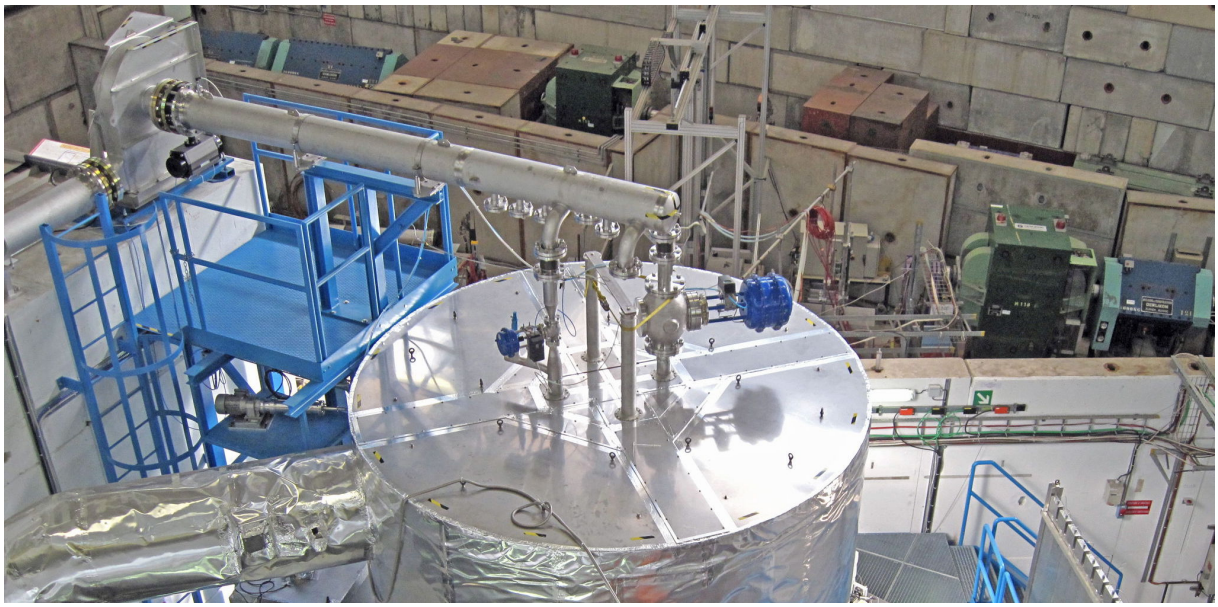

Fig. 2: Pressure regulation valves and fast exhaust system installed above the CLOUD chamber for the CLOUD6 run, June–July 2012, to produce controlled adiabatic pressure reductions (volume expansions) and create liquid and ice clouds inside the chamber ("cloudy" experiments).

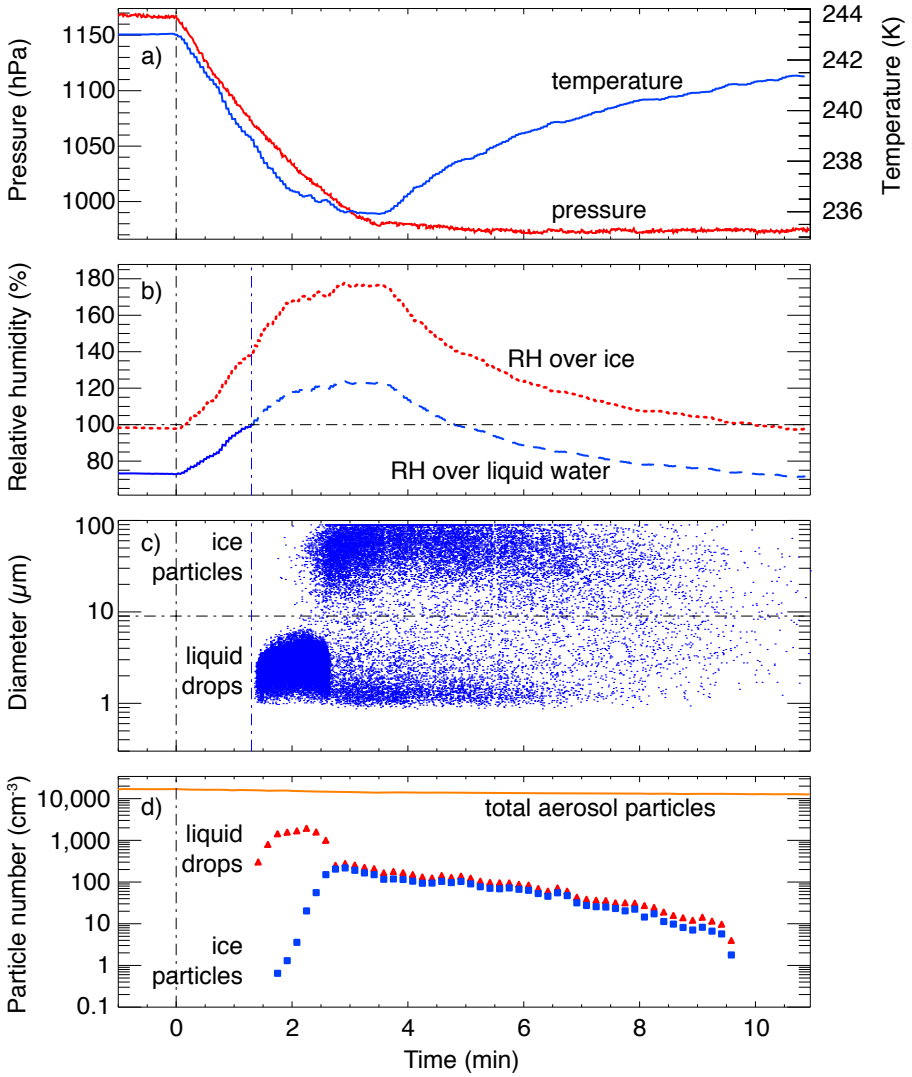

Fig. 3: Example of cloud formation vs. time during CLOUD6: a) adiabatic pressure reduction and air temperature measured inside the CLOUD chamber with fast-response thermocouples, b) total relative humidity (RH) over water and ice, c) particle counts and size detected by the Welas instrument mounted underneath the chamber, with a vertical sampling probe, and d) number concentrations of aerosol particles, cloud liquid drops and cloud ice particles. As the pressure falls, the temperature falls and the RH rises (panels a and b). When the RH exceeds 100% over liquid water, a liquid cloud forms and the droplets grow to a maximum size of about 6 μm (panel c). When the temperature falls below about 237 K, the liquid droplets start to freeze homogeneously and the ice particles experience a high water vapour supersaturation near 140%, which drives very rapid growth to large sizes near 100 μm. The rapid growth of ice particles depletes the available water vapour and so the remaining liquid droplets evaporate. This reproduces the *Bergeron-Findeisen mechanism* which is an important atmospheric process responsible for rainfall from supercooled liquid clouds (caused by sedimentation of large ice particles). After the pressure reduction ceases, the air is re-heated by the relatively warm chamber walls and the cloud eventually evaporates completely.

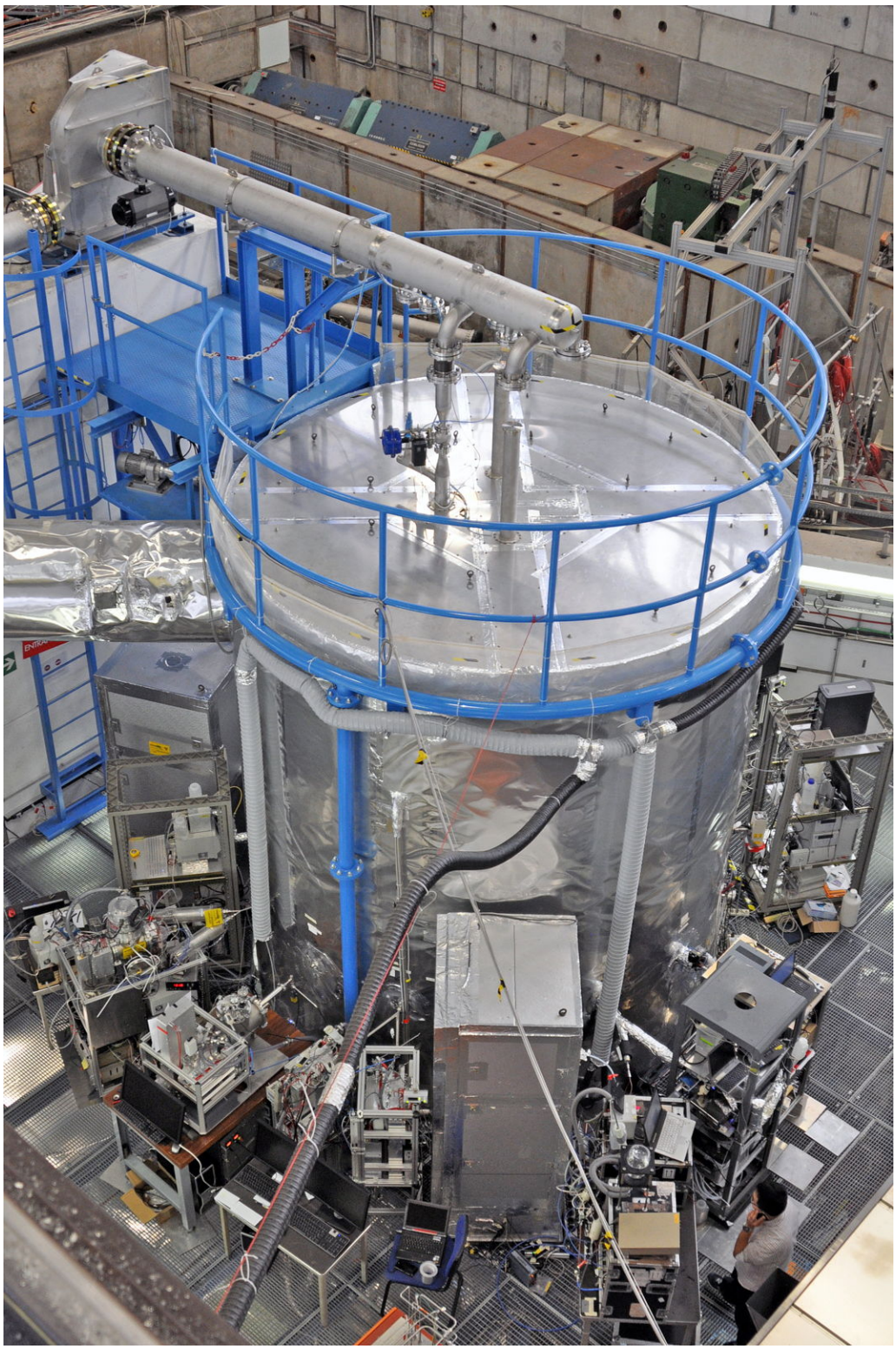

Fig. 4: Experimental configuration during the CLOUD7 campaign at CERN, October-December 2012. A total of 25 sampling instruments surround the chamber at mid-height, including 9 state-of-art mass spectrometers to provide molecular analysis of vapours and nucleating clusters.

Table 1: CLOUD instrumentation during the CLOUD7 run, October–December 2012.

Mass spectrometers	Particle counters	Particle sizers	Particle analysers	Gas analysers
APITOF-	PSM_1.1	scanning PSM	HTDMA	IC
APITOF+	DEG-CPC_1.7	radial DMA	OTDMA	LOPAP
CI-APITOF_1	DEG-CPC_2.0	LDT		SO_2
CI-APITOF_2	CPC_3.2	SMPS		O_3
CIMS	CPC 3010			dewpoint
IMS-TOF				
NAIS				
PTRTOF				
TDCIMS				
Key:				
APITOF	atmospheric pressure interface time of flight mass spectrometer			
CI-APITOF	chemical ionisation APITOF			
CIMS	chemical ionisation mass spectrometer			
CPC	condensation particle counter			
DEG-CPC	diethylene glycol CPC			
DMA	differential mobility analyser			
HTDMA	hygroscopic tandem DMA			
IC	ion chromatograph			
IMS-TOF	ion mobility spectrometer - time of flight mass spectrometer			
LDT	laminar diffusion tube CPC			
LOPAP	long path absorption photometer			
NAIS	neutral cluster and air ion spectrometer			
OTDMA	organic tandem DMA			
PSM	particle size magnifier			
PTRTOF	proton transfer reaction time of flight mass spectrometer			
SMPS	scanning mobility particle sizer			
TDCIMS	thermal desorption chemical ionisation mass spectrometer			

Further trace gases used during CLOUD7 included ozone, sulphur dioxide, ammonia and dimethylamine. Aerosol nucleation and growth in the presence of α-pinene was studied under three oxidation conditions to isolate the underlying physico-chemical processes:

1. Pure OH oxidation: OH production by UV photolysis of HNO_2 in the absence of ozone.
2. Pure O_3 oxidation: dark oxidation in the presence of ozone, with the addition of 0.1% H_2 to scavenge OH radicals produced from ozonolysis reactions.
3. Combined OH and O_3 oxidation: oxidation in the presence of ozone and UV light.

The nucleation and growth studies included both α-pinene alone and also α-pinene in the presence of ammonia and/or dimethylamine, all at atmospheric concentrations. A cocktail of these atmospheric vapours was investigated that closely simulates the Hyytiälä field station in Finland [4, 5]. Other studies during CLOUD7 included measurements of neutral molecular cluster compositions using new CI-APITOF instruments (Table 1) and production of H_2SO_4 from sulphur dioxide by ionizing radiation.

CLOUD7 was a very successful run. All the experimental goals were achieved and the results are being analysed for presentations at conferences during 2013 and journal publications.

Fig. 5: Participants at the CLOUD-ITN Conference, Königstein, Germany, 22–25 May 2012.

3 CLOUD MEETINGS IN 2012

The following CLOUD Collaboration meetings and data workshops were held in 2012:

CLOUDy workshop, University of Manchester, 11–12 Jan. Scientific goals and planning for CLOUD6.

CLOUD4–5 data workshop, Hyytiälä, Finland, 27 Feb – 1 Mar. Analysis of data from CLOUD4–5.

Aerosol growth rate workshops, Helsinki, 13–16 Apr and 10–12 Sep. Analysis of aerosol growth rates from CLOUD2–5.

CLOUD-ITN Conference, Königstein, Germany, 22–25 May. Presentation and discussion of results from all CLOUD runs, together with 30 invited external experts from Europe and the United States (Fig. 5), followed by 1-day CLOUD collaboration meeting.

CLOUD collaboration meeting and CLOUD-TRAIN kickoff, CERN, 15–16 Oct. CLOUD collaboration meeting and startup of EU FP7 CLOUD-TRAIN Marie Curie network (15 CLOUD Marie Curie fellows during 4-year project).

4 PUBLICATIONS AND PRESENTATIONS

A summary of all CLOUD runs since the beginning of data taking is shown in Table 2. A major effort is underway to complete the analyses of a large fraction of the new findings from these data during 2013 and to submit publications. Some of the results are either already published [6]–[10] or under current review [11]–[15]. At least ten further manuscripts are expected to be submitted for publication in 2013, including several to high profile journals.

This year's main conference is the 19^{th} International Conference on Nucleation and Atmospheric Aerosols, ICNAA 2013, Fort Collins, Colorado, 24–28 June 2013, <http://chem.atmos.colostate.edu/icnaa/>. Reflecting the high interest of the aerosol-cloud-climate community in the experiment, CLOUD is presenting the opening plenary talk at ICNAA 2013 and a Special Session spanning the first day is devoted to CLOUD results, <http://chem.atmos.colostate.edu/icnaa/ICNAA_2013_Program_sized.pdf>. A total of almost 30 CLOUD abstracts were submitted to the conference.

Table 2: CLOUD runs at the CERN PS to investigate the influence of ionising particles on aerosol nucleation and growth, and cloud microphysics.

Run	Month	Year	Aim
CLOUD1	Nov–Dec	2009	Commissioning
CLOUD2	Jun–Jul	2010	Binary H_2SO_4, NH_3 ternary nucleation
CLOUD3	Oct–Nov	2010	Binary H_2SO_4, NH_3 ternary nucleation
CLOUD4	Jun–Jul	2011	Dimethylamine & pinanediol ternary nucleation
CLOUD5	Oct–Nov	2011	Free tropospheric binary H_2SO_4, NH_3 ternary nucleation
CLOUD6	Jun–Jul	2012	Initial cloud formation experiments
CLOUD7	Oct–Dec	2012	NH_3, dimethylamine & α-pinene ternary nucleation & growth

5 CLOUD8 RUN IN 2013

A single 8-week CLOUD8 run is planned in 2013, 14 Oct – 20 Dec, aimed at cloudy physics experiments to follow on from the successful CLOUD6 technical run. Although the PS beam will not be available, some ion measurements can be made with cosmic rays and, for unipolar charges, with the CIGAR. Several developments are underway to improve the performance of CLOUD for these experiments. Among these is an active system to control the expansion rate according to the thermodynamic state of the chamber and thereby to extend and control the lifetime of the cloud. Improvements are also being made in the uniformity of the air temperature inside the chamber and in its measurement. The latter involves the development of a thermometer string that can measure the vertical temperature profile of the air inside the CLOUD chamber to a sensitivity near 0.01 K with a response time near 1 s and, moreover, is able to operate in a high electric field. Around 30 sampling instruments will be attached to the CLOUD chamber; all these instruments are being tested and, where necessary, modified to operate at up to 200 mbar atmospheric overpressure.

6 FUTURE PLANS

CLOUD is tackling one of the most challenging problems in atmospheric science - to understand the interaction of aerosol particles with clouds and climate, and to quantify the influence of cosmic rays.

Aerosol forcing of climate over the industrial age is estimated to have caused a global average cooling of -0.4 to -1.8 Wm^{-2} due to changes of cloud albedo. This large uncertainty compares to a much better understood forcing of 1.7 ± 0.2 Wm^{-2} from anthropogenic CO_2 emissions. Aerosols therefore have a major influence on climate change but the magnitude of the uncertainty, which has persisted through all IPCC assessments since 1996 [2, 16], limits our confidence in climate change projections [3].

Since ions in the free troposphere and marine boundary layer result mainly from galactic cosmic rays, their role in aerosol-cloud processes is of considerable interest as a possible physical mechanism for solar-climate variability [17].

The basic aims of CLOUD are to:

1. Resolve and quantify the fundamental physical and chemical processes involved in the formation and growth of cloud-active aerosols and the interaction of these aerosols with clouds.

2. Measure the effects of cosmic rays on aerosols and clouds, and settle the question of whether cosmic rays exert a climatically-significant effect on climate and, if so, under what conditions.

3. Develop a mechanistic understanding of the underlying physico-chemical processes and incorporate them into global models capable of simulating the behaviour of ions, aerosols and clouds under realistic meteorological conditions.

4. Reduce the uncertainty in the climate impact of aerosols and their interaction with clouds, leading to more robust climate projections.

CLOUD combines three major research programmes in a single experiment (Fig. 6):

1. Formation of cloud-active aerosols: These experiments identify the condensable vapours contributing to nucleation of new particles in the atmosphere and their growth to sufficient size to form cloud droplets. Nucleation and growth rates are measured over the full range of atmospheric conditions and ion concentrations for each of the participating vapours and their combinations. These experiments have been the main focus of CLOUD so far.

2. Aerosol-cloud interactions: These experiments measure and interpret the activation of cloud condensation nuclei (CCN) to haze or cloud droplets over a wide range of environmental parameters and ionisation conditions. Cloud formation results in substantial modification of aerosol properties which, upon evaporation, feeds into the next cloud formation cycle. Aerosol-cloud processing has barely been previously studied under controlled conditions in the laboratory. So far only a single technical run (CLOUD6) has been made involving cloud formation in the chamber. Aerosol-cloud interactions will be measured for the first time in CLOUD8.

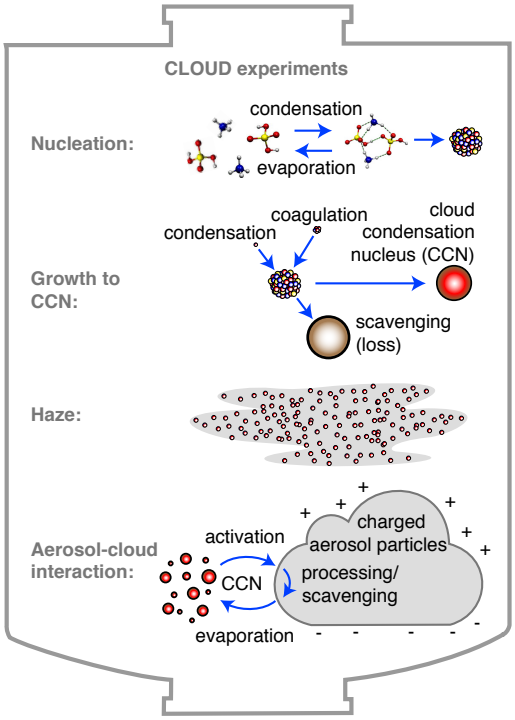

Fig. 6: Present and future CLOUD experiments on ion-aerosol-cloud processes in the laboratory from the nanometer molecular scale to the meter cloud scale.

3. Aerosol-cloud modelling and climate impact: Incorporation of CLOUD measurements into global models is a key aspect of the experiment. In a break from the normal way of using models, the model simulations will increasingly be used to guide the experimental programme, ensuring that most important measurements are made under the key environmental conditions. The new data will be fed into the models and then used to reduce uncertainties and sharpen the predictions. Two new and highly advanced models will be used, and each will be substantially developed by the Leeds CLOUD team. The UK Met

Office Unified Model (UM) is a global weather and climate model capable of nesting down to regional domains of a few hundred km with a spatial resolution of about 100×100 m to resolve individual clouds. The UM can simulate real meteorological situations for any location on the globe to study particular real cloud systems. The Met Office Large Eddy Model (LEM) can simulate cloud-scale processes down to the scale of metres and provides an idealised controlled environment for studying aerosol-cloud interaction processes. Both models will be substantially enhanced to include coupling of aerosols and cloud microphysics on these scales, based on the GLOMAP aerosol model developed by Leeds [18], which in turn will be based on the CLOUD measurements. The first GLOMAP model results from CLOUD are presently in draft manuscript form and will be submitted for publication within a few weeks [19].

This method of directly combining a global aerosol-cloud model with an experiment to specifically quantify and reduce uncertainty is unprecedented in atmospheric science. The global model will resolve individual clouds so that the CLOUD process measurements can be tested, while still allowing the global cycling of aerosols to be simulated. This approach will overcome a fundamental limitation of many previous studies using low resolution (>100 km resolution) global models, which cannot explicitly simulate individual clouds, cloud microphysics or aerosol-cloud interactions.

The CLOUD facility at CERN has established itself as the world's leading experiment for ion-aerosol-cloud studies. CLOUD has also brought together a world-class team of atmospheric and modeling experts to conduct the experiments, analyse and interpret the data, and assess the climate impact of the measurements. A wide range of processes are involved, such as nucleation rates, growth rates, droplet and ice particle activation, and liquid and ice cloud microphysics. Moreover, the dependence of these processes must be measured on numerous variables such as temperature, relative humidity, ionisation, trace gas combinations and their concentrations, and CCN characteristics. Even with careful selection of the parameter space in which to make detailed measurements, we estimate around ten more years will be required to carry out the experimental programme outlined above.

The projected CLOUD experimental programme is therefore likely to extend well beyond the planned upgrade of the East Hall beamlines. After several year's experience in the T11 beamline we would like to request that T11 be retained for CLOUD in the new East Hall beamline layout, ideally with 1–2 m additional space in the experimental zone in the direction towards T10. A dedicated T11 beamline for CLOUD will maximise the efficiency and output of the experiment, and provide the maximum availability of the T9 and T10 beamlines for test-beam users.

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