EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN-SPSC-2015-026
SPSC-SR-165
June 21, 2015

2014 PROGRESS REPORT ON PS215/CLOUD

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

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 concentrations of greenhouse gases in the atmosphere [1]. However aerosol-cloud processes are poorly understood and constitute the largest present uncertainty in radiative forcing of the climate [1].

CLOUD [2, 3] aims to measure the fundamental physico-chemical mechanisms involved in aerosol-cloud processes and, in particular, the influence of galactic cosmic rays. This work should contribute to a sharper understanding of Earth's climate sensitivity and improved confidence in future warming projections for the 21^{st} century. In addition CLOUD will answer the question of whether cosmic rays exert a significant influence on the climate.

CLOUD is now established as the world's most advanced laboratory experiment on atmospheric aerosol particle nucleation [4]. CLOUD has published 17 papers [6]–[22] in peer-reviewed journals so far, of which three are in *Nature* or *Science* [8, 13, 15] and two in the *Proceedings of the National Academy of Sciences* [14, 16]. Some example results from recent publications are shown in Figs.1–3. Four papers [23]–[26] are currently under review and around 10 further papers [27]–[35] are expected to be submitted in 2015. The latter will include the first papers on cloud formation ("cloudy" experiments) in the CLOUD chamber from the CLOUD9 run [34, 35] (an example result is shown in Fig. 4).

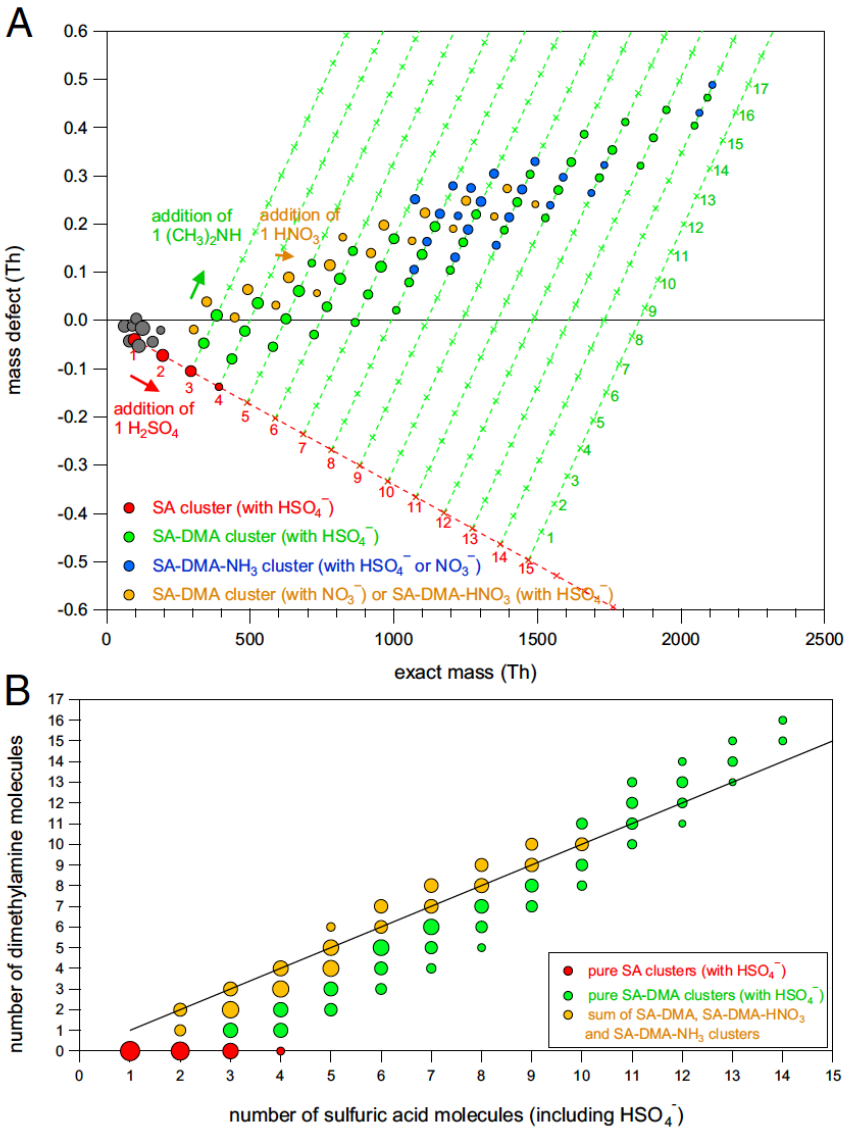

Fig. 1: First measurement of the molecular composition of neutral nucleating clusters [16]. Molecular weight and composition of neutral clusters during a new particle formation event with sulphuric acid and dimethlyamine (DMA). A) Mass defect plot measured with the CI-APi-TOF mass spectrometer for an experiment with 10 pptv DMA and $3 \times 10^7 \mathrm{cm}^{-3}$ H_2SO_4. The mass defect is the difference between the exact mass and the nominal (integer) mass of the cluster. The symbol size is proportional to the logarithm of the signal intensity (count rate). Background ions and clusters (not containing H_2SO_4 or DMA) are indicated by grey symbols. B) The same data with the signals grouped to show the number of SA and DMA molecules in the cluster. Clusters are detected containing up to 14 SA molecules and 16 DMA molecules, corresponding to a mobility diameter near 2.0 nm.

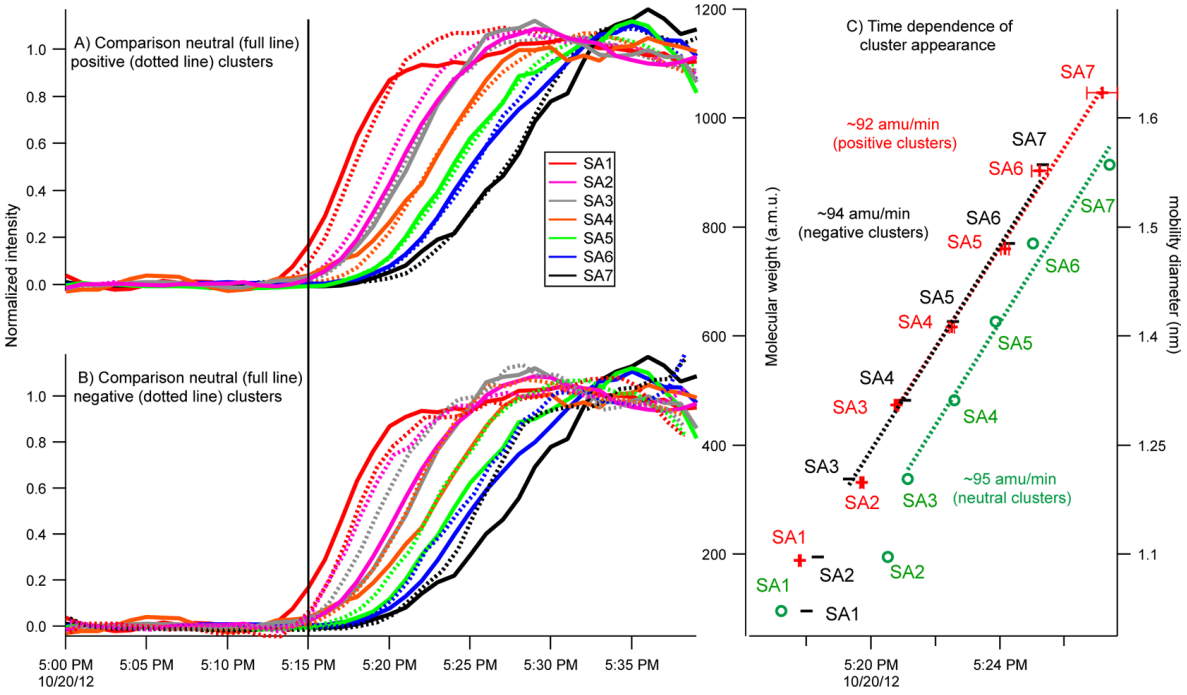

Fig. 2: Molecule-by-molecule growth of nucleating clusters of sulphuric acid (SA) and dimethylamine [18]. The curves in panels A) and B) show the time evolution of clusters containing up to 7 sulphuric acid molecules (SA7). Each curve sums over all clusters containing the indicated number of SA molecules. The black line at 5:15 pm marks the time when the UV illumination was switched on, which initiated the production of H_2SO_4 in the chamber. Panel A compares positive (dotted curves) and neutral clusters (solid curves); panel B compares negative (dotted curves) and neutral clusters (solid curves). Panel C shows the molecular weight and mobility diameter of the clusters versus time. The appearance time of a cluster is taken to be when it reaches 50% of its equilibrium intensity. The curves show the slow rate at which molecules arrive on an embryonic aerosol particle under atmospheric conditions: about one molecular collision per 90 s.

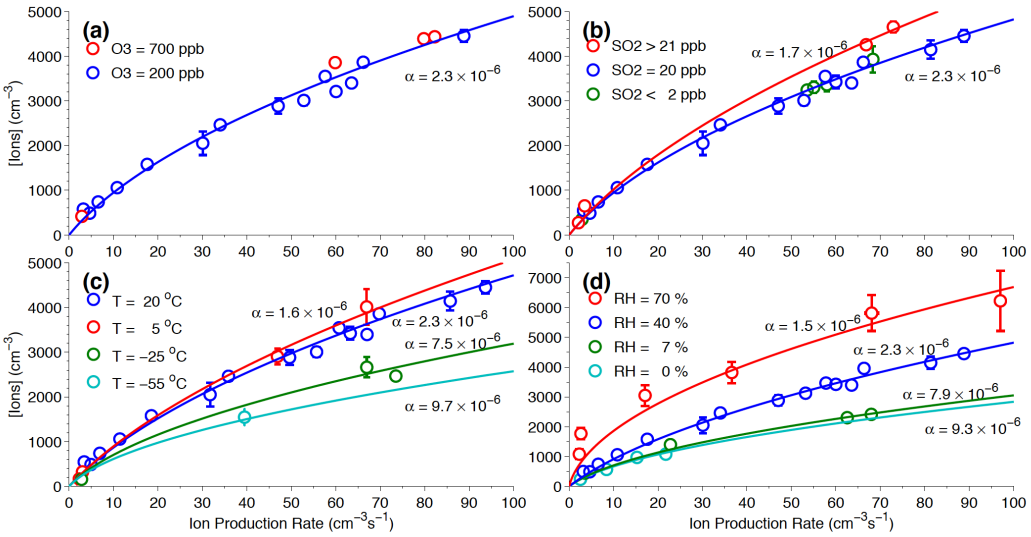

Fig. 3: Dependency of the ion-ion recombination rate on ambient conditions [22]. The curves show the equilibrium ion pair concentrations measured in the CLOUD chamber as a function of the ion production rate (beam intensity) under various conditions: a) ozone concentrations of 200 ppbv and 700 ppbv, at 20°C, 40% relative humidity (RH) and 20 ppbv SO_2; b) SO_2 concentrations between 2 and 30 ppbv, at 20°C, 40% RH and 200 ppbv O_3; c) temperatures of 20°C, 5°C, −25°C and −55°C; and d) 0%, 7%, 40% and 70% RH, at 20°C. Ion-ion recombination causes the ion concentrations to increase more slowly at higher ion production rates. The curves show that the ion-ion recombination rate depends on temperature and relative humidity, which affect the small-ion cluster sizes and diffusion rates.

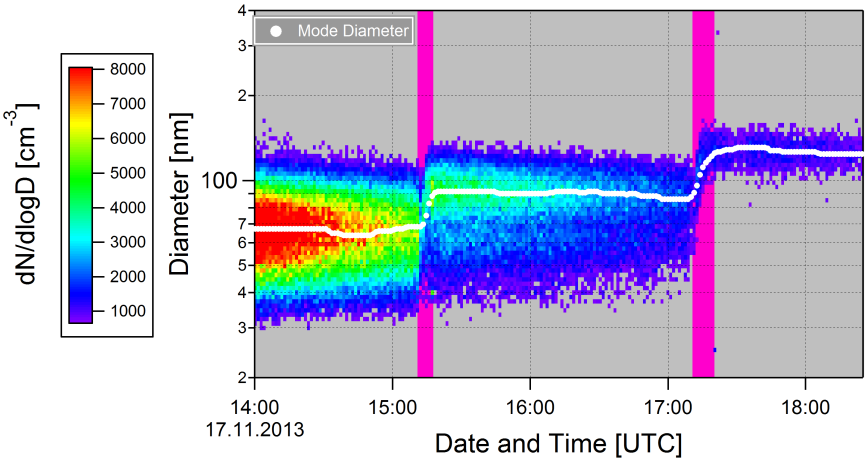

Fig. 4: Aerosol processing of SO_2 into H_2SO_4 in cloud droplets [34]. The data show the evolution of the dry size distribution of cloud condensation nuclei (CCN) over a period of around four hours. The vertical axis indicates the CCN diameter, with a colour scale indicating number concentration (cm^{-3}). The white line shows the mode of the CCN size distribution. During this period there were two adiabatic pressure reductions in the CLOUD chamber, which activated liquid clouds over periods of about 5 minutes (indicated by purple bands). The chamber was re-pressured after the first expansion to prepare for the second cloud activation. Rapid aerosol growth can be seen during both clouds: from 70 to 90 nm in the first cloud and 85 to 110 nm in the second. The figure shows that CLOUD can reproduce cloud processing of trace atmospheric vapours into aerosol under controlled conditions in the laboratory. Cloud processing is a major mechanism for aerosol growth in the atmosphere but very few laboratory data exist so far.

2 CLOUD MEETINGS, 2013–2014

The following CLOUD Collaboration meetings and data workshops were held in 2013–2014:

CLOUD6–7 data workshop, Königstein, Germany, 25 Feb – 1 Mar 2013. Analysis of data from CLOUD6–7.

Aerosol growth rate workshop, Stockholm, Sweden, 2–3 May 2013. Analysis of aerosol growth rates from CLOUD4–7.

Ion workshop, CERN, 6–8 Aug 2013. Analysis of air ion production and loss rates from CLOUD3–7.

CLOUD-TRAIN summer school, Braunfels, Germany, 9–14 Sep 2013. Summer school on aerosol-cloud interactions as part of the CLOUD-TRAIN Marie Curie training network (15 CLOUD Marie Curie fellows during 4-year project).

CLOUD collaboration meeting, CERN, 30 Oct – 1 Nov 2013. CLOUD collaboration meeting and Finance Review Committee meeting (FRC2).

Global modelling workshop, University of Leeds, United Kingdom, 17–20 Feb 2014. Implementation of CLOUD measurements in the Leeds global aerosol model, GLOMAP.

CLOUD7–8 data workshop, Manchester, United Kingdom, 24–28 Mar 2014. Analysis of data from CLOUD7–8.

Aerosol nucleation and growth rate workshop, Paul Sherrer Institute, Switzerland, 2–4 Jul 2014. Analysis of aerosol nucleation and growth rates from CLOUD5–8.

CLOUD collaboration meeting, CERN, 30 Sep – 2 Oct 2014. CLOUD collaboration meeting and Finance Review Committee meeting (FRC3).

3 CLOUD RUNS, 2013–2015

Since the last annual CLOUD Progress Report (CERN-SPSC-2013-014), there have been three CLOUD runs at the PS:

1. *CLOUD8 (14 Oct – 8 Dec 13):*
   - Nucleation of biogenic particles at very low sulphuric acid concentrations.
   - First operation of new cloud particle detectors and cloud condensation nuclei (CCN) generators at CLOUD in preparation for the CLOUD9 run. Extensive development of the software controls of the CLOUD gas system was also involved to generate adiabatic pressure reductions in the CLOUD chamber for liquid and ice cloud activation (see Fig. 5).
2. *CLOUD9 (10 Sep – 24 Nov 14):*
   - Aqueous phase chemistry (chemical processing in liquid clouds). An example is shown in Fig. 4.
   - Ice microphysics.
   - Glassy (highly viscous) secondary organic aerosol and their cloud nucleating properties.
   - Ion production and loss mechanisms.
3. *CLOUD10T (20 Apr – 7 Jun 15):*
   - Technical run to test and develop new instruments for CLOUD10. The new instruments developed at CLOUD10T comprised: acetate (CH_3COOH) CI-APi-TOF, FIGAERO-CIMS, iodine (I^-) CIMS, oxygen (O_2^-) CIMS, DMA-TRAIN, and VIPER. An example of the results obtained with the FIGAERO is shown in Fig. 6.

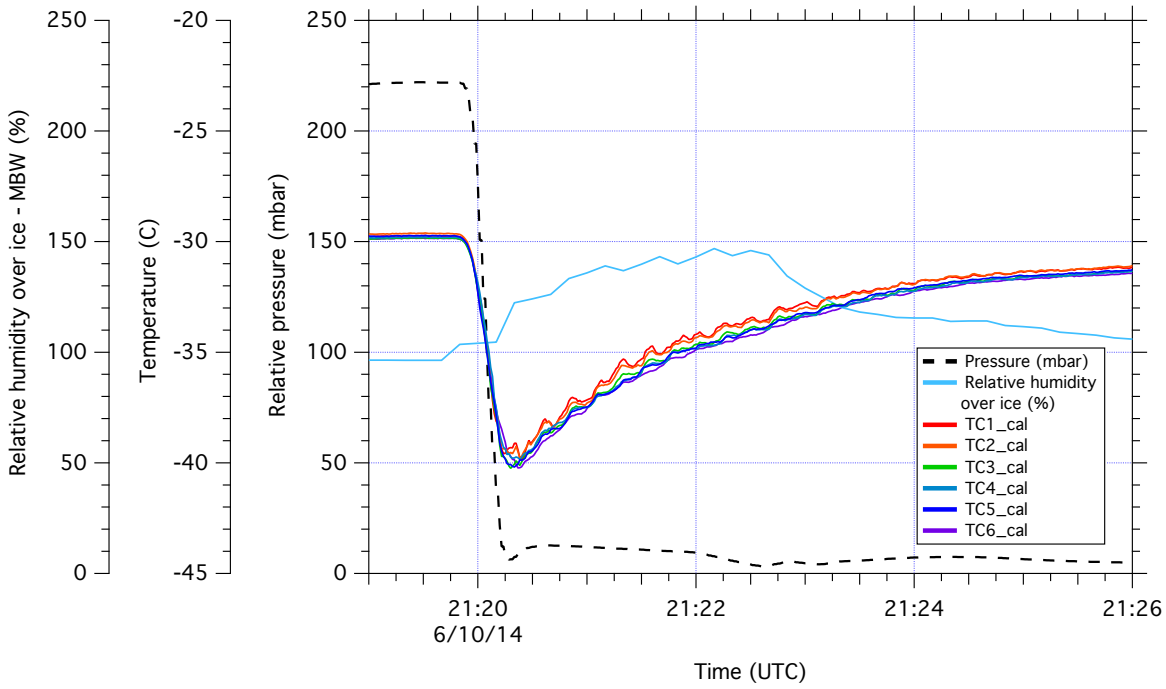

Fig. 5: Activation of clouds in the CLOUD chamber. Clouds are activated by a controlled adiabatic pressure reduction (dashed curve) starting from a relative humidity (blue curve) near 95%. This reduces the air temperature and raises the relative humidity above 100%, leading to the formation of a liquid or ice cloud. Clouds form in the atmosphere by the same process. In the example shown here, the temperature fell below the homogeneous freezing point of water (near -37°C) and so an ice cloud formed. The coloured curves show a string of thermocouples measuring the CLOUD internal air temperature, which falls from -30°C to -40°C during the adiabatic pressure reduction. Clouds can be maintained in the chamber for 5–10 minutes, depending on the selected conditions, until heating by the walls and sedimentation of cloud particles reduces the relative humidity below 100%, at which point the cloud dissipates.

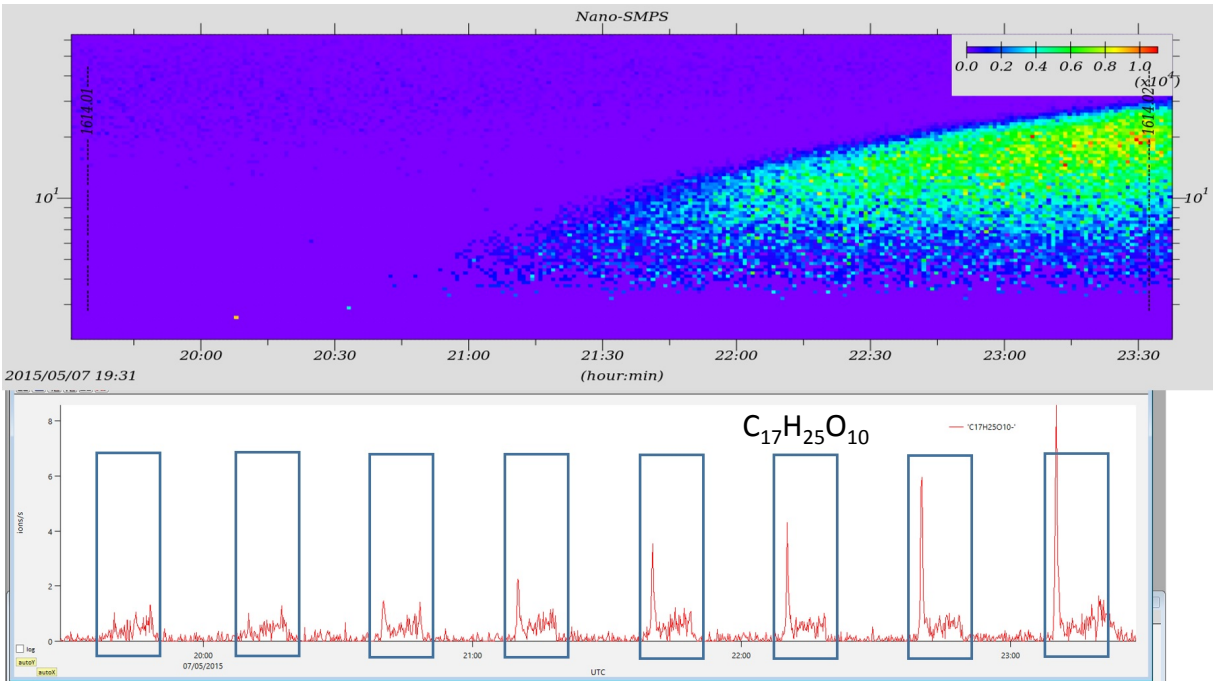

Fig. 6: First measurements from the FIGAERO mass spectrometer during CLOUD10T, May 2015. The newly-developed FIGAERO (Filter Inlet for GAs and AEROsol) mass spectrometer was first operated during the CLOUD10T technical run, May 2015. The instrument is designed to measure the molecular identity and relative contribution of organic compounds responsible for aerosol particle growth at sizes of a few nanometres and above. The first hours of a new particle formation event are shown. Rapid ozonolysis of the biogenic vapour α-pinene ($C_{10}H_{16}$) leads to the production of extremely low volatility organic vapours which contribute to the formation of new particles in the nanometre size range. Subsequent condensation of low volatility vapours causes the particles to grow up to 30 nm diameter. The upper panel shows the number size distribution of particles during the event as a function of time, measured by a scanning mobility particle sizer, SMPS (the cutoff size of the SMPS is around 7 nm). The lower panel exhibits 8 measurement intervals of a semi-online mass measurement from the FIGAERO mass spectrometer, which demonstrates that it is capable of resolving the organic composition of these nanoparticles in the few nm size range. The method involves collection of an extremely small mass of aerosol particles on a filter (detection limit $\sim$0.5 ng, equivalent to the mass of a single 8 μm particle), followed by their thermal evaporation at the entrance of a high-resolution time-of-flight mass spectrometer. The blue rectangles indicate time intervals when sampled particles are vapourised from the filter and analysed after chemical ionisation in the mass spectrometer. The high mass resolution of the spectrometer enables the exact mass of the condensing organic species to be determined, and hence the molecular identity of the ion, $[M-H]^-$, can be determined. The red vapourisation curves (where temperature increases with time) are obtained after sampling the aerosols for fixed time periods of 20 minutes at 10 lpm flow rate. The curves show a background plus a gradually increasing prompt signal at a single selected mass channel corresponding to the compound $C_{17}H_{25}O_{10}$. The measurements clearly identify this compound to be a component of aerosol particles in the few nanometre size range.

4 CLOUD10 RUN, 28 Sep – 4 Dec 2015

The CLOUD10 run is planned for 28 Sep – 4 Dec 2015, with the final 4 weeks using cosmic rays (no East Area beams). The aims of the CLOUD10 run are:

1. Simulation of aerosol particle nucleation and growth at the Hyytiälä boreal forest station under daytime and night-time conditions, involving mixtures of gases that include SO_2, O_3, α-pinene, δ3-carene, NH_3, NO, NO_2 and H_2O.

2. Systematic data at a range of vapour concentrations, beam intensities and temperatures to parametrise H_2SO_4/biogenic nucleation for evaluation of its climate impact using the Leeds global aerosol model, GLOMAP.

3. Isoprene (C_5H_8) oxidation chemistry, and the effect of ions.

4. First look at particle production from anthropogenic volatile organic compounds.

This will be the most ambitious run undertaken by CLOUD so far, involving almost 40 instruments attached to the CLOUD chamber, of which 11 are mass spectrometers (Fig. 7). New features of the CLOUD facility include the use of several new gases as well as a 248 nm KrF UV excimer laser light source for the optical fibre system. The UV laser is estimated to increase the photolysis rate of ozone by around a factor 100 relative to the present Hg-Xe lamps. This will extend the performance of CLOUD to reproduce atmospheric concentrations of the climatically-important hydroxyl $(OH\cdot)$ radical.

Mass Spectrometers:		**Particle counting / sizing:**		**Gas measurements / UV / DP / other:**	
NO3- CI-API-TOF	UHEL	NanoSMPS	PSI	O3 monitor	CERN/EMPA
NO3- CI-API-ToF + TD-DMA	Frankfurt	SMPS	PSI	O3 monitor	Frankfurt
Acetate CI-API-TOF	Kuopio	CPC 2.5	PSI	SO2 monitor	Frankfurt
Acetate CIMS (NH3)	Frankfurt	CPC 3010	PSI	NO 10 ppt	PSI
API-TOF (-ve)	UHEL	DMA Train	Vienna	NO2 10 ppt	PSI / CU-Boulder
NH4+ CI-APi-ToF	PSI/Tofwerk	PSM scanning	UHEL	NO2 30 ppt	PSI
IMS-TOF	PSI	PSM 1.3 nm	UHEL	CO/ CO2 monitor	Frankfurt
PTR-TOF	Innsbruck	PSM 1.5 nm	UHEL	DOAS (glyoxal)	CU-Boulder/PSI
FIGAERO I- CIMS	Aerodyne/CERN	PSM Ion	UHEL	Dew Point Mirror	Tropos
TDCIMS	Kuopio	NAIS	UHEL	Dew Point Mirror	CERN
Peroxy-CIMS	CMU	Nano HT-DMA	Kuopio	Dew Point TDL	KIT
		VIPER	Frankfurt	GCR counter & Hodoscope	Lebedev
		nano radial DMA	Caltech	Temperature sensors	CERN
				UV spectrometer	CERN / CMU
				UV laser (248 nm)	CERN
				absolute air pressure	CERN
				cryogenic N2 and O2 gauges	CERN

Fig. 7: Analysing instruments for the CLOUD10 run, 28 Sep – 4 Dec 2015. The instruments will include 11 state-of-art mass spectrometers, listed in the first column.

5 CLOUD REQUESTS

In our 2014 report to the SPSC [3], we presented the long-term scientific plans for CLOUD and summarised:

> *"...We estimate that CLOUD will need a further ten years' operation at the CERN PS to carry out the important experimental and modelling programme described here."*

Following its review of the report, the SPSC concluded [5]:

> *"The SPSC congratulates the consortium on the internationally leading results, which were achieved since 2009 . . .*
> *The Committee also recommends CERN to provide adequate support to the collaboration."*

However, despite the recommendation of the SPSC, there are several aspects related to CERN support of CLOUD that could seriously impact future CLOUD operations if they are not resolved. The main items of concern are summarised below.

5.1 T11 beamline

CLOUD has requested in its long-term planning document for the SPSC [3] that the T11 beamline be retained in the East Area upgrade planned for LS2 (long shutdown 2). LS2 will involve a shutdown of all CERN accelerators for a period of 2 years during 2019 and 2020. Maintaining 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. If CLOUD were moved to T9 then not only would less T9 test beam time be available for other users but also their access to the upstream T9 experimental zone would be blocked during CLOUD operation.

The highest priorities for CLOUD operation in future are a) flexibility in the availability of beam time, and b) open access to the experimental area during non-beam periods so that the chamber and support systems can be upgraded and developed for the upcoming run. The scientific potential of CLOUD has proven to be very high and the field of studies still ahead is large. At present CLOUD is the only laboratory chamber in the world where atmospheric aerosol nucleation and growth experiments can be carried out under controlled conditions. To exploit this potential, the CLOUD facility and instruments are re-configured and extended for each new measurement campaign. These modifications are done during non-beam time, and require continual free access to the beam area.

The above requirements are well met in the present T11 beamline. The current beam size and energy are suitable for CLOUD. The T11 experimental zone is small, but well suited for CLOUD. The space currently available on the T10 side of the CLOUD chamber is tight, leading to difficulties in placing measurement instruments and in maintaining the necessary access paths around the chamber. A study has therefore been made to enlarge the T11 experimental zone slightly in the direction towards T10. This is compatible with the East Area beamline upgrades planned for LS2, shown in Fig. 8.

We have recently carried out a preliminary but detailed cost estimate of the resources required for the CLOUD beamline during LS2, under two scenarios:

1. CLOUD stays in T11 (and the experimental zone is enlarged slightly, as shown in Fig. 8):
   - Manpower: 0.5 years FTE
   - Costs: 90 kCHF
2. CLOUD moved to T9:
   - Manpower: 3.1 years FTE
   - Costs: 860 kCHF

Considerably higher costs would be involved in a possible move to T9. The manpower situation is perhaps even more critical. There are no full-time CERN expert staff for CLOUD. During LS2 the CLOUD expert persons will work on many critical LHC tasks, like LHCb and ALICE upgrade installations. In the current manpower plans, the expert manpower needed to execute a move of CLOUD to T9 is not available, and no plan exists for solving this.

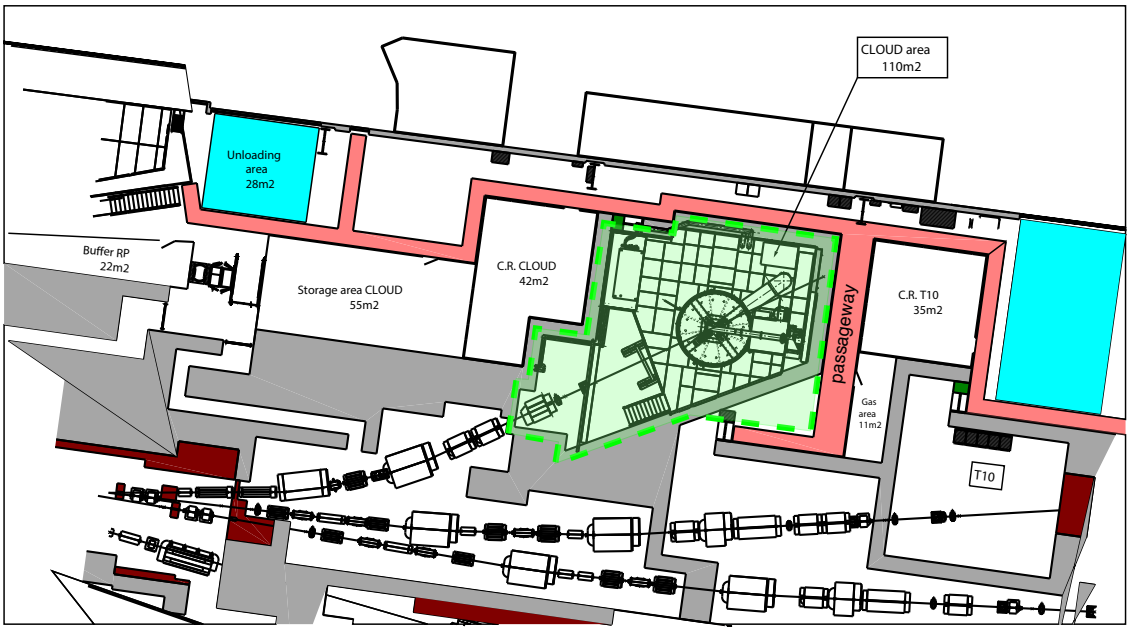

Fig. 8: Proposed T11 experimental zone in the LS2 East Area upgrade (2019–2020). The green shaded area represents the current T11 zone with an extension in the direction towards T10 to provide more space for analysing instruments attached to the CLOUD chamber.

Moreover, a possible move of CLOUD during LS2 would have a major impact on the physics output of the experiment. As during LS1, CLOUD plans to run for physics measurements during LS2, using cosmic rays. Based on CLOUD's previous publications and increasing rate of output, we could anticipate about 15 peer-reviewed publications from data to be obtained during the two-year LS2 period, including about 2 in Nature or Science. These would be lost if CLOUD were moved to T9. Although CLOUD is presently unique, there are serious discussions underway to build a competing chamber elsewhere, so this loss of physics may not be recoverable. And, subsequently, if the T9 beamline were shared with other users, there would be further constraints and limitations on beam time which would negatively impact the productivity of CLOUD.

5.2 Permanent CLOUD open office/meeting room

An additional request made by CLOUD in the long-term planning document to the SPSC [3] concerns a dedicated 50 m^2 open office space and meeting room for CLOUD. No room was available during the CLOUD9 run, Sep–Dec 2014, and so the collaboration had to rent and equip a temporary meeting room (barrack 652) near the East Hall for that period, paid by the CLOUD common fund. In addition, CERN has provided no offices for the five CLOUD research fellows in the CERN team; they are currently using 3 temporary offices borrowed from PH-DT. Furthermore, no offices are provided at CERN for any of the visiting CLOUD experimenters.

CLOUD needs a dedicated 50 m^2 open office space and meeting room for its daily run coordination meetings and for use of the experimenters at CERN, which number up to 30 or more during runs at the PS. An example of a suitable office is shown in Fig. 9. For radiation protection reasons this open office/meeting room cannot be inside the East Hall. However it needs to be close to the T11 experimental zone where all the work takes place. The CLOUD run team is required to do daily setup, calibration and modification work continually throughout the measurement campaign. The meeting room needs adequate video-conferencing equipment to allow for on-line participation of researchers at CLOUD institutes in the run coordination meetings.

Fig. 9: Example of a meeting room/open office suitable for the CLOUD collaboration. The photograph shows bat. 3893, made from two pre-fabricated units (full dimensions 6×7.5 m^2) at the LHCb site (LHC Point 8). A similar office, located near the T11 experimental zone, would be suitable for CLOUD.

The CLOUD meeting room/open office would be a multi-purpose room serving the following functions:

- Daily data analysis and run coordination meetings during data taking at the CERN PS.
- Weekly technical planning and physics meetings for the CERN team throughout the year, including audio-visual communications with the CLOUD Analysis Working Group members in other institutes.
- An open office for the CERN CLOUD team.
- Open office space for CLOUD experimenters during experimental runs at the CERN PS. Without adequate space in the proximity of T11, the experimenters use the T11 CLOUD counting room which, for radiation protection reasons, should not be used for this purpose.

Despite continual requests over several years, there has been no progress on CERN providing a dedicated open office/meeting room for CLOUD. We currently have no solution for a meeting room near the East Hall for the CLOUD10 run, Sep–Dec 2015. This will negatively impact CLOUD operations. We urgently request that CERN finds a permanent solution for a dedicated CLOUD open office/meeting room under its responsibilities as host laboratory, as stated in the General Conditions Applicable to Experiments at CERN:

> *"5.8. CERN normally provides, free of charge and within the limits and constraints imposed by the available resources and schedules of accelerators, the following standard services and facilities for the duration of the Experiment:*
> ...
> *g) office space, equipped with standard furniture and infrastructure facilities including network connections, telephones and electricity."*

5.3 CERN technical support staff for CLOUD

The CERN CLOUD team is responsible for the maintenance and development of the CLOUD facility. In addition the CERN team is responsible for a wide range of support tasks required for the operation and data taking of the facility by the CLOUD researchers: technical and resource coordination, safety matters, host institute services (technical, logistics, administrative), handling of the analysing instruments from the CLOUD institutes (reception, crane work, operational support needs, removal and shipping of instruments back to the institutes) and gas system support. In addition the CERN CLOUD team is responsible for guiding visiting groups to CLOUD, which currently number about 30 per year.

CLOUD is a "living detector" that is continually improved and upgraded to extend its physics reach as new knowledge is obtained. Each run at the PS involves a completely new configuration of analysing instruments and places new demands on the technical performance of the CLOUD facility and the range of gases under study. So far the small CERN CLOUD team has managed to carry out these tasks, but the situation is becoming critical. If CLOUD does not receive adequate technical support from CERN in future then there is a real danger that CLOUD will be unable to carry out its planned scientific programme.

The CERN staff on CLOUD has recently fallen by 1 FTE and now totals 1.0 FTE, comprising:

- 0.3 FTE engineer (PH-DT).
- 0.4 FTE technician (PH-DT).
- 0.3 FTE (EN-MME) - but uncertain to continue.

Within PH-DT there is a direct conflict of these staff with the needs for the LHC experiment upgrades, so there is great pressure even on this extremely low level of technical support staff. CLOUD needs a minimum of 2 FTE CERN technical support staff just to prepare CLOUD for each run at the PS, without even considering detector upgrades.

Acknowledgements

We would like to thank CERN PH-DT, EN-MME, Lau Gatignon, the PS Coordinator and the CERN PS machine team for their support of CLOUD.

References

[1] IPCC, Climate Change 2013 - The Physical Science Basis. Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press (2014).

[2] CLOUD Collaboration. A study of the link between cosmic rays and clouds with a cloud chamber at the CERN PS.
CERN-SPSC-2000-021; SPSC-P-317 (2000), <http://cds.cern.ch/record/444592>
CERN-SPSC-2000-030; SPSC-P-317-Add-1 (2000), <http://cds.cern.ch/record/462623>
CERN-SPSC-2000-041; SPSC-P-317-Add-2 (2000), <http://cds.cern.ch/record/497173>
CERN-SPSC-2006-004; SPSC-P-317-Add-3 (2006). <http://cds.cern.ch/record/923140>

[3] CLOUD Collaboration. CLOUD status and long-term plans.
CERN-SPSC-2014-018; SPSC-P-317-Add-4 (2014), <http://cds.cern.ch/record/1706253>

[4] Andreae, M.O. The aerosol nucleation puzzle. *Science* **339**, 911–912 (2013).

[5] CERN SPSC. Minutes of the 114^{th} meeting of the SPSC. CERN-SPSC-2014-024 SPSC-114 (June 2014).

[6] Duplissy, J., *et al.* Results from the CERN pilot CLOUD experiment. *Atmos. Chem. Phys.* **10**, 1635–1647 (2010).

[7] Kupc, A., *et al.* A fibre-optic UV system for H_2SO_4 production in aerosol chambers causing minimal thermal effects. *J. Aerosol Sci.* **42**, 532–543 (2011).

[8] Kirkby, J., *et al.* Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation. *Nature* **476**, 429–433 (2011).

[9] Voigtländer, J., *et al.* Numerical simulations of mixing conditions and aerosol dynamics in the CERN CLOUD chamber. *Atmos. Chem. Phys.* **12**, 2205–2214 (2012).

[10] Bianchi, F., *et al.* On-line determination of ammonia at low pptv mixing ratios in the CLOUD chamber. *Atmos. Meas. Tech.* **5**, 1719–1725 (2012).

[11] Praplan, A.P., *et al.* Dimethylamine and ammonia measurements with ion chromatography during the CLOUD4 campaign. *Atmos. Meas. Tech.* **5**, 2161–2167 (2012).

[12] Keskinen, H., *et al.* Evolution of particle composition in CLOUD nucleation experiments. *Atmos. Chem. Phys.* **13**, 5587–5600 (2013).

[13] Almeida, J., *et al.* Molecular understanding of sulphuric acid-amine particle nucleation in the atmosphere. *Nature* **502**, 359–363 (2013).

[14] Schobesberger, S., *et al.* Molecular understanding of atmospheric particle formation from sulfuric acid and large oxidized organic molecules. *Proc. Natl. Acad. Sci.* **110**, 43, 17223–17228 (2013).

[15] Riccobono, F., *et al.* Oxidation products of biogenic emissions contribute to nucleation of atmospheric particles. *Science* **344**, 717–721 (2014).

[16] Kürten, A., *et al.* Neutral molecular cluster formation of sulfuric acid-dimethylamine observed in real time under atmospheric conditions. *Proc. Natl. Acad. Sci.* **111**, 15019–15024 (2014).

[17] Schnitzhofer, R., *et al.* Characterisation of organic contaminants in the CLOUD chamber at CERN. *Atmos. Meas. Tech.* **7**, 2159–2168 (2014).

[18] Bianchi, F., *et al.* Insight into acid-base nucleation experiments by comparison of the chemical composition of positive, negative, and neutral clusters. *Environ. Sci. Technol.* **48**, 13675–13684 (2014).

[19] Schobesberger, S., *et al.* On the composition of ammonia–sulfuric-acid ion clusters during aerosol particle formation. *Atmos. Chem. Phys.* **15**, 55–78 (2015).

[20] Kürten, A., *et al.* On the derivation of particle nucleation rates from experimental formation rates. *Atmos. Chem. Phys.* **15**, 4063–4075 (2015).

[21] Praplan, A.P., *et al.* Elemental composition and clustering behaviour of α-pinene oxidation products for different oxidation conditions. *Atmos. Chem. Phys.* **15**, 4145–4159 (2015).

[22] Franchin, A., *et al.* Experimental investigation of ion-ion recombination at atmospheric conditions. *Atmos. Chem. Phys. Discuss.* **15**, 3667–3702 (2015).

[23] Kürten, A., *et al.* Thermodynamics of the formation of sulfuric acid dimers in the binary $(H_2SO_4{-}H_2O)$ and ternary $(H_2SO_4{-}H_2O{-}NH_3)$ system. *Atmos. Chem. Phys. Discuss.* **15**, 13957–14006 (2015).

[24] Lehtipalo, K., *et al.* Towards understanding the early steps of atmospheric nano-particle growth. (submitted, 2015).

[25] Duplissy, J., *et al.* Effect of ions on sulfuric acid-water binary particle formation II: Experimental data 1 and comparison with QC-normalized classical nucleation theory. (submitted, 2015).

[26] Ehrhart, S., *et al.* Comparison of the SAWNUC model with CLOUD measurements of sulphuric acid-water nucleation. (submitted, 2015).

[27] Dunne, E., *et al.* Global aerosol formation from sulfuric acid, ammonia, organics and ions. (to be submitted, 2015).

[28] Kürten, A., *et al.* Experimentally determined particle formation rates over a wide range of tropospheric conditions: influence of sulfuric acid, ammonia, ions, and temperature. (to be submitted, 2015).

[29] Almeida, J., *et al.* Parameterisation of neutral and charged binary and ternary nucleation rates from 250 to 310 K. (to be submitted, 2015).

[30] Kirkby, J., *et al.* Biogenic nucleation. (to be submitted, 2015).

[31] Rondo, L., *et al.* Effect of dimethylamine on the gas phase sulfuric acid concentration measured by Chemical Ionization Mass Spectrometry (CIMS). (to be submitted, 2015).

[32] Kim, J., *et al.* Hygroscopicity of nanoparticles produced from homogeneous nucleation in the CLOUD experiment. (to be submitted, 2015).

[33] Tröstl, J., *et al.* Organic aerosol particle growth. (to be submitted, 2015).

[34] Hoyle, C., *et al.* Aqueous phase oxidation of sulphur dioxide by ozone in cloud droplets. (to be submitted, 2015).

[35] Järvinen, E., *et al.* Observation of viscosity transition in secondary organic aerosol. (to be submitted, 2015).