EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

CERN-SPSC-2016-028
SPSC-SR-192
June 14, 2016

2015 PROGRESS REPORT ON PS215/CLOUD

CLOUD Collaboration

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CERN-SPSC-2016-028 / SPSC-SR-192
14/06/2016

1 SCIENCE HIGHLIGHTS

The CLOUD experiment at CERN [1, 2] is investigating under atmospheric conditions the formation and growth of aerosol particles, and the chemistry and microphysics of liquid and ice clouds. A unique focus of the experiment is to evaluate the influence of galactic cosmic rays on aerosols, clouds and climate. The impact of the experimental measurements on climate are evaluated with a state-of-art global aerosol model. In turn, the global model, together with tightly-integrated field measurements, help to guide the choice and range of variables for experimental study by CLOUD.

The experiments conducted so far have mainly focused on atmospheric aerosol nucleation and growth involving various chemical systems, and how these processes are influenced by galactic cosmic rays. In addition, two runs (CLOUD8, 2013, and CLOUD9, 2014) were largely devoted to 'cloudy" experiments where the chemistry and microphysics of liquid and ice clouds were investigated, including ion-related processes. The results have been published in more than forty journal articles, including eight high-impact publications: four letters in *Nature* (Kirkby *et al.*, 2011; Almeida *et al.*, 2013; Kirkby *et al.*,, 2016, Tröstl *et al.*, 2016), one in *Science* (Riccobono *et al.*, 2014), two articles in the *Proceedings of the National Academy of Sciences of the United States of America* (Schobesberger *et al.*, 2013; Kürten *et al.*,

2014), and one in *Nature Communications* (Lehtipalo *et al.*, 2016). The *Nature* article by Kirkby *et al.*, 2011, has been cited over 320 times and is in the 96^{th} percentile of all *Nature* articles of a similar age.

During 2015–2016, sixteen CLOUD articles so far have been published in journals [3]–[18], of which four report the first results from the liquid and ice cloud experiments [15]–[18]. Four further articles are currently under review [19]–[22]. Some highlights from these publications are presented below.

The major discovery reported in *Nature* in May 2016 (Kirkby *et al.*) is that highly oxidised biogenic vapours produce abundant particles in the atmosphere in the absence of sulphuric acid. The highly oxygenated molecules (HOMs) appear rapidly in the CLOUD chamber upon exposure of α-pinene to ozone (Fig.1). Previously it was thought that sulphuric acid—which largely arises from sulphur dioxide emitted by fossil fuels—was essential to initiate particle formation. CLOUD found that ions from galactic cosmic rays strongly enhance the production rate of pure biogenic particles, by a factor 10-100 compared with particles without ions (Fig.2). CLOUD also showed (Tröstl *et al.*, *Nature*, 2016) that oxidised biogenic vapours dominate particle growth in unpolluted environments, starting just after the first few molecules have clustered together and continuing all the way up to sizes above 50-100 nm where the particles can seed cloud droplets. The growth rate was quantitatively explained with a model of organic condensation.

Ion-induced nucleation of pure biogenic particles may have important consequences for pristine climates since it provides a hitherto-unknown mechanism by which nature produces particles without pollution. CLOUD found that, once embryonic biogenic particles have formed, related but more abundant oxidised biogenic vapours cause particle growth to accelerate. Rapid growth of the new particles while they are still small and highly mobile implies a larger fraction will avoid coagulation with pre-existing larger particles and eventually reach sizes where they can seed cloud droplets and influence climate. There are at least two important consequences for the climate. First, pure biogenic nucleation and growth may raise the baseline aerosol state of the pristine pre-industrial atmosphere and so may reduce the estimated anthropogenic radiative forcing from increased aerosol-cloud albedo over the industrial period. Second, pure biogenic nucleation shows the highest sensitivity of all nucleation processes so far studied by CLOUD and may potentially shed new light on the long-standing question of a physical mechanism for solar-climate variability in the pristine pre-industrial climate.

The first publications from CLOUD's liquid and ice cloud experiments include liquid-phase chemistry, where fast uptake and oxidation of SO_2 was measured in super-cooled cloud droplets (Hoyle *et al.*, 2016). The formation of glassy (highly viscous) biogenic aerosol particles was also studied in experiments at various temperatures down to -38°C and increasing relative humidity. The glassy transition of aerosol particles was measured *in situ* with the scattering depolarisation instrument, SIMONE, developed by KIT (Järvinen *et al.*, 2016). In addition, the ice activation properties of glassy biogenic particles were studied using another newly-developed instrument: the ice nucleus counter SPIN (Ignatius *et al.*, 2016).

2 CLOUD10, 28 Sep – 12 Dec 2015

CLOUD10 was the most ambitious run undertaken so far, involving almost 40 instruments attached to the CLOUD chamber, of which 11 were mass spectrometers (Figs. 3 and 4). New features of the CLOUD facility included the use of several new gases as well as an 8W, 248 nm KrF UV excimer laser and a UVA LED light source (Fig. 5). The UV laser extended the performance of CLOUD to reproduce atmospheric concentrations of the climatically-important hydroxyl $(OH\cdot)$ radical. Several new analysing instruments were successfully operated at CLOUD10 (an example is shown in Fig. 6).

The primary aim of the CLOUD10 run was to simulate aerosol particle nucleation and growth at the Hyytiälä boreal forest field station under daytime and night-time conditions, involving mixtures of gases that included SO_2, O_3, α-pinene, δ3-carene, NH_3, NO, NO_2 and H_2O. This was largely successfully achieved, except that the available UVA power was insufficient to generate atmospheric NO_2

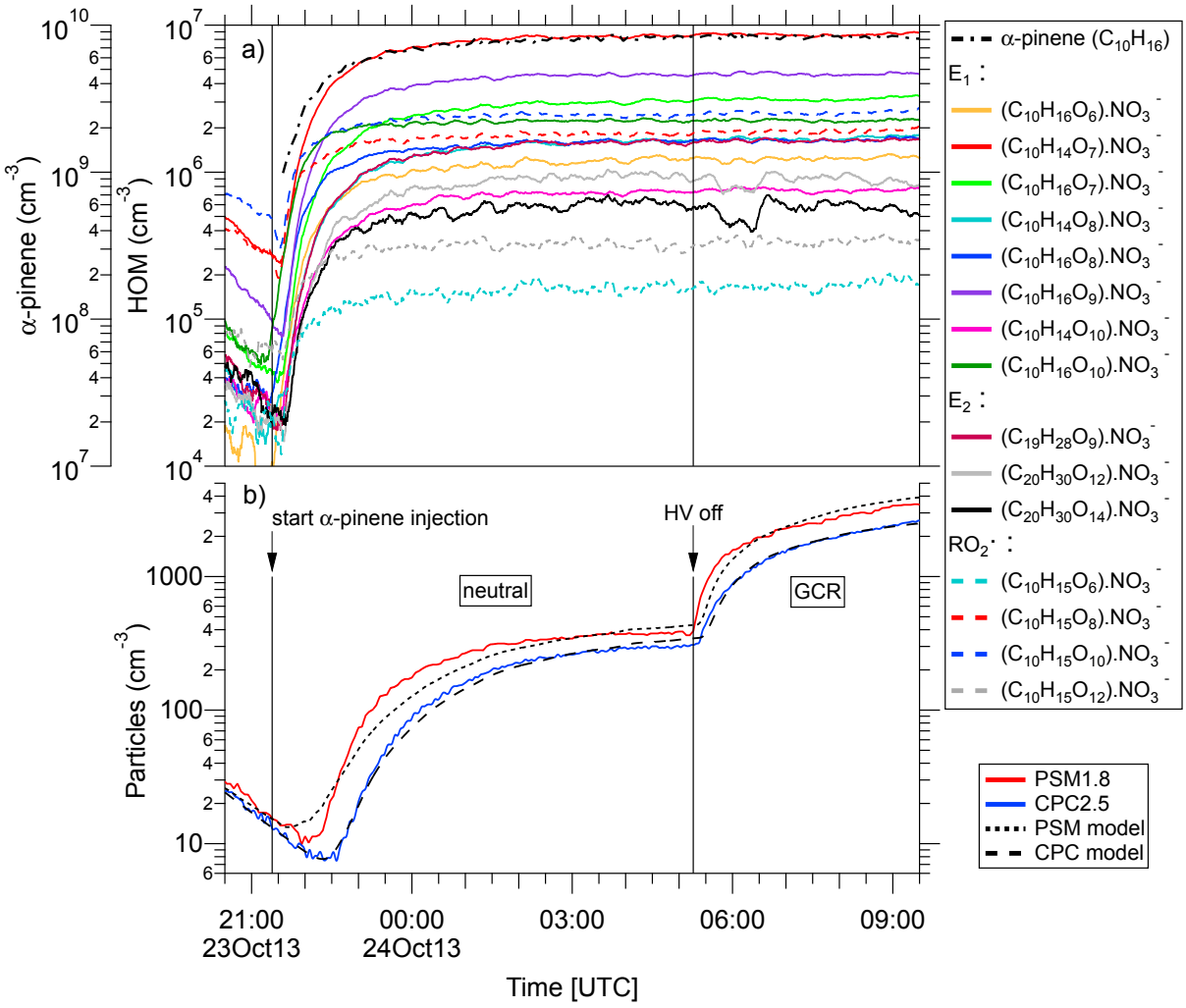

Fig. 1: Evolution of HOMs and particles during a typical run. a) Evolution of selected HOM monomers (E_1), dimers (E_2) and peroxy ($RO_2\cdot$) radicals at 300 pptv α-pinene, 33 ppbv O_3, zero H_2 or HONO, 38% RH, 278 K, and $[H_2SO_4]$ below 5×10^4 cm^{-3}. The HOMs start to appear soon after first injection of α-pinene into the chamber at 21:22, 23 October. A HOM monomer is a highly oxygenated molecule derived from α-pinene ($C_{10}H_{16}$), and a HOM dimer is a covalently-bound pair of monomers. Peroxy radicals are identified by an odd H number. The HOMs are charged with an NO_3^- ion in the CI-APi-TOF mass spectrometer. b) Evolution of the particle number concentrations measured in the PSM1.8 (red curve) and CPC2.5 (blue curve) particle counters. The HV clearing field was switched off at 05:16, 24 October, marking the transition from neutral (ion-free) to GCR conditions in the chamber. A sharp increase in the rate of particle formation is seen, due to ion-induced nucleation of pure biogenic particles. However, no change occurs in the HOM concentrations (panel a) since these are predominantly neutral gas-phase molecules. The dotted and dashed curves in panel b show the PSM1.8 and CPC2.5 distributions, respectively, simulated for this run with the AEROCLOUD kinetic model, which is used to derive the experimental nucleation rates.

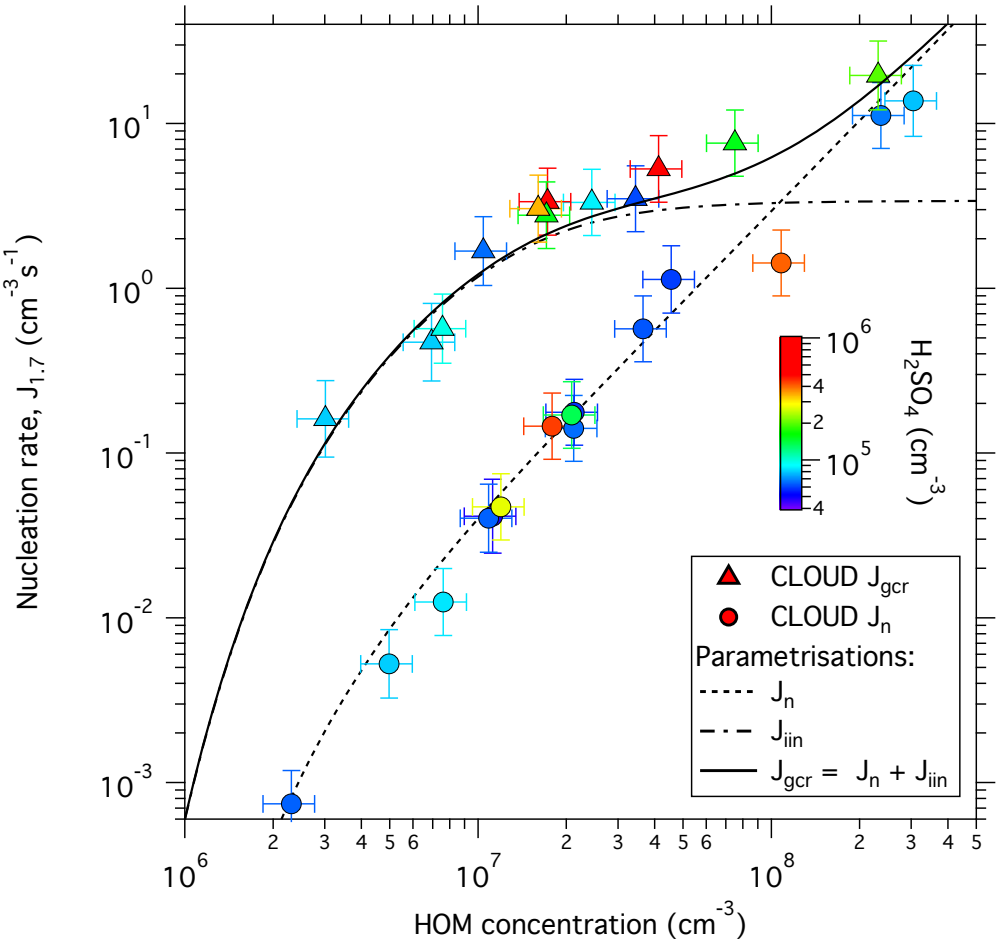

Fig. 2: Pure biogenic nucleation rates against HOM concentration (1 pptv = 2.4×10^7 cm^{-3}). Neutral (J_n; circles) and GCR (J_{gcr}; triangles) nucleation rates versus total HOM concentration, [$RO_2\cdot$ + E_1 + E_2]. The experimental conditions are 10–1300 pptv α-pinene (for measurements below $J_{1.7}$ = 10 cm^{-3}s^{-1}), 30–35 ppbv O_3, zero H_2 or HONO, 38% RH, 278 K, and $< 8 \times 10^5$ cm^{-3} H_2SO_4. The colour scale shows [H_2SO_4]; purple and blue points correspond to contaminant-level (below the detection threshold); other colours correspond to measurements after SO_2 was added to the chamber. The fitted curves show parametrisations for J_n (dashed), J_{gcr} (solid) and ion-induced nucleation (J_{iin} = J_{gcr} - J_n; dot-dashed). The J_{iin} parametrisation assumes that the nucleation rate falls steeply at HOM concentrations below the experimental measurements, following a similar slope to that for J_n.

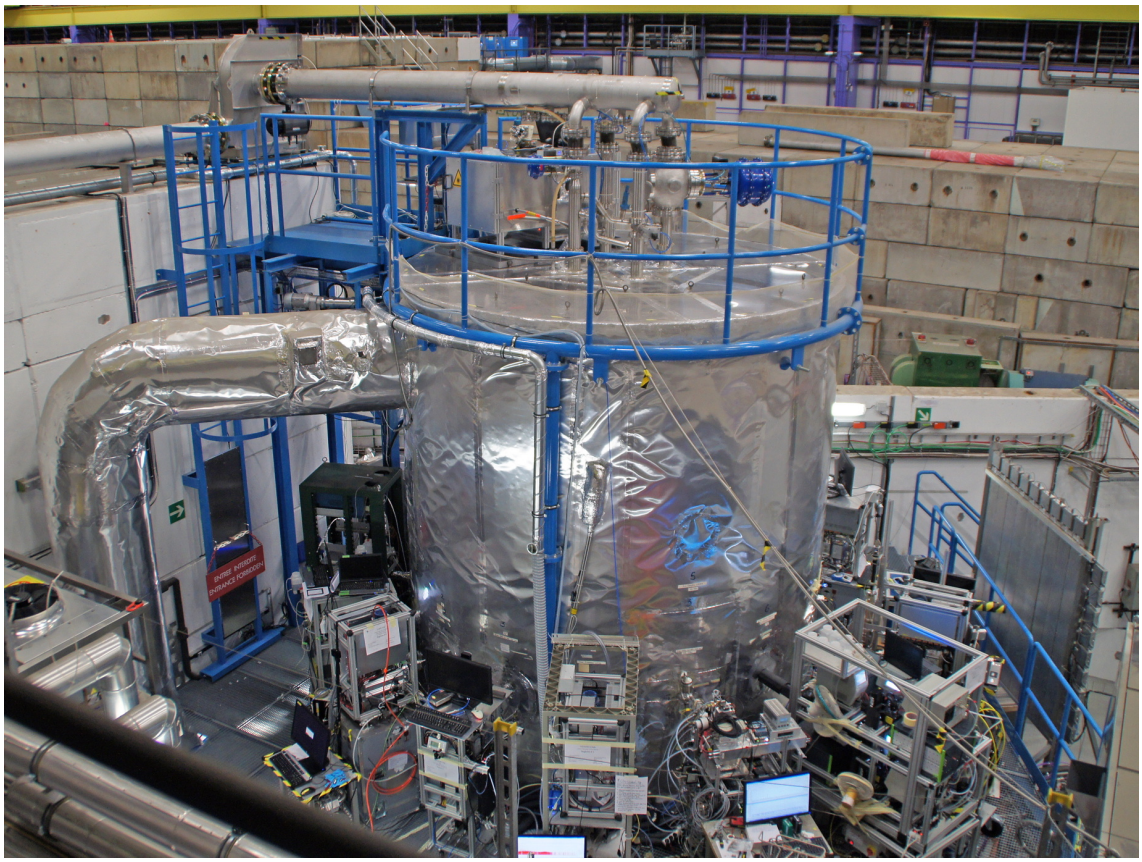

Fig. 3: CLOUD10. CLOUD with analysing instruments in the East Hall, November 2015.

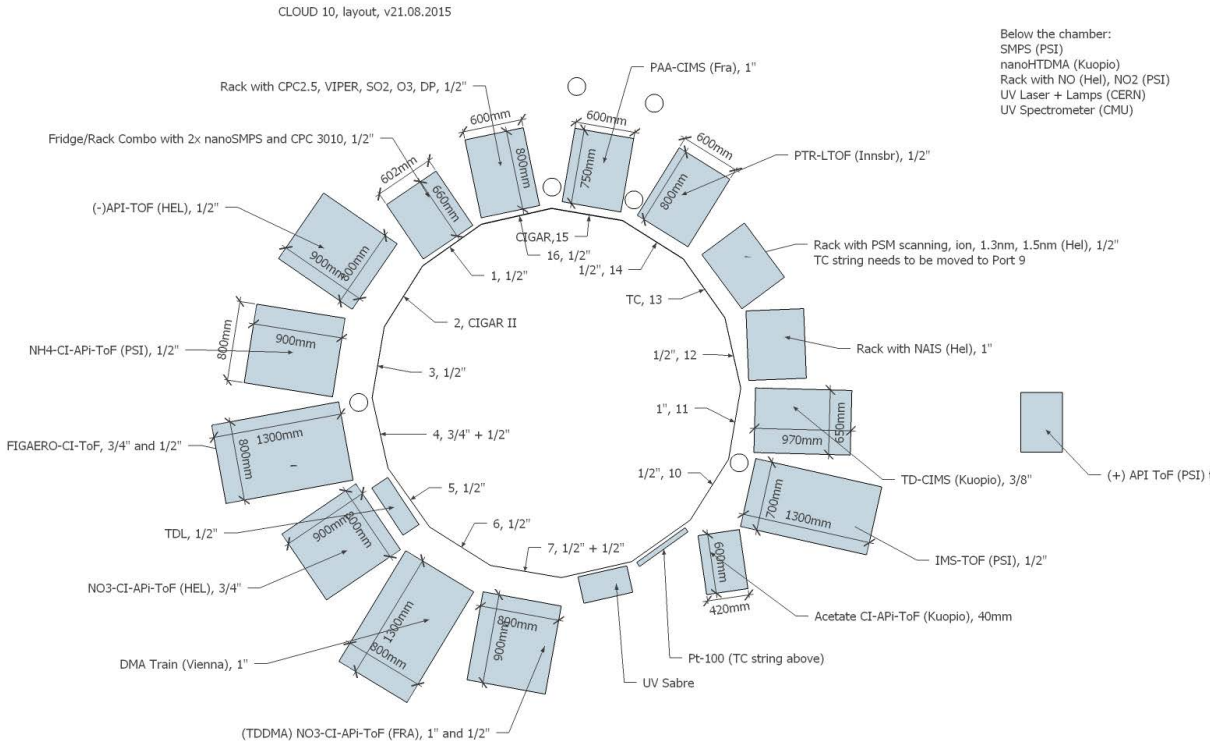

Fig. 4: CLOUD10 layout. Layout of the analysing instruments around the chamber during CLOUD10.

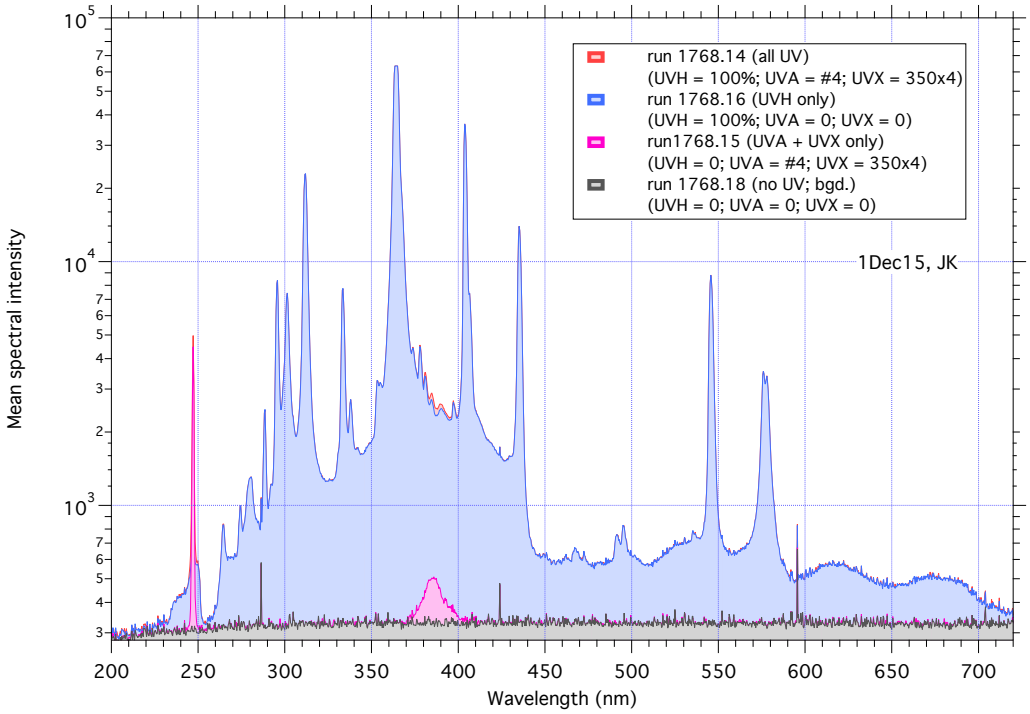

Fig. 5: CLOUD10 UV spectral intensity. The spectral intensity from the three UV sources in operation during CLOUD10: Hamamatsu Hg-Xe lamp system (blue curve, UVH), KrF excimer laser (pink curve at 248 nm, UVX) and UVA LED (pink curve at 385nm, UVA). The grey curve indicates the spectrometer background for the UVH curve. The UV spectrometer has different acceptances for each UV source so the relative pulse sizes are arbitrary.

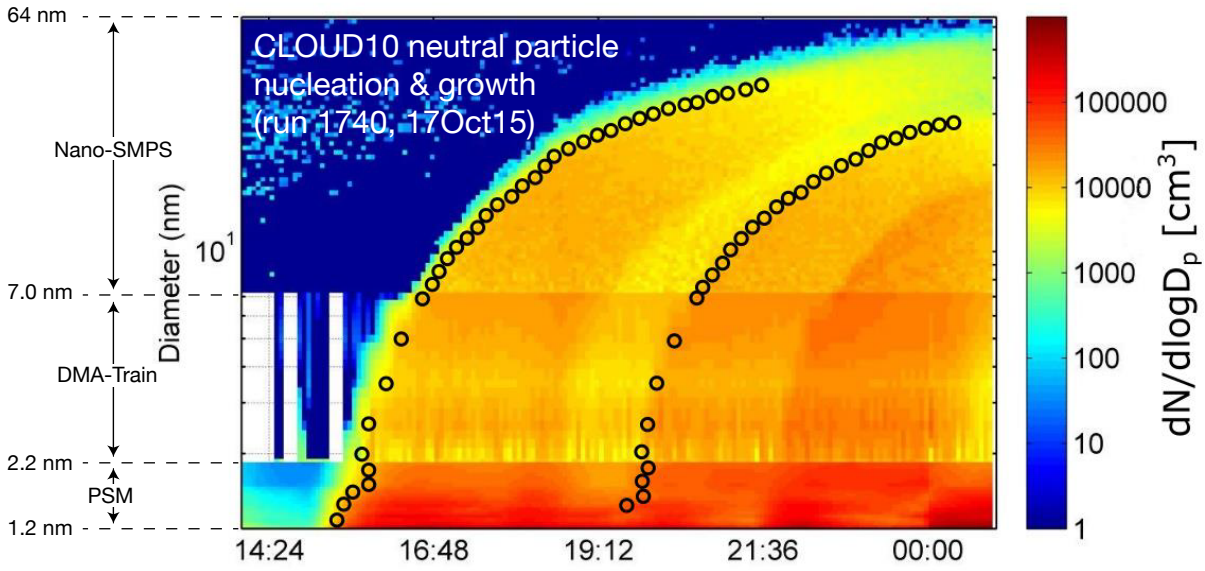

Fig. 6: CLOUD10, first continuous size measurement of neutral particle formation and growth. The figure shows the first time a continuous size versus time measurement has been measured for freshly-nucleated *neutral* particles. The important and previously-inaccessible region between 2 nm and 7 nm diameter is successfully measured by the new Vienna DMA train.

photolysis rates. A new 200 W UVA source has therefore been designed and is under construction for CLOUD11. Systematic measurements were carried out over the atmospheric range of vapour concentrations, beam intensities and boundary layer temperatures to parametrise H_2SO_4/biogenic nucleation for evaluation of its climate impact using the Leeds global aerosol model, GLOMAP. Other studies in CLOUD10 included short periods devoted to studying isoprene (C_5H_8) oxidation chemistry, and the effect of ions, as well as a first look at particle production from anthropogenic volatile organic compounds in preparation for CLOUD11.

3 CLOUD MEETINGS, 2015–2016

The following CLOUD Collaboration meetings and data workshops were held in 2015–2016:

CLOUD7–9 data workshop, Paul Scherrer Institute, 2–6 Feb 2015. Analysis of data from CLOUD7–9.

CLOUD-TRAIN summer school, Cacais, Portugal, 7–13 Jun 2015. Summer school for young CLOUD scientists given by CLOUD PIs and invited international experts.

CLOUD collaboration meeting, CERN, 21–23 Oct 2015. CLOUD collaboration meeting and Finance Review Committee meeting (FRC4).

CLOUD10 data workshop, Hyytiälä Forestry Field Station, Finland, 1–4 Feb 2016. Analysis of data from CLOUD10.

CLOUD-TRAIN final conference, Königstein, Germany, 14–17 Jun 2016. Open presentation and discussion of latest unpublished CLOUD results by young scientists, together with recent results from 20 invited external experts from Europe and the United States, followed by a half-day closed CLOUD collaboration meeting.

4 CLOUD11 BEAM REQUEST, 27 Sep – 21 Nov 2016

The CLOUD11 run is planned for 27 Sep – 21 Nov 2016, with the final week using cosmic rays (no East Area beams). The aims of the CLOUD11 run are:

1. **Anthropogenic aerosol particle nucleation and growth.** Three anthropogenic volatile organic compounds will be studied: toluene (methyl benzene, $C_6H_5.CH_3$), 1,2,4 trimethylbenzene ($C_6H_3.3(CH_3)$) and naphthalene ($C_{10}H_8$). Nucleation and growth will be studied under polluted urban conditions (high concentrations of NO_x, ozone, O_3, and hydroxyl radicals, OH·).

2. **Pure biogenic aerosol particle nucleation and growth.** The interplay of three vapours will be investigated: isoprene (C_5H_8), the monoterpene, α-pinene ($C_{10}H_{16}$), and the sesquiterpene, β-caryophyllene ($C_{15}H_{24}$). These experiments follow up on CLOUD's discovery of pure biogenic nucleation reported in *Nature* (Kirkby *et al.*, 2016), which involved α-pinene alone and showed a large ion enhancement (factor 10–100). The study of pure biogenic nucleation will be extended beyond the CLOUD10 measurements to include mixtures of these three biogenic vapours, as found in the atmosphere, as well as the influence of NO_x, oxidant concentrations (O_3 and OH·) and temperature, under various ion conditions (neutral, GCR and π beam). These measurements will allow a more robust evaluation to be made of the atmospheric importance of pure biogenic nucleation, using the Leeds global aerosol model, GLOMAP.

Several improvements in the CLOUD detector will be made with respect to CLOUD10. These include a factor 10 increase in NO_2 photolysis rate with the UV sabre 3 (200 W optical power at 385 nm). Several new and improved mass spectrometers will also be available for CLOUD11, such as a HO_x/RO_x mass spectrometer, an NH_3 mass spectrometer and a long-TOF (high resolution) FIGAERO-CI-APiLTOF for analysing the precise chemical species in freshly-nucleated aerosol particles.

5 CLOUD REQUESTS

5.1 Permanent CLOUD open office/meeting room

CLOUD has requested in its long-term planning document to the SPSC [2] a dedicated 50 m^2 open office space and meeting room. CLOUD needs this room for its daily run coordination meetings and for use by CLOUD experimenters at CERN, which number up to 30 or more during runs at the PS. No offices have been provided for CLOUD research fellows in the CERN team or for any of the visiting CLOUD experimenters. The meeting room is therefore also needed as an open office for CLOUD researchers.

No meeting room was available during the CLOUD9 run (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. For the CLOUD10 run (2015), a temporary meeting room in bat. 510 was provided by EP. The same temporary meeting room will be used for the CLOUD11 run in fall 2016.

We request that EP actively finds a permanent solution this long-standing problem of the lack of a dedicated open office/meeting room for CLOUD. The room needs to be close to the T11 experimental zone since the CLOUD run team is required to do daily setup, calibration and modification work continually throughout the run period.

5.2 CERN support staff for CLOUD

The CERN CLOUD team is responsible for the maintenance and development of the CLOUD facility. 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.

The CERN technical support for maintenance and development of the CLOUD facility has recently improved a little (increasing from 1.0 to 1.2 FTE, made up from 3 part-time CERN staff), but it remains at the limit of viability.

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), coordinating the analysing instruments from the CLOUD institutes (instrument configuration and platform layout for each campaign, special gas requirements, radioactive sources, instrument safety aspects, instrument reception, crane work, operational support needs, removal and shipping of instruments back to the institutes) and gas system support. In addition the CLOUD DAQ system is an integral part of the CLOUD experiment and CERN has a *de facto* responsibility to ensure the DAQ system is properly staffed.

To carry out these responsibilities, in addition to technical support, CLOUD needs at least 2 fellows in the CERN team: a) a research fellow to coordinate the analysing instruments for each campaign, and related tasks, and b) a technical fellow to take responsibility for the CLOUD DAQ and control systems, and instrument integration for each campaign. So far, these CERN-based fellows have been funded by two successful EU Marie Curie ITNs. The second of our EU ITNs is now about to end, and unfortunately CLOUD was not successful with its third ITN proposal, submitted in 2016.

We therefore request that the CERN Fellows and Associates Committee will agree in principle to consider young researchers with an atmospheric research background as candidates for a CERN research fellowship, and that they be judged on an equal basis of excellence alongside particle physics candidates. We also request that CLOUD be assigned a technical fellows position so that a suitable young DAQ expert can be hired to take responsibility for the CLOUD DAQ system.

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.

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