



2016 PROGRESS REPORT ON PS215/CLOUD

CLOUD Collaboration

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1 SCIENCE HIGHLIGHTS

Atmospheric aerosol nucleation is poorly understood yet has a major influence on climate change. Aerosols and their effect on clouds are the largest source of uncertainty in anthropogenic radiative forcing of climate [1]. Ion-induced nucleation may also shed new light on the long-standing puzzle of a physical mechanism for solar-climate variability in the pristine pre-industrial climate [2, 3]. Particle nucleation and growth (condensation) is also a major source of fine urban smog particles (PM_{2.5}), which are a significant contributor to mortality in urban environments. Life expectancy is reduced by 0.6 y per 10 $\mu\text{g}/\text{m}^3$ increase in PM_{2.5} [4]. Concentrations in the range 10–25 $\mu\text{g}/\text{m}^3$ are typical for European cities, and far higher annual means near 100 $\mu\text{g}/\text{m}^3$ are typical for Chinese mega-cities.

CLOUD [5, 6] is the world's leading experiment for laboratory studies of atmospheric aerosol nucleation and growth. Unique features include condensable contaminants well below 1 pptv, control of ionisation throughout tropospheric conditions (π beam, ground-level galactic cosmic rays - GCRs, ion-free conditions with a HV clearing field), precise control of all experimental conditions (trace precursor vapour concentrations, relative humidity, UV, etc.), highly stable operation at any temperature in the range -75°C to 50°C , and the ability to create liquid or ice clouds from aerosol particles nucleated and grown in the chamber and then study their chemistry and microphysics. Moreover, in a break from



the normal approach of global models—where they are separated from experiments and at the end of the research chain—CLOUD brings together experimentalists and modellers into a single integrated project. CLOUD is pioneering an innovative synergistic approach in which the model simulations guide the experiments as well as exploit the experimental results. The outcome is a state-of-the-art global aerosol model, GLOMAP, that is grounded on laboratory-measured physical and chemical processes.

So far during 2016–2017, seventeen CLOUD articles [11]–[27] have been published in journals, of which two are in *Nature* [18, 19], one in *Science* [26], one in *Nature Communications* [17] and one in the *Proceedings of the National Academy of Sciences* [22]. A further four manuscripts [28]–[31] are currently under review. Some highlights from these publications are presented below.

A major CLOUD discovery reported in *Nature* [18] is that highly oxydised 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 the abundant biogenic monoterpene, α -pinene ($C_{10}H_{16}$), to ozone. 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. CLOUD also reported in a second paper in *Nature* [19] 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 pure biogenic nucleation is important for our understanding of climate since it provides a hitherto-unknown mechanism by which nature produces particles without pollution. And, once embryonic particles have formed, related but more abundant oxidised biogenic vapours cause the 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. In 2016 CLOUD reported in *Proc. Natl. Acad. Sci.* [22] that pure biogenic nucleation raises the baseline aerosol state of the pristine pre-industrial atmosphere and so reduces estimates of anthropogenic radiative forcing (by 27% in our global model) due to increased aerosol-cloud albedo over the industrial period (Fig. 1).

In a comprehensive study published in *Science* [26], we built a global model of aerosol formation using CLOUD-measured nucleation rates involving sulphuric acid, ammonia, ions and organic molecules. Although sulphuric acid has long been known to be important for nucleation, our results show for the first time that observed concentrations of particles throughout the atmosphere can be explained only if additional molecules—organic compounds or ammonia—participate in nucleation. The model results compare well with atmospheric observations (Fig. 2). The results also show that ionisation of the atmosphere by cosmic rays accounts for nearly one-third of all particles formed, although small changes in cosmic rays over the solar cycle do not affect aerosols enough to influence today’s polluted climate significantly. During CLOUD12 we plan to collect the necessary data to extend this parameterisation and global model study to include multi-component nucleation involving highly oxygenated biogenic vapours (H_2SO_4 – NH_3 – HOM – H_2O , where HOM refers to Highly Oxygenated Molecules) (§5).

CLOUD finds that ions are important for nucleation for almost all chemical systems studied so far [7, 8, 18, 26]. The notable exception is amine base-stabilised sulphuric acid nucleation, which forms highly stable neutral particles at a rate near the kinetic limit [9, 10], so ions add little useful additional stability to the nucleating clusters. Pure biogenic particles are the least stable (i.e. have the highest evaporation rates) measured so far by CLOUD and so have the highest ion enhancement factors (10–100 at GCR intensities). Ion-induced pure biogenic nucleation was especially important in the pristine pre-industrial climate (Fig. 1) and may bear on the question of the mechanism for solar-climate variability. CLOUD is currently evaluating the climate impact of ion-induced pure biogenic nucleation in more realistic environments than the pure α -pinene system where it was discovered [18] (§2.2).

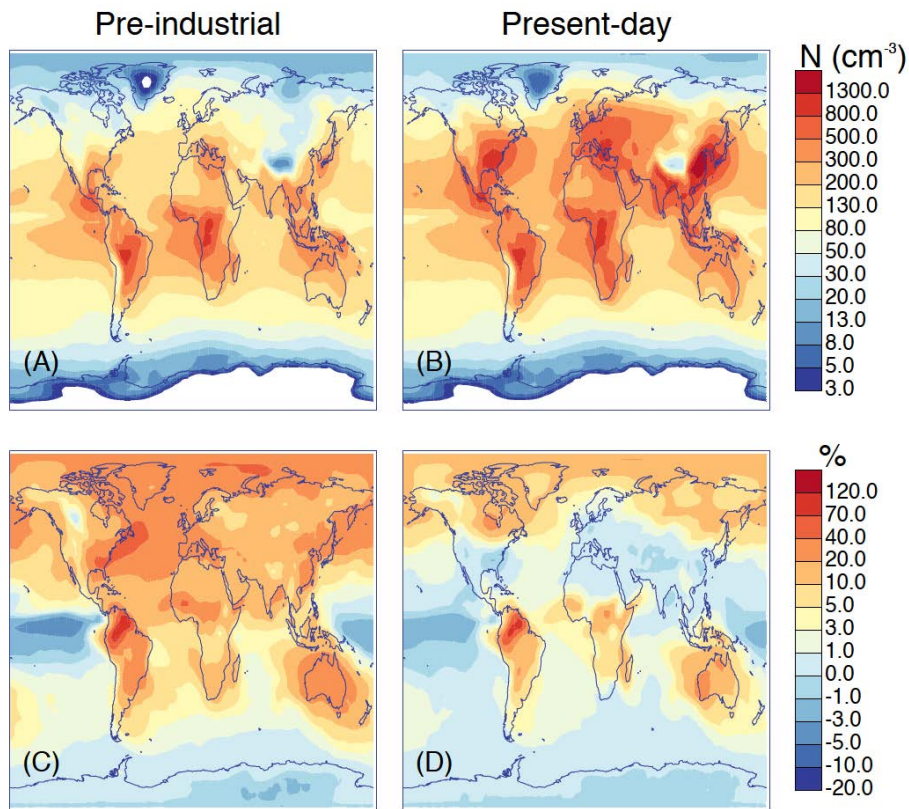


Fig. 1: Impact of pure biogenic nucleation on pre-industrial and present-day climates [22]. Modelled concentrations of cloud condensation nuclei (CCN) are shown at 0.2% supersaturation, annually averaged at cloud base level in A) pre-industrial and B) present-day climates, after including pure biogenic nucleation. The percentage changes in CCN concentrations when pure biogenic nucleation is introduced are shown in C) pre-industrial and D) present-day climates. The model assumes pure biogenic nucleation proceeds at the rates measured in Kirkby *et al.*, 2016 [18]. Pure biogenic nucleation is seen to have a larger effect in the pristine pre-industrial atmosphere, thereby changing the baseline from which anthropogenic aerosol radiative forcing is evaluated.

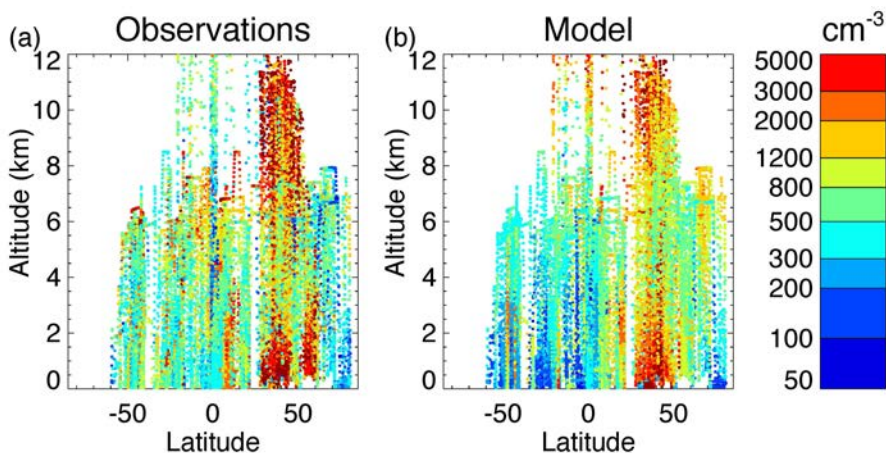


Fig. 2: Comparison of CLOUD measurements with global observations of new particle formation [26]. Comparison of observed (left) and modelled (right) particle concentrations (>3 nm diameter) versus latitude and altitude. This is the first time that a global aerosol model has been based solely on experimentally-measured particle nucleation and grow rates.

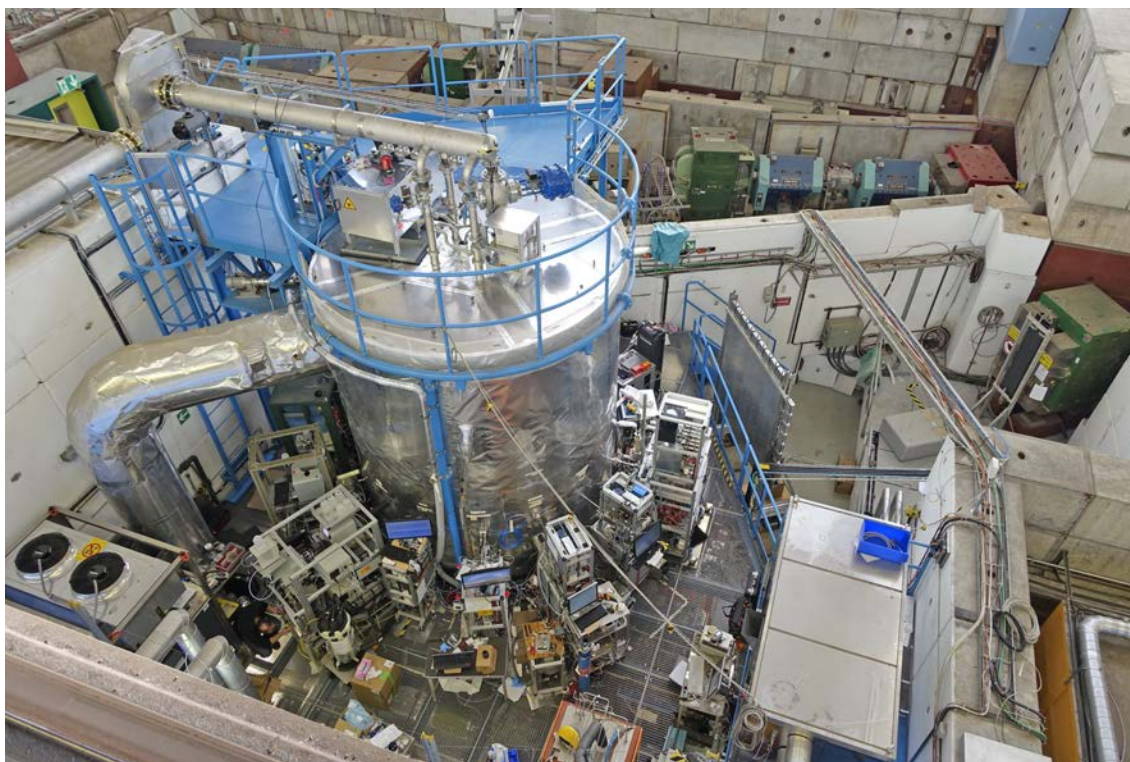


Fig. 3: CLOUD11. CLOUD with its analysing instruments in the East Hall, November 2016. The re-located UV excimer laser and Hamamatsu UV lamps can be seen above the chamber, together with the new upper platform.

2 CLOUD11 run, 26 Sep – 25 Nov 2016

2.1 Analysing instruments and facility upgrades

Around 40 analysing instruments were attached to the chamber during CLOUD11, of which 8 were mass spectrometers (Figs. 3, 4 and 5). Upgrades of the CLOUD facility for the CLOUD11 run included:

Upper platform: A new upper platform was constructed for CLOUD11 to accommodate anticipated additional instrumentation above the chamber (Fig. 3).

UV fibre optic system: The UV excimer laser (UVX) and Hamamatsu UV lamps (UVH) were re-located above the chamber and a new shorter quartz fibre optic system installed for the UVH lamps. The new fibre optics provided a factor 6 increase in ozone photolysis rate (wavelengths below 320 nm). The spectral intensities of the UV systems are shown in Fig. 6.

UV sabre 3: UV sabre 3 (Fig. 7) provides 400 W optical power at 385 nm for simulating actinic photolysis rates of NO_2 to NO , which is then oxidised back to NO_2 and the cycle repeated. In association with organic vapours from biogenic and anthropogenic sources, the NO_x cycle is the major pathway for ozone production in the troposphere. Nitric oxide is a major chemical reactant in the atmosphere and so it is important for CLOUD to realistically simulate atmospheric concentrations.

Gases: In CLOUD11 several new gases were introduced into the CLOUD for the first time: the sesquiterpene β -caryophyllene ($\text{C}_{15}\text{H}_{24}$), toluene (methyl benzene, $\text{C}_6\text{H}_5\cdot\text{CH}_3$), 1,2,4 trimethylbenzene ($\text{C}_6\text{H}_3\cdot 3(\text{CH}_3)$) and naphthalene (C_{10}H_8). Several of these vapours (β -caryophyllene and naphthalene, together with α -pinene and δ 3-carene) were provided from newly-constructed stainless-steel evaporators in thermally-controlled water baths. Among all the CLOUD gases, ammonia has

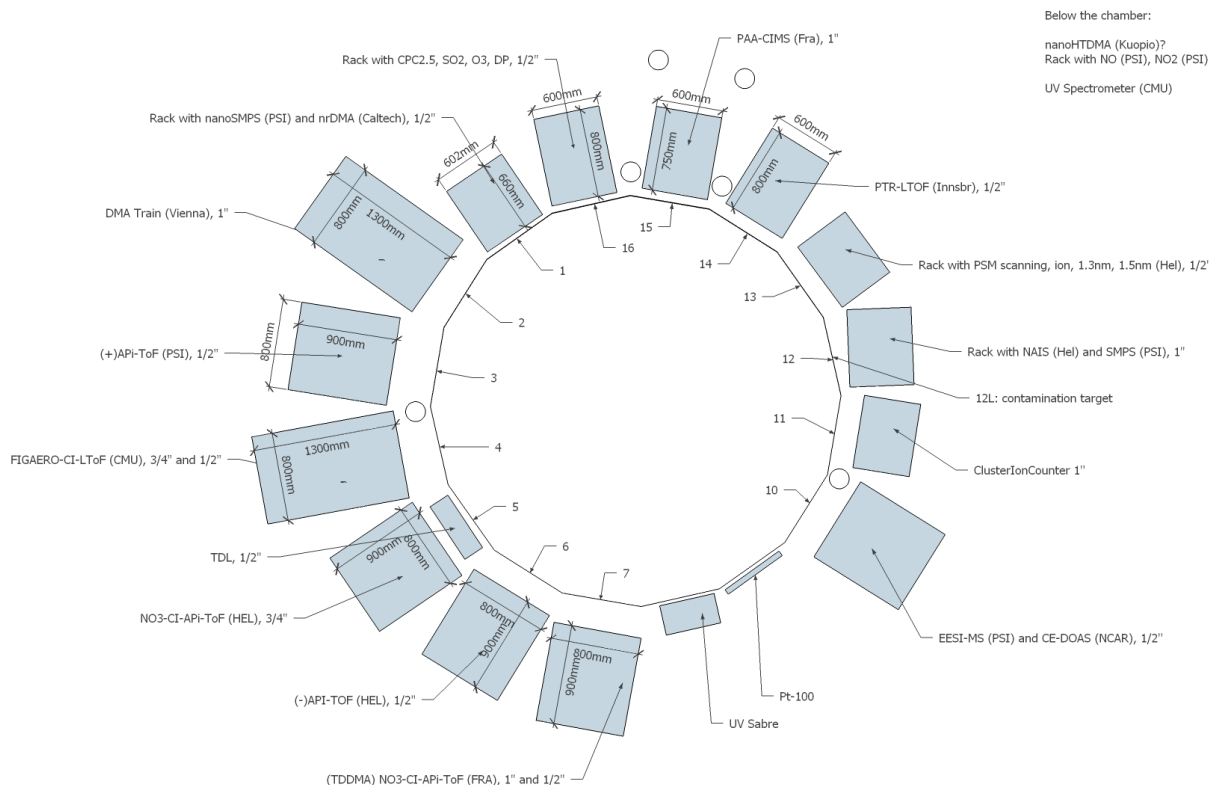


Fig. 4: CLOUD11 layout. Layout of the analysing instruments around the chamber during CLOUD11.

CLOUD11 INSTRUMENT SUMMARY (~40 instruments, including 8 mass spectrometers)

Port #	instrument name	Acronym	Institute	Function
1	nano scanning mobility particle sizer	nanoSMPS	PSI	size particle size distribution 5-60 nm
1	nano radial differential mobility analyzer	nrDMA	Caltech	size particle size distribution 3-10 nm
2	differential mobility analyzer train (6 DMAs, 6 CPCs, 2 PSMs)	DMA Train	UVienna	size particle size distribution 1.4-10 nm
3	(+)Atmospheric pressure interface time of flight mass spectrometer	(+)Api-ToF	PSI	positive ions
4	Filter Inlet for gases and aerosols for Chemical Ionization Long ToF	FIGAERO(-)LTOF	CMU	gases & particle composition with iodide ions
5	Tunable Diode Laser	TDL	KIT	water vapour
5	HOxROx(NO3-)ToF	HOxROx(NO3-)CI-API-ToF	UHel	HOMs, sum of ROx and HOx
6	(-)Atmospheric pressure interface time of flight mass spectrometer	(-)Api-ToF	UHel	negative ions
7	Thermal Desorption DMA coupled to NO3-CI-API-ToF	TDDMA(NO3)CI-API-ToF	UFra	HOMs, TDDMA: HOMs in the particle phase
8	UV Sabre 3 (385 nm)	UVS3	CERN	photolyse NO2 > NO
9	Pt100 string	Pt100_1-5	CERN	T, supplies TDL with T
10	capillary enhanced differential optical absorption spectrometer	CE-DOAS	CU Boulder	glyoxal, NO2
10	extractive electrospray ionization mass spectrometer	EESI-TOFMS	PSI	particle phase composition (w/o sampling)
11	Cluster Ion Counter	CIC	UHEL	positive and negative ions
11	scanning mobility particle sizer	SMPS	PSI	size particle size distribution 15-600 nm
12	Neutral Cluster and Air ion Spectrometer	NAIS	UHEL	size distribution of ions
13	Particle Size Magnifiers (scanning x2, fixed threshold x2)	PSM	UHEL and PSI	particle number at different cutoff sizes
14	Proton Transfer Reaction Long TOF MS	PTR-LTOF	UINNSBR	VOCs and oxidizedVOCs in the gas phase
15	Proton Transfer TOF	H3O+ TOF	UFRA	ammonia
16	Picarro G2103	Picarro	EMPA	ammonia
17-19	O3 monitor, SO2 monitor, CPC 2.5	O3, SO2, CPC2.5	EMPA, UFRA, PSI	CPC 3776 (butanol)
Below the chamber				
1	Cavity attenuated phase shift spectrometer	CAPS	PSI	NO2
2	NO monitor	NO monitor	PSI	NO
3	Dewpoint monitor	DP	CERN	relative humidity, RH
4	UV spectrometer and UV photodiodes (x2)	UVS, UVPD1, UVPD2	CERN, CMU	spectral and total UV intensity
Above the chamber				
1	UV excimer laser (248 nm) + fibre optics	UVX	CERN	photolyse O3 (OH production)
2	UV Hg-Xe lamps + fibre optics	UVH	CERN	broad UV source (general photolysis)

Fig. 5: CLOUD11 instruments summary.

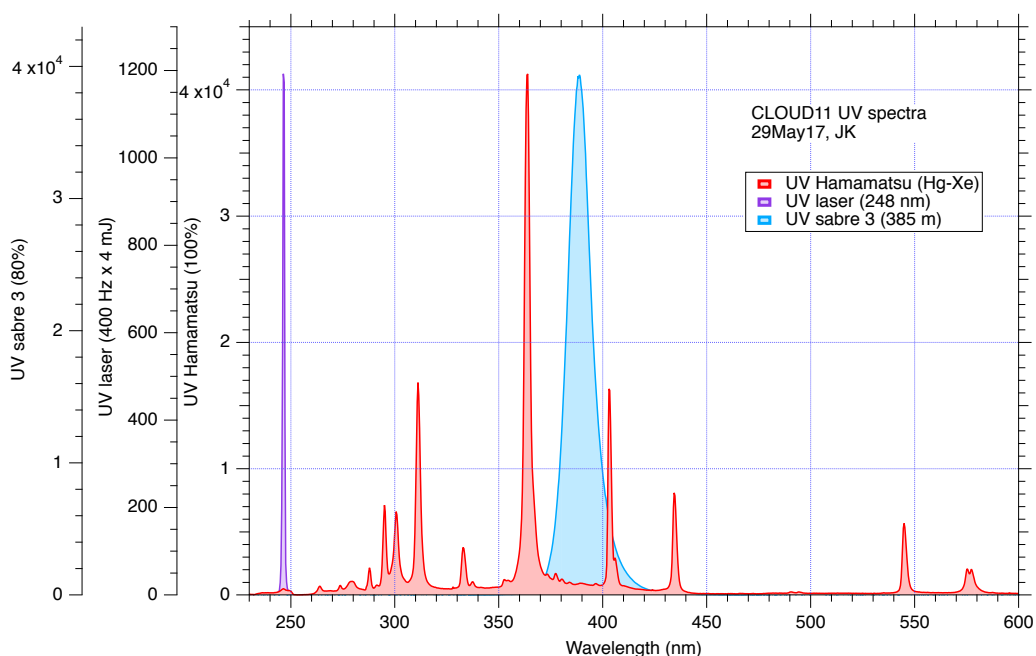


Fig. 6: CLOUD11 UV spectral intensity. The spectral intensity from the three UV sources operated during CLOUD11: Hamamatsu Hg-Xe lamp system (red curve), KrF excimer laser (purple curve at 248 nm) and the new UV sabre 3 (cyan curve at 385 nm). The UV laser is optimised for O_3 photolysis (hydroxyl, $OH\cdot$, radical production) and UV sabre 3 is optimised for NO_2 photolysis to NO . The UV spectrometer has different acceptances for each UV source so the relative signal intensities are arbitrary.



Fig. 7: UV sabre 3. UV sabre 3 provides 400 W optical power at 385 nm for simulating actinic photolysis of NO_2 to NO . The LED array is mounted on a liquid-cooled copper bar to remove 800 W thermal power. To maintain ultra clean and charge-free conditions in the CLOUD chamber, the assembly is housed in a quartz tube which is partially coated with Ti to remove surface charge. The UV sabre is designed to withstand temperature transitions in the CLOUD chamber between $-90^\circ C$ and $+100^\circ C$.

proved to be the most difficult to control owing to its adsorption onto stainless steel surfaces and subsequent release. We greatly improved the ammonia responsivity for CLOUD11 by installing sulfonated-coated tubes on the ammonia supply lines.

Isoprene cryotrap: CLOUD is investigating the influence of isoprene (C_5H_8) on atmospheric nucleation and growth, which is globally the most abundant biogenic vapour. During CLOUD10 we detected trace high-C contaminants in our isoprene supply, which interfered with the measurements. We therefore installed a cryotrap in the isoprene line for CLOUD11, which very successfully eliminated all contaminants.

2.2 CLOUD11 scientific programme

The primary aims of the CLOUD11 run were as follows:

1. **To investigate pure biogenic particle nucleation and growth under realistic environmental conditions.** The interplay of three vapours were investigated: isoprene (C_5H_8), the monoterpene, α -pinene ($C_{10}H_{16}$), and the sesquiterpene, β -caryophyllene ($C_{15}H_{24}$). These experiments followed up on CLOUD's discovery of pure biogenic nucleation reported in *Nature* [18], which involved α -pinene alone and showed a large ion enhancement (factor 10–100). The study included mixtures of these three biogenic vapours, as found in the atmosphere, as well as the influence of NO_x , oxidant concentrations (O_3 and $OH\cdot$), relative humidity (RH) and temperature, and under various ion conditions (neutral, GCR and π beam). These measurements are currently being analysed and will allow a more robust evaluation to be made of the atmospheric importance of pure biogenic nucleation, using the Leeds global aerosol model, GLOMAP.
2. **Anthropogenic aerosol particle nucleation and growth.** We studied three anthropogenic volatile organic compounds: toluene (methyl benzene, $C_6H_5\cdot CH_3$), 1,2,4 trimethylbenzene ($C_6H_3\cdot 3(CH_3)$) and naphthalene ($C_{10}H_8$). Nucleation and growth were studied under various conditions of NO_x , ozone, O_3 , and hydroxyl radicals, $OH\cdot$, and the analysis is in progress.
3. **Completion of the Hyytiälä, Finland, boreal forest simulation.** During CLOUD10 we simulated 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, δ 3-carene, NH_3 , NO , NO_2 and H_2O . This programme was successfully achieved with one exception that the available UV power was insufficient to reach atmospheric NO_2 photolysis rates. For that reason we constructed and operated an intense new UV source 3 at CLOUD11. This successfully reproduced actinic NO_2 photolysis rates and we were able to complete the daytime simulation of Hyytiälä conditions, closely reproducing the trace vapour spectra observed at that site. A detailed study and comparison is currently underway between the Hyytiälä observations and CLOUD simulation. One important result is that CLOUD finds that ion-induced nucleation is more important under Hyytiälä conditions than is observed [31]. The origin of this discrepancy is not yet understood and further studies are planned, both at Hyytiälä and at CLOUD.

The results from CLOUD10 and CLOUD11 have been discussed in several workshops during 2017 (see §3). Four papers from CLOUD10/11 have been submitted so far [28]–[31] and around 20 more manuscripts are in preparation, of which around half are planned to be submitted in 2017.

3 CLOUD MEETINGS, 2016–2017

The following CLOUD Collaboration meetings and data workshops were held in 2016–2017:

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 the latest unpublished CLOUD results by young scientists, together with recent results from invited external experts from Europe and the United States, followed by a half-day closed CLOUD collaboration meeting.



Fig. 8: Participants at the CLOUD-TRAIN final conference, Königstein, Germany, 14–17 Jun 2016.

CLOUD11 status and CLOUD collaboration meeting, CERN, 26–28 Oct 2016. Assessment of the in-progress CLOUD11 run, followed by a CLOUD collaboration meeting and Finance Review Committee meeting (FRC5).

CLOUD10/11 data workshop, Bad Zurzach, Switzerland, 30 Jan – 3 Feb 2017. Analysis of data from CLOUD10 and CLOUD11.

CLOUD10/11 data workshop, University of Vienna, Austria, 29 May – 2 Jun 2017. Analysis of data from CLOUD10 and CLOUD11.

4 COLLABORATION ISSUES

4.1 CLOUD Collaboration

In 2016 Manchester University left the CLOUD Collaboration after completion of the PhD of its CLOUD-TRAIN ESR. In 2016 CLOUD was joined by a new partner (Rainer Volkamer group) from the University of Colorado Boulder, Department of Chemistry & Biochemistry, with special theoretical and experimental expertise that is needed for CLOUD in multiphase chemistry in clouds and aerosols.

4.2 CERN fellows

CLOUD wishes to thank CERN EP for their generous support of an Applied Fellow for CLOUD Run Coordinator, starting 1 Jan 2017. The CERN CLOUD team has also been joined by an open-choice Research Fellow, starting 1 Jan 2017.

4.3 CLOUD-MOTION

CLOUD has been awarded an unprecedented third Marie Curie Innovative Training Network grant within the framework of the EC Horizon 2020 research and innovation programme. CLOUD-MOTION involves 15 ESRs (PhD students) who will carry out their research on CLOUD and are based at 10 beneficiaries from within the CLOUD Collaboration. Among these, two will be CERN ESRs who will be enrolled at Goethe University Frankfurt and University of Lisbon, respectively, for their academic requirements. CLOUD-MOTION is coordinated by Goethe University Frankfurt and the award will be around 3.9M€. It is expected to run from 1 September 2017 until 31 August 2021.

5 CLOUD12 BEAM REQUEST, 18 Sep – 27 Nov 2017

The CLOUD12 run is planned for 18 Sep – 27 Nov 2016, with the first week involving instrument setup and the final week using cosmic rays (no East Area beams)—a 9-week data period in total. The aims of the CLOUD12 run are (with approximate duration in parentheses):

1. **Marine nucleation and growth involving iodine compounds (2 wk).** This is a new study for CLOUD. Iodine-associated particle formation has been observed in coastal regions when seaweed is exposed at low tide. Two pre-cursor vapours will be studied in CLOUD: diiodomethane (CH_2I_2) and elemental iodine (I_2). A new green light sabre (LS4, 100 W at 528 nm) is under construction to photolyse I_2 for these experiments.
2. **Growth rates of pure sulphuric acid particles at small sizes (3 d).** Newly-formed particles must grow to sizes above around 50 nm before they become cloud condensation nuclei (CCN) and affect clouds and climate. The fraction of new particles that grow to CCN is highly sensitive to their growth rates at small sizes, when they are highly mobile and susceptible to scavenging by pre-existing aerosol. In 2016 CLOUD reported in *Nature Communications* [17] dramatic differences in growth rates for various mixtures of sulphuric acid and base vapours (ammonia and dimethylamine). Using newly-developed instrumentation—first used in CLOUD10/11—we will repeat these measurements with far higher precision, measuring the growth rates of pure sulphuric acid particles at small sizes, and comparing with growth rates measured when ammonia is present.
3. **Multi-component aerosol particle nucleation and growth (4 wk).** In 2016 CLOUD published in *Science* [26] the first global aerosol model study based entirely on experimental measurements, which compared closely to atmospheric observations throughout the free troposphere. This involved parameterisation of extensive CLOUD measurements of binary ($\text{H}_2\text{SO}_4\text{--H}_2\text{O}$) and ternary inorganic ($\text{H}_2\text{SO}_4\text{--NH}_3\text{--H}_2\text{O}$) nucleation over the full range of tropospheric conditions. During CLOUD12 we plan to collect the necessary data to extend this parameterisation and global model study to include multi-component nucleation involving highly oxygenated biogenic vapours ($\text{H}_2\text{SO}_4\text{--NH}_3\text{--HOM--H}_2\text{O}$, where HOM refers to Highly Oxygenated Molecules). The new parameterisation will also use extensive measurements performed in CLOUD10/11 on pure biogenic ($\text{HOM--H}_2\text{O}$) nucleation and on the simulation of daytime and night-time conditions at the Hyytiälä boreal forest station in Finland. During the CLOUD12 study, further pure biogenic runs will be performed under the cold conditions corresponding to convective cloud outflows over the Amazon, where vigorous particle formation has been observed from research aircraft. The end result will be the most globally-comprehensive set of atmospheric nucleation measurements achieved so far by CLOUD between the boundary layer and the top of the troposphere.
4. **Anthropogenic aerosol particle nucleation and growth (2 wk).** Particle nucleation and growth from anthropogenic vapours is a major source of PM_{2.5} in urban environments, but the underlying processes are not well understood. PM_{2.5} is readily transported deep into the lungs and is a major cause of mortality in urban environments, not only in highly polluted megacities in developing countries but also in the cities of Europe and North America. During CLOUD12 we will extend

previous CLOUD11 studies of particle nucleation and growth from the anthropogenic volatile organic compounds 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, $OH\cdot$, and various levels of pre-existing aerosol loadings).

Each of the individual experiments in the four programmes above will be performed under various ionisation conditions to measure the effects of ions on the physical and chemical processes under study: neutral (ion-free; with the high voltage clearing field on and beam stopper closed), GCR (galactic cosmic rays; with the HV off and beam stopper closed) and π (pion beam; with the HV off, beam stopper open and collimators set to the desired beam intensity).

6 CLOUD REQUESTS

6.1 Permanent CLOUD open office/meeting room

CLOUD has requested in its long-term planning document to the SPSC [6] a dedicated 50 m² 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. The meeting room is also needed as an open office for CLOUD researchers and for the weekly work of the CERN CLOUD team. During the last two CLOUD runs (2015-2016), a temporary meeting room in bat. 510 was provided by EP. However, other uses are planned for bat. 510 in future, and it will become unavailable.

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.

6.2 CLOUD operation during LS2 East Area Renovation, 2019–2020

During LS2, 2019–2020, the CERN accelerators will be off and the renovation of the beam lines and infrastructure in the East Area will be carried out.

Although the CLOUD experimental programme will be reduced without beam, we are requesting to run with cosmic rays during the fall 2018 and fall 2019 periods. There is considerable urgency to continue with CLOUD data collection over this 2-year period in view of the important impact CLOUD results are having on the understanding of aerosols on climate change, which represents the largest current uncertainty in anthropogenic radiative forcing. Moreover, a large number of PhD students—including 15 from the new H2020 Marie Curie Initial Training Network, CLOUD-MOTION—will rely on CLOUD data collected during the 2018–2020 period.

CLOUD appreciates that the East Area Renovation project takes priority during LS2 and that its work schedule in the East Hall cannot be impacted. We are in close discussion with the East Area Renovation team (project leader, Sebastien Evrard /EN-EA-AC) and are exploring how essential CLOUD services (access, limited crane use, electricity, de-ionised water, chilled water, counting room, etc.) could be maintained to allow operation of CLOUD in fall 2019 and fall 2020.

Acknowledgements

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