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CLOUD Collaboration

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1 SCIENCE HIGHLIGHTS

CLOUD is studying, under controlled conditions in the laboratory, how aerosol particles form and grow from trace atmospheric vapours, and how these processes are affected by ions from galactic cosmic rays [1, 2, 3]. The experiment involves a large chamber whose contents are continuously sampled and analysed by a suite of mass spectrometers and other instruments. A 3.5 GeV/c π^+ beam from the CERN PS simulates galactic cosmic ray (GCR) ionisation in the chamber corresponding to any chosen altitude in the troposphere. CLOUD is also studying the effects of ions and aerosols on cloud microphysics, using adiabatic pressure reductions to form liquid or ice clouds in the chamber. The clouds are analysed both *in situ* with optical instruments and also via sampling probes to external instruments that measure the liquid droplet or ice particle activation properties of the aerosol particles.

A summary of CLOUD publications, 2011–2022, is shown in Fig. 1. The publications in peer-reviewed journals total around 70 and they now receive almost 1000 citations per year. During 2022–2023, twelve papers have been published so far [5]–[16], including one each in *Nature* [7], *Nature Chemistry* [9], *Nature Communications* [13] and *Science Advances* [16]. Five more papers are currently under review [17]–[21]. Some selected results from the published papers are presented below.

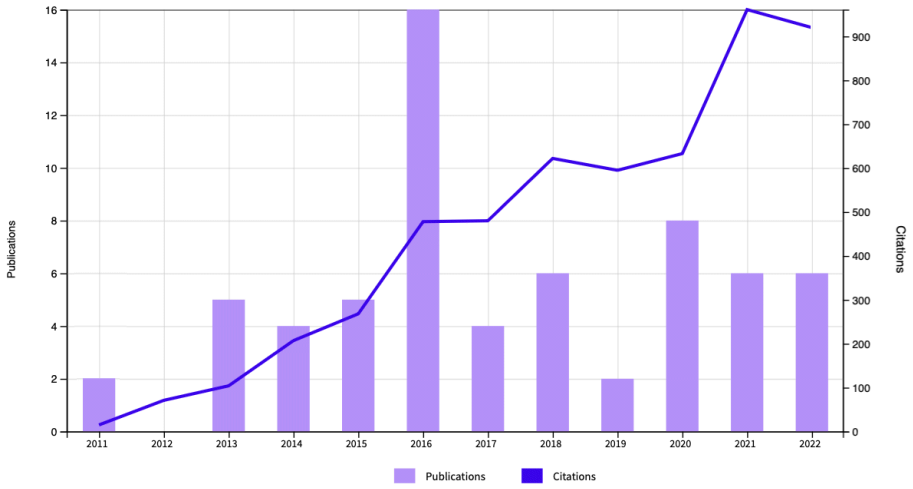

Fig. 1: Summary of CLOUD publications, 2011–2022. CLOUD peer-reviewed publications (purple bars) and annual citations (blue line) versus year. The CLOUD publications over this period total around 70 and the citations total around 5800. The CLOUD h-index is 32, which includes seven papers with over 200 citations and one with 900 citations. *(Clarivate Web of Science.)*

Chemical pathway for formation of iodic acid in the atmosphere [9]: Experiments at CLOUD have shown the importance of iodine oxoacids (iodic acid, HIO_3, and iodous acid, HIO_2) for particle formation in pristine regions of the atmosphere [4, 11]. The iodine arises from coastal seaweeds and the ocean surface. Iodine emissions from the sea surface have tripled since 1950 and are projected to keep increasing with rising ozone concentrations and retreating sea ice. The CLOUD experiments use green illumination to photolyse molecular iodine into iodine atoms without producing hydroxyl radicals from ozone in the chamber. This revealed a new and highly efficient chemical pathway for producing iodic acid via iodooxy hypoiodite, I_2O_2, which can explain daytime observations of HIO_3 production in the remote lower troposphere (Fig. 2) [9]. Iodooxy hypoiodite is rapidly produced in weak daylight from $I\cdot + O_3 \rightarrow IO\cdot + O_2$ followed by $IO\cdot + IO\cdot \rightarrow IOIO$, and then iodic acid is produced from the reactions $IOIO + O_3 \rightarrow IOIO_4$ followed by $IOIO_4 + H_2O \rightarrow HIO_3 + HOI + O_2$. Overall, the CLOUD measurements indicate that iodic acid is a highly efficient source of new particles in the atmospheric boundary layer: it competes in rate with sulfuric acid-ammonia in pristine regions, it proceeds in weak, overcast daylight and a single precursor vapour drives both rapid nucleation and growth.

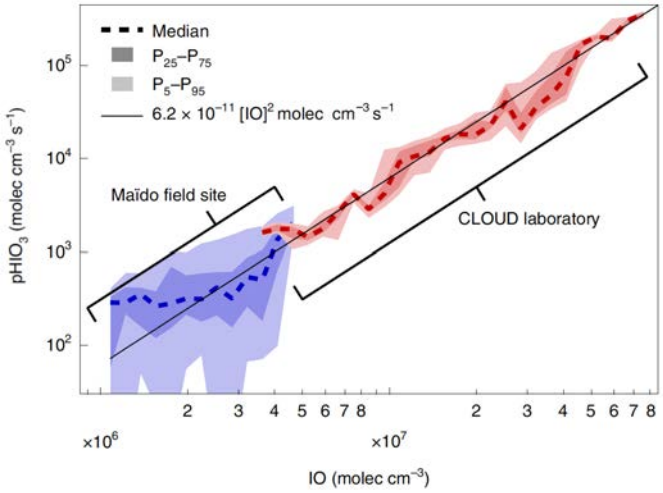

Fig. 2: Comparison of CLOUD and field measurements for production of iodic acid in the atmosphere [9]. Good agreement is observed between iodic acid (HIO_3) production rates measured at CLOUD (red curve) and measured by our CLOUD colleagues at the Maïdo observatory, La Réunion (2154 m altitude; blue curve). IO radical concentrations at CLOUD overlap with those found in the remote lower free troposphere. The solid black line is the IOIO formation rate expected from IO radicals at 283 K. This corresponds to the rate-limiting step of HIO_3 formation under both field and laboratory conditions, confirming the new chemical pathway discovered by CLOUD.

Ion-charged particle recombination rates [15]: As well as studying the effects of galactic cosmic ray ionisation on aerosol particle nucleation and growth, we are performing experiments at CLOUD to investigate possible direct effects of charge on the microphysics of fair-weather (non-thunderstorm) clouds. The charge state of aerosol particles is affected by the unipolar space charge in the vicinity of cloud boundaries arising from ion-drift in Earth's electric field. Moreover, cloud droplets in fair-weather clouds are frequently found to have charges of up to around 100 e, probably resulting from fragmentation processes. There are no experimentally-established significant effects that fair-weather charges exert on cloud microphysics, so these are highly speculative experiments. However, possible effects include electroscavenging (enhanced aerosol-droplet scavenging), aggregation (or repulsion) of water droplets or ice particles, droplet activation at lower water vapour supersaturation or higher freezing points of supercooled water droplets. To investigate these processes requires the production of cloud condensation nuclei with multiple charges which, because of electrostatic repulsion, cannot be achieved by diffusion charging. Initial experiments were therefore performed during CLOUD14 using a newly-developed electrospray CHarged AeRosol GEnerator (CHARGE). The experimental setup is shown schematically in Fig. 3 together with our recently-published measurements of the ion–particle rate coefficients versus number of charges on the particle in the range 1–9 e [15].

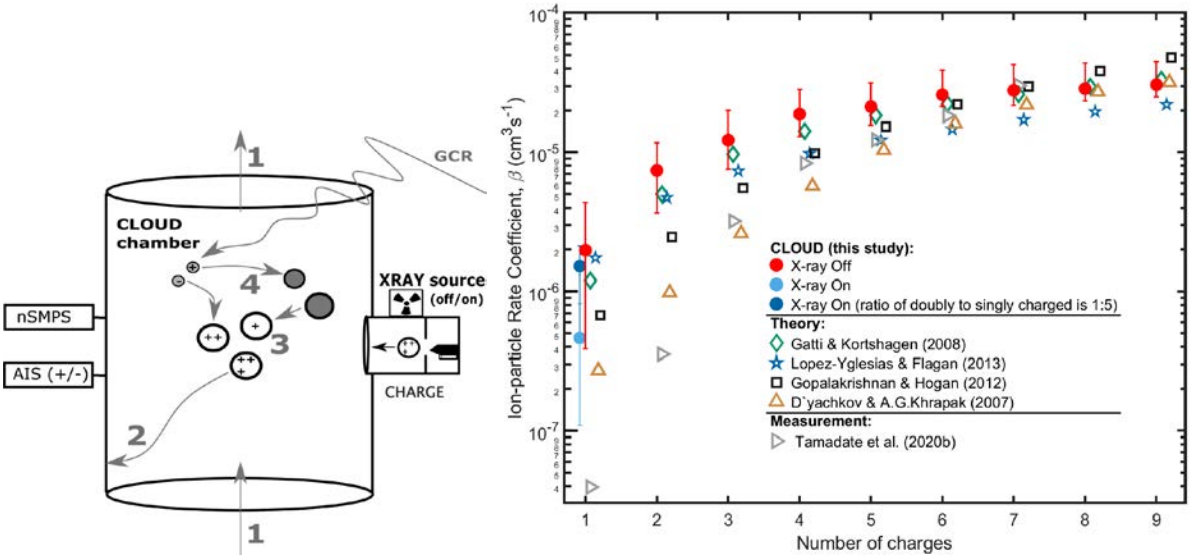

Fig. 3: Measurement of the collision rate coefficients between small ions and multiply-charged ultrafine aerosol particles [15]. *Left panel:* Schematic representation of the experimental setup. Charged particles are injected into the CLOUD chamber from the CHarged AeRosol GEnerator (CHARGE). An X-ray source can be turned on to neutralise the particles, i.e. reduce their charges to the Boltzmann distribution, in order to perform control experiments with single- and zero-charged particles. After an injection period of around 30 min, particle injection from CHARGE is switched off and the evolution of the particle charges during a decay stage is monitored by sampling instruments such as the nano scanning particle mobility sizer (nSMPS) and air ion spectrometer (AIS). The evolution of particle and ion charges in the chamber is analysed as a function of the production and loss terms: (1) dilution loss, (2) wall loss, (3) coagulation loss, and (4) collisions of aerosols with oppositely-charged small ions produced by galactic cosmic rays. *Right panel:* Experimental results showing the ion–particle rate coefficients versus number of charges on the particle [15]. The CLOUD measurements are indicated by filled circles which are coloured red for X-ray Off experiments, blue for X-ray On experiments assuming full steady-state ion distributions, and cyan for X-ray On and assuming incomplete steady-state ion distributions. Theoretical predictions are shown in open symbols for 1 nm negative ions recombining with positively-charged particles of 6 nm diameter. Good agreement is found between the CLOUD measurements and the theoretical model of Gatti and Kortshagen (2008). The only previous experimental measurements for multiply-charged particles are shown in grey triangles (Tamadate et al., 2020). The $\pm 1\sigma$ error bars indicate total statistical plus systematic errors. The symbols are offset from integer charges to improve their visibility.

Fig. 4: CLOUD15 run (19 Sep - 22 Nov 2022). CLOUD and some of its analysing instruments in the T11 experimental zone during the CLOUD15 run. During operation, the T11 beam emerges from the lower right hand side of the figure and passes through two counter planes (B and VH; the latter is seen at the right hand edge of the image) before crossing the CLOUD chamber located inside a thermal housing. Mass spectrometers and other instruments mounted on the platform continuously extract air from the CLOUD chamber via sampling probes to analyse its contents. FLOTUS is located on the opposite side of CLOUD and is not visible in the image.

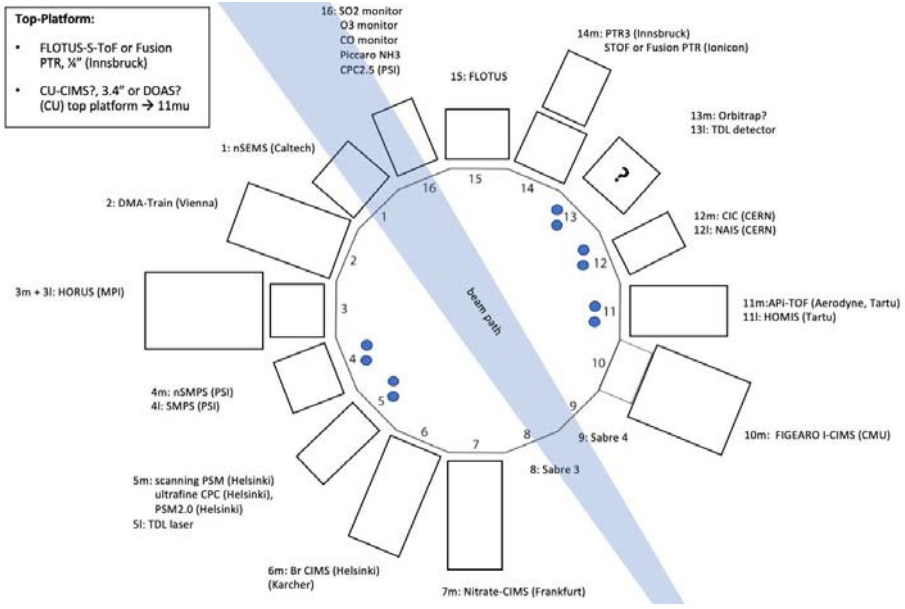

Fig. 5: CLOUD15 instrument layout. Layout of the analysing instruments around the chamber during the CLOUD15 run, 19 Sep - 22 Nov 2022.

2 CLOUD15 RUN, 19 Sep – 22 Nov 2022

The CLOUD15 run (Figs. 4 and 5) took place 19 Sep – 22 Nov 2022, with the following science goals:

1. **Marine nucleation and growth with methanesulphonic acid:** Sulphuric acid vapour is produced by the oxidation of sulphur dioxide (SO_2) in the atmosphere. The dominant source of SO_2 in the marine atmosphere is from the oxidation of dimethylsuphide, DMS (CH_3SCH_3), which is emitted from phytoplankton at the sea surface. As well as sulphuric acid, oxidation of DMS produces methanesulphonic acid, MSA (CH_3SO_3H), with a high yield that we measure to exceed H_2SO_4 below 10 °C [10]. Methanesulphonic acid is likely to nucleate via an acid-base mechanism but experiments at atmospheric conditions remain ambiguous since MSA and H_2SO_4 always appear together from DMS oxidation. To resolve this ambiguity, we conducted experiments during CLOUD15 where MSA either was obtained from DMS oxidation or was directly injected into the chamber from an evaporator, i.e. without H_2SO_4. The experiments were performed with ammonia and dimethylamine (DMA; $(CH_3)_2NH$), and at temperatures between +20°C and -50°C.

2. **Upper free tropospheric nucleation and growth with iodine oxoacids, sulphuric acid and biogenic vapours:** Although largely absent in the Amazon rainforest itself, copious new particle formation is observed high overhead in convective cloud outflows at altitudes between 8 and 15 km. Similarly, on a much broader scale, intense new particle formation has been observed in the tropical upper free troposphere over both the Pacific and Atlantic oceans, covering about 40 per cent of Earth's surface. We therefore performed experiments during CLOUD15 to study new particle formation and growth involving iodic acid (HIO_3), iodous acid (HIO_2), sulphuric acid and biogenic vapours at upper tropospheric conditions: low temperatures (-30°C → -50°C), low vapour concentrations and high ion concentrations.

3. **FLOTUS commissioning:** During the CLOUD15 run we installed the new FLOw TUbe System (FLOTUS) at CLOUD (Fig. 6) and carried out the initial commissioning of the combined CLOUD-FLOTUS system. The commissioning was completed during the dedicated CLOUD16T technical run in Spring 2023, and initial experiments performed on new particle formation from aged α-pinene vapours.

3 CLOUD16T RUN, 17 Apr – 12 May 2023

We simulate in CLOUD selected regions of Earth's atmosphere. The 26 cubic meter stainless steel chamber is filled with humidified ultra-pure synthetic air, made from evaporated cryogenic nitrogen and oxygen into which various trace atmospheric vapours are injected (ozone, sulphur dioxide, nitric acid, ammonia, organic vapours, iodine, etc.). By adjusting parameters such as vapour concentrations, temperature, humidity, illumination and beam intensity, we can simulate and very precisely control the atmospheric conditions under study. However, due to losses of vapours and particles when they come into contact with the wall of the CLOUD chamber, experiments can only last a few hours. This gives sufficient time to study many processes but not the slow transformation through several oxidation steps that some organic vapours undergo in the atmosphere over the course of several days.

FLOTUS (Fig. 7) fills that gap. It is a 60-litre quartz chamber of 3 m length and 20 cm diameter, with gas occupancy times of 1–3 minutes. We commissioned FLOTUS, measured its technical performance and performed initial experiments during the CLOUD15 and CLOUD16T runs ('T' denotes a technical run). The integration of FLOTUS with CLOUD introduces substantial additional complexity to the experiments since, in essence, two coupled and independently-controlled experimental systems are in operation simultaneously. During CLOUD15 we measured that FLOTUS simulates hydroxyl radical oxidative exposures of organic vapours up to the equivalent of 100 h in the atmosphere (Fig. 8). An example physics experiment with FLOTUS during CLOUD16T is shown in Fig. 9.

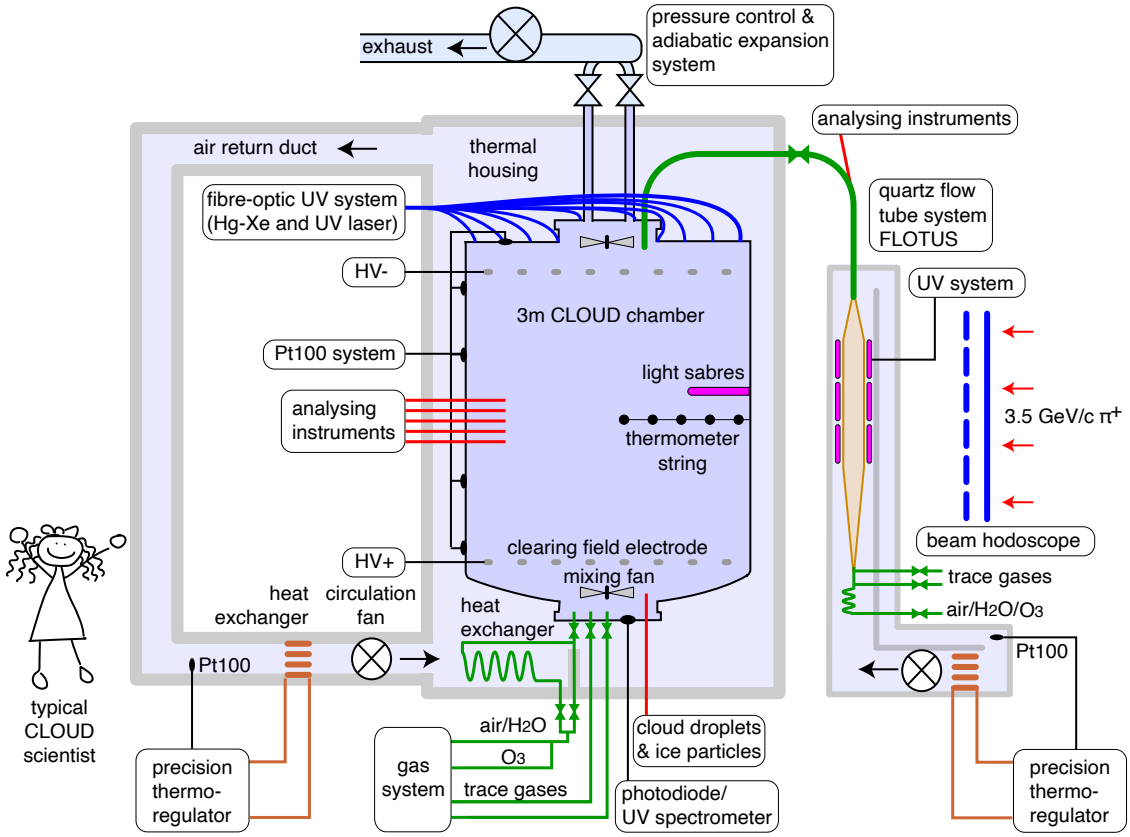

Fig. 6: Schematic of CLOUD with the new FLOw TUbe System (FLOTUS). Organic vapours can be pre-aged in FLOTUS before injection into the CLOUD chamber, where their role in aerosol nucleation and growth is investigated with the full suite of CLOUD instruments. The output from FLOTUS is transferred into CLOUD via a wide-bore pipe (thick green line). In addition, the FLOTUS output can be directly sampled via a pickoff pipe (thin red line) for analysis by instruments mounted on the upper platform. All the operating conditions of FLOTUS (gases, UV irradiation, relative humidity and temperature) are controlled independently of the CLOUD chamber.

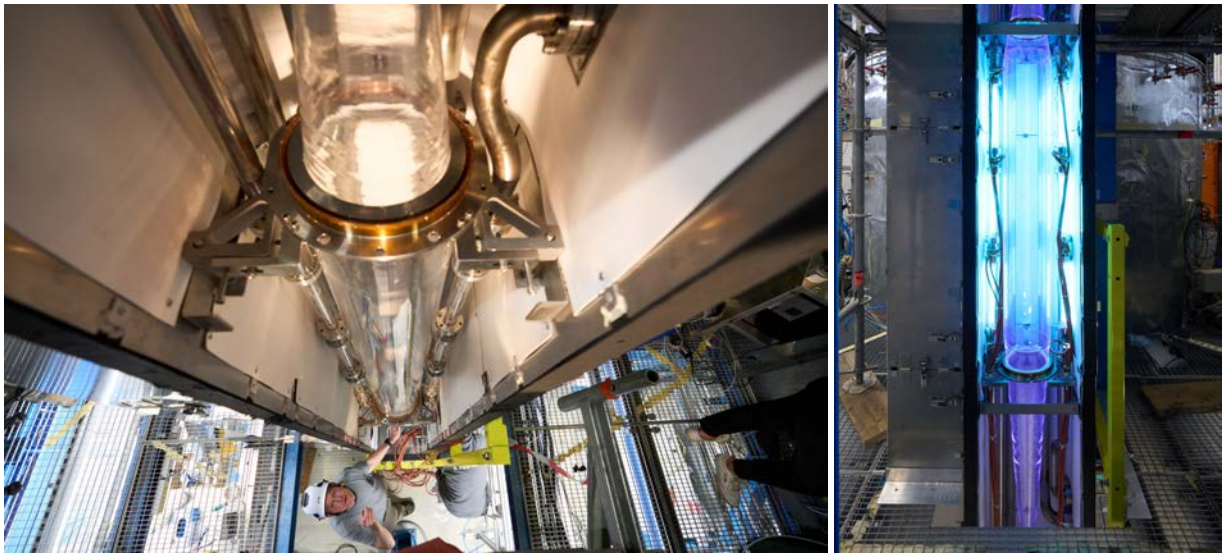

Fig. 7: FLOTUS at CLOUD. FLOTUS is a 60 litre laminar-flow quartz drift tube of length 3 m in which organic vapours can be aged by exposure to hydroxyl radicals corresponding to several days in the real atmosphere. The hydroxyl radicals are produced by photolysis of ozone under intense UVC irradiation (right panel).

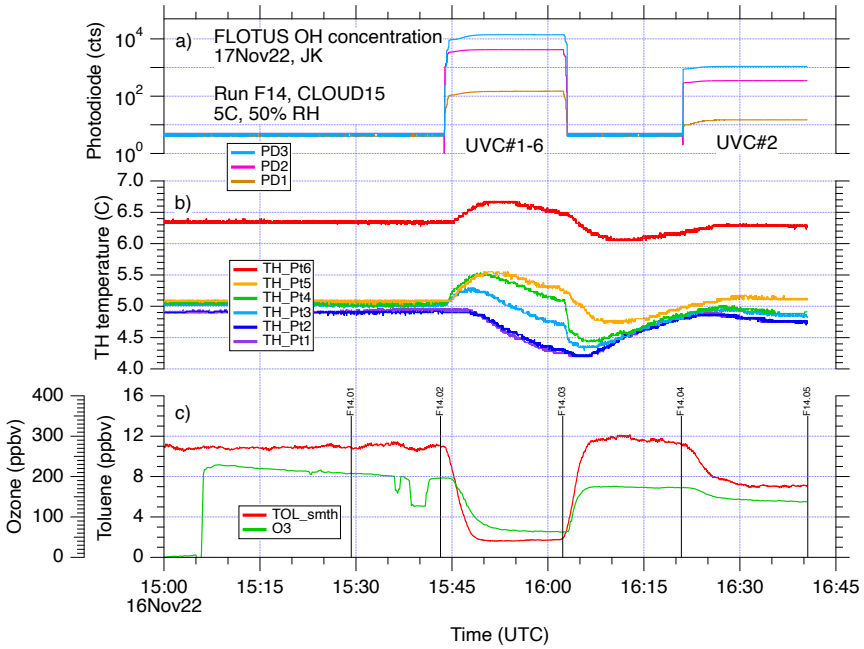

Fig. 8: Measurement of the technical performance of FLOTUS during CLOUD15. a) Response of photodiodes mounted inside the thermal housing versus time. There are two periods when the UVC (254 nm) lamps were switched on: i) run F14.02 with all 6 lamps and ii) run F14.04 with only a single lamp. b) Temperature of the air circulating through the FLOTUS thermal housing, measured by Pt100 sensors. Sensors TH_Pt1 to TH_Pt5 correspond to the full length of the section that houses the quartz chamber, which show a thermal gradient of $\sim$1.0 °C when all 6 lamps (70 W power each) are switched on. The thermoregulation system is seen reacting to the increased heat load to bring the temperature back to the 5 °C setpoint. The vertical orientation of the quartz chamber inhibits convective flows, preserving laminar flow along its length. c) Concentrations of ozone (green curve) and toluene ($C_6H_5CH_3$; red curve) measured at the exit of FLOTUS. When the UVC lamps are on, ozone is photolysed to produce hydroxyl (OH·) radicals, which then react with toluene, reducing the concentrations of both vapours. After accounting for the 90 s flow time in FLOTUS at 40 l/min, the measured toluene loss rate implies 1.3×10^{10} cm^{-3} OH· for six UVC lamps under these conditions, equivalent to around 100 h exposure to sunlight in the real atmosphere.

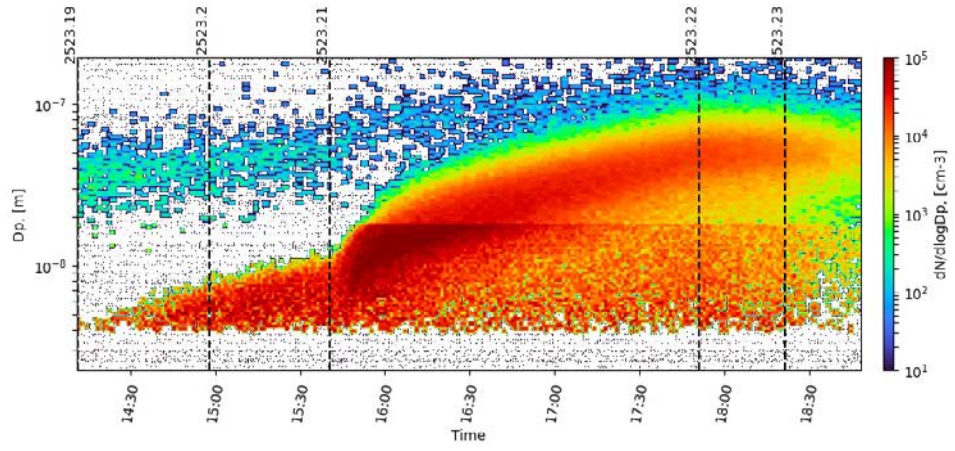

Fig. 9: Evolution of particles measured in CLOUD with aged biogenic vapours injected from FLOTUS during CLOUD16T. Particle size spectrum measured in the CLOUD chamber versus time, during the CLOUD16T run. In this experiment, α-pinene is oxidised in FLOTUS and the products are then transferred into CLOUD. Run 2523.19 corresponds to zero illumination in CLOUD (which switches off hydroxyl production), and then weak UVC is added in CLOUD at the start of run 2532.20, followed by stronger UVC in CLOUD at run 2532.21, when a sharp increase in particle formation and growth rate is seen.

4 CLOUD MEETINGS, 2022–2023

The following in-person CLOUD Collaboration meetings and data workshops were held in 2022–2023:

CLOUD15 status and CLOUD collaboration meeting, CERN, 11–14 Oct 2022. Assessment of the CLOUD15 run in progress, followed by a CLOUD collaboration meeting and Finance Review Committee meeting FRC11.

CLOUD15 data workshop, Bad Zurzach, Switzerland, 13–17 Feb 2023. Initial results from CLOUD15, instrument developments, CLOUD modelling and planning for CLOUD16T/16.

CLOUD-DOC summer school and CLOUD collaboration meeting, Gras-Ellenbach, Germany, 12–20 Jun 2023. Summer school on organic new particle formation, ice nuclei, cloud condensation nuclei, global climate change, health effects of particles, creative problem solving, outreach, scientific writing and developing a start-up company. The collaboration meeting discussed results from CLOUD15 and CLOUD16T, and the planning for the CLOUD16 experiments.

5 COLLABORATION

CLOUD has been awarded an unprecedented fourth Marie Curie Innovative Training Network grant within the framework of the EC Horizon Europe research and innovation programme. The project, named CLOUD-DOC, started on 1 September 2022 and will run for four years until 31 August 2026. CLOUD-DOC involves 12 PhD students (including 2 with Swiss partners) who will carry out their doctoral research on CLOUD. The students are based at twelve CLOUD institutes, including CERN. CLOUD-DOC is coordinated by Goethe University Frankfurt, and the EC award totals EU 2.7 M€, plus an additional 20% Swiss contribution.

6 BEAM REQUEST 2023

CLOUD requests to run from 25 Sep to 3 Dec 2023 for CLOUD16 (10 wk). Our instrument installation week will be 18–24 Sep 2023. We understand that the CERN PS will stop on 30 Oct 2023 to reduce energy costs and we will continue with a GCR run after that time. The science goals mainly focus on new particle formation under cold conditions, as follows:

1. **Upper free tropospheric nucleation and growth over the Amazon:** We will extend our CLOUD15 studies of upper free tropospheric new particle formation involving sulphuric acid and organic vapours to include dimethylsulphide (CH_3SCH_3) and NO_x at temperatures between -10 °C and -50 °C.

2. **Upper free tropospheric nucleation and growth involving surfactants:** Surprisingly high concentrations (several pptv) of long-chained fatty aldehydes ($(CH_2)_nO$) have been reported by our CLOUD colleagues at the Chacaltaya observatory in the Bolivian Andes (5200 m altitude). These surfactants originate from the surface of the tropical Pacific Ocean and, after convection, are transported long distances of up to 1000 km to reach the observatory, so their measured concentrations are highly diluted compared with those expected in the upper troposphere nearer their sources. During CLOUD16 we will study the contributions of two fatty aldehydes—nonanal ($C_9H_{18}O$) and dodecanal ($C_{12}H_{24}O$)—to new particle formation and growth at temperatures between -15 °C and -50 °C. These experiments will also include sulphuric acid and NO_x.

3. **Nucleation and growth in the Arctic:** We will study new particle formation and growth involving dimethylsulphide, methanesulphonic acid (MSA), iodic acid and ammonia at concentrations found in the Arctic and at temperatures between 10 °C and -10 °C. The MSA will be injected into

CLOUD via FLOTUS to minimise the effect of high vapour concentrations of MSA and ammonia in nearby 'plumes' around their injection points before the vapours are well mixed. We will also include experiments to investigate the contribution of the dialdehyde glyoxal ($CHO.CHO$) to particle growth via oligomerisation in the particle phase. Glyoxal is one of the most prevalent carbonyl compounds in the atmosphere, with concentrations frequently exceeding 100 pptv.

4. **Interaction of biogenic and anthropogenic vapours:** Urban environments involve a mixture of biogenic vapours (from trees and parks) and anthropogenic vapours (from automobiles, domestic stoves and industrial products). However, the impact on smog formation of the interactions between these two classes of vapours remains poorly known. During CLOUD16, we will investigate the interactions between biogenic organic vapours (α-pinene and isoprene) and anthropogenic vapours (SO_2, O_3, NO_x, HONO and the aromatic organic, trimethybenzene - C_9H_{12}) at 5 °C and 30 °C. In addition, we will add propene (C_3H_6) for certain experiments in order to vary the concentration of light RO_2 radicals.

7 SUMMARY

Experiments at CLOUD are measuring new particle formation at the molecular level, and helping to interpret observations in the ambient atmosphere. This mechanistic understanding has enabled the development of laboratory-based implementations of particle nucleation in global atmospheric chemistry and climate models. This is effectively catching up with gas-phase chemical kinetics where, for more than 40 years, laboratory experiments have provided straightforward kinetic equations that could be inserted directly into models, i.e. explicit mechanisms. In the aerosol world, a similar level of "nucleation kinetics" has largely been achieved through CLOUD experiments over the last twelve years—but more work remains to be done.

Acknowledgements

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