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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 and also with sampling instruments that measure the liquid droplet or ice particle activation properties of the aerosol particles. During 2021–2022, nine papers have been published so far [4]–[12], including one in *Science* [5] and one in *Nature* [11]. Three more papers are under review [13]–[15]. Some results from these papers are presented below.

1.1 Role of iodine in atmospheric aerosol particle formation

A paper from CLOUD published in *Science* [5] reported how new particles form from iodine-containing vapours under marine boundary layer conditions. We measured the nucleation and growth rates (Fig. 1) as well as the molecular composition of freshly formed particles from iodic oxoacids (iodic acid, HIO_3, and iodous acid, HIO_2) from +10 °C to -10 °C Fig. 2). These vapours derive from photolysis and oxidation of molecular iodine emitted from the ocean surface. We found that the conversion to iodine oxoacids takes place rapidly even under weak daylight conditions, without requiring any ultraviolet.

We found that the nucleation rates of iodic acid particles are extremely rapid, even exceeding those of sulphuric acid-ammonia under similar conditions (Fig. 1). The particles form especially rapidly on ions from galactic cosmic rays, limited only by the rate of collision with iodic acid molecules (termed the kinetic limit) (Figs. 1 A and 3A). We measured the growth rate of embryonic particles during ion-induced nucleation to be a factor 6 higher than for neutral nucleation (Fig. 3B). The faster growth rate helps to shepherd freshly nucleated particles rapidly through the smallest size range where they are most vulnerable to scavenging loss, thus greatly increasing the survival probability of ion-induced particles.

We also found that the particles are composed almost entirely of iodic acid, which drives rapid growth at the kinetic limit. However, iodous acid plays a key role in stabilising the initial steps of neutral (uncharged) particle formation. Our global boundary layer measurements of iodic acid indicate the conditions for abundant iodine new particle formation and rapid growth are frequently reached at coastal mid-latitude and polar sites.

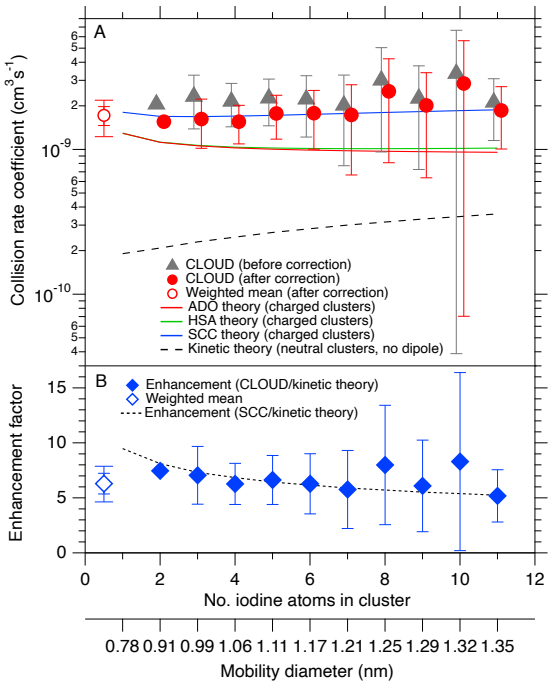

Fig. 3: Collision rate coefficients for ion-induced iodic acid nucleation [5]. A) Collision rate coefficients measured between neutral HIO_3 monomers and charged clusters containing up to 11 iodine atoms, at (0.8-2.0)×10^7 cm^{-3} HIO_3 (red circles). The solid curves show theoretical expectations for the charged collision rate coefficients from average dipole orientation theory (ADO) and its extensions. The expected collision rate coefficients between neutral monomers and neutral clusters, ignoring dipole-dipole interactions, are shown by the dashed black curve. B) Measured enhancement factors for charged versus neutral collision rate coefficients. The hollow markers show the weighted mean values from the trimer to 11-mer, with ±1σ errors indicating statistical without (inner caps) and with systematic errors (outer). These are the first experimental measurements of the charge-enhanced collision rates during ion induced nucleation under atmospheric conditions.

These findings are important for our understanding of climate. Iodic acid particle formation can compete with sulphuric acid in pristine regions of the atmosphere. Moreover, it is a very efficient source of cloud condensation nuclei since a single vapour species drives both nucleation and rapid growth. Iodic acid particle formation is likely to be especially important in marine regions where sulphuric acid and ammonia concentrations are extremely low. Indeed, frequent new particle formation over the high Arctic pack ice has recently been reported, driven by iodic acid with little contribution from sulphuric acid. Global iodine emissions have increased three-fold over the last 70 years and may continue to increase as sea ice becomes thinner in future. The resultant increase of iodic acid cloud condensation nuclei could increase longwave radiative forcing from clouds and provide a positive feedback that accelerates the loss of sea ice in the Arctic.

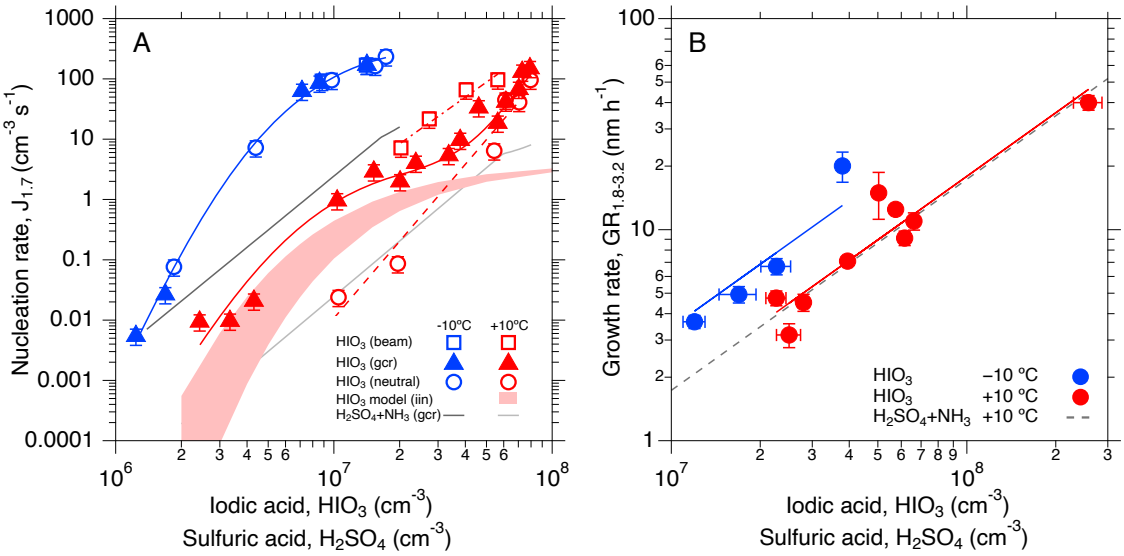

Fig. 1: Nucleation and growth rates of iodic acid (HIO_3) particles reported in *Science* [5]. a) Nucleation rates measured at 1.7 nm, $J_{1.7}$, versus HIO_3 concentration at +10°C and -10°C, and under three ionisation conditions: neutral, J_n (ions eliminated from the chamber by a 20 kV/m electric field); galactic cosmic ray, J_{gcr} (boundary layer ion pair concentrations of around 700 cm^{-3}) and beam, J_π (ion pair concentrations around 2500 cm^{-3}, comparable to the upper free troposphere). The nucleation rates show a strong dependency on HIO_3 concentration, charge and temperature. There is a large ion enhancement of the nucleation rate at +10 °C (e.g. J_{gcr}/J_n = 30 at 10^7 cm^{-3} HIO_3). The red band shows a model prediction for HIO_3 ion-induced nucleation, J_{iin} (= $J_{gcr} - J_n$) at +10 °C, at the kinetic limit. The nucleation rate of iodine oxoacids exceeds that of H_2SO_4–NH_3 at the same acid concentrations. b) Particle growth rates between 1.8 and 3.2 nm, versus HIO_3 concentration at +10 °C and -10 °C. The growth rates of HIO_3 particles at +10 °C are identical to CLOUD measurements for H_2SO_4–NH_3 particles.

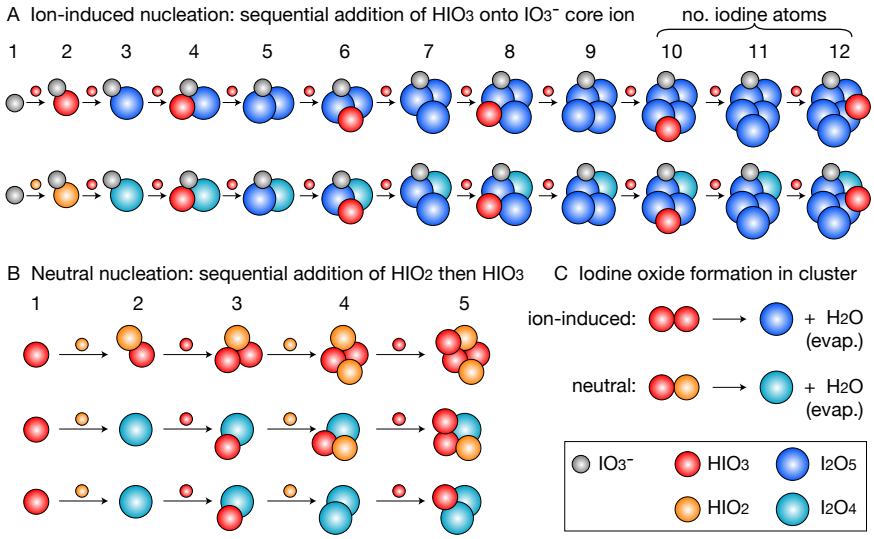

Fig. 2: Molecular composition of embryonic iodine oxoacid clusters during nucleation [5]. Schematic representations of the nucleation mechanisms for A) ion-induced (charged) and B) neutral (uncharged) iodine oxoacid clusters, derived from CLOUD measurements. Ion-induced nucleation involves condensation of iodic acid (HIO_3) alone onto an IO_3^- ion, whereas neutral nucleation involves repeated stepwise condensation of iodous acid (HIO_2) followed by iodic acid, revealing the previously-unknown importance of HIO_2 to stabilise embryonic iodic acid particles. Iodine oxide formation takes place in the clusters, as shown in panel C).

1.2 New mechanism for rapid particle formation in the upper troposphere

A paper from CLOUD published in *Nature* [11] reported synergistic particle formation in the upper troposphere from a novel mixture of vapours. Particle formation at high altitudes is enhanced by the low air temperature but limited by the availability of suitable vapours. Nevertheless, newly-formed particles are persistently observed throughout Earth's upper troposphere. However, the vapours and mechanisms that drive the formation of these particles are not understood.

CLOUD discovered a new mechanism for extremely rapid particle formation and growth in the upper troposphere via an unexpected synergy between nitric acid, sulphuric acid and ammonia vapours. We measured that these three vapours together form new particles 10–1,000 times faster than sulphuric acid–ammonia nucleation alone, which has previously been shown by CLOUD to be rate-limited by the scarcity of sulphuric acid in the upper troposphere. Once particles have formed, co-condensation of ammonia and abundant nitric acid alone is sufficient to drive rapid growth to larger sizes where they can seed clouds.

Moreover, CLOUD showed that these particles—even with trace amounts of sulphuric acid as low as 1%—are highly efficient at seeding ice crystals, comparable to desert dust particles, which are thought to be the most widespread and effective ice seeds in the atmosphere. When a supercooled cloud droplet freezes, the resulting ice particle will grow at the expense of any unfrozen droplets nearby, so ice has a major influence on cloud microphysical properties and precipitation (around 75% of global precipitation originates from ice particles).

The new mechanism discovered by CLOUD may be the dominant source of new particles in regions of the upper troposphere where ammonia is efficiently convected, such as over the Asian monsoon. Indeed, abundant ammonium nitrate particles have recently been reported in the Asian tropopause aerosol layer. Our global model simulations show that particles formed this way can spread across the mid-latitude Northern Hemisphere, influencing Earth's climate on an intercontinental scale. In the upper troposphere, nitric acid is abundant from lightning while ammonia originates from surface emissions—livestock and fertilizer—and is carried aloft by convective clouds and then released when droplets freeze. The atmospheric concentrations of all three vapours were much lower in the pre-industrial era, and each is likely to follow different trajectories under future air pollution controls. The new CLOUD results can inform policies for pollution regulations as well as improve the ability of global models to predict how the climate will change in future.

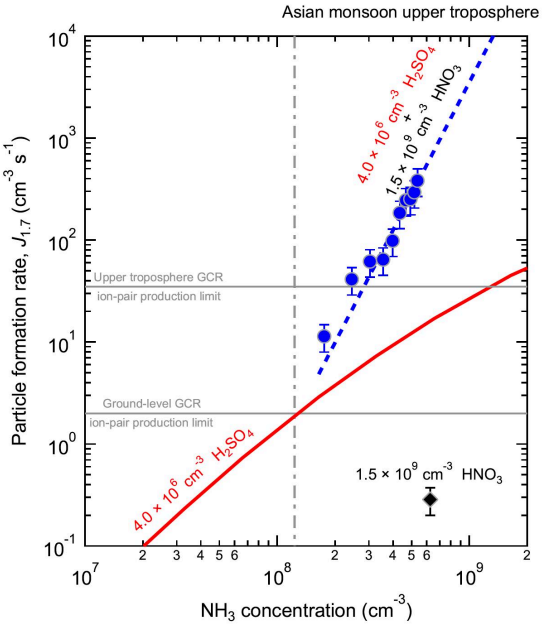

Fig. 4: Particle formation rates from nitric acid, sulphuric acid and ammonia at upper tropospheric conditions [11]. The chemical systems are HNO_3–NH_3 (black), H_2SO_4–NH_3 (red) and HNO_3–H_2SO_4–NH_3 (blue) at 223 K and 25% relative humidity. The black diamond shows the CLOUD measurement of HNO_3–NH_3 nucleation. The red solid curve is $J_{1.7}$ versus ammonia concentration from previous CLOUD measurements. Regions to the right of the vertical grey dotted line indicate the Asian monsoon conditions. The horizontal grey solid lines show $J_{1.7}$ upper limits for ion-induced nucleation resulting from the GCR ionisation rate of around 2 ion pairs cm^{-3}s^{-1} at ground level and 35 ion pairs cm^{-3}s^{-1} in the upper troposphere. Among the three nucleation mechanisms, H_2SO_4–NH_3 nucleation dominates in regions with low ammonia, whereas HNO_3–H_2SO_4–NH_3 nucleation dominates at higher ammonia levels characteristic of the Asian monsoon upper troposphere.

2 CLOUD15T RUN

During the East Area Renovation, 2019–2021, which took place during Long Shutdown 2 (LS2), the T11 beamline was completely rebuilt with new magnets, power supplies and optics. The East Hall experimental zones were also re-configured, providing substantially better access for instruments around the CLOUD chamber. All the experimental control rooms for the East Area beamlines were also replaced. CLOUD added a chemicals lab (including a fume hood) adjacent to the new T11 control room to handle the wide range of chemicals required to operate the facility and its instruments. The East Area Renovation necessitated the dismantling and rebuilding of almost all the CLOUD facility infrastructure including, in particular, the gas supply and distribution system. Trace gases for the chamber are provided either from evaporators (for chemicals that are liquid or solid at room temperature) or from bottles (for volatile vapours), which are housed in temperature-controlled cabinets outside the East Hall, adjacent to CLOUD's liquid nitrogen and liquid oxygen dewars. The rebuilt gas distribution system has been consolidated in a single area within the T11 experimental zone, located underneath a new platform extension.

The CLOUD15T run took place 28 Mar – 2 May 2022, after completion of the East Area Renovation and CLOUD rebuild. The "T" signifies "Technical" since the primary goal of CLOUD15T was to re-commission CLOUD and the T11 beamline after their re-build and to confirm operation at their previous performances. The characteristics of the T11 beam during CLOUD15T are shown in Fig. 5. The T11 beam intensity is 1.8 10^6 π^+ per spill at 3.5 GeV/c (Fig. 5a), which matches the previous performance. The wide transverse beam profile in x and y also meet design expectations (Figs. 5b and c).

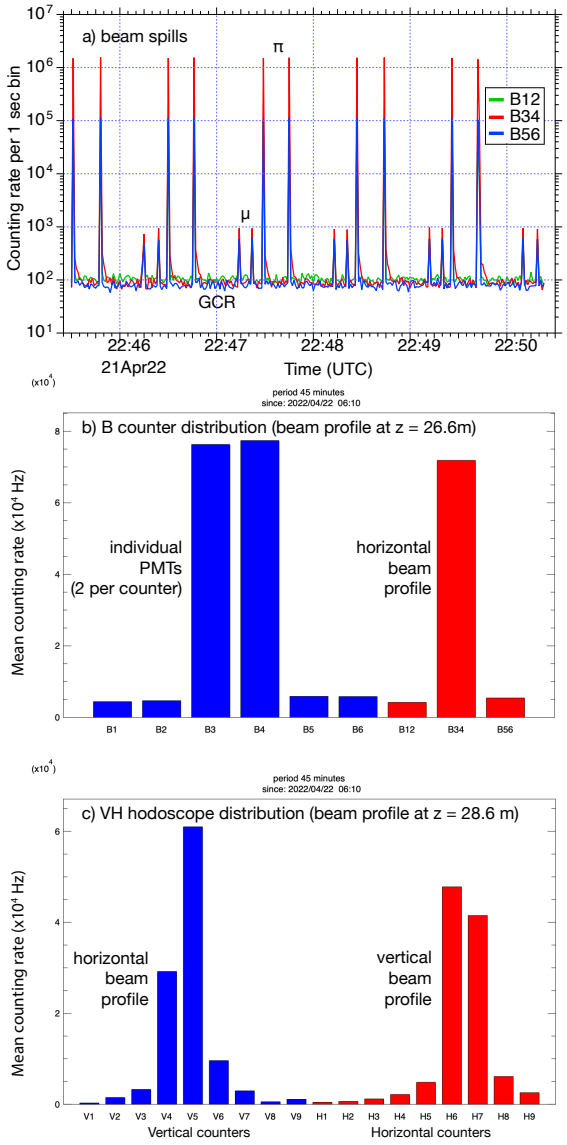

Fig. 5: T11 3.5 GeV/c π^+ beam characteristics. a) Beam flux at 2 spills per supercycle (59 s). Also evident are two low intensity μ spills per supercycle from PS extractions to another East Area target. Beam transverse profile measured in the b) B counters and c) VH xy hodoscope. Each counter is 20 cm width and the horizontal angle of the beam axis with respect to the normal of the counter planes is 32°. The z coordinates for these counters refer to their distance from the PS beam target for T11. For context, the T11 beam horizontal focus is at 23.1 m and the centre of CLOUD is 33.6 m from the target.

The CLOUD facility was also successfully re-commissioned during CLOUD15T (Figs. 6 and 7). In addition, initial experiments were carried out on marine aerosol particle nucleation from methanesulphonic acid (CH_3SO_3H, MSA) and ammonia. MSA is produced in the atmosphere along with sulphur dioxide and sulphuric acid from the oxidation of dimethyl sulphide (CH_3SCH_3, DMS), which is emitted from the ocean surface by phytoplankton. As the most abundant biogenic sulphur compound emitted to the atmosphere, DMS is a major precursor for aerosol particles in the marine atmosphere.

Fig. 6: CLOUD15T run (23 Mar - 2 May 2022). CLOUD and its analysing instruments in the T11 experimental zone during the CLOUD15T technical run to re-commission CLOUD following the rebuild of the T11 beamline, the CLOUD facility and its infrastructure during LS2. During operation, the T11 beam emerges from the concrete shielding blocks on the right hand side of the figure and passes through two counter planes (B and VH; the latter is seen mounted on the edge of the platform) 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. The HORUS instrument is in the left foreground.

Instrument	Institute
Mass Spectrometers:	
LTOF Nitrate-CIMS	UFRA
+/- APITOF	Aerodyne
PTR3	Innsbruck
PTR-STOF	Innsbruck
Particle counters/sizing:	
nano-SMPS (5-60 nm)	PSI
long-SMPS (15-600 nm)	PSI
CPC 3776 2.5 nm	PSI
PSM scanning	UHEL
NAIS	CERN
CIC	CERN
HOM-AIS	Tartu
Gas measurements:	
CO monitor	EMPA/PSI
O3 monitor	CERN/EMPA
SO2 monitor	UFRA
PICARRO (CO2)	PSI
PICARRO (NH3)	PSI
HORUS	MPI
Dew Point Mirror	CERN
TDL-Hygrometer (in situ)	KIT / CERN

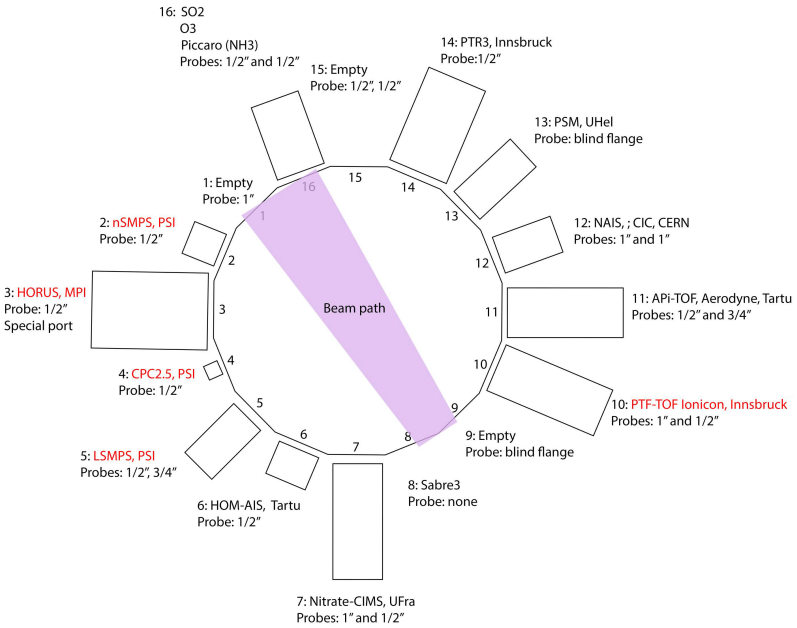

Fig. 7: CLOUD15T instrument layout. Layout of the analysing instruments around the chamber during the CLOUD15T run.

HORUS (HydrOxyl Radical measurement Unit based on fluorescence Spectroscopy): During CLOUD15T an important new instrument named HORUS was commissioned to measure hydroxyl (OH) and hydroperoxyl (HO_2) radicals (collectively referred to as HO_x). The most abundant oxidants in the atmosphere are O_2 and O_3 but these have large bond energies, leaving OH as the primary oxidising species in the atmosphere. The hydroxyl radical is produced by photolysis of ozone, $O_3 + h\nu \rightarrow O_2 + O(^1D)$, followed by reaction with water, $O(^1D) + H_2O \rightarrow 2$ OH. The reaction requires electronically excited atomic oxygen, $O(^1D)$, and so photolysis is restricted to wavelengths below 320 nm. The presence of OH in the troposphere underpins the contemporary understanding of the chemistry of the troposphere, and especially the relationship between emissions, air quality, aerosol particle formation and anthropogenic climate change. For example, OH concentrations in the atmosphere determine the 12-year lifetime of the important greenhouse gas, methane; without OH, methane and other vapours such as carbon monoxide would simply accumulate in the atmosphere. HO_x radicals also have a major influence on the chemical pathways that transform volatile vapours in the atmosphere into the extremely low volatility vapours that drive particle nucleation and growth. However, measurements of OH are highly challenging because daytime levels in the atmosphere (and in CLOUD) are around 0.1 pptv and the OH lifetime is 1 s or less. Nevertheless, MPIC Mainz has developed a sensitive instrument that detects the fluorescence of OH radicals after their excitation by a Nd:Yag pumped pulsed dye laser at 308 nm. A special sampling probe was designed to couple HORUS to the CLOUD chamber, and the instrument successfully measured both OH and HO_2 concentrations during CLOUD15T (Fig. 8).

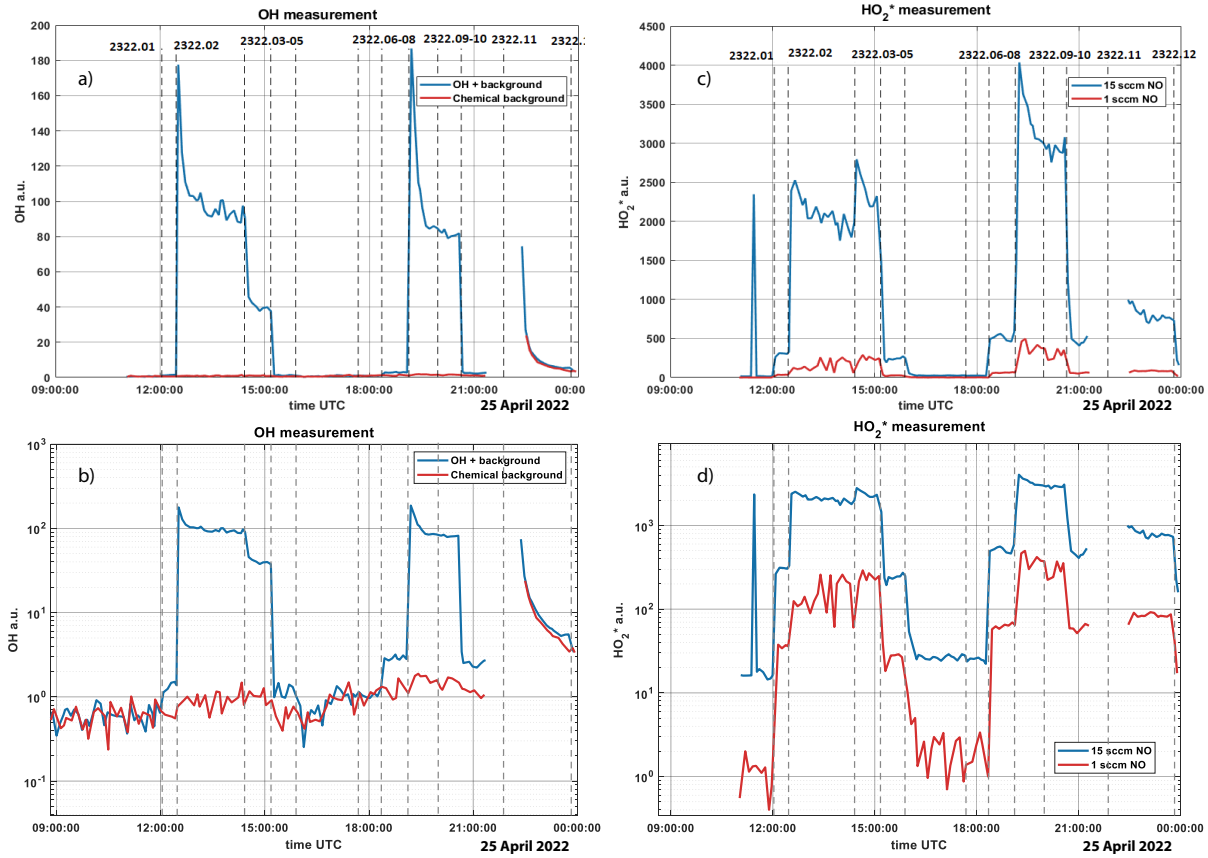

Fig. 8: Measurement by HORUS of the hydroxyl (OH) and hydroperoxyl (HO_2) radicals during CLOUD15T. The left-hand panels show OH measurements on a) linear and b) logarithmic vertical scales. The right-hand panels show HO_2 measurements on c) linear and d) logarithmic vertical scales. The vertical dashed lines indicate transitions of UV light sources in CLOUD, which control the photolysis rate of ozone in the chamber and hence the production rate and concentrations of HO_x (= OH + HO_2). The blue curves show the signal+background measurements of HORUS and the red curves show the background alone.

3 FLOTUS (FLOw TUbe System)

So far, CLOUD has been able to study rapid gas phase oxidation of organic precursor vapours on timescales of a few hours, limited by the wall loss lifetime of vapours and particles. At present, CLOUD is blind to a broad class of oxidation processes that are occurring in the real atmosphere on longer timescales, involving repeated independent oxidation steps.

However, CLOUD will soon be able to study aerosol particle formation and growth from these "aged" organic vapours using the FLOw TUbe System, FLOTUS, attached to the chamber (Fig. 9). FLOTUS is currently under construction at CERN and will become a permanent part of the CLOUD facility. It is a laminar-flow quartz drift tube of 3 m length and 20 cm diameter, with gas occupancy times of a few minutes. In the presence of ozone and intense UV light, FLOTUS simulates hydroxyl radical oxidative exposures of organic vapours equivalent to several days in the atmosphere. The output of FLOTUS will be transferred to the CLOUD chamber where the full power of its instrumentation will be used to study particle formation and growth with aged organics. The first operation of FLOTUS will be at CLOUD15 in fall 2022 (§5).

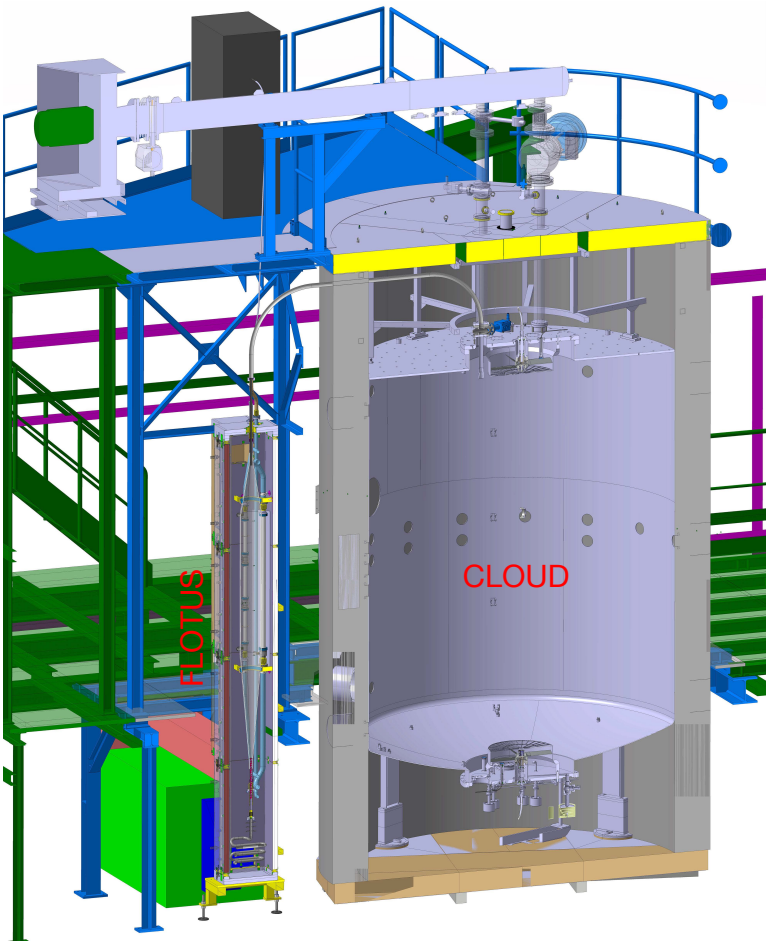

Fig. 9: FLOTUS (FLOw TUbe System) at CLOUD. FLOTUS is a 3 m laminar-flow quartz drift tube in which organic and other vapours receive hydroxyl radical exposures corresponding to several days in the real atmosphere. The outflow from FLOTUS is directed into the CLOUD chamber for studies with "aged" organics. All the operating conditions of FLOTUS (gases, UV irradiation, relative humidity and temperature) are controlled independently of the CLOUD chamber.

4 SCIENTIFIC GOALS 2022-2026

The CLOUD partners were recently informed by the European Commission that our CLOUD-DOC proposal for a new Marie Curie training network has received very favourable reviews and the project will be funded within the Horizon Europe Framework Programme. This will be the fourth consecutive Marie Curie training network for CLOUD, coordinated by Joachim Curtius, Goethe University Frankfurt. The grant of 2.7M € will support 12 PhD students on CLOUD during the period 1 Sep 2022 – 31 Aug 2026 (10 are EU-funded plus 2 by matching Swiss National Science Foundation funds). The scientific goals of the CLOUD-DOC proposal will be incorporated into the CLOUD experimental programme over this period. The overarching theme is to investigate new particle formation (NPF) taking place in cold regions of the atmosphere—which are extremely sensitive to particle concentrations and likely also to ions—namely polar and upper tropospheric regions, as follows:

Arctic: We know from field observations that NPF occurs frequently in Arctic areas, at least in summer. Recent observations show that Arctic/polar NPF and growth involve a complex mixture of chemical compounds: sulphuric acid, ammonia, iodic acid and methanesulphonic acid. The relative importance of these vapours varies with location and season. Due to the low condensation sink and low temperatures, NPF can occur at lower vapour source rates than those found at lower latitudes. However, since the particle growth rates in the Arctic are slow, it is also important to know which vapours are available for growth. Furthermore, the Arctic is currently warming at an unprecedented rate due to climate change and the atmospheric composition is affected by the declining sea ice and melting permafrost. For example, global iodine emissions have increased three-fold over the last 70 years and may continue to rise in the future with the thinning of sea-ice. The research questions we will address include:

- What is controlling NPF rates in different parts of the Arctic and in different seasons?
- How is NPF in polar regions expected to change in future with increasing temperature? Do we expect the NPF rates to decrease (due to less pollution and higher temperatures) or increase (due to increasing iodine and volatile organic compounds)?
- What is the role of ions in the Arctic?
- Can secondary particles from polar NPF act as ice nucleating particles?

Upper troposphere in the monsoon outflow over Asian megacities and agricultural regions: NPF in the free troposphere is a major global source of cloud condensation nuclei (CCN). However, what drives the process in the relatively cold upper troposphere remains poorly known, especially over the tropical convective regions where abundant newly formed particles have been consistently observed. Almost half the world's population lives in the inflow region of the Asian monsoon. Ammonia transported by the Asian monsoon convective systems to the upper troposphere can drive intense nucleation and growth of ammonium nitrate/sulphate particles to CCN sizes. These particles are long-lived and so can be transported by the subtropical jet stream far outside the region of formation and growth, potentially influencing cirrus cloud formation on continental scales and beyond. Other potentially important vapours are aromatic organics, which are ubiquitous in the polluted Asian boundary layer. These vapours are potent precursors of low volatility oxygenated organic compounds, but only after multi-step oxidation. Thus, their oxidation products can be readily transported to the upper troposphere, where they can contribute to NPF. Fundamental questions remain about the transport efficiency of ammonia and aromatic species, their role in upper tropospheric NPF, and their impact on the climate. The research questions we will address include:

- *NPF from nitric acid, sulphuric acid and ammonia:* Using the CLOUD chamber, we will investigate synergistic NPF and growth by nitric acid, sulphuric acid and ammonia under upper tropospheric conditions. The uptake efficiency of NH_3 in supercooled cloud droplets formed in the CLOUD chamber through adiabatic expansions at various temperatures will also be studied at different cloud water pH values. Parameterisations of the experimental data will be incorporated in a state-of-the-art global climate-chemistry model (EMAC).
- *NPF from aromatic precursors:* Using the FLOTUS-CLOUD combination, we will measure the oxidation chemistry and particle formation and growth from aromatic precursors during transport to the monsoon upper troposphere. Parameterisations of the experimental data will be incorporated in EMAC.
- *Atmospheric and climate implications:* Using EMAC and process models for convective cloud transport, we will quantify the climatic effects of NPF on global cloud condensation nuclei (CCN),

ice nucleating particles (INP) and radiative forcing, and quantify the climate impact of agricultural ammonia emissions. Our results will inform emissions control policies.

Upper troposphere above pristine tropical rainforests: Although NPF is not observed in the boundary layer of pristine tropical rainforests such as the Amazon, high concentrations of freshly-formed particles are found in the upper troposphere overhead. Deep convective clouds are thought to act as a pump of precursor vapours from the boundary layer to the upper troposphere . Particles formed at high altitudes in the outflow of convective clouds may subsequently be carried downwards to provide a source of particles at the surface, but large gaps in knowledge remain. To build an accurate picture of the role of convective clouds as sources of new CCN, we need to understand the multicomponent chemistry involving isoprene, monoterpenes and sesquiterpenes with different atmospheric oxidants, as well as the uptake and release of the gases and their oxidation products to the liquid and ice hydrometeors during convective transport. Studies coupling all the relevant pieces in this complex puzzle do not exist to date but CLOUD is ideally suited to addressing the problem. The research questions we will address include:

- What are the multicomponent NPF pathways relevant for the tropical upper troposphere (including neutral and ion-induced mechanisms)?
- What are the processes controlling the convective transport and the simultaneous chemical evolution of gas-phase species found in tropical rainforests?
- What is the importance of NPF in the outflow of convective clouds in the tropical upper troposphere as a source of CCN at lower altitudes?

Southern Ocean: Southern Ocean clouds are poorly represented in current climate models that informed the recent 6th Assessment Report of the Intergovernmental Panel on Climate Change (IPCC). Modelled clouds overestimate the amount of sunlight reaching the ocean surface, causing a warm sea-surface temperature bias. This results in an underestimation of the sea ice extent, a displaced location of the ocean circulation belts, and a southward displacement of storm tracks. Such misrepresentations of cloud properties reduce the precision of climate models to reproduce important global climate properties such as Earth's climate sensitivity. A major part of the problem is incomplete knowledge of the microphysical properties (e.g. droplet size distribution) and phase state (liquid or frozen) of Southern Ocean clouds, which are influenced by the combined effects of aerosol particles and boundary layer dynamics. An important unknown is the source of CCN in the Southern Ocean. There is evidence for a significant fraction of CCN originating from new particle formation from gaseous precursors, and subsequent growth to CCN sizes by condensation of organic and inorganic vapours. However, new particle formation is rarely observed in the marine boundary layer, suggesting that CCN are forming in the free troposphere and then mixing down into the marine boundary layer where they become available for cloud formation. Concerning ions, observations suggest that ions and variations of the fair-weather electric field are affecting meteorological conditions over Antarctica such as ground level pressure and temperature. The mechanism is unknown but may involve cloud formation from vapours and particles transported from the Southern Ocean to the Antarctic interior by the katabatic circulation. The research questions we will address include:

- NPF and growth under conditions found over the Southern Ocean
- Comparison of the CLOUD data with ship and research station campaigns in the Southern Ocean and Antarctic, e.g., ACE-SPACE25 (Antarctic Circumnavigation Experiment – Study of Preindustrial-like Aerosol Climate Effects).

- Large-scale modelling of the Southern Ocean and Antarctic region using CLOUD data, including process model studies.
- Investigation of the effects of charge on new particle formation and on cloud microphysics under Southern Ocean conditions.

5 BEAM REQUEST 2022

CLOUD requests 9 weeks of beam in fall 2022 (Sep/Oct/Nov) for the CLOUD15 run. The science goals are as follows:

1. **Pure biogenic nucleation with aged organics:** We will use FLOTUS to age mixtures of biogenic vapours (alpha-pinene and isoprene) corresponding to up to several days OH exposure in the atmosphere, and inject these aged vapours into the CLOUD chamber. Using the full power of the analysing instruments attached to CLOUD, we will study the chemistry of these aged organic vapours and their ability to form and grow new particles under neutral and charged (GCR and π beam) conditions. We will compare these measurements with previous CLOUD experiments involving fast (below one hour) autooxidation of the same biogenic vapours under otherwise similar conditions.
2. **Carbon closure:** As part of the above study, and together with the new HORUS instrument to measure HO_x radicals, we will seek to explain quantitatively the fate of *all* carbon molecules injected into FLOTUS or CLOUD—so-called 'carbon closure'. Our ability to track the oxidative evolution of organic carbon over its entire atmospheric lifetime is critical for the understanding the impact of organic emissions on human health and Earth's climate.
3. **Marine nucleation and growth with methanesulphonic acid ($\mathbf{CH_3SO_3H}$, MSA):** We will extend our initial CLOUD15T studies of MSA and ammonia new particle formation and growth to vapour and ion concentrations characteristic of marine and polar regions between the boundary layer and upper troposphere.
4. **Iodic acid nucleation and growth under upper tropospheric conditions:** We will study new particle formation and growth involving iodine vapours at upper tropospheric conditions: low temperatures (-30°C $\rightarrow$ -50°C), low vapour concentrations and high ion concentrations.

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