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CLOUD Collaboration

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1 SCIENCE HIGHLIGHTS

An overview of the scientific contributions made over the last ten years by the Cosmics Leaving OUtdoor Droplets (CLOUD) experiment at CERN (1; 2), and the projected scientific programme over the next ten years, are provided in Addendum 5 of the CLOUD proposal (3). CLOUD published 7 papers in 2018 (6)–(12) and has published 1 paper so far in 2019 (13). Four more papers are under review (14)–(17).

Experiments at CLOUD have shown that there is a limited set of atmospheric vapours with sufficiently low volatility to form new particles. They comprise sulphuric acid, extremely low volatility organic compounds (ELVOCs, which form a subset of highly oxygenated molecules, HOMs, from biogenic and anthropogenic organic precursors), and iodine oxides. However, in most regions of the lower atmosphere, these vapours are insufficient to nucleate alone, and they require additional stabilising vapours such as ammonia, amines or ions. Ions are especially important in stabilising new particles involving semi-volatile organic vapours, and may enhance their formation rates by up to one or two orders of magnitude. However, although the main players in atmospheric aerosol nucleation and growth may have been identified, there remain many unanswered questions and barely-explored areas—including the role of ions. Moreover, in contrast with single-species inorganic vapours such as sulphuric acid or iodic acid, the HOMs comprise hundreds and even thousands of different organic vapours with different

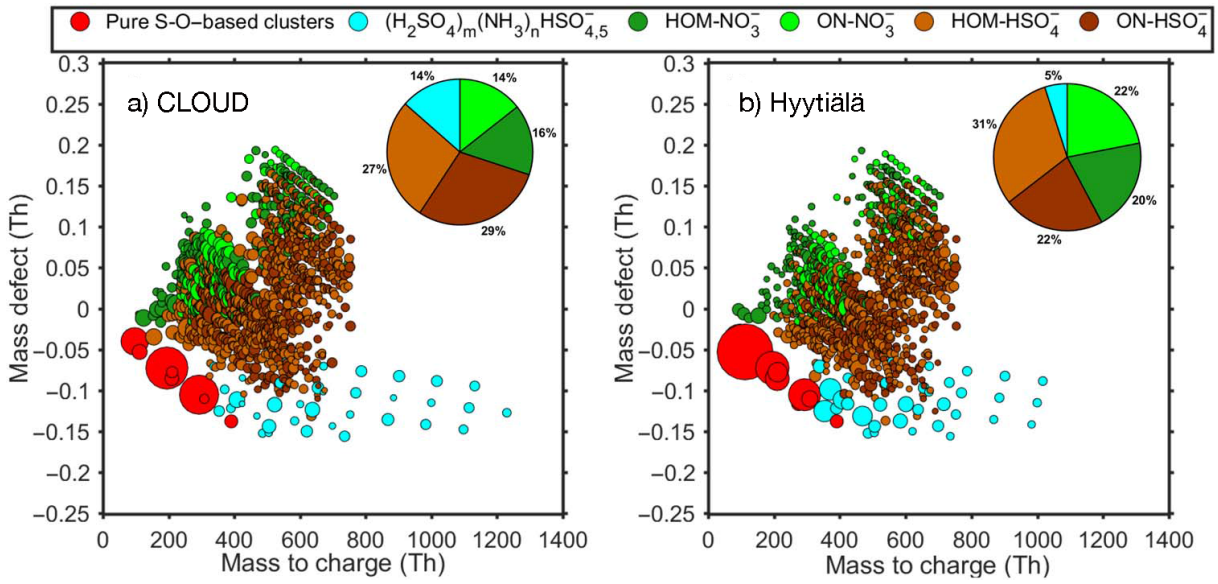

Fig. 1: Simulation of boreal forest new particle formation in the CLOUD chamber: Negatively-charged ions and molecular clusters during new particle formation events measured a) in the CLOUD chamber and b) in the boreal forest at Hyytiälä, Finland. The x axis shows the cluster mass, and the y axis shows the mass defect (difference of the cluster mass from integer mass). Each circle represents a distinct ion or charged molecular cluster, and the diameter corresponds to the signal intensity on a logarithmic scale. The CLOUD data are averaged over several experiments with 10^6–10^7 cm^{-3} H_2SO_4, 1 ppbv NOx, 200–500 pptv NH_3, 600 pptv monoterpenes, and 40 ppbv ozone, at 38% relative humidity and 278 K. The colours indicate the molecular composition of the ions and clusters: pure sulphuric acid and SO_5^- (red), H_2SO_4-NH_3-HSO_4^- (cyan), HOM-NO_3^- (dark green), organonitrate-NO_3^- (light green), HOM-HSO_4^- (light brown), and organonitrate-HSO_4^- (dark brown). The pie charts indicate the relative amounts of all identified species, excluding the pure S–O species. With a multicomponent mixture of sulphur dioxide, ammonia, monoterpenes, NOx and ozone, CLOUD was able to simulate closely the new particle formation and growth rates, and the gas-phase chemical composition, observed in the boreal forest at Hyytiälä (11).

volatilities—and, in addition, the volatilities of *all* vapours are strongly temperature-dependent. This illustrates the breadth and complexity of the parameter phase space that CLOUD is investigating.

During recent runs, CLOUD has carried out experiments that simulate specific environments in order to interpret the atmospheric observations made at those locations. A major effort has been devoted to reproducing boreal forest conditions, which have been characterised in great detail over the last twenty years at the Hyytiälä Forestry Field Station, Finland. Figure 1 shows a comparison of the negatively-charged ions and molecular clusters in CLOUD and at Hyytiälä, when the environmental conditions are matched (11). CLOUD has closely reproduced the particle formation and growth rates observed at Hyytiälä, and shown that the key process driving new particle formation is a multicomponent mechanism involving sulphuric acid, ammonia and biogenic HOMs. NOx—which largely arises from anthropogenic sources—substantially reduces the particle formation rates by prematurely terminating the self-oxidation mechanism that leads to the lowest-volatility HOMs, to form less-volatile organonitrates (Figure 1). On the other hand, ammonia (again, largely from anthropogenic sources) strongly enhances the particle formation rates, as do ions, up to the limit set by the ion-pair production rate of a few cm^{-3} s^{-1}.

The impact of aerosol particles on Earth's radiative balance involves both direct effects (by scattering and absorbing light) and indirect effects (through their influence on cloud microphysics). The indirect effects are thought to be the more important, but are less well understood. A nucleated particle

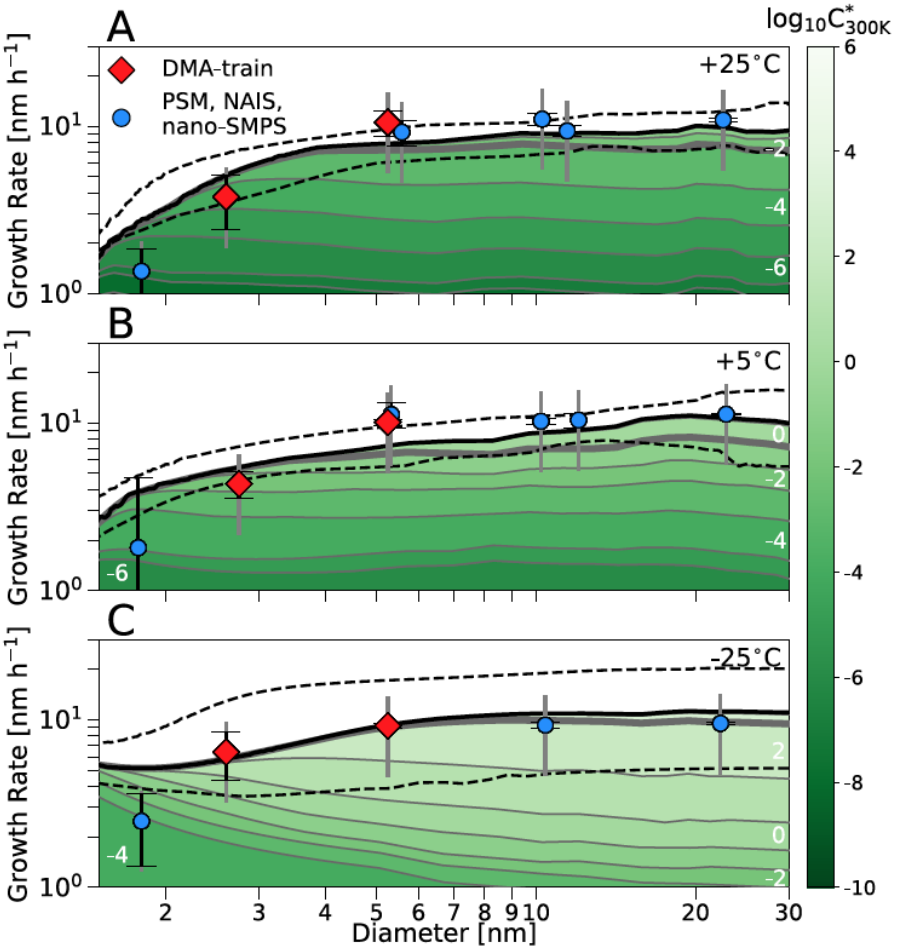

Fig. 2: Modelled and measured grow rates of biogenic particles: Examples of modelled and measured growth rates of biogenic particles versus size for 3 experiments at a) +25°C, b) +5°C and c) -25°C. The experiments are at made under comparable conditions for reaction rates of α-pinene ozonolysis [$(1.8 \pm 0.5)10^6$ cm^{-3}s^{-1}]. The data points show growth rates measured by the DMA-train (red) and PSM/NAIS/nano-SMPS (blue) instruments. The modelled contributions from highly oxygenated organic molecules (HOMs) to the growth rates are indicated by the green bands. The contributions are directly derived from gas-phase HOM measurements, without any adjustments. The HOMs are binned according to their saturation mass concentrations, $\log(C^\star_{300K}$ [μg m^3]). The HOM volatilities are calculated from the measured atomic and molecular compositions and are experimentally verified by particle-phase volatility measurements with the FIGAERO CI-APiTOF. The uncertainties in the model calculations (± 1 in $\log\ C^\star_{300K}$) are indicated by dashed lines. The data reveal several important features of organic particle growth: i) suppression of the growth rate at small sizes due to the Kelvin (curvature) effect, and ii) fast organic growth rates over a wide temperature range, which result from iii) near-complete compensation of the reduced oxygenation (higher $C^\star_{300K}$) at lower temperatures by the lower overall volatilities. (The reduced oxygenation is due to a slowing of the self-oxygenation rate.) To state the last point another way: organic vapours with 5–6 orders-of-magnitude higher volatility at 300 K can drive particle formation and initial growth at 248 K. This is the first time complete closure (agreement) has been achieved between biogenic particle growth rates and gas-phase HOM measurements (9).

must grow to sizes above around 50 nm before it can act as a cloud condensation nucleus (CCN). So, as well as nucleation rates, particle growth rates need to be fully characterised in order to understand the impact of aerosols on clouds and climate. The CLOUD institutes have developed several state-of-the-art instruments dedicated to particle size measurements in the critical region between molecular sizes near 1 nm and 10–20 nm, where commercial instruments are available. They include the U Helsinki scanning PSM, the U Vienna DMA-Train and the Caltech nano-SEMS. These counters have uncovered a treasure trove of new results during recent CLOUD runs. One example is shown in Fig. 2 (9).

2 CLOUD13T run, 11 June – 9 July 2018

2.1 CLOUD13T analysing instruments

Since the entire CLOUD13T technical run was dedicated to ion studies, only a few sampling instruments were connected to the chamber. They comprised two Cluster Ion Counters (CIC), a Neutral cluster and Air Ion Spectrometer (NAIS), and two gas analysers (SO_2 and ozone). One CIC counter sampled from the mid-plane of the chamber, and the other from a port located near the upper HV electrode (which was set at 0 V for the entire run). Experiments were performed for a wide range of conditions of beam intensity, fan settings, relative humidities and temperatures.

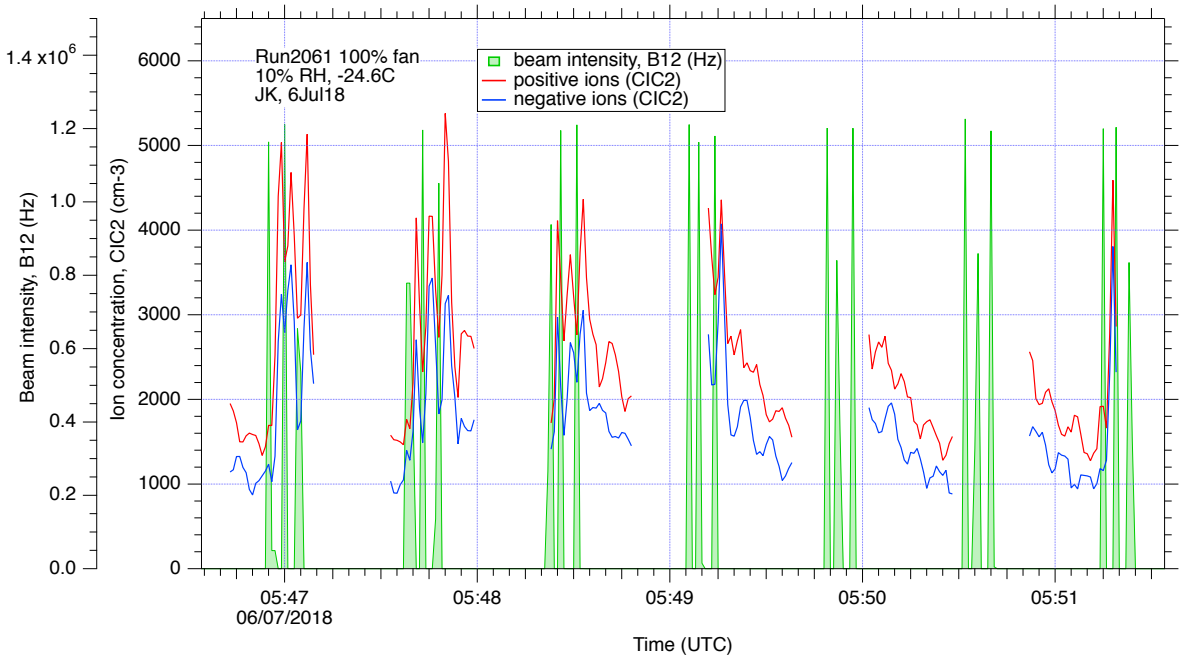

Fig. 3: Ion bursts in the CLOUD chamber during CLOUD13T: Small-ion concentrations measured during CLOUD13T, 6 July 2018. The green trace shows the beam intensity at 1 Hz sampling rate; three bursts can be seen per 43 s supercycle of the CERN PS. The red and blue traces show the positive and negative small-ion concentrations, respectively, measured by a Cluster Ion Counter (CIC) sampling air from the mid-plane of the chamber. The CIC records continuously at 1 Hz rate for 27 s and then calibrates for the following 24 s, resulting in data gaps. The difference between the supercycle and CIC periods leads to complete sampling of the small-ion time structure after four supercycles. The measurements show sharp spikes in ion concentrations when the beam passes through the chamber, followed by rapid mixing and dilution of the ions on timescales of a few tens of seconds. The 3 s delay between passage of the beam pulse and the corresponding ion spike is due to the finite time for ions to reach the entrance of the sampling probe and pass out of the chamber into the CIC.

2.2 CLOUD13T scientific programme

The CLOUD13T run took place 11 June – 9 July 2018, with the first week for instrument setup and the following three weeks for data taking. The studies comprised:

1. **Ion production and loss rates.** We measured ion production and loss rates as a function of variables such as beam intensity, mixing fan speeds, relative humidity and temperature. We compared wall loss rates of ions with those for neutral sticky molecules such as sulphuric acid. These measurements will provide precise measurements of the ion-ion recombination coefficient as a function of relative humidity and temperature, which is a fundamental quantity of importance for understanding atmospheric ion concentrations.

2. **Ion non-uniformities in the CLOUD chamber.** The 3.5 GeV/c π beam from the CERN PS deposits ions in the 3 m CLOUD chamber within a volume of about 1.2 m transverse size. Furthermore the ions are deposited in a few (3) intense pulses per supercycle of 30–40 s. The ionisation is then distributed throughout the CLOUD chamber by a combination of diffusion and bulk transport by the internal mixing fans. During CLOUD13T we measured the ion non-uniformities in the chamber under various beam and mixing fan conditions (an example is shown in Fig. 3). We will compare the measurements with our Computational Fluid Dynamics (CFD) simulations and with air flow measurements inside the CLOUD chamber, and then use the results to assess the impact of non-uniformities of ions and vapours on CLOUD measurements.

During the CLOUD13T run, the Huber chiller that controls the chamber temperature failed and so measurements could not be made over the full temperature range initially planned (although data were recorded at -50°C for several days). Nevertheless, the quality of the ion measurements is excellent, and a comprehensive analysis of ion production and loss rates is underway, including a comparison of ionisation in the CLOUD chamber with atmospheric ion measurements made at field stations.

3 CLOUD13 run, 17 September – 28 November 2018

3.1 Analysing instruments and facility upgrades

Around 40 analysing instruments were attached to the chamber during CLOUD12, including 11 mass spectrometers, 18 particle counters/sizers, 9 gas analysers and 5 light sources (Figs. 4, and 5). Upgrades of the CLOUD facility for the CLOUD13 run included:

Light sabre 3: LS3 is a 400W optical power UV sabre (385 nm) for NO_2 photolysis to NO, and HONO photolysis to OH· radicals. It was damaged during CLOUD12 and repaired for CLOUD13, together with a new chiller to remove the 1.2 kW parasitic heat. A hardware failsafe was installed to prevent any possibility of overheating in case of chiller failure.

Light sabre 1: LS1 is a 50W UV sabre (254 nm, not adjustable) for photolysis of ozone to produce hydroxyl radicals (OH·). It was equipped with a new control and cooling system for CLOUD13.

CLOUD DAQ and slow control: A new DAQ architecture, new servers and several new slow control systems were introduced for CLOUD13, together with a new software tool for retrieving and analysing CLOUD data in quasi real time. The CLOUD DAQ and slow control systems performed excellently throughout the entire CLOUD13 campaign.

New gases: New gases for the CLOUD13 run included dimethylsulphide, nitric acid, cresol and glyoxal. The nitric acid vapour was generated with an ultra-clean inox evaporator containing 70%w/w HNO_3 at a precisely-controlled temperature. In addition, the HONO generator was re-installed, together with improved control of the rate of delivery of reactants (sodium nitrite and sulphuric acid). Photolysis of HONO provided an intense source of OH· radicals for the urban experiments.

Fig. 4: CLOUD13. CLOUD with its analysing instruments in the East Hall, viewed from vertically above the beam axis, 19 Oct 2018 (Max Brice, CERN).

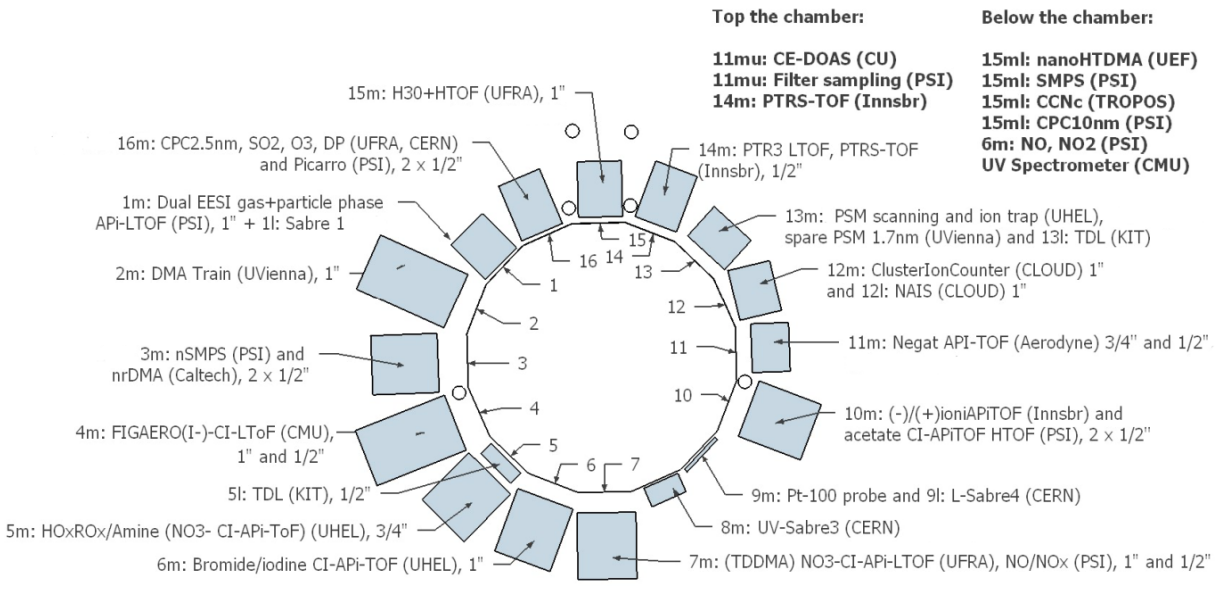

Fig. 5: CLOUD13 instrument layout. Layout of the analysing instruments around the chamber during CLOUD13, Sep-Nov 2018. The beam enters the chamber in the region between ports 7 and 10.

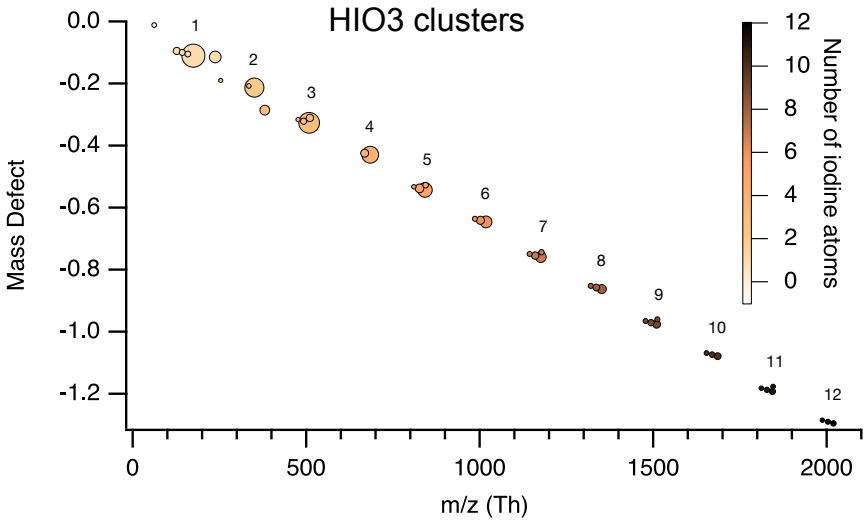

Fig. 6: Coastal marine nucleation of iodic acid particles during CLOUD13: Nucleating iodic acid (HIO_3) molecular clusters measured with the Br^- CI-APiTOF. The iodic acid derives from gas-phase oxidation of molecular iodine during experiments to simulate coastal marine conditions. The x axis shows the cluster mass, and the y axis shows the mass defect (difference of the cluster mass from integer mass). The high-resolution mass measurements identify the precise atomic/molecular composition of each of the clusters, which grow to contain up to 12 iodine atoms (corresponding to 1.35 nm mobility diameter).

T11 experimental zone improvements: Several improvements were made to the T11 experimental zone, in particular a heat-extraction system to remove the hot air produced by the close-packed analysing instruments above and below the platform.

3.2 CLOUD13 scientific programme

The CLOUD13 run took place 17 September – 26 November 2018, with the first week involving instrument setup and the final two weeks using cosmic rays (no East Area beams)—a 9-week data period in total. The aims of the CLOUD13 run were to extend and explore the new findings from CLOUD12, as follows:

1. **Marine nucleation and growth involving iodine compounds and dimethylsulphide.** Experiments were carried out to elucidate the chemical mechanism for producing iodic acid (HIO_3) in the atmosphere. Iodic acid has extremely low volatility and homogeneously nucleates under atmospheric conditions (Fig. 6). We also investigated marine nucleation in the presence of dimethylsulphide (DMS). DMS is produced by phytoplankton and is the largest natural source of sulphur dioxide in the atmosphere, accounting for around 20% of global SO_2 in the absence of volcanoes.

2. **Multi-component aerosol particle nucleation and growth.** We completed the measurements needed to parameterise multicomponent nucleation over the entire troposphere for global aerosol and climate models. Multi-component nucleation involves four basic components: $H_2SO_4 - NH_3 - HOM - H_2O$, where HOM refers to Highly Oxygenated biogenic Molecules. The nucleation and growth rates from these vapours were measured as a function of NO_x, relative humidity, temperature and ions. We also extended our measurements with biogenic vapours at low temperatures since recent observations from research aircraft indicate the presence of abundant particle nucleation in the tropical free troposphere at 8–12 km altitude in the outflows of deep convective clouds. For the first time, we also studied the influence of nitric acid on multi-component nucleation and urban nucleation in the presence of ammonia. Since nitric acid is a volatile vapour, it

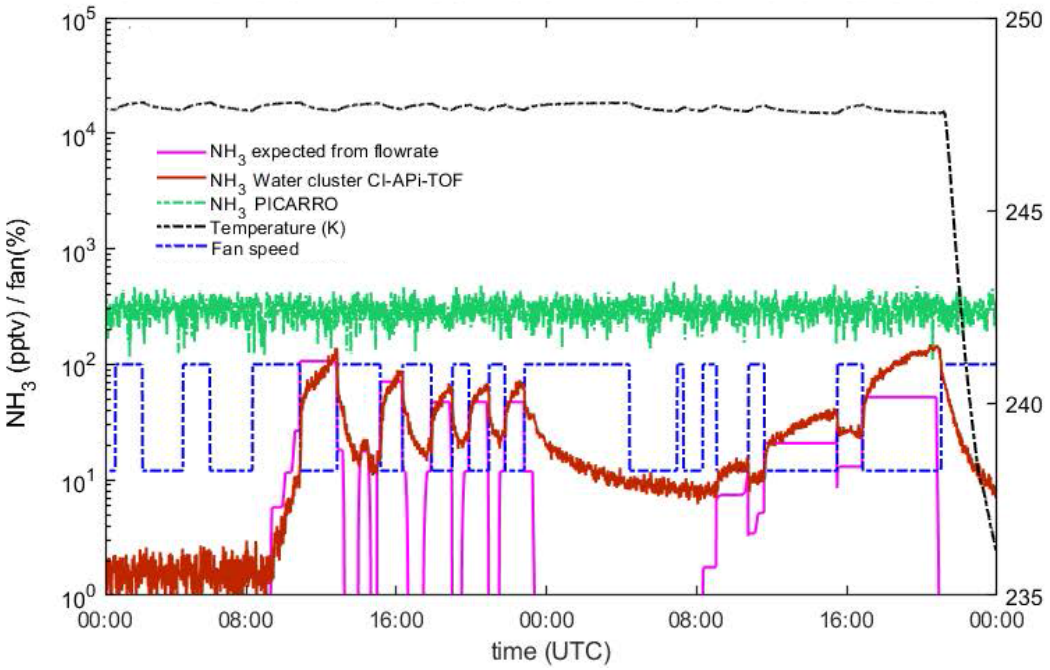

Fig. 7: High-sensitivity real-time ammonia measurements during CLOUD13: Ammonia measured with the newly-developed H_3O^+ CI-APiTOF (17). The brown curve shows gas-phase ammonia measurements from the H_3O^+ CI-APiTOF at 0.25 s sampling rate. The magenta curve shows the ammonia concentrations expected from the mass flow controller settings, assuming ammonia is permanently lost on colliding with the chamber walls. The dot-dashed blue trace shows the mixing fans speeds, which were varied between 12% and 100% (the wall loss rate increases by a factor 5 in going from 12% to 100%). The green trace shows a commercial ammonia analyser, which has a limit of detection of around 300 pptv. The limit of detection of the H_3O^+ CI-APiTOF is near 0.5 pptv for ammonia, and about 0.1 pptv for dimethylamine (not shown). The brown NH_3 trace shows a low contaminant level at the start and close initial agreement with the expected ammonia concentrations, but with a gradual excess appearing towards the end of the 2-day period. This signifies gradual conditioning of the chamber walls as they adsorb NH_3 and it starts partially to evaporate. These are the most sensitive gas-phase ammonia measurements so far achieved by any instrument, and represent a major advance for CLOUD. For comparison, CLOUD's limit of detection for NH_3 in Kirkby *et al.*, Nature, 2011, was 35 pptv, and each measurement required around 1 hour sampling time (4; 5).

is generally not thought to be important for particle nucleation. However it is often present in the atmosphere in quite large mixing ratios of up to several 100 pptv, whereas sulphuric acid levels are typically around only 0.1–1 pptv, even under relatively polluted conditions. For these experiments we required highly-sensitive ammonia measurements, which were beautifully achieved with a new H_3O^+ CI-APiTOF developed by the Frankfurt CLOUD team (Fig. 7 (17)).

3. **Anthropogenic aerosol particle nucleation and growth.** During CLOUD13 we extended the previous CLOUD11/12 studies of particle nucleation and growth from the anthropogenic volatile organic compounds to a new aromatic vapour, cresol (C_7H_8O), together with toluene (methyl benzene, $C_6H_5.CH_3$). The cresol vapour was generated from an inox evaporator. Nucleation and growth were, in particular, studied under high condensation sink conditions to understand why new particle formation is observed in Chinese megacities under such conditions, in contradiction with expectations.

Overall, CLOUD13 was a highly successful run which achieved its goals and included several major results that will shortly be submitted to Nature/Science (as could be inferred from Figure 8).

Fig. 8: CLOUD13 experimenters: CLOUD experimentalists at a "3 o'clock meeting" at the end of the CLOUD13 run, 28 November 2018.

4 CLOUD MEETINGS, 2018–2019

The following CLOUD Collaboration meetings and data workshops were held and are planned in 2018–2019:

CLOUD12 data workshop, Innsbruck, Austria, 5–9 Feb 2018. Analysis of data from CLOUD10–12, discussion of CLOUD manuscripts and planning for CLOUD13T and CLOUD13 runs.

CLOUD12 data workshop, Cascais, Portugal, 4–8 June 2018. Analysis of data from CLOUD10–12, discussion of CLOUD manuscripts and planning for CLOUD13T and CLOUD13 runs.

CLOUD-MOTION summer school, Hyytiälä Field Station, Finland, 13–23 August 2018. Summer school on aerosol nucleation, growth, and climate effects, together with practical sessions on CLOUD data analysis.

CLOUD13 status and CLOUD collaboration meeting, CERN, 24–26 Oct 2018. Assessment of the CLOUD13 run in progress, followed by a CLOUD collaboration meeting and Finance Review Committee meeting (FRC7).

CLOUD13 data workshop, U Stockholm, Sweden, 11–15 Feb 2019. Analysis of data from CLOUD10–13, discussion of CLOUD manuscripts and planning for CLOUD14.

CLOUD-MOTION mass spectrometer workshop, U Wuppertal, Germany, 25–28 Mar 2019. Theory and practice of chemical ionisation mass spectrometers for atmospheric research.

CLOUD-MOTION summer school and CLOUD collaboration meeting, Wengen, Switzerland, 14–20 June 2019. Summer school on climate modelling, clouds, ice particles, aerosol-cloud interactions, atmospheric chemistry, and urban particle formation, together with practical sessions on scientific writing. Planning for CLOUD manuscripts and CLOUD14 experiments.

CLOUD14 status and CLOUD collaboration meeting, CERN, 29 Oct – 1 Nov 2019. Assessment of the CLOUD14 run in progress, followed by a CLOUD collaboration meeting and Finance Review Committee meeting (FRC8).

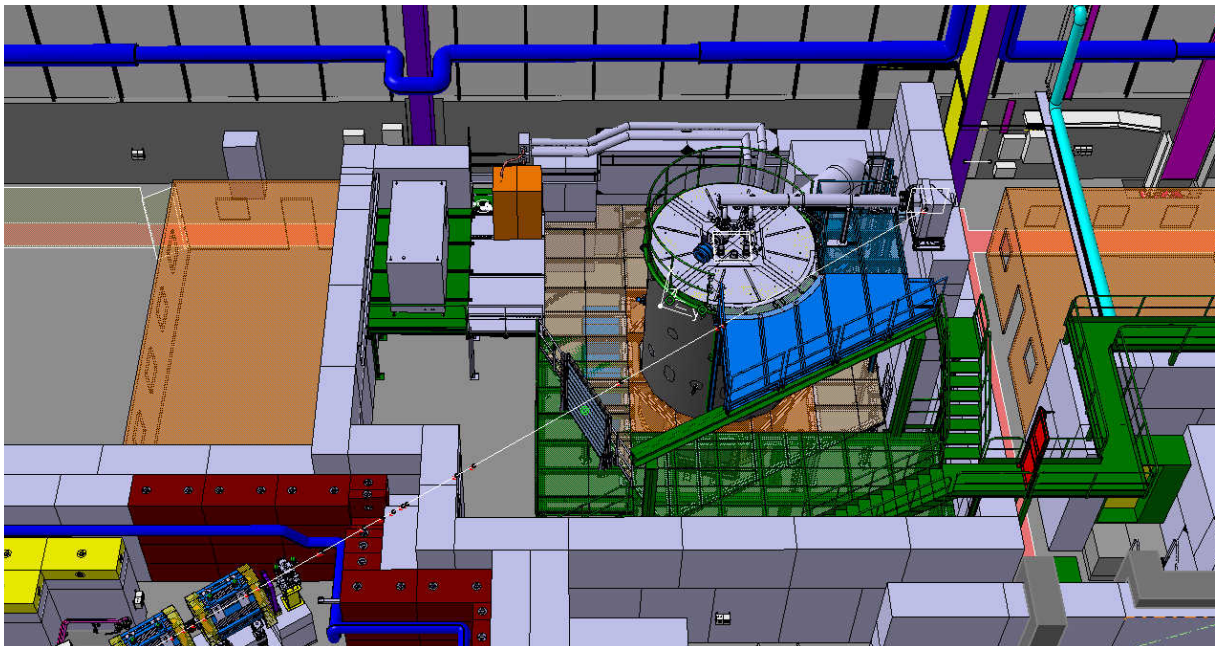

Fig. 9: Model drawing of CLOUD in the T11 zone at the CERN Proton Synchrotron after completion of the East Area renovation, 2019–2020. The improved space for sampling instruments can be seen on the green-shaded section of the platform. A major re-configuration of the control room, chemistry lab, gas system and other CLOUD facility systems will take place after LS2 to benefit from the improved space around CLOUD.

5 COSMIC RAY RUN, 2019

5.1 CLOUD14 cosmic ray run request, 16 September – 25 November 2019

The CLOUD14 'CLOUDy' run is planned for 16 September – 25 November 2019, using cosmic rays (no East Area beams during LS2, 2019–2020)—a 9-week data period in total. The aims of the CLOUD14 run are as follows:

1. **Activation properties of secondary aerosol for cloud droplets and ice particles:** Aerosol particles will be nucleated and grown in the CLOUD chamber from vapours under various conditions (chemical species, relative humidity, temperature, and ion concentrations). A wide range of secondary aerosol will be investigated (inorganic, pure biogenic, multicomponent, marine, and urban). The cloud activation properties will be investigated for liquid droplets (cloud condensation nuclei, CCN) and ice particles (ice nuclei, IN).

2. **Effect of charge on cloud microphysics:** A new CCN generator is being developed to produce highly charged CCN of either polarity. Expansion (CLOUDy) experiments will be performed with highly-charged CCN and compared with similar experiments using uncharged CCN. There are no experimentally-established fair weather charge effects 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.

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

The CLOUD Collaboration wishes to thank the East Area Renovation team (project leader, Sebastien Evrard /EN-EA-AC) for maintaining essential CLOUD services (access, crane use, electricity, de-ionised water, chilled water, counting room, etc.) to allow the CLOUD14 run in fall 2019.

We understand that a CLOUD run in 2020 is excluded by the East Area Renovation work, and so the CLOUD15 run will be planned for fall 2021. There will be a large workload for the small CERN CLOUD team and the EP-DT gas group to prepare CLOUD for the CLOUD15 run in the re-configured T11 zone after LS2 (Fig. 9).

Acknowledgements

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