CLOUD'S 10-YEAR SCIENTIFIC PROGRAMME

CLOUD Collaboration

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1   INTRODUCTION

Overview: The Cosmics Leaving OUtdoor Droplets (CLOUD) experiment at CERN (Fig. 1) was conceived and constructed to examine the influence of ionisation from galactic cosmic rays (GCRs) on aerosols, clouds and climate [76, 78], as a potential mechanism that could explain extensive evidence for solar-climate variability in the palaeoclimate [14, 77]. The essential concept was to reproduce well-controlled and precisely-known atmospheric conditions inside a large chamber and to simulate cosmic rays with an ionising particle beam from the CERN Proton Synchrotron.

The purpose of this document is to take stock of what has been learnt during the first 10 years of CLOUD operation and to use this experience to foresee how the scientific programme may evolve over the next 10 years. Since starting data collection, CLOUD experiments have provided an unprecedented molecular understanding of aerosol particle formation and growth in the atmosphere (Fig. 2), and measured particle nucleation and growth rates over a wide range of atmospheric conditions. Following a series of high-impact publications, CLOUD is now widely recognised as the world's leading experiment for laboratory studies of atmospheric aerosol nucleation and growth [4, 113]. Indeed, CLOUD is the world's first—and, so far, unique—laboratory experiment to reach the demanding technical performance required to measure nucleation and growth of aerosol particles under controlled atmospheric conditions.

Fig. 1: CLOUD in the East Hall T11 beam line of the CERN Proton Synchrotron, during the CLOUD13 run, September-November 2018. The stainless-steel chamber (3.0 m diameter, 26 m^3 volume) is located inside a thermal housing around which air circulates at a temperature between 208 K and 313 K, with a stability near 0.01 K (the air return duct can be seen to the left of the thermal housing). The chamber is surrounded by an array of state-of-the-art instruments that continuously sample and analyse the contents of the chamber. Five different lights sources for the chamber mimic sunlight by targeting specific photochemical reactions. A 3.5 GeV/c π^+ beam of about 1.5 m transverse dimension provides controlled ionisation rates over the full range occurring in the troposphere. The beam emerges from the green dipole magnet at the right-hand edge of the image and passes through an $x - y$ counter hodoscope before crossing the chamber. Neutral (ion-free) conditions are simulated with a high-voltage clearing field, which sweeps ions from the chamber in under 1 s. The chamber is supplied with synthetic air from the evaporation of liquid nitrogen and liquid oxygen. The air is humidified with ultrapure water, and ozone is added from a UV generator. A flexible, ultra-clean and highly-stable gas system supplies the chamber with up to around 25 trace gases at concentrations as low as 1 pptv (part per trillion by volume) from concentrated sources that include gas bottles, evaporators and generators. Two stainless-steel fans at the top and bottom of the chamber, respectively, ensure the contents of the chamber are well-mixed. In order to maintain cleanliness inside the chamber, the fans are magnetically coupled to external drive shafts and motors. For 'CLOUDy' experiments, the chamber is operated in a classical Wilson cloud chamber mode by performing controlled adiabatic pressure reductions of a humid air parcel. This maintains a liquid or ice cloud in the chamber for 5-10 minutes, to allow investigation of direct ion-aerosol-cloud interactions and other cloud microphysical processes.
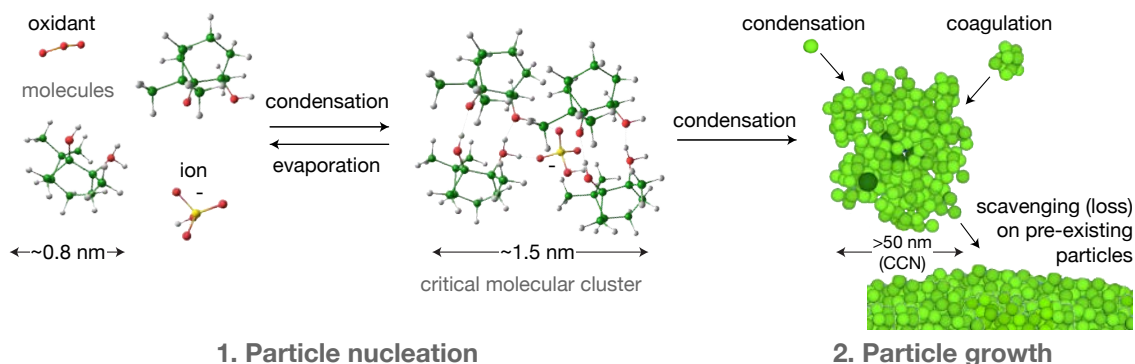
Fig. 2: Nucleation and growth of atmospheric aerosol particles. Oxidation processes in the atmosphere produce a few vapours with extremely low volatility (saturated vapour pressures below 10 pg m^{-3}) such as sulphuric acid or biogenic highly oxygenated molecules (HOMs). These vapours can form embryonic clusters of only a few molecules, but the clusters generally evaporate without the presence of a suitable stabilising agent such as ammonia or ions from galactic cosmic rays. Once the molecular clusters exceed a critical size of around 1.5 nm, they become more stable and continue to grow by further vapour condensation. The new aerosol particles may eventually grow above 50 nm, where they become cloud condensation nuclei (CCN) and can seed cloud droplets, provided they are not previously lost by collision with pre-existing particles.

Science: New particle formation and growth is important for climate change since it gives rise to more than half of global cloud condensation nuclei (CCN) [54]. It is also a major health issue since it produces particle pollution and smog episodes in urban environments. CCN and clouds have increased since pre-industrial times, which has offset a significant but poorly-understood fraction of the warming from increased greenhouse gases. A large part of this uncertainty results from the poorly-known baseline aerosol state of the pristine pre-industrial atmosphere [15]. Anthropogenic increases of aerosols and clouds are considered by the Intergovernmental Panel on Climate Change (IPCC) to be the major uncertainty limiting our understanding of Earth's climate sensitivity (the mean surface temperature change for a given radiative forcing), which, in turn, limits the ability of climate models to make precise climate projections for the 21st century [69].

CLOUD experiments have shown that ions from galactic cosmic rays can stabilise nucleating clusters, and, for otherwise weakly bound clusters (such as ammonia sulphuric acid, organics with sulphuric acid, and pure organics), that ions can enhance nucleation rates by a factor 10–100, depending on conditions [78, 123, 79, 83]. The sensitivity to ions is especially strong for pure biogenic nucleation, which may have dominated during pristine pre-industrial times, suggesting a higher sensitivity to solar variations in the past [79]. However, climate-model simulations using CLOUD measurements show very little sensitivity of current global CCN to variations of ionisation over a solar-cycle [37, 57, 54].

Does this close the door on a significant GCR-cloud-climate connection? No, for several reasons. Firstly, CLOUD is still in a discovery mode where important new mechanisms involving ions are being found, such as pure biogenic nucleation in the absence of sulphuric acid [79]. Secondly, aerosol particles must reach sizes near 50 nm before they constitute cloud condensation nuclei (CCN), and charge is likely to enhance particle growth rates as well as their formation rates. Thirdly, cosmic rays may modulate cloud properties independently of aerosol nucleation processes. Space charge and relatively highly-charged aerosol build up around clouds due to droplet evaporation and to ion drift in Earth's fair-weather electric field. This could alter droplet or ice formation processes and provide a mechanism for cosmic rays to modulate cloud properties directly [14]. CLOUD will study these processes experimentally during the 'CLOUDy' runs. Finally, although *global* CCN seem to be relatively insensitive to solar variations of cosmic ray ionisation, it cannot yet be excluded that there are climatically-significant *regional* effects. All these topics require further experimental and modelling studies by CLOUD before the cosmic ray-climate question can finally be settled.

CERN's role: CLOUD would not be possible without CERN. As well as the availability of a π^+ beam, the design, construction and unprecedented performance of the CLOUD facility and its infrastructure have depended on CERN technical know-how—and continue to do so. Both the CLOUD facility and the state-of-art analysing instruments that sample the contents of the chamber are continually adapted and developed for each run to match the experimental goals. The small CERN team is responsible for maintaining and developing the CLOUD facility and its infrastructure. In addition, members of the CERN team are responsible for overall coordination of the CLOUD experiment in their capacities as spokesperson (Kirkby), technical coordinator (Mathot), resources coordinator (Onnela), run coordinator (Manninen), and DAQ coordinator (Weber).

A less-obvious, but nevertheless important, role of CERN concerns 'cultural exchange'. CLOUD has introduced to atmospheric science an experimental approach developed over several decades in particle physics whereby institutes pool their resources to build and operate a large "general-purpose" detector that comprehensively measures the processes under study with combined instrumentation beyond what can be achieved any single institute. This approach has been highly successful in particle physics but was previously unfamiliar to atmospheric science. It has proved pivotal to CLOUD's success. So too has been the experience of the CERN team in organising and coordinating a successful international collaboration.

Another scientific culture introduced to climate science by CLOUD is to couple experiments and global modelling within a single project. In a break from the normal way of building global models— where they are separated from experiments, at the end of the research chain—the global model simulations are tightly integrated in CLOUD and are used to guide the experiments as well as to exploit the experimental results. The parameterised CLOUD measurements are embedded in the advanced aerosol chemistry and physics model, GLOMAP, developed by the University of Leeds CLOUD partner. This has led to the first global model study of aerosol production based solely on laboratory measurements [37]. GLOMAP is, in turn, incorporated into the UK Earth System Model (UKESM), so the CLOUD measurements are providing a firm experimental foundation for the IPCC modelling community to investigate the full coupling of aerosols, clouds, radiation and climate response.

Scientific objectives: CLOUD has three key overall scientific objectives during the next 10 years:

1. **Climate change:** Identify and quantify the mechanisms for aerosol particle formation and growth in the present-day atmosphere, and determine the aerosol state of the pristine pre-industrial climate. This will improve our understanding of anthropogenic aerosol radiative forcing and help sharpen estimates of projected climate change in the 21st century, as emissions of polluting vapours are reduced.

2. **Cosmic-ray-climate mechanism:** Answer whether cosmic rays provide a physical mechanism for solar-climate variability in the pristine pre-industrial climate and determine to what extent they may contribute to present-day climate change.

3. **Urban particle pollution:** Identify and quantify the mechanisms for smog formation and growth in polluted urban environments. This will provide an improved scientific understanding for informed public-health policy decisions on urban pollution reduction , as well as allow an assessment of the impact of 'urban plumes' on regional climate.

In the remainder of this document we will first outline the importance of atmospheric particle formation and CLOUD's contributions to date. Building on this experience, we will then present CLOUD's 10-year experimental and modelling programme that meets the overall objectives above. Finally we will summarise the resources required from CERN.

2 IMPORTANCE OF ATMOSPHERIC PARTICLE FORMATION

2.1 Climate change

The change in mean global surface temperature since pre-industrial time, ΔT, can be simply expressed as $\Delta T = \lambda F$. The effective radiative forcing, F, is the change of Earth's mean surface energy input, and the climate sensitivity, λ, is the equilibrium surface temperature response to the forcing. The parameter λ includes all feedback processes such as changes in atmospheric water vapour (greenhouse gas), ice cover (albedo) or rate of heat sequestering by the deep ocean. The feedback processes are hard to predict from first principles but λ can, in principle, be determined from the 'experiment' currently underway if the temperature change and forcing are known. Once λ is known, future climate warmings for given emissions scenarios can be reliably predicted. The uncertainty in ΔT (0.8°C) is small since it is directly measured by thermometers. The current understanding of F and its components are summarised in Fig. 3a, and the overall uncertainties are shown in Fig. 3b. Whereas the radiative forcing from greenhouse gases is well-understood, there are much large uncertainties from aerosol forcing.
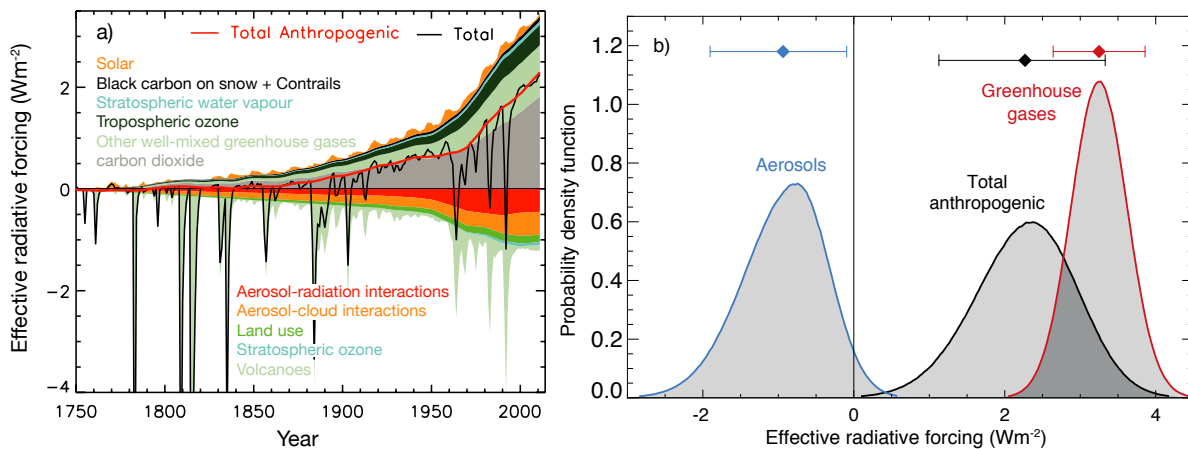

Fig. 3: The effective radiative forcing of climate since pre-industrial times. a) Evolution of effective radiative forcings of the climate since 1750 from natural and anthropogenic sources. b) Present-day global mean effective radiative forcings and uncertainties due to changes in greenhouse gases and aerosols since pre-industrial times. The central values and 5% to 95% confidence ranges are indicated by the points and error bars above the curves. The confidence ranges represent model diversity rather than strict statistical quantities. Positive forcings (warming effects) from greenhouse gases are reasonably well understood but negative forcings (cooling effects) from aerosols are not. The total anthropogenic forcing was 2.3 Wm^{-2} in 2011, and its uncertainty range of 1.1 to 3.3 Wm^{-2} is dominated by aerosols. Figs. 8.18 and 8.16 from IPCC 5th Assessment Report [69].

Aerosols exert a strong influence on Earth's radiative balance through a series of complex and poorly-quantified processes involving scattering and absorption of radiation (Figs. 4 a-b) and interactions with clouds (Figs. 4 c-d) . In consequence, one of the most challenging and persistent problems in atmospheric science has been to understand how changes in aerosol particles due to anthropogenic activities are affecting the climate. The IPCC considers that the major factor limiting our ability to make accurate projections of long-term climate change is due to the uncertainty in how much aerosols and clouds have changed between the pre-industrial era and the present, and how they may continue to change in the future (Fig. 3a) [69]. The magnitude of aerosol radiative forcing since 1750 is estimated to lie between -0.1 and -1.9 Wm^{-2}, compared with a much better understood forcing of 2.6 to 3.8 Wm^{-2} due to greenhouse gases (Fig. 3b). The resultant factor three uncertainty in total anthropogenic radiative forcing (between 1.1 and 3.3 Wm^{-2}) directly translates into a factor three uncertainty in Earth's climate sensitivity (1.5°C to 4.5°C for a doubling of CO_2). Aerosols therefore have a major influence on climate change, but their uncertainty has persisted through all IPCC assessments since 1996 despite a huge
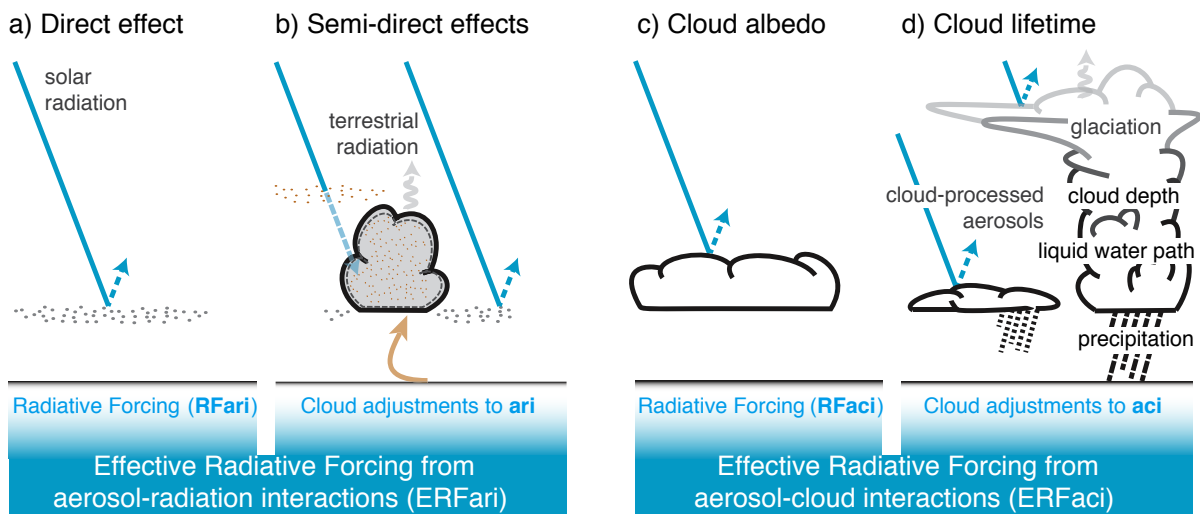
a) Direct effect   **b) Semi-direct effects**   **c) Cloud albedo**   **d) Cloud lifetime**

Radiative Forcing (**RFari**)   Cloud adjustments to **ari**   Radiative Forcing (**RFaci**)   Cloud adjustments to **aci**

Effective Radiative Forcing from aerosol-radiation interactions (ERFari)   Effective Radiative Forcing from aerosol-cloud interactions (ERFaci)

Fig. 4: Effective radiative forcings (ERF) from changes in atmospheric aerosol: a–b) aerosol-radiation interactions (ari) and c–d) aerosol-cloud interactions (aci). The ERF include rapid but poorly-understood 'cloud adjustments' in response to aerosol changes (b and d) . Adapted from Fig. 7.3 of the IPCC 5th Assessment Report [69].

research effort. Indeed, forty years ago, the seminal 1979 Charney report of the US National Academy of Sciences estimated the same value for Earth's climate sensitivity, namely $3°C$ for a doubling of CO_2, with an uncertainty between $1.5°C$ and $4.5°C$ [17].

2.2 Human health

Ambient fine particles with sizes below 2.5 μm (PM2.5) also have a major impact on human health. The mass concentration of PM2.5 has been extensively monitored since the 1970's, and the adverse health effects are well-established by numerous epidemiological studies. PM2.5 is now recognised as the fifth-highest risk factor of mortality worldwide, causing 4.2 million premature deaths per year [23]. Life expectancy is reduced by around 0.6 years per 10 μg m^{-3} mean exposure to PM2.5 [116]. A major source of smog episodes in polluted urban environments is due to new particle formation and growth involving soot and volatile organic compounds. They largely arise from incomplete combustion of fossil fuels, wood fires and natural gas heaters, and from the rapid cooling of hot exhaust gases from vehicles, which produce particles from low volatility precursor vapours such as unburnt engine lubrication oil.

Although ultrafine particles (those with diameter below 100 nm) contribute less than a few per cent of PM2.5 mass, they dominate particle *number* concentrations. Whereas PM2.5 are predominantly deposited in the upper respiratory tract, ultrafine particles penetrate deeper into the tracheobronchial and alveolar regions, where they are deposited with high efficiency. Once deposited, ultrafine particles—in contrast to larger particles–can readily translocate to extrapulmonary sites via the blood and lymph circulation, and even via axons. However, the particular health hazards of ultrafine particles remain uncertain due to the lack of long-term environmental monitoring for epidemiological studies.

3 CLOUD'S SCIENTIFIC CONTRIBUTIONS TO DATE

3.1 CLOUD experimental runs and publications

When CLOUD was designed there was enormous uncertainty surrounding atmospheric new particle formation. Even the predominant nucleation mechanism was unknown, although there was clear evidence that sulphuric acid was involved [81]. Theoretical considerations suggested that nucleation by sulphuric acid vapour alone together with water vapour (so-called 'binary' nucleation) was too slow to explain atmospheric observations. Numerous experimental measurements of the binary sulphuric acid nucleation

rate had been made but they differed by many orders of magnitude, with some claiming it could account for atmospheric observations, but others not. The role of additional vapours or ions in stabilising new particle formation was unknown.

In its first experimental run in 2009, CLOUD discovered a key base stabilisation mechanism in which each additional sulphuric acid molecule in a nucleating cluster is stabilised by one additional ammonia molecule. CLOUD also found that ions helped to stabilise the clusters even further, increasing the nucleation rates by up to a factor 10. These represented the world's first measurements of atmospheric nucleation at the molecular level. CLOUD also showed that binary sulphuric acid was around one million times too slow to account for nucleation in the warm boundary layer. These first results from CLOUD were published in Nature in 2011 [78] and have received more than 560 citations.

Table 1: A decade of CLOUD experimental runs at the CERN PS. 'NPF' indicates new particle formation i.e. nucleation and growth.

Run	Year	Month	Aim
CLOUD1	2009	Nov-Dec	Commissioning and binary sulphuric acid (H_2SO_4) NPF
CLOUD2	2010	Jun-Jul	Binary H_2SO_4 and ammonia (NH_3) ternary NPF
CLOUD3	2010	Oct-Nov	Binary H_2SO_4 and NH_3 ternary NPF
CLOUD4	2011	Jun-Jul	Dimethylamine (C_2H_7N) and pinanediol ($C_{10}H_{18}O_2$) ternary NPF
CLOUD5	2011	Oct-Dec	Free tropospheric (cold) binary H_2SO_4 and NH_3 ternary NPF
CLOUD6	2012	Jun-Jul	Commission cloud formation (CLOUDy) adiabatic expansion experiments
CLOUD7	2012	Sep-Dec	NH_3, C_2H_7N and α-pinene ($C_{10}H_{16}$) ternary NPF
CLOUD8	2013	Oct-Dec	Pure biogenic ($C_{10}H_{16}$) NPF and CLOUDy commissioning (no PS)
CLOUD9	2014	Sep-Nov	CLOUDy (aqueous phase, ice & glassy aerosol) and ion studies
CLOUD10T	2015	Apr-May	Instrument development (FIGAERO, DMA-TRAIN, TDDMA, acetate TOF)
CLOUD10	2015	Sep-Dec	Boreal forest NPF ($C_{10}H_{16}$, $\delta3$-carene, H_2SO_4, NH_3 and NO_x)
CLOUD11	2016	Sep-Nov	Pure biogenic NPF ($C_{10}H_{16}$, isoprene (C_5H_8), β-caryophyllene, NO_x), boreal NPF ($C_{10}H_{16}$, H_2SO_4, NH_3 and NO_x), urban aromatic NPF (toluene (C_7H_8), trimethylbenzene (C_9H_{12}), naphthalene ($C_{10}H_8$), H_2SO_4, NH_3 and NO_x)
CLOUD12	2017	Sep-Nov	Marine NPF (iodine (I_2) and diiodomethane (CH_2I_2)), precision H_2SO_4-NH_3 growth rates, multi-component NPF ($C_{10}H_{16}$, C_5H_8, H_2SO_4, NH_3 and NO_x), urban NPF (C_7H_8, C_9H_{12}, $C_{10}H_8$, H_2SO_4, NH_3 and NO_x)
CLOUD13T	2018	Jun-Jul	Ion production and loss rates; ion non-uniformities in the chamber
CLOUD13	2018	Sep-Nov	Marine NPF (I_2 and dimethylsulphide (C_2H_6S)), multi-component NPF ($C_{10}H_{16}$, C_5H_8, H_2SO_4, NH_3 and NO_x), nitric acid NPF (HNO_3 and NH_3), urban NPF (C_7H_8, cresol (C_7H_8O), H_2SO_4, NH_3, C_2H_7N and NO_x)

In a series of fifteen experimental runs (Table 1) since 2009, CLOUD has systematically explored the physics and chemistry of atmospheric particle formation and growth, and investigated the role of ions. At every step, CLOUD findings have solidified our fundamental scientific understanding of these processes while refining their representation in climate models. Yet, at every step, surprises have emerged upending existing assumptions about particle formation and growth and, in several cases, extending more broadly into atmospheric chemistry and our understanding of oxidation mechanisms.

Those thirteen experimental runs have led to sixty-four published papers, one in 2010 [39], three in 2011 [87, 78, 82], five in 2012 [88, 153, 118, 38, 11], six in 2013 [42, 136, 165, 2, 32, 73], six in 2014 [92, 134, 129, 12, 84, 123], six in 2015 [86, 166, 48, 119, 89, 135], twenty-seven in 2016 [91, 37, 57, 1, 47, 68, 63, 43, 83, 146, 79, 93, 67, 128, 138, 105, 70, 109, 75, 40, 101], nine in 2017

[27, 157, 54, 106, 21, 150, 13, 9, 144], and seven in 2018 [94, 143, 10, 155, 132, 85, 49]. In addition, four are under review [103, 131, 171, 62], eighteen are nearing submission [55, 99, 161, 117, 139, 169, 50, 152, 60, 26, 112, 160, 162, 174, 160, 133, 65, 126], a further twenty-four are in preparation [110, 90, 53, 98, 163, 74, 111, 44, 3, 66, 56, 145, 25, 104, 36, 45, 61, 170, 18, 137, 151, 46, 156, 158], and several dozen more are in early stages of writing.

The published work includes four papers in *Nature* [78, 2, 146, 79], two in *Science* [123, 37], four in *PNAS* [136, 84, 57, 143], one in *Nature Communications* [93], and one in *Science Advances* [94]. One is under review in *Science Advances* [171], one in *PNAS* [62] and a further two manuscripts are being prepared for submission to *Nature* [160, 169]. Overall, CLOUD papers have been cited more than 2500 times, and the h-index for the sixty-four published papers is 22.

3.2 CLOUD results on new particle formation and growth

3.2.1 *Sulphuric acid and ammonia*

Experiments started (CLOUD1–3 and CLOUD5) with the ternary sulphuric acid, ammonia and water system, with and without ions at various intensities. Measurements helped resolve a long-standing controversy about the magnitude and power-law dependence of sulphuric acid nucleation [168, 5, 176, 7, 8, 140]. Pure binary nucleation of sulphuric acid and water vapour is extremely slow at ambient concentrations. Even ternary nucleation rates remain lower than those observed in the warm planetary boundary layer for a given H_2SO_4 concentration, though charge substantially enhances nucleation rates at relevant ion-pair production rates [78, 135, 86]. The CLOUD dataset represents the first extensive mapping of ternary nucleation rates over the full range of tropospheric conditions.

Instrumentation and technical developments were central to these findings. Particle measurements at $d_{mob} = 1.2$ nm and associated theory showed that particle microphysics down to the smallest sizes can be a confounding factor when the lower limit cutoff sizes for particle measurement are relatively large ($d_{mob} \gtrsim 3$ nm) [92, 42, 165, 83]. Critical to quantifying nucleation and growth rates were gas-phase measurements of sulphuric acid [87], organic constituents [134], ammonia, [11] and most importantly, the composition of small growing clusters. Both ionisation in the CLOUD chamber [135] and nitrate ion chemical ionisation [83] have been used to unequivocally identify the controlling pathways for nucleation and growth.

The composition measurements revealed that very small particles grow as ammonium bisulphate, with an acid:base stoichiometry just above 1:1 rather than fully as neutralized ammonium sulphate with a 2:1 stoichiometry [78, 135, 83], once the gas-phase ammonia reaches roughly 10-100 times the gas-phase sulphuric acid concentration [11].

3.2.2 *Sulphuric acid and dimethylamine*

Subsequent to finding that the sulphuric acid–ammonia system could not explain boundary layer new particle formation at warm temperatures, experiments in CLOUD4 and CLOUD7 investigated a stronger base: dimethylamine. Nucleation and growth was found to proceed at the kinetic limit in the presence of only 5 pptv dimethylamine. This was subsequently confirmed with well-controlled experiments in flow tubes [71]. As a result, the CLOUD experiments revealed that nucleation and growth rates for the sulphuric-acid dimethylamine system overlap those observed in the atmosphere [2]. The particle growth rates are also fast because the rates are kinetic i.e. governed only by the collision rate of sulphuric acid vapour. A large fraction of the available sulphuric acid rapidly evolves into small clusters with 2-4 sulphuric acid molecules [84]. Coagulation of these small clusters with growing particles can drive up to 90% of the particle growth [93]. As with ammonia, the small clusters grow with a 1:1 stoichiometry once the amine vapour reaches a concentration greater than roughly 10 times the sulphuric acid vapour. Theory suggests that the transition to a 2:1 stoichiometry occurs for $5 \lesssim d_{mob} \lesssim 10$ nm, depending on the amine concentration [1].

Sulphur dioxide—the precursor of sulphuric acid—largely derives from coal combustion [51], while ammonia and amine emissions are largely agricultural in origin [115]. Concentrations of all these vapours have risen dramatically since the industrial revolution. However, although the nucleation rates of sulphuric acid–amines match those observed in the atmosphere, the composition of the growing clusters does not resemble the composition observed in the boreal forest [136]. On the other hand, there is recent evidence that amines may play a crucial role in new particle formation in polluted megacities like Shanghai [173, 172]. The CLOUD results were key to interpreting these urban observations.

3.2.3 Biogenic highly oxygenated organic molecules (HOMs)

Before CLOUD started to study organic vapours, there was considerable evidence that condensation of organic vapours often drives the growth of nucleated particles [124, 114, 35, 125], but whether they participated in nucleation itself was uncertain [177]. CLOUD experiments thus turned to the role of biogenic terpenes in new particle formation [52, 80, 59].

The first organic system studied in CLOUD was oxidation of pinanediol in the presence of sulphuric acid (CLOUD4). Pinanediol is a surrogate for first-generation oxidation products of α-pinene, with a vapour pressure similar to cis-pinonic acid. The experiments thus explored the hypothesis that multi-generational aging is a source of new particle formation [30, 29], and that the organic oxidation products may have a special affinity with sulphuric acid [32, 109]. The results strongly confirmed this hypothesis, establishing that OH oxidation of pinanediol results in rapid formation of highly oxidised products that cluster effectively with sulphuric acid, generating a multitude of organics that typically form clusters with oxidised-organic–acid ratios between 2:1 and 1:1 under atmospheric conditions [136]. This suggests that, rather than acid-base chemistry enforcing a reaction stoichiometry, the organic–inorganic pathway involves a broad valley in free-energy space associated with hydrogen bonding [109]. Global modelling with GLOMAP incorporating a parameterisation of this organic-inorganic pathway significantly improves agreement of the model with measurements in the boundary layer [123].

During this time, a picture was emerging in the literature describing rapid oxidation of organics ('auto-oxidation') to form first-generation highly oxygenated molecules (HOMs), which would have extremely low volatilities. This peroxy-radical auto-oxidation [24, 41, 127, 72] explains the high oxidation state of the pinanediol products [136]. However, auto-oxidation also means that first-generation oxidation of α-pinene and other organics can be a source of particles. CLOUD thus embarked on a series of experiments involving oxidation of α-pinene and other terpenes by both ozone and OH radicals, which continue to the present day since these processes turn out to be of great importance for atmospheric particle formation and growth. Thus far, CLOUD has explored new particle formation by α-pinene oxidation products alone, mixed with sulphuric acid (CLOUD8), mixed with sulphuric acid and ammonia (CLOUD10), and in richer mixtures of other terpenoids (CLOUD10 – CLOUD13).

The most notable finding was that the first-generation oxidation products of α-pinene alone could drive nucleation and growth at rates and under conditions observed in the atmosphere, in the absence of sulphuric acid (Fig. 5) [79]. The 'pure biogenic' nucleation rates depend strongly on charge, with boundary-layer ionisation rates enhancing nucleation by up to a factor of 100. Particle growth accelerates by a factor of 5 or so as particles grow from 1 to 3 nm, whereas condensation of non-volatile species would *decelerate* growth by a factor of 5, strongly indicating that the finite vapour pressure of condensing organics inhibits condensation to the smallest particles via the Kelvin effect [146, 21].

The importance of this new nucleation mechanism is that it implies the ubiquitous presence of new particle formation in the pristine pre-industrial atmosphere, despite sulphur dioxide levels that were around 5 times lower than today. In the pre-industrial era, broad regions with high terpene emissions imply higher CCN concentrations than previously thought (Fig. 6), raising the baseline aerosol state from which the current radiative forcing is determined. Climate model simulations with and without pure biogenic nucleation reduce the magnitude of the estimated aerosol radiative forcing by 27% [57].
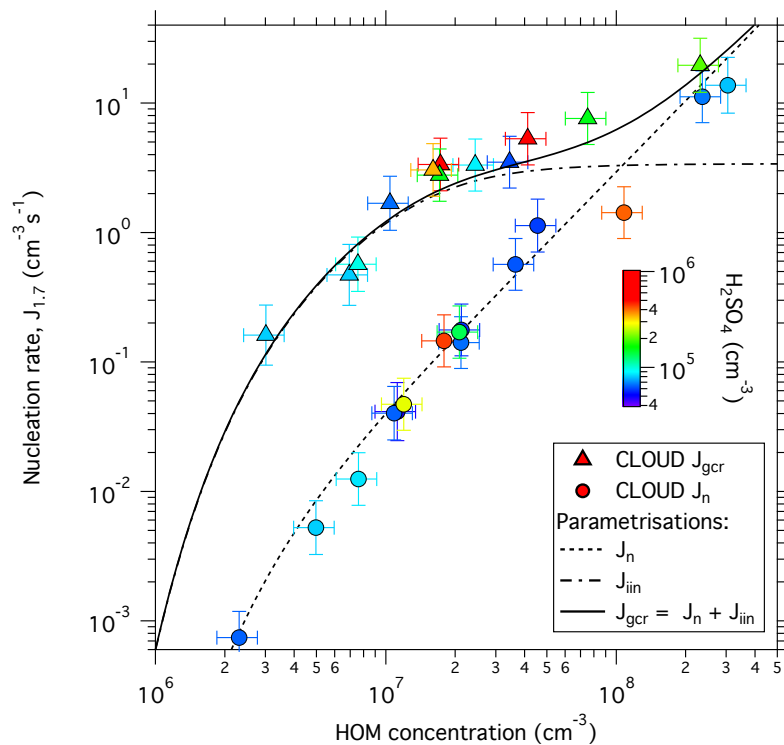
Fig. 5: The discovery of pure biogenic nucleation [79]. Pure biogenic nucleation rates versus highly oxygenated organic molecule (HOM) concentration (1 pptv = 2.4×10^7 cm^{-3}). Neutral (J_n; circles) and galactic cosmic ray (J_{gcr}; triangles) nucleation rates are shown. The colour scale shows [H_2SO_4]; purple and blue points correspond to contaminant-level (below the detection threshold); other colours correspond to SO_2 added to the chamber. The fitted curves show parameterisations for J_n (dashed), J_{gcr} (solid) and ion-induced nucleation ($J_{iin} = J_{gcr} - J_n$; dot-dashed). The experimental conditions are 10–1300 pptv α-pinene, 30–35 ppbv O_3, 38% RH, 278 K, and $< 8 \times 10^5$ cm^{-3} H_2SO_4.
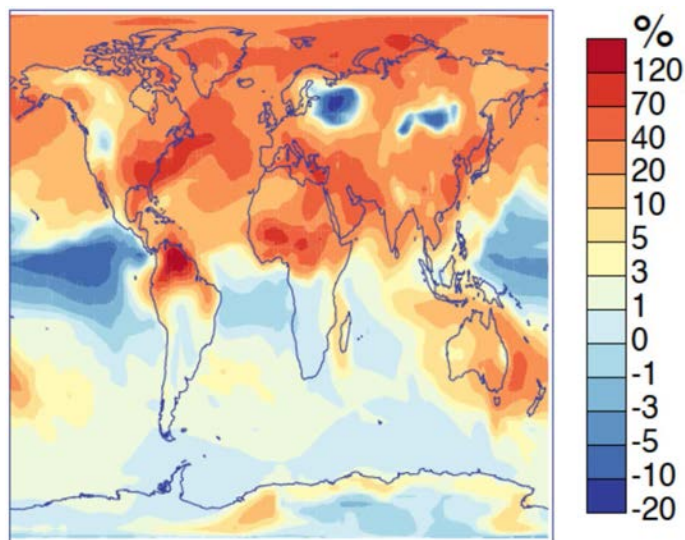

Fig. 6: Percentage change in cloud condensation nuclei (CCN) at cloud base level, at 0.2% supersaturation, for pre-industrial conditions in August, after including ion-induced pure biogenic nucleation in the model [57].

The pure α-pinene oxidation experiments revealed that nucleation itself is driven by covalently bound, extremely low-volatility organic compounds, C_{20} 'dimers' (ELVOCs), formed in the gas phase [79, 49] via association of two C_{10} peroxy radicals. The subsequent growth is largely controlled by highly oxidised, low-volatility organic compounds, C_{10} monomers (LVOCs), formed via peroxy-radical auto-oxidation [146]. Initial modelling of CLOUD growth rates for α-pinene + ozone biogenic nucleation suggested that nitrate chemical ionisation mass spectrometers (nitrate CIMS) observe some, but not all, LVOCs [146, 21]. This has been confirmed by subsequent nitrate CIMS and proton transfer reaction mass spectrometer (PTR-3 [13]) measurements [143, 46].

Findings from the most recent CLOUD experimental runs have confirmed that the production of ELVOCs and LVOCs is strongly temperature dependent [49, 143, 46, 174]. The yields are highly sensitive to NO_x, with ELVOC yields decreasing at increased NO concentration, presumably because RO_2 + NO termination impedes RO_2 + RO_2 ELVOC formation [171, 103, 133]. For the first-generation α-pinene + ozone system, we now have complete closure (agreement) between gas-phase observations (combining nitrate CIMS and PTR-3), measured and modelled particle growth rates between 1 and 30 nm, and the particle-phase composition measured with the I^- FIGAERO CIMS [143, 174]. The latter instrument is measuring the chemical composition of particles at ultra low concentrations below 1 ng m^{-3}.

Simultaneous oxidation of isoprene can inhibit nucleation, again presumably because the isoprene-derived RO_2 inhibit ELVOC formation, either by leading to more volatile (C_{15}) dimers or to separate molecular (C_{10} and C_5) products [62]. This can account for the near-absence of new particle formation in the biogenically-rich Amazonian boundary layer. The nucleation rates are sensitive to ultra-violet illumination near 360 nm [65, 139] similar to prior observations of secondary organic aerosol (SOA) formation [120, 175, 64].

3.2.4 Anthropogenic highly oxygenated organic molecules (HOMs) and nitric acid

In recent campaigns, CLOUD has found that anthropogenic aromatic vapours associated with urban pollution also produce HOMs that participate in particle nucleation and growth, but generally with a somewhat lower yield of the ELVOC component that drives nucleation and initial particle growth [169]. CLOUD has also investigated nucleation involving sulphuric acid, dimethylamine and anthropogenic organics in a highly polluted urban environment that simulates conditions in Shanghai [173].

The presence of high NO_x levels in urban environments leads to high mixing ratios of nitric acid, in the few ppbv range. CLOUD has studied the effect of these high nitric acid concentrations on particle nucleation and growth, and a manuscript on the results will soon be submitted to Nature [160].

The biogenic and anthropogenic organic particles have different attributes when particles collected on a teflon filter are thermally desorbed into an I^- FIGAERO CIMS [96]. Biogenic particles tend to thermally decompose, with C_{10} fragments appearing at desorption temperatures consistent with C_{20} products. C_{20} products also appear with lower mass fractions than the C_{10} fragments [174]. Products from an urban mix of anthropogenic aromatics are much more thermally stable [162]. This may indicate a difference in the fraction of organic particle growth that occurs via condensation of low vapour-pressure species vs reactive uptake of more volatile species for the different precursors [21, 162].

3.2.5 Coastal marine nucleation: iodine oxides and dimethylsulphide

Iodine oxides in biologically-active coastal regions are found to be an important source of new particles [108, 141]. CLOUD has investigated iodine-related coastal marine nucleation during the CLOUD12-13 runs. Whereas the importance of iodic acid (HIO_3) has already been shown in atmospheric observations [141], the nucleation rates, growth rates and chemical pathways remain unknown. The CLOUD experiments have elucidated these aspects and the results will be submitted soon to Nature [61]. During CLOUD13 the marine studies were extended to include dimethylsulphide (C_2H_6S), which is emitted by phytoplankton and, together with sporadic volcanoes, is the most important natural source of sul-

phur dioxide, amounting to about 20% of anthropogenic emissions. It is important to understand the role of dimethylsulphide in new particle formation in order to determine the aerosol state of the pristine pre-industrial climate.

The iodine data have also provided a precision measurement of the enhanced collision rate between a neutral condensing monomer and a charged cluster, due to the charge-dipole attractive force [60]. This is the first measurement of the charge-neutral collision rate to be made under atmospheric conditions, and it represents a fundamental physical property of ion-induced nucleation that needs to be accounted for in global aerosol simulations.
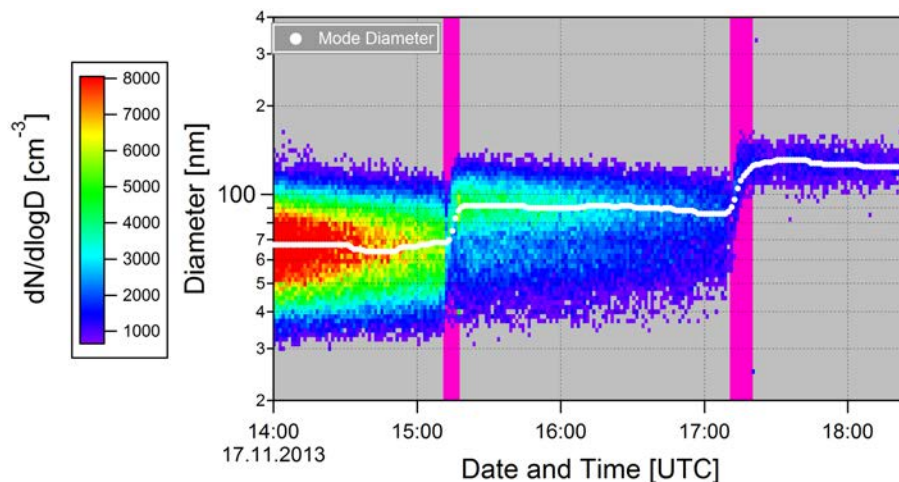

Fig. 7: Processing of SO_2 into H_2SO_4 in cloud droplets [67]. The data show the evolution of the dry size distribution of cloud condensation nuclei (CCN) over a period of around four hours. The vertical axis indicates the CCN diameter, with a colour scale indicating number concentration (cm^{-3}). The white line shows the mode of the CCN size distribution. During this period there were two adiabatic pressure reductions in the CLOUD chamber, which activated liquid clouds over periods of about 5 minutes (indicated by purple bands). The chamber was re-pressured after the first expansion to prepare for the second cloud activation. Rapid aerosol growth can be seen during both clouds: from 70 to 90 nm in the first cloud and 85 to 110 nm in the second. Aqueous-phase processing in cloud droplets is a major mechanism for aerosol growth but very few laboratory data exist so far.

3.3 CLOUDy experimental results

The chamber also has the capacity to employ adiabatic expansions in order to study water and ice nucleation, and subsequent particle properties and aqueous-phase chemistry (an example is shown in Fig. 7 [67]). For the expansions, the chamber is first brought to a high relative humidity at 220 mbar relative pressure, and then cooled adiabatically with a controlled pressure drop to 5 mbar [105]. Through *in situ* optical depolarization measurements, experiments on α-pinene secondary organic aerosol have constrained the glass transition points versus relative humidity, which rises from 35% RH at 263 K to 80% RH at 235 K [70]. These same glassy organic particles were found to nucleate ice for saturation ratios (with respect to ice) between 1.3 and 1.4 at 235 K, significantly below the homogeneous freezing value [68]. Ozonolysis of isoprene also forms secondary organic aerosol in aqueous droplets. Furthermore, the expansions were able to separate transient increases in condensed-phase organics during supersaturated (liquid) conditions from irreversible condensation of new organic mass [50].

Initial ion studies were carried out during the CLOUD8–9 CLOUDy runs but the instrumentation was insufficient to properly address the question of a possible direct effect of ions on cloud microphysics, independent of their effect on aerosols [14]. This will be addressed in the upcoming CLOUD14 run, Sep-Nov 2019, which will include more advanced instruments for measuring ions and ice particles, as well as a generator for highly-charged cloud condensation nuclei.

3.4 Modelling of CLOUD findings

A key objective of CLOUD research is to transfer the laboratory measurements into global climate models in order to simulate aerosol-climate interactions in both the present-day and pre-industrial atmospheres based CLOUD measurements rather than empirical parameterisations. Model simulations emphasise comparison with ambient observations and other constraints; they also are tested to ensure that when models and measurements agree they do so for the right reasons. The complexity and many interconnected scales from the intricate chemistry and microphysics in CLOUD to global circulation and climate also require parameterisations to be computationally tractable. This motivates model developments ranging in scale from chamber process (box) models to atmospheric large-eddy (cloud-resolving) models to high-resolution regional models to the general circulation models (GCMs) used for weather and climate forecasts.
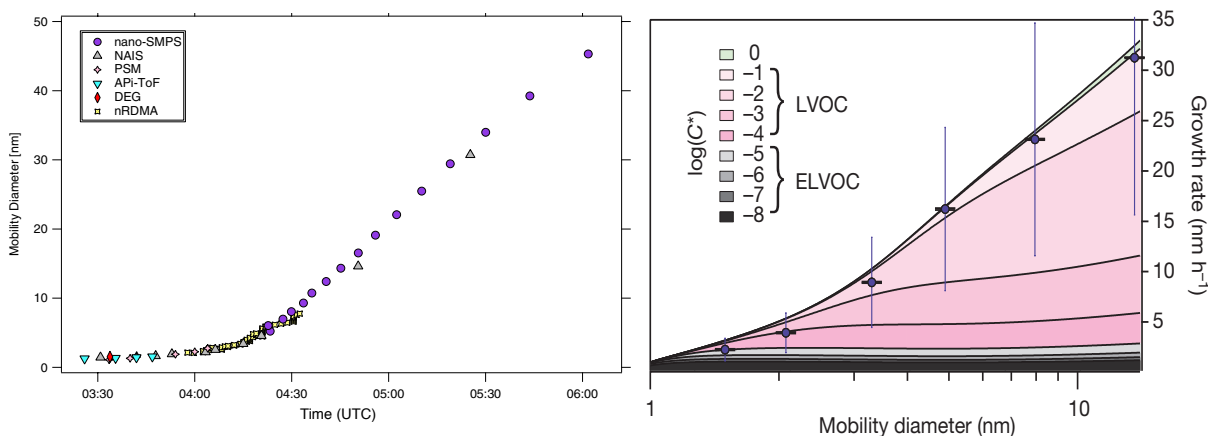

Fig. 8: Particle diameter versus time and modelled growth rate vs diameter from a nucleation experiment involving pure α-pinene oxidation [146]. Size is constrained by multiple instruments as indicated in the legend. Dynamical volatility basis set modelling with product volatility distribution constrained by mass spectrometer observations reproduces the growth rates as shown.

3.4.1 Dynamical and process models

A critical element of CLOUD data analysis is dynamical modelling. This is especially complicated for experiments involving condensation of organic vapours. There are hundreds of individual compounds with a wide range of vapour pressures in these experiments. The dynamical version of the CMU Volatility Basis Set (VBS) [34, 28, 31, 20] accounts for the time-varying excess gas-phase concentration of condensible species and the effect of curvature (Kelvin effect) on the activity of organics in very small particles [35, 149, 147, 148, 146, 21, 143]. The Kelvin term can be expressed as a simple power law, $10^{d_{K10}/d_p}$, where the 'decadel Kelvin diameter' (d_{K10}) is the particle diameter at which the vapour pressure of a species is a factor of ten higher than that over a flat surface. For a surface tension typical of organics, i.e. 30 mN/m, $d_{K10} \simeq 4.5$ nm [35]; thus organics in particles with $d_p = 2.25$ nm will have vapour pressures enhanced by a factor of 100. The dynamical VBS successfully predicts the observed size dependence of growth rates given the volatility distribution of observed gas-phase oxidation products [146, 21]. For a given vapour saturation ratio and particle-phase activity (which, together, give the condensation driving force) the dynamical VBS then calculates the growth rate due to condensation of organics over a wide volatility range. Broadly, the extremely low volatility organics (EVLOCs), with volatilities $C^* \leq 10^{-5}\,\mu\text{g m}^{-3}$, condense almost irreversibly, whereas the more abundant, but more volatile, low-volatility organics (LVOCs) start condensing significantly only when $d_p \gtrsim d_{K10}$ (depending on the gas-phase saturation ratio) [146, 21]. This explains the acceleration of the growth rate with

particle size that has been observed in CLOUD, as shown in Figure 8 over the full temperature range $250 < T < 300$ K as both chemistry and vapour pressures change dramatically [143, 174, 133].

3.4.2 Global aerosol-climate models

Global aerosol simulations using the University of Leeds GLOMAP model have been an essential element of CLOUD's research [123, 37, 57, 54]. Uncertainty in aerosol climate forcing has been a recalcitrant problem for climate change despite a huge research effort over the last 40 years. This is because the multi-scale coupled dynamics in the climate system is a complex problem but most importantly because a fundamental basis has been lacking to constrain the aerosol–climate forcing based on physics and chemistry rather than educated guesses. CLOUD/GLOMAP model results are beginning to impact this problem (e.g. Fig. 9 [57]).
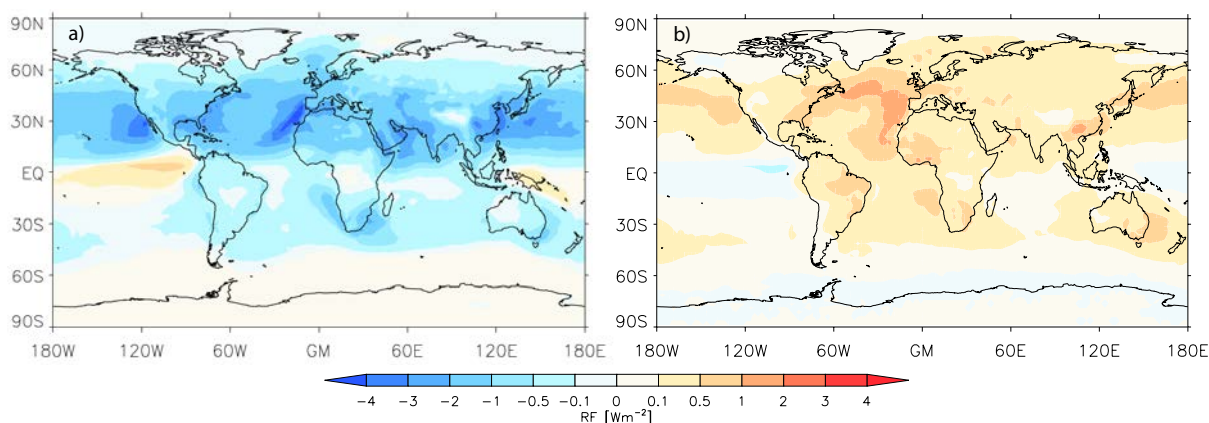

Fig. 9: GLOMAP simulations [57] based on CLOUD measurements showing a) the distribution of cloud-albedo radiative forcing (RF) between the pre-industrial climate and the present day, after including pure biogenic nucleation (left panel) and b) the change to this distribution when pure biogenic nucleation is included in the model (right panel). The global average radiative forcing is -0.60 W m^{-2}, and the change due to pure-biogenic nucleation is $+0.22$ W m^{-2} (a 27% reduction in magnitude).
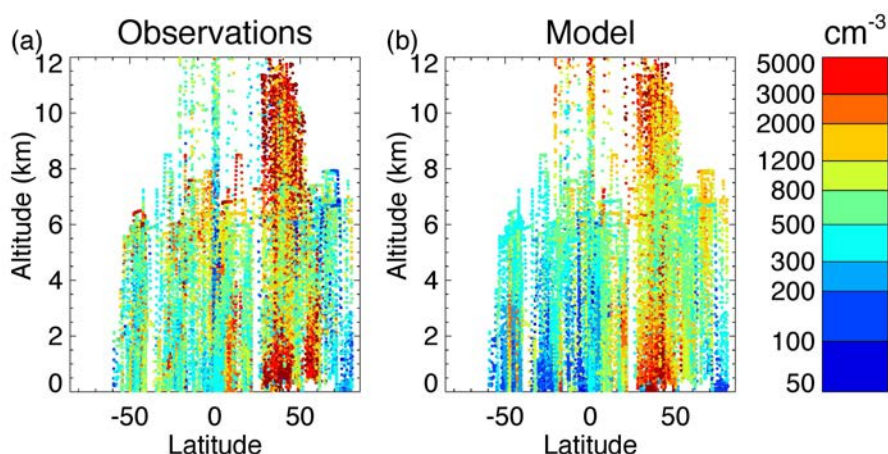

Fig. 10: Comparison of CLOUD measurements with global observations of new particle formation [37]. Comparison of observed (left) and modelled (right) particle concentrations (>3 nm diameter) versus latitude and altitude. This was the first time that a global aerosol model had been based solely on experimentally-measured particle nucleation and grow rates.

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Results from GLOMAP [97] using a parameterisation directly interpolating the CLOUD1–5 data show that ternary nucleation, usually involving ions, is ubiquitous throughout the free troposphere (Fig. 10 [37]). The CLOUD data show—for the first time—the importance of ammonia, together with sulphuric acid, for global particle formation. However, no significant solar influence via ion-induced nucleation was found on cloud microphysics [37]. These global modelling studies have now been extended to include pure biogenic nucleation [57]. They are currently being further extended using the extensive new results from CLOUD10–13 on multicomponent new particle formation involving sulphuric acid, ammonia, biogenic vapours (isoprene and α-pinene), water and NO_x [62, 171, 25, 56].

4   CLOUD'S 10-YEAR SCIENTIFIC PROGRAMME

CLOUD is firmly established as the world-leading experiment addressing atmospheric new particle formation. It defines the state of the art. We now know 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 organic molecules, HOMs), 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. These few statements would not have been possible before CLOUD started 10 years ago; at that time sulphuric acid alone was thought to be responsible for atmospheric particle formation.

Results from the first 15 experimental runs have greatly enhanced our understanding of the physics and chemistry of atmospheric aerosol and are providing a firm experimental foundation for rigorous climate simulations. 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 volatilities—and, in addition, the volatilities of *all* vapours is strongly temperature-dependent. This illustrates the breadth and complexity of the parameter phase space that CLOUD is investigating.

4.1   Experimental objectives

Several key areas have emerged at the limits of our knowledge, largely as a consequence of CLOUD research. The overall scientific objectives for CLOUD over the next 10 years are summarised in §1, and specific experimental objectives that we can identify today include the following (see §4.3 for the atmospheric modelling objectives):

1. Further experiments and modelling to develop a comprehensive treatment of highly oxygenated organic molecules (HOMs) participating in atmospheric particle formation and growth. This will include elucidating and parameterising the coupling between different HOM sources (isoprene, monoterpenes and sesquiterpenes) and oxidation mechanisms (ozone, hydroxyl radicals and nitrate radicals), largely via peroxy-radical (RO_2) cross reactions to form extremely low volatility peroxide 'dimers' (ROOR) or (with NO) less-volatile organonitrates.

2. The roles of humidity, temperature and ions throughout the phase space of reactions critical to atmospheric new particle formation.

3. The role of charge in cloud microphysics, including aerosol-droplet scavenging, nucleation of liquid water droplets and ice particle formation.

4. Marine new particle formation over biologically-productive regions of the open ocean—especially in regions of marine stratocumulus decks, which are important for Earth's radiative balance—including the roles of dimethylsulphide, methylsulphonic acid, iodic and other halogen-containing compounds, and amines.

5. Reactive uptake of highly oxidised light organics such as glyoxal to newly-formed particles.

6. The mechanisms for smog formation in highly polluted urban environments, including the effects of inorganic acids (sulphuric and nitric acid), bases (ammonia and amines) and aromatic organic oxidation products from vehicles, domestic fuels and domestic heating. Studies of their coupling with biogenic organics (from parks or in mixed rural/urban environments).

7. The effect on sulphuric acid particle nucleation of different amine species, e.g. methylamine, dimethylamine, trimethylamine or diamines.

It should be noted that CLOUD continues to make discoveries and so the experimental objectives are likely to be adapted according to new findings. These objectives motivate the 10-year run plan shown in Table 2.

Table 2: 10-year experimental run plan for CLOUD. The anticipated primary aims of each run are indicated. In general, each run will include shorter periods dedicated to other experimental topics from the list in order to make progress in all areas on roughly the same time scale.

Run	Year	Month	Primary aim
CLOUD14	2019	Nov-Dec	CLOUDy: CCN formed in CLOUD, charge effects on cloud microphysics
	2020		East Area renovations (no CLOUD run)
CLOUD15	2021	Sep-Nov	Nitric acid NPF in different environments (polluted to pristine)
CLOUD16	2022	Sep-Nov	Biogenic NPF: peroxy-radical (RO$_2$) reactivity & RO$_2$ + HO$_2$ competition
CLOUD17	2023	Sep-Nov	Urban NPF with ammonia, amines and high condensation sink
CLOUD18	2024	Sep-Nov	CLOUDy: reactive uptake, aqueous processing, charge & cloud microphysics
CLOUD19	2025	Sep-Nov	Marine NPF over the open ocean (dimethylsulphide, methylsulphonic acid)
CLOUD20	2026	Sep-Nov	NPF versus relative humidity, temperature, ions phase space for specific environments (Amazon, boreal, free troposphere, polar, marine stratocumulus)
CLOUD21	2027	Sep-Nov	Biogenic NPF: sesquiterpenes, multi-generation oxidation (ageing)
CLOUD22	2028	Sep-Nov	Sulphuric acid-amine NPF versus amine species, diamines
CLOUD23	2029	Sep-Nov	CLOUDy: charge effects on ice nucleation

4.2 Discussion

We will present in this section a discussion of some of the detailed scientific aspects for the proposed experimental run plan.

4.2.1 Organic oxidation

A key factor in past and future CLOUD experiments is the role of organic oxidation chemistry in new particle formation and growth. Simply put, our objective is to identify the rate-limiting step or steps in both particle formation (J) and CCN formation (J, growth rates, and survival probability) under a comprehensive range of pre-industrial and present-day conditions. Major challenges are that very small yields can matter—so that key steps can be quite minor pathways—and that experiments must be performed under atmospheric conditions. Fortunately, CLOUD is up to this task.

We care about nucleation rates $J \leq 1$ cm^{-3} s^{-1} and growth rates GR ≤ 10 nm/h. For a gas-kinetic process with a rate constant of 10^{-10} cm^3 molec^{-1} s^{-1}, two compounds with concentrations of 10^5 cm^{-3} will collide at a rate of 1 cm^{-3} s^{-1}. For them to nucleate, in very round numbers they require a saturation ratio $S \geq 10^4$ or so unless the nucleation involves a chemical reaction such as acid-base dissociation rather than hydrogen bonding and Van der Waals interactions (i.e., vapour-pressure

driven clustering). This means an effective saturation number concentration of $10 \, \text{cm}^{-3}$, which is a mass saturation concentration of $6 \times 10^{-9} \, \mu\text{g m}^{-3}$ for a molar weight of $350 \, \text{g mole}^{-1}$. This is why we regard ELVOCs with $C^\circ \lesssim 10^{-8} \, \mu\text{g m}^{-3}$ as potential nucleating species, provided their vapour mass concentration (in total) is $C^v \gtrsim 10^{-4} \, \mu\text{g m}^{-3}$. vapour concentrations of $0.1 \, \text{ng m}^{-3}$ matter to nucleation.

Growth rates scale differently. The growth rate for a particle size, $d_p \simeq 10 \, \text{nm}$, is $\simeq 226 \, \text{nm hr}^{-1}$ / $(\mu\text{g m}^{-1})$ [35], and so $2 \, \text{nm hr}^{-1}$ of growth requires $10^{-2} \, \mu\text{g m}^{-1}$ of condensable vapours with excess saturation ratios $\gtrsim 1$. Growth is, thus, potentially decoupled from nucleation, and both can be small contributors to overall secondary organic aerosol production. vapour concentrations as low as $10 \, \text{ng m}^{-3}$ matter to growth.

A typical background condensation sink is of order $10 \, \text{hr}^{-1}$ [164, 33], and so $10 \, \text{ng m}^{-3}$ of vapours driving nano-particle growth will produce $100 \, \text{ng m}^{-3} \, \text{hr}^{-1}$ of total secondary organic aerosol (SOA); a full day of photochemistry would produce roughly $1 \, \mu\text{g m}^{-3}$ of SOA. This is about half of a typical SOA formation rate at the boreal forest site Hyytiälä, Finland, which consequently explains the conclusion that half of the SOA formation needs to be effectively non-volatile to reproduce observed particle growth rates [114, 124, 125]. Finally, SOA molar yields from common precursors (monoterpenes, alkyl benzenes, etc.) are typically of order 0.2 [80, 59]. In summary, SOA yields from oxidation reactions are often 0.2, yields of species driving nano-particle growth are of order 0.1, and yields of species driving nucleation are of order 0.001. One tenth of one percent is a small tail on a large dog; we thus expect nucleation to be very sensitive to conditions, and experimental constraints to be correspondingly delicate.

The rate-limiting processes for these various products are likely to differ. Nucleation itself (when driven by vapour pressure alone and not by a chemical reaction) is controlled by saturation ratios, and so depends sensitively on the steady-state concentration of potential nucleating species; increasing the oxidation rate in an experiment by a factor of ten will increase saturation ratios by a factor of ten, which in turn will activate species that are a factor of ten more volatile, all else being equal. It is thus critical that experiments reproduce ambient conditions, or at least scale with ambient conditions (the balance of production rates and condensation sinks). Even with this requirement, given molar yields of 0.1% or so, extremely minor processes may have a major effect on production of nucleator species. The yields of species contributing to growth are larger and, thus, somewhat less likely to be extremely sensitive to conditions, potentially decoupling the production of nucleating from that of condensing species driving growth. Furthermore, the volatility distribution of condensing LVOC species governs their contribution through the critical 1-10 nm size range, where growth rates and survival probabilities change dramatically with size [146, 21]. The above discussion serves to show the necessity for nucleation and growth experiments in the laboratory to reproduce atmospheric conditions. Here, CLOUD is so far in a class by itself.

4.2.2 Urban nucleation

New particle formation is frequently observed in urban areas as well as in the remote atmosphere. Nucleation itself is not hard to explain. In spite of the high condensation sinks in polluted settings, production rates of condensible vapours are also high, so the steady-state saturation ratios of nucleating vapours can be similar in remote and urban locations [142, 58]. However, if the saturation ratios of condensible vapours and, thus, growth rates were also similar, the survival probabilities of those nucleated particles should be extremely small under urban conditions because of the large coagulation sink. Resolution of this paradox is a key objective for CLOUD, including progressively higher condensation sinks combined with addition of HONO photolysis as a hydroxyl (OH) radical source that will allow us to reduce relative humidity in the study of anthropogenic volatile organics nucleation and growth without weakening the OH production (from $O^1D + H_2O$).

4.2.3 Marine nucleation

New particle formation in the marine atmosphere has been a topic of interest since before the CLAW hypothesis [16]. Progress over the past 20 years suggests the oxidation of dimethylsulphide (DMS) [100, 6] contributes to the formation of new particles primarily in the free troposphere, at the edges of deep convective clouds, but a comprehensive assessment of the molecular picture of growth to CCN sizes is missing. The response of clouds to changes in aerosol is also much more complex than the boundary layer DMS chemistry originally envisioned in CLAW [121], with contributions from marine organics [102, 107, 122] and iodine in coastal environments [108, 141]. Understanding the sources of new particles and contributions to CCN budgets in pristine environments (a proxy for pre-industrial conditions) remains one of the most important requirements for accurate assessments of aerosol climate forcing [15].

New particle formation in the pristine marine and tropical free troposphere will be an increasingly important focus at CLOUD in the coming years. Recent field evidence suggests that iodic acid is the nucleating species that drives new particle formation and growth in iodine-rich, coastal atmospheric environments [141]. However, the mechanism for iodic acid formation remains elusive, and there are currently no data to constrain the temperature dependence of new particle formation and growth rates from iodic acid. Future CLOUD experimental runs will further investigate the temperature dependencies of the mechanism of DMS oxidation (ratio of sulphuric acid and methane sulphonic acid production), and of fatty acid photolysis as a source of HOMs that can grow nanoparticles [19, 130, 159] and appear to be ubiquitous over oceans and in the tropical free troposphere [22, 95, 154, 167]. Forthcoming CLOUD experimental runs will target missing fundamental knowledge of the reaction mechanisms, and the associated new particle formation and growth rates.

4.2.4 CLOUDy

Previous CLOUDy experiments have monitored the phase of secondary organic aerosol, finding that the transition from a liquid to glassy state alters its behaviour as CCN and ice nucleating particles (INP) [70, 68]. The CLOUDy experiments will investigate the ice and cloud droplet nucleation properties of freshly-nucleated and grown, chemically-aged, and cloud-processed organic and inorganic aerosol particles, with particular emphasis on the role of viscosity transitions (glassy particles). The latter result from changing chemical composition due to temperature, relative humidity, both gas- and particle-phase chemical reactions, and particle charge. Experiments performed during expansion-chamber runs will be augmented by measurements of CCN and INP behaviour of the particles formed during the isobaric CLOUD experiments, enabling parameterisations that are needed for atmospheric models.

4.3 Atmospheric modelling objectives

CLOUD's objectives for atmospheric modelling over the next 10 years are:

1. **Climate modelling in which the changes in aerosols influence cloud cover and precipitation.** So far in CLOUD the simulated changes in aerosols have been coupled to albedo (Fig. 4c) but not to the cloud physics. However, it is known that changes in aerosol can affect precipitation and cloud cover. We expect these so-called 'cloud adjustments' (Fig. 4d) to be particularly important in the pristine pre-industrial environment, so understanding them will help to determine more realistically the aerosol-cloud state of the pre-industrial climate and how it can be affected by changes in cosmic rays.

2. **Cloud-scale modelling at very high spatial resolution** to understand the behaviour of aerosols and ions around clouds. So far all the atmospheric modelling of CLOUD results has been at the global scale with model grid spacings of about 100 km. However, we know from field measurements that aerosols and ions can behave very differently in the vicinity of clouds where humidity is high, trace

vapours can be evaporated out of droplets, and elevated charge densities can occur. We also know that aerosol concentrations can become extremely low (1 cm^{-3} compared to typically 100 cm^{-3}) following precipitation, which may create ideal conditions for new particle formation. We will develop a new range of atmospheric aerosol and chemistry models capable of simulating these processes at the scale of 10's of metres (so-called large-eddy models) as well as regional models similar to those used in the most high-resolution weather forecasts. Studies will be performed over regions where new particle formation could make a big difference to the development of clouds, such as the Arctic, tropical and continental deep convection, and shallow-cloud marine environments with extremely low aerosol concentrations.

3. Near-cloud space charge and the effect on cloud droplet formation. Ion production and loss processes will be incorporated in the new cloud-resolving models to understand how the build up of near-cloud unipolar space charge affects the charge state of aerosols that are mixed into the clouds. It has been hypothesised that highly-charged aerosol particles could alter droplet formation processes and provide a means for cosmic rays to modulate cloud properties independently of aerosol nucleation processes. CLOUD will study these processes experimentally during the 'CLOUDy' runs.

4. Aerosol formation in highly polluted environments. New discoveries in CLOUD show that a range of anthropogenic vapour pollutants could be involved in new particle formation and growth. This could have important implications for the future evolution of aerosols and clouds as pollutant emissions are reduced. Emissions are already declining rapidly in China. We will simulate new particle formation and growth on the scales of 'urban plumes' using our new high-resolution models. We aim to understand the potential for non-linear changes in cloud condensation nuclei caused by rapid air pollution reduction policies. One hypothesis is that cloud condensation nuclei concentrations could be buffered because the reduction in primary particle emissions could be offset by increases in nucleation. This is advantageous since it would maintain the present negative aerosol-cloud radiative forcing (cooling). The precise mechanisms and rates for new particle formation and growth will need to be elucidated in the CLOUD chamber so that these finely-balanced processes can be simulated accurately.

5 CLOUD RESOURCES

5.1 CLOUD organisation

The collaboration responsibilities on CLOUD are essentially that the CERN team is responsible for the CLOUD facility and experimental coordination, and the external institutes are responsible for the analysing instruments attached to CLOUD for each experimental run and for CLOUD modelling. There is joint responsibility of all collaboration members for designing the experiments, operating CLOUD, analysing the data, carrying out the modelling studies and preparing the results for presentation and publication.

CLOUD runs at the CERN PS have evolved into one per year, held between September and November/December, as providing the most efficient net science output from CLOUD in consideration of the necessary time for offline data analysis, preparation of manuscripts and technical developments of the CLOUD facility and the analysing instruments. The full CLOUD team—in particular the doctoral students and postdocs—attend each CLOUD experimental run, with a typical average of 25–30 external researchers at CERN over the 8–10 week run. This is critical to the experimental run, since the experiments run continuously and the facility requires constant staffing by at least two operators. The researchers from the member institutions are not only responsible for the maintenance and operation of their own instruments, but must also understand all of the instruments as well as the scientific objectives for that shift's run plan in order to ensure success. The run plan for each day is developed in a "3 o'clock"

meeting the day before, and so, in addition to shifts in the control room, the representatives from each institution are responsible for rapid preliminary data analysis to inform the experimental run planning. CLOUD offers a unique training opportunity for young scientists not only in application of their own instruments and methods, but also in logistics and planning of a multi-faceted experimental run in an international environment.

CLOUD has two collaboration meetings each year, one during the fall experimental run at CERN, and one in the late winter or early spring in another location. The PIs and most researchers attend these meetings (60 CLOUD researchers attended the last meeting in Stockholm, 11–15 February 2019). The meetings combine extensive in-person work on specific data analysis tasks, planning and vetting of paper story-boards, and planning for future experimental runs. In addition to the collaboration meetings, members participate in one or two more focused CLOUD data workshops each year as well as ongoing video conferences to facilitate data reduction, collaborative data exchange, and overall interpretation.

5.2 CLOUD personnel

5.2.1 CLOUD Marie Curie ITN support

CLOUD has been supported by an unprecedented string of three Marie Skłowdonska Curie International Training Network (ITN) grants from the EC to support a large number of CLOUD doctoral students (15 in the current CLOUD-TRAIN project), as well as collaborative grants by the US National Science Foundation to support doctoral students and research fellows from the US. The success of these proposals is essential to ongoing CLOUD research, but the unparalleled scientific output of CLOUD has demonstrated the ability of the collaboration members and the entire team to secure funding ranging from individual investigator to ITN grants. The national funding agencies for collaboration member institutions have also supported institutional payments to the CLOUD Common Fund. During the next ten-year research period we plan to aggressively pursue a fourth ITN grant.

5.2.2 CERN team

The small CERN team is responsible for maintaining and developing the CLOUD facility and its infrastructure, comprising the chamber, thermal regulation system, thermometer system, sampling probes, mixing fans, HV clearing field, light sources, water purification and humidification system, cryogenic system, gas system, slow-control system, DAQ system, beam control and measurement, and T11 experimental zone infrastructure. In addition, the CERN team members are responsible for overall coordination of the CLOUD experiment in their capacities as spokesperson (Kirkby), technical coordinator (Mathot), resources coordinator (Onnela), run coordinator (Manninen), and DAQ coordinator (Weber). CLOUD is also very strongly supported by the EP-DT gas team. The small size of the CERN team means that each individual is critical for CERN to continue to fulfil its obligations to CLOUD.

CLOUD is not typical of a CERN experiment for reasons more than just its science. It is a 'living experiment' that is continually upgraded to improve its physics reach. The facility is continually developed in areas such as new light sources and optical components, new thermometer strings, specialised sampling probes, aerosol generators, gas generators, new trace gases and T11 zone infrastructure. The state-of-art mass spectrometers and other analysing instruments that sample the contents of the chamber are re-configured for each experimental run to match the experimental goals. They are shipped to CERN at the start of each run (Fig. 11), attached to the CLOUD chamber and brought into operation within a single installation week, to create a single, integrated experiment, and then removed and shipped back to the institutes at the end of each run. The very same instruments that analyse the CLOUD chamber are deployed in the field, providing an essential connection between the laboratory measurements and the real atmosphere that has proved to be a key component of CLOUD's success.

It represents a huge logistical challenge for the run coordinator to smoothly coordinate around 40 CLOUD instruments from 20 different institutes for each run—a task that extends throughout the

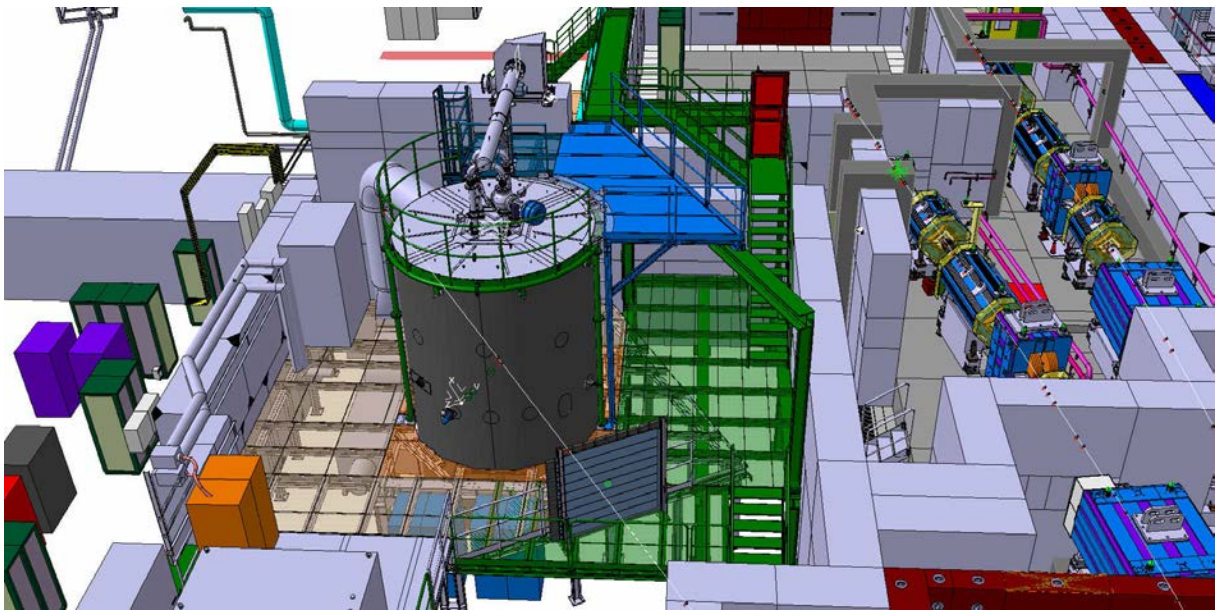**Fig. 11:** Boxes in the East Hall used to ship instruments to CERN for the CLOUD13 run, Sep–Nov 2013.

Fig. 12: 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.

year, not just during the experimental runs themselves. This includes all the safety requirements for each instrument, special gases, radioactive sources, etc., as well as coordinating accommodation for around 60 different people at the CERN hostel over a 10-week period. The work requires a full-time postdoctoral fellow at CERN with expert knowledge of atmospheric aerosol instrumentation and the scientific questions driving CLOUD's research. It is very efficiently carried out at present by a CERN applied fellow (Manninen). It is essential that CERN continues to support a run coordinator position for CLOUD at the end of the current applied fellow's contract (end 2019).

5.3 East Area renovation

A persistent difficulty for CLOUD has been the restricted space around the chamber to fit the mass spectrometers and other sampling instruments. There has been a similar lack of space for the gas system, which has grown in complexity with each year as more gases are added to the chamber (a total of around 25 different primary gases have been studied so far in the CLOUD chamber). With the East Area Renovation during LS2, 2019–2020, the space problem will be considerably improved (Fig. 12).

6 CONCLUDING REMARKS

CLOUD began its experimental journey 10 years ago. It was designed with a specific goal—to settle the cosmic-ray-cloud-climate question—but it was also designed as a precision and highly flexible experimental instrument that could be adapted to meet new science goals. The chamber and its associated gas system were built to the highest technical standards achievable, and relied heavily on CERN know-how. Together with new advanced instruments developed within the collaboration, CLOUD has become the world's leading experiment for studying atmospheric aerosol nucleation and growth in the laboratory—a subject of great importance to society through its impact on climate change and health.

We have outlined in this document the scientific contributions of CLOUD over the 10 years since it started to take data. With this experience, we can see into CLOUD's future far more clearly today than we could in 2009, and have outlined a 10-year run plan extending to 2029. We have no doubt that the next ten years for CLOUD will be as important and exciting as the first ten.They will allow CLOUD to fulfil its objectives to settle the cosmic-ray-climate mechanism, to reduce the uncertainties in anthropogenic climate change and to elucidate the mechanisms for urban particle pollution.

References

[1] L. Ahlm, T. Yli-Juuti, S. Schobesberger, A. P. Praplan, J. Kim, O.-P. Tikkanen, M. J. Lawler, J. N. Smith, J. Tröstl, J. C. A. Navarro, U. Baltensperger, F. Bianchi, N. M. Donahue, J. Duplissy, A. Franchin, T. Jokinen, H. Keskinen, J. Kirkby, A. Kürten, A. Laaksonen, K. Lehtipalo, T. Petäjä, F. Riccobono, M. P. Rissanen, L. Rondo, S. Schallhart, M. Simon, P. M. Winkler, D. R. Worsnop, A. Virtanen, and I. Riipinen. Modeling the thermodynamics and kinetics of sulfuric acid-dimethylamine-water nanoparticle growth in the CLOUD chamber. *Aerosol Science and Technology*, 50:1017–1032, 2016.

[2] J. Almeida, S. Schobesberger, A. Kürten, I. K. Ortega, O. Kupiainen, A. P. Praplan, A. Amorim, F. Bianchi, M. Breitenlechner, A. David, J. Dommen, N. M. Donahue, A. Downard, E. Dunne, J. Duplissy, S. Ehrhart, R. C. Flagan, A. Franchin, R. Guida, A. Hansel, H. Junninen, M. Kajos, H. Keskinen, A. Kupc, T. Kurtén, A. N. Kvashin, A. Laaksonen, K. Lehtipalo, J. Leppä, V. Loukonen, V. Makhmutov, S. Mathot, M. J. McGrath, T. Nieminen, T. Olenius, A. Onnela, T. Petäjä, F. Riccobono, I. Riipinen, L. Rondo, F. D. Santos, S. Schallhart, R. Schnitzhofer, J. H. Seinfeld, M. Sipilä, Y. Stozhkov, F. Stratmann, A. Tomé, G. Tsagkogeorgas, Y. Viisanen, A. Vrtala, P. E. Wagner, E. Weingartner, H. Wex, D. Wimmer, P. Ye, T. Yli-Juuti, K. S. Carslaw, M. Kulmala, J. Curtius, U. Baltensperger, D. R. Worsnop, H. Vehkamäki, and J. Kirkby. Molecular understanding of amine-sulphuric acid particle nucleation in the atmosphere. *Nature*, 502:359–363, 2013.

[3] D. Amaladhasan, C. Fuchs, C. Hoyle, PSI, and A. Zünd. Isoprene SOA AIOMFAC calculations. *Atmospheric Chemistry and Physics*, planned, Jul 2019.

[4] M. O. Andreae. The aerosol nucleation puzzle. *Science*, 339(6122):911–912, Feb. 2013.

[5] S. M. Ball, D. R. Hanson, F. L. Eisele, and P. H. McMurry. Laboratory studies of particle nucleation: Initial results for H_2SO_4, H_2O, and NH_3 vapors. *Journal of Geophysical Research: Atmospheres*, 104(D19):23709–23718, 1999.

[6] A. Bandy, I. C. Faloona, B. W. Blomquist, B. J. Huebert, A. D. Clarke, S. G. Howell, R. L. Mauldin, C. A. Cantrell, J. G. Hudson, B. G. Heikes, J. T. Merrill, Y. Wang, D. W. O'Sullivan, W. Nadler, and D. D. Davis. Pacific Atmospheric Sulfur Experiment (PASE): dynamics and chemistry of the south Pacific tropical trade wind regime. *Journal of Atmospheric Chemistry*, 68:5–25, 2011.

[7] D. R. Benson, L.-H. Young, F. R. Kameel, and S.-H. Lee. Laboratory-measured nucleation rates of sulfuric acid and water binary homogeneous nucleation from the SO_2 + OH reaction. *Geophysical Research Letters*, 35(11):L11801, 2008.

[8] T. Berndt, F. Stratmann, M. Sipilae, J. Vanhanen, T. Petaja, J. Mikkila, A. Gruener, G. Spindler, R. L. Mauldin, III, J. Curtius, M. Kulmala, and J. Heintzenberg. Laboratory study on new particle formation from the reaction OH + SO2: influence of experimental conditions, H2O vapour, NH3 and the amine tert-butylamine on the overall process. *Atmospheric Chemistry and Physics*, 10:7101–7116, 2010.

[9] A.-K. Bernhammer, M. Breitenlechner, F. N. Keutsch, and A. Hansel. Technical note: Conversion of isoprene hydroxy hydroperoxides (ISOPOOHs) on metal environmental simulation chamber walls. *Atmospheric Chemistry and Physics*, 17(6):4053–4062, 2017.

[10] A.-K. Bernhammer, L. Fischer, B. Mentler, M. Heinritzi, M. Simon, and A. Hansel. Impact of gas bottle impurities in the case of isoprene on oxidation product distribution observed by ultra-sensitive high resolution mass spectrometry. *Aerosol Measurement Techniques*, 11:4763–4773, 2018.

[11] F. Bianchi, J. Dommen, S. Mathot, and U. Baltensperger. On-line determination of ammonia at low pptv mixing ratios in the CLOUD chamber. *Atmospheric Measurement Techniques*, 5(7):1719–1725, 2012.

[12] F. Bianchi, A. P. Praplan, N. Sarnela, J. Dommen, A. Kürten, I. K. Ortega, S. Schobesberger, H. Junninen, M. Simon, J. Tröstl, T. Jokinen, M. Sipilä, A. Adamov, A. Amorim, J. Almeida, M. Breitenlechner, J. Duplissy, S. Ehrhart, R. C. Flagan, A. Franchin, J. Hakala, A. Hansel, M. Heinritzi, J. Kangasluoma, H. Keskinen, J. Kim, J. Kirkby, A. Laaksonen, M. J. Lawler, K. Lehtipalo, M. Leiminger, V. Makhmutov, S. Mathot, A. Onnela, T. Petäjä, F. Riccobono, M. P. Rissanen, L. Rondo, A. Tomé, A. Virtanen, Y. Viisanen, C. Williamson, D. Wimmer, P. M. Winkler, P. Ye, J. Curtius, M. Kulmala, D. R. Worsnop, N. M. Donahue, and U. Baltensperger. Insight into acid-base nucleation experiments by comparison of the chemical compositions of positive, negative and neutral clusters. *Environmental Science & Technology*, 48:13675–13684, 2014.

[13] M. Breitenlechner, L. Fischer, M. Hainer, M. Heinritzi, J. Curtius, and A. Hansel. PTR3: An instrument for studying the lifecycle of reactive organic carbon in the atmosphere. *Analytical Chemistry*, 89:5824–5831, 2017.

[14] K. S. Carslaw, R. G. Harrison, and J. Kirkby. Cosmic rays, clouds, and climate. *Science*, 298(5599):1732–1737, 2002.

[15] K. S. Carslaw, L. A. Lee, C. L. Reddington, K. J. Pringle, A. Rap, P. M. Forster, G. W. Mann, D. V. Spracklen, M. T. Woodhouse, L. A. Regayre, and J. R. Pierce. Large contribution of natural aerosols to uncertainty in indirect forcing. *Nature*, 503:67–71, 2013.

[16] R. J. Charlson, J. E. Lovelock, M. O. Andreae, and S. G. Warren. Ocean phytoplankton, atmospheric sulfur, cloud albedo, and climate. *Nature*, 326:655 – 661, 1987.

[17] J. G. Charney, A. Arakawa, D. J. Baker, B. Bolin, R. Dickinson, R. M. Goody, C. E. Leith, H. M. Stommel, and C. I. Wunsch. *Carbon Dioxide and Climate: A Scientific Assessment*. National Academy of Sciences, 1979.

[18] D. Chen, X. He, CMU, UHEL, and M. Sipilä. Constraints on particle composition during I_2 photo-oxidation using a Br^- FIGAERO CIMS. *Atmospheric Chemistry and Physics*, planned, Jun 2019.

[19] R. Chiu, L. Tinel, L. Gonzalez, R. Ciuraru, F. Bernard, C. George, and R. Volkamer. UV photochemistry of carboxylic acids at the air-sea boundary: A relevant source of glyoxal and other oxygenated VOC in the marine atmosphere. *Geophysical Research Letters*, 44(2):1079–1087, 2017.

[20] W. K. Chuang and N. M. Donahue. A 2-dimensional volatility basis set – part 3: Prognostic modeling and NO_x dependence. *Atmospheric Chemistry and Physics*, 16:123–134, 2016.

[21] W. K. Chuang and N. M. Donahue. Extremely Low Volatility Organic Compounds (ELVOCs), dynamics, and smog-chamber secondary organic aerosol formation. *Atmospheric Chemistry and Physics*, 17:10019–10036, 2017.

[22] S. Coburn, I. Ortega, R. Thalman, B. Blomquist, C. W. Fairall, and R. Volkamer. Measurements of diurnal variations and eddy covariance (EC) fluxes of glyoxal in the tropical marine boundary layer: description of the Fast LED-CE-DOAS instrument. *Atmospheric Measurement Techniques*, 7(10):3579–3595, 2014.

[23] A. Cohen, M. Brauer, R. Burnett, H. R. Anderson, J. Frostad, K. Estep, K. Balakrishnan, B. Brunekreef, L. Dandona, R. Dandona, V. Feigin, G. Freedman, B. Hubbell, A. Jobling, H. Kan, L. Knibbs, Y. Liu, R. Martin, L. Morawska, C. A. P. III, H. Shin, K. Straif, G. Shaddick, M. Thomas, R. V. Dingenen, A. V. Donkelaar, T. Vos, C. J. L. Murray, and M. H. Forouzanfar. Estimates and 25-year trends of the global burden of disease attributable to ambient air pollution: an analysis of data from the Global Burden of Diseases Study 2015. *The Lancet*, 389(10082):1907–1918, May 2017.

[24] J. D. Crounse, L. B. Nielsen, S. Jørgensen, H. G. Kjaergaard, and P. O. Wennberg. Autoxidation of organic compounds in the atmosphere. *Journal of Physical Chemistry Letters*, 4(20):3513–3520, 2013.

[25] L. Dada, J. Kirkby, and CLOUD. New particle formtion from pristine biogenic to polluted environments. *Nature*, planned, Jul 2019.

[26] L. Dada, UHEL, and J. Kirkby. The effect of sesquiterpene oxidation products on nucleation and growth of particles. *Atmospheric Chemistry and Physics*, in preparation, Feb 2019.

[27] A. Dias, S. Ehrhart, A. Vogel, C. Williamson, J. Almeida, J. Kirkby, S. Mathot, S. Mumford, and A. Onnela. Temperature uniformity in the CERN CLOUD chamber. *Aerosol Measurement Techniques*, 10:5075–5088, 2017.

[28] N. M. Donahue, S. A. Epstein, S. N. Pandis, and A. L. Robinson. A two-dimensional volatility basis set: 1. Organic mixing thermodynamics. *Atmospheric Chemistry and Physics*, 11:3303–3318, 2011.

[29] N. M. Donahue, K. M. Henry, T. F. Mentel, A. K. Scharr, C. Spindler, B. Bohn, T. Brauers, H. P. Dorn, H. Fuchs, R. Tillmann, A. Wahner, H. Saathoff, K. H. Naumann, O. Möhler, T. Leisner, L. Müller, M.-C. Reinnig, T. Hoffmann, K. Salow, M. Hallquist, M. Frosch, M. Bilde, T. Tritscher, P. Barmet, A. P. Praplan, P. F. DeCarlo, J. Dommen, A. S. H. Prévôt, and U. Baltensperger. Aging of biogenic secondary organic aerosol via gas-phase OH radical reactions. *Proceedings of the National Academy of Sciences*, 109:13503–13508, 2012.

[30] N. M. Donahue, K. E. Huff Hartz, B. Chuong, A. A. Presto, C. O. Stanier, T. Rosenørn, A. L. Robinson, and S. N. Pandis. Critical factors determining the variation in SOA yields from terpene ozonolysis: A combined experimental and computational study. *Faraday Discussions*, 130:295–309, 2005.

[31] N. M. Donahue, J. H. Kroll, A. L. Robinson, and S. N. Pandis. A two-dimensional volatility basis set - part 2: Diagnostics of organic-aerosol evolution. *Atmospheric Chemistry and Physics*, 12:615–634, 2012.

[32] N. M. Donahue, I. K. Ortega, W. Chuang, I. Riipinen, F. Riccobono, S. Schobesberger, J. Dommen, U. Baltensperger, M. Kulmala, D. R. Worsnop, and H. Vehkamaki. How do organic vapors contribute to new-particle formation? *Faraday Discussions*, 16:91–104, 2013.

[33] N. M. Donahue, L. N. Posner, D. M. Westervelt, Z. Li, M. Shrivastava, A. A. Presto, R. C. Sullivan, P. J. Adams, S. N. Pandis, and A. L. Robinson. Where did this particle come from? Sources of particle number and mass for human exposure estimates. In *Airborne Particulate Matter: Sources, Atmospheric Processes and Health*, pages 35–71. The Royal Society of Chemistry, 2016.

[34] N. M. Donahue, A. L. Robinson, C. O. Stanier, and S. N. Pandis. Coupled partitioning, dilution, and chemical aging of semivolatile organics. *Environmental Science & Technology*, 40:2635 – 2643, 2006.

[35] N. M. Donahue, E. R. Trump, I. Riipinen, and J. R. Pierce. Theoretical constraints on pure vapor-pressure driven condensation of organics to ultrafine particles. *Geophysical Research Letters*, 38:L16801, 2011.

[36] D. Draper, M. Lawler, and J. Smith. A comparison of nanoparticle composition from a Finnish boreal forest and representative chamber studies. *Atmospheric Chemistry and Physics*, planned, Jun 2019.

[37] E. M. Dunne, H. Gordon, A. Kürten, J. Almeida, J. Duplissy, C. Williamson, I. K. Ortega, K. J. Pringle, A. Adamov, U. Baltensperger, P. Barmet, F. Benduhn, F. Bianchi, M. Breitenlechner, A. Clarke, J. Curtius, J. Dommen, N. M. Donahue, S. Ehrhart, R. C. Flagan, A. Franchin, R. Guida, J. Hakala, A. Hansel, M. Heinritzi, T. Jokinen, J. Kangasluoma, J. Kirkby, M. Kulmala, A. Kupc, M. J. Lawler, K. Lehtipalo, V. Makhmutov, G. Mann, S. Mathot, J. Merikanto, P. Miettinen, A. Nenes, A. Onnela, A. Rap, C. L. S. Reddington, F. Riccobono, N. A. D. Richards, M. P. Rissanen, L. Rondo, N. Sarnela, S. Schobesberger, K. Sengupta, M. Simon, M. Sipilä, J. N. Smith, Y. Stozhkov, A. Tomé, J. Tröstl, P. E. Wagner, D. Wimmer, P. M. Winkler, D. R. Worsnop, and K. S. Carslaw. Global atmospheric particle formation from CERN CLOUD measurements. *Science*, 354:1119–1124, 2016.

[38] E. M. Dunne, L. A. Lee, C. L. Reddington, and K. S. Carslaw. No statistically significant effect of a short-term decrease in the nucleation rate on atmospheric aerosols. *Atmospheric Chemistry and Physics*, 12(23):11573–11587, 2012.

[39] J. Duplissy, M. B. Enghoff, K. L. Aplin, F. Arnold, H. Aufmhoff, M. Avngaard, U. Baltensperger, T. Bondo, R. Bingham, K. Carslaw, J. Curtius, A. David, B. Fastrup, S. Gagné, F. Hahn, R. G. Harrison, B. Kellett, J. Kirkby, M. Kulmala, L. Laakso, A. Laaksonen, E. Lillestol, M. Lockwood, J. Mäkelä, V. Makhmutov, N. D. Marsh, T. Nieminen, A. Onnela, E. Pedersen, J. O. P. Pedersen, J. Polny, U. Reichl, J. H. Seinfeld, M. Sipilä, Y. Stozhkov, F. Stratmann, H. Svensmark, J. Svensmark, R. Veenhof, B. Verheggen, Y. Viisanen, P. E. Wagner, G. Wehrle, E. Weingartner, H. Wex, M. Wilhelmsson, and P. M. Winkler. Results from the CERN pilot CLOUD experiment. *Atmospheric Chemistry and Physics*, 10(4):1635–1647, 2010.

[40] J. Duplissy, J. Merikanto, A. Franchin, G. Tsagkogeorgas, J. Kangasluoma, D. Wimmer, H. Vuollekoski, S. Schobesberger, K. Lehtipalo, R. C. Flagan, D. Brus, N. M. Donahue, H. Vehkamäki, J. Almeida, A. Amorim, P. Barmet, F. Bianchi, M. Breitenlechner, E. M. Dunne, R. Guida, H. Henschel, H. Junninen, J. Kirkby, A. Kürten, A. Kupc, A. Määttänen, V. Makhmutov, S. Mathot, T. Nieminen, A. Onnela, A. P. Praplan, F. Riccobono, L. Rondo, G. Steiner, A. Tome, H. Walther, U. Baltensperger, K. S. Carslaw, J. Dommen, A. Hansel, T. Petäjä, M. Sipilä, F. Stratmann, A. Vrtala, P. E. Wagner, D. R. Worsnop, J. Curtius, and

M. Kulmala. Effect of ions on sulfuric acid-water binary particle formation II: Experimental data and comparison with QC-normalized classical nucleation theory. *Journal of Geophysical Research Atmospheres*, 212:1752–1775, 2016.

[41] M. Ehn, J. A. Thornton, E. Kleist, M. Sipila, H. Junninen, I. Pullinen, M. Springer, F. Rubach, R. Tillmann, B. Lee, F. Lopez-Hilfiker, S. Andres, I.-H. Acir, M. Rissanen, T. Jokinen, S. Schobesberger, J. Kangasluoma, J. Kontkanen, T. Nieminen, T. Kurten, L. B. Nielsen, S. Jorgensen, H. G. Kjaergaard, M. Canagaratna, M. D. Maso, T. Berndt, T. Petaja, A. Wahner, V.-M. Kerminen, M. Kulmala, D. R. Worsnop, J. Wildt, and T. F. Mentel. A large source of low-volatility secondary organic aerosol. *Nature*, 506:476–479, 2014.

[42] S. Ehrhart and J. Curtius. Influence of aerosol lifetime on the interpretation of nucleation experiments with respect to the first nucleation theorem. *Atmospheric Chemistry and Physics*, 13(22):11465–11471, 2013.

[43] S. Ehrhart, L. Ickes, J. Almeida, A. Amorim, P. Barmet, F. Bianchi, J. Dommen, E. M. Dunne, J. Duplissy, A. Franchin, J. Kangasluoma, J. Kirkby, A. Kupc, K. Lehtipalo, T. Nieminen, F. Riccobono, L. Rondo, S. Schobesberger, G. Steiner, A. Tomé, D. Wimmer, U. Baltensperger, P. E. Wagner, and J. Curtius. Comparison of the SAWNUC model with CLOUD measurements of sulphuric acid-water nucleation. *Journal of Geophysical Research Atmospheres*, 121:12,401–12,414, 2016.

[44] H. Finkenzeller, CMU, and CU. Effects of humidity on the composition and growth of nanoparticles. *Atmospheric Chemistry and Physics*, planned, Aug 2019.

[45] H. Finkenzeller, UHEL, and CU. Photochemical modeling of iiodic acid yields during photochemical oxidation of I_2. *Journal of Physical Chemistry A*, planned, Jun 2019.

[46] L. Fischer and UINS. Detailed chemical mechanism in CLOUD-10,11 and PTR-3 data. *Atmospheric Chemistry and Physics*, planned, Jun 2019.

[47] A. Franchin, A. J. Downard, J. Kangasluoma, T. Nieminen, K. Lehtipalo, G. Steiner, H. E. Manninen, T. Petäjä, R. C. Flagan, and M. Kulmala. A new high transmission inlet for the Caltech nano-RDMA for size distribution measurements of sub-3 nm ions at ambient concentrations. *Atmospheric Measurement Techniques*, 9:2709–2720, 2016.

[48] A. Franchin, S. Ehrhart, J. Leppä, T. Nieminen, S. Gagné, S. Schobesberger, D. Wimmer, J. Duplissy, F. Riccobono, E. M. Dunne, L. Rondo, A. Downard, F. Bianchi, A. Kupc, G. Tsagkogeorgas, K. Lehtipalo, H. E. Manninen, J. Almeida, A. Amorim, P. E. Wagner, A. Hansel, J. Kirkby, A. Kürten, N. M. Donahue, V. Makhmutov, S. Mathot, A. Metzger, T. Petäjä, R. Schnitzhofer, M. Sipilä, Y. Stozhkov, A. Tomé, V.-M. Kerminen, K. Carslaw, J. Curtius, U. Baltensperger, and M. Kulmala. Experimental investigation of ion-ion recombination at atmospheric conditions. *Atmospheric Chemistry and Physics*, 15:7203–7216, 2015.

[49] C. Frege, I. K. Ortega, M. P. Rissanen, A. P. Praplan, G. Steiner, M. Heinritzi, L. Ahonen, A. Amorim, A.-K. Bernhammer, F. Bianchi, S. Brilke, M. Breitenlechner, L. Dada, A. Dias, J. Duplissy, S. Ehrhart, I. El-Haddad, L. Fischer, C. Fuchs, O. Garmash, M. Gonin, A. Hansel, C. R. Hoyle, T. Jokinen, H. Junninen, J. Kirkby, A. Kürten, K. Lehtipalo, M. Leiminger, R. L. Mauldin, U. Molteni, L. Nichman, T. Petäjä, N. Sarnela, S. Schobesberger, M. Simon, M. Sipilä, D. Stolzenburg, A. Tomé, A. L. Vogel, A. C. Wagner, R. Wagner, M. Xiao, C. Yan, P. Ye, J. Curtius, N. M. Donahue, R. C. Flagan, M. Kulmala, D. R. Worsnop, P. M. Winkler, J. Dommen, and U. Baltensperger. Influence of temperature on the molecular composition of ions and charged clusters during pure biogenic nucleation. *Atmospheric Chemistry and Physics*, 18:65–79, 2018.

[50] C. Fuchs and PSI. Vapour wall loss correction of SOA mass yields at sub-saturated conditions in the CLOUD chamber: Assessing the effect of NO_x, relative humidity and temperature on SOA yields. *Atmospheric Measurement Techniques*, in preparation, Mar 2019.

[51] T. M. Gaydos, C. O. Stanier, S. N. Pandis, and S. N. Pandis. Modeling of in situ ultrafine atmospheric particle formation in the eastern United States. *Journal of Geophysical Research Atmospheres*, 110:D07S12, 2005.

[52] A. H. Goldstein and I. E. Galbally. Known and unexplored organic constituents in the Earth's atmosphere. *Environ. Sci. Technol.*, 41:1515–1521, 2007.

[53] L. Gonzales, D. Stolzenburg, UVIE, and CLOUD. Size-dependent growth rates of particles between 1 and 10 nm for a range of chemical systems. *Atmospheric Chemistry and Physics*, planned, Sep 2019.

[54] H. Gordon, J. Kirkby, U. Baltensperger, F. Bianchi, M. Breitenlechner, J. Curtius, A. Dias, J. Dommen, N. M. Donahue, E. M. Dunne, J. Duplissy, S. Ehrhart, R. C. Flagan, C. Frege, C. Fuchs, A. Hansel, C. R. Hoyle, M. Kulmala, A. Kürten, K. Lehtipalo, V. Makhmutov, U. Molteni, M. P. Rissanen, Y. Stozhkov, J. Tröstl, G. Tsagkogeorgas, R. Wagner, C. Williamson, D. Wimmer, P. M. Winkler, C. Yan, and K. S. Carslaw. Causes and importance of new particle formation in the present-day and pre-industrial atmospheres. *Journal of Geophysical Research Atmospheres*, 122:8739–8760, 2017.

[55] H. Gordon, Leeds, and CLOUD. Can cloud biogenic really represent boundary layer npf? *Atmospheric Chemistry and Physics*, in preparation, Dec 2019.

[56] H. Gordon, S. Schuchmann, and CLOUD. Parameterization of multicomponent nucleation throughout the troposphere beased on CLOUD results. *Nature Climate Change*, planned, Jul 2019.

[57] H. Gordon, K. Sengupta, A. Rap, J. Duplissy, C. Frege, C. Williamson, M. Heinritzi, M. Simon, C. Yan, J. Almeida, J. Tröstl, T. Nieminen, I. K. Ortega, R. Wagner, E. M. Dunne, A. Adamov, A. Amorim, A.-K. Bernhammer, F. Bianchi, M. Breitenlechner, S. Brilke, X. Chen, J. S. Craven, A. Dias, S. Ehrhart, L. Fischer, R. C. Flagan, A. Franchin, C. Fuchs, R. Guida, J. Hakala, C. R. Hoyle, T. Jokinen, H. Junninen, J. Kangasluoma, J. Kim, J. Kirkby, M. Krapf, A. Kürten, A. Laaksonen, K. Lehtipalo, V. Makhmutov, S. Mathot, U. Molteni, S. A. Monks, A. Onnela, O. Peräkylä, F. Piel, T. Petäjä, A. P. Praplan, K. J. Pringle, N. A. D. Richards, M. P. Rissanen, L. Rondo, N. Sarnela, S. Schobesberger, C. E. Scott, J. H. Seinfeld, S. Sharma, M. Sipilä, G. Steiner, Y. Stozhkov, F. Stratmann, A. Tomé, A. Virtanen, A. L. Vogel, A. C. Wagner, P. E. Wagner, E. Weingartner, D. Wimmer, P. M. Winkler, P. Ye, X. Zhang, A. Hansel, J. Dommen, N. M. Donahue, D. R. Worsnop, U. Baltensperger, M. Kulmala, J. Curtius, and K. S. Carslaw. Reduced anthropogenic aerosol radiative forcing caused by biogenic new particle formation. *Proceedings of the National Academy of Sciences*, 113(43):12053–12058, 2016.

[58] S. Guo, M. Hu, M. L. Zamora, J. Peng, D. Shang, J. Zheng, Z. Du, Z. Wu, M. Shao, L. Zeng, M. J. Molina, and R. Zhang. Elucidating severe urban haze formation in china. *Proceedings of the National Academy of Sciences*, 111(49):17373–17378, 2014.

[59] M. Hallquist, J. C. Wenger, U. Baltensperger, Y. Rudich, D. Simpson, M. Claeys, J. Dommen, N. M. Donahue, C. George, A. H. Goldstein, J. F. Hamilton, H. Herrmann, T. Hoffmann, Y. Iinuma, M. Jang, M. E. Jenkin, J. L. Jimenez, A. Kiendler-Scharr, W. Maenhaut, G. McFiggans, T. F. Mentel, A. Monod, A. S. H. Prévôt, J. H. Seinfeld, J. D. Surratt, R. Szmigielski, and J. Wildt. The formation, properties and impact of secondary organic aerosol: current and emerging issues. *Atmospheric Chemistry and Physics*, 10:5155–5236, 2009.

[60] X. He, J. Kirkby, UHEL, CLOUD, and M. Kulmala. Direct measurement of ion-molecule collision rates. *Physics Review Letters*, in preparation, Feb 2019.

[61] X. He, UHEL, and CLOUD. From the formation of HIO_3 to the growth of iodic particles. *Nature*, planned, Jun 2019.

[62] M. Heinritzi, L. Dada, M. Simon, D. Stolzenburg, A. C. Wagner, L. Fischer, L. R. Ahonen, A. Baccarini, P. S. Bauer, B. Baumgartner, F. Bianchi, S. Brilke, D. Chen, A. Dias, J. Dommen, J. Duplissy, H. Finkenzeller, C. Frege, C. Fuchs, O. Garmash, H. Gordon1, I. E. Haddad, X. He, J. Helm, V. Hofbauer, C. R. Hoyle1, J. Kangasluoma, T. Keber, C. Kim, A. Kürten, J. Lampilahti, T. M. Laurila, K. Lehtipalo, M. Leiminger, H. Mai1, V. Makhmutov, S. Mathot, B. Mentler, U. Molteni, W. Nie, T. Nieminen, A. Onnela, E. Partoll, M. Passananti, T. Petäjä, V. Pospisilova, L. L. J. Quéléver, M. P. Rissanen, C. Rose, M. Sipilä, Y. Stozhkov1, C. Tauber, A. L. Vogel, R. Volkamer, R. Wagner, M. Wang, L. Weitz1, D. Wimmer, M. Xiao, C. Yan, P. Ye1, Q. Zha, A. Amorim, U. Baltensperger, A. Hansel, M. Kulmala, A. Tomé, P. M. Winkler, D. R. Worsnop, N. M. Donahue, J. Kirkby, and J. Curtius. Molecular understanding of the suppression of aerosol nucleation by isoprene. *Proceedings of the National Academy of Sciences*, submitted, 2019.

[63] M. Heinritzi, M. Simon, G. Steiner, A. C. Wagner, A. Kürten, A. Hansel, and J. Curtius. Characterization of the mass-dependent transmission efficiency of a CIMS. *Atmospheric Measurement Techniques*, 9(4):1449–1460, 2016.

[64] K. M. Henry and N. M. Donahue. Photochemical aging of α-pinene secondary organic aerosol: effects of OH radical sources and photolysis. *Journal of Physical Chemistry A*, 116:5932–5940, 2012.

[65] V. Hofbauer, H. Finkenzeller, P. Ye, D. Chen, M. Wang, Q. Ye, R. L. Mauldin III, CLOUD, R. Volkamer, and N. M. Donahue. Determining photolysis rates in the CLOUD chamber from multiple illumination sources. *Atmospheric Measurement Techniques*, in preparation, Jan 2019.

[66] V. Hofbauer, M. Heinritzi, U. Molteni, UFRA, PSI, CMU, and CLOUD. Condensation properties of HOMs onto particles and chamber walls. *Atmospheric Chemistry and Physics*, planned, Dec 2019.

[67] C. R. Hoyle, C. Fuchs, E. Järvinen, H. Saathoff, A. Dias, I. El Haddad, M. Gysel, S. C. Coburn, J. Tröstl, A.-K. Bernhammer, F. Bianchi, M. Breitenlechner, J. C. Corbin, J. Craven, N. M. Donahue, J. Duplissy, S. Ehrhart, C. Frege, H. Gordon, N. Höppel, M. Heinritzi, T. B. Kristensen, U. Molteni, L. Nichman, T. Pinterich, A. S. H. Prévôt, M. Simon, J. G. Slowik, G. Steiner, A. Tomé, A. L. Vogel, R. Volkamer, A. C. Wagner, R. Wagner, A. S. Wexler, C. Williamson, P. M. Winkler, C. Yan, A. Amorim, J. Dommen, J. Curtius, M. W. Gallagher, R. C. Flagan, A. Hansel, J. Kirkby, M. Kulmala, O. Möhler, F. Stratmann, D. R. Worsnop, and U. Baltensperger. Aqueous phase oxidation of sulphur dioxide by ozone in cloud droplets. *Atmospheric Chemistry and Physics*, 16:1693–1712, 2016.

[68] K. Ignatius, T. B. Kristensen, E. Järvinen, L. Nichman, C. Fuchs, H. Gordon, P. Herenz, C. R. Hoyle, J. Duplissy, S. Garimella, A. Dias, C. Frege, N. Höppel, J. Tröstl, R. Wagner, C. Yan, A. Amorim, U. Baltensperger, J. Curtius, N. M. Donahue, M. W. Gallagher, J. Kirkby, M. Kulmala, O. Möhler, H. Saathoff, M. Schnaiter, A. Tomé, A. Virtanen, D. Worsnop, and F. Stratmann. Heterogeneous ice nucleation of viscous secondary organic aerosol produced from ozonolysis of α-pinene. *Atmospheric Chemistry and Physics*, 16:6495–6509, 2016.

[69] IPCC. *Climate Change 2013: The Physical Science Basis*. Cambridge University Press, 32 Avenue of the Americas, New York, NY 10013-2473, USA, 2013.

[70] E. Järvinen, K. Ignatius, L. Nichman, T. B. Kristensen, C. Fuchs, C. R. Hoyle, N. Höppel, J. C. Corbin, J. Craven, J. Duplissy, S. Ehrhart, I. El Haddad, C. Frege, H. Gordon, T. Jokinen, P. Kallinger, J. Kirkby, A. Kiselev, K.-H. Naumann, T. Petäjä, T. Pinterich, A. S. H. Prevot, H. Saathoff, T. Schiebel, K. Sengupta, M. Simon, J. G. Slowik, J. Tröstl, A. Virtanen, P. Vochezer, S. Vogt, A. C. Wagner, R. Wagner, C. Williamson, P. M. Winkler, C. Yan, U. Baltensperger, N. M. Donahue, R. C. Flagan, M. Gallagher, A. Hansel, M. Kulmala, F. Stratmann, D. R. Worsnop, O. Möhler, T. Leisner, and M. Schnaiter. Observation of viscosity transition in α-pinene secondary organic aerosol. *Atmospheric Chemistry and Physics*, 16:4423–4438, 2016.

[71] C. N. Jen, P. H. McMurry, and D. R. Hanson. Stabilization of sulfuric acid dimers by ammonia, methylamine, dimethylamine, and trimethylamine. *Journal of Geophysical Research Atmospheres*, 119(12):7502–7514, 2014. 2014JD021592.

[72] T. Jokinen, T. Berndt, R. Makkonen, V.-M. Kerminen, H. Junninen, P. Paasonen, F. Stratmann, H. Herrmann, A. B. Guenther, D. R. Worsnop, M. Kulmala, M. Ehn, and M. Sipilä. Production of extremely low volatile organic compounds from biogenic emissions: Measured yields and atmospheric implications. *Proceedings of the National Academy of Sciences*, 112(23):7123–7128, 2015.

[73] H. Keskinen, A. Virtanen, J. Joutsensaari, G. Tsagkogeorgas, J. Duplissy, S. Schobesberger, M. Gysel, F. Riccobono, J. G. Slowik, F. Bianchi, T. Yli-Juuti, K. Lehtipalo, L. Rondo, M. Breitenlechner, A. Kupc, J. Almeida, A. Amorin, E. M. Dunne, A. J. Downward, S. Ehrhart, A. Franchin, M. Kajos, J. Kirkby, A. Kürten, T. Nieminen, V. Makhmutov, S. Mathot, P. Miettinen, A. Onnela, T. Petäjä, A. Praplan, F. Santos, S. Schallhart, M. Sipilä, Y. Stozhkov, A. Tomé, P. Vaattovaara, D. Wimmer, A. Prevot, J. Dommen, N. M. Donahue, R. Flagan, E. Weingartner, Y.Viisanen, I. Riipinen, A. Hansel, J. Curtius, M. Kulmala, D. R. Worsnop, U. Baltensperger, H. Wex, F. Stratmann, and A. Laaksonen. Evolution of particle composition in CLOUD nucleation experiments. *Atmospheric Chemistry and Physics*, 12:5587–5600, 2013.

[74] C. Kim, S. Amanatidis, S. Kong, R. Flagan, Caltech, and CLOUD. Nano rDMA and ROMIAC characterization. *Aerosol Science and Technology*, planned, Aug 2019.

[75] J. Kim, L. Ahlm, H. Keskinen, M. Lawler, J. Tröstl, J. Duplissy, T. Yli-Juuti, E. Lehtinen, J. Smith, I. Riipinen, A. Kürten, F. Bianchi, N. M. Donahue, P. Miettinen, A. Amorim, A. Laaksonen, A. Tomé, C. Williamson, D. Wimmer, J. Hakala, J. Kirkby, K. Lehtipalo, K. Sengupta, L. Rondo, M. Heinritzi, P. M. Winkler, M. P. Rissanen, M. Simon, P. Ye, R. C. Flagan, S. Schobesberger, T. Jokinen, T. Petäjä, M. Kulmala, and A. Virtanen. Hygroscopicity of nanoparticles produced from homogeneous nucleation in the CLOUD experiments. *Atmospheric Chemistry and Physics*, 16:293–304, 2016.

[76] J. Kirkby. Beam measurements of a CLOUD (Cosmics Leaving OUtdoor Droplets) chamber. *CERN-OPEN-2001-028*, 1998. https://arxiv.org/abs/physics/0104041v2.

[77] J. Kirkby. Cosmic rays and climate. *Surveys in Geophysics*, 28(5-6):333–375, Feb. 2008.

[78] J. Kirkby, J. Curtius, J. Almeida, E. Dunne, J. Duplissy, S. Ehrhart, A. Franchin, S. Gagne, L. Ickes, A. Kurten, A. Kupc, A. Metzger, F. Riccobono, L. Rondo, S. Schobesberger, G. Tsagkogeorgas, D. Wimmer, A. Amorim, F. Bianchi, M. Breitenlechner, A. David, J. Dommen, A. Downard, M. Ehn, R. C. Flagan, S. Haider, A. Hansel, D. Hauser, W. Jud, H. Junninen, F. Kreissl, A. Kvashin, A. Laaksonen, K. Lehtipalo, J. Lima, E. R. Lovejoy, V. Makhmutov, S. Mathot, J. Mikkila, P. Minginette, S. Mogo, T. Nieminen, A. Onnela, P. Pereira, T. Petaja, R. Schnitzhofer, J. H. Seinfeld, M. Sipila, Y. Stozhkov, F. Stratmann, A. Tome, J. Vanhanen, Y. Viisanen, A. Vrtala, P. E. Wagner, H. Walther, E. Weingartner, H. Wex, P. M. Winkler, K. S. Carslaw, D. R. Worsnop, U. Baltensperger, and M. Kulmala. Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation. *Nature*, 476(7361):429–433, Aug 2011.

[79] J. Kirkby, J. Duplissy, K. Sengupta, C. Frege, H. Gordon, C. Williamson, M. Heinritzi, M. Simon, C. Yan, J. Almeida, J. Tröstl, T. Nieminen, I. K. Ortega, R. Wagner, A. Adamov, A. Amorim, A.-K. Bernhammer, F. Bianchi, M. Breitenlechner, S. Brilke, X. Chen, J. Craven, A. Dias, S. Ehrhart, R. C. Flagan, A. Franchin, C. Fuchs, R. Guida, J. Hakala, C. R. Hoyle, T. Jokinen, H. Junninen, J. Kangasluoma, J. Kim, M. Krapf, A. Kürten, A. Laaksonen, K. Lehtipalo, V. Makhmutov, S. Mathot, U. Molteni, A. Onnela, O. Peräkylä, F. Piel, T. Petäjä, A. P. Praplan, K. Pringle, A. Rap, N. A. Richards, I. Riipinen, M. P. Rissanen, L. Rondo, N. Sarnela, S. Schobesberger, C. E. Scott, J. H. Seinfeld, M. Sipilä, G. Steiner, Y. Stozhkov, F. Stratmann, A. Tomé, A. Virtanen, A. L. Vogel, A. Wagner, P. E. Wagner, E. Weingartner, D. Wimmer, P. M. Winkler, P. Ye, X. Zhang, A. Hansel, J. Dommen, N. M. Donahue, D. R. Worsnop, U. Baltensperger, M. Kulmala, K. S. Carslaw, and J. Curtius. Ion-induced nucleation of pure biogenic particles. *Nature*, 530:521–526, 2016.

[80] J. H. Kroll and J. H. Seinfeld. Chemistry of secondary organic aerosol: Formation and evolution of low-volatility organics in the atmosphere. *Atmospheric Environment*, 42(16):3593–3624, MAY 2008.

[81] M. Kulmala, L. Laakso, K. E. J. Lehtinen, I. Riipinen, M. Dal Maso, T. Anttila, V.-M. Kerminen, U. Hõrrak, M. Vana, and H. Tammet. Initial steps of aerosol growth. *Atmospheric Chemistry and Physics*, 4:2553–2560, 2004.

[82] A. Kupc, A. Amorim, J. Curtius, A. Danielczok, J. Duplissy, S. Ehrhart, H. Walther, L. Ickes, J. Kirkby, A. Kürten, J. Lima, S. Mathot, P. Minginette, A. Onnela, L. Rondo, and P. Wagner. A fibre-optic UV system for H$_2$SO$_4$ production in aerosol chambers causing minimal thermal effects. *Journal of Aerosol Science*, 42(8):532 – 543, 2011.

[83] A. Kürten, F. Bianchi, J. Almeida, Kupiainen-Maata, E. M. Dunne, J. Duplissy, P. Barmet, M. Breitenlechner, J. Dommen, N. M. Donahue, R. C. Flagan, A. Franchin, J. Hakala, A. Hansel, M. Heinritzi, L. Ickes, T. Jokinen, J. Kangasluoma, J. Kim, J. Kirkby, A. Kupc, K. Lehtipalo, M. Leiminger, V. Makhmutov, A. Onnela, I. K. Ortega, T. Petäjä, A. P. Praplan, F. Riccobono, M. Rissanen, L. Rondo, R. Schnizhofer, S. Schobesberger, J. N. Smith, G. Steiner, Y. Stozhkov, A. Tomé, J. Tröstl, G. Tsagkogeorgas, P. E. Wagner, C. Williamson, D. Wimmer, P. Ye, U. Baltensperger, K. Carslaw, M. Kulmala, and J. Curtius. Experimental particle formation rates spanning tropospheric conditions of sulfuric acid, ammonia, ions, and temperature. *Journal of Geophysical Research Atmospheres*, 121:12,377–12,400, 2016.

[84] A. Kürten, T. Jokinen, M. Simon, M. Sipilä, N. Sarnela, H. Junninen, A. Adamov, J. Almeida, A. Amorim, F. Bianchi, M. Breitenlechner, J. Dommen, N. M. Donahue, J. Duplissy, S. Ehrhart, R. C. Flagan, A. Franchin, J. Hakala, A. Hansel, M. Heinritzi, M. Hutterli, A. Laaksonen, K. Lehtipalo, M. Leiminger, V. Makhmutov, S. Mathot, A. Onnela, T. Petäjä, A. P. Praplan, L. Rondo, S. Schobesberger, J. H. Seinfeld,

F. Stratmann, A. Tomé, J. Tröstl, P. E. Wagner, E. Weingartner, C. Williamson, D. Wimmer, P. Ye, U. Baltensperger, K. S. Carslaw, M. Kulmala, D. R. Worsnop, J. Kirkby, and J. Curtius. Neutral molecular cluster formation of sulfuric acid-dimethylamine observed in real time under atmospheric conditions. *Proceedings of the National Academy of Sciences*, 111:15019–15024, 2014.

[85] A. Kürten, C. Li, F. Bianchi, J. Curtius, A. Dias, N. M. Donahue, J. Duplissy, R. C. Flagan, J. Hakala, T. Jokinen, J. Kirkby, M. Kulmala, A. Laaksonen, K. Lehtipalo, V. Makhmutov, A. Onnela, M. P. Rissanen, M. Simon, M. Sipilä, Y. Stozhkov, J. Tröstl, P. Ye, and P. H. McMurry. New particle formation in the sulfuric acid-dimethylamine-water system: Reevaluation of CLOUD chamber measurements and comparison to an aerosol nucleation and growth model. *Atmospheric Chemistry and Physics*, 18:845–863, 2018.

[86] A. Kürten, S. Münch, L. Rondo, F. Bianchi, J. Duplissy, T. Jokinen, H. Junninen, N. Sarnela, S. Schobesberger, M. Simon, M. Sipilä, J. Almeida, A. Amorim, J. Dommen, N. M. Donahue, E. M. Dunne, R. C. Flagan, A. Franchin, J. Kirkby, A. Kupc, V. Makhmutov, T. Petäjä, A. P. Praplan, F. Riccobono, G. Steiner, A. Tomé, G. Tsagkogeorgas, P. E. Wagner, D. Wimmer, U. Baltensperger, M. Kulmala, D. R. Worsnop, and J. Curtius. Thermodynamics of the formation of sulfuric acid dimers in the binary (H_2SO_4-H_2O) and ternary (H_2SO_4-H_2O-NH_3) system. *Atmospheric Chemistry and Physics*, 15:10701–10721, 2015.

[87] A. Kürten, L. Rondo, S. Ehrhart, and J. Curtius. Performance of a corona ion source for measurement of sulfuric acid by chemical ionization mass spectrometry. *Atmospheric Measurement Techniques*, 4(3):437–443, 2011.

[88] A. Kürten, L. Rondo, S. Ehrhart, and J. Curtius. Calibration of a chemical ionization mass spectrometer for the measurement of gaseous sulfuric acid. *The Journal of Physical Chemistry A*, 116(24):6375–6386, 2012.

[89] A. Kürten, C. Williamson, J. Almeida, J. Kirkby, and J. Curtius. On the derivation of particle nucleation rates from experimental formation rates. *Atmospheric Chemistry and Physics*, 15:4063–4075, 2015.

[90] H. Lamkaddam, M. Xiao, and PSI. Quantitative driven by both condensation of vapors and coagulation of small molecular clusters. *Environmental Science & Technology*, planned, Sep 2019.

[91] M. J. Lawler, P. M. Winkler, J. Kim, L. Ahlm, J. Tröstl, A. P. Praplan, S. Schobesberger, A. Kürten, J. Kirkby, F. Bianchi, J. Duplissy, A. Hansel, T. Jokinen, H. Keskinen, K. Lehtipalo, M. Leiminger, T. Petäjä, M. Rissanen, L. Rondo, M. Simon, M. Sipilä, C. Williamson, D. Wimmer, I. Riipinen, A. Virtanen, and J. N. Smith. Unexpectedly acidic nanoparticles formed in dimethylamine-ammonia-sulfuric acid nucleation experiments at CLOUD. *Atmospheric Chemistry and Physics*, 16:13601–13618, 2016.

[92] K. Lehtipalo, J. Leppä, J. Kontkanen, J. Kangasluoma, A. Franchin, D. Wimmer, S. Schobesberger, H. Junninen, T. Petäjä, M. Sipilä, J. Mikkilä, J. Vanhanen, D. R. Worsnop, and M. Kulmala. Methods for determining particle size distribution and growth rates between 1 and 3 nm using the particle size magnifier. *Boreal Environment Research*, 19:215–236, 2014.

[93] K. Lehtipalo, L. Rondo, S. Schobesberger, T. Jokinen, N. Sarnela, A. Franchin, T. Nieminen, M. Sipilä, A. Kürten, F. Riccobono, S. Erhart, T. Yli-Juuti, J. Konkanen, A. Adamov, J. Almeida, A. Amorim, F. Bianchi, M. Breitenlechner, J. Dommen, A. J. Downard, E. M. Dunne, J. Duplissy, R. C. Flagan, R. Guida, J. Hakala, A. Hansel, W. Jud, J. Kangasluoma, H. Keskinen, J. Kim, A. Kupc, A. Laaksonen, S. Mathot, I. K. Ortega, A. Onnela, A. Praplan, , M. P. Rissanen, T. Ruuskanen, F. D. Santos, S. Schallhart, R. Schnitzhofer, J. N. Smith, J. Tröstl, G. Tsagkogeorgas, A. Tomé, P. Vaattovaara, A. E. Vrtala, P. E.Wagner, C. Williamson, D. Wimmer, P. M. Winkler, A. Virtanen, N. M. Donahue, K. S. Carslaw, U. Baltensperger, J. Kirkby, I. Riipinen, J. Curtius, M. Kulmala, and D. R. Worsnop. The effect of acid–base clustering and ions on the growth of atmospheric nano-particles. *Nature Communications*, 7:11594, 2016.

[94] K. Lehtipalo, C. Yan, L. Dada, F. Bianchi, M. Xiao, R. Wagner, D. Stolzenburg, L. R. Ahonen, A. Amorim, A. Baccarini, P. S. Bauer, B. Baumgartner, A. Bergen, A.-K. Bernhammer, M. Breitenlechner, S. Brilke, A. Buckholz, S. B. Mazon, D. Chen, X. Chen, A. Dias, J. Dommen, D. C. Draper, J. Duplissy, M. Ehn, H. Finkenzeller, L. Fisher, C. Frege, C. Fuchs, O. Garmash, H. Gordon, J. Hakala, X. C. He, L. Heikkinen, M. Heinrizi, J. C. Helm, V. Hofbauer, C. R. Hoyle, T. Jokinen, J. Kangasluoma, V.-M. Kerminen, C. Kim, J. Kirkby, J. Kontkanen, A. Kürten, M. J. Lawler, H. Mai1, S. Mathot, R. L. Mauldin III, U. Molteni, L. Nichman, W. Nie, T. Nieminen, A. Ojdanic, A. Onnela1, M. Passananti, T. Petäjä, F. Piel, V. Pospisilova,

L. L. J. Quélévér, M. P. Rissanen, C. Rose, N. Sarnela, S. Schallhart, K. Sengupta, M. Simon, C. Tauber, A. Tomé, J. Tröst, O. Väisänen, A. L. Voge, R. Volkamer, A. C. Wagner, M. Wang, L. Weitz, D. Wimmer, P. Ye, A. Ylisirniö, Q. Zha, K. Carslaw, J. Curtius, N. Donahue, R. C. Flagan, A. Hansel, I. Riipinen, A. Virtanen, P. M. Winkler, U. Baltensperger, M. Kulmala, and D. R. Worsnop. Multi-component new particle formation from sulfuric acid, ammonia and biogenic vapors. *Science Advances*, 4:1–9, 2018.

[95] C. Lerot, T. Stavrakou, I. De Smedt, J.-F. Müller, and M. Van Roozendael. Glyoxal vertical columns from GOME-2 backscattered light measurements and comparisons with a global model. *Atmospheric Chemistry and Physics*, 10(24):12059–12072, 2010.

[96] F. D. Lopez-Hilfiker, C. Mohr, M. Ehn, F. Rubach, E. Kleist, J. Wildt, T. F. Mentel, A. Lutz, M. Hallquist, D. Worsnop, and J. A. Thornton. A novel method for online analysis of gas and particle composition: description and evaluation of a filter inlet for gases and aerosols (FIGAERO). *Atmospheric Measurement Techniques*, 7(4):983–1001, 2014.

[97] G. W. Mann, K. S. Carslaw, D. V. Spracklen, D. A. Ridley, P. T. Manktelow, M. P. Chipperfield, S. J. Pickering, and C. E. Johnson. Description and evaluation of GLOMAP-mode: a modal global aerosol microphysics model for the ukca composition-climate model. *Geoscientific Model Development*, 3:519–551, 2010.

[98] H. E. Manninen and UHEL. Comparison of inoization and ion-induced nuclation in cloud with field observations. *Atmospheric Chemistry and Physics*, planned, Aug 2019.

[99] L. Mauldin, M. P. Rissanen, C. Yan, V. Hofbauer, M. Schervish, UHEL, and CMU. HO_x–RO_x chemistry in CLOUD. *Journal of Physical Chemistry A*, in preparation, May 2019.

[100] R. Mauldin, C. Cantrell, M. Zondlo, E. Kosciuch, F. Eisele, G. Chen, D. Davis, R. Weber, J. Crawford, D. Blake, A. Bandy, and D. Thornton. Highlights of OH, H2SO4, and methane sulfonic acid measurements made aboard the NASA P-3B during Transport and Chemical Evolution over the Pacific. *Journal of Geophysical Research Atmospheres*, 108(D20), OCT 29 2003.

[101] J. Merikanto, J. Duplissy, A. Määttänen, H. Henschel, N. M. Donahue, D. Brus, S. Schobesberger, M. Kulmala, and H. Vehkamäki. Effect of ions on sulfuric acid-water binary particle formation I: Theory for kinetic and nucleation-type particle formation and atmospheric implications. *Journal of Geophysical Research Atmospheres*, 121:1736–1751, 2016.

[102] N. Meskhidze and A. Nenes. Phytoplankton and cloudiness in the Southern Ocean. *Science*, 314(5804):1419–23, 2006.

[103] U. Molteni and PSI. α-Pinene ozonolysis mechanism and HOM formation via auto-oxidation. *Earth and Space Chemistry*, submitted, Jan 2019.

[104] U. Molteni and PSI. Ion-mobility spectroscopy coupled to nitrate chemical ionization mass scpectrometry in the CERN CLOUD chamber. *Aerosol Measurement Techniques*, planned, Jul 2019.

[105] L. Nichman, C. Fuchs, E. Järvinen, K. Ignatius, N. F. Höppel, A. Dias, M. Heinritzi, M. Simon, J. Tröstl, A. C. Wagner, R. Wagner, C. Williamson, C. Yan, F. Bianchi, P. J. Connolly, J. R. Dorsey, J. Duplissy, S. Ehrhart, C. Frege, H. Gordon, C. R. Hoyle, T. B. Kristensen, G. Steiner, N. M. Donahue, R. Flagan, M. W. Gallagher, J. Kirkby, O. Möhler, H. Saathoff, M. Schnaiter, F. Stratmann, and A. Tomé. Discrimination of water, ice and aerosols by light polarisation in the CLOUD experiment. *Atmospheric Chemistry and Physics*, 16:3651–3664, 2016.

[106] L. Nichman, E. Järvinen, J. Dorsey, P. Connolly, J. Dupplisy, C. Fuchs, K. Ignatius, K. Sengupta, F. Stratmann, O. Möhler, M. Schnaiter, and M. Gallagher. Intercomparison study and optical asphericity measurements of small ice particles in the CERN CLOUD experiment. *Atmospheric Measurement Techniques*, 10:3231–3248, 2017.

[107] C. D. O'Dowd, M. C. Facchini, F. Cavalli, D. Ceburnis, M. Mircea, S. Decesari, S. Fuzzi, Y. J. Yoon, and J.-P. Putaud. Biogenically driven organic contribution to marine aerosol. *Nature*, 431(7009):676–680, 2004.

[108] C. D. O'Dowd, J. L. Jimenez, R. Bahreini, R. C. Flagan, J. H. Seinfeld, K. Hameri, L. Pirjola, M. Kulmala, S. G. Jennings, and T. Hoffmann. Marine aerosol formation from biogenic iodine emissions. *Quarterly Journal of the Royal Meteorological Society*, 417(6889):632–636, 2002.

[109] I. K. Ortega, N. M. Donahue, T. Kurtén, M. Kulmala, C. Focsa, and H. Vehkamäki. Can highly oxidized organics contribute to atmospheric new-particle formation? *Journal of Physical Chemistry A*, 120:1452–1458, 2016.

[110] J. Pfeifer and CERN. Design and performance of a charged CCN generator. *Aerosol Measurement Techniques*, planned, Sep 2019.

[111] J. Pfeifer, T. Nieminen, A. Kürten, M. Simon, M. Heinritzi, CLOUD, J. Curtius, and J. Kirkby. Molecular understanding of multicomponent nucleation involving ammonia. *Proceedings of the National Academy of Sciences*, planned, Aug 2019.

[112] J. Pfeifer and UFRA. An ion source for highly sensitive measurements of ammonia, amines and HIO_3. *Aerosol Measurement Techniques*, in preparation, Feb 2019.

[113] J. R. Pierce. Cosmic rays, aerosols, clouds, and climate: Recent findings from the CLOUD experiment. *Journal of Geophysical Research: Atmospheres*, 502(5599):359–5, Aug. 2017.

[114] J. R. Pierce, I. Riipinen, M. Kulmala, M. Ehn, T. Petäjä, H. Junninen, D. R. Worsnop, and N. M. Donahue. Quantification of the volatility of secondary organic compounds in ultrafine particles during nucleation events. *Atmospheric Chemistry and Physics*, 11:9019–9036, 2011.

[115] R. W. Pinder, P. J. Adams, and S. N. Pandis. Ammonia emission controls as a cost-effective strategy for reducing atmospheric particulate matter in the eastern United States. *Environmental Science & Technology*, 41(2):380–386, JAN 15 2007.

[116] C. A. Pope, M. Ezzati, and D. W. Dockery. Fine-particulate air pollution and life expectancy in the United States. *The New England Journal of Medicine*, 360(4):376–386, Jan. 2009.

[117] V. Pospisilova, F. D. Lopez-Hilfiker, and PSI. EESI particle composition and instrumentation description. *Atmospheric Chemistry and Physics*, in preparation, Mar 2019.

[118] A. P. Praplan, F. Bianchi, J. Dommen, and U. Baltensperger. Dimethylamine and ammonia measurements with ion chromatography during the CLOUD4 campaign. *Atmospheric Measurement Techniques*, 5(9):2161–2167, 2012.

[119] A. P. Praplan, S. Schobesberger, F. Bianchi, M. P. Rissanen, M. Ehn, T. Jokinen, H. Junninen, A. Adamov, A. Amorim, J. Dommen, J. Duplissy, J. Hakala, A. Hansel, M. Heinritzi, J. Kangasluoma, J. Kirkby, M. Krapf, A. Kürten, K. Lehtipalo, F. Riccobono, L. Rondo, N. Sarnela, M. Simon, A. Tomé, J. Tröstl, P. M. Winkler, C. Williamson, P. Ye, J. Curtius, U. Baltensperger, N. M. Donahue, M. Kulmala, and D. R. Worsnop. Elemental composition and clustering behaviour of α-pinene oxidation products for different oxidation conditions. *Atmospheric Chemistry and Physics*, 15:4145–4159, 2015.

[120] A. A. Presto, K. E. Huff Hartz, and N. M. Donahue. Secondary organic aerosol production from terpene ozonolysis: 1. Effect of UV radiation. *Environmental Science & Technology*, 39:7036–7045, 2005.

[121] P. K. Quinn and T. S. Bates. The case against climate regulation via oceanic phytoplankton sulphur emissions. *Nature*, 480(7375):51–56, 2011.

[122] P. K. Quinn, T. S. Bates, K. S. Schulz, D. J. Coffman, A. A. Frossard, L. M. Russell, W. C. Keene, and D. J. Kieber. Contribution of sea surface carbon pool to organic matter enrichment in sea spray aerosol. *Nature Geoscience*, 7(3):228–232, 2014.

[123] F. Riccobono, S. Schobesberger, C. E. Scott, J. Dommen, I. K. Ortega, L. Rondo, J. Almeida, A. Amorim, F. Bianchi, M. Breitenlechner, A. David, A. Downard, E. Dunne, J. Duplissy, S. Ehrhart, R. C. Flagan, A. Franchin, A. Hansel, H. Junninen, M. Kajos, H. Keskinen, A. Kupc, O. Kupiainen, A. Kürten, T. Kurtén, A. N. Kvashin, A. Laaksonen, K. Lehtipalo, V. Makhmutov, S. Mathot, T. Nieminen, T. Olenius, A. Onnela, T. Petäjä, A. P. Praplan, F. D. Santos, S. Schallhart, J. H. Seinfeld, M. Sipilä, D. V. Spracklen, Y. Stozhkov,

F. Stratmann, A. Tomé, G. Tsagkogeorgas, P. Vaattovaara, H. Vehkamäki, Y. Viisanen, A. Vrtala, P. E. Wagner, E. Weingartner, H. Wex, D. Wimmer, K. S. Carslaw, J. Curtius, N. M. Donahue, J. Kirkby, M. Kulmala, D. R. Worsnop, and U. Baltensperger. Oxidation products of biogenic emissions contribute to nucleation of atmospheric particles. *Science*, 344:717–721, 2014.

[124] I. Riipinen, J. R. Pierce, T. Yli-Juuti, T. Nieminen, S. Häkkinen, M. Ehn, H. Junninen, K. Lehtipalo, T. Petäjä, J. Slowik, R. Chang, N. C. Shantz, J. P. D. Abbatt, W. R. Leaitch, V.-M. Kerminen, D. R. Worsnop, S. N. Pandis, N. M. Donahue, and M. Kulmala. Organic condensation: a vital link connecting aerosol formation to cloud condensation nuclei (CCN) concentrations. *Atmospheric Chemistry and Physics*, 11:3865–3878, 2011.

[125] I. Riipinen, T. Yli-Juuti, J. R. Pierce, T. Petäjä, D. R. Worsnop, M. Kulmala, and N. M. Donahue. Contribution of organics to atmospheric nanoparticle growth. *Nature Geoscience*, 5:453–458, 2012.

[126] M. Rissanen, C. Yan, L. Mauldin, V. Hofbauer, N. M. Donahue, and UHEL. Peroxy radical chemistry of monoterpenes during CLOUD organic oxidation campaigns. *Physical Chemistry Chemical Physics*, in preparation, Jun 2019.

[127] M. P. Rissanen, T. Kurtén, M. Sipilä, J. A. Thornton, J. Kangasluoma, N. Sarnela, H. Junninen, S. Jørgensen, S. Schallhart, M. K. Kajos, R. Taipale, M. Springer, T. F. Mentel, T. Ruuskanen, T. Petäjä, D. R. Worsnop, H. G. Kjaergaard, and M. Ehn. The formation of highly oxidized multifunctional products in the ozonolysis of cyclohexene. *Journal of the American Chemical Society*, 136(44):15596–15606, 2014. PMID: 25283472.

[128] L. Rondo, S. Ehrhart, A. Kürten, A. Adamov, F. Bianchi, M. Breitenlechner, J. Duplissy, A. Franchin, J. Dommen, N. M. Donahue, E. M. Dunne, R. C. Flagan, J. Hakala, A. Hansel, H. Keskinen, J. Kim, T. Jokinen, K. Lehtipalo, M. Leiminger, A. Praplan, F. Riccobono, M. P. Rissanen, N. Sarnela, S. Schobesberger, M. Simon, M. Sipilä, J. N. Smith, A. Tomé, J. Tröstl, G. Tsagkogeorgas, P. Vaattovaara, P. M. Winkler, C. Williamson, D. Wimmer, U. Baltensperger, J. Kirkby, M. Kulmala, T. Petäjä, D. R. Worsnop, and J. Curtius. Effect of dimethylamine on the gas phase sulfuric acid concentration measured by chemical ionization mass spectrometry (CIMS). *Journal of Geophysical Research Atmospheres*, 121:3036–3049, 2016.

[129] L. Rondo, A. Kürten, S. Ehrhart, S. Schobesberger, A. Franchin, H. Junninen, T. Petäjä, M. Sipilä, D. R. Worsnop, and J. Curtius. Effect of ions on the measurement of sulphuric acid in the CLOUD experiment at CERN. *Atmospheric Measurement Techniques*, 7(7):3849–3859, 2014.

[130] S. Rossignol, L. Tinel, A. Bianco, M. Passananti, M. Brigante, D. J. Donaldson, C. George, U. Lyon, U. Claude, and B. Lyon. Atmospheric photochemistry at a fatty acid–coated air-water interface. *Science*, 353(6300):699–702, 2016.

[131] M. Sahyoun, E. Freney, J. Brito, J. Duplissy, A. Colomb, R. Dupuy, T. Bourianne, C. Yan, T. Petäjä, M. Kulmala, A. Schwarzenboeck, C. Planche, and K. Sellegri. Evidence of new particle formation within volcanic passive plumes and its parameterization from airborne in-situ measurements. *Journal of Geophysical Research, Atmospheres*, submitted, 2019.

[132] N. Sarnela, T. Jokinen, J. Duplissy, C. Yan, T. Nieminen, M. Ehn, S. Schobesberger, M. Heinritzi, S. Ehrhart, K. Lehtipalo, J. Tröstl, M. Simon, A. Kürten, M. Leiminger, M. J. Lawler, M. P. Rissanen, F. Bianchi, A. P. Praplan, J. Hakala, A. Amorim, M. Gonin, A. Hansel, J. Kirkby, J. Dommen, J. Curtius, J. Smith, T. Petäjä, D. R. Worsnop, M. Kulmala, N. M. Donahue, and M. Sipilä. Measurement-model comparison of stabilized Criegee intermediate and highly oxygenated molecule production in the CLOUD chamber. *Atmospheric Chemistry and Physics*, 18:2363–2380, 2018.

[133] M. Schervish, V. Hofbauer, M. Wang, D. Chen, P. Ye, CLOUD, and N. M. Donahue. VBS model of multiple chemical systems in CLOUD. *Atmospheric Chemistry and Physics*, in preparation, Mar 2019.

[134] R. Schnitzhofer, A. Metzger, M. Breitenlechner, W. Jud, M. Heinritzi, L.-P. D. Menezes, J. Duplissy, R. Guida, S. Haider, J. Kirkby, S. Mathot, P. Minginette, A. Onnela, H. Walther, A. Wasem, A. Hansel, and T. CLOUD. Characterisation of organic contaminants in the CLOUD chamber at CERN. *Aerosol Measurement Techniques*, 7:2159–2168, 2014.

[135] S. Schobesberger, A. Franchin, F. Bianchi, L. Rondo, J. Duplissy, A. Kürten, I. K. Ortega, A. Metzger, R. Schnitzhofer, J. Almeida, A. Amorim, J. Dommen, E. M. Dunne, M. Ehn, S. Gagné, L. Ickes, H. Junninen, A. Hansel, V.-M. Kerminen, J. Kirkby, A. Kupc, A. Laaksonen, K. Lehtipalo, S. Mathot, A. Onnela, T. Petäjä, F. Riccobono, F. D. Santos, M. Sipilä, A. Tomé, G. Tsagkogeorgas, Y. Viisanen, P. E. Wagner, D. Wimmer, J. Curtius, N. M. Donahue, U. Baltensperger, M. Kulmala, and D. R. Worsnop. On the composition of ammonia–sulfuric-acid ion clusters during aerosol particle formation. *Atmospheric Chemistry and Physics*, 15:55–78, 2015.

[136] S. Schobesberger, H. Junninen, F. Bianchi, G. Lönn, M. Ehn, K. Lehtipalo, J. Dommen, S. Ehrhart, I. K. Ortega, A. Franchin, T. Nieminen, F. Riccobono, M. Hutterli, J. Duplissy, J. Almeida, A. Amorim, M. Breitenlechner, A. J. Downard, E. M. Dunne, R. C. Flagan, M. Kajos, H. Keskinen, J. Kirkby, A. Kupc, A. Kürten, T. Kurtén, A. Laaksonen, S. Mathot, A. Onnela, A. P. P. L. Rondo, F. D. Santos, S. Schallhart, R. Schnitzhofer, M. Sipilä, A. Tomé, G. Tsagkogeorgas, H. Vehkamäki, D. Wimmer, U. Baltensperger, K. S. Carslaw, J. Curtius, A. Hansel, T. Petäjä, M. Kulmala, N. M. Donahue, and D. R. Worsnop. Molecular understanding of atmospheric particle formation from sulfuric acid and large oxidized organic molecules. *Proceedings of the National Academy of Sciences*, 110:17223–17228, 2013.

[137] K. Sengupta, Leeds, and CLOUD. Investigating the impact of biogenic organics on CCN. *Atmospheric Chemistry and Physics*, planned, Jun 2019.

[138] M. Simon, M. Heinritzi, S. Herzog, M. Leiminger, F. Bianchi, A. Praplan, J. Dommen, J. Curtius, and A. Kürten. Detection of dimethylamine in the low pptv range using nitrate chemical ionization-atmospheric pressure interface-time of flight (CI-APi-TOF) mass spectrometry. *Atmospheric Measurement Techniques*, 9:2135–2145, 2016.

[139] M. Simon, Q. Ye, CMU, and UFRA. Photochemistry of different systems in CLOUD. *Atmospheric Chemistry and Physics*, in preparation, Mar 2019.

[140] M. Sipilä, T. Berndt, T. Petäjä, D. Brus, J. Vanhanen, F. Stratmann, J. Patokoski, R. L. Mauldin, A.-P. Hyvärinen, H. Lihavainen, and M. Kulmala. The role of sulfuric acid in atmospheric nucleation. *Science*, 327(5970):1243–1246, 2010.

[141] M. Sipilä, N. Sarnela, T. Jokinen, H. Henschel, H. Junninen, J. Kontkanen, S. Richters, J. Kangasluoma, A. Franchin, O. Peräkylä, M. P. Rissanen, M. Ehn, H. Vehkamäki, T. Kurten, T. Berndt, T. Petäjä, D. Worsnop, D. Ceburnis, V.-M. Kerminen, M. Kulmala, and C. O'Dowd. Molecular-scale evidence of aerosol particle formation via sequential addition of HIO3. *Nature*, 537(7621):532–534, 2016.

[142] C. O. Stanier, A. Y. Khlystov, and S. N. Pandis. Nucleation events during the Pittsburgh Air Quality Study: Description and relation to key meteorological, gas phase, and aerosol parameters. *Aerosol Science and Technology*, 38:253–264, 2004.

[143] D. Stolzenburg, L. Fischer, A. L. Vogel, M. Heinritzi, M. Schervish, M. Simon, A. C. Wagner, L. Dada, L. R. Ahonen, A. Amorim, A. Baccarini, P. S. Bauer, B. Baumgartner, A. Bergen, F. Bianchi, M. Breitenlechner, S. Brilke, S. Buenrostro Mazon, D. Chen, A. Dias, D. C. Draper, J. Duplissy, I. El Haddad, H. Finkenzeller, C. Frege, C. Fuchs, O. Garmash, H. Gordon, X. He, J. Helm, V. Hofbauer, C. R. Hoyle, C. Kim, J. Kirkby, J. Kontkanen, A. Kürten, J. Lampilahti, M. Lawler, K. Lehtipalo, M. Leiminger, H. Mai, S. Mathot, B. Mentler, U. Molteni, W. Nie, T. Nieminen, J. B. Nowak, A. Ojdanic, A. Onnela, M. Passananti, T. Petäjä, L. L. J. Quéléver, M. P. Rissanen, N. Sarnela, S. Schallhart, C. Tauber, A. Tomé, R. Wagner, M. Wang, L. Weitz, D. Wimmer, M. Xiao, C. Yan, P. Ye, Q. Zha, U. Baltensperger, J. Curtius, J. Dommen, R. C. Flagan, M. Kulmala, J. N. Smith, D. R. Worsnop, A. Hansel, N. M. Donahue, and P. M. Winkler. Rapid growth of organic aerosol nanoparticles over a wide tropospheric temperature range. *Proceedings of the National Academy of Sciences*, 115:9122–9127, 2018.

[144] D. Stolzenburg, G. Steiner, and P. M. Winkler. A DMA-train for precision measurement of sub-10 nm aerosol dynamics. *Atmospheric Measurement Techniques*, 10(4):1639–1651, 2017.

[145] D. Stolzenburg and UVIE. The role of ammonia in stabilizing sulfuric acid growth. *Atmospheric Chemistry and Physics*, planned, Jul 2019.

[146] J. Tröstl, W. K. Chuang, M. Heinritzi, C. Yan, U. Molteni, L. Ahlm, C. Frege, F. Bianchi, R. Wagner, M. Simon, K. Lehtipalo, C. Williamson, J. S. Craven, J. Duplissy, A. Adamov, J. Almeida, A.-K. Bernhammer, M. Breitenlechner, S. Brilke, A. Dias, S. Ehrhart, R. C. Flagan, A. Franchin, C. Fuchs, H. Gordon, R. Guida, M. Gysel, A. Hansel, C. R. Hoyle, T. Jokinen, H. Junninen, J. Kangasluoma, H. Keskinen, J. Kim, M. Krapf, A. Kürten, A. Laaksonen, M. Lawler, M. Leiminger, S. Mathot, O. Möhler, T. Nieminen, A. Onnela, T. Petäjä, F. M. Piel, P. Miettinen, M. P. Rissanen, L. Rondo, N. Sarnela, S. Schobesberger, K. Sengupta, M. Sipilä, J. N. Smith, G. Steiner, A. Tomè, A. Virtanen, A. C. Wagner, E. Weingartner, D. Wimmer, P. M. Winkler, P. Ye, K. S. Carslaw, J. Curtius, J. Dommen, J. Kirkby, M. Kulmala, I. Riipinen, D. R. Worsnop, N. M. Donahue, and U. Baltensperger. The role of low-volatility organic compounds in initial particle growth in the atmosphere. *Nature*, 530:527–531, 2016.

[147] E. R. Trump and N. M. Donahue. Oligomer formation within secondary organic aerosol: Equilibrium and dynamic considerations. *Atmospheric Chemistry and Physics*, 14:3691–3701, 2014.

[148] E. R. Trump, S. A. Epstein, I. Riipinen, and N. M. Donahue. Wall effects in smog chamber experiments: A model study. *Aerosol Science and Technology*, 50(11):1180–1200, 2016.

[149] E. R. Trump, I. Riipinen, and N. M. Donahue. Modeling the condensational growth of nanoparticles in the atmosphere. *Boreal Environment Research*, 19:352–362, 2014.

[150] G. Tsagkogeorgas, P. Roldin, J. Duplissy, L. Rondo, J. Tröstl, J. G. Slowik, S. Ehrhart, A. Franchin, A. Kürten, A. Amorim, F. Bianchi, J. Kirkby, T. Petäjä, U. Baltensperger, M. Boy, J. Curtius, R. C. Flagan, M. Kulmala, N. M. Donahue, and F. Stratmann. Evaporation of sulphate aerosols at low relative humidity. *Atmospheric Chemistry and Physics*, 17:8923–8938, 2017.

[151] O. Väisänen, UEF, and CLOUD. Hydroscopicity of iodide aerosol. *Aerosol Science and Technology*, planned, Jun 2019.

[152] A. L. Vogel, M. Wang, V. Hofbauer, PSI, and CLOUD. Production of nitroxy-organosulfates via reactive uptake of SO_2 to organic particles. *Nature Chemsitry*, in preparation, Mar 2019.

[153] J. Voigtländer, J. Duplissy, L. Rondo, A. Kürten, and F. Stratmann. Numerical simulations of mixing conditions and aerosol dynamics in the CERN CLOUD chamber. *Atmospheric Chemistry and Physics*, 12(4):2205–2214, 2012.

[154] R. Volkamer, S. Baidar, T. L. Campos, S. Coburn, J. P. DiGangi, B. Dix, E. W. Eloranta, T. K. Koenig, B. Morley, I. Ortega, B. R. Pierce, M. Reeves, R. Sinreich, S. Wang, M. a. Zondlo, and P. a. Romashkin. Aircraft measurements of BrO, IO, glyoxal, NO2, H2O, O2–O2 and aerosol extinction profiles in the tropics: comparison with aircraft-/ship-based in situ and lidar measurements. *Atmospheric Measurement Techniques*, 8(5):2121–2148, 2015.

[155] A. C. Wagner, A. Bergen, S. Brilke, C. Fuchs, M. Ernst, J. Hoker, M. Heinritzi, M. Simon, B. Bühner, J. Curtius, and A. Kürten. Size resolved online chemical analysis of nano aerosol particles: A thermal desorption differential mobility analyzer coupled to a chemical ionization time of flight mass spectrometer. *Atmospheric Measurement Techniques*, 18:5489–5506, 2018.

[156] A. C. Wagner, UFRA, and CLOUD. Should I stay or should I go? Gas to particle transitions of HOMs from anthropogenic VOCs. *Atmospheric Chemistry and Physics*, planned, Jun 2019.

[157] R. Wagner, C. Yan, K. Lehtipalo, J. Duplissy, T. Nieminen, J. Kangasluoma, L. R. Ahonen, L. Dada, J. Kontkanen, H. E. Manninen, A. Dias, A. Amorim, P. S. Bauer, A. Bergen, A.-K. Bernhammer, F. Bianchi, S. Brilke, S. B. Mazon, X. Chen, D. C. Draper, L. Fischer, C. Frege, C. Fuchs, O. Garmash, H. Gordon, J. Hakala, L. Heikkinen, M. Heinritzi, V. Hofbauer, C. R. Hoyle, J. Kirkby, A. Kürten, A. N. Kvashnin, T. Laurila, M. J. Lawler, H. Mai, V. Makhmutov, R. L. Mauldin III, U. Molteni, L. Nichman, W. Nie, A. Ojdanic, A. Onnela, F. Piel, L. L. J. Quéléver, M. P. Rissanen, N. Sarnela, S. Schallhart, K. Sengupta, M. Simon, D. Stolzenburg, Y. Stozhkov, J. Tröstl, Y. Viisanen, A. L. Vogel, A. C. Wagner, M. Xiao, P. Ye, U. Baltensperger, J. Curtius, N. M. Donahue, R. C. Flagan, M. Gallagher, A. Hansel, J. N. Smith, A. Tomé, P. M. Winkler, D. Worsnop, M. Ehn, M. Sipilä, V.-M. Kerminen, T. Petäjä, and M. Kulmala. The role of ions in new-particle formation in the cloud chamber. *Atmospheric Chemistry and Physics*, 17:15181–15197, 2017.

[158] D. Wang, PSI, and CLOUD. Calculation of α-pinene kinetics at different temperatures. *Atmospheric Chemistry and Physics*, planned, May 2019.

[159] L. Wang, A. F. Khalizov, J. Zheng, W. Xu, Y. Ma, V. Lal, and R. Zhang. Atmospheric nanoparticles formed from heterogeneous reactions of organics. *Nature Geoscience*, 3(4):238–242, 2010.

[160] M. Wang, CLOUD, CMU, and N. M. Donahue. Nitric acid and ammonia particle formation. *Nature*, in preparation, Jun 2019.

[161] M. Wang and CMU. The relative contribution of reactive uptake and condensation of extremely low volatility molecules to growth of biogenic and anthropogenic organic particles. *Atmospheric Chemistry and Physics*, in preparation, Apr 2019.

[162] M. Wang, M. Xiao, PSI, and CMU. Aging versus auto-oxidation in anthropogenic and biogenic organic new-particle formation. *Atmospheric Chemistry and Physics*, planned, Feb 2019.

[163] S. Weber and CERN. CLOUD data acquision system. *Aerosol Measurement Techniques*, planned, Aug 2019.

[164] D. M. Westervelt, J. R. Pierce, I. Riipinen, W. Trivitayanurak, A. Hamed, M. Kulmala, A. Laaksonen, S. Decesari, and P. J. Adams. Formation and growth of nucleated particles into cloud condensation nuclei: model–measurement comparison. *Atmospheric Chemistry and Physics*, 13(15):7645–7663, 2013.

[165] D. Wimmer, K. Lehtipalo, A. Franchin, J. Kangasluoma, F. Kreissl, A. Kürten, A. Kupc, A. Metzger, J. Mikkilä, T. Petäjä, F. Riccobono, J. Vanhanen, M. Kulmala, and J. Curtius. Performance of diethylene glycol-based particle counters in the sub-3 nm size range. *Atmospheric Measurement Techniques*, 6(7):1793–1804, 2013.

[166] D. Wimmer, K. Lehtipalo, T. Nieminen, J. Duplissy, S. Ehrhart, J. Almeida, L. Rondo, A. Franchin, F. Kreissl, F. Bianchi, H. E. Manninen, M. Kulmala, J. Curtius, and T. Petäjä. Technical note: Using DEG CPCs at upper tropospheric temperatures. *Atmospheric Chemistry and Physics*, 15:7547–7555, 2015.

[167] F. Wittrock, A. Richter, H. Oetjen, J. P. Burrows, M. Kanakidou, S. Myriokefalitakis, R. Volkamer, S. Beirle, U. Platt, and T. Wagner. Simultaneous global observations of glyoxal and formaldehyde from space. *Geophysical Research Letters*, 33(16):L16804, aug 2006.

[168] B. E. Wyslouzil, J. H. Seinfeld, R. C. Flagan, and K. Okuyama. Binary nucleation in acid–water systems. ii. sulfuric acid–water and a comparison with methanesulfonic acid–water. *The Journal of Chemical Physics*, 94(10):6842–6850, 1991.

[169] M. Xiao, PSI, CLOUD, and C. Hoyle. Particle formation and growth for urban conditions. *Nature*, in preparation, Mar 2019.

[170] M. Xiao, M. Wang, B. Baumgartner, L. Dada, D. Stolzenburg, I. E. Haddad, and PSI. Production of highly oxygenated molecules during oxidation of aromatic compounds typical of polluted urban environments. *Atmospheric Chemistry and Physics*, planned, Jun 2019.

[171] C. Yan, W. Nie, A. L. Vogel, L. Dada, K. Lehtipalo, D. Stolzenburg, R. Wagner, M. P. Rissanen, M. Xiao, L. Ahonen, L. Fischer, R. Clémence, F. Bianchi, H. Gordon, O. Garmash, A. Dias, M. Simon, M. Heinritzi, P. Ye, V. Hofbauer, A. Amorim, P. S. Bauer, A. Bergen, A.-K. Bernhammer, M. Breitenlechner, S. Brilke, A. Buchholz, S. B. Mazon, M. R. Canagaratna, X. Chen, A. Ding, J. Dommen, D. C. Draper, J. Duplissy, C. Frege, C. Fuchs, R. Guida, J. Hakala, L. Heikkinen, C. R. Hoyle, T. Jokinen, J. Kangasluoma, J. Kirkby, J. Kontkanen, A. Kürten, M. J. Lawler, H. Mai, S. Mathot, R. L. M. III, P. Miettinen, U. Molteni, L. Nichman, T. Nieminen, J. Nowak, A. Ojdanic, A. Onnela, A. Pajunoja, T. Petäjä, F. Piel, L. L. J. Quéléver, N. Sarnela, S. Schallhart, K. Sengupta, M. Sipilä, J. N. Smith, A. Tomé, J. Tröstl, O. Väisänen, A. C. Wagner, A. Ylisirniö, Q. Zha, U. Baltensperger, K. S. Carslaw, J. Curtius, R. C. Flagan, A. Hansel, I. Riipinen, A. Virtanen, P. M. Winkler, N. M. Donahue, M. Ehn, V.-M. Kerminen, M. Kulmala, and D. R. Worsnop. How NO_x suppresses the formation of atmospheric biogenic particles. *Science Advances*, submitted, 2019.

[172] C. Yan, R. Yin, Y. Lu, L. Dada, Y. Fu, J. Kontkanen, C. Deng, D. Yang, R. Baalbaki, K. Dallenbach, M. Schervish, R. Cai, M. Bloss, T. Chan, T. Chen, X. Chen, Y. Chen, B. Chu, B. Foreback, O. Garmash, X. He, L. Heikkinen, T. Jokinen, H. Junninen, J. Kangasluoma, T. Kokkonen, M. Kurppa, K. Lehtipalo, H. Li, H. Li, X. Li, Y. Liu, Q. Ma, P. Paasonen, R. Pileci, P. Rantala, A. Rusanen, N. Sarnela, P. Simonen, W. Wang, Y. Wang, M. Xue, G. Yang, L. Yao, Y. Zhou, J. Kujansuu, T. Petäjä, M. Ge, H. He, N. Donahue, D. Worsnop, V.-M. Kerminen, Y. Ma, J. Zheng, L. Wang, Y. Liu, J. Jiang, and M. Kulmala. The synergistic role of sulfuric acid, dimethylamine and oxidized organics governing new-particle formation in Beijing. *Nature*, submitted, 2019.

[173] L. Yao, O. Garmash, F. Bianchi, J. Zheng, C. Yan, J. Kontkanen, H. Junninen, S. B. Mazon, M. Ehn, P. Paasonen, M. Sipilä, M. Wang, X. Wang, S. Xiao, H. Chen, Y. Lu, B. Zhang, D. Wang, Q. Fu, F. Geng, L. Li, H. Wang, L. Qiao, X. Yang, J. Chen, V.-M. Kerminen, T. Petäjä, D. R. Worsnop, M. Kulmala, and L. Wang. Atmospheric new particle formation from sulfuric acid and amines in a chinese megacity. *Science*, 361(6399):278–281, 2018.

[174] Q. Ye, M. Wang, D. Stolzenburg, V. Hofbauer, , D. Chen, M. Schervish, A. L. Vogel, CMU, and CLOUD. Temperature-dependent composition of particles formed via α-pinene ozonolysis measured with a FIGAERO-CIMS. *Journal of the American Chemical Society*, in preparation, Jan 2019.

[175] J. Zhang, K. E. Huff Hartz, S. N. Pandis, and N. M. Donahue. Secondary organic aerosol formation from limonene ozonolysis: Homogeneous and heterogeneous influences as a function of NO_x. *Journal of Physical Chemistry A*, 110:11053–11063, 2006.

[176] R. Zhang, I. Suh, J. Zhao, D. Zhang, E. Fortner, X. Tie, L. Molina, and M. Molina. Atmospheric new particle formation enhanced by organic acids. *Science*, 304(5676):1487–1490, JUN 4 2004.

[177] R. Zhang, L. Wang, A. F. Khalizov, J. Zhao, J. Zheng, R. L. McGraw, and L. T. Molina. Formation of nanoparticles of blue haze enhanced by anthropogenic pollution. *Proceedings of the National Academy of Sciences of the United States of America*, 106(42):17650–17654, Oct. 2009.