



ADDENDUM TO THE CLOUD PROPOSAL

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1 Introduction

1.1 Overview

Following the request of the CERN SPSC at its meeting on 23 May 2000, this report provides further details on the experimental programme and beam schedule of CLOUD. Specifically, we describe (in §4 and §5) two examples of the type of experiments to be performed by CLOUD, and their expected precisions. The experimental and theoretical motivation for these experiments is summarised in §3. The beam requirements and schedule are elaborated in §7. We also summarise the progress on the detector design since the proposal [1] in §2, and the expected scope of the aerosol and cloud simulations for the experiment in §6.

1.2 Choice of experiments

The ions and radicals produced in the atmosphere by galactic cosmic rays (GCRs) may influence cloud microphysics in several ways:

1. Nucleation (creation) of new condensation nuclei (CN) from trace condensable vapours.
2. Growth of CN into cloud condensation nuclei (CCN), which seed cloud droplets.
3. Activation of CCN into cloud droplets in the presence of supersaturated water vapour.
4. Creation of ice nuclei, which cause the freezing of supercooled liquid droplets in clouds.

If the rate or efficiency of any of these processes is significantly modified by ions in the atmosphere, then the GCR intensity could modify the droplet number concentrations of clouds and thereby affect their lifetimes and radiative properties. Motivated by the experimental and theoretical considerations summarised in §3, we have selected for this report one example experiment from each of the first two processes listed above, specifically:

Exp.1: Binary homogeneous nucleation of ultrafine condensation nuclei (UCN) of sulphuric acid and water, under atmospheric conditions (§4).

Exp.2: Growth of these UCN into CCN in the presence of additional vapours: sulphuric acid, water, ammonia and low volatility organic compounds (LVOCs), again under atmospheric conditions (§5).

These examples are part of the category I experiments (“Creation & growth of aerosols”) listed in Table 3 (p.29) of the CLOUD proposal.

2 Progress on the detector design

2.1 Summary

Since the proposal we have improved the detector design by replacing the buffer expansion tank and flow chamber (described in §4 of the proposal) by a single device, a *reactor chamber* (Figs. 1 and 2).

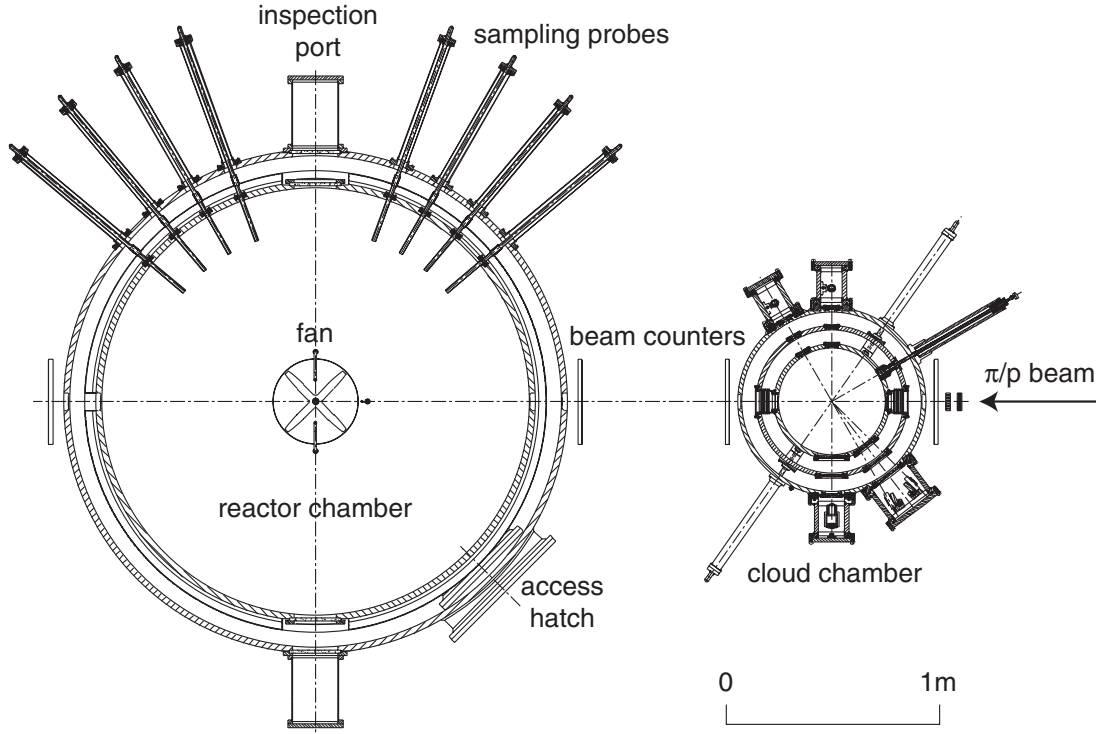


Figure 1: Horizontal section of the cloud chamber and the new reactor chamber.

The reactor chamber will be located in the beamline, immediately behind the cloud chamber. It performs the same functions as the devices it replaces, but with improved performance, namely:

1. Buffer tank for small expansions. This functions in the same way as described previously in the proposal, providing a second method to produce small expansions.
2. A reactor chamber for providing the source of reacted gas/aerosols for the external detectors. This replaces the function of the original flow chamber but with the improvement that it is now effective for long growth-time experiments extending for several days, since diffusion losses to the walls are strongly reduced. Since the aerosol and trace gas lifetimes from wall losses should scale as the linear dimension of the vessel (i.e. the ratio of its volume to surface area), we expect the reactor chamber will have about a factor four longer particle lifetime than the cloud chamber. The cloud chamber is suitable for growth experiments lasting up to about a day. For longer growth-time experiments the reactor chamber can now provide samples of

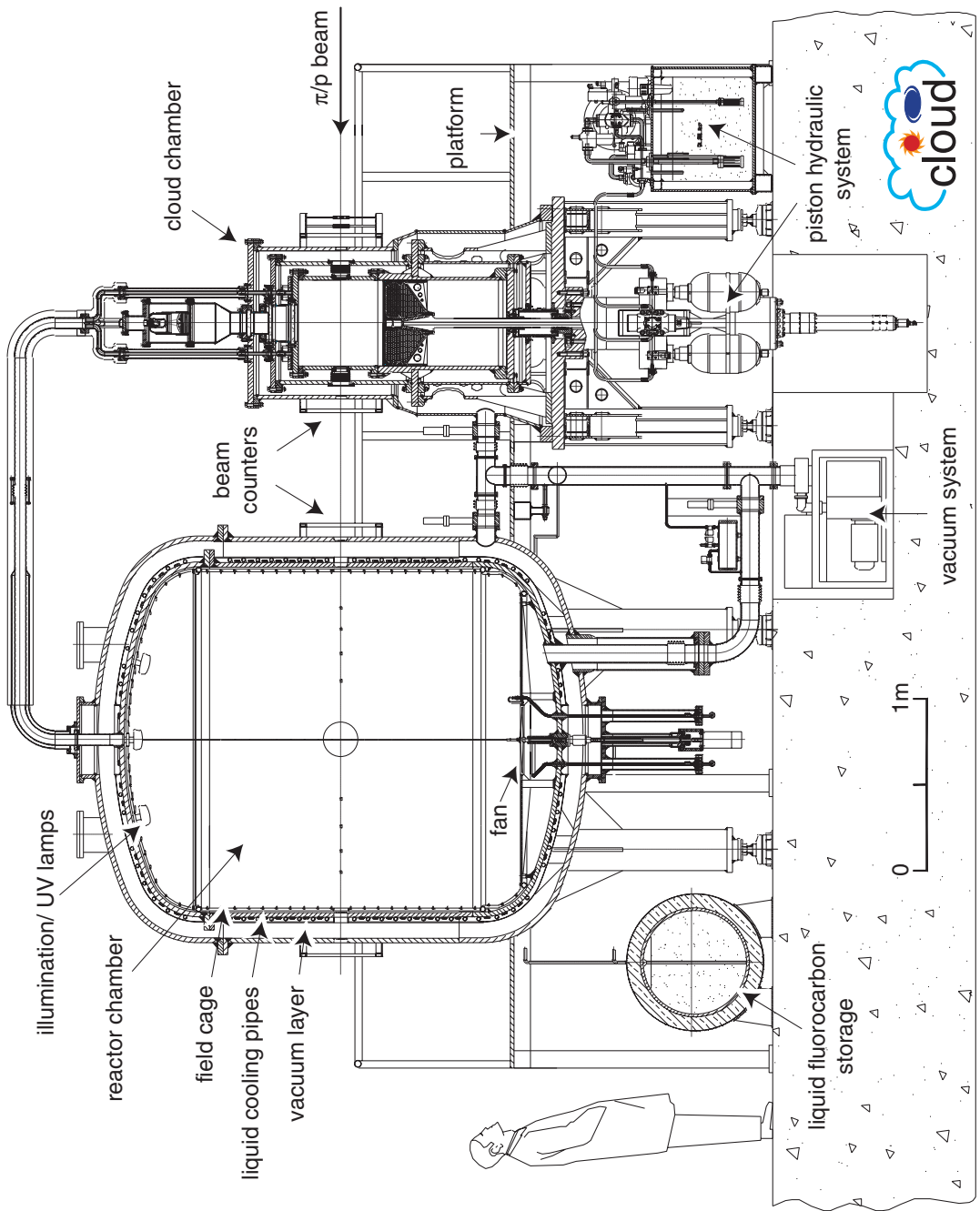


Figure 2: Vertical section showing the cloud chamber and the new reactor chamber. The latter combines the functions of the original buffer expansion tank and flow reactor chamber, and replaces them.

reacted gas/aerosols for analysis in the cloud chamber itself since its volume is much larger than the cloud chamber (by a factor 64).

The estimated extra cost of the reactor chamber over the original buffer expansion tank and flow chamber it replaces is 30 kCHF.

2.2 Reactor chamber design

The reactor chamber is a cylinder of internal dimensions 2 m (diameter) \times 2 m (height). The inner vessel can be operated between atmospheric pressure and a vacuum. It is constructed from aluminium, with black teflon lining the inner surface. The teflon is made partly conductive in order to prevent any charge buildup. An inner field cage provides a simple clearing field.

The reactor chamber is exposed to the same beam as the cloud chamber and operated at the same temperature and pressure. However, since it simply provides a container for trace gases and aerosols to react in the presence of the beam (i.e. no droplet activation is involved), the requirements on the temperature stability are less demanding than for the cloud chamber, and a stability of a few \times 0.1 K is adequate. The temperature is controlled with liquid fluorocarbon circulated in pipes around the outer surface of the reactor vessel. Heater cables are also wrapped around the reactor vessel to allow cleaning by bakeout under vacuum. A simplified thermal insulation is employed, comprising superinsulation and a vacuum layer. The liquid system is independent of the one used for the cloud chamber in order to maintain the precise temperature stability of 0.01 K required for the cloud chamber.

The reactor chamber is equipped with gas/aerosol inlet and outlet pipes. A small fan is installed inside the vessel to provide the option of slow stirring of the gas filling to assist, where necessary, homogeneous mixing throughout the large volume. A special inlet pipe located near the fan provides fresh trace gas to replace losses to the walls or to aerosol growth. The reactor vessel is also equipped with sampling probes to extract gas and aerosols for the external detectors (mass/ion/mobility spectrometers, trace gas analysers and aerosol particle sizers). The tip of each sampling probe can be independently adjusted to any radial position ($0 < r < 1$ m) inside the vessel. The reactor vessel also includes ports connecting to the vacuum system and to the active volume of the cloud chamber.

Access to the inside of the reactor vessel is provided by a removable hatch in the side wall and, for full access, the top of the vacuum tank and reactor vessel are also removable. The vessel is equipped with internal illumination and two inspection ports. Internal UV lamps are also provided for experiments involving photochemical processes.

2.3 Reactor chamber performance

The performance of the reactor chamber principally concerns the lifetime of trace gases and aerosols due to wall losses. Aerosols that strike the walls will be lost due to attractive van der Waals forces. Since the diffusion coefficient of aerosol particles is small, the wall losses due to thermal diffusion are small (see Fig. 42 of the proposal). Trace gas molecules, on the other hand, have a high diffusion rate. However these molecules strike the walls at high velocity and mostly rebound (which is, after all, the origin of gas pressure in a

vessel). The fraction that do not rebound depends on temperature and on the nature of the gas molecules and the surface material.

The experience of the AIDA aerosol facility [2] at Forschungszentrum Karlsruhe is instructive in this regard. AIDA is a large cylindrical vessel of internal dimensions 4 m (diameter) \times 7 m (height) with an inner ceramic lining. The temperature is controlled to 0.5 K precision by cold air circulating around the outside of the vessel. It is equipped with an internal fan to ensure homogeneous mixing. Operation of the fan is found to have only a small effect on aerosol lifetimes. The measured $1/e$ lifetimes in AIDA for trace O_3 and NO_2 gases at room temperature are 70 h and 370 h, respectively. Since the lifetimes should scale as the linear dimension of the vessel, this implies the CLOUD reactor chamber should have corresponding lifetimes of about 30 h and 150 h, respectively. In fact, since teflon is superior to ceramic as a lining material, these lifetime figures are probably conservative. During long-duration experiments with CLOUD, the losses of trace vapours due to wall adhesion or to aerosol growth will be compensated by a continuous small inflow of trace gases.

3 Experimental and theoretical motivation

3.1 Present experimental knowledge

There are only sparse experimental data on the effect of ions in the atmosphere on new particle formation—and none, to our knowledge, on the effect of ions on particle growth from CN to CCN. Observations have been made of nucleation bursts of CN in the atmosphere that cannot be explained by classical theories. For example, Hörrak *et al.* [3] reported the spontaneous formation of bursts of intermediate size ions in urban air, which they suggest may be due to ion-induced nucleation. Also, Clarke *et al.* [4] observed formation of new ultrafine particles in the tropical marine (Pacific) boundary layer that could not be explained by classical binary ($\text{H}_2\text{SO}_4\text{-H}_2\text{O}$) homogeneous nucleation theory at the low ambient concentrations of sulphuric acid ($1\text{--}5\cdot 10^7$ molecules cm^{-3}).

However a recent study by Yu and Turco [5] based on an ion-mediated model *is* able to reproduce the observations of Clarke *et al.*, as described below. Their model indicates that the nucleation rate of fresh CN in the marine boundary layer¹ is generally limited by the available ion production rate from GCRs. In contrast, the nucleation rate in the upper atmosphere is generally limited by the trace vapour concentration since the temperatures are lower and the trace vapour saturation ratios correspondingly higher, and so binary homogeneous nucleation can occur at an appreciable rate. This could explain why satellite observations show a solar modulation signal only in clouds below about 3 km (Fig. 6 in the CLOUD proposal).

Direct experimental evidence that ions are involved in the nucleation of new particles under atmospheric conditions is lacking. However positive effects with ions have been seen. For example, Bricard *et al.* [6] observed new particle production in filtered (aerosol-free) Paris air exposed to very high radiation doses ($3\cdot 10^8$ Bq $\text{m}^{-3} \times 300$ s). Vohra *et al.* [7], on the other hand, carried out experiments with radon at naturally-occurring ionisation

¹The *boundary layer* extends from the Earth's surface up to about 1 km; the region extending from about 1 km to the tropopause is known as the *free troposphere*.

levels of 3–15 Bq m⁻³ and observed new particle production proportional to ionisation rate, but using artificial air containing high concentrations of trace gases (300 ppb SO₂, 100 ppb C₂H₄ and 160 ppb O₃).

In short, many of the CLOUD experiments will be the first of their kind and, as such, there are good prospects for making important advances in our understanding of the importance of ionising particle radiation on aerosol and cloud physics.

3.2 Physical model of ion-mediated nucleation and growth

Despite intensive research over several decades, the origin of the ubiquitous background of ultrafine aerosols in the troposphere has not yet been determined. Moreover even the fundamental mechanism that leads to new particle formation remains poorly understood. It has been suggested that ionisation from GCRs plays a key role in the formation of new aerosol particles [8]–[12], and indeed this will be one of the major areas of study for CLOUD.

The classical theory of binary H₂SO₄-H₂O homogeneous nucleation fails to explain observations of new ultrafine particle formation in clean regions of the lower atmosphere, such as occurs over oceans and in pristine continental air [4, 13, 14, 15]. Typically the nucleation rates predicted by classical theory are far lower (by as much as 10 orders of magnitude) than the experimentally-observed rates. Recent modelling work [5] demonstrates that thermodynamically-stable charged clusters, caused by vapours condensing onto ions produced by GCRs, can form at much lower ambient vapour concentrations and grow significantly faster than neutral clusters.

The steps involved in the creation of CCN from condensable vapours (in this case, sulphuric acid) are shown in Fig. 3. Molecular H₂SO₄-H₂O clusters form and evaporate continually (step 1) by kinetic motion. Under suitable conditions some clusters will reach the critical size of about 1–2 nm diameter—known as *nucleation* (step 2). Once the critical size is reached, continued growth of the cluster becomes preferential thermodynamically.

The nucleation of aerosols in the atmosphere involves several competing processes which include molecular clustering, evaporation, scavenging of condensable vapour by pre-existing particles, and sedimentation by rainfall. In this environment, electrically charged embryos have a competitive advantage over neutral embryos. Charged clusters provide additional electrostatic attractions with polar molecules, allowing critical embryos to form with fewer molecules than for neutral clusters. Therefore ions can greatly enhance the rate of formation of new particles in regions where the concentration of condensable vapours is too low for stable neutral clusters to form at an appreciable rate, such as frequently occurs in the marine boundary layer. The key parameters controlling the rate of new particle formation are the concentrations of condensable vapours, the GCR ionisation rate, and the surface area of pre-existing particles.

Once formed, the ultrafine condensation nuclei (UCN) continue to grow (step 3). The main growth process up to diameters of about 10 nm is molecular condensation; for larger sizes the main growth mechanism is coagulation of existing CN. During the growth of CN to CCN, other vapours in the atmosphere such as ammonia, nitric acid and organic compounds are known to be important. The growth rate is also expected to be enhanced by the presence of ions, becoming less significant with increasing size of the aerosol particle. This enhancement is largest when the two colliding particles have opposite sign (+–),

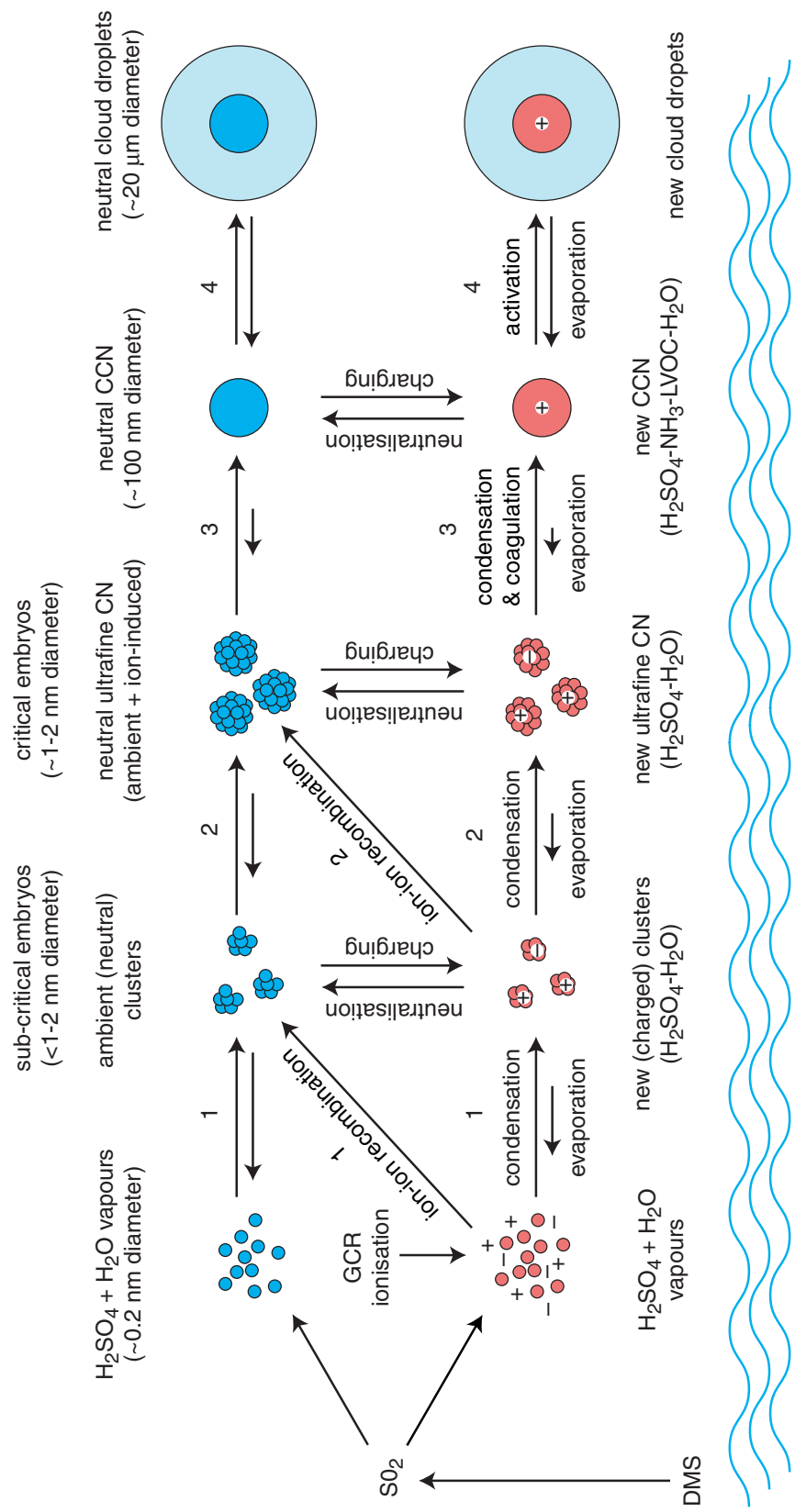


Figure 3: Possible influence of galactic cosmic rays (GCRs) on the nucleation of new condensation nuclei (CN) and on their growth into cloud condensation nuclei (CCN) in the marine boundary layer, ultimately increasing the number of cloud droplets. The labelled steps comprise: 1) formation of molecular clusters from condensable vapours, 2) formation of critical embryos (ultrafine CN) from molecular clusters, 3) growth of CN into CCN, and 4) activation of CCN into cloud droplets. The upper path corresponds to neutral (uncharged) aerosol particles and the lower path to charged aerosol particles (from GCR ionisation). Charged aerosols are expected to have an enhanced growth rate and reduced evaporation relative to neutral aerosols. Dimethyl sulphide (DMS) from plankton is the major source of sulphur dioxide—the precursor of sulphuric acid—in remote marine environments.

but there is also an increased rate between one charged and one neutral particle (+0 or -0), when compared with two neutral particles (00). Finally the activation of CCN into cloud droplets (step 4) may also be influenced by charge. However the effect is expected to be small for the typical electric charges found on aerosol particles under fair-weather conditions.

There is a continual interchange between charged and neutral particles as small ions diffuse onto existing CN and CCN, either neutralising or charging them in the process. This implies that ions may potentially affect the production rate of a large fraction of the CCN produced by gas-to-particle conversion, regardless of whether or not the original UCN were produced via ion-mediated processes.

It is important to note that aerosol particles and trace vapours are continually being scavenged from the atmosphere by rainfall. Following a precipitation event, the air is left with a relatively low aerosol particle number concentration and a low aerosol surface area. It is under these conditions that the nucleation of fresh UCN is most likely to occur. Furthermore, for clean environments, this indicates that the *rate* at which new particles are produced and grow into CCN can strongly affect the lifetime and radiative properties of the clouds in these regions, since there is rarely sufficient time for a large CCN population to form.

3.3 Aerosol effects on cloud radiative properties

The effect that a change in the CCN number concentration has on the radiative properties of a cloud can be quantitatively estimated as follows [16, 17]. Assuming the liquid water content and depth of the cloud is fixed, then its optical thickness, τ , is given by $\tau \propto Nr^2$, where N is the droplet number concentration and r the mean droplet radius. Since $N \propto r^{-3}$, this indicates $\tau \propto N^{1/3}$. Therefore a change of the droplet number concentration by ΔN leads to a change of the optical thickness by $\Delta\tau$, where

$$\frac{\Delta\tau}{\tau} = \frac{1}{3} \cdot \frac{\Delta N}{N} \quad (1)$$

The albedo (reflectivity), A , of a cloud is the fraction of incident radiation that is reflected into the backward hemisphere. For the scattering of solar radiation by clouds [16, 17],

$$A \approx \frac{\tau}{\tau + 6.7} \quad (2)$$

Differentiating Eq. 2 and combining with Eq. 1 gives,

$$\frac{\Delta A}{A} = (1 - A) \cdot \frac{\Delta N}{N} \quad (3)$$

The rather thin stratiform clouds that cover an appreciable fraction of the Earth's surface, and especially marine regions, have an albedo of about 0.5 and a droplet number concentration of about 100 cm^{-3} or less (see Fig. 9). Equation 3 shows that these clouds are very sensitive to changes in the CCN number concentration; their reflectance changes by 0.5% or more per single additional cloud droplet per cubic centimetre of air! This in turn indicates that GCR-induced changes in the CCN number concentration of only a few per cent could produce significant effects on the radiative properties of such clouds.

4 Exp.1: Nucleation of ultrafine CN

4.1 Model predictions

The recent model study of Yu and Turco [5] provides quantitative predictions of the ion-mediated nucleation of ultrafine condensation nuclei (UCN) and their subsequent growth, under the ambient conditions measured in the Pacific boundary layer by Clarke *et al.* [4]. Their model incorporates the effects of electrostatic interactions between charged and neutral molecules and clusters. Although the basic physics is well known, several parameters of the model are poorly constrained by experimental data—such as the ‘sticking’ probability when two particles collide—and, in these cases, reasonable values have been estimated. The model predictions should therefore be taken as representative of the magnitude of ion-mediated effects, rather than as definitive. Nevertheless the model studies of Yu and Turco represent the most quantitative that presently exist on the possible effects of GCR ionisation on aerosol formation and growth under atmospheric conditions.

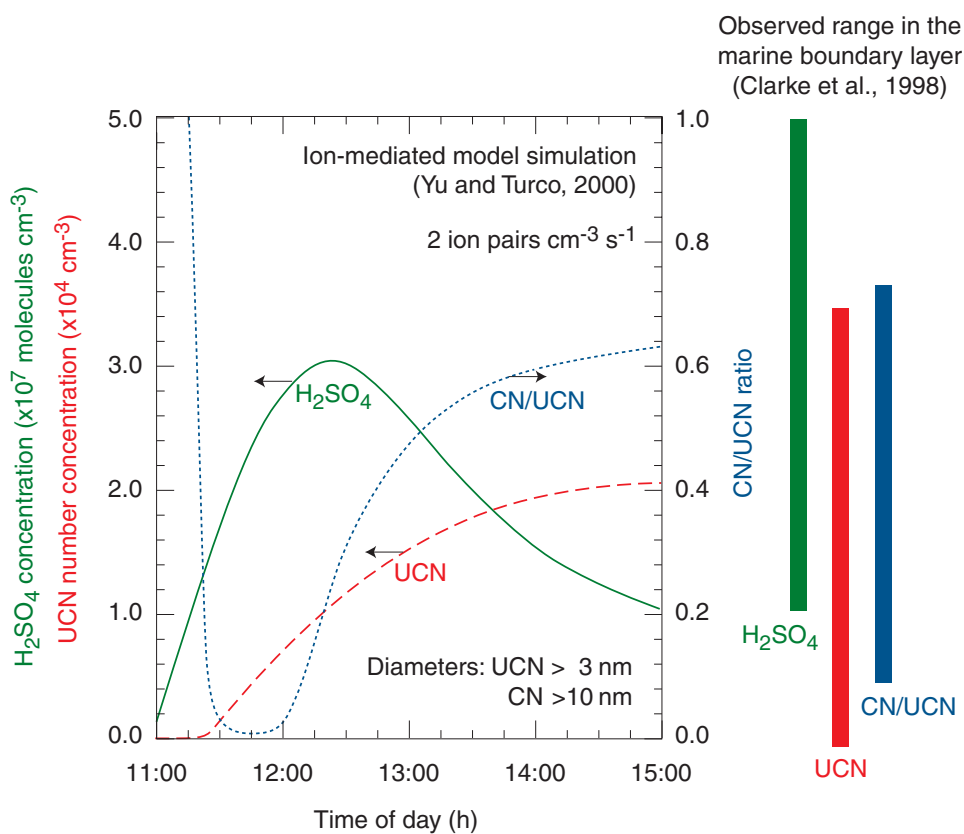


Figure 4: Ion-mediated model simulation by Yu and Turco [5] of the binary nucleation of new ultrafine condensation nuclei (UCN) from H₂SO₄-H₂O vapour in the marine boundary layer. An initial background CN concentration of 100 cm⁻³ is assumed. The simulations agree well with the range of values measured by Clarke *et al.* [4], indicated by the vertical bars on the right hand side. Classical binary homogeneous nucleation theory, which does not include ion-mediated effects, predicts no nucleation under these conditions (see Fig. 5).

To compare directly with the data of Clarke *et al.*, the simulation is initialised at 11:00 local time under the experimentally-observed conditions, namely $T = 298$ K, relative humidity (RH) = 95% and an initial background aerosol distribution of 100 cm^{-3} . Sulphuric acid vapour is generated *in situ* at the observed rate of

$$10^4 \cdot \sin[\pi(t - 6)/12] \text{ molecules cm}^{-3}\text{s}^{-1} \quad (4)$$

where $6 < t < 18$ h is the local time. The H_2SO_4 production rate is set to zero at other times. The ion pair production rate is $2 \text{ cm}^{-3}\text{s}^{-1}$, corresponding to GCRs at ground level.

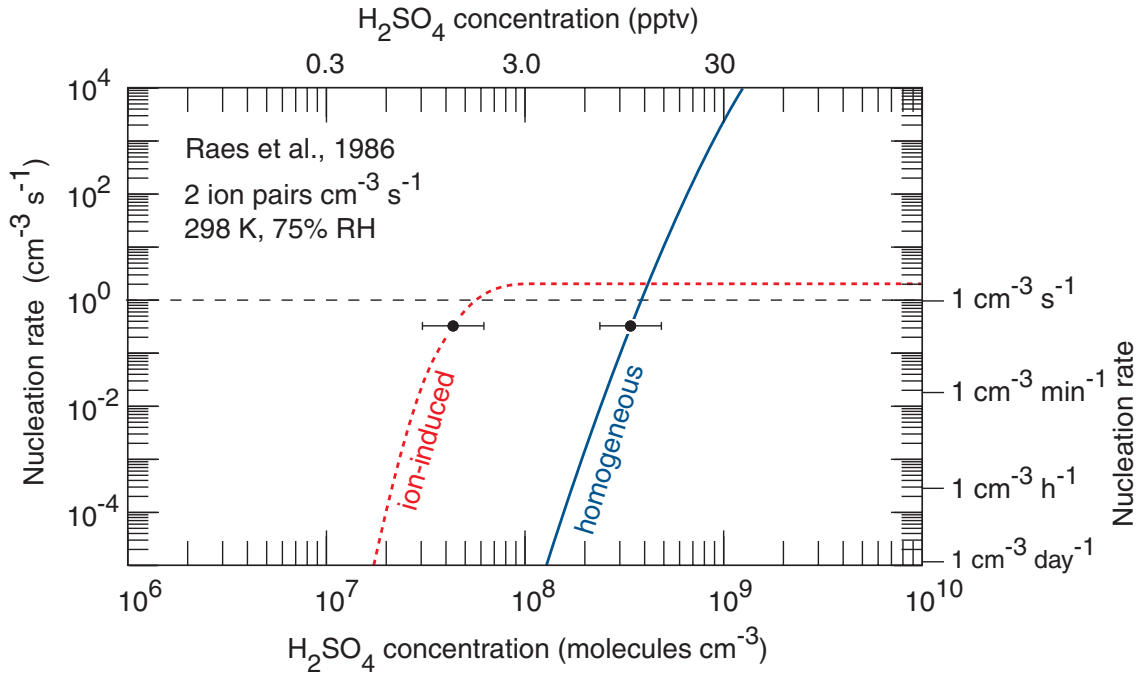


Figure 5: The rates of ion-induced and classical binary homogeneous nucleation of H_2SO_4 - H_2O aerosol particles as a function of H_2SO_4 concentration in the marine boundary layer, estimated by Raes *et al.* [10]. A background ($d = 100$ nm) CN concentration of 400 cm^{-3} is included. The expected measurement precisions for CLOUD are indicated by points with error bars. More recent estimates by Viisanen, Kulmala and Laaksonen [18, 19] suggest the curve labelled ‘homogeneous’ in the figure should be shifted to the right by about a factor five higher H_2SO_4 concentrations.

The results of the simulation are shown in Fig. 4, in comparison with the experimental data. The model predicts negligible UCN (>3 nm diameter) formation until the H_2SO_4 concentration reaches about 10^7 cm^{-3} (at about 11:15 local time) and then, within one hour, a UCN population of about 10^4 cm^{-3} has formed. The H_2SO_4 concentration falls after 12:00 due to a decreased production rate and to scavenging by the aerosol particles. The newly-formed particles continue to grow, and after 12:00 an appreciable fraction

exceed 10 nm diameter (labelled ‘CN’). The simulation agrees well with the experimental observations of Clarke *et al.*, which are indicated by the vertical bars in Fig. 4.

Under these conditions classical binary homogeneous nucleation theory predicts an insignificant production of new particles. Figure 5, for example, shows the results of an earlier calculation by Raes, Janssens and Van Dingenen [10], which indicates negligible new particle formation (i.e. below the so-called *critical nucleation rate* of $1 \text{ cm}^{-3}\text{s}^{-1}$) until the H_2SO_4 concentration reaches about $4 \cdot 10^8$ molecules $\text{cm}^{-3}\text{s}^{-1}$. Raes *et al.* estimate the threshold for ion-induced nucleation occurs at an H_2SO_4 concentration about one order of magnitude lower. (Yu and Turco estimate the ion-mediated threshold to be still lower by about a factor of four, i.e. near 10^7 molecules $\text{cm}^{-3}\text{s}^{-1}$. This is mainly due to the higher RH and lower background aerosol concentration in Yu and Turco’s simulation.) Note also from Fig. 5 that the maximum production rate of new aerosols by ion-induced nucleation is predicted to be limited by the GCR intensity ($2 \text{ cm}^{-3}\text{s}^{-1}$ at ground level).

4.2 Experimental goals

The purpose of this experiment is to measure the effect of ionising particle radiation on the rate of formation of ultrafine condensation nuclei in the few-nm size range, in the presence of sulphuric acid and water vapours. Sulphuric acid is chosen because field measurements indicate it is an important precursor vapour for gas-to-particle conversion in the atmosphere.

The basic parameter to be measured is the *nucleation rate*, J [$\text{cm}^{-3}\text{s}^{-1}$], of UCN as a function of the primary experimental variables, namely, sulphuric acid vapour concentration, relative humidity, temperature, background aerosol concentration and ion-pair production rate. A representative expectation of the variation of J with sulphuric acid vapour concentration and ion-pair production rate is shown in Fig. 5. This figure indicates that a large measurement range is required for the nucleation rate since it is a steep function of parameters such as vapour concentration (and also temperature).

The nucleation rate is determined by a measurement of the UCN number concentration [cm^{-3}] after a time t [s]. In the case of the cloud chamber, the minimum detectable particle number (after activation to visible droplets) is about 1 particle per 10 cm^3 . Assuming an exposure time of an hour (3600 s), this indicates the minimum measurable nucleation rate is $J \sim 3 \cdot 10^{-5} \text{ cm}^{-3}\text{s}^{-1}$. The maximum nucleation rate that can be measured is about 12 orders of magnitude higher. The maximum measurable droplet density in the cloud chamber is about 10^7 cm^{-3} . Depending on the variable under study, if the effective exposure time is adjusted to about 1 s duration then a maximum J of $10^7 \text{ cm}^{-3}\text{s}^{-1}$ can be measured. If the variable under study is ion pair concentration then this is achieved by initiating a cloud chamber expansion cycle 1 s after the arrival of the beam pulse. If the variable is sulphuric acid saturation ratio, then a short, precise saturation pulse can be generated by a piston expansion during the nucleation phase, followed by a small re-compression which arrests nucleation but allows droplet growth and observation. We have used the latter technique extensively for nucleation studies with the Vienna cloud chambers (see, for example, ref. [20]).

4.3 Experimental measurements

The experimental conditions for the first run are as follows:

H ₂ SO ₄ molecular concentration	5 · 10 ⁷	cm ⁻³
Relative humidity	100	%
Temperature	298	K
Background aerosol concentration	0	cm ⁻³
Pressure	101	kPa
Beam intensity	10 ³	π/p pulse ⁻¹

The carrier gas is pure artificial air (80:20 N₂:O₂). The procedure is first to adjust the cloud chamber and reactor chamber to the desired temperature and then to fill both of them with the specified gas mixture at the desired pressure.

Before starting the run, thermodynamic equilibrium is established. During this period the clearing field is used to clear ions produced by GCRs from the chambers. The clearing time for small ions is about 2 s in the cloud chamber and 8 s in the reactor chamber. This should be compared with the typical ion lifetime of ~ 1000s under atmospheric conditions and with the time needed for an ion to attract an H₂SO₄ molecule of ~ 50 s at these concentrations. Therefore when the clearing field is present it reduces the ambient ion pair concentration from GCRs by about a factor 100 below the natural level, and also essentially excludes ion-mediated nucleation processes. The clearing field is therefore a very effective tool for measurements at “zero GCR” ionisation.

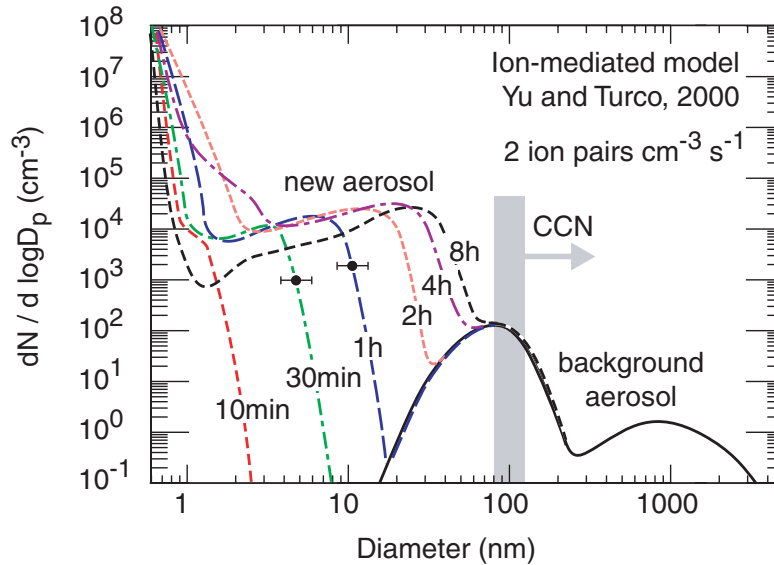


Figure 6: Evolution of the CN size spectra from ion-mediated nucleation and growth under conditions typical of the marine boundary layer [5]. The expected measurement precisions for CLOUD are indicated by the points with error bars. The solid curve indicates the initial assumed background aerosol concentration of 100 cm⁻³. When sulphuric acid-water CN reach diameters of about 100 nm they become efficient CCN for seeding clouds.

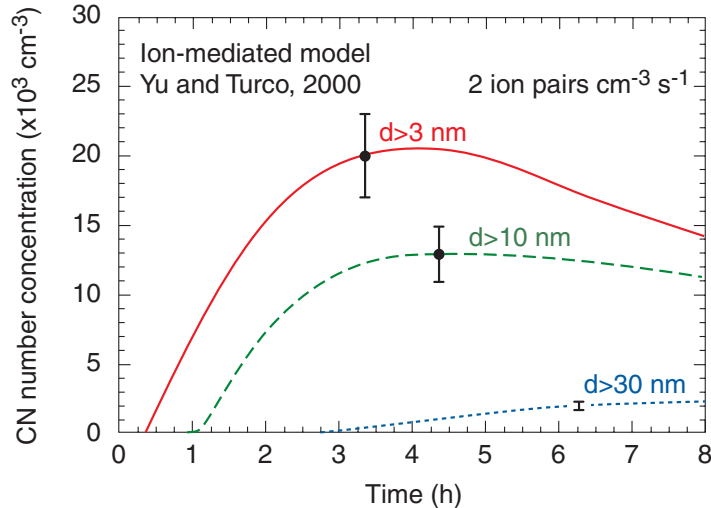


Figure 7: Ion-mediated nucleation and growth of CN under conditions typical of the marine boundary layer [5]. Each curve shows the evolution of the number of CN above an indicated minimum diameter, d . The expected measurement precisions for CLOUD are indicated by points with error bars.

After thermodynamic equilibrium is established, the chambers are exposed to the beam for the required period. In the case of UCN nucleation and growth to ~ 10 nm diameter, the characteristic times at atmospheric vapour concentrations are up to about one hour (Figs. 6 and 7), with the first UCN appearing after about 10 minutes. During the beam exposure small quantities of gas are continually drawn off from the reactor chamber via the sampling probes for analysis in the external mass spectrometers, mobility spectrometers, and particle sizers and counters. In this way the evolution of the UCN nucleation and growth can be analysed and recorded continuously during the 1-hour beam exposure.

In order to detect and measure UCN concentrations in the cloud chamber, a large volume expansion ratio of about 35% is required to create a water vapour supersaturation of about 600%. Under these conditions, the cloud chamber is sensitive to all particles above 1.2 nm diameter and to all *charged* particles down to molecular sizes of 0.2 nm (see Fig. 49 in the proposal). In the case of commercial particle sizers the lowest detection limit is about 3 nm diameter. Therefore, when studying UCN nucleation, the cloud chamber will provide information complementary to the external particle sizers. The latter will be used to measure the UCN size spectra versus time (i.e. the distributions shown in Fig. 6). The cloud chamber, in contrast, integrates the particle count above a certain minimum size which is set by the expansion ratio. Once an expansion and droplet activation cycle has taken place, the cloud chamber must be refilled with a fresh mixture since the residual aerosols after droplet evaporation may have been altered during the activated phase.

During each run the following quantities will be measured with the detectors² indicated:

<u>Quantity</u>	<u>Detector</u>
UCN nucleation rate, J [$\text{cm}^{-3}\text{s}^{-1}$]	Cloud chamber, CPC
UCN size spectrum	SMPS
UCN mass spectrum ($< 10^4$ amu)	ToFMS, IOMAS
UCN chemical composition	PITMAS
Charged UCN mobility spectrum	PIMS
Trace gas concentrations	CIMS
Small ion concentration	PIMS
Ion pair production rate	BCS

The expected experimental precisions for the first two quantities are indicated by the error bars in Figs. 5–7. In addition the various thermodynamic conditions—such as temperature, pressure and relative humidity—will be recorded.

After completing a run with a fixed set of conditions, the ‘primary’ variable under study is changed and the next run is started. The primary variables are those for which the nucleation rate may be most sensitive, and which will therefore be studied in detail. They include the H_2SO_4 concentration, relative humidity, temperature, background aerosol concentration, and ionisation rate. Pressure is a secondary variable. The approximate ranges of study for the primary variables are as follows:

H_2SO_4 concentration	$3 \cdot 10^6 - 10^{10}$	cm^{-3}
Relative humidity	40 – 100	%
Temperature	268 – 298	K
Background aerosol concentration	0 – 1000	cm^{-3}
Beam intensity	0 – 10^3	π/p pulse ⁻¹

This series of measurements should allow CLOUD to determine the effect of GCR ionisation on the nucleation of sulphuric acid-water UCN under conditions corresponding to the marine boundary layer. This experiment is estimated to require 4 weeks beamtime (§7.2).

5 Exp.2: Growth of CN into CCN

5.1 Model predictions

Yu and Turco have extended their ion-mediated simulation in time to follow the growth of freshly formed UCN into CCN over a period of several days [21]. The initial conditions are the same as described in §4.1 except that organic vapours are now included in the simulation, and an initial background aerosol distribution of 10 cm^{-3} is assumed, which corresponds to clean air, freshly scavenged by precipitation. Although H_2SO_4 vapour is

²The acronyms are as follows: BCS (beam counter system), CIMS (chemical ionisation mass spectrometer), CPC (condensation particle counter), IOMAS (ion mass spectrometer), PIMS (programmable ion mobility spectrometer), PITMAS (quadrupole ion trap mass spectrometer), SMPS (scanning mobility particle sizer) and ToFMS (time-of-flight mass spectrometer).

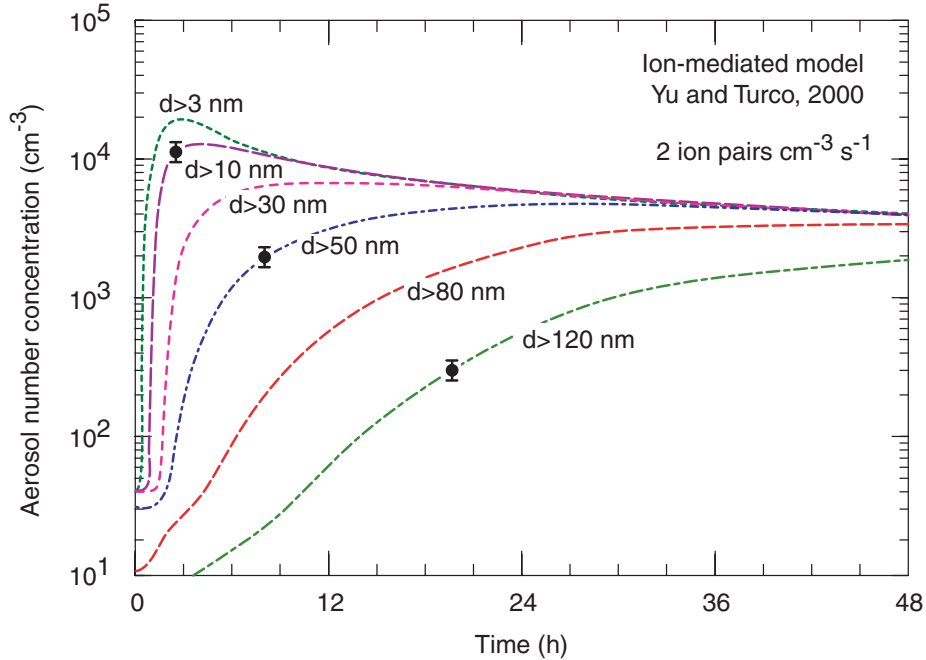


Figure 8: Ion-mediated model predictions of the nucleation of UCN and growth into CCN in the presence of trace H_2SO_4 and organic vapours, under conditions typical of the marine boundary layer [21]. When these aerosols reach diameters of about 100 nm they become efficient CCN. The expected measurement precisions for CLOUD are indicated by the points with error bars. An initial background CN number concentration of 10 cm^{-3} is assumed. Note that dilution (mixing) has not been considered in these simulations, so the final aerosol concentrations are much higher than are typically found in the marine boundary layer ($\sim 100 \text{ cm}^{-3}$).

important for the nucleation of new aerosols, the atmospheric concentrations are generally too small to account for all of the subsequent rate of growth of CN into CCN, and so additional vapours are implicated. The model assumes constant production rates for non-volatile organic compounds of $0.5 \mu\text{g m}^{-3} \text{ day}^{-1}$ and for low volatile organic compounds of $5 \mu\text{g m}^{-3} \text{ day}^{-1}$, as suggested by field observations.

The results are shown in Fig. 8. The simulations indicate a significant production of CCN (diameters above $\sim 100 \text{ nm}$) by ion-mediated processes after about 24 h. These characteristic times for the *in situ* growth of new CN and CCN are consistent with our field measurements [15, 22]. Note in Fig. 8 that, although the H_2SO_4 vapour production rate peaks each day at 12:00 (Eq. 4), it does not lead to the formation of new UCN after the first day because of scavenging by the existing aerosols.

5.2 Experimental goals

The purpose of this experiment is to measure the effect of ionising particle radiation on the rate of growth of CN into CCN (i.e. from $\sim 5 \text{ nm}$ diameter to $\sim 100 \text{ nm}$) in the presence of

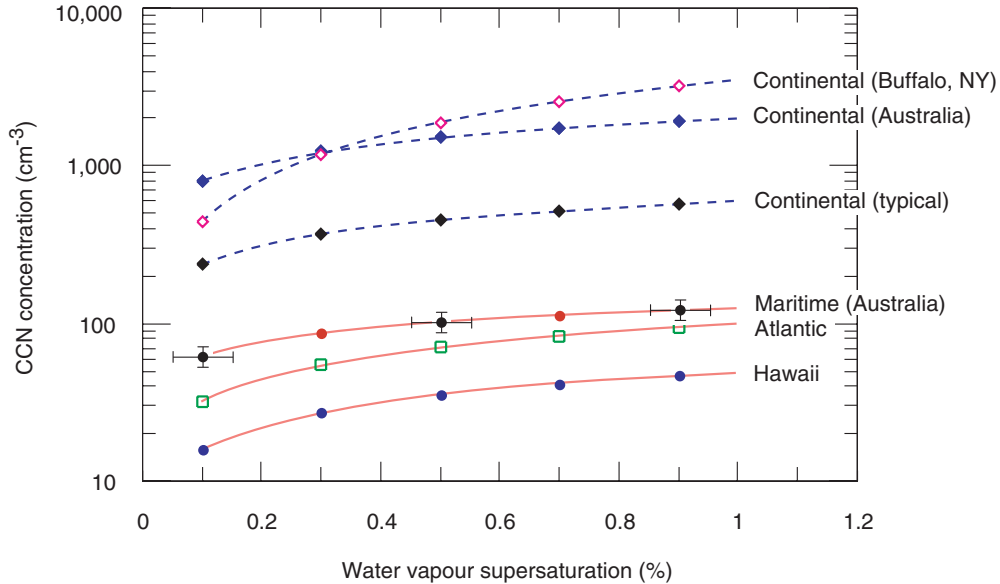


Figure 9: Measurements of CCN concentrations at several sites: marine (solid curves) and continental (dashed curves) as a function of the water vapour supersaturation [24]. The CCN concentrations are equal to the cloud droplet concentrations at a given supersaturation. The expected measurement precisions for CLOUD are indicated by the points with error bars.

condensable vapours that are known to be important in the atmosphere: sulphuric acid, water, ammonia and organic compounds.

The basic parameter to be measured is the CCN concentration, $CCN(s)$. The CCN are the subset of the CN population that activate into droplets at a specified water vapour supersaturation, s . The fraction of the CN that constitute CCN depends not only on their size but also on their chemical composition and other properties. Field measurements show that $CCN(s)$ [cm^{-3}] can often be parametrised as [23]

$$CCN(s) = c s^k$$

where s [%] is the water vapour supersaturation, and c [cm^{-3}] and k are empirical parameters that embed the information about the size and chemical composition of the aerosol population. Measurements of $CCN(s)$ versus s at several sites are shown in Fig. 9 [24]. This figure illustrates the low CCN population ($10\text{--}100\text{ cm}^{-3}$) that characterises clean marine regions. The peak values of s reached in the atmosphere depend on the spectrum of CCN and on other conditions such as the updraft velocity of the air parcel. For marine stratiform clouds, the typical values of s are in the range 0.1 to 0.5%. Even for strongly convective clouds the peak values of s are rarely above 1% since droplet activation arrests further increase. In the CLOUD experiment the CCN concentration is measured with the cloud chamber operated with a small volume expansion ratio of $\sim 10^{-3}$, corresponding to a piston movement of $\sim 500\text{ }\mu\text{m}$ (see Table 5 on p.65 of the proposal). The expected measurement precision of $CCN(s)$ is indicated in Fig. 9.

Other important parameters to measure in this experiment are the size-discriminated aerosol number concentrations (Fig. 8) and the aerosol size spectra (Fig. 6). Both will be measured versus time over the duration of each run, which lasts about 2 days.

5.3 Experimental measurements

The experimental conditions for the first run are the same as for the previous experiment (§4.3), with the addition of ammonia and low volatility organic compounds (LVOCs)³ in trace amounts:

NH ₃ concentration	1 ppbv
LVOC concentration	5 $\mu\text{g m}^{-3} \text{ day}^{-1}$

The procedure is similar to the previous experiment, except in the following respects. The beam exposure time for a single run is now 2 days. During this time small quantities of the trace vapours are introduced into the cloud chamber and the reactor chamber to compensate for wall losses and for aerosol growth. As before, during the beam exposure small samples of gas are continually drawn off from the reactor chamber via the sampling probes for analysis in the external detectors. In this way the evolution of the CN into CCN can be measured over the course of the beam exposure.

After the first expansion of the cloud chamber to measure $CCN(s)$ at, say, $s = 0.3\%$, the cloud chamber is cleared and re-filled from the reactor chamber. The volume of the cloud chamber is less than 2% of that of the reactor chamber. Therefore a total of about 5 samples for $CCN(0.3\%)$ measurements can be made this way over the course of the 2 day exposure period without serious impact on the contents of the reactor chamber. At the end of the 2-day run, several samples will be drawn off sequentially from the reactor chamber into the cloud chamber in order to measure the final $CCN(s)$ distribution over the range $0.1 < s < 1.0\%$ ⁴ (Fig. 9).

During each run the following quantities will be measured:

<u>Quantity</u>	<u>Detector</u>
CCN concentration, $CCN(s)$ [cm^{-3}]	Cloud chamber
CN concentration	CPC
CN size spectrum	SMPS
UCN mass spectrum ($< 10^4$ amu)	ToFMS, IOMAS
CN chemical composition	PITMAS
Charged CN mobility spectrum	PIMS
Trace gas concentrations	CIMS
Small ion concentration	PIMS
Ion pair production rate	BCS

As for the previous experiment, after completing a run with a fixed set of conditions, the primary variable under study is changed and the next run is started. In view of

³LVOCs that may be important for aerosol growth in the atmosphere include terpenes, acetone, isoprene and the higher n -alkanes.

⁴The cloud chamber hydraulic system can be programmed to provide a continuous, precise slow piston expansion if required. This may allow a continuous scan measurement of $CCN(s)$ to be made over the range $0.1 < s < 1.0\%$ with a single chamber filling.

the long duration of these runs, the primary variables in the first round experiment will be limited to trace gas concentrations, background aerosol concentrations, and beam intensity. Their approximate ranges of study are as follows:

LVOC concentration	0.5 - 50	$\mu\text{g m}^{-3} \text{ day}^{-1}$
Background aerosol concentration	0 - 1000	cm^{-3}
Beam intensity	$0 - 10^3$	$\pi/\text{p pulse}^{-1}$

For the run at zero beam intensity, it may be necessary to start the experiment with a beam exposure of about 1 hour in order to generate the initial CN population. For the remainder of this run, the beam would be turned off so the growth of the CN into CCN could be measured under zero-beam conditions.

This series of measurements should allow CLOUD to determine the effect of GCR ionisation on the growth of CN into CCN under conditions characteristic of the marine boundary layer. The CCN concentrations will be parametrised as a function of water vapour supersaturation and other variables so the results can be readily incorporated into cloud models. This experiment is estimated to require 8 weeks beamtime (§7.2).

6 Goals of the aerosol and cloud simulations

The experimental results obtained with CLOUD will be evaluated with aerosol and cloud simulations. The basic physics aims of these simulations are as follows:

1. To incorporate the microphysics of ion-mediated processes into the aerosol and cloud models.
2. To examine the sensitivity of clouds under atmospheric conditions to variations in the GCR intensity, in the presence of other sources of natural variability.

As is familiar with Monte Carlo simulations in particle physics experiments, there will be a close feedback between the simulations and the experimental observations to confirm a) that the simulations closely reproduce the experimental data and b) that the underlying microphysics is understood. We expect the simulations will also eventually become important in guiding the direction of the experimental programme.

As well as a detector simulation, this work will require a complete simulation of the microphysical processes under study. Already existing within our collaboration are detailed models of aerosol nucleation, growth and activation, and also two sophisticated cloud models—for cumulus and marine stratus clouds, respectively. Ion-mediated microphysics will be incorporated into these models based on the experimental data from CLOUD.

It is important to make clear that the present modelling expertise of our collaboration extends as far as single clouds. If item 2 above reveals discernible GCR effects on single clouds, we would seek to provide suitable parametrisations for the climate modelling community to explore the influence of GCR effects on the global climate.

7 Accelerator requirements

7.1 Beam requirements

As described in the proposal (§7), we request to install CLOUD at the CERN PS in the T11 beamline of the East Hall. The experimental area layout including the new reactor chamber is shown in Fig.10. A dedicated counting room for CLOUD will be installed above the current T11 counting room. The cloud chamber and reactor chamber will be permanently located at the downstream end of the T11 beamline, with the external detectors installed nearby. The upstream half of the present free space along the T11 beamline will be left open for installing small test experiments during the beam periods that CLOUD is not operating. However, during its operation periods, CLOUD would be the sole user of T11.

The present energy, maximum intensity and particle composition of the T11 beam is suitable for CLOUD. However, we have a special requirement that the minimum beam intensity can be adjusted to about 100 particles per pulse spread over an area of about $30 \times 30 \text{ cm}^2$ (i.e. near to the natural GCR ionisation rate). The low intensity may be reached by closing beam collimators and perhaps adding a beam absorber. By suitable defocusing of the quadrupole magnets in the T11 beamline, a transverse size at the detector of $30 \times 30 \text{ cm}^2$ seems feasible. The transverse dimensions of the beam scintillation counters have been increased to $40 \times 40 \text{ cm}^2$ to accommodate the larger beam size (Figs. 1 and 2).

Table 1: Estimated beam requirements and schedule during the first year of CLOUD operation.

Date	No. weeks	Activity or Experiment
Mar - Apr 2002	8	Checkout
May 2002	4	Exp.1: UCN nucleation
Jun 2002	4	Exp.2: CN→CCN growth
Oct 2002	4	Exp.2: CN→CCN growth
Nov 2002	4	Exp.3: Creation of ice nuclei
Totals:	8 weeks 16 weeks	Checkout Data taking

7.2 Beam schedule

The estimated beamtime required during the first year of CLOUD operation is 24 weeks, as summarised in Table 1. The checkout period includes setting up the correct operation of the hardware and data acquisition, and carrying out technical performance tests.

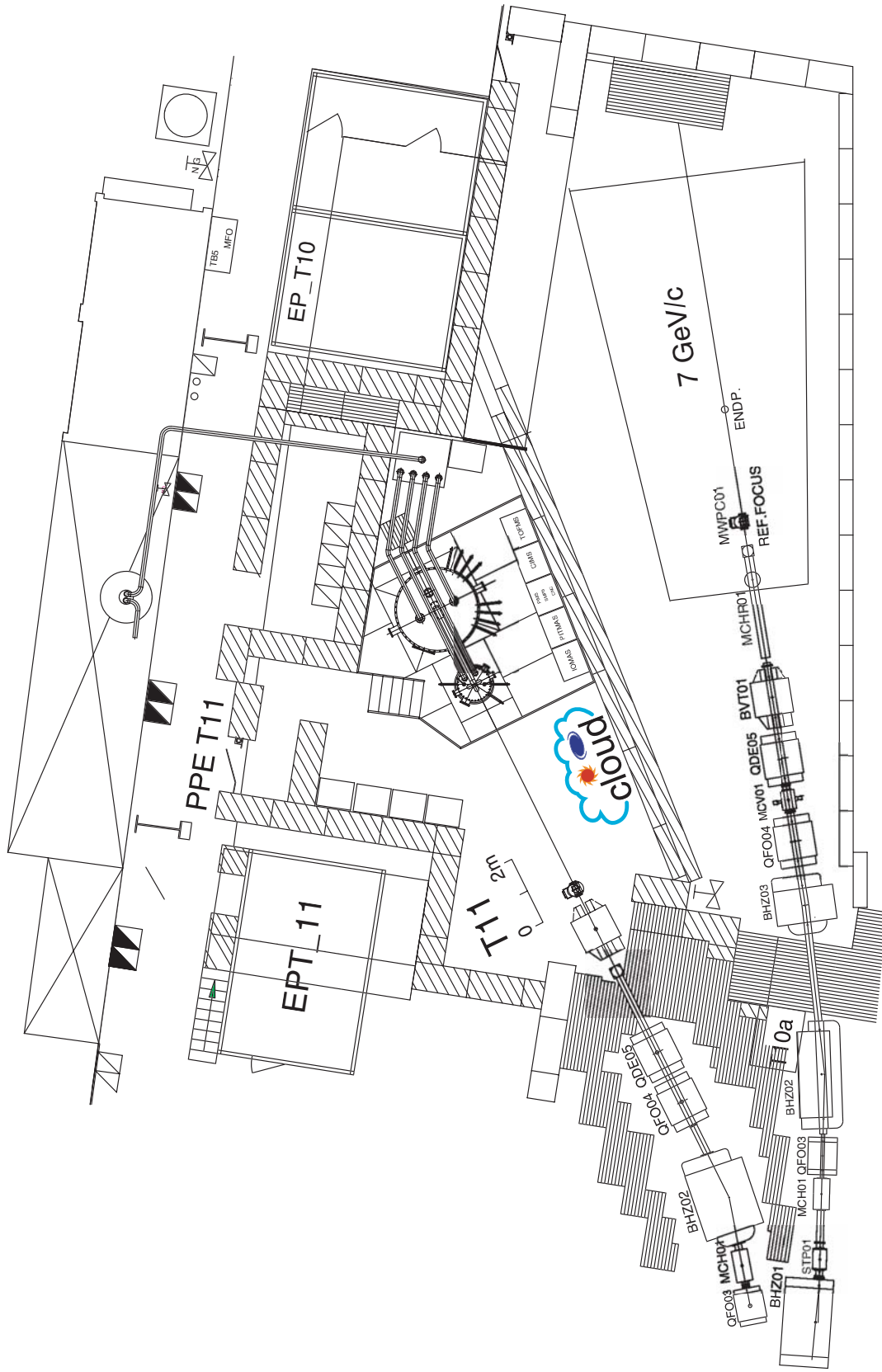


Figure 10: Experimental area layout of the CLOUD experiment at the CERN PS in the T11 beamline of the East Hall.

The estimated beamtime required for each of the two experiments described in this document is 4 weeks and 8 weeks, respectively, as follows:

Exp.1. Nucleation of UCN: Each run is estimated to require 1 shift (8h) for setup and beam exposure. The experimental variables and the approximate number of different values that will be measured are as follows:

H ₂ SO ₄ concentration	6 measurements
Relative humidity	3 "
Temperature	4 "
Background aerosol concentration	3 "

This totals 16 runs, assuming only the variable under study is changed in a given family of measurements. Each of these 16 different sets of operating conditions will be measured at 4 beam intensities: 0, 1, 3 and 10 × the natural GCR ionisation rate in the boundary layer. (A value of ‘0’ corresponds to the beam-off measurement.) The total beam time is therefore 64 shifts (3 weeks); or a calendar time of 4 weeks after allowing for downtime, chamber cleaning, etc.

Exp.2. Growth of CN into CCN: Each run is estimated to require 6 shifts (48h) for setup and beam exposure. The experimental variables and the approximate number of different values that will be measured are as follows:

Trace gas concentrations	4 measurements
Background aerosol concentration	3 "

This totals 7 runs, each requiring 6 shifts, to total 42 shifts. Each of these 7 different sets of operating conditions will be measured at 3 beam intensities: 0, 1 and 10 × the natural GCR ionisation rate. The total beam time is therefore 126 shifts (6 weeks); or a calendar time of 8 weeks.

As regards the experimental programme for the subsequent years, 2003 and 2004, at this stage it is not possible to define the schedule precisely since it will depend on the outcome of the initial studies as well as developments elsewhere in the field. However we can estimate the beamtime required for the various experiments summarised in Table 3 (p.29) of the proposal as follows:

<u>Experiment</u>	<u>Beamtime [weeks]</u>
NO _x formation	2
Polar stratospheric aerosols	6
Droplet activation	4
UCN nucleation II (ternary vapours)	8
CN → CCN growth II	8
Creation of ice nuclei II	4
Total:	32 weeks

Therefore during 2003 and 2004 we estimate at least 16 weeks of beamtime will be required for CLOUD each year, which represents just over one half of the normal yearly operation of the East Hall beamlines.

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