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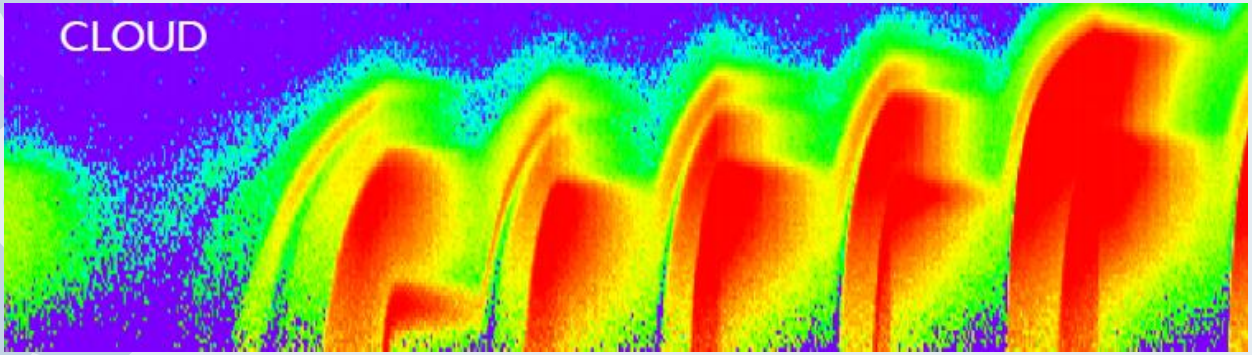
Status and plans of the CLOUD experiment



Joachim Curtius and the CLOUD team

Institute for Atmospheric and Environmental Sciences
Goethe-University of Frankfurt am Main

Outline

Introduction

The CLOUD experiment in 2012

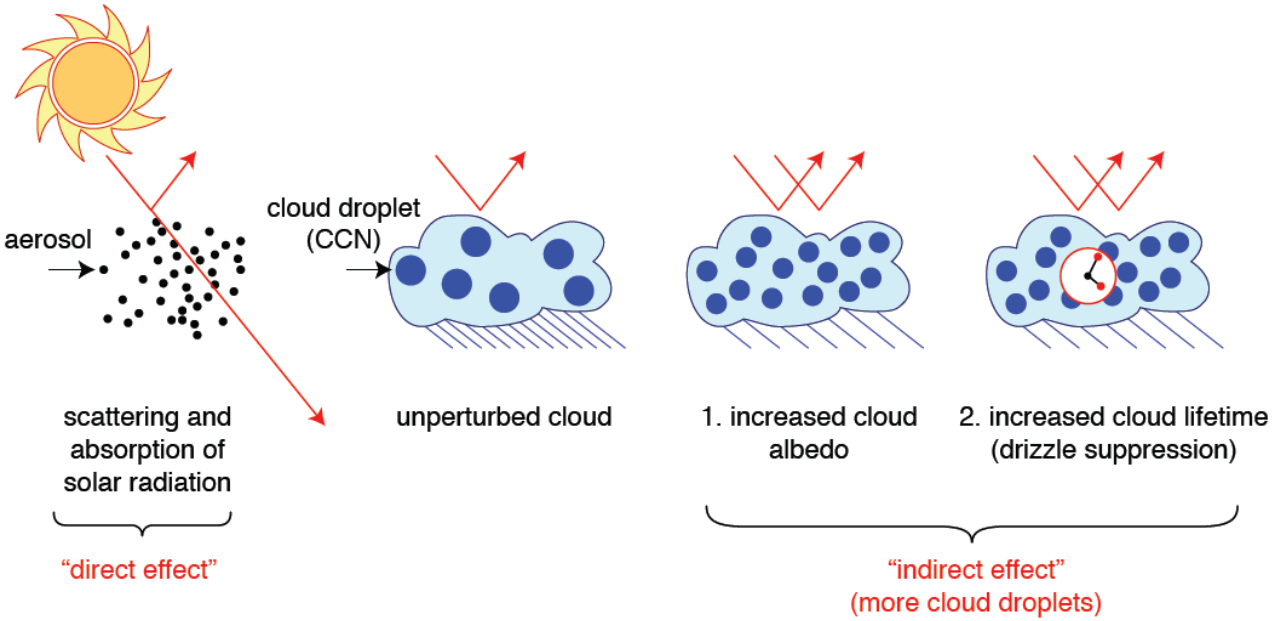
- CLOUD 6: first adiabatic expansion runs
- CLOUD 7: aerosol nucleation and growth runs:
  - dimethylamine-ternary system
  - α-pinene-ternary system

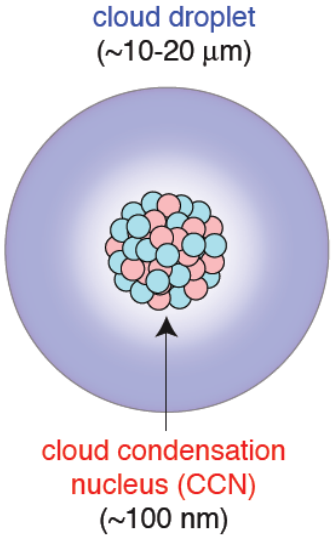
Future plans

CLOUD Collaboration

Austria:	University of Innsbruck
	University of Vienna
Finland:	Finnish Meteorological Institute
	Helsinki Institute of Physics
	University of Eastern Finland
	University of Helsinki
Germany:	Goethe-University of Frankfurt
	Karlsruhe Institute of Technology
	Institute for Tropospheric Research
Portugal:	University of Beira Interior
	University of Lisbon
Russia:	Lebedev Physical Institute
Sweden	University of Stockholm
Switzerland:	CERN
	Paul Scherrer Institute
	Tofwerk
United Kingdom:	University of Leeds
	University of Manchester
USA:	Aerodyne Research Inc.
	California Institute of Technology
	Carnegie Mellon University

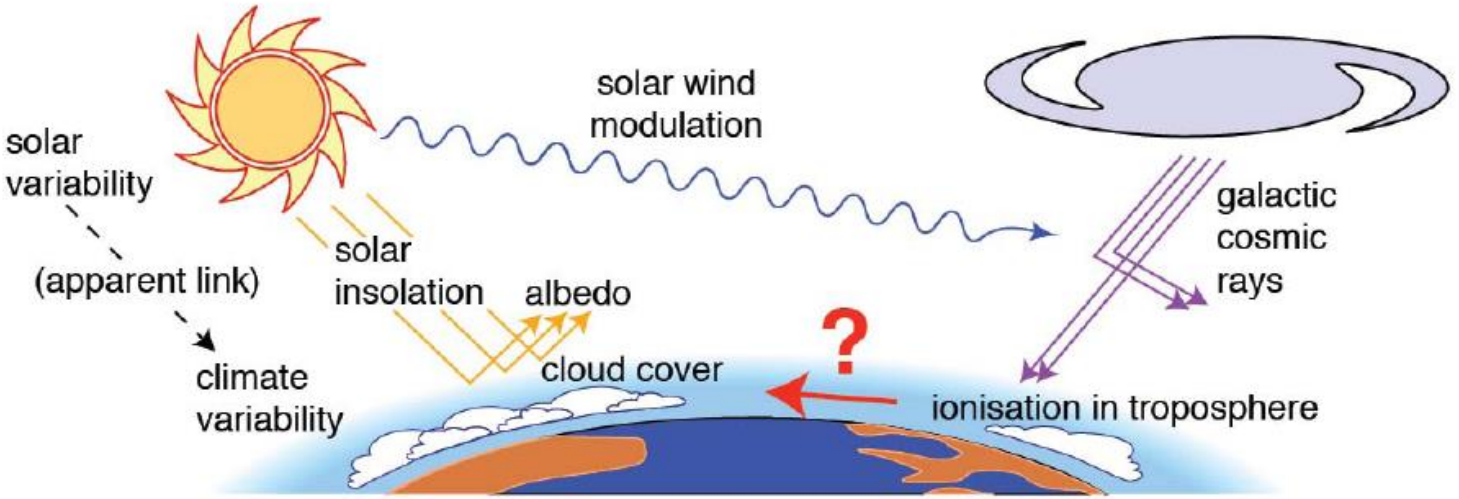
Importance of aerosols for the climate

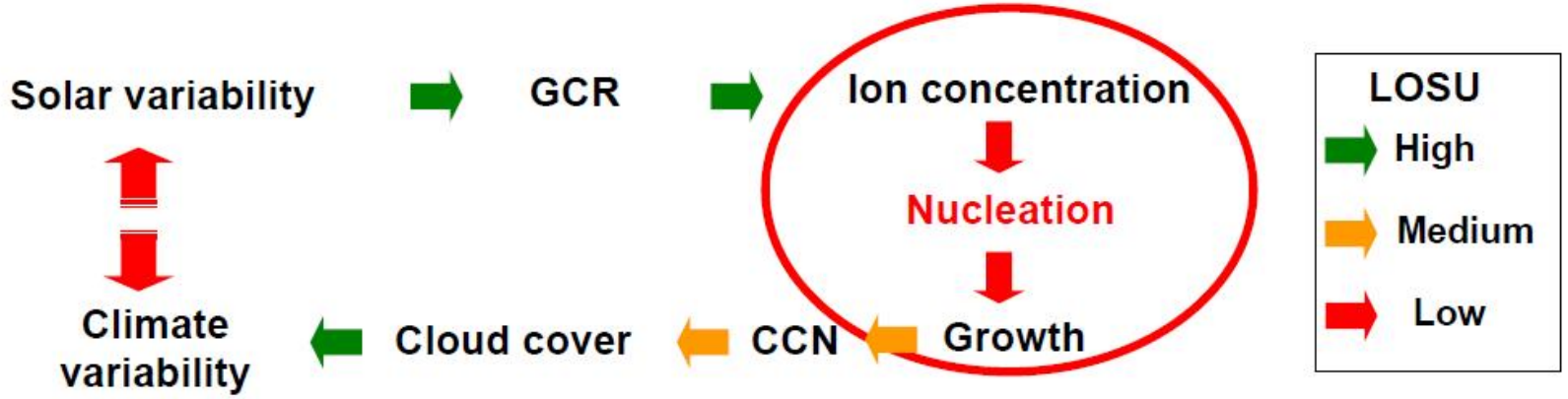




- All cloud droplets form on aerosol "seeds" known as cloud condensation nuclei - CCN
- Cloud properties are sensitive to number of droplets
- More aerosols/CCN:
  - brighter clouds, with longer lifetimes
- Sources of atmospheric aerosols:
  - direct (dust, sea salt, fires)
  - indirect (gas-to-particle conversion)

A possible mechanism for a link between galactic cosmic rays and clouds

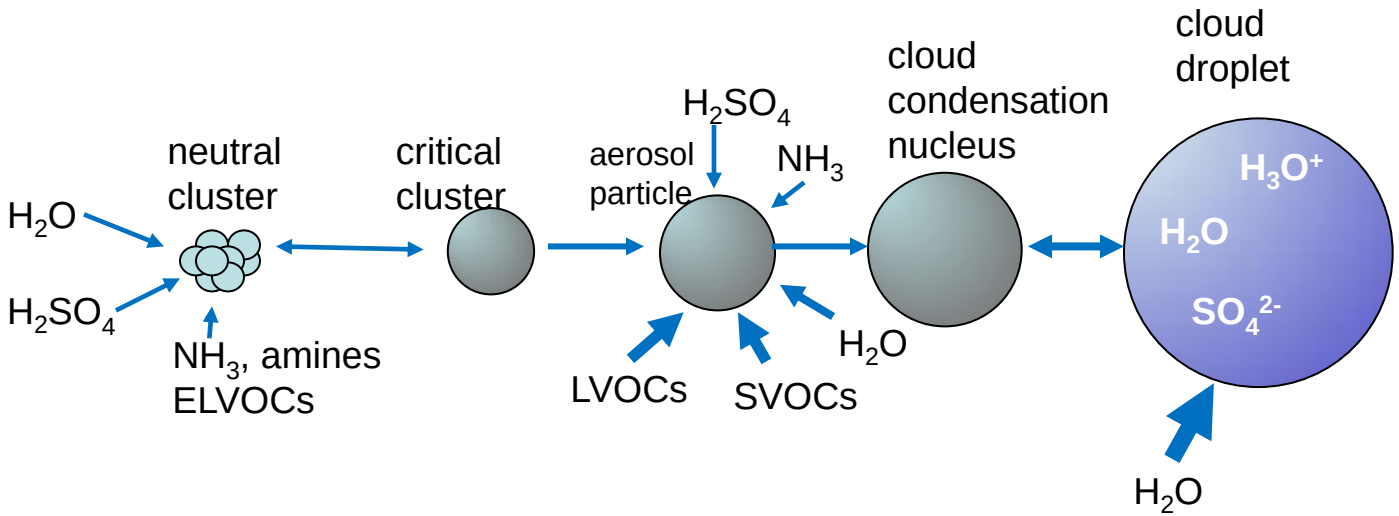




Courtesy J. Kirkby

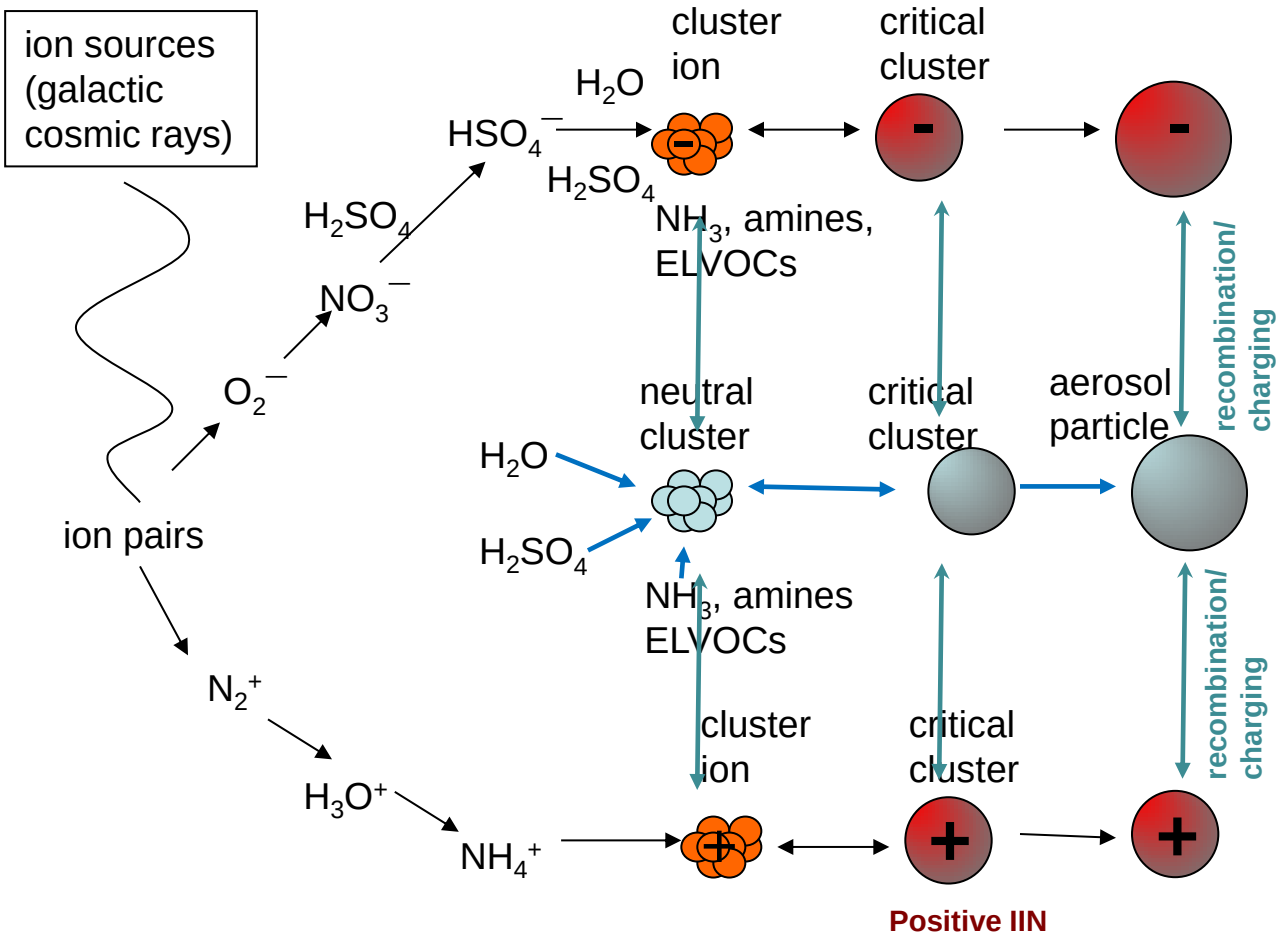
LOSU: Level of Scientific Understanding

Nucleation processes



0.3 nm 1 nm 100 nm > 1 µm

Ion-induced nucleation



0.3 nm 1 nm 100 nm > 1 µm

CLOUD 2012:
The white spots on the nucleation map are now being filled!

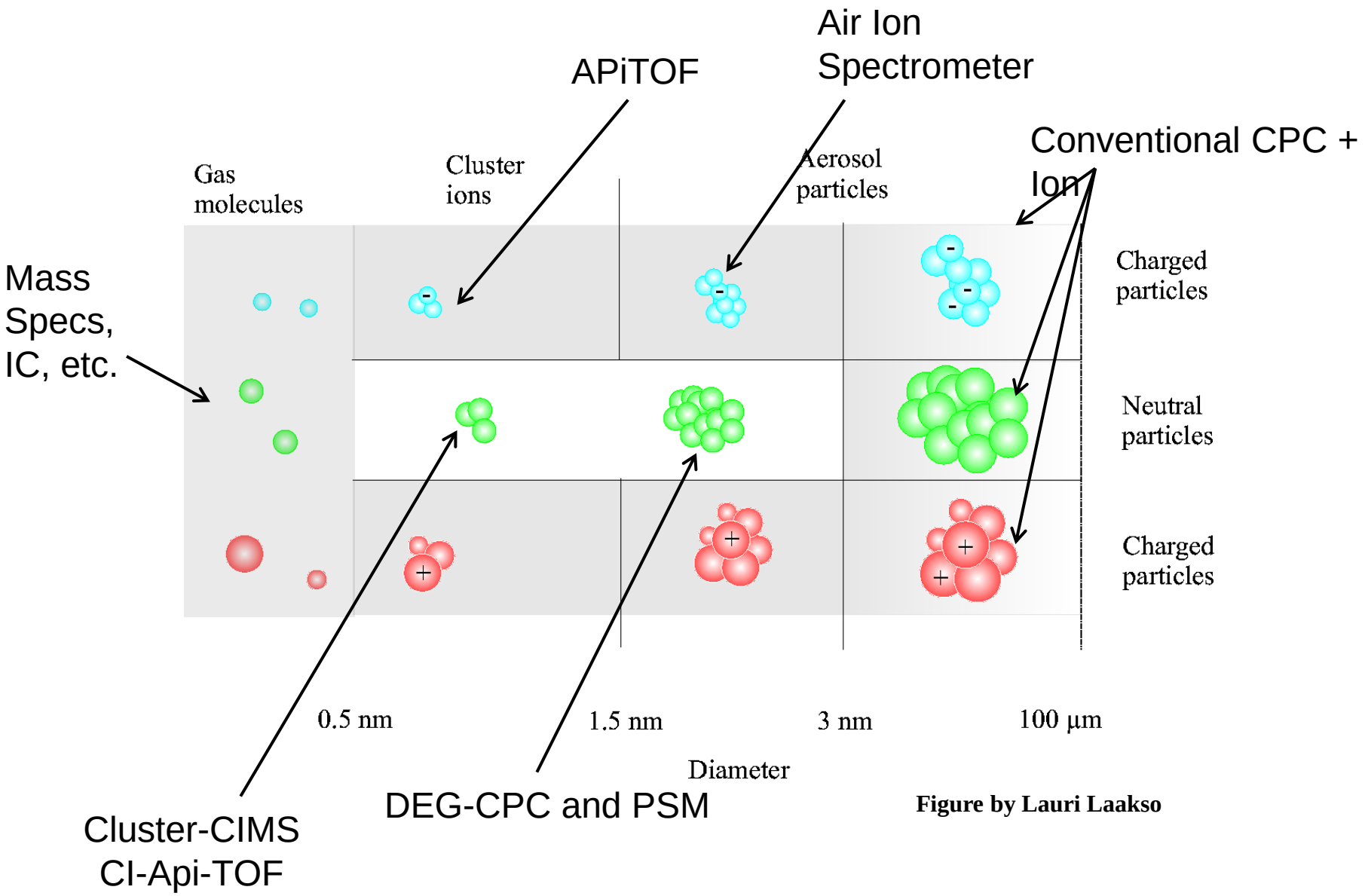
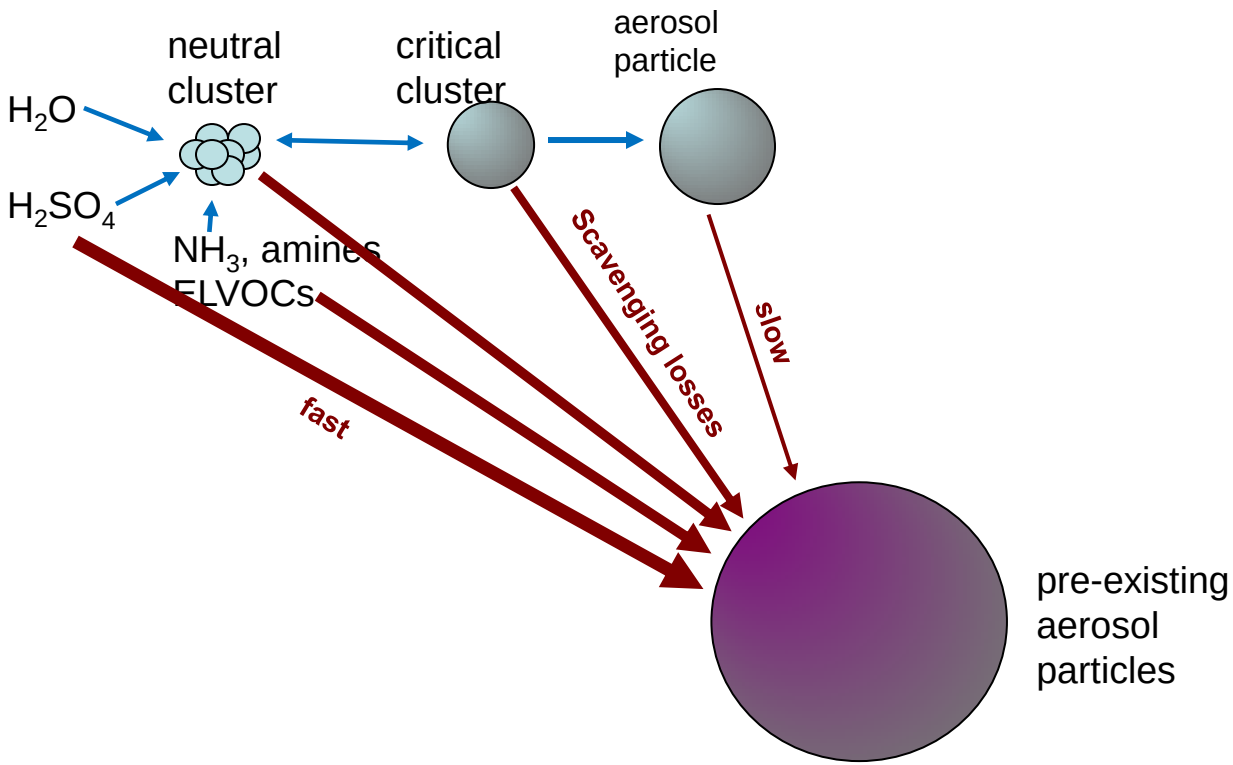
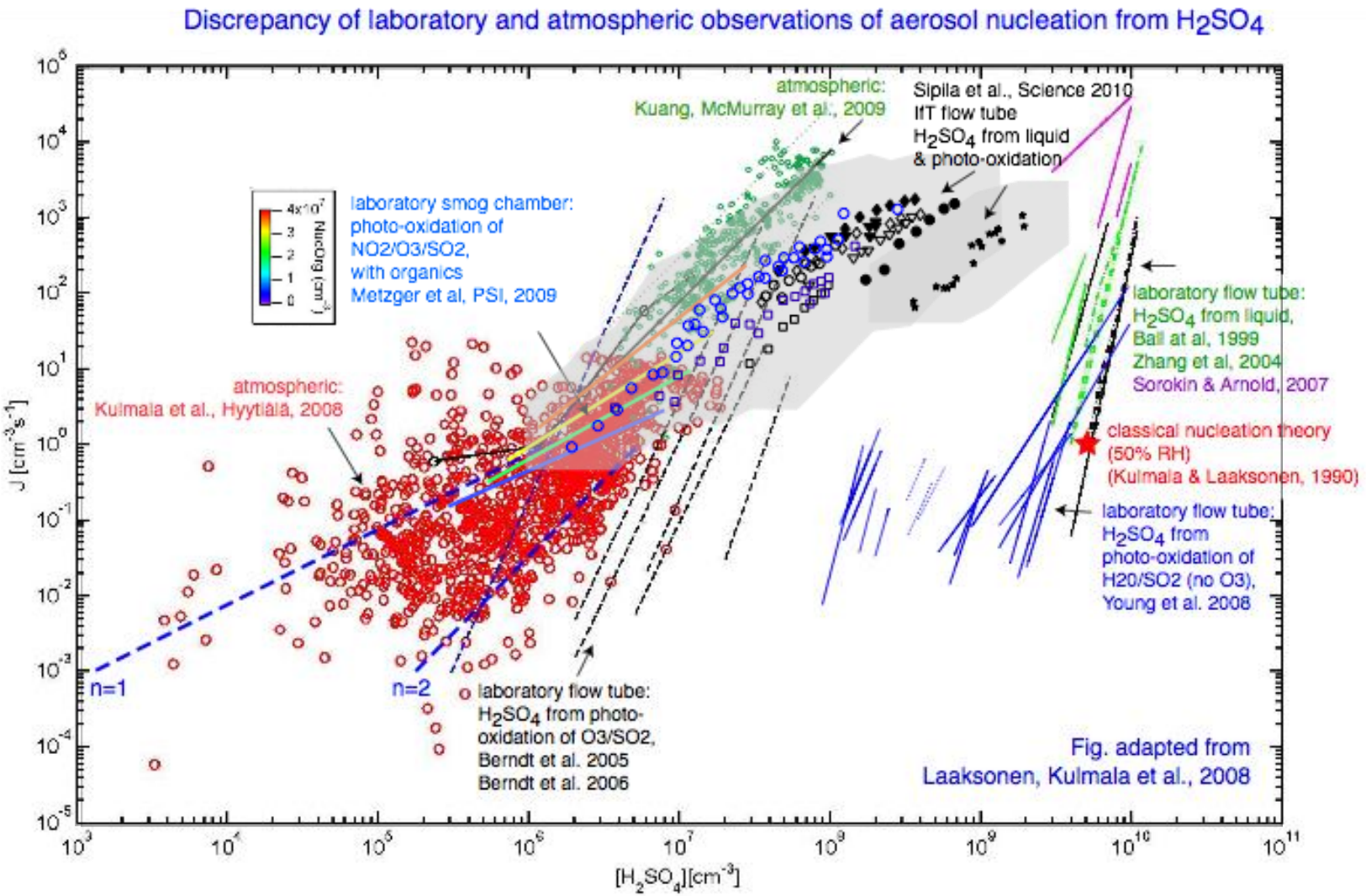


Figure by Lauri Laakso

Competition of nucleation and scavenging: “grow or die!”

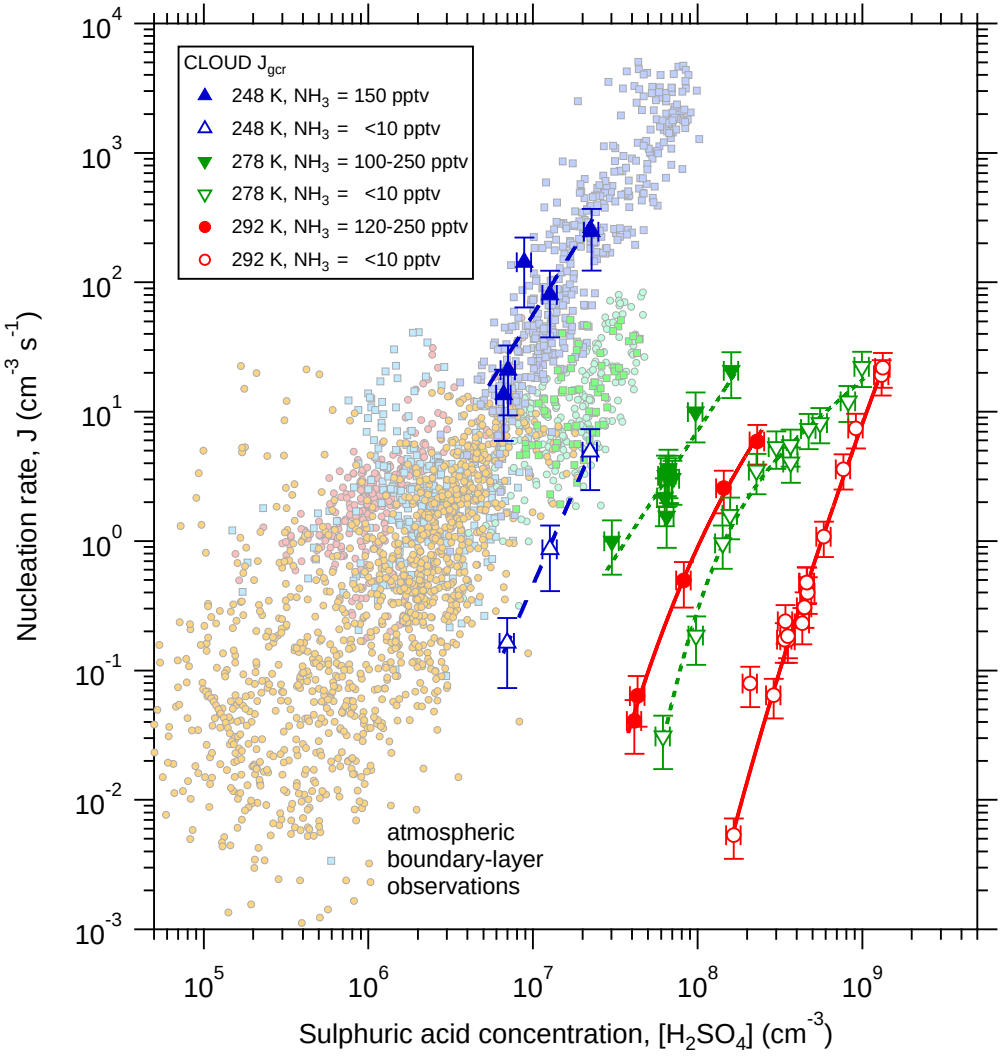


Previous measurements of nucleation rate vs [H_2SO_4]



- Many orders of magnitude discrepancy between previous experiments!
- Slopes (~ critical cluster size) between 1 and >10

CLOUD nucleation rate vs [H_2SO_4]



(Kirkby et al., Nature, 476, 2011)

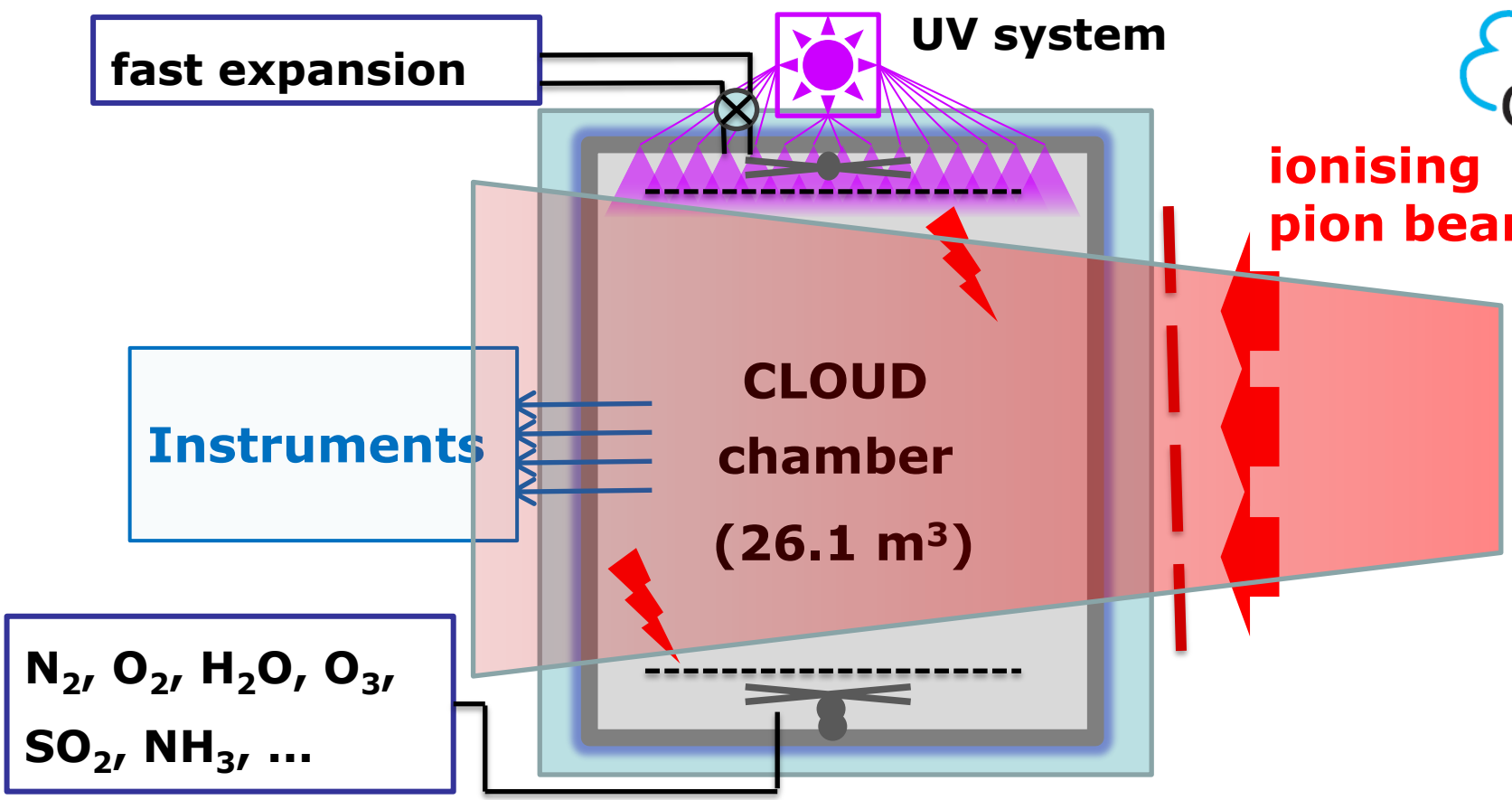
- Boundary layer nucleation cannot be explained by H_2SO_4 + NH_3 + ions (factor 10-1000 too slow)

CLOUD

Goals:

- **Understand and quantify microphysics and chemistry of of nucleation, growth and cloud-aerosol interaction**
- **Study full range of atmospheric conditions**
  - ✓ **with clean chamber (<pptv contamination of key vapours, well-controlled conditions**
  - ✓ **using a suite of state-of-the-art instrumentation**
  - ✓ **ion-induced processes: CERN beam to simulate GCRs**
- **Parameterize results and feed into models (scales…)**

- **7 experimental phases since Dec 2009, > 1150 nucleation measurements**
- **In 2012: CLOUD 6 (June), CLOUD 7 (Oct-Dec)**



- ✓ Electropolished stainless steel surfaces, no teflon…)
- ✓ Temperature range +40°C → -80°C (stabilized to <0.05°C)
- ✓ Ultra-pure gas supplies
- ✓ Exposed to adjustable 3.5 GeV/c π+ beam from CERN PS
- ✓ Homogeneous UV illumination by 250 quartz fibres
- ✓ Continuously mixed by 2 fans
- ✓ 60 kV clearing field
- ✓ Contents analysed by instruments via sampling probes
- ✓ Can be operated as an expansion chamber for droplet & ice activation

ATMOSPHERIC SCIENCE

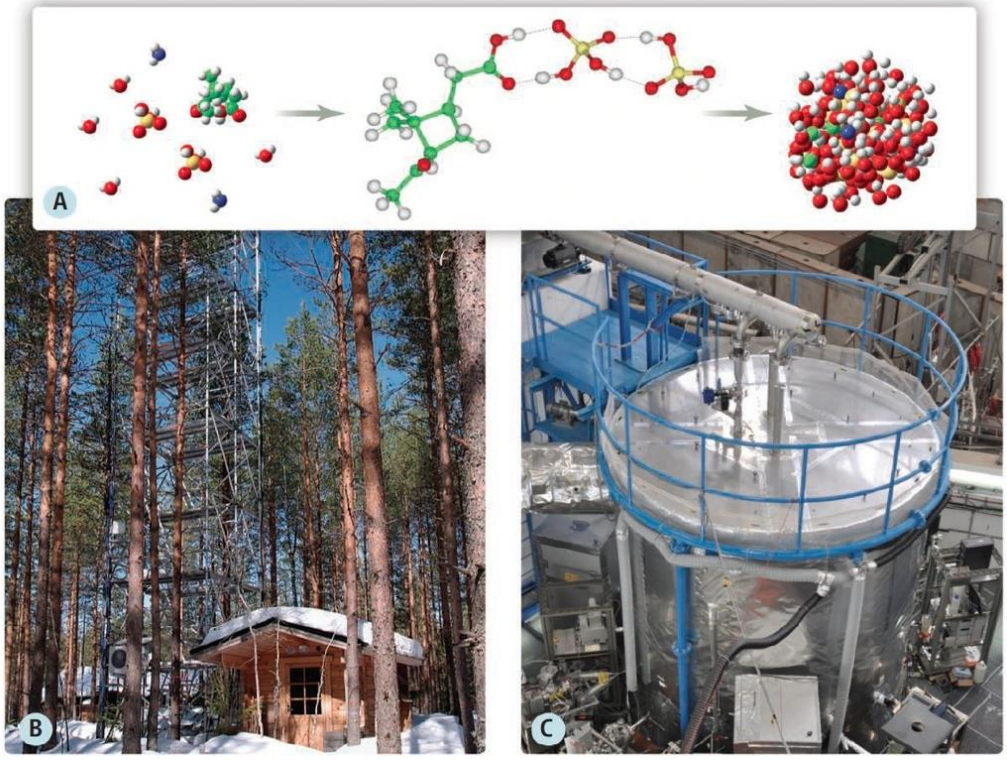
The Aerosol Nucleation Puzzle

Field studies in a boreal forest and laboratory experiments reveal how atmospheric aerosols are formed from gas molecules.

Meinrat O. Andreae

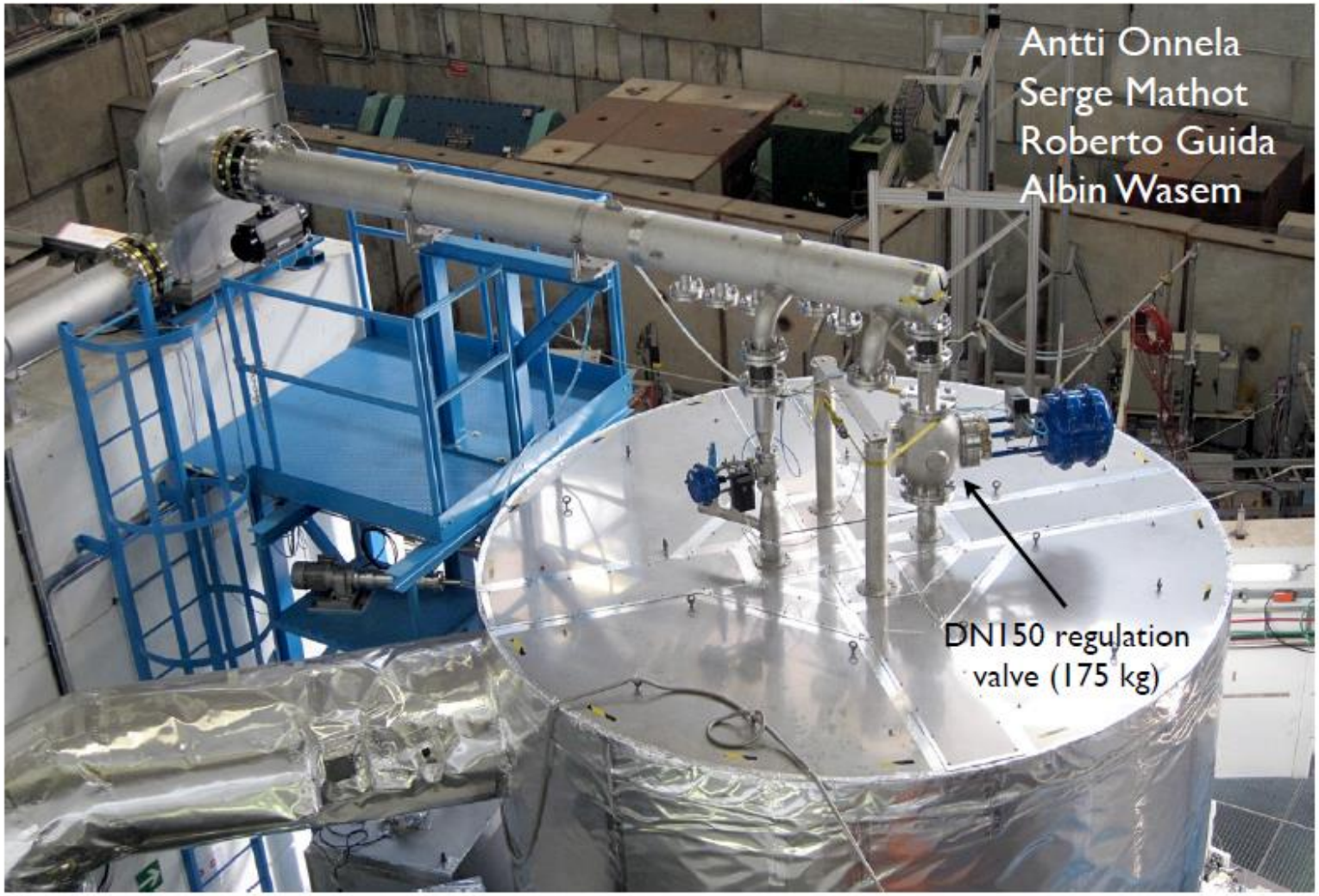
www.sciencemag.org SCIENCE VOL 339 22 FEBRUARY 2013

Published by AAAS



may be able to provide some answers. In the most sophisticated laboratory study of the nucleation process so far, researchers brought together H_2SO_4 and organic molecules in the CLOUD (Cosmics Leaving Outdoor Droplets) chamber at CERN at environmentally relevant concentrations (see the figure, panel C). They followed, step-by-step, how molecular species come together in orderly progression and grow into clusters (*3*). The interplay of field and laboratory studies must continue in order to improve our understanding of the interface between molecules and particles.

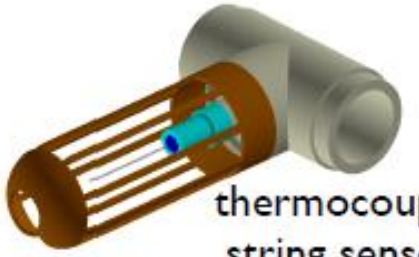
CLOUD6 (Jun12)



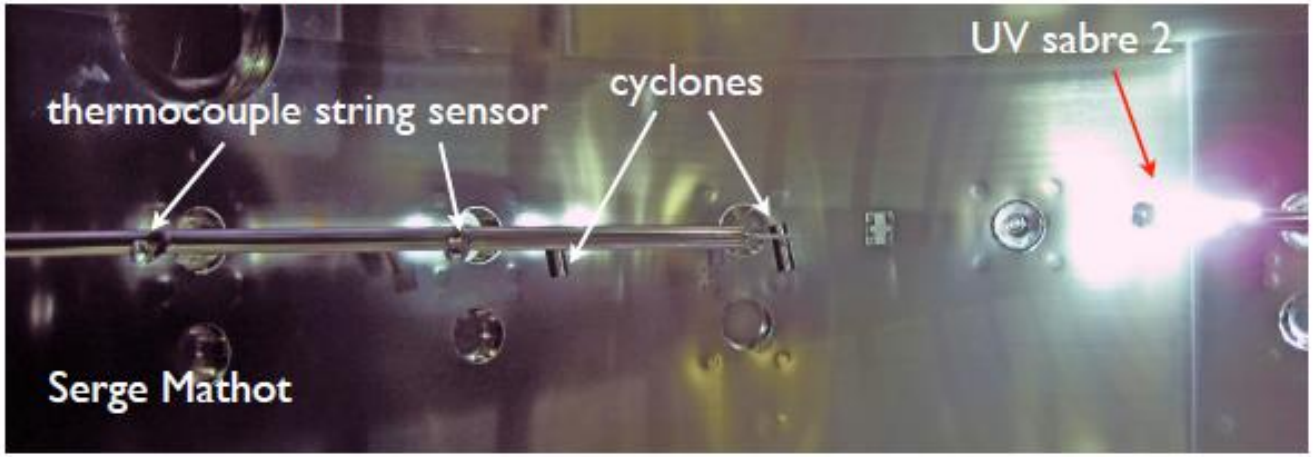
- Commissioning of fast expansion system
- Commissioning of various new instruments for characterization of cloud droplets, ice particles, cloud condensation nuclei and ice nuclei.

CLOUD6 sampling instruments

Gas:		
	CIMS H2SO4	Frankfurt
	IC NH3	PSI
	O3	EMPA
	SO2	Frankfurt
RH:		
	DP 1 stage (heated probe)	Leipzig
	DP 2 stages (heated probe)	CERN
Ions:		
	NAIS	Helsinki
ICE/CCN:		
	Welas	KIT
	PPD-2	Hertford/ Manchester
	3V-CPI	Manchester
	CAPS-CAS-DPOL	Manchester
	CCNC	Leipzig
Particles:		
	SMPS (with cyclone)	PSI
	CPC 2.5	PSI
	charge fraction CPC	PSI
	AMS	PSI

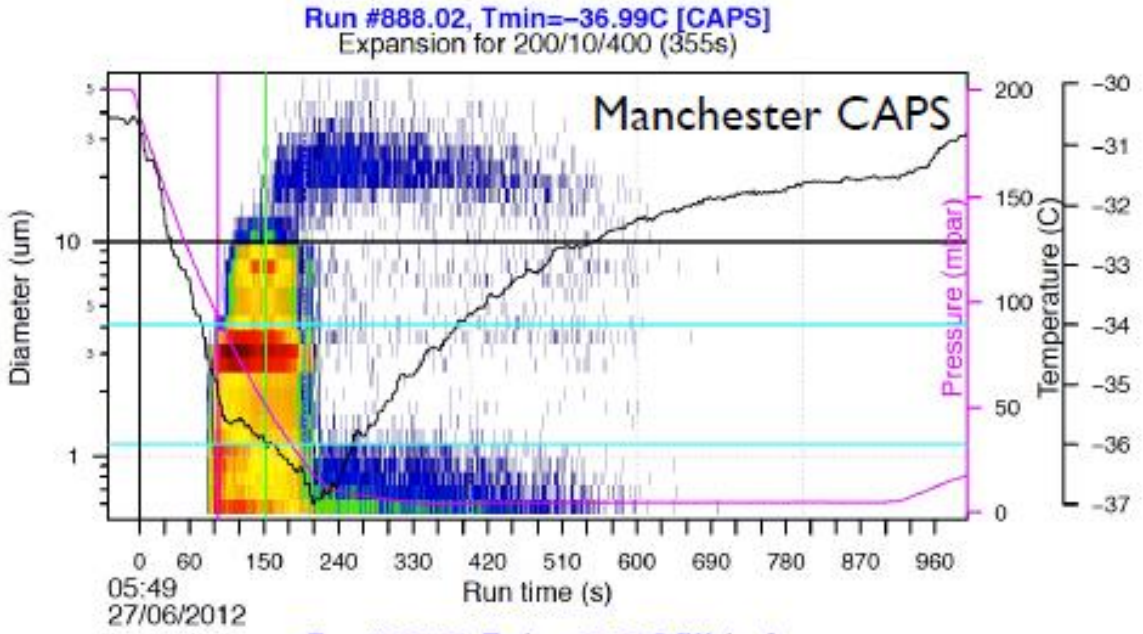


thermocouple string sensor



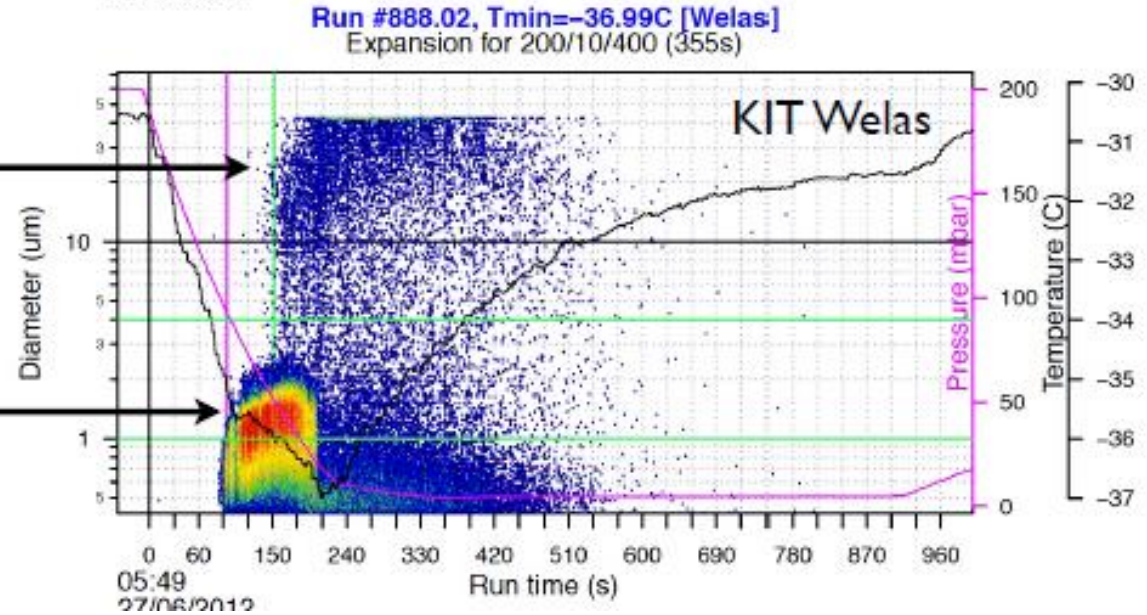
- Also new CLOUD components:
  - thermocouple string
  - cyclone sampler
  - CIGAR
  - UV sabre 2

CLOUD6 liquid and ice clouds near -37C



T = -30.2C

Gary Lloyd



ice particles

liquid droplets

Thea Schmitt

CLOUD7 (Oct-Dec 2012)

PTRTOF
(U Innsbruck)

LDT (PSI)

SMPS (PSI)
DP (CERN)

CPC2.5 (PSI)
CPC 3010 (U Frank.)

TDCIMS (NCAR)

IMS-TOF (U Helsinki)

APITOF- (U Helsinki)

APITOF+ (PSI)

SO2 (U Frankfurt)
O3 (EMPA)

2xPSM (U Helsinki)
2xDEGCPC (U Frankfurt)

CI-APITOF (U Helsinki)

CI-APITOF (U Frankfurt)

LOPAP (PSI)
IC (PSI)

CIMS (U Frankfurt)

rDMA (Caltech/U HEL)
HTDMA/OTDMA (UEF)

NAIS (U Helsinki)

Σ: 7 ToF + 1 Quad + 1 ion mobility spec

CLOUD7 experiments (1Oct - 13Dec12)

	Chamber clean (Karcher + 100C cycle) + monitor bgds with PTRTOF
1	Chamber contaminants: add in stages: dry air, H2O, O3, SO2
2	Binary H2SO4 nucleation at contaminant NH3 (<10 ppt)
3	NH3-H2SO4 nucleation at 20-40 pptv NH3
4	CIMS calibration
5	Effect of DMA on CIMS H2SO4 measurement (1SA & 2SA)
6	Ion-induced nucleation vs diffusion charging for DMA-H2SO4 nucleation
7	Neutral cluster distribution for DMA-H2SO4 nucleation (& DMA hi GR)
8	Pure ozonolysis of alpha-pinene (high DMA) - suppress OH
	Chamber clean (100C cycle only)
9	Pure ozonolysis of alpha-pinene (low NH3/DMA) - suppress OH
10	Pure OH oxidation of alpha-pinene (lower NH3/DMA) - no O3
11	Pure OH oxidation of alpha-pinene (lowest DMA) - no O3
	Chamber clean (100C cycle only)
12	Nucleation of alpha-pinene/DMA without H2SO4 (no SO2)
13	Hyytiälä simulation (alpha-pinene + 1. contaminant NH3/DMA 2. low NH3/DMA)
	Chamber clean (100C cycle only)
14	Beam production of H2SO4 (+parasitic H2SO4 calibrations of CIMS & CI-APIs)
15	DMA nucleation at 1ppt (+parasitic rDMA+DEG2.0 calibration)

- Most complex series of experiments carried out so far with CLOUD
- New gases:
  - H_2 (0.1%)
  - HONO (0-1000 ppt)
  - alpha-pinene (0-1200 ppt)
- Very successful run; all experimental goals in table achieved

Next runs

CLOUD8 (Oct 2013-Dec 2013), CLOUDy experiments (with GCR)

CLOUD9 (spring 2014), aerosol nucleation and aerosol processes

- Further clarification of competing chemical systems (ions, DMA, NH3, organics…)
- Role of water vapour for nucleation and growth
- Identification of multi-component clusters
- Early growth rates
- Nucleation process molecule-by-molecule
- Parametrizations for models → assessments of climate impacts
- Role of GCR ionisation for CCN activation, ice nuclei activation, riming, formation of precipitation

Summary

- CLOUD allows *precise and reproducible* measurement of aerosol nucleation and growth rates over *full range of atmospheric conditions*. (cleanliness of chamber, observation of nucleation molecule-by-molecule, all conditions well-controlled - including ionisation, ...)

- Role for climate includes *anthropogenic* (H_2SO_4, NH_3 levels…) and *natural* variable factors (ionization by GCRs)

- *Ion* clusters and *neutral* clusters observed during nucleation at atmospherically-relevant concentrations (time-resolved).
  → chemistry and mechanisms
  → fundamental understanding of atmospheric nucleation and growth

- CLOUD measurements are being incorporated into regional and global models to assess impact on clouds and climate. Manuscript of first global model results to be submitted within few weeks:
  Dunne et al, Impact of cosmic rays on global aerosol, clouds and climate

Future CLOUD plans

1. Resolve and quantify the fundamental physical and chemical processes involved in the formation and growth of cloud-active aerosols and the interaction of these aerosols with clouds.

2. Measure the effects of cosmic rays on aerosols and clouds, and settle the question of whether cosmic rays exert a climatically-significant effect on climate and, if so, under what conditions.

3. Develop a mechanistic understanding of the underlying physico-chemical processes and incorporate them into global models simulating the behaviour of ions, aerosols and clouds under realistic meteorological conditions.

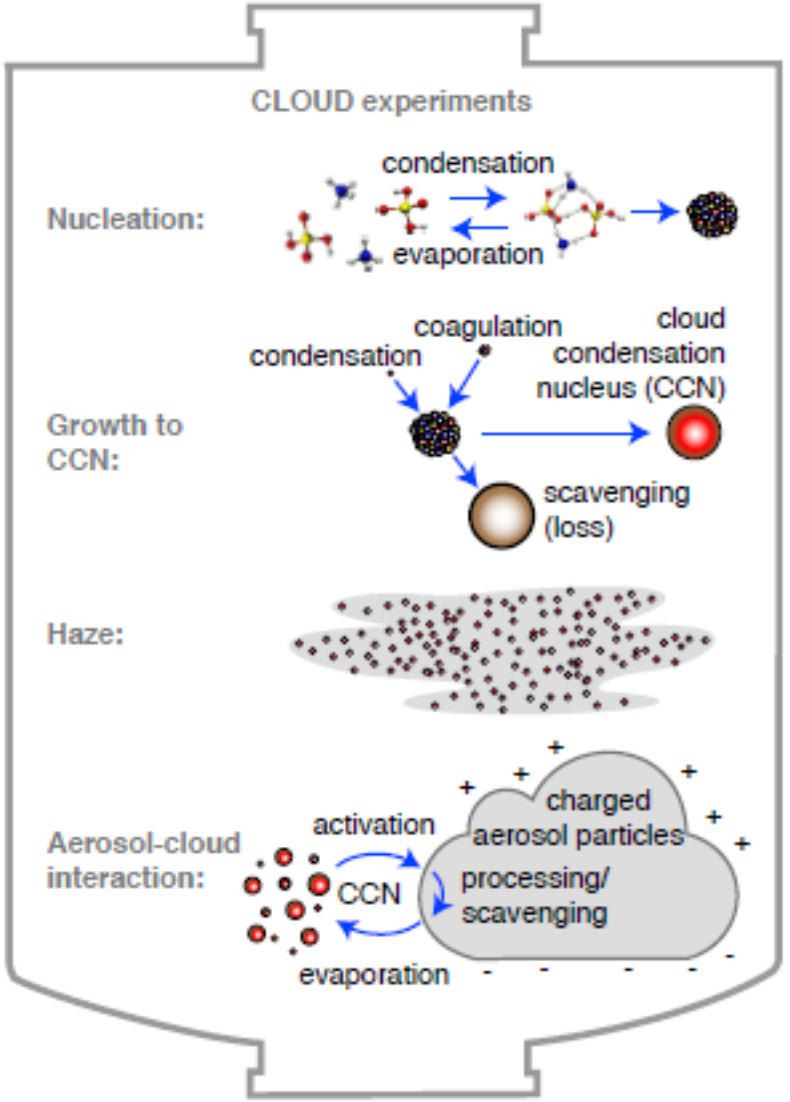
4. Reduce the uncertainty in the climate impact of aerosols and their interaction with clouds, leading to more robust climate projections.

→ CLOUD aims to resolve the fundamentals of ion-aerosol impact on clouds and climate

→ Measurement of 1-2 vapours at a *single* temperature and relative humidity requires a full 8-week run. So the future CLOUD experimental programme is estimated to require 10 years, even with careful limitation of the range of experimental variables

Future work

- Formation of cloud-active aerosols
- Aerosol-cloud interactions
- Aerosol-cloud modelling and climate impact

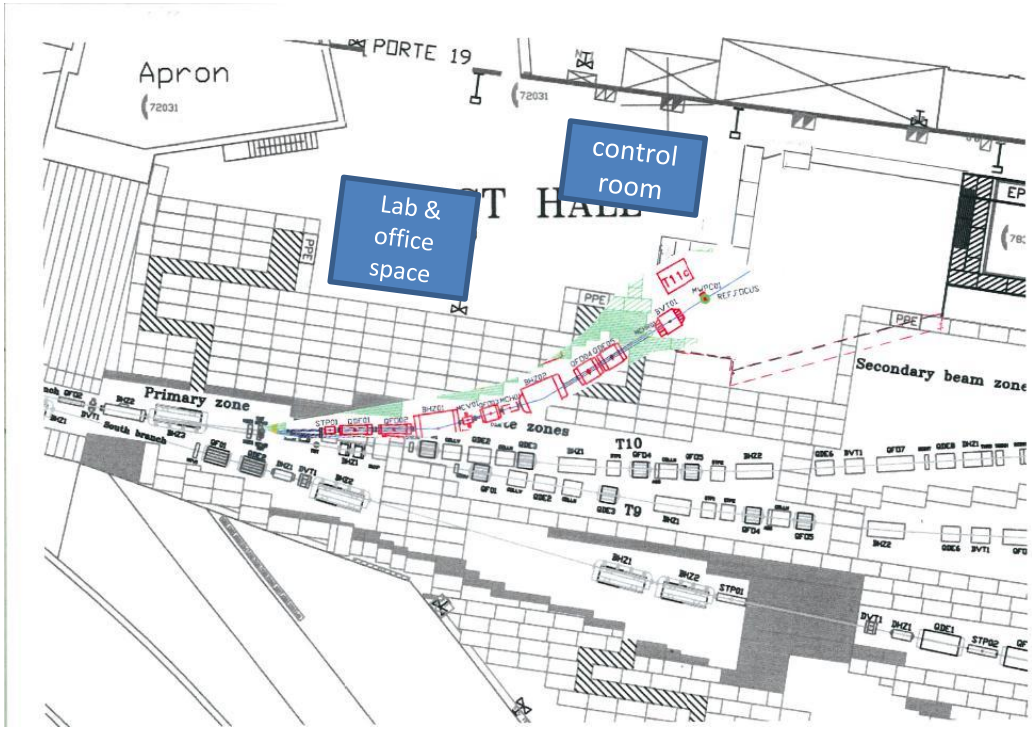


Present and future CLOUD experiments on ion-aerosol-cloud processes.

Request for office and lab space

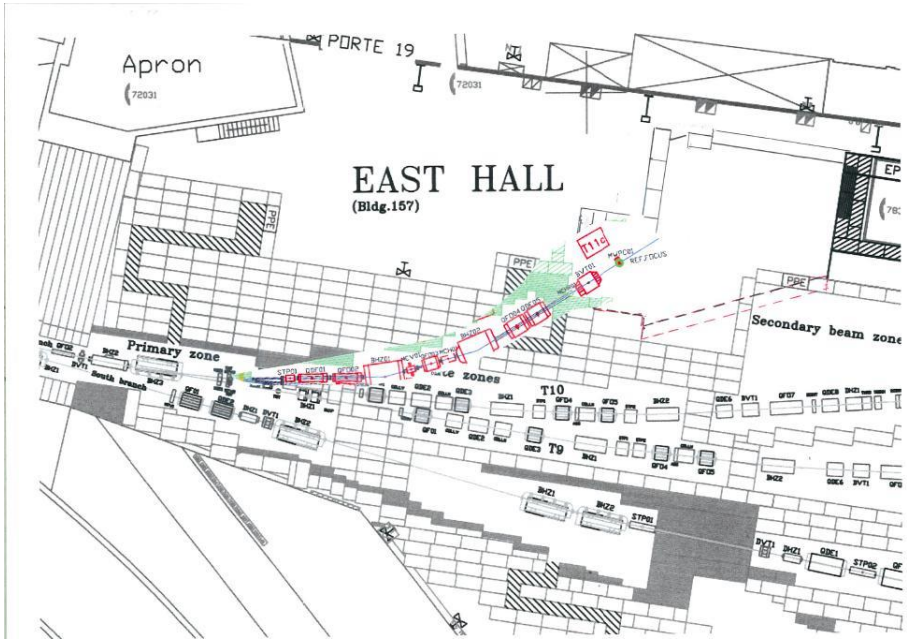
- CLOUD has typically 20-30 persons present during experiments.
- CLOUD control room is over-crowded, too loud, not air-conditioned; additional office space/meeting room and room for instrument testing & repairs for CLOUD is urgently needed.

  → Request for office & lab space nearby and enlarged control room, at least double container, a/c, 30 m^2.



Request for continuation of T11 beamline

- CLOUD experimental programme likely to extend well beyond planned upgrade of East Hall beamlines
- CLOUD requests that T11 beamline be retained in the new East Hall configuration (ideally with 1-2 m extra space in T10 direction):
  - Maximise CLOUD efficiency and output
  - Maximise availability of T9 and T10 beamlines for test-beam users (if CLOUD shares T10 then a) less beam-time available for other users and b) no access to T10 zone while CLOUD using beam).
  - Present T9+T10+T11 users would completely saturate new T9+T10 beamlines



Request for technical support

- Expert CERN technical engineering support has been key to CLOUD's success and will continue to be so in future
- Present CERN staff support for CLOUD totals 1.8 FTE
- After July 2013 this falls to 0.8 FTE (JK retirement)

- The CLOUD collaboration requests that CERN maintains 1.8 FTE support for CLOUD after July 2013 with the full-time assignment of Serge Mathot to CLOUD. Serge has made extensive contributions to the experiment so far and his knowledge of the experiment and his skills are crucial to CLOUD's future success.

CLOUD MEETINGS IN 2012

- **CLOUDy workshop**, University of Manchester, 11–12 Jan. Goals and planning for CLOUD6.
- **CLOUD4–5 data workshop**, Hyytiälä, Finland, 27 Feb – 1 Mar. Analysis of data from CLOUD4–5.
- **Aerosol growth rate workshops**, Helsinki, 13–16 Apr and 10–12 Sep. Analysis of aerosol growth rates from CLOUD2–5.
- **CLOUD-ITN Conference**, Königstein, Germany, 22–25 May.
  Presentation and discussion of results from all CLOUD runs, together with 30 invited external experts, followed by 1-day CLOUD collaboration meeting.
- **CLOUD collaboration meeting and CLOUD-TRAIN kickoff**, CERN, 15–16 Oct.
  CLOUD collaboration meeting and startup of EU FP7 CLOUD-TRAIN Marie Curie network.

Invited talks 2012-2013 (selection)

Deutsche Physikalische Gesellschaft:
Symposium on solar influence on Earth‘s climate
28 Feb 2013, Jena, Germany
invited talk, Joachim Curtius

ICNAA 2013
International Conference on Nucleation and Atmospheric Aerosols
24-28 June 2013, Fort Collins, CO
Keynote plenary speaker: Jasper Kirkby
Special Session on CLOUD (>25 contributions)

Gordon Research Conference for Atmospheric Chemistry 2013:
28 July – 2 August 2013, Snow Mountain Res., VT
Invited talk: Joachim Curtius

Many other invited talks at AGU, EGU, CAWSES, ISSI, MPG, etc.

Publications:

Kirkby, J., et al. Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation. *Nature* 476, 429–433 (2011).

Kupc, A., et al.. A fibre-optic UV system for H2SO4 production in aerosol chambers causing minimal thermal effects. *J. Aerosol Sci.* 42, 8, 532–543 (2011).

Voigtländer, J., et al. Numerical simulation of flow, H2SO4 cycle and new particle formation in the CERN CLOUD chamber. *Atmos. Chem. Phys.* 12, 2205–2214 (2012).

Bianchi, F., et al. On-line determination of ammonia at low pptv mixing ratios in the CLOUD chamber. *Atmos. Meas. Tech.* 5, 1719–1725 (2012).

Praplan, A.P., et al. Dimethylamine and ammonia measurements with ion chromatography during the CLOUD4 campaign. *Atmos. Meas. Tech.* 5, 1719–1725 (2012).

Kürten, A. et al. Performance of a corona ion source for measurement of sulfuric acid by chemical ionization mass spectrometry, Atmos. Meas. Tech., 4, 437-443, (2011).

Kürten, A. et al. Calibration of a Chemical Ionization Mass Spectrometer for the Measurement of Gaseous Sulfuric Acid, J. Phys. Chem. A., 116, 6375-6386. (2012).

Keskinen, H., et al. Evolution of particle composition in CLOUD nucleation experiments. *Atmos. Chem. Phys. Discuss.* 12, 31071–31105 (2012).

Wimmer, D., et al. Performance of diethylene glycol based particle counters in the sub 3 nm size range. *Atmos. Meas. Tech. Discuss.* 6, 2152–2181 (2013).

Almeida, J., et al. Molecular understanding of amine-sulphuric acid particle nucleation in the atmosphere. (submitted, 2013).

Schnitzhofer, R., et al. Characterisation of organic impurities in the CLOUD chamber at CERN. (submitted, 2013).

Schobesberger, S., et al. Molecular understanding of the first steps of atmospheric particle formation from sulfuric acid and large oxidized organic molecules. (submitted, 2013).

More than 25 publications in preparation…

Funding

- CLOUD-TRAIN, EU Marie Curie Initial Training Network, Oct 2012 – Sep 2016:
  12 PhD students, 3 Post-Docs, 3.8 Mio Euro

- Regular support by national funding, e.g., by German BMBF (CLOUD-12 project until 2015), Swiss National Science Foundation, the Academy of Finland Center of Excellence program, additonal national funding agencies…

- Proposal for ERC Synergy grant submitted in Jan 2013.

- CLOUD MoU signed by 19 CLOUD partners (Apr 2012) and CLOUD Financial Review Committee established (next meeting Oct 2013). CLOUD collaboration institutes pay annual CF contribution to fund CLOUD M&O costs. CERN support is gratefully acknowledged.

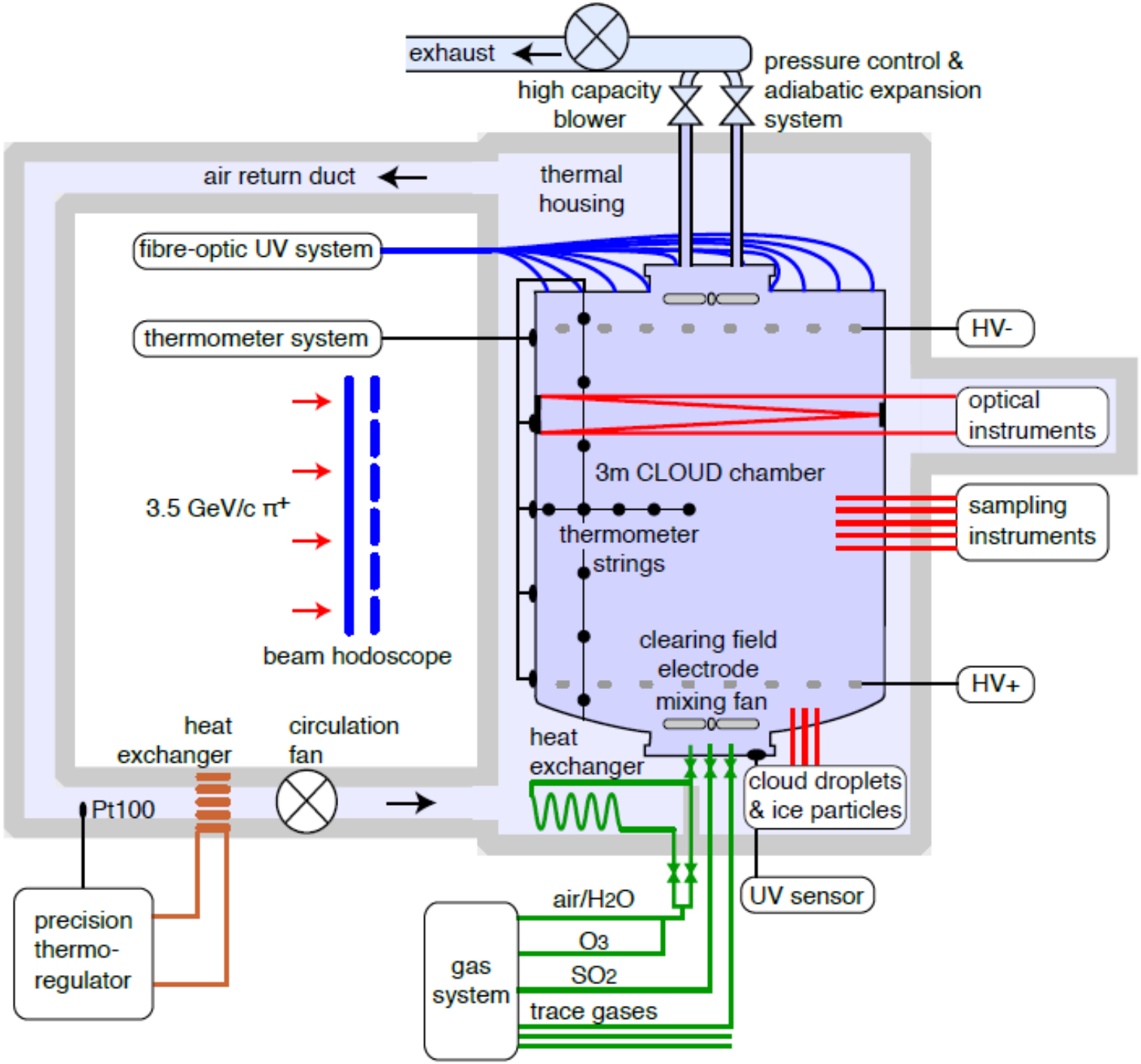
MARIE CURIE ACTIONS

GEFÖRDERT VOM
Bundesministerium
für Bildung
und Forschung

CERN

Acknowledgements

We would like to thank CERN PH-DT, EN-MME, EN-MEF and TE-VSC for their excellent support of CLOUD and, in addition, to thank the CERN PS machine team and the PS Coordinator for their strong support of CLOUD and for efficient operation of the PS!


exhaust
high capacity blower
pressure control & adiabatic expansion system
air return duct
thermal housing
fibre-optic UV system
thermometer system
HV-
optical instruments
3m CLOUD chamber
3.5 GeV/c π+
sampling instruments
thermometer strings
beam hodoscope
clearing field electrode
mixing fan
HV+
heat exchanger
circulation fan
heat exchanger
cloud droplets & ice particles
Pt100
UV sensor
precision thermo-regulator
air/H2O
O3
SO2
gas system
trace gases

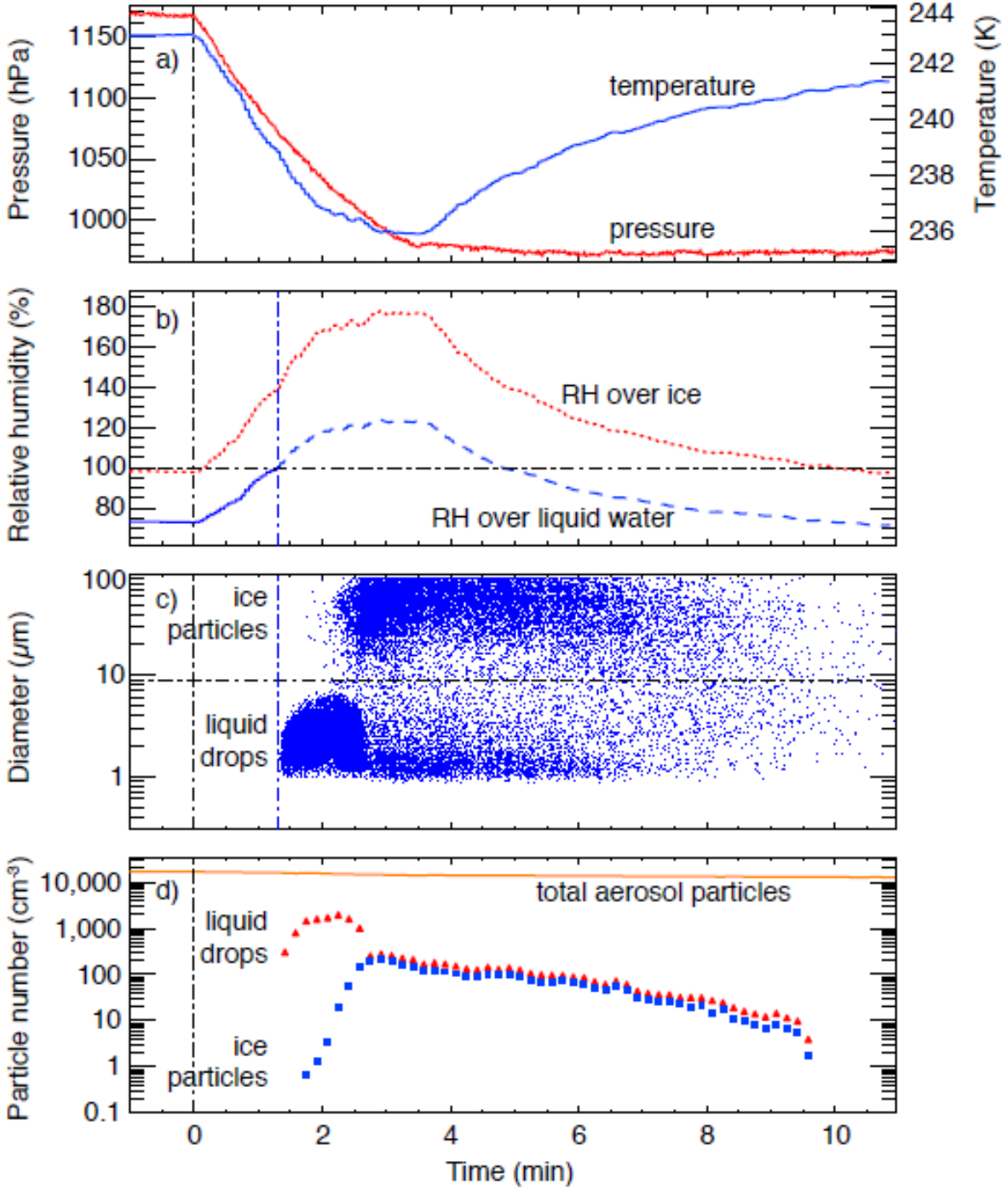

a)
Pressure (hPa)
1150
1100
1050
1000
244
242
240
238
236
Temperature (K)
temperature
pressure
b)
Relative humidity (%)
180
160
140
120
100
80
RH over ice
RH over liquid water
c)
Diameter (μm)
100
10
1
ice particles
liquid drops
d)
Particle number (cm⁻³)
10,000
1,000
100
10
1
0.1
total aerosol particles
liquid drops
ice particles
0
2
4
6
8
10
Time (min)

Table 2: CLOUD runs at the CERN PS to investigate the influence of ionising particles on aerosol nucleation and growth, and cloud microphysics.

Run	Month	Year	Aim
CLOUD1	Nov–Dec	2009	Commissioning
CLOUD2	Jun–Jul	2010	Binary H_2SO_4, NH_3 ternary nucleation
CLOUD3	Oct–Nov	2010	Binary H_2SO_4, NH_3 ternary nucleation
CLOUD4	Jun–Jul	2011	Dimethylamine & pinanediol ternary nucleation
CLOUD5	Oct–Nov	2011	Free tropospheric binary H_2SO_4, NH_3 ternary nucleation
CLOUD6	Jun–Jul	2012	Initial cloud formation experiments
CLOUD7	Oct–Dec	2012	NH_3, dimethylamine & α-pinene ternary nucleation & growth