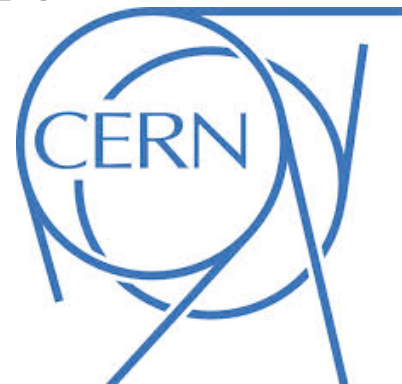


The CLOUD experiment

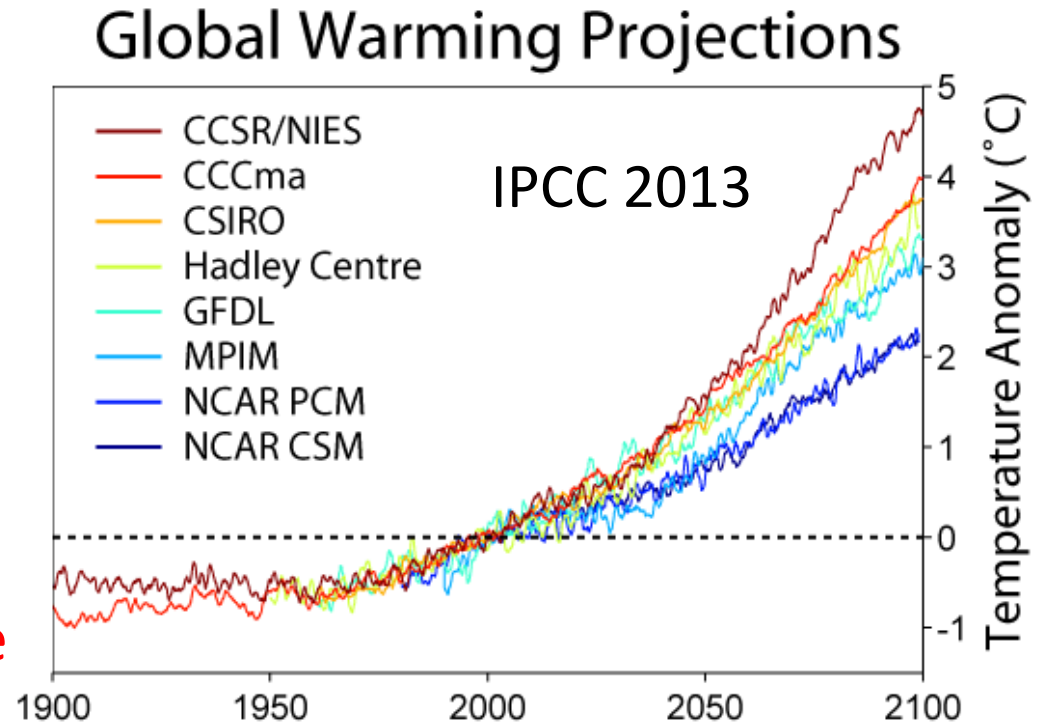
Hamish Gordon (CERN)

Louvain-la-Neuve, 3rd December 2014



Clouds and climate

- 0.5°C warming since 1960
- IPCC 2013: “It is extremely likely (95-100%) that human influence has been the dominant cause of the observed warming since the mid-20th century.”

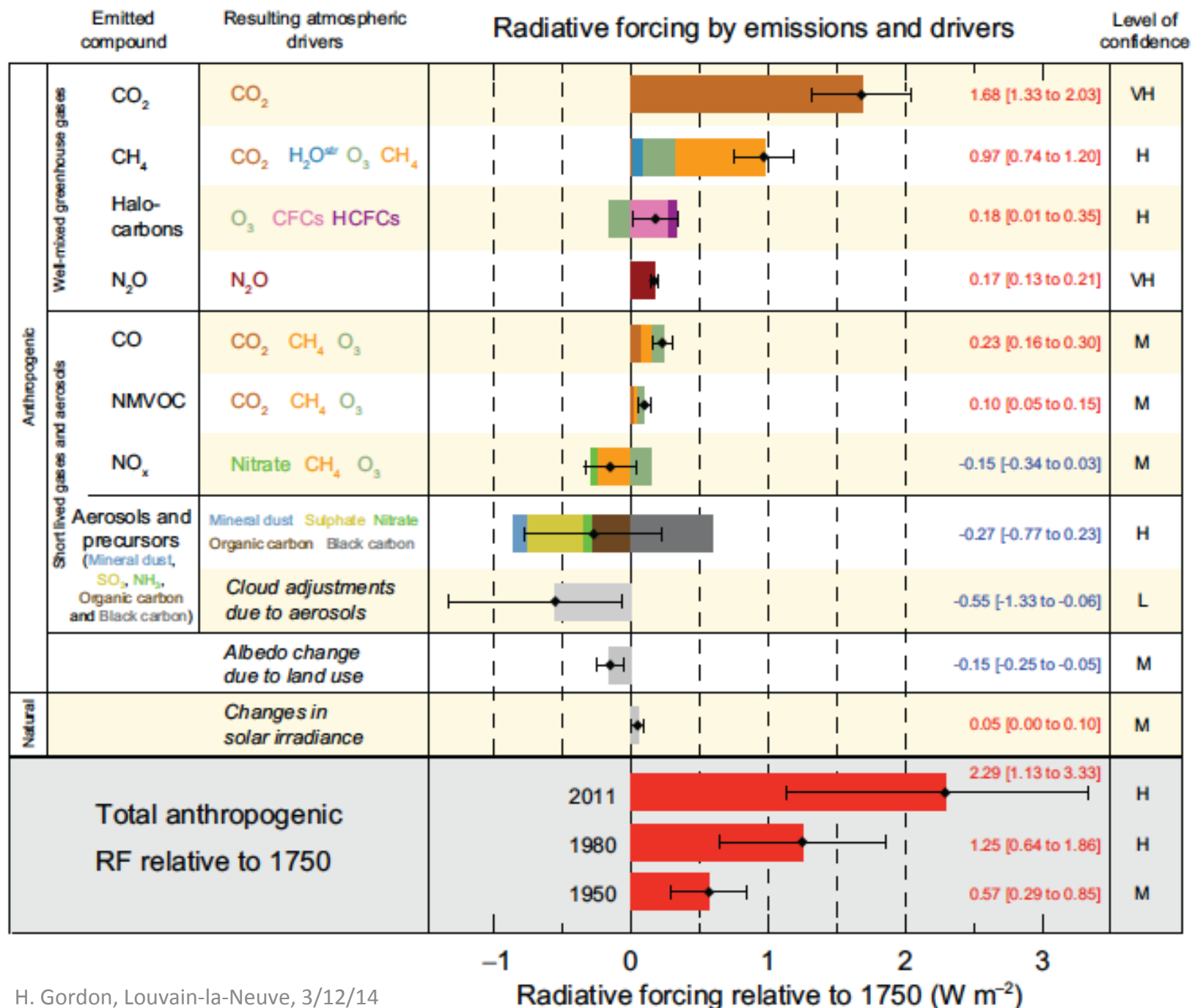


- Clouds are (more-or-less) white, the Earth is black, and ~60% of Earth's surface is covered by clouds

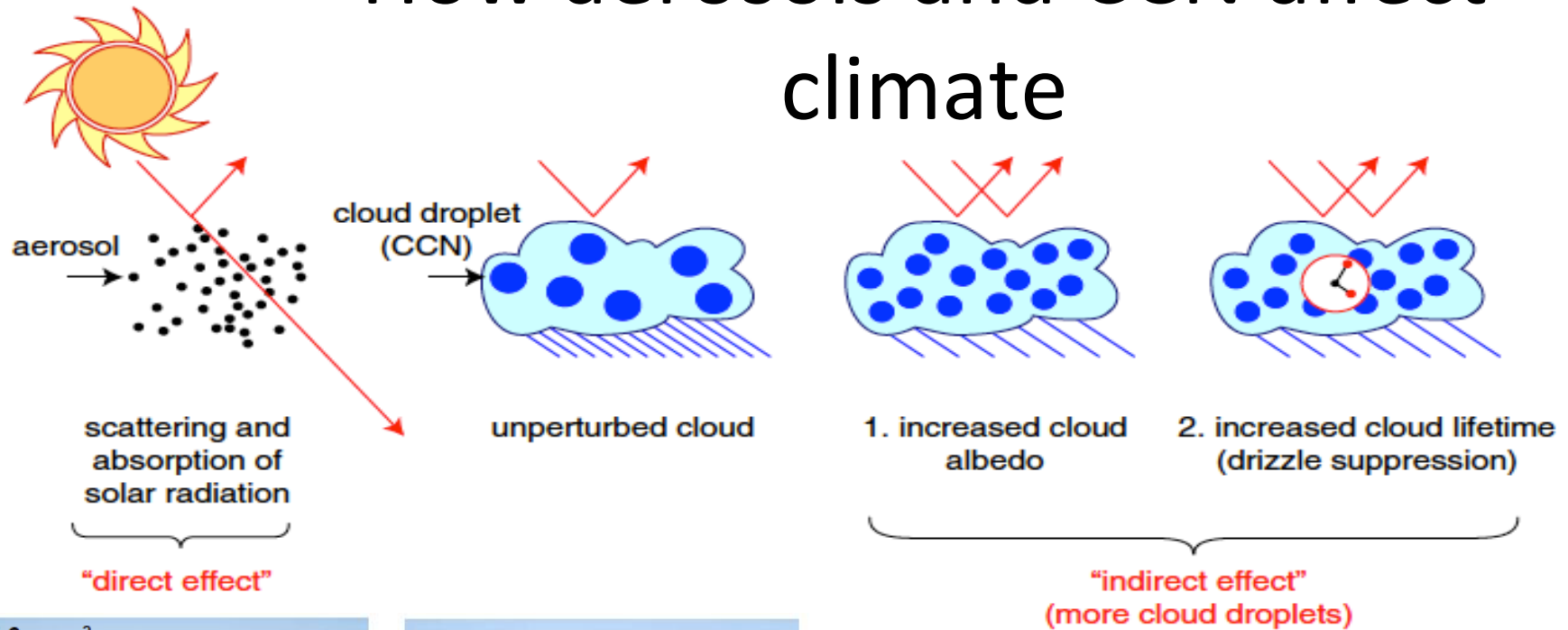
Cloud formation

- Humid air **rises** due to topography, weather systems or convection, and **cools** adiabatically
- At the dew point (100% RH) it wants to condense into clouds but **needs particulate matter** that can act as cloud condensation nuclei
- CCN/IN are mostly dust, sea salt, soot or **nucleated aerosol**





How aerosols and CCN affect climate



(a)



(b)



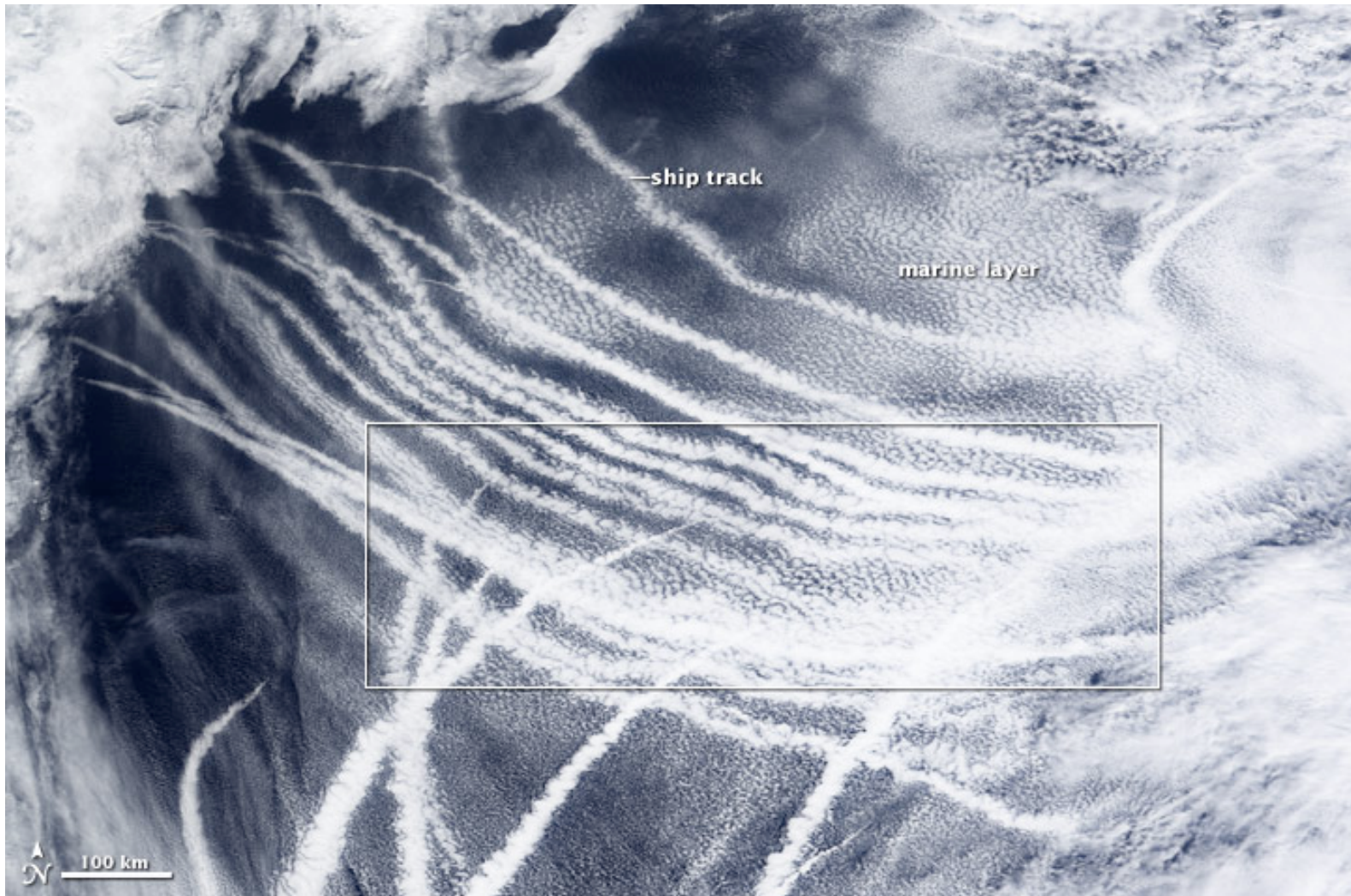
(c)



(d)

n-la-Neuve, 3/12/14

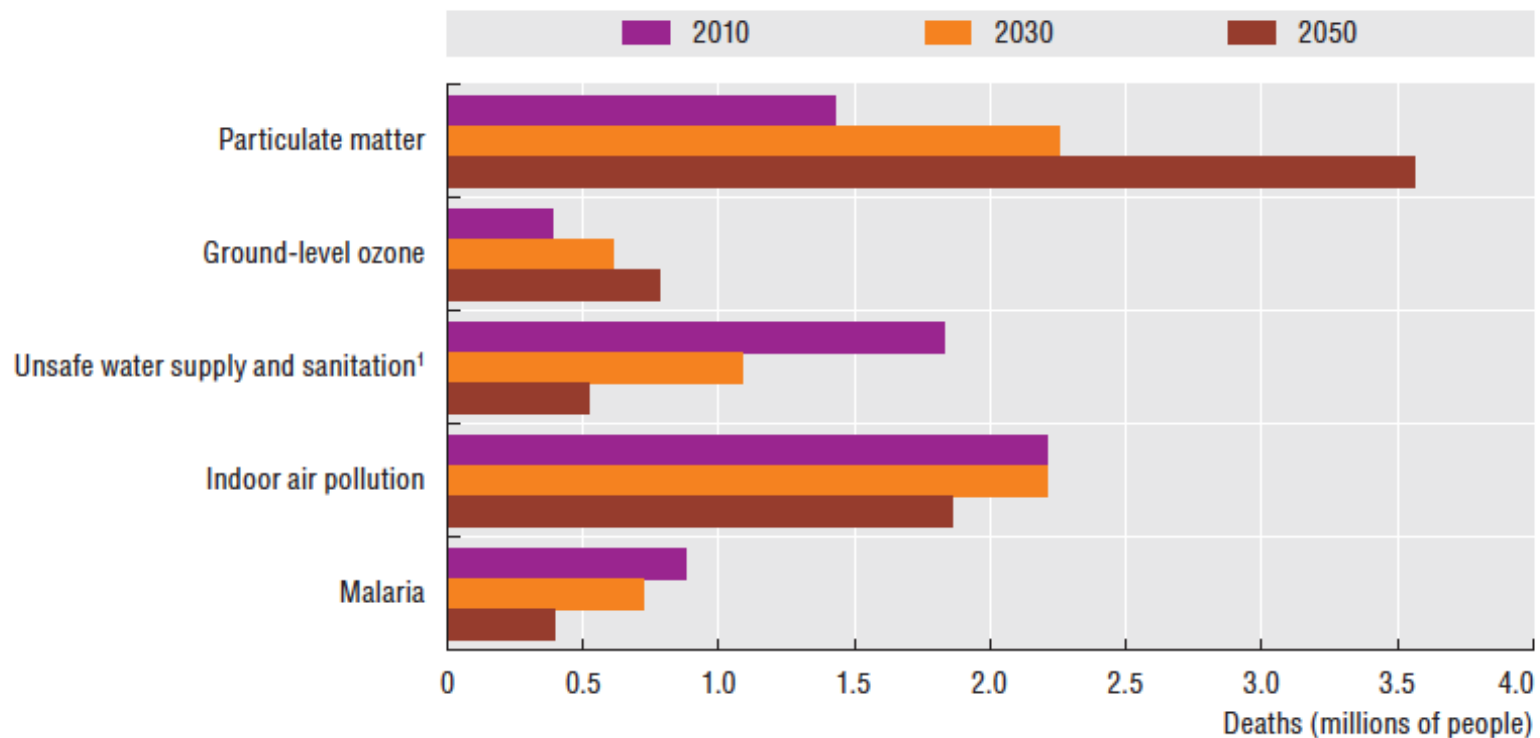
MODIS satellite, Pacific Ocean, March 4 2009



Digression...

Global premature deaths from selected environmental risks, OECD 2010 to 2050

Figure 0.4. **Global premature deaths from selected environmental risks:**
Baseline, 2010 to 2050



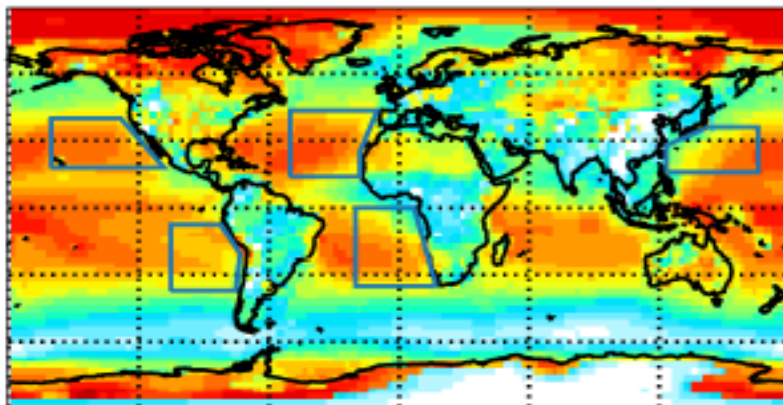
1. Child mortality only.

Source: OECD Environmental Outlook Baseline: output from IMAGE.

<http://dx.doi.org/10.1787/888932571855>

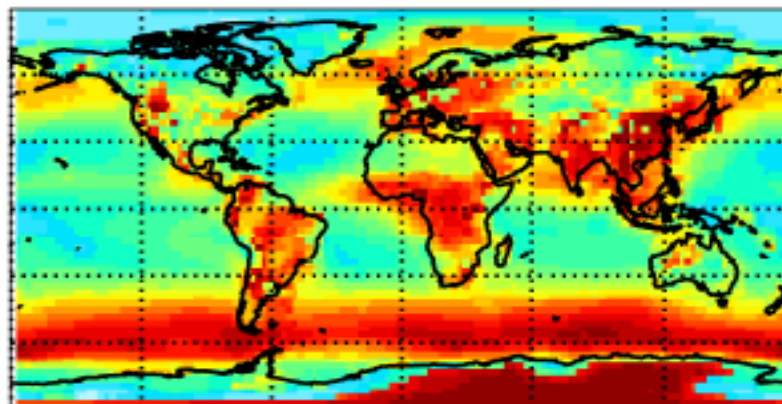
How important is nucleation?

A: CCN(0.2%) contribution from nucleation



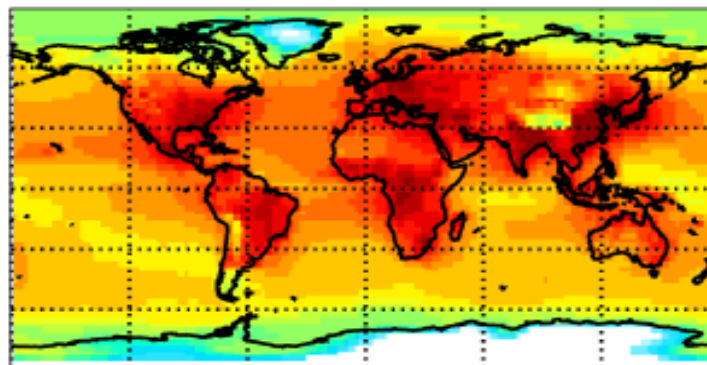
0 25 50 75 100 %

B: CCN(0.2 %) contribution from Primaries



0 25 50 75 100 %

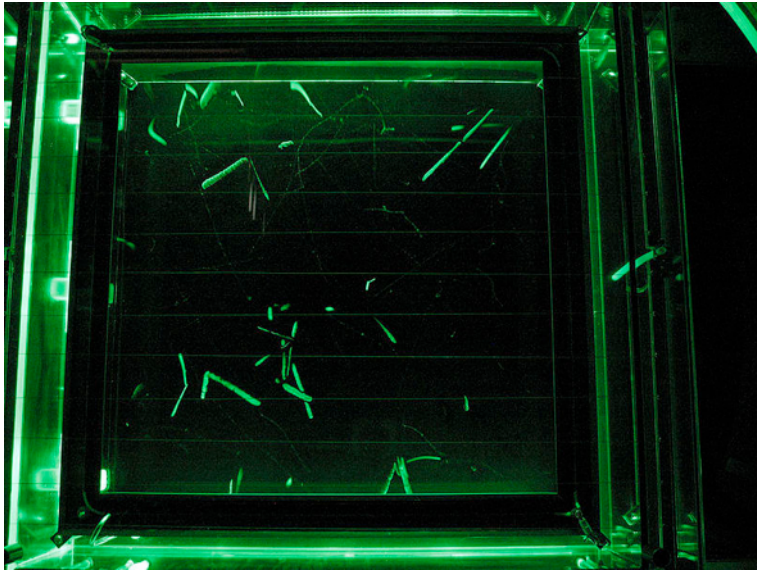
A: Total CCN (0.2 %)



1 6 44 299 2000 cm⁻³

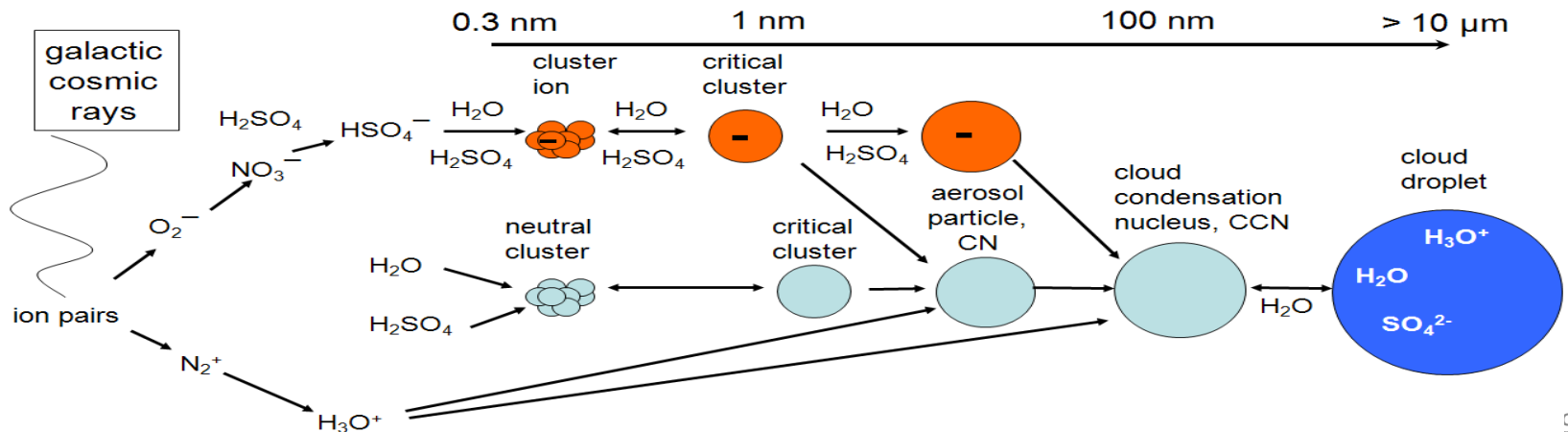
Merikanto et al, ACP 9 8601 (2009)

Could cosmic rays affect clouds?



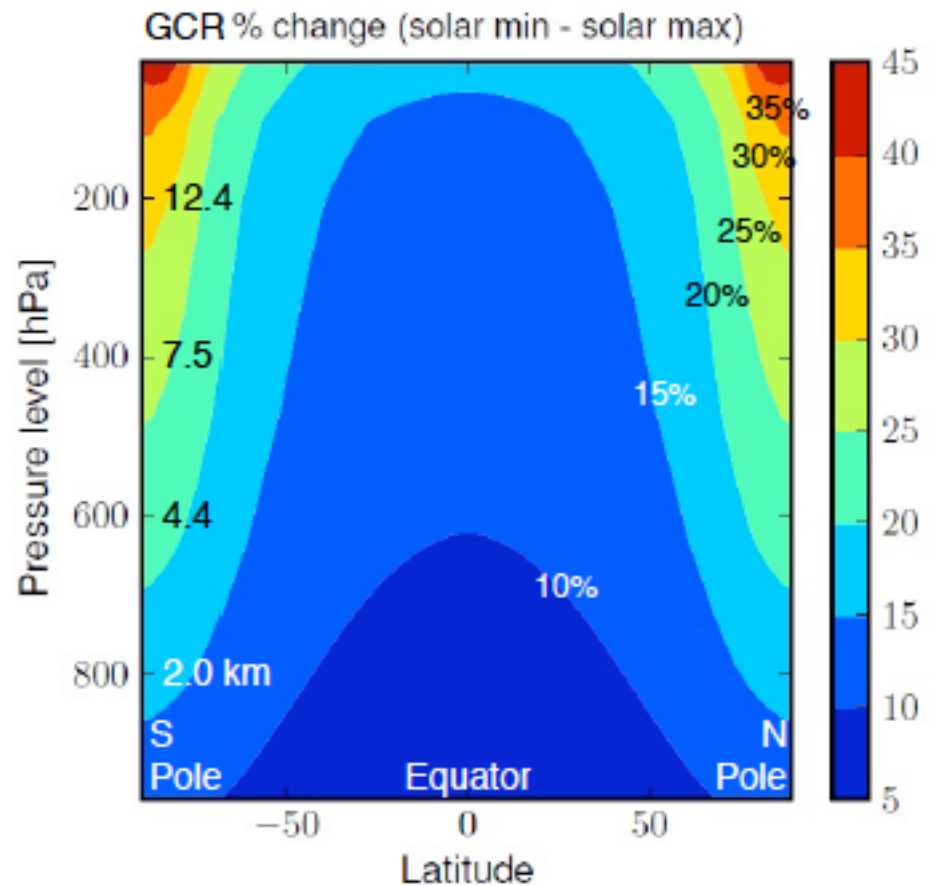
Saint Mary's University

- Cosmic rays produce ~ 2 ion pairs $\text{cm}^{-3}\text{s}^{-1}$ at the surface and ~ 40 at the top of the troposphere
- Ions from radon also important
- Charge-stabilised clusters could mean more CCN



GCR intensity variation

- Over the 11-year solar cycle, changes in solar wind cause on average a 15% change in GCR intensity (5% near the geomagnetic equator to 40% at the poles) (Carslaw et al, Science 2002)
- Over millennial time scales the GCR intensity can vary by a factor 3 or 4.

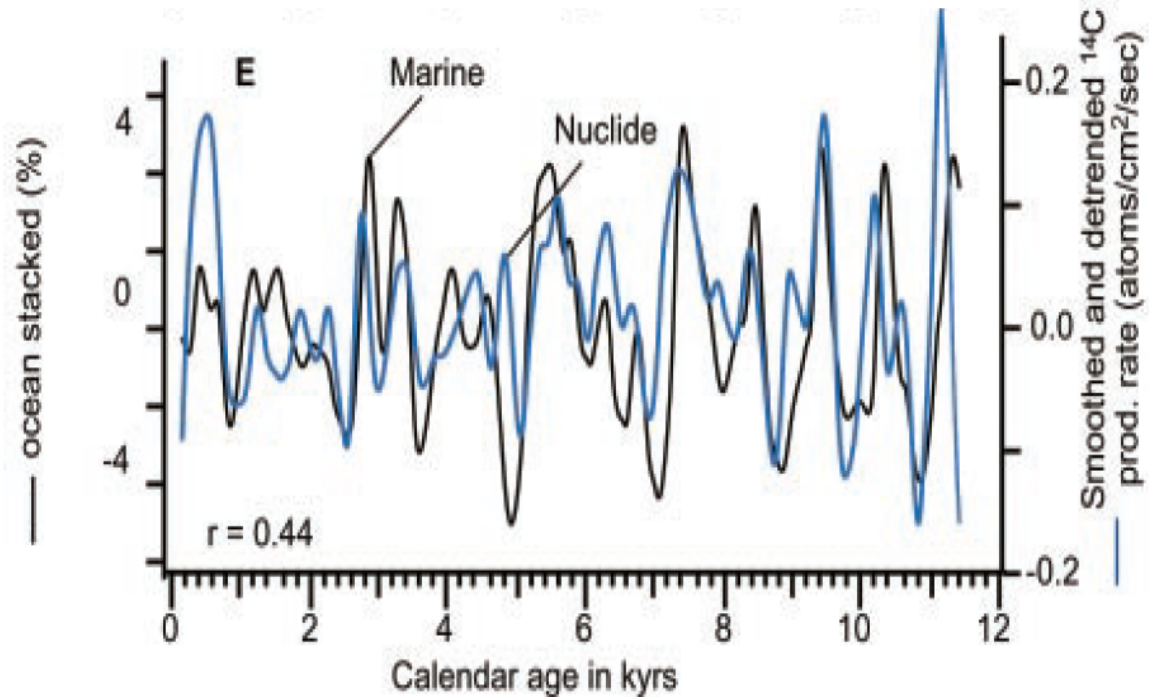


Could cosmic rays affect climate?

- Evidence for this in Bond et al, Science 2001:
e.g. correlation of North Atlantic ice mass with
C-14 (formed by GCRs)

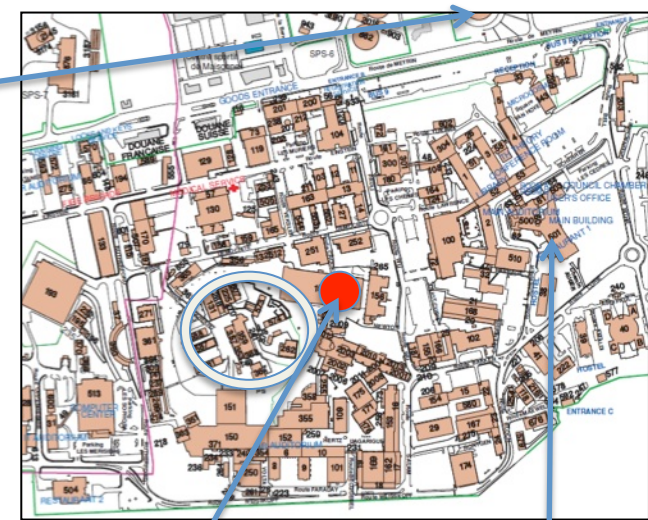
But cosmic ray flux also
correlated to total solar
irradiance, may be other
factors...

Correlation $\neq$ causation!



The CLOUD experiment

Globe



R1

- Chamber at CERN in the PS East Hall
- Study nucleation of aerosol from carefully controlled gas mixtures
- Investigate the effect of ions from cosmic rays or the PS beam on CCN formation
- Study the chemical processing of gases in clouds, focusing on possible effects of charges

Stainless steel chamber



Add an atmosphere and sunlight

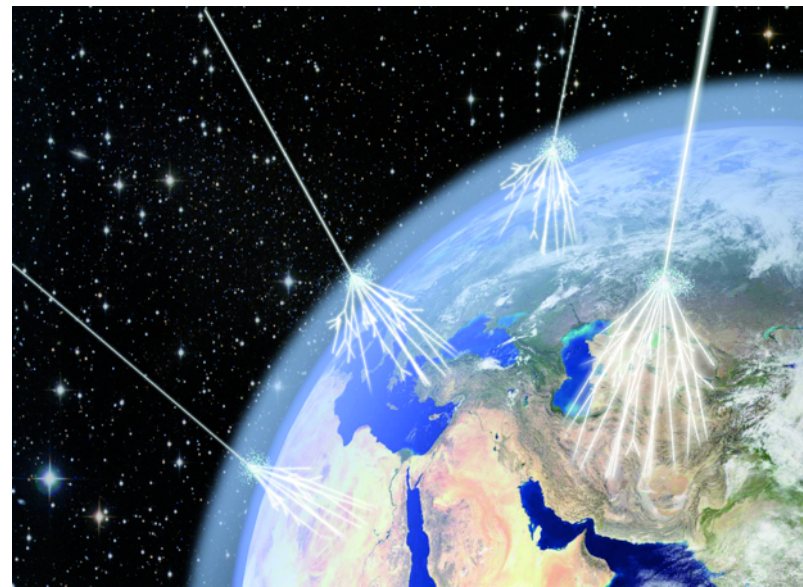
- Artificial air and trace gases from evaporators or bottles
- Ensure well mixed



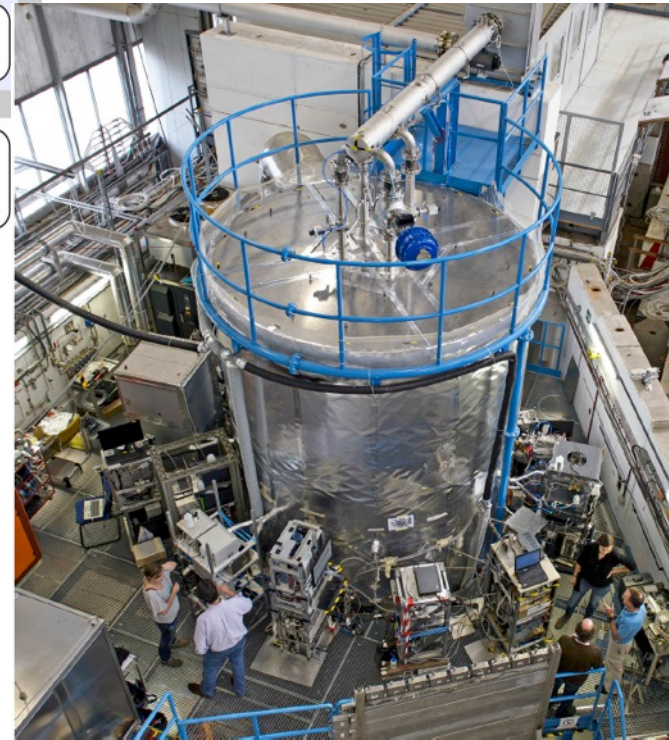
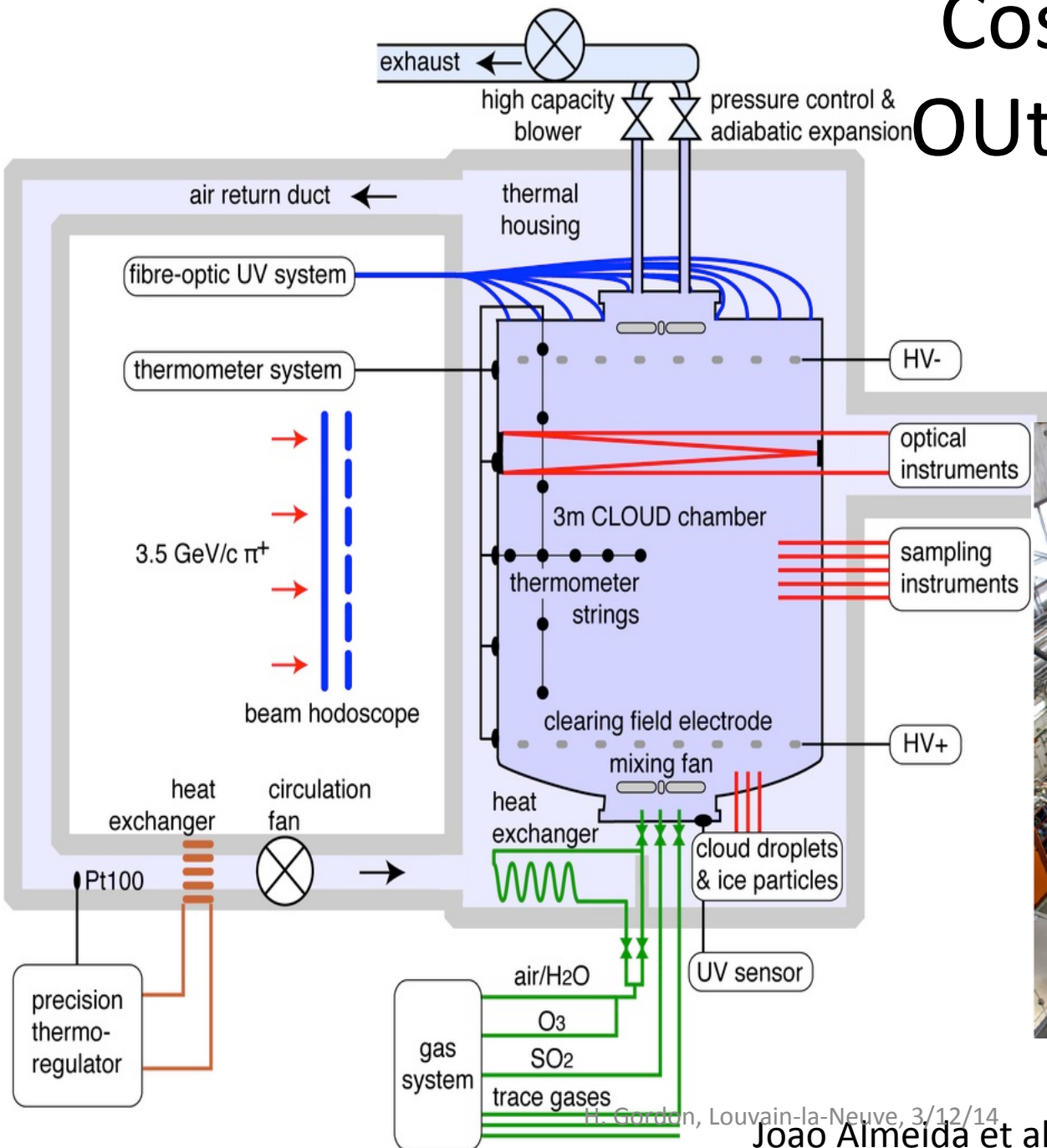
Add UV “sunlight” to
photolyse ozone



Add or remove ions



Cosmics Leaving Outdoor Droplets



H. Gordon, Louvain-la-Neuve, 3/12/14

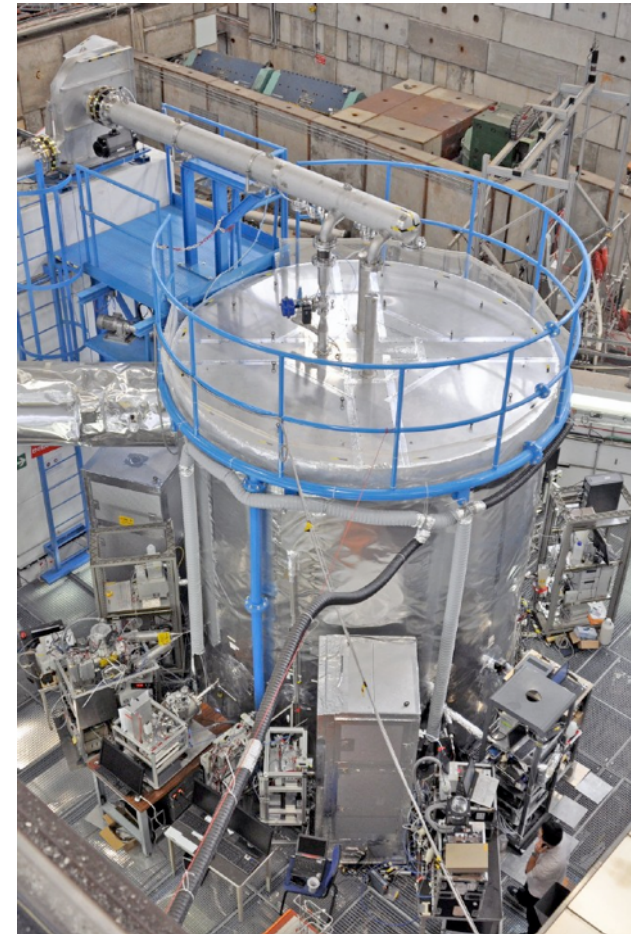
Joao Almeida et al, Nature **502** 359, (2013)

The CLOUD collaboration



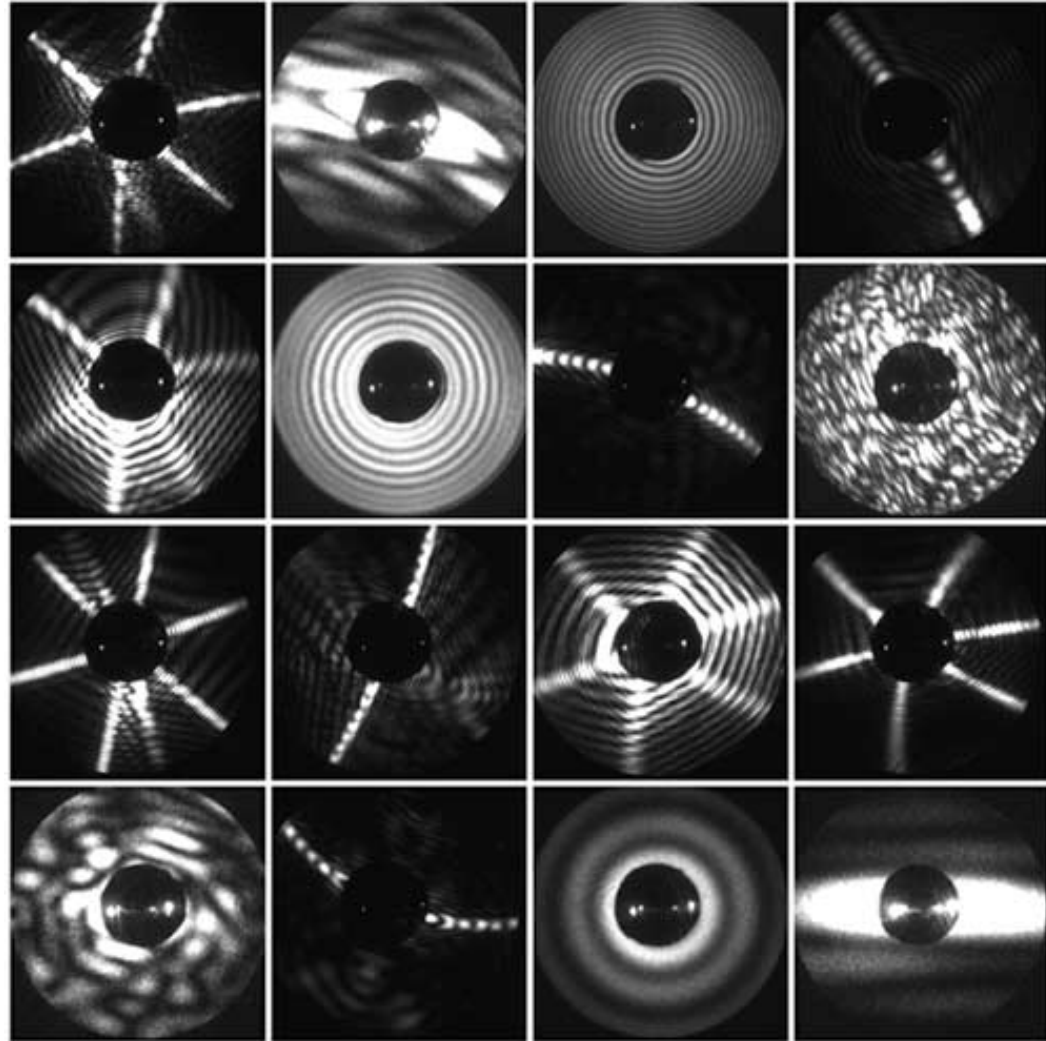
Cloudy experiments

- Inject water and cloud condensation nuclei into chamber, and pressurise to 200mbar over atmospheric pressure
- Then fast expansion, during which $P^{1-\gamma}T^{\gamma}=\text{constant}$, cools the chamber to make a cloud
- Cloud slowly evaporates
- Cloud droplets sampled by instruments connected to the chamber and optically monitored in situ



Cloud/ice experiments

- Investigate in-cloud processing of chemicals (reactions in clouds may not be the same as reactions in beakers)
- Investigate effectiveness of ice nuclei and ice crystal roughness, scattering properties
- Effect of charge on clouds (atmospheric E field $\sim 100 \text{ Vm}^{-1}$)



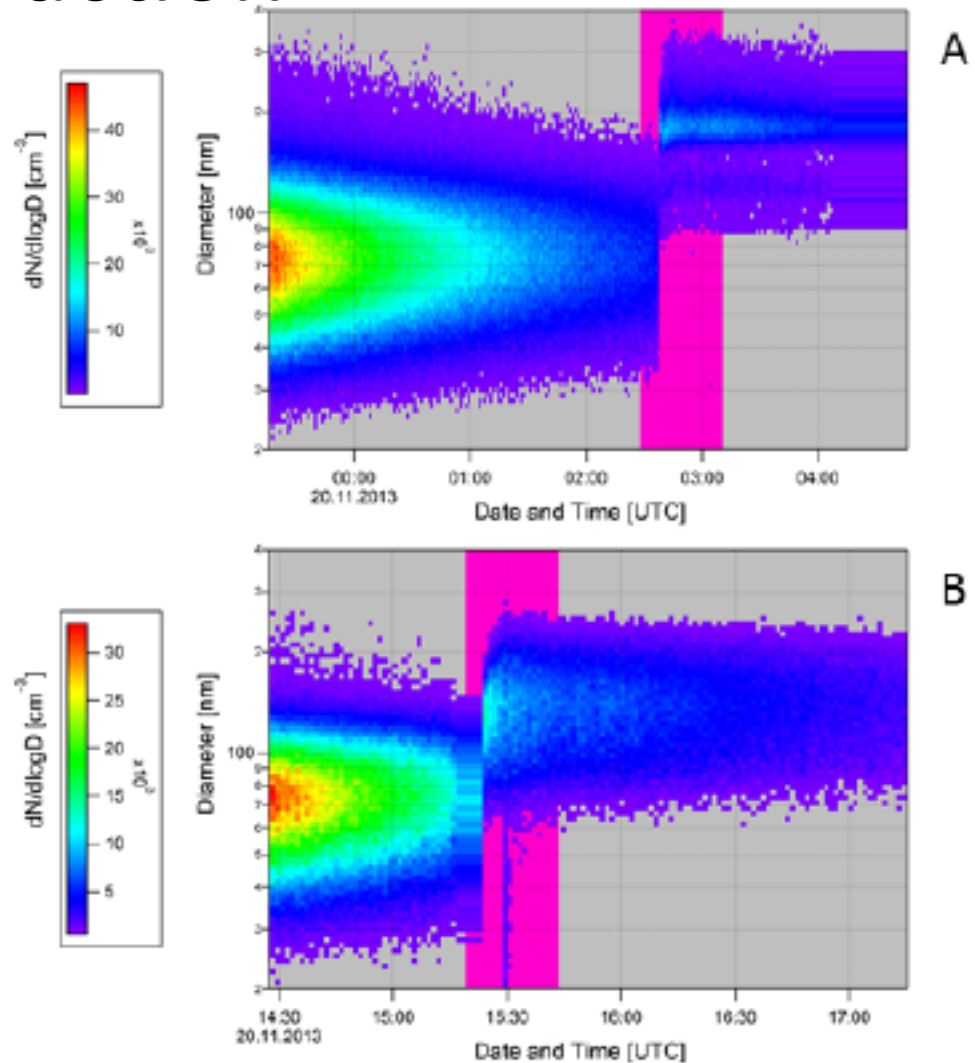
Cloud instrumentation



Mostly optical:
e.g. Cloud Aerosol and
Precipitation
Spectrometer (CAPS)
Measures droplet size
and depolarisation of
optically scattered light,
which helps discriminate
between water and ice
Study CCN as well as
cloud droplets
In-situ studies with
SIMONE/CCD

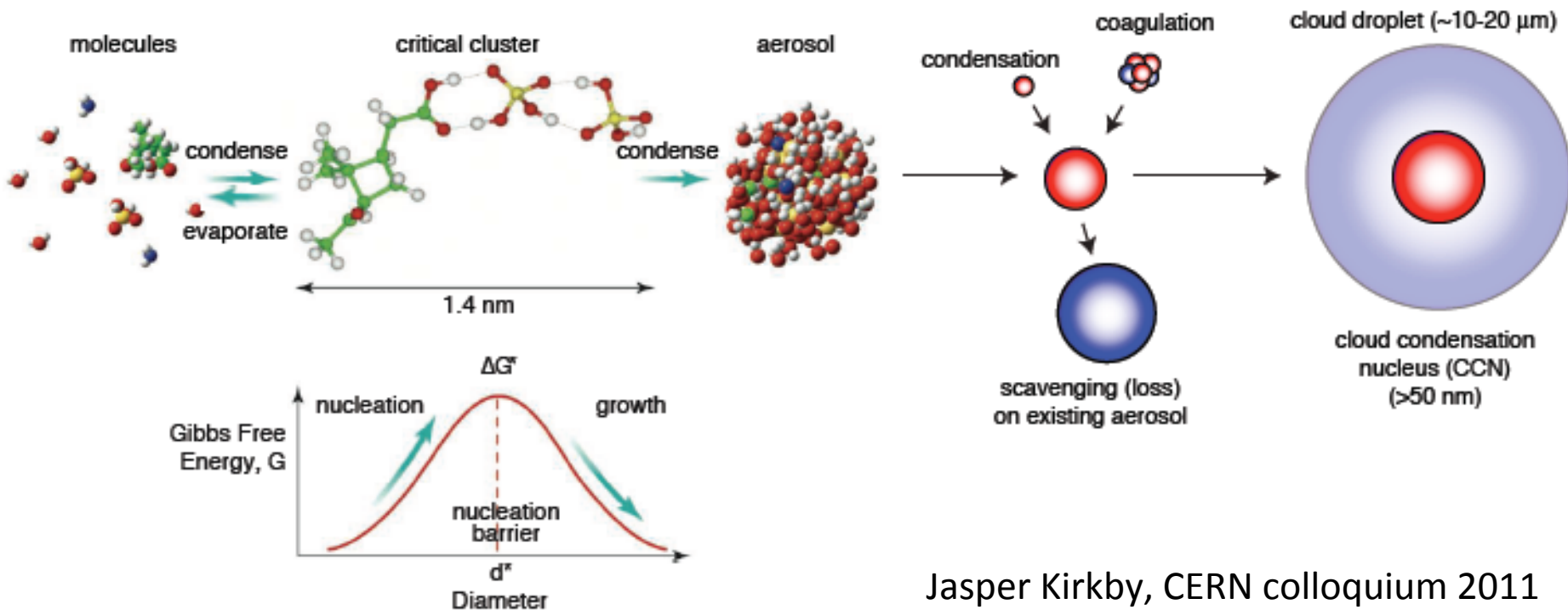
Aqueous chemistry and cloud-aerosol interaction

- Aerosol size distribution (right, at +10C and -10C) grows during a cloud (pink period) due to reactive uptake of SO₂ on ammonium sulphate seed aerosol



Nucleation experiments

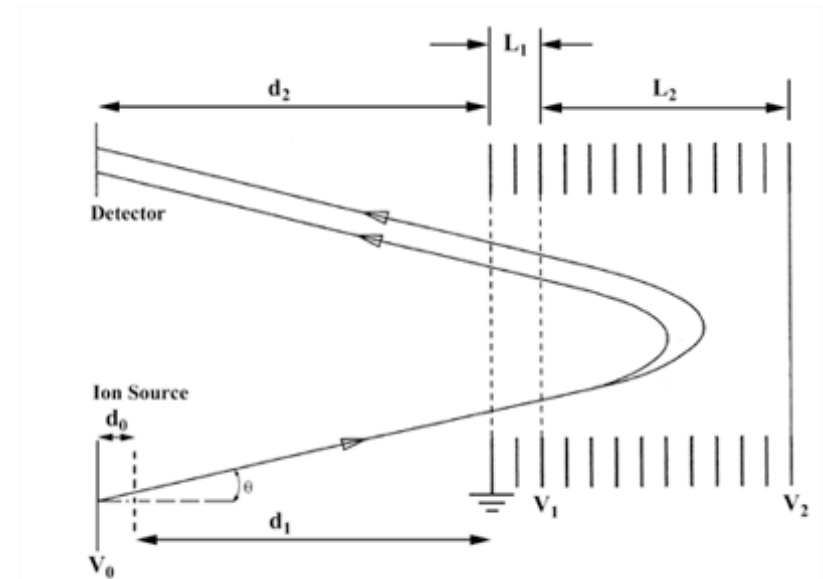
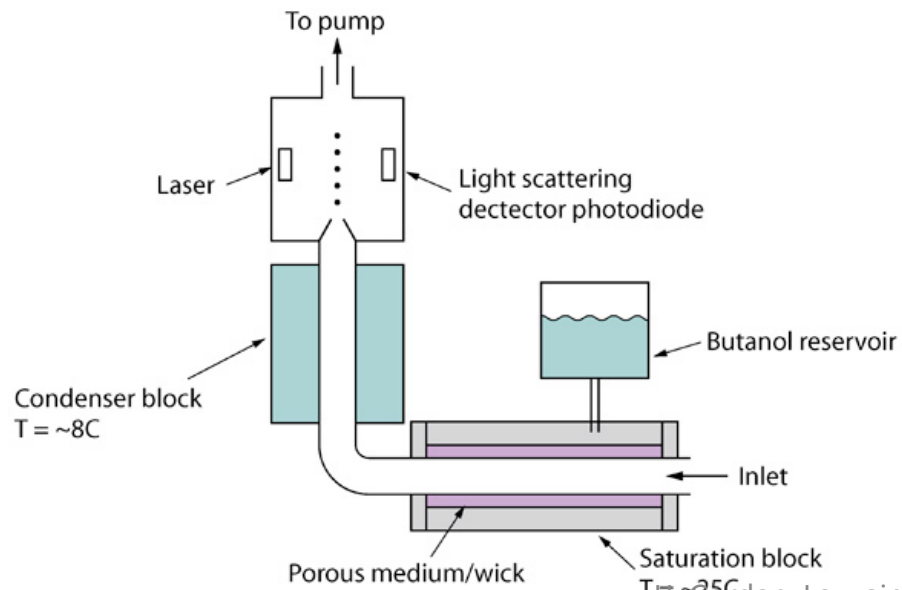
- Focus on formation of secondary atmospheric aerosol particles and their growth to CCN size



Jasper Kirkby, CERN colloquium 2011

Nucleation instrumentation

- Particle counters
- How many particles of a given size?
- CPC, SMPS, CCNC, nRDMA
- Mass spectrometers (mostly TOF)
- What do the particles contain?

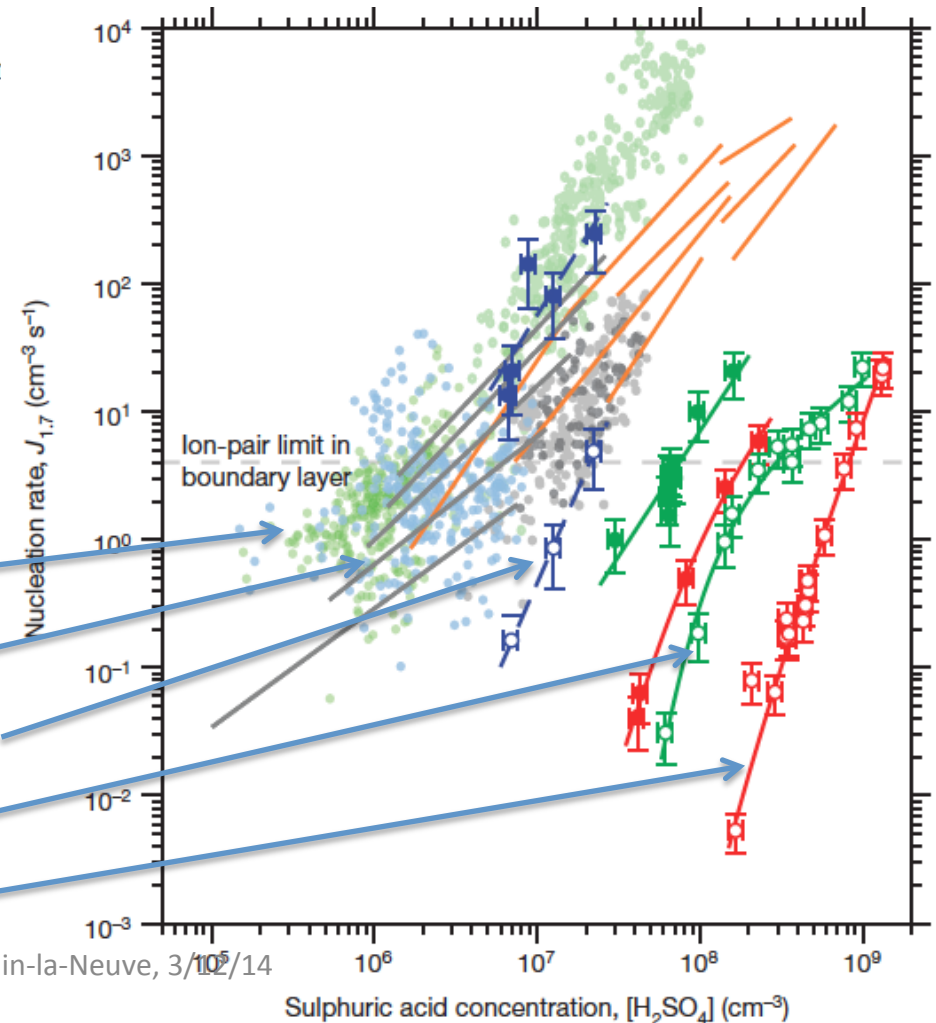


Role of sulphuric acid, ammonia and galactic cosmic rays in atmospheric aerosol nucleation

Jasper Kirkby¹, Joachim Curtius², João Almeida^{2,3}, Eimear Dunne¹

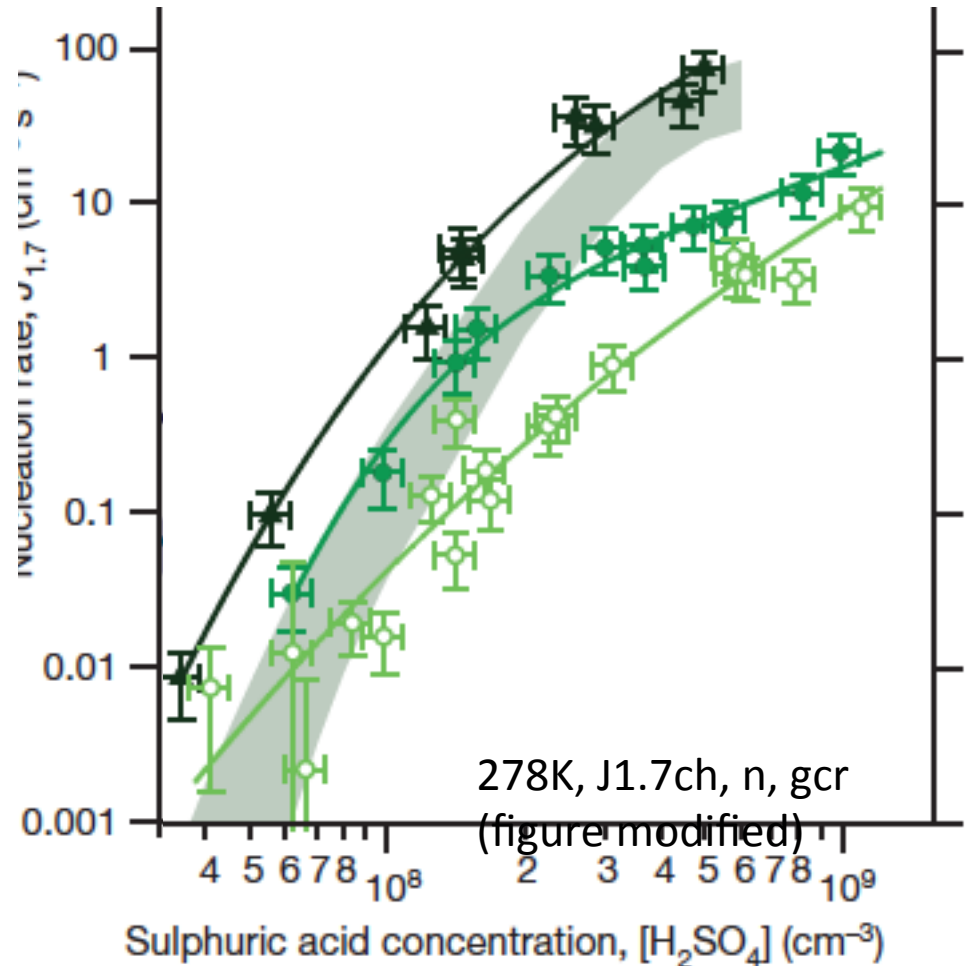
Sulphuric acid and ammonia alone cannot explain observed BL new particle formation rates

- Atmospheric data (dots)
- Other lab measurements (lines)
- CLOUD data at 248K (blue, filled=higher NH₃)
- CLOUD data at 278K (green)
- CLOUD data at 292K (red)



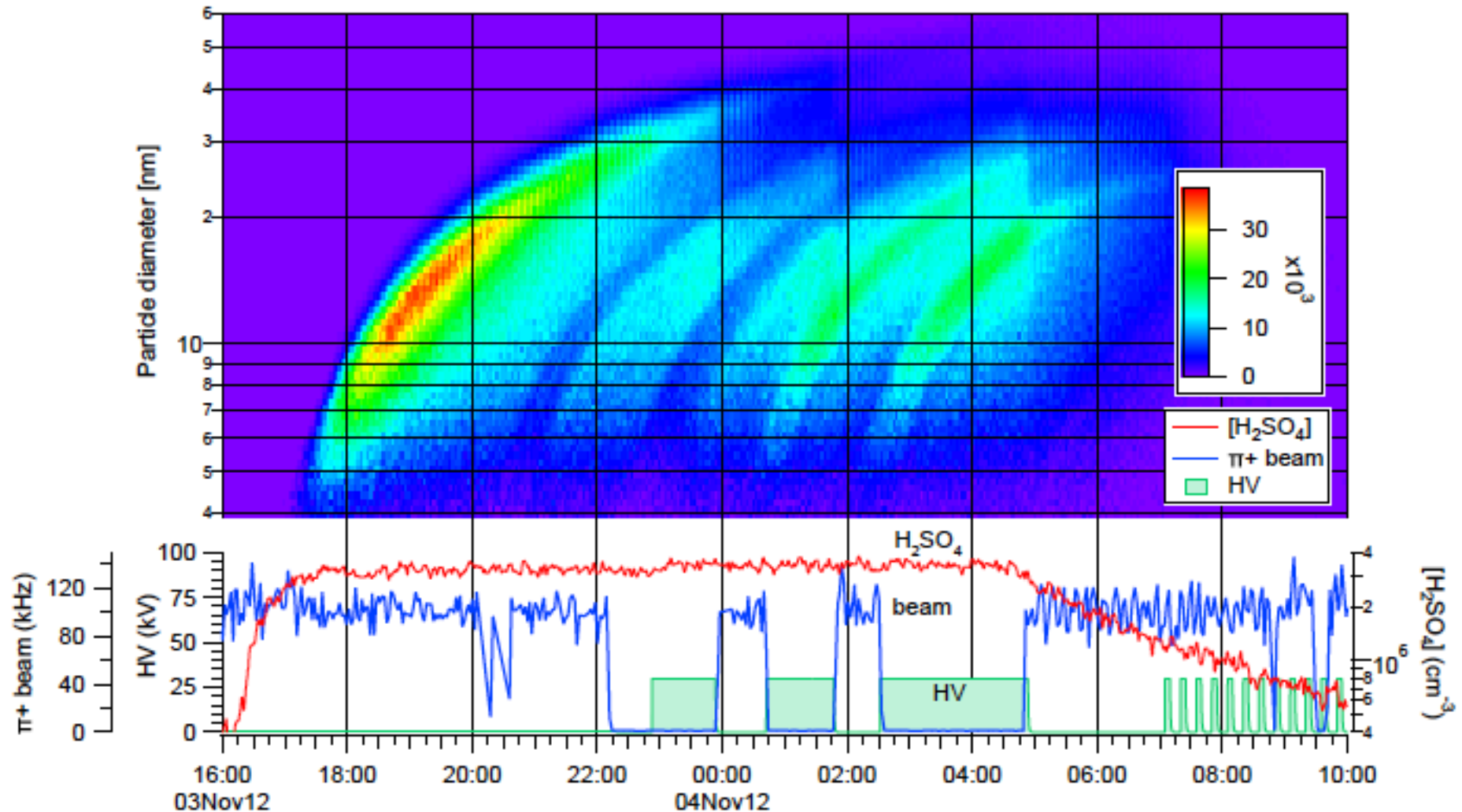
Ion-induced nucleation

Cosmic rays enhance rates of new particle formation by more than a factor of 10 in specific conditions (if the formation rate is very low to begin with)
May be relevant in FT!



Example ion-induced nucleation experiment

UV switched on at start, then only ion concentration varied



CLOUD report to SPSC, 2014

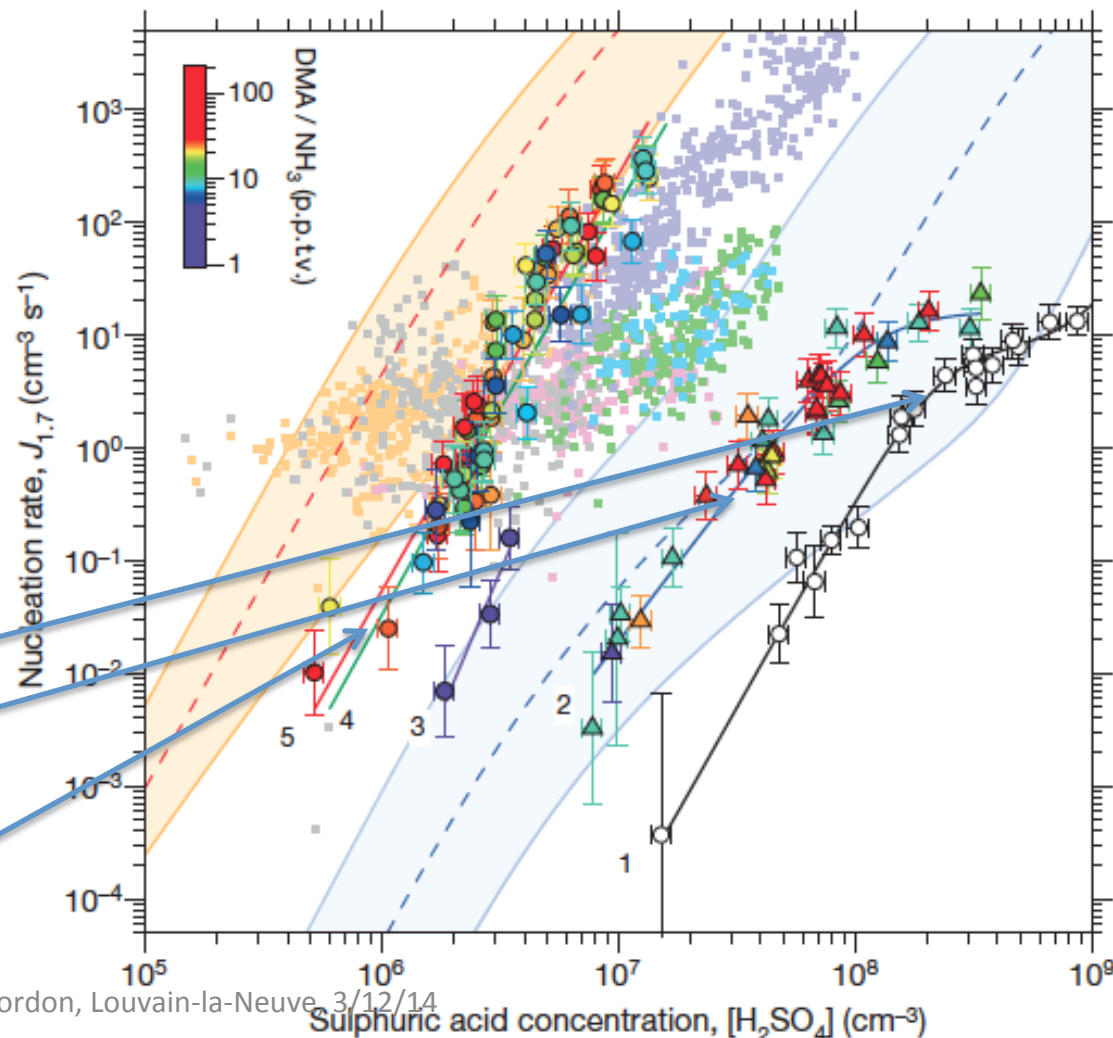
Molecular understanding of sulphuric acid–amine particle nucleation in the atmosphere

João Almeida^{1,2}, Siegfried Schobesberger³, An

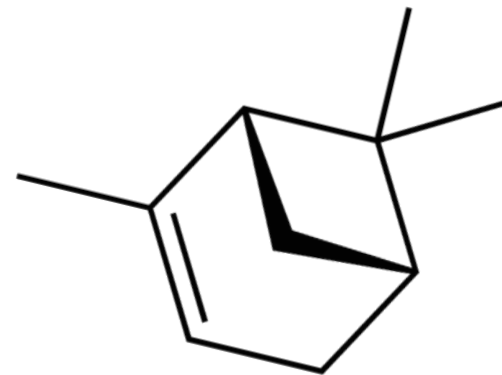
Sulphuric acid and amines can explain observed new particle formation rates if the concentrations of amines are high enough (only true some of the time)

Sulphuric acid (+water)
H₂SO₄ and ammonia (colour scale for NH₃), + theory blue band

H₂SO₄, a little ammonia (10pptv), and DMA (colour scale for DMA)
+theory orange band

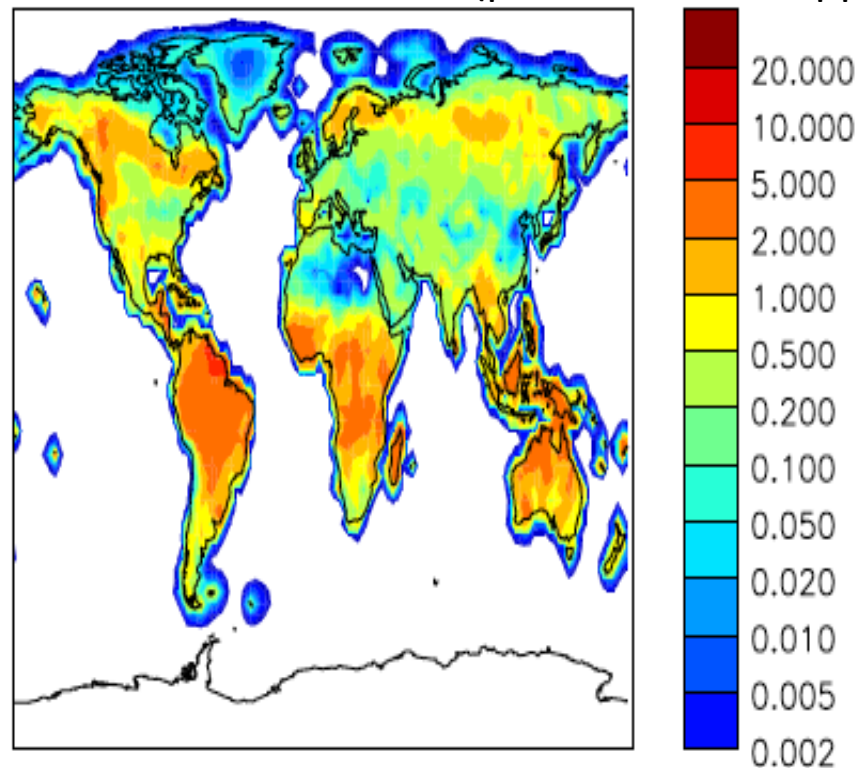


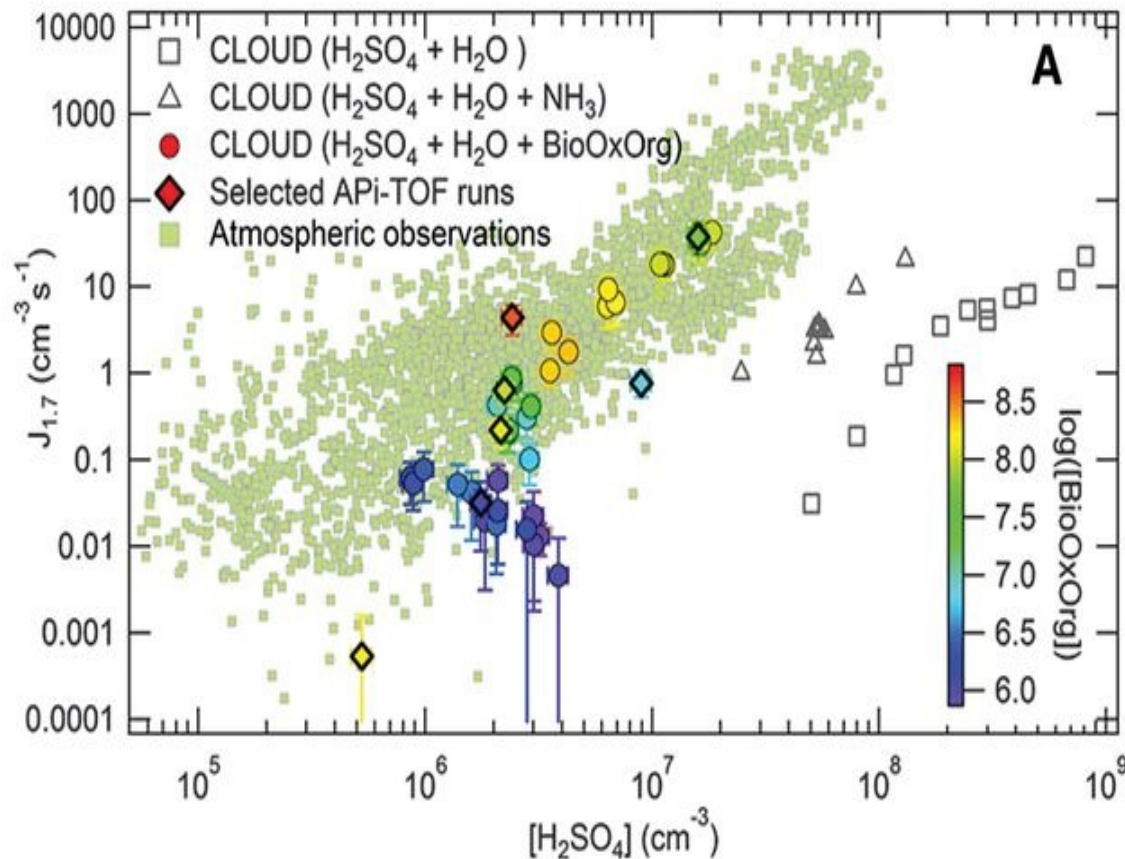
Pinenes and ELVOCs



- Cyclic carbon compounds with 10 carbon atoms (pinenes) are emitted by trees in large quantities (of order 0.1% of the annual CO₂ mass flux, mostly in spring/summer)
- Pinenes are oxidised by ozone, OH or NO_x to form **extremely low volatility organic compounds** (ELVOCs) which readily condense onto atmospheric aerosols and participate in particle formation
- State-of-the-art summarised in Ehn et al, A large source of low-volatility secondary organic aerosol, Nature **506** 476 (2014)

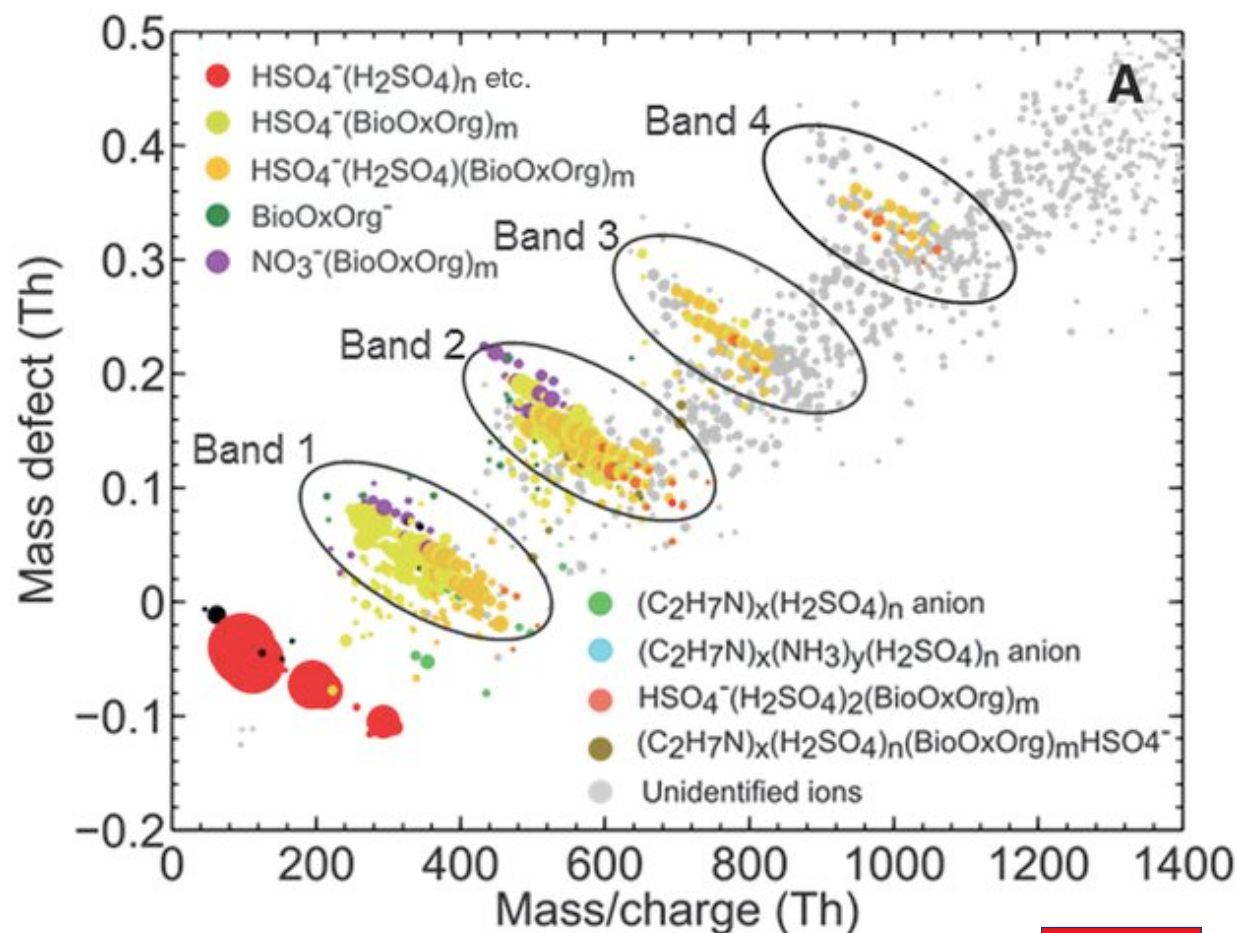
Concentration (ppt) of oxidised terpenes in GLOMAP aerosol model (peak SA conc 0.4ppt)





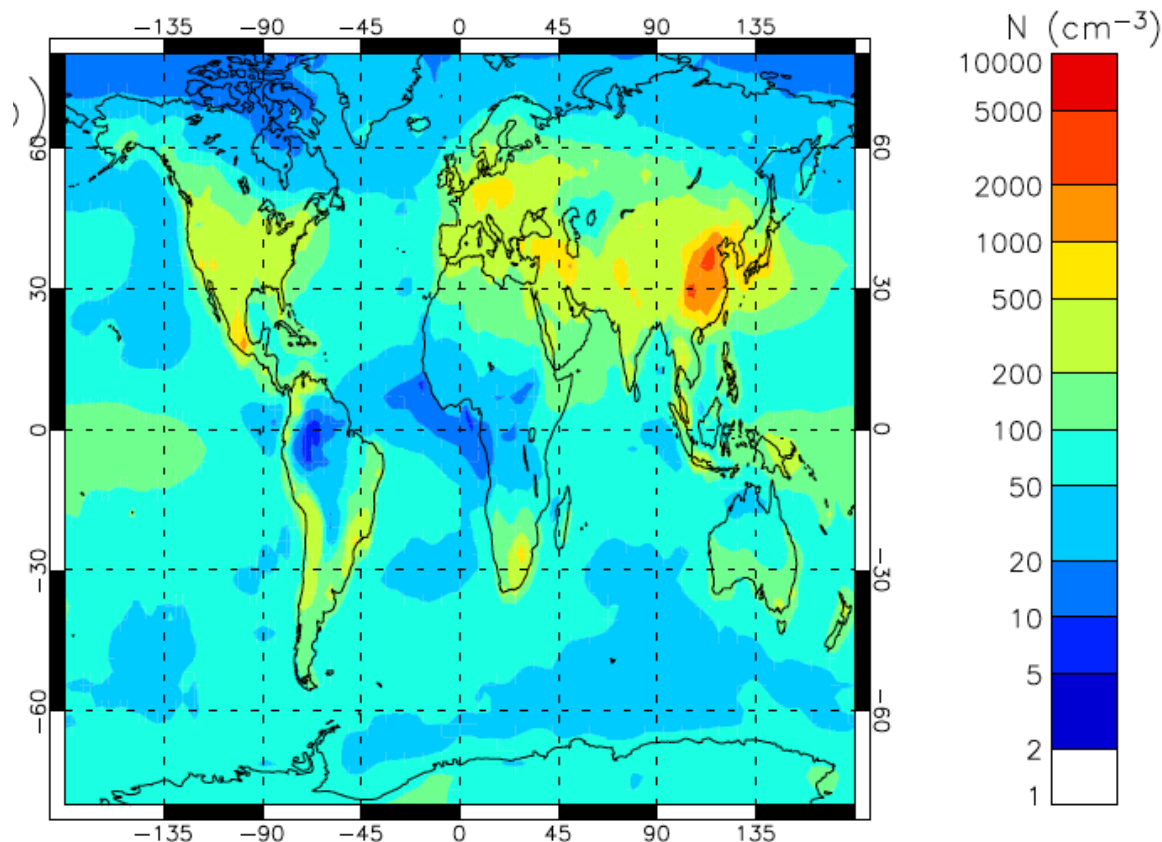
These organic compounds have a much stronger effect than ammonia and are much more abundant than DMA

Nucleation as seen by TOF-MS



Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles
 Francesco Riccobono *et al.*
Science **344**, 717 (2014);
 DOI: 10.1126/science.1243527

Global model of aerosol processes



Form, grow and transport nanoparticles and CCN around the atmosphere
Simulate cloud-aerosol interactions and particle loss rates

Can try to reproduce measured particle number concentrations

Reproduce gas phase chemistry

Aitken soluble mode number concentration, Mann et al, GMD 3 512 (2010)

Host Chemical Transport Model

- Forced by ECMWF Winds
- Convective transport
- Convective and resolved rain

GLOMAP

- Aerosol size spectrum ($\sim 3 \text{ nm} - 24 \mu\text{m}$)
- 2-moment (m and N) sectional or modal scheme

Sources

Emissions

- Anthrop + volcanic SO_2 emissions
- DMS emissions from wind stress and DMS sea surface concentration
- Sea salt aerosol generation function

Sulfur Chemistry

- 8 sulfur species, 8 sulfur reactions
- Aqueous phase chemistry
- Oxidants from full chemistry run

Microphysics

Nucleation and Condensation

- Binary $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ nucleation
- Condensational growth

Coagulation

- Semi implicit fast numerical solution

Hygroscopic Growth

- Equilibrium size given by Kohler equation

Removal

Dry Deposition

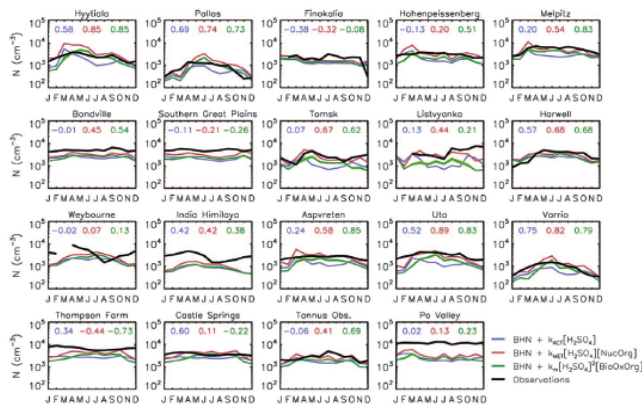
- Dry deposition of aerosol

Clouds

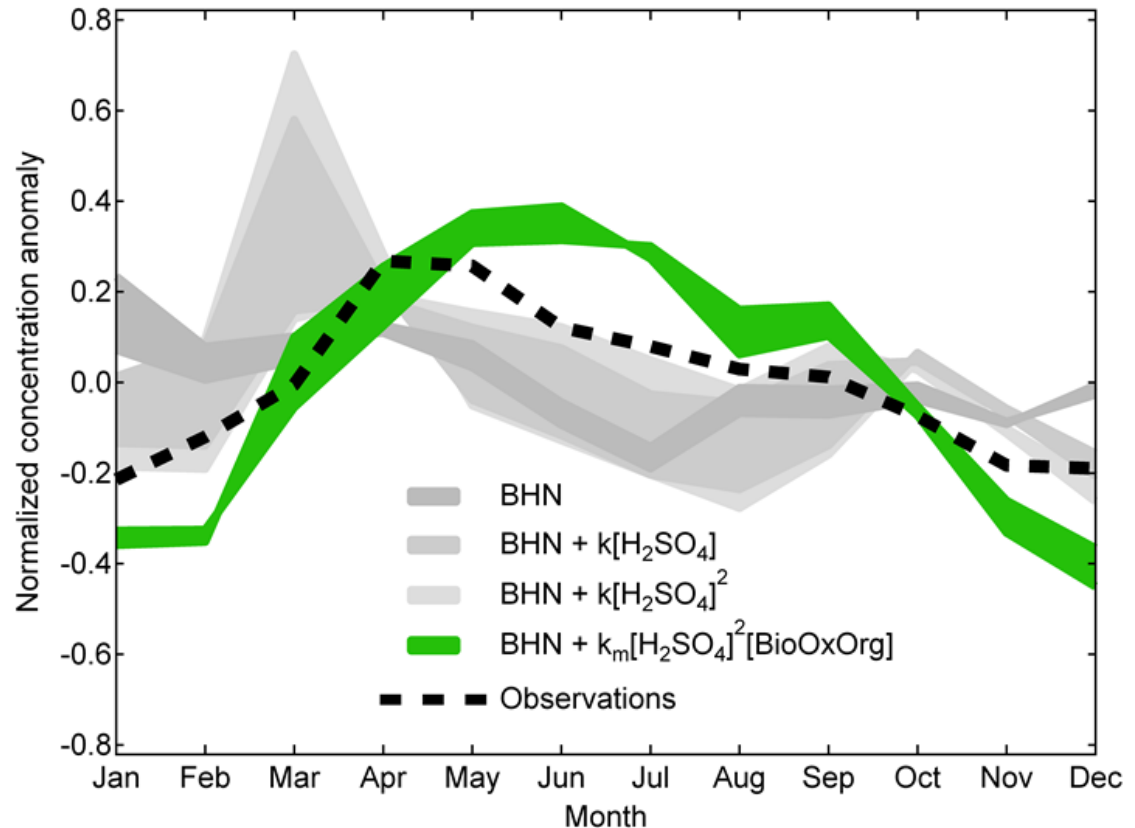
- Prognostic convective and frontal rain
- In-cloud and below-cloud aerosol/gas scavenging
- Cloud-processing in diagnosed low-level cloud

Dominick Spracklen

Normalised anomaly in particle concentrations (3-14 nm) at 19 northern hemisphere continental sites

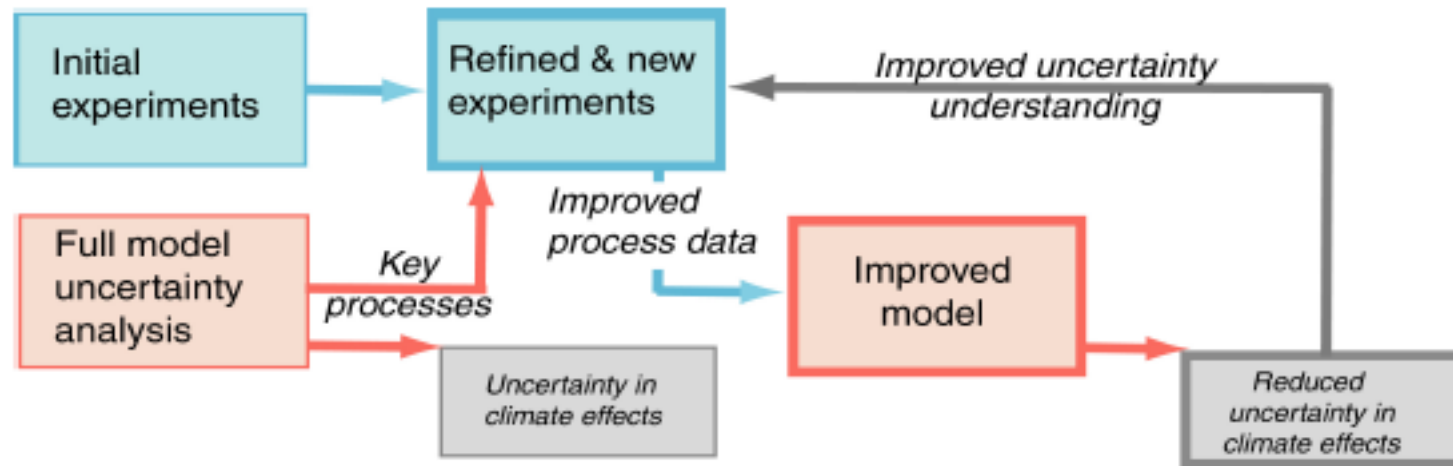


Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles
Francesco Riccobono *et al.*
Science **344**, 717 (2014);
DOI: 10.1126/science.1243527



Improved explanation
of particle formation
in the atmosphere in
the summer months

Next steps for CLOUD



- Study nucleation of biogenic organic compounds more comprehensively with upgraded UV light source
- Key question: can we get significant new particle formation without sulphuric acid, ozone, NO_x, etc? (i.e. in the **unpolluted pre-industrial atmosphere** over land)
- Feedback to GLOMAP model– first papers on atmospheric relevance of CLOUD findings to be submitted for publication soon

Summary

- CLOUD is an expansion chamber at CERN studying **new particle formation** and **clouds** in the atmosphere
- A key aim is to investigate effect of **cosmic rays** on cloud formation
- Key result so far is that **ammonia and sulphuric acid cannot explain particle formation** in the lower atmosphere, as previously assumed
- The **organic vapours** that (with sulphuric acid) do explain the nucleation are under investigation
- Possibility to use CLOUD to study **pre-industrial atmosphere**
- Good prospects for **cloud chemistry and ice microphysics** studies in parallel

<http://cloud.web.cern.ch>

Long-term natural climate change

- Milankovitch cycles: millennial time scale variations in Earth's orbit.
- TSI: 0.1% irradiance variation over one solar cycle translates to up to 0.1K temperature change
- Similar variation between Maunder Minimum (1645-1715, reign of the Sun King!) and present day
- TSI increased in the first half of the 20th century a little, and decreased a little since 1970