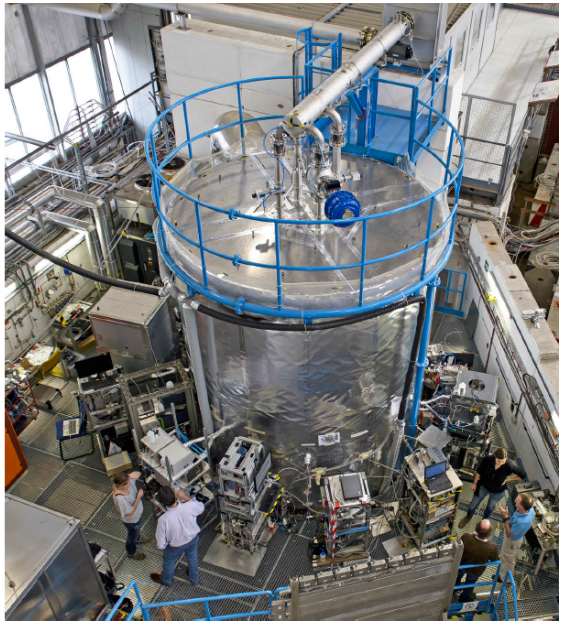


T. Vatanen

Results from the CLOUD experiment

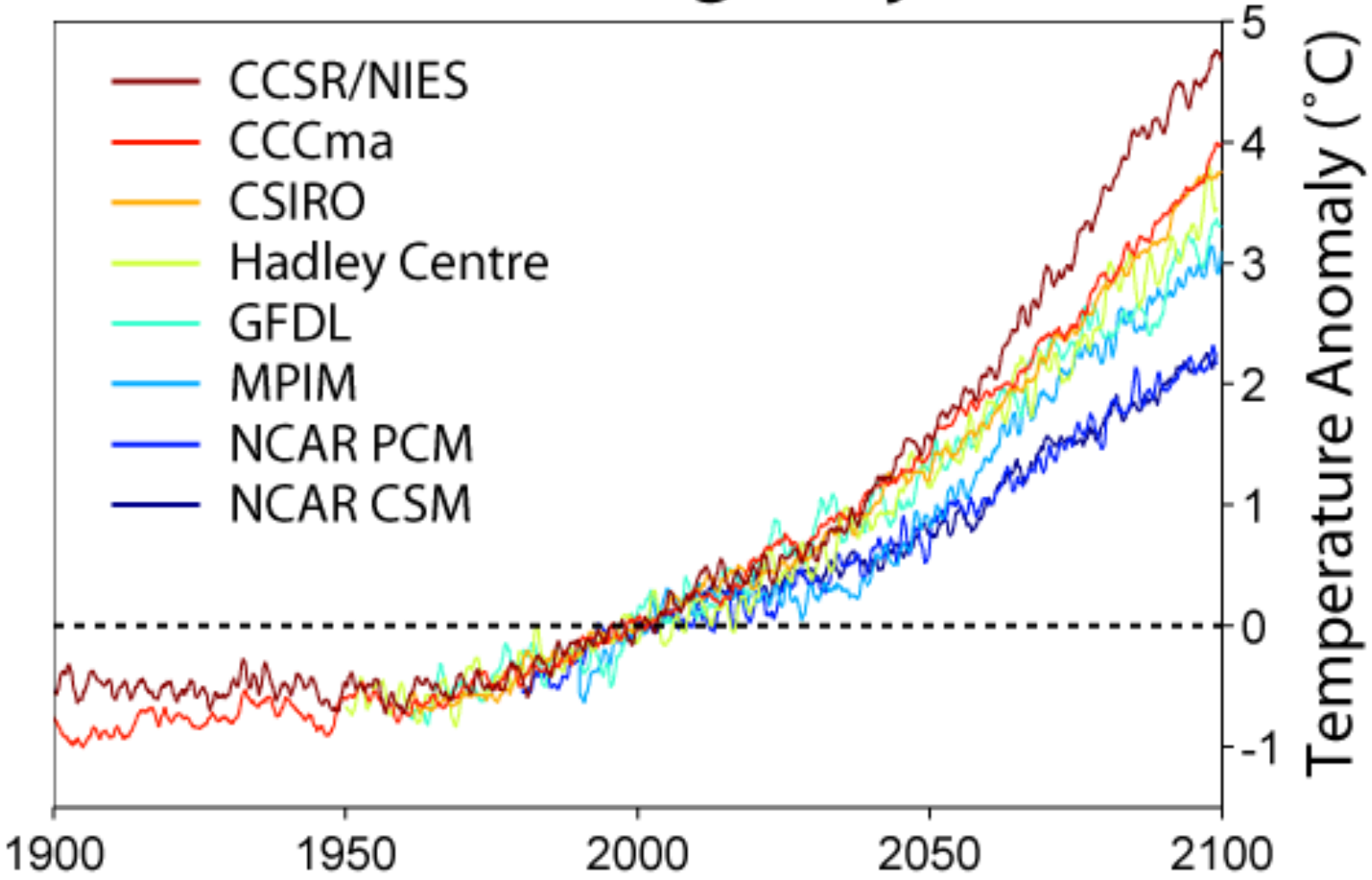
Hamish Gordon (CERN)

LPNHE HEP seminar 2/7/15

CERN

IPCC 2013

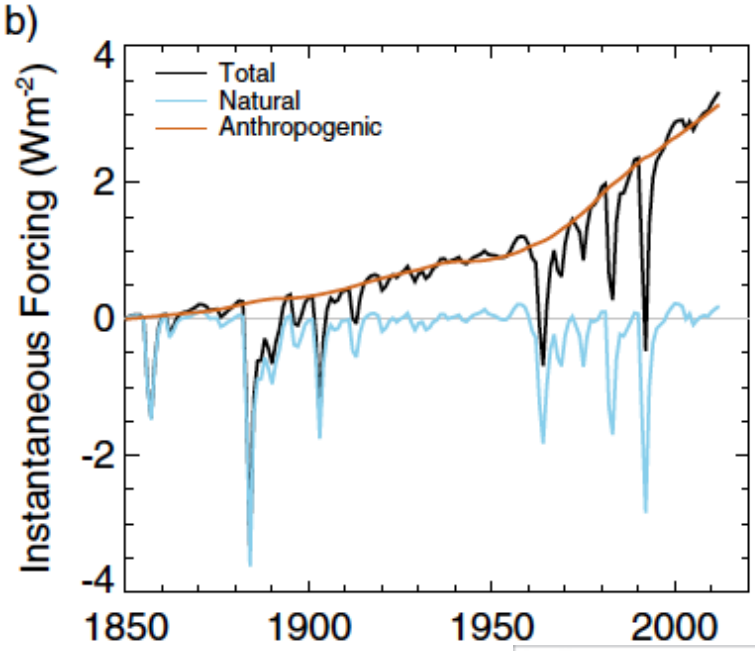
Global Warming Projections



- 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."

Agenda

- Effect of clouds and aerosols on climate
- CLOUD chamber experiment
- Highlights of CLOUD results 2011-2015
- Implications for understanding the atmosphere: interpretation via global aerosol modelling
- Future prospects



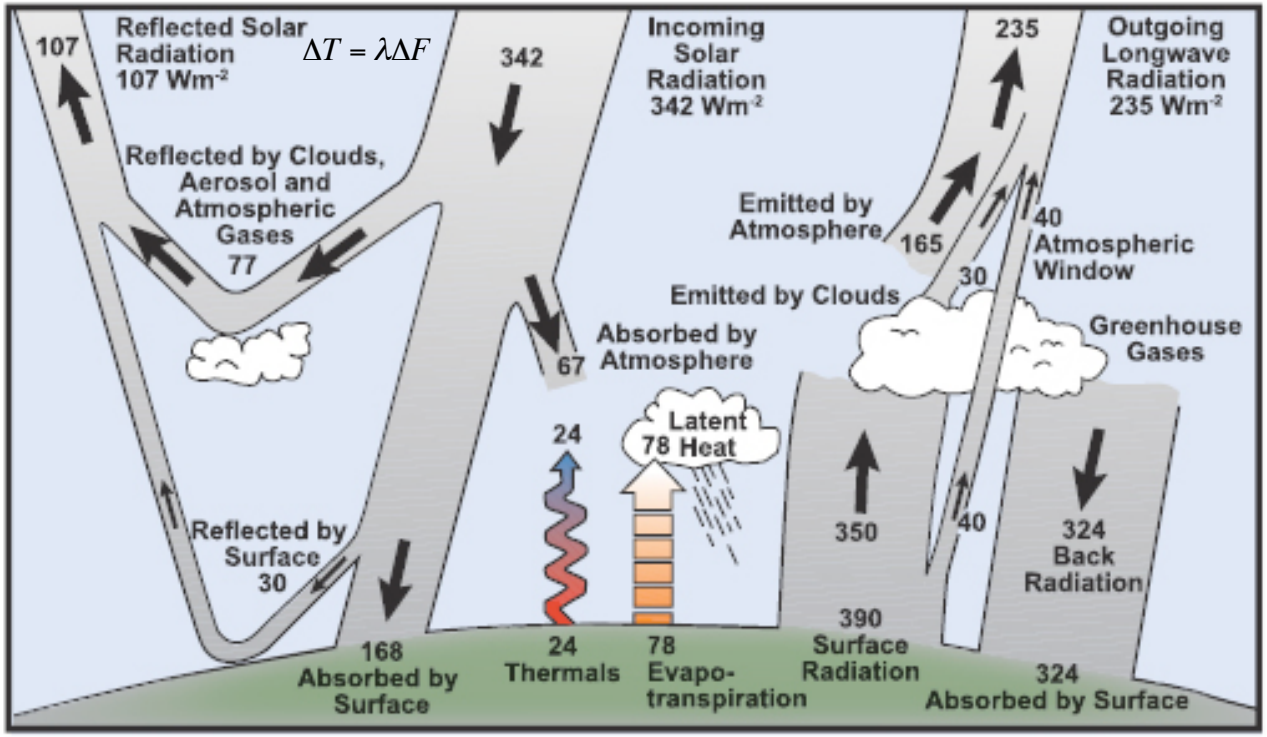
Radiative forcing

Instantaneous global radiative forcing at the tropopause in the E2-R NINT ensemble. (Miller et al, 2014)

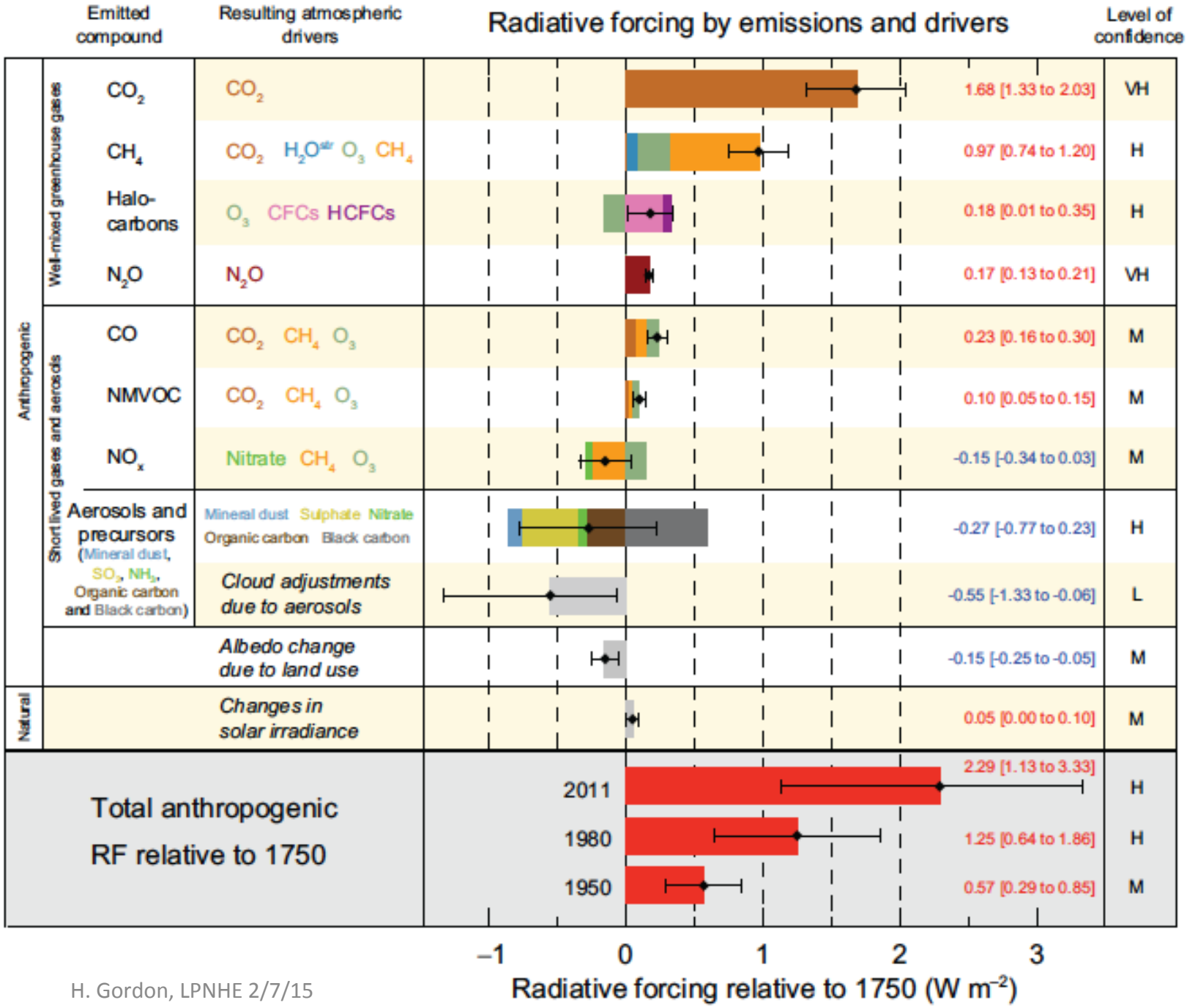
IPCC: "effective radiative forcing": radiative forcing after rapid adjustments.

Climate sensitivity:

$$\Delta T = \lambda \Delta F$$



IPCC 2007

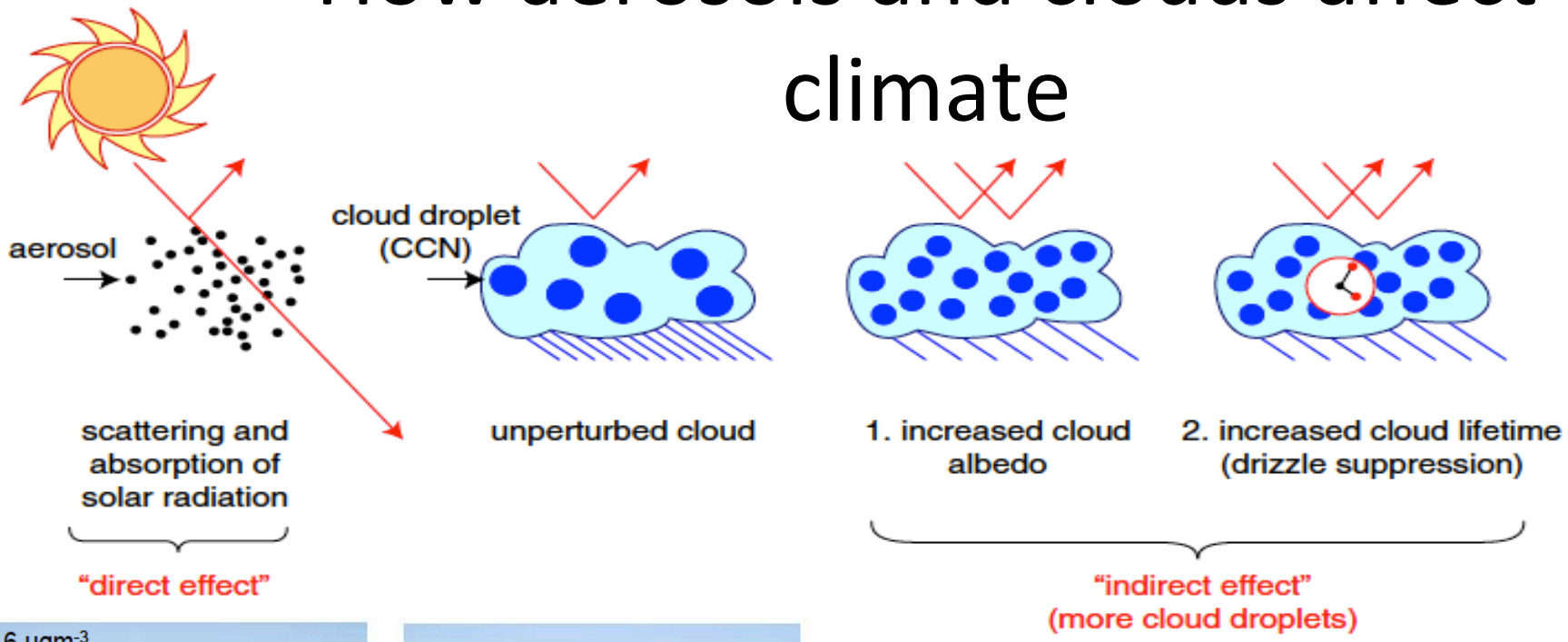

Emitted compound
Resulting atmospheric drivers
Radiative forcing by emissions and drivers
Level of confidence
Anthropogenic
Natural
Well-mixed greenhouse gases
Short lived gases and aerosols
CO2
CH4
Halo-carbons
N2O
CO
NMVOC
NOx
Aerosols and precursors (Mineral dust, SO2, NH3, Organic carbon and Black carbon)
CO2
CO2 H2Osstr O3 CH4
O3 CFCs HCFCs
N2O
CO2 CH4 O3
CO2 CH4 O3
Nitrate CH4 O3
Mineral dust Sulphate Nitrate Organic carbon Black carbon
Cloud adjustments due to aerosols
Albedo change due to land use
Changes in solar irradiance
1.68 [1.33 to 2.03]
0.97 [0.74 to 1.20]
0.18 [0.01 to 0.35]
0.17 [0.13 to 0.21]
0.23 [0.16 to 0.30]
0.10 [0.05 to 0.15]
-0.15 [-0.34 to 0.03]
-0.27 [-0.77 to 0.23]
-0.55 [-1.33 to -0.06]
-0.15 [-0.25 to -0.05]
0.05 [0.00 to 0.10]
VH
H
H
VH
M
M
M
H
L
M
M
Total anthropogenic RF relative to 1750
2011
1980
1950
2.29 [1.13 to 3.33]
1.25 [0.64 to 1.86]
0.57 [0.29 to 0.85]
H
H
M
–1
0
1
2
3
Radiative forcing relative to 1750 (W m–2)

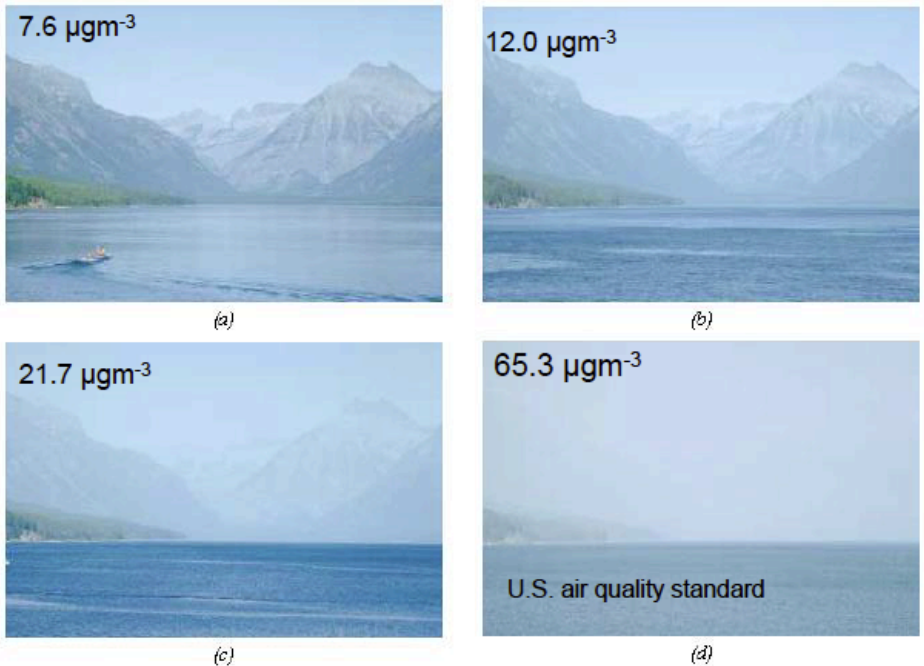
Cloud formation

- 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

Cloud Appreciation Society

How aerosols and clouds affect climate



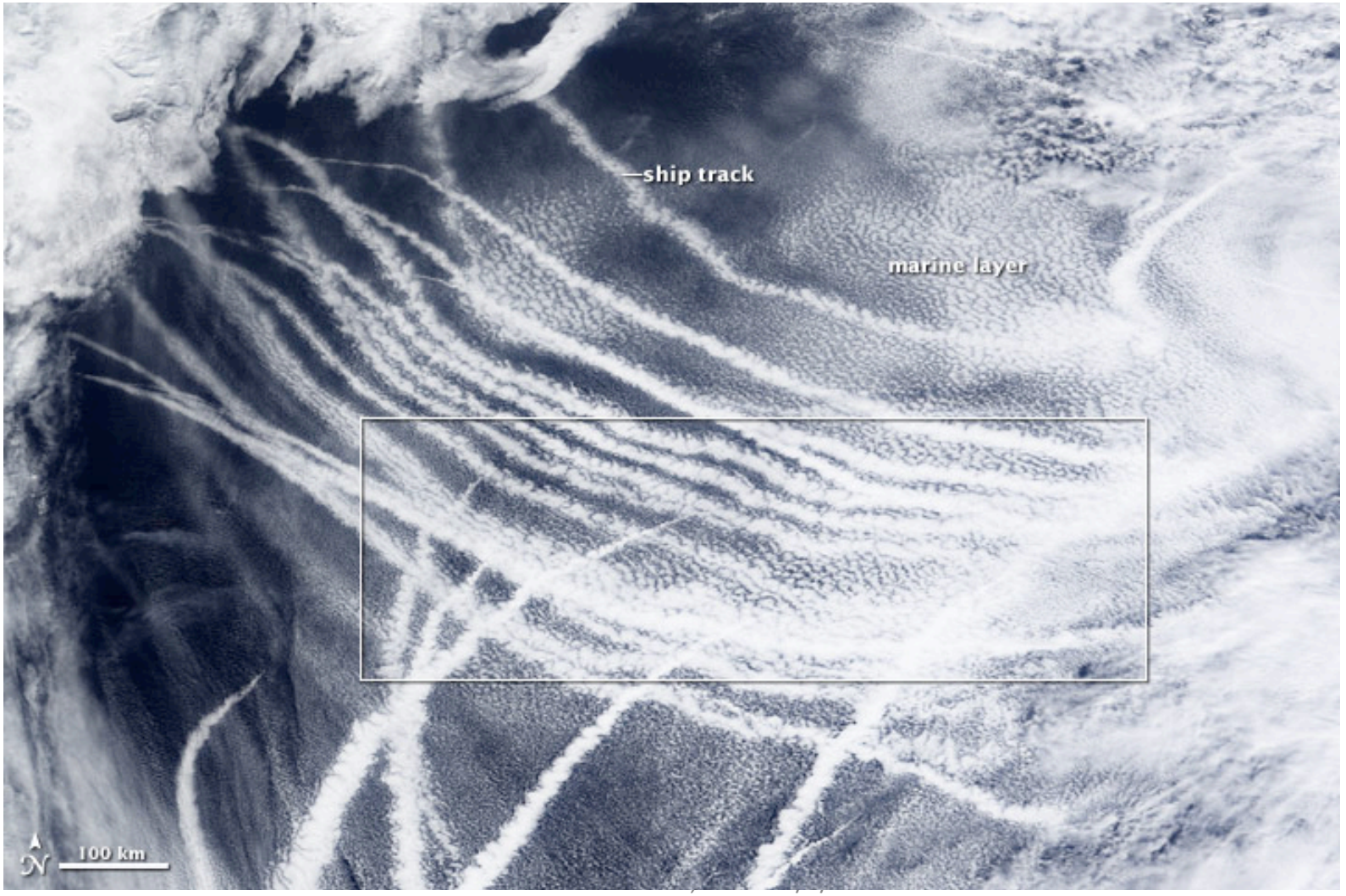


- Clouds are (more-or-less) white, the Earth is black, and ~60% of Earth's surface is covered by clouds

Picture credit: Jasper Kirkby, Surv Geophys **28** 333 (2007)

J.M.W. Turner, Calais Pier, 1803

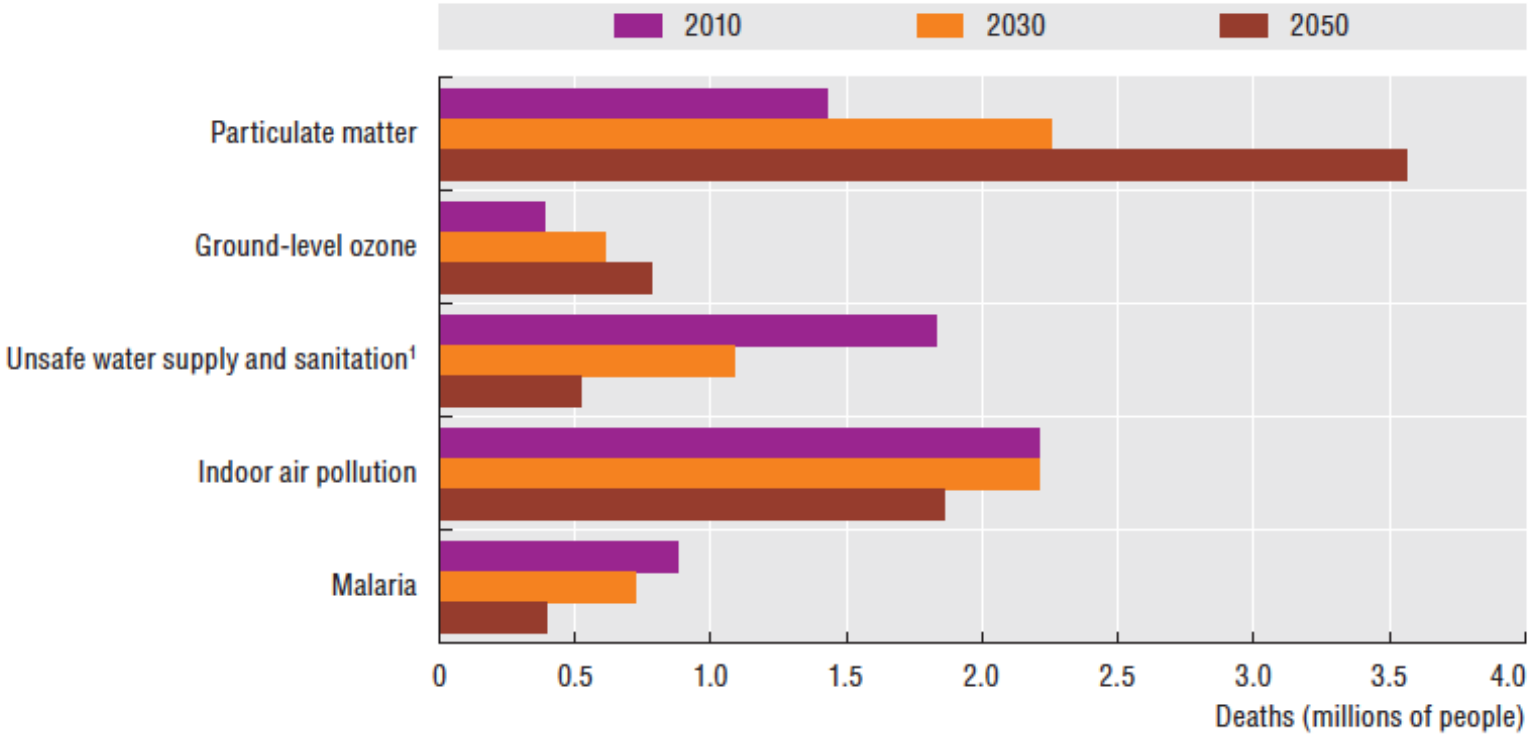
MODIS satellite, Pacific Ocean, March 4 2009



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**

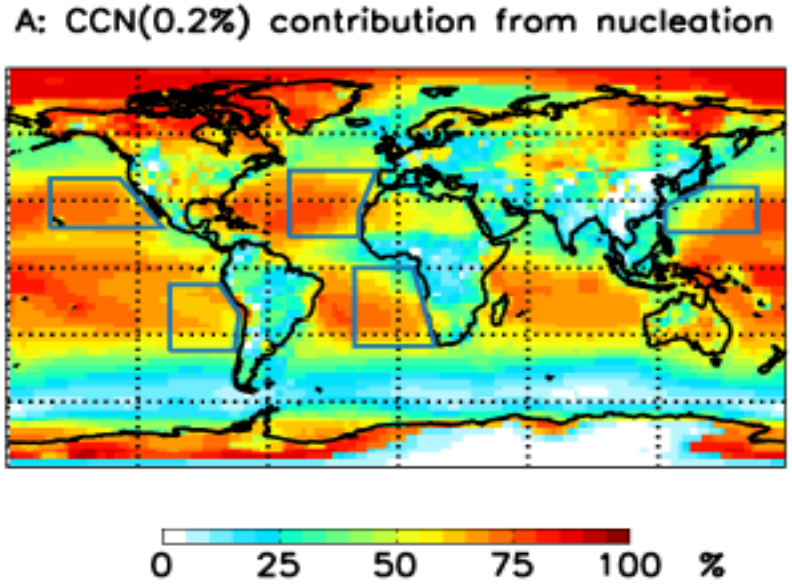


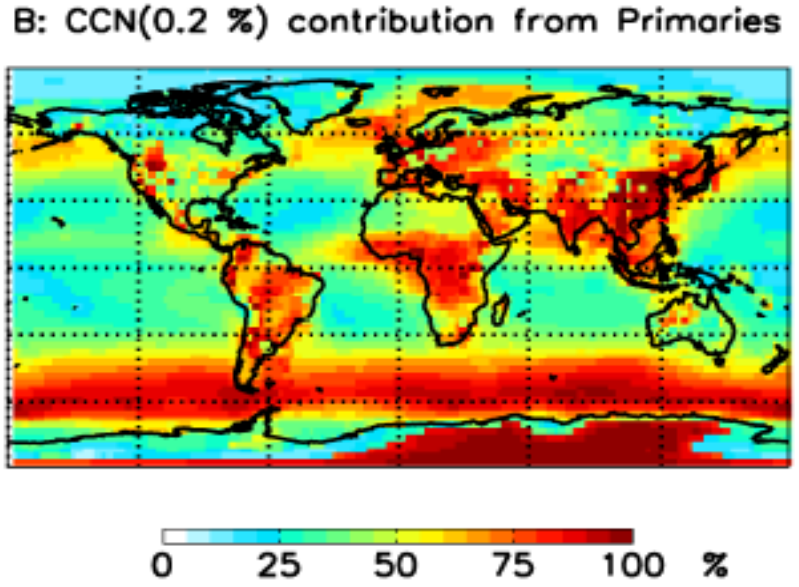
1. Child mortality only.

Source: OECD Environmental Outlook Baseline: output from IMAGE

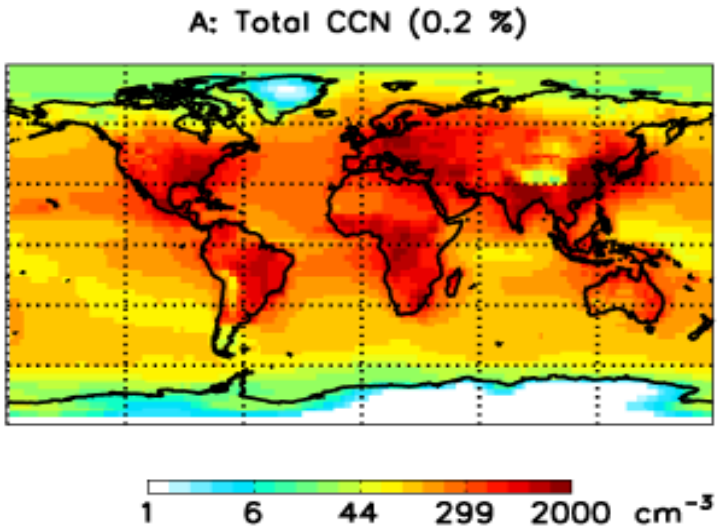
http://dx.doi.org/10.1787/888932571855

How important is nucleation?



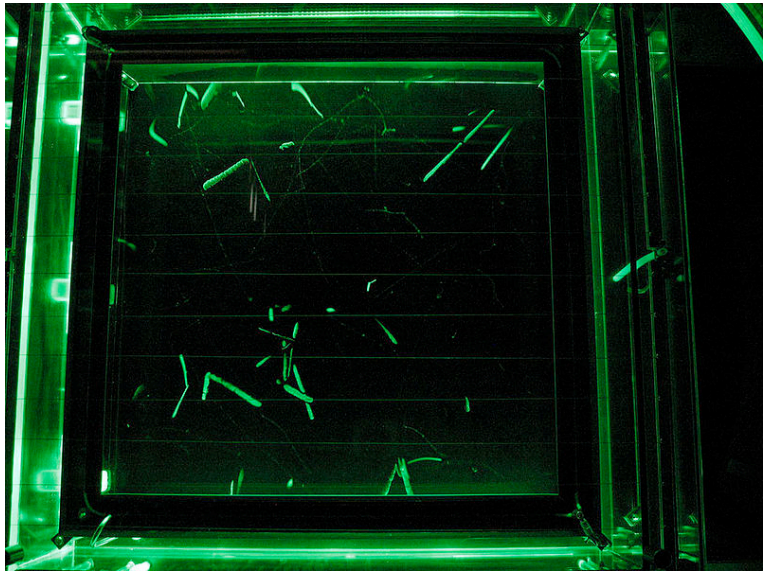


40-70% of CCN.



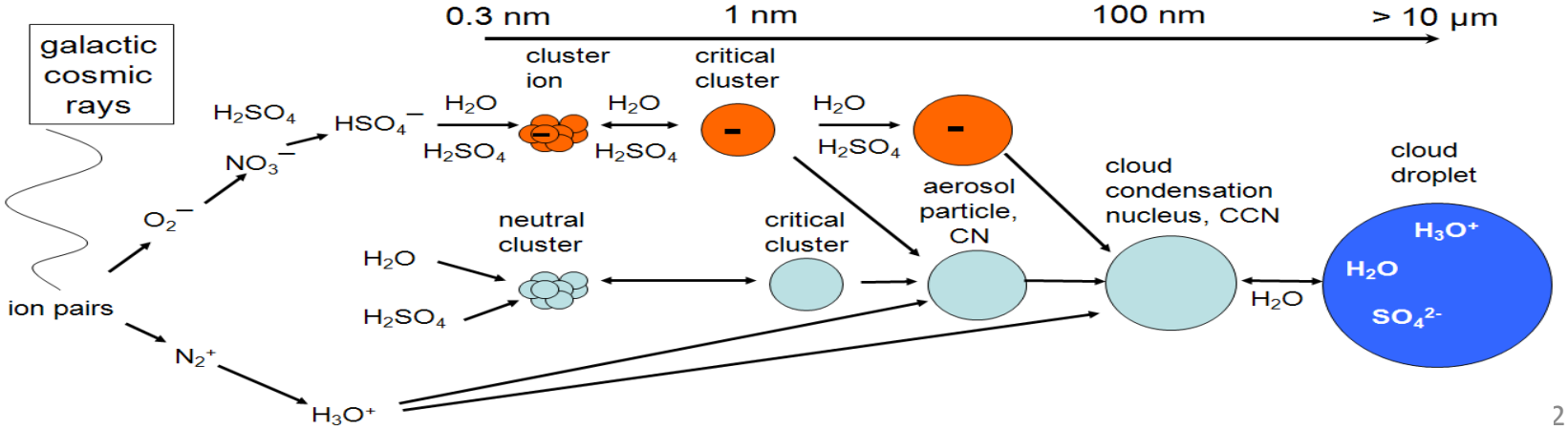
Merikanto et al, ACP 9 8601 (2009)

Could cosmic rays affect clouds?



Saint Mary's University

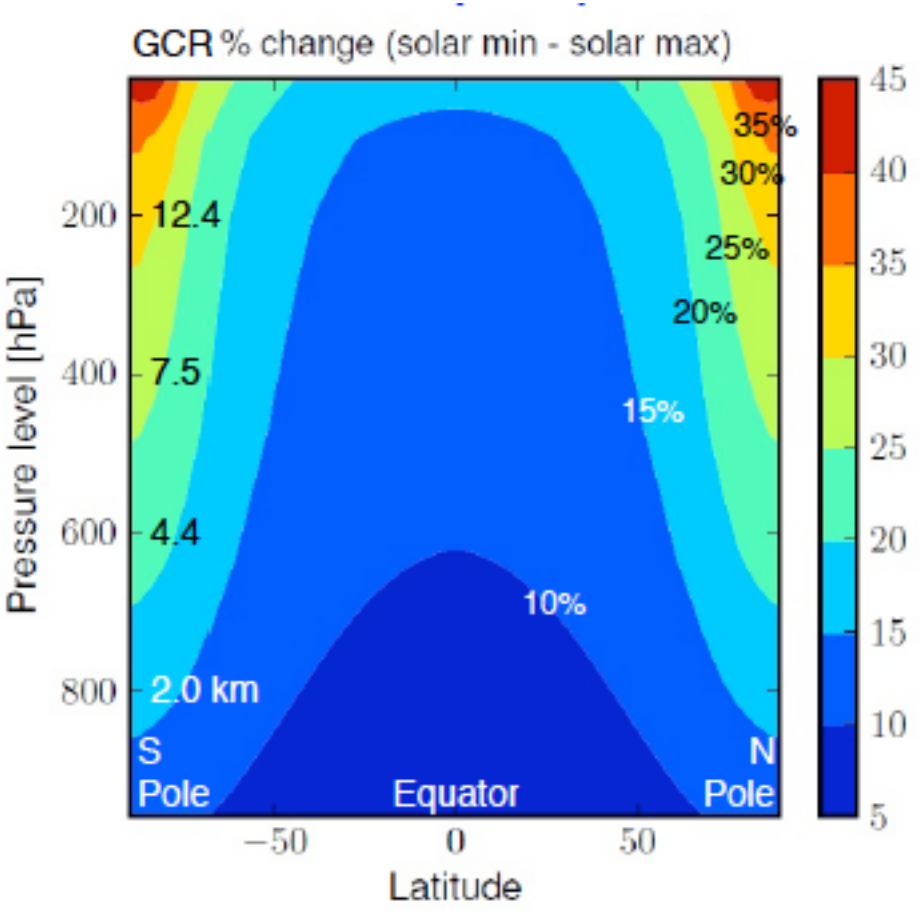
- Cosmic rays produce ~2 ion pairs $cm^{-3}s^{-1}$ at the surface and ~40 at the top of the troposphere
- Ions from radon also important
- Charge-stabilised clusters could mean more CCN



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 millenial time scales the GCR intensity can vary by a factor 3 or 4.

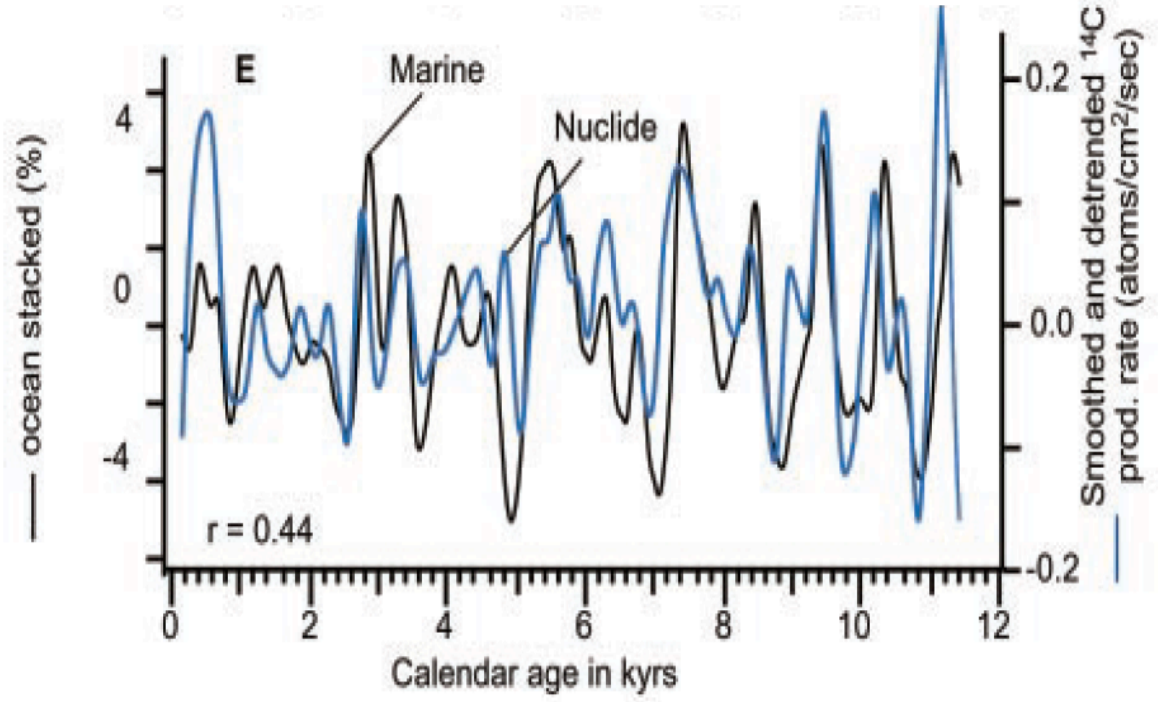


Could cosmic rays affect climate?

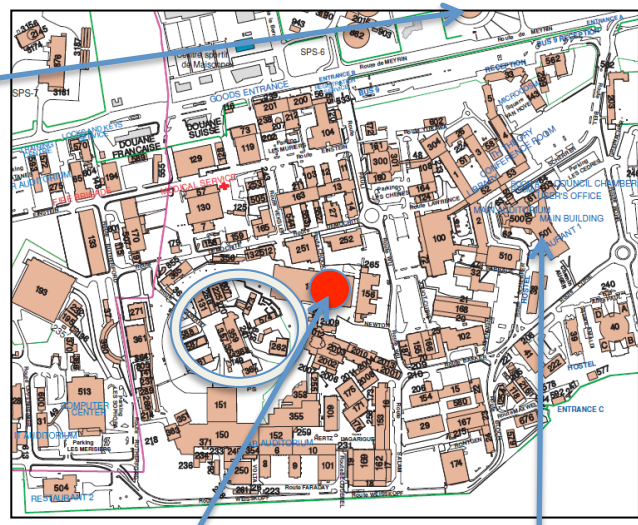
- Evidence for this in Bond et al, Science 2001: e.g. correlation of North Atlantic ice mass with C-14 (formed by GCRs)

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

Correlation ≠ causation!



The CLOUD experiment



- 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

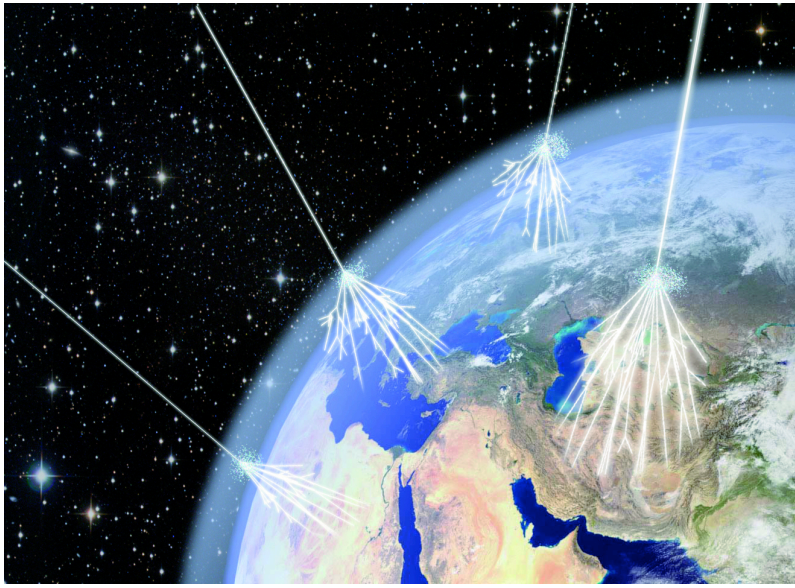
Stainless steel chamber

Add an atmosphere and sunlight

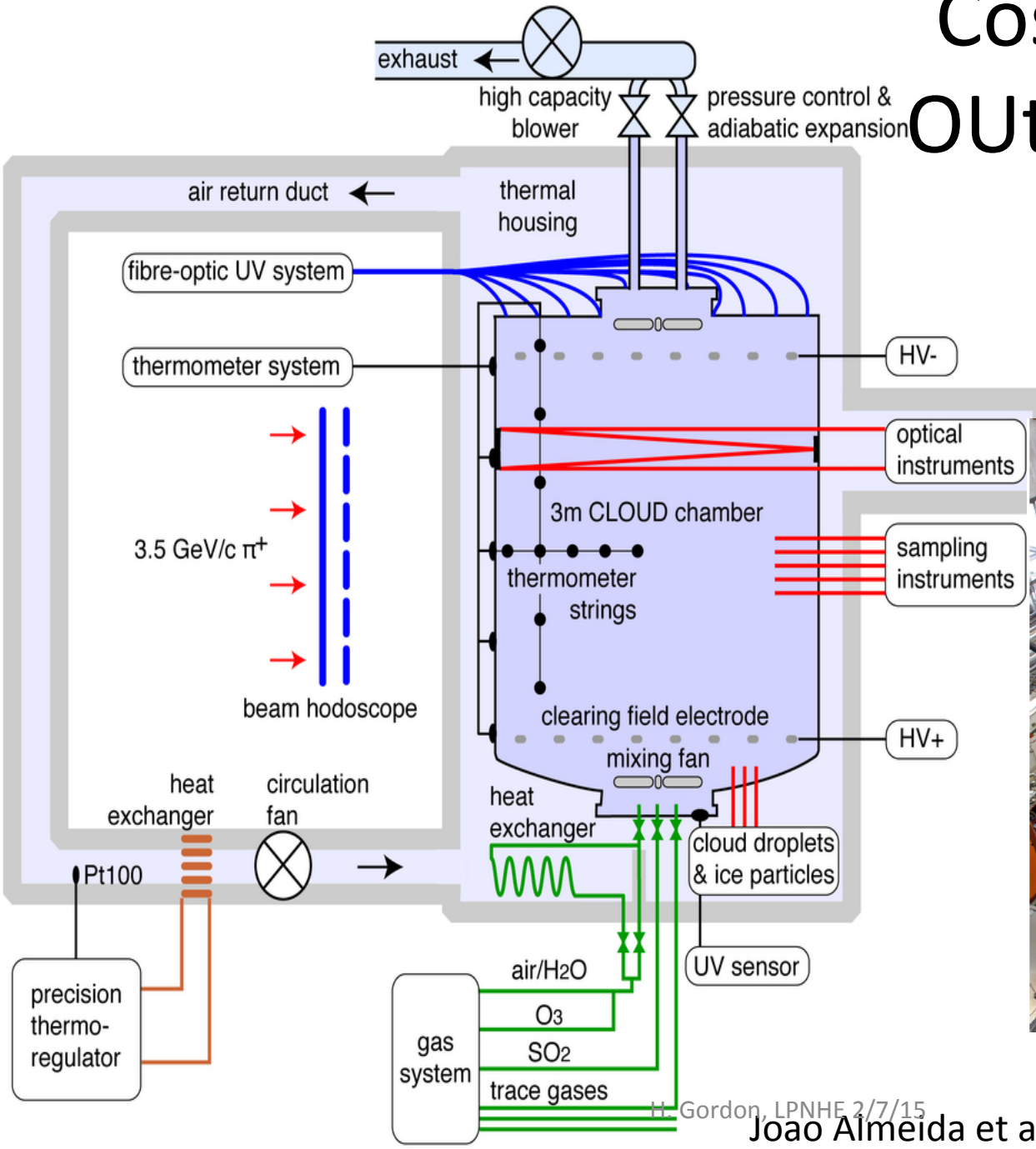
- Artificial air and trace gases from evaporators or bottles
- Ensure well mixed

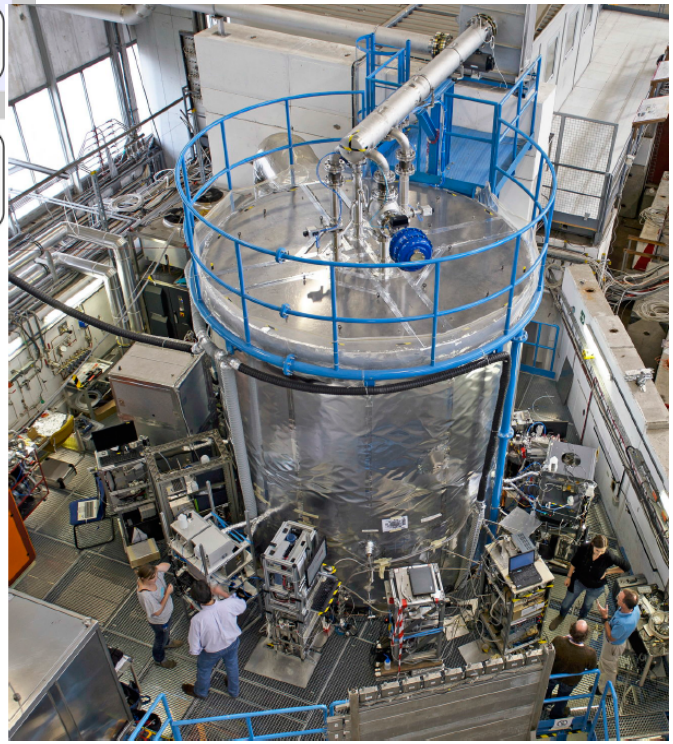
Add UV “sunlight” to photolyse ozone

Add or remove ions



Cosmics Leaving OUtdoor Droplets





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

The CLOUD collaboration

Aerodyne Research

UNIVERSITY OF LEEDS

MANCHESTER 1824
The University of Manchester

Stockholm University

UNIVERSITY OF HELSINKI

UNIVERSITY OF EASTERN FINLAND

FINNISH METEOROLOGICAL INSTITUTE

GOETHE UNIVERSITÄT FRANKFURT AM MAIN

CALIFORNIA INSTITUTE OF TECHNOLOGY 1891

CERN

LEIBNIZ-INSTITUT FÜR TROPOSPHÄRENFORSCHUNG

Carnegie Mellon University

SIM

KIT
Karlsruher Institut für Technologie

PAUL SCHERRER INSTITUT
PSI

TOFWERK
Time-of-Flight Mass Spectrometry

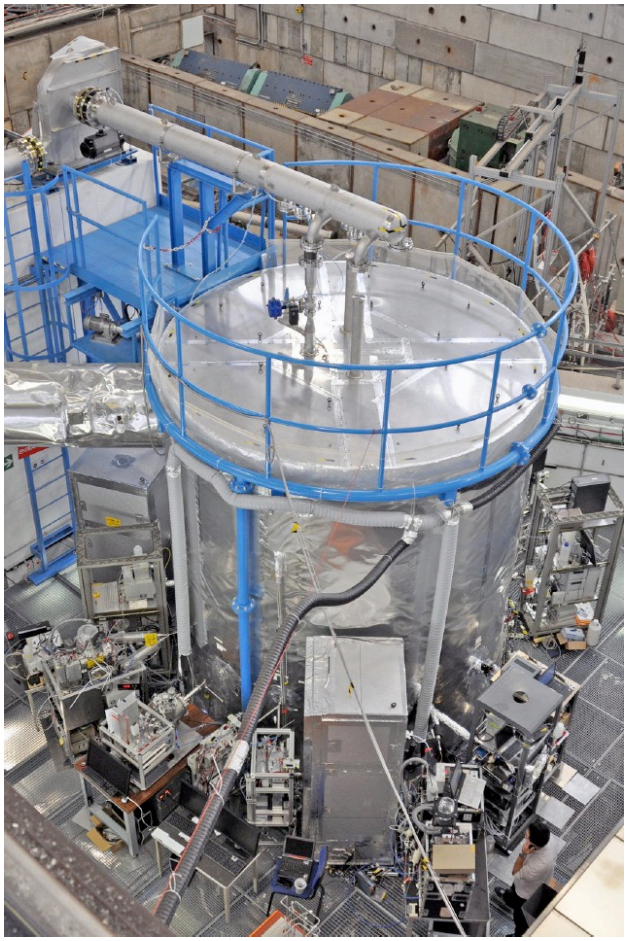
IONICON

universität innsbruck

universität wien

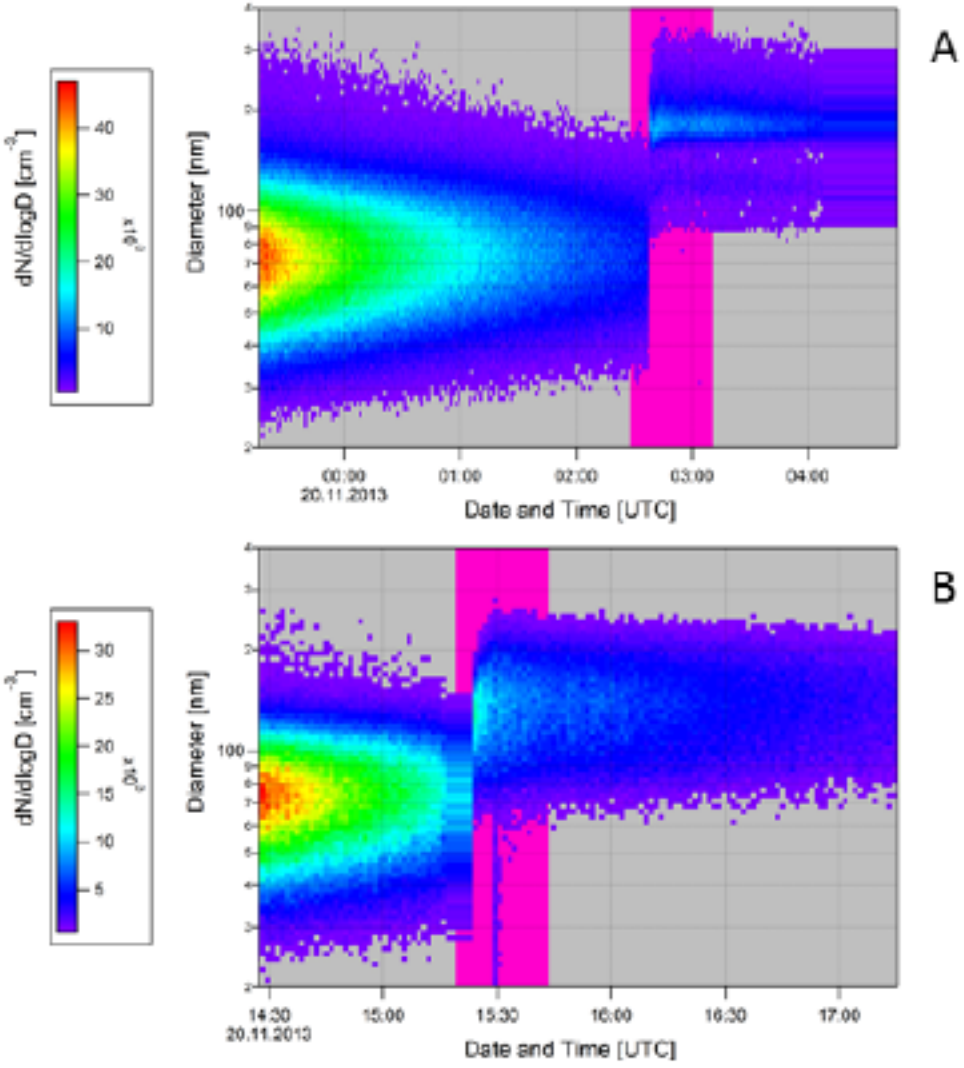
Cloudy experiments

- Inject water and cloud condensation nuclei into chamber, and pressurise to 200mbar over atmospheric pressure
- Then fast expansion, during which $P^{1-\gamma}T^{\gamma}$=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



Aqueous chemistry and cloud-aerosol interaction

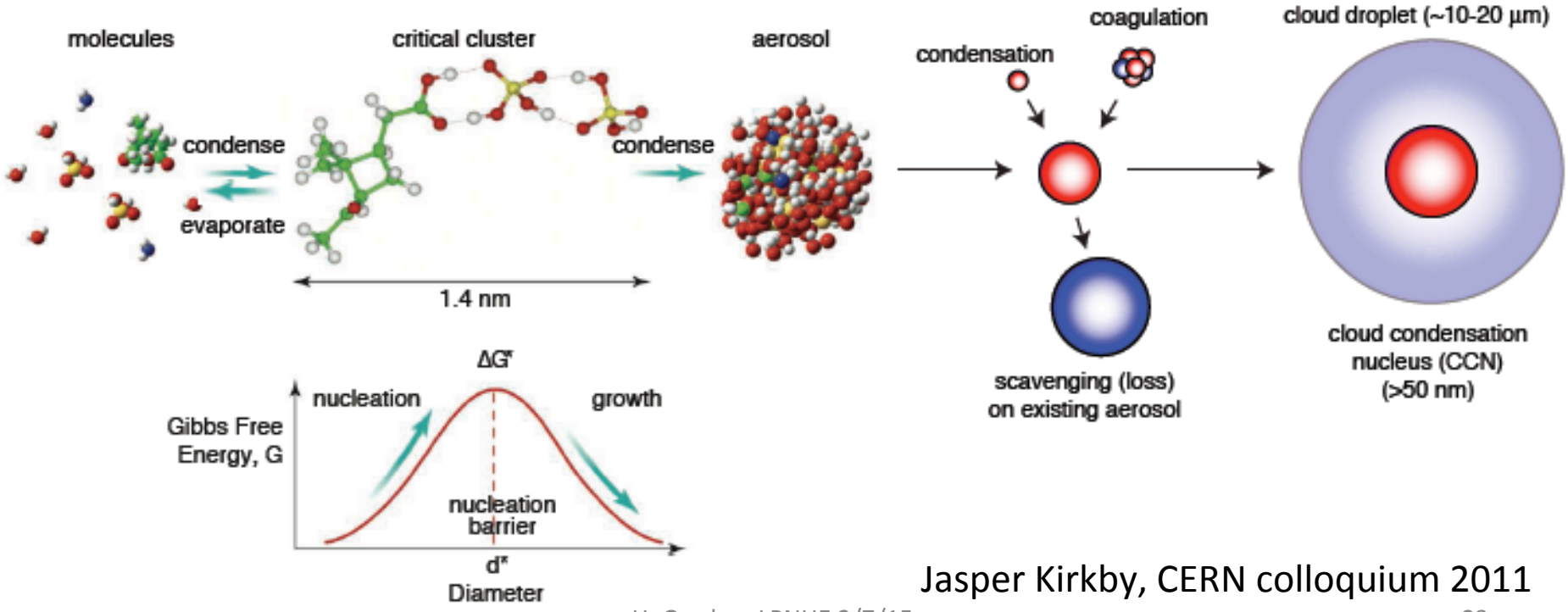
- Aerosol size distribution (right, at +10C and -10C) grows during a cloud (pink period) due to reactive uptake of SO2 on ammonium sulphate seed aerosol



SPSC report, 2014

Nucleation experiments

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

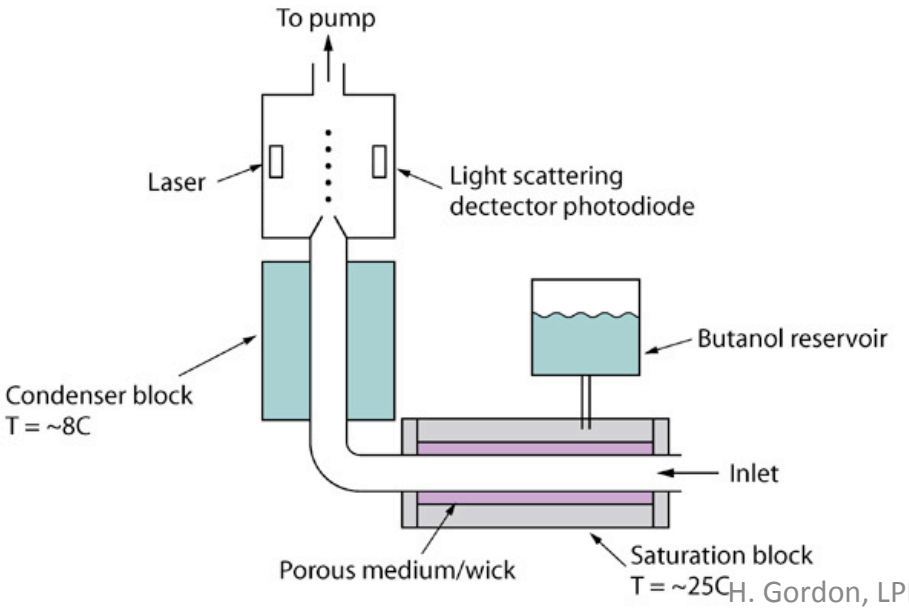


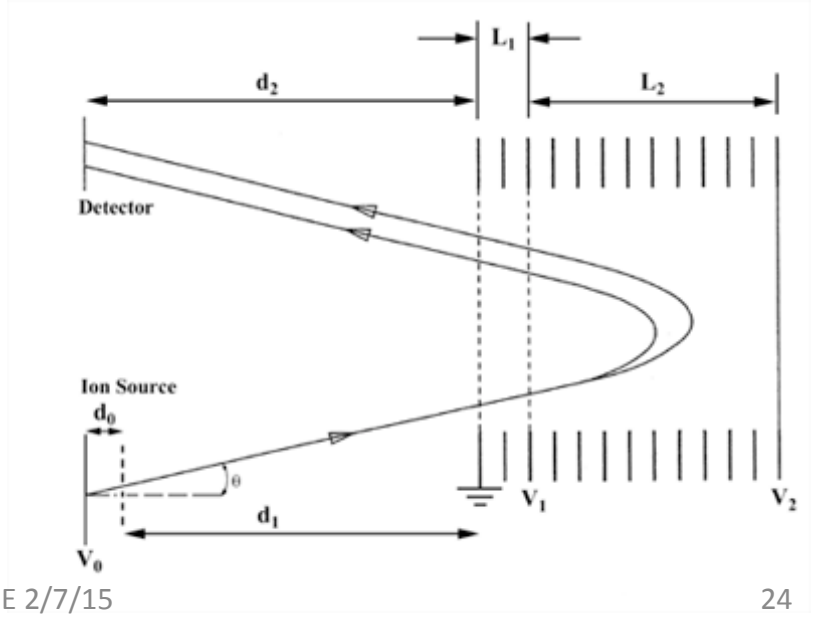
Jasper Kirkby, CERN colloquium 2011

Nucleation instrumentation

- Particle counters
- How many particles of a given size?
- CPC, SMPS, CCNC, nRDMA

- Mass spectrometers (mostly TOF)
- What do the particles contain?





Mass spectrometers in use

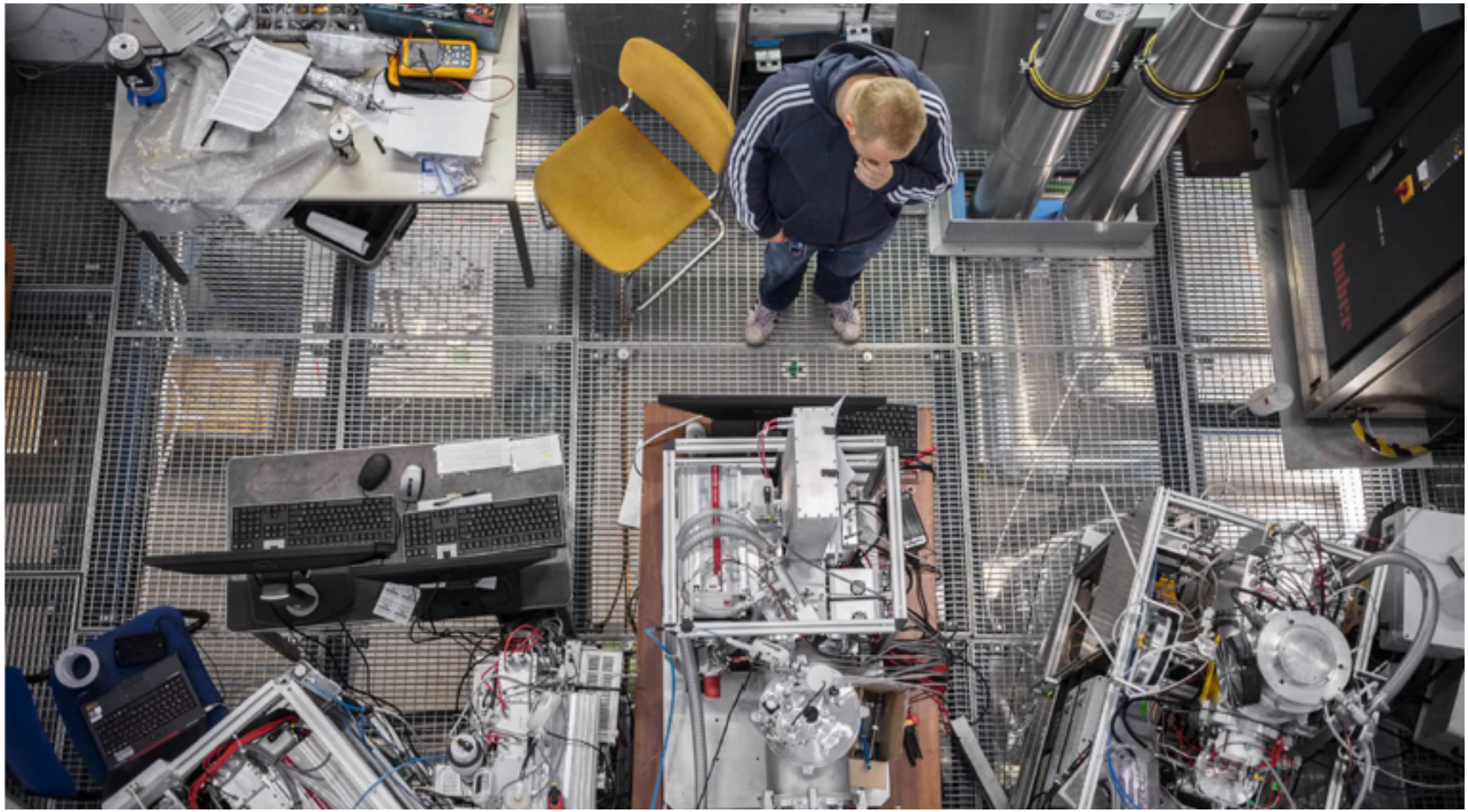


Photo: Andri Pol

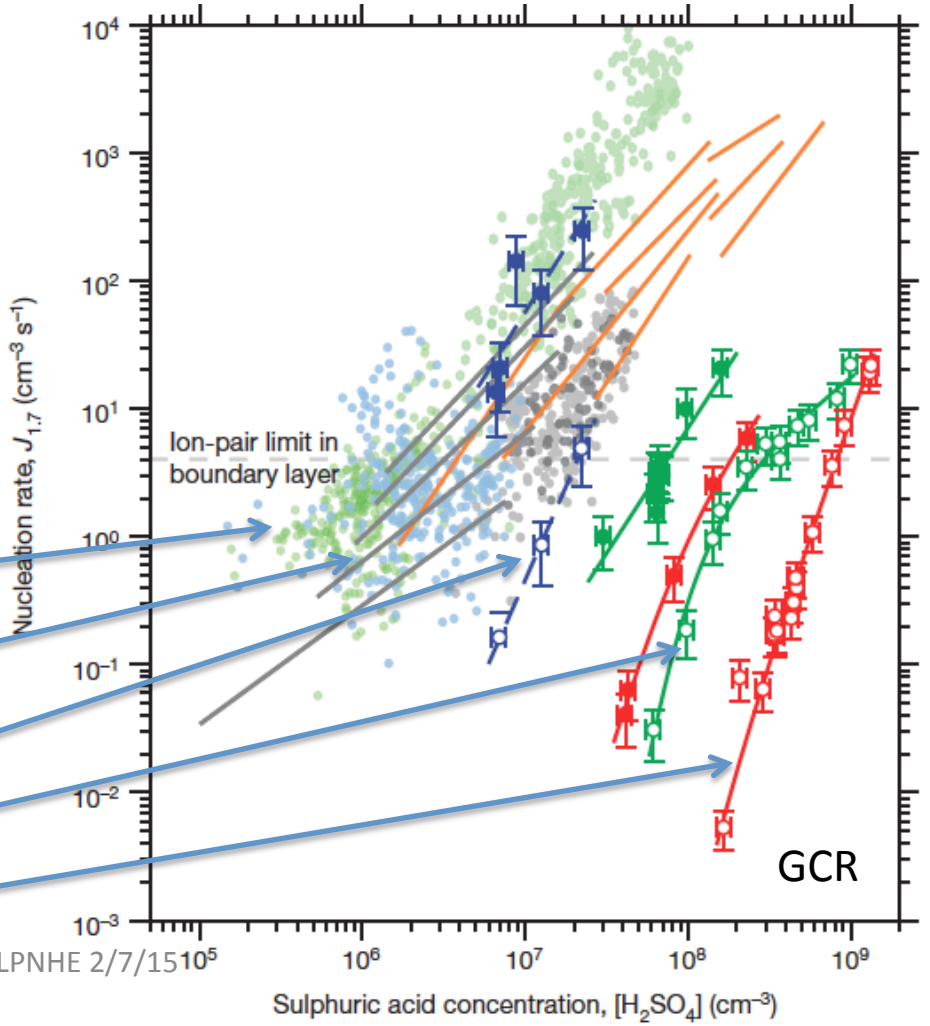
LETTER

doi:10.1038/nature10343

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

Jasper Kirkby[1], Joachim Curtius[2], João Almeida[2,3], Eimear Dunn

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



Atmospheric data (dots)

Other lab measurements (lines)

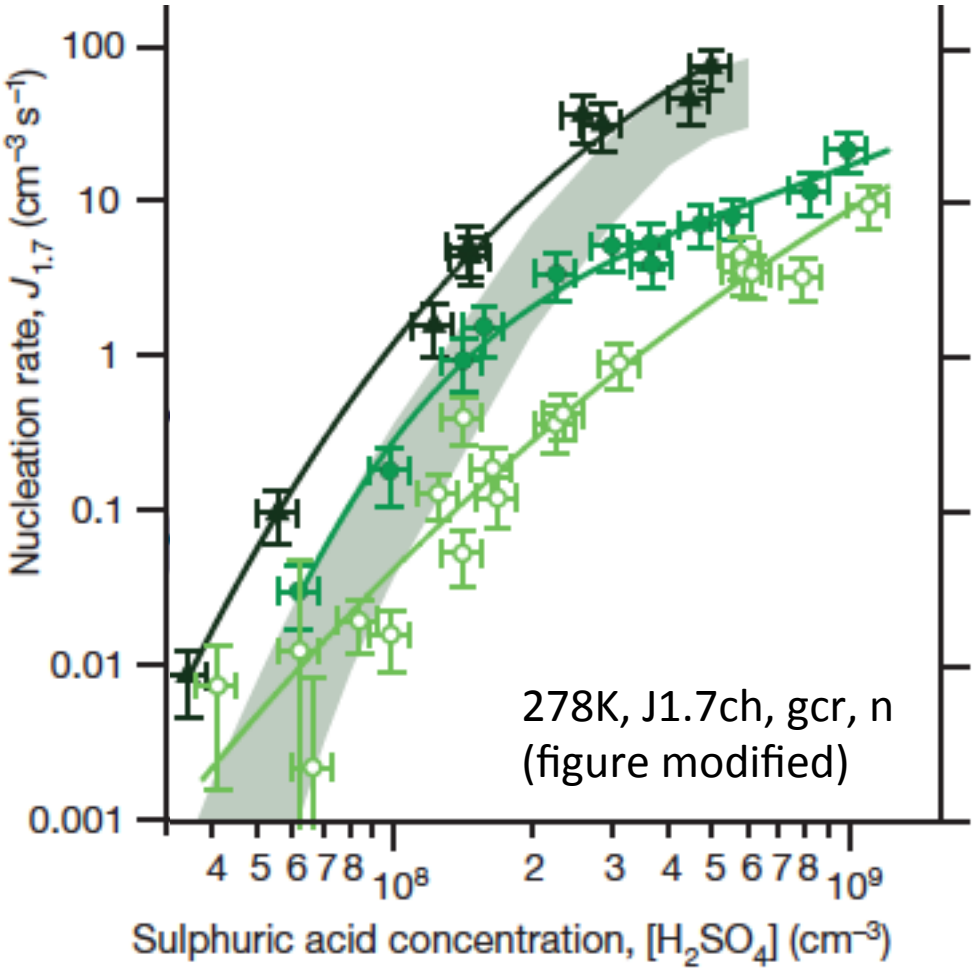
CLOUD data at 248K (blue, filled=higher NH3)

CLOUD data at 278K (green)

CLOUD data at 292K (red)

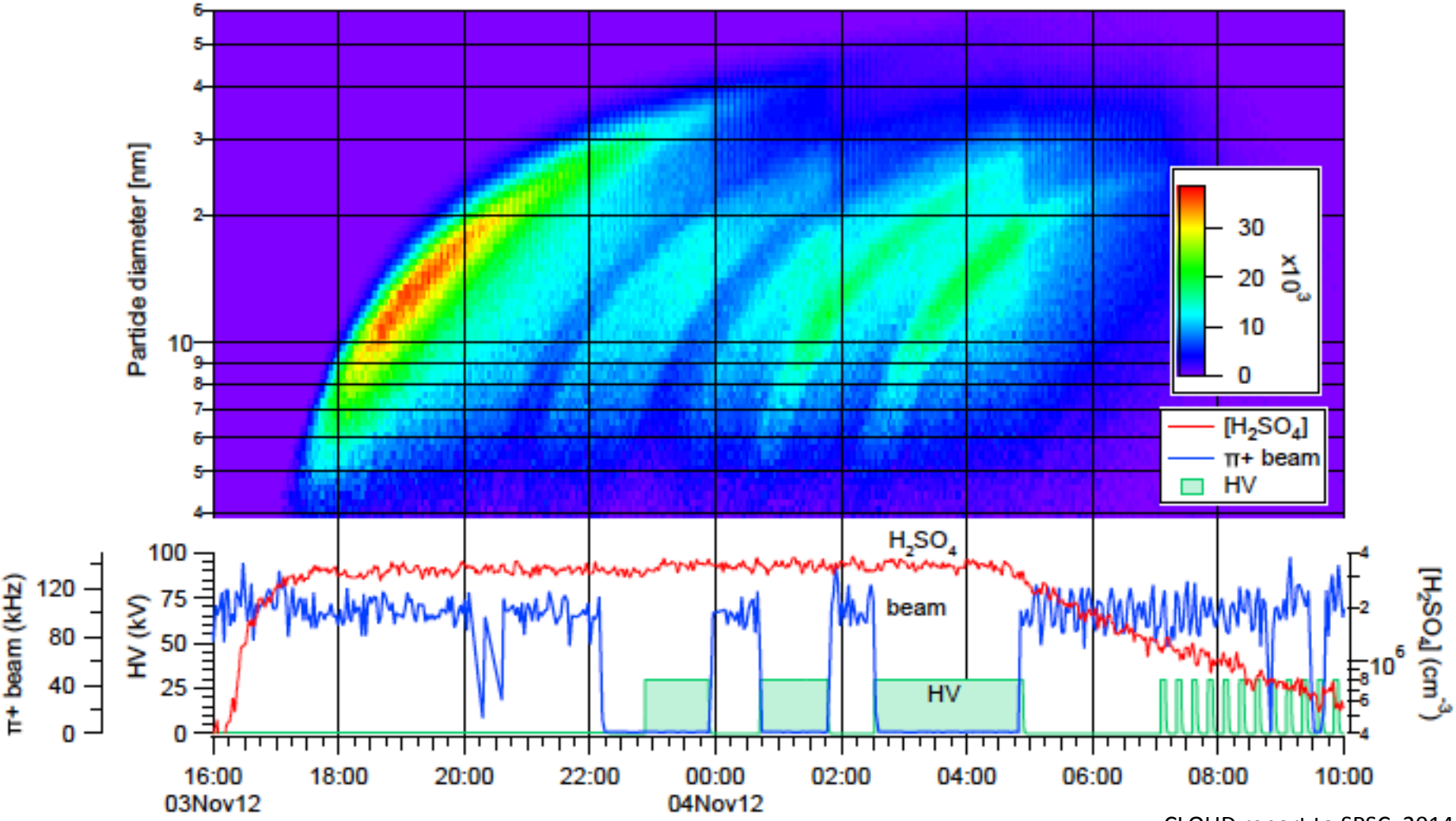
Ion-induced nucleation

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

LETTER

OPEN

doi:10.1038/nature12663

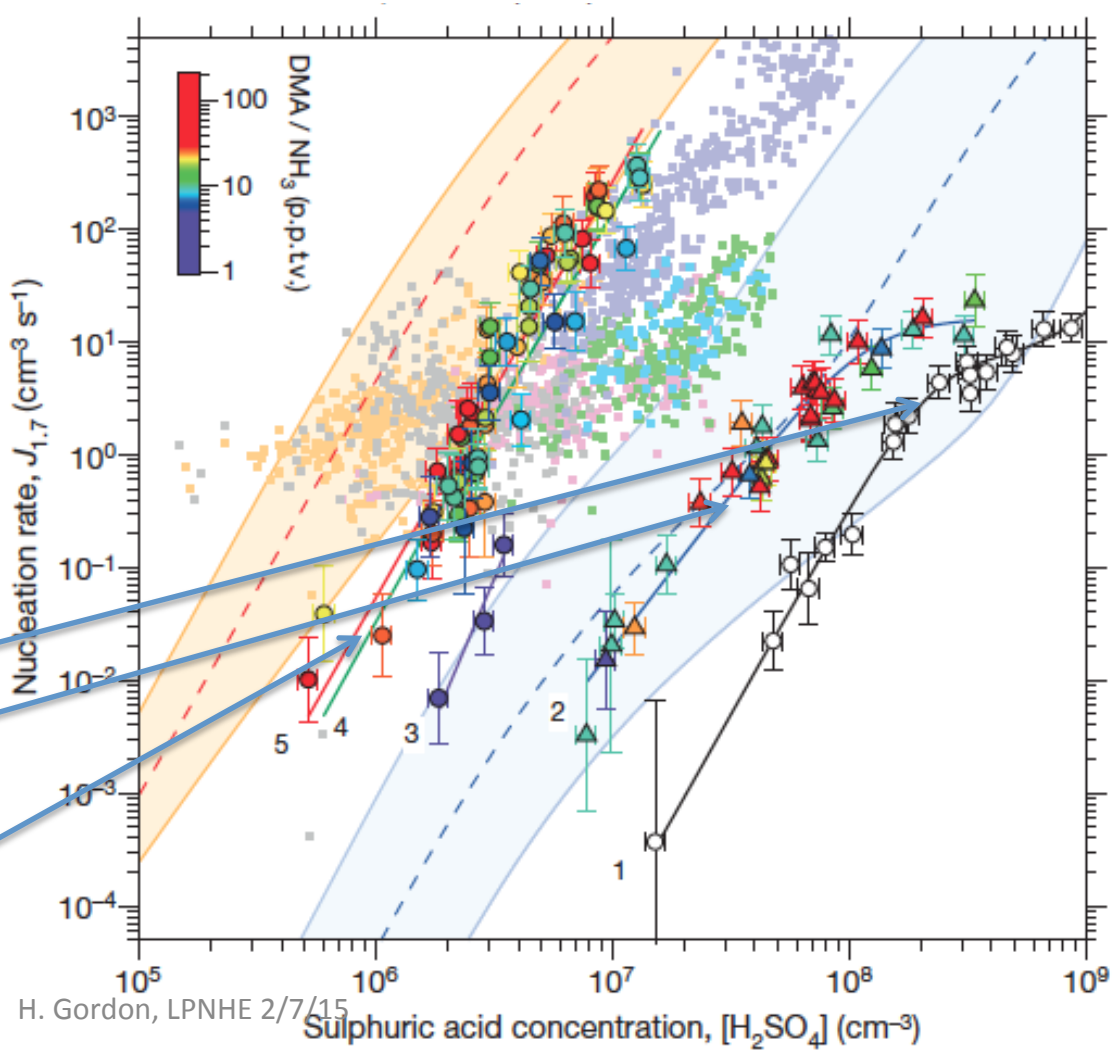
Molecular understanding of sulphuric acid–amine particle nucleation in the atmosphere

João Almeida[1,2], Siegfried Schobesberger[3], 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)

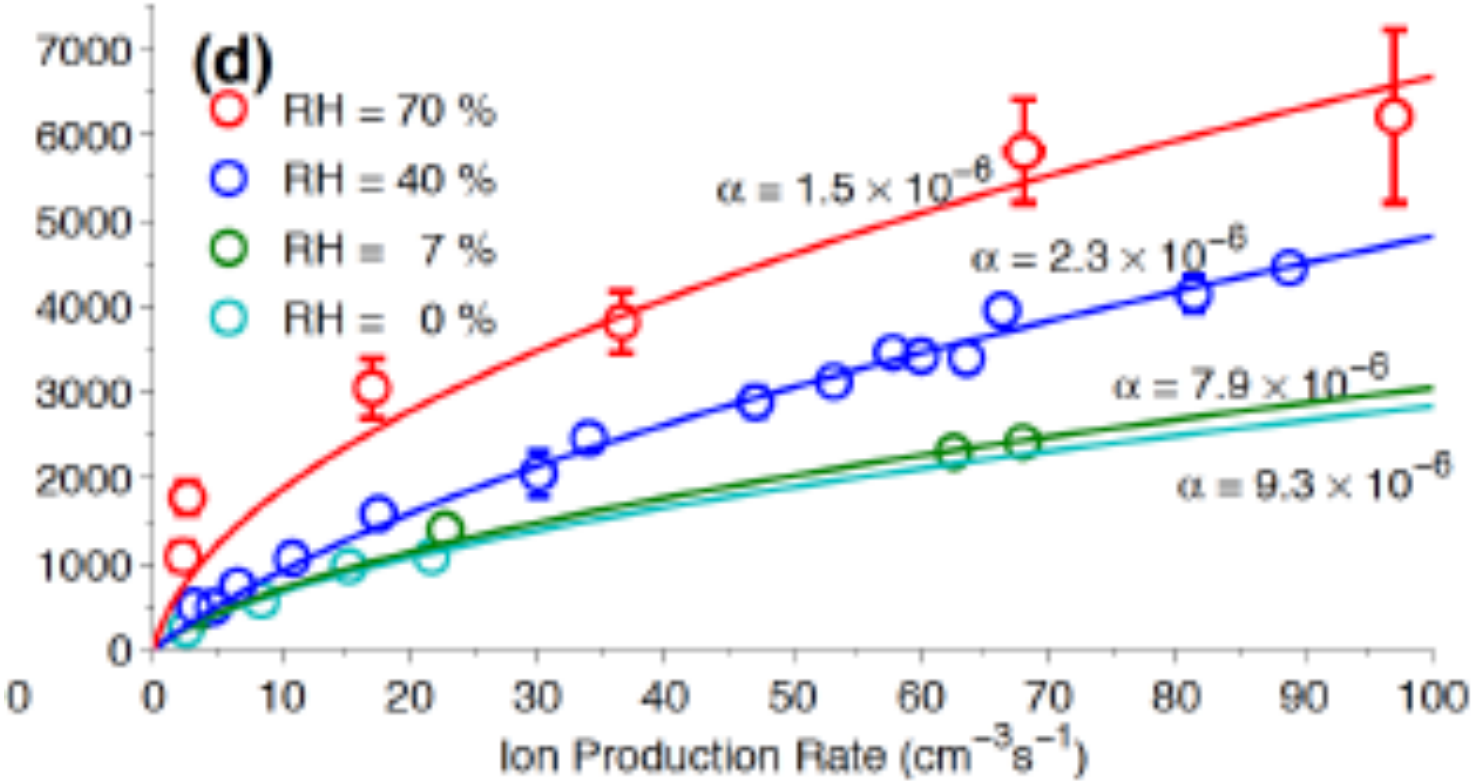
Sulphuric acid (+water) H2SO4 and ammonia (colour scale for NH3), + theory blue band

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



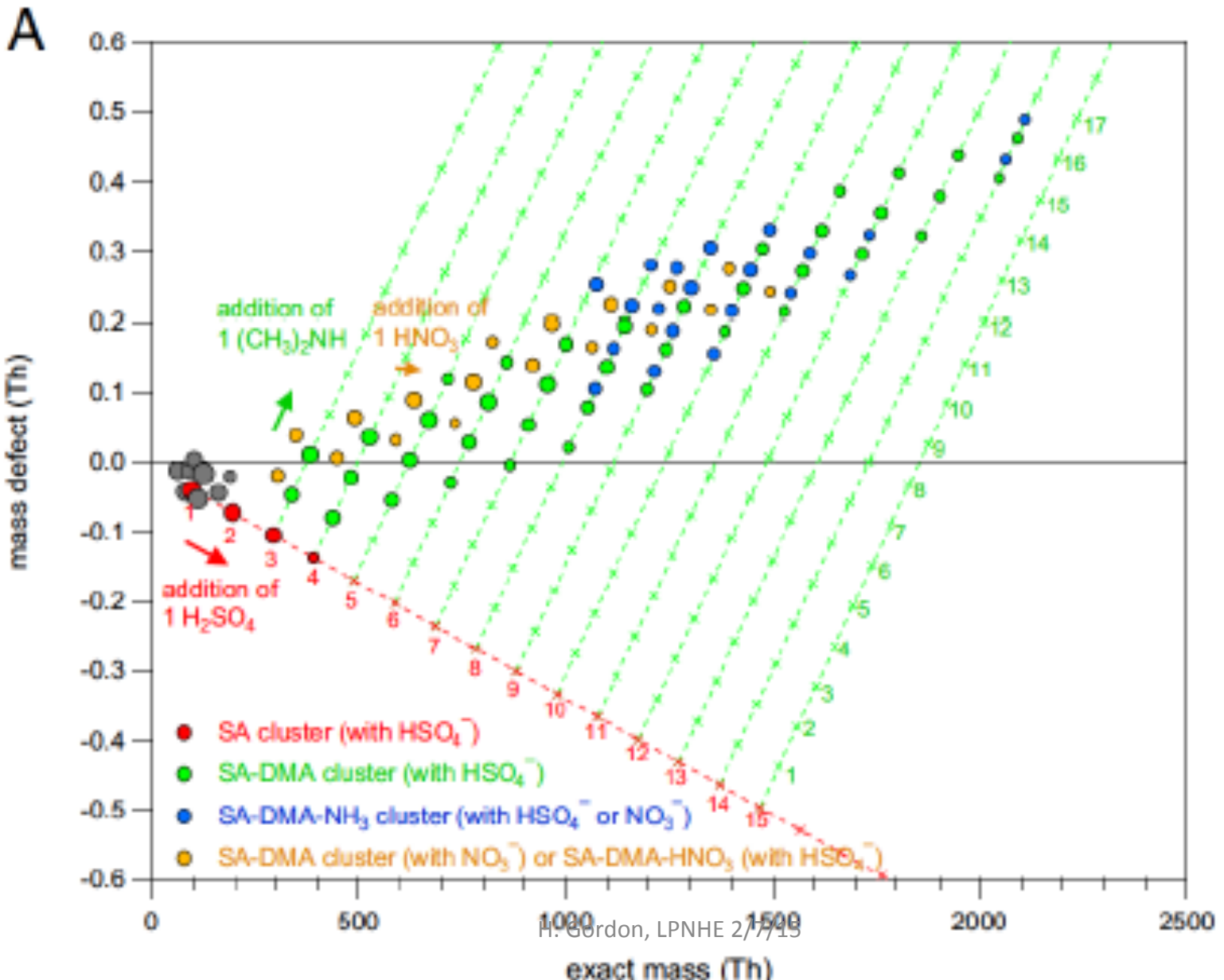
CLOUD results since July 2014

- Ion-ion recombination in the atmosphere: dependence on T, RH



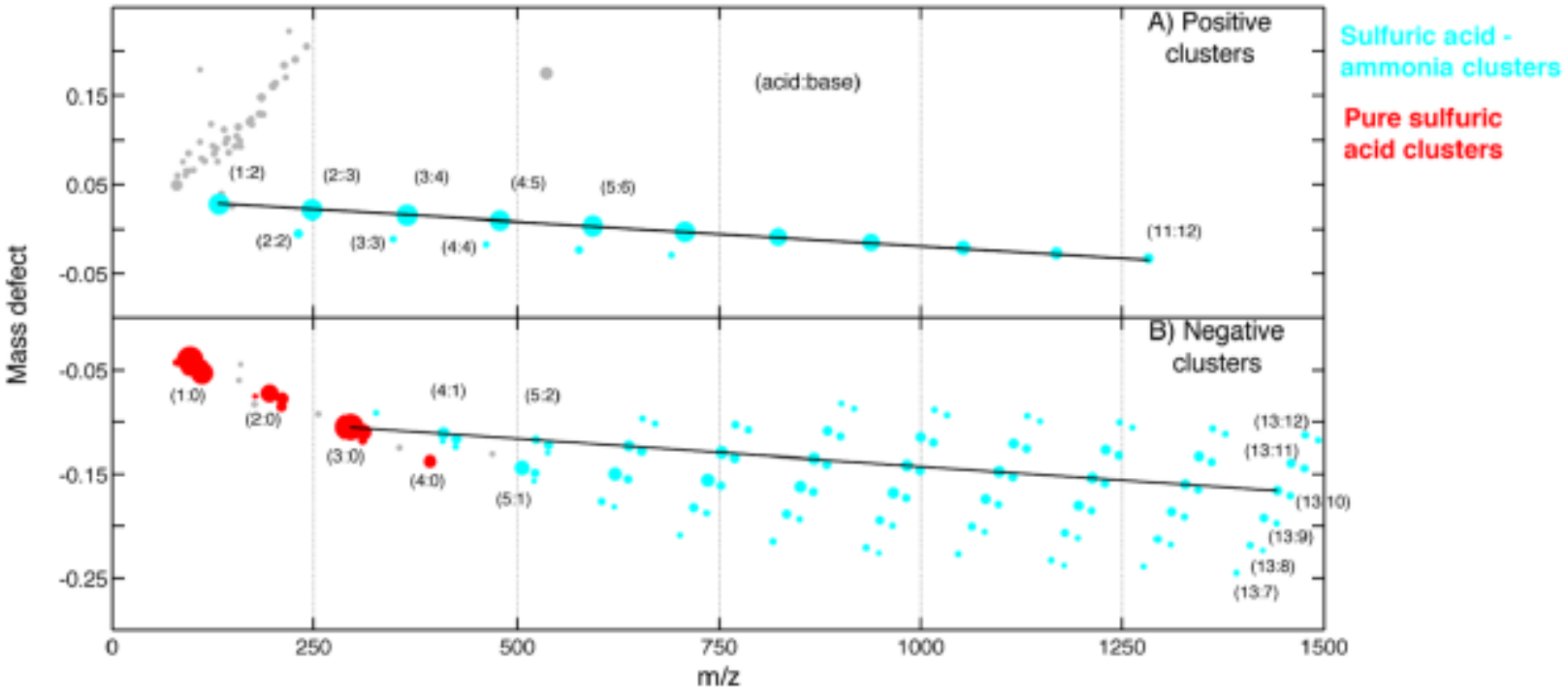
Franchin A. et al, Experimental investigation of ion-ion recombination at atmospheric conditions, Atmos. Chem. Phys. Discuss. 15, 3667 (2015)

Neutral clusters



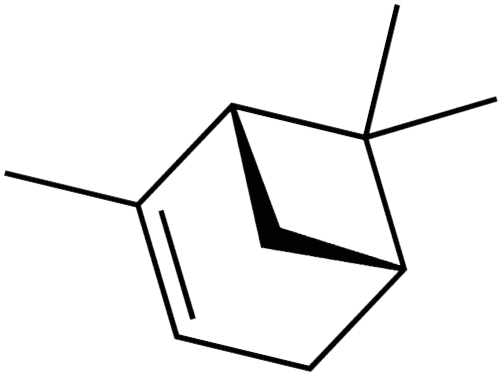
Kuerten A. et al, Neutral molecular cluster formation of sulfuric acid-dimethylamine observed in real time under atmospheric conditions, Proc. Nat. Acad. Sci. 111, 15019 (2014)

Charged clusters



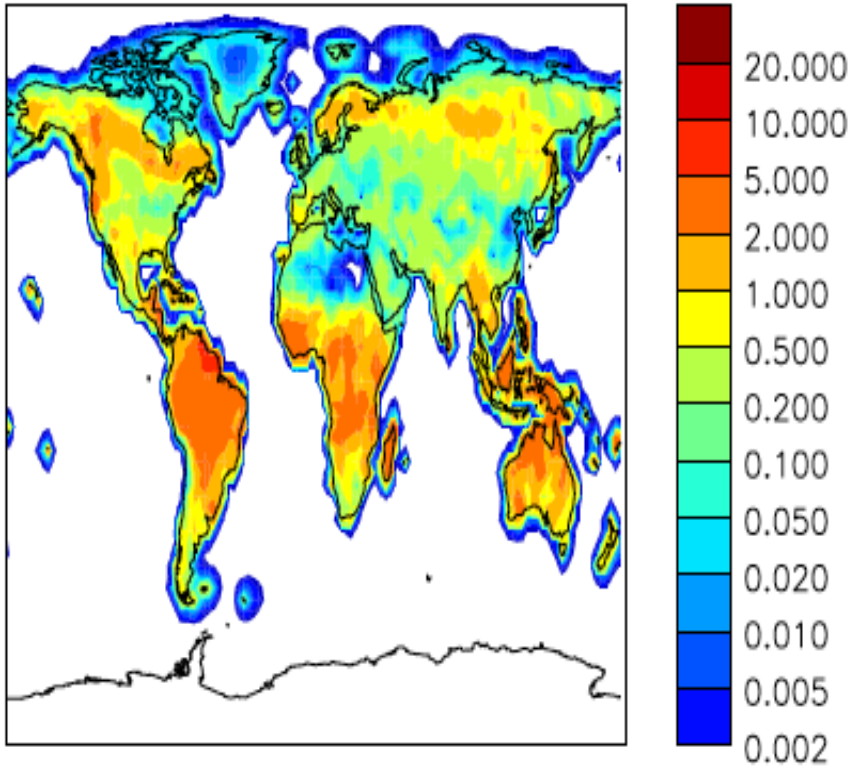
Bianchi et al, Insight into acid-base nucleation experiments by comparison of the chemical composition of positive, negative, and neutral clusters, Envir. Sci. Tech. 48 13675 (2014)

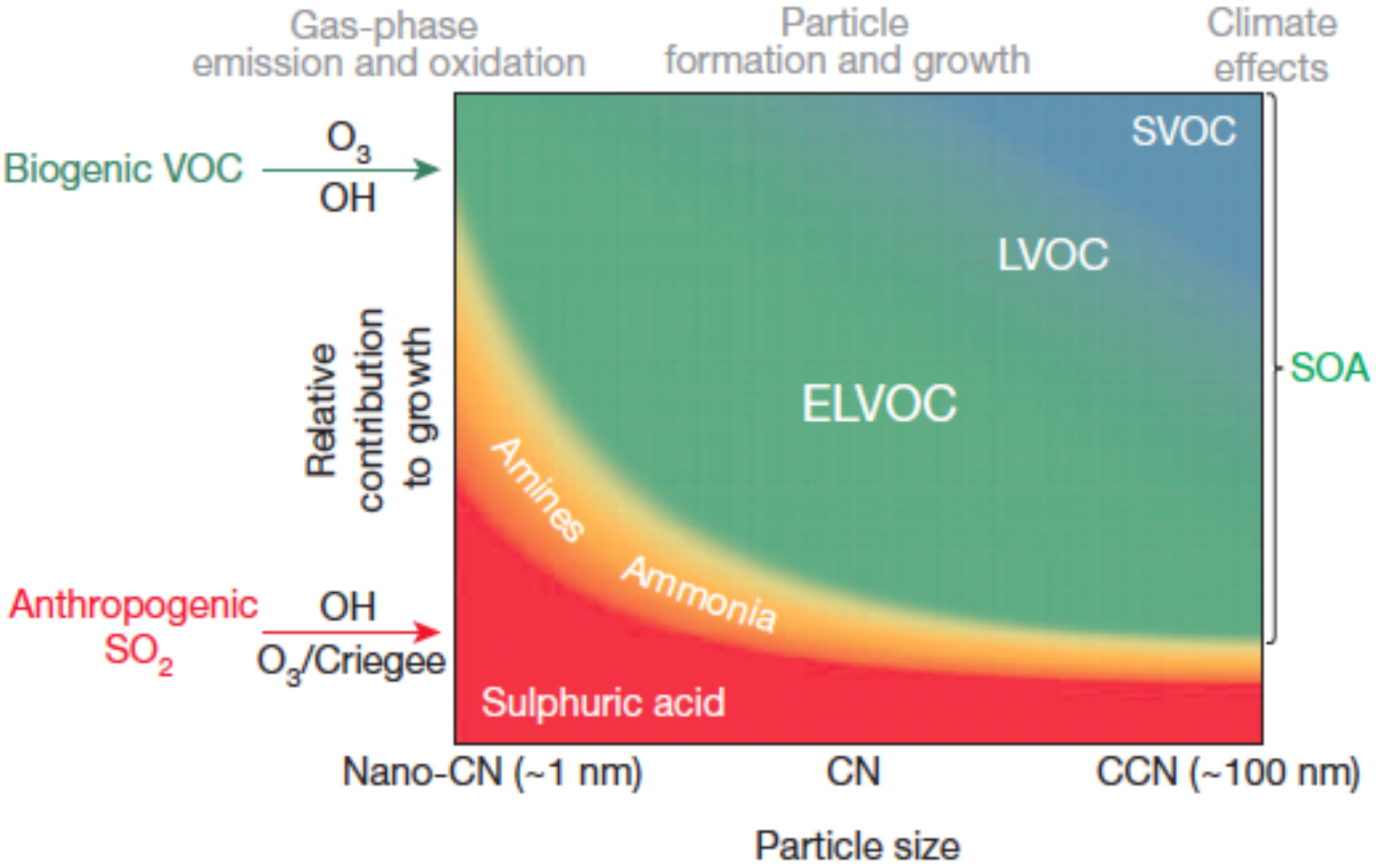
Pinenes and ELVOCs



- 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 NOx 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)





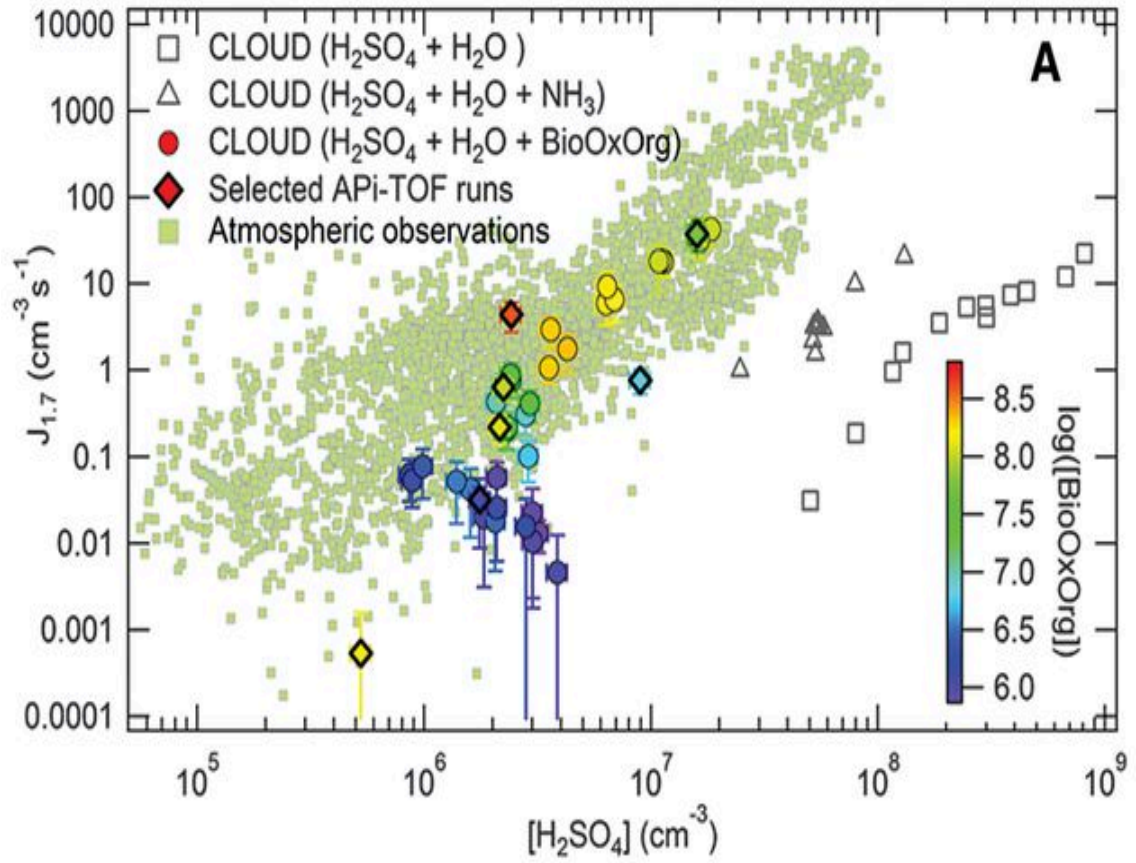
Ehn et al, Nature **506** 476 (2014)

Science
AAAS

Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles

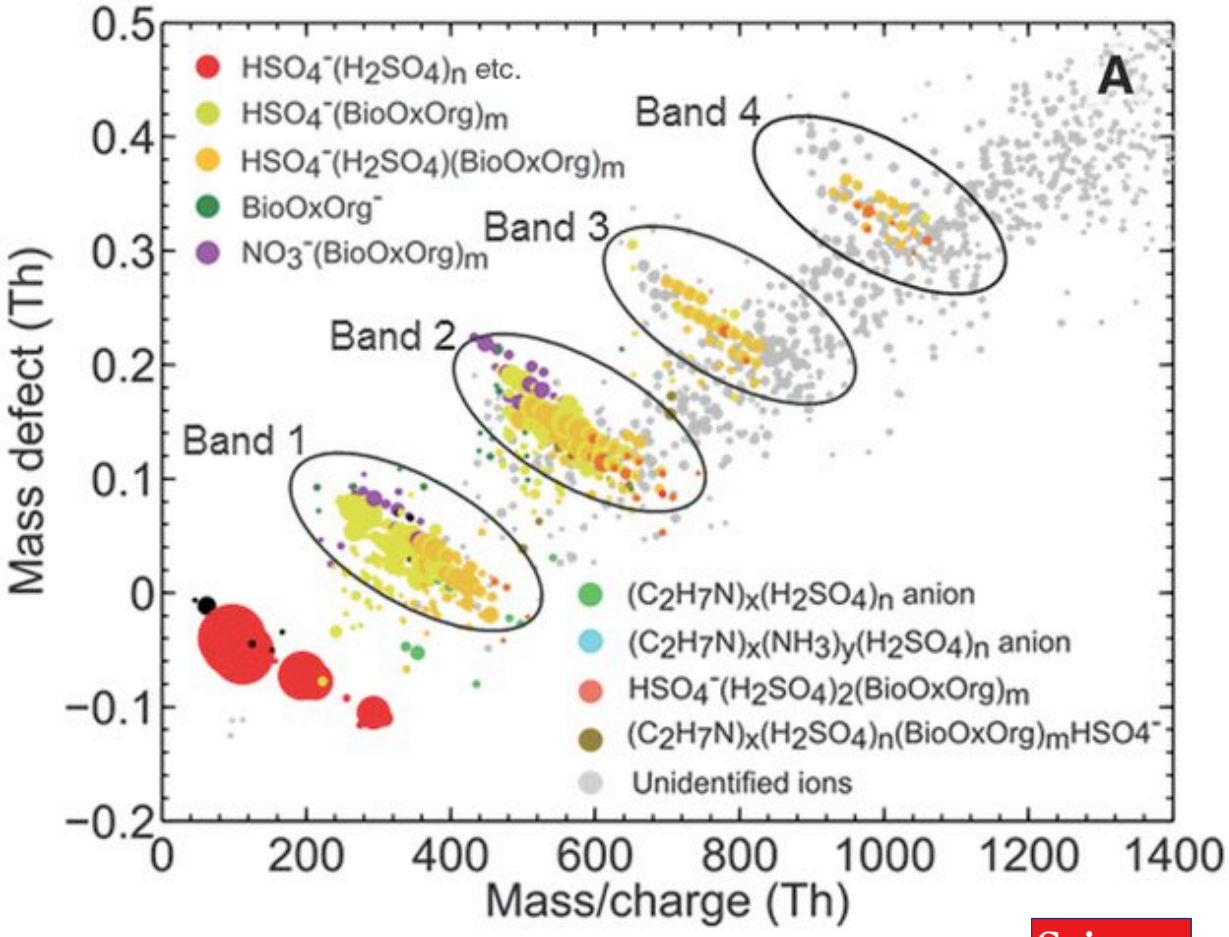
Francesco Riccobono *et al.*

Science **344**, 717 (2014);
DOI: 10.1126/science.1243527



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

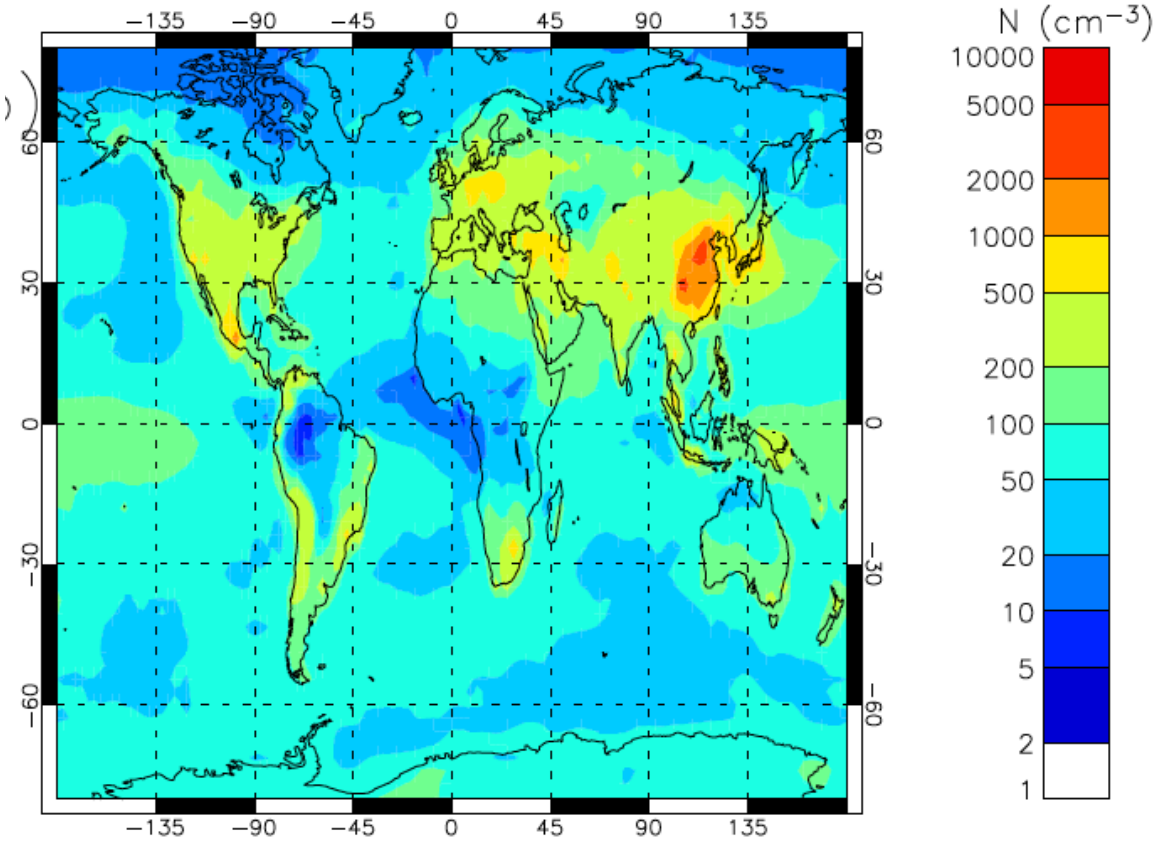
Nucleation as seen by TOF-MS



Science
AAAS

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



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

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

Host Chemical Transport Model

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

https://www.youtube.com/watch?v=zgFSpHL2k8I

GLOMAP

- Aerosol size spectrum (~ 3 nm – 24 μm)
- 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 H_2SO_4/H_2O nucleation
- Condensational growth

Coagulation
- Semi implicit fast numerical solution

Hygroscopic Growth
- Equilibrium size given by Kohler equation

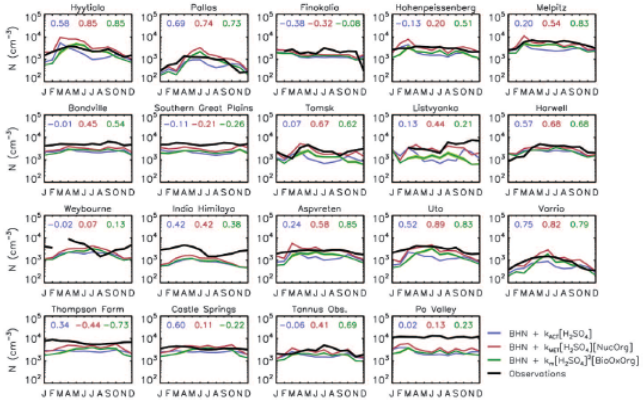
Removal

Dry Deposition
- Dry deposition of aerosol

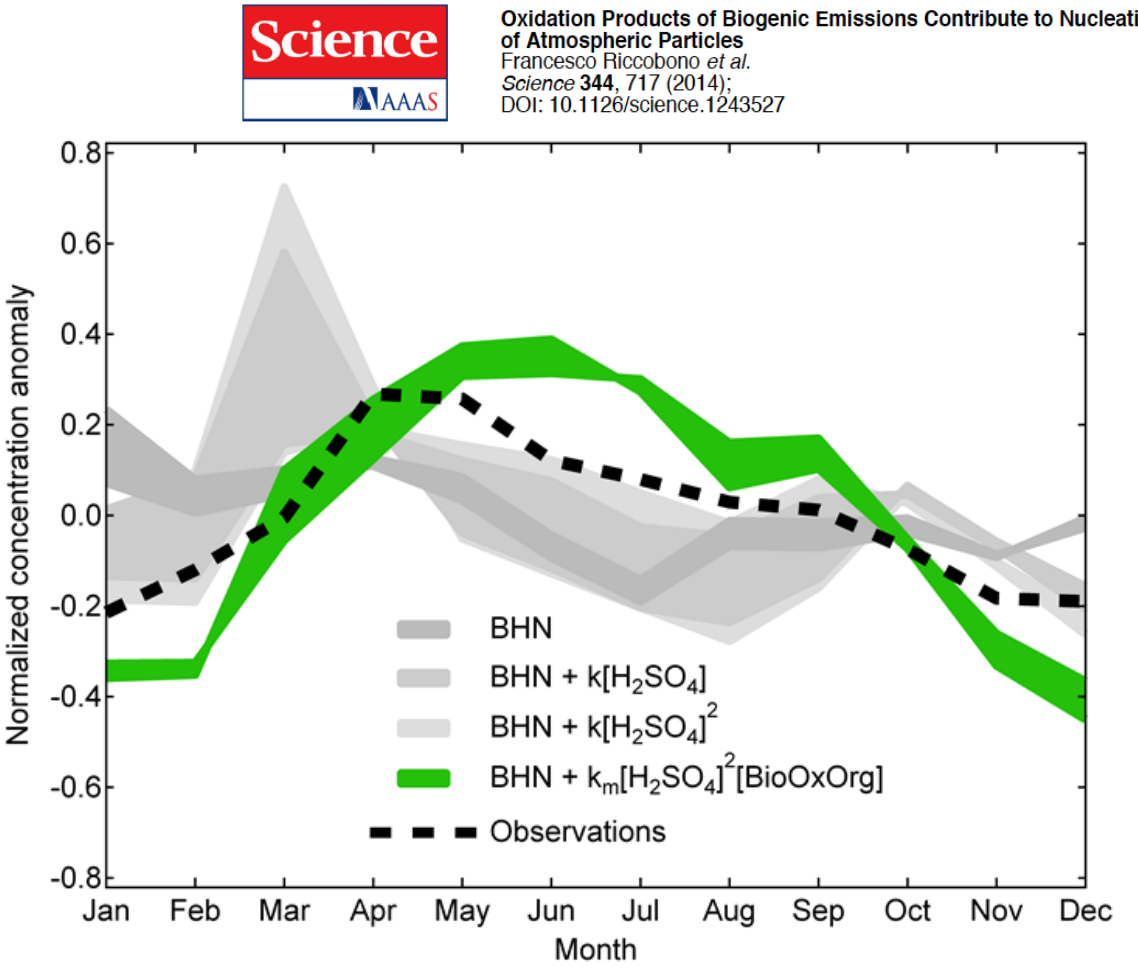
Clouds
- Prognostic convective and frontal rain
- In-cloud and below-cloud aerosol/gas scavenging
- Cloud-processing in diagnosed low-level cloud

Dominick Spracklen

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

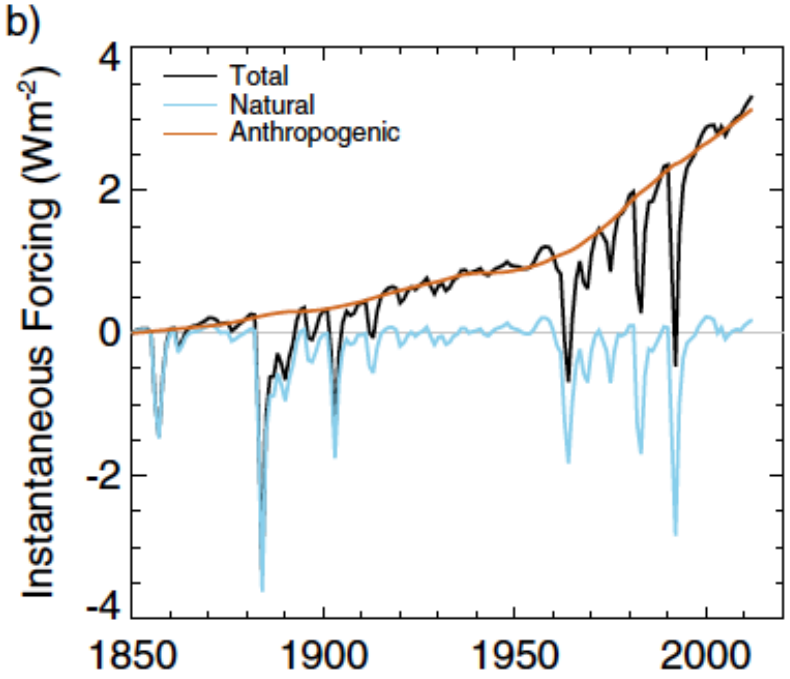


Improved explanation of present-day particle formation in the atmosphere in the summer months

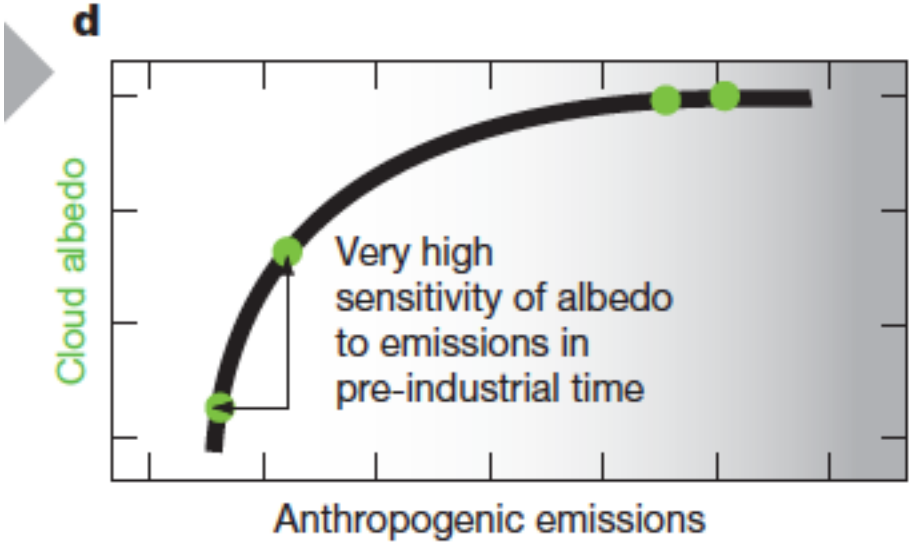


Big questions in aerosol science

- Carslaw et al (Nature, 2013): Pre-industrial climate is much more sensitive to aerosol perturbations than present-day climate.
- Imprecise knowledge of aerosol forcing allows models with a range of climate sensitivity to reproduce the warming already observed (Kiehl, J. T. et al (2007))



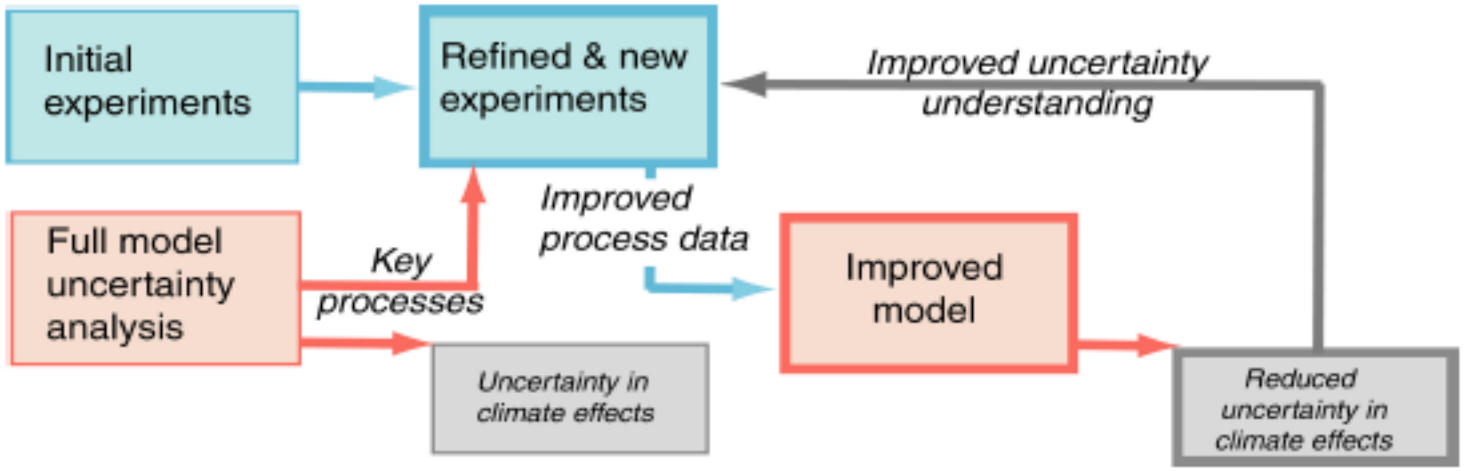
$$\Delta T = \lambda \Delta F$$



Pre-industrial atmosphere

- SO2 emissions ~75% lower
- NH3 emissions ~80% lower
- Biogenic organic emissions not too different
- Uncertainties enormous
- 45 per cent of the variance of aerosol forcing since about 1750 arises from uncertainties in natural emissions, 34 per cent of the variance is associated with anthropogenic emissions

Next steps for CLOUD



- Daytime nucleation of biogenic organic compounds
- Temperature dependence of organic nucleation
- Key question: can we get significant new particle formation without sulphuric acid, ozone, NOx, etc? (i.e. in the unpolluted pre-industrial atmosphere over land)

Summary

http://cloud.web.cern.ch

- 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
- Also cloud chemistry and ice microphysics studies
- Possibility to use CLOUD to study pre-industrial atmosphere
- As nucleation is a much more complicated process than originally thought, CLOUD results uniquely able to give predictive power to atmospheric aerosol models

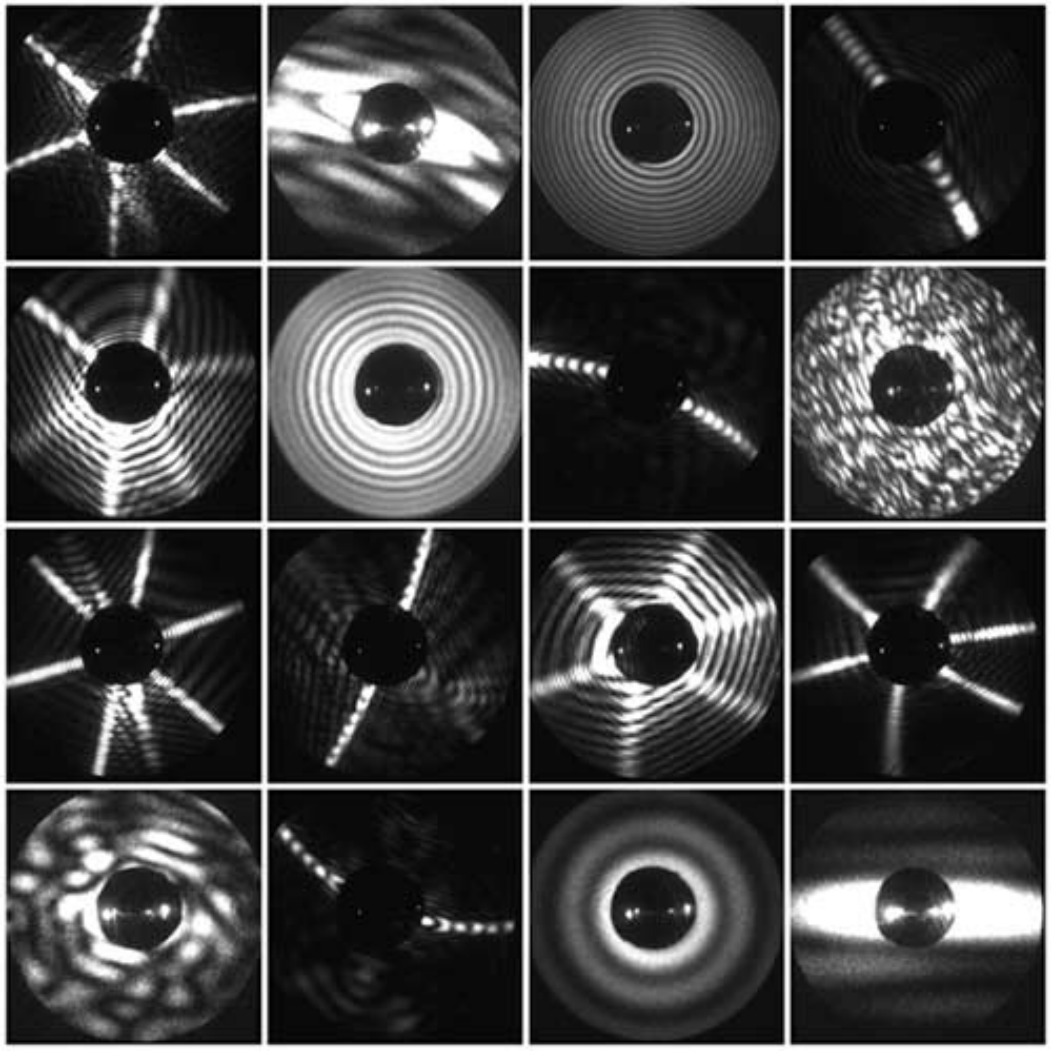
Thanks for listening

Long-term natural climate change

- Milankovitch cycles: millenial 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

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 ~100 Vm^{-1})



Particle Phase Discriminator, University of Hertfordshire

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

Cloud aerosol and precipitation spectrometer, University of Manchester

Ice clouds in CLOUD