The CLOUD experiment

Cosmics Leaving Outdoor Droplets

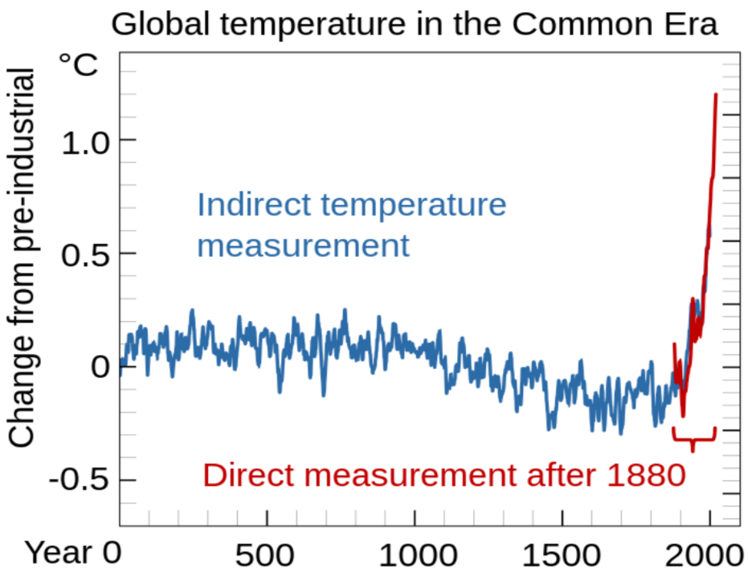
Photo: NASA ISS007E10807

Agenda

- Background: Earth’s climate, cosmic rays, aerosols and clouds
- CLOUD Experiment: Concept, methods, results

- (Visit to CLOUD, if time allows)

Earth’s temperature



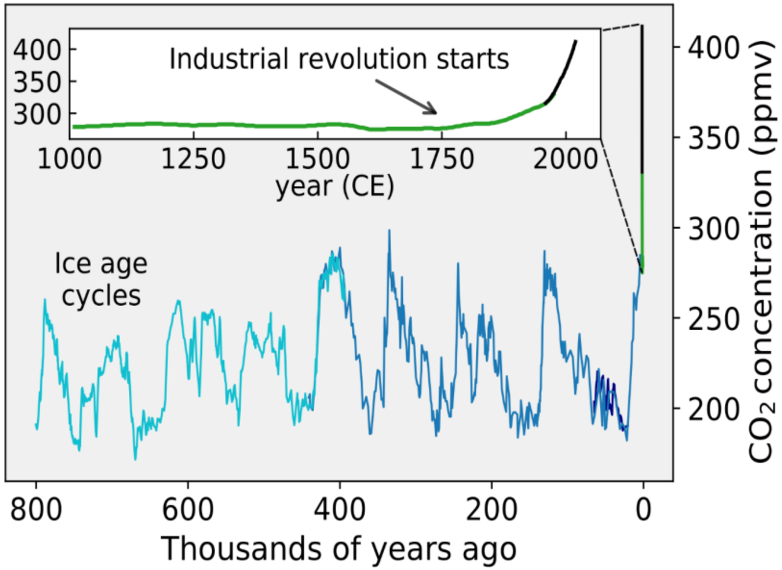
Source: Wikipedia

Global surface temperature reconstruction over the last 2000 years using proxy data from tree rings, corals, and ice cores (blue) [1] and directly observed data (red) [2].

[1] Neukom, R., et al., (2019b), Consistent multidecadal variability in global temperature reconstructions and simulations over the Common Era. *Nature Geoscience*. **12** (8): 643–649. doi:10.1038/s41561-019-0400-0

[2] Global Annual Mean Surface Air Temperature Change“, https://data.giss.nasa.gov/gistemp/graphs_v4/, NASA. Retrieved 23 Feb 2020.

Recent major increase in CO_2



Source: Wikipedia

CO_2 concentrations over the last 800,000 years as measured from ice cores [3][4][5][6] (blue/green) and directly [7] (black)

[3] Lüthi, D., et al., (May 2005). High-resolution carbon dioxide concentration record 650,000–800,000 years before present, *Nature*, **453** (7193): 379-382 doi:10.1038/nature06949

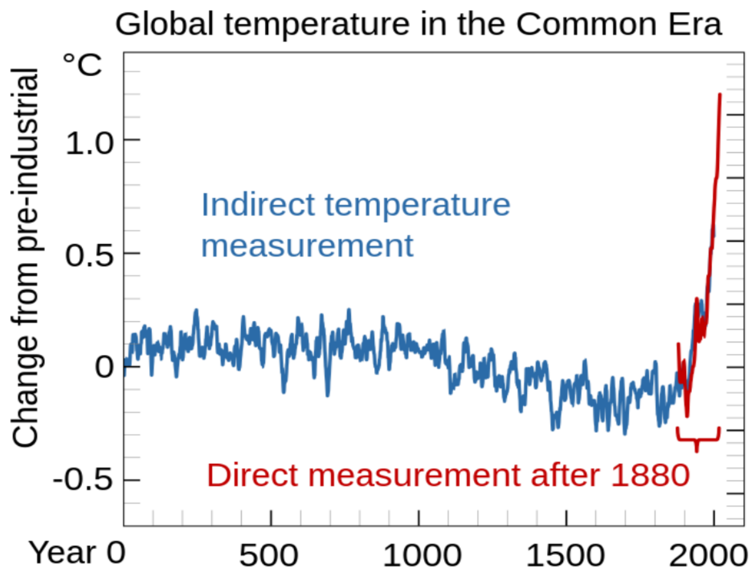
[4] Fischer, H., et al, (12 March 1999), Ice Core Records of Atmospheric CO 2 Around the Last Three Glacial Terminations, *Science*, **283** (5408): 1712–1714. doi:10.1126/science.283.5408.1712

[5] Indermühle, A., et al. (1 March 2000), Atmospheric CO 2 concentration from 60 to 20 kyr BP from the Taylor Dome Ice Core, Antarctica, *Geophysical Research Letters*. **27** (5): 735–738. doi:10.1029/1999GL010960

[6] Etheridge, D., et al, (1998). Historical CO2 Records from the Law Dome DE08, DE08-2, and DSS Ice Cores. *Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory*. U.S. Department of Energy. Retrieved 20 Nov 2022.

[7] Keeling, C.; Whorf, T. (2004). Atmospheric CO2 Records from Sites in the SIO Air Sampling Network". *Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory*. U.S. Department of Energy. Retrieved 20 Nov 2022.

Increase of CO_2

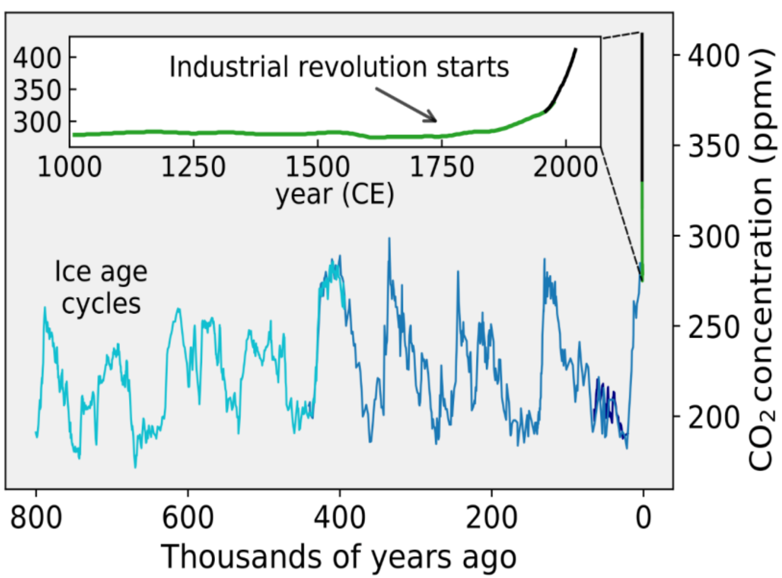


Source: Wikipedia

Global surface temperature reconstruction over the last 2000 years using proxy data from tree rings, corals, and ice cores (blue) [1] and directly observed data (red) [2].

[1] Neukom, R., et al., (2019b), Consistent multidecadal variability in global temperature reconstructions and simulations over the Common Era. *Nature Geoscience*. **12** (8): 643–649. doi:10.1038/s41561-019-0400-0

[2] Global Annual Mean Surface Air Temperature Change", https://data.giss.nasa.gov/gistemp/graphs_v4/, NASA. Retrieved 23 Feb 2020.



Source: Wikipedia

CO_2 concentrations over the last 800,000 years as measured from ice cores [3][4][5][6] (blue/green) and directly [7] (black)

[3] Lüthi, D., et al., (May 2005). High-resolution carbon dioxide concentration record 650,000–800,000 years before present, *Nature*, **453** (7193): 379-382 doi:10.1038/nature06949

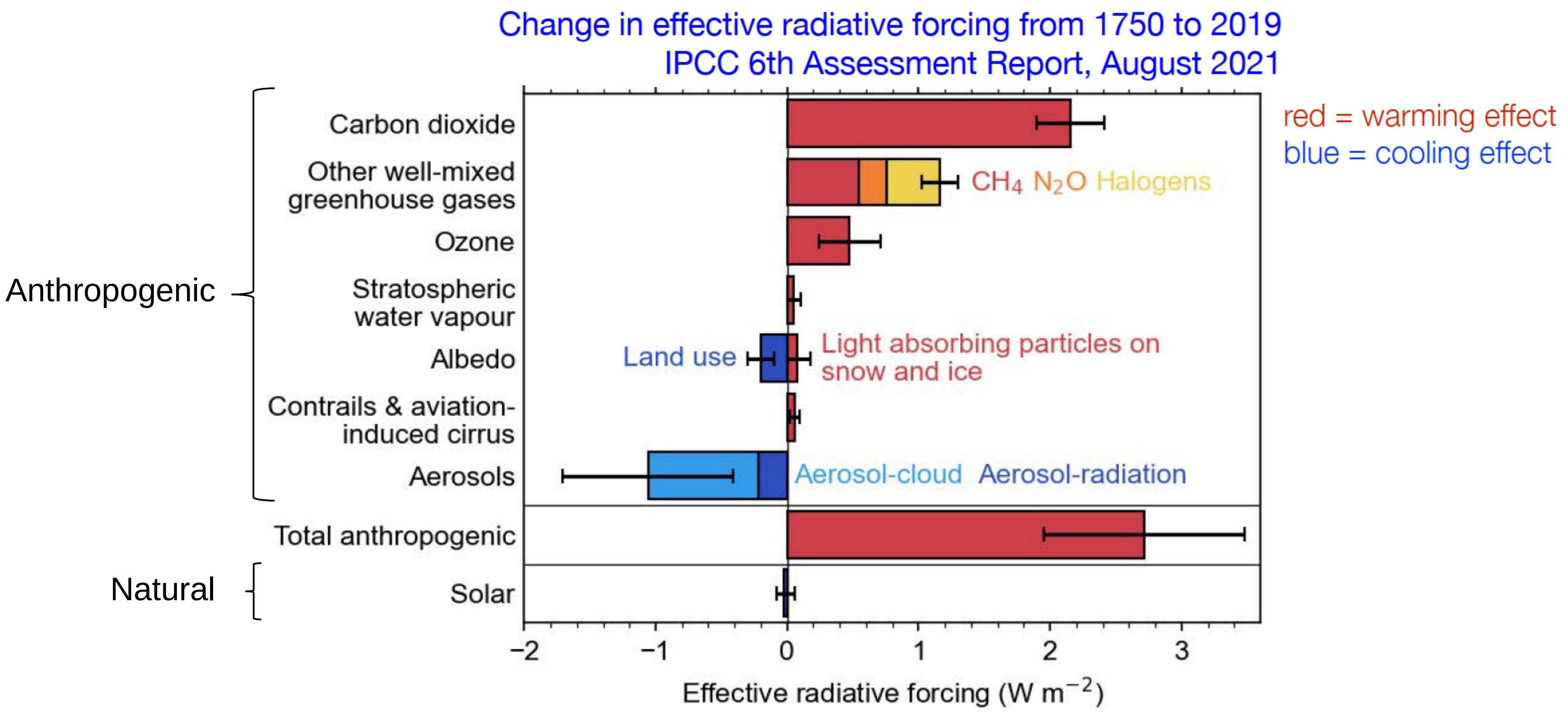
[4] Fischer, H., et al, (12 March 1999), Ice Core Records of Atmospheric CO 2 Around the Last Three Glacial Terminations, *Science*, **283** (5408): 1712–1714. doi:10.1126/science.283.5408.1712

[5] Indermühle, A., et al. (1 March 2000), Atmospheric CO 2 concentration from 60 to 20 kyr BP from the Taylor Dome Ice Core, Antarctica, *Geophysical Research Letters*. **27** (5): 735–738. doi:10.1029/1999GL010960

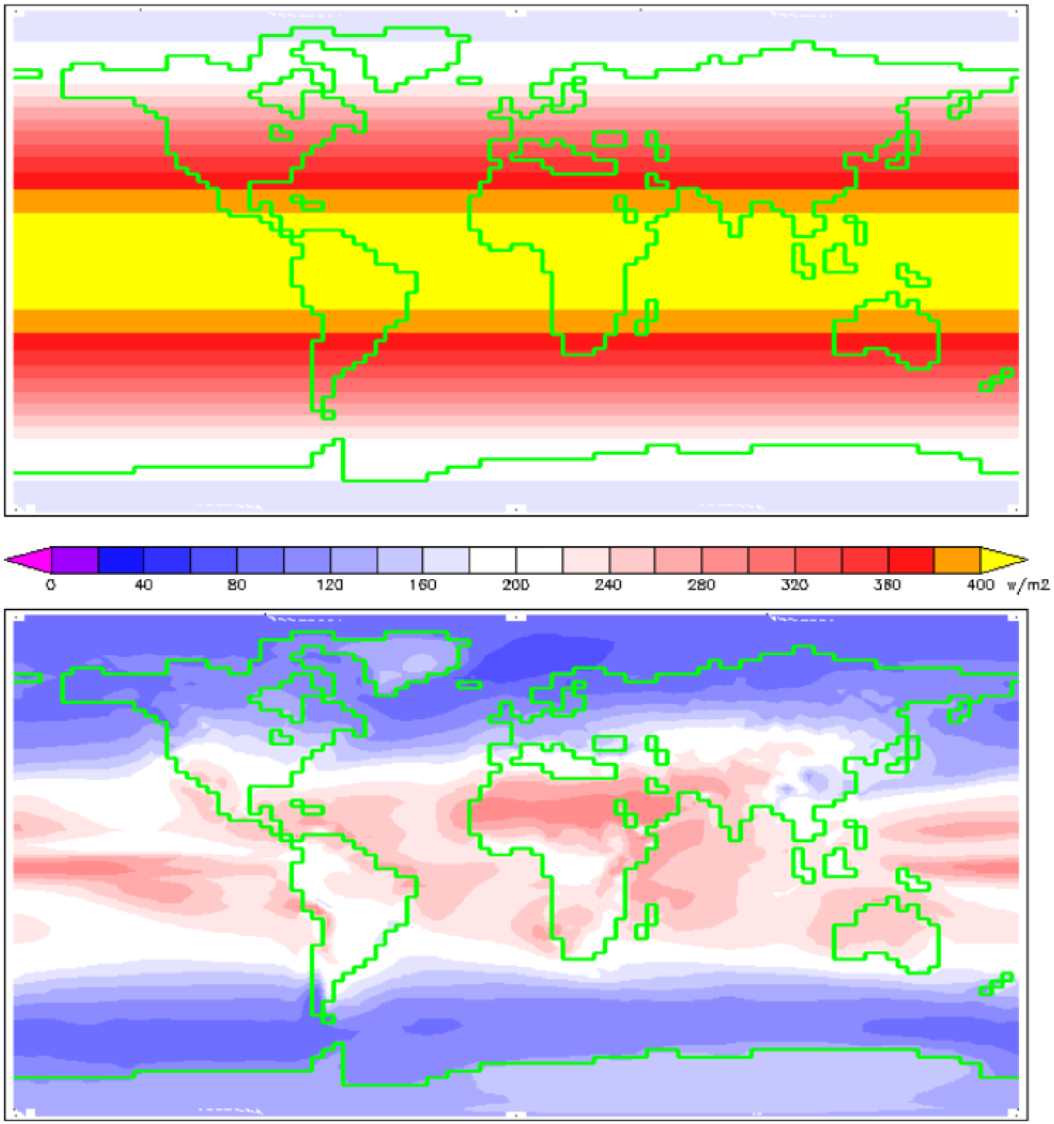
[6] Etheridge, D., et al, (1998). Historical CO2 Records from the Law Dome DE08, DE08-2, and DSS Ice Cores. *Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory*. U.S. Department of Energy. Retrieved 20 Nov 2022.

[7] Keeling, C.; Whorf, T. (2004). Atmospheric CO2 Records from Sites in the SIO Air Sampling Network". *Carbon Dioxide Information Analysis Center, Oak Ridge National Laboratory*. U.S. Department of Energy. Retrieved 20 Nov 2022.

But the climate change is not only about CO_2 !



Solar irradiation in total

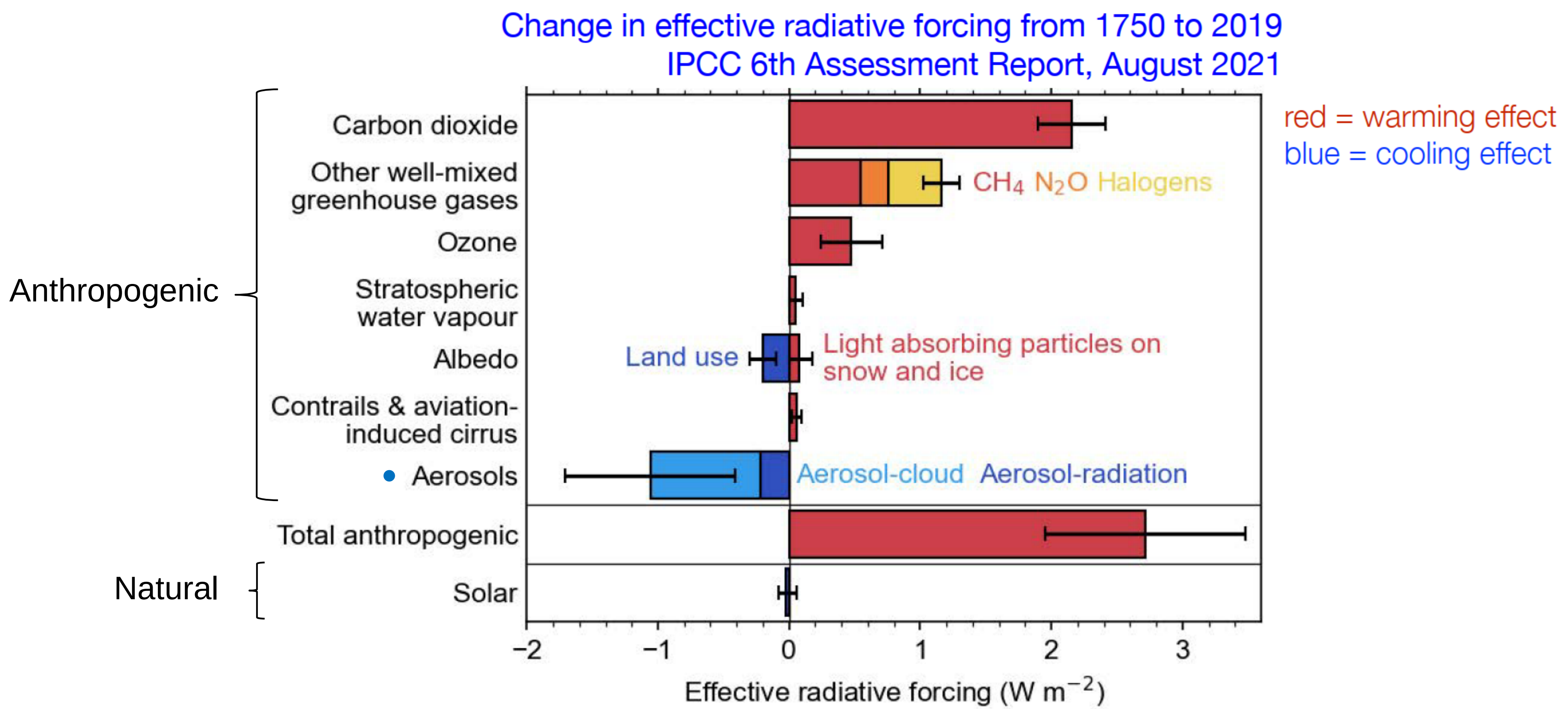


At the top of Earth's atmosphere

At ground level
(uses the same colour scale)

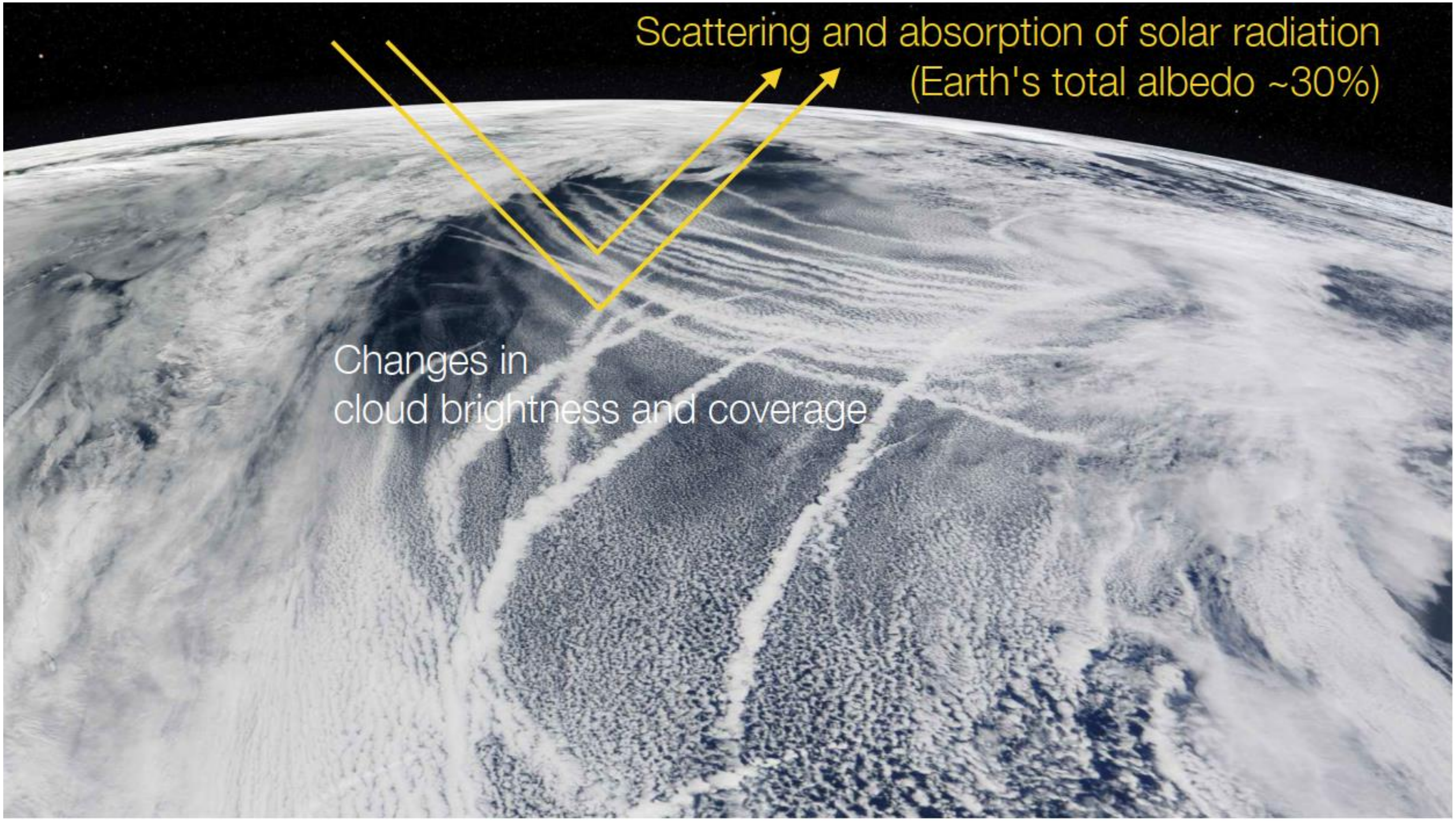
Source Wikipedia:
William M. Connolley using HadCM3 data

Effect of aerosols



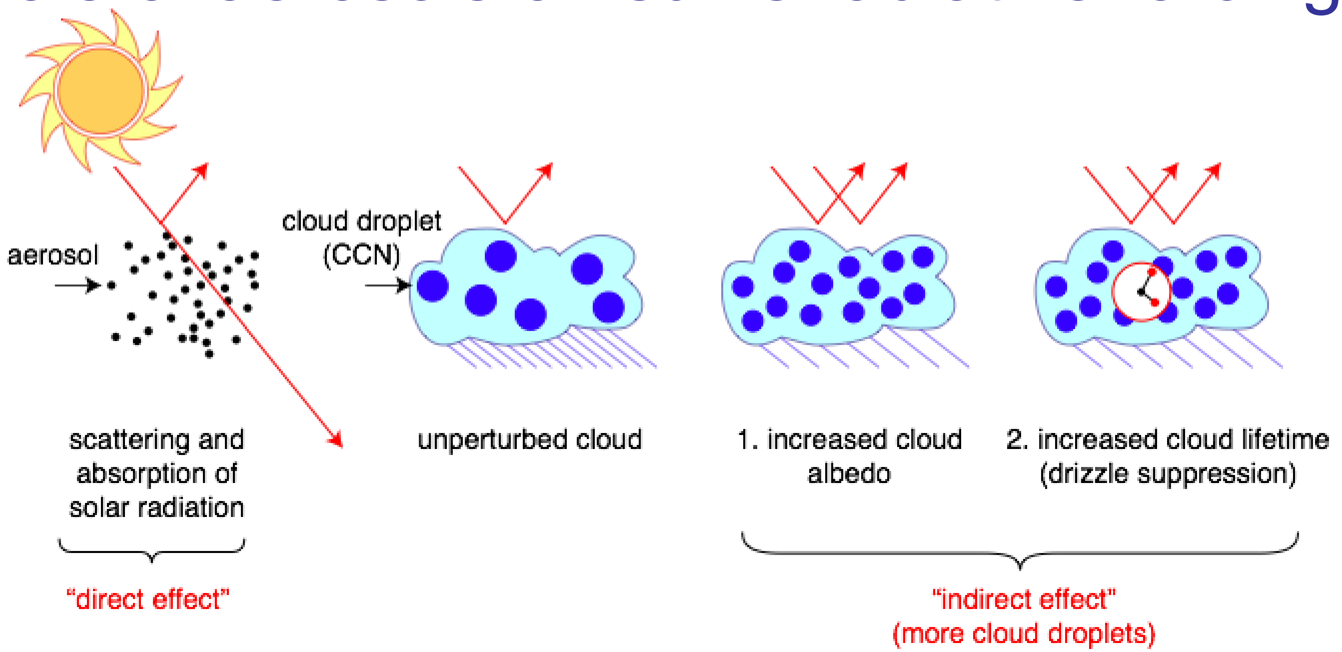
- Aerosols have a cooling effect.
  - They have counter-acted a large but poorly understood fraction of warming from greenhouse gases
  - The uncertainty in total anthropogenic radiative forcing is dominated by aerosols
  - Future emission reductions (e.g. SO_2) will reduce the cooling from aerosols/clouds. But by how much?

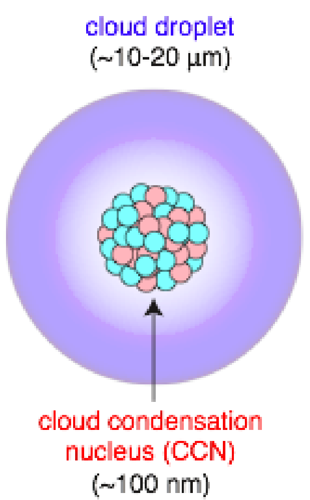
Aerosol and clouds



North Pacific, NASA MODIS satellite, 4 March 2009

Role of aerosols on sun's radiative forcing





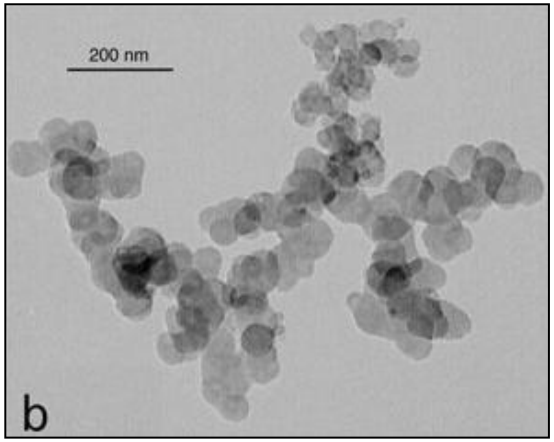
- All cloud droplets form on aerosol "seeds" known as cloud condensation nuclei - CCN
- Cloud properties are sensitive to number of droplets
- More aerosols/CCN:
  - Brighter clouds, with longer lifetimes
- Sources of atmospheric aerosols:
  - Primary (dust, sea salt, fires)
  - Secondary (gas-to-particle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

See youtube: "No particles no fog" https://www.youtube.com/watch?v=EneDwu0HrVg

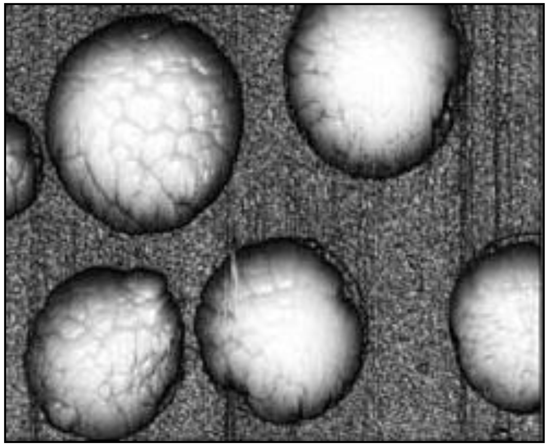
What is an aerosol?

Definition: Suspension of small (liquid or solid) particles in a gas

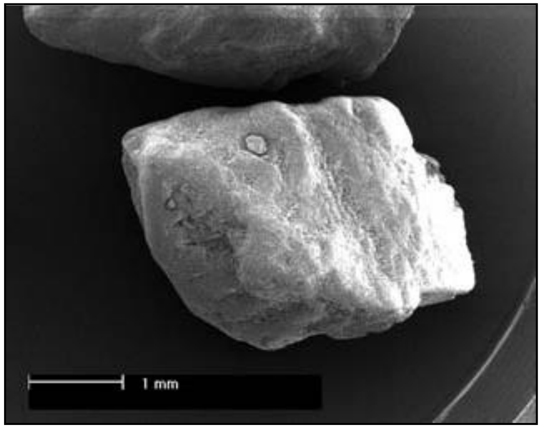
Diesel soot: ca. 0.1 μm



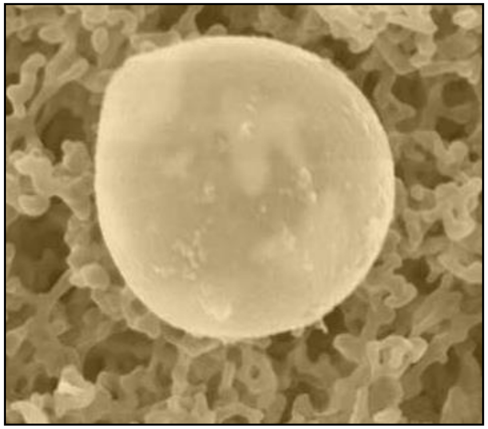
Ammonium sulfate: ca. 0.1 μm



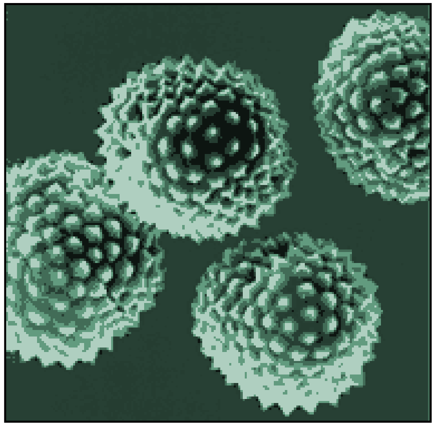
Sea salt: 0.2 - 10 μm



Mineral dust: 0.2 - 10 μm

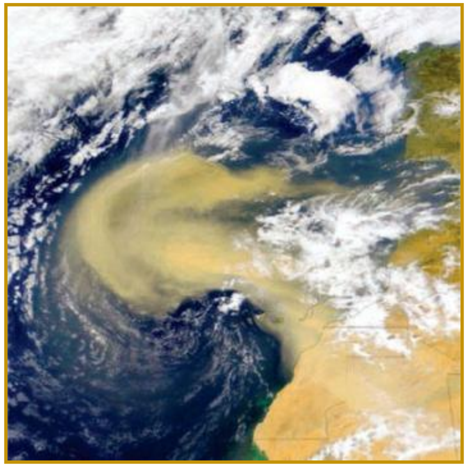


Pollen: 10 - 100 μm



Primary Aerosol Sources

Sea spray



Mineral dust

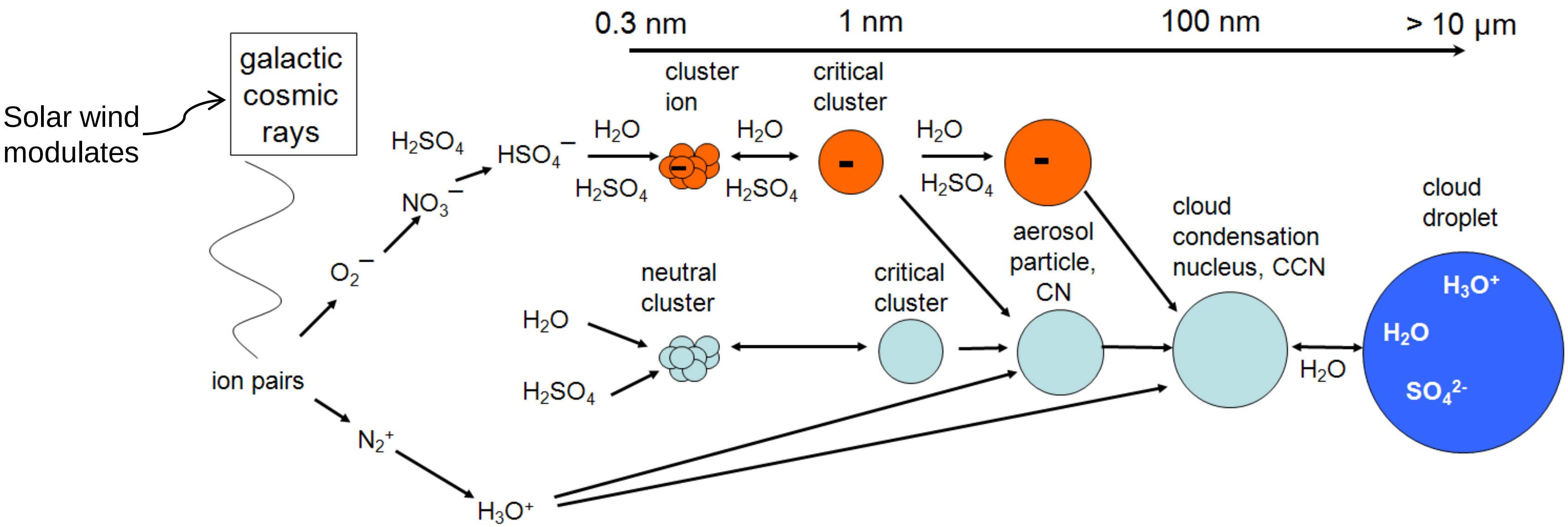
Volcano ► Sulfates, dust

Biomass burning ► Organics

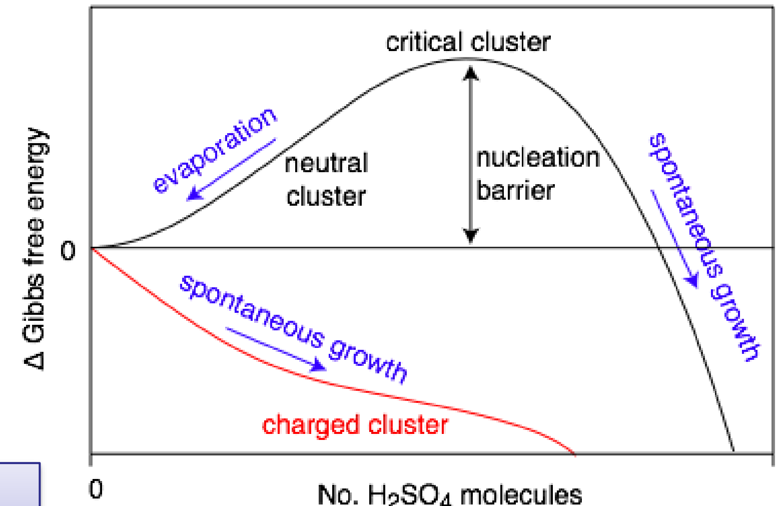
Traffic emissions ► Soot

Industrial Emissions

Secondary aerosol production: Gas-to-particle conversion



- Trace condensable vapour → CN → CCN
- But contributing vapours and nucleation rates poorly known
- H_2SO_4 is thought to be the primary condensable vapour in atmosphere (sub ppt)
- Ion-induced nucleation pathway is energetically favoured but limited by the ion production rate and ion lifetime
- *Candidate mechanism for solar-climate variability*



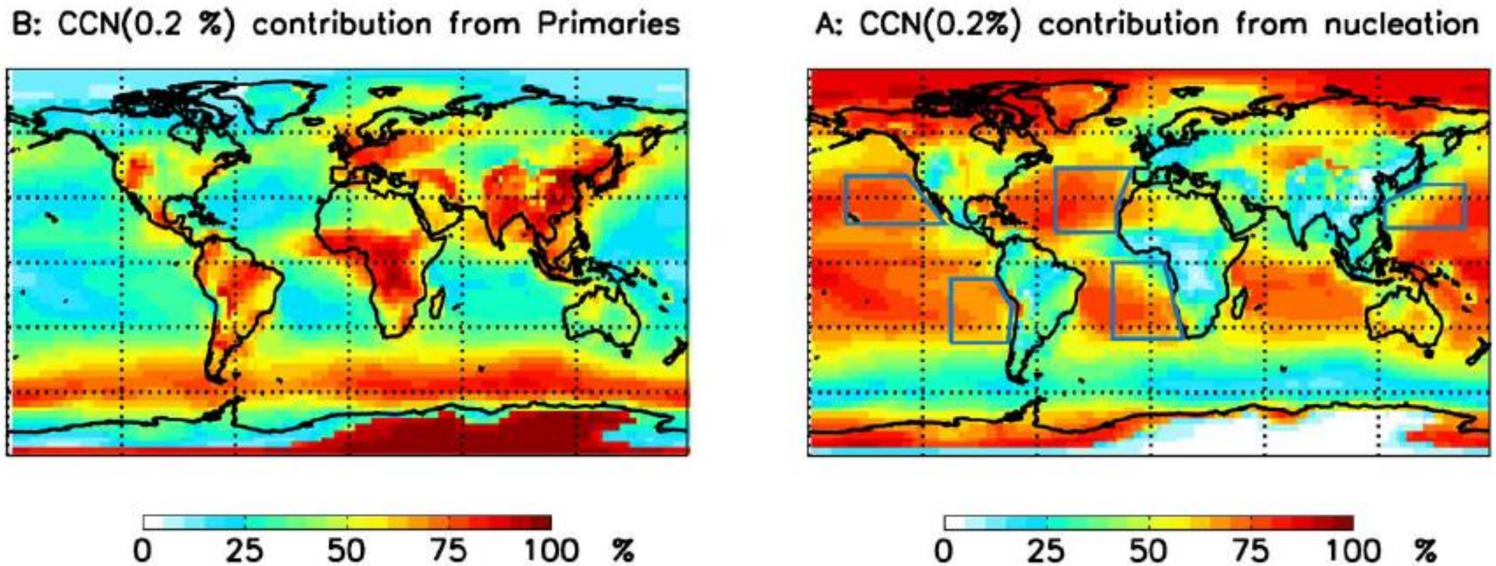
This secondary aerosol formation is the key object of study in CLOUD

Primary vs. secondary aerosols

Origin of global cloud condensation nuclei, CCN, 500-1000 m above ground level

Primary production (dust, sea-spray, biomass burning)

Secondary production - nucleation (gas-to-particle conversion)



Merikanto et al., ACP, 2009

About 50% of all cloud drops are formed on secondary aerosols

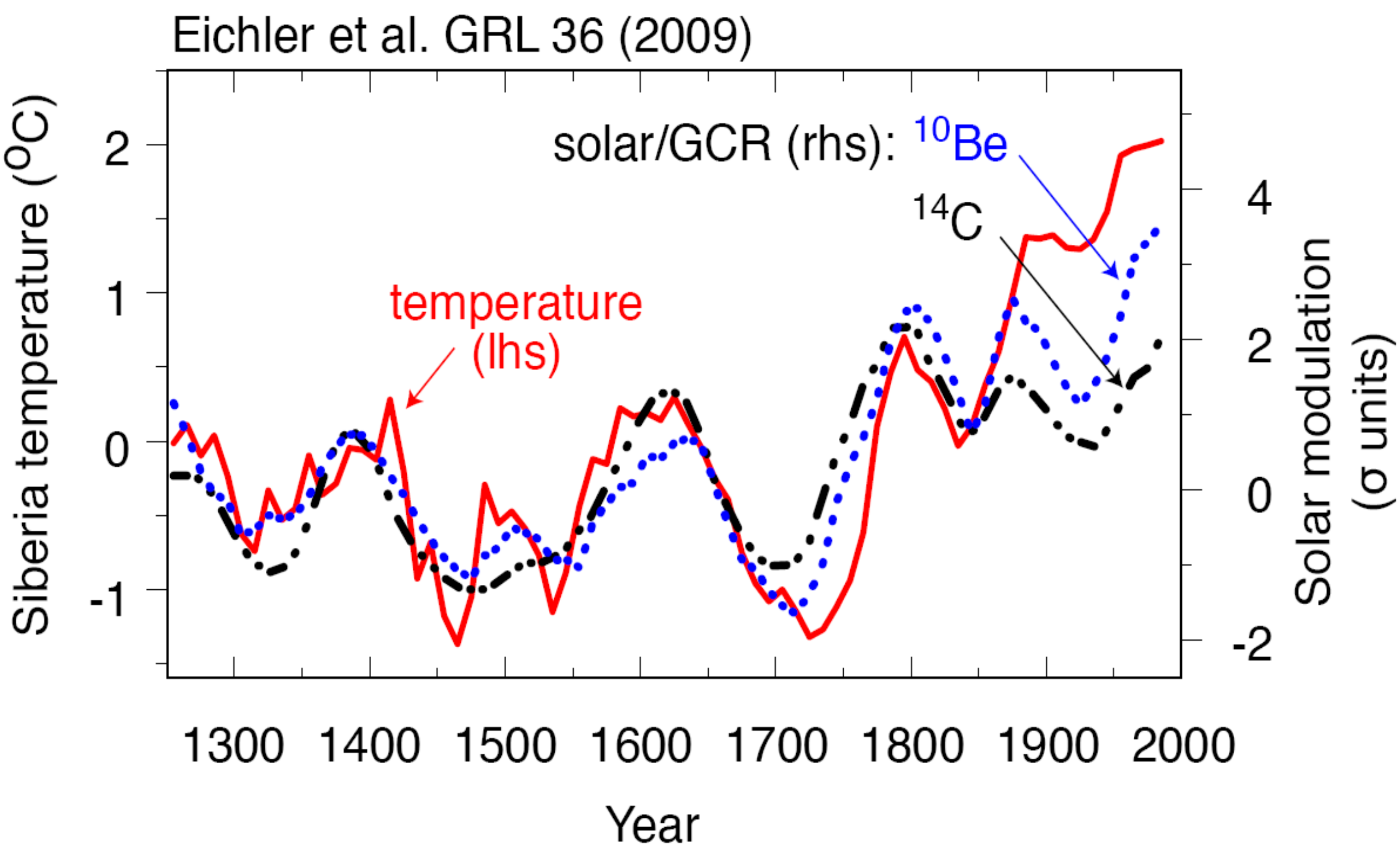
Secondary aerosol formation – nucleation is poorly understood and is the key object of study in CLOUD

Original reason why CLOUD is at CERN:
Study link from Cosmic Rays via aerosols & clouds to Climate

- Numerous correlations suggest GCR-climate connection.
- But no established mechanism to explain this.

Several recent observations, e.g. by Eichler et al., ACP, 2009:

Correlation between GCRs and temperature in Siberia from glacial ice core data.



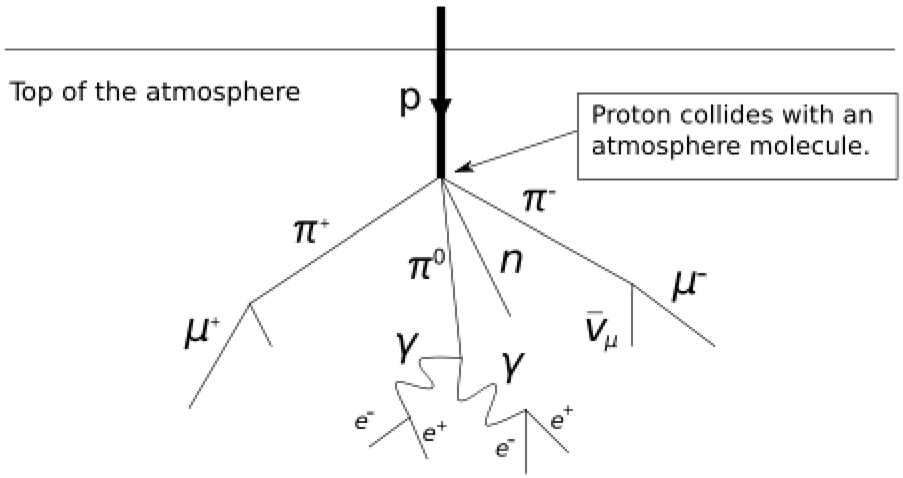
Cosmic rays

High energy particles from outer space

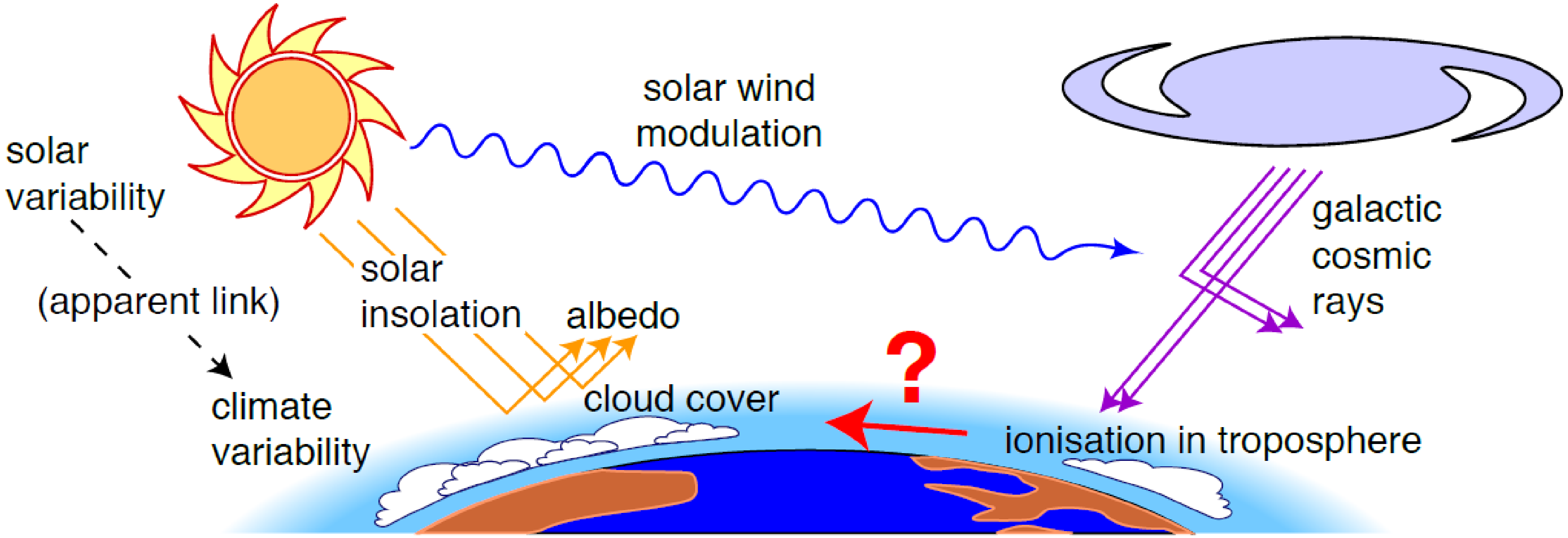
- Mostly protons; ~90%
- Helium nuclei (alpha particles); ~9%
- Others: Electrons, heavy nuclei; 1%

Earth atmosphere protects from the cosmic rays

- Lacking protection against cosmic rays is a major problem for long space travels.



Solar → Cosmic ray → Climate mechanism?



- Higher solar activity → reduced GCRs →(?) reduced cloud cover → warmer climate
- Satellite observations not yet settled:
  Significant GCR-cloud correlations reported by some (Svensmark, Laken...) and weak or excluded by others (Kristjansson, Wolfendale...)

CLOUD

- CLOUD studies the above topics by
  - Recreating atmosphere in a large chamber to study aerosol particle formation and influence of ions
  - Studying aerosol-cloud interactions by forming liquid or ice clouds

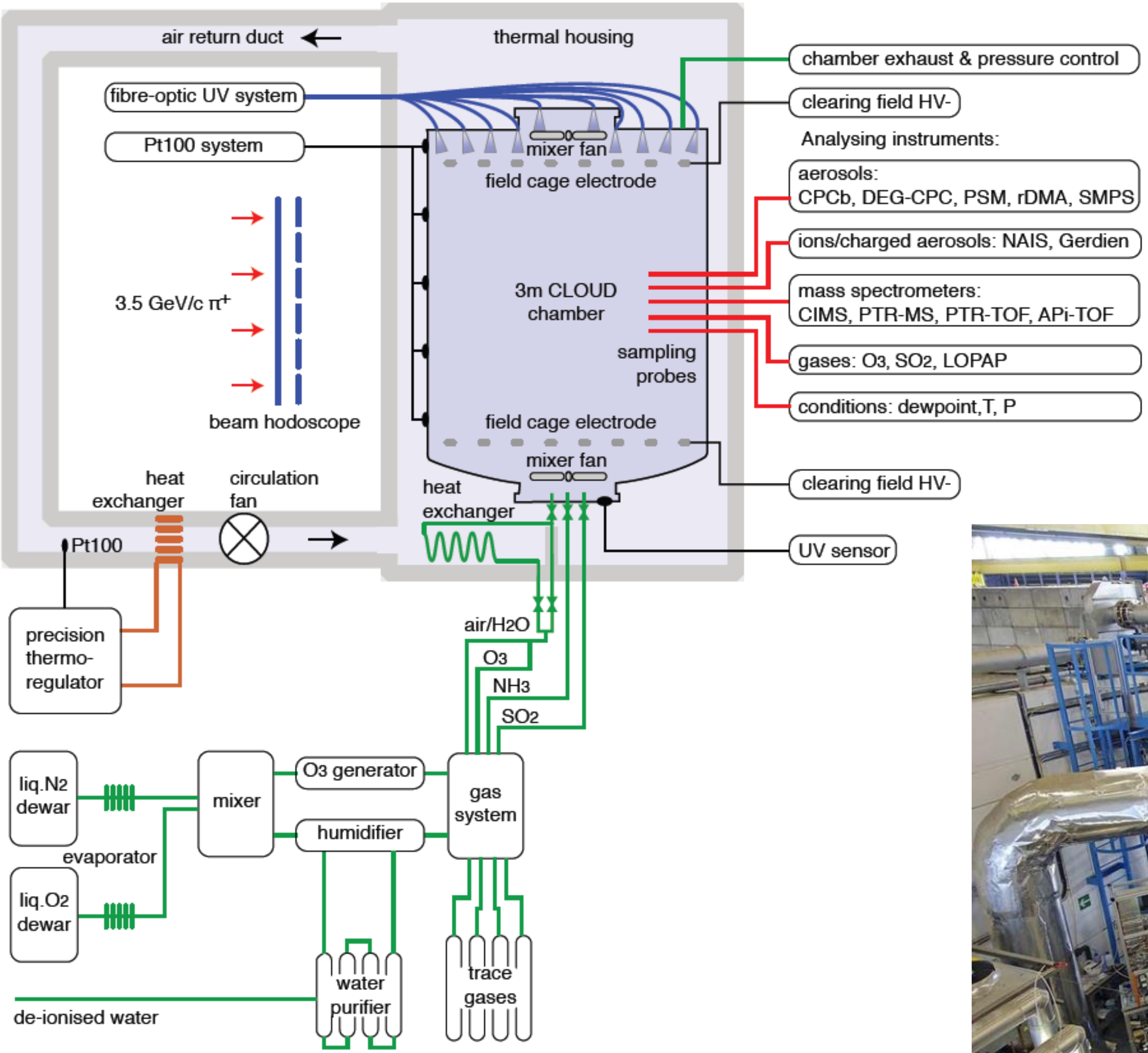
- CLOUD's unique features:
  - As realistic as possible atmospheric conditions:
    - Requires large chamber with contaminants < 1 pptv (10^{-12} or 30 sec / 1 million years)
    - World's cleanest large atmospheric chamber
  - Measurement and control of ionizing radiation (cosmic rays and CERN's beam) and UV light
  - Operates over full tropospheric T range (+30°C → -65°C)
  - State-of-art instruments analyse contents of chamber

CLOUD Aerosol chamber

- 27 m^3
- Pressure: Atmospheric ± 0.3 bar
- Only metallic seals
- Electropolished inner surfaces

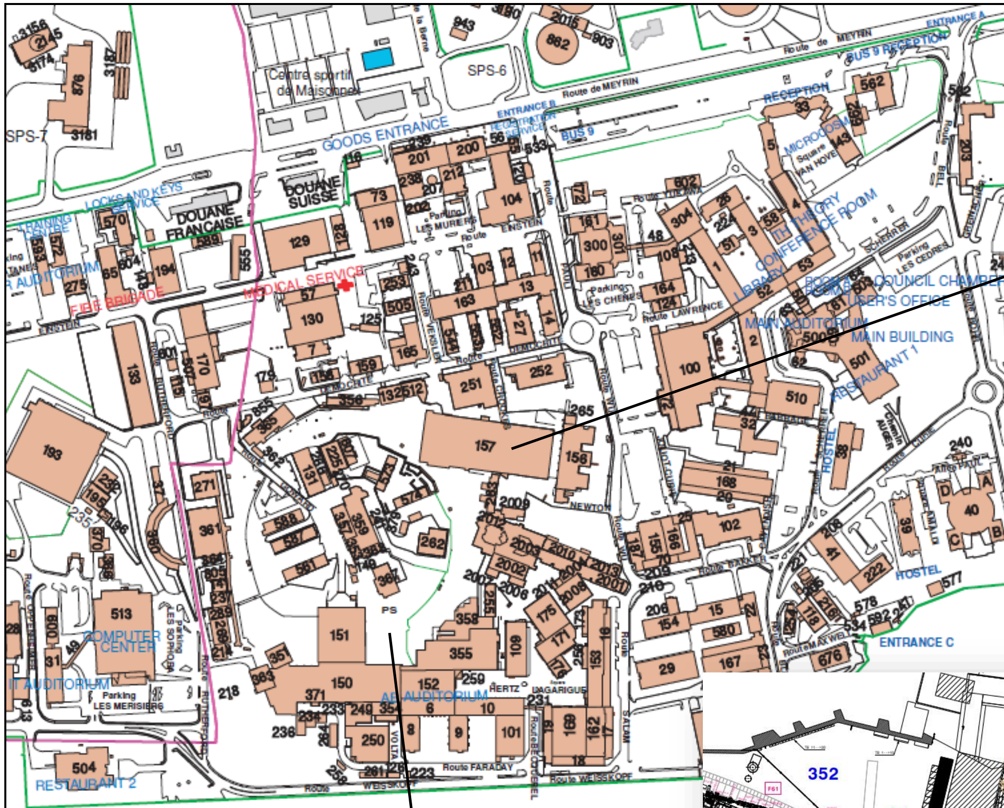
Aerosol chamber in T11

CLOUD



air return duct
thermal housing
fibre-optic UV system
Pt100 system
mixer fan
field cage electrode
3.5 GeV/c π+
beam hodoscope
3m CLOUD chamber
sampling probes
field cage electrode
mixer fan
heat exchanger
circulation fan
heat exchanger
Pt100
precision thermo-regulator
air/H2O
O3
NH3
SO2
liq.N2 dewar
evaporator
liq.O2 dewar
mixer
O3 generator
humidifier
gas system
water purifier
trace gases
de-ionised water
chamber exhaust & pressure control
clearing field HV-
Analysing instruments:
aerosols: CPCb, DEG-CPC, PSM, rDMA, SMPS
ions/charged aerosols: NAIS, Gerdien
mass spectrometers: CIMS, PTR-MS, PTR-TOF, APi-TOF
gases: O3, SO2, LOPAP
conditions: dewpoint,T, P
clearing field HV-
UV sensor

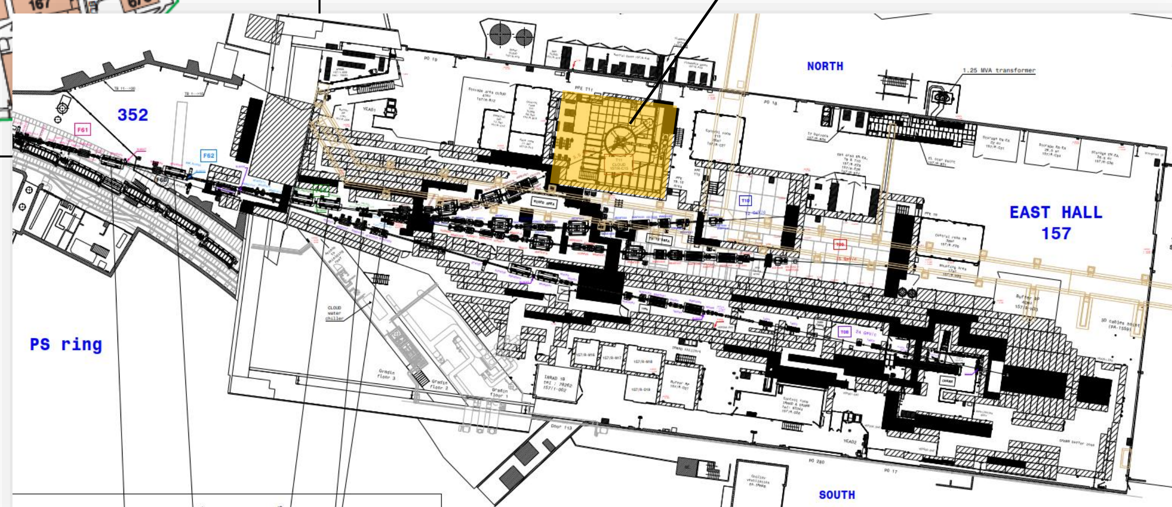
CLOUD in CERN PS East Hall



PS East Hall

Proton Synchrotron (PS) accelerator, first operation in 1959

CLOUD beam area in PS East Hall



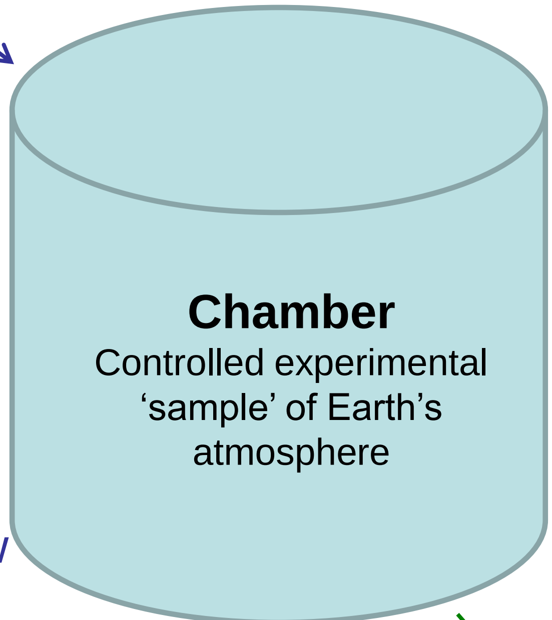
CLOUD in CERN PS East Hall

CLOUD measurement principle

1. Fill chamber with clean air + water vapour

2. Set temperature and pressure

3. Add trace gases, condensable species in atmospheric, extremely low concentrations
~1 molecule in 10^{12} air molecules



4. Expose to ionizing beam, and possibly to UV-light

5. Observe
- Particle growth size distribution
- Electrical charge distribution
- Cloud droplet/ice particle concentrations,
- etc.

6. Repeat experiment (typically some hours), possibly with varying parameters

7. Carefully flush the chamber and clean the chamber walls between experiments

Now: CLOUD16 run (25 Sep - 3 Dec 2023)

- Tropical rainforest upper free troposphere:
  - α-pinene, isoprene
  - sulfuric acid
  - NOx
- Marine surfactants in the upper free troposphere:
  - nonanal ((CH2)9O)
  - sulfuric acid
  - NOx
- Cool boreal forest boundary layer:
  - α-pinene
  - sulfuric acid
- Arctic boundary layer:
  - dimethylsulfide (methanesulfonic acid, sulfuric acid)
  - iodine (iodic acid, iodous acid)
  - ammonia
  - glyoxal (dialdehyde, CHOCHO)
- Interaction of biogenic and anthropogenic vapours in urban environments:
  - biogenic vapours (trees): α-pinene, isoprene
  - anthropogenic vapours (automobiles, industry...): sulfuric acid, ammonia, dimethylamine, aromatic organics, NOx

Results: Improved understanding of atmospheric processes

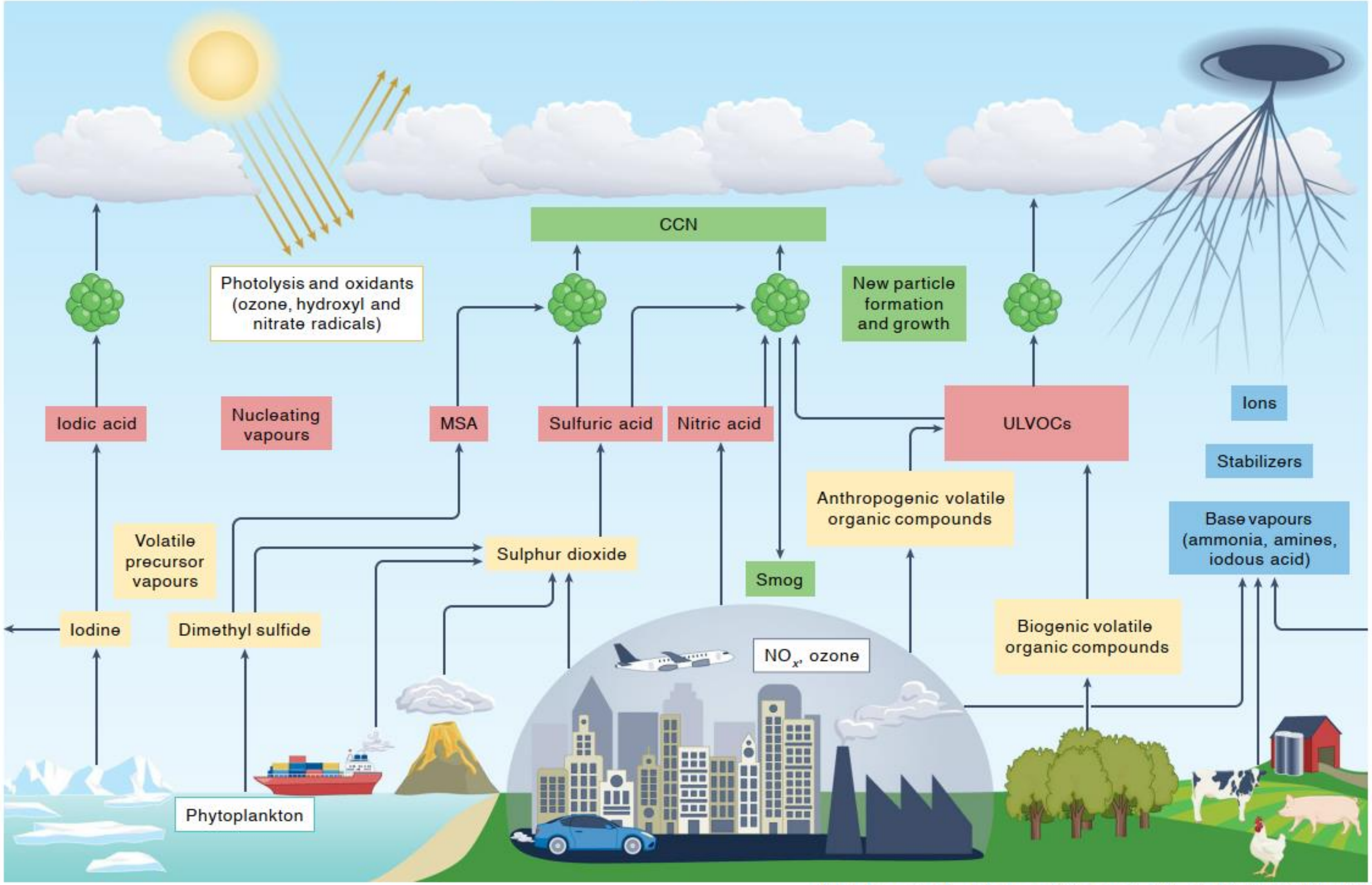
nature geoscience

Published online: 7 November 2023

Perspective

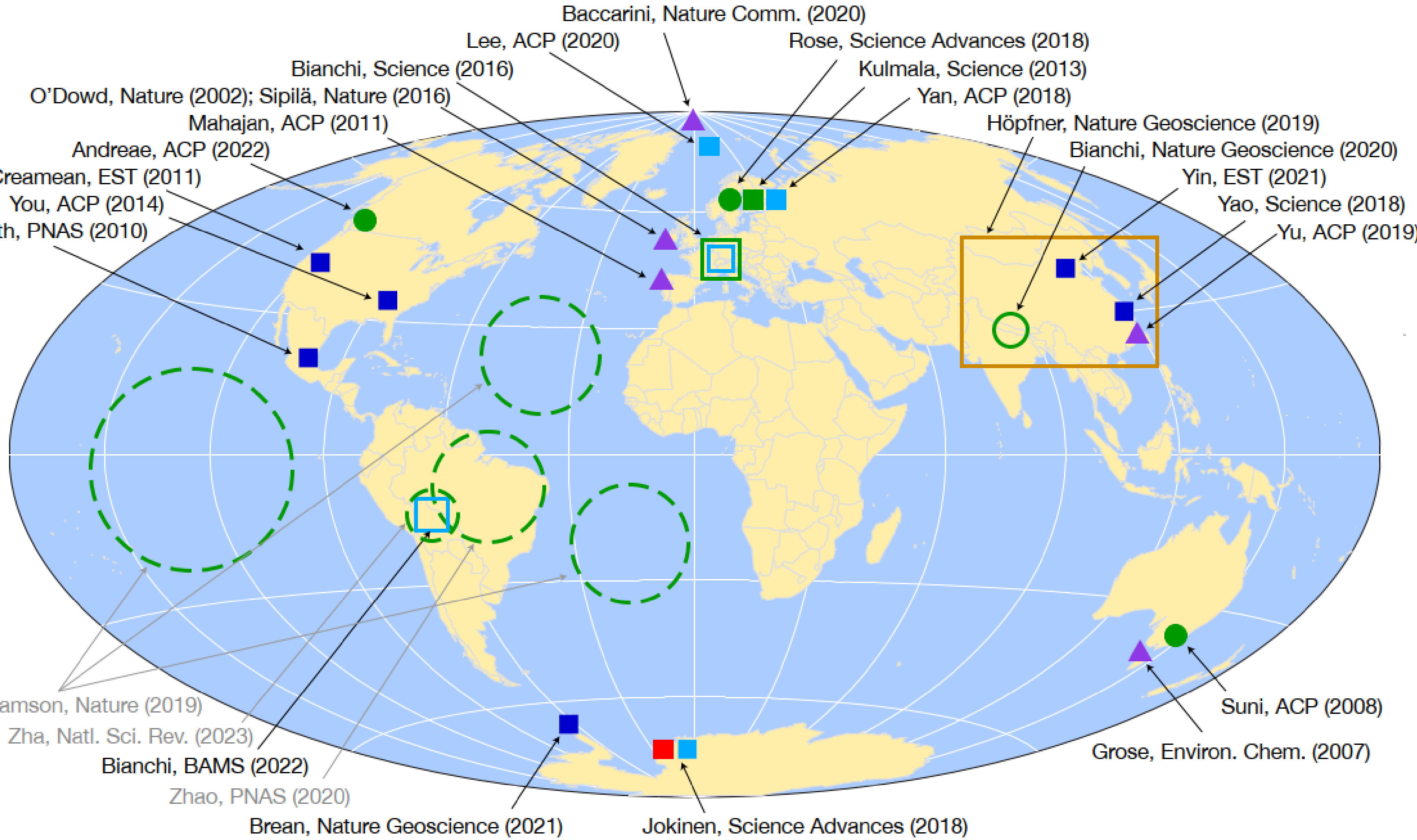
https://doi.org/10.1038/s41561-023-01305-0

Atmospheric new particle formation from the CERN CLOUD experiment



Kirkby et al., Nature Geosci. 16, 948-957 (2023)

Geographical locations of nucleation mechanisms measured by CLOUD



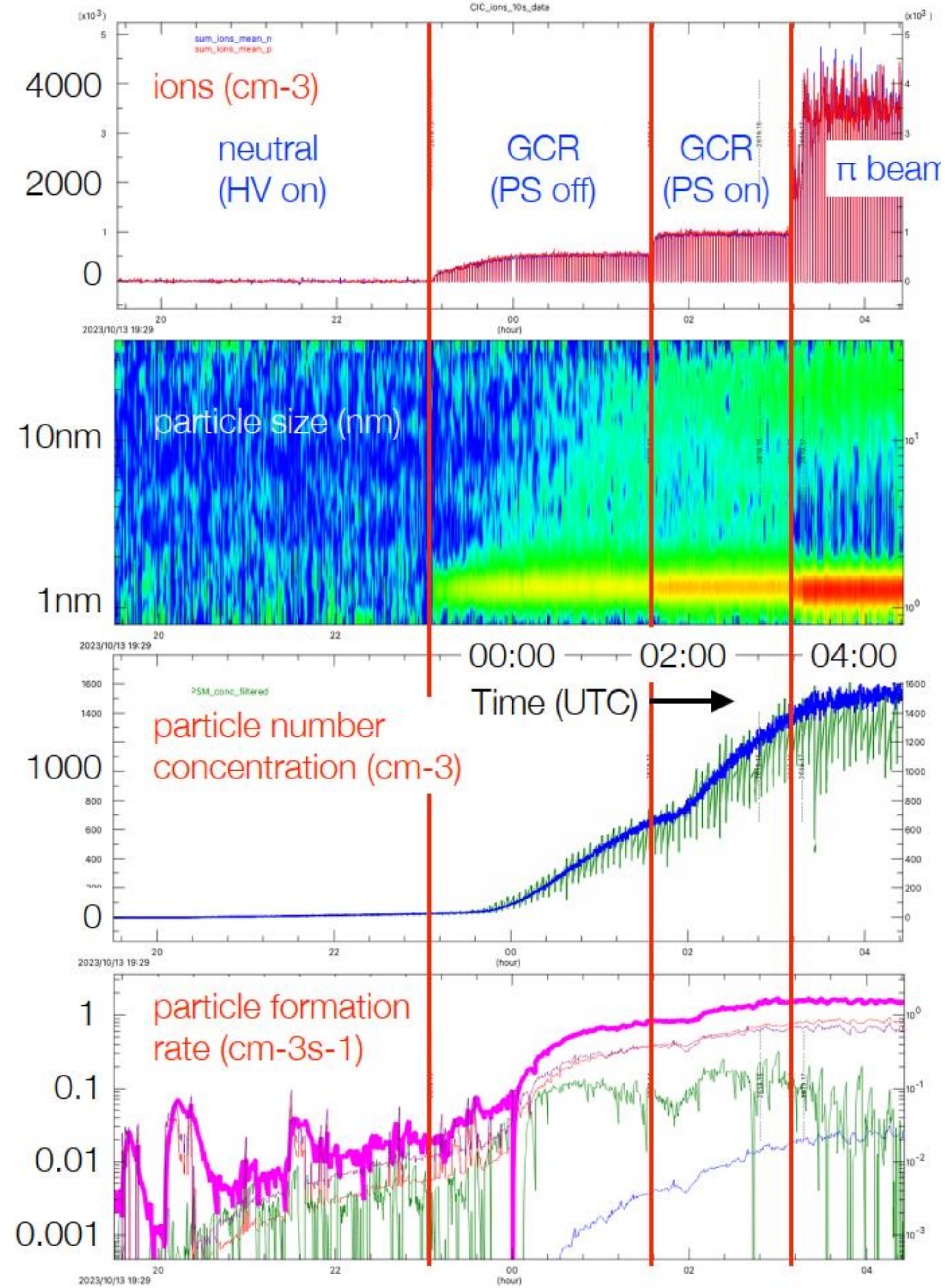
Legend:
filled symbols = boundary layer
large open symbols = free troposphere
dashed open symbols = not yet confirmed

- sulfuric acid
- sulfuric acid-ammonia
- sulfuric acid-ammonia-biogenic HOM
- sulfuric acid-ammonia-nitric acid
- sulfuric acid-amines
- biogenic HOM
- iodic acid

Kirkby et al., Nature Geosci. 16, 948-957 (2023)

Example from CLOUD16

- Aerosol particle formation in the tropical rainforest upper free troposphere (CLOUD chamber at -50°C)



CLOUD's near-term future plans

- Aerosol particle formation and growth in cold regions:
  - Tropical Atlantic and Pacific upper free troposphere
  - Asian monsoon upper free troposphere
  - Southern Ocean upper free troposphere
  - Particle evaporation in passing from cold to warm environments

- "CLOUDy" experiments:
  - Effect of aerosol charge on cloud microphysics (aerosol scavenging)
  - Asian monsoon ice nucleation from HNO3-H2SO4-NH3 particles
  - Transport of vapours to the upper free troposphere: release of NH3 (and other dissolved vapours) upon supercooled droplet freezing or evaporation

- Parameterise CLOUD measurements for global climate models, and evaluate the impact on present and future climates

CLOUD collaboration & Finland

20 institutions from
9 countries with
150 contributing scientists and technical staff.

Finnish institutes and scientists are centrally involved in CLOUD since the beginning:

- State-of-the-art measurement instruments, operated at CLOUD.
- Contributions to scientific studies and publications, in lead role in many of them.
- Essential links to field measurements done in Finland and elsewhere.

Country	Institution
	European Organization for Nuclear Research (CERN), Geneva
Austria	University of Innsbruck, Institute for Ion and Applied Physics, Innsbruck
	University of Vienna, Faculty of Physics
Cyprus	The Cyprus Institute, Climate & Atmosphere Research Center, Nicosia
Estonia	University of Tartu, Laboratory of Environmental Physics
Finland	Finnish Meteorological Institute, Helsinki
	Helsinki Institute of Physics
	University of Eastern Finland, Department of Applied Physics, Kuopio
	University of Helsinki, Department of Physics
Germany	Goethe University Frankfurt, Institute for Atm. and Env. Sciences
	Karlsruhe Institute of Technology, Institute for Meteorology and Climate Research
	Leibniz Institute for Tropospheric Research, Leipzig
	Max-Planck Institute for Chemistry, Department of Atmospheric Chemistry, Mainz
Portugal	University of Lisbon, Department of Physics and University of Beira Interior, Covilha
Sweden	University of Stockholm, Department of Applied Environmental Science
Switzerland	Paul Scherrer Institute, Laboratory of Atmospheric Chemistry, Villigen
United States	California Institute of Technology, Div. of Chemistry and Chemical Eng., Pasadena, CA
	Carnegie Mellon University, Department of Chemical Eng., Pittsburgh, PA
	Aerodyne Research Inc., Billerica, MA with Tofwerk AG, Thun, Switzerland
	University of Colorado, Dep. of Chemistry and Biochemistry & CIRES, Boulder, CO

CLOUD during a physics run

Spare slides

CLOUD in short

CLOUD experiment's scientific goals:

1. Settling questions on cosmic ray – aerosol – cloud – climate links
2. Sharpening understanding on past & present aerosol – cloud radiative forcing
3. Understanding drivers for urban smog formation

2006: Approved in CERN's scientific program.

2007-2009: Design and construction.

2009-2019: Operation and data taking. Between beam runs continuous adaptations to improve and broaden the experimental measurement reach of CLOUD.

2020-2022: Major upgrade as part of CERN PS East Hall renovation.

2022- : Operation and data taking.

Peer-reviewed publications (2011-2022): ~75 papers, including 10 in Nature or Science

For more on CLOUD see: http://home.cern/science/experiments/cloud

Key questions addressed by CLOUD

For each system of precursor vapours and ambient conditions (T, relative humidity...):

- What is the aerosol particle formation rate vs vapour concentrations?
- What is the influence of ions from galactic cosmic rays between 0 and 10 km altitude?
- How fast do the particles grow from molecular (~1 nm) to CCN sizes (~50 nm)?
- Which chemical compounds are involved in a) nucleation and b) growth?
- What are the gas-phase chemical pathways transforming volatile precursor vapours into ultra-low-volatility nucleating vapours?

Summary of CLOUD

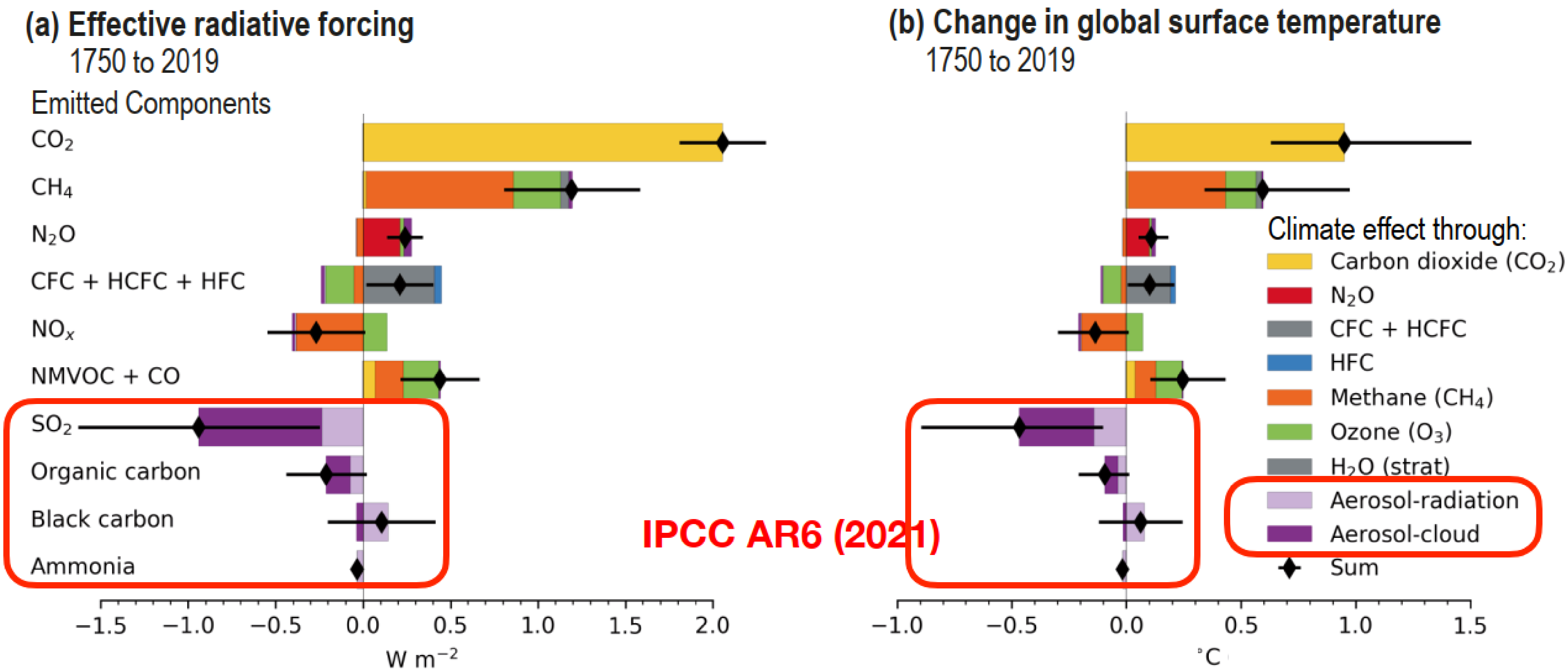
- CLOUD is providing a mechanistic understanding of aerosol particle formation and growth for global atmospheric chemistry and climate models

- This is effectively catching up with gas-phase chemical kinetics where - since more than 40 years! - laboratory experiments have provided straightforward kinetic equations that could be inserted directly into models—that is, explicit mechanisms

- In the aerosol world, a similar level of ‘nucleation kinetics’ has largely been achieved through CLOUD experiments over the past 12 years - but there is still much more to do

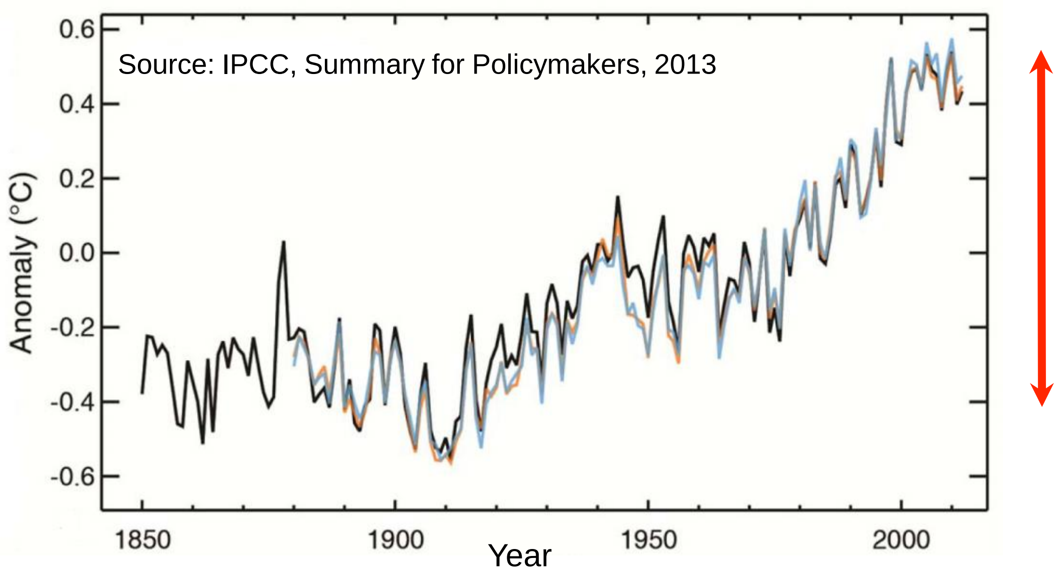
- CLOUD has transformed how aerosols are represented in global climate models

Radiative forcings since 1750 (IPCC AR6)



- Estimated aerosol effective radiative forcing = -(1.3±0.7) W/m^2
- Effective radiative forcing from CO2 = (2.1±0.3) W/m^2

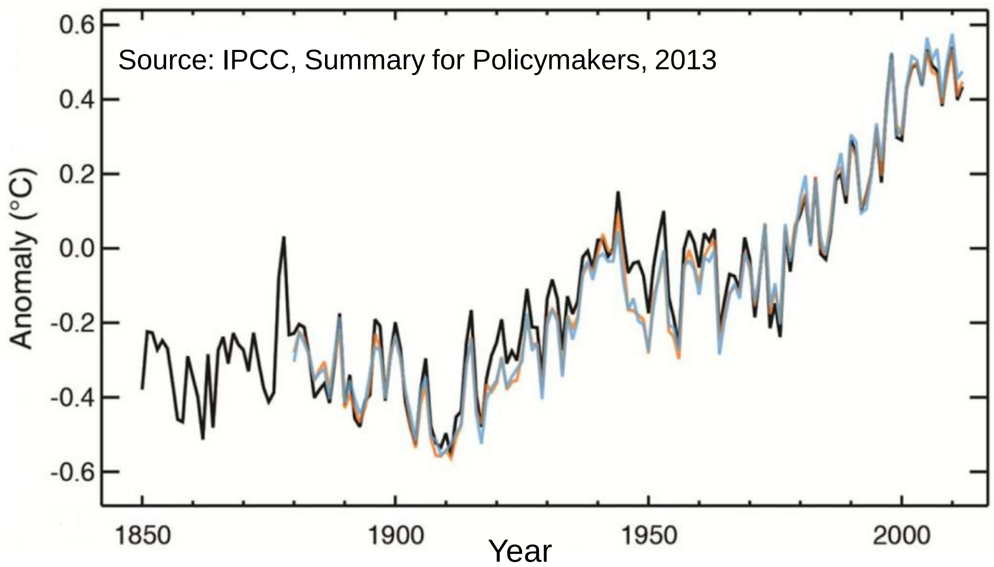
Global surface temperature



ΔT = 1 °C
since 1850

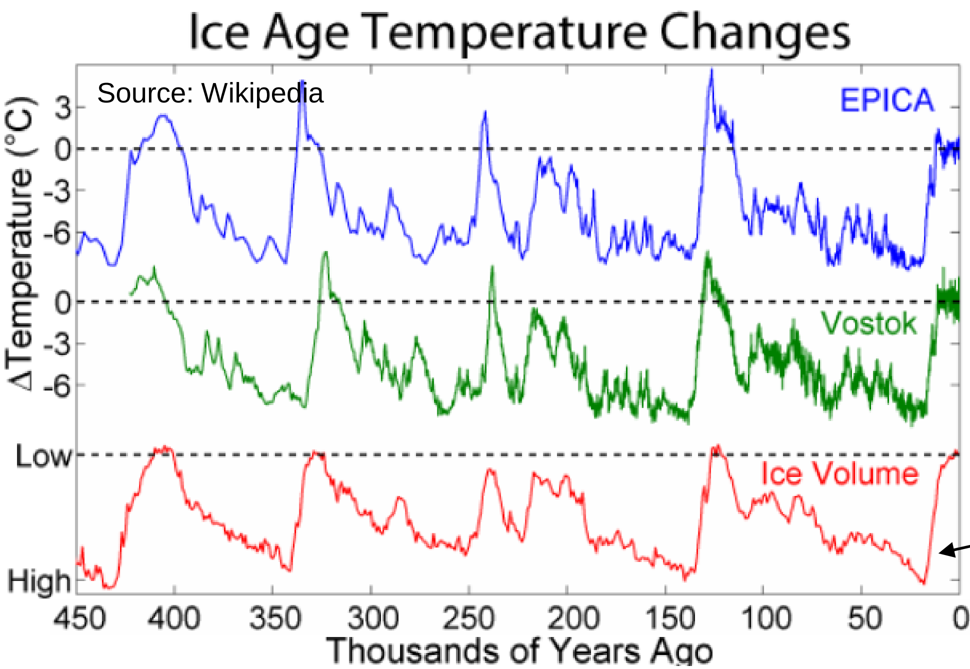
Predictions for next 100 years:
Increase of 1.5 to 4.5 °C
Goal of Paris climate agreement:
Limit increase to max 2 °C

Global surface temperature



$\Delta T = 1\ °C$
since 1850

Predictions for next 100 years and doubling of CO2 in atmosphere:
Increase of 1.5 to 4.5 °C
Goal of Paris climate agreement:
Limit increase to max 2 °C

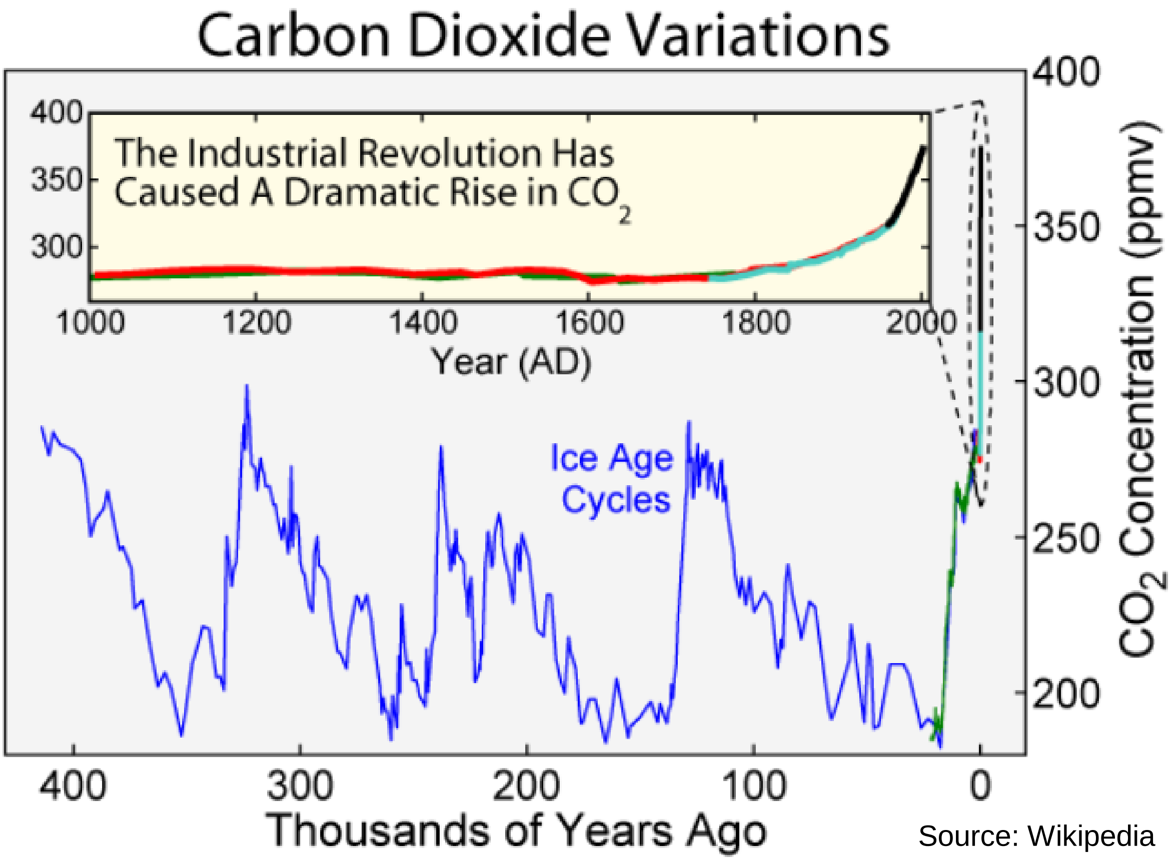


$\Delta T = 6\ °C$
since last Ice age.

EPICA = European Project for Ice Coring in Antarctica
Vostok = Ice core measurements at Russian Vostok Antartic base

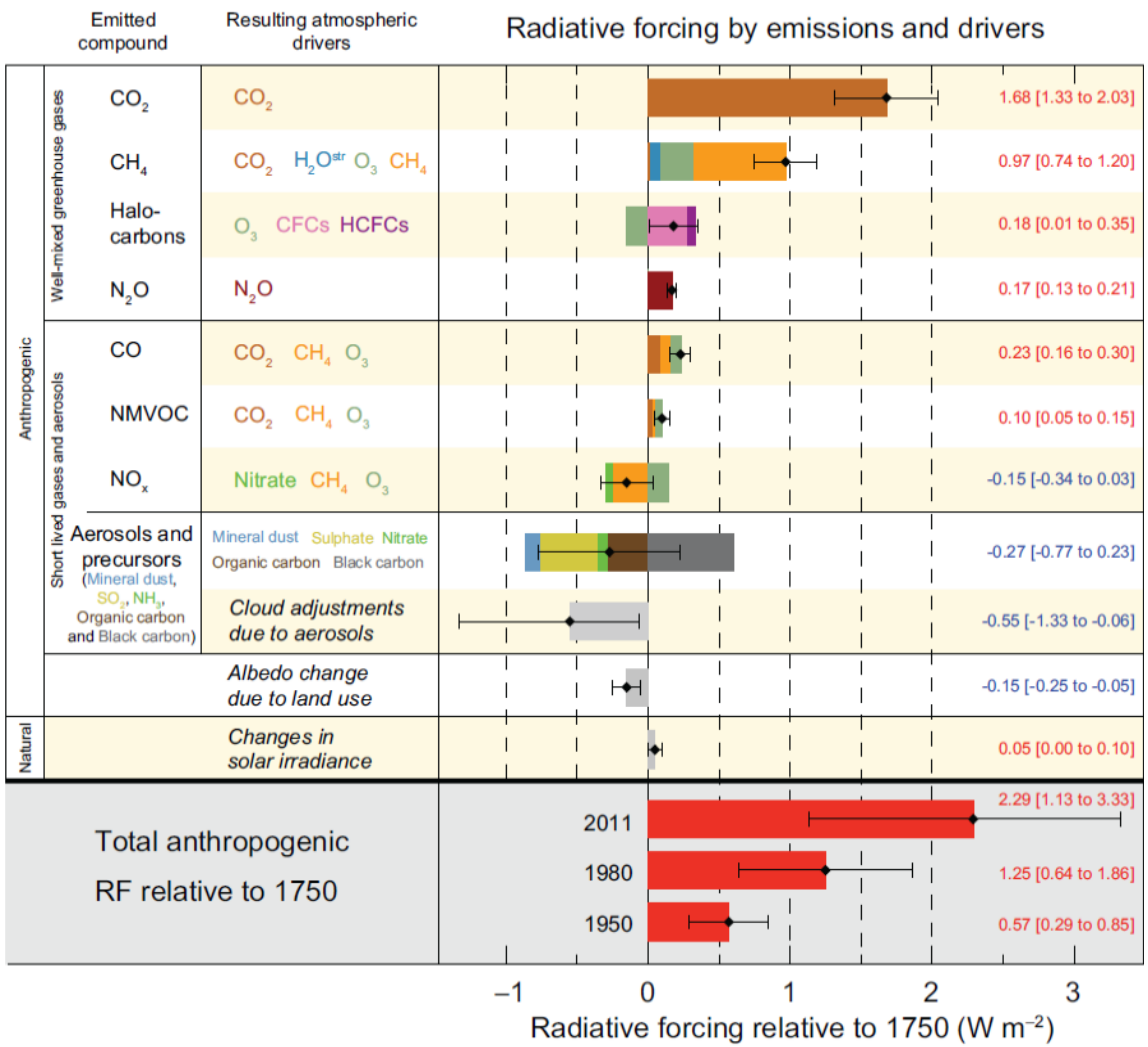
3 km thick ice on Northern Europe!

CO_2 in atmosphere



Source: Wikipedia

It's not only about CO_2 !



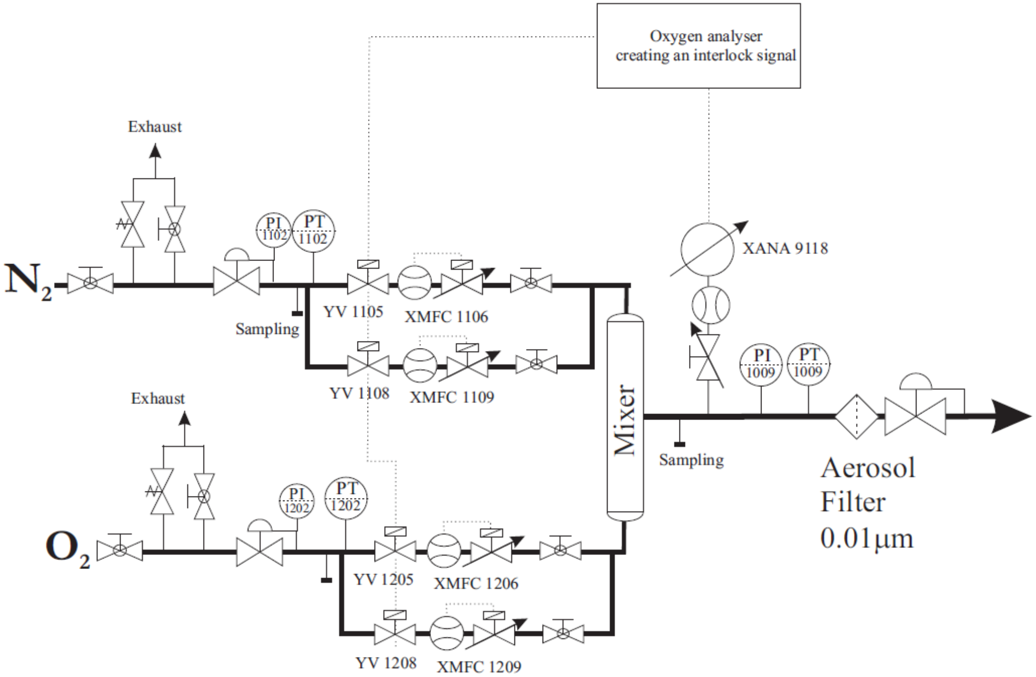
A. Anthropogenic aerosol forcings are poorly understood.

B. Natural part is very small. Is there a missing natural forcing? Is that from varying cosmic ray flux, modulated by sun?

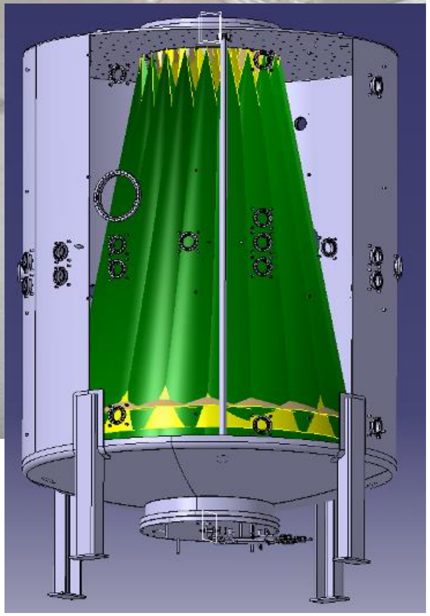
Source: IPCC, Summary for Policymakers, 2013

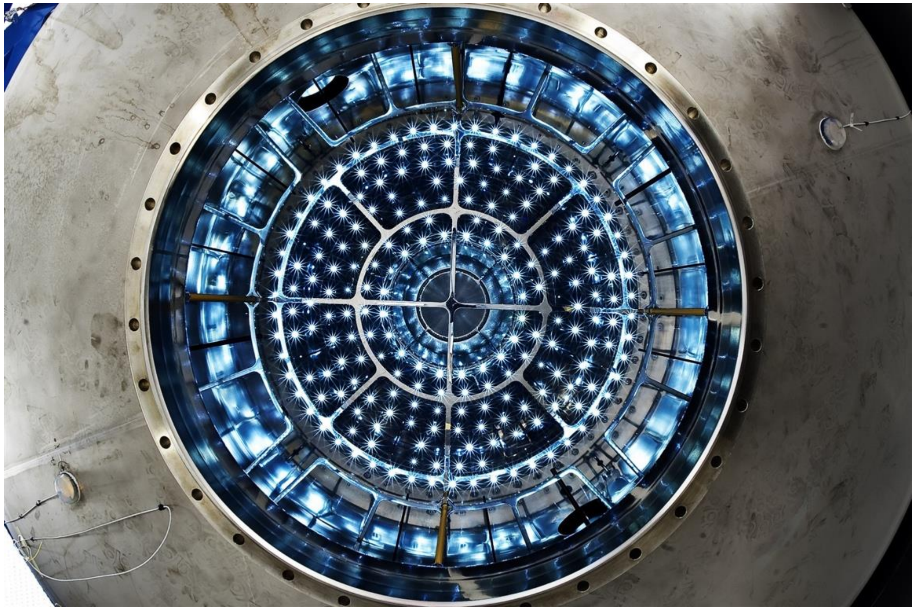
A + B → The CLOUD experiment

Ultra-pure air

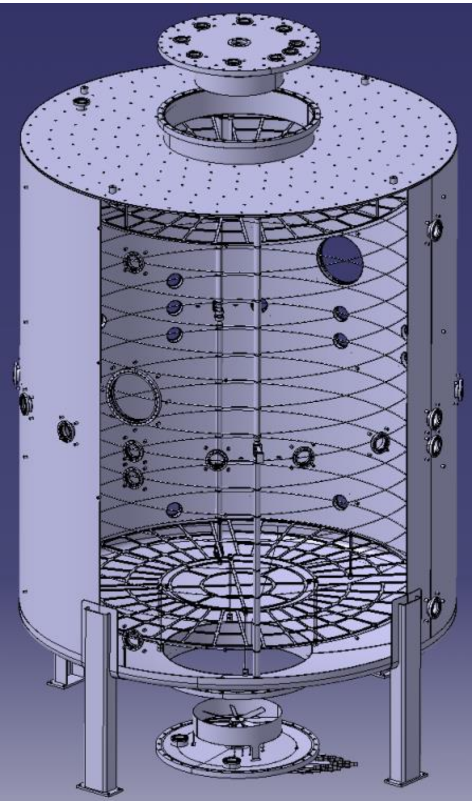


UV system





HV field cage



EU Horizon Europe Marie Curie Doctoral Network: CLOUD-DOC

- 12 PhD students
  at 12 CLOUD institutes
  (Frankfurt, CERN, Helsinki, Stockholm, Ionicon, Tropos, Cyprus Inst., Tartu, Vienna, KIT, PSI, Tofwerk)

- 1Sep22-31Aug26 (2.7M€)