Status and plans of the CLOUD experiment

CLOUD's contribution to the understanding of global aerosol and climate

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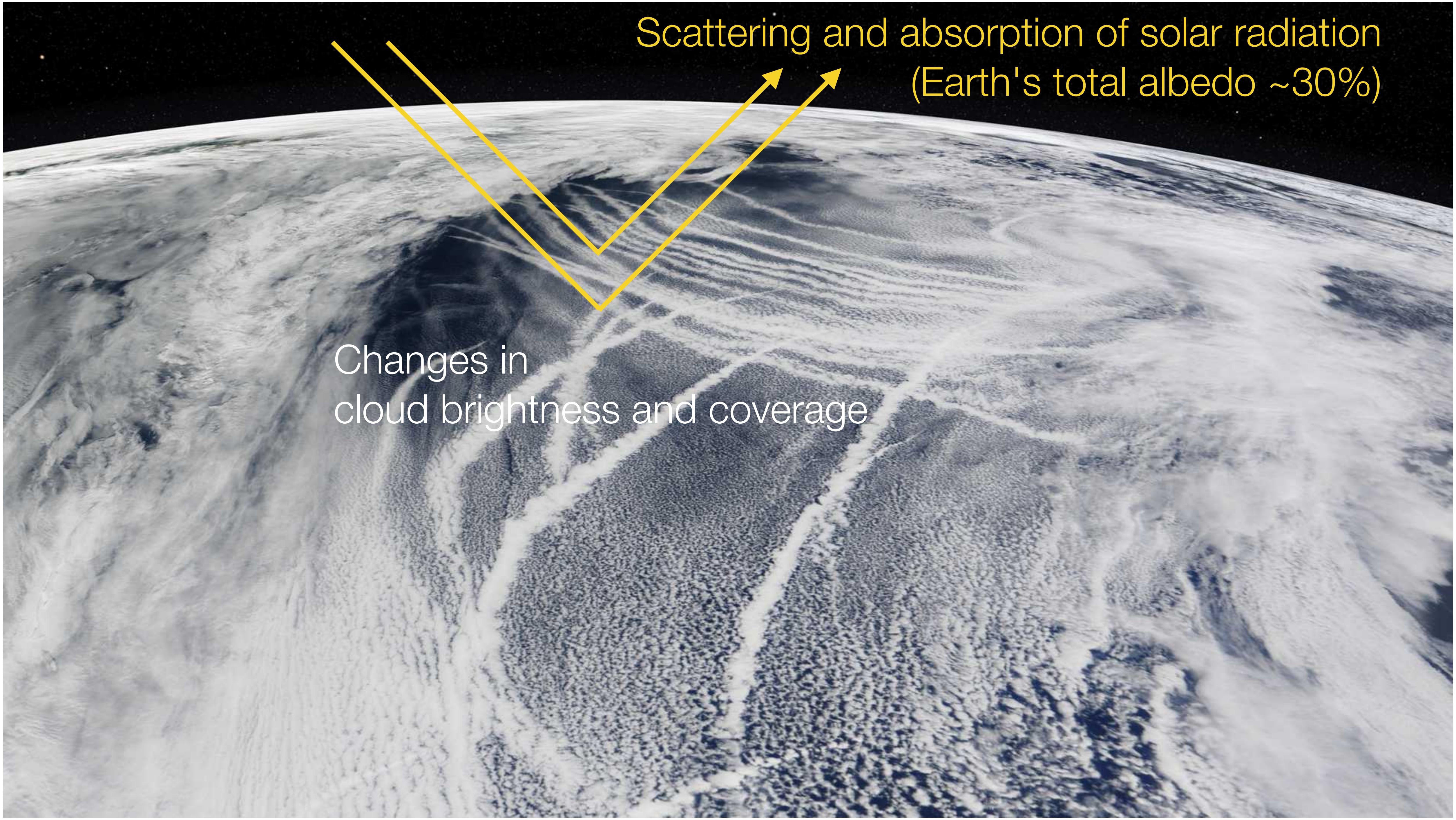
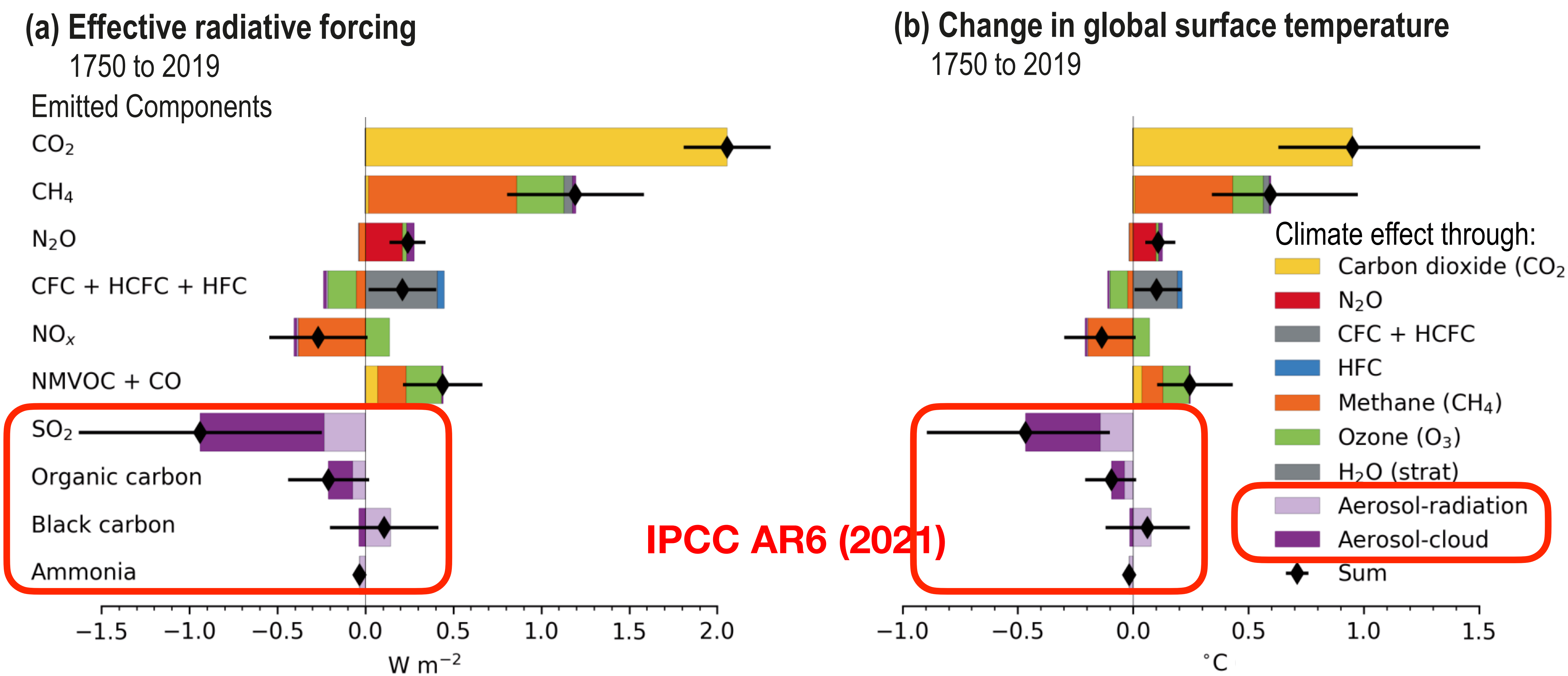
SPSC open session, CERN, 14Nov23

Influence of aerosol particles on climate



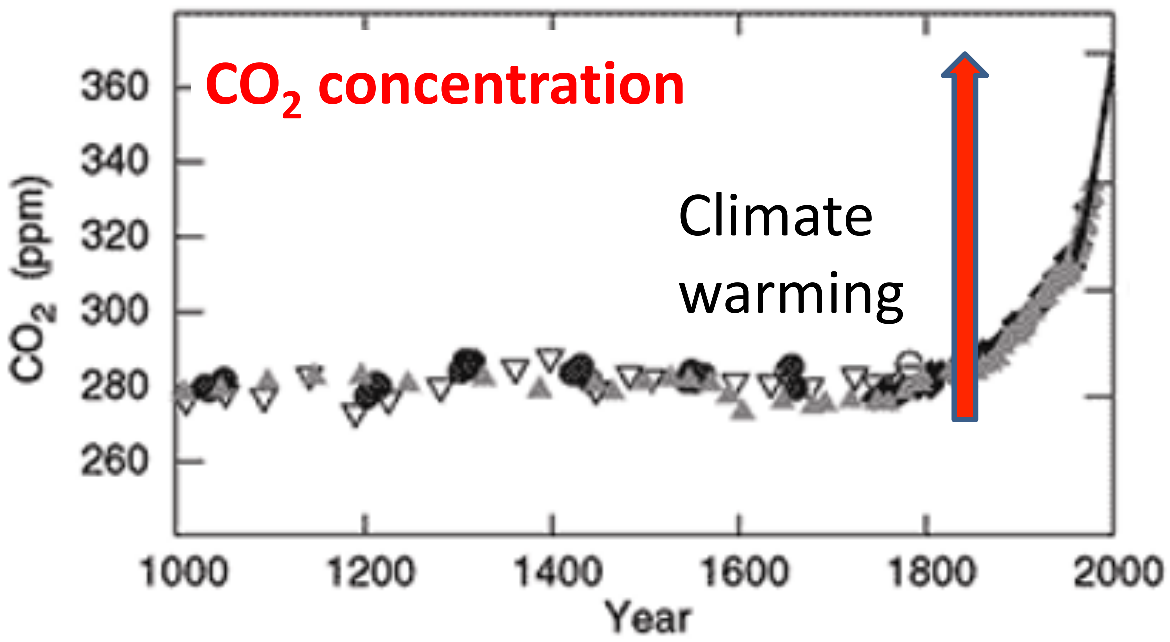
North Pacific, NASA MODIS satellite, 4Mar09

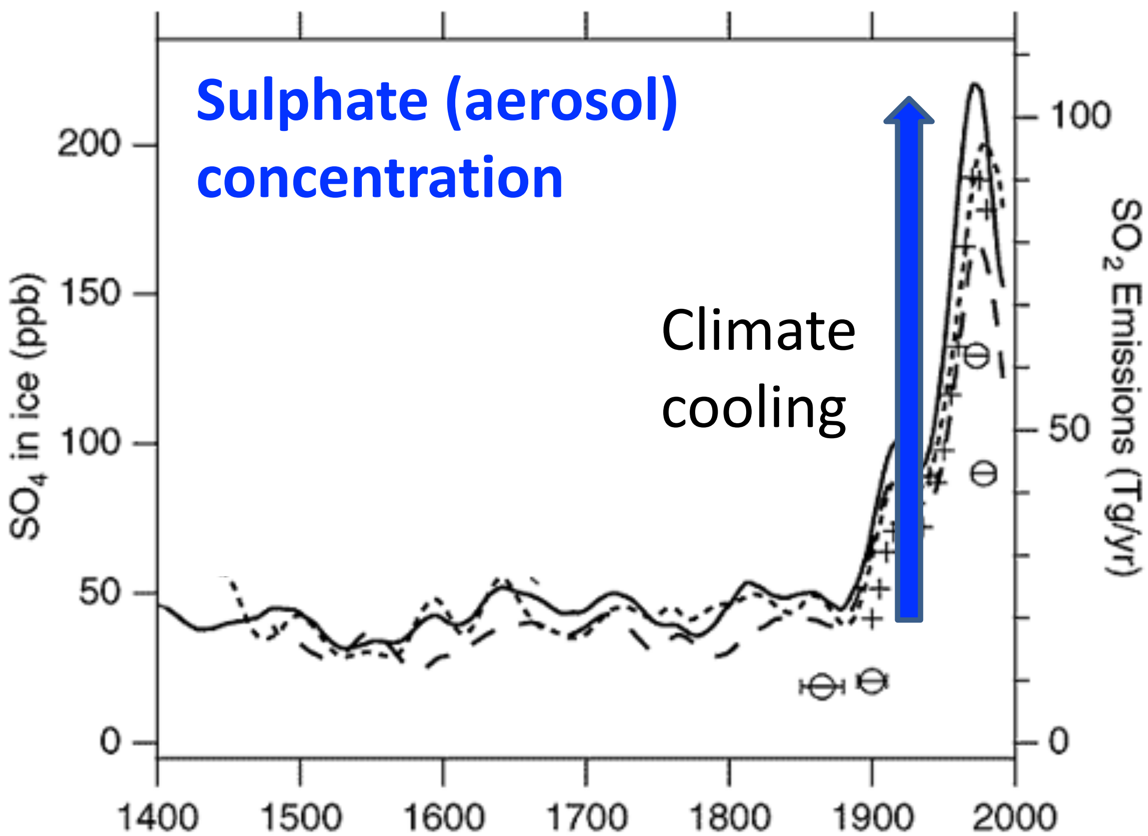
Radiative forcings since 1750 (IPCC AR6)



- Estimated aerosol effective radiative forcing = $-(1.3 \pm 0.7)$ Wm^{-2}
- Effective radiative forcing from CO_2 = (2.1 ± 0.3) Wm^{-2}

Climate prediction needs mechanistic representations for aerosol particles

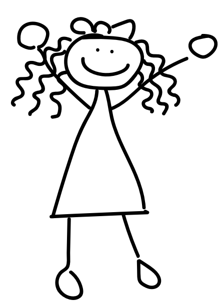




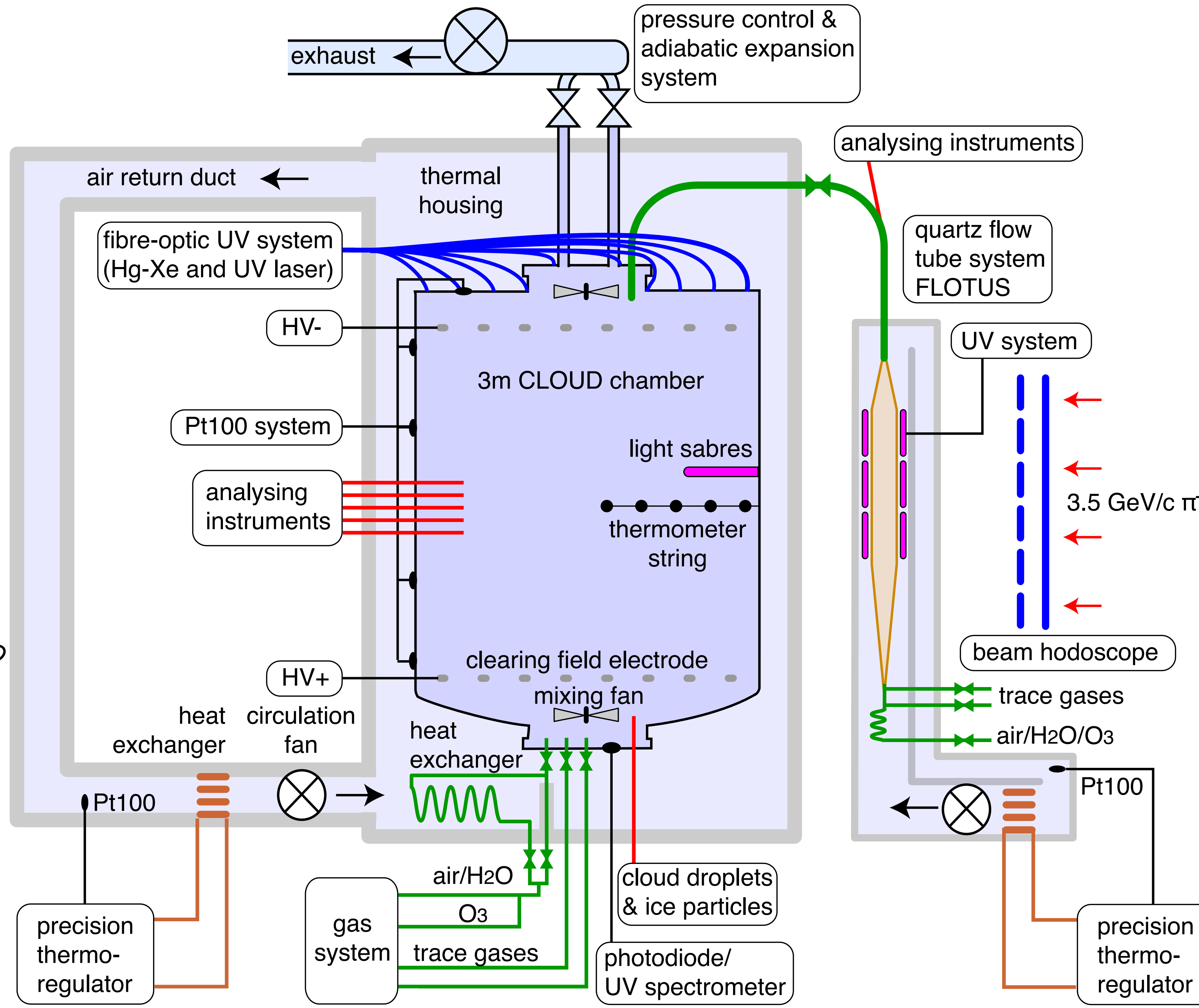
- Ice cores record greenhouse gas concentrations
- They also record sulphate (aerosol) mass concentrations
- But they provide no information on the *number* of aerosol particles, and hence cloudiness in the pre-industrial era
- We need climate models that include the underlying atmospheric chemical and physical mechanisms to predict:
  - Pre-industrial aerosol and clouds (=> Earth's climate sensitivity)
  - Earth's future climate without SO_2 emissions

CLOUD

- Key features::
  - Ultra-low contaminants
  - Atmospheric concentrations
  - Precise and steady control of all conditions over long periods:
    - vapours
    - T, UV
    - ionisation (n, gcr, π)
  - Comprehensive analysis instruments
  - FLOTUS (FLOw TUbe System)

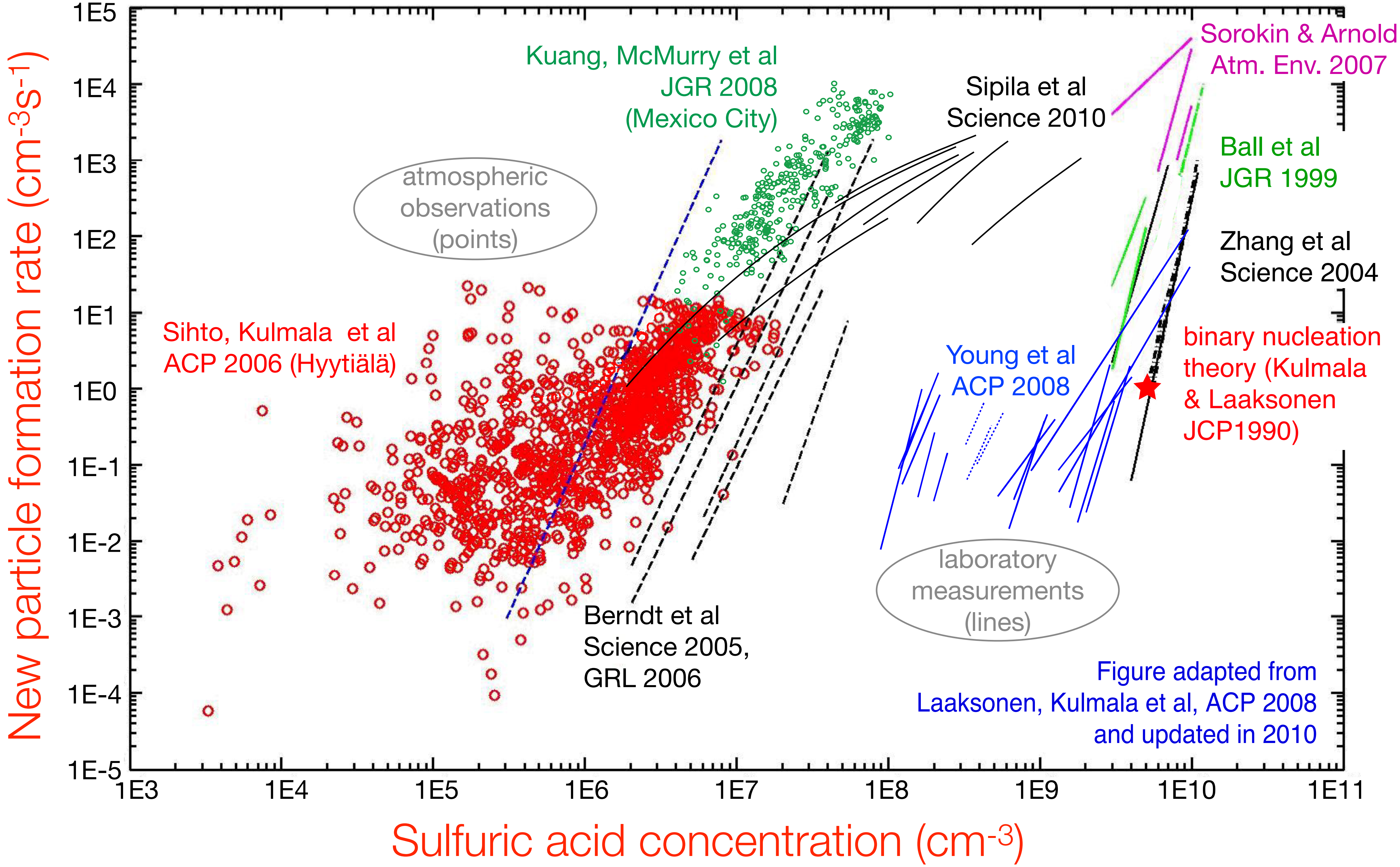


typical CLOUD scientist



CLOUD16 run (25Sep-3Dec23)

Understanding of atmospheric aerosol BC (Before CLOUD)



- In 2010, sulfuric acid was thought to account for almost all particle formation in the atmosphere - but laboratory experiments disagreed by many orders of magnitude

CLOUD advances in atmospheric aerosol 2011-23

	Year (cites.)	
nature	2011 (912)	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 Dunne[4], Jonathan Duplissy[1,5,6], Sebastian Ehrhart[2],
nature	2013 (658)	LETTER OPEN doi:10.1038/nature12663 **Molecular understanding of sulphuric acid–amine particle nucleation in the atmosphere** João Almeida[1,2], Siegfried Schobesberger[3], Andreas Kürten[1], Ismael K. Ortega[3], Oona Kupiainen-Määttä[3], Arnaud P. Praplan[4],
Science	2014 (370)	**Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles** Francesco Riccobono,[1]* Siegfried Schobesberger,[2] Catherine E. Scott,[3] Josef Dommen,[1]
nature	2016 (349)	LETTER OPEN doi:10.1038/nature17953 **Ion-induced nucleation of pure biogenic particles** Jasper Kirkby[1,2], Jonathan Duplissy[3,4], Kamalika Sengupta[5], Carla Frege[6], Hamish Gordon[2], Christina Williamson[1]†,
nature	2016 (454)	LETTER OPEN doi:10.1038/nature18271 **The role of low-volatility organic compounds in initial particle growth in the atmosphere** Jasmin Tröstl[1], Wayne K. Chuang[2], Hamish Gordon[3], Martin Heinritzi[4], Chao Yan[5], Ugo Molteni[1], Lars Ahlm[6], Carla Frege[1],
Science	2016 (238)	RESEARCH ARTICLE ATMOSPHERIC SCIENCE **Global atmospheric particle formation from CERN CLOUD measurements** Eimear M. Dunne,[1]*† Hamish Gordon,[2]*‡ Andreas Kürten,[3] João Almeida,[2,3]
nature	2020 (98)	Article **Rapid growth of new atmospheric particles by nitric acid and ammonia condensation** Mingyi Wang[1,2,30], Weimeng Kong[3,30], Ruby Marten[4], Xu-Cheng He[5], Dexian Chen[1,6],
Science	2021 (66)	RESEARCH ARTICLE AEROSOL FORMATION **Role of iodine oxoacids in atmospheric aerosol nucleation** Xu-Cheng He[1]*, Yee Jun Tham[1], Lubna Dada[1], Mingyi Wang[2], Henning Finkenzeller[3],
nature	2022 (13)	Article **Synergistic HNO_3–H_2SO_4–NH_3 upper tropospheric particle formation** https://doi.org/10.1038/s41586-022-04605-4 Mingyi Wang[1,2,7], Mao Xiao[3], Barbara Bertozzi[4], Guillaume Marie[5], Birte Rörup[6],

- CLOUD publications total around 75, including:
  - 6 in Nature
  - X 4 in Science
  - 1 in Nature Geosc.
  - 1 in Nature Chem.
  - 4 in Proc. Natl. Acad. Sci. USA
  - 2 in Nature Com.
  - 4 in Science Adv.

Science 2023 (embargoed) He et al

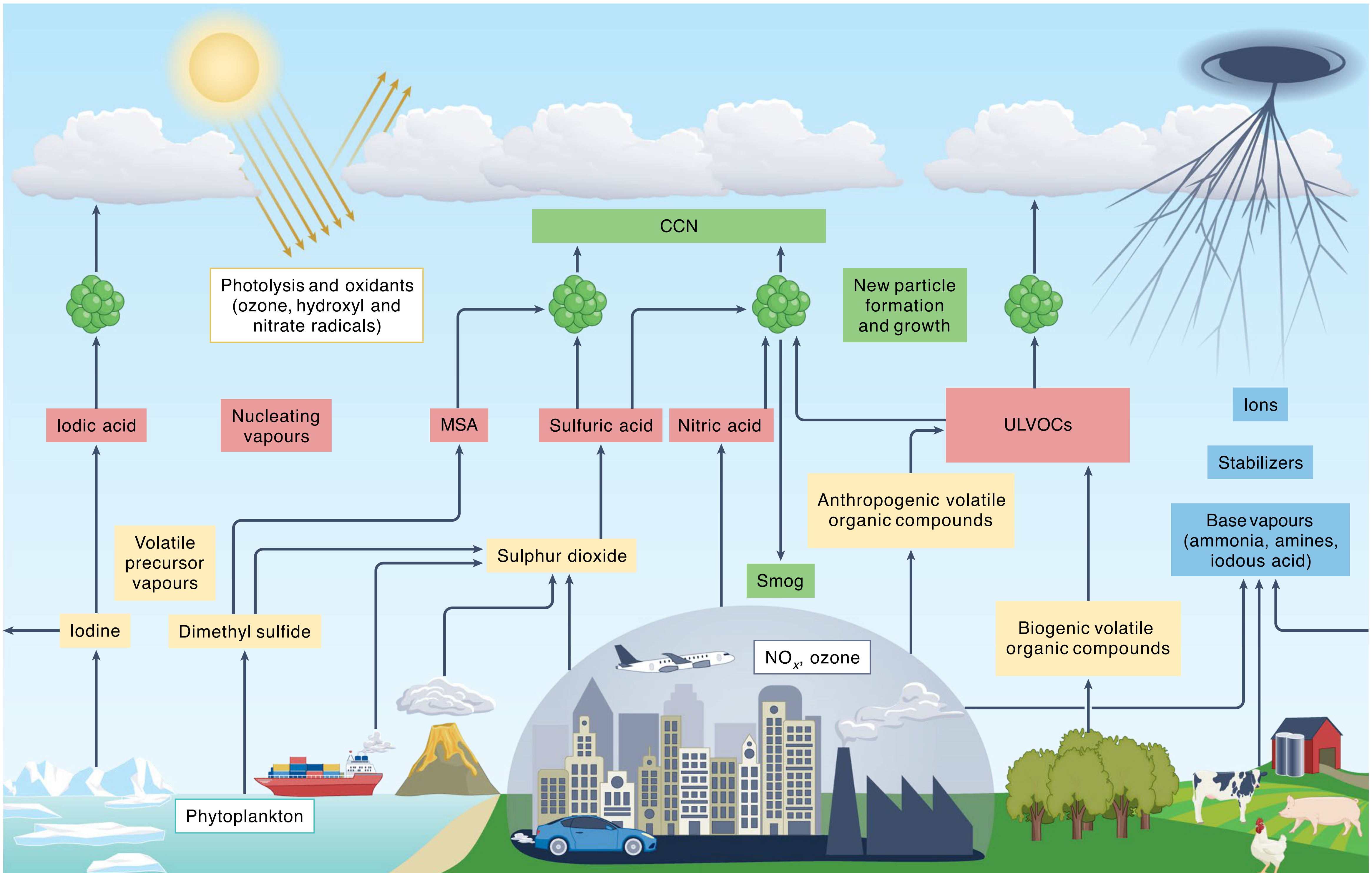
nature geoscience

Published online: 7 November 2023

Perspective

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

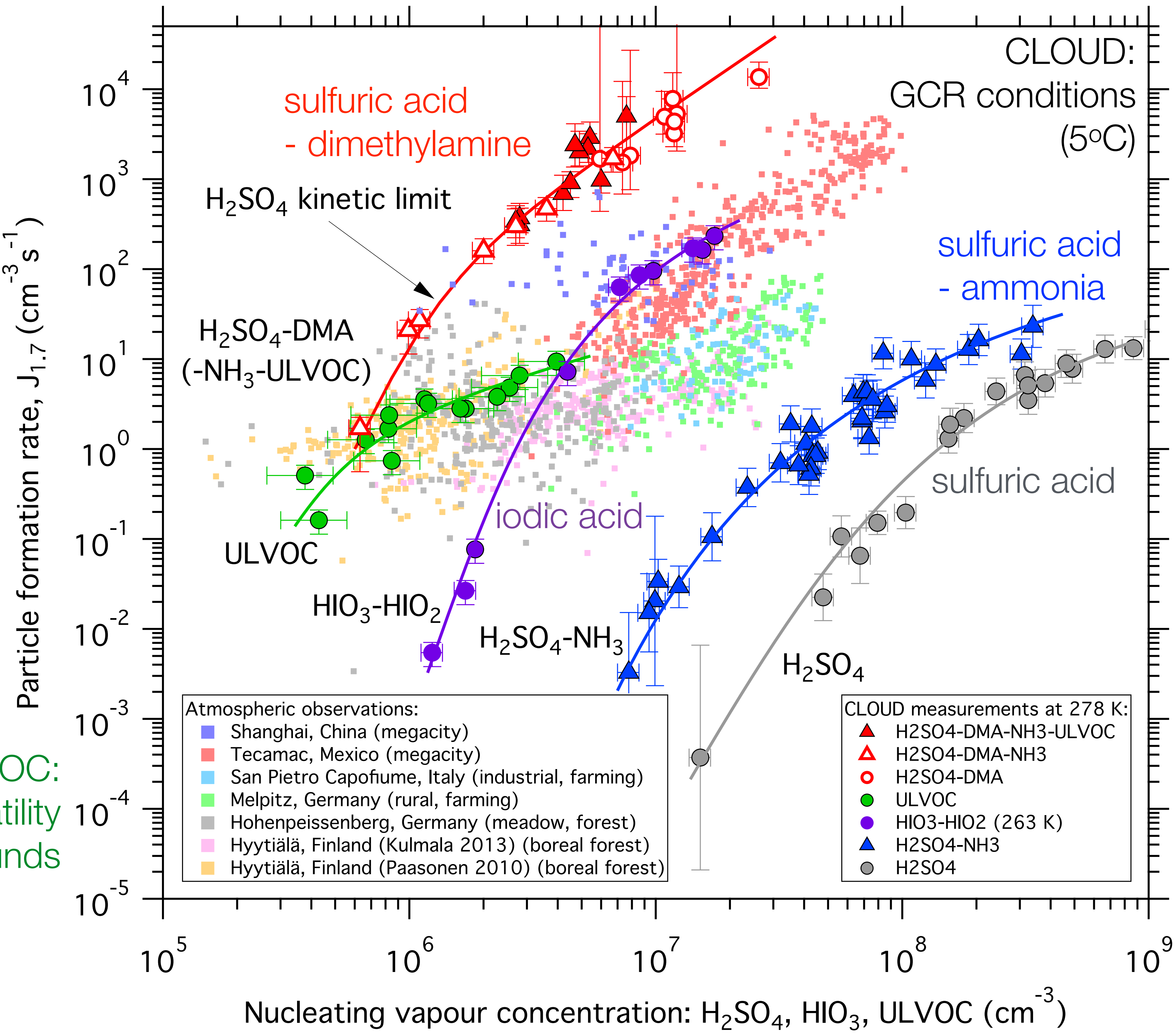
Atmospheric new particle formation from the CERN CLOUD experiment



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

New particle formation from CLOUD

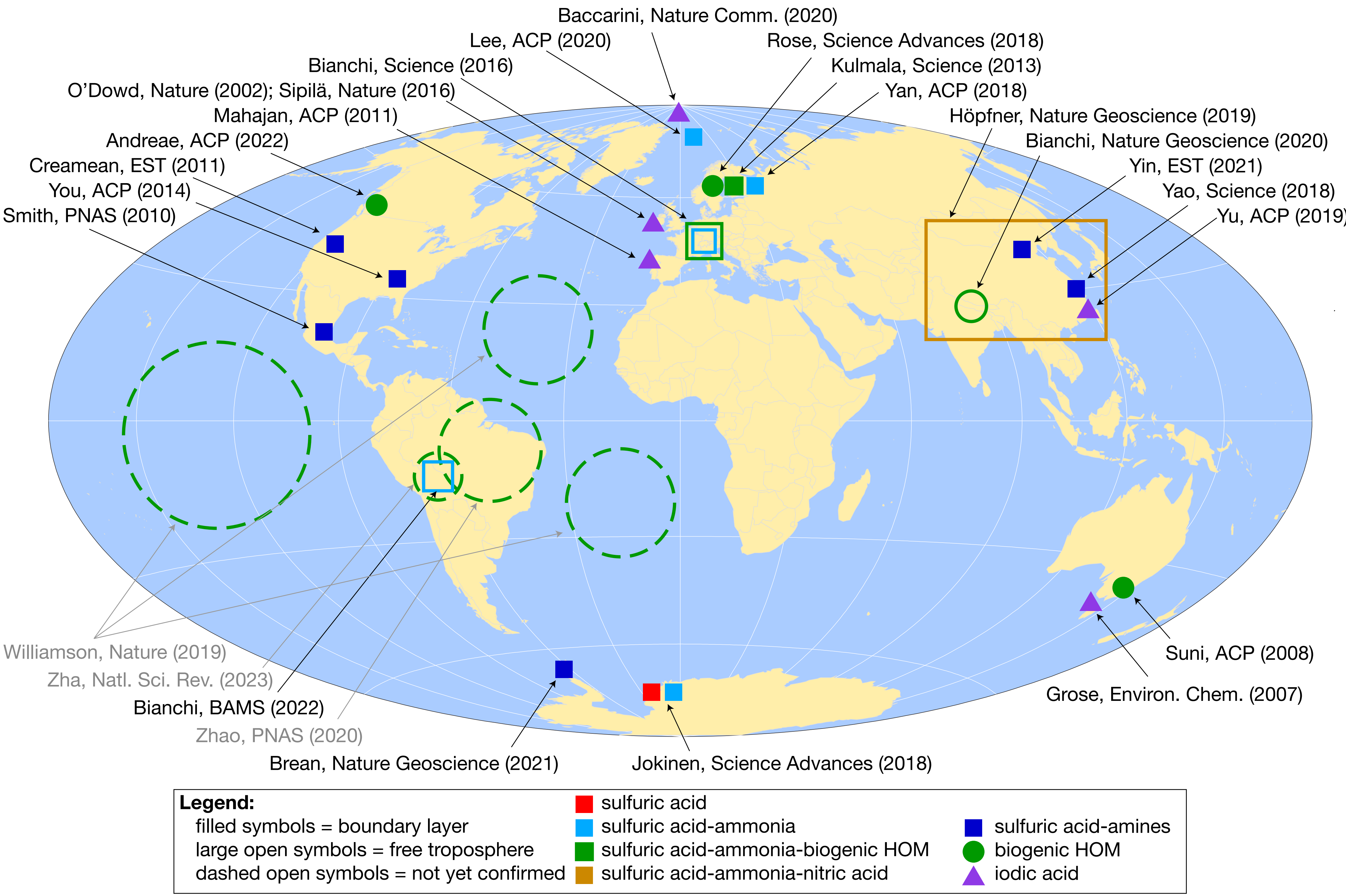
Kirkby et al., Nature Geosci. 16, 948-957 (2023)



ULVOC:
Ultra-low-volatility
organic compounds

- CLOUD measurements so far indicate that GCR ions account for 27% of global CCN for low clouds
- However GCR variations over solar cycle change CCN by only 0.2%, which is not climatically significant

Geographical locations of nucleation mechanisms measured by CLOUD



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

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?

CLOUD16 run (25Sep-3Dec23)

- Tropical rainforest upper free troposphere:
  - α-pinene, isoprene
  - sulfuric acid
  - NOx

- Marine surfactants in the upper free troposphere:
  - nonanal ($(CH_2)_9O$)
  - sulfuric acid
  - NOx

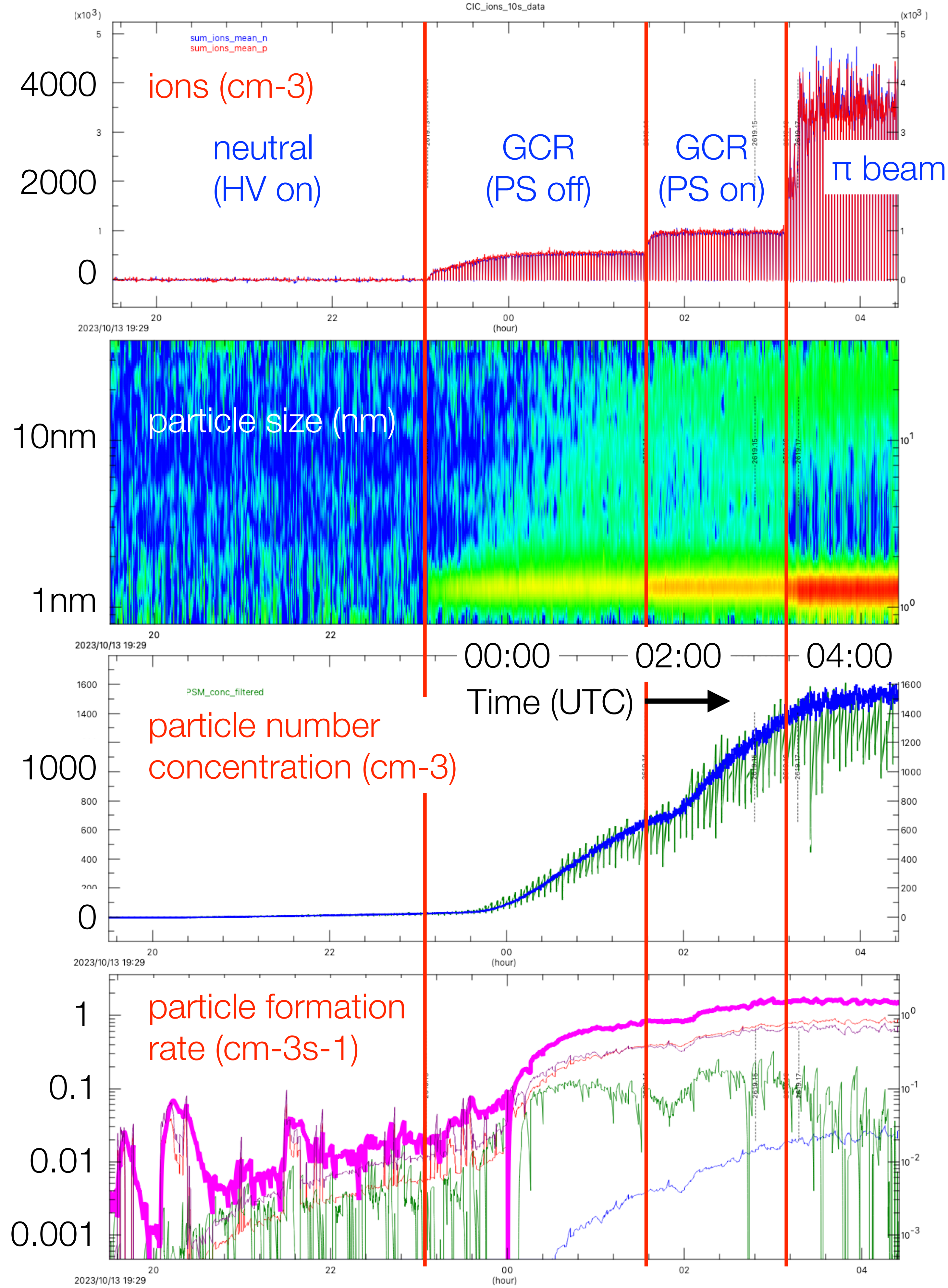
- Cool boreal forest boundary layer:
  - α-pinene
  - sulfuric acid

- Arctic boundary layer:
  - dimethylsulfide (methanesulfonic acid, sulfuric acid)
  - iodine (iodic acid, iodous acid)
  - ammonia
  - glyoxal (dialdehyde, CHOCHO)

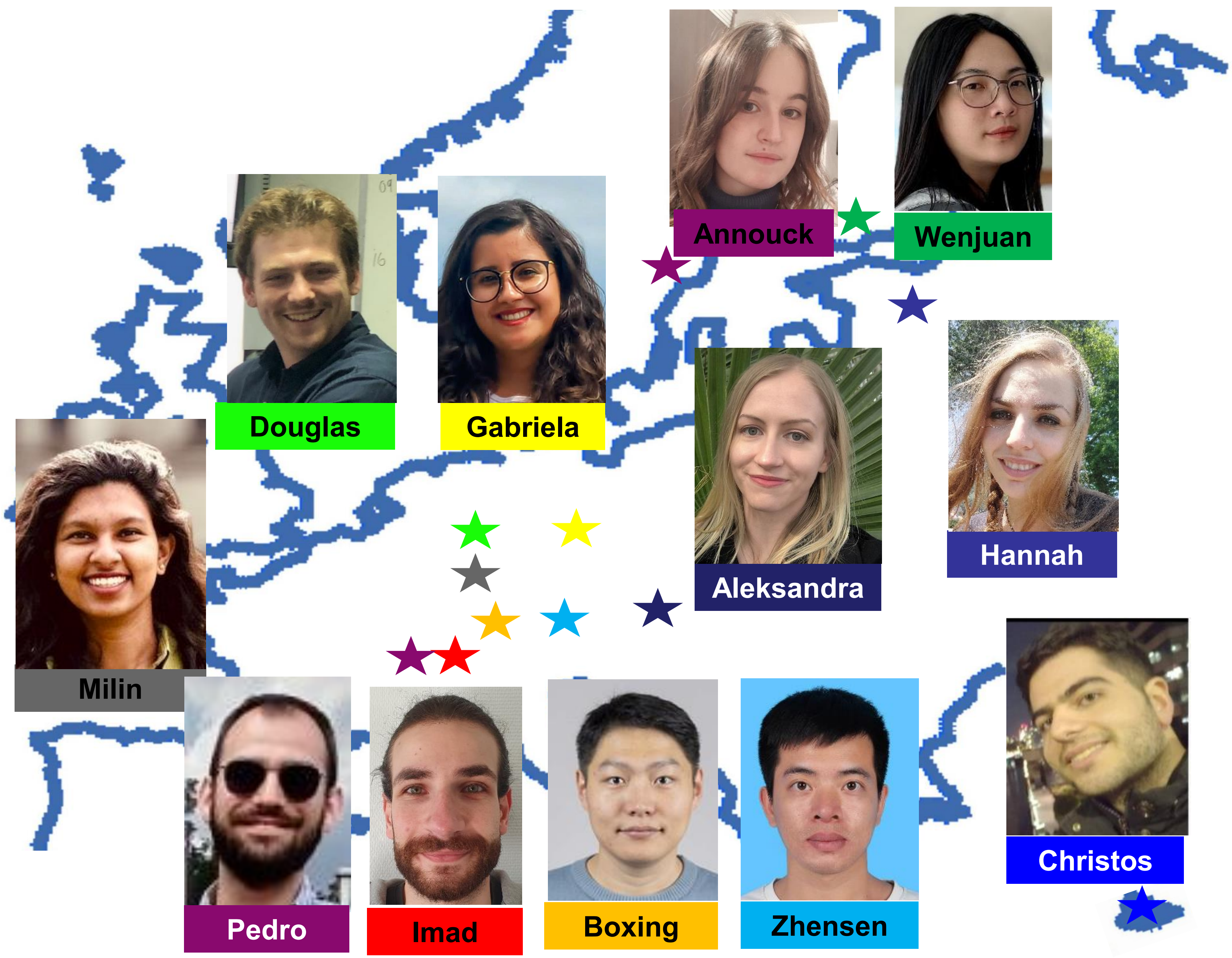
- Interaction of biogenic and anthropogenic vapours in urban environments:
  - biogenic vapours (trees): α-pinene, isoprene
  - anthropogenic vapours (automobiles, industry...): sulfuric acid, ammonia, dimethylamine, aromatic organics, NOx

CLOUD16 example run
(13-14Oct23)

- Aerosol particle formation in the tropical rainforest upper free troposphere (CLOUD chamber at -50^{o}C)



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€)

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

Summary

- 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