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Status and plans of the CLOUD experiment

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On behalf of the CLOUD collaboration:

Austria: University of Innsbruck
University of Vienna
Cyprus: Cyprus Institute
Estonia: Univ. of Estonia, Tartu
Finland: Finnish Meteorological Institute
University of Helsinki, INAR
University of Eastern Finland
Germany: Goethe University Frankfurt
Karlsruhe Institute of Technology
Leibniz Institute for Tropospheric Res.
Max Planck Institute for Chemistry

Portugal: University of Beira Interior
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Russia: Lebedev Physical Institute
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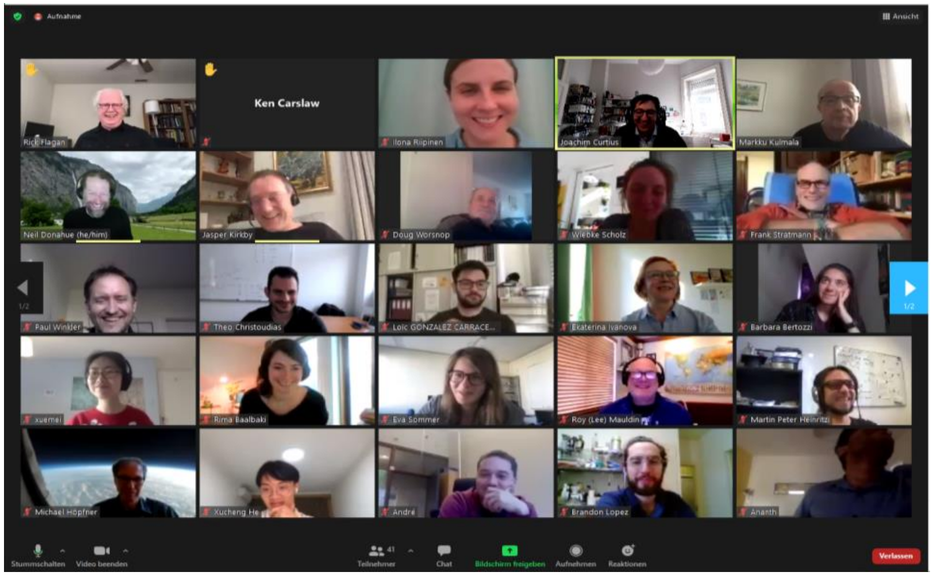
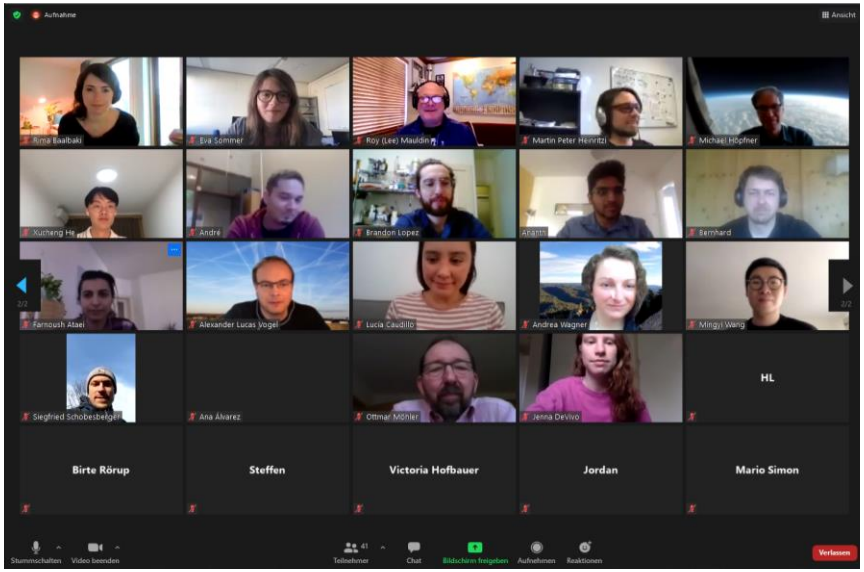




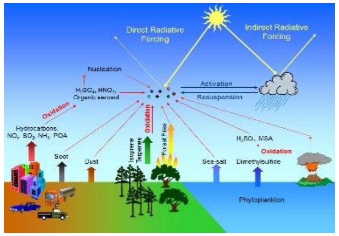
CLOUD-MOTION, final conference, online, April 2021

Relevance of atmospheric aerosol

- **Cloud formation**
  (CCN & INP, Dusek et al., Science, 2007;
  Froyd et al., Nature Geosci., 2022)

- **Visibility**
  (e.g., v. Marle et al., Nature, 2022)

- **Climate**
  (IPCC AR6 2021; Bellouin et al., Rev. Geophys. 2019)



- **Health effects**
  (7 Mio premature deaths)
  (WHO report; Lelieveld et al., Cardiovasc. Res, 2020)

- **Heterogeneous reactions**
  (e.g., on PSCs; Peter, Ann. Rev. Phys. Chem., 1997)

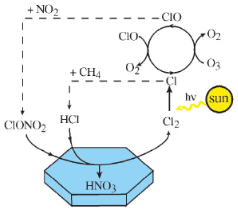
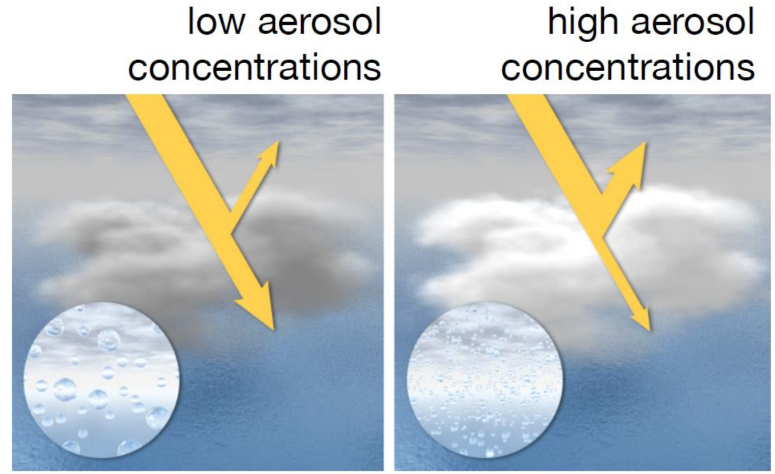
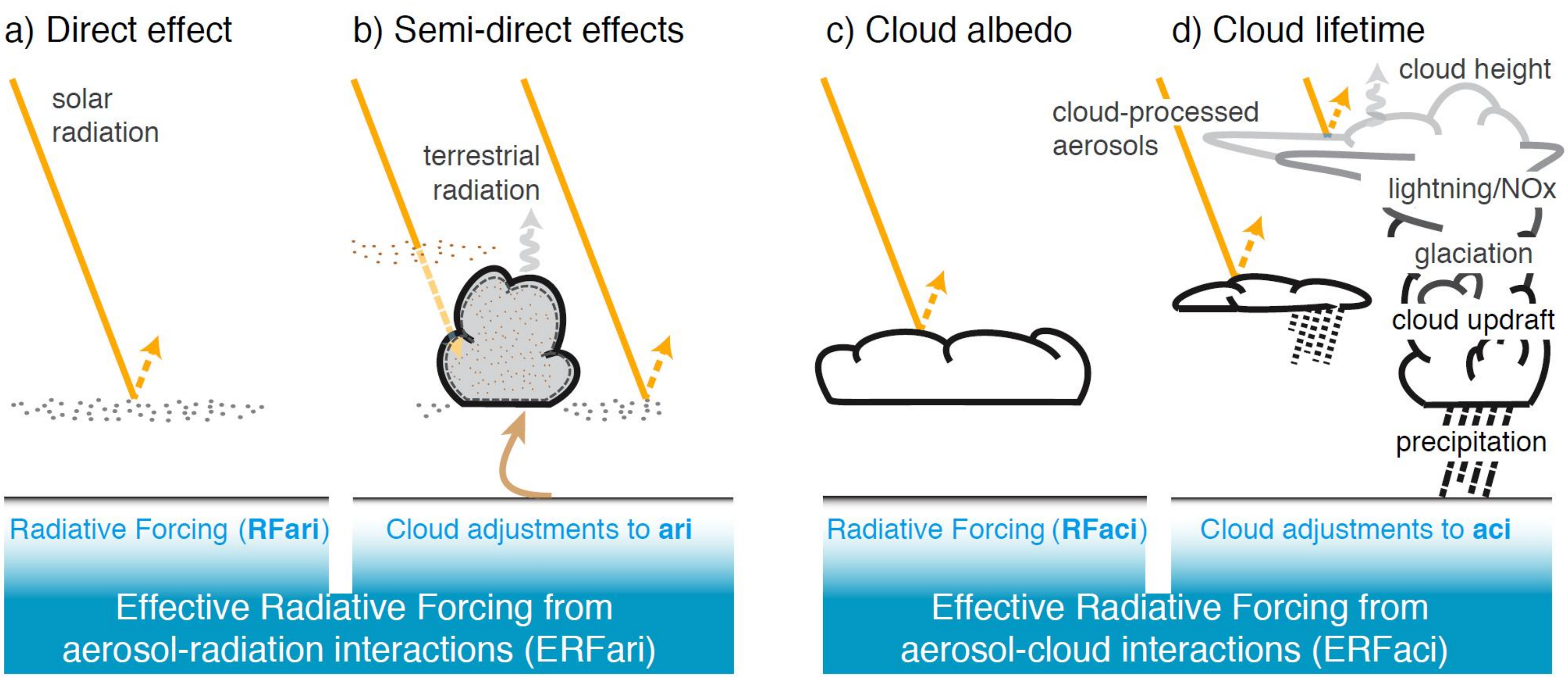


Figure 2 Schematic diagram of the reactions leading to polar ozone depletion.

Aerosol effects on clouds and climate

- Aerosols above ~50nm constitute Cloud Condensation Nuclei (CCN)

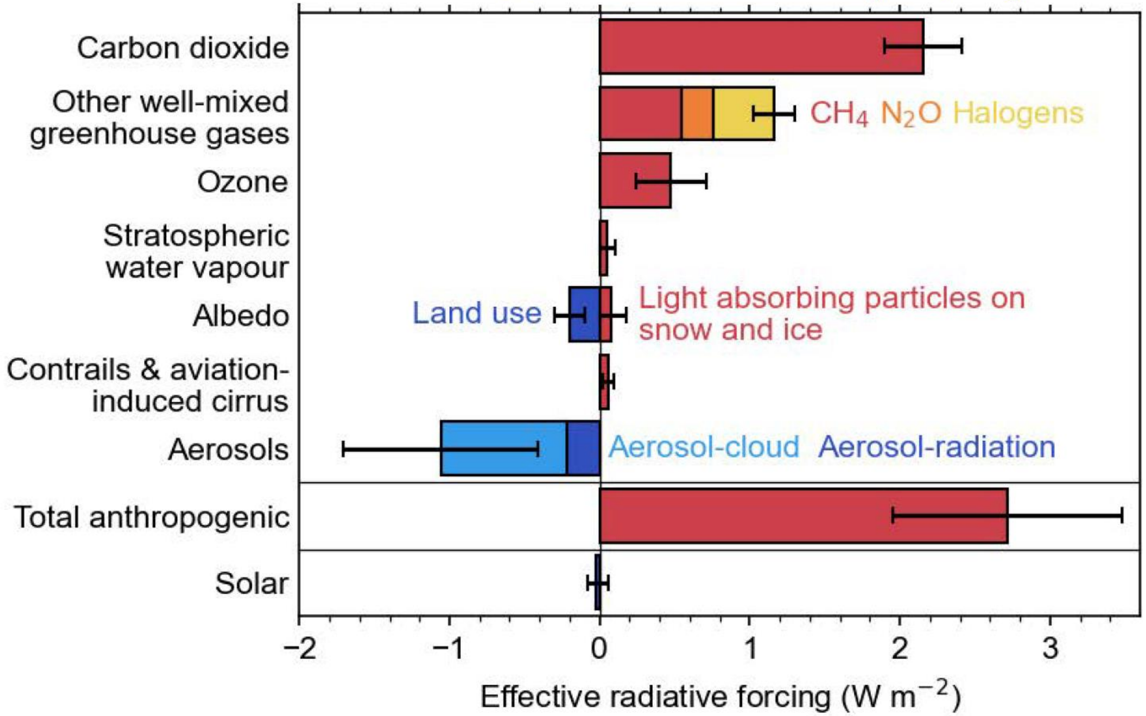




IPCC AR5, 2013

Aerosols are important for climate change

Change in effective radiative forcing from 1750 to 2019
IPCC 6th Assessment Report, August 2021

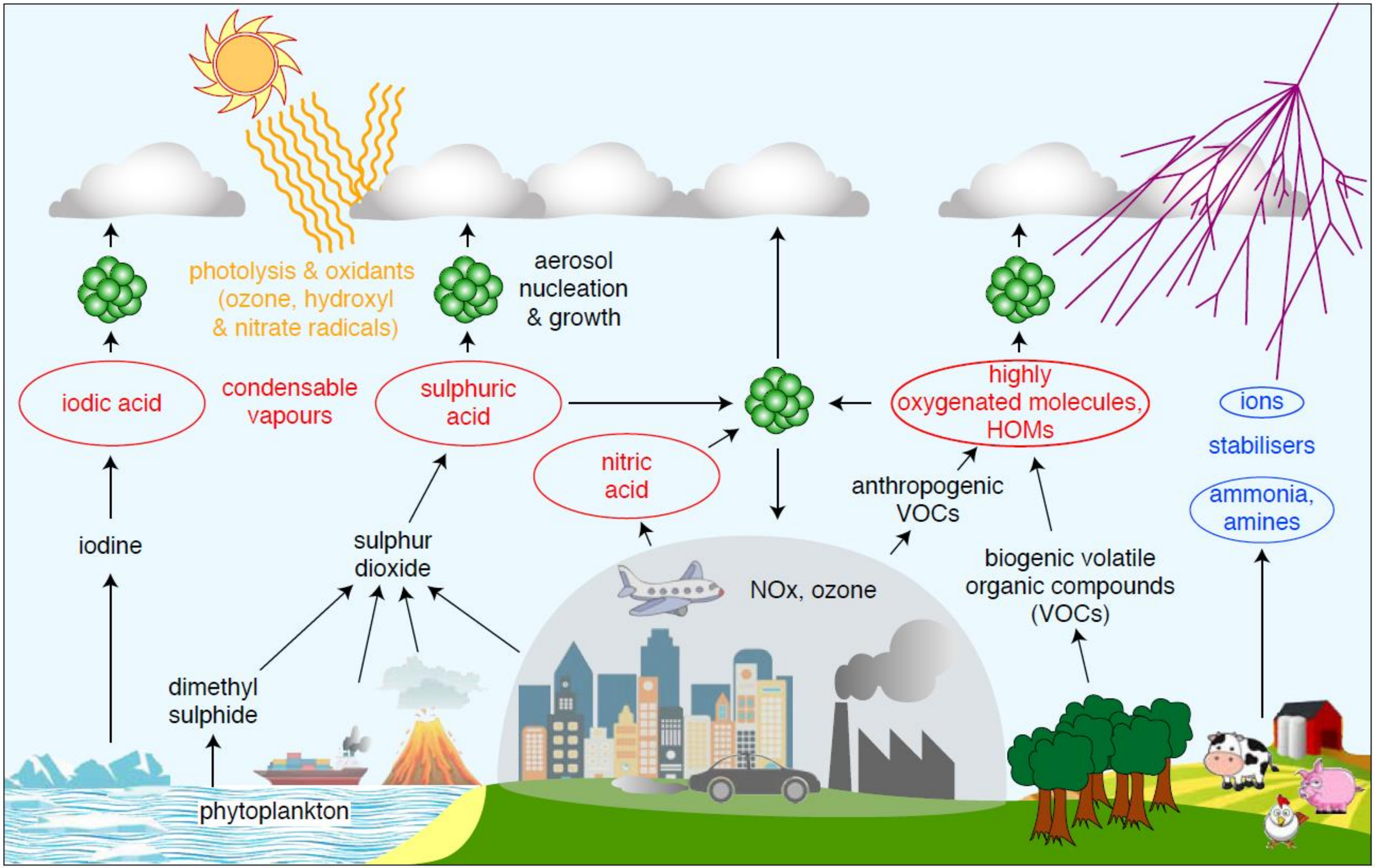


red = warming effect
blue = cooling effect

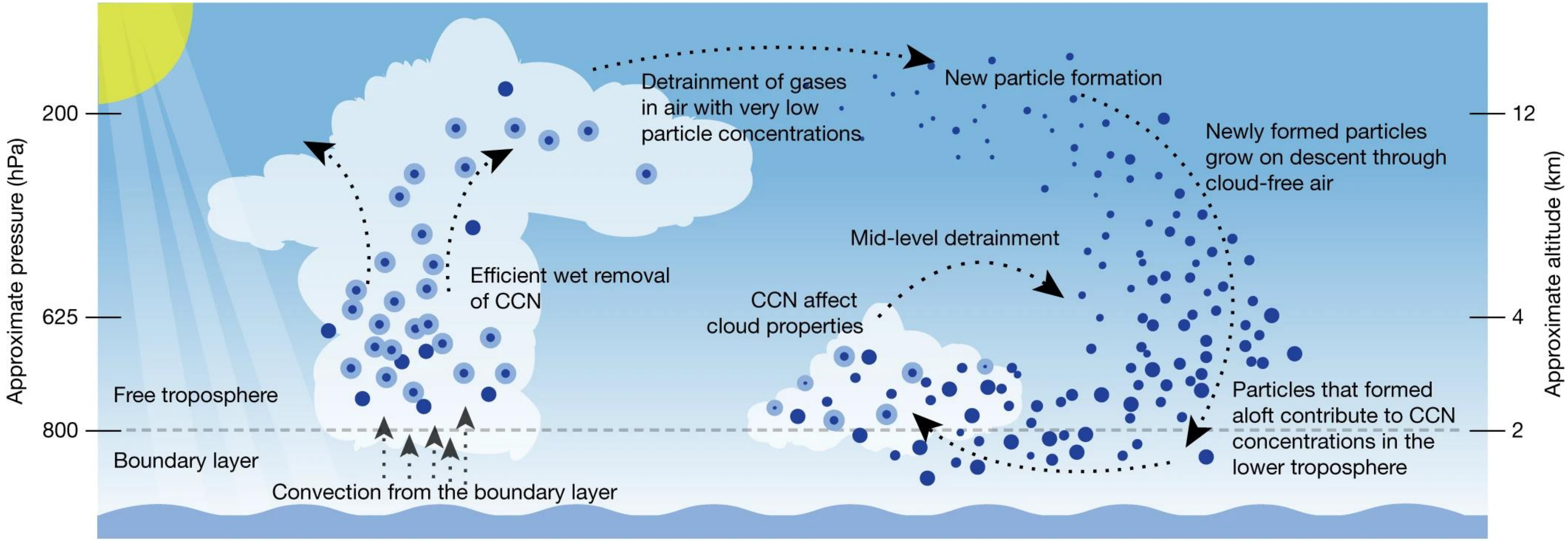
- Changes of aerosols since pre-industrial times have offset a large but poorly understood fraction of warming from greenhouse gases
- The uncertainty in total anthropogenic radiative forcing is dominated by aerosols
- This results in a factor 3 uncertainty in Earth's climate sensitivity and expected warming in 21st century:
    - 1.5-4.5^{o}C for a doubling of CO_2 (AR5)
- Future emissions reductions eg. SO_2 will reduce the cooling from aerosols. But by how much?

CLOUD studies the formation of new aerosol particles from precursor gases

Several gases were discovered to play a role for atmospheric new particle formation



Recent measurements in the tropical upper troposphere show occurence of high levels of aerosol nucleation

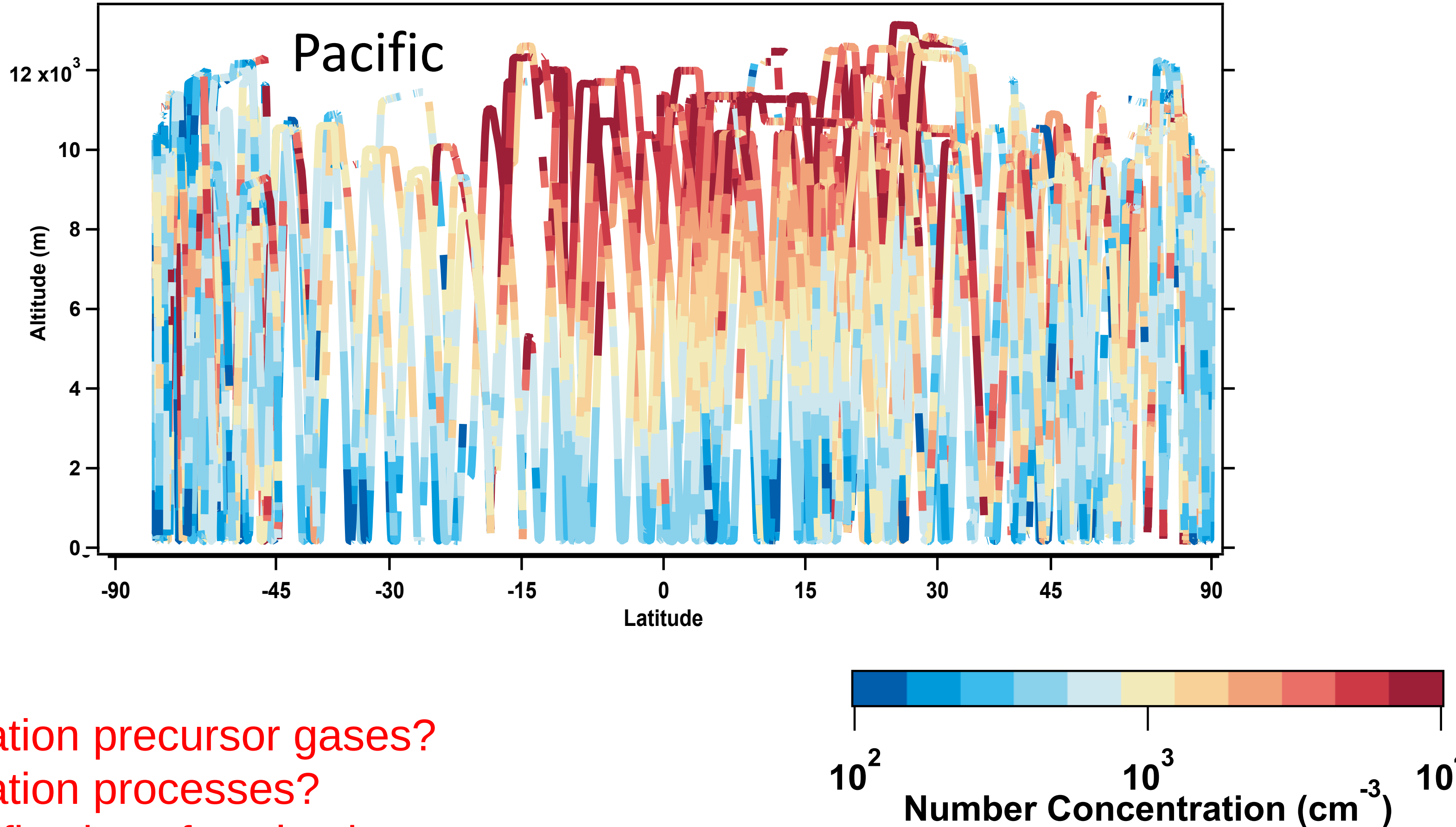


Williamson et al., Nature, 2019

Nucleation precursor gases?
Nucleation processes?
Quantification of nucleation and growth rates?

→ CLOUD

Recent measurements in the tropical upper troposphere show frequent occurence of high levels of aerosol nucleation



But:
Nucleation precursor gases?
Nucleation processes?
Quantification of nucleation rates and growth rates?

→ CLOUD

Williamson et al., Nature, 2019

T11 beam area with CLOUD Experiment
after East Hall Renovation during CLOUD15T run in spring 2022

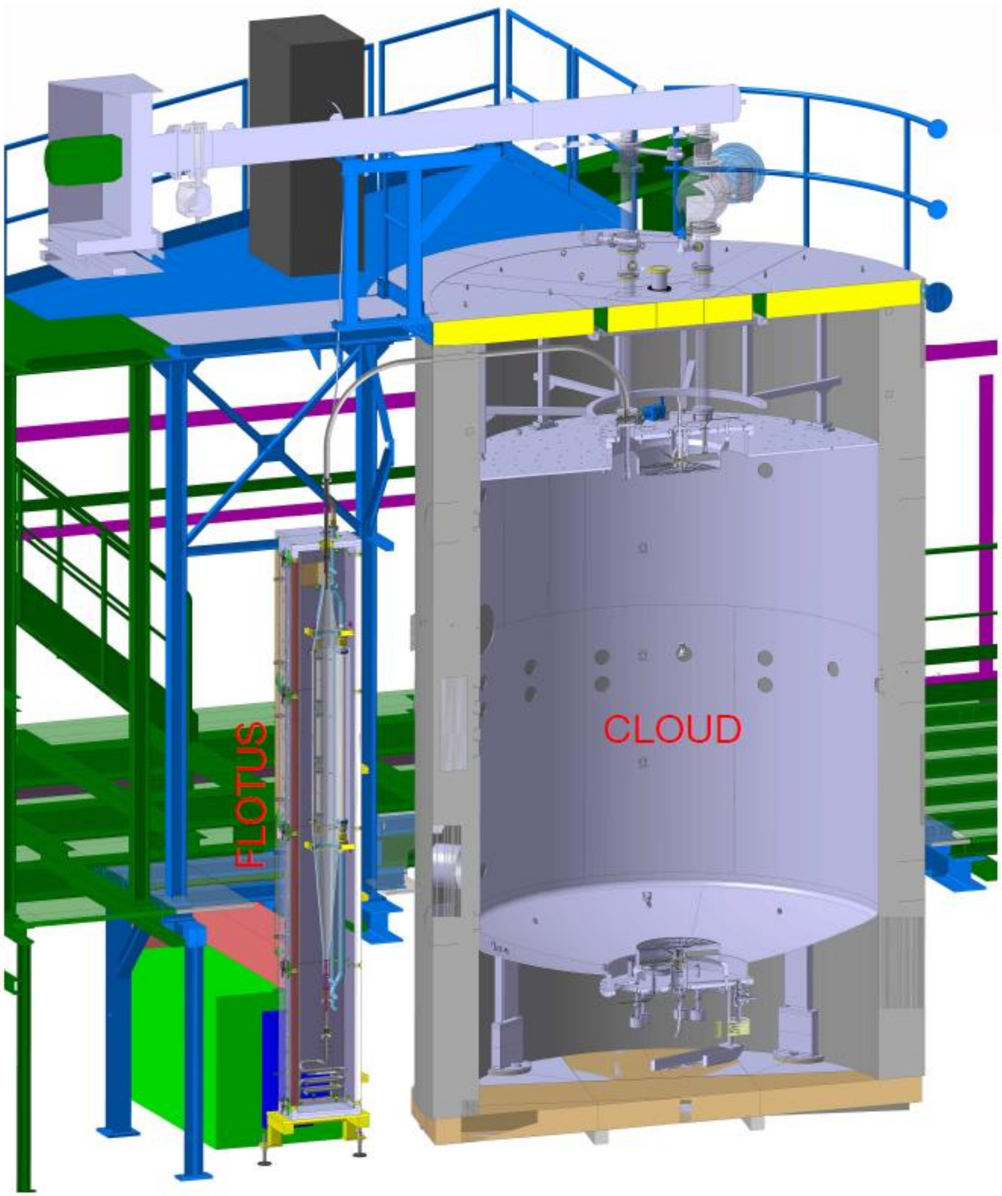
- Improved space and infrastructure
- New control room, new chemical room
- Rebuilt gas system & IT network
- New T11 beamline magnets and power supplies

CLOUD 2022

New experimental facility
FLOTUS (FLOw TUbe System)
set up for preparation
of pre-aged aerosol particles

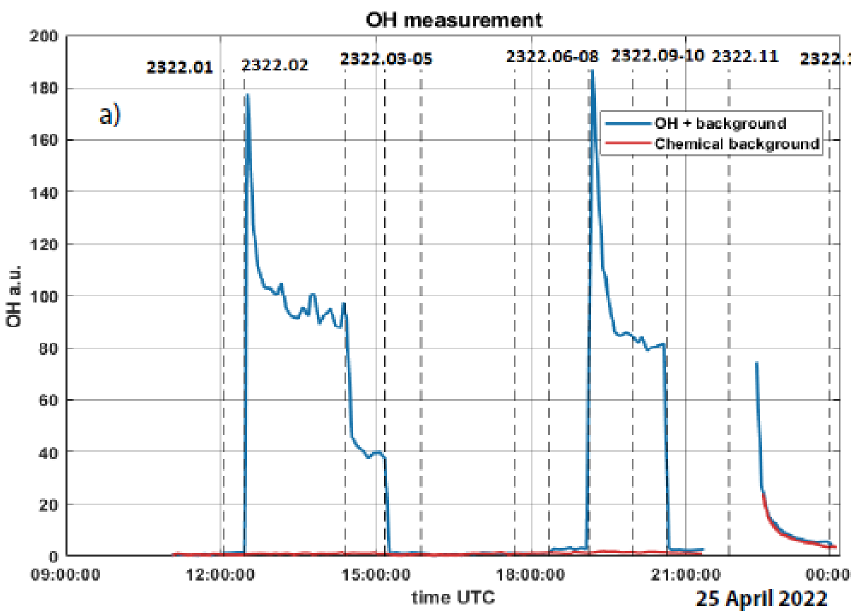
Intense UV exposure to simulate
Several days of oxidative
exposure.

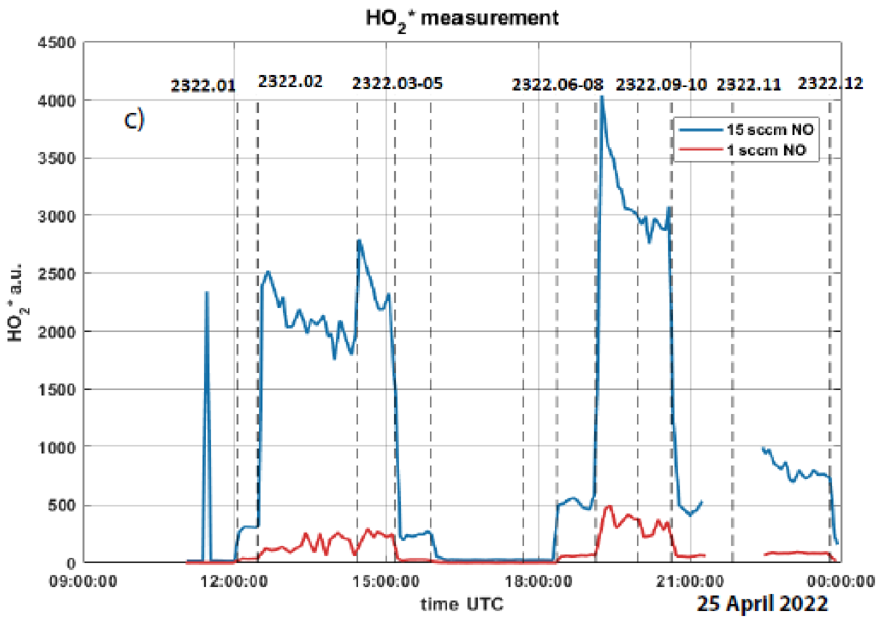
Greatly extends science
reach of CLOUD.



Extension of experimental capabilities
through **HORUS instrument** from MPI-Chemistry:

- Allows direct measurement of OH and HO_2 radicals
- OH is the most important reactive chemical in the troposphere
- Highly challenging measurement: OH concentrations around 0.1 pptv and OH lifetime below 1 s

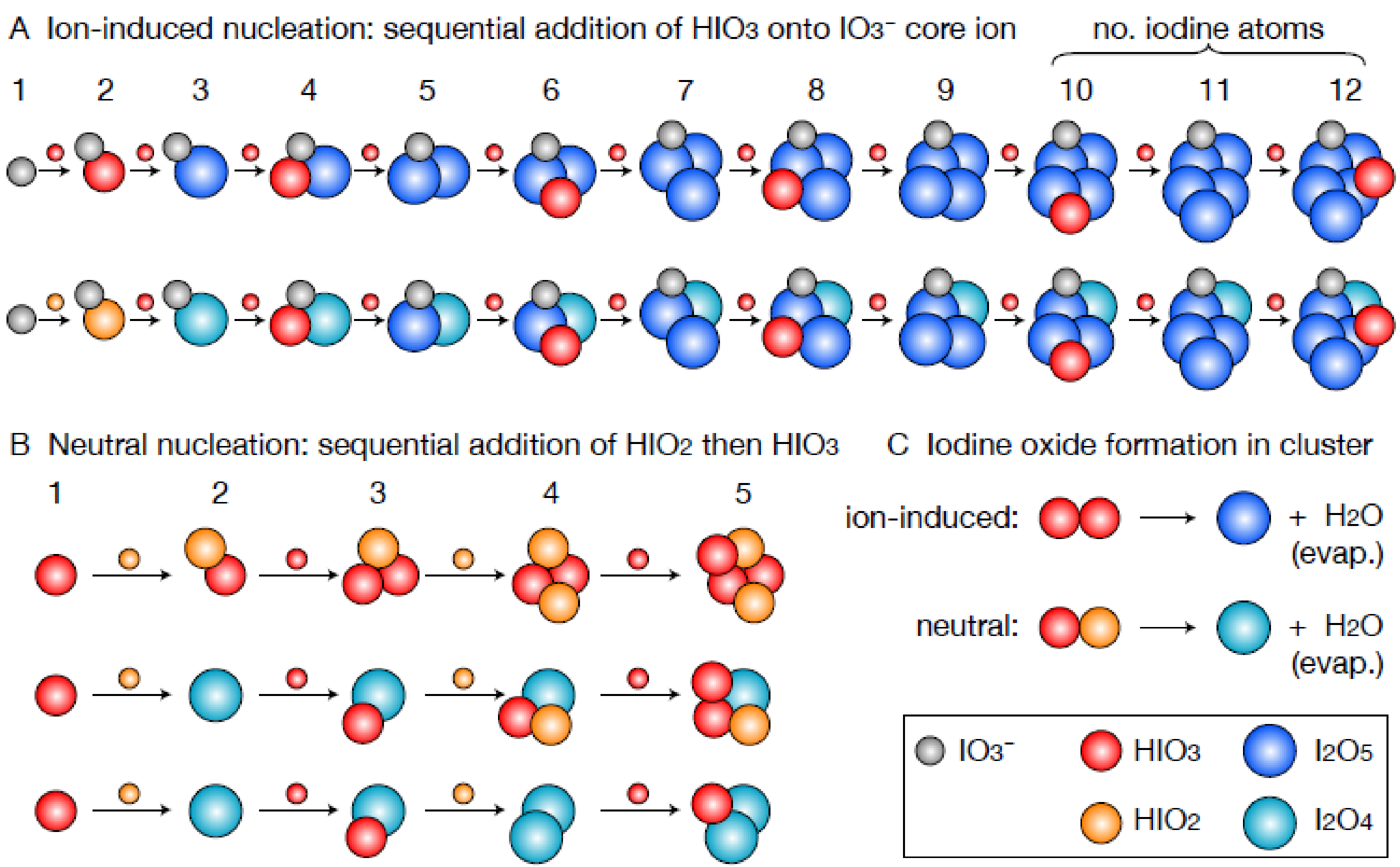




Results

Study of nucleation for iodic acid HIO_3:
Detailed mechanism for charged and neutral nucleation identified:

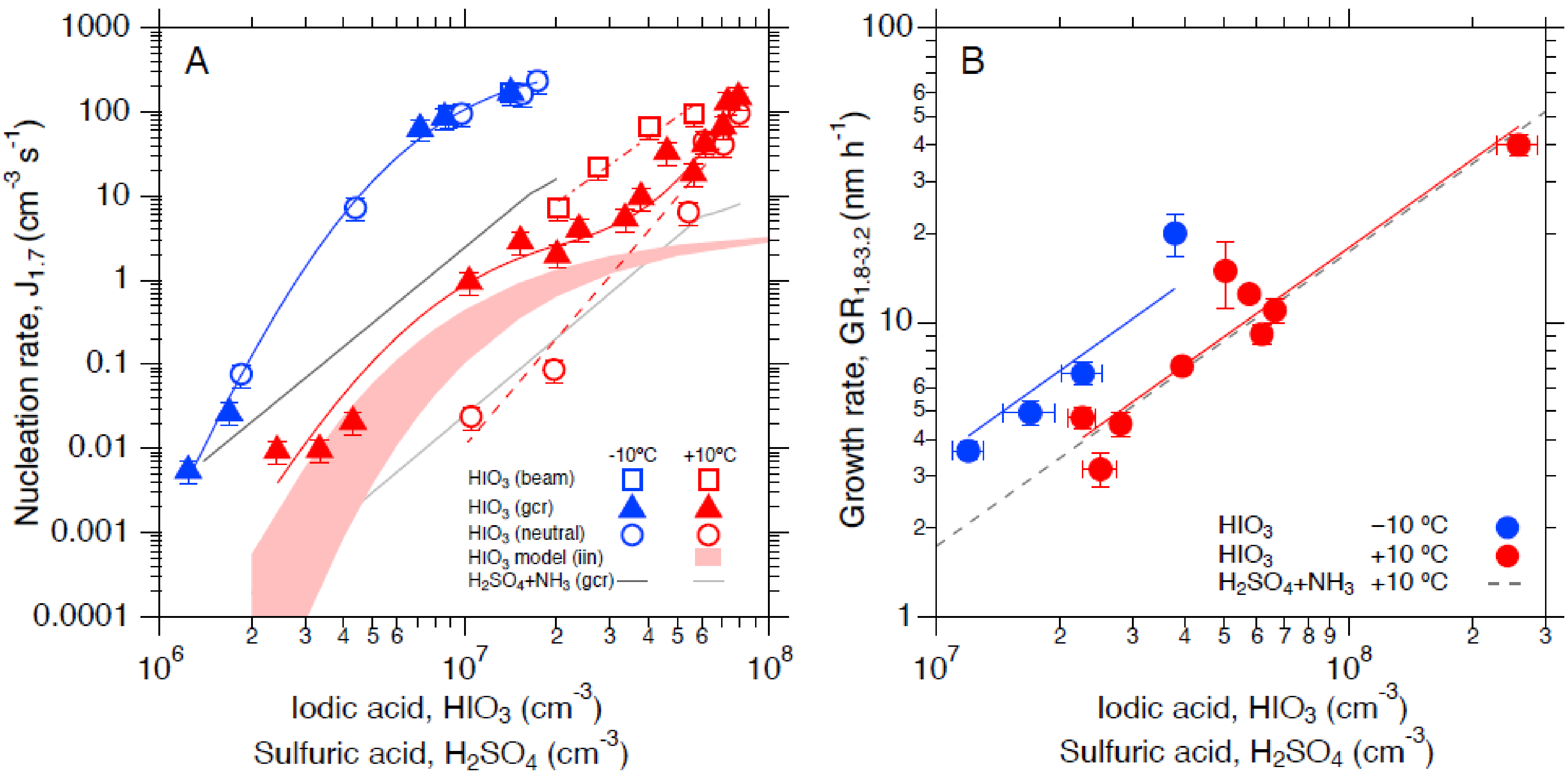
He et al., Science, 2021: Importance for Arctic marine and coastal nucleation.
3-fold increase of atmospheric iodine due to sea ice thinning



Results

Study of nucleation for iodic acid HIO_3:
Determination of nucleation rates (A) and growth rates (B);
HIO_3 identified as potent nucleator
Strong enhancement of particle formation rate at +10C from GCR ions (up to x100)

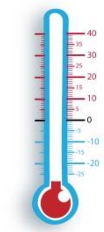
He et al., Science, 2021



CLOUD Experiments

Can we replicate atmospheric conditions for deep convective lifting of precursors
in East Asian monsoon?

UV



T

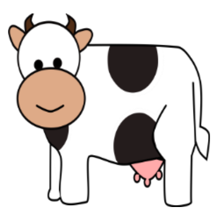
Neutral
GCR
Beam

Ionization

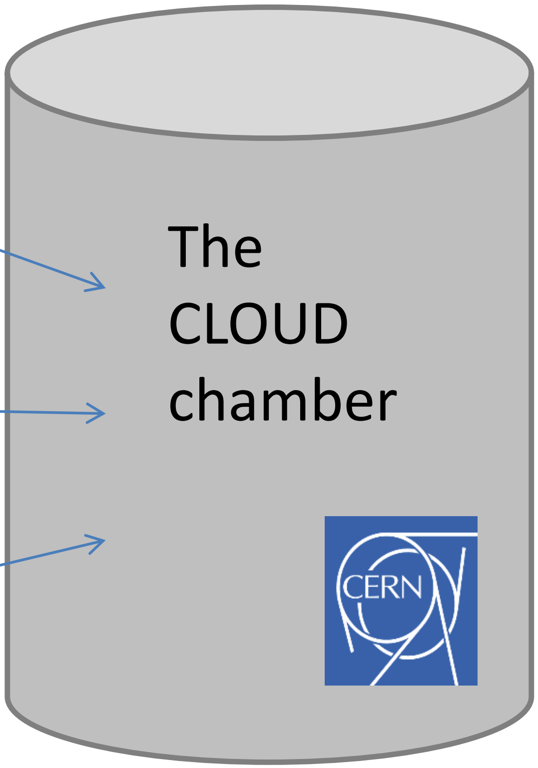
Instrumentation:
particle counters
size spectrometers
mass spectrometers
gas monitors etc.

H_2SO_4

$NO_2 + OH \rightarrow HNO_3$



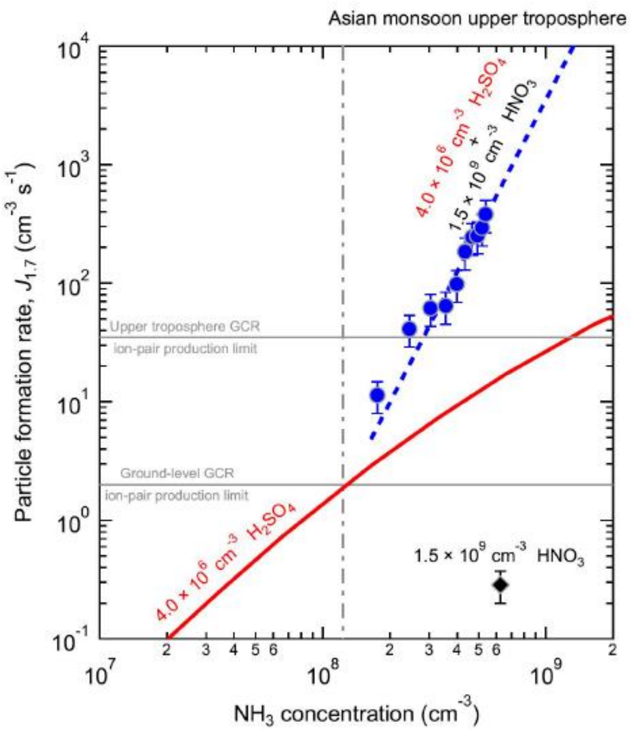
NH_3

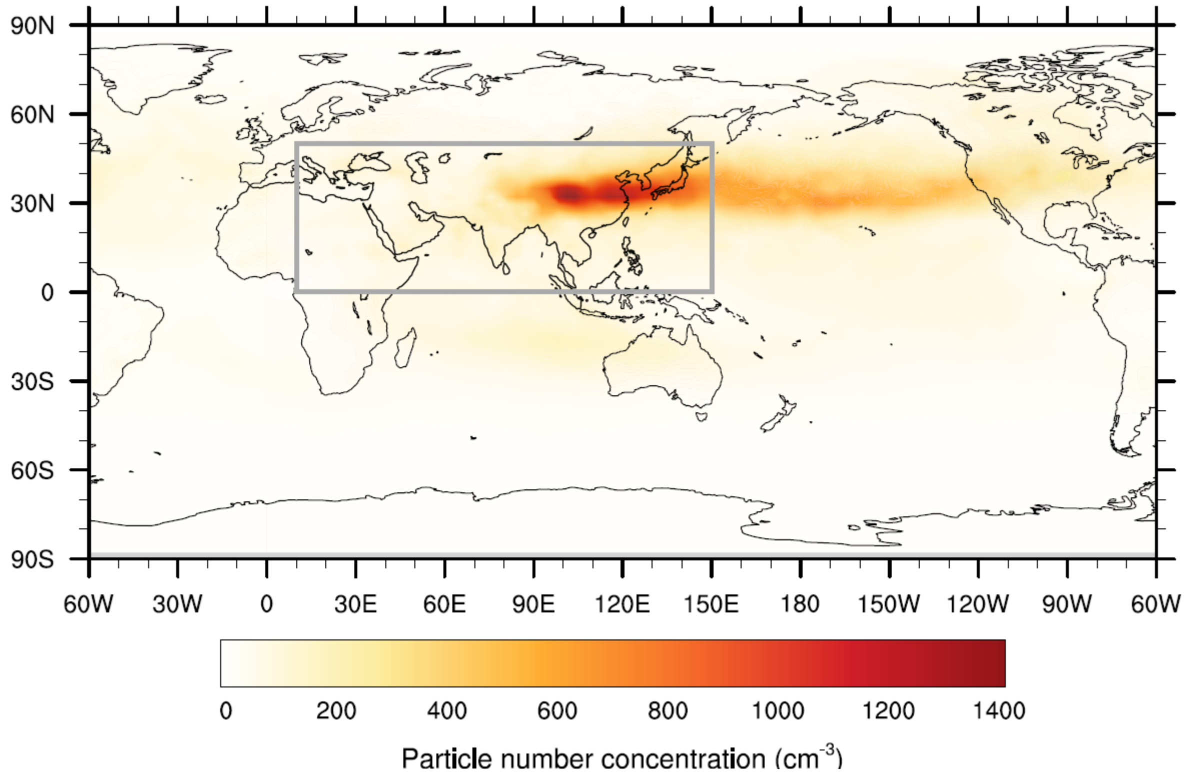


Results

Study of nucleation for nitric acid - ammonia - sulfuric acid system:
Determination of nucleation rates at upper troposphere conditions

Wang et al., Nature, 2022: highly efficient aerosol nucleation in the outflow of East Asian monsoon.
Ammonium nitrate particles also efficient INP for cirrus





CLOUD-DOC

EU Marie Skłodowska Curie Action - Doctoral Network “CLOUD-DOC“ awarded:
Sep 2022 – Aug 2026, 12 PhD students, 2.7 Mio Euro

Research plan: Focus on four scientific areas

- ***Arctic***
- ***Upper Troposphere in the monsoon outflow over Asian megacities and agricultural regions***
- ***Upper troposphere above pristine tropical rain forests***
- ***Southern Ocean/Antarctic***

CLOUD15 experiment in fall 2022

Science goals: Cold conditions.

1. **Pure biogenic nucleation with aged organics**
   Use FLOTUS to age mixtures of biogenic vapours and inject these aged vapours into the CLOUD chamber. Study the chemistry of aged organic vapours and their ability to form and grow new particles

2. **Carbon closure**
   Explain quantitatively the fate of all carbon molecules injected into FLOTUS/CLOUD. Track the oxidative evolution of organic carbon over its entire atmospheric lifetime.

3. **Marine nucleation and growth with methanesulphonic acid (CH_3SO_3H, MSA)**
   Extend our initial CLOUD15T studies of MSA and ammonia new particle formation and growth to characteristic of marine and polar regions between the boundary layer and upper troposphere.

4. **Iodic acid nucleation and growth under upper tropospheric conditions**
   Study new particle formation and growth involving iodine vapours at upper tropospheric conditions: low temperatures (-30°C →-50°C), low vapour concentrations and high ion concentrations.

Beam requests 2022 for CLOUD15 run

CLOUD requests 9 weeks of beam in fall 2022 (Sep-Nov) for the CLOUD15 run (as in previous years).

CLOUD Run Coordinator

- Major component of CLOUD Run Coordinator's work involves dealing with CERN safety requirements for the CLOUD facility and its more-than-50 instruments:
  - electrical safety, chemical safety, flammable and toxic gases, radioactive sources, X ray sources, lasers

- CLOUD Run Coordinator handles all planning & logistics for 50-or-so analysing instruments that are shipped to CERN at the start of each run and then returned at end, in coordination with:
  - CERN shipping and receiving personnel
  - CERN crane operators
  - CERN radiation personnel

- Previous Run Coordinators were CLOUD full-time CERN research staff (Kirkby 2009-2013, Gordon 2014-2017, Manninen 2017-2019)

- Run Coordinator tasks cannot be carried out by small CERN CLOUD technical team (1.7 FTE CERN staff, none is full-time)

- Critical position for CLOUD. Not yet solved

Summary

- CLOUD is eager to restart science runs after LS2 & Covid shut-down.
- Renovation during LS2 lead to greatly improved experimental area.
- CLOUD15T technical run took place in spring 2022.
- CLOUD15 experiment planned for fall 2022.
- Continued achivement scientific results with high impact.
- CLOUD-DOC funding secured through EU MSCA-DN.
- Ambitious experimental programme for 2022-2026.

CLOUD publications (2021-2022):

[4] He, X.-Ch., *et al.* Determination of the collision rate coefficient between charged iodic acid clusters and iodic acid using the appearance time method. *Aerosol Sci. Technol.*, **55**, 231–242 (2021).

[5] He, X.-Ch., *et al.* Role of iodine oxoacids in atmospheric aerosol nucleation. *Science* **371**, 589–595 (2021).

[6] Wang, M. *et al.* Measurement of iodine species and sulfuric acid using bromide chemical ionisation mass spectrometers. *Atmos. Meas. Tech.* **14**, 4187–4202 (2021).

[7] Surdu, M. *et al.* Molecular characterization of ultrafine particles using extractive electrospray time-of-flight mass spectrometry. *Environ. Sci.: Atmos.* **1**, 434–448 (2021).

[8] Xiao, M., *et al.* The driving factors of new particle formation and growth in the polluted boundary layer. *Atmos. Chem. Phys.* **21**, 14275–14291 (2021).

[9] Caudillo, L., *et al.* Chemical composition of nanoparticles from α-pinene nucleation and the influence of isoprene and relative humidity at low temperature. *Atmos. Chem. Phys.* **21**, 17099–17114 (2021).

[10] Amaladhasan, D. A., *et al.* Modelling the gas–particle partitioning and water uptake of isoprene-derived secondary organic aerosol at high and low relative humidity. *Atmos. Chem. Phys.* **22**, 215–244 (2022).

[11] Wang, M., *et al.* Synergistic HNO_3-H_2SO_4-NH_3 upper tropospheric particle formation. *Nature* **605**, 483–489 (2022).

[12] Marten, R. *et al.* Survival of newly formed particles in haze conditions. *Environ. Sci.: Atmos.* **2**, 491–499 (2022).

CLOUD manuscripts currently under review:

[13] Xiao, M., *et al.* Second-generation chemistry drives secondary aerosol formation from aromatic hydrocarbons. *Science Advances*, submitted (2022).

[14] Shen, J., *et al.* High methanesulfonic acid production in the OH-initiated oxidation of dimethylsulfide at low temperatures. *Environ. Sci. Technol.*, submitted (2022).

[15] Finkenzeller, H., *et al.* The gas-phase source mechanism of iodic acid as a driver of aerosol formation. *Nature Chem.*, submitted (2022).

cloud

Funding

- CLOUD-DOC, EU Marie Skłodowska Curie Action - Doctoral Network, Sep 2022 – Aug 2026, 12 PhD students, 2.7 Mio Euro

- Regular support by national funding, e.g., by German BMBF, Swiss National Science Foundation, the Academy of Finland Center of Excellence program, other national funding agencies…

- Regular meetings of CLOUD Financial Review Committee at CERN.
  CERN support is gratefully acknowledged.

MARIE CURIE ACTIONS

GEFÖRDERT VOM
Bundesministerium
für Bildung
und Forschung

CERN

Acknowledgements

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