21 June 2016

Status and plans of the CLOUD experiment

Joachim Curtius

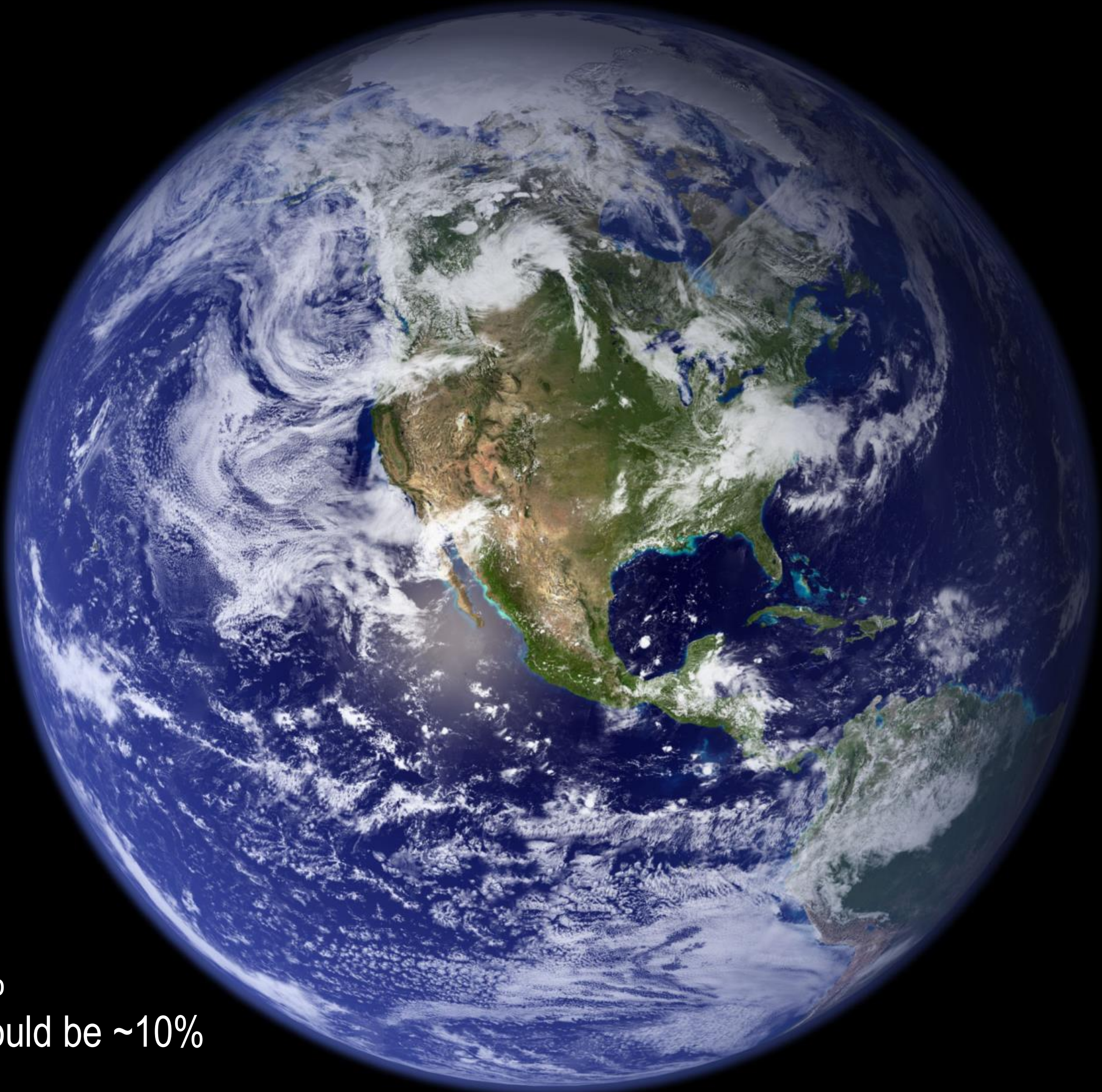
Institute for Atmospheric and Environmental Sciences
Goethe University Frankfurt

On behalf of the CLOUD collaboration:

Austria:	University of Innsbruck
	University of Vienna
Finland:	Finnish Meteorological Institute
	Helsinki Institute of Physics
	University of Eastern Finland
	University of Helsinki
Germany:	Johann Wolfgang Goethe University Frankfurt
	Karlsruhe Institute of Technology
	Leibniz Institute for Tropospheric Research
Portugal:	University of Beira Interior
	University of Lisbon
Russia:	Lebedev Physical Institute
Sweden:	University of Stockholm
Switzerland:	CERN
	Paul Scherrer Institute
	TOFWERK
United Kingdom:	University of Manchester
	University of Leeds
USA:	Aerodyne Research
	California Institute of Technology
	Carnegie Mellon University

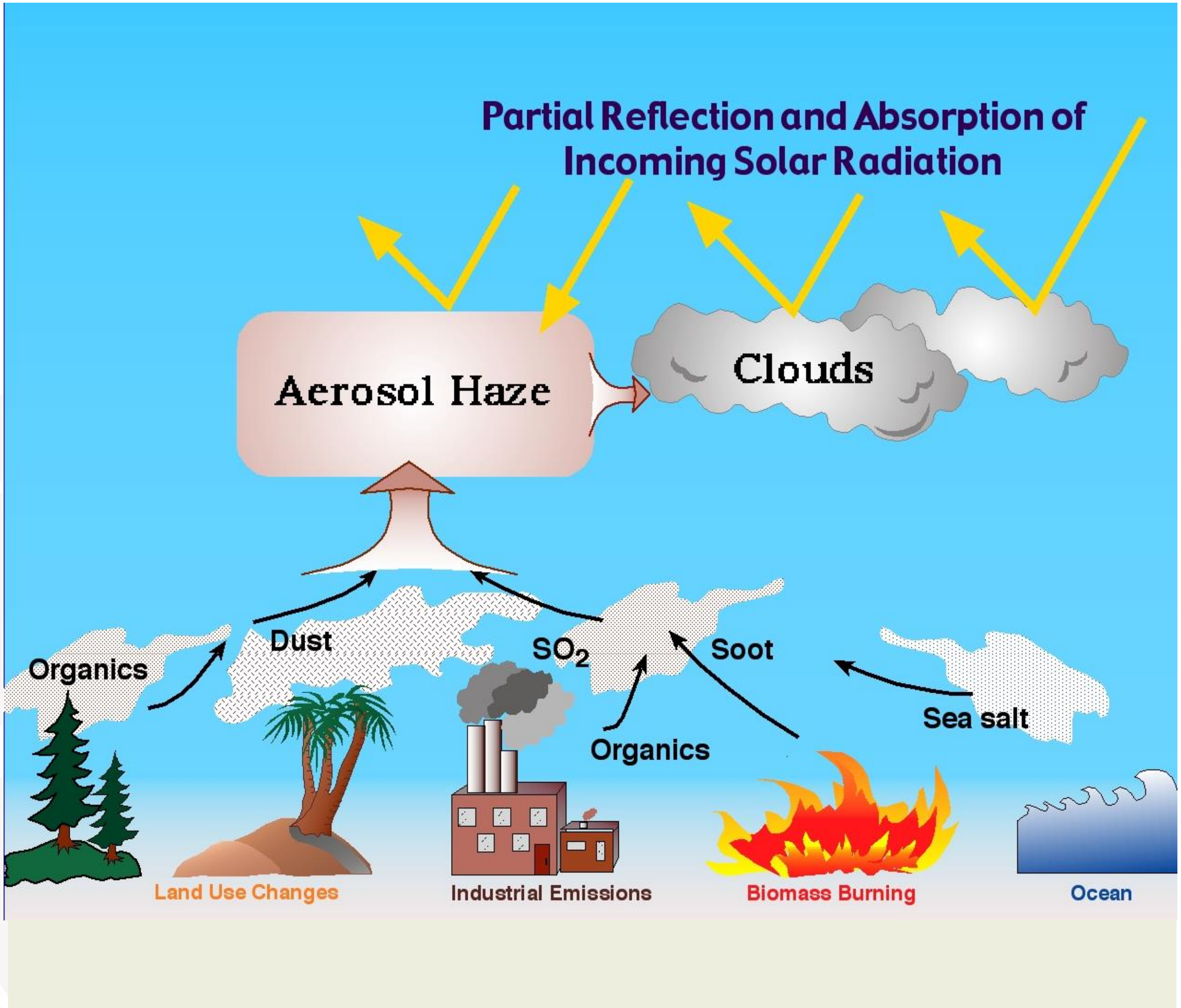
CLOUD data workshop, Hyytiälä, FIN, Feb 2016.

Clouds



- cover more than 60% of the Earth‘s surface on average
- average albedo ~30%
  without clouds this would be ~10%
- strong effect on radiation budget: “more clouds = cooler climate”

Aerosols, clouds and climate



Direct effect

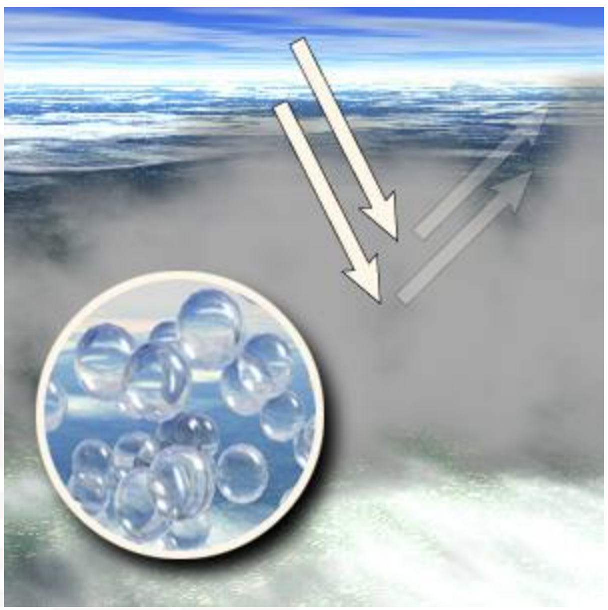
Scattering und absorption of incoming sunlight by aerosol particles.

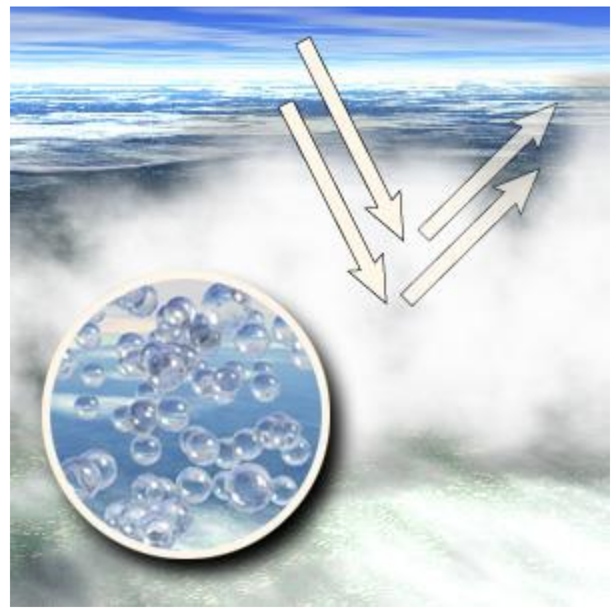
Indirect effect

Changes of cloud albedo and cloud lifetime through additional anthropogenic aerosol particles.

Cloud adjustments due to aerosols

More aerosol particles → more Cloud Condensation Nuclei (CCN)
→ more (but smaller) cloud droplets → more sunlight reflected to space
→ surface cooling





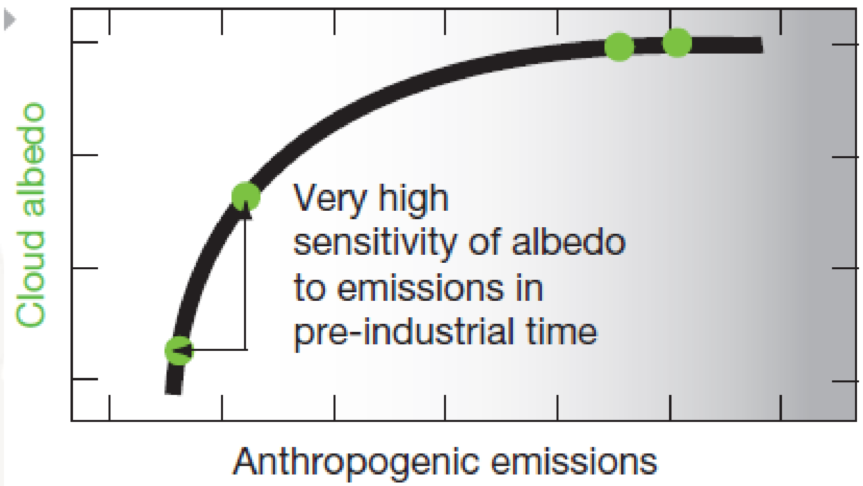
More but smaller droplets:

Whiter clouds

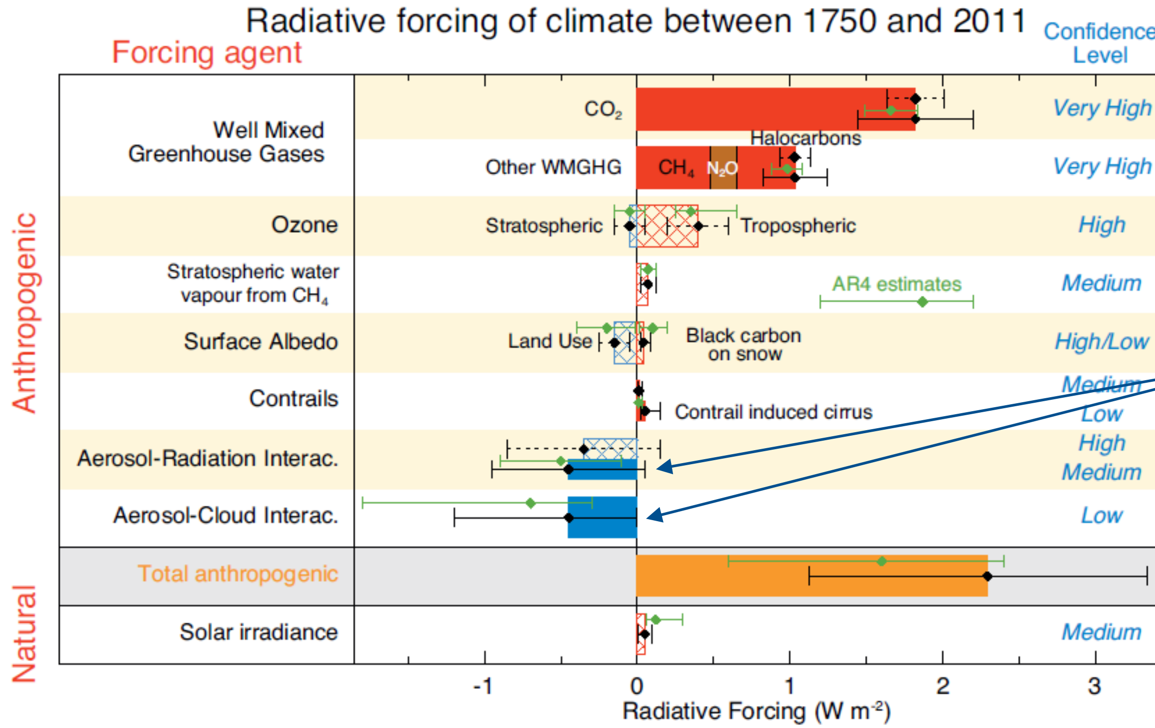
How many aerosol particles, CCN and cloud droplets were present in pre-industrial times?

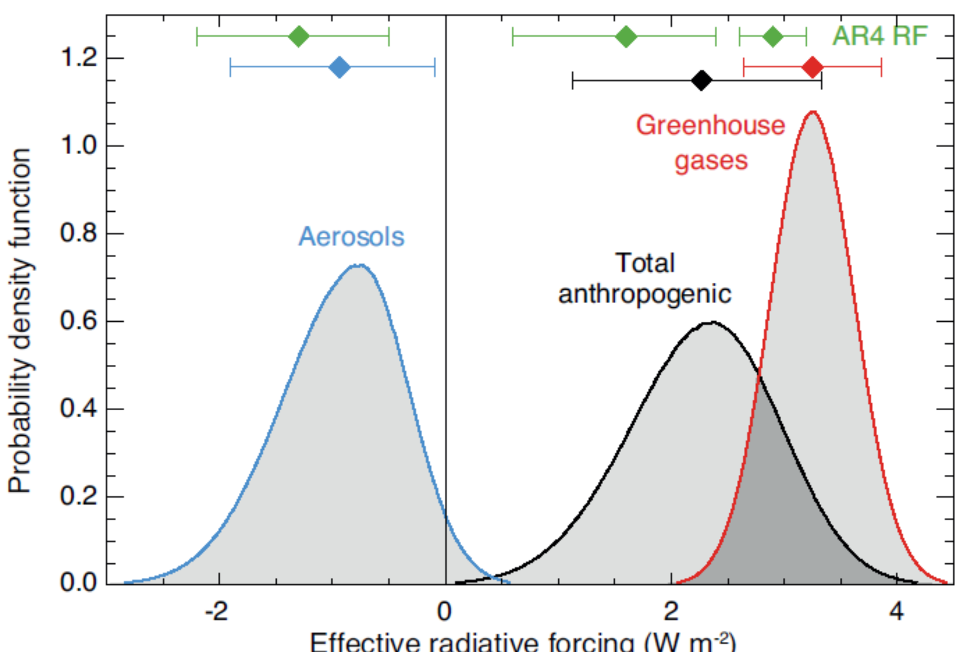
Large contribution of natural aerosols to uncertainty in indirect forcing

Carslaw et al., Nature, 2013



"The results point to the importance of understanding pristine pre-industrial-like environments, with natural aerosols only"





Radiative forcing due to anthropogenic aerosols:

- Net cooling effects
- Largest uncertainty

[IPCC,2013]

Nucleation of terpene oxidation products without sulfuric acid

biogenic particle production

Blue haze

biogenic particle formation

LETTER

Ion-induced nucleation of pure biogenic pa

Jasper Kirkby[1,2], Jonathan Duplissy[3,4], Kamalika Sengupta[5], Carla Frege[6], Hamish Gordon[2], Christina Willi
Martin Heinritzi[1,7], Mario Simon[1], Chao Yan[3], João Almeida[1,2], Jasmin Tröstl[6], Tuomo Nieminen[3,4], Ismael K
Marti
Aless
Juha
Vladi
Arna
Nina
Yuri S
Ernes
Neil M

LE

The
initi

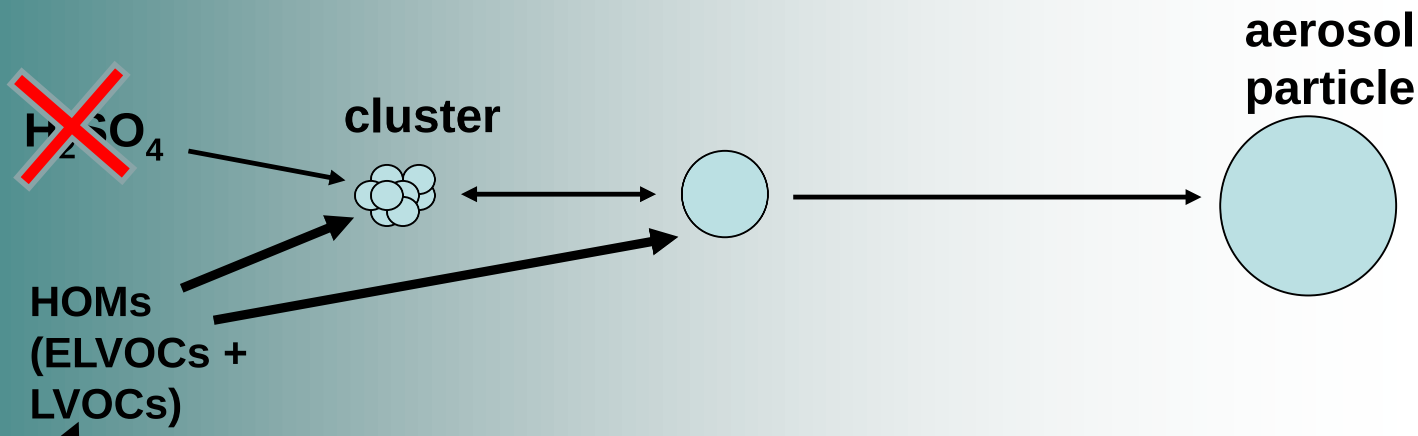
Jasmin Tr
Federico
Jonathan
Sophia Br
Martin Gy
Helmi Ke
Markus L
Pasi Miet
Mikko Si
Ernest W
Josef Don
Urs Balte

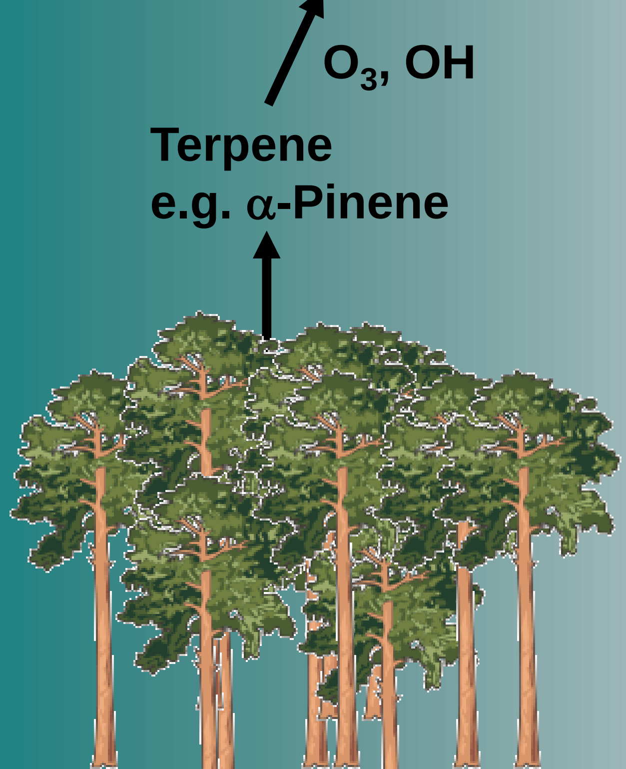
oposphere: A question of chemistry and timing

F. Bianchi,[1,2,3*] J. Tröstl,[1] H. Junninen,[3] C. Frege,[1] S. Henne,[4] C.R. Hoyle,[1,5] U. Molteni,[1] E. Herrmann,[1] A. Adamov,[3] N. Bukowiecki,[1] X. Chen,[3] J. Duplissy,[3,6] M. Gysel,[1] M. Hutterli,[7] J. Kangasluoma,[3] J. Kontkanen,[3] A. Kürten,[8] H. E. Manninen,[3] S. Münch,[8] O. Peräkylä,[3] T. Petäjä,[3] L. Rondo,[8] C. Williamson,[8†] E. Weingartner,[1‡] J. Curtius,[8] D. R. Worsnop,[3,9] M. Kulmala,[3] J. Dommen,[1] U. Baltensperger[1*]

Coordinated release by *Nature* and *Science* on 26 May 2016

Role of Highly Oxygenated Multifunctional Organic Molecules for nucleation and growth of particles



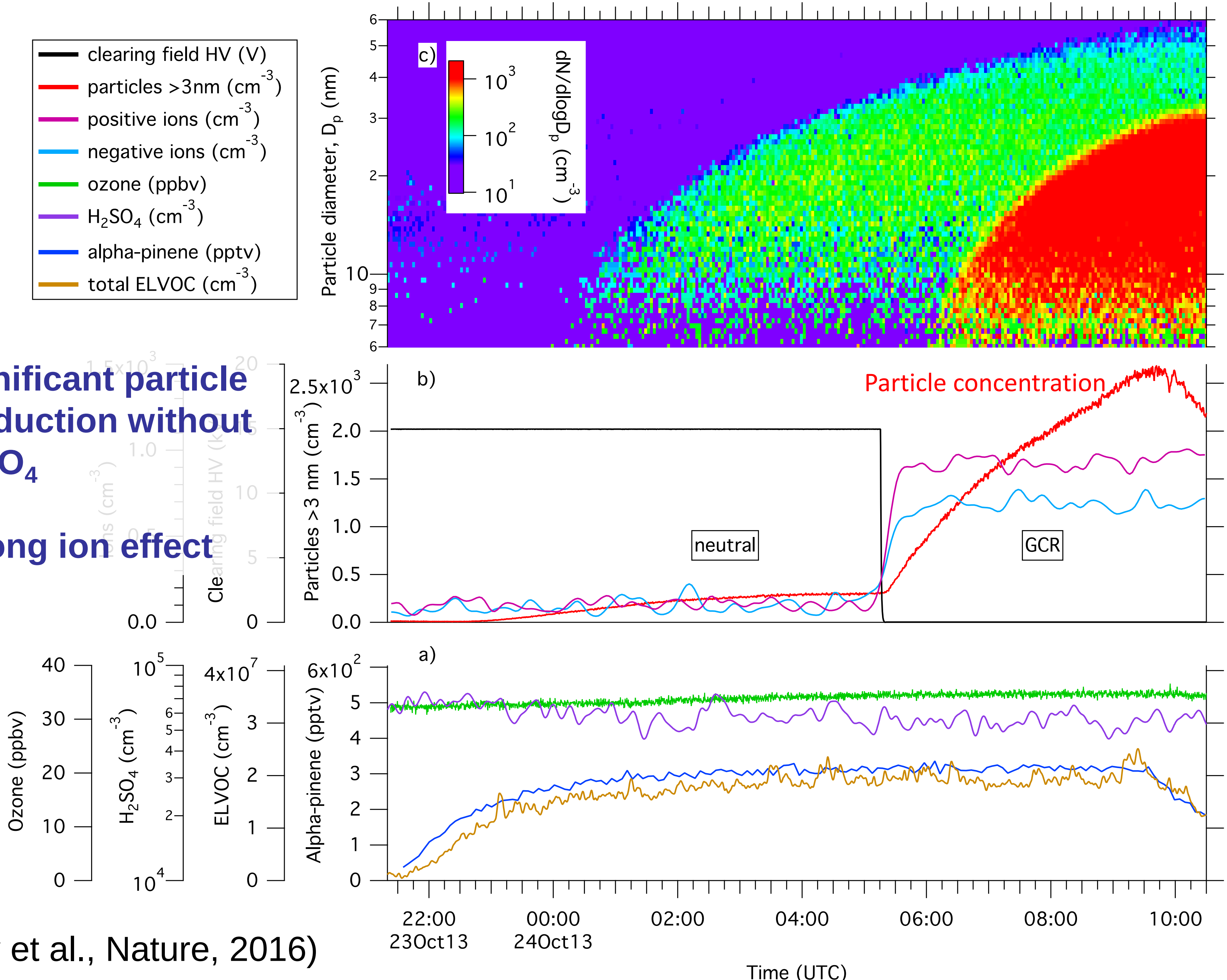


α-Pinene: most important monoterpene ($C_{10}H_{16}$)

HOMs = Highly Oxygenated Multifunctional organic molecules
ELVOCs = extremely low volatility organic compounds
LVOCs = low volatility organic compounds

In how far do HOMs contribute to atmospheric nucleation and growth?

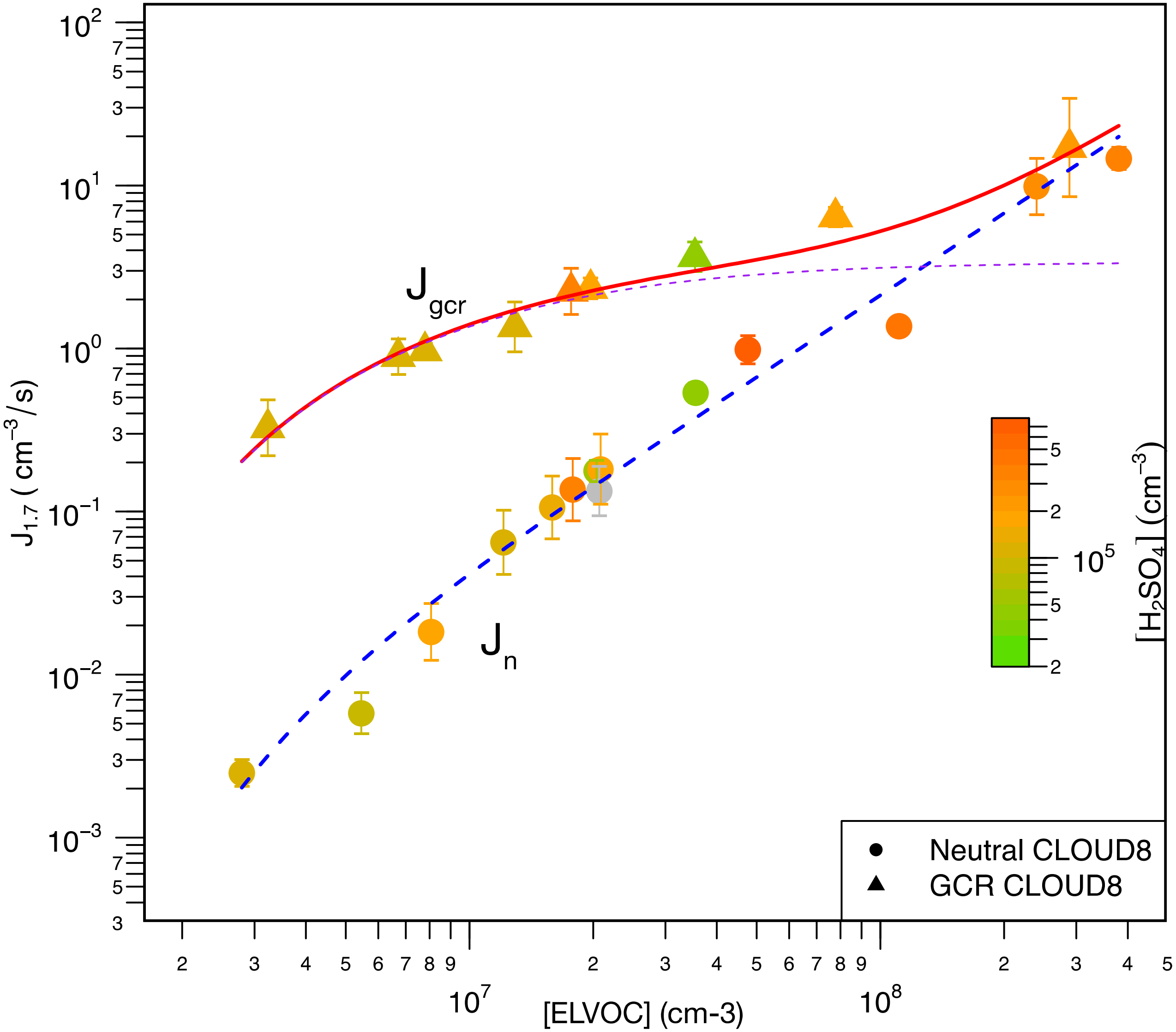
Particle formation from purely biogenic sources



- **Significant particle production without H_2SO_4**
- **Strong ion effect**

(Kirkby et al., Nature, 2016)

- **Substantial nucleation of α-pinene oxidation products without sulfuric acid**
- **Strong ion effect**

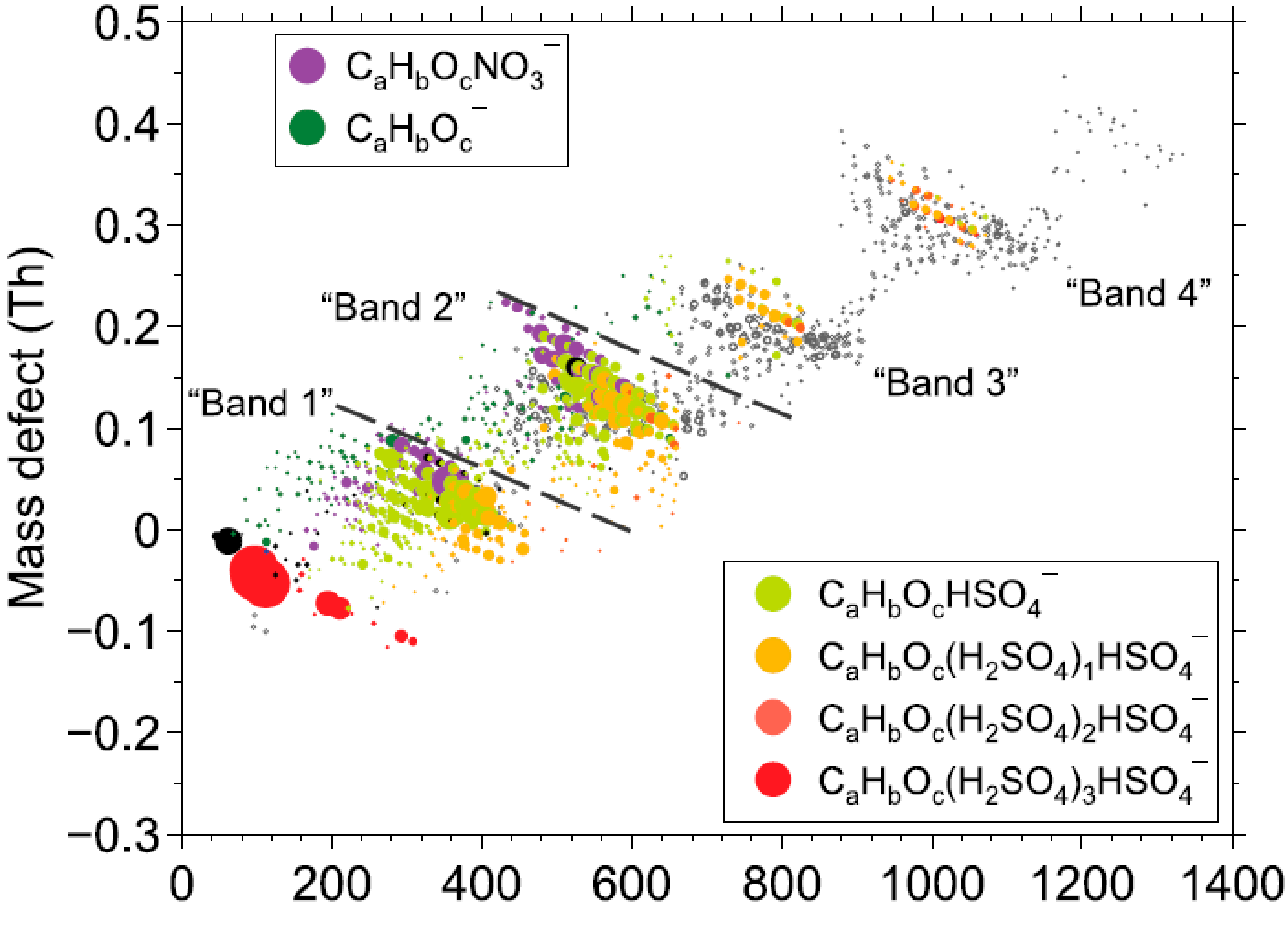


Important mechanism for pre-industrial atmosphere when H_2SO_4 was low.

Effect of temperature, NO_x, other terpenes, RH, etc. needs to be studied…

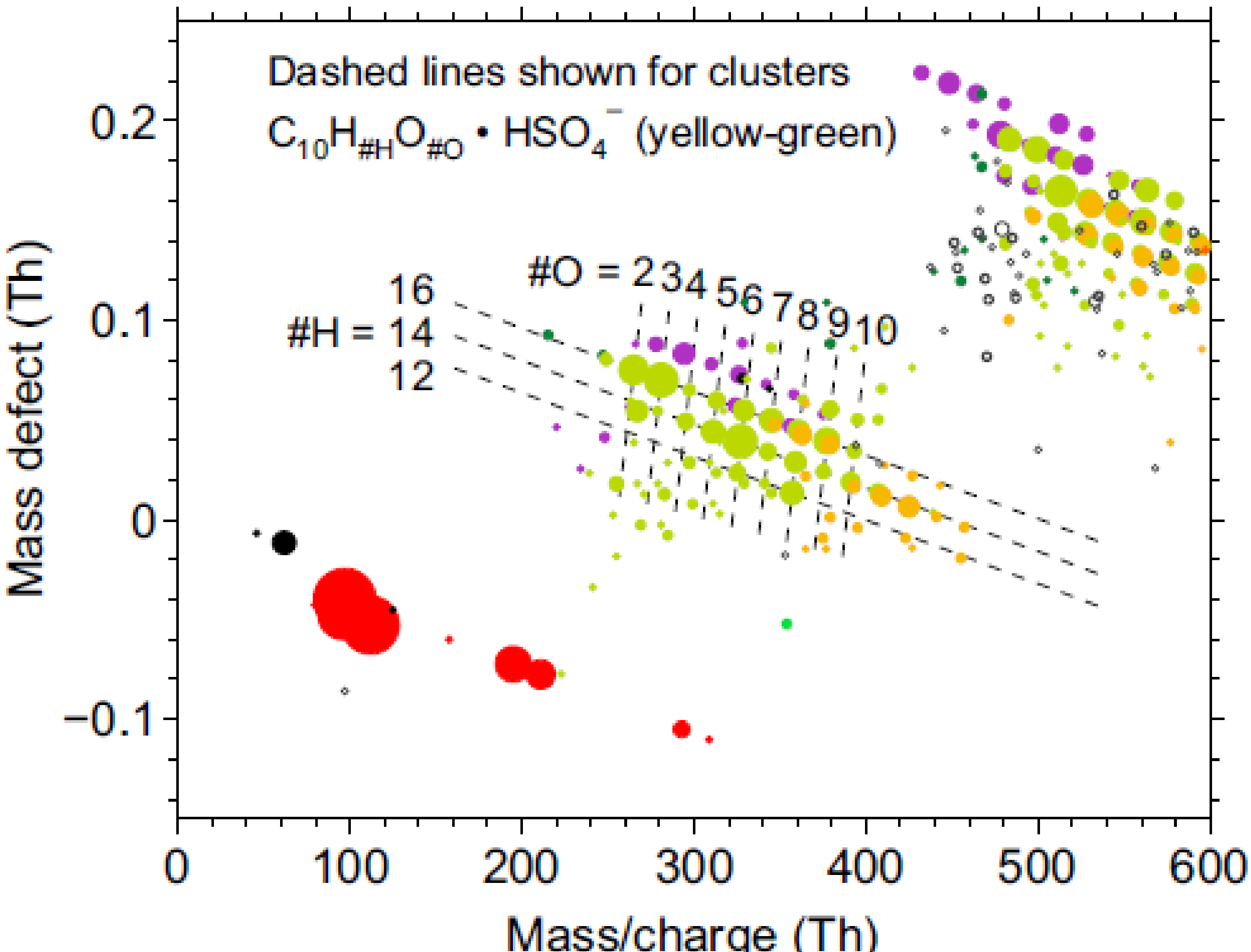
(Kirkby et al., Nature, 2016)

Molecular composition of cluster ions : Hundreds of oxidation products



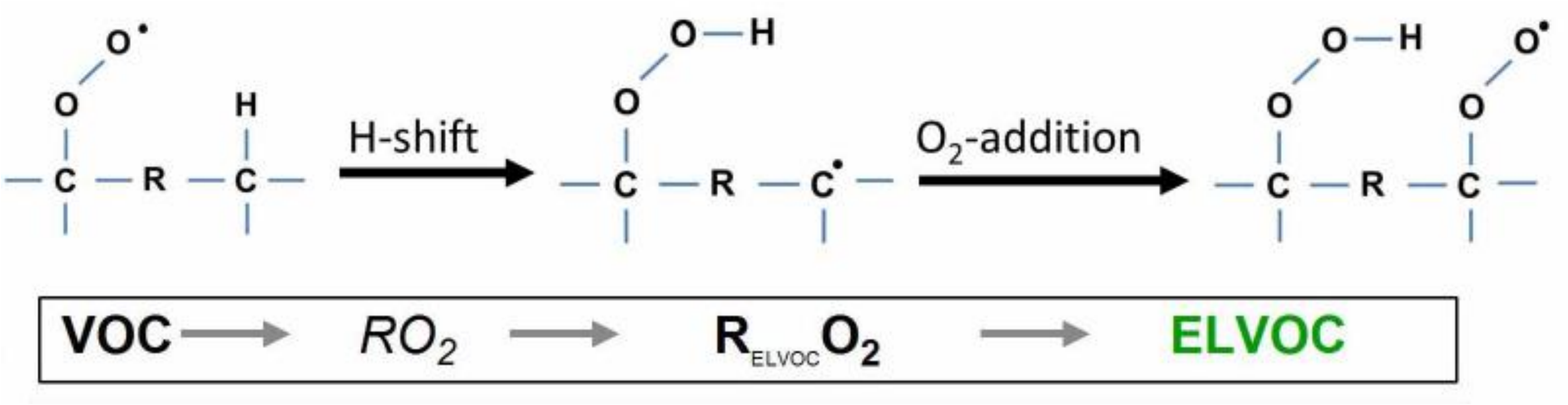
[Schobesberger et al., PNAS 2013]

Highly oxygenated products form within minutes: ELVOCs



[Schobesberger et al., PNAS, 2013]

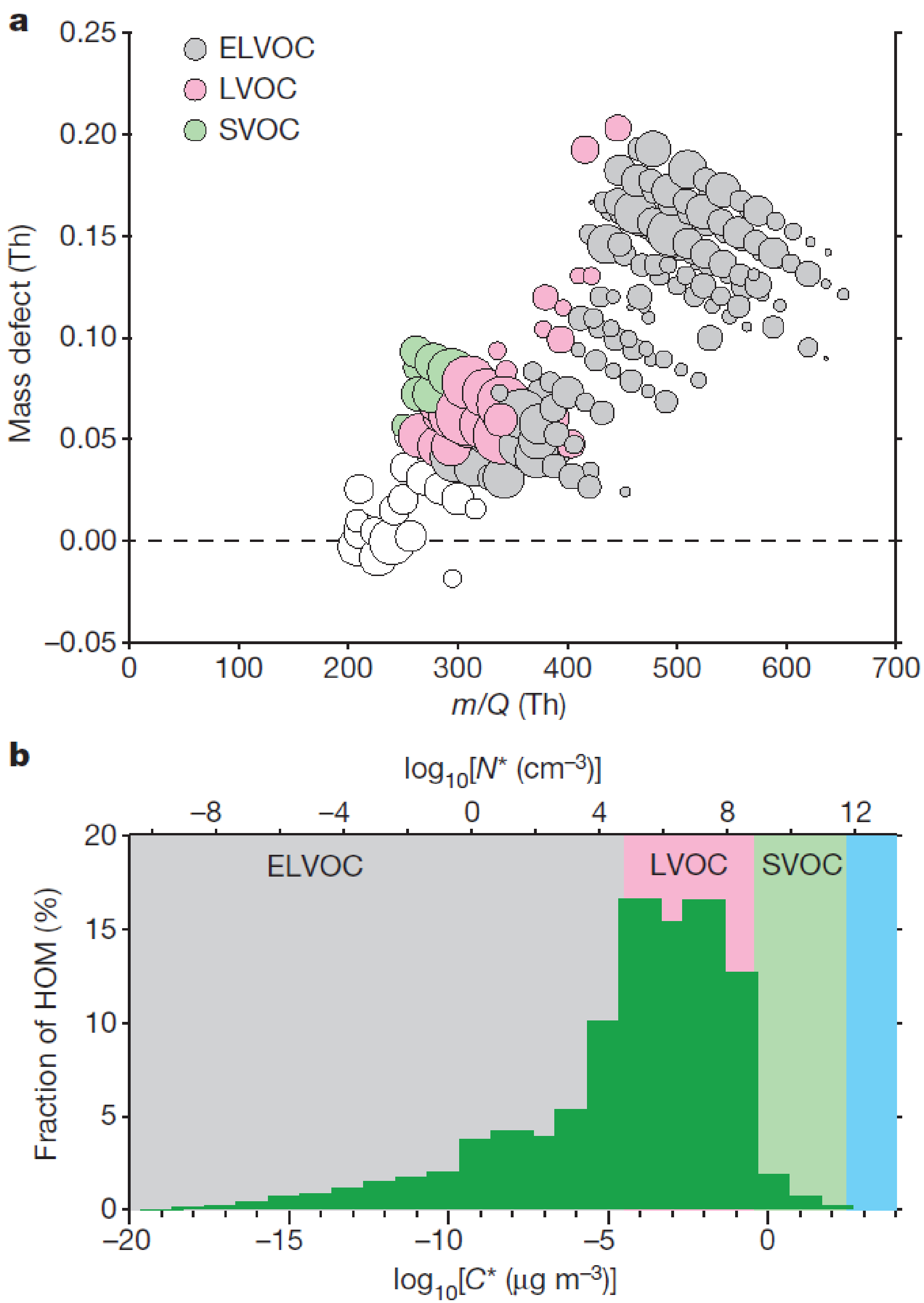
Fast and efficient formation of ELVOCs from "autoxidation" (Crounse et al, JPCL, 2013; Ehn et al., Nature, 2014)



ELVOCs form within minutes…

particle growth

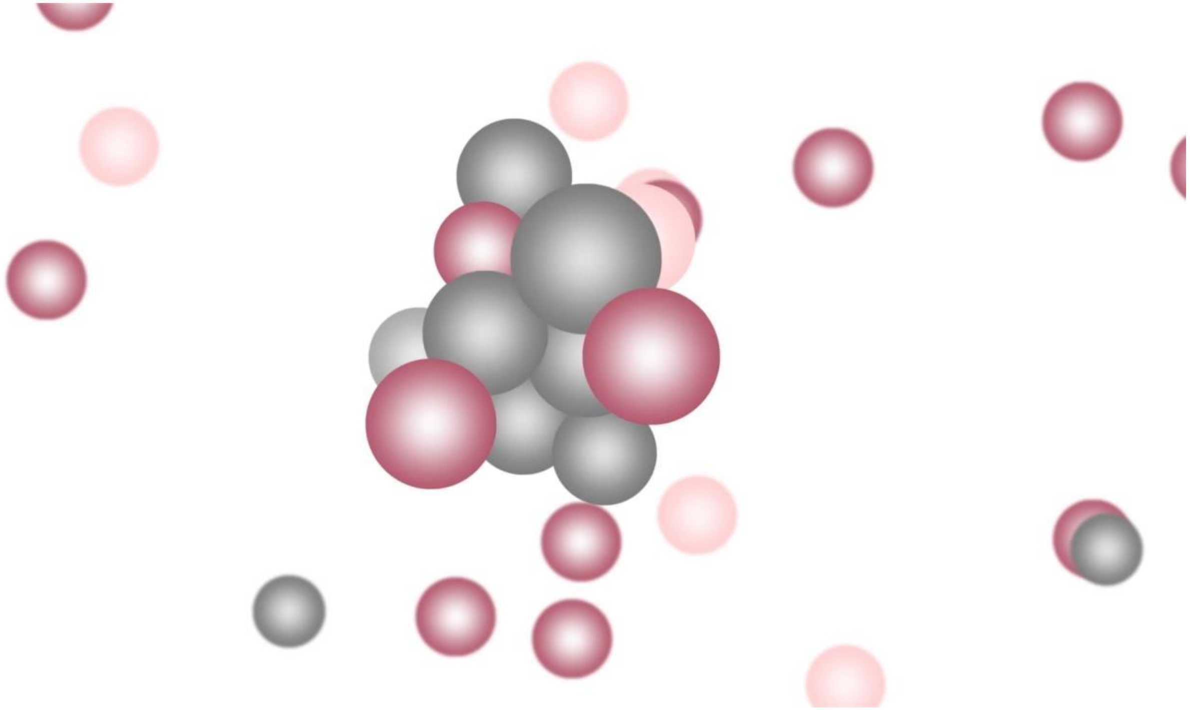
Growth



- ELVOCs (36% of HOM signal) contribute to nucleation and growth from beginning
- LVOCs contribute to growth at >2nm size.

[Tröstl et al., Nature, 2016]

Growth

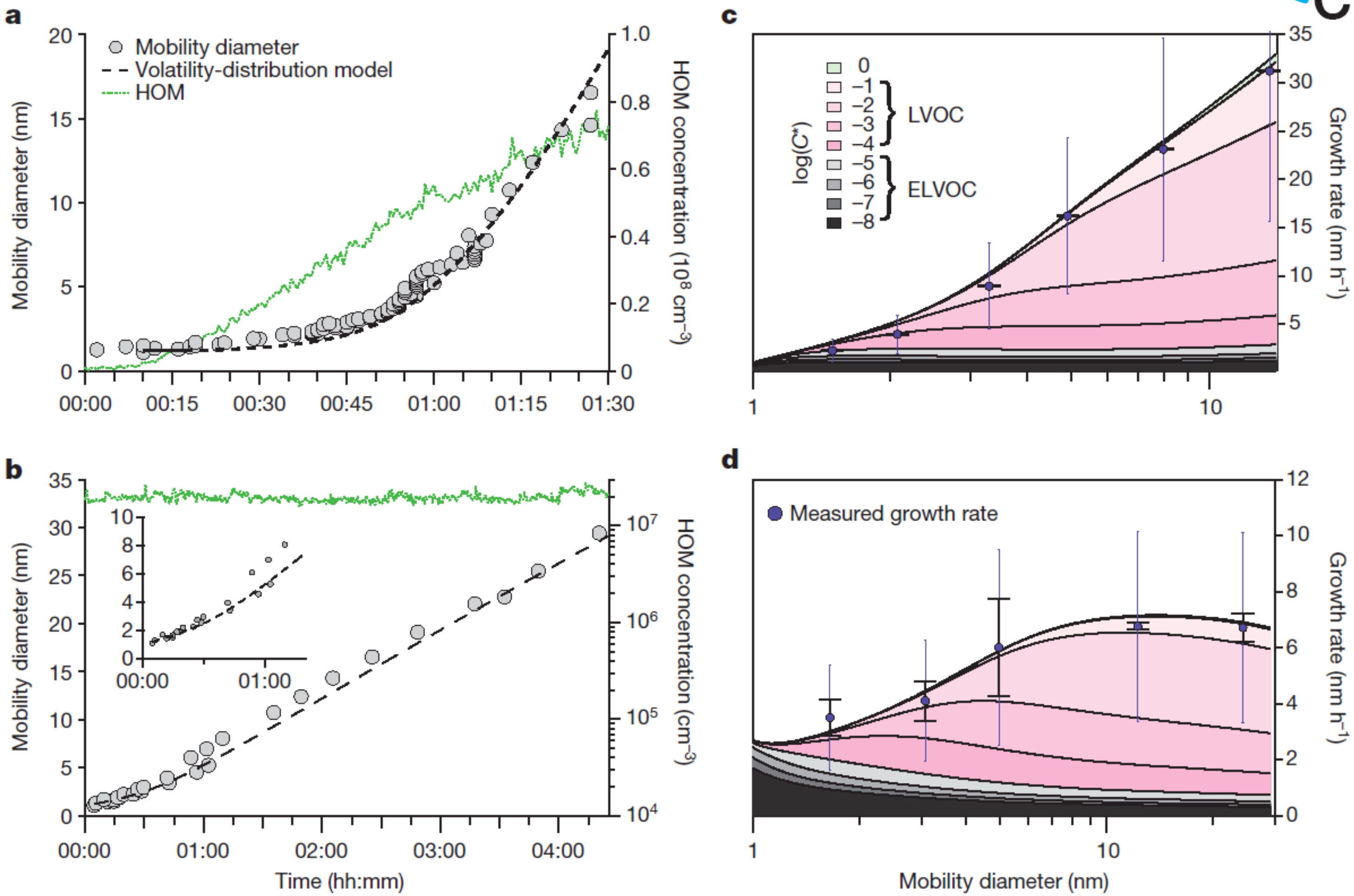


- ELVOCs (36% of HOM signal)
  contribute to nucleation and growth from the beginning

- LVOCs contribute to growth starting at ~2 nm.

[Carnegie Mellon University 2016]

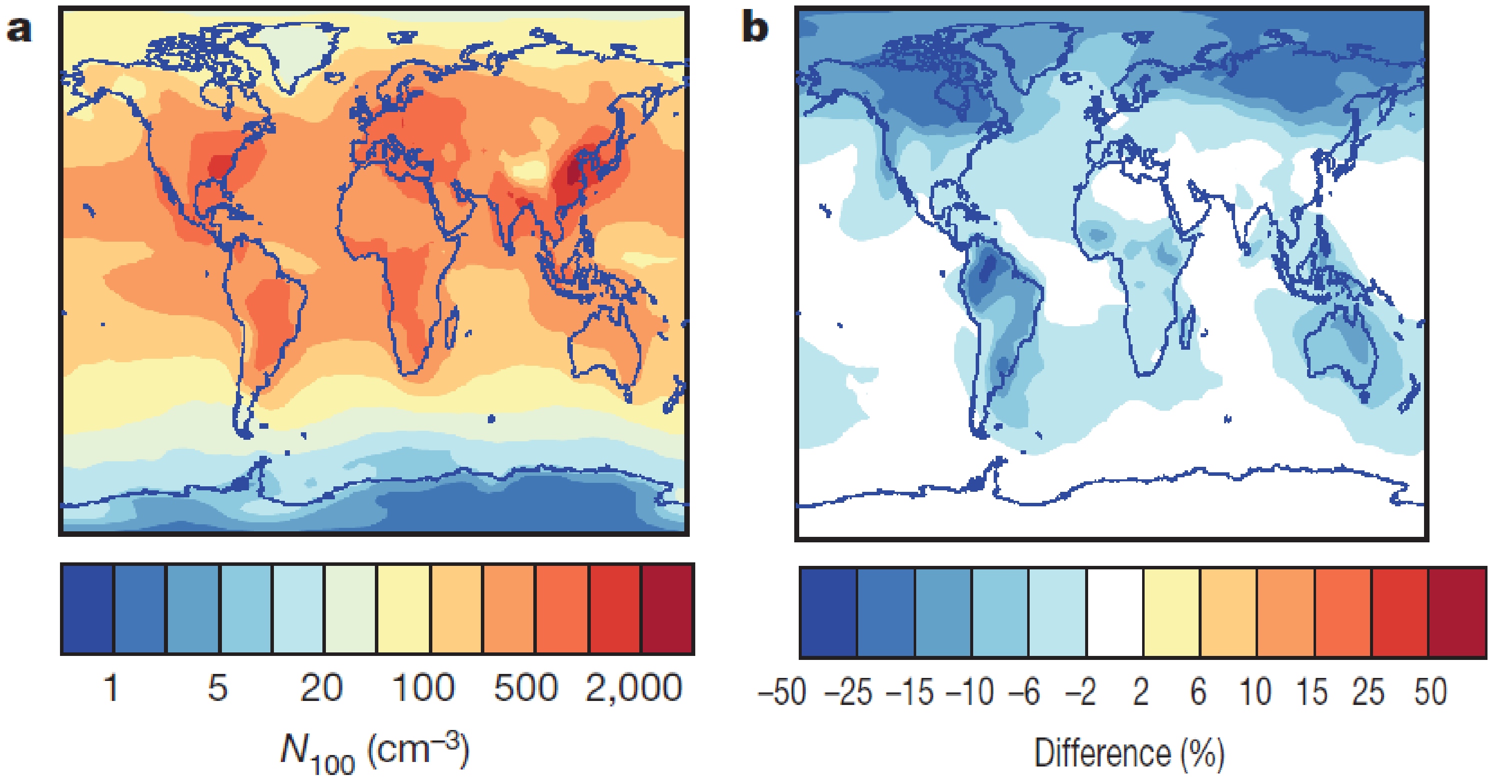
Growth from ELVOCs and LVOC



[Tröstl et al., 2016]

- Growth accelerates as subsequently more LVOCs can condense.

Growth

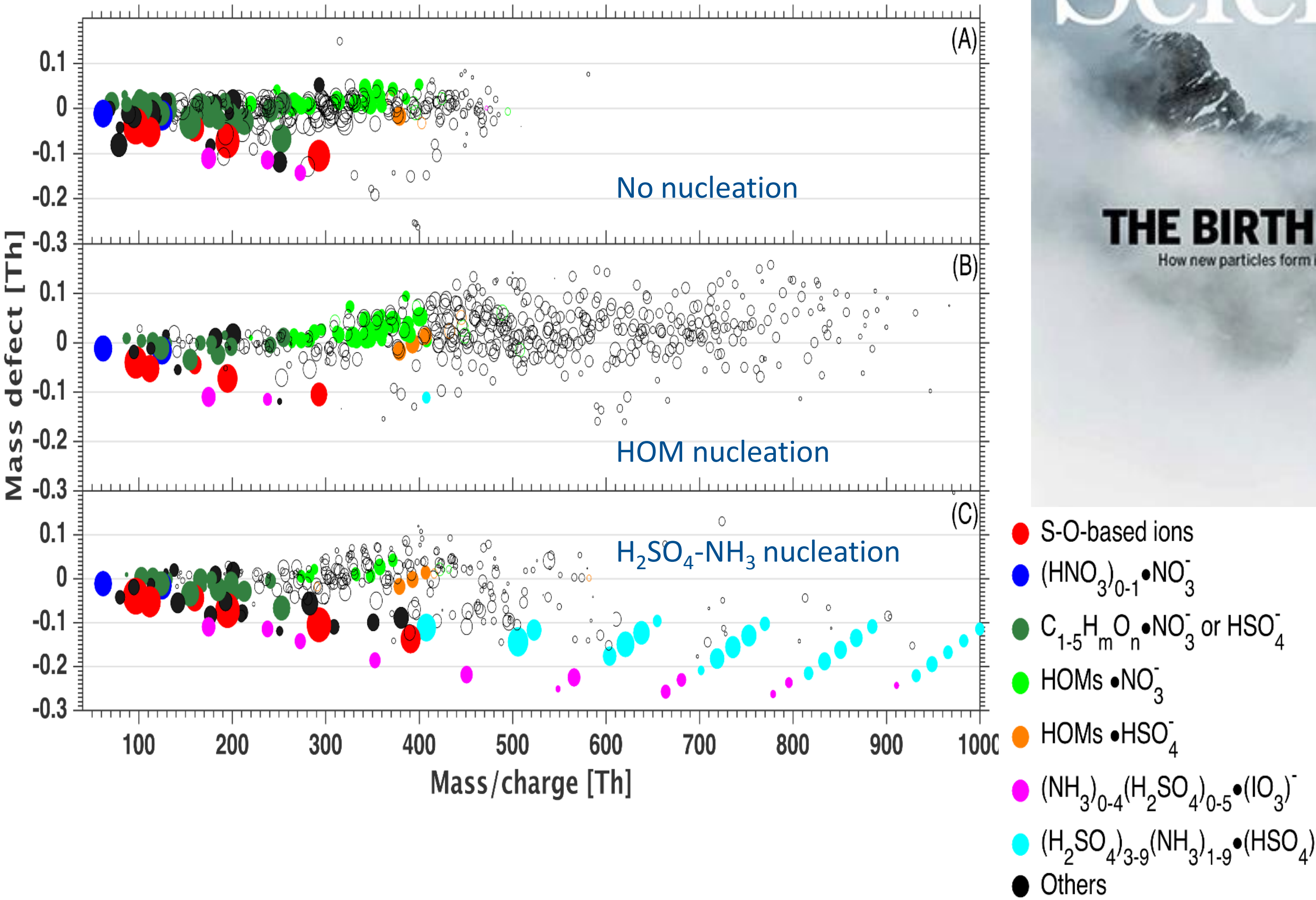


a) Modelled global CCN-concentrations at cloud base (growth by HOMs and H_2SO_4, im 1.7-3 nm),
b) Difference when growth by HOMs is neglected (H_2SO_4 only)

→ Initial growth by HOMs has significant impact on CCN concentration!

[Tröstl et al., Nature, 2016]

Atmospheric observations

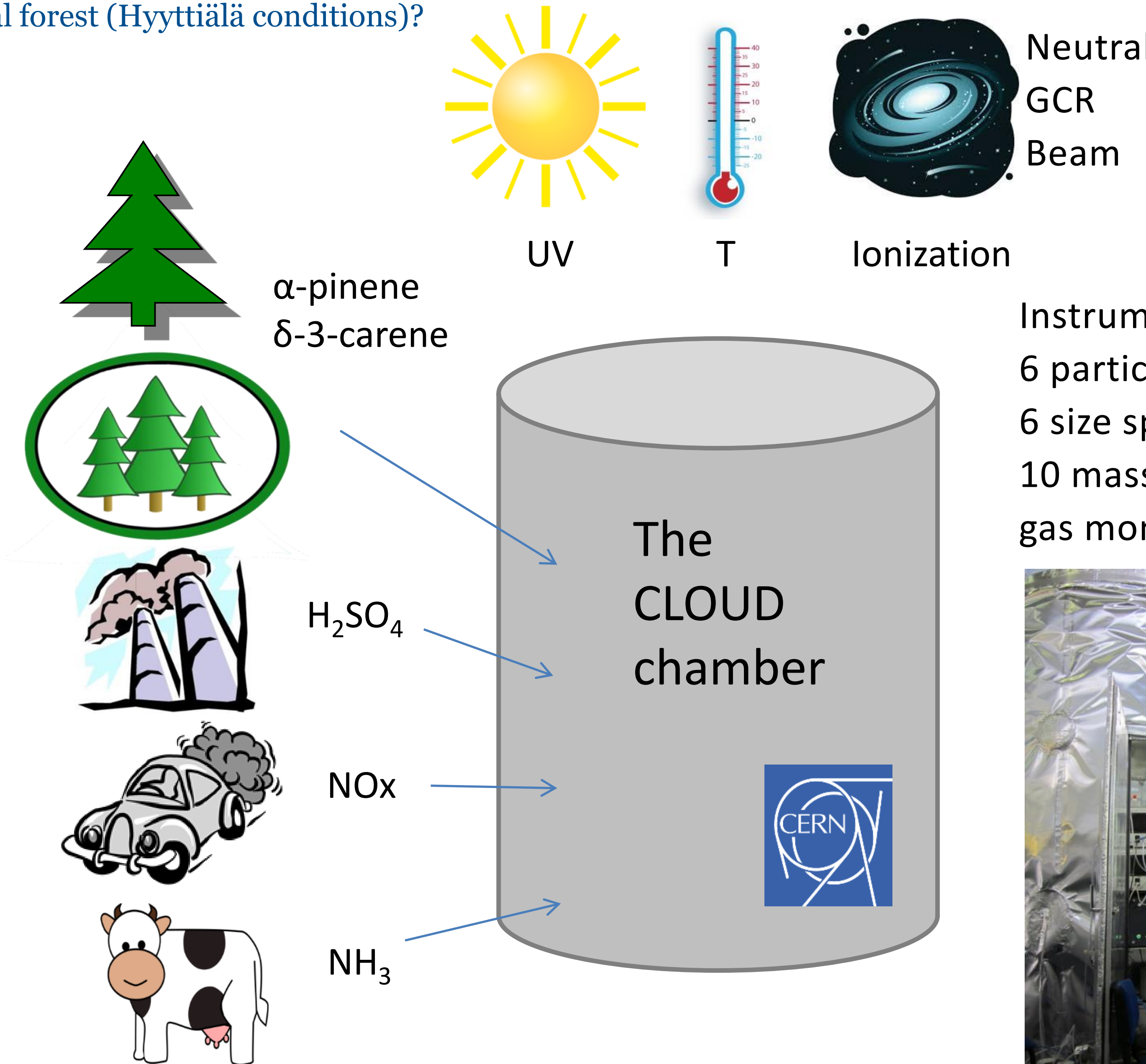


- ➢ HOM nucleation and growth also observed in free troposphere
- ➢ Sulfuric acid-ammonia nucleation more frequent

[Bianchi et al., Science, 2016]

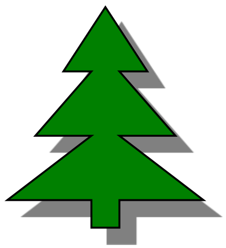
CLOUD-10 Experiment (Sep-Dec 2015)

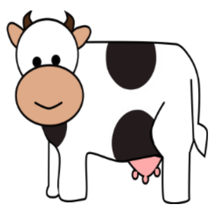
Can we replicate conditions
in boreal forest (Hyytiälä conditions)?



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

CLOUD10 results





- Complex interplay of H_2SO_4 , NH_3 and biogenic nucleators!
- Composition of nano-particles (2-10 nm)
- Role of NO_x
- Role of temperature
- Correlation with groups of ELVOCs

CLOUD11 plans for fall 2016

CLOUD11 experiment is scheduled for 27 Sep – 21 Nov 2016
(final week using cosmic rays, no East Area beams).

Aims:

1. *Anthropogenic* aerosol particle nucleation and growth.
   Toluene, trimethylbenzene and naphthalene.
   Nucleation and growth under polluted urban conditions.

2. *Pure biogenic* aerosol particle nucleation and growth
   - Interplay of *isoprene* (C_5H_8), α*-pinene* ($C_{10}H_{16}$), *and* β*-caryophyllene* ($C_{15}H_{24}$).
   - Influence of NO_x, oxidant concentrations (O_3 and OH), humidity and temperature.
   - Various ion conditions (neutral, GCR and beam).
   - Assess tmospheric importance of pure biogenic nucleation, using the global aerosol model GLOMAP.

Beam requested during CLOUD11.

Requests

Office space:

- CLOUD has typically 30 persons present during experiments

→ *Request for a permanent open office/meeting room (~50 m^2) for CLOUD close to the T11 experimental zone.*

Personnel:

- CLOUD is supported by CERN with 1.2 FTE technical support (0.2 increase).

- In addition to the technical support, CLOUD needs at least 2 fellows in the CERN team:

  a) a research fellow to coordinate the analysing instruments for each campaign and related tasks.

  b) a technical fellow to take responsibility for the CLOUD DAQ and control systems.

CLOUD request that the CERN Fellows and Associates Committee will agree to consider young researchers with an atmospheric research background as candidates for a CERN research fellowship.

CLOUD request to be assigned a technical fellow's position so that a suitable DAQ expert can be hired.

CLOUD MEETINGS IN 2015-2016

CLOUD7–9 data workshop, Paul Scherrer Institute, 2–6 Feb 2015. Analysis of data from CLOUD7–9.

CLOUD-TRAIN summer school, Cacais, Portugal, 7–13 Jun 2015.

CLOUD collaboration meeting, CERN, 21–23 Oct 2015. incl. FRC4.

CLOUD10 data workshop, Hyytiälä Forestry Field Station, Finland, 1–4 Feb 2016.

CLOUD-TRAIN final conference, Königstein, Germany, 14–17 Jun 2016. incl. 20 invited external experts.

Honors

Andreas Kürten from U Frankfurt received the **Smoluchowski Award** at the European Aerosol Conference in 2015 in recognition of his 2014 PNAS paper on CLOUD results for neutral amine-sulfuric acid clusters. It is the most prestigeous award for early career researchers in aerosol science.

CLOUD publications 2015-2016:

Nucleation and growth:

1. Bianchi, F, et al.: New particle formation in the free troposphere: A question of chemistry and timing, Science, doi: 10.1126/science.aad5456, 2016.
2. Duplissy, J, et al.: Effect of ions on sulfuric acid-water binary particle formation: 2. Experimental data and comparison with QC-normalized classical nucleation theory, *J. Geophys. Res.- Atmos.*, 121, 1752–1775, doi: 10.1002/2015JD023539, 2016.
3. Ehrhart, S, et al.: Comparison of the SAWNUC model with CLOUD measurements of sulphuric acid-water nucleation, *JGR.- Atmos.*, in press, 2016.
4. Heinritzi, M, et al.: Characterization of the mass dependent transmission efficiency of a CIMS, *Atmos. Meas. Tech*., 9, 1449–1460, doi:10.5194/amt-9-1449-2016, 2016.
5. Kirkby, J, et al.: Ion-induced nucleation of pure biogenic particles, *Nature*, 533, doi 10.1038/nature17953, 2016.
6. Kim, J., et al.: Hygroscopicity of nanoparticles produced from homogeneous nucleation in the CLOUD experiments, *Atmos. Chem. Phys.* 16, 293–304, doi:10.5194/acp-16-293-2016, 2016.
7. Kürten, A, et al.: On the derivation of particle nucleation rates from experimental formation rates, *Atmos. Chem Phys.*, 15, 4063–4075, doi: 10.5194/acp-15-4063-2015, 2015.
8. Kürten, A, et al.:: Thermodynamics of the formation of sulfuric acid dimers in the binary (H_2SO_4–H_2O) and ternary (H_2SO_4–H_2O–NH_3) system, *Atmos. Chem. Phys.*, 15, 10701–10721, 10.5194/acp-15-10701-2015, 2015.
9. Kürten, A, et al.: Experimental particle formation rates spanning tropospheric conditions of sulfuric acid, ammonia, ions, and temperature, accepted by *J. Geophys. Res. – Atmos.*, 2016.
10. Lehtipalo, K, et al.: The effect of acid-base clustering on the growth of atmospheric nano-particles, *Nature Communications*, 7, 11594, doi:10.1038/ncomms11594, 2016.
11. Praplan, A, et al.: Elemental composition and clustering behaviour of –pinene oxidation products for different oxidation conditions, *Atmos. Chem. Phys.,* 15, 4145–4159, doi: 10.5194/acp-15-4145-2015, 2015.
12. Rondo, L, et al.: Effect of dimethylamine on the gas phase sulfuric acid concentration measured by Chemical Ionization Mass Spectrometry (CIMS), *J. Geophys. Res. Atmos.,* doi:10.1002/2015JD023868, 2016.
13. Simon, M, et al.: Detection of dimethylamine in the low pptv range using nitrate Chemical Ionization-Atmospheric Pressure interface-Time Of Flight (CI-APi-TOF) mass spectrometry, *Atmos. Meas. Tech.*, 9, 2135–2145, doi:10.5194/amt-9-2135-2016, 2016.
14. Tröstl, J, et al.: Low-volatility organic compounds are key to initial particle growth in the atmosphere, *Nature*, 533, doi 10.1038/nature18271, 2016.
15. Wimmer, D, et al.: Technical Note: Using DEG-CPCs at upper tropospheric temperatures, *Atmos. Chem. Phys.*, 15, 7547–7555, doi: 10.5194/acp-15-7547-2015, 2015.

Liquid and ice microphysics of clouds:

16. Hoyle, CR, et al., Aqueous phase oxidation of sulphur dioxide by ozone in cloud droplets, *Atmos. Chem. Phys. 16, 1693–1712, doi:10.5194/acp-16-1693-2016*, 2016.
17. Ignatius, K, et al.: Heterogeneous ice nucleation of viscous secondary organic aerosol produced from ozonolysis of α-pinene, *Atmos. Chem. Phys., Atmos. Chem. Phys., 16, 6495–6509,* doi:10.5194/acp-16-6495-2016, 2016.
18. Järvinen, E, et al.: Observation of viscosity transition in α-pinene secondary organic aerosol, *Atmos. Chem. Phys.*, 16, 4423–4438, doi:10.5194/acp-16-4423-2016, 2016.
19. Nichman, L, et al.: Discrimination of water, ice and aerosols by light polarisation in the CLOUD experiment, *Atmos. Chem. Phys. Discuss*., 15, 31433–31469, doi: 10.5194/acpd-15-28575-2015, 2015.

Under review:

20. Lawler, M., et al.: Unexpectedly acidic nanoparticles formed in dimethylamine-ammonia-sulfuric acid nucleation experiments at CLOUD, *Atmos. Chem. Phys. Discuss*. 15, 2016.
21. Ahlm, L., et al.: Modeling the thermodynamics and kinetics of sulfuric acid-dimethylamine-water nanopartice growth in the CLOUD chamber, submitted to *J. Aerosol Sci*., 2016.
22. Dunne, E, et al.: Global particle formation from CERN CLOUD measurements, submitted to *Science*, 2016.
23. Gordon et al.: Reduced anthropogenic aerosol radiative forcing caused by biogenic new particle formation, submitted to *Proc. Natl. Acad. Sci.*, 2016.

Funding

- CLOUD-TRAIN, EU Marie Curie Initial Training Network, Oct 2012 – Sep 2016:
  12 PhD students, 3 Post-Docs, 3.8 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. Scheme to reduce deficit established and well on track. Current deficit <100 kCHF, balanced sheet expected for 2017. CERN support is gratefully acknowledged.

MARIE CURIE ACTIONS

GEFÖRDERT VOM
Bundesministerium für Bildung und Forschung

CERN

Acknowledgements

We would like to thank CERN EP-DT, EN-MME, EN-MEF and TE-VSC for their excellent support of CLOUD and, in addition, to thank the CERN PS machine team and the PS Coordinator for their strong support of CLOUD and for efficient operation of the PS!