Nucleation and particle growth

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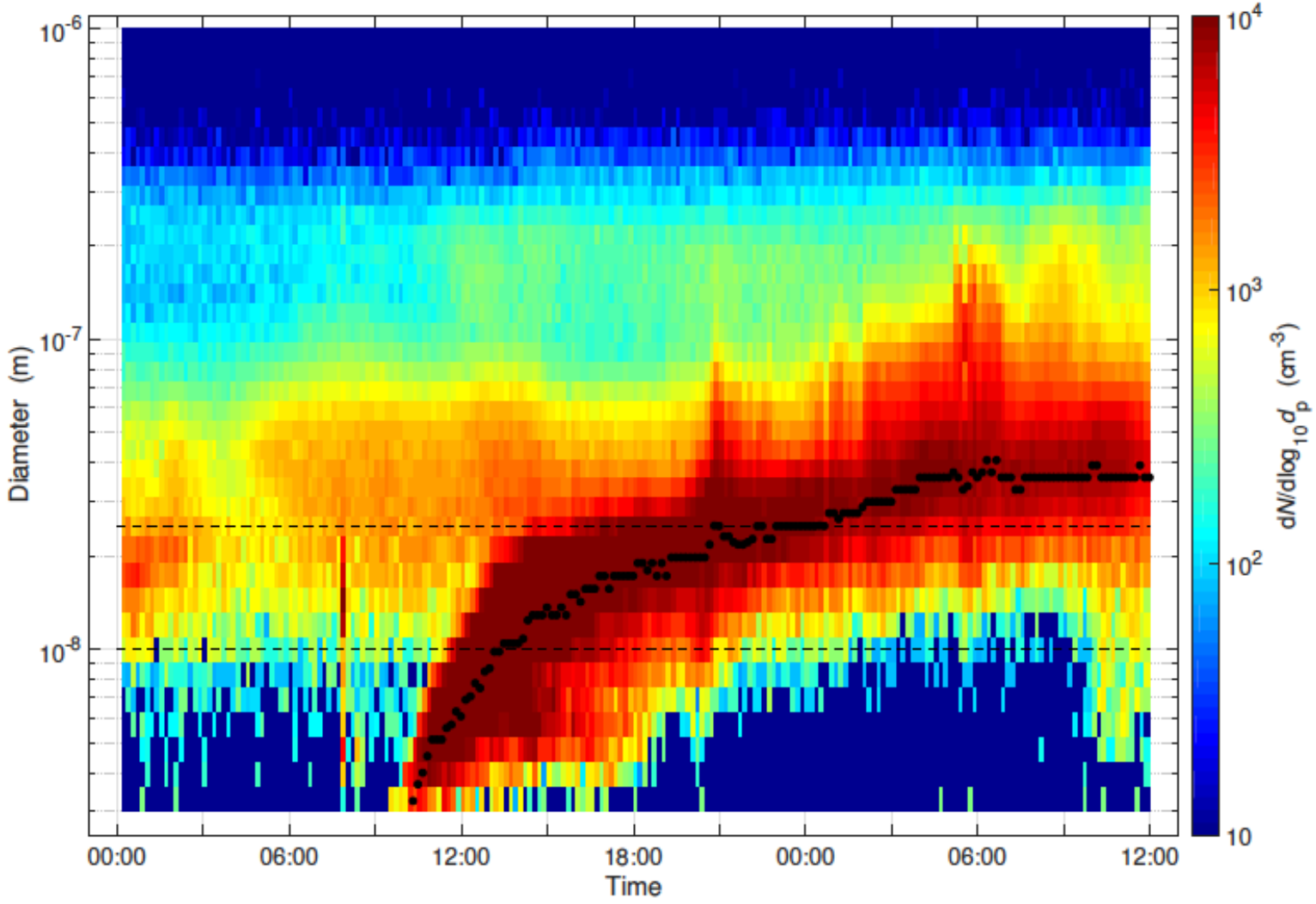
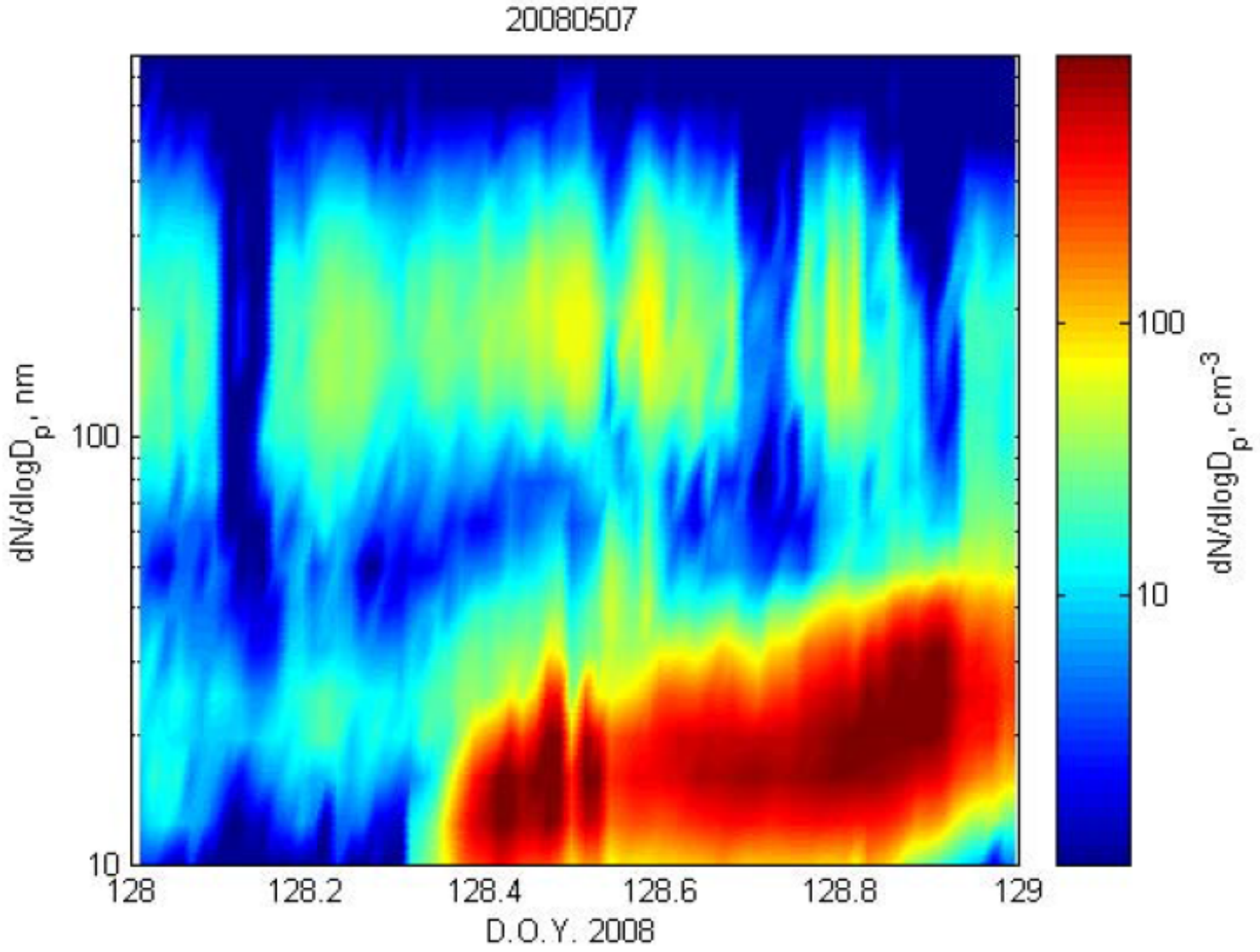


An example of a new particle formation event observed in Hyytiälä, Finland, 15–16 March 2011, illustrating the continuous growth of the newly formed aerosol particles for about 25 h.

Nieminen et al, ACP, 2018



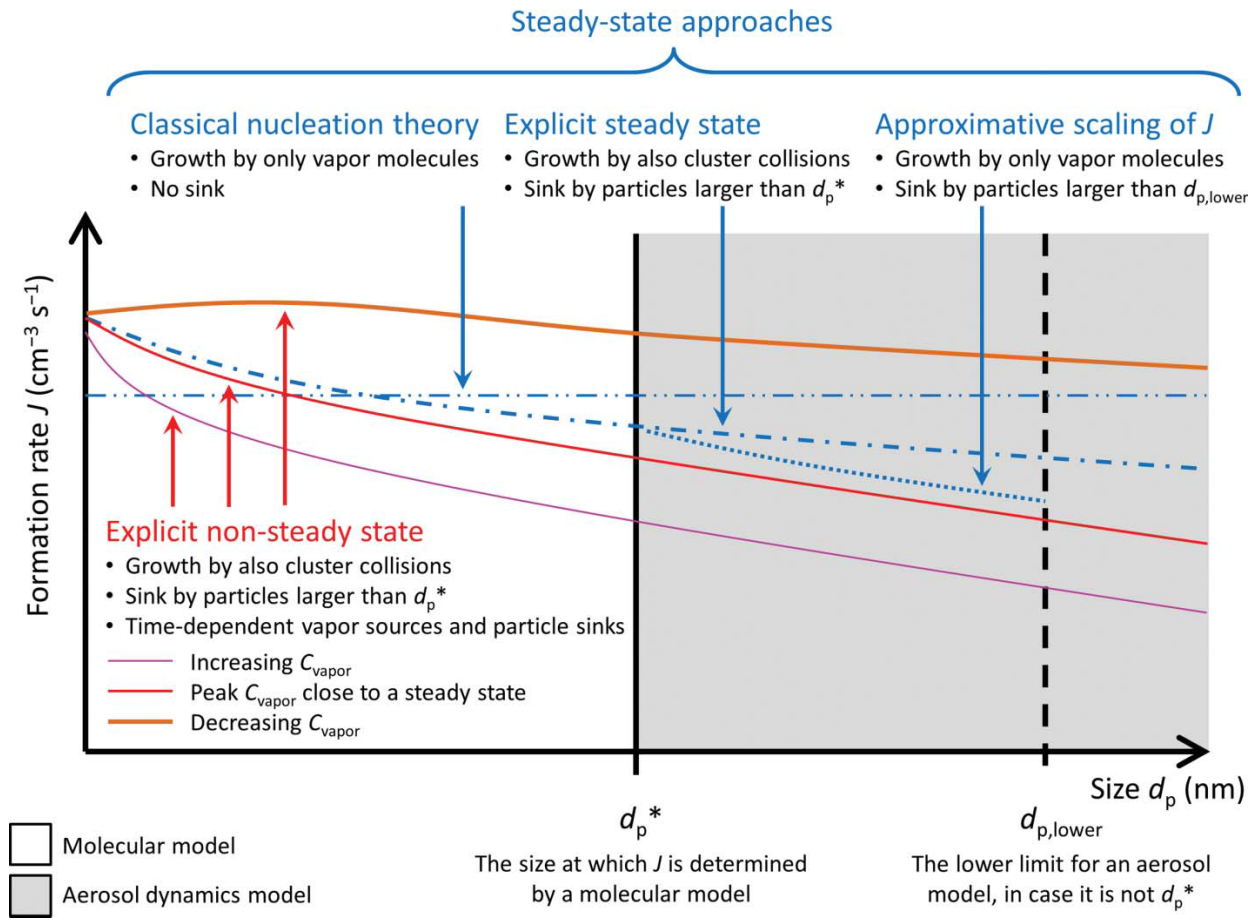
Example of an Arctic nucleation event as observed 7 May 2008.

dN/dlogDp (cm−3) versus decimal day of year (D.O.Y.).

Tunved et al., ACP, 2013

What is nucleation?

- Recommendation! Read the review “New Particle Formation in the Atmosphere: From Molecular Clusters to Global Climat” by Lee et al., JGR 2018
- Nucleation; A two step process
  - First step; formation of a critical nucleus during the phase transformation from vapor to liquid or solid
  - Second step; growth of the critical nucleus to a larger size (>2–3 nm) that competes with removal by preexisting aerosols
- The further growth of the particles are by condensation

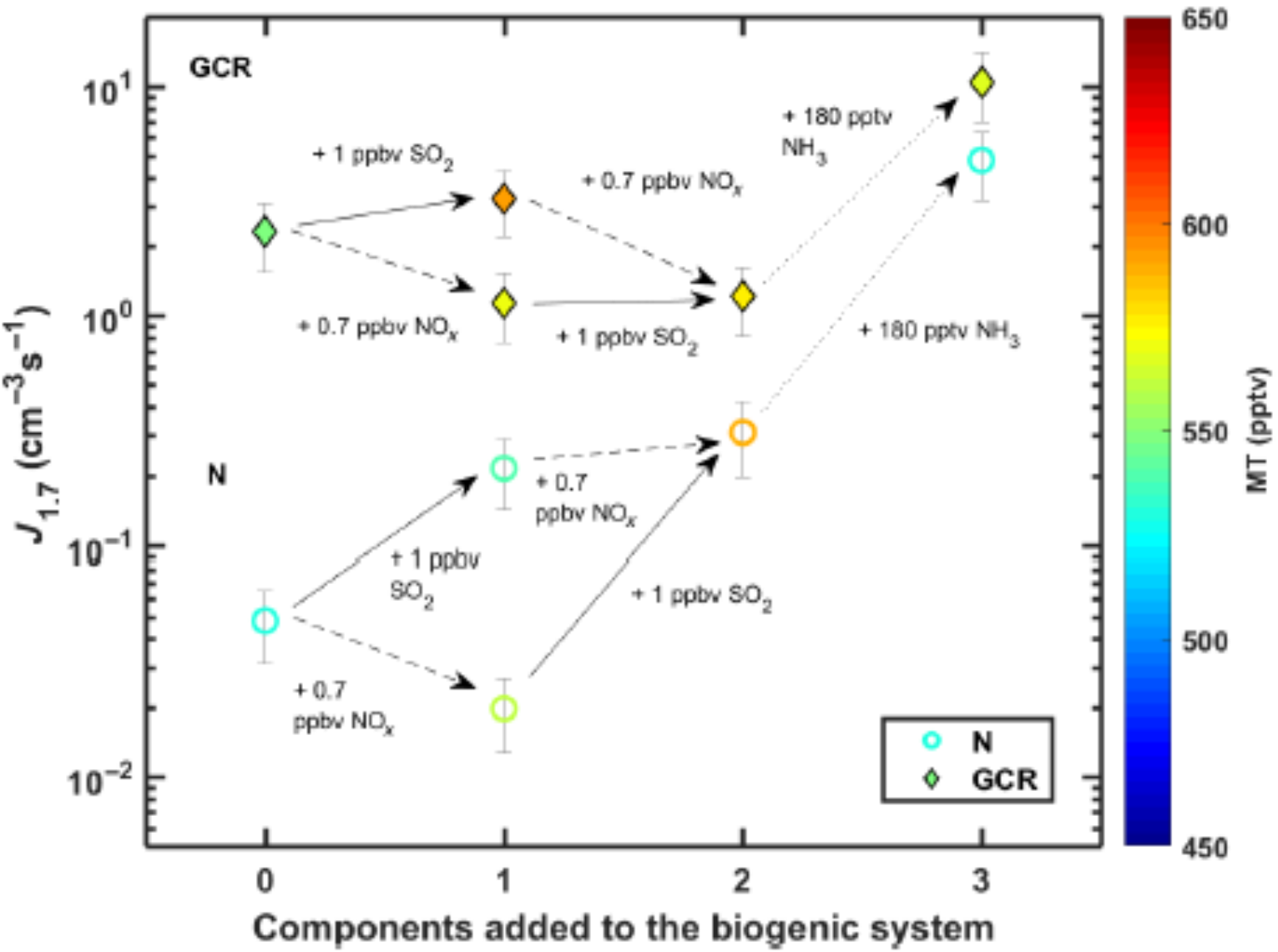


Olenius and Riipinen, AST, 2016

Schematic figure of the size-dependent particle formation (nucleation) rate described by a molecular model, and its incorporation in an aerosol dynamics model.

The formation rates are qualitative, and demonstrate the differences of the different steady-state approaches (blue) to each other, as well as to the true dynamic formation rate in the case of a time-dependent situation (red hues). The solid (red-hued) lines illustrate an example behavior of the dynamic formation rate relative to the steady-state rate in a diurnal cycle where the vapor concentration C_{vapor} first increases, then stays at a maximum, and finally decreases. The relations of the rates may be different depending on the ambient conditions; e.g., the scaling approach may give values either lower or higher than the explicit steady-state solution.

The primary goal of CLOUD is to understand the influence of galactic cosmic rays on aerosols and clouds, and their implications for climate.



Lehtipalo, 2018, Sci Advances

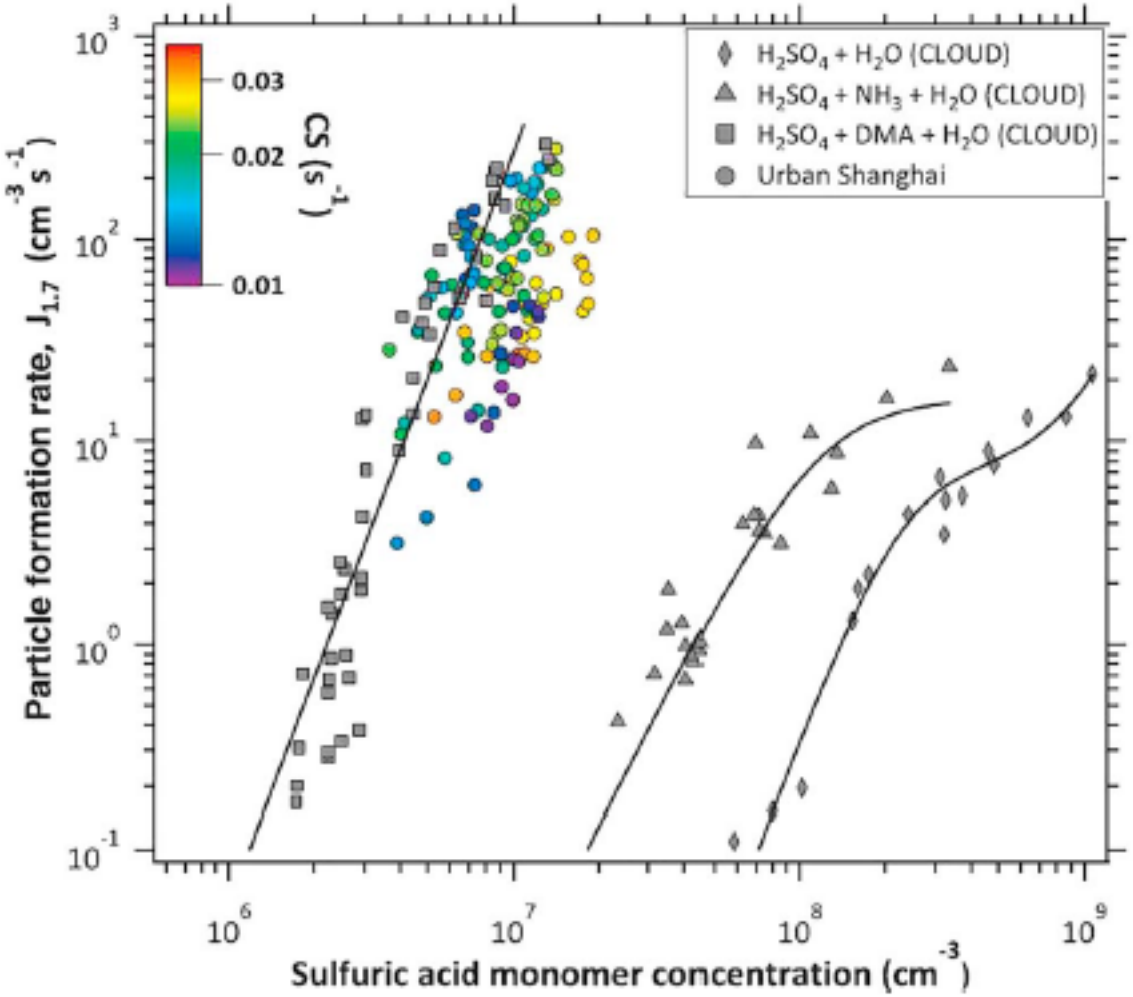
The effect of adding different vapors on biogenic nucleation rates (J1.7). All points have similar MT (530 to 590 pptv) and ozone (40 ppbv) mixing ratios.

The leftmost points were measured with only MTs added to the chamber, and each step to the right represents addition of one more component to the system.

Solid arrows describe the addition of ca. 1 ppbv of SO_2 (resulting in an H_2SO_4 concentration of 1 × 10^7 to 2 × 10^7 cm^{-3}), dashed arrows describe the addition of ca. 0.7 ppbv of NOx , and dotted arrows describe the addition of ca. 180 pptv of NH_3.

Circles are experiments at neutral conditions (N), and diamonds are experiments at GCR conditions.

Colors of the symbols indicate the measured MT mixing ratio.

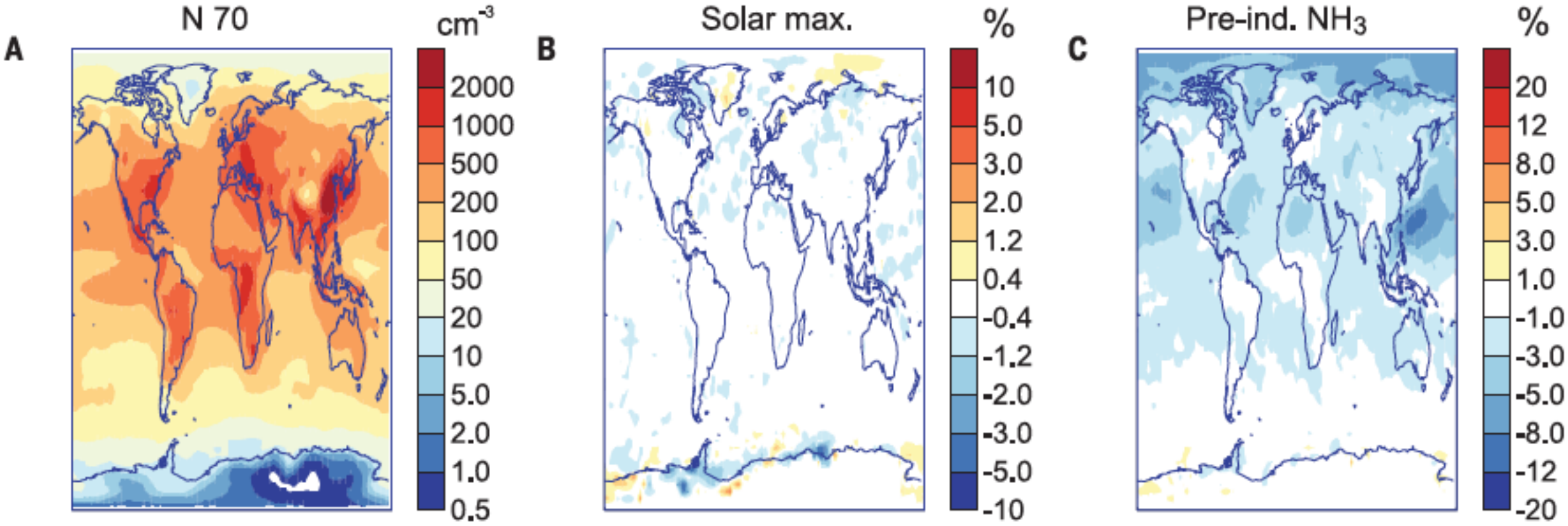


Lee et al., 2018, JGR

Measured particle formation rates for 1.7-nm particles (J1.7) as a function of sulfuric acid concentrations and condensation sink (CS) in urban Shanghai, China (colored circles).

In comparison, binary (diamonds) and ternary nucleation with ammonia (triangles; Kirkby et al., 2011) and diamine (squares; Almeida et al., 2013) experimental results taken from CLOUD chamber experiments are also included. Adapted from L. Yao et al. (2018).

Cosmic rays variations do not affect CCN while anthropogenic ammonia does!



Modeled present-day CCN concentrations and the effect of perturbations. Hygroscopic particles above 70 nm diameter are used as a proxy for CCN.

(A) Annual mean CCN concentrations at about cloud base altitude (915 hPa)

(B) Effect of changing the heliospheric modulation potential from solar minimum to maximum.

(C) Effect of reducing ammonia concentrations to preindustrial levels.

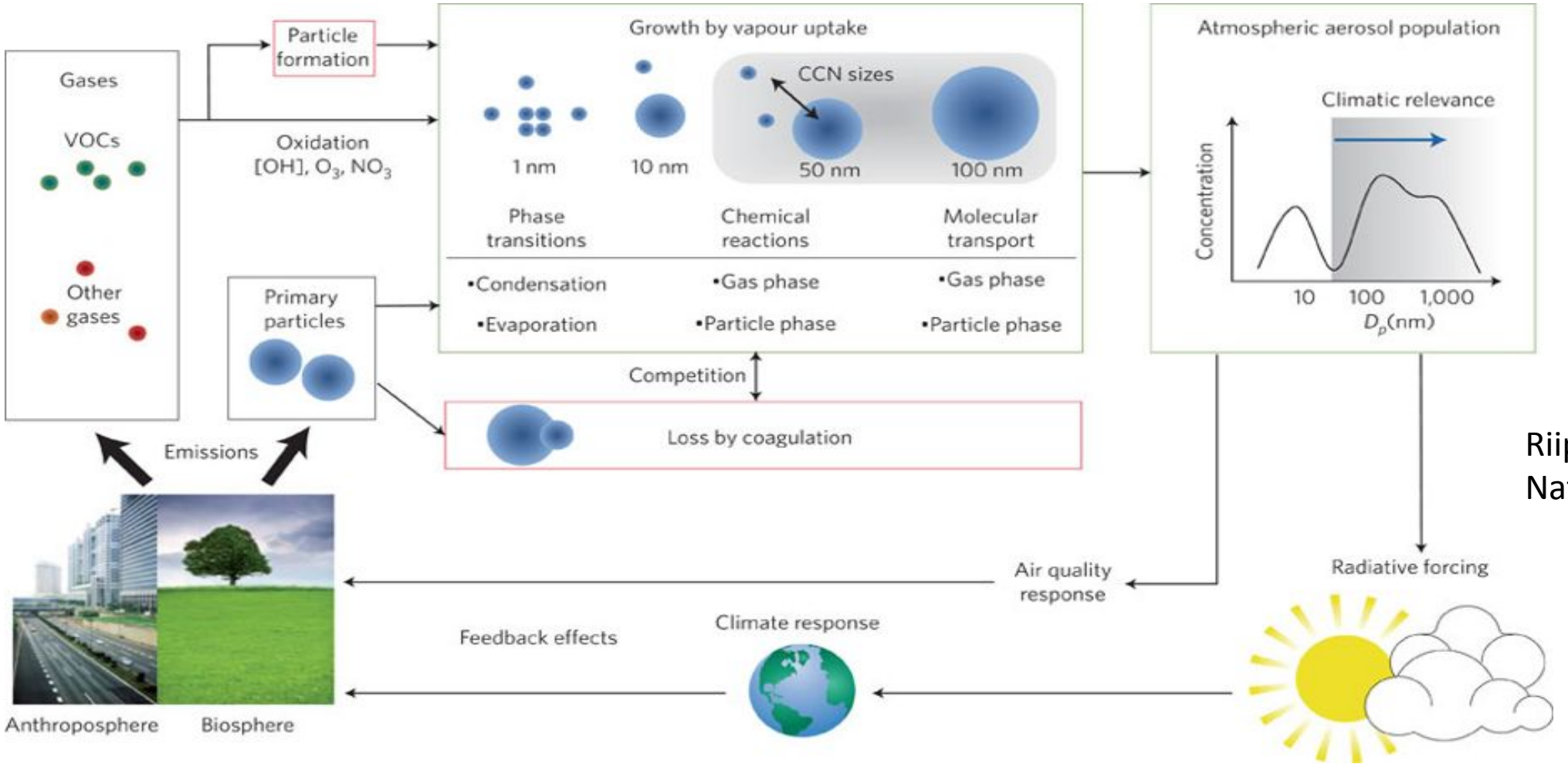
Gasphase chemistry provides the precursor compounds

- Always H_2SO_4 but also
  - Ammonia / Amines
  - Organics
  - Ions
  - Combined give highest nucleation rates

- How is not really known
  - ”Currently, the mechanism and chemical species responsible for atmospheric NPF are still highly uncertain. In particular, consistent chemical mechanisms to explain NPF under diverse atmospheric conditions are still lacking.” (Lee et al., JGR, 2018)

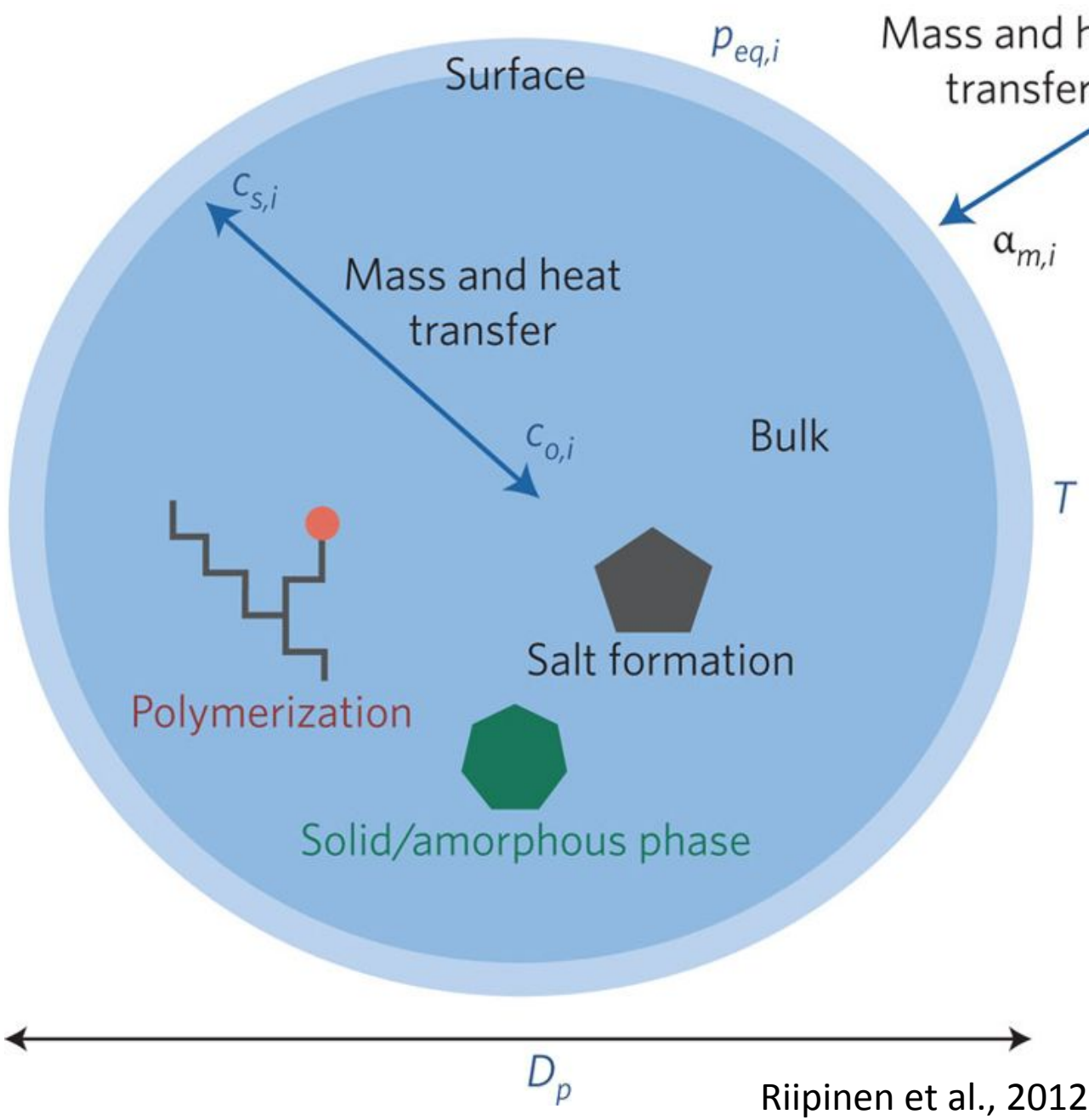
- Nucleation is strongly dependent of thermodynamic parameters as RH and T
  - “Furthermore, the atmospheric conditions regulating NPF events are also poorly understood, including temperature, RH, and preexisting background aerosols.” (Lee et al., JGR, 2018)

Connections between volatile organic compound emissions and nanoparticle growth.



Riipinen et al., 2012, Nature Geoscience

VOCs are oxidized by O_3, OH and NO_3, and the vapours formed in these reactions condense on to particles. Freshly formed particles must grow from ~1 nm to 30–100 nm before colliding with larger particles to have an impact on climate. Uptake of organic vapours is a major source of the growth of these small particles.



Riipinen et al., 2012, Nature Geoscience

The processes influencing organic vapour uptake by atmospheric nanoparticles.

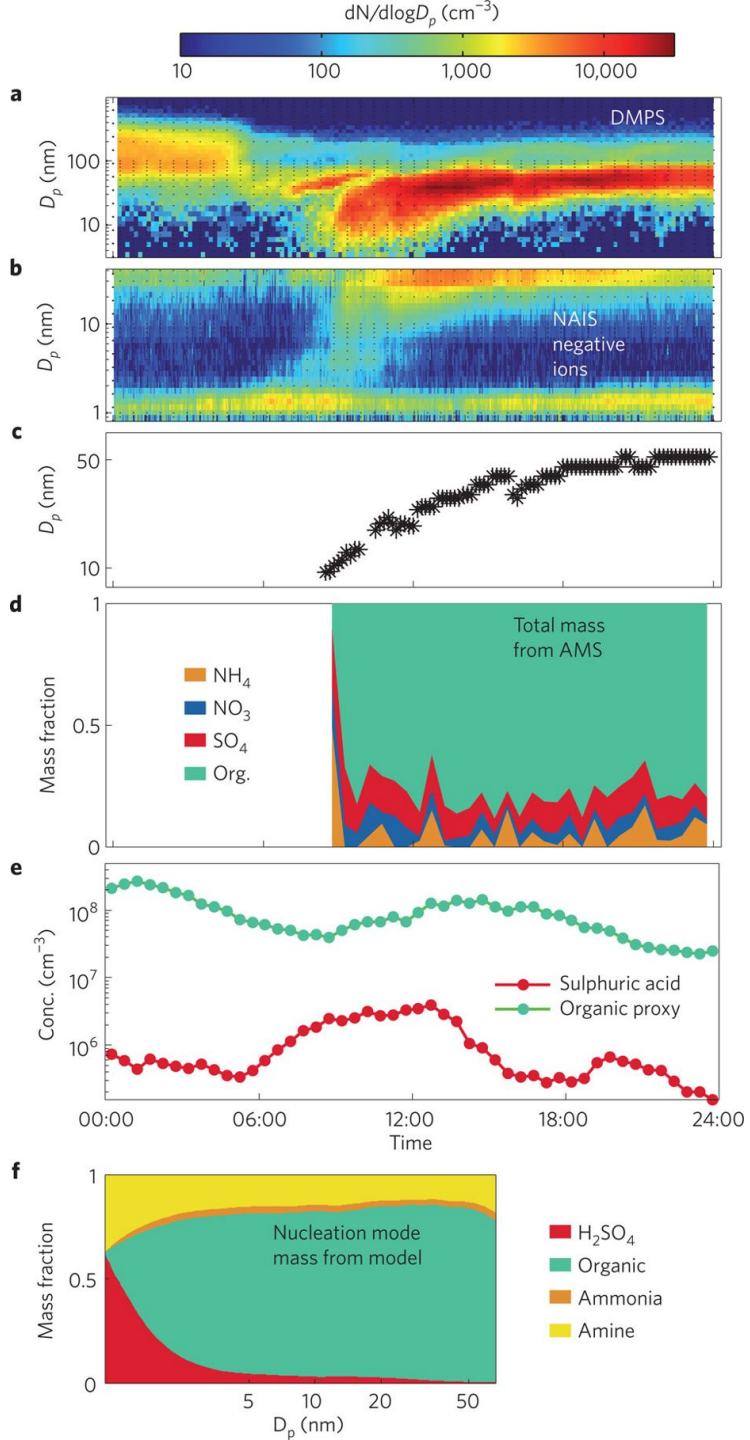
cs,i is the concentration of compound *i* at the particle surface;

co,i is the concentration of *i* in the bulk;

peq,i is the equilibrium partial pressure of *i* at the particle surface and *p∞,i* far away from the surface;

α*mi* is the mass accommodation coefficient of *i*. *T*, temperature; *Dp*, particle diameter.

Mass transport is proportional to the vapour pressure or concentration gradients in the gas or particulate phase, and, in practice, coupled with heat transport (due to the latent heat of the phase transition reactions).



Particle formation event on 23 July 2010 in Hyytiälä, Finland

a,**b**, Aerosol size distribution measured with a differential mobility particle sizer (DMPS) and a neutral cluster and air ion spectrometer (NAIS).

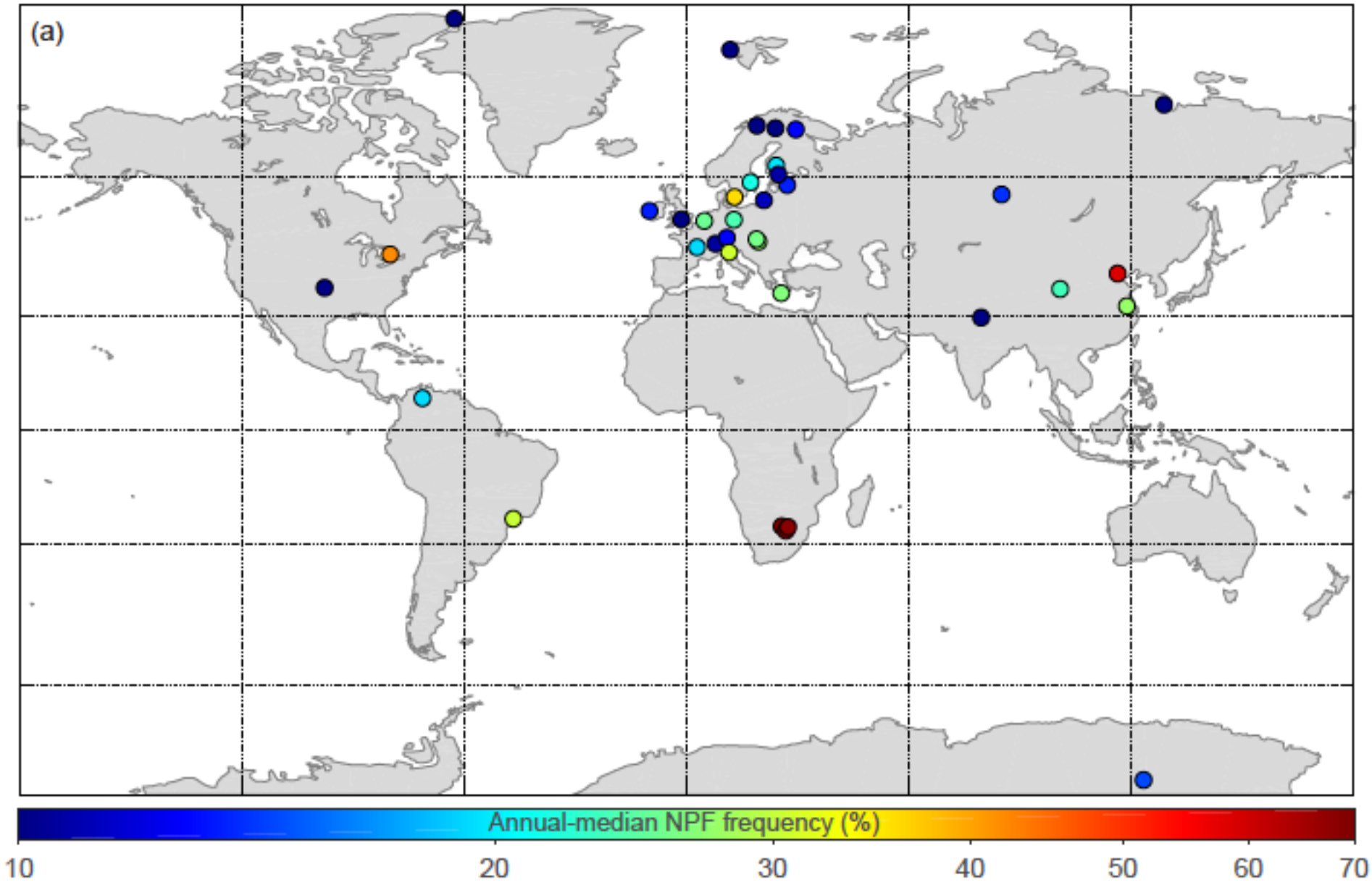
c, Median diameter of the growing particles. *Dp*, particle diameter.

d, >20 nm particle composition measured with the aerosol mass spectrometer (AMS).

e, Concentrations of gas-phase sulphuric acid from the chemical ionization mass spectrometer (CIMS) and organic acid proxy, whose diurnal profile was constrained by measured malonic acid concentration and level by organic flux to the aerosols.

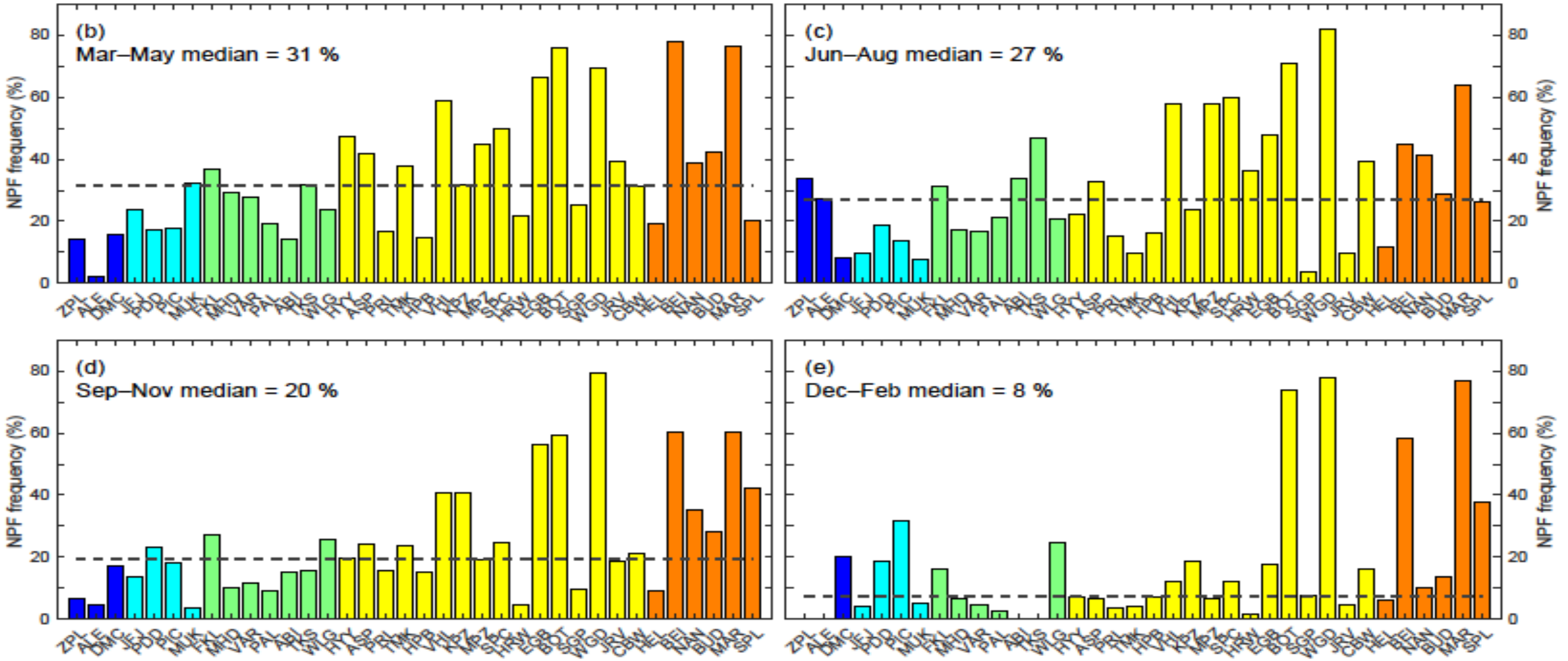
f, Modelled composition of particles growing by uptake of sulphuric acid, organic acid, trimethylamine (5×10^7 cm^{-3}) and ammonia (2×10^{10} cm^{-3}), where the initial cluster contains sulphuric acid and trimethylamine, saturation vapour pressure of the organic acid is 10 times higher than sulphuric acid, and the stability of aminium salts is 1000-fold higher than ammonium salts

Riipinen et al., 2012, Nature Geoscience



Nieminen et al, ACP, 2018

Annual-median frequency of the NPF formation events at the different measurement sites.



Seasonal-median frequency of the NPF formation events at the different measurement sites. The dashed lines show the median seasonal values, and the color scheme represents the classification of the sites into polar, high-altitude, remote, rural, and urban environments.

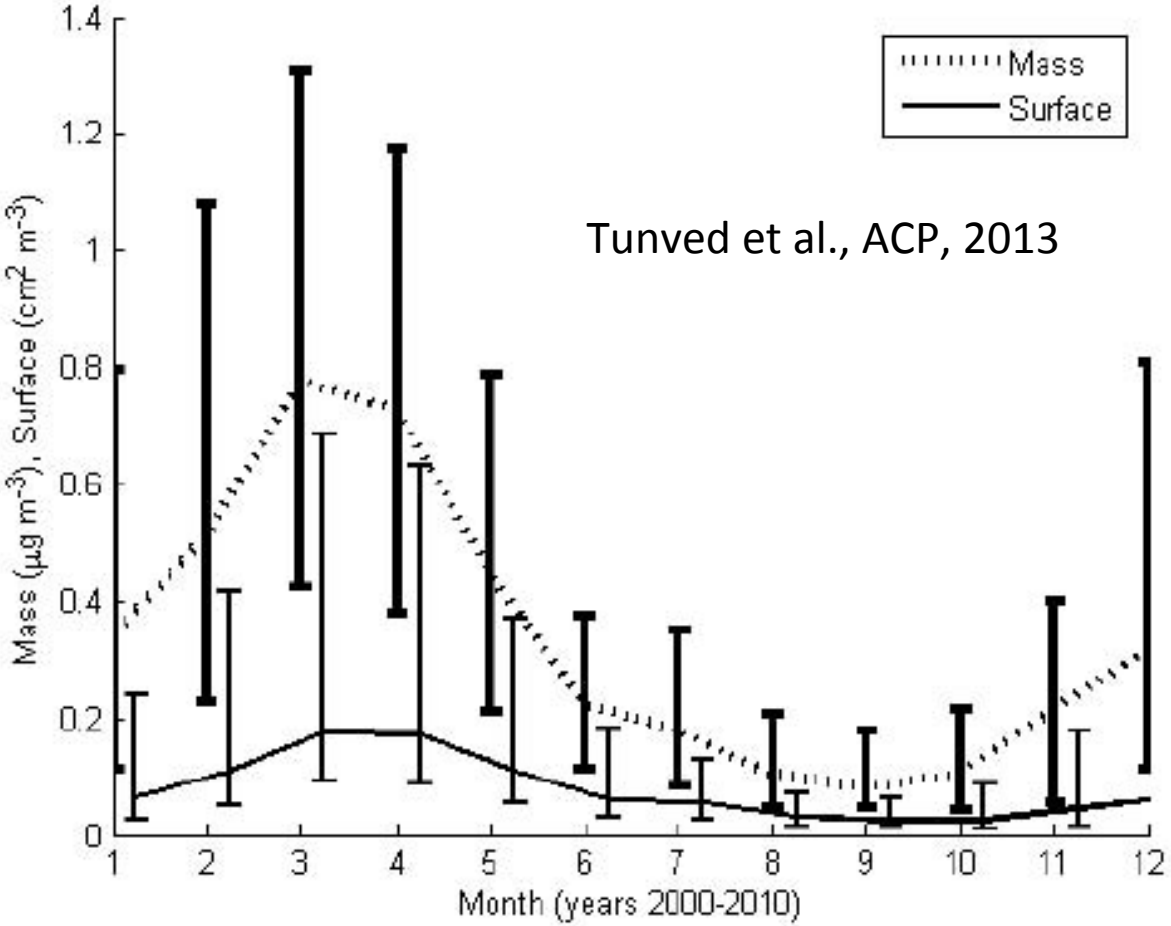
Nieminen et al, ACP, 2018

Well perhaps looking in more detail ……..

- Polar seems less nucleating, but shown to be strongly seasonal
- Amazon, excluded shows almost NO nucleation
- Central Siberia reevaluated show 11 events in 3 years (Wiedensohler et al., 2018)
- Why is that??

Zeppelin station, Ny Ålesund, Svalbard

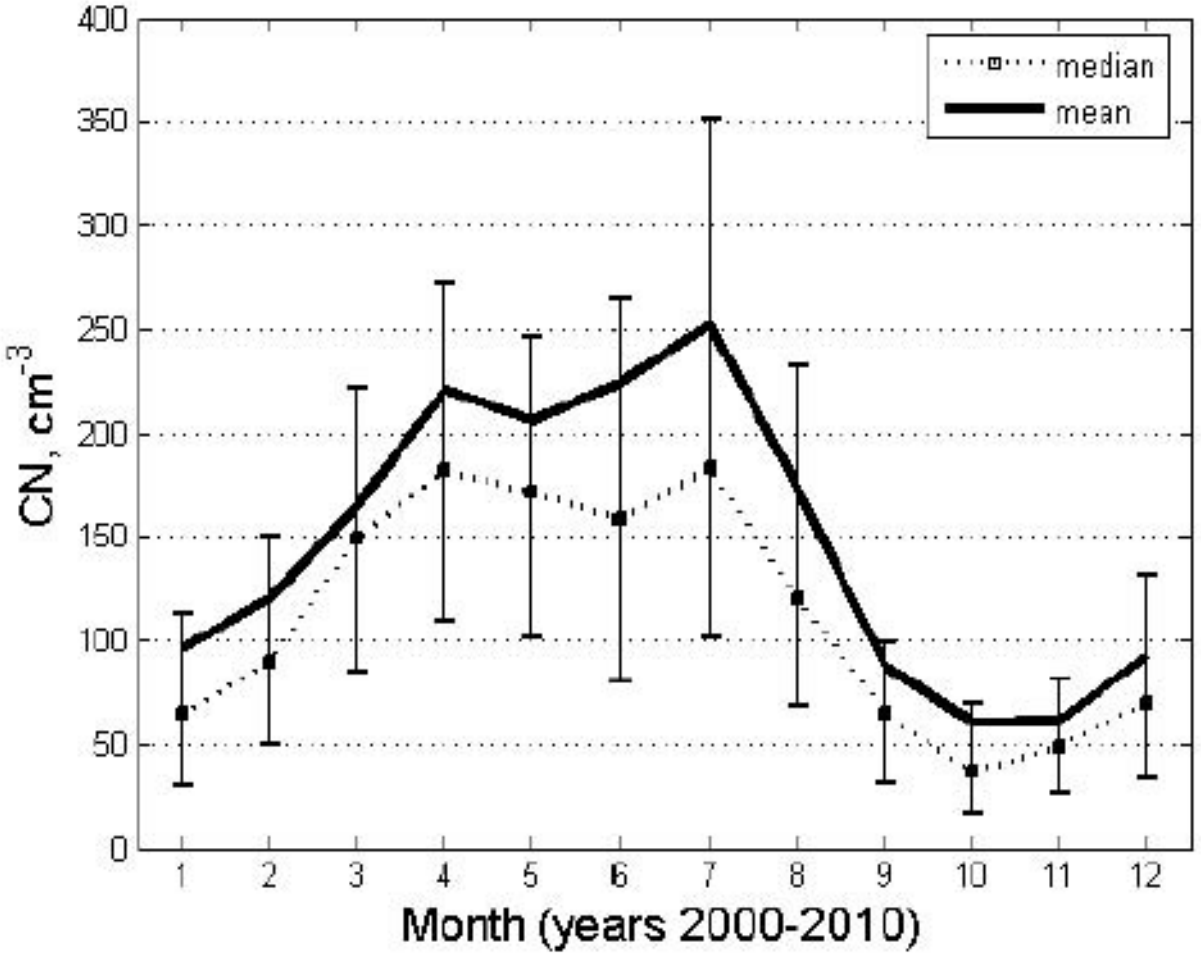
On my way to Zeppelin flying over Svalbard 1st of April 2019



Tunved et al., ACP, 2013

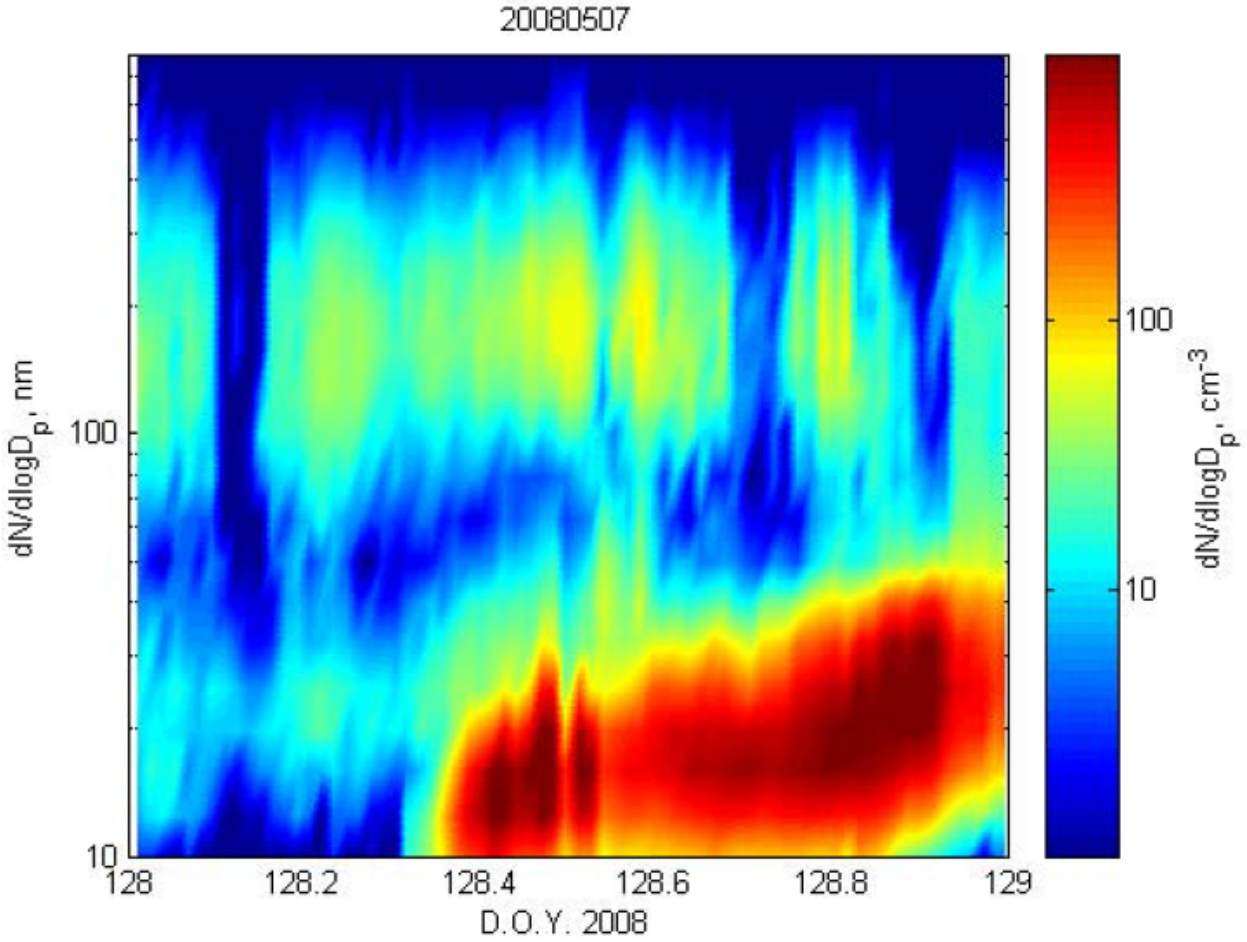
Annual average variation of integrated surface and mass, March 2000–March 2010.

Mass data calculated from aerosol number size distribution assuming a density of = 1 g cm−3. 25–75th percentile ranges indicated by errorbars.



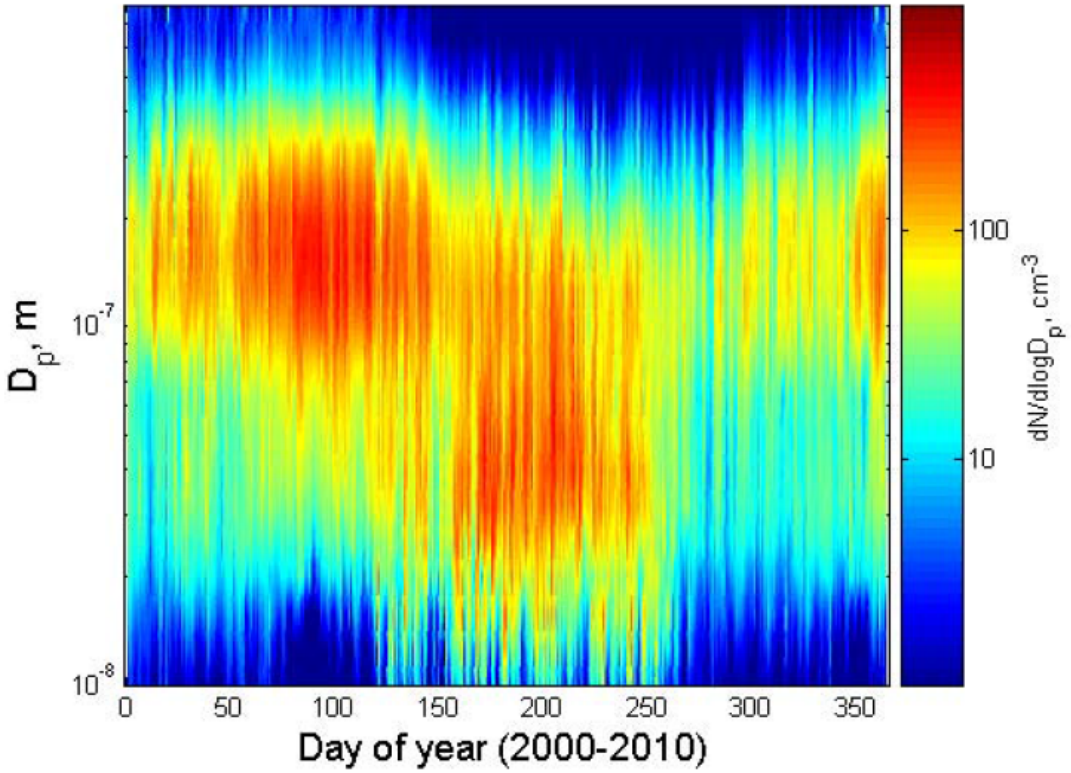
Annual average variation of median and mean integrated number concentration per month March 2000–March 2010.

25–75th percentile ranges indicated by vertical "error bars".



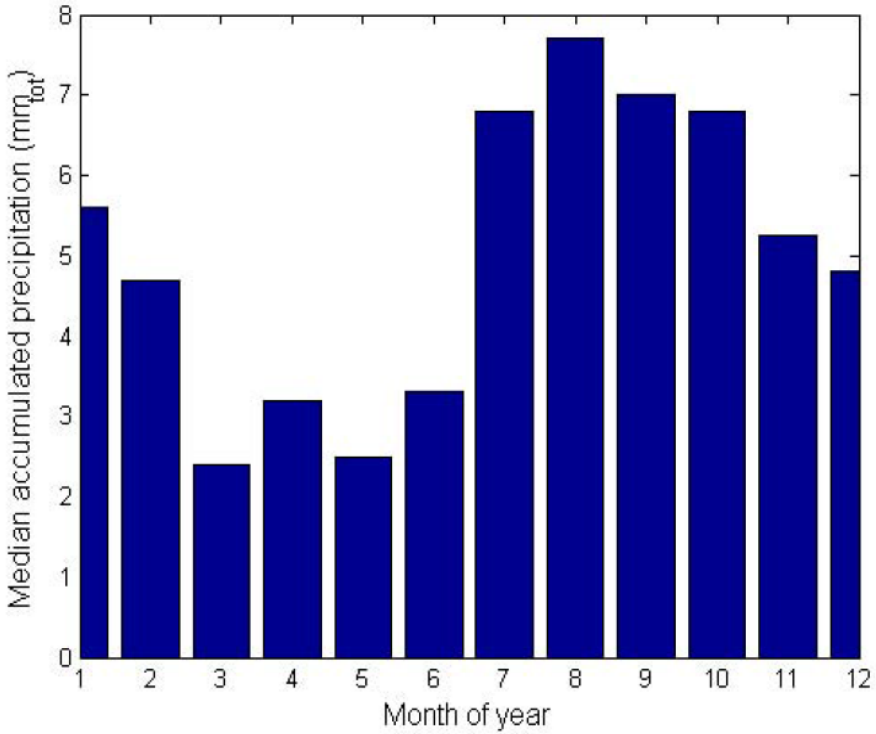
dN/dlogDp (cm−3) versus decimal day of year (D.O.Y.).

Tunved et al., ACP, 2013



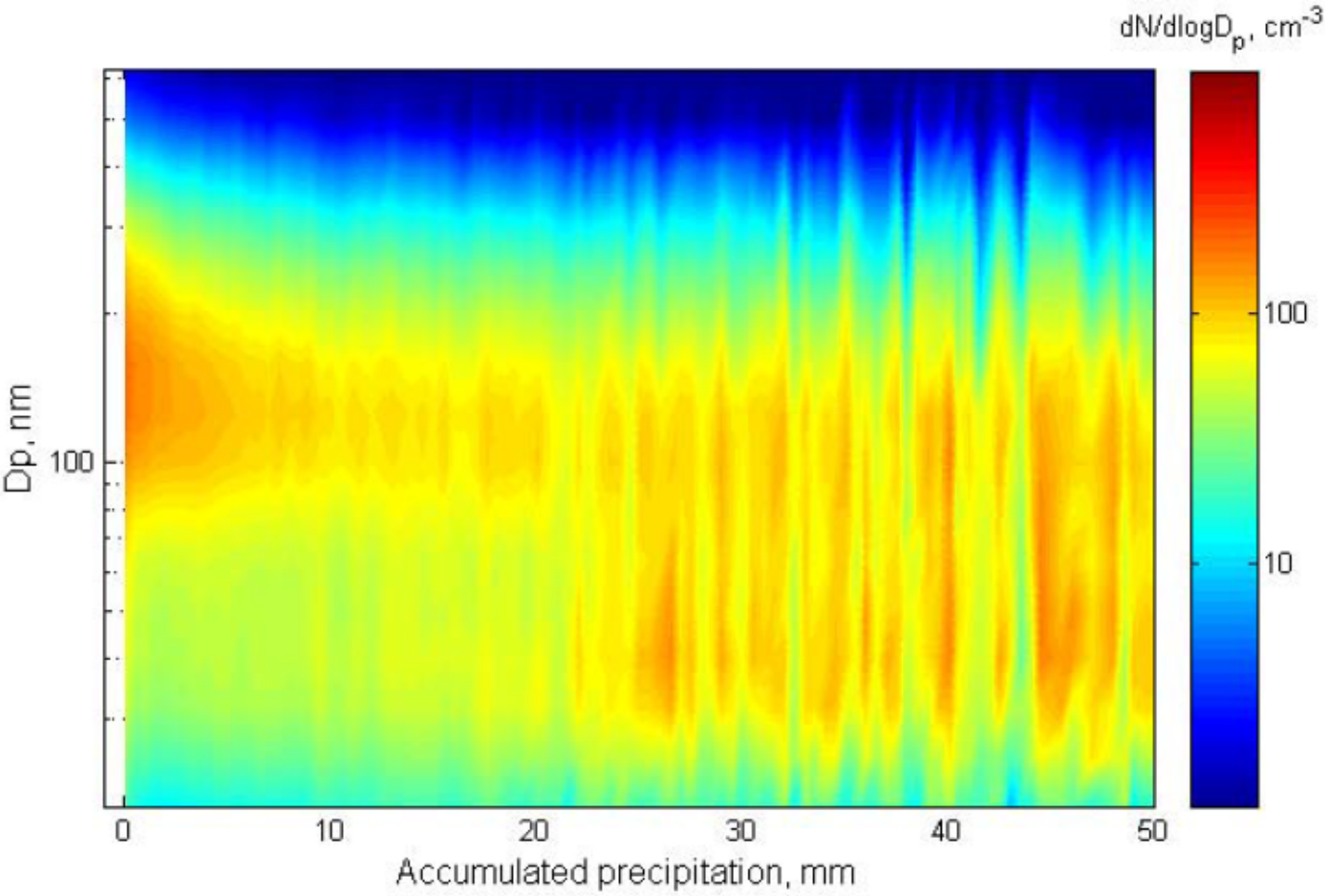
Spectral plot of daily average aerosol number size distributions, March 2000–December 2010.

Units on x-axis as day of year.



Monthly median of accumulated precipitation experienced by trajectories arriving at Svalbard during the period of 2000–2010.

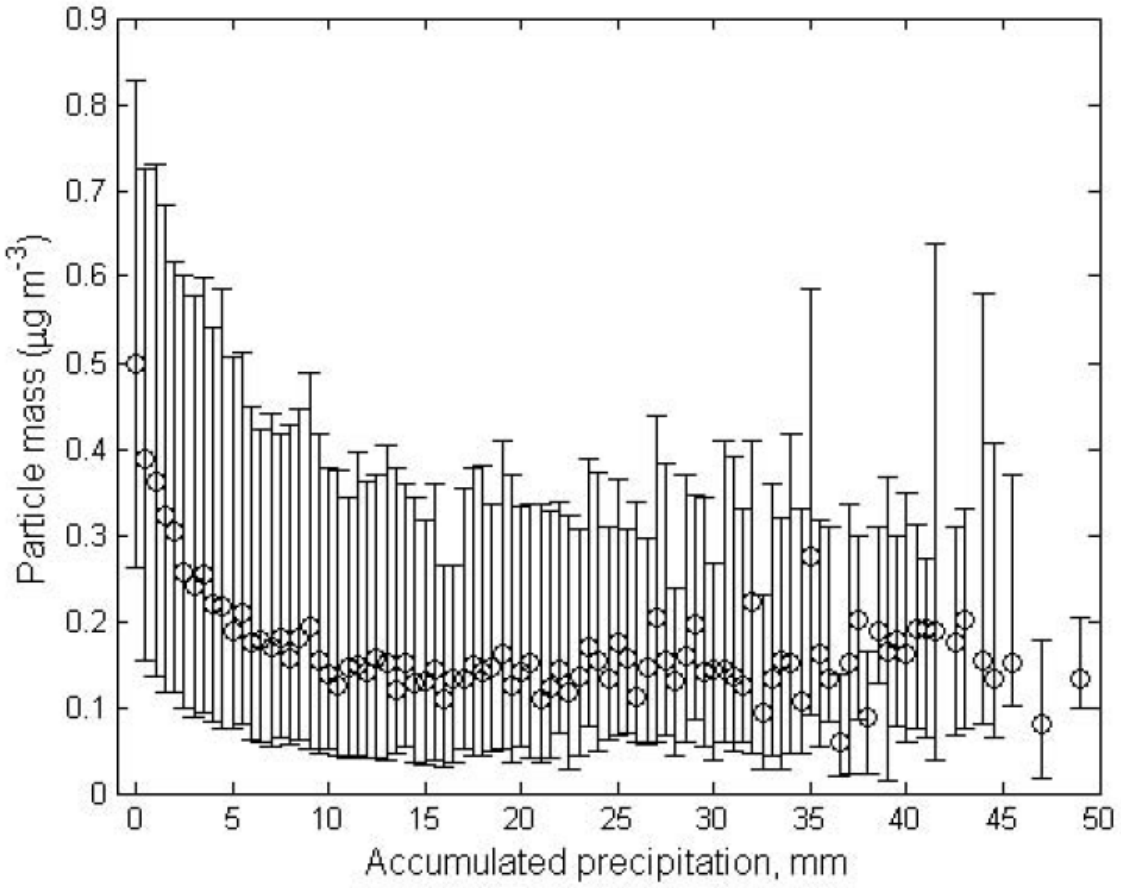
Tunved et al., ACP, 2013



Evolution of aerosol number size distribution as a function of accumulated precipitation (mmtot) along 240 h trajectories.

Data are shown binned over a step size of 0.5mmtot, and the corresponding size distributions over this ranges of precipitation is presented as median values.

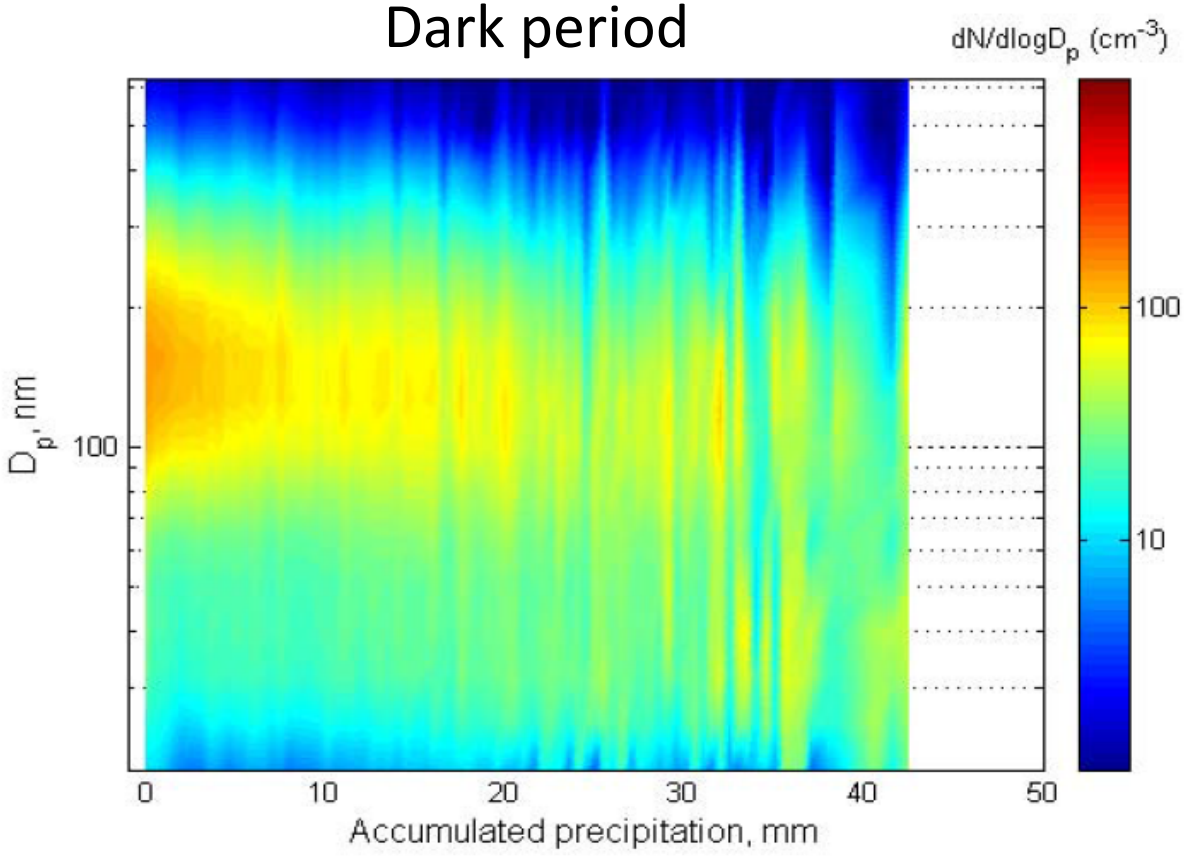
Tunved et al., ACP, 2013



Submicron aerosol mass (10–630 nm; 1 g cm−3) as a function of accumulated precipitation along the trajectories. All data collected between 2000 and 2010.

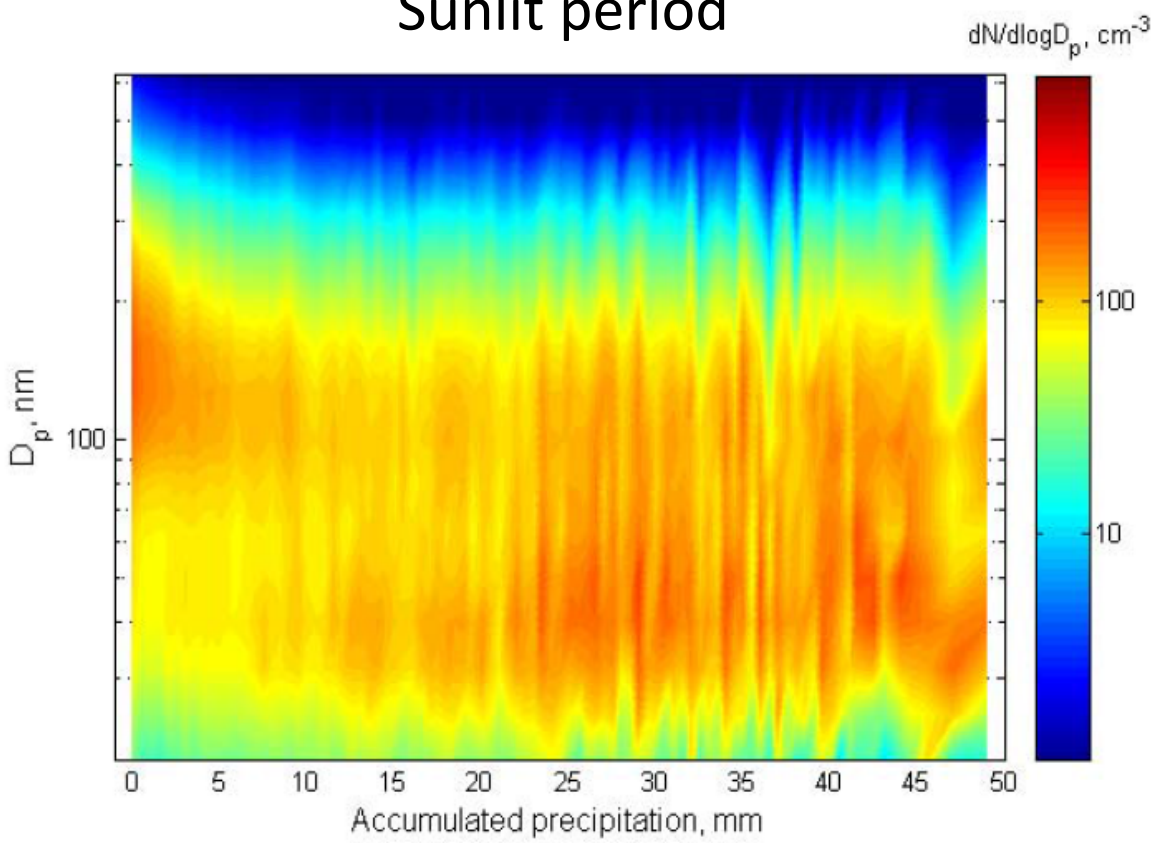
Data are shown as 25–75th percentile ranges per bin (solidlines) and median (circles).

Dark period



Evolution of aerosol number size distribution as a function of accumulated precipitation (mmtot) along 240 h trajectories for the dark period (October–February). Data from 2000–2010

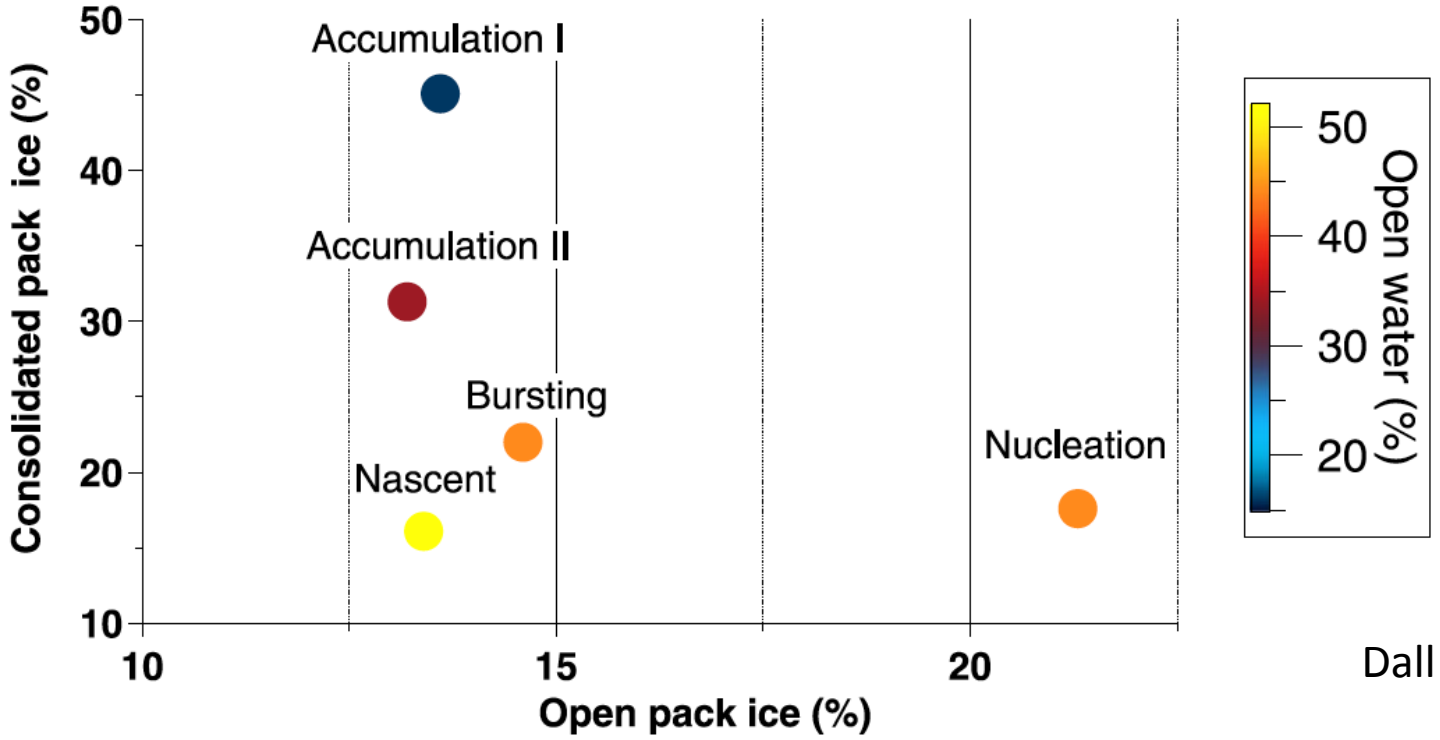
Sunlit period



Evolution of aerosol number size distribution as a function of accumulated precipitation (mmtot) along 240 h trajectories for the sunlit period (March–September). Data from 2000–2010.

Tunved et al., ACP, 2013

Origin of precursor gases seem to be the open pack ice?



Dall Osto, Sci Report, 2017

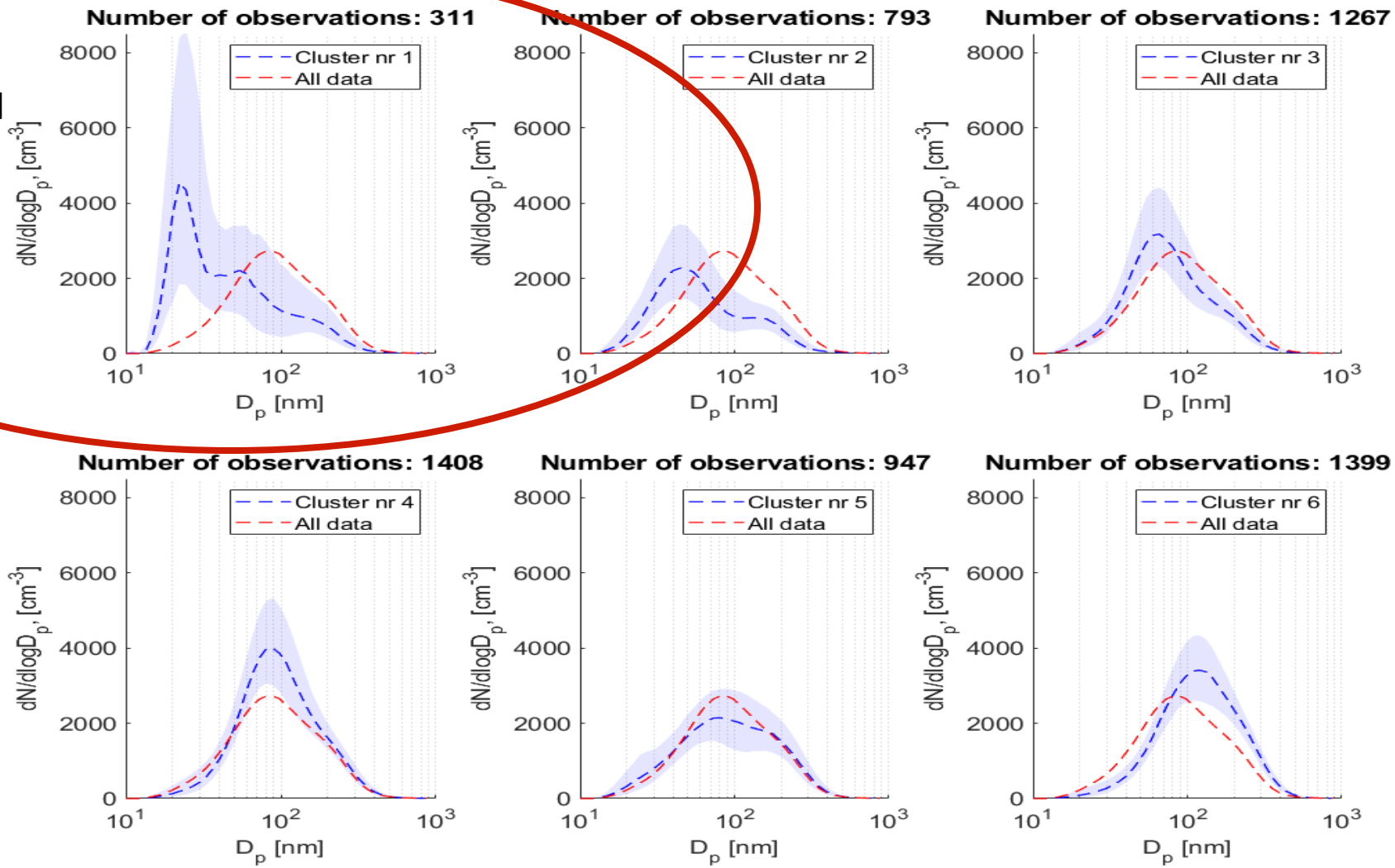
Percentages of total time (hours) of air mass back trajectories travelling over different sea ice areas for each of the 5 aerosol categories.

Arctic sources, sinks and nucleation

- Seasonal variations in size reveal different source processes
- Long distant transport can be facilitated by meteorology
- Precipitation dominating deposition process
- Sink processes can induce formation processes
- The nucleation observed is probably natural but how is still unclear
  - DMS, ion induced binary nucleation ?
  - Organics microgels?
  - Sun light needed!

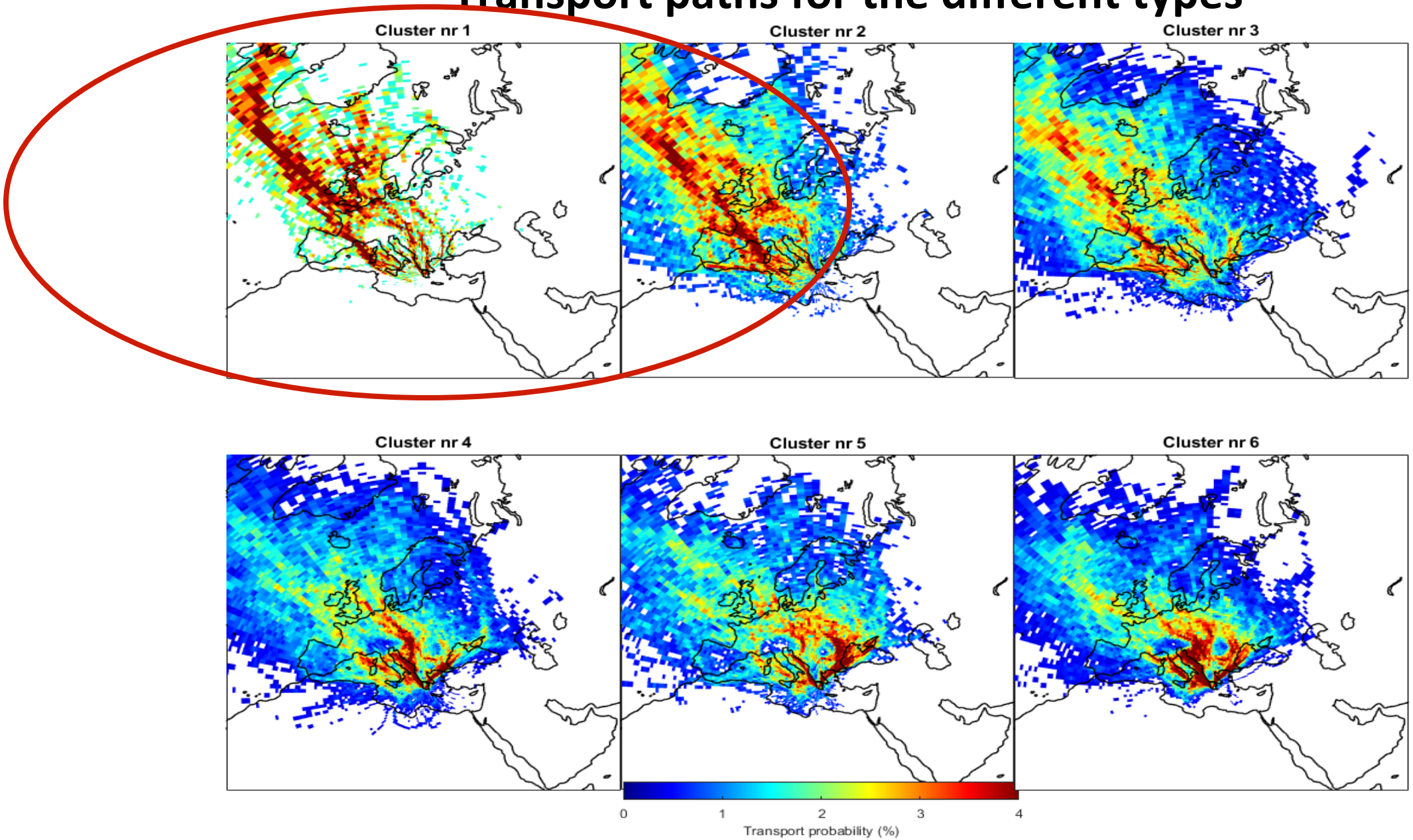
Background aerosol over the Mediterranean

About 5% of all

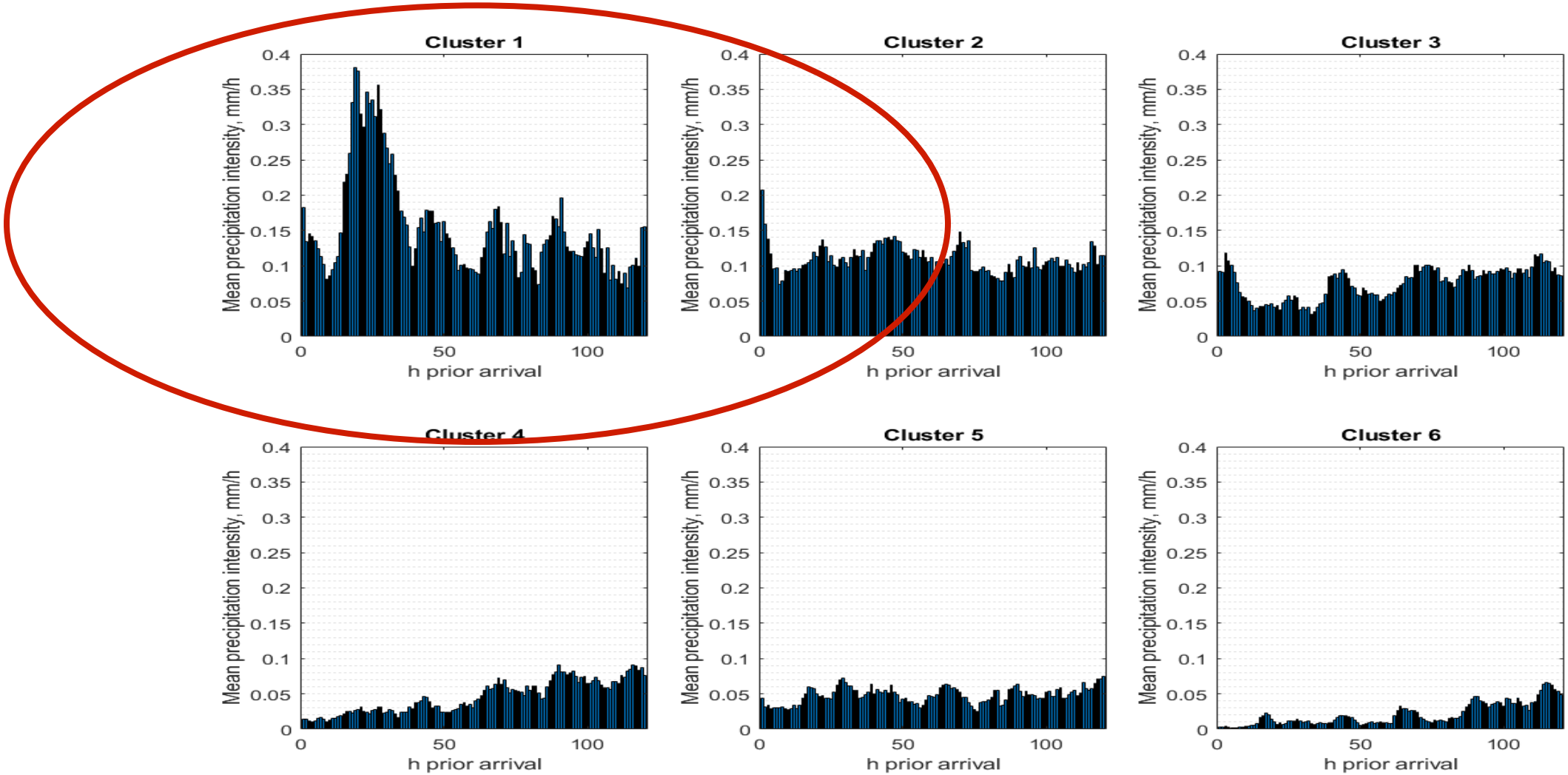


Six most abundant size distribution types

Transport paths for the different types



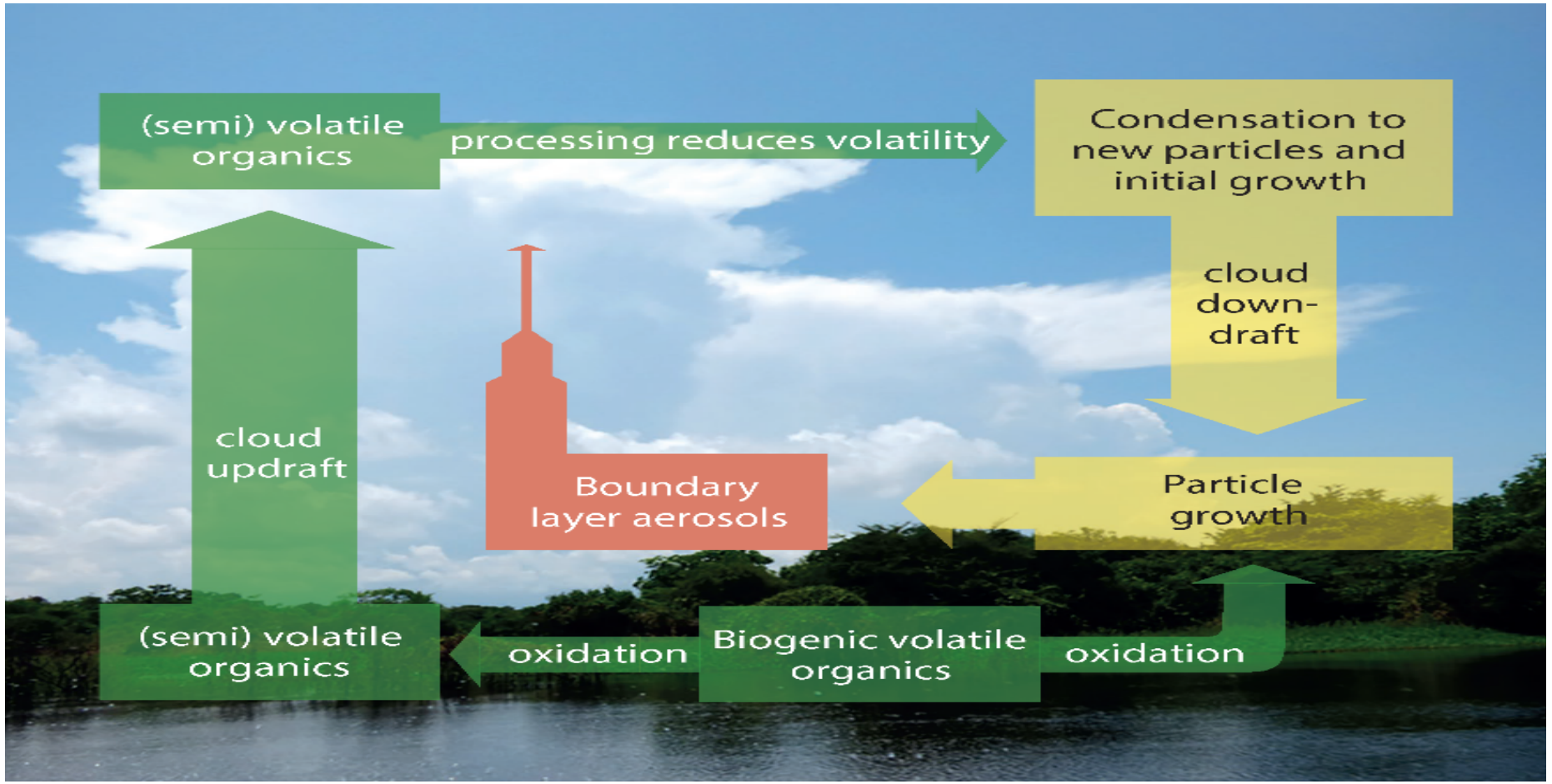
Precipitation history for the different size distribution types



Meteorology, sources, transport and precipitation controls nucleation

- Sources brings the needed nucleation precursor gases
- Atmospheric chemistry including needed photochemistry during transport providing nucleation precursors and condensable gases
- Precipitation scavenge condensation sink

Nucleation in the Amazonas occurs in the outflow from the top of the clouds

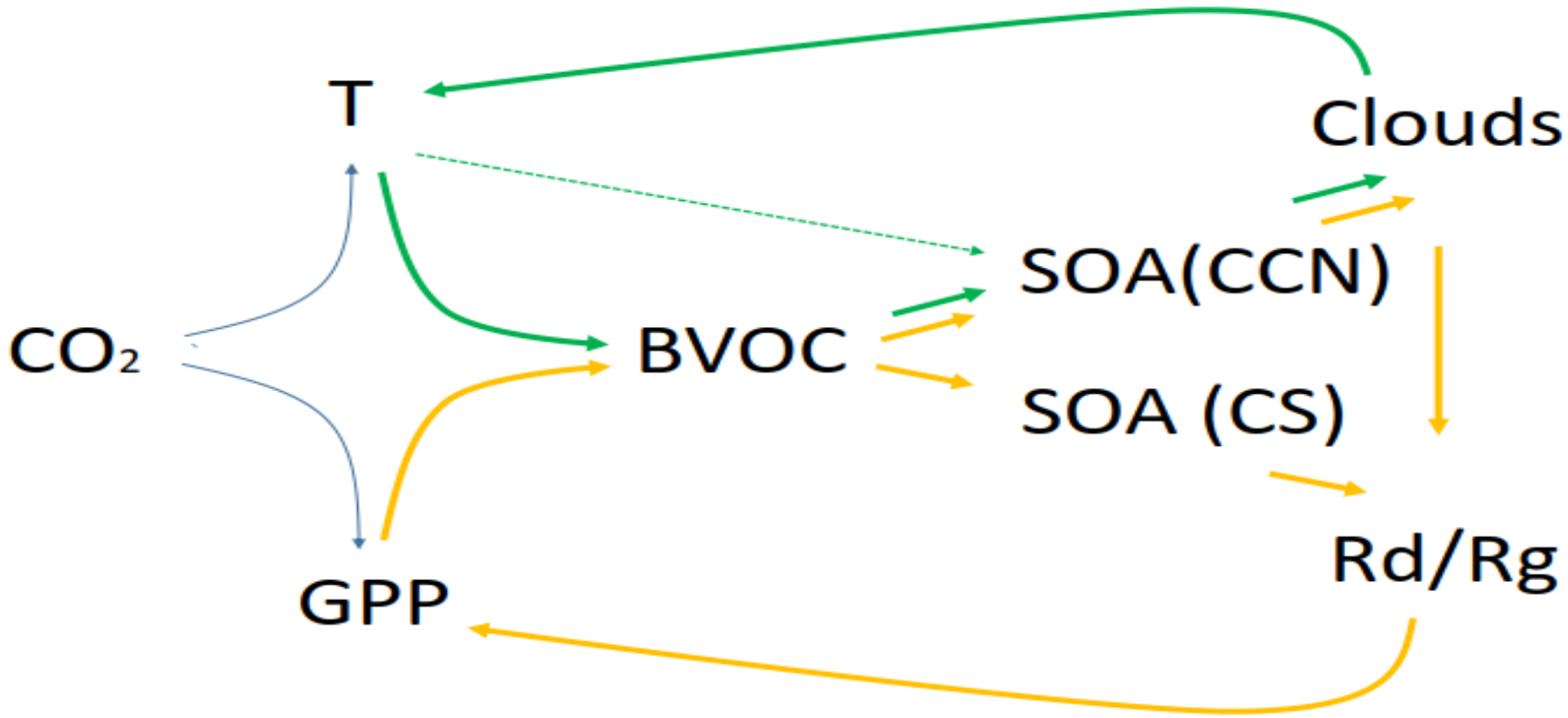


Conceptual model of the aerosol life cycle over the Amazon Basin (from Andreae et al, 2018).

Questions arise

- Central Siberia similar few observations of nucleation as Amazonas. Why? Where is the nucleation occurring?
- Why are the nucleation so frequent in Scandinavia?
- Will “nature” always provide new particles when needed?
- Is there a natural balance? And how has that been changed due to anthropogenic emissions?


Thank you for listening and discussing
See you Friday afternoon



Schematic of the carbon-based Continental Biosphere-Atmosphere-Cloud-Climate COBACC feedback loops. The temperature-related feedback loop is in green color, the GPP-related feedback loop in orange. CO_2 – concentration of carbon dioxide, GPP – gross primary production, BVOC – biogenic volatile organic compounds, SOA – secondary organic aerosol, CS – condensation sink, CCN – cloud condensation nuclei, R_d/R_g – fraction of diffuse radiation in global radiation, T – temperature.