Carnegie Mellon

Atmospheric data over a solar cycle: No connection between galactic cosmic rays and aerosol formation events

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4.8.2010 ECRS, Turku, Finland

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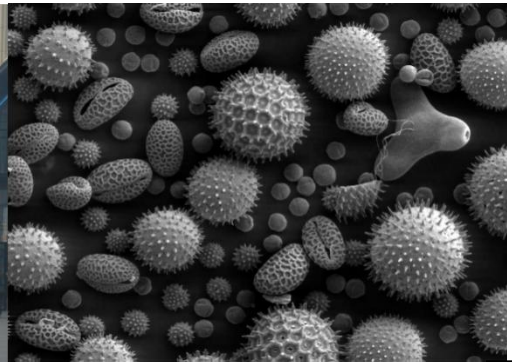
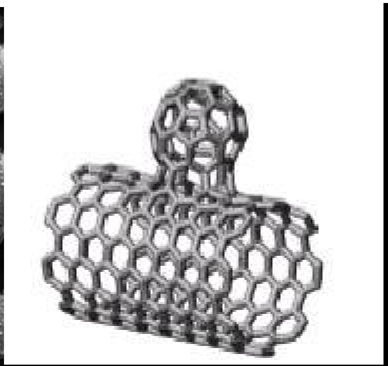
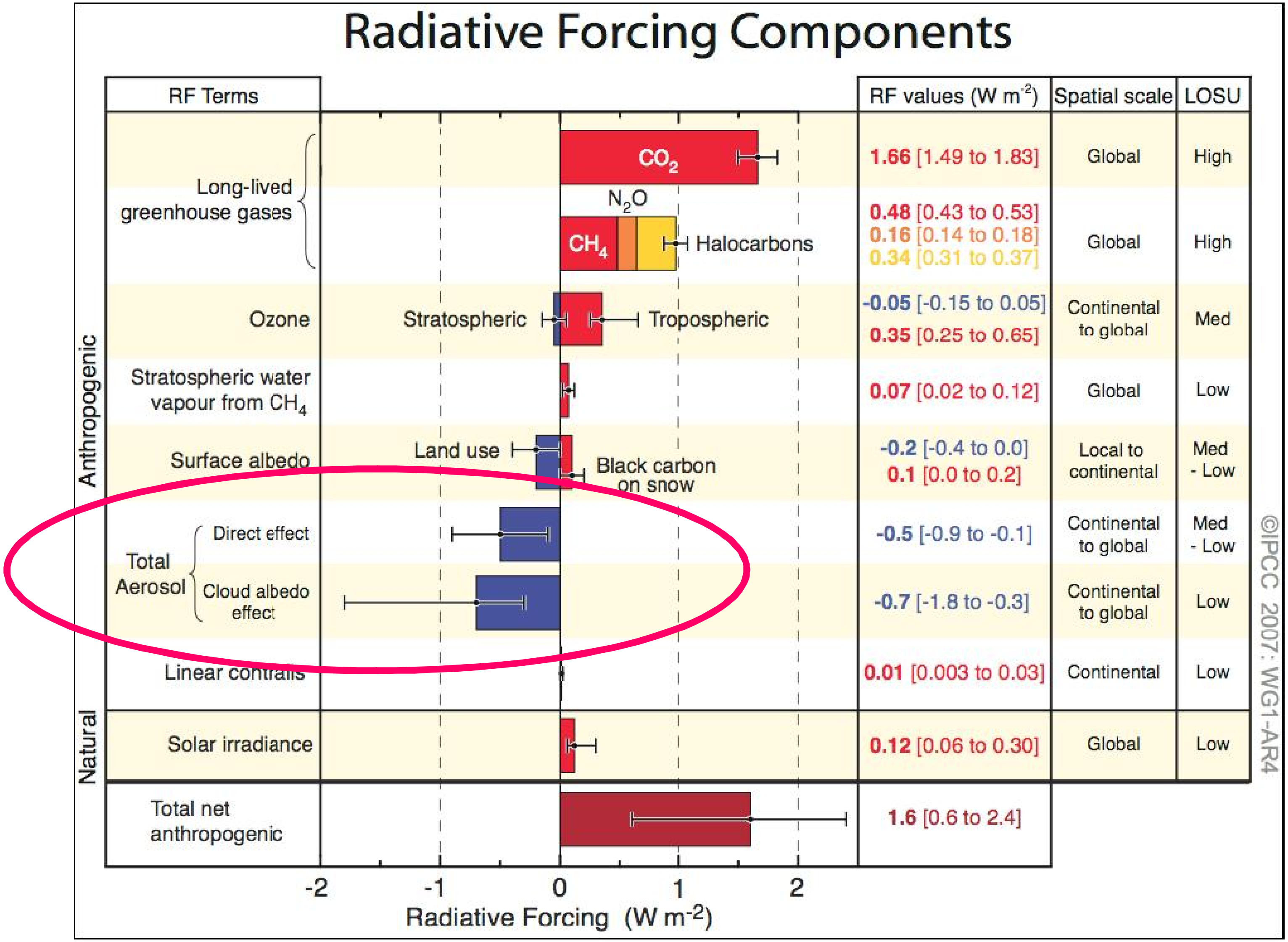
Atmospheric aerosols: Ubiquitous and variable

- Liquid or solid particles suspended in air
- Diameters ~10^{-9} – 10^{-4} m
- Concentrations ~10^{0} – 10^{5} cm^{-3} (10^{-1} – 10^{2} $\mu g\ m^{-3}$)

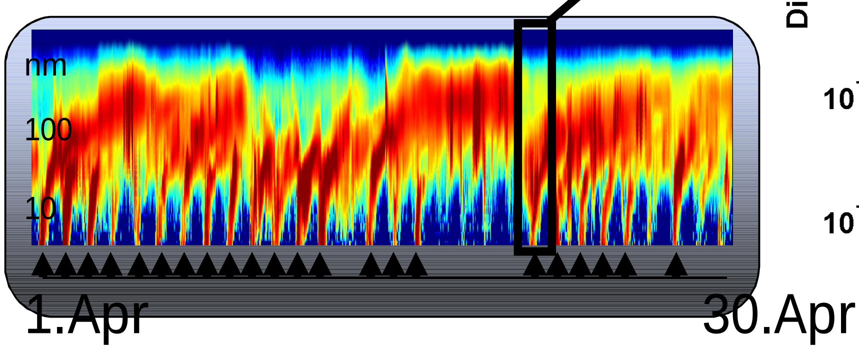
Aerosols are the largest single uncertainty in climate forcing calculations

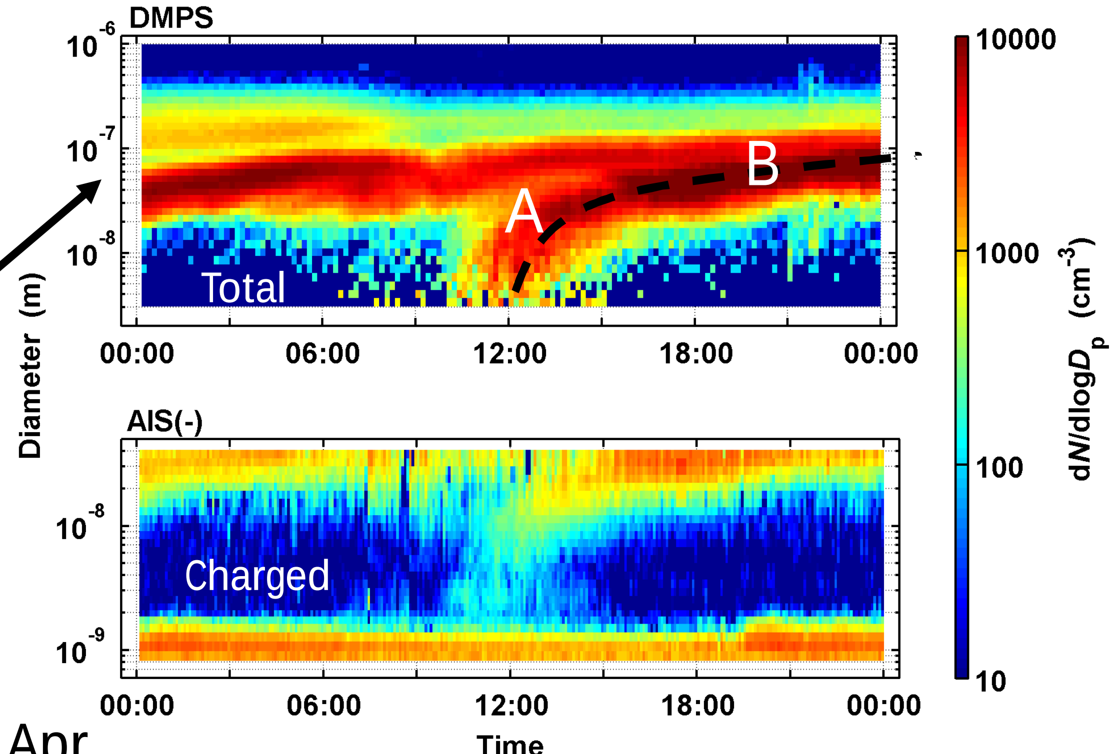


Particle formation events at the SMEAR II station in Hyytiälä, Finland

- Nucleation (A) + condensational growth (B) to climatically relevant sizes

- Persistence of the mode = regional phenomenon

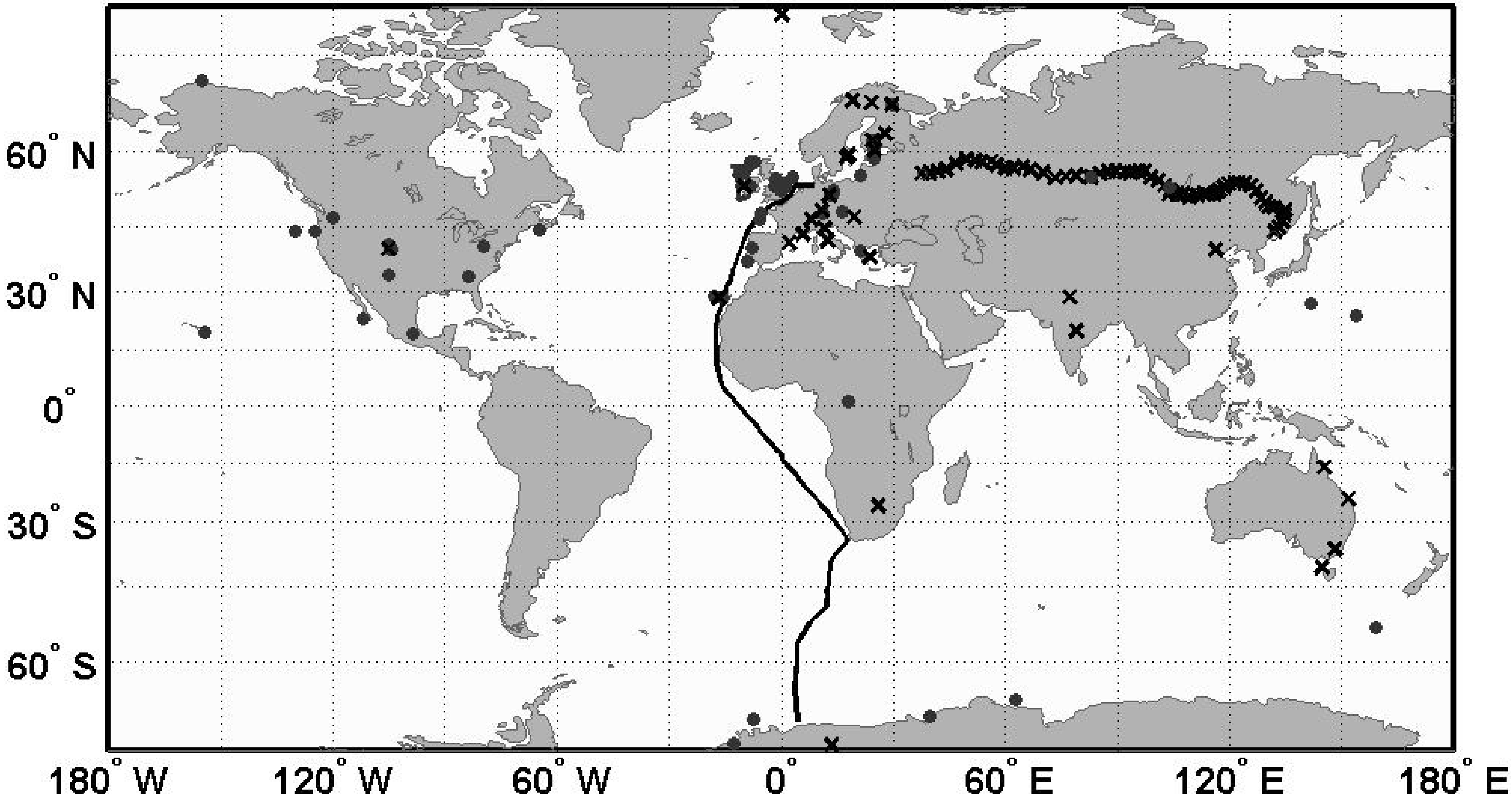




- Mechanistic understanding still incomplete

Nucleation is a significant source of atmospheric aerosol

- 30-75 % of continental particle numbers



Kulmala et al., *J. Aerosol Sci.* 2004; Merikanto et al., *Atmos. Chem. Phys* 2009

Are atmospheric nucleation and galactic cosmic rays connected?

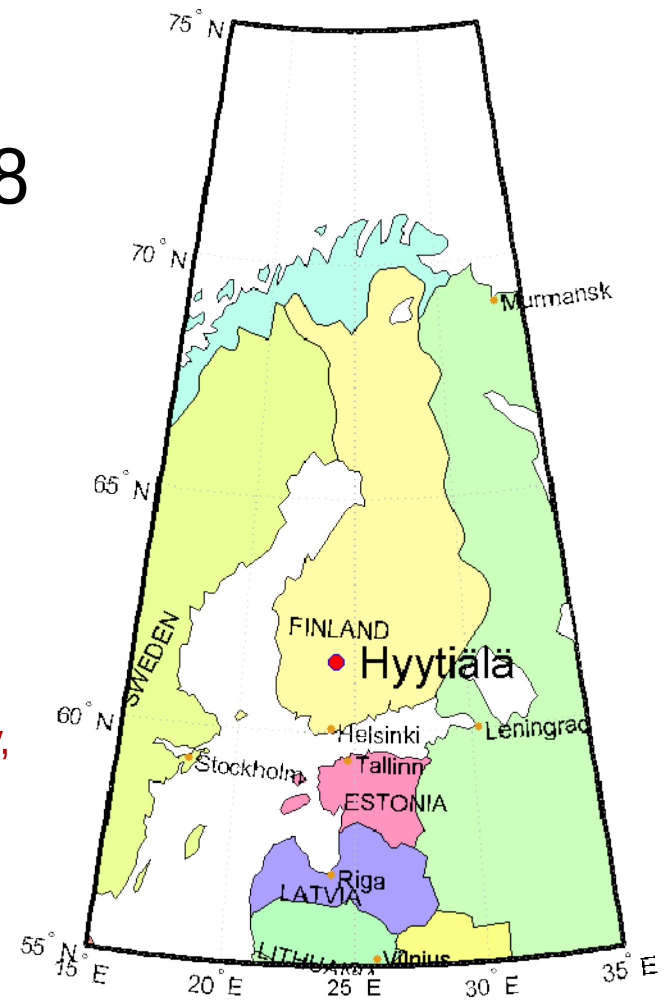
- Ion-induced or -mediated nucleation proposed as a major mechanism of atmospheric nucleation (e.g. Yu and Turco, *GRL* 2000; Yu, *ACP* 2006)
  - Charge helps stabilize sulfuric acid/water/ammonia clusters (e.g. Winkler et al., *Science* 2008)

- Indications of correlation between low level clouds and galactic cosmic rays (Svensmark and Friis-Christensen, *J. Atmos. Sol. Terr. Phys*. 2006)
  - Production of CCN by ion-induced nucleation?

- Do we find indications of cosmic rays controlling atmospheric nucleation?

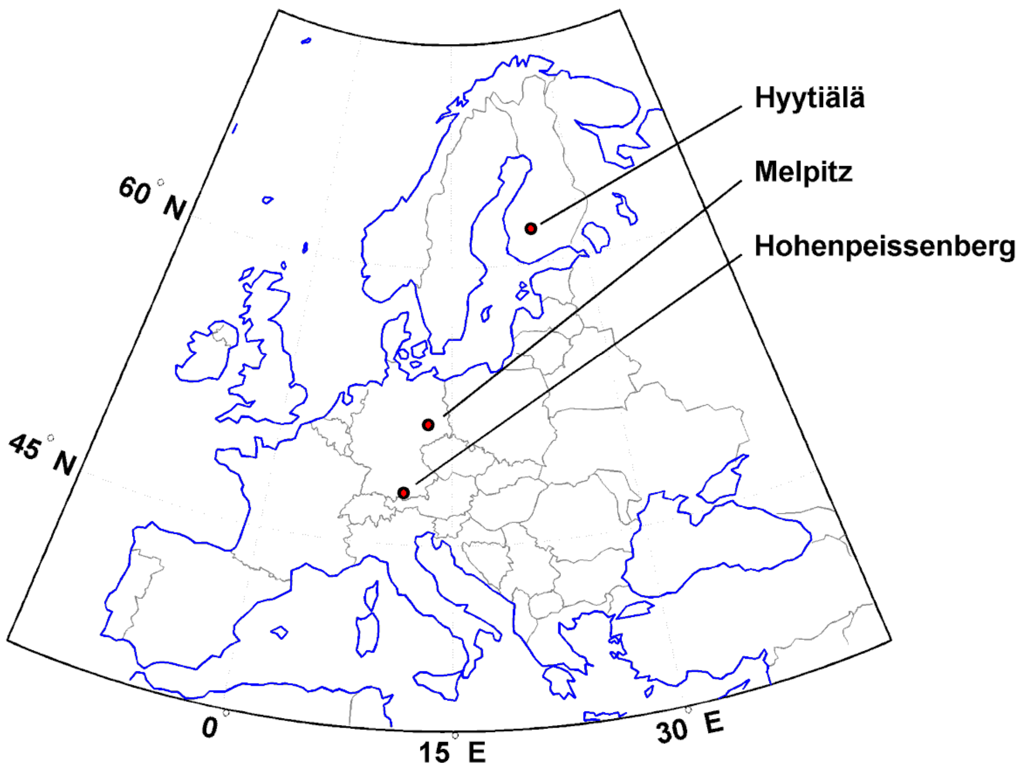
Approach 1: 13 years of aerosol formation data vs. CRII

- Continuous aerosol size distribution measurements in Hyytiälä, Finland, years 1996-2008
  - Differential Mobility Particle Sizer (DMPS) instrument
- Cosmic Ray Ionization Intensity (CRII) time series calculated with the CRAC:CRII model (Usoskin and Kovaltsov, *JGR* 2006)
  - Comparison between CRII and particle formation



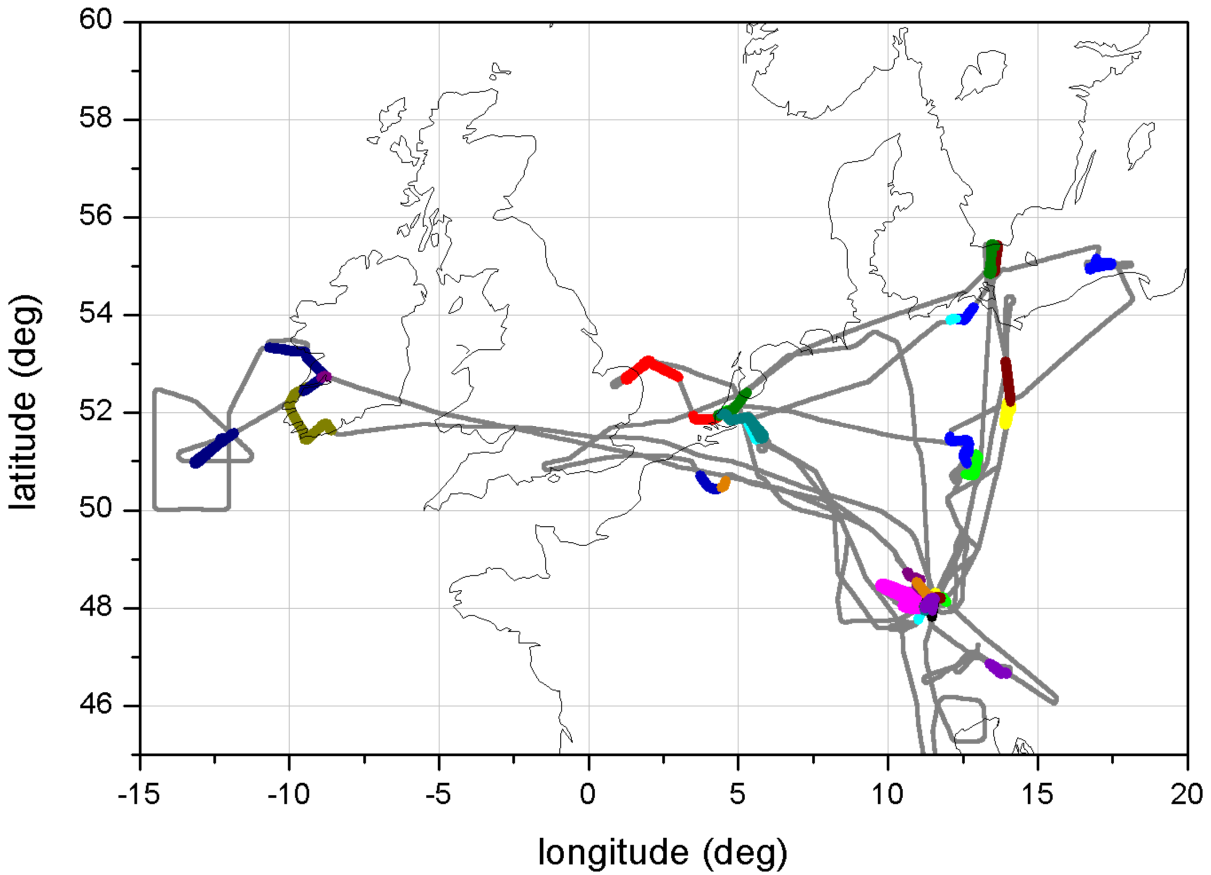
Approach 2: Ion-induced fraction of atmospheric nucleation in Europe

- Charged and total aerosol size distributions at 3 European sites
  - (Neutral cluster and) Air Ion Spectrometers (N)AIS
  - Ion-induced fraction of particle formation rates



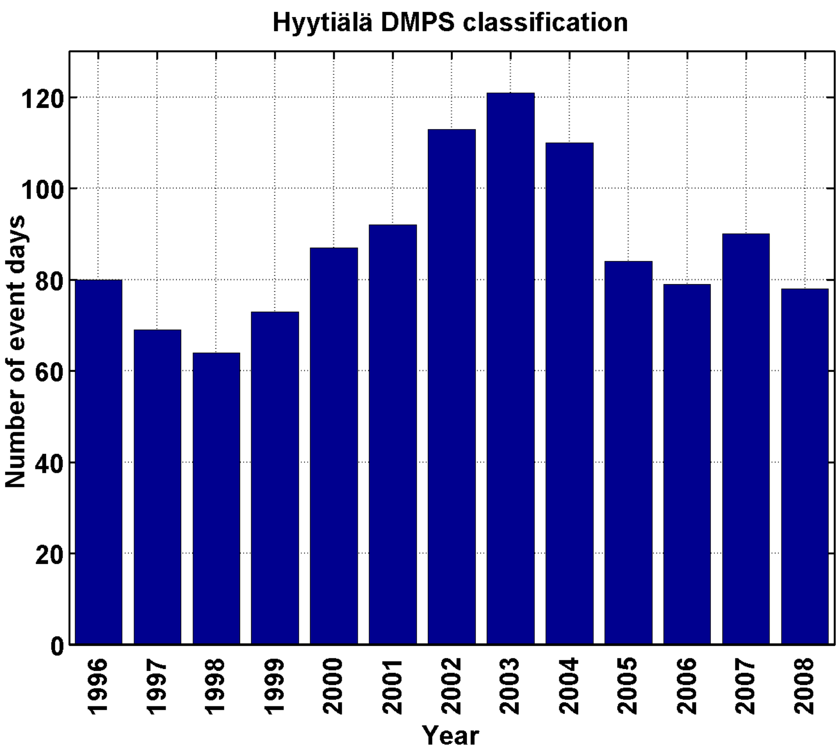
Approach 3:Vertical profiles of charged and neutral particles over Europe

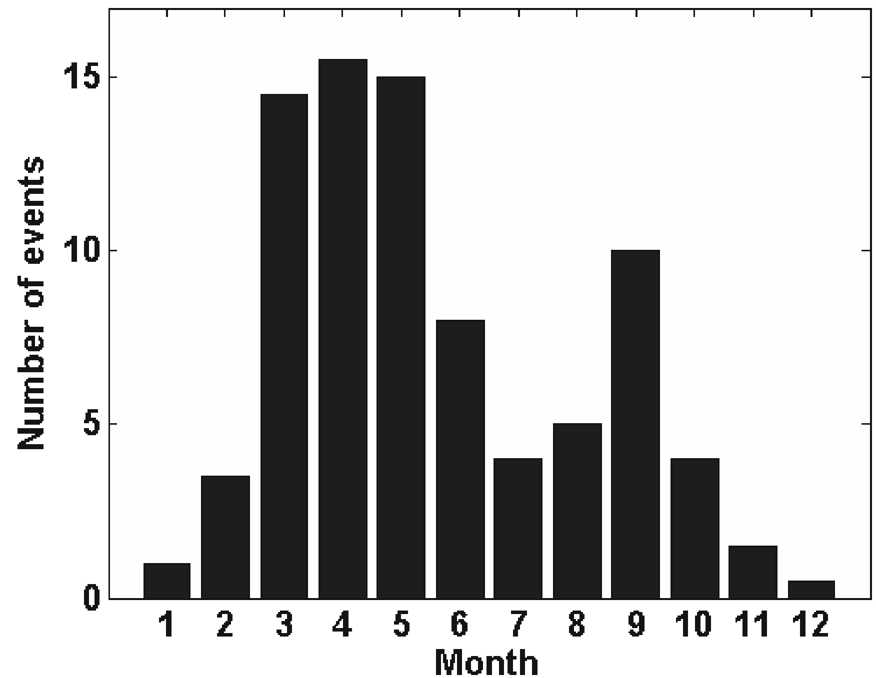
- Airborne NAIS measurements of aerosol and ion size distributions over Central Europe



Results: Aerosol formation in Hyytiälä, 1996-2008

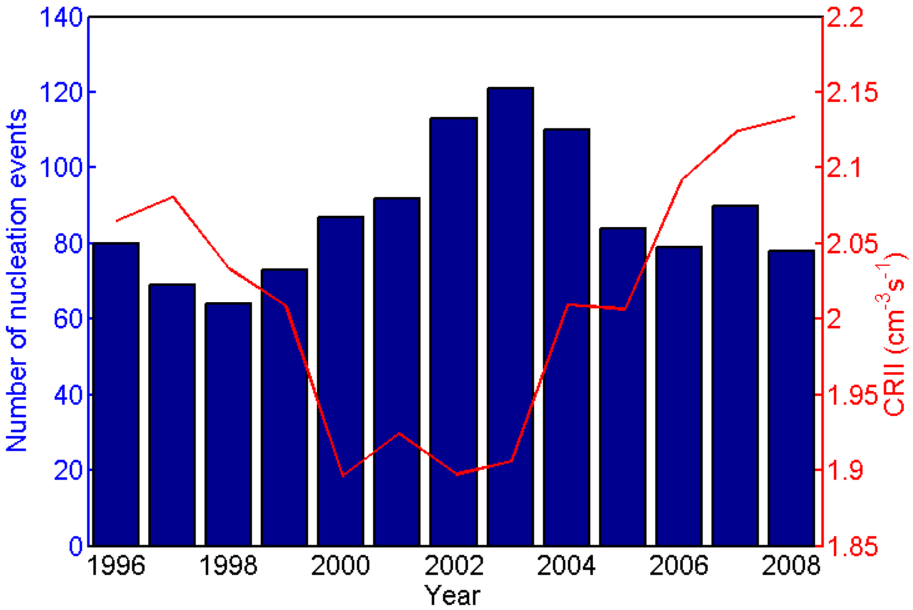
- Clear seasonal and annual trends

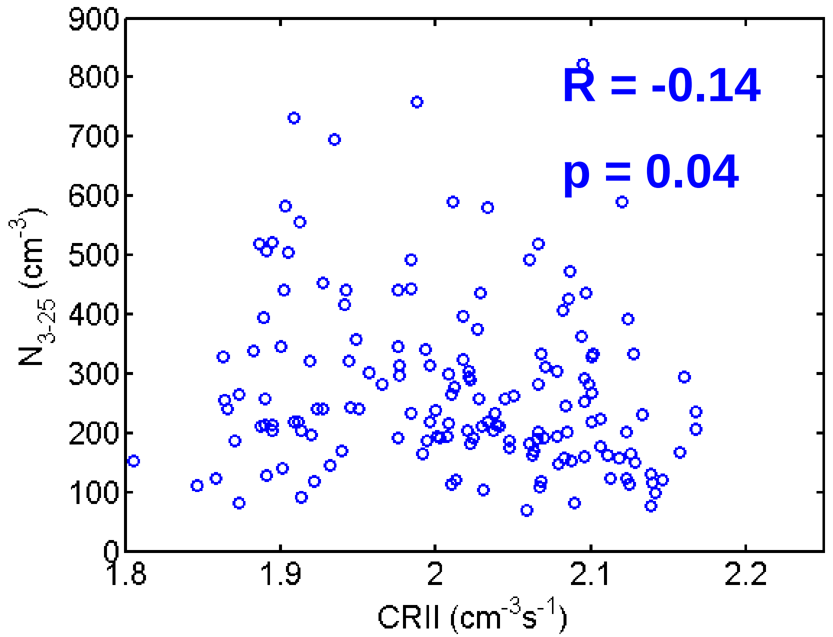




Aerosol formation in Hyytiälä 1996-2008: No correlation with CRII

- Similar results in all particle size classes



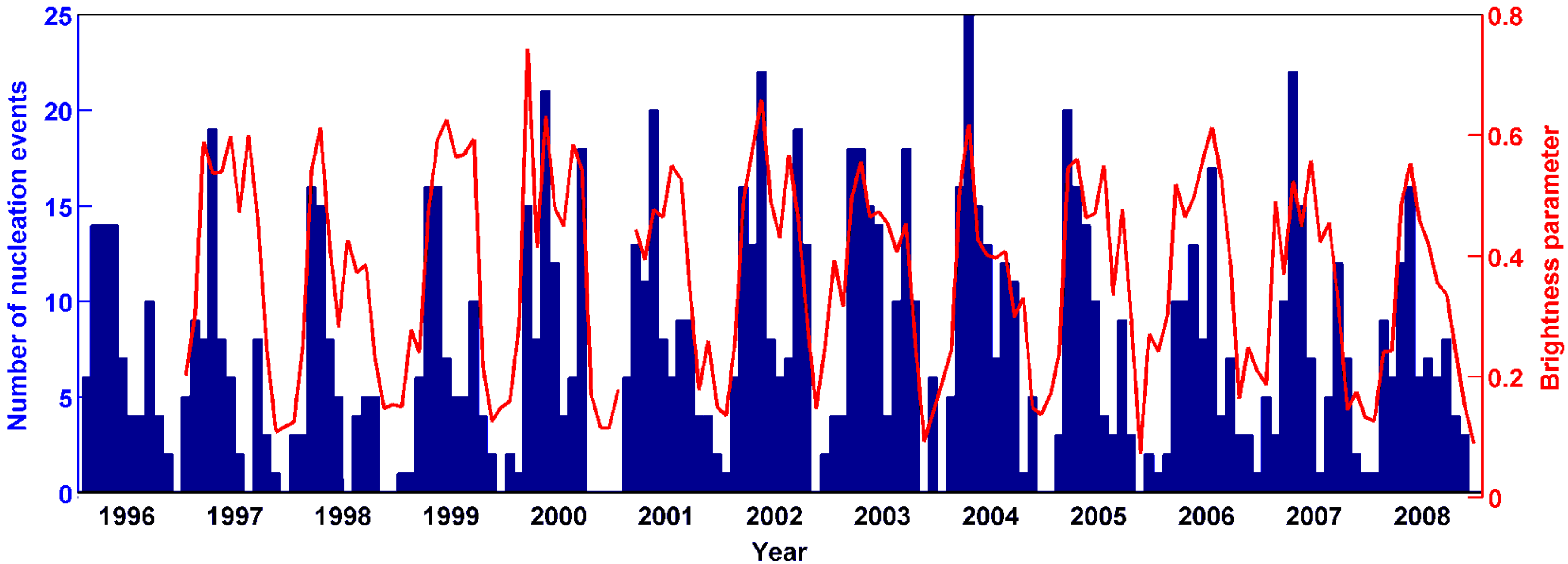


- No clear connection with geomagnetic activity either

Brightness and nucleation events in Hyytiälä

- Brightness parameter

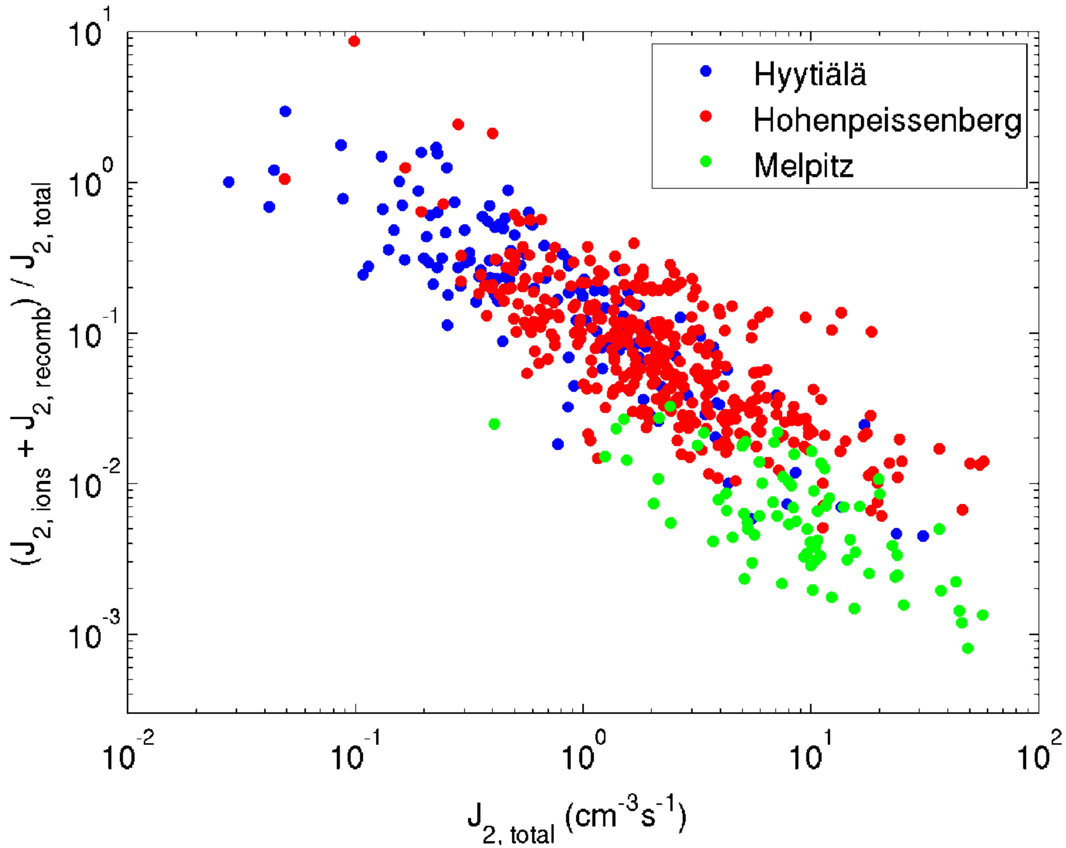
$$P = \int^{16} R_{obs} \Big/ \int^{16} R_{theor}$$



- $R = 0.60$, $p < 10^{-10}$
- Related to photochemical production of condensable vapors

Ion-induced fraction of atmospheric nucleation at 3 European sites

- Ion-induced fraction typically < 10 %, ions favored at cleaner conditions

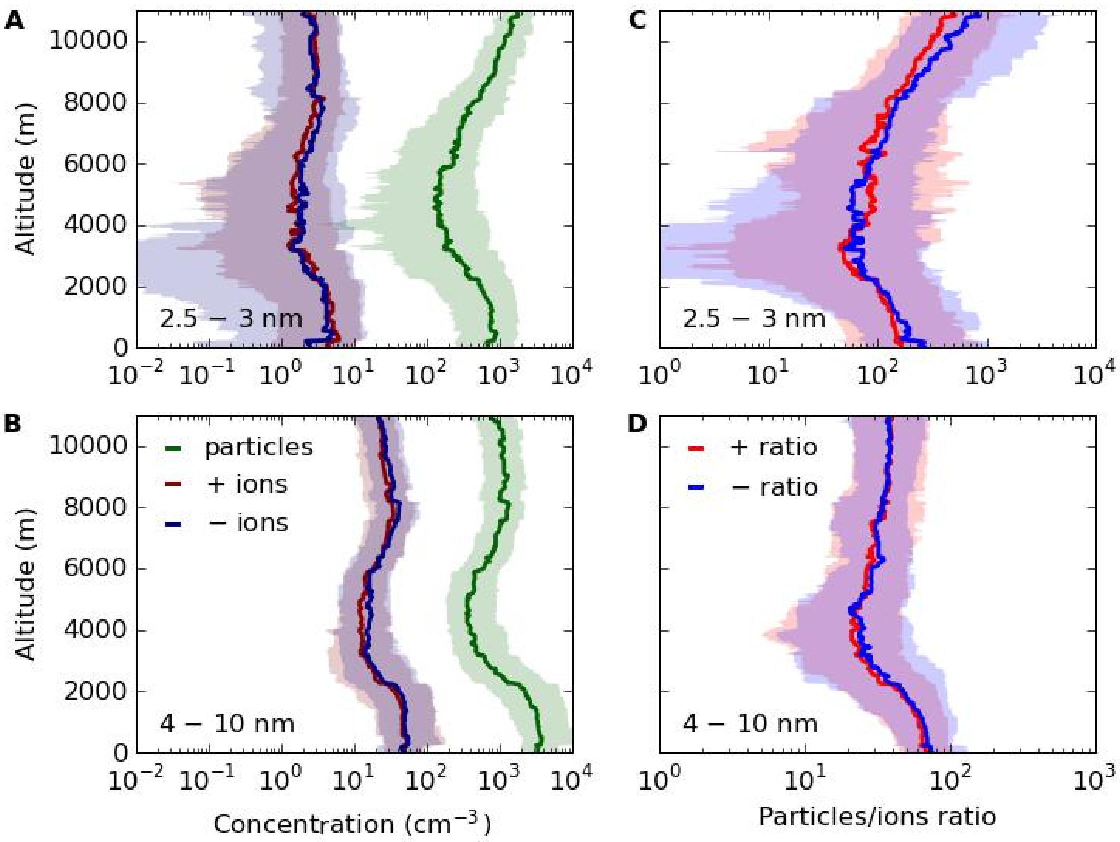


- Consistent with the lacking correlation with CRII

Iida et al., *JGR* 2006; Manninen et al., *ACP* 2009

Vertical profiles of ions and neutral particles

- No significant variation inside troposphere
  - More data needed!



Mirme et al., *ACP* 2009

Conclusions and future directions

- Cosmic rays do not seem to control atmospheric nucleation – at least not in Hyytiälä
  - No correlation with geomagnetic activity either

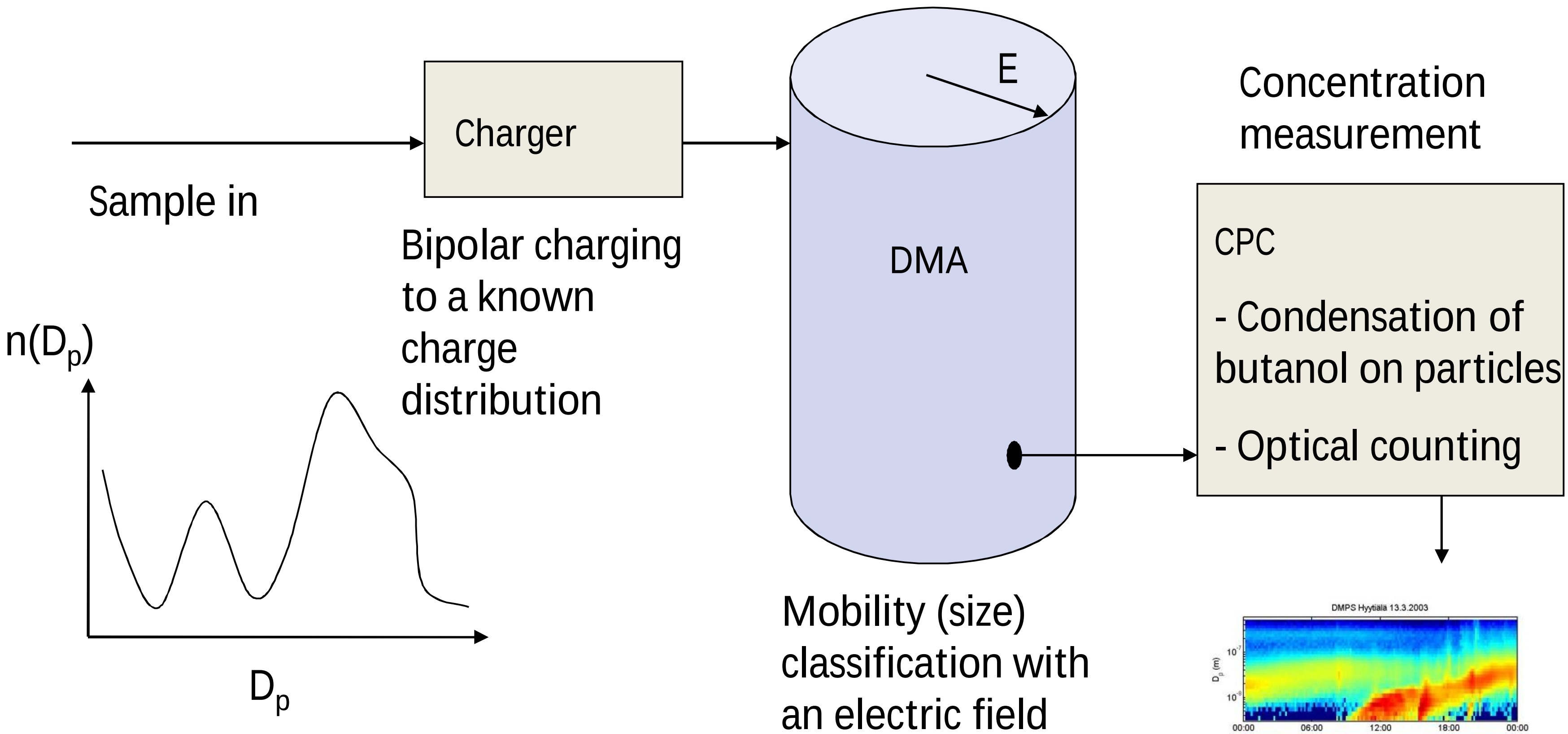
- Neutral mechanisms dominate over ion-induced nucleation in continental Europe
  - Ion-induced fraction usually < 10%
  - No clear signs of increasing ion-induced fraction with altitude
  - Availability of condensable vapors more important
  - Other sites / altitudes / pristine atmosphere?

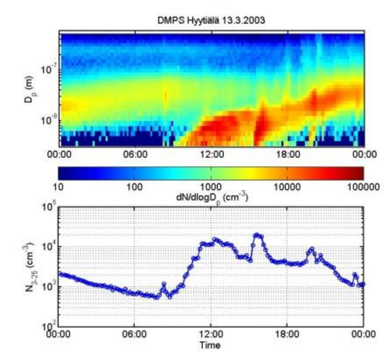
- Other connections between cloudiness and cosmic rays?

For details of this work, see e.g. Kulmala *et al.*, *Atmos. Chem. Phys.* 10, 1885-1898, 2010.

Extra slides

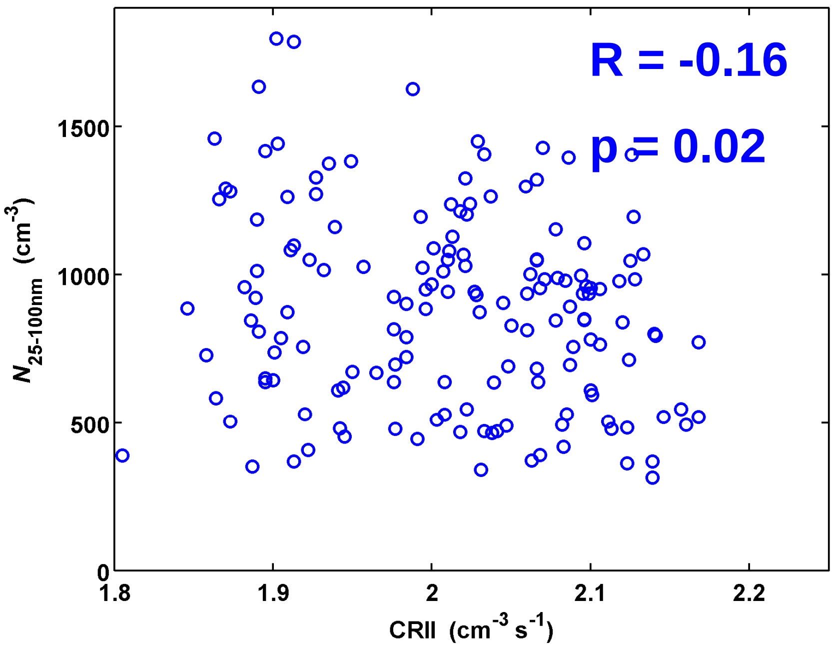
The DMPS system at the SMEAR II: particle size distribution measurements since 1996

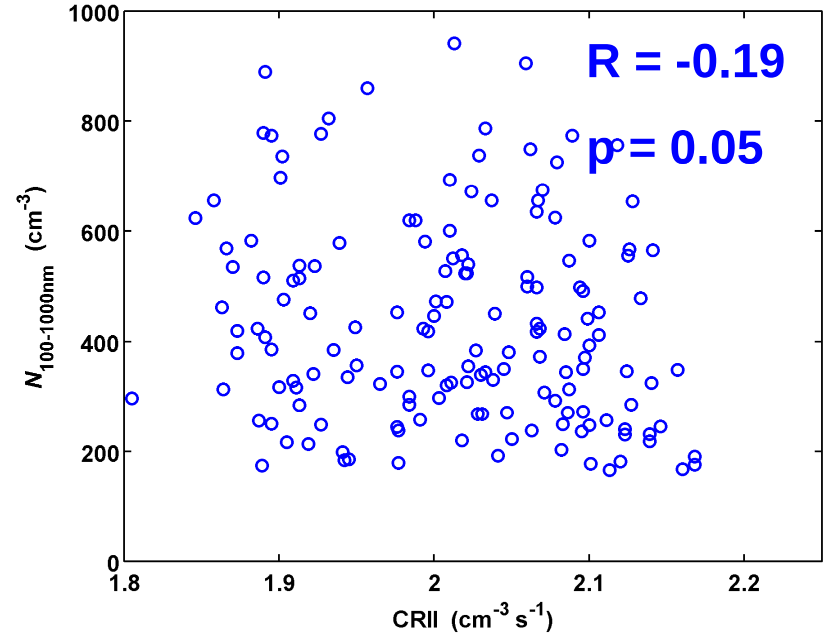




- Size range 3 – 1000 nm
- Time resolution 10 min

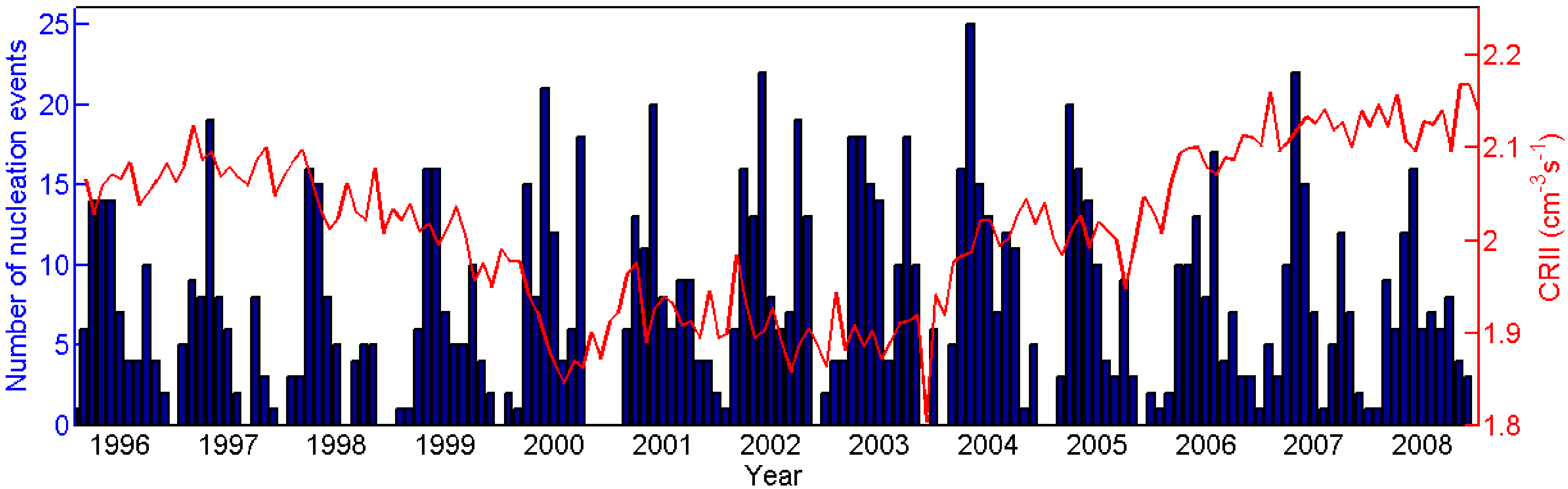
Cosmic radiation and particle number





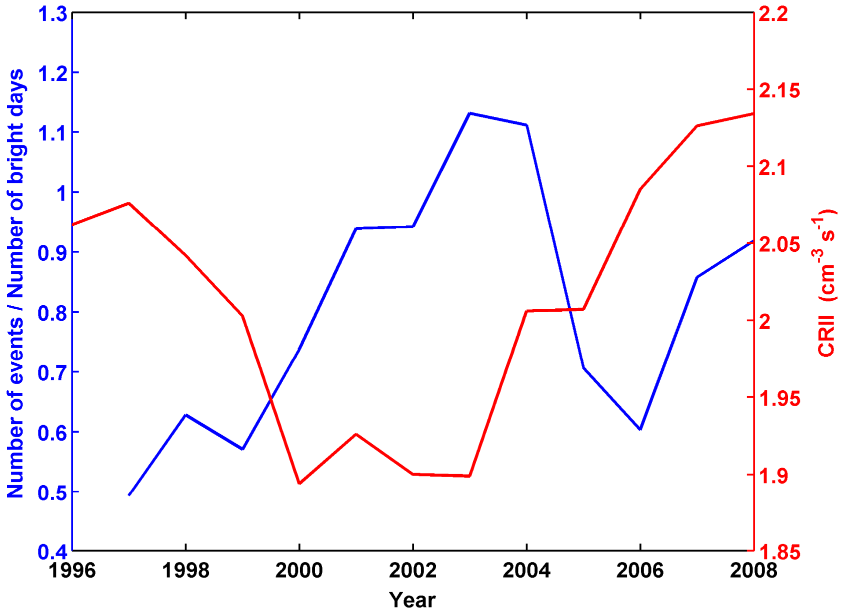
- Similar results for geomagnetic activity

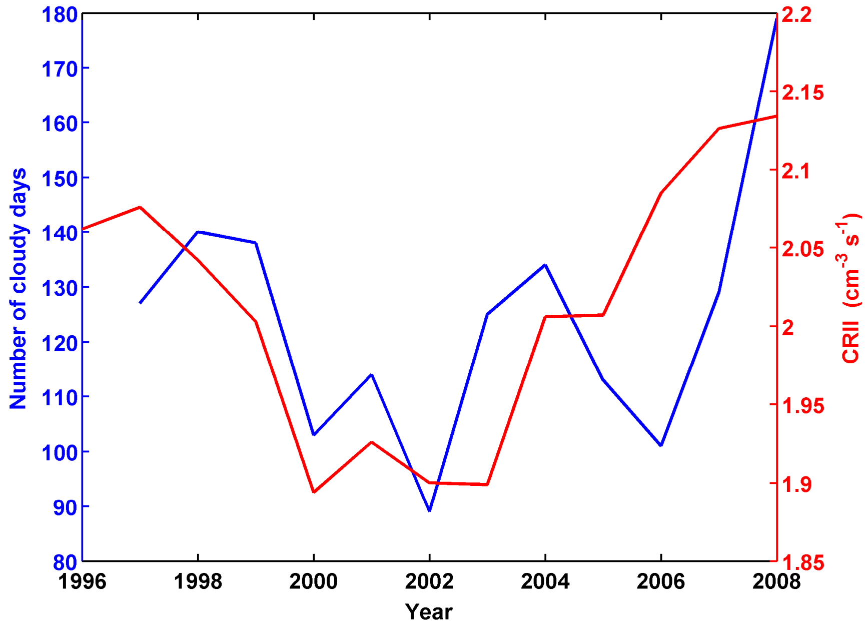
Cosmic radiation and nucleation events, monthly numbers



- CRII shows no signs of controlling NPF or particle numbers

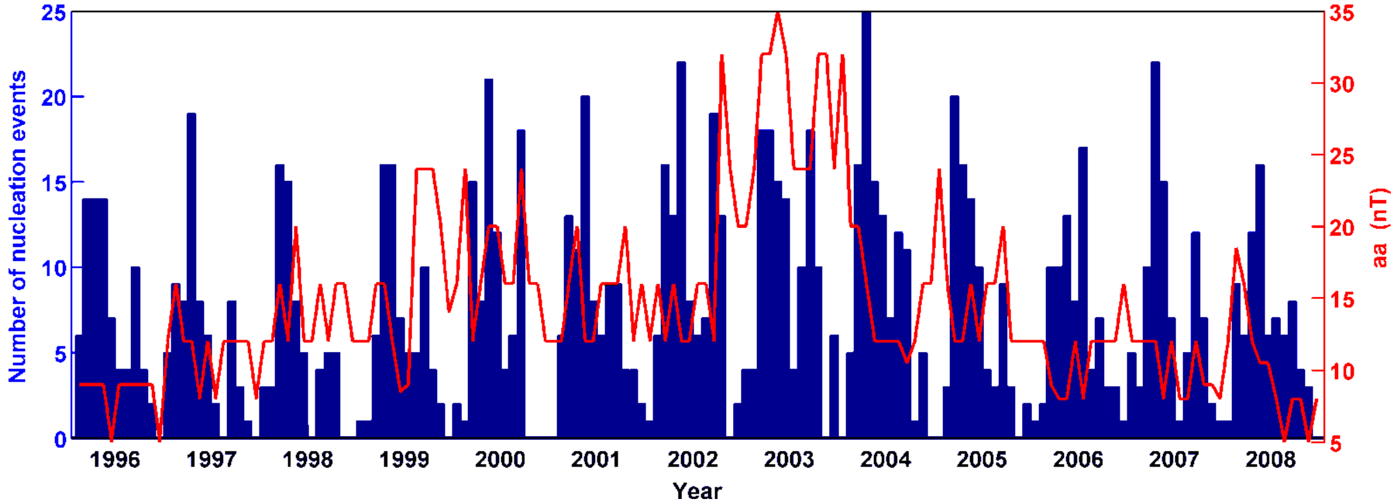
Cosmic radiation and cloudiness in Hyytiälä

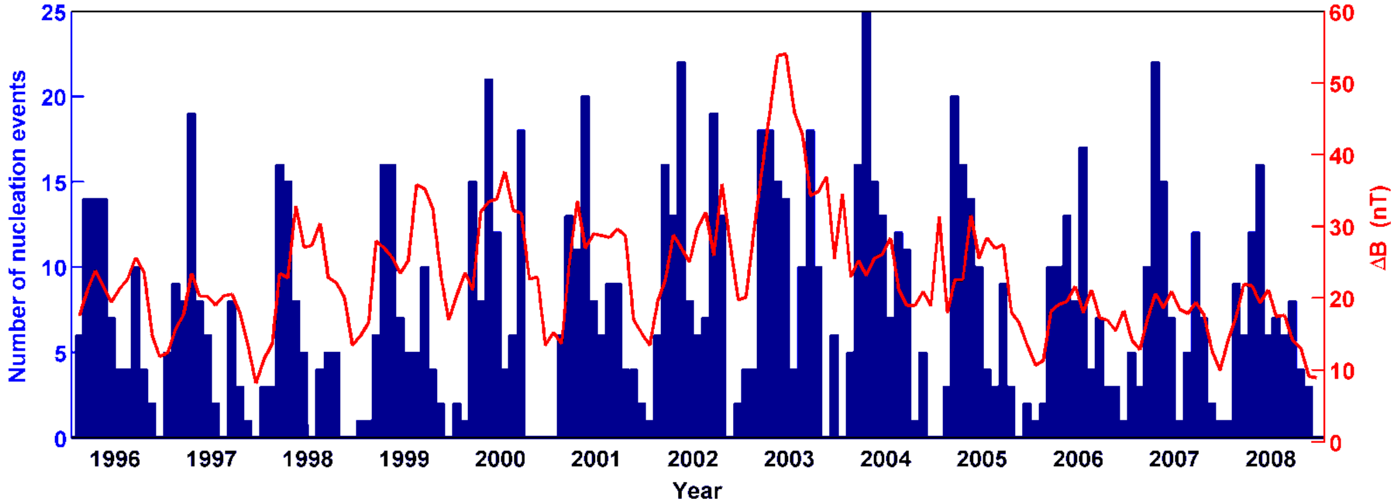




- Connection cannot be ruled out – no significant correlation found though

Nucleation and geomagnetic activity



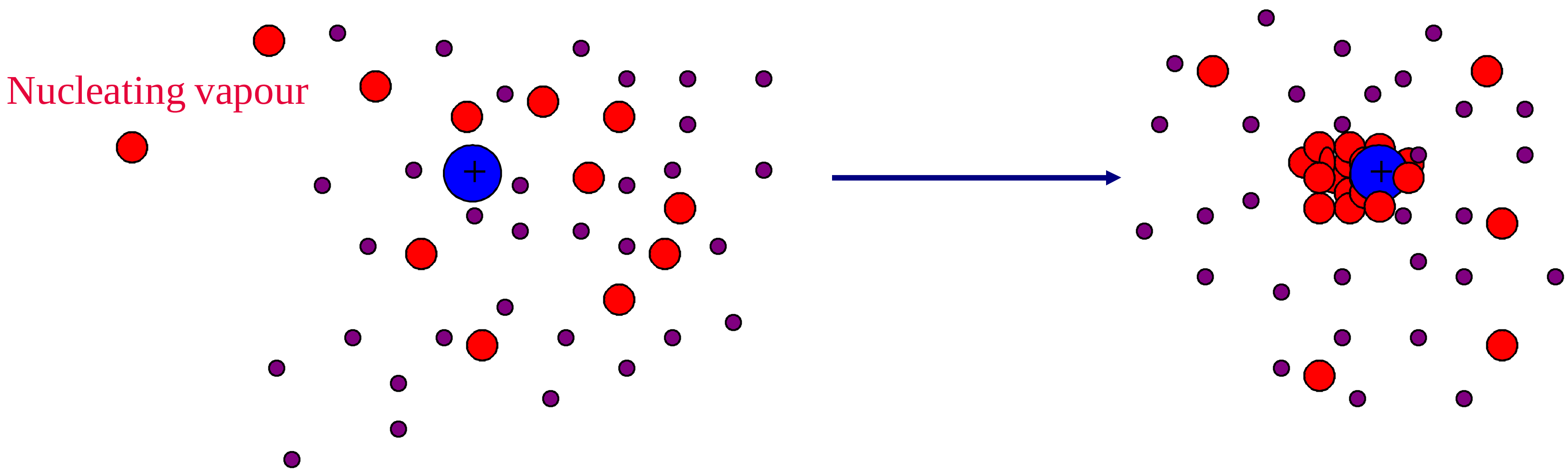


Binary correlation coefficients

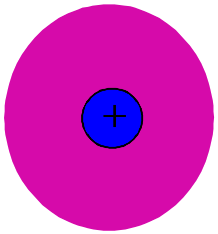
	Number of nucleation events	Number of events/ number of bright days	CRII (cm^{-3} s^{-1})
CRII ($cm^{-3}s^{-1}$)	-0.14 (0.09)	-0.04 (0.69)	-
Brightness parameter	0.60 (<10^{-10})	-	-0.09 (0.27)
aa (nT)	0.11 (0.15)	0.20 (1.9×10^{-2})	-0.65 (<10^{-10})
Kp	0.14 (0.09)	0.13 (0.12)	-0.68 (<10^{-10})
ΔB (nT)	0.38 (1.1×10^{-6})	0.10 (0.27)	-0.65 (<10^{-10})
J_3 (cm^{-3} s^{-1})	0.39 (1.5×10^{-5})	0.013 (0.90)	0.05 (0.63)
GR (nm h^{-1})	-0.11 (0.23)	-0.17 (0.08)	-0.08 (0.41)
$N_{3\text{-}25nm}$ (cm^{-3})	0.69 (<10^{-10})	0.19 (0.03)	-0.16 (0.04)
$N_{25\text{-}100nm}$ (cm^{-3})	0.66 (<10^{-10})	-0.03 (0.73)	-0.19 (0.02)
$N_{100\text{-}1000nm}$ (cm^{-3})	0.11 (0.18)	-0.35 (3.7×10^{-5})	-0.16 (0.05)

Nucleation around electrically charged molecules or particles

- Electrostatic charge-dipole interaction induces cluster formation



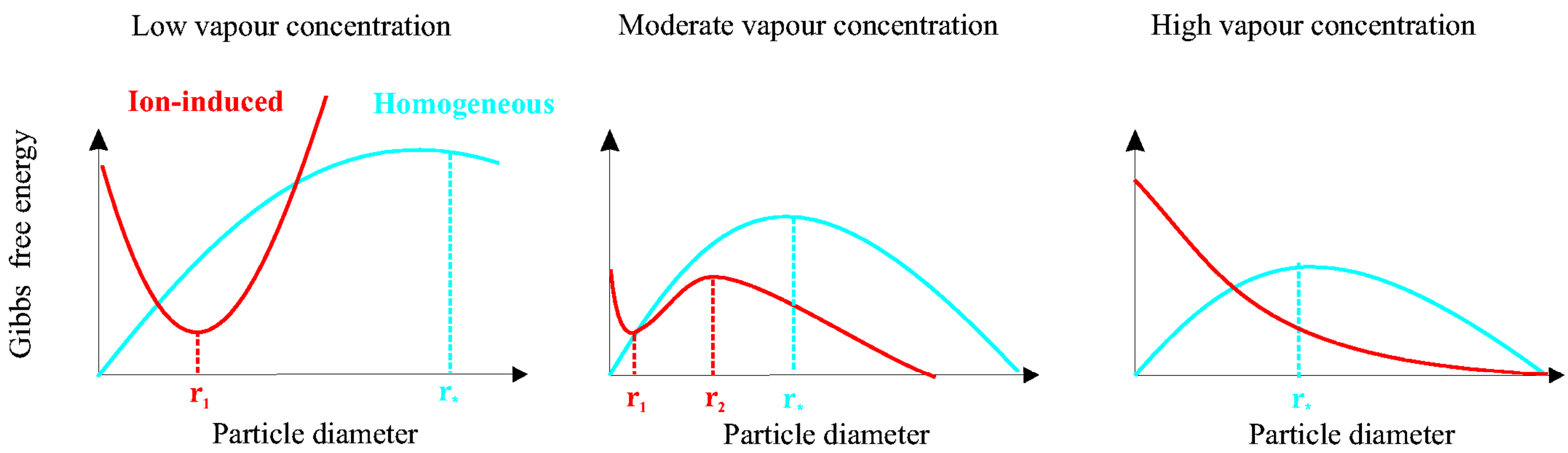
$$\Delta G = -kT\ln S + A\sigma + \frac{q^2}{8\pi\varepsilon_0}\left(1-\frac{1}{\varepsilon_r}\right)\left(\frac{1}{r}-\frac{1}{r_{ion}}\right)$$



ε_0 permittivity of vacuum, ε_r dielectric constant

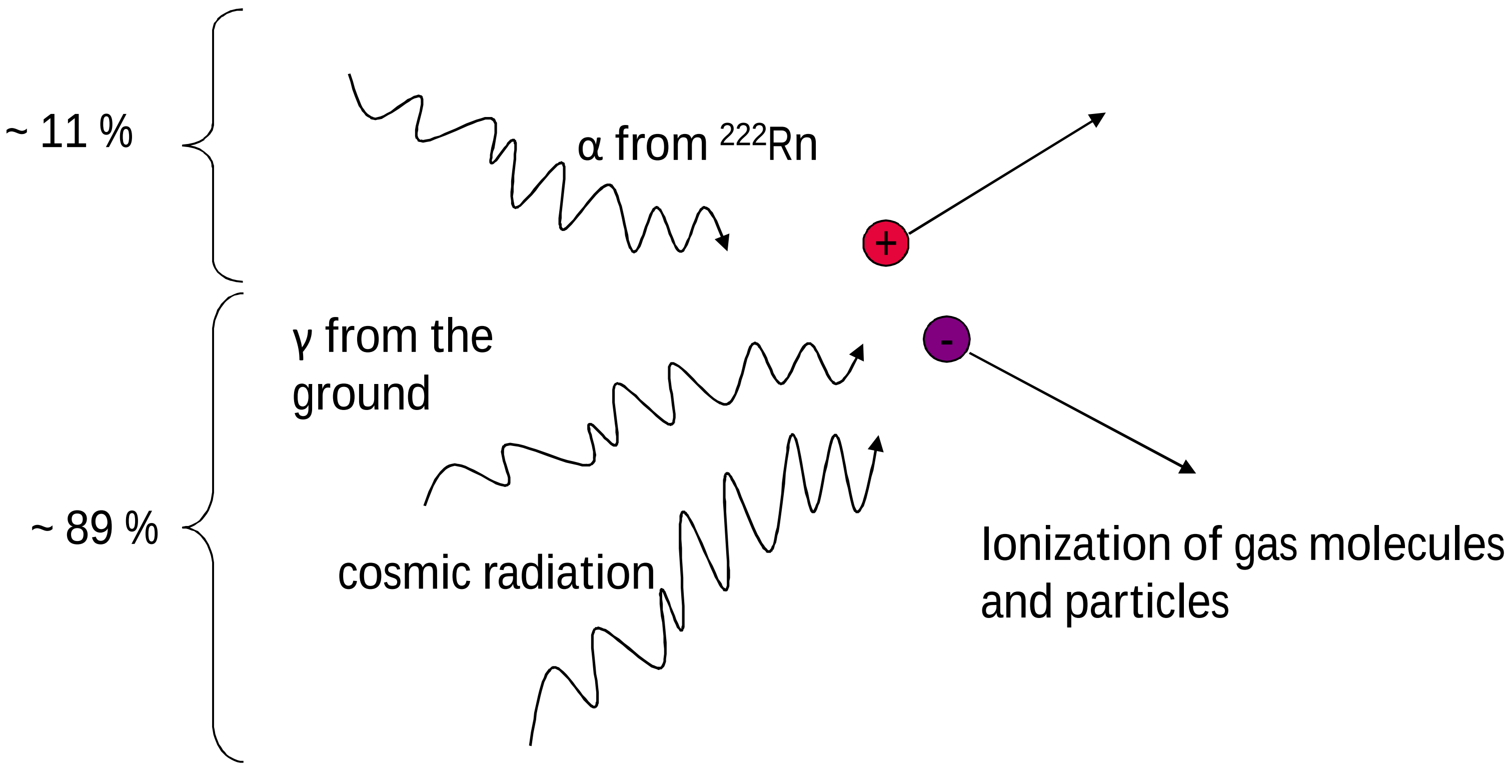
Free energy curve has a minimum and a maximum for moderate S

- Nucleation barrier is the difference in free energies between the minimum and the maximum



Atmospheric ion production in Hyytiälä

- Total ionization rate in Hyytiälä 4.2 – 17.6 ion pairs $cm^{-3}\ s^{-1}$ (mean ~ 6 ion pairs $cm^{-3}\ s^{-1}$)



Hirsikko et al., *BER* 2007

Ion spectrometers, AIS and BSMA

(Mirme et al., *BER* 2007; Tammet, *Atmos. Res.* 2006)

- Sizes even below 1 nm can be reached
  - No charging required – measure naturally charged fraction
  - High flow rates can be used (particle counting with electrometers)

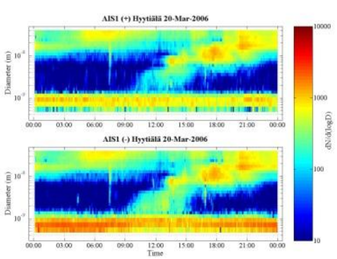
Concentration measurement

Sample in

DMA / Capacitor

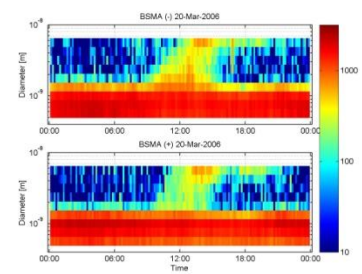
Series of electrometers / Electrometer

Mobility classification



AIS:

0.8 – 40 nm



BSMA:

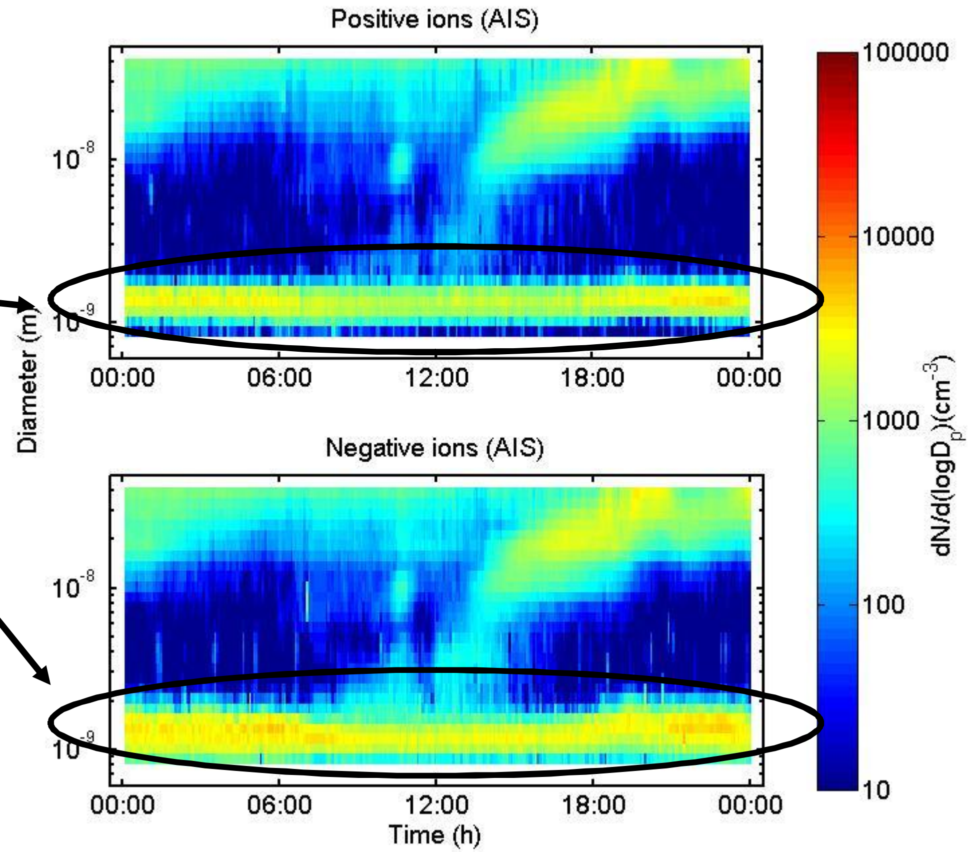
0.8 – 7 nm

Size distributions of charged atmospheric particles in Hyytiälä

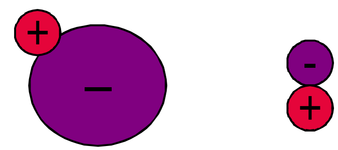
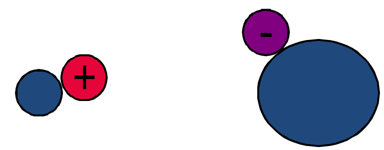
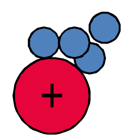
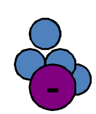
- Concentrations ~1- 10 % of total concentrations

Ion clusters,

~ 100 – 1000 cm^{-3}

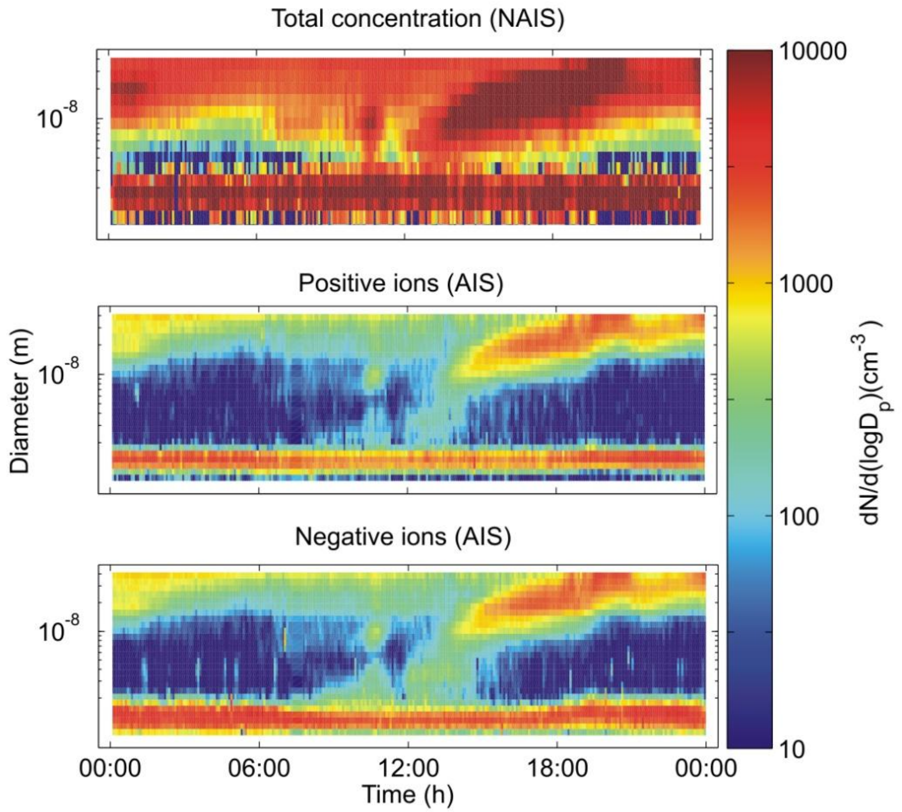


Air ion / charged particle dynamics

Recombination / neutralisation	
Charging / coagulation	
Condensational growth	
Nucleation	

How to assess the importance of ion-related nucleation mechanisms?

- Measurements on both neutral and charged particles now possible: NAIS, newly-developed CPCs



- Comparison of total vs. charged particle concentrations

Kulmala et al., *Science* 2007; Sipilä et al., *Science* 2010