National Institute for Research in Amazonia - INPA

Is size distribution or chemical composition the key parameter to aerosol act as CCN?

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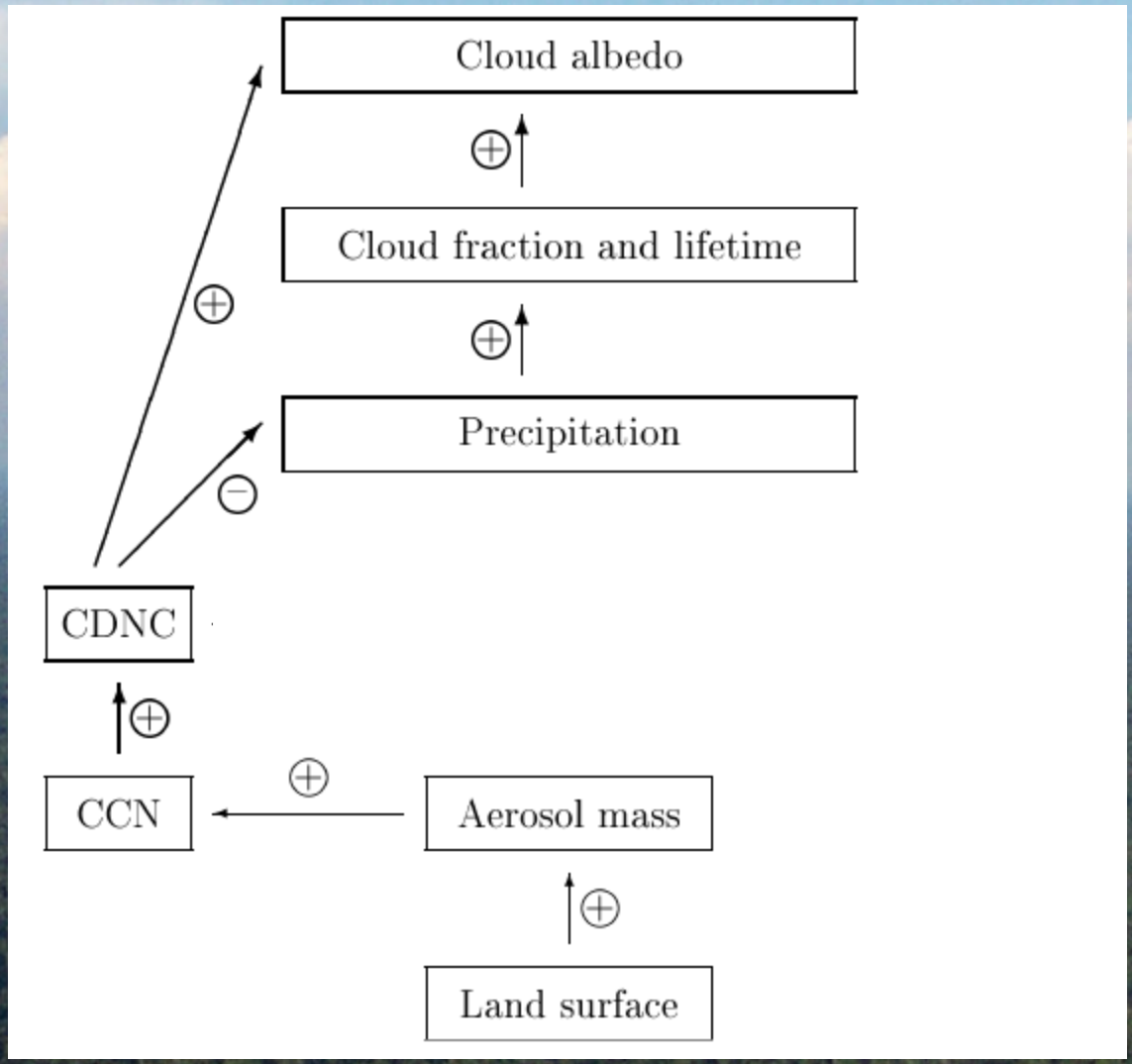
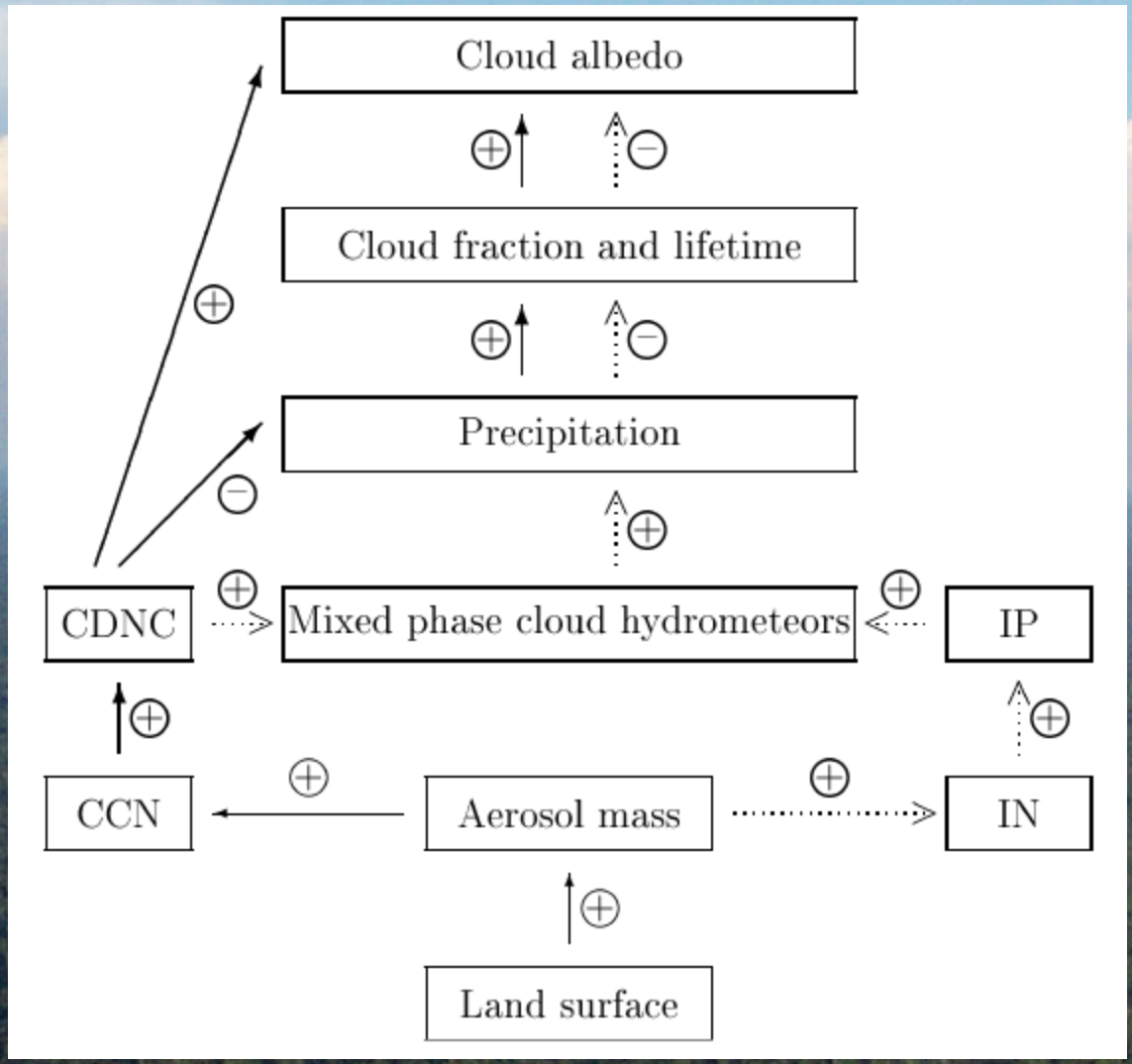
CCN acts on…

- Cloud droplet formation;
- Rainwater chemistry (in-cloud processes);
- Delaying or not precipitation in warm clouds;
- Terrestrial radiative balance through clouds albedo
- Hydrological cycle (?);
- Ice nuclei (?)

Feedbacks in warm clouds…



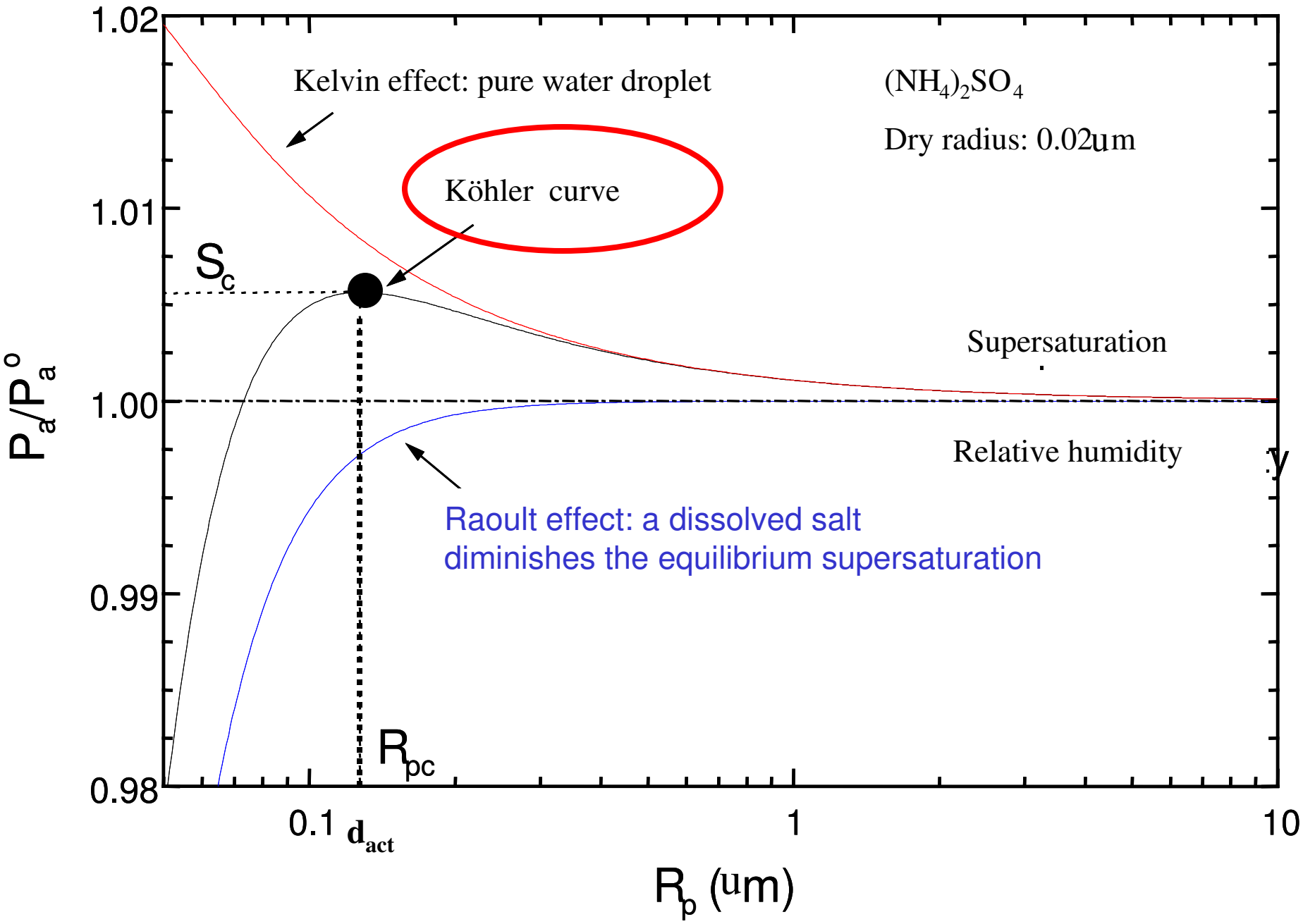
…become more complex in mixed phase clouds.



The question

- What properties are critical for the nucleation ability of aerosol?
  - Size distribution?
  - Chemical composition?
  - Updraft velocities?
  - Maximum supersaturation reached incloud?

Köhler theory on aerosol particle nucleation



Köhler equation for a single particle

$$\frac{e}{e_{sat}} = \exp\left[\frac{A}{D_p} - \frac{B}{D_p^3}\right]$$

$$A = \frac{M_w \sigma_w}{RT\rho_w} \qquad B = \frac{6\nu n_s M_w}{\pi \rho_w}$$

Chemical information

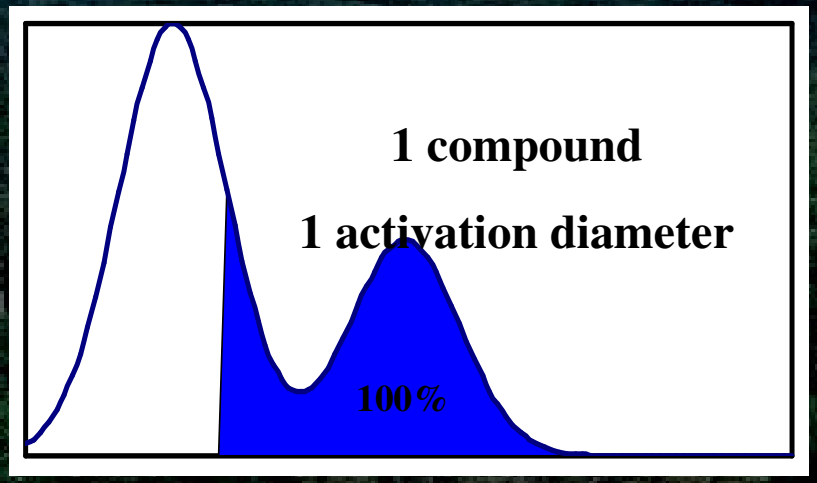
Term A (Kelvin effect)

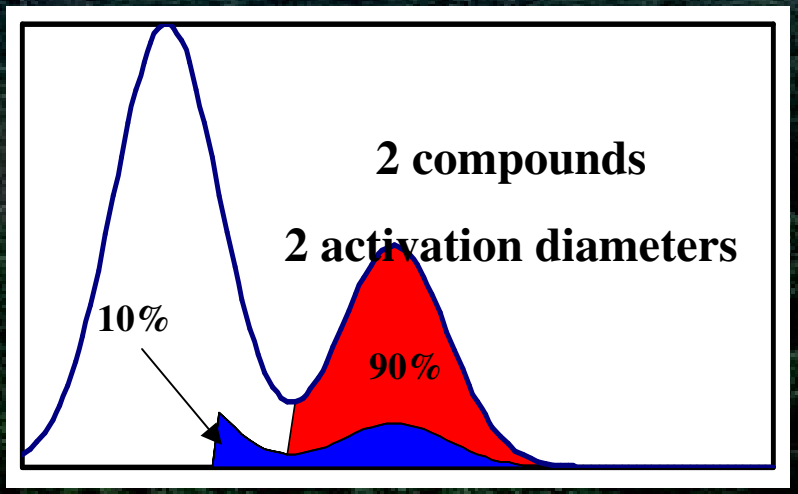
e/e_{sat}: vapor ratio
M_w: Molecular weight of water
σ_w: surface tension between air and water
ρ_w: water density
R: Universal gas constant
T: temperature,

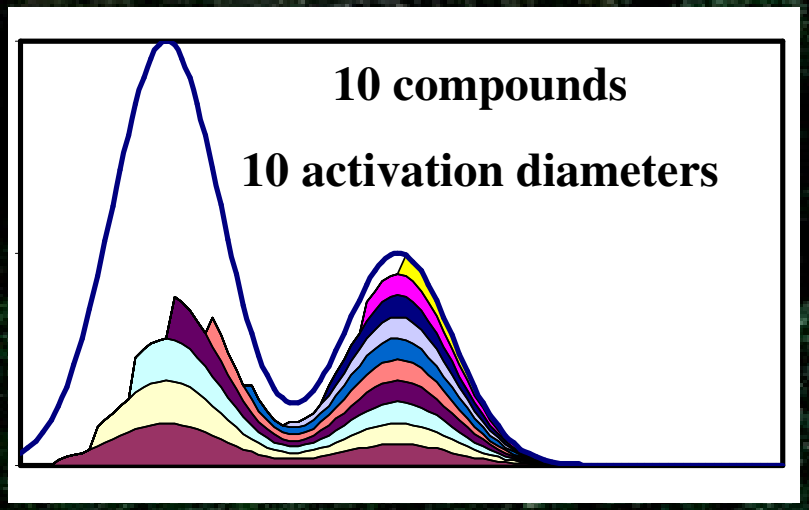
Term B (solute [or Raoult] effect)

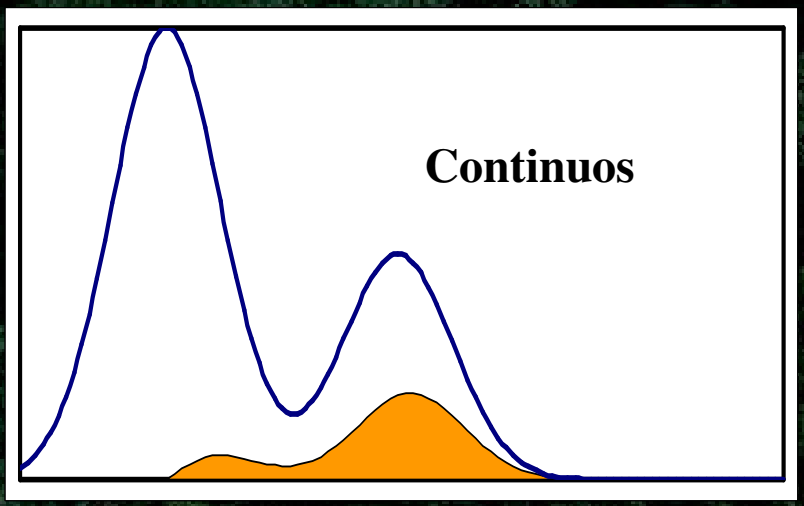
ν: number of ions result of the dissociation of a single solute molecule (van't Hoff's factor)
n_s: number of solute molecules

The role of chemical composition of aerosols

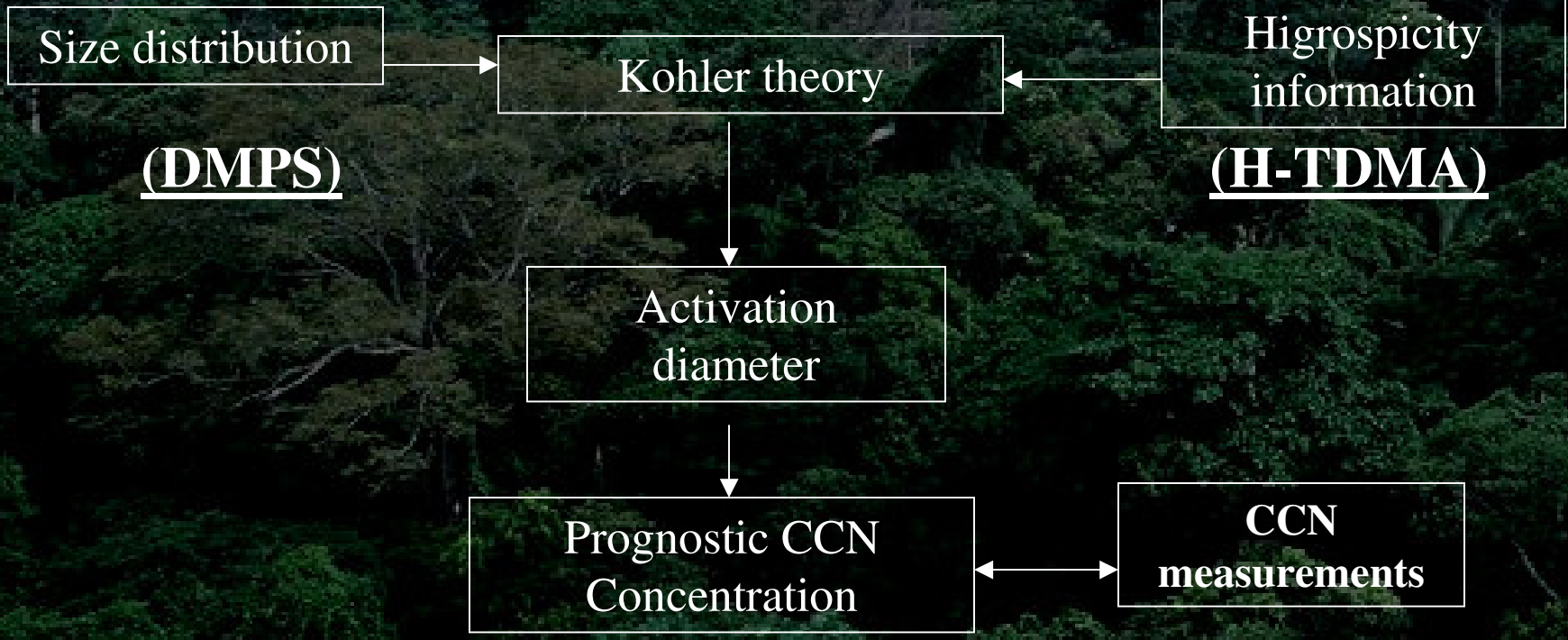








Building a closure relationship to estimate CCN concentration

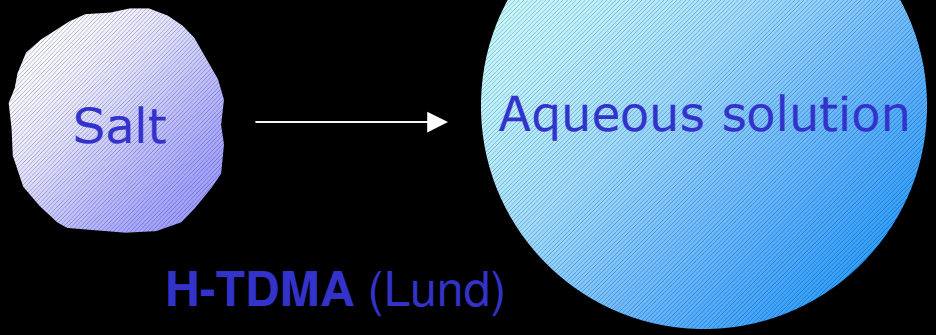


Validation

For parameterization details, see Rissler et al., 2004

Humidified particle at RH = 90%

Dry particle

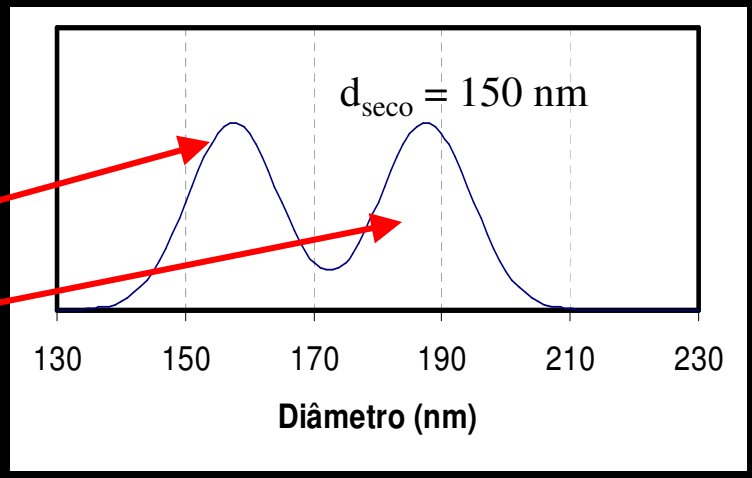


H-TDMA (Lund)

Growth factor (g):

$$g = \frac{d\left(UR = 90\%\right)}{d\left(UR = 5\%\right)}$$

Mode	Growth factor
Hidrofobic (HF)	1,05
Higroscopic (HG)	1,25



Data from Lund University group (see Rissler et al., ACP, 2004 and 2006

Determination of dry activation diameter

Soluble fraction and growth factor (GF)

$$\varepsilon = \frac{g_\varepsilon^3 - 1}{\left[g_{sol}(d_a)\right]^3 - 1}$$

g_ε : GF of a particle composed by insoluble and soluble fractions

$g_{sol:}$: GF considering the particle as composed only by soluble matter (ε = 1).

Number of dissociated ions

$$N_i = \varepsilon \cdot \frac{\pi \cdot d_s^3}{6} \cdot \frac{\rho_s}{M_s}$$

ρ_s: solute density
M_s :molar weigh
d_s: particle diameter

$$B = \frac{6\nu n_s M_w}{\pi \rho_w}$$

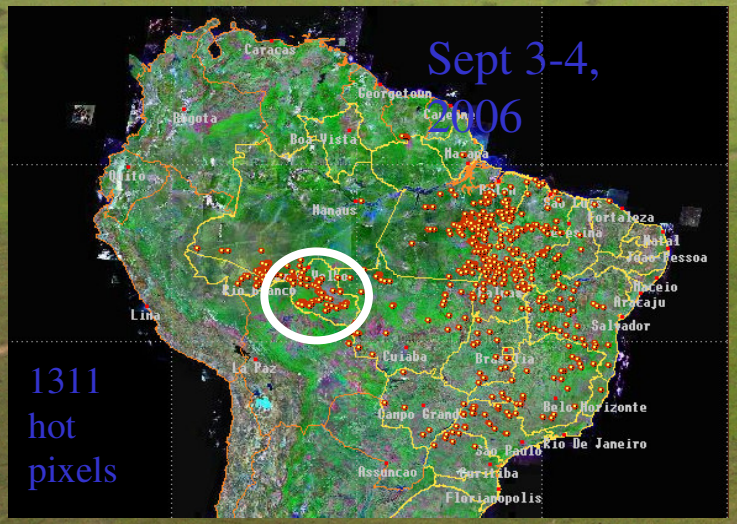
$$\ln S_c = \left(\frac{4A^3 M_s \rho_w}{27 \nu M_w \rho_s \varepsilon d_s^3} \right)^{1/2} = s_c$$

Relationship between dry diameter and activation supersaturatioin

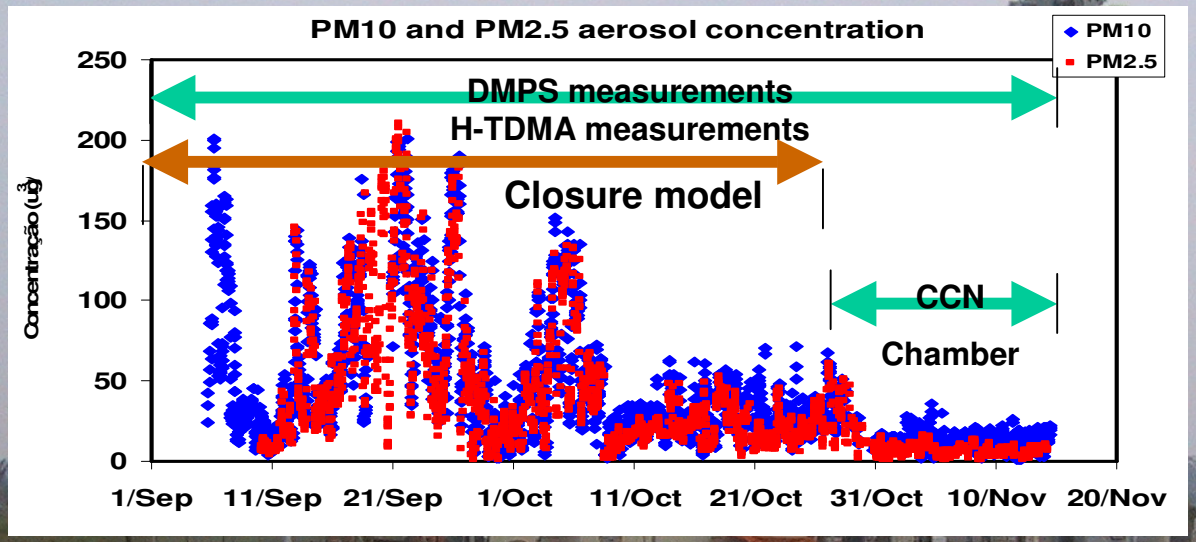
Rissler et al., 2004, Atm. Chem. Phys.

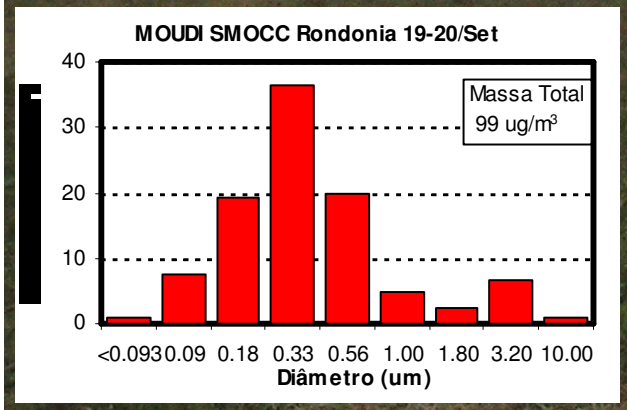
LBA/SMOCC 2002 results

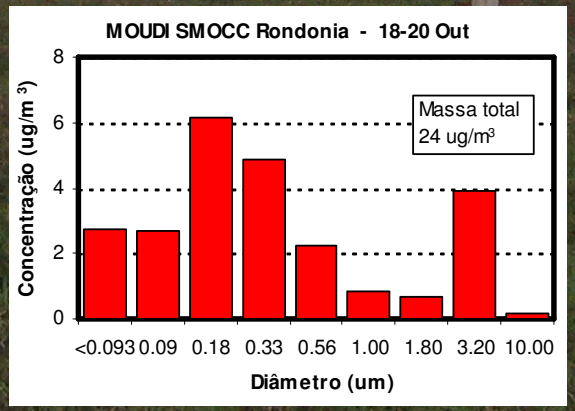
SMOCC – SMOke aerosols, Clouds, rainfall and Climate

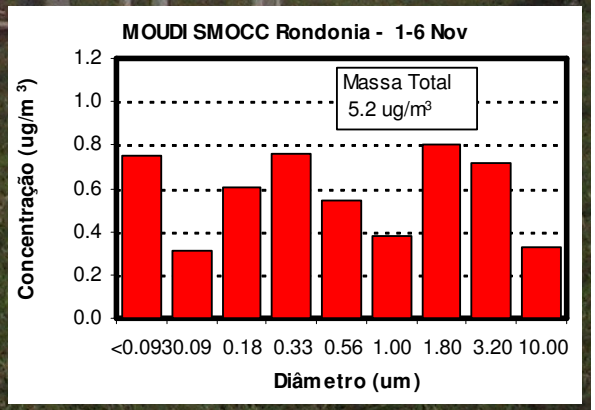


Size distribution, particle number and mass concentration varied significantly during the SMOCC







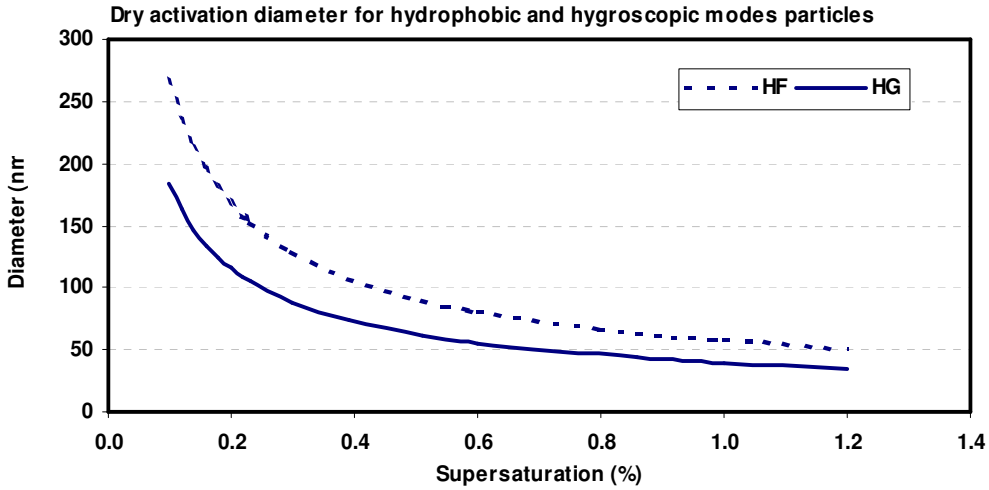


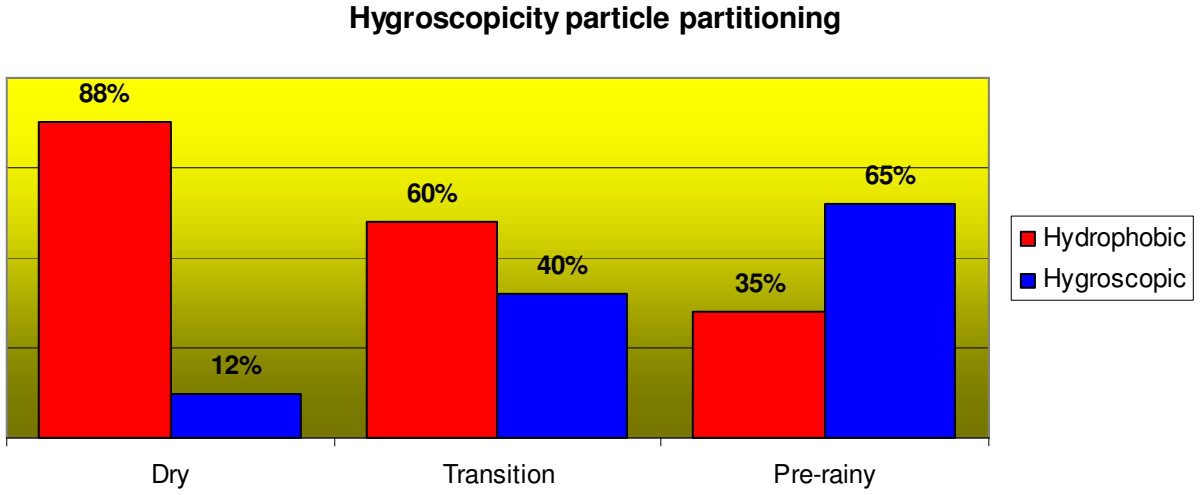
Intensive biomass-burning

Transition

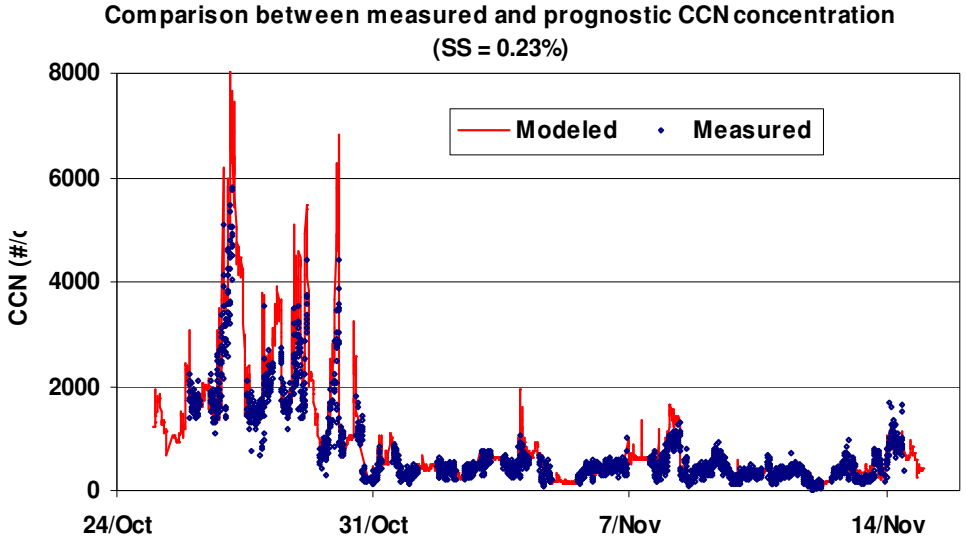
Pre-rainy sub-period

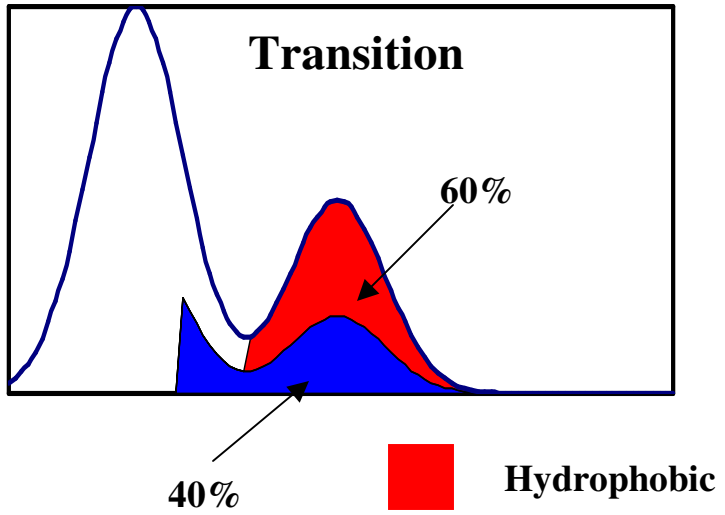
Hygroscopicity results with H-TDMA

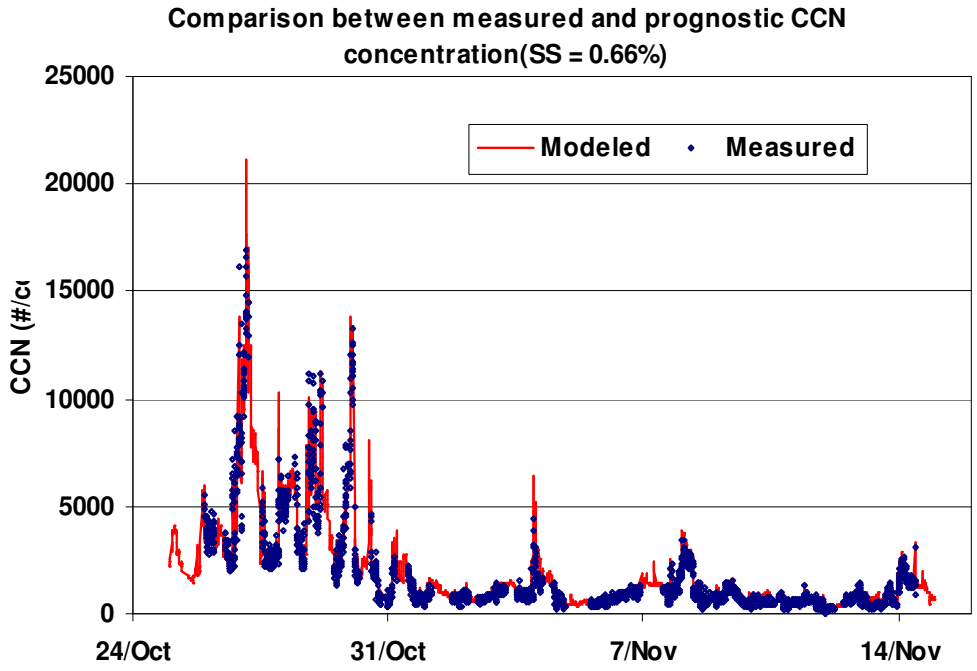


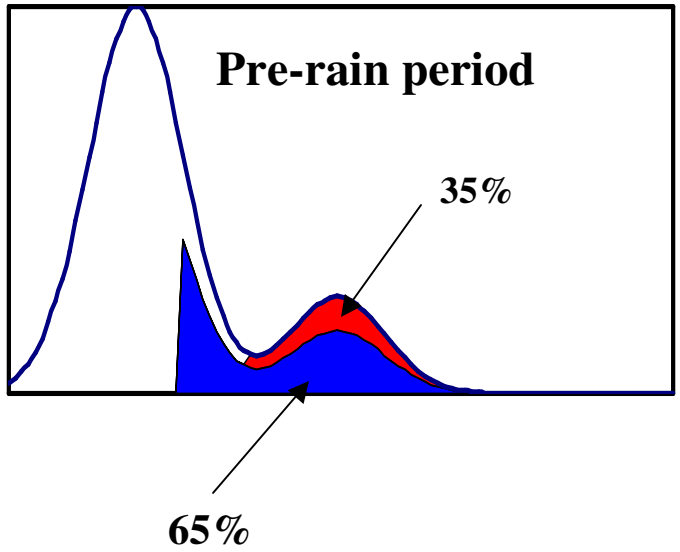


Prognostic model validation

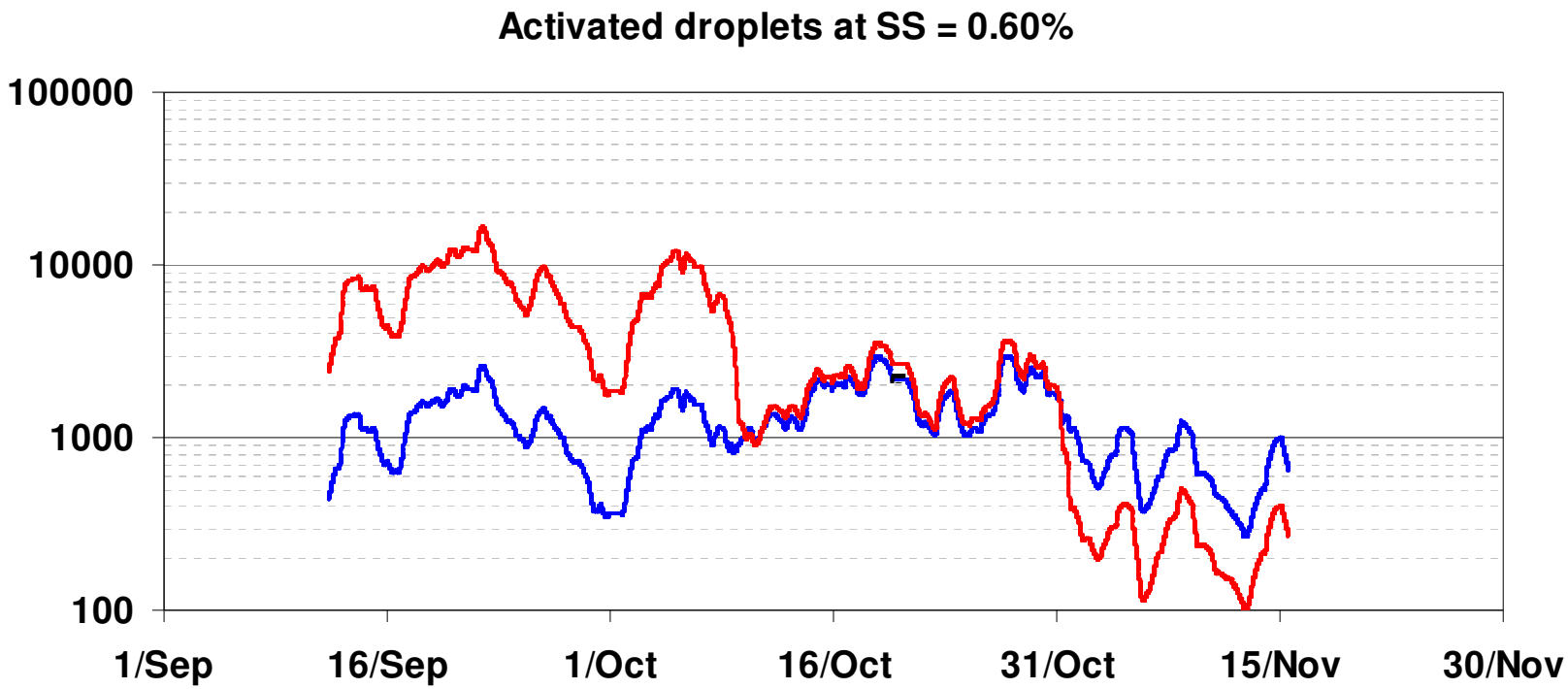






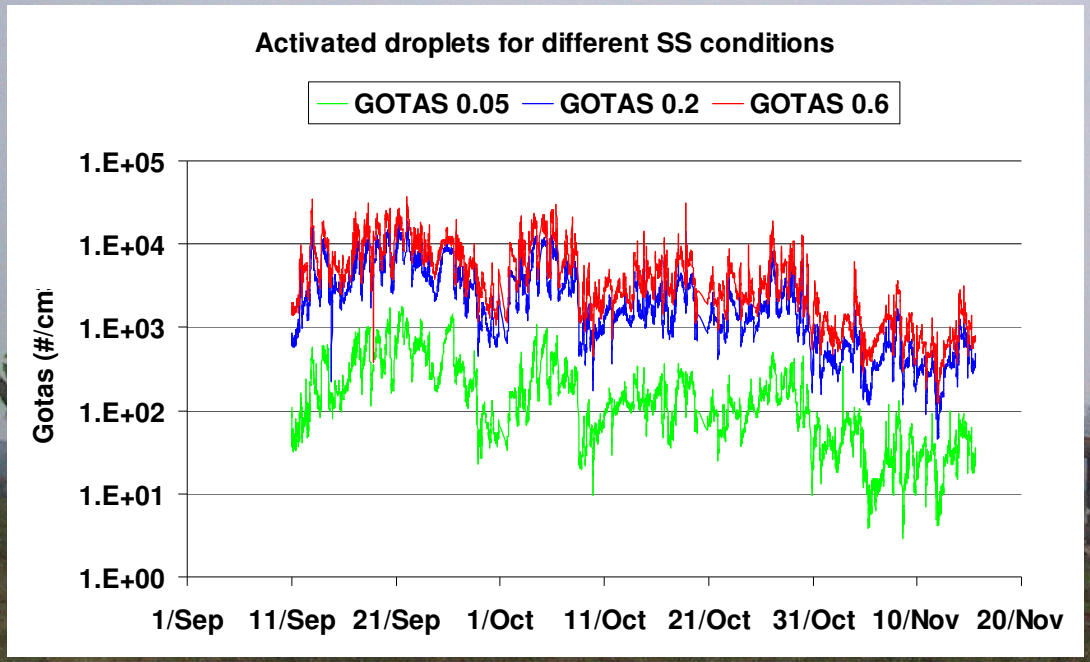


Extrapolation to intense and remaining transition



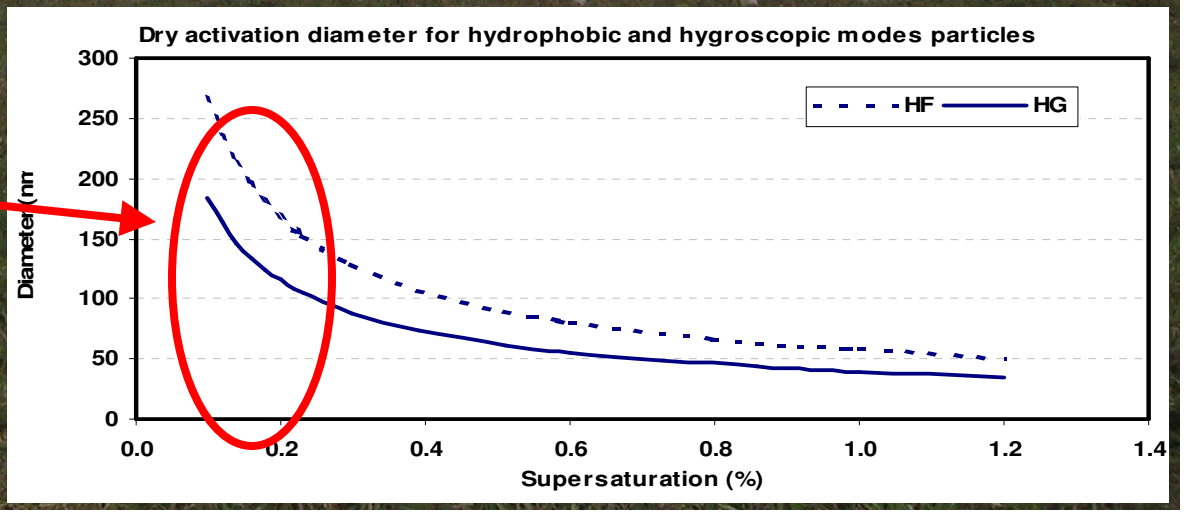
- The concentration of hydrophobic particles was correlated with particulate mass concentration
- On the other hand, the concentration of hygroscopic particles was stable suggesting a rather constant source

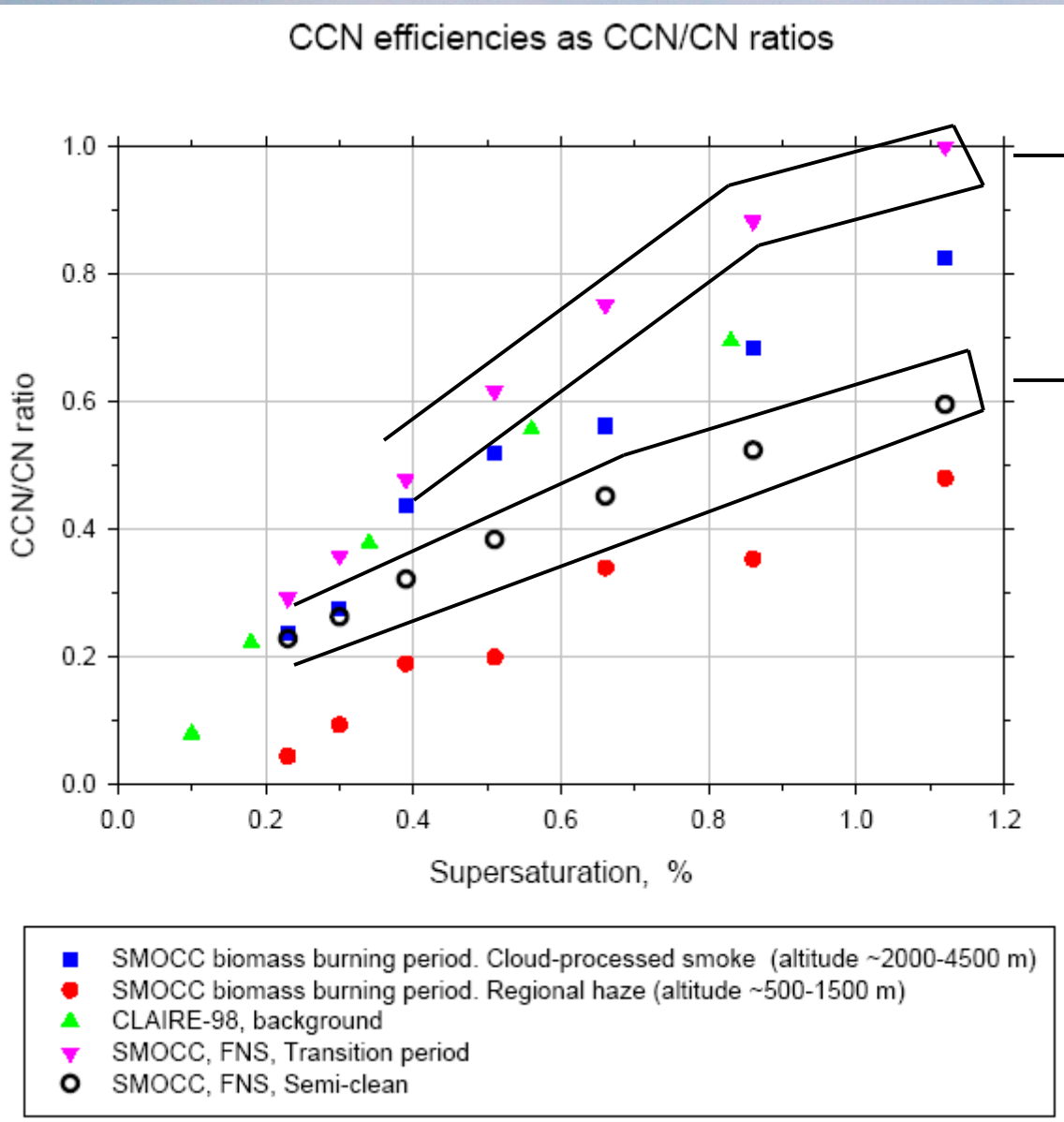
Results from the prognostic model: chemical composition is important under very low ranges of supersaturation



Number of activated droplets change orders of magnitude under low SS (< 0.2%)

- **Activation diameter: high sensibility in low SS**
- **Hydrophobic particles need greater SS to be activated**





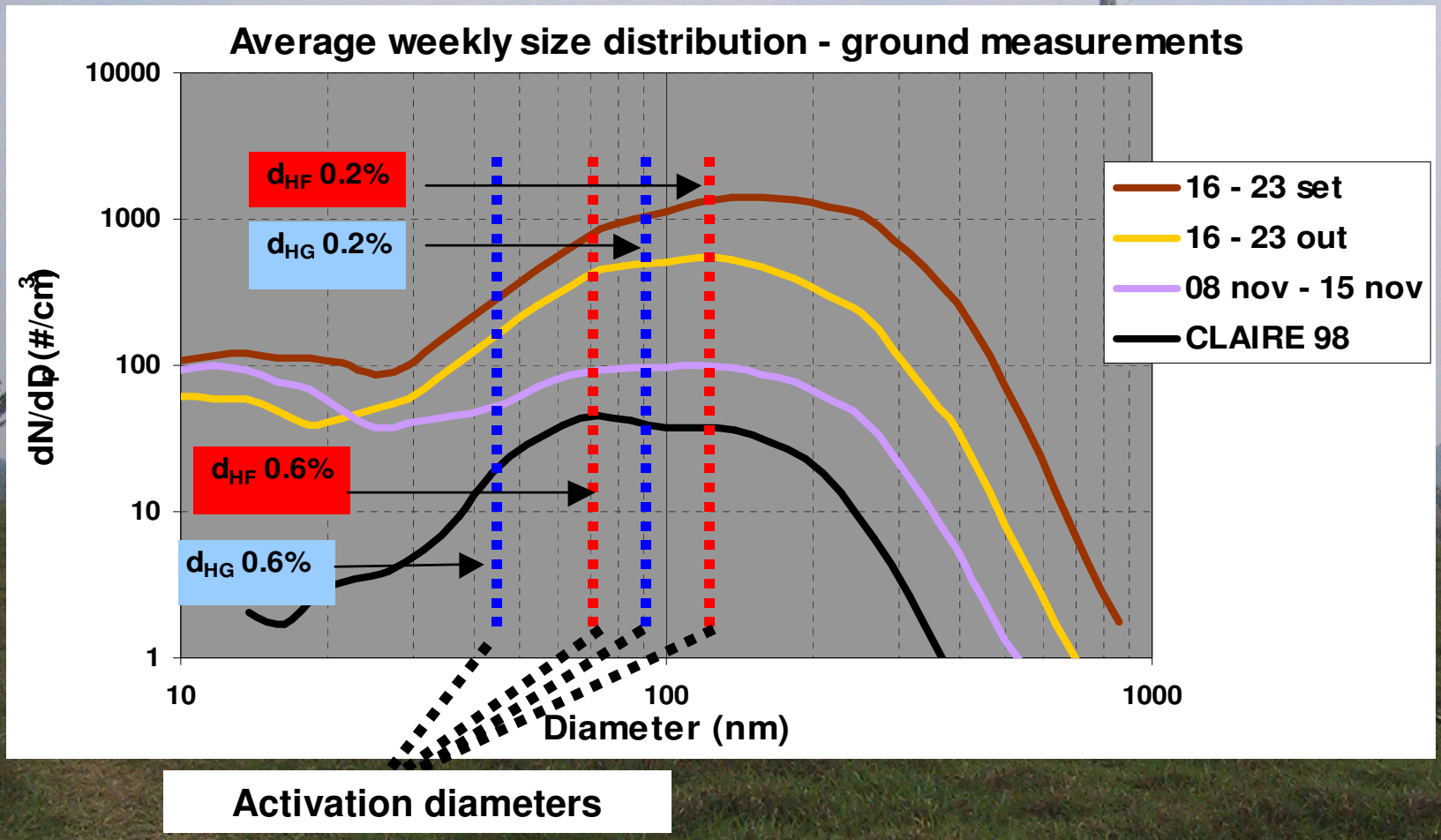
Transition

Pre-rainy period

A surprisingly result: the "semi clean" period has less efficient CCN than during transition period

Average sub period size distribution

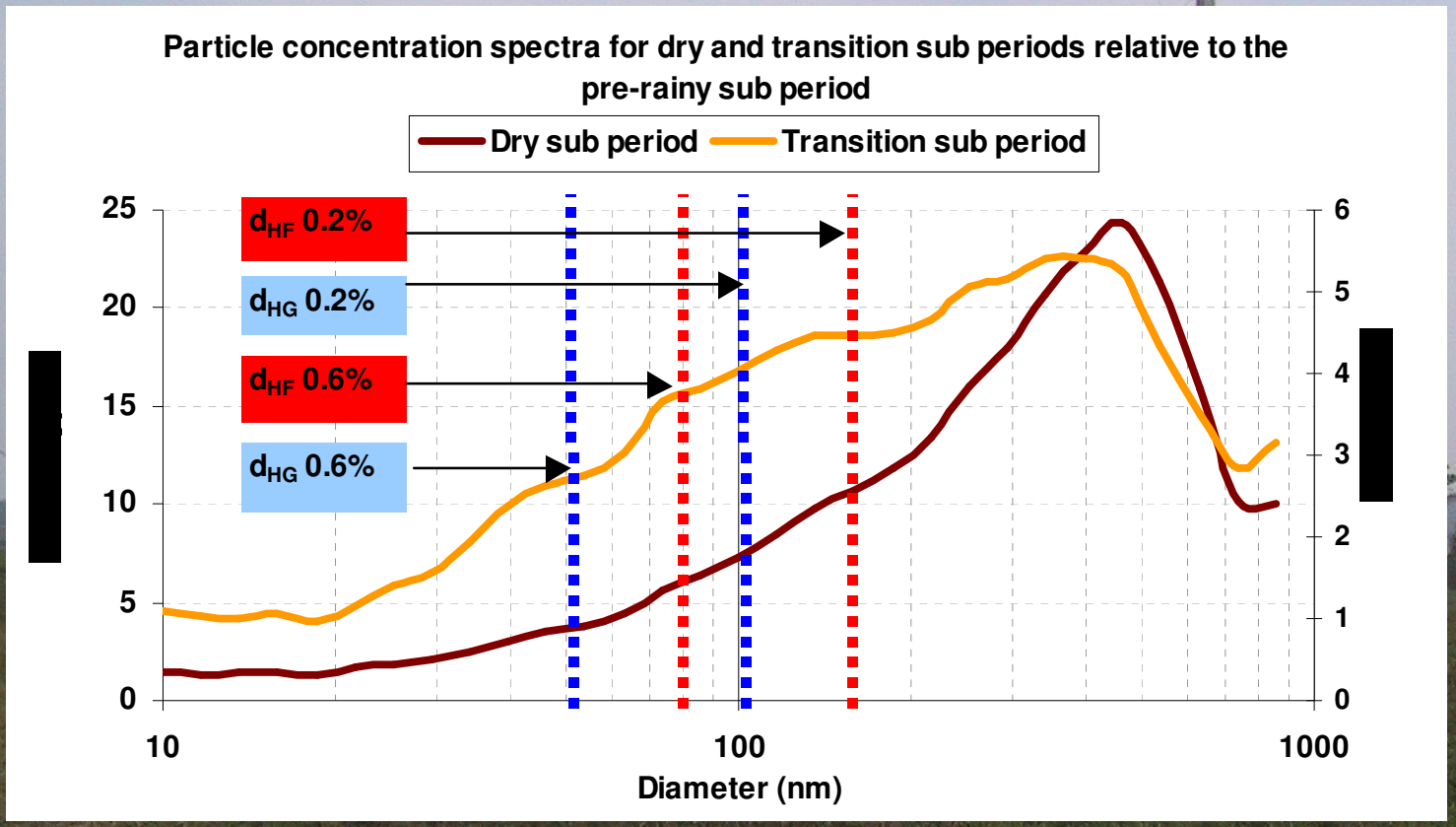
Understanding differences among sub periods



- Transition to rainy period: reduction in particle concentration with $d > d_{act}$
- Pre-rainy period: high amount of particles below activation ($d < d_{act}$)
- CLAIRE 98 (very clean condition): few particles below activation ($d < d_{act}$)

Average sub period size distribution

Understanding differences among sub periods



- Differences between dry and pre-rainy sub periods explained by different size distribution and not by chemical composition
- Excess of particles in the range $d > d_{act}$
- Particles from biomass burning are good CCN (at least its average features)

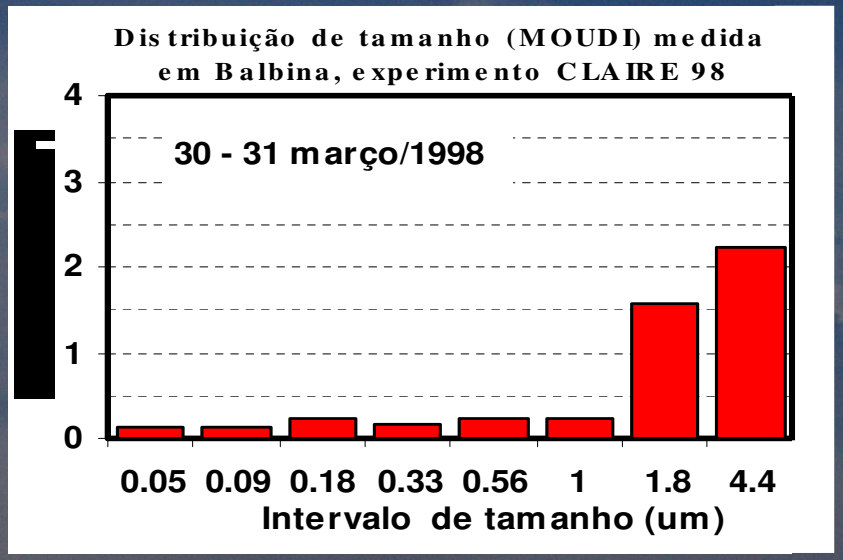
Conclusion

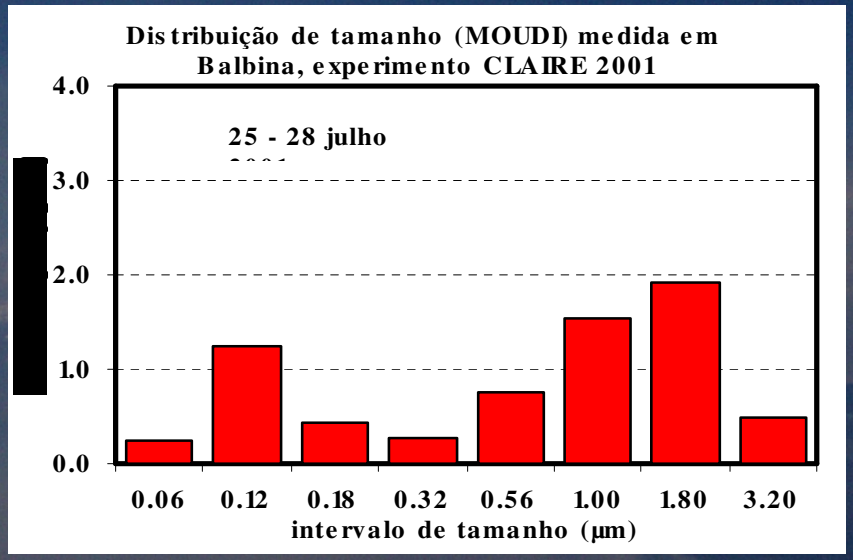
- A parameterization model based on two kind of aerosol is viable to provide reliable prognostic CCN concentration
- Size distribution is the key parameter to estimate CCN concentration if SS > 0.2%
- For SS < 0.2% the chemical composition plays na important role

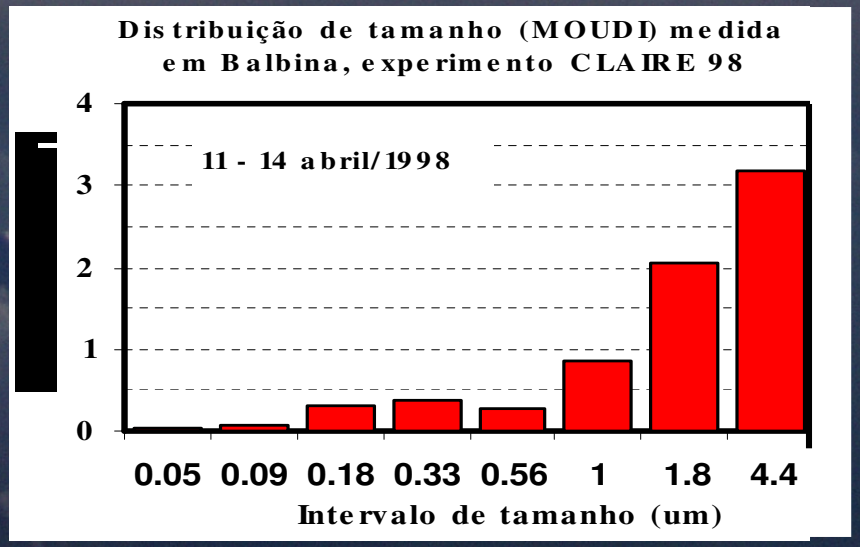
New challenges

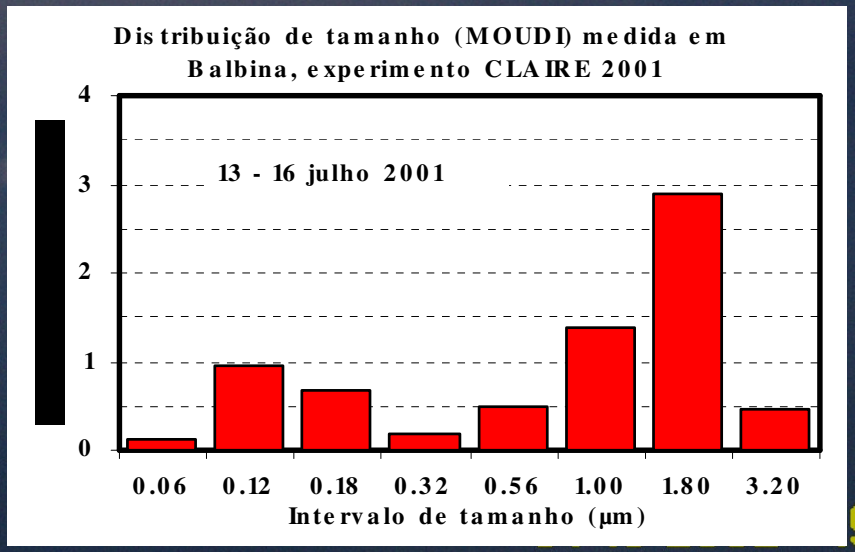
- To test the parameterization in column models with explicit microphysics and in mesoscale models (e.g.: RAMS)
- To extend hygroscopicity measurements for coarse particles – critical to evaluate its role in collision-coalescence.

Size distributions in Central Amazônia, LBA/CLAIRE 1998 (mar/apr) and 2001 (jun/jul)









LBA/CLAIRE 1998 – Central Amazonia

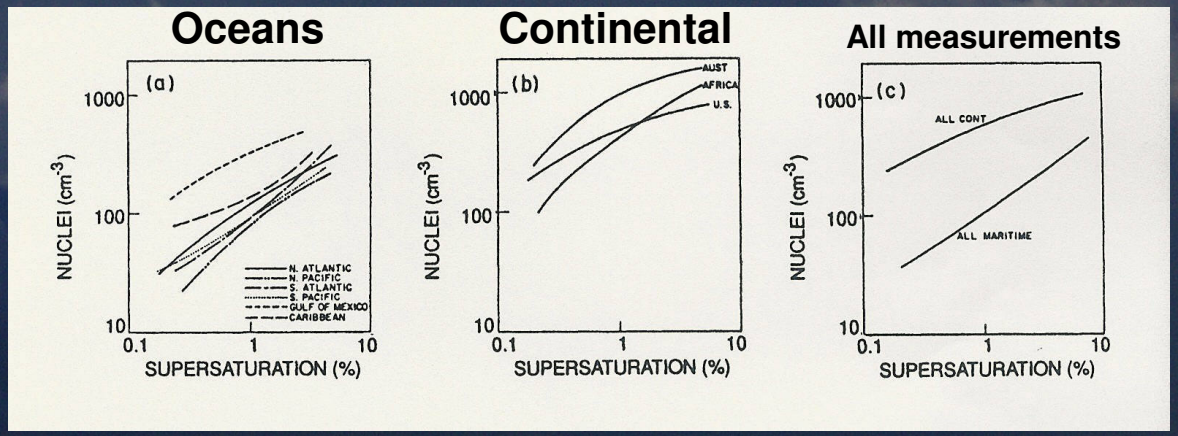
Cloud condensation nuclei in the Amazon Basin: "Marine" conditions over a continent?

Gregory C. Roberts[1] and Meinrat O. Andreae
Biogeochemistry Department, Max Planck Institute for Chemistry, Mainz, Germany

Jingchuan Zhou
Division of Nuclear Physics, Lund University, Lund, Sweden

Paulo Artaxo
Institute of Physics, University of São Paulo, São Paulo, Brazil

S^a, %	$N_{CCN\ or\ CN}$, cm^{-3}
0.15	33 ± 24
0.30	101 ± 60
0.60	182 ± 92
1.00	267 ± 132
1.50	320 ± 164
CN[b]	390 ± 250



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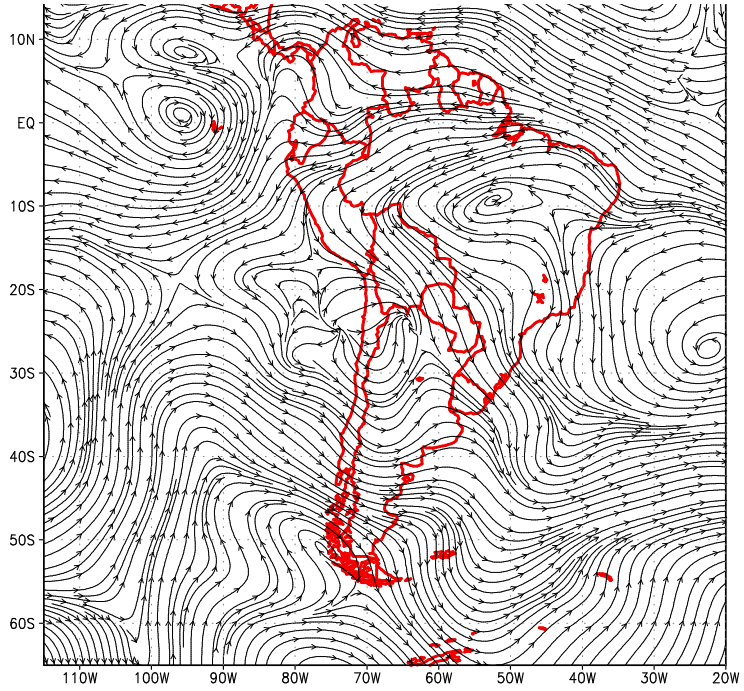
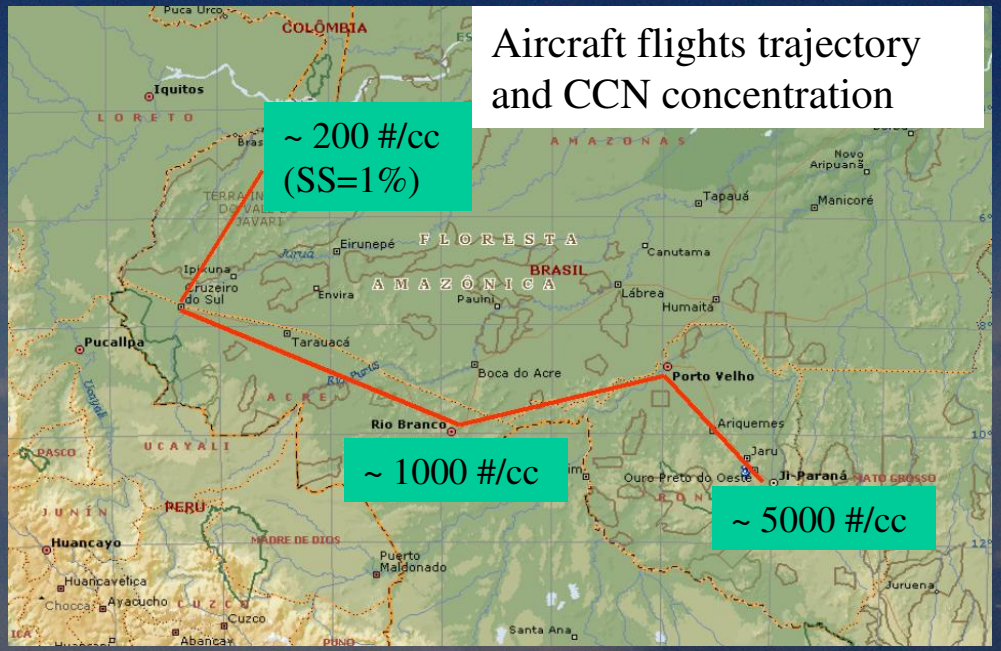
Aircraft measurements during SMOCC

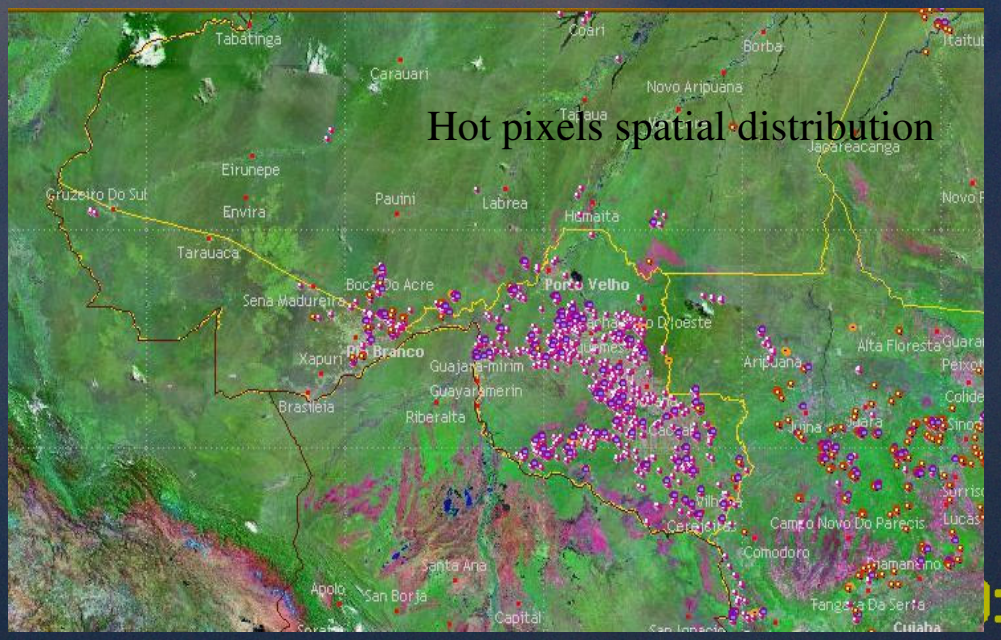
Horizontal profiles: comparing diferent air masses respect to biomass burning influence

Period: October 04 – 07, 2002

850 mb wind streamlines

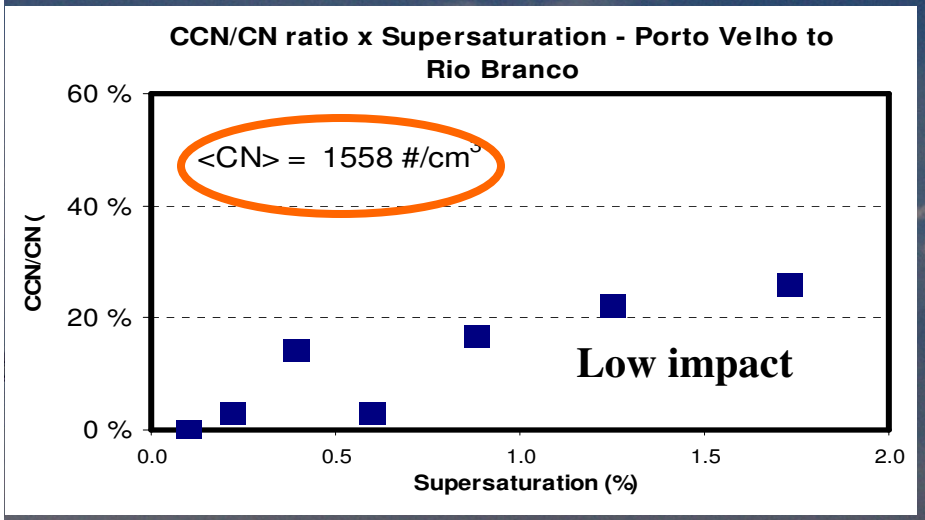


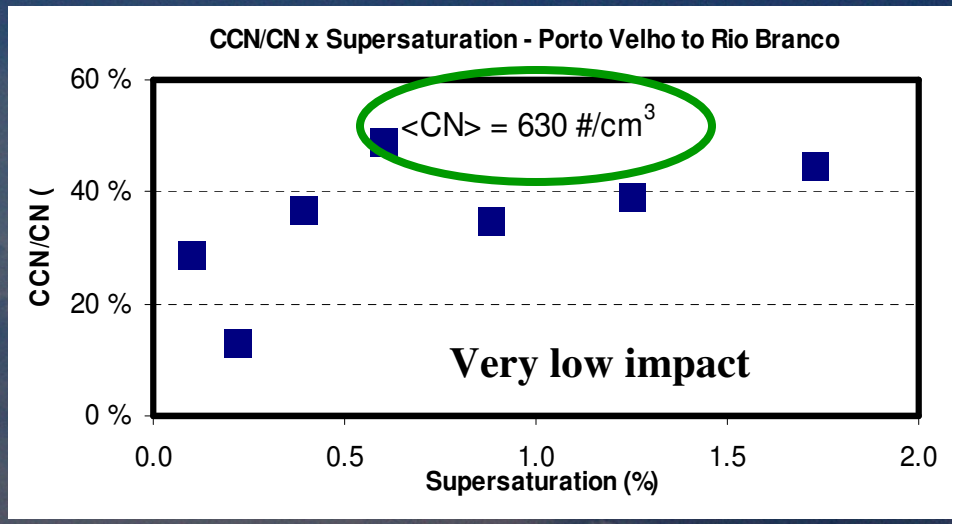




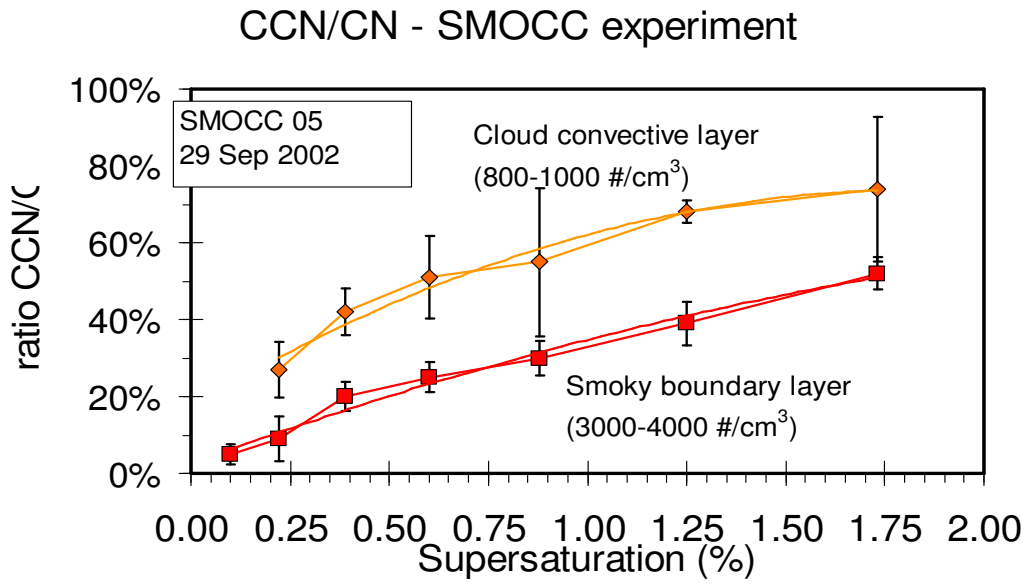
Aircraft measurements

CCN/CN ratio measured in the Porto Velho – Rio Branco flight

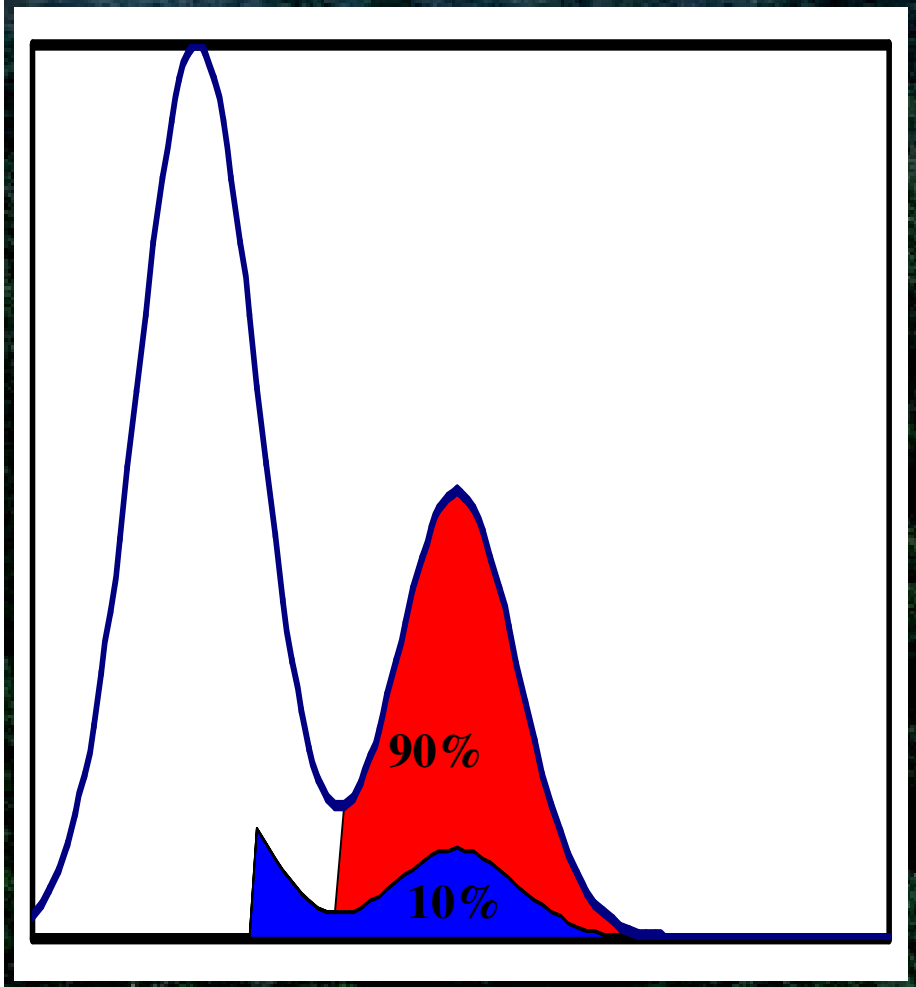


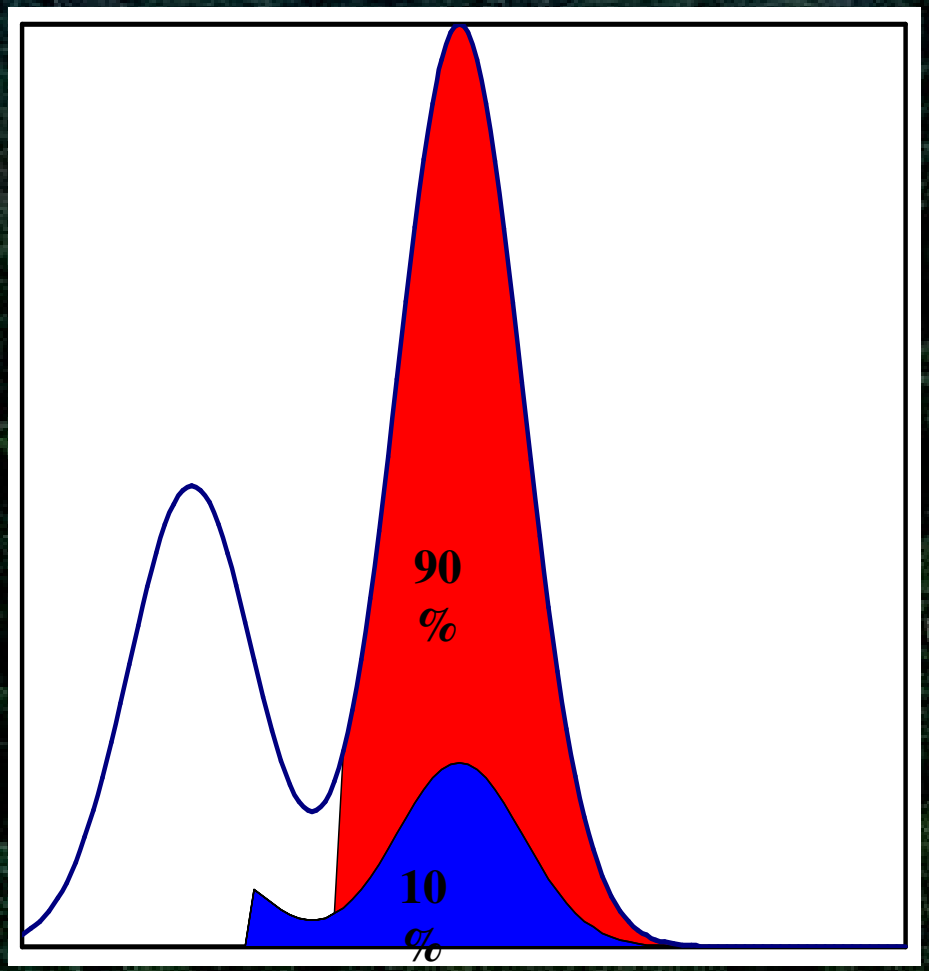


Vertical profile in Ji-Paraná.

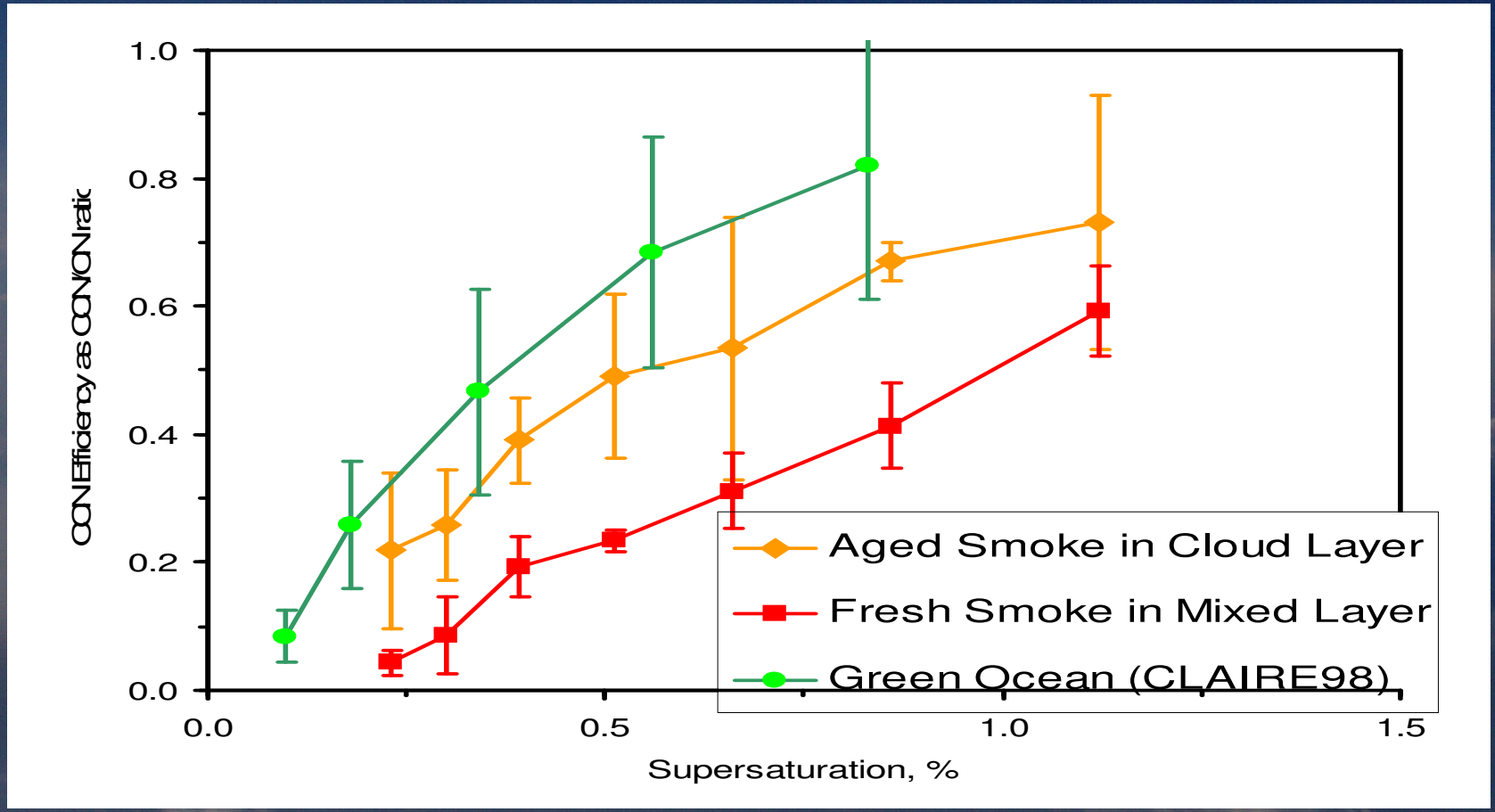


The effect of size distribution





Aircraft measurements - average



Results suggest that:

Particle concentration ⇔ biomass burning impact

Biomass burning impact ⇔ CCN efficiency

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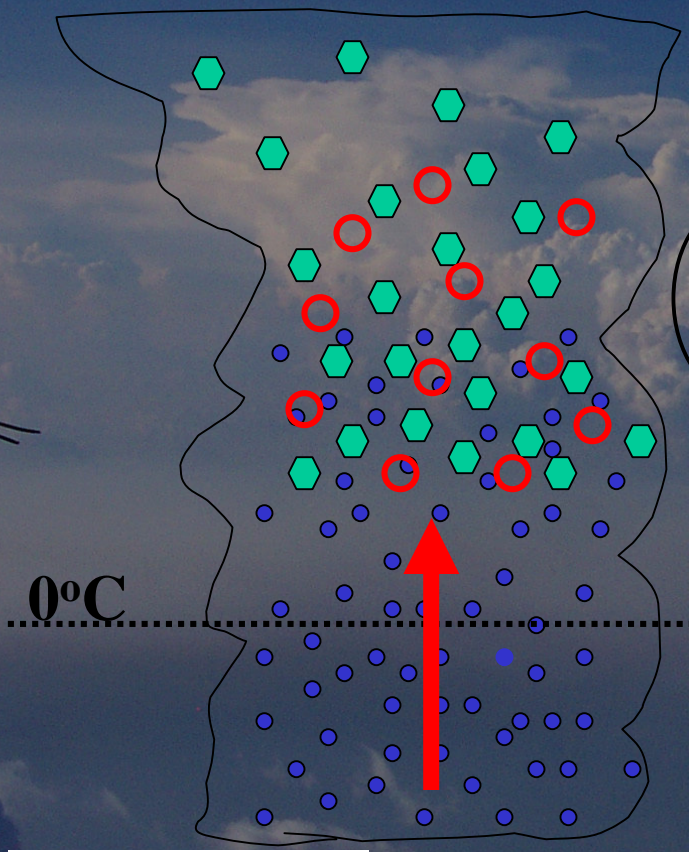
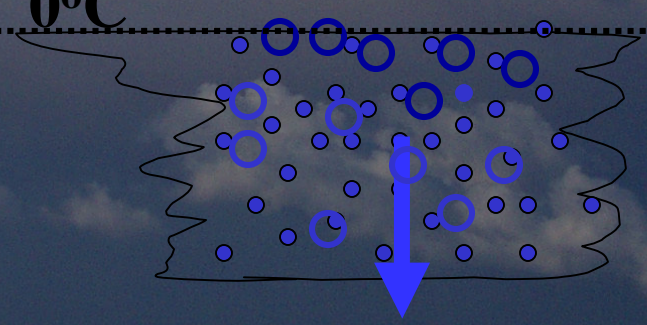
Continental clouds:
+ CCN, strong updraft, lighting



Cloud drop
Rain drop
Ice crystal
Ice precipitation

Oceanic clouds: low CCN conc., weaker updrafts, presence of giant CCN

Cloud drop
Rain drop
Ice crystal
Ice precipitation



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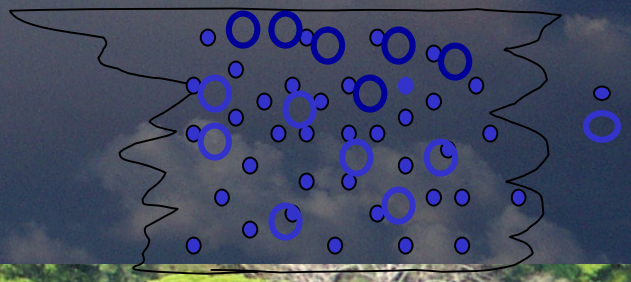
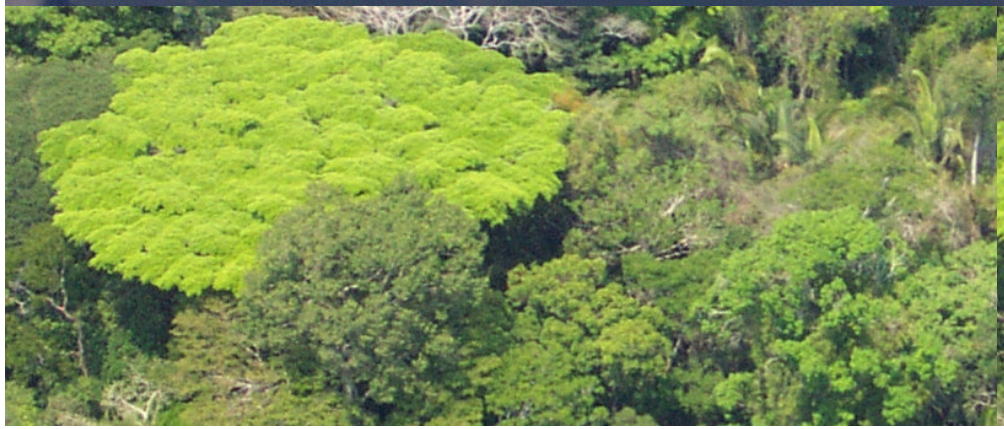
- **Forested surface: Weaker updrafts, low CCN conc., High humidity;**
- **Warm clouds**
- **Almost no lightning during the wet season; low vertical development.**

0 ºC

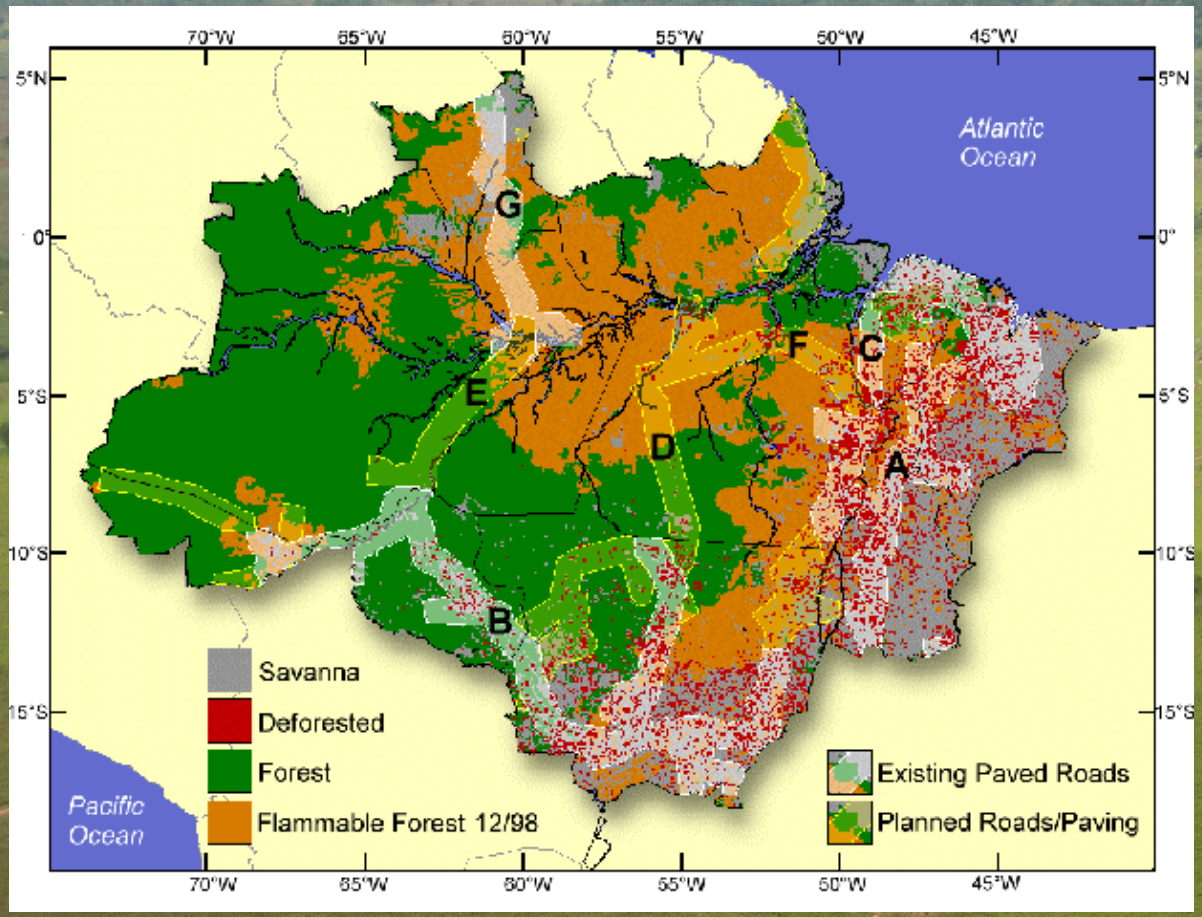


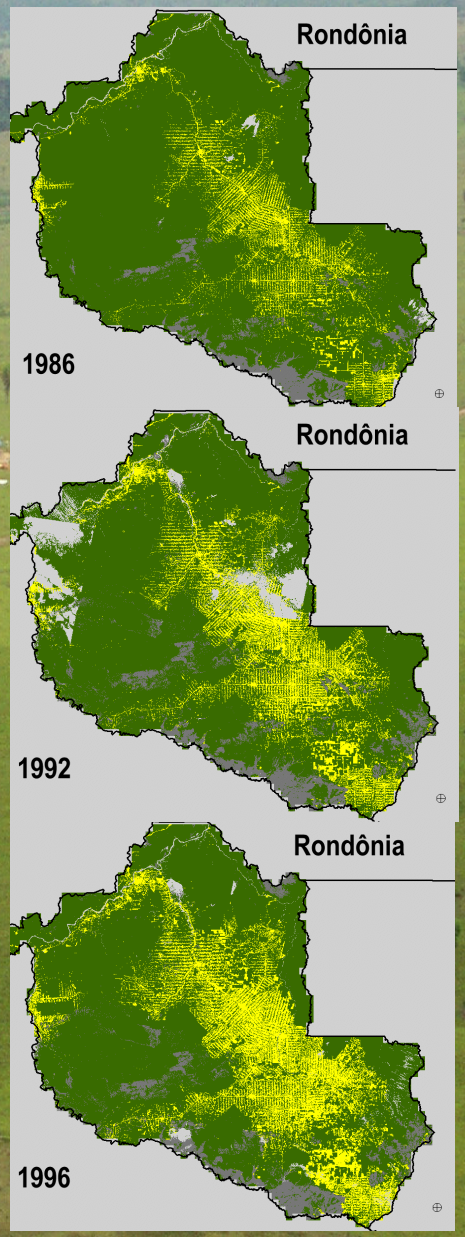


Background

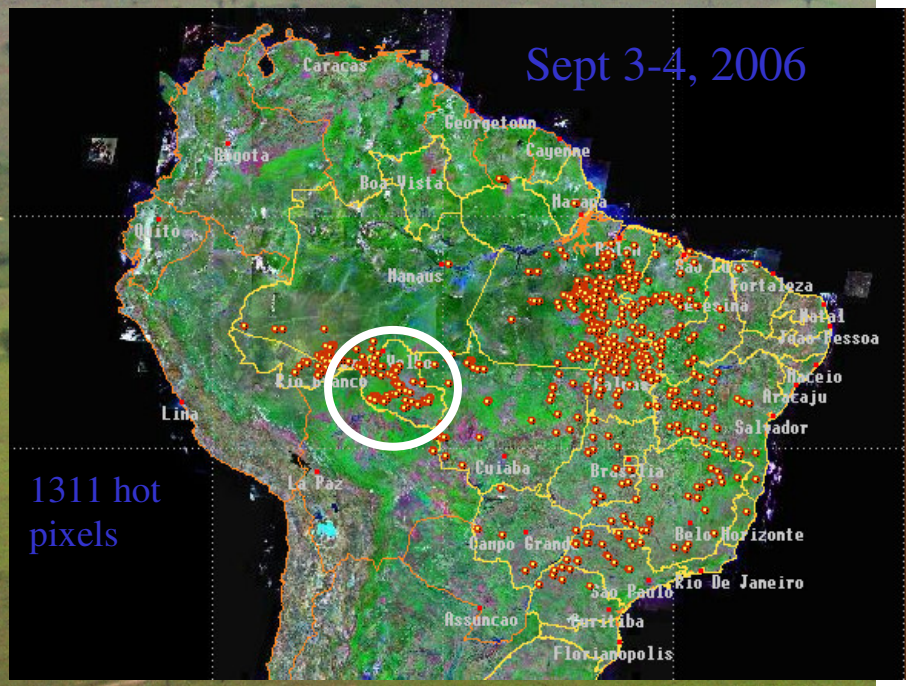
- CCN ability of aerosols are dependent of
  - Size distribution
  - Chemical composition
- These properties are usually related with number concentration, because higher concentration areas are usually polluted areas.

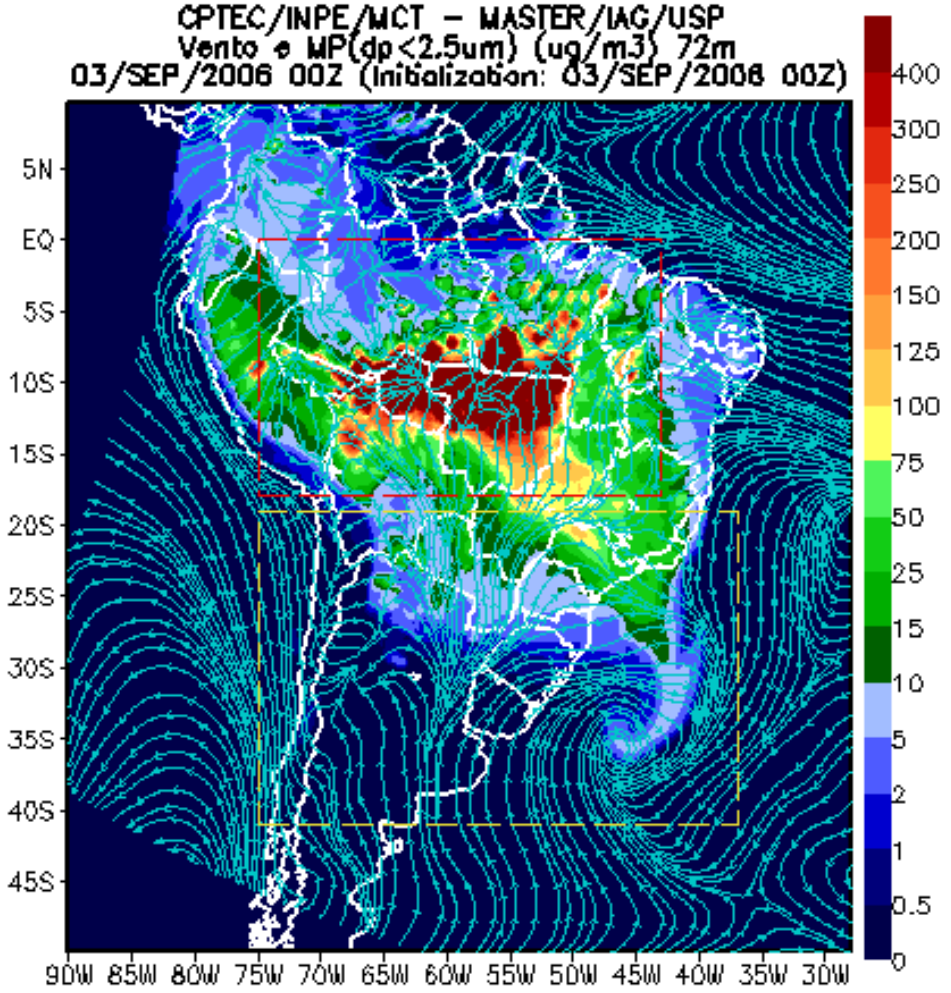
Sampling area

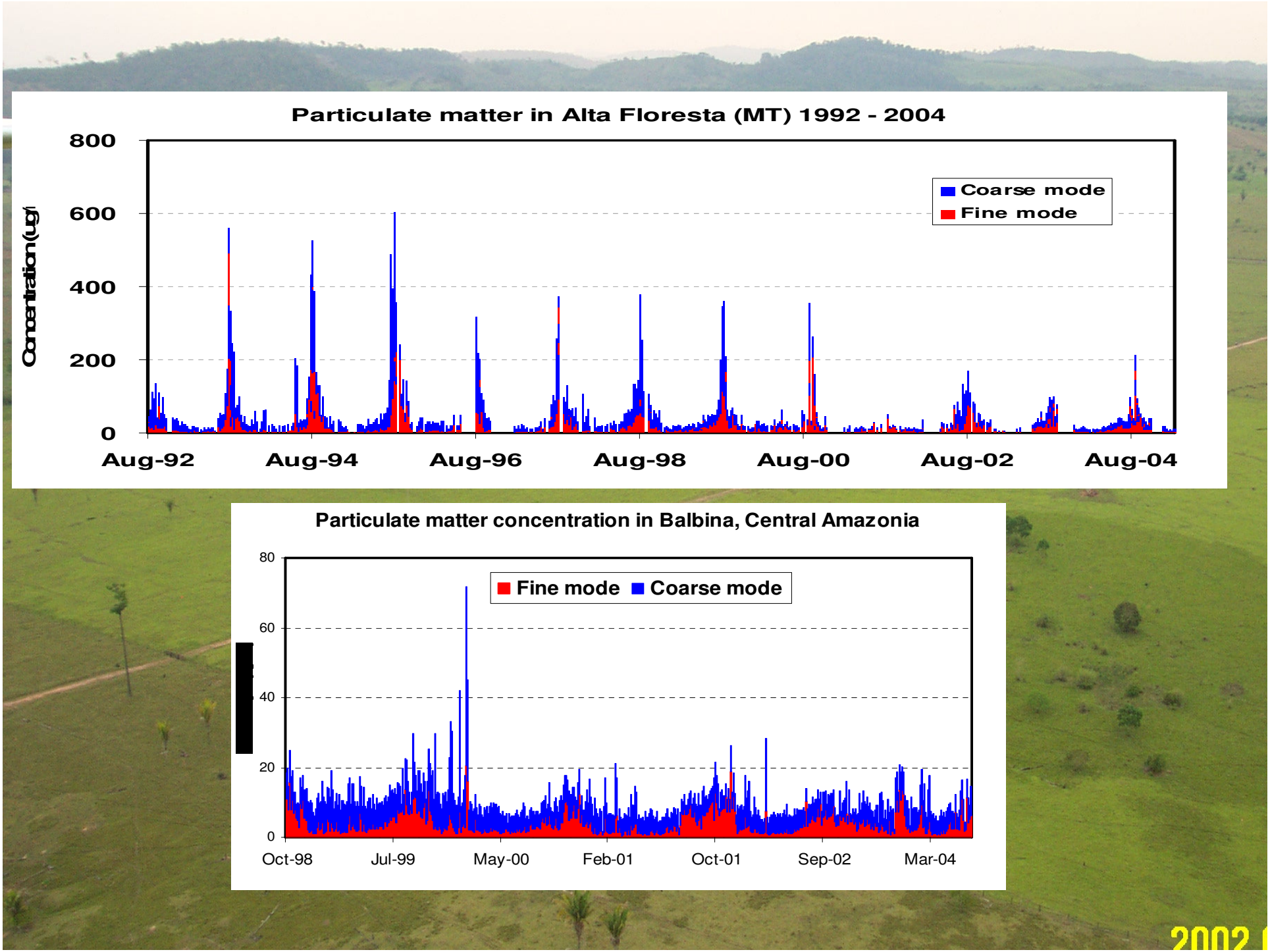




1-2 days ago…


Sept 3-4, 2006
1311 hot pixels
Caracas
Georgetown
Cayenne
Bogota
Boa Vista
Macapa
Quito
Manaus
Fortaleza
Natal
Joao Pessoa
Maceio
Aracaju
Salvador
Lima
Cuiaba
Brasilia
La Paz
Campo Grande
Belo Horizonte
Rio De Janeiro
Sao Paulo
Assuncao
Curitiba
Florianopolis


CPTEC/INPE/MCT - MASTER/IAG/USP
Vento e MP(dp<2.5um) (ug/m3) 72m
03/SEP/2006 00Z (Initialization: 03/SEP/2006 00Z)
5N
EQ
5S
10S
15S
20S
25S
30S
35S
40S
45S
90W 85W 80W 75W 70W 65W 60W 55W 50W 45W 40W 35W 30W
400
300
250
200
150
125
100
75
50
25
15
10
5
2
1
0.5
0


Particulate matter in Alta Floresta (MT) 1992 - 2004
Concentration (ug/
800
600
400
200
0
Coarse mode
Fine mode
Aug-92
Aug-94
Aug-96
Aug-98
Aug-00
Aug-02
Aug-04
Particulate matter concentration in Balbina, Central Amazonia
80
60
40
20
0
Fine mode
Coarse mode
Oct-98
Jul-99
May-00
Feb-01
Oct-01
Sep-02
Mar-04