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# Towards the simulation of size-resolved aerosol over Europe: model and observations

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# Outlook

- Main features of EMEP-MAFOR model
- Model results for size-resolved aerosol and comparison with observations for 2008 and 2010:
  - European aerosol data, 2008 (Asmi et al., 2011)
  - Selected ebas sites, 2010
- Process analysis: effect of BVOC and AVOC on new particle formation and growth (test runs vs. Observations)
- Summary of main findings

# Motivation

- Knowing the aerosol size distribution is essential for studies of:
- Health implications of air pollution: ultrafine particles (<100 nm)
  - penetrate the epithelial cells of the lungs and accumulate in lymph nodes (Nel et al., Science 2006).
  - epidemiological and toxicological studies show clear correlation between ultrafine particles and health endpoints (Daher et al., Environ. Science, 2013).
- Aerosol radiative effects
- Various aerosol processes (aerosol-clouds interaction, new particle formation and secondary aerosols etc.)

# MAFOR Aerosol Dynamics

- Solves size distribution of a mixed multicomponent aerosol on a fixed sectional grid (invariable volume)
- Documentation: Karl et al., Tellus, 2011; http://mafor.nilu.no
- Evaluated with chamber data (Karl et al., ACP 2012) and PNC measurements at motorway (Keuken et al., AE, 2012); compared to other AD models (AEROFOR, Pirjola & Kulmala, 2001).
- Consistent initiation and time integration of particle number and mass concentrations
- Condensation/evaporation of $H_2SO_4$ and condensable OV follows explicit formulation based on APC scheme (M.Z. Jacobson, 2000).
- SOA precursors: isoprene, monoterpenes, aromatics (in reactions with OH, $O_3$ and $NO_3$)

# The EMEP/MSC-W standard model

- Eulerian chemical transport model – Open Source http://emep.int
- Horizontal grid: flexible wrt projection and resolution:
  - Regional domain (Europe+N. Atlantic): 50x50 km$^2$ to 1x1 km$^2$ PS → LatLon
  - Global domain: 1º x 1º
- Vertical grid: 20 layers (up to 100 hPa) planned flexibility of vertical resolution
- Meteorology: ECMWF-IFS (standard), HIRLAM, WRF
- Chemistry: 130 species, about 160 reactions
- Aerosols: $SO_4$, $NO_3$, $NH_4$, EC, OA (POA+SOA), sea salt, min. dust
  Bulk mass: fine and coarse modes ($PM_{2.5}$, $PM_{10}$)
- SOA: 1D-VBS with 4 volatility bins
- Use for policy applications: Status, trends and projections of long-range trans-boundary pollution under the UN ECE LRTAP Convention

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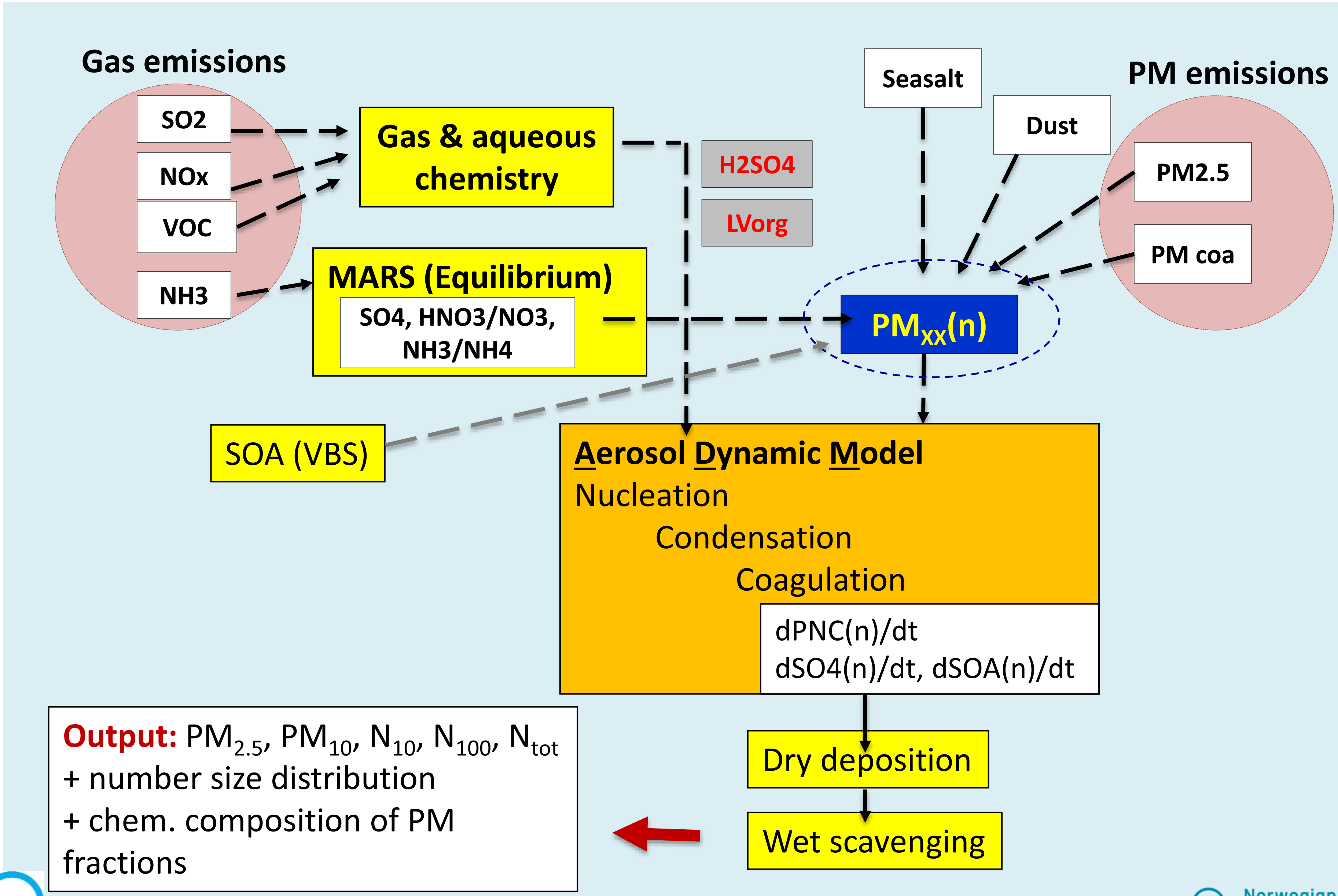
# Computational structure of EMEP-MAFOR



# Nucleation parameterizations

## Boundary Layer

$H_2SO_4$ cluster activation

$$J_{act} = A \cdot C_{g,H_2SO_4}$$

Sulphuric acid – biogenic organic

$$J_{b,het} = K_b \cdot C_{g,BLOC} \cdot C_{g,H_2SO_4}$$

Sulphuric acid – aromatic acid

$$J_{a,het} = K_a \cdot C_{g,ALOC} \cdot C_{g,H_2SO_4}$$

$A$ = 2.4 e-7 $s^{-1}$

$K_b$ = 1.1 e-14 $cm^3\ s^{-1}$

$K_a$ = 7.5 e-14 $cm^3\ s^{-1}$

**Sulphuric acid production:**

$$[OH] + [SO_2] \longrightarrow H_2SO_4 \qquad k = 2.0\text{ e-12 } cm^3 s^{-1}$$

**Critical Limit for nucleation onset**

**Wexler et al. (1994):**

$$C_{crit} = 0.16 \cdot \exp(0.1T - 3.5RH - 27.7) \times 0.05$$

## Free Troposhere

**Binary homogeneous $H_2SO_4/H_2O$ nucleation, parameterization from Vehkamaki et al. (2002)**

*Nucleated particles are assigned to 1st bin (1nm diameter) grow due condensation of $H_2SO_4$ and COV*

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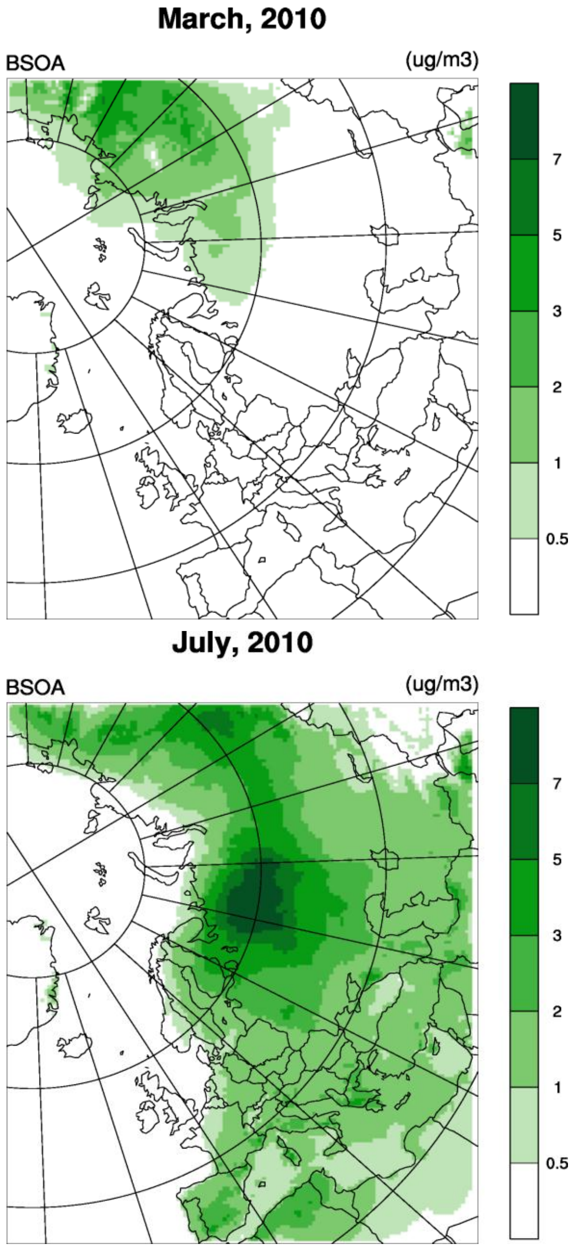
# Simulation setups

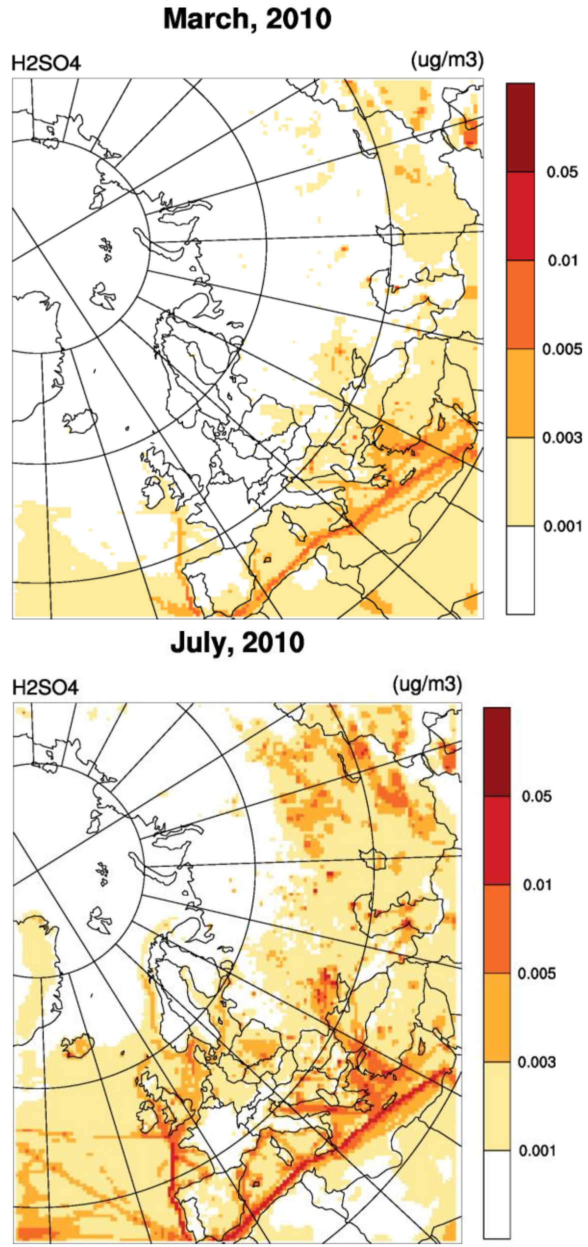
- **REFerence ARO run:** $H_2SO_4$ –aromatic acid nucleation, condensation of $H_2SO_4$ + biogenic + aromatic OVs
- **Test BIO run:** $H_2SO_4$ –biogenic organics nucleation, condensation of $H_2SO_4$ + biogenic OVs
- **Test SO4 run:** Cluster activation nucleation, only $H_2SO_4$ condensation
- **Observation data sources:**
  - Asmi et al. Year: 2008 (annual/seasonal PNC & size distribution)
  - EBAS (various sites): Year 2010, dN/dlogDp
  - SPC (San Pietro Capofiume): Year 2010, dN/dlogDp
  - SmartSMEAR (Hyytiälä) : Summer 2010, VOC, $H_2SO_4$, OH

# Seasonal differences

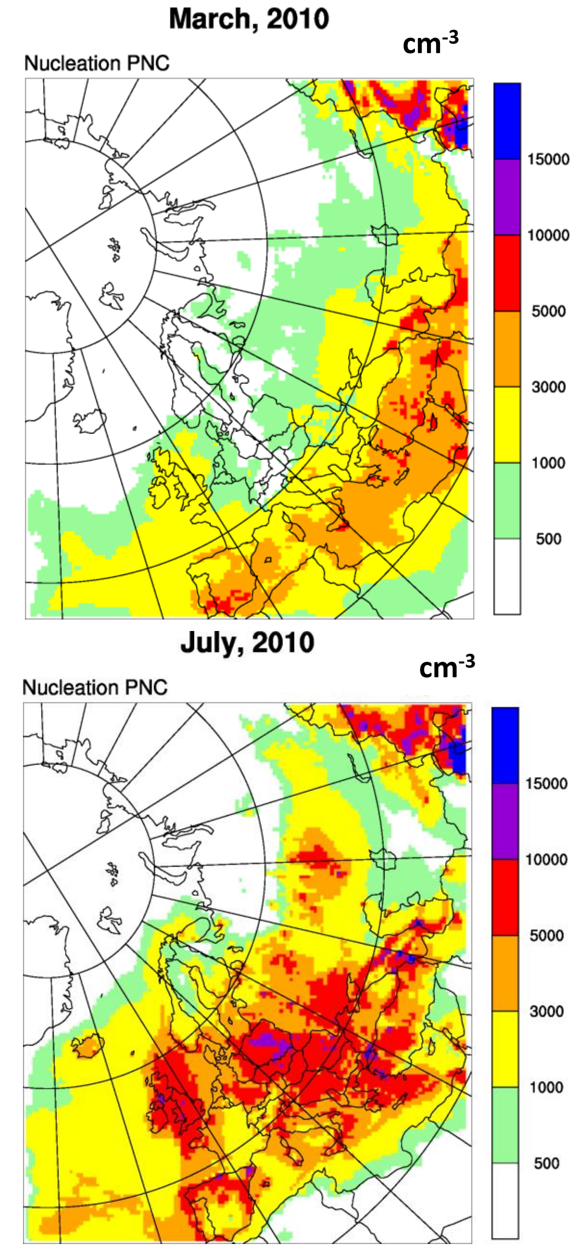
## BSOA



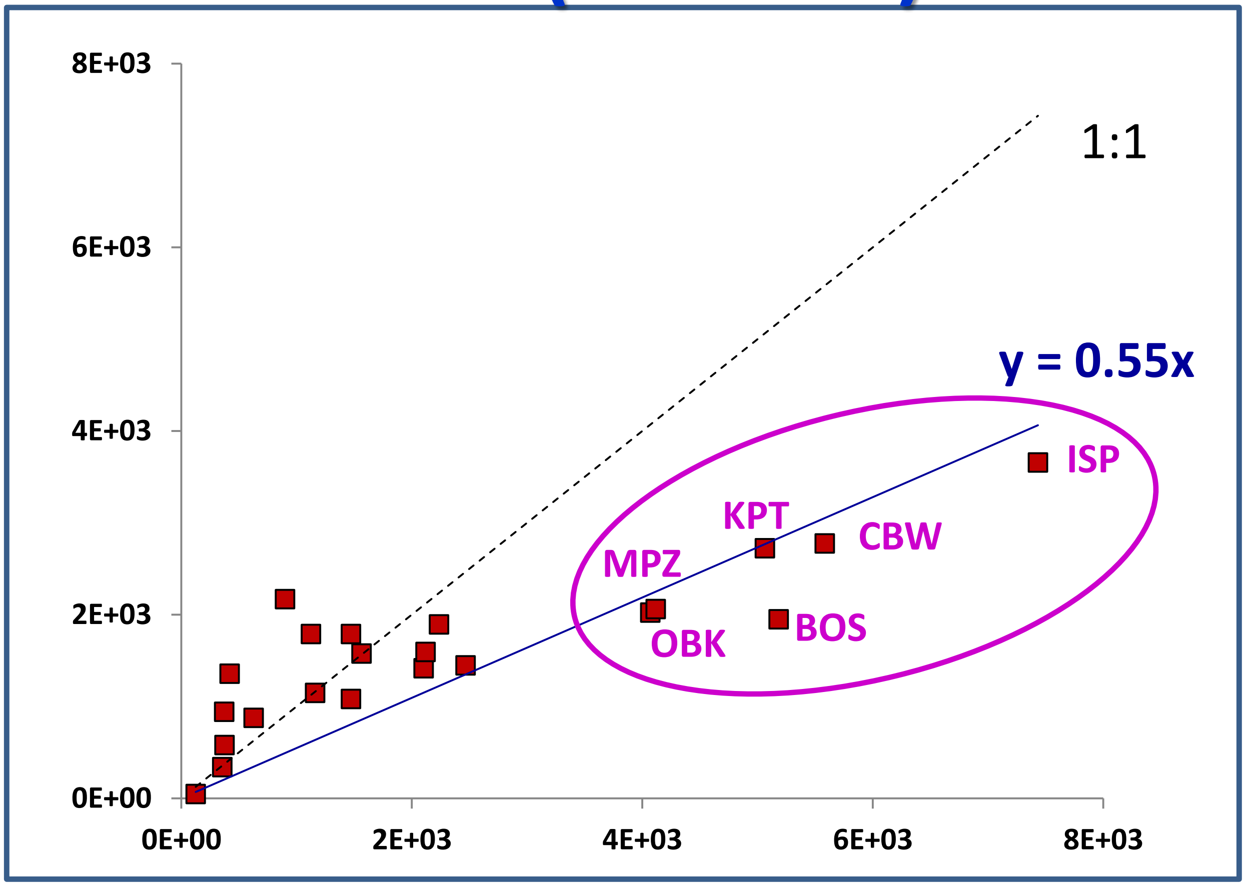

## Sulphuric acid




## PNC nucleation mode



# Scatter-plot: modelled vs. observed total PNC (d>10nm) in 2008




- Birkenes
- Aspvreten
- Pallas
- Vavihill
- Boesel
- K_Puszta
- Melpitz
- Kosetice
- Waldhof
- Finokalia
- Ispra
- Harwell
- MaceHead
- Cabauw
- Zugspitze
- Jungfraujoch
- Ny_Alesund2
- Hyytiala
- Hohenpeissenber
- Puy_deDome
- Schauinsland
- Mt_Cimone

**Observational data from Asmi et al., ACP, 2011**
**http://www.atm.helsinki.fi/eusaar/**

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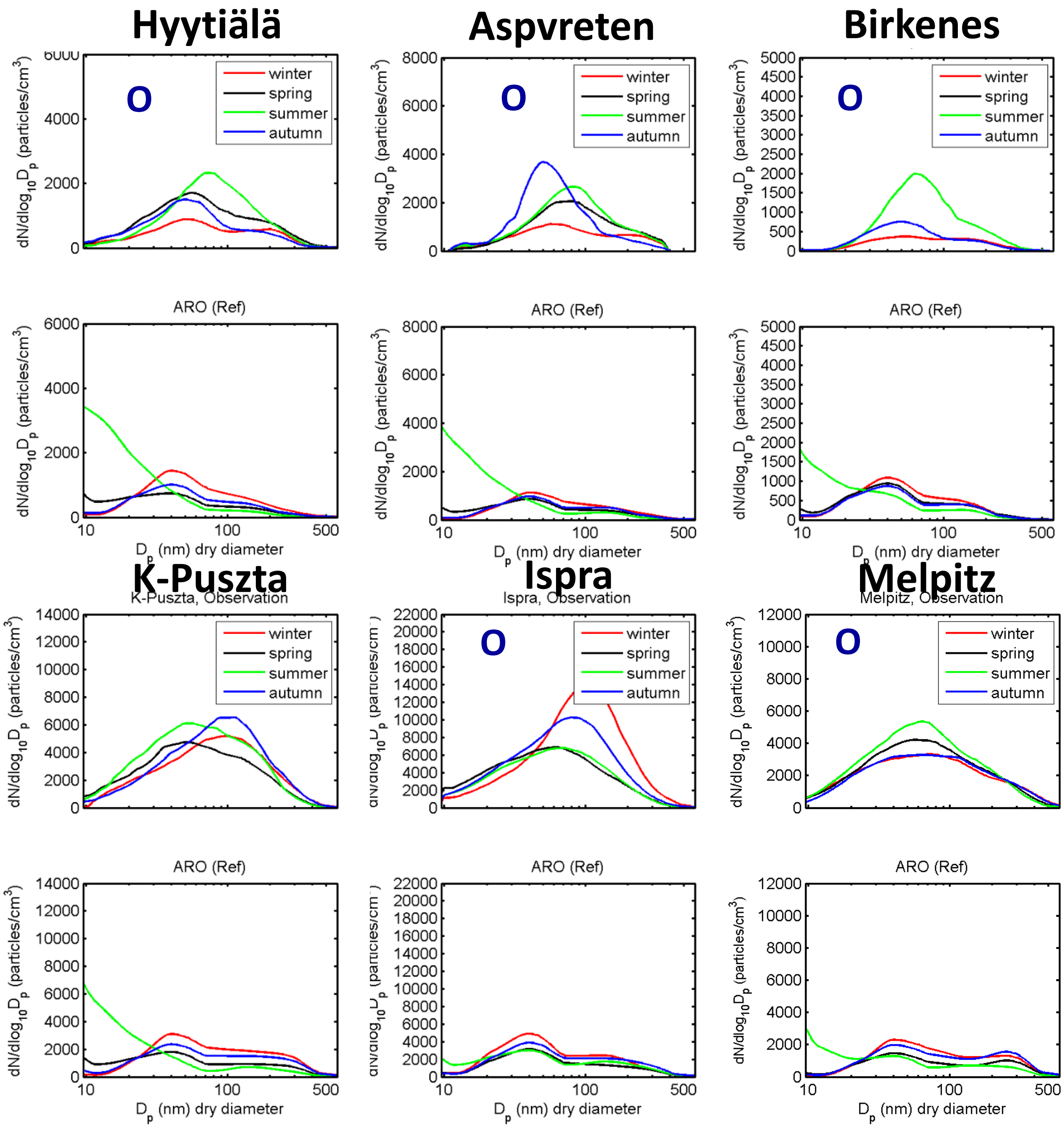
# Annual median size distribution (2008)




## MODEL:

- bimodal size distribution (Observed is closer to unimodal distribution)
- Summer: too many particles <20nm – nucleated particles not growing to observed size
- In summer: VBS-SOA and agriculture biomass burning missing
- In winter: wood burning (residential heating) underestimated. Other indications: from EC & OC comparisons

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# NPF Event Analysis (15-50 nm)

**Formation rate**

$$J_{15} = \frac{dN_{15-50}}{dt} + F_{coag} + F_{growth}$$

Nucleation = total number change + Loss by coagulation + Loss by growth

**Growth rate**

Linear fit to the maximum value of the size distribution (dN/dlogDp) as it varies over time.

$GR_{15,50}$ between 15 and 50 nm

Condensational growth

**Condensation sink**

$$CS = 2\pi D * \sum_{i=1}^{max} \beta_i * Dp_i * N_i$$

Proportional to condensational scavenging of vapours

**Coagulation sink**

$$CoagS_{15} = \frac{1}{2} K_{1,1} N_1 + \sum_{j=2}^{max} K_{1j} N_j$$

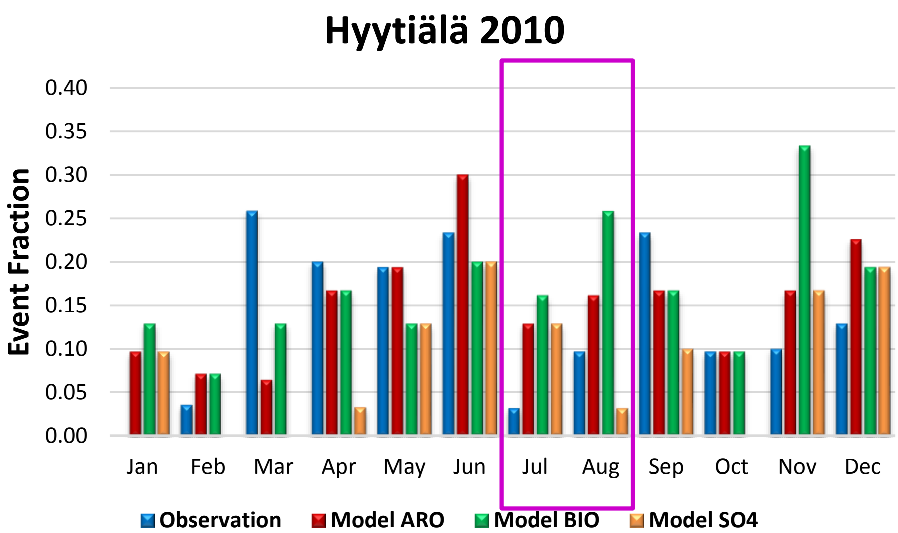
**Survival probability**

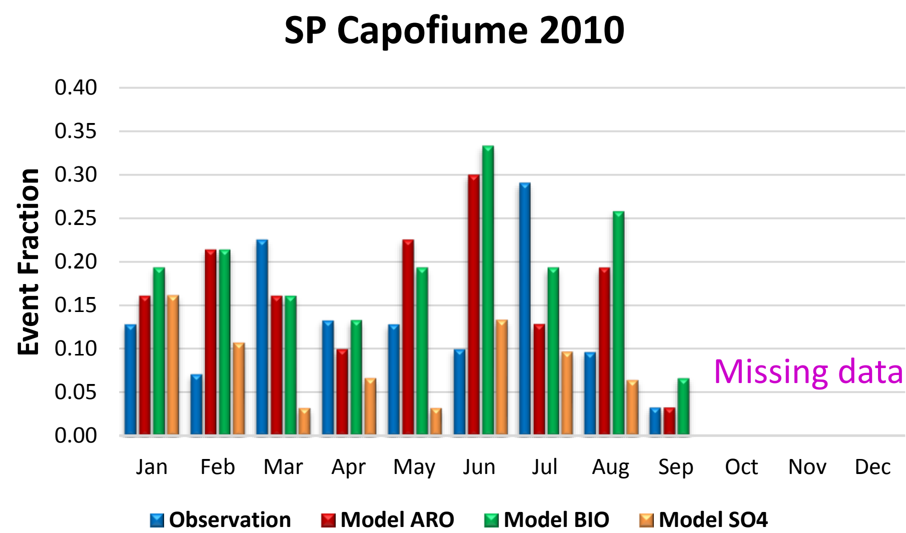
$$SP_{15,50} = exp\left(\frac{-\tau_{15,50}^{cond}}{\tau_{15}^{coag}}\right)$$

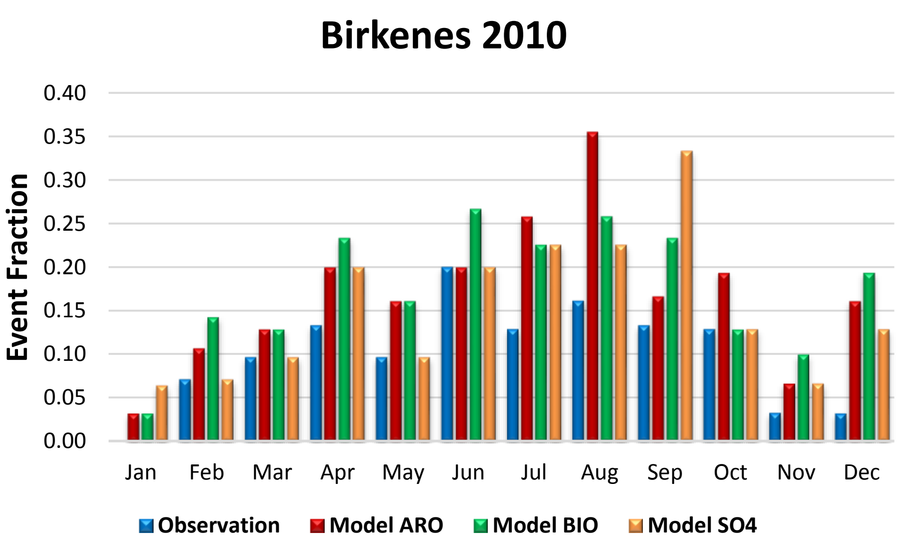
Survival probability from 15 to 50 nm.

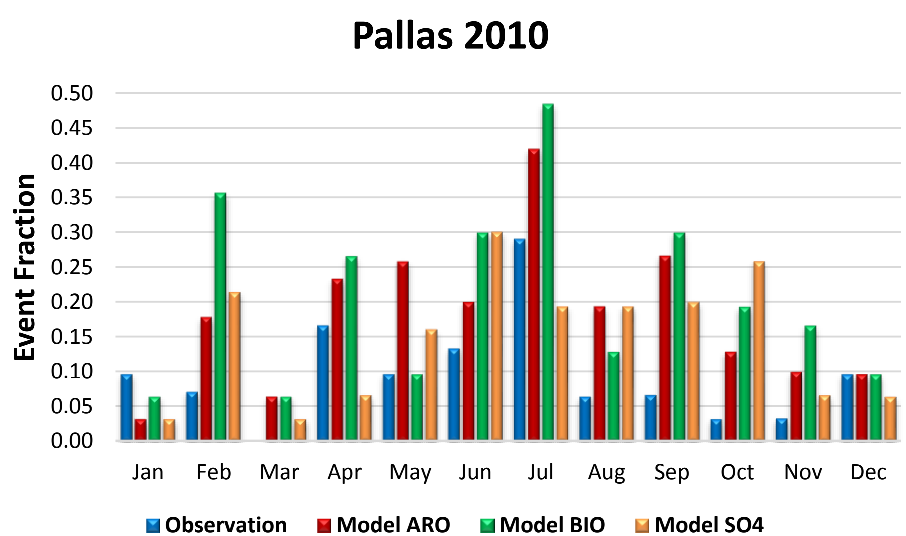
with $\tau_{15,50}^{cond} = \frac{50-15nm}{GR_{15,50}}$; $\tau_{15}^{coag} = \frac{1}{CoagS_{15}}$

# Event Frequency




**Definition of event:** $dN_t$ (15-30nm) > $5 \cdot dN_{t-1}$ (15-30nm) *[hourly averages]*
growth of 15-50 nm particles for > 2 hours and GR > 0.28 nm/h

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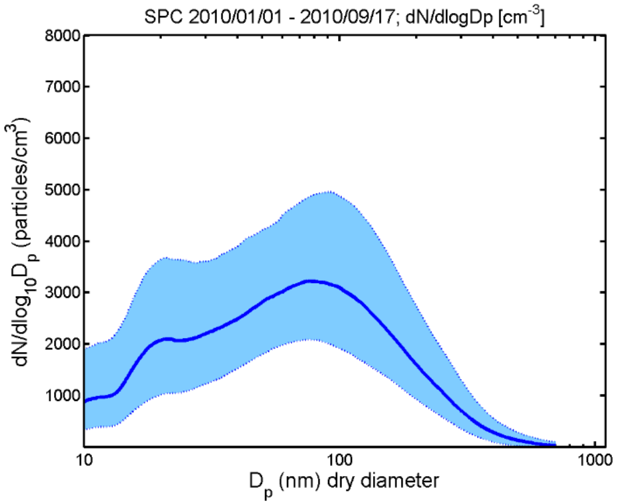
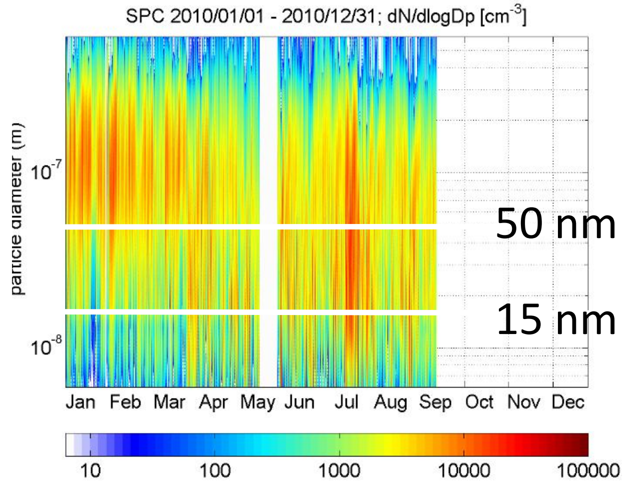
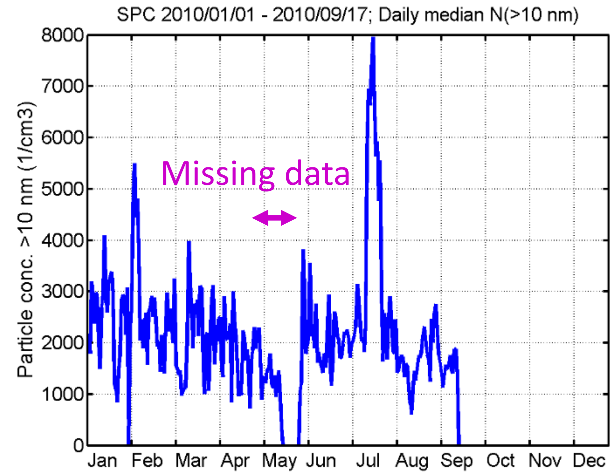
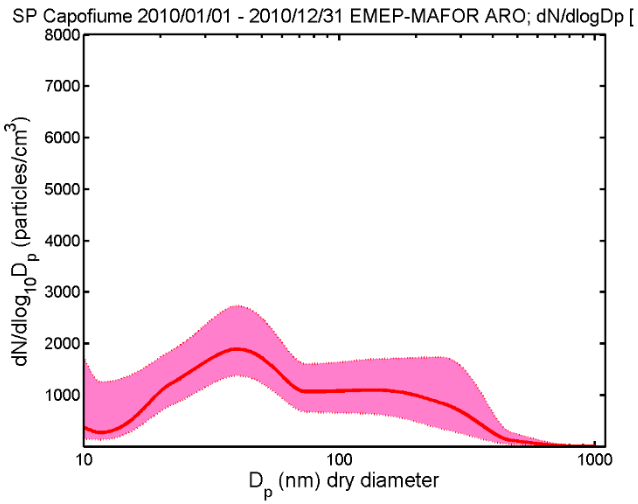
# SP Capofiume 2010 - Observations

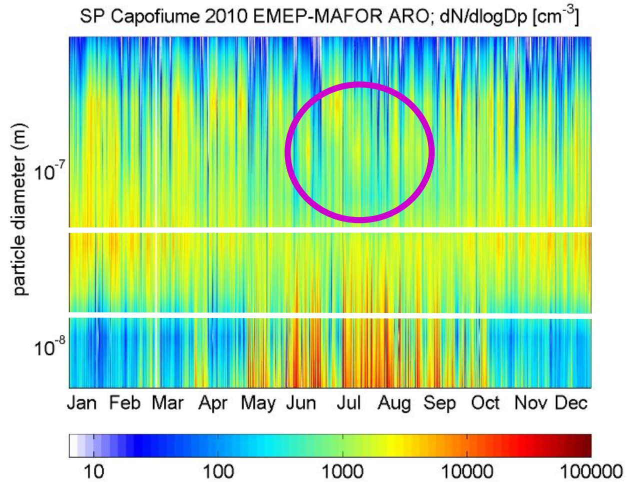


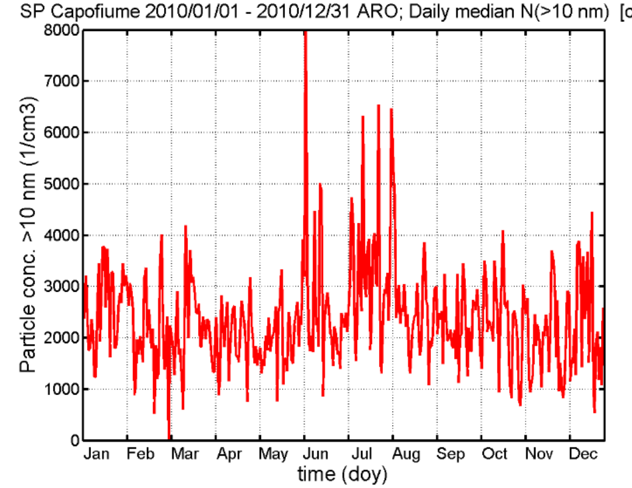




# SP Capofiume 2010-ARO

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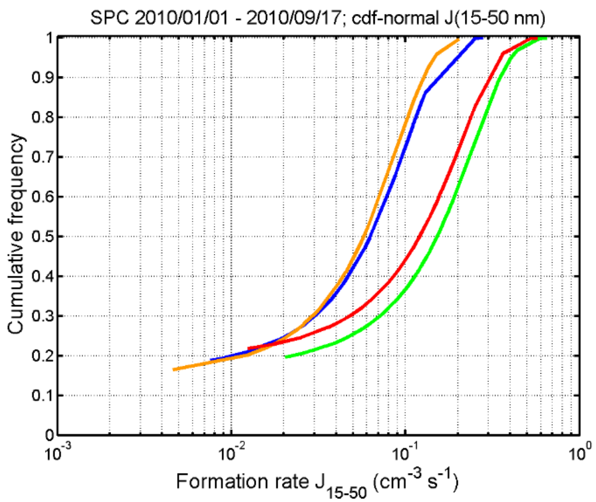
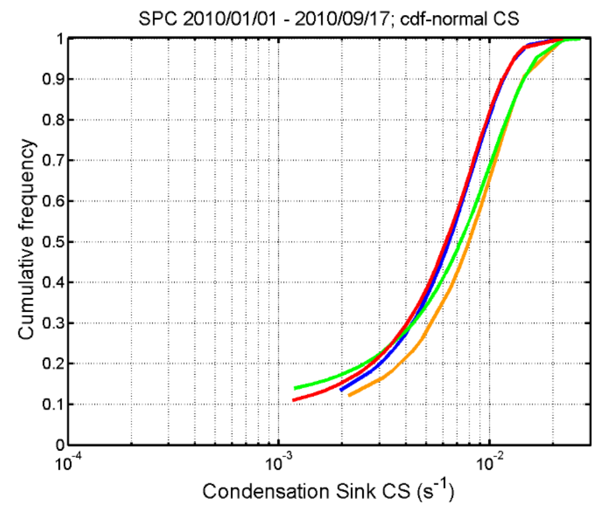
# SPC 2010

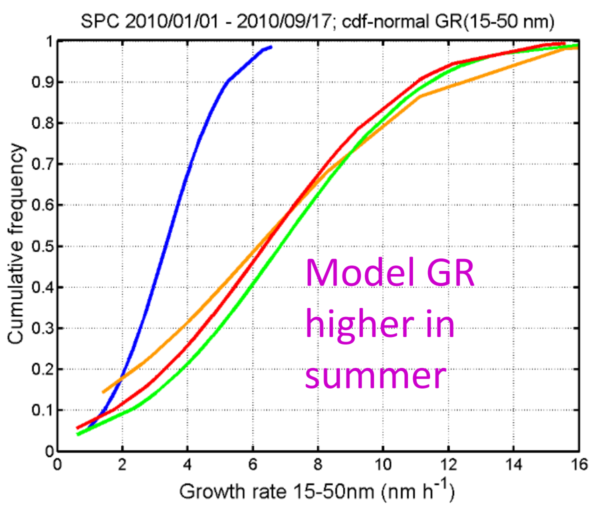
-SO4

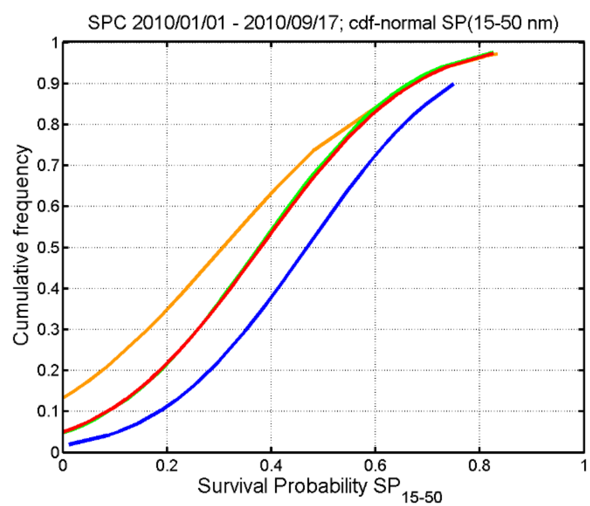
-BIO

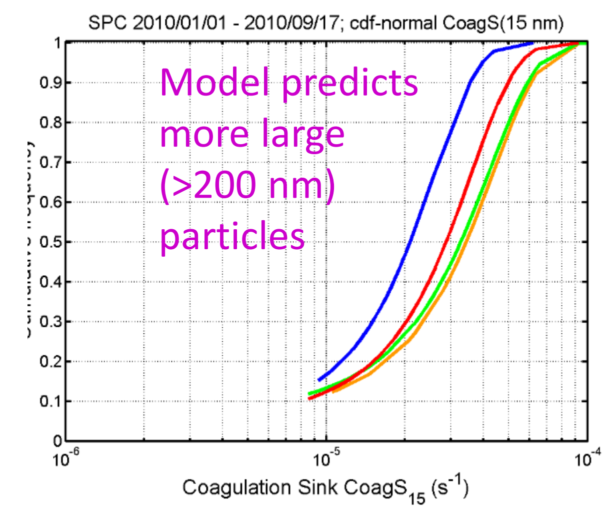
-ARO




***Cumulative distribution functions only from event days.***
***Smooth appearance due to assumption of normal distributed data***

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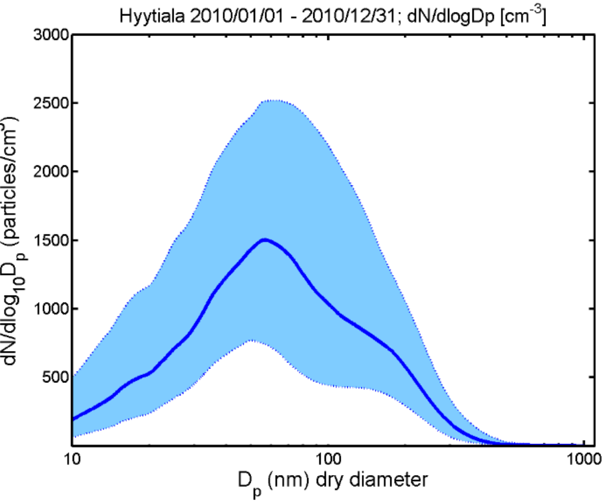
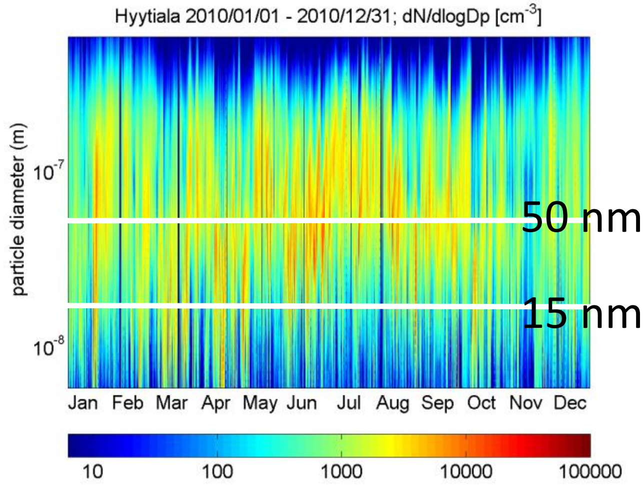
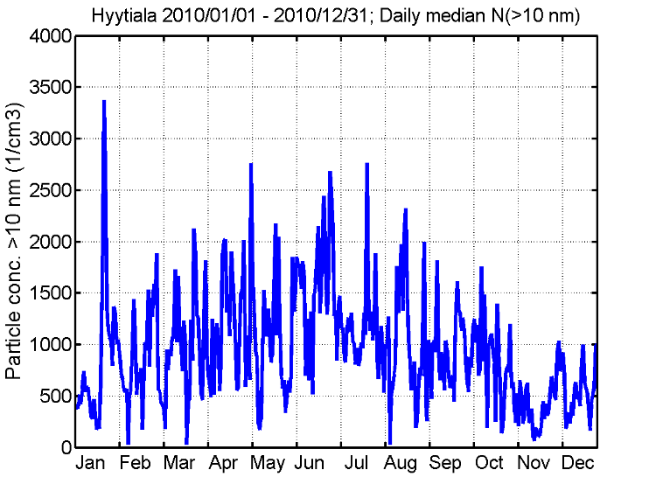
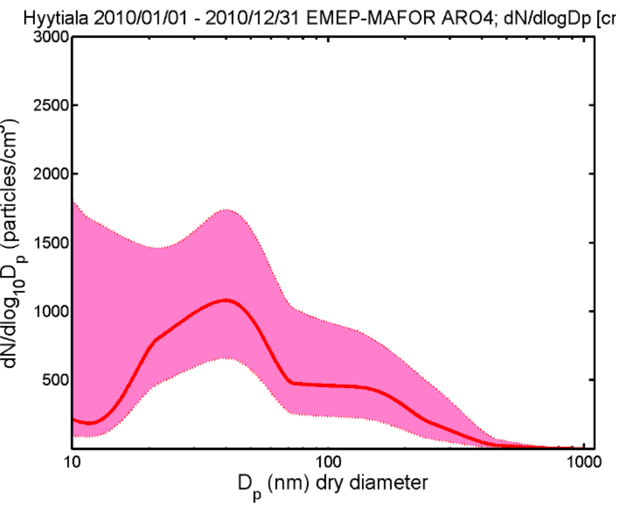
# Hyytiälä 2010 - Observations

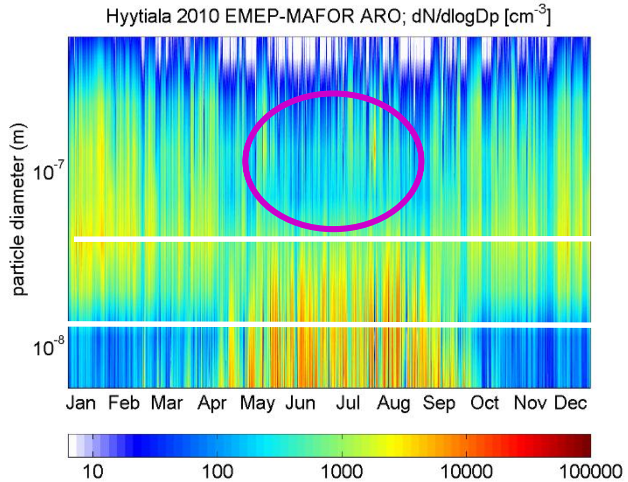


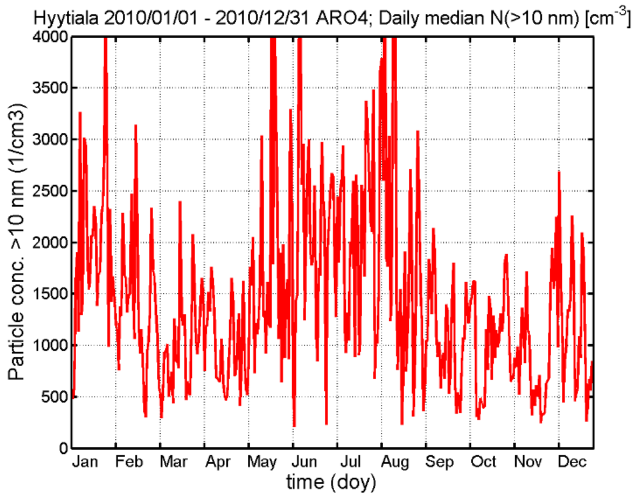




# Hyytiälä 2010-ARO

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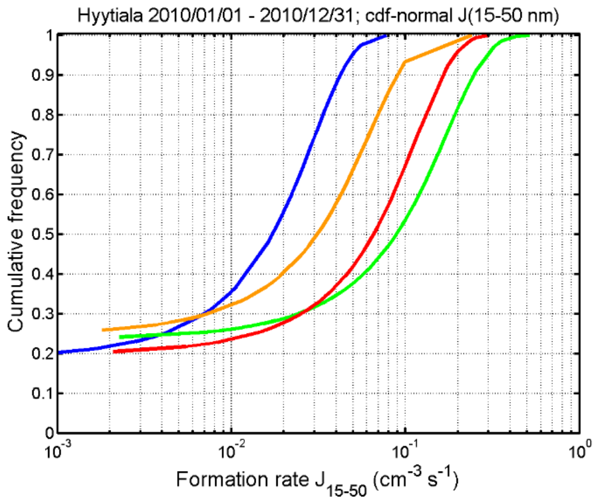
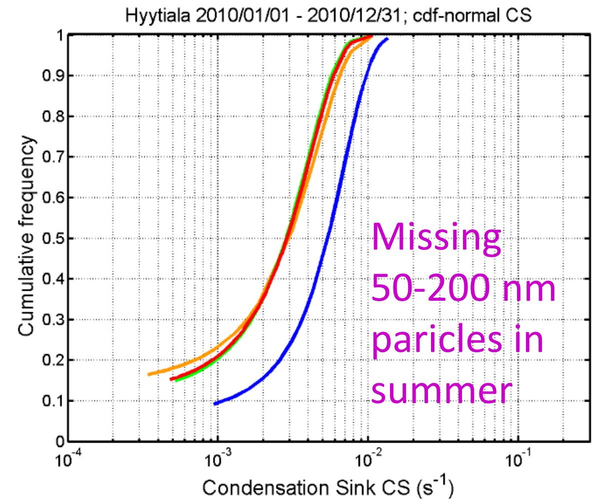


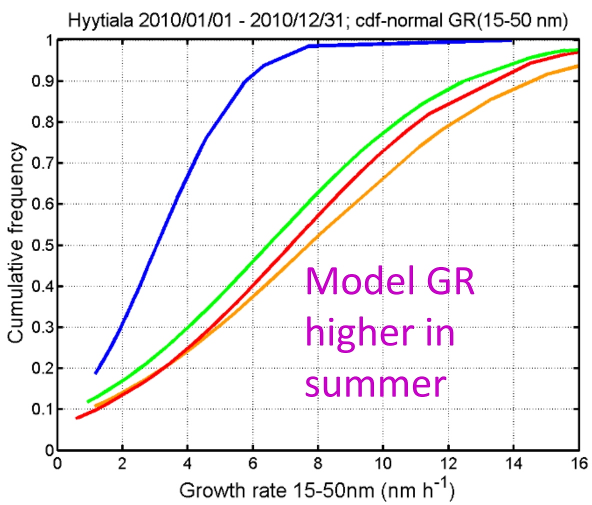

# Hyytiälä 2010

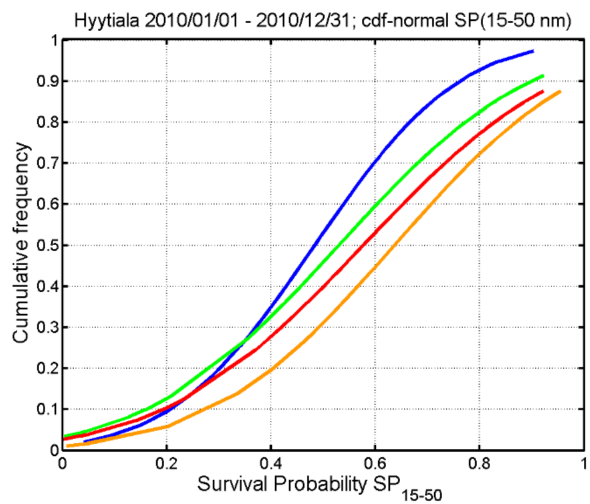
-SO4

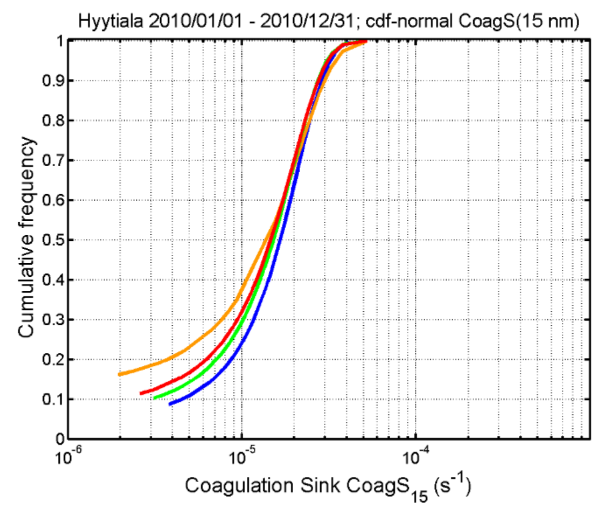
-BIO

-ARO

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***Cumulative distribution functions only from event days.***
***Smooth appearance due to assumption of normal distributed data***

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# «Golden Day Events» Hyytiälä, 24-30 March 2003

**Observations:**

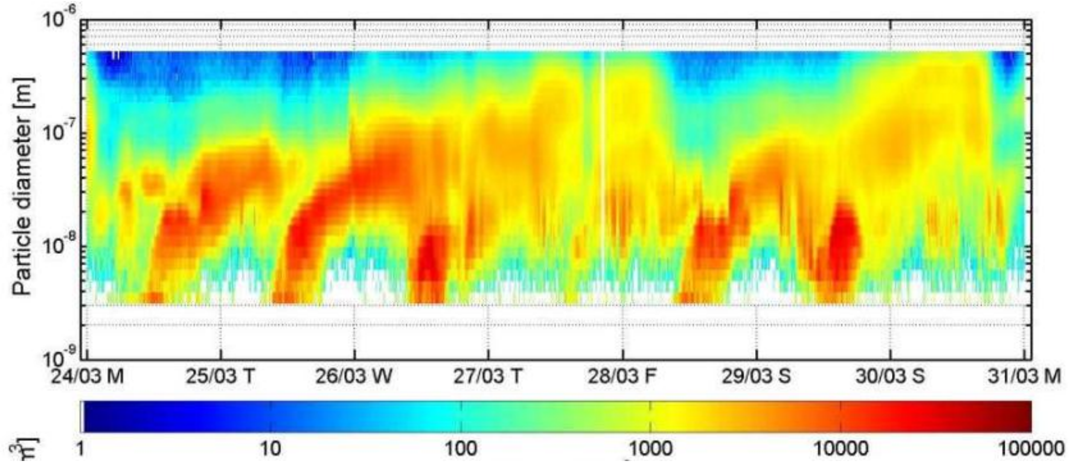
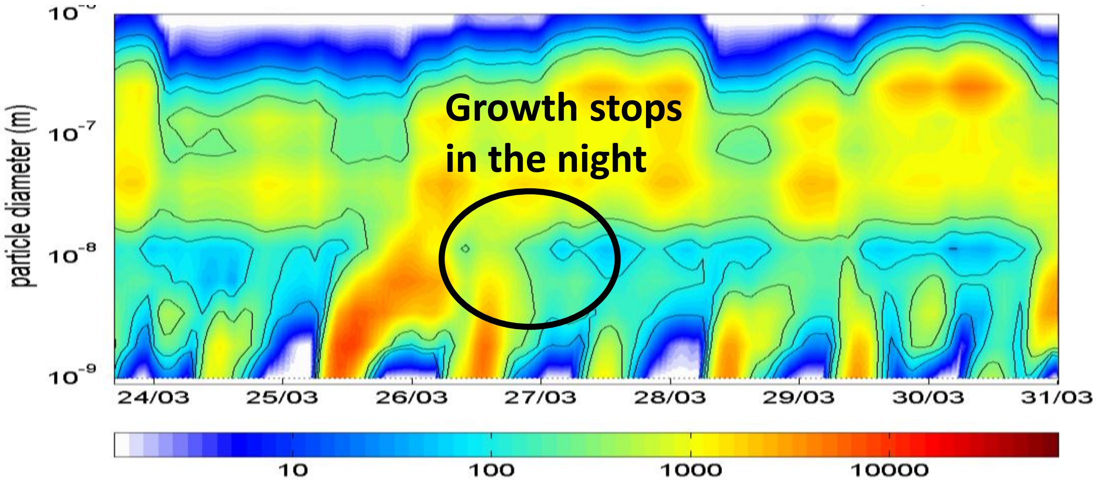



**Figure from: O'Dowd et al., ACP, 2009**

**Model (Ref):**



# Summary of main findings

- Missing particle sources in winter and in summer
  - Wood burning (domestic heating) in winter
  - Agriculture biomass burning in summer
  - SOA from VBS model is currently missing
- Based on the event analyses it cannot be decided whether biogenic or aromatic VOC are involved in nucleation. On average, nucleation involving biogenic organics results in more events than observed.
- Underestimation of 50-200 nm particles in summer at remote sites, possibly due to:
  - Model predicts too many events in summer with too high GR
  - BVOC chemistry at night identified as possible reason for interruption of particle growth

# Acknowledgement

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- Maija Kajos (Univ. Helsinki) for SMEAR data on VOC
- Tuomo Nieminen (Univ. Helsinki) and Michael Boy (Univ. Helsinki) for SMEAR data on H2SO4 and OH
- Jorma Joutsensaari (UEF) and Ari Laaksonen (FMI) for SPC data
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- Matthias Karl thanks NILU for financial support

**Thank you for your kind attention!**

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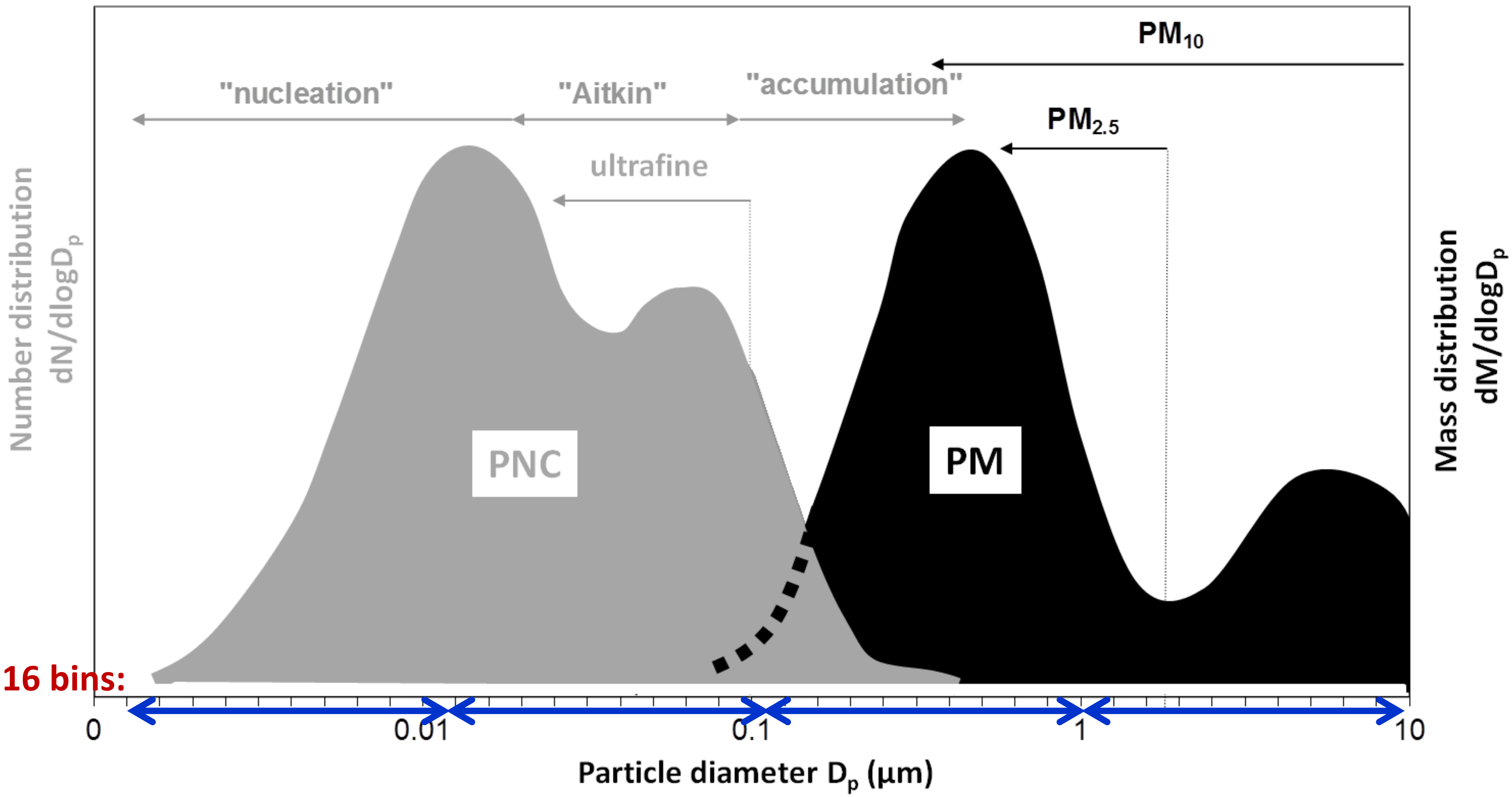
# Appendix

# PNC and PM size-resolved definition




| $PM_{XX}$: | 0.008 | 0.080 | 0.300 | 3 | GMD(µm) |
|---|---|---|---|---|---|
| | 1.45 | 1.5 | 1.55 | 1.80 | σ |

(PPM emissions, dry / wet deposition)

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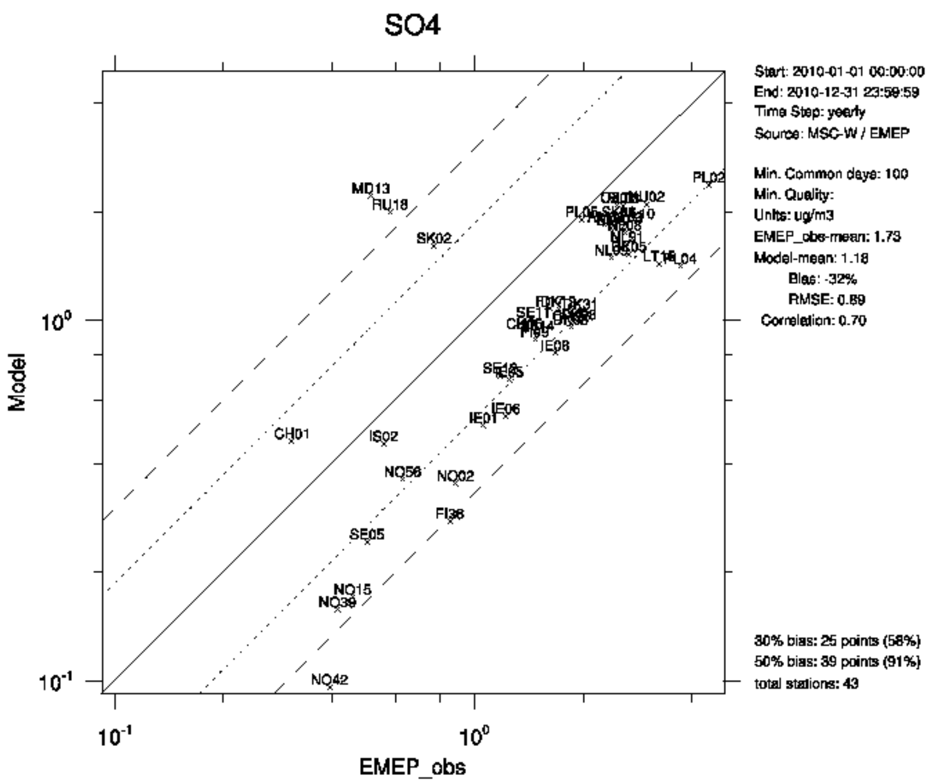
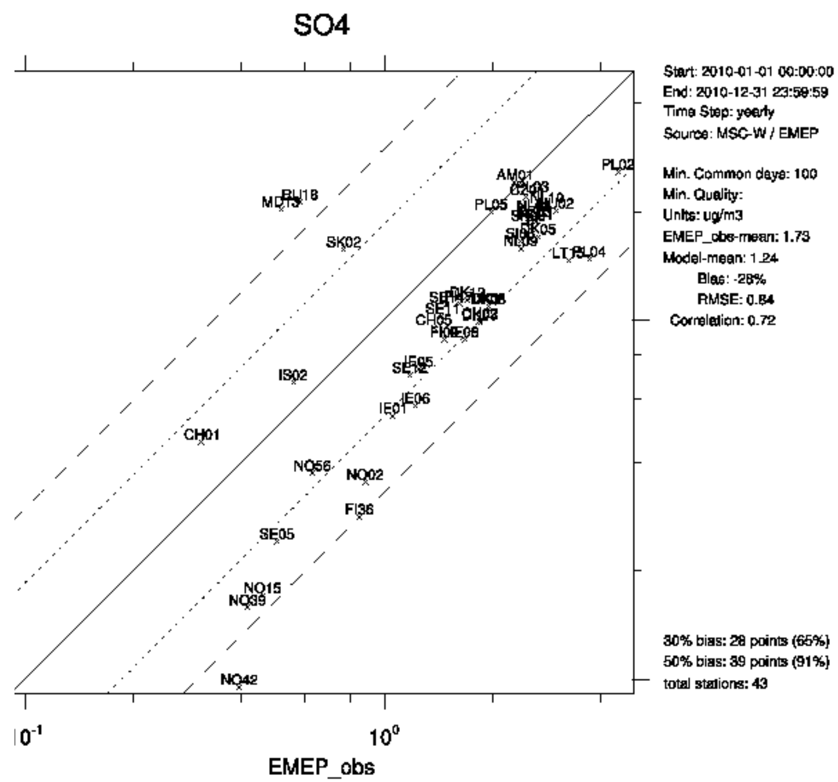
# Bulk vs. Size-resolved Sulphate

# 2010 mean compared to EMEP-stations




## Bulk

Correlation: 0.70

## Size-resolved

Mean 5% higher

Correlation: 0.72

# MAFOR Gas-phase reactions

- **SOA precursors: isoprene, a-pinene, o-xylene**

| **Educts** | **Products** | **Rate constant** |
|---|---|---|
| C5H8 + OH | ISRO2 (peroxy radical of isoprene) | 2.7e-11*exp(390.0/*T*) |
| [ISRO2] + [NO] | 0.003 BLOC + 0.101 BSOC | KRO2NO |
| [ISRO2] + [HO2] | 0.024 BLOC + 0.119 BSOC | 0.706·KHO2RO2 |
| α-pinene + O3 | 0.2 BLOC + 0.8 TERPRO2 (peroxy radical of monoterpenes) | 6.3e-16*exp(-580.0/*T*) |
| α-pinene + OH | TERPRO2 | 1.2e-11*exp(444.0/*T*) |
| [TERPRO2] + NO | 0.052 BLOC + 0.184 BSOC + 1.6 HCHO + MGLYOX + 0.6 MAL + NO2 | KRO2NO |
| [TERPRO2] + [HO2] | 0.327 BLOC + 0.180 BSOC + 1.0 HCHO + MGLYOX + 0.9 CO | KHO2RO2 |
| BSOC + [OH] | BLOC | 4.0e-11 |
| OXYL + OH | OXYRO2 (peroxy radical of aromatics) | 1.36e-11 |
| OXYRO2 + NO | 0.003 ALOC | KRO2NO |
| OXYRO2 + HO2 | 0.106 ALOC | KHO2RO2 |

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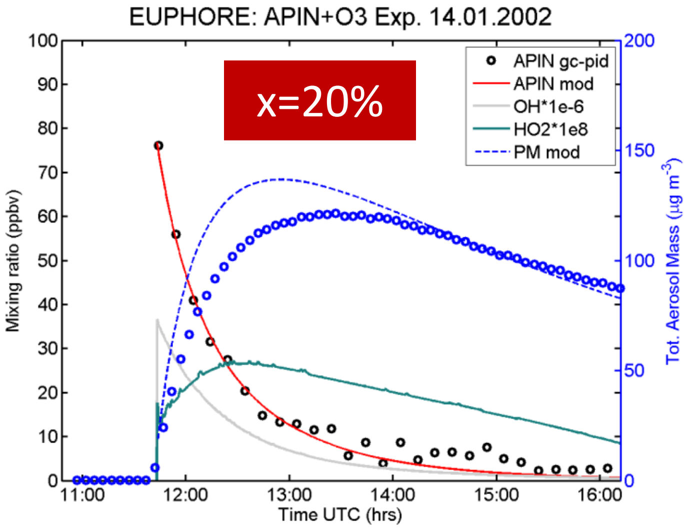
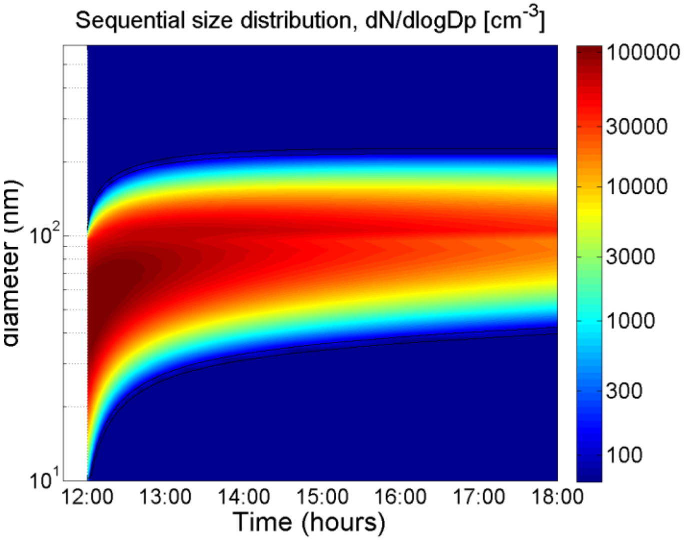
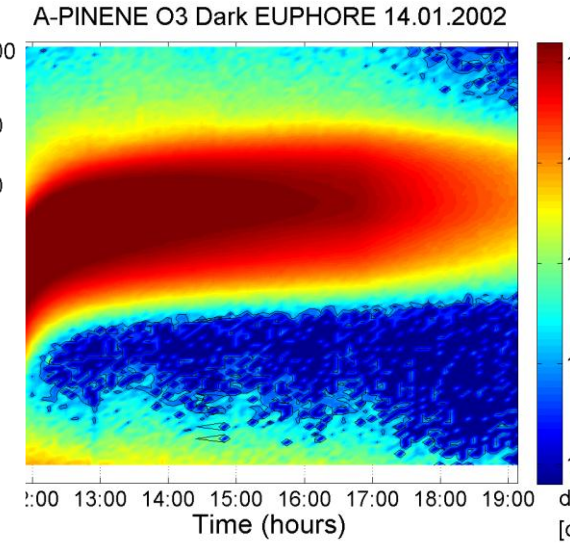
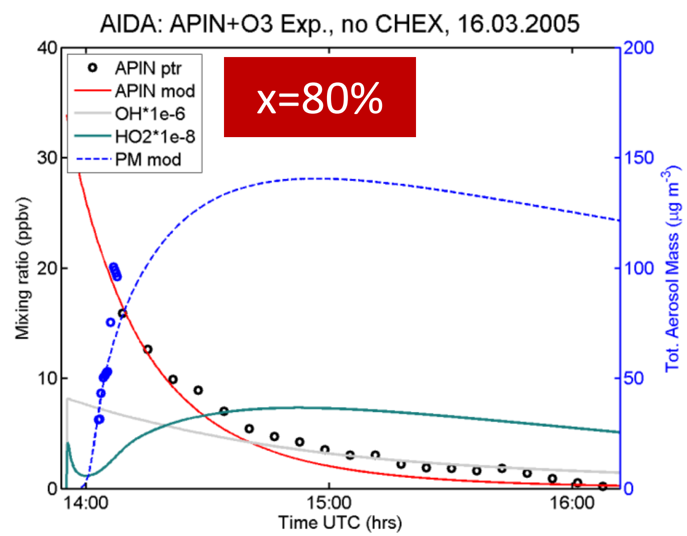
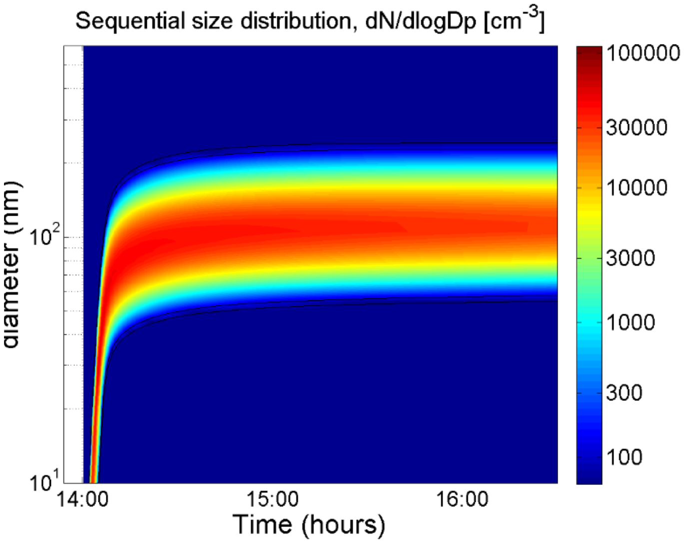
# Chamber SOA: a-pinene+O3 → x BLOC+ (1-x)TERPRO2

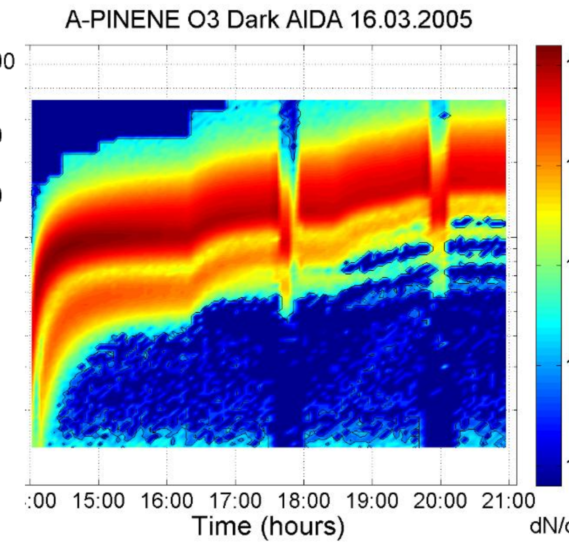
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