Atmospheric aerosols and its impact on climate and health

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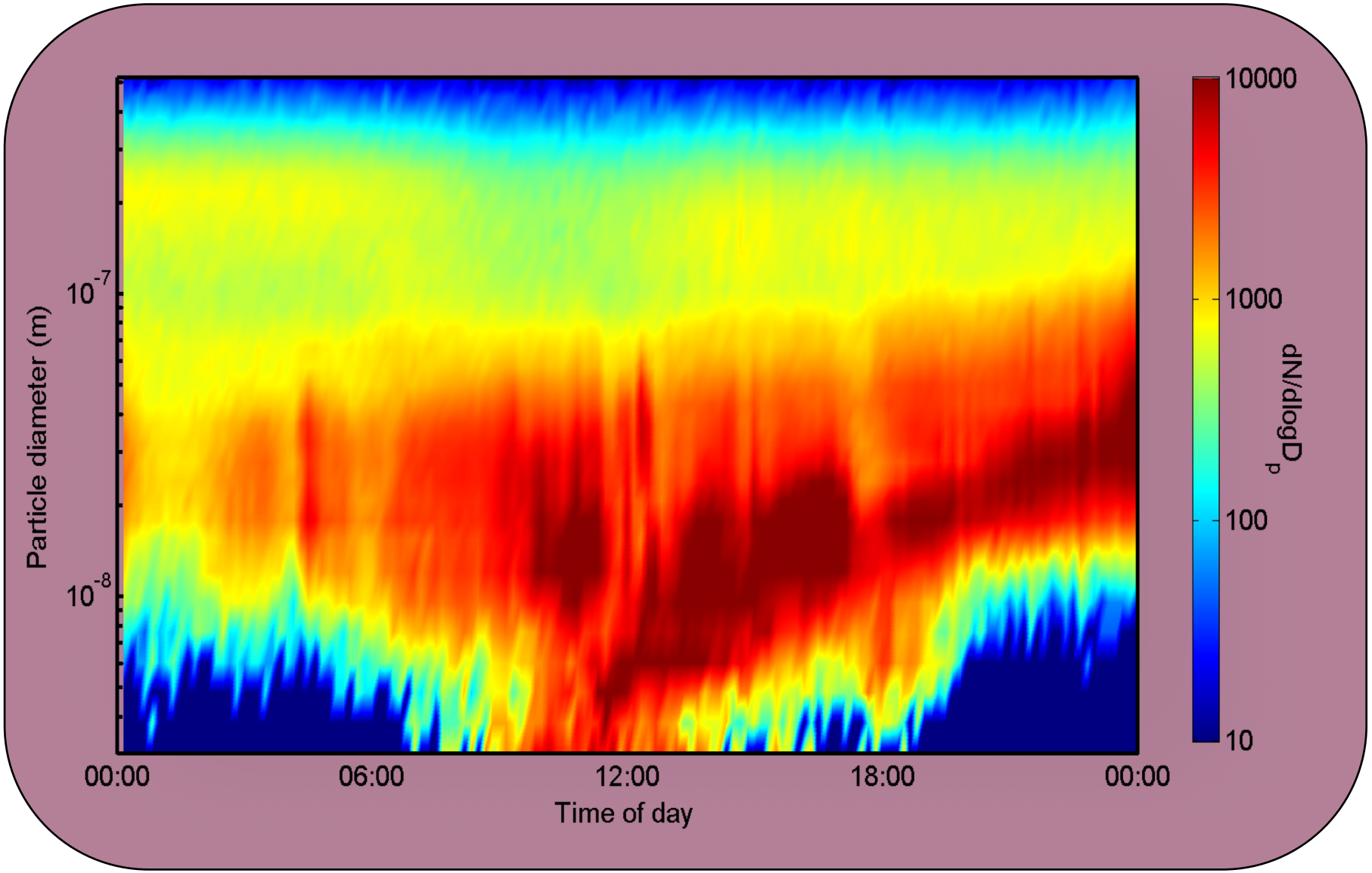
Why we care about aerosols in the atmosphere?

- visibility
- health (respiration)
- radiative balance
- cloud formation
- heterogeneous reactions
- delivery of nutrients
- disease carrier
- …..


Particle diameter (m)
10^-7
10^-8
00:00
06:00
12:00
18:00
00:00
Time of day
10000
1000
100
10
dN/dlogD_p

Cluster – tiny aerosol particles

Ultrafine particles with Dp<100nm

Semi, low and extreme low volatile organic compounds

H_2SO_4, NH_3, DMA

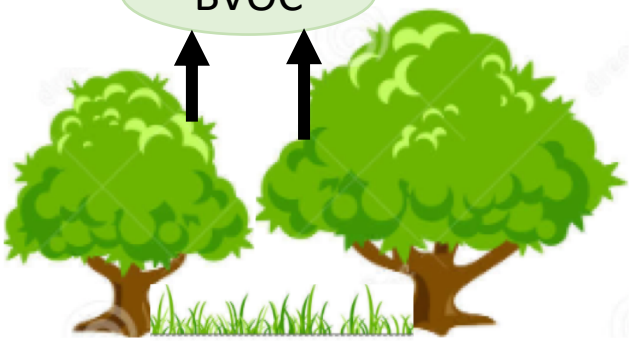
Reaction with OH, O_3 and NO_3 - Autoxidation

$$C_zH_yO_xO_2\bullet \xrightarrow{\text{H-shift + } O_2} C_zH_yO_{x+2}O_2\bullet \xrightarrow{\text{H-shift + } O_2} C_zH_yO_{x+4}O_2\bullet$$

Emissions of ammonia and amines from anthropogenic and biogenic sources

AVOC, SO_2, NO_x, etc

BVOC



Formation of highly oxidized multifunctional (HOM; O:C≥0.7) organic compounds by autoxidation of monoterpenes (MT)

$MT\ (C_{10}H_{16}) + O_3 \nrightarrow \gamma C_{10}H_{15}O_2\text{-}OO\cdot$ (γ = 9 % for α-pinene)

$C_{10}H_{15}O_2\text{-}OO\cdot \xrightarrow{\text{H-shift}+O_2} C_{10}H_{15}O_4\text{-}OO\cdot$ (1st H-shift + O_2)

$C_{10}H_{15}O_4\text{-}OO\cdot \xrightarrow{\text{H-shift}+O_2} C_{10}H_{15}O_6\text{-}OO\cdot$ (2nd H-shift + O_2)

$C_{10}H_{15}O_6\text{-}OO\cdot \xrightarrow{\text{H-shift}+O_2} C_{10}H_{15}O_8\text{-}OO\cdot$ (3rd H-shift + O_2)

$C_{10}H_{15}O_8\text{-}OO\cdot \xrightarrow{\text{H-shift}+O_2} C_{10}H_{15}O_{10}\text{-}OO\cdot$ (4th H-shift + O_2)

Reactions terminating the autoxidation, leading to formation of stable HOMs:

$C_{10}H_{15}O_x\text{-}OO\cdot + HO_2 \rightarrow C_{10}H_{15}O_x\text{-}OH$

$C_{10}H_{15}O_x\text{-}OO\cdot + RO_2 \nrightarrow \alpha\cdot C_{10}H_{15}O_x\text{-}OH + \beta\cdot C_{10}H_{14}O_x{=}O + \gamma\cdot C_{10}H_{15}O_x\text{-}O\cdot + (1\text{-}\alpha\text{-}\beta\text{-}\gamma)\cdot C_{20}H_{30}O_{x+4}$

$C_{10}H_{15}O_x\text{-}OO\cdot + NO \rightarrow \alpha\cdot C_{10}H_{15}O_x\text{-}ONO_2 + \beta\cdot C_{10}H_{14}O_x{=}O + \gamma\cdot C_{10}H_{15}O_xO\cdot + (1\text{-}\alpha)\cdot NO_2$

$C_{10}H_{15}O_xOO\cdot \rightarrow C_{10}H_{15}O_xOH$

We have developed a HOM mechanism that comprise 1773 reactions and 208 new compounds.

The mechanism has been coupled to the Master Chemical Mechanism v 3.3

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ARTICLE

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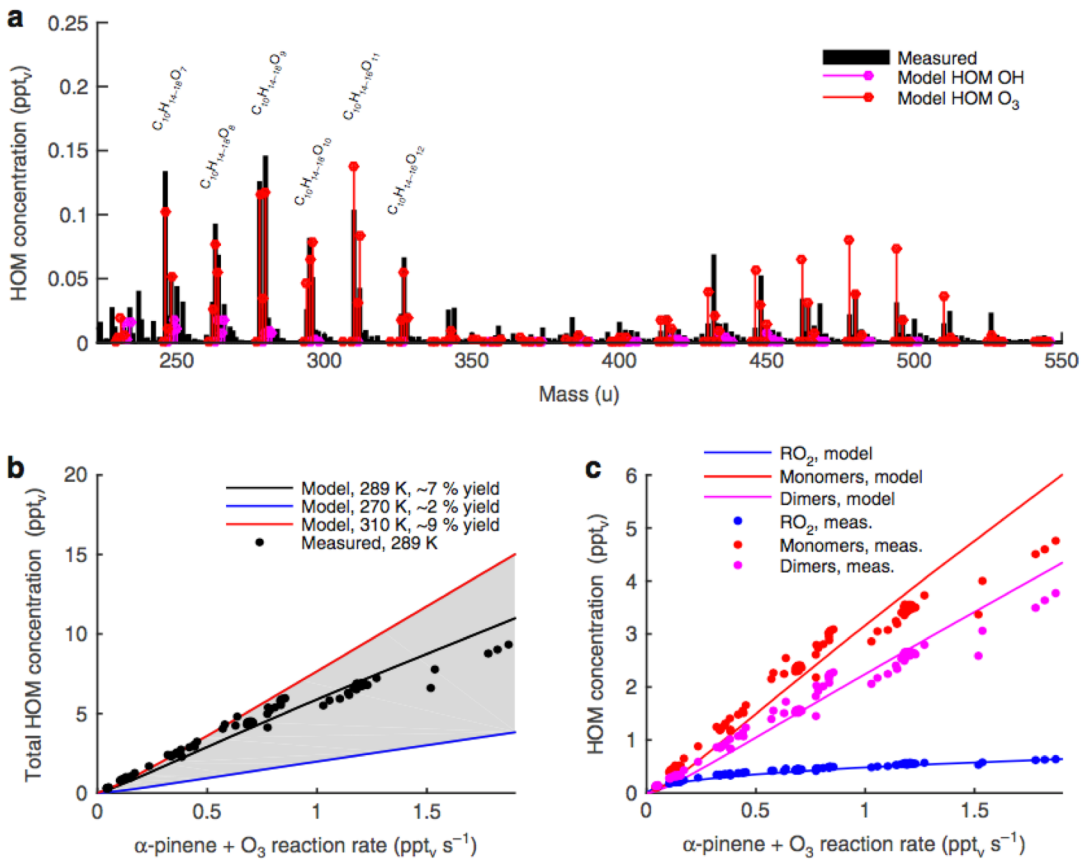
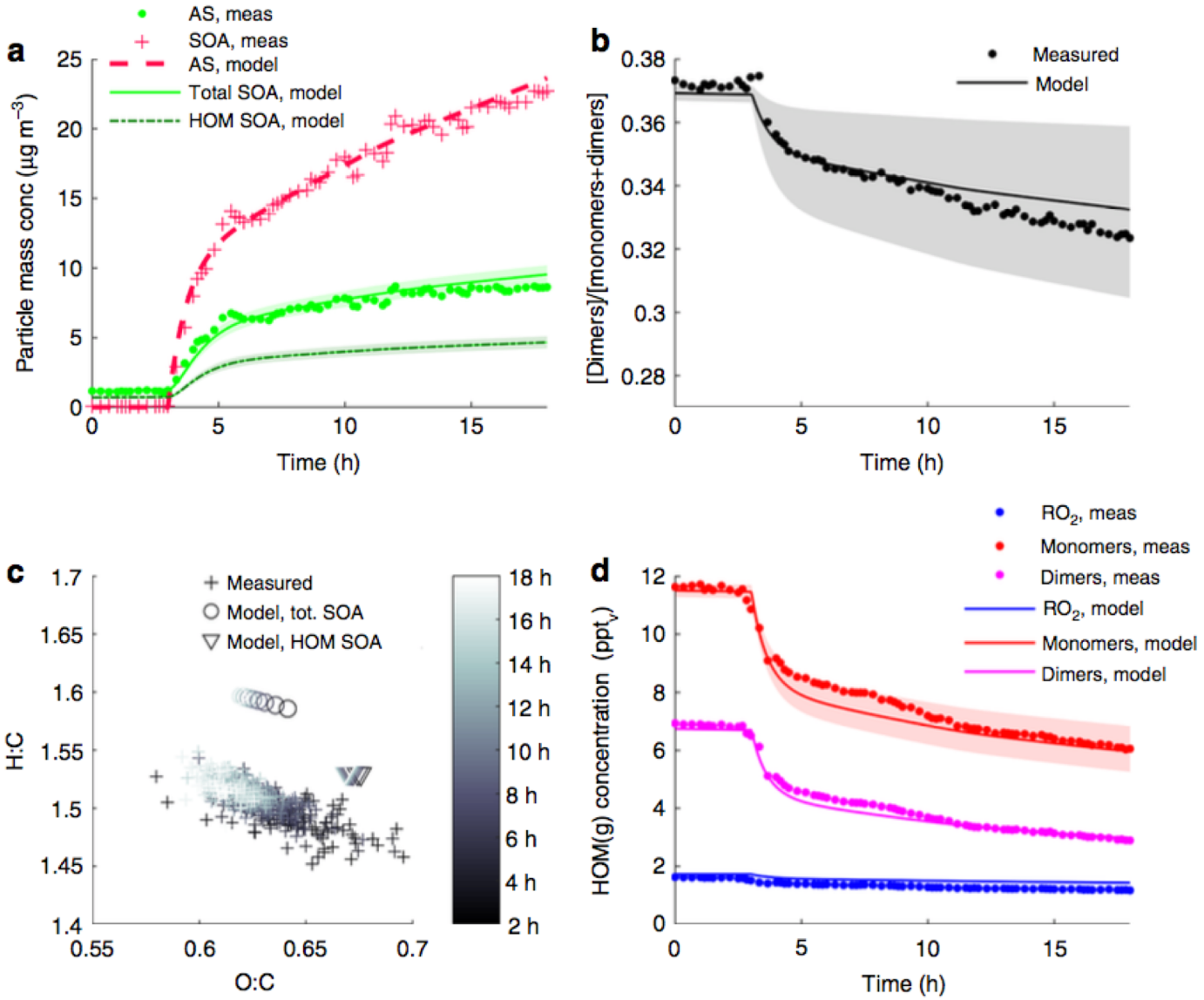
The role of highly oxygenated organic molecules in the Boreal aerosol-cloud-climate system

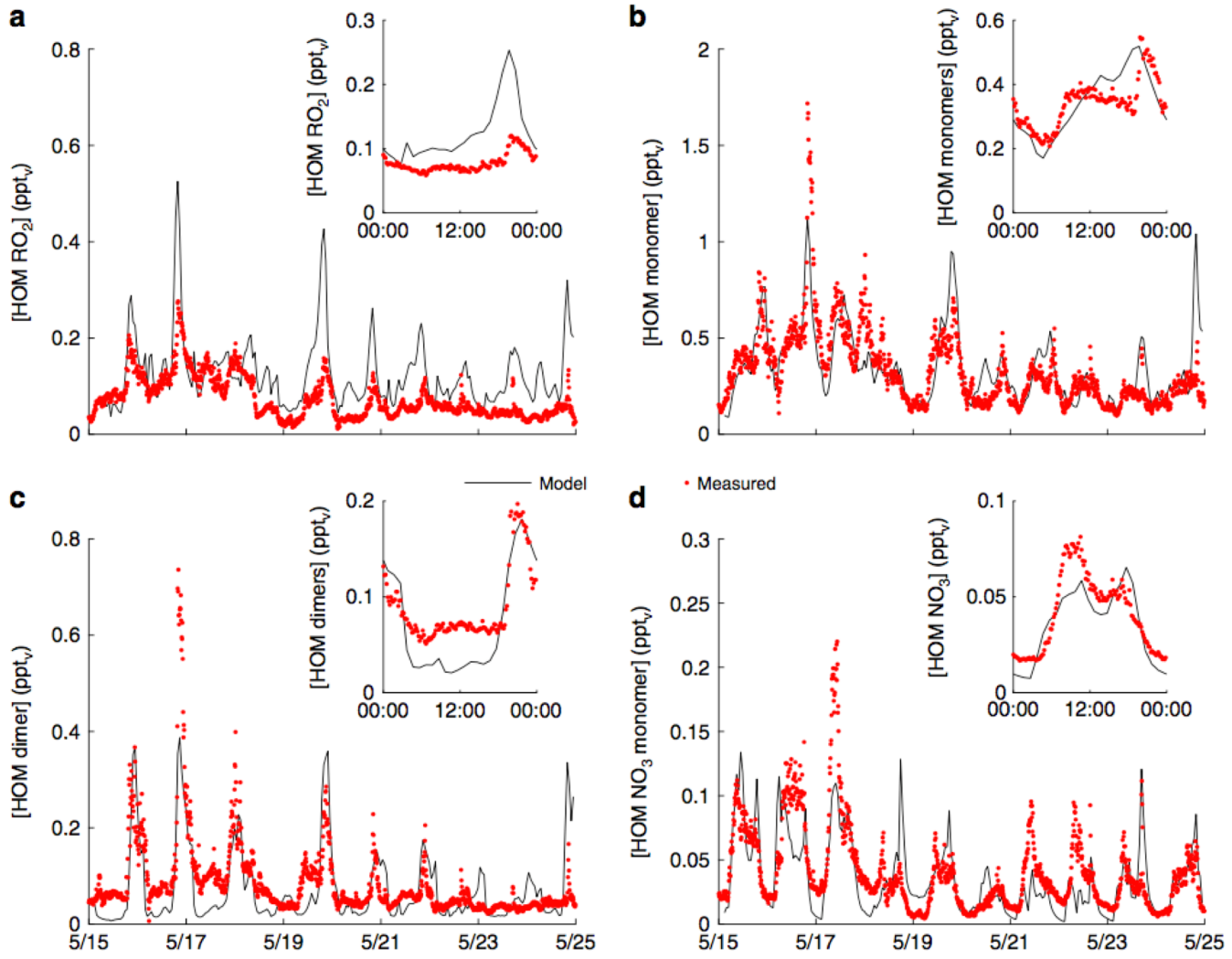
Pontus Roldin[1*], Mikael Ehn[2], Theo Kurtén[3], Tinja Olenius[4], Matti P. Rissanen[2], Nina Sarnela[2], Jonas Elm[5], Pekka Rantala[2], Liqing Hao[6], Noora Hyttinen[7], Liine Heikkinen[2], Douglas R. Worsnop[2,8], Lukas Pichelstorfer[2,9], Carlton Xavier[2], Petri Clusius[2], Emilie Öström[1], Tuukka Petäjä[2], Markku Kulmala[2], Hanna Vehkamäki[2], Annele Virtanen[6], Ilona Riipinen[4] & Michael Boy[2]

Highly oxygenated organic molecule (HOM) formation from α-pinene. Modelled and measured HOM(g) concentrations during a JPAC α-pinene ozonolysis experiment



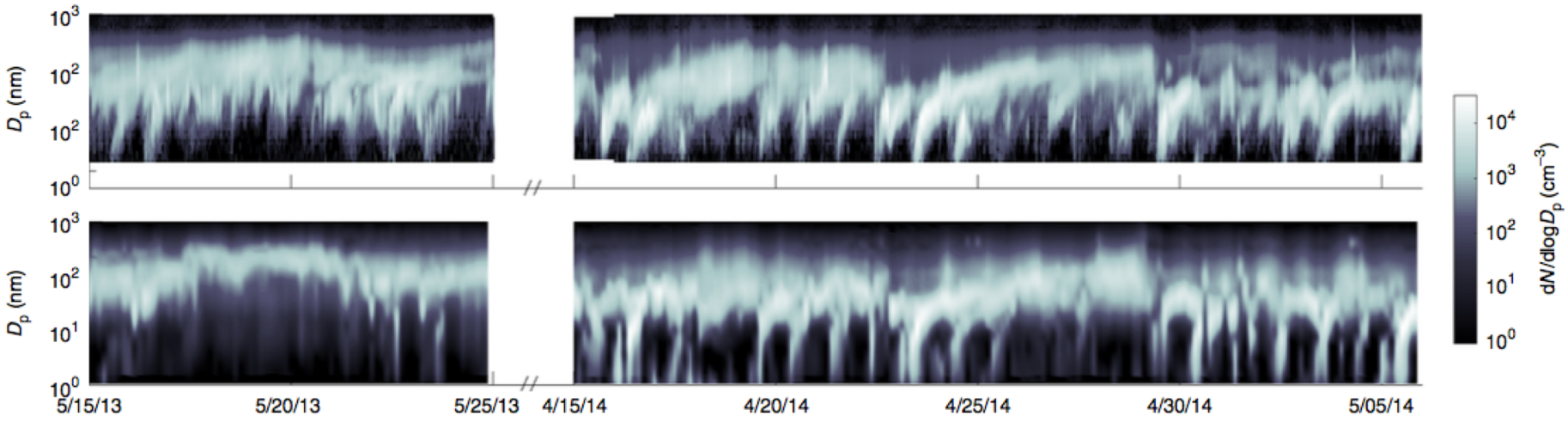


Highly oxygenated organic molecule (HOM) gas-particle partitioning. Model and measurement results from an α-pinene ozonolysis experiment with ammonium sulfate (AS) seed particles.



Highly oxygenated organic molecules (HOM) in the boreal forest. Modelled and measured HOM gas-phase concentrations at the Station for Measuring Ecosystem-Atmosphere Relations II (SMEAR II) between 15 and 24 May 2013.

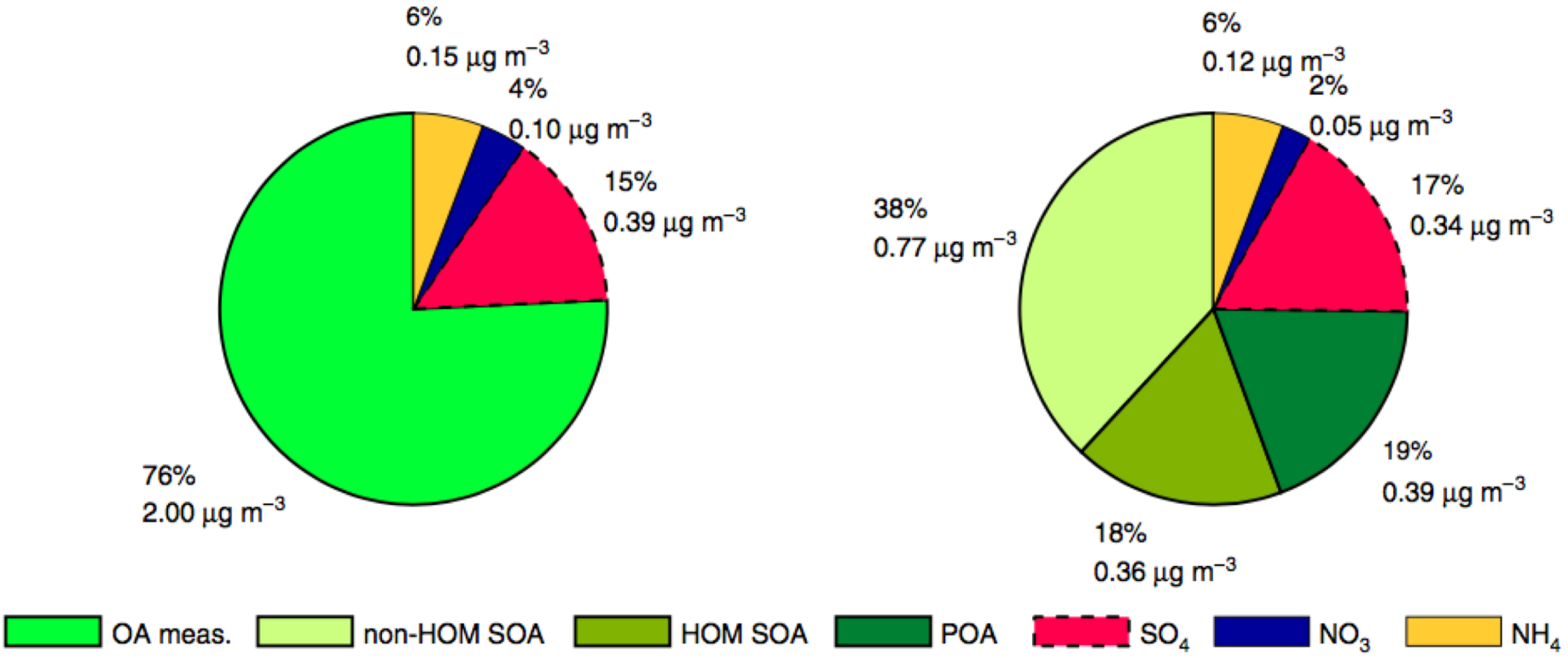
Measured and modelled particle number concentrations at the SMEAR II (Hyytiälä, Finland)



Upper plot: measurements
Lower plot: model

Roldin et al., Nature communication, 2019

Average non-refractory submicron particle chemical composition



Left plot: measurements
Right plot: model

Roldin et al., Nature communication, 2019

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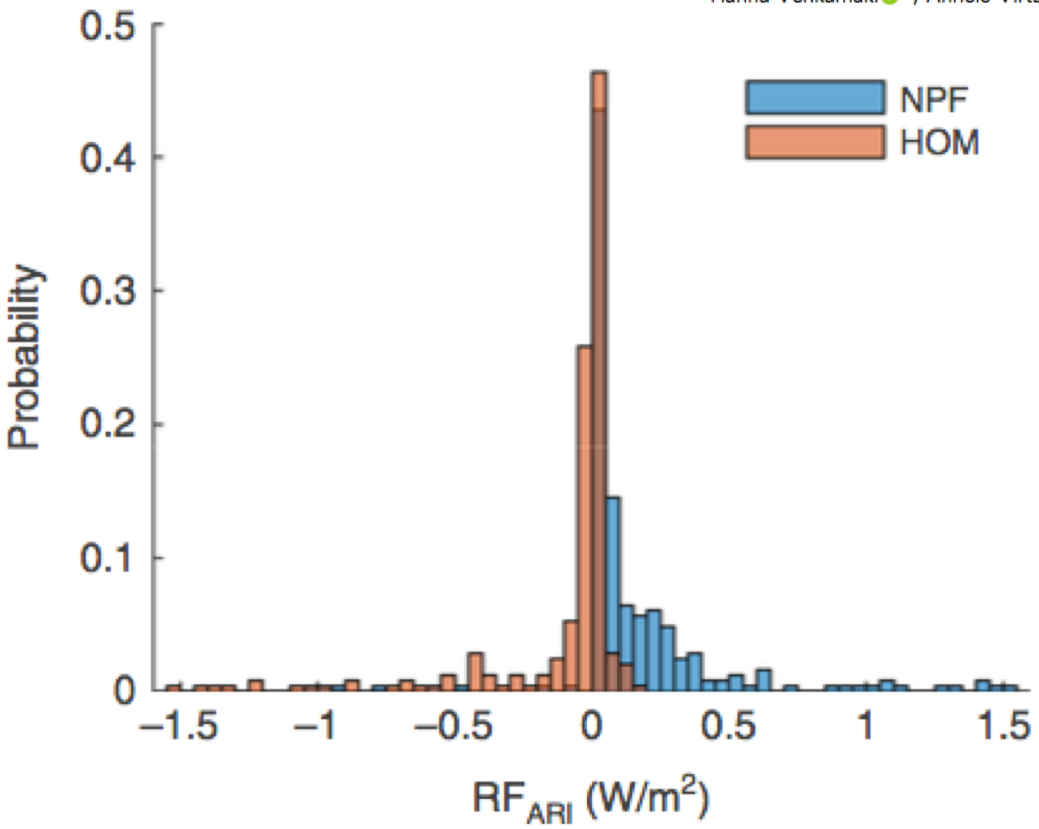

Modelled top of the atmosphere direct aerosol radiative forcing probability distributions caused by new particle formation (NPF) and HOM secondary organic aerosol (HOM SOA) formation, during clear sky conditions.