Hygroscopicity prediction Impacted by particle Mixing in Sub-urban environment (HIMS)

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Leibniz Association

ACROSS – Atmospheric Chemistry of the Suburban Forest

AAAR American Association for Aerosol Research

LSCE

TROPOS Leibniz Institute for Tropospheric Research

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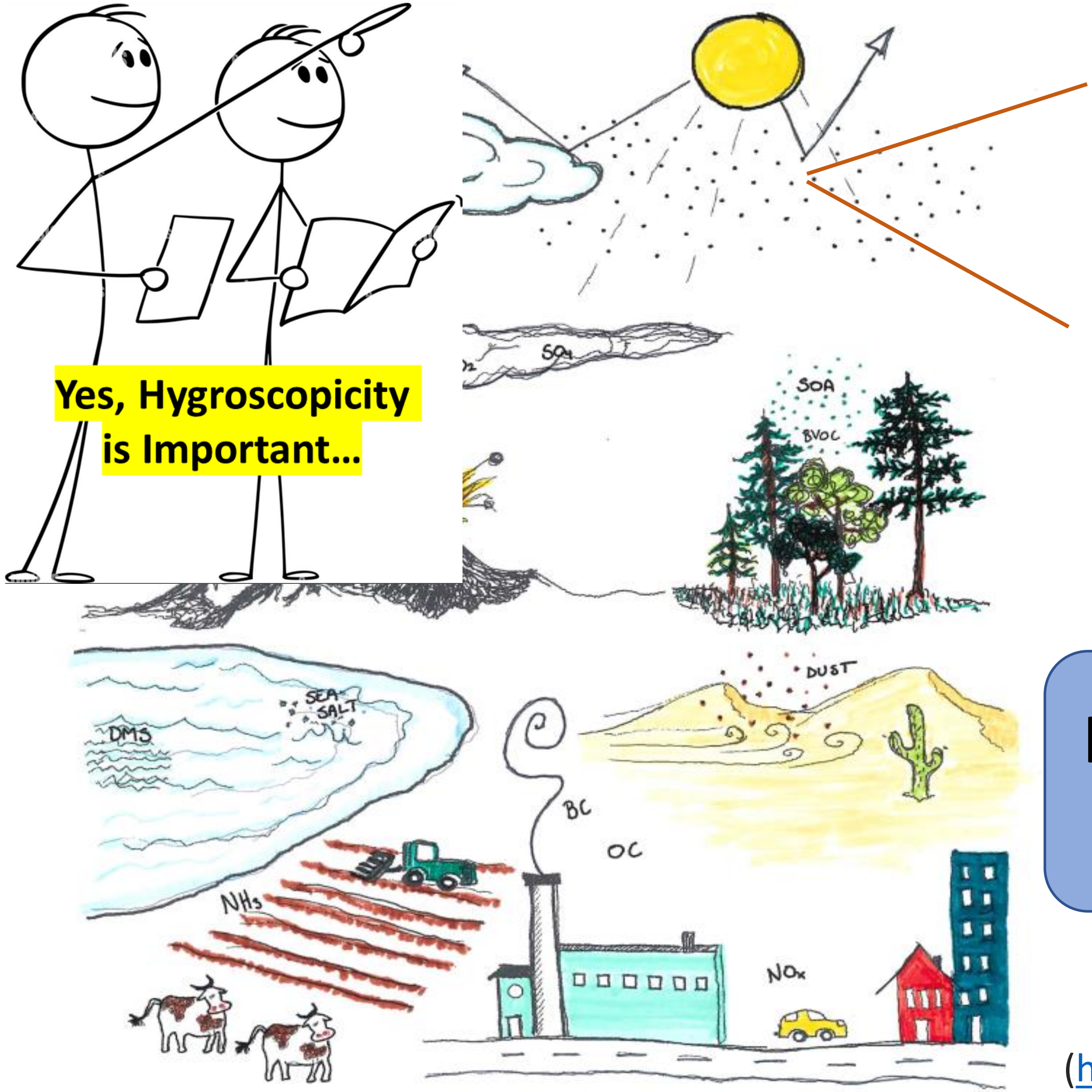
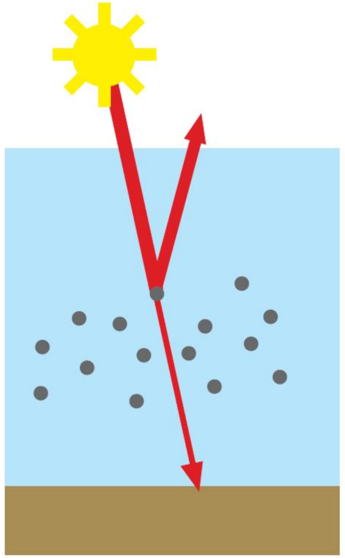
Introduction

Hygroscopicity





Aerosol direct effect
Scattering and absorption of solar radiation

- **Hygroscopicity** is a particle's ability to **absorb moisture / take up water** from the environment.
- Subsaturate – Relative Humidity (RH) ≤ 100% affects the **aerosol size, phase and chemical properties.**

Hygroscopicity primarily depends on aerosol size and chemical composition

(https://www.noresm.org/)

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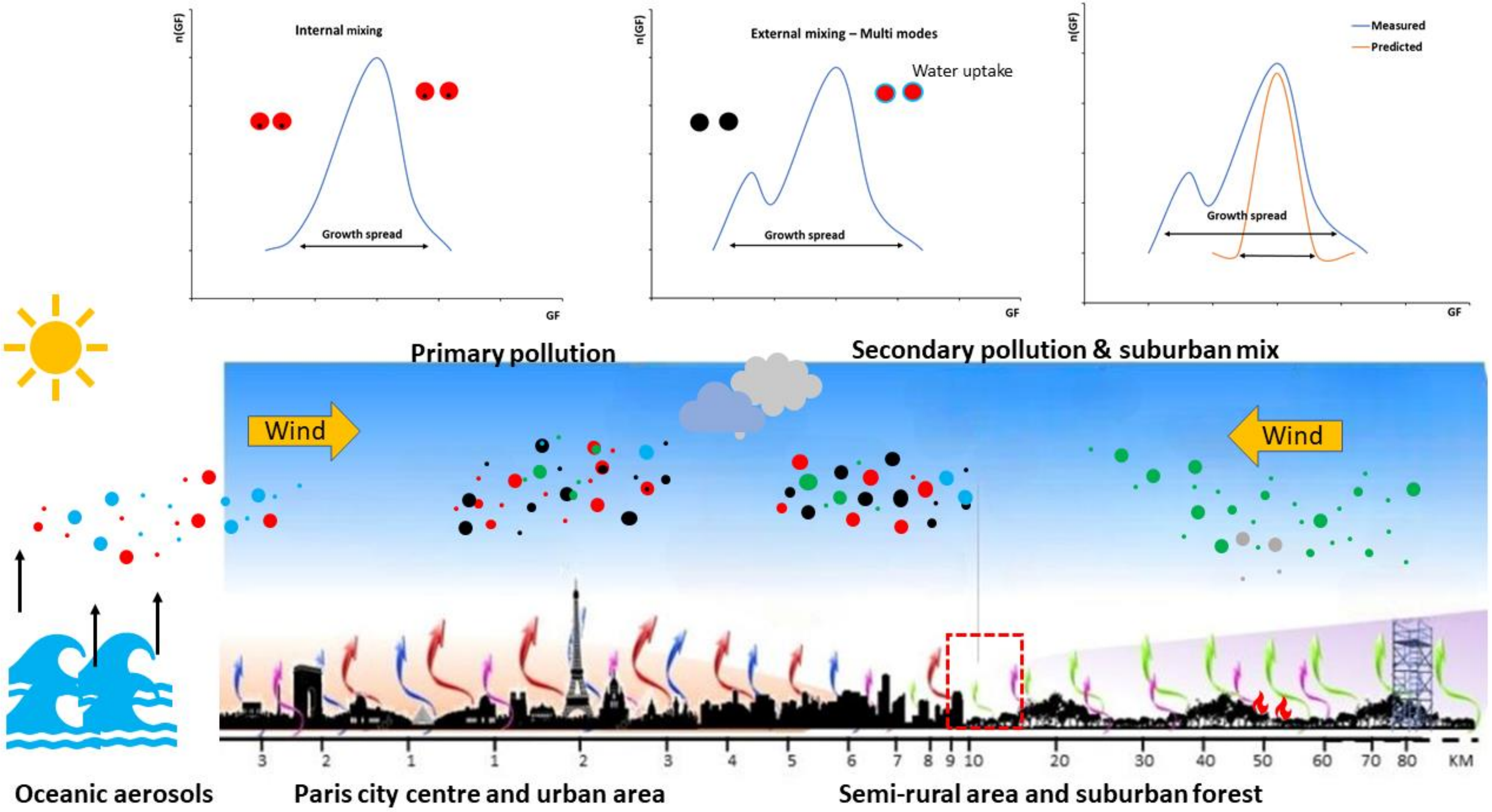
Overview of the ACROSS campaign



- Illustrative figure showing the general behaviour of hygroscopicity over the measurement site during the ACROSS 2022 campaign at LSCE-SIRTA.

- ACROSS (**A**tmospheric **C**hemist**R**y **O**f the **S**uburban fore**S**t)

  (13 June – 25 July 2022)

- Aim: To understand detailed chemistry and physics of urban airmass mixed with biogenic emissions. https://across.cnrs.fr/

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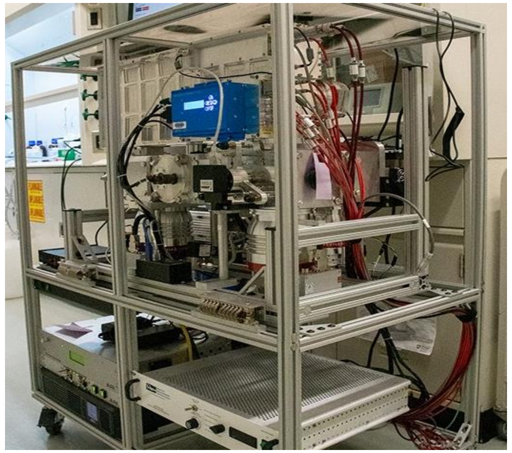
Methodology

Steps and Instrumentation

(a)

(c)

(b)



- HTDMA was run for four different diameters (100,150,200 and 250nm at RH = 90%± 3)
- **Collocated measurements at SIRTA**: On-site ACSM, Aethalometer and PTR-CHARON.
- Processed the data and applied the ZSR rule for closure study between measured and predicted hygroscopicity.

(a) HTDMA - Hygroscopicity / Humidified Tandem Differential Mobility Analyser
(b) HR-ToF-AMS - High Resolution-Time of Flight-Aerosol Mass Spectrometer, figure DeCarlo et al., 2006
(c) MPSS - Mobility Particle Size Spectrometer (a.k.a SMPS)

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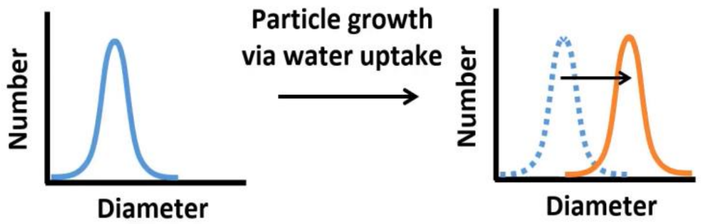
Hygroscopicity parameter Kappa (κ)

- Hygroscopic growth factor (GF)

$$GF\ (RH, D_0) = \frac{Dp\ (RH)}{D_0} \qquad (1)$$

$$\kappa_{measured} = (GF^3 - 1)\left(\frac{exp\left(\frac{A}{D_p dry \cdot GF}\right)}{RH} - 1\right) \qquad (2)$$



- Illustrative of particle growth diameter (D_P)

- The Zdanovskii-Stokes-Robinson (ZSR) mixing rule

$$\kappa_{chem} = \sum_i V_{f\,i} \,.\, \kappa_i \qquad (3)$$

- Illustrative of ZSR mixing rule

$A = \frac{4\,\sigma_{s/a} M_w}{RT\rho_w}$, *Petters & Kreidenweis et al., 2007*

Kappa derived from Köhler's theory.

D_p dry = the initial dry particle diameter; $\sigma_{s/a}$ = the droplet surface tension (0.0728 Nm^{-2}); M_w = molecular weight of water; ρ_w = the density of liquid water; R = universal gas constant; T = absolute temperature.

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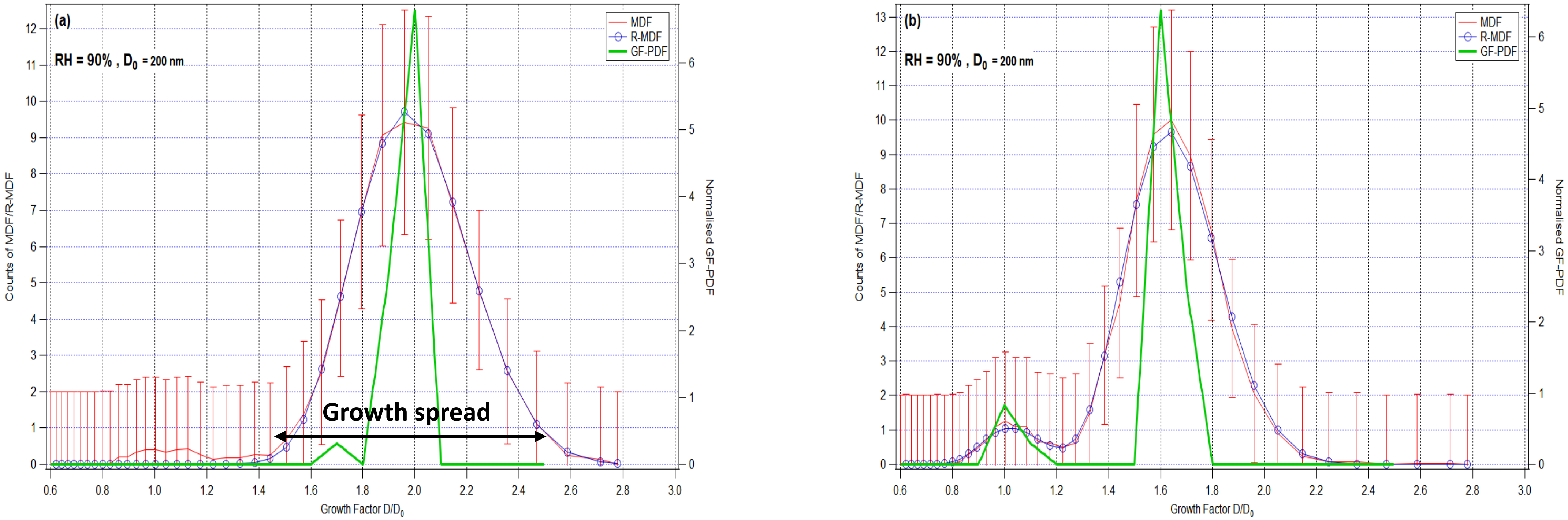
Sigma growth spread for mixing state



The standard deviation (σ) is used to measure the growth factor's spread to describe the mixing state *(Sjogren et al., 2008)*. The inverted data were grouped into two cases, representing the aerosol mixing.

Specifically, σ ≤ 0.08 indicates an internally mixed aerosol (Fig. a),

σ ≥ 0.10 describes an externally mixed aerosol with two distinct modes (Fig. b)

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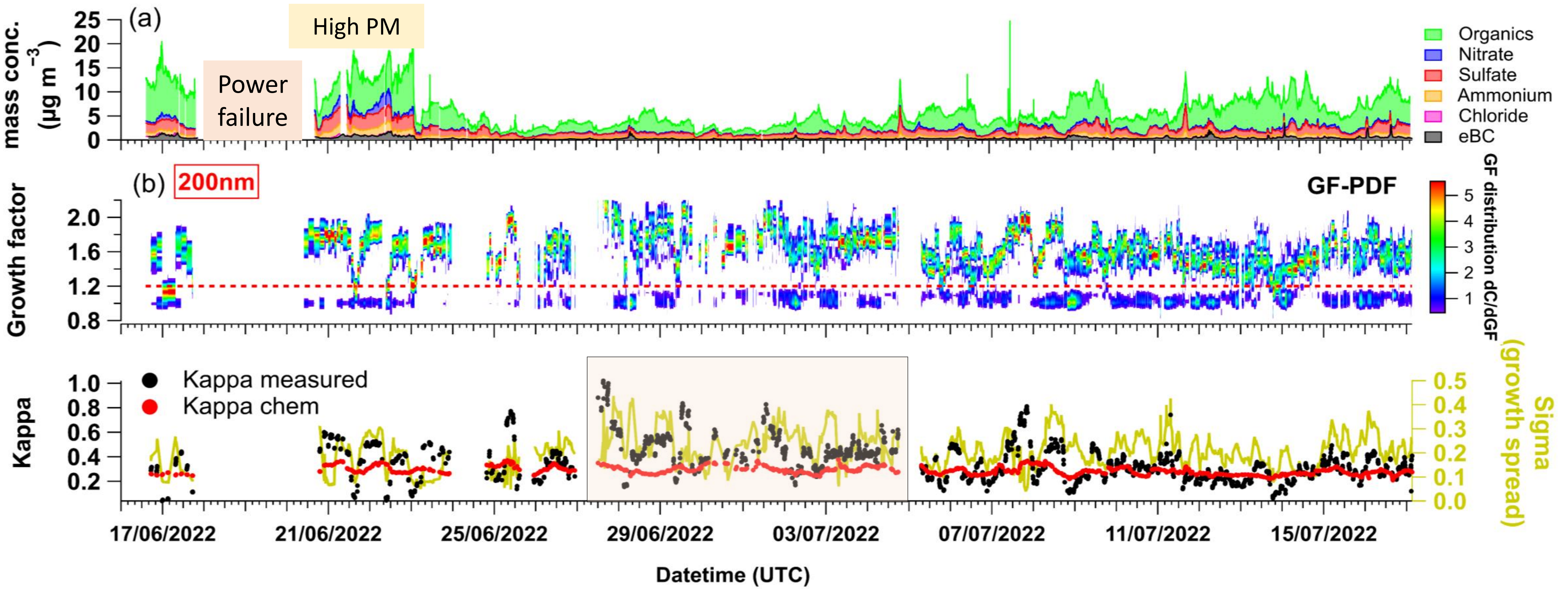
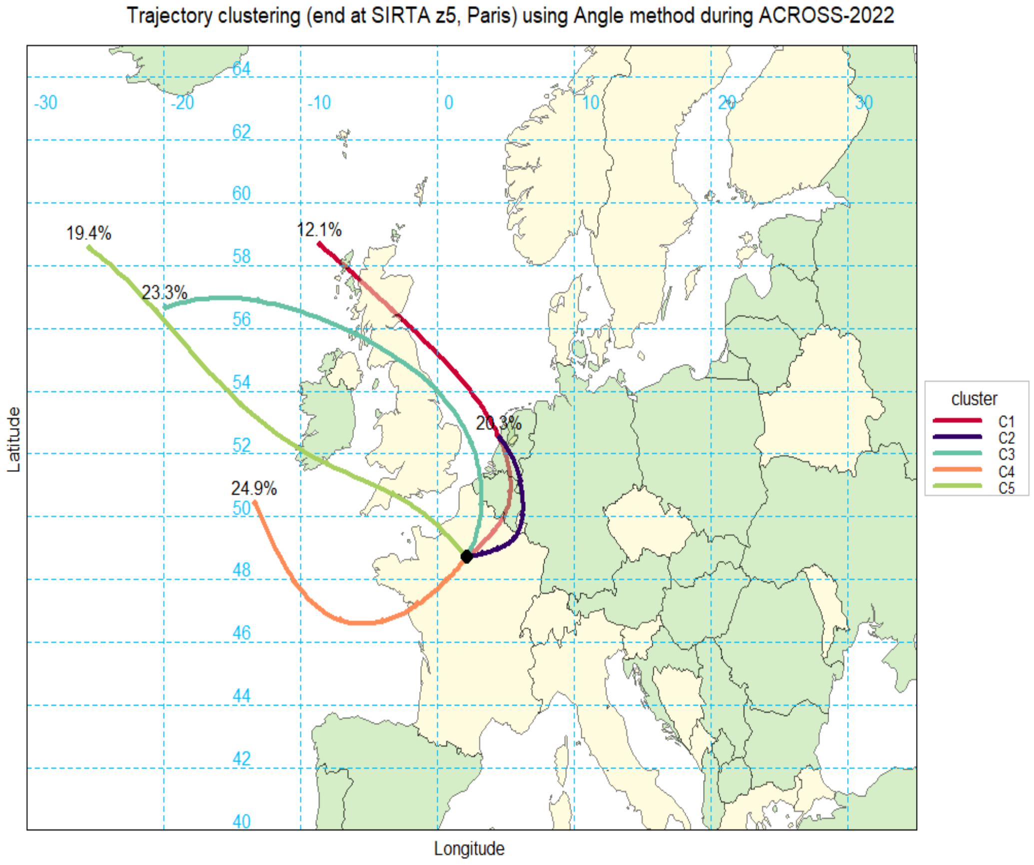
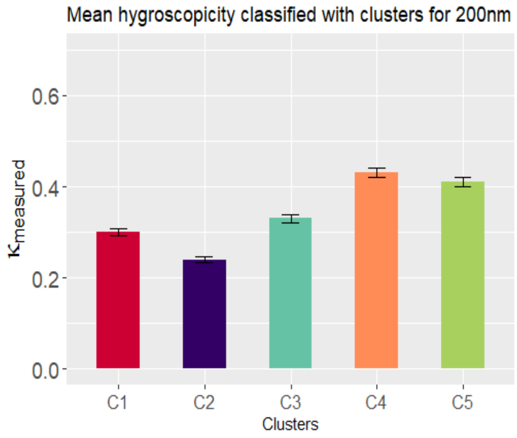
ACROSS Campaign results

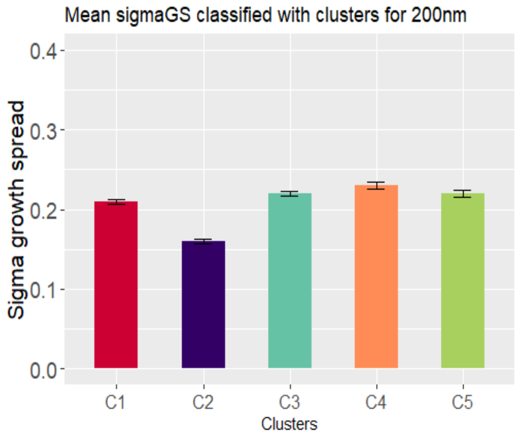


- GF-PDF (Growth factor probability distributions function) shows two distinct modes, hydrophobic (GF≤ 1.2) and hygroscopic (GF > 1.2), and also size dependence.
- ZSR mixing rule: kappa measured and chemically derived shows R=0.49 for 200nm and gets worse for a larger diameter of 250nm.

Trajectory cluster







- 72 h back trajectories were calculated on an hourly basis using the Hysplit with GDAS data at 100m.
- There were five distinct clusters.
- Mostly, the airmass was marine, carrying coarse particles

- High GF is associated with clusters of marine airmass influencing high sigma growth spread.

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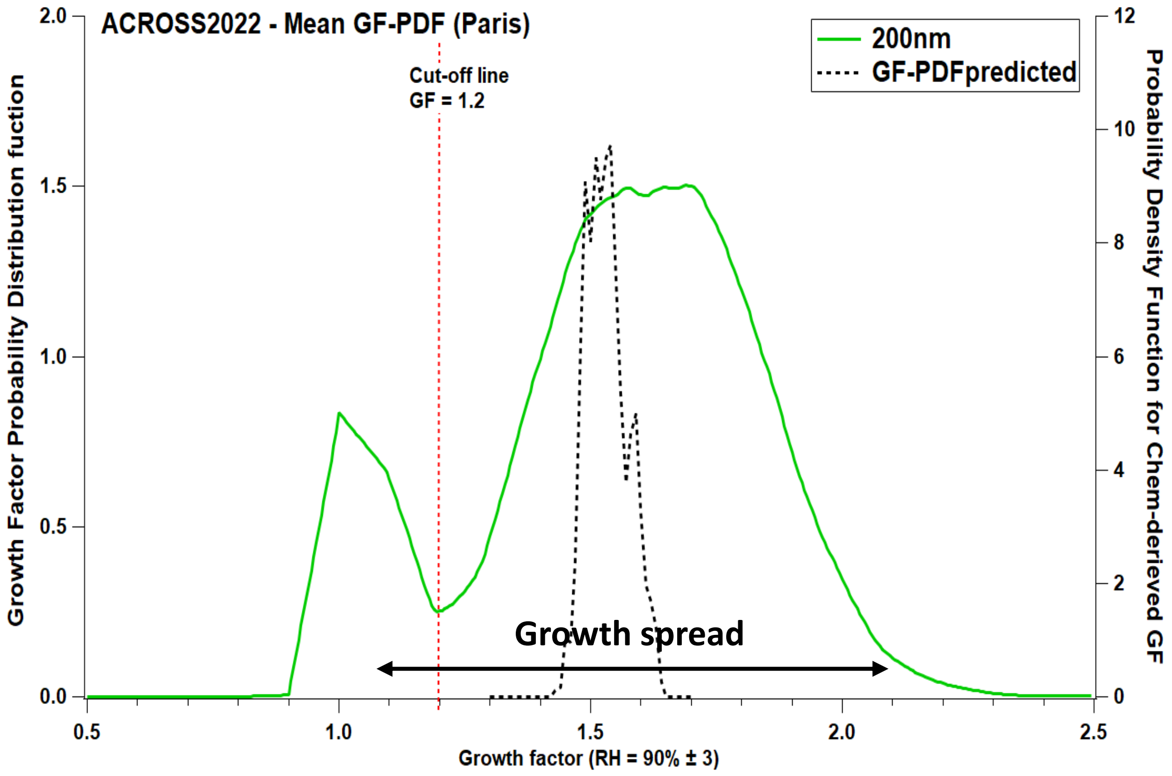
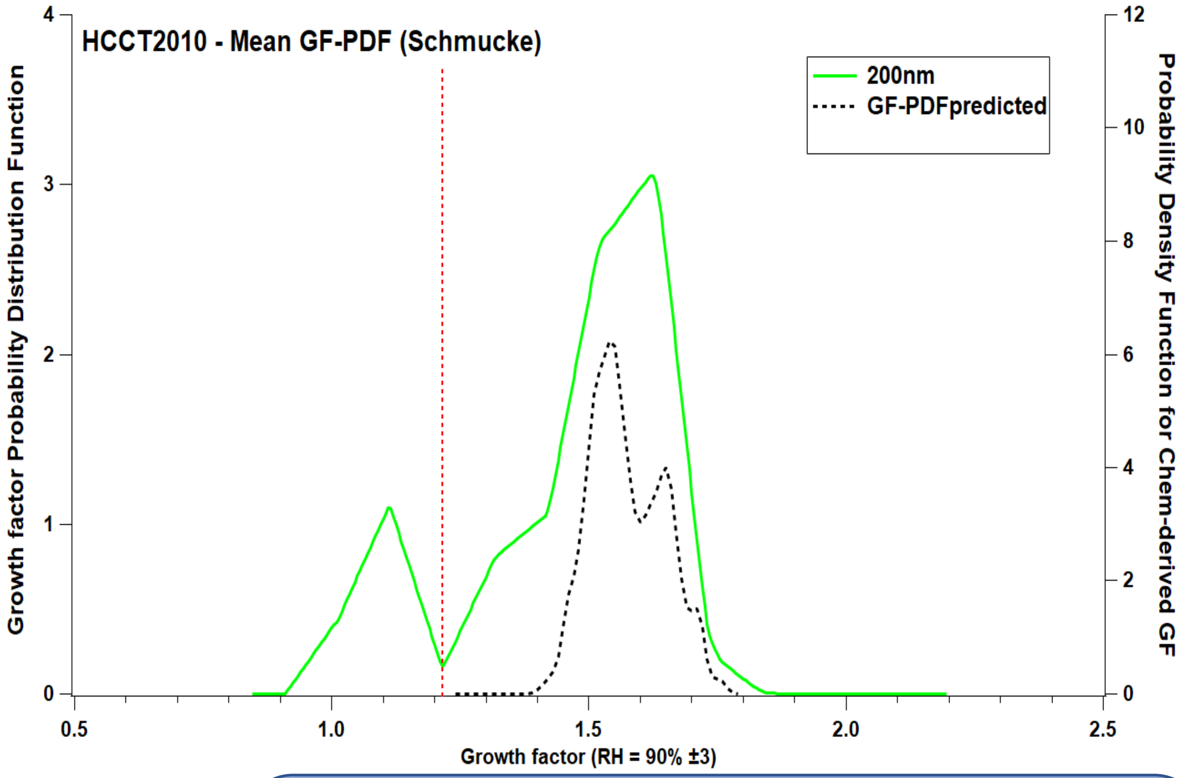
Mixing state influencing prediction of hygroscopicity

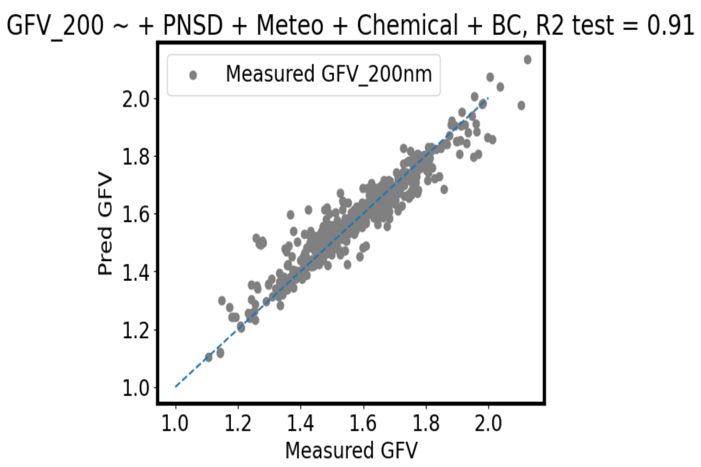


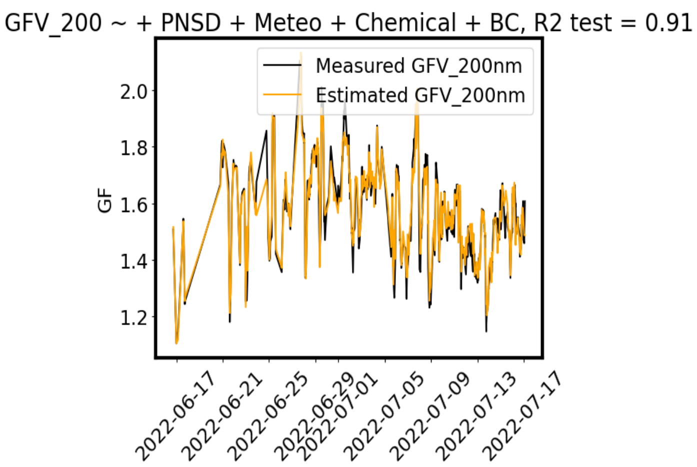


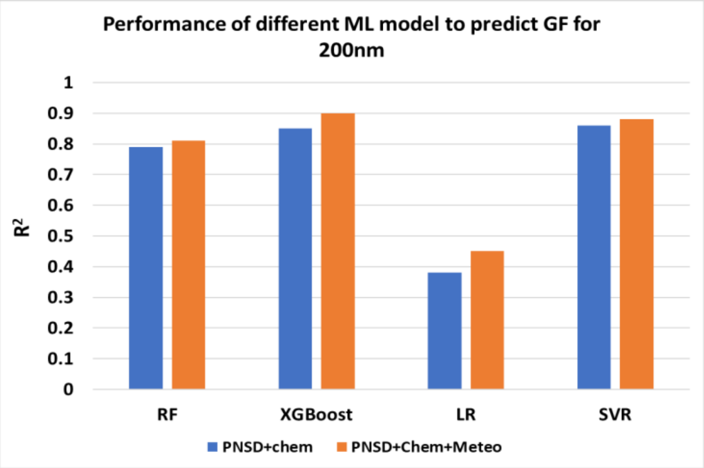
- The closure study during HCCT 2010 was close to a 1:1 line with a correlation ranging from 0.43-0.70.
- Applying the classical ZSR rule gives mean GF values exclusively but does not distinguish growth modes, which doesn't work for external mixed aerosol.

ZSR mixing rule doesn't work for external mixed aerosol

Predicted hygroscopicity through ML







- Different ML models are run on the Paris dataset to estimate aerosol hygroscopicity.
- Among the four ML model approaches, the XGBoost and SVR perform well (also for the India dataset).
- PNSD and chemicals together are predicted very well GF.

ML predicts hygroscopicity very well, but how can R2 be improved?
By improving κ_{org}

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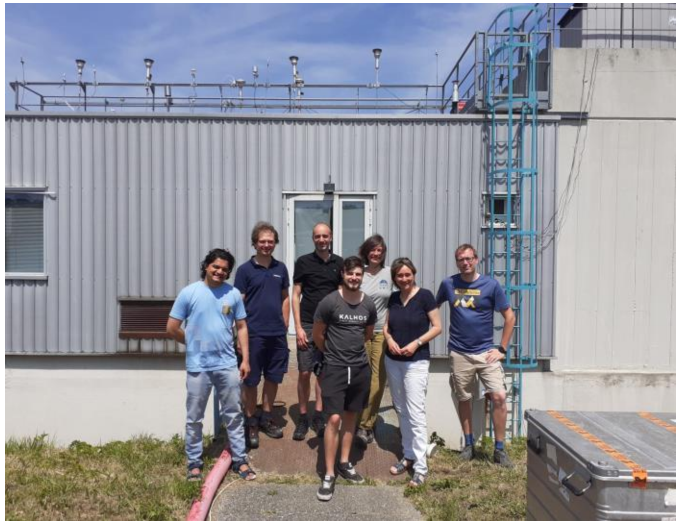
Summary

Why? To reduce the knowledge gap in terms of kappa in the subsaturate regime, urged by the lack of field hygroscopicity measurements.

- The standard deviation (σ) of GF is used to describe the mixing state of aerosols.

- High GF is associated with clusters of marine airmass influencing high σ.

- Particle mixing influencing prediction of hygroscopicity, classical ZSR method does not work for external mixed and fresh urban background aerosol.



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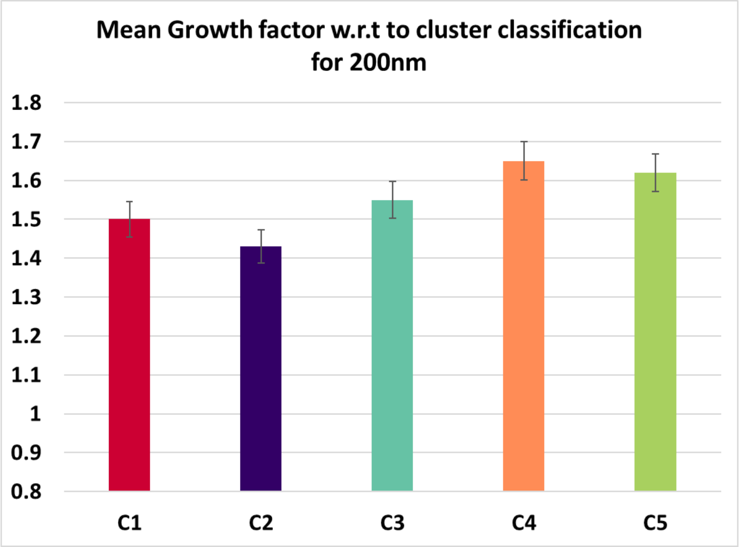
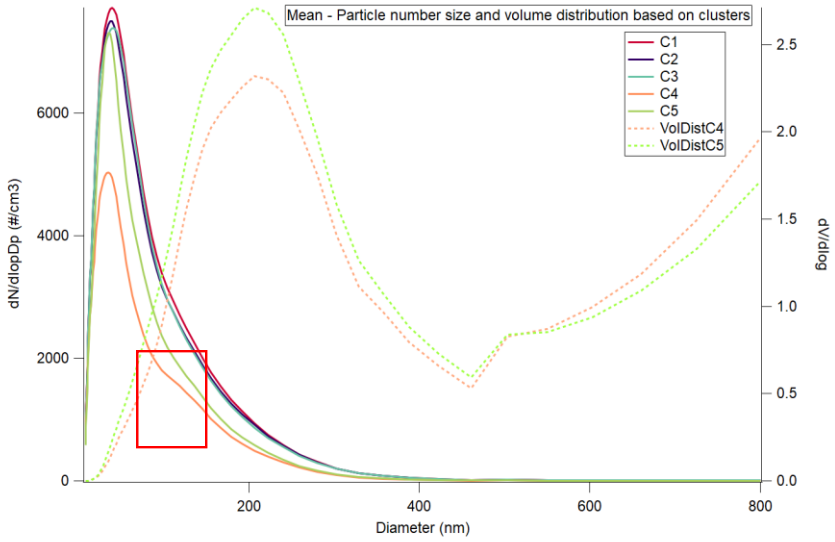
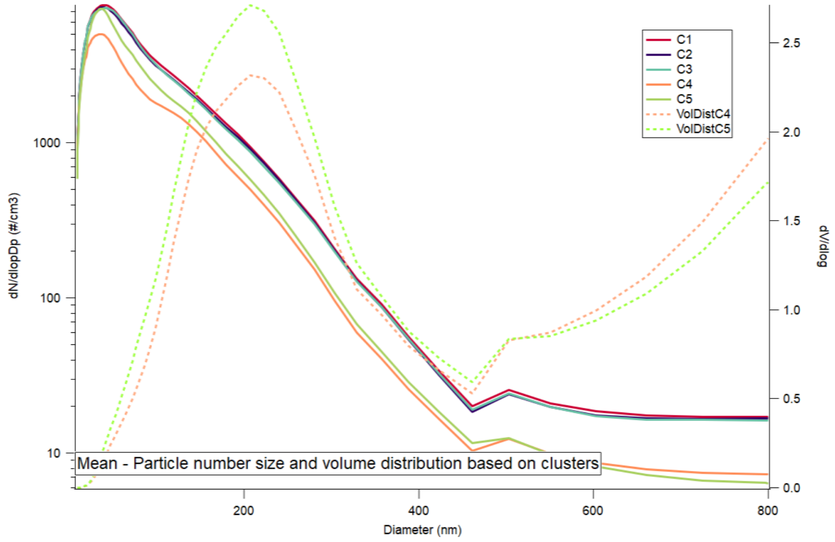
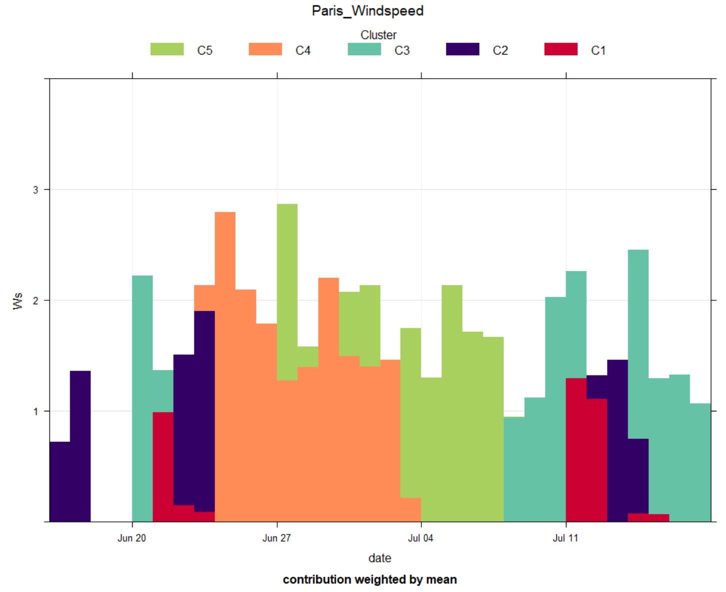
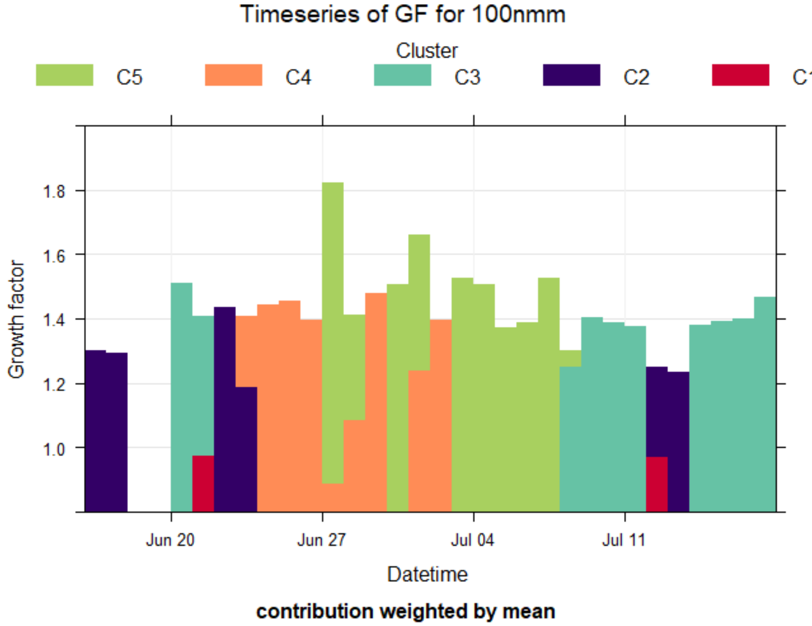
Backup slides

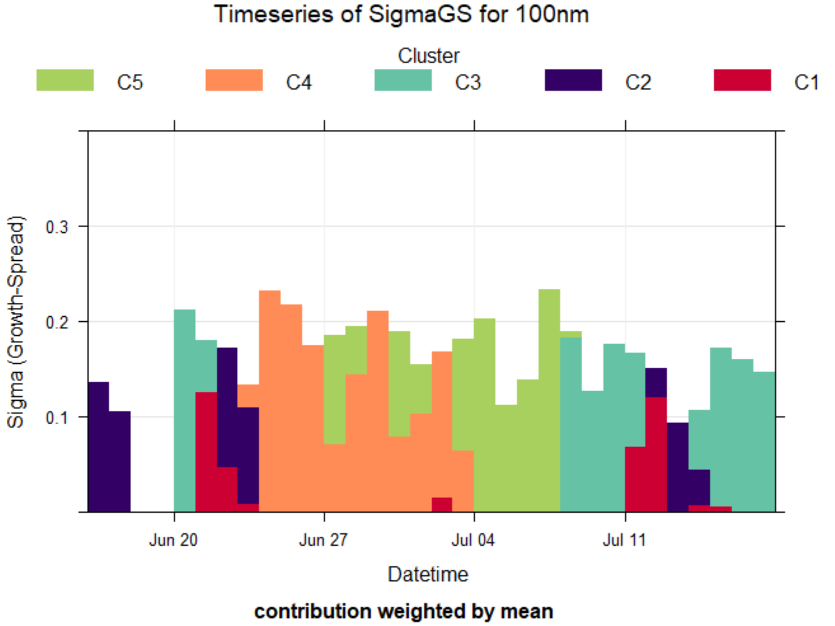












Backup slides