UNIVERSITY OF
EASTERN FINLAND

Modeling uncertainties of aerosol properties and processes

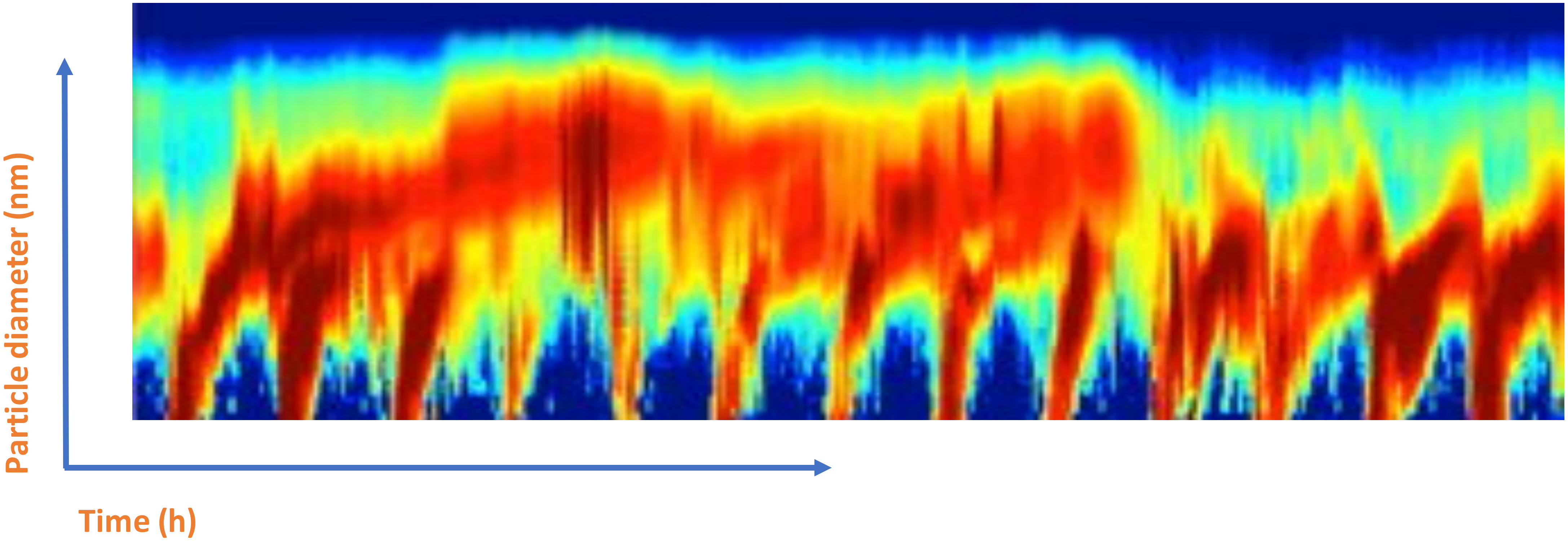
Kari Lehtinen

Team: Prof. Aku Seppänen, Dr. Matti Niskanen, Dr. Matthew Ozon, Väinö Hämäläinen, Teemu Salminen

VILMA CoE Virtual laboratory for molecular level atmospheric transformations

Finnish Centre of Excellence in Inverse Modelling and Imaging

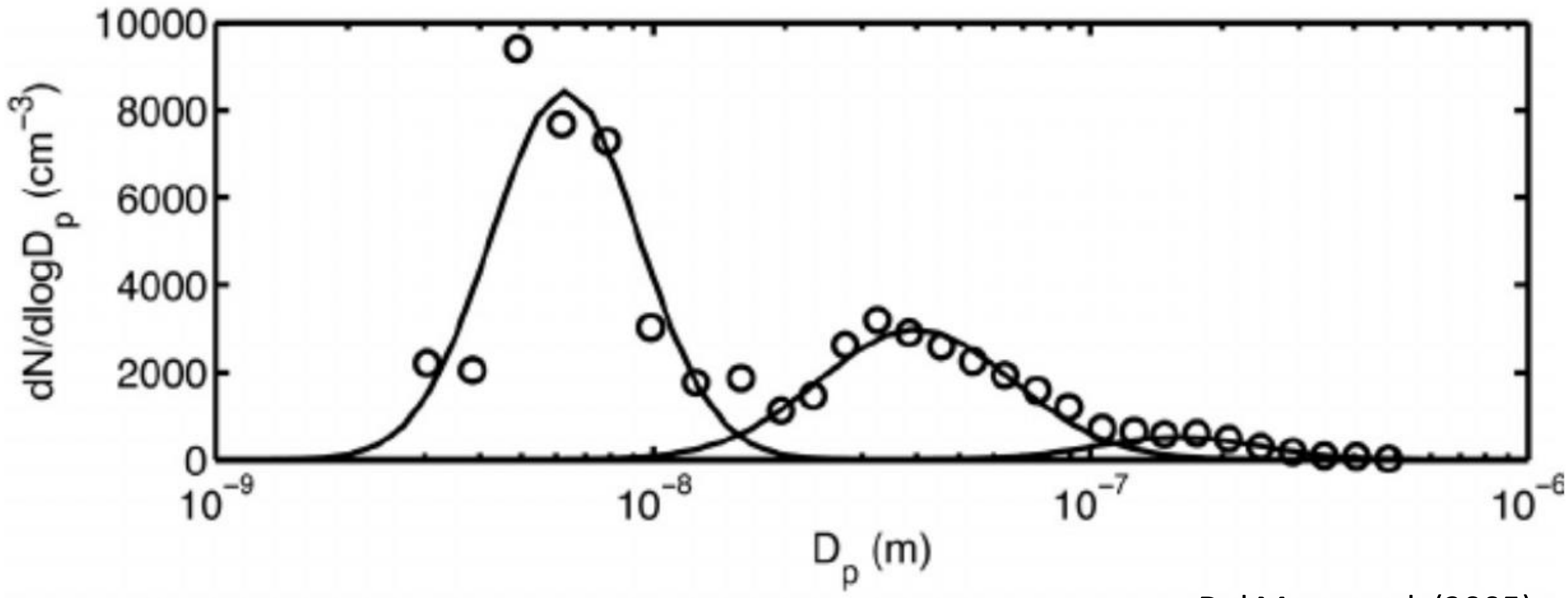
Sequential particle counter measurements



Aims:

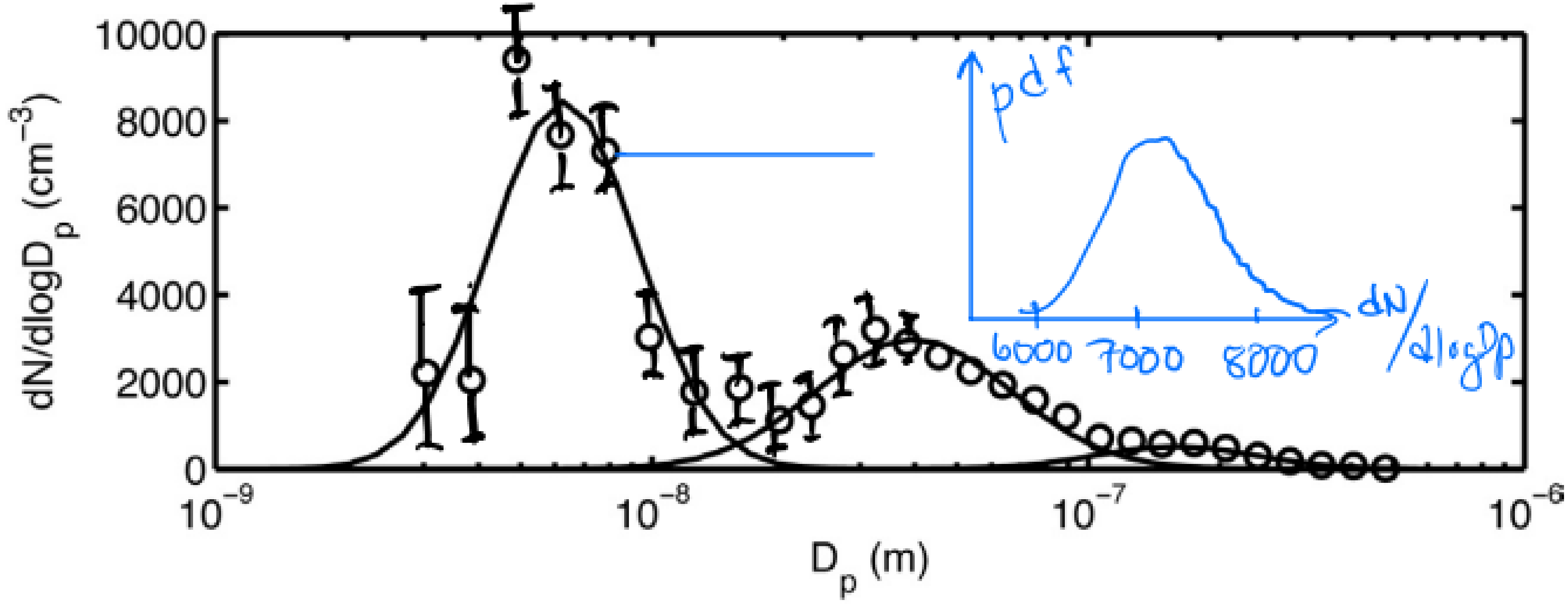
- Reconstruct the particle size distribution, with uncertainties
- Estimate the process rates (nucleation, condensational growth, deposition), with uncertainties

A typical number concentration distribution



Dal Maso et al. (2005)

Number concentration distribution as pdf:s



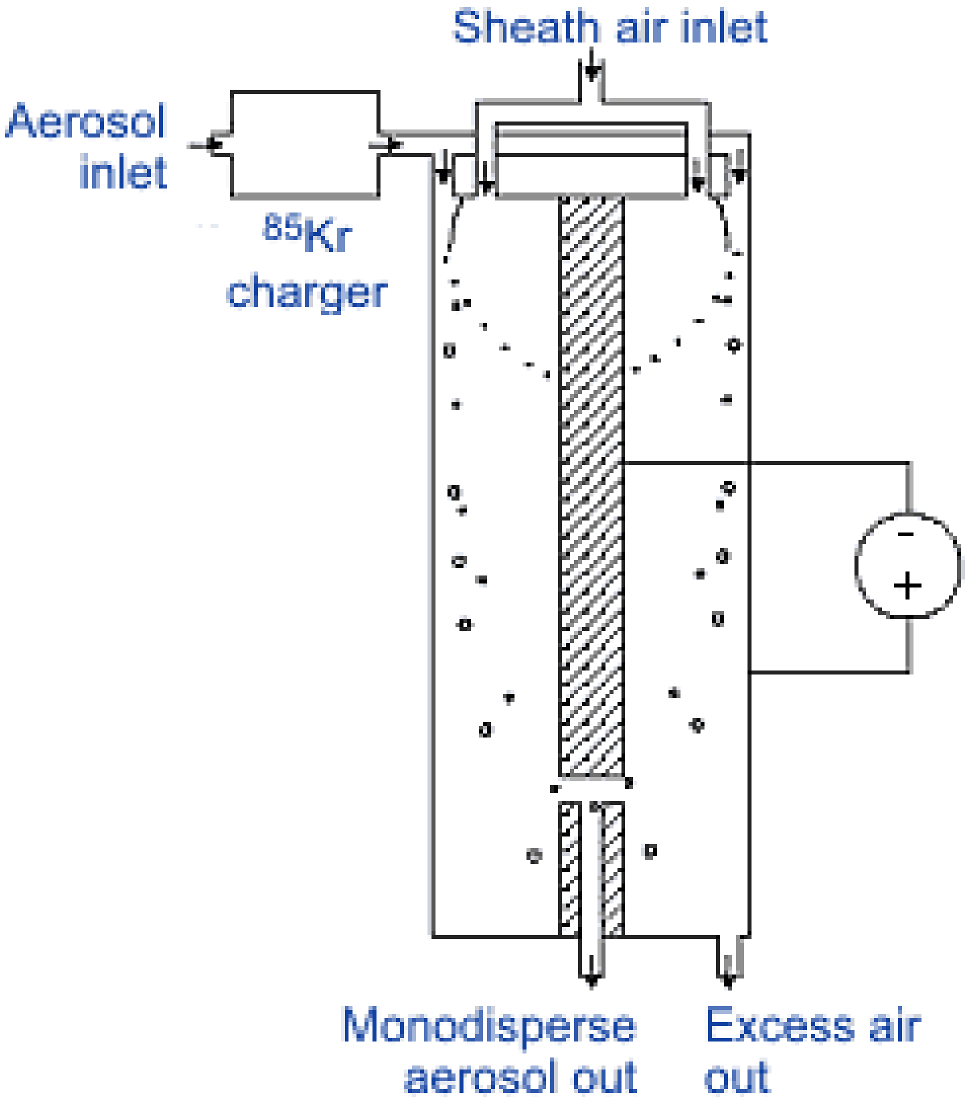
MEASUREMENT UNCERTAINTIES

Many aerosol measurement devices are based on classification by electrical mobility

Challenge:
Charge as a function of particle size needs to be known

Our aim is to find out:
How uncertain are the charging probabilities?

Other sources of uncertainty:
Diffusion losses, low counting statistics



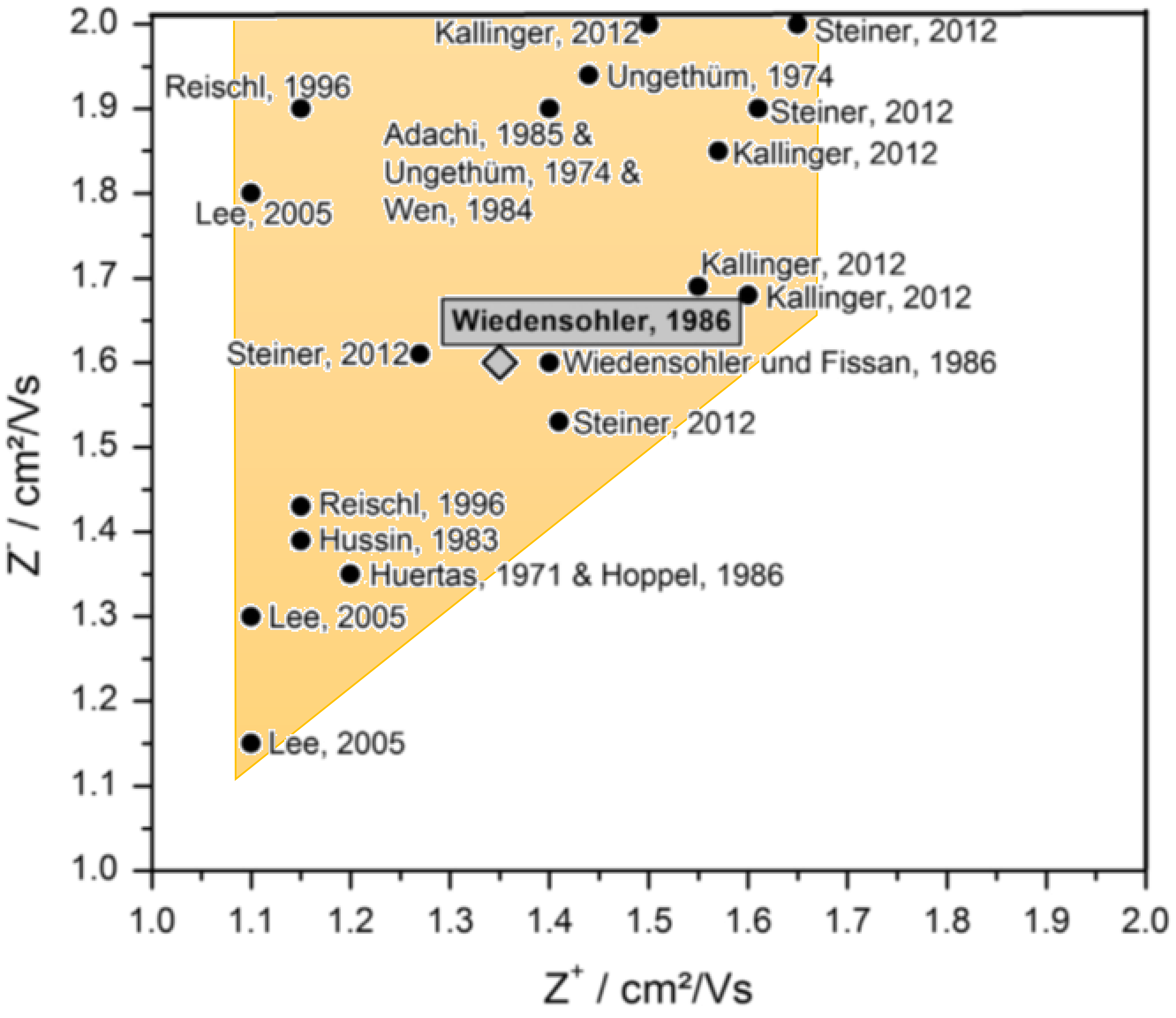
MODEL SETUP: (following Lopez-Yglesias&Flagan, 2013)

- Specify ion properties
- Boltzmann statistics for the steady state ion distributions
- Calculation of flux coefficients
- Solution of population balances

↓

Charging probabilities for different charging states as a function of particle size (Hoppel&Frick, 1986 -> Wiedensohler, 1988)

Variability in charger ion properties



Tigges et al. (2015)

CHARGING PROBABILITY UNCERTAINTY

Wiedensohler (xxxx) curve fits as pale dotted lines.

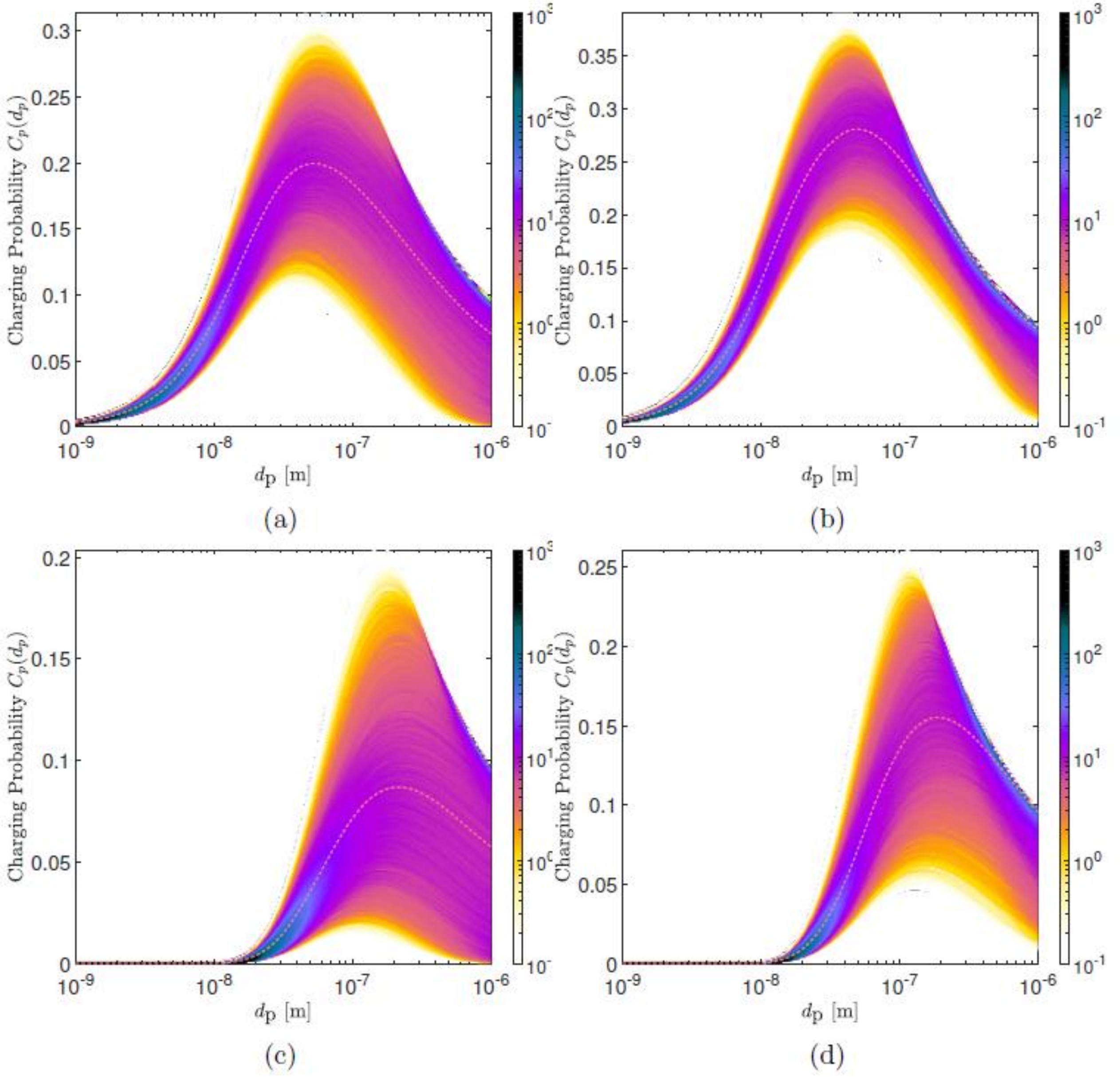
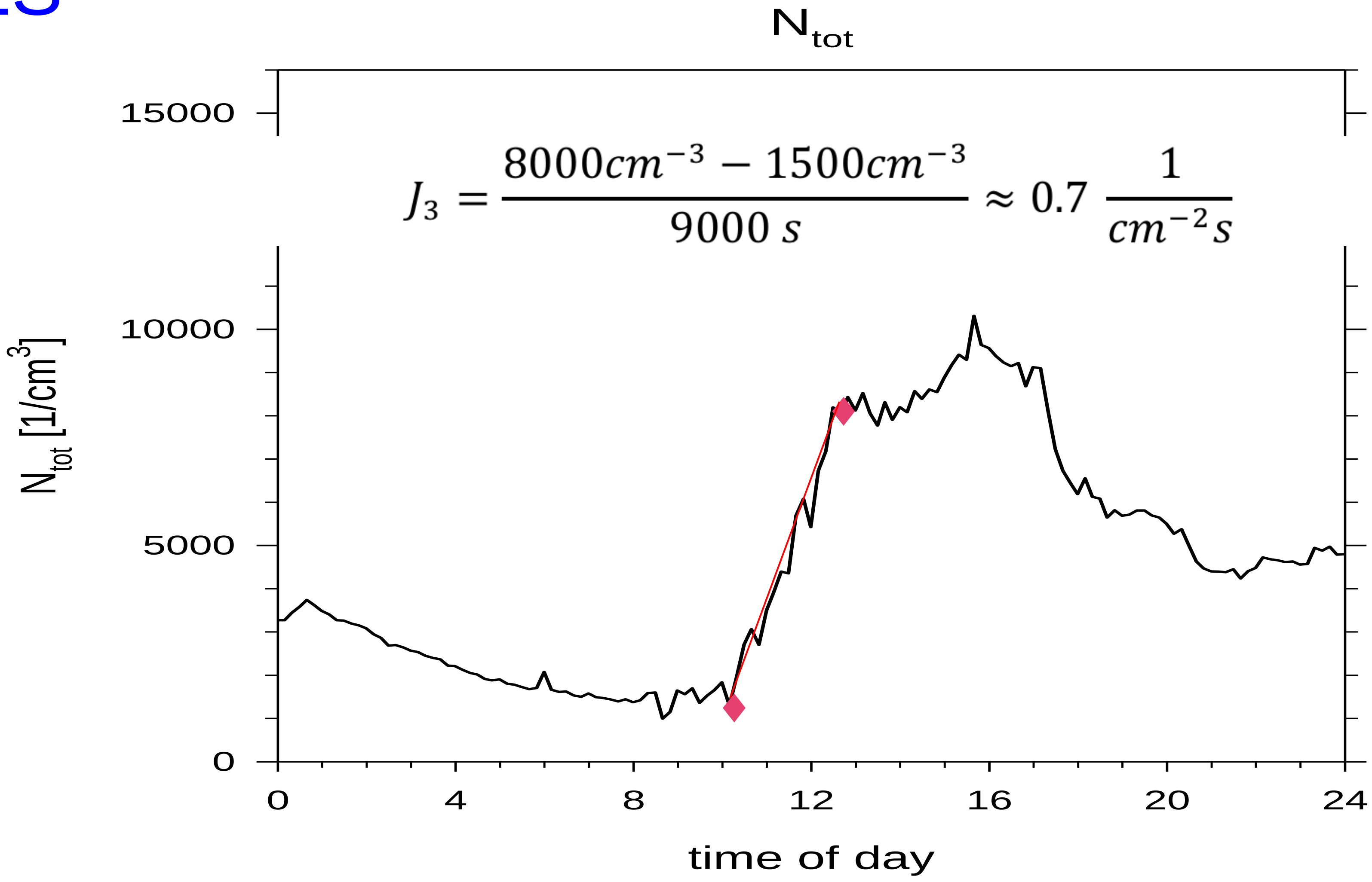


Figure 3: The curve densities corresponding to charge states: a) $p = 1$, b) $p = -1$, c) $p = 2$, d) $p = -2$, e) $p = 3$, and f) $p = -3$. The dashed orange lines show the respective charging probabilities evaluated at the expected values of the parameters.

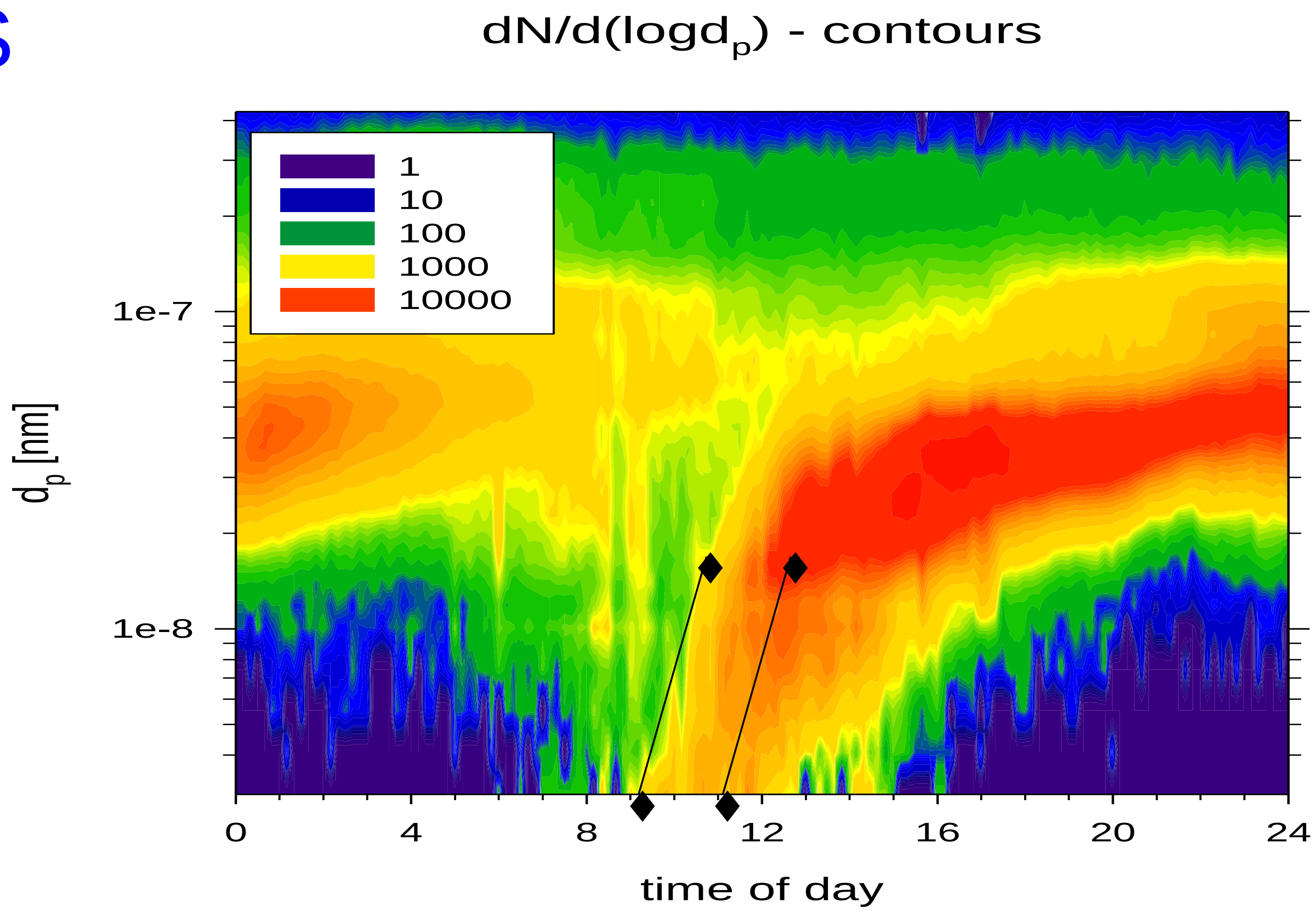
FORMATION RATES

May 19th, 1999



$$J_3 = \frac{8000cm^{-3} - 1500cm^{-3}}{9000\ s} \approx 0.7\ \frac{1}{cm^{-2}s}$$

GROWTH RATES



$$GR \approx \frac{20nm - 3nm}{2h}$$

$$= 8.5\frac{nm}{h}$$

ESTIMATION OF RATES: KALMAN FILTER

- An algorithm that is used to estimate system parameters, including those that cannot be measured directly
- Input: noisy and/or inaccurate and/or missing measurements
- Output: less noisy, more complete and more accurate estimates, with uncertainty estimations
- System parameters are modeled as probability density distributions
- Integrates physical knowledge and intuition into Bayesian inference
- Used widely e.g. in process control and tracking systems

BAYESIAN STATE ESTIMATION

- Sequence of measurements: $y^1,\dots,y^T$
- State variable (model unknowns) $X^k = [N_i^k \quad g_i^k \quad \lambda_i^k \quad J^k]$
- State-space model

$$X^{k+1} = F(X^k) + w^k$$ ← Evolution model (GDE)

$$y^k = HX^k + v^k$$ ← Observation model (DMPS)

- State estimation: Given $y^1,\dots,y^t$ estimate X^k

$k > t$: Prediction
$k = t$: Filtering
$k < t$: smoothing

PHYSICAL KNOWLEDGE AND INTUITION

- Time evolution of the size distribution is described by the aerosol GDE

$$\frac{\partial n}{\partial t}(d_{\mathrm{p}},t) = -\underbrace{\frac{\partial g(d_{\mathrm{p}},t)n(d_{\mathrm{p}},t)}{\partial d_{\mathrm{p}}}}_{\text{growth by condensation}} \underbrace{- n(d_{\mathrm{p}},t)\int_{d_0}^{\infty} \beta(d_{\mathrm{p}},s)n(s,t)\mathrm{d}s}_{\text{coagulation sink}}$$

$$\underbrace{+\frac{1}{2}\int_{0}^{d_{\mathrm{p}}} \beta\left(\sqrt[3]{d_{\mathrm{p}}^3 - q^3}, q\right) n\left(\sqrt[3]{d_{\mathrm{p}}^3 - q^3}, t\right) n(q,t)\mathrm{d}q}_{\text{coagulation source}} \underbrace{-\lambda(d_{\mathrm{p}},t)n(d_{\mathrm{p}},t)}_{\text{loss by deposition}}$$

- Prior ‘guesses’ for the variables can be taken to make sense, and can include correlations both in time and size

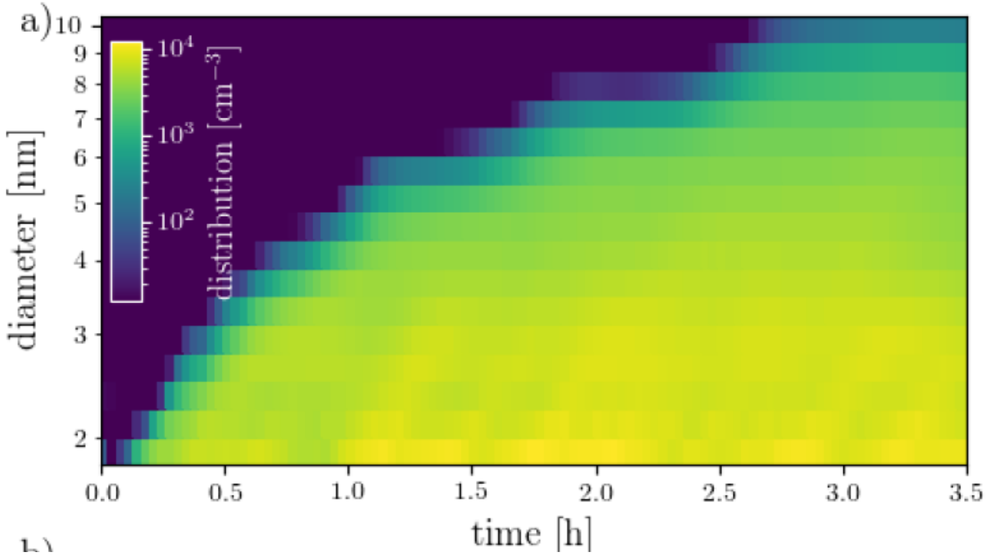
General dynamic equation (GDE) of aerosols

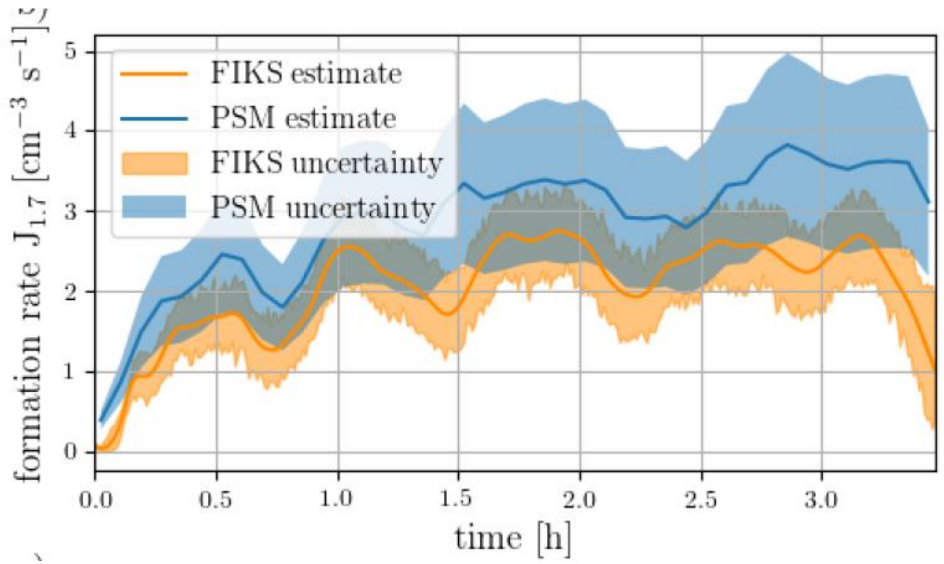
Unknowns in the model
$n(d_p,t)$*, *$g(d_p,t)$ and ***$\lambda(d_p,t)$***
and $J(t)$

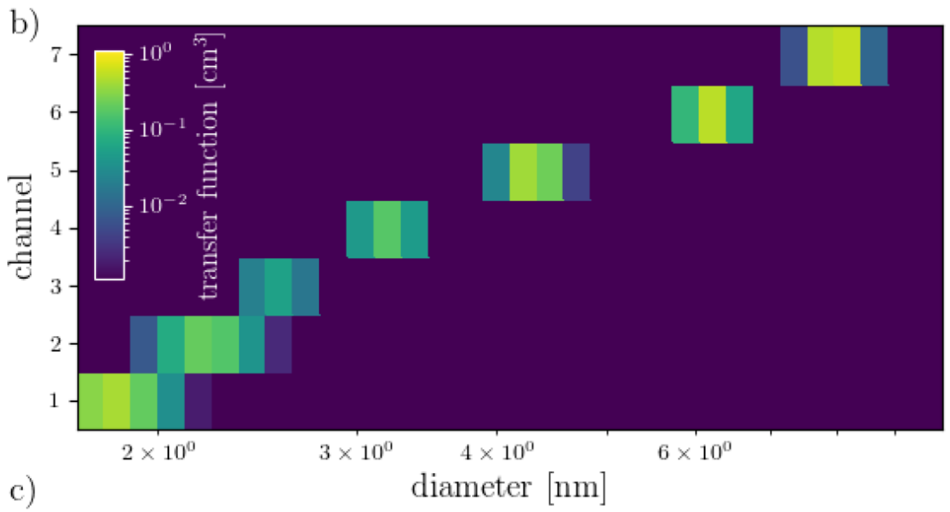
$$\frac{\partial n}{\partial t}(d_\mathrm{p},t) = -\underbrace{\frac{\partial g(d_\mathrm{p},t)n(d_\mathrm{p},t)}{\partial d_\mathrm{p}}}_{\text{growth by condensation}} \underbrace{-\, n(d_\mathrm{p},t)\int_{d_0}^{\infty}\beta(d_\mathrm{p},s)n(s,t)\mathrm{d}s}_{\text{coagulation sink}}$$

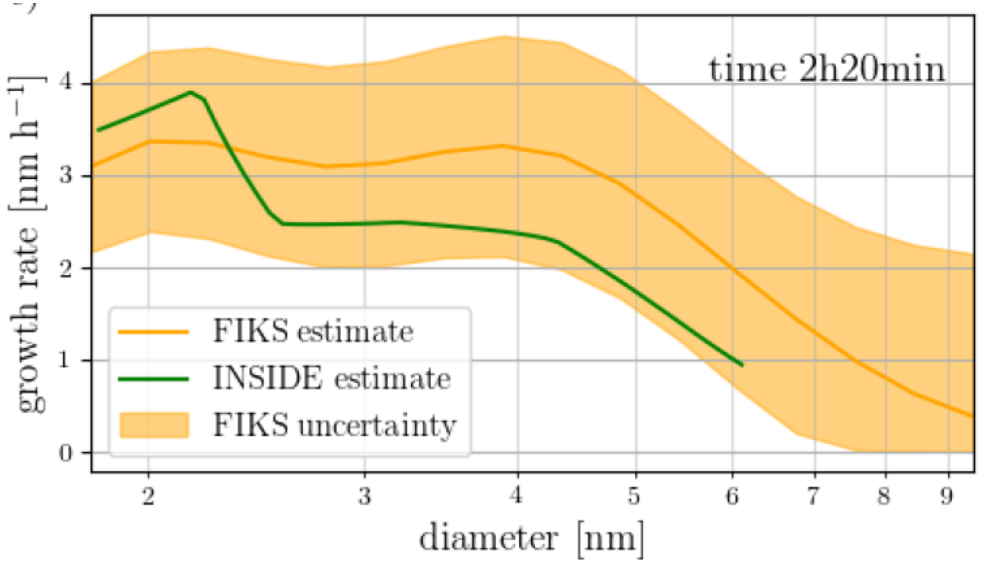
$$\underbrace{+\frac{1}{2}\int_{0}^{d_\mathrm{p}}\beta\left(\sqrt[3]{d_\mathrm{p}^3-q^3},q\right)n\left(\sqrt[3]{d_\mathrm{p}^3-q^3},t\right)n(q,t)\mathrm{d}q}_{\text{coagulation source}} \underbrace{-\,\lambda(d_\mathrm{p},t)n(d_\mathrm{p},t)}_{\text{loss by deposition}}$$

DMA-train measurements of sulphuric acid-ammonia nucleation and growth in CLOUD



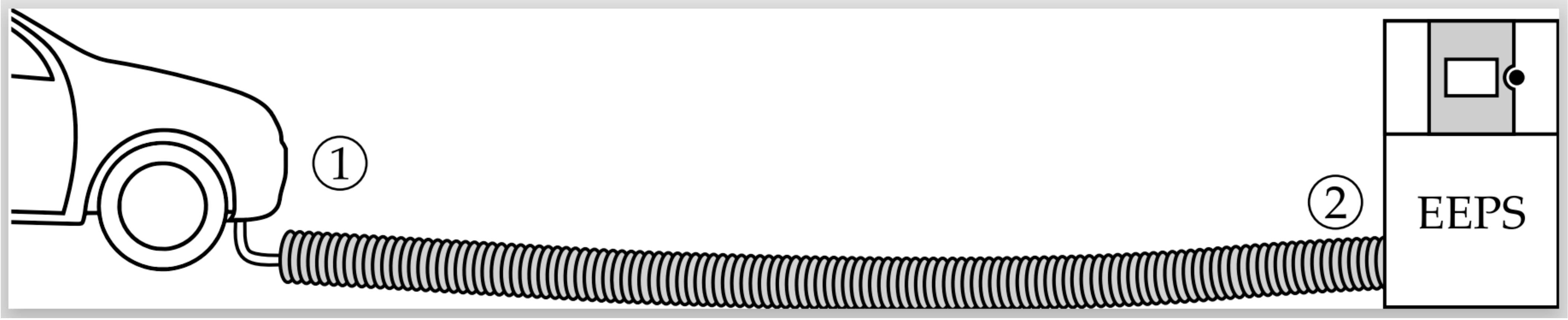






Thanks to Lubna Dada and Dominik Stolzenburg who made the comparisons to CLOUD data possible

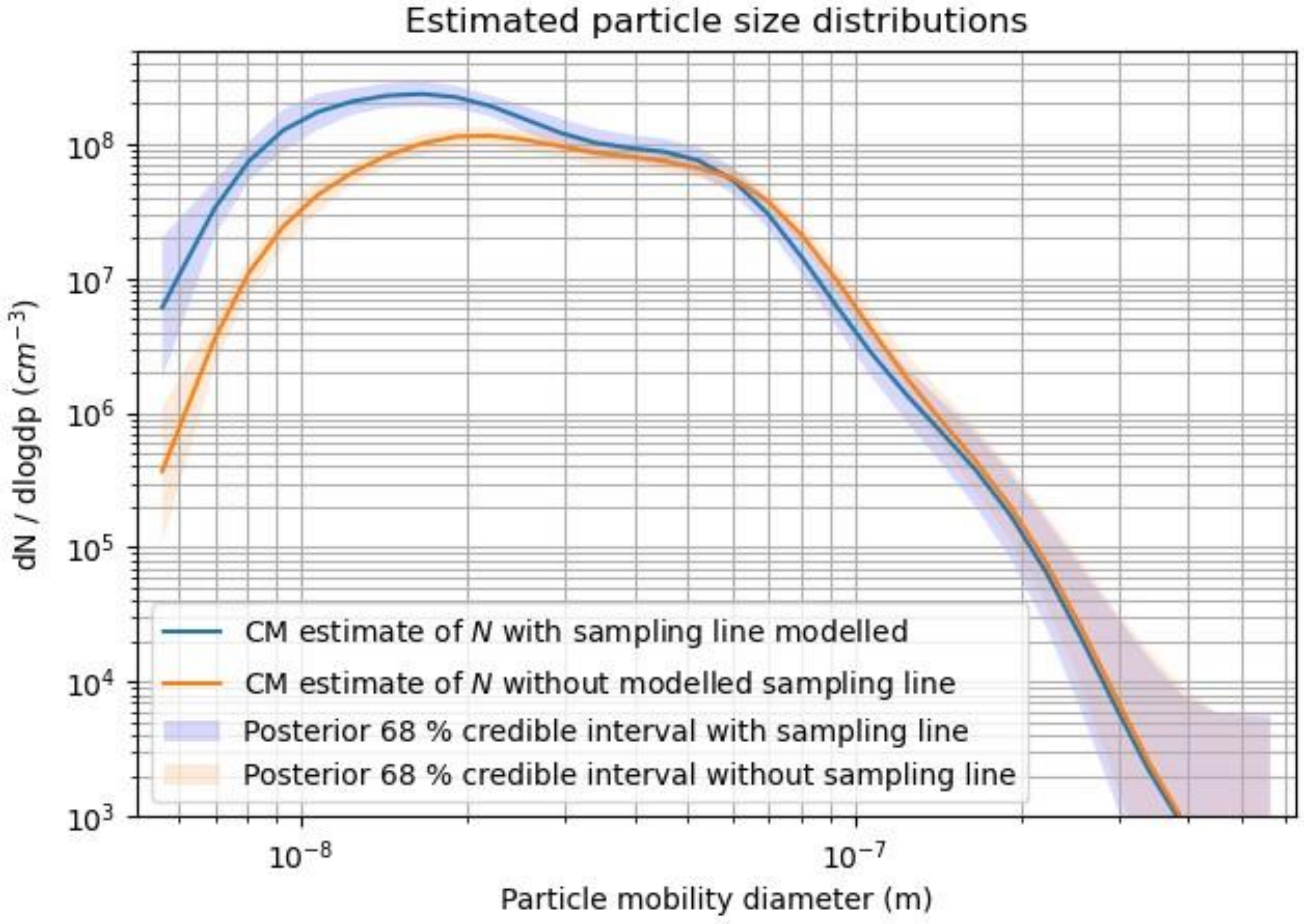
Example: Inverse modeling of coagulation and deposition with aerosol GDE & MCMC



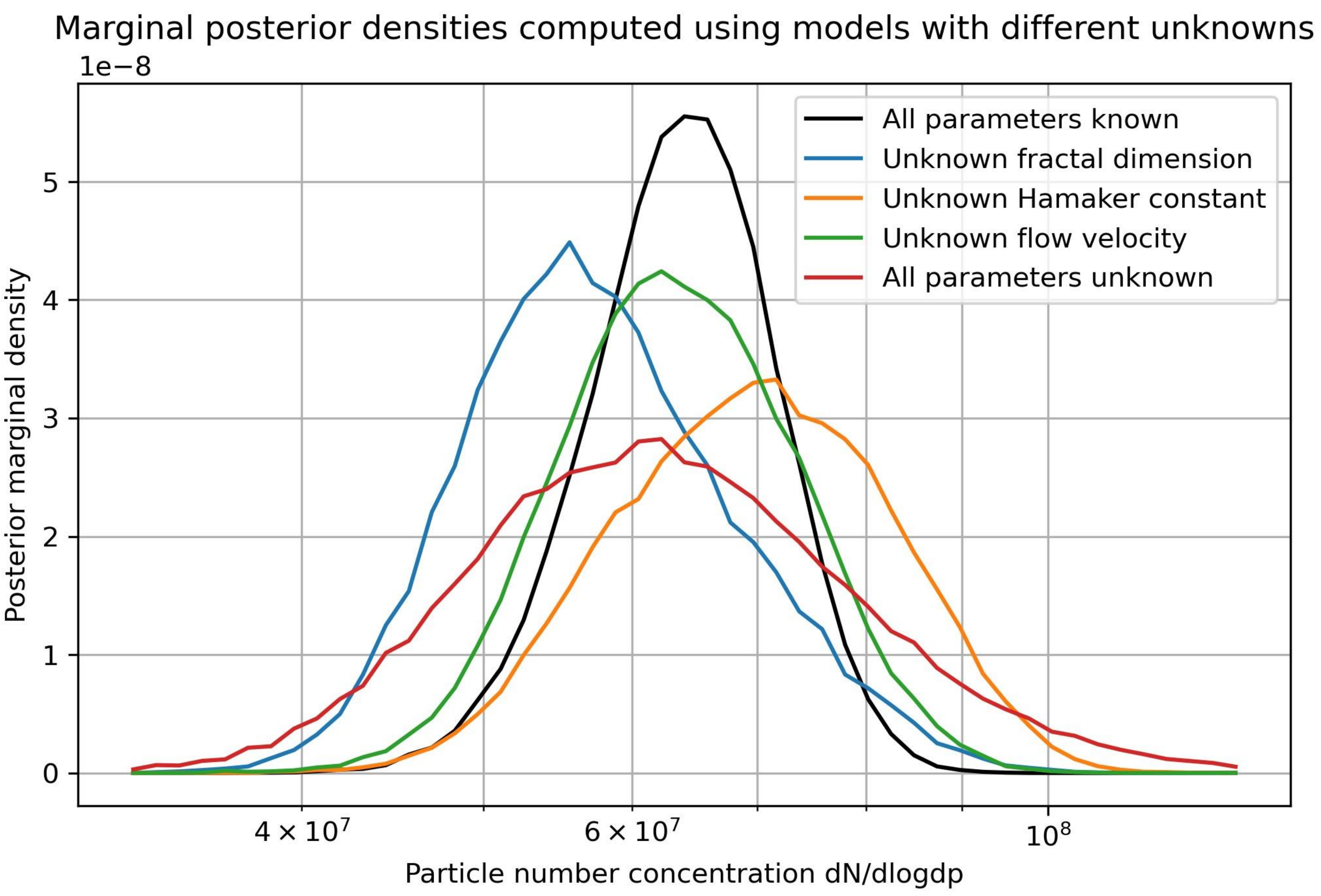
Engine
Exhaust
Particle
Sizer

Variables with uncertainty:

Fractal shape parameters (fractal dimension, primary particle size)
Van Der Waals force (Hamaker constant)
Flow velocity



Estimated particle size distributions
dN / dlogdp (cm^{-3})
10^8
10^7
10^6
10^5
10^4
10^3
10^{-8}
10^{-7}
Particle mobility diameter (m)
CM estimate of N with sampling line modelled
CM estimate of N without modelled sampling line
Posterior 68 % credible interval with sampling line
Posterior 68 % credible interval without sampling line



Marginal posterior densities computed using models with different unknowns
1e−8
All parameters known
Unknown fractal dimension
Unknown Hamaker constant
Unknown flow velocity
All parameters unknown
Posterior marginal density
Particle number concentration dN/dlogdp
0
1
2
3
4
5
4 × 10^7
6 × 10^7
10^8

Geoscientific Model Development
Geosci. Model Dev., 14, 3715–3739, 2021
https://doi.org/10.5194/gmd-14-3715-2021

Retrieval of process rate parameters in the general dynamic equation for aerosols using Bayesian state estimation: BAYROSOL1.0

Matthew Ozon[1], Aku Seppänen[1], Jari P. Kaipio[1,3], and Kari E. J. Lehtinen[1,2]
[1]Department of Applied Physics, University of Eastern Finland, Kuopio, Finland
[2]Finnish Meteorological Institute, Kuopio, Finland
[3]Department of Mathematics, Faculty of Science, University of Auckland, Auckland, New Zealand

UNIVERSITY OF EASTERN FINLAND

Thank you!

Atmos. Chem. Phys., 21, 12595–12611, 2021
https://doi.org/10.5194/acp-21-12595-2021

Atmospheric Chemistry and Physics
EGU

Aerosol formation and growth rates from chamber experiments using Kalman smoothing

Matthew Ozon[1], Dominik Stolzenburg[2], Lubna Dada[2,3,4], Aku Seppänen[1], and Kari E. J. Lehtinen[1,5]
[1]Department of Applied Physics, University of Eastern Finland, 70210 Kuopio, Finland
[2]Institute for Atmospheric and Earth System Research/ Physics, University of Helsinki, 00014 Helsinki, Finland
[3]EPFL, School of Architecture, Civil and Environmental Engineering, 1951 Sion, Switzerland
[4]Paul Scherrer Institute, Laboratory of Atmospheric Chemistry, 5232 Villigen PSI, Switzerland
[5]Atmospheric Research Centre of Eastern Finland, Finnish Meteorological Institute, 70210 Kuopio, Finland

ELSEVIER
Available online at www.sciencedirect.com
ScienceDirect
Journal of Aerosol Science 00 (2023) 1–19
Journal of Aerosol Science

Uncertainty Analysis of the Aerosol Charge Distribution in a Bipolar Environment

Väinö Hämäläinen[1,1,*], Aku Seppänen[1,1], Kari Lehtinen[1,1,1]