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INSTITUTE FOR ATMOSPHERIC AND EARTH SYSTEM RESEARCH

UNIVERSITY OF HELSINKI

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Quantifying the contributions of NPF and traffic emissions on urban UFP concentrations

P. Paasonen (with thanks to many)

UFP symposium, Brussels, 3.-4.7.2024

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Why…

- Motivation:
  - Adverse (not well quantified) UFP health effects & cooling climate impacts
  - Decreasing PM mass enhances new particle formation (NPF) and increases UFP lifetime
  - Shares of traffic and NPF in urban (and global) UFP unclear
- Uncertainties in how to reduce UFP health burden and how it affects climate

Why and how?

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- Direct observations of the source contributions not available and the methods to estimate them require “educated guesses”:
  - New/improved data analysis methods needed
- To improve modelling of particle numbers, description of nucleation mode (<30nm) particle emissions and their near-source processes require more attention

From concentrations to source rates and from observations to models

- Shares of NPF and traffic sources to observed <u>concentrations</u>
    - NPF and traffic tracer approach
    - The next step: better long-term estimates and momentary probabilities

- Determining <u>source rates</u> for NPF and emission
    - New “NPF” classification routine for more information, including emissions
    - Towards continuous J and E timeseries

- Improving PN emissions in <u>models</u> and testing NPF mechanisms
    - Emission models and sub-grid scale process parameterisations
    - Testing emission and NPF mechanisms with Lagrangian models

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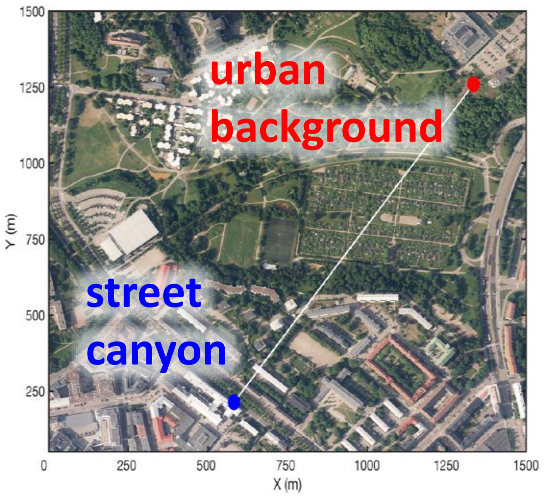
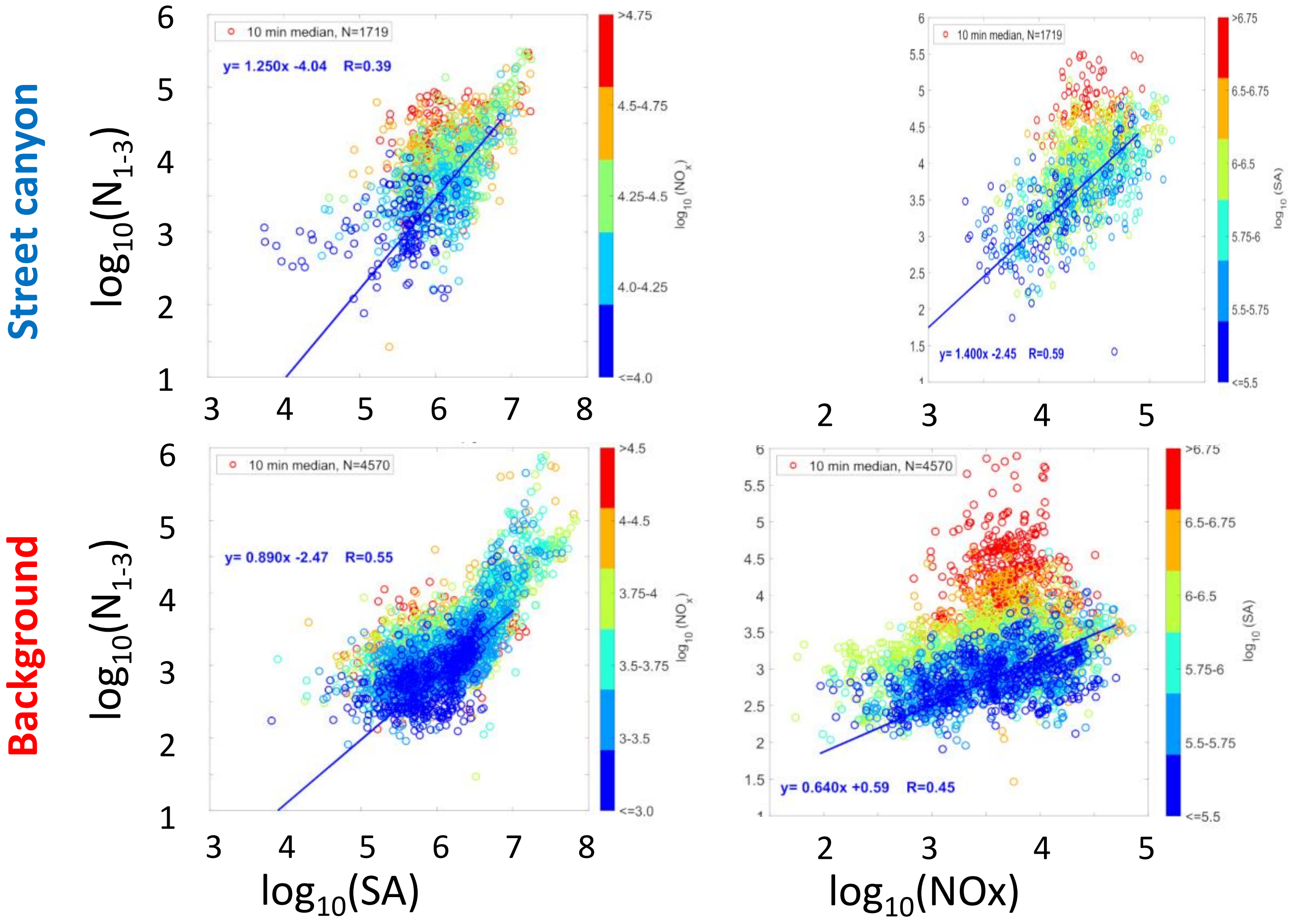
Helsinki – roughly similar contributions of traffic and NPF on sub-3 nm particle concentration



- One month data (May, 2018)
- NOx as tracer for traffic emissions, sulphuric acid (SA) for NPF
- Street canyon N_{1-3} dominated by traffic at nighttime
- Background N_{1-3} dominated by NPF at daytime
- Traffic daytime and background night roughly 50-50



Okuljar et al., ACP, 2021

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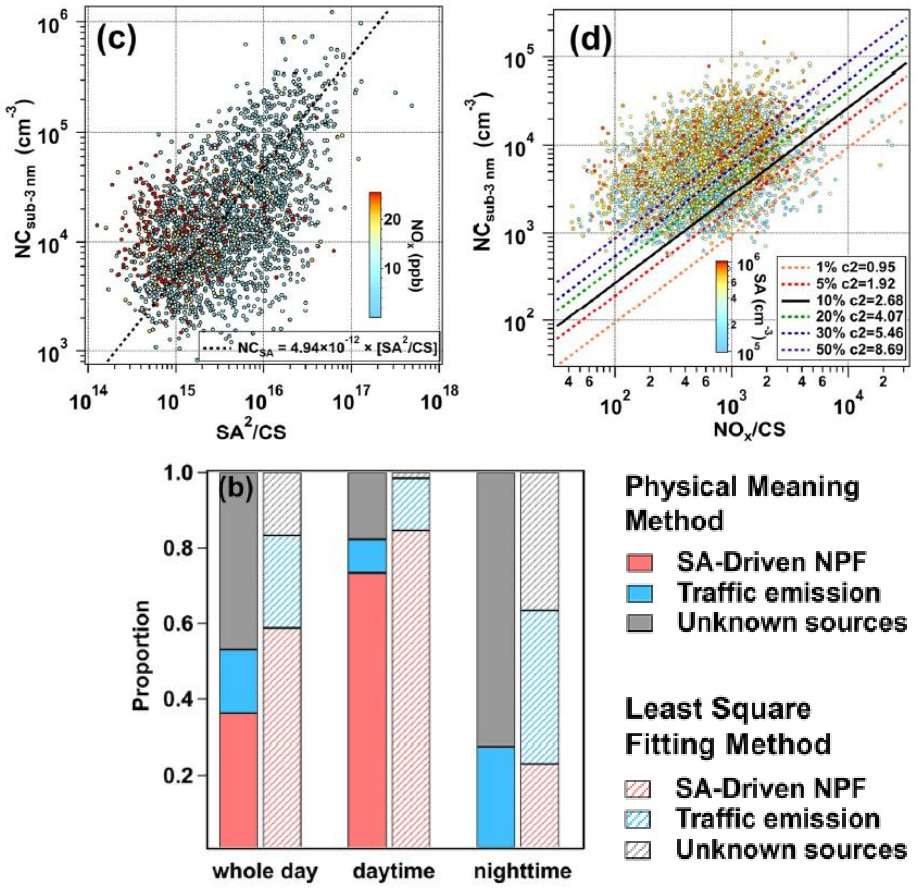
Nanjing urban background – daytime $N_{1\text{-}3}$ dominated by NPF, nighttime traffic and ”undefined sources”

- In methods, some modifications to Okuljar et al. (2021): some improvements, some drawbacks and some issues remaining
- My notes-to-self from Okuljar and Chen papers:
  - Linear least squares is a good fitting method for these purposes
  - Defining contribution of “x-axis related source” with n^th^ percentile requires educated guesses (& being careful with log-scales)



Chen et al., JGR: Atmos., 2023

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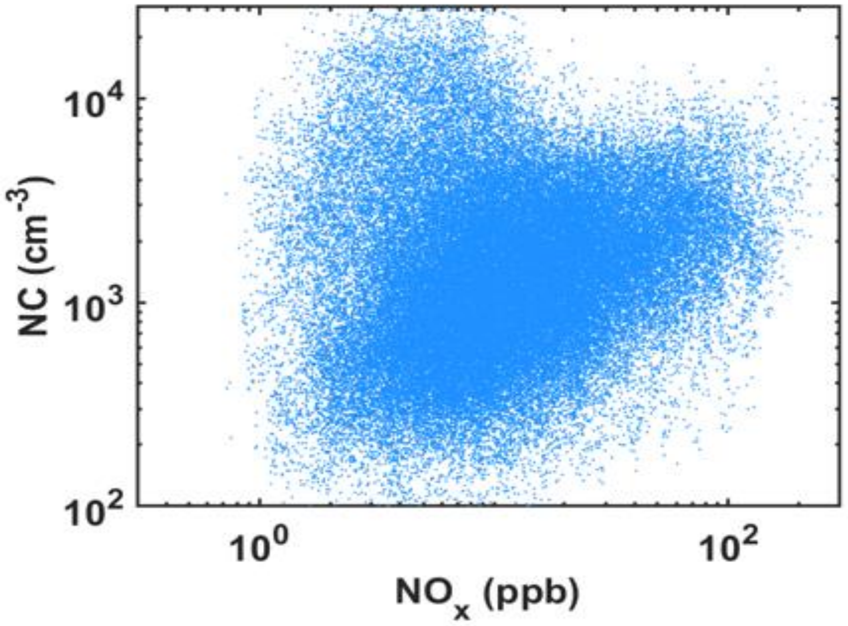
The next step: use source-specific probability modes for PN to determine source shares

- Hypothesis: UFP concentrations from certain source represent a (log-normal) probability distribution mode. For traffic: μ (and possibly σ) = f(NOx)

Nanjing $N_{6\text{-}25}$



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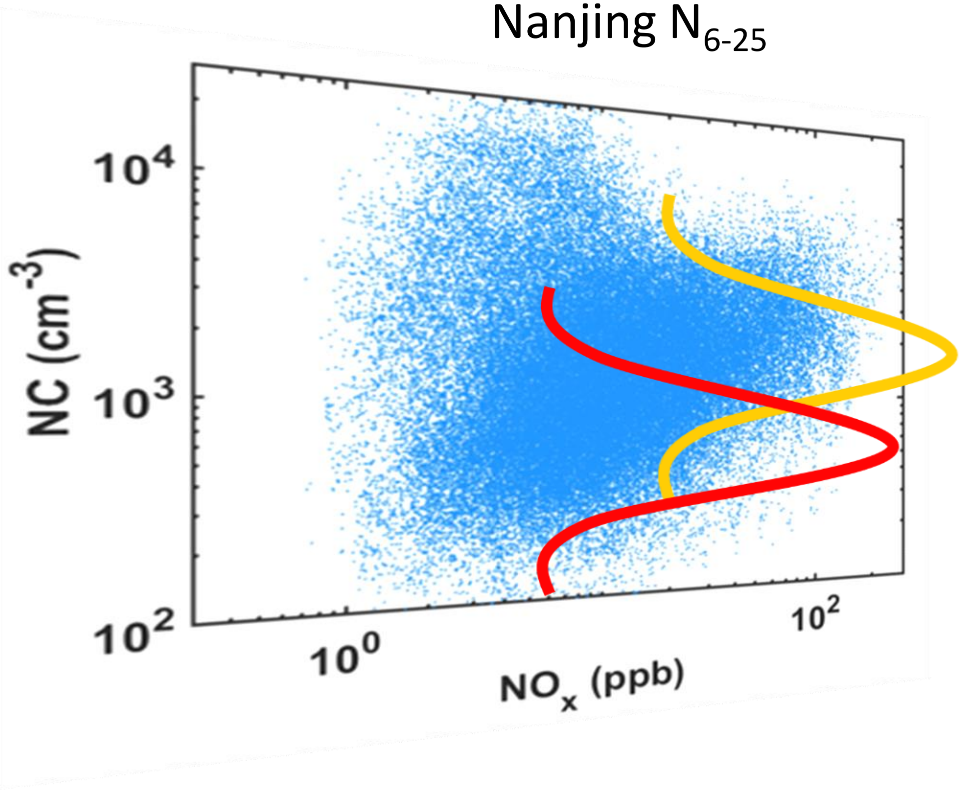

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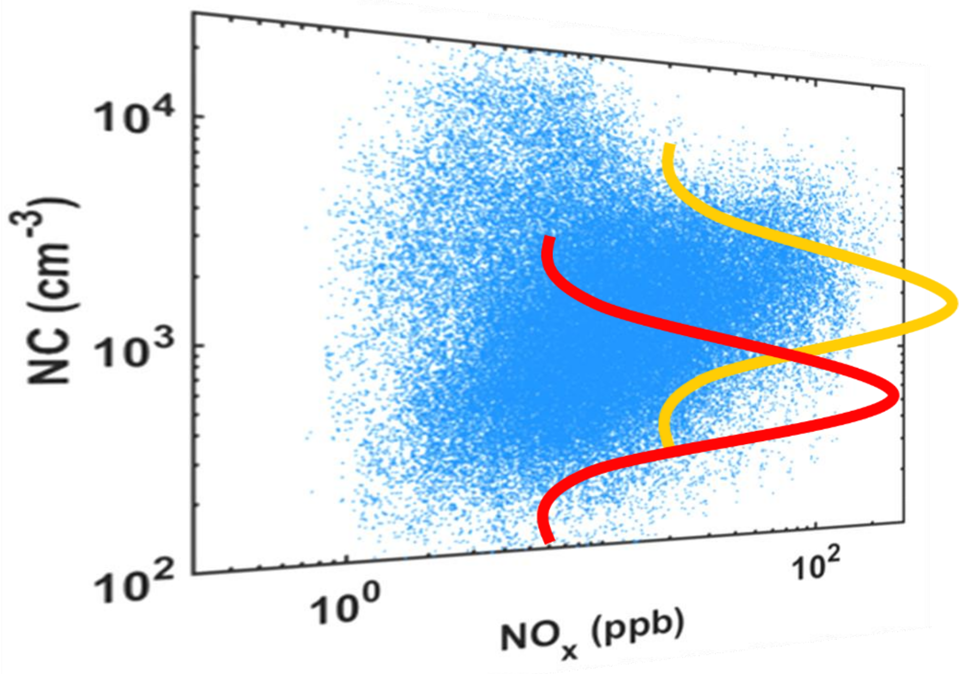
The next step: use source-specific probability modes for PN to determine source shares

- Hypothesis: UFP concentrations from certain source represent a (log-normal) probability distribution mode. For traffic: μ (and possibly σ) = f(NOx)
- The difference between the observation and this mode equals to UFP from other sources
- A routine to separate traffic mode and the rest, calculate overall (e.g., seasonal) shares and estimate probabilities of shares on momentary concentrations

Nanjing $N_{6\text{-}25}$



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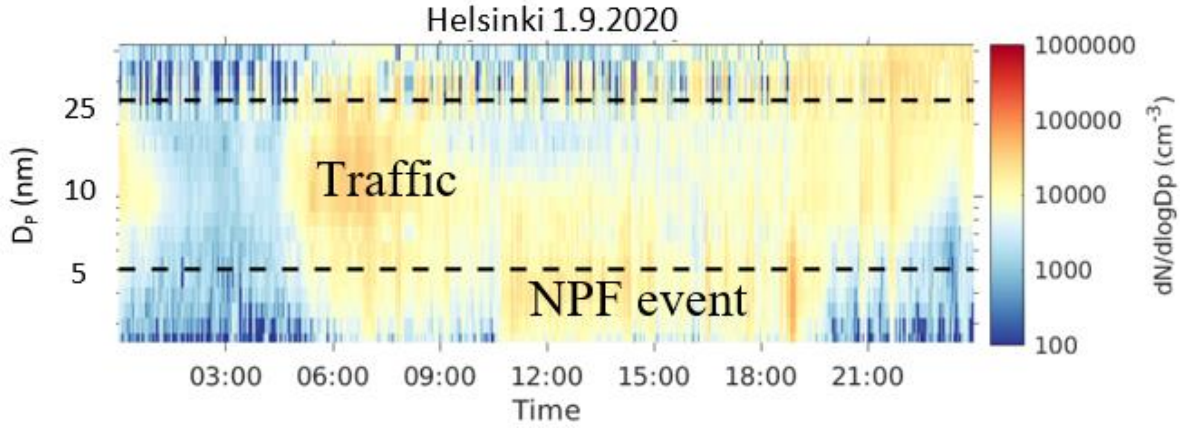
Rethinking ”NPF classification”: spatio-temporal information and recognition of nucleation mode emissions

Traditional classification (dal Maso et al., Boreal Env. Res., 2005) divides days to NPF event, non-NPF event and undefined days: NPF event if a new mode appears @ <25 nm and diameter grows.

New approach:

- Spatial information based on the observed mode:
    - Reaches <5 nm: NPF (or emission) event *in situ*
    - Mode diameter grows: wide (upwind) area
- Temporal information: start and end times of NPF (or emission) event
- Likelihood of emission or NPF event based on appearing mode width:
    - Reaching from <5 to >25 nm, likely emissions
    - Further analysis with additional observations (NOx, rush hours, radiation…)



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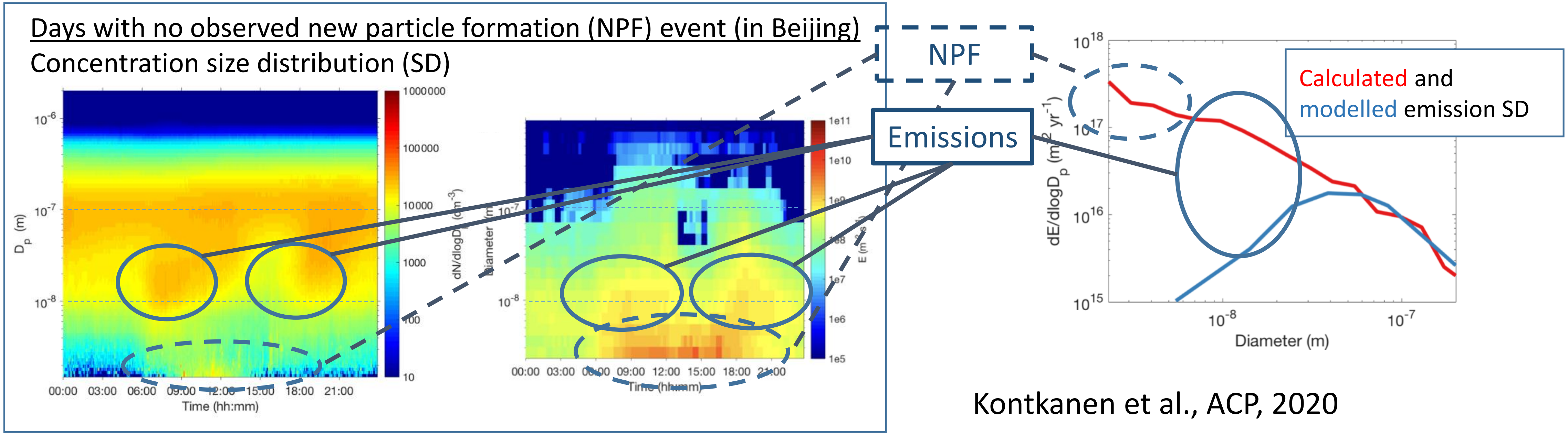
Continuous time-series for NPF and PN emission rates near

- Solving NPF rate (*J*) requires:
  - Coagulation sink (directly from particle size distribution measurements)
  - Particle growth rate (GR)
  - (and the assumption of spatial and temporal homogeneity of past upwind aerosol processes…)
- Determining GR is challenging, but several advances taking place
  - GR observed to vary less than concentrations of condensable vapours, also on “non-NPF event” days (Kulmala et al., 2024, Aerosol Res.)
  - Automatic derivation of growth rates from observation data generated (Paasonen et al., 2018, ACP, and *in preparation*)

Beijing – annual level of emitted >30nm particles from observations ≈ from GAINS model; challenges in < 30 nm, time series yet to test


Days with no observed new particle formation (NPF) event (in Beijing)

Concentration size distribution (SD)

NPF

Emissions

Calculated and modelled emission SD

Kontkanen et al., ACP, 2020

- Modelled PN emissions (Paasonen et al., 2016) do not capture the traffic mode, but improvements hopefully soon (co-operation between INAR, Tampere University, IIASA, SYKE, TNO(?)…)
- Calculating timeseries for (upwind) *J* is “simple”, for (upwind) emissions more complicated

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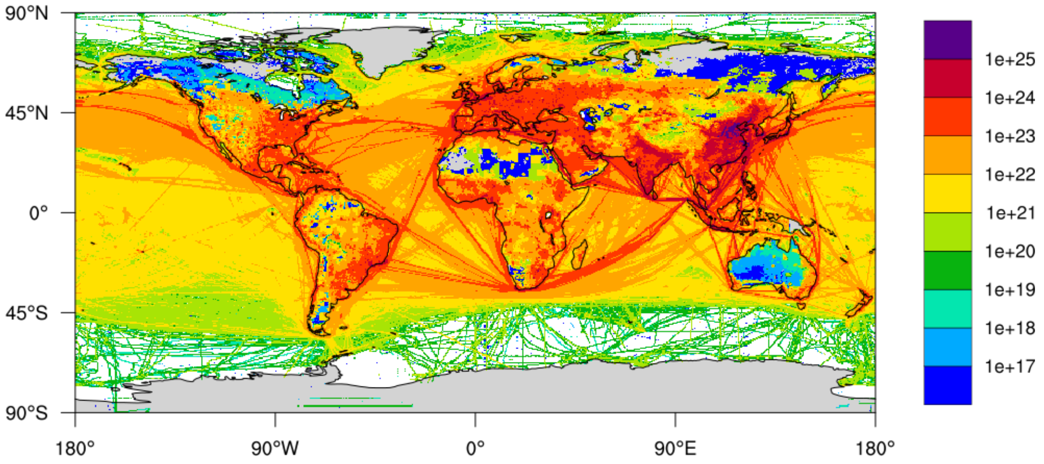
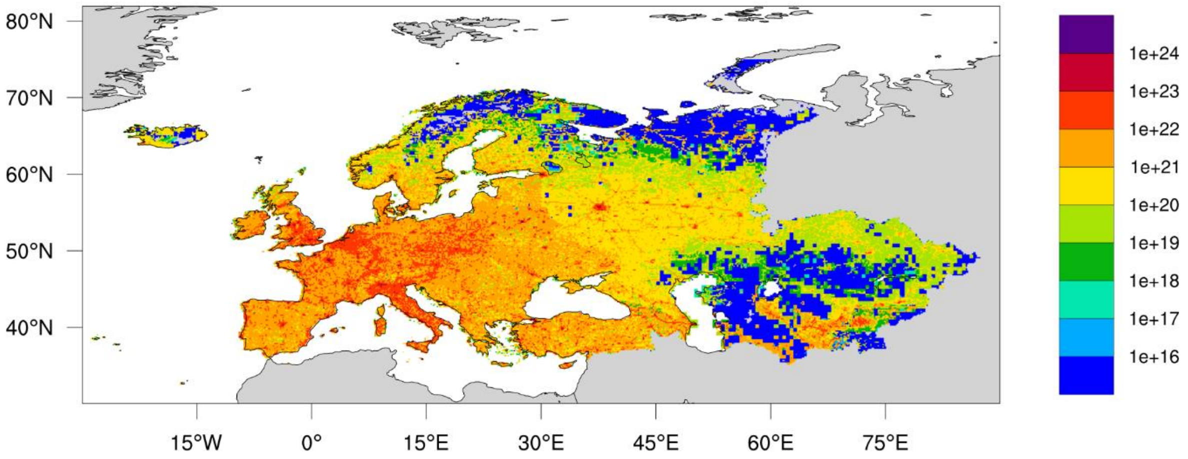
Size-segregated global anthropogenic particle number emissions

- Produced with GAINS emission scenario model, ECLIPSE_v6a CLE scenario
- Spatial resolution: global 0.5°×0.5°, Europe 0.1°×0.1°, Finland with FRES model by SYKE 0.25km×0.25 km
- Size resolution: range 3-1000 nm with flexible number of bins
- Improvements for sub-30 nm emissions to be implemented



Anthropogenic particle number emissions (#/grid cell)



Figures by Dian Ding

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Sub-grid scale (SGS) process parameterisations with theory and multi-scale simulations

Model configurations

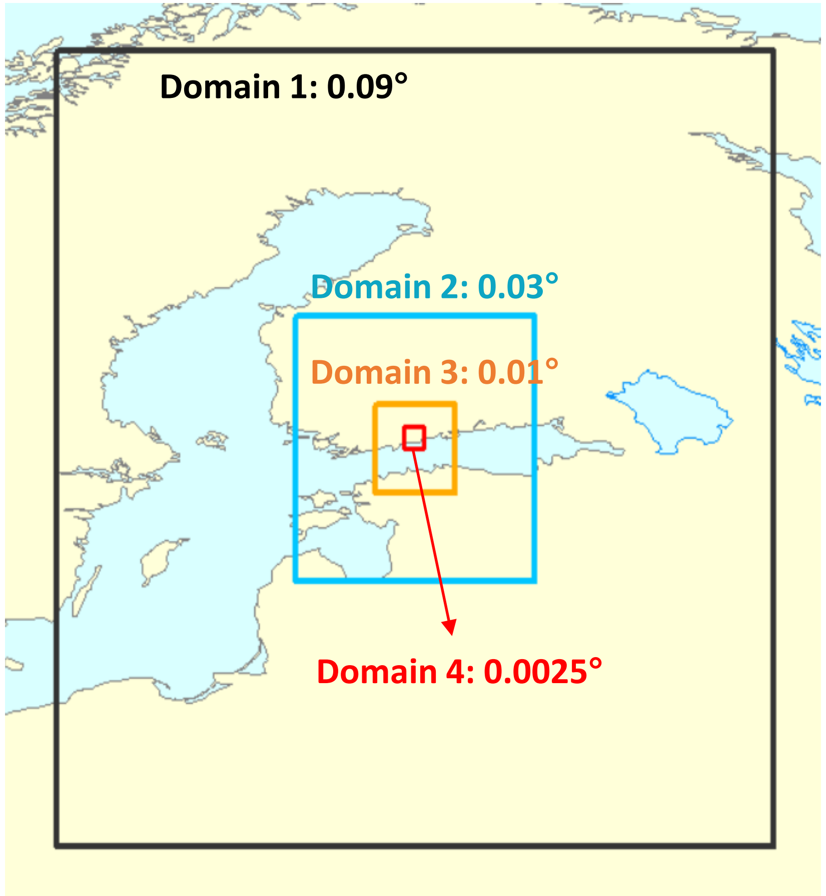
- Meteorology: WRF
- Aerosol: SILAM with sectional aerosol module

PN emission with size distribution

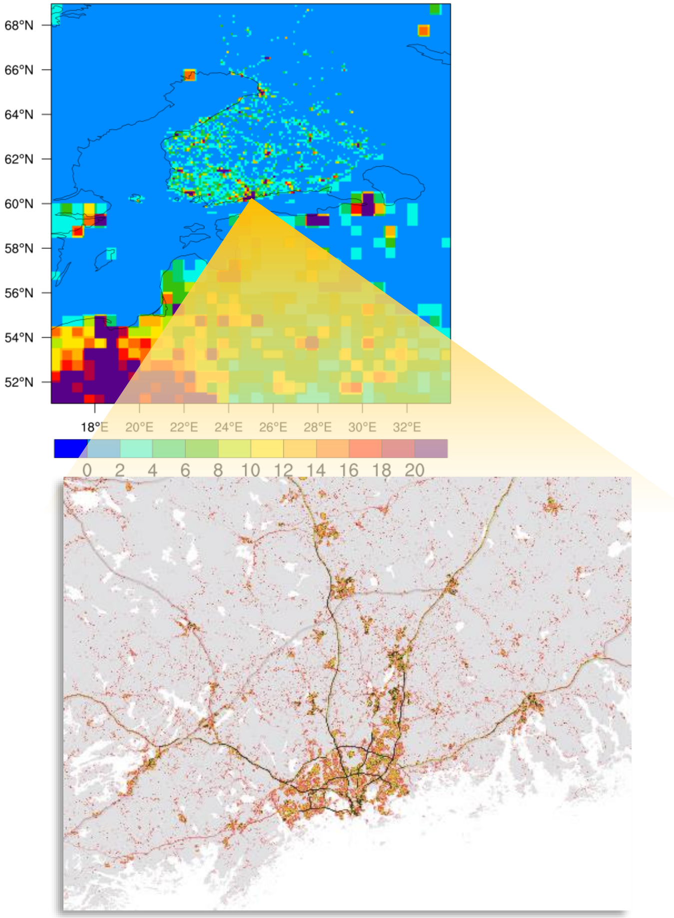
- Global: GAINS (0.1°×0.1°)
- Local: FRES (250m×250m)

SGS parameterisations

- Theoretical parameterisations, e.g., Pierce et al., *J. Aero. Sci.*, 2009
- Simulations with varying spatial resolution



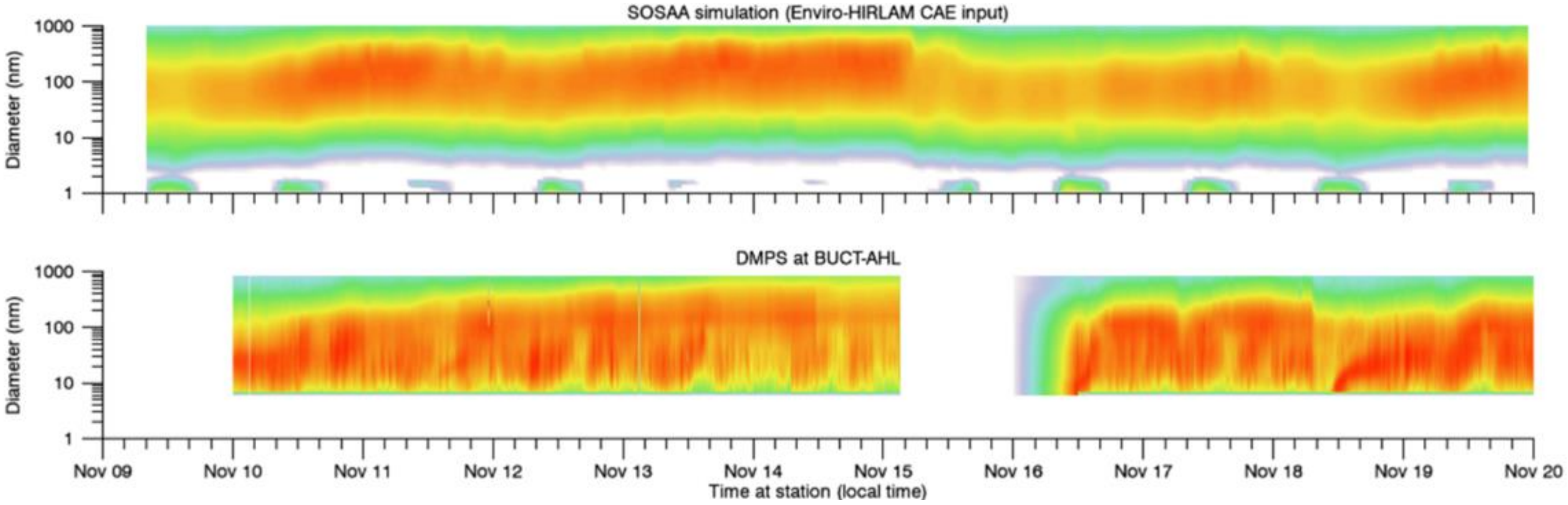
PM$_1$ emissions in domain 1 (kg/year)



Figures by Dian Ding

Lagrangian simulations for testing emission levels and size distributions, and NPF mechanisms

- Simulations with Lagrangian 1D model SOSAA (Zhou et al., 2014, Bor. Env. Res.)
  - 72 hour back trajectories for each hour of *in situ* observations
  - PN emissions from GAINS implemented
  - Currently testing the impact of adding nucleation mode traffic emissions and altering agricultural waste burning emissions for Beijing simulations



8.7.2024

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Summary

- To better predict the impacts of altered urban emissions on health and climate, we are working on
  - Improving the methods to separate traffic and NPF particles and their source rates from observations
  - Improving the description of PN emissions and sub-grid scale processes in regional/global models
- Improved methods for the above will be published by 2025

- Lagrangian models offer a good tool to search the possibly missing sources (or processes)