State of knowledge - Importance of precursor substances for the formation of UFP

Symposium: Ultrafine Particles – Air Quality and Climate

Session B.4

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Introduction

Literature survey on behalf of and funded by UBA (German Environment Agency)

Aims:

- identification of the most relevant sources of UFP, focus on SOA
- comparison of these groups regarding their contributions to PN/UFP concentrations at different sites, their emissions of UFP precursors, and the associated SOA formation potential,
- compilation of reduction potentials

For this,

- > 120 publications were evaluated, of these
- > 110 published within the last 10 years,
- nearly 50 published within the last four years

Overview

Source groups

Road traffic, air traffic, shipping, residential heating, and large combustion systems were identified as the most relevant anthropogenic sources for UFP and its precursor substances

- In this talk, the focus is on road traffic as an example
    - currently largest anthropogenic contributor to ambient UFP
    - effects shown here exemplary for road traffic also occur for other combustion sources
- Selected papers on air traffic in the appendix

Ambient PN measurements

- Compilation of long-time PN measurements at different sites
- Comparison of PN measurements in the vicinity of sources (road, airport, seaside)

Summary

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Selected papers on road traffic

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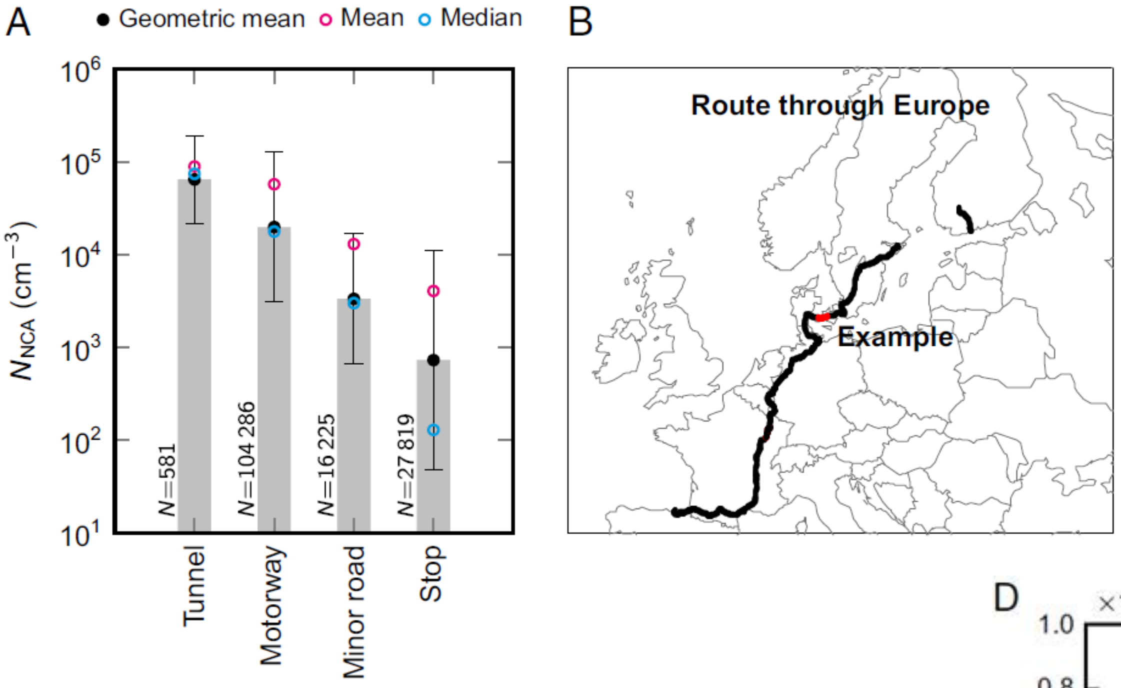
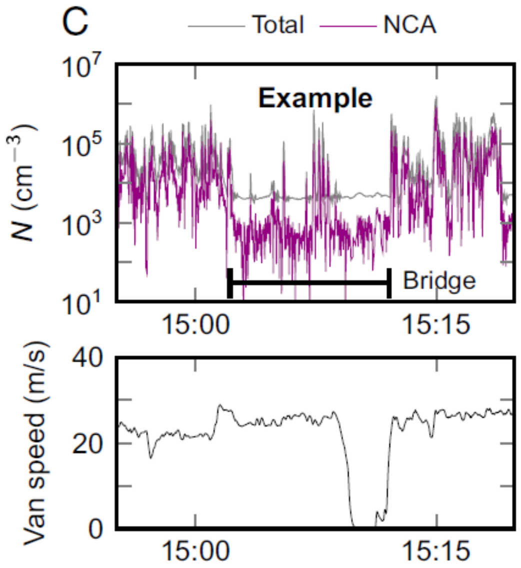
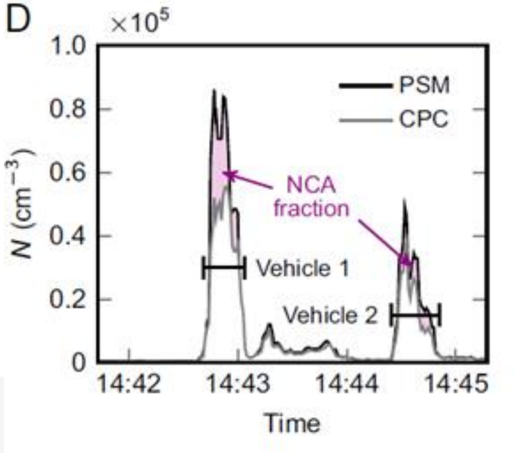
Selected papers on road traffic

Rönkkö et al. (2017): *Traffic is a major source of atmospheric nanocluster aerosol*

NCA (nanocluster aerosol, $N_{1.3}$) measurements during an on-road experiment through Europe of instruments: PSM (particle size magnifier) and CPS (condensation particle counter)







A, C: Measured NCA number concentrations ($N_{1.3}$) during the on-road experiment
D: Example for PSM measurements (including NCA) and CPC measurements (excluding NCA)

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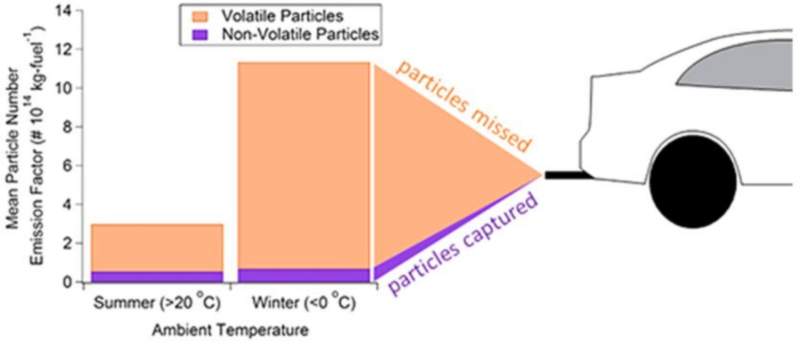
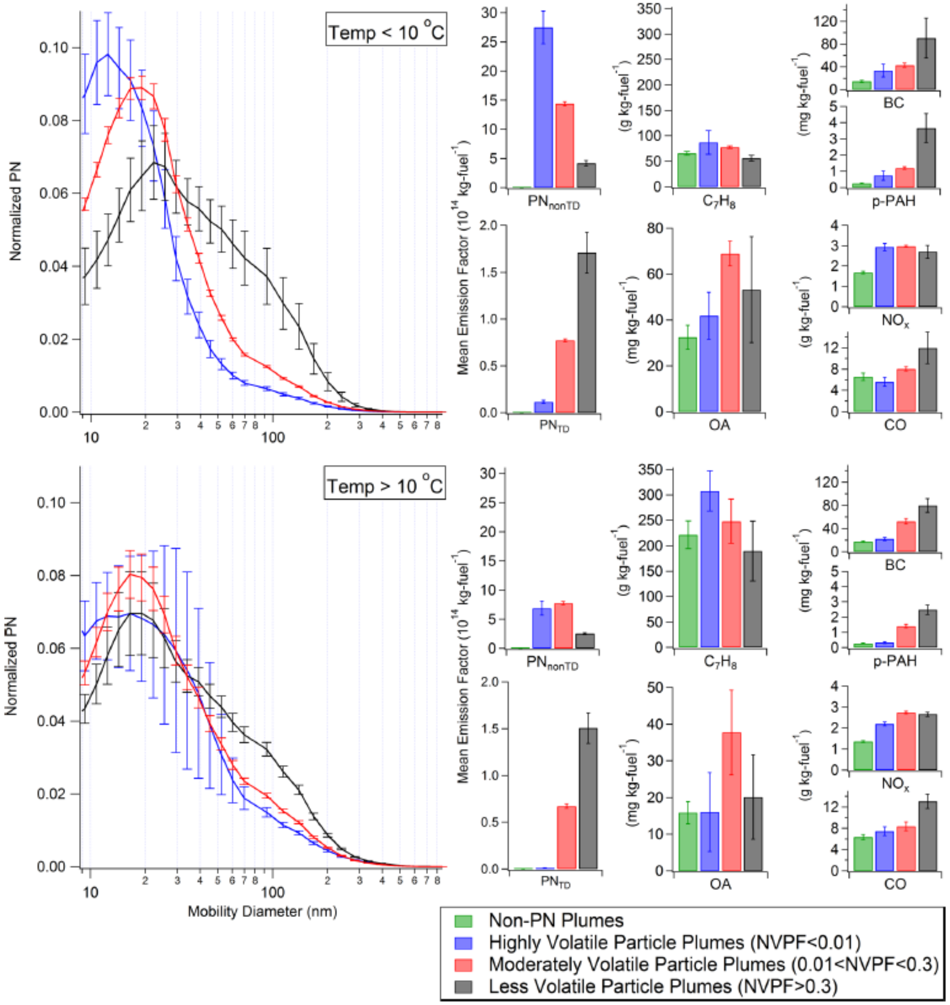
Selected papers on road traffic

Wang et al. (2017): *Real-World Emission of Particles from Vehicles: Volatility and the Effects of Ambient Temperature*

Several point sampling campaigns at a traffic influenced site in Toronto between 2013 and 2015, automated evaluation of ca. 130’000 plumes





Top: Fleet-average emission factors for solid and volatile particles in Summer and in Winter

Right: particle size distributions for 3 plume categories defined by their non-volatile particle fraction (NVPF) and corresponding average emission factors (EF) or classical air pollutants for T<10°C (top) and T>10°C (bottom)

Gas-phase EF lead to the assumption that

- Non-PN plumes could be emitted by vehicles of higher emission standards
- Less volatile PN plumes could be emitted by diesel vehicles of lower emission standards

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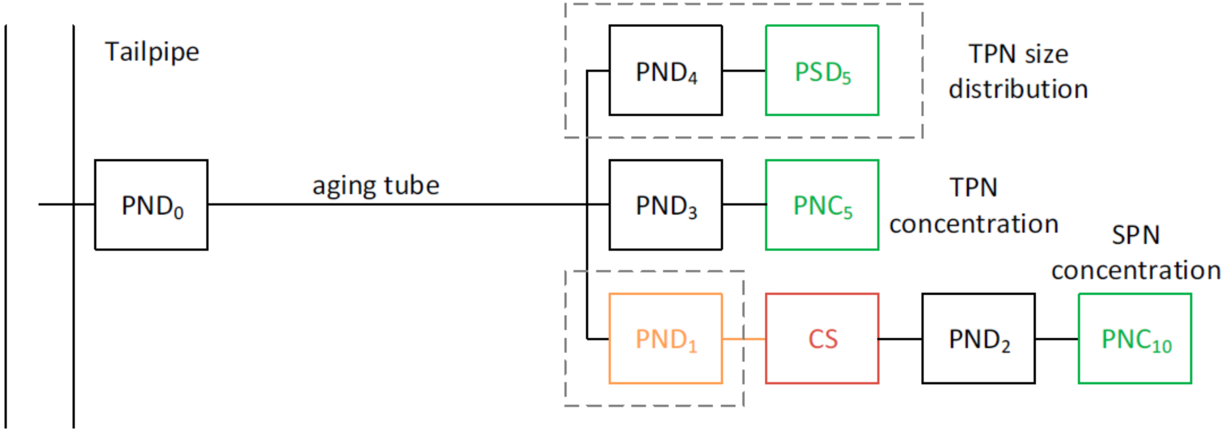
Selected papers on road traffic

Giechaskiel et al. (2022): *Revisiting Total Particle Number Measurements for Vehicle Exhaust Regulations*

Suggested measurement setup for TPN and SPN for regulation

Chose conditions in diluters and in aging tube such that nucleation is favored
-> volatile PN formation potential



green:	Particle Counters/Detectors
red:	Catalytic Stripper (CS): Temperature: 300°C
PNC:	Particle Number Counter
PND:	Particle Number Diluter
PSD:	Particle Size Distribution Instrument

dotted boxes: optional parts

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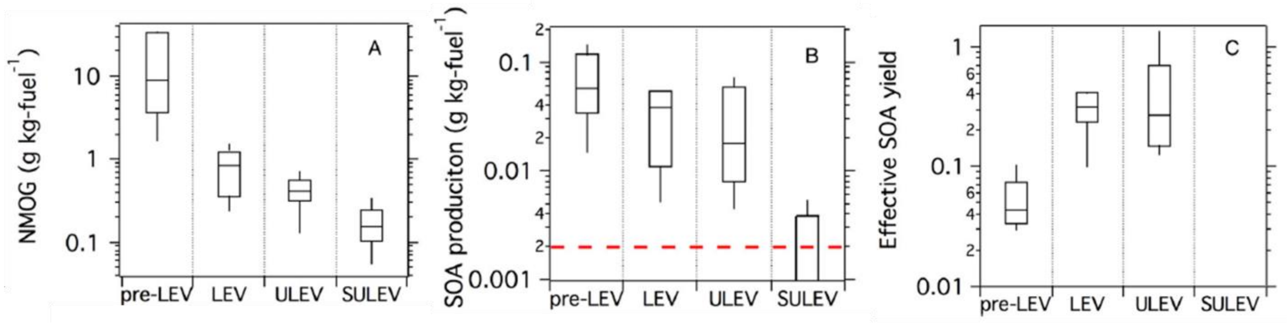
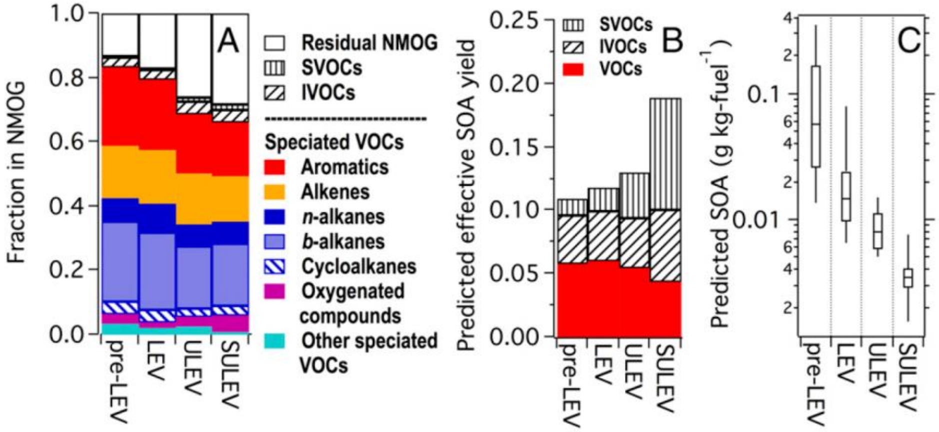
Selected papers on road traffic

Zhao et al. (2017): *Reducing secondary organic aerosol formation from gasoline vehicle exhaust*

Emission data and SOA-production data from <u>photooxidation experiments</u> with diluted gasoline exhaust for different emission standards



NMOG composition und <u>predicted</u> SOA production



- NMOG emissions <u>decrease</u> with higher emission standards
- Measured and predicted effective SOA yields <u>increase</u> with higher emission standards
- Measured SOA yields increase stronger than predicted SOA yields, Possible reason: non-linear dependency of SOA production from NMOG/NO_X-ratio
- <u>NMOG emission decrease dominates</u>: SOA production decreases with higher emission standards

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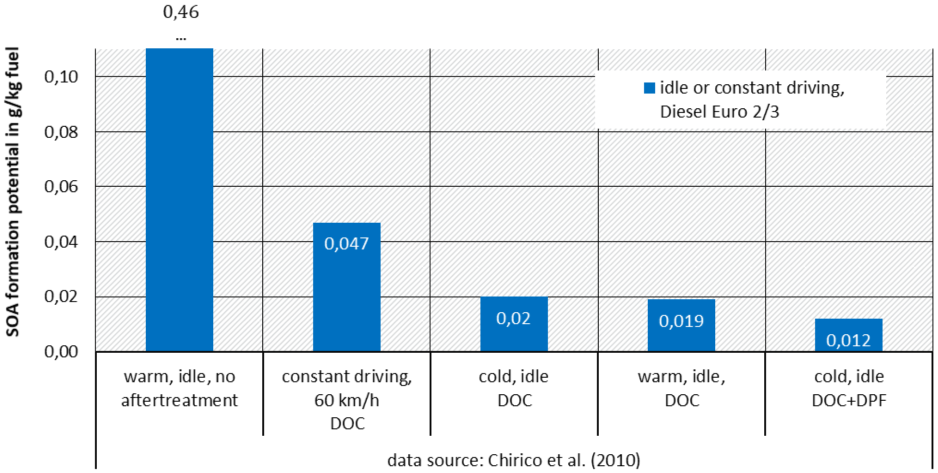
Compilation of SOA formation potentials for PC from several papers

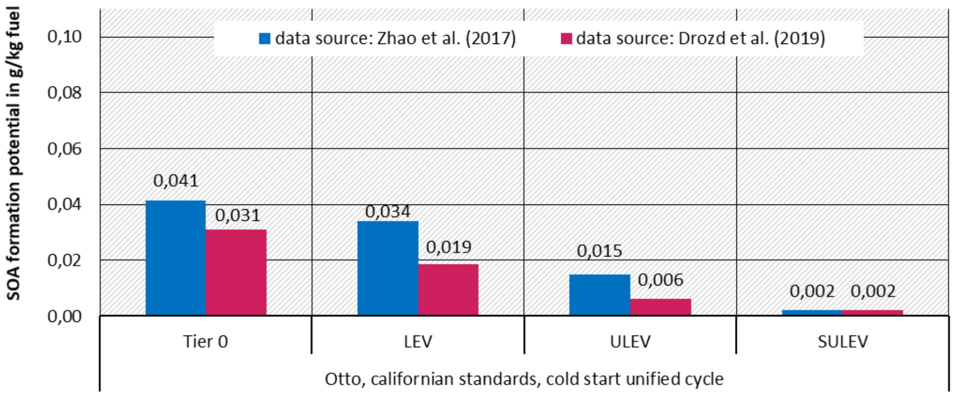
(not shown in detail)

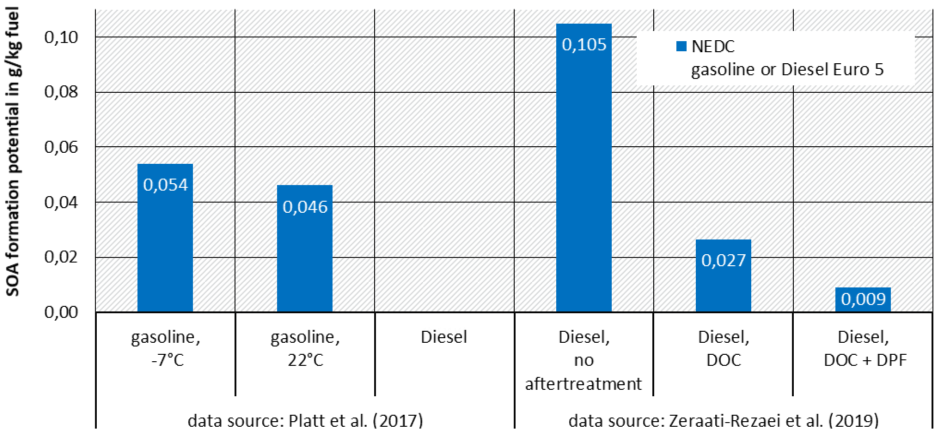
Diesel Euro 2/3:
without aftertreatment system: ~0,5 g SOA / kg fuel
DOC: reduction by factor 24

Diesel Euro 5:
for working DOC and DPF:
reductions of over 90% to 0,009 g SOA / kg fuel

Gasoline, SULEV:
0,002 g SOA / kg fuel







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Ambient PN measurements

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Long-time PN measurements: Trend

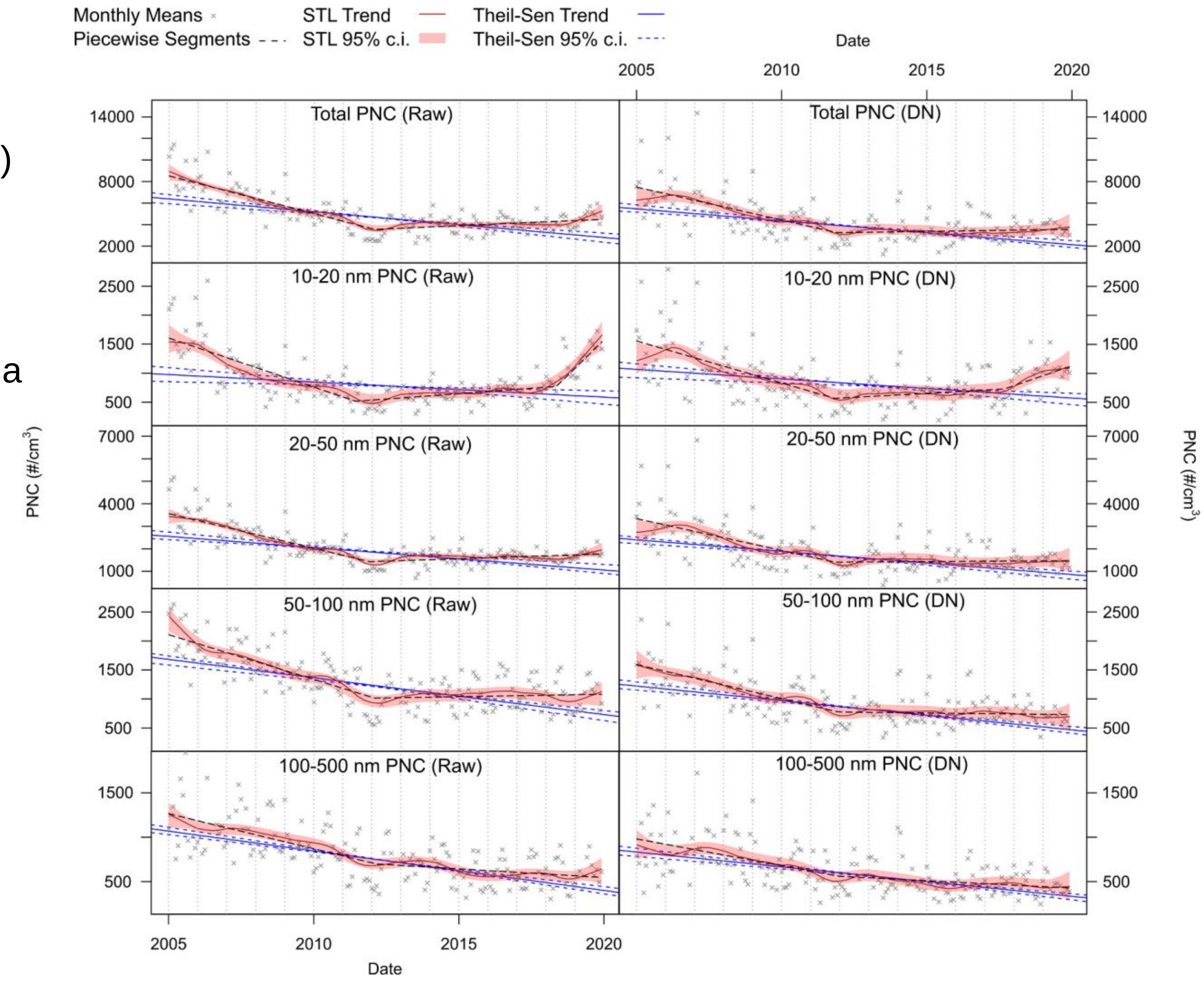
Chen et al. (2022): ***Long-term trends of ultrafine and fine particle number concentrations in New York State: Apportioning between emissions and dispersion***

Hourly PN measurements
at an urban background station in
Rochester (town in New York State)
2005 to 2019

Plot: Monthly mean PNCs
by size class (grey crosses) and
trends according to several statistica
approaches

left column: Row data,
right column:
Dispersion-normalized data

- ➢ Overall: decreasing trend
- ➢ Since ~2018 increase for nucleation particles
- ➢ Possible explanation: Cleaner air can lead to increasing particle formation



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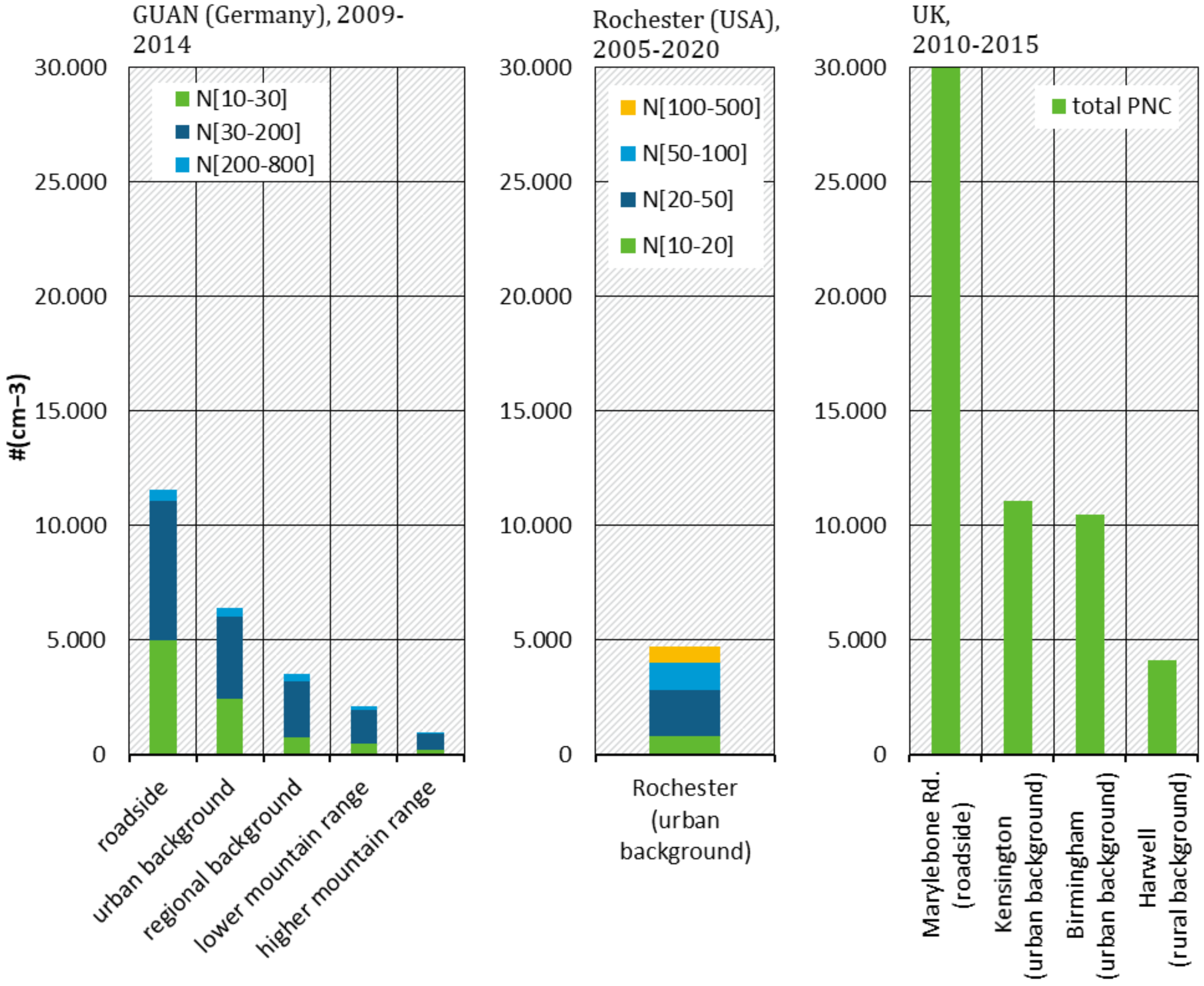
Comparison of long-time PN measurements at different sites

Measurements from the GUAN network (German Ultrafine Aerosol Network), from Rochester (previous slide) and from the UK (published by DEFRA, 2018)

<u>Caveat</u>: Comparison only meant to be <u>qualitative</u>:

- particle size ranges differ
- time periods differ

- ➢ Marylebone Road:
  traffic hotspot,
  very high PN concentration
- ➢ urban background PN
  from GUAN and Rochester:
  comparable
- ➢ GUAN and UK:
  strong gradient between traffic stations and rural background



data source: Sun et al. (2019)

data source: Chen et al. (2022)

data source: DEFRA(2018)

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PN measurements in the vicinity of sources

Caveat: Comparison only meant to be qualitative:

- particle size ranges differ
- time periods differ
- distance measurement site – source differ

- ➢ PN high in vicinity of sources (~20.000 #/cm^3)
- ➢ strong gradients
- ➢ contribution Heathrow in Harlington: ~1/3 (not shown: measurements near Schiphol comparable)
- ➢ Road traffic in vicinity of airports still important

Distance measurement site - source

0 km | **1,2 km** | **5 / 6 km** | **7-20 km**

data source: Rönkkö et al. (2017)

data source: Masiol et al. (2017)

data source: Jacobi et al. (2016)

data source: Ausmeel et al. (2019)

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Summary

Summary (general)

(including papers not selected to show here)

Which source groups are important?

- All the year at ground-based measurement stations in Germany:
  Road traffic contributes most to total PN and SOA
- Winter:
  SOA contributions from wood burning for residential heating important
- Near large point or line sources (e.g., airports, shipping lines, harbours) contributions of these sources may dominate

For all source groups: Volatile contributions dominate

Which gas-phase precursors are important?

- Inorganic gases
  NOx, NH_3, SO_2 as precursors for SIA, secondary inorganic aerosol
- Organic gases:
  - the less volatile they are, the higher is their SOA formation potential (2-dim volatility basis set by Donahue et al., 2012)
  - SOA-formation also by aromatics and other cyclic HC
- Condensates und „delayed primary“ PN:
  actually POA (primary organic aerosol), enter aerosol phase after cooling without further chemical reactions. Should be subject to abatement measures as well as the gas-phase precursors

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Summary (Road Traffic)

(including papers not selected to show here)

- For UFP, exhaust emissions are more important than non-exhaust emissions (abrasion)
- Currently, emissions of solid PN > 10 nm are limited
  larger contributions of volatile and secondary traffic related PN are not accounted for
- Emission limits for the formation potential of volatile PN are theoretically possible, conditions have to be agreed upon
- For increasingly stringent emission standards:
  VOC emissions are reduced, their specific SOA yields are increased, overall, for the SOA formation potential, the reduction dominates
- If emission standards are observed, reductions of traffic related ambient UFP mass are expected in future, so that the relative importance of other source groups will grow
- A decrease in UFP mass not necessarily also reduces PN, cleaner air may favor nucleation over condensation

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Summary (Abatement potentials)

(including papers not selected to show here)

Where are abatement potentials?

- Road traffic:
  - Substantial reductions (~90%) for PN (total) and SOA achieved in the past (Euro 5) by catalysts and filters, further reductions (Euro 6, 7) expected if standards are observed
  - Currently limited: Solid particles: up to Euro 6: > 23 nm, Euro 7: > 10 nm
    Theoretically possible: Limitation of PN formation potential for volatile PN (delayed primary)
- Air traffic and shipping:
  - Air traffic: first limits for solid PN since January 2023, no PN limits for shipping
  - EF for SOA-precursors as high as for road traffic before the introduction of limits
  - EF for PN (total) for air traffic even higher (high temperatures)
  - ➢ Abatement potentials exist
- Residential heating:
  - Potential to reduce emissions of condensables by replacing traditional manual stoves by automatic stoves
- Large combustion plants:
  - Reduction potential by switching off coal-fired power plants

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Thank you!

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Selected papers on air traffic

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Selected papers on air traffic

Masiol et al. (2017): ***Sources of sub-micrometre particles near a major international airport***

Measurement campaigns:

Summer 2014 (warm season)
Winter 2014/2015 (cold season)

in **Harlington**,
village 1,2 km north of
London Heathrow airport

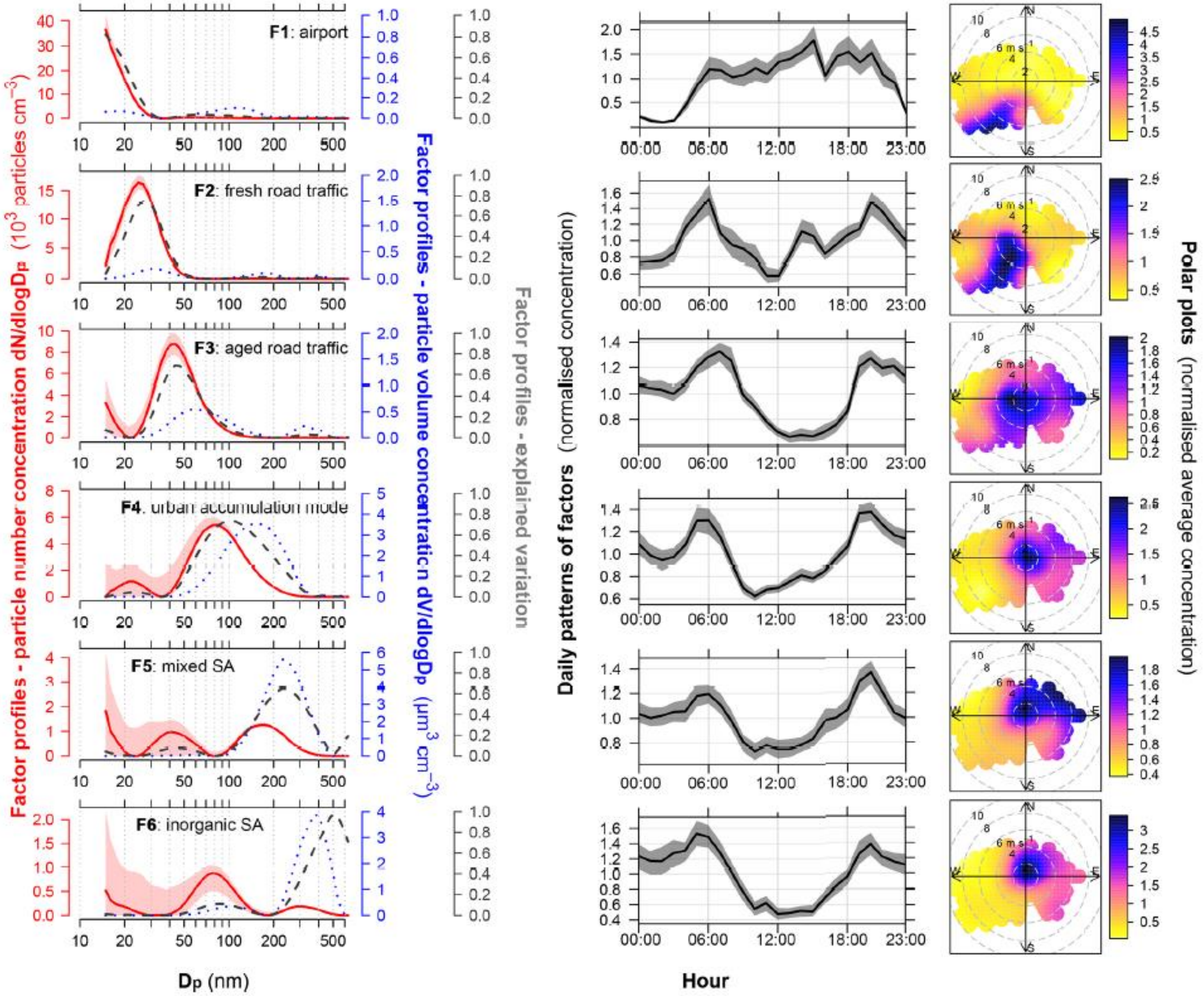
right: PMF-analysis
(warm season)

Main results both seasons:

PN contributions at the site

Airport:
30% to 35%

Fresh road traffic:
24% to 36%
Aged road traffic:
16% to 21%



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Selected papers on air traffic

Kılıç et al. (2018): ***Identification of secondary aerosol precursors emitted by an aircraft turbofan***

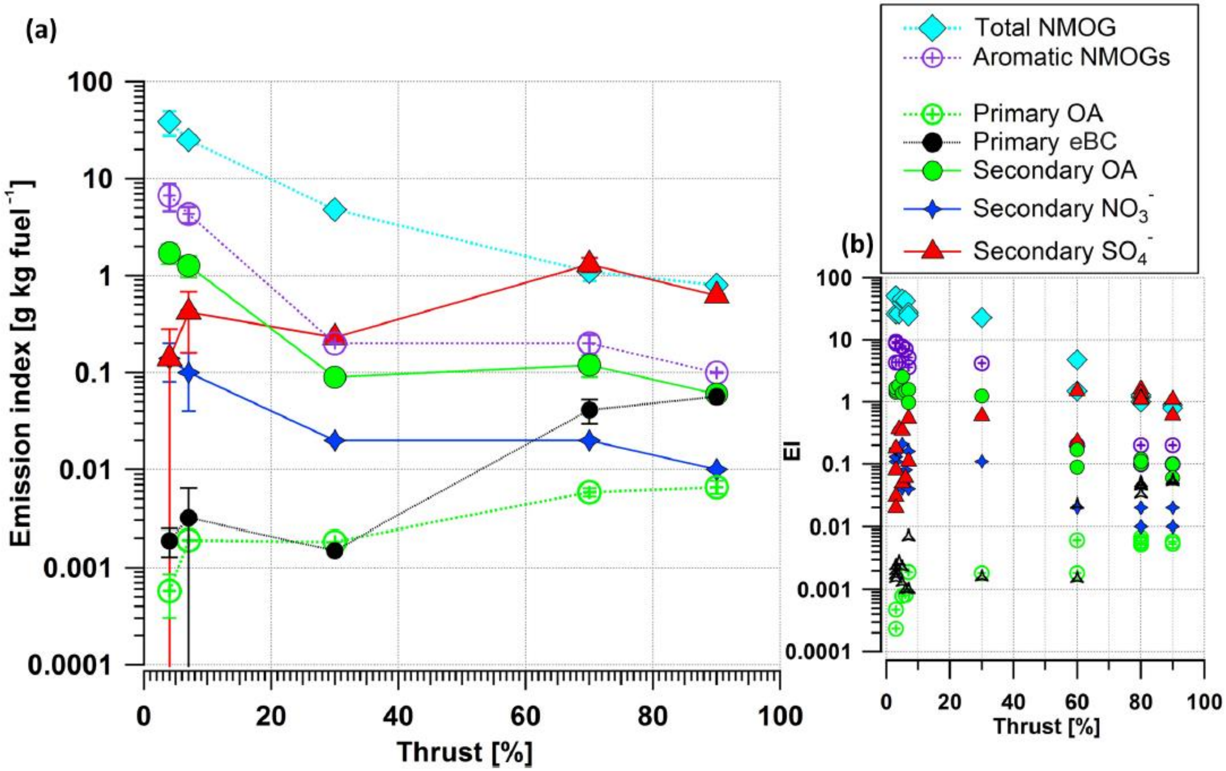
Measurements of emission indices for a CFM56 variant turbofan motor in a test cell at Zurich Airport after aging

right:
Results for emission indices

From these, Kılıç et al. (2018)
Estimate for Zurich 2010:
SOA formation from
Zurich airport
between 5,5 and 13,2 t

For comparison: Road traffic:
in an area of 200 km^2
around Zurich: ca. 94 t

But: Airport is a point source, so
its influence may dominate locally



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Literature

Literature

Ausmeel et al. (2019): *Methods for identifying aged ship plumes and estimating contribution to aerosol exposure downwind of shipping lanes.* Atmospheric Measurement Techniques, 12(8), 4479–4493. https://doi.org/10.5194/amt-12-4479-2019

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Jacobi et al. (2016): *Messung ultrafeiner Partikel im Umfeld des Frankfurter Flughafens.* Informationen zum Thema UFP Fluglärmkommission, 28. September 2016

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