What goes up must come down –
studying urban aerosol dynamics by quantifying
vertical particle exchange
Atmospheric aerosol budget

Quantitative atmospheric aerosol budget:
- Identify and quantify major aerosol sources
- Identify and quantify major aerosol sinks
- Identify and quantify transport mechanisms

Climate effects

Health effects

Atmospheric chemistry
Global scale aerosol sources and horizontal transport

[https://gmao.gsfc.nasa.gov/research/science_snapshots/2020/Australia_fires_smoke.php]
Urban scale aerosol sources and transport

- Multiple ground-based and atmospheric particle sources and sinks
- Vertical and horizontal transport

- Long-range transport
- Secondary aerosol formation
- Wet and dry deposition
- Emission sources at the ground

Advection
Urban boundary layer

Urban boundary layer = urban pollution island

driven by local emission sources and boundary layer dynamics
Turbulent particle transport in boundary layer

Visualization of particle transport by LIDAR measurements in Dixon, CA:

- 2007/05/22 06 - 07 PDT
- 2007/05/22 07 - 08 PDT
- 2007/05/22 12 - 13 PDT
- 2007/05/22 13 - 14 PDT
Atmospheric turbulence - the energy cascade

Big whirls have little whirls that feed on their velocity;
And little whirls have lesser whirls, and so on to viscosity. (L. F. Richardson)
Aerosol dynamics in urban boundary layer

In urban boundary layer, highly dynamic

- particle concentrations
- particle size distributions
- chemical composition
- spatial distribution

due to variability of

- emission sources
- emission strengths
- turbulent transport
- boundary layer height
- atmospheric chemistry

Direct measurement of turbulent transport to quantify net vertical particle exchange
Quantification of turbulent particle exchange by micrometeorological methods:

- Eddy Covariance
- Relaxed Eddy Accumulation
- Flux-Gradient relationship (K approach)
- Disjunct Eddy Covariance, Mixed Layer Gradient, …

**Eddy Covariance:**

Relating vertical wind speed $w$ and particle concentration $c$;

The turbulent aerosol flux is covariance of $w$ and $c$: $F_{\text{aerosol}} = \overline{w'c'}$

**Instantaneous measurement:**

- wind fluctuation, here updraft: $w > 0$
- concentration fluctuation, here: $c > \overline{c}$

Averaging yields net particle exchange
Eddy covariance aerosol flux measurements

Requirements:
- Fast measurements – to resolve all relevant eddy sizes
- Steady state conditions – typically in 30 min averaging intervals
- Horizontal homogeneity – separate wind sectors in urban environments

Measurements:
- vertical wind speed, e.g. with sonic anemometer
- total particle number concentration, e.g. with condensation particle counter
- particle mass concentration, e.g. with optical particle counter
- size-resolved particle concentration, e.g. with optical particle counter
- chemically resolved particle concentration, e.g. with aerosol mass spectrometer
PM2.5 mass concentrations and fluxes

PM2.5 concentrations and fluxes at two different sites in the province of Lecce (Donateo et al., 2006):

Maglie, December 2004

Lecce, May 2003

[Donateo et al., 2006]
Diurnal cycles of PM2.5 fluxes in Florence, and proxies for road traffic activity and domestic heating intensities (Gioli et al., 2013):

- Early morning: net deposition
- Afternoon and evening: two net emission peaks
- Morning: rush hour and increased gas consumption
- Evening: secondary peak

No direct temporal relation of potential PM sources and net PM fluxes
Particle number fluxes and traffic

Total particle number fluxes in Helsinki, in wind sectors (Ripamonti et al., 2013):
- Strong emission fluxes in urban and road sectors
- Less traffic and particle emission fluxes on weekends
Particle number fluxes and traffic: flux footprints

**Total particle number fluxes** in Lecce (Conte et al., 2018; 2021):

- Flux footprint indicates measurement mostly influenced by road traffic
- Diurnal cycle of particle flux and traffic activity are similar
- Linear relationship of traffic intensity and total particle number fluxes
## EC-derived particle emission factors of road traffic

Number of particles emitted from road traffic per kilometer:

\[ 0.8 \cdot 10^{14} \text{ km}^{-1} – 6.0 \cdot 10^{14} \text{ km}^{-1} \]

<table>
<thead>
<tr>
<th>City</th>
<th>Emission factor ([10^{14} \text{ km}^{-1}])</th>
<th>Lower diameter</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stockholm</td>
<td>1.4 ± 0.1</td>
<td>&gt; 11 nm</td>
<td>Maartensson et al. (2006)</td>
</tr>
<tr>
<td>Helsinki</td>
<td>3.0 ± 1.1</td>
<td>&gt; 5 nm</td>
<td>Järvi et al. (2009)</td>
</tr>
<tr>
<td>Lecce</td>
<td>5.0 ± 2.4</td>
<td>&gt; 9 nm</td>
<td>Contini et al. (2012)</td>
</tr>
<tr>
<td>Helsinki</td>
<td>6.0 ± 0.2 (cold)</td>
<td>&gt; 6 nm</td>
<td>Ripamonti et al. (2013)</td>
</tr>
<tr>
<td></td>
<td>3.7 ± 0.1 (warm)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Innsbruck</td>
<td>0.8 ± 0.1</td>
<td>&gt; 11 nm</td>
<td>Deventer et al. (2018)</td>
</tr>
<tr>
<td>Lecce</td>
<td>2.2 ± 0.6</td>
<td>&gt; 9 nm</td>
<td>Conte &amp; Contini (2019)</td>
</tr>
</tbody>
</table>
Spatial and temporal variability of particle fluxes

If spatial distribution of ground-based emission sources is known:
Eddy covariance flux measurement and footprint analysis provide spatially averaged net particle exchange.

If emission strengths of dominant particle sources change in time:
Weekly cycles and seasonal differences indicate temporal trends in emission strengths.

But:
- Variable atmospheric conditions and variable efficiency of turbulent transport
- Only few long-term particle flux measurements to study seasonal differences

Measurement approaches for additional information:
- Size-resolved particle flux measurements
- Chemically resolved particle flux measurements
Urban emission sources and particle sizes

Characteristic particle number size distributions (normalized) of various urban aerosol sources (Wu and Boor, 2020):

- **Road traffic** (exhaust of various vehicle types)
- **Brake wear** (non-exhaust traffic emission)
- **Biomass burning** (various fuels)
- **Cooking** (various stoves)
- **Energy and heating production**
Size-resolved aerosol number and mass fluxes

Size-resolved particle emission factors from road traffic (Conte and Contini, 2019)
Size-resolved urban aerosol fluxes

Innsbruck Air Quality Study INNAQS 2015
(Deventer et al., 2018)

- Strong emission of small particles
- Deposition of larger particles
- Bi-directional transport of particles

Size-resolved aerosols number fluxes

D = 6 nm
D = 30 nm
D = 400 nm

[Deventer et al., 2018]
Size-resolved aerosol number and mass fluxes

Innsbruck Air Quality Study INNAQS 2015 (Deventer et al., 2018)

- Aerosol number flux is controlled by smallest particles (diameter < 100 nm): emission of particles
- Aerosol mass flux is controlled by larger particles (diameter > 100 nm): emission of particles (D < 300 nm) and deposition of particles (D > 300 nm)

\[
\frac{dF_N}{d \log D} [10^6 \text{ m}^{-2} \text{s}^{-1}]
\]

\[
\frac{dF_M}{d \log D} [\text{ng m}^{-2} \text{s}^{-1}]
\]

Aerosol number flux

Aerosol mass flux

Green: with storage and deliquescence correction
Grey: without storage and deliquescence correction
Black carbon aerosol fluxes in Beijing

Diurnal cycle of black carbon number and mass fluxes measured by Eddy covariance combining sonic anemometer and SP2 (Joshi et al., 2020):

- Morning rush hour clearly visible in summertime BC number fluxes
- Similar total mass emission fluxes in summer (6.1 ng m\(^{-2}\) s\(^{-1}\)) and winter (5.5 ng m\(^{-2}\) s\(^{-1}\))
- Road traffic dominant source of black carbon fluxes
Comparison of measured black carbon emission fluxes and estimates based on MEIC 2013 emission inventory (Joshi et al., 2020):

- Large deviations between top-down and bottom-up approaches
- Measured total black carbon emission fluxes smaller than emission inventory estimates (all sectors) by a factor of 59 in winter and 47 in summer, and by a factor of 38 compared to transport sector only
Chemically resolved urban aerosol fluxes

Aerosol mass spectrometry and eddy covariance in Mexico City, 2006 (Zalakeviciute et al., 2012)

Hydrocarbon-like organic aerosol

Nitrate aerosol

Oxygenated organic aerosol

Biomass burning organic aerosol

[Zalakeviciute et al., 2012]
Summary of previous studies

In urban environments...

- **Net vertical particle exchange**
  depending on urban sources and atmospheric conditions

- **Strong vertical exchange of traffic-related aerosol**
  with dominant contribution of sub-100 nm particles

- **Bi-directional size-resolved fluxes**
  particle number: typically net emission
  particle mass: net emission or deposition

- **Chemically resolved particle fluxes**
  strongly site-specific and dependent on flux footprint

**Measurement challenges due to rapid changes of aerosol concentrations**

→ Spectral analysis
Rapid changes in urban particle concentrations

**Time series** of particle number concentration and vertical wind speed $w$:

Eddy covariance requires steady state conditions: often violated

Steady state assumption not required for flux reconstruction by wavelet analysis

→ reconstruction of instantaneous particle fluxes
Wavelet analysis: spectral decomposition

**Wavelet analysis** of particle number time series:
Comparison of eddy covariance and wavelet fluxes

**Wavelet flux estimates** confirm validity of eddy covariance measurements:

- Particle number [cm$^{-3}$]
- $w$ [m s$^{-1}$]
- Aerosol number flux [$10^6$ m$^{-2}$ s$^{-1}$]
- 1 min CWT flux [a.u.]

[Graph showing wavelet flux estimates and eddy covariance measurements]
Spectral flux estimates: added benefits

- Particle flux estimates for time periods shorter than 30 min
- Check for spectral similarity of particle fluxes
- Cospectral peak indicates dominant time scale/eddy size
- Contribution of different time scales/eddy sizes to particle flux

Median diurnal cycle of particle number fluxes reconstructed from wavelet analysis

[Conte et al., 2021]
Turbulence and secondary aerosol formation
Turbulence and secondary aerosol formation

size-resolved fluxes:

size distribution
20 nm – 3 µm
32 m above ground

size distribution
8 nm – 700 µm
at ground level

Average of 7 NPF

00:00 04:00 08:00 12:00 16:00 20:00 24:00
Turbulence and secondary aerosol formation

[Graph showing diameter [μm] over time from 00:00 to 24:00]

[Diagram illustrating chemical time scale vs. transport time scale]

[Deventer et al., 2015]
Turbulence and secondary aerosol formation

size-resolved fluxes:

size distribution
20 nm – 3 µm
32 m above ground

size distribution
8 nm – 700 µm
at ground level

24 August 2013
Conclusions

**Vertical particle exchange** can be measured and quantified by micrometeorological methods

**Eddy covariance** is a valuable tool to study urban aerosol sources and dynamics

**Urban particle emission fluxes** are frequent but deposition periods and bi-directional fluxes occur

**Spectral analysis tools** such as wavelet analysis are important in highly dynamic urban environments

**Integrated field experiments** including flux measurements of particles and trace gases are needed to study chemistry-transport interactions
Grazie

M. Conte, M. J. Deventer, T. Karl, O. Klemm, S. Mayor, S. Schmitt, L. von der Heyden, and many others.

Funding by German Research Foundation DFG, German Academic Exchange Service DAAD, TU International Office

https://www.leccenelsalento.it/basilica-s-croce