Since 2004 the aerosol measurement station in Augsburg, Southern Germany, has supplied data for detailed specification of physical and chemical particle properties by novel measurement techniques. The measurement site and measurement program was described more detailed by Pitz et al. (2008a and b). Here we used the particle size distribution data (PSD) collected in winter 2006/07 to identify the most relevant local and regional sources of ambient particles in Augsburg by positive matrix factorization (PMF) method. PMF is a multivariate tool that decomposes a matrix of data sample into two sub-matrices, the factor profiles and factor contributions. It has been developed by Paatero and Tapper (1994) and Paatero (1997) and was used to resolve sources on the basis of observation without detailed prior knowledge of the sources and source profiles. The model used in this study was EPA PMF 3.0, developed by U.S. EPA based on the ME-2 algorithm (http://www.epa.gov/heasd/products/pmf/pmf.html).

In Figure 1 the source profiles for number and volume size distribution are shown for all sources identified by PMF in our study: two traffic factors, secondary aerosols, stationary combustion, nucleation particles, re-suspended dust and a long range transported dust factor. Two traffic factors were dominated by ultrafine particles (diameter <100 nm), contributed 25% and 40% to total particle number concentration.
The stationary combustion factor consisted of particles around 100 nm, and accounted for 26% of total NC. Re-suspended dust was composed of particles with diameter >2.5 µm. The source types were identified by the following information, (I) factor profiles with particle number and volume size distribution, (II) contribution of each factor to total number and volume concentration, (III) diurnal variation of each factor, (IV) Spearman correlation coefficients between each factor and measured gaseous species as well as chemical composition of particles, and (V) the directionality of sources provided by CPF (conditional probability function) analysis.

Small-scale spatial and temporal variation measurements of total particle number concentration (PNC) suggested that the Augsburg site is representative for urban background conditions (Cyrys et al., 2008) as strong temporal correlations were observed between this site and three other background sites in Augsburg. On the other hand, the PNCs were not homogeneously distributed over the whole study area. Also the impact of specific particle sources differs for different measurement sites as shown in Figure 2. Whereas the traffic factor contributes 45% to the PM$_{10}$ mass concentration at KP (traffic site), the contribution of this source to PM$_{10}$ mass at the rural sites We and Ho is much lower (5%).

![Figure 2: Mean PM$_{10}$ mass concentrations measured at eight different monitoring sites in Augsburg and the impact of different particle sources to the PM mass concentration (KP - traffic site, BP - traffic influenced urban background site, LO, LfU, Bifa & Ki_Li - urban background sites, We & Ho - Rural/ Tower sites)](image)

It implicates that exposure assessment to ambient fine and ultrafine particles for long-term epidemiological studies remains a difficult challenge. To assess small-scale air pollution contrasts land-use regression approach was introduced (Briggs et al. 1997, Hoek et al., 2008). Land-use regression combines measuring of air pollution at a limited number of sites across the study area and development of prediction models using variables obtained by geographic information systems (GIS). The model can then be applied to a large number of the home addresses of the study cohort. In the framework of a European Study of
Cohorts for Air Pollution Effects (ESCAPE) we measured PM$_{2.5}$ and PM$_{10}$ concentrations at 20 sites in the region Munich/Augsburg. The measurements were conducted from October 2008 to September 2009 in different seasons. After an adjustment for season, annual means for each location were calculated. The annual average for PM$_{10}$ varied between 16 and 34 µg/m$^3$, and between 10 and 18 µg/m$^3$ for PM$_{2.5}$. To explain the variability of the PM concentrations across the study area we collected both European-wide GIS variables (such as CORINE land use, road network) as well as local GIS variables (such as traffic intensity, street configuration, population density). The preliminary regression models have an adjusted R$^2$ of 0.64 and 0.92 for PM$_{2.5}$ and PM$_{10}$, respectively. It suggests that the modelling approach described here can be used to predict small-scale spatial variation of ambient particles in an urban and rural area.

The monitoring station in Augsburg is also a part of the Germany-wide network for characterization of fine and ultrafine particles (GUAN: German Ultrafine Aerosol Network). It was established in 2008 and it combines eleven monitoring stations located at different environments (rural, urban background and traffic) spread out across Germany (Birmili et al., 2009). Due to the harmonization of the measurement techniques and common quality assurance strategy at all involved monitoring sites, GUAN will allow a common analysis of the data, e.g. with regard to spatial and temporal variation of source contributions to the ambient aerosol over Germany.


Briggs DJ; Collins S; Elliott P; Fischer P; Kingham S; Lebret E; Pryl K; VanReeuwijk H; et al. (1997) Mapping urban air pollution using GIS: a regression-based approach. Int J Geograph Inform Sci. 11, 699-718.


Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung

J. Cyrys
J. Gu, M. Pitz, S. von Klot, J. Schnelle-Kreis, W. Birmili, A. Peters
Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung

Stadt Augsburg

Region Augsburg

Deutschland

Messstation Augsburg
Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung

Source apportionment
Spatial and temporal variation

Deutschland
Region Augsburg
Stadt Augsburg
Messenstation Augsburg
Exposure characterization relevant for health research: Aerosol measurement site in Augsburg
Mean ambient particle size distribution Augsburg
14th Mar 07 - 17th Dec 08
Positive matrix factorization (PMF)

How does PMF work?

Particle data sample

\[ X_{ij} = \sum_{k=1}^{p} g_{ik} f_{kj} + e_{ij} \]

\[ X = GF + E \]
Seven factors were resolved: Re-suspended dust, fresh traffic emission, aged traffic emission, stationary combustion, nucleation particles, secondary aerosols and long range transported dust.
Source Apportionment Using PSD data at FH site
(Dec 21st, 2006 – Mar 23rd, 2007)

Contribution to

**particle number**

- Combustion sources: 26%
  - Aged traffic emissions: 40%
  - Fresh traffic emissions: 25%
  - Secondary aerosols: 1%
  - Nucleation: 4%
  - Re-suspended dust: 3%
  - Long range dust: 1%

**particle mass**

- Combustion sources: 41%
  - Aged traffic emissions: 10%
  - Fresh traffic emissions: 1%
  - Secondary aerosols: 29%
  - Nucleation: 1%
  - Re-suspended dust: 9%
  - Long range dust: 9%
Spatial Variation of Particle Sources in Augsburg (PM$_{10}$) (winter 2007/08)

Four site categories

- **Traffic site**: KP
- **Traffic influenced urban background**: BP
- **Urban background**: LO, LfU, Bifa & Ki_Li
- **Rural/Tower site**: We & Ho
Spatial Variation of Particle Sources in Augsburg (PM$_{10}$) (winter 2007/08)

- **Königsplatz**
  - PM$_{10}$: 41.5 µg m$^{-3}$
  - Dust: 5%
  - Traffic: 45%
  - Nitrate: 14%
  - Biomass: 14%
  - NaCl: 12%
  - Sulfate: 10%

- **Bourgsplatz**
  - PM$_{10}$: 26.6 µg m$^{-3}$
  - Dust: 6%
  - Traffic: 22%
  - Nitrate: 22%
  - Biomass: 25%
  - NaCl: 8%
  - Sulfate: 17%

- **Lo, LfU, Bifa & Ki_Li**
  - LO: 23.4 µg m$^{-3}$
  - Dust: 6%
  - Traffic: 13%
  - Nitrate: 22%
  - Biomass: 31%
  - NaCl: 7%
  - Sulfate: 19%

- **Ho & We**
  - Ho: 19.0 µg m$^{-3}$
  - Dust: 7%
  - Traffic: 5%
  - Nitrate: 37%
  - Biomass: 22%
  - NaCl: 7%
  - Sulfate: 22%

- **We**: 17.8 µg m$^{-3}$
  - Dust: 8%
  - Traffic: 5%
  - Nitrate: 30%
  - Biomass: 29%
  - NaCl: 7%
  - Sulfate: 22%

- **LfU**: 23.6 µg m$^{-3}$
  - Dust: 6%
  - Traffic: 13%
  - Nitrate: 24%
  - Biomass: 28%
  - NaCl: 7%
  - Sulfate: 22%

- **Ki_Li**: 19.6 µg m$^{-3}$
  - Dust: 8%
  - Traffic: 13%
  - Nitrate: 26%
  - Biomass: 28%
  - NaCl: 6%
  - Sulfate: 22%
Temporal Variation of Particle Sources in Augsburg (PM$_{10}$) (winter 2007/08)
Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung

Source apportionment
Spatial and temporal variation

Deutschland
Region Augsburg
ESCAPE

Stadt Augsburg

Messstation Augsburg

HelmholtzZentrum münchen
German Research Center for Environmental Health
ESCAPE project

European Study of Cohorts for Air Pollution Effects

http://www.escapeproject.eu/index.php
ESCAPE Locations
Exposures modelled for Augsburg study area based on measurements within ESCAPE

$\text{PM}_2.5$

$\text{PM}_{10}$

$\text{PM}_{\text{coarse}} (\text{PM}_{10} - \text{PM}_{2.5})$

Absorption coefficient $\text{PM}_{2.5}$

$\text{NO}_x$ and $\text{NO}_2$

“Source-specific” elements from XRF

Background versus Traffic exposures
### Annual averages for NO₂ and PM₁₀ in Munich/Augsburg

<table>
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<th>Site Type</th>
<th>GPS LAT</th>
<th>GPS LON</th>
<th>NO₂ adj</th>
<th>NO₂ unadj</th>
<th>PM₁₀ unadj</th>
<th>PM₁₀ adj</th>
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ESCAPE – Entwicklung von LUR

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<th>Land\textsubscript{300}</th>
<th>Alt</th>
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<td>45</td>
<td>5</td>
</tr>
<tr>
<td>45</td>
<td>20,000</td>
<td>50</td>
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</tr>
<tr>
<td>37</td>
<td>18,000</td>
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<td>6</td>
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<tr>
<td>36</td>
<td>13,000</td>
<td>25</td>
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Mean PM2.5 = 11.83 + (0.004*Tvol\textsubscript{300}) + (0.268*Land\textsubscript{300}) – (0.036*Alt)

<table>
<thead>
<tr>
<th>Mean PM2.5</th>
<th>Tvol\textsubscript{300}</th>
<th>Land\textsubscript{300}</th>
<th>Alt</th>
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<td>7</td>
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<td>?</td>
<td>1,000</td>
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<td>?</td>
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Development of GIS variables for land use regression (LUR)

Traffic intensity on the nearest (major) road

Inverse distance to the nearest (major) road

Total traffic load on major roads in a 100m buffer

Indicator of within 50m of motorway

Total length of majors roads within a 100m buffer
# LUR model for PM$_{10}$ (Munich/Augsburg)

<table>
<thead>
<tr>
<th>Variable</th>
<th>Slope</th>
<th>SE</th>
<th>p value</th>
<th>R$^2$</th>
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\[
y = 1.098x - 2.1615 \\
R^2 = 0.9317
\]
Geocoded addresses of study participants in the region Augsburg and PM10 mass concentration

\[
\text{PM}_{10} (x, y) = \\
a \times \text{TRAF LOAD 50} (x, y) + \\
b \times \text{ROAD LENGTH 500} (x, y) + \\
c \times \text{HEAVY TRAF LOAD 300} (x, y) + \\
d \times \text{ROAD LENGTH 25} (x, y) - \\
e \times \text{NATURAL_LVA 100} (x, y) + \\
\text{intercept}
\]
Das Ziel: räumliche Verteilung von Luftschadstoffen in der Studienregion

PM$_{2.5}$ absorbance modelled exposure
Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung

Source apportionment
Spatial and temporal variation

Stadt Augsburg
Region Augsburg
ESCAPE

Deutschland
GUAN

Messstation Augsburg
German Ultrafine Aerosol Network (GUAN)
Anzahl-Größenverteilungen (TDMPS, DMPS, SMPS)

Nichtflüchtige Bestandteile (Thermodenuder)

Absorption (MAAP)

Berner-Impaktor (5)
Time series of soot and PM$_{10}$ concentration (5 hour running average)

Time series of soot concentration in 2009 (24 hour average)
Time series of soot concentration in 2009 (24 hour average)
Summary

• Traffic is the major contributor of particle number concentrations at a background monitoring site in Augsburg

• Stationary combustion and secondary aerosols are the major volume/mass contributors

• The impact of specific particle sources differs for different measurement sites (spatial variation). Better exposure for long-term studies is needed (LUR, GIS)

• Strong temporal correlation for source contribution within the city area of Augsburg was observed

• Characterization of the spatial-temporal variation of ultrafine, fine particles and soot over Germany is ongoing (GUAN)
Messstationen in Städten und auf dem Land
Major current research themes with regard to particle characterization for epidemiological studies

- Long-term health effect studies for other particle measures than PM10 or PM2.5 are needed (spatial variation, GIS and land use regression modeling)
- Short-term health effect studies on ultrafine particles, other particle characteristics and air pollution mixtures are needed (temporal variation)
- Better characterization of organic compounds (spatial and temporal)
- Which particles and sources are responsible for the observed health associations?
- Evaluation of measures to improve air quality (German focus: Low emission zones)
- „Near road studies“ with personal monitoring for better understanding the mechanisms
- Health effects of “hotspot” exposures in the cities
Back up Folien
Exposure characterization relevant for health research: Aerosol measurement site in Augsburg

- Continuous physical and chemical particle characterization for epidemiological studies
- Source apportionment
- Spatial variation of particles (urban background sites, traffic sites)
- Personal exposure assessment
- Development of new measurement techniques
- Organic characterization of samples for toxicological and epidemiological studies
Measurement program (hourly measurements with automated devices)

- **Physical characterization**
  - Particle size distribution (PSD) in the size range from 3 nm to 20 µm (number counts)
  - Non-volatile PSD in the size range 3 – 800 nm (300°C)
  - Particle length 10 nm – 1 µm (EAD)
  - Active particle surface 10 nm – 1 µm (DC)
  - Particle mass <2.5 µm, <10 µm (TEOM)

- **Chemical characterization**
  - sulfate <2.5 µm
  - nitrate <2.5 µm
  - soot (EC) <2.5 µm
  - polycyclic aromatic hydrocarbons (PAHs)
Further parameters

**Length concentration (LC)** for *X* selected size bins *K* with the diameter *d*:

\[
LC_{K=1 \text{ to } (K+X)} = \sum_{K=1}^{K+X} (NC_K \cdot d_K)
\]

**Surface concentration (SC)** for *X* selected size bins *K* with the diameter *d* and assuming spherical shape of the particles:

\[
SC_{K=1 \text{ to } (K+X)} = \sum_{K=1}^{K+X} (NC_K \cdot \pi \cdot d_K^2)
\]

**Mass concentration (MC)** for *X* selected size bins *K* with the diameter *d* and an apparent mean density *ρ*, assuming spherical shape of the particles:

\[
MC_{K=1 \text{ to } (K+X)} = \sum_{K=1}^{K+X} (NC_K \cdot \frac{\pi \cdot d_K^3}{6})
\]
Positive matrix factorization (PMF)
How does PMF work?

• Give the number of sources to the model
• A pair of G and F matrix obtained at each calculation circle (or each Q)
• Calculation continued until a minimum Q was got (hundreds of rotations)
• Give another number of sources
• Choose the right number

\[ Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left( \frac{x_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}} \right)^2 \]

Data uncertainty

residuals

The objective of PMF is finding out G and F at minimum Q value

- **NaCl, de-icing agent**: 6.7% PM10
- **Secondary sulfate**: 13.0% PM10
- **Combustion**: 13.3% PM10
- **Secondary nitrate**: 30.5% PM10
- **Traffic emission**: 16.5% PM10
- **Re-suspended dust**: 20.0% PM10
Comparison between PCC and PSD method

Factor temporal comparison between PCC and PSD method

- Chemical composition data
- Size distribution data
- Aged traffic/ Traffic emission
  - Great influence at KP
  - Regional/ Long range transported

- Stationary combustion/ Residential & commercial heating
  - $r = 0.72$

- Re-suspended dust
  - $r = 0.62$

- Nitrate & sulfate / Secondary aerosols
  - $r = 0.92$

Date range:
TRAPCA Modell – Verteilung der Messstandorte (TRAPCA 1999/2000)
### ESCAPE – Entwicklung von LUR

\[
\text{Mean PM2.5} = 11.83 + (0.004 \times \text{Tvol}_{300}) + (0.268 \times \text{Land}_{300}) - (0.036 \times \text{Alt})
\]

<table>
<thead>
<tr>
<th>Mean PM2.5</th>
<th>Tvol$_{300}$</th>
<th>Land$_{300}$</th>
<th>Alt</th>
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</table>
BS$_{2.5}$ Konzentrationen für 1757 TRAPCA Probanden in München

PM$_{2.5}$ absorbance modelled exposure
Ziele

• Aufbau, Wartung und Betrieb der Messtechnik zur Erfassung des Ultrafeinstaubes

• Charakterisierung des raum-zeitlichen Vorkommens feiner, ultrafeiner und rüschhaltiger Partikel in der Außenluft in Deutschland über einen Zeitraum von 3 Jahren

• Bewertung der erhobenen Parameter im Hinblick auf ihre Eignung für epidemiologische Studien

• Diskussion der Repräsentativität der Messungen im Messnetz in Bezug auf die Bevölkerung im Einzugsbereich

• Ermittlung des Forschungsbedarfs in Deutschland vor dem Hintergrund der Messungen, des Wissensstand in 2010 und auf internationaler Ebene laufender Forschungsaktivitäten