Characterization of particulate matter in urban and rural area

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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).

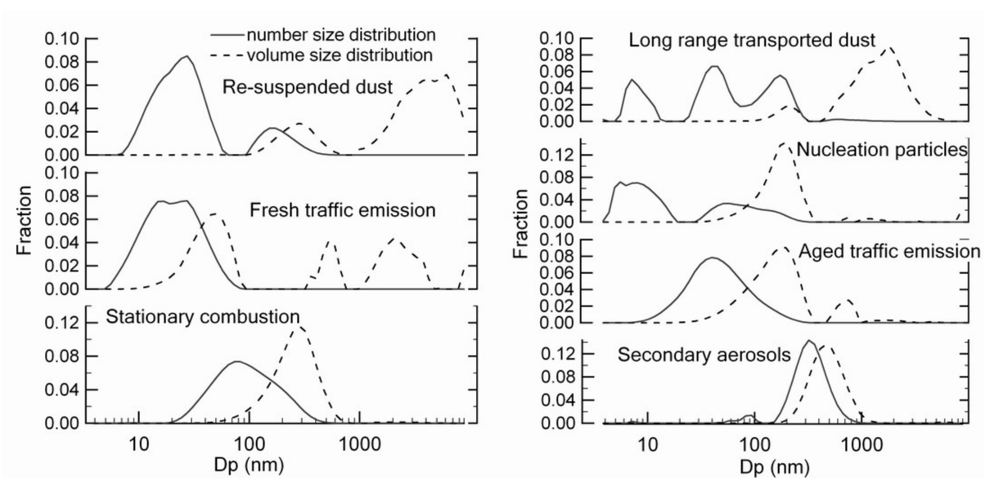

Figure 1: Number and volume size distribution of seven factor identified by PMF in Augsburg in winter 2006/07

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

(NC). 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 homogonously 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%).

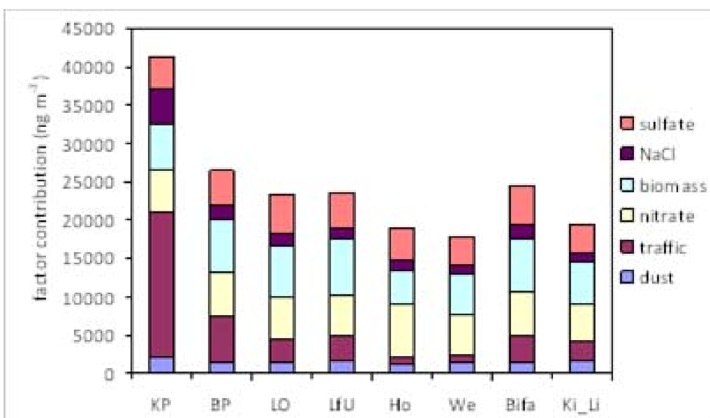

Figure 2: Mean PM_{10} mass concentrations measured at eight different monitoring sites in Augsburg and the impact of different particle sources to the PM mass concentration (KP - traffic site, BP - traffic influenced urban background site, LO, LfU, Bifa & Ki_Li - urban background sites, We & Ho - Rural/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

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.

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Charakterisierung von Feinstaub-partikeln in städtischer und ländlicher Umgebung

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14th ETH Conference on Combustion Generated Nanoparticles

Focus-Event 2010 (1): Feinstaub in Städten und ländlichen Raum, Zürich, 04.08.2010

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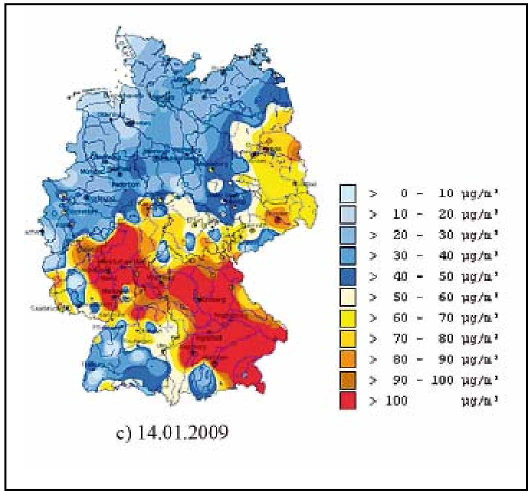
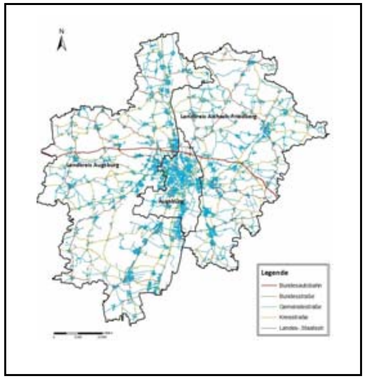
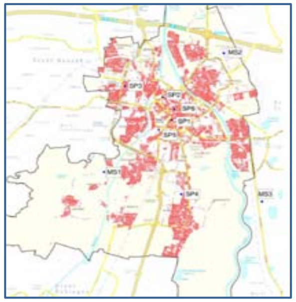
Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung



Deutschland



Region Augsburg



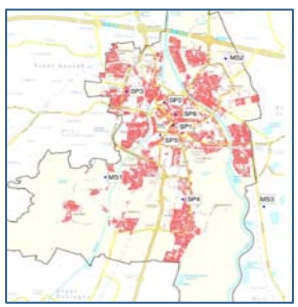
Stadt Augsburg

Messstation Augsburg

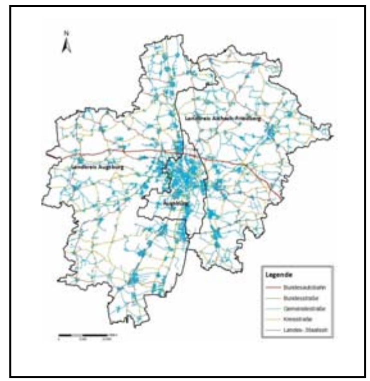
Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung

Source apportionment
Spatial and temporal variation

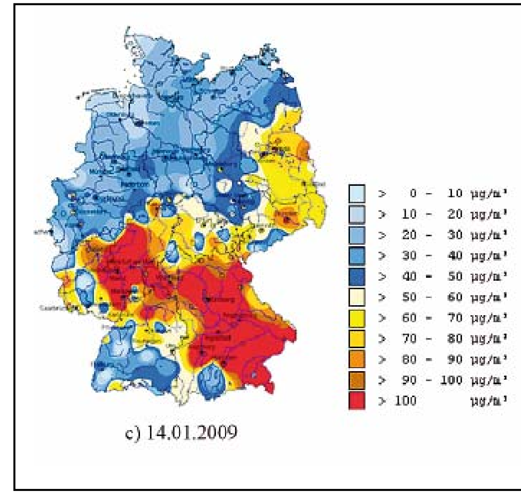
Messstation Augsburg



Stadt Augsburg

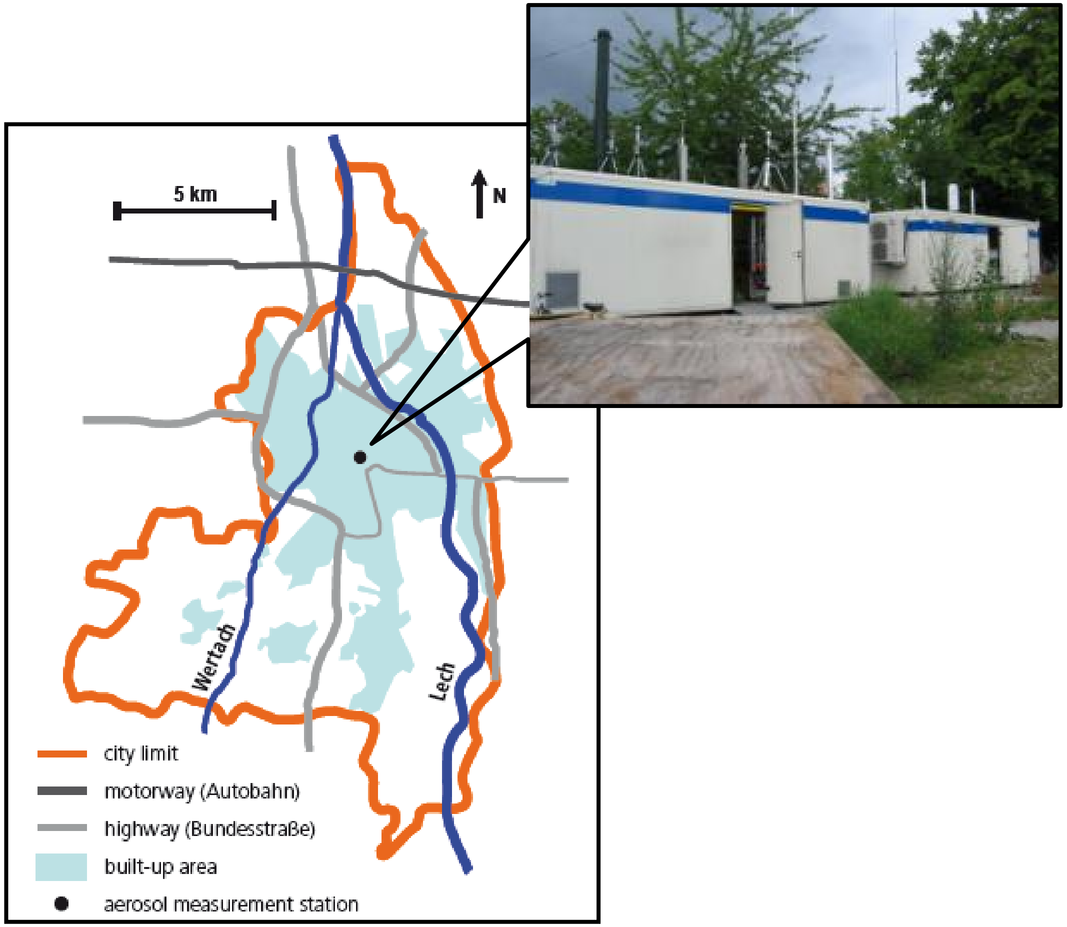


Region Augsburg

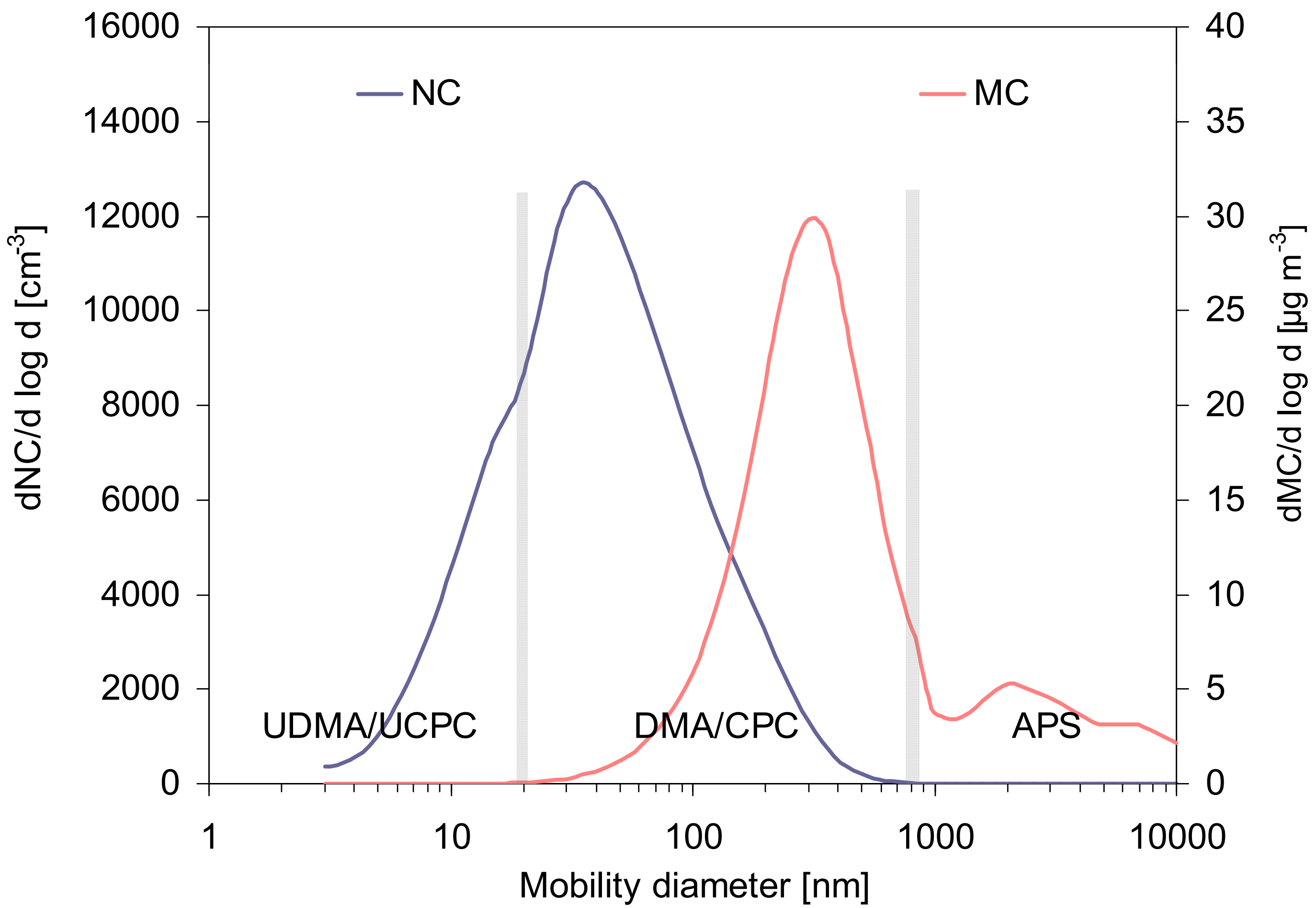


Deutschland

Exposure characterization relevant for health research: Aerosol measurement site in Augsburg



Mean ambient particle size distribution Augsburg
14th Mar 07 - 17th Dec 08



	Ambient NC
3	376,1993468
3,392	392,2488502
3,836	513,629653
4,337	682,2917866
4,905	977,0967234
5,546	1384,249006
6,271	1877,018347
7,091	2465,490648
8,019	3134,93575
9,067	3868,178995
10,253	4665,310308
11,593	5507,623724
13,109	6383,75068
14,824	7213,004082
16,762	7750,722139
18,954	8249,328276
21,433	9199,375833
24,235	10442,15199
27,404	11721,60382
30,988	12433,34916
35,04	12697,19492
39,622	12563,44367
44,803	12121,09967
50,662	11516,64825
57,286	10801,06606
64,777	10029,11403
73,248	9200,7831
82,826	8330,925908
93,657	7476,710233
105,903	6656,925364
119,752	5892,72697
135,411	5192,668867
153,118	4538,393931
173,14	3903,506804
195,781	3280,126616
221,382	2665,411451
250,331	2079,182909
283,065	1544,470744
320,08	1087,501143
361,935	725,1153218
409,263	453,0055423
462,78	267,3780821
523,295	156,032137
591,723	86,25724316
669,1	47,4328031
756,594	25,71571085
855,529	13,51752966
967,402	5,249243279
1093,903	3,16233623
1236,947	2,161161597
1398,695	1,652325035
1581,594	1,317591515
1788,41	1,030822052
2022,27	0,759677617
2286,711	0,515322015
2585,73	0,338390661
2923,851	0,218281137
3306,186	0,139676143
3738,517	0,088311777
4227,381	0,055148118
4780,171	0,034604846
5405,246	0,023536275
6112,058	0,016570818
6911,297	0,011236871
7815,047	0,007138543
8836,975	0,004371879
9992,534	0,002576711

Positive matrix factorization (PMF)
How does PMF work?

Positive matrix factorization (PMF)

m elements

n samples

	1	2	3............	i	.... m
1	X11	X21	X31	Xi1	.... Xm1
2	X12	X22	X32	Xi2	.... Xm2
3	X13	X23	X33	Xi3	.... Xm3
.	.	.	.	.	....
.	.	.	.	.	....
j	X1j	X2j	X3j........	**Xij**	.... Xmj
.		.	.	.	....
.		.	.	.	....
n	X1n	X2n	X3n	Xin	.... Xmn

Particle data sample

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

$$X = GF + E$$

m elements

f sources

	1	2	3............	i	.... m
1	F11	F21	F31	Fi1	.... Fm1
2	F12	F22	F32	Fi2	.... Fm2
3	F13	F23	F33	Fi3	.... Fm3
.	.	.	.	.	....
.	.	.	.	.	....
k	F1k	F2k	F3k	**Fik**	.... Fmk
.		.	.	.	....
.		.	.	.	....
f	F1f	F2f	F3f	Fif	.... Fmf

Source profiles

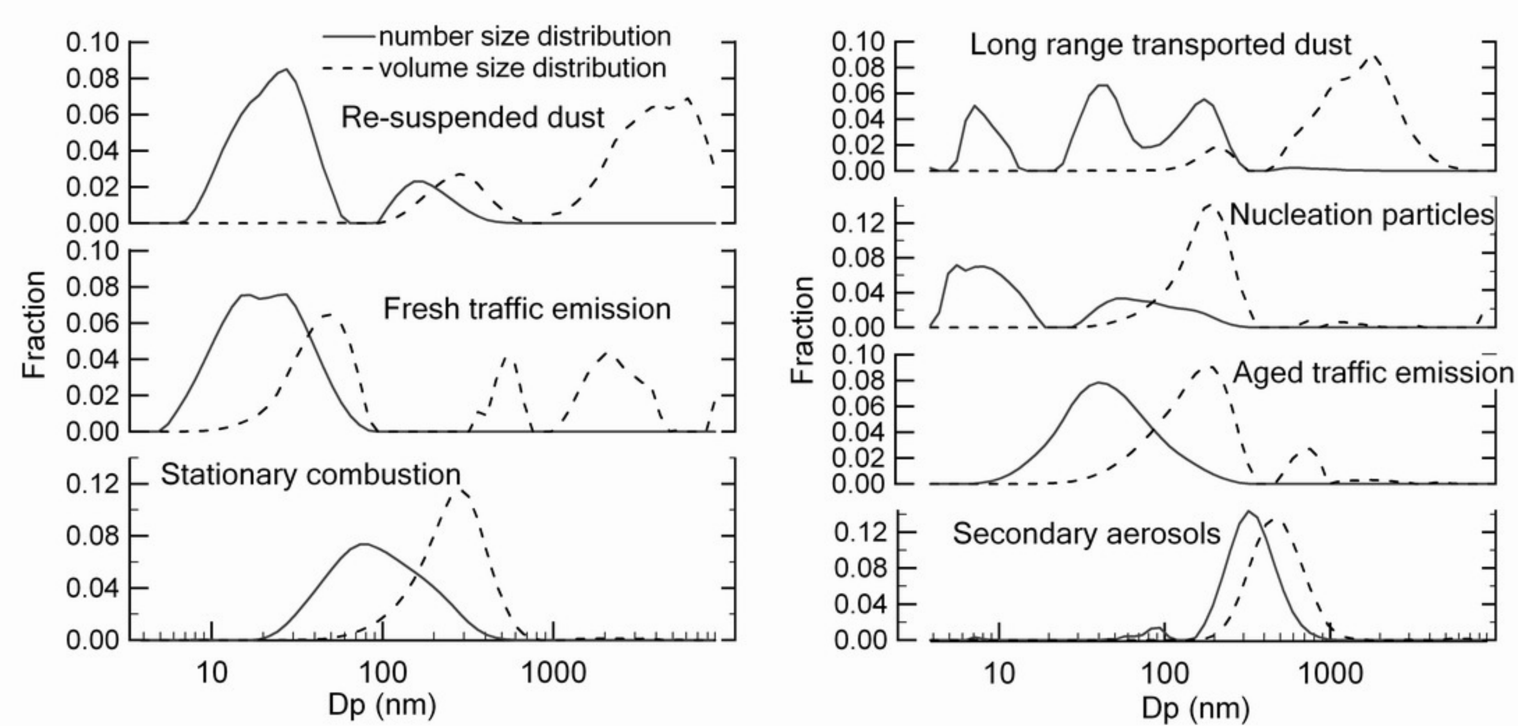
f sources

n samples

	1	2	3........	k......	f
1	G11	G21	G31....	Gk1	Gf1
2	G12	G22	G32....	Gk2	Gf2
3	G13	G23	G33....	Gk3	Gf3
.	.	.	.	.	.
.	.	.	.	.	.
j	G1j	G2j	G3j....	**Gkj**	Gfj
.		.	.	.	.
.		.	.	.	.
n	G1n	G2n	G3n....	Gkn	Gfn

Source contributions

Source Apportionment using PSD data at FH site (Dec 21st,2006 – Mar 23rd, 2007)

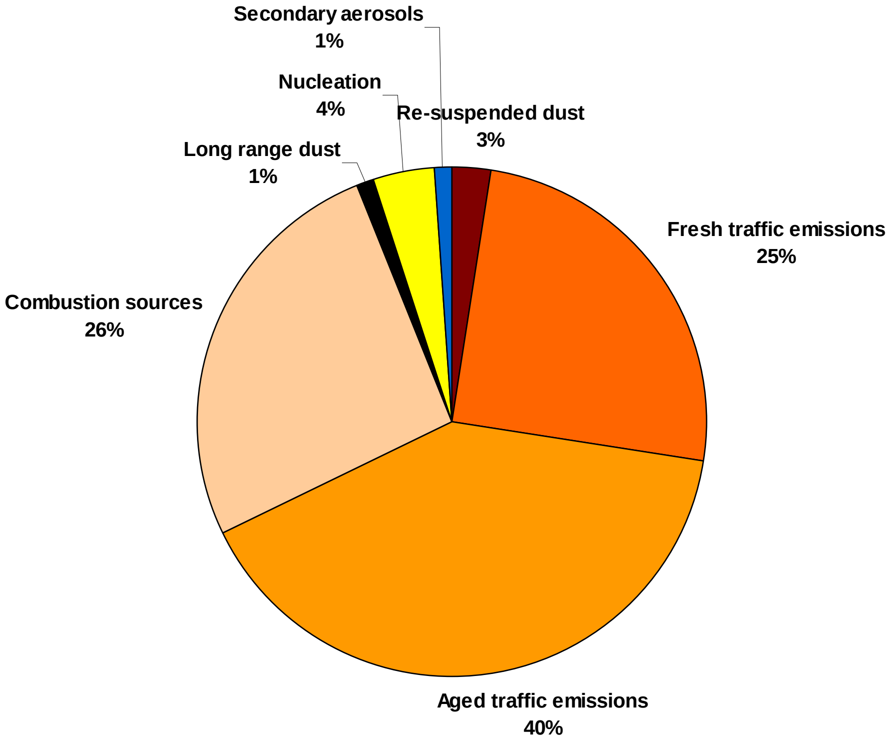


Seven factors were resolved: Re-suspended dust, fresh traffic emission, aged traffic emission, stationary combustion, nucleation particles, secondary aerosols and long range transported dust.

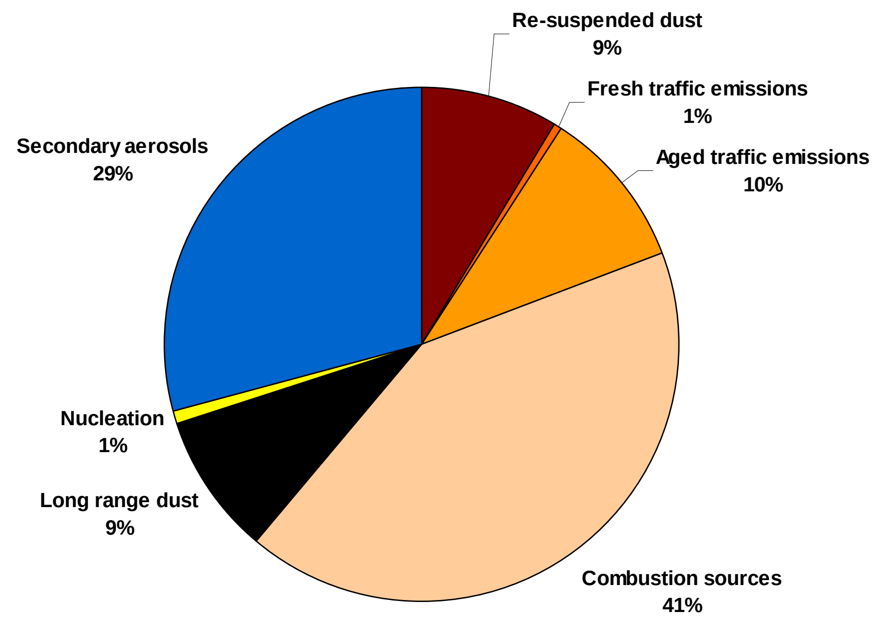
Source Apportionment Using PSD data at FH site (Dec 21st,2006 – Mar 23rd, 2007)

Contribution to

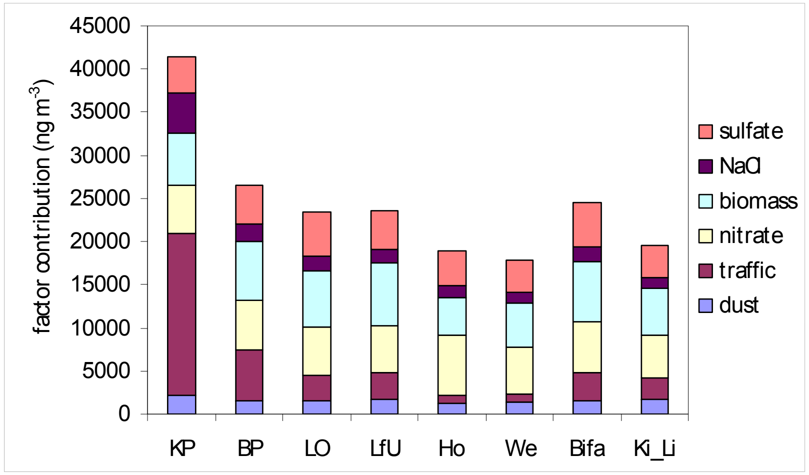
particle number

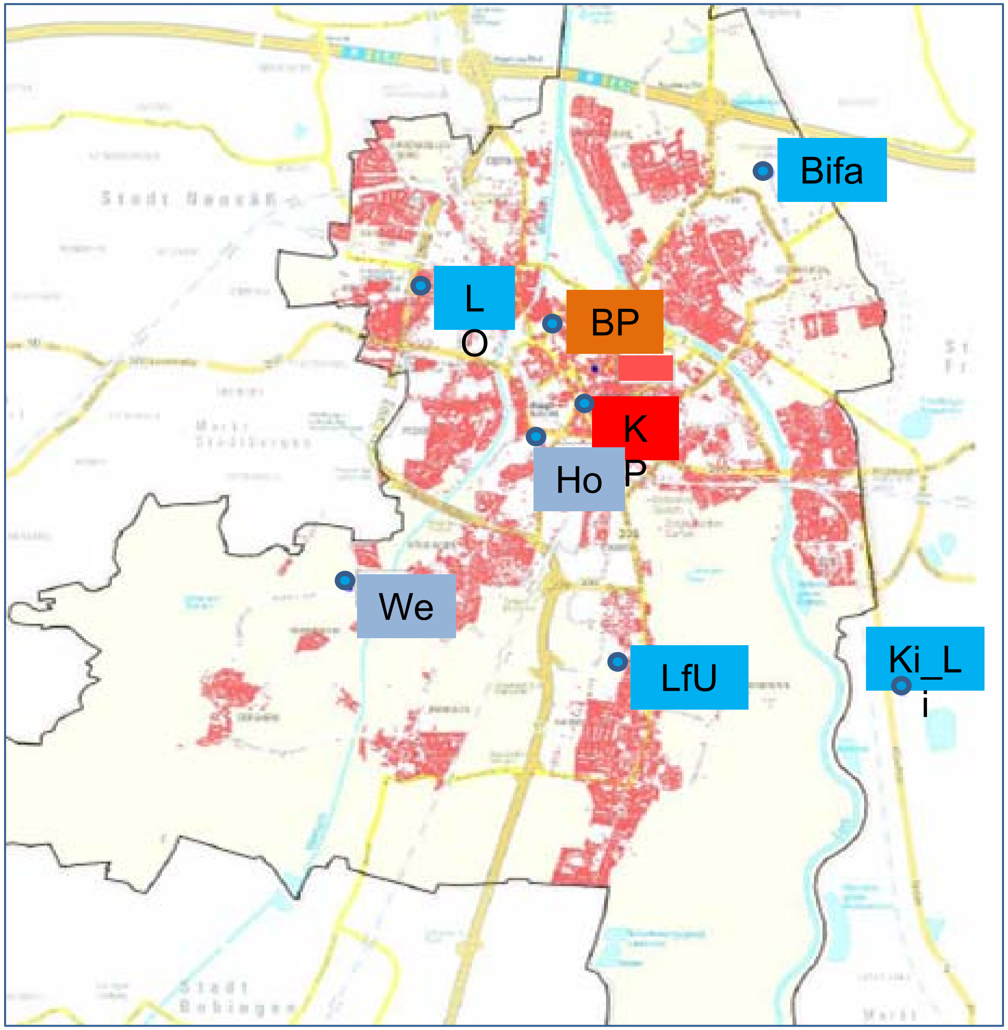


particle mass



Spatial Variation of Particle Sources in Augsburg (PM_{10}) (winter 2007/08)



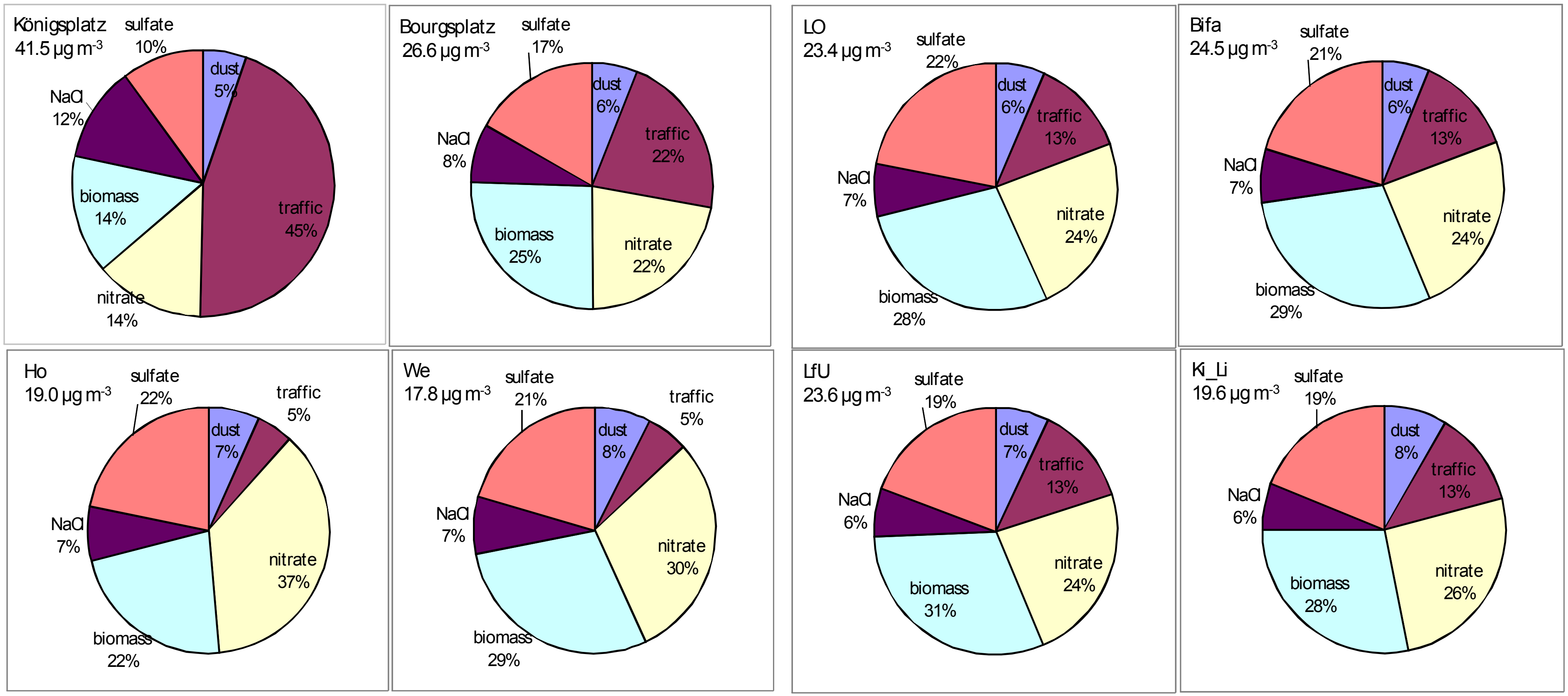


Four site categories

Traffic site: KP
Traffic influenced urban background: BP
Urban background: LO, LfU, Bifa & Ki_Li
Rural/ Tower site: We & Ho

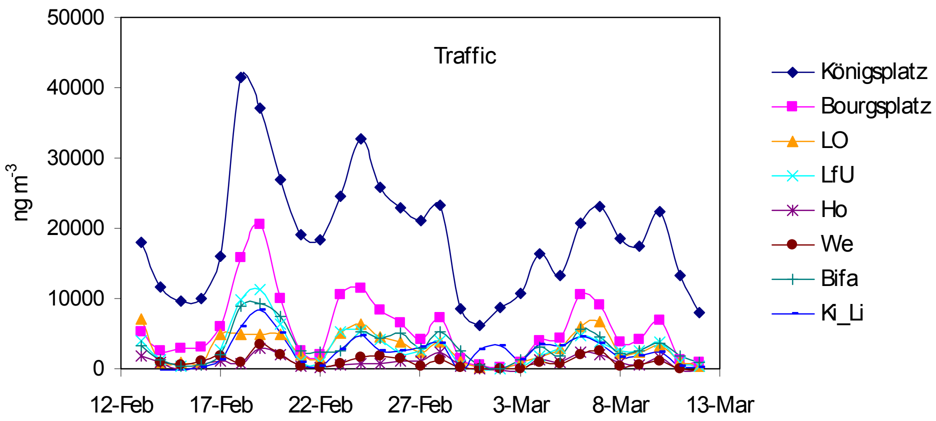
Spatial Variation of Particle Sources in Augsburg (PM_{10}) (winter 2007/08)

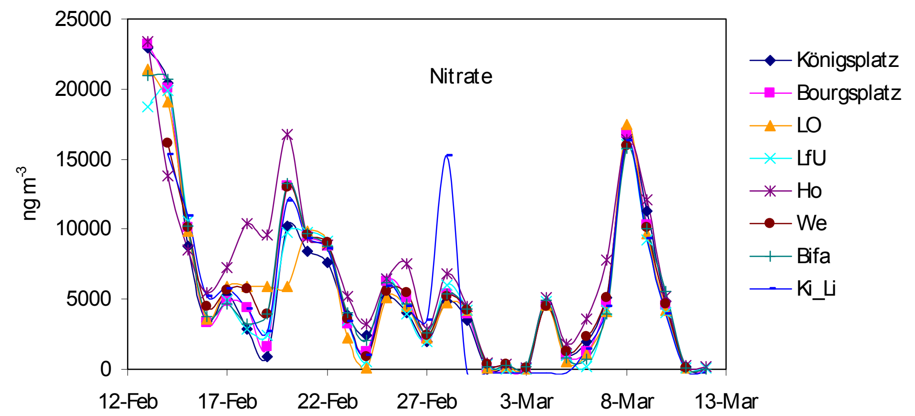
Königsplatz **Bourgsplatz** **Lo, LfU, Bifa & Ki_Li**

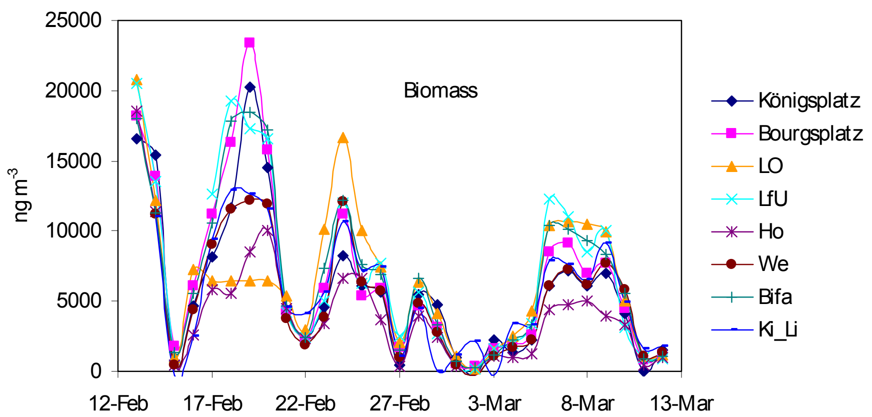


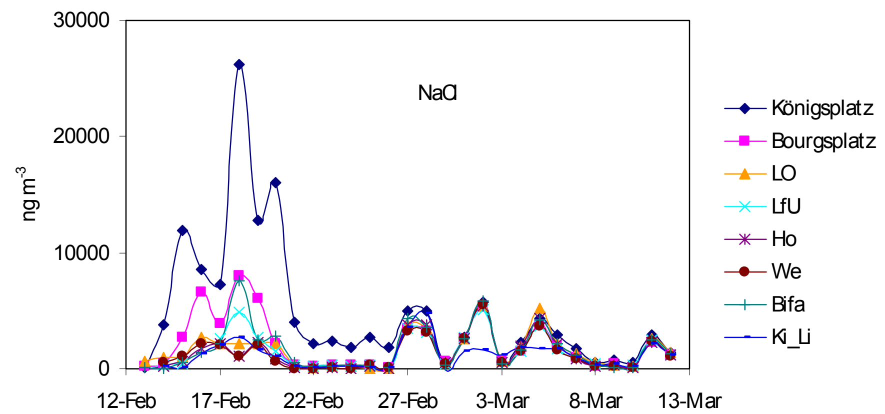
Ho & We

Temporal Variation of Particle Sources in Augsburg (PM_{10}) (winter 2007/08)





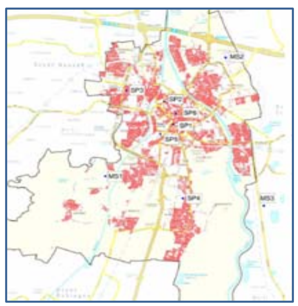




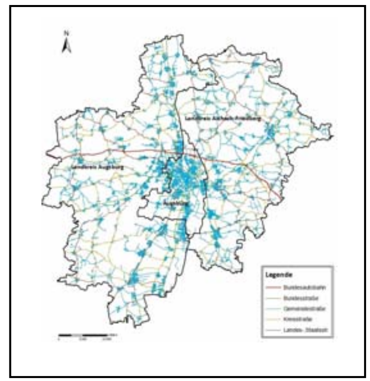
Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung

Source apportionment
Spatial and temporal variation

Messstation Augsburg

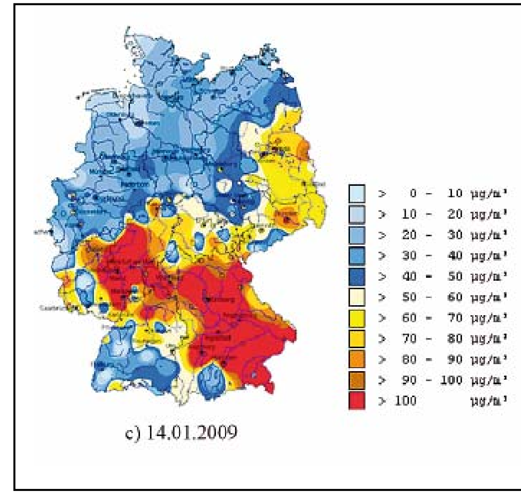


Stadt Augsburg



Region Augsburg

ESCAPE



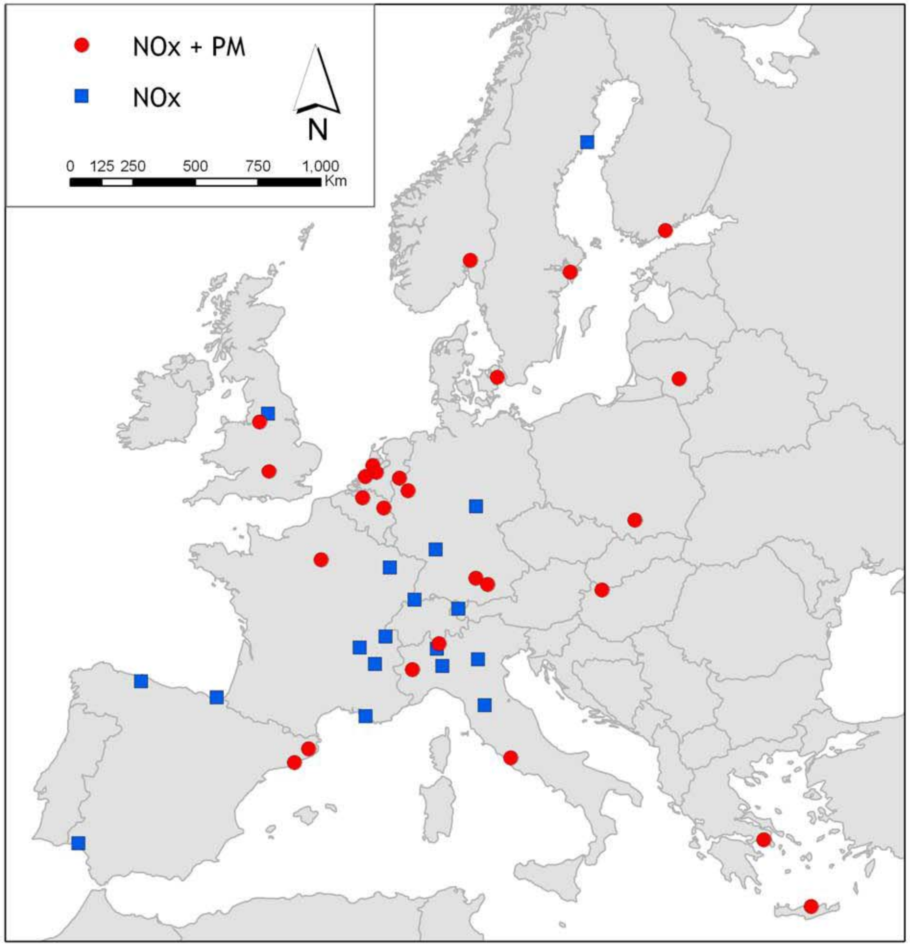
Deutschland

E S C A P E

*E*uropean *S*tudy of *C*ohorts for *A*ir *P*ollution *E*ffects

http://www.escapeproject.eu/index.php

ESCAPE Locations



Exposures modelled for Augsburg study area based on measurements within ESCAPE

$PM_{2.5}$

PM_{10}

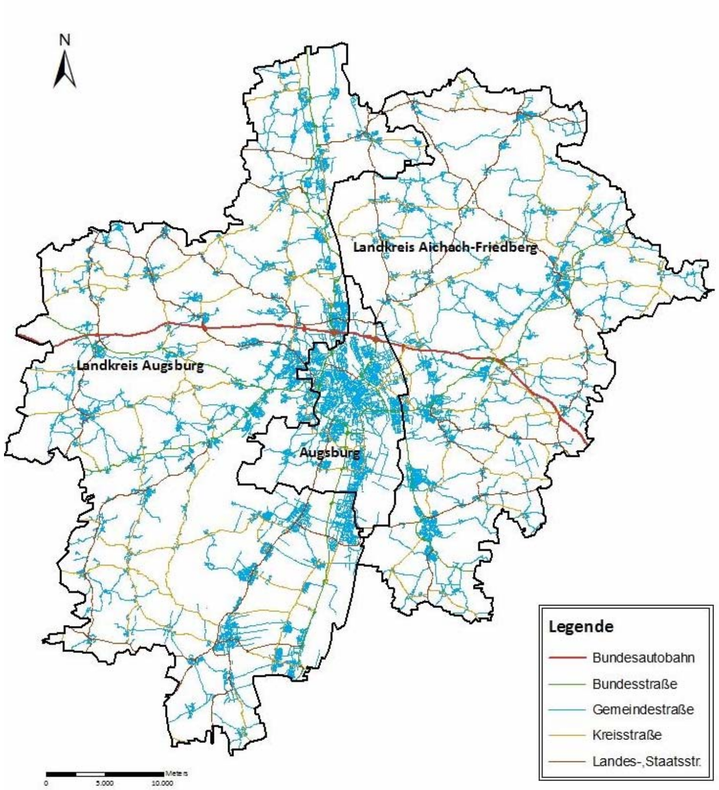
PM_{coarse} (PM_{10}-$PM_{2.5}$)

Absorption coefficient $PM_{2.5}$

NO_x and NO_2

"Source-specific" elements from XRF

Background versus Traffic exposures



Annual averages for NO_2 and PM_{10} in Munich/Augsburg

Site	Type	GPS LAT	GPS LON	NO2 adj	NO2 unadj	PM10 una	PM10 adj
1	UB	4,814,036	1,161,832	26.43	28.01	19.05	20.98
2	UB	4,816,254	116,398	21.52	22.15		
3	S	4,814,999	1,143,745	26.87	28.45	18.54	21.56
4	S	4,813,888	1,150,266	29.93	31.50		
5	S	4,812,568	1,159,534	28.27	29.84	17.70	20.72
6	RB	4,818,853	1,186,533	17.99	18.62		
7	UB	481,173	1,157,799	32.31	32.26	20.64	23.37
8	UB	4,820,612	1,154,956	21.68	22.32		
9	RB	4,817,591	112,586	17.18	18.75	16.88	18.81
10	S	4,816,401	1,148,543	30.67	31.30		
11	UB	4,815,565	1,156,749	27.82	31.02	20.90	19.38
12	S	4,809,209	11,503	24.20	26.72		
13	S	4,813,033	1,155,591	48.56	51.07	31.32	29.89
14	UB	4,813,859	1,147,038	18.33	21.31		
15	S	481,269	1,162,969	28.53	31.05	21.78	20.35
16	UB	4,811,765	1,165,369	23.65	26.64		
17	UB	4,809,137	1,163,817	16.53	19.51	20.35	15.67
18	S	4,815,106	1,161,094	32.32	34.83		
19	S	4,825,886	1,142,349	23.79	26.30	22.30	20.88
20	RB	4,831,587	1,165,991	26.74	29.26		
21	RB	4,832,603	1,090,303	17.83	21.30	17.85	18.41
22	RB	4,837,633	1,085,353	22.27	27.06		
23	S	4,836,463	1,089,498	50.27	52.84	32.96	33.52
24	S	4,838,049	1,086,172	29.64	33.66		
25	UB	4,837,661	1,088,835	28.43	32.50	21.22	21.79
26	UB	4,837,759	1,091,989	19.93	25.25		
27	S	4,811,019	1,152,512	53.46	59.44	23.42	23.99
28	UB	4,816,872	1,157,135	21.74	27.79		
29	RB	4,830,591	1,189,897	20.35	22.10	21.25	21.81
30	RB	4,787,617	1,169,704	16.18	18.82		
31	S	4,858,485	1,108,859	19.69	17.35	19.03	22.65
32	S	4,845,721	1,113,133	31.72	29.38		
33	RB	4,858,231	1,108,641	13.59	11.25	17.39	21.02
34	RB	4,849,026	1,085,348	17.23	14.89		
35	S	4,816,474	1,158,826	41.35	39.01	23.07	26.69
36	S	4,810,497	115,838	39.29	36.94		
37	UB	4,810,362	1,160,893	25.69	23.35	16.65	20.27
38	S	4,810,601	1,165,944	23.85	21.50		
39	RB	4,808,015	1,196,432	17.12	14.78	15.13	18.75
40	S	4,800,129	1,134,333	39.94	37.60		

ESCAPE – Entwicklung von LUR

Mean PM2.5	$Tvol_{300}$	$Land_{300}$	Alt
40	10,000	45	5
45	20,000	50	10
37	18,000	30	6
36	13,000	25	9

Mean PM2.5 = 11.83 + (0.004*Tvol300) + (0.268*Land300) – (0.036*Alt)

Mean PM2.5	$Tvol_{300}$	$Land_{300}$	Alt
?	11,000	40	7
?	1,000	34	10
?	2,700	25	4

Development of GIS variables for land use regression (LUR)

Traffic intensity on the nearest (major) road

inverse distance to the nearest (major) road

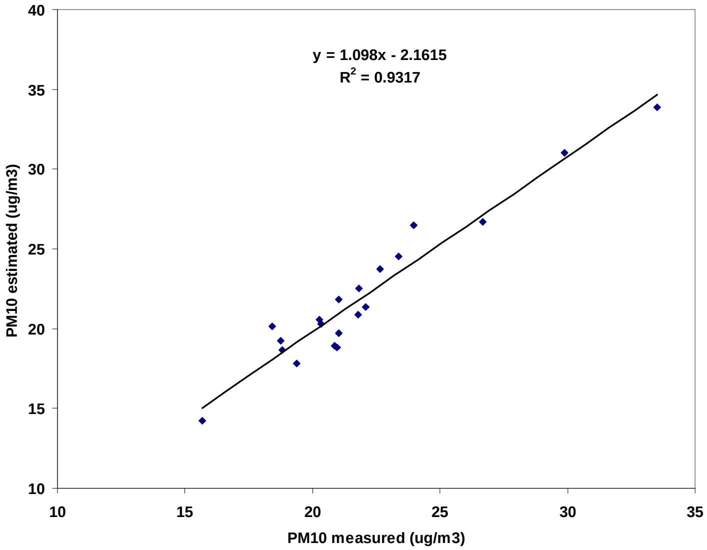
Total traffic load on major roads in a 100m buffer

Indicator of within 50m of motorway

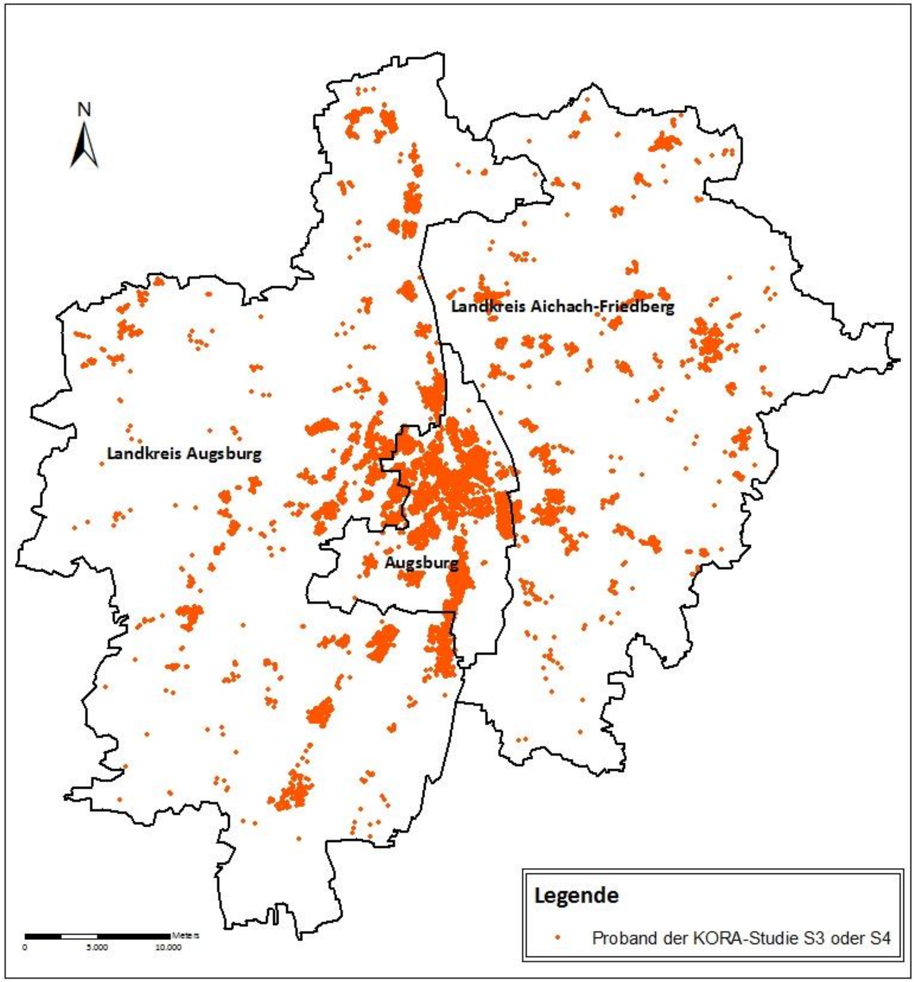
Total length of majors roads within a 100m buffer

LUR model for PM_{10} (Munich/Augsburg)

Variable	Slope	SE	p value	R2
TRAF LOAD 50	7.07E-06 (a)	1.04E-06	<.0001	0.67
ROAD LENGTH 500	4.42E-04 (b)	1.28E-04	0.0039	0.16
HEAVY TRAF LOAD 300	1.27E-06 (c)	3.46E-07	0.0026	0.04
ROAD LENGTH 25	4.05E-02 (d)	1.19E-02	0.0041	0.05
NATURAL_LVA 100 NEG	3.27E+01 (e)	1.76E+01	0.0841	0.02
				0.94



Geocoded addresses of study participants in the region Augsburg and PM10 mass concentration



PM_{10} (x,y) =

a x **TRAF LOAD 50 (x,y)** +

b x **ROAD LENGTH 500 (x,y)** +

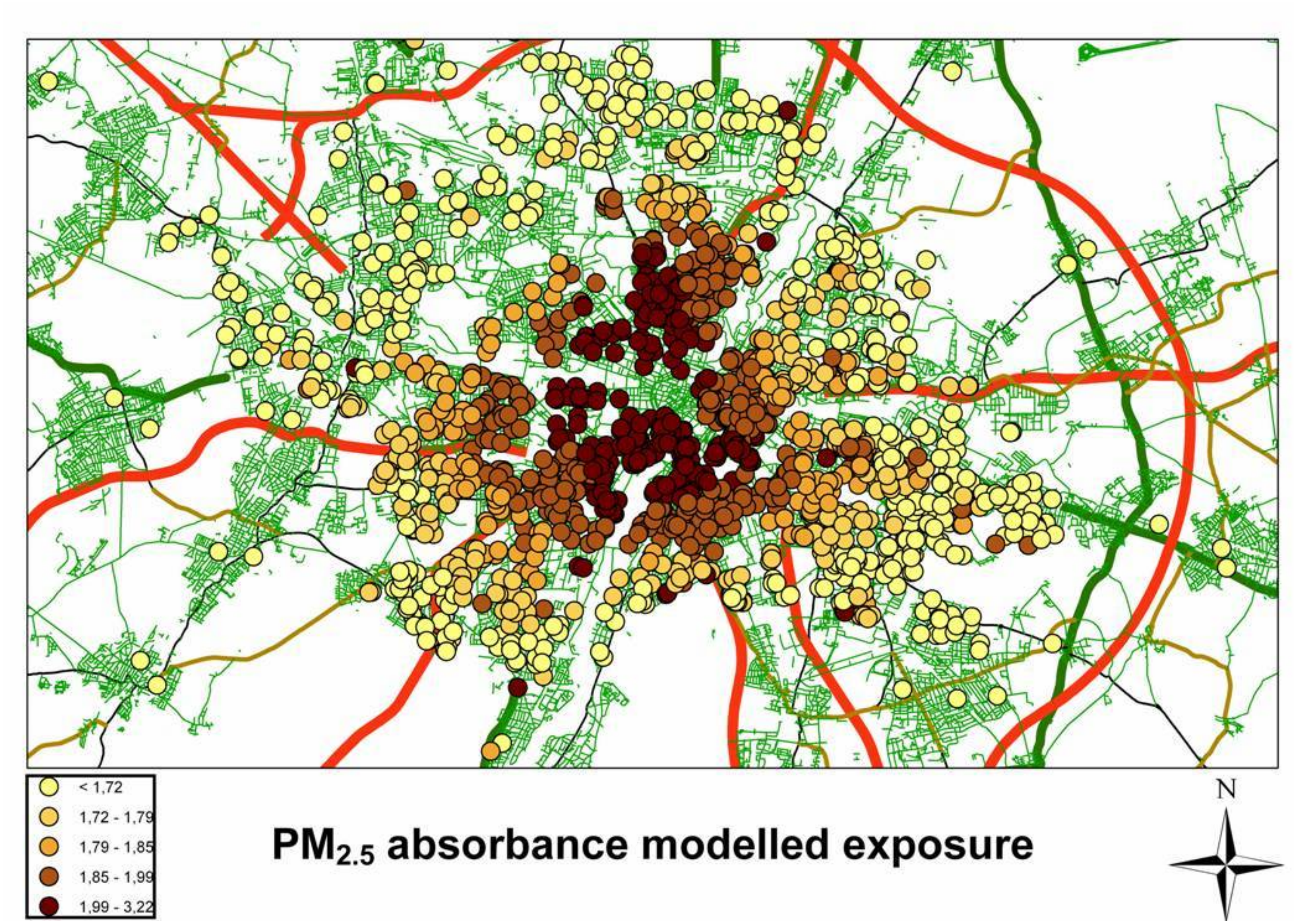
c x **HEAVY TRAF LOAD 300 (x,y)** +

d x **ROAD LENGTH 25 (x,y)** -

e x **NATURAL_LVA 100 (x,y)** +

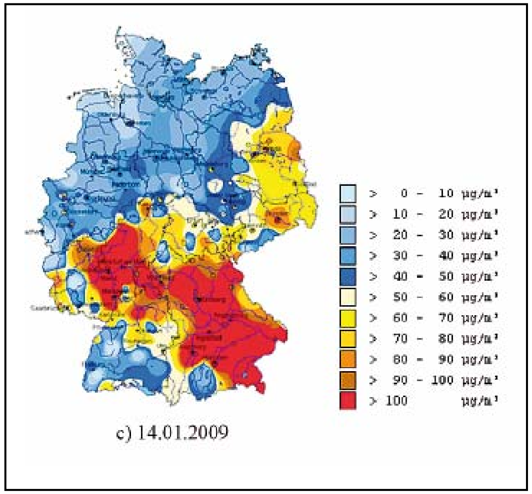
intercept

Das Ziel:
räumliche Verteilung von Luftschadstoffen in der Studienregion



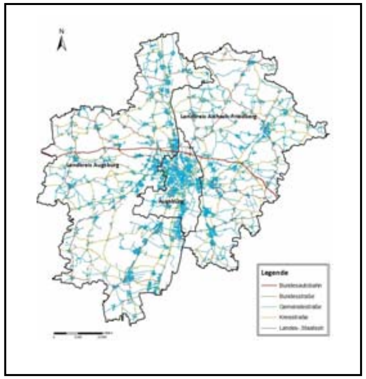
Charakterisierung von Feinstaubpartikeln in städtischer und ländlicher Umgebung

Source apportionment
Spatial and temporal variation



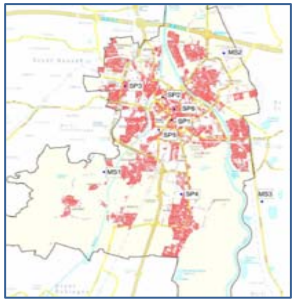
Deutschland

GUAN



Region Augsburg

ESCAPE

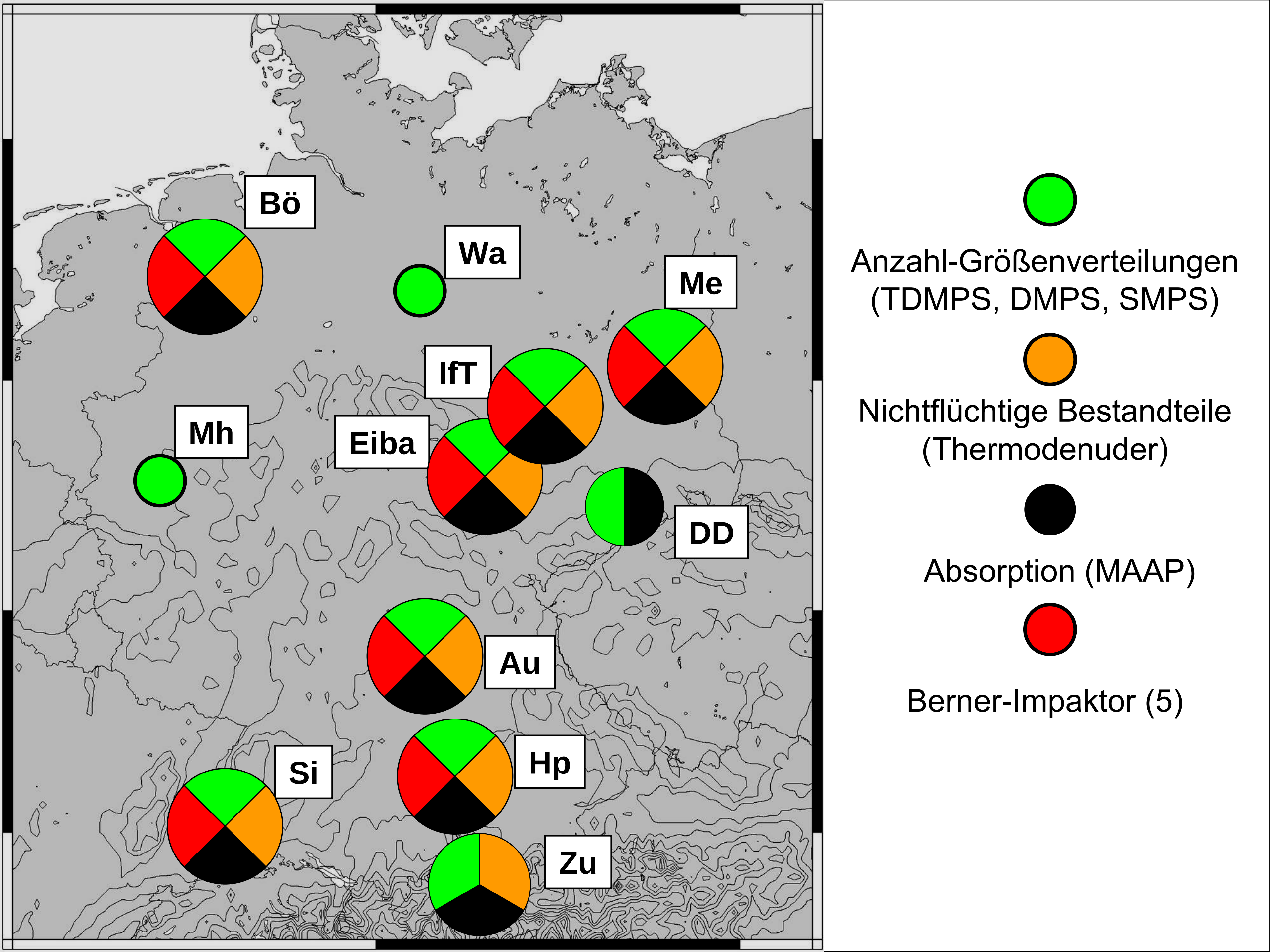


Stadt Augsburg

Messstation Augsburg

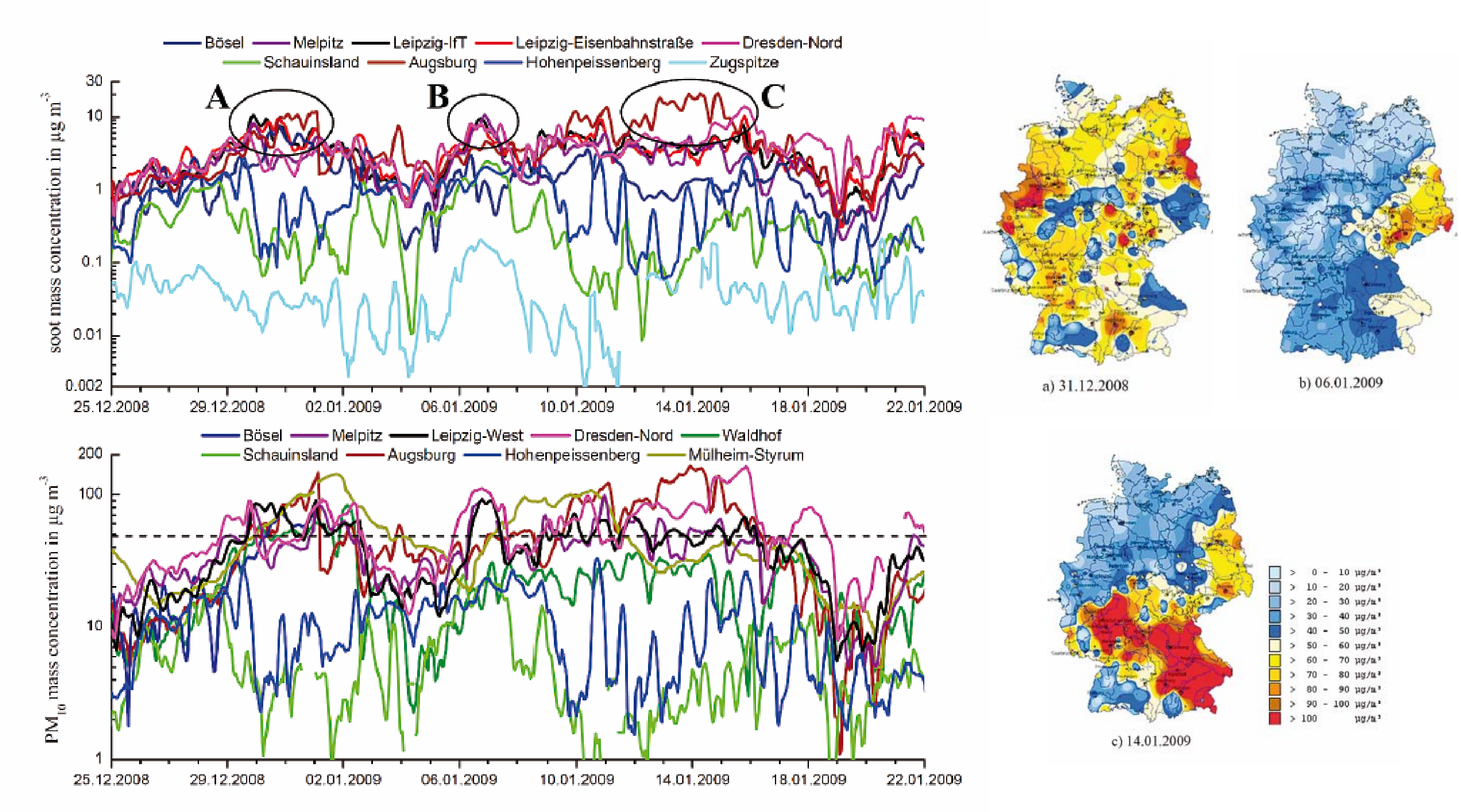
GUAN

German Ultrafine Aerosol Network (GUAN)



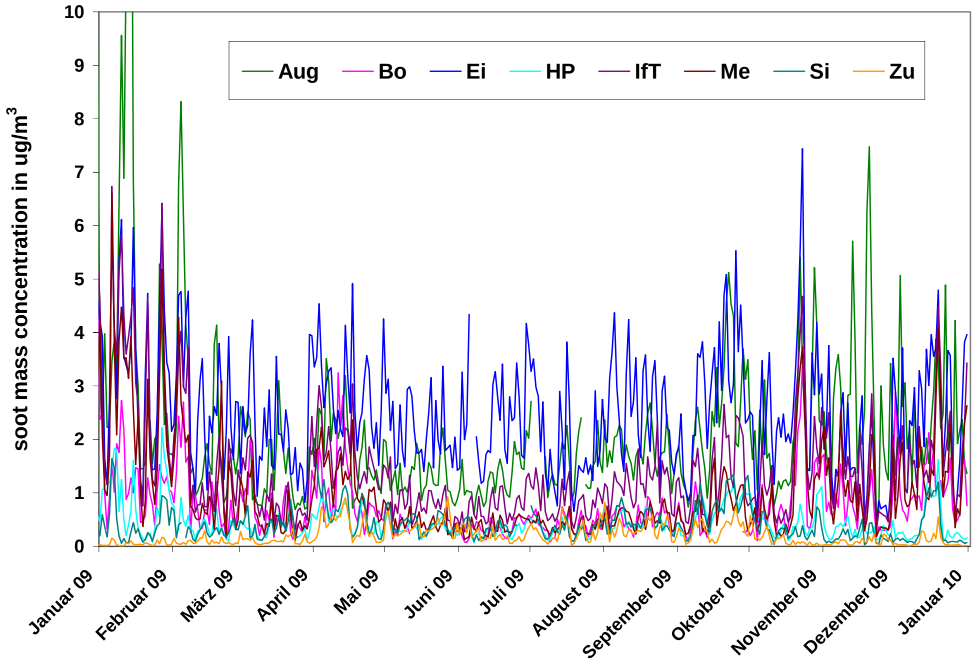
Bö
Wa
Me
IfT
Mh
Eiba
DD
Au
Hp
Si
Zu
Anzahl-Größenverteilungen (TDMPS, DMPS, SMPS)
Nichtflüchtige Bestandteile (Thermodenuder)
Absorption (MAAP)
Berner-Impaktor (5)

Time series of soot and PM_{10} concentration (5 hour running average)

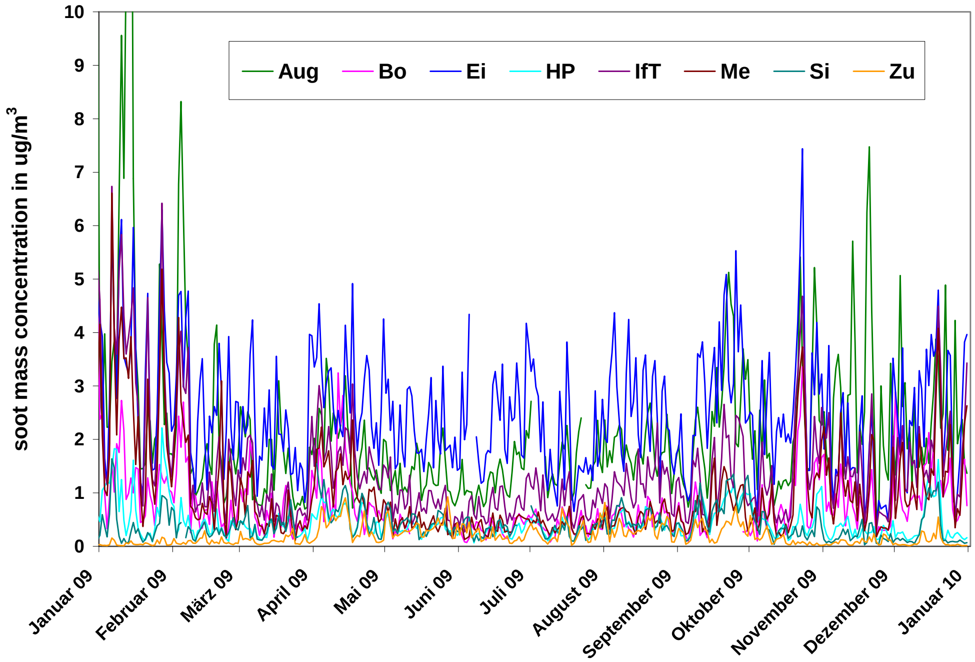


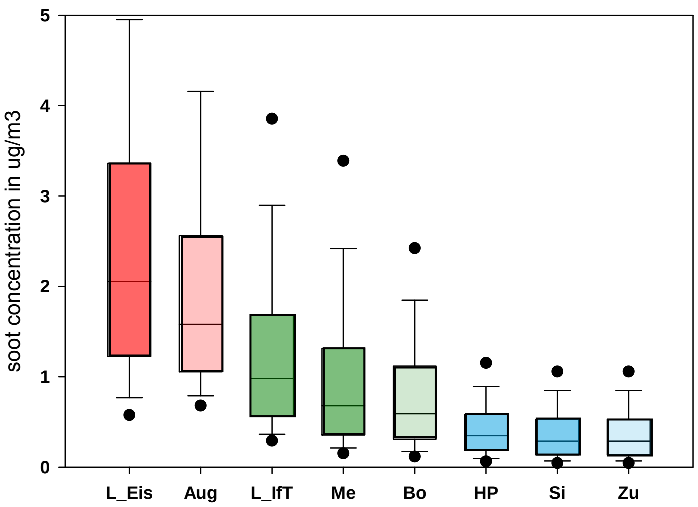
Birmili et al.: Atmospheric aerosol measurements in the German Ultrafine Aerosol Network (GUAN): Part 1 - soot and particle number size distri-but-ions. *Gefahrst. Reinh. Luft*, **69**(4): 137-145, 2009.

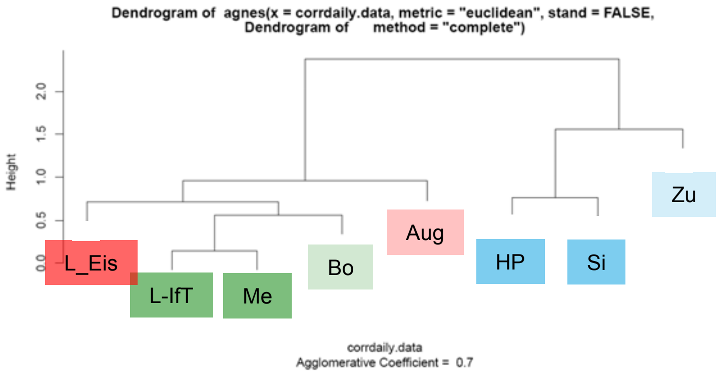
Time series of soot concentration in 2009
(24 hour average)



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







HelmholtzZentrum münchen
German Research Center for Environmental Health

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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



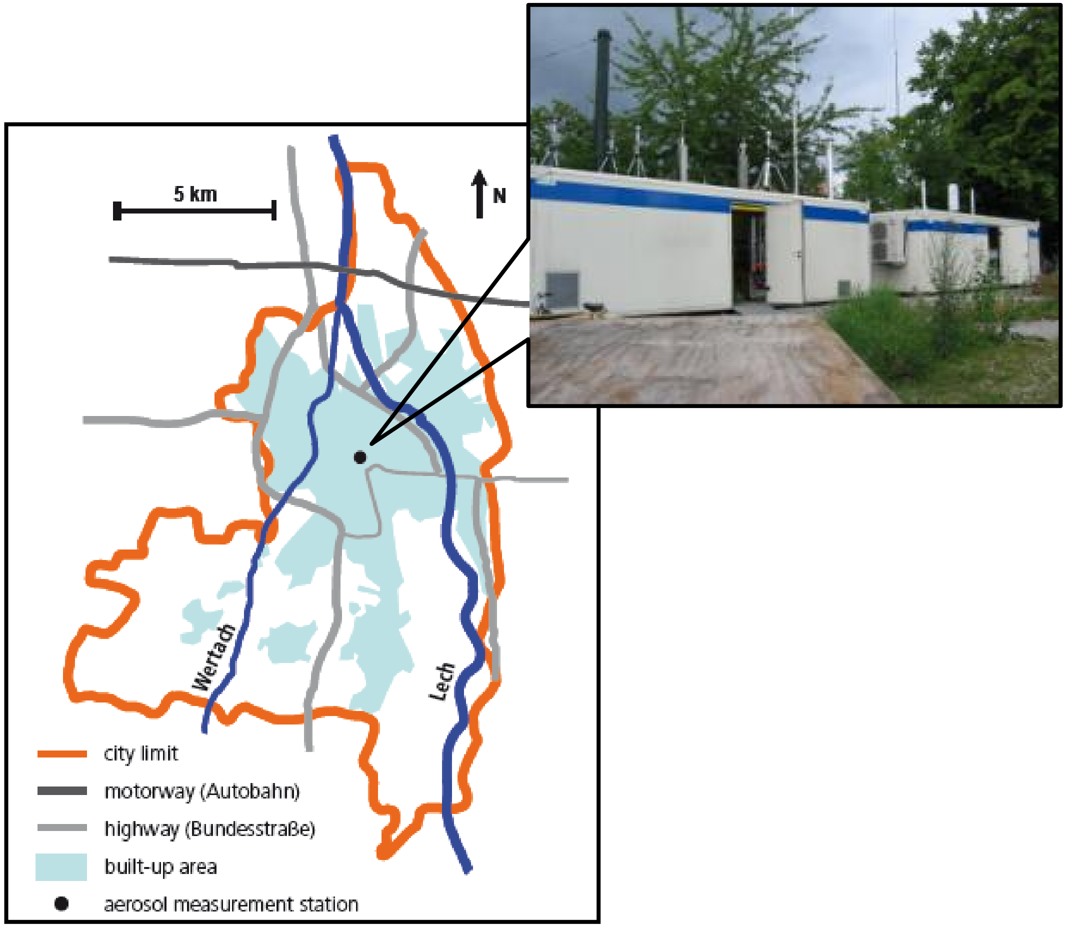

Major current research themes with regard to particle characterization for epidemiological studies

- 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_K:

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

Surface concentration (SC) for X selected size bins K with the diameter d_K, 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_K and an apparent mean density ρ, assuming spherical shape of the particles:

$$MC_{K=1\text{ to }(K+X)} = \sum_{K=1}^{K+X} (NC_K \cdot \frac{\pi \cdot d_K^3}{6} \cdot \rho)$$

Positive matrix factorization (PMF)
How does PMF work?

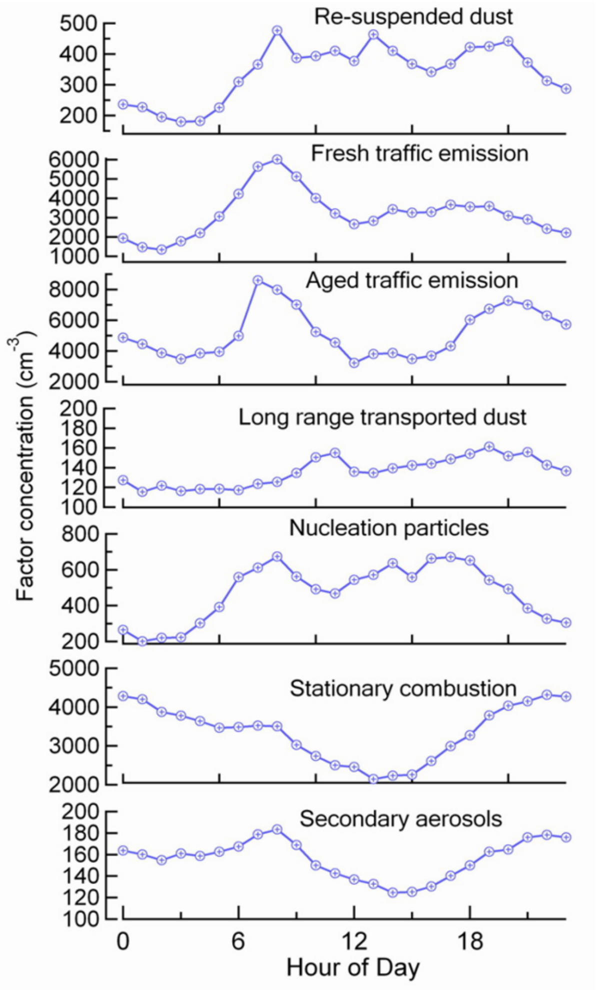
- Give the number of sources to the model
- A pair of G and F matrix obtained at each calculation circle (or each Q)
- Calculation continued until a minimum Q was got (hundreds of rotations)
- Give another number of sources
- Choose the right number

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

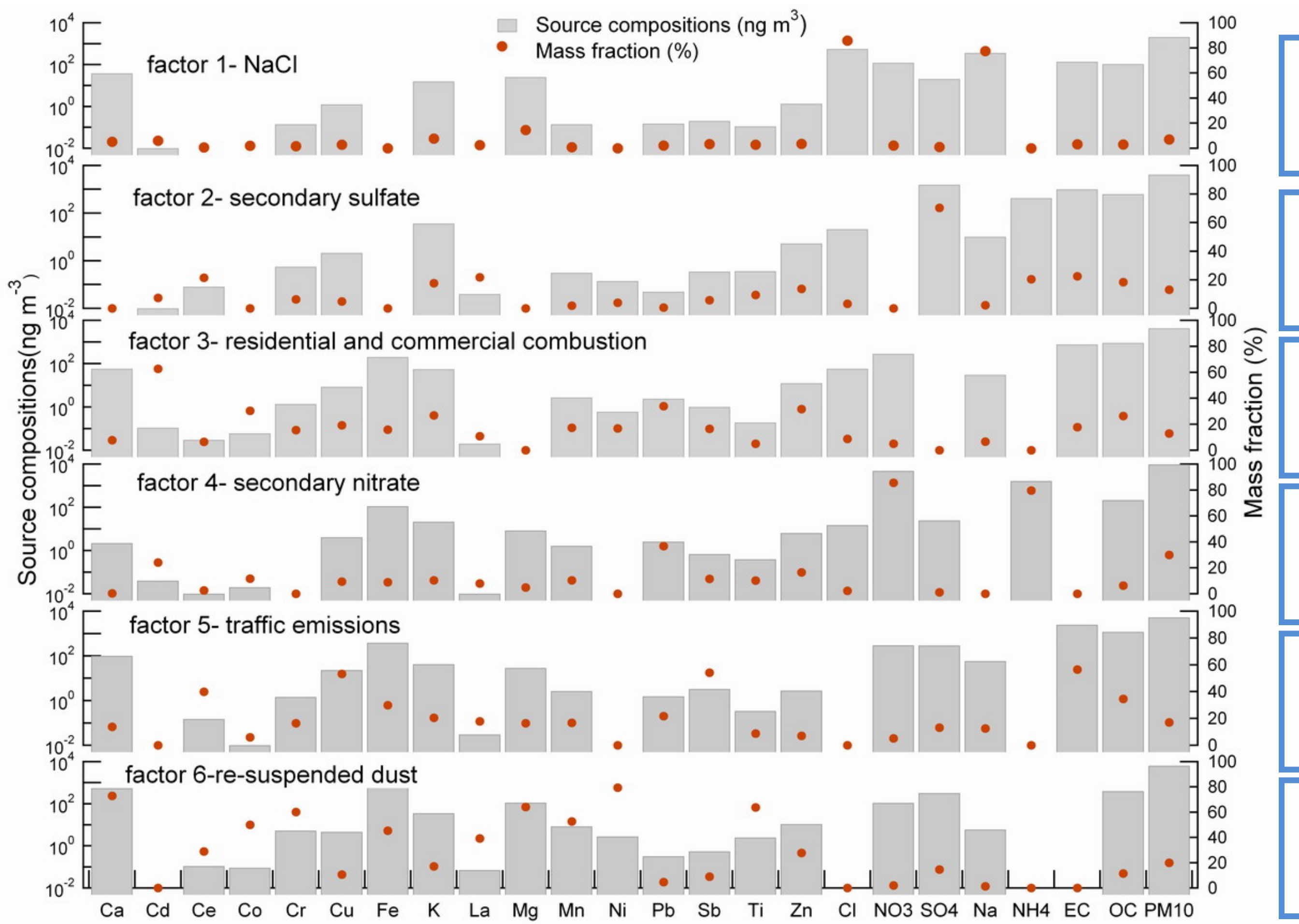
$$\frac{\boxed{\text{residuals}}}{\boxed{\text{Data uncertainty}}}$$

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

Source Apportionment Using PSD data at FH site
(Dec 21st,2006 – Mar 23rd, 2007)



Source Apportionment Using PCC data at KP site
(Dec 21st,2004 – Mar 23rd, 2005)



NaCl, de-icing agent
6.7% PM10

Secondary sulfate
13.0% PM10

Combustion
13.3% PM10

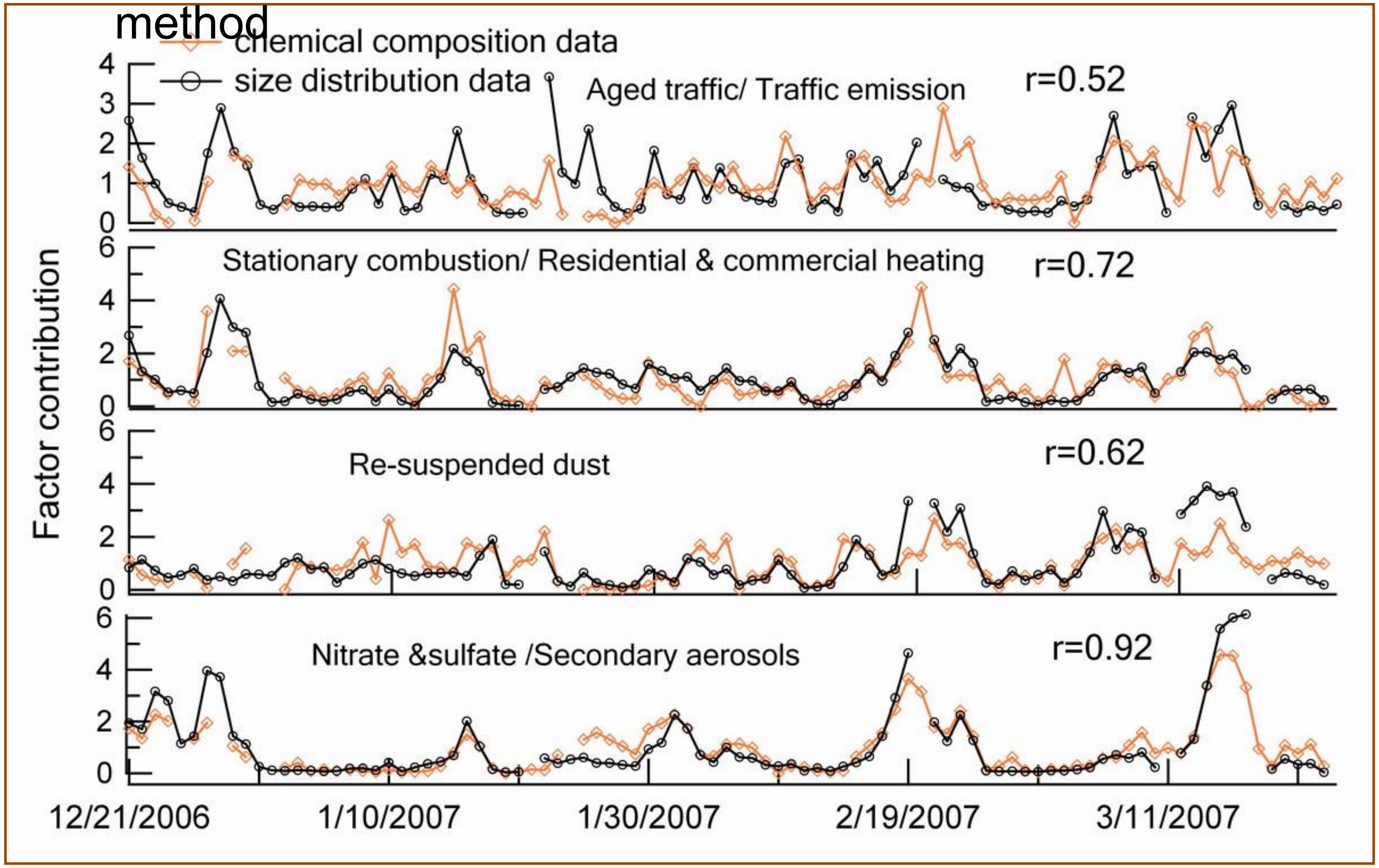
Secondary nitrate
30.5% PM10

Traffic emission
16.5% PM10

Re-suspended dust
20.0% PM10

Comparison between PCC and PSD method

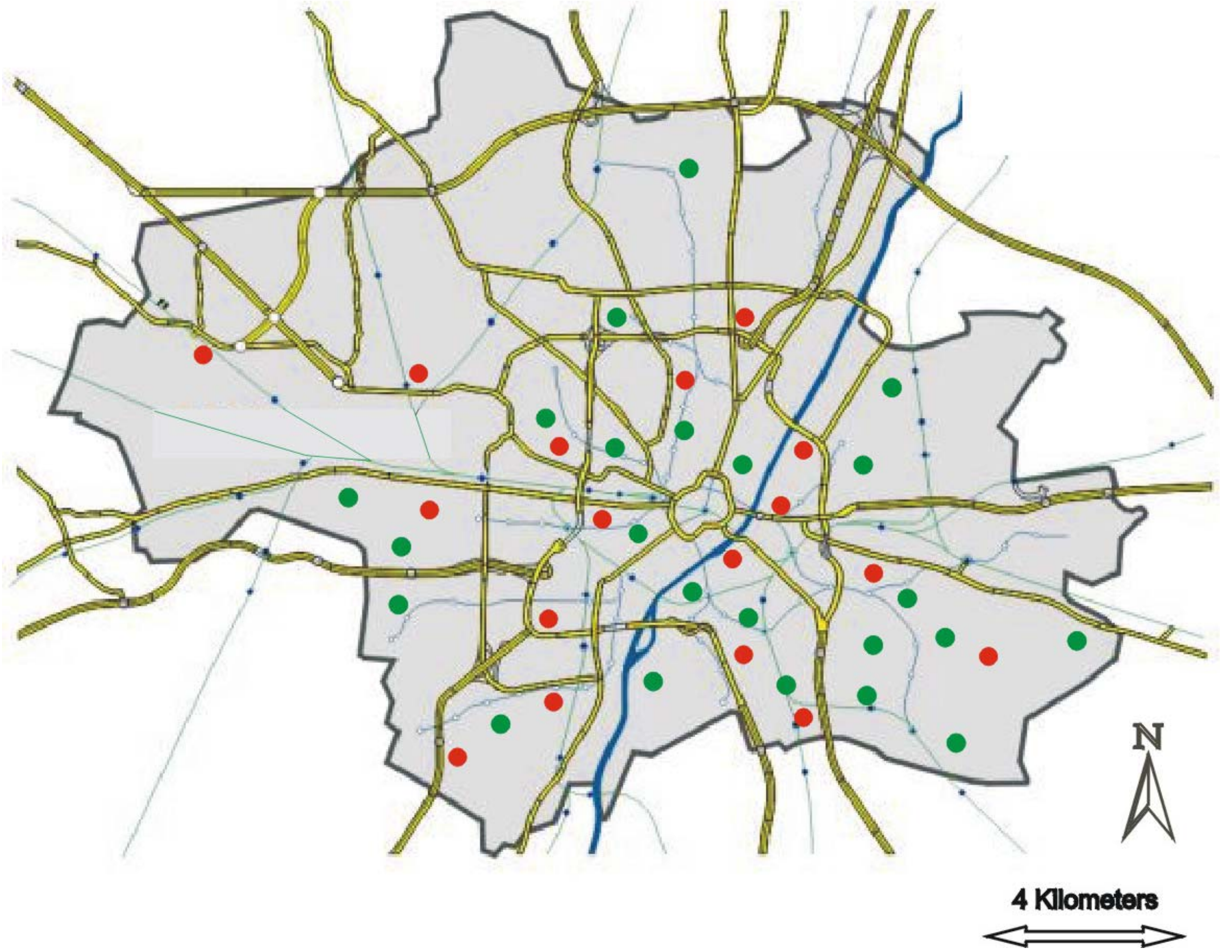
Factor temporal comparison between PCC and PSD method



Great influence at KP

Regional/ Long range transported

TRAPCA Modell – Verteilung der Messstandorte (TRAPCA 1999/2000)



ESCAPE – Entwicklung von LUR

Mean PM2.5	$Tvol_{300}$	$Land_{300}$	Alt
40	10,000	45	5
45	20,000	50	10
37	18,000	30	6
36	13,000	25	9

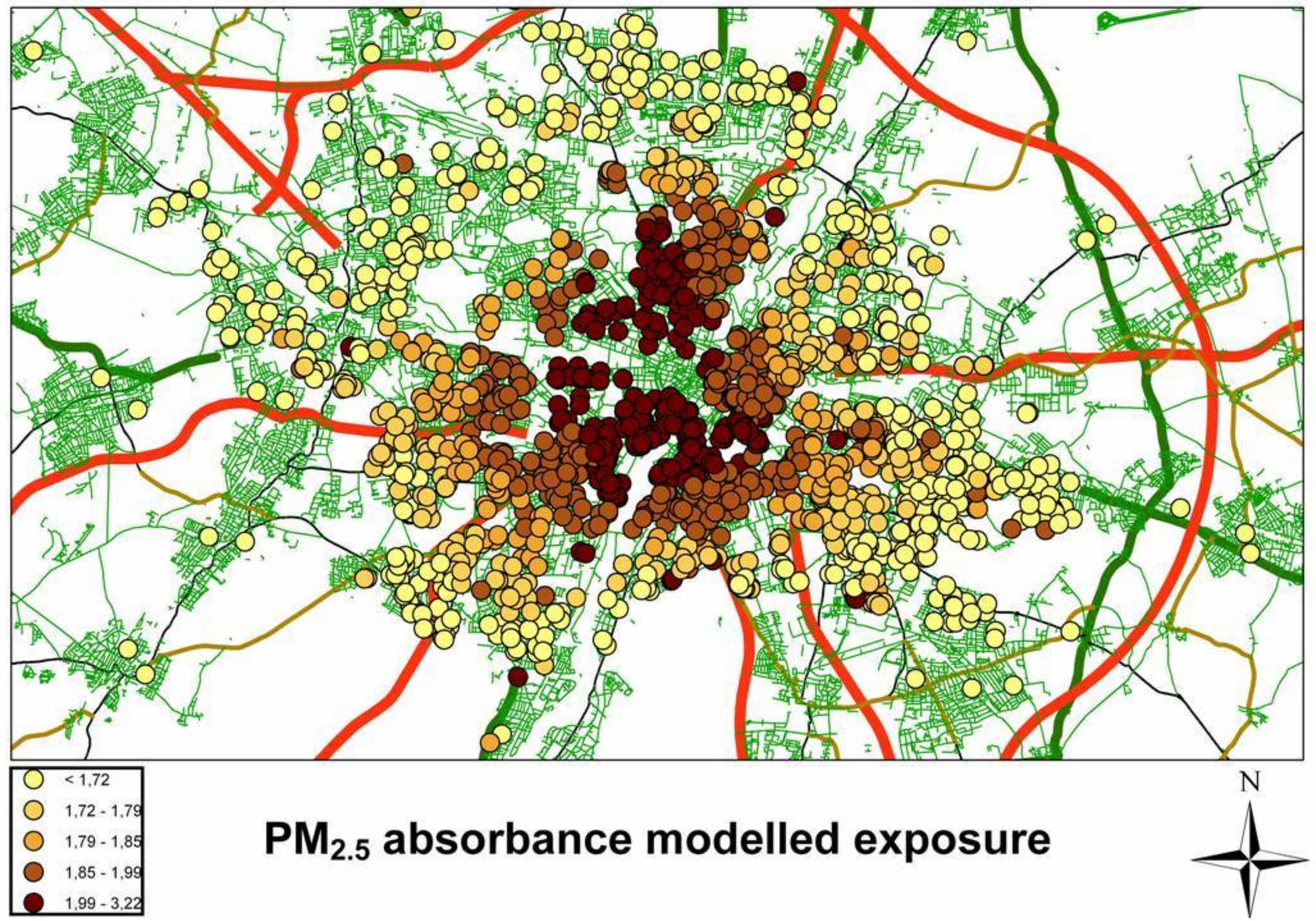
⇩

Mean PM2.5 = 11.83 + (0.004*Tvol300) + (0.268*Land300) – (0.036*Alt)

⇩

Mean PM2.5	$Tvol_{300}$	$Land_{300}$	Alt
?	11,000	40	7
?	1,000	34	10
?	2,700	25	4

$BS_{2.5}$ Konzentrationen für 1757 TRAPCA Probanden in München



Ziele

- Aufbau, Wartung und Betrieb der Messtechnik zur Erfassung des Ultrafeinstaubes
- Charakterisierung des raum-zeitlichen Vorkommens feiner, ultrafeiner und russhaltiger 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