

# Chemical composition of aerosol particles including UFPs in Saxony

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- Adverse human health effects by exposure to fine particles depending on size and chemical composition
- Observation of the EU limit value of  $\text{PM}_{10} = 50 \mu\text{gm}^{-3}$ , the Federal Administration Court in Leipzig recently pronounced a judgement confirming the right to clean air for every citizen
- Influence of fine particles on radiative forcing by direct (scattering or absorbing of radiation) or indirect (formation of CCN, thereby influencing cloud droplet size distribution and albedo) processes
- Influence on atmospheric multiphase chemistry: reactions in gas and liquid phase, reactions on particle surfaces, uptake and equilibria reactions, radical reactions  
⇒ alteration of existing particles, formation of new particles



## Motivation

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## **Investigation of**

- particle sources
- fractions of different particle sources
- long range transport effects

## **was performed by combination of**

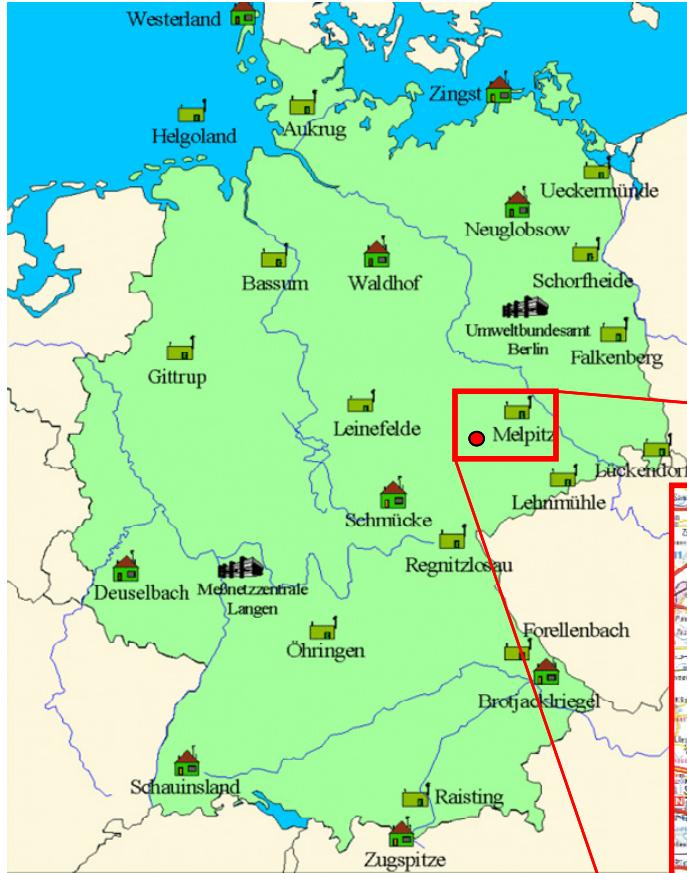
- size-segregated particle sampling using impactors
- sampling during different seasons (mostly summer/winter)
- sampling at different polluted sites in Saxony
- chemical analysis of all particle size ranges
- trajectory analysis (NOAA HYSPLIT) to estimate air mass origin



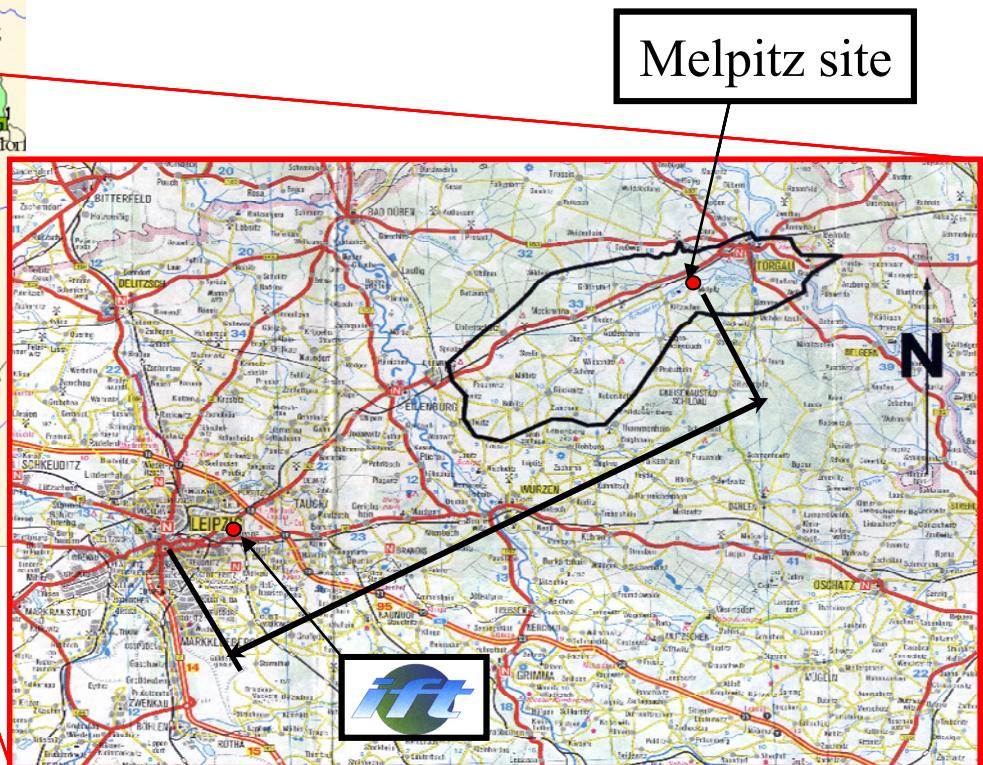
Projects in Saxony dealing with particle investigation by size-segregated sampling and chemical analysis performed during the last decade (without claim of completeness) were a.o.:

- „MINT“ (autumn 1997) at the research station Melpitz of the IfT
- „Feinstaub“, a source study (1999/2000) at 3 stations in Leipzig and Melpitz - **LfUG I**
- „Schwebstaub“, a study of size-segregated chemical composition at 2 stations in Dresden (2003/04) - **LfUG II**
- „Ferneintrag“, a study of long range transport at 5 stations in Saxony (2006/07) - **LfUG III**
- FAT project „Carbon in nanoparticles“ (2004/05), investigation of particles in a busy street canyon
- „Holzfeuerungen“ (wood burning) starting Oct. 2007 - **LfUG IV**





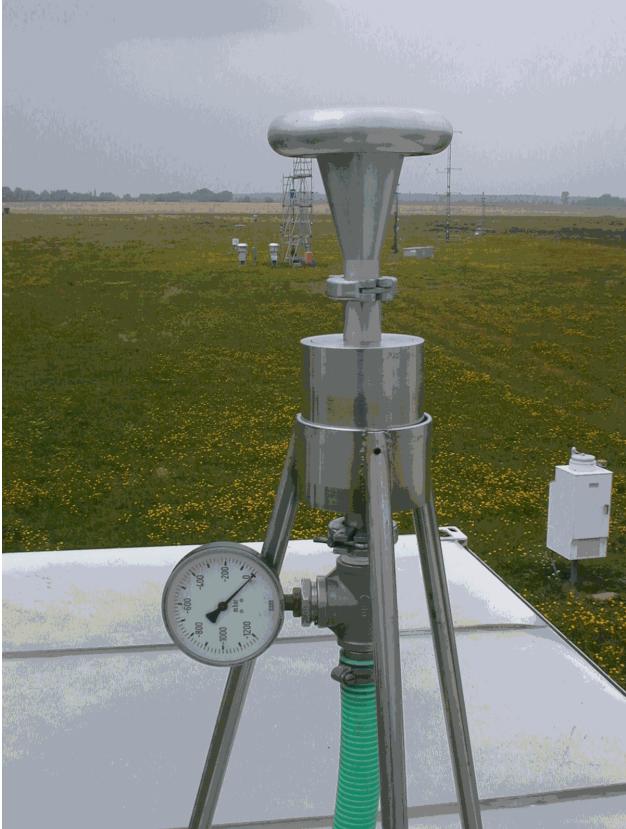
Distance:  
Leipzig (downtown) to Melpitz  
about 50 km  
( $12^{\circ}56' E, 51^{\circ}32' N$ ,  
Altitude 86 m above sea level)



UBA network and the IfT research station Melpitz

## **MOUDI/Nano-MOUDI – System** (Micro-Orifice Uniform Deposit Impactor)

MOUDI: size range  $D_{p,aer} = 0.056\text{--}18 \mu\text{m}$ /flow:  $1.8 \text{ m}^3\text{h}^{-1}$   
Nano-MOUDI:  $D_{p,aer} = 0.010\text{--}0.056 \mu\text{m}$ /flow:  $0.6 \text{ m}^3\text{h}^{-1}$   
Sampling time: (24) 48 – 96 hours



### **BERNER impactor**

Cuts:  $0.05/0.14/0.42/1.2/3.5/10 \mu\text{m}$   
aerodynamic diameter

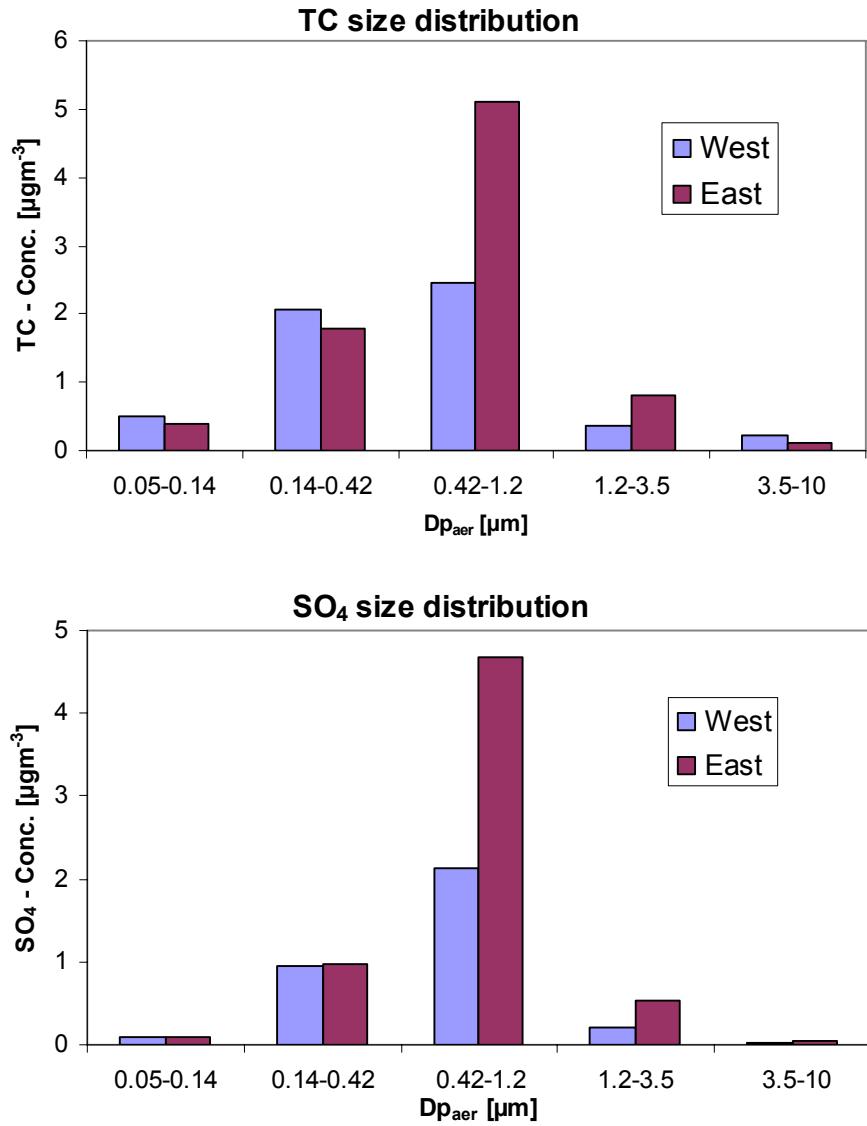
Flow:  $4.5 \text{ m}^3\text{h}^{-1}$

Sampling time: 24 hours



## **Sampling instruments**

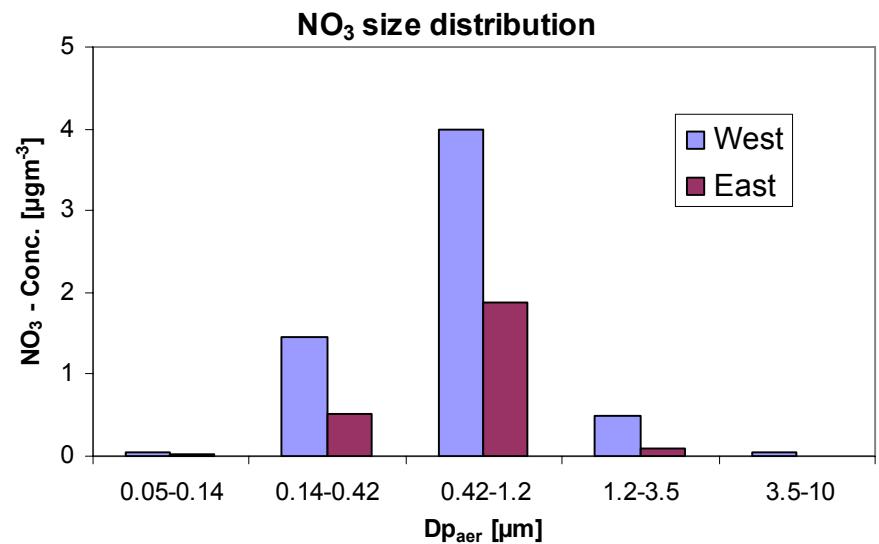




TC: significant W/E-differences on size ranges  
 $D_p_{aer} = 0.42\text{-}1.2 \text{ and } 1.2\text{-}3.5 \mu\text{m}$ , trend E > W

SO<sub>4</sub>: significant W/E-difference on size range  
 $D_p_{aer} = 0.42\text{-}1.2 \mu\text{m}$ , trend E > W

NO<sub>3</sub>: significant W/E-differences on size ranges  
 $D_p_{aer} = 0.14\text{-}0.42, 0.42\text{-}1.2, \text{ and } 1.2\text{-}3.5 \mu\text{m}$ , trend W > E



Significance was confirmed by Student's t-test  
(5% error probability)



**Some MINT 97 results (west/east – contrast)**

**Air stream North: northern Atlantic**

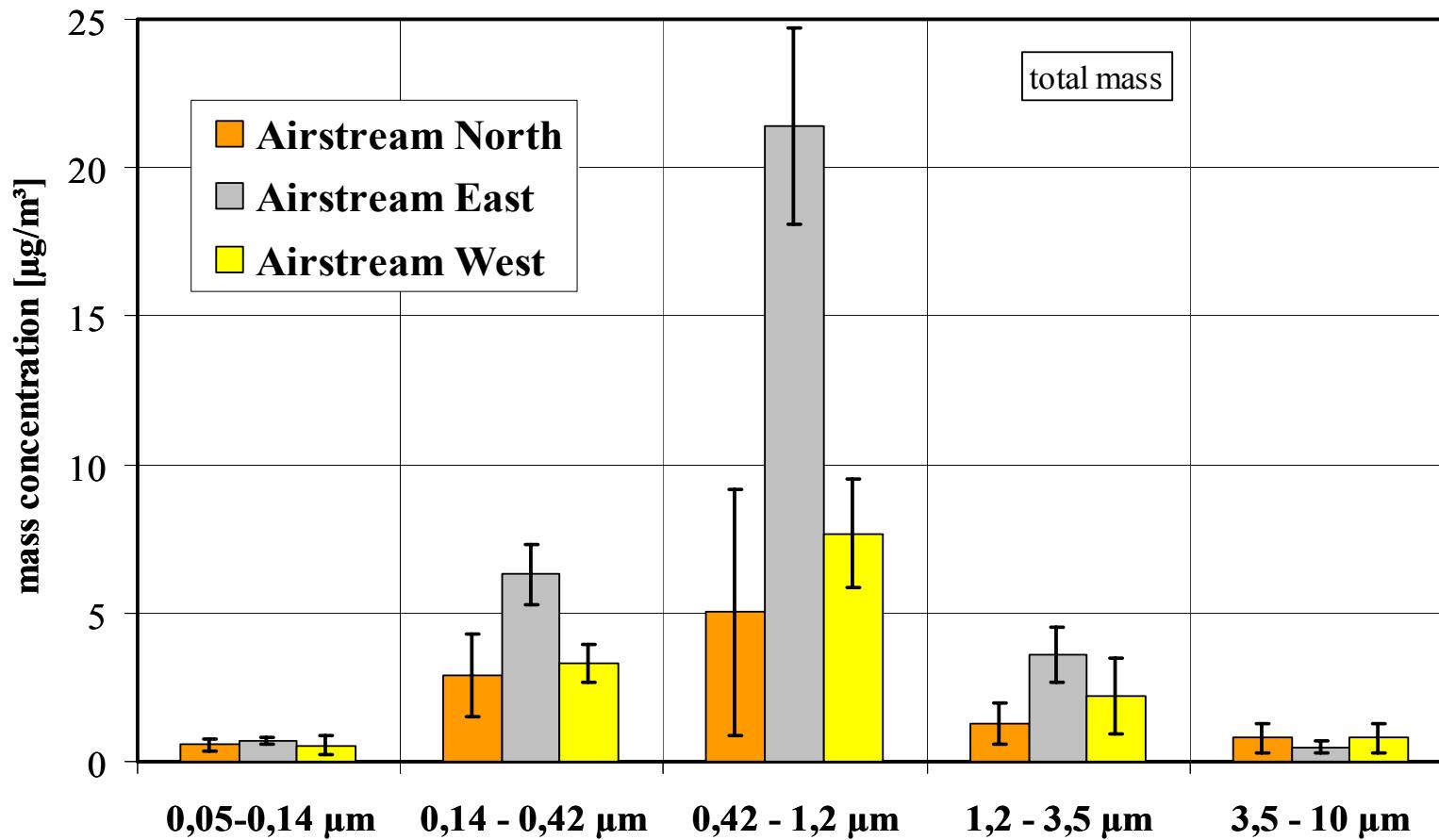
2003: 10-27, 10-30 and 11-03

**Air stream West: Atlantic and Western Europe**

2003: 11-17, 11-20 and 11-24

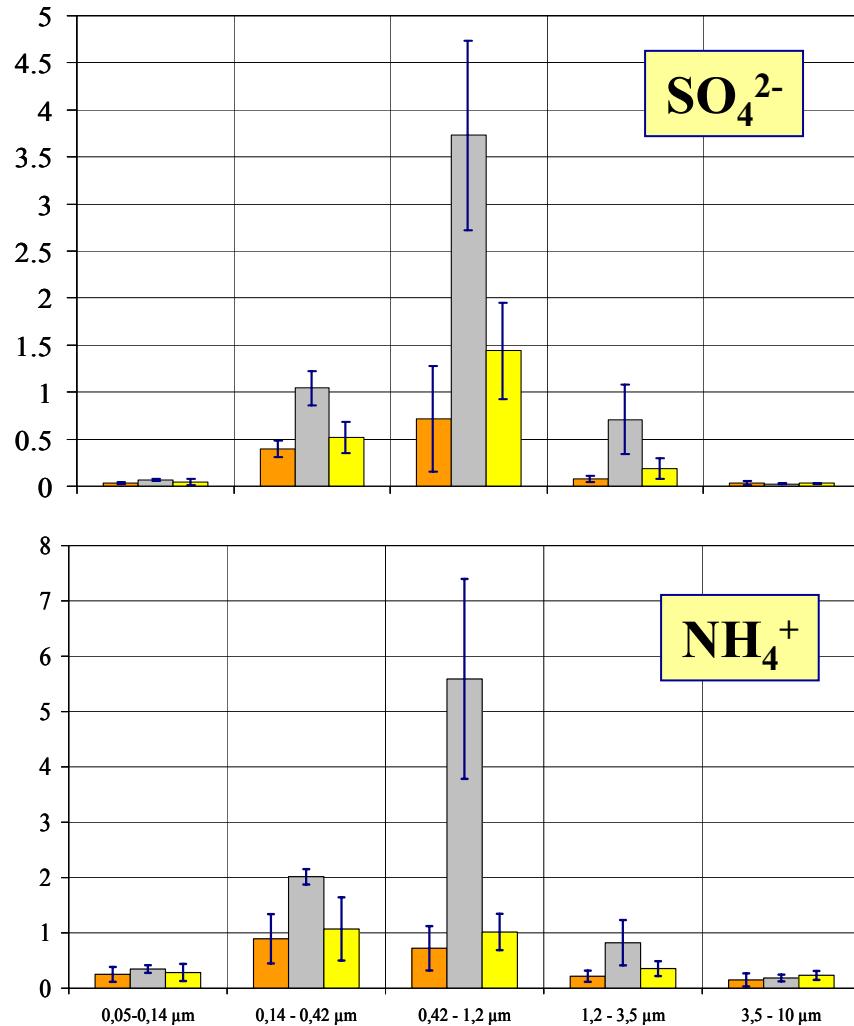
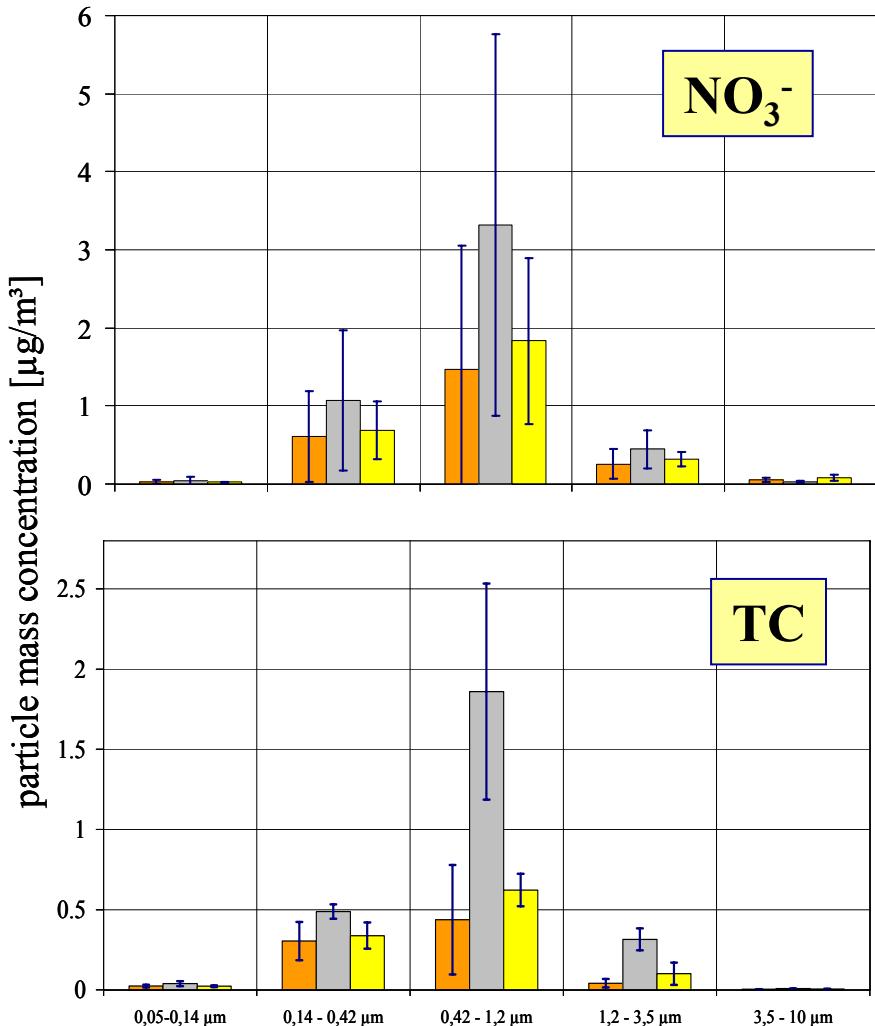
**Air stream East: continental, Eastern Europe**

2003: 11-08, 11-09 and 11-13

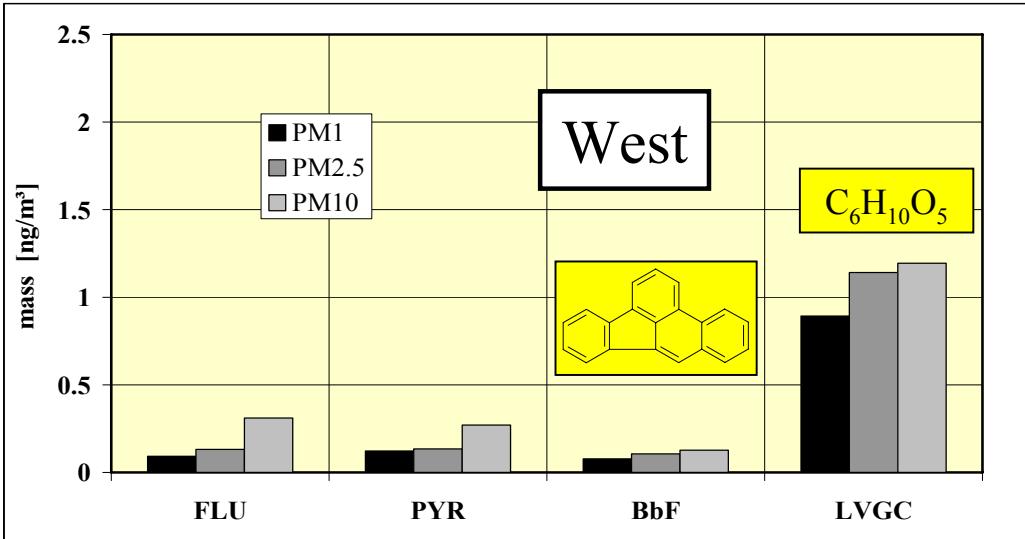


**Size segregated particle mass in 2003 Melpitz episodes**





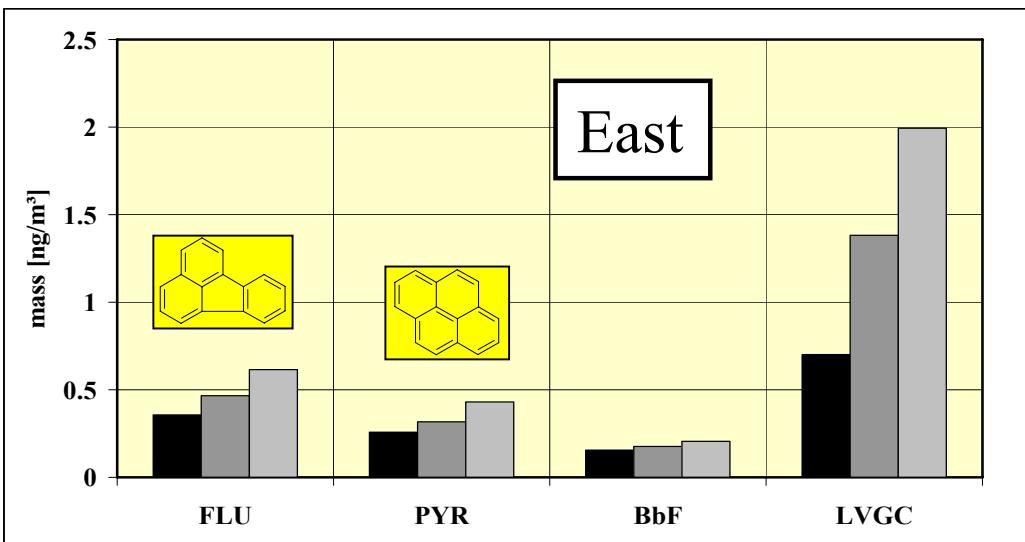
Air stream: North, West, and East



GC-MS  
(for Levoglucosan derivated with  
Trimethylsilylaminiumhydroxide)

2004-03-15

Melpitz

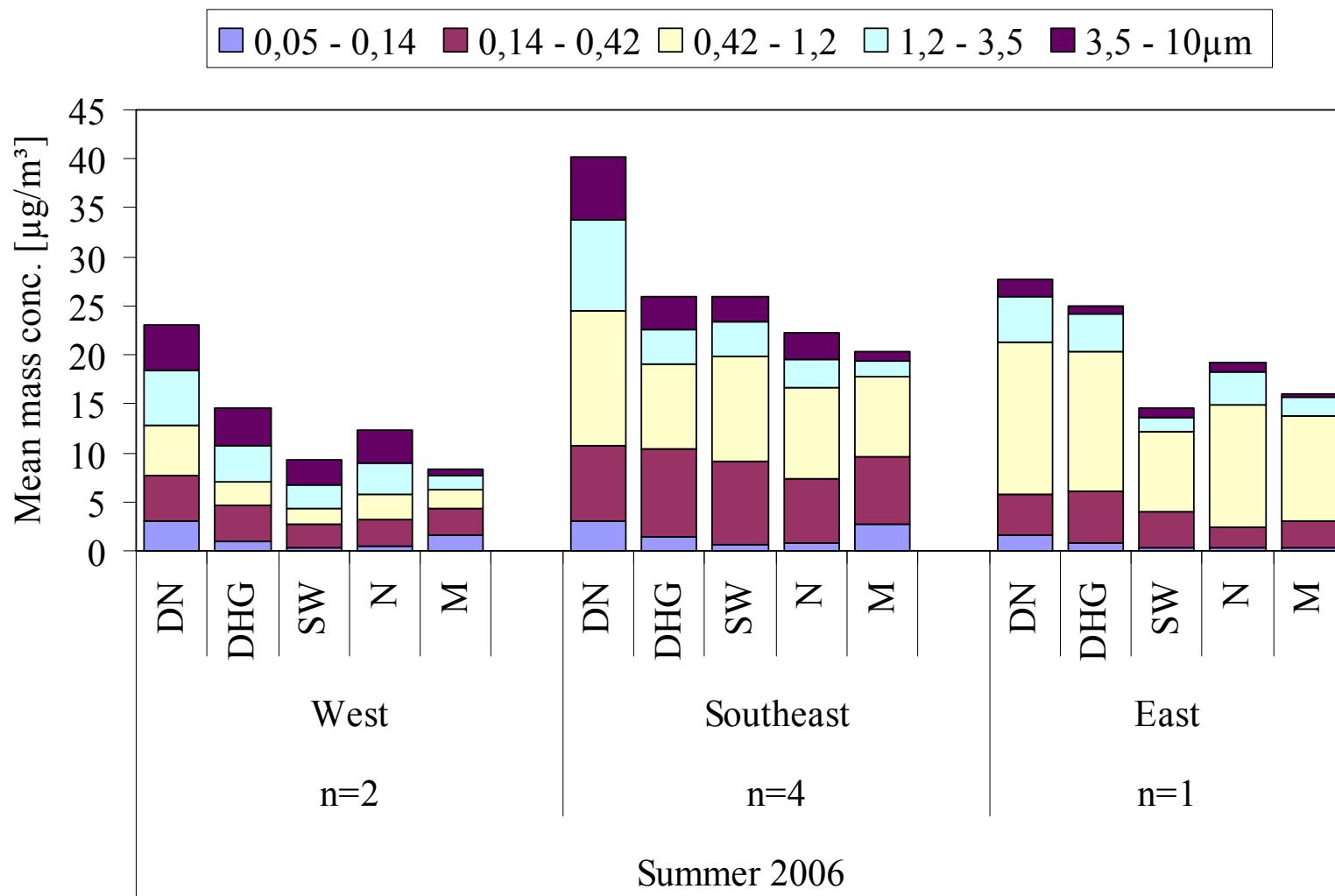


2004-03-12

FLU - Fluoranthene	combustion
PYR - Pyrene	combustion
BbF - Benzo(b)-fluoranthene	gasoline marker
LVGC Levoglucosan	biomass burning



Selected organic species according to air mass origin



**Particle mass distribution in summer 2006 (Melpitz)**



Traffic influenced  
urban site Leipzig

Urban background site  
on the IfT roof (4 km NE)



Regional background  
research site Melpitz  
(50 km NE)



**LfUG I measurement sites**

## **Source apportionment from particle measurement data using a set of basic assumptions and logical deductions:**

- 1) Traffic density is assumed to be nearly constant throughout the year
- 2) Particulate carbon in the size range  $D_{p,aer} = 0.05\text{-}0.14 \mu\text{m}$  originate entirely from traffic emissions with a characteristic OC/EC ratio
- 3) Higher size ranges include for EC aged traffic emissions and domestic heating emissions in winter (for OC the same), as well as aged traffic emissions in summer and for OC aged traffic emissions together with biogenic emissions
- 4) Diesel engine vehicles emit 90% of the particles originating from traffic in Saxony
- 5) Mixing Layer Height (MLH) is assumed to be in summer twice as high than in winter, for winter/summer comparison MLH is formally adapted by doubling summer concentrations

**Considering these assumptions calculation can be performed using following equations:**

$$(1) \text{ EC(traffic)} = \text{EC(summer)}$$

$$(2) \text{ EC(diesel)} = \text{EC(traffic)} \cdot 0.9$$

$$(3) \text{ EC(petrol)} = \text{EC(traffic)} \cdot 0.1$$

$$(4) \text{ OC(traffic)} = \text{EC(traffic)} \cdot \text{OC/EC(traffic)}$$

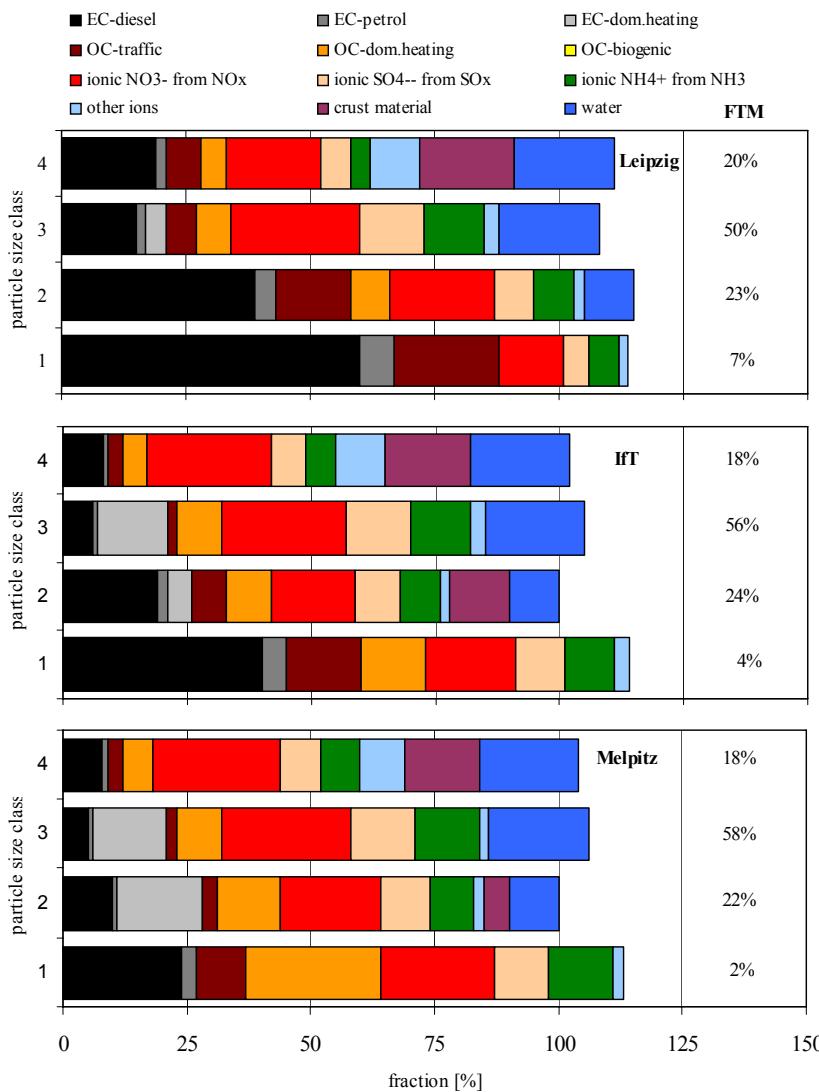
$$(5) \text{ EC(dom. heating)} = \text{EC(winter)} - \text{EC(summer)}$$

$$(6) \text{ OC(dom. heating)} = \text{OC(winter)} - \text{OC(traffic)}$$

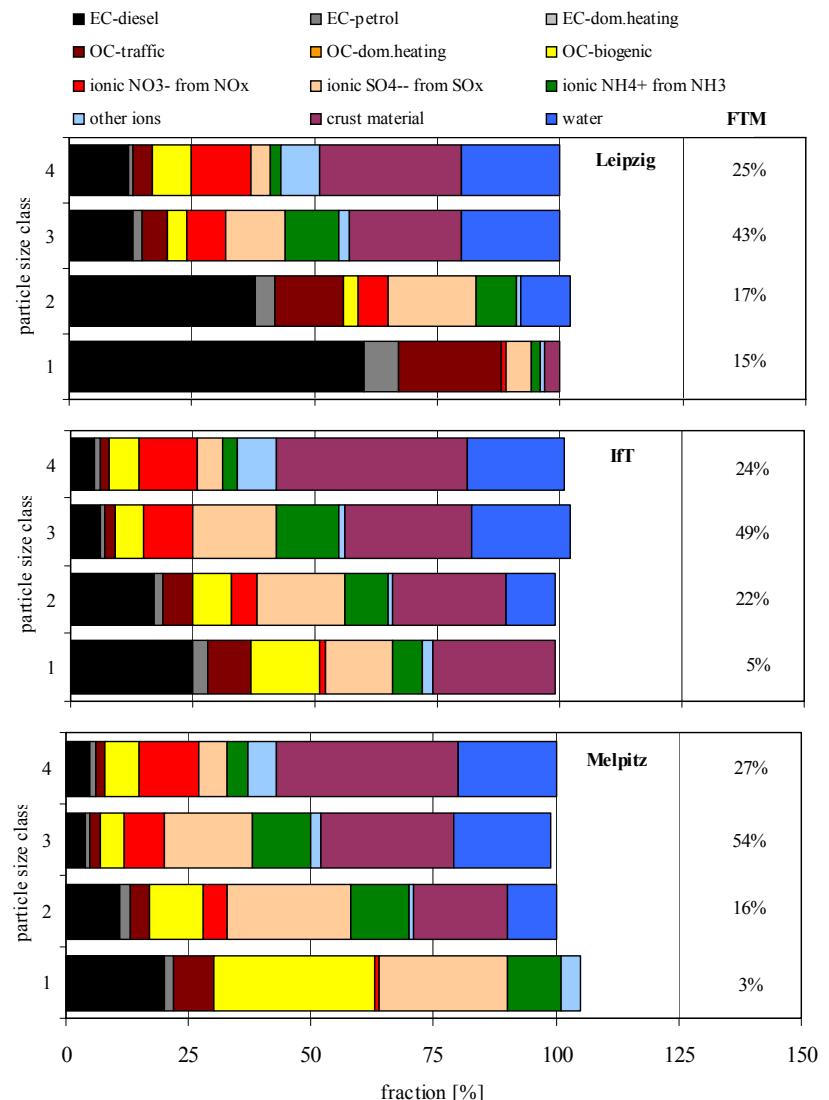
$$(7) \text{ OC(biogenic)} = \text{OC(summer)} - \text{OC(traffic)}$$



# Winter 1999/2000



# Summer 2000



Fine particle study (LfUG I): source attribution



Dresden – Nord (DN)  
Urban traffic site

Traffic density: 55000 vehicles/day

Particle sampling: PM<sub>10</sub>, PM<sub>2.5</sub>  
Size-segregated: BERNER, MOUDI

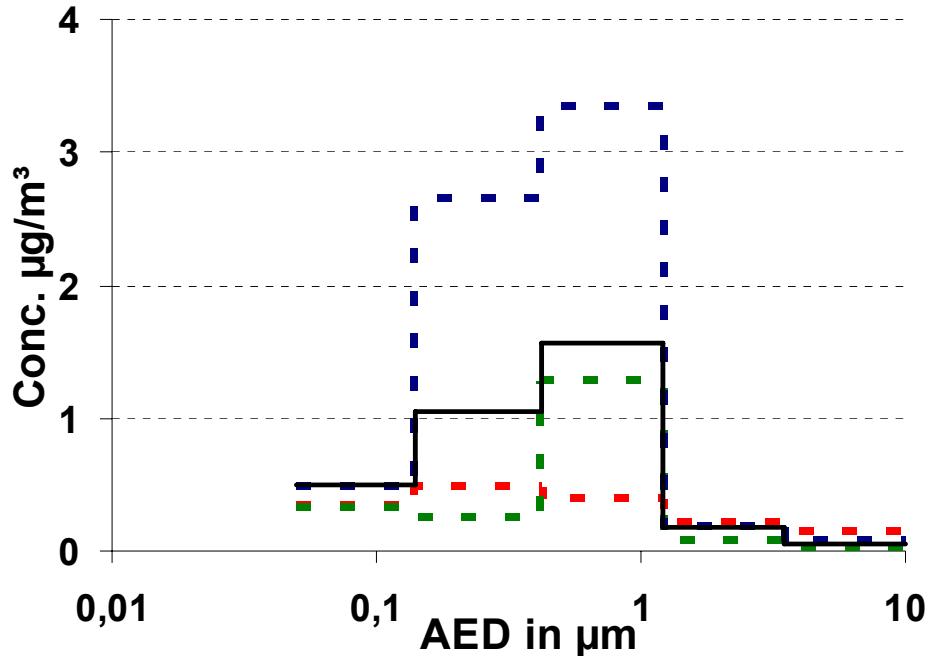


Urban background site  
Traffic density < 5000 veh./day

Winter and summer sampling



**LfUG II measurement sites in Dresden**



BERNER impactor  
5 stage sampling

Average line:  $n = 8 / 6^{\circ}\text{C}$

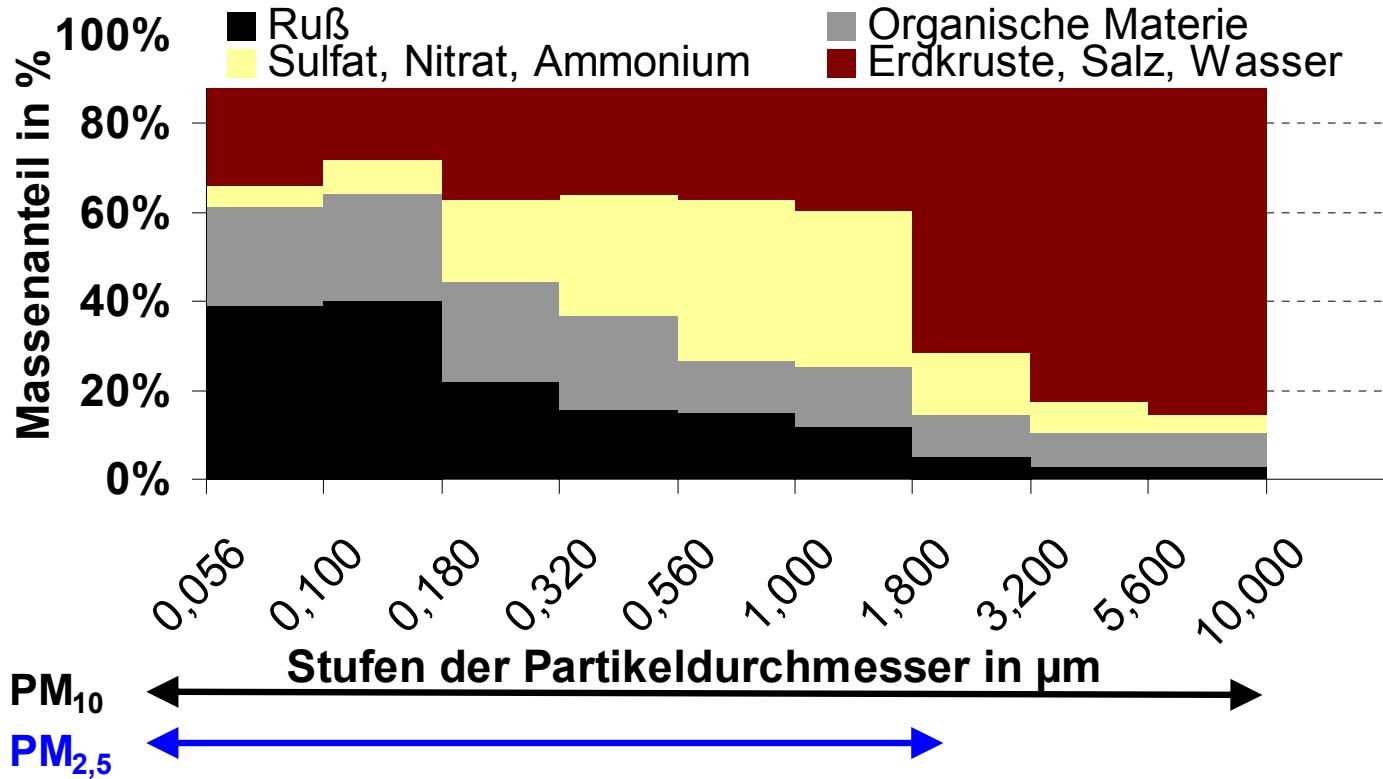
Weekday before heating period  
 $n = 2 / 12^{\circ}\text{C}$

Weekday during heating period  
 $n = 2 / 2^{\circ}\text{C}$   
domestic heating emissions

Saturday + New Year  
during heating period ( $n = 2 / 2^{\circ}\text{C}$ )  
less vehicles on the road: a half of passenger cars  
a quarter of trucks



EC before and during heating period

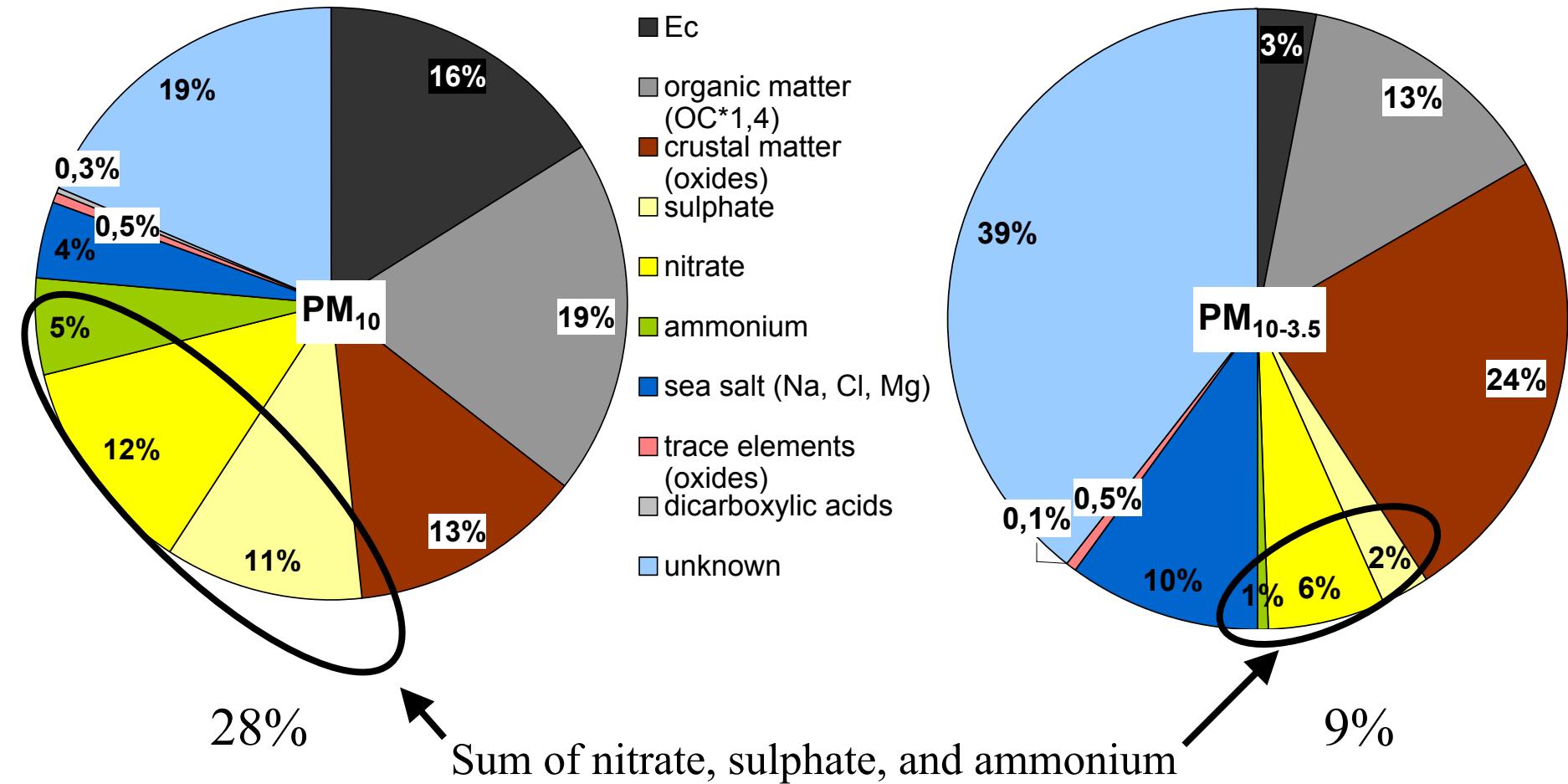


Soot fraction decreases with growing particle diameter, ultrafine particles consists to about 60% of carbonaceous material

Coarse particles mainly consists of crustal material, sea salt or thawing salt

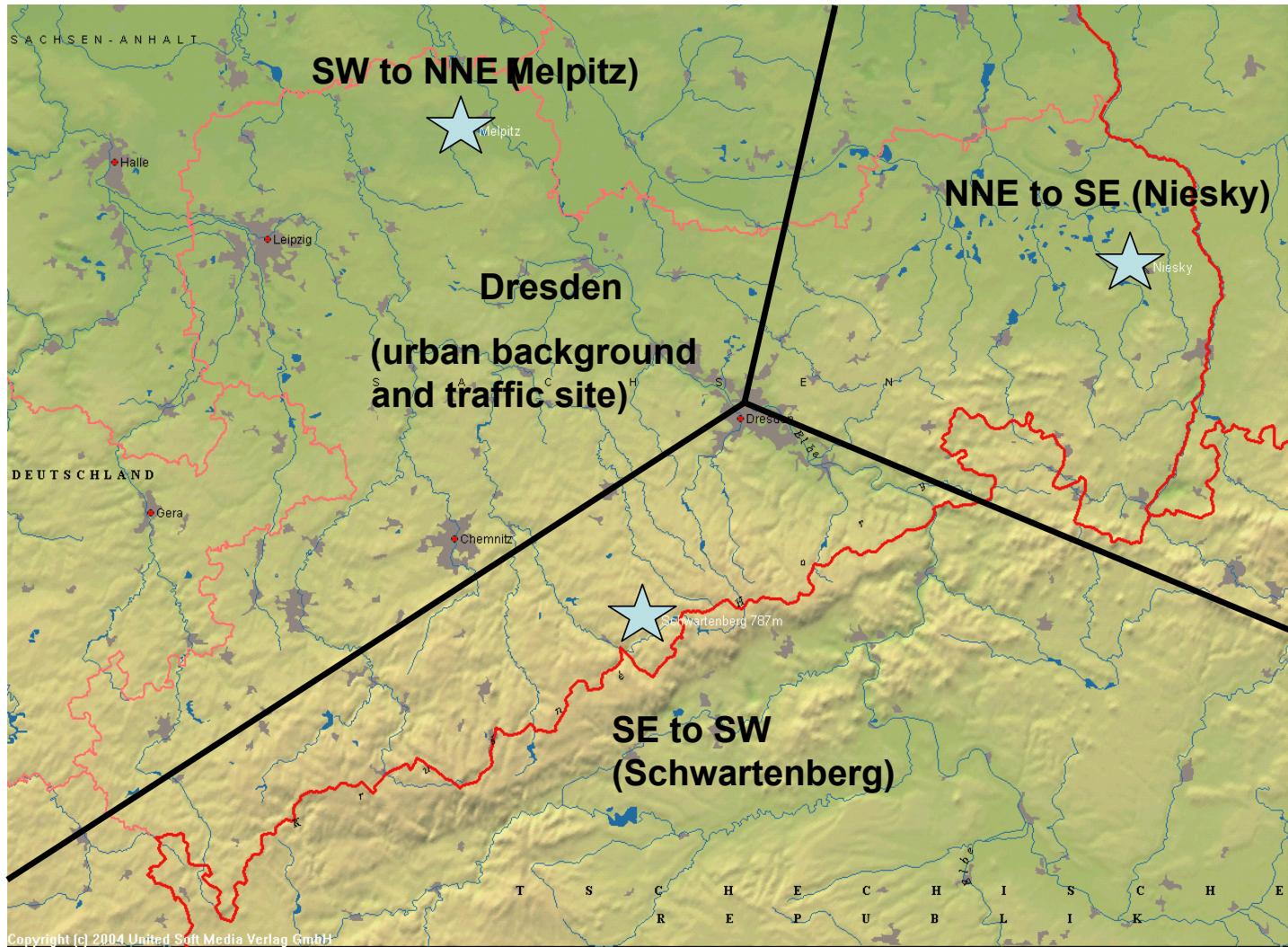


**Distribution of particle main components (MOUDI)**



## Chemical composition of PM<sub>10</sub> and the coarse fraction at the kerbside station in Dresden-Nord

## Sector arrangement for long range transport events



**SLUG III measurement sites in Saxony**

Determination of air mass origin by meteorological analysis and 96 hours backward trajectories (NOAA HYSPLIT)

Source apportionment of particle components was performed following the **Lenschow approach**:

**Local traffic = Urban traffic site – Urban background site**

**Urban background = Urban background site – Rural background site**

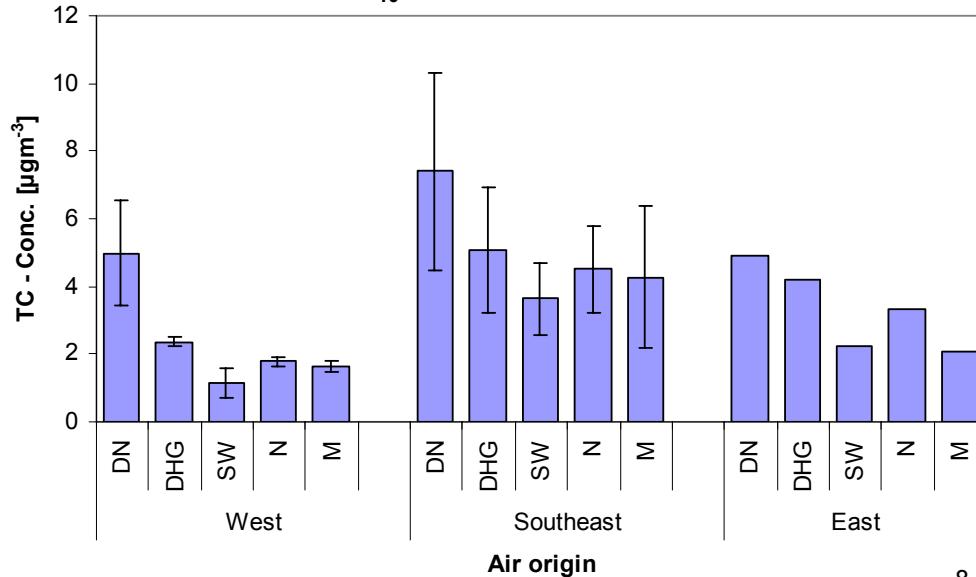
**Regional background = Rural background site**

Urban traffic site:	Dresden - North	DN
Urban background site:	Dresden - Herzogingarten	DHG
Rural background sites:	Melpitz (sector SW to NNE)	M
	Niesky (sector NNE to SE)	N
	Schwartenberg (sector SE to SW)	SW



**Estimating of particle fractions by Lenschow approach**

## PM<sub>10</sub>-TC: Summer 2006



Highest TC at DN site (traffic) at all air origin directions

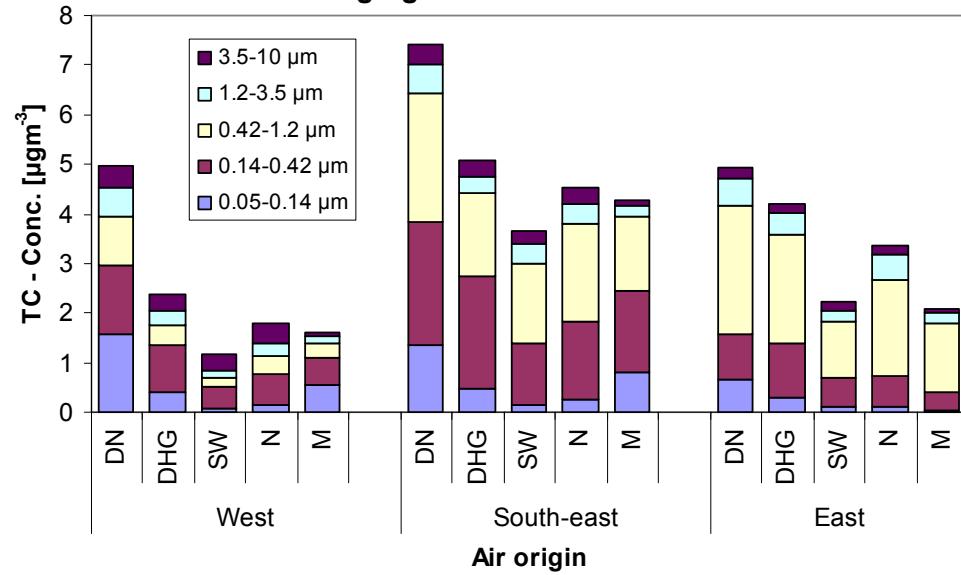
During SE and E air origin TC higher than at W direction at all sites

Size-segregation shows:

Highest fraction of UFP at DN site (fresh traffic emissions), esp. during air origin from west

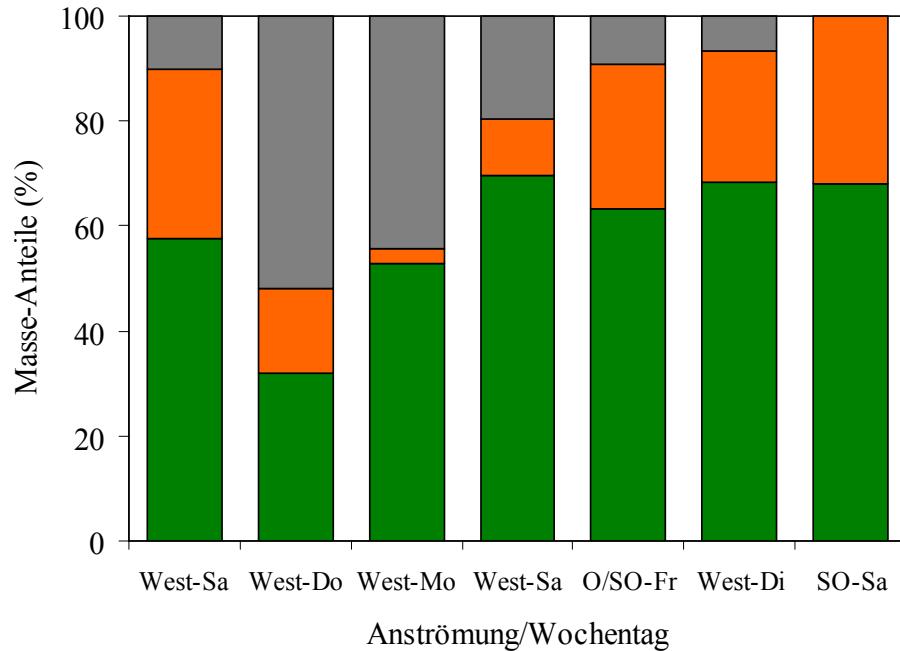
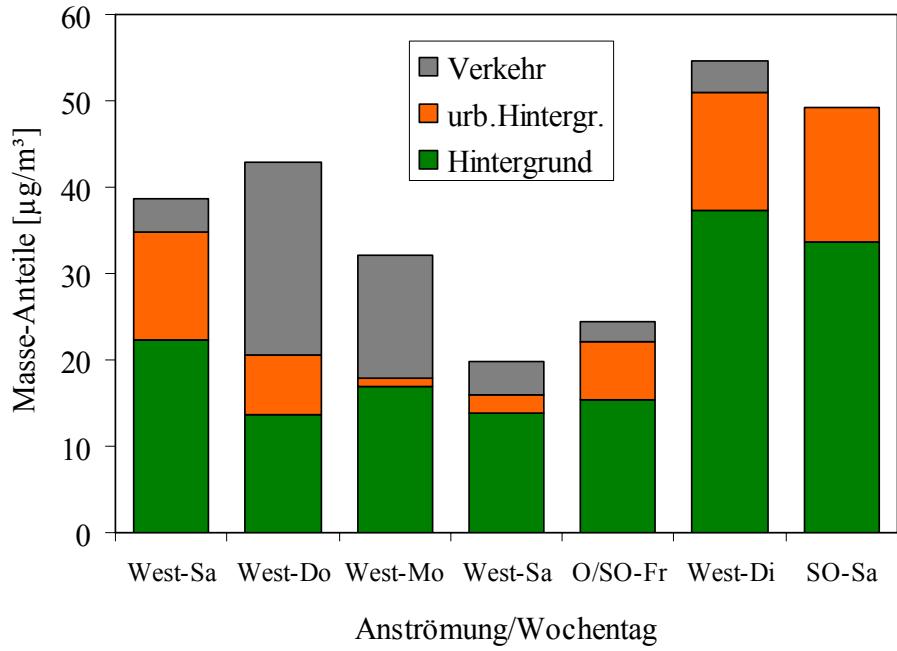
Highest fractions of 0.14-1.2  $\mu\text{m}$  particles showing residence time in air > 1 week (long range transport) during SE and E air origin

## Size-segregated TC: Summer 2006



**TC concentrations depending on air origin**



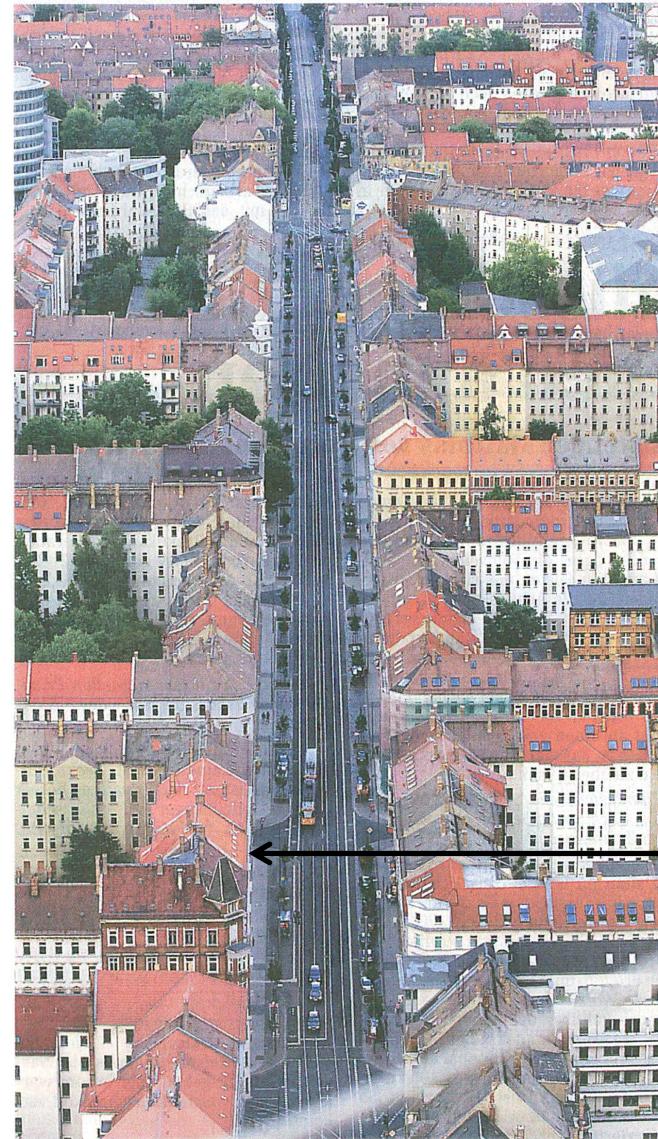


Maximum local traffic fraction of PM<sub>10</sub> was found at west air origin on workdays (up to 50%)

Regional background fraction can rise up to 60% independent of air origin and weekday

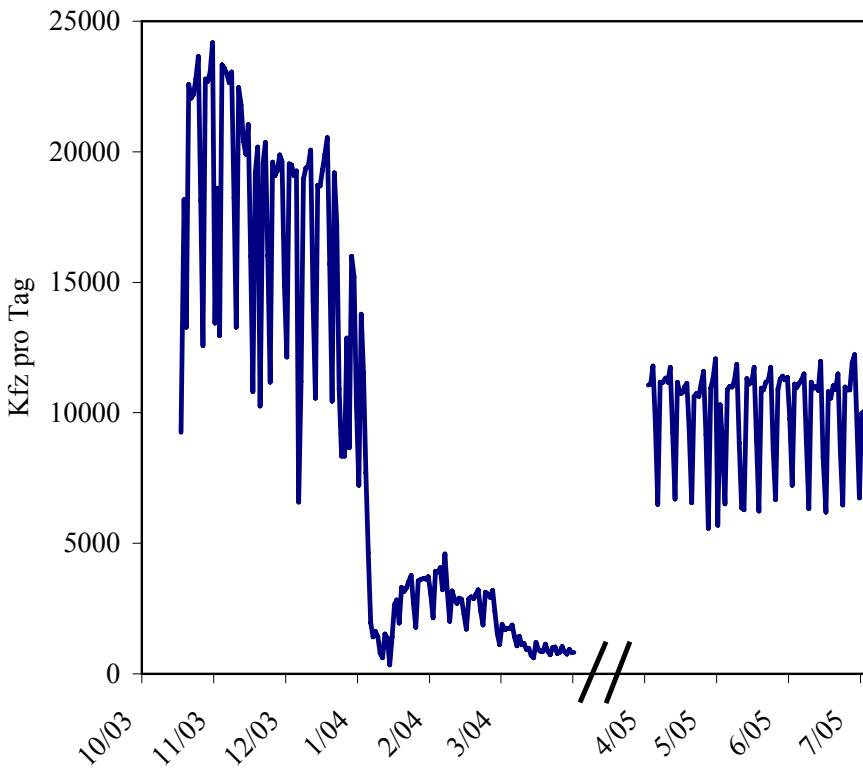
Sampling site in the Eisenbahnstraße, situated in the section between the crossroad with the H.-Liebmann-Straße (in the foreground) and the crossroad with the Torgauer Straße (in the background)

Distance to the IfT and to the city site (near main station) both about 2 km



FAT:  
Forschungs-  
vereinigung  
Automobiltechnik e.V.

Site E  
measurement inlet  
7m above ground



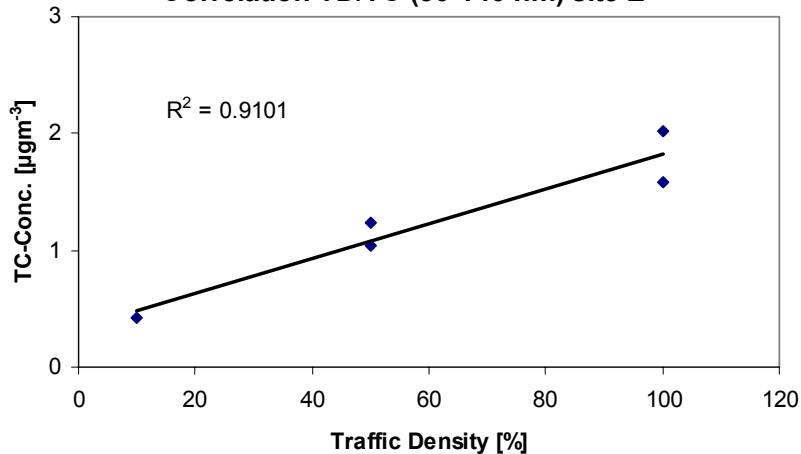
Traffic density (20000 vehicles/day = 100%) was changing because of reconstruction of the street:

Summer 2003	100%	Winter 2003	100%
Winter 2004	10%		
Winter 2005	50%	Sommer 2005	50%



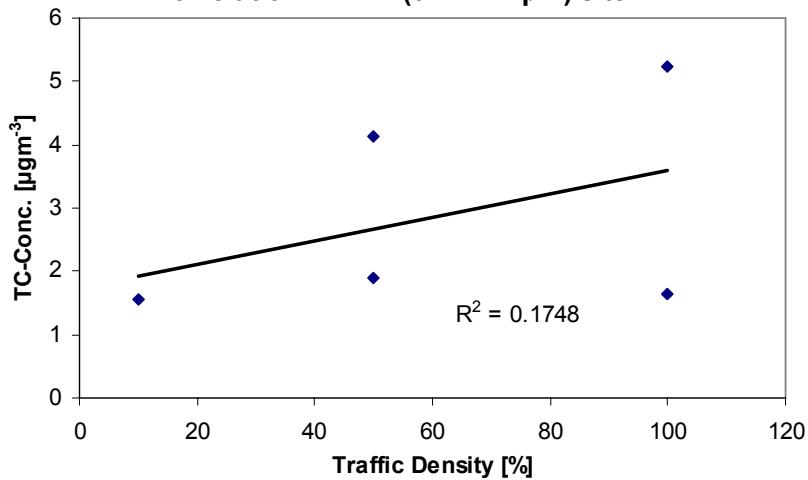
## Traffic density during FAT project

**Correlation TD/TC (50-140 nm) site E**



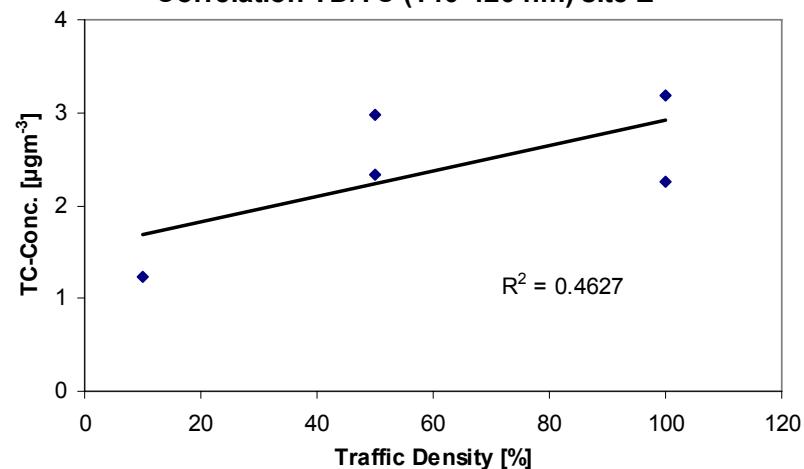
Good correlation in the UFP range

**Correlation TD/TC (0.42-1.2 µm) site E**



Weak correlation in the FP range I

**Correlation TD/TC (140-420 nm) site E**



No correlation in the FP range II



**TD / TC correlation depending on particle size range**

	Aerodynamic particle diameter [nm]		
	50-140	140-420	420-1200
Concentration [ $\mu\text{gm}^{-3}$ ]	$R^2$		
PM	0.78	0.23	0.17
TC	0.91	0.46	0.18
Fraction of local traffic [ $\mu\text{gm}^{-3}$ ]			
LT-PM	0.76	0.45	0.43
LT-TC	0.89	0.52	0.02
LT- $\Sigma$ alk [ $\text{ngm}^{-3}$ ]	0.99	0.87	0.46

$R^2 \cdot 100\%$  yields the percentage of measurement values explicable by the linear approach

Local traffic is the dominating source in the UFP size range, affect on Fine Particles in the 140-420 nm size range, but has only negligible Influence on Fine Particles in the size ranges beyond.



## Comparison of regression parameters

- West / East air pollution differences still exist
- Long-range transport of particles occurs depending on meteorological situation (esp. 0.42-1.2  $\mu\text{m}$  particles)
- EU PM<sub>10</sub> limit value of 50  $\mu\text{gm}^{-3}$  may be (nearly) achieved in such air masses before entering city areas
- Simultaneous size-segregated sampling at three sites makes possible the apportionment of particle components to the fractions local traffic, urban background and regional background for every size range.
- PM<sub>10</sub> mass distribution shows in average only about 5% and 10-15% of the total mass in the range of 0.05 to 0.14  $\mu\text{m}$  and 0.14 to 0.42  $\mu\text{m}$ , resp. Traffic emission causes the most of the particle number concentration (esp. in the UFP range) but only a minor fraction of the total particle mass.



## Conclusions and outlook I

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- Steps to observe the PM<sub>10</sub> limit value have to consider the fine particle range of 0.42 to 1.2 µm (50-60% of total mass) and the coarse mode particles beyond (resuspension).
- Steps to a real improvement of exposition situations relevant to human health should not be focussed to the observation of the mass limit value alone, but should consider also the fine and ultrafine particles existing in high number concentrations (without influencing strongly the mass limit value) and the composition of such particles including health hazardous components.
- Investigation of particles emitted by individual wood burning heating systems and their effects on air quality in Saxony are the next steps in this way.



## Conclusions and outlook II

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Thank you for your attention!



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