Criteria For Health Effects Evaluation

Of Diesel Particulate Matter

Dr. Norbert Metz
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Abstract
Mechanisms of diesel particulate matter health effects are still an open issue despite numerous investigations all over the world. Therefore additionally to the mass the parameters size and size distribution, number and number distribution, ultrafine particles, surface, chemical composition, shape and morphology have to be analysed to find the most effective criteria for health effects. Exhaust emissions regarding these criteria from passenger cars and duty vehicles are given and possible mechanism for health effects are discussed. Additionally to the size, the deposited particle mass in the lung, the chemical composition of adsorbed material and the hydrophobic, respectively hydrophilic character and also the surface are considered. Due to the complex interaction of all parameters health effects cannot be concentrated on one parameter alone. It is necessary to evaluate all parameters to understand the mechanism of health effects and to be able to derive effective measures to improve the situation. If all these parameters and their relation to each other are known, the gravimetrical determination of PM from diesel engines is the most efficient and suitable way for certification purposes.

Introduction

The dilemma regarding the evaluation of health effects from diesel particles is that whether animal tests nor epidemiological studies are able to give a reliable background for risk assessments. The defence and lung clearance mechanism is not yet fully clear: Assuming that all particles are 0.2 µm aerodynamic diameter and a particle concentration of 100 000 Particles per cm^3 is a typical urban exposure (APEG 1999-Maynard) the daily particle intake of an adult can be calculated

100 000 x 780 000 cm3/h x 24 h x 29 = 5.4×10^{11} /day.

3.35×10^{11} are penetrating to the aveoli .

Assumption is that all alvoeoli are ventilated equally. If alveoli number in the adult is 296×10^{10} macrophages (ICPR 66) then number of particles deposited per alveolus is 1130 /day. Crapo (1982) estimate 2.16×10^{10} macrophages in the human alveolar region, implying 73 macrophages per alveolus. If all particles are collected by the macrophages one macrophage ingest 1130/73 = 15 particles.

An overload is estimated at 900 000 particles, when the macrophage ingest 60 % of his own volume, which is estimated to 1000 μm^3

Oberdörster demonstrated this with particles of an diameter of 50 nm with a volume of 0.000 065 μm^3. Therefore an overload situation is far far away.

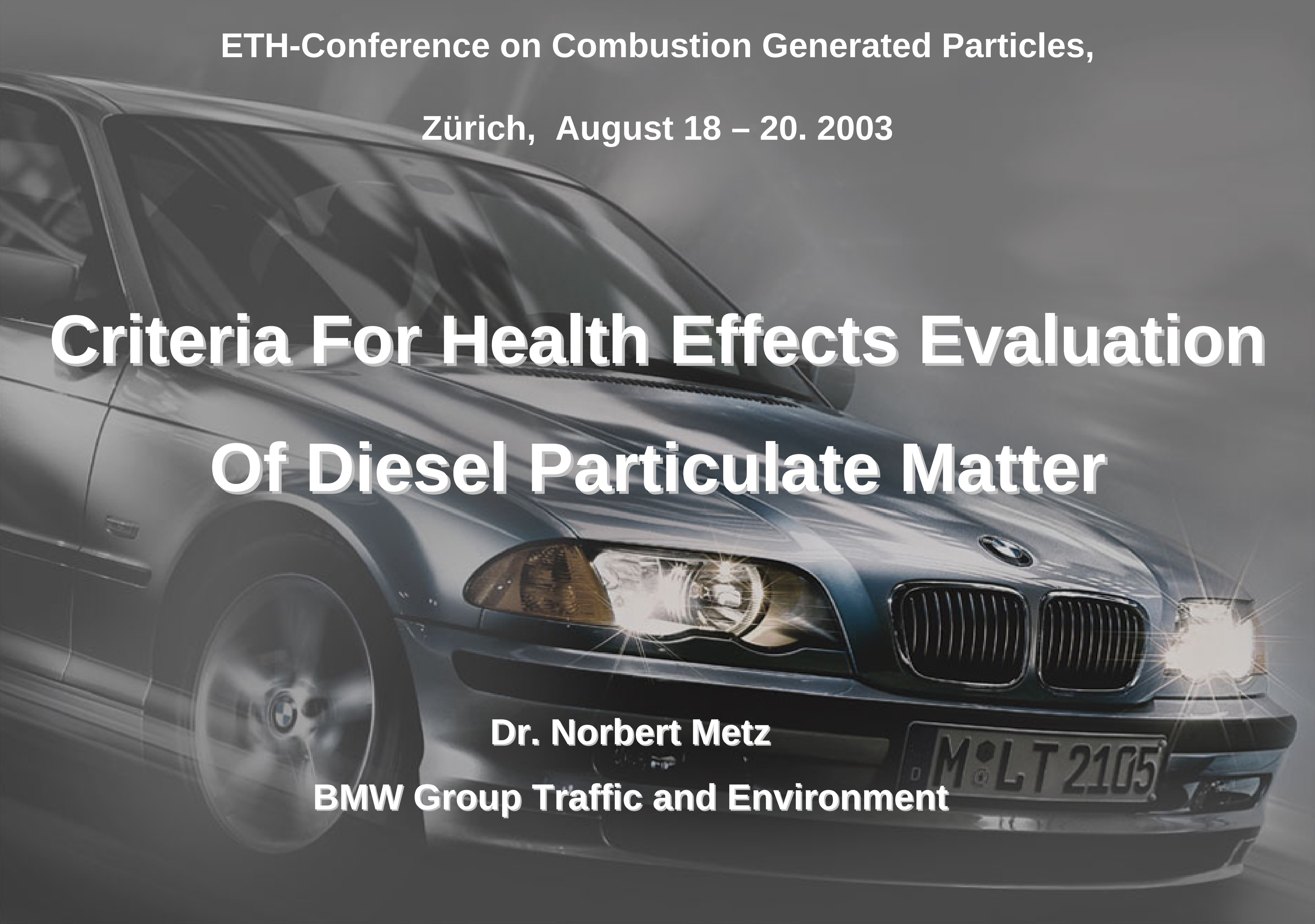
Therefore a lot of parameters have to be investigated.

Main conclusions

- The evaluation of health effects from Diesel exhaust is complex and can not be explained with one single parameter.
- Mass is certainly the most important criteria, which determine dose

response. Prerequisite is that for the number measurements a differentiation between liquid and solid aerosols must be done.

- Important criteria for the evaluation of health effects are:
  - PM mass, size and size distribution, number and number distribution, PM concentration in ambient air, PM surface, shape, morphology, chemical composition inclusive adsorbed material and hygroscopic properties together with bioavailability
- PM mass emission is declining, despite increasing mileage.
- A lower sulphur- content in the diesel fuel strengthen this trend.
- PM emission both mass and number decreases continuously.
- For certification the gravimetric measurement of mass is still the most efficient and suitable procedure.



ETH-Conference on Combustion Generated Particles,

Zürich, August 18 – 20. 2003

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Introduction	Dilemma for the Evaluation of Health Effects
PM Mass	Particulate Matter Emission (PM_{10} , $PM_{2.5}$, Diesel Soot) Emission Contribution of Different Sources in Germany Development of PM-Emissions from Road Transport regarding PM_{10} , $PM_{2.5}$, Soot (EC) and Number
Mass and Number	Correlation between Mass and Number, Fuel Influence
PM Size	Size Distribution (Emission / Air Quality)
PM Surface	Surface for Euro III and Euro IV Passenger Cars
PM Morphology	Diesel PM, Urban Aerosol and Particles in the Lung
PM Chemistry	Chemical Composition (Diesel PM and Ambient)
Health Effects	Hypothesis for Effects of $PM_{2.5}$ and Diesel Soot
Summary and Conclusions	

Tools For Research Upon Health Effects

In Vitro-Test

Ames-Test

Cytogenetic Test

DNA-Repair Test

Cell trans-formation Test

In Vivo-Test

epicutaneous
subcutaneous
intratracheal
intrapereonal
inhalative

Animal Species

Mice (e.g. Scenar)
Hamsters
Rats
(e.g. Fisher 344, Wistar, Sprague Dawley)

Epidemiology

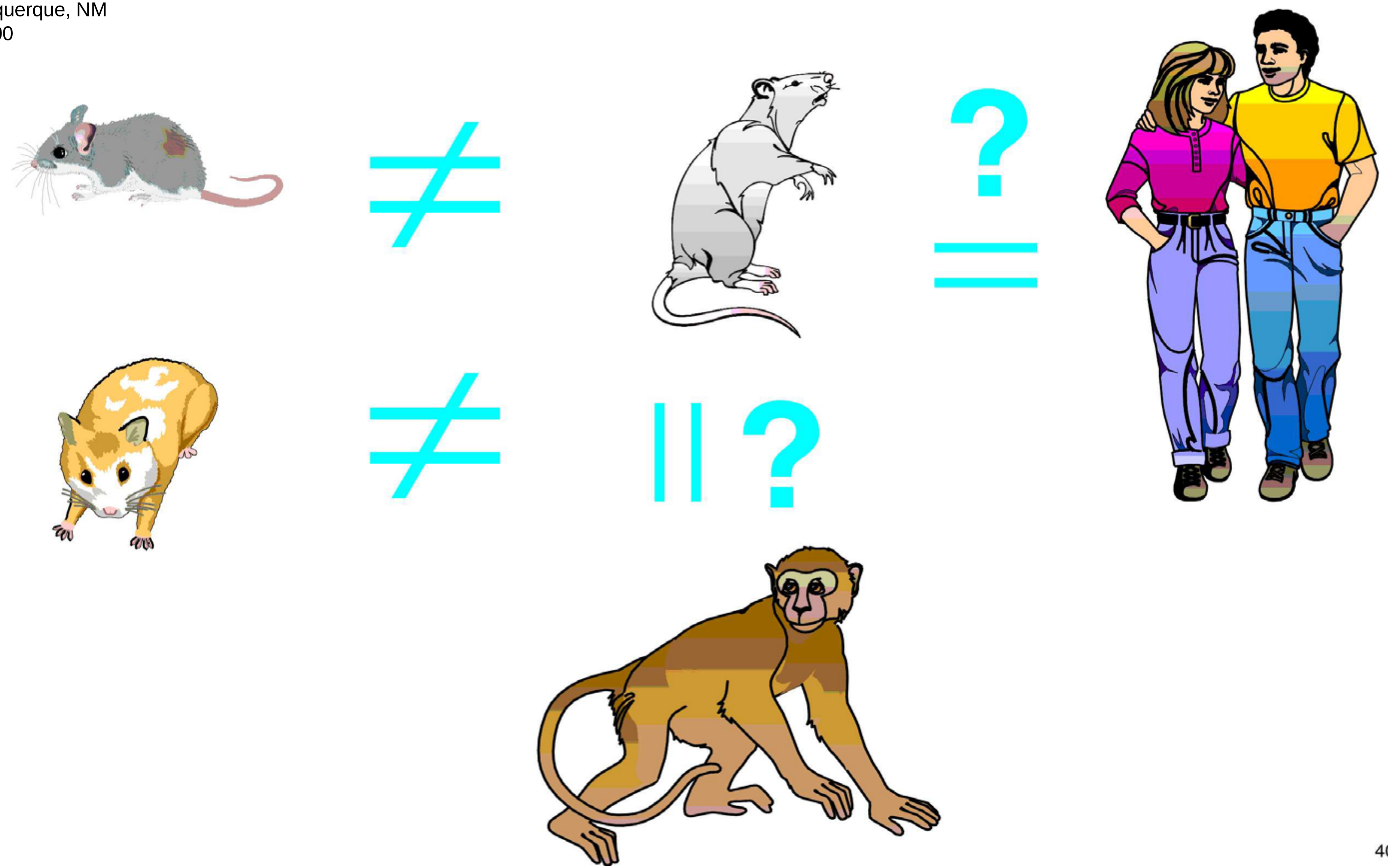
Case Studies

Cohort Studies

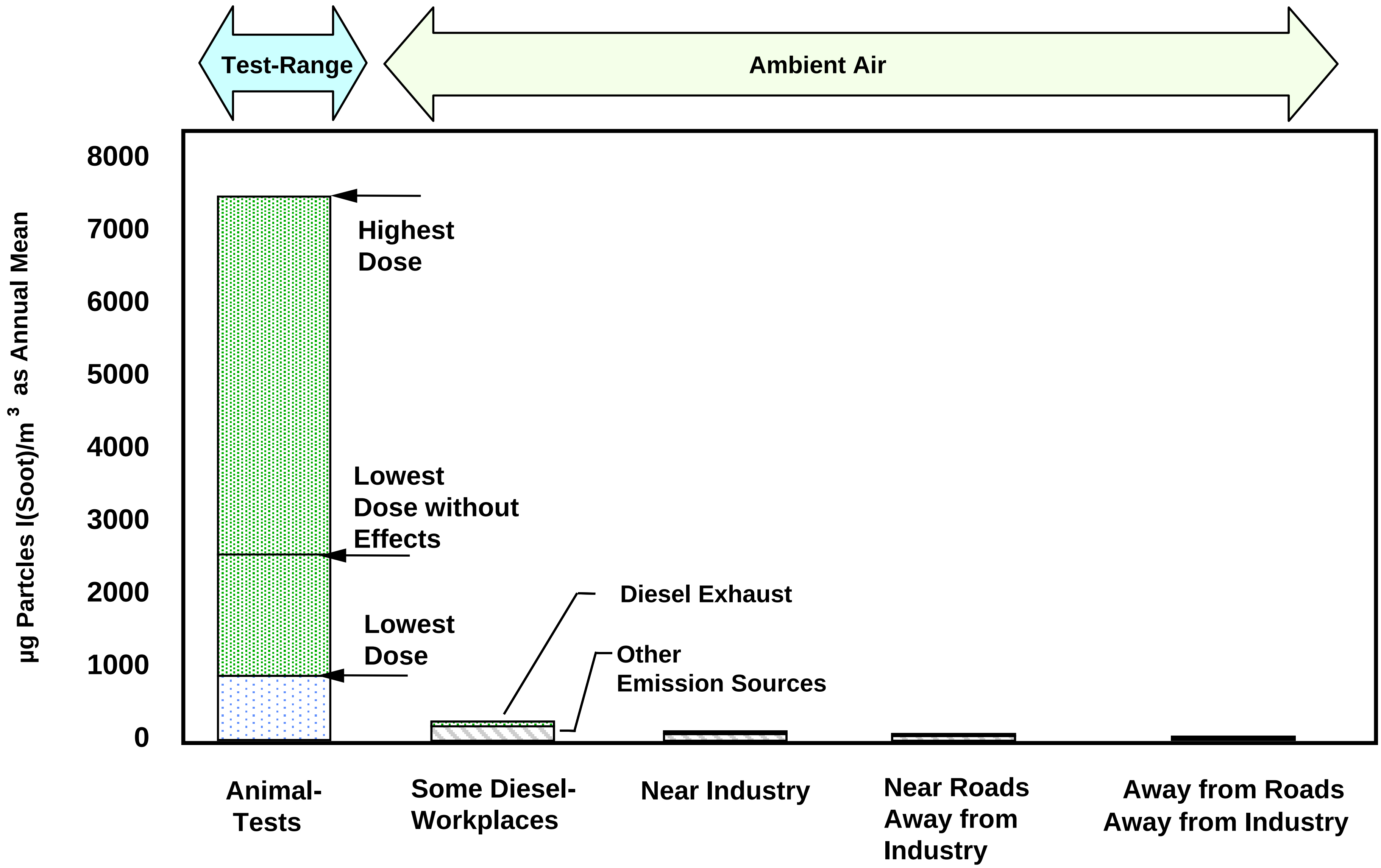
Teratogenic Studies

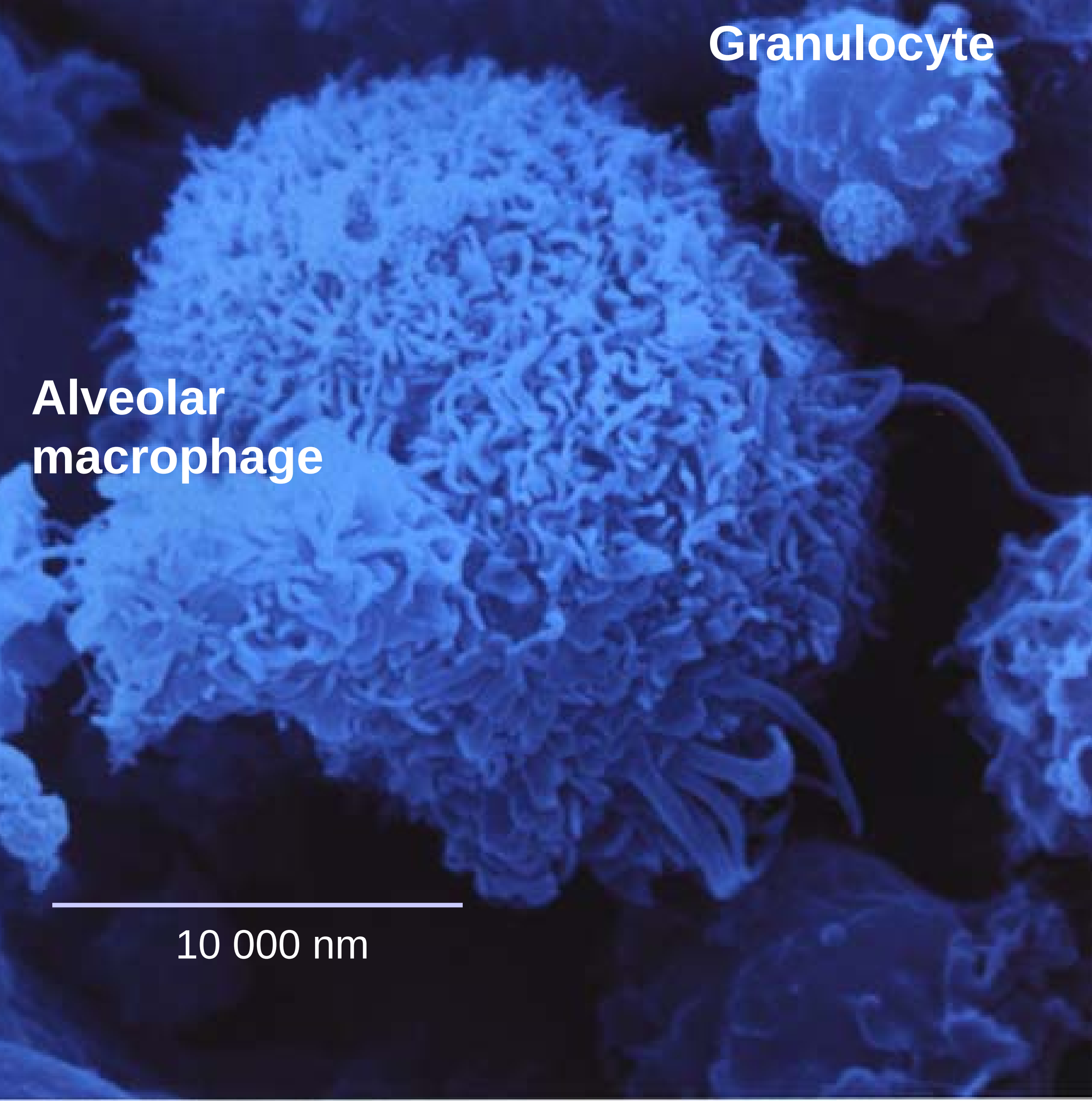
Risk Assessment for Humans

THE QUESTION IS RELEVANCE TO HUMANS



Comparison Of Particle Exposed Concentration Ranges





Important cells of the lung on the way to particle-induced tumor genesis.

Main function: Defence and lung clearance

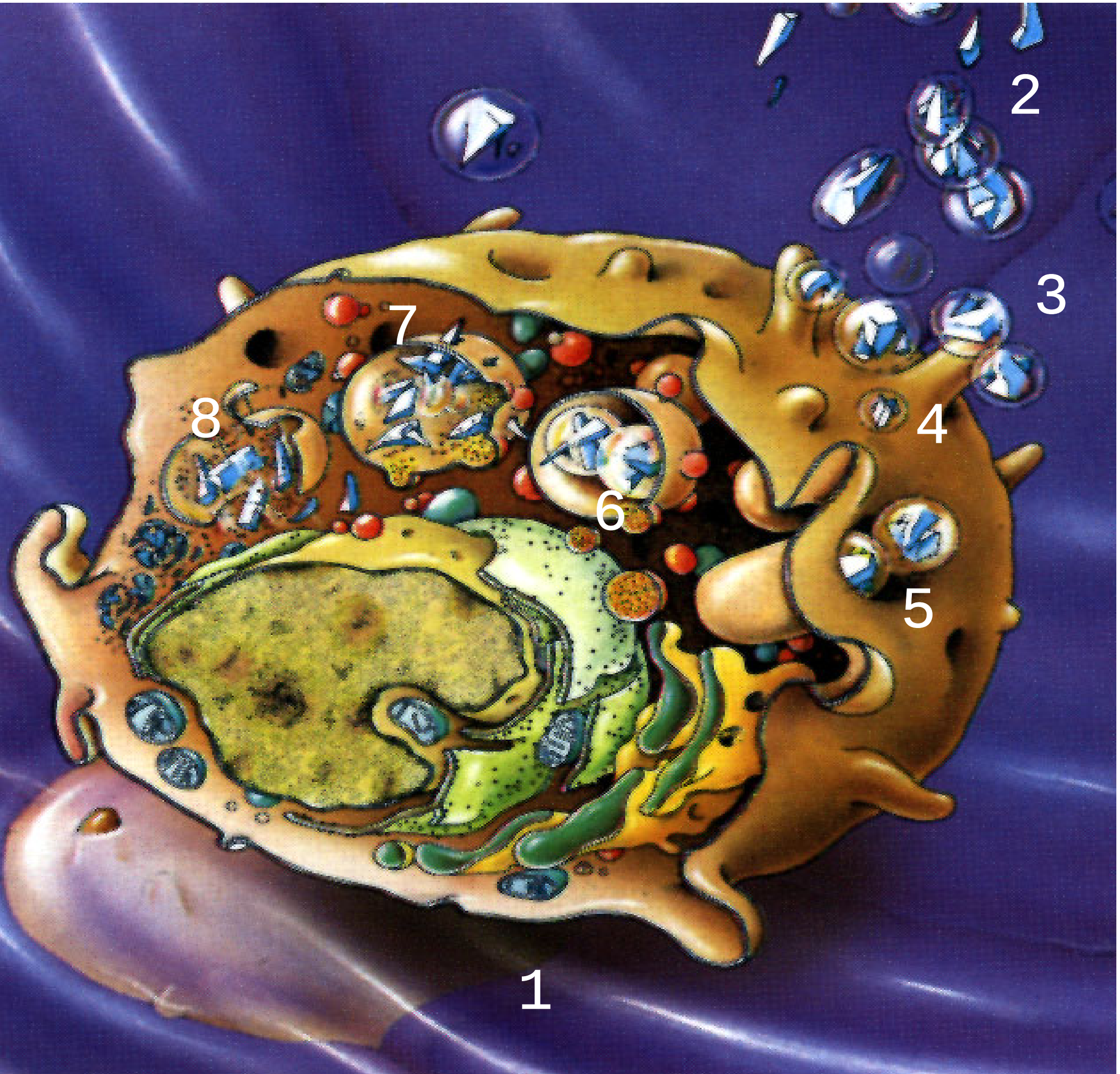
Activated Alveolar Macrophage (center)
Granulocyte (right upper corner)

REM Image from a lung cell

Ref.: Bruch J., Uni Essen 2003

Me-3806

Macrophages bind with „Surfactant“ inhaled Particles in the Lung



1 = Surface ot the alveolar epithel

2 = Particles coming in the alveole

3 = Wrapping of particles

4 = Contact with the macrophage

5 = Entrance in the Cytoplasma

6 = Formation of Phagolysomes

7 = Direct Reaction

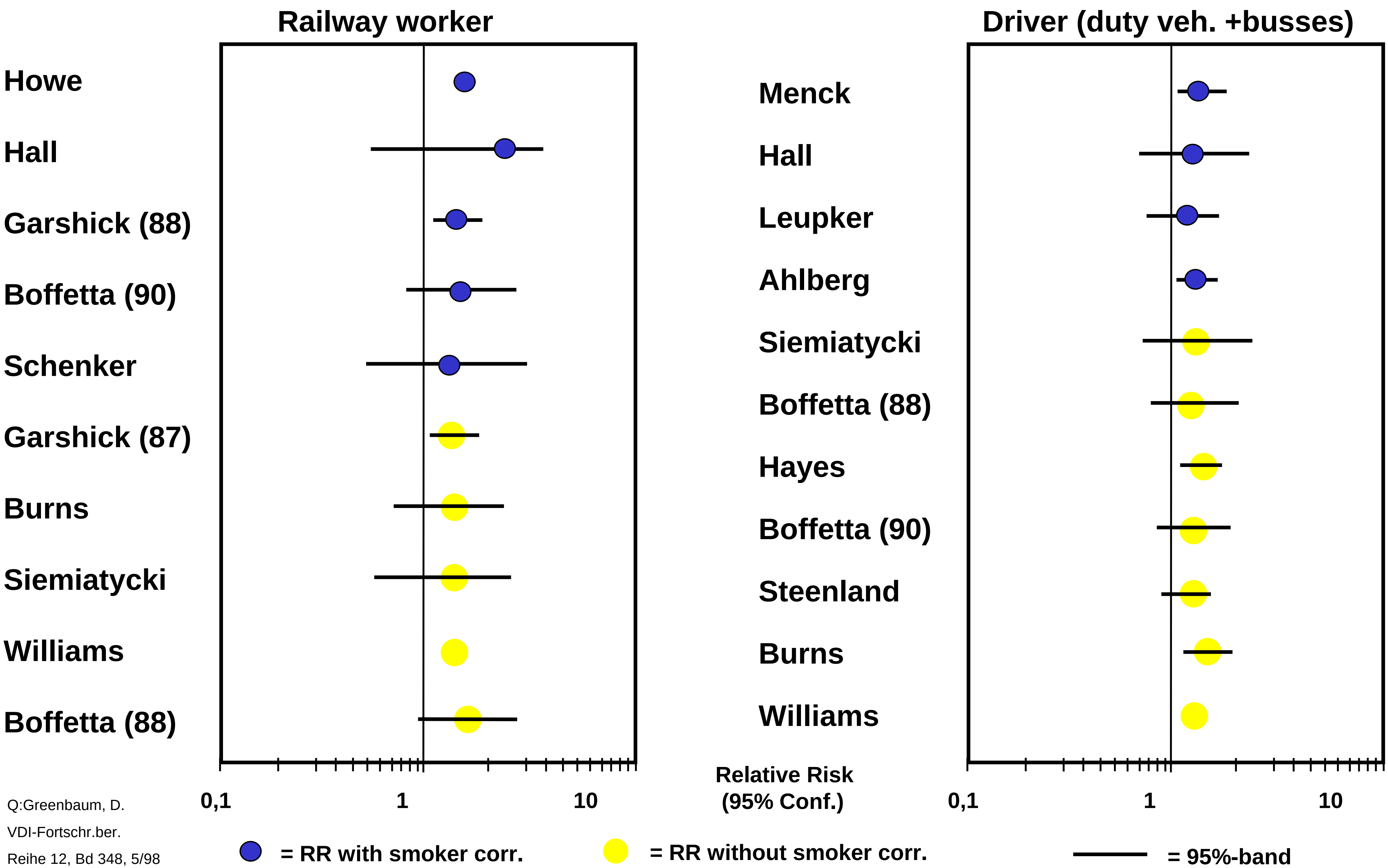
8 = Release of lytic encymes

Questionable Relevance Of Rat Test Results To Humans

- Hamster don´t show even with high dose tumors, with mice the reactions don´t allow clear conclusions.
- Rats exposed to low dosis developed no tumors.
- With high dose rats showed tumors, which are not diesel specific. They also appeared with TiO_2, elemental carbon and other fine particles fractions.
- The formation of tumors is due to the overload of the rat lung with particles.
- Locations of the particle deposition and reactions in the lung, due to inhaled particles are total different in rats and humans.
- Metabolismus in rats and in humans is not comparable.

➡ **Rat data can not be used therefore for risk assessments to estimate a danger for humans.**

Epidemiological Studies And Relative Risk



Q:Greenbaum, D.
VDI-Fortschr.ber.
Reihe 12, Bd 348, 5/98

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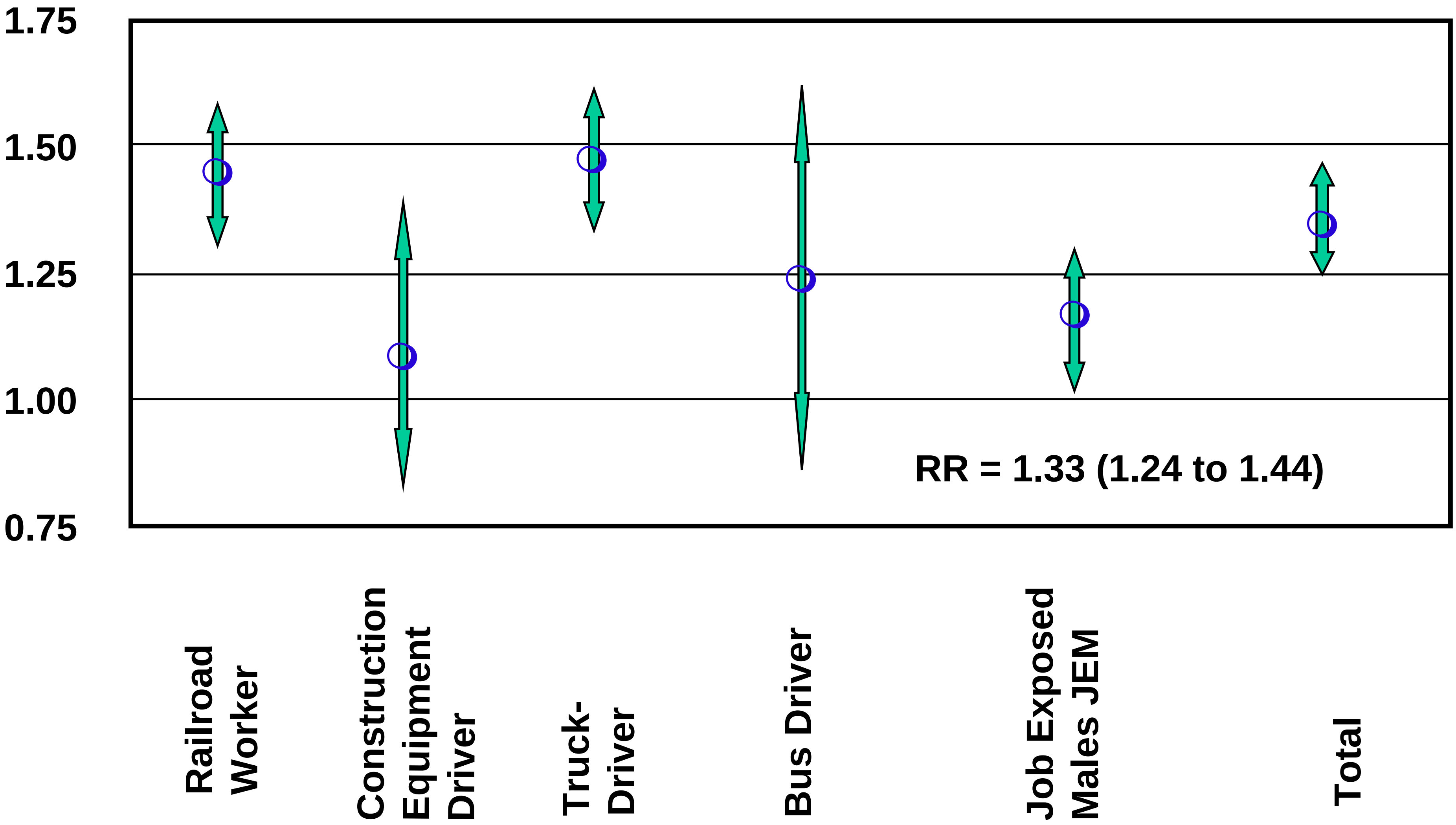
Me- 3810
Greenbaum, D.
VDI-Fortschr.ber.
Reihe 12, Bd 348, 5/98

Relative Risk (derived from Epidemiology)

Metaanalysis Summary of Bhatia



Weak Points in Epidemiology Regarding Risk Estimations

- Exact exposition data are mostly unknown and later not deductible.
- Collectives exposed only to diesel exhaust are not available.
- Other confounders (e.g. smoking, alcohol use, drugs, etc.) are often not - or insufficient - taken into account.
- Smoking as a strong carcinogen has even at low concentrations stronger effects as particles in urban aerosols, since particles are a weak carcinogen, if at all.
- Job induced nutrition and life style habits are hard to be taken into account adequate.
- Also genetic burden and illness can not be taken into account.

➡ **Therefore results of existing epidemiological studies can not be used for risk assessments.**

Criteria to Characterize Diesel Particulate Matter for Health Effects Evaluations

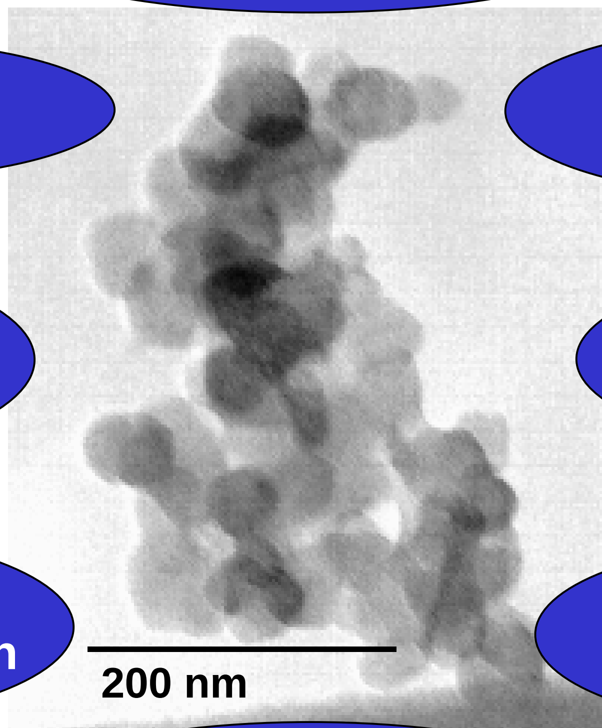
Concentration [g/m³]

Mass [g/km]

Shape and Morphology

Size [µm oder nm]
Size Distribution

Hydrophobic resp.
Hydrophilic Character



Number and
Number Distribution

Surface [m²]

Chemical Composition

Which Particles Are Responsible for Health Effects ?

- Total Suspended Particulate Matter
- Particles with a size below 10 µm (PM_{10})
- Particles with a size below 2.5 µm ($PM_{2.5}$)
- Particles with a size below 0.1 µm ($PM_{0.1}$)
- Ultrafine Particles (below 50 nm)
- Soot Particles, Total Carbon
- Organic Carbon (OC)
- Elemantal Carbon (EC)

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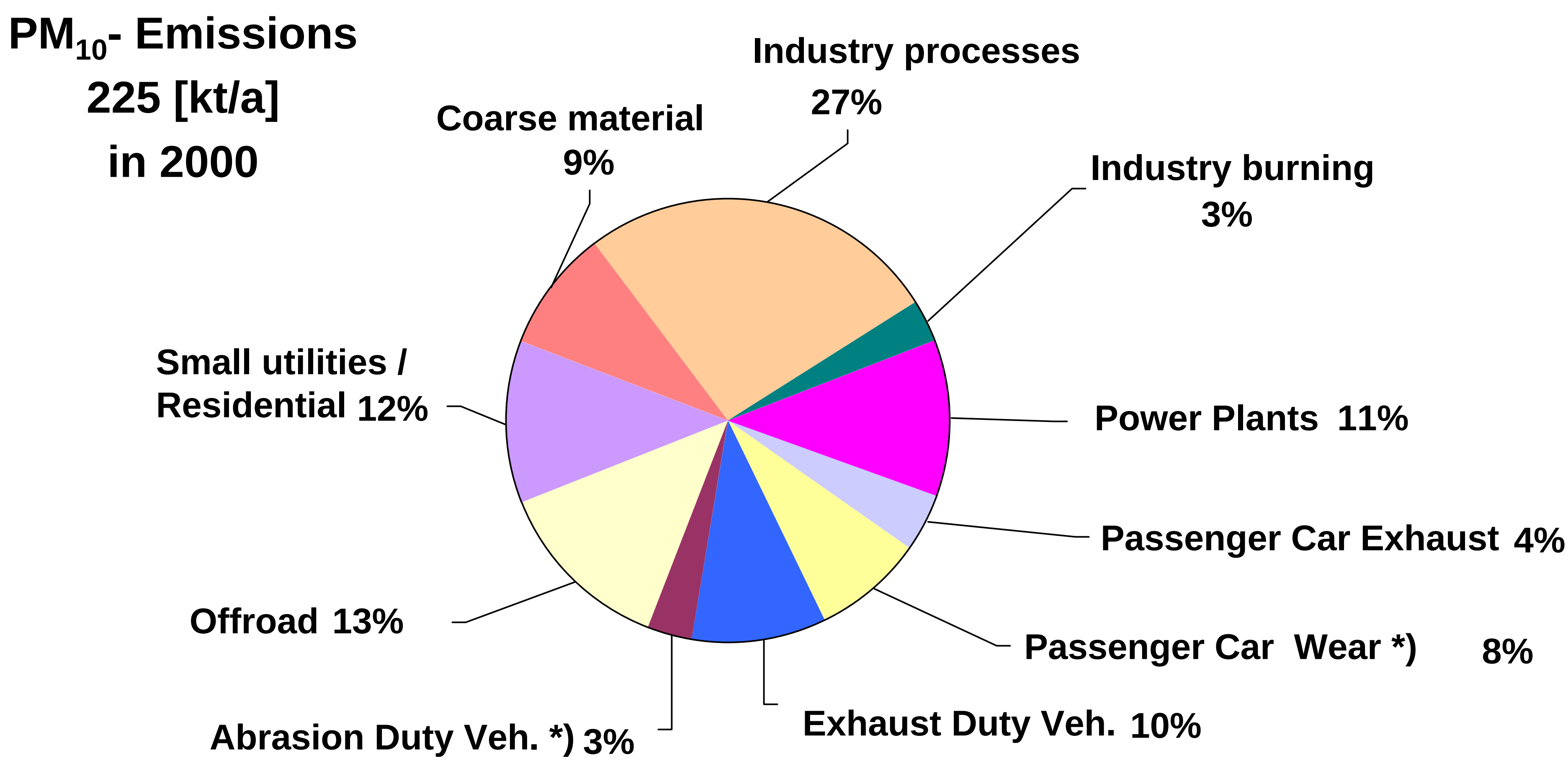
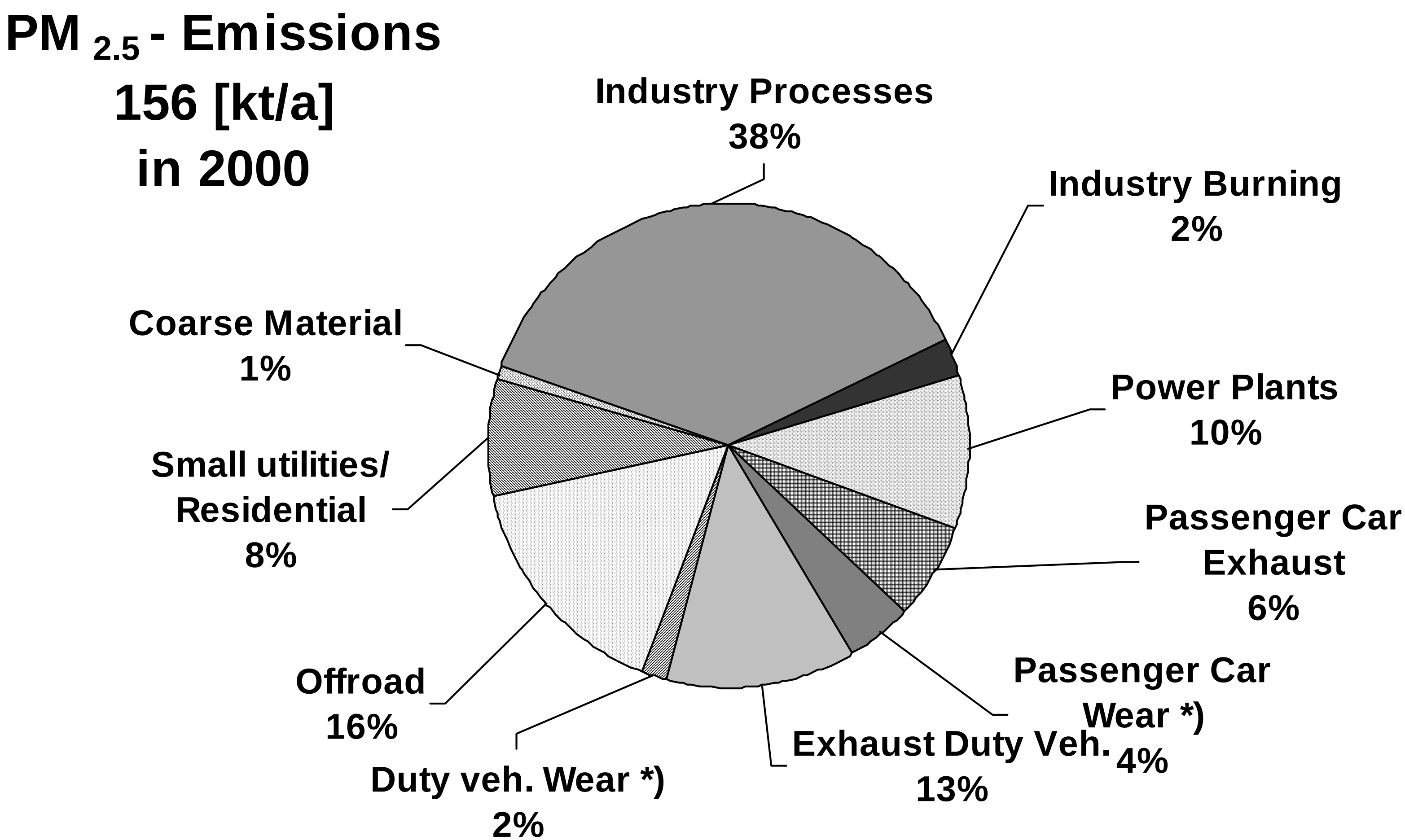
Q: Prüller, AK-EL
Wien, Juni 2002

Contribution of Different Sources to PM_{10} Emissions in Germany in 2000



*) Wear: Tyre, Brake, Road

Contribution of Sources to $PM_{2.5}$ Emissions in Germany in 2000



*) Wear: Tyre, Brake, Road

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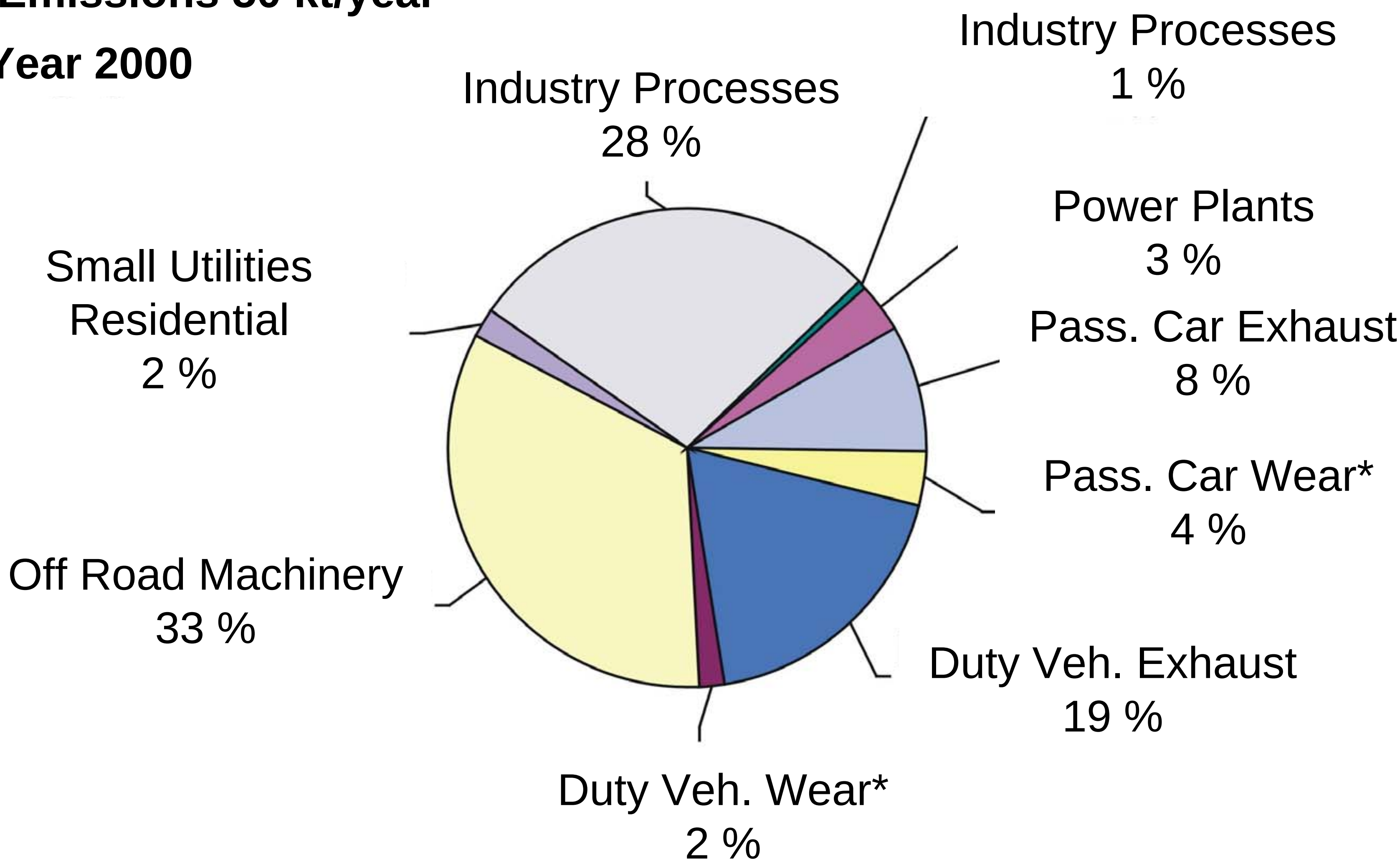
Me- 3816
Ref: Nader, TU Wien, 2002

Anthropogenic PM $_{0.1}$ Emissions in Germany in 2000

PM 0.1 Emissions 50 kt/year in the Year 2000



* Wear: Tyre, Brake, Road

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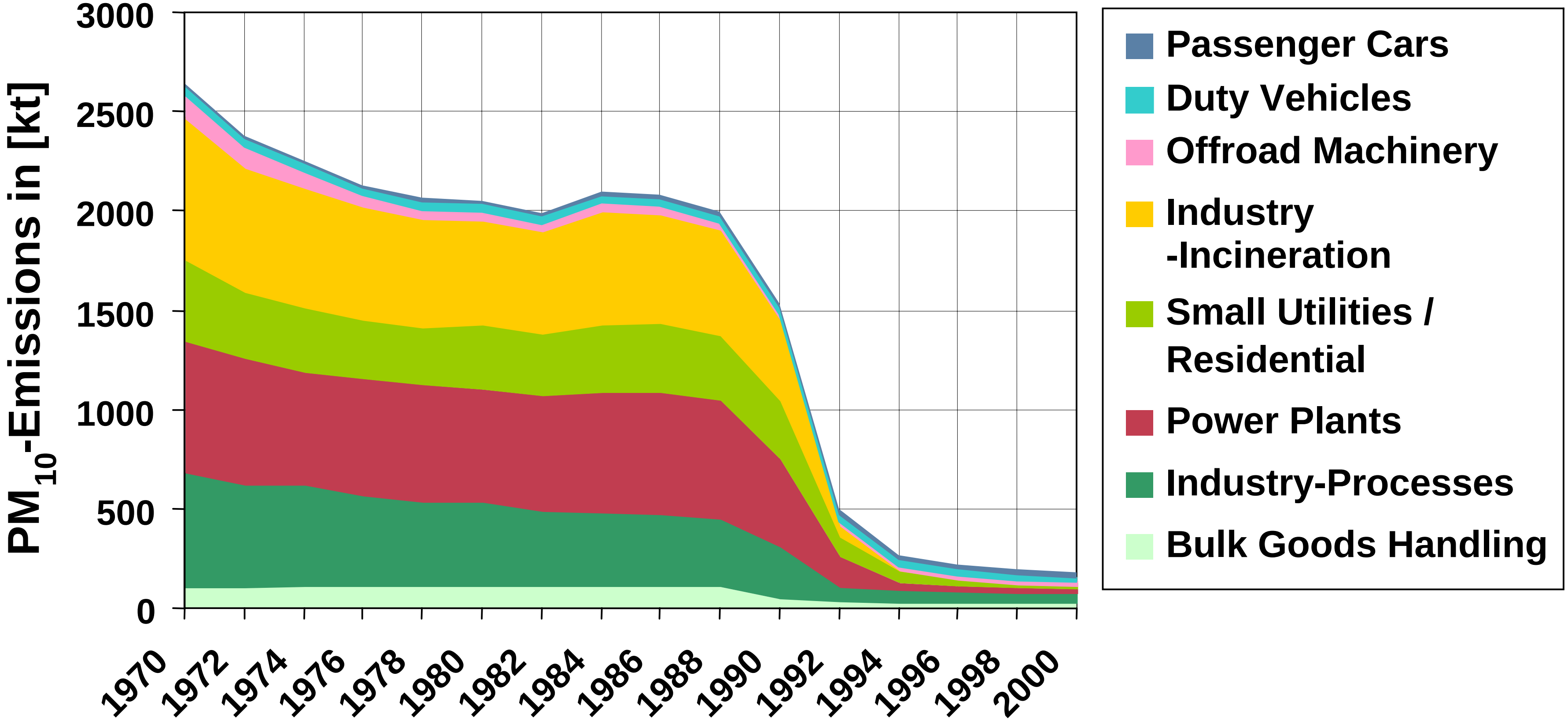
Me- 3817
Q: Prüller, AK-EL
Wien, Juni 2002

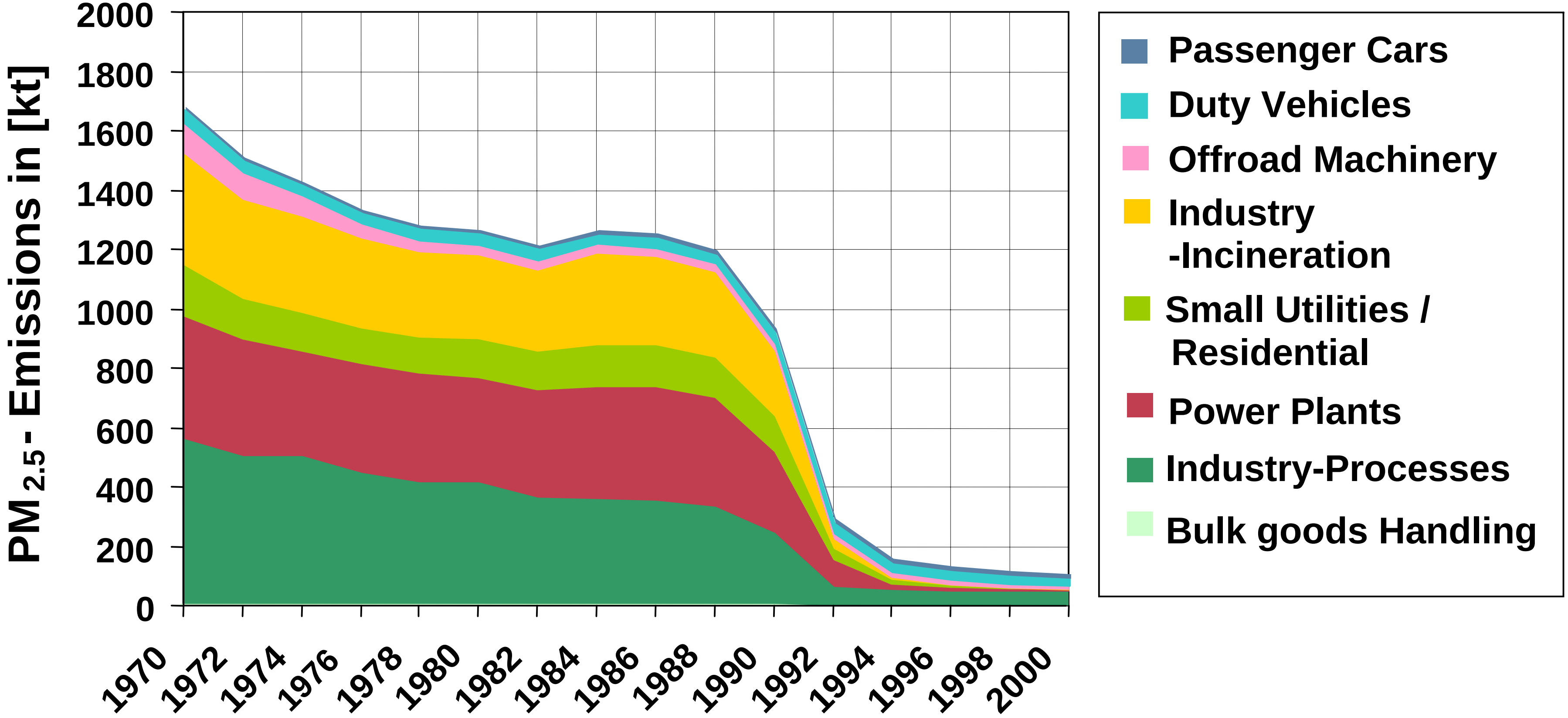
PM_{10} Emission Trend
Sources in Germany



(inclusive emissions of Road Transport due to Wear (Tyres, Brakes and Road))

$PM_{2.5}$ Emission Trend

Sources in Germany



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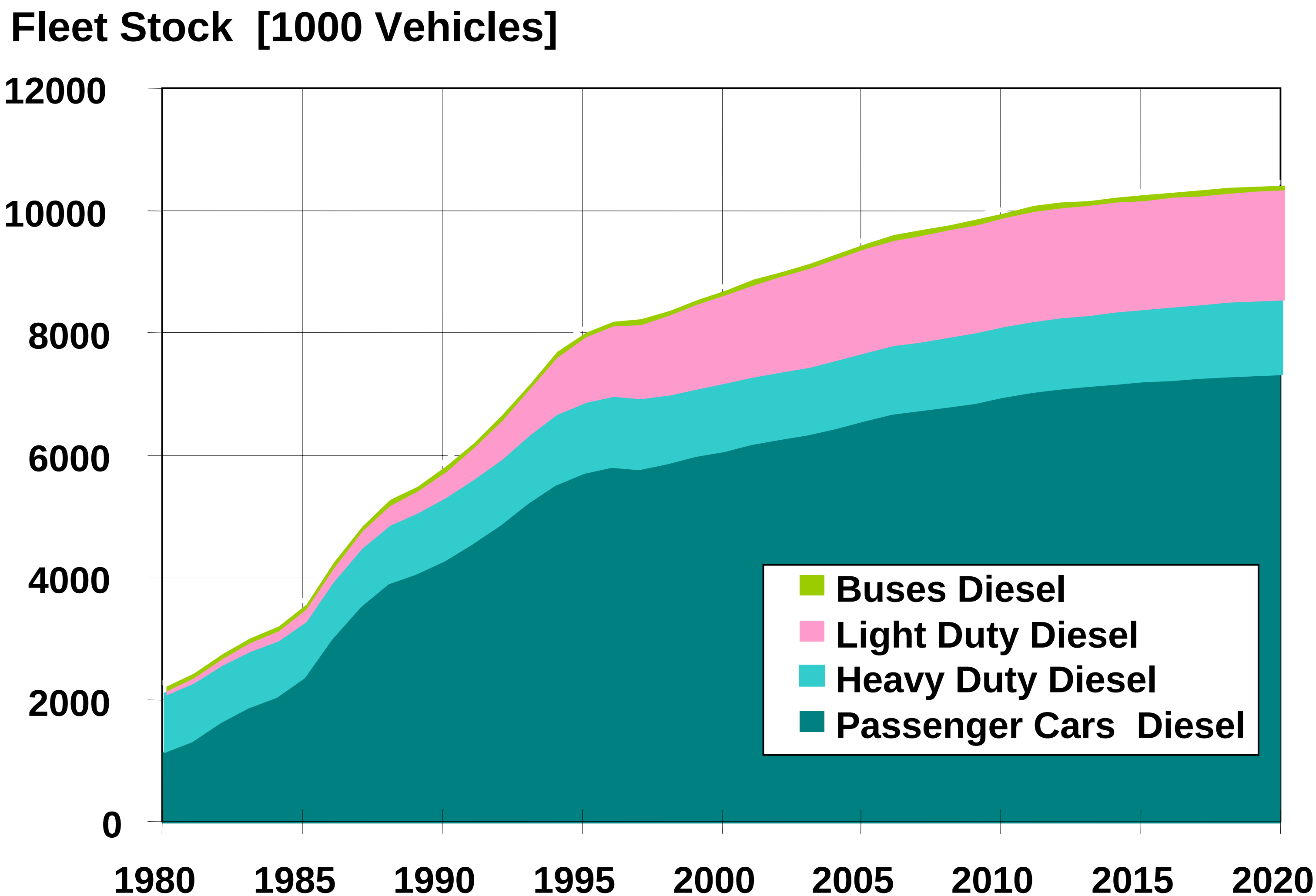
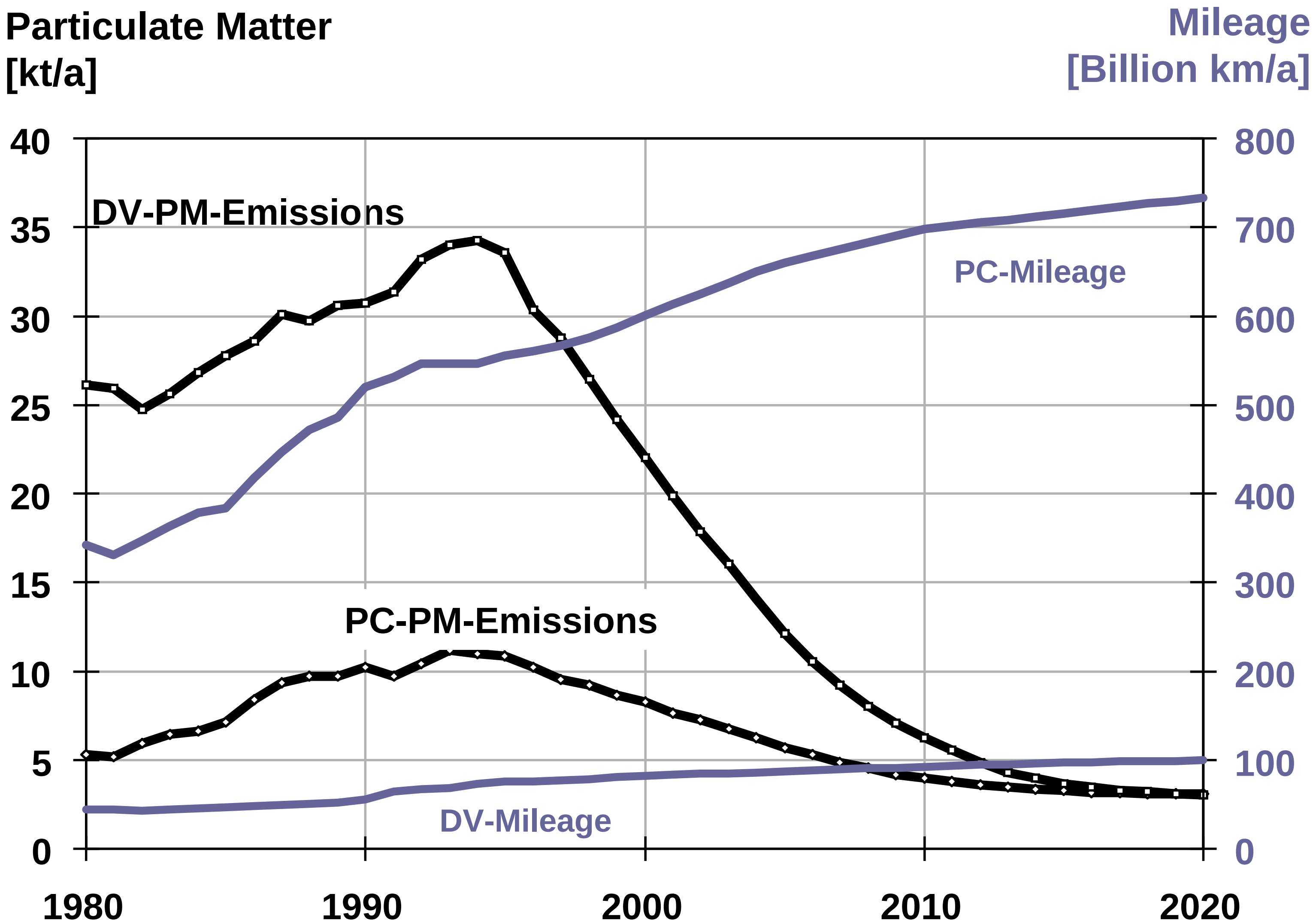
Q: Prüller S., TU Wien, AK EL 2002

Diesel Vehicles Trend in Germany

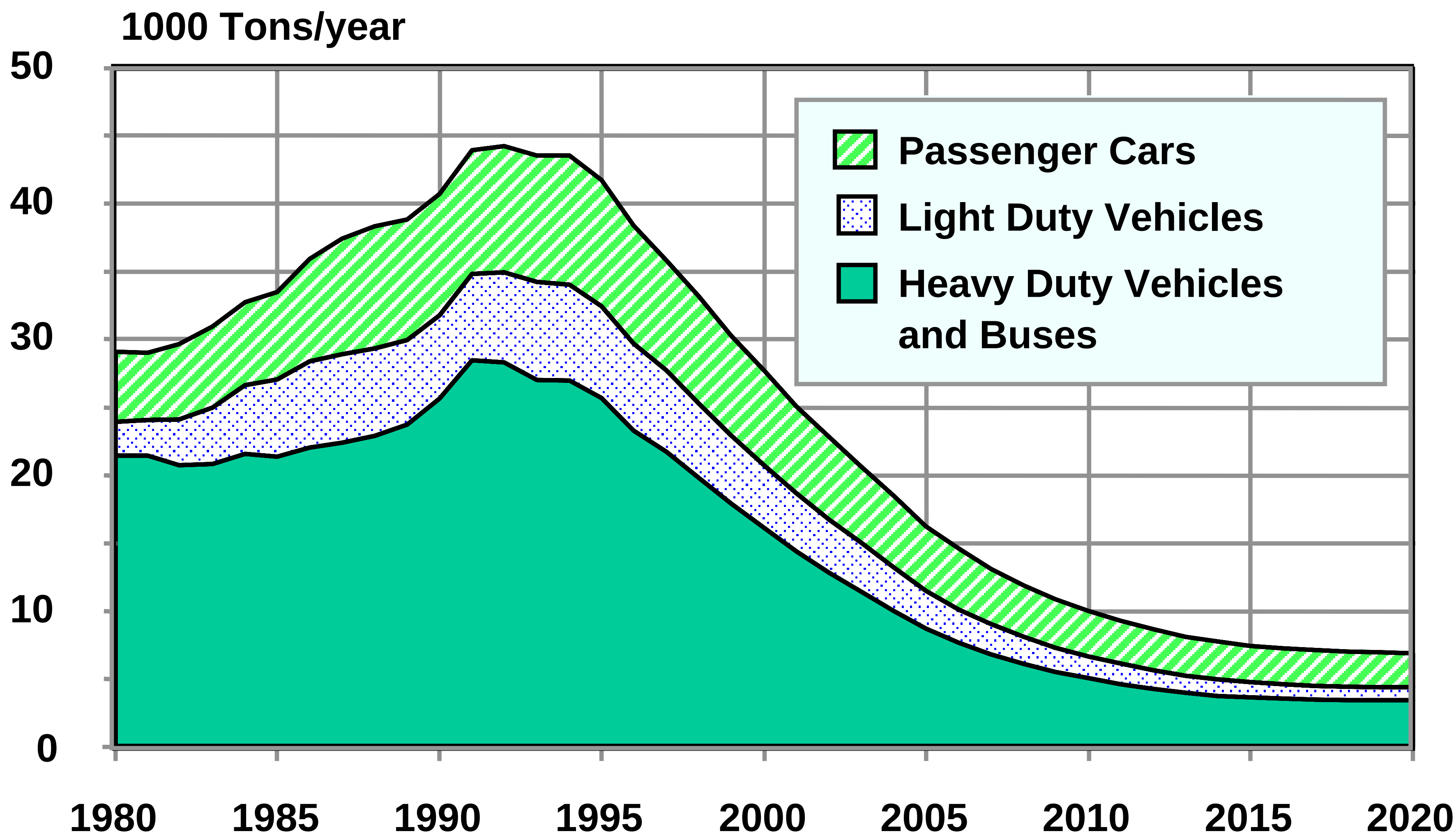


Particulate Matter Exhaust Emissions

Passenger Cars and Duty Vehicles in Germany



Development of PM Emissions Of Road Transport in Germany



Quelle: IFEU Heidelberg 03.09.1998

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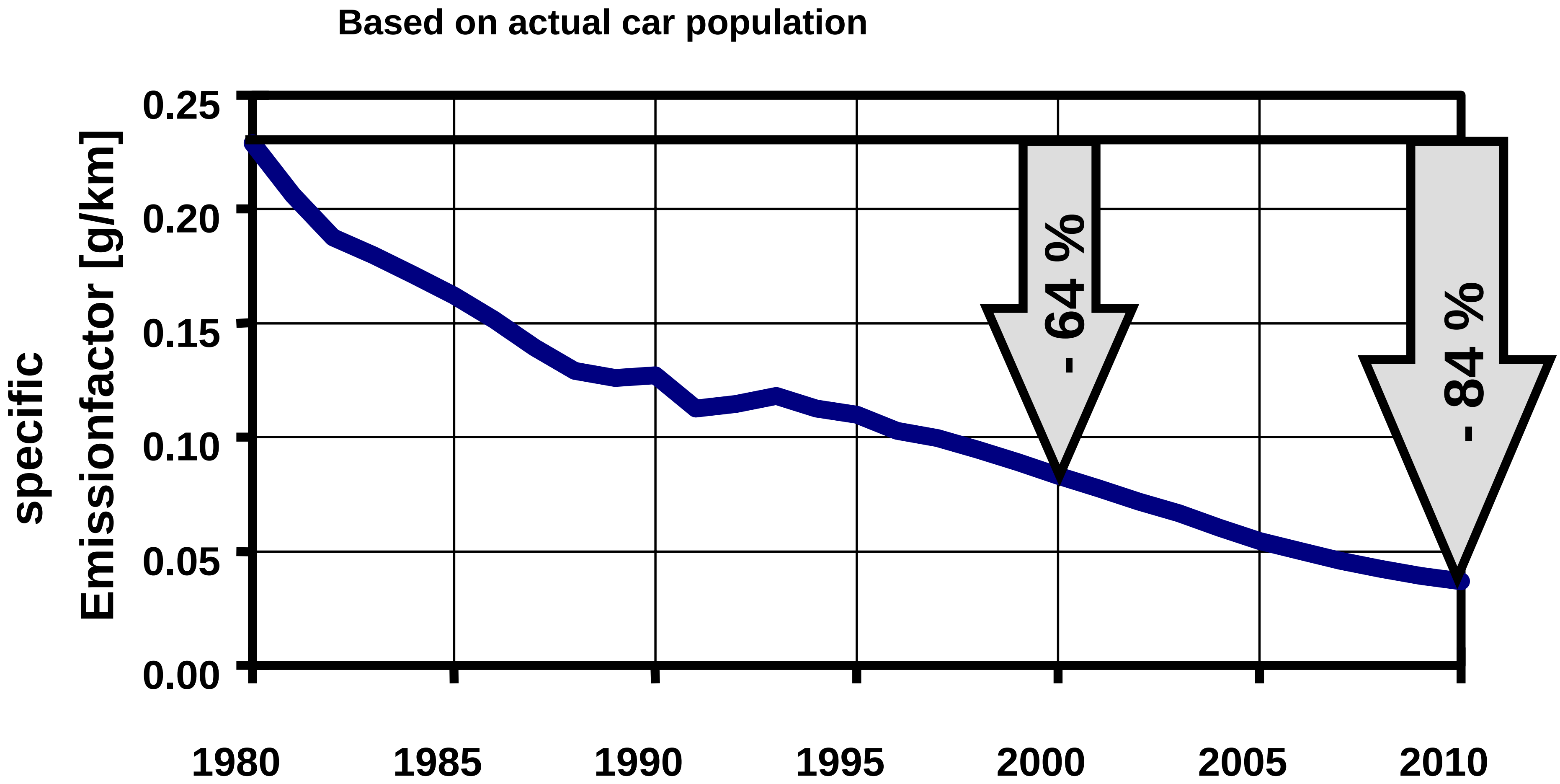
Q: IFEU aus
CHC-9008.ppt

Development of Diesel Passenger Car Fleet Emission Factors in Germany



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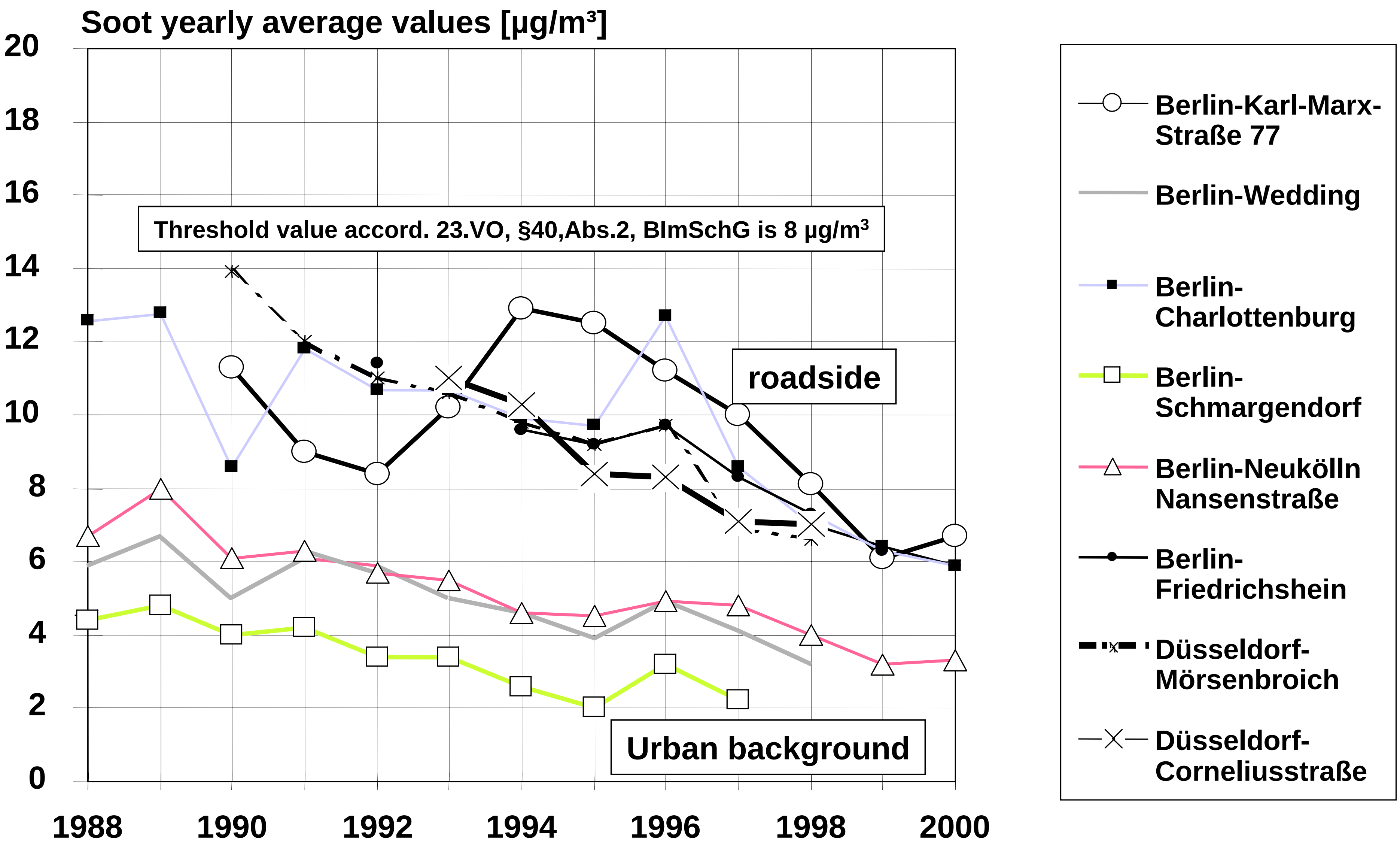
Soot (EC) Air Quality Trend In Some German Cities



aus H.P. Lenz, Ch. Cozzarini , S. Prüller, Bericht Emissionen und Luftqualität 1998, Stuttgart, 25.6.1998

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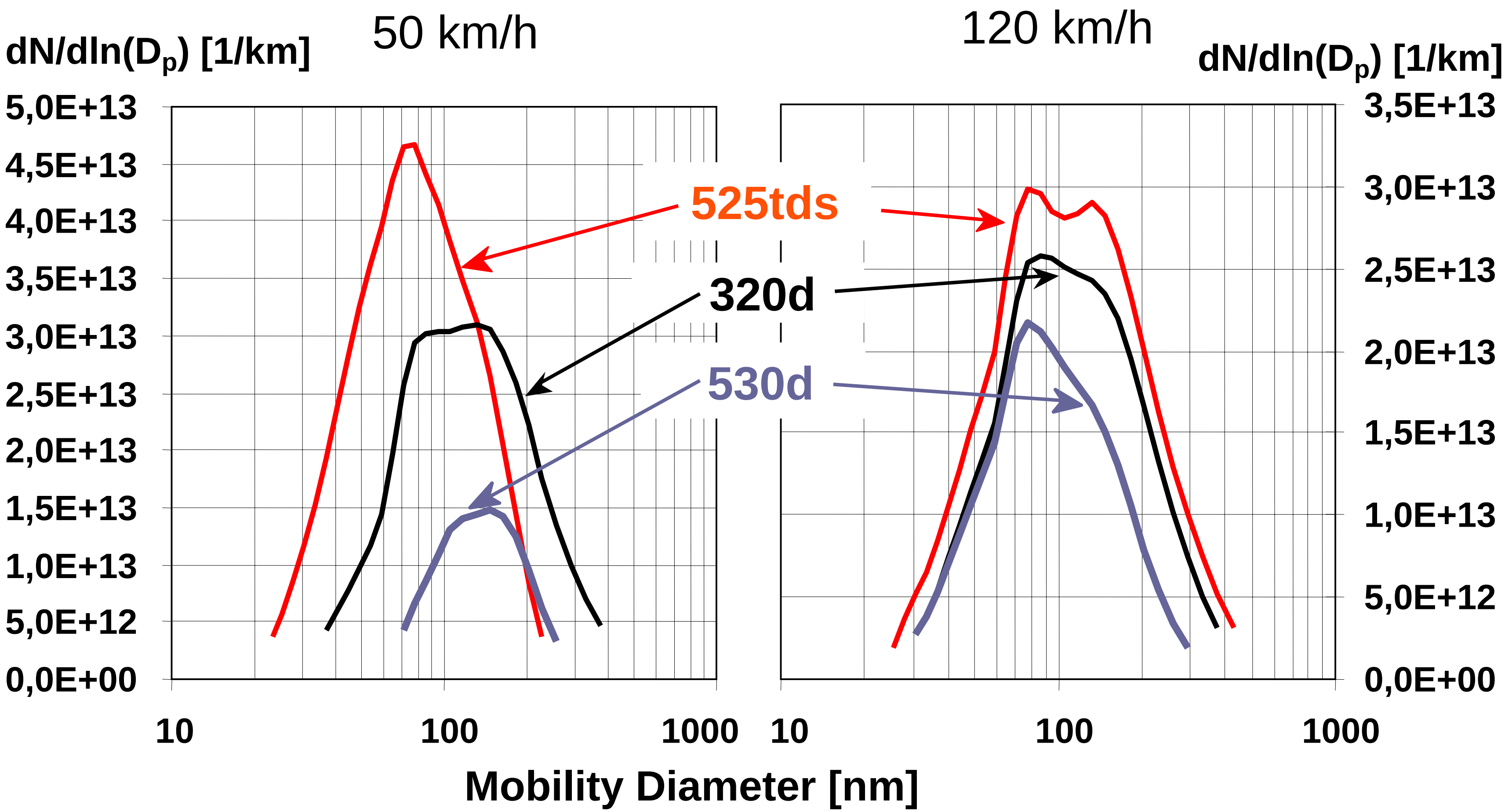
Q: Resch, Steinparzer F. ATZ

Size Distribution Of Exhaust Gas Particles

50 km/h And 120 km/h Constant Speed



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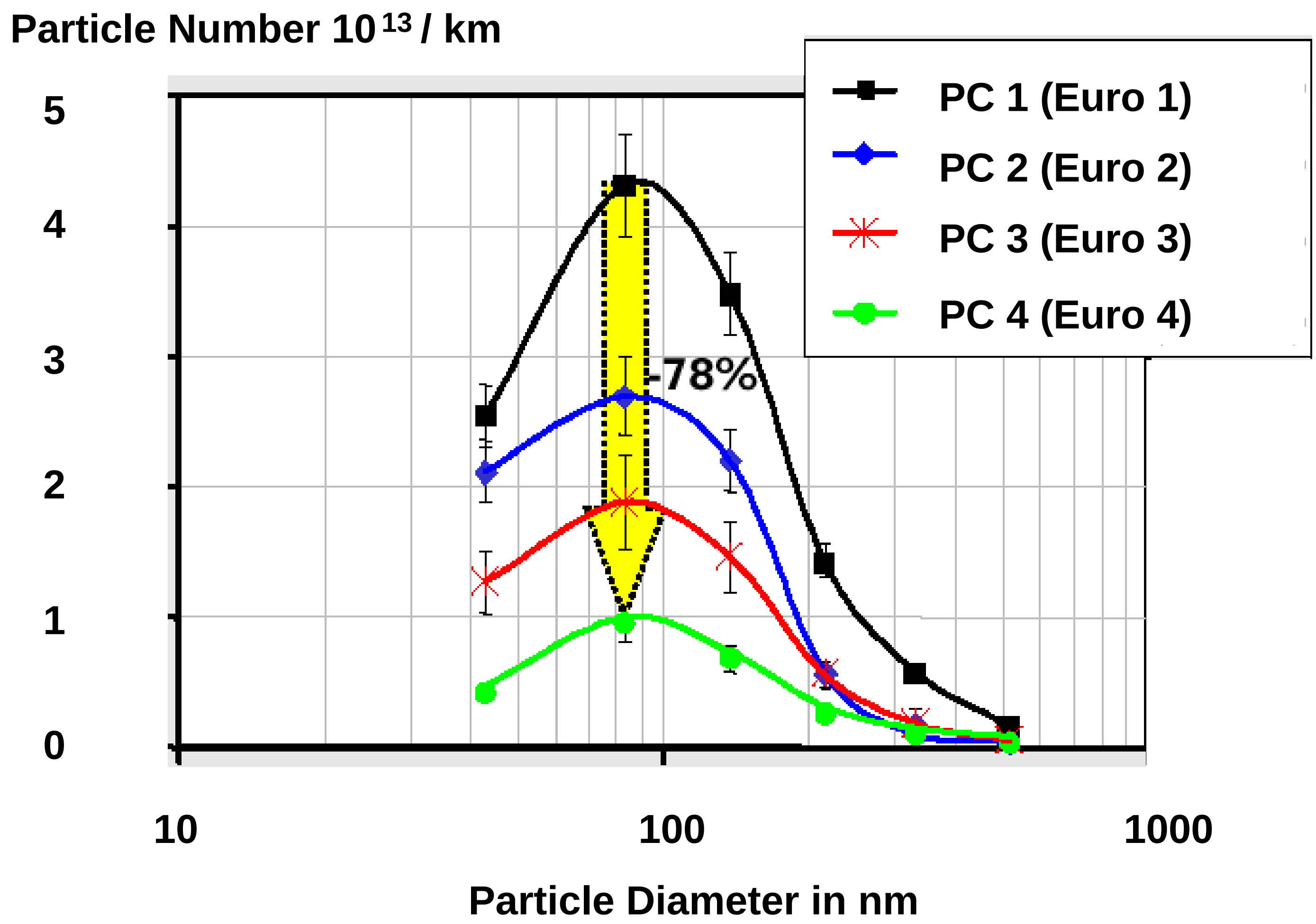
Q: Schindler.
VDA-Kongress
Frankfurt,2000

Particle Emission Reduction Of Diesel Passenger Cars In Mass And Number



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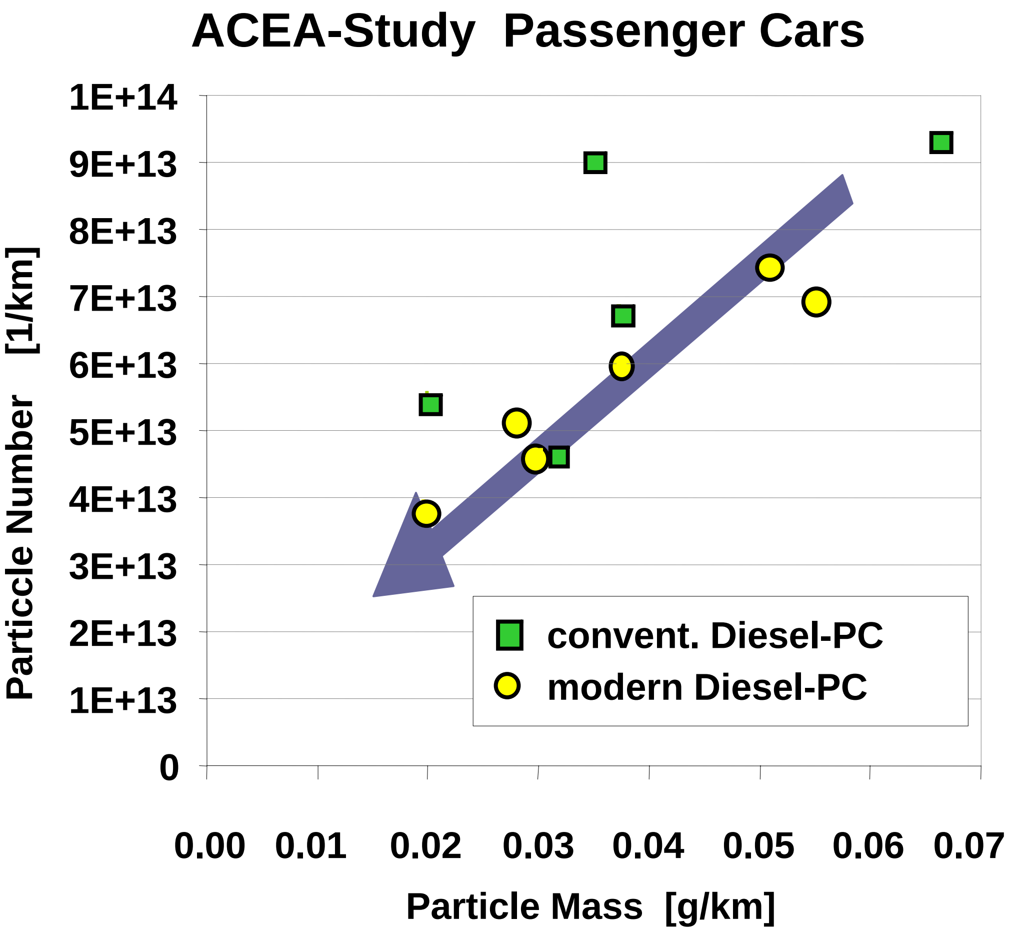
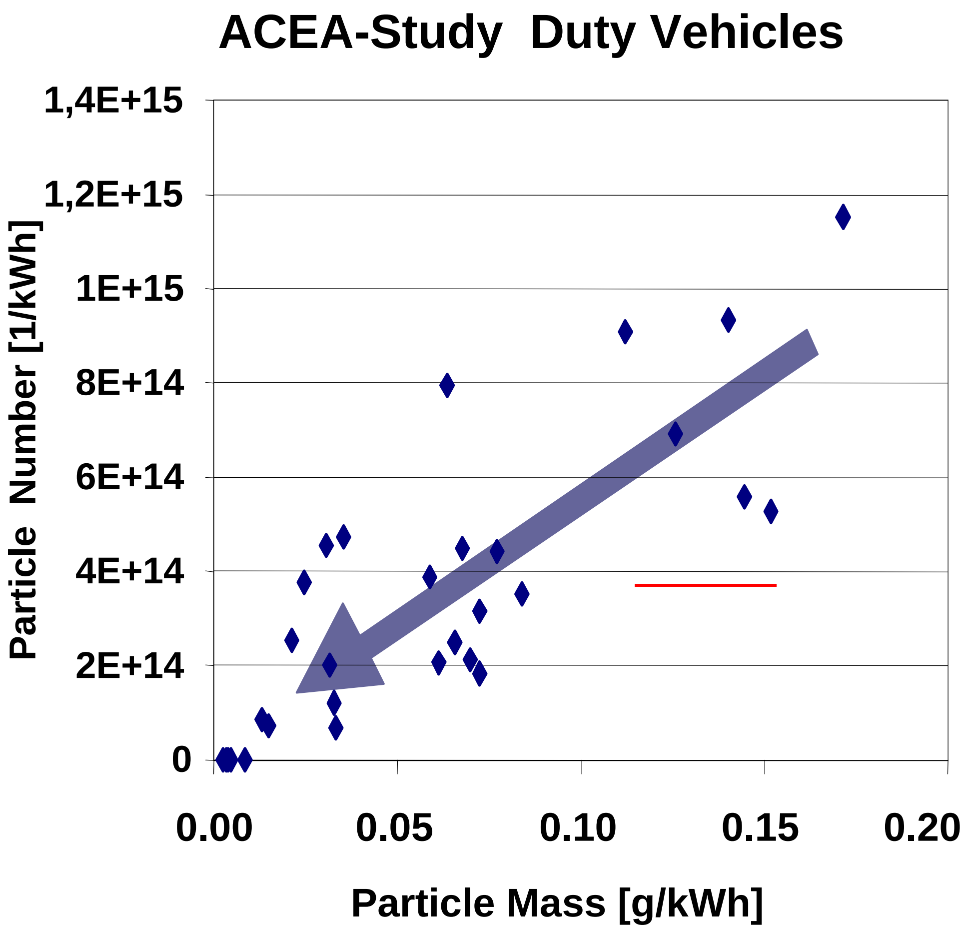
Q: ACEA, Rep.Fine Part.
Brüssel 1999, Stein J. VDA-
Techn.Kongr.9/2000,Frankf.

Reduction Of Particle Emissions

Mass And Number Of Diesel Vehicles





ACEA-Programm zur Partikelemission von Pkw
gemessen mit SMPS bei 100 km/h Konstantfahrt

ACEA-Programm zur Partikelemission von Nfz
gemessen mit SMPS bei Konstantlast

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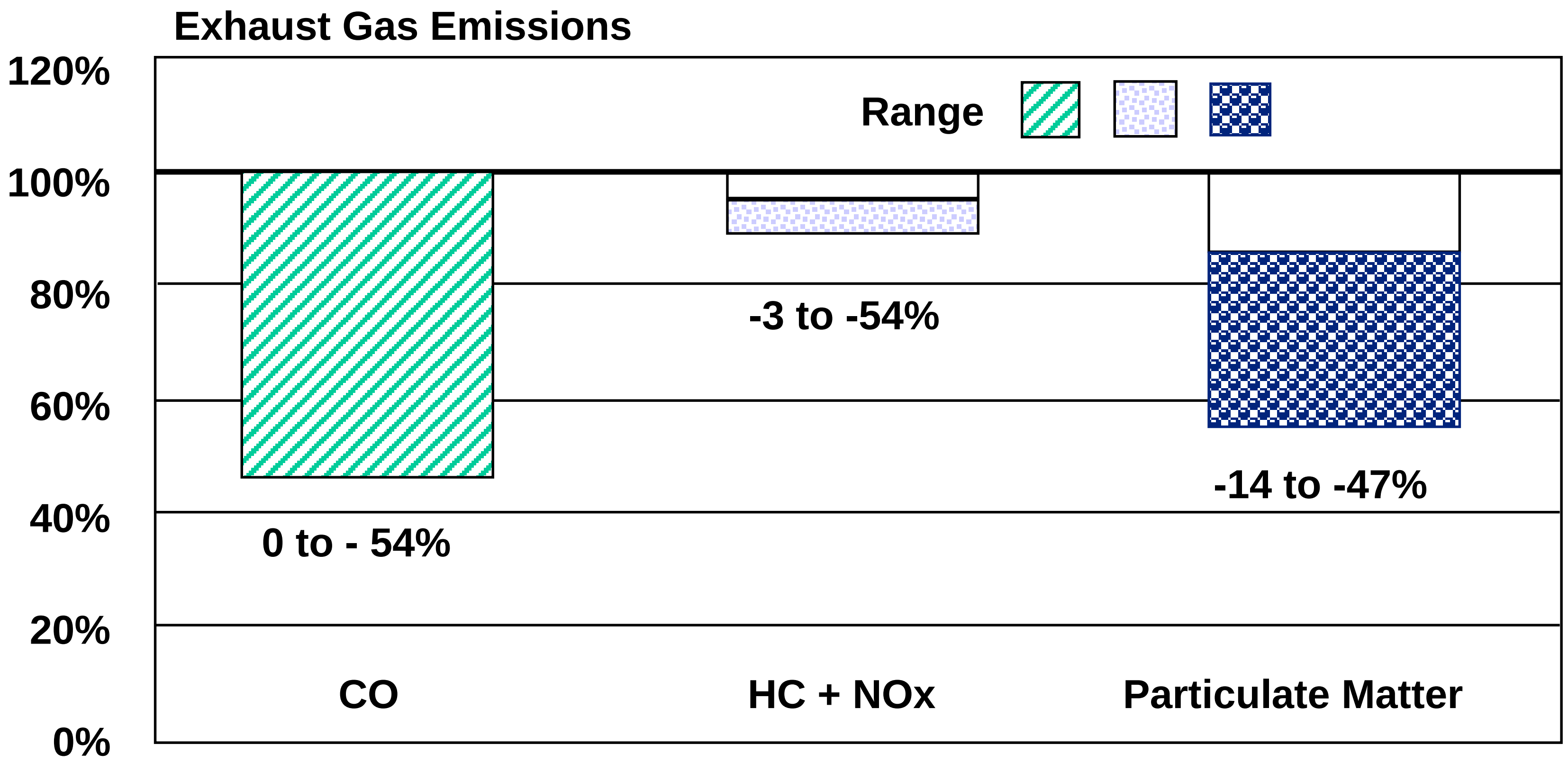
Ref: Schindler
VDA-Workshop 3.6.98

Passenger Car Emission Improvements By Reducing Sulphur Content In Diesel Fuel From 350ppm to 10ppm

Measurement on different vehicles (Audi, BMW, DC, VW) in Europe-driving cycle

Basis: Normal Diesel Fuel = 100% compared with Swedish Diesel Fuel Class 1



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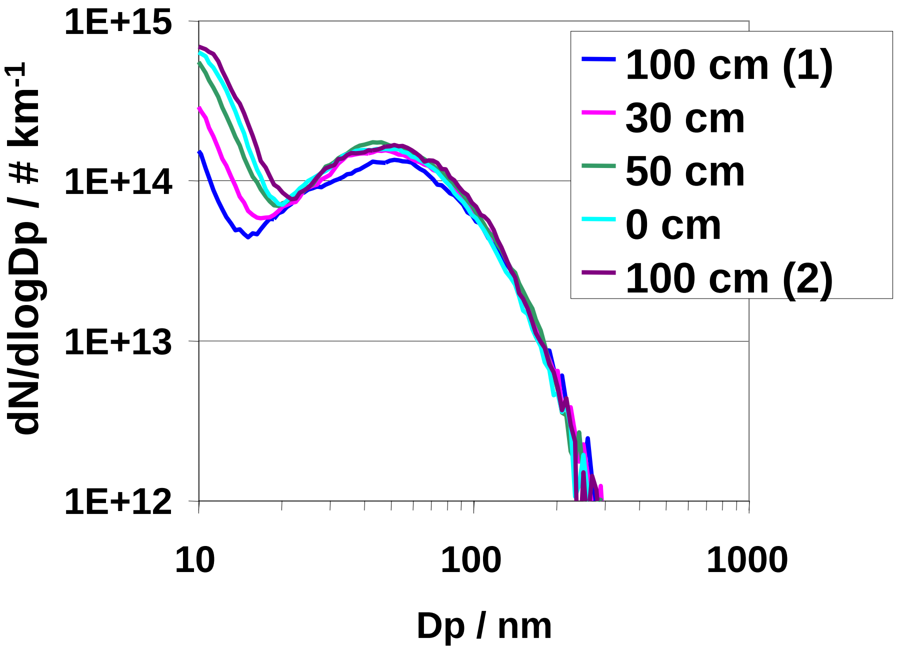
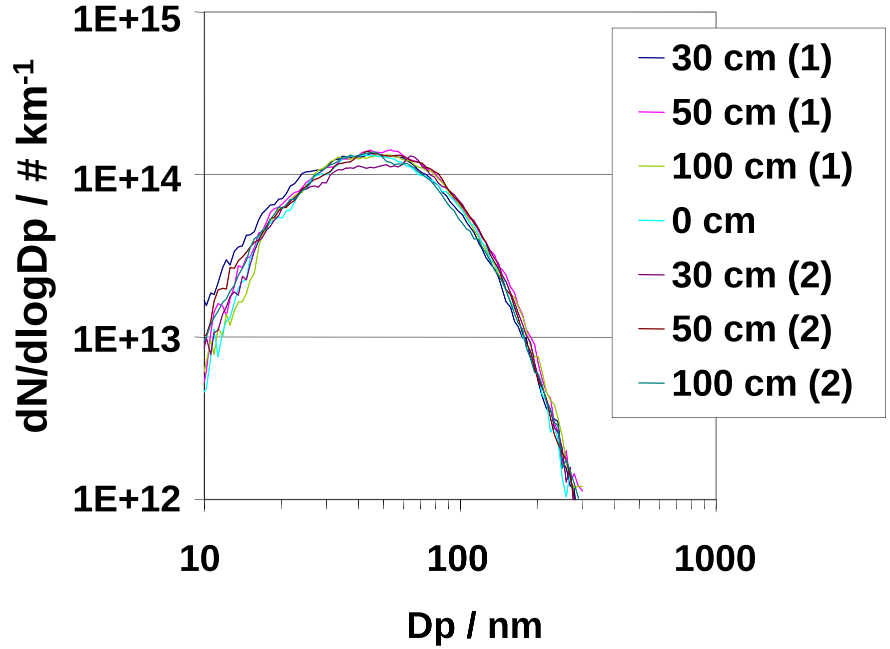
Q:Vogt R., ETH-Nanopart-Conference, Zürich, 2002

Typical Diesel Particle Size Distribution With 320 ppm Sulfur And With 40 ppm Sulfur

Diesel car, 320 ppm S fuel, speed 100 km/h, distance 14 m, T=21°C, RH=53-59%



Diesel car, 40 ppm S fuel, speed 100 km/h, distance 14 m, T=17-20°C, RH=56-80%



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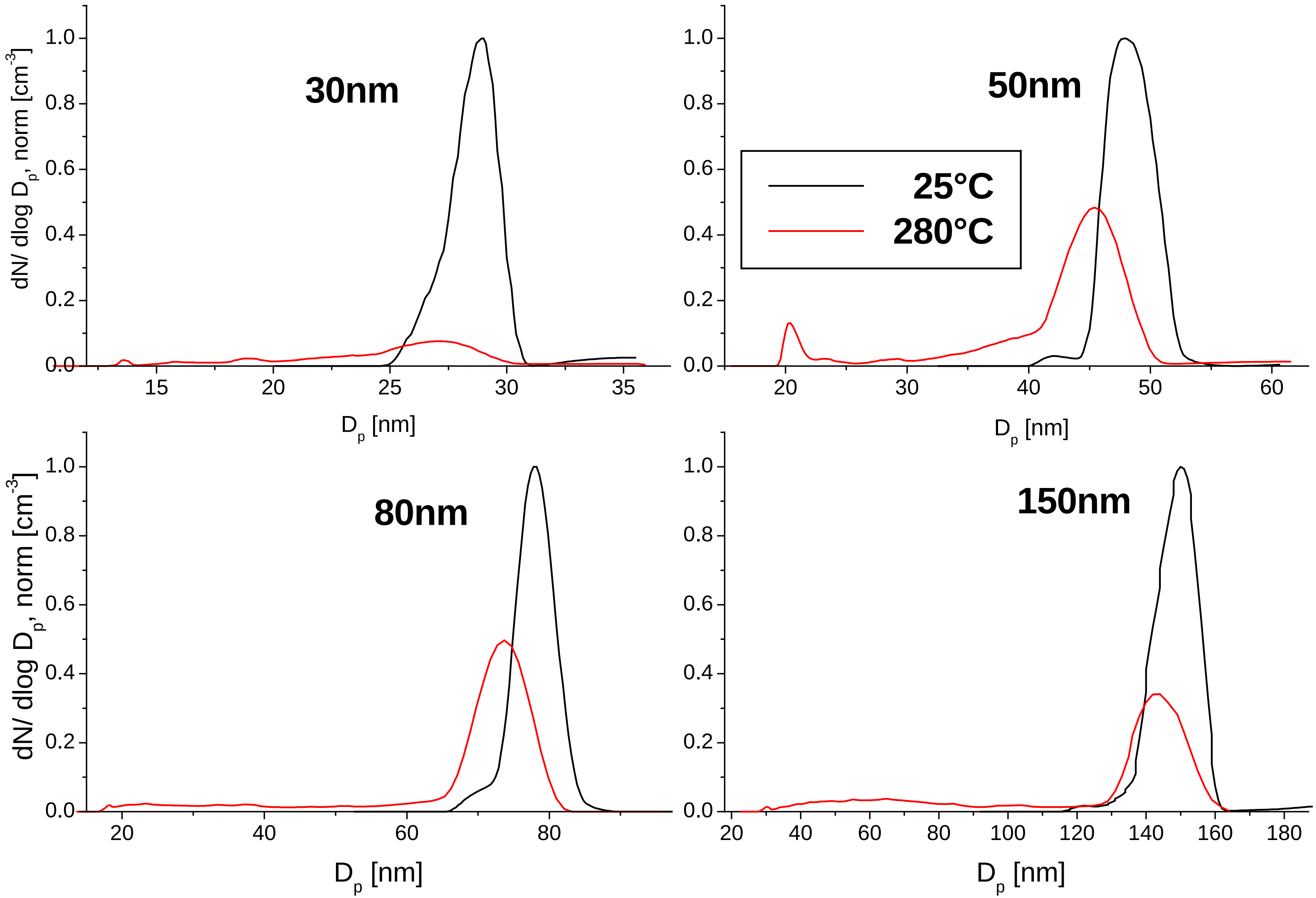
Ref.: Wehner, B., Wiedensohler A. 2001

Preliminary Highway Sampling Results For Some Size Fractions In Germany



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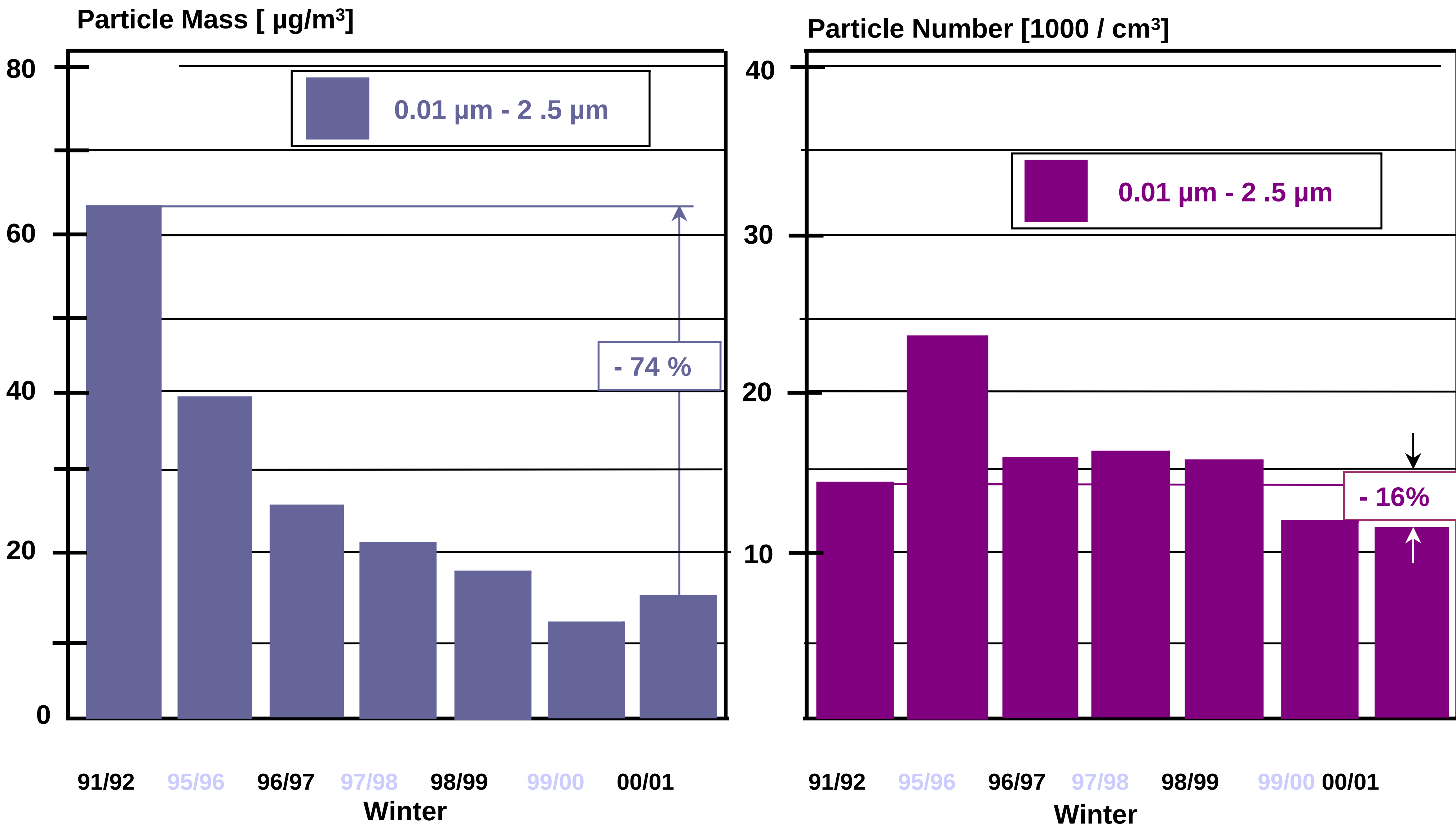
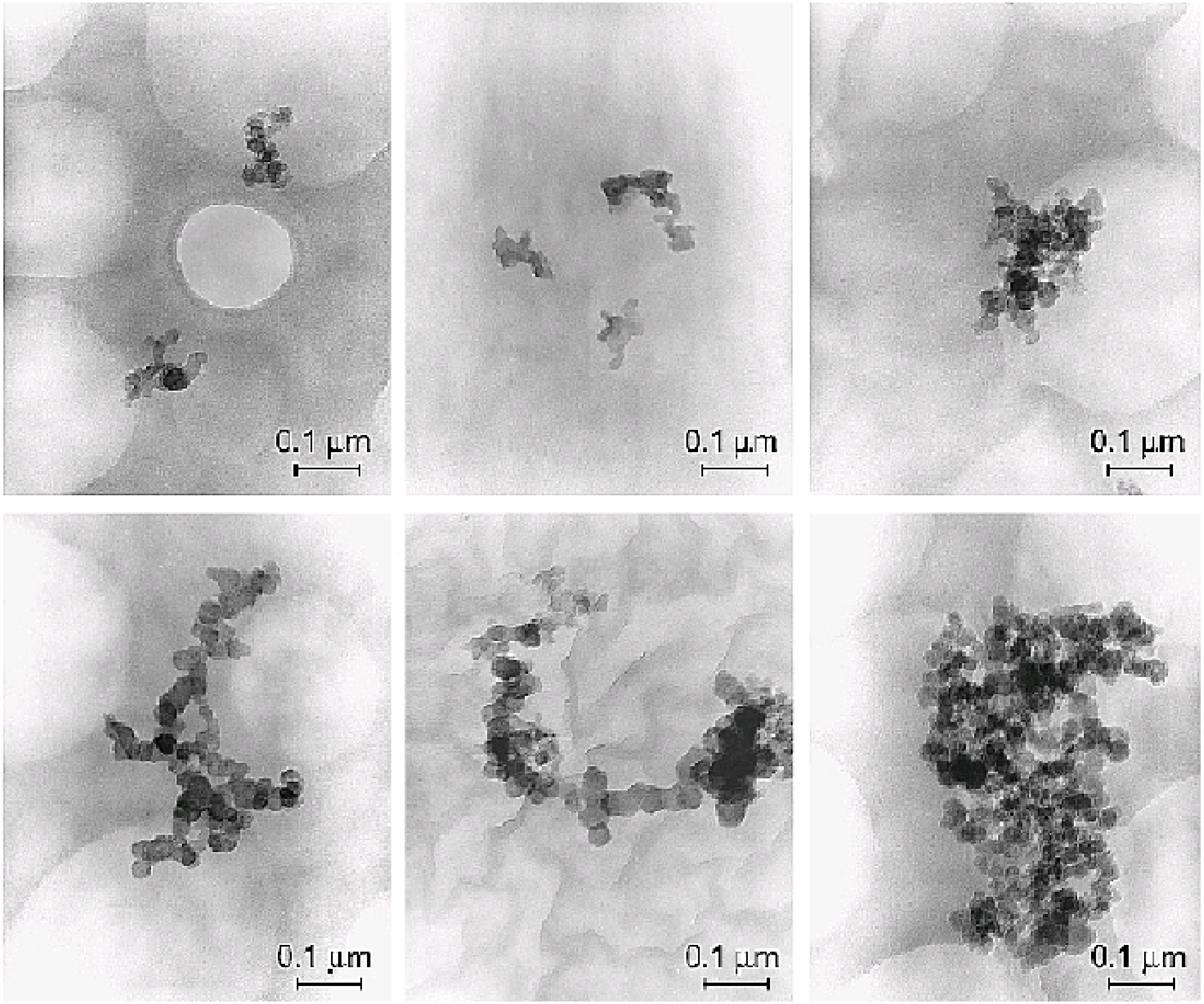
Mass And Number Of 2.5 Particles
In Urban Ambient Air In Erfurt

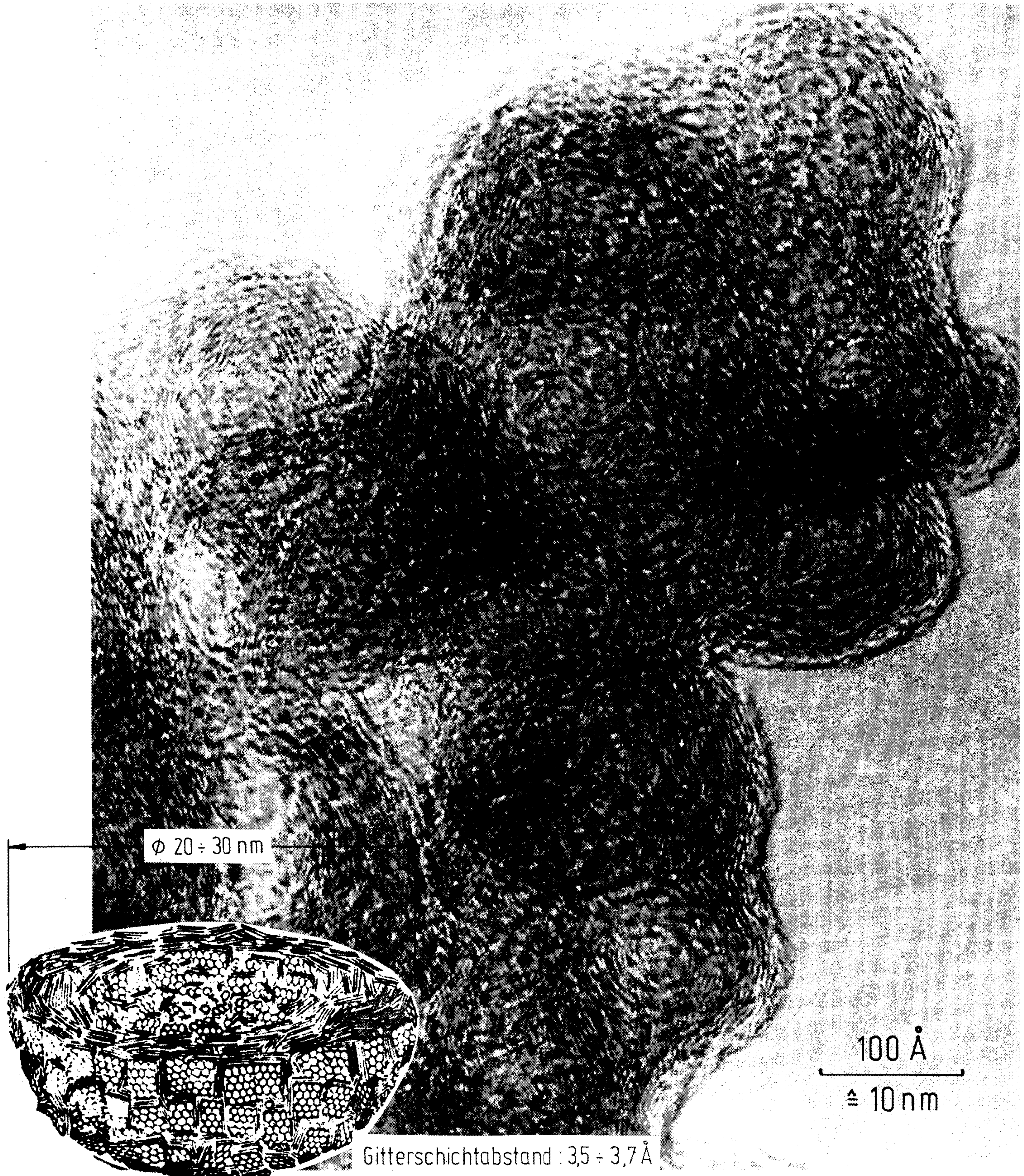


Ref: Kreyling W.G. et al, Atmospheric Environm. 7/2003 in press

Dieselparticles Measured With Transmission Electromicroscopy



Diesel Particle With High Resolution
Magnification 400 000 With Zeiss EM100



A typical aggregate particle consisting of several primary particles, which diameter lies between 20 and 30 nm. Primary particles are strong connected with each other.

In the lower left corner the schematic structure of a primary particle is shown. It consists out of many carbon layers packed together. The distance of the grids is about 0.36 nm or 3.6 Angström.

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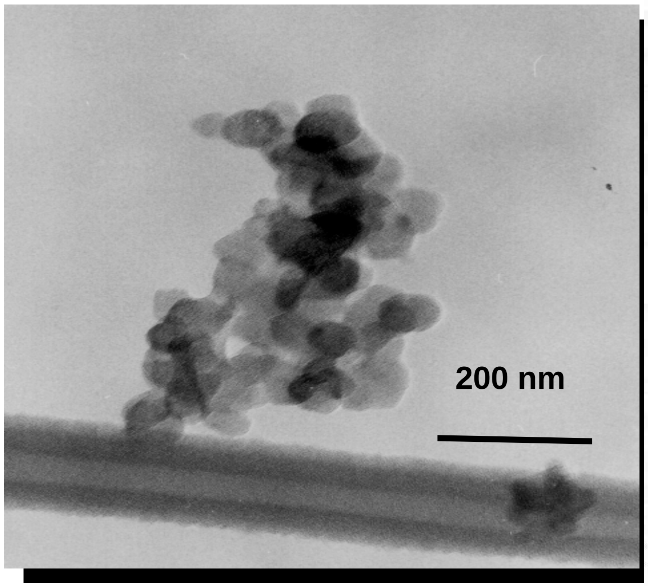
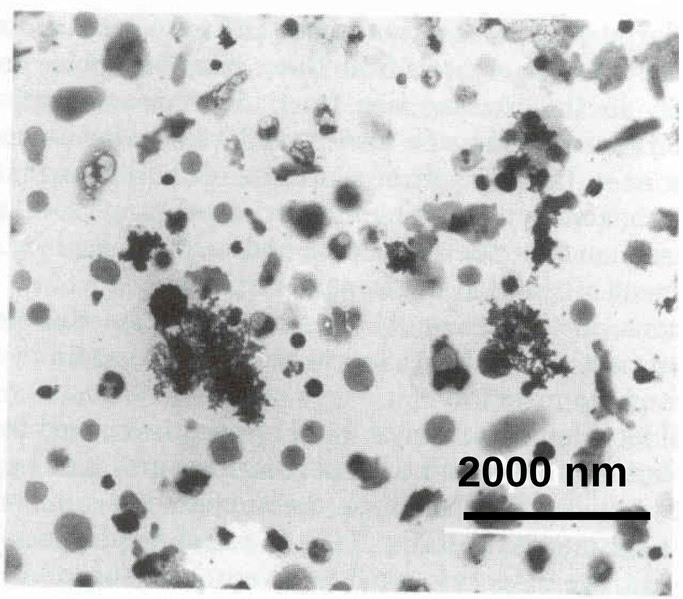
Q:Metz N., MTZ, et al. Richards R.J. in PM, Properties and Effects upon Health ed. B.Maynard a. Howell BIOS,1999, Page 111

Particle From A Diesel Engine, Particles In Ambient Air And Deposited In The Lung

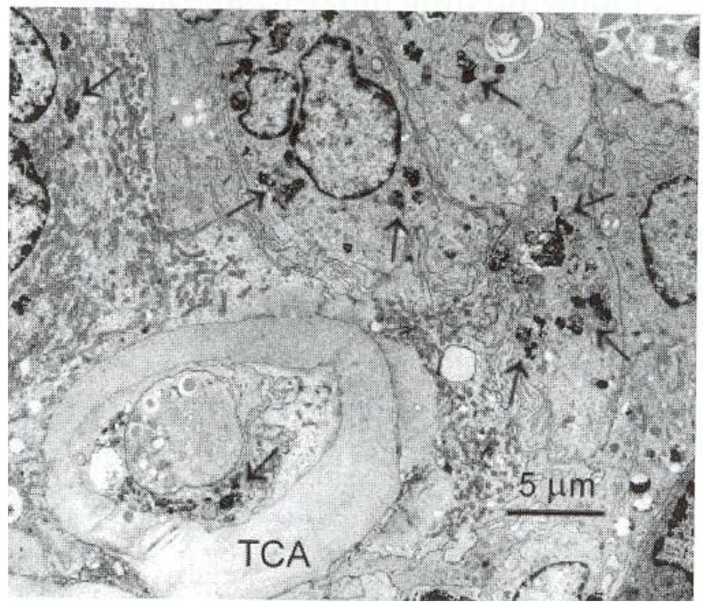
Diesel Engine, CR, 120km/h



Urban Aerosol, UK



DPM administered to rats, UK



On the way from the exhaust tailpipe into the ambient air and to the deposition in the respiratory system diesel particles tend to increase their diameter from 100-200 nm over 300-500 nm (ambient air) to 600-1000 m (alveols and interstitium)

Surface Of Diesel Aggregate Particles

The weight of particles of a Euro 3 passenger car is
0,04 g/km = 40 mg/km = 40 000 µg/km = 40 000 000 ng/km.

The weight of one aggregate particle is approximately
$580 * 10^{-9}$ ng.
This means that in the Exhaust there are

$$\frac{40\,000\,000 \text{ ng} * 1000\,000\,000}{580 \text{ ng} * \text{km}}$$

$= 69 * 10^{12}$ Particles /km with a Surface of 81 500 nm^2 /Particle

Surface Euro 3 = $69 * 81{,}4 * 10^{15} = 5.6\ m^2/km$.

The Surface of Particles from Euro 4 Passenger Cars is 2.8 m^2/km.

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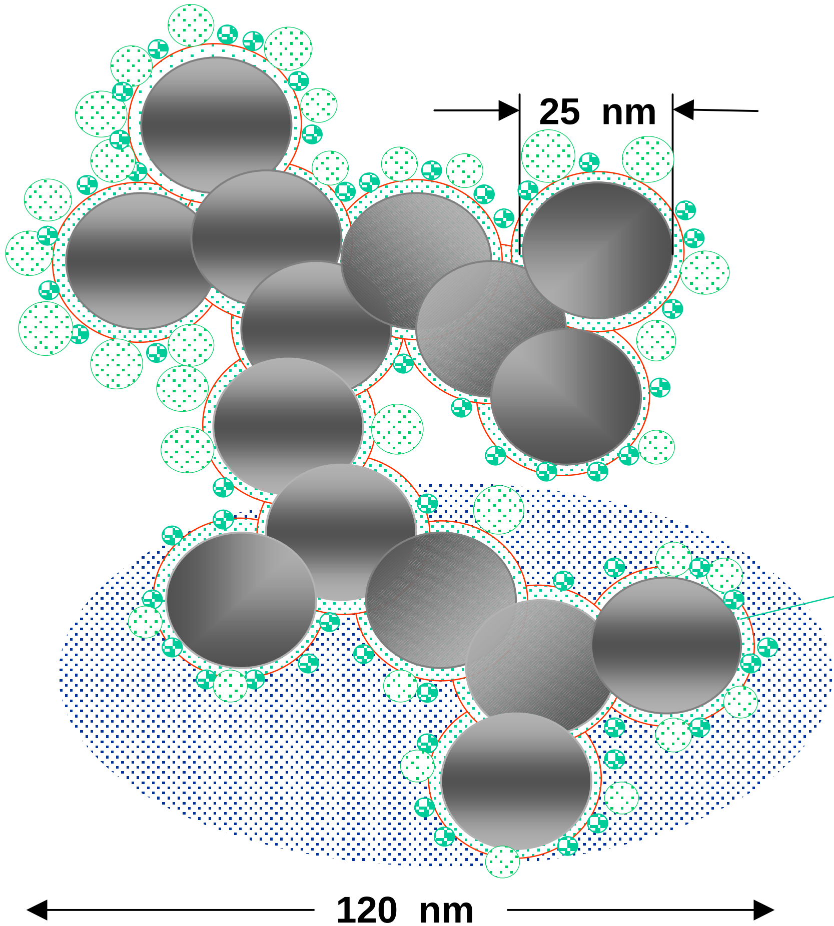
Ref: Greenbaum D., 2/2003

Aggregate Particle Composition Of A Modern PC Diesel Engine



Aggregate Particle Diameter (AD) = 120 nm

Primary Particle Diameter (PD) = 25 nm

EC = Elemental Carbon

Sulfate SO_4

Soluble organic HC-Fraction OC

Gaseous HC

Adsorbed HC

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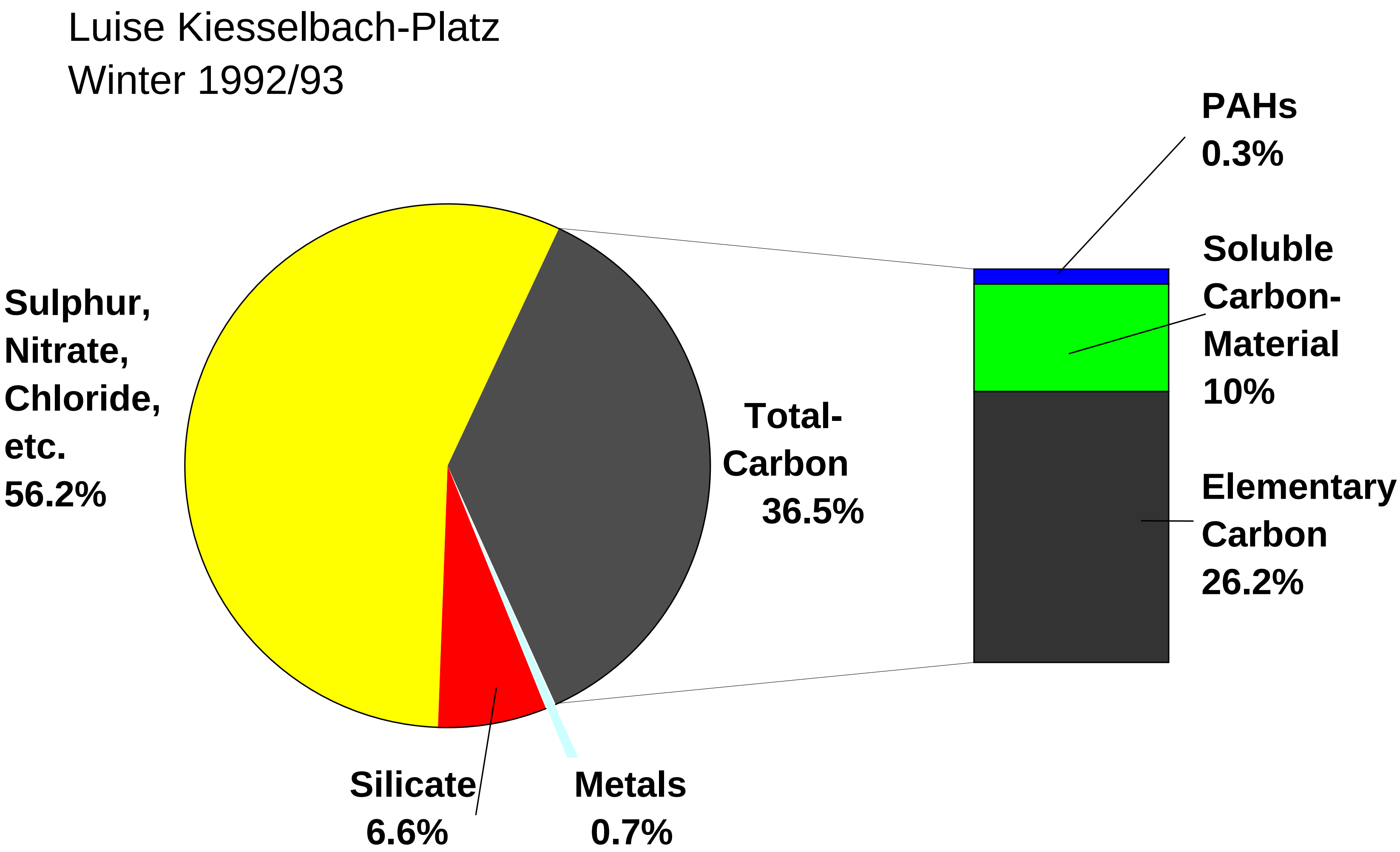
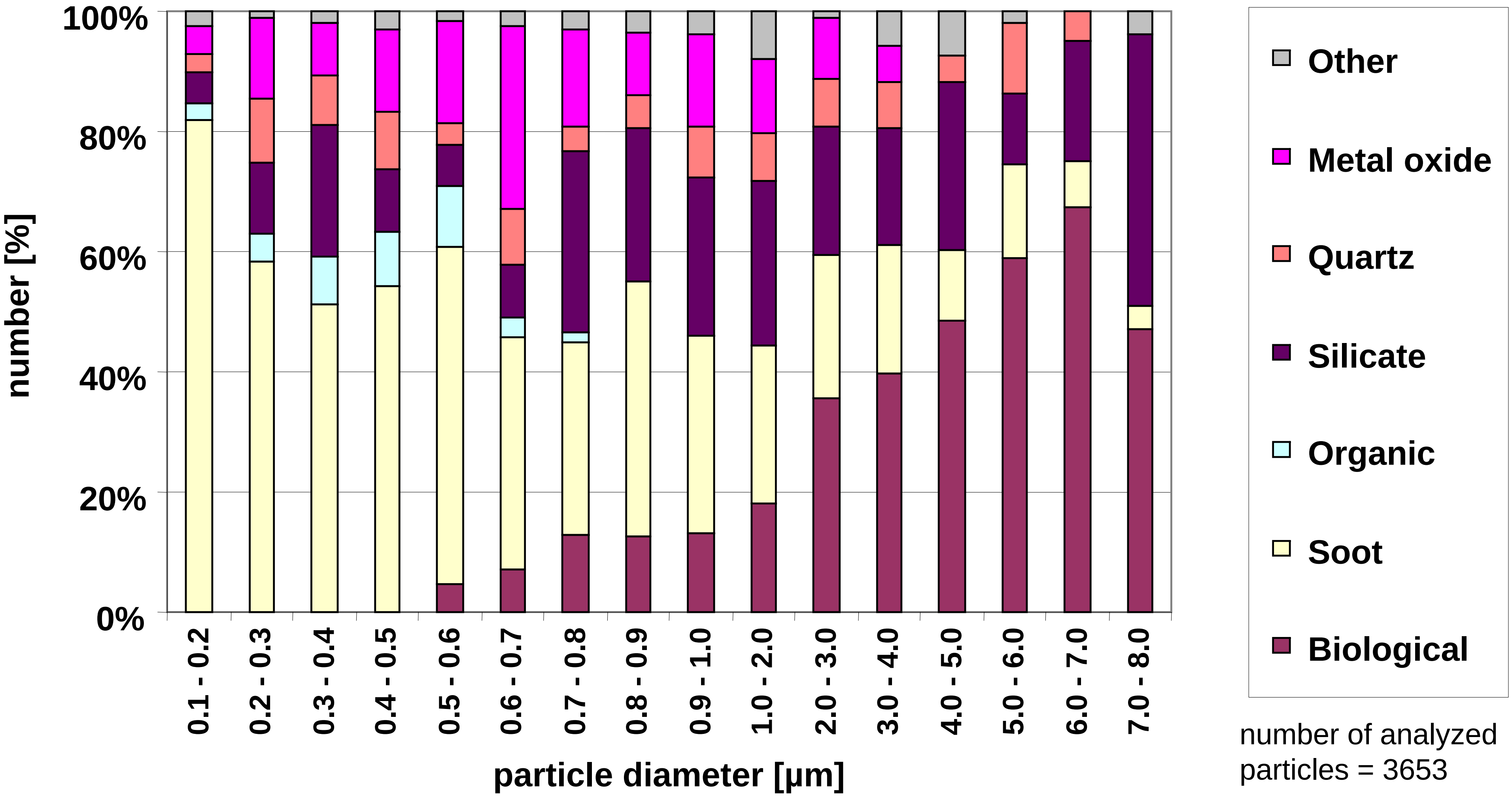
me-3836
Q: Niessner R.
München 1995

Composition Of Particulate Matter Near Kerbside In Munich, Germany



Relative Abundance Of Particle Groups At Black Forest „Kleiner Feldberg“ (13.08.1997)

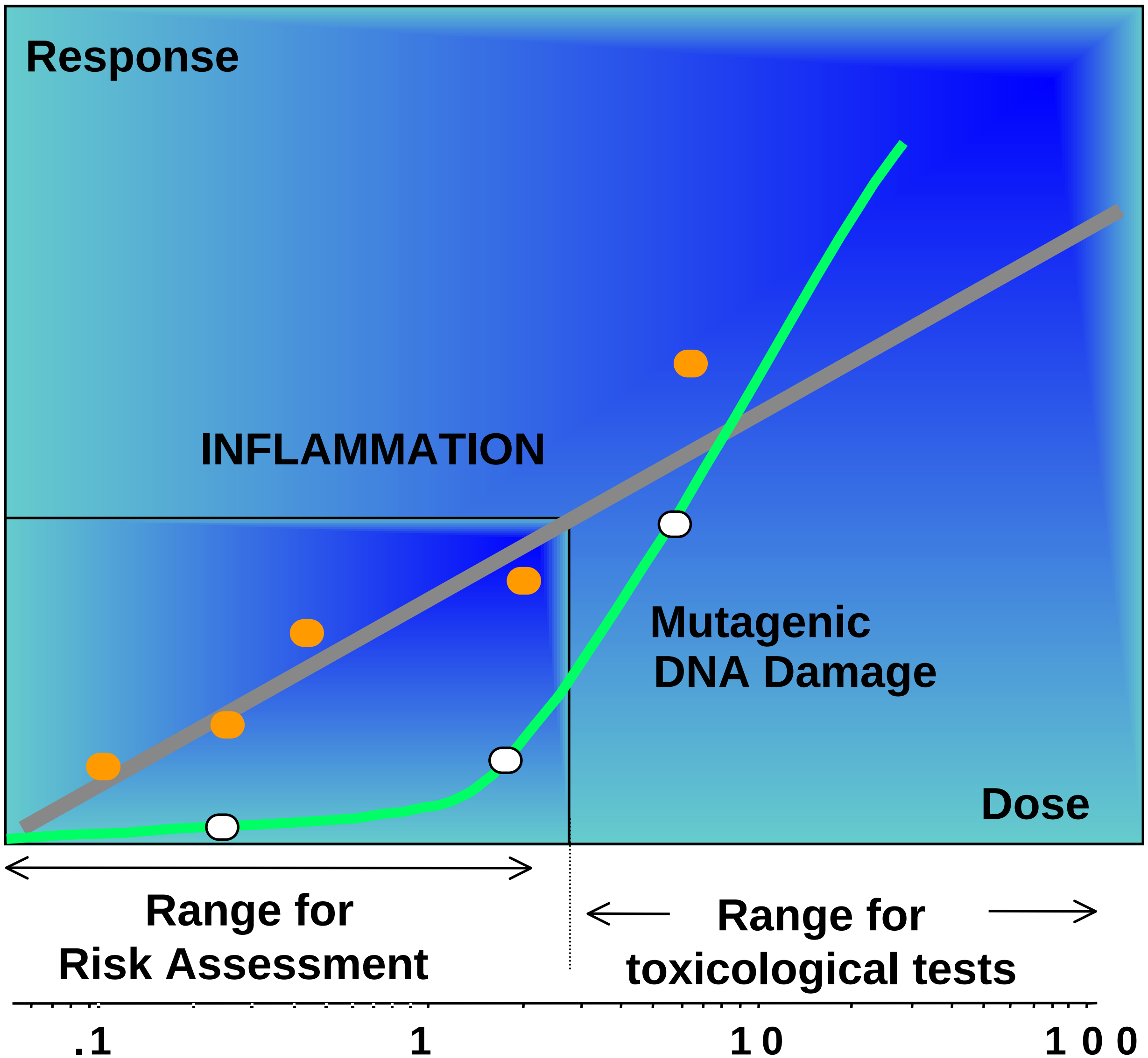


number of analyzed particles = 3653

contact:
Martin Ebert (mebert@geo.tu-darmstadt.de)
Stephan Weinbruch (dh6d@hrzpub.tu-darmstadt.de)

Umweltmineralogie

Linear Dose Response For Inflammation And J-Shaped Response For Genotoxic Substances

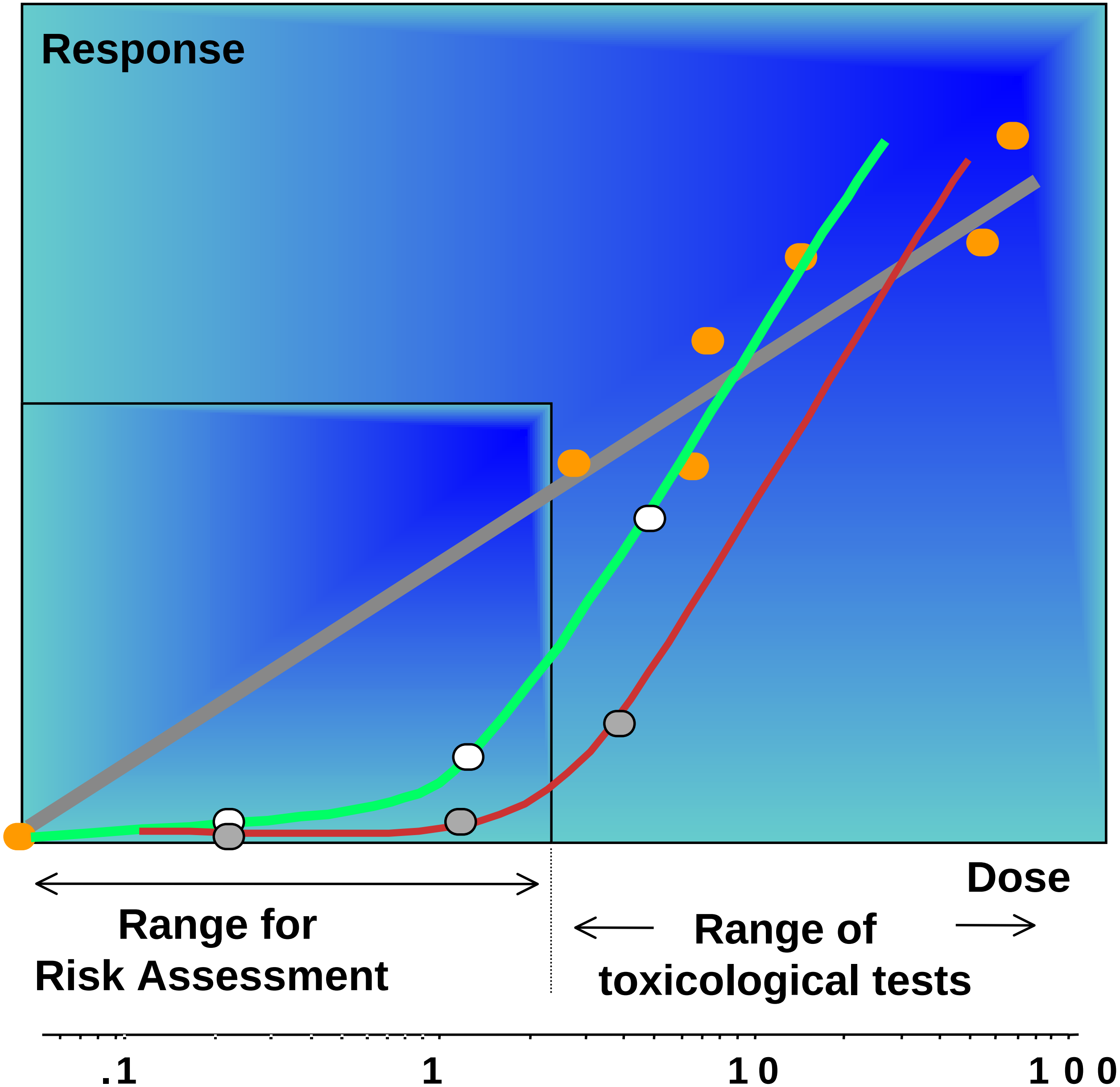


Dose response (DR) for inflammation and DNA damage for hygienic risk assessment for Quartz

J-shaped DR and threshold for a gentoxic effect from Quartz
(white circles, green curve)

Linear DR for an inflammatoric effect
(orange points, gray line)

Questionable Linearity Of Dosis-Response Relation

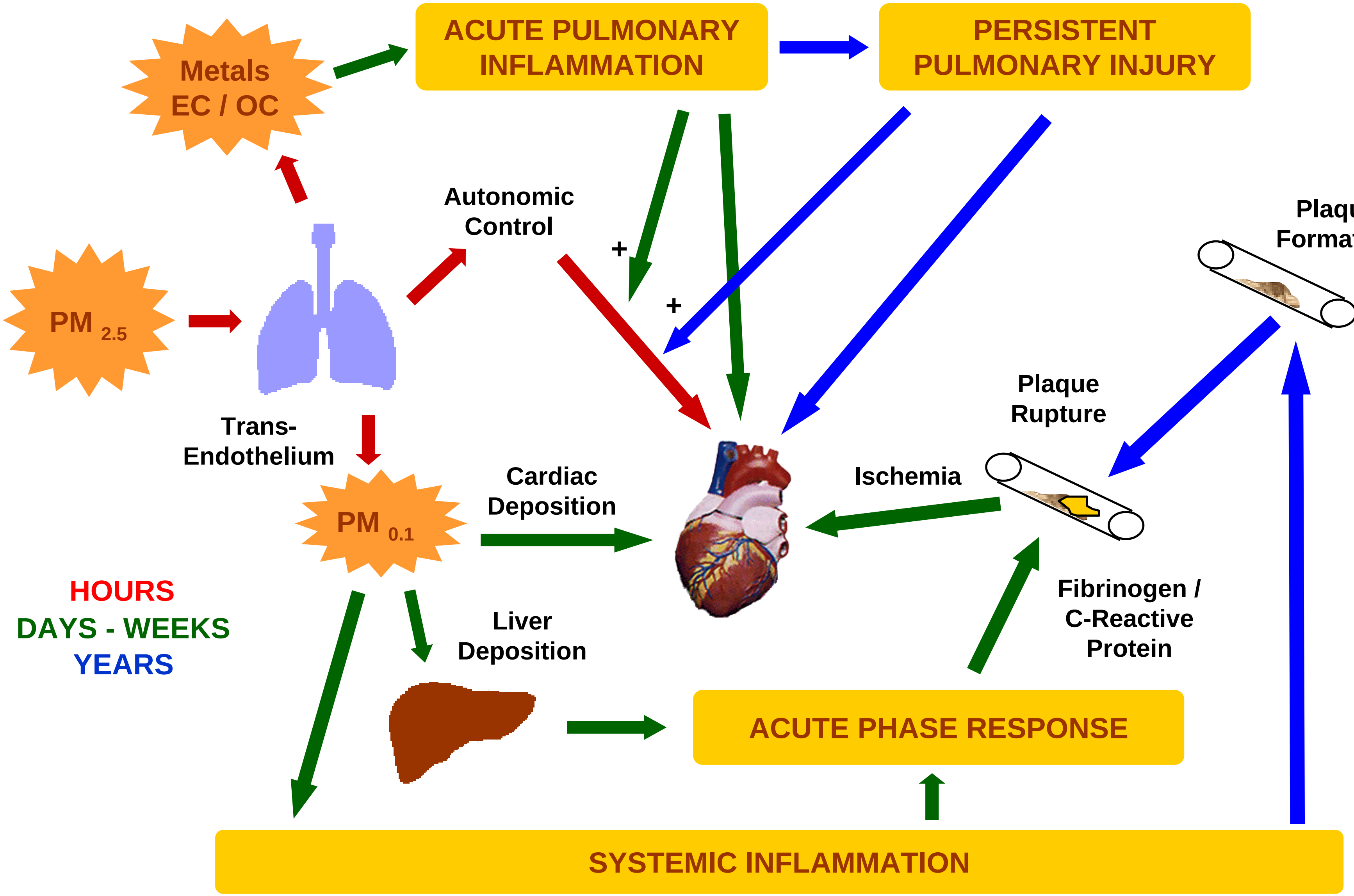


Semi quantitative schema for different threshold curves for different samples

Measured:
red and green line for different aerosols

Hypothesis:
Extrapolation from high dose to low dose (gray line)

Hypothetical Modes of Action for Fine PM



Summary and Conclusions

- **The evaluation of health effects from Diesel exhaust is complex and can not be explained with one single parameter.**
- **Mass is certainly the most important criteria, which determine dose response. Prerequisite is that for the number measurements a differentiation between liquid and solid aerosols must be done.**
- **Important criteria for the evaluation of health effects are:**
  - **PM mass, size and size distribution, number and number distribution, PM concentration in ambient air, PM surface, shape, morphology, chemical composition inclusive adsorbed material and hygroscopic properties together with bioavailability**
- **PM mass emission is declining, despite increasing mileage.**
- **A lower sulphur- content in the diesel fuel strengthen this trend.**
- **PM emission both mass and number decreases continuously.**
- **For certification the gravimetric measurement of mass is still the most efficient and suitable procedure.**