

N. Metz
BMW
München
Germany

38

Development of particle numbers in cities of the diesel passenger car fleet in Germany

Development of particle numbers in cities of the diesel passenger car fleet in Germany

Dr. Norbert Metz and Dr. Christian Cozzarini BMW Group Munich

Abstract

On the basis of the ACEA study on the emissions of fine particles from passenger cars with correlations of particle mass emission and number of fine particles emission calculations were conducted with the emission calculation tool TREMOD. This emission forecast model takes into account the penetration of new technologies into the car fleet as well as the increase in mileage and the distribution of the yearly mileage for the different types of roads. The results show that despite the increase of diesel passenger cars into the car fleet and the increase in the yearly mileage the number of fine particles are going down since 1995. At the air quality site in Erfurt this is reflected for the particle mass and regarding the number of particle classes for fine but not for ultra-fine particles. A correlation between traffic density and the number of ultra-fine particles are sometimes assumed. The question where those ultra-fine particles are coming from and what chemical composition they have is still pending. Due to experiences on dynamometer test benches with different temperatures and dilution ratios it is likely that those ultra-fine particles are not solid. Since the mass emission of solid diesel particles are going down significantly –which is undoubtedly also reflected in air quality measurements – there is evidence that a significant part of ultra-fine particles are liquid aerosol particles from different origin. Secondary aerosols may play a role as well as the humidity of the atmosphere.

Introduction

Fine and ultra-fine particles are penetrating deep into the lung of human beings and animals [1] and are therefore in the focus of science, the public and the regulating agencies, see figure1.

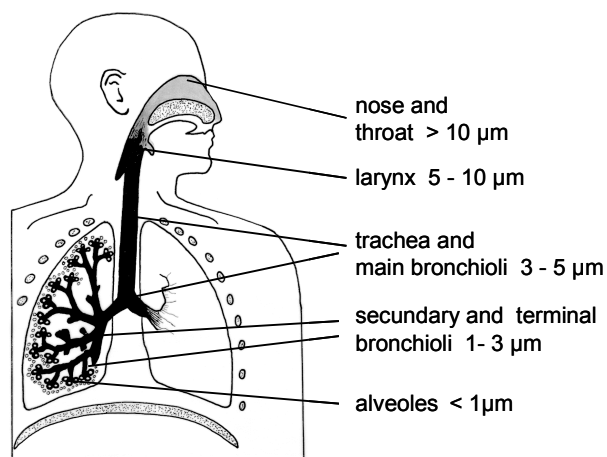


Figure1: Penetration of particles into the lung dependent from their size

Dependent from the chemical composition and the physical shape liquid particles are cleared quickly within hours or in a solid form they stay in the lung for about one year. It is known that solid particles, smaller than 100 nm, are retained for 300 days and more [2].

Particle morphology

Many results with measurements of the so called mobility diameter have been published in recent years [3,4,5,6] and only a few of them tried to correlate the mobility diameter with the aerodynamic respectively the mean geometric diameter of the particles [7,8], see figure 2.

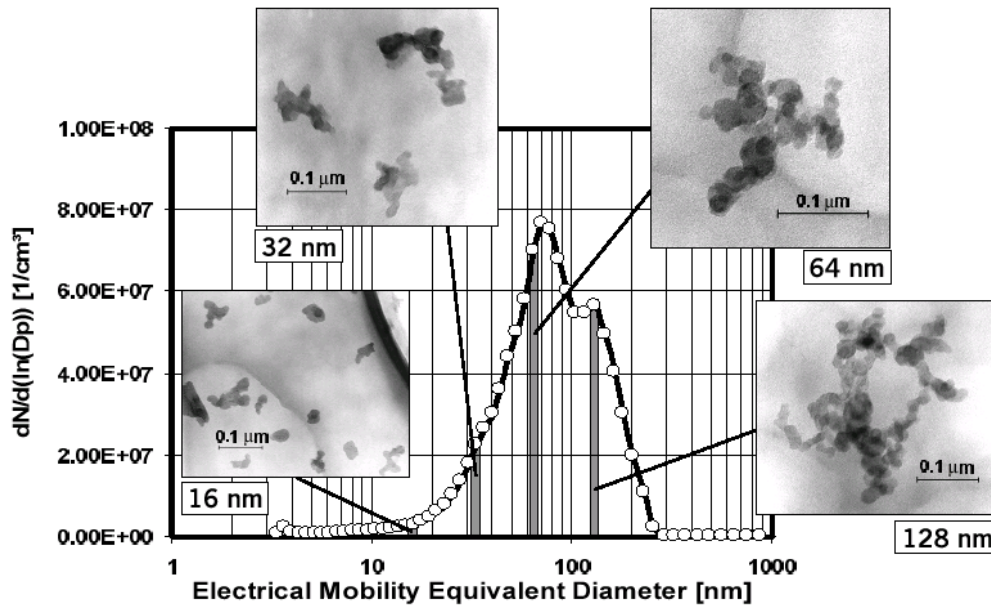


Figure 2: Comparison of morphological investigated geometrical diameters with indirect obtained mobility diameter from diesel soot particles [8]

It seems that a mobility diameter of 80 nm correlates with an mean geometric diameter measured with an electronic microscope of about 110 nm or even more.

A typical diesel soot particle is an aggregate particle with a geometrical size of around 100nm consisting of about 15 primary particles with a typical size of 25 nm [9], see figure 3.

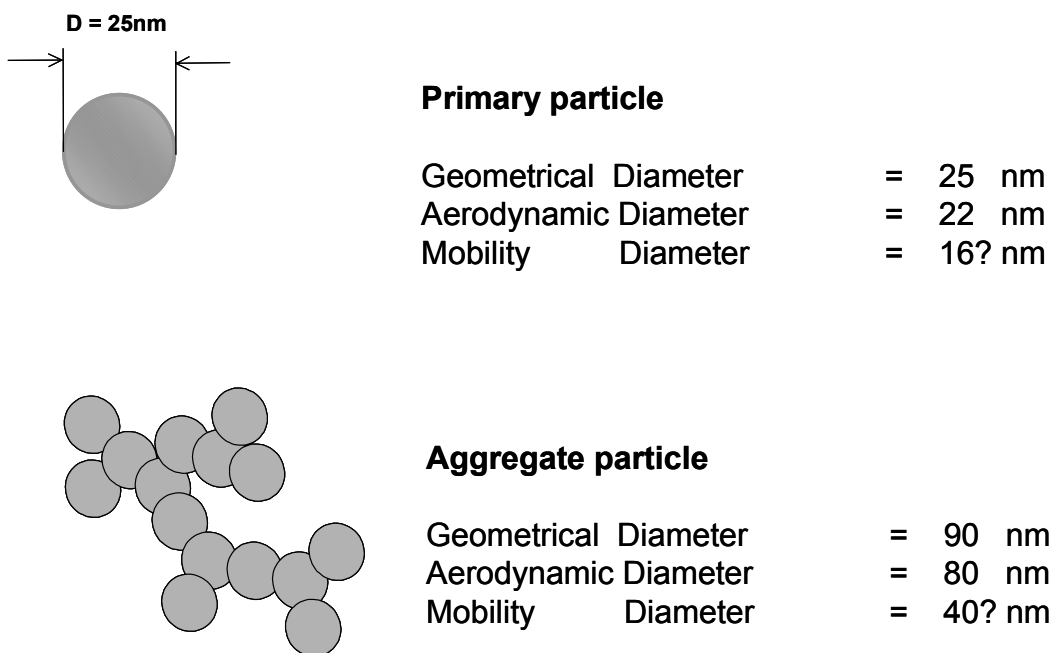
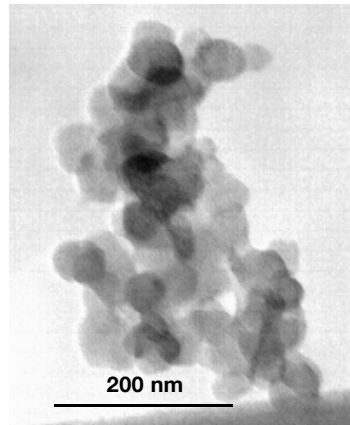


Figure 3: Typical diesel soot particle from a diesel passenger car engine

In most discussions about the size of particles and their effects a distinction of the different kinds of diameter definitions are missing. Therefore it is important to consider which diameter is regarded, see figure 4.

**TEM: 530 d, 120 km/h, Common-rail
High pressure, direct injection**



**Geometrical dimensions
of a typical diesel particle**

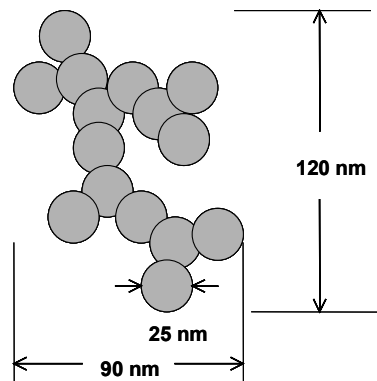


Figure 4: Definition of different diameters

Health effects

Particles of the size range of diesel soot particles are able to reach the alveolar region. More than 50 % of those particles are exhaled again [10], see figure 5.

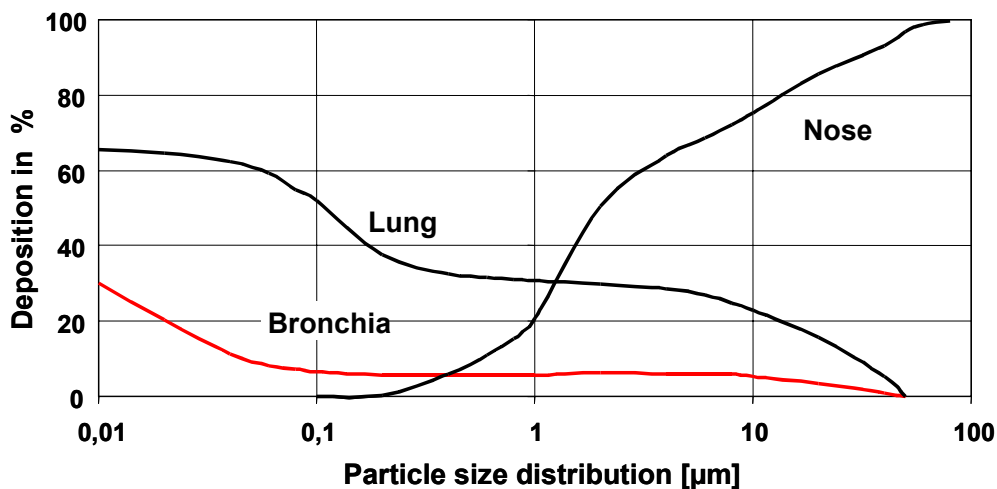


Figure 5: Deposition of particles dependent from their size in the respiratory tract

Those particles, which reach the alveolar region, are eliminated slower. There are different explanations how the cleaning mechanism takes place. One route is the phagocytosis. Macrophages along the respiratory airways and in the alveolar lumen eat the particles and transport them slowly to the upper airways. One hypothesis assumes that ultra-fine particles are so small that macrophages are not able to detect them. It is unknown how big particles must be that macrophages detect and eat them. Particles from the nuclei mode – which are liquid – are eliminated from the lung by a different route, see figure 6 [11,12].

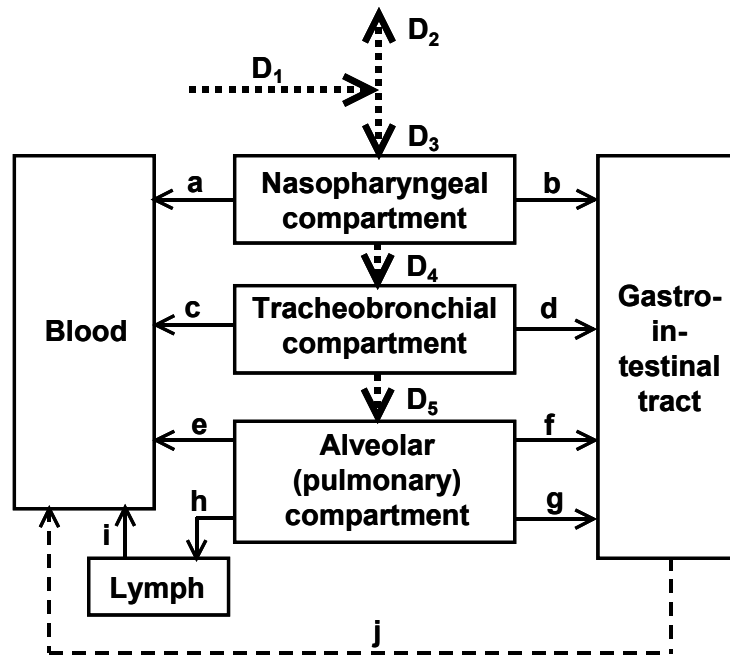


Figure 6: Different clearance routes in the human lung

Dependent from their character - hydrophilic or hydrophobic - they are deposited in the surfactant, which cover the whole airways down to the alveolar region and eliminate all substances over the lymph nodes. Therefore especially the particles from the accumulation mode must be considered with care, see figure 7.

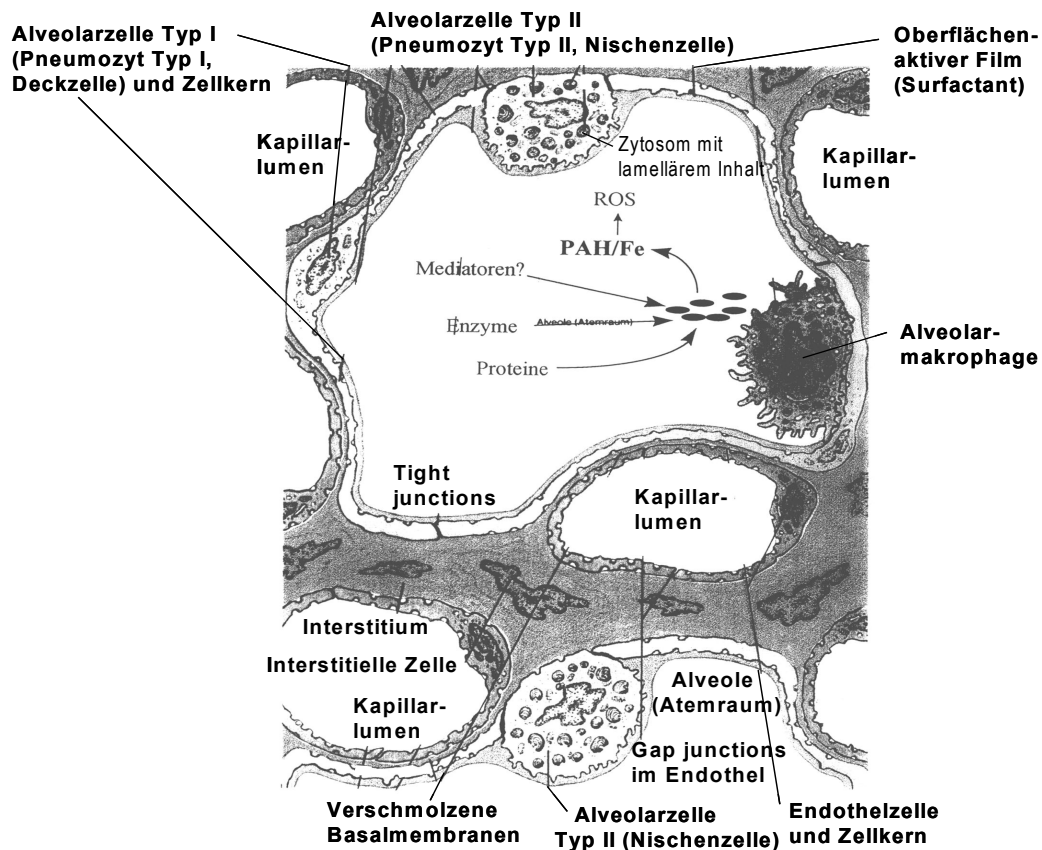


Figure 7: Alveolar sac section with surface active surfactant and an alveolar macrophage

Nevertheless one hypothesis assumes that solid particles below 50 nm will undergo the cleaning mechanism of the macrophages and are able to penetrate into the interstitium around the alveolar walls. A recent study in the Lovelace Laboratory in Albuquerque, New Mexico showed that there are clear differences in the deposition of this type of particles between rats, monkeys and coal workers [13], see figure 8.

DISTRIBUTION OF PARTICULATE MATERIAL IN RATS, MONKEYS, AND HUMANS EXPOSED TO COAL DUST

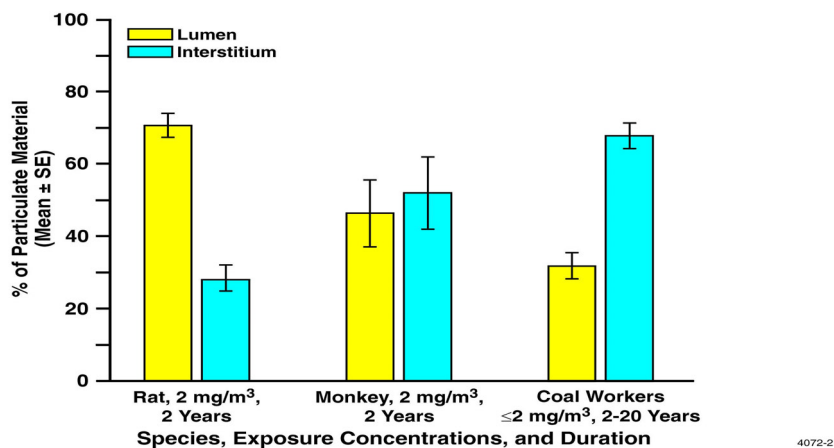


Figure 8: Differences in the deposition of particulate matter in animals and humans

High dose exposures of fine particles showed clearly that solid soot particles are deposited in man in the walls between the alveolar sacs (Interstitium), while in the rat almost all particles are deposited in the alveolar lumen, see also figure 9.

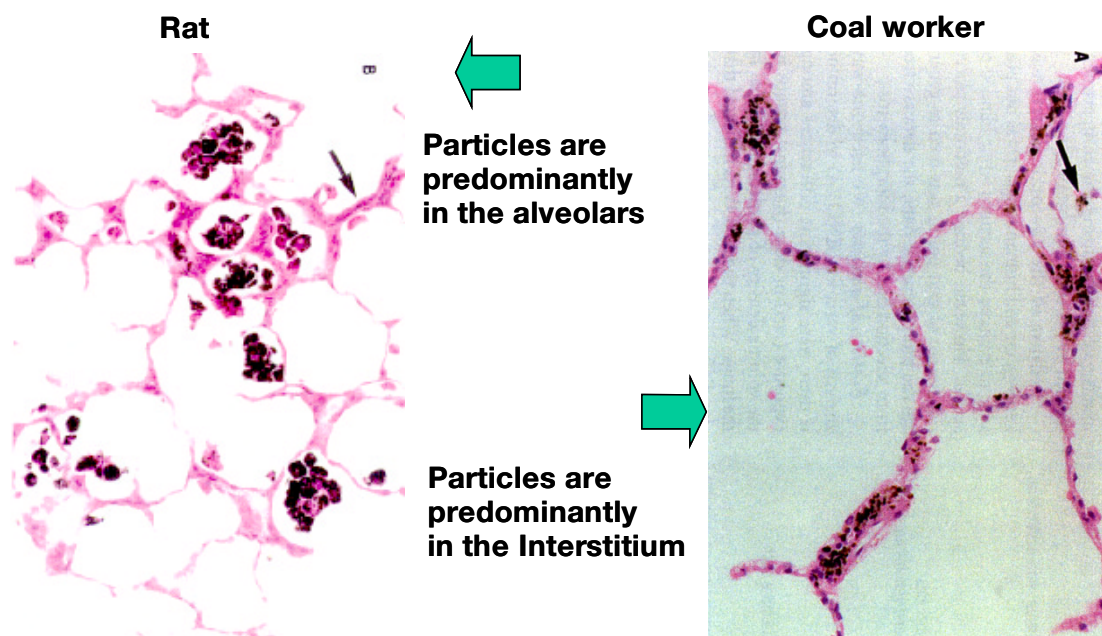


Figure 9: Particulate matter deposited in the lumen (rats) and in the interstitium (man)

It is clear that the results of rat exposures are not suitable to calculate a risk for humans. In earlier investigations were rats, hamsters and mice were exposed to higher doses of exhaust emissions rats happened to be the most sensitive species, see figure 10.

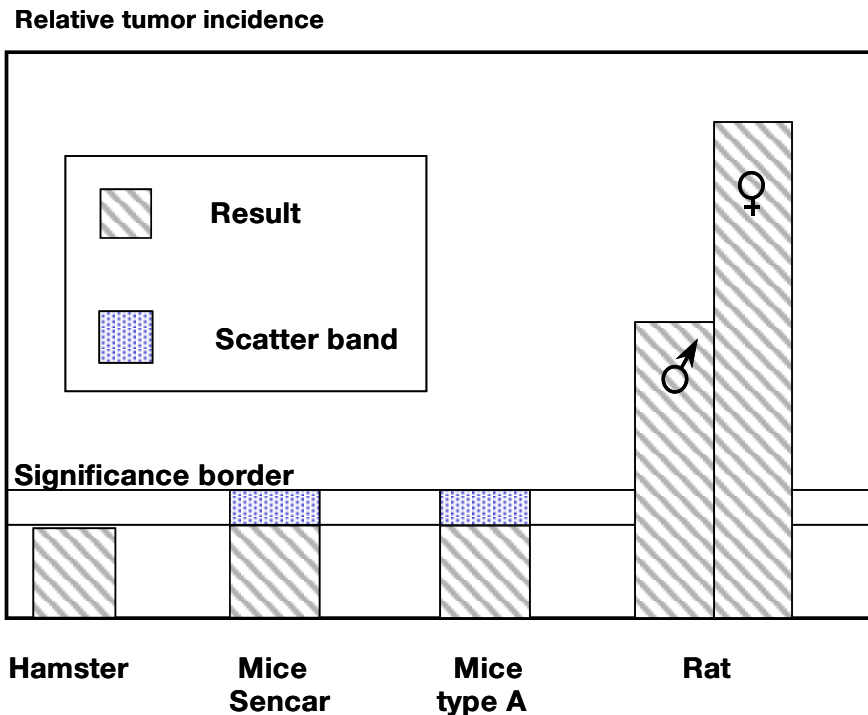


Figure 10: Tumor response to 6.6 mg/m³ diesel particulate matter exposure for two years

Only in rats a tumor response was detected at very high doses, in hamsters and mice no significant results were obtained and also with lower doses no tumor response was seen [14]. With liquid particles in this size range no adverse health effect results are known.

Contribution of road transport to PM₁₀ particulate matter emission in Germany

Searching for the sources of fine and ultra-fine particles also road transport has been investigated [15]. Especially diesel vehicles are blamed to be a main contributor to this size class of particles [16]. Additionally there are fears that new technologies with high pressure injection and common rail might have even an adverse effect on the emission of the number of ultra-fine particles, while there is no doubt that the particle mass is going down significantly in diesel passenger cars and in diesel duty vehicles [17]. Investigations of the Association of Constructeurs of European Automobiles ACEA tested a variety of diesel vehicles with the result that new technologies in the vehicles reduce the particle mass and as well the particle number [18].

Development of the particulate matter emission in Germany

Since more than 15 years particulate matter emissions are going down significantly due to improvement from all contributing sources [19], see figure 11.

TSP in Mt

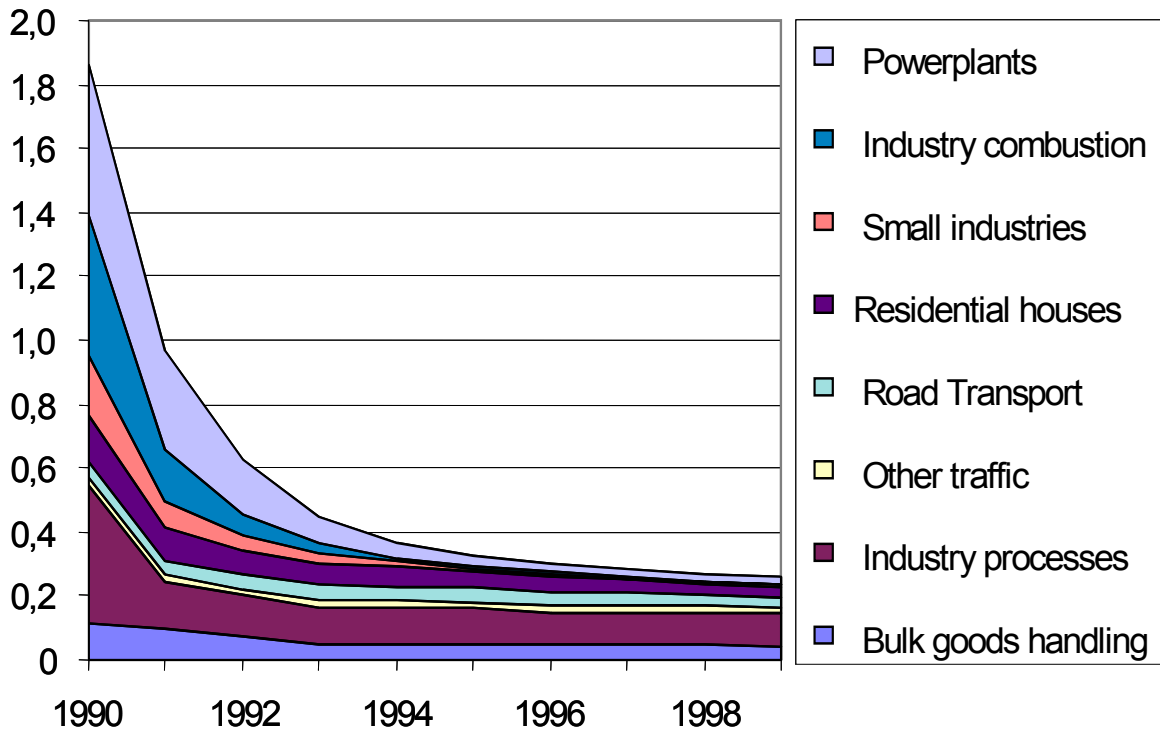


Figure 11: Development of total suspended particulate matter in Germany from 1990 to 1999

Coarse particles with a size over $10\ \mu\text{m}$ have only a minor health effect, therefore particles below $10\ \mu\text{m}$ (PM_{10}) are of special interest. In 2000 about 230 kt are estimated for Germany [20], see figure 12.

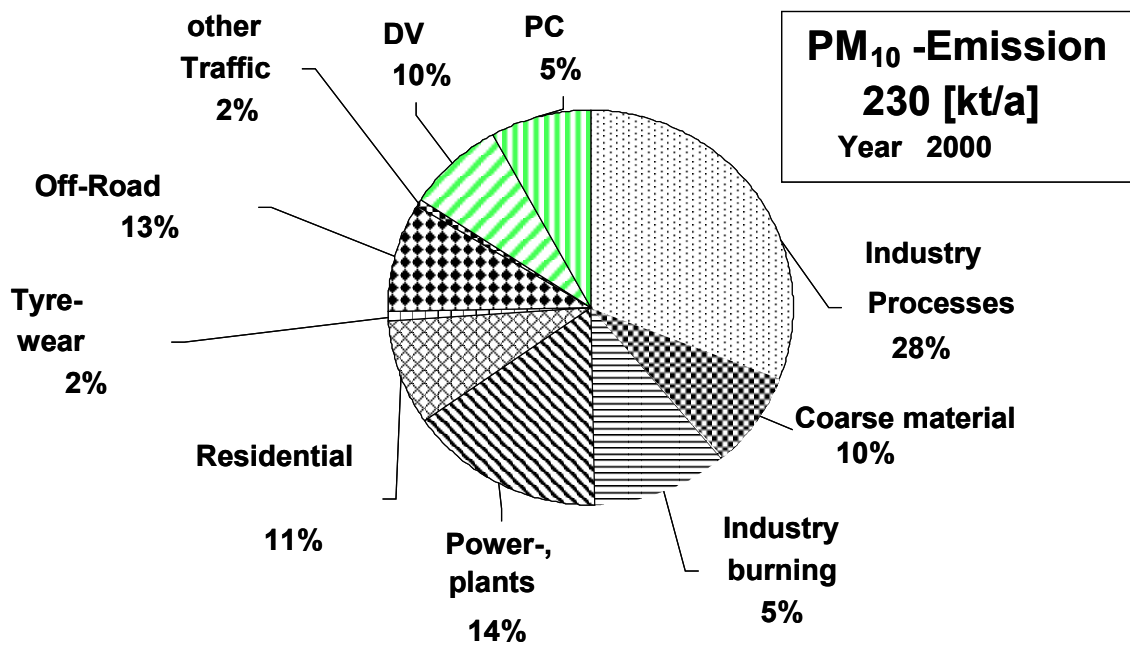


Figure 12: Contribution of different sources to PM_{10} emission in Germany in 2000

Duty vehicles contribute about 10 %, passenger cars about 5 % with a decreasing trend. Also other sources are reducing their emission, therefore it can be expected that air quality regarding PM₁₀ and PM_{2.5} is improving further in the future.

Regarding air quality total suspended particle-concentrations as well as PM₁₀-, PM_{2.5}- and soot-concentrations are going down. Figure 13 shows the results for a typical urban measurement site, partly from measured data and since 1998 calculated with a simple box model SEM [21].

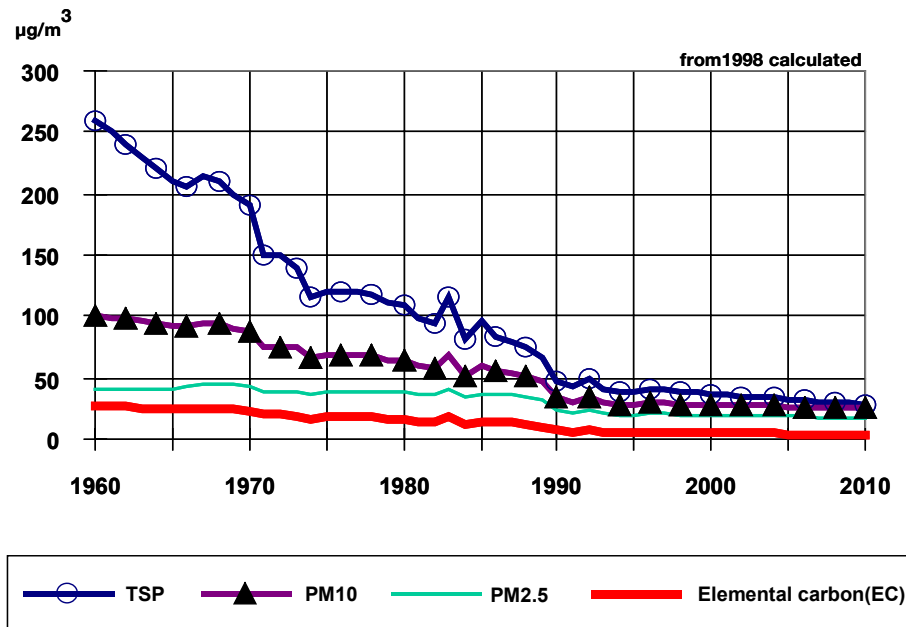


Figure 13: Development of air quality regarding different size classes of PM in Germany

Besides the measurements from the particle mass concentration only a few measurements of ambient particle numbers are available [12], see figure 14.

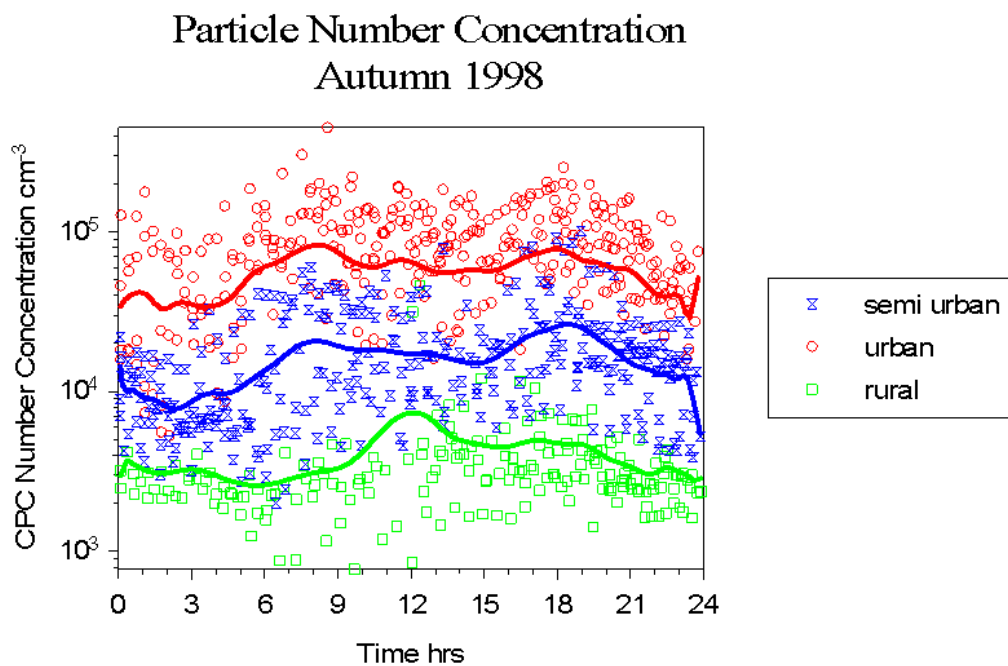


Figure 14: Results from a remote site (Hohenpeissenberg), a semi-urban site (Neuherberg) and a city site (Luise Kiesselbachplatz, direct on a crossing of busy traffic roads).

The particle numbers near the curbside of the busy street are higher. The share of road transport is unknown. Therefore this paper estimates the number of particles, which are coming from diesel passenger cars for the mileage of the total diesel passenger car fleet in all German cities.

For this purpose it is appropriate to use the TREMOD road transport emission model, which was developed for the conventional exhaust gas emission compounds [22]. Figure 15 shows the schematic of the calculation principle (This model, developed at the university of Vienna, is very close to the TREMOD calculation principle).

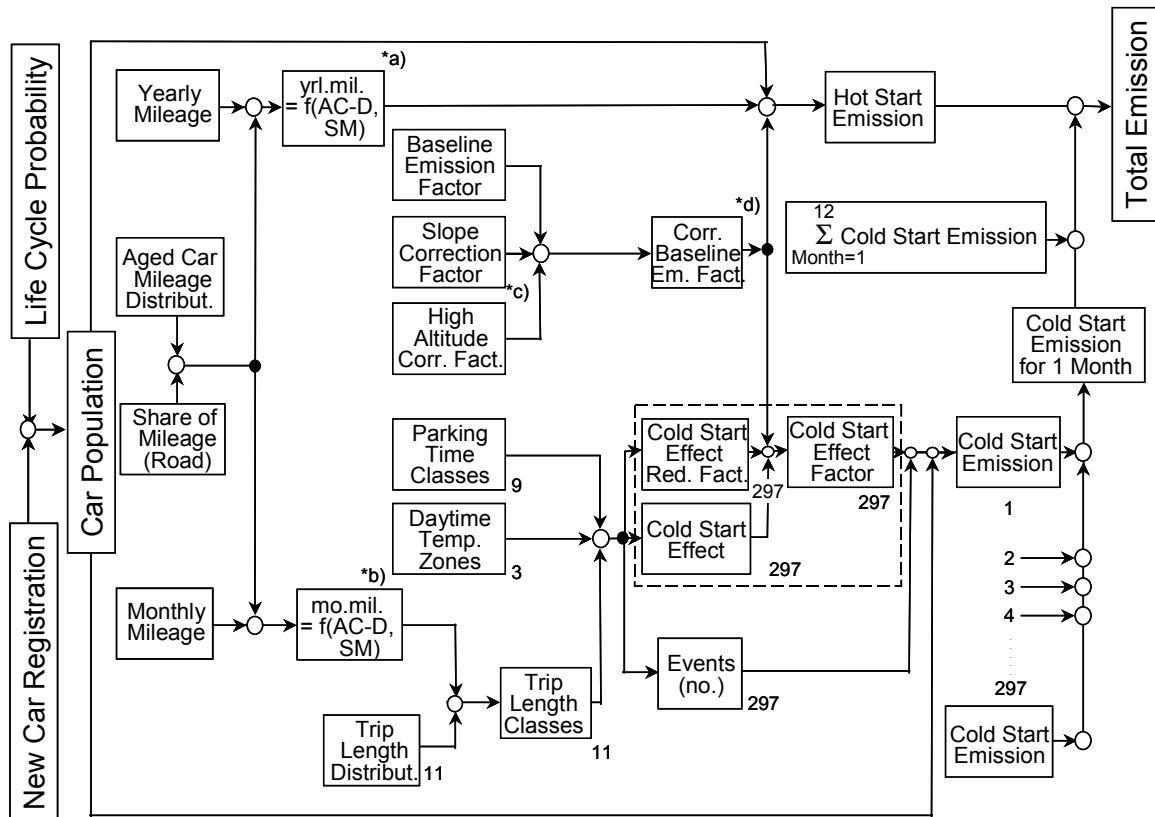


Figure 15: Calculation principle for the determination of annual emissions from cars [23]

*a) yrl. mil.: Yearly Mileage, AC-D: Aged Car Mileage Distribution
SM: Share of Mileage on different Road Classes

*b) mo. mil.: Monthly Mileage, AC-D: Aged Car Mileage Distribution

*c) Corr. Fact.: Correction Factor *d) Em. - Fact.: Emission Factor,

This model takes into account the increase and penetration of new vehicle technologies into the car fleet, the distribution of the mileage on different road categories and the relevant emission factors as well as cold start effects.

Figure 16 depicts the development of the vehicle numbers divided for all passenger car classes, were on the upper part all diesel technology categories can be seen.

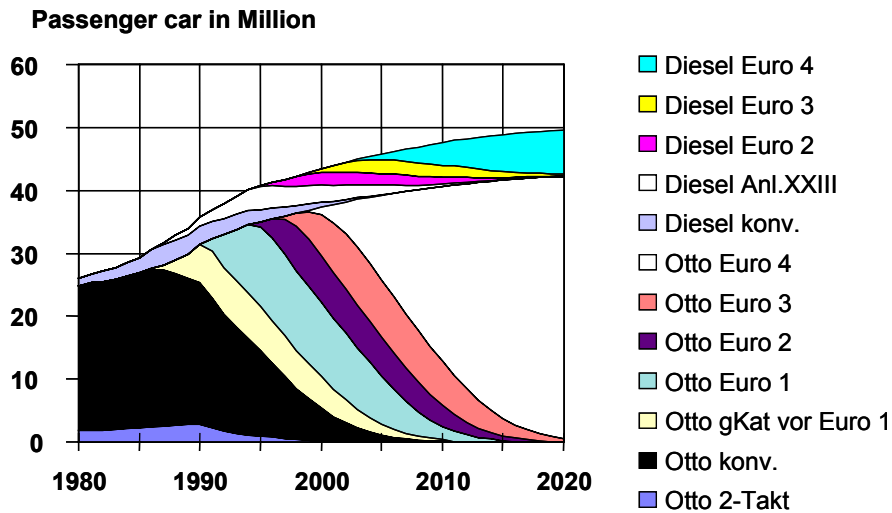


Figure 16: Development of the car population in Germany from 1980 to 2020

Figure 17 shows the development of the mileage in Germany. For this calculation the part for city roads are used, because cities are the place where most people are exposed.

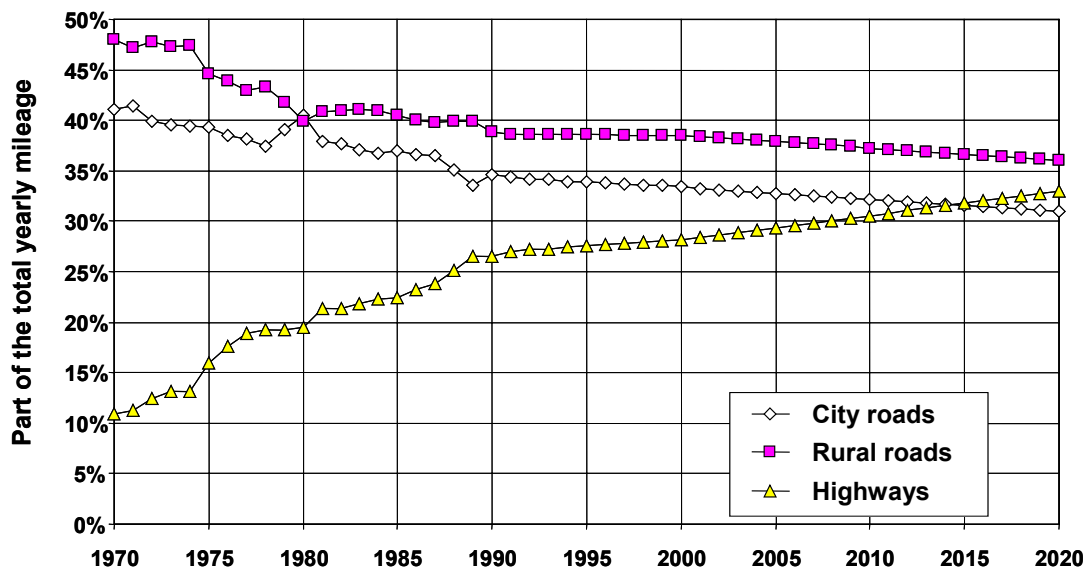


Figure 17: Distribution of the annual mileage on different road types

Since 1990 the particle mass emission standards for passenger cars were reduced by 93 %, see figure 18.

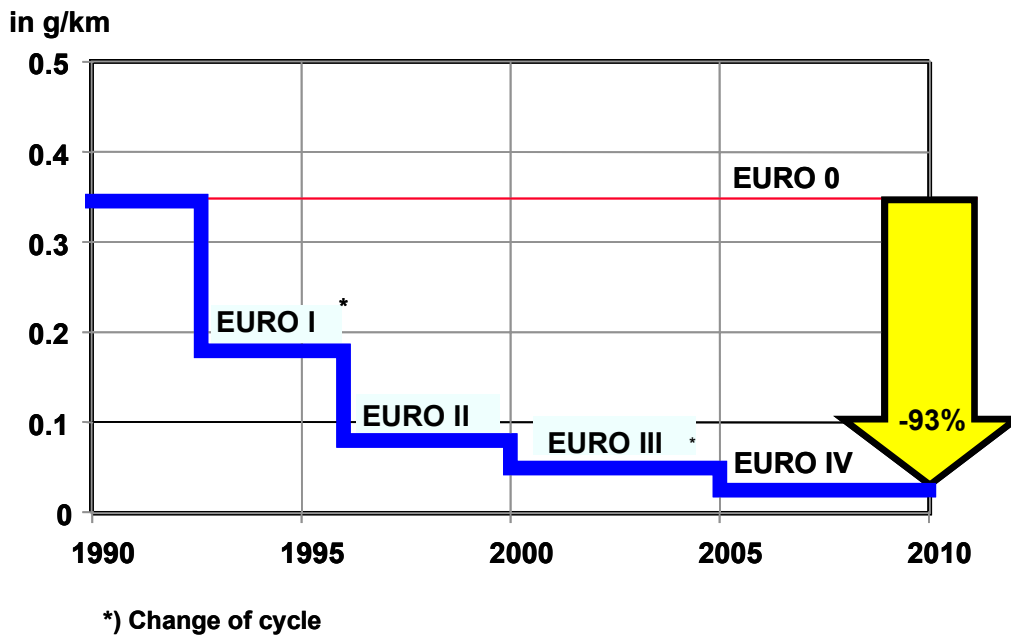


Figure 18: Development of the particle emission standards in the European Union [17]

As a result of the reduction of particle emissions from heavy and light duty vehicles as well as from passenger cars the total particulate matter emission from road transport is declining since 1994, see figure 19.

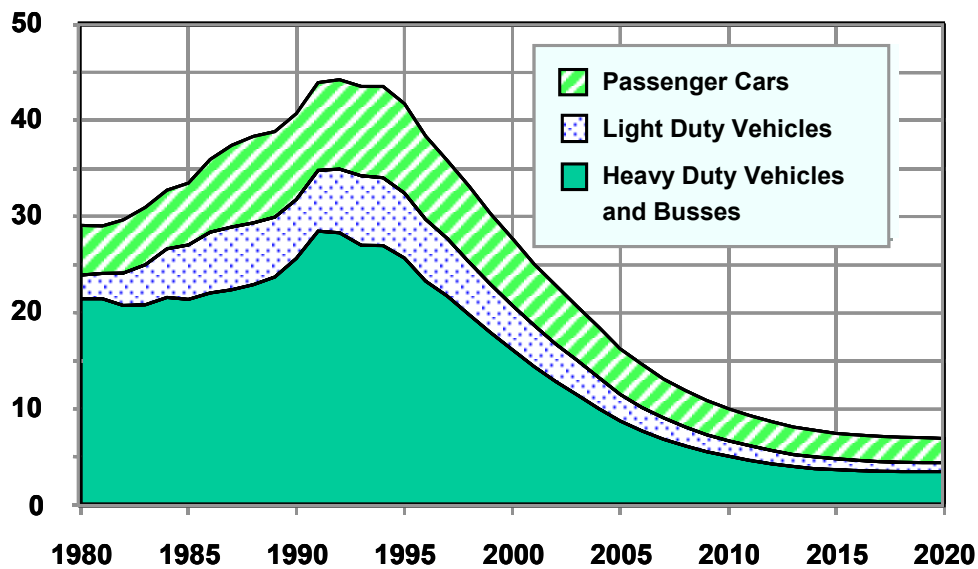


Figure 19. Development of the particle mass from road transport in Germany

The number factors used in the following calculation are derived with two approaches. The first one is based on the correlation with mass calculated from an ACEA program [18] with 11 diesel passenger cars, from a CONCAWE program [24] with four diesel passenger cars and from VW measurements with two diesel passenger cars in the European urban driving cycle [25] were parallel to the number also the mass was determined gravimetrically. The second approach is based on measurements of the particle number on dynamometers tests with different technology classes and test cycles.

One result of the ACEA study is shown in figure 20. For a certain mass emission factor a relevant number factor can be derived. It must be noted that the data base for the number factors are not so sound as it is the case for the conventional compounds but as a first estimation the data are sufficient accurate.

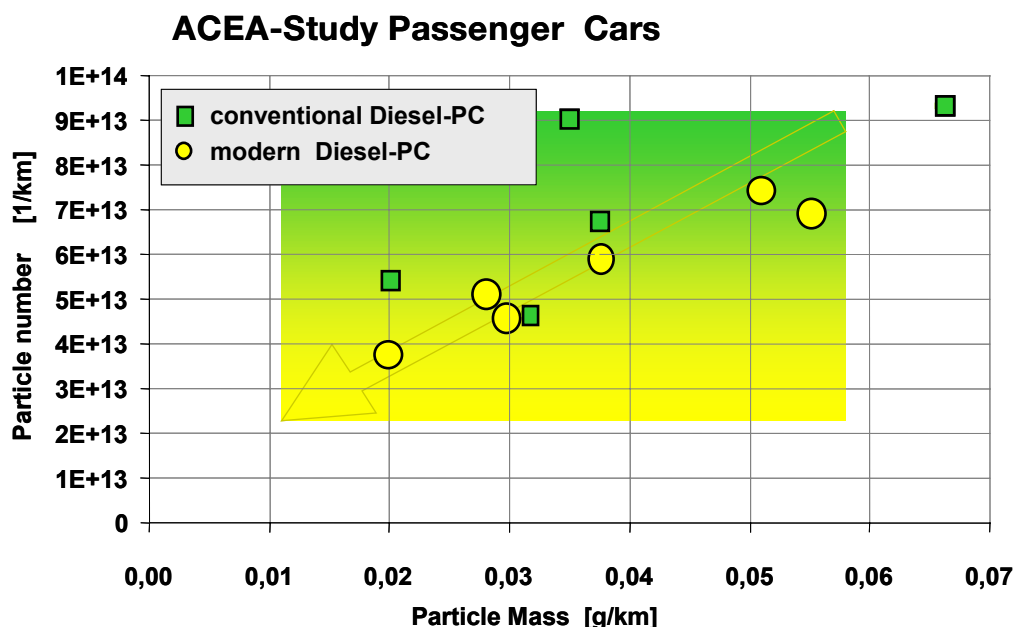


Figure 20: Correlation of particle mass with particle number of passenger cars [18]

Table 1 summarizes the input data for this approach.

Table 1: Input data for the calculation of particle numbers for the diesel passenger car fleet

Vehicle category	Road Group	Dim.	1980	1990	2000	2010	2020
PKW Diesel 86-88	City road	g/km	0,142	0,152	0,167	0,000	0,000
PKW Diesel Anl. XXIII	City road	g/km	0,000	0,109	0,120	0,127	0,000
PKW Diesel EURO2	City road	g/km	0,000	0,000	0,086	0,081	0,084
PKW Diesel EURO3	City road	g/km	0,000	0,000	0,058	0,054	0,057
PKW Diesel EURO4	City road	g/km	0,000	0,000	0,000	0,033	0,033
PKW Diesel vor 86	City road	g/km	0,324	0,354	0,000	0,000	0,000
Vehicle category	Road Group	Dim.	1980	1990	2000	2010	2020
PKW Diesel 86-88	City road	E direct (t)	194,63	2.326,41	412,24	0,00	0,00
PKW Diesel Anl. XXIII	City road	E direct (t)	0,00	1.320,72	1.261,22	176,53	0,00
PKW Diesel EURO2	City road	E direct (t)	0,00	0,00	1.115,13	289,77	8,08
PKW Diesel EURO3	City road	E direct (t)	0,00	0,00	250,32	396,10	66,27
PKW Diesel EURO4	City road	E direct (t)	0,00	0,00	0,00	687,98	1.160,50
PKW Diesel vor 86	City road	E direct (t)	2.359,47	699,82	0,00	0,00	0,00
Vehicle category	Road Group	Dim.	1980	1990	2000	2010	2020
PKW Diesel 86-88	City road	Number	2,9194E+23	3,49E+24	6,18E+23	1,00E-02	1,00E-02
PKW Diesel Anl. XXIII	City road	Number	1,00E-02	1,98E+24	1,89E+24	2,65E+23	1,00E-02
PKW Diesel EURO2	City road	Number	1,00E-02	1,00E-02	1,67E+24	4,35E+23	1,21E+22
PKW Diesel EURO3	City road	Number	1,00E-02	1,00E-02	3,75E+23	5,94E+23	9,94E+22
PKW Diesel EURO4	City road	Number	1,00E-02	1,00E-02	1,00E-02	1,03E+24	1,74E+24
PKW Diesel vor 86	City road	Number	3,5392E+24	1,05E+24	1,00E-02	1,00E-02	1,00E-02

Figure 21 gives the result. Starting in 1993 the numbers emitted from diesel passenger cars in German cities are going down and reaching a level, which is lower that the one for 1980.

Number emitted from diesel PC during city operation

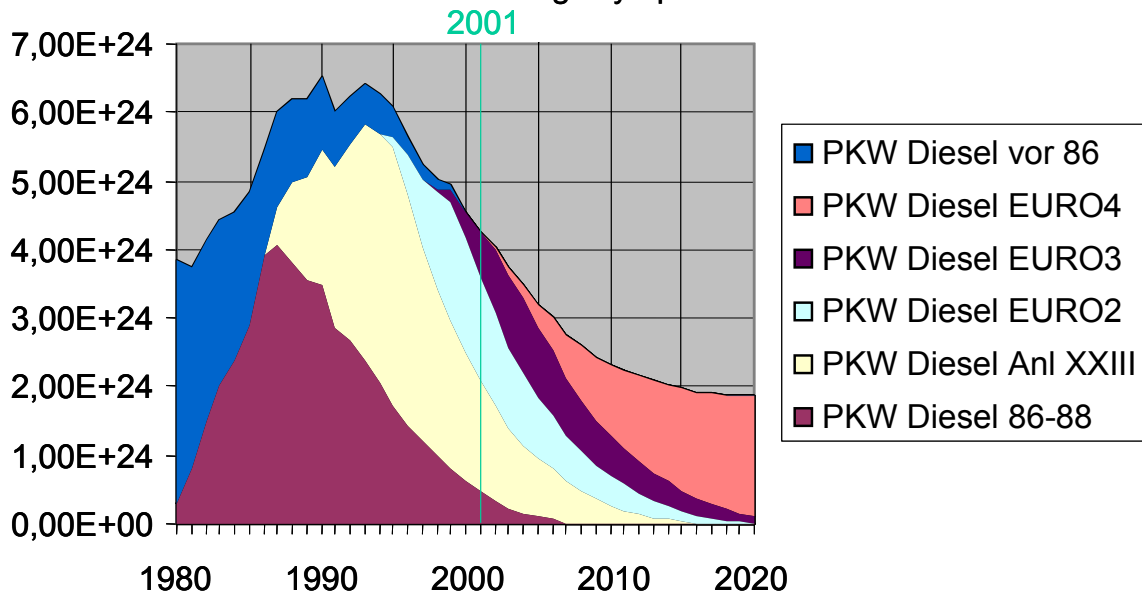


Figure 21: Number development of the diesel passenger car fleet in cities in Germany

Regarding the contribution of the different vehicle categories, e.g. in 2001 a small portion is related to diesel cars produced between 1986 and 1988, the biggest portion from diesel cars certified with the requirements of “Anlage XXIII”, which correspond EURO 1, the second biggest portion comes from EURO 2 diesel cars, followed by EURO 3 diesel cars. Only in the years coming EURO 4 cars are penetrating into the fleet and thus contribute to the result. For the second approach direct number measurements for different diesel car technologies are used. The input data are summarized in table 2.

Table 2: Number factors used as input for the second approach

Vehicle Category	year	number factor (number / km)
Pre 1986	before 1986	1,90 E+14
86 to 88	1986	1,60 E+14
EURO 1	1990	1,42 E+14
EURO 2	1995	7,10 E+13
EURO 3	1999*	4,97 E+13
EURO 4	2004*	3,55 E+13

* = due to taxation incentives, starting one year earlier as demanded

The relation of the different vehicle categories is shown in figure 22. Overall a reduction in the particle numbers of 78 % is reached with diesel cars fulfilling EURO 4.

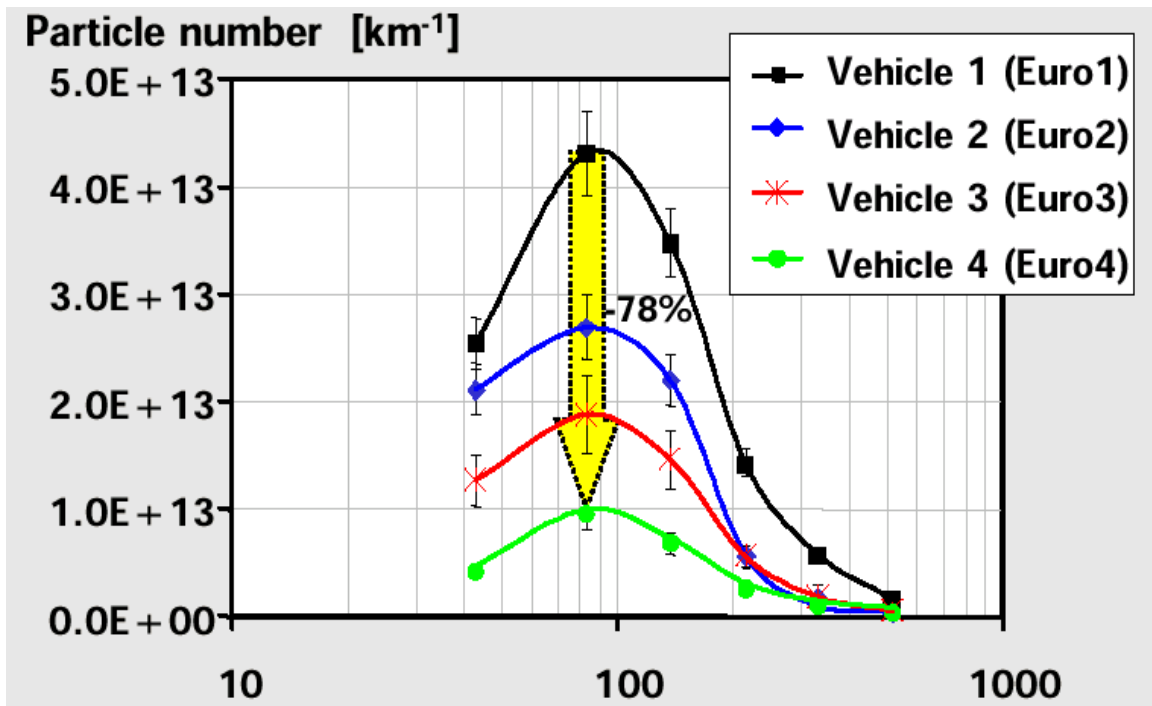


Figure 22: Number distribution of different diesel passenger car categories [25]

The number result for the operation within cities in Germany is shown in figure 23.

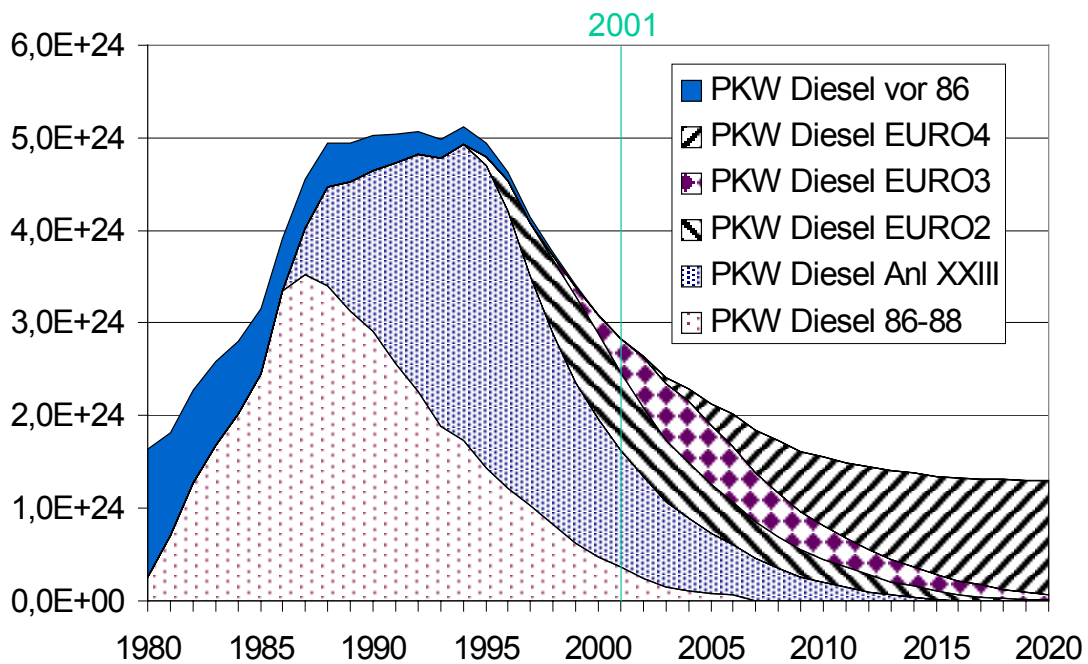


Figure 23: Number development of the diesel passenger car fleet in German cities

The numbers are going down since 1996. The level is somewhat lower, may be that this approach do not reflect enough the influence of the cold start. The contribution of the different diesel car categories is about the same as in the first approach. In 2010 the number level of 1980 is reached again.

Figure 24 shows the comparison of the two approaches. The scatter band, which results from the two approaches, will reflect the development from the diesel car fleet in the cities in Germany. The trend to lower numbers will continue the next decade.

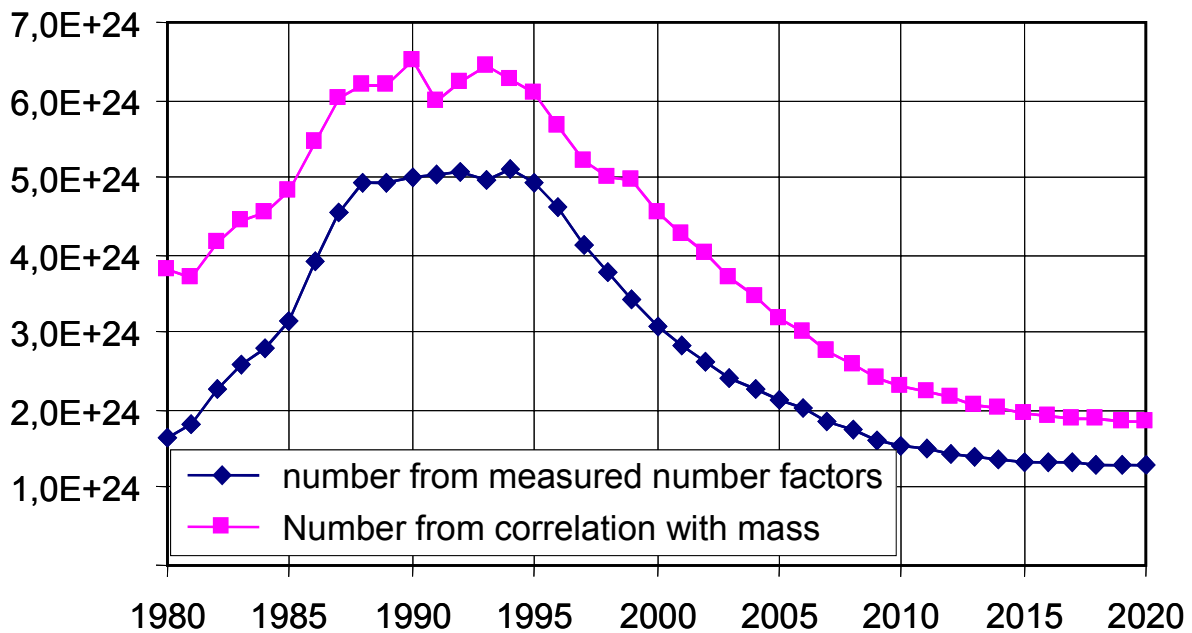


Figure 24: Comparison of two approaches to calculate the number development of the diesel passenger car fleet in cities in Germany

Conclusions

Passenger cars, especially those with diesel engines are still increasing in Germany. Despite the rising share of diesel engines in the passenger car fleet exhaust gas emissions are going down. The calculations of the resulting number of particles from diesel passenger cars show that also the numbers of particles which are emitted are decreasing. It would therefore be consequent that particles in the ambient air also are going down if road transport is the main contributor. Therefore number measurements on different sites in cities have to be analyzed to see the trend and to find out which contributing sources are important.

References

- [1] Andersen, Utah
- [2] Muhle, Personal communication 3/2001
- [3] Kittelson D.B., Engines and Nanoparticles, J. Aerosol Sci., V29 (1998) Nr. 5 and 6
- [4] Kasper M, Matter U. et Burtscher H. SAE-Paper 2000-01-1998
- [5] Nanoparticle measurement conferences, Zürich, 1997 to 2001
- [6] Khalek I.S., Kittelson D.B., Graskow B.R., Wei O., Brear F., SAE-Paper 980525
- [7] Metz N., Resch G., Schönberger K. et Steinparzer F., MTZ 61.Jg. (2000) 24-30
- [8] Cartus T., Dorfer F., Schindler W., Reischl G.P., 4th ETH-Nanoparticle Conference 2000
- [9] Metz N., 4th ETH-Nanoparticle Conference 2000
- [10] Wichmann et al., Handbuch der Umweltmedizin, ecomed-Verlag, Landsberg
- [11] Goldsmith J.R., Friberg L.T., effects of air poll. on human health in Stern Air Poll., Vol.II, Chapt.7, p465, Acad press, N.Y., 1977
- [12] Karg E., Ferron G.A., Busch B., Heyder J., BayFORKLIM-Report B II 7, 4/1999
- [13] Nikula K.J., Vallyathan V., Green F.H.Y., Hahn F.F. Infl. of exposure conc. or dose on the distr. of PM in rat and human lungs, Environm. Health Persp., V109 (2001) 311-318
- [14] Stöber W., On the health hazards of part. diesel eng. exh. Emissions SAE-Paper 871988

- [15] Lamprecht U., 4th ETH Conf. on nanoparticle-measurement, Session 7, Zürich 7.8.2000
- [16] Brand P., Heyder J., Wichmann H.E., Tuch T., Particle Number and mass concentration in East. Germany, *Atm. Environm.*31(1997)4193 and personal communication 2 / 2001
- [17] Kohoutek P., Metz N., Richter K., Part. Matter emissions from modern diesel engines 7th Inhalation Symp. ILSI, 24.2.1999 Hannover
- [18] Margaria G., ACEA programme on emissions of fine particles from passenger cars Report Dec. 1999, Brussels
- [19] Strogies UBA, Daten zur Umwelt, Berlin, Personal communication 4/2001
- [20] Prüller S., TU Wien, WG Emission and air quality 3 / 2001
- [21] Metz N., Air quality and exposure of TSP, PM10, PM2.5 and soot, 7th Inhalation Symp. ILSI, 24.2.1999 Hannover
- [22] TREMOD emission model Höpfner U. et al. Heidelberg, 2000
- [23] Cozzarini C., Theses 1999 TU Wien
- [24] Hall D., CONCAWE Report 98 / 51, Bruxelles
- [25] Schindler K.P., IAA-Technical Conference, 28.9. 2000 Frankfurt