EMISSION FACTORS OF SEVERAL PARTICLE PROPERTIES FROM CURRENT DIESEL PASSENGER CARS

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Introduction

Atmospheric particulate matter (PM) is responsible for serious health effects, especially in urban areas. PM is by definition a mass metric, but recent studies have been seeking for the definition and quantification of additional physical parameters for airborne particles such as surface area, number, and size, to associate with adverse health effects (1). On the other hand, emission standards for vehicle exhaust particle emissions become increasingly stringent, leading the current gravimetric procedure to its detection limit. This, by turn, leads to the necessity of exploring more sensitive particle measures to regulate exhaust emissions.
To meet these needs, this paper presents an overview of particle exhaust emission factors, including several particle proprieties, such as mass [g km^{-1}], active surface area [cm^2 km^{-1}], particle number [km^{-1}] and particle size distribution, collected in the European "PARTICULATES" project. The main aim is to compare new and old vehicle technologies and investigate how mass emission standards have been reflected in additional particle properties.
These emission factors are based on measurements conducted in a number of laboratories around Europe, following a strict protocol which included the fuel use, the driving patterns, the vehicle conditioning and the experimental conditions and set-up. For all particle metrics, we adopt the approach used in COPERT (2), that is the emission factors are presented as a function of the mean speed of real-world driving patterns. These speed-dependent curves were calculated considering either hot cycles or hot cycles and steady states.

Data and method

Sampling system

The sampling system for aerosol measurements consists of two branches, downstream of the porous dilutor, which samples aerosol directly from the exhaust line. One branch (termed "dry" branch) consists of a thermodenuder to remove volatiles and an ELPI (Electrical Low Pressure Impactor), which provides the size specific particle concentration. No volatile particle separation takes place in the other "wet" branch. In this branch, particles are sampled by the diffusion charger (DC) which monitors the real-time concentration of the active surface. Then Condensation Particle Counter (CPC) records the number concentration in real-time. Moreover, for steady state conditions, the number-weighted size distribution is collected using the Scanning Mobility Particle Sizer (SMPS) (Table 1). More information on this sampling system and the sampling conditions established are given elsewhere (3).

Vehicles and fuels

The vehicle sample consisted of 19 cars, including conventional diesel ones meeting several Euro standards, vehicles equipped with diesel particle filters (DPF), port fuel injected and direct injection spark ignition cars (Table 2). Moreover, in order to evaluate the effect of fuel properties on the emission level four diesel and three gasoline fuels were used, mainly

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differentiated with respect to their sulphur content which was ranging from 300 to below 10 mg/kg (Table 3). We only deal with diesel vehicles in this paper due to size constraints.

Table 1 Particle related information collected with the sampling system

Instrument	Property	Size resolution	Temporal resolution
Condensation Particle Counter (CPC)	Particle number concentration	One channel >7 nm	1 s (transients)
Scanning Mobility Particle Sizer (SMPS)	Particle sizing and concentration	64 channels per decade	90 s (steady states)
Electrical Low Pressure Impactor (ELPI) + thermodenuder (TD)	Solid particle sizing and concentration	First 8 channels with filter stage 7nm-1 μm	1 s (transients)
Diffusion Charger (DC)	Active surface	One channel 7nm – 1 μm	1 s (transients)

Table 2 Vehicle sample characteristics

	Lab	Vehicle	Engine Size [l]	Emission Standard	After-Treatment
Diesel	LAT	VW Golf TDI	1.9	Euro I	None
	LAT	VW Golf TDI	1.9	Euro II	OxiCat
	AVL MTC	Peugeot 406 HDI	2.0	Euro II	OxiCat
	LAT	Renault Laguna dCi	1.9	Euro III	OxiCat
	IFP	VW Golf TDI	1.9	Euro III	OxiCat
	FFA	VW Golf TDI	1.9	Euro III	OxiCat
	EMPA	Ford Galaxy TD	1.9	Euro III	OxiCat
	AVL MTC	VW Golf TDI	1.9	Euro III	OxiCat
	Shell	VW Golf TDI	1.9	Euro III	OxiCat
Diesel + DPF	EMPA	Peugeot 406 HDI/FAP	2.0	Euro III+DPF	OxiCat + DPF
	IFP	Peugeot 307 SW	2.0	Euro III+DPF	OxiCat + DPF
	LAT	Renault Laguna dCi	1.9	Euro III+DPF	OxiCat +2xDPF
	AVL MTC	Peugeot 607 HDI	2.2	Euro III+DPF	OxiCat + DPF
	Shell	Peugeot 607 HDI	2.2	Euro III+DPF	OxiCat + DPF
Gasoline Port Fuel Injection	LAT	BMW 318ti	1.8	Euro I	TWC
	IFP	Renault Mégane II	1.6	Euro III	TWC
	LAT	Toyota Corolla TS	1.8	Euro III	TWC
	EMPA	Alfa 146 TS 16V	2.0	Euro III	TWC
	EMPA	RenaultMégane16V	1.6	Euro III	TWC
Stoich. DISI	EMPA	Toyota Avensis	2.0	Euro III	TWC
	Shell	Renault Mégane IDE	2.0	Euro III	TWC
Lean DISI	AVL MTC	Mitsubishi Carisma GDI	1.8	Euro III	TWC/NOx Storage
	IFP	Peugeot 406 HPI	2.0	Euro III	TWC/NOx storage
	Shell	Citroen C5 HPI	2.0	Euro III	TWC/NOx Storage

Table 3 Gasoline and diesel fuels separated according to fuel sulphur content

Diesel		Gasoline	
Fuel	Sulphur content (mg/kg)	Fuel	Sulphur content (mg/kg)
D2	280	G1	143
D3	38	G2	45
D4	8	G3	6
D5	3		

Driving Cycles

The driving cycles used for the development of emission factors were:

- Hot UDC;
- Hot EUDC;
- 2 ARTEMIS Motorway (one reaches maximum average speed of 120 km/h, the other one of 150 km/h depending on the vehicle maximum power)
- ARTEMIS Road
- ARTEMIS Urban
- Steady state 50, 90, 120 km/h

The three real-world ARTEMIS cycles (Figure 1) were build on the basis of a statistical analysis conducted on the real speed patterns measured on board 80 passenger cars, representative of European driving conditions (4). They describe the speed profiles of urban, rural and motorway driving conditions Each of these cycles is actually a compilation of different individual sub-cycles, each characteristic of a special traffic situation within the wider definition. For example the "urban" cycle consists of sub-cycles typical for free-flow or congested urban traffic. These are also shown in Figure 1. For these PM metrics where real-time concentration data were available (active surface and particle number concentration) the emission factors were calculated using the ARTEMIS sub-cycles. In these cases, the speed patterns were divided into parts, characteristic of different driving conditions, and for each part the emission factors were calculated on the basis of instantaneous data.

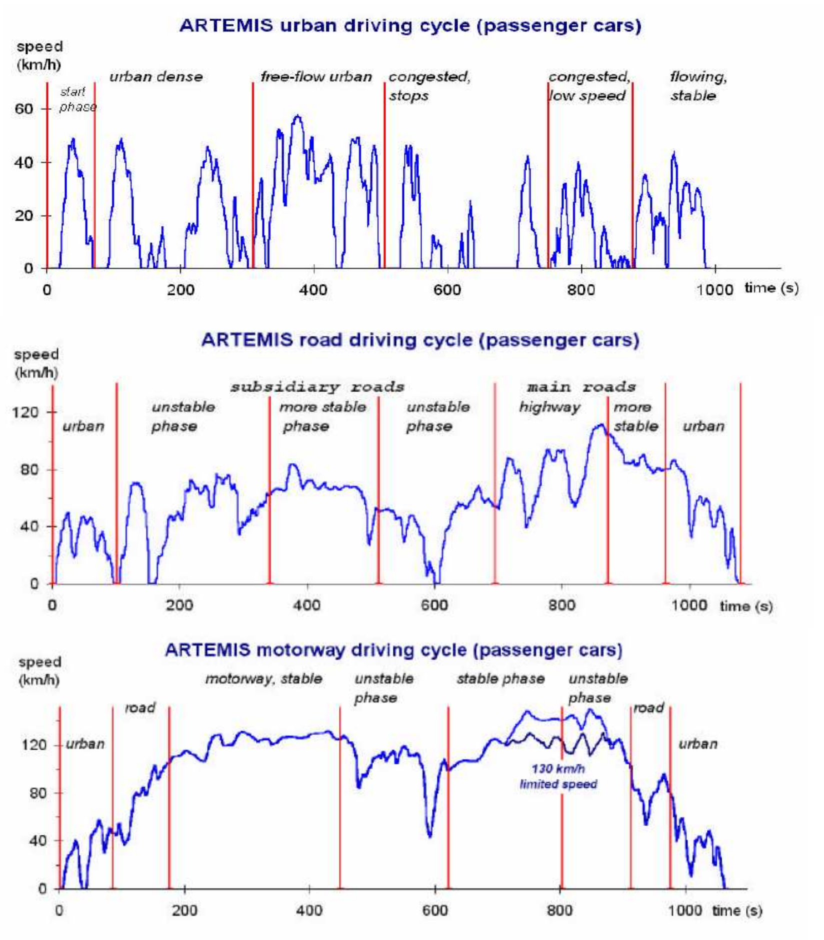

Figure 1 The ARTEMIS cycles and the individual sub-cycles

Results

Methodology

The emission factors were built and evaluated using the following methodological steps:

- Drawing the regression lines for all metrics on the basis of hot cycles and comparing them to the one calculated using both hot cycles and steady states.
- Analysis of effect of vehicle technology improvements and sulphur fuel content on particle emission factors.
- Examination of the statistical validity of the emission factors, by calculating the coefficient of variance and conducting an analysis of variance.
- Investigation of the effect of developing emission factors once with the composite Artemis cycles and once with the individual sub-cycles. For each sub-cycle, new emission factors were calculated on the basis of instantaneous collected data.

Effect of steady state speeds on the emission factors

The comparison between the speed-dependant curves calculated using either only hot cycles or both hot cycles and steady state speeds reveals that differences in estimations are negligible. The only remarkable difference was observed for DPF vehicles and in particular for the total particle number. In this case, the maximum relative variation between the emission factors curve drawn using both hot cycle and steady state and the emission factors curve obtained using only hot cycle is 42%). Steady states for DPF vehicles is a particular case due to nucleation mode formation. However, even in this case, the variability of the emission values within one driving cycle is larger than between the two cases. So, in this paper, it is proposed to consider the emission curves obtained using both transient cycles and steady state speeds.

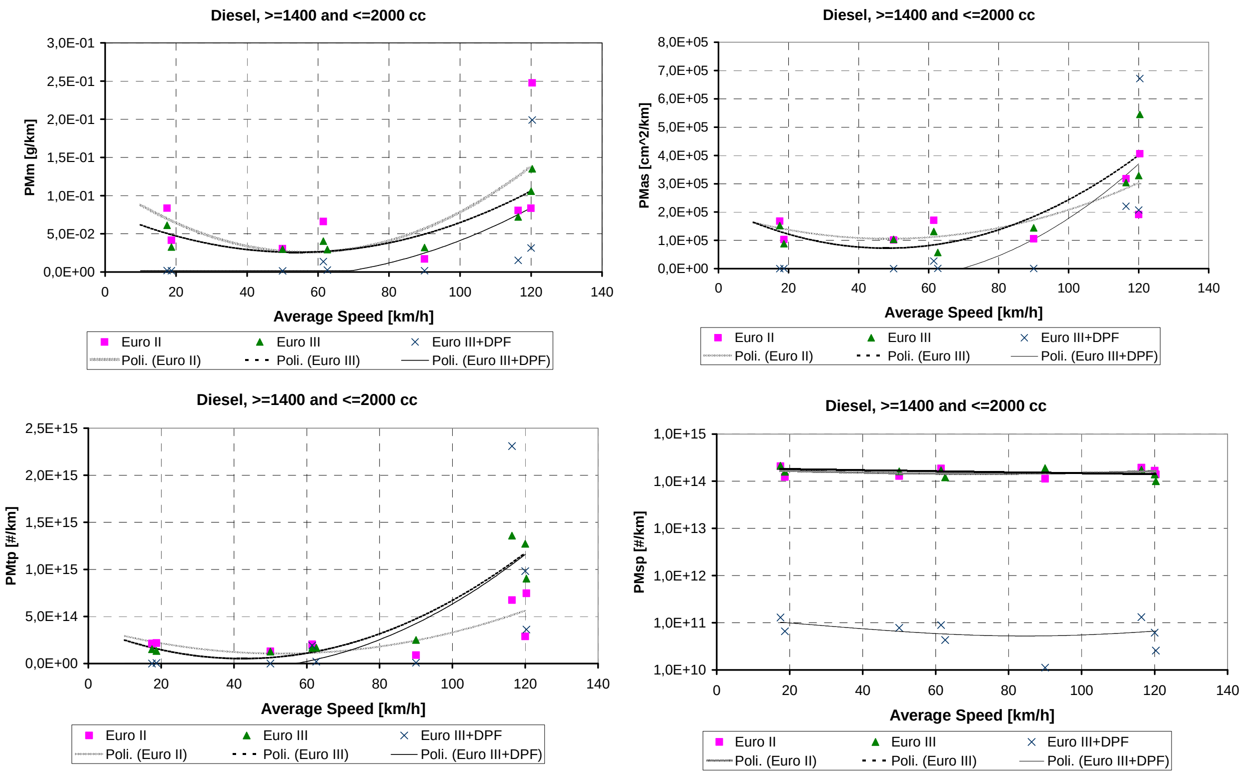

Figure 2 **Speed dependent emission factors for different exhaust particle metrics: mass (PMm), active surface (PMas), total (PMtp) and solid number (PMsp)**

Effect of vehicle emission technology

Figure 2 compares the emission performance of diesel vehicles of different technology, D2 fuelled, using a number of particle metrics. DPF equipped vehicles represent an important improvement in vehicle technology with their emission factors found much below the conventional vehicles, at least for mean speeds below 90 km/h. For high speed cycles and high sulphur fuel though, the DPF vehicle emission factors increase significantly for all particle metrics except solid number, reaching similar levels with older vehicles. This is probably related to the high exhaust temperature related with high speed cycles and the

formation of sulphates under these conditions. Filter regeneration may also contribute to the increase of emission levels. However, solid particle number remains at three orders of magnitude below conventional levels, even over these high speed/temperature conditions.
The shape of the regression lines is similar for all the metrics except solid particle number, showing a minimum in the range 30-60 km/h. The emission levels increase at lower and higher average speeds. Usually the highest values are measured for motorway cycle or for high-speed steady state. The only exception is the solid particle concentration regression line. This metric seems not to be influenced by the driving cycle or Euro level.

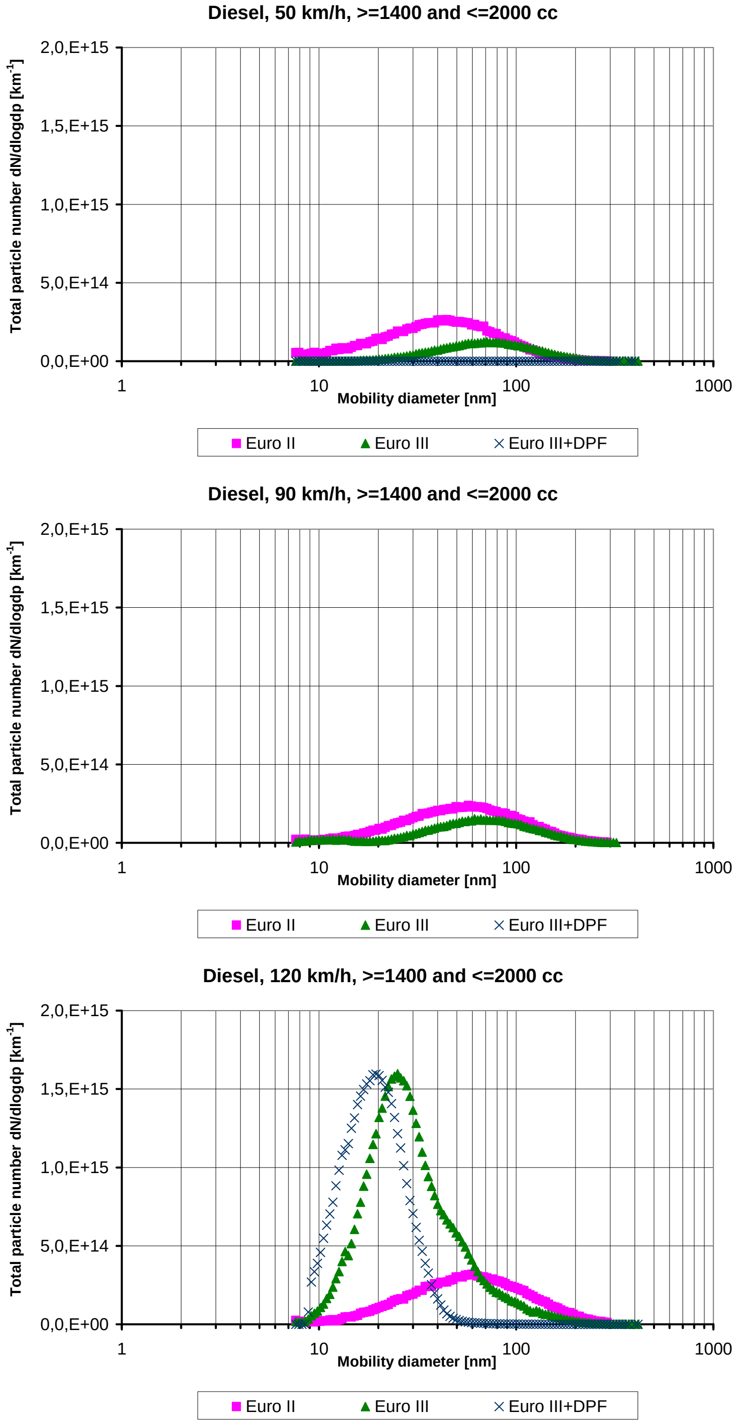

Figure 3 **Particle size distributions of different technology vehicles determined at various speeds**

The particle size distributions clearly depend on vehicle technology for D2 fuelled vehicles, based on measurements conducted at LAT. At low speed, the reduction in particle number concentration is evident as the technology improves. At high speeds though, the EURO III and DPF vehicles present high peaks of nucleation mode particles. For the EURO III vehicle the peak is observed at 30 nm, while for the DPF vehicle at 10 nm (Figure 3). The reasons for these high peaks for low-soot vehicles may on one hand be the limited number of condensation sites which would allow for heterogeneous condensation of volatiles and, on the other, the efficient oxidation of SO_2 to SO_3 by oxidation catalysts at high speeds.

Analysis of variance

In order to explore whether there are significant effects on emissions using DPF, an analysis of variance was applied to the emission factors, considering three speed ranges (<40 km h^{-1}, 40-90 km h^{-1} and >90 km h^{-1}).

The Kruskal-Wallis test, applied to evaluate the differences between the vehicle technology emission factors, revealed that:

- For PM mass, there is a significant difference between the DPF vehicles and the older ones over all speed ranges. The same test, when applied only to the older vehicle categories samples shows that the differences between their emission factors means (these groups) are not significant.
- For particle active surface, there is a significant difference between emission factors of the newer vehicles and the older ones for speeds lower than 90 km/h. At higher speed the statistical test reveals no significant differences between technologies.
- For total particle number, there are still significant differences between DPF equipped vehicles and conventional one for the three speed ranges ones. In this case, there is a significant difference also between conventional vehicle technologies for speeds lower than 40 km h^{-1} and higher than 90 km h^{-1}.
- For solid particle emission factors the test confirms the difference between DPF equipped vehicle and the conventional ones.

The coefficients of variance of emission factors change on the basis of EURO level, driving cycles and PM metrics. In general, the highest values of the coefficient are detected for DPF equipped vehicles: this could be because the same vehicle could operate either at normal mode or trap regeneration mode for different repetition of the same cycle. The most variable DPF emission factors relate to the active surface and the total particle number concentration: the maximum level of variability is 200% and 160% respectively. For all metrics, the average coefficient of variance of EURO II and EURO III vehicles, calculated using all cycles, are similar, included the interval 20-40%. Additionally, there is not clear dependence of the variability on driving cycles because the coefficients calculated for different metrics on the same cycle are different. The only exception is the similarity of variance calculated for active surface and total particle number : for both the highest values are recorded for lower speed cycles.

Effect of fuel

Evaluating the effect on emissions of decreasing sulphur content in the fuel is not straightforward. In general, the improvement of reduced sulphur is always evident for high-speed cycle and all metrics (except for solid particle concentration): the D2 emission levels are higher than the one measured with other fuels, while the emissions levels with D3 and D4 are quite similar. D5, also having a difference chemical character on emissions, also affects solid particle number concentration. In particular:

- With regard to PM, D2 has significant effect on emission over high-speed conditions (more than 90 km/h) for all vehicle technologies. Al lower speeds, an overlapping between emission factors for different sulphur content is observed. Emission factors for D3, D4 and D5 fuels reach similar values.
- Similar to the mass, the effect of using high sulphur fuel on active surface is explicit at high-speed, while an overlapping of emission levels with different fuels is observed at lower speeds. However, for DPF vehicle some emission factors reached high values

even using lower sulphur fuels at high speed cycles. It has to be considered that there is a wide spread of DPF emission factors, even using a low sulphur fuel, due to the different operation mode of the vehicles during the different repetition of the same cycles.

- There is an increase of number of total particle emitted when D2 fuel is used over the high-speed cycle. Like for the others metrics, the emission level for all fuels is similar at the low-speed conditions. For not DPF equipped vehicles the emission obtained using D3, D4 and D5 are similar. For DPF vehicle, the emission for fuels with low sulphur content are similar for lower speed, while for motorway cycle the emission level using D4 increases.
- For solid particle the effect of using fuels with different sulphur seems to be negligible.
- For EURO III and DPF vehicle the effect of decreasing fuel sulphur content is evident on size particle size distribution at 120 kmh^{-1}: the number of particle extremely decreases for smaller size diameters (lesser than 50 nm), especially using D4 fuel . For EURO II vehicle and for the other steady states an overlapping of the size distribution is observed using different fuels.

<u>Effect of sub-cycles on emission factor estimation</u>

In order to obtain more detailed functions of emission factors with average speed, the ARTEMIS cycles were analyzed in their individual sub-cycles. In this way, it is possible to increase the number of data to calculate the regression line: for every new cycle there will be a new emission factor. In total 16 sub-cycles are used, covering a wide range of mean speeds from 9 to 134 km h^{-1}, including different traffic congestion levels.

Figure 4 shows that the emission factor functions obtained using sub-cycles are similar to the these calculated using only the composite cycles. A change is observed for DPF vehicles where the function slope for the active surface area and the total particle number is less marked when sub-cycles are used. This is only related to the use of larger number of points in developing the functions. The second difference is observed for the active surface of EURO IV vehicles. However, this was due to differences in the input data because it was not possible to obtain real time data for one laboratory (AVL MTC).

Summary and Conclusion

Emission factors for emissions of several particle related metrics are given for a number of diesel passenger cars operating on four different fuels. The results show that it is possible to construct speed dependant functions for the emission factors, the same way done in the past for conventional gaseous pollutants. The shape and level for each function depends on the metric considered as different phenomena are responsible for the formation of solid and volatile particles. It is expected that such emission factors can be used as input to air quality models to eventually relate ambient emission levels with exhaust aerosol concentrations.

Narrow

References

1. G. Obersdörster, ‘Toxicology of ultrafine particles: in vivo studies’. Philosophical Transaction of the Royal Society of London, A 358, 2719-2740.
2. L. Ntziachristos, Z. Samaras, “COPERT III. Methodology and emission factors (Version 2.1)”, Technical report No 49, European Environment Agency, 2000.
3. L. Ntziachristos, Giechaskiel, B., Pistikopoulos, P., Samaras, Z., Mathis, U., Mohr, M., Ristimäki, J., Keskinen, J., Mikkanen, P., Casati, R., Scheer, V., Vogt, R. "Performance evaluation of a novel sampling and measurement system for exhaust particle characterization", SAE Technology Paper 2004-01-1439.
4. ARTEMIS, “Assessment and reliability of Transport emission Models and Inventory systems”, European Commission, 2003.

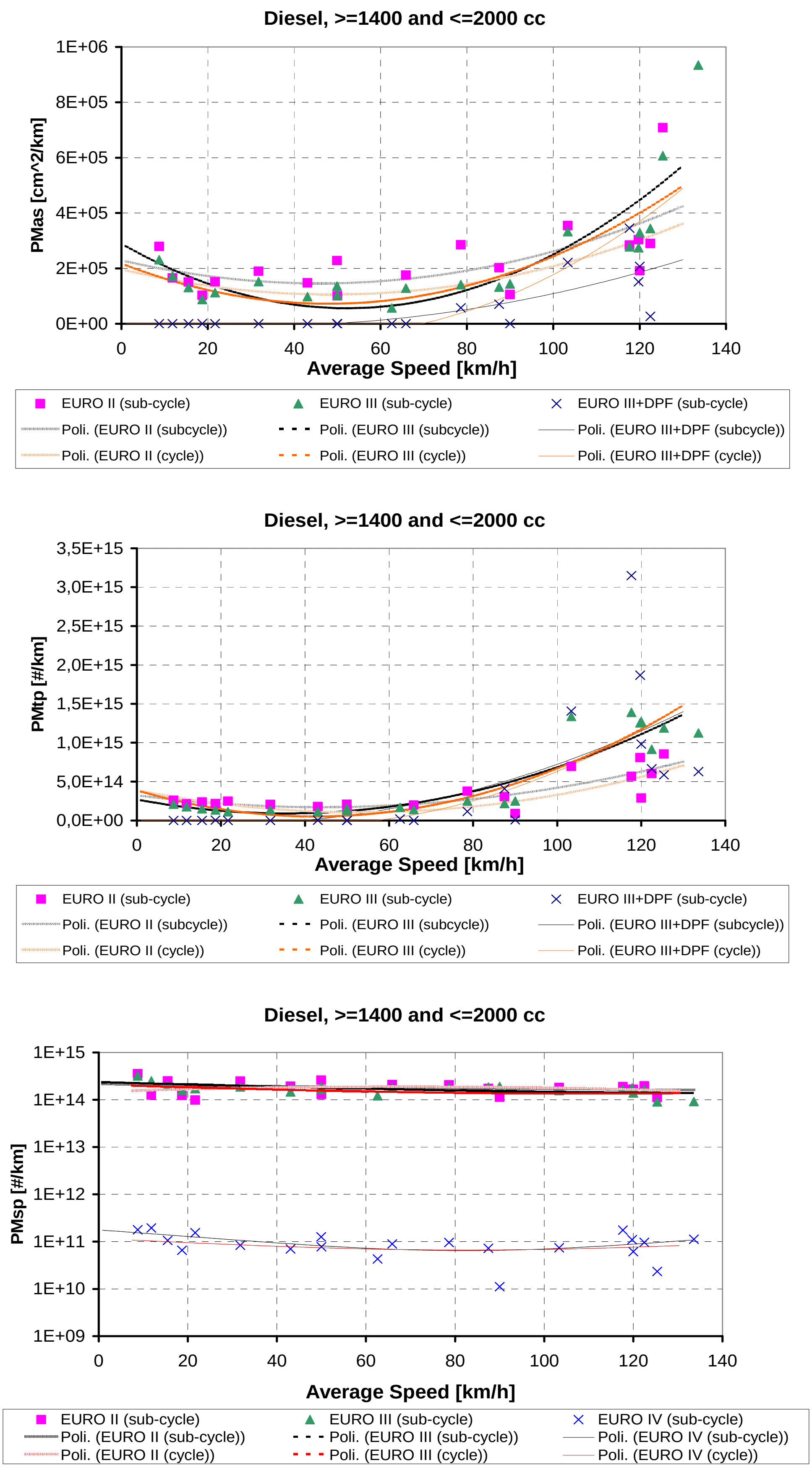

Figure 4 **Comparison between the speed-dependent emission factors functions obtained using the composite Artemis cycles and the individual sub-cycles.**