Particulate matter produced by compression ignition engines has a significant adverse impact on public health. Traditionally, particulate matter emissions from vehicles have been assessed using mass-based techniques. European Union legislation has steadily reduced the mass-based emissions limits for Diesel cars, to the point where the legislation effectively mandates the usage of a Diesel particulate filter, commonly referred to as a DPF. The Euro 5 limit of 5 mg/km (soon to be reduced to 4.5 mg/km, but using a revised measurement method which tends to generate slightly lower results) represents an 80 % reduction from the previous limit for Euro 4 CI vehicles.

In spite of the rapid reduction of the particulate mass emission limit, toxicological research performed over the past two decades has revealed particle number, size and surface area to be attributes more closely correlated with the health effects of particulates. The Euro 5+ standard introduces a particle number limit, which is widely recognized as being more stringent than the mass limit. The particle number limit has no precedent for comparison, but can be considered to represent a reduction in particle number concentration of >>99 %, compared to unfiltered exhaust. The existence of a correlation between the number of particles emitted and the mass collected on the filter is of interest for a number of practical engineering reasons. However, the correlation of these two metrics is problematic, and many studies have failed to find any meaningful correlation [e.g. 1].

It is possible to speak of a correlation between particle number and particle mass [2], and these metrics are sometimes observed to correlate in samples of ambient air from polluted urban settings. However, such correlations are not always observed; the ambient mass concentration reveals relatively little about the ambient number concentration. Correlation of particle mass and particle number in automotive exhaust is problematic for a variety of reasons. Firstly, as defined in the legislation [3], the particle number metric does not quantify certain types of particles, whereas all particles contribute (somewhat) to the total particulate mass. The particle number counter’s sampling probe is deliberately designed to exclude some large particles, and the system features a tube heated to between 300 °C and 400 °C to vaporize all volatiles and prevent them being included in the total particle count. Ideal measurement conditions for optimal quantification of number and mass do not overlap entirely, and the particulate mass and particle number sampling may take place at slightly different locations within the dilution tunnel. Before the sample is pre-treated in any way, it can be assumed that particle mass will be conserved [2]. Conversely, particle number can be radically and rapidly reduced through agglomerational processes [2]. Thus, the numerical relationship between number and mass is likely to be different at each of the following points: within the cylinder, immediately downstream of the DPF, in the dilution tunnel or ambient air (following dilution). Therefore, knowledge of the relationship at any one of these points provides little insight into the relationship at any of these other locations. Additionally, gravimetric measurement techniques may not be sensitive enough to produce meaningful results if the exhaust has passed through a high-efficiency DPF. Lastly, the massive disparity of the magnitude of the two metrics...
(often a factor of tens of billions) complicates mathematical analysis and restricts precision to orders of magnitude only.

In order to further investigate filtered particle emissions phenomena, a series of tests were performed in BOSMAL’s climate-controlled Euro5/6 exhaust emission laboratory on a range of Euro 5 compression ignition vehicles, all equipped with particulate filters. In total, 35 vehicle-aftertreatment combinations were tested on a chassis dynamometer over the New European Driving Cycle, with exhaust gas passing through a dilution tunnel to facilitate particle number quantification by a condensation particle counter and mass quantification by filtering diluted exhaust through TX40 foil-backed filters. As the ageing, dimensions and chemistry of the aftertreatment systems varied, a broad range of emissions results were collected, covering around 4 orders of magnitude of particle number and 2 orders of magnitude for mass.

At higher emission levels, strong positive correlations were found for two vehicles, both of which revealed a correlation of $3 \times 10^{12}$ particles/km per mg/km (Figure 1). This value is broadly similar to a correlation reported recently [4]. The observed correlation further confirms that the particle number limit is over an order of magnitude more stringent than the mass limit. The higher-emission vehicles displayed a range of results; the strength of the correlation was generally proportional to the mass emission result. Mass emission factors below $\sim 1$ mg/km generally correlated relatively poorly, indicating a severe lack of sensitivity of the gravimetric method below this level.

![Figure 1: Correlation between particle mass and particle number for a higher-emission vehicle, tested with a range of aftertreatment systems over the New European Driving Cycle](image)

For low-emission vehicles, no overall correlation was found (Figure 2). This finding is in line with certain other studies which have failed to find correlations when investigating vehicles equipped with DPFs. However, despite the absence of a correlation for the low-emission vehicles (with a range different aftertreatment systems), comparing the number and mass values for each phase of the New European Driving Cycle yielded linear correlations with widely varying gradients, some of which were negative (Figure 2).
These varying yet strong correlations were observed for two lower-emission vehicles, over a large number of tests. This phenomenon suggests that the properties of the particles emitted can vary significantly – to the extent where the usual positive correlation between mass and number can even be reversed. While any negative correlation between number and mass appears highly counterintuitive, the aforementioned caveats that apply to comparison of the two metrics may go some way towards explaining this unusual finding.

The overall steep, positive correlation confirms that measures to reduce particle number emissions will rapidly reduce mass emissions. This fact is significant, as for very low emission vehicles the mass method can be insufficiently sensitive to accurately quantify emission. However, the correlation process remains problematic, and further work is required in this area. The future introduction of particle mass and number limits for other engine types will raise a great many more questions regarding the numerical relationship between mass and number in automotive exhaust.

Select references:

Traditionally, particulate matter emissions from vehicles have been assessed using mass-based techniques. The Euro 5+ standard introduces a particle number limit, which is widely recognised as being more stringent than the mass limit. However, this has been problematic, and many studies have failed to find any meaningful correlation [e.g. 1]. In order to further investigate filtered particle emissions phenomena, a series of NEDC tests were performed in BOSMAL’s climate-controlled Euro5/6 exhaust emission laboratory on a range of Euro 5 compression ignition passenger cars, all equipped with particulate filters (see Figures 1-4b). At lower emission levels, strong positive linear correlations were found for two vehicles, both of which revealed a correlation of $3 \times 10^{12}$ particles/mg (Figure 5) over the NEDC, broadly similar to a value reported in a recent study [2]. Relationships of varying gradient were found for the two phases of the NEDC (Figure 6). This correlation confirms that the PN limit is over an order of magnitude more stringent than the PM limit. For lower-emitting vehicles, no overall linear correlations with widely varying gradients, some of which were negative (Figure 8).