Vehicles Exhaust Particulate Matter Emissions

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In the automotive field the term “particulate matter (PM)” is used for the collected matter on a flow-through filter under specific conditions, and the term “particle” for aerosol particles measured while airborne (suspended matter). Particles are divided into “volatile” and “non-volatile” (or solid) at tailpipe conditions (high temperature, high concentration). Species that at tailpipe conditions appear volatile, may partition toward the particulate phase at atmospheric conditions (low temperature), and the term semi-volatile better characterizes them. The term “semi-volatiles” (instead of “volatiles”) will be used loosely in this text to indicate species not counted after dilution and thermal pre-treatment at 300–400 °C. The term ultrafine particles (i.e., particles < 100 nm) is not so common in the automotive community. Even though the majority of particles has sizes <100 nm, the tail extends to larger sizes. A recent review argued that a better definition for ultrafine particles (focusing on the automotive field) would be particles <500 nm.

primary aerosol fresh aerosol secondary aerosol nucleation mode vehicle emissions road transport urban pollution air quality PMP PEMS

1. Introduction

The atmospheric aerosol is a complex and dynamic mixture of solid and liquid particles in the air, generated from natural (such as pollen, sea salt, volcanic ash, and soot particles from natural fires) and anthropogenic sources (e.g., combustion, waste incineration, and road abrasion). Anthropogenic emissions of atmospheric aerosol and its precursors have increased over the past century and are known to have significant impacts on human health [1][2] and climate change [3]. Studies have actually shown that the particulate matter (PM) has a greater impact on health than the gaseous components [4][5]. Globally, >50% of the population lives in urban areas with poor PM air quality [6].

Ambient PM concentrations are monitored in many places around the world, together with several gaseous species. Traffic is an important source of PM and the exhaust emissions of vehicles have been regulated for many years. In the last decade, a solid particle number (SPN) limit has also been imposed in the European Union (EU) legislation [7] and other countries in Asia have followed. There are a few concerns about the representativeness and the usefulness of the SPN limit, as it is today proposed: (i) exhaust and atmospheric particles usually extend to sizes much lower than the cut-point of 23 nm currently included in the regulations; (ii) only solid particles following a thermal treatment are regulated, while atmospheric include additional higher volatility species [8]. Thus, the

current standard does not address particles which are representative of real-world exposure to traffic particulate emissions [9][10].

2. PM Emitted from Vehicles

This section will focus on the physical characterization and chemical composition of the PM emitted by vehicles, along with the potential for secondary aerosol formation.

2.1. Primary "Tailpipe" Particles

Figure 1 (left lower corner) plots schematically the exhaust aerosol at the tailpipe and the atmosphere within a few seconds (i.e., primary and delayed primary aerosol) [11][12]. It also shows sampling and measurement (left part), as well as: (i) the resulting PM mass and chemical characterization (lower right part) of a filter [13]; and (ii) particle number size distribution before and after thermal pre-treatment (upper right part) with a solid particle number (SPN) instrument [14].

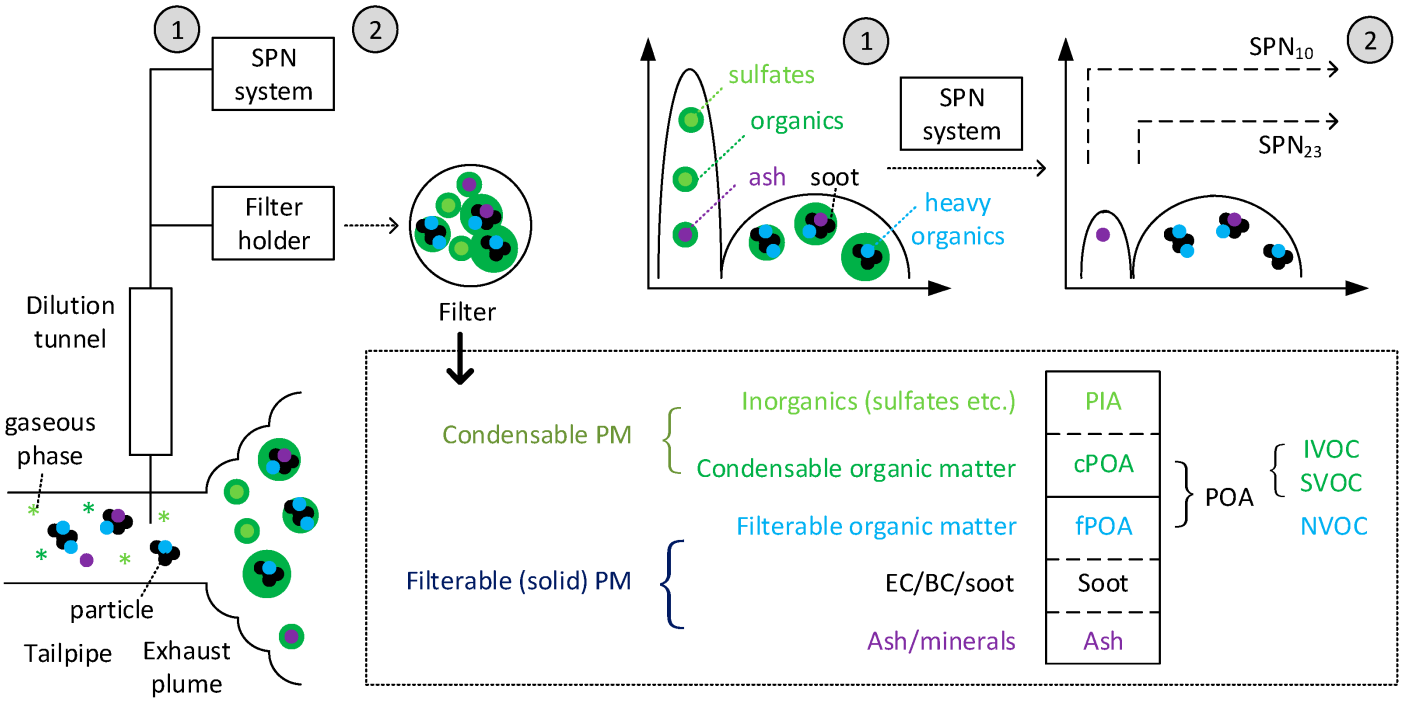

Figure 1. Tailpipe "primary" and fresh aged "delayed primary" aerosol consisting of inorganics, organics at the gaseous or particle phase, and soot and ash particles. On the right side the filter based and solid particle number (SPN) counting methods are plotted. Based on [11][12][13][14]. cPOA = condensed primary organic aerosol; IVOC = intermediate volatility organic compound; NVOC = non-volatile organic compounds; PIA = primary inorganic aerosol; POA = primary organic aerosol; PM = particulate matter; SPN = solid particle number; SVOC = semi-volatile organic compound.

At the vehicles' tailpipe, only primary non-volatile (solid) particles can be found due to the high exhaust gas temperatures, while the majority of precursors are in the gaseous phase (i.e., "semi-volatiles" in this paper). Typically, a soot or accumulation mode is measured at the tailpipe with a mean size >50 nm [15]. Studies of modern gasoline vehicles have found smaller means, around 30 nm [16][17]. The accumulation mode particles consist of many spherules (sometimes called primary particles, a term that will not be used in this text) of elemental carbon [18] with fuel and lubricating oil components [19]. Particle-bound polycyclic aromatic hydrocarbons (PAHs) are also commonly reported [20][21][22][23][24]. A solid core or nano mode with a mean size of 10 nm has also been found [25][26]. This mode consists of amorphous carbonaceous compounds, PAHs, or metallic ash from fuel or lubricant [27][28][29][30][31][32][33]. In one case two separate core modes were found (derived from lubricant and fuel respectively) [34]. Recently, urea non-volatile particles have also been reported at sizes around 20 nm [35][36]. Non-volatile clusters <3 nm have also been reported for compressed natural gas (CNG) engines [37][38]. The modes depend, among others, on the engine, fuel, combustion strategy, and aftertreatment devices [11][39][40][41]. Recently, in addition to the combustion process related solid particles, nanoparticles during braking (motoring) have been reported, even when no fuel injection and combustion process take place in the cylinder [42][43][44]. Engine and aftertreatment wear particles can also been found [45]. The introduction of particulate filters has reduced significantly the concentration of primary particles in the exhaust gas of modern vehicles [7][46]. Sometimes, larger coarse mode particles appear originating from the crankcase ventilation, wear, or soot re-entrainment [47][48]. Regarding the precursor gases, oxidation catalysts reduced hydrocarbons, but in many cases increased the SO_2 to SO_3 conversion and NH_3 [49][50]. NO_x reduction aftertreatment decreased NO_x [51].

2.2. Delayed Primary "Fresh" Particles

At the tailpipe outlet the aerosol is diluted and cools down [52][53][54]. The precursors (e.g., sulfuric acid, hydrocarbons) [55][56] that were in gaseous phase at the tailpipe (due to the high exhaust gas temperatures) may nucleate to form new nucleation mode particles or condense on other particles (e.g., non-volatile core or accumulation mode). Thus, the "fresh" exhaust aerosol comprises the solid particles in the tailpipe (primary PM) and the newly formed particles during the seconds of mixing of the exhaust gas with ambient air [11][57][58] (**Figure 1**).

The formed nucleation mode (in the absence of a solid core) peaks at approximately 10 nm depending on the availability of the precursors [47][59][60][61]. Sulfuric acid is the key nucleating compound as measurements [62] and models show [63][64][65][66]. With low sulfur fuels, lubricants play an important role [62][67] and aftertreatment devices enhance the SO_2 to SO_3 conversion [49]. Relatively high SO_2 concentrations can be measured at diesel vehicles compared to gasoline and gas engines due to the higher oxidative environment in the exhaust [68]. Hydrocarbons are then necessary for the subsequent growth of such sulfate core particles [26][69][70]. A nucleation mode can be typically seen with high sulfur fuel (300 ppm) and/or lubricant [62][71], high speeds (exhaust gas temperatures) [49][72], and during regenerations [73][74] (see also discussion in [75]). Without aftertreatment devices, hydrocarbons (alkanes, PAHs) may also form a separate nucleation mode [60]. Without any aftertreatment devices, this nucleation mode has high particle number concentration and large mean size [76], but with aftertreatment devices the concentration and size is usually low [77]. Both fuel and oil are significant sources of hydrocarbons [23][33][78][79][80][81]

[82][83]. Fuel contributes to volatile organic compounds (VOC) and intermediate volatility organic compounds (IVOC), while oil to semi-volatile organic compounds (SVOC) [84]. More on chemical composition of vehicles exhaust can be found elsewhere [39][85][86][87][88].

At this primary atmospheric dilution stage no significant chemical transformations take place. When looking the complete particle size distribution, the exhaust aerosol formed by different processes is frequently allocated to separate modes with different concentrations and particle size ranges [47]. The size distribution may consist of a cluster mode, one or two core modes, the soot mode and the coarse mode. The formation and properties of each mode (size, chemical composition) depends on the vehicle (engine, aftertreatment, fuel, lubricant) [59][89][90][91][92], driving conditions and the ambient conditions (temperature humidity) [8][93]. Some of the modes may often appear blended and are difficult to distinguish, unless combined with thermal separation techniques, such as treatment with a catalytic stripper or thermodenuder, followed by microscopy.

In terms of mass, under laboratory conditions the soot and ash particles and heavy compounds comprise the solid part (see **Figure 1**). Primary organic aerosol (POA) and primary inorganic aerosol (PIA) compounds condensed on particles or as a separate nucleation mode consist the condensable PM. It should be mentioned that the compounds that are emitted in the gaseous phase under relevant atmospheric conditions are secondary organic aerosol (SOA) precursors.

Vehicular emissions are eventually diluted by a factor of 1000 or more in the atmosphere [94], but dilution near roads may be lower [95]. A study that summarized measured dilution ratios in function of distance from moving vehicles, reported dilution ratios of 200–500:1 at a distance of 10 m [75][96]. Other researchers provided equations to estimate the dilution ratio [97]. At lower speeds the dilution in the wake of the vehicle can be much higher at the same distance [98][99]. Measurements at different short distances behind moving vehicles (10–50 m) did not find significant evolution of the nucleation mode, after correcting for dilution [96]. In the time scale of a few seconds this nucleation mode seems stable. However, these particles may evaporate after some time in the atmosphere and may subsequently contribute to the formation of secondary aerosol of higher mass [100][101][102]. For example, some studies found that the organic to total carbon ratio is higher for roadside nanoparticles compared to typical exhaust soot particles [103]. On the other hand, other studies found an increase of the particle size from 1.5 m to 15 m from the road [104][105]. Models usually divide the analysis into separate ‘tailpipe to road’ and ‘road to ambient’ parts [6][106]. Most of the changes to particle number concentration and size distribution occur rapidly with the dilution and cooling of the exhaust gas. Later, interaction of fresh particles with relatively aged particles also takes place [107].

2.3. Secondary Particles

In addition to the primary and delayed primary PM, large amounts of secondary particulate matter forms after the exhaust gases are released into the atmosphere [108]. After some hours or days under atmospheric conditions, secondary PM is formed due to oxidation of gaseous precursors. Studies of secondary aerosol formation rate and quantity (yield) are done in smog chambers [109] or oxidation flow reactors [110].

Recent chamber studies have shown that secondary particulate matter from combustion engines consists mainly of organic compounds and ammonium nitrate [111][112][113] and that the secondary PM formation can be significantly larger than primary PM emission [112][114][115][116][117]. Studies that focused on the SOA also found that the contribution of vehicles to SOA can be higher than primary aerosol [112][118][119]. The emissions of secondary PM precursors from internal combustion engines depend on fuel properties [120][44][111][113][121][122][123]. The advantages of the oxidative exhaust aftertreatment and especially the use of diesel particulate filters (DPF) on decreasing aged PM has clearly been shown [124][44][89][122][125][126][127][128]. Catalyzed DPFs on road vehicles were also demonstrated to yield very low SOA over transient operation. Gasoline engines have greater secondary aerosol precursor emissions than diesel engines [124][112][121][125][128][129][130]. However, it was also shown that SOA emissions follow emission standards [116] and recent Euro 6 gasoline vehicles had very low SOA [10]. Gasoline particle filters (GPF) have not shown similar reduction potential against SOA as DPFs [131]. For a natural gas engine, the mass of the aged particles produced by an oxidation flow reactor was hundreds of times higher than the mass of primary particles [91]. Cold start emissions have been shown to have a significant contribution to SOA [10][116][132][133]. These studies highlight the need to better characterize (semi)volatile compounds from vehicles in order to better estimate their contribution to secondary aerosol formation.

2.4. Emission Levels of Solid and Volatile Particles

The introduction of a solid particle number (SPN) limit clearly resulted to a significant decrease of vehicle emissions. For example, DPF vehicles have by a factor of >10,000 lower SPN emissions than vehicles without DPFs (from $>10^{14}$ #/km to $<10^{10}$ #/km) [7]. Similarly, SPN emission levels of gasoline direct injection vehicles dropped from $>10^{12}$ to $<10^{11}$ #/km [134] with the use of a gasoline particle filter (GPF). In contrast, no SPN emission reductions were observed for vehicle technologies that were not covered by relevant regulations. For example, the SPN emissions of port fuel injection vehicles have remained at the same level (mostly between 10^{11} and 10^{12} #/km) for the last 30 years [134]. Mopeds and motorcycles also exhibit high SPN emissions which, depending on engine tuning, can often reach more than 10^{11} #/km [135]. Any decreases in emission levels of this category were attributed to technology improvements (two-stroke vs. four-stoke, carburetor vs. electronic injection) which were forced by stricter limits in gaseous pollutants [136]. This is an example that demonstrates that satisfactory control of SPN can be achieved by regulating co-pollutants.

One question is how different formation mechanisms of solid and total particle emissions are; this would have an impact on the emission control technologies in each case. In general, SPN control does not necessarily result in a decrease of total particle number (TPN), because the semi-volatile part is formed by ions and organics while the solid part is mostly elemental carbon and ash. In an exaggerated example of the past, a DPF equipped engine was shown to result to higher particle number emissions than the non-DPF one [137]. Later it was shown that due to the low soot concentration post DPF available volatile species preferentially nucleated and formed new particles in the absence of solid cores on where they could condense [138]. Similar findings have been observed in the atmosphere where high particle number levels can be seen when PM is low [139]. A recent study with 130,000 plume measurements found that the number of semi-volatile particles comprised 85% to 94% of total particles [8]. Even though semi-volatile particles can be dominant in terms of number, their contribution to mass depends on the

existence or not of a particulate filter. Detailed studies with heavy-duty engines equipped with aftertreatment devices to fulfil the 2007 and 2010 standards (i.e., oxidation catalyst, DPF and selective catalytic reduction (SCR) for NO_x) had elemental carbon <20% of total mass [87][88]. The organic carbon on the other hand was 30–65% and the rest were sulfates and nitrates. A constant nucleation mode over a test cycle (10^{13} #/km, e.g., 10^7 #/cm^3 with mean size 20 nm) would correspond to only 0.1 mg/km (<10 μg on the filter at the end of the cycle). To put these numbers into context, the current SPN limit is 6×10^{11} #/km, with the mass limit at 4.5 mg/km. For heavy-duty vehicles, the same number concentration (10^7 #/cm^3) would translate to >5 times higher emissions due to the higher exhaust flow rate.

There is a significant body of studies that have measured both solid and total number concentrations. For example, large projects funded by the industry [140], and the European Commission, such as the Particulates project which ended in 2004 [141], showed small differences between TPN and SPN at low speeds, but high at high speeds. Other smaller scale studies reported differences of 50–100% between TPN and SPN for Euro1–4 vehicles [74] or recent Euro 5 and Euro 6 [142] or 2009–2012 model years [143] for typical cycles. A review showed that for type approval cycles the trends for solid particles were followed also for total particles (i.e., decreasing for GDIs, no decrease for PFIs) [134]. This decrease is not always so evident in real life [59]. For example, during cold start of gasoline and gas engines nucleation mode particles can be formed [144][145], but not always [146]. Relatively high differences have been reported when fuel specifications change [133][147], and at high speed cycles [143][148][149]. Even for the same vehicle and fuel, different operating points can result to varying TPN/SPN ratios [150][151]. Recent research projects, such as the DownToTen (DTT), which ended in 2020, presented results from many vehicles where the TPN emissions were more than one order of magnitude higher than the SPN [152]. Of particular interest were cases such as gasoline vehicles with GPF, compressed natural gas (CNG) vehicles with and without particulate filter and plug-in hybrids that all under certain conditions exhibited a large range of TPN/SPN values. Another study found more than one order of magnitude higher TPN than SPN for hybrid vehicles even at city driving [153]. All studies mentioned conducted measurements directly from the tailpipe so any volatile particles cannot be attributed to desorption artifacts from the sampling lines.

Similar conclusions have been also drawn for heavy duty engines [154]. A study showed that the total particle number emissions increased from 10^{11} #/km to 10^{13} #/km when the exhaust gas temperature was >310 °C [155]. In general, due to the high exhaust gas temperatures and consequently high release of desorbed species from the aftertreatment devices and exhaust line, and high SO_2 to SO_3 conversion, high TPN concentrations are reported [36][49][156]. Tests with L-category vehicles (e.g., mopeds and motorcycles) also resulted in high TPN, especially at high speeds [135][157]. Different combustion technologies (e.g., temperature reactivity controlled compression ignition (RCCI), hot or low exhaust gas recirculation (EGR) combustion etc.) can also have various TPN to SPN ratios [158].

Specific events, such as DPF regeneration can also produce high concentrations of both solid and semi-volatile particles [155][159][160][161][162][163]. SPN emissions can reach or even exceed the limit of 6×10^{11} #/km [160][163], while total particle emissions can be one to three orders of magnitude higher (up to 2×10^{14} #/km) [73][160][163]. Studies with light-duty and heavy-duty vehicles have also shown that even when the emissions during regeneration events

are considered, the weighted (over regeneration distance) solid particle number emissions remain below the current SPN limit [36][164][165]. The regeneration frequency is on average around 400–500 km, with a tendency of shorter distance for newer vehicles [166]. Regarding semi-volatiles, many studies have shown that the concentration of sulfates and organics in the exhaust increases during regenerations and this is often linked to the formation of a distinct nucleation mode [73][163][167]. However, one study found that increased particle number emissions during DPF regeneration were still by 83–99% lower than those without DPF [90]. Furthermore, considering the regeneration frequency, the apparent total particulate matter filtration efficiency was reduced by less than 2% over the average driving conditions for medium- and heavy-duty diesel vehicles [125]. Still, the weighted (total) particle number concentrations over the regeneration distance can be up to one order of magnitude higher than the current limit for solid particles.

The collected evidence suggests that there can be technologies, fuels, and operation conditions that lead to SPN and TPN levels and trends exhibiting significant deviations. The same evidence also suggests that the metric chosen for regulatory control may influence which technologies are promoted for future vehicles and what specifications for fuels and lubricants are decided. All of these factors will have an impact, not only on the specific metric, but on other co-pollutants as well. Therefore, deciding on the proper metric for particle number control will be decisive for the wider environmental impacts of road transport.

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