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SUSTAINABILITY OF HYDRATED ETHANOL FUEL FROM THE PERSPECTIVE OF PARTICLE NUMBER EMISSIONS ON FLEX-FUEL VEHICLES

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ABSTRACT

In Europe, the EURO 6 Light Duty emission standards, being a consequence from Diesel vehicles, introduced limits for particle number (PN) of Gasoline Direct Injection (GDI) engines. Given the concern of the extremely small particles on health effect, the same limits were set (6.0×10^{11} #/km) for both diesel and GDI cars. In Brazil, the current phase for light vehicles of PROCONVE L6 enforced particulate matter (PM) mass emission standard for Diesel vehicles only, applying the limits of 25 mg/km for passenger cars and 30 mg/km for commercial vehicles. Similar to the European concern, even not considering yet the particle number (PN) emissions, future Brazilian phase L7, which is under study, considers a significant reduction on the limit of the particulate matter (PM) mass emission, from current limits down to 6 mg/km for both Diesel and DI engines, for passenger and commercial vehicles. Aiming for contributing on hydrated ethanol fuel sustainability discussion compared to gasoline and for generating reference data regarding particle number (PN) emissions which can be used for future emissions regulations of Flex-Fuel vehicles in Brazilian market, this article explores particle number (PN) measurements of Flex-Fuel Vehicles equipped with direct injection (DI) and port fuel injection (PFI) engines, fueled with ethanol (E100) and gasohol (E22). The methodology followed the Brazilian standard driving cycle NBR6601(EPA75) in a standard vehicle emission laboratory configured with Particle Number Counter (PNC) model AVL 489, according to UNECE Regulation 83 (Light Duty Vehicles). The results show that ethanol (E100) fuel produces significantly less particle number (PN) emissions than gasohol (E22), about 90% and 30% less on DI and PFI engines respectively.

The theoretical portion explores the particulate matter concerning definitions, sources, characterization, regulamentation, impacts to human health and mitigation. Also, it highlights the advantages that Ethanol fuel usage brings to human health compared to gasoline.

Keywords: Ethanol, Sustainability, Particle Number, Particle Matter.

1. INTRODUCTION

World Health Organization declared on its update (WHO, 2016) that more than 80% of people living in urban areas that monitor air pollution are exposed to elevated levels of particulate matter that exceed the air quality limits. This update on air quality database registers that only 2% of cities in low and middle-income countries with more than 100,000 inhabitants meet WHO air quality guidelines. In high-income countries, the scenario increases to 44%. Similarly, according to Irish Environmental Protection Agency, the WHO estimates show that more than 400,000 premature deaths are attributable to poor air quality in Europe annually. Locally in Brazil this matter is not different, the numbers of deaths are also scary. Figures from the Institute for Health Metrics and Evaluation, 2016 related to 2015 showed that the cause of 52,284 deaths were related to exposure to particulate matter of fine particles, $PM_{2.5}$ (Dallmann, 2017). Still according to (WHO, 2014) the Ambient Air Quality Database reports annual average $PM_{2.5}$ concentrations in 40 Brazilian cities. It shows that only one city is below the WHO air quality guideline of 10 µg/m3, the other 39 cities are exceeded, figure 1.

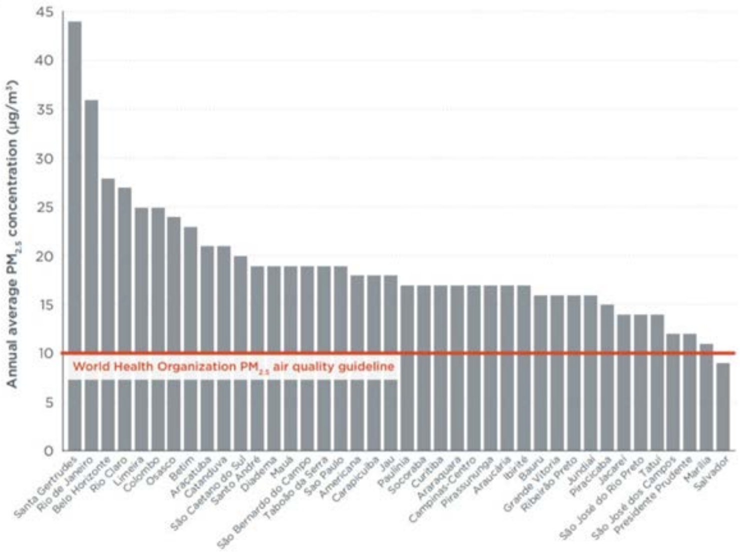


Figure 1: Annual Mean $PM_{2.5}$ concentrations in Brazilian cities (WHO, 2014).

Regarding particulate matter produced by flex fuel vehicles, Salvo et al. (2017) presented a recent paper with an investigation about the effect of ethanol – gasohol fuels usage shifts in ultrafine particles generation (from 6 nm to 100 nm diameter particles). It was found that the shift from gasohol to ethanol reduced the amount of ultrafine concentration in on third and the opposite occurred when flex fuel vehicles shifted from ethanol to gasohol.

Based on all of these facts, the control of PM pollution is extremely important for human well-being. The effect of ethanol in flex fuel vehicles, as well as the injection technology, PFI or DI, are relevant factors in this topic. This paper aims to contribute in this discussion by generating data with current production vehicles.

2. THEORETICAL FOUNDATION

2.1 Definition & Sources

2.1.1 DEFINITIONS OF PARTICULATE MATTER & PARTICLE NUMBER

It is important to disambiguate the different meanings that the term particulate matter has been subjected to. Sometimes, the different terms Particulate Matter, Particulates, Particles, PM, PN, Smoke and Soot can be treated interchangeably, on emissions discussions for general purpose, without the rigor of the academic, while discussions of emissions measurements use a distinction between Smoke, PM and PN.

Regarding to this, according to EPA, Particulate Matter can be also called particle pollution, which is defined by a mixture of solid particles and liquid droplets found in the air. Some of them are big enough to be seen with the naked eye, such as dust, soot, or smoke, while others are so tiny that they can only be seen on an electronic microscope.

A good explanation is given by EEA when they outline Particulate Matter (PM) as "a collective name for fine solid or liquid particles added to the atmosphere by processes at the earth's surface. Particulate matter includes dust, smoke, soot, pollen and soil particles". Related to Smoke, the same institution defines it as "an aerosol, consisting of visible particles and gases, produced by the incomplete burning of carbon-based materials, such as wood and fossil fuels". As for Soot, they state it as an "impure black carbon with oily compounds obtained from the incomplete combustion of resinous materials, oils, wood, or coal". At last, the EEA definition for Particle Number (PN) is "a variety of measurements characterizing the number of particles in an aerosol sample".

2.1.2 PARTICULATE MATTER SOURCES

For EPA, the particulate matter sources can come from either natural or anthropogenic source. Natural source is generated by the nature itself, such as salt spray, wildfires, sand, dust, volcanoes, etc. It is considered an anthropogenic source anything that is caused by humans or their activities, highlighting the products of combustion, agriculture, among others. Figure 2 demonstrates that anthropogenic sources, in 2011, were responsible for 44% of the $PM_{2.5}$ emissions by mass, while natural sources were responsible for 56%.

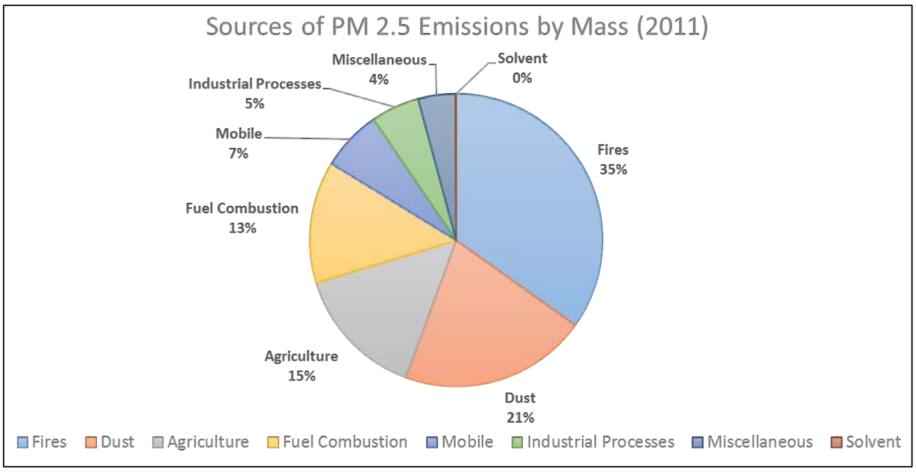


Figure 2: Contribution of the natural and anthropogenic sources on PM 2.5 emissions (EPA).

Particulate matter is divided in two distinct categories, primary and secondary. Primary particles are directly emitted and secondary particles form as a result of atmospheric reactions involving gaseous emissions. Both are regulated, though secondary particles are regulated indirectly.

2.2 Particulate Matter in Engine Exhaust and Importance

2.2.1 COMPOSITION

Once diesel fuel is one of the main responsible for the particulate matter emissions, its composition is highlighted. According to (Johnson et al., 1994), particle matter diesel emissions is divided in three phases, gas, solid and liquid/vapor phases. The gas phase emission includes NOx, CO, and sulfur dioxide (SO_2). Figure 3 demonstrates the components of the solid and liquid/vapor phases. It shows that solid phase emissions are initially constituted of small (10-80 nm) solid carbon cores – SOL – and agglomerates (50-1,000 nm). The liquid/vapor phase is composed of the organic, named SOF – Soluble organic fraction – and hydrocarbon component and sulfate (SO_4), which can be removed by water. Part of the hydrocarbons are absorbed onto the SOL, and part of them remains as a vapor.

Generally speaking, the whole composition involves some hydrocarbons absorbed in an agglomerate of SOL. Those hydrocarbons can be removed by the SOF, which is an organic solvent of the hydrocarbons.

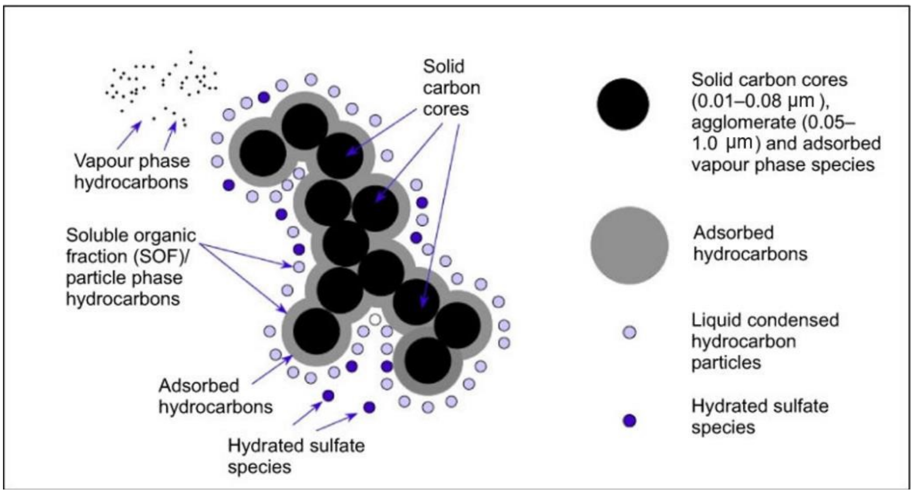


Figure 3 - Schematic of Diesel Particles and Vapor Phase Compounds (Johnson et al., 1994).

2.2.2 CHARACTERIZATION

It is noticeable that the European Regulatory Agency evolved when went from mass-based air pollution regulation for particulate matter to particle number, that is more accurate about the physical properties of the particulate matter regarding to the effects on human health. Surface area can also be measured as a parameter to be regulated.

There is an important correlation between mass, particle number and surface area of the particulate matter that is explained by (Kittelson et al., 2006), which includes the size range defined for the atmospheric particles as PM_{10} (diameter < 10 μm), fine particles $PM_{2.5}$ (diameter

< 2.5 µm), ultrafine particles $PM_{0.1}$ (diameter < 0.10 µm or < 100 nm), and nanoparticles (diameter < 0.05 µm or < 50 nm). A diesel fuel distribution is exposed in figure 4, and according to the author, distribution from a spark ignition engine would be similar but with relatively less material in the accumulation mode region.

The practical meaning of this demonstration is that the total of mass does not reflect properly the number of small particles, which are the ones harmful to human health.

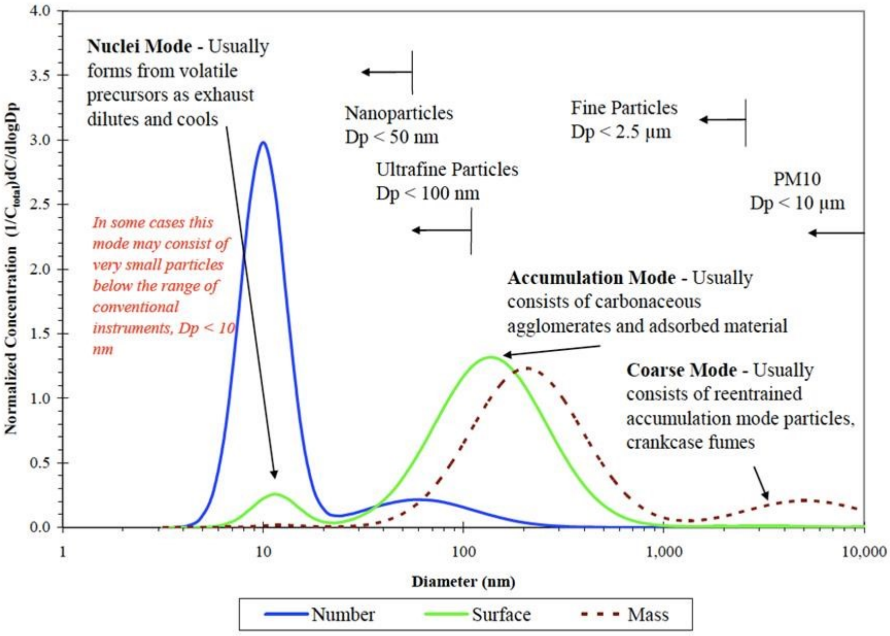


Figure 4: Typical PM Size Profile of the Emissions from a Diesel Engine Indicating the Nuclei and Accumulation Mode Size Regions (Kittelson et al., 2006).

Complementing the explanation related to the size of the particles, EPA provides a helpful description about the size of the particles when compared to a human hair. It shows that the average human hair is about 70 µm in diameter – making it 30 times larger than the largest fine particle. EPA calls the particles sizes as: PM_{10}: inhalable particles, with diameters that are generally 10 µm and smaller; $PM_{2.5}$: fine inhalable particles, with diameters that are generally 2.5 µm and smaller; $PM_{0.1}$, with diameters that are generally 0.1 µm and smaller, as seen on figure 5.

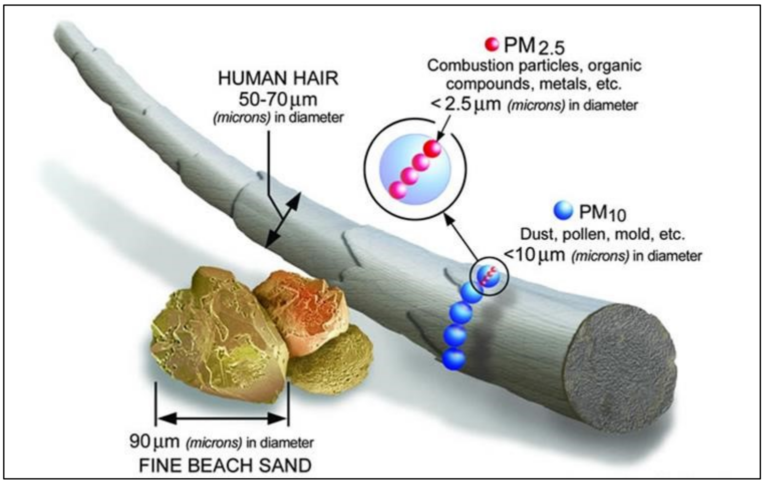


Figure 5: Size comparison between human hair and particle matter (EPA).

2.2.3 IMPACT ON HEALTH AND CLIMATE CHANGE

Several studies about health of the past decades have pointed out strong evidences that elevated levels of particulate matter air pollution are associated with increased cardiovascular and respiratory diseases (Kayser, 2018). In this matter, small particles from engines are particularly worrisome.

(Tollefson, 2018) stated that soot is a major contributor to climate change. Similarly, defined that soot, also known as black carbon, *"is the second most important human emission in terms of its climate forcing in the present-day atmosphere; only carbon dioxide is estimated to have a greater forcing"* (Bond et al., 2013). (Bullis, 2013) expound is his article "Cleaning Up Diesel Trucks and Cooking Stoves Could Reduce Climate Change", that selective reductions of particulate pollution might help the climate change issue.

2.3 Regulations and Test Cycles

In the U.S. and Europe, PM_{10} and $PM_{2.5}$ are currently regulated.

2.3.1 US EMISSIONS STANDARDS

In the United States Emissions standards have two bodies regulating emissions – the EPA and CARB. Both regulate particulate matter using a mass limit. EPA established Tier III 3mg/mi on FTP driving cycle in 2017, while CARB took LEV III 3mg/mi in 2017, 1mg/mi (begin phase-in 2025), also on FTP driving cycle. Figure 6.

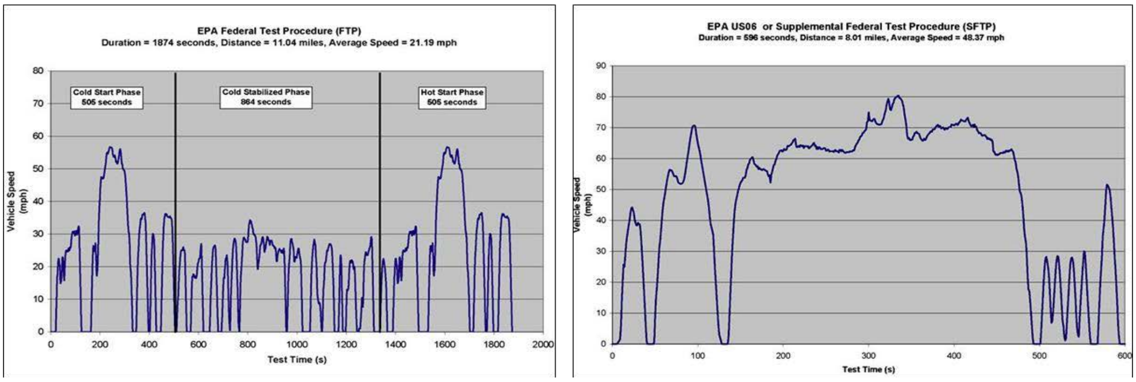


Figure 6: Speed profile of FTP 75 and US 06 driving cycles.

2.3.2 EUROPEAN EMISSION STANDARDS

The European Union implemented a Particle Number limit in addition to a mass standard. Started with Euro5b, Diesel vehicles were limited to 6x10^{11} particles/km. For Direct Injection Gasoline engines, Euro 6, in 2014, initially met a limit of 6x10^{12} particles/km, and from 2017, with Euro 6c met full standard of 6x10^{11} particles/km. Particle number standard must be met in addition to the 5mg/km particle mass standard. NEDC driving cycle has been replaced in 2017 by WLTP and the RDE on road cycle was included as a supplemental test to verify real-world compliance with emissions limits.

Particulate measurement protocol – PMP – was established to very carefully define the sampling and measurement conditions in an attempt to obtain repeatable measurements. In order to achieve repeatable measurements, only solid particles are counted, volatile particles are removed by using an evaporation tube or thermodenuder and, to further ensure volatile particles are not counted, only particles between 23 nm and 1,000 nm are counted.

2.4 Formation in Engines

Basically, the formation of the soot happens during the combustion of hydrocarbons. In the presence of oxygen (stoich or lean) and sufficient temperature, however, most of it will be oxidized. In rich flames, there is insufficient oxygen to oxidize all of the soot – that which remains is emitted.

(Smith, 1981) explains that the mechanism of the formation of the soot particles in diesel engines follows a sequence which starts with pyrolysis, that is the decomposition of the molecules, brought about by high combustion temperatures. As a result, the platelets are formed and then become microscopic crystals, also known as crystallite. All the crystallites together transform in turbostratic particles. After that, there is the coagulation phase and the surface growth. The grown particles, later on, form an aggregation. As mentioned in figure 3, there is the absorption and condensation of hydrocarbons. The increasing on the size of the particles along the mechanism of their formation is also shown on figure 7. They start at 0.35 nm as platelets and end from 0.1 to 10 μm at the aggregation of the grown surface particles.

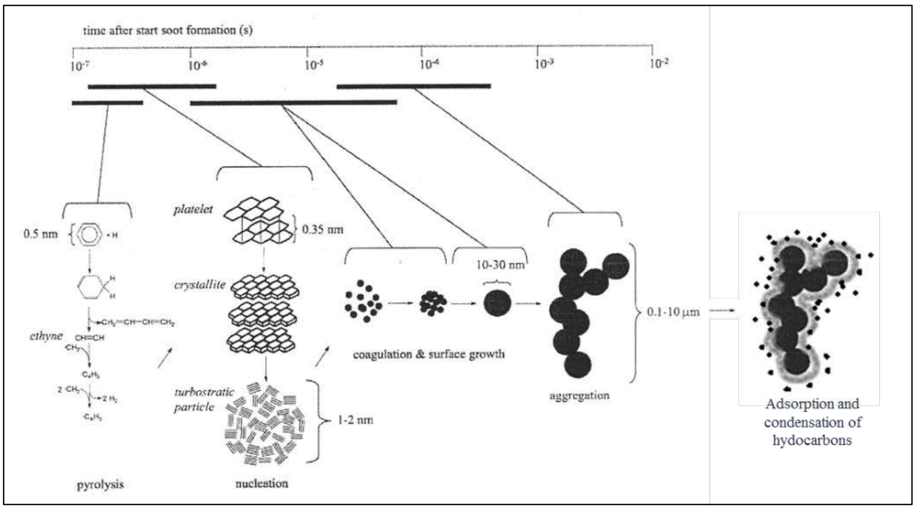


Figure 7: Schematic mechanism of the formation of soot particles (Smith, 1981).

Generally speaking, in internal combustion engines, soot forms as a result of reactions in localized fuel-rich regions. While the mechanisms differ, this is true for both Diesel and Gasoline engines. In Diesel engines soot forms on the fuel-side of the diffusion flame.

In Gasoline engines, the fuel-rich regions may be split into two categories: liquid fuel in-cylinder, in which either aerosol droplets or surface films (or pools) of fuel result from poor fuel spray characteristics; and pockets of fuel-rich mixture, that is a result of poor mixing.

(Ketterer, 2016) describes that in DI engines the sources of PM formation are directly related to the combustion chamber surfaces. Its influence is impacted by the following characteristics: (1) piston crown (poorly chosen injection timing); (2) bore liner (excessive penetration may be due to poorly chosen injection timing, spray pattern, insufficient or mismatched charge motion); (3) combustion chamber roof (direct – poor spray pattern, indirect – may splash off of the piston); (4) intake valves (Incorrect spray pattern); (5) liquid droplets in the chamber volume (poor atomization from poorly designed injector or excessive deposits on the injector); (6) residual fuel on the injector tip (may be leaking, have an excessive sac volume, or fuel may be adsorbed by deposits); (7) fuel (liquid or vapor) may collect in the top land crevice; and (8) vapor-phase fuel-rich pockets (poor mixing – may be due to poor injection or charge motion characteristics). Figure 8.

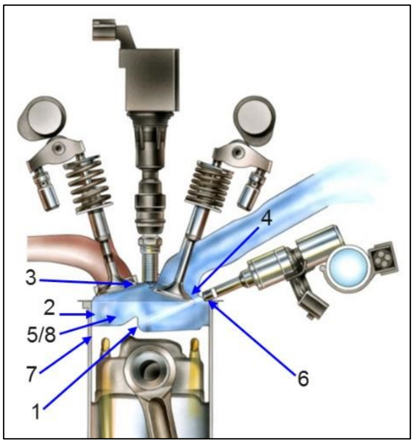


Figure 8: Impact on combustion chamber surfaces as a source of PM formation in DI engines (Ketterer, 2016).

2.5 Factor Influencing Soot Formation

As already described the soot formation is influenced by several parameters such as temperature, air-fuel ratio, chamber pressure, fuel composition, residence time and addition of additives.

Experimental studies from (Boing et al., 1990) and (Pires da Cruz 2010) have shown that soot formation increases with combustion temperature in the region below 1500 K, since the pyrolysis and crystallite rate production depends mainly on temperature, which contributes to aggregation growth and finally to form the structure of the adsorption and condensation of hydrocarbons. Differently, in the high temperature region around 1500 to 1700 K, where the oxidation process is dominant the soot formation is greatly reduced, figure 9. Many experimental investigations confirm that temperature is the dominant influence parameter on soot formation even at elevated pressure levels.

Other parameter that strongly impacts the soot formation is the air-fuel ratio. In general, the soot mass and particle diameter increase with the equivalence ratio (EQR) as shown in figure 9 with the combined effect of temperature and air-fuel ratio (as the C/O ratio). It can be observed a critical C/O ratio below which no soot is formed.

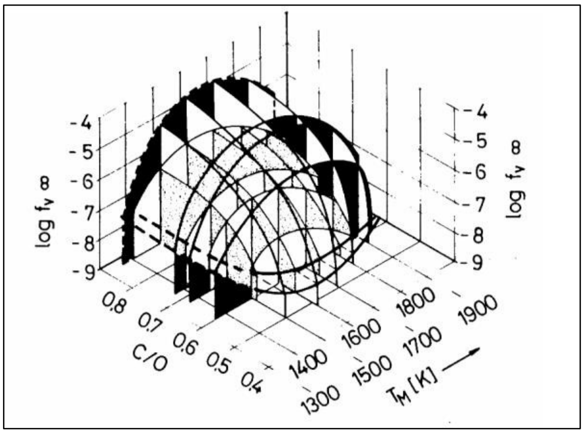


Figure 9: Soot volume fraction as function of the molar ratio C/O and the temperature behind a laminar premixed ethylene/air flame at a pressure of 10 bar (Boing et al., 1990).

The type of fuel and their composition is another important parameter. As the aggregation process results from the fuel thermal composition, its chemical structure strongly impacts the kind and quantity of products. The soot formation increases with the hydrocarbon chain size. Polyaromatics and aromatics chains produce more aggregation than alkenes (Boing, 1990). In this way, E100 fuel has chemically less tendency to soot formation than E22 fuel since its chemical structure is small (C_2H_6O) and without aromatic molecules (E22 fuel contains 15 to 35% in volume of aromatic chain).

As result of this efficiency on soot formation, some recent studies have shown the environmental and social benefits of E100 fuel. (Salvo et al., 2017) reported that the ultrafine particle (7-100 nm diameter) fall by one-third during the morning commute when higher gasoline price induces 2 million drivers in the real-world megacity of São Paulo to refuel their vehicle with E100 fuel instead of E22 fuel. The opposite trend was measured for fueling shifts from E100 to E22, figure 10.

In addition, (Pires da Cruz, 2010) relates that soot formation can also be changed by additives. He also explains that when inert additives like H_2O, CO_2 or SO_2 are added to the fuel (from dilution via internal or external gas recirculation) the profiles of species concentration and temperature are changed, decreasing the shooting tendency and overriding the temperature effect. His studies showed that the addition of water is an efficient way to reduce the soot formation.

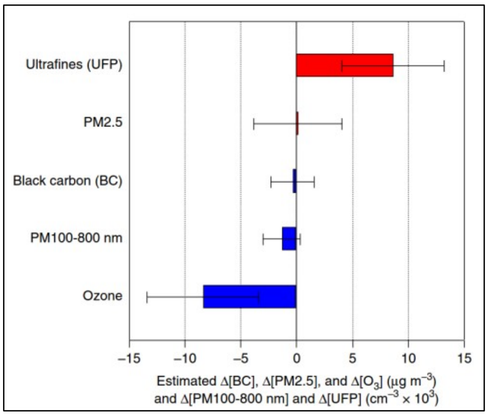


Figure 10: Estimated changes in pollutant concentrations in the São Paulo metropolitan area as the gasoline share in the flex-fuel fleet rises from 30 to 80% points (Salvo et al., 2017).

2.6 Mitigation

One way to mitigate particulate matter is by reducing engine-out emissions. Design and calibration efforts can be aimed at reducing the amount of particulate matter entering the exhaust stream. Related to exhaust aftertreatment, the use of devices in the exhaust stream is necessary to capture or oxidize the particles after they have entered the exhaust stream. Unfortunately, it is not totally effective, because it is tightly dependable on engine out reductions and generally it has larger impacts on vehicle performance and/or fuel consumption, and requires the use of particulate filters. (Whitaker, 2011) provides an example of the efforts that come from improvements obtained on engine hardware combined with improvements made on engine calibration, in order to reduce the PN emissions on a Gasoline DI engine at NEDC driving cycle, is shown on figure 11. These improvements together achieved 78% reduction on PN emissions at NEDC driving cycle. The results of this study demonstrated that the majority of the particulates are created during the cold start and at transient accelerations during cold engine operation. The contribution of each work package used for the huge reduction of particulate emissions are as follows: cold start 10%; catalyst heating 10%; transients during cold engine operation 60%; steady state and transient operation of a hot engine 20%. Details of the PN formation mechanisms, calibration actions and influencing hardware related to each of these work packages are shown on figure 12.

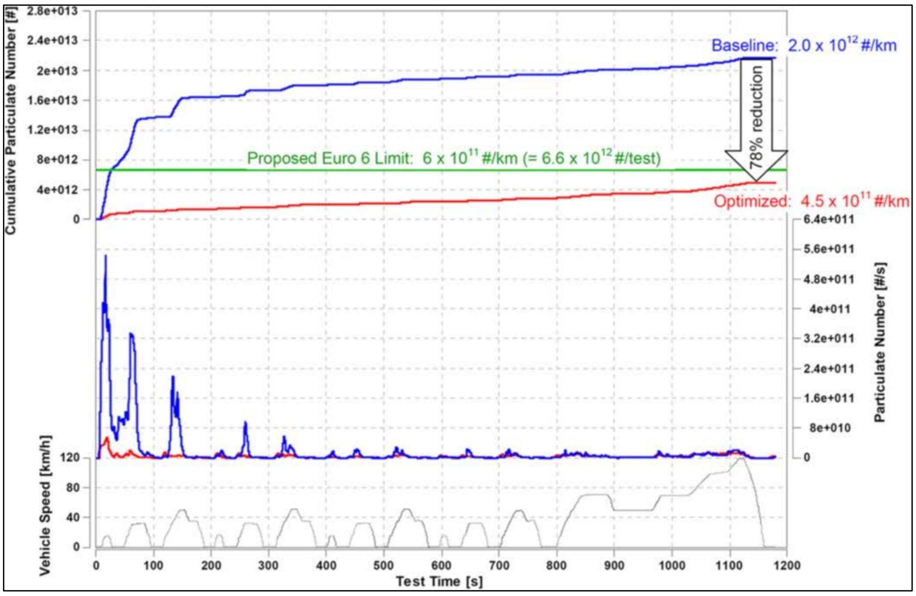
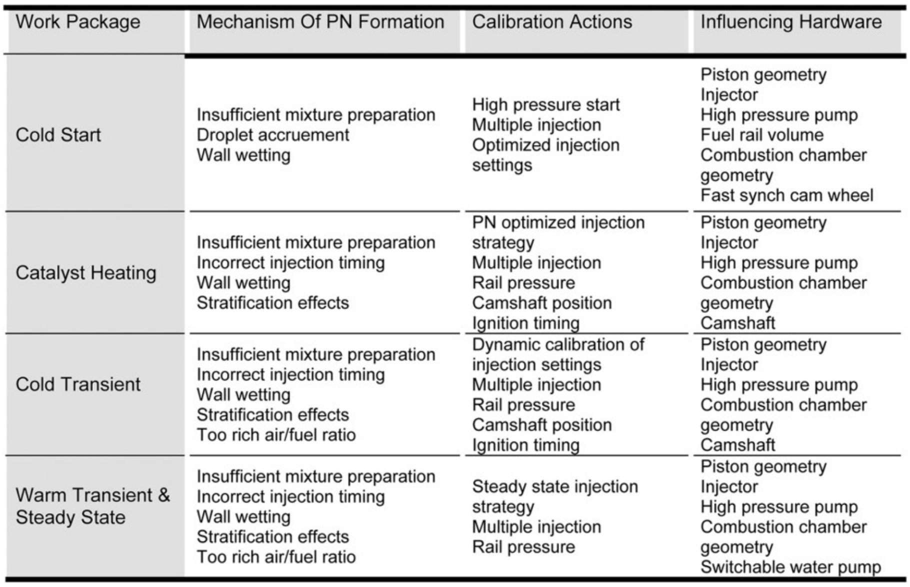


Figure 11: Particulate number results of DI engine at NEDC driving cycle. Euro-5 baseline versus engine optimized for Euro-6 PN emissions (Whitaker, 2011).



Work Package	Mechanism Of PN Formation	Calibration Actions	Influencing Hardware
Cold Start	Insufficient mixture preparation Droplet accruement Wall wetting	High pressure start Multiple injection Optimized injection settings	Piston geometry Injector High pressure pump Fuel rail volume Combustion chamber geometry Fast synch cam wheel
Catalyst Heating	Insufficient mixture preparation Incorrect injection timing Wall wetting Stratification effects	PN optimized injection strategy Multiple injection Rail pressure Camshaft position Ignition timing	Piston geometry Injector High pressure pump Combustion chamber geometry Camshaft
Cold Transient	Insufficient mixture preparation Incorrect injection timing Wall wetting Stratification effects Too rich air/fuel ratio	Dynamic calibration of injection settings Multiple injection Rail pressure Camshaft position Ignition timing	Piston geometry Injector High pressure pump Combustion chamber geometry Camshaft
Warm Transient & Steady State	Insufficient mixture preparation Incorrect injection timing Wall wetting Stratification effects Too rich air/fuel ratio	Steady state injection strategy Multiple injection Rail pressure	Piston geometry Injector High pressure pump Combustion chamber geometry Switchable water pump

Figure 12: Influence of engine calibration and hardware on the PN-result (Whitaker, 2011).

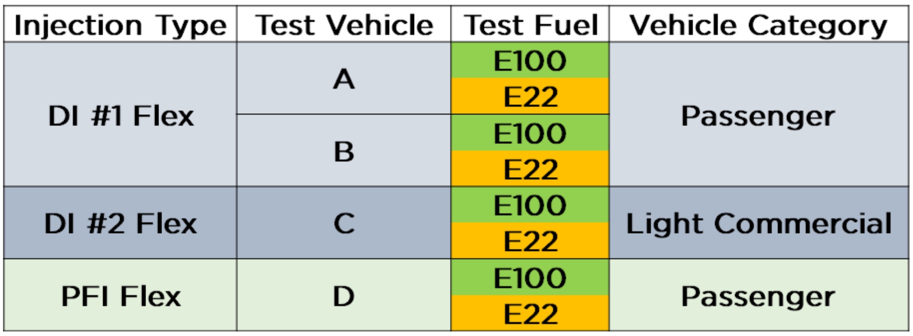
3. METHODOLOGY

Once exposed the mechanisms and characteristics of the particulate matter and its importance and relation with the human health and climate change, developed countries adopted regulations for a very careful PN control. Considering PN is generated in all engine technologies, and with the use of all fuels, it is certain that discussions about PN emissions for passengers' vehicles in Brazil will eventually consider Flex-Fuel engines of all injection technologies that run with E100 and E22, and any blend that comes from that. As an attempt to provide reference data to help the initial discussions, a group of tests were conducted on four normal production FlexFuel vehicles, fueled with E100 and E22, focusing on PN emissions on Brazilian standard driving cycle NBR6601 (EPA75). Those vehicles belong to two distinct groups of Flex-Fuel engine technologies, DI and PFI.

All measurements were performed on chassis dynamometer (48-inch roller diameter) at GM Mercosul Emissions Laboratory configured with Particle Number Counter (PNC) model AVL 489 which works with the particle's diameter between 23 nm to 2.5 µm, according to UNECE Regulation 83 (Light Duty Vehicles), figure 13.

Figure 13: PNC installed in the CVS dilution tunnel.

Two sets of DI engines and one PFI engine were used in this study, according to figure 14.



Injection Type	Test Vehicle	Test Fuel	Vehicle Category
DI #1 Flex	A	E100	Passenger
		E22	
	B	E100	
		E22	
DI #2 Flex	C	E100	Light Commercial
		E22	
PFI Flex	D	E100	Passenger
		E22	

Figure 14: Distribution of tests comparing injection technologies and fuels.

A total of 26 tests were run to generate all test batches and allow the comparison between different injection technologies and fuel blends.

4. RESULTS

For the measurements with DI engines, two normal production passenger vehicles of the same model were tested, both equipped with the same engine type (vehicle A run 3 test batches and vehicle B run 6 test batches with each fuel), and also one light commercial vehicle with a different DI engine (1 test batch with each fuel), figure 15. Another passenger vehicle assembled with a PFI engine was tested (1 test batch with each fuel), figure 16.

The results of this research showed that PN emissions with E100 fuel is much lower than PN emissions with E22 fuel in all tests. E100 PN emissions with both DI and PFI engines are lower, as seen in figures 15 and 16.

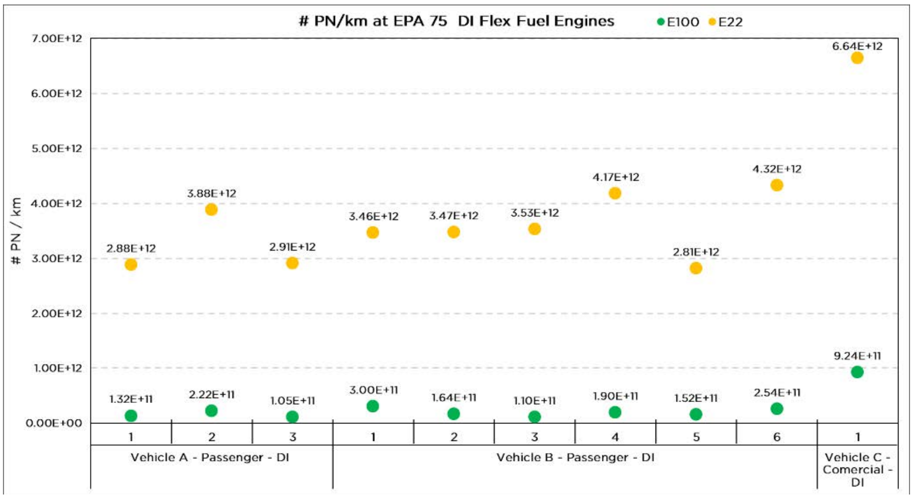


Figure 15: PN emissions with DI Flex-Fuel engines at EPA 75 driving cycle.

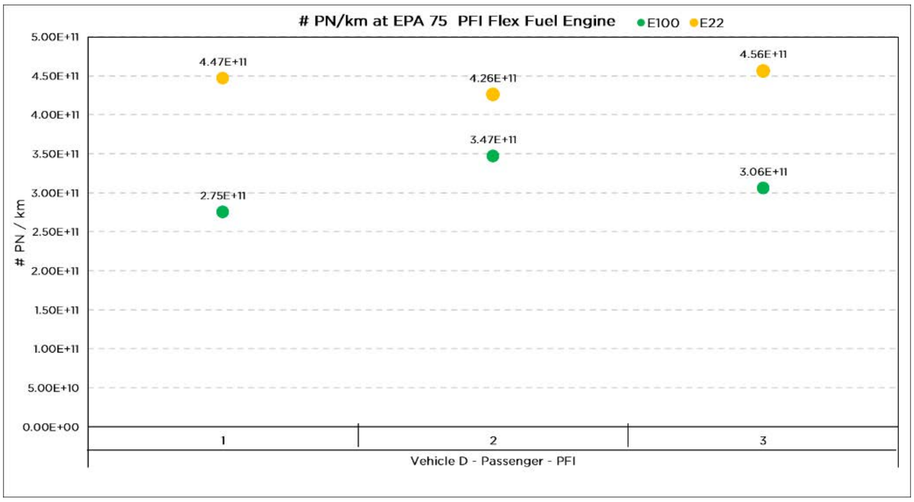


Figure 16: PN emissions with PFI Flex-Fuel engine at EPA 75 driving cycle.

The majority of the PN are generated during cold engine operation of the emissions test. Figure 17 shows the PN picks with both E100 and E22 fuels after cold start and during cold drivability transient accelerations.

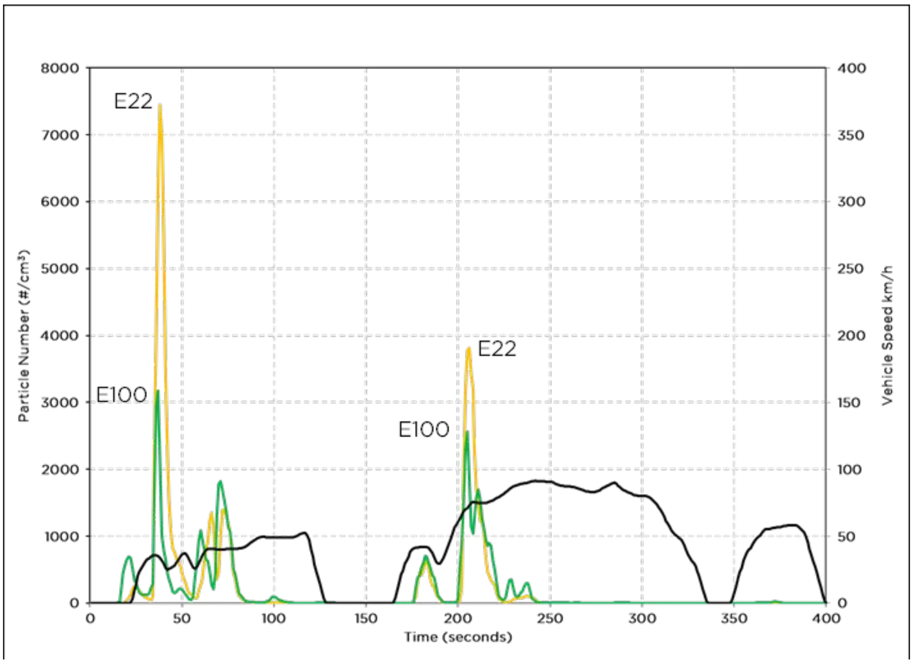
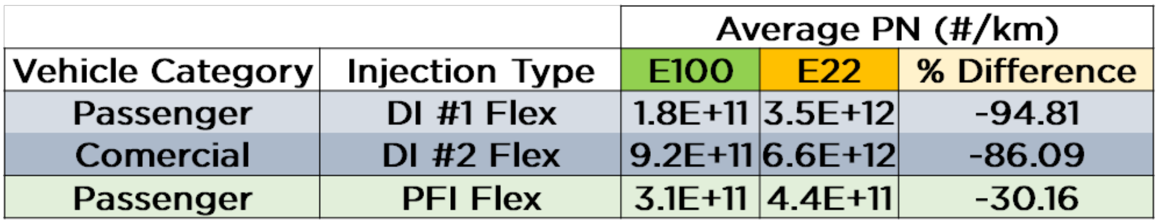


Figure 17: PN emissions on PFI Flex-Fuel engine at EPA 75 driving cycle.

Figure 18 shows the average results with each Flex-Fuel engine technology, DI and PFI show that E100 fuel produces significantly less PN emissions. On both DI engines, E100 combustion produces about 90% less PN than E22. On PFI engine, PN emissions is much lower than seen on DI engines and E100 performance is even better, generating 30% less than E22.



		Average PN (#/km)		
Vehicle Category	Injection Type	E100	E22	% Difference
Passenger	DI #1 Flex	1.8E+11	3.5E+12	-94.81
Comercial	DI #2 Flex	9.2E+11	6.6E+12	-86.09
Passenger	PFI Flex	3.1E+11	4.4E+11	-30.16

Figure 18: Final average results with E100 and E22 fuels.

5. CONCLUSIONS

Tests confirmed that DI engines produce higher amount of particle numbers than PFI engines. However, E100 particle number in both DI engines is one order of magnitude lower than E22 emissions level and gets closer to the amount of PNs generated by the PFI engine.

Based on the results E100 is a strategic fuel for reducing and controlling PM pollution. Its application is a key factor for obtaining environmental, social and economic sustainability.

6. ACKNOWLEDGMENT

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8. ABBREVIATIONS

CARB	California Air Resources Board
CO	Carbon Monoxide
CVS	Constant Volume Sampler
DI	Direct Injection
E100	Hydrated Ethanol Fuel
E22	Gasoline with 22% anhydrous ethanol
EEA	European Environment Agency
EPA	US Environmental Protection Agency
EURO 5b	European Emission Standard stage 5b
EURO 6	European Emission Standard stage 6
EURO 6c	European Emission Standard stage 6c
FTP	US Federal Test Procedure
GDI	Gasoline Direct Injection
L6	Current emissions limits phase of PROCONVE
LEV III	California light-duty vehicle emission standards
NBR 6601	Brazilian city driving test standard
NEDC	New European Driving Cycle
NOx	Nitrogen Oxide
PFI	Port Fuel Injection
PM	Particulate Matter
$PM_{0.1}$	Particulate Matter diameter < 0.10 µm
$PM_{2.5}$	Particulate Matter diameter < 2.5 µm
PM_{10}	Particulate Matter diameter < 10 µm
PMP	Particulate Measurement Protocol
PN	Particle Number
PROCONVE	Brazilian Program for Control of Air Pollution by Automotive Vehicles
RDE	Real Driving Emissions
SO2	Sulfur Dioxide
SO4	Sulfate
SOF	Soluble organic fraction
SOL	Solid carbon cores
STOICH	Stoichiometric mixture
Tier III	US federal exhaust emission standards for light-duty vehicles
US	United States of America
WHO	World Health Organization

WLTP	Worldwide harmonized Light Vehicles Test Procedure