

# Secondary Organic Aerosol Formation from Untreated Exhaust of Gasoline Four-Stroke Motorcycles

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This document is the accepted manuscript version of the following article:  
Esmailirad, S., Setyan, A., Wang, J., & Hosseini, V. (2021). Secondary organic aerosol formation from untreated exhaust of gasoline four-stroke motorcycles. *Urban Climate*, 36, 100778 (13 pp.). <https://doi.org/10.1016/j.uclim.2021.100778>

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## ***ABSTRACT***

This study investigates the secondary organic aerosol (SOA) formation potential of carburetor motorcycles exhaust. This type of two-wheeler is a popular means of transport in many Asian cities. A volatility-based numerical model was employed to predict SOA formation from a fleet of motorcycles in Tehran, capital of Iran. The fleet was a combination of four-stroke, gasoline-powered motorcycles with different engine displacement volumes. Total hydrocarbon (THC) emission factors of all motorcycles were previously measured in a chassis dynamometer laboratory according to cold start Euro-3 emissions certification test procedures. Due to incomplete combustion and lack of control on exhaust emissions, unburned fuel was assumed to be a good surrogate for the exhaust of carburetor motorcycles, regarding SOA formation. 150 *cc* engine and 200 *cc* engine motorcycles had the highest SOA formation potential, under atmospheric oxidant concentration, while 125 *cc* engine motorcycles had the highest SOA emission factor (travel- and fuel-based). It was found out that SOA emission factor of 125 *cc* engine motorcycles could increase up to 20%, three to five years after production. Average SOA formation from carburetor motorcycles in the present study was 4 times higher than Euro-4 passenger cars and 20 times higher than direct emission of particles from Euro-2 motorcycles and on (according to EMEP/EEA levels for PM<sub>2.5</sub> emission factors). Carburetor motorcycles with 180 *cc* engine volume in the present study, had the lowest SOA formation potential.

Keywords: carburetor motorcycles; emission factor; SOA; unburned fuel

## 1. Introduction

Particulate matter (PM) in the atmosphere is the subject of great concern, due to its known adverse effects on the environment and human health. PM is either directly emitted into the atmosphere from polluting sources (primary) or is formed inside the atmosphere, as a result of chemical reactions of gas-phase precursors (secondary). Organic aerosol (OA) forms 20%–90% of submicron particles mass, depending on the location around the globe (Jimenez et al., 2009). Mobile sources (i.e. automobiles) are the major anthropogenic sources of OA, which contribute to both primary OA (POA) and secondary OA (SOA) (Gentner et al., 2017).

Motor vehicles emit a complex mixture of hydrocarbons, including single- and multiple-ring aromatics, normal/ cyclic/ branched alkanes and olefins, a lot of which are SOA precursors (Dhital et al., 2019; Lu et al., 2018). Previous studies have shown that internal combustion engines exhaust have a chemical structure similar to unburned fuel (Gentner et al., 2012; Miracolo et al., 2012; Presto et al., 2011). Sampling and gas chromatography analysis of fuel is more straightforward than engine exhaust. Therefore, using unburned fuel as a surrogate for exhaust can provide reasonable estimates regarding the SOA formation potential of some vehicles exhaust. Jathar et al. (2013) investigated SOA formation from eleven different unburned fuels (commercial gasoline, three types of jet fuel, and seven different diesel fuels) in a smog chamber. They observed SOA production trends largely consistent with differences in carbon number and molecular structure of the fuels. They also found that both the mass yield and composition of the SOA formed from unburned fuel was similar to those from uncontrolled emissions of engines operating on those fuels. Therefore, unburned fuel may be appropriate surrogate for uncontrolled engines exhaust, but not for emissions from engines equipped with after-treatment devices such as catalytic converters.

Early attempts to simulate SOA formation from gas-phase oxidation of organic compounds only accounted for high-flux VOCs like single-ring aromatics, alkanes and alkenes with less than or equal to 12 carbons, isoprene and terpenes (Goldstein and Galbally, 2007; Odum et al., 1997). Later studies showed that this traditional approach resulted in considerable underestimation of SOA formation in urban and remote regions (De Gouw et al., 2009; Dzepina et al., 2009; Volkamer et al., 2006). Further studies revealed that combustion sources emit a substantial amount of lower

volatility organics, known as semi- and intermediate VOCs (S/IVOCs), which are difficult to speciate, hence not included in models (Grieshop et al., 2009; Robinson et al., 2007).

A large part of S/IVOCs mass cannot be quantified using one-dimensional gas chromatographic techniques and they are classified as unresolved complex mixtures (UCM). Due to uncertainty regarding the molecular structure of these compounds, it is not possible to study SOA formation from their oxidation in similar manners to traditional precursors. Alternatively, non-traditional models have been developed which employ volatility-based oxidation mechanisms (Esmaeilirad and Hosseini, 2018; Jathar et al., 2012; Pye and Seinfeld, 2010). In all of these non-traditional models, volatility of the precursor species is decrease by one or multiple orders of magnitude after each oxidation reaction. SOA mass yields data either come from a surrogate species assumption or are directly derived from smog chamber experimental results.

Motorcycles are a popular means of transport in many Asian countries. Haworth (2012) reported that 77% of the total number of worldwide motorcycles were used in Asia. Although motorcycles are considered economically efficient and easily mobile, they commonly emit more pollutants (per power unit) than passenger cars. The main reason is the use of cheaper engines with limited technological advances, which lack effective emission control or after-treatment technologies. Unburned hydrocarbon (HC) is one of the important regulated pollutants emitted from motorcycles. HC emission factor from motorcycles can be 100-200 times higher than passenger cars (Vasic and Weilenmann, 2006; Zhou et al., 2014).

Motorcycles constitute 18% of the vehicles fleet in Tehran, consisting almost entirely of the air-cooled gasoline four-stroke engine type with carburetor fuel delivery system. Currently, all the on-road motorcycles in Tehran were produced under the so-called Euro-3 certification and, according to the DoE<sup>1</sup> regulations, Euro-4 certification for motorcycles came into force only in late 2019. Emission inventory studies in Tehran have shown that 31% of total emitted VOCs during morning rush hours in Tehran is related to motorcycles (Shahbazi et al., 2016a), which is equivalent to more than 27000 tons per year (Shahbazi et al., 2016b). Such extent of organic compounds emission is a significant source of SOA formation in the atmosphere.

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Carburetor motorcycles emit a considerable amount of unburned fuel, due mainly to two reasons: 1) Rich fuel-air mixture leads to low combustion efficiency and incomplete combustion of hydrocarbons. 2) Air circulation around outer walls of the combustion chamber leads to undesirable cooling of chamber wall and quenching the flame near it (Heywood, 1989). For the majority of carburetor motorcycles in Tehran, exhaust emissions are completely uncontrolled (Hassani and Hosseini, 2016). Therefore, unburned fuel can be a reasonable surrogate for the exhaust of carburetor motorcycles, in terms of SOA formation potential (Jathar et al., 2013). For this purpose, chemical composition of the gasoline, including molecular weight, carbon number, carbon intensity and volatility distribution, must be determined.

In the present study, we used a volatility-dependent model to estimate SOA formation potential from carburetor motorcycles fleet in Tehran, using gasoline fuel as a surrogate for engine exhaust.

## 2. Materials and methods

### 2.1. Fuel speciation

One sample of regular gasoline, commonly consumed by motor vehicles in Tehran, was analyzed to determine its molecular structure, using gas chromatography/mass spectrometry. The instrument used for the analysis was constituted of an Agilent 6890 GC and an Agilent 5973 mass selective detector (MSD). The GC was equipped with an Agilent J&W DB-624 column (length: 30 m, inner diameter: 0.25 mm, film thickness: 1.4  $\mu$ m). The gasoline was characterized using the EPA method 8260D. Figure 1 plots the fuel speciation data in the volatility basis set (VBS). The VBS is a modeling framework which classifies organic compounds into logarithmically spaced bins of effective saturation concentration ( $C^*$ ) (Donahue et al., 2006).  $C^*$  is proportional to the saturation vapor pressure of each species and is used to describe the gas-particle partitioning of an organic mixture at 298 K.

In order to determine the  $C^*$  for each species in the fuel, mathematical formulations, developed by Jathar et al. (2014), were used. Those formulations were based on NIST vapor pressure data for different organics and they were as following:

$$C^* = \exp\left(\frac{(24.5 - C^\#)}{0.899}\right) \quad \text{for alkanes and alkenes} \quad (1)$$

$$C^* = \exp\left(\frac{(22.3 - C^\#)}{0.806}\right) \quad \text{for aromatics} \quad (2)$$

where,  $C^\#$  is the carbon number of the hydrocarbon. Presented data in Figure 1 is based on the quantified mass, which was around 70% of total mass of the gasoline sample. Previous analyses on Tehran gasoline fuel have reported total aromatic + naphthalene content of approximately 40%, therefore, the portion of the fuel mass which was not quantified is mostly of aliphatic structure (paraffin and olefin). The mass fraction distribution, for the quantified fuel mass, is presented in Table 1. Total quantified density ( $521 \text{ kg m}^{-3}$ ) was divided by the actual density ( $744 \text{ kg m}^{-3}$  from previous measurements) to scale the volatility distribution and reduce the error. This is the reason the sum of all mass fractions in Table 1 is not equal to unity. Since the volatility distribution of the not-quantified mass was unknown, its contribution to SOA formation was ignored. Thus, the predictions of the present study are the lower limit for the actual SOA formation potential of the carburetor motorcycles exhaust in Tehran.

## 2.2. Motorcycles tests

A study was performed on the tailpipe emissions of sixty carburetor motorcycles, rented from private owners in Tehran. Table 2 summarizes the studied fleet, which was so chosen to resemble the real fleet of in-use motorcycles in Tehran, with more than 70% consisting of 125 cc displacement volume engines (Hassani and Hosseini, 2016). All of the motorcycles were tested on a standard chassis dynamometer under the proper ECE driving cycle<sup>2</sup> for cold start Euro-3 emissions tests. Concentrations of CO<sub>2</sub>, CO, total hydrocarbons (THC) and NO<sub>x</sub> were measured in each test. Details about the instrumentation and test setup can be found in Hassani and Hosseini (2016).

In the present study, we assumed that the organic molecular composition of motorcycle exhaust can be approximated by unburned fuel, in terms of SOA formation. Thus, we need to know the THC concentration at the tailpipe. Table 3 summarizes the data from chassis dynamometer measurements required for calculating THC concentration. Engines with larger than 125 cc volume were tested during two phases, phase one under ECE and phase two under EUDC<sup>3</sup> driving cycle.

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<sup>2</sup> - ECE (Economic Commission for Europe) is an Urban Driving Cycle, also known as UDC.

<sup>3</sup> - Extra Urban Driving Cycle

The two phases of the tests are shown in Figure 2. EUDC cycle has been designed to represent more aggressive, high speed driving modes. The maximum speed of the EUDC cycle is 120 *km/h*, while low-powered motorcycles barely reach 60 *km/h*. Values reported in Table 3 are arithmetic mean  $\pm$  one standard deviation in each group of engine volume. Higher engine loads during EUDC cycle result in higher exhaust temperature and eventually higher emission rate of NO<sub>x</sub> and lower emission rate of THC.

In order to calculate THC concentration inside emission bags, two separate methods were used. In the first method, total THC mass is calculated from THC emission factor and distance travelled in the test. THC mass divided by the bag volume results in the THC concentration:

$$C_{\text{Bag}} = \frac{EF_{\text{THC}} \times \text{distance} \times MW_{\text{fuel}} \times 10^6}{V \times MW_{C_1} \times \text{Fuel } C^\#}, \quad (3)$$

where,  $C_{\text{bag}}$  is THC concentration inside the emission bags ( $\mu\text{g } m^{-3}$ ),  $EF_{\text{THC}}$  is THC emission factor ( $\text{g } km^{-1}$ ),  $MW_{\text{fuel}}$  is molecular weight of fuel ( $104.3 \text{ g } mole^{-1}$ ),  $MW_{C_1}$  is molecular weight of methane ( $16.04 \text{ g } mole^{-1}$ ),  $V$  is the bag volume ( $m^3$ ) and  $C^\#$  is carbon number of the fuel (8.2). Fuel properties were derived from GC analysis of Tehran gasoline sample.

In the second method, ideal gas assumption was used, as following:

$$C_{\text{Bag}} = \frac{C_{\text{THC}} / \text{Fuel } C^\# \times P_{\text{air}} \times MW_{\text{fuel}}}{8.314 \times T_{\text{air}}}, \quad (4)$$

where,  $C_{\text{THC}}$  is reported THC concentration inside emission bags ( $ppmC_1$ ),  $P_{\text{air}}$  is dilution air pressure ( $Pa$ ) and  $T_{\text{air}}$  is dilution air temperature ( $K$ ). Since we have assumed that chemical composition of the exhaust is similar to that of unburned gasoline, average carbon number of fuel was used to convert  $ppmC_1$  to  $ppm$ . Total THC concentration (sum of two phases) at tailpipe is:

$$C_{TP} = C_{\text{Bag},1} \times DF_1 + C_{\text{Bag},2} \times DF_2, \quad (5)$$

where,  $C_{TP}$  is tailpipe concentration of THC,  $C_{\text{Bag},1}$  and  $C_{\text{Bag},2}$  is THC concentration in phase 1 and phase 2 corresponding bags, respectively, and  $DF$  is dilution factor for each phase. Finally, ambient concentration of THC ( $C_{\text{amb}}$ ), is calculated, using proper assumption for ambient dilution factor ( $DF_{\text{amb}}$ ):

$$C_{\text{amb}} = \frac{C_{\text{TP}}}{\text{DF}_{\text{amb}}}. \quad (6)$$

Considering that SOA mass yields employed in the present study (section 2.3), were derived from smog chamber experimental results,  $\text{DF}_{\text{amb}}$  was so chosen that THC concentration in the ambient ( $C_{\text{amb}}$ ) was close to the smog chamber initial concentrations. Hence, a factor of 1000 was fit to that purpose.  $C_{\text{amb}}$  calculated from both methods had an average disagreement of about 2% for all cases. Table 4 reports the average ambient THC concentrations, which will be used as an input to the SOA prediction model.

Another method of reporting emission factors of hydrocarbons from vehicles, is based on the unit mass consumption of the fuel ( $g \text{ kg-fuel}^{-1}$ ). Fuel-based emission factors for the motorcycles under study are determined as:

$$\text{Fuel-based EF} = \frac{\text{EF}_{\text{THC}} \times 100}{\text{Fuel}_{\text{cons.}} \times 10^{-3} \times \rho_{\text{fuel}}}. \quad (7)$$

where,  $\text{EF}_{\text{THC}}$  is THC emission factor ( $g \text{ km}^{-1}$ ),  $\text{Fuel}_{\text{cons.}}$  is fuel consumption in each phase ( $L/100 \text{ km}$ ) and  $\rho_{\text{fuel}}$  is the density of the fuel existing in the motorcycle tank ( $kg \text{ m}^{-3}$ ).

Table 5 compares the THC emission factors of the present Iranian motorcycles fleet with those published in the literature for a number of 4-stroke two-wheelers (motorcycles, mopeds, scooters). THC emission factors of Iranian carburetor motorcycles are close to the measured values in Asia and Europe. Carburetor fuel delivery system, without any after-treatment device, generally emits more unburned hydrocarbons than fuel injection system, for the same engine volume.

May et al. (2014) have characterized tailpipe emissions of light duty gasoline vehicles (LDGVs) of varying ages, in addition to several medium- and heavy-duty diesel vehicles with varying levels of after-treatment. Figure 3 compares THC emission factors of motorcycles in the present study, with different groups of vehicles studied by May et al. (2014). It is noticed that THC emission factor from carburetor motorcycles in Tehran is 3 times higher than older gasoline vehicles, while 17 times higher than newer gasoline vehicles and non-DPF diesel vehicles. Banitalebi and Hosseini (2015) measured hot exhaust emission factors of regulated gas pollutants from Iranian-made Euro-2 certified LDGVs and reported a value of  $0.24 \text{ g km}^{-1}$  for their THC emission factor. For carburetor motorcycles in the present study, this value equals to 0.48 on average, which is two

times as high as Euro-2 gasoline vehicles. Consequently, it is anticipated that SOA formation potential from carburetor motorcycles in Tehran is much higher than LDGVs, even with an inferior pollution standard.

### 2.3. SOA model

In the present study, SOA mass yields data, presented by Jathar et al. (2014) were employed. They have used SOA production data from an ensemble of unburned fuels, measured in a smog chamber, to fit an SOA model, solely based on the volatility of the precursors. Their model could describe the observed SOA formation with relatively few parameters. Smog chamber data were from the work of Jathar et al. (2013) in which eleven different fuels (3 gasoline; 2 Fischer–Tropsch derived from coal; 2 Fischer–Tropsch derived from natural gas; 6 Jet Propellant-8 and 10 diesels), were tested in Carnegie Mellon University smog chamber. Initial concentration of gasoline vapor was between 500 and 2000  $\mu\text{g m}^{-3}$ . Experiments were conducted under low- $\text{NO}_x$ , dry condition ( $\text{RH} < 5\%$ ) and 298 K. The average OH-exposure in each experiment was  $1.7 \times 10^7 \text{ molec-h cm}^{-3}$ ; this corresponds to 17 hours of photo-oxidation at an OH concentration of  $10^6 \text{ molec cm}^{-3}$ .

In the SOA model, decay of the SOA precursors and the production of semi-volatile species is described using the following set of equations:

$$\frac{d[X_j]}{dt} = -k_{\text{OH},X_j}[\text{OH}][X_j] \quad (8)$$

$$\frac{d[M_i]_{g+p}}{dt} = \sum_j \alpha_{i,j} k_{\text{OH},X_j}[\text{OH}][X_j] \quad (9)$$

where,  $X_j$  is an SOA precursor  $j$  (each bin in the volatility distribution of Table 1),  $k_{\text{OH},X_j}$  is the reaction rate constant of precursor  $j$  with OH radical,  $M_i|_{g+p}$  is the total gas + particle organic mass in bin “ $i$ ” of the VBS and  $\alpha_{i,j}$  is the SOA mass yield.  $[\ ]$  symbols are used to represents concentration. The reaction rate of each organic species with OH radical was calculated using mathematical formulations developed by Jathar et al. (2014), in which,  $k_{\text{OH}}$  was related to  $C^*$ :

$$k_{\text{OH}} = -1.84 \times 10^{-12} \log(C^*) + 4.27 \times 10^{-11} \quad \text{for alkanes} \quad (10)$$

$$k_{\text{OH}} = -5.7 \times 10^{-12} \log(C^*) + 1.14 \times 10^{-10} \quad \text{for aromatics} \quad (11)$$

A weighted average  $k_{OH}$ , using the mole fractions obtained from GC-MS analysis of the fuel sample, was calculated for each volatility bin. Average reaction rate with OH radical for each SOA precursor is presented in Table 1. Gas particle partitioning is calculated as following:

$$\zeta_i = \left(1 + \frac{C_i^*}{C_{OA}}\right)^{-1}$$

$$C_{OA} = \sum_{i=1}^N \zeta_i \times M_i \big|_{g+p} \quad (12)$$

where  $\zeta_i$  is the fraction of mass in volatility bin “ $i$ ” in the particle-phase,  $C_i^*$  is the effective saturation concentration corresponding to bin “ $i$ ” in  $\mu g m^{-3}$ ,  $C_{OA}$  is the total particulate OA concentration in  $\mu g m^{-3}$  and  $N$  is the total number of volatility bins in the VBS.

Multi-generation oxidation is not accounted for in this model, since SOA mass yields are derived from smog chamber experiments which had similar reaction time and OH-exposure. Thus, SOA mass yields already include some multigenerational oxidation.

In this model, it is assumed that SOA formation only depends on precursors volatility (or carbon number) and it does not account for differences in molecular structure (e.g. alkane vs aromatic). Scheme of this model is similar to that of Jathar et al. (2012) for aircraft exhaust or Esmaeilirad and Hosseini (2018) for vehicle exhaust. It is assumed that each  $C^*$  bin is a precursor species that reacts with OH radical to form a distribution of semi-volatile species, 3 to 7 orders of magnitude lower in volatility than the original precursor. Each higher (or lower) precursor  $C^*$  bin is assumed to form the same product distribution but shifted by one  $C^*$  bin.

In order to determine the matrix of SOA mass yields ( $\alpha_{i,j}$ ), Jathar et al. (2014) fitted the time-dependent SOA data from all of the fuels, simultaneously and obtained the SOA yields presented in Table 6. According to the volatility distribution of Tehran gasoline sample, 8 precursor species exist in a  $C^*$  range of  $10^3$  to  $10^{10} \mu g m^{-3}$  and their corresponding oxidation products are distributed across a  $C^*$  range of  $10^{-4}$  to  $10^7 \mu g m^{-3}$ .

Condensed-phase chemistry of semi-volatile organic compounds largely falls under process known as “oligomerization” which leads to higher carbon-number products, without new OA mass addition (Miracolo et al., 2010). Therefore, the model employed in the present study, only accounts for the gas-phase oxidation of organic compounds, present in the vehicle exhaust and no particle-

phase aging processes are considered, as the mechanisms involved are highly complicated and uncertain. Apart from particle-phase aging, SVOCs can evaporate upon dilution in the ambient and undergo gas-phase oxidation. However, it has been previously shown that SVOC oxidation has very little contribution to final SOA predicted by this model (Esmailirad and Hosseini, 2018).

### 3. Results and discussion

Key input parameters for the volatility-dependent model, employed in the present study, are volatility distribution of the gasoline fuel, ambient concentration of THC emitted from motorcycles and OH reaction rate for each precursor bin ( $C^*$ ). All of these inputs were determined in section 2. OH radical concentration was taken as  $10^6 \text{ molec cm}^{-3}$  (typical ambient concentration) and oxidation time for each experiment was 17 hours, as suggested by Jathar et al. (2014). SOA prediction results are shown in Figure 4, for different engine volumes of carburetor motorcycles in Tehran. Findings of this graph are in agreement with reported values for average ambient concentrations of unburned hydrocarbons for each group of engine volume (Table 4). The highest THC and SOA concentration correspond to 150 cc engines and the lowest correspond to 125 cc engines. The higher the THC concentration, the more the SOA precursor concentration among combustion products.

Fuel-based SOA emission factor ( $g \text{ kg fuel}^{-1}$ ) is calculated as:

$$EF = \frac{\Delta m \times C_f}{MW_C} \times \left( \frac{\Delta CO_2}{MW_{CO_2}} + \frac{\Delta CO}{MW_{CO}} + \frac{\Delta THC}{MW_{THC}} \right)^{-1} \quad (13)$$

where, EF is SOA emission factor,  $\Delta m$  is SOA concentration after 17 h of photo-oxidation ( $\mu g \text{ m}^{-3}$ ),  $\Delta CO_2$  is carbon dioxide net concentration ( $\mu g \text{ m}^{-3}$ ),  $\Delta CO$  is carbon monoxide net concentration ( $\mu g \text{ m}^{-3}$ ),  $\Delta THC$  is net concentration of unburned hydrocarbons ( $\mu g \text{ m}^{-3}$ ), MW is molecular weight of carbon-containing combustion products and  $C_f$  is carbon intensity of the fuel ( $g\text{-Carbon kg-fuel}^{-1}$ ). Using equation (13), fuel density and average fuel consumption, one can also calculate the travel-based emission factor of SOA. Calculated factors are reported in Table 7, for different groups of motorcycle engine volumes. It is inferred from Table 7 that 125 cc motorcycles, which constitute 70% of in-use motorcycles fleet of Tehran, have the highest emission factor of secondary organic aerosols, as well. It was also noticed in Figure 3 that THC emission factor of

125 cc motorcycles was higher than all other engine volumes. Generally, an increase in the engine volume causes increased consumption of fuel and air. However, higher emission factors of THC and SOA for 125 cc motorcycles indicates lower combustion efficiency, meaning more unburned fuel in the combustion products for less fuel combustion. Previous measurements on the same set of motorcycles had shown that NO<sub>x</sub> emission factor in 125 cc motorcycles is much lower than other engine volumes (Hassani and Hosseini, 2016). This is another evidence for lower combustion efficiency in 125 cc motorcycles. Because NO<sub>x</sub> formation increases as the combustion temperature increases. Higher combustion efficiency in higher engine volumes can be due to better design of combustion chamber, which results in less fuel trapped in different parts of the chamber, hence remain unburned. Better adjustment and functioning of carburetor can also affect the combustion efficiency. Among different groups of carburetor motorcycles under study, 180 cc engine volumes had the best condition, regarding the emission of regulated pollutants and SOA formation potential.

Figure 5 compares SOA emission factors of motorcycles in the present study, with different groups of vehicles studied by May et al. (2014). All the EFs in this figure were calculated for 17 h of photo-oxidation in the atmosphere (OH concentration =  $10^6 \text{ molec cm}^{-3}$ ). According to Figure 5, average SOA formation from Tehran motorcycles fleet is 28 times higher than average SOA formation from gasoline and diesel vehicles studied by May et al. (2014).

Effect of aging on THC emission and SOA formation was assessed for 125 cc engine volume motorcycles, due to the larger number of tests carried out and the wider span of production dates in that group. 125 cc motorcycles were divided into three categories (all tests were performed in 2014):

- Motorcycles produced before 2010 (9 tests)
- Motorcycles produced between 2010 and 2012 (15 tests)
- Motorcycles produced between 2013 and 2014 (9 tests)

Figure 6 shows the aging assessment results. Calculation basics for THC and SOA emission factors in Figure 6 are the same as Figure 3 and Figure 5, respectively. It is understood that newer motorcycles emit less unburned hydrocarbons, which results in decreased SOA formation. Three years after production and later on, SOA emission factor from these motorcycles increased up to 17-22%. Since these motorcycles produced in different years, did not have various engine and

emission control technologies (all Euro-3 with carburetor), the slight increase in THC and SOA emission factors for older motorcycles can be related to deteriorated carburetor performance and combustion characteristics as a result of aging.

Due to the absence of primary PM measurements from in-use motorcycles in Tehran, EMEP/EEA data for exhaust emissions from road transport (passenger cars, light commercial trucks, heavy-duty vehicles including buses and motorcycles) were used for comparison purpose. EMEP/EEA exhaust emission factors are Tier 2 emission factors that were calculated on the basis of the Tier 3 methodology. Figure 7(a) presents a comparison between the SOA emission factors of the motorcycles fleet in the preset study and direct PM<sub>2.5</sub> emission factors of different classes of euro-certified vehicles. Average SOA formation from motorcycles in Tehran can be 60 times higher than direct PM emissions from a Euro-5 passenger car. This quantity is 3 times higher than PM emission from Euro-5 heavy duty diesel vehicles, 1.5 times higher than Euro-5 urban buses and 20 times higher than primary emission of particles from Euro-2 motorcycles and on. In addition, according to direct measurements of PM emissions from two-wheelers (reported in Table 5), SOA emission factor from Tehran motorcycle fleet ( $\sim 80 \text{ mg km}^{-1}$ ) can be 10 times as high as direct PM emission factor from a 125 cc carburetor motorcycle ( $\sim 8 \text{ mg km}^{-1}$ ) and over 50 times higher than fuel-injection motorcycles, equipped with catalyst ( $\sim 1.5 \text{ mg km}^{-1}$ ). Figure 7(b) compares the average SOA emission factor from Tehran motorcycles with primary PM emission factors from motorcycles in different European countries, based on a fleet composition in 2005. The average SOA EF reported in Figure 7(b) for Tehran is a weighted average based on the fleet composition in 2014. It is found that average fuel-based SOA emission factor for Tehran motorcycles is more than 3 times higher than direct PM emissions from motorcycles in European developed countries and almost close to the values corresponding to less developed countries of eastern Europe.

Many studies have proved the effect of fuel injection system and catalytic converters on reducing the emission of unburned hydrocarbons (HC) and VOCs from motorcycles (Costagliola et al., 2014; Dhital et al., 2019; Katijan et al., 2019; Tsai et al., 2017). Decreased levels of organic compounds in the exhaust will ultimately result in lower SOA formation in the atmosphere.

#### 4. Conclusion

Studies have shown that, in terms of SOA formation, unburned fuel may be appropriate surrogate for uncontrolled engines exhaust, due to the similarity between the composition and mass yield of their SOA products. Carburetor motorcycles are good example of vehicles with no control on their combustion generated pollutants. Such motorcycles are the third popular means of transport in the city of Tehran and have a considerable contribution to the severe air pollution problem of this city. Unburned hydrocarbons in the exhaust of motorcycles is a rich source of SOA precursors that can further contribute to the particulate pollution. In the present study, we applied a volatility-dependent model to THC emission data measured from a set of motorcycles, in order to predict the SOA formation potential of those vehicles exhaust. The motorcycles were so selected that represent the fleet in Tehran (a combination of 125 *cc*, 150 *cc*, 180 *cc* and 200 *cc* engine displacement volumes). THC, CO and CO<sub>2</sub> emission factors of those motorcycles were previously measured in chassis dynamometer tests.

THC emission factor of the studied fleet was on average, 12 times higher than the gasoline vehicles and 20 times higher than the diesel vehicles, reported in the literature. Results of the SOA prediction model showed that 125 *cc* motorcycles, which constitute more than 70% of the total motorcycles fleet in Tehran, also have the highest SOA emission factor. The average SOA formation from exhaust of motorcycles in Tehran is 28 times higher than the reported values for gasoline and diesel vehicles in the literature. The study also showed that the exhaust of a motorcycle in the city of Tehran can produce secondary organic aerosols which is 60 times higher than the direct emission of PM<sub>2.5</sub> from a Euro-4 passenger car and 3 times higher than the direct emission of particles from motorcycles in European countries. Among different groups of carburetor motorcycles under study, 180 *cc* engine volumes had the best condition, regarding the emission of regulated pollutants and SOA formation potential. An increase in THC and SOA emission factors was observed for older 125 *cc* engine volume motorcycles, which highlights the importance of technical inspection of motorcycles, as soon as three years after production. Replacing carburetor fuel delivery system with fuel injection and the use of catalytic converters are considered effective ways of lowering unburned hydrocarbons in the motorcycles combustion products, which can result in decreased SOA formation potential of the exhaust.

The novel approach to determine SOA formation potential of untreated exhaust can be applied towards determining the emission certification levels and annual inspection emissions of vehicles,

emphasizing on potential of particle production rather than direct exhaust emissions. The implemented method in the present study can be applied to any other untreated engine exhaust, including diesel vehicles without catalysts (DOC, SCR) and particulate filters (DPF) or aircraft engines. For that purpose, the volatility distribution of the fuel and the emission factor of organic compounds present in the exhaust, must be determined.

## **Acknowledgments**

The authors would like to acknowledge the Air Quality Control Company (AQCC, a subsidiary of Tehran Municipality) for their help and support during this work. We would like to thank Mr. Amin Hassani for providing detailed data from chassis dynamometer tests of motorcycles.

## **Funding**

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

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## Tables

**Table 1.** Mass fraction distribution and average reaction rate with OH radical for each volatility bin, corresponding to the quantified mass of the gasoline in the present study.

Volatility bin ( $\mu\text{g m}^{-3}$ )	$10^3$	$10^4$	$10^5$	$10^6$	$10^7$	$10^8$	$10^9$	$10^{10}$
Mass Fraction	0.000	0.000	0.031	0.038	0.162	0.241	0.189	0.038
$k_{\text{OH}}$ ( $\text{cm}^3 \text{molec}^{-1} \text{s}^{-1}$ )	3.7e-11	3.6e-11	8.3e-11	7.2e-11	6.7e-11	5.9e-11	3.0e-11	2.5e-11

**Table 2.** Number and composition of the motorcycles fleet under study. All selected from Tehran in-use fleet with carburetor fuel supply system, without any after-treatment devices.

Engine displacement volume (cc)	Sample size		
	Produced before 2012	Produced between 2012 and 2014	Total
125	23	13	36
150	2	6	8
180	6	4	10
200	4	2	6

**Table 3.** Chassis dynamometer test results, according to Euro-3 emissions certification test procedures for each group of engine displacement volume. Data are reported as arithmetic mean  $\pm$  one standard deviation.

Quantity		Engine displacement volume (cc)			
		125	150	180	200
Dilution air pressure ( <i>kPa</i> )	Phase 1	$87.6 \pm 0.2$	$87.7 \pm 0.4$	$87.4 \pm 0.2$	$87.4 \pm 0.2$
	Phase 2	–	$87.7 \pm 0.4$	$87.4 \pm 0.2$	$87.4 \pm 0.2$
Dilution air temperature ( $^{\circ}\text{C}$ )	Phase 1	$24.3 \pm 1.1$	$24.6 \pm 0.5$	$25.0 \pm 0.8$	$25.0 \pm 0.9$
	Phase 2	–	$26.9 \pm 1.0$	$27.7 \pm 1.1$	$27.7 \pm 1.0$
Dilution factor	Phase 1	$82.0 \pm 6.7$	$72.5 \pm 5.8$	$68.6 \pm 4.5$	$64.4 \pm 2.7$
	Phase 2	–	$27.6 \pm 1.4$	$29.1 \pm 1.1$	$25.4 \pm 0.7$
Emission bag volume ( $\text{m}^3$ )	Phase 1	$119.2 \pm 0.4$	$119.2 \pm 0.5$	$118.8 \pm 0.5$	$118.6 \pm 0.6$
	Phase 2	–	$40.1 \pm 0.2$	$40.0 \pm 0.2$	$40.0 \pm 0.2$
THC net concentration in bag ( <i>ppmC<sub>I</sub></i> )	Phase 1	$38.2 \pm 8.2$	$39.1 \pm 13.9$	$28.8 \pm 9.9$	$39.5 \pm 11.7$
	Phase 2	–	$49.2 \pm 14.7$	$54.8 \pm 25.1$	$49.4 \pm 16.4$
Fuel consumption ( <i>L/100 km</i> )	Phase 1	$2.4 \pm 0.4$	$2.9 \pm 0.4$	$2.8 \pm 0.4$	$2.9 \pm 0.2$
	Phase 2	–	$2.7 \pm 0.2$	$2.6 \pm 0.1$	$2.8 \pm 0.1$
Distance travelled ( <i>km</i> )	Phase 1	$6.0 \pm 0.1$	$5.9 \pm 0.2$	$6.0 \pm 0.03$	$5.6 \pm 0.8$
	Phase 2	–	$6.6 \pm 0.04$	$6.6 \pm 0.1$	$6.7 \pm 0.1$
THC emission factor ( <i>g km<sup>-1</sup></i> )	Phase 1	$0.44 \pm 0.1$	$0.46 \pm 0.17$	$0.33 \pm 0.11$	$0.45 \pm 0.13$
	Phase 2	–	$0.17 \pm 0.05$	$0.19 \pm 0.09$	$0.17 \pm 0.06$

**Table 4.** Average THC ambient concentrations for chassis dynamometer tests, according to Euro-3 emissions certification test procedures, for each group of engine displacement volume.

Engine volume (cc)	125	150	180	200
$C_{\text{amb}} (\mu\text{g m}^{-3})$	$1398 \pm 268$	$1706 \pm 511$	$1596 \pm 617$	$1699 \pm 530$

**Table 5.** Total hydrocarbon (THC) and particulate mass (PM) emission factors for a number of 4-stroke two-wheelers in Asia and Europe.

Fuel Supply System	Engine Volume (cc)	After-treatment	Test Cycle	THC EF <sup>a</sup> (g km <sup>-1</sup> )	PM EF (mg km <sup>-1</sup> )	Country	Reference
Carburetor	125	– <sup>b</sup>	ECE	–	8.48	Taiwan	Yang et al. (2008)
	125	None	ECE	0.65	–	Taiwan	Yao et al. (2009)
	< 150	2-way catalyst	ECE	0.63	–	Taiwan	Chiang et al. (2014)
	100	None	IDC <sup>c</sup>	9.5	1.6	India	Momenimovahed et al. (2014)
	50 (scooter)	2-way catalyst	ECE (cold/hot)	0.56/0.19	6.94/0.33	European example	Zardini et al. (2014)
	125	3-way catalyst	ECE	0.19	–	China	Li et al. (2015)
	150			0.15			
	125		ECE	0.51			
	150	None	ECE+EUDC	0.32	–	Iran	Hassani and Hosseini (2016)
	180		ECE+EUDC	0.25			
	200		ECE+EUDC	0.32			
	50 (Moped)	–	ECE	1	7.9	Netherlands	Eijk et al. (2017)
	125	–	ECE	0.9	–	Taiwan	Tsai et al. (2017)
	100	–	ECE	0.46	–	Vietnam	Duy et al. (2019)
	100	None	ECE	0.7	–	Vietnam	Hoang et al. (2019)
Injection	1000	3-way catalyst	ECE+EUDC	0.12	–	Italy	Iodice and Senatore (2015)
	1000	3-way catalyst	ECE+EUDC (hot & cold)	0.34	1.5	Italy	Costagliola et al. (2016)
	280	3-way catalyst	ECE+EUDC	0.14	–	Italy	Iodice and Senatore (2016)
	Variable	3-way catalyst	WMTC <sup>d</sup>	–	< 1.5	Italy	Giechaskiel et al. (2019)
	Variable	3-way catalyst	WMTC/ECE	–	1.4/3.4	European example	Kontses et al. (2020)

<sup>a</sup> Emission Factor

<sup>b</sup> – is unknown

<sup>c</sup> Indian Driving Cycle

<sup>d</sup> World-harmonized Motorcycle Test Cycle

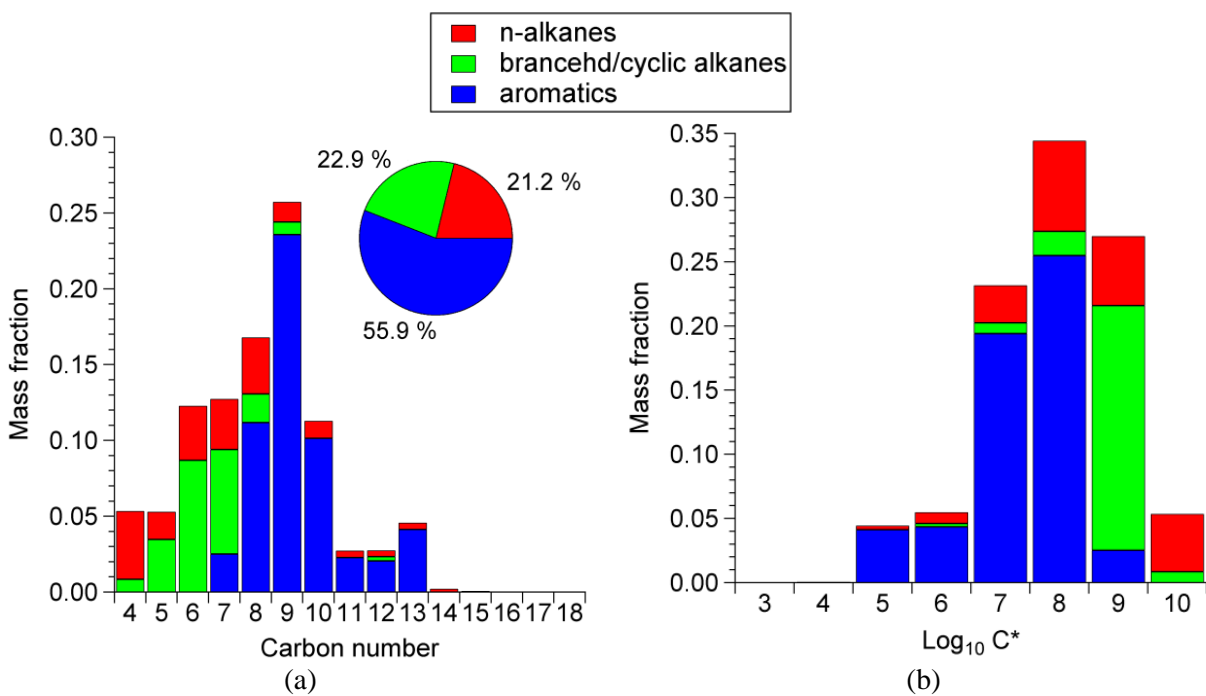
**Table 6.** SOA mass yield for the precursors in the volatility-dependent model (Jathar et al., 2014).

Precursor $C^*$ ( $\mu\text{g m}^{-3}$ )	Product $C^*$ ( $\mu\text{g m}^{-3}$ )											
	$10^{-4}$	$10^{-3}$	$10^{-2}$	$10^{-1}$	$10^0$	$10^1$	$10^2$	$10^3$	$10^4$	$10^5$	$10^6$	$10^7$
$10^3$	0.011	0.078	0.034	0.006	0.297	—	—	—	—	—	—	—
$10^4$	—	0.011	0.078	0.034	0.006	0.297	—	—	—	—	—	—
$10^5$	—	—	0.011	0.078	0.034	0.006	0.297	—	—	—	—	—
$10^6$	—	—	—	0.011	0.078	0.034	0.006	0.297	—	—	—	—
$10^7$	—	—	—	—	0.011	0.078	0.034	0.006	0.297	—	—	—
$10^8$	—	—	—	—	—	0.011	0.078	0.034	0.006	0.297	—	—
$10^9$	—	—	—	—	—	—	0.011	0.078	0.034	0.006	0.297	—
$10^{10}$	—	—	—	—	—	—	—	0.011	0.078	0.034	0.006	0.297

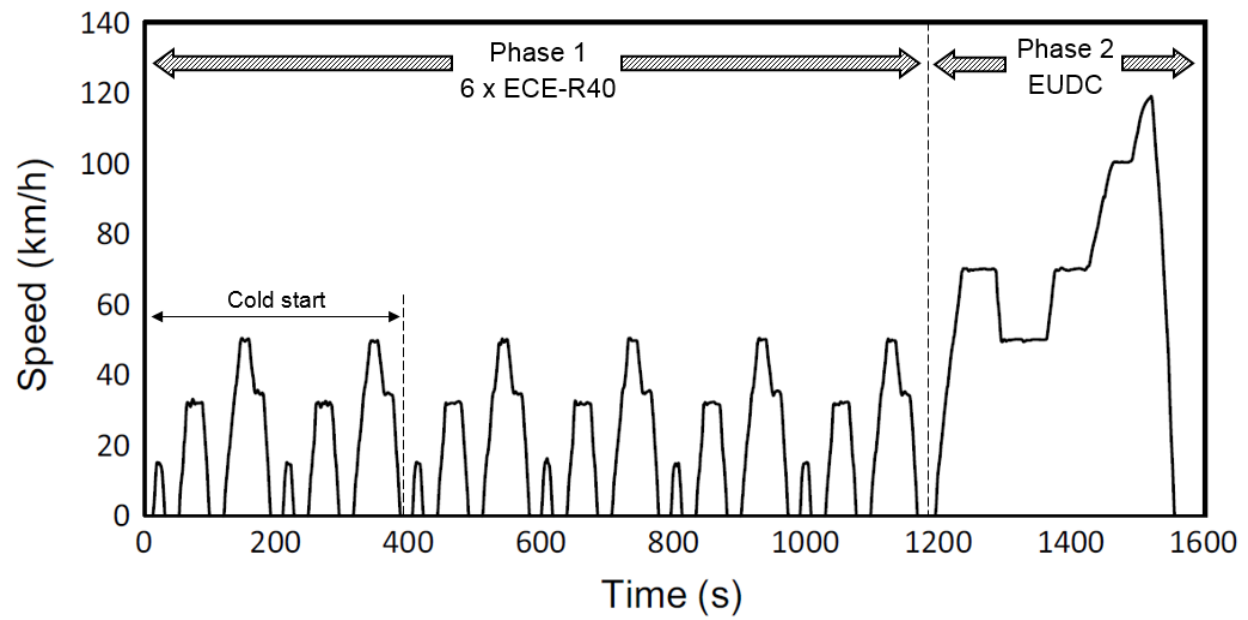
**Table 7.** SOA emission factors after 17 hours of photo-oxidation under ambient conditions, for different motorcycle engine volumes.

Engine volume (cc)	125	150	180	200
SOA concentration after 17 h of photo-oxidation ( $\mu\text{g m}^{-3}$ )	265	330	307	329
Fuel-based emission factor ( $\text{g kg-fuel}^{-1}$ )	4.6	2.5	2.4	2.6
Travel-based emission factor ( $\text{mg km}^{-1}$ )	80	50	50	60

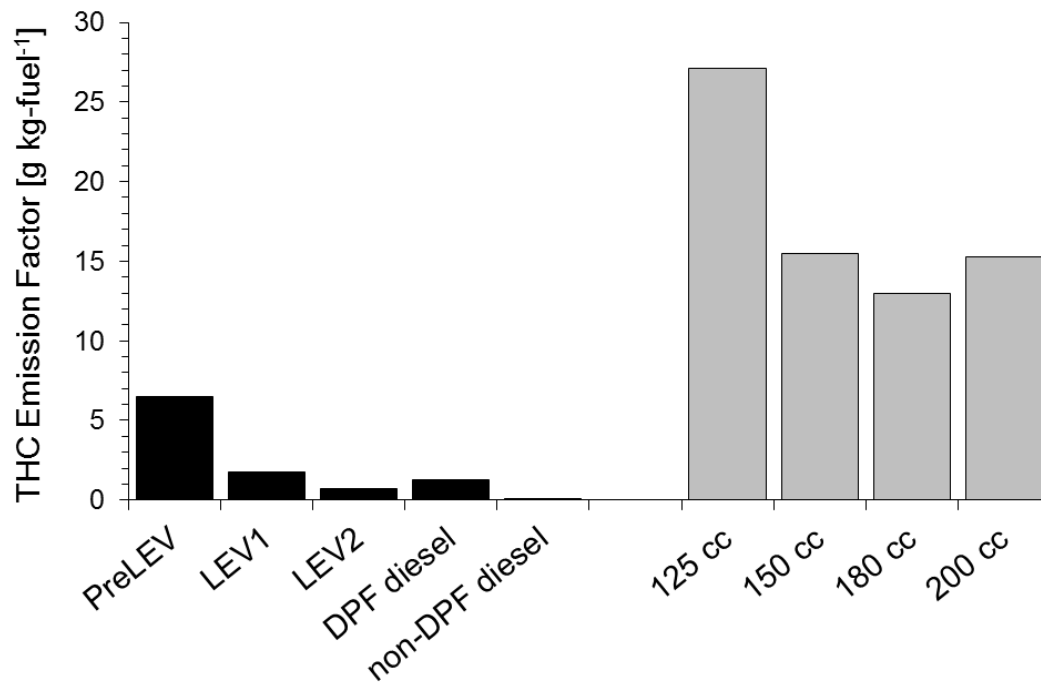
## Figures



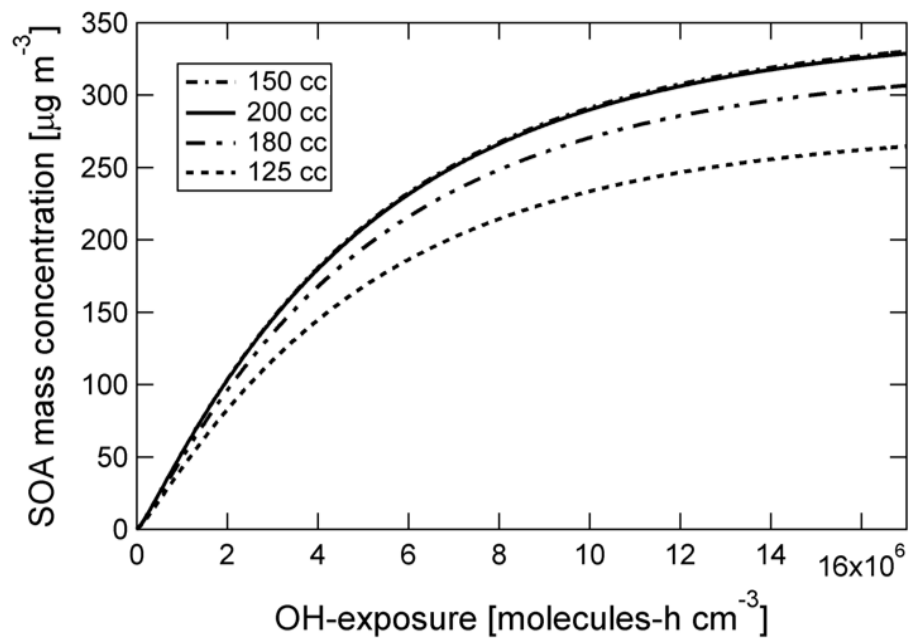
**Figure 1.** (a) Carbon number and (b) volatility, along with molecular structure distribution for Tehran regular gasoline. Inset pie shows the relative fraction of normal alkanes, branched/cyclic alkanes and aromatics.



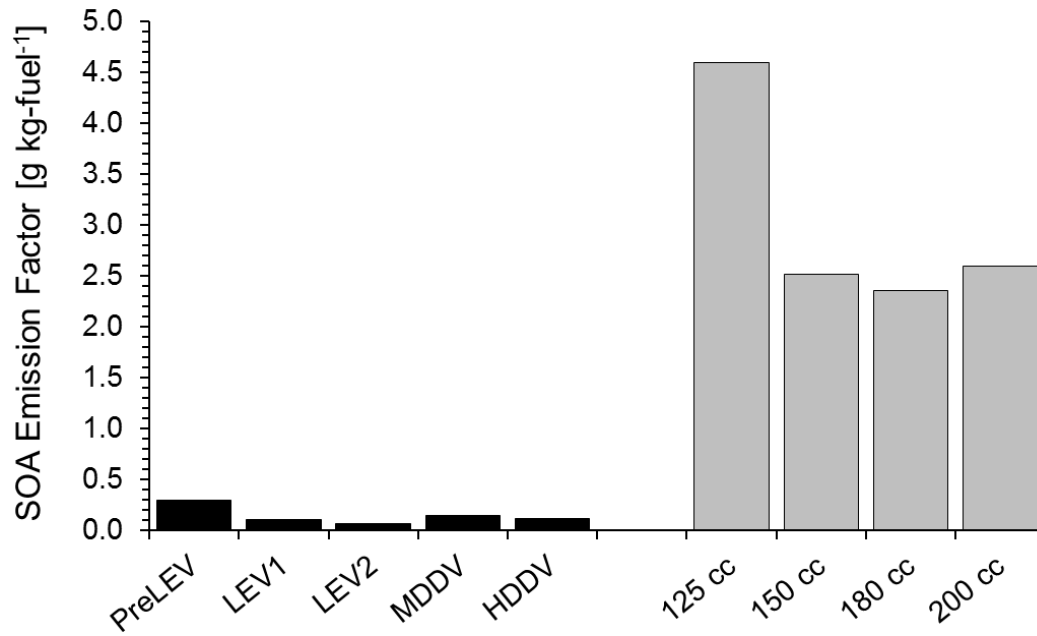
**Figure 2.** Driving cycle for testing motorcycles under Euro-3 emission standard. The EUDC is only used for motorcycles with engine volume larger than 125 cc.



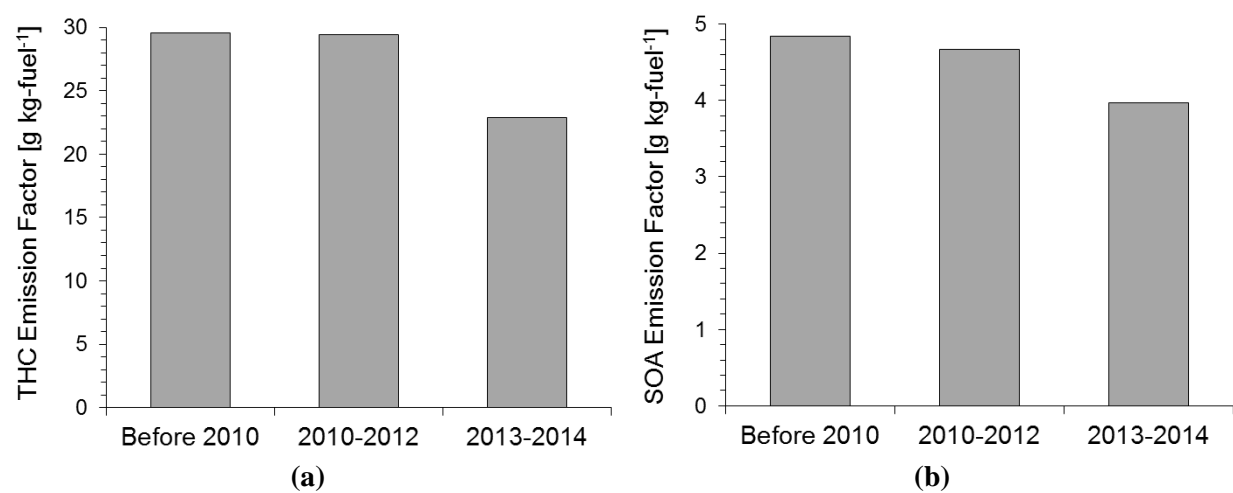
**Figure 3.** Comparison between fuel-based THC emission factors of carburetor motorcycles in the present study and vehicles under study in May et al. (2014) (PreLEV: LDGVs manufactured prior to 1994; LEV1: LDGVs manufactured between 1994 and 2003; LEV2: LDGVs manufactured in 2004 or later).



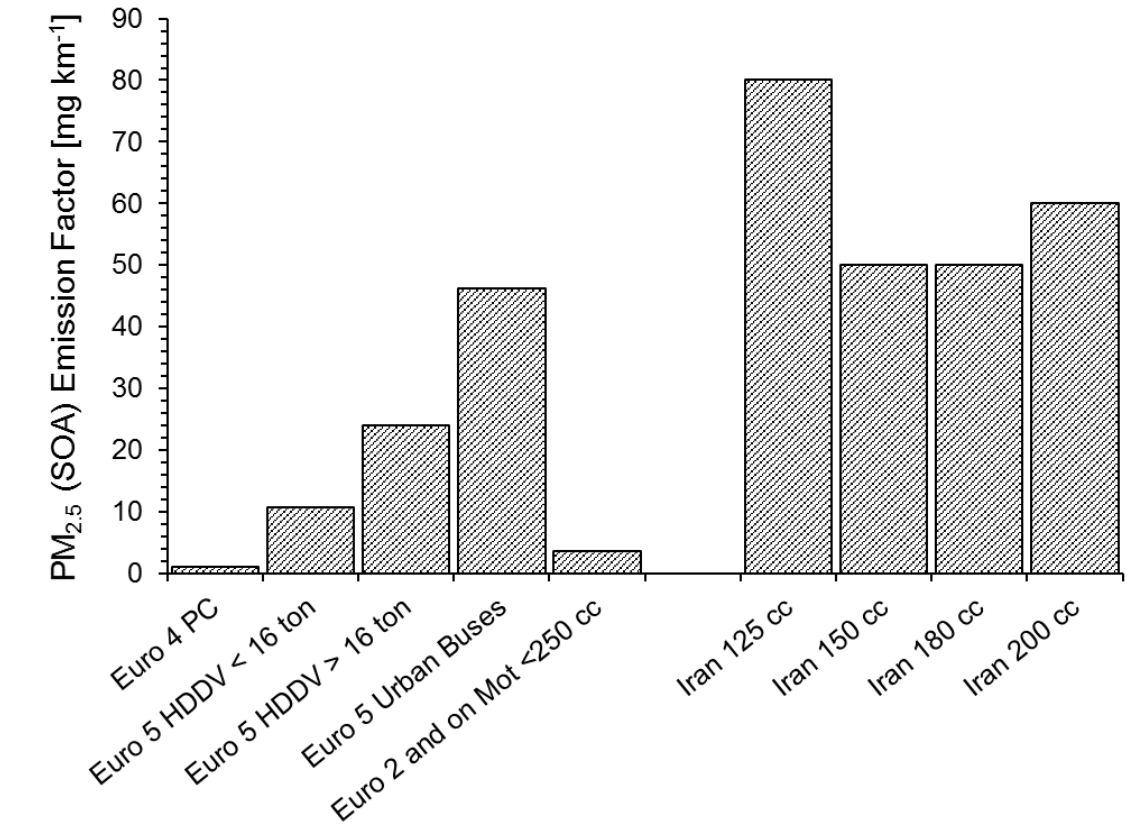
**Figure 4.** SOA formation potential of the exhaust from Tehran carburetor motorcycles, for different engine volumes, after 17 hours of photo-oxidation, using SOA mass yields matrix of Table 6.



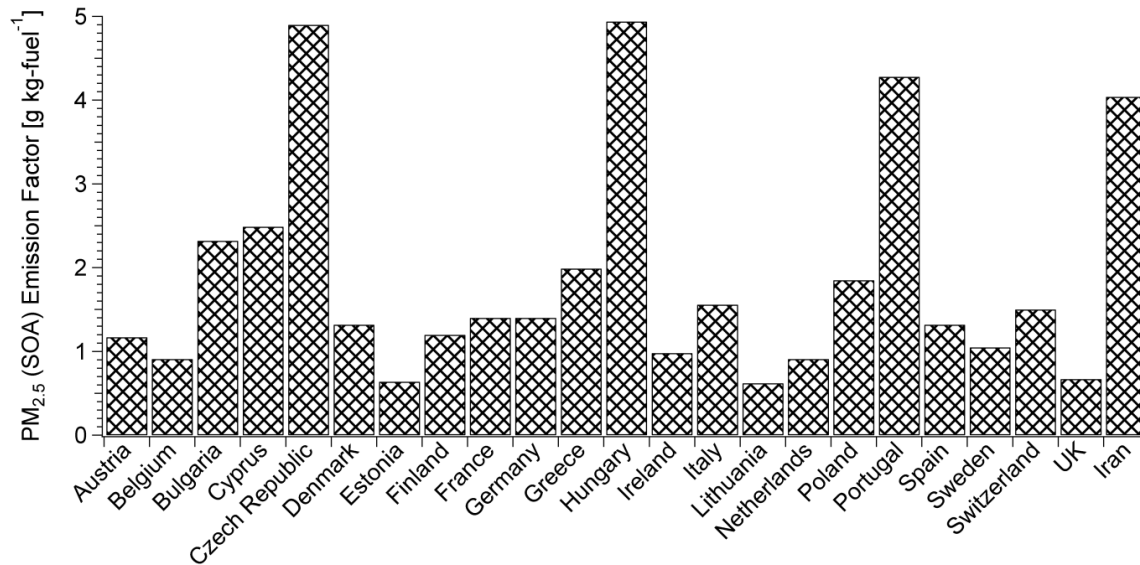
**Figure 5.** Comparison between fuel-based SOA emission factors of carburetor motorcycles in the present study and vehicles under study in May et al. (2014) (PreLEV: LDGVs manufactured prior to 1994; LEV1: LDGVs manufactured between 1994 and 2003; LEV2: LDGVs manufactured in 2004 or later).



**Figure 6.** Effect of aging on THC emission and SOA formation from 125 cc engine volume motorcycles. X-axis is categorized based on the production year of the motorcycles.



(a)



(b)

**Figure 7.** (a) Comparison between SOA emission factors of Tehran carburetor motorcycles and primary PM emission factors of different classes of euro-certified vehicles. (b) Comparison between average SOA emission factor of Tehran motorcycles with primary PM emission factors of motorcycles in different European countries (EMEP/EEA, 2018). Pay attention to the emission factor unit in each graph.