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In-situ Estimation of Non-Regulated Pollutant Emission Factors in an Urban Area of Nantes, France, with Fleet Composition Characterization

Simon Martinet[a], Yao Liu[a,*], Liliane Jean-Soro[b], Mathieu Goriaux[b], Michel André[a]

[a] Univ Lyon, IFSTTAR, AME, LTE, Bron, 69675, France
[b] GERS, LEE, IFSTTAR, Bouguenais, 44344, France
[*] Corresponding author: yao.liu@ifsttar.fr, Phone: +33 472 14 24 75, Fax: +33 472 37 68 37;
Address: 25 avenue François Mitterrand, 69675 Bron, France

Abstract

The purpose of this study is to estimate the in-situ emission factors of several pollutants (particle number [PN], black carbon [BC] and several volatile and semi-volatile organic compounds [VOCs and SVOCs]) in an urban area of Nantes, France, with real-world traffic conditions and characterization of the fleet composition. The fleet composition and driving conditions are characterized by the number of vehicles, their speeds and their types (passenger cars [PCs], light commercial vehicles [LCVs], heavy-duty vehicles [HDVs]) as well as their characteristics (make, model, fuel, engine, EURO emission standard, etc.). The number of vehicles passing on the boulevard is around 20,000 per day with about 44% of Euro 5 and Euro 6 vehicles. The impacts of fleet composition on emission were analyzed by ANOVA. The results show that the fleet composition has a significant impact on emissions for different pollutants. Higher percentage of gasoline PCs between Euro 4 to Euro 6 and Euro 4 diesel PCs induces more BC emission. Higher percentage of old gasoline and diesel vehicles (≤ Euro 3) induces higher emission of toluene, ethylbenzene and m+p- and o-xylene. Furthermore, emission factors estimated in this work were compared to those calculated in other in-situ studies that show a good agreement. For the chassis bench comparison, the in-situ PN and BC emission factors are in the same range as those measured for diesel vehicles without particle filter and gasoline vehicles with direct injection system. These EFs are also comparable with old heavy duty vehicles without particle filter ($5x10^{13}$-$2x10^{14}$ #/km).

Keywords

In-situ emission factors; Unregulated pollutants; Fleet composition

1. Introduction

On-road vehicle emissions are the main cause of atmospheric pollution in urban areas. Road transport induces particles, black carbon (BC) and of several VOCs and SVOCs (Volatile and Semi-Volatile Organic Compounds) emissions, such as carbonyl compounds (formaldehyde, hexanal), and BTEX (benzene, toluene, ethylbenzene, xylene) as well as various alkanes and alkenes. These VOCs and SVOCs are non-regulated compounds that could serve as secondary particle precursors and have serious negative impacts on human health (Sydbom et al., 2001; Lewtas et al., 2007) and air quality in cities.

The European Union is imposing emission limits for regulated pollutants in order to reduce road-traffic emissions. Facing on these vehicle emission standards, emission factors are derived from dynamometer bench test (Alves et al., 2015; Yang et al., 2015; Louis et al., 2016; Martinet et al., 2017) or from on-board emissions measurements (O’Driscoll et al., 2016; Ntziachristos et al., 2016). These emission factors constituted an input database with different vehicle categories using by emission models (e.g., COPERT, HBEFA, PHEM and MOVES) for air quality studies. However, for these emission models, the emission factors inventories for recent Euro 5 and Euro 6 vehicles are quite poor (Rexeis et al., 2013). Only eighty Euro 5 vehicles and twenty Euro 6 vehicles (with 13 different vehicle models, and only one Euro 6 gasoline car) were added to HBEFA Version 3.2 for regulated compounds, which may not be representative of the entire fleet composition. The emissions of non-regulated pollutants are rarely measured and integrated in emission models. As consequence, their emission factors for an entire fleet could not be estimated correctly actually due to this deficiency of database, and their impact on air quality and human health could not be investigated accurately.

Emission factors could be estimated in-situ for a part or an entire fleet using various methods. The first is the chasing method, where pollutant concentrations are measured by driving a mobile measurement platform behind either a single vehicle (Karjalainen et al., 2014; Ježek et al., 2015) or part of the fleet present on the road (Yli-Tuomi et al., 2005; Wang et al., 2009; Westerdahl et al., 2009; Kam et al., 2012; Ning et al., 2012; Hudda et al., 2013). In the second method, traffic pollutant concentrations are collected by a fixed measurement platform placed on the roadside. This method makes it possible to measure emissions for an entire fleet driving near the measurement site (Ketzel et al., 2003; Imhof et al., 2005; Rose et al., 2005; Jones et al., 2006; Bukowiecki et al., 2010). For most of these studies, the number of LDVs, HDVs and buses are counted and the traffic speeds are measured in some cases. However, the fleet compositions with vehicle engine, capacity, combustion, age and Euro emission standard were not fully characterized. As consequence, the impacts of fleet composition on non-regulated pollutant emissions are hardly investigated. Furthermore, the on-road emission factors in these cited studies were mainly calculated for particles and BC. Very few on-road emission factors studies were focused on secondary particle precursors (carbonyl compounds, BTEX and alkanes…). Ning *et al.* (2012) determined on-road emission factors for butane, but only for an HDV/bus fleet.

In this paper, the emission factors of particle number (PN), BC, and several aliphatic, aromatic and carbonyl compounds were estimated using the different concentrations of NO_X and a pollutant between a background measurement site and a traffic measurement site with the fleet composition observed during the measurements campaign. Furthermore, the fleet composition was characterized based on the vehicle type, make, model, fuel, engine, age, Euro emission standard, as well as the traffic conditions, traffic speed, and congestion.

ANOVA statistical analyses were performed to characterize the impact of fleet composition on emissions. Furthermore, the emission factors estimated in this study were compared to other emission factors calculated in previous in-situ studies and those measured on a dynamometer bench.

2. Experimental Method

2.1 Measurement Sites

The measurements were conducted at two sites in the city of Nantes, France, between April 19 and 30, 2017. The first site was an urban background (47°13'20.3"N 1°32'15.2"W) site to measure urban background pollutant concentrations over a period of four days. The second site was a traffic site in the city center (47°12'16.0"N 1°33'10.9"W). It is an urban boulevard with two lanes of traffic in each direction, a speed limit of 30 km/h, and traffic lights (Fig. 1). The number of vehicles passing on the boulevard is around 20,000 per day. Trucks and buses pass on the boulevard but with a low frequency. Measurements were conducted over a period of seven days.

2.2 Traffic Characterization

The fleet composition and traffic conditions on the traffic site were characterized by AlyceSofreco (a private company specialized in the field of traffic measurement). These data were recorded in both directions of traffic and over a period of seven days using two video cameras and pneumatic-tube automatic traffic counters. The counters determined the number of vehicles and the driving conditions (speed, traffic congestion, etc.) and the video cameras collected the license plate numbers of each vehicle. Using these license plate numbers, AAAData (a private company) provided the characteristics of each vehicle, including its make, model, vehicle type, fuel, engine, date of entry into circulation and Euro emission standard according to the vehicle type (LDV and HDV).

2.3 Sampling Devices

The sampling devices were installed in a truck that been specially fitted with an array of analyzers to sample the ambient air. The truck was placed along the edge of the road on the traffic site and the sampling was carried out around at a height about two meters and at a distance of 0.5 m of the road (Fig. 1). A Fast Mobility Particle Sizer (FMPS™; TSI) was used to measure the distribution and total particle number ranging from 5.6 to 560 nm with 1 scan/s at a flow rate of 8 L/min, with a concentration range from 0 to 10^7 particle/cm^3. An Aethalometer® (AE-33-7, Magee Scientific) was used to measure the BC concentration. The data are collected once a second and at a flow rate of 5 L/min. The concentration ranges from 10 to 10^5 ng/m^3 with a detection limit of 5 ng/m^3 for 1 hour. The device measures the light attenuation for seven wavelengths from UV to IR (370, 470, 525, 590, 660, 880 and 940 nm). The 880 nm wavelength corresponding to the maximum amount of BC was used for the quantification in this study.

VOCs and SVOCs were sampled on different cartridges with 1 sample per hour. DNPH and Tenax® cartridges were used to collect respectively carbonyl compounds and BTEX and five majority alkanes with a flow rate of 1 L/min and 0.1 L/min. A private laboratory, TERA Environnement, analyzed the cartridges (78 TENAX and 78 DNPH, including 28 for the background site, 49 for the traffic site and 1 for the transport blank for each type of cartridge) using standardized analytical methods (ISO-16000-6, ISO 16000-3, NIOSH 2549, NIOSH 5506 and NF X 43-025). The complete list of compounds (six BTEX, five alkanes and 11 carbonyl compounds) analyzed on the cartridges is given below.

- BTEX: benzene, toluene, ethylbenzene, m-xylene, p-xylene, o-xylene.
- Alkanes: nonane, decane, undecane, cyclopropane, ethyl, cyclohexane, ethyl.
- Carbonyl compounds: formaldehyde, acetaldehyde, acetone, acrolein, propionaldehyde, crotonaldehyde, methacrolein, butanal, benzaldehyde, pentanal, hexanal.

Fig. 1. Diagram of the traffic site used for the measurements.

2.4 Emission Factors

The variation of NO_X and pollutant concentrations between the background site and the traffic site were used for estimating road-traffic emission factors (Imhof et al., 2005; Jones et al. 2006; Johansson et al., 2009; Krecl et al., 2017). This method is based on the assumption that the dilution of the pollutant between the exhaust outlets of vehicles and the sampler device inlets and the behavior in the atmosphere are comparable for NO_X and the other pollutants (Lohmeyer et al., 2002; Amato et al., 2010; Bukowiecki et al., 2010; Gietl et al., 2010). Using this assumption, it is considered that the dilution of other pollutants may be approximated by the dilution of NO_X. The wind speed (Fig. 2) and directions during the campaign varied between 0 and 7.6 m/s, with principally the north direction and few times the south and west direction. The temperature varied between 0 and 19 °C with relative humidity at about 30-99%. The following equation is used to calculate the emission factor for a given pollutant:

$$EF_P = \Delta P \times \frac{EF_{NOx}}{\Delta NOx} \qquad (1)$$

where EF_P and EF_{NOx} are the emission factor for pollutant P and NO_X, respectively, given in mass or number of particles per vehicle per kilometer (#/veh/km or µg/veh/km). ΔP and ΔNOx are the difference of concentrations between background and traffic sites for pollutant P and NO_X respectively. The emission factors for NO_X used in the equation (1) were obtained using COPERT 4 (COmputer Programme to calculate Emissions from Road Transport) for each time step (one-hour period) with the corresponding fleet composition characterized at the same time step. For urban driving conditions, COPERT 4 estimates a NO_X emission that takes into account cold and hot emissions.

For all cartridges, the sampling time is one hour to have enough material for chemical analysis. For PN and BC, the data resolution is 1 second. However, in order to be able to analyze the emission factors with the fleet for all measured pollutants, the PN and BC measurement were averaged in one-hour period, corresponding to the cartridge sampling time. In addition, the NO_X concentrations used for ΔNO_X in Equation 1 are given as a 15-minute average concentration. It is therefore impossible to go below this 15-minute time step for calculating emission factors.

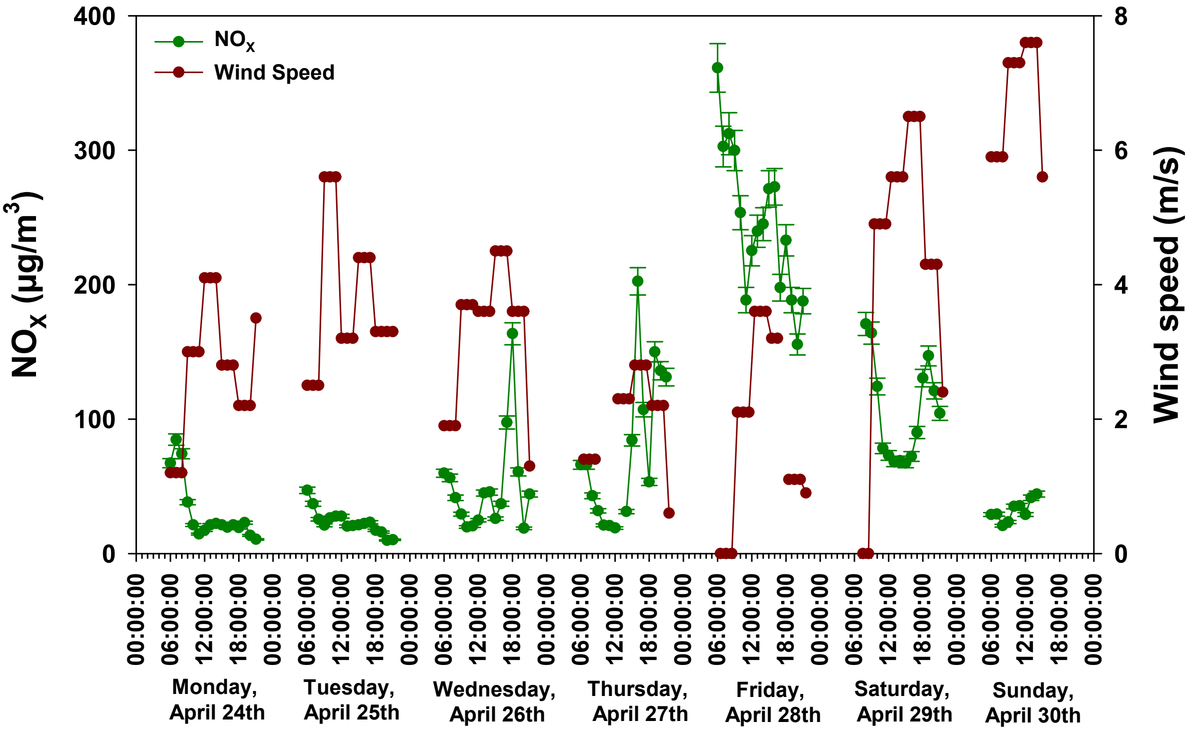

Fig. 2. Time profiles of NO_X concentration and wind speed on the measurement week.

2.5 ANOVA Analysis

The analysis of variance (ANOVA) is a statistical technique for assessing the differences between the dependent variables, which are the emissions, of a nominal variable with several categories (composition of the fleet). The null hypothesis (H0) for the analysis represents the fact that there is no significant difference between the groups. The alternative hypothesis considers that there is at least one significant difference among the groups. For the ANOVA test, the F-ratio and associated probability value (p-value) are calculated. If the p-value associated with the F is smaller than 0.05, then the H0 is rejected and the alternative hypothesis (H1) is retained (Fanelli et al., 2018) and this implies that the groups have a significant impact on emissions (Wildt and Ahtola, 1978). It can be concluded that the means of all groups are not equal and we can determine which groups are different from others.

In this work, ANOVA was performed by SPAD (data analysis and data mining software) to determine impact of fleet composition on emission factors. The nominal-leval variables used are the number of HDVs, the percentage of diesel vehicles between pre-Euro and Euro 3, the percentage of Euro 4 diesel, the percentage of diesel between Euro 5 and Euro 6, the percentage of gasoline vehicles between pre-Euro and Euro 3 and the percentage of gasoline between Euro 4 and Euro 6.

3. Results and Discussion

3.1 Fleet Composition

The number of vehicles was characterized during the measurement on the traffic site. A total of 140,076 vehicles drove along the boulevard during the seven-day measurement period. Many vehicles were registered twice or more times during the week. The number of unique vehicles after removing these duplicates is 57,220. The number of vehicles varied between 21,147 and 23,401 from Monday to Friday. On Saturday and Sunday, 18,763 vehicles and 9,860 vehicles were counted, respectively. The number of vehicles per hour was between 600 and 1800 with an average number of 1400/h for all five weekdays and between 200 and 1600 on the weekend with an average number of 900/h. The traffic speed was characterized for each 5 minutes. The average traffic speed for one hour was between 16 and 34 km/h on all five weekdays and 21 and 38 km/h on the weekend (

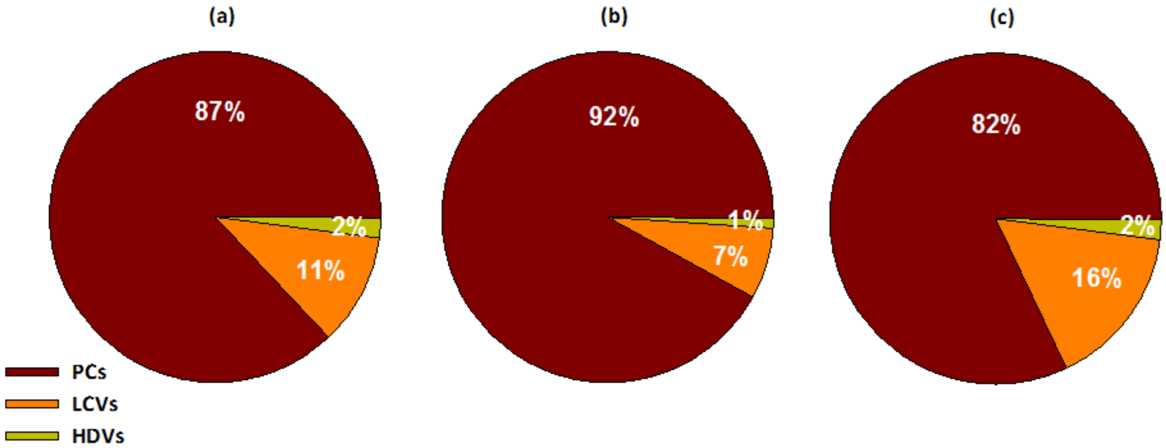

Fig. 3. Weekdays (a) and weekend (b) characterization of the fleet composition during the measurement campaign. 2017 composition of the French fleet (c).

Furthermore, the fleet composition characterized during the measurement campaign was composed of 87% passenger cars (PCs), 11% light commercial vehicles (LCVs), and 2% heavy-duty vehicles (HDVs) on weekdays. On the weekend, it consisted of 92% PCs, 7% LCVs, and 1% HDVs. The number of LCVs and HDVs decreased over the weekend, particularly on Sundays. According to André *et al.* (2014), the entire French fleet was composed of 82% PCs, 16% LCVs, and 2% HDVs in 2017 (Fig. 3). The comparison between this study and the French fleet composition shows the slightly higher percentage of PCs and the slightly lower percentage of LCVs. These differences are explained by the fact that the measurements were conducted in an urban environment, where LCV and HDV traffic is generally lower.

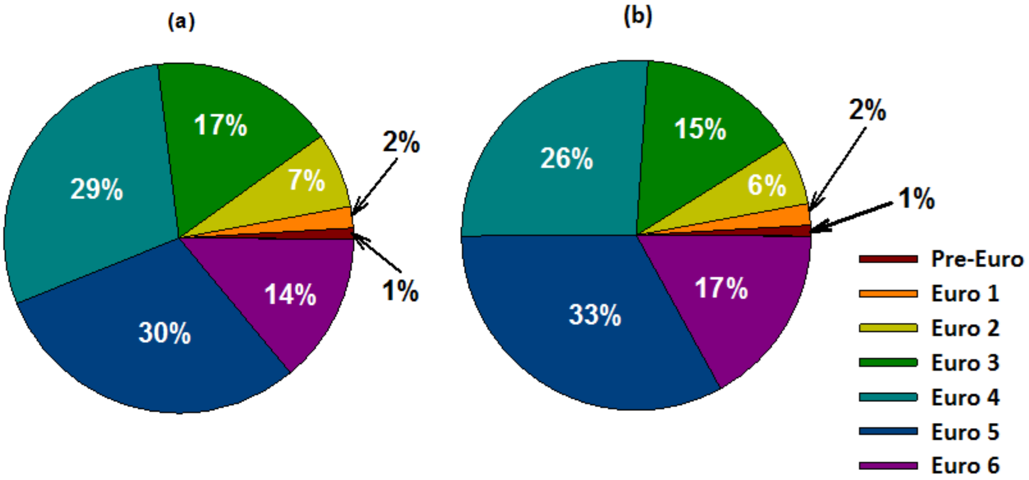

Fig. 4. Distribution of the various Euro emission standards for PCs (a) during the measurement campaign and (b) for the 2017 French fleet.

The distribution of the various Euro emission standards for PCs during the measurements was 14% Euro 6, 30% Euro 5, 29% Euro 4, 17% Euro 3, 7% Euro 2, 2% Euro 1, and 1% pre-Euro vehicles (Fig. 4a). This distribution is consistent with André et al. (2014), who estimated the PCs French fleet composed 17% Euro 6, 33% Euro 5, 26% Euro 4, 15% Euro 3, 6% Euro 2, 2% Euro 1, and 1% pre-Euro in 2017 (Fig. 4b). As regards drive technology systems, the observed PC fleet was composed of 68% diesel vehicles, 30% gasoline vehicles, and 2% other such as gasoline or diesel hybrid vehicles, electric vehicles, gasoline/compressed natural gas and gasoline/liquefied petroleum gas vehicles. The LCV fleet was composed of 97% diesel vehicles. For the HDVs, all the trucks were powered by diesel engines and all the buses were natural gas combustion or other gaseous hydrocarbons.

3.2 Estimation of Global Fleet Emission Factors

The global fleet emission factors for PN, BC, carbonyl compounds, BTEX, alkanes, NO_X concentrations, number of vehicles, and traffic speeds are presented in Fig. 5 as a function of the measurement time and day on the traffic site. The average emission factor of one vehicle (equation (1)) has been multiplying by the vehicle number that gives us the pollutant emission factors for a global fleet, over the one-hour measurement period. This allows us to study the pollutant emissions in relation with the current traffic.

The particle number emission factors varied between 2×10^{16} and 9.6×10^{17} #/km on five weekdays and between 2.3×10^{16} and 4.6×10^{17} #/km on the weekend. The BC emission factors varied between 2.4×10^{9} and 2.4×10^{11} ng/km on weekdays and between 6.6×10^{9} and 1.2×10^{11} ng/km on the weekend. The emission factors for carbonyl compounds varied between 2×10^{4} and 2.4×10^{6} µg/km on weekdays and between 2.7×10^{3} and 4.9×10^{5} µg/km on the weekend. The hexanal emission factors ranged between 2.3×10^{3} and 4.1×10^{5} µg/km on weekdays and between 1.5×10^{4} and 3.2×10^{5} µg/km on the weekend. For the three alkanes, the emission factors were between 1.2×10^{4} and 7.1×10^{6} µg/km on weekdays and between 5.3×10^{3} and 2×10^{6} µg/km on the weekend. The BTEX emission factors ranged between 5.4×10^{3} and 3.3×10^{6} µg/km on weekdays and between 6.3×10^{4} and 5.1×10^{6} µg/km on the weekend. The missing points for VOCs are either pollutant quantities sampled on the cartridges below their quantification limit; or negative values by subtracting the background value.

The urban background concentrations for the pollutants used in the equation (1) were measured on the urban background site used by the air quality association “Air Pays de la Loire”. This background site is not affected by road traffic emissions, the measured NO_X, PN and BC concentrations vary little on the different measurement time (16h/day during 4 days) (between 3200 and 4800 #/cm^3 for PN, between 400 and 700 ng/m^3 for BC, and between 5 and 50 µg/m^3 for NO_X) and provides a general background concentration with all different sources. The subtraction in the equation (1) allows removing only the general back ground levels but not the local background levels. This might induce a potential bias with an over estimation of EF for PN and other pollutants if the local background levels are higher than general background levels. The local background levels could also be measured when there is no traffic. However, depending on the weather conditions, especially the wind speeds, the accumulation phenomena under no traffic condition might also induce a potential bias on our measurement. The first point in the morning (at around 6:00 a.m.) was taken as the background level at the traffic site. Using this local background levels, the emission factors of PN and BC are respectively between 5×10^{15} and 9×10^{17} #/km and 1.2×10^{8} and 2.3×10^{11} ng/km. The local background values induce an underestimation of 4 and 20 times for PN and BC comparing to the 'Air Pays de la Loire' background site, but only for low emission period. These background values do not induce a significant difference at high emission period for both PN and BC. Overall, if we use the local background value, it induced an average underestimation around 30% for all period of measurement.

In general, the highest emission factors were measured between 7:00 a.m. and 10:00 a.m. and between 5:00 p.m. and 8:00 p.m. on all five weekdays. For the Saturday, the highest emission factors were measured between 10:00 a.m. and 1:00 p.m. and between 5:00 p.m. and 8:00 p.m. For the Sunday, between 6:00 a.m. and 3:00 p.m., high emission factors were measured for carbonyl compounds, BTEX and alkanes. The higher emission factors measured in the morning can be explained by vehicle cold start, which emits large amounts of pollutants such as BC, PN, BTEX, and carbonyl compounds (Westerholm et al., 1996; Joumard et al., 2000; Sluder et al., 2000; Caplain et al., 2006; Louis et al., 2016; Martinet et al., 2017).

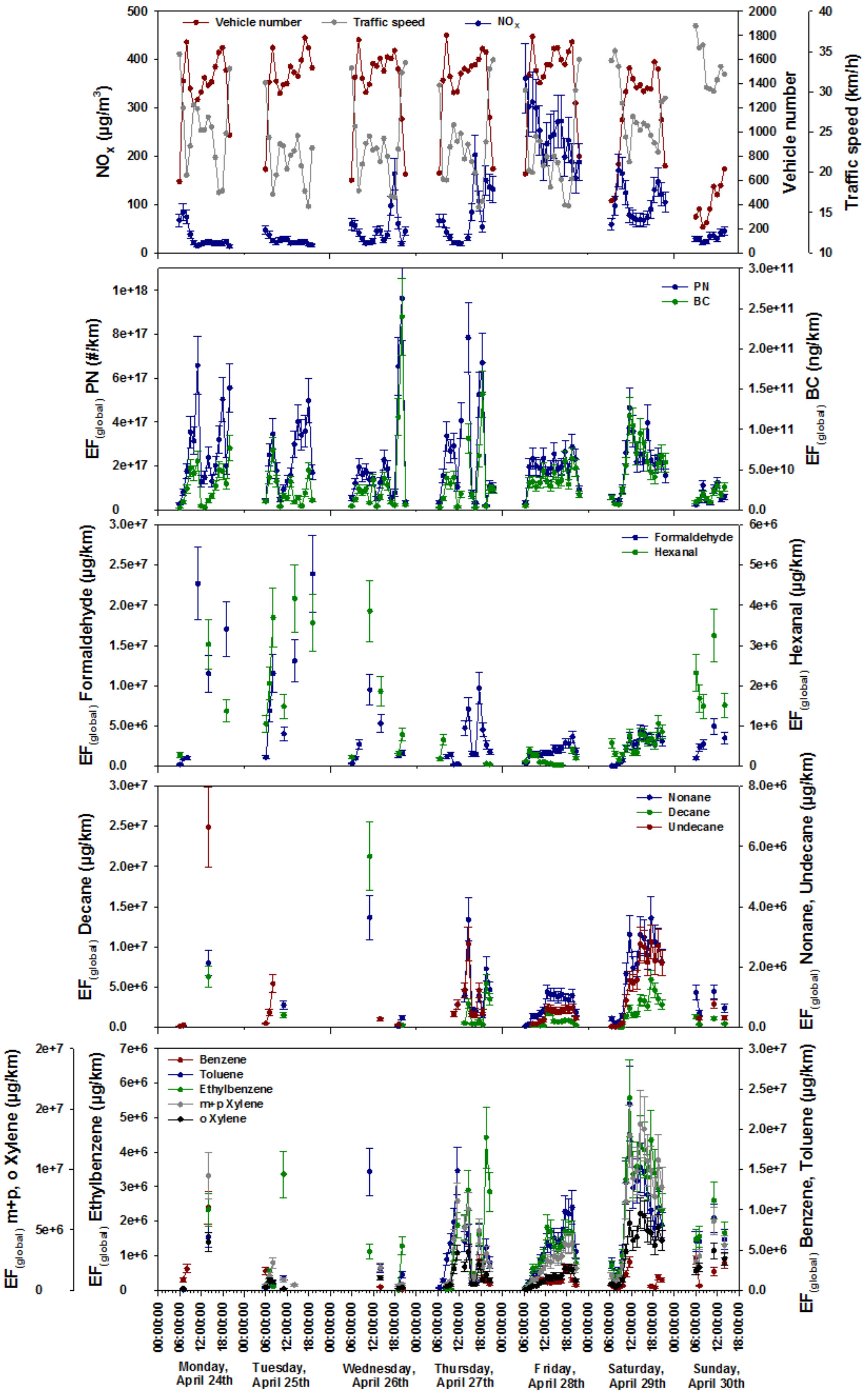

Fig. 5. Global fleet emission factors for PN, BC, carbonyl compounds, BTEX, alkanes, NO_X concentrations, number of vehicles, and traffic speeds as a function of the time and day of measurement on the traffic site.

The PN and BC emission factors showed a good correlation, following the same tendency during the week of measurements (Fig. 5). Apart from Tuesday afternoon between 2:00 p.m. and 8:00 p.m., the PN emission factors did not follow the same tendency as the BC emission factors. The emission factor values for BC decreased while those for PN increased sharply. This could imply that the particle number emissions measured during this afternoon were not due to road traffic (See Section below).

3.3 PN Concentration and Size Distribution Time Profiles

The number of vehicles, BC, NO_X and PN for three size ranges ([0-50] nm, [50-100] nm, and [100-500] nm) concentrations were followed from Monday (April 24th) to Sunday (April 30th), which show the similar results in general for the five weekdays, except for Tuesday afternoon. Fig. 6 shows the time profiles of NO_X, BC, PN concentrations and vehicles number on Monday, Tuesday and Wednesday. Concentration peaks of BC, PN and NO_X was observed over a short period between 7:00 a.m. and 9:00 a.m. on weekdays corresponding to morning rush-hour traffic (See Section 3.2).

Moreover, Fig. 6 shows a large peak for PN with size range [0-50] nm between 2:00 p.m. and 8:00 p.m. on Tuesday that is not correlated with BC, NO_X and PN size ranges [50-100] nm and [100-500] nm. Since NO_X and BC are considered as traffic tracers (Pant et al., 2013), which might indicates that the [0-50] nm PN on Tuesday afternoon is not generated by the road traffic.

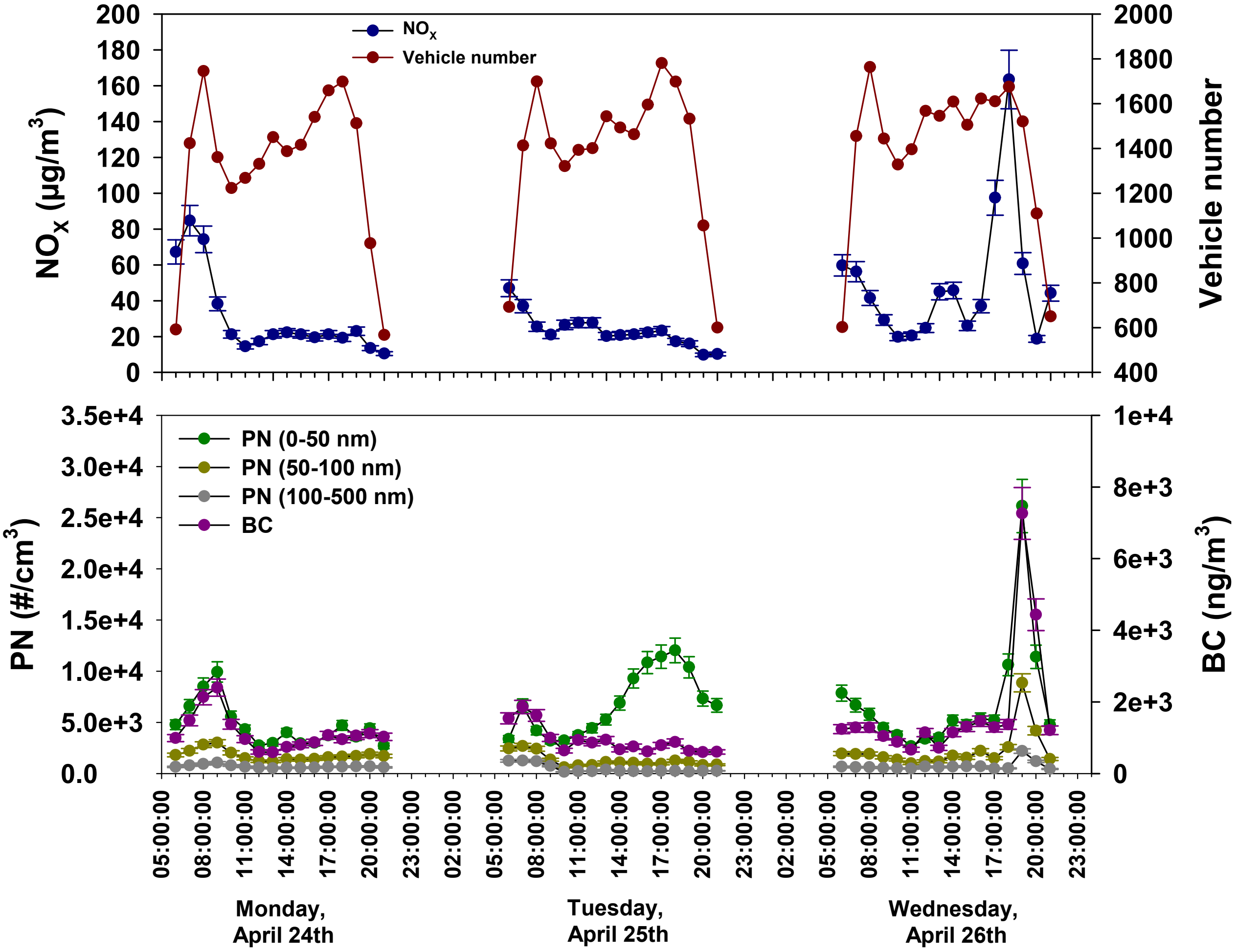

Fig. 6. Time profiles of NO_X concentration and number of vehicles and PN and BC concentrations on Monday, Tuesday and Wednesday.

3.4 ANOVA Analysis of Global Fleet Emission Factors

The pollutant emission factors for a global fleet, over the one-hour measurement period, have been analyzed by ANOVA statistical analysis to investigate impact of fleet composition on measured pollutant emissions (Table 1). The emission factors were analyzed with 6 categories: number of HDVs (0 to 43 with groups of every 10 HDVs), percentage of diesel PCs between pre-Euro to Euro 3 (5 to 25% with 5% interval), percentage of Euro 4 diesel PCs (10 to 30% with 5% interval), percentage of diesel PCs Euro 5 and Euro 6 (15 to 35% with 5% interval), percentage of gasoline PCs between pre-Euro and Euro 3 standard (5 to 15% with

5% interval) and percentage of gasoline PCs between Euro 4 and Euro 6 (5 to 25% with 5% interval). The groups of variables for passenger cars have been made according to the implementation of new after-treatment or engine technologies. For diesel vehicles, the diesel particle filter (DPF), which significantly reduces PN and BC emissions, is considered as variables. Thus, the first category includes the percentage of pre-Euro to Euro 3 diesel vehicles that are not equipped with DPF; the second category includes the percentage of Euro 4 diesel vehicles that are partially equipped with DPF; and the last category includes the percentage of Euro 5 and Euro 6 diesel vehicles that are all equipped with DPF. For gasoline vehicles, the first category therefore includes the percentage of gasoline vehicles from pre-Euro to Euro 3 standards, all of which have indirect injection engines; and the second category includes the percentage of gasoline vehicles from Euro 4 to Euro 6 standards, as the introduction of direct injection engines has begun on Euro 4 standard vehicles. Moreover, for ANOVA analysis, the fleet composition has been classed by groups (different gaps of vehicle number or percentage) to investigate their impact on emissions. Each group has to contain enough samples to be significant and not too large to have a good sensibility. Tuesday afternoon PN data have not been taken into account because of its strange behavior show in section 3.3.

For a result of the ANOVA analysis to be significant, the p-value must be ≤ 0.05, which is called a "significant result" in Table 1. However, to increase the power of the ANOVA analysis, it is possible to consider that a result with a p-value between 0.05 and 0.1 are significant, which is called "result considered as significant" in Table 1, but with a great uncertainty. This analysis should be read with a special attention because they might also indicate that the impacts of the analyzed group on emission could be significant but not clearly significant.

Table 1

Results of the ANOVA analysis for the effect of the fleet composition on the emission factors

Fleet composition	Pollutants	Unit	p-value	Groups having a significant impact	
				Negative impact	Positive impact
Number of HDVs	PN	#/km	0.6	--	--
	BC	ng/km	0.2	--	--
	Formaldehyde	µg/km	0.2	--	--
	Hexanal	µg/km	0.6	--	--
	Benzene	µg/km	0.6	--	--
	Toluene	µg/km	0.004*	20-30; 30-40	10-20
	Ethylbenzene	µg/km	0.04*	30-40	0-10; 10-20
	m+p-Xylene	µg/km	0.2	--	--
	o-Xylene	µg/km	0.1	--	--
	Nonane	µg/km	0.5	--	--
	Decane	µg/km	0.4	--	--
	Undecane	µg/km	0.7	--	--
Percentage of diesel PCs between pre-Euro to Euro 3 standard	PN	#/km	0.08**	15-20%	10-15%
	BC	ng/km	0.6	--	--
	Formaldehyde	µg/km	0.3	--	--
	Hexanal	µg/km	0.2	--	--
	Benzene	µg/km	0.6	--	--
	Toluene	µg/km	0.02*	10-15%	15-20%
	Ethylbenzene	µg/km	0.07**	10-15%	15-20%
	m+p-Xylene	µg/km	0.02*	10-15%	15-20%
	o-Xylene	µg/km	0.02*	10-15%	15-20%

	Nonane	µg/km	0.1	--	--
	Decane	µg/km	0.9	--	--
	Undecane	µg/km	0.7	--	--
Percentage of diesel PCs Euro 4 standard	PN	#/km	0.1	--	--
	BC	ng/km	0.001*	10-15%; 15-20%	20-25%; 25-30%
	Formaldehyde	µg/km	0*	20-25%; 25-30%	10-15%; 15-20%
	Hexanal	µg/km	0.7	--	--
	Benzene	µg/km	0.3	--	--
	Toluene	µg/km	0.1	--	--
	Ethylbenzene	µg/km	0.02*	15-20%	20-25%
	m+p-Xylene	µg/km	0.04*	15-20%	20-25%
	o-Xylene	µg/km	0.02*	15-20%	20-25%
	Nonane	µg/km	0.3	--	--
	Decane	µg/km	0.9	--	--
	Undecane	µg/km	0.6	--	--
Percentage of diesel PCs between Euro 5 to Euro 6 standard	PN	#/km	0.006*	30-35%	20-25%
	BC	ng/km	0.2	--	--
	Formaldehyde	µg/km	0.06**	15-20%	30-35%
	Hexanal	µg/km	0.3	--	--
	Benzene	µg/km	0.07**	15-20%	30-35%
	Toluene	µg/km	0.2	--	--
	Ethylbenzene	µg/km	0.2	--	--
	m+p-Xylene	µg/km	0.1	--	--
	o-Xylene	µg/km	0.06**	15-20%	30-35%
	Nonane	µg/km	0.2	--	--

	Decane	µg/km	0.6	--	--
	Undecane	µg/km	0.007*	15-20%	30-35%
Percentage of gasoline PCs between pre-Euro to Euro 3 standard	PN	#/km	0.5	--	--
	BC	ng/km	0.7	--	--
	Formaldehyde	µg/km	0.9	--	--
	Hexanal	µg/km	0.07	--	--
	Benzene	µg/km	0.4	--	--
	Toluene	µg/km	0.07**	5-10%	10-15%
	Ethylbenzene	µg/km	0.004*	5-10%	10-15%
	m+p-Xylene	µg/km	0.007*	5-10%	10-15%
	o-Xylene	µg/km	0.02*	5-10%	10-15%
	Nonane	µg/km	0.2	--	--
	Decane	µg/km	0.9	--	--
	Undecane	µg/km	0.7	--	--
Percentage of gasoline PCs between Euro 4 to Euro 6 standard	PN	#/km	0.004*	5-10%	20-25%
	BC	ng/km	0*	5-10%; 10-15%	20-25%
	Formaldehyde	µg/km	0.5	--	--
	Hexanal	µg/km	0.6	--	--
	Benzene	µg/km	0.4	--	--
	Toluene	µg/km	0.3	--	--
	Ethylbenzene	µg/km	0.1	--	--
	m+p-Xylene	µg/km	0.1	--	--
	o-Xylene	µg/km	0.1	--	--
	Nonane	µg/km	0.3	--	--
	Decane	µg/km	0.5	--	--

	Undecane	µg/km	0.07**	5-10%; 10-15%	20-25%

* Significant results

** Results considered as significant

-- No results

For BC, the groups of “20-25%” and “25-30%” Euro 4 diesel PCs and “20-25%” gasoline PCs between Euro 4 to Euro 6 have a significant positive impact on the emissions. This observation indicates that higher percentage of these two categories present in the fleet induces more BC emission. For the gasoline PCs between Euro 4 to Euro 6, the group “20-25%” has also a significant positive impact on the PN emissions. The positive impact of gasoline PCs between Euro 4 and Euro 6 on the PN and BC emissions can be explained by the introduction of direct injection technology on certain gasoline vehicles, which induces more PN and BC emissions than multipoint injection gasoline vehicles. These emissions could reach the level of some diesel vehicles without a particulate filter (Liang et al., 2013, Martinet et al., 2017).

The percentage of diesel and gasoline PCs between pre-Euro and Euro 3 has a significant positive impact on toluene, ethylbenzene and m+p- and o-xylene emission factors, more particular the groups “15-20%” and “10-15%” respectively. Comparing to the average emission factors (1.4×10^{6} µg/km for toluene, 3.4×10^{5} µg/km for ethylbenzene, 9×10^{5} µg/km for m+p-xylene and 4.2×10^{5} µg/km for o-xylene), the group “15-20%” of Pre-Euro to Euro 3 diesel induces 1.5, 1.4, 1.6 and 1.6 times higher emission respectively for toluene, ethylbenzene, m+p-xylene and o-xylene. The group “10-15%” of Pre-Euro to Euro 3 gasoline induces 1.2, 1.4, 1.3 and 1.3 times higher emission respectively for toluene, ethylbenzene, m+p-xylene and o-xylene. Moreover, the group “20-25%” of diesel Euro 4 PCs has also a positive impact on ethylbenzene and m+p- and o-xylene emission factors, 1.4, 1.4 and 1.5

times higher respectively comparing to the average emission factors. For the analyses on the number of HDVs, emission factors estimated in the weekend have not been taken into account because of the HDV driving ban. The Table 1 showed that the impact of the number of HDVs on pollutant emission does not show a significant positive impact. This observation might be explained on one side by the small percentage of HDVs in the fleet (0 to 43 HDVs per hour); and on the other side by the low HDVs emission since 80% of these HDVs are recent vehicles (≥ Euro 4).

3.5 Emission Factors Per Vehicle and Comparison with Other Studies

In this section, the average emission factors calculated by equation (1) were used (# or mass/veh/km) in order to compare with other studies. Fig. 7 shows the box-and-whisker plot for these average emission factors per vehicle for carbonyl compounds, BTEX, alkanes, PN, and BC. In these plots, the boxes contain 50% of the emission factors around the median (black line in the box). The upper and lower halves of the boxes represent respectively the 75th and 25th percentiles and the whiskers represent the 90th and 10th percentiles. The emission factors that are above or below the whiskers are considered to be atypical values.

Fig. 7 shows the average emission factors per vehicle for PN and BC, which are between 1.2×10^{13} and 8.7×10^{14} #/km/veh and between 1.7×10^{6} and 2.16×10^{8} ng/km/veh, respectively. For both emission factors, the median is in the middle of the box, which indicates a symmetric distribution. Half the emission factors are between 9.1×10^{13} and 1.87×10^{14} #/km/veh for PN and between 1.1×10^{7} and 3.8×10^{7} ng/km/veh for BC. In addition, the boxes are comparatively short, suggesting that the PN emission factors have a high level of agreement between them, and the BC emission factors have also a high level of agreement between them. Fig. 7 shows also the average emission factors per vehicle for the measured VOCs and SVOCs. For all these compounds, the median is in the lower half of the box, which assumes an asymmetric distribution toward the low values. Their emission factors varied between 14 and 2.1×10^4 µg/veh/km.

For each compound, the atypical values are presented by black dots. Here, only the atypical values above the upper whisker limit are further analyzed with the corresponding fleet composition. The emission factors above the upper whisker for PN and BC are, respectively, 2.3 to 6.6 times higher and 2.7 to 9.6 times higher compared to the median. The highest emission factor for both corresponds to the same fleet composition with 69% of LDVs diesel and 58% of PCs ≤ Euro 4 (composition 8 in the Fig. 7) for the Wednesday between 8:00 p.m. and 9:00 p.m. The second highest emission factor for BC and the third highest for PN (composition 7 in the Fig. 7) correspond to the Sunday between 8:00 a.m. and 9:00 a.m.

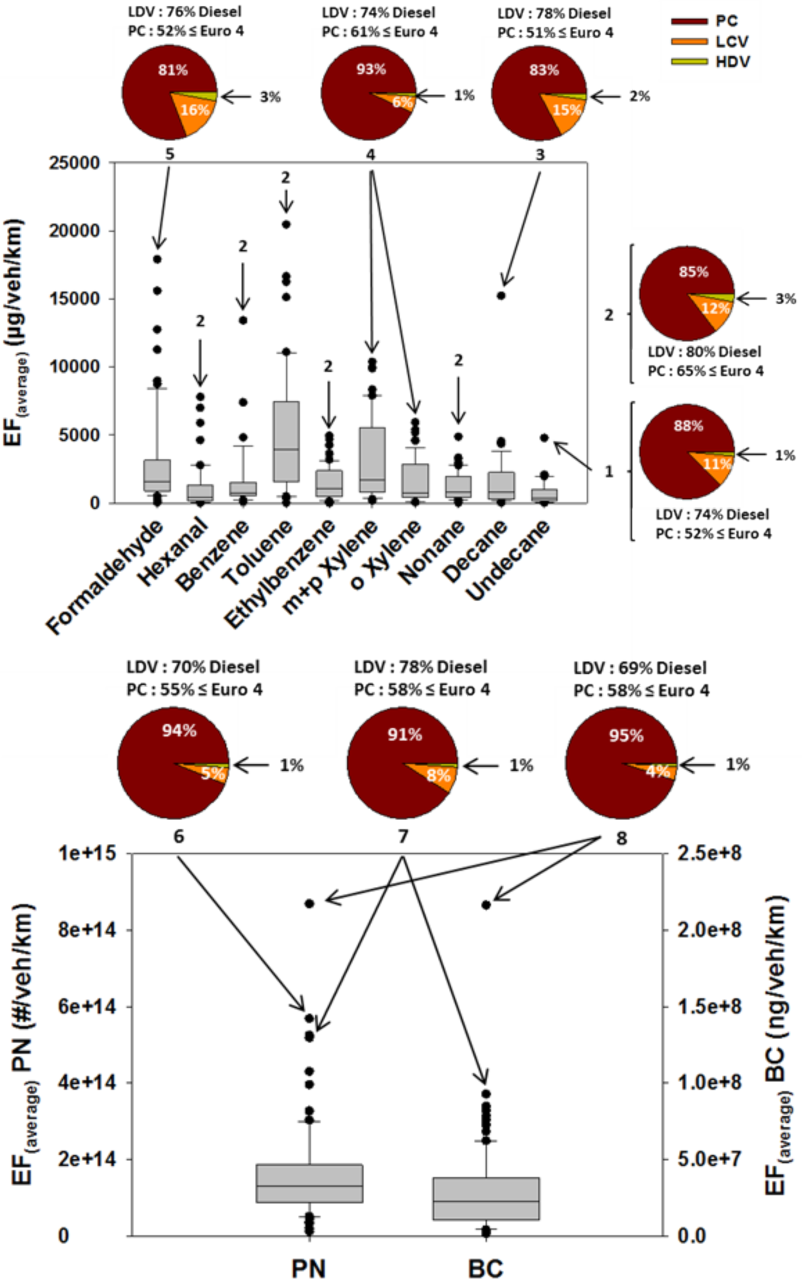

Fig. 7. Box-and-whisker plot of the average vehicle emission factors for PN, BC, carbonyl compounds, BTEX and alkanes. Fleet composition of 1: Monday 2:00 – 3:00 p.m.; 2: Sunday 6:00 - 7:00 a.m.; 3: Wednesday 11:00 - 12:00 a.m.; 4: Sunday 11:00 - 12:00 a.m.; 5: Monday 11:00 - 12:00 a.m.; 6: Monday 8:00 – 9:00 p.m.; 7: Sunday 8:00 - 9:00 a.m.; 8: Wednesday 8:00 - 9:00 p.m.

The emission factors for carbonyl compounds above the upper whisker are 5 to 11 times higher and 6 to 17 times higher than the median for formaldehyde and hexanal, respectively. For the three alkanes, the emission factors above the upper whisker are 3 to 18 times higher compared to their median. The BTEX emission factors above the upper whisker are 1.4 to 8 times higher compared to their median. The highest emission factor for hexanal, benzene, toluene, ethylbenzene and nonane correspond to the same fleet composition (composition 2 in the Fig. 7) for the Sunday between 6:00 a.m. and 7:00 a.m. We remark that, in this time interval, the fleet composition composed relatively high percentage of LDVs diesel (80%), PCs ≤ Euro 4 (65%) and HDVs (3%), which have been showed by the ANOVA analysis to have significant impacts on emission of these compounds (Table 3). For m+p and o xylene compounds, the highest emission factor corresponds to the fleet composition of the Sunday between 11:00 a.m. and 12:00 a.m. (composition 4 in the Fig. 7), with relatively high percentage of PCs ≤ Euro 4. For formaldehyde, the highest emission factor corresponds to the Monday between 11:00 a.m. and 12:00 a.m. (composition 5) with 3% of HDVs.

Fig. 8 shows the comparison of PN, BC, formaldehyde and BTEX emission factors estimated in this study and with those estimated during lab bench measurements with Artemis urban driving cycles that is not exactly the same driving condition but might represent the most similar driving condition (average speed from 8.7 to 31.8 km/h from congestion to fluid situations) comparing to our traffic site (hourly average speed between 16 and 34 km/h). The detail of bench measurement of Euro 1 to Euro 6 gasoline and diesel PCs were presented in our previous works (Rehn 2013; Louis et al., 2016; Martinet et al., 2017). Moreover, the PN and BC emission factors estimated in this study were also compared with other in-situ studies with similar site characteristics (Ketzel et al., 2003; Gratmonev et al., 2004; Rose et al., 2005; Imhof et al., 2005; Jones et al. 2006; Westerdahl et al., 2009; Krecl et al., 2018).

The emission factors estimated by other in-situ studies are between 5.8×10^{13} and 9.3×10^{14} #/veh/km for PN; and between 1.7×10^{7} and 3.5×10^{7} ng/veh/km for BC, which are in the same range as those estimated in this paper (Fig. 8), even fleet compositions are different in different countries and different years.

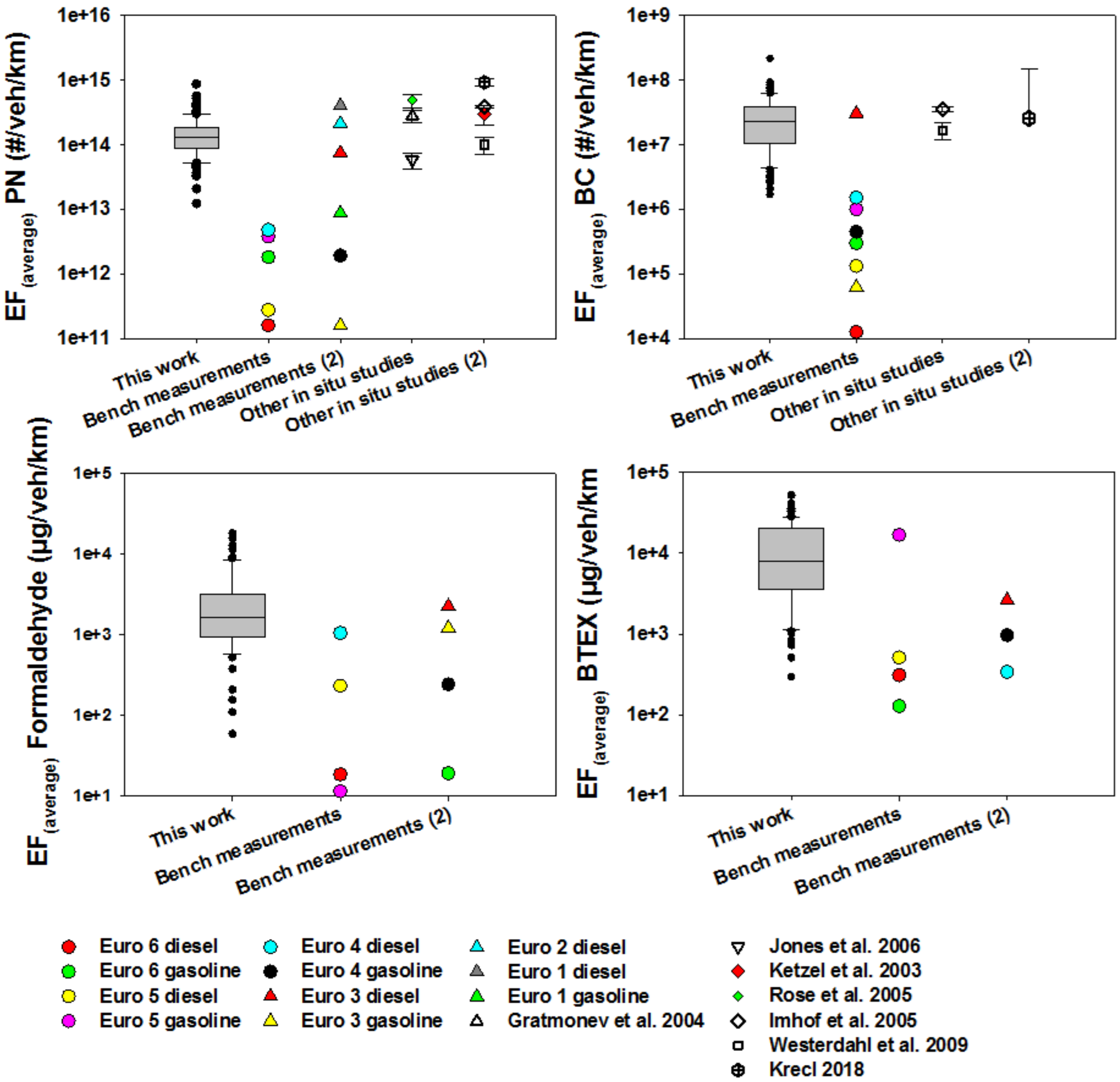

Fig. 8. Comparison of PN, BC, formaldehyde and BTEX emission factors per vehicle in this work with emission factors estimated by bench measurements (Rehn 2013; Louis et al., 2016; Martinet et al., 2017) or by other in-situ studies (Ketzel et al., 2003; Gratmonev et al., 2004; Rose et al., 2005; Imhof et al., 2005; Jones et al., 2006; Westerdahl et al., 2009; Krecl et al., 2018).

The PCs emission factors in this work are also compared to the bench measurement (previous work). The bench emission factors of PN and BC obtained from diesel vehicles without particle filter and gasoline vehicles with direct injection system are in the same range as the box plot. The impact of gasoline with direct injection system and Euro 4 diesel vehicles on PN and BC emission has been observed with ANOVA analysis. However, for the pre-Euro to Euro 3 old diesel vehicles, even they induce a high BC and PN emission with individual vehicle measurement, their impact on BC and PN emission could be considered either as significant but with a great uncertainty or not significant because they are omnipresent in each one-hour period with very closed percentage in the fleet (10 to 20%). These in-situ emission factors are also comparable with old HDVs without particle filter (PN emission factors between 5×10^{13} and 2×10^{14} #/veh/km) (Giechaskiel et al., 2012), contrary to recent HDVs equipped with DPF that induce lower PN emissions (between 5×10^{10} and 2×10^{12} #/veh/km) (Giechaskiel et al., 2018). For fomaldehyde, the emission factors of Euro 3 and Euro 4 diesel and gasoline vehicles are in the same range as the emission factors estimated in this work. More recent Euro 5 and Euro 6 PCs seem to contribute less emission. For BTEX, the Euro 5 gasoline DI vehicle is located above the box that might indicate its high impact on BTEX emission. However, we want to attract special attention here, because only one Euro 5 gasoline DI vehicle data from bench has been provided (Louis et al., 2016). More vehicles for each category should be tested on the chassis bench under similar experimental conditions to confirm this observation.

4. Conclusion

This paper aimed to estimate the emission factors of PN, BC, and several aliphatic, aromatic and carbonyl compounds with a real fleet present on the measurement site. The fleet composition characterized during the measurement campaign was comparable with the 2017 French fleet. A total of 140,076 vehicles were counted for seven days with about 21,000 to 23,000 vehicles in the weekdays and 10,000 to 18,000 in the weekend.

The highest emissions were measured during the morning between 7:00 a.m. - 10:00 a.m. and during the end of afternoon between 5:00 p.m. - 8:00 p.m. for the weekdays, and between 10:00 a.m. - 1:00 p.m. and between 5:00 p.m. - 8:00 p.m. for the Saturday. These periods correspond to traffic peaks. The higher emission measured in the morning can be explained by vehicle cold start, which emits large amounts of pollutants such as BC, PN, BTEX, and carbonyl compounds. PN and BC emission factors show a good correlation, except the Tuesday afternoon between 2:00 p.m. - 8:00 p.m. The emission factor values for BC decreased while those for PN increased sharply. This could imply that the particle number emissions, especially the PN size between [0-50] nm, measured during this afternoon were not due to road traffic.

The impacts of the fleet composition on the pollutant emissions were studied by ANOVA analyses. These analyses show the positive impact of the higher percentage of gasoline PCs between Euro 4 and Euro 6 and Euro 4 diesel PCs on the BC emissions and the higher percentage of diesel and gasoline PCs between pre-Euro and Euro 3 on toluene, ethylbenzene, m+p-xylene and o-xylene emissions. The higher percentage of Euro 4 diesel PCs induces higher emission of ethylbenzene, m+p-xylene and o-xylene. And the number of HDVs present in the weekdays does not induce a significant impact on measured pollutant emissions since 80% of HDVs in the fleet is recent HDVs (≥ Euro 4).

The emission factors per vehicle were studied with boxplots and compared to other emission factors calculated in previous in-situ studies and with bench measurement. PN and BC emission factors assume a symmetric distribution contrary to the BTEX, alkanes and carbonyl compounds emission factors. For PN and BC, the highest emission factors correspond to the same fleet composition with high percentage of diesel PCs. The highest emission factors for hexanal, benzene, toluene, ethylbenzene and nonane correspond to a fleet composition with high percentage of diesel PCs (85%) and old PCs (65%), and for m+p and o Xylene, they correspond to high percentage of old PCs (61%). These results have also been observed by the ANOVA analyses for toluene, ethylbenzene, m+p-xylene and o-xylene. The PN and BC emission factors estimated by other in-situ studies are in the same range as the emission factors estimated in this work. For the chassis bench comparison, the PN and BC emission factors estimated in this work are in the same range as those measured for diesels PCs without particle filter, gasoline PCs with direct injection system and old HDVs. For BTEX, bench emission factors are in the same range as in-situ emission factors estimated in this work.

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Appendices

Table A.1

Analytical methods for BTEX, carbonyl compounds, and PAH samples in the gas and particulate phases with quantification limit and uncertainty

Compound family	Cartridge type	Analytical technique	Standardized method	Quantification limit	Analytical uncertainty
BTEX					
Benzene	Tenax	ATD-GC/MS*	ISO 16000-6	10 ng/cartridge	20%
Toluene				1 ng/cartridge	
Ethylbenzene					
m-p,Xylene					
o-Xylene					
Alkanes					
Nonane	Tenax	ATD-GC/MS*	ISO 16000-6	1 ng/cartridge	20%
Decane					
Undecane					
Cyclopropane, Ethyl					
Cyclohexane, Ethyl					
Carbonyl					
Formaldehyde	DNPH	HPLC/UV$^{\#}$	ISO 16000-3	30 ng/cartridge	20%
Acetaldehyde					
Acetone					
Acrolein					
Propionaldehyde					
Crotonaldehyde					
Methacrolein					
Butanal					

Benzaldehyde					
Pentanal					
Hexanal					

* ATD-GC-MS: Automated Thermal Desorption – Gas Chromatograph – Mass Spectrometer

HPCL/UV: High Performance Liquid Chromatography/Ultra Violet Detector

Table A.2

Vehicle emission factors per hour for PN, BC, carbonyl compounds, BTEX and alkanes.

Pollutants		**Emission factors (Min-Max)**	**Mean**	**Standard deviation**
PN	Weekdays	1.2×10^{13} – 8.7×10^{14}	2×10^{14}	1.9×10^{14}
(#/veh/km)	Weekend	6.1×10^{13} – 5.3×10^{14}	1.7×10^{14}	1×10^{14}
BC	Weekdays	1.7×10^{6} – 2.2×10^{8}	2.4×10^{7}	2.8×10^{7}
(ng/veh/km)	Weekend	9×10^{6} – 9.3×10^{7}	4.7×10^{7}	2.1×10^{7}
Formaldehyde	Weekdays	1.6×10^{-1} – 18	3	3.9
(mg/veh/km)	Weekend	6×10^{-2} – 13	3.2	3.1
Hexanal	Weekdays	1.4×10^{-2} – 2.8	7.1×10^{-1}	8.8×10^{-1}
(mg/veh/km)	Weekend	2.1×10^{-1} – 7.8	1.8	1.8
Benzene	Weekdays	9.1×10^{-2} – 7.4	1.2	1.5
(mg/veh/km)	Weekend	2.3×10^{-1} – 13.4	2.4	3.5
Toluene	Weekdays	3.4×10^{-2} – 11	3	2.6
(mg/veh/km)	Weekend	3.3 – 20.5	9.6	4.7
Ethylbenzene	Weekdays	1.3×10^{-2} – 2.6	8.5×10^{-1}	7×10^{-1}
(mg/veh/km)	Weekend	7×10^{-1} – 5	2.6	1.2

m+p Xylene	Weekdays	1.8×10^{-1} – 6.8	1.6	1.5
(mg/veh/km)	Weekend	1.4 – 10.4	6.4	2.8
o Xylene	Weekdays	7×10^{-2} – 2.9	7.1×10^{-1}	6.4×10^{-1}
(mg/veh/km)	Weekend	6.1×10^{-1} – 5.9	3.2	1.6
Nonane	Weekdays	3.5×10^{-2} – 3.3	8×10^{-1}	7.5×10^{-1}
(mg/veh/km)	Weekend	3.3×10^{-1} – 4.9	2	1.1
Decane	Weekdays	1.5×10^{-1} – 15.2	1.5	3.1
(mg/veh/km)	Weekend	4.1×10^{-2} – 4.5	1.7	1.3
Undecane	Weekdays	5.5×10^{-2} – 4.8	5.3×10^{-1}	8..9 × 10^{-1}
(mg/veh/km)	Weekend	3.5×10^{-2} – 2.1	1.1	7×10^{-1}

Table A.3

Example of COPERT input data for calculating NO_X emission factors

Category	Fuel	Segment	Euro Standard	Stock [n]	Mean Activity [km/year]	Urban Speed [km/h]
Passenger Cars	Gasoline	Small	PRE ECE	0	0	25
Passenger Cars	Gasoline	Small	Euro 1	1	4077	25
Passenger Cars	Gasoline	Small	Euro 2	2	5055	25
Passenger Cars	Gasoline	Small	Euro 3	1	6820	25
Passenger Cars	Gasoline	Small	Euro 4	2	8508	25
Passenger Cars	Gasoline	Small	Euro 5	1	11007	25
Passenger Cars	Gasoline	Small	Euro 6	5	13309	25
Passenger Cars	Gasoline	Medium	PRE ECE	0	0	25
Passenger Cars	Gasoline	Medium	Euro 1	2	5398	25
Passenger Cars	Gasoline	Medium	Euro 2	4	6672	25
Passenger Cars	Gasoline	Medium	Euro 3	5	8995	25
Passenger Cars	Gasoline	Medium	Euro 4	0	11319	25

Passenger Cars	Gasoline	Medium	Euro 5	2	14555	25
Passenger Cars	Gasoline	Medium	Euro 6	3	17592	25
Passenger Cars	Gasoline	Large-SUV-Executive	PRE ECE	0	0	25
Passenger Cars	Gasoline	Large-SUV-Executive	Euro 1	0	5610	25
Passenger Cars	Gasoline	Large-SUV-Executive	Euro 2	0	7069	25
Passenger Cars	Gasoline	Large-SUV-Executive	Euro 3	0	9523	25
Passenger Cars	Gasoline	Large-SUV-Executive	Euro 4	1	12000	25
Passenger Cars	Gasoline	Large-SUV-Executive	Euro 5	0	15454	25
Passenger Cars	Gasoline	Large-SUV-Executive	Euro 6	3	18619	25
Passenger Cars	Diesel	Mini	Euro 4	10	15608	25
Passenger Cars	Diesel	Mini	Euro 5	5	17572	25
Passenger Cars	Diesel	Mini	Euro 6	5	19	25
Passenger Cars	Diesel	Small	Euro 1	1	10957	25
Passenger Cars	Diesel	Small	Euro 2	3	12152	25
Passenger Cars	Diesel	Small	Euro 3	7	13560	25
Passenger Cars	Diesel	Small	Euro 4	16	15236	25
Passenger Cars	Diesel	Small	Euro 5	12	17146	25
Passenger Cars	Diesel	Small	Euro 6	17	18122	25
Passenger Cars	Diesel	Large-SUV-Executive	Euro 1	0	11376	25
Passenger Cars	Diesel	Large-SUV-Executive	Euro 2	1	12489	25
Passenger Cars	Diesel	Large-SUV-Executive	Euro 3	2	14286	25
Passenger Cars	Diesel	Large-SUV-Executive	Euro 4	2	15807	25
Passenger Cars	Diesel	Large-SUV-Executive	Euro 5	2	17770	25
Passenger Cars	Diesel	Large-SUV-Executive	Euro 6	2	18788	25
Light Commercial Vehicles	Diesel	N1-II	Conventional	0	2313	25
Light Commercial Vehicles	Diesel	N1-II	Euro 1	0	5012	25
Light Commercial Vehicles	Diesel	N1-II	Euro 2	0	7490	25
Light Commercial Vehicles	Diesel	N1-II	Euro 3	2	10979	25
Light Commercial Vehicles	Diesel	N1-II	Euro 4	9	15840	25
Light Commercial Vehicles	Diesel	N1-II	Euro 5	13	21249	25
Light Commercial Vehicles	Diesel	N1-II	Euro 6	23	25636	25
Heavy Duty Trucks	Diesel	Rigid <=7,5 t	Euro I	0	846	25
Heavy Duty Trucks	Diesel	Rigid <=7,5 t	Euro II	0	4475	25

Heavy Duty Trucks	Diesel	Rigid <=7,5 t	Euro III	0	11207	25
Heavy Duty Trucks	Diesel	Rigid <=7,5 t	Euro IV	0	19789	25
Heavy Duty Trucks	Diesel	Rigid <=7,5 t	Euro V	0	32935	25
Heavy Duty Trucks	Diesel	Rigid <=7,5 t	Euro VI	0	50409	25
Heavy Duty Trucks	Diesel	Rigid 7,5 - 12 t	Euro I	0	903	25
Heavy Duty Trucks	Diesel	Rigid 7,5 - 12 t	Euro II	0	4404	25
Heavy Duty Trucks	Diesel	Rigid 7,5 - 12 t	Euro III	0	11191	25
Heavy Duty Trucks	Diesel	Rigid 7,5 - 12 t	Euro IV	1	19733	25
Heavy Duty Trucks	Diesel	Rigid 7,5 - 12 t	Euro V	1	33797	25
Heavy Duty Trucks	Diesel	Rigid 7,5 - 12 t	Euro VI	0	53438	25
Heavy Duty Trucks	Diesel	Rigid 14 - 20 t	Euro I	0	952	25
Heavy Duty Trucks	Diesel	Rigid 14 - 20 t	Euro II	0	4598	25
Heavy Duty Trucks	Diesel	Rigid 14 - 20 t	Euro III	0	11195	25
Heavy Duty Trucks	Diesel	Rigid 14 - 20 t	Euro IV	0	19669	25
Heavy Duty Trucks	Diesel	Rigid 14 - 20 t	Euro V	0	33306	25
Heavy Duty Trucks	Diesel	Rigid 14 - 20 t	Euro VI	1	50609	25
Heavy Duty Trucks	Diesel	Rigid 20 - 26 t	Euro I	0	983	25
Heavy Duty Trucks	Diesel	Rigid 20 - 26 t	Euro II	0	4800	25
Heavy Duty Trucks	Diesel	Rigid 20 - 26 t	Euro III	0	11470	25
Heavy Duty Trucks	Diesel	Rigid 20 - 26 t	Euro IV	0	19795	25
Heavy Duty Trucks	Diesel	Rigid 20 - 26 t	Euro V	0	33762	25
Heavy Duty Trucks	Diesel	Rigid 20 - 26 t	Euro VI	0	50832	25
Heavy Duty Trucks	Diesel	Rigid 26 - 28 t	Conventional	0	0	25
Heavy Duty Trucks	Diesel	Rigid 26 - 28 t	Euro I	0	995	25
Heavy Duty Trucks	Diesel	Rigid 26 - 28 t	Euro II	0	4999	25
Heavy Duty Trucks	Diesel	Rigid 26 - 28 t	Euro III	0	11940	25
Heavy Duty Trucks	Diesel	Rigid 26 - 28 t	Euro IV	0	19884	25
Heavy Duty Trucks	Diesel	Rigid 26 - 28 t	Euro V	2	34084	25
Heavy Duty Trucks	Diesel	Rigid 26 - 28 t	Euro VI	0	50947	25
Buses	Diesel	Urban Buses Midi <=15 t	Euro IV	0	27800	25
Buses	Diesel	Urban Buses Midi <=15 t	Euro V	0	38074	25
Buses	Diesel	Urban Buses Midi <=15 t	Euro VI	0	45582	25
Buses	Diesel	Urban Buses Articulated >18 t	Conventional	0	2000	25
Buses	Diesel	Urban Buses Articulated >18 t	Euro I	0	5823	25

Buses	Diesel	Urban Buses Articulated >18 t	Euro II	0	11357	25
Buses	Diesel	Urban Buses Articulated >18 t	Euro III	0	19120	25
Buses	Diesel	Urban Buses Articulated >18 t	Euro IV	1	27763	25
Buses	Diesel	Urban Buses Articulated >18 t	Euro V	1	37992	25
Buses	Diesel	Urban Buses Articulated >18 t	Euro VI	0	45633	25

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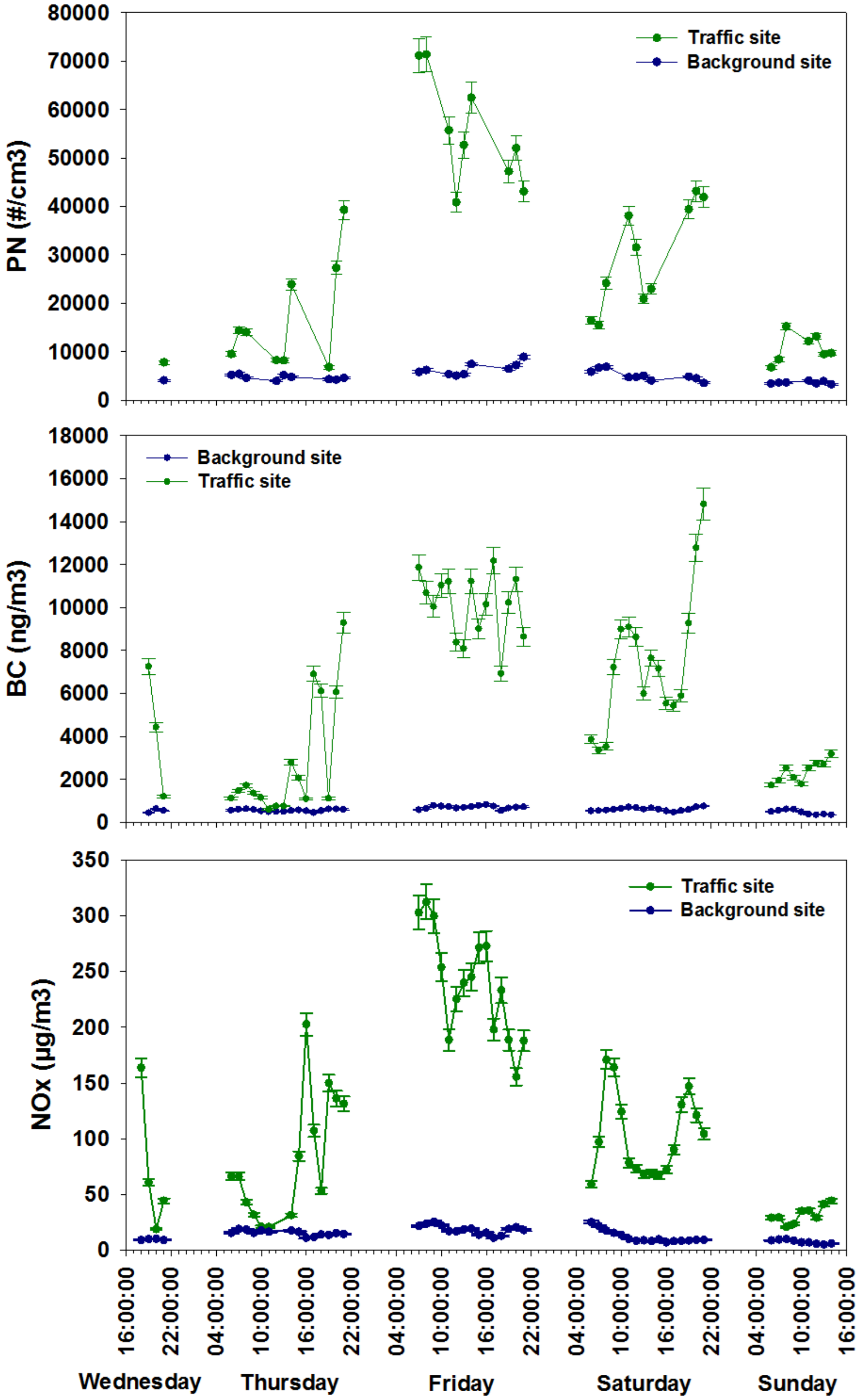

Fig. A.1. Time profiles of PN, BC and NO$_X$ concentrations on the traffic site and the background site From Wednesday to Sunday.

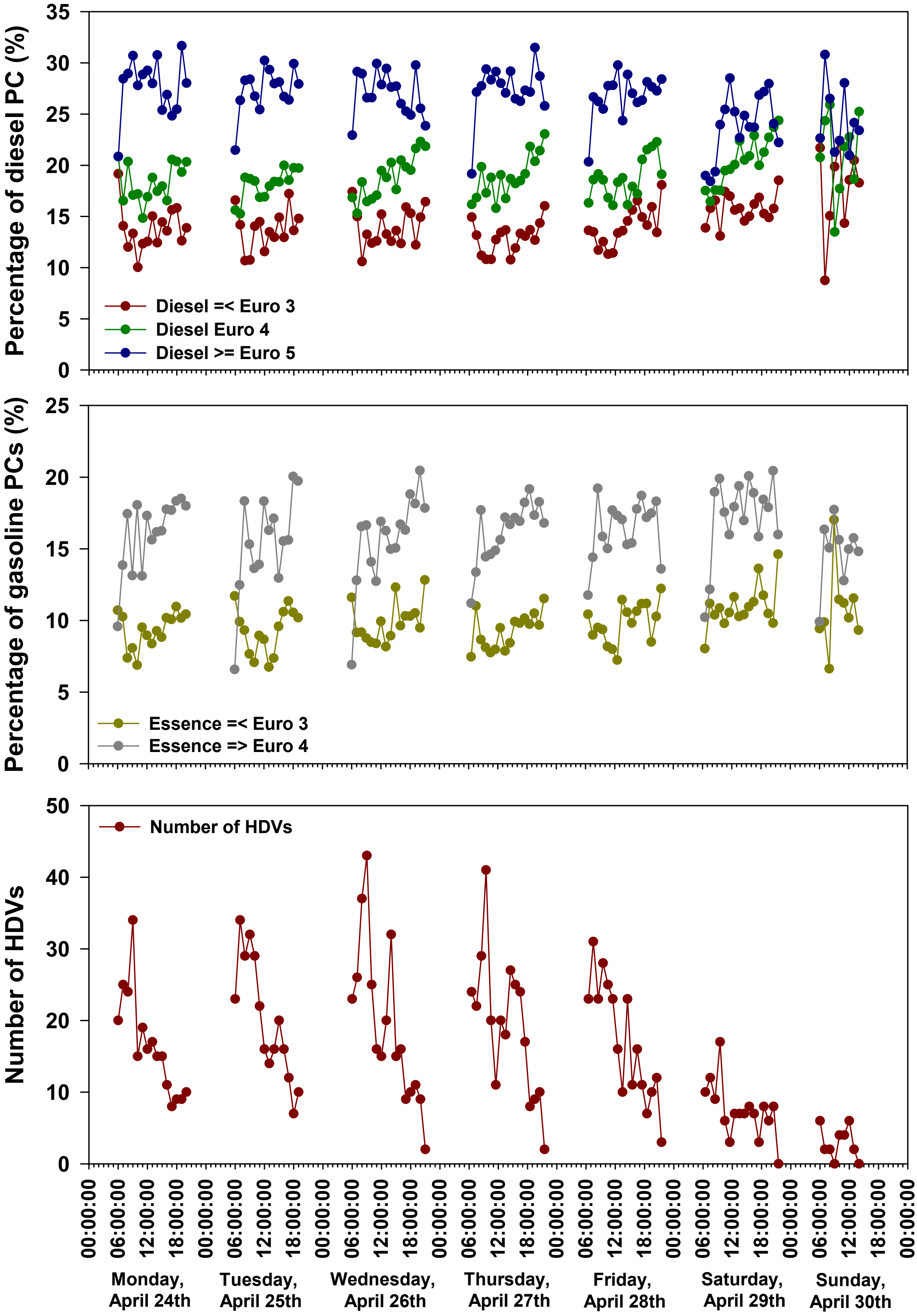

Fig. A.2. Time profiles of the percentages of the diesel PCs, the percentages of gasoline PCs and the numbers of HDVs on the measurement week.

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