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VOC emissions from Euro 6 vehicles

Christian Ferrarese[1,2], Jacopo Franzetti[1,2], Tommaso Selleri[1,3] and Ricardo Suarez-Bertoa[1*]

Abstract

Background Air pollution is a major health concern in worldwide. Non-methane volatile organic compounds (NMVOCs) are precursors of secondary air pollutants, with road transport being responsible of ~ 90% for the EU-27's NMVOCs transport emissions in 2021. A series of VOC emissions from 17 modern gasoline, Diesel and Plug-in hybrid (PHEV) vehicles were investigated under various driving conditions and temperatures. All tested vehicles meet the latest European emission standard (Euro 6d and Euro 6d-TEMP). The different VOC species were measured with a Fourier-Transform Infrared Analyzer (FTIR).

Results Diesel vehicles presented the lowest VOC emissions, while PHEVs operating in charge sustaining mode, with a depleted battery, exhibited very similar behavior to conventional gasoline. Among the VOCs, C_5 compounds were the primary contributors to total NMVOCs over WLTC at 23 °C for gasoline and PHEV vehicles. A proportional increase in VOC emissions at colder temperatures, affecting all the studied species, was observed. Significant increases were observed for Aromatics, with an important contribution of $< C_5$ as well. On the other hand, VOC emissions from Diesel vehicles were consistently low and little affected by temperature, except for Aldehydes in tests at − 7 °C. VOC emissions primarily occurred during cold starts, with urban cycle showing higher emission factors due to its shorter distance. VOC emissions remained consistently low during the highway cycle, highlighting a significant reduction in VOC emissions once the after-treatment system (ATS) was warmed up, even under demanding conditions. In Diesel vehicles, total VOCs measured with the FTIR exhibited a slight tendency to exceed Total Hydrocarbons (THC) measured with a Flame Ionization Detector (FID), while for gasoline vehicles and PHEVs, the trend was temperature-dependent.

Conclusions In summary, the study shows that VOC emissions from Diesel vehicles are significantly lower compared to modern gasoline and PHEV vehicles. Moreover, gasoline and PHEV vehicles exhibit similar levels and emission profiles of VOC emissions. Additionally, ambient temperatures and driving conditions have a significant impact on VOC emissions for all the powertrain technologies investigated.

Keywords Volatile organic compounds, Pollutants emissions, Laboratory vehicle testing, Euro6d, Euro6d-TEMP, FTIR, Non-methane volatile organic compounds

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Background

Air pollution significantly impacts the health of the European population [1]. The European Environment Agency reported in their 2022 briefing on Europe's air quality status that, despite reductions in air pollutant emissions, > 95% of the EU urban population was exposed to PM2.5 and O_3 concentrations above the WHO guidelines in 2021 [2]. These compounds are considered secondary air pollutants and non-methane volatile organic compounds (NMVOCs) are one of their precursors [1, 3]. The road transport sector is an important source

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of NMVOCs. EU-27's NMVOC emissions from road transport have decreased by ~ 90% since 1990. Nevertheless, this sector was responsible for ~ 90% of EU-27's NMVOC transport emissions in 2021 [4, 5]. Reductions in NMVOC emissions from the road transport have been achieved following an increase in the stringency on the total hydrocarbon (THC or HCs) limits included in the EU vehicle emission standards (also known as Euro standards), which have led to improvements in engine combustion strategies as well as the introduction of more efficient exhaust after-treatment technologies [6].

During vehicle certification, THC emissions are measured using a Flame Ionization Detector (FID). In FID, the sample is introduced into a hydrogen flame, and a portion of the HCs present is ionized [7, 8]. FID measurement is performed at 191 °C to ensure that most of the hydrocarbons of interest are volatile. Since the sensitivity of the FID depends on each HC component, a response factor is defined that gives the relative sensitivity compared to propane (C_3H_8) as a calibration gas. In this process, the ions obtained generate an electric current that is nearly proportional to the carbon atoms in the sample. For this reason, the HCs concentration is referred to as "THC" and the units are "ppmC". However, vehicles not only emit hydrocarbons but also other VOCs resulting from the partial combustion/oxidation of the fuel or the oxygenated fuel itself, e.g., ethanol [9–12]. The FID has a lower sensitivity for oxygenated molecules. For example, with some of them, such as formaldehyde, it yields a zero response. For this reason, and to overcome the potential underestimation of such emissions, the California Air Resource Board (CARB) and the US EPA have introduced the concept of non-methane organic gases (NMOG), which include Non-Methane Hydrocarbons (NMHCs), alcohols and carbonyls [13]. Nonetheless, FTIR spectroscopy has been proposed as an alternative to FID because all of the species contributing to NMOG are infrared active. In fact, FTIRs are widely used in vehicle emission testing. Their implementation is already prescribed in the UNECE GTR No 15 [14] and EU 582/2011 [15] for the measurement of NH_3, N_2O, ethanol, acetaldehyde or formaldehyde.

Some studies comprehensively characterized NMVOCs emitted by light and heavy-duty vehicles, both in laboratory and on-road, using offline measurements such as gas chromatography [16–19], which do not allow for a detailed time resolution. Others carried out online measurements using PTR-ToF-MS [9, 20], which can result in molecular fragmentation, complicating the differentiation between small alkanes and alkane fragments. It has been shown that speciation of NMVOCs is progressively more difficult for Fourier Transform Infrared Spectroscopy (FTIR) as the number of carbons increases

[21] because: (i) the individual molecular rotation-vibration lines coalesce into broader bandshapes and (ii) they progressively overlap with each other. This will require FTIRs with high resolution performances.

The present study investigates the emissions of a series of VOCs, including: methane (CH_4), hydrocarbons with less than 5 carbon atoms ($< C_5$), pentanes (C_5), hydrocarbons with more than 5 carbon atoms ($> C_5$), aromatics, alcohols and aldehydes from seventeen Euro 6d/Euro 6d-TEMP passenger cars (5 Diesel, 6 conventional gasoline and 6 plug-in hybrid vehicles (PHEVs)) measured using FTIR spectroscopy under a wide variety of driving and ambient conditions. These results provide a good overview of light-duty vehicle NMVOC emissions at the stage when the latest EU emission regulation, EU 2017/1151 [22], was introduced for modern Euro 6d vehicles with different powertrain technologies.

Methods

13 Euro 6d-TEMP and 4 Euro 6d light-duty vehicles were tested at the Vehicle Emission Laboratory (VELA) of the European Commission Joint Research Centre (EC-JRC) Ispra, Italy. Vehicles were selected either through the European Commission's Market Surveillance vehicle selection procedure [23] or through other vehicle selection procedures within other test programmes that attempted to be representative of the European market. The facility has been extensively described in previous publications [24, 25]. The specific facility used includes a climatic test cell with controlled relative humidity and temperature to simulate different ambient conditions. Tests were conducted at different temperatures (from − 30 °C to 50 °C) on a 4 wheel-drive chassis dynamometer, made of two roller benches with a diameter of 48 inches (1.219 m). Tailpipe emissions were collected in a Constant Volume Sampler (CVS, AVL, Austria). A critical Venturi nozzle was used to regulate the flow (CVS flow range: 2–20 m^3 min^{-1}). The exhaust flow rate was determined by subtracting the flow of dilution air introduced into the tunnel, measured with a Venturi system, to the total flow of the dilution tunnel, measured by a critical flow Venturi.

Testing conditions and vehicles characteristics

The selected fleet included: six gasoline vehicles (two Gasoline Direct Injection (GDI) with three-way-catalyst (TWC) and a gasoline particulate filter (GPF) (GV1, GV6) and four Port Fuel Injection (PFI) equipped with a TWC (GV2, GV3, GV4 and GV5); five Diesel vehicles (all equipped with selective catalytic reduction system (SCR), Diesel oxidation catalyst (DOC) and Diesel particulate filter (DPF) (DV1-DV5)); six plug-in hybrid (PHEV) gasoline vehicles (all equipped with TWC and

GPF (HV1-HV6)). The main vehicle characteristics are summarized in Table 1. All vehicles tested were fueled with B7 Diesel or E10 commercial petrol, the chemical composition of which complies with European Directive 2009/30/EC [26] on the environmental specifications of market fuels. Environmental specifications for market fuels are listed in Additional file 1: Table S1.

Tests were performed on the worldwide harmonized light vehicles test cycle (WLTC), which was designed to be representative of real driving conditions as it is based on real world vehicle trips from several countries. The cycle consists of four phases with different duration and speed profiles: low speed (589 s), medium speed (433 s), high speed (455 s) and extra-high speed (323 s) phases [22]. Two vehicles (GV6 and HV6) were also tested on the Transport for London (TFL) driving cycle, which simulates an urban driving cycle characterized by stop and go traffic in congested conditions; the same two vehicles were also tested on a high speed driving cycle, with sharp and frequent acceleration, developed by ADAC (Allgemeiner Deutscher Automobil-Club e.V), known as the Bundesautobahn motorway cycle (BAB). Speed profiles of the performed cycles can be found in Additional file 1: Fig. S1).

Vehicles' emissions were studied across a wide range of temperatures and starting conditions. "Starting condition" refers to the after-treatment system (ATS) temperature at cycle starting point. The "COLD" condition refers

to a test where the ATS temperature at the starting point is that of the climatic cell temperature ($T_{CELL} \pm 3$ °C). The "HOT" condition refers to a test carried out immediately after another test so that the ATS is already warmed up. All PHEVs were tested with a completely depleted battery state of charge (SOC), a mode commonly known as charge sustaining mode, as defined by regulation [22].

The driving cycles, temperatures and starting conditions on which vehicles were tested are summarized in Table 2.

Analytical instrumentation

Gaseous compounds were analyzed from the raw exhaust by a high resolution Fourier-Transform Infrared Analyzer (FTIR – Nicolet Antaris IGS Analyzer-Thermo Electron

Table 2 Overview of the test cycles, temperatures and starting conditions performed on the fleet of vehicles

Cycles	Temperatures	Starting condition	Vehicles
WLTC	− 7 °C, 23 °C, 35 °C, 50 °C	COLD, HOT	DV1-DV5, GV1-GV5, HV1-HV5
TFL	− 30 °C, − 10 °C, − 7 °C, 5 °C, 23 °C, 50 °C	COLD	GV6, HV6
BAB	− 30 °C, − 10 °C, − 7 °C, 5 °C, 23 °C, 50 °C	HOT	GV6, HV6

Table 1 Main characteristics of the fleet of vehicles tested

Name	Euro Standard	Powertrain	Engine type	After-treatment	Engine displacement (cm^3)	ICE Engine power (kW)	Odometer (km)
DV1	Euro 6d-TEMP	ICE Diesel	CI	DPF + SCR + DOC	1461	70	4293
DV2	Euro 6d-TEMP	ICE Diesel	CI	DPF + SCR + DOC	2287	115	16,335
DV3	Euro 6d-TEMP	ICE Diesel	CI	DPF + SCR + DOC	1597	118	3102
DV4	Euro 6d-TEMP	ICE Diesel	CI	DPF + SCR + DOC	1499	96	9680
DV5	Euro 6d-TEMP	ICE Diesel	CI	DPF + SCR + DOC	1968	110	3480
GV1	Euro 6d-TEMP	ICE Gasoline	SI GDI	TWC + GPF	1499	100	9666
GV2	Euro 6d	ICE Gasoline	SI PFI	TWC	1199	61	3263
GV3	Euro 6d-TEMP	ICE Gasoline	SI PFI	TWC	1199	61	7578
GV4	Euro 6d-TEMP	ICE Gasoline	SI PFI	TWC	1248	55	628
GV5	Euro 6d-TEMP	ICE Gasoline	SI PFI	TWC	1498	110	46,257
GV6	Euro 6d-TEMP	ICE Gasoline	SI GDI	TWC + GPF	1998	135	23,373
HV1	Euro 6d	Hybrid Gasoline	SI GDI	TWC + GPF	1598	133	3003
HV2	Euro 6d-TEMP	Hybrid Gasoline	SI GDI	TWC + GPF	1580	77	1472
HV3	Euro 6d	Hybrid Gasoline	SI GDI	TWC + GPF	1997	221	2485
HV4	Euro 6d-TEMP	Hybrid Gasoline	SI GDI	TWC + GPF	1998	90	6400
HV5	Euro 6d-TEMP	Hybrid Gasoline	SI GDI	TWC + GPF	1499	100	4201
HV6	Euro 6d	Hybrid Gasoline	SI GDI	TWC + GPF	1598	133	8293

ICE: internal combustion engine; CI: compression ignition; SI GDI: spark ignition gasoline direct injection; SI PFI: spark ignition port fuel injection; DPF: diesel particulate filter; SCR: selective catalytic reduction; DOC: diesel oxidation catalyst, TWC: three-way-catalyst, GPF: gasoline particulate filter

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Scientific Instruments LLC, Madison, WI, USA). The FTIR spectrometer was composed of a Michelson interferometer (spectral resolution: 0.5 cm^{-1}, spectral range: 600–4000 cm^{-1}), a multipath gas cell with 2 m of optical path with a working pressure of 860 hPa, a down-stream sampling pump (6.5 l min^{-1} flowrate) and a liquid nitrogen cooled mercury cadmium telluride detector. The system had an acquisition frequency of 1 Hz. The instrument was connected to the vehicle tailpipe by a polytetrafluoroethylene (PTFE) heated sampling line (191 °C). For more information on the FTIR methodology see ([27] and references there-in, [28]).

Among the different compounds measured in this work we focused on the following VOCs: CH_4, C_5, Aromatics, Alcohols (methanol CH_4O and ethanol C_2H_6O), Aldehydes (formaldehyde CH_2O or HCHO and acetaldehyde C_2H_4O) and hydrocarbons with less than 5 carbon atoms, grouped as $< C_5$. $< C_5$ group comprises: ethane C_2H_6, propane C_3H_8, ethene C_2H_4, propene C_3H_6, butadiene C_4H_6 and ethyne C_2H_2. The hydrocarbon with the highest number of carbon atoms that the instrument was able to measure was octane C_8H_{18}. Each component was analyzed in real-time obtaining a concentration (ppmV). To smooth the emission profile of each compound, all concentrations below the limit of detection were set to zero.

The exhaust flow rate was obtained as the difference between the total flow of the dilution tunnel, measured by a critical flow Venturi, and the flow of dilution air introduced into the tunnel, determined with a Venturi system. The emission rates (mg s^{-1}) were derived from the exhaust flow rate (m^3 s^{-1}) and the concentration (ppmV). The mass emissions of each individual component were added up to obtain the total VOC emission value over a given driving cycle. Emission factors (mg km^{-1}) were derived from the mass emissions of each component and from the total driving distance of the test. THC were measured through a heated (191 °C) Flame Ionization Detector (FID) to compare the mass values obtained with two different analytical techniques. The FID used was embedded in an AMA i60 bench (AVL, Graz, Austria) designed for regulated emission measurement. Detection limits of the different compounds measured with FTIR and THC measured with FID are listed in Additional file 1: Table S2.

Aggregated data were presented by means of descriptive statistic tools namely boxplots. Boxes represent the InterQuartile Range (IQR), that extended from the first quartile (Q1) to the third quartile (Q3) of the data distribution. The horizontal line represents the median (Q2). Whiskers extended no more than 1.5 times the IQR from the edges of the box, ending at the furthest data point within the interval. Outliers are presented as separate dots representing points outside boundaries identified by

lower and upper whiskers. It should be noted that outliers were actual emission factors.

Results and discussion

VOC overall emissions over regulated laboratory cycles

Figure 1 illustrates the aggregate results of tests conducted on WLTC at 23 °C for five Diesel (DV1-DV5), five conventional gasoline (GV1-GV5), and five PHEV (HV1-HV5) vehicles. Vehicles were tested between 2 and 5 times over the WLTC. The tests were performed in COLD starting conditions and emission factors for different VOCs were obtained as described in section "Analytical instrumentation". Overall emissions for all NMVOC groups and CH_4 were generally low, with a few outliers, representing actual emission factors, exceeding 10 mg km^{-1}. Despite the variability of the parameters between the different vehicles in the different categories, as can be seen from Table 1, the variability of the emission factors of the different compounds is small.

The emissions of NMVOCs from Diesel vehicles were very low, typically less than 3 mg km^{-1} with negligible contributions from C_5 and Alcohols, which is likely due to C_5 and Alcohols absence in the chemical composition of Diesel fuel (see additional details on fuel composition in Additional file 1: Table S1). The primary contributor to total VOC emissions for Diesel vehicles was CH_4, with a median of 1.9 mg km^{-1} and low variability (1.2–5.3 mg km^{-1}). The medians of $< C_5$, Aromatics, and Aldehydes were very low, at 0.9 mg km^{-1}, 0.1 mg km^{-1}, and 0.1 mg km^{-1} respectively, with small variability (0.2–1.8 mg km^{-1}, 0–0.2 mg km^{-1}, and 0–0.7 mg km^{-1}, respectively). VOC emissions in Diesel vehicles are expected to be low also due to the efficiency of the DOC, which is related to the air–fuel ratio, light-off temperature and light-off time, space velocity, and flow restriction characteristics [29].

Emissions of NMVOCs from PHEVs and gasoline vehicles showed no significant differences between the two technologies. This is in line with previous research, which has shown that there are no substantial differences in hydrocarbon emissions when using a PHEV in charge sustaining mode compared to an ICE gasoline vehicle. Furthermore, it has been shown that the NMHC emissions of a PHEV tested in charge sustaining mode are comparable to those obtained by testing the vehicle with a fully charged battery [30].

Emissions of $< C_5$ were minimal with medians of 1.1 mg km^{-1} for PHEVs and 1 mg km^{-1} for gasoline vehicles. The variability for $< C_5$ emissions was low for PHEVs (0.4–2.3 mg km^{-1}) and higher for gasoline vehicles (0.4–3.8 mg km^{-1}). It can be observed that the emissions of all three technologies, Diesel, gasoline, and PHEVs, were very similar. The medians differed by less than 0.2 mg km^{-1} and the variability was similar, except for gasoline
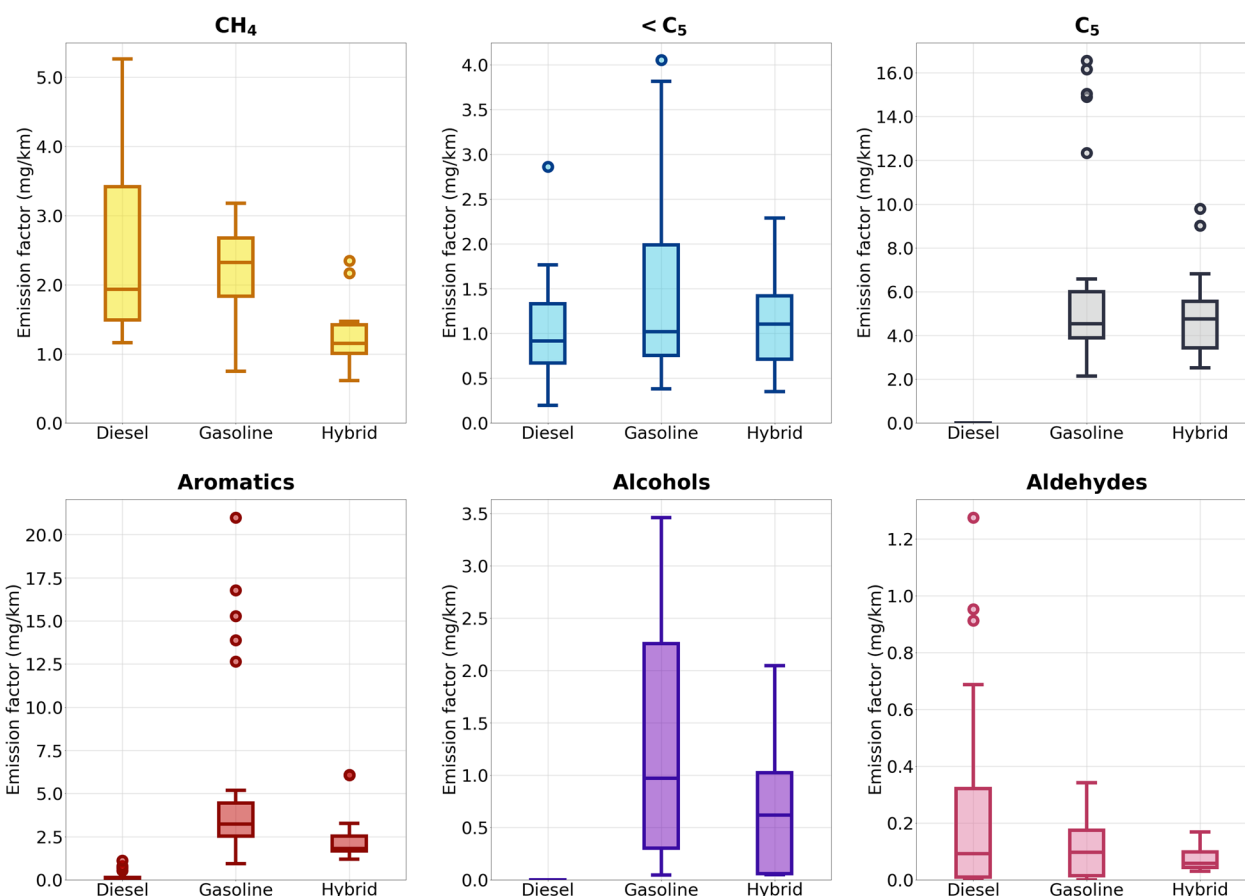
Fig. 1 Overall VOC emissions of 5 Diesel (DV1-DV5), 5 conventional gasoline (GV1-GV5) and 5 plug-in hybrid gasoline (HV1-HV5) vehicles over regulated laboratory cycles WLTC at 23 °C

vehicles. This can be attributed to a single gasoline vehicle (GV4) whose emissions during WLTCs at 23 °C were consistently higher than others, ranging from 3.3 to 4 mg km^{-1}. Unburned fuel is linked to low carbon atom hydrocarbon emissions in gasoline vehicles [20].

For gasoline and PHEV vehicles, C_5 emissions were the primary contributors to NMVOCs, accounting for 30–45% and 45–60%, respectively. As expected, the medians were comparable, at 4.5 mg km^{-1} for gasoline and 4.8 mg km^{-1} for PHEV vehicles driven in charge sustaining mode, and the variability was low (2.1–6.6 mg km^{-1} and 2.5–6.8 mg km^{-1}, respectively). A small number of high emission outliers were recorded for gasoline vehicles, ranging from 12 to 17 mg km^{-1}, all caused by one vehicle (GV4). Some outliers were observed for PHEV vehicles around 9–10 mg km^{-1}, also in this case related to one vehicle (HV4).

Emissions of Aromatics in gasoline vehicles were low with a median of 3.2 mg km^{-1} and small variability (0.9–5.2 mg km^{-1}). For C_5 emissions, gasoline vehicles presented some high emission outliers ranging from 12.5 to

21 mg km^{-1}, all caused by the same vehicle (GV4). The high emission outliers observed in this vehicle for C_5 and Aromatics can be attributed to an extended light-off time of the TWC. Furthermore, the elevated carbon monoxide (CO) emissions, coupled with reduced nitrogen oxide (NOx) emissions during the cold start phase, suggest a potentially rich engine operating condition [31], resulting in increased VOC emissions for this vehicle. Lower Aromatics emissions were recorded for PHEVs, with a median of 1.8 mg km^{-1} and very low variability (1.6–3.4 mg km^{-1}). Alcohols and Aldehydes emissions were very low for both gasoline and PHEV vehicles. Medians of Alcohols emissions were 1 and 0.6 mg km^{-1} for gasoline and PHEV vehicles, respectively, while Aldehydes emissions for both powertrain technologies were lower than 0.5 mg km^{-1}.

Effect of vehicle starting condition on VOC emissions
NMVOC emissions from vehicles are typically associated with unburned fuel and incomplete combustion products, and are primarily released during the initial

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stages of the cycle, most of them before the activation of the after-treatment systems (DOC or TWC). As already discussed, NMVOC emissions from Diesel vehicles were low under COLD conditions. Hence, an effect of the starting conditions was not observed for Diesel vehicles. For that reason, the effects of starting conditions on VOC emissions for gasoline and plug-in hybrid electric vehicles (PHEVs) were investigated.

VOC emissions of gasoline vehicles and PHEVs are largely related to the time required for the light-off of the TWC, whose function also includes oxidizing hydrocarbons to carbon dioxide (CO_2) and water (H_2O). Since in the first seconds of the test cycle in HOT starting conditions the average temperature of ATS is higher, thus the

TWC warm-up phase is shorter, leading to very low VOC emissions throughout the test compared to COLD conditions. Additional file 1: Fig. S2 illustrates an example of VOC emission profiles on WLTC at 23 °C as a function of engine coolant temperature for both gasoline and PHEV vehicles.

Figure 2 presents the aggregate results of VOC emissions of gasoline vehicles and PHEVs on WLTC at 23 °C as a function of starting conditions, as defined in section "Analytical instrumentation". COLD refers to a test where the ATS temperature at the starting point is that of the climatic cell temperature (23 °C ± 3 °C), while HOT refers to a test carried out immediately after another test so that the after-treatment system is already warmed up.
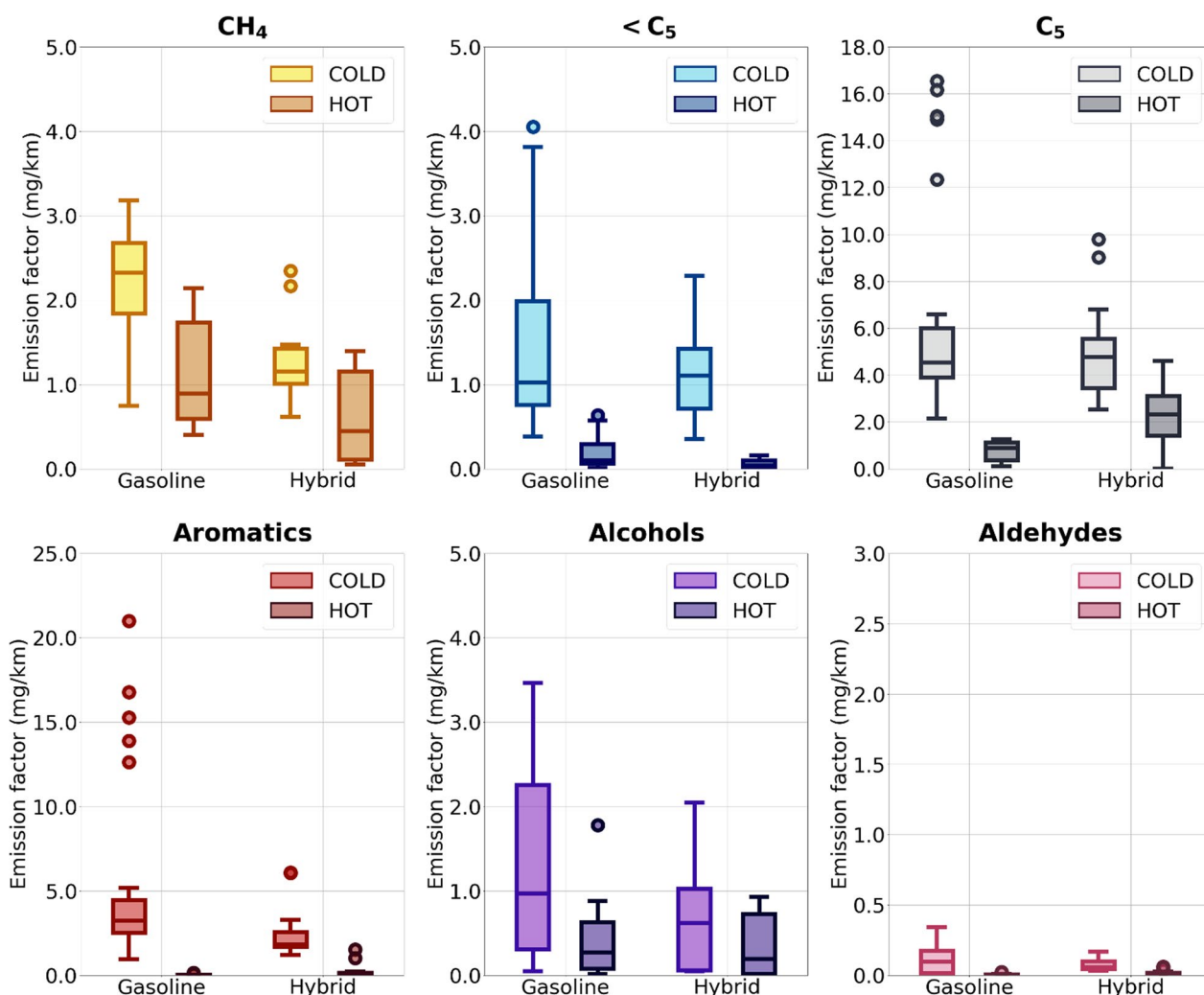

Fig. 2 Overall VOC emissions of gasoline and PHEV vehicles on WLTC at 23 °C as a function of the starting condition. COLD refers to a test where the ATS temperature at the starting point is the same as the climatic cell temperature (23 °C ± 3 °C) and are the same emission results of those reported in Fig. 1, included to ease the comparison. HOT refers to a test carried out immediately after another test so that the after-treatment system is already warmed up

In general, HOT starting conditions significantly lower VOC emissions, with a reduction of approximately 75% for gasoline vehicles and 63% for PHEVs. All NMVOC emissions became negligible or less than 1 mg km^{-1}, except for C_5 in PHEVs, with a median of 2.3 mg/km^{-1} and variability ranging from 0 to 4.6 mg km^{-1}. CH_4 emissions exhibited a lower reduction of approximately 60% for both gasoline vehicles and PHEVs. As demonstrated by previous studies [32], CH_4 shows to be more difficult to oxidize than other hydrocarbons under stoichiometric combustion over the TWC also in modern technologies.

Effect of ambient temperature on VOC emissions
Regulated cycles
Figure 3 displays the results of emissions tests conducted at −7 °C and 23 °C for Diesel, gasoline, and PHEVs on WLTC. Additionally, Diesel vehicles were tested under hot ambient temperatures of 35 °C and 50 °C. The height of the bars represents the average emissions factor of all tests performed on different vehicles at a given ambient temperature, while the error bars indicate the maximum and minimum values resulting from the testing. Detailed emission factors (mg km^{-1}) of all the species at the
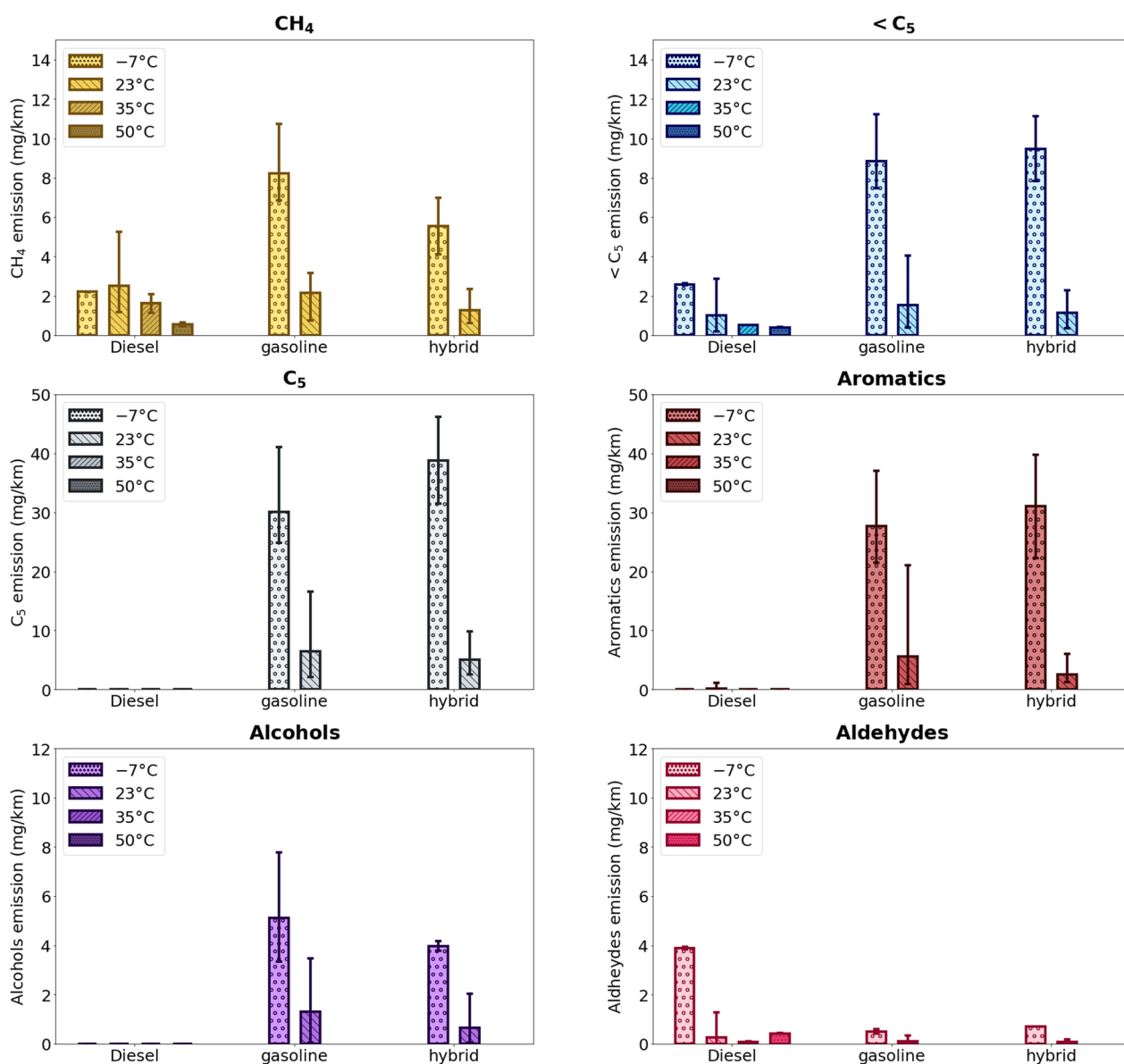
Fig. 3 Emissions of VOCs on WLTC as a function of environmental temperature. The height of the bars represents the average of all tests performed on different vehicles at a given temperature, while error bars indicate the maximum and minimum values resulting from a test

different testing temperatures are reported in Additional file 1: Table S3.

VOC emissions in Diesel vehicles were little affected by temperature except for Aldehydes at -7 °C, increasing up to 4 mg km^{-1}, more than 15 times of those at 23 °C. Among the Aldehydes, formaldehyde (HCHO) was the major contributor. HCHO emissions in Diesel vehicles are linked to oxidation process of unburned fuel on DOC. Since the extent of HCHO emissions depends on the after-treatment efficiency and light-off time, the reason for the increase of aldehydes emission at subzero temperatures may be the worse combustion and lower efficiency with longer light-off times of the ATS [33].

Colder temperature induced a proportional increase in emissions for gasoline and PHEVs, affecting all the studied species. The major contributor to NMVOC emissions at -7 °C was C_5 for both gasoline and PHEV, with emission factors of 30 and 38 mg km^{-1}, respectively. These emission factors represent increases of more than 4 and 7 times compared to those at 23 °C. The most substantial rise in emissions at lower temperatures was observed for Aromatics, with emission factors of 28 and 31 mg km^{-1}, signifying increases of more than 5 and 11 times, respectively, compared to those at 23 °C. VOC emissions increase at lower temperatures is in line with previous literature studies [34, 35] as lower temperatures require an extended warm-up period for after-treatment systems to reach optimal operating conditions. It is important to note that the different polycyclic aromatic compounds (PAHs) in the Aromatic group are associated with different toxic health effects. In addition, due to their low vapour pressures and similar chemical structure to the extended aromatic systems, PAHs tend to adsorb to soot particles and co-released at low temperatures [36]. This may lead to a further underestimation of the total VOCs emitted at low temperatures. Aldehydes emission remained very low (< 1 mg km^{-1}) at -7 °C.

Figure 4 shows the cumulative VOC emissions during the regulated cycles. Darker colors correspond to tests at 23 °C, while lighter colors represent -7 °C tests. Within each group, the solid lines denote the average of the cumulative emissions over regulated cycles, while the bandwidth represents the maximum and minimum cumulative emissions among all the tests within the specific vehicle category and ambient temperature.

The cumulative emissions for all vehicle categories and pollutants considered in this study exhibited an initial step increase in the first few seconds of the test, followed by a relatively constant level throughout the rest of the test. The difference between the -7 °C and 23 °C tests is mainly because in the initial phase of colder tests (in Fig. 4 represented by lighter color), the increase in emissions is greater and lasts longer, resulting in significantly

higher final emissions than in the tests at 23 °C. The observed relatively large variability, as indicated by the bandwidths, could be attributed to the cumulative effect of variability deriving from differences among the five cars within each category, as well as the inherent differences between individual tests.

Interestingly, Alcohols at -7 °C exhibited a different trend, with cumulative emission increasing throughout the entire test. The fact that alcohol may not be catalytically controlled may explain these trends. This behavior was unexpected, when considering previous literature studies [9, 10], where alcohols emissions from petrol vehicles were mainly in the first seconds of the test.

Unregulated cycles

To evaluate the effect of the driving cycle on VOC emissions under a wide range of ambient temperatures, two distinct unregulated test cycles were used to simulate urban and highway driving conditions. The TFL cycle was used to simulate stop-and-go in congested traffic conditions, while the BAB cycle was used to simulate high speed driving with sharp and frequent acceleration to simulate motorway over takes. It is important to note that TFL was performed under "COLD" conditions, while BAB was performed under "HOT" conditions as defined in the "Methods" section "Testing conditions and vehicles characteristics". Due to experimental constraints, focus was directed toward a Euro 6d-TEMP ICE gasoline vehicle (GV6), tested across a wide range of environmental temperatures: -30 °C, -10 °C, -7 °C, 5 °C, 23 °C, and 50 °C. Additionally, a Euro 6d PHEV (HV6) was tested at -7 °C and 23 °C to estimate the effect of hybridization on VOC emissions under urban and highway driving conditions.

Figure 5 displays the results of emission tests conducted at various ambient temperatures during a TFL cycle for a gasoline vehicle (GV6) and a PHEV (HV6). Detailed emission factors (mg km^{-1}) of all the species at the different testing temperatures are reported in Additional file 1: Table S4.

Similar to the WLTC tests, VOC emissions in TFL cycles generally rise as the temperature decreases. It is evident that small temperature variations lead to small differences in VOC emissions for the gasoline vehicle (e.g., -7 °C vs -10 °C and 5 °C vs 23 °C). At 50 °C, emissions exhibit different trends depending on the pollutant. With engine intake air at 50 °C, combustion may be inefficient, leading to an increase in hydrocarbon emissions [37]. CH_4 and $< C_5$ emissions were very similar to those registered at 23 °C, whereas the emissions of C_5, Aromatics and Aldehydes were more than halved. For Aromatics, whose emission factor in TFL at 50 °C (10 mg km^{-1}) was approximately a third of that at 23 °C (26 mg km^{-1}),
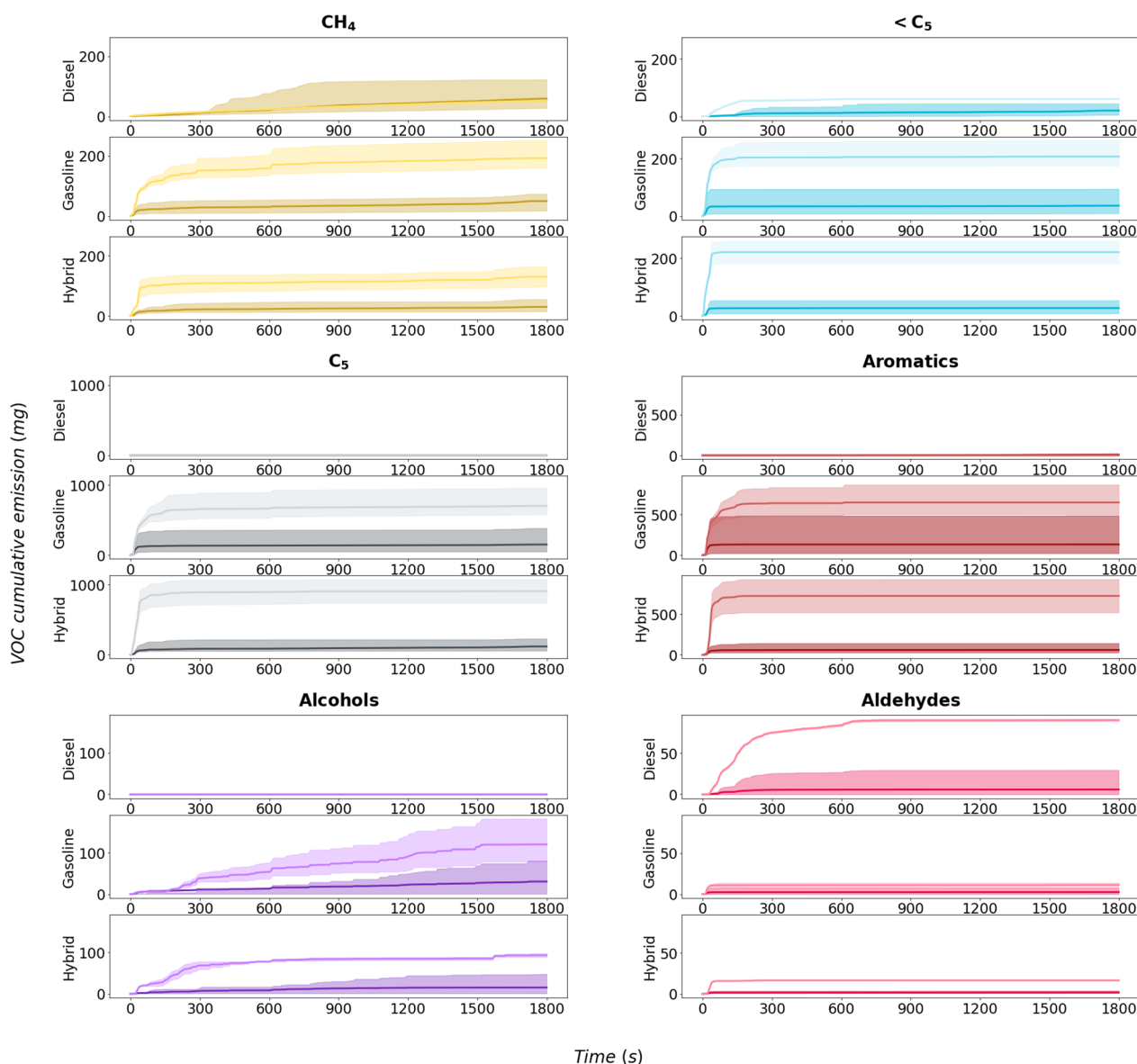
Fig. 4 Cumulative emissions of VOCs on WLTC for Diesel, gasoline and PHEV vehicles at − 7 °C (lighter colors) and 23 °C (darker colors). The solid line represents the average of the cumulative emissions obtained during WLTCs, while the bandwidth represents the maximum and minimum cumulative emissions among all the tests belonging to the specific vehicle category and environmental temperature

appeared to show the greatest reduction in emissions. TFL at − 30 °C proved to be the most severe condition, resulting in emissions more than twice as high as those at − 7 °C and over fivefold higher than those at 23 °C. As in regulated cycles, PHEV showed a similar trend to gasoline vehicles for all pollutants, with emissions increasing by 2–3 times at lower temperatures.

The emission factors (mg km^{-1}) for TFL were much higher compared to those obtained during the regulated WLTC cycle, due to the shorter distance travelled during TFL (approx. 9 km) in comparison to WLTC (approx. 23 km), which resulted in a greater impact of the cold start on the final emission factor. At 23 °C during a TFL, the total VOC emission factor was 85 mg km^{-1}, while the highest value obtained during a WLTC at 23 °C for gasoline vehicles (GV1-GV5) was 45 mg km^{-1}. Nevertheless, when considering the total mass emission (mg), VOC mass emissions amount to 755 mg, which is lower than the maximum VOC mass emissions observed in a WLTC at 23 °C, namely 1033 mg.

For this gasoline vehicle (GV6), Aromatics and C$_5$ constituted the primary fractions of VOC emissions across
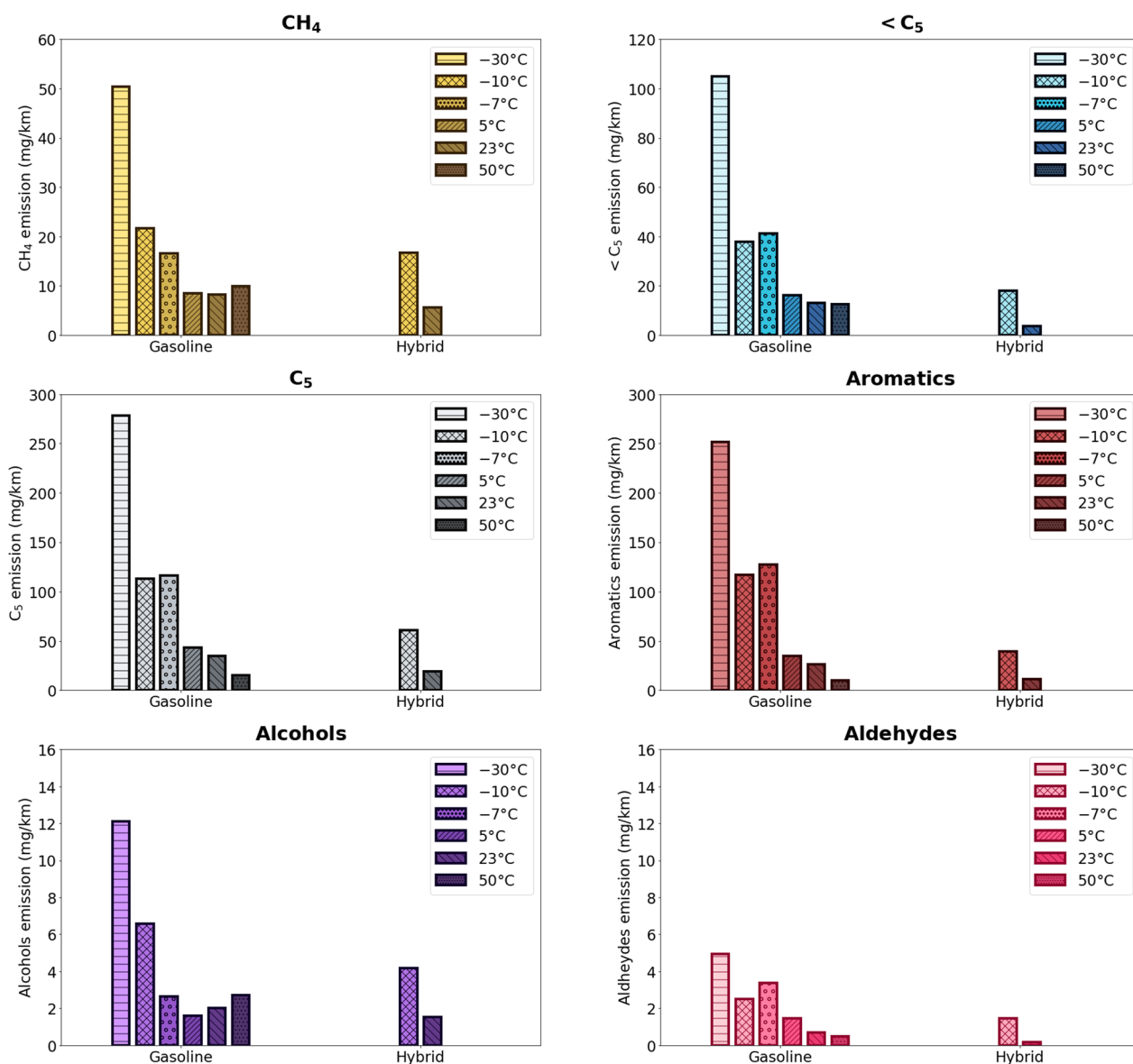
Fig. 5 VOCs emission on TFL tests as a function of environmental temperature

all conditions, jointly contributing to over 70% of its total VOC emissions. Additionally, $< C_5$ made an important contribution (approx. 13–15%). Aldehydes emissions were found to remain low for all testing temperatures, with a slight increase at lower temperatures. Alcohols emissions remained very low as well, but showing a slightly different trend with respect to other VOCs, with an emission factor at $- 10$ °C (7 mg km^{-1}) that was more than double the one at $- 7$ °C (3 mg km^{-1}) which in turn was comparable to the TFL Alcohols emission at 23 °C (2 mg km^{-1}).

Figure 6 displays the results of emission tests conducted at various ambient temperatures during a BAB cycle for a gasoline vehicle (GV6) and a PHEV (HV6). Detailed emission factors (mg km^{-1}) of all the species at the different testing temperatures are reported in Additional file 1: Table S5.

Emissions were generally very low for the gasoline vehicle, with all VOC emissions being below 10 mg km^{-1} at all studied temperatures and negligible for Aromatics, Alcohols and Aldehydes. This confirms the behaviour observed in section "Effect of vehicle starting condition on VOC emissions", where in HOT starting conditions, VOC emissions decrease significantly as the ATS is already warmed up and the cold start effect is almost absent. This effect is independent of
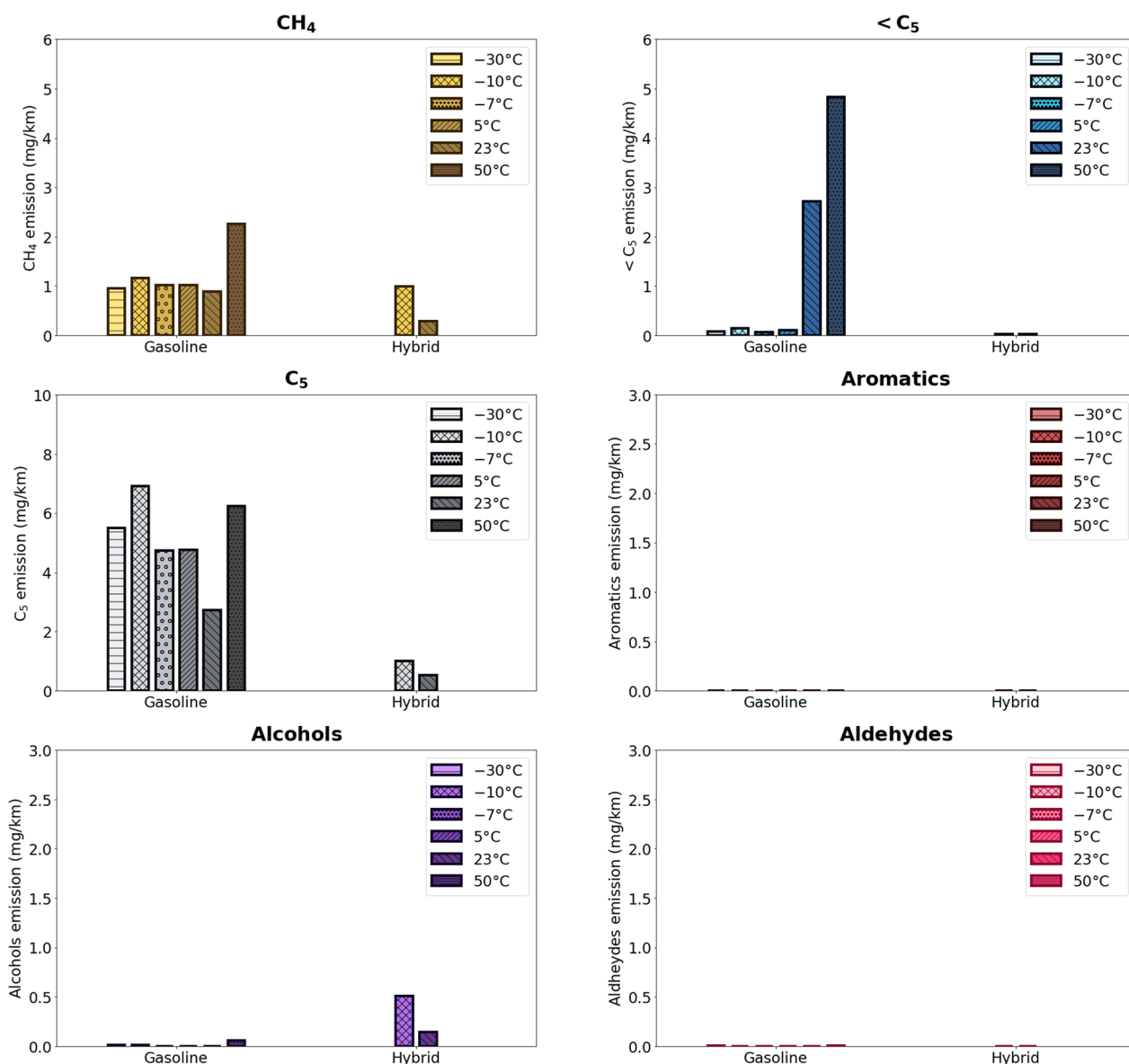
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Fig. 6 VOC emissions on BAB cycles as a function of temperature

the environmental test temperature and although the cycle is very demanding, with significant and frequent high-speed accelerations, VOC emissions remain very low. Interestingly, $< C_5$ emissions were negligible at lower temperatures and increased at higher temperatures, while remaining very low (< 5 mg km^{-1}), with an emission factor at 50 °C that was almost double that at 23 °C.

VOC emissions recorded for this particular PHEV vehicle were very low on a BAB cycle in HOT starting condition at both -7 °C and 23 °C.

THC$_{FID}$ vs VOC$_{FTIR}$

In the process of vehicle certification, THC are quantified using a FID. In the present work, in addition to FTIR measurement, THC emission factors from FID were evaluated (THC$_{FID}$). This allowed to compare them with the total VOC emissions obtained from FTIR (VOC$_{FTIR}$). VOC$_{FTIR}$ emission factors were estimated as the sum of the emission factors for each different compound groups, as described in section "Analytical instrumentation". Figure 7 illustrates the difference between THC$_{FID}$ and VOC$_{FTIR}$ emission factors (mg km^{-1}) observed in WLTC
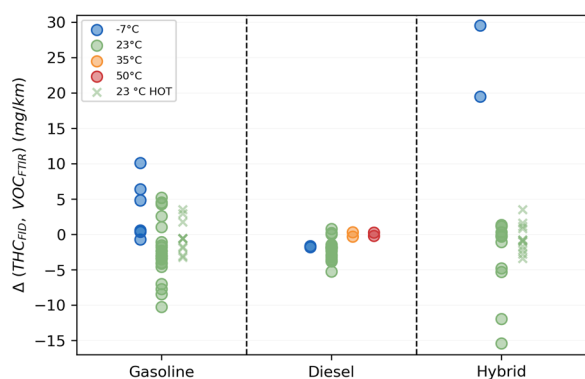
Fig. 7 Difference between THC_{FID} and VOC_{FTIR} emission factors (mg km^{-1}) obtained in WLTC tests performed on Diesel, conventional gasoline and PHEV vehicles, at different ambient temperatures and starting conditions

tests conducted on Diesel (DV1-DV5), conventional gasoline (GV1-GV5) and PHEVs (HV1-HV5) vehicles, under various ambient temperatures (−7 °C and 23 °C for gasoline vehicles and PHEVs, and −7 °C, 23 °C, 35 °C and 50 °C for Diesel vehicles). Tests at 23 °C in HOT starting conditions were also included in the analysis.

It is important to note that a high variability was detected in the difference between THC_{FID} and VOC_{FTIR} emission factors among different vehicles. Therefore, only qualitative discussions of the results are possible. For instance, the significant difference recorded in hybrid vehicles at −7 °C (> 20 mg km^{-1}) was related to tests performed on only one vehicle (HV2).

VOC_{FTIR} on Diesel vehicles exhibited a slight tendency to exceed FID measurements, corresponding to negative value of Δ (THC_{FID}, VOC_{FTIR}), as the temperature decreased. Conversely, they approach values very close to zero at higher temperatures. It should be noted that hydrocarbon emission factors in Diesel were consistently low in all tests performed, below 10 mg km^{-1}. HCHO emissions constituted a major fraction of VOC emissions in Diesel vehicles at lower temperatures, as evident from Fig. 3, accounting for more than 30% of VOC_{FTIR} emissions. Since FID has a very low sensitivity to HCHO [13], this could explain the negative values of Δ (THC_{FID}, VOC_{FTIR}) in Diesel vehicles at −7 °C.

On the other hand, gasoline vehicles and PHEVs showed a tendency toward positive Δ (THC_{FID}, VOC_{FTIR}) at −7 °C, and to negative values at 23 °C. A positive value of Δ (THC_{FID}, VOC_{FTIR}) may be linked to the potential presence of other HCs, such as long chain hydrocarbons or polyaromatic compounds, that remain undetected by the FTIR as not present in common libraries. Tests performed in HOT starting conditions are homogeneously distributed between −5 and +5 mg km^{-1} for both

gasoline vehicles and PHEVs, bearing in mind that under these conditions emissions were very low (< 10 mg km^{-1}).

Conclusion

Thirteen Euro 6d-TEMP and four Euro 6d light-duty vehicles, including six gasoline vehicles, five Diesel vehicles, and six Plug-in Hybrid vehicles (PHEV), were tested at the Vehicle Emission Laboratory (VELA) of the European Commission Joint Research Centre (EC-JRC) Ispra, Italy. Tests were performed over the type-approval Worldwide harmonized Light-duty vehicles Test Cycle (WLTC). The vehicles' VOC emissions were studied across a wide range of ambient temperatures (−7 °C, 23 °C, 35 °C, 50 °C), with and without an after-treatment system warmed up at the start of the test. Two vehicles (a conventional gasoline and a PHEV) were also tested on the Transport for London (TFL) driving cycle, which simulates an urban driving cycle characterized by stop and go traffic in congested conditions and on a high speed driving cycle, with sharp and frequent accelerations (BAB). Emissions of various VOCs, including CH_4, C_5, Aromatics, Alcohols, Aldehydes and hydrocarbons with less than 5 carbon atoms (< C_5), were measured using a Fourier-Transform Infrared Analyzer (FTIR). Additionally, Total Hydrocarbons (THC) were measured with a Flame Ionization Detector (FID).

The results showed that the overall emissions of VOCs over WLTC at 23 °C were generally low, with few cases exceeding 10 mg km^{-1}. Diesel vehicles, presented the lowest VOC emissions, with CH_4 as the primary contributor at a median emission factor of 2 mg km^{-1}. The primary contributor to total NMVOCs for gasoline vehicles and PHEVs was C_5, accounting for 30–45% and 45–60%, respectively. Moreover, emissions of NMVOCs from PHEVs driven in charge sustaining mode showed no substantial differences with respect to those measured for conventional gasoline vehicles.

Colder temperatures induced an increase in emissions for gasoline and PHEVs, affecting all the studied species. The most significant rise in emissions at lower temperatures was observed for Aromatics, with emissions increasing up to 11 times compared to those at 23 °C. This rise in emissions is mainly due to the longer light-off time of the catalytic system, resulting in significantly higher final emissions than in the tests at 23 °C. VOC emissions in Diesel vehicles were little affected by temperature except for Aldehydes at −7 °C, increasing up to 4 mg km^{-1}. Among the Aldehydes, formaldehyde (HCHO) was the major contributor. Similar to the WLTC tests, VOC emissions in TFL cycles generally rise as the temperature decreases. As expected, small temperature variations lead to small differences in VOC emissions for the gasoline vehicle (e.g., −7 °C

vs -10 °C and 5 °C vs 23 °C). The main fraction of the VOCs for these vehicles at all conditions were Aromatics and C_5, with an important contribution of $<C_5$ as well. VOC emissions took place mainly in the cold start phase independently of the cycle, but TFL, covering a shorter distance, results in higher emission factors. VOC emissions over the BAB cycle were generally very low for the gasoline vehicle, with all VOC emissions being below 10 mg km^{-1} at all studied temperatures and negligible for Aromatics, Alcohols and Aldehydes. This confirms that VOC emissions decrease significantly once the ATS is already warmed. This effect is independent of the environmental test temperature and although the cycle is very demanding, with significant and frequent high-speed accelerations, VOC emissions remained very low.

Finally, a comparison between THC emission measured with FID (THC_{FID}) and total VOC emission measured with FTIR (VOC_{FTIR}) showed that VOC_{FTIR} in Diesel vehicles exhibited a slight tendency to yield higher emission than FID measurements, possibly due to FID's low sensitivity to HCHO. On the other hand, gasoline vehicles and PHEVs showed a temperature-dependent trend: VOC_{FTIR} were lower than THC_{FID} at -7 °C, and higher at 23 °C.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s12302-024-00854-4.

Additional file 1: Figure S1. Speed profiles of WLTC, TFL and BAB test cycles. **Figure S2.** Comparison of VOC emission profiles and Engine Coolant Temperature (ECT) during WLTC at 23 °C for a conventional gasoline vehicle and a PHEV. Darker colours and dark blue line represent the VOC emission profile and ECT, respectively, over the WLTC test in "COLD" starting condition. Lighter colours and red line represent the VOC emission profile and ECT respectively, over the WLTC test in "HOT" starting condition. **Table S1.** Environmental specification of market fuels (petrol and Diesel), which complies with European Directive 2009/30/EC. **Table S2.** Limit of detection of the different hydrocarbon compound measured with FTIR and THC measured with FID. **Table S3.** Emission factors (EF) (mg km^{-1}) of all the VOCs for Diesel, gasoline and PHEV vehicles, during WLTC at the different testing temperatures (-7 °C, 23 °C, 35 °C, 50 °C). **Table S4.** Emission factors (EF) (mg km^{-1}) of all the VOCs for gasoline (GV6) and PHEV (HV6) vehicles, during TFL test cycle at the different testing temperatures (-30 °C, -10 °C, -7 °C, 5 °C, 23 °C, 50 °C). **Table S5.** Emission factors (EF) (mg km^{-1}) of all the VOCs for gasoline (GV6) and PHEV (HV6) vehicles, during BAB test cycle at the different testing temperatures (-30 °C, -10 °C, -7 °C, 5 °C, 23 °C, 50 °C).

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Declarations

Ethics approval and consent to participate
Not applicable.

Consent for publication
Not applicable.

Competing interests
The authors declare that they have no competing interests.

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