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POLYCHLORINATED DIBENZODIOXINS AND DIBENZOFURANS EMISSIONS FROM TRANSPORTATION

Measurement of Persistent Organic Pollutants (POPs) was performed in order to complement the set of known emission factors. Methodology for gasoline engines emission factors determination that uses POPs concentrations measured in exhaust gasses, limited pollutants emission factors (carbon monoxide CO, nitrogen oxides NO_x and hydrocarbons HC) and other compounds (carbon dioxide CO₂) was elaborated. Empirical value of exhaust gasses production announced by specialists of TUV SUD was used for diesel. Emission factors of four vehicle-fuel systems were determined particularly 11 congeners of Polychlorinated Dibenzodioxins (PCDDs), 34 congeners of Polychlorinated Dibenzofurans (PCDFs) and their toxic equivalents (TEQ) for each of the measured system. TEQ PCDD/F congener profile typical for POPs sources from transportation was compiled on the basis of measured data.

1. Introduction

Persistent Organic Pollutants (POPs) represent group of organic compounds with specific physico-chemical and environmental-chemical properties. The most important among them are resistance to diverse degradation processes, low solubility in water, lipophilic character. These properties results in high tendency for bioaccumulation, semi volatility enabling global atmospheric transport and significant adverse effects on human health or environment. Polychlorinated Dibenzodioxins (PCDDs) and Polychlorinated Dibenzofurans (PCDFs) belong among others to this group of compounds. They originate during communal, hospital or dangerous wastes incineration, also during coal, peat and wood combustion and can be also determined in vehicles emissions [1]. Their content in exhaust gasses is in very small concentrations and because of difficult and complicated analytical determination these compounds are measured in exhaust gasses only sporadically [2].

2. Measurements methodology

Emission factors (Ef) measurements of three vehicles using four different types of fuel were performed in two sampling campaigns in 2009 and 2010. Standard city cycle in accordance with ECE 83.04B consisting of 4 cycles ECE 15 in 195 sec. (Fig. 1) in three repetitions with previous 5 minutes engine heating up to running temperature (SDC) was chosen to simulate urban driving. Sufficient amount of exhaust gasses for PCDD/Fs and Polyaromatic Hydrocarbons (PAH) determination was sampled during these three cycles. Sampling was performed by using apparatus described in Fig. 2 connected directly to exhaust pipe. All measurements were performed in authorized station for vehicles homolo-

gation TUV SUD on dynamometer SCHENCK 364/GS56 that simulates flywheel mass and running resistance as if vehicle moves on the road.

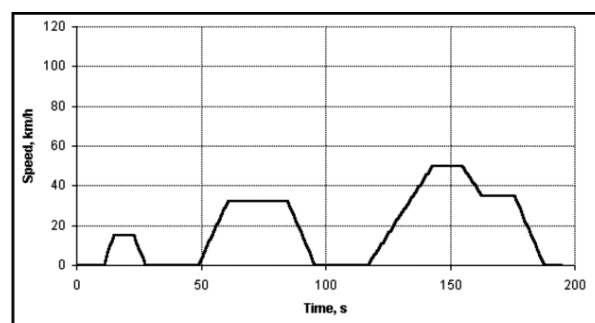


Fig. 1 Standard city cycle ECE 15

PCDD/Fs isotopic marked congeners were placed before the sampling to the sorption system to ensure control of PCDD/Fs sorption efficiency. Sampling and analyses were performed in accordance with CSN EN 1948-1-3 considering that measurements next to exhaust pipe was not possible to perform isokinetically. The same apparatus was used also for PAH sampling. Basic emission parameters such as CO₂, CO, HC, NO₂ were also measured at the same time.

Vehicle thus passed through the test in accordance with speed profile that is standard and described in ECE 83 regulation. Exhaust gasses were sampled in the whole volume using HORIBA CVS 7300 T device and were diluted to prevent condensation.

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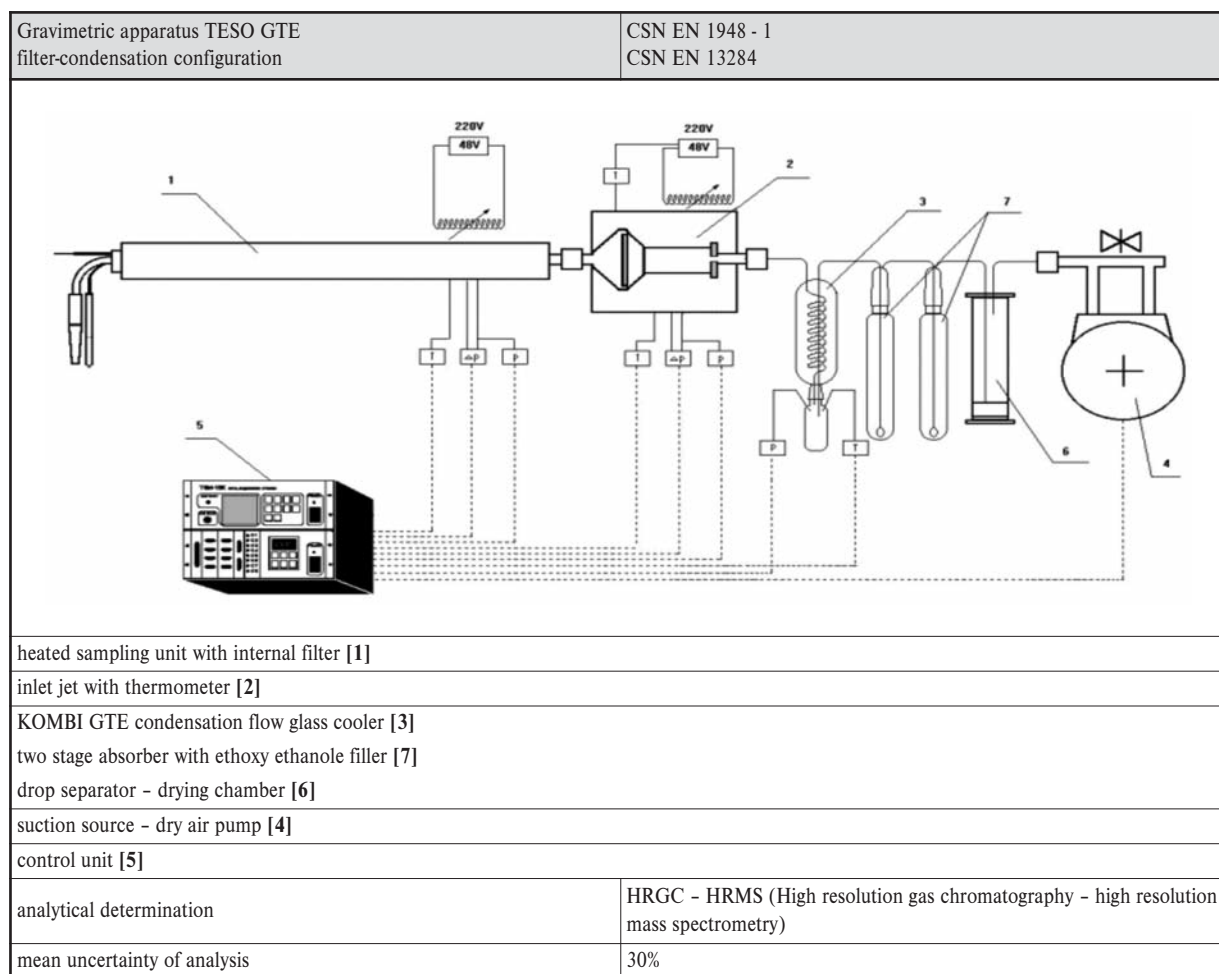


Fig. 2 Apparatus for PCDD/F and PAH sampling

Fuels used for measurements were tanked in common petrol stations. Their average chemical composition is characterised by parameters shown in Tab. 3. Symbols n_H and n_C mean the amount of hydrogen and carbon atoms in the mean molecule of fuel, ρ_p means fuel density, BA is 95 octane gasoline and MN summer diesel fuel.

3. Ef calculation methodology

Methodology results from Ef values of CO, NO_x, CO₂ and HC measured in dry exhaust gasses on dynamometer by standard methods and from fuel consumption. Ef of pollutants are calculated from measured concentration and measured volumes of dry exhaust gasses used for chosen pollutants sampling under normal conditions (101.325 kPa, 0 °C).

Symbols marked with upper index on the right side are related to dry gasses that means to gasses without any water vapours. Amount of substances entering the combustion process are marked with capital N with appropriate indexes, amount of substances of

Used symbols

Table 1

A_i	atomic weight of i -th element [g.mol ⁻¹]
a, b, c	stoichiometric coefficients for elements in mean molecule of fuel in sequence of C, H, O
c_i^s	i -th component concentration in dry exhaust gasses [g.m ⁻³]
$conc_i^s$	i -th component content in dry exhaust gasses of the sample [g.sample ⁻¹]
β	ratio of hydrogen atoms versus carbon atoms in fuel
γ	ratio of oxygen atoms versus carbon atoms in fuel
Ef_m^i	i -th component emission factor [g.kg ⁻¹]
Ef_v^i	i -th component emission factor [g.km ⁻¹]
FC	fuel consumption [l. (100 km) ⁻¹]
L	route passed by vehicle [km]
m_i	i -th component weight [kg]
M_i	i -th component molar weight [g.mol ⁻¹]
μ_i	i -th component molar weight relative to one carbon atom in the molecule [g.mol ⁻¹]

n_i	amount of substance of i-th component content in exhaust gasses [mol]
N_i	amount of substance of i-th component content in air [mol]
N_p	amount of substance of consumed fuel [mol]
N_V	amount of substance of air entering the engine during fuel combustion [mol]
π_i	i-th element percentage content in fuel [%]
V_a	gas one mol volume under standard conditions [L.mol ⁻¹]
V^s	volume of dry gasses sample [m ³ .sample ⁻¹]
V_{exh}	volume of dry exhaust gasses originated during fuel combustion [m ³]
ρ_p	fuel density [kg.l ⁻¹]

gasses in exhaust gasses are marked with n with appropriate indexes. Index p means fuel.

Physical constants used in calculations Table 2

Fuel	BA95	LPG	CNG	MN
Constant	Constant value			
M_{O_2} [mol.g ⁻¹]	31.9988			
M_{H_2O} [mol.g ⁻¹]	18.0015			
M_{CO} [mol.g ⁻¹]	28.01055			
M_{NO} [mol.g ⁻¹]	30.0061			
M_{NO_2} [mol.g ⁻¹]	46.0055			
M_{CO_2} [mol.g ⁻¹]	44.00995			
V_a [mol.g ⁻¹]	22.414			
M_p [mol.g ⁻¹]	104.07	53.53	16.014	173.64
A	7.328	3.684	1	12.36
β	1.808	2.523	4	1.913
ω	0.4389	0.6307	1	0.4739
π_o [%]	2.73	0	0	0.92
ρ_p [kg.l ⁻¹]	0.748	0.538	0.000654	0.832

It is necessary to know fuel composition when calculating Ef. The meanings of factors used for calculations are described in Table 1, physical constants in Table 2. Stoichiometric coefficients are introduced for simplification of further equations and their meaning is evident from equation (4)

$$\omega = \frac{b - 2 \cdot c}{4 \cdot a} = \frac{\beta}{4} - \frac{\gamma}{2} \quad (1)$$

$$\frac{N_p}{n^s} = \frac{Y_{o_2}^s - y_{o_2}^s + (1 - Y_{o_2}^s) \cdot \frac{y_{CO}^s}{2} - \left(1 - \frac{Y_{o_2}^s}{2}\right) \cdot y_{NO_2}^s - \frac{y_{NO}^s}{2} + \left(a \cdot (1 + \omega) - (1 + \omega) \cdot Y_{o_2}^s\right) \cdot y_p^s}{a \cdot (1 + \omega \cdot (1 - Y_{o_2}^s))} \quad (12)$$

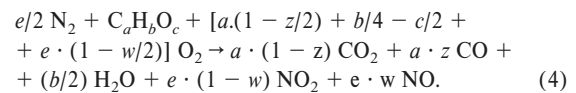
If atomic ratio H:C (= β) and oxygen percentage content π_o are known, then

$$\omega = \frac{\beta}{4} - \frac{\pi_o \cdot (A_c + \beta \cdot A_H)}{2 \cdot (100 - \pi_o) \cdot A_o} \quad (2)$$

From the equation for fuel molecular weight relative to one carbon atom after substitution from (1) and (2), it holds

$$\mu_p = \frac{M_p}{a} = A_c + \beta \cdot A_H + \gamma \cdot A_o = \frac{100 \cdot (A_c + \beta \cdot A_H)}{(100 - \pi_o)} \quad (3)$$

Emission factors of limited pollutants calculation results from equations of fuel combustion, mass balance and measured values. Fuel combustion is considered in some simplification (it is assumed that hydrocarbons contained in exhaust gasses are identical to those contained in fuel, origin of trace concentrations of PAH, N₂O, NH₃ and others are not considered):



Mass balance of real fuel combustion considering presumptions characterised above can be described by following equations for separate elements:

$$C: a \cdot (N_p - n_p) = n_{CO_2} + n_{CO} \quad (5)$$

$$H: b \cdot (N_p - n_p) = 2 \cdot n_{H_2O} \quad (6)$$

$$(N_{O_2} - n_{O_2}) + c \cdot (N_p - n_p) = n_{H_2O} + 2 \cdot n_{CO_2} + n_{CO} + 2 \cdot n_{NO_2} + n_{NO} \quad (7)$$

$$N: (N_{N_2} - n_{N_2}) = n_{NO_2} + n_{NO} \quad (8)$$

Amount of substance of dry gasses enters combustion process

$$N_V^s = N_V - N_{H_2O} = N_{N_2} + N_{O_2} + N_{CO_2} \quad (9)$$

and for amount of substance of dry gasses getting off the exhaust pipe holds

$$n^s = n - n_{H_2O} = n_{N_2} + n_{O_2} + n_{CO_2} + N_{CO_2} + n_{CO} + n_p + n_{NO_2} + n_{NO} \quad (10)$$

Symbol for i-th component concentration in entering air and in exhaust gasses formulated as molar fraction is described as

$$Y_i^s = \frac{N_i}{N_V^s}, y_i^s = \frac{n_i}{n^s}, y_i = \frac{n_i}{n} \quad (11)$$

and equation solution for unknown parameters N_V^s a N_p brings formula necessary for further calculations

If pollutant concentration is expressed in mass units per volume unit, Ef of separate components of exhaust gasses relative to unit of consumed fuel can be described as

$$Ef_m^i = \frac{m_i}{m_p} = c_i^s \cdot \frac{V_a}{M_p} \cdot \frac{n^s}{N_p} \quad (13)$$

Ef expressed in mass units per passed route and concentration input data in mass unit per volume unit of exhaust gasses are calculated as

$$Ef_i^f = Ef_m^i \cdot \frac{m_p}{l} = \frac{conc_i^s}{V^s} \cdot \frac{V_{exh}}{l} \quad (14)$$

Then results from equation (14) for rates for conversion from separate pollutants concentrations in exhaust gasses (POPs in this case) to Ef after substitution of the molar fraction from equation (11) and after neglecting NO_2 concentration (NO predominantly originates during combustion):

$$\frac{V_{exh}}{l} = \frac{V_a}{Y_{O_2}^s} \cdot \left(a \cdot \frac{FC \cdot \rho_p}{M_p} \cdot (1 + \omega \cdot (1 - Y_{O_2}^s)) + \frac{Ef_i^{CO}}{M_{O_2}} - (1 - Y_{O_2}^s) \cdot \frac{Ef_i^{CO}}{2 \cdot M_{CO}} - (1 + \omega) \cdot (a - Y_{O_2}^s) \cdot \frac{Ef_i^p}{M_p} + \frac{Ef_i^{NO_x}}{2 \cdot M_{NO_x}} \right) \quad (15)$$

The result from equation (14) is used for Ef calculation relative to passed route. After substitution of the result from equation (15) in dimensions described in Table 2 and after neglecting oxygen concentration in exhaust gasses, the formula for calculation of Ef of pollutants holds:

$$Ef_i^f = \frac{conc_i^s \cdot V_a}{1000 V^s \cdot Y_{O_2}^s} \cdot \left(a \cdot (1 + \omega \cdot (1 - Y_{O_2}^s)) \cdot \frac{10 \cdot FC \cdot \rho_p}{M_p} - (1 - Y_{O_2}^s) \cdot \frac{Ef_i^{CO}}{2 \cdot M_{CO}} - \frac{Ef_i^{NO_x}}{2 \cdot M_{NO_x}} + (1 + \omega) \cdot (a - Y_{O_2}^s) \cdot \frac{Ef_i^p}{M_p} \right) \quad (16)$$

Stoichiometric calculations without knowledge of oxygen concentration in exhaust gasses are not possible for diesel engines. Because of this fact it possible to use empirical value $1.2181 \text{ m}^3 \cdot \text{km}^{-1}$ for recalculation of POPs concentrations in exhaust gasses in accordance with recommendation of authorized station for vehicles homologation TÜV SÜD.

4. Results and discussion

Emission factors of common pollutants that were also used for POPs Ef calculations are described in Table 3. Upper index p means unburned hydrocarbons

Congener profiles for j -th measurement (17) or cumulative congener profiles (18) were compiled on the basis of measured data in accordance with following equations

$$Ef TEQ_j^i = Ef_j^i \cdot TEQ_j^i \quad (17)$$

$$Ef TEQ_i = \sum_i Ef_j^i \cdot TEQ_j^i, \quad (18)$$

where index i represents appropriate PCDD or PCDF congener. Cumulative Ef s are counted above all PCDD and PCDF congeners. Following PCDD congeners and their toxic equivalents were considered:

2378TCDD	12378PeCDD	123478HxCDD
1	0.5	0.1
123678HxCDD	123789HxCDD	1234678HpCDD
0.1	0.1	0.01
OCDD		
0.001		

Results of limited pollutants emission factors measurements

Table 3

Date	Vehicle type	Fuel	ρ_p kg.l ⁻¹	nH/nC	% O	nC	Tachom.	$EF_i^{CO_2}$ g.km ⁻¹	EF_i^{CO} g.km ⁻¹	EF_i^p g.km ⁻¹	$EF_i^{NO_x}$ g.km ⁻¹	$FC \cdot 10^2$ l.km ⁻¹
2.12.09	SKODA Felicia 1.3/50 kW	BA 95t	0.748	1.85	2.7	6	184965	242	0.716	0.136	0.178	10.27
2.12.09	SKODA Felicia 1.3/50 kW	LPG	0.538	0.538	0	3.5	184981	193	10.49	0.91	0.223	13.05
2.12.09	SKODA Fabia 1.4/44 kW	BA 95t	0.748	1.85	2.7	6	250389	243	0.826	0.105	0.497	10.28
2.12.09	SKODA Octavia 1.9 TDI/77 kW	MNt	0.832	1.91	0.92	12.36	156295	233	0.058	0.129	0.455	8.86
22.9.10	SKODA Felicia 1.3/50 kW	BA 95t	0.748	1.85	2.7	6	188083	267	0.682	0.088	0.133	11.3
22.9.10	SKODA Felicia 1.3/50 kW	LPG	0.538	0.538	0	3.5	188091	223	12.05	0.576	0.049	15.0
22.9.10	SKODA Fabia 1.4/44 kW	BA 95t	0.748	1.85	2.7	6	266232	215	0.568	0.042	0.443	9.1
22.9.10	SKODA Octavia 1.9 TDI/77 kW	NMt	0.832	1.91	0.92	12.36	179595	218	0.086	0.054	0.465	8.2

Legend: t means tanked in petrol station (same in Table 4)

and following PCDF congeners and their toxic equivalents were considered:

2378TCDF	12378PeCDF	23478PeCDF
0.1	0.05	0.5
123478HxCDF	123678HxCDF	
0.1	0.1	
234678HxCDF	123789HxCDF	1234678HpCDF
0.1	0.1	0.01
1234789HpCDF	OCDF	
0.01	0.001	

TEQ PCDD a PCDF Ef congener profiles shown in Figs. 3 and 4 indicate higher values in 2010 but profiles for separate years of measurement can be considered as similar. TEQ 2378TCDD Ef was the highest in 2009 whereas 12378PeCDD Ef was the highest in 2010 among all PCDD congeners. 23478PeCDF Ef was the highest among all PCDF congeners in both measuring campaigns. TEQ OCDD and OCDF Ef were the lowest although TEQ 123789HxCDF Ef was once (Octavia 2010) the lowest.

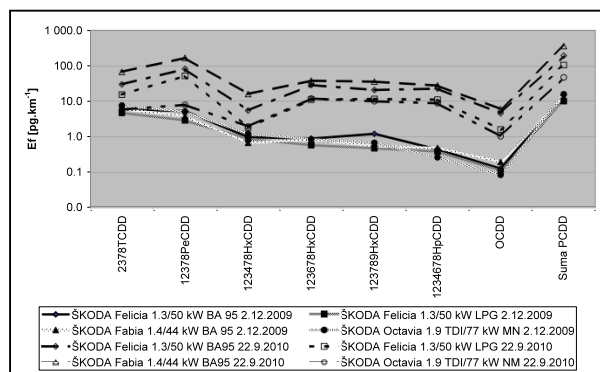


Fig. 3 TEQ-PCDD congener profiles (logarithmic scale of y axis)

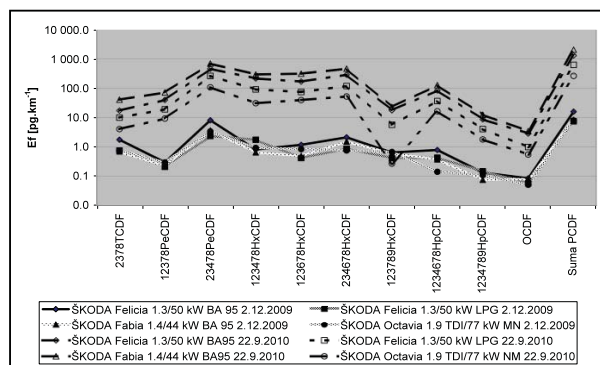


Fig. 4 TEQ-PCDF congener profiles (logarithmic scale of y axis)

Significant differences were found out among cumulative TEQ Ef in particular years. Predominant influence of Ef measured in

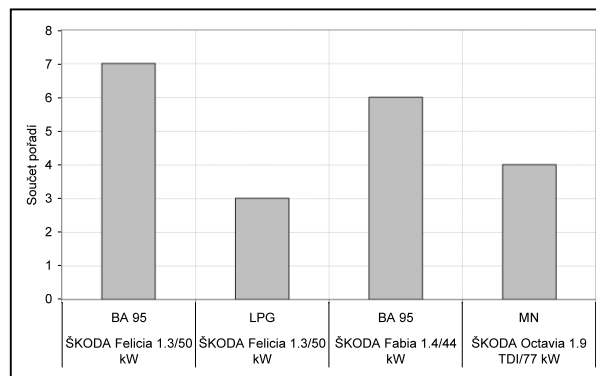


Fig. 5 Sum of rank of separate TEQ-PCDD/F emission factors of tested vehicles

2010 was demonstrated by comparison of arithmetic and geometric averages. Usage of sum of sequences of Ef measured in appropriate campaign and year according to their value is not dependent on absolute values of measured cumulative Ef (Fig. 5). In accordance with this criterion, PCDD/F Ef of Felicia with gasoline fuel had the highest toxic equivalent while PCDD/F Ef of the same vehicle using LPG fuel had the lowest toxic equivalent. Emission factors TEQ for conventional fuels (gasoline, diesel) decreased with decreasing age of vehicle. Usage of LPG in the same vehicle significantly decreased POPs emission in comparison with usage of conventional fuel. LPG had the lowest PCDD/F Ef among studied fuels.

Determined POPs Efs were compared with Efs measured by CDV for a wider selection of vehicles during 2005 - 2006 [3, 4]. Summary of measuring conditions in 2005 - 2010 is outlined in Table 4. Most of fuels were certificated. Trend of dependency of cumulative PCDD/F Ef sums for separate vehicles and fuel on measuring date for separate measuring campaign (one day or two days) is shown in Fig. 6. Efs of cold starts (CS) are also shown in this graph. Cold start represents SDC without pre heating after vehicle parking outside whole night and its movement onto the dynamometer with engine off.

Efs had similar character of their time progress. This time progress was in four campaigns similar to time progress of PAH Efs. Data variance was larger within the sampling campaigns than among campaigns. Geometric averages of Efs measured in campaigns were in the range of units up to tens of pg.km^{-1} (in the frame in Fig. 6). However, PCDD/F Efs were in two campaigns in 2006 lower (tenths to units of pg.km^{-1}) and in the last campaign in 2010, on the contrary, hundreds to thousands of pg.km^{-1} .

Since differences in PAH Efs measurements in contrast to PCDD/F Efs were not relevant, entrance of different amounts of chlorine or its compounds to the measuring systems in these three campaigns than in others was considered. Probably these compounds were contained in the air entering combustion process in vehicles engines.

Cold starts Efs were higher than Efs measured under running conditions for all fuels which is probably determined by different

Summary of cumulative emission factors measuring conditions in 2005-2010

Table 4

Measurement No.	Identification	Vehicle type	Fuel	Comment	Passed km
6523	F-BA 95	SKODA Fabia 1.4/44 kW	Natural 95	Certificated	139 500
6522	F-BA 95	SKODA Fabia 1.4/44 kW	Natural 95	Certificated	139 512
6520	F-BA 95-E5	SKODA Felicia 1.3/50 kW	Natural 91 s 5 % EtOH	Certificated	139 524
6521	F-BA 5-EE15	SKODA Felicia 1.3/50 kW	Natural 91 s 15 % ETBE	Certificated	139 536
6518	OS-MN	SKODA Octavia 1.9 SDI combi	MN, summer	Certificated	82 100
6519	OS-MN-M5	SKODA Octavia 1.9 SDI combi	MN summer with 5% MERO	Certificated	82 112
10542	F-LPG	SKODA Felicia 1.3/50 kW	LPG		142 900
10544	Fa-BA 95	SKODA Fabia 1.4/44 kW	Natural 95	Certificated	179 600
10543	Fa-BA 95-E5	SKODA Fabia 1.4/44 kW	Natural 91 s 5 % EtOH	Certificated	179 612
10545	Fa-BA 95-EE15	SKODA Fabia 1.4/44 kW	Natural 91 s 15 % ETBE	Certificated	179 624
10546	OS-MN-M31	SKODA Octavia 1.9 SDI combi	MN summer with 31% MERO	Certificated	93 900
14	Fa-BA 95t-cs	SKODA Fabia 1.4/44 kW	BA95 tanked	Cold start	183 053
15	OS-MNt-cs	SKODA Octavia 1.9 SDI combi	MN tanked	Cold start	96 902
16	FF-E85	Ford Focus Flexifuel	85% EtOH		2 900
17	FF-E85-cs	Ford Focus Flexifuel	85% EtOH	Cold start	2 912
2398	FM-CNG-cs	Fiat Multipla (manufactured CNG)	CNG	Cold start	n/a
2399	FM-CNG	Fiat Multipla (manufactured CNG)	CNG		n/a
2400	FF-E85-cs	Ford Focus Flexifuel	85% EtOH	Cold start	19 888
2401	FF-E85	Ford Focus Flexifuel	85% EtOH		19 900
2402	Fa-BA 95t-cs	SKODA Fabia 1.4/44 kW	BA95 tanked	Cold start	185 914
2403	OS-MNt-cs	SKODA Octavia 1.9 SDI combi	MN tanked	Cold start	105 588
2404	OS-MN-M5	SKODA Octavia 1.9 SDI combi	MN summer with 5% MERO	Certificated	105 600
2405	OS-MN-M31	SKODA Octavia 1.9 SDI combi	MN summer with 31% MERO	Certificated	105 612
5774	Fa-BA 95t-cs	SKODA Fabia 1.4/44 kW	BA95 tanked	Certificated	192 888
5775	Fa-BA 95	SKODA Fabia 1.4/44 kW	Natural 95	Certificated	192 900
5776	Fa-95-E5	SKODA Fabia 1.4/44 kW	Natural 91 with 5 % EtOH	Certificated	192 912
5777	Fa-BA 95-EE15	SKODA Fabia 1.4/44 kW	Natural 91 with 15 % ETBE	Certificated	192 924
5778	FaP-CNG	SKODA Fabia 1,4 Combi (reconstruction to CNG)	CNG		108 200
5779	OS-N-M5	SKODA Octavia 1.9 SDI combi	MN summer with 5% MERO	Certificated	114 700
5780	OS-N-M31	SKODA Octavia 1.9 SDI combi	MN summer with 31% MERO	Certificated	114 712
61700	F-BA 95t	SKODA Felicia 1.3/50 kW	BA95 tanked		185 000
61701	F-LPG	SKODA Felicia 1.3/50 kW	LPG		185 012
61702	Fa-BA 95t	SKODA Fabia 1.4/44 kW	BA95 tanked		250 400
61703	OT-MNt	SKODA Octavia 1.9 TDI/77 kW	MN, summer, tanked		156 300
68843	F-BA 95t	SKODA Felicia 1.3/50 kW	BA95 tanked		188 083
68844	F-LPG	SKODA Felicia 1.3/50 kW	LPG		188 091
68842	Fa-BA 95t	SKODA Fabia 1.4/44 kW	BA95 tanked		266 232
68845	OT-MNt	SKODA Octavia 1.9 TDI/77 kW	MN, summer, tanked		179 595

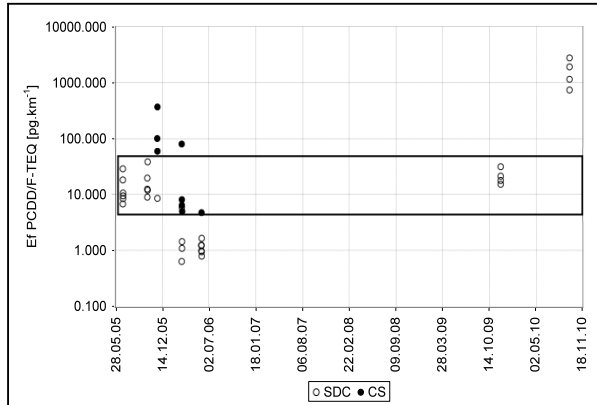


Fig. 6 Summary of cumulative PCDD/F emission factors measured by CDV (logarithmic scale)

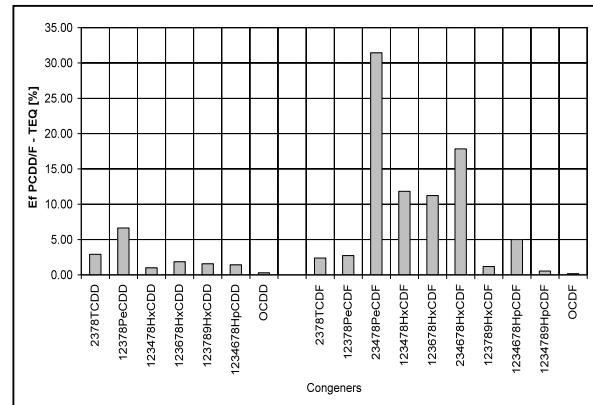


Fig. 7 Congener profile of TEQ PCDD/F emission factors mean values of all tested Vehicles

conditions of combustion (lower temperatures, incomplete combustion)

Cumulative TEQ emission factors for all the measuring campaigns, all the tested vehicles and fuels shown in Fig. 7 in percentage were calculated in accordance with equation (19)

$$EfTEQ^i = \frac{\sum_j Ef_j^i \cdot TEQ_j^i}{\sum_j \sum_i Ef_j^i \cdot TEQ_j^i} \cdot 100. \quad (19)$$

Congener profile calculated in accordance with equation (19) represents congener profile estimation of sources from individual passenger road transport.

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