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

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.
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, $\rho_p$ 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\textsubscript{x}, CO\textsubscript{2}, 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 different elements in mean molecule of fuel are marked with $a$, $b$, and $c$, stoichiometric coefficients for elements in mean molecule of fuel in sequence of C, H, O.

### Table 1

| 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 |
| $c_i^c$      | $i$th component concentration in dry exhaust gasses [g.m\(^{-1}\)] |
| $c_i^s$      | $i$th component content in dry exhaust gasses of the sample [g.sample\(^{-1}\)] |
| $\beta$      | ratio of hydrogen atoms versus carbon atoms in fuel |
| $\gamma$     | ratio of oxygen atoms versus carbon atoms in fuel |
| $Ef_i^c$     | $i$th component emission factor [g.kg\(^{-1}\)] |
| $Ef_i^l$     | $i$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\(^{-1}\)] |
| $\mu_i$      | $i$th component molar weight relative to one carbon atom in the molecule [g.mol\(^{-1}\)] |
gases 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 |      |      |      |      |     |
| \( M_{H_2} \) [mol.g\(^{-1}\)] |   | 31.998 | | | |
| \( M_{CO_2} \) [mol.g\(^{-1}\)] | 18.0015 | | | | |
| \( M_{CO} \) [mol.g\(^{-1}\)] | 28.01055 | | | | |
| \( M_{NO} \) [mol.g\(^{-1}\)] | 30.0061 | | | | |
| \( M_{SO} \) [mol.g\(^{-1}\)] | 46.0055 | | | | |
| \( M_{CO_2} \) [mol.g\(^{-1}\)] | 44.00995 | | | | |
| \( V_{exh} \) [mol.g\(^{-1}\)] | 22.414 | | | | |
| \( V_{a} \) [mol.g\(^{-1}\)] | 104.07 | 53.51 | 16.014 | 173.64 |
| \( A \) | 7.328 | 3.684 | 1 | 12.36 |
| \( \beta \) | 1.808 | 2.523 | 4 | 1.913 |
| \( \omega \) [%] | 0.4389 | 0.6307 | 1 | 0.4739 |
| \( \eta_a \) [%] | 2.73 | 0 | 0 | 0.92 |
| \( \rho_p \) [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)

\[
\omega = \frac{b - 2 \cdot c}{4 \cdot a} = \frac{\beta}{4} - \frac{\gamma}{2} \tag{1}
\]

If atomic ratio \( H:C = (\beta) \) and oxygen percentage content \( \pi_o \) are known, then

\[
\omega = \frac{\beta}{4} - \frac{\pi_o}{2} \left( A_c + \beta \cdot A_o \right) \tag{2}
\]

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

\[
\mu_p = \frac{M_p}{a} = A_c + \beta A_o + \gamma A_o = \frac{100 \left( A_c + \beta A_o \right)}{100 - \pi_o} \tag{3}
\]

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 \( \text{PAH, N}_2\text{O, NH}_3 \) and others are not considered):

\[
\begin{align*}
\frac{\varepsilon}{2} \text{N}_2 + \text{C}_n\text{H}_m\text{O}_z & \rightarrow a \cdot (1 - z/2) + b/4 - c/2 + \\
+ e \cdot (1 - w/2) \text{O}_2 & \rightarrow a \cdot (1 - z) \text{CO}_2 + a \cdot z \text{CO} + \\
+ (b/2) \text{H}_2\text{O} & \rightarrow e \cdot (1 - w) \text{NO}_2 + e \cdot w \text{NO}.
\end{align*} \tag{4}
\]

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

\[
\begin{align*}
\text{C:} & \quad \left( N_c - n_a \right) = n_{co} + n_{co} \tag{5} \\
\text{H:} & \quad b \cdot \left( N_p - n_a \right) = 2 \cdot n_{h_2} \tag{6} \\
\text{O:} & \quad \left( N_o - n_a \right) + c \cdot \left( N_c - n_a \right) = n_{m_o} + 2 \cdot n_{co} + \\
& \quad + n_{co} + 2 \cdot n_{no} + n_{no} \tag{7} \\
\text{N:} & \quad \left( N_n - n_a \right) = n_{no} + n_{no}. \tag{8}
\end{align*}
\]

Amount of substance of dry gasses enters combustion process

\[
N'_v = N_v - N_{no} = N_n + N_o + N_{co} \tag{9}
\]

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

\[
\begin{align*}
n' = n - n_{h_2} = n_{o} + n_{o} + n_{co} + N_{co} + n_{co} + \\
& + n_{o} + n_{no} + n_{no}. \tag{10}
\end{align*}
\]

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

\[
Y'_i = \frac{N_{i'}}{N'_v}, \quad y_i = \frac{n_i}{n'} \tag{11}
\]

and equation solution for unknown parameters \( N_{i'} \) a \( N_p \) brings formula necessary for further calculations

\[
\frac{N_i}{n'} = \frac{Y'_{o_i} - Y'_{o_i} + \left( 1 - Y'_{o_i} \right) \frac{Y'_{o_i}}{2} - \left( 1 - \frac{Y'_{o_i}}{2} \right) Y'_{o_i} - Y'_i + \left( a \cdot \left( 1 + \omega \right) \left( 1 + \omega \right) Y'_{o_i} \right) Y'_i}{a \cdot \left( 1 + \omega \cdot \left( 1 - Y'_{o_i} \right) \right)} \tag{12}
\]
If pollutant concentration is expressed in mass units per volume unit, $E_f$ of separate components of exhaust gases relative to unit of consumed fuel can be described as

$$E_{f_i} = \frac{m_i}{m_c} = \frac{c_i \cdot V_i}{n_i \cdot N_v}.$$  \hspace{1cm} (13)

$E_f$ expressed in mass units per passed route and concentration input data in mass unit per volume unit of exhaust gases are calculated as

$$E_{f_i} = \frac{m_i}{l} = \frac{\text{conc}_{i}}{V} \cdot \frac{V_{exh}}{l}. \hspace{1cm} (14)$$

Then results from equation (14) for rates for conversion from separate pollutants concentrations in exhaust gases (POPs in this case) to $E_f$ after substitution of the molar fraction from equation (11) and after neglecting NO$_2$ concentration (NO predominantly originates during combustion):

$$\frac{V_{exh}}{l} = \frac{V_i}{Y_{o_i}} \left( a \cdot \frac{FC \cdot \rho_{c}}{M_p} (1 + \omega (1 - Y_{o_i})) + \frac{E_{f_i}}{2} \cdot \frac{\rho_{c}}{M_p} - (1 - Y_{o_i}) \right) \cdot \frac{E_{f_i}^{CO}}{2 \cdot M_{CO}} - (1 + \omega) \left( a - Y_{o_i} \right) \frac{E_{f_i}^{NO}}{M_p} + \frac{E_{f_i}^{NO}}{2 \cdot M_{NO}} \hspace{1cm} (15)$$

The result from equation (14) is used for $E_f$ calculation relative to passed route. After substitution of the result from equation (15) in dimensions described in Table 2 and after neglecting oxygen concentration (O$_2$), the formula for calculation of $E_f$ of pollutants holds:

$$E_{f_i} = \frac{\text{conc}_{i}}{1000V} \cdot \frac{V_i}{Y_{o_i}} \left( a \left(1 + \omega \left(1 - Y_{o_i}\right)\right) \cdot \frac{10 \cdot FC \cdot \rho_{c}}{M_p} - (1 - Y_{o_i}) \cdot \frac{E_{f_i}^{CO}}{2 \cdot M_{CO}} - \frac{E_{f_i}^{NO}}{2 \cdot M_{NO}} + (1 + \omega) \left( a - Y_{o_i} \right) \frac{E_{f_i}^{NO}}{M_p} \right). \hspace{1cm} (16)$$

Stoichiometric calculations without knowledge of oxygen concentration in exhaust gases are not possible for diesel engines. Because of this, it was possible to use empirical value 1.2181 m$^3$.km$^{-1}$ for recalculation of POPs concentrations in exhaust gases 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 $E_f$ 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:

$$E_f \cdot TEQ_j = E_{f_i} \cdot TEQ_{j_i} \hspace{1cm} (17)$$

$$E_f \cdot TEQ = \sum_{i} E_{f_i} \cdot TEQ_{j_i} \hspace{1cm} (18)$$

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

| Congener   | TEQ |
|------------|-----|
| $12378$TCDD | 2378 |
| $12378$PeCDD | 12378 |
| $123478$HxCDD | 123478 |
| $123478$HpCDD | 123478 |
| OCDD       | 0.1 |
| $0.001$    |     |

Results of limited pollutants emission factors measurements

| Date       | Vehicle type | Fuel | $\rho_{p}$ kg.l$^{-1}$ | $nH/nC$ | % O | nC | Tachom. | $E_{Fl}^{CO2}$ g.km$^{-1}$ | $E_{Fl}^{CO}$ g.km$^{-1}$ | $E_{Fl}^{NO}$ g.km$^{-1}$ | $E_{Fl}^{NOx}$ g.km$^{-1}$ | $FC.10^{13}$ Lkm$^{-1}$ |
|------------|--------------|------|------------------------|---------|-----|----|---------|--------------------------|--------------------------|----------------------------|---------------------------|--------------------------|
| 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| 184891 | 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 | MNT  | 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:

\[
\begin{align*}
2378\text{TCDF} & \quad 12378\text{PeCDF} & \quad 23478\text{PeCDF} \\
0.1 & \quad 0.05 & \quad 0.5 \\
123478\text{HxCDF} & \quad 123678\text{HxCDF} & \\
0.1 & \quad 0.1 & \\
234678\text{HxCDF} & \quad 123789\text{HxCDF} & \quad 1234678\text{HpCDF} \\
0.1 & \quad 0.1 & \quad 0.01 \\
1234789\text{HpCDF} & \quad \text{OCDF} & \\
0.01 & \quad 0.001 &
\end{align*}
\]

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.

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
| 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          |              | 183 053   |
| 15             | OS-MNt-cs      | SKODA Octavia 1.9 SDI combi | MN tanked          |              | 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              |              | 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          |              | 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 s 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-M15         | SKODA Octavia 1.9 SDI combi | MN summer with 5% MERO | Certificated  | 114 700   |
| 5780           | OS-M13         | 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   |
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)

\[
Ef_{TEQ} = \frac{\sum_j Ef_i \cdot TEQ_j}{\sum_j Ef_i \cdot TEQ_j} \cdot 100.
\]  

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

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

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