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THE PROBLEM OF EVALUATION OF INDIVIDUAL PERSISTENT ORGANIC POLLUTANTS EMISSIONS FROM ROAD TRANSPORT (ILLUSTRATED BY THE CASE OF ODESSA INDUSTRIAL-AND-URBAN AGGLOMERATION)

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Abstract. One of the main environmental polluters, especially in large cities of Ukraine, is road transport. In particular, road transport is one of the main sources of persistent organic pollutants in the environment. Therefore, monitoring of air pollution by road transport is one of the most relevant questions in Ukraine, especially considering its intentions to join the European Union. Analysis of the regulatory and legislative frameworks of Ukraine shows that, unfortunately, today, there are no regulatory documents that motivate to reduce the load of mobile sources, including road transport, on the environment. Moreover, even though the only methodology for calculating pollutant emissions from mobile sources expired in 2015, no new methodology has been developed yet. This paper calculates the masses of pollutants formed during the combustion of gasoline and diesel fuel, and proves the necessity to take into consideration persistent organic pollutants when assessing the toxicity of exhaust fumes, considering their cumulative effect and half-life. The calculation was performed according to European and national (Ukrainian) methodologies, which are supplemented by the calculation of polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) formation. Using the relative hazard factor, the relative masses of these substances are calculated, the obtained masses of all components of the exhaust fumes are ranked, the contribution of each component to the toxicity of emissions is revealed. Using emission factors, the specific emissions of these persistent organic pollutants during the combustion of diesel fuel, gasoline, liquefied and compressed gases are calculated. By the results, the types of organic fuels with the highest priority for use by mobile sources are identified.

Keywords: mobile sources, road transport, persistent organic pollutants, polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans, toxicity of emissions.

1. Introduction

All over the world, much attention is paid to the monitoring of environmental pollution by road transport (Jason at al., 2021; Chengyong at al., 2021; Deya, Mehtab, 2020), in particular, to search for the most environmentally friendly fuel and transport (Longo at al., 2018; Falbo at al., 2020). This problem is also relevant to Ukraine, which seeks to meet the strict requirements of the European Union (EU) in environmental safety and adapts the procedures of atmospheric monitoring to the approach in EU countries (Ukrainian Ministry, 2019). As road transport is one of the main sources of unintentional production of persistent organic pollutants (POPs), especially with their extreme toxicity, taking into account their emissions when using organic fuel is necessary to ensure environmental safety in urban areas. According to (UNEP, 2013), the combustion of organic fuel by road transport produces such POPs as polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). As in all combustion processes, POPs are formed after the combustion and cooling of exhaust fumes. That is why the emissions of PCDDs and PCDFs largely depend on the conditions under which the cooling of the furnace

and exhaust gases is conducted. The main ways of getting into the environment are emissions into the air basin, especially as part of fly ash.

It is important to note that road transport is one of the main air polluters in industrial-and-urban agglomerations (IUA). Atmospheric air monitoring data of the Odessa IUA indicates that 70–80 % of air pollution is from mobile sources. Therefore, the calculation of the pollutants generation, including POPs, from the combustion of organic fuel is carried out using the example of Odessa IUA.

As road transport is a constant source of air pollution, the choice of a more environmentally friendly type of fuel can significantly reduce the anthropogenic load on the urban environment components. However, in Ukraine, there are no effective mechanisms to encourage people to choose road transport with a safer type of internal combustion engine. In particular, the Law of Ukraine (VRU, 2015), which came into force on 01.01.2015, stopped the taxation of pollutant emissions into the atmosphere by mobile sources by the environmental tax.

At the same time, the national (Ukrainian) methodology of 2008 (VRU, 2008), which allowed to calculate emissions of pollutants and greenhouse gases from vehicles expired on 19.02.2015 (State Statistical Service of Ukraine, 2015), and to date, there is no methodology that would allow this calculation. Thus, the negative impact of emissions from road transport at the legislative level has not been regulated since 2015. Moreover, even the previous method of 2008, which expired, did not take into account PCDD and PCDF in the calculation, and the quality of statistical information does not allow the use of modern European methods of calculation. This determines the urgency of assessing the impact of road transport on the environment, taking into account the toxicity of PCDD and PCDF.

Thus, the purpose of this study is to carry out the evaluation of individual persistent organic pollutants emissions from road transport (illustrated by the case of Odessa industrial-and-urban agglomeration) using available methodologies, to identify priority pollutants in exhaust fumes and the safest type of organic fuel.

2. Materials and Methods

The calculation of pollutant production, including POPs, from the combustion of organic fuel by road transport in the Odessa IUA, is performed based on two methods: national (Ukrainian) (VRU, 2008) and European (EMEP, 2019). This is done to compare the best existing approaches in Ukraine with the current European, which can be used based on the quality of statistical information in Ukraine. The calculation of

pollutant emissions is carried out according to the following formula:

$$E_{pollutant} = EF_{pollutant} \cdot M_{fuel}, \quad (1)$$

$EF_{pollutant}$ is emission factor of pollutant; M_i is mass of burned organic fuel, t; E_i is emission of pollutant, g (for all pollutants except POPs) or g TEQ PCDDs and PCDFs (for POPs).

TEQ is toxicological (dioxin) equivalent, which is a unit of cumulative toxicity of POPs. This value expresses the cumulative toxicity of complex mixtures of PCDDs and PCDFs due to the toxicity of the most toxic of the mixture – 2,3,7,8-TCDD. It is common to express the toxicity of mixtures of PCDDs and PCDFs in TEQ TCDD, which is equal to 1.

Given that the combustion of organic fuels by road transport is one of the main sources of unintentional formation of POPs, in particular PCDDs and PCDFs, we have supplemented the existing methodology of calculating these toxic substances. For this purpose, the corresponding emission factors from the European methodology are used (UNEP, 2013).

Also, based on the properties of PCDDs and PCDFs, we have improved the existing method of their calculation, taking into account the accumulation of these substances and the half-life. Given the extreme persistency of these substances in the environment and the human body, it is proposed to consider the formation of these pollutants not only for the current year but also taking into account the accumulation of these substances over the previous 10 years. Thus, the amount of PCDDs and PCDFs in the environment in 2017 will consist of the total mass of these pollutants formed in 2017 and the mass of PCDDs and PCDFs that did not decompose in the environment during 2007–2016. This technique is designed as a scientific work, which is protected by a copyright certificate (Mykhailenko, Safranov, 2021) and has the following form:

$$A_t = A \cdot e^{-\frac{t}{\tau}}, \quad (2)$$

A_t is mass of POPs after half-life for a period of time t ; A is initial mass of POPs; t is the period of time under consideration; τ is the period of time during which the concentration decrease in e times.

It should be noted that for PCDDs and PCDFs the average half-life in the environment is 10 years [4], so τ (PCDDs and PCDFs) is 14.5 years.

To compare the toxicity of exhaust fumes components, we used the concept of reduced mass – a conditional value that allows in a comparable form to reflect the harmfulness or environmental hazard of some pollutants in the emission relative to others. To calculate this value, use the formula:

$$MP_{pollutant} = A_{pollutant} \cdot M_{pollutant} \quad (3)$$

A_i is the relative hazard factor of the pollutant, calculated by the formula:

$$A_{pollutant} = 1/MAC_{pollutant} \quad (4)$$

$MAC_{pollutant}$ is the daily-average maximum allowable concentration (MACda) of pollutant in the air of

populated areas, mg/m^3 (in the absence of MACda, we used the maximum one-time MAC (MACmot) of pollutant in the atmospheric air of populated areas, mg/m^3 ; in the absence of MACmot, a temporary safe reference action level (TSRAL) was used; in the absence of TSRAL, we used the maximum allowable concentration of pollutant in the air of the working area (MACwa), reduced by 10 times.

3. Results and Discussion

Using national (Ukrainian) (VRU, 2008) and European (EMEP, 2019) methodologies, we calculated the value of pollutant emissions from road transport in the Odesa IUA for 2017 (Table 1). It should be noted that the calculation of the two methods was carried out to compare the methodologies available for use, although the Ukrainian method is not valid since 2015.

Table 1

Calculation of the pollutants emission into the atmosphere from road transport in the Odesa IUA for 2017 according to European and national (Ukrainian) methodologies

Pollutant	Gasoline				Diesel fuel			
	European methodology		Ukrainian methodology		European methodology		Ukrainian methodology	
	Emission factor, kg/t	Emission, kg	Emission factor, kg/t	Emission, kg	Emission factor, kg/t	Emission, kg	Emission factor, kg/t	Emission, kg
CO	152.3	$2.08 \cdot 10^7$	201.8	$1.92 \cdot 10^7$	7.4	$7.68 \cdot 10^5$	36.2	$3.76 \cdot 10^6$
NMVOG	14.59	$1.99 \cdot 10^6$	53	$5.06 \cdot 10^6$	1.54	$1.60 \cdot 10^5$	3.08	$3.20 \cdot 10^5$
PM	0.02	$2.73 \cdot 10^3$	0	0	1.52	$1.58 \cdot 10^5$	3.85	$4.00 \cdot 10^5$
NO _x	13.22	$1.80 \cdot 10^6$	21	$2.00 \cdot 10^6$	14.91	$1.55 \cdot 10^6$	31.4	$3.26 \cdot 10^6$
N ₂ O	0.186	$2.54 \cdot 10^4$	n.a	n.a	0.056	$5.81 \cdot 10^3$	n.a	n.a
NH ₃	0.667	$9.10 \cdot 10^4$	0.004	$3.82 \cdot 10^2$	0.038	$3.94 \cdot 10^3$	0	0
B(a)P	$4.2 \cdot 10^{-6}$	$5.73 \cdot 10^{-1}$	0	0	$15.8 \cdot 10^{-6}$	1.64	0.03	$3.11 \cdot 10^3$
Pb	$3.3 \cdot 10^5$	4.5	0.013	$1.24 \cdot 10^3$	$5.2 \cdot 10^5$	5.40	0	0
CO ₂	3180	$4.34 \cdot 10^8$	3183	$3.04 \cdot 10^8$	3140	$3.26 \cdot 10^8$	3138	$3.26 \cdot 10^8$
CH ₄	n.a	n.a	0.94	$8.97 \cdot 10^4$	n.a	n.a	0.083	$8.61 \cdot 10^3$

Note: NMVOG – non-methane volatile organic compounds; PM – particulate matter; B(a)P – benzopyrene; emission factor of pollutant (EF), emission (E) – see formula (1); n.a – not available.

The obtained calculations show that a larger number of pollutant emissions by the national (Ukrainian) method is higher than similar values of pollutants in the European method. This may be due to the fact that the emission factors in the national methodology were established long ago and took into account the state of the car fleet, which was relevant at the time of the methodology. It should be noted that the value of Tier 1 coefficients was used to calculate emissions from motor transport according to the European methodology (EMEP, 2019), which takes into account the specifics of the EU car fleet as of 1995.

One of the main limitations of the national (Ukrainian) methodology is the value of the emission factor “0” for such pollutants as benzopyrene and Pb. This is unacceptable, given the specifics and hazards of these substances. Moreover, road transport is the main source of urban environment pollution by heavy metals and benzopyrene.

According to the national methodology, the combustion of gasoline does not produce particulate matter, which is a false statement, because when burning gasoline in internal combustion engines, especially in the old models, particulate matter is present in the

exhaust fumes. Moreover, particulate matter is the main carrier (sorber) of heavy metals, which are formed in the exhaust fumes and settle to the surface with it. Therefore, taking into account the calculation of particulate matter and Pb in the national method is interrelated.

The considered European methodology at Tier 1 does not take into account the formation of CH₄ during the combustion of organic fuel, in contrast to the method (VRU, 2008). Estimation of CH₄ formation from mobile sources allows to present more accurately the picture of greenhouse gases formation in Ukraine and to make more thorough decisions to reduce their production, which is the main condition of the Paris Agreement.

As considerable attention is usually paid to environmental pollution with POPs, we have supplemented the above methodologies by calculating the emission of PCDDs and PCDFs. Based on information sources (Head of Department of Statistics in the Odesa region, 2017; Kopytov, 2012; Conversion calculator, 2021) we calculate the gross unintentional emissions of POPs from road transport of the Odesa IUA in 2017 based on updated data using the appropriate volumes of fuel. Thus, PCDDs and PCDFs

emissions are 0.3 g TEQ THDD for gasoline and 0.015 g TEQ THDD for diesel. According to (UNEP, 2013), during natural gas combustion, PCDDs and PCDFs are not produced.

According to formula (2), we determined the annual masses of POPs inflows into the environment of the Odesa IUA during the combustion of organic fuel

during 2007-2017, and also the accumulated masses of PCDDs and PCDFs, taking into account the half-life and cumulative effect of these substances. Based on the obtained results, graphs are constructed (Figs. 1 and 2), which clearly show the dynamics of changes in the mass of PCDDs and PCDFs with each passing year, taking into account their half-life and cumulative effect.

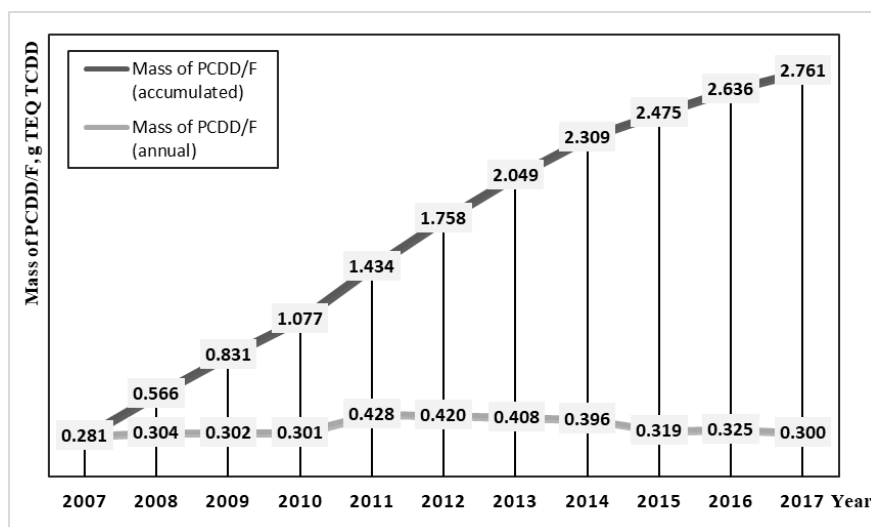


Fig. 1. Inputs of PCDDs and PCDFs in the environment of the Odesa IUA from gasoline combustion, taking into account and without taking into account the cumulative effect

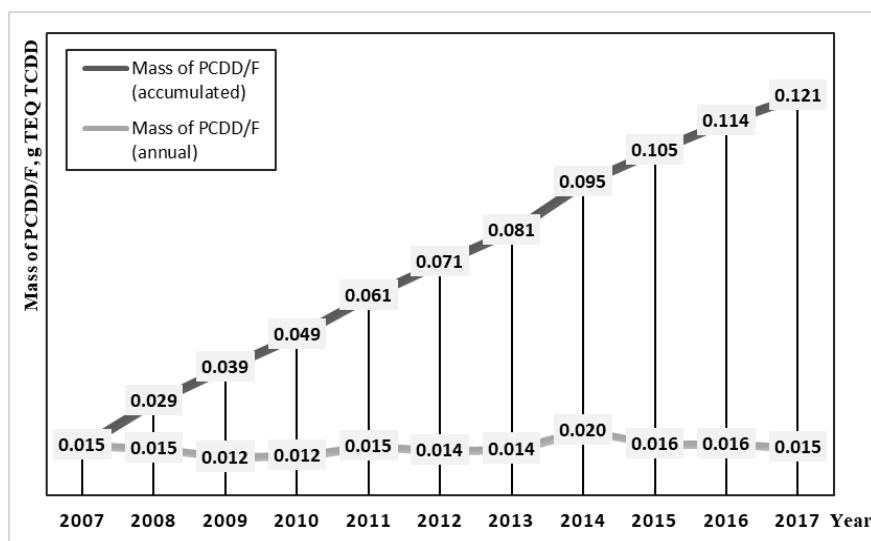


Fig. 2. Inputs of PCDDs and PCDFs in the environment of the Odesa IUA from diesel fuel combustion, taking into account and without taking into account the cumulative effect

The results show that the value of PCDDs and PCDFs, taking into account the half-life and cumulative effect is much greater than the annual mass of PCDDs and PCDFs, which enters the air of the Odesa IUA, and more accurately reflects the real picture of pollution, given the specifics of considered substances. In particular, the difference between the values of annual income without taking into account the half-life and the

cumulative effect is almost an order of magnitude smaller.

Based on the calculations obtained using formula 3, the values of the reduced masses for all pollutants were calculated using Ukrainian and European methods. The results are presented in Tables 2 and 3.

The percentage toxicity of the components for each fuel is shown in Figures 3 and 4.

Table 2

Calculation of the reduced mass values of pollutants in the combustion of gasoline by mobile sources in the Odesa IUA for 2017 according to the Ukrainian and the EU methodologies

Pollutant	MAC, mg/m ³	A	Ukrainian methodology			European methodology		
			E, t	E, reduced t	Percent	E, t	E, reduced t	Percent
CO	3.00	0.33	19248.62	6416.21	9.23	20773.72	6924.57	11.13
NM VOC	1.50	0.67	5055.38	3370.26	4.85	1990.08	1326.72	2.13
PM	0.05	20	0	0	0	2.73	54.56	0.09
NO _x	0.04	25	2003.08	50076.92	72.02	1803.21	45080.20	72.48
N ₂ O	0.06	16.67	–	–	–	25.37	422.84	0.68
NH ₃	0.04	25	0.38	9.54	0.01	90.98	2274.47	3.66
B(a)P	0.1·10 ⁻⁵	1·10 ⁶	0	0	0	0.0006	572.88	0.92
Pb	0.3·10 ⁻³	3.3·10 ³	1.24	4133.33	5.94	0.0045	15.00	0.02
CO ₂	–	–	303609.23	–	–	433752.00	–	–
CH ₄	50.00	0.02	89.66	1.79	0.00	–	–	–
PCDD/F	0.5·10 ⁻⁹	2·10 ⁹	0.19·10 ⁻⁵	3820.00	7.94	0.19·10 ⁻⁵	3820.00	8.88

Table 3

Calculation of the reduced mass values of pollutants in the combustion of diesel fuel by mobile sources in the Odesa IUA for 2017 according to the Ukrainian and the EU methodologies

Pollutant	MAC, mg/m ³	A	Ukrainian methodology			European methodology		
			E, t	E, reduced t	Percent	E, t/t	E, reduced t	Percent
CO	3.00	0.33	3757.31	1252.44	0.04	768.07	256.02	0.58
NM VOC	1.50	0.67	319.68	213.12	0.01	159.84	106.56	0.24
PM	0.05	20.00	399.60	7992.07	0.25	157.77	3155.31	7.12
NO _x	0.04	25.00	3259.10	81477.59	2.54	1547.56	38688.88	87.33
N ₂ O	0.06	16.67	–	–	–	5.81	96.87	0.22
NH ₃	0.04	25.00	0	0	0	3.94	98.60	0.22
B(a)P	0.1·10 ⁻⁵	1·10 ⁶	3.11	3113793.10	97.16	0.0016	1639.93	3.70
Pb	0.3·10 ⁻³	3.3·10 ³	0	0	0	0.0054	17.99	0.04
CO ₂	–	–	325702.76	–	–	325910.3	–	–
CH ₄	50.00	0.02	8.61	0.17	0.0001	–	–	–
PCDD/F	0.5·10 ⁻⁹	2·10 ⁹	0.84·10 ⁻⁷	168.00	0.01	0.84·10 ⁻⁷	168.00	0.55

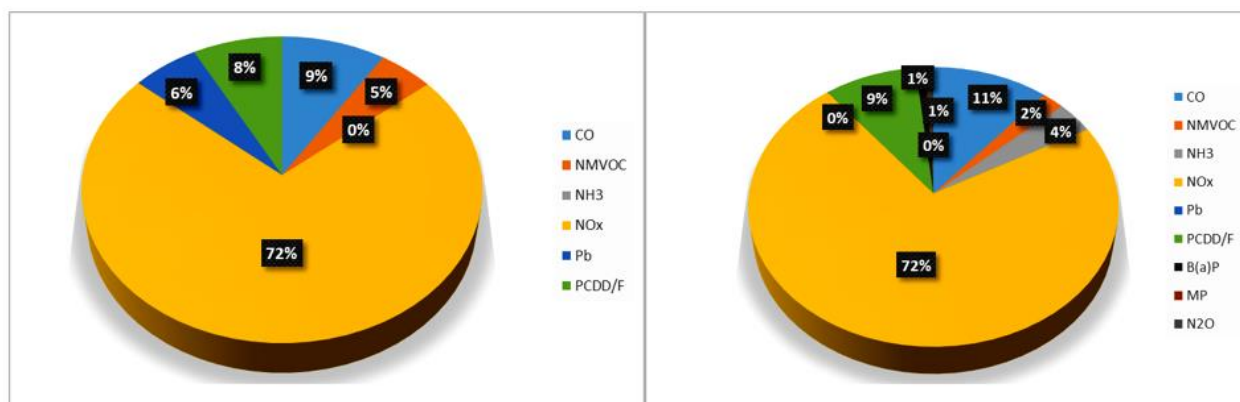


Fig. 3. Percentage ratio of the toxicities of the of individual components of the emissions from the gasoline combustion in the Odesa IUA for 2017, calculated using the Ukrainian (a) and European (b) methodologies

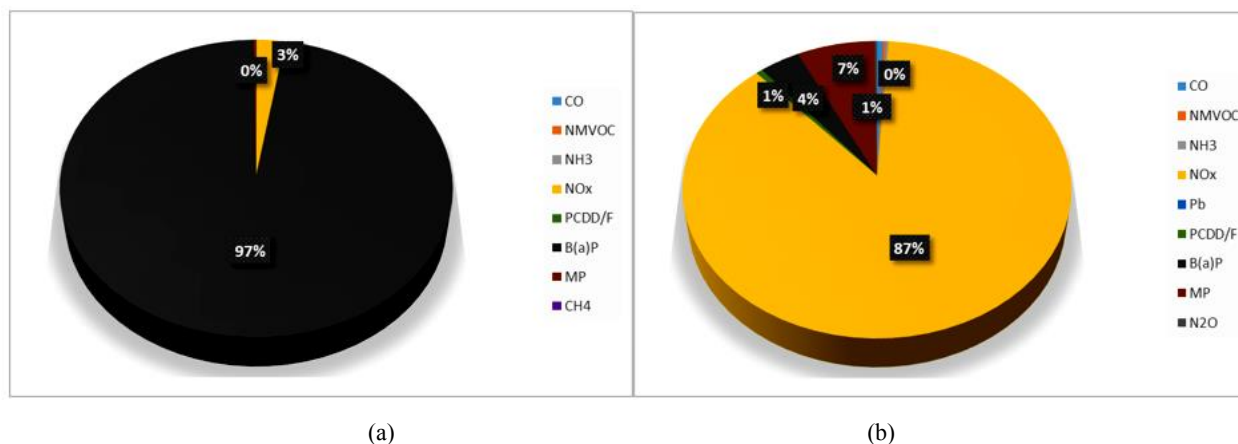


Fig. 4. Percentage ratio of the toxicities of the of individual components of the emissions from the diesel fuel combustion in the Odesa IUA for 2017, calculated using the Ukrainian (a) and European (b) methodologies

The obtained results prove the importance of taking into account PCDDs and PCDFs in state methodologies of calculating emissions from combustion of organic fuel from mobile sources, especially in the case of gasoline because for this fuel, according to European methods, PCDD and PCDF have 3rd priority, after NOx and CO.

According to the European methodology, the main pollutants formed during the combustion of liquid organic fuels are nitrogen oxides. However, there are some differences between the Ukrainian methodology and the EU methodology, in particular, in the calculation of benzopyrene emissions. In the case of the Ukrainian method, the reduced mass of benzopyrene is 97 % of all pollutants in the exhaust gases, which, in our opinion, is a very exaggerated value. The value of the emission factor given in the EU methodology and established experimentally is more possible. The high emission factor of benzopyrene can explain the small values of the reduced masses of other pollutants calculated for diesel fuel according to the Ukrainian methodology. In the case of the EU methodology, nitrogen oxides

remain the priority pollutants, particulate matter and benzopyrene have the 2nd and 3rd priorities, respectively, and PCDD and PCDF take the 5th priority. This is due to the fact that the combustion mode of diesel fuel prevents the access of oxygen, and the efficiency of diesel engines is higher than that of gasoline.

According to the obtained results, it is advisable to compare the environmental safety of available fossil fuel types. Due to the lack of initial information on the use of gaseous fuels by road transport in the Odesa IUA in 2017, we compared the relative specific emissions from combustion of 1 ton of each organic fuel using emission factors of relative hazards for these fuels. Note that the calculation was made according to the method (EMEP, 2019), which, based on the above calculations, is more accurate. Also, the calculation takes into account that the fuel consumption for diesel fuel is on average 20 % lower than gasoline (Favorit Motors, 2022), and gas consumption – 10 % higher than gasoline (Chelgas.ru, 2022). The results of the calculation are presented in Table 4.

Table 4

Specific emissions from the combustion of 1 ton of each organic fuel type using emission factors for diesel and gasoline fuels, liquefied and compressed gases

Pollutant	EF, kg/t				Emission, kg			
	diesel	gasoline	liq. gas	comp. gas	diesel	gasoline	liq. gas	comp. gas
CO	7.4	152.3	84.7	5.7	1.95	50.26	30.75	2.07
NMVOC	1.54	14.59	13.64	0.26	0.83	9.78	10.05	0.19
PM	1.52	0.02	0	0.02	24.32	0.40	0.00	0.44
NO _x	14.91	13.22	15.2	13	298.20	330.50	418.00	357.50
N ₂ O	0.056	0.186	0.089	n.a.	0.75	3.10	1.63	-
NH ₃	0.038	0.667	0.08	n.a.	0.76	16.68	2.20	-
B(a)P	1.58·10 ⁻⁵	4.2·10 ⁻⁶	2·10 ⁻⁶	n.a.	12.64	4.20	0.22	-
Pb	5.2·10 ⁻⁵	3.3·10 ⁻⁵	n.a.	n.a.	0.14	0.11	-	-
PCDD/F	1·10 ⁻¹⁰	2.2·10 ⁻⁰⁹	0	0	0.16	4.40	0.00	0.00

Note that the methodology does not allow to calculate the specific masses of pollutants such as N_2O , NH_3 and benzopyrene for compressed natural gas, as well as Pb for liquefied and compressed gases. It should also be noted that the emission factors for compressed gas in the methodology are based on a study of emissions

from freight bus transport. However, since the percentage of contribution to the toxicity of exhaust gases from these substances is relatively small, the results allow us to compare these fuels with each other. In Fig. 5, a comparative histogram of total specific emissions from combustion of different types of organic fuel is shown.

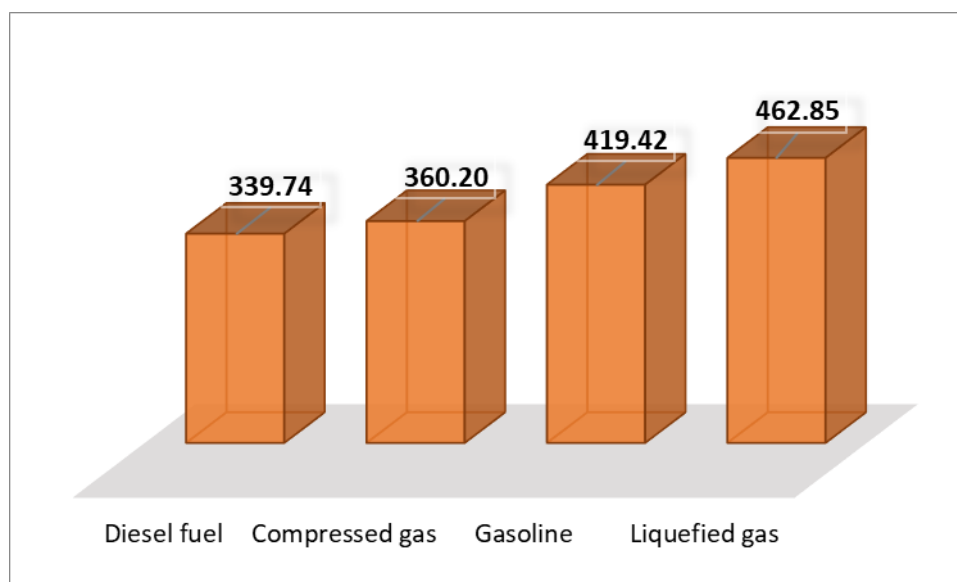


Fig. 5. Histogram of total specific emissions from combustion of different types of organic fuel

As can be seen from Fig. 5, the lowest total emissions are observed for diesel fuel and compressed natural gas combustion due to lower values of emission factors for NO_x . However, the choice of organic fuel should also take into account not only the total toxicity of emissions but also the emissions of each specific pollutant. However, for a more complete picture, it is necessary to have a methodology that contains updated emission factors and calculate the formation of all pollutants. Today, unfortunately, such a methodology does not exist in Ukraine.

4. Conclusions

As a result of research, the following conclusions can be drawn:

- In Ukraine, there are no effective mechanisms to encourage people to choose road transport with a more environmentally friendly type of internal combustion engine. There is virtually no methodological and legislative framework for Ukraine in monitoring air pollution and environmental taxation of mobile sources. Former methods, which are not valid, also do not allow an objective assessment of environmental pollution by road transport, and the quality of statistical information does not allow the use of new EU methodologies.

- When assessing the toxicity of exhaust fumes, it is proposed to use not the annual masses of

unintentionally formed POPs but to take into account their cumulative effect and half-life, which will provide more accurate results of POPs entry into the environment.

- Emissions of pollutants (including POPs) into the air basin from mobile sources for the Odesa IUA are calculated using Ukrainian and European methodologies. The relative masses of all pollutants are calculated using the relative hazard coefficient of pollutants, and the contribution of each pollutant to the toxicity of the exhaust fumes is estimated. A similar calculation is made according to national (Ukrainian) and European methodologies. The conclusion about the need to develop a national methodology was made.

- The main pollutants that make the greatest contribution to the toxicity of exhaust fumes from the combustion of diesel fuel and gasoline are nitrogen oxides. However, the development of new methodologies should consider the unintentional formation of PCDDs and PCDFs, which contribute significantly to emissions toxicity, especially in gasoline combustion, and provide their monitoring, which is obligatory for Ukraine as a part of the Stockholm Convention.

- Based on the values of pollutant emission factors, their total specific masses, which are formed during the combustion of diesel fuel, gasoline, liquefied and compressed gases, are compared. The lowest values were obtained for diesel fuel and compressed natural

gas, which indicates a greater environmental friendliness of these fuels compared to gasoline and liquefied gas.

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