Aggregated toxicity of road vehicles as basis for future regulation in the field of atmospheric air protection

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Abstract

This study presents ascertainment of priorities of automotive emission future state regulation based on analysis of the contribution of a comprehensive set of 64 known pollutants to aggregated toxicity of road vehicles with internal combustion engines powered by different fuels and electric vehicles as well. Aggregated mass emissions of pollutants are estimated for conventional 'Euro-0'–'Euro-6' vehicles and electric vehicles on a unified basis. In-use mass emission of pollutants and its relative toxicities that are established based on a ratio of the maximum permissible concentration of carbon monoxide and interested pollutant in the atmosphere is considered. Wear debris of pneumatic tires, road surface, and brake pads are also included. It has been discovered that aggregated mass emissions of modern:—cars with gasoline engines are only four times higher than the in-use emissions of electric vehicles;—cars with diesel engines are twice as high as those of cars with gasoline engines;—diesel-powered heavy-duty vehicles are only five times higher than the emissions of electric vehicles. The contribution of different types of contaminants to aggregated toxicity of road vehicles of different technologies is assessed. The results give essential substantiation and new ideas for future legislative regulation of emissions of various harmful substances by transport, also considering not yet regulated contaminants, and fuel quality standards as well. The results can also serve as the basis for an introduction of efficient and fair environmental labeling of vehicles that is directly proportional to the environmental damage in populated areas.

Keywords Road vehicles · Atmospheric pollution · Toxicity-weighting · Aggregated toxicity · Automotive emission regulation

1 Introduction

Many regions of the world, in efforts to struggle with a negative impact of road transport on human health, quality of life, and premature mortality in populated areas [1, 2], have implemented and continue to implement various and mostly incompatible systems of technical [3, 4], fiscal [5], and other types of harmful emissions regulation from road vehicles. These systems include first of all the establishment of maximum permissible limits of emission for certain types of hazardous substances, taxes, as well as differentiation of vehicle access to the road infrastructure, including fees for the usage of roads and the introduction

of so-called 'low emission zones' [6, 7] with limited access of 'environmentally dirty' cars.

However, while the emission standards become more and more severe, we should take into account also the following:

1. This leads to significant complications in design, lower reliability, and higher cost of vehicles production and operation.
2. The complexity of the design and a decrease in reliability negatively affect the retention of the established emission limits compliance during the operation of a vehicle (the result can be a more rapid deterioration

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in the environmental performance of modern vehicles compared to their predecessors, despite the current requirements for onboard diagnostic systems).

3. The rise in the cost of new, 'cleaner' vehicles can, for economic reasons, lead to a more extended operating period of old vehicles. The latter emits enormous amounts of harmful substances and still play a decisive role in overall urban air pollution despite an increased share of even pure electric vehicles in traffic.
4. Actual on-road emissions can significantly exceed the type-approval values obtained in a laboratory. This problem is known as 'off-cycle' emission. In this context, higher emission standards often in a greater extent, demonstrate rather virtual technological capabilities in idealized laboratory conditions that have little to do with actual pollution reduction.
5. The structure of emissions and the contribution of individual components to their general toxicity often change with new emission standards and related technologies.
6. Finally, modern 'environmentally friendly' technologies may even increase total emission of pollutants considering full life cycle assessment.

Thus, future emission standards for the automotive industry, as well as technical, fiscal and other types of regulation in this field, should be justified and set in some optimal way. Optimal emission standards for different pollutants should consider the cost of its further reduction-to-benefit ratio.

Theoretical limits of a further possible and justified reduction of the toxicity of vehicles with internal combustion engines are a subject of interest undoubtedly. It can be determined as 'in-use' emissions of pollutants with wear debris from electric vehicles, which are studied in the research too.

For instance, in accordance to [8], the increased popularity of electric vehicles will likely not have a significant effect on particulate matter (PM) levels since non-exhaust emissions already account for over 90% of PM_{10} and 85% of $PM_{2.5}$ emissions from traffic. Furthermore, non-exhaust PM in both fine and coarse sizes is rich in heavy metals [9].

Regulated and non-regulated emissions, including non-exhaust emissions due to abrasion of brakes, tires, and road surface were considered in [9]. Vehicles with internal combustion engines (ICE) are compared with electric vehicles (EV) utilizing such criteria and indicators as human toxicity, particulate matter formation, human health impacts. It is shown that the harmful impact of vehicles with modern petrol-powered ICE can be comparable to some extent with EVs given the criteria as mentioned above.

All the above mentioned, therefore proves the need for more grounded and reliable approaches to assessing of the environmental impact and therefore aggregated toxicity of road vehicles as a basis for future regulation in the field of atmospheric air protection.

To conduct a comprehensive assessment of the environmental impact of EVs and ICE vehicles [10, 11] examined full life cycle assessment (LCA). Brennan and Barder [11] covered the sourcing of raw materials, R and D, manufacturing and in-use, and the ultimate disposal of the vehicle and considering five secondary environmental impacts: human toxicity potential (HTP), terrestrial toxicity potential (TTP), freshwater toxicity potential (FTP), mineral depletion potential (MDP), and fossil fuel depletion potential (FFDP). It argued that EVs total environmental impact is a challenge.

However, [12] confirms the high complexity associated with exploiting of the HTP, as a quantitative toxic equivalency potential (TEP) that has been introduced to express the potential harm of a unit of chemical released into the environment in the full LCA.

Generally speaking, toxicity-weighting can be considered as a grounded, precise and reliable way to estimate aggregated toxicity and prioritize pollutants reduction strategy if the number of types of pollutants is large, as in the case of road vehicles. Toxicity-weighting is used by Risk-Screening Environmental Indicators (RSEI) Model by the United States Environmental Protection Agency [13]. Different pollutants aggregation functions are part of many air quality indexing (or air pollution indexing) systems around the world [14]. The analysis of toxicity-weighted emissions is the approach utilized in [15] as well.

However, the review [14] of different approaches to air quality indexing and methods of toxicity weighting, show that no standard practice today is widely recognized yet. Furthermore, road vehicles generally emit an extensive range of harmful contaminants. And, there are many unknowns regarding the data needed to implement reasonably the approaches mentioned above.

[16] gives an overview of four road vehicle environmental ranking methods covering 'Greenscore' in the US, 'Ecoscore' in Belgium, 'VCD Environmental Car list' in Germany, and 'Australian Green Vehicle Guide'. One drawback of mentioned methods is that the pollutants emissions effect is attenuated by considering 'standard' values instead of real emission values. Furthermore, the pollutants list is too limited. Weighting by different impact categories is highly subjective and cannot be considered as entirely justified.

Fortunately, in [17, 18] aggregated toxicity calculated merely as the sum of known pollutants, each multiplied by its relative toxicity coefficient derived as a carbon monoxide equivalent. Carbon monoxide is taken as reference as it

is a well-known poisoning substance and has, at the same time, one of the lowest toxicity in comparison with other 'automotive' pollutants. The toxicity coefficients are based on a comparison of the maximum permissible concentrations of pollutants in the atmospheric air and assumed as a ratio of maximum permissible concentrations of carbon monoxide to an interested pollutant.

In addition to setting emission limits for new vehicle types, an important and beneficial regulation area is the differentiation of vehicle access conditions to road infrastructure, aimed at limiting the use of environmentally 'dirty' vehicles.

Ideally, such distinction, if being fair and reasonable, should be applied in proportion to the amount of environmental damage. The development of approaches to assessing such harm and appropriate basis for vehicles labeling is also an essential direction of progress in this area.

However, for the time being, there is a severe lack of studies and literature related to this field. It is especially regarding a comprehensive analysis of the share of different contaminants to aggregated toxicity of road vehicles of different technologies of different years, respectively, emission standards.

The study aims to define the contribution of a comprehensive set of known pollutants to aggregated toxicity of road vehicles of different technological levels as a base to ascertain priorities of future state regulation in the field of atmospheric air protection.

The main objectives of the study are:

1. To determine an acknowledged and feasible method to estimate of average values of aggregated in-use toxicity of road vehicle.
2. To determine relative to carbon monoxide toxicity ratios of the most significant known pollutants.
3. To determine aggregated emissions of road vehicles of different technology levels.
4. To assess the contribution of different types of contaminants to aggregated toxicity of road vehicles of different technological levels.
5. To ascertain justified priorities of future state regulation in the field of automotive emission.

2 The method

A comprehensive estimation of total environmental damage due to ingredient pollution of road vehicles is a complicated task. It is due to a broad range of contaminants, their different ways and spread in the environment, complex chemical reactions occurring in the atmosphere, which, in turn, are also functions of many

factors and finally simultaneous exposition and action on the human body with many unknowns.

On practice, the task needs some simplification. In the first approach, total environmental damage can be considered merely as an aggregated toxicity of known today pollutants taking into account their mass emission and relative toxicity level of each.

In this study, aggregated emissions of ΣCO_{add} from the primary pollutants are calculated based on the coefficients of their relative toxicity (R_t) corresponding to carbon monoxide according to the approach outlined in [17, 18], which has become widely used

$$\Sigma CO_{add} = \sum_{i=1}^n m(i) \times R_t(i) \quad (1)$$

where ΣCO_{add} is aggregated emissions of significant pollutants (in factitious grams of carbon monoxide (CO), the toxicity (harmfulness) of which is taken as 1, $m(i)$ is mass emissions of i th pollutant, $R_t(i)$ is the coefficient of the relative toxicity (harmfulness) of the i th contaminant [relative to carbon monoxide (CO)], n is the total number of types of pollutants taken into account.

Mass emissions per unit of the mileage of a wide range of pollutants that are close to actual operational emissions (as opposed to 'Euro' limit values) can be calculated based on, for example, the method of the European Environment Agency [18, 19].

Source [19] contains 'in-use' emission factors for a broad range of contaminants as part of exhaust gases. Emission factors expressed in g/km per vehicle depending on:

- vehicle category;
- mass;
- engine volume;
- type of fuel;
- emission control technology with disaggregation on 'Euro' environmental standards;
- and many other factors, including traffic particularities, weather conditions as well.

In this study, the Tier 2 approach of [19] is used. It considers the average emission factors of different vehicle categories of different fuels and emission control technologies appointed to 'Euro' emission standards.

Emission factors of [19] can be considered as closer to real driving conditions sure than the results of certification tests utilizing strictly prescribed driving cycles and all the more so emission standards figures.

Source [20] contains 'in-use' emission factors for road vehicle tire, brake wear, and road surface wear expressed in g/km per vehicle depending on the vehicle category.

This method is widely used in many EU countries and other countries of the world for the inventory of pollutants by road transport.

Appropriate methods can also be used to calculate emissions of vehicles from the North American market and other non-European approved markets.

In this work, from the list of pollutants, taken into account in [19, 20], 64 known contaminants are used, an essential part of which is the most influential in terms of determining the total toxicity of emissions.

The descriptive statistics for the pollutants are already done in [19, 20]. It is included all the datasets about the mass emission of each pollutant from each source. Furthermore, this is a subject of different study going far away from the scope of this paper.

Source [19] contains specific emission factors in g/km for different weight classes of heavy-duty vehicles (HDV). Mass emission in g/km is heavily dependent on vehicle mass. In order to compare different heavy-duty vehicles with a broad range of masses on a unified system of coordinates, it is useful to use g/tkm unit instead of g/km. It is discovered that the best alignment of specific emission factors of [19] from g/km unit to a unified g/tkm system achieved using as divisor square root of HDV full mass. To do this, specific values of each weight class in g/km simply divided on a square root of HDV full mass average estimation.

It should be noted that toxicity ratio of PM here is differentiated on its source in the first approach, taking into account fuel type and non-exhaust sources as well. Furthermore, black carbon (BC) emission considered separately in the model.

Also, this is considered absorbed and adsorbed harmful substances emitted with PM based on existing knowledge about the PM content of different engine technologies and fuels. These substances expressed as 'TCOM' [toxic component of organic matter (OM)], and apparently, can be considered as the most dangerous component of suspended particles. TCOM consisted of adsorbed on a developed particle surface and absorbed by their liquid part of polycyclic aromatic hydrocarbons and persistent organic pollutants ('PAHs and POPs'), aldehydes, ketones, and aromatic hydrocarbons.

Emission factors of metals and PM are given in [19, 20] for various sources, covering ICE exhaust and non-exhaust. 'Non-exhaust' wear debris emissions considered tire, brake, and road pavement abrasion as well.

In this context, it should be noted that electric vehicles wear debris emissions would be a subject of more in-depth investigations in a further development of [20] method. In the first approach, electric vehicle, due to heavy batteries and bigger total weight, expected to emit more tire, brake and road pavement wear debris. But, electric vehicle (EV)

should lower to some extent brake wear thanks to energy recuperation feature.

To reflect further development of EV technology, a concept of hypothetical EV in which practically only an electric drive is used to brake in all (100%) modes, is proposed. Below it is marked as EV (CR) that means EV with 'Clean Recuperation' (CR). But, EVs wear debris investigation goes far beyond the study aim.

In general, the above-considered method to estimate road vehicle aggregated toxicity is quite transparent, simple to reproduce, and provides comprehensible results.

3 Relative to carbon monoxide toxicity ratios of significant pollutants

The coefficients of harmfulness (R_i) relative to carbon monoxide of 64 primary pollutants have been adopted. The toxicity ratios are assumed as the ratio of carbon monoxide maximum permissible concentration to maximum permissible concentration of another interested pollutant.

Data from [21–28] regarding the maximum permissible concentrations of pollutants in the atmospheric air of settlements and the air of the working area, as well as [18] data, were used.

For ease of use, pollutants are grouped into eight groups (Table 1).

Abovementioned contaminants had known emission factors derived in [19, 20] which is the criterion for their selection. It is believed that such comprehensive set of contaminants is quite enough to reflect if not the total aggregated toxicity of conventional road vehicles and EVs, then at least to compare different technologies on a more grounded and comprehensive basis than known before.

4 Results

Aggregated emissions of road vehicles of different technology levels are estimated using the method, described in part 2, on the base of widely recognized in-use emission factors of [19, 20] of 64 known primary pollutants and its relative harmfulness, as being adopted in part 3 above.

The results of the calculations are shown in graphical form on Figs. 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, and 13.

The following abbreviations are applied below to the names of the pollutants and their combinations (groups):

- 'AKA' is the total combined emissions of groups II (aldehydes), III (ketones), and IV (aromatics);
- 'TSP' is aggregate suspended particles of Group VI;
- 'TCOM' is the designation of the most dangerous, apparently, a component of suspended particles, con-

Table 1 Relative to carbon monoxide (CO) toxicity ratios (R_t) of 64 significant pollutants

Group	Chemical formula or denotation	Name	R_t
Group I (the bulk of the exhaust emissions)	CO	Carbon monoxide	1
	CO ₂	Carbon dioxide	0.0022
	NO _x	Oxides of nitrogen	75
	N ₂ O	Nitrous oxide	188
	NH ₃	Ammonia	75
	'LHC'	'Light hydrocarbons'	3.16
Group II (aldehydes)	CH ₂ O/H ₂ CO	Formaldehyde	1000
	C ₂ H ₄ O/CH ₃ CHO	Acetaldehyde	300
	C ₃ H ₄ O/CH ₂ =CHCHO	Acrolein	100
	C ₇ H ₆ O/C ₆ H ₅ CHO	Benzaldehyde	50
	C ₄ H ₆ O/CH ₃ CH=CHCHO	Crotonaldehyde	5
	C ₄ H ₆ O/CH ₂ =C(CH ₃)	Methacrolein	5
	C ₄ H ₈ O/CH ₃ (CH ₂) ₂ CHO	Butyraldehyde	5
	C ₄ H ₈ O/(CH ₃) ₂ CHCHO	Isobutanaldehyde	5
	C ₃ H ₆ O/CH ₃ CH ₂ CHO	Propionaldehyde	5
	C ₆ H ₁₂ O/CH ₃ (CH ₂) ₄ CHO	Hexanal	5
	C ₅ H ₁₀ O/CH ₃ (CH ₂)	<i>i</i> -Valeraldehyde	5
	C ₅ H ₁₀ O/CH ₃ (CH ₂)	Valeraldehyde	5
	C ₈ H ₈ O/CH ₃ C ₆ H ₄ CHO	<i>o</i> -Tolualdehyde	5
	C ₈ H ₈ O/CH ₃ C ₆ H ₄ CHO	<i>m</i> -Tolualdehyde	5
	C ₈ H ₈ O/CH ₃ C ₆ H ₄ CHO	<i>p</i> -Tolualdehyde	5
Group III (ketones)	C ₃ H ₆ O/CH ₃ -CO-CH ₃	Acetone	8.57
	C ₄ H ₈ O/CH ₃ -C-CH ₂ -CH ₃	Methylethylketone	5
Group IV (aromatics)	C ₇ H ₈ /C ₆ H ₅ CH ₃	Toluene	5
	C ₈ H ₁₀ /C ₆ H ₅ C ₂ H ₅	Ethylbenzene	150
	C ₈ H ₁₀ /C ₆ H ₄ (CH ₃) ₂	<i>m,p</i> -Xylene	15
	C ₈ H ₁₀ /C ₆ H ₄ (CH ₃) ₂	<i>o</i> -Xylene	15
	C ₉ H ₁₂ /C ₆ H ₃ (CH ₃)	1,2,3 trimethylbenzene	100
	C ₉ H ₁₂ /C ₆ H ₃ (CH ₃) ₃ (asym.)	1,2,4 trimethylbenzene	200
	C ₉ H ₁₂ /C ₆ H ₃ (CH ₃) ₃ (sym.)	1,3,5 trimethylbenzene	300
	C ₈ H ₈ /C ₆ H ₅ CH=CH ₂	Styrene	1500
	C ₆ H ₆	Benzene	30
	C ₉	C ₉	5
	C ₁₀	C ₁₀	10
	C > 13	C > 13	20
Group V (PAHs & POPs)	C22H12/C22H10O2/ID(1,2,3,cd)P	Indeno(1,2,3-cd)pyrene	1,500,000
	C20H12/C20H10O2/B(k)F	Benzo(k)fluoranthene	3,000,000
	C20H12/B(b)F	Benzo(b)fluoranthene	3,000,000
	C20H12/B(a)P	Benzo(a)pyrene	3,000,000
Group VI (particulate matter (PM))	'BC'	Black carbon	41.5
	PM _{2.5} petrol	PM originated from petrol engines exhaust gases	300
	PM _{2.5} diesel	PM originated from diesel engines exhaust gases	200
	PM _{2.5} tire	PM originated from pneumatic tire wear debris	100
	PM _{2.5} brake	PM originated from brake wear debris	150
	PM _{2.5} road	Pm originated from road surface wear debris	50
Group VI (sulfur compounds)	SO ₄	Sulfates	30
	SO ₂	Sulfur dioxide	22

Table 1 (continued)

Group	Chemical formula or denotation	Name	R_t
Group VIII (metals and its compounds (MS))	Cd	Cadmium	2,000
	Cu	Copper	40
	Cr	Chromium	1000
	Ni	Nickel	4000
	Se	Selenium	200
	Zn	Zinc	40
	Hg	Mercury	4000
	As	Arsenic	2000
	Fe	Iron	75
	Mg ₂₊	Magnesium ion	60
	Mo	Molybdenum	150
	Sb	Antimony	150
	Si	Silicon	60
	Sn	Tin	150
	Ti	Titanium	20
	Pb	Lead	400
	C ₈ H ₂₀ Pb	Tetraethyllead	224,000

Fig. 1 Aggregated pollutants emission of petrol-powered passenger cars and EVs (g/km)

Fig. 2 Aggregated pollutants emission of diesel-powered passenger cars and EVs (g/km) **Fig. 3** Aggregated pollutants emission of diesel-powered heavy-duty vehicles and EVs (g/tkm)

sisting of adsorbed on a developed particle surface and absorbed by their liquid part of polycyclic aromatic hydrocarbons and persistent organic pollutants ('PAHs and POPs'), aldehydes, ketones, and aromatic hydrocarbons;

- 'Exh' is a supplement to the designation of particles whose source of origin is exhaust gases of engines;
- 'SD (TBR)' is the aggregate emissions with wear debris of a pneumatic tire, road pavement and brake pads fol-

Fig. 4 Contribution (%) of different pollutants to aggregated toxicity of 'Euro-2' passenger cars with petrol engines

Fig. 7 Contribution (%) of different pollutants to aggregated toxicity of 'Euro-6' passenger cars with compressed ignition engines

Fig. 5 Contribution (%) of different pollutants to aggregated toxicity of 'Euro-6' passenger cars with petrol engines

Fig. 8 Contribution (%) of different pollutants to aggregated toxicity of 'Euro-2' city buses with compressed ignition engines

Fig. 6 Contribution (%) of different pollutants to aggregated toxicity of 'Euro-2' passenger cars with compressed ignition engines

Fig. 9 Contribution (%) of different pollutants to aggregated toxicity of 'Euro-6' city buses with compressed ignition engines

Fig. 10 Contribution (%) of different pollutants to aggregated toxicity due to tire, road, and brake pads wear debris

lowing components: total suspended particles (TSP), polycyclic aromatic hydrocarbons and persistent organic pollutants (PAHs and POPs), and metals and their compounds (MS).

The charts contain the following abbreviations regarding the type of vehicle and fuel:

- 'PC' is passenger cars;
- 'EV' is electric vehicles;

Fig. 11 Contribution (%) of different pollutants to aggregated toxicity of petrol-powered cars depending on the 'Euro' class

- 'EV (CR)' is electric vehicles with clean recuperation. That is meaning electric vehicles of the next generation with close to 100% utilization of the energy recovery system during braking. The abbreviation '(CR)' means here 'Clean Recuperation' of a hypothetical wheeled vehicle of a future in which practically only an electric drive is used to brake in all (100%) modes. Thus, there is no brake wear debris emission. Sure, current EV has recuperation, but still emit brake wear debris to some extent. Definition of 'Clean Recuperation' used in the paper to reflect further development of EV technology;
- 'LDV' is light-duty vehicles;
- 'HDV' is heavy-duty vehicles, including high capacity city buses;
- 'P' is petrol (gasoline);
- 'D' is diesel oil.

4.1 Aggregated emissions of road vehicles of different technology levels

Figures 1 and 2 show aggregated emissions (ΣCO_{add}) in g/km of relatively new (within 30,000–60,000 km mileage) passenger cars with internal combustion engines of the environmental classes 'Euro-0'–'Euro-6' and electric vehicles.

Consolidated total emissions of modern 'Euro-5' and 'Euro-6' passenger cars with petrol engines (Fig. 1) are only about four times higher than aggregated emissions of current electric vehicles (it is assumed that the latter consist of a pneumatic tire, road, and brake wear debris).

Fig. 12 Contribution (%) of different pollutants to aggregated toxicity of diesel-powered cars depending on the 'Euro' class

Fig. 13 Contribution (%) of different pollutants to aggregated toxicity of diesel-powered urban buses depending on the 'Euro' class

Used on Fig. 1, classes 'Euro-6+' and 'Euro-6++' are not connected to petrol-powered passenger cars, and shown for direct comparison of the data with diesel-powered cars on Fig. 2. Here, 'Euro-6', 'Euro-6+', 'Euro-6++' designations are used for simplification and meant 'Euro-6b', 'Euro-6c' + 'Euro-6d-temp', 'Euro-6d' classes, respectively.

The small increase in average aggregated emissions is due to grows of the share of gasoline direct injection technology that is lead to PM emission side effect.

Aggregated emissions of 'Euro-6++' cars with diesel engines (Fig. 2) are twice as high as those of 'Euro-5'–'Euro-6' cars with petrol engines (Fig. 1).

Figure 3 shows aggregated emissions ($\Sigma\text{CO}_{\text{add}}$) in g/tkm by diesel-powered heavy-duty vehicles (HDV). It is calculated average aggregate specific emissions per tonne of a total mass of a vehicle. Modern 'Euro-6' HDVs emit on average only about five times more aggregated emissions than pure electric transport.

Figures 1, 2, and 3 gives a clear, simplified representation of aggregated toxicity of conventional road vehicles of different 'Euro' classes, powered by petrol and diesel fuel, and EVs, which speaks for itself.

4.2 The contribution of different types of pollutants to aggregated toxicity of road vehicles of different technological levels

The contribution of different types of pollutants to aggregated toxicity of road vehicles of different technological levels is a subject of great interest. It is desired to design more substantiated future emission standards for the automotive industry, also considering not yet regulated contaminants, and fuel quality standards as well.

Figures 4, 5, 6, 7, 8, 9, and 10 give a representation of the defined contribution of different kinds of contaminants to the aggregated toxicity of road vehicles of different technological levels. The numbers near pollutant's name mean the contribution in %.

Figures 4 and 5 show the contribution (%) of contaminants to aggregated emissions of passenger cars according to the 'Euro-2' and 'Euro-6' levels with petrol engines.

Figures 6 and 7 show the contribution (%) of pollutants to aggregated emissions of passenger cars according to the 'Euro-2' and 'Euro-6' levels with diesel engines.

Figures 8 and 9 show the contribution (%) of pollutants to aggregated emissions of city buses according to the 'Euro-2' and 'Euro-6' levels with diesel engines.

Figures 4, 5, 6, 7, 8, and 9 provide an overview of the fundamental differences in the distribution of the contribution of different pollutants to the aggregate total toxicity of 'Euro-2' and 'Euro-6' vehicles.

Figure 10 shows the contribution (%) of pollutants to aggregated emissions of a pneumatic tire, road pavement, and brake pads wear.

Figures 11, 12, and 13 show the contribution change of different pollutants to the total toxicity of correspondingly passenger cars with petrol engines, passenger cars with diesel engines and for city buses with diesel engines of different environmental classes.

Below is an analysis of the contribution of different types of pollutants to the total aggregated toxicity of vehicles of different technological levels.

4.2.1 The contribution of carbon dioxide (CO_2)

The main part of the gross mass emissions from the exhaust gases of internal combustion engines is in the group I of gaseous components (Table 1), in which CO_2 is dominated sure. Despite the shallow (0.0022) CO_2 relative harmful factor (which is 455 times less than CO), its high gross emissions (the lion's share of fuel combustion products account for CO_2) lead to a noticeable toxic effect in large cities.

The contribution of CO_2 emissions to the total aggregated toxicity during the last two decades is raised from 0.2 to 0.9% for diesel-powered and from 0.8 to 2.3% for gasoline-powered vehicles. Here, more significant values naturally correspond to a higher environmental class of vehicles. It is a result of lowering other pollutants mass emissions share. It proves the expediency to take into account CO_2 not only as a greenhouse gas, but also as a toxic pollutant to some extent.

4.2.2 The contribution of nitrogen oxides (NO_x)

Nitrogen oxides (NO_x) contribute significantly to aggregated toxicity.

Contribution of NO_x of petrol-powered cars decreased from 77% for 'Euro-0' (Fig. 11), then to 35% for 'Euro-2' (Figs. 4, 11), and to 24% for 'Euro-6' (Figs. 5, 11).

Contribution of NO_x of diesel-powered cars was 24% for 'Euro-0' (Fig. 12) due to high share of PM-products, then was increased to about 1/2 for 'Euro-2'–'Euro-6', and becoming significantly lower (to about 30%) only for 'Euro-6++' (Figs. 6, 7, 11).

Contribution of NO_x of diesel-powered city buses was 68% for 'Euro-0' (Fig. 13), then 79% for 'Euro-2' (Fig. 8), and becoming significantly lower (to about 31%) only for 'Euro-6' (Figs. 8, 9, 13).

4.2.3 The contribution of aldehydes, ketones, and aromatic hydrocarbons

The emissions of aldehydes and ketones together with the emissions of aromatic hydrocarbons (respectively, Groups II, III and IV) can make a significant (3–18% for diesel and 19–26% for petrol engines) contribution to the total toxicity (Figs. 4, 5, 6, 7, 8, 9, 10, 11, 12, 13).

Its share has shown principal differences for city buses of 'Euro-4' and 'Euro-5' vs. 'Euro-6' classes' (Fig. 13). Probably it can be seen as the non-desirable result of modern selective catalytic reduction technology.

4.2.4 The contribution of polycyclic aromatic hydrocarbons and persistent organic pollutants

Particularly, dangerous is the emissions of polycyclic aromatic hydrocarbons and persistent organic pollutants (Group V).

Their 'contribution' for petrol engines increases from 4% for 'Euro-0' (Fig. 11), then to 6% for 'Euro-2' (Figs. 4, 11), and to 18% for 'Euro-6' (Figs. 5, 11).

It increases for diesel-powered passenger cars from 11% for 'Euro-0' (Fig. 11), to 16% for 'Euro-2' (Figs. 6, 12), and to 42% for 'Euro-6' (Figs. 7, 12).

It increases for diesel-powered city buses from 2% for 'Euro-0' (Fig. 13), to 4% for 'Euro-2' (Figs. 8, 13), and to 27% for 'Euro-6' (Figs. 9, 13).

4.2.5 The contribution of particulate matter

Particulate matter (PM) emissions (Group VI) from various sources, especially from diesel (up to 27% for 'Euro-0' and 10% for 'Euro-2', for example) also contribute significantly to the overall toxicity past years. Nevertheless, 'Euro-6' diesel-powered vehicles demonstrate negligible small share of PM.

'Euro-6' diesel-powered vehicles demonstrate a small share of PM (and even TCOM for HDVs) based on mass emission measurement. Notwithstanding above mentioned, actual PM toxicity, including particles number (PN), finest particulates toxicity, and its real contribution to aggregated toxicity should be a subject for further research sure.

4.2.6 The contribution of sulfur compounds

Emissions of sulfur compounds (Group VII) and metals (Group VIII) do not have a decisive effect (up to 0.1% of the contribution) however are essential methodologically from many reasons as it is discussed in Sect. 6.

4.2.7 The contribution of wear debris

Emissions of Group VI, VII, VIII and V pollutants with pneumatic tire, road pavement, and brake pads wear products make an essential contribution to the overall toxicity of high-class ('Euro-5' and 'Euro-6') environmental vehicles (up to 16–25%). It also takes into account emissions from electric vehicles that are not actually 'zero-emission' vehicles, as previously thought.

The justified priorities of future state regulation in the field of atmospheric air protection from road vehicles pollution can be based on the research results and layout below.

5 Discussion

The method used for the estimation of aggregated toxicity of road vehicle can be considered as quite feasible, being grounded on previous knowledge.

The coefficients of relative toxicity for the 64 types of the most significant and investigated pollutants, sources of emissions of which are road transport, have been established. They can also be used as reference data in further research in this area.

The list of contaminants taken into account allows for an objective assessment of the total toxicity of vehicles of different technological levels, including comparing vehicles with internal combustion engines powered by different types of fuel and electric vehicles as well.

It can be used in the same manner for vehicles that have no 'Euro' designations, including vehicles imported to European countries from North America. In such a way, vehicles from different markets, that are being certified within incompatible systems, can be considered in a common (universal) system of coordinates for state regulation in a country.

The ordinal numbers of the 'Euro' standards sure characterize only the successive stages of the implementation of increasingly stringent environmental requirements. Vehicles of the same 'Euro' level, but using different types of fuel and engines, can differ significantly in aggregated toxicity.

Aggregated emission per unit of mileage is a more strong base for the implementation of environmental hazard labeling road vehicles that is directly proportional to the environmental damage.

The results obtained make it possible to do some conclusions and determine priorities in the further legislative regulation of emissions of various harmful substances by transport:

1. Nitrogen oxides (NO_x) is still a significant but already not most decisive subject to establish more stringent emission norms. Its contribution to aggregated toxicity shows the tendency for a decrease.
2. Emissions of particularly dangerous polycyclic aromatic hydrocarbons (PAH) and persistent organic pollutants (POPs), being added by aldehydes, ketones, and aromatic hydrocarbons, becoming the most important subject for future vehicle's emission standards as well as fuel quality standards.
3. Although the latest 'Euro-6' diesel-powered vehicles demonstrate negligible small share of PM, at first sight, its emission should be further restricted. The reason is the content of the most dangerous components of suspended particles, consisting of adsorbed on a

developed particle surface and absorbed by their liquid component of PAHs, POPs, aldehydes, ketones, and aromatic hydrocarbons. The gasoline direct injection technology should also be the subject for further regulation of PM, taking into account nanoparticles as well.

4. Nitrous oxide (N_2O) becoming noticeable component for diesel engines.
5. Further lowering of sulfur content in fuels is probably meaningless given direct sulfur compounds emission, but needs further investigation regarding PM formation processes, as well as exhaust gases after-treatment devices and fuel equipment performance and durability issues. Sulfur content in fuels and acid precipitation issue should be further investigated sure.
6. The aggregate emissions with wear debris of a pneumatic tire, road pavement and brake pads becoming an essential factor. Potential of lowering these emissions is an open question.
7. The aggregate emissions with wear debris are crucial that can be considered as the lower limit of theoretically possible lowering of aggregated emission of vehicles with internal combustion engines. Therefore, it serves as a background of future emission standards substantiation to make them reasonably adequate or sufficient.

In general, Figs. 4, 5, 6, 7, 8, 9, 10, 11, 12, and 13 provide inputs and insights into substantiated reduction strategy for various pollutants to reduce aggregated total toxicity of modern 'Euro-5' and 'Euro-6' vehicles, future vehicles, and vehicles of prior technological levels that are still in use.

At the same time, the results obtained for relatively new vehicles, that is within the initial range of 30–60 thousand kilometers [19]. However, in operation for a considerable time emission can significantly exceed these values [29]. It should be a subject of additional research and the main direction of development of the method of calculation of aggregated emissions of pollutants of vehicles in operation.

The study focused on in-use emissions of all categories of road vehicles in populated areas. Total life cycle assessment (LCA) combining the presented approach should be addressed in further investigations.

6 Conclusions

In this paper, the contribution of a comprehensive set of known pollutants to aggregated toxicity of road vehicles of different technological levels is studied as a base to ascertain of priorities of future state regulation in the field of atmospheric air protection.

The acknowledged and feasible method to estimate of average values of aggregated in-use toxicity of road vehicle is determined.

Relative to carbon monoxide toxicity ratios of the most significant known pollutants is determined.

Aggregated emissions of road vehicles of different technology levels are determined.

Aggregated mass emissions of pollutants, both from traditional vehicles with internal combustion engines and electric vehicles, including wear debris of a pneumatic tire, road pavement, and brakes are defined in a common coordinate system. Aggregated emissions reflect the average aggregate degree of damage that a vehicle causes to the environment through ingredient contamination in populated places.

It has been discovered that:

- aggregated total mass emissions of passenger cars of level 'Euro-5' and 'Euro-6' with gasoline engines are only four times higher than the emissions of electric vehicles;
- aggregated total mass emissions of 'Euro-6++' passenger cars with diesel engines are twice as high as those of 'Euro-5' and 'Euro-6' cars with gasoline engines;
- aggregated mass emissions of diesel-powered heavy-duty vehicles of the 'Euro-6' level are on average only five times higher than the emissions of electric vehicles.

Justified priorities of future state regulation in the field of automotive emission are elaborated in Sect. 5.

It can be useful to design more substantiated future emission standards for the automotive industry, also considering not yet regulated contaminants, and fuel quality standards as well.

The emissions of the wear debris mentioned above thus determine the lower limit (or theoretical potential) for further reduction of aggregated toxicity of internal combustion engines, that is, about 3–5 g/km sum of reduced to carbon monoxide ($\Sigma\text{CO}_{\text{add}}$) pollutants for passenger cars and about 3–6 g/tkm of $\Sigma\text{CO}_{\text{add}}$ for heavy-duty vehicles.

Furthermore, a new coordinate system has been obtained that can offer general principles for the development and subsequent implementation of a universal approach to environmental hazard labeling of road vehicles and low emission zones (LEZs) access rules. Here, 3–5 g/km for PC and LDV (and 3–6 g/tkm for HDV) is the lowest value corresponds to pure electric propulsion technology. ICE-powered vehicles occupy higher values, spreading as it is presented on Figs. 1, 2, and 3, depending on the initial 'Euro' standard compliance. Both vehicles and (LEZs) can use aggregated toxicity as environmental hazard level (EHL) values grouped in several classes as a common system. Such an approach is technology-neutral and

combines in unified coordinates vehicles of different markets approved within incompatible emission standards. Moreover, EHL can be dynamic to reflect vehicle environment properties degradation in time. It gives principally new opportunities to improve the efficiency of vehicle access restriction schemes in cities and can serve as a basis for future fiscal policy as well.

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Compliance with ethical standards

Conflict of interest The author declares that he has no conflict of interest.

References

- Connolly R, Pierce G, Gattaciecchia J, Zhu Y (2020) Estimating mortality impacts from vehicle emission reduction efforts: the Tune In and Tune Up program in the San Joaquin Valley. *Transp Res Part D* 78:102190. <https://doi.org/10.1016/j.trd.2019.11.019>
- Barnes JH, Chatterton TJ, Longhurst JWS (2019) Emissions vs. exposure: increasing injustice from road traffic-related air pollution in the United Kingdom. *Transp Res Part D Transp Environ* 73:56–66. <https://doi.org/10.1016/j.trd.2019.05.012>
- CPT Group GmbH (2019) Worldwide emission standards and related regulations: passenger cars/light and medium duty vehicles. <https://www.continental-automotive.com/getattachment/8f2dedad-b510-4672-a005-3156f77d1f85/EMISSIONBOOKLET2019.pdf>. Accessed 29 Aug 2020
- Nesbit M, Fergusson M, Colsa A, Ohlendorf J, Haves C, Paquel K, Schweitzer J-P (2016) Comparative study on the differences between the EU and US legislation on emissions in the automotive sector. European Parliament Directorate-General for internal policies. Institute for European Environmental Policy. [https://www.europarl.europa.eu/RegData/etudes/STUD/2016/587331/IPOL_STU\(2016\)587331_EN.pdf](https://www.europarl.europa.eu/RegData/etudes/STUD/2016/587331/IPOL_STU(2016)587331_EN.pdf). Accessed 29 Aug 2020
- ACEA Tax Guide (2019) https://www.acea.be/uploads/news_documents/ACEA_Tax_Guide_2019.pdf. Accessed 29 Aug 2020
- Low Emission Zones. ACEA position paper (2015) https://www.acea.be/uploads/publications/ACEA_LEZ_Position_May_2015.pdf. Accessed 30 Aug 2020
- Amundsen AH, Sundvor I (2018) Low emission zones in europe requirements, enforcement and air quality. TØI report 1666/2018. <https://www.toi.no/getfile.php?mmfileid=49204>. Accessed 10 Jan 2020
- Timmers VRJH, Achten PAJ (2016) Non-exhaust PM emissions from electric vehicles. *Atmospheric environment* 134. <http://www.soliftec.com/NonExhaust%20PMs.pdf>. Accessed 29 Aug 2020
- Pant P, Harrison RM (2013) Estimation of the contribution of road traffic emissions to particulate matter concentrations from field measurements: a review. <https://doi.org/10.1016/j.atmosenv.2013.04.028>. Accessed 30 Aug 2020
- Hoofman N, Oliveira L, Messagie M, Coosemans T, Mierlo V (2016) Environmental analysis of petrol, diesel and electric passenger cars in a belgian urban setting. *J Energies* 9:84. <https://doi.org/10.3390/en9020084>
- Brennan JW, Barder TE (2017) Battery electric vehicles vs. internal combustion engine vehicles: a united states-based comprehensive assessment. Arthur D. Little 2016. <https://www.adlittle.com/en/insights/viewpoints/battery-electric-vehicles-vs-internal-combustion-engine-vehicles>. Accessed 29 Aug 2020
- McKone TE, Hertwich EG (2001) The human toxicity potential and a strategy for evaluating model performance in life cycle impact assessment. *Int J LCA* 6:106–109. <https://doi.org/10.1007/BF02977846>
- Risk-Screening Environmental Indicators (RSEI) Model. US EPA (2020). <https://www.epa.gov/rsei/rsei-toxicity-weights>. Accessed 29 Aug 2020
- Kanchan K, Gorai AK, Goyal P (2015) A review on air quality indexing system. *Asian J Atmos Environ* 9–2:101–113. <https://doi.org/10.5572/ajae.2015.9.2.101>
- Waste: A Handbook for Management (2011) Edited by Trevor Letcher, Daniel Valerio. Elsevier Inc. ISBN 978-0-12-381475-3. <https://searchworks.stanford.edu/view/9518829>. Accessed 29 Aug 2020
- Marques S et al (2014) Eco-rating methodologies for private cars: driving cycle influence. *Proc Soc Behav Sci* 111:682–691. <https://doi.org/10.1016/j.sbspro.2014.01.102>
- Vremennaya tipovaya metodika opredeleniya e'konomicheskoy e'ffektivnosti osushhestvleniya prirodookhrannykh meropriyatij i ocenki e'konomicheskogo ushherba, prichinyaemogo narodnomu khozyajstvu zagryazneniem okruzhayushhej sredy. A.S.By'strov, V.V.Varankin, M.A.Vilenskij i dr. M.: E'konomika, 1986.–96 s (Временная типовая методика определения экономической эффективности осуществления природоохранных мероприятий и оценки экономического ущерба, причиняемого народному хозяйству загрязнением окружающей среды. А.С.Быстров, В.В.Варанкин, М.А.Виленский и др. М.: Экономика, 1986.–96 с.). <https://files.stroyinf.ru/Data2/1/4293750/4293750177.pdf>. Accessed 29 Aug 2020
- Gutarevych YuF, Zerkalov DV, Hovorun AH, Korpach AO, Merzhyevska LP (2006) *Ekologiya ta avtomobilnyi transport*. Kyiv, 292. (Гутаревич Ю. Ф., Зеркалов Д. В., Говорун А. Г., Корпач А. О., Мержиевська Л. П. *Экологія та автомобільний транспорт*, К.—2006.—292 с.). https://library.kre.dp.ua/Books/2-4%20kurs/Osnovi%20ekologii/Gutarevich_Yu_F_Ekologiya_avtomobilnogo_transportu_2002%20.pdf. Accessed 29 Aug 2020
- EMEP/EEA Air pollutant emission inventory guidebook 2019: passenger cars, light commercial trucks, heavy-duty vehicles including buses and motorcycles. <https://www.eea.europa.eu/publications/emep-eea-guidebook-2019/part-b-sectoral-guidance-chapters/1-energy/1-a-combustion/1-a-3-b-i/view>. Accessed 29 Aug 2020
- EMEP/EEA Air pollutant emission inventory guidebook 2019: road transport: automobile tyre and brake wear, automobile road abrasion. <https://www.eea.europa.eu/publications/emep-eea-guidebook-2019/part-b-sectoral-guidance-chapters/1-energy/1-a-combustion/1-a-3-b-vi/view>. Accessed 29 Aug 2020
- GN 2.1.6.695-98 (1998) Predel'no dopustimye kontsentratsii (PDK) zagryaznyayushchih veshchestv v atmosfornom vozduhe naselennykh mest (ГН 2.1.6.695-98 'Предельно допустимые концентрации (ПДК) загрязняющих веществ в атмосферном воздухе населенных мест'). <http://docs.cntd.ru/document/1200004633>. Accessed 29 Aug 2020
- GN 2.1.6.3492-17 (2017) Predel'no dopustimye kontsentratsii (PDK) zagryaznyayushchih veshchestv v atmosfornom vozduhe gorodskih i sel'skih poseleniy (ГН 2.1.6.3492-17 'Предельно допустимые концентрации (ПДК) загрязняющих веществ в атмосферном воздухе городских и сельских

- поселений'). <http://docs.cntd.ru/document/556185926>. Accessed 29 Aug 2020
23. GN 2.2.5.3532-18 (2018) Predel'no dopustimye kontsentratsii (PDK) vrednykh veshchestv v vozduhe rabochey zony (ГН 2.2.5.3532-18 'Предельно допустимые концентрации (ПДК) вредных веществ в воздухе рабочей зоны'). <http://docs.cntd.ru/document/557235236> Accessed 29 Aug 2020
 24. GOST 12.1.005-88 (1988) Sistema standartov bezopasnosti truda (SSBT). Obshchie sanitarno-gigienicheskie trebovaniya k vozduhu rabochey zony (s izmeneniem No. 1) (ГОСТ 12.1.005-88 'Система стандартов безопасности труда (ССБТ). Общие санитарно-гигиенические требования к воздуху рабочей зоны (с Изменением № 1)'). <http://docs.cntd.ru/document/1200003608>. Accessed 29 Aug 2020
 25. Hranychno dopustymi kontsentratsiyi khimichnykh i biolohichnykh rechovin v atmosferному povitri naselenykh mist. Zatverdzeni v 2015 r. t.v.o. holovnoho derzhavnoho sanitarnoho likaria Ukrainy S.V. Protasom. ('Гранично допустимі концентрації хімічних і біологічних речовин в атмосферному повітрі населених місць', затверджені в 2015 р. т.в.о. головного державного санітарного лікаря України С.В. Протасом.—19 с.). http://online.budstandart.com/ua/catalog/doc-page.html?id_doc=81980. Accessed 29 Aug 2020
 26. DSP-201-97 (1997) Derzhavni sanitarni pravyla okhorony atmosferного povitria naselenykh mist (vid zabrudnennia khimichnuyu ta biolohichnuyu rechovinamy); zatverdzeni nakazom Ministerstva okhorony zdorovia Ukrainy vid 9 lyupnia 1997 r. No. 201. (ДСП-201-97 'Державні санітарні правила охорони атмосферного повітря населених місць (від забруднення хімічними та біологічними речовинами)', затверджені наказом Міністерства охорони здоров'я України від 9 липня 1997 р № 201.). <https://zakon.rada.gov.ua/rada/show/v0201282-97#Text>. Accessed 29 Aug 2020
 27. Hranychno dopustymi kontsentratsii (HDK) ta oriientovni bezpechni rivni diiannia (OBRD) zabrudniuiuchykh rechovin v atmosferному povitri naselenykh mist (Гранично допустимі концентрації (ГДК) та орієнтовні безпечні рівні діяння (ОБРД) забруднюючих речовин в атмосферному повітрі населених місць. Підготовлено Управлінням атмосферного повітря Мінекоресурсів України (головний спеціаліст Трофименко Н.С.), Українським науковим центром технічної екології (співробітники лабораторії № 13), Головним санітарно-епідеміологічним Управлінням Міністерства охорони здоров'я України (головний спеціаліст Карпенко Т.Ф.), Комітетом з питань гігієнічної регламентації МОЗ України (Горбань Л.М.) та Українським науковим гігієнічним центром (зав. лабораторією гігієни атмосферного повітря д.м.н. Присяжнюк В.Є., н.с. Федоришин О.П.). 2017.—с. 42) <https://netref.ru/granichno-dopustimi-kontsentratsiyi-gdk-ta-oriientovni-bezpechni.html>. Accessed 29 Aug 2020
 28. Hranychno dopustymi kontsentratsii khimichnykh i biolohichnykh rechovin v atmosferному povitri naselenykh mist. Approved by the order of the Ministry of Health of Ukraine dated January 14, 2020 No. 52 (Гранично допустимі концентрації хімічних і біологічних речовин в атмосферному повітрі населених місць, затверджені наказом Міністерства охорони здоров'я України від 14 січня 2020 року № 52). <https://zakon.rada.gov.ua/laws/show/z0156-20#Text>. Accessed 29 Aug 2020
 29. Klymenko O, Ustyomenko V, Kolobov K, Rychok S, Hora M, Naumenko N (2019) Analysis of emissions in the European driving cycle of used light-duty vehicles imported to Europe from North America. SAE Int J Sust Trans Energy Environ Policy. <https://doi.org/10.4271/13-01-01-0001>

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