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Article

Analysis of the National Annual Emission of Pollutants from Road Transport in Poland in the Years 1990–2020

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Abstract: This article presents the official results of the inventory of pollutants in Poland from road transport, developed in the National Centre for Emissions Management of the Institute of Environmental Protection–National Research Institute in Warsaw as part of the reporting of national pollutant emissions. Considerations concern pollutants harmful to the health of living organisms. Source information is available in official reports. The national emission of pollutants covers the years of balancing 1990–2020. The aim of the work is to assess the dynamic properties of the processes of national emission of pollutants. The list of substances whose emission is examined in the article is consistent with the list of substances in accordance with the European Monitoring and Evaluation Programme/European Environment Agency (EMEP/EEA) procedure. These are pollutants harmful to the health of living organisms. The inventory of pollutants contributing to the intensification of the greenhouse effect in the atmosphere is carried out as part of separate reports. The energy consumption of road vehicles was presented. The ratio of national annual emissions of pollutants in 2020 and in 1990 is the lowest for sulfur oxides and lead, followed by non-methane volatile organic compounds and carbon monoxide. The analysis of the national pollutant emission concerned the energy emission factor, which characterizes the ecological level of the accumulated category of road vehicles. This is an original element, not seen before in world literature and official reports of research institutes. An unambiguous trend of decreasing the energy emission factor of pollutants, apart from ammonia, was found, which is related to the use of catalytic flue gas purification systems. This is confirmed by the analysis of the ratio of the energy emission factor in 2020 and 1990, as well as the relative derivative of the energy emission factor in relation to the national annual emission of pollutants.

Keywords: energy emission factor; road transport; inventory of pollutant emission

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1. Introduction

The pollution emission inventories are now a widely used tool in environmental protection [1–4], which has been supported by sanctioned procedures enabling the critical assessment of the impact of civilization-related activities on the environment. Pollutant emissions from road transport play a special role in environmental protection, largely due to the extensive use of road vehicles, especially in the regions with high population densities, i.e., large urban agglomerations.

The inventories of emissions from road transport are now carried out in all developed countries [5–10]. For example, the emissions from road transport in the Greater Dublin area are referred to in [5]. The emissions from road transport in England, Wales, Scotland and

Northern Ireland between 2005 and 2020 are analyzed in [6]. Large importance is given to the emissions from automotive sources in rapidly developing countries. The study in [9] presents the results of the inventory of pollutant emissions from road transport in India in 2020 and the study in [10] in the years 1998–2015 in China. The work in [11] presents the assessment of the daily electricity demand on selected roads in Turkey. It is this case study that shows the development of electromobility on Turkish highways. The study in [12] is another work in which the problems of electromobility are discussed together with the problems of power systems and energy storage systems referring to Turkish traffic. In [13], Valido et al. show vehicle pollution and fuel consumption monitoring on the basis of artificial intelligence-based (AI) system and gas emission estimator model. This work demonstrates a state-of-the-art solution that includes already invented technologies, such as convolutional neural networks and emission models enabling a camera as an emission detector. Another paper that discusses the problems of particulate matter (PM) emission from traffic is the study in [14] by Salva et al. In this paper, the authors state that the PM emission is influenced by such factors as vehicle category, emission standard, vehicle velocity and weather conditions. The work in [15] by Storch et al. discusses the problem of PM emission because of the brake wear. The paper in [16] by Bondorf et al. presents the results of investigation of PM emission resulting from brakes and tires exploitation wear. Another paper in which emission of pollutants in real driving conditions is estimated based on the data from on-board diagnostic system (OBD) and machine learning is the article in [17] by Rivera-Campoverde et al. Such an approach is original as it includes the standard onboard equipment of automobile.

The study in [18] by Baghestani et al. presents the problems of traffic congestion, discussing how to relieve traffic jams and minimize traffic impact on environment, health and economy. Congestion pricing was presented by the authors as one of the most popular concepts.

When discussing the problem of pollution from traffic, a very crucial issue is traffic modeling, which is presented in [19] by Oskarbski and Biszko and in [20] by Zourlidou et al.

Poland is one of the countries that has dynamically developed in the last 30 years.

In Poland, the inventory of pollutant emissions is carried out by the National Centre for Emissions Management (KOBiZE) of the Institute of Environmental Protection–National Research Institute. The work of KOBiZE [7,8] is reported to the Convention on Long-Range Transboundary Air Pollution (CLRTAP) [21] and to the European Union in accordance with Directive (EU) 2016/2284 of the European Parliament and of the Council of 14 December 2016 on the reduction in national emissions of certain types of atmospheric pollutants (the so-called ceilings directive) [1]. KOBiZE reports are also used by the Statistics Poland.

The rapid progress of motorization over the last three decades can be measured by the number of vehicles and the intensity of their use. However, this method is not an objective enough way for related calculations, as many cars are not used on a regular basis by their owners. Instead, a good measure of vehicle use intensity is fuel consumption. Figure 1 shows the national energy consumption-L by road vehicles in Poland between 1990 and 2020.

Figure 1 clearly shows a progressive increase in fuel consumption in Poland, which was over 3.5 times greater in 2020 as compared to that in 1990.

Evidently, such dynamic motorization progress may be accompanied by an increase in pollutant emissions. This adverse effect can be counteracted by improving vehicle technical parameters related to emissions [22–24], as well as by applying appropriate technical and organizational measures to vehicle traffic.

The formal results of the pollutant emissions inventory carried out by the National Centre for Emissions Management (KOBiZE), operating at the Institute of Environmental Protection–National Research Institute (IOŚ-PIB), were included the framework of the present study.

The aim of the work is to assess the dynamic properties of the processes of the national emission of pollutants and the process of the energy emission factor, which characterizes the ecological level of the cumulative category of road vehicles. The original element of the novelty in the publication is the fact that it was not limited to the study of the national annual emission of pollutants from road transport. The energy emission indicator was also analyzed in an objective way characterizing the impact of technical progress on the emission of pollutants. It was also examined how fast the energy emission indicator decreased in the years of balancing.

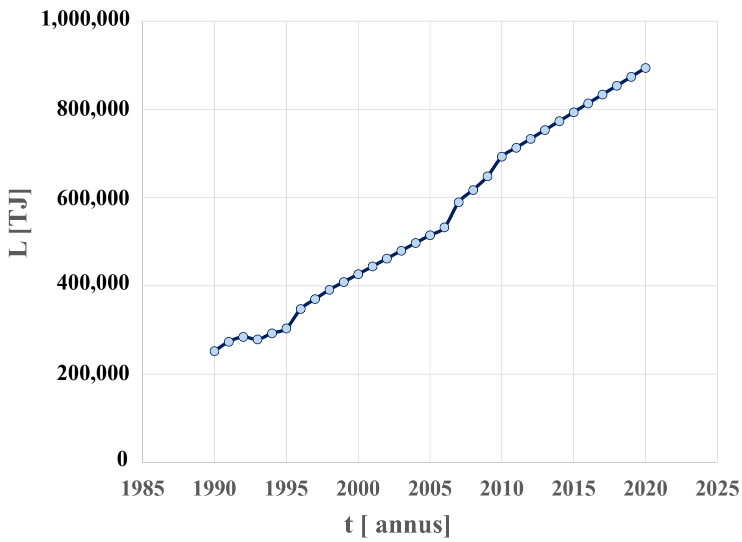

Figure 1. Energy consumption by road vehicles at a national level (L) [7,8].

2. Research Methodology

The COPERT software is used for the inventory of pollutant emissions from road transport. The data for the software concern the number and traffic conditions of road vehicles of all elementary categories, as well as the properties of vehicles [2,25–32].

A category is a class of objects that have specific characteristics and are related to each other. Categories of road vehicles are determined according to the following criteria: application, contractual size, construction properties, fuel and technical level [2,25–32].

The elementary category of road vehicles includes vehicles with all the same criteria. The cumulative category of motor vehicles includes vehicles with not all the same criteria.

Due to the use of the inventory of pollutant emissions from road transport, cumulative categories are distinguished: passenger cars, light trucks, trucks, city buses, long-distance buses and L-category road vehicles (motorcycles, mopeds, quads and microcars).

For light vehicles (passenger cars, light trucks, minibuses and L-category vehicles, i.e., motorcycles and mopeds as well as quads and microcars), the criterion for the contractual size of vehicles is the cubic capacity of the internal combustion engine, and for heavy vehicles (trucks and city and long-distance buses) it is their maximum mass [2,25–32].

In elementary categories, there is a differentiation due to [2,25–32]:

- type of cycle: spark-ignition and compression-ignition engines and two-stroke and four-stroke engines,
- fuel, mainly: gasoline, diesel oil and esters of vegetable oils,
- for light vehicles: engine displacement,
- for heavy vehicles: the maximum weight of the vehicle,
- pollutant emission class for light vehicles: Pre Euro, Euro 1–Euro 6; for heavy vehicles: Pre Euro, Euro I–Euro VI.

The criterion value of pollutant emissions for light vehicles is the specific distance emission determined in the type-approval driving tests. In the European Union, this is the WLTP procedure (Worldwide Harmonized Light Vehicles Test Procedure) [22,23]. In addition, the specific distance emission of nitrogen oxides and the road number of particulate matter in real vehicle traffic conditions are determined in the RDE (Real Driving Emissions) test [22,23]. The specific distance emission of pollutants is a derivative of pollutant emission (mass) in relation to the length of the road covered by the vehicle. The specific distance number of particulate matter is a derivative of the number of particulate matter in relation to the length of the road covered by the vehicle.

The emission criterion for heavy-duty vehicles refers to the engine driving the vehicle. This value is the specific brake emission of pollutants and the specific brake number of particulate matter determined in the European Union in the static WHSC (World Harmonized Steady Cycle) test and in the dynamic WHTC (World Harmonized Transient Cycle) test [22,23]. The specific brake emission of pollutants is the derivative of the pollutant emission in relation to the work performed by the engine, and the specific brake number of particulate matter is the derivative of the number of particulate matter in relation to the work performed by the engine.

For each elementary category of vehicles, two values are adopted, characterizing the intensity of use: the number of vehicles in the category and the annual mileage of vehicles. The product of the number of vehicles and their mileage can be taken as a measure of the intensity of use of vehicles of a given elementary category.

The change in the structure of vehicles in particular years consists primarily in the systematic decrease in the number and annual mileage of vehicles of lower ecological categories and, as a consequence, in the increase in the number and annual mileage of vehicles of higher ecological categories. In addition, cars with two-stroke engines were practically eliminated. There is also a tendency to increase the share of the intensity of use of larger vehicles: passenger cars and light trucks with engines with larger cylinder volumes and heavy vehicles with a higher total weight.

Pollutant emissions are balanced for four model vehicle traffic conditions: in traffic jams, in cities outside of traffic jams, outside cities and on motorways and expressways [2,26]. The national annual pollutant emission is equal to the sum of pollutant emission for each of the model traffic conditions.

For each of these conditions, the average velocity of vehicles and the share of the length of the road covered in the given model traffic conditions in the total length of the road covered in all traffic models are assumed [2,26]. The average velocity of vehicles is significant for the nature of their traffic. It is typical that a low average velocity corresponds to significant traffic impediments, resulting in frequent braking and acceleration and a large proportion of time with zero velocity.

The COPERT software contains the characteristics of pollutant emissions for individual elementary categories of vehicles in the form of dependence of specific distance emissions on average vehicle velocity [2].

The present work examines the national annual emissions-E_a of harmful to the health of living organisms pollutants, such as:

- carbon monoxide—CO,
- non-methane volatile organic compounds—NM VOCs,
- nitrogen oxides—NO_x, represented by nitrogen dioxide—NO_2,
- total suspended particulates—TSP,
- particulate matter—PM2.5,
- particulate matter—PM10,
- black carbon—BC,
- sulphur oxides—SO_x, represented by sulphur dioxide—SO_2,
- lead—Pb,
- ammonia—NH_3.

Formally, the national annual pollutant emission is the intensity of pollutant emission averaged over the period of a calendar year. The national annual emission of pollutants is extensive. Comparing the value of the national annual emission of pollutants makes it possible to assess the dynamics of the total emission of pollutants.

The energy emission factor was examined:

$$EN = \frac{E_a}{L} \quad (1)$$

The energy emission factor is an intensive value. The energy index of pollutants enables an objective assessment of the quality of the research subject—the focus of this work—a road vehicle due to the emission of pollutants. For most pollutants, the energy emission factor characterizes the quality of the internal combustion engine in terms of pollutant emissions. Only in the case of dust emissions, tribological pairs occurring in the vehicle and in the conditions of its cooperation with the environment are their additional source. These are primarily tribological pairs in the braking system, in the clutch, as well as the tires of the wheels cooperating with the road surface. The reduction in the energy pollutant emission indicator results primarily from the technical improvement of internal combustion engines, mainly due to the use of equipment systems for reducing pollutant emissions.

The subject of subsequent analyses was the relative derivative of the energy emission factor in relation to the national annual pollutant emissions:

$$\delta[EN] = \frac{\frac{dE_a}{dt}}{E_a} \quad (2)$$

The relative derivative of the energy emission factor in relation to time in relation to the national annual emission of pollutants from road transport in Poland in the years 1990–2020 characterizes the dynamic properties of the energy emission factor.

The analysis of the relative derivative of the energy emission factor was conducted in consideration of the possibility of assessing the sensitivity of the energy emission factor to pollutants in relation to the years of balancing pollutant emissions.

The ratio of national annual emissions of pollutants in 2020 and in 1990 is

$$\delta(E_a(2020), E_a(1990)) = \frac{E_a(2020)}{E_a(1990)} \quad (3)$$

The ratio of the national annual emission of pollutants in 2020 and in 1990 characterizes the changes in the national annual emission of pollutants in the years of pollutant emission balancing.

The ratio of the energy pollutant emission factor in 2020 and in 1990 is determined in the form of

$$\delta(EN(2020), EN(1990)) = \frac{EN(2020)}{EN(1990)} \quad (4)$$

The ratio of the energy emission factor in 2020 and in 1990 is used to assess changes in the ratio of the energy emission factor in 2020 and in 1990 in the years of pollutant emission balancing.

3. National Annual Emissions from Road Transport

Figures 2–11 show the national annual emissions of pollutants from road transport in Poland in the years 1990–2020.

Despite the dynamic increase in the intensity of the use of road vehicles in subsequent years, in particular their number, the national annual emission of carbon monoxide is systematically decreasing. This is primarily the result of improving the technical quality of internal combustion engines, mainly spark-ignition engines. This applies primarily to light vehicles because spark-ignition engines are predominant in them. The factors affecting the reduction in the national annual carbon monoxide emissions are significantly affected by the decreasing share of the use of two-stroke engines for driving vehicles.

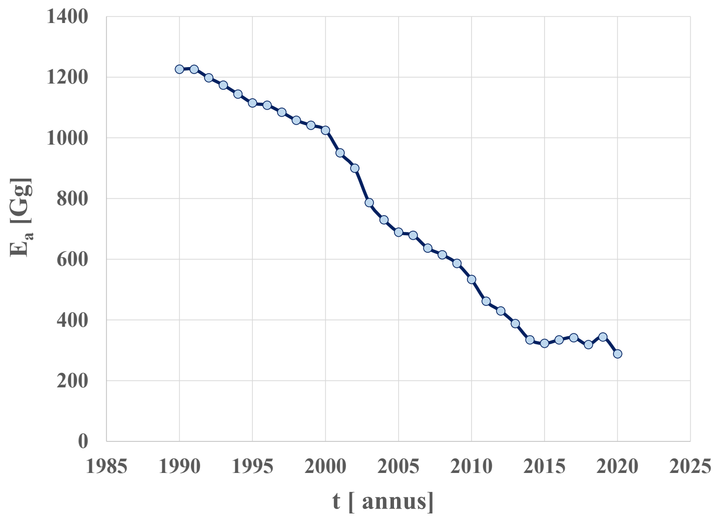

Figure 2. National annual emissions (E_a) of carbon monoxide (CO) [7,8].

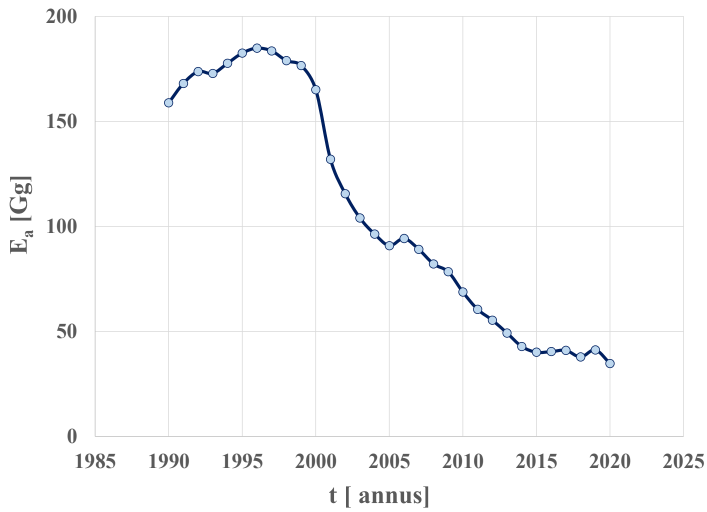

Figure 3. National annual emissions (E_a) of non-methane volatile organic compounds (NM VOCs) [7,8].

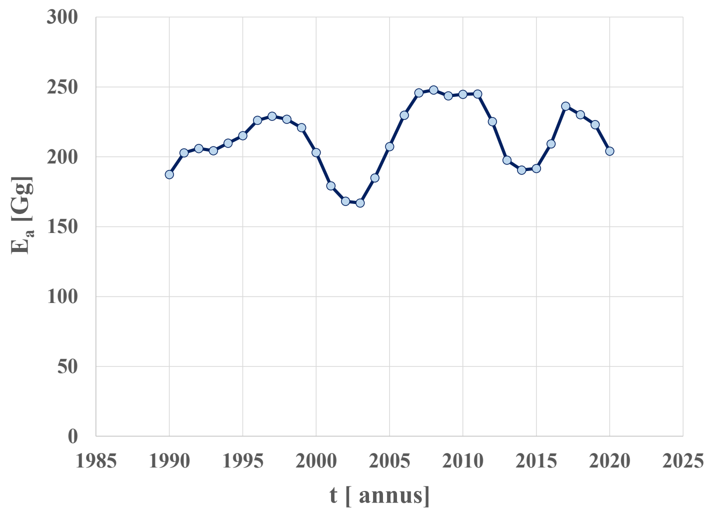

Figure 4. National annual emissions (E_a) of nitrogen oxides (NO_x) [7,8].

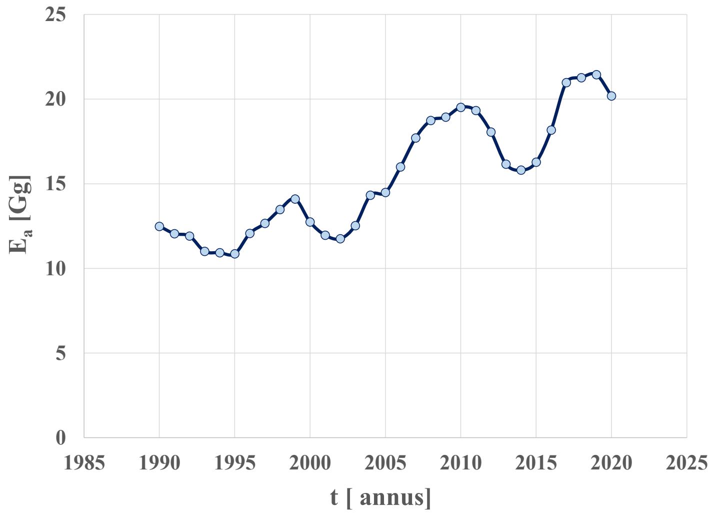

Figure 5. National annual emissions (E_a) of total suspended particulates (TSP) [7,8].

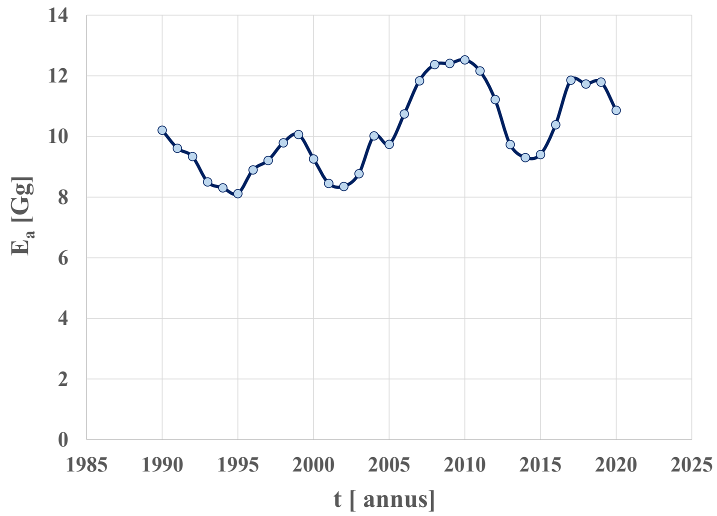

Figure 6. National annual emissions (E_a) of particulate matter (PM2.5) [7,8].

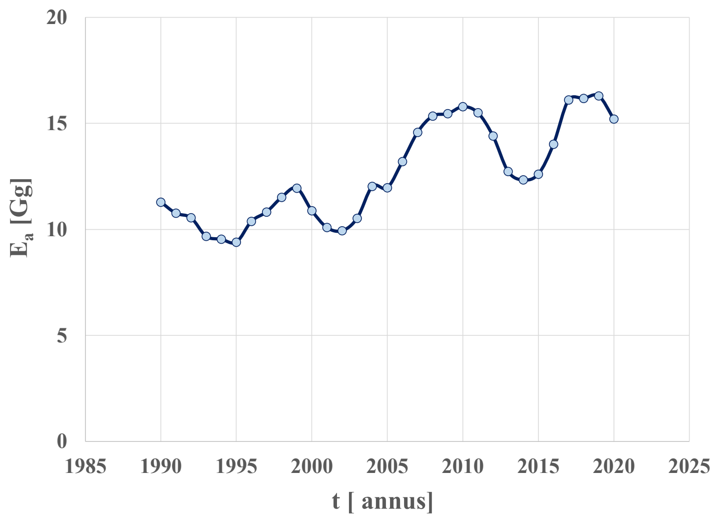

Figure 7. National annual emissions (E_a) of particulate matter (PM10) [7,8].

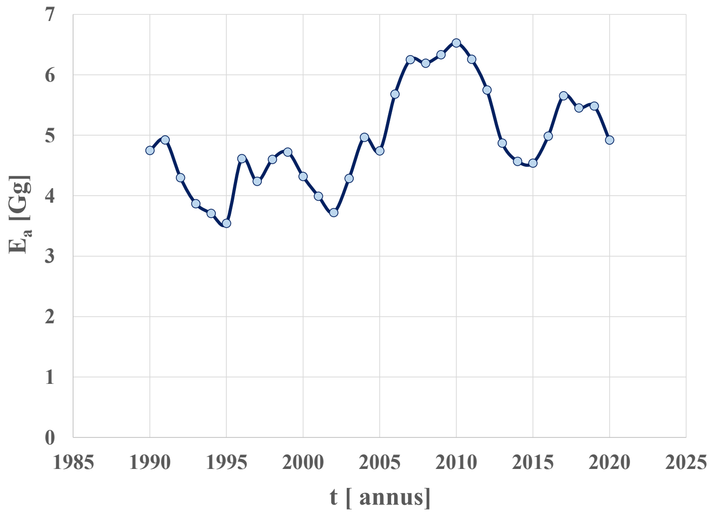

Figure 8. National annual emissions (E_a) of black carbon (BC) [7,8].

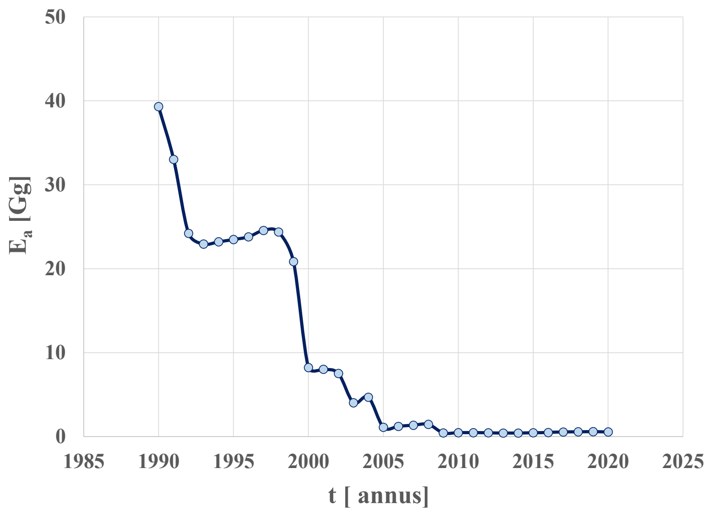

Figure 9. National annual emissions (E_a) of sulphur oxides (SO_x) [7,8].

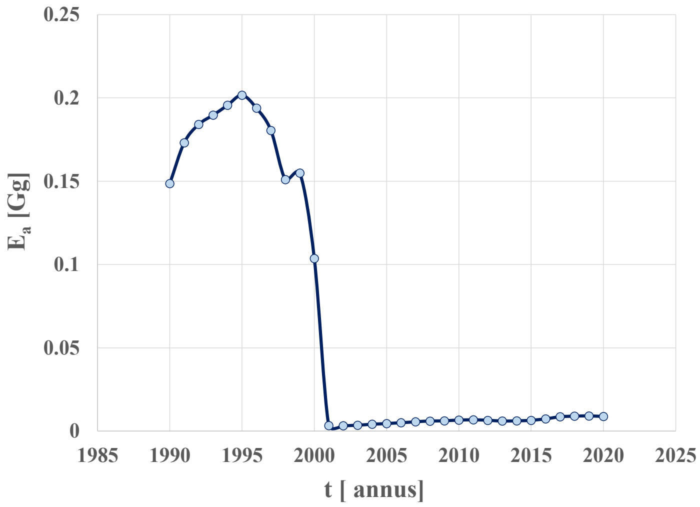

Figure 10. National annual emissions (E_a) of lead (Pb) [7,8].

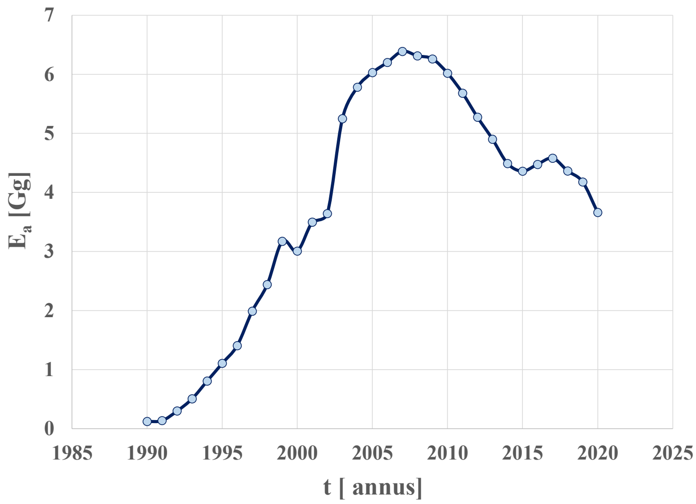

Figure 11. National annual emissions (E_a) of ammonia (NH_3) [7,8].

The national annual emissions of non-methane volatile organic compounds have been systematically decreasing since 1996. The reasons for the decrease in national annual emissions of non-methane volatile organic compounds are similar to the national annual emissions of carbon monoxide. In the case of non-methane volatile organic compounds, the declining share of the use of two-stroke engines for driving cars plays a particularly important role.

In the case of the national annual emission of nitrogen oxides, there is no such significant trend as in the case of carbon monoxide and non-methane volatile organic compounds. This is mainly due to the greater technical difficulty of the methods of reducing nitrogen oxide emissions from internal combustion engines. It should be noted that the dominant factor contributing to the emission of nitrogen oxides is the maximum temperature of the thermodynamic medium during combustion. Therefore, the postulates of reducing nitrogen oxide emissions and reducing fuel consumption are in a sense contradictory.

The similarity of the national annual emission of particulate matter—total-TSP and their components PM10, PM2.5 and soot—is significant, as these values are approximately linearly dependent on each other.

The tendency of increasing the national annual emission of particulate matter is influenced by the increasing intensity of road vehicle use. Technical methods of reducing particulate emissions, primarily from diesel engines, are among the most difficult to implement. A breakthrough event was the development of diesel particulate filters and methods of their regeneration. Nevertheless, a significant increase in the number of heavy vehicles and the intensity of their use, which is related to the development of the economy (transport of goods by trucks), has a decisive influence on the tendency to increase the national annual emission of dust.

Another important factor affecting the emission of particulates is their origin from tribological pairs. First of all, the cooperation between the tires of the road wheels and the road surface plays a dominant role in this case. For safety reasons, high adhesion between the road wheels and the road surface is more important than dust emissions.

In the case of sulfur oxides, there is a significant decrease in the national annual emission until 2005. Since 2005, the emission of sulfur oxides has been practically insignificant. This is related to a radical reduction in the share of sulfur in fuels, especially in diesel oil. In the 20th century, the mass fraction of sulfur in diesel fuel for automotive applications was 1.5%. The share of sulfur in fuels for large internal combustion engines, e.g., marine ones, was even more than 5%. A large share of sulfur in fuels had a beneficial effect on reducing the wear of the surfaces of the pairs cooperating with each other, especially the cylinder surface and the side surface of the piston. A particularly big problem in this case occurs in large engines with a large displacement, for which high friction work occurs at a large piston stroke. Therefore, a large share of sulfur in fuels was justified, but at the same time it was conducive to the emission of nitrogen oxides that are very harmful to the environment.

The decisive factor contributing to the radical reduction in the sulfur fraction in fuels was the widespread use of catalytic reactors in engines to reduce the emission of pollutants, mainly carbon monoxide, organic compounds and nitrogen oxides, as well as catalytic particulate filters. Sulfur contained in fuels affected the destruction of catalytic reactors and catalytic particulate filters due to the coating of active surfaces of catalytic reactors and catalytic particulate filters with sulphates. Currently, there is a sulfur limit of 50 ppm (parts per million) in automotive diesel fuel, and very often it is even less than 10 ppm.

Since 2001, there has been a very large decrease in the national annual lead emission. This is due to the radical reduction in the proportion of lead compounds (primarily tetraethyl lead) added to motor gasolines, used to increase the resistance to knocking combustion, which is particularly dangerous for internal combustion engines. The threat of knocking combustion is particularly dangerous for spark-ignition engines with a high compression ratio in order to increase effective efficiency. The use of the addition of lead compounds to fuels resulted in the emission of lead compounds that are very harmful to the health of living organisms. The reasons for the practical elimination of lead compounds from gasoline are similar to those in the case of the sulfur-durability of catalytic reactors. Lead coated the active surface of catalytic reactors, damaging the reactors. Eliminating the addition of lead compounds to gasolines required a different composition of gasolines in order to ensure sufficient resistance to knocking combustion. Straight-chain aliphatic hydrocarbons have a particularly low resistance to knock combustion; aromatic hydrocarbons have a high resistance, but they are much more harmful to the health of living organisms than aliphatic ones. The solution is the use of branched aliphatic hydrocarbons (mainly isooctane-2,2,4-trimethylpentane). Such gasolines are produced to a large extent by the use of reformulation techniques and for this reason they are called reformulated gasolines.

The dominant factor in the process of increasing the national annual ammonia emission is the increasing intensity of the use of motor vehicles. Ammonia emission is related to catalytic methods of exhaust gas purification of impurities. The improvement of these methods means that since 2007, despite a significant increase in the intensity of motorization, there has been a decrease in national annual motorization emissions.

In most cases, irrespective of the dynamic development of motorization in Poland (dynamic increase in energy consumption at a country level—Figure 1), there was a clear trend toward a decrease in the national annual emissions of carbon monoxide starting from the beginning (1996) of balancing pollutant emissions and non-methane volatile organic compounds. These results are associated with the major progress in design solutions for internal combustion engines with the aim of reducing pollutant emissions [22–24]. There was an evident reduction in the national emission of sulphur oxides, starting from 1998, and of lead, starting from 1999. This marked decrease in the national emissions of sulphur and lead oxides is associated with a significant reduction in sulphur and lead in fuels. In the case of nitrogen oxides, there was no explicit trend in the national annual emission patterns, and as for the national annual emissions of particulate matter, a slightly increasing trend was observed. The emission of nitrogen oxides is strongly dependent on many factors related to both the properties of vehicle combustion engines [22–24] and the way they are used, i.e., the models of their motion [26]. If we consider the properties of engines, it is important what category of vehicles they are in terms of pollutant emissions and whether they are spark-ignition or compression-ignition engines [29,31,33]. Therefore, it is difficult to unambiguously interpret the process of national annual emission of nitrogen oxides from road vehicle engines. In the case of particulate emissions, the share of particulate emissions from internal combustion engines is significantly reduced, while the emission of particulate matter from tribological pairs is maintained [27]. In particular, ensuring high adhesion between tyred wheels and road surface is a priority for safety reasons, and this is conducive to increased dust emissions.

Table 1 shows the ratio of national annual emissions of pollutants in 2020 to 1990.

Table 1. The ratio of pollutant national annual emissions in 2020 to 1990.

L	CO	NM VOC	NO_x	TSP	PM2.5
3.542	0.235	0.219	1.089	1.617	1.064
PM10	**BC**	**SO_x**	**Pb**	**NH_3**	
1.347	1.036	0.0140	0.0593	29.9	

The largest reductions are, evidently, in the national emissions of sulphur oxides and lead, followed by non-methane volatile organic compounds. National annual emissions of ammonia increased the most, but overall, these were not large when compared to other civilization-related activities.

4. Analysis of the Energy Factor of Pollutant Emissions from Road Transport

Figures 12–21 show the energy emission factor for road transport in Poland in the years 1990–2020.

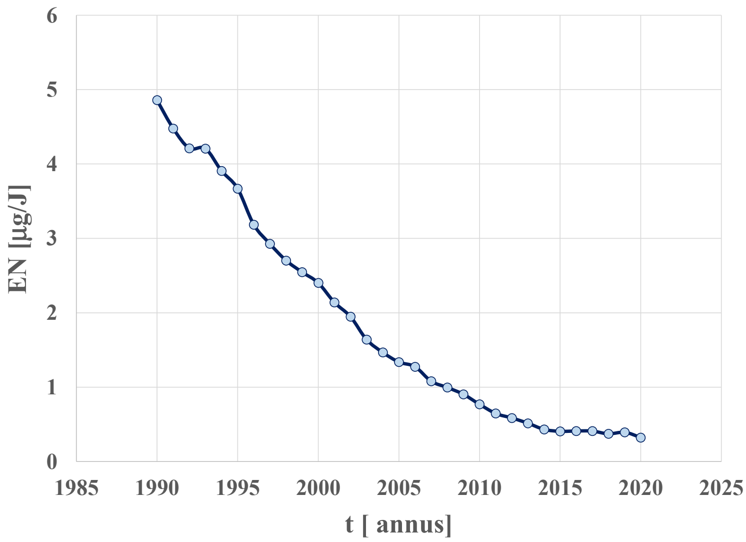

Figure 12. The energy emission factor (EN) of carbon monoxide (CO).

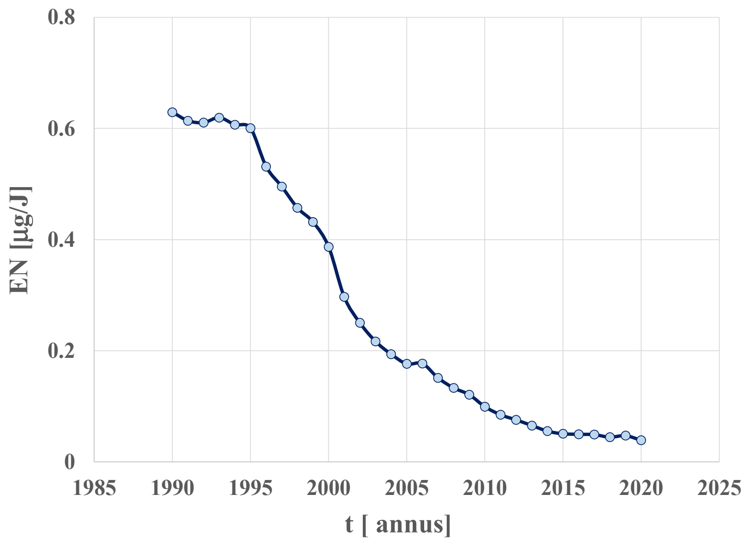

Figure 13. The energy emission factor (EN) of non-methane volatile organic compounds (NM VOCs).

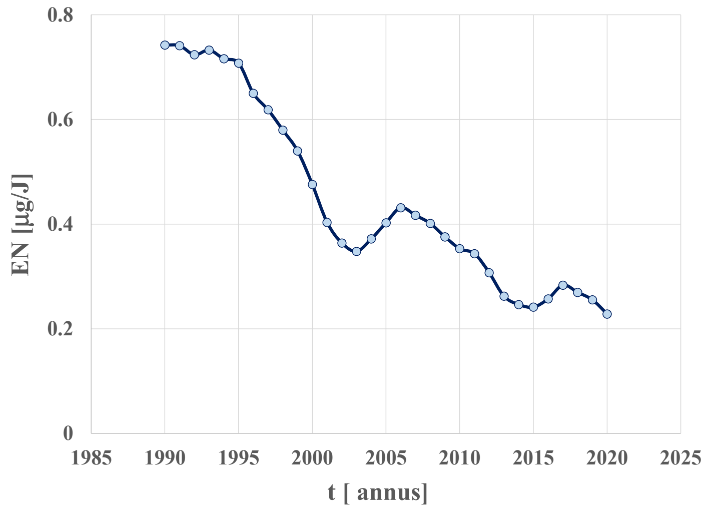

Figure 14. The energy emission factor (EN) of nitrogen oxides (NO_x).

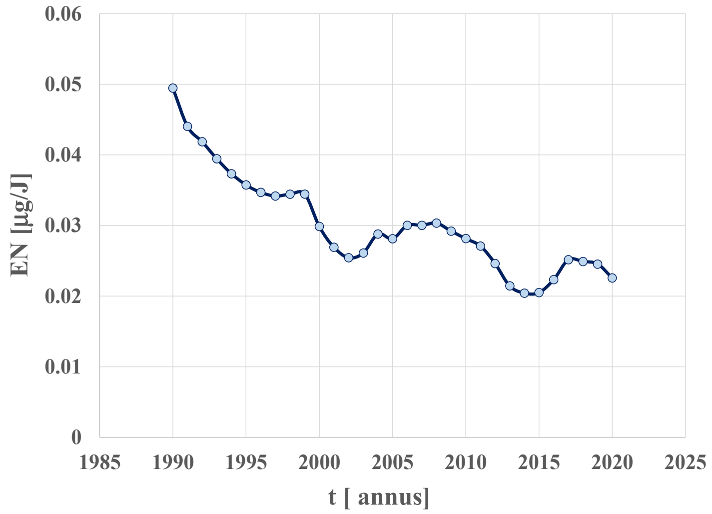

Figure 15. The energy emission factor (EN) of total suspended particulates (TSP).

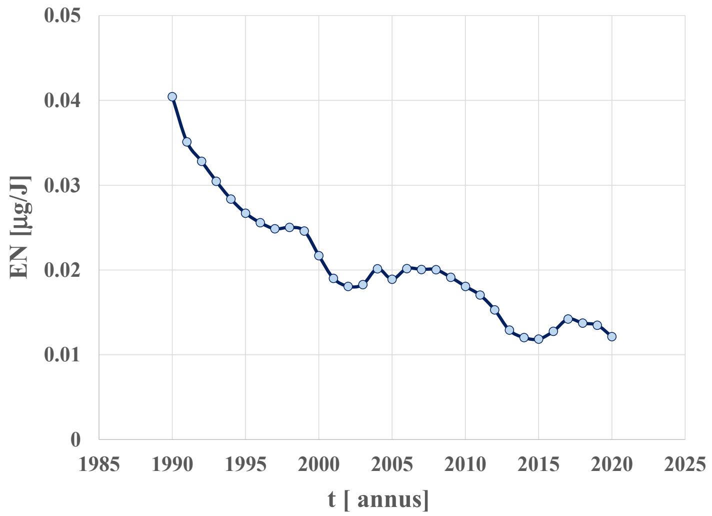

Figure 16. The energy emission factor (EN) of particulate matter (PM2.5).

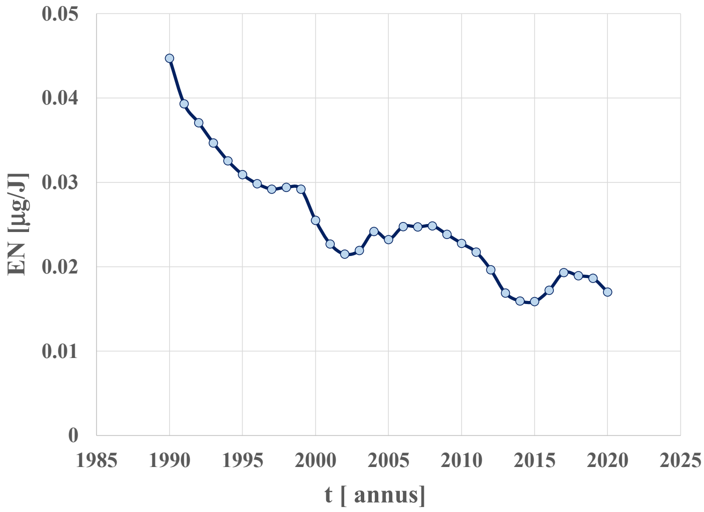

Figure 17. The energy emission factor (EN) of particulate matter (PM10).

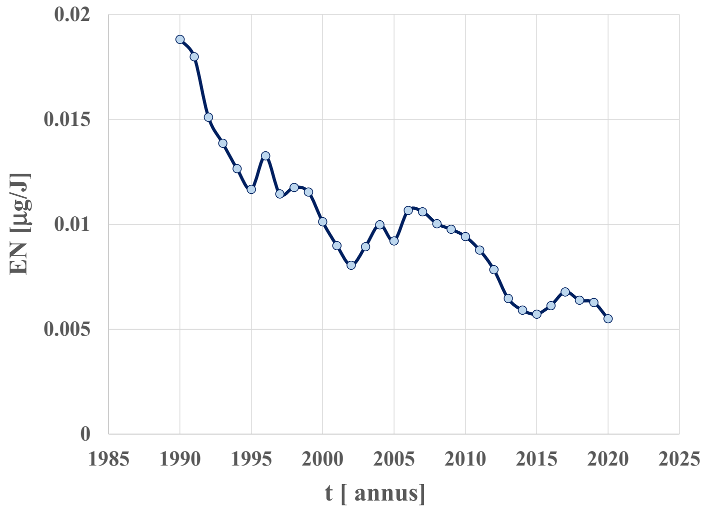

Figure 18. The energy emission factor (EN) of black carbon (BC).

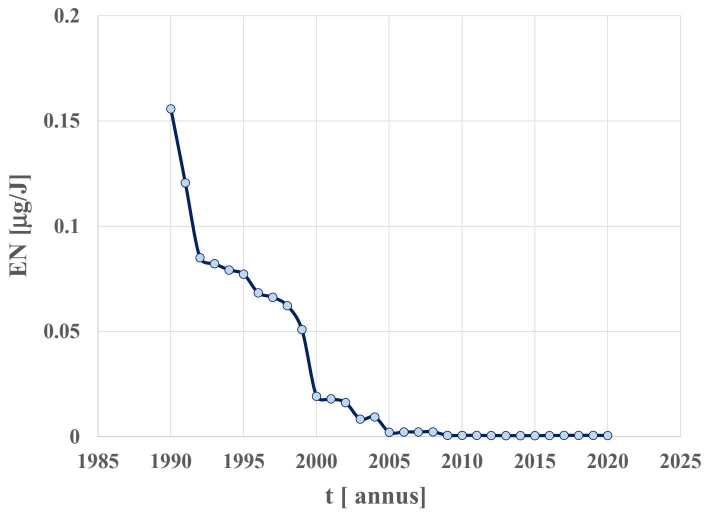

Figure 19. The energy emission factor (EN) of sulphur oxides (SO_x).

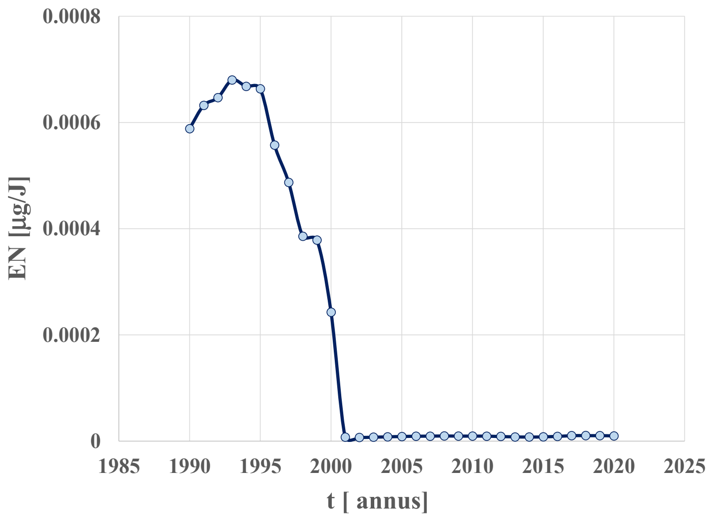

Figure 20. The energy emission factor (EN) of lead (Pb).

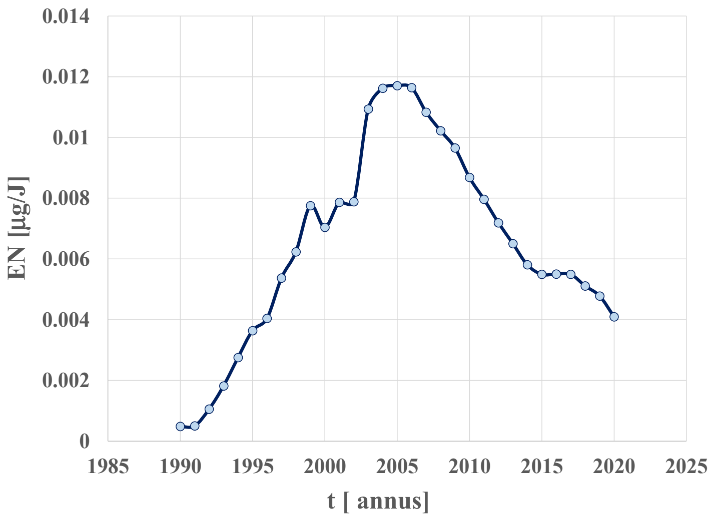

Figure 21. The energy emission factor (EN) of ammonia (NH_3).

There is an unambiguous tendency to a clear decrease in the energy indicator of carbon monoxide emission, which results from technical progress in internal combustion engines and a decrease in the share of two-stroke engines in the number of cars.

As with the carbon monoxide energy factor, there is a clear decrease in the energy factor of non-methane volatile organic compounds. The causes of this condition are similar to those in the case of carbon monoxide.

In the case of the national annual emission of nitrogen oxides, there is no definite downward trend, while in the case of the energy emission factor, there is a clear downward trend. This is the result of technical progress, primarily in the field of exhaust fumes cleaning methods.

As in the case of nitrogen oxides, the increasing intensity of motor vehicle use is of dominant importance for the national annual emission of particulate matter. On the other hand, the energy indicator of dust emission is clearly decreasing due to technical progress. In the case of diesel engines, a significant reduction in particulate emissions has been achieved thanks to a significant increase in pressure and rational control of fuel injection rate, controlled air turbulence, and above all the use of catalytic particulate filters and regeneration systems.

The decrease in the sulfur oxides emission factor is related to the change in the composition of fuels, especially diesel oil.

As in the case of sulfur oxides, the energy factor of lead emission is mainly influenced by the practical elimination of lead compounds from motor gasolines.

The regular reduction in the energy factor of ammonia emission since 2005 is the result of technical progress in the field of catalytic flue gas cleaning methods.

The decreasing trends of national annual emissions of all the pollutants examined excluding ammonia were evident.

Table 2 shows the ratio of the energy emission factor of pollutants in 2020 to 1990.

Table 2. The ratio of energy emission factor in 2020 to 1990.

CO	NM VOC	NO_x	TSP	PM2.5
0.066	0.062	0.308	0.457	0.300
PM10	**BC**	**SO_x**	**Pb**	**NH_3**
0.380	0.293	0.004	0.017	8.454

The lowest ratios were observed for sulphur oxides, lead, followed by carbon monoxide and non-methane volatile organic compounds.

Figures 22–31 show the relative derivative of the energy emission factor versus time in relation to the national annual emission of pollutants from road transport in Poland in the years 1990–2020. The graphs presented below also show the linear trends of the derivative of the energy emission factor of the pollutants studied.

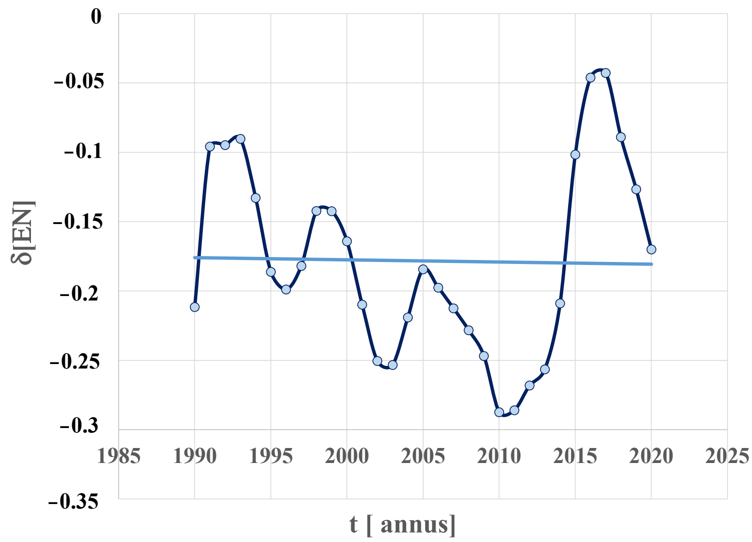

Figure 22. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of carbon monoxide (CO) (blue line—linear approximation).

The linear trend of the relative derivative of the energy carbon monoxide emission factor over time in relation to the national annual carbon monoxide emission is practically constant and negative, which means that the relative derivative of the energy carbon monoxide emission factor over time decreases in a similar way in the years 1990–2020.

The linear trend of the derivative of the energy indicator of non-methane volatile organic compounds emissions versus time in relation to the national annual emission of non-methane volatile organic compounds is negative and tends to decrease, i.e., the energy indicator of emissions of non-methane volatile organic compounds decreases faster and faster in the years 1990–2020.

The linear trend of the derivative of the energy indicator of nitrogen oxides emission against time in relation to the national annual emission of nitrogen oxides is practically constant and negative. This means that, despite the great technical difficulties in reducing nitrogen oxide emissions, the developed methods are effective.

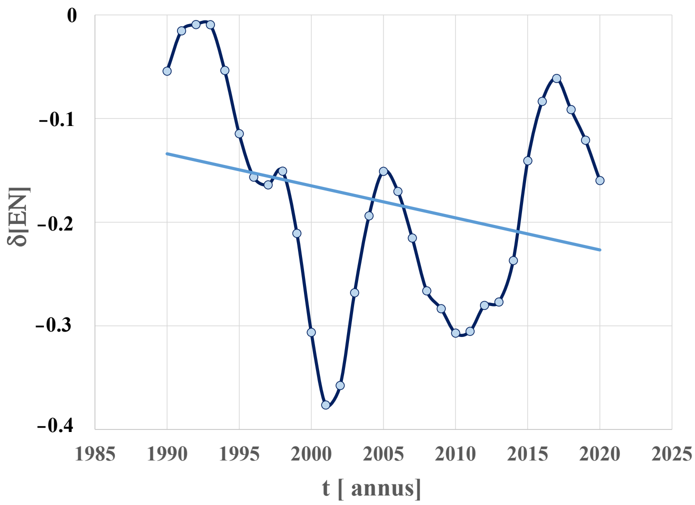

Figure 23. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of non-methane volatile organic compounds (NM VOCs) (blue line—linear approximation).

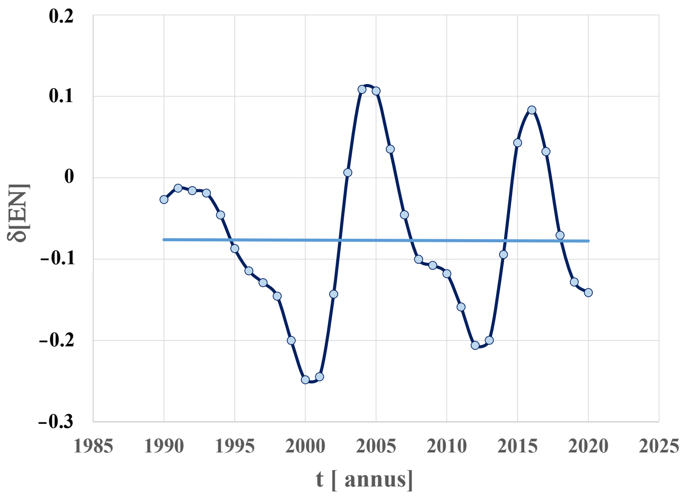

Figure 24. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of nitrogen oxides (NO_x) (blue line—linear approximation).

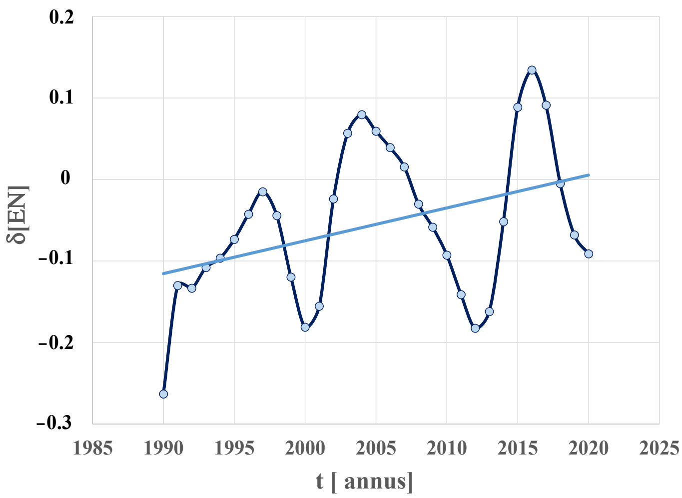

Figure 25. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of total suspended particulates (TSP) (blue line—linear approximation).

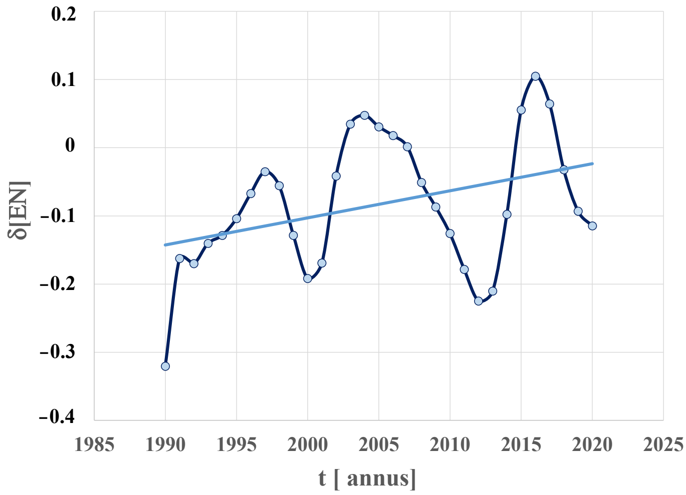

Figure 26. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of particulate matter (PM2.5) (blue line—linear approximation).

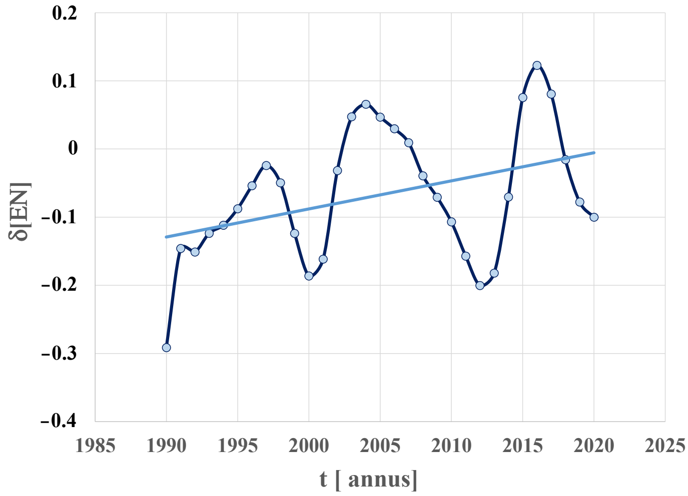

Figure 27. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of particulate matter (PM10) (blue line—linear approximation).

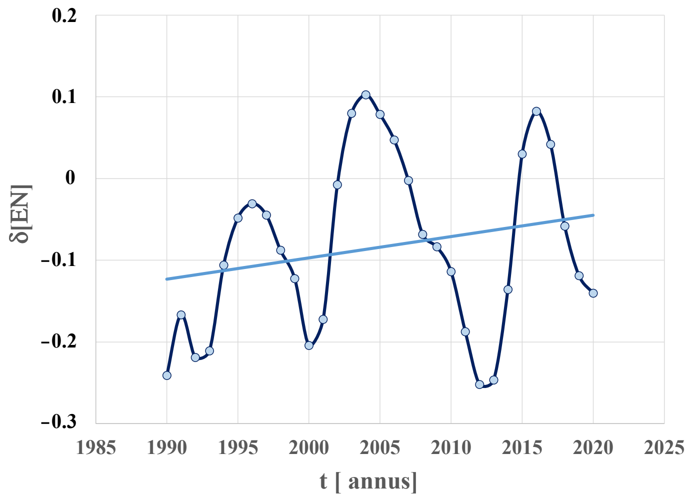

Figure 28. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of black carbon (BC) (blue line—linear approximation).

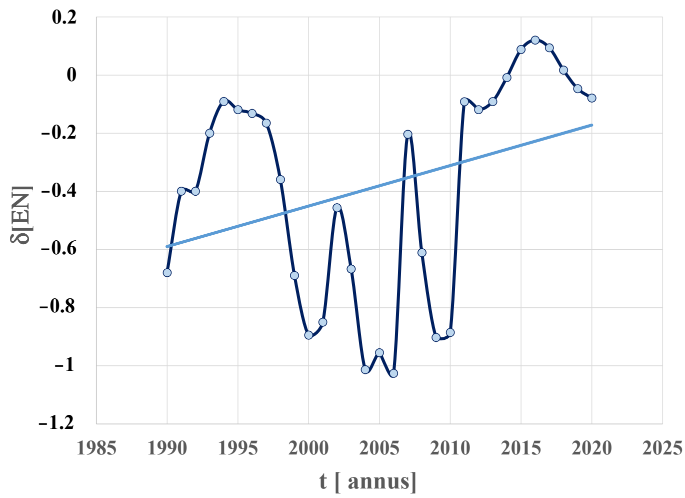

Figure 29. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of sulphur oxides (SO_x) (blue line—linear approximation).

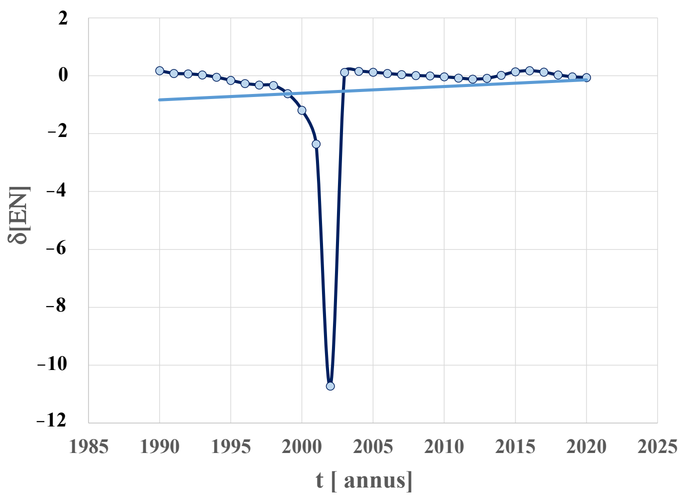

Figure 30. The relative derivative (δ [EN]) (black line) of the energy emission factor relative to the national annual emissions of lead (Pb) (blue line—linear approximation).

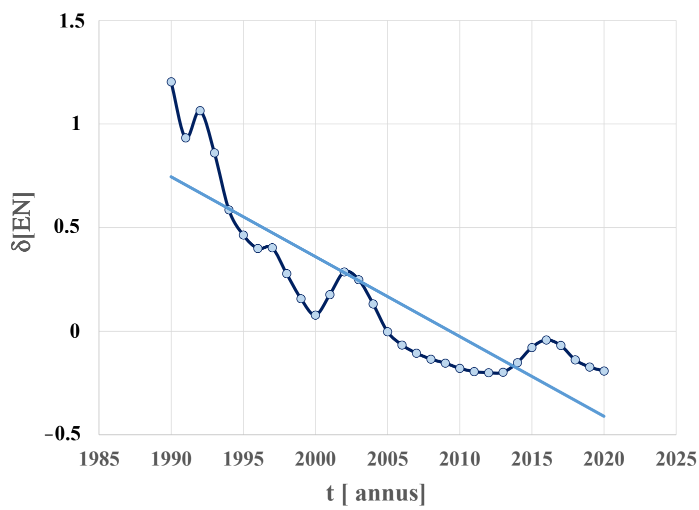

Figure 31. Relative derivative (δ [EN]) (black line) of the energy emission factor relative to national annual emissions of ammonia (NH_3) (blue line—linear approximation).

The linear trend of the derivative of the energy dust emission index versus time in relation to the national annual dust emission is negative and tends to increase. This means that there is a tendency for a slower and slower decrease in the energy indicator of dust emission. This is significantly influenced by the share of particulate matter components from tribological pairs in road transport.

The linear trend of the derivative of the sulfur oxides emission index over time in relation to the national annual emission of sulfur oxides is negative and tends to increase. In the years 2004–2006, there was a significant decrease in the energy indicator of sulfur oxides emission due to a radical decrease in the share of sulfur content in fuels.

The linear trend of the derivative of the energy lead emission factor over time in relation to the national annual lead emission is practically constant. Significant is the abrupt change in the value around 2003.

The linear trend of the derivative of the energy ammonia emission factor versus time in relation to the national annual ammonia emission tends to decrease regularly.

The energy emission factors for carbon monoxide and nitrogen oxides remained almost unchanged over the period under the study. For particulate matter, the energy emission factor decreased more and more rapidly. This is evident in Table 3 as well.

Table 3. Difference between the relative derivative of the energy emission factor of pollutants relative to the national annual emission of pollutants in 2020 and in 1990.

CO	**NM VOC**	**NO_x**	**TSP**	**PM2.5**
−0.00600	−0.09300	−0.00150	0.12000	0.12000
PM10	**BC**	**SO_x**	**Pb**	**NH_3**
0.12300	0.07800	0.41700	0.69300	−1.15500

5. Conclusions

The paper presents the processes of national annual emission of the considered substances in the years of pollutant emission balancing. Despite the significant development of the automotive industry in Poland, which is accompanied by a significant increase in energy consumption, the national annual emission of pollutants either decreases or increases only slightly. For carbon monoxide, the emission of pollutants has been constantly decreasing, and for non-methane volatile organic compounds since 1996. The most significant is the decrease in the national annual emission of sulfur and lead oxides since the early 2000s and tetraethyl lead in gasoline due to the durability of catalytic exhaust aftertreatment systems. The substances whose test results are presented in the article are carbon monoxide, non-methane volatile organic compounds, total nitrogen oxides, along with their fractions: particulate matter PM2.5 and PM10, soot and sulfur oxides, lead and ammonia.

Furthermore, on the basis of the results of the present study, the following conclusions can be drawn:

1. The national annual emissions of pollutants from road transport are not increasing to a degree corresponding with the dynamic pattern of motorization development. These results are related to the substantial progress in the design solutions of internal combustion engines toward the reduction in pollutant emissions.
2. The value characterizing the dynamic increase in the intensity of the development of the automotive industry in Poland is the annual energy consumption by means of transport. The ratio of energy consumption by means of transport in 2020 and 1990 is over 4. This means a very dynamic development of road transport in Poland.
3. Despite the dynamic increase in the intensity of road vehicle use in subsequent years, in particular their number, the national annual emission of carbon monoxide and non-methane volatile organic compounds is systematically decreasing.
4. In the case of nitrogen oxides, there is no longer such a significant tendency of decreasing the national annual emission. This is mainly due to the greater technical difficulty of the methods of reducing nitrogen oxide emissions from internal combustion engines.

5. The tendency of increasing the national annual emission of particulate matter is influenced by the increasing intensity of road vehicle use. Another important factor affecting the emission of particulates is their origin from tribological pairs. First of all, the cooperation between the tires of the road wheels and the road surface plays a dominant role in this case.
6. Since 2005, there has been a significant decrease in the national annual emission of sulfur oxides. This is related to a radical reduction in the share of sulfur content in fuels, especially in diesel oil. The decisive factor contributing to the radical reduction in the sulfur content in fuels was the widespread use of catalytic reactors.
7. Since 2001, there has been a radical decrease in the national annual lead emission. This is related to the practical elimination of the addition of lead compounds (mainly tetraethyl lead) in motor gasolines, used to increase the resistance to knocking combustion.
8. The energy factors of carbon monoxide and nitrogen oxides emissions remain almost unchanged in the period of balancing emissions. For particulate matter, the energetic emission factor is decreasing more and more rapidly.
9. The lowest is the ratio of the energy pollutant emission factor in 2020 and 1990 for sulfur oxides and lead, followed by non-methane volatile organic compounds and carbon monoxide.

The most significant original achievements of the work are the following:

1. The analysis of the dynamic properties of the processes of national annual emission of pollutants from road transport in Poland was accomplished. The study of the national annual emission of pollutants enables an objective assessment of the harmfulness of transport to the environment, in particular to the health of living organisms.
2. The analysis of the energy pollutant emission factor in the balancing period is completely original and not present in the scientific literature and reports so far. The analysis of the energy process of the pollutant emission factor enables an objective assessment of the improvement in the impact of the automotive industry on the environment thanks to the improvement of the quality of road vehicles due to the emission of pollutants and thanks to the ways of using vehicles, e.g., thanks to rationalization in the field of traffic engineering.

Balancing the emissions from road transport constitutes a tool for rationalization with the aim of protecting the environment from motorization impacts [3]. Detailed studies make it possible to assess the sensitivity of pollutant emissions to the traffic patterns of road vehicles and their structure related to technical characteristics that determine pollutant emissions [25–31,33]. These studies may concern, among others: average velocity of vehicles of general categories in traffic conditions: in traffic jams in cities outside traffic jams, outside cities and on motorways and expressways.

The most important method of research concerns the structure of vehicles in generalized categories due to the use of vehicles, primarily due to the level of pollutant emissions. This is crucial because it is difficult to come to terms with the institutional restriction of the use of road vehicles. In fact, the way to improve the situation in terms of pollutant emissions from road transport consists mainly in improving the quality of vehicles and the ways they are used, and thus in the rational and not arbitrary treatment of the problem of emissions of pollutants harmful to the health of living organisms from road transport, the intensity of which cannot be arbitrarily minimized due to the requirements of modern civilization, in particular the economy.

It is advisable to continue research on the emission of pollutants harmful to health of living organisms from road transport as simulation research in two directions. The first direction is the study of the impact of the structure of road vehicles on pollutant emissions. The second direction is the study of the impact of vehicle traffic models on pollutant emissions.

It is also advisable to continue work in the field of greenhouse gas emissions, fuel consumption and energy consumption.

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Nomenclature

BC	black carbon
E_a	national annual pollutant emissions
EMEP/EEA	European Monitoring and Evaluation Programme/European Environment Agency
TSP	total suspended particulates
EN	energy emission factor of pollutants
L	national annual energy consumption
NM VOCs	non-methane volatile organic compounds
t	time
$\delta(E_a(2020), E_a(1990))$	ratio of the pollutant national annual emissions in 2020 to 1990
$\delta(EN(2020), EN(1990))$	ratio of the energy emission factor of pollutants in 2020 to 1990
$\delta[EN]$	the relative derivative of the energy emission factor

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