referent) concentrations. The Kyiv streets were ranged by the value of danger indices: the central city streets were at the first place. It was determined, that mortal effects from the exposure of exhaust gases were possible only in the incidents. In conditions of intensive motor traffic (over 7000-1000 automobile/year) a risk of the increase of total morbidity of population (especially of the system of blood circulation and nervous system, in occupational conditions – the respiratory system) is possible. Limiting allowable concentrations of priority components are advisably used as a health risk assessment criterion from the exhaust gases exposure. Exhaut gases concentrations that picked up the smell and caused the development of discomfort (for petrol engine at the level of 6 mg/m^3, for diesel engine 4,4 mg/m^3) were offered as a criterion of admissible risk.

POLLUTANTS EMISSION FROM THE AUTOMOTIVE TRANSPORT EXEMPLIFIED IN POLAND SINCE 1990

Irena Twardowska and Joanna Kyziol

Polish Academy of Sciences, Institute of Environmental Engineering,
34 M. Sklodowska-Curie St., 41-819 Zabrze, Poland

Introduction

In the last decade of XX century, significant changes of vehicle number and structure in countries undergoing transition from planned to market economy, among them in Poland, have been observed along with a parallel development of the environmental legislation and rendering emission standards more stringent. Here, the share of the automotive transport in major air pollutant emission in Poland has been presented and analyzed on the background of its quantitative and qualitative development. The dynamics of car fleet displayed fast growth in 1990-2002 (for 76%), but in the next years distinctly slowed down due to the market saturation. The vehicle number growth in this period was not even, and was particularly high for cars (for 114%, i.e. more than twice) and trucks (for 121%), while the number of tractors showed weak increase, and the number of buses and motors decreased (Table 1). At present, car share in total vehicle number comprises 70%.

Transport is a well known mobile source of air, and also soil contamination along the roads. Change in the number and structure of automotive transport means along with the engine and fuel improvement should have resulted in changes of emission loads.

Table 1.

Changes of vehicle and tractor number in Poland since 1990 (GUS, 2004)

Vehicles	1990	1995	2000	2001	2002	2003	
	In thousands						2000=100%
Cars	5261	7517	9991	10503	11029	11244	112.5
Buses	92	85	82	82	83	83	101.2
Trucks	1045	1354	1879	1979	2163	2313	123.1
Motors	1357	929	803	803	869	845	105.2
Tractors	1192	1212	1253	1257	1294	1322	105.5
TOTAL	**9041**	**11186**	**14106**	**14724**	**15525**	**15899**	**112.7**

Methods

Analysis of statistical data on air pollution

The analyzed statistical data concerning emission of air pollutants from vehicles with fuel engines has been estimated by the Institute of Automotive Transport either as a product of a fuel consumption and a specific emission factor that defines the mean load of a specific pollutant emitted from the incineration of the fuel mass unit, or as a product of total annual mileage for a given kind of a vehicle and a road emission factor that defines mean mass of a specific pollutant emitted per one-kilometer mileage. Emission factors for the road transport have been evaluated on the basis of extensive studies of vehicles representative for the Polish vehicle fleet. These emission data comprise greenhouse gases (GHG) and other organic and inorganic pollutants: CO_2, NH_4, N_2O, CO, volatile hydrocarbons, NO_x, solid particulates, SO_2 and Pb. Data on emission of persistent organic pollutants (POPs) comprise emission of dioxins and furans defined by TEQ (toxic equivalent factor with respect to the most toxic dioxin 2,3,7,8-TCDD), as well as a sum of 4PAHs (including BaP – benzo[a]pyrene).

Contamination of soils with PAH emission

Taking into consideration that adverse impact of PAHs emission to air from vehicle engines affect negatively also soils along the roads, the own survey has been carried out in 1994 along the roads of different traffic intensity in two selected sites: **(G) –** the motorway

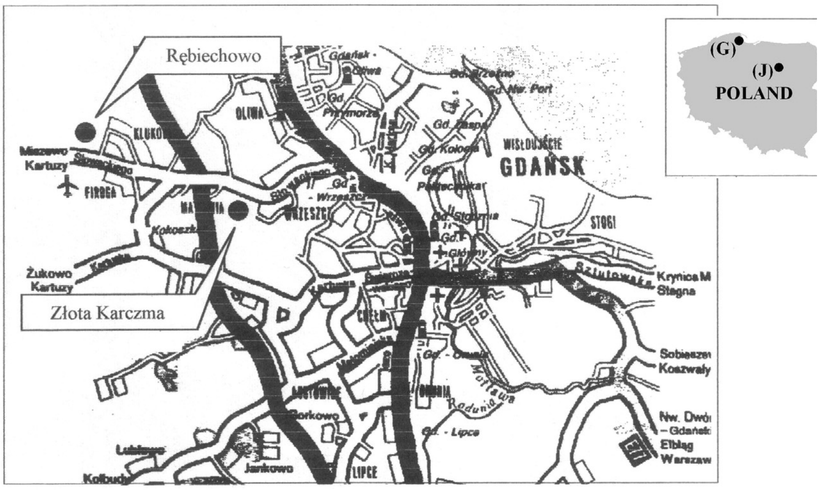

Fig. 1. (**G) –** the motorway connecting Gdansk with the Rembiechowo airport. Studied intersects location: (i) **Grb –** Rembechowo and (ii) **Gzk –** Zlota Karczma.

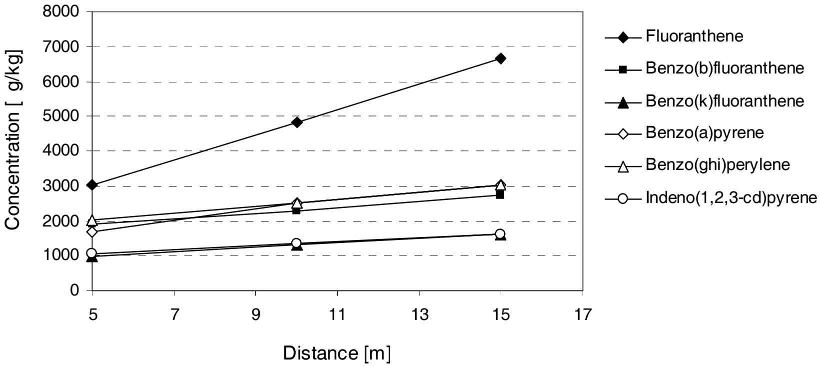

Fig. 2. Concentrations of PAHs along the **Grb** (Rebiechowo) intersect.

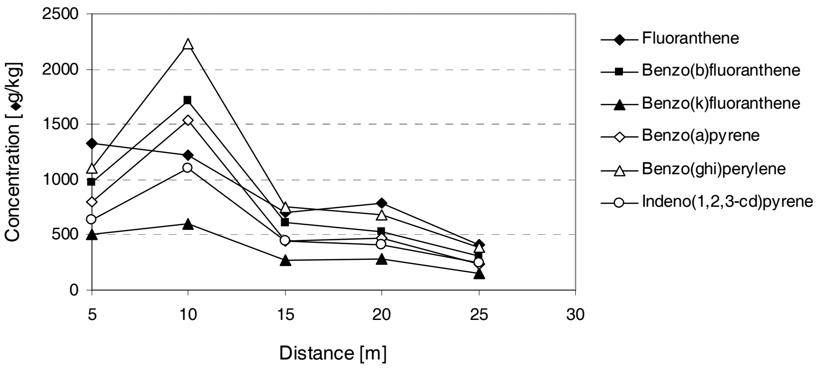

Fig. 3. Concentrations of PAHs along the **Gzk** (Zlota Karczma) intersect.

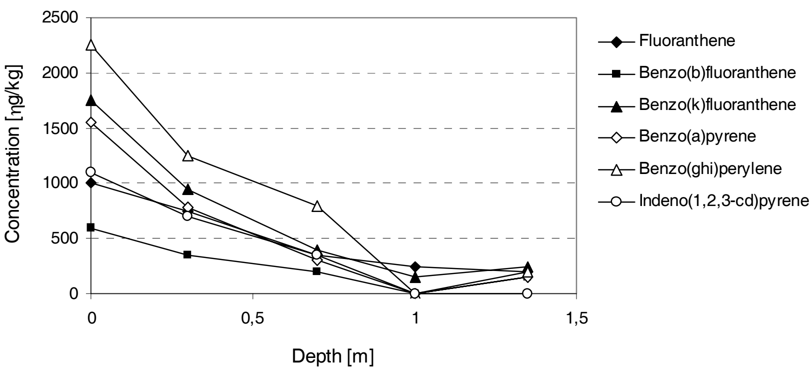

Fig. 4. PAH distribution pattern along the soil profile 0 -1.5 m (**Gzk** intersect, 10 m distance from the motorway)

connecting Gdansk with the Rembiechowo airport (Fig. 1-4), and for comparison (**J**) **-** along the local road in the almost pristine low-populated Jerutki rural area (Fig. 5-6). In the site (**G)** the soils from humus layer 0-20 cm were sampled in two intersects: (i) close to the airport (Rembiechowo, **Grb**) and (ii) in the vicinity of junction (Ziota Karczma, **Gzk**), in different distances from the motorway, up to 25 m from the road borderline. In the site (**J**), humus layer 0-20cm of soil horizon A, was sampled in 7 randomly selected points along the road.

For analysis, PAHs were extracted with dichloromethane in ultrasonic bath, with following centrifugation, evaporation of solvent in nitrogen stream, dissolution of the residue with methanol and filtration with use of 0.22мм syringe filter. For very complex samples, the extracts were additionally purified using SPE silica gel columns. In that case, the residue was dissolved in *n*-hexane. Analyte was applied at SPE column conditioned with *n*-hexane; as an eluent, *n*-hexane and *n*-hexane/ dichloromethane mixture 1:1 was used. The reproducibility of results was high (standard deviation <5%). In detail, the extraction procedure has been presented elsewhere (Twardowska and Kolodziejczyk, 1998). Recovery of 6 PAHs from soil samples ranged from 49% to 67%, depending on the constituent extracted, at standard deviation ranging from 2 to 5% (in particular, recovery of BaP was 47±5%, Fl 49±3%, BbFl 64±4%, BkFl 49±2%, BghiP 35±2% and IP 67±5%).

Soil samples were analyzed for 6 PAHs (according to the elution order): fluoranthene Fl, benzo[b]fluoranthene BbF, benzo[k]fluoranthene BkF, benzo[a]pyrene BaP, benzo[ghi]perylene BghiP and indeno[1,2,3-cd]pyrene IP with use of high performance liquid chromatography HPLC (Hewlett-Packard 1090 with time-programmed fluorescence detector HP 1046 A and integrator HP 3392A (detection limit <1мg/kg) (reference method).

Table 2.

Share of transport in gaseous and particulate emission to air in Poland in 2002 (after GUS, 2004)*

Emission source	Particu-Lates	SO_2	NO_x	CO	VOC	NH_3	GHG		
							CO_2	CH_4	N_2O
	In Gigagrams (% total)								
TOTAL	**473 (100%)**	**1456 (100%)**	**796 (100%)**	**3410 (100%)**	**898 (100%)**	**325 (100%)**	**308276.9 (100%)**	**1799.6 (100%)**	**73.0 (100%)**
Transport total	63 (13.3%)	43 (2.9%)	330 (41.5%)	756 (22.2%)	172 (19.1%)	-	29552.5 (9.6%)	4.8 (0.3%)	2.0 (2.7%)
Automotive transport	54 (11.4%)	35 (2.4%)	237 (29.8%)	636 (18.7%)	126 (14.0%)	-			

* Source: Data of the National Center of Emissions Inventory approved by Minister of Environment

Share of Automotive Transport in Pollutant Emission to Air

Gas and particulate emission

Transport, in particular automotive one, is a serious source of air pollution with gaseous and particulate emission, though its share in the particular gases emissions is uneven (Table 2). Transport is a major source of NO_x emissions and is responsible for about one fifth of CO and VOCs emitted into the air from various sources, while its share in SO_2 emission is marginal. Emission of major greenhouse gases GHG in 2002 comprised 30273.3 Gg i.e. 8.2% of total annual emission expressed in CO_2 equivalent, of this CO_2 was the predominant GHG being emitted by transport: its share in the total GHG balance was close to 10%, while the emission of N_2O and CH_4 was insignificant.

Automotive transport plays the major role in the emissions of air pollutants from transportation means (Table 2). It should be noted that in Poland the role of transport as a source of pollutant emission is somewhat suppressed by the emissions from power/thermal plants due to absolute domination of coal as a fuel for generating electricity (94.7%); in other countries less dependent on coal the share of pollutant emission, in particular of CO_2 from the automotive transport in total emission can be higher.

Heavy metals and organic pollutants

Emission of heavy metals from automotive transport in total balance appears to be minor compared to other sources, of which fuel burning processes in households and industry are the major sources of heavy metal emission (Table 3). Of all estimated metal emissions, that of lead was the highest.

Table 3.

Share of automotive transport in total heavy metal emission in Poland in 2002 (after GUS, 2004)

Emission source	As	Cr	Zn	Cd	Cu	Ni	Pb	Hg
	Mg (% of total)							
TOTAL	**49.4**	**55.5**	**1639.0**	**48.7**	**388.2**	**257.8**	**588.0**	**19.8**
Transport total	- -	2.3 (4.1%)	- -	0.3 (0.6%)	3.1 (0.8%)	5.3 (2.0%)	19.1 (3.2%)	- -
Automotive transport	- -	2.2 (4.0%)	- -	0.2 (0.4%)	2.6 (0.7%)	4.4 (1.7%)	18.6 (3.16%)	- -

Source: Data of the National Center of Emissions Inventory approved by Minister of Environment

The load of persistent POPs emitted from transport is relatively low compared to the major sources of emission that are primarily fuel burning in households, and to the lesser extent incineration processes in the industry. Of emitted highly toxic organic contaminants, though, the share of BaP appeared to be the most significant and in 2002 comprised 3.9% of total emission (Table 4).

It should be added that the lead emission from the road vehicles is unevenly distributed in the area, and shows the highest intensity, depending upon the traffic density, along the roads. The emission of polycyclic aromatic hydrocarbons (PAHs), in particular of the most toxic compound benzo[a]pyrene from automotive transport is a source of health hazard in the thickly populated urban areas and in the vicinity of highways and motorways, and close to the cross-roads.

Table 4.

Share of automotive transport in total POPs emission in Poland in 2002 (after GUS, 2004)

Emission sources	PCDD/PCDF		PAH			
			4 PAHs		BaP	
	Mg TEQ[a)]	%	kg	%	kg	%
TOTAL	**433365.9**	**100.0**	**160104.0**	**100.0**	**46004.0**	**100.0**
Transport total	717.2	0.1	1829.0	1.1	1823.5	3.9
Automotive transport	638.2	0.1	1349.5	0.8	1345.4	2.9

a) TEQ – Toxic equivalent; [b)] Sum of 4 WWA

Source: Data of the National Center of Emissions Inventory approved by Minister of Environment

Temporal Trends of Pollutant Emission from Automotive Transport

Dynamic growth of the vehicle number in

the last decade of XX century, with parallel over 2-fold increase of cars number and their share from 58% in 1990 to 71% in 2003 in total vehicle fleet (Table 1) did not result in the proportional increase of pollutant emission as it might have been expected. Moreover, by the end of the century (in 2000), either moderate increase of emissions from all means of transport (CO_2.for 34.5%, SO_2 for 9.1% ; particulates for 9.0%) or significant decrease (VOC for 37.7% , CO for 31.3%, CH_4 for 28.7%, NO_x for 13.5%) has been observed, with an exception of N_2O emission that showed an increase almost proportional to the increase of vehicle number (for 65.1%). A particularly striking decrease of emission occurred with respect to lead (93.6%) (Table 5).

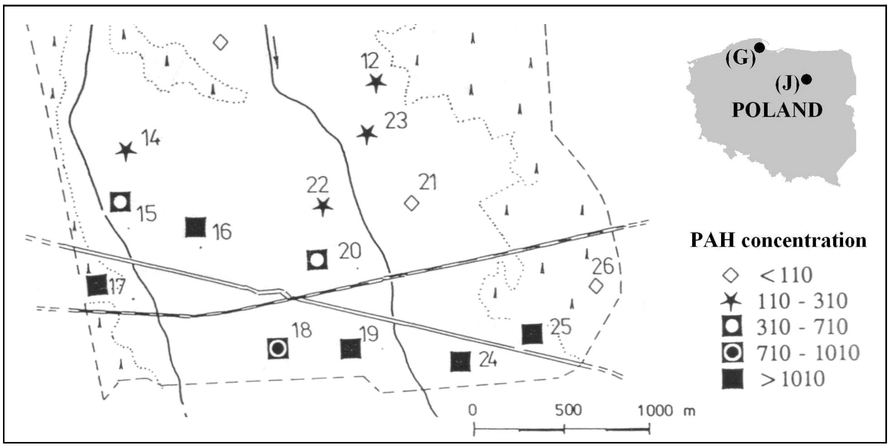

Fig. 5. Soil (0-20cm) sampling points for PAHs along a local road in Jerutki (**J**) site.

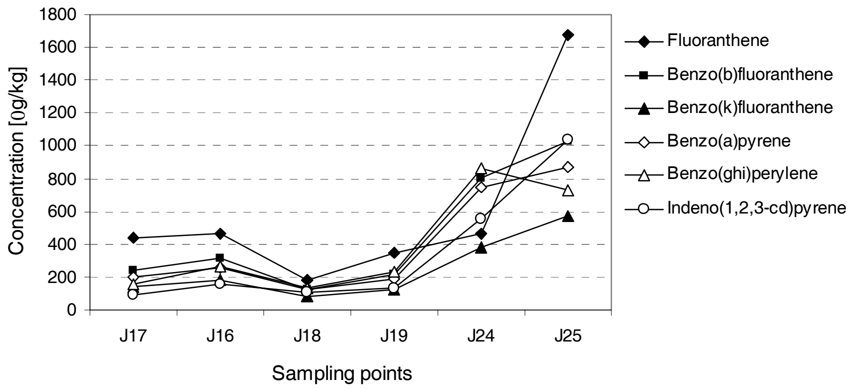

Fig. 6. Concentrations of PAHs in soil (0-20 cm) along the local road in the pristine Jerutki (**J**) area.

The statistical data for automotive transport available since 1998 show even deeper decrease of emissions of all pollutants, in spite of the fact that this area of transport underwent the most significant quantitative changes. In particular, emission of GHG in 1998-2002 declined by 10.4% for CO_2 and 37.4% for CH_4 , while N_2O showed only +10.5% growth. Reduction of other gaseous pollutants was as high as 14.8% NO_x, 30.4% CO, 43.2% VOC, and 68.6% SO_2, while reduction of particulates accounted for 18.9%. Lead emission from automotive transport in this period decreased by 87.2% (Table 5).

Such significant reduction of emissions from automotive transport despite of its quantitative increase was due to the deep qualitative changes supported by a stringent regulations that resulted in parallel modernization of the vehicle fleet, at present equipped with efficient engines, catalysts as a compulsory equipment, and a promotion of unleaded gasoline in the fuel market.

Also construction of ring roads, two-level crossings and speedways reduces significantly the direct health impact of dense traffic. The adverse impact of emissions from automotive transport in urban areas on the human health cannot be, though, thoroughly eliminated due to the old urban infrastructure in the city centers.

Nevertheless, the successful efforts directed to the reduction of emissions from the automotive transport contribute to the visible improvement of the air quality in Poland, both with respect to reduction of GHG in accordance with Kyoto Protocol, that obliges Poland to reduce GHG emission by 6% (1990 to 2008/2012) (WCI, 2005), and also regarding direct health impact on humans.

Traffic Impact on PAH Concentrations in Soil

Motorway with dense traffic

PAH burden of soil predominantly results from aerosols deposition and washout from the atmosphere, also of those emitted from the vehicle engines. Assessment of 6PAHs, among them of BaP in the surface (0-20cm) soil layer in two intersects along the motorway from Gdansk to airport (Fig 1) confirmed that dense road traffic resulted in high PAH enrichment in soils (Fig. 2, 3).

In the humus soil layer of both intersects the concentrations of analyzed standardized PAHs (BaP, Fl, BghiP) (Directive of Minister of Environment, 2002) exceeded from over an order (Fl, BghiP) up to two orders of magnitude (BaP) MCL in soils of A (protected areas) and B groups (agricultural, forest and urban areas) in all points, falling within concentration range for C group (industrial and traffic areas) (Fig. 2).

In **Gzk** (Ziota Karczma) intersect maximum concentrations were found to occur 10 m from the motorway, where the contents of BaP, similarly as in **Grb** intersect, were two orders of magnitude higher, while Fl and BghP exceeded maximum concentration levels MCL over an order of magnitude (Fig. 3).

Further decrease with the distance was observed, though even 25 m from the motorway the concentrations of standardized PAHs in the humus layer 0-20 cm exceeded MCL for soils of A and B group. Along this intersect, concentration of BaP in soil layer 0-20 cm ranged from 239 мg/kg (25 m from the motorway) to 1539 мg/kg (10 m from the motorway), at MCL for A class soils 20 мg/kg, and for B class 30 мg/kg. The surveyed soils were found to belong to class C (MCL for this class in 0-2 m layer is 5000 мg BaP/kg d.m., and 250000 мg/kg s.m. for a sum of 10PAHs).

The concentrations of all analyzed PAHs showed gradual decrease with depth along the soil profile, though the PAH concentration pattern *vs* depth proved vertical migration of these compounds and enrichment also in deeper soil layers (Fig. 4).

Local road in the rural area

PAHs burden of 0-20 cm soil layer along the local road in the almost pristine Jerutki area (Fig. 5) appeared to be comparable with the concentrations recorded along the Gdansk motorway with dense traffic that can be attributed to the kind of road users (tractors, harvesters, other agricultural machines, trucks etc. using diesel fuel). BaP concentrations in analyzed soil samples ranged from 128 to 874 мg/kg s.m., i.e. from several times to over 30-40 times higher than MCL for A and B class (20 and 30 мg/kg, respectively).

Conclusions

Increase of automotive transport by 76%, in that more than duplication of car number since 1990 has not resulted in the proportional increase of pollutant emission. Due to modernization of car fleet and motorways, and stringent environmental regulations, total emissions from transport in the decade 1991-2000 appeared to be much lover than its quantitative increase and since 1998 displayed decrease with respect to practically all monitored parameters.

Long-term emissions of PAH from automotive transport resulted in accumulation and vertical migration of PAHs in soil along the motorways and local roads that disqualify this soils for agricultural use.

References

Directive of the Minister of Environment of 9 September 2002 on standards of soil quality and standards of subsoil quality. *Dz.U.*02.165.1359 of 4 October 2002.

GUS – Central Statistical Office: *Environment Protection 2004. Information and Statistical Studies*. Statistical Publishing Establishment, Warsaw 2004, pp. 508 (*in Polish*).

Twardowska, I., and Kolodziejczyk, A.M.: Benzo[a]pyrene in soils and ground water: occurrence, sources, distribution, interrelation. *Toxicol. Environ.Chem.* 66:127-144 (1998).

WCI : Kyoto is coming. *Ecoal,* 52:1-6 (January 2005)

Abstract

POLLUTANTS EMISSION FROM THE AUTOMOTIVE TRANSPORT EXEMPLIFIED IN POLAND SINCE 1990

Irena Twardowska and Joanna Kyziol

On the background of statistical data on the dynamic quantitative growth of automotive transport (ATM) that has more than doubled since 1991, temporal changes and a share of gaseous and particulate emissions from this source in total emission have been analyzed. Most of the emissions, of which major ones were NO_x, CO, VOC and CHG, were found to decrease substantially due to the stringent environmental policy. Survey of 6 Polycyclic Aromatic Hydrocarbons (6PAHs) along a motorway and a local road displayed high accumulation of benzo[a]pyrene BaP and other PAHs in soil (0-20 cm) and their vertical migration to deeper layers. Concentrations of PAHs in soil, both along a motorway and a local road appeared to exceed MCL in soils of A class (protected areas) and B class (agricultural soils) for one-two orders of magnitude that disqualifies these lands for agricultural use.