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COMPARISON OF BENZENE AND ITS ALKYLATED DERIVATES PROFILES IN CAR'S FUELS, ENGINE EXHAUST GASES AND IN AIR IN THE VICINITY OF COMMUNICATION ARTERIES

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Abstract: Main sources of emission of benzene and its alkylated derivatives to the atmospheric air, particularly the vehicular ones, are characterized in the paper. Growing use of fuels, especially unleaded gasoline, leads to emission of considerable amounts of these aromatic hydrocarbons. The paper presents and discusses results of determinations of the content of benzene and its alkylated derivatives in engine fuels. Also emission factors of these hydrocarbons from commonly used in Poland types of cars, both with spark- and self-ignition engines, are considered. Profiles of investigated hydrocarbons in fuels and exhaust gases were determined. Respectively compared profiles proved the motor vehicles responsible for main part of the benzene and its alkylated derivatives in air in the vicinity of communication arteries.

INTRODUCTION

Benzene and its alkylated derivatives: toluene, xylenes, ethylbenzene, isopropylbenzene and trimethylbenzenes are monocyclic aromatic hydrocarbons. They belong to the volatile organic compounds (VOCs). In the atmosphere, in favorable conditions, the tropospheric photo-chemical ozone may arise from VOCs [3]. Methane, due to its considerably higher concentrations in the air, is very often considered separately; other VOCs are then called non-methane volatile organic compounds (NMVOCs). In the 15 Europe Union countries, in 2002, the total emission of NMVOCs was 9.6 million Mg, in Poland – 0.9 million Mg [13].

Road transport and usage of solvents are the most important anthropogenic sources of NMVOCs emission in majority of European countries: e.g. in Great Britain, Germany, France, Norway, Finland, Denmark [9]. The third dominating source is extraction, production and distribution of fuels. Three groups of sources: usage of solvents, road transport and combustion processes in the municipal and living sectors are responsible for the anthropogenic emission of NMVOCs in Poland. These sources contribute over 69% to the total emission [13].

The greatest amounts of NMVOCs arise from the combustion of liquid fuels. For example, in Great Britain such NMVOCs constitute 86.5% of the total emission of these compounds [14]. The greatest amounts of vehicular NMVOCs come from gasoline fuelled vehicles (77% in 1997), smaller from Diesel engines (19%) and the smallest from liquid gas fuelled cars (3%) [12]. Nearly 60% of the vehicular NMVOC emission comes from light (passenger) cars [13].

The emission of vehicular NMVOCs has been systematically decreasing for the recent two decades, e.g. in Great Britain in 2000 the emission was 2.5 times lower than in 1990 and since 1999 the emission has been systematically reduced by 15% every year [5]. Average NMVOC emission drop in the EU countries in the period 1990–1999 was 42% [8]. In Poland, from 1990 to 2000, emission was reduced by half and between 2000 and 2003 by 25% [14, 21]. Simultaneously, the number of vehicles was continuously increasing. In Poland, from 1990 to 2004, the number of vehicles almost doubled, the number of passenger cars increased 2.3 times [14] and is still growing [21]. The simultaneous occurrence of two opposing trends: increasing vehicle number and decreasing NMVOC emission is due to improvement of technical condition of vehicles and changes in quality of engine fuels. Newer cars are equipped with catalytic converters of exhaust gases, they more efficiently utilize fuel and their engines may be fuelled with liquid gas, what significantly reduces emission of vehicular pollutants.

Due to relatively high concentrations of VOCs in the atmospheric air, especially in vicinity of communication tracts and communication supporting utilities, decreasing trends in emission of VOCs from communication sources do not prevent hazard from high both toxicity of some VOCs and photochemical reactivity of others.

For some recent years, among all VOCs, benzene and its alkylated derivatives have drawn attention of investigators. These VOCs affect the natural environment and living organisms both indirectly, by contributing to formation of ozone and photochemical smog, as directly. In the direct influence, benzene, as a carcinogenic, mutagenic, teratogenic, embryotoxic and generally toxic to the blood-forming system compound, destroying also the immune system, is the most dangerous VOC [23]. The other hydrocarbons are less harmful in direct effect on humans. Nevertheless, toluene, ethylbenzene and xylenes, along with benzene, are among the 21 substances considered to be toxic compounds coming from vehicles with internal combustion engines [9].

Mainly, benzene, but their alkylated derivative too, is emitted from combustion of liquid fuels. For instance, in Great Britain, as much as 65% of emitted to the air benzene comes from this source [5]. In Great Britain, nine out of fifty emitted in the greatest amounts chemical compounds are monocyclic aromatic hydrocarbons (benzene, toluene, xylenes, ethylbenzene, and trimethylbenzenes). Motor cars are the main source of majority of these hydrocarbons [18]. The emission amount and kind of hydrocarbons in car exhaust depend on construction and exploitation properties of engines as well as on a kind and composition of fuel.

Emission of benzene and its alkylated derivatives comes mainly from spark ignition engines because these compounds are components of gasoline. Considerable amount of aromatic hydrocarbons, also benzene and its alkylated derivatives, in gasoline is due to limitations imposed on using of anti-knock lead compounds. To maintain properly high octane number, increased content of high-octane components in gasoline, such as the aromatic hydrocarbons, appeared necessary. Now, due to harmfulness of some aromatic hydrocarbons, especially benzene, measures are being undertaken to limit their usage as anti-knock additives to gasolines. It is expressed by successive introducing of more and more restrictive standards for gasoline composition.

Since 2002, permissible content of benzene and aromatic hydrocarbons in Polish gasolines has been the same as in gasolines used in the EU countries. Till then, the quality standards for the Polish gasolines essentially differed from requirements set for the European fuels. The present European standards are still less rigorous than the American, and especially Californian, ones.

Lowering benzene content in gasolines does not suffice to lower its emission in exhaust gases. As it is known from investigations, benzene arises as a product of chemical transformations of its alkylated derivatives during processes of fuels' combustion [17, 25].

OUR OWN STUDIES

Outline and scope of the investigations

To evaluate emission of benzene and its alkylated derivatives from vehicular sources, taking account of quality of Polish gasolines and number of vehicles with catalytic converters, determinations of concentrations of these compounds in gasolines, car exhaust gases and in atmospheric air in vicinity of roads were performed. The cause-effect relationship between composition of fuels, composition of exhaust gases and air pollution due to road traffic was assumed to be reflected in profiles of investigated compounds in fuel, exhaust gases and in the air. Nine monocyclic aromatic hydrocarbons were investigated: benzene, toluene, m,p-xylene (determined together), o-xylene, ethylbenzene, isopropylbenzene, 1,2,4-trimethylbenzene and 1,3,5-trimethylbenzene. On the basis of received results, the emission factors for the investigated substances and motor vehicles were determined.

Methods

Samples of exhaust gases were taken during tests conducted on a chassis dynamometer SCHENCK 500GS/60 at the Exhaust Emissions Laboratory of the BOSMAL Automotive Research and Development Centre in Bielsko-Biala.

The cars selected for exhaust sampling were conditioned for 18 h in closed compartment, at temperature 22°C and relative air humidity 50%. The pollutant concentrations were determined for conditions of the so called "cold start". Sequential sample taking of exhaust gases and their dilution with air in 150 dm^3 Tedlar bags by using the AVL CEC-Q20 CFV-CVS sampling system was performed during the New European Driving Cycle driving test (NEDC) according to the valid investigating procedures. The methodology provides separate averaged samples for both the urban driving cycle (UDC) and the extra-urban driving cycle (EUDC).

Concentration of total hydrocarbons, recalculated into atoms of carbon (C), was measured with the use of a flame ionization detector (FID) calibrated with propane.

Exhaust gases were sampled by using glass sorption tubes filled with Tenax GR. After thermal desorption, they were analyzed with the use of a Hewlett-Packard 5890 gas chromatograph with FID having an RTX-5, 30 m × 0.32 mm, capillary column with a 1.3 μm film stationary phase (Resteck).

Concentrations of benzene and its alkylated derivatives in exhaust gases were determined for six selected car brands differing with engine piston displacement, mileage and technical condition. Two of them are obsolete construction cars, but still met on Polish roads. Characteristics of the examined cars are presented in Table 1.

Table 1. Characteristic of the tested cars

Brand	Engine cubic capacity [cm^3]	Ignition type	Age [years]	Mileage [km]	Feeding system	After treatment system	Fuel
Fiat Palio Weekend	1600	SI	new	150	Injection	Catalytic converter	Gasoline
Fiat Seicento Young	900	SI	new	70	Injection	Catalytic converter	
Honda Civic	1500	SI	4	76000	Injection	Catalytic converter	
Fiat 126p	650	SI	8	35000	Carburetor	None	
Polonez SLE	1500	SI	11	150000	Carburetor	None	
Ford Fiesta	1800	CI	4	97000	Injection	None	ON

To determine the content of the investigated compounds and total aromatic hydrocarbons, gasoline (ethyl gasoline ET94, unleaded Euro Super 95, Euro Super Plus 98, universal U95) and petroleum diesel were chromatographically analyzed. A gas chromatograph PE 8700 with FID and a capillary column WAX-EH having dimensions 60 m × 0.35 mm × 1.5 μm was applied.

PRESENTATION AND DISCUSSION OF THE RESULTS

Car fuels

The chromatographic analyses of five fuel samples provided information on the content of benzene and its alkylated derivatives in various engine fuels used in Poland. The total content of benzene and its alkylated derivatives in the examined gasolines varied between 20.5% vol. and 28.5% vol. In the diesel oil, the examined compounds occurred in much lower amounts, they were only 2.7% of the fuel volume. The greatest contributions to the fuels, in descending order, were observed for: toluene, m,p,o-xylene, benzene and 1,2,4-trimethylbenzene.

The content of benzene in the gasolines was from 1.96% to 2.33% vol. Also high content of toluene was found. Dealkylation of toluene in the gasoline combustion process may contribute to formation of benzene in exhaust gases. The content of benzene in diesel oil was on average 6 times lower than in gasolines and was equal to 0.34% vol.

The content of benzene in examined gasolines was then lower than the permissible content, equal to 5.0% vol., defined in national regulations valid [22], but considerably higher than that defined in the European norm EN 228:1999, reverting to the Directive 98/70/EC, that permitted no more than 1.0% vol. content of benzene in gasolines [1].

Car exhaust gases

Benzene and its alkylated derivatives were found in exhaust gases from spark-ignition (SI) engines as well as in exhaust of compression-ignition (CI) engines. Considerably greater amounts were in exhaust of SI engine cars, especially the cars without catalytic converters. CI engine cars contribute to emission of benzene and its alkylated derivatives in a significantly lower degree than SI engine cars, even than the SI engine cars with catalytic converters.

The concentrations of benzene and its alkylated derivatives measured in exhaust gases were used to determine emission factors and mass contribution of these compounds to the exhaust. They were determined on the basis of the computed values of adequate corrected concentrations. The emission factors were determined for all driving cycles: UDC, EUDC and also for the whole NEDC. Examples of the emission factors are presented in Figure 1, where the vertical axis is scaled logarithmically.

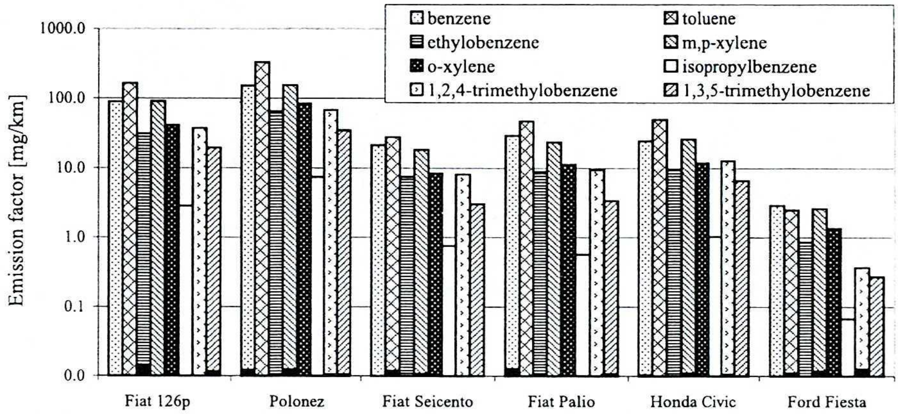

Fig. 1. The emission factors for benzene and its alkylated derivatives emitted from car vehicles in the urban driving cycle of the NEDC test for cold start (logarithmic scale)

The emission factor profiles for all the SI engine cars are similar; the profiles for SI and CI engine cars differ. From among all examined compounds toluene, xylenes and benzene were emitted in the greatest amounts. The mass percentage of benzene and its alkylated derivatives in total determined hydrocarbons were computed on the basis of the analysis of exhaust gases. The contributions of all SI engine cars were similar while contributions of SI and CI engine cars differed.

Comparison of the emission factors proved at least 5 times lower in emissions of the examined compounds from the SI engine cars with than without catalytic converters. Also, exhaust gases from the SI engine cars with catalytic converters comprised about 10 times more examined hydrocarbons than the CI engine cars without the converters.

The driving tests showed considerably lower efficiency of catalytic converters in varying conditions of an urban cycle than in stable conditions of an extra-urban cycle.

The factors for emission of benzene and some of its alkylated derivatives from cars [6, 7, 11, 15, 26, 27], published worldwide, differ from each other. They refer to new cars tested in the homologation process or for marketing purposes, or they are averaged factors for vehicles of various engine cubic capacity, mileage, feed system, kind and condition of exhaust gas purification system, using fuels of various composition. Therefore, factors from investigations carried out in quite specific national conditions cannot be compared with the factors from literature. In the authors' opinion, presented investigations are an original contribution to knowledge on the contamination of atmospheric air with BTX in countries where motorization develops fast and where, although transiently, obsolete cars that are in bad condition dominate.

Comparison of profiles of benzene and its alkylated derivatives in car fuels, car exhausts and atmospheric air

The computed mass content of benzene and its alkylated derivatives in the total hydrocarbons in exhaust gases and car fuels allowed for comparing the received profiles of the content (Fig. 2).

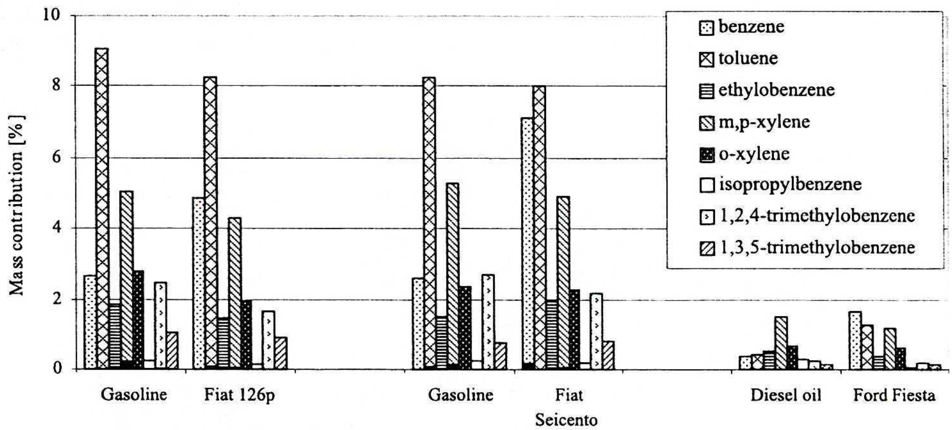

Fig. 2. Profiles of mass content of benzene and its alkylated derivatives in car fuels and car exhaust gases

There is clear similarity of the profiles of the examined compounds in gasolines and exhaust gases for SI engine cars. It is worth mentioning that the benzene mass content in exhaust gases for all investigated SI engine cars is greater than its content in fuel, what confirms the opinion that some part of emitted benzene comes from dealkylation of its alkylated derivatives and also that its conversion in a catalytic converter is relatively less efficient. Supposition that benzene and toluene may arise from the dealkylation reaction may be confirmed by comparing the profiles for CI engine exhaust and diesel oil.

To determine if the vehicular emission affects the air pollution in vicinity of roads, profiles of relative concentrations of benzene and its alkylated derivatives in car exhaust and in the atmospheric air near communication arteries were compared. The relative concentrations were computed by relating concentrations of particular alkylated derivatives to the benzene concentration assumed to be 1. The received profiles are presented in Figure 3.

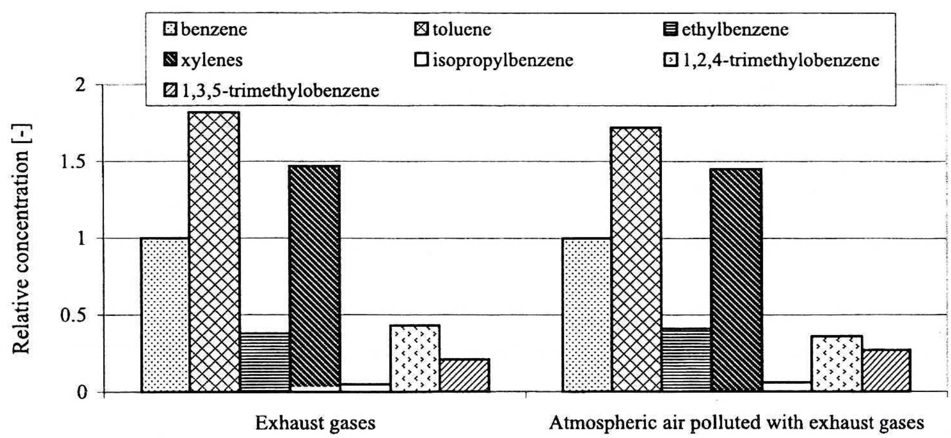

Fig. 3. The profiles of relative concentrations of benzene and its alkylated derivatives in car exhaust gases and atmospheric air

The BTX profiles were determined by computing the weighted arithmetic mean of relative concentrations in exhaust gases from cars with Spark Ignition and Self Ignition engines. The means were weighted by using percentages of SpI and SeI cars in the total of the Silesian cars. The BTX profile of the atmospheric air was determined by computing the arithmetic mean of the relative concentrations of BTX in air at 11 measuring points in Gliwice, Katowice Bytom and Zabrze. Profiles presented in Figure 3 refer to urban traffic. Relative concentrations of BTX in car exhaust were computed from an analysis of flue gases taken during an urban driving cycle. Relative concentrations of BTX in air were determined by analyzing air from measuring points located in the city centers, close to communication arteries, at crossroads, street canyons and built over vicinities of roads [30].

A comparison of the profiles shows their considerable similarity. The differences observed for some compounds may be due to the process of mixing exhaust gases with previously polluted air, in which content of some hydrocarbons, particularly highly reactive compounds such as isopropylene and trimethylbenzenes, changed due to photochemical reactions.

The computed relative concentrations in the atmospheric air may be compared with results of other measurements from over the world [2, 4, 16, 19, 20, 24, 28, 29]. The comparison shows closeness of the profile of investigated substance concentrations in air to those received in Krakow [16] and Szczecin [28], where the proportion toluene/benzene is lower than 2. In other countries, the toluene/benzene proportion of concentrations is higher. It may be due to different structure of the motor vehicle fleet or composition of fuels. Another comparison was yet done. Namely, the profiles of relative concentrations of benzene and its alkylated derivatives in air in vicinity of roads and in air near industrial, non-vehicular, sources of emission of these compounds were compared. The profiles are presented in Figure 4.

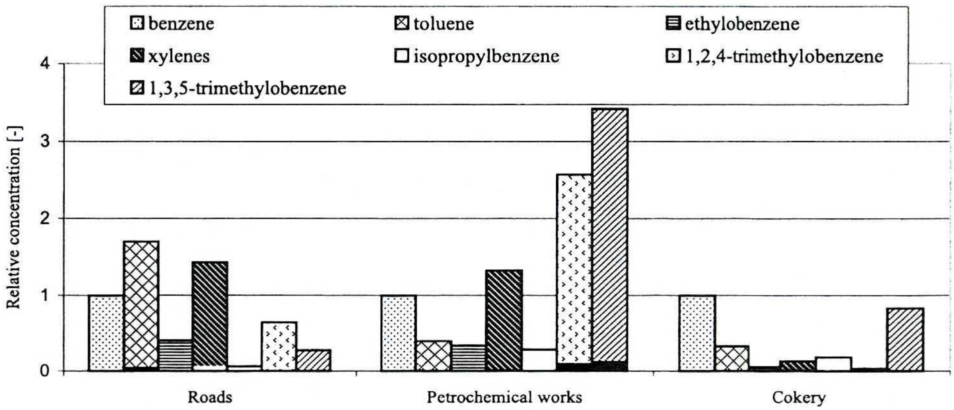

Fig. 4. The profiles of relative concentrations of benzene and its alkylated derivatives in air in neighborhood of various emission sources

The profiles are essentially different. It proves that the levels of concentrations of benzene and its alkylated derivatives in neighborhood of communication tracts are due to vehicular sources [30]. According do the authors' knowledge, the presented above profiles and their interpretation are the original results of the performed investigations and therefore cannot be compared with data from the literature.

CONCLUSIONS

The internal combustion car engines are an important source of emission of benzene and its alkylated derivatives. Mostly, the pollution of these compounds comes from car vehicles equipped with spark-ignition engines. The concentration of benzene and its alkylated derivatives in exhaust of spark-ignition engine cars without catalytic converters is about 50 times greater than in exhaust of compression-ignition engine cars.

Application of catalytic converters of exhaust gases allows for above fivefold on average lowering of total emission of benzene and its alkylated derivatives.

The mass content of benzene in exhaust gases is distinguishably higher than in fuel, while shares of its alkylated derivatives in fuel and in exhaust gases are similar. Probably, it is due to the process of dealkylation of the benzene homologues leading to formation of benzene in combustion of fuel-air mixture in an engine.

The profiles of concentrations of benzene and its alkylated derivatives in car fuels, car exhaust gases and atmospheric air in vicinity of communication arteries are very similar. It proves that car vehicles are the main source of emission of these compounds near roads.

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PORÓWNANIE PROFILI BENZENU I JEGO ALKILOWYCH POCHODNYCH W PALIWACH, SPALINACH SILNIKOWYCH I POWIETRZU ATMOSFERYCZNYM W SĄSIEDZTWIE SZLAKÓW KOMUNIKACYJNYCH

W artykule scharakteryzowane zostały główne źródła emisji benzenu i jego pochodnych alkilowych do powietrza atmosferycznego, ze szczególnym uwzględnieniem źródeł motoryzacyjnych. Wzrost zużycia paliw silnikowych, a zwłaszcza benzyn bezołowiowych, prowadzi do emisji dużych ilości tych aromatycznych węglowodorów do powietrza atmosferycznego. W artykule zaprezentowano i omówiono wyniki badań własnych dotyczących określenia zawartości benzenu i jego pochodnych alkilowych w paliwach silnikowych oraz wskaźników emisji tych węglowodorów z powszechnie eksploatowanych w Polsce typów pojazdów samochodowych z silnikami z zapłonem iskrowym (ZI) i zapłonem samoczynnym (ZS). W oparciu o uzyskane wyniki określono profile badanych węglowodorów w paliwach silnikowych i spalinach z silników samochodowych. Porównanie odpowiednich profili dowiodło, że pojazdy samochodowe są głównym źródłem emisji benzenu i jego pochodnych alkilowych w rejonach tras komunikacji samochodowej.