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Volatile Organic Compounds in Private Cars and Public Vehicles

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1. Introduction

The quality of air is a significant parameter, affecting people's health and well-being. This concerns both the indoor and outdoor air, although for quite a long time it was the atmospheric air that attracted researchers' concern. The change occurred with the development of civilization and increasing time spent indoors. By indoor air we understand all closed environments, which are not under regulation related to occupational safety. Studies on indoor air have been focused mainly on the places of residence, offices, schools and education premises.

But in the modern societies, the automobile cabin is an important part of the leaving environment. In many countries, time spend on return commute to a workplace is growing and easily exceed one hour. Statistics on the USA citizens way of living show that they spend 87% of their time indoors, 8% outdoors and 5% in transportation (by car, bus, train or plane), and in Europe similar tendencies are observed: 90% indoors, 6%

outdoors and 4% transportation for French people [1]. There is not such an evaluation for Poland, but due to the increased traffic, time spend in either private car or public transport vehicle increased last years significantly.

Therefore the exposure from transport vehicles is of growing importance. For person whose occupation required longer periods to be spent inside a vehicle (policemen, taxi, bus and truck drives, servicemen, sales representatives), the relative contribution of in-vehicle exposures to overall is greater than 30%.

Research on indoor air quality in car compartments was rather scare until very recently. Toxic substances originate from interior materials, gasoline loss, infiltration of outdoor air pollutants. Some of published in-vehicles studies have identified elevated levels of many unleaded and diesel fuel related pollutants, such as volatile organic compounds, carbon monoxide, and particulate matters compare to other indoor environments [16].

Yoshida and Matsunaga [31] identified over 160 substances in the cabin of the new car, including BTEX and formaldehyde, but their research was limited to one private vehicle only. More research was done by Zhang et al. [32] in unoccupied, parked cars. The main gaseous substances identified were: benzene, toluene, ethylbenzene and xylenes (BTEX). The same substances are found to show the higher levels in the buses in China [3], Hong Kong [17], Taiwan [12] and Spain [25]. Concentrations were strongly influenced by trip time and area (such as downtown compare with suburbs), so emission from fuel was the most probable dominant source.

Besides BTEX, eye irritants including a number of aldehydes, should be of special concern [29]. Due to the analytical methods some aldehydes are measured, although levels of only a few of them are regulated, due to known hazardous effects. National Institute for Occupational Safety and Health, USA (NIOSH) established recommended exposure limits (REL) in the work environment for some of them: formaldehyde (methanal), acetaldehyde (ethanal), valeraldehyde (pentanal) and acrolein (propenal) [21]. Only formaldehyde is regulated in most of countries (including Poland) but in workplaces, which does not include automobile compartments.

Methanal (formaldehyde) has been known as a major eye and nose irritant in indoor environment for many years. It is probably responsible for many health treats [11]. Some studies have suggested that people exposed to formaldehyde levels ranging from 50 to 100 ppb for long periods of time are more likely to experience asthma-related respiratory symptoms, such as coughing and wheezing [30].

Researchers agree that formaldehyde is sensory irritant but vary in the opinion about its cancerogenity [14]. Exposure to moderate levels of formaldehyde (hundreds of ppb or greater) can cause a number of irritant symptoms, including temporary burning of the eyes or nose, and a sore throat [29].

Acetaldehyde is a recognized eye and lung irritant. Nasal cancer connected to acetaldehyde exposure was confirmed in animals [21] therefore Occupational Safety and Health Administration, USA (OSHA) recommends that acetaldehyde be considered a potential occupational carcinogen.

Propenal (acrolein) has been recognized as pulmonary toxicant and listed among 188 the most hazardous air pollutants identified by U.S. EPA [26]. It can act synergistically with other carcinogents in the development of lung cancer. Studies linked acrolein with children asthma and lung cancer in women exposed to cooking fumes [18]. But nasal irritation appears to be the most sensitive respiratory effect.

Pentanal (valeraldehyde) may cause skin, eye, nose and respiratory system irritation. Other aldehydes, such as hexanal and fenylomethanal (benzaldehyde) irritate mucous membranes and respiratory system. Fenylomethanal (benzaldehyde) is also a skin irritant. Low levels can cause breathing problems or skin redness. Permanent exposure, on very low levels may cause headaches or chronic fatigue.

Pollutants from groups of aldehydes and ketones may cause numerous ailments, starting from a common headache, sleepiness, feeling of fatigue or irritation of mucous membranes, up to lesions of internal organs and even carcinogenic changes. The main sources of indoor air pollution by these substances include building and finishing materials, as well as the outdoor air and the users of compartments themselves.

These syndromes are widely described in the literature as sick building syndrome (SBS), but they were studied in the traditional indoor environments, what means internal spaces in the buildings.

No single chemical or biological pollutant can be seen as the origin of SBS, because many factors and substances, usually under measurement thresholds, act synergistically to cause this effect. However, examination of spaces in which occupants suffer from SBS included typical eye irritants, such as carbonyls, and these substances might be found in the automobile compartments.

Levels and sources of aldehydes (carbonyl compounds) have been widely studied in residential indoor areas, office environments and other public spaces [2, 4, 9, 10, 13, 20, 26]. Less is known about levels of carbonyl compounds in vehicles. Among over 160 compounds detected by Yoshida and Matsunaga [31] only formaldehyde belongs to the carbonyl group. He found formaldehyde concentration to be at 46.4 $\mu g/m^3$, but his studies were based on one car. Formaldehyde and acetaldehyde were measured by Jo and Lee [15] in 40 cars in Taegu, South Korea. Cars varied in engine volume, mileage and year of production. All of them were produced in Korea. Formaldehyde was within 8.8÷39.8 $\mu g/m^3$ (median 19.1 $\mu g/m^3$), while acetaldehyde 1.4÷19.1 $\mu g/m^3$ (median 7.0 $\mu g/m^3$).

Knowledge about indoor air quality in cars is important, as substances causing eye irritation can cause lack of concentration and reduced work efficiency of occupants [8] and similar effects in car compartments may impact not only drivers well being but road users safety. Therefore the studies to evaluate levels of carbonyl compounds and their impact on the indoor air quality in public and private cars have been undertaken.

2. Materials and Methods

The concentrations of carbonyl compounds were measured in 10 private passenger cars and 10 buses owned by the Municipal Transport Company in Lublin. Lublin, the city of 400 thousand inhabitants suffers from traffic problems and a lot of car and bus users complain about growing amount of time spent in vehicles. Lublin is situated in south-eastern, less developed part of Poland and one of the poorest regions of EU. A lot of cars used by private owners are second hand cars, older than 10 years.

Private automobiles varied in the model, producer, kilometrage and air condition system. Details of the examined vehicles are gathered in Table 1.

All busses selected for the experiment were the one model buses, bought by the City of Lublin at the same time (2009) from the same producer and identically equipped. They served at different routes (lines) in the Lublin city, therefore small differences in the kilometrage (between 25 000 to 32 000 km) have been observed.

Table 1. Characteristic of the examined private automobiles
Tabela 1. Charakterystyka badanych samochodów osobowych

Car	Car code	Producer	kilometrage (km)	Age (years)	AC
1	C-A1	A	110000	6	climatronic
2	C-A2	A	280000	11	climatronic
3	C-B1	B	130000	7	climatronic
4	C-B2	B	50000	4	manual
5	C-C1	C	80000	4	climatronic
6	C-D1	D	240000	4	climatronic
7	C-E1	E	171000	6	climatronic
8	C-F1	F	190000	1	manual
9	C-G1	G	200000	10	manual
10	C-H1	H	180000	12	manual

Samples for measurements of eye irritants were collected during seven days of normal occupation (including the weekend). Aldehydes were sorbed using a passive sampling method on Radiello dosimeters (RAD165) (exposure time: 168 hours). Dosimeters were placed in drivers cabins (for buses) and in the front of the car cabin, near driver seat (for private vehicles). All measurements were performed in triplicate and repeated in two following weeks.

Adsorbed aldehydes were extracted with acetonitrile (HPLC grade) and analyzed using the RP HPLC method (Waters) with a Restek Allure AK column; acetonitrile/water elution and UV detection set at 365 nm.

The calibration of dosimeters was performed in a 5 L glass chamber under controlled conditions of temperature, air flow and aldehyde concentration. Detection limits were 0.1 $\mu g/m^3$ for formaldehyde and hexanal and 0.2 $\mu g/m^3$ for other carbonyl compounds. Blank concentrations of carbonyl compounds were lower than detection limits.

3. Results and Discussion

The performed studies allowed for the identification of 12 volatile organic compounds from the carbonyl group, including: methanal, ethanal, propenal, propanal, butanal, pentanal, hexanal, phenylomethanal, butane-2-n, 2-methyl-2-propenal and 3-methyl-benzaldehyde.

Eight carbonyl compounds: methanal (formaldehyde), ethanal (acetaldehyde), propenal (acrolein), propanal (propion aldehyde), butanal, pentanal (valeraldehyde), hexanal and phenylomethanal (benzaldehyde) were detected in all the examined vehicles, either buses or private cars. The data are presented in figures 1 and 2 for buses and automobiles, respectively.

Concentrations in buses were generally lower than in private vehicles. No big discrepancies were found between concentrations of target compounds in particular buses, as they were the same model buses, bought in 2009 from the same producer and similarly equipped. The concentration of eight aldehydes in the examined buses were within the range 5.0÷8.7 $\mu g/m^3$ (mean 6.6 $\mu g/m^3$) for methanal, 4.2÷10.3 $\mu g/m^3$ (mean 6.7 $\mu g/m^3$) for ethanal, 1.5÷3.6 $\mu g/m^3$ (mean 2.6 $\mu g/m^3$) for propenal, nd. – 1.5 $\mu g/m^3$ (mean 1.1 $\mu g/m^3$) for propanal, 3.2÷8.6 $\mu g/m^3$ (mean 5.5 $\mu g/m^3$) for butanal, 0.1÷3.3 $\mu g/m^3$ (mean 2.1 $\mu g/m^3$) for pentanal, 2.6÷5.5 $\mu g/m^3$ (mean 3.8 $\mu g/m^3$) for hexanal and 0÷0.1 $\mu g/m^3$ (mean 0.03 $\mu g/m^3$) for phenylomethanal.

Acetaldehyde (ethanal) concentrations measured in private cars showed the biggest discrepancies and varied from 0.8 to 44.1 $\mu g/m^3$, followed by butanal – from 4.5 to 51.6 $\mu g/m^3$. Formaldehyde, the most ubiquitous indoor carbonyl compound was found in the range 7.9÷28.3 $\mu g/m^3$.

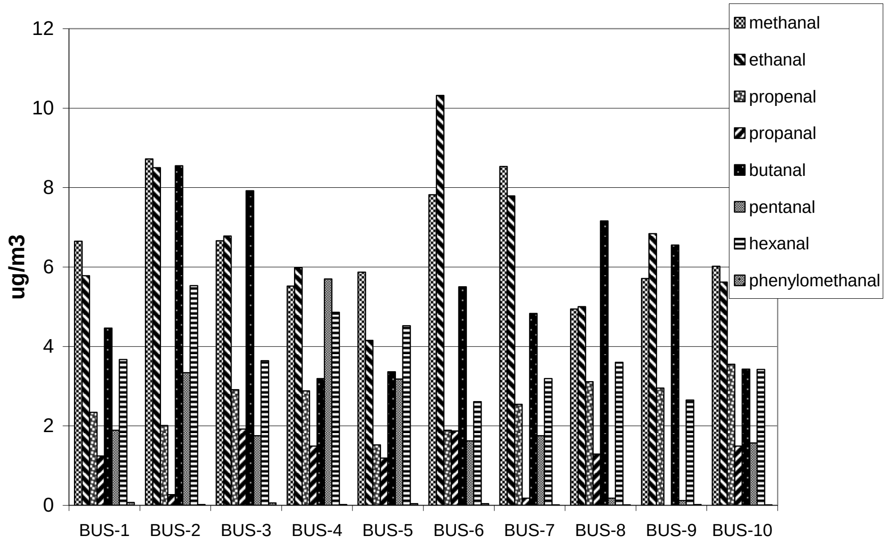

Fig. 1. Concentration of detected aldehydes in public buses in Lublin city
Rys. 1. Stężenia aldehydów w badanych autobusach miejskich w Lublinie

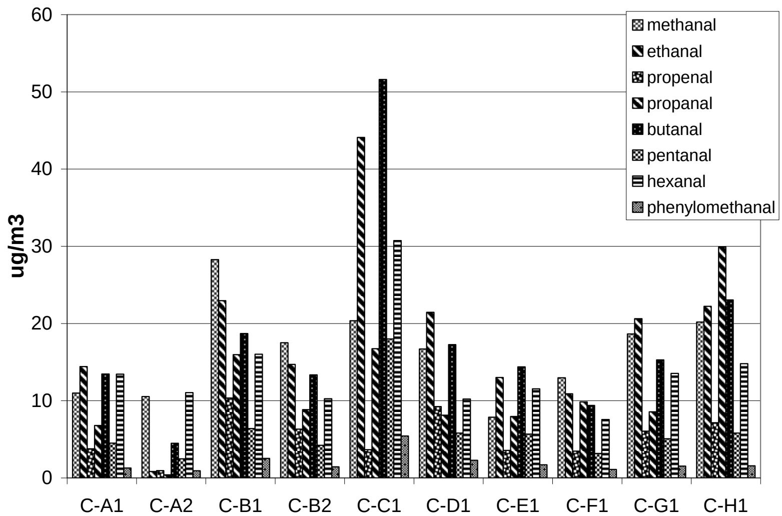

Fig. 2. Concentrations of detected aldehydes in private vehicles
Rys. 2. Stężenia badanych aldehydów w samochodach osobowych

Formaldehyde alone or in combination with other chemicals, serves a number of purposes in manufactured products. It is used to add permanent-press qualities to clothing and draperies, as a component of glues and adhesives, and as a preservative in some paints and coating products [22]. Therefore, sources of formaldehyde in indoor air include building materials, smoking, household products [27, 30]. Formaldehyde has been found to be a major secondary pollutant from cleaning products in the presence of ozone [27, 30]. In the car HCHO may originate from finishing materials, cigarette smoking and cleaning products.

The main indoor source of acetaldehyde (ethanal) is tobacco smoke, although particle board furniture and carpeting are also recognized sources [5].

Concentrations of acetaldehyde and formaldehyde in the examined vehicles may be caused by the fabric upholstery and the dyes used for its manufacturing. Lower concentrations of acetaldehyde were measured in cars with leather seat covering. This is in agreement with findings of Dassonville et al. [5] for homes, that slightly higher concentrations of formaldehyde and acetaldehyde were found in offices with wall-to-wall carpeting than those with panels.

Smoking is not permitted in public buses, although some of drivers are smokers. Similar situation occurs for private vehicles, as none of owners claim to smoke in the car. But environmental tobacco smoke might be a minor source of formaldehyde and acetaldehyde.

The National Institute for Occupational Safety and Health (NIOSH), USA has set the recommended exposure limit for formaldehyde to REL = 0.016 ppm, which might be converted to approximately 20 $\mu g/m^3$ [21]. The levels of formaldehyde permitted in non-occupied indoor spaces in Poland are set at 100 $\mu g/m^3$, while in occupied spaces at 50 $\mu g/m^3$ [23]. The current regulations are under evaluation, and the new standard is proposed to be set at 20 $\mu g/m^3$ for both occupied (residential) and non-occupied (workplace) spaces. The new value is in compliance with exposure limits recommended by NIOSH, however NIOSH REL values apply only to workplace hazards, whereby the term “workplace” is still understand in the traditional sense as, for example industrial sites with chemical emissions from processes and materials used.

Only in one car, formaldehyde concentration exceeded 20 $\mu g/m^3$. This car was four years old, with not very high kilometrage. Elevated concentrations of acetaldehyde (44.1 μg/m^3) were measured in this particular vehicle too, although acetaldehyde concentration was below the NIOSH REL (100 μg/m^3). Based on other results it might be presumed that textile finishing materials in this car were source of elevated levels formaldehyde and acetaldehyde.

Relatively high concentrations of hexanal (7.6 to 30.7 μg/m^3) were found in all of the automobiles examined. Hexanal is used as masking agent in cosmetics and fragrances. This is in agreement with our previous findings that hexanal in indoor spaces occupied by female subject using strong fragrances showed higher concentration then in offices occupied by male subjects [6]. Main users of every examined car were male, but concentrations of hexanal were higher in the "family cars" and cars with fragrances masking agents.

Pentanal (valeraldehyde) is common in natural and synthetic fragrances too, but concentrations of pentanal were lower for the examined cars and buses compare to those of hexanal.

Benzaldehyde, due to almond like smelt, is used in cosmetic and fragrance industry, in spice and dies production and as a solvent. It may be released from many interior materials in the vehicle cabins, but in the examined vehicles concentrations of benzaldehyde (phenylomethanal) were slightly above the detection limit.

Concentrations of propenal (acrolein), a known strong eye irritant was found within the range 0.9÷10.3 μg/m^3. Acrolein is another aldehyde, ubiquitous in the environment, produced by the incomplete combustion of organic materials as well as the oxidation of other atmospheric pollutants such as 1,3-butadiene, a component of motor vehicle exhaust. Less is known about indoor air sources, and all of suspected actions are connected with smoking and cooking activities [10]. In the automobiles it may originate from engine exhaust or tobacco smoke. Acrolein is not listed in the ordinance of the Polish Ministry of Health. NIOSH has set the exposure limit of acrolein to REL = 250 μg/m^3 [22]. Concentrations of acrolein were lower then REL in all the examined vehicles.

Pang and Mu [25] found methanal (formaldehyde), ethanal (acetaldehyde), propanal (propionaldehyde) and phenylomethanal (benzaldehyde) to be dominant in the cars, they examined in China. Comparison of the results from this study with findings for formaldehyde and acetaldehyde of other authors is presented in Table 2. Literature data for carbonyl compounds in cars and busses are scare, but measurements for limited number of aldehydes made in Korea [15] and China [24] showed higher mean values for formaldehyde then in Lublin, Poland.

Table 2. Concentration of formaldehyde and acetaldehyde (μg/m^3) in cars – literature data

Tabela 2. Stężenia formaldehydu i aldehydu octowego (μg/m^3) w pojazdach – dane literaturowe

Place /author	vehicle	formaldehyde		acetaldehyde	
		mean	range	mean	range
Korea [15]	car	20.0	8.8÷39.8	8.9	1.4÷19.1
	bus	21.2	8.7÷28.8	9.1	2.5÷15.5
China [24]	car	27.0	13.0÷34.0	30.0	18.0÷84.0
	bus	20.0	13.0÷30.0	18.0	14.0÷26.0
Poland [this study]	bus	6.2	4.9÷8.7	6.7	4.2÷10.3
	car	16.4	7.9÷28.3	18.5	0.8÷44.1

The detected levels in bus driver cabins were far below the recommendations of NIOSH REL and OSHA exposure limits. Concentrations of most of the detected carbonyl compounds in the private cars were higher, but below the limits too, with one exception. For one car (four years old) concentrations of formaldehyde were at 20 μg/m^3 (at NIOSH REL limit), concentration of acetaldehyde exceeded 40 μg/m^3, for butanal exceeded 50 μg/m^3, for hexanal exceeded 30 μg/m^3. Based on owner interview and car inspection, environmental tobacco smoke (ETS) and intensive use of air fresheners might be the main source of the indoor pollutants in this particular car.

4. Conclusions

Both in the case of cars and buses, the highest concentrations among volatile organic compounds were found for ethanal (acetaldehyde) and butanal. Formaldehyde and acrolein, compounds accused of Sick Building Syndrome, as well as hexanal, were ubiquitous in all the examined vehicles.

In the case of ethanal (acetaldehyde) and butanal, this is caused by the fabric upholstery and the dyes used in manufacturing. High concentrations of the remaining compounds were caused by the applied air fresheners, perfumes and fragrant cleaning agents. Environmental tobacco smoke has an impact on the levels of formaldehyde and acrolein. The detected levels in bus driver cabins were far below the recommendations of NIOSH REL and OSHA exposure limits. Concentrations of most of the detected carbonyl compounds in the private cars were higher, but below the limits too, with the exception of one car.

Substances causing eye irritation are ubiquitous in the indoor spaces, including private and public vehicles and can cause lack of concentration and reduced work efficiency of occupants.

The quality of indoor air is of growing importance now-a-days, as time spent by man in different indoors environment grows rapidly. In the future it will become more relevant to consider cars as workplaces, and apply exposure limits for drivers, especially as the impact of formaldehyde on memory skills has recently been confirmed by Lu et al. [19].

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Lotne związki organiczne w prywatnych i publicznych środkach transportu

Streszczenie

Od końca XX wieku w krajach rozwiniętych, jakość powietrza wewnętrznego jest ważniejsza dla jakości naszego życia i zdrowia niż jakość powietrza zewnętrznego. Wynika to przede wszystkim ze zmian w stylu życia – człowiek coraz więcej czasu spędza w pomieszczeniach. Są to nie tylko pomieszczenia mieszkalne i miejsca pracy, ale także wypoczynku (kluby, restauracje, centra gimnastyczne i centra handlowe) oraz środki transportu.

Jakość powietrza w mieszkaniach, biurach i pomieszczeniach edukacyjnych od kilkunastu lat jest przedmiotem badań naukowców z wielu krajów. Mniej uwagi przywiązywano do powietrza w środkach transportu, mimo że coraz więcej ludzi spędza nawet ponad godzinę dziennie w drodze do pracy i z pracy, a coraz więcej zawodów jest związanych z przebywaniem w środkach transportu (przedstawiciele handlowi, kierowcy zawodowi, policjanci). W kabinach samochodowych zidentyfikowano ponad 160 substancji, jednak większość badań ograniczała się do BTEX, które mogą pochodzić ze spalin samochodowych.

Do zanieczyszczeń, które są oskarżane o syndrom chorego budynku i mogą mieć wpływ na efektywność i wydajność pracy, a co za tym idzie bezpieczeństwo na drodze, należą związki z grupy aldehydów o małych masach cząsteczkowych. Mają one przede wszystkim działanie drażniące błony śluzowe. W pomieszczeniach pochodzą one z materiałów wyposażenia wnętrz i chemii domowej, ale tkaniny i wykładziny oraz odświeżacze powietrza spotykamy też w samochodach.

Aby ocenić jakość powietrza w środkach transportu, wykonano pomiary związków karbonylowych w 10 samochodach osobowych i 10 autobusach, poruszających się po Lublinie, mieście we wschodniej Polsce. Badane samochody osobowe różniły się rokiem produkcji, przebiegiem, marką i producentem oraz typem klimatyzacji. Badane autobusy pochodziły z tej samej partii, zakupionej

w 2009 roku, miały zatem podobne wyposażenie i przebieg. Poboru próbek dokonano metodą dozymetrii pasywnej, a analizy jakościowej i ilościowej metodą HPLC.

Zidentyfikowano 12 związków karbonylowych, a 8 z nich: metanal (formaldehyd), etanal (acetaldehyd), propenal (akroleina), propanal (aldehyd propionowy), butanal, pentanal (aldehyd walerianowy), heksanal i fenylometanal (benzaldehyd), występowało we wszystkich badanych samochodach i autobusach powyżej poziomu detekcji.

Stężenia wszystkich badanych związków w autobusach były niższe niż w samochodach osobowych i nie przekraczały stężeń zalecanych przez NIOSH i OSHA. Stężenia w samochodach prywatnych były także niższe niż zalecane maksymalne, z wyjątkiem jednego samochodu, który charakteryzował się najwyższymi stężeniami prawie wszystkich mierzonych substancji. Zarówno dla autobusów, jak i samochodów osobowych, najwyższe stężenia zmierzono dla etanalu i butanalu. Stosunkowo wysokie stężenia wykazywał także metanal (formaldehyd) i propenal (akroleina) – czyli substancje oskarżane o powodowanie syndromu chorego budynku. Etanal i butanal mogły pochodzić z tapicerki oraz barwników, ponieważ wyższe stężenia zmierzono w samochodach z tapicerką welurową niż skórzaną. Natomiast formaldehyd i akroleina są produktami niepełnego spalania i ich wyższe stężenia odnotowano w samochodach palaczy. Jakkolwiek w kabinach autobusów palenie jest zabronione, i nikt z palących właścicieli samochodów prywatnych nie przyznawał się do palenia w samochodzie, to środowiskowy dym tytoniowy powoduje podwyższenie stężeń tych substancji. Źródłem heksanalu natomiast były prawdopodobnie odświeżacze powietrza i perfumy użytkowniczek pojazdów.

Substancje drażniące błony śluzowe są powszechnie obecne w pojazdach i mogą powodować obniżenie koncentracji, dlatego wydaje się uzasadnione objęcie badaniami monitoringowymi jakość powietrza w środkach transportu.