RESEARCH ARTICLE

The influence of meteorological conditions and anthropogenic activities on the seasonal fluctuations of *BTEX* in the urban air of the Hanseatic city of Gdansk, Poland

Mariusz Marć[1] · Michalina Bielawska[2] · Waldemar Wardencki[1] · Jacek Namieśnik[1] · Bożena Zabiegała[1]

Abstract The results of studies conducted in Gdansk in the period from January to December 2013 and focused on the determination of *BTEX* in the atmospheric air are presented. At the stage of the isolation and/or enrichment of analytes from the gaseous medium, a passive sampling technique—*Radiello*® diffusive passive samplers, was applied. The time-weighted average annual concentration of benzene, toluene, ethylbenzene, and total xylenes determined in the monitored area was as follows: 0.66 ± 0.32, 1.63 ± 0.94, 0.67 ± 0.61, and 2.9 ± 2.7 $\mu g/m^3$. As a result of the research, the potential emission sources of *BTEX*, which activity has a significant impact on the concentration of these compounds in the atmosphere in the Gdansk area, were identified. A comparison of the accuracy and precision of the results of benzene concentrations in the atmosphere obtained by the *BTEX* automatic analyzer and passive techniques was performed. Moreover, a significant influence of meteorological conditions, such as air temperature, intensity of solar radiation, velocity and direction of wind, humidity, and rainfall on the benzene content in ambient air was shown. Additionally, in order to determine the conditions in the area covered by the monitoring, information about atmospheric stability and the height of the mixing layer was presented.

✉ Bożena Zabiegała
bozena.zabiegala@pg.gda.pl

1 Department of Analytical Chemistry, Gdańsk University of Technology, ul. Narutowicza 11/12, 80-233 Gdańsk, Poland

2 Agency of Regional Air Quality Monitoring in the Gdansk Metropolitan Area (ARMAAG), ul. Brzozowa 15A, 80-245 Gdańsk, Poland

Introduction

According to the guidelines proposed by experts from the World Health Organization (WHO) and the European Commission (EC), the term "volatile organic compounds" refers to those organic compounds whose boiling points, measured at a pressure of 1,013 hPa, oscillate in the range of 50–100 to 240–260 °C (WHO 1980; Directive 2004/42/EC). In urban areas, the presence of chemical compounds from the volatile organic compounds (VOCs) group (in the original form) in the atmosphere can significantly affect human health, causing respiratory diseases, headaches, and eye irritation (Tong et al. 2013; Kountouriotis et al. 2014). In addition, under appropriate climatic and meteorological conditions in urban areas, VOCs are substrates/precursors (including nitrogen oxides) in the reactions leading to the formation of tropospheric ozone. According to information included in the Directive of the European Parliament and Council 2008/50/EC as precursors of tropospheric ozone formation in ambient air of urban areas, the following 31 organic compounds from VOCs group were considered: 1,3-butadiene, formaldehyde, compounds from the trimethylbenzenes group, and organic compounds known as BTEX (benzene, toluene, ethylbenzene, *m*, *p*-xylene, *o*-xylene). The *BTEX* organic compounds group is defined as indicators to obtain information about the degree of exposure of the human body to the harmful effects of the

VOCs (Ly-Verdú et al. 2010). Special attention in the monitoring research of air quality is paid to the level of benzene content in urban areas. According to European legislation, annual average benzene concentration in the ambient air of urban areas should not exceed 5 μg/m^3 (Directive 2000/69/EC). Long-term human exposure to benzene present in ambient air can cause asthma, acute myeloid leukemia, and liver, kidney, respiratory system, and central nervous system damage (Zhang et al. 2012; Ramírez et al. 2012; Kim et al. 2013; Simpson et al. 2013; Cakmak et al. 2014).

The main sources of emissions of the *BTEX* in urban areas include the following: combustion of liquid fuels in motor vehicle engines (traffic); the burning of fossil fuels; the chemical, coke oven, metallurgical and refinery industries; fuel stations (liquid fuel spills or leaks from distributor tanks) and asphalt concrete factories (Alyuz and Alp 2014; Dumanoglu et al. 2014; Hien et al. 2014; Słomińska et al. 2014). Furthermore, the weather conditions such as wind velocity and direction, temperature, humidity, type and intensity of precipitation, the extent and duration of the temperature inversion, and the thickness/height of the boundary layer can significantly affect concentration of the *BTEX* in the ambient air in urban areas (Yılmaz Civan et al. 2012; Zheng et al. 2013; Kountouriotis et al. 2014; Zhang et al. 2014).

In the article, the results of studies conducted from January to December 2013, focused on the determination of *BTEX* in the atmospheric air in Gdansk, are presented. At the stage of the isolation and/or enrichment of analytes from the gaseous medium, the passive sampling technique—*Radiello®* diffusive passive samplers was applied. Moreover, using *t-Student* and *F-Snedecor* statistical tests, the attempt was made to compare the techniques of the passive sampling of analytes with the automatic on-line *BTEX* analyzer in terms of accuracy and precision. The concentration levels of benzene measured in the ambient air in Gdansk in 2011–2013 were adopted as the comparison criterion.

An analysis of the impact of weather condition variability (temperature, solar radiation, velocity and direction of wind, humidity, and precipitation) on the seasonal variation of the benzene concentration in ambient air in the monitoring area was conducted in the next part of the research. Primary meteorological data were supplemented by information from *The Weather Research and Forecasting* (*WRF*) *Model* regarding the atmospheric stability and the mixing layer height (www.wrf-model.org). Synoptic conditions are based on the synoptic maps of the atmospheric pressure distribution (www.wetterzentrale.de). According to information listed in the literature, the *WRF Model* is the mesoscale meteorological model designed to simulate and forecast atmospheric circulation based on both actual and theoretical data. This model can be used for different spatial scales, from a few to several hundred kilometers (Zepka et al. 2014).

The *WRF Model* was developed by several institutions, such as the *National Centre for Atmospheric Research* (NCAR), the *National Centres for Environmental Prediction* (NCEP), the *National Oceanic and Atmospheric Administration* (NOAA), the *Forecast Systems Laboratory* (FSL), and the *Air Force Weather Agency* (AFWA) (Diagne et al. 2014; Hariprasad et al. 2014; Shrivastava et al. 2015).

As input data for the model, information extracted from a shared *NCEP/NCAR Reanalysis* project can be used, which includes all the measurement data from the ground-based network, aerological and precipitation measurements, as well as data from surveys and satellite observations (www.esrl.noaa.gov).

As additional information, the paper presents the results of seasonal fluctuations in the concentration of toluene, ethylbenzene, and total xylenes determined in the atmosphere in Gdansk from January to December 2013. In order to identify the main sources of *BTEX* emissions in the monitoring area, numerical values of *BTEX* inter-species ratio—*toluene/benzene* (T/B); m, p-Xylene/Benzene (m, p-X/B); o-Xylene/Benzene (o-X/B); m, p-xylene/ethylbenzene (m, p-X/E) were determined.

Materials and methods

Description of sampling area and location of monitoring/sampling stations

Research aimed at the determination of the *BTEX* in the atmospheric air was carried out on the territory of one of the largest Polish cities—Gdansk. Gdansk is located in the northern part of Poland, on the Gdansk Bay. The city covers an area of approx. 262 km^2 and is inhabited by approx. 461,000 residents. The Gdansk area is one of the largest and fastest growing areas in Poland in terms of industry, transport, and urban development. The main industries whose centers are located in Gdansk include the shipbuilding industry (*Gdansk Ship Repair Yard*), the petrochemical industry (*LOTOS Group*), the energy industry (*EC Wybrzeże*), and the clothing, metal, and construction industries (Król et al. 2012; Marć et al. 2014a). Approximately 70,000 economic entities were registered in Gdansk in 2013 (www.gdansk.pl).

In the case of road infrastructure, the main communication junctions include streets such as *Marynarki Polskiej Street*, *Grunwaldzka Street*, *Gen. J. Hallera Street*, and *Armii Krajowej Street* (Marć et al. 2014c). The numerical value of the vehicle use rate, the number of registered cars in the city per 1,000 inhabitants, amounted to approx. 523.5 in the area of Gdansk in 2013 (www.gdansk.pl).

In order to obtain reliable and credible analytical information about the current state of the atmosphere and air quality in the monitoring area, it is very important to properly select the location of measurement points. The following logistical, urban, and weather conditions may have a significant impact on

the distribution of the monitoring area measuring/monitoring stations: suitable distance of the measuring/monitoring station from individual or continuous intensive sources of emissions; regular exchange of ambient air around the area of the measuring/monitoring site occurring as a result of air mass movement in different directions; location of water bodies with a large surface area, i.e., lakes, seas, oceans; location of green areas with a large surface area, i.e., meadows, fields, forests; population density in the area around the measuring/monitoring site; meteorological conditions in the area around the measuring/monitoring site; the role of the measuring/monitoring site: the measurement of the urban background, the measurement of hot spots along the main traffic routes.

Taking into account the above criteria, five measuring/monitoring stations were located in the area of Gdansk, which the *Agency of the Regional Air Quality Monitoring Foundation in the Gdansk Metropolitan Area* (*ARMAAG Foundation*), the user and administrator, founded in 1993. In each of the stations are installed stationary analyzers and monitors operating in *on-line* mode, which provide information about ambient air quality and meteorological conditions in the monitoring area (www.armaag.gda.pl). In Fig. 1, the locations of measuring/monitoring stations in the Gdansk area are shown schematically. Detailed information about the location of measuring/monitoring stations and meteorological conditions in the monitoring area is presented in previous literature reports (Zabiegała et al. 2010; Marć et al. 2014a).

BTEX sampling techniques

Sampling of *BTEX* analytes from the atmospheric air from the area around the measuring/monitoring stations located in the city of Gdansk was done using passive sampling technique. As a tool for sampling analytes from the gaseous medium, Radiello® diffusive passive samplers (*Fondazione Salvatore Maugeri, Padova, Italy*) were applied, which consist of three main components: a plastic tripod allowing the installation of a passive sampler, a microporous polyethylene diffusion membrane, and a cylindrical steel net filled with graphitized charcoal *Carbograph 4* functioning as a sorption bed. Detailed information about the construction and metrological characteristics of Radiello® diffusive passive samplers is presented in previous literature data (Król et al. 2012; Krupińska et al. 2013; Marć et al 2014b).

On each of the five measuring/monitoring stations located in the monitoring area, Radiello® diffusive passive samplers were placed at a height of approx. 3 m from the ground surface in steel cages specially constructed for this purpose. The purpose of these frames was to protect the samplers from the adverse effects of the weather (rain, snow, hail, heavy gusts of wind, and intense sunlight) and to ensure proper air circulation around the sampler diffusion zone (reducing the phenomenon of passive sampler "starvation"). Inside the cages were installed two passive samplers. In order to avoid the phenomenon of competitiveness, passive samplers were
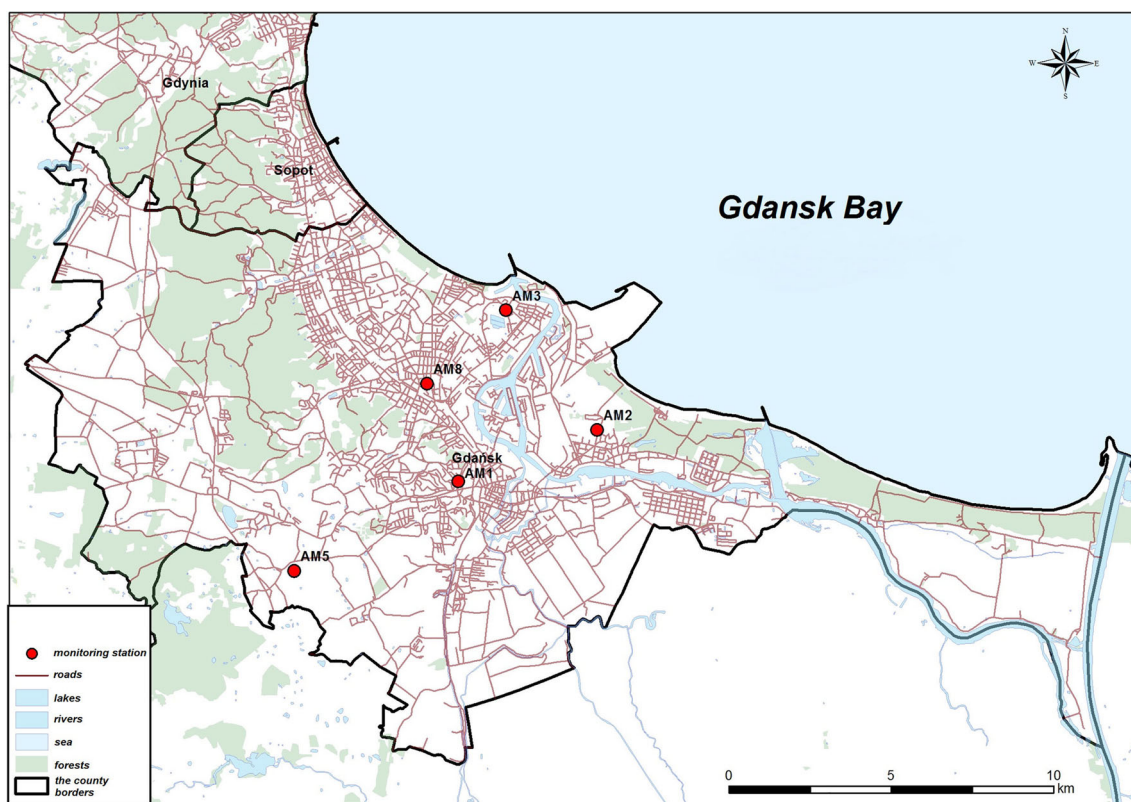

Fig. 1 Location of monitoring/measurement stations in Gdansk

spaced at least 20 cm from each other. Figure 2 shows schematically how and where Radiello® diffusive passive samplers were installed at the measuring/monitoring sites. The exposure time of the Radiello® diffusive passive samplers installed on each of the five measuring/monitoring stations was 14 days. After the predetermined exposure time, cylindrical steel nets filled with the sorption bed were removed from the interior of the passive sampler and transferred to glass containers sealed with a polyethylene caps. The containers were then transported to the laboratory in order to perform the liberation, separation, identification, and quantification of analytes retained on the sorption bed. During the measurement period, which lasted from January to December 2013, 240 analyte samples in total were collected from the gaseous medium from all five 5 measuring/monitoring stations (48 samples from each of the measuring/monitoring station).

The application of the passive sampling technique in monitoring research of ambient air quality allows the obtaining of analytical information defined as the time-weighted average concentration (TWA) of the analyte in the gaseous medium. In the case of applying Radiello® diffusive passive samplers in the research, the average time-weighted concentration of BTEX analytes present in the atmosphere is determined based on the following equation (www.radeillo.com) (1):

$$C = \frac{m}{Q_{298} \cdot \left(\frac{T}{298}\right)^{1.5} \cdot t} \cdot 10^6 \qquad (1)$$

where:

m	mass of the analyte retained on a sorption bed [μg]
t	exposure time [min]
C	time-weighted average concentration of the compound determined in the ambient air [μg/m^3]
Q_{298}	uptake/sampling rate of a particular analyte from the BTEX group at a temperature of 298 K and a pressure of 1,013 hPa [ml/min]
T	temperature [K]

The Q_{298} coefficient defining the sampling rate of the BTEX analytes from a gaseous medium by Radiello®

diffusive passive samplers at 298 K and pressure of 1, 013 hPa were determined by the manufacturer (*Fondazione Salvatore Maugeri*, *Padova*, *Italy*), and were respectively: $Q_{289,\text{Benzene}}$ 26.8 ml/min, $Q_{289,\text{Toluene}}$ 30.0 ml/min, $Q_{289,\text{Ethylbenzene}}$ 25.7 ml/min, $Q_{289,m\text{-Xylene}}$ 26.6 ml/min, $Q_{289,p\text{-Xylene}}$ 26.6 ml/min, $Q_{289,o\text{-Xylene}}$ 24.6 ml/min.

The on-line determination of BTEX present in the atmospheric air over the Gdansk area was performed by means of the automatic BTEX on-line analyzer—GC955-600 (*Synspec b.v.*), one of the standard pieces of equipment of the *ARMAAG Foundation* measuring/monitoring station. This type of analyzer contains the following main elements: pump, sample system—pre-concentration with *Tenax GR* 35-60 mesh, oven with a temperature range from 50 to 100 °C, analytical capillary column *SY5* (13 m×0.53 mm×1.2 μm), stripper capillary column *AT5* (2 m×0.32 mm×1 μm), and photoionization detector (PID) operating at 10.6 eV. This type of analyzer requires high-purity nitrogen (99.999 % pure) as a carrier gas. The *GC955-600* on-line analyzer allows the determining of the concentration levels of BTEX present in the atmospheric air at 1-h intervals.

Instrumental analysis—extraction/liberation and final determination of BTEX

The BTEX were desorbed from cylindrical tubes filled with sorption medium using a two-stage thermal desorption technique—a thermal desorber designed, constructed, and made by *Omnisfera*, Gdansk, Poland. Each of the cylindrical tubes was heated up to 280 °C for 20 min. At this stage, the flow rate of high-purity helium was set to 50 ml/min. Liberated analytes were transported and focused into a microtrap (37 mg of *Tenax TA* and 27 mg of *Carbotrap*), which was cooled to 5 °C using a *Peltier* cooler (thermoelectric cooling). Next, the microtrap was rapidly heated (ballistic) to 280 °C for 5 min. Analytes liberated from the microtrap were transported by high-purity helium (flow rate 2.2 ml/min) into the chromatographic column. More information about the two-stage thermal desorption, technique can be found in previous

Fig. 2 General view of the monitoring/measurement stations, together with the installation of Radiello® diffusive passive samplers
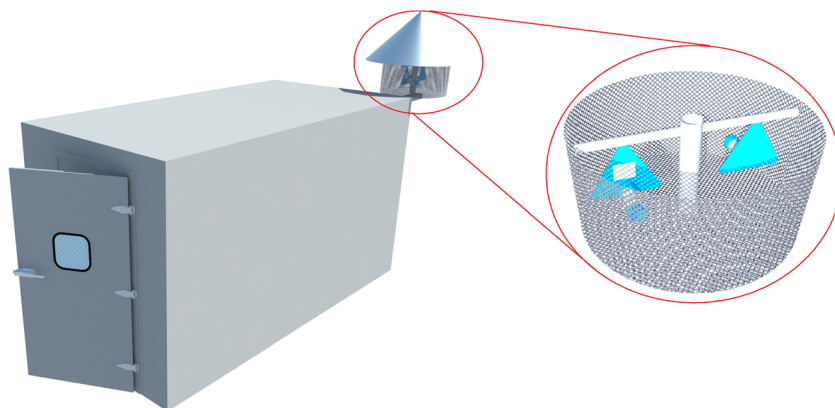
literature data (Zabiegała et al. 2007; Król et al. 2012; Marć et al. 2014b).

A *Hewlett-Packard 5890 GC Series II* gas chromatograph coupled with a flame ionization detector (FID) was applied at the stage of separation, identification, and final determination of the *BTEX* released from the cylindrical tubes filled with sorption medium. The *DB-1* (*J&W*) capillary column (30 m×0.32 mm×5 μm) was used. The high-purity helium was applied as the carrier gas, with a constant flow rate of 2.2 ml/min. The *GC* oven temperature program was initially set to 40 °C for 1 min, then raised at a rate of 15 °C/min up to 120 °C, then increased at a rate of 10 °C/min up to 220 °C, and finally maintained for 5 min at 220 °C. The temperature of the flame ionization detector was set to 250 °C, and the transfer line temperature was set to 150 °C.

Chemical standards and calibration of *TD-GC-MS* system

The quantitative determinations of *BTEX* adsorbed on the sorption medium *Carbograph 4* were performed using the external standard (*ESTD*) method. External calibration was carried out using a mixture of 13 volatile organic compounds (including *BTEX*) at the concentration level 2,000 μg/ml per component in methyl alcohol (*EPA VOC Mix 2*, *Supelco*, *USA*).

Two five-point calibration curves for *BTEX* were developed for two main ranges of analytes mass introduced and retained on the Radiello® diffusive passive sampler sorption medium *Carbograph 4*: first a range of calibration curves was created from 50 to 400 ng per sorption medium (the coefficient of determination (R^2) for the calibration curves of *BTEX* was from 0.995 to 0.999), and the second range of calibration curves was created from 400 to 2,000 ng per sorption medium (the coefficient of determination (R^2) for the calibration curves of *BTEX* was from 0.994 to 0.996). Further detailed information about the procedure of preparing *BTEX* chemical standards and extensive information about the entire calibration procedure can be found elsewhere (Zabiegała et al. 2010; Król et al. 2012; Marć et al. 2014a, b, c).

Quality assurance and quality control of *BTEX* measurements

Quality assurance (QA)/quality control (QC) control procedures consisted of the analysis of field blanks and parallel duplicate samples. For this purpose, on one of the monitoring stations located in the monitoring area, some of the Radiello® diffusive passive samplers were left unexposed during the exposure time (14 days). Then, field blank samples were analyzed using the same analytical protocol as in the case of passive samplers exposed to atmospheric air. The calculated amounts of the *BTEX* were corrected, taking the blank test into account. Furthermore, the effectiveness of analyte liberation

from the sorption medium, *Carbograph 4*, was studied. The recoveries of *BTEX* after extraction at high temperature (two-stage thermal desorption) were in the range of 97–103 %.

The uncertainty (2σ) of the sampling rates (Q) estimated by the manufacturer of the Radiello® diffusive passive sampler were from 7.5 % for benzene to 11.3 % for *p*-xylene. The expanded uncertainty of the determinations of *BTEX* in atmospheric air by the use of the *GC955-600 on-lin*e analyzer was estimated by *ARMAAG Foundation* staff at 25 %.

The method quantitation limit (MQL) for the analytical protocol of *BTEX* determination in the atmospheric air over the Gdansk area, using the Radiello® diffusive passive sampler, were estimated in the range of 0.02–0.05 μg/m³.

Results and discussion

Seasonal fluctuations of benzene concentration level measured in the atmospheric air in the monitoring area in 2013

According to information published by experts from the *International Agency for Research on Cancer*, benzene is classified into Group I of chemical compounds with carcinogenic properties. According to the literature, prolonged exposure of the human body to benzene can affect significantly the number of people diagnosed with cancer, e.g., leukemia (I.A.R.C. 1982). Due to the adverse effects of benzene on the human body, European law clearly defines the maximum concentration of the compound in the atmosphere in the monitoring area. According to the information presented in Directive 2000/69/EC of the European Parliament and of the Council of Europe, the average benzene concentration in the atmosphere in the monitoring area (values allowed for the protection of human health) must not exceed 5 μg/m³.

As a result of the monitoring study conducted in 2013, it was concluded that the limit value of annual average benzene concentration in the atmosphere was not exceeded at any of the monitoring/measuring stations located in Gdansk. The average annual time-weighted benzene concentration determined in the atmosphere in the areas located around the monitoring/measuring stations were (result presented as TWA±standard deviation) as follows: AM1—0.7±0.4 μg/m³; AM2—0.5±0.2 μg/m³; AM3—0.7±0.4 μg/m³; AM5—0.6±0.4 μg/m³; AM8—0.8±0.5 μg/m³, respectively. The average annual time-weighted benzene concentration determined in the atmosphere in Gdansk was 0.7±0.4 μg/m³ in 2013.

According to information published in the scientific literature, there is a clear relationship between the concentration of benzene in ambient air in urban areas, the ambient air temperature, and solar radiation intensity (Zabiegała et al. 2010; Król et al. 2012; Marć et al. 2014a, b, c). Figure 3 presents a

Fig. 3 The average monthly time-weighted benzene concentration in the atmosphere determined in Gdansk area in 2013: a in relation to ambient air temperature; b in relation to the intensity of solar radiation
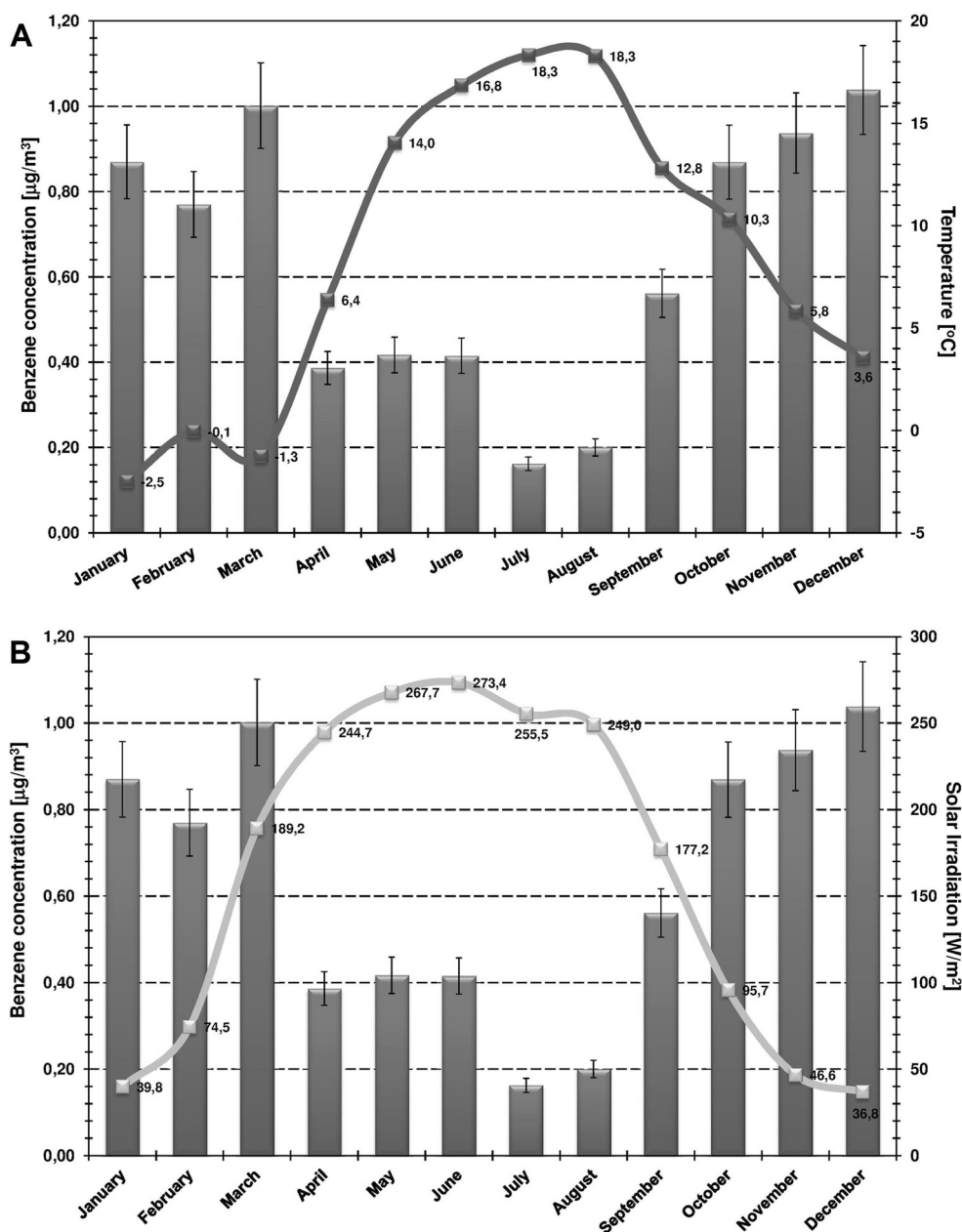
graphical representation of data illustrating the values of average monthly time-weighted benzene concentrations determined in ambient air in Gdansk in 2013.

By analyzing the information presented in Fig. 3a, it can be noted that there is a clear relationship between the average monthly time-weighted benzene concentration determined in ambient air and the temperature of the ambient air. In the monitoring area during the heating season, covering the period from 1 October to 30 April, the average concentration of benzene determined in ambient air was 0.8 ± 0.2 $\mu g/m^3$ (average temp. in this period: 3.2 °C), while in the summer (the period from May 1 to September 30), the average concentration of benzene in ambient air was 0.4 ± 0.2 $\mu g/m^3$ (average temp. in

this period: 16.1 °C). The presence of a clear inverse relationship between the temperature of the ambient air and the average monthly time-weighted benzene concentration determined in the atmosphere in the urban monitoring area depend on the seasonal activity of the three major sources of emissions:

1. The combustion of liquid fuels in motor vehicle engines—low ambient air temperature in the autumn and winter increases the density of the oil contained in combustion engines, causing the time for reaching the optimum fuel combustion temperature (total combustion) longer. The occurrence of this type of phenomenon increases

fuel consumption (liquid fuel combustion is incomplete), which in turn increases the number of *BTEX* (including benzene) emitted to the ambient air;

2. The combustion of solid fuels used for domestic heating—in the autumn and winter, low atmospheric temperature results in the need for domestic heating. In residential buildings in which heat is not supplied by thermal power stations, low-efficiency individual heating systems are used. In such systems, the main heating fuel is low-quality coal. The drop in ambient air temperature in urban areas increases the intensity of the use of low-efficiency individual heating systems in residential areas, thereby increasing the amount of compounds from the *BTEX* group emitted into the air;

3. The seasonal use of recreational and residential gardens—in the summer season, covering the period from May to September, increased human activity in gardens and recreation areas, resulting from the use of these areas for leisure and recreation. In the case of the autumn period (October and November), human activity in residential gardens and recreation areas is due to cleaning work, which is often accompanied by biomass or plastics burning. Moreover, in exceptional cases, in the winter, small-sized buildings located in residential gardens and recreation areas can be operated by users and heated by low capacity furnaces fueled with different kinds of (often inappropriate) materials. Any kind of human activity resulting from the seasonal use of recreational residential gardens can contribute to an increase in emissions of the *BTEX* emitted into the air in urban areas.

Furthermore, the activity of industrial centers located in Gdansk may have an impact on the ambient air quality, and thereby also on the levels of *BTEX*. However, in most cases, the activity of industrial centers (excluding power plants) is not significantly dependent on the ambient air temperature.

Another meteorological factor influencing the benzene concentration in the atmosphere in the Gdansk area is the intensity of solar radiation (Fig. 3b). In the spring and summer, including the months from April to September, the intensity of solar radiation is maintained at a high level (the average radiation intensity in this period was approx. 245 W/m^2), which in turn causes an increase in the intensity of photocatalytic reactions occurring in the atmospheric air. The presence in the atmosphere of compounds classified as oxidizing agents, i.e., tropospheric ozone and hydroxyl radicals, results in a significant reduction in the amount of benzene in ambient air in urban areas (Alghamdi et al. 2014) (the rate constant for reaction of hydroxyl radicals with benzene—$k_{OH}=1.21 \times 10^{-12}$ cm^3 molecules^{-1} s^{-1}) (Monod et al. 2001). In the case of the autumn-winter period, covering the months from October to March, a significant decrease in the intensity of solar radiation was reported—the average radiation intensity

in this period was only approx. 80 W/m^2. The lower intensity of solar radiation in autumn-winter causes photocatalytic reactions to occur with much less intensity, due to the smaller, and in some cases negligible, concentration of strong oxidants (O$_3$ or OH) in the atmosphere. The occurrence of this type of phenomenon causes the residence time of benzene in ambient air in urban to be extended (average atmospheric lifetime is 9.4 days based on the reaction with the OH) (Monod et al. 2001), which in turn may contribute to the increased migration/transportation of benzene from/or to monitoring area.

Referring to the information presented in Fig. 3, it can be noted that there is an interesting relationship affecting the amount of benzene in the atmospheric air. In April and November, the ambient air temperature in the monitoring area was at a similar level, and amounted to 6.4 and 5.8 °C, respectively. However, the average monthly time-weighted benzene concentration in the atmosphere determined in Gdansk during these months was significantly different, and amounted to 0.39±0.06 and 0.9±0.3 μg/m^3. The occurrence of this type of phenomenon is caused by the differences in the intensity of solar radiation recorded in April and November. The average intensity of solar radiation in urban monitoring areas in April was 245 W/m^2, while in November, the value was more than five times smaller, and amounted to approx. 47 W/m^2. Noticing such a relationship confirms the earlier hypothesis that in addition to the ambient air temperature, the solar radiation intensity also has a significant effect on the concentration of benzene in ambient air in urban areas.

Both the ambient air temperature and the intensity of solar radiation have a significant effect on the thickness/height of the boundary layer and the intensity of the mixing of air masses in urban areas. According to information listed in the literature, in the spring and summer, the thickness/height of the boundary layer is much higher than in the autumn and winter. In the spring and summer, the high temperature of the ambient air and the high intensity of solar radiation cause an increase in the intensity of photochemical reactions involving strong oxidants (O$_3$ and OH) and organic compounds present in ambient air (Tao et al. 2007; Choi et al. 2011).

Comparison of the concentration levels of benzene measured using the on-line analyzer and the passive sampling technique in the period 2011–2013

In Fig. 4, a graph showing the results of studies conducted in Gdansk in 2011–2013 is presented, aimed at the determination of benzene in ambient air using the automatic on-line analyzer (*GC955-600*) and passive sampling of analytes from the gaseous medium (Radiello® diffusive passive samplers).

Referring to the information presented in Fig. 4, it can be noticed that the concentrations of benzene in the atmosphere determined using the passive sampling technique are in most

Fig. 4 Average monthly concentrations of benzene in ambient air determined in Gdansk in 2011–2013 using passive sampling technique and automatic on-line *BTEX* analyzer
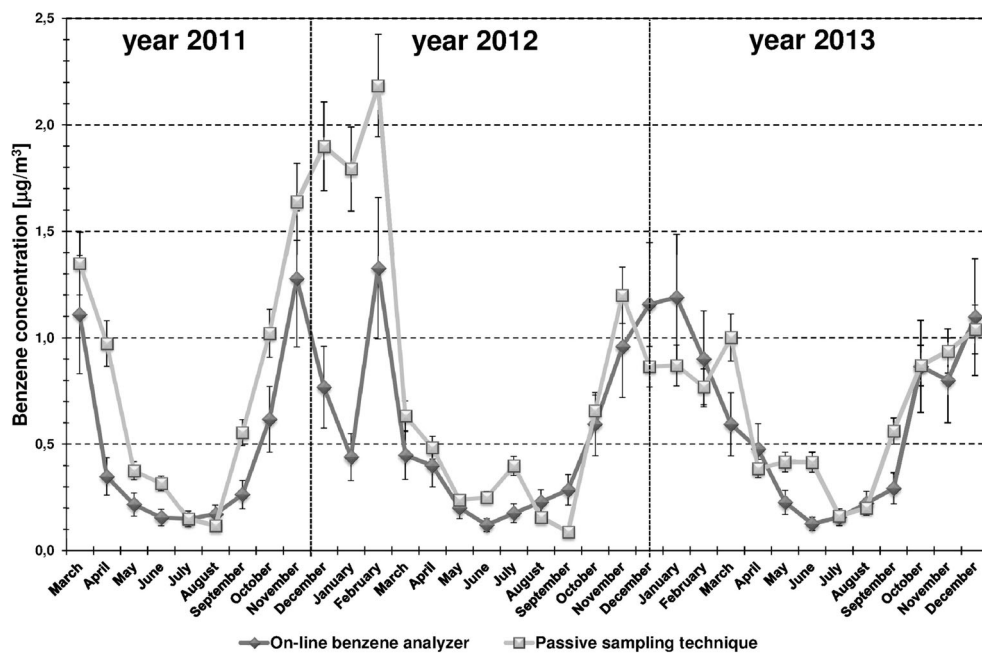
cases higher than the concentrations of benzene determined using a stationary on-line automatic analyzer. A clear distinction in the obtained results can be seen in the measurement period lasting from September 2011 to March 2012. The occurrence of a discrepancy between the obtained results of benzene concentration in the atmosphere may be due to the following phenomena or factors: number of samples and the sampling period of the analytes from the atmosphere. In the case of an automatic on-line analyzer, benzene concentration measurement in the atmosphere is performed at 1-h intervals, and the exposure time of passive samplers (period of the analytes sampling from the atmosphere) during one round of measurement was 14 days (336 h); operating mode of the equipment used (on-line monitor, *off-line* passive sampler); the impact of weather conditions such as temperature, or velocity and direction of wind, which significantly affect the uptake/sampling rate of the passive samplers; time needed to obtain analytical information (benzene concentration) since the end of the sampling (on-line monitor—real time concentration, passive sampler—time-weighted average concentration).

Moreover, as presented in the previous section, it can be noticed that there is a clear difference between the levels of benzene content in the atmosphere determined in the autumn-winter and spring-summer seasons.

A very important phenomenon observed during the analysis of the information shown in Fig. 4 is the fact that from year to year, while conducting the monitoring research, the levels of benzene slowly decreases in the ambient air in Gdansk. This phenomenon may be due the effective information policy about the environmental impact of low-efficiency heating systems, and replacing old car models with newer ones, in which

combustion engines have better efficiency, and in which the combustion of liquid fuels emits lower amounts of pollutants into the air.

In order to compare the standard deviations of the data sets obtained from 2011 to 2013 using automatic on-line analyzer and passive sampling technique, the statistical *F-Snedecor* test was performed. As a result of the *F-Snedecor* statistical test, it was found that for each measurement period, the calculated F_{calc} parameter (2011—2.32; 2012—2.71; 2013—1.45) is less than the F_{crit} parameter (2011—3.18; 2012 and 2013—2.82). This type of information leads to the conclusion that the compared analytical procedures do not differ in a statistically significant way in terms of precision. In order to compare the mean values of the three measuring periods of the previously mentioned data sets obtained using the two different analytical devices/techniques, the *t-Student* statistical test was performed. As a result of the *t-Student* statistical test, it was observed that the determined parameter t_{calc} (2011—1.38; 2012—0.97; 2013—0.57) is less than the t_{crit} parameter (2011—2.10; 2012 and 2013—2.07) for each measurement period. This information allows the thesis that the compared two analytical procedures do not differ statistically significantly in terms of accuracy/correctness.

For this reason, it can be concluded that the passive sampling of analytes from a gaseous medium can provide an alternative solution to expensive automated instruments in the field of monitoring air quality. The presence of a statistically significant relationship between passive techniques and automated analyzers used in research allowing for determining the VOCs in ambient air in urban areas was also shown in previous papers (Gallego et al. 2011; Walgraeve et al. 2011; Król et al. 2012).

Effect of meteorological conditions in the monitoring area on the levels of benzene in ambient air in 2011–2013

In addition to local emission sources of organic compounds into the air in the monitoring area, meteorological conditions are one of the main factors affecting air quality (Czarnecka and Kalbarczyk 2008; Ramsey et al. 2014; Saha et al. 2014). The most important meteorological factors that may affect the type and amount of pollutants in ambient air in urban areas include the following: temperature, velocity and direction of wind, and the extent and duration of temperature inversions. Table 1 summarizes the information about the Pearson correlation coefficients (at a significance level of $p<0.05$) determined between the level of benzene content in the atmosphere and the various meteorological factors recorded in the monitoring area in 2011–2013.

Interpreting the information listed in Table 1, it can be noticed that the clearest correlation occurred between the concentration of benzene in the atmospheric air and wind velocity. The presence of a moderate negative correlation (from −0.59 to −0.25) allows the conclusion that in the monitoring area a decrease in wind velocity increases the concentration of benzene in the atmosphere. It was also noted that depending on the measurement period, the relationship between the concentration of benzene in the atmosphere and wind direction is moderate or low (from −0.39 to −0.012). In the case of the influence of humidity on the content of benzene in the atmosphere, it can be noted that there is a moderate positive correlation, mainly during the heating season (from 0.32 to 0.37). In addition, analyzing the data presented in Table 2, it can be concluded that there is no influence of rainfall on the concentration of benzene in ambient air in the monitoring area (the presence of very low–near zero correlation). As indicated in Table 2, it was noted that all the correlation coefficients between meteorological factors and the concentration of benzene in the atmosphere reach higher values in the heating season (autumn-winter).

Another way to determine the relationship between meteorological factors and the concentration of benzene in the atmosphere is to determine the linear relationship between the factors redefined by the determination coefficient R^2. Table 2 summarizes information on the set of the determination coefficient specified between meteorological factors and the benzene content in the atmospheric air in Gdansk in 2011–2013. Interpreting the data summarized in Table 2, it can be concluded that wind velocity can be in a range from 6 to 35 % responsible for the seasonal fluctuations of benzene in the ambient air in the monitoring area. In the case of atmospheric pressure, wind direction, and atmospheric humidity, their impact on the content of benzene in the ambient air in the monitoring area was smaller, and ranged from 0.30 to 28 %; from 0.70 to 15 %; and from 0.90 to 15 %, respectively.

The detailed analysis of high episodes of benzene helps to find the main factors responsible for its increasing concentration. The study covers the analysis of three selected episodes from 2011 to 2013 described in detail in following subsections. Taking into consideration the meteorological data enables understanding the cause of high benzene concentration appearance. The most important factors which contribute to increasing concentrations of benzene are as follows: anticyclone atmospheric circulation, stable atmosphere conditions, low wind velocity, and the occurrence of inversion layer.

Episode I—the period from 01.03.2011 to 03.03.2011

The information on the variability of the concentration of benzene in the atmosphere observed during episode I (from 01.03.2011 to 03.03.2011) and the area around monitoring/measuring station AM2, together with meteorological conditions is shown in Supplementary Fig. S1. During episode I, it was recorded at the beginning of March 2011, in the area around monitoring/measuring station AM2, that there were adverse weather conditions affecting the increase in the benzene concentration in the atmosphere. The high benzene concentration was caused by the occurrence of anticyclonic circulation, which brought cold weather with a negative temperature and low-velocity wind. For most of the time, stable atmospheric conditions with the inversion layer were reported. All of these factors significantly contributed to the increase in the

Table 1 The Pearson correlation coefficient (at a significance level of $p<0.05$) for a linear relationship between the levels of benzene determined in the atmosphere and various meteorological factors in 2011–2013

The Pearson coefficient	2011			2012			2013		
	Heating season	Summer season	Year	Heating season	Summer season	Year	Heating season	Summer season	Year
pressure	0.44	0.28	0.41	0.33	−0.22	0.27	0.061	−0.021	0.011
wind direction	−0.39	−0.32	−0.21	−0.37	−0.19	−0.08	−0.012	−0.022	0.012
wind velocity	−0.59	−0.39	−0.38	−0.53	−0.40	−0.35	−0.58	−0.25	−0.32
humidity	0.35	0.011	0.38	0.32	−0.10	0.36	0.37	−0.011	0.37
precipitation	–	–	–	−0.16	−0.13	−0.17	−0.021	−0.021	−0.031

Table 2 The determination coefficient for the linear relationship between the concentrations of benzene and various meteorological elements for the period 2011–2013

The determination coefficient [%]	2011			2012			2013		
	Heating season	Summer season	Year	Heating season	Summer season	Year	Heating season	Summer season	Year
pressure	19	8.1	17	28	5.1	7.5	0.30	–	–
wind direction	15	10	4.0	14	3.6	0.70	–	–	–
wind velocity	35	15	14	28	16	12	34	6.4	10
humidity	12	–	15	10	0.90	13	14	–	14
precipitation	–	–	–	2.6	1.7	3.0	–	–	–

level of benzene in the ambient air in the area around measuring station AM2 up to 7.0–9.0 μg/m³, especially at night.

Episode II—the period from 05.02.2012 to 07.02.2012

The data on the variability of the concentration of benzene in the atmosphere during episode II (from 05.02.2012 to 07.02.2012) recorded in the area around monitoring/measuring station AM2, along with meteorological conditions are shown in Supplementary Fig. S2. At the beginning of February 2012, during episode II, elevated levels of benzene in the ambient air in the area around the monitoring/measuring station were reported, resulting from the occurrence of adverse meteorological conditions. The high concentration of benzene in the atmosphere was associated with the occurrence of a high pressure circuit, together with the occurrence of freezing temperatures and little wind velocity (low air circulation). Similarly, as in episode I, table atmospheric conditions were reported most of the time. All of these factors were unfavorable for the dispersion of benzene and contributed to the increase in the levels of this compound in the atmosphere, especially at night.

Episode III—the period from 11.05.2013 to 07.11.2013

The information on the variability of benzene concentration in the atmosphere observed during episode III (from 11.05.2013 to 07.11.2013) in the area around monitoring/measuring station AM2, together with meteorological conditions, is shown in Supplementary Fig. S3. Adverse weather conditions affecting the increase of benzene concentrations were observed at the beginning of November 2013. Cyclonic circulation with a positive temperature and low-velocity wind at night occurred during episode III. The stable atmospheric conditions with inversion layer were reported on the night of November 5–6, which contributed to an increase in the levels of benzene in the atmosphere, especially at night. The air quality improved due to changes in the circulation, bringing higher wind velocity on 6–7 March.

Seasonal fluctuations of toluene, ethylbenzene, and total xylenes concentration level measured in the atmospheric air in the monitoring area in 2013

Data on average monthly variation in the time-weighted concentrations of toluene, ethylbenzene, total xylenes, and the amounts determined in the atmosphere at the individual measuring/monitoring stations located in Gdansk are shown in Fig. 5.

The average annual time-weighted concentration of toluene, ethylbenzene, and total xylenes determined in the atmospheric air in the areas located around each monitoring/measuring station were (result shown as TWA±standard deviation) as follows: AM1—1.9±1.3, 0.5±0.3, and 2.0±1.3 μg/m³; AM2—1.3±0.7, 1.5±1.7, and 6.5±7.6 μg/m³; AM3—1.8±1.0, 0.7±0.5, and 2.8±2.0 μg/m³; AM5—1.1±0.4, 0.3±0.1, and 1.1±0.6 μg/m³; AM8—2.0±1.2, 0.5±0.4, and 2.1±1.8 μg/m³, respectively. The average annual time-weighted concentration of toluene, ethylbenzene, and total xylenes determined in the atmosphere in the area of Gdansk in 2013 were 1.6±0.9, 0.7±0.6, and 2.9±2.7 μg/m³, respectively. The highest time-weighted average concentration of toluene was observed in the ambient air around measuring/monitoring station AM8, while the smallest concentration was observed in the area around measuring/monitoring station AM5. This is due to the fact that one of the main roads— *Gen. J. Haller Street* is located around the measuring/monitoring station, where the average daily traffic is recorded as being from 45,526 to 60,118 vehicles (Marć et al. 2014c). On the other hand, measuring/monitoring station AM5 is located in the least urbanized area, distant from the main traffic routes, and in a place where the heat for residences is supplied from a power plant (no heating by residential heating systems characterized by low efficiency).

By analyzing the information presented in Fig. 5, it can be seen that there is no clear relationship between the average monthly time-weighted concentration of toluene, ethylbenzene, and total xylenes determined in the atmosphere and the season of the year (between the heating season and the summer season). According to literature data, the lack of a

Fig. 5 The average monthly time-weighted concentrations of toluene, ethylbenzene, and total xylenes determined in the atmosphere in the area around each monitoring/measuring station located in Gdansk in 2013
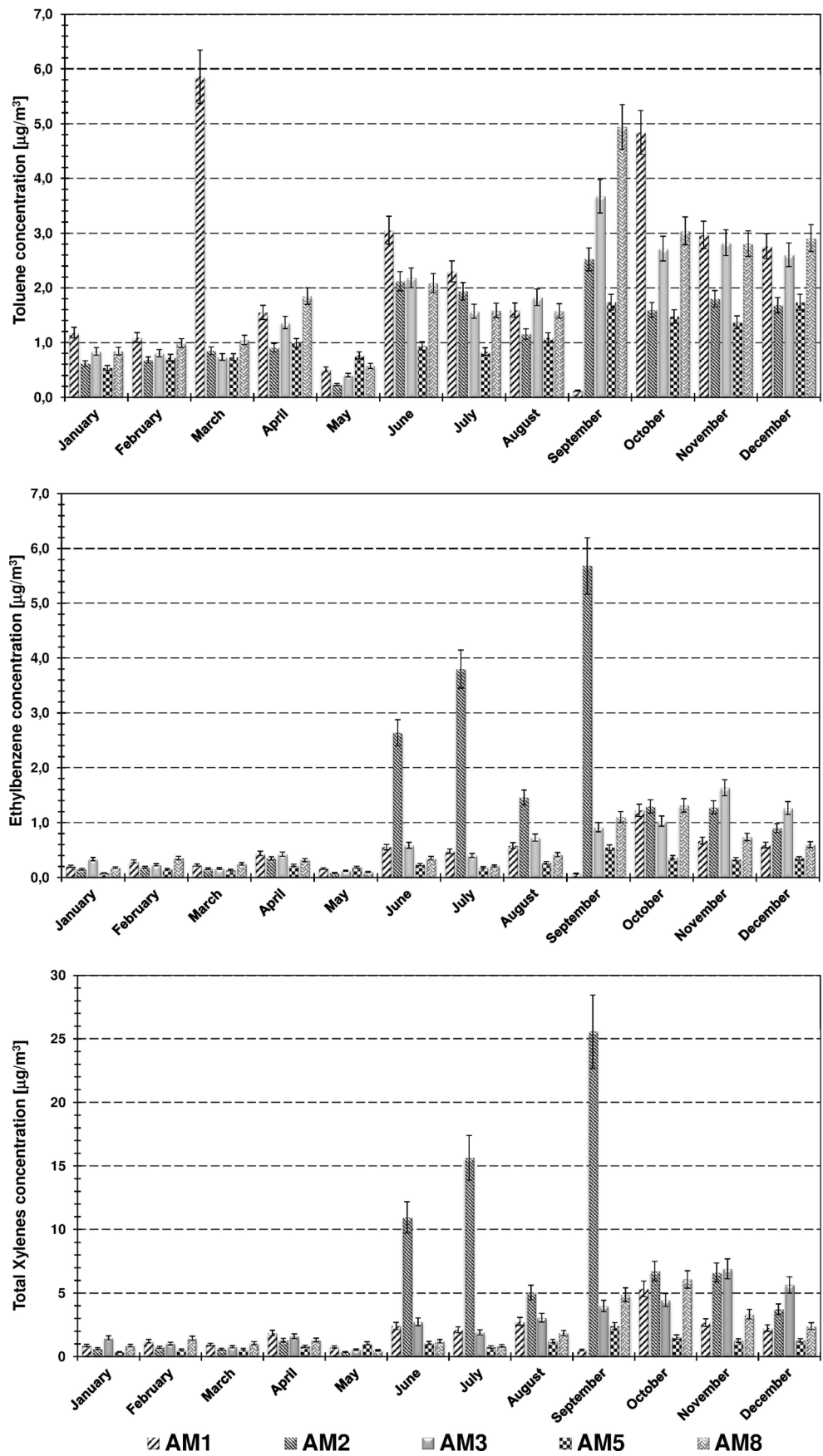
correlation between the time-weighted average concentration of toluene, ethylbenzene, and xylenes, and the meteorological conditions (mainly the temperature of the ambient air) is due to the fact that these compounds are much less stable in the ambient air than the above-mentioned benzene. According to information published by Monod et al. 2001, the shorter residence time of toluene (1.9 days), ethylbenzene (1.6 days), and xylenes (average 17.2 h) in the ambient air compared to benzene (9.4 days) is due to the fact that these compounds undergo chemical reactions with the hydroxyl radicals (OH) present in the atmospheric air much more easily and faster (Monod et al. 2001).

Moreover, it was noted that in areas located around the majority of the measuring/monitoring stations, the average monthly time-weighted concentration of toluene, ethylbenzene, and total xylenes, determined in the atmosphere in the months from June to December, were at a much higher level than from January to May.

Referring to the data presented in Fig. 5, it can be seen that in the atmosphere in the area around measuring/monitoring station AM2, significantly higher levels of ethylbenzene and total xylenes were determined from June to September. A sudden increase in the concentration of ethylbenzene and total xylenes in the ambient air in the area around measuring/monitoring station AM2 may have been caused by intensive construction works in close proximity to the measuring/monitoring station. Near measuring/monitoring station AM2 construction works started, involving road tunneling under the Vistula river, in late May 2013. The road transport of earth excavated from the site ran through the area where measuring/monitoring station AM2 was located. The excavated earth was transported by trucks powered by diesel fuel.

Variations in the inter-species ratios of *BTEX* in 2013

The determination of numerical values of *BTEX* inter-species ratio parameters allows the obtaining of analytical information about the main sources of *BTEX* emissions to the air located in the monitoring area. The numerical values of *BTEX* inter-species ratio parameters largely depend on the climatic conditions of the area, the types of organic compound emission sources into the air, and the age of air parcels (Vardoulakis et al. 2011). The numerical values of *BTEX* inter-species ratio parameters for each of the measuring/monitoring station, and for the entire urbanized area covered by monitoring, are summarized in Table 3.

By analyzing the information summarized in the literature, it can be taken into account that the interpretation includes two main factors: the numerical values of the parameter *T/B* (*toluene/benzene* concentration ratio), and the numerical values of the parameter *m, p-X/E* (*m, p-xylene/ethylbenzene* concentration ratio) (Hsieh et al. 2006; Yurdakul et al. 2013). According to the literature, the parameter *T/B* allows the specification of

Table 3 The numerical values of inter-species BTEX concentration ratios

	T/B	*m, p-X/B*	*o-X/B*	*m, p-X/E*
AM1	2.66	1.78	0.95	2.81
AM2	2.88	5.71	8.95	1.77
AM3	2.62	2.48	1.66	2.61
AM5	1.88	1.18	0.68	2.68
AM8	2.77	1.94	0.99	2.87
Heating season in Gdansk area	1.93	1.68	1.05	2.67
Summer season in Gdansk area	4.71	4.98	6.19	4.98
Annual in Gdansk area	2.56	2.62	2.65	2.55

to what extent the traffic affects air quality, and thus the concentration of the *BTEX* in the air in the monitoring area. In the case when the numerical value of the *T/B* parameter fluctuates in the range from 1.5 to 4.3 or 1.3 to 3.0, it can be concluded that the traffic is a major source of *BTEX* emissions into the air in the monitoring area. The greater the numerical value of the *T/B* parameter, the greater the impact of the traffic on the concentration of *BTEX* in the atmosphere in the monitoring area (Khoder 2007; Miller et al. 2011, 2012). Interpreting the information summarized in Table 3 , it can be noticed that the largest numerical values of the *T/B* parameter were determined for the areas around the measuring/monitoring stations AM2 and AM8. The obtained results lead to the conclusion that the main source of the emission of *BTEX* into the air in the area around the measuring/monitoring stations AM2 and AM8 is the traffic. This information is consistent with the previously presented data concerning the characteristics of the area around measuring station AM8, where one of the main roads is located. In the case of the area around measuring/monitoring station AM2, results indicate that the construction traffic (trucks) works in close proximity to the measuring/monitoring station has a significant effect on the *BTEX* concentration in the atmospheric air. The smallest numerical value of the *T/B* parameter was recorded for the areas around measuring/monitoring station AM5. This type of information allows the conclusion that the movement of motor vehicles is not the only source of emission of *BTEX* into the air (as in the previously described cases). This is due to the fact that the measuring/monitoring station is in a location characterized by a low degree of urbanization and the lack of a main traffic route in close proximity. Significant differences were noted in the obtained results in the case of the determined seasonal (heating season and summer season) numerical values of the *T/B* parameter. The significantly lower numerical value of the *T/B* parameter determined during the heating season can indicate that the movement of motor vehicles powered by combustion engines is not a major source of *BTEX* emissions into the air in Gdansk. During the heating season, the concentration of *BTEX* (mainly benzene) can be affected by the low

efficiency of domestic heating systems which use low-quality coal as fuel. However, in the summer, the calculated numerical value of the *T/B* parameter clearly shows that the main source of emissions of *BTEX* into air in Gdansk is the movement of motor vehicles powered by combustion engines. The differences between the numerical values of *T/B* parameters calculated for the areas around the measuring/monitoring stations may be caused by the following factors: the intensity of motor vehicle usage, the age and type of cars on main roads, the quality and type of liquid fuel used, and the type and activity of industrial centers located in the area of measuring/monitoring stations (Murena 2007).

Another parameter that allows the obtaining of analytical information about air quality in the urban areas covered by monitoring is the *m, p-X/E* parameter. Determination of this parameter allows the estimation of the "freshness" of emission sources located in the area covered by the monitoring, and the obtaining of preliminary information about the intensity of the photochemical reactions occurring in the atmosphere—photochemical age of air mass (Chang et al. 2006; Hsieh et al. 2011). In the scientific literature devoted to topics related to air quality studies in urban areas, *BTEX* are transported from fresh emission sources (fresh air mass), where the numerical value of the determined *m, p-X/E* parameter is greater than 3.3 (Miller et al. 2011). Referring to the data presented in Table 3, it can be concluded that the numerical value of the *m, p-X/E* parameter was greater than 3.3 only in the spring in Gdansk. This leads to the conclusion that in the summer, *BTEX* are emitted from "fresh" emission sources, i.e., as a result of the combustion of liquid fuels in motor vehicle combustion engines. On the other hand, considering the numerical value of the *m, p-X/E* parameter determined for the whole area of Gdansk (2.56), it can be concluded that *BTEX* emitted from different types of emission sources (motor vehicles, low-efficiency domestic heating systems, the activity of different types of industries) may undergo a degradation process as a result of photochemical reactions taking place in the atmosphere involving substances classified as oxidizing agents, i.e., hydroxyl radicals or tropospheric ozone.

Conclusions

As a result of monitoring studies in Gdansk from January to December 2013, it was found that the determined annual average benzene concentration in ambient air does not exceed the value of 5 $\mu g/m^3$ defined in the European legislation. Interpreting the obtained results, it was found that the level of benzene in the ambient air in Gdansk may be affected by three main sources of emissions: the movement of wheeled vehicles powered by combustion engines (combustion of liquid fuels), the combustion of low-quality coal used as fuel in individual heating systems with low efficiency, and seasonal activity in

gardens and residential recreation areas. It was also shown that the temperature of the ambient air has a significant effect on the concentration of benzene in the atmosphere in the monitoring area—during the heating season significantly higher levels of benzene in the atmosphere than in the summer were reported.

According to the analysis of data related to weather conditions and the benzene concentrations in ambient air in 2011–2013, it was concluded that knowledge about wind velocity can explain the value of the daily variation of benzene levels from 6.0 to 35 % in ambient air in different seasons.

According to the analysis, meteorological conditions during the heating season have the greatest effect on benzene concentration in the atmosphere in Gdansk. Analysis of episodes where high concentrations of benzene were reported showed that the most important factors causing the occurrence of this type of phenomenon are as follows: anticyclonic atmospheric circulation, stable atmosphere equilibrium, low wind velocity, the occurrence of an inversion layer, and the height of the mixing layer. As a result of *t-Student* and *F-Snedecor* statistical tests, it was found that the determined benzene concentrations using the passive sampling of analytes from the gaseous medium in 2011–2013 do not differ statistically significantly in terms of accuracy and precision in comparison with the application of the automatic on-line analyzer. It has been shown that the passive sampling technique can be successfully applied as an alternative to costly stationary automated analyzers. By interpreting the determined numerical values of inter-species ratios of *BTEX* parameters, it was found that the presence in the atmosphere of *BTEX* in Gdansk results mainly from the use of motor vehicles powered by combustion (combustion of liquid fuel). Furthermore, it has been shown that active industrial centers located in the monitoring area can have an influence on the amount of analytes from the *BTEX* group (mainly toluene, ethylbenzene, and total xylenes) in the atmosphere. In addition, it was concluded that human activity resulting from construction activities in close proximity to a monitoring/measuring station significantly affect the results of research, mainly the concentration of toluene, ethylbenzene, and total xylenes in atmospheric air.

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