

BENZENE AIR POLLUTION IN THE SILESIAN REGION

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ZANIECZYSZCZENIE BENZENEM POWIETRZA W WOJEWÓDZTWIE ŚLĄSKIM

W artykule przedstawiono wyniki monitoringu benzenu w powietrzu, w 19 punktach na terenie województwa śląskiego. Punkty monitoringu wytypowano z uwagi na gęstość zaludnienia i intensywność ruchu samochodowego lub ze względu na obecność przemysłowych źródeł emisji benzenu (koksownie, rafinerie). Pomiary prowadzono metodą pasywną, z zastosowaniem rurek sorpcyjnych wypełnionych węglem aktywnym. Czas ekspozycji rurki – 2 tygodnie. W każdym punkcie, w okresie od sierpnia 2002 r. do lipca 2003 r. wykonano 12 ekspozycji. Wyniki pomiarów wykazały zróżnicowane narażenie mieszkańców śląskich miast na ponadnormatywne stężenie benzenu. Stężenie średnioroczne przyjmowało wartości z zakresu 4,09 $\mu\text{g}/\text{m}^3$ do 9,26 $\mu\text{g}/\text{m}^3$, a więc nie przekroczyło wartości dopuszczalnej wraz ze 100% marginesem tolerancji. W większości punktów zostało przekroczone średnioroczne stężenie dopuszczalne 5 $\mu\text{g}/\text{m}^3$. Przedstawiono również wyniki przykładowej sesji pomiarowej w Zabrzu przy użyciu automatycznego analizatora BTX. W równoległe prowadzonej ekspozycji próbnika pasywnego uzyskano podobną wartość stężenia benzenu, jak w pomiarach automatycznych.

Summary

The paper presents results of benzene monitoring in ambient air in the Silesian Region (Poland). Sampling points within the Region were chosen with respect to the population density, traffic intensity and presence of industrial emission sources of benzene (coking plants, refineries). Benzene concentrations were measured by means of the passive sampling by using sorption tubes filled with charcoal. The exposure time of each tube was 2 weeks. At each sampling point 12 tubes were exposed from August 2002 to July 2003. The results of the experiment proved differentiated degrees of exposure of the Silesian inhabitants to elevated, exceeding the limit value (5.0 $\mu\text{g}/\text{m}^3$), concentrations of benzene. The mean annual concentration of benzene varied over the Region between 4.09 and 9.26 $\mu\text{g}/\text{m}^3$, not exceeding the doubled standard (i.e. the limit plus 100% margin of tolerance, i.e. 10 $\mu\text{g}/\text{m}^3$ in total). The limit value for benzene concentration was exceeded at the majority of the sampling points. Results of the exemplary measurement performed in Zabrze with the use of an automatic BTX analyzer are also presented. The parallel exposure of a passive sampler gave similar benzene concentration.

INTRODUCTION

Benzene is the air pollutant commonly occurring in the urban ambient air. It comes mainly from combustion of liquid fuels in car engines. Processes such as coal coking, power production or some petrochemical ones also introduce benzene and its homologues into the atmosphere. Benzene and also some other monocyclic aromatic hydrocarbons are especially harmful to living organisms – they are carcinogens.

In Poland, like in EU countries, for the purposes of health protection, the limit of $5.0 \mu\text{g}/\text{m}^3$ is imposed on the annual mean concentration. However, the 100% margin of tolerance, functioning as a provisional mitigation, is allowable till the end of 2005, what, at the time being, gives the impassable value of $10 \mu\text{g}/\text{m}^3$.

Benzene is one of a few air pollutants the monitoring of which was not obligatory in urban areas till the Framework Directive 96/62/EC [4] and its Daughter Directive 2000/69/EC [6] came into force. Implementation of these directives yielded, in midst of 2002, introduction of new regulations on the atmospheric air quality assessment in Poland [14], where, among other things, new requirements concerning monitoring of benzene were specified.

Till then, in Poland, systematic measurements of benzene concentration had been carried out only around important industrial sources of BTX (e.g. installations for ethylbenzene production, big coking plants, etc). Also some emergency measurements had been performed.

Systematic measurements of benzene concentrations in the Silesian Region have been performed since September 2001. In the beginning, the concentrations were measured at 6 sites in the Upper Silesian Agglomeration – heavily urbanized central part of the Region; and since August 2002 – at 19 sampling points located in 18 cities scattered over the whole Region [9, 10]. This (intended as initial) system of measurements was to help developing of the regional monitoring system of benzene. Now it is being used to create an optimum program of monitoring (a method and timetable of measurements) for a system of urban background, traffic and industrial stations.

The measurements were done by using passive sampling (no active intake system needed). The method is based on the phenomenon of diffusion from an environment of higher concentration (polluted air) to an environment with lower concentration (sorption material) [1, 2, 11, 12]. This way of sampling has been applied for years to the air pollution measuring, especially at work-stands. Recent years have brought an increasing interest in this relatively cheap method allowing sampling of benzene to observe the atmospheric air quality (projects MACBETH, RESOLUTION [3]).

The paper presents the results of using the passive sampling to determine distributions of the mean annual concentrations of benzene and other monocyclic aromatic hydrocarbons in the Silesian Region.

At one of the nineteen measuring points (Zabrze, IEE – Institute of Environmental Engineering), some extra measurements of BTX were performed automatically. These measurements, for lack of technical means for receiving reasonable continuous series of results, provided information on variability of the BTX concentrations only within some several-weeks' periods. Although the received information may be rather complementary to the passive measurements performed at this measuring point, it allows verifying of the indications of the diffusive samplers.

SAMPLING AND ANALYTICAL METHODS

METHOD WITH PASSIVE SAMPLING

BTX were sampled by using ORSA 5 samplers: glass tubes, 28 mm long, with inner diameter 8 mm. The ORSA 5 tube is filled with charcoal (sorption material). Both ends of the tube are closed with "diffusion barrier" – material regulating rate of the diffusion and preventing from excessive desorption. The rate of diffusion depends on a difference between the concentrations inside and outside the tube, and on meteorological conditions (mainly temperature and wind speed) – respectively to which, in the following calculations, proper corrections are taken into account.

The samplers were placed in casings protecting them from precipitation and installed from 3 to 5 meters above the ground, with free access of atmospheric air provided. The time of exposure of a sampler was 14 days. The samplers were not "doubled". There was only one snap-holder for one ORSA 5 sampler in the original mounting case, what was due to relatively high consistency of results of laboratory tests in the controlled atmosphere of BTX and results of experiments in field conditions. As the producer claims, the median of relative standard deviations of three-sample expositions at the near limit concentration of benzene ($5 \mu\text{g}/\text{m}^3$) is 3.4% and sharply decreases with concentration increasing [8]. In another experiment with the ORSA 5 samplers (Duisburg, Germany), consisting in 1324 exposures of pairs of samplers, the standard deviation from the measurements was $0.14 \mu\text{g}/\text{m}^3$, what makes 2.8% of the benzene limit value $5 \mu\text{g}/\text{m}^3$ [13].

After exposure, the sorption material underwent desorption – solvent extraction with carbon disulphate (CS_2) with addition of butyl acetate as an internal standard. Next, the extract was analyzed with the use of a gas chromatograph. The main part of the analytical work was done in the laboratory of the Institute of Environmental Engineering (IEE) and laboratories of Silesian Regional Inspectorate of Environment Protection (SRIEP) in Częstochowa and Bielsko-Biała. Methodologies used in all laboratories were the same – the difference consisted in the gas chromatograph configurations only. In the SRIEP laboratories the configuration GC/FID was used, in the IEE – GC/MS. The configuration GC/MS, applied in the IEE, is described below.

The qualitative and quantitative analyses were performed with the use of a Varian Star 3400 CX gas chromatograph equipped with a Saturn-3 mass spectrometer with electron ionization, and DB-1 capillary column (60 m x 0.32 mm, film thickness 1 μm). The conditions of the analysis were following:

- temperature of the column was programmed to increase from 60 to 200°C at 4°C/min, and from 200 to 250°C at 14°C/min,
- temperature of the injector was 250°C,
- temperature of the source (EI) was 230°C.

The qualitative analysis was performed on the basis of the retention times and library of mass spectra. The method of an internal standard (butyl acetate) was applied in the quantitative analysis.

According to the producer suggestion, the recovery coefficients for determined compounds were assumed to be 1.00. The sampling rates at 20°C were $6.44 \text{ cm}^3/\text{min}$ for benzene and $5.72 \text{ cm}^3/\text{min}$ for toluene. The ambient concentrations C [$\mu\text{g}/\text{m}^3$] were calculated from the following formula:

$$C = \frac{(m_d - m_b) \cdot 10^6}{SR \cdot t}$$

where: m_d – mass of desorbed analyte [μg],
 m_b – blank of analyte [μg],
 SR – sampling rate [cm^3/min],
 t – exposure time [min].

To assess accuracy of the method for quantitative determination of BTX in the extract, the analysis was repeated ten times. The results allowed determining of the relative standard deviations (RSDs). The RSD value ($n = 10$) was 5.4% for benzene and 8.5% for toluene.

Moreover, the detection limits were determined. The detection limit was $0.3 \mu\text{g}/\text{m}^3$ for benzene and $0.4 \mu\text{g}/\text{m}^3$ for toluene [9, 10].

Neither the IEE nor SRIEP provided conditions for estimating the expanded uncertainty. The expanded uncertainty is defined by the producer and in it – among other parameters – uncertainty of sampling rate (repeated experiments in standard atmospheres), variations in geometry of samplers and microenvironmental factors (field experiments with multiple samplers), influences of extreme temperature, wind speed, humidity are taken into account.

The uncertainty budget of input parameters is discussed in [8]. According to [8], the expanded uncertainty for measurements of benzene with use of ORSA 5 samplers equals 20%, what is below the 30% accuracy for indicative measurements demanded by the Directive 2000/69/EC [6].

METHOD INVOLVING BTX ANALYZER

An automatic chromatograph AirmoBTX (Chromato-Sud), comprising a sampling system with a sampling unit, thermodesorber, analytical column (MTX-5, 9 m x 0.52 mm, film thickness 1 μm) and flame ionization detector (FID) was used.

A 750 cm^3 sample of gas was automatically taken at the rate of $50 \text{ cm}^3/\text{min}$. Volatile compounds were adsorbed on Carbotrap B bed. The sampling system comprised three sorption tubes. Two tubes worked alternately – while the desorption from one of them was occurring, the second one was sorbing pollutants. The thermodesorption of analyte lasted 3 min at the temperature of 300°C .

The quantitative determination was performed with FID, an integral part of the system. The temperature of the analytical column was changing from 45 to 150°C at $20^\circ\text{C}/\text{min}$ and the terminal temperature of the column was maintained for 2 min. The flow-rate of hydrogen (carrier gas) in the column was $0.5 \text{ cm}^3/\text{min}$. The temperature of the detector was 150°C . Determined compounds were identified by their retention times with the use the external standard method. The standard mixture contained benzene, toluene, ethylbenzene, m-xylene, p-xylene, o-xylene, 1,2,3-trimethylbenzene, 1,3,5-trimethylbenzene. Concentration of each compound was 0.03 ppm.

For the statistical purposes, the standard mixture was analyzed ten times by using the automatic method. The results allowed determining the relative standard deviations (RSDs) and confidence intervals for the method. The RSD ($n = 10$) was 0.81% for benzene

and 1.00% for toluene. The confidence interval was $116 \pm 0.50 \mu\text{g}/\text{m}^3$ for benzene and $145 \pm 0.62 \mu\text{g}/\text{m}^3$ for toluene.

To determine the detection limit, the standard mixture was diluted in nitrogen so that each of the mixture components was at a concentration of $500 \text{ ng}/\text{m}^3$. The detection limit was $0.3 \mu\text{g}/\text{m}^3$ for benzene and $0.4 \mu\text{g}/\text{m}^3$ for toluene [5]. The detection limit determined in the laboratory conditions was congruent with the parameters guaranteed by the producer.

MONITORING SITES AND SCHEME OF MEASUREMENTS

The sampling points were located in 18 cities of the Silesian Region (Tab. 1). Two sampling points of different kinds were in Częstochowa (points No. 1 and 11). The location sites of these points were of great population density. In seven cities, out of the total 18, industrial utilities related to unorganized emission of BTX exist. These are coking plants in Częstochowa, Zabrze, Dąbrowa Górnicza, Czerwionka Leszczyny, Bytom and an oil refinery in Czechowice Dziedzice. The sampling point in Wodzisław Śląski is affected by the nearby coking plant "Radlin" and also by a few Czech coking plants. The sampling points in Częstochowa (No. 1), Bielsko-Biała (No. 10), and Tychy (No. 13) are representative of situation in the vicinity of roads (traffic stations). The rest of the points represent the urban background.

In each of these 19 sampling points, 12 sample takings were performed (12 two-week exposures). The sampling started in August 2002, ended in July 2003, and, generally, each next exposure began a month after beginning of the previous one.

At the point No. 6 in Zabrze (premises of the IEE), the automatic BTX analyzer, working in irregularly distributed time intervals (several weeks' continuous measurements), is installed. In this paper, results of the measurement series, simultaneous with a passive sampler exposure in the period from May 20 to July 30, 2003, are presented.

RESULTS AND DISCUSSION

METHOD WITH PASSIVE SAMPLING

Measured concentrations of benzene are presented in Fig. 1. The stack-bar charts present results received for a sampling point in consecutive exposures. To enhance readability of the charts, the bars filling is related with the concentration: below limit value (LV), between LV and LV + margin of tolerance (MT), above LV + MT. The horizontal line denotes the mean concentration.

The bars on the left and right sides of the charts represent results for the hot season, the inner bars – for the heating season of the year.

Mean concentrations of benzene and toluene are compiled in Tab. 2. The mean values of concentrations are taken over the whole year, summer (April – September) and winter (the rest of the year), respectively.

The total time of exposure of a single diffusive sampler was about a half of the year – it is a good basis for estimating the annual mean concentration of benzene.

The mean concentration of benzene at the 19 investigated points varied from point to point between $4.09 \mu\text{g}/\text{m}^3$ (No. 5, Katowice Załęże) and $9.26 \mu\text{g}/\text{m}^3$ (No. 10, Bielsko-Biała) –

Table 1. Characteristics of the sampling points

Point No.	City	Kind of the sampling point	Notes about localization
1	Częstochowa	traffic station (cokery)	city centre, crossroad with heavy traffic
2	Dąbrowa Górnicza	urban background (cokery)	city centre, about 200 m from main communication arteries, quarter of low houses.
3	Czechowice-Dziedzice	urban background (refinery)	quarter of low houses, about 400 m from quite busy road, 1 km from industrial terrains
4	Wodzisław Śląski	urban background (cokery near to Radlin)	vicinity of living quarters, far from roads
5	Katowice	urban background	neighborhood of a station for monitoring of country background pollution, near to big living quarter and trade and service centre
6 *	Zabrze	urban background (cokery)	centre of densely populated area, 100–200 m from roads
7	Ruda Śląska	urban background	quarter of blocks of flats, near to trade and service centre
8	Bytom	urban background (cokery)	crossing of main roads in city centre, old houses (heating furnaces).
9	Chorzów	urban background	old low houses (heating furnaces), near to chemical plant
10	Bielsko-Biała	traffic station	city centre, main communication tract
11	Częstochowa	urban background	quarter of multifamily houses, periphery of city
12	Sosnowiec	urban background	city centre, about 50 m from traffic circle
13	Tychy	traffic station	cross road in city centre
14	Gliwice	urban background	city centre, living and service quarter, 20 m from quite busy road
15	Czerwionka Leszczyny	urban background (cokery)	living and service quarter, parking lot
16	Rybnik	urban background	urban terrain, neighborhood of old houses with individual furnaces
17	Racibórz	urban background	neighborhood of bus terminal
18	Jastrzębie Zdrój	urban background	densely populated living quarter
19	Cieszyn	urban background	sampling point oriented towards assessment of transboundary transport of pollutants

* additionally – point of automatic measurements

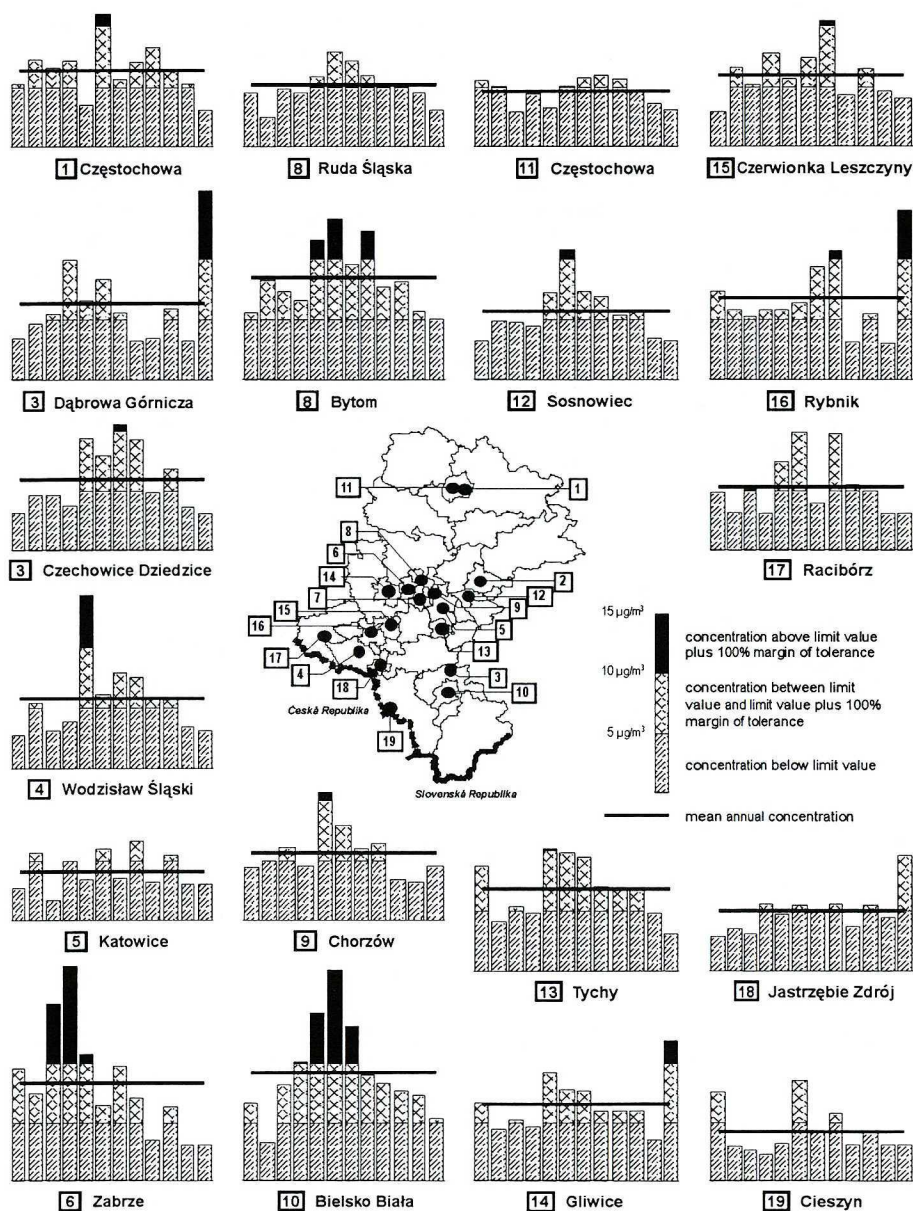


Fig. 1. The results of passive measurements of benzene concentrations in the Silesian Region, August 2002 – July 2003

the permissible value $10 \mu\text{g}/\text{m}^3$ (LV+MT) was never exceeded. The value LV – $5 \mu\text{g}/\text{m}^3$ – was exceeded at majority of the points. At 4 points: Katowice Załęże (No. 5), Częstochowa (No. 11), Jastrzębie Zdrój (No. 18), and Cieszyn (No. 19), the annual mean concentrations of benzene were between 4.0 and $5.0 \mu\text{g}/\text{m}^3$.

Table 2. Summer, winter and annual mean concentrations of benzene and toluene in 19 sampling points in the Silesian Region; the results of passive measurements in the period August 2002 – July 2003

Measurement point Point No. – City	Mean concentration* [$\mu\text{g}/\text{m}^3$]					
	Benzene			Toluene		
	summer	winter	year	summer	winter	year
1 – Częstochowa (traffic station)	5.50	7.01	6.38	9.32	9.18	9.24
2 – Dąbrowa Górnicza	6.39	6.16	6.27	4.41	5.09	4.75
3 – Czechowice-Dziedzice	4.29	7.64	5.97	4.40	6.01	5.21
4 – Wodzisław Śląski	3.90	7.56	5.73	4.22	5.84	5.03
5 – Katowice	3.59	4.59	4.09	4.23	4.46	4.34
6 – Zabrze	7.37	9.28	8.33	5.36	5.47	5.41
7 – Ruda Śląska	4.09	6.15	5.12	4.88	5.89	5.38
8 – Bytom	6.61	10.17	8.39	10.34	12.01	11.18
9 – Chorzów	4.61	6.46	5.62	6.86	8.20	7.53
10 – Bielsko-Biała	6.39	12.13	9.26	12.08	20.33	16.20
11 – Częstochowa (urban background)	4.13	4.99	4.56	6.03	5.31	5.67
12 – Sosnowiec	4.21	6.96	5.58	8.72	9.04	8.88
13 – Tychy	5.43	8.00	6.71	8.61	10.86	9.73
14 – Gliwice	6.48	6.60	6.55	7.24	8.23	7.82
15 – Czerwonka Leszczyny	4.93	6.80	5.95	3.97	5.60	4.86
16 – Rybnik	6.77	6.81	6.79	4.17	5.33	4.80
17 – Racibórz	3.98	6.61	5.29	3.73	6.57	5.28
18 – Jastrzębie Zdrój	5.16	4.69	4.89	4.11	4.57	4.38
19 – Cieszyn	3.84	4.47	4.16	3.27	3.73	3.50

* arithmetic mean of all results („year”) or results of exposure in summer („summer” – 6 months from April to September) or results of exposure in winter („winter” – the rest of the year)

Visibly higher concentrations of benzene occurred in the direct vicinity of roads (Bielsko Biała, No. 10) and at sites (cities) where effects of industry add to the transportation effect (Zabrze – No. 6, Bytom – No. 8).

Analysis of the benzene/toluene concentration ratios yields the BTX profiles typical of communication for:

- Bielsko-Biała and Bytom (street canyons along main communication tract in the city centre),
- Tychy and Częstochowa (crossroads in the city centers).

For both seasons, in Zabrze, Dąbrowa Górnicza, Rybnik, and Czerwionka Leszczyny, higher concentrations of benzene than of toluene were observed – it means presence of other, than the transportation, sources of benzene (chemical industry, coking plants).

METHOD INVOLVING BTX ANALYZER

The measurements conducted in Zabrze with the use of a BTX analyzer, despite of their good time resolution (15 min), may be considered merely „indicative measurements“ – the total measuring period covers less than one half of a year. However, they provide valuable information on instantaneous and diurnal variability of the BTX concentrations – the information that cannot be received by using the passive sampling due to the two-week’ averaging period.

In Fig. 2, the series of 1 h measurements of the benzene concentration and air temperature, covering the whole period of one passive sampler exposure (May 20 – June 03, 2003), is presented. The horizontal line denotes the mean benzene concentration equal $7.0 \mu\text{g}/\text{m}^3$. Concentration yielded by the passive method within this same period was $5.3 \mu\text{g}/\text{m}^3$. The relative error of the less sensitive passive measurements was then 24%, not exceeding the uncertainty level of 30% allowable in periodic measurements of benzene concentration [7].

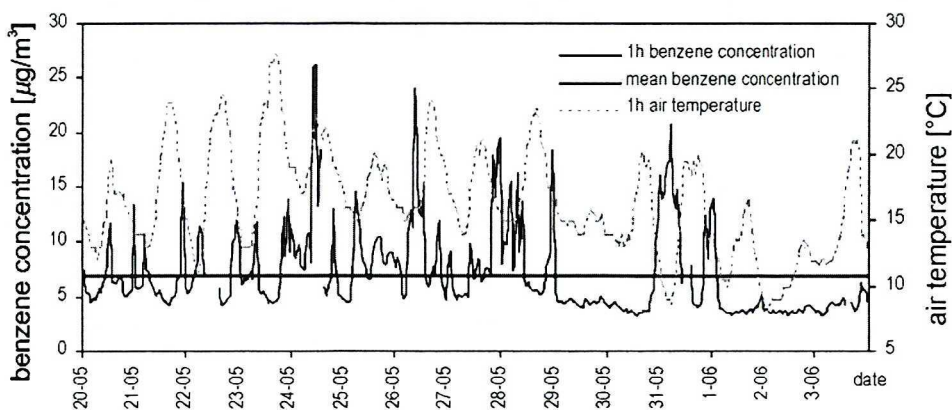


Fig. 2. Series of 1h measurements of benzene concentration and air temperature in the period from May 20 to June 03, 2003

In Fig. 3, the diurnal distributions of the benzene and toluene concentrations for the analyzed period are presented. Consistency of the two distributions suggests common origin of the pollutants. Till noon, strong effect of the very busy access road, distant by about 170 m from the measuring point and connecting the centre of Zabrze with the state road number 4 (E40), may be noted. The lowest concentrations of benzene and toluene, slightly increasing between 4:00 and 5:00 p.m., occur in the afternoon matching the maximum temperature period. Diurnal variability of BTX concentrations fits the variability of concentrations of main air pollutants and is caused by changes of thermodynamic parameters of the mixing layer. Both morning and evening peaks of concentration occur, but hours of their occurrence change over a year (in winter higher concentrations occur in later hours).

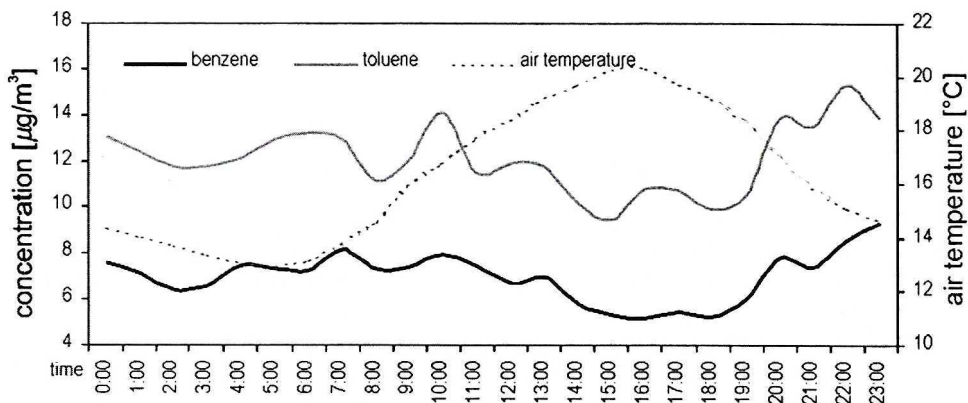


Fig. 3. Diurnal distributions of benzene and toluene concentrations and air temperature in the period from May 20 to June 03, 2003

Fig. 4 also illustrates diurnal distribution of the benzene concentration within the considered period. Minimum, maximum and mean concentrations for each of 24 hours of a day are taken over the whole period and plotted in the bar-chart. The mean concentration is denoted by the horizontal line. The widest concentration variability intervals for benzene and the most frequent episodes of its high concentration occur for hours 10:00 a.m. and 11:00 p.m.

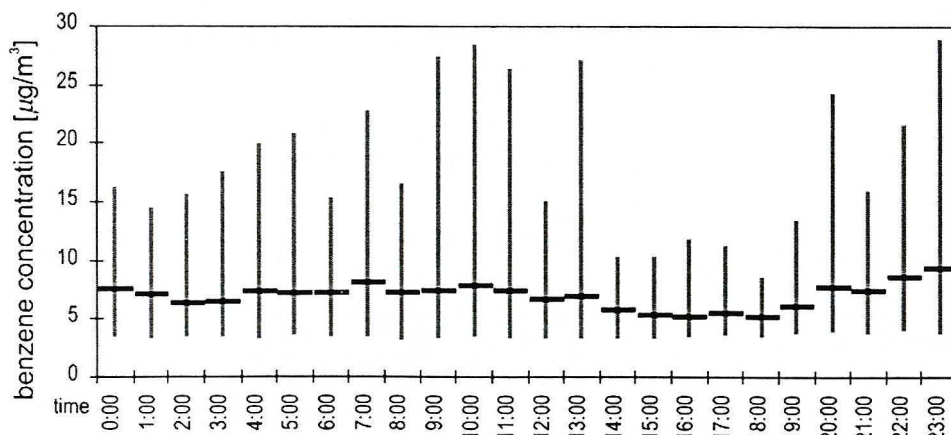


Fig. 4. Diurnal distribution of variability range of benzene concentration in the period from May 20 to June 03, 2003; the vertical line denotes the minimum-maximum range, horizontal – mean concentration

Differences between distributions of instantaneous and diurnal concentrations of benzene may be seen in Fig. 5. Within two weeks of the measuring period, the maximum 24 h concentration of benzene was $18.5 \mu\text{g}/\text{m}^3$ (May 31, 2003 – low 24 h wind speed, about 0.3 m/s). The maximum 1 h concentration of benzene in this period was $26.1 \mu\text{g}/\text{m}^3$. Distinctly lower concentrations of benzene were noted in the end part of the considered period, what was due to rains and higher than average wind speeds. Increase in concentration was observed on May 31, 2003, when it stopped to rain and the greater part of the day was windless.

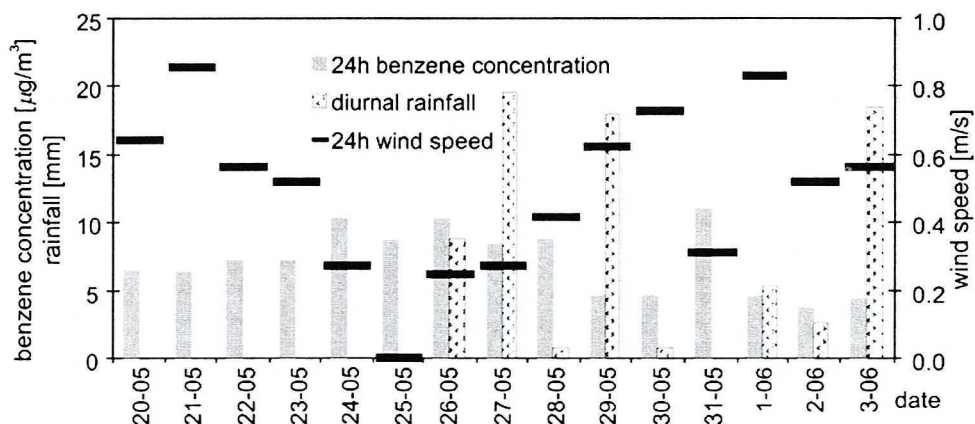


Fig. 5. The 24 h benzene concentration, 24 h wind speed and diurnal rainfall in the period from May 20 to June 03, 2003

CONCLUSIONS

The results of passive measurements confirm the hypothesis that in the Silesian Region there exists only limited risk for benzene concentration to exceed the standard $10 \mu\text{g}/\text{m}^3$ (LV + MT), valid till 2005. Instead, at almost all sites, where elevated level of benzene concentration was expected, the limit value of $5 \mu\text{g}/\text{m}^3$ (assumed for health reasons), was exceeded. Thus, it should be stated that the air quality is not satisfactory in majority of Silesian cities.

Using passive samplers to determine mean concentrations of benzene for purposes of yearly assessment of air quality seems to be a good solution because of low costs. However, when the annual concentrations, measured at non-traffic stations, exceed summed together the admissible concentration and tolerance margin, some other, more detailed and of better time resolution, approach to the situation is necessary. This especially concerns densely populated areas with elevated health hazard. Within the urban background areas, unaffected directly either by traffic or elevated sources, the results from the passive method are in agreement with – for example – data from an automatic BTX analyzer, as it was in the presented here Zabrze case. But if a very effective industrial source of BTX exists in a city, frequent high concentration episodes are not scored by a passive sampler. The sampler records lower concentrations than an automatic analyzer does – as it happened within the period from September 2001 to April 2002 in Zabrze, when two cokery objects were operating simultaneously (the installation for permanent tar distillation and “Makoszowy” Cokery, the latter liquidated in April 2002). Such a situation demands an additional, manual or automated aspiratory method, supported by the parallel observations of the meteorological parameters, to confirm concentrations coming from the passive measurements. Only such an approach, implemented within the scope of an air protection program, allows spotting the causes of and undertaking measures against violations of the air quality standards.

Considering the passive sampling quite uncertain method, one must notice necessity for continuing investigations to confirm the regularities in the spatial distribution of concentrations and to reveal eventual trends.

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