Environment Protection Engineering

Vol. 42 2016 No. 4

DOI: 10.5277/epe160402

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WIND ROSE AND MODELLING OF THE FATE OF POLYNUCLEAR AROMATIC HYDROCARBONS IN THE CENTRAL POLAND

The wind rose analysis has been performed for the town of Radom in Central Poland. It was found that West and South-West wind directions significantly dominate in the wind rose, causing PAHs influx from coal power stations. The analysis of the PAHs accumulation in water, soil and bottom sediments based on the fugacity and partition coefficients model has been performed for Domaniów Lake. The results have been compared with the experimental PAHs concentrations in the lake, in- and out-flowing waters as well as in the dust over the lake.

1. INTRODUCTION

Environmental problems have invariably a transboundary, regional or global dimension. Increasing amounts of chemicals produced in the world and development of analytical methods for the analysis of pollutants have already turned the attention of researchers towards the environmental issues. The accumulation of DDT in the livers of birds, presence of mercury in the metabolism of Japanese fishermen, smog in great agglomerates (London), presence of polynuclear aromatic hydrocarbons (PAHs) in the air around industrialized areas as well as the emission of CFCs, including refrigerants, destroying the ozone layer can serve as the examples.

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The distribution of pollution is frequently calculated for different regions in all over the world mainly because of their importance for air quality monitoring and propagation of pollutants in the natural environment. Many of such calculations have been done recently for North America [1, 2], Asia [3, 4], North Africa [5] and Europe [6–8]. Some of them concern pollution distribution in a big area such as the part of country [8] or countries [6–8], some are focused on the cities [1, 3, 4] and some on very small, very polluted [7] urban [5] or environmentally important [9] places.

Many algorithms were elaborated for calculation of the pollution distribution [10–12] which allow for very accurate determination of the pollutants partition between different environmental compartments. They are especially important in the case of ecological disasters [9] or precise pollution monitoring [3,5,6].

By many different transport mechanisms long lived chemical pollutants appear in the all environmental compartments: air, water, soil, sediment, aerosols, water-suspended particulates, vegetative biota and animal biota. However, different chemicals appear in these environmental compartments in different amounts. It is important to be able to quantify the distribution of each chemical pollutant among different environmental compartments for a variety of reasons. Firstly, it allows to determine whether a chemical is likely to appear in a compartment (e.g. in animal biota) in such quantity that would make harm or promote chronic disease. Secondly, one should know in which environmental compartment a chemical will appear in the highest concentration, and therefore will be the easiest to detect. It could then be the basis for the efficient environmental monitoring and help to develop new legal regulations on hazardous chemicals.

In Poland, electricity is generated almost exclusively in coal power stations, located mostly in the western part of the country. This causes the presence of polynuclear aromatic hydrocarbons (PAHs – carcinogenic compounds) in the atmosphere. The pollution distribution depends strongly on the winds in the area investigated. So, the knowledge concerning set and speed of the wind is absolutely necessary for proper description of pollution propagation caused by the fall out. This work is concerned with analysis of the wind rose and PAHs accumulation in water, soil and bottom sediments in the central Poland. The town of Radom – the second largest city in Mazowsze region and Domaniów Lake – the second largest lake in Mazowsze region, which can serve as good patterns of pollution distribution in industrial area of the central Poland, have been selected for calculation of the wind rose and PAHs accumulation in water, soil and bottom sediments, respectively.

The analysis is based on the fugacity and partition coefficients model widely used by environmentalists [10–12]. The results of calculation have been compared with the measurements of concentration of PAHs (fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene and benzo[b]fluoranthene) in the lake, in flowing and out flowing waters as well as in the dust over the lake.

2. EXPERIMENTAL PROCEDURES

2.1. THE WIND ROSE

Despite meteorological reports [13, 14], only a few information is available about precisely calculated wind roses for various parts of Poland. They are mainly focused on the Baltic See [15] and northern [16] or southern (mountains) part of Poland [9].

Radom (area of 112 km^2) is located in the southern part of Mazowsze Voivodeship, close to Domaniów lake, on lowlands surrounded by three rivers: the Radomka River from North, the Vistula River from East and the Iłżanka river from South and forests: Kozieniecka Forest from South-East and Stromiecka Forest from North.

The experimental data collected from 1994 to 2003 and used for the calculations of the wind rose were taken from the weather station located in Radom air-field (South-East border of Radom) (*Annual Meteorological Report, 1994–2003*). The area around the station is flat and a little bit sloping down into West and North-West direction. The measurements were carried out 12 m above the ground using an electric wind meter M-47. The left data, for years 2004–2005, were taken from the Voivodeship Inspectorate of Environmental Protection in Radom [13, 14].

The speed and set of wind was measured and recorded every day at 7/8 a.m., 1/2 and 7/8 p.m. Mean day wind speed was calculated according to the equation:

$$\bar{v} = \frac{v_{07/08} + v_{13/14} + v_{19/20}}{3} \tag{1}$$

where: $\bar{v}$ is mean day wind speed, $v_{07/08}$ – wind speed measured at 7/8 a.m., $v_{13/14}$ – wind speed measured at 1/2 p.m., $v_{19/20}$ – wind speed measured at 7/8 p.m.

Based on the mean day wind speeds, the mean monthly wind speed at 7/8 a.m., 1/2 and 7/8 p.m. was calculated. In the case of 16-direction wind rose (Fig. 1), the main 8 directions are N, NE, E, SE, S, SW, W, NW and 8 indirect ones – NNE, ENE, ESE, SSE, SSW, WSW, WNW, NNW. The 16-directions wind rose can be recalculated into 8-direction wind rose [17] by adding to the main set of wind half of the sum of neighbouring sets of wind according to the following equations:

$$N_8 = N_{16} + 0.5(NNW_{16} + NNE_{16})$$

$$NE_8 = NE_{16} + 0.5(NNE_{16} + ENE_{16})$$

$$E_8 = E_{16} + 0.5(ENE_{16} + ESE_{16})$$

$$SE_8 = SE_{16} + 0.5(ESE_{16} + SSE_{16})$$

$$S_8 = S_{16} + 0.5(SSE_{16} + SSW_{16})$$

$$SW_8 = SW_{16} + 0.5(SSW_{16} + WSW_{16})$$

$$W_8 = W_{16} + 0.5(WSW_{16} + WNW_{16})$$

$$NW_8 = NW_{16} + 0.5(WNW_{16} + NNW_{16}) \quad (2)$$

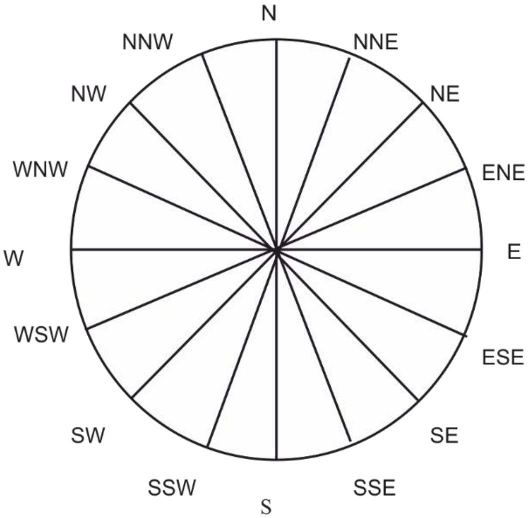

Fig. 1. 16-direction wind rose with main and indirect sets of wind

The wind rose with smaller number of sets of wind can be recalculated also into wind rose with higher number of sets of wind [17]. In this case, the main sets of wind should be divided into indirect sets of wind. If we recalculate 8-direction wind rose into 16-direction one, the main set of wind of this rose is equal to the half of main set of wind of 8-direction wind rose and indirect set of wind of the rose consists of 25% of the sum of neighbouring sets of wind of 8-direction wind rose according to the following equations:

$$N_{16} = 0.5N_8$$

$$NNE_{16} = 0.25(N_8 + NE_8)$$

$$NE_{16} = 0.5NE_8$$

$$ENE_{16} = 0.25(NE_8 + E_8)$$

$$E_{16} = 0.5E_8$$

$$ESE_{16} = 0.25(E_8 + SE_8) \text{ etc.} \tag{3}$$

The above procedure can be applied to recalculation of 16-direction wind rose into 36-direction wind rose. In this case, the circle is divided on 36 parts(10° each) and for main sets of wind is assumed 44% of main sets of winds of 16-direction wind rose and for indirect sets of wind the appropriate values of the of the sum of neighbouring sets of wind of 16-direction wind rose according to the following equations:

$$D_{01} = 0.44N_{16}$$

$$D_{02} = 0.06(N_{16} + 6NNE_{16})$$

$$D_{03} = 0.44NNE_{16}$$

$$D_{04} = 0.09(2NNE_{16} + 3NE_{16})$$

$$D_{05} = 0.44NE_{16}$$

$$D_{06} = 0.15(2NE_{16} + ENE_{16})$$

$$D_{07} = 0.44ENE_{16}$$

$$D_{08} = 0.06(6ENE_{16} + E_{16})$$

$$D_{09} = 0.44E_{16} \text{ etc.} \tag{4}$$

The experimental data (measured wind speed from 1 to 11 m/s and 12 sets of wind) were stored in the file for each month as meteorological cases. In the file one can distinguish 133 different meteorological cases being a possible combination of 11 different wind speeds, 12 different sets of wind and calm cases. Then the number of meteorological cases is calculated for the whole year and presented in the form of table showing dependence of wind speed on the main sets of wind as a function of meteorological cases – yearly wind rose. Further, the wind rose can be presented in a graphical form usually only for 4 following ranges of wind speed: 1–2 m/s, 1–4 m/s, 1–6 m/s and 1–11 m/s. The wind rose obtained in this way is a statistical presentation of particular wind speed in a selected direction, and of the atmosphere state equilibria.

The monthly experimental data of speeds and sets of wind collected for 12 years (from 1994 to 2005) has been recalculated according to the above procedure and twelve (one for each year) 12-direction wind roses have been obtained. As an example, the wind rose for year 2005 is presented in Table 1 and Fig. 2. In the table, dependence of the wind speed on main sets of wind as a function of all meteorological cases taken for the calculation (together with information about calm cases) is presented. Numbers from 1 to 12 define the main sets of wind and every set of wind describes wind from an angle equal to 360°/12 = 30° of the wind rose.

Table 1

Wind rose of The town of Radom in 2005

Wind speed [m/s]	Main sets of wind											
	1	2	3	4	5	6	7	8	9	10	11	12
	Number of meteorological cases											
1	14	72	99	154	96	147	223	174	145	136	122	80
2	18	58	79	219	317	263	292	301	263	164	72	68
3	27	42	48	143	301	215	136	230	331	209	101	26
4	23	17	20	74	117	79	55	157	248	175	63	9
5	4	9	23	48	52	20	17	122	187	97	26	5
6	0	2	9	17	8	8	6	72	176	41	11	0
7	1	0	1	19	3	5	0	35	104	12	1	0
8	0	0	0	0	0	0	0	10	46	10	1	0
9	0	0	0	0	0	0	0	2	14	4	0	0
10	0	0	0	0	0	0	0	0	9	2	0	0
11	0	0	0	0	0	0	0	0	1	1	0	0
Calm cases	414	total number of meteorological cases taken into account										7970

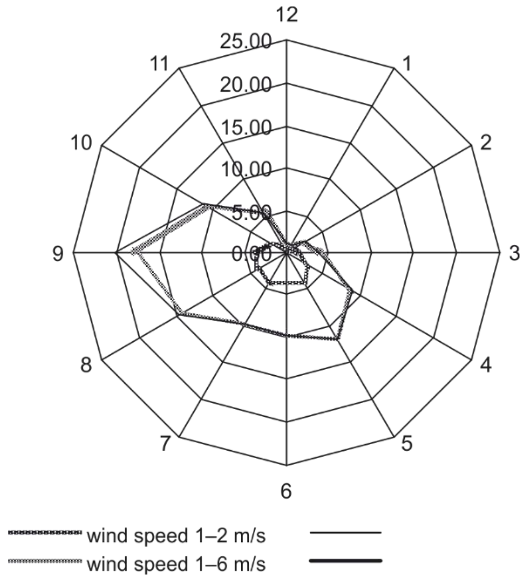

Fig. 2. Wind rose of the town of Radom in 2005

The period of observations is very important for proper construction of a wind rose. If it is too short, the results obtained from the analysis can be very erroneous. The World Meteorological Organization recommends 30 year period [18] for a wind rose analysis but usually is very difficult to analyze such long period. The period of 10 years is assumed in Poland as a standard one for the wind rose analysis. It seems to be adequate for proper determination of aerodynamic conditions in the investigated area, enabling sufficient information on contaminants distribution in the area. Basing on our yearly wind roses from 1994 to 2005 and using the previous calculation procedure, we have calculated the twenty-year wind rose of the town of Radom. It is presented in Table 2 and Fig. 3.

Table 2

Twelve-year wind rose of The town of Radom

Wind speed [m/s]	Main sets of wind											
	1	2	3	4	5	6	7	8	9	10	11	12
	Number of meteorological cases											
1	796	813	1102	847	783	914	1039	1326	1337	1545	1465	1018
2	1058	1062	1859	1684	1771	1664	1847	2412	2413	2397	1925	1398
3	485	657	1437	1891	2006	1634	1759	2630	2512	1856	1305	737
4	239	392	1033	1606	1577	1105	1331	2157	1912	1170	721	323
5	97	209	790	1315	1117	769	1093	1799	1464	676	399	228
6	34	64	487	709	538	458	644	1080	918	359	166	61
7	7	37	327	534	290	269	476	770	728	230	119	39
8	6	19	231	322	205	179	396	667	556	139	66	26
9	0	10	106	177	77	84	171	262	207	45	25	7
10	4	2	19	44	15	39	120	127	90	33	11	26
11	6	1	8	2	2	7	23	22	17	2	3	3
Calm cases	8567	Total number of meteorological cases taking into account										99436

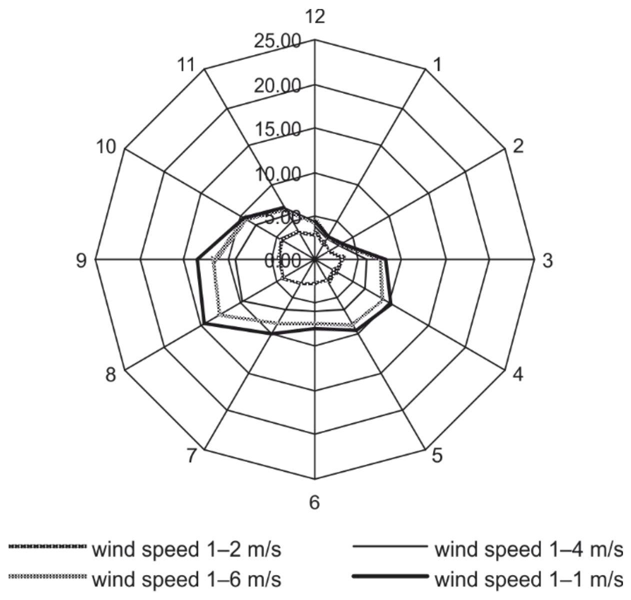

Fig. 3. Twenty-year wind rose of the town of Radom, 1994–2005

Figure 3 shows clearly that West and South-West are the predominant directions of wind in the town of Radom and, as it was stated, in the central part of Poland. Independently of a year studied, these directions significantly dominate in the wind rose what causes the influx of polynuclear aromatic hydrocarbons from coal power stations in the atmosphere of this part of Poland.

2.2. THE PAHS ACCUMULATION

PAHs concentrations in the river waters were determined both in the dissolved phase and adsorbed on suspended solid particles. The surface water samples collected according to the Polish Standard [19] were filtered through a membrane (Baker Separex 47) filter, 0.45 μm . The filtrate (0.6–1.0 L) and the suspended particles left on the filter were analyzed by different procedures:

- Dissolved PAHs were isolated from the filtrate using extraction to the solid phase using the Empore^{TM} extraction disks.
- PAHs adsorbed on the suspended particles retained on the filter were extracted (after drying and weighing) in an ultrasonic field using methanol as a solvent. A similar procedure was used in the analysis of the bottom sediments.

After purification on columns with silica gel, the extracts were analyzed chromatographically by use of a HP 1050 liquid chromatograph equipped with a programmable HP 1046 fluorescence detector. The column used was Vydac 5 PAH 250×4.6 mm with a pre-column Vydac RP C_{18}. Acetonitrile–water mixture was used as a mobile phase. The separation was performed using a gradient system with qualitative identification being carried out based on the standard solutions prepared from the mixture PAH Mix 9 produced by Dr. Ehrenstorfer GmbH containing 16 PAHs chosen by the EPA for determination in environmental samples.

3. THE MODEL

One of the best basic models describing propagation of pollution in the natural environment is the fugacity model [10, 11]. It assumes that pollutant distribution between the environment compartments is strictly connected with its fugacity [10, 11, 20, 21] and refers to the fugacity of octanol-1 and water partition coefficient. Introducing the basic thermodynamic relation [10, 11] defining the fugacity coefficients and their relation for a compound *i* in liquid and vapor phases we get the equation describing vapor–liquid equilibria, which can be further simplified for environmental calculation assuming that in environment each chemical component *i* is present only at very low concentration and air can be treated as an ideal gas phase.

Octanol-1 was selected as a reference compound because of its chemical composition (74% C, 14% H i 12% O) similar to the composition of fat and the fact that mixed with water, it forms two liquid phases. The partition of component i K_i^{O-W} between water (hydrophilic) and organic (octanol-1 – hydrophobic) phases can be defined as follows:

$$K_i^{O-W} = \frac{C_i^O}{C_i^W} \tag{5}$$

where C_i^O, C_j^W are the concentrations of ith component in octanol-1 (O) and water (W).

Hansch et al. [22] have found a linear dependence of the logarithms of the solubility of organic liquid i in water and its octanol-1–water partition coefficients, the logarithm of the partition coefficient being the additive-constitutive property of organic matter. K_i^{O-W} coefficients started to be used as a measure of hydrophobicity and fate of organic matter in environment. Their values for octanol-1–water systems for most pollutants can be found in the literature [23, 24]. Based on the above assumptions, we can derive [10, 11, 20, 21] dependences for vapor–liquid and liquid–liquid equilibria in environment at constant temperature, and further, taking into account K_i^{O-W} values, the partition coefficients of components i for all important environmental compartments (air–water, animal biota–water, vegetable biota–water, soil–water, sediment–water, etc.):

$$K_i^{n-W} = f_i^n K_i^{O-W} \tag{6}$$

where: f_i^n is the fraction of i component in compartment n (fat, vegetable biota, the organic contents of the soil, sediment, and suspended matter).

Modeling of the propagation of selected component i in the environment [11, 20, 21] requires considering integration of various processes such as partition between various environmental compartments, chemical transformation and degradation, emission to the environment, precipitation and transport processes as well as time dependences of concentrations of the components. Having these all necessary equations [11, 20, 21], the calculation of distribution of long lived chemical pollutants in all environmental compartments: air, water, soil, sediment, aerosols, water-suspended particulates, vegetative biota and animal biota can be performed [25].

4. RESULTS

The samples of PAHs (fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, chrysene and benzo[b]fluoranthene) have been collected from four sampling points (Fig. 4) located on two inflow rivers (Szabasówka River – point 1, Radomka River I – point 2), one outflow river (Radomka River II – point 4) and on the lake (point 3

– collecting the dust). The essential inflow and outflow parameters of Lake Domaniów are given in Table 3.

Table 3

Lake Domaniów inflow and outflow parameters

Inflow	
Szabasowka River, m^3/s	2
Radomka I River, m^3/s	4
Outflow	
Radomka II River, m^3/s	6
Volume and area of the lake	
Volume, m^3	6 500 000
Area, km^2	7.5

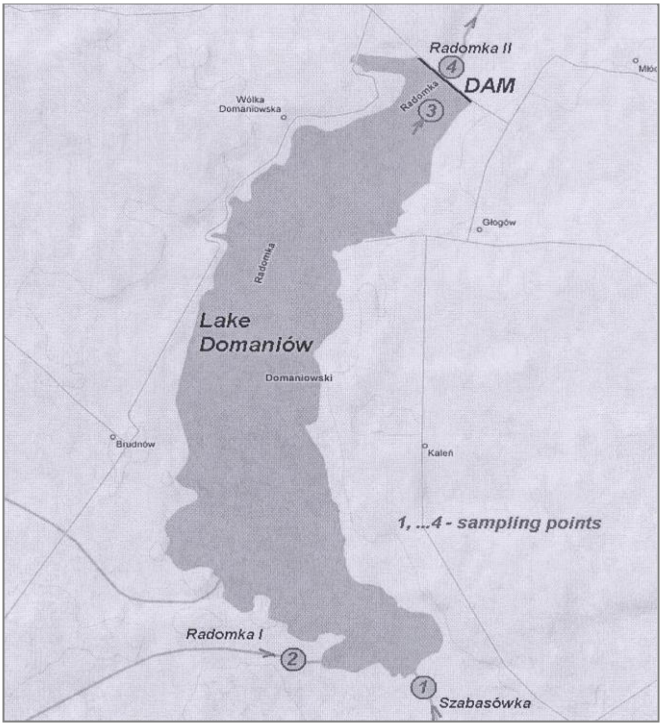

Fig. 4. Sampling points at Domaniów Lake

The measured concentrations of the all investigated PAHs in the sampling points are presented in Table 4. The results show that the concentration of PAHs in this region is very low. The total amount of PAHs in air equals 6.3 ng/m^3, corresponding to ca. 5% of PAHs' concentration in the Silesia region [26]. It means that, despite of the South-

-West wind direction, the influence of the coal power stations located in the South-West part of Poland on the air pollution in the central Poland is very small.

Table 4

Concentrations of the investigated PAHs in the dust over Lake Domaniów and in the inflow and outflow rivers

PAH	Point 3		Point 1 [ng/L]	Point 2 [ng/L]	Point 4 [ng/L]
	Fall [kg/year]	Air [ng/m^3]			
Fluorene	0.1152	1.60	3.48	1.82	5.30
Phenanthrene	0.0432	0.60	5.98	2.85	8.50
Anthracene	negligible	negligible	5.38	4.05	1.99
Fluoranthene	0.1224	1.70	13.17	15.93	16.20
Pyrene	0.072	1.00	7.40	12.57	15.60
Benz[a]anthracene	0.0144	0.20	6.46	13.41	6.76
Chrysene	0.0432	0.60	6.15	10.72	10.46
Benzo[*b*]fluoranthene	0.0432	0.60	0.61	0.86	3.70

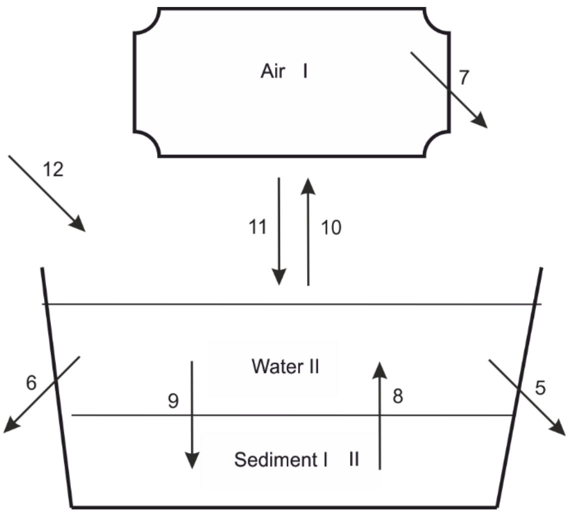

Fig. 5. Flows taken into account during the model calculations:
5–7 – irreversible reaction, 8–11 – exchange, 12 – fallout

The measured concentrations of PAHs for the outflow river (point 4) have been compared with the predicted results given by the fugacity model [25]. The calculations were based on the experimental data for inflow rivers (points 1, 2) and the fallout (point 3). Figure 5 shows all the flows taken into account during the calculation. All necessary auxiliary PAHs data needed for the program running taken from Mackay et al. handbook [23] and Stephenson and Malanowski [24] are summarized in Tables 5 and 6.

Table 5

Physico-chemical properties of the investigated PAHs at 25 °C [23, 24]

PAH	Vapour pressure p^{sat} [Pa]	Solubility in water C_W [mmol/m^3]	$\log K^{O-W}$	Henry's constant H [Pa·m^3/mol]
Fluorene	0.0900	11.43	4.18	7.893
Phenanthrene	0.0200	33.00	4.57	3.240
Anthracene	0.0010	18.84	4.54	3.961
Fluoranthene	0.0012	8.805	5.22	0.957
Pyrene	0.0006	12.89	5.18	0.919
Benz[a]anthracene	2.3×10^{-5}	1.030	5.91	0.581
Chrysene	5.7×10^{-7}	1.599	5.60	0.065
Benzo[*b*]fluoranthene	5.2×10^{-8}	0.150	5.80	0.014

Table 6

Half-life times *t* [h] of the investigated PAHs
in various environmental compartments at 25 °C [23]

PAH	Air	Water	Soil	Sediment
Fluorene	55	550	5500	17 000
Phenanthrene				
Anthracene				
Fluoranthene	170	1700	17 000	55 000
Pyrene				
Benz[a]anthracene				
Chrysene				
Benzo[*b*]fluoranthene				

The results of prediction (absolute and percentage deviations) are summarized in Table 7.

Table 7

Measured and calculated PAH concentrations
in the outflow river Radomka II (point 4) [ng/L]

PAH	Concentration in point 4		Deviation	
	Measured	Calculated	Absolute	Percentage
Fluorene	5.30	6.1	0.8	15.55
Phenanthrene	8.50	8.8	0.3	3.88
Anthracene	1.99	2.4	0.4	22.25
Fluoranthene	16.20	16.1	–0.1	–0.78
Pyrene	15.60	16.5	0.9	5.58
Benz[a]anthracene	6.76	6.9	0.2	2.91
Chrysene	10.46	11.4	1.0	9.97
Benzo[*b*]fluoranthene	3.70	2.8	–0.9	–24.42

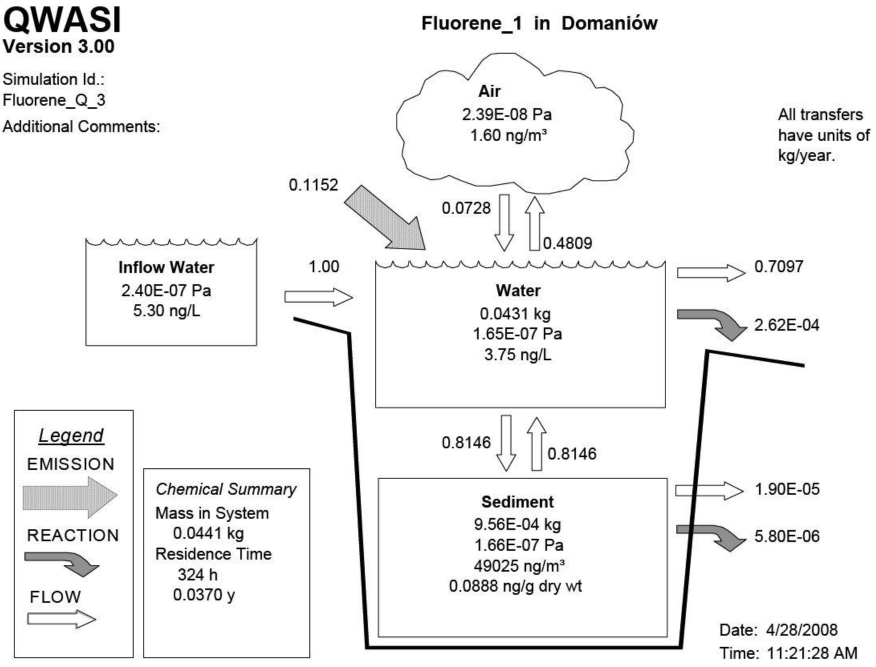

Fig. 6. Distribution of fluorene between all compartments in Domaniów Lake

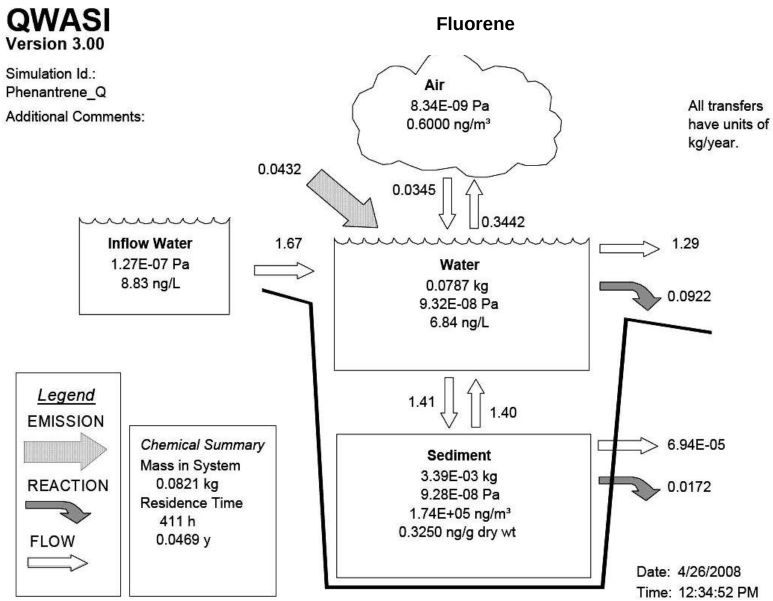

Fig. 7. Distribution of phenanthrene between all compartments in Domaniów Lake

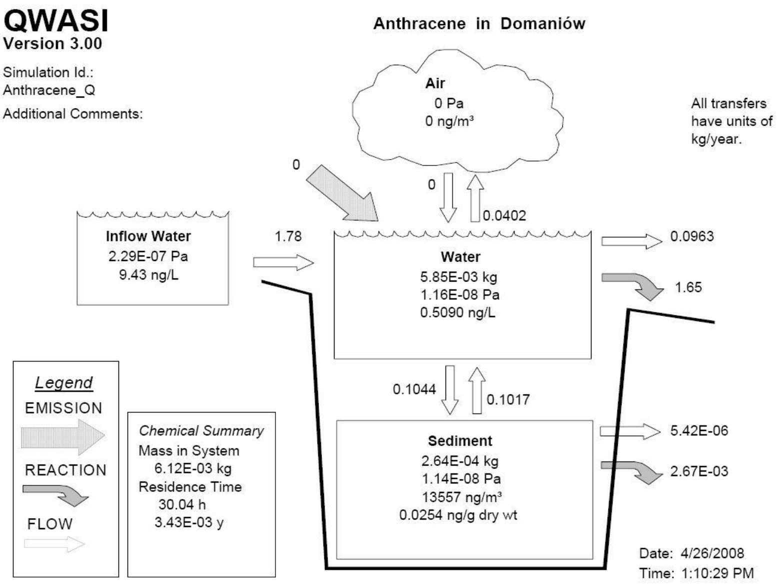

Fig. 8. Distribution of anthracene between all compartments in Domaniów Lake

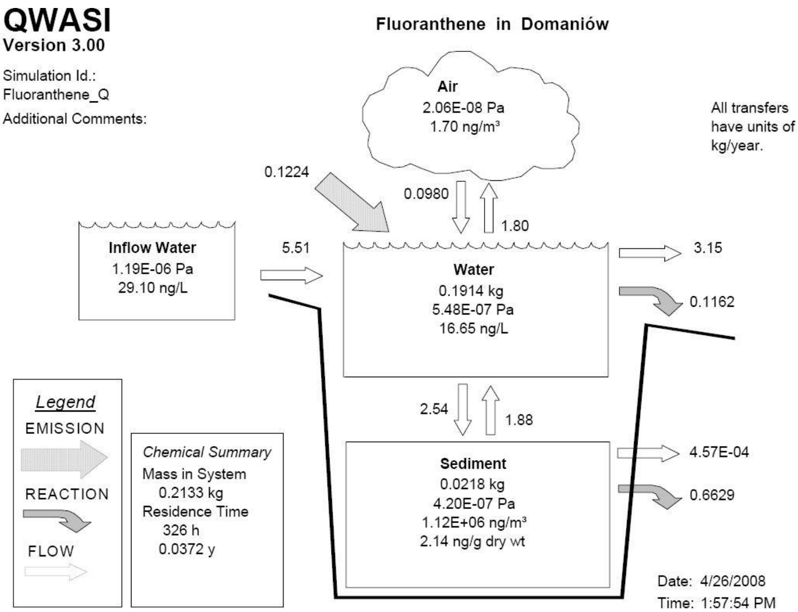

Fig. 9. Distribution of fluoranthene between all compartments in Domaniów Lake

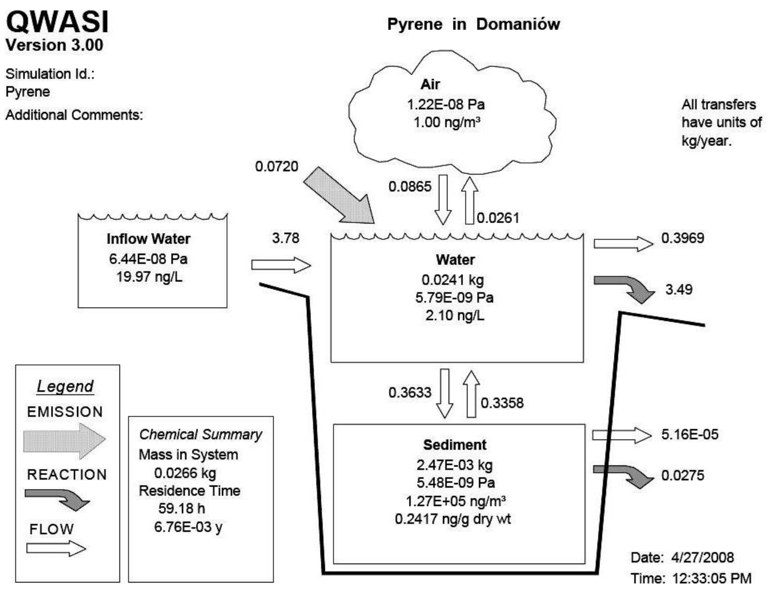

Fig. 10. Distribution of pyrene between all compartments in Domaniów Lake

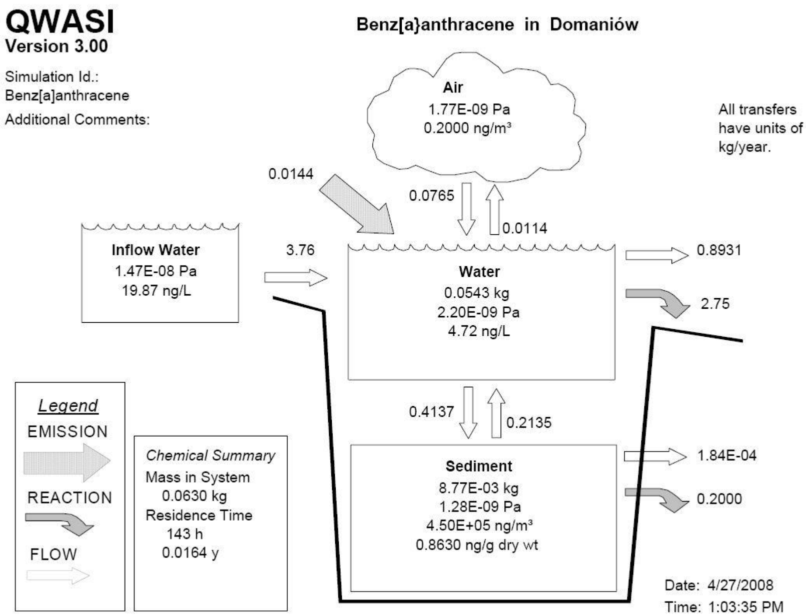

Fig. 11. Distribution of benz[a]anthracene between all compartments in Domaniów Lake

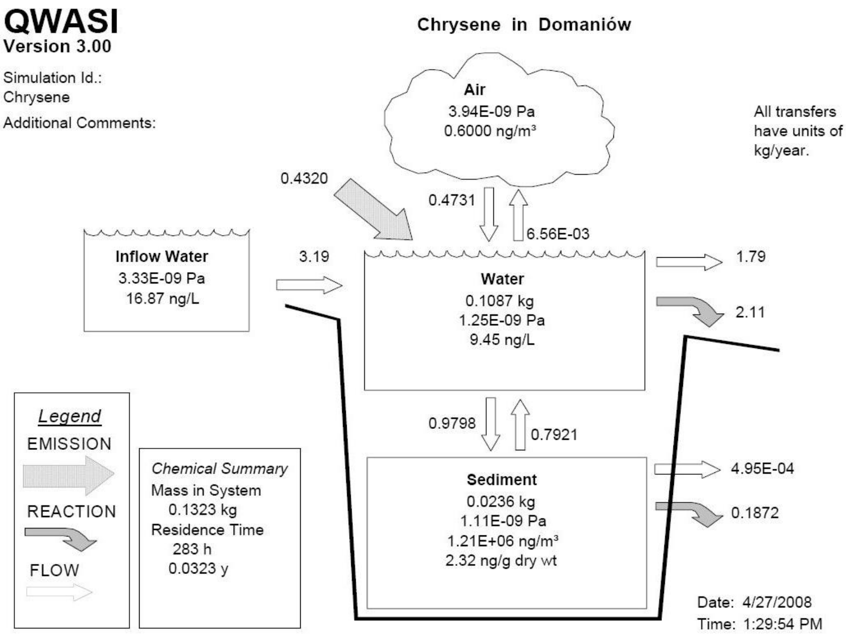

Fig. 12. Distribution of chrysene between all compartments in Domaniów Lake

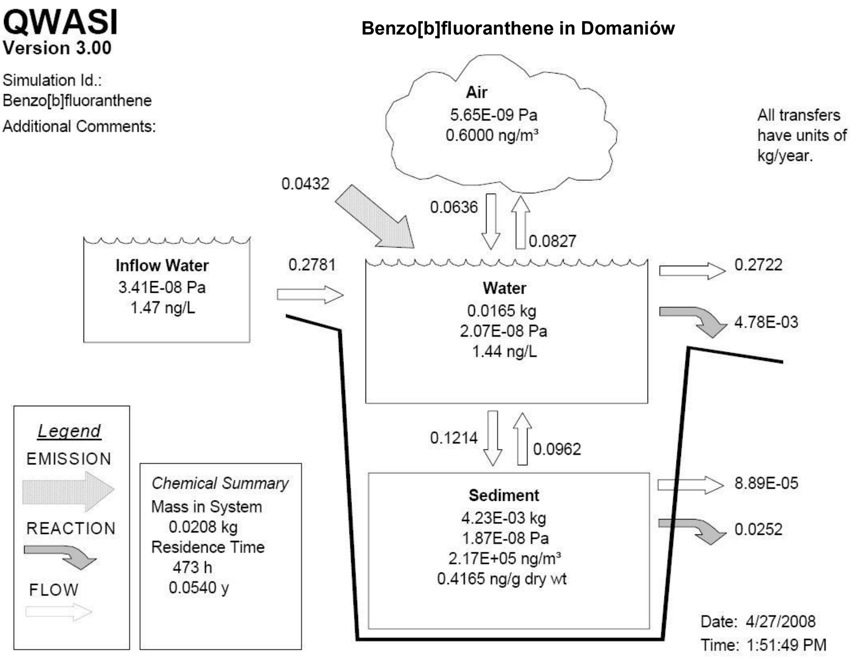

Fig. 13. Distribution of benzo[b]fluoranthene between all compartments in Domaniów Lake

The used model [25] quite well predicts the PAHs concentrations in the outflow river. The absolute deviations do not exceed 1 ng/L. The higher percentage deviations (22–24%) obtained for some PAHs are connected with their small content in the outflow (e.g. absolute deviation equal to 0.4 ng/L for anthracene corresponds to its percentage deviation equal to 22.25%).

The detailed calculations (the QWASI III model [25]) of distribution of the investigated components between all environmental compartments (air, water and sediment), describing amounts of all flows taken into account (Fig. 5) are shown in Figs. 6–13. The results of calculations agree with the global results presented by the model and allow detailed analysis of the PAHs distribution in the all investigated compartments in respect of their influence on the natural environment. In the case of Domaniów lake, the concentrations of all investigated PAHs both in air, water and sediment are very low, what means that in this region (central Poland) the pollution caused by PAHs is very small.

The results of prediction show good applicability of the model [25] for this type of calculations and prove that having information on the inflow of water, amounts of introduced chemicals and their thermodynamic properties we are able to predict properly the chemicals distribution in outflowing water.

5. CONCLUSIONS

The performed wind rose analysis showed that West and South-West are the predominant directions of wind both in the town of Radom as well in the central part of Poland. Independently of a year studied, these directions significantly dominate in the wind rose introducing polynuclear aromatic hydrocarbons (PAHs), originating from coal power stations, in the atmosphere in this part of Poland.

The measured concentrations of the all investigated PAHs are very low. This means that the influence of the coal power stations on the air pollution in the central Poland is very small and has no impact on the air pollution in other Baltic countries.

The PAH's concentrations in the investigated region calculated by the fugacity model agreed very well with the experimental data which proved the correctness of the model. The detailed results of calculations describing distribution of the all investigated PAHs between air, water and sediment, confirm the experimental results concerning very small PAH's pollution in the central Poland.

The results confirm usefulness of the fugacity model under consideration for prediction and monitoring of the fate of PAH's in the environment.

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