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Modelling of gas–particle partitioning of PAHs according to ab/adsorption approach

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Abstract: The new approach of the study was to assess the consistency between the gas–particle partition coefficients of 16 EPA (Environmental Protection Agency) polycyclic aromatic hydrocarbons (PAHs) predicted by the Dachs–Eisenreich ab/adsorption model and the experimental results obtained within the field measurements. A total number of 29 air samples was obtained at 9 locations in Serbia. High volume air sampler was applied, with quartz filters for collecting the atmospheric particles and polyurethane foam filters (PUF) for retaining the free gas molecules of PAHs. The results predicted by the model and the experimental data were compared. The deviations between the measured and predicted *f* (fraction) values were less than one order of magnitude for Flo, Phe, Ant, Flu, Pyr, B(a)A and Chr. For the PAHs with high molecular mass, B(b)F, B(k)F, B(a)P, I(1,2,3-cd)P, D(ah)A and B(ghi)P, very good agreement was confirmed, except for the data measured at the Oil refinery in Pančevo. The applied model underestimated the concentrations of PAHs in gas-phase for the low-molecular mass PAHs.

Keywords: Dachs–Eisenreich model; high volume sampling; air pollution.

INTRODUCTION

Gas/particle partitioning is a complex and sophisticated mechanism affecting the fate, transmission and transport of semivolatile organic compounds (SOCs), such as PAHs.¹ It gets the modelling of atmospheric distribution into the focus of recent investigations. During the previous decades, numerous methods have been suggested for modelling the sorption processes, ad/absorption and gas–particle

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partitioning of SOC.^{1–5} The atmosphere is the dominant medium for the transport and transformation processes of generated PAHs. In the atmosphere, PAHs can be present as free gas molecules (lower molecular mass PAHs) or can be adsorbed onto/into aerosol particles (higher molecular mass PAHs) and transmitted over the long distances.⁶

US EPA classified 16 PAHs (naphthalene, Nap; acenaphthylene, Acy; acenaphthene, Ace; fluorene, Flo; phenanthrene, Phe; pyrene, Pyr; fluoranthene, Flu; anthracene, Ant; chrysene, Chr; benz[*a*]anthracene, B(a)A; benzo[*b*]fluoranthene, B(b)F; benzo[*k*]fluoranthene, B(k)F; benzo[*a*]pyrene, B(a)P; dibenz[*a,h*]anthracene, D(ah)A; benzo[*ghi*]perylene, B(ghi)P; indeno[1,2,3-*c,d*]pyrene, I(123cd)P) as priority pollutants, due to their high frequency of detection in soil, air and water samples.⁵ The selected 16 PAHs can be classified according to the value of vapour pressure and molar mass in three groups: PAHs as free gas molecules (Nap, Acy, Ace, Flo, Phe, Ant), PAHs distributed between gas and particle phase (Chr, B(a)A, Pyr, Flu) and PAHs associated with particle phase (B(b)F, B(k)F, B(a)P, D(ah)A, B(ghi)P, I(123cd)P).⁵

The mostly investigated pathological effect of PAHs is their high toxicity and the most hazardous compound of the group is cancerogenic, teratogenic and genotoxic B(a)P. The main source of atmospheric PAHs is the incomplete combustion of organic matter. Soot particles are byproducts of the liquid and gaseous fuels combustion and PAHs perform an important role in soot formation and particle growth.⁷ PAHs have a significant affinity for carbonaceous materials, so the adsorption of PAHs onto the soot fraction of atmospheric aerosols, or primary aerosol carbon with which it is highly correlated, may be a dominant mechanism affecting the gas-particle partitioning of PAHs.¹

The main purpose of our study was to assess the consistency between the gas-particle partition coefficients of 16 EPA priority PAHs predicted by the Dachs–Eisenreich adsorption model and the experimental results gained within the field measurements.

EXPERIMENTAL

Air samples (gas and particulate) were taken in the urban and industrial area of the three cities in Serbia (Novi Sad, Pančevo, Kragujevac).

The three sampling sites in the city of Novi Sad were: N1, Oil Refinery Novi Sad (N 45° 16' 23,3"; E 19° 52' 12,1"), N2, Šangaj settlement, near the oil refinery (N 45° 16' 22,7"; E 19° 52' 24,1") and N3, city centre (N 45° 14' 54,3"; E 19° 50' 42,9"). The three sampling sites in the city of Pančevo were: P1, Oil refinery Pančevo (N 44° 49' 56,3"; E 20° 41' 25,4"), P2, Chemical Industry Pančevo (N 44° 49' 57,0"; E 20° 40' 17,0") and P3, city centre (N 44° 52' 12,8"; E 20° 38' 24,1"). In the city of Kragujevac, two samples were collected in the "Zastava" car factory: K1 (N 44° 0' 10,5"; E 20° 54' 46,6") and K2 (N 44° 0' 13,7"; E 20° 54' 45,9"). The third sample, K3 was collected in the city centre (N 44° 1' 4,2" E 20° 54' 45,9").⁵

Three air samples (gaseous and particle phase) were collected at sampling sites N1, N2, N3, P2, K1, K2 and K3 and four samples at the sites P1 and P3, therefore the total number of 29 air samples was obtained from 9 representative monitoring stations.

High volume air sampler, HVAS, was used, with quartz filters for collecting the atmospheric particles (GF) and polyurethane foam filters (PUF) for collecting the free gas molecules. The samples were analyzed using GC-MS instrument (HP 6890 – HP 5972) supplied with a J&W Scientific fused silica column DB-5MS. All analytical procedures were done in the laboratories of the Research Centre for Environmental Chemistry and Ecotoxicology (RECETOX), Masaryk University, Brno, Czech Republic.

The gas-particle partition coefficient K_p and particle bound fraction ϕ

The gas-particle partitioning could be quantified using partitioning coefficient, K_p , or particle-bound fraction, ϕ . Fraction is defined as a ratio of compound concentration associated with the particle phase ($F / \text{ng m}^{-3}$) over the sum of compound gaseous phase concentrations ($A / \text{ng m}^{-3}$) and particle phase concentrations:⁸

$$\phi = \frac{F}{A + F} = \frac{K_p c_{\text{TSP}}}{1 + K_p c_{\text{TSP}}} \quad (1)$$

Coefficient K_p is a ratio of F/c_{TSP} over A , where c_{TSP} is the concentration of total suspended particles (TSP) in the air ($\mu\text{g m}^{-3}$):⁸

$$K_p = \frac{F / c_{\text{TSP}}}{A} \quad (2)$$

Dachs-Eisenreich sorption model

The Dachs-Eisenreich sorption model, unlike other previously widely discussed adsorption and absorption models,¹ considers the partitioning of pollutants into two aerosol components: absorption into organic matter (OM) and adsorption to aerosol elemental carbon (EC).¹

$$K_p = 10^{-12} [(f_{\text{OM}} M_{\text{oct}} \gamma_{\text{oct}} / \rho_{\text{oct}} M_{\text{OM}} \gamma_{\text{OM}}) K_{\text{OA}} + f_{\text{EC}} (a_{\text{EC}} / a_{\text{SC}}) K_{\text{SA}}] \quad (3)$$

where: f_{OM} and f_{EC} are fractions of organic matter and elemental carbon on the TSP, respectively, K_{OA} is octanol/air partition coefficient, K_{SA} is soot/air partition coefficient, L kg^{-1} , M_{oct} is molecular weight of the octanol, $130.23 \text{ g mol}^{-1}$, M_{OM} is molecular weight of the organic matter phase, g mol^{-1} , ρ_{oct} is density of octanol, 0.82 kg L^{-1} , 10^{-12} comes from: $10^{-9} \text{ kg } \mu\text{g}^{-1} \times 10^{-3} \text{ m}^3 \text{ L}^{-1}$, units conversion factor, a_{EC} and a_{SC} are specific surface areas ($\text{m}^2 \text{ g}^{-1}$) of the aerosol elemental carbon and the soot carbon, respectively, used to measure K_{SA} (the D-E model¹ assumes this ratio is equal to 1).

The model further presumes that $M_{\text{oct}}/M_{\text{OM}}$ and $\gamma_{\text{oct}}/\gamma_{\text{OM}}$ are equal to 1. The model assumes that well-known octanol and elemental carbon are good surrogates of organic matter and soot, respectively.

If the partition is calculated only with the first part of the equation, K_p values would be underpredicted by 10 to 50 times. It is in accordance with the known fact that the majority of PAHs family members express a greater tendency to sorb onto the soot phase than into the organic matter per mass of sorption phase.¹

RESULTS AND DISCUSSION

TSP, EC and atmospheric PAHs concentrations

The concentrations of TSP and EC applied in the modelling process are given in Table I.

TABLE I. TSP and EC concentrations

Parameter	P1-P3	N1	N2	N3	K1-K3 ^a	S1-S4 ^a	T1-T5 ^a
$c_{\text{TSP}} / \mu\text{g m}^{-3}$	114 (47–173) ^b	187 (116–267) ^b	187 (116–267) ^b	205 (124–256) ^b	100	100	100
$c_{\text{EC}} / \mu\text{g m}^{-3}$	3.4 (8–13) ^b	5 (1–13) ^b	5 (1–13) ^b	4 (1–12) ^b	–	–	–
f_{EC}	0.0298	0.0267	0.0267	0.0195	0.05	0.05	0.05

^aLiterature data; ^brange of concentrations

In the vicinity of the Pančevo centre, TSP concentrations ranged from 47 to 173 $\mu\text{g m}^{-3}$, with the average value of 114 $\mu\text{g m}^{-3}$. In Šangaj settlement, Novi Sad, TSP concentrations ranged from 116 to 267 $\mu\text{g m}^{-3}$ (average 187 $\mu\text{g m}^{-3}$) and in the Novi Sad centre, from 124 to 256 $\mu\text{g m}^{-3}$ (average 205 $\mu\text{g m}^{-3}$). The TSP concentration values measured at P3 were also applied for the modelling at the P1 and P2 locations. In the city of Pančevo, the EC concentrations ranged from 8 to 13 $\mu\text{g m}^{-3}$ (average 3.4 $\mu\text{g m}^{-3}$; $f_{\text{EC}} = 0.0298$). In Šangaj settlement, the EC levels ranged from 1 to 13 $\mu\text{g m}^{-3}$ (average 5 $\mu\text{g m}^{-3}$; $f_{\text{EC}} = 0.0267$) and, in the Novi Sad centre, from 1 to 12 $\mu\text{g m}^{-3}$ (average 4 $\mu\text{g m}^{-3}$; $f_{\text{EC}} = 0.0195$). At the location P3 the TSP levels were lower than in Novi Sad, but the fraction of elemental carbon (soot phase), f_{EC} , was higher. There were no measured data for TSP, EC and f_{EC} for the cities of Kragujevac, Sarajevo and Tuzla, therefore the literature values were used. The TSP concentrations for different spatial conditions (suburban: 60 $\mu\text{g m}^{-3}$, urban: 100 $\mu\text{g m}^{-3}$) are given by Whitby.⁹ The mass fraction of elemental carbon f_{EC} in aerosol particles for urban conditions ($f_{\text{EC}} = m_{\text{EC}}/m_{\text{TSP}} = 0.05$) is published by Jonkers and Koelmans,¹ Seinfeld and Pandis,⁷ Vardar *et al.*¹⁰ and Dachs and Eisenreich.¹¹

The measured atmospheric concentrations of gas and particle phase of PAHs (ng m^{-3}) at locations in Serbia are shown on Fig. 1.

Fig. 1 displays that gas molecule concentrations of lower molecular mass PAHs (Phe, Flu and Pyr) were about two to four times higher at Oil Refinery Pančevo and Chemical Industry Pančevo comparing to urban locations in Novi Sad. Gas concentrations at Oil Refinery Novi Sad are higher than in the city, but still lower than at Oil Refinery Pančevo and Chemical Industry Pančevo. The variability in the aerosol and gas molecule concentrations of PAHs is related to wind direction and EC concentrations in the aerosols.¹

The most notably variations of measured ϕ fractions are recorded for the group of PAHs presented in both, gas and particle phase (B(a)A, Chr), Fig. 2.

The particle bound fractions of 16 EPA PAHs (in percentage) were calculated for all nine locations in Serbia using the Eq. (1) and it is shown later in Figs. 3–7 as the “measured value”, when measured and predicted values are compared.

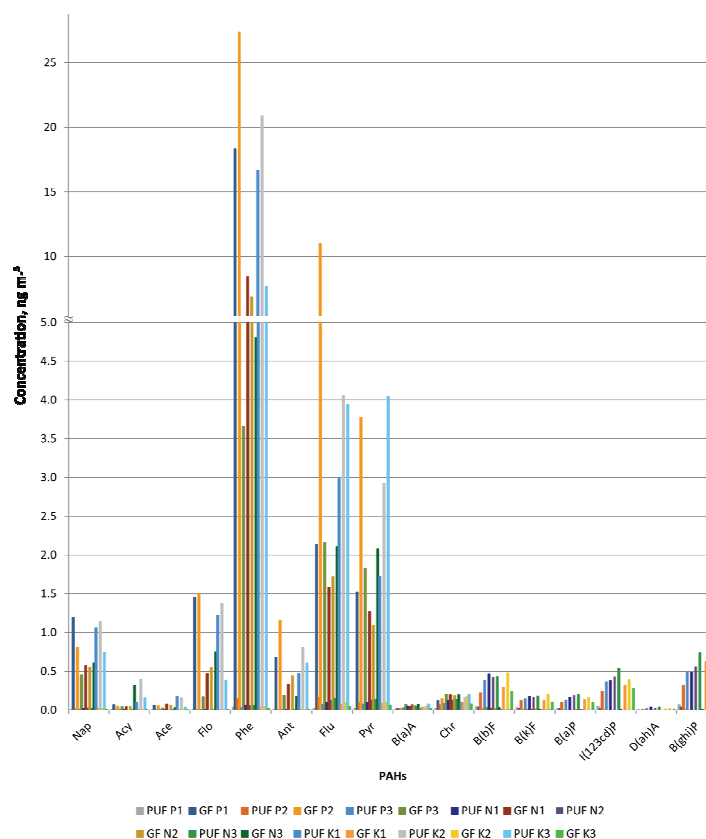


Fig. 1. Median values of measured atmospheric concentrations of PAHs gas (PUF) and particle phase (GF) at locations in Serbia.

Dachs–Eisenreich model

In order to perform the modelling process, values for the vapour pressure p_L^0 and $\log K_{OA}$ were obtained from Mackay *et al.*,¹² and the values for $\log K_{SA}$ were calculated using the Eq. (4) except for the values for Flo, Phe, Ant, Flu, Pyr, Chr and B(a)P, which were taken from the Lohmann and Lammel:⁴

$$K_{SA} = -0.85 \log p_L^0 + 8.94 - \log \left(\frac{998}{a_{EC}} \right) \quad (4)$$

The data were not available for the organic matter fraction f_{OM} , the K_p and ϕ values, so they were calculated using $f_{OM} = 0.40$ for urban conditions and $f_{OM} = 0.25$ for suburban conditions, recommended by Seinfeld and Pandis,⁷ and also using $f_{OM} = 0.20$, proposed by Jonkers and Koelmans.¹¹ Values from Table I for TSP and f_{EC} were used to calculate the partition coefficient K_p by the Dachs–Eisenreich Equation:¹

$$K_p = \frac{f_{OM}}{10^{12} \rho_{oct}} K_{OA} + \frac{f_{EC}}{10^{12}} K_{SA} \quad (5)$$

and the particle bound fraction ϕ was calculated using Eq. (1).

The values for vapour pressure, $\log K_{OA}$ and $\log K_{SA}$ of 16 EPA priority PAHs are presented in Table II.

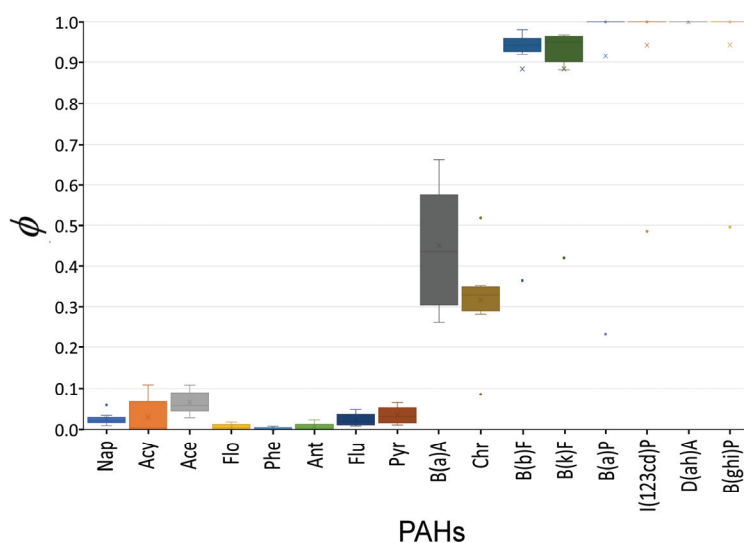


Fig. 2. Variations of ϕ values at the locations in Serbia.

TABLE II. Vapour pressure, $\log K_{OA}$ and $\log K_{SA}$ for 16 EPA priority PAHs used in Dachs–Eisenreich model

PAH	p_L^0 / Pa	$\log K_{OA}$	$\log (K_{SA} / \text{L kg}^{-1})$
Nap	1.00	5.13	6.89
Acy	0.90	6.23	7.78
Ace	0.30	6.22	8.18
Flo	0.50	6.68	8.60
Phe	0.08	7.45	9.40
Ant	0.07	7.34	9.50
Flu	6×10^{-3}	8.60	10.50
Pyr	5×10^{-4}	8.61	10.60
B(a)A	5×10^{-4}	9.54	10.54
Chr	1.7×10^{-4}	10.44	12.10
B(b)F	10^{-6}	10.98	12.84
B(k)F	10^{-6}	11.19	12.84
B(a)P	3×10^{-6}	10.77	13.00
I(123cd)P	10^{-9}	11.56	15.39
D(ah)A	10^{-8}	13.90	14.54
B(ghi)P	10^{-8}	11.01	14.54

Modelling gas-particle partition of PAHs

The discrepancy between the measured and the predicted values for higher molecular mass PAHs (B(b)F, B(k)F, B(a)P, I(1,2,3 cd)P, D(ah)A, B(ghi)P) is negligible for all sampling locations in Serbia, except for Oil Refinery Pančevo (P1), where the measured values for ϕ were 30–50 % lower than predicted (Fig. 3). The reason is higher rate of PAHs emission in the atmosphere at the location Oil Refinery Pančevo than the rate of sorption onto the soot in the air, causing the specific gas-particle partition comparing to urban conditions.

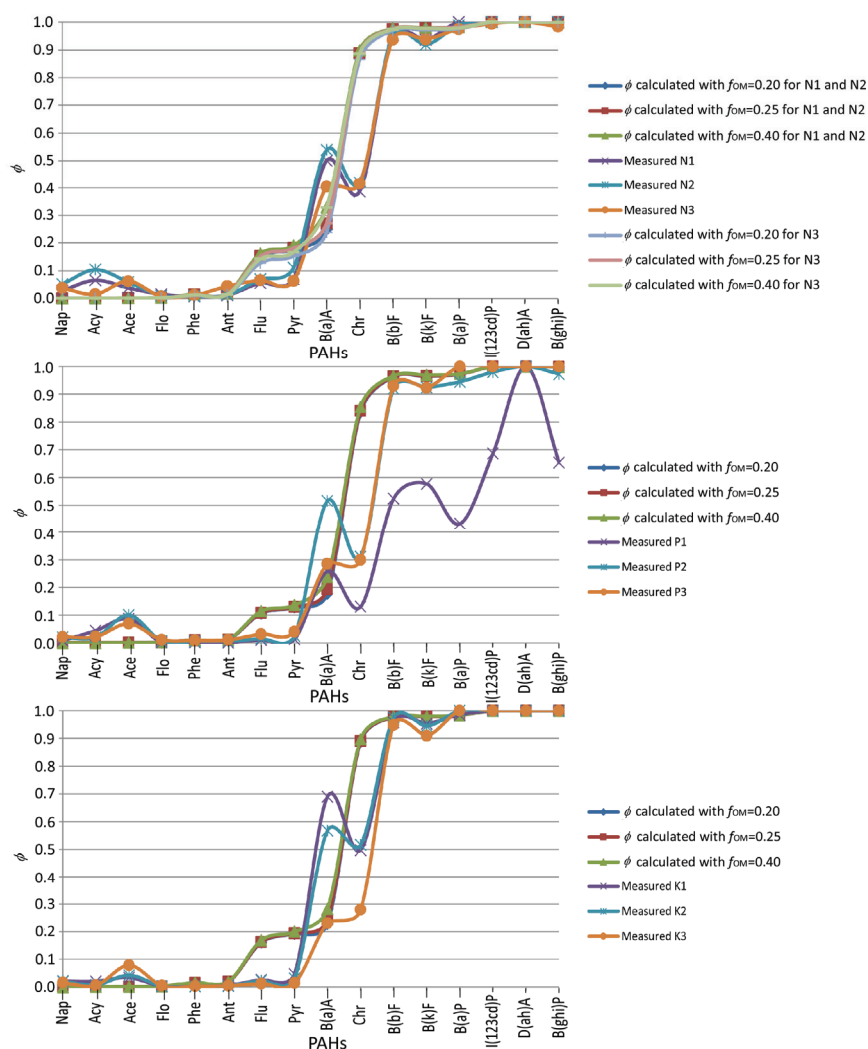


Fig. 3. Comparison of measured and predicted ϕ values at all locations using Dachs–Eisenreich model.

Other reasons for the presented discrepancy could be:

- the variations in the chemical contents of the atmospheric particles which were not adequately presented by the uniform values for f_{OM} and f_{EC} ;
- as indicated by Dachs and Eisenreich,¹ the temperature exerts a strong influence on the values of K_{OA} and K_{SA} ; the coefficient K_{SA} depends on the dimensionless Henry's law constant H and its value varies at different temperatures; the temperature influence on K_{OA} is defined by $\log K_{OA} = A + B/T$, whereas A and B are coefficients estimated by Harner and Bidleman by measurements of K_{OA} at different temperatures – T / K ,³ but in our prediction, it was assumed that the temperature was 25 °C; the prediction should be more precise if the temperature influence was taken into account;
- since the liquid film of organic matter covers the elemental carbon, the reversible processes of SOCs ad/desorption onto and from the atmospheric aerosols are very slow and the reaching of equilibrium between gas and particle phase is delayed, the gaps between measured and real concentrations can be potentially caused;¹
- according to the specific source of PAHs emissions at the Oil Refinery Pančevo (P1), the modelled values could be improved using the measured values for TSP and f_{EC} on the site;
- breakthrough of volatile PAHs from the PUF can lead to underestimation of the concentrations of gas-phase PAHs.

Three different scenarios for the f_{OM} value gave approximately identical levels of PAHs particle fraction ϕ , which means that the process of absorption doesn't affect so much the overall partition of PAHs in the atmosphere. The research has shown that the Dachs and Eisenreich model provided the good prediction for urban locations, but underestimated the values in rural areas for one order of magnitude.^{10,13}

To compare the results, the ratio of measured and modelled values is shown on logarithmic scale (for selected locations N1 and P1, Fig. 4). It is obvious that the Dachs and Eisenreich model points out the good predictions of gas-particle partition for higher molecular mass PAHs considering all three scenarios. As well, the predicted values obtained by Dachs and Eisenreich model for lower molecular mass PAHs are slightly closer to the measured ones, comparing to the other previously wide used mathematical models.^{14–16} Nevertheless, the model needs the correction, at least for the Nap, Acy, Ace and Flo.

The differences between measured and modelled values for three cities are presented using QGIS software and shown on Figs. 5–7. The difference is positive if the measured value is higher than the modelled one, and *vice versa*. Only the ϕ for Chr and B(a)A are pictured on the maps because these compounds showed the highest gaps between the measured and the modelled values.

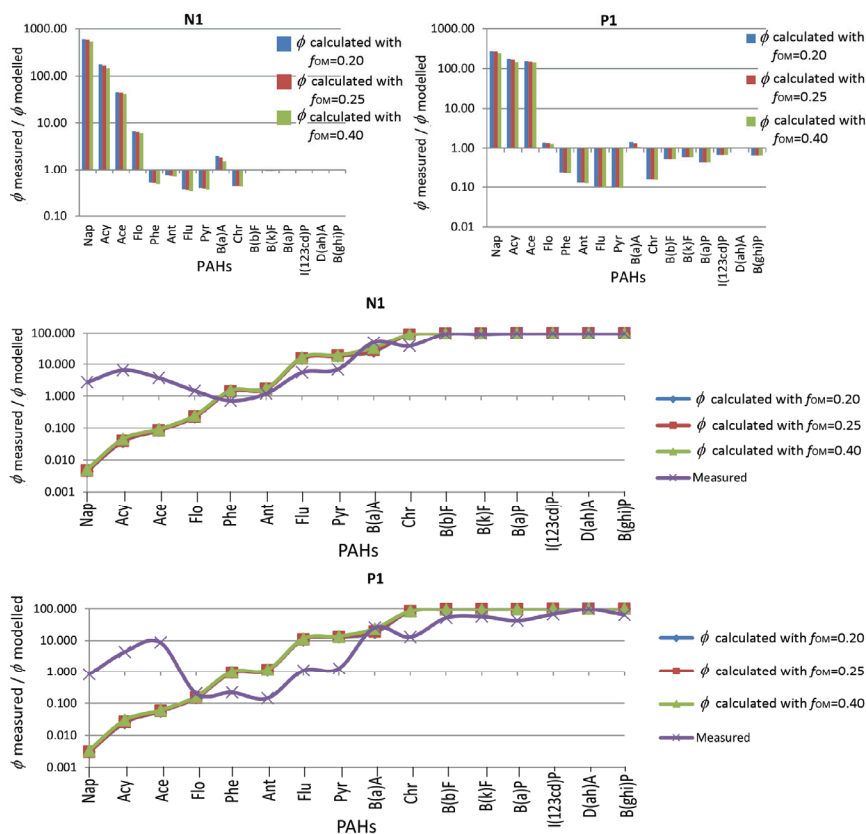


Fig. 4. Measured and modelled ϕ values at locations N1 and P1.

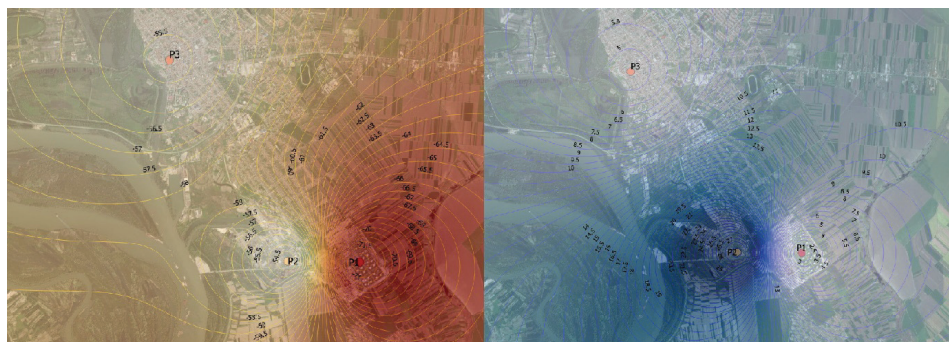


Fig. 5. The differences between measured and modelled ϕ values of Chr (a) and B(a)A (b) for the city of Pančevo, Serbia.

The ϕ for Chr in Pančevo is significantly overestimated using the Dachs and Eisenreich model at the locality P1 and it is obvious that the model needs to be corrected. Particle bound fraction for Chr was also overestimated at the P2 and P3

locations, but not in such high extent. While the partition of B[a]A was acceptable, as predicted by the model at P1 and P3, it was underestimated at the locality Petrochemical Industry Pančevo.



Fig. 6. The differences between measured and modelled fractions ϕ of Chr and B(a)A for the city of Novi Sad, Serbia.

The results were similar at the Oil Refinery Novi Sad. The model, once again, overestimated the particle fraction of Chr and underestimated the particle fraction of B(a)A, although the differences between the Oil Refinery and the city centre were minor.

For the city of Kragujevac (Fig. 7), the model overestimated Chr particle fraction for the city centre (60 %) and for the car factory (40 %). B(a)A particle fraction was modelled accurately for the city centre (K3), but again underestimated in the industrial area.

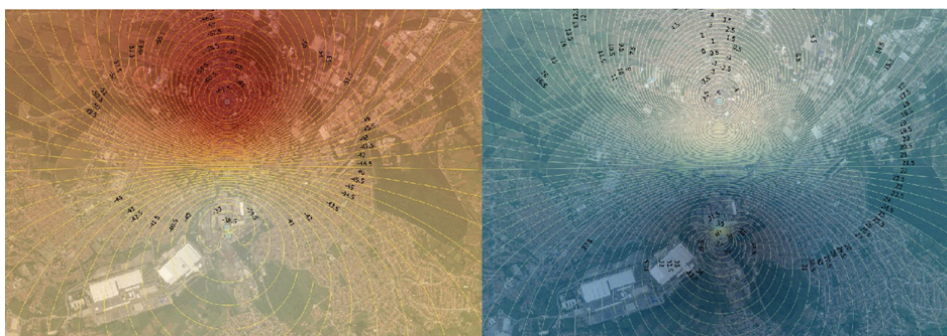


Fig. 7. The differences between measured and modelled fractions ϕ of Chr and B(a)A for the city of Kragujevac, Serbia.

CONCLUSION

The results on modelling of the PAHs atmospheric distribution using the Dachs–Eisenreich ab/adsorption model displays gaps between the measured and predicted ϕ values less than one order of magnitude for Flo, Phe, Ant, Flu, Pyr,

B(a)A and Chr. For the high molecular mass PAHs, namely B(b)F, B(k)F, B(a)P, I(1,2,3-cd)P, D(ah)A and B(ghi)P, very good correlation was confirmed.

Similar variability of the measured/modelled ϕ values was obtained using the previously discussed theoretical and empirical, absorption and adsorption models, indicating the presence of non-exchangeable, inert fraction of PAHs in the suspended particles, which cannot be exchanged by the air. One of the possible reasons for the discrepancy between the measured and modelled ϕ values could be the assumed constant values of f_{OM} and f_{EC} . For each specific locality, the influence of the local ambient conditions on both physical and chemical structure, concentration and dynamic of suspended particles formation should be considered.

For the PAHs with lower molecular mass, all models have significantly underestimated the concentrations of PAHs as the free gas molecules, but applying the Dachs and Eisenreich approach, the modelled values are slightly closer to the measured ones. For the second group of PAHs, in both the gas and particle phase, the variations between the measured and the predicted values are recorded using all models. For the third group of PAHs, the Dachs and Eisenreich model has predicted the partition processes precisely.

The application of the Dachs–Eisenreich sorption approach was examined and demonstrated and resulted in the modified regional-scale gas/particle atmospheric modelling. Our results can provide ground for the new research area development including the gas/particle spatial-temporal variations of ϕ values for the variety of atmospheric pollutants distribution balance models, without the need for extensive additional field data collection.

ИЗВОД

МОДЕЛОВАЊЕ ГАС-ЧЕСТИЧНЕ РАСПОДЕЛЕ ПОЛИАРОМАТИЧНИХ УГЛОВОДОНИКА ПРИМЕНОМ АПСОРПЦИОНО/АДСОРПЦИОНОГ МОДЕЛА

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Мотив истраживања био је да се процени конзистентност између гас-честичне расподеле 16 ЕРА полиароматичних угљоводоника предиктоване апсорпционо/адсорпциононим Dachs–Eisenreich моделом и резултата добијених експерименталним теренским мерењима. На 9 репрезентативних локација у Србији прикупљено је укупно 29 узорака ваздуха. Примењена је експериментална метода прикупљања узорака ваздуха узorkивачима велике запремине (high volume air sampler, HVAS), са кварцним филтерима за атмосферске суспендоване честице и филтерима од полиуретанске пене (PUF) за слободне гасне молекуле полиароматичних угљоводоника. Упореджени су резултати добијени Dachs–Eisenreich моделом и подаци добијени теренским мерењима. Одступања између измерених и вредности модела била су мања од једног реда величине за Flo, Phe, Ant, Flu, Pir, B (a) A и Chr. За полиароматичне угљоводонике велике молекулске масе, B (b) F, B (k) F, B (a) P, I (1,2,3-cd) P, D (ah) A и B (ghi) P, потврђена је веома добра

корелација, осим код података са територије Рафинерије нафте у Панчеву. Примењени модел је показао знатно ниже вредности концентрација за слободне гасне молекуле полиароматичних угљоводоника мале молекулске масе.

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