

GAS PARTICLE PARTITIONING OF PERSISTENT ORGANIC POLLUTANTS IN A PILOT AREA OF THE CITY OF KRAGUJEVAC, SERBIA

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ABSTRACT

Destruction of the industrial targets, accompanied by explosions and combustion during the NATO operation in Serbia (March-June 1999), caused contamination by persistent organic pollutants of the atmosphere, surface and ground water, sediment, soil and biota. Air sampling campaign from June/July 2004 was conducted using active sampling method and gas/particle partitioning of sixteen priority PAHs and seven PCB congeners were analyzed. The standard method for air sampling of nonpolar aromatic compounds like polychlorinated biphenyls and polycyclic aromatic hydrocarbons is active sampling method using high or medium volume samplers (HiVols/MeVols) containing a glass fiber filter and polyurethane foam filters or XAD-2. Samples were collected once a day for the period of three days, simultaneously using three HiVol samplers at the three selected localities in the city of Kragujevac (two samplers have been placed at the industrial area and the third sampler was used as background sampler located in the city centre. Gas chromatography coupled with mass spectrometry analysis was carried out in laboratories of Research Centre for Environmental Chemistry and Ecotoxicology (RECETOX), Masaryk University in Brno, Czech Republic. The relatively high concentration of PCBs was measured on the locality inside the Zastava Car Factory (industrial locality), in vicinity of the power generation building and two large transformers. Some minor variations in concentration levels of PAHs were detected, but still the highest concentration was noticed at the main production building, inside the car factory. From the results on concentration levels of the PCB congeners in the air samples, from gaseous and particulate phase, it is evident that the most of the investigated congeners existed in the gaseous phase. Analysis of the gas and aerosol partitioning of polycyclic aromatic hydrocarbons in Kragujevac emphasized the presence of PAHs in both, gaseous and particulate phase. Gas particle partitioning of polycyclic aromatic hydrocarbons showed uniform distribution at the investigated locations.

Keywords: Gas particle partitioning, Persistent organic pollutants, Polychlorinated biphenyls, Polycyclic aromatic hydrocarbons

INTRODUCTION

Persistent organic pollutants (POPs) are a class of organochlorine chemicals characterized by long-range transport and processes of bioaccumulation and biomagnification in humans and animal fat tissues, and by high resistance to environmental degradation processes. POPs are toxic organic compounds of natural or anthropogenic origin and include substances such as polycyclic aromatic hydrocarbons (PAHs), dioxins, furans, polychlorinated biphenyls (PCBs) and organochlorine pesticides (HCH, DDT, HCB, etc). Being capable of long-range transport, POPs released to the

environment could be transported through air and water, due to advection and diffusion processes, to regions far from their original sources according to concentration, temperature and pressure gradient. In these remote regions, POPs are accumulated in living organisms, including humans, to levels that can injure human health and the environment. The removal of POPs from the atmosphere mostly depends on the gas particle phase partitioning, since the deposition process of gases is different than for particles, and it is obvious that the environmental fate of POPs in the environment is determined by the partition of the compound between different media, such as air, water and soils, or between different phases of the same media. Distribution of the released pollutant between various environmental compartments depends on its numerous physical-chemical properties.

During the conflict situation in Serbia (especially from 24th March – 11th June 1999), a huge amount of PAHs, PCBs, dioxins and furans was generated and released into the atmosphere, ground and surface water, sediment, soil and biota, as the result of the destruction of industrial plants and installations, military targets and storage spaces, filled with toxic chemicals and by combustion caused by explosions. After the air strikes, UNEP has defined four key hot-spots in Serbia: Bor, Novi Sad, Pancevo and Kragujevac. The air strikes in Kragujevac caused extensive environmental pollution, with damage to soil, water and air. The main environmental problem was the leaking of 2500 kg of PCBs (contained in transformer oil) into the Morava river and contamination of groundwater and soil (Kaisarevic 2007, UNEP UNCHS 1999).

In order to define the relevant phase into which chemicals primarily partition in air, the gas particle partitioning of sixteen US EPA priority PAHs and seven PCB congeners was investigated and analyzed. The obtained gas particle distribution was compared with the values of fraction (%) of a compound associated with particles estimated using Junge-Pankow adsorption model. The atmospheric levels of PAHs/PCBs used in the study were measured separately in the gas and particle phase of the atmosphere using active sampling method during the sampling campaign conducted in June 2004 at one of the hot-spots in Serbia, the city of Kragujevac, within the project Assessment of the selected POPs in the atmosphere and water ecosystems from the waste generated by warfare in the area of former Yugoslavia.

1 MATERIALS AND METHODS

1.1 Sampling locations

Air samples were collected during the 24 hours for the period of three days (June 23–26, 2004) simultaneously using three HiVol samplers at the three selected highlighted localities in Kragujevac. Two samplers have been placed at the industrial area, inside the Zastava Car Factory: one at the power generation building (locality K1) and the other one at the main production building (locality K2). The third sampler was used as a background sampler located at the University of Kragujevac, city centre, (locality K3). Taking into consideration the leaking of transformer oil during the air strikes and the permanent usage of the charcoal in the boilers, localities K1 and K2 are assumed to be the major source of the air pollution with PCB congeners and PAHs, while the motor vehicles at the locality K3 could be the source of polycyclic aromatic hydrocarbons emission.

1.2 Sampling methods

Ambient air sampling was conducted in accordance with the U.S. EPA methods for the determination of toxic organic compounds in ambient air (U.S. EPA 1999) using the active sampling method with high volume sampler (HiVols) containing a quartz fibre filter (which collects particle-bound compounds) and two polyurethane foam filters (which retain gaseous compounds).

1.3 Sample analysis

Polycyclic aromatic hydrocarbons and polychlorinated biphenyls were analyzed by the laboratories of the Research Centre for Environmental Chemistry and Ecotoxicology (RECETOX), Masaryk University, Brno, Czech Republic, using the gas chromatography-mass spectrometry method for determining PAHs and gas chromatography-electron capture detector for analyzing PCB congeners (APOPSBAL 2005, Klánová 2007).

2 RESULTS

2.1 Gas/particle partitioning

Partitioning of semi volatile organic compounds between the gas and particulate phases in the atmosphere controls their rate of removal by wet and dry deposition, their atmospheric degradation and their long-range transport (Mandalakis & Euripides 2007, Bidleman 1988, Scheringer 1997).

Fractions of polycyclic aromatic hydrocarbons and polychlorinated biphenyls adsorbed at the atmospheric particles, ϕ [%], at the localities K1, K2 and K3 are shown in Table 1.

Table 1: PAH/PCB fraction adsorbed at the atmospheric particles, ϕ [%], at the localities K1, K2 and K3

Locality Component	K1	K2	K3
Naphthalene	2.04	3.06	1.58
Acenaphthylene	2.00	0.76	0.64
Acenaphthene	3.37	4.29	7.89
Fluorene	0.24	0.29	0.52
Phenanthrene	0.22	0.22	0.31
Anthracene	0.64	0.37	0.49
Fluoranthene	2.61	2.36	1.23
Pyrene	4.69	3.12	1.46
Benz(a)anthracene	68.96	56.81	23.08
Chrysene	49.75	51.63	27.91
Benz(b)fluoranthene	95.79	97.55	94.84
Benz(k)fluoranthene	95.31	94.49	90.90
Benz(a)pyrene	98.52	100	100
Dibenz(a,h)anthracene	100	100	100
Indeno(123cd)pyrene	100	100	100
Benz(g,h,i)perylene	100	100	100
PCB 28	0.11	0.39	2.86
PCB 52	0.21	1.70	7.69
PCB 101	0.85	2.74	8.69
PCB 118	1.43	7.10	11.11
PCB 138	2.62	9.09	9.09
PCB 153	1.64	5.74	10
PCB 180	6.30	17.5	0

The most often used model for describing the gas/particle partitioning of the persistent organic pollutants was proposed by Junge-Pankow (Junge 1977, Pankow 1987). According to the Junge-Pankow model, the fraction of a compound associated with particles, ϕ , is calculated using vapor pressure of the sub cooled liquid at the ambient temperature, p_{OL} , available aerosol surface, θ , (m^2 particles per m^3 air) and parameter c (Pa m) that depends on the difference between the heat of desorption from the particle surface and the heat of vaporization of the compound and the surface properties of the suspended particles, $\phi=c\theta/(p_{OL}+c\theta)$ (Lammel et al. 2007, Mandalakis & Euripides 2007).

For the estimation of the fraction ϕ of the investigated compounds, the sub cooled liquid vapor pressure at the average temperature during the three days of 24 hours sampling periods was used. Calculations were made assuming $c=0.172$ Pa m (Junge 1977) and $\theta=1.1\times10^{-3} m^2 m^{-3}$, which are representative of aerosols from urban areas (Van Pul et al. 1998). Comparison of the observed and estimated PAHs/PCBs fractions adsorbed at the atmospheric particles is shown at the Fig. 1 and 2 (subcooled liquid vapour pressure for Indeno(123cd)pyrene is not available).

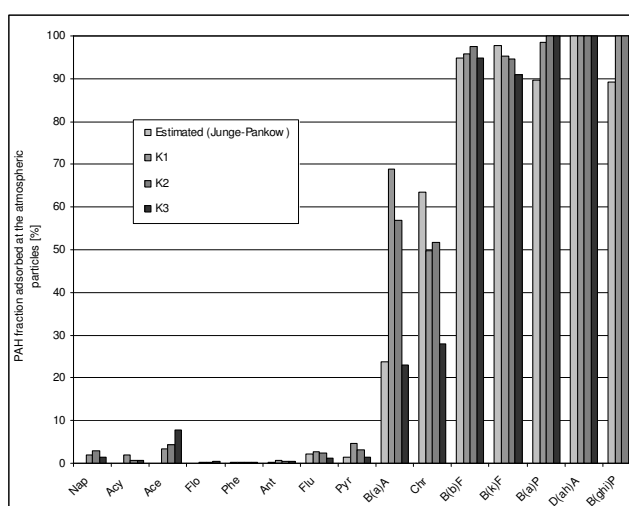


Fig. 1: Comparison of the estimated and calculated PAH fractions adsorbed at the atmospheric particles

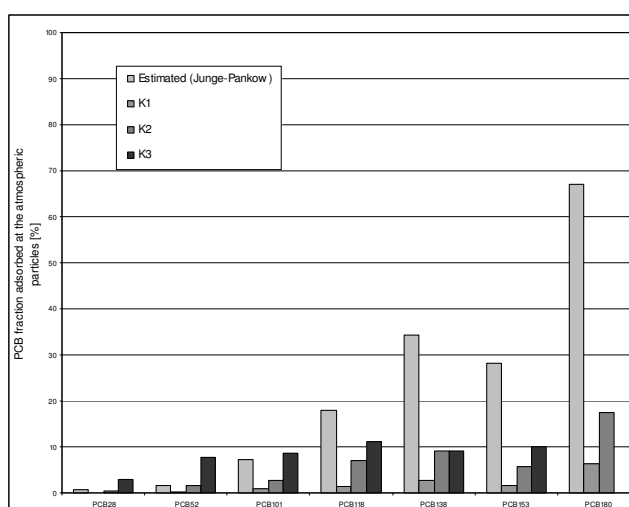


Fig. 2: Comparison of the estimated and calculated PCB fractions adsorbed at the atmospheric particles

Relative deviations of estimated ϕ values using Junge-Pankow approach to the calculated values obtained from the measured concentration levels of polycyclic aromatic hydrocarbons and polychlorinated biphenyls at the localities K1, K2 and K3 are shown in Table 2.

Table 2: Relative deviations of estimated ϕ values using Junge-Pankow approach to the calculated values at the localities K1, K2 and K3 [%]

Locality Component	K1	K2	K3
Naphthalene	99.97	99.98	99.97
Acenaphthylene	99.78	99.41	99.30
Acenaphthene	99.63	99.71	99.84
Fluorene	89.00	90.89	94.92
Phenanthrene	24.37	24.37	46.33
Anthracene	62.45	35.04	50.95
Fluoranthene	19.47	10.94	70.88
Pyrene	67.34	50.90	4.92
Benz(a)anthracene	65.66	58.31	2.61
Chrysene	27.81	23.15	127.82
Benz(b)fluoranthene	1.02	2.81	0.03
Benz(k)fluoranthene	2.68	3.57	7.66
Benz(a)pyrene	8.91	10.26	10.26
Dibenz(a,h)anthracene	0.05	0.05	0.05
Benz(g,h,i)perylene	10.69	10.69	10.69
PCB 28	581.25	92.15	73.80
PCB 52	655.47	6.68	79.37
PCB 101	759.37	166.59	15.94
PCB 118	1152.58	152.28	61.22
PCB 138	1212.02	278.16	278.16
PCB 153	1614.65	389.90	181.20
PCB 180	963.89	283.00	not defined

2.2 Ambient air PAH/PCB concentrations

PAH/PCB concentration levels in the atmosphere are presented on the basis of individual compounds. The average concentrations of all investigated PAHs and PCBs in the gaseous phase and the average total concentration (gaseous and particulate phase) of PAHs/PCBs for the field sites K1, K2 and K3 are presented in Table 3.

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Table 3: Average concentration levels of PAHs/PCBs (gaseous phase and gaseous + particulate phase / Σ G+P) at the field sites K1, K2 and K3 (ng/m³) (APOPSBAL 2005)

Locality	K1		K2		K3	
Type of area	Industrial		Industrial		Urban	
Phase Component	Gaseous phase	Σ G+P	Gaseous phase	Σ G+P	Gaseous phase	Σ G+P
Naphthalene	1.056	1.078	1.141	1.177	0.746	0.758
Acenaphthylene	0.098	0.100	0.394	0.397	0.154	0.155
Acenaphthene	0.172	0.178	0.156	0.163	0.035	0.038
Fluorene	1.220	1.223	1.374	1.378	0.385	0.387
Phenanthrene	16.664	16.701	20.886	20.933	7.665	7.689
Anthracene	0.469	0.472	0.809	0.812	0.608	0.611
Fluoranthene	2.984	3.064	4.053	4.151	3.943	3.992
Pyrene	1.727	1.812	2.923	3.017	4.045	4.105
Benz(a)anthracene	0.018	0.058	0.038	0.088	0.080	0.104
Chrysene	0.100	0.199	0.163	0.337	0.204	0.283
Benz(b)fluoranthene	0.013	0.309	0.012	0.489	0.013	0.252
Benz(k)fluoranthene	0.006	0.128	0.012	0.218	0.010	0.110
Benz(a)pyrene	0.002	0.135	0.000	0.158	0.000	0.097
Dibenz(a,h)anthracene	0.000	0.318	0.000	0.390	0.000	0.276
Indeno(123cd)pyrene	0.000	0.013	0.000	0.021	0.000	0.011
Benz(g,h,i)perylene	0.000	0.621	0.000	0.580	0.000	0.514
Total PAHs	24.529	26.409	31.961	34.309	17.888	19.382
PCB 28	1.812	1.814	1.008	1.013	0.034	0.035
PCB 52	11.141	11.164	0.577	0.587	0.024	0.026
PCB 101	11.424	11.522	0.495	0.510	0.021	0.023
PCB 118	6.816	6.915	0.170	0.183	0.008	0.009
PCB 138	5.096	5.233	0.190	0.209	0.010	0.011
PCB 153	5.028	5.112	0.230	0.244	0.009	0.010
PCB 180	0.639	0.682	0.032	0.040	0.001	0.001
Total PCBs	39.685	40.149	2.704	2.785	0.107	0.112

In order to investigate the presence of significant local source of pollution, relative contributions of the PAHs grouped according to number of rings in structure and relative contributions of individual PCB congeners were analyzed (Fig. 3 and 4)

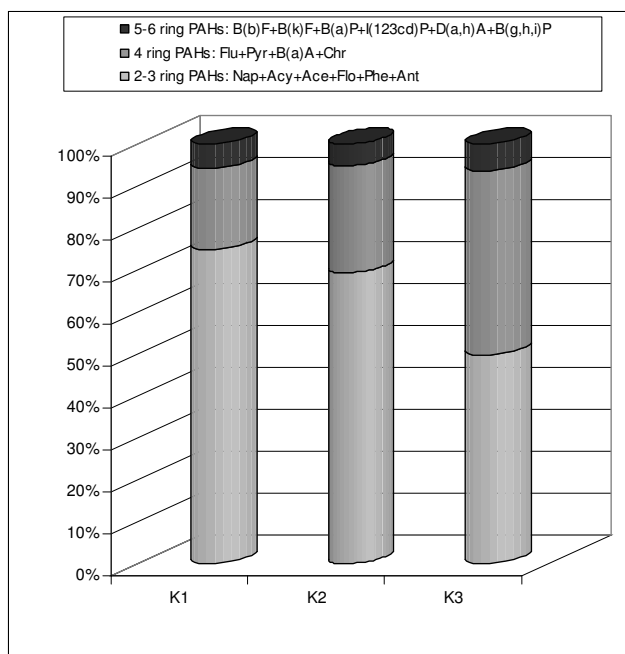


Fig. 3: Relative contributions of the PAHs (PUF+GF) grouped according to number of rings in structure in the ambient air (K1, K2 and K3)

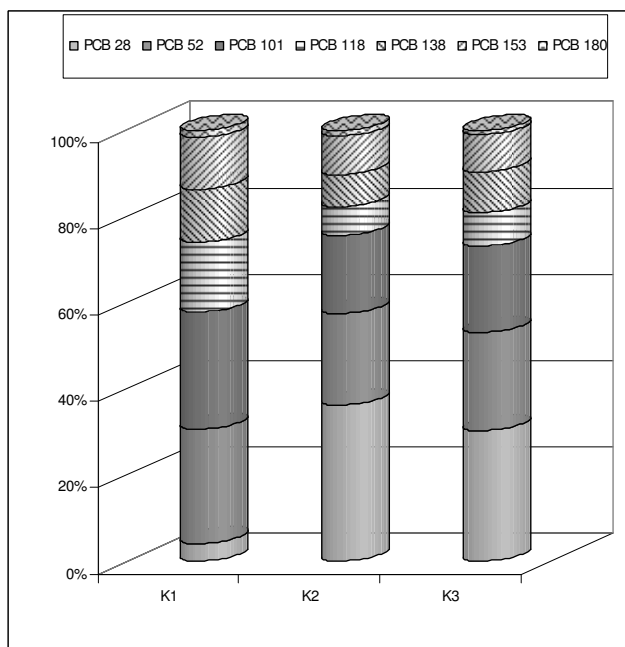


Fig. 4: Relative contributions of the individual PCBs (PUF+GF) in the ambient air (K1, K2 and K3)

3 DISCUSSION

PAHs in this study range from those which generally can only be detected in the vapour phase (such as 2 or 3-ring PAHs) to compounds which can be detected only in collected particles (5 and 6-ring PAHs). Gas/particle partitioning analysis shows that while Flo, Phe and Ant were collected almost completely on the polyurethane-foam filters (vapour adsorbent medium), B(b)F, B(k)F, B(a)P, D(a,h)A, I(123cd)P and B(g,h,i)P could only be detected in the glass-fiber filters at all localities (Table 1).

Results of the gas particle distribution behaviour analysis emphasize presence of PCB congeners mainly in the vapour phase (Table 1).

Analyzing the relative deviation of estimated ϕ values using Junge-Pankow approach to the calculated values obtained from the measured concentration values (see Fig. 1, Fig. 2 and Table 2), good concurrence is observed for high molar mass of polycyclic aromatic hydrocarbons in the atmosphere (with five or more condensed rings), as well as for the Pyr and B(a)A at the locality K3. Relative error for B(b)F, B(k)F, B(a)P, D(ah)A and B(ghi)P ranges from 0.048746 to 10.69297 %; for Pyr and B(a)A relative error amounts to 4.92% and 2.61%, respectively. The highest relative deviations of estimated/calculated ϕ values are detected for all investigated PCB congeners (relative deviations of ϕ ranges from 6.68% to extremely high values) and at the low molecular mass of PAHs - Nap, Acy and Ace (up to 99.98%) and in the case of Chr, at the city centre (127.82%). On the other hand, results obtained by the other authors cite that the vapour particle partitioning of the low molecular mass of PAHs can be predicted by Junge-Pankow model within the reasonable limits.

Detected concentrations of B(a)P (PUF+GF) ranged from levels near the detection limit (about 0.01 ng/m³) in air samples detected at locality K3 to levels of 0.135 (locality K1) and 0.158 (locality K2). Levels of B(a)P at the sites K1 and K2 are above immission limit value for the B(a)P regulated in Serbia (0.1 ng/m³). Concentration levels of other PAHs recognized by IARC as carcinogens (IARC 1987) (B(a)A and D(a,h)A) are near the detection limit for all investigated locations (Table 3). Obtained values demonstrate presence of B(a)P above immission limit value at the all field sites.

At the investigated locations, the highest total PAH concentration value was detected on the locality K2 (car factory – main production building), Σ PAH=34.309 ng/m³. Total PAH concentration levels at the other two locations amounted to Σ PAH=26.409 ng/m³ and Σ PAH=19.382 ng/m³, respectively. The most abundant component at all localities was Phe (20.933 ng/m³ at the locality K2), while D(ah)A was present in the least extent. Incomplete combustion in the coal furnace seems to be the major source of Phenantrene in the Zastava Car Factory.

PAHs with two and three rings (Nap, Acy, Ace, Flo, Phe, Ant) constitute the major fraction of atmospheric PAHs for all three field sites. The fraction of PAHs with four rings (Flu, Pyr, B(a)A, Chr) in the atmosphere is neglected for the locations K1 and K2, while it is near the fraction of PAHs with two and three rings at the site K3, the locality with the lowest overall PAH concentration. For all field sites, the contribution of PAHs with five and six rings in the atmosphere is insignificant (Fig. 3). Similar concentration trends of the investigated PAHs point out that detected levels result from the permanent incomplete combustion of organic matters at the selected sites, global atmospheric transport and persistence, without presence of the significant and specific source of pollution.

As the direct result of NATO operation in Serbia / the city of Kragujevac, high concentration levels of PCB congeners in ambient air are detected at the selected localities of the investigated region. Total concentration of the seven PCB congeners at the locality K1 inside the Zastava Car Factory has reached the value of 40.149 ng/m³ (Table 3). Locality K1, in the vicinity of the big transformer filled with pyralene, with the highest detected PCBs level presents the important source of pollution. PCB 52 and PCB 101 constitute the major fractions of the total PCBs at the locality K1 indicating the major source of contamination in the area, while the fractions of PCB 28 and PCB 180 are neglected. At the field sites K2 and K3, the similar concentration trend of individual PCB congeners with the major fraction of PCB 28 is noticed (Fig. 4).

4 CONCLUSIONS

GC/MS analysis of the air samples points out on the presence of residues of PCBs and PAHs, sampled on three localities in Kragujevac, Serbia. Since vapour pressure value is considered to be the dominant physico-chemical property of the compounds which influence the vapour particle partitioning, obtained distribution of the investigated pollutants is as expected. Initial results suggest that the gas particle partitioning of the high molar mass of PAHs is correspondant to the Junge-Pankow adsorption model. Extremely high levels of PCB congeners result from the damages on transformers filled with pyrelenes during the destruction of the industrial complex in Kragujevac. Similar concentration trends of the tested polycyclic aromatic hydrocarbons point out that detected levels result from the permanent incomplete combustion of organic matters.

5 RECOMMENDATIONS AND PERSPECTIVES

Determination of the gas particle partitioning behaviour of the selected persistent organic pollutants is of extraordinary importance for general estimation of wet and dry deposition from the atmosphere, modelling of the environmental fate and transport phenomena through the atmosphere and other environmental compartments and exposure of human population to these very toxic pollutants at micro, mezzo and macro level.

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