

Sources, Concentrations, Fate and Effects of Polycyclic Aromatic Hydrocarbons (PAHs) in the Environment. Part A: PAHs in Air

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Abstract

This paper provides a review of contemporary information on the concentrations, burdens and fate of polycyclic aromatic hydrocarbons (PAHs) in the atmosphere, with reference to the historical trends in PAH emissions. The main anthropogenic sources of PAHs and their effect on the concentrations of these compounds in air on a national-regional scale are discussed. The fate of PAHs in the air, their persistence and the main mechanisms of their losses are presented. Health hazards associated with PAH air pollution is stressed.

Keywords: polycyclic aromatic hydrocarbons, PAH, benzo(a)pyrene, organic pollutants, air pollution

Introduction

Polycyclic aromatic hydrocarbons (PAHs) belong to the group of persistent organic pollutants (POPs). These are organic contaminants that are resistant to degradation, can remain in the environment for long periods, and have the potential to cause adverse environmental effects. Some of them are susceptible to dispersion on a global scale because, in addition to having environmental persistence, they are "semi-volatile", i.e. under environmental conditions they move between the atmosphere and the Earth's surface in repeated, temperature-driven cycles of deposition and volatilisation [34]. POPs are truly multimedia contaminants which occur in all parts of the environment: atmosphere, inland and sea waters, sediments, soils and vegetation [22, 34, 36]. They are mainly of anthropogenic origin and have no significant natural sources.

PAH (which are known for their strong mutagenic, carcinogenic and toxic properties) are composed of carbon and hydrogen atoms arranged in the form of fused benzene rings (linear, cluster or angular arrangement) [28]. There are thousands of PAH compounds in the environment but in practice PAH analysis is restricted to the determination of 6 to 16 compounds. Individual PAHs differ substantially in their physical and chemical properties [15, 28, 35].

Generally, the high molecular weight compounds $i \geq 4$ aromatic rings) are less water-soluble, less volatile and more lipophilic than lower molecular weight ones [2, 15, 17, 22, 33, 34, 36] - Table 1. The best known model compound from this group is highly carcinogenic benzo(a)pyrene (BaP). PAHs are listed by the United States Environmental Protection Agency and the European Commission as priority pollutants.

The widespread occurrence of PAHs is largely due to their formation and release in all processes of incomplete combustion of organic materials. The last century of industrial development caused a significant increase of PAH concentrations in the natural environment [2, 7, 34, 36]. Investigations of the content of PAHs in ice cores from Greenland [10] showed that the present level of these compounds is about 50 times higher than in preindustrial periods; changes in their qualitative distribution suggest that the sources of PAHs shifted from biomass burning to fossil fuels combustion in the last 200 years. The general trends in PAH concentrations in the ice core were in agreement with the historical record of world petroleum production [10].

PAHs emitted to the air can be transported over long distances before they are deposited with atmospheric precipitation on soils, vegetation or sea and inland waters [33,

36]. The presence of PAHs in all these elements of the environment may create a risk not only to humans but to all living organisms.

The aim of this paper is to review contemporary information on concentrations, burdens, the fate and risk associated with the presence of these compounds in the atmosphere.

Sources of PAH Emission

PAHs are produced in all processes of incomplete combustion of organic substances [2, 12, 17, 28, 36]. Their production is favoured by an oxygen-deficient flame, temperatures in the range of 650-900°C and fuels which are not highly oxidized. Natural sources of pyrogenic PAH such as volcanic activity and forest fires do not significantly contribute - for the present - to overall PAH emission [2, 28, 36]. Anthropogenic sources can be divided into two categories: the combustion of materials for energy supply (*e.g.* coal, oil, gas, wood, *etc.*) and combustion for waste minimization (*e.g.* waste incineration) [2, 23, 36]. The first category includes stationary sources like industry (mainly coke and carbon production, petroleum processing, aluminium sintering, *etc.*), residential heating (furnaces, fireplaces and stoves, gas and oil burners), power and heat generation (coal, oil, wood and peat power plants) and mobile sources like cars, lorries, trains, airplanes and sea traffic

Table 1. Properties of PAHs

Property	PAHs ≤ 3 rings	PAHs ≥ 4 rings
Water solubility (mg/l)	32 ÷ 0.07	0.26 ÷ 0.0003
Volatility (Henry's constant - Atm. m ³ .mol ⁻¹)	5.0E-2 + 7.9E-4	3.5E-4 + 2.2E-6
Sorption affinity (log K _{ow})	3.4 + 4.6	5.2 + 7.7
Condensation temperature (°C)	-50 + -10	-10 + +30
Ecotoxicity	+	+/-
Carcinogenicity	-	++
Examples	Naphthalene (2-rings) Acenaphthene (3-rings) Fluorene (3-rings) Anthracene (3-rings)	Pyrene (4-rings) Chrysene (4-rings) Benzo[a]pyrene (5-rings) Benzo[ghi]perylene (6-rings)

(gasoline and Diesel engines). The second category covers incineration of municipal and industrial wastes. Other miscellaneous sources contain unregulated fires such as agricultural burning, recreational fires, crematoria, *etc.*, cigarette smoking as well as volatilization from soils, vegetation and other surfaces [2, 23, 36].

Table 2. Estimated annual PAH emission in different countries.

Country	Stationary fuel combustion		Industrial processes	Transport	Incineration	Total	Reference
	Domestic heating	Power stations					
Tonnes/year							
The US	1 380	400	3 500	2 170	1 150	8 600	[23]
Sweden	142	2	313	47	4	510	[23]
Sweden	-	-	-	-	-	200	[20]
Poland	265	118	320	80	5	800	[2]
Poland	464		67	35	-	566	[5]
The UK	604	6	19	80	6	712*	[36]
Germany	-	-	-	-	-	8 218	[20]
The Netherlands	-	-	-	-	-	1 116	[20]
EUROPE**	9 863		324	845	-	14 090	[33]
Percent of total							
The US	16.0	4.7	40.7	25.2	13.4	100	[23]
Sweden	28.7	0.4	61.3	9.2	0.8	100	[23]
Poland	33.1	14.8	40.0	11.3	0.6	100	[20]
Poland	82		11.8	6.2	-	100	[2]
The Netherlands	56		24	20	-	100	[36]
The UK	84.6	0.8	2.6	11.2	0.8	100	[36]
EUROPE**	70		23.0	6.0	-	99	[33]

*) more detailed estimations indicate emission of PAHs to the UK atmosphere from primary combustion sources to be greater than 1000 t/year [36].

***) estimated on the base of BaP emission data (PAHs emission = 10 x BaP emission)

Table 2 compares the estimated anthropogenic emissions of PAHs in different countries. According to the above evaluations PAH emission in individual European countries varies from as low as 200 t/year in Sweden in 1988 [20] to as high as 8,000 t/year in Germany in 1985 [20, 33, 36]. The observed discrepancies are most probably due not only to real differences in PAH emissions but rather to different calculation methods, the amount of potential sources included, and the number of PAH compounds taken into account. However, the values of 500-1000 t/year for individual Central and West European countries (i.e. 4-7% of total European emission) seem to be the most realistic at the present. While the older publications indicated industrial processes as a dominant PAH source (40-60% of total PAH emission for the US [23], Sweden [23] and Poland [2]), according to the more recent evaluations for the UK [36], Poland [5] and the whole Europe (including the European part of the former USSR [33]) most PAHs (70-85%) derive from domestic fuel and coal combustion with industry being responsible for 10-20% and transport for 6-10% of total PAH release (Table 2).

Fate and Transformations of PAHs in Atmosphere

PAHs entering the atmosphere derived from the combustion and from volatilization (Figure 1). They are present in the ambient air as vapours or adsorbed into airborne particulate matter [3, 9, 16, 17, 19, 29, 33, 36]. Gas-to-particle partition of PAHs depends on the molecular weight of the compounds, temperature, humidity and precipitation [29, 33]. In general, low-volatile PAHs with > 5 rings, characterized by relatively high temperature of condensation, are adsorbed on the airborne particles [33, 34]. They are classified in the low mobility category of POPs subjected to rapid deposition and retention close to the source [34]. The lower-molecular weight compounds with 2-3 rings, exhibiting low temperatures of condensation, are more abundant in the gas phase [29, 32, 33, 34, 36]. These hydrocarbons (included in the high or moderately high mobility categories [34]), undergo world-wide atmospheric dispersion and preferentially accumulate in polar latitudes [33, 34]. Semi-volatile 4-ring PAHs (like pyrene or phenanthrene) can be found in both phases and their gas-to-particle partition coefficients are most susceptible to the influence of environmental factors [29]. They are deposited and accumulate mainly in mid latitudes [34]. With high summer temperatures (or in tropical regions), the concentrations of PAHs in the gas phase increase whereas during winters (or in Arctic regions) particulate phase PAHs dominate [3, 17, 29, 34]. Bodek *et al.* [3] found that the content of particulate-phase PAHs in winter air samples from the Upper Silesia region in Poland is about 10-20 times higher than in summer samples (Table 3). Lenicek *et al.* [13] reported alike relationship between winter/spring air samples from the North Bohemia Region in the Czech Republic. Similar alterations were observed by other authors [17, 25]. In the case of gas phase PAH the difference was much smaller [17]. Higher PAH concentrations (in general measured in the particulate phase) in the winter months were related also to the higher emission of these pollutants from domestic heating sources [3, 36]. The adsorption of PAHs onto particulate phases may be affected not only by temperature but by humidity as well: it was found [17, 29] that the gas-to-particle PAH ratio decreases

with increasing humidity. The range of PAH adsorption on atmospheric sorbents depends also on the quantity of the suspended particulates and their nature (soot, dust, fly-ash, pyrogenic metal oxides, pollens, *etc.*, of different particle size) [17, 29, 33] and a significant correlation was found between the amount of dust in the air and PAH concentrations in the particulate phase [32, 36].

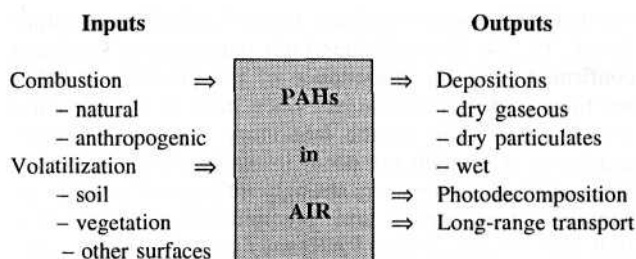


Fig. 1. Schematic diagram of PAH fate in atmosphere.

PAHs present in the atmosphere are subject to complex physico-chemical reactions and transformations in the atmosphere; dry and wet deposition, photochemical transformations and reactions with other pollutants [9, 19, 24, 32] (Figure 2). The physical mechanism of PAHs loss from the atmosphere is deposition [17, 19, 32, 33, 36]. PAH associated with particulates are subject to gravitational settling and scavenging by precipitation/water vapour with efficiency related to the depository surface type. Van Jaarsveld *et al.* [33] found that the dry deposition rate of BaP over land surface is nearly twice as high as over sea surface. The differences in these velocities the authors [33] ascribed partly to the contrast in the roughness length and partly to the different atmospheric stability regimes. For PAHs in particle phase the removal rates are a function of the physical parameters of the carrier particle, of which particle size is the most important [17, 32, 33]; e.g. mean dry deposition velocity for particle class with median aerodynamical diameter of 0.2 μm was calculated to be 0.000065 $\mu\text{m}/\text{sec}$ while for the particles with diameter of 40 μm -0.067 m/sec [33]. Higher molecular weight PAHs, like BaP, are supposed to be emitted with substantial fraction on large particles [33]. According to Van Jaarsveld *et al.* [33] the variations in particle size distribution and in the BaP conversion rates have a dramatic effect on air concentrations and the dry and wet depositions individually, while total BaP deposition varies much less due to mass conservation mechanisms. Gustafsson *et al.* [6] calculated that about 6 t of pyrene and 3 t of BaP *per year* are entering the western part of the North Atlantic through atmospheric precipitation. Wild and Jones [36] estimated that about 23% of total PAH input to the UK atmosphere are deposited each year to the ground surface by wet/dry deposition. Simonich and Hites [27] evaluated for the northeast US that about 60% of all PAH entering this airshed are deposited directly to soils, waters and vegetation. Hence, from about 75 to 40% [27, 36] of all PAHs are removed from air by other processes, and photochemical transformations are considered to be the most important among them. Observation made so far indicate that PAH in the vapour phase are more susceptible to such reactions than associated with particulates [9, 32, 36]. Thus adsorption on highly porous, carbonaceous particles like soot or fly-ash may provide some protection from photooxidation [19]. Kamens *et al.* [9] found that at moderate temperatures the rate of PAH photodecomposi-

tion may decrease with increased PAH particle loadings. In the absence of light some PAHs may react with molecular oxygen but these reactions appear to be very slow and to represent an insignificant degradation pathway [19]. PAHs have been found to react as well with atmospheric ozone, with NO_x (to produce nitro-PAHs which are potentially more mutagenic and carcinogenic than PAH precursors), with SO_x and OH radicals [1, 19, 24, 26, 32, 36]. Whereas nitrated PAH compounds have been identified in air samples [1, 19, 24, 35] and their high mutagenicity has been confirmed [26], the importance of the products of other reactions is difficult to assess since most of the literature data is based only on the laboratory experiments under conditions that might not occur in reality [32]. The persistence of PAHs in the air is strongly influenced by sunlight, humidity, temperature and precipitation [19, 33, 36]. Half-lives of atmospheric PAHs may vary from hours (sunlight, moderate temperatures and humidity) to days or even weeks (low intensity sunlight, low temperatures, low humidity) [9]. Long-range transport of PAHs reported for both Europe and the US (being one of the outputs of PAHs from the atmosphere in the given area) confirms low degradation of these contaminants, particularly those adsorbed on very small particles of suspended matter [6, 10, 19, 32, 33].

The Content of PAHs in the Air

Air contributes to a very small degree (< 0.5%) to total PAH burden in the environment (Table 4), even though all PAHs released in the natural and anthropogenic combustion processes enter the atmosphere. According to Wild and Jones [36] the UK air mass contains only ~ 4% of PAHs emitted each year in this country. From this observation authors [36] concluded that "the atmosphere is not a repository and collector of PAH but more likely to be a transporter, dilutor and reactor."

Comprehensive information on the calculated (TREND model using 1990 emission data) area-average concentrations of BaP in air for different European countries was given recently by Van Jaarsveld *et al.* [33]. The highest concentrations of BaP in air were found in the former Czechoslovakia (2.9 ng/m³), Hungary (1.9 ng/m³) Germany (1.5 ng/m³), Poland (1.4 ng/m³) and Austria (1.3 ng/m³). The lowest in: Finland (0.034 ng/m³), Sweden (0.040 ng/m³), Norway (0.042 ng/m³) and Portugal (0.067 ng/m³). Assuming BaP being about 1/10 of total PAH content [5, 12, 28], this would correspond to approximately 10-30 ng of ΣPAH/m³ in the central part of Europe and about 0.3-0.7 ng/m³ in the least polluted peripheral countries of the

Table 3. The content of PAHs in air as affected by the location and the season.

Location	PAH concentrations : mean {range} (ng/m ³)		References
	ΣPAH	BaP	
Czech Republic – Košetice	15.4 {0.54 – 101}	0.04	[7]
Czech Republic – Prachatice		3.5 {n.d. – 8.6}	[12]
Czech Republic – Teplice		4.8 {n.d. – 11.7}	[12]
/winter/summer/		5.50 / 0.51	[13]
France – Paris		7.9 / 0.8	[17]
particulate phase /winter/summer/		0.05 / 0.03	[17]
gas phase /winter/summer/			
The Netherlands – Rotterdam		1.2 / 0.20	[33]
/winter/summer/			
Norway – Elverum /annual/	158 {24 – 494}		[25]
/winter: cold/	372 {180 – 494}		[25]
/winter: mild/	34 {23 – 48}		[25]
Poland – Upper Silesia region			
/winter/summer/	{3 – 400} / {1 – 60}		[3]
Poland – Gdańsk		6	[5]
Poland – Biała Podlaska	20.4		[21]
Slovakia (14 localities)	{36 – 389}	{0.37 – 8.36}	[11]
The UK (urban air)	{148}	{0.65}	[36]
The UK – Lancaster		0.40	[30]
gas + particulate phase		0.004	[30]
gas phase			
The US	5.7 {0.2 – 65}		[9]
The US, Columbus, OH	13 {2 – 75}		[9]
The US, Great Lakes	{0.2 – 0.3}		[9]
particulate phase		{0.012 – 0.090}	[4]
gas phase		{0.004 – 0.024}	[4]
Remote aerias – "background concentration"		0.001	[33]
Atlantic – tropical zone	{0.10 – 0.20}		[16]

Table 4. Partition of PAHs in natural environment (Ref. [36])

Element of the environment	Percent of total	
	ΣPAH	BaP
Soil	94.4	92.9
Freshwater sediments	5.4	7.1
Water	< 0.01	< 0.01
Air	0.1	< 0.01
Vegetation	0.1	< 0.01
Biota	< 0.01	< 0.01

continent. The results of these evaluations confirm that the concentrations of higher molecular PAHs in the atmosphere follows their emission pattern (Table 2). The concentrations of BaP in air over the Baltic and North Seas were evaluated as 0.78 ng/m³ and 0.75 ng/m³, respectively, which is comparable to the calculated average level of this pollutant in the atmosphere over countries such as the former Yugoslavia, Romania or Belgium [33].

Examples of typical concentrations of PAH and BaP in air as affected by the localization of the sampling point and the season are given in Table 3. They vary widely from as low as < 0.1 ng/m³ in remote areas such as the Atlantic tropical zone [16] or the Arctic [33] to as high as nearly 600,000 ng/m³ in a coke-oven plant (the vicinity of the furnace) [14]. Highly urbanized cities in the US show levels of carcinogenic PAH (being only a fraction of total PAHs) of 15-50 ng/m³, whereas in rural areas the median carcinogenic PAH content in air ranges from 2.6 ng/m³ to 4.4 ng/m³ [18]. In the UK the median total PAH content in urban atmosphere was reported to be much higher - 148 ng/m³ [30, 36], which is very similar to the concentrations found in the town of Elverum in Norway (158 ng/m³ [25]). Even higher values were noted in the most urbanized and industrialized (coal-mining) Upper Silesia region of Poland, where mean annual PAH concentrations were reported to be in the range 150-900 ng/m³ [3,21], or in the industrialized town of Kosice in Slovakia (689 ng/m³ [11]). Unpolluted regions of these countries were characterized by much lower levels of PAHs in air: 20 ng/m³ in Biala

Podlaska in Poland [21], and 36 ng/m³ in Starina in Slovakia [11]. This corresponds very well with the estimate by Van Jaarsveld *et al.* [33] average content of these hydrocarbons in air in Central European countries (15-30 ng/m³).

Risk to Humans

Most of the particle-bound PAHs are associated with small-size respirable particles [19]. In general, chemicals administered to the lung as fine particulates, in aerosol form, are expected to be more bioavailable than those administered in solid matrices like food [18]. According to Menzieef *al.* [18] estimated potential doses of carcinogenic PAHs by inhalation range between about 0.02 µg/day and 3 µg/day with a median value of 0.16 µg/day, which is nearly 20 times less than the calculated food dose and about 25 times more than the potential dose with drinkable water (Table 5). Research shows [18, 22, 31] that air contributes 3-20% of total human exposure to PAHs and comes in second position (after food) as a source of these pollutants for humans. In certain cases, where urban/industrial atmospheres contain the highest ambient concentrations of PAHs, air is a predominant exposure route [18]. Cigarette smoke can significantly contribute to potential PAH doses via inhalation (over 50% of total exposure - Table 5). The ambient standards regarding exposure to PAHs refer usually to BaP. For example in Germany and Poland permissible BaP concentration in air as a daily exposure is 5 ng/m³ and as a year exposure - 1 ng/m³ [3]. The similar atmospheric BaP levels are accepted in the US (year exposure in the range 0.3-0.7 ng/m³) [8].

Summary

Contemporary information on the concentrations and fate of PAHs in air were reviewed. The partitioning of these compounds in the atmosphere is evaluated, with reference to the historical trends in PAH emissions. The main anthropogenic sources of PAHs were discussed. Domestic coal combustion, coal-fired power stations, industrial processes and vehicles are estimated to make major contributions to contemporary national-regional PAH emissions, although the relative proportions attributed to these sources will vary for different PAH compounds and between countries. The presented information indicates that large uncertainties still exist in atmospheric sources, loads, fates and degradation rates of PAHs and further data enabling more accurate general model evaluations are needed.

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Table 5. Potential doses of carcinogenic PAHs (Ref. [18]).

Source of PAHs	Intake			
	Nonsmokers		Smokers	
	µg/day	Percent of total	µg/day	Percent of total
Food	3	93.0	3	44.6
Air	0.16	4.9	0.16	2.4
Water	0.006	0.2	0.006	< 0.01
Soil (accidental ingestion)	0.06	1.9	0.06	1
Cigarette smoke	-	-	3.5*	52
Total	3.22	100	6.72	100

*) average of 2-5 µg/day.

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