

## SOURCES AND DISTRIBUTION OF POLYCYCLIC AROMATIC HYDROCARBONS IN WET DEPOSITION IN URBAN AND SUBURBAN AREAS OF GIZA, EGYPT

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**ABSTRACT:** Wet deposition (rain) samples were collected at three locations, two in urban and suburban areas of Giza (Dokki and Kafr Tohormos) and background ones in a rural area of Menofiya province (Kafr El-Akram) during the period of November, 2004 – April, 2005. In the suburban area, the particle-bound PAH concentrations in suspended particulate matter were also determined before and after rainfall. The highest concentrations of PAH compounds in wet deposition were found in the urban area, whereas the lowest concentrations were detected in the rural area and the differences in mean concentrations were statistically significant ( $p < 0.001$ ). The concentrations of the total PAH compounds were 25.897, 13.647, and 0.023  $\mu\text{g/L}$  in the particulate phase, 11.288, 6.365, and 0.0871  $\mu\text{g/L}$  in the dissolved phase, and 37.185, 20.012, and 0.1101  $\mu\text{g/L}$  in the particulate+dissolved phases in rain water at the urban, suburban, and rural areas, respectively. Benzo(b)fluoranthene, benzo(ghi)perylene, and benzo(a)anthracene were the dominant PAH compounds in wet deposition at both urban and suburban sites, indicating that a significant fraction of heavy PAH compounds were emitted from traffic due to the incomplete combustion of fuel. On the other hand, naphthalene, phenanthrene, anthracene, and fluorine were the predominant PAH compounds in wet deposition in the rural area, indicating that the PAHs of lower molecular weight predominated. The total carcinogenic PAH compounds represented 59.68, 59.19, and 21.62% of the total PAH concentrations in wet deposition in the urban, suburban, and rural areas, respectively. In suspended particulate matter, the concentrations of the total PAH compounds in the suburban area were 1588.7 and 609.65  $\mu\text{g/g}$  before and after rainfall, respectively. Motor vehicles were the major source of PAHs in the atmosphere of the areas studied and, consequently, in the wet deposition, and they mainly originated from pyrolytic processes.

**KEY WORDS:** Urban/suburban/rural areas, wet deposition, suspended particulate matter, polycyclic aromatic hydrocarbons, carcinogenic compounds

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**Abbreviations:**

ACE = acenaphthene	FLU = fluorine
ACY = acenaphthylene	FLT = fluoranthene
ANT = anthracene	IND = indeno(1, 2, 3,-cd)pyrene
BaA = benzo(a)anthracene	LMW = low molecular weights
BaP = benzo(a)pyrene	NA = naphthalene
BbF = benzo(b)fluoranthene	PAHs = polycyclic aromatic hydrocarbons
BGP = benzo(ghi)perylene	PHE = phenanthrene
CRY = chrysene	PYR = pyrene
DBA = dibenzo(a,h)anthracene	

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## INTRODUCTION

The urban atmosphere is subjected to huge inputs of anthropogenic contaminants produced by both stationary and mobile sources (Schauer et al., 1996; Bilos et al., 2001). Polycyclic aromatic hydrocarbons (PAHs) are a group of environmental contaminants composed of two or more fused aromatic rings. They are ubiquitous in the environment (Baek et al., 1991; Wilcke, 2000). In the atmosphere, PAHs are semi-volatile organic compounds distributed both in gaseous and particle-associated (airborne particles) phases, depending on the ambient temperature and particulate matter levels; they are subjected to removal mechanisms such as oxidative and photolytic reactions and wet and dry deposition (Garban et al., 2002). Low molecular weight PAHs (two to three ring-PAHs) are found mostly in the gas phases, whereas high molecular weight PAHs (five- to six-ring PAHs) are often associated with airborne particles (Gundel et al., 1995; Lighty et al., 2000; Zheng and Fang, 2000). Semivolatile four-ring PAHs were found in both gaseous and particulate phases (Park et al., 2002; Sin et al., 2003). PAHs are produced by incomplete combustion of fossil and non-fossil fuels (Venkataraman and Friedlander, 1994; Khalili et al., 1995; Harrison et al., 1996; Mastral et al., 2003; Di Lella et al., 2006; Luo et al., 2006). In the big cities, the atmospheric PAH levels remain high due to increasing vehicular traffic and low dispersion of atmospheric pollutants (Miguel et al., 1998). PAHs can also be found in rural areas (Gevao et al., 2000; Garban et al., 2002) and remote regions (Kallenborn et al., 1998; Migaszewski, 1999) due to their persistence in the environment and their ability to be transported over long distances (Aamot et al., 1996; Halsall et al., 2001).

The lower molecular weight PAHs are known to be less toxic and found predominantly in the gaseous phase in the urban atmosphere (Halsall et al., 1994). They can react with other pollutants such as ozone and nitrogen oxides to form more toxic derivatives such as carcinogenic nitro-derivatives which are formed from the reaction of PAHs with  $\text{NO}_3$  (Arey et al., 1989). Metabolites of some high molecular weight PAHs such as benzo(a)anthracene, chrysene, benzo(b,k)fluoranthene, and benzo(a)pyrene were reported to be mutagenic and carcinogenic in fish, mammals, and humans (Zedeck, 1980; Menzie et al., 1992; Moore, 1995).

The removal of chemical pollutants from the atmosphere takes place through chemical transformations as well as by wet and dry deposition (Duce et al., 1983; Prospero et al., 1983). Atmospheric transportation and deposition of contaminants have been linked to uptake in aquatic and terrestrial biota (Bidleman, 1999). PAHs are globally cycled in the atmosphere due to their chemical/microbiological stability, low water solubility, lipophilic properties, and to existent vapour pressure (Duce et al., 1991) and they may accumulate in both aquatic and terrestrial systems. They can be deposited by direct impact of airborne particulate and gaseous molecules on surfaces (dry deposition) and/or by precipitation (wet deposition) (Motelay-Massei et al., 2003). Dry deposition predominates in the vicinity of urban/industrial areas, whereas wet deposition predominates in more remote areas (Golomb et al., 1997). The mean concentrations of the total PAH compounds were 450 and 140 ng/L in the dissolved and particulate phases of rain water, respectively, at an urban area in



Switzerland (Leuenberger et al., 1988). Total PAHs concentration in precipitated rain was 1422.3 ng/L at Lake Balaton, Hungary (Kiss et al., 1997). The total PAH levels ranged from 50 to 312 ng/L in rain samples at Seabrook, TX, USA (Park et al., 2001). The concentration of the total PAHs varied from 84.25 to 777.49 ng/L in rain water at Mumbai, India (Sahu et al., 2004). There always exists a possibility that the atmospheric precipitation of the PAHs may cause ecological damage in surrounding regions. Atmospheric deposition is a significant source of PAHs to the surface waters of lakes, estuaries, and the remote ocean, especially to waters downwind of urban and industrialized areas (Eisenreich et al., 1981; Duce et al., 1991; Leister and Baker, 1994; Golomb et al., 1997). Jones et al. (1989) found that atmospheric deposition of particulate-bound PAHs was the principal source of PAHs in Welsh soils. The levels of PAHs in urban soils and in the sediments of lakes and bays were high due to the deposition of atmospheric particulates containing PAHs from urban areas (Kennicutt et al., 1993; Wild and Jones, 1995; Wang et al., 2003). Therefore, the necessity for the investigation of PAHs in wet precipitation has been raised in urban and suburban areas of Giza, Egypt, too.

The present study aimed to obtain information on the concentrations, distribution, and origin of PAHs in wet deposition at three sampling sites, two in urban and suburban areas and one in a rural area, and to investigate the effect of rainfall on the atmospheric concentrations of PAHs in the suburban area studied.

## MATERIALS AND METHODS

### Sampling sites

Wet deposition (rain) samples were collected at three locations: two in urban and suburban areas of Giza (Dokki and Kafr Tohormos, respectively) and one in a rural area of Menofiya province. The urban area (Dokki) is located to the southwest of the Cairo city centre; it is characterized by heavy traffic and various human activities yielding air pollutants, as nitrogen and sulfur oxides, carbon oxides, hydrocarbons, ozone, and suspended particulate matter. The suburban area (Kafr Tohormos) lies southwest of the Cairo city centre near to an agricultural region. It is characterized by a relatively low traffic density, low-level human activities, and unpaved roads. On the other hand, the background rural area is located in Menofiya province, namely Kafr El-Akram, about 55 km to the north of Cairo, where the major activities of the inhabitants are usually associated with agriculture and some light industries.

Climatologically, both Giza and Menofiya are in the subtropical climatic zone. The general climate of both cities is cold, moist, and rainy in winter, whereas it is characterized by high temperature, high solar radiation, clear sky, and aridity during summer season. According to the data obtained from the Egyptian Meteorological Authority, the monthly rainfall ranged from 1.4 mm (February, 2005) to 8 mm (March, 2005) in the urban and suburban areas of Giza, and 0.1 mm (December, 2004) to 4.6 mm (January, 2005) in the rural area during the period of study.



### Sample collection

At each site, the wet deposition (rain) samples were collected during the period of November, 2004 – April, 2005. Rain water was collected through a glass funnel fitted into a glass receiving bottles according to Kiss et al. (1997). The collectors were placed on the top of four-storey buildings in about 10 m height from ground level on an event basis. They were deployed as soon as the rain began and retrieved immediately after the rain stopped. Upon arrival at the laboratory, the rain samples were filtered through a glass fiber filter to separate particulates. The PAH compounds on the glass fiber filter were defined as the particulate phase, while those in the filtrate as the dissolved phase. Both glass fiber filters and filtrates were stored at 4°C in darkness. Due to low volume of rain water in Egypt, the glass fiber filters as well as the filtrates during each month were combined to produce large sample volume for analysis. Both particulate and dissolved phases of PAHs were separately determined in each sample.

To investigate the effect of rainfall on the concentration of the atmospheric PAHs, total suspended particulate matter in the ambient air of the suburban area was collected before and after rainfall. The samples were collected on glass fiber filters (Whatmann, 20 cm × 25 cm) using a high volume sampler at approximately 1.2 m<sup>3</sup>/min. Prior to sampling, the glass fiber filters were combusted at 450°C for 4 h (Park et al., 2001).

### Sample extraction

The glass fiber filters were Soxhlet extracted with methylene chloride (CH<sub>2</sub>Cl<sub>2</sub>) for 24 h (Baker et al., 1992). The extract was then concentrated to about 3 mL on a rotary evaporator. On the other hand, a liquid-liquid technique was used to extract the PAH compounds in the filtrate, i.e., the dissolved phase in rain water samples. The filtrate was directly taken into a separating funnel of suitable capacity and 100 mL of a hexane/methylene chloride mixture (v/v 85/15) was added for each liter of the sample (Ollivon et al., 1999), followed by vigorous shaking for 20 min. The hexane/methylene layer containing extracts was then collected on anhydrous sodium sulfate layer in a beaker. This procedure was repeated 3 times to ensure complete extraction. The extracts were combined and reduced to about 3 mL on a rotary evaporator.

Before gas chromatography (GC) analysis, the extracts were purified using silica gel/alumina column chromatography consisting of 5 g anhydrous sodium sulfate, 20 g silica gel (deactivated 5% with distilled water), 10 g alumina (deactivated 1% with distilled water), 5 g sand, and glass wool to remove interferences according to Park et al. (2001). The extracts were concentrated, exchanged to 2 mL hexane, placed on the columns, and eluted with 1:1 pentane-dichloromethane (200 mL). The eluted extract was concentrated on a rotary evaporator and exchanged to 1 mL hexane and stored at 4°C till analysis.

A gas chromatography (Hewlett-Packard HP6890) fitted with a Flame Ionization Detector (FID) was used. A HP-5 (30 m × 320 μm × 0.25 μm) capillary column with hydrogen as carrier gas was used. It was calibrated with a diluted standard so-



lution of 15 compounds (PAH mixture, Supelco, Inc., Bellefonte, PA), and the concentrations of the target PAH compounds were quantified using this external standard solution. The concentrations of the following PAHs were determined: naphthalene (NA), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FLU), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLT), pyrene (PYR), benzo (a)anthracene (BaA), chrysene (CRY), benzo(b)fluoranthene (BbF), benzo(a)pyrene (BaP), dibenzo(a,h)anthracene (DBA), benzo(ghi)perylene (BGP), and indeno(1,2,3,-cd)pyrene (IND).

### Statistical analysis

Student's *t* test was used to estimate the significant difference between the mean concentrations of the individual PAH compounds in the study areas and between the mean concentrations before and after rainfall according to Gregory (1963).

## RESULTS

The monthly range and total period mean concentrations of the PAH compounds in the particulate and dissolved phases in rain water samples at the three sampling sites are summarized in *Table 1*. It is clear from it that the more abundant PAH compounds were BbF and BGP in the particulate phase and NA in the dissolved phase in rain water at each of the three sites. In both phases, the highest concentrations of the individual PAH compounds were found in the urban area, whereas the lowest levels were detected in the rural one, and the differences in mean concentrations were statistically significant ( $p < 0.001$ ). The mean concentrations of the individual PAH compounds in particulate phase ranged from 0.0130  $\mu\text{g/L}$  for NA to 6.2881  $\mu\text{g/L}$  for BbF in the urban area, 0.0065  $\mu\text{g/L}$  for NA to 3.4730  $\mu\text{g/L}$  for BbF in the suburban area, and 0.00015  $\mu\text{g/L}$  for NA to 0.0041  $\mu\text{g/L}$  for BbF in the rural area during the period of study. In addition, the concentrations of the total PAH compounds in particulate phase in rain water were 25.897, 13.647, and 0.023  $\mu\text{g/L}$  at the urban, suburban, and rural site, respectively (*Fig. 1*). In the dissolved phase in rain water, the mean levels of the individual PAH compounds ranged from 0.2551  $\mu\text{g/L}$  for DBA to 3.690  $\mu\text{g/L}$  for NA in the urban area, 0.116  $\mu\text{g/L}$  for BaP to 2.099  $\mu\text{g/L}$  for NA in the suburban area, and 0.0001  $\mu\text{g/L}$  for DBA to 0.0145  $\mu\text{g/L}$  for NA in the rural area during the period of study (*Table 1*). Moreover, the concentrations of the total PAH compounds in the dissolved phase in rain water were 11.288, 6.365, and 0.0871  $\mu\text{g/L}$  at the urban, suburban, and rural sites, respectively (*Fig. 1*). The mean levels of the individual PAH compounds in rain water (particulate + dissolved phases) ranged from 0.3630  $\mu\text{g/L}$  for ANT to 6.5536  $\mu\text{g/L}$  for BbF in the urban area, 0.193  $\mu\text{g/L}$  for ACE to 3.598  $\mu\text{g/L}$  for BbF in the suburban area, and 0.0029  $\mu\text{g/L}$  for DBA to 0.0146  $\mu\text{g/L}$  for NA in the rural area during the period of study (*Table 2*). The concentrations of the total PAH compounds in wet deposition were 37.185, 20.012, and 0.1101  $\mu\text{g/L}$  in the urban, suburban, and rural areas, respectively (*Fig. 1*). In addition, the total PAH concentrations in the particulate phase were higher than those found in the dissolved phase in rain water in both urban and

TABLE 1. Monthly range and total period mean concentrations ( $\mu\text{g/L}$ ) of PAH compounds in particulate and dissolved phases in rain water in the urban, suburban, and rural areas

	Urban		Suburban		Rural		
	Particulate	Dissolved	Particulate	Dissolved	Particulate	Dissolved	
NA	Range	0.0091 – 0.0227	2.5092 – 6.2730	0.0044 – 0.0110	1.3853 – 3.4633	0.00007 – 0.00025	0.00725 – 0.02537
	Mean	0.0130	3.6900	0.0065	2.0990	0.00015	0.0145
ACY	Range	0.0092 – 0.0229	0.3243 – 0.8177	0.0051 – 0.0128	0.1993 – 0.4983	0.00010 – 0.00036	0.00280 – 0.00980
	Mean	0.0133	0.4810	0.0075	0.3020	0.0002	0.0056
ACE	Range	0.0098 – 0.0244	0.2686 – 0.6715	0.0052 – 0.0132	0.1221 – 0.3052	0.00011 – 0.00040	0.00310 – 0.01085
	Mean	0.0132	0.3950	0.0080	0.1850	0.0002	0.0062
FLU	Range	0.0105 – 0.0263	0.3603 – 0.9008	0.0059 – 0.0148	0.1868 – 0.4669	0.00017 – 0.00061	0.00445 – 0.01557
	Mean	0.0155	0.5299	0.0090	0.2830	0.0003	0.0089
PHE	Range	0.0173 – 0.0436	0.2950 – 0.7379	0.0105 – 0.0264	0.1841 – 0.4603	0.00021 – 0.00073	0.00500 – 0.01750
	Mean	0.0257	0.4341	0.0160	0.2790	0.0004	0.0100
ANT	Range	0.0101 – 0.0253	0.2344 – 0.5917	0.0053 – 0.0132	0.1511 – 0.3778	0.00020 – 0.00072	0.00485 – 0.01697
	Mean	0.0149	0.3481	0.0080	0.2290	0.0004	0.0097
FLT	Range	0.0413 – 0.1037	0.2923 – 0.7308	0.0251 – 0.0627	0.1828 – 0.4570	0.00035 – 0.00123	0.00390 – 0.01365
	Mean	0.0610	0.4299	0.0380	0.2770	0.0007	0.0078
PYR	Range	0.0180 – 0.0451	0.2525 – 0.6376	0.0125 – 0.0313	0.1637 – 0.4092	0.00024 – 0.00085	0.00375 – 0.01312
	Mean	0.0265	0.3751	0.0190	0.2480	0.0005	0.0075



	Urban		Suburban		Rural	
	Particulate	Dissolved	Particulate	Dissolved	Particulate	Dissolved
BaA	Range 1.8553 – 4.8199	1.1264 – 2.8161	0.9008 – 2.2522	0.6191 – 1.5477	0.00073 – 0.00257	0.00325 – 0.01137
	Mean 2.9176	1.6577	1.4650	0.9380	0.0014	0.0065
CRY	Range 1.2973 – 3.2444	1.0600 – 2.6500	0.6248 – 1.5619	0.5782 – 1.4454	0.00105 – 0.00367	0.00350 – 0.01225
	Mean 1.9320	1.5588	0.9860	0.8760	0.0021	0.0070
BbF	Range 4.1868 – 10.467	0.1797 – 0.4513	2.3640 – 5.9100	0.0825 – 0.2062	0.00190 – 0.00665	0.00075 – 0.00262
	Mean 6.2881	0.2655	3.4730	0.1250	0.0041	0.0015
BaP	Range 1.8456 – 4.6416	0.1813 – 0.4607	0.7887 – 1.9717	0.0766 – 0.1914	0.00154 – 0.00540	0.00030 – 0.00105
	Mean 2.8186	0.2710	1.3950	0.1160	0.0031	0.0006
DBA	Range 2.4190 – 6.0975	0.1765 – 0.4336	1.3028 – 3.2571	0.0970 – 0.2425	0.00140 – 0.00490	0.00005 – 0.00017
	Mean 3.5868	0.2551	1.9740	0.1470	0.0028	0.0001
BGP	Range 2.8591 – 7.3978	0.2092 – 0.5054	1.6303 – 4.0758	0.0917 – 0.2293	0.00175 – 0.00612	0.00035 – 0.00122
	Mean 4.3340	0.2973	2.1520	0.1390	0.0035	0.0007
IND	Range 2.6190 – 6.5225	0.2098 – 0.5086	1.3134 – 3.2835	0.0805 – 0.2013	0.00160 – 0.00560	0.00025 – 0.00087
	Mean 3.8368	0.2992	2.0900	0.1220	0.0032	0.0005

All differences of the mean values in the Table are statistically significant at  $p < 0.001$ .  
For the full terms of abbreviations see the List of Abbreviations.

suburban areas (Fig. 1). On the other hand, the total PAH levels in the dissolved phase were higher than those found in the particulate phase in rain water of the rural area (Fig. 1).

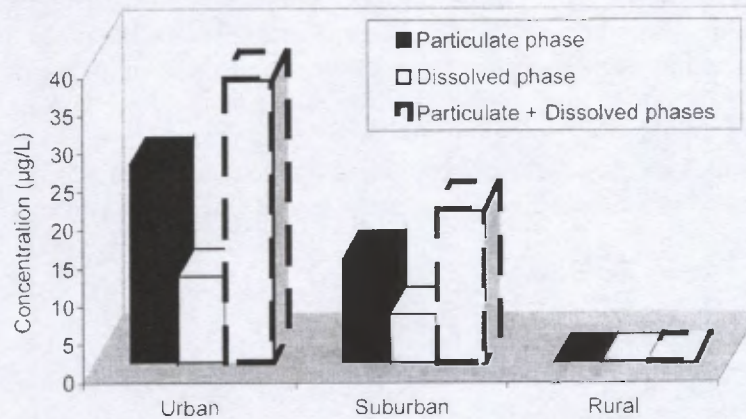


Fig.1. The total concentrations of PAH compounds in the particulate phase, dissolved phase and particulate + dissolved phases in rain water during the period of study at the three sampling sites

TABLE 2. Monthly range and total period mean concentrations (µg/L) of PAH compounds in rain water (particulate + dissolved phases) in the urban, suburban, and rural areas

	Urban		Suburban		Rural	
	Range	Mean	Range	Mean	Range	Mean
NA	2.5193 – 6.2937	3.7030	1.3810 – 3.4738	2.1055	0.00734 – 0.02560	0.0146
ACY	0.3339 – 0.8400	0.4943	0.2048 – 0.5108	0.3095	0.00293 – 0.01011	0.0058
ACE	0.2790 – 0.6951	0.4082	0.1278 – 0.3180	0.1930	0.00325 – 0.01120	0.0064
FLU	0.3711 – 0.9265	0.5454	0.1929 – 0.4812	0.2920	0.00464 – 0.01615	0.0092
PHE	0.3130 – 0.7810	0.4598	0.1949 – 0.4862	0.2950	0.00525 – 0.01820	0.0104
ANT	0.2451 – 0.6160	0.3630	0.1568 – 0.3908	0.2370	0.00508 – 0.01765	0.0101
FLT	0.3341 – 0.8339	0.4909	0.2082 – 0.5193	0.3150	0.00427 – 0.01486	0.0085
PYR	0.2710 – 0.6820	0.4016	0.1765 – 0.4401	0.2670	0.00399 – 0.01397	0.0080
BaA	2.9828 – 7.6315	4.5753	1.5209 – 3.7990	2.4030	0.00401 – 0.01390	0.0079
CRY	2.3588 – 5.8900	3.4908	1.2038 – 3.0065	1.8620	0.00458 – 0.01590	0.0091
BbF	4.3678 – 10.9170	6.5536	2.4473 – 6.1150	3.5980	0.00268 – 0.00923	0.0056
BaP	2.0275 – 5.1015	3.0896	0.8659 – 2.1620	1.5110	0.00186 – 0.00640	0.0037
DBA	2.5960 – 6.5300	3.8419	1.3998 – 3.4996	2.1210	0.00145 – 0.00507	0.0029
BGP	3.0693 – 7.9020	4.6313	1.7229 – 4.3040	2.2910	0.00213 – 0.00731	0.0042
IND	2.8298 – 7.0300	4.1360	1.3946 – 3.4835	2.2120	0.00185 – 0.00647	0.0037

All the differences of mean values in the Table are statistically significant at  $p < 0.001$ .  
For the full terms of abbreviations see the List of Abbreviations.



The concentration of various PAH compounds in suspended airborne particulate matter before and after rainfall in the suburban area is presented in Fig. 2. It can be seen that the highest concentrations of individual PAH compounds in suspended particulate matter were found before rainfall, whereas the lowest levels of these compounds were observed after rainfall, and the difference of the mean concentrations was statistically significant ( $p < 0.001$ ). The mean concentrations of the individual PAH compounds in suspended particulate matter ranged from 19.7  $\mu\text{g/g}$  for NA to 301  $\mu\text{g/g}$  for BbF before rainfall and from 7.55  $\mu\text{g/g}$  for NA to 115.33  $\mu\text{g/g}$  for BbF after rainfall. In addition, the concentrations of the total PAHs were 1588.7  $\mu\text{g/g}$  before rainfall and 609.65  $\mu\text{g/g}$  after rainfall; the total PAHs concentration ratio before and after rainfall was 2.61 (Fig. 3).

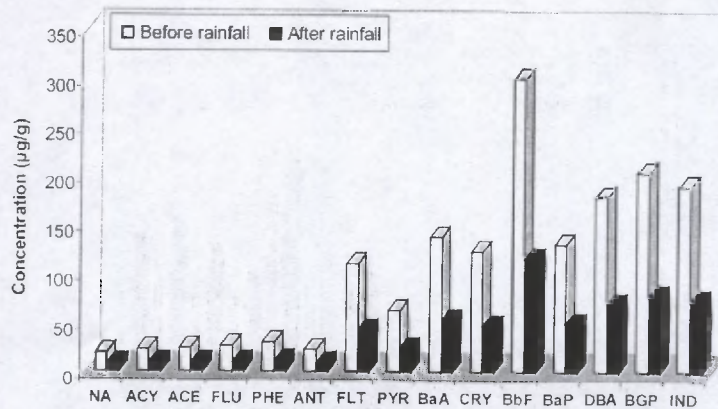


Fig. 2. Concentrations of the individual PAH compounds in suspended particulate matter in air before and after rainfall at the suburban site. The full terms of abbreviations see in the list of abbreviation

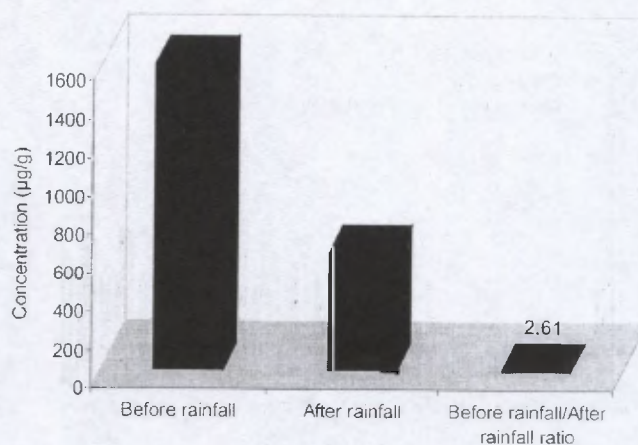
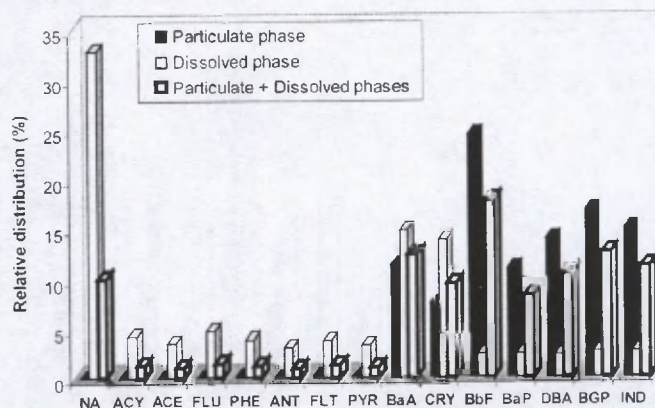
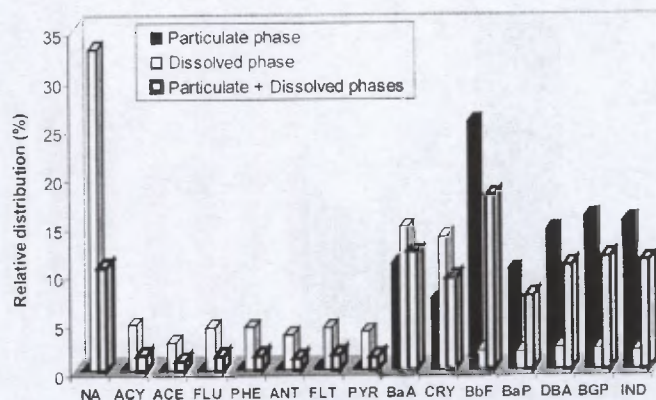


Fig. 3. The total concentrations of PAH compounds before and after rainfall in the suburban area

The contribution of an individual PAH to the total PAH concentration in the particulate phase, dissolved phase and particulate and dissolved phases at the three sampling sites are graphically represented in *Figs. 4–6*. It is clear that BbF, BGP, IND, and DBA were predominant in the particulate phase, whereas NA, BaA, and CRY were predominant in the dissolved phase in rain water in both the urban and suburban areas (*Figs. 4 and 5*). In addition, BbF, BGP, and BaA were the dominant PAH compounds in rain sampling (particulate + dissolved phases) at the same sites. At the rural site (Kafr El-Akram), the predominant PAH compounds in the particulate phase were BbF, BGP, IND, and BaP, whereas NA, PHE, ANT, and FLU were those in the dissolved phase in rain water (*Fig. 6*). Moreover, NA, PHE, ANT, and FLU (two–three aromatic rings) were the predominant PAH compounds in rain sampling (particulate + dissolved phases).



*Fig. 4. Relative distribution of PAH compounds in the particulate phase, dissolved phase and particulate + dissolved phases in rain water during the period of study at the urban site. The full terms of abbreviations see in the list of abbreviation*



*Fig. 5. Relative distribution of PAH compounds in the particulate phase, dissolved phase and particulate + dissolved phases in rain water during the period of study at the suburban site. The full terms of abbreviations see in the list of abbreviation*



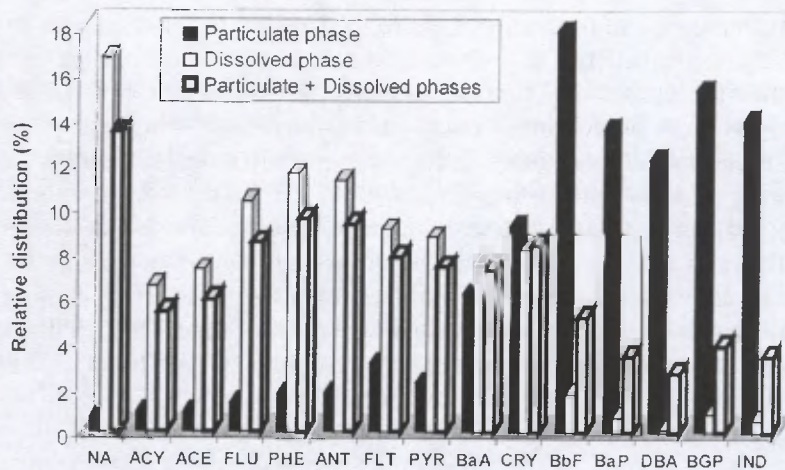


Fig. 6. Relative distribution of PAH compounds in the particulate phase, dissolved phase and particulate + dissolved phases in rain water during the period of study at the rural site. The full terms of abbreviations see in the list of abbreviation

The contribution of an individual PAH compounds to the total PAH concentration in the suspended airborne particulate matter before and after rainfall at the suburban site is graphically presented in Fig. 7. As it can be seen, the distribution of the individual PAH compounds before and after rainfall are very similar, with BbF as the most abundant compound followed by BGP, IND, DBA, BaA, BaP, CRY, FLT, PYR, PHE, FLU, ACE, ANT, ACY, and NA. Such distribution of individual PAH compounds are similar to that found in the particulate phase in rain water at the same site.

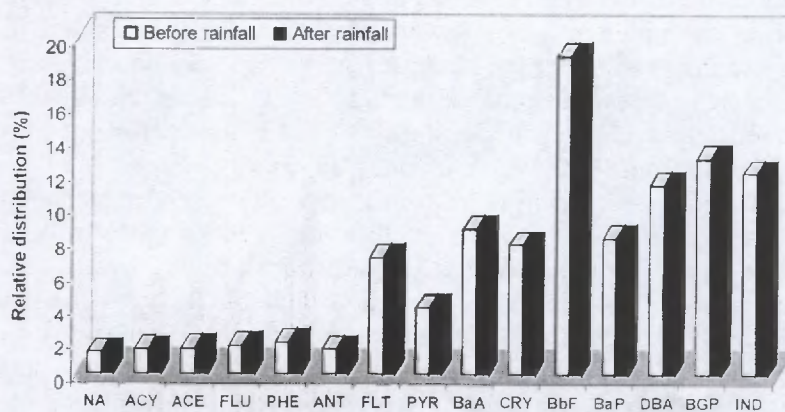


Fig. 7. Relative distribution of PAH compounds in suspended particulate matter in air before and after rainfall at the suburban site. The full terms of abbreviations see in the list of abbreviation

## DISCUSSION

The observed high levels of the PAH compounds in wet deposition in the urban area were due to the high traffic densities which led to an increase of the emission of PAHs. Motor vehicles are the major source of atmospheric PAHs (Oanh et al., 2000; Zheng and Fang, 2000; Wada et al., 2001; Khillare et al., 2005; Chang et al., 2006). In the present study, the total PAHs concentration in wet deposition at Giza was higher than those reported in other cities in the world (Leuenberger et al., 1988; Kiss et al., 1997; Park et al., 2001; Sahu et al., 2004). This may be due to high emission of PAHs from high traffic density in Giza city. Moreover, the low rain volume at Giza city leads to concentration of PAHs in wet deposition. This finding is in agreement with Park et al. (2001) who found an inverse relationship between PAHs concentration and rain volume, indicating a possible dilution effect. More PAHs are removed during the early stage of a rain event and the subsequent rain simply dilutes the initial PAH concentrations (Leister and Baker, 1994; Simcik et al., 1997).

In the present study, the particulate/dissolved concentration ratios were 2.29, 2.14, and 0.26 at the urban, suburban, and rural sites, respectively. These results indicate that PAHs in particle phase were predominating in the atmosphere of both urban and suburban areas, whereas gaseous PAHs were predominating in the air of the rural area. This finding confirms that of Lee et al. (1995) reporting higher carbon content and more PAHs adsorbed in the young aerosol emitted by the automobile exhaust. They added that the PAHs shift from the particle phase to gas phase during the transport process from place to place and this leads to an increase of the total PAHs fraction in the gas phase in the rural area. The particle-bound PAHs were regionally deposited in urban area, whereas more volatile PAHs were transported over long distances (Bucheli et al., 2004).

The highest concentrations of the individual PAH compounds in the suspended airborne particulate matter before rainfall at the suburban site indicate that the wet precipitation is an important factor for the removal of PAH compounds from the atmosphere and the most important processes are wet and dry depositions (Baek et al., 1991), whereas the minor removal process is gas absorption (Buehler et al., 1998). Precipitation accelerates the deposition of fine particles from the atmosphere (Dickhut and Gustafson, 1995; Wild and Jones, 1995).

Traffic emission was the main source of the atmospheric PAH compounds in the urban and suburban areas of Giza. Therefore, the predominance of heavy PAH compounds in rain water indicates that a significant fraction of them was emitted from traffic due to the incomplete combustion of fuels. In the rural area, the PAHs of lower molecular weight (mostly dominant in the gaseous fraction) were predominating in the atmosphere and consequently in wet deposition in it, since it was far from the point sources and there were no significant anthropogenic sources of PAHs in it. Kafr El-Akram is located far from dense traffic; it is not exposed to any direct emission from any adjacent road. Therefore, the PAHs in the atmosphere of Kafr El-Akram were exclusively a result of diffusion and dispersion of PAHs produced by the traffic of the surrounding cities. The time required for the transportation of the PAHs is enough to give rise to significant photochemical reactions caus-



ing a reduction of particle-bound PAHs at Kafr El-Akram. In addition, a part of the PAH mass was shifted from the particle phase to the gas phase during the transport process from the traffic source of the surrounding cities to the rural area. This finding is in agreement with Lee et al. (1995) who reported that the depletion of PAHs in the particle phase may be partly due to photochemical reactions as well as the shift of PAH mass from the particle phase to the gas phase in order to reach thermodynamic equilibrium during the transport process from traffic source to the rural atmosphere.

The carcinogenic PAH compounds have high molecular weight and are especially bound to suspended particulate matter. According to the International Agency for Research on Cancer (IARC, 2001), the total carcinogenic PAH compounds (BaA, BbF, BaP, DBA, and IND) represent 75.10, 76.18, and 63.47% of the total concentration of PAHs in the particulate phase, 24.35, 22.75, and 10.56% of that in the dissolved phase, and 59.68, 59.19, and 21.62% of that in the particulate + dissolved phases in wet deposition of the urban, suburban, and rural areas, respectively. Moreover, the total carcinogenic PAH compounds represent 59.17 and 59.07% of the total concentration of PAHs in suspended particulate matter before and after rainfall at the suburban site, respectively.

In the present study, the distribution of the individual PAH compounds before and after rainfall at the suburban site are very similar. This similarity implies similar emission sources for rain water and suspended particulate matter in the atmosphere of the suburban area in Giza city, in which the vehicle exhaust emissions are the dominant ones.

The distribution of various PAH compounds differs according to their production sources. Their distribution in particulate phase in rain water and suspended particulate matter in the air allows the elucidation of their origin. PAHs distribution patterns in both particulate phase in rain water and suspended particulate in air before and after rainfall are characterized by a dominance of heavy PAHs (four–six aromatic rings) at the urban, suburban, and rural sites (Figs. 4–7). In addition, all the light molecular weight (LMW; two–three aromatic rings)/high molecular weight (HMW; four–six aromatic rings) PAH concentration ratios were below 0.1 in the particulate phase in rain water at all of the three sampling sites and in suspended particulate in air at the suburban area. This result reflects the predominance of HMW PAH compounds and indicates, on the other side, a pyrolytic origin of the PAH pollution. This finding is in agreement with many authors (Zheng and Wan, 1997; Yang et al., 1998; Wang et al., 1999; Soclo et al., 2000; Azimi et al., 2005) reporting that the petrogenic contamination is characterized by the abundance of LMW PAHs, whereas the pyrolytic contamination is characterized by the dominance of HMW PAHs. The presence of CRY and BaA, which are considered as markers of combustion processes, indicates the impact of pyrolytic contamination (Moreda et al., 1998; Soclo et al., 2000).

The PHE/ANT and FLT/PYR concentration ratios can also be used to distinguish between pyrolytic and petrogenic origins. The former is temperature dependent; high temperature processes result in low PHE/ANT values and petrogenic contamination leads to much higher PHE/ANT values (Azimi et al., 2005). Budzinski et al.



(1997) reported PHE/ANT $>10$  as characteristic of petrogenic sources, whereas PHE/ANT $<10$  as that of pyrolytic ones. Moreover, values of the FLT/PYR concentration ratio above unity indicate that the contamination by PAHs is of combustion origin, whereas values below unity indicate a petrogenic origin (Azimi et al., 2005). In the present study, the PHE/ANT concentration ratios were 1.72, 2.00, and 1.00 in particulate phase in rain water in urban, suburban, and rural areas, respectively, and 1.30 in suspended particulate in air before and after rainfall in the suburban area. In addition, the FLT/PYR concentration ratios were 2.3, 2.0, and 1.4 in the particulate phase in the rain water of urban, suburban, and rural areas, respectively, and 1.75 in suspended particulate in air before and after rainfall in the suburban area. These ratios indicate that PAHs in the atmosphere and consequently in the wet deposition at the urban, suburban, and rural sites originate mainly from pyrolytic processes. The pyrolytic origin in the urban and suburban areas of Giza may be linked with the high traffic densities which emit huge quantities of PAHs into the atmosphere. In the rural area, the pyrolytic origin may be produced by the high density traffic of the surrounding cities. This finding is confirmed by the BaP/BGP concentration ratio which was used to investigate the emission sources of PAHs. The BaP/BGP concentration ratio higher than 0.6 indicates the presence of traffic emission (Pandey et al., 1999). In the present study, the BaP/BGP concentration ratios were 0.65, 0.65, and 0.89 in the particulate phase in rain water at urban, suburban, and rural sites, respectively, and 0.63 in the airborne particulate before and after rainfall at the suburban site. These ratios indicate that motor vehicles are the major source of PAH compounds in the atmosphere of the study areas and consequently in the wet deposition.

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