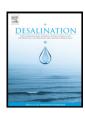


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# Distribution of polycyclic aromatic hydrocarbons in drinking water in Egypt

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#### ABSTRACT

The levels of total PAHs in River Nile water ranged from 1112.7 to 4351.2 ngl<sup>-1</sup>. It is significant that the mean values of the total PAHs found in water samples collected from four sites along the River Nile were among the highest values recorded in other world areas. The obtained results show that PAHs in sediments are within or lower than the residue levels reported in certain other areas. It was found that the PAHs in River Nile were mainly derived from combustion coming from the traffics, heating system and run off. Meanwhile, the sources of pollution include petroleum-derived hydrocarbons which contributed to minor inputs. B(a)P, a carcinogenic PAH, was not detected in treated drinking water samples.

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#### 1. Introduction

The availability of water of acceptable quality in Egypt is limited and getting even more restricted, while at the same time, the needs for water increase as a result of population growth, industrial development and of cultivation desert land. Egyptian industry uses 638 Mm<sup>3</sup> yr<sup>-1</sup> of water, of which 549 Mm<sup>3</sup> yr<sup>-1</sup> are discharged to the drainage system. The River Nile supplies 65% of the industrial water needs and receives more than 57% of its effluents [1]. More than 95% of Egyptian water resources come from River Nile whose quality, however, dramatically worsened in the past few years because of pollution due to growing industrialization.

In Egypt there are no national monitoring program concerning the identification and determination of organic micro pollutants in drinking water resources. Few studies were carried to measure the concentration of pesticide in aquatic environment [1–3]. However, there are no available data concerning the distribution of PAHs in drinking water.

PAHs, in the environment, consist of parent compounds and alkylated homologous. Such compounds (PAHs) are of great environmental concern due to their persistence, bioaccumulation and toxic effects [4–6]. PAH distributions dominated by high molecular weight un-substituted species are mainly generated from combustion of fuels. Meanwhile, PAHs dominated by low molecular weight compounds and alkylated homologous are mainly generated from petrogenic sources [1,7–9].

The aim of this work is to investigate the distribution of some polycyclic aromatic hydrocarbons in drinking water main source and to evaluate the efficiency of conventional drinking water treatment plants in the removal of such compounds.

#### 2. Description of study area

The distribution of PAHs in River Nile system, which forms the main water source of Egypt, was investigated. Four sites were selected to represent different sector in River Nile (Fig. 1). These sites are Beni-Suif (Upper Egypt) and Greater Cairo at the intake of Rod El Farag DWTP. At this location the electrical power station, ship maintenance center and Nile transportation discharged their wastewater into the River. In addition to that site is considered downstream of most Greater Cairo activities, El-Mansoura (Eastern Part of Delta), and Kafr El-Sheikh (Western Part of Delta). Three DWTPs were selected as a case study to evaluate the efficiency of conventional drinking water treatment methods in the removal of PAHs. The first one is located in Upper Egypt and supplied Beni-Suif City with drinking water derived from River Nile. The second DWTP is located at Fowa City (Kafr El-Sheikh Province) and designed to supply 900 ls<sup>-1</sup> treated water. The third one at Meet Fairs (EL-Mansoura Province) derives its raw water supply from a fresh water irrigation canal (Meet Tanah) located in the eastern area of delta of River Nile. Pre-chlorination is applied to the raw at a dose of  $4 \text{ mg l}^{-1}$ , which is followed by addition of alum solution to yield a concentration of 35 mg  $l^{-1}$  of the coagulant. The mean RTs of flocculation and sedimentation were 40 min and 3.5 h, respectively. Post chlorination of treated water amounted to 2 mg l

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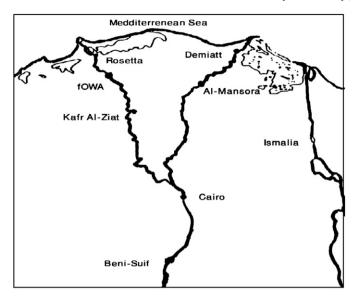


Fig. 1. Sampling sites.

and residual chlorine in the final effluent and distribution system ranged between 0.8 and 1.2 mg  $l^{-1}$ .

#### 3. Materials and methods

#### 3.1. Materials

In this study the sixteen PAHs were analyzed with the USEPA Method SW-846 [10]. They include naphthalene (NaP), acenaphthylene (Acy), acenaphthene (Ace), fluorine (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo[a]anthracene (BaA), chrysene (Chr), benzo[a]pyrene (BaP), benzo[b]fluoranthene (BbF), Benzo[k]fluoranthene (BkF), dibenzo[a,h]anthracene (DahA), indeno [1,2,3-cd]pyrene (IcdP) and benzo[g,h,i]perylene (BghiP) which were purchased from Supleco Company (Germany). All the chemicals and solvents used were pesticide residue grade or equivalent (Merck, Germany).

#### 3.2. Sampling and extraction

A total of 51 samples, including 19 raw water; 16 treated water and 16 sediment samples were collected during March, May, June, August, and October 2005. Raw and treated water grab samples were collected according to APHA (1998) [11] in 2.5 l amber colored glass bottles previously cleaned with hot water, chromic acid, tap water, and acetone and finally rinsed three times with redistilled dichloromethane (CH<sub>2</sub>Cl<sub>2</sub>). The samples were transported in icebox and analysis was performed as soon as possible. If the extraction of the samples was not started within 4–6 h, the samples should be preserved by using 50 ml of CH<sub>2</sub>Cl<sub>2</sub> and stored in dark, in this case the holding time did not exceed 7 days. An adequate volume of water samples (2 l) was acidified with 1.0 M sulfuric acid to pH2 and extricated twice with redistilled CH<sub>2</sub>Cl<sub>2</sub>. The combined extracts were evaporated, dried with anhydrous Na<sub>2</sub>SO<sub>4</sub> and then concentrated to 1 ml by using stream of pure nitrogen.

Surface sediments were collected with stainless steel Van Veen grab samplers operated by hand line. The surface layer (top 1 cm) was carefully removed with spatula. The samples were transported in jars previously cleaned, as previously given. The analysis was carried out according to USEPA Method SW-846 [10]. Sediment was air dried, homogenized in an agate mortar and passed through 1 mm stainless steel sieve. Ten grams of dry sediment was extracted using Soxhlet extractor with 100 ml of CH<sub>2</sub>Cl<sub>2</sub> solvent for 24h at 4–6 cycles. Elemental

sulfur was removed by reaction with activated copper. The sulfur free extract was concentrated to 10 ml by rotary evaporator and then to 1 ml by using gentle steam of pure nitrogen. The extract solvent was exchanged for cyclohexane, and then concentrated to 2 ml for the clean-up procedure.

#### 3.3. Clean up and separation

The extract was cleaned and separated by silica gel column chromatography. Ten grams of 35/70 silica gel in  $CH_2CI_2$  was placed in 10-mm glass column. About 2 g of anhydrous  $Na_2SO_4$  was added to the top of silica gel. The packed materials were washed with 40 ml of hexane at a rate of 2 ml min $^{-1}$ . Discarding the elution and just before exposure of  $Na_2SO_4$  layer to air, the extract was transferred to the column. Twenty ml of redistilled hexane was added and the first fraction was collected and discarded. The second fraction, which contains PAHs, was eluted using 25 ml of  $CH_2CI_2$  in hexane (4:6 v/v). The elution was concentrated to 1 ml and sealed in a vial with screw cap and Teflon silicon desk and preserved at 4 °C till chromatographic analysis was carried out.

#### 3.4. Chromatographic analysis

Identification of PAHs was carried out by using a Varian 3400GC coupled to a Finnigan Mat SSQ 700, equipped with capillary column (30 m  $\times$  0.25 mm and a 0.25  $\mu$ m film thickness fussed silica SE54). The injections were made in split less injector mode with delay time of 3 min and kept at 280 °C; the GC oven was held at 80 °C for 2 min and programmed to rise to 280 °C at a rate of 8 °C min<sup>-1</sup> for 30 min. Helium was used as a carrier gas at a flow rate of 1 ml min<sup>-1</sup>. The transfer line was marinated at 300 °C, and the mass spectrometer was scanned from 40- to 500 a.m.u. every second.

Quantification was carried out by using HP6890 plus GC equipped with FID detector, SE54 capillary column and split less injector mode. The initial column temperature was 80 °C. After an initial hold time for 2 min the temperature was programmed to rise to 280 °C at a rate of 8 °C min $^{-1}$  for 30 min. The injector and detector temperatures were 200 °C and 300 °C, respectively. Helium was used as a carrier gas at a flow rate of 2 ml min $^{-1}$ .

Standard PAHs (Mixture 610-M), which contained 16 PAHs in methanol/methylene chloride (1:1 v/v) was obtained from Supleco Company. Calibration standards, at a minimum of five concentration levels, were prepared.

The quality control program was carried out. This program includes the following:

1- Blank was run with each set of analysis. 2- Quantification of PAHs was carried out using external standards with coefficient for calibration curves higher than 0.995.3- The Calibration program was verified on each working day by the measurement of one or more standards. 4- A random sample was run in triplicate. Laboratory control sample was analyzed with each series of samples (10 samples). Q-chart was used and two values of  $\pm 2$  slandered deviations are the lower and upper limits. The efficiency of the extraction was determined by addition of known concentration of 4, 4' difluorobiphenyl as surrogate standard. Recovery for water samples ranged from 83 to 103%. However for sediment the recovery percent ranged from 80 to 110%

### 4. Results and discussion

#### 4.1. Levels of PAHs in River Nile surface water

Results represented in Table 1 showed that naphthalene(NaP), acenaphthene (Ace), fluorine (Fl), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benzo(a) anthracene [B(a)A], chrysene (Chr), benzo(b) fluoranthene [B(b)F], and benzo(k) fluoranthene [B(k)F],

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