Human Health Assessment of Sixteen Priority Polycyclic Aromatic Hydrocarbons in Contaminated Soils of Northwestern Algeria

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic molecules that contain more than two fused aromatic benzene rings. The majority of PAHs present in the environment are due to incomplete combustion and pyrolysis of organic matter. Recently, toxicity studies of 16 PAHs listed as priority pollutants by the United States Environmental Protection Agency (USEPA) have received considerable attention. These PAHs are of particular interest due to their mutagenic activities as some are classified as probable human carcinogens and others are potentially carcinogenic. Furthermore, benzo[a]pyrene (BaP) has been characterized as having a local and systemic carcinogenic effect, as well as widespread persistence in diverse environmental matrices such as soil, dust, sediments, and water due to its lipophilic and hydrophobic properties. The International Agency for Research on Cancer (IARC) classified benzo(a)pyrene as carcinogenic to humans, along with seven PAHs: Group I, benzo(a)anthracene, dibenzo(a,l)pyrene, and dibenzo(a,h)anthracene as probable carcinogens; and Group II, naphthalene, chrysene, benzo(b)fluoranthene, benzo(j)fluoranthene, benzo(k)fluoranthene and indeno(1,2,3-c,d)pyrene as possible carcinogens. These PAHs have been shown to have toxic effects on human reproductive, developmental, cardiorespiratory, and immune systems. Most of the questions
Research

about the toxicological effects of PAHs on humans are related to their carcinogenesis, mutagenesis, and endocrine disruption. Furthermore, PAH toxicity may have estrogen and antiestrogen effects, which may influence the risk of breast cancer, stomach cancer, dermatitis, gastroenteritis, and pneumonia. The soil system is considered to be a good reservoir of organic contaminants, including PAH compounds, owing to their ubiquity, stability, and long-term enrichment in soils. A high concentration of PAHs has been found in many surface soils around the world, among the numerous environmental matrices (water, dust, soil, plant, and sediments). However, PAH-contaminated surface soil can directly or indirectly expose humans, so it is critical to assess the risk of PAH-contaminated soils to protect human health. Health and cancer risk evaluations focused on various methods of specific exposure criteria and age ranges have been extensively described for populations in various locations around the world, but there are only a few cases of studies recorded for the African community in this context. It is therefore necessary to assess the potentially harmful effects of PAHs in soils in order to inform future remediation decisions.

Polycyclic aromatic hydrocarbons are widespread in the studied area of northwestern Algeria as a result of industrial, commercial and transportation activities, and cereal cultivation.

This research represents the first evaluation of the cancer risk for three population groups: children (2 to 12 years old), adolescents (13 to 20 years old), and adults (over 20 years old) who were exposed to 16 PAHs through contaminated surface soils via three different exposure pathways: ingestion, inhalation, and dermal contact in northwestern Algeria.

Methods

The population of Algeria’s northwestern region is concentrated in five cities (Tlemcen, Sidi Bel Abbes, Ain Temouchent, Mascara, and Oran) (Figure 1), which covers an area of 33,030 km² and have a density of 1,582 inhabitants. With a median age of about 28.5 years, this population accounts for 3.65% of all Algerian residents. The study area has a Mediterranean climate with moderate, wet winters and warm, dry summers, with annual average temperatures ranging from 43°F to 86°F, annual

| Abbreviations |
|---------------|
| **BaPeq** | BaP toxic equivalent concentration |
| **CDI** | Chronic daily intake |
| **EC** | Toxic equivalent quotient |
| **USEPA** | United States Environmental Protection Agency |

Figure 1 — Map of study area showing sampling points and population concentration in Northwestern Algeria
Rainfall ranging from 400 to 670 mm, and an average hourly wind speed of 8.4 to 11.1 miles per hour. In the study area, PAH-contaminated surface soil samples were localized in several locations from different fields: industrial park area surrounding cement factories, as well as urban/rural, residential, cereal agricultural and industrial zones.19,20

Polyaromatic hydrocarbons in soil

Previous studies and published results in this area of northwestern Algeria have reported monitoring data on 16 individual PAHs in surface soils, as seen in Table 1. The concentration of 20 PAHs identified in surface soils from industrial cement, urban, cereal agricultural, and rural sites were 10724, 741, 385, and 246 µg/kg, respectively.19 In addition, concentrations for the Σ16 PAHs ranged from 188.07–2068.04, 246.86–1918.0, and 133.72–249.69 µg/kg for industrial, urban, and agricultural soils, respectively.20 The concentrations of the 16 PAHs detected in surface soil samples from all regions were converted into maximum, minimum, and means concentrations in the present investigation. The present study calculated the health risk to the population in this area from inhalation, ingestion, and dermal contact exposure to soils polluted with...
16 PAHs based on these data (Table 1).

Toxic equivalent concentrations

Benzo[a]pyrene is one of the most potent carcinogenic PAHs. In the present study, the toxicities of PAHs of contaminated surface soil samples were evaluated by calculating the toxicity value of each PAH based on the set of toxicity equivalency factors (TEFs) developed by Nisbet and Lagoy (1992) (Table 1). The total BaP equivalent concentration (BaPeq) was calculated by the sum of BaPeq for each PAH using toxicity equivalent factors, and the toxic equivalent quotient (TEQ) of soil was calculated by collecting the products of each individual PAH content, following Equations 1 and 2:

\[ \text{BaPeq} = \text{Ci} \times \text{TEFi} \]

\[ \text{TEQ} = \sum \text{(PAHi} \times \text{TEFi}) \]

where, BaPeq is a toxic equivalent concentration, Ci is a concentration of PAHi, and TEFI is a toxicity equivalence factor for each PAH.

Risk assessment of polyaromatic hydrocarbon models

In this research, chronic daily intakes (CDIs) associated with PAH exposure in the soil through the three pathways (ingestion, inhalation, and dermal contact) were evaluated by the formulae reported by the USEPA (1991)\textsuperscript{25}, following Equation 3–5:

\[ \text{CDI}_{\text{soil ingestion}} = \frac{\text{CS}_{\text{soil}} \times \text{IR}_{\text{soil}} \times \text{CF} \times \text{EF} \times \text{ED}}{(\text{BW} \times \text{AT})} \]

\[ \text{CDI}_{\text{inhalation}} = \frac{\text{CS}_{\text{soil}} \times \text{ED} \times \text{HR} \times \text{EF}}{(\text{PEF}_{\text{soil}} \times \text{AT} \times \text{BW})} \]

\[ \text{CDI}_{\text{dermat contact}} = \frac{\text{CS}_{\text{soil}} \times \text{CF} \times \text{ED} \times \text{EF} \times \text{AF} \times \text{DAF} \times \text{SA}}{(\text{BW} \times \text{AT})} \]

where CDI\textsubscript{inhalation} is the chronic daily intake related to soil particle ingestion (mg/kg/day). The CDI of PAHs in this medium was not considered as this study only addresses the PAHs carried by soil dust particles. CS\textsubscript{soil} is the concentration of PAHs in soil (mg/kg), IR\textsubscript{soil} is the ingestion rate of soils (mg/day), EF is the exposure frequency (d. year\textsuperscript{-1}), ED denotes exposure duration (years), BW is the bodyweight of the exposed individual (kg), LT is a lifetime (WHO 2006), AT is the average time (days) for lifetime exposure of cancer risk (AT=LTx365), CF is a conversion factor (1. 10\textsuperscript{-6} kg/mg), CDI\textsubscript{inhalation} is the chronic daily intake via inhalation of soil particles (mg kg\textsuperscript{-1} d\textsuperscript{-1}), HR is the air inhalation rate (m\textsuperscript{3}/d), PEF\textsubscript{soil} is the soil particle emission factor, CDI\textsubscript{dermal contact} is the chronic daily intake for dermal contact of soil (mg/kg/d), SA is the skin surface area available for contact soil (cm\textsuperscript{2}/day), AF is the relative skin adherence factor for soil (mg/cm\textsuperscript{2}), and ABS is the dermal absorption fraction. The values of all variables are reported in Table 2.

Human health risk assessment

According to the USEPA’s Exposure Factor Handbook,\textsuperscript{28} the incremental lifetime cancer risk (ILCR) model was used to evaluate the risk to residents exposed to PAHs in soil. The ILCRs for each of the three contact pathways resulting from intake of daily contact products (CDIs) to oral slope factor cancer (CSFs) is shown in Equation 6:

\[ \text{ILCR} = \text{CDI} \times \text{Cancer oral slope factor (CSF)} \]

Equation 6

Where CSF is the carcinogenic slope factor (mg/kg/day). CSF\textsubscript{ingestion}, CSF\textsubscript{dermal}, and CSF\textsubscript{inhalation} of BaP were considered to be 7.3, 25, and 3.85 (mg/kg/day), respectively, based on the cancer-causing ability of BaP.\textsuperscript{29,30}

R represents the total risk of cancer resulting from the sum of the ILCR associated with each exposure contact as follows in Equation 7:

\[ R = \text{ILCR}_{\text{inhalation}} + \text{ILCR}_{\text{dermat}} + \text{ILCR}_{\text{ingestion}} \]

Equation 7

Where Cs is the PAH concentration of soil samples (mg/kg), other variables are shown in Table 1, and the cancer risks for children (2 to 12 years), adolescents (13 to 20 years), and adults (>20 years) were calculated separately.

According to the USEPA and regulatory programs, an ILCR of ≤10\textsuperscript{-6} denotes the level of risk considered acceptable or inconsequential, an ILCR of ≥10\textsuperscript{-4} is considered a serious risk requiring high priority attention, and an ILCR from <10\textsuperscript{-6} to <10\textsuperscript{-4} reflects a potential risk to human health.\textsuperscript{28,31-33} A cancer risk (R) value greater than 10\textsuperscript{-6} is considered unacceptable according to the USEPA.\textsuperscript{34,35}

Results

Only benzo(a)pyrene, from of the 16 PAHs considered in the present study, has toxicity data that can be used to
The carcinogenic risk due to exposure to each PAH was estimated by the toxicity equivalent of BaP (BaPeq). In the present study, the toxicity of PAHs in soil samples from northwest Algeria was evaluated using the TEQs of 16 PAHs and the BaPeq was computed from the soil sample based on the TEF values. The BaPeq and concentrations of 16 PAHs (µg/kg) detected in soil samples are shown in Table 1.

The human health risk is assessed based on possible exposures to PAH-contaminated soil surfaces, either directly or indirectly, and according to various parameters for children, adolescents, and adults such as exposure frequency, exposure duration, body weight, and average life span (Table 2). Intake daily contacts (CDIs) and carcinogenic risks (ILCRs) for each PAH for three age groups (children, adolescents, adults) and exposure pathways (inhalation, ingestion, dermal contact) in the study area were evaluated based on Equation 2 to 6 and the results are reported in Tables 3-5 and Supplemental Material Table S2.

In the present assessment of human health exposure to PAH-contaminated soils, the ILCR was chosen to assess and diagnose the potential cancer risk (R) for different age groups via three pathway exposures, based on the CSF and CDI. The results of ILCRs and R values (min, max and mean) for three population groups: children (2-12 years of age), adolescents (13-20 years of age), adults (>20 years of age). The carcinogenic risk due to exposure to each PAH was estimated by the toxicity equivalent of BaP (BaPeq).

| Definition                          | Units   | Children 2-12 years | Adolescents 13-20 years | Adults >20 years | Reference |
|------------------------------------|---------|---------------------|-------------------------|-----------------|-----------|
| Average body weight                | Kg      | 24.2               | 56.8                    | 69.6            | 26        |
| Unit conversion factor             | Kg/mg   | 1.00x10^6          | 1.00x10^6               | 1.00x10^6       | 25, 28    |
| Exposure frequency                 | Days/years | 350       | 350                     | 350             | 25        |
| Exposure duration                  | Years   | 7                  | 10                      | 41              | 28        |
| Inhalation rate                    | m³/day  | 10                 | 16                      | 16              | 53, 28    |
| Soil ingestion rate                | mg/day  | 200                | 100                     | 100             | 25        |
| Dermal surface exposure            | cm²/day | 2800               | 5700                    | 5700            | 53        |
| Dermal adherence factor            | mg/cm² | 0.2                | 0.2                     | 0.07            | 53        |
| Dermal adsorption fraction         | Unitless| 0.13               | 0.13                    | 0.13            | 53        |
| Lifetime                           | Years   | 72                 | 72                      | 72              | 25        |
| Average life span                  | Days    | 26 280             | 26 280                  | 26 280          | 53        |
| Particulate emission factor        | m³/kg   | 1.36x10⁹           | 1.36 x10⁹               | 1.36 x10⁹      | 53        |
| Cancer slope factor ingestion      | mg/kg/day | 7.3         | 7.3                     | 7.3             | 29        |
| Cancer slope factor dermal         | mg/kg/day | 25           | 25                      | 25              | 29        |
| Cancer slope factor inhalation     | mg/kg/day | 3.85         | 3.85                    | 3.85            | 29        |

Table 2 — Exposure Variables for the Health Risk Assessment for Children, Adolescents and Adults via Ingestion, Inhalation and Dermal Exposure Pathways
years of age), and adults (>20 years of age), via different exposure pathways (inhalation, ingestion, and dermal contact) are presented in Tables 3–6.

The average values of total ILCRs calculated for the study area indicate that risk levels via dermal contact for children, adolescents, and adults ranged from $1.37 \times 10^{-5}$ to $1.99 \times 10^{-5}$ (Table 3), while the risk levels of cancer via ingestion ranged from $3.35 \times 10^{-6}$ to $1.12 \times 10^{-5}$ (Table 4) and inhalation exposures ranged from $2.08 \times 10^{-10}$ to $6.96 \times 10^{-10}$ (Table 5). In addition, the cancer risk level for adults was $3.12 \times 10^{-5}$, $2.48 \times 10^{-5}$ for children, and $2.04 \times 10^{-5}$ for adolescents (Table 6).

### Discussion

The present study revealed that BaPeq values for 16 PAHs in the study area averaged $280.28$ g/kg, which is less than $300$ g/kg and much lower than the Canadian Soil Quality Guideline recommendation of TEQ < 600 g/kg. In addition, the TEQ values also reflect the relative carcinogenic potential of all 16 PAHs to BaP in soil samples (Table 1).

The contribution of BaPeq for each PAH to total TEQ estimates were in the following order: benzo[a]pyrene (BaP) (38.6%) > dibenzo(a,h)anthracene (38.3%) > benzo[a]anthracene > benzo[b]fluoranthene > benzo[g,h,i]perylene. However, the BaPeq levels for surface soils in the study area exceeded the Dutch target value of 33 g. kg$^{-1}$. In addition, the TEQ values also reflect the relative carcinogenic potential of all 16 PAHs to BaP in soil samples (Table 1).

| Individual PAH | ILCR Dermal contact |
|----------------|---------------------|
|                | Children | Adolescents | Adults |
| Naphthalene    | $1.33 \times 10^{-7}$ | $1.65 \times 10^{-7}$ | $1.93 \times 10^{-7}$ |
| Acenaphthylene | $1.86 \times 10^{-7}$ | $2.31 \times 10^{-7}$ | $2.70 \times 10^{-7}$ |
| Acenaphthene   | $3.74 \times 10^{-8}$ | $4.63 \times 10^{-8}$ | $5.42 \times 10^{-8}$ |
| Fluorene       | $3.75 \times 10^{-8}$ | $4.64 \times 10^{-8}$ | $5.44 \times 10^{-8}$ |
| Phenanthrene   | $6.91 \times 10^{-7}$ | $8.56 \times 10^{-7}$ | $1.00 \times 10^{-6}$ |
| Anthracene     | $2.85 \times 10^{-7}$ | $3.53 \times 10^{-7}$ | $4.13 \times 10^{-7}$ |
| Fluoranthene   | $2.84 \times 10^{-6}$ | $3.52 \times 10^{-6}$ | $4.13 \times 10^{-6}$ |
| Pyrene         | $2.84 \times 10^{-6}$ | $3.52 \times 10^{-6}$ | $4.13 \times 10^{-6}$ |
| Benzo[a]anthracene | $2.08 \times 10^{-6}$ | $2.57 \times 10^{-6}$ | $3.01 \times 10^{-6}$ |
| Chrysene       | $5.90 \times 10^{-7}$ | $7.31 \times 10^{-7}$ | $8.56 \times 10^{-7}$ |
| Benzo[b]fluoranthene | $1.18 \times 10^{-6}$ | $1.46 \times 10^{-6}$ | $1.71 \times 10^{-6}$ |
| Benzo[k]fluoranthene | $6.30 \times 10^{-7}$ | $7.80 \times 10^{-7}$ | $9.14 \times 10^{-7}$ |
| Benzo[a]pyrene (BaP) | $7.91 \times 10^{-7}$ | $9.81 \times 10^{-7}$ | $1.15 \times 10^{-6}$ |
| Indeno(1,2,3-c,d)pyrene | $6.42 \times 10^{-7}$ | $7.96 \times 10^{-7}$ | $9.32 \times 10^{-7}$ |
| Dibenz(a,h)anthracene | $1.57 \times 10^{-7}$ | $1.95 \times 10^{-7}$ | $2.28 \times 10^{-7}$ |
| Benzo(g,h,i)perylene | $6.14 \times 10^{-7}$ | $7.61 \times 10^{-7}$ | $8.91 \times 10^{-7}$ |
| $\sum 2$-3-ring | $1.37 \times 10^{-6}$ | $1.70 \times 10^{-6}$ | $1.99 \times 10^{-6}$ |
| $\sum 4$ring | $8.35 \times 10^{-6}$ | $1.04 \times 10^{-5}$ | $1.21 \times 10^{-5}$ |
| $\sum 5$ring | $2.76 \times 10^{-6}$ | $3.42 \times 10^{-6}$ | $4.00 \times 10^{-6}$ |
| $\sum 6$ring | $1.26 \times 10^{-6}$ | $1.56 \times 10^{-6}$ | $1.82 \times 10^{-6}$ |
| $\sum 16$ PAHs | $1.37 \times 10^{-5}$ | $1.70 \times 10^{-5}$ | $1.99 \times 10^{-5}$ |

Table 3 — Incremental Lifetime Cancer Risk (ILCR) for Human Exposure to Individual Polyaromatic Hydrocarbons (PAHs) via Dermal Contact Exposure to Surface Soils in the Study Area.
Table 4— Incremental Lifetime Cancer Risk (ILRC) for Human Exposure to Individual Polyaromatic Hydrocarbons (PAHs) via Ingestion to Surface Soils in the Study Area

| Individual PAH                  | Children          | Adolescents       | Adults           |
|---------------------------------|-------------------|-------------------|------------------|
| Naphthalene                     | $1.07 \times 10^{-7}$ | $3.24 \times 10^{-8}$ | $1.08 \times 10^{-7}$ |
| Acenaphthylene                  | $1.50 \times 10^{-7}$ | $4.55 \times 10^{-8}$ | $1.52 \times 10^{-7}$ |
| Acenaphthene                    | $3.00 \times 10^{-8}$ | $9.12 \times 10^{-9}$ | $3.05 \times 10^{-8}$ |
| Fluorene                        | $3.01 \times 10^{-8}$ | $9.15 \times 10^{-9}$ | $3.06 \times 10^{-8}$ |
| Phenanthrene                    | $5.54 \times 10^{-7}$ | $1.69 \times 10^{-7}$ | $5.65 \times 10^{-7}$ |
| Anthracene                      | $2.29 \times 10^{-7}$ | $6.96 \times 10^{-8}$ | $2.33 \times 10^{-7}$ |
| Fluoranthene                    | $2.28 \times 10^{-6}$ | $6.94 \times 10^{-7}$ | $2.32 \times 10^{-6}$ |
| Pyrene                          | $2.28 \times 10^{-6}$ | $6.94 \times 10^{-7}$ | $2.32 \times 10^{-6}$ |
| Benzo[a]anthracene              | $1.67 \times 10^{-6}$ | $5.07 \times 10^{-7}$ | $1.70 \times 10^{-6}$ |
| Chrysene                        | $4.73 \times 10^{-7}$ | $1.44 \times 10^{-7}$ | $4.82 \times 10^{-7}$ |
| Benzo[b]fluoranthene            | $9.48 \times 10^{-7}$ | $2.88 \times 10^{-7}$ | $9.65 \times 10^{-7}$ |
| Benzo[k]fluoranthene            | $5.05 \times 10^{-7}$ | $1.54 \times 10^{-7}$ | $5.14 \times 10^{-7}$ |
| Benzo[a]pyrene (BaP)            | $6.35 \times 10^{-7}$ | $1.93 \times 10^{-7}$ | $6.46 \times 10^{-7}$ |
| Indeno(1,2,3-c,d)pyrene         | $5.15 \times 10^{-7}$ | $1.57 \times 10^{-7}$ | $5.25 \times 10^{-7}$ |
| Dibenzo(a,h)anthracene          | $1.26 \times 10^{-7}$ | $3.84 \times 10^{-8}$ | $1.28 \times 10^{-7}$ |
| Benzo(g,h,i)pyrene             | $4.93 \times 10^{-7}$ | $1.50 \times 10^{-7}$ | $5.02 \times 10^{-7}$ |
| $\Sigma$2-3-ring                | $1.10 \times 10^{-6}$ | $3.34 \times 10^{-7}$ | $1.12 \times 10^{-6}$ |
| $\Sigma$4-ring                 | $6.70 \times 10^{-6}$ | $2.04 \times 10^{-6}$ | $6.82 \times 10^{-6}$ |
| $\Sigma$5-ring                 | $2.21 \times 10^{-6}$ | $6.74 \times 10^{-7}$ | $2.25 \times 10^{-6}$ |
| $\Sigma$6-ring                 | $1.01 \times 10^{-6}$ | $3.07 \times 10^{-7}$ | $1.03 \times 10^{-6}$ |
| $\Sigma$16 PAHs                | $1.10 \times 10^{-5}$ | $3.35 \times 10^{-6}$ | $1.12 \times 10^{-5}$ |

Other studies have indicated that BaP has a similar influence on TEQ.\textsuperscript{39,42-44} This further confirms the importance of BaP as an indicator of overall PAH carcinogenicity. The average BaPeq value for the seven carcinogenic PAHs (BaPeq$\Sigma_{7PAHs_{Carcin}}$) contributes more than 96.5% of the total $\Sigma$16 BaPeq level in the present study, explaining why the seven carcinogenic PAHs are the most toxic of the 16 PAH total components.

The results of TEQ in northwestern Algerian surface soils (Table 1) varied between 14.81 to 1038.35 μg/kg, with an arithmetic mean of 280.28 μg/kg, which is lower than those in soils from regions of Kucming, Shenfu, and Beijing, China (100–3400 μg/kg)\textsuperscript{45,46} and Lagos, Nigeria (523–1046 μg/kg),\textsuperscript{38} but higher than reported in Delhi, India (4.39–717.06 μg/kg, mean of 131.46 μg/kg),\textsuperscript{47} Zhongyuan oilfield, (9.10–75.83 μg/kg),\textsuperscript{48} Beijing, China (39.4–559.5 μg/kg),\textsuperscript{49} and Isola Delle...
To date, no environmental guidelines have yet been established in Algeria for PAH-contaminated soils. According to most regulatory programs and guidelines around the world, an ILCR less than or equal to $10^{-6}$ denote a negligible risk, ILCRs of $10^{-6}$ to $10^{-4}$ indicate potential risk, and ILCRs greater than $10^{-4}$ indicate potential high risk.\textsuperscript{25,51,52}

The results of the ILCR carcinogenic risk of individual PAH surface soil (Tables 3–5) polluted with fluoranthene, pyrene, benzo[b]fluoranthene, and benzo[a]anthracene contributes substantially to the potential cancer risk (ILCR $\geq 10^{-6}$) of children and adults by ingestion and dermal contact exposures, and these compounds also contribute to BaPeq and increasing CDIs among the 16 PAHs.

Femmine, Italy (3.3–69 μg/kg).\textsuperscript{39}

### Table 5 — Incremental Lifetime Cancer Risk (ILRC) for Human Exposure to Individual Polycyclic Aromatic Hydrocarbons (PAHs) via Inhalation Exposure to Surface Soils in the Study

| Individual PAH          | ILCR Carcinogenic risk Inhalation |
|-------------------------|----------------------------------|
|                         | Children | Adolescents | Adults     |
| Naphthalene             | $2.07 \times 10^{-12}$ | $2.01 \times 10^{-12}$ | $6.73 \times 10^{-12}$ |
| Acenaphthylene          | $2.90 \times 10^{-12}$ | $2.82 \times 10^{-12}$ | $9.45 \times 10^{-12}$ |
| Acenaphthene            | $5.81 \times 10^{-13}$ | $5.66 \times 10^{-13}$ | $1.89 \times 10^{-12}$ |
| Fluorene                | $5.83 \times 10^{-13}$ | $5.68 \times 10^{-13}$ | $1.90 \times 10^{-12}$ |
| Phenanthrene            | $1.08 \times 10^{-11}$ | $1.05 \times 10^{-11}$ | $3.50 \times 10^{-11}$ |
| Anthracene              | $4.43 \times 10^{-12}$ | $4.32 \times 10^{-12}$ | $1.44 \times 10^{-11}$ |
| Fluoranthene            | $4.42 \times 10^{-11}$ | $4.31 \times 10^{-11}$ | $1.44 \times 10^{-10}$ |
| Pyrene                  | $4.42 \times 10^{-11}$ | $4.31 \times 10^{-11}$ | $1.44 \times 10^{-10}$ |
| Benzo[a]anthracene      | $3.23 \times 10^{-11}$ | $3.14 \times 10^{-11}$ | $1.05 \times 10^{-10}$ |
| Chrysene                | $9.18 \times 10^{-12}$ | $8.94 \times 10^{-12}$ | $2.99 \times 10^{-11}$ |
| Benzo[b]fluoranthene    | $1.84 \times 10^{-11}$ | $1.79 \times 10^{-11}$ | $5.99 \times 10^{-11}$ |
| Benzo[k]fluoranthene    | $9.80 \times 10^{-12}$ | $9.54 \times 10^{-12}$ | $3.19 \times 10^{-11}$ |
| Benzo[a]pyrene (BaP)    | $1.23 \times 10^{-11}$ | $1.20 \times 10^{-11}$ | $4.01 \times 10^{-11}$ |
| Indeno(1,2,3-c,d)pyrene  | $9.99 \times 10^{-12}$ | $9.73 \times 10^{-12}$ | $3.26 \times 10^{-11}$ |
| Dibenzo(a,h)anthracene  | $2.44 \times 10^{-12}$ | $2.38 \times 10^{-12}$ | $7.97 \times 10^{-12}$ |
| Benzo(g,h,i)perylene    | $9.55 \times 10^{-12}$ | $9.30 \times 10^{-12}$ | $3.11 \times 10^{-11}$ |
| $\Sigma 2$-3-ring       | $2.13 \times 10^{-11}$ | $2.08 \times 10^{-11}$ | $6.94 \times 10^{-11}$ |
| $\Sigma 4$ring          | $1.30 \times 10^{-10}$ | $1.27 \times 10^{-10}$ | $4.23 \times 10^{-10}$ |
| $\Sigma 5$ring          | $4.29 \times 10^{-11}$ | $4.18 \times 10^{-11}$ | $1.40 \times 10^{-10}$ |
| $\Sigma 6$ring          | $1.95 \times 10^{-11}$ | $1.90 \times 10^{-11}$ | $6.37 \times 10^{-11}$ |
| $\Sigma 16$ PAHs        | $2.14 \times 10^{-10}$ | $2.08 \times 10^{-10}$ | $6.96 \times 10^{-10}$ |
potential cancer risk (ILCR ≥ 10^{-6}) of adolescents via dermal contact.

In the present study, the high molecular weight polycyclic aromatic hydrocarbons (HMW PAHs) with four rings contributed the most to the ILCRs for all age groups (Table 3-5). The 5- and 6-ring HMW PAHs such as benzo[a]pyrene (BaP), benzo[b]fluoranthene, benzo[k]fluoranthene, indeno(1,2,3-c,d)pyrene, and benzo(g,h,i)perylene have a moderate contribution to the cancer risk for adults and children. The remaining 2- and 3-ring PAHs with ILCRs lower than 10^{-6} are classified as lower cancer risk for humans, according to USEPA standards, where the acceptable risk of cancer from individual PAHs is (10^{-6} – 10^{-4}).

In addition, the risks from individual PAH exposure pathways were summarized to determine the key exposure pathway for each PAH compound contaminant surface soil in the study area. The results presented in Tables 3–5 clearly show a large difference between cancer risks for all 16 PAHs detected in soils via the inhalation route with a magnitude of (10^{-13}~10^{-10}), and the dermal contact and ingestion routes with a magnitude of (10^{-8}~10^{-6}) and (10^{-9}~10^{-6}), respectively. The results indicate that the dermal contact and ingestion pathways greatly contribute to the carcinogenic risk of human exposure to each PAH. Thus, higher dermal exposure contact can be explained by the emission of PAHs during traffic road, industrial and agricultural activities, which can be transferred to the surface of tools and clothing to the skin.

In summary, the carcinogenicity of each PAH depends on its chemical structure and physical-chemical properties. The HMW PAHs are more potent carcinogens, and many studies have reported that exposure to HMW PAHs is linked to increased incidences of leukemia, bone, brain, bladder and scrotal cancers, and adverse pregnancy outcomes.

Cancer risk

In the present study, the cancer risk (R) of a total of 16 PAHs for children, adolescents, and adults had means
of 2.48 x 10^{-5}, 2.04 x 10^{-5}, and 3.12 x 10^{-5} mg/kg/d, respectively (Table 6). These values are in the unacceptable range (10^{-5}~10^{-4}) an indicate a potential cancer risk to human health. In addition, the results showed that the ILCRs through the ingestion and dermal contact pathways ranged from 10^{-7} and 10^{-5} in soil samples, while the cancer risk through inhalation was 10^{-12} to 10^{-9}, about 10^4 to 10^5 times lower than that through ingestion and dermal exposures. As shown in Table 6, the average values of total ILCRs calculated for the study area were similar to results found by Zha et al. (2018) in Nanjing, China.10

For children and adults, the cancer risk levels via dermal contact were in the same order of magnitude (10^{-5}), showing that both dermal contact and ingestion had a significant contribution to the cancer risk in children and adults. Nevertheless, the risk of ingestion for adolescents (< 10^4) was lower than for children and adults, because children are more likely to be exposed to environmental pollutants and considered the most sensitive group age with frequent hand-to-mouth or object-to-mouth activities which facilitates ingestion of contaminated soils.56 For adults, a longer exposure time to the indoor/ outdoor environment, greater body weight and greater skin surface area can also lead to a higher potential cancer risk.57 In the present study, the cancer risk for adults via dermal contact (1.99 x 10^{-9}) was higher than for children and adolescents. This result is similar to the human cancer risk resulting from PAH exposure to surface soil in China cities (Beijing, Guangzhou, and Lanzhou), due to the larger dermal exposure with a long exposure period for adults.34 However, for adults, although the ingestion rate for soil (IRsoil) was relatively small, the longer exposure time may result in increased cancer risk, where can be similar to the risk of soil ingestion for children.

For adolescents, (13 to 20 years), the ILCRs for adolescents were lower due to lower soil ingestion rates and shorter exposure time. As shown in Table 3, the risk of cancer in children exposed to PAHs via dermal contact was significantly higher than that reported for adolescents. Furthermore, the risk of cancer in children from ingestion was equivalent to that of adults but higher than that of adolescents (Table 4).

Based on national and international regulatory agencies’ delineated ILCR values, ≤ 10^{-6}, 10^{-6} to 10^{-4}, and >10^{-4} correspond to negligible risk, potential risk, and high risk of cancer, respectively.14,58 The ILCRs reported in the present study for children, adolescents, and adults due to PAH exposure (Table 6) were comparable with the ILCRs reported for various African locations like Warri city, Nigeria (children, 3.07 x 10^{-5}; adults, 2.3 x 10^{-5}),59 Kumasi city, Ghana (children, 8.5 x 10^{-6}; adults 2.1 x 10^{-4}),60 which indicate potential cancer risk. Values in the present study were lower than in Lagos, Nigeria (children, 6.66 x 10^{-5}; adults, 5.11 x 10^{-5}), Tamale, northern Ghana (children, 9.26 x 10^{-5}; adults 1.02),60 dust from Warri city Nigeria (children, 3.11 x 10^{-5}; adults, 1.49 x 10^{-4}),62 and Delta state, Nigeria (children, 3.34 x 10^{-5}; adults, 2.56 x 10^{-5}),63 where a high risk of cancer was observed. Furthermore, the ILCRs were also comparable with those reported for locations in other countries such as Delhi-Kolkata, India (adults, 6.92 x 10^{-5}; children 6.22 x 10^{-5}) and Dhanbad, India (adults, 1.82 x 10^{-5}; children 1.85 x 10^{-5}).64 The present results were lower than values reported in Mahshahr, Iran (adults 1.20 x 10^{-4}; children 1.56 x 10^{-4}),65 Guangzhou, China (adults, 2.08 x 10^{-4} – 1.13 x 10^{-5}; children, 2.26 x 10^{-5} – 1.23 x 10^{-5}), and higher than in Kobra, India (adults, 5.6 x 10^{-8}; children, 2.9 x 10^{-7}).66

The R level for adults in the present study was higher than for children, while adolescents had the lowest risk (Table 6). Similar results have been reported in surface soils from the Tianjin coastal new region in China by Rongguang Shi et al. (2020),66 possibly due to fact that IRsoil and EF in children and adults were higher than in adolescents.

In addition, the R for children and adults in the study area was greater than the acceptable value of 1 x 10^{-6}, indicating that the study area has the potential for human carcinogenic risk. Compared to other studies, the obtained results show that the total R values were in agreement with those reported in contaminated soils from Hong Kong metropolitan soils and urban soil from Nigeria.59,67

In general, preliminary health assessment results from the study area indicate that PAH-contaminated soils present a cancer risk to the resident population via cumulative ingestion and through dermal contact routes. The R for the three age groups in the present study indicate a potential health risk for adults and children, indicating that special attention should be paid to the health risks of children.

**Study limitations**

Benarba et al. found a high incidence of liver cancer in the province, the highest in Algeria, suggesting a high prevalence of risk factors.54 Therefore, evaluation of PAH levels in soil and air, as well as other organic pollutants in specific sites with diverse activities (industrial, urban/residential) is recommended to better understand the potential effects on human health in this area.
Conclusions

The present study represents the first health risk assessment from PAH-contaminated soils in northwestern Algeria. Total cancer risks of 16 PAHs for children, adolescents and adults were 2.48 x 10^{-5}, 2.04 x 10^{-5} and 3.12 x 10^{-5} mg/kg/d. The risk of cancer for adolescents was within the acceptable range, with no potential risk of cancer, but there was a higher risk of moderate to severe cancer in children and adults in the study area. Among different exposure pathways, the observed cancer risks through dermal contact had the highest contribution to total cancer risks in the study area. Further research should focus on risk assessment of PAHs in specific areas as agricultural, residential/urban, and industrial, and should include exposure through air.

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References

1. Abdel-Shafy HI, Mansour MS. A review on polycyclic aromatic hydrocarbons: source, environmental impact, effect on human health and remediation. Egypt J Petr. 2016;25(1):107-123. https://doi.org/10.1016/j.ejpe.2015.03.011
2. Dat N-D, Chang MB. Review on characteristics of PAHs in atmosphere, anthropogenic sources and control technologies. Sci Total Environ. 2017;609:682-693. https://doi.org/10.1016/j.scitotenv.2017.07.204
3. Olayinka OO, Adewusi AA., Olarenwaju OO, Aladesida A A. Concentration of polycyclic aromatic hydrocarbons and estimated human health risk of water samples around Atlas Cove, Lagos, Nigeria. J Health Pollution. 2018; 8(20): 181210. https://doi.org/10.5969/j.2156-9614-8.20.181210
4. Keith LH. The source of US EPA’s sixteen PAH priority pollutants. Polycycl Aromat Compd. 2015;35(2):147-160. https://doi.org/10.1080/10406638.2014.892886
5. Szopińska M, Szumińska D, Białik RJ et al. Determination of polycyclic aromatic hydrocarbons (PAHs) and other organic pollutants in freshwaters on the western shore of Admiralty Bay (King George Island, Maritime Antarctica). Environ Sci Pollut Res. 2019;26(18):18143-18161. https://doi.org/10.1007/s11356-019-05945-w
6. Zango ZU, Sambudi NS, Jumbri K et al. An Overview and Evaluation of Highly Porous Adsorbent Materials for Polycyclic Aromatic Hydrocarbons and Phenols Removal from Wastewater. Water. 2020;12(10):2921. https://doi.org/10.3390/w12102921
7. On the Evaluation IWG. BENZO [a] PYRENE. In: Chemical Agents and Related Occupations. IARC Monogr Eval Carcinog Risks Hum. 2012.
8. Hamid N, Syed JH, Kamal A et al. A review on the abundance, distribution and ecobiological risks of PAHs in the key environmental matrices of South Asia. Rev Environ Contam Toxicol. 2016;240:1-30. https://doi.org/10.1007/978-3-319-5007
9. Cancer IARC. Some non-heterocyclic polycyclic aromatic hydrocarbons and some related exposures. IARC Press, IARC Monogr Eval Carcinog Risks Hum. 2010.
10. Sansom GT, Kirsch KR, Casillas GA, Camargo K, Wade TL, Knap AH, Horney JA. (2021). Spatial Distribution of Polycyclic Aromatic Hydrocarbon Contaminants after Hurricane Harvey in a Houston Neighborhood. J Health Pollution 2021; 11(29):210308. https://doi.org/10.5969/j.2156-9614-11.29.210308
11. Kumar B, Verma V, Sharma C et al. Estimation of toxicity equivalency and probabilistic health risk on lifetime daily intake of polycyclic aromatic hydrocarbons from urban residential soils. Hum Ecol Risk Assess. 2015;21(2):434-444. https://doi.org/10.1080/10807039.2014.921530
12. Osiyemi, O, Abiodun-Solanke AJ, Mangai E, Okeke E, Jahnezim B. Comparison of Polyaromatic Hydrocarbon Residue Concentrations in Clarias gariepinus Smoked with Traditional and Mechanical Kilns. J Health Pollution. 2020; 10 (28): 201215. doi: https://doi.org/10.5696/2156-9614-10.28.201215
13. Nam JY, Sweetman AJ, Jones KC. Polynuclear aromatic hydrocarbons (PAHs) in global background soils. J Environ Monit. 2009;11(1):45-48. https://doi.org/10.1039/b813841a
14. Chen S-C, Liao C-M. Health risk assessment on human exposed to environmental polynuclear aromatic hydrocarbons pollution sources. Sci Total Environ. 2006;366(1):112-123. https://doi.org/10.1016/j.scitotenv.2005.08.047
15. Gope M, Masto RE, George J et al. Exposure and cancer risk assessment of polycyclic aromatic hydrocarbons (PAHs) in the street dust of Anchal city, India. Sustain Cities Soc. 2018;38:616-626. https://doi.org/10.1016/j.scs.2018.01.006
16. Qu C, Li B, Hu H et al. Multi-pathway assessment of human health risk posed by polycyclic aromatic hydrocarbons. Environ Geochem Health. 2015;37(3):587-601. https://doi.org/10.1007/s10653-014-9675-7
17. Chen D, Feng Q, Liang H et al. Distribution characteristics and ecological risk assessment of polycyclic aromatic hydrocarbons (PAHs) in underground coal mining environment of Xuzhou. Hum Ecol Risk Assess. 2019;25(6):1564-1578. https://doi.org/10.1080/10807039.2018.1489715
18. Koblis K, Day C, Heath JS. Impact of surrogate selection on risk assessment for total petroleum hydrocarbons. Soil Sediment Contam. 1993;2(2):125-136. https://doi.org/10.1080/15320389309383433
19. Mebarka DH, Taleb S, Benghalem A et al. Residue analysis of some PAHs in some Algerian soil: a preliminary environmental impact assessment. Energy Procedia. 2012;18:1125-1134. https://doi.org/10.1016/j.egypro.2012.05.127
20. Halfadji A, Touabet A, Portet-Koltalo E et al. Concentrations and source identification of polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) in agricultural, urban/residential, and industrial soils, east of Oran (Northwest Algeria). Polycycl Aromat Compd. 2019;39(4):299-310. https://doi.org/10.1007/s10806-017-1326947
21. Population Statistics. Algeria. 2021 Jan 27; Accessed [2021 April 20], Available from http://citypopulation.de/Algeria-Cities.html
22. Meddi M, Meddi H, Toumi S et al. Regionalization of rainfall in north-western Algeria. Geographia Technica. 2013;17:56-69.
23. Nisbet IG, Lagoy PK. Toxic equivalency factors (TEFs) for polycyclic aromatic hydrocarbons (PAHs). Regul Toxicol Pharmacol. 1992;16(3):290-300. https://doi.org/10.1016/0034-673X(92)80030-T
Contaminants of Concern. USA, New York: The US Department of Health and Human Services. 2007.
34. Wang W, Huang M-J, Kang Y et al. Polycyclic aromatic hydrocarbons (PAHs) in urban surface dust of Guangzhou, China: Status, sources and human health risk assessment. Sci Total Environ. 2011;409(21):4519-4527. https://doi.org/10.1016/j.scitotenv.2011.07.030
35. Yang W, Lang X, Li G. Cancer risk of polycyclic aromatic hydrocarbons (PAHs) in the soils from Jiaozhou Bay wetland. Chemosphere. 2014;112:289-295. https://doi.org/10.1016/j.chemosphere.2014.04.074
36. Coatu V, Oros A, Tiganus D et al. Assessment of chemical contamination in biota from Romanian marine waters in respect with maximum admissible levels regulated by legislation for human consumption. J Environ Protect Ecol. 2015;16:117-125.
37. Environment CCoMot. Canadian Soil Quality Guidelines for Carcinogenic and Other Polycyclic Aromatic Hydrocarbons (PAHS): Environmental and Human Health Effects Scientific Supporting Document. Canadian Council of Ministers of the Environment. 2008.
38. Elghobor MJ, Iwegbue CM, Eguavoen OI et al. Occurrence, sources and ecological and human health risks of polycyclic aromatic hydrocarbons in soils from some functional areas of the Nigerian megacity, Lagos. Environ Geochem Health. 2020;42(9):2895-2923. https://doi.org/10.1007/s10653-020-00528-z
39. Devi NL, Yadav IC, Shihua Q et al. Environmental carcinogenic polycyclic aromatic hydrocarbons in soil from Himalayas, India: Implications for spatial distribution, sources apportionment and risk assessment. Chemosphere. 2016;144:493-502. https://doi.org/10.1016/j.chemosphere.2015.08.062
40. Kumar B, Verma VK, Kumar S et al. Polycyclic aromatic hydrocarbons in residential soils from an Indian city near power plants area and assessment of health risk for human population. Polycycl Aromat Compd. 2014;34(3):191-213. https://doi.org/10.1080/01406638.2014.883414
41. Li G, Lang Y, Yang W et al. Source contributions of PAHs and toxicity in reed wetland soils 493 of Liaohe estuary using a CMB-TEQ method. Sci Total Environ. 2014;490:199-204. https://doi.org/10.1016/j.scitotenv.2014.05.001
42. Enuneku A, Ozekeke O, Okpara B et al. Ingestion and Dermal 494 Cancer Risk Via Exposure to Polycyclic Aromatic Hydrocarbons (PAHs)
52. Chen P, Liang J. Polycyclic aromatic hydrocarbons in green space soils in Shanghai: source, distribution, and risk assessment. J Soils Sediments. 2021;21(2):967-977. https://doi.org/10.1007/s11368-020-02838-2

53. United States Environmental Protection Agency (USEPA). Risk assessment guidance for superfund. Volume I: human health evaluation manual (Part A and Part E supplemental guidance for dermal risk assessment). EPA/540/1-89/002 Washington DC, USA E: Office of Emergency and Remedial Response. 2001

54. Srogi K. Monitoring of environmental exposure to polycyclic aromatic hydrocarbons: a review. Environ Chem Lett. 2007;5(4):169-195. https://doi.org/10.1007/s10311-007-0095-0

55. Nieuwoudt C, Pieters R, Quinn LP et al. Polycyclic aromatic hydrocarbons (PAHs) in soil and sediment from industrial, residential, and agricultural areas in central South Africa: An initial assessment. J Soils Sediments. 2011;20(2):188-204. https://doi.org/10.1080/15320383.2011.546443

56. Guney M, Zagury GJ. Bioaccessibility and other key parameters in assessing oral exposure PAH-contaminated soils and dust: A critical review. Hum Ecol Risk Assess. 2016;22(6):1396-1417. https://doi.org/10.1080/10807039.2016.1185691

57. Moore F, Akhbarizadeh R, Keshavarzi B et al. Ecotoxicological risk of polycyclic aromatic hydrocarbons (PAHs) in urban soil of Isfahan metropolis, Iran. Environ Monit Assess. 2015;187(4):1396-1417. https://doi.org/10.1007/s10661-015-4433-6

58. Wei C, Han Y, Bandowe BAM et al. Occurrence, gas/particle partitioning and carcinogenic risk of polycyclic aromatic hydrocarbons and their oxygen and nitrogen containing derivatives in Xi’an, central China. Sci Total Environ. 2015;505:814-822. https://doi.org/10.1016/j.scitotenv.2014.10.054

59. Iwegbue CM, Obi G, Aganbi E et al. Concentrations and health risk assessment of polycyclic aromatic hydrocarbons in soils of an urban environment in the Niger Delta, Nigeria. Toxicol Environ Health Sci. 2016;8(3):221-233. https://doi.org/10.1007/s13530-016-0279-8

60. Bandowe BAM, Nkansah MA. Occurrence, distribution and health risk from polycyclic aromatic compounds (PAHs, oxygenated-PAHs and azaarenes) in street dust from a major West African Metropolis. Sci Total Environ. 2016;553:439-449. https://doi.org/10.1016/j.scitotenv.2016.02.142

61. Ofori S, Cobbinah S, Imoro A. PAH Quantification and Estimated Carcinogenic Risks at Selected Fuel Stations in Tamale Metropolis, Ghana. Ghana Sci J. 2020;61(1):60-72. https://doi.org/10.4314/gjs.v61i1.5

62. Iwegbue CM, Obi G. Distribution, sources, and health risk assessment of polycyclic aromatic hydrocarbons in dust from urban environment in the Niger Delta, Nigeria. Hum Ecol Risk Assess. 2016;22(3):623-638. https://doi.org/10.1080/10807039.2015.1100157

63. Emoyan OO. Quantification and cancer risk evaluation of polycyclic aromatic hydrocarbons in soil around selected telecom masts in Delta state Nigeria. Egypt J. Chem. 2020;63(2):433-448. https://doi.org/10.21608/ejchem.2019.17620.2081

64. Tarafdar A, Sinha A. Health risk assessment and source study of PAHs from roadside soil dust of a heavy mining area in India. Arch Environ Contam Toxicol. 2019;74(5):252-262. https://doi.org/10.1007/9338244.2018.1444575

65. Rastegari Mehr M, Keshavarzi B, Moore F et al. Contamination level and human health hazard assessment of heavy metals and polycyclic aromatic hydrocarbons (PAHs) in street dust deposited in Mahshahr, southwest of Iran. Hum Ecol Risk Assess. 2016;22(8):1726-1748. https://doi.org/10.1080/10807039.2016.1219221

66. Ke C-L, Gu Y-G, Liu Q. Polycyclic aromatic hydrocarbons (PAHs) in exposed-lawn soils from 28 urban parks in the megacity Guangzhou: occurrence, sources, and human health implications. Arch Environ Contam Toxicol. 2017;72(4):496-504. https://doi.org/10.1007/s00244-017-0397-6

67. Shi R, Li X, Yang Y et al. Contamination level and human health risks of polycyclic aromatic hydrocarbons in surface soils from Tianjin coastal new region, China. Environ. Pollut. 2020;268:115938. https://doi.org/10.1016/j.envpol.2020.115938

68. Benarba B, Meddah B, Hamdani H. Cancer incidence in North West Algeria (Mascara) 2000-2010: results from a population-based cancer registry. Excli J. 2014;13:709-723.