

Polycyclic aromatic hydrocarbons (PAHs) in crude oil-contaminated water and soil and their removal using locally available plant materials

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ABSTRACT

The presence of polycyclic aromatic hydrocarbons (PAHs) in water and soil can be harmful to human life when ingested. PAHs are determined in the water and soil of the B-Dere community of Rivers State, Nigeria. The concentration level, source identification, and treatment were carried out. The water samples were treated with garlic and Moringa seed extracts, while the soil samples were treated with garlic and Fenton oxidation reagents. PAHs were extracted before and after treatment. The gas chromatograph mass spectrometer analyses showed 13 PAHs in the water and 10 PAHs in the soil. The highest concentration in water was recorded for benzo(ghi)perylene, with a mean value of $27.7 \pm 0.25 \text{ ngL}^{-1}$, while that of soil was recorded for benz(a)anthracene, with a mean value of $14.4 \pm 0.631 \text{ ngkg}^{-1}$. The source of PAHs in water was pyrogenic, while that of soil was petrogenic. Garlic extract removed 100% of benz(a)anthracene and benzo(b)fluorothane from the water, while Moringa removed 100% biphenylene from the water. However, garlic extract removed 2.59% of acenaphthylene, while Fenton reagents removed 100% of anthracene, phenanthrene, and chrysene from the soil. Moringa seed and garlic extracts can be used in PAH's polluted water treatment.

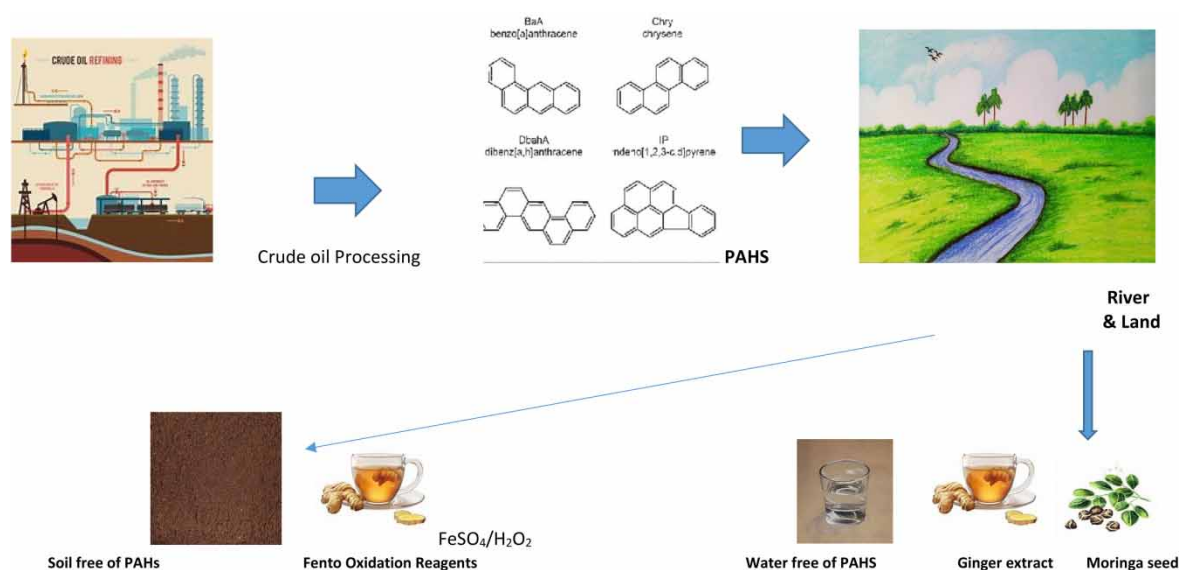
Key words: B-Dere, garlic, Moringa, PAHs, soil, water

HIGHLIGHTS

- PAHs were determined in the water and soil of the B-Dere Community (an oil-producing community).
- Moringa seed extracts removed 100% biphenylene from the water.
- Garlic extracts removed 100% of benz(a)anthracene and benzo(b)fluorothane from the water.
- Garlic extract removed a fraction of Acenaphthylene from the soil.

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GRAPHICAL ABSTRACT



1. INTRODUCTION

Crude oil processing releases substances that can affect humans, plants and aquatic organisms if ingested through drinking water, consumption of plants through the food chain or inhalation. Most of the crude oil constituents and byproducts are categorized as persistent organic pollutants (Jacquin *et al.* 2017). The pollution of water bodies in Nigeria was commonly caused by oil spills. This affects vital ecosystem balance such as nutrient recycling and water purification processes (Akpan & Bassey 2020; Erwin *et al.* 2023). In Nigeria, a substantial number of activities that affect the ecosystem occur regularly, especially in the Niger Delta region. This includes waste discharged from industries, flaring of gases and water and soil pollution which cause health challenges (Ite *et al.* 2018). The natural environment contaminated by the spillage of oil is very difficult to remove from marshes and mangroves (Wali *et al.* 2019). The oil spill causing the contamination of environmental media (soil, water, and air) has been linked to respiratory challenges, such as cancer risk, skin diseases, and gastrointestinal disorders for the residents (Kuppusamy *et al.* 2020; Onyena & Sam 2020). Toxic effects, such as asphyxiation of plants and death of organisms, may occur, if it contains a high amount of light aromatic hydrocarbons (Linden & Jonas 2013).

The pollution of surface water bodies has led to a decrease in the quantity of fish, aquatic animals, and species. Aquatic animals not trapped by the pollutants migrate away from more polluted areas to less polluted areas of the water body. When crude oil contaminates water bodies, micro-organisms migrate to the uncontaminated areas (Omozue 2021). This causes economic hardship for fishermen (Iwubeh *et al.* 2020). Plants cannot move in most cases and get destroyed, thereby putting the ecosystem in a state of disequilibrium. The wastewater, heavy metals, and toxic chemicals discharged due to crude oil processing and agricultural practices contributed to the quality issues of the water. These further complicate the sustainable treatment and remediation of water systems (Eyankware *et al.* 2021).

Polycyclic aromatic hydrocarbons (PAHs) are toxic, mutagenic, teratogenic, and carcinogenic (USEPA 2009). PAHs in the soil can be encapsulated in minerals, and are present in non-aqueous liquid, which can serve as a route for human exposure (Tarafdar *et al.* 2018). Other sources are industrial activities, vehicular emissions, horticultural composts, and the combustion of fossil fuels (Kumar *et al.* 2020; Ambade *et al.* 2021). The presence of PAHs everywhere stimulated research interest in different remediation techniques (Gan *et al.* 2009).

The low-molecular-weight (Lmw) PAHs in contaminated soils have a half-life of 5–7 years while the high-molecular-weight (Hmw) PAHs have a half-life of 9–10 years (Wild *et al.* 1991). There is a need to remove PAHs from contaminated soils because of adverse human health effects associated with its ingestion (Haritash & Kaushik 2009). Degradation of PAHs by chemical oxidation has been used as an effective method of remediation of the soil. Permanganate ion is a strong oxidant for organic contaminants that can break the double bonds present

in PAH structure (Ferrarese *et al.* 2008; Brown *et al.* 2003). The oxidants (Fenton reagents) can rapidly remove PAHs in soils (Lindsey *et al.* 2003; Yap *et al.* 2011).

PAHs are chemically stable and not biodegradable. The tendencies to cause cancer of the lung, bladder, and skin make the need to determine PAH levels in water, soil and air very important. The route and frequency of exposure go a long way in determining the severity of adverse effects (Yu *et al.* 2018). PAHs in water should not exceed $0.01 \mu\text{g L}^{-1}$ and they should be considered priority pollutants in drinking water (Wang *et al.* 2007; Ma *et al.* 2020). Sources of PAHs in water could be petroleum spills, fuel combustion, run-off from roads, creosote-coated wood leachate, and effluents from industries (Fouad *et al.* 2022; Mojiri *et al.* 2019).

Ruptures of old pipes have been recorded severally in the B-Dere community and the odor of the oil can be perceived some distance away from the spill location. Gokana and B-Dere people have protested oil spills for affecting their source of livelihood (Wizor & Eludonyi 2020).

Many varieties of sorbent materials have been used for the removal of oil by absorption or adsorption. Synthetic, inorganic, and organic materials are effective with one or more drawbacks. Synthetic materials, such as polyurethane and butyl rubber, are non-biodegradable causing environmental challenges. Fouling occurs in clay and zeolites when used as adsorbent materials for oil because of water absorption in their matrices (Al-Jammal & Juzsakova 2017; Kumar *et al.* 2021). The used agro-based materials, such as rice husk, corn cobs, and silica, have been effective. It is necessary to further explore extracts from plant materials because of their low cost and environmental degradability.

This study aimed to determine environment-friendly ways of removing PAHs in contaminated water and soil of a rural community. Garlic and Moringa seed extracts were used to remove PAHs from water, while garlic and Fenton reagents were used to remove them from the soil of the study area.

2. MATERIALS AND METHODS

2.1. The study area

The study area is the B-Dere community in the Gokana Local Government Area of Rivers State, Nigeria. The town covers an area of 963 square kilometers. It is one of the many communities that are affected by the oil spillage problems in Ogoni land. B-Dere, Gokana, Nigeria is only 20 meters 65.62 feet above sea level. It is a coastal community which is affected by heavy flooding. Most inhabitants are fishermen and because of the availability of dryland within the vicinity, when the sea level rises, most coastal communities around the area relocated to B-Dere. It is located between Longitude $3^{\circ}48' 84''$ E – Latitude $5^{\circ}17' 87.5''$ N and Longitude $3^{\circ}77' 86''$ E – Latitude $5^{\circ}25'92''$ N (Figure 1). Oil spillage in the area impacts negatively on the adjoining creek which rendered large agricultural land unsuitable for the intended use (Weli & Arokoyu 2014).

2.2. Chemicals and reagents

All reagents used for preparations are of analytical grade which require no further purification. The water used was doubly deionized water using a Millipore purification system (Millipore, France). The Fenton reagents used are Hydrogen peroxide (50%) and iron (II) sulfate (0.1%) (Akpoveta *et al.* 2018).

2.3. Treatment of PAH-contaminated water with garlic and moringa extracts

Garlic and moringa seeds were obtained from the market and identified by the plant expert from the School of Agriculture of the University. The garlic was washed and rinsed with distilled water before blending to extract its water content. The water was filtered using filter paper and kept in the refrigerator. The moringa seed shell was removed and 10 g of the seeds were soaked in 250 mL of distilled water for 6 h. 300 mL of contaminated water sample was added to 100 mL of the plant extracts and the mixture was shaken on a rotary shaker for 15 min at 105 cycles per minute (Hussein *et al.* 2011; Idris *et al.* 2014).

2.4. Extraction of PAHs from water

A standard method was used for the liquid–liquid PAH extraction with a mixture of dichloromethane and n-hexane. The extraction was carried out using Moringa seed extract (250 mL) and garlic extract (250 mL), respectively. It was shaken rigorously with the addition of a 100 mL mixture of dichloromethane and n-hexane.

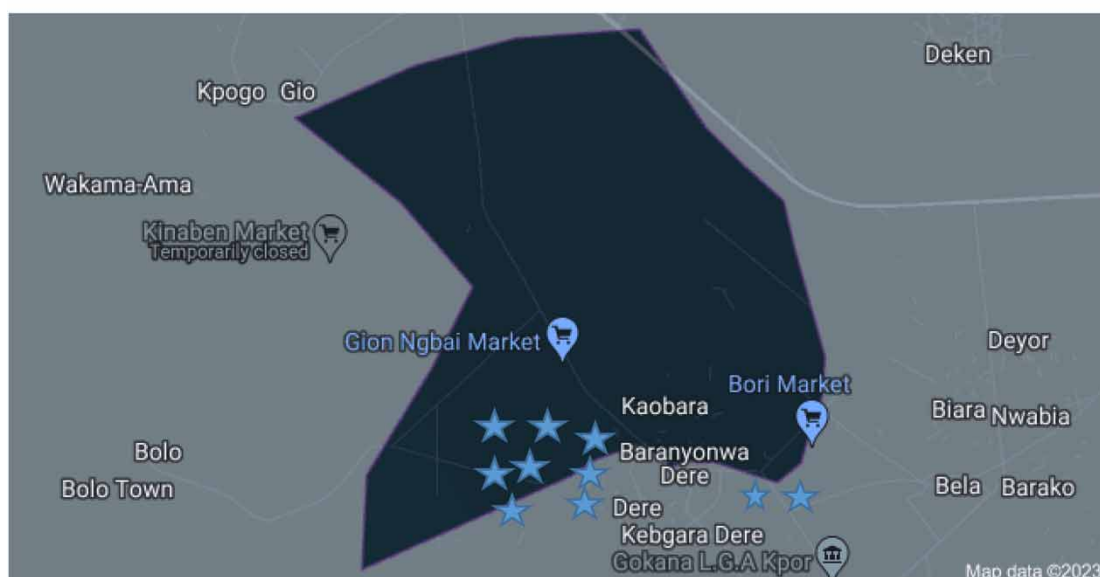
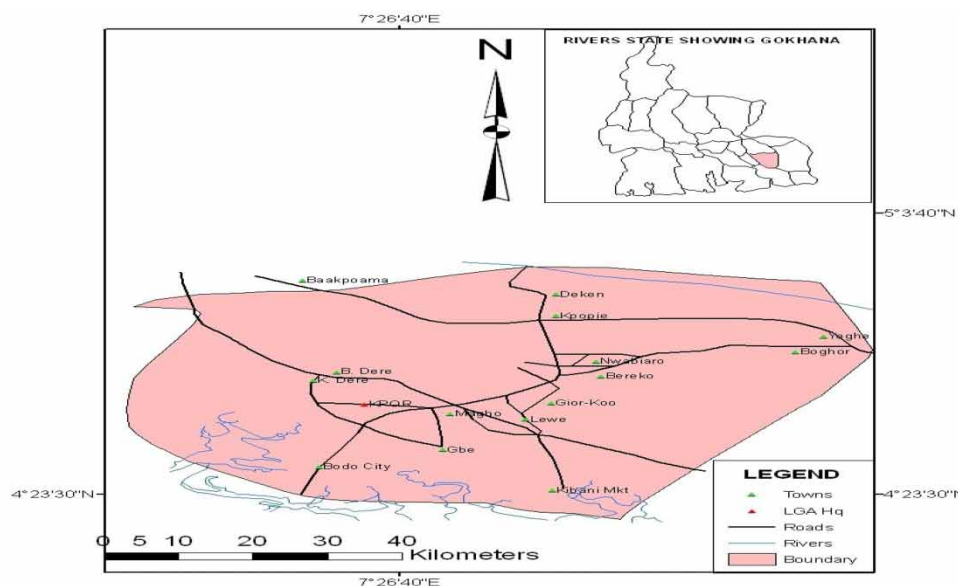


Figure 1 | Map showing B Dere in Gokana Local Government Area, Rivers State, Nigeria and its google map.

2.5. Treatment of PAH-contaminated soil with garlic extracts and Fenton reagents

20 mL of garlic and 20 mL of Fenton reagents were added, respectively to 2 g of soil in each beaker. They were mixed and allowed to settle. The reagents were optimized following [Akpoveta *et al.* \(2018\)](#). The sample was filtered into a clean solvent-rinsed extraction bottle, using filter paper fitted into Buchner funnels. The extract was concentrated to 2 mL by freeze drying and then transferred for cleanup/separation.

2.6. Extraction of PAHs from the soil

10 mL of dichloromethane was added to 2 g of the soil, mixed, shaken on a rotary shaker and filtered using Buchner funnels ([Lau *et al.* 2010](#)). The extract was concentrated to 2 mL and then transferred for cleanup.

2.7. Cleanup

An optimized solid phase extraction (SPE) was used to co-extract PAHs from the matrix of Moringa extract and garlic extract, respectively. The column was first conditioned with dichloromethane and n-hexane, while the plant extracts were loaded on a normal phase column ($-NH_2$ and Alumina) ([Blasco *et al.* 2007](#)). 2 mL of hexane was used to elute the PAHs from the column.

2.8. Instrumental settings

PAH analyses were carried out using a gas chromatograph (GC/MS, Agilent, 7890 series, USA). The flow rate was set at 1.5 mL/min with nitrogen as the carrier gas. The oven temperature was set at 80 °C, holding time of 0.5 min; ramped 80–220 °C at 20 °C/min, then 220–300 °C at 10 °C/min.

2.9. Quality control

The double-distilled water samples were spiked with 16 PAHs and extracted. The results were validated through recovery studies.

2.10. Statistical analysis

Principal components analysis (PCA) was used for data evaluation of PAH sources in water and soil samples. Varimax rotation and eigenvalue of 1 were used to reduce a set of original data.

2.11. Calculation

(i) The %PAHs removed was calculated by

$$\% \text{PAHs removed} = (C_i - C_f) / C_i \times 100 \quad (1)$$

where C_i is the mean concentration of PAHs in the raw water and C_f is the mean concentration after treatment.

(ii) The tendency of BaP to cause cancer was calculated using (BaP_{TEQ}):

$$\text{BaP}_{\text{TEQ}} = \sum C_i \times \text{BaP}_{\text{TEF}} \quad (2)$$

where BaP_{TEF} is the cancer potency relative to BaP and C_i is the individual PAH concentration

(iii) The BaP mutagenic equivalency factor (BaP_{MEQ}) for the PAHs was evaluated using the following Equation:

$$\text{BaP}_{\text{MEQ}} = \sum C_i \times \text{BaP}_{\text{MEF}} \quad (3)$$

where BaP_{MEF} is the mutagenic potency factor while C_i is the PAH concentration.

(iv) Incremental Lifetime Cancer Risk (ILCR)

By ingestion, dermal contact and inhalation (USEPA 2009) were calculated with

$$\text{ILCR}_{\text{ing}} = \frac{(C \times \text{IngR} \times \text{EF} \times \text{ED} \times \text{CF} \times \text{SFO})}{\text{BW} \times \text{AT}} \quad (4)$$

where ILCR_{ing} is the risk via ingestion; C is the concentration of PAHs (mg/kg soil; ng/L water); IngR is the ingestion rate of the soil (mg/kg), (200 for children and 100 for adults); EF is the exposure frequency (313 days/year for both children and adults); ED is the exposure duration (6 years for children and 24 years for adults); BW is the average body weight (15 kg for children; 60 kg for adults); AT is the average time ($52 \times 365 = 18,980$ days for both children and adults); CF is the conversion factor ($1 \times 10^{-6} \text{ kgmg}^{-1}$); SFO is the oral slope factor. The SFO ($\text{mg}^{-1}\text{kg}^{-1}\text{day}^{-1}$) values are BaA = (7.3×10^{-1}), Chry = (7.3×10^{-6}), BbF = (7.3×10^{-1}), BkF = (7.3×10^{-2}), and BaP = 7.3 (USEPA 2009).

$$\text{ILCR}_{\text{derm}} = \frac{(C \times \text{SA} \times \text{AF} \times \text{ABF} \times \text{EF} \times \text{ED} \times \text{CF} \times \text{SFO} \times \text{GLABS})}{\text{BW} \times \text{AT}} \quad (5)$$

where $\text{ILCR}_{\text{derm}}$ is the risk via dermal contact; SA is the surface area of the skin that is in contact with the soil ($2,800 \text{ cm}^2/\text{day}$ for children; $5,700 \text{ cm}^2/\text{day}$ for adults), (Ferreira-Baptista & De Miguel 2005); AF is the skin adherence factor for dust (0.2 mg/cm^2 for children and 0.07 mg/cm^2 for adults); ABF is the dermal absorption factor (chemical specific) (0.13 for both children and adults); EF is the exposure frequency (313 days/year for both children and adults); ED is the exposure duration (6 years for children and 24 years for adults); CF is the conversion factor (1×10^{-6} for both children and adults); GIABS is the gastro-intestinal absorption factor

(8 hrs/day for both children and adults).

$$ILCR_{inh} = \frac{(C \times EF \times ET \times ED \times IUR)}{BW \times AT} \quad (6)$$

where $ILCR_{inh}$ is the risk via inhalation; ET (Exposure Time) = (52 yrs $\times$ 365 days/yr $\times$ 24 h/day = 455,520 h/day for both children and adults); IUR values for BaA, BaF, and BkF are 1.1×10^{-4} (mg/m³)⁻¹ for Chry is 1.1×10^{-5} for BaP is 1.1×10^{-3} (mg/m³)⁻¹ for BaP (USEPA 2012); BW (Body Weight) = (15 kg for children; 60 kg for adults); AT is the average time (52 yrs $\times$ 365 days/yr $\times$ 24 h/day = 455,520 h/day for both children and adults). *Note:* The average life expectancy of Nigerians is 52 years (Iwegbue & Obi 2016); PEF (Particle Emission Factor) for respirable particle (PM₁₀) = 1.36×10^9 m³/kg. (Iwegbue & Obi 2016).

3. RESULTS AND DISCUSSION

The percent PAHs recovered was explained under recovery studies while physicochemical parameters, such as pH, temperature, and conductivity of both water and soil, were discussed under various headings (Tables 1 and 3). The range of concentration of PAHs in water and soil is presented in Tables 2 and 4. The source identification of PAHs and lifetime risk are discussed in Table 5 and 6, respectively. The results of treatment with moringa, garlic, and fento-oxidation reagents are presented in Table 7.

Table 1 | Physiochemical parameters of water

Parameters	Minimum	Maximum	Mean $\pm$ SD	WHO 2011
pH	6.50	7.1	6.7 $\pm$ 0.11	6.5–8.5
Temperature °C	28.5	30.5	29.6 $\pm$ 0.35	NA
Electrical conductivity (μ Scm ⁻¹)	102.5	206.1	159 $\pm$ 21.3	250
	200	240.0	220 $\pm$ 7.07	250
Alkalinity (mgL ⁻¹)	121	140.1	129 $\pm$ 3.59	NA
Total hardness (mgL ⁻¹)	256	450.3	322 $\pm$ 36.3	250
Cl ⁻ (mgL ⁻¹)	255.8	350.7	301.9 $\pm$ 35.9	250
TDS (mgL ⁻¹)	50.3	105.4	86.9 $\pm$ 10.1	250

Table 2 | Range of PAHs in water samples of the B-Dere community

PAHs	Abbreviation	Min	Max	Mean $\pm$ SD	Henley Dam water, South Africa (Munyengabe <i>et al.</i> 2017)	Water of Lodz-Choim Area, Poland (Kabzinski <i>et al.</i> 2002)	Shamal Helman water, Cairo Egypt (Fouad <i>et al.</i> 2022)
Naphthalene	Nap	7.01	7.03	7.02 $\pm$ 0.006	13.52 $\pm$ 0.01	326 $\pm$ 632	208 $\pm$ 118
Acenaphthylene	Acy	10.80	10.84	10.8 $\pm$ 0.012	34.31 $\pm$ 0.03	478 $\pm$ 1,210	ND
Biphenylene	Bip	10.88	10.91	10.9 $\pm$ 0.009	ND	ND	ND
Acenaphthene	Ace	11.37	11.41	11.4 $\pm$ 0.012	ND	38 $\pm$ 41	ND
Fluorene	Flu	12.61	12.64	12.6 $\pm$ 0.009	39.34 $\pm$ 0.02	175 $\pm$ 181	103 $\pm$ 117
Anthracene	Ant	14.97	15.02	14.9 $\pm$ 0.015	ND	69 $\pm$ 205	475 $\pm$ 124
Phenanthrene	Phe	15.01	15.12	15.1 $\pm$ 0.035	ND	12 $\pm$ 27	315 $\pm$ 132
Pyrene	Pyr	18.04	18.08	18.1 $\pm$ 0.012	ND	22 $\pm$ 37	225 $\pm$ 143
Tryphenylene	Trp	22.04	22.07	22.1 $\pm$ 0.009	ND	ND	ND
Benz(a)anthracene	BaA	22.16	24.61	22.9 $\pm$ 0.813	ND	25 $\pm$ 62	ND
Benzo(e)pyrene	BeP	24.62	25.28	24.8 $\pm$ 0.218	ND	8 $\pm$ 05	ND
Benzo(b)fluorothane	BbF	25.28	27.42	26.0 $\pm$ 0.71	ND	24 $\pm$ 30	ND
Benzo(ghi)perylene	BghiP	27.21	28.06	27.7 $\pm$ 0.25	ND	2 $\pm$ 4	ND
SUM				224.463 $\pm$ 0.023		1,178 $\pm$ 2,247	

ND, not detected.

Table 3 | Physiochemical parameters of the soil

	Minimum	Maximum	Mean $\pm$ SD
pH	3.7	6.6	5.37 $\pm$ 1.49
Moisture (%)	6.7	7.09	6.90 $\pm$ 0.195
Density (g/cm ³)	0.79	0.95	0.873 $\pm$ 0.79
Organic carbon (mg/kg)	0.92	2.11	1.527 $\pm$ 0.595
Electrical conductivity, μScm^{-1}	150	263	196.3 $\pm$ 59.18
Sand (%)	70.4	86.9	76.8 $\pm$ 8.85
Silt (%)	0.66	0.78	0.71 $\pm$ 0.062
Clay (%)	23.4	66	44.9 $\pm$ 21.3

Table 4 | Range of PAHs in soil samples of B-Dere

PAHs	Min	Max	Mean $\pm$ SD	The soil of Shatt Al-Arab Iran (Al-Sad <i>et al.</i> 2019)	The soil of South Russia (Dudrikova <i>et al.</i> 2023)
NaP	6.001	6.324	6.14 $\pm$ 0.163	10.20 $\pm$ 0.20	555 $\pm$ 19.4
Acy	7.92	8.865	8.24 $\pm$ 0.439	0.47 $\pm$ 0.10	9.6 $\pm$ 9.4
Ace	9.009	9.563	9.33 $\pm$ 0.234	0.46 $\pm$ 0.40	9.6 $\pm$ 6.9
Flu	8.865	9.857	9.48 $\pm$ 0.430	0.75 $\pm$ 0.10	18.4 $\pm$ 21.3
Phe	10.76	11.24	10.9 $\pm$ 0.199	7.92 $\pm$ 0.20	94.9 $\pm$ 92.1
Ant	10.5	11.43	10.98 $\pm$ 0.38	1.55 $\pm$ 0.30	1.4 $\pm$ 3.6
Flr	11.131	12.872	11.9 $\pm$ 0.771	9.54 $\pm$ 0.20	155 $\pm$ 302.6
Pyr	12.952	13.205	13.02 $\pm$ 0.125	8.04 $\pm$ 0.10	85.5 $\pm$ 194.4
Chr	13.546	14.945	14.01 $\pm$ 0.063	5.90 $\pm$ 0.50	77.4 $\pm$ 155.2
BaA	14.091	14.945	14.4 $\pm$ 0.631	3.27 $\pm$ 0.40	80.7 $\pm$ 212.8
SUM			108.5 $\pm$ 0.289		

Table 5 | Source identification of PAHs in water and soil of B-Dere

PAHs	Limit	sources	soil	water
Ant/Ant + Phe	<0.1	petrogenic		
	>0.1	combustion	0.5	0.498
	<0.4	petroleum		
Flr/Flr + Pyr	0.4–0.5	fuel	0.494	-
	>0.5	coal		
	<0.2	petrogenic		
BaA/BaA + Chry	0.2–0.35	fuel		
	>0.35	coal	0.5	1
	<0.2	Petrogenic		
LMW/HMW	>0.1	petrogenic	1.012	
	<1.0	pyrogenic		0.461

3.1. Recovery studies

The results from recovery studies showed that percentage recovery of naphthalene, acenaphthylene, biphenylene, acenaphthene, fluorene, anthracene, phenanthrene, pyrene, triphenylene, benz(a)anthracene, benzo (e)pyrene,

Table 6 | ILCR and human health risk in soil and water samples of the D-Bere

Risk parameters	PAHs used for computation	Soil	Water
ILCR _{ing} (Adult)	Sum of PAHs	1.28×10^{-7}	2.57×10^{-7}
ILCR _{ing} (Children)	Sum of PAHs	1.031×10^{-6}	2.05×10^{-6}
ILCR _{derm} (Adult)	Sum of PAHs	1.605×10^{-7}	1.046×10^{-9}
ILCR _{derm} (Children)	Sum of PAHs	2.252×10^{-6}	4.40×10^{-6}
ILCR _{inh} (Adult)	Sum of PAHs	1.092×10^{-5}	2.273×10^{-8}
ILCR _{inh} (Children)	Sum of PAHs	2.729×10^{-6}	5.39×10^{-9}
BaP _{Req} BaP _{Meq}	BaA	1.495	2.22
	Chr	0.0149	Nil
	BaP	Nil	0.253
	BbF	Nil	24.7
	BaA	1.225	1.829
	Chr	0.254	Nil
	BaP	Nil	6.325
	BbF	Nil	24.7

Table 7 | Percentage of PAHs removed from water and soil using garlic extract, Moringa extracts, and Fento reagents

Water			Soil		
	% removed by garlic extract	% removed by Moringa extract		% removed by garlic extract	% removed by Fenton reagent
NaP	0.64	Nil	NaP	1.34	1.45
Acy	Nil	0.139	Acy	2.59	0.417
Ace	0.184	0.044	Ace	0.429	0.265
Flu	0.734	0.126	Flu	0.78	Nil
Ant	0.207	0.033	Ant	0.177	100
Phe	Nil	0.033	Phe	Nil	100
Pyr	0.171	0.116	Pyr	Nil	0.06
BaA	100	0.305	BaA	0.368	0.468
Bip	0.28	0.0001	Chr	Nil	100
Tph	100	0.045	Flr	0.186	Nil
BeP	0.316	0.19			
BbF	0.14	0.059			
Bghip	2.5	100			
BbT	100	0.338			

benzo(b)fluorothane, benzo(ghi)perylene, and BbT are $90.3 \pm 1.79\%$, $90.1 \pm 2.16\%$, $91.7 \pm 2.61\%$, $90.3 \pm 1.55\%$, $91.8 \pm 2.12\%$, $90.9 \pm 2.83\%$, $94.2 \pm 1.8\%$, $92.1 \pm 1.71\%$, $89.9 \pm 1.02\%$, $91.6 \pm 2.03\%$, $93.31 \pm 2.03\%$, $95.1 \pm 1.94\%$, $94.3 \pm 1.04\%$, and $92.7 \pm .04\%$, respectively, while the limit of detection ranged between 0.02 and 0.1. Kabzinski *et al.* (2002) reported a higher percentage recovery value for phenanthrene and similar values for other PAHs. SPE and reverse-phase high-performance chromatography (RHPLC) were used to achieve the results.

3.2. Physicochemical parameters of water

The pH recorded from B-Dere, Gokana Creek water ranged from 6.5 to 7.1 with an average value of 6.7 ± 0.11 while the temperature ranged from 28.5 to 30.5 °C with an average value of 29.6 ± 0.35 °C (Table 1). The pH range is in agreement with Nkpa *et al.* (2013), who studied crabs and shrimps in Bodo City, B-Dere, and Kaa river water and reported a similar range of values. Electrical conductivity recorded in the water ranged from

102.5 to 206.1 μSCm^{-1} with an average value of $159 \pm 21.3 \mu\text{SCm}^{-1}$. The low value of conductivity implies lower mineralization of organic matter in the water (Abida & Harikrishna 2008). Alkalinity ranged from 121 to 140 $\text{mgCaCO}_3\text{L}^{-1}$ with an average value of $129 \pm 3.59 \text{mgCaCO}_3\text{L}^{-1}$. Total hardness ranged from 120.9 to 140.1 mgL^{-1} with a mean value of $131.1 \pm 7.83 \text{mgL}^{-1}$.

Chloride concentration ranged from 255.8 to 350.7 mgL^{-1} with a mean value of $301.9 \pm 35.9 \text{mgL}^{-1}$. All chloride concentrations are above the WHO limit in drinking water. This is in agreement with Oladimeji *et al.* (2009). The TDS concentration ranged between 50.3 and 105.4 mgL^{-1} with a mean value of $86.9 \pm 10.1 \text{mgL}^{-1}$ which was below the WHO limit. Extremely high TDS concentrations can cause depletion of oxygen, affect the growth of many aquatic lives and can even cause their death.

3.3. PAHs in water

The naphthalene (Nap) concentration in water ranged from 7.01 to 7.03 ngL^{-1} with a mean value of $7.02 \pm 0.006 \text{ngL}^{-1}$. Acenaphthylene (Acy) ranged from 10.80 to 10.84 ngL^{-1} with an average value of $10.8 \pm 0.012 \text{ngL}^{-1}$ (Table 2). Grmasha *et al.* (2024) reported a higher range of naphthalene and acenaphthylene in the Danube river, Budapest.

Biphenylene (Bip) ranged from 10.88 to 10.91 ngL^{-1} with an average value of $10.9 \pm 0.009 \text{ngL}^{-1}$. Acenaphthene (Ace) ranged from 11.37 to 11.41 ngL^{-1} with an average value of $11.4 \pm 0.012 \text{ngL}^{-1}$. Fluorene (Flu) ranged from 12.61 to 12.64 ngL^{-1} with an average value of $12.6 \pm 0.009 \text{ngL}^{-1}$. Anthracene (Ant) ranged from 14.97 to 15.02 ngL^{-1} with an average value of $14.9 \pm 0.015 \text{ngL}^{-1}$. Phenanthrene (Phe) ranged from 15.01 to 15.12 ngL^{-1} with an average value of $15.1 \pm 0.035 \text{ngL}^{-1}$. Munyengabe *et al.* (2017) reported Phenanthrene with a higher concentration value in the raw water of Msundusi River in South Africa than the results obtained from the present study. Naphthalene and Phenanthrene found in the water may be due to heavy traffic and aircraft passing through the study area. The emission produced may precipitate into the water body (Mojiri *et al.* 2019). Pyrene (Pyr) ranged from 18.04 to 18.08 ng/L with an average value of $18.1 \pm 0.012 \text{ngL}^{-1}$. Tryphenylene (Trp) ranged from 22.04 to 22.07 ng/L with an average value of $22.1 \pm 0.009 \text{ngL}^{-1}$. Benz(a)anthracene (BaA) ranged from 22.16 to 24.61 ngL^{-1} with an average value of $22.9 \pm 0.813 \text{ngL}^{-1}$. Benzo(e)pyrene (BeP) ranged from 24.62 to 25.28 ngL^{-1} with an average value of $24.8 \pm 0.218 \text{ngL}^{-1}$. Benzo(b)fluorothane (BbF) ranged from 25.28 to 27.42 ngL^{-1} with a mean value of $26.0 \pm 0.71 \text{ngL}^{-1}$. Benzo(ghi)perylene (BghiP) ranged from 27.21 to 28.06 ngL^{-1} with an average value of $27.7 \pm 0.25 \text{ngL}^{-1}$. Kabzinski *et al.* (2002) reported higher values for all the PAHs detected in the drinking water of Lodz, Poland except for Pyrene (Pyr), Phenanthrene (Phe), Benzo(e)pyrene (BeP), and Benzo(ghi)perylene (BghiP) (Table 2). Fouad *et al.* (2022) reported a higher value of PAHs in Shamal Helman water in Cairo, Egypt. The order of dominance is Benzo(ghi)perylene > benzo(b)fluorothane > Benzo(e)pyrene > Benz(a)anthracene > Tryphenylene > Pyrene > Phenanthrene > Anthracene > Fluorene > Acenaphthene > Biphenylene > Acenaphthylene > Naphthalene. The light PAHs are found in all the water samples due to their hydrophilic nature which makes them persistent; however, factors, such as aeration, absorption, adsorption, and precipitation, are also responsible (Rojo-Nieto *et al.* 2013). The sum of PAHs before treatment was 224.463ngL^{-1} which can be classified as lightly polluted (Table 2). A higher value of a sum of PAHs was reported in the rivers of Dhaka, Bangladesh (Nahar *et al.* 2023). Similarly, Fouad *et al.* (2022) reported higher values in the river of Cairo, Egypt. Surface water contaminations involving PAHs can be categorized into four, namely, micro-polluted, if PAHs concentration ranges between 10 and 50 ngL^{-1} ; lightly polluted, if it ranges between 50 and 250 ngL^{-1} ; moderately polluted, if the range is between 250 and 1,000 ngL^{-1} , and heavily polluted if the concentration is greater than 1,000 ngL^{-1} (Feng-Chen *et al.* 2020).

3.4. Physiochemical parameters of soil

The pH recorded from B-Dere soil ranged from 3.7 to 6.6 with a mean value of 5.37 ± 1.49 while the percentage moisture ranged from 6.7 to 7.09% with a mean value of $6.90 \pm 0.195\%$ (Table 3). The pH range and its average value imply that the soil is acidic which may need to be limed to reduce the acidity for agricultural purposes. The soil density ranged 0.79–0.95 g/cm^3 with a mean value of $0.873 \pm 0.79 \text{g/cm}^3$.

The organic carbon ranged from 0.92 to 2.11 mg/kg with an average value of $1.527 \pm 0.595 \text{mg/kg}$, while the electrical conductivity ranged between 150.0 and 263.0 μSCm^{-1} with an average value of $196.3 \pm 59.18 \mu\text{SCm}^{-1}$. Ghare & Kumbhar (2021) reported a higher pH and a lower %organic carbon in the soil of the Gujarat region of India. The organic carbon can be low, while the high electrical conductivity confers salinity to the soil as a result of the different ions present (Zeeshan *et al.* 2014). The percentage of sand ranged from 70.4 to 86.9% with an

average value of $76.8 \pm 8.85\%$, while the percentage of silt ranged from 0.66 to 0.78% with a mean value of $0.71 \pm 0.062\%$. The percentage of clay ranged from 23.4 to 66% with a mean value of $44.9 \pm 21.3\%$.

3.4.1. PAHs in the soil of B-Dere

The naphthalene concentration in the soil ranged from 6.001 to 6.324 ng/g with an average value of 6.14 ± 0.163 ng/g. A higher naphthalene concentration was reported in the soil of Shatt Al- Arab, Iran (Al-Sad *et al.* 2019). Acenaphthylene ranged from 7.92 to 8.865 ng/g with an average value of 8.24 ± 0.439 ng/g (Table 4). Acenaphthene ranged from 9.009 to 9.563 ng/g with a mean value of 9.33 ± 0.234 ng/g.

Fluorene ranged from 8.865 to 9.857 ng/g with an average value of 9.48 ± 0.430 ng/g. Phenanthrene ranged from 10.76 to 11.24 ng/g with an average value of 10.9 ± 0.199 ng/g. Anthracene ranged from 10.5 to 11.43 ng/g with an average value of 10.98 ± 0.380 ng/g. Floranthene ranged from 11.131 to 12.872 ng/g with an average value of 11.9 ± 0.771 ng/g. Pyrene ranged from 12.952 to 13.205 ng/g with an average value of 13.02 ± 0.125 ng/g. Chrysene ranged from 13.546 to 14.945 ng/g with an average value of 14.01 ± 0.063 ng/g. Benz(a)anthracene ranged from 14.091 to 14.945 ng/g with a mean value of 14.4 ± 0.631 ng/g. Natural and anthropogenically transformed coastal soils of Southern Russia showed an extremely higher value of naphthalene, floranthene, pyrene, chrysene, and benz(a)anthracene in comparison with the present study (Dudrikova *et al.* 2023). Some PAHs were not found in the soil sample due to possible evaporation and biological activity (Sanches *et al.* 2011; Ukiwe *et al.* 2013). The order of PAHs dominance in the soil of the study area is as follows: Benz(a)anthracene > Chrysene > Pyrene > Floranthene > Anthracene > Phenanthrene > Fluorene > Acenaphthene > Acenaphthylene > Naphthalene.

3.5. Source identification

The sources of PAHs in soil and water of B-Dere are mainly due to combustion as the values obtained from Ant/Ant + Phen are 0.5 and 0.498, respectively (Table 5).

The BaA/(BaA + Chry) ratio in soil samples from B-Dere was 0.5 which indicated a pyrogenic origin. The ratio >0.35 showed pyrogenic sources, while <0.20 indicated petrogenic sources (Yunker *et al.* 2002). The results indicated sewage deposit or compost origin. Nwaichi *et al.* (2020) reported lower value in the soil collected from Abuloma and Okujagu-ama, Port Harcourt, Nigeria.

The Flr/Flr + Pyr ratios in soil samples from B-Dere were 0.494, which indicated a source to be from fuel combustion, while there is no fluoranthene in the water to compute the value. The presence of PAHs in surface water is known to be caused by oil spillage, exhaust, smoke, fire, industries, and precipitation (Wang *et al.* 2000). The ratio (Lmw/Hmw) PAHs was 1.012 in the soil, which indicated petrogenic sources, while in water it was 0.461 which is pyrogenic. This is in agreement with the previous studies (Rocher *et al.* 2004).

3.6. Principal component analysis

The principal component analysis of water data showed two major components (Figure 2).

Component 1 consists of Flu, BbF, BeA, BeP, NaP, and Bghip, which represents 59.49% of the total variance observed. Component 2 is mainly dominated by Ant, Phe, and Pyr, which represents 40.508% of the total variance observed. This further supports the possibility of petrogenic sources.

The principal component analysis of soil data showed two major components (Figure 3).

Component 1 consists of NaP, Chr, Flr, Pyr, Ant, Phe, Acy, Ace, and Flu which represents 80.631% of the total variance observed. Component 2 is mainly dominated by BaA and a few others Ace, Flu, Phe, and Ant. All these further suggest pyrogenic sources of PAHs in the soil.

3.7. Lifetime risk

The ILCR when the soil is ingested by an adult $ILCR_{ing}$ (Adult) is 1.28×10^{-7} while that of children was 1.031×10^{-6} (Table 6). The cancer risk through the dermal route ($ILCR_{derm}$ (Adult)) is 1.605×10^{-7} for adults, while that of children is 2.252×10^{-6} .

Nasher *et al.* (2013) reported a higher dermal risk value than the results from the present study. The human and animal skin sensitizers among the PAHs are anthracene and benzo(a)pyrene (IPCS 1998). If the soil dust is inhaled, the incremental lifetime cancer risk ($ILCR_{inh}$ (Adult)) is 1.092×10^{-5} , while that of children is 2.729×10^{-6} . PAHs can enter and exit the human body through breathing, and ingestion from water, soil, and food (ATSDR 1999; Nwaichi *et al.* 2020). The toxicity equivalent of soil (BaP_{Teq}) was evaluated with BaA

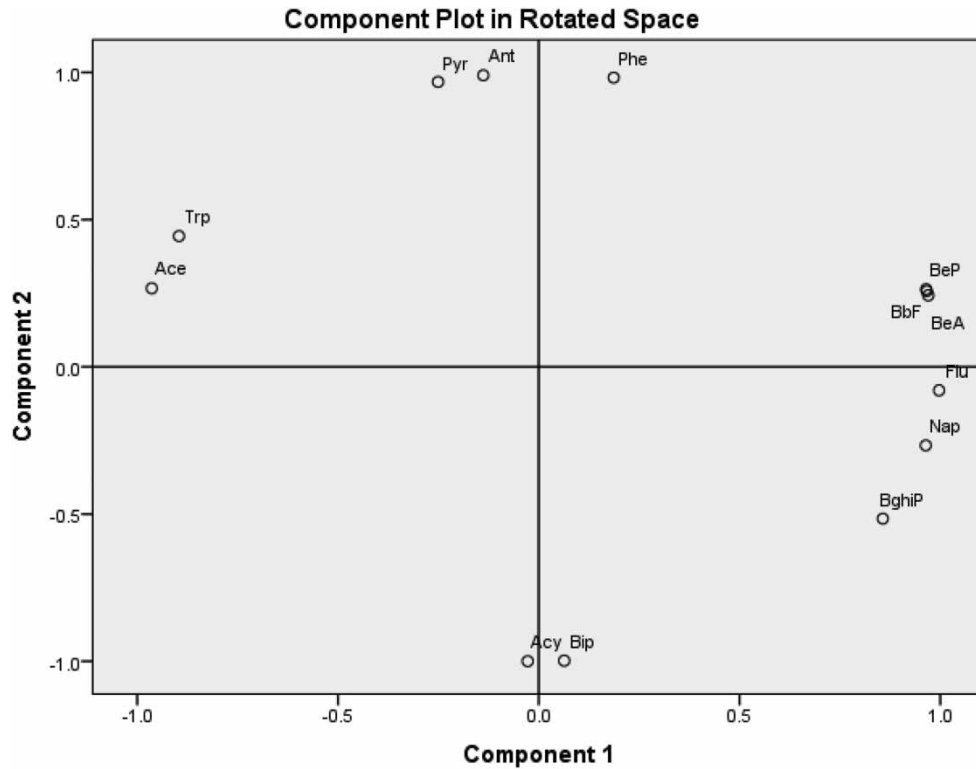


Figure 2 | Results of PCA analysis of PAHs in water of D-Bere.

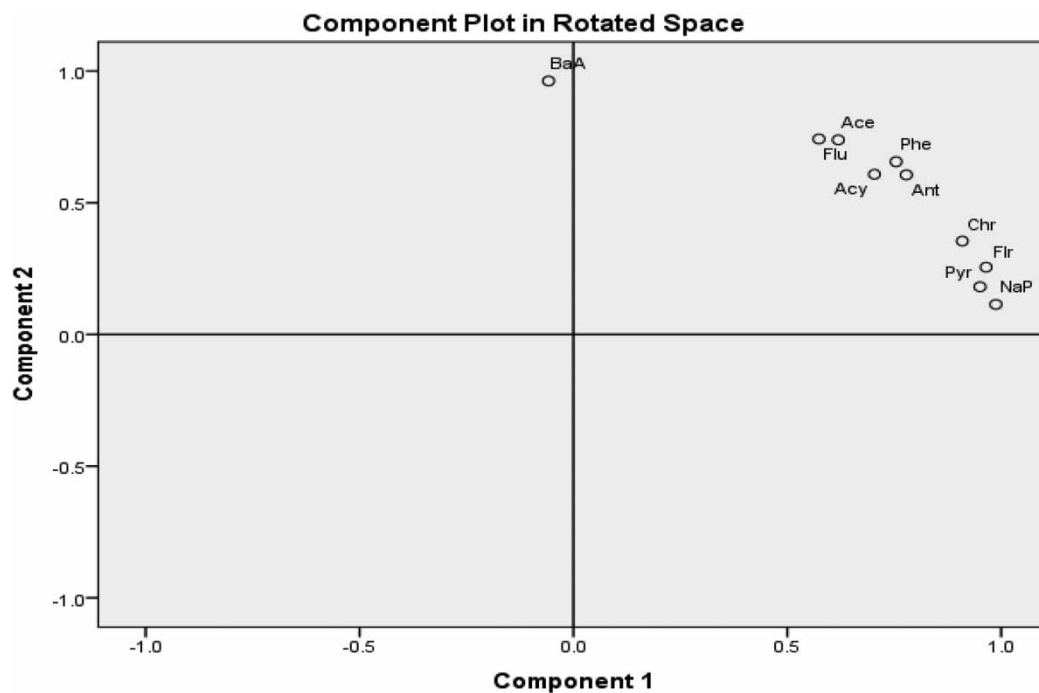


Figure 3 | Results of PCA analysis of PAHs in soil of D-Bere.

and Chr values of 1.495 and 0.0149, respectively, while mutagenic equivalents (BaP_{Meq}) are 1.225 and 0.254, respectively.

The results of PAHs in water samples showed that the ILCR when ingested by an adult $ILCR_{ing}(Adult)$ is 2.57×10^{-7} , while that of children $ILCR_{ing}(Children)$ is 2.05×10^{-6} (Table 6). The dermal version of it, when the water is used to bathe ($ILCR_{derm}(Adult)$) is 1.046×10^{-9} for adults, while that of children is 4.40×10^{-6} .

The ILCR of PAHs through inhalation by adults (ILCR_{inh}(Adult)) is 2.273×10^{-8} , while that of children is 5.39×10^{-9} . The toxicity equivalent of water (BaP_{Req}) was evaluated with BaA, BaP, and BbF values of 2.22, 0.253, and 24.7, respectively, while mutagenic equivalent (BaP_{Meq}) was 1.829, 6.325, and 24.7 respectively. The results from the present study with respect to ingestion and dermal risk agree with the results obtained by Yao *et al.* (2023), in the study of Don and Tangxum lake of China.

3.8. Percentage of PAHs garlic and moringa

Garlic and Moringa seed extracts were used for the treatment of water, while garlic and Fenton reagents were used for the treatment of the soil. The garlic extract removal of PAHs from water ranged between 0.14% for BbF and 100% (BaA, Tph, and BbT) (Table 7). The higher removal of BaA, Tph, and BbT is in agreement with the study of Gutierrez-Urbano *et al.* (2021).

Others are 0.64% NaP, 0.184% Ace, 0.734% Flu, 0.207% Ant, 0.171% Pyr, 0.28% Bip, 0.316% BeP, 0.14% BbF, and 2.5% Bghip. None of Acy and Phe is removed by garlic extract. Non-removal of phenanthrene might be due to its non-planar structure and its high stability (Rani & Shanker 2020). A higher success rate was recorded when PAH-contaminated water and soil samples were remediated using a tropical plant (*Eleocharis ochrostachys*) through a surface flow system (Alsbani *et al.* 2020). PAHs can be reduced by garlic, onions, shallots, and pepper (Neves *et al.* 2021). The ability to remove PAHs is due to the presence of a bioactive compound (allicin) in garlic (Janoszka *et al.* 2019; Saputro *et al.* 2022).

However, Moringa seed extract removal of PAHs ranged between 0.0001% for Bip and 100% for Bghip. Others are 0.139% Acy, 0.044% Ace, 0.126% Flu, 0.033% Ant, 0.033% Phe, 0.116% Pyr, 0.305% BaA, 0.045% Tph, 0.19% BeP, 0.059% BbF, and 0.338% BbT. Nap is not removed by the Moringa seed extract.

The results of treatment of polluted soil with the garlic extract showed removal of 1.34% NaP, 2.59% Acy, 0.429% Ace, 0.78% Flu, 0.177% Ant, 0.368% BaA, and 0.186% Flr. None of Phe, Pyr, and Chr is removed. The removal of PAHs using garlic extract acting as a chelating agent facilitates the removal of organic pollutants in the soil (Kiosnar *et al.* 2019).

Fenton reagents removed 1.45% NaP, 0.417% Acy, 0.265% Ace, 100% Ant, 100% Phe, 0.06% Pyr, 0.468% BaA, and 100% Chr. None of Flu and Flr is removed. The results of the percentage of PAHs removed from this study is lower than reported results from the literature (Jonsson *et al.* 2007; Nam *et al.* 2021). Phenanthrene and pyrene removal was achieved efficiently by graphene oxide-coated microbubbles (Yahya & Lau 2021). The electrophilic aromatic substitution mechanism was used to describe PAH oxidation and removal (Liu *et al.* 2020). Moringa seed and garlic extracts can be incorporated in water treatment that requires the removal of PAHs in water for their effectiveness in removing 100% of BaA, Tph, BbT, and Bghip.

4. CONCLUSION

The study investigated PAH levels in the B-Dere Community, which is an oil-producing area in Nigeria. The water and soil of the area were examined for their PAHs and their physicochemical parameters. The PAHs were removed using Moringa seed extracts, garlic extracts, and by the Fenton-oxidation chemical method. The results showed that B-Dere water contained a high concentration of chloride and 13 PAHs. The order of dominance of PAHs in water is benzo(ghi)perylene > benzo(b)fluorothane > benzo(e)pyrene > benz(a)anthracene > triphenylene > pyrene > phenanthrene > anthracene > fluorene > acenaphthene > biphenylene > acenaphthylene > naphthalene. The soil contains 10 PAHs in the following order: benz(a)anthracene > chrysene > pyrene > fluoranthene > anthracene > phenanthrene > fluorene > acenaphthene > acenaphthylene > naphthalene. The sources of PAHs in water are pyrogenic, while those of soil are petrogenic. The lifetime risk was higher in children than in adults if the water is ingested through drinking and if the water is used to bathe. However, the incremental lifetime risk of cancer through inhalation from the soil was higher for adults than for children. Garlic extract is effective in removing benz(a)anthracene, BaA, and benzo(b)fluorothane from water, while moringa extract is effective in removing biphenylene from water. The garlic is not as effective in removing PAHs from the soil as the Fenton oxidation reagents. The community may adopt the green method of water and soil treatment described in this study for better utilization of water and soil for agricultural purposes.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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