

Assessment of Levels of Polycyclic Aromatic Hydrocarbons in the Oil from Kuwait Oil Lakes

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Abstract. In an attempt to assess the potential impacts resulting from the oil fires started during the Gulf War, levels of polycyclic aromatic hydrocarbons (PAHs) were determined in Kuwaiti oil lakes. Samples were collected from two oil lakes within each of the four oil fields heavily damaged during the 1991 Gulf War. The oil samples were fractionated and the aromatic fraction analyzed quantitatively by gas chromatography/mass spectrometry (GC/MS). In addition, the effect of weathering on the PAH content was monitored over a 21-month period. The results indicated that total PAH contents initially ranged from 52.4 to 425.7 mg/kg in oil lakes. Phenanthrene and chrysene were the major PAHs in most of the samples. Benzo(a)pyrene was detected in all samples in varying amounts (0.46 to 5.3 mg/kg). Total PAH contents were higher, after 21 months, ranging from 59.55 to 616.9 mg/kg. Concentration of benzo(a)pyrene also increased considerably (from 0.16 to 22.2 mg/kg). The hazard potential of these compounds in Kuwait's environment was assessed and indicated that large amounts of the remaining oil will continue to have serious ecological implications unless immediate measures are taken to deal with the problem.

The environmental consequences of the Gulf War and its aftermath have been the subject of many recent studies (UNEP 1991; EPC 1991; Khordagui 1991; Greenpeace 1992). Many of these studies have been concerned with the marine ecosystem (Price and Robinson 1993; Readman *et al.* 1992); Sadiq and McCain (1993) estimated that at least 800 km of shoreline was oiled as a result of the Gulf War. They also summarized the adverse impacts of the oil slick to the intertidal communities, mangroves, salt marshes, subtidal communities, coral reefs, fish and shrimp eggs and larvae. Air pollution and fallout from the oil fires have been investigated (Johnson *et al.* 1991; Mulholland *et al.* 1991; Penner 1991). The ground level measurements indicated that the oil fires increased the ambient concentrations of inhalable air particulates (PM10) in Kuwait and

northeastern Saudi Arabia. These studies also established that the fires caused a significant increase in the concentrations of CO₂, SO₂, and NO_x. Analysis of PM10 and total suspended air particles (TSP) showed that the levels of Ni, Cd, V, Mo, Ti, and Cr were higher during oil fires as compared to the levels before. Concentration of total PAHs in PM10 air particulates were significantly higher compared with those from Saudi Arabia and Bahrain. Potential risks associated with the contaminated soil were assessed by Al-Awadhi *et al.* (1993), which indicated that PAHs together with trace metals may accumulate and migrate in the subsurface soil layers and in desert plants, creating potential hazard to ground water aquifers as well as to grazing animals. The possible serious impact of desert soil pollution on ground water aquifers was further discussed by Literathy (1992). The oil lakes, however, did not receive much attention.

A large number of oil wells in Kuwaiti oil fields were blown up along with other oil installations (crude oil gathering centers, oil pipelines, loading terminals etc.) in 1991 by the retreating Iraqi army. At the end of the war, a total of 616 wells were on fire, while 76 wells were destroyed and were gushing oil but did not catch fire. About 100 wells were damaged but did not leak any oil. As a result of this damage, large areas of desert were flooded with crude oil, together with partially burned crude oil, soot, and associated pollutants forming large pools, commonly known as "oil lakes." The formation of these oil lakes constitutes a unique event, as such a massive and widespread terrestrial oil spill has never occurred before.

Kuwait oil fields are generally categorized into northern (Raudatain and Sabriya) and southern (Greater Burgan, Managish, Umal Gudair and Wafra) oil fields (Figure 1). Greater Burgan consists of three distinct oil fields, namely Burgan, Ahmadi, and Magwa. Most of the damage was suffered by Greater Burgan, Raudatain, and Sabriya oil fields and most of the oil lakes were formed in these areas. Approximately 200 lakes were formed, covering a total of 27.2 km² (Al-Sulaimi *et al.* 1992). The lakes varied widely in location, shape, size, depth, and type of crude oil. Most of the lakes had an irregular shape and their depth initially varied from very shallow to 1.5 m. The revised estimates for the total amount of oil contained in lakes, according to the Ministry of Oil (Kuwait), was about 25 million barrels (Arab Times 1992).

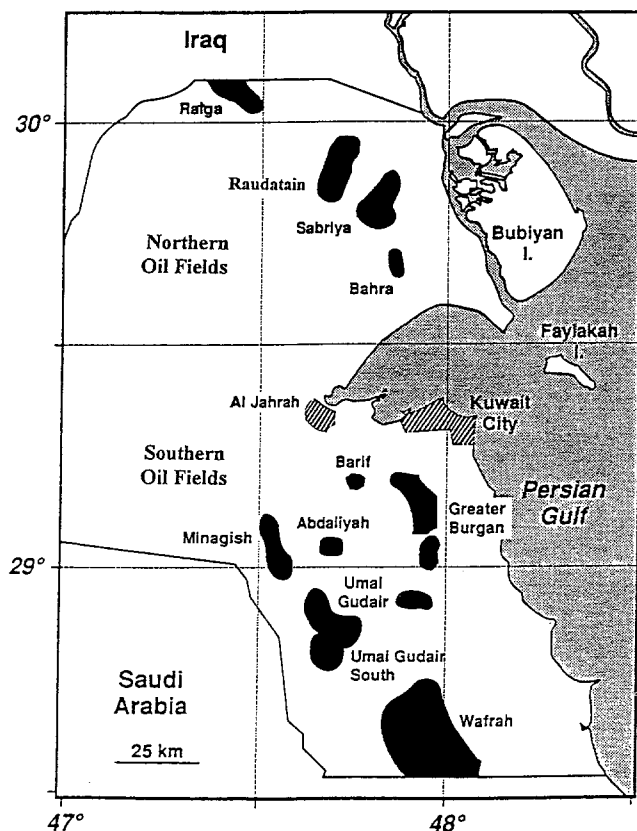


Fig. 1. Map of Kuwait showing the location of oil fields

The composition of the oil in different lakes varied greatly during their formation. Those formed around the wells that were damaged but did not catch fire contained mostly crude oil. On the other hand, lakes formed around the burning wells contained oil whose composition depended on many variables (crude oil composition, degree of combustion, surface temperature of the lake, fallout of the partially burned crude oil, and the amount of seawater used for fire-fighting).

The presence of such a large amount of oil, exposed to a hot climate and spread over a relatively large area, poses a potentially serious environmental hazard. It not only contaminates a vast area, but also threatens to pollute the groundwater, a scarce resource in arid country with no natural surface water. More important, these lakes could continue to pollute the air by emitting volatile compounds present in the oil. In addition, petroleum-derived compounds are produced over time by photooxidation, autooxidation, and biodegradation of the oil. Many of the lakes (in Burgan and Magwa oil fields) were in close proximity to population centers, whereas others (Raudatain and Sabriya) were upwind from Kuwait city.

Polycyclic aromatic hydrocarbons (PAHs) constitute the most toxic fraction of the oil, especially in partially burned residues (IARC 1983; Zedeck 1980; Shubik and Hartwell 1957). The objective of this study, therefore, was to assess the amounts of PAHs in lakes in order to provide useful information for hazard and risk assessment as well as for future studies dealing with long-term effects of these lakes and remedial measures for the restoration of contaminated soils.

Materials and Methods

Sample Collection

In September 1992, samples were collected from two lakes within each of the following oil fields: Greater Burgan, Magwa, Raudatain, and Sabriya. These four fields were the most heavily impacted. Sampling, from the same lakes, was repeated every 3 months for a period of 9 months. To study the quantitative changes in the PAH fraction of the oil in the lakes, PAH composition of the weathered oils was compared to the samples collected while the oil fires were still burning. This required the procurement of authentic samples of the oil from the lakes, stored properly. Unfortunately, these samples were not available.

However, early samples of oil from different lakes were obtained from the Petroleum Technology Department (PTD) of the Kuwait Institute for Scientific Research. These samples were collected in September, 1991 for some unrelated studies. Samples were kept in large containers at ambient temperature and mostly without airtight caps. Sampling of the oil in the two oil lakes from each of the four oil fields that suffered the most damage was undertaken. The samples were collected from the same lakes over a period of 21 months to appraise the effect of oil weathering on PAH content.

The sample collection procedure for early samples was adopted for all of the subsequent collections. Samples from the lakes were collected in precleaned glass vials (40 ml). The vial was allowed to dip in the oil, just below the surface, at 3 m from the edge of the lake. Oil was collected, avoiding any visible extraneous material. The vial was tightly closed with a teflon-faced screw cap, kept in an ice box, and transported to the laboratory where it was stored in a freezer at -4°C .

Fractionation of Oil

The oil samples were fractionated into different classes of compounds according to the method outlined by Romeu (1986). A 250 mg sample was accurately weighed in a 9 ml vial, 7 ml *n*-pentane was added, and the mixture was shaken vigorously for a few minutes to allow the oil to mix with the solvent. Insoluble asphaltene were settled by centrifuging the sample at 2500 rpm for 15 minutes. The supernatant pentane layer was removed carefully. The extraction was repeated 2 more times or until the pentane layer did not show any color. The pentane soluble extracts were concentrated to 1 ml and were fractionated on a chromatographic column (23 cm \times 0.8 mm ID). Silica gel (chromatography grade, 40–200 mesh) was activated at 130°C overnight and subsequently deactivated with 5% water. The first fraction was eluted with *n*-hexane (16 ml) and contained saturated hydrocarbons. Aromatics were recovered by eluting the column with 24 ml of hexane:chloroform mixture (1:1, v/v). For gas chromatographic analysis, the volume of aromatic fraction was reduced to 1 ml and an aliquot (0.1 μl) was injected, whereas for GC/MS analysis, the final volume was adjusted to 100 μl .

Gas Chromatographic Analysis

The aromatic fraction was analyzed by gas chromatography using a Varian gas chromatograph (GC-3400) equipped with a universal injector, flame ionization detector, and a fused silica capillary column (0.2 mm ID, 30 m length), coated with a SE-30 stationary phase. The injector temperature was 260°C , and the detector was kept at 280°C . The initial temperature of the column (80°C) was held for 2 min and then programmed to 260°C at $4^{\circ}\text{C}/\text{min}$. The final temperature was held for 10 minutes. Helium was used as carrier gas (flow rate: 2 ml/minute).

Table 1. List of PAHs and the ions monitored

PAH Compound	Abbreviation	Ion
Naphthalene	Naph	128
Acenaphthylene	Acey	152
Acenaphthene	Acen	154
Fluorene	Fluo	166
Phenanthrene	Phen	178
Anthracene	Anth	178
Fluoranthene	FlAn	202
Pyrene	Pyr	202
Benzo(a)anthracene	BaA	228
Chrysene	Chry	228
Benzo(b)fluoranthene	BbFl	252
Benzo(k)fluoranthene	BkFl	252
Benzo(a)Pyrene	BaP	252
Indeno(1,2,3-cd)pyrene	Ind	276
Benzo(ghi)perylene	BPY	276
Dibenzo(a,h)anthracene	Dibn	278

Gas Chromatography/Mass Spectrometry

Qualitative and quantitative analysis of the aromatic fraction was carried out on a Shimadzu bench-top GC/MS (QP-2000), using single ion monitoring (SIM). Gas chromatographic conditions were similar to those given for GC analysis, except the column used for GC/MS analysis was coated with a SE-52 stationary phase. The injection used the splitless mode. Injection volume was 1 μ l with injection port temperature of 280°C. The interface was heated to 280°C. The ion source temperature was kept at 260°C. Ionization energy in the mass spectrometer was 70 eV. The PAHs and the corresponding ions monitored are given in Table 1.

An external standard method was used for quantifying PAHs in the aromatic fraction. A commercial standard mixture of PAHs (PAH mixture 610-M, Supelco) containing the compounds listed in Table 1 was diluted so that the BaP concentration was 2 μ g/ml. The standard mixture was injected at the start of each working day and analyzed under the same GC conditions.

Quality Control

One oil lake sample from each of the four oil fields was duplicated as a quality control sample. These samples were processed and analyzed as independent samples. The percentage of recovery was established for each individual PAH by spiking some of the samples with known amounts of aromatic hydrocarbons, and ranged from 81.3% (naphthalene) to 91% [benzo(a)pyrene].

Results and Discussion

Immediately after the oil fires were put out, massive operations were underway to restore the oil production. Production areas were cleaned up and crude oil from the lakes was drained into a nearby lake or lagoon in order to prepare the sites for drilling. Many small lakes disappeared or were drained into larger lakes in these operations. The situation in the oil fields was very

dynamic over the study period and it was difficult to verify the authenticity of some of the samples (Sabriya 10).

In general, the oil thickened with time. The deepest lakes contained oil that flowed relatively easily, especially during the summer months when the temperature of the oil reached close to 75°C (ambient temperature up to 50°C), whereas the oil in very shallow lakes became very viscous.

Standard Kuwait crude oil contains 27% aromatics (Romeu 1986). During weathering, especially under very hot ambient conditions in Kuwait, volatile aromatics (benzene, toluene, ethylbenzene, xylenes, 1,3,5-trimethylbenzene, isopropylbenzene, and isopropyltoluenes) were quickly lost. In fact, gas chromatographic analysis (Figure 2) revealed that monoaromatics were not detected in most of the samples. High ambient temperatures and the intense heat of the oil fires resulted in complete loss of these compounds. As can be seen from these chromatograms, the aromatic fraction did not show volatile aromatic compounds. As a result, higher molecular weight aromatics were predominant in the aromatic fraction. More noticeable was the hump that indicated a large amount of unresolved complex mixture accounting for the bulk of the fraction.

PAH Levels in Oil Lakes

The results of GC/MS analysis showed that, in general, volatile PAHs (naphthalene, acenaphthylene, and acenaphthene) were not detected in any of the samples. For early samples, this was a little surprising. The possible explanations include exposure to high temperatures in and around the oil fields or inadequate sample storage. Subsequent samples did not show the presence of any of these compounds, probably due to evaporative losses.

Burgan Oil Lakes: PAHs levels in Burgan oil lakes are given in Table 2a. The total PAH content of Burgan 1 lake sample was relatively low (57.6–72.53 mg/kg). Phen and Fluo constituted the bulk of PAH content. Benzo(a)pyrene (BaP), a potent carcinogenic PAH (IARC 1987), was present in relatively small amounts; its concentration after 12 months was unexpectedly low. The apparent trend from these results indicated that lower molecular weight compounds decreased with time, while higher molecular weight compounds increased in relative concentration as the oil weathering progressed. Total PAH levels increased from 57.6 mg/kg to 71.3 mg/kg over the 21 months.

The PAHs contents of the oil sample from lake Burgan 6 were also low. Phen and Chry were the main components. Levels of BaP were significantly higher than those of Burgan 1. The weathering pattern observed for this lake was similar to that of Burgan 1. The total PAH content increased from 77.9 mg/kg to 100.7 mg/kg.

Magwa Oil Lakes: The total PAH contents were in the same range as those of Burgan oil field lakes (Table 2b). Phen constituted the bulk of total PAHs. BaP was present in relatively low concentration. The weathering pattern of PAH fraction of the oil from this lake was different from samples from other lakes. The total PAH contents decreased (24.7% loss) over 21 months. The reason for this decrease is the larger percentage of most volatile PAHs lost over the 21 months. In contrast, relatively higher molecular-weight PAHs increased with time (e.g., BaP increased from 0.7 to 1.29 mg/kg).

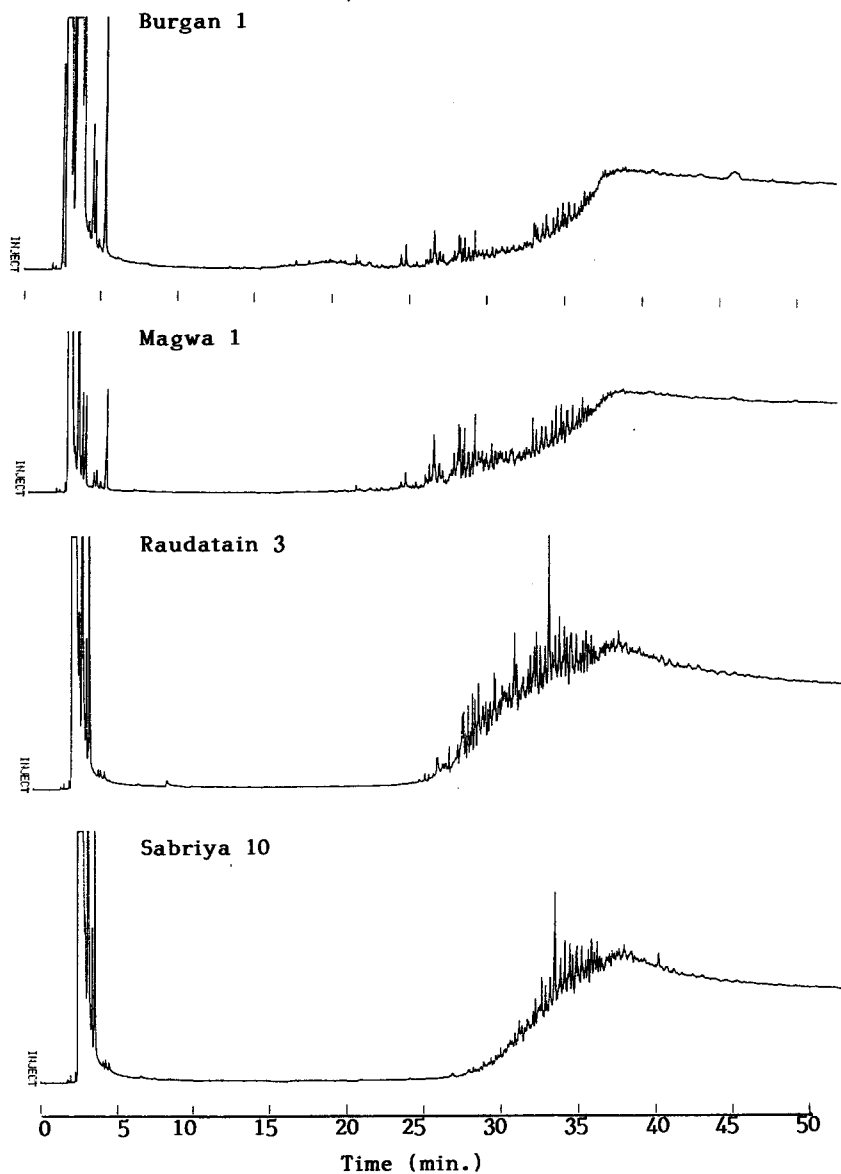


Fig. 2. GC profile of aromatic fraction of some selected samples (Batch 1)

PAH composition of the oil from Magwa oil lake 11, on the other hand, was distinctly different. The PAH contents were much higher, with Phen and Chry as major components. All other PAHs found were also at high levels. Weathering affected the PAHs in the oil in a similar way as the sample from lake Magwa 1, i.e., the volatile PAHs (Fluo and Phen) decreased with time, while higher molecular weight PAHs increased. BaP increased from 5.3 to 14.4 mg/kg over the study period.

Raudatain Oil Lakes: The results of PAHs from the oil from Raudatain oil lakes are given in Table 2c. The total PAH contents for the early samples of Raudatain 3 lake were remarkably low as compared to the subsequent samples. The overall PAH levels were not very high. Phen and Chry showed much higher concentration as compared to other PAHs. BaP levels were relatively very low when compared to the oil from other lakes. The oil weathering caused a significant increase in the concentration of all PAHs present. Even the volatile fraction of PAHs considerably increased. The PAH contents of the oil from lake Raudatain 1 were distinctly higher than that of Rau-

datain 3 lake. Phen and Chry were the main constituents of this fraction. The weathering pattern for this sample was similar to that of the Raudatain 3 sample.

Sabriya Oil Lakes: The concentrations of different PAHs in the oil from lake Sabriya 5 are given in Table 2d. The overall levels of PAHs in the sample were moderate. As in other samples, Phen and Chry formed the bulk of PAHs. Benzo(a)pyrene was detected in appreciable concentrations (4.8 to 8.2 mg/kg). The weathering trend indicated that the more volatile PAHs (Fluo and Phen) decreased with time, while the higher molecular weight compounds increased progressively. The total PAH contents increased by 17.65% over the 21-month period.

The oil from lake Sabriya 10 was unusual in many respects (Table 2d). First, the total PAHs found in this sample were much higher compared to the samples collected from the oil lakes of other oil fields. Apart from increased levels of the components, Chry showed very high levels. BaP was also present in much higher concentration (up to 22 mg/kg). Secondly, higher PAHs (Ind and BP_y) were also detected in the last

Table 2. Levels of PAHs (mg/kg) in the oil from different lakes**Table 2a.** Burgan Field

PAHs	Lake Burgan 1 Weathering Time (months)					Lake Burgan 6 Weathering Time (months)				
	0	12	15	18	21	0	12	15	18	21
Fluo	14.67	11.68	8.81	7.84	6.60	7.9	13.6	13.1	10.5	6.4
Phen	28.4	41.96	46.14	43.49	46.21	24.0	35.4	33.5	31.8	22.0
FlAn	3.11	3.26	3.19	3.20	3.32	3.7	4.1	4.0	4.4	4.3
Pyr	4.68	4.74	4.99	4.83	4.93	4.3	2.4	5.4	5.7	5.9
Chry	5.74	8.18	8.21	8.59	8.72	26.3	32.3	34.6	36.6	38.1
BbFl+BkFl	0.54	0.73	0.73	0.81	0.84	7.1	12.5	13.0	15.1	17.0
Bap	0.46	0.16	0.46	0.5	0.64	4.5	6.5	6.7	6.8	7.0
Total	57.6	70.69	72.53	69.26	71.26	77.9	106.9	110.3	111.0	100.7

Table 2b. Magwa Field

PAHs	Lake Magwa 1 Weathering Time (months)					Lake Magwa 11 Weathering Time (months)				
	0	12	15	18	21	0	12	15	18	21
Fluo	14.97	4.95	4.76	4.24	3.93	34.50	18.7	16.5	15.4	10.5
Phen	44.28	37.47	35.58	37.51	32.03	92.7	82.5	84.8	66.2	41.1
FlAn	3.66	3.31	3.6	3.86	4.61	14.0	15.6	22.1	24.0	24.2
Pyr	5.28	5.32	5.69	5.66	5.77	20.1	23.1	22.5	27.5	28.2
Chry	8.47	8.70	8.88	9.00	9.22	114.6	174.9	268.9	270.1	282.3
BbFl+BkFl	1.73	1.73	2.31	2.49	2.7	12.5	19.0	19.5	20.2	23.7
Bap	0.7	1.11	1.13	1.24	1.29	5.3	6.1	9.4	12.0	14.4
Total	79.09	62.60	61.95	64.00	59.55	293.8	339.9	443.7	435.7	424.5

Table 2c. Raudatain Field

PAHs	Lake Raudatain 3 Weathering Time (months)					Lake Raudatain 1 Weathering Time (months)				
	0	12	15	18	21	0	12	15	18	21
Fluo	3.8	15.9	12.6	10.9	8.3	7.9	12.6	11.8	11.6	11.1
Phen	27.3	58.4	59.6	67.0	67.6	44.2	79.8	78.5	97.3	98.4
FlAn	2.4	5.2	5.5	6.4	6.6	8.7	20.1	20.3	24.2	26.2
Pyr	2.1	6.3	7.9	8.8	8.9	12.1	21.5	21.8	31.2	35.0
Chry	11.9	21.9	22.1	25.1	26.9	19.5	83.9	89.3	94.2	101.2
BbFl+BkFl	3.8	4.0	4.6	6.9	7.8	7.8	15.3	16.5	26.5	26.8
Bap	1.2	1.2	2.4	2.7	2.9	1.1	4.4	5.6	6.8	6.8
Total	52.4	112.8	114.5	127.8	129.0	101.4	237.6	243.9	291.7	305.4

Table 2d: Sabriya Field

PAHs	Lake Sabriya 5 Weathering Time (months)					Lake Sabriya 10 Weathering Time (months)				
	0	12	15	18	21	0	12	15	18	21
Fluo	20.97	12.3	5.5	4.40	4.20	20.50	31.3	6.6	3.4	1.2
Phen	69.4	69.40	69.10	49.40	45.90	70.9	75.0	95.9	55.1	4.9
FlAn	9.0	12.1	14.4	15.3	16.3	12.4	43.6	56.6	44.9	31.9
Pyr	10.3	18.4	16.1	16.0	16.0	24.3	87.4	78.3	71.1	58.8
Chry	49.1	77.8	85.3	86.5	97.9	270.9	320.9	334.7	388.4	380.4
BbFl+BkFl	12.1	19.8	18.0	17.4	18.1	23.8	18.5	55.0	64.7	76.1
Bap	4.8	5.0	7.5	7.2	8.2	3.0	6.7	19.7	21.4	22.2
Ind								20.0	19.9	19.5
BPY								24.4	22.2	21.9
Total	175.6	214.8	215.9	196.3	206.6	425.7	583.4	691.2	691.1	616.9

Table 3. Estimated amounts of oil and PAHs in the lakes

Lake	Area (m ²)	Initial amount oil (tons)	Initial PAHs (kg)	Expected leftover oil (tons)	Expected leftover PAHs (kg)
Burgan 1	520,000	520,000	29,952	31,200	2,134
Burgan 6	840,000	420,000	32,718	50,400	3,295
Magwa 1	102,000	65,000	5,194	6,120	309
Magwa 11	280,000	450,000	132,210	16,800	56,123
Raudatain 1	180,000	63,000	3,301	10,800	426
Raudatain 3	2,500	1,500	152	150	46
Sabriya 5	15,000	3,000	527	900	109
Sabriya 10	300,000	60,000	25,542	18,000	15,757
Total	2,239,500	1,582,500	229,596	134,370	78,200

three samples. These PAHs were not detected in any sample from other lakes. The fluctuation in concentrations of Phen, FlAn, and Pyr were also unusual. For example, levels of Phen increased slowly from the first to the second sample (over 12 months). In sample 3, its concentration increased sharply and then quickly decreased over the next 3 months. These unexpected results indicate strongly that different oils were mixed in the lake about the time sample 3 was collected (December 1992).

Hazard Potential

It is apparent from the above results that oil lakes represent a potentially serious source of hazardous pollutants to the environment. Since November, 1991, when the last of the oil fires were extinguished, a large amount of oil spilled into the lakes has been removed. However, as a result of three years of weathering, the oil has thickened and is not easy to remove completely. In fact, the field observations indicate that a layer of 5–20 cm is left, which consists mostly of thickened oil with some suspended soil. The oil content of the leftover layer may be as high as 90%. On average, the oil makes up about 60% of this sludge-like material (Dr. Kazmer Puskas, KISR, personal communication). Table 3 lists the estimated total amount of PAHs present in the oil at the time of first sampling for each of the lakes investigated. The total amount of PAHs remaining after the oil has been removed has also been estimated. These calculations indicate that lakes Magwa 11 and Sabriya 10 would leave the largest amount of PAHs. Assuming the oily layer to be constant, the total oil and consequently, PAHs, leftover depend on two variables, i.e., area of the lake and the concentration of PAHs in the oil. Therefore, a larger lake whose PAH concentration is relatively low would leave relatively smaller amounts of PAHs. Lake Burgan 1, Burgan 6, Magwa 1, and Raudatain 1 are good examples of these.

Similarly, the amount of oil leftover from all of the oil lakes can be estimated roughly by assuming an average layer of 10 cm and average oil content of 60%. The calculations indicate that approximately 1.55 million tons of oil will be left in the lake beds. Average total PAH content of the oil from the lakes is 240 mg/kg. Therefore, the total amount of PAH in the remaining oil will be approximately 372,096 kg (about 372 tons).

The potential impacts of such a large amount of hazardous pollutants to the environmental health and ecology are many fold. First, after prolonged weathering and as a result of wind erosion, the dust contaminated with the oil may be resuspended

in the air and expose populated areas (Al-Yakoob and Saeed 1994). Secondly, the weathered oil may undergo photochemical changes and biodegradation and release degradation products into the environment. The kinetics, magnitude, and toxicology of degradation products needs to be studied. Thirdly, the contaminated soil may be transported over long distances with time as a result of sand and wind movement, thus polluting a much larger area. This may expose the fauna and flora of the affected region for some time to come. Lastly, fresh ground water aquifers in northern Kuwait are under serious threat from the contaminants leaching from the oil lakes. Raudatain and Sabriya oil lakes are present in the catchment areas of Raudatain and Um al Aish fresh water fields. Low pollution levels in these aquifers have already been detected (Al-Sulaimi *et al.* 1993). As the average rain fall in Kuwait is very low (120 mm/year), it may be many years before significant levels of pollution may be detected. A study to assess the long-term pollution of ground water caused by the events of 1991 is currently under way.

At present, efforts are underway to find a suitable technique for the restoration of the soils contaminated as a result of the atmospheric fallout of the oil well fires. Safe disposal of leftover oily sludge from the oil lakes merits immediate attention as it has the potential for serious long-term environmental consequences.

Conclusions

Analysis of PAHs in the oil samples from different lakes over a 21-month period indicate that different lakes contained PAHs in different concentrations. PAH content of the oil lakes increased considerably over the study period. Benzo(a)pyrene, present in all samples, also increased significantly. Calculations indicate that large amounts of the oil will remain after the lakes have been drained. This would leave large amounts of PAHs in the environment, which may have long-term adverse effects on the population as well as on the ecosystem and the groundwater. The magnitude of the problem calls for immediate measures to deal with this issue of serious ecological implications.

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