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Investigation of polar and nonpolar material in the groundwater of Raudhatain and Umm Al-Aish fresh groundwater fields of Kuwait

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ABSTRACT

Parts of the fresh groundwater of Raudhatain and Umm Al-Aish freshwater fields have been contaminated with petroleum hydrocarbons leached from petroleum lakes and petroleum-contaminated soil resulting from the 1991 Gulf War. Pump and treat methodology has been suggested for treatment of contaminated groundwater. Selection and design of methodologies for the treatment of contaminants require knowledge related to types and physiochemical properties of contaminants. Therefore, as a preliminary step toward treatment of contaminated water, a study was conducted in order to identify and quantify contaminants in terms of polar and nonpolar material of contaminated water. Groundwater samples were collected from 10 monitoring wells (nine contaminated and one uncontaminated) of water fields. Collected samples were analyzed for total petroleum hydrocarbon (TPH), total organic carbon (TOC), phenol, tannin and lignin, volatile acids and Ultraviolet (UV)-induced fluorescence before and after activated silica gel. The silica gel splits aggregated polar and nonpolar material by retaining polar material on its activated surfaces. The results of the study indicated that both polar and nonpolar materials are present in the contaminated groundwater. TPH, UV-induced fluorescence, phenol, and volatile acid contents were dominantly composed of polar material at 86%, 84%, 73%, and 90%, respectively; whereas TOC, tannin and lignin contents were dominantly composed of nonpolar material at 58% and 60%, respectively. The results suggest that treatment techniques specific for polar and nonpolar contents will be required for the treatment of petroleum hydrocarbon-contaminated groundwater.

Keywords: Fresh groundwater; Contamination; Petroleum hydrocarbons; Treatment

1. Introduction

The Raudhatain–Umm Al-Aish area is situated in the northeastern part of Kuwait. Two oil fields, namely, Raudhatain and Sabriya, are located in the Raudhatain– Umm Al-Aish area (Fig. 1). Sole fresh groundwater resource of country is also located under topographic depressions of Raudhatain and Umm Al-Aish areas within and in the vicinity of Raudhatain and Sabriya oil fields. During the 1991 Gulf War, a total of 613 oil wells were set on fire by the retreating Iraqi troops. Out of these 613 wells, 101 wells were in Raudhatain and Sabriya oil fields. The resulting spillage of huge volumes of crude oil from oil wells gave rise to oil lakes and crude oil-impregnated soil (generally known as sludge). Moreover, products of crude oil combustion spread over a large area in the vicinity of the oil fields, causing widespread contamination of the soil. Oil lakes, sludge, and products of combustion are sources of contamination for the fresh groundwater source of the country. During rainy seasons, these sources of contamination generate petroleum hydrocarbon-contaminated runoff that infiltrated into groundwater and percolated into the fresh groundwater through unsaturated zone of pebbly, gravely, silty, and slightly clayey sand intercalated lenses of clay.

Studies conducted so far in this area, to assess the impact of sources of contamination on fresh groundwater, have

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focused only on the detection and quantification of nonpolar hydrocarbons, such as total petroleum hydrocarbon (TPH), polycyclic aromatic hydrocarbons (PAH) and benzene, toluene, ethylbenzene, and xylene (BTEX) in groundwater, as they are toxins and carcinogenic. However, the detection and the quantification of polar material are equally important not only to assess pollution levels but also for modeling the transport processes in the aquifer to forecast the pollution prospects in the long-term (Al-Awadi et al., 2006), as well as for the selection and designing of methodologies for the treatment of contaminants. Knowing the types and physicochemical properties of contaminants that are present would allow most treatment methods to be selectively eliminated without spending significant amounts of time and money on feasibility studies (Nyer, 1993).

The present study attempts to identify and quantify contaminants in terms of polar and nonpolar materials in the petroleum hydrocarbon-contaminated groundwater of the Raudhatain–Umm Al-Aish fresh groundwater fields.

2. Materials and methods

A total of 10 groundwater monitoring wells in the Raudhatain and Umm Al-Aish fresh groundwater fields were sampled for an investigation of the polar and nonpolar materials in the groundwater. The locations of the wells are depicted in Fig. 1. The wells P1, P12, P34R and P36R are located in Raudhatain fresh groundwater field, whereas, the wells P17, P18, P19, P33UA, P25UA and P27UA-1 are located in Umm Al-Aish fresh groundwater field. Details on the wells are presented in Table 1.

Before the collection of groundwater samples, the wells were purged. Grundfos MP1 submersible pump was used to purge the monitoring wells. During the well purging, temperature, pH, electrical conductivity (EC), dissolved oxygen (DO), and oxidation–reduction potential (ORP) of the produced water were measured at appropriate intervals using standard probes attached to a flow-through cell. After purging three volumes of the water in the well and stabilization of pH and EC values of groundwater, the groundwater samples were collected in laboratory-cleaned, sterilized ambercolored glass bottles. Groundwater recharge to the wells P12, P34R, and P36R was too low to purge using Grundfos MP1 submersible pump, even at its minimum pumping capacity. Therefore, the groundwater samples from these wells were collected using Teflon bailers.

Quality control-quality assurance (QA/QC) samples, field replicates and field blanks (FBs) were also collected. Field replicates, P33UA, and P33UA-D were collected from Well P33UA from the Umm Al-Aish fresh groundwater field. Samples to be used for TPH analysis were preserved with diluted (1:1) hydrochloric acid (HCl) to bring the pH of the sample to below 2 in order to cease possible degradation of biodegradable material. The samples were then packed into ice boxes to reduce and maintain their temperature at 4°C.

The collected groundwater samples were analyzed for total organic carbon (TOC), TPH, ultraviolet (UV)-induced fluorescence emission, phenol, tannin, and lignin and volatile acids. The analysis for TOC, TPH, UV-induced fluorescence emission, phenol, tannin and lignin, and volatile acids was carried out in the laboratories of the Water Research Center (WRC); whereas analysis for VOCs was carried out at the Central Analytical Laboratory of Kuwait Institute for Scientific Research (KISR). Table 2 presents the analytical methods and instruments used for analyses of groundwater samples.

The samples and/or the extract of the samples were treated (mixed) with silica gel to split the polar and nonpolar materials in the samples. Silica gel (silicon hydroxide



Fig. 1. Locations of the monitoring wells in the Raudhatain-Umm Al-Aish area.

				G. Elevation	Screen
Well No.	Location	North (m)	East (m)	a msl	Interval m bgl
P1	Raudhatain	3,314,176.18	755,579.84	37.88	29.00-34.00
P12	Umm Al-Aish	3,308,251.85	761,864.59	36.92	32.00-37.00
P17	Umm Al-Aish	3,303,177.53	771,147.22	27.21	15.00-20.00
P18	Umm Al-Aish	3,301,243.66	770,052.68	27.35	15.00-43.00
P19	Umm Al-Aish	3,301,953.06	770,678.21	26.76	16.00-21.00
P25UA	Umm Al-Aish	3,300,638.69	770,650.27	26.50	16.00-19.00
P27UA-1	Umm Al-Aish	3,301,306.88	770,417.23	26.99	16.50-19.50
P33UA	Umm Al-Aish	3,301,341.79	770,175.17	26.47	17.50-20.50
P34R	Raudhatain	3,309,584.02	761,254.53	40.25	33.00-36.00
P36R	Raudhatain	3,308,813.19	761,617.55	42.44	32.00-35.00

Table 1		
Details on	the monitoring	wells

Note: R: Raudhatain; UA: Umm Al-Aish.

Table	2					
Analy	tical methods	used for a	nalysis of	groundwate	r samp	oles

Parameter	Method	Instrument
TOC	USEAP Method 5310B	TOC analyzer
TPH	USEPA Method 418.1	FT/IR Spectrometer
Fluorescence	Saenz et al. 1991	Spectrofluorophotometer
Phenol	4-Aminoantipyrine method	DR/2000 Spectrophotometer
Tannin and lignin	Tyrosine method	DR/2000 Spectrophotometer
Volatile acids	Esterification method	DR/2000 Spectrophotometer

[SiOH]) is a chemically inert, nontoxic, polar, amorphous form of silicon dioxide (SiO₂). In analytical chemistry, silica gel has been used to separate polar material from solutions. For example, USEPA Methods 418.1 (1993) (Total Petroleum Hydrocarbon) and 1664 (Oil and Grease) (2009) prescribe the use of silica gel to remove polar material from samples to be analyzed for TPH. Similarly, Method 5520 F (hydrocarbons) (APHA, 1998) also prescribes the use of silica gel to remove polar materials. It states that if a solution of hydrocarbons and fatty materials in a nonpolar solvent is mixed with silica gel, the fatty acids are removed selectively from the solution. The materials not eliminated by silica gel adsorption are designated as hydrocarbons. The highly active polar surfaces of the silica gel adsorb polar material by the means of hydrogen or dipole-dipole interaction that binds material onto the surface of the silica gel.

To isolate nonpolar material and measure it as TOC, the water samples were passed through silica gel (4 g) taken into a glass column. The effluent collected, 10 mL, was then analyzed for TOC using a TOC analyzer. To split nonpolar material and to measure it as TPH, the carbon tetra chloride (CCl_4) extracts of the water samples were mixed with 1.5 g of silica gel for 5 min using Teflon-coated magnetic stirrer. The silica-gel-treated extracts were then analyzed for TPH using Fourier transform infrared (FTIR) spectrometer. Same silica-gel-treated extracts of the samples were exposed to 265 nm of UV light in order to estimate the quantity of nonpolar material using a spectrofluorophotometer. To isolate

the nonpolar material and measure it as phenol, tannin and lignin, and volatile acids, 100 mL of the sample was mixed with 3 g of silica gel in capped bottles for 15 min using a shaker. The silica-gel-treated water sample was then filtered through Whatman No. 42 filter paper and analyzed for phenol, tannin, and lignin, and volatile acids.

3. Results

Mixing of water from different strata in a well, and in some instances, exposure of the water to the atmosphere may induce chemical instability, even though the original water in place is in equilibrium with its surroundings. This chemical instability may cause changes in certain constituents and requires sample preservation or on-site determination of certain properties of the water (Hem, 1985). Temperature, pH, DO, ORP, CO₂, and alkalinity are so closely related to the environment of the water that they are likely to be altered during sampling and storage. Therefore, meaningful values of these quality parameters can be obtained only when they are measured immediately on-site. Therefore, after completion of well purging, pH, EC, DO, and ORP were measured during purging at appropriate intervals and at the end of well purging. The final values of these parameters are presented in Table 3.

The concentrations of combined polar and nonpolar material in the groundwater samples are presented in Table 4. The concentration of nonpolar material represented the

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Table 3	
Groundwater quality parameters measured o	on site

Well No.	Temperature (°C)	pН	EC (µS/cm)	DO (mg/L)	ORP (mg/L)	Odor
P1	28.4	7.82	698	5.33	15.4	_
P12	28.0	6.80	5,240	0.00	-192.4	H_2S
P17	28.0	6.90	900	0.00	-30.4	Hydrocarbon
P18	27.6	6.50	8,190	0.00	-78.8	Hydrocarbon
P19	29.0	6.72	2,753	0.00	-51.1	-
P25UA	28.4	6.98	3,230	5.29	40.0	-
P27UA-1	29.0	6.67	6,750	0.00	-34.0	Hydrocarbon
P33UA	27.7	6.91	10,130	0.00	-56.7	Hydrocarbon
P34R	28.0	7.87	4,650	6.36	41.8	-
P36R	28.0	7.54	12,880	3.24	91.0	-
FB	28.0	7.83	10.50	3.07	2.15	-

EC = electrical conductivity, DO = dissolved oxygen, ORP = oxidation reduction potential.

material that remains in the water sample and/or in the extract of the sample after treatment with silica gel. The concentrations of polar material represent the material removed (adsorbed) by silica gel from the water sample and/ or from the CCl_4 extract of the sample. The concentration of polar material was obtained by subtracting the concentration of polar and nonpolar material in the sample and/or extract of the sample before silica-gel treatment. Table 5 presents minimum, maximum, and average (%) concentrations of TOC, TPH, UV-induced fluorescence, phenol, tannin and lignin, and volatile acids in terms of polar and nonpolar material as percentage.

3.1. Polar and nonpolar materials as total organic carbon

The concentrations of polar and nonpolar materials as TOC in the groundwater samples from the Raudhatain–Umm Al-Aish fresh groundwater field are presented in Table 4 and plotted in Fig. 2. The TOC (polar and nonpolar) of the groundwater samples ranged between 0.53 and 38.12 mg/L, with a mean value of 12.29 mg/L.

The results with regard to polar and nonpolar materials as TOC indicated material in the groundwater dominance of nonpolar material (58%) compared with polar material (42%; Table 5). Polar material as TOC was not detected in the groundwater samples from the monitoring wells P1, P12, P25UA, P34R, and P36R.The slight higher amount of nonpolar material as TOC in the samples from monitoring wells P1, P12, P34R, and P36R may be attributed to leaching of trace amounts of organics either from the silica gel or from the glass wares during testing.

3.2. Polar and nonpolar materials as total petroleum hydrocarbon

TPH is the total concentration of the hydrocarbons extracted and measured in groundwater samples. The concentrations of polar and nonpolar materials as TPH in the groundwater samples are presented in Table 4 and plotted in Fig. 3.

The combined concentration of polar and nonpolar material in the groundwater ranged between 0.06 and 2.21 mg/L, with a mean value of 0.75 mg/L. The overall comparison of polar and nonpolar materials indicated that the groundwater was dominantly composed of polar material (86%) as compared with nonpolar material (14%) (Table 5). However, it was dominantly composed of nonpolar material (67%) when compared with polar material (33%) in the groundwater samples from the wells P1.

3.3. Ultraviolet-induced fluorescence

UV-induced fluorescence intensity is a qualitative measure used for identification of the presence of organic contaminants (Al-Awadi et al., 2001). Fluorescence is extremely sensitive and many factors (type of solution, pH, ionic strength, temperature, ORP, and interactions with metal ions and organic substances) affect its emission (Senesi, 1990). Of the dissolved organic matter, only 40% to 60% was fluorescent; this fluorescent material principally comprised of protein and organic acids (Senesi, 1993).

The UV-induced fluorescence intensities due to polar and nonpolar materials in the CCl_4 extracts of the samples are presented in Table 4. Fluorescence was not detected in the CCl_4 extracts of water samples from wells P1, P12, and P34R. A high fluorescence intensity indicated a high concentration of the material. The comparison indicated that the material in the groundwater samples was dominantly composed of polar material (84%) as compared with nonpolar material (16%; Table 5). While it was dominantly composed of polar material in the groundwater samples of the monitoring wells P17, P18, P19, P27UA-1, and P33UA, it was entirely of polar material in groundwater samples from wells P25U and P36R. A comparison of fluorescence intensities of groundwater samples is presented in Fig. 4.

3.4. Polar and nonpolar materials as phenol

Phenols are organic compounds which contain a hydroxyl (–OH) group attached to a carbon atom in a benzene ring or in a more complex aromatic ring system. The simplest member of this group is phenol. It is also known as carbolic acid, benzenol, phenylic acid, hydroxybenzene, and phenic acid. Due to hydrogen bonding, most low-molecular-weight

Polar and nc	npolar mí	aterials in	l ground	łwater sam	iples													
Well	Ē	OC (mg/l	L L	E	l/gm) Hc	Ē	Fl	lorescene	e	μJ	enol (mg	L)	Tanni	in and li (mg/L)	gnin	Volatil	e acids (n	ıg/L)
No.	P&NP	Ъ	NP	P&NP	Ь	ЪР	P&NP	Ь	NP	P&NP	Ь	NP	P&NP	Ъ	NP	P&NP	Ъ	NP
P1R	0.74	I	0.80	0.06	0.02	0.04	I	I	I	I	I	I	I	I	I	I	I	I
P12R	1.31	I	1.40	0.20	0.17	0.03	I	I	I	0.016	0.016	0.000	2.40	1.60	0.80	27.00	2.00	25.00
P17U	10.61	6.50	4.11	0.42	0.37	0.05	3.23	2.39	0.84	I	I	I	0.10	0.10	0.00	10.00	10.00	0.00
P18R	38.12	31.69	6.43	2.21	2.19	0.02	13.05	9.51	3.54	0.019	0.019	0.000	2.80	0.20	2.60	24.00	24.00	0.00
P19R	11.80	8.42	3.38	0.38	0.34	0.04	4.53	3.24	1.29	0.001	0.001	0.000	0.60	0.10	0.50	18.00	18.00	0.00
P25UA	0.69	I	0.75	0.11	0.08	0.03	2.24	2.24	I	I	I	I	I	I	I	I	I	I
P27UA-1	21.89	17.57	4.32	1.45	1.38	0.07	15.51	13.53	1.98	0.034	0.020	0.014	1.90	0.50	1.40	24.00	23.00	1.00
P33UA	24.79	20.01	4.78	1.63	1.54	0.09	16.48	13.69	2.79	0.005	0.001	0.004	3.20	1.00	2.20	1.00	1.00	0.00
P33UA-D	24.15	19.48	4.67	1.54	1.47	0.07	16.63	14.08	2.55	0.016	0.005	0.011	3.20	1.00	2.20	1.00	1.00	0.00
P34R	0.53	I	0.60	0.10	0.09	0.01	I	I	I	I	I	I	0.50	0.10	0.40	2.00	2.00	0.00
P36R	0.59	I	0.61	0.13	0.13	I	1.29	1.29	I	0.001	0.001	0.000	0.80	0.50	0.30	1.00	1.00	0.00
FB	0.37	I	0.50	0.23	0.16	0.07	I	I	I	I	I	Ι	I	Ι	Ι	I	I	Ι
Note: P&NP:	Polar and r	nonpolar;	P: polar;	: NP: nonpc	olar. –: no	t detecte	.pa											



Fig. 2. Polar and nonpolar materials as total organic carbon.



Fig. 3. Polar and nonpolar materials as total petroleum hydrocarbon.



Fig. 4. Comparison of fluorescence intensities.

phenols are water-soluble (i.e., polar) while high-molecular weight phenols are less water-soluble (i.e., nonpolar).

Phenol and its structurally related compounds are toxic at relatively low concentration and have been listed as priority pollutants by USEPA. Natural waters normally contain less than 1 µg/L (0.001 mg/L), but concentrations up

Table 4 Polar an

to 20 μ g/L (0.02 mg/L) may occur in some areas (Rittmann and McCarty, 2001).

The observed concentrations of phenol (polar and nonpolar) and phenol (as polar) and phenol (as nonpolar) materials in the groundwater are presented in Table 4 and plotted in Fig. 5. The concentration of phenol in the groundwater samples ranged between 0.001 and 0.034 mg/L, with a mean value 0.013 mg/L. Phenol was not detected in groundwater samples from the wells P1, P17, P25UA, and P34R. The monitoring well P1 is located at uncontaminated site of the Raudhatain fresh groundwater field. The monitoring wells P17, P25UA, and P34R are located at contaminated sites, and TOC and TPH analysis of water samples from these wells indicated presence of hydrocarbon contaminants. However, the lack of phenol in the groundwater samples from monitoring wells P17, P25UA, and P34R may be attributed to lack of phenol in the source of contamination and/or its biodegradation at these monitoring well locations.

Overall comparison indicated that phenol was dominantly composed of polar material (73%) as compared with nonpolar material (27%) (Table 5), and it was entirely composed of polar material in the groundwater samples from monitoring wells P12R, P18UA, P19UA, and P36R.

3.5. Polar and nonpolar materials as tannin and lignin

Tannin and lignin are naturally occurring chemical compounds, most frequently found where large quantities of vegetation have decayed. Water that has tannin and lignin appears to be faint yellow to brown in color and may have a slightly bitter taste. The USEPA has no guidelines for expected levels in drinking water, because there is no risk associated with these compounds other than possibly that of a slight stimulant; however, some migraine sufferers believe that tannin can trigger a migraine (Belkraft.com, 2005).

The concentrations of tannin and lignin (polar and nonpolar materials) and polar and nonpolar materials as tannin and lignin concentrations in the groundwater samples are presented in Table 4 and plotted in Fig. 6. Tannin and lignin were not detected in the groundwater samples from the wells P1 and P25UA.

The concentrations of tannin and lignin (polar and nonpolar) in the groundwater ranged between 0.10 and



Fig. 5. Polar and nonpolar material as phenol.

3.20 mg/L, with a mean value 1.72 mg/L. The comparison of polar and nonpolar materials as tannin and lignin indicates that it is dominantly composed of nonpolar material (60%) as compared with nonpolar material (40%; Table 5).

3.6. Polar and nonpolar materials as volatile acids

The method employed to measure the concentrations of volatile acids in the groundwater samples is based on the esterification of carboxylic acids present and the determination of the esters by ferric hydroxamate reaction. Carboxylic acids are organic acids characterized by the presence of a carboxyl group, -C(=O)OH, usually written as -COOH (Mc Naught and Wilkinson, 1997). The simplest series of carboxylic acids is the alkanoic acids, R–COOH, where R is hydrogen or an alkyl group $(C_n H_{2n+1})$. They are produced by oxidation of primary alcohols or aldehydes; they may also be produced by the oxidative cleavage of olefins (unsaturated open-chain hydrocarbons) by means of ozonolysis. In particular, any alkyl group on a benzene ring is fully oxidized to a carboxylic acid, regardless of its chain length. Hydrolysis of nitriles, esters, or amins can also produce carboxylic acids. Lower carboxylic acids (1 to 4 carbon) are miscible (i.e., polar) with water; whereas, higher carboxylic acids are less soluble (nonpolar) due to the increasing hydrophobic nature of the alkyl chain (Morrison and Boyd, 1992).

The concentrations of volatile acids (polar and nonpolar combined), polar and nonpolar components of volatile

Table 5

Comparison of polar and nonpolar materials in the groundwater of the study areas

Parameter	Polar material (%) Min, Max, (Avg)	Nonpolar material (%) Min, Max, (Avg)
TOC	0, 83.13, (41.58)	16.87, 100, (58.42)
TPH	33.33, 100, (85.71)	0, 66.67, (14.29)
UV-induced	71.52, 100, (84.17)	0, 28.48, (15.83)
Phenol	20, 100, (72.87)	0, 68.75, (27.13)
Tannin and lignin	7.14, 66.67, (40.20)	0, 92.86, (59.80)
Volatile acids	7.41, 100, (89.96)	0, 92.59, (10.72)



Fig. 6. Polar and nonpolar material as tannin and lignin.



Fig. 7. Polar and nonpolar material as volatile acids.

acids observed in the groundwater samples are presented in Table 4 and plotted in Fig. 7. The concentrations of volatile acids (polar and nonpolar combined) ranged between 1.00 and 27.00 mg/L, with a mean value of 12.00 mg/L. Volatile acids were not detected in the groundwater samples from the wells P1 and P25UA.

A comparison of polar and nonpolar materials as volatile acids indicated that the material in the groundwater samples was dominantly composed of polar material (89%) as compared with nonpolar material (11%; Table 5), and was entirely composed of polar material in groundwater samples from monitoring wells P17, P18, P19, P33UA, P34R, and P36R.

4. Conclusion and recommendations

The results of groundwater quality measurements, taken on site, indicated the presence of slightly acidic, fresh to brackish groundwater in reduced environments at organicand hydrocarbon-contaminated groundwater sites in the Raudhatain and Umm Al-Aish fresh groundwater fields. The acidic character of the groundwater was ascribed to the presence of organic acids and H₂S.

The TPH, UV-induced fluorescence, phenol, and volatile acids results indicated that the contaminants in the groundwater of the Raudhatain and Umm Al-Aish fresh groundwater fields were dominantly composed of polar material (86%, 84%, 73%, and 90%, respectively), whereas, the TOC, and tannin and lignin results indicated that they were dominantly composed of nonpolar material (58% and 60%, respectively). The higher concentration of polar material in the groundwater may be attributed to weathering of crude oil and its combustion products. When crude oil is exposed to atmospheric conditions, a number of natural processes such as evaporation, emulsification, dispersion, sedimentation, dissolution, photo-oxidation, and biodegradation take place simultaneously. The most significant weathering process for oil spill is evaporation, due to which, more volatile fraction of oil was lost within the few hours. Oxidation of crude oil by the action of ultraviolet radiation in sunlight (photo-oxidation) and by microbes which utilize them as food source (bio-oxidation or bio-degradation) produces oxidation products which are generally more soluble (polar). The results suggest that treatment techniques specific for polar and nonpolar contents will be required for the treatment of petroleum hydrocarbon-contaminated groundwater.

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