



The origin of hydrogen sulphide found in abnormally saline groundwaters in Kuwait City, Kuwait

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Abstract

The paper presents a qualitative assessment water geochemistry study to understand the deep non-anthropogenic origin of dissolved H₂S in anomalously abnormal saline groundwaters in Kuwait City. The groundwater samples show geographical variation in water chemistry, salinity, CO₂ and H₂S concentrations and δ³⁴S isotope data. The variation was found to be controlled by the proximity to the abandoned Medina Field. Three geographical areas in Kuwait City with different groundwater properties were identified: Area-A with very high H₂S concentration (> 100 mg/l); Area-B with moderate H₂S concentration (50–100 mg/l) and Area-C with low H₂S concentration (< 50 mg/l). The δ³⁴S isotope data of dissolved H₂S in the groundwater samples showed a range of –8.9 to –12.9‰. The variation of δ³⁴S values indicate the origin of H₂S sourced from mixing of low-temperature sulphate reduction (BSR) with high-temperature thermochemical sulphate reduction (TSR) fluids migrated up-fault from deeper and hotter oil reservoirs through reactivated NE-SW deep-seated faults during recent tectonic events.

Keywords Kuwait city groundwater · H₂S · Sulphur isotopes · Groundwater salinity · Kuwait · Upfault water migration

Introduction

In Kuwait, it has been proved that deep subsurface geological fluids (hydrocarbon and/or non-hydrocarbon gases associated with formation waters/ oilfield brines) migrate up-fault during tectonic events (Al-Hajeri 2018; Al-Hajeri et al. 2020; Al-Hajeri and Bowden 2017; Al-Hajeri and Bowden 2018; Prakash et al. 2019). When deeper and hotter fluids reach shallower sections, diagenetic reactions are promoted and enhanced through water–rock interactions. From the rock phase perspective, Gunatilaka (1989) found that the occurrence of primary spheroidal dolomite crystals in fluid-filled pore spaces of rock samples from the Kuwait Formation (Fig. 1) occurs close to groundwater aquifers with a low dolomitising potential in southern Bahrah Field (Fig. 2). He found that hydrocarbon species dissolved in groundwater had oxidised

to CO₂ close to the surface, after which gas bubbles had formed nuclei for spheroidal dolomite growth. El-Sayed et al. (1991) studied δ¹³C data of shallow dolomite in Kuwait and suggest that samples close to oil fields show relatively lighter δ¹³C values (~–10‰ PDB), resulting from the oxidation of seeping hydrocarbons. Khalaf and Abdullah (2013) evaluated the development of diagenetic alunites with dolomitised duricrust (dolocretes) in Dammam Formation samples obtained from a quarry close to the Ahmadi residential area (Fig. 1). They found that the occurrence of these diagenetic dolocretes was probably controlled by hydrocarbon migration. However, from a water phase perspective, formation waters of Cretaceous and Tertiary reservoirs within the Kuwait petroleum system are anomalously abnormal in salinity at encountered depths (Al-Hajeri and Bowden 2017). In addition, Al-Hajeri et al. (2017) evaluated the paleothermal history of the Tayarat Formation reservoirs in Burgan Field by conducting two geochemical studies: fluid inclusion microthermometry of calcite-filled veins and water geochemical thermometry. They found that homogenisation temperatures of fluid inclusions and water thermometry suggest that paleofluids were trapped at temperatures about 200 °C, four times higher than present-day reservoir temperatures of 50 °C. In Kuwait City, one avenue is famous for its unpleasant smell of H₂S gas. Therefore, Mukhopadhyay et al. (2007) found that

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Age	Formation	Age	Formation	Age	Formation
Jurassic	Hith	Cretaceous	Tayarat	Tertiary	Dibdibba
	Gotnia		Mishrif		Lower Fars
	Najmah		Ahmadi		Ghar
	Sargelu		Wara		Dammam
	Dhrumah		Mauddud		Rus
Triassic	Minjur		Burgan	Radhuma	Kuwait*
	Jilh		Shuaiba	Evaporites	
	Sudair		Zubair	Carbonates	
Per. Permian	Khuff		Ratawi	Siliciclastics	
	Unayzah		Minagish	Shale	
		Makhul			

Fig. 1 Simplified stratigraphic column of Kuwait. Kuwait Group has been renamed Kuwait Formation by Amer and Al-Hajeri (2019)

the increase of water salinities and dissolved H_2S gas concentrations in the groundwater of Kuwait City probably originated from non-anthropogenic (non-sewage) source.

The reaction between dissolved SO_4 and hydrocarbons is thermodynamically unstable in all diagenetic environments (Machel 2001). There are two mechanisms generating H_2S : BSR and TSR. The BSR develop in low-temperature (< 80 °C) thermal regime, whereas TSR operates in high-temperature (127–180 °C) thermal regime (Machel 2001). The BSR fluids show lighter $\delta^{34}S$ ratio because of depletion in ^{34}S isotope fractionation, while TSR fluids tend to display heavier $\delta^{34}S$ ratio as a result of ^{34}S enrichment lowers isotope fractionation (Brunner and Bernasconi 2005; Machel 2001; Manzano et al. 1997). SO_4 and sulphide products of TSR process show low $\delta^{34}S$ fractionations because aqueous pools of SO_4 are typically limited, and thus, fractionation is limited (Manzano et al. 1997). Finally, the objective of this study is to use the sulphur isotope data to identify the source of dissolved H_2S in the groundwater of Kuwait City.

Geological setting

The Phanerozoic sedimentary section of Kuwait in the depocenter of the Arabian platform basin is around 35,000 ft. in thickness (Alsharhan and Nairn 1997; Kamen-Kaye 1970). The Kuwait stratigraphic column is poorly documented albeit most information obtained from deep drilling wells penetrating the Unayzah Formation of Permo-Carboniferous age (Fig. 1). The Khuff Formation represents a carbonate/evaporite system of Upper Permian age that was deposited in a shallow water environment. The Sudair, Jilh and Minjur formations of the Triassic period are characterised by a mixed sequence of carbonates, evaporites and continental siliciclastics, respectively. However, the Jurassic age sediments are characterised by the deposition of shallow marine water carbonate/evaporite system of the Marrat, Sargelu, Najmah, Gotnia and Hith formations. In Kuwait, the Gotnia and Hith Formation evaporites represent a major regional seal

between the Jurassic and Cretaceous sections (Al-Hajeri et al. 2006; Ali 1995). The entire Cretaceous period primarily consists of carbonates interrupted by two marine regression events that deposited the thick Zubair and Burgan formation siliciclastic sequences during the Middle Cretaceous. The Tertiary section unconformably sits on the Tayarat Formation of Upper Cretaceous age. The early Tertiary deposits of the Radhuma, Rus and Dammam formations are carbonate and evaporite systems separated by an unconformity from late Tertiary and Quaternary siliciclastic deposits of Kuwait Formation (Amer and Al-Hajeri 2019).

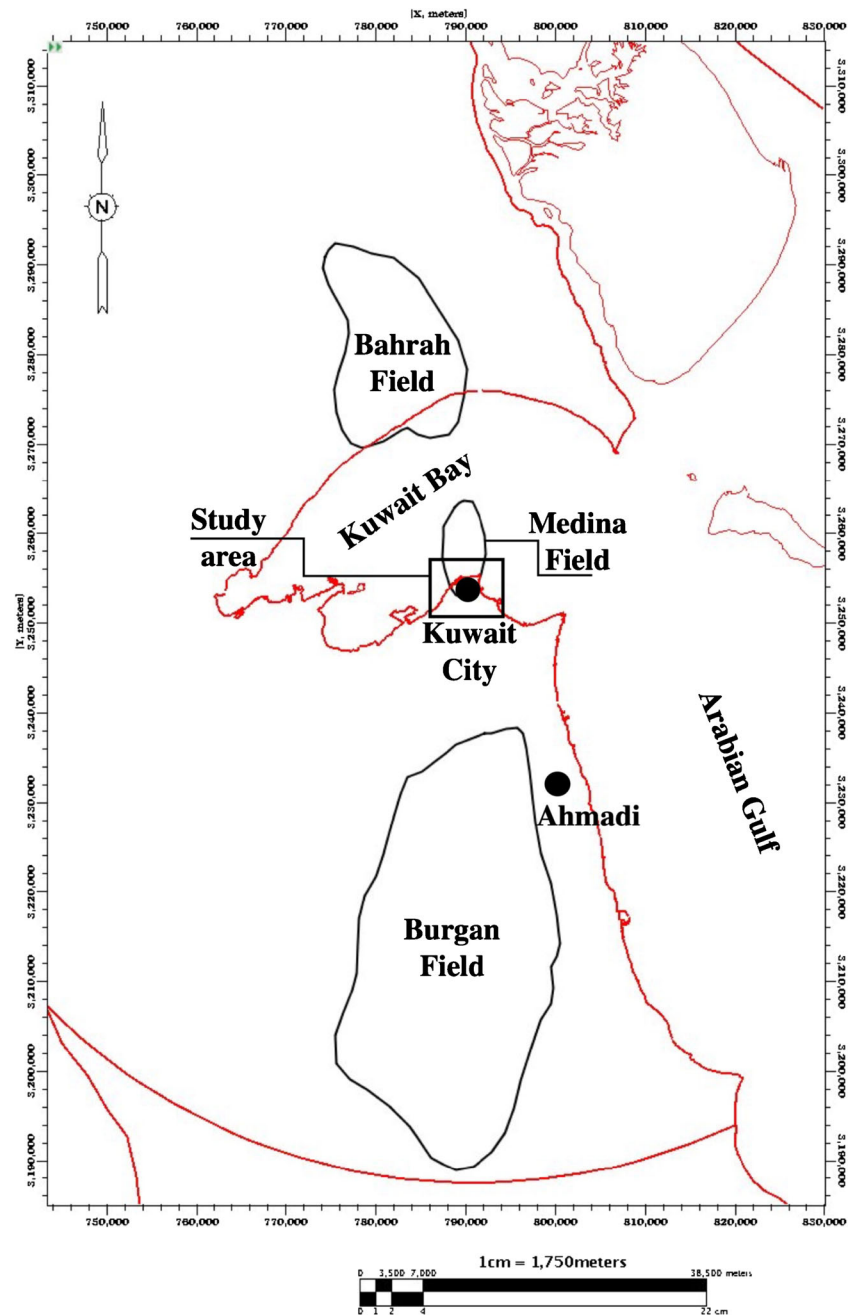
Structural geology

The northern Arabian Gulf state of Kuwait is located in a passive margin bounded by the Arabian Shield craton to the West and the Zagros subduction zone to the East. The regional Hercynian Orogeny that initiated during the mid-Carboniferous and continued until the late Permian formed the widely distributed NE-SW trending structural lineaments (Faqira et al. 2009). The Hercynian structures in Kuwait were later followed by a Triassic regional extension forming a series of horsts and grabens that cut across the Hercynian mega-structures (Faqira et al. 2009), forming a complex fault network. These events were followed by a regional phase of major deformation during Jurassic rifting (Filbrandt et al. 2006; Sharland et al. 2001). The Alpine Orogeny initiated during the Cretaceous and continued to the mid-to-late Tertiary (Corley et al. 2006; Richard et al. 2014). The Alpine-1 phase that occurred during the Cretaceous was characterised by transtensional tectonics creating the majority of the NW-SE trending structures. The Alpine-2 phase occurred during the mid-to-late Tertiary and was characterised by tectonic inversion and reactivation of the pre-existing NE-SW trending structures (Filbrandt et al. 2006; Richard et al. 2014). Bou-Rabee (2000) thinks that recent seismic activities or earthquakes in Kuwait are attributed to the Alpine-2 event. The NE-SW fault system was most likely reactivated during the late Holocene following the final stages of Kuwait's uplift and emergence and is reflected in the natural fracture orientations observed over the Bahrah Field area (Fig. 3) (Al-Hajeri et al. 2020; Amer and Al-Hajeri 2020; Amer et al. 2018).

Hydrogeological setting

In Kuwait, the Tertiary deposits of the Dibdibba, L. Fars/Ghar and Dammam formations are considered hydrodynamic layers, whereas the deeper hydrogeologic layers are identified as hydrostatic layers, i.e. no groundwater movement (Mukhopadhyay et al. 1996; Omar et al. 1981; SNEA 1983). The Rus Formation (Early Eocene in age) anhydrites are identified as a regional seal/hydrodynamic barrier between a shallower active hydrodynamic system and a deeper

Fig. 2 A location map of the study area in Kuwait City



hydrostatic system. Kuwait is part of the Saudi Arabia hydrodynamic system sourced from distal outcrops in the southwest of Saudi Arabia with a very weak hydrodynamic gradient groundwater movement direction towards the northeast (Al Mahmoud 2017; SNEA 1983). Therefore, Tertiary hydrodynamic layers in southwest Kuwait of SO_4 -rich less-saline waters change to Cl -rich higher-saline brines towards the northeast because of hydrodynamic stagnant conditions and the development of a static saline water body (Mukhopadhyay et al. 1996; Omar et al. 1981). However, it has been suggested that the Cl -rich waters are anomalously abnormal with regard to salinity for its depth, indicating this probably migrated

upward from Rus Formation anhydrites and/or the Gotnia Formation (Upper Jurassic in age) halite (Al-Hajeri and Bowden 2017; SNEA 1983).

Methods and dataset

In Kuwait City, 10 groundwater samples with abnormally high salinity containing total dissolved solids (TDS) > 10,000 mg/L were collected. These samples were stratigraphically collected from the base Kuwait Group at a depth > 80 m, which is recently known as Kuwait Formation

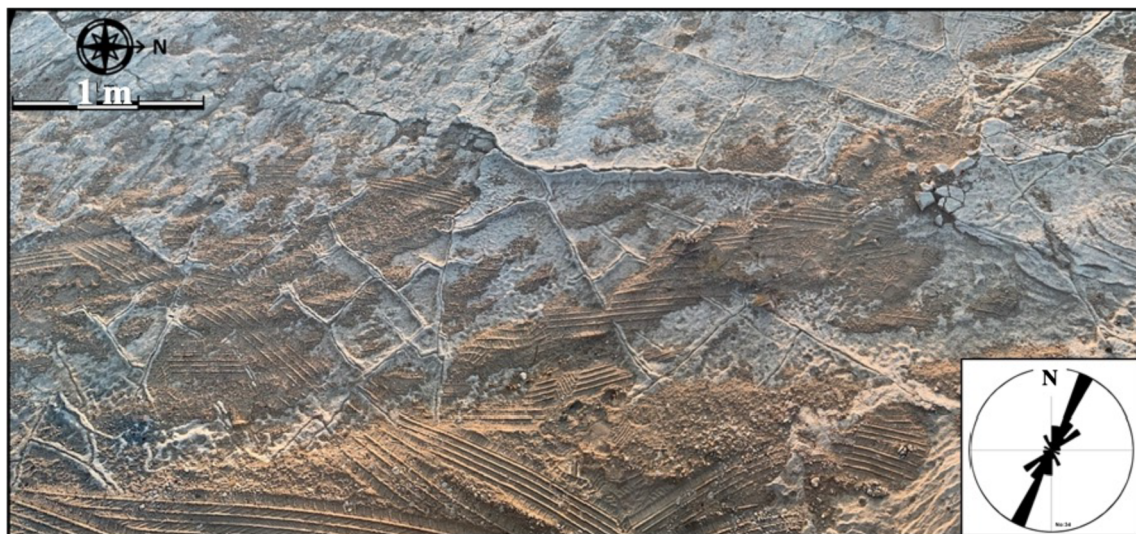


Fig. 3 Natural joints and fracture network observed in outcrops of southern Bahrah Field with the rose diagram of 34 measurements showing fractures with the dominance of N30° trends

(Amer and Al-Hajeri 2019). The water samples were analysed for major ions (Na, Ca, Mg, K, Cl, SO₄ and HCO₃), TDS, pH, dissolved CO₂ gas and H₂S gas. In addition, sulphur isotope data were measured for the dissolved H₂S in water. The standard methods adopted for the determination of major ions, carbonate and bicarbonate (Alkalinity) samples were as follows: anions-ASTM 4327, cations-ASTM 6919 and alkalinity-SMEWW 2320. The standard methods for measuring dissolved CO₂ and TDS are ASTM D513 and ASTM D5907, respectively. However, standard method used to determine H₂S was SMEWW 4500S2-F, whereas IRMS standard method was used after converting sulphate into sulphide to measure sulphur isotopes ($\delta^{34}\text{S}$). The $\delta^{34}\text{S}$ of dissolved sulphate is determined by the barium sulphate precipitation technique. Prior to preparation, the sulphate concentration must be determined using a Thermo AQUAFast II™ colorimeter; a minimum sulphate concentration of 5 ppm is required for the analysis. Once the sulphate concentration is known, the amount of sample needed to obtain 25mg of barium sulphate is weighed into a clean beaker. Samples are then filtered to remove particulate matter before being brought to a boiling temperature. Calcium carbonate in the sample is removed by adding 1N hydrochloric acid until the sample reaches a pH of 1–3. An appropriate amount of barium chloride solution is added based on the concentration of sulphate in the sample. The reaction is given sufficient time to complete under heat; a testing solution is used to ensure that all dissolved sulphate has precipitated. The precipitant is then filtered and dried, and the final amount is weighed. The $\delta^{34}\text{S}$ of the dried barium sulphate is then determined by combustion to SO₂ gas using an Elementar Vario Cube EA. Approximately 1.2 mg BaSO₄ is weighed into a tin capsule and loaded into an autosampler. The sample is then combusted and passed over a reduction furnace. The resulting SO₂ gas is passed through a

ConFlo II interfaced to the IRMS, where it is compared against a reference gas.

The sulphur isotope ratio was reported as $\delta^{34}\text{S}$ value in per mil (or ‰) notation. $\delta^{34}\text{S}$ is defined as follows:

$$\delta^{34}\text{S} = \left(\frac{{}^{34}\text{S}}{{}^{32}\text{S}} \right)_{\text{Sample}} - \left(\frac{{}^{34}\text{S}}{{}^{32}\text{S}} \right)_{\text{Standard}} / \left(\frac{{}^{34}\text{S}}{{}^{32}\text{S}} \right)_{\text{Standard}} \times 1000$$

Results

Table 1 reports chemical, physical and isotopic data. The groundwaters of Kuwait City show variation in chemistry, salinities, gas concentrations and sulphur isotope values. The variation is geographically controlled related to the proximity to Medina oil Field. The field was abandoned after drilling two discovery wells that proved oil accumulations in Cretaceous reservoirs in the year 1963. Therefore, three areas with different readings were identified (Figs. 4 and 5): Area-A with very high H₂S and CO₂ concentrations and water salinities, lower pH and heavier $\delta^{34}\text{S}$ data (wells KC-WW-01, KC-WW-02, KC-WW-04, KC-WW-08, KC-WW-16 and KC-WW-17); Area-B with moderate readings (wells KC-WW-09 and KC-WW-14) and Area-C with lower H₂S and CO₂ concentrations and water salinities, higher pH and lighter $\delta^{34}\text{S}$ data (wells KC-WW-07 and KC-WW-10)

Discussion

We propose that variation in $\delta^{34}\text{S}$ ratio values (–12.9 to –8.9‰) of the dissolved H₂S associated with anomalously abnormal salinity groundwaters of Kuwait City indicative of

Table 1 The physicochemical properties and sulphur isotopes of Lower Kuwait Formation groundwater samples in Kuwait City

Well name	Formation Area	Na (mg/L)	Ca (mg/L)	Mg (mg/L)	Cl (mg/L)	SO4 (mg/L)	HCO3 (mg/L)	pH @ 25 °C	Density (g/cc)	TDS (mg/L)	Dissolved CO2 (mg/L)	Dissolved H2S (mg/L)	$\delta^{34}\text{S}$ of Dissolved H2S (CDT) ‰
Kuwait Bay Seawater		12,061	532	1610	22,771	3100	175	8.3	1.03	41,852			+20.9*
KC-WW-1	Kuwait	17,326	2825	1390	43,914	2387	649	6.5	1.04	75,547	240	168	
KC-WW-2		29,226	6152	2212	56,838	1822	1072	6.0	1.07	13,9757	456	184	-8.9
KC-WW-4		33,892	4586	1596	58,300	1787	1051	5.9	1.05	98,915	253	234	-7.8
KC-WW-8		19,820	4218	1426	36,659	1885	1702	6.4	1.05	90,189	362	197	-11.8
KC-WW-16		26,756	4074	1379	51,368	1986	1045	6.3	1.05	85,380	297	219	-11.6
KC-WW-17		14,228	237	1075	32,319	2305	640	6.5	1.02	62,427	178	213	-11.4
KC-WW-9	B	4142	951	375	5754	3232	1010	7.0	1.01	17,227	182	94	-11.8
KC-WW-14		3737	1104	421	6526	1918	1010	8.0	1.01	19,693	282	58	-11.7
KC-WW-7	C	2521	877	340	4372	1711	1006	7.5	1.01	13,619	106	32	-13.2
KC-WW-10		3044	885	357	5442	2117	1008	7.7	1.01	16,073	98	47	-12.9
Bahrah Field	Mauddud												+21
	Zubair												+14.3
	Ratawi												+22.1

*Robinson and Gunatilaka (1991)

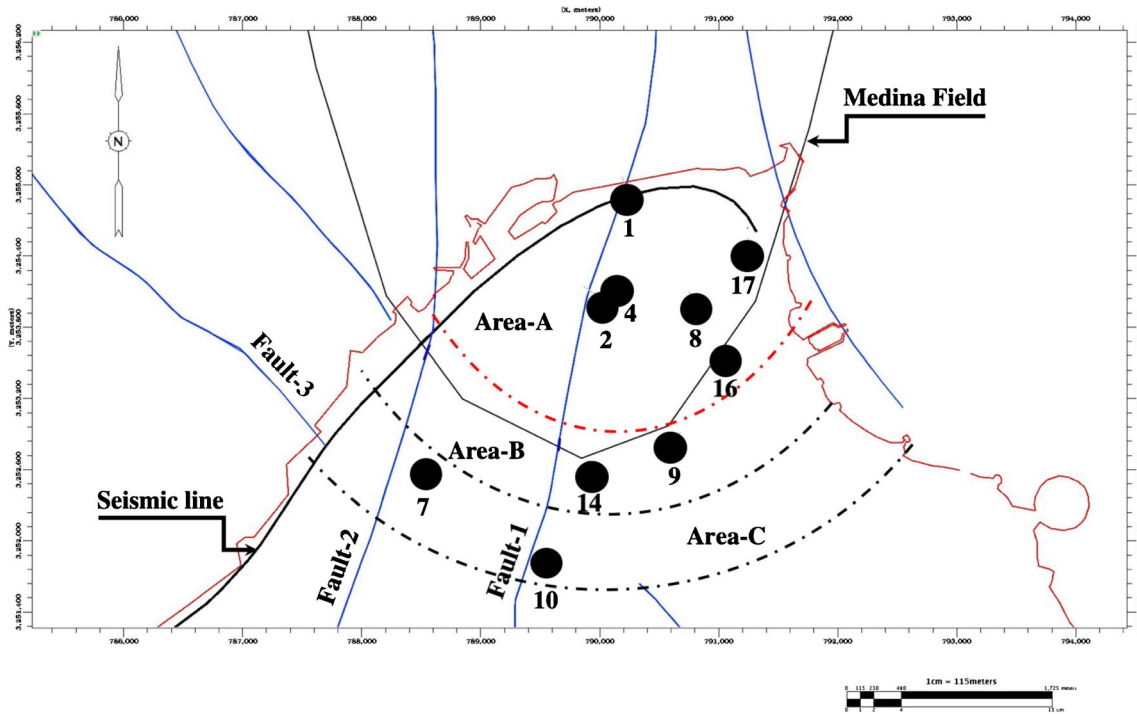


Fig. 4 The histogram plot showing the average values of groundwater parameters (Cl, pH, CO₂, H₂S and δ³⁴S) of Area-A, Area-B and Area-C in Kuwait City tabulated in Table 1

a BSR source with possible mixing with a thermogenic source (Al-Hajeri 2018; Colling 2005). The δ³⁴S data values of groundwater samples in Kuwait City are distinguishably different than Cretaceous values (Table 1). The formation waters of Cretaceous reservoirs in Bahrah Field show dissolved H₂S with heavy δ³⁴S values range from +14 to +20‰ suggesting TSR source probably from reservoirs Palaeozoic in age (Al-Hajeri 2018; Al-Hajeri and Bowden 2018; Claypool et al. 1980). Therefore, we also propose that an upward and up-fault migration mechanisms of deep and hot overpressured (Hunt 1990) probably responsible for migrating TSR fluids to reach near-surface deposits.

Colling (2005) evaluated the δ³⁴S data of H₂S gas for the Cretaceous oil reservoirs in Burgan Field and found that variation in δ³⁴S values attributed to the mixing of BSR and TSR processes (Brunner and Bernasconi 2005; Machel 2001; Manzano et al. 1997). Therefore, to characterise the source of H₂S in Burgan Field based on δ³⁴S values, Colling

identified three classes: (1) +5‰ to +14‰ indicative of a TSR source; (2) 0‰ to +5‰ indicative of mixing between TSR and a BSR source; (3) -10 to 0‰ indicative of BSR source mixing with TSR source. The present-day temperature of Cretaceous oil reservoirs in Kuwait is < 100 °C; therefore, it is unlikely that TSR operates in situ. Therefore, more likely that TSR fluids have migrated from deeper and hotter formations representing an overprint of previous isotopic signature with thermochemically generated reduced sulphide (Al-Hajeri 2018; Al-Hajeri and Bowden 2018). Moreover, a native sulphur field sample was obtained from a naturally occurring H₂S seep in Burgan Field (Fig. 2) for petrographic and δ³⁴S analyses (Al-Hajeri and Bowden 2018). The scanning electron microscope images of the native sulphur field sample show the occurrence of bladed sulphur species and globular sulphate precipitates. The sulphur forms are sublimated in the pore system, and the globular sulphate precipitate tends to form a grain coating rather than occurring as bladed sulphur.

Fig. 5 The geographical distribution of the three study areas with different groundwater properties and its proximity to Medina Field: Area-A, -B and -C (Table 1). The location map also shows the seismic line used in the study

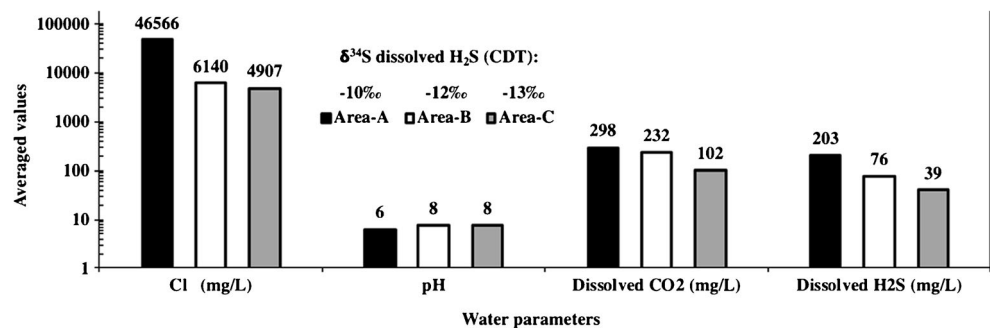
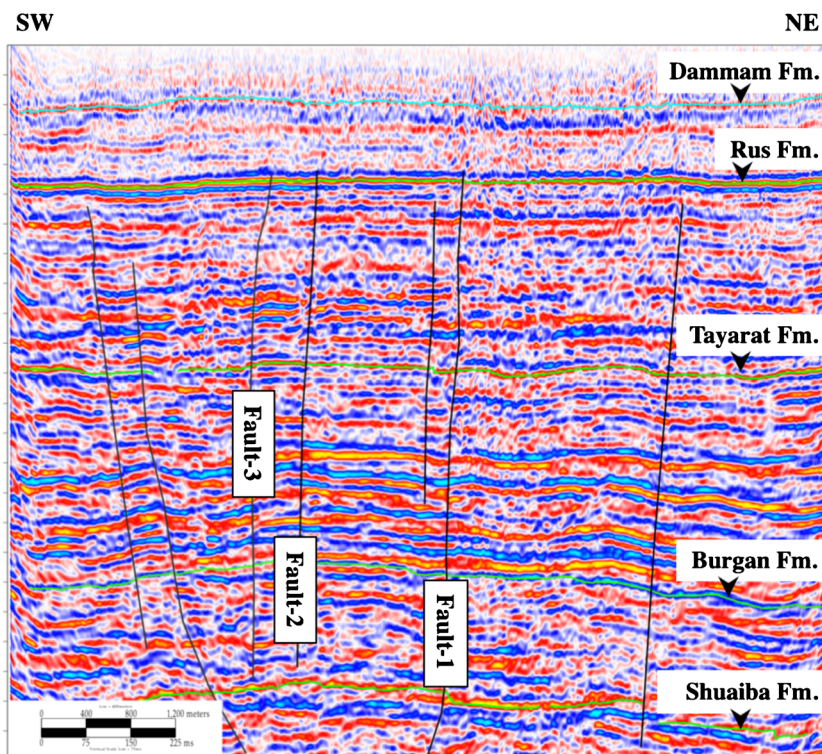


Fig. 6 The three interpreted faults reached top Rus Formation: Fault-1, -2 and -3 (see Fig. 5).



It indicates that sulphate globules developed first, followed by bladed sulphur filling the remnant pore space. The $\delta^{34}\text{S}$ value of the native sulphur sample is +12.9‰, which represents a relatively low degree of fractionation for a reduced sulphur phase. The present-day Arabian Gulf seawater $\delta^{34}\text{S}$ of +20.9‰ (Robinson and Gunatilaka 1991) imply that the source of sulphur species in the H_2S seep of Burgan Field is not related to a contemporary deposit. However, based on Claypool et al. (1980), $\delta^{34}\text{S}$ universal curve for ancient seawater SO_4 corresponds to the Khuff Formation (Permian in age, Fig. 1).

In Kuwait, deep-seated fault reactivation facilitated up-fault migration of geological fluids during Upper Cretaceous and Neogene tectonics (Al-Hajeri and Bowden 2017; Al-Hajeri and Bowden 2018; Al-Hajeri et al. 2017; Corley et al. 2006). In Kuwait City, seismic data display the occurrence of faults at the top of the Rus Formation with two trends of different orientations NE-SW (N30°) and NW-SE (Fig. 6). The same NE-SW fault system was reactivated during the final uplift and emergence stage of Kuwait during the late Holocene (Amer and Al-Hajeri 2020; Amer et al. 2018). The deep subsurface NE-SW in orientation normal faulting has been responsible for recent earthquake activities recorded in north Kuwait (Al-Jeri et al. 2020). Interestingly, field trip observations show the occurrence of joints and a fracture network oriented towards NE-SW in the vicinity of southern Bahrah Field (Fig. 3). Recent field studies documented the

occurrence of unusual surface sandstone pipes along weak zones of fractures network with NE-SW trend in southern Bahrah Field (Al-Hajeri et al. 2020). Thus, it has been suggested that N30° fault system acted as a possible present-day conduit for subsurface geological fluid flow (Fig. 7).

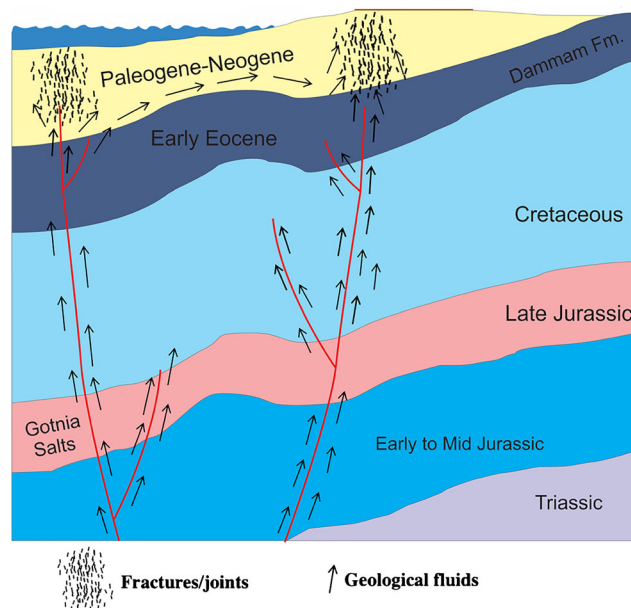


Fig. 7 A conceptual model of the up-fault/upward migration of subsurface geological fluids in Kuwait City

Conclusion

The occurrence of non-anthropogenic dissolved H₂S gas in anomalously abnormal saline groundwaters in Kuwait City suggests these fluids were probably sourced from a deeper origin. The $\delta^{34}\text{S}$ data indicates that H₂S gas was originated from a biogenic source with possible mixing with a thermogenic source. The chemical, physical, salinity and isotopic properties of the groundwater dataset vary geographically due to proximity to the abandoned Medina oil Field. Water wells close to Medina Field tend to show higher groundwater properties, whereas wells more distal from the field show lower groundwater properties. It has been suggested that faults are possible conduits for subsurface fluid flow. Recent tectonic events reactivated deep-seated fault facilitating up-fault migration of deeper fluids. It appears that the NE-SW fault system is the responsible conduit for present-day subsurface fluid flow in Kuwait. Thus, we recommend to pay more attention to NE-SW fault system to better understand surface- and near-surface manifestations of subsurface geological activities in Kuwait.

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Declarations

Conflict of interest The authors declare that they have no competing interests.

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