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The hydrogeochemistry of shallow groundwater from Lut Desert, Iran: The hottest place on Earth

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ABSTRACT

This paper presents the first shallow groundwater geochemical data from the Lut Desert (Dasht-e-Lut), one of the hottest places on the planet. The waters are Na–Cl brines that have undergone extensive evaporation, but they are unlike seawater derived brines in that the K⁺ is low and the $Ca^{2+} > Mg^{2+}$ and $HCO_3^- > SO_4^{2-}$. In addition to evapo-concentration, the most saline samples indicate that the dissolution of previously deposited salt also acts as a major control on the geochemistry of these waters. High concentrations of gypsum in surface soils along with the water geochemistry indicate the dissolution, precipitation, and reprecipitation of evaporite salts are very important to the overall chemistry of the near surface environment. As demonstrated by the high H₄SiO₄ concentrations, the weathering of aluminosilicate minerals also contributes to the solutes present. Fixed nitrogen (NO₃⁻ + NO₂⁻) concentrations suggest little to no denitrification is occurring in these waters. The processes controlling the geochemistry of the Lut waters are similar to those in other hyperarid regions of the Earth.

1. Introduction

The Lut Desert (Dasht-e-Lut) in southeast Iran is one of the largest deserts in Iran, receiving < 30 mm of precipitation annually. Its bioclimate is classified has 'tropical hyperdesertic,' the hottest and driest bioclimate in the world in the Global Bioclimatic Classification (Djamali et al., 2011). The region has been referred to as the thermal pole of the Earth and is recognized as one of the hottest, if not the hottest, locations on planet Earth with surface temperatures as high as 70.7 °C recorded via satellite in the mid 2000's (Mildrexler et al., 2006). A recent measurement during the sampling campaign in Yalan dunes (March 4, 2017) returned skin temperature as high as 78.2 °C (Trescher, 2017). The mean annual precipitation (7-year average, 2003-2010) recorded at the nearest meteorological station, Shahdad synoptic weather station, located at the west flank of the Lut desert with the altitude of 400 m.a.s.l, does not exceed 30 mm. Precipitation is highest from January through March and the dry season lasts from April to November. Mean annual temperature at the station is 27.5 °C and the mean minimum and maximum daily air temperatures of the warmest and coldest months of the year are -2.6 °C and 50.4 °C respectively (Fig. S1). Remote sensing analysis from 2002 to 2003 report average daily summer temperatures as high as 45.3–55.7 C, with surface soil temperatures as high as 66.5 to 71.1 (Azizi et al., 2007). The Lut Desert has a total area of ~51,800 km², and consists of sand dunes, ergs, mega-yardangs (kaluts), desert pavement, as well as large flat areas of salty soils and playas (Alavipanah et al., 2007; Ehsani, 2011). A semi-permanent river (Rud-e Shur) flows from NW of the area and passes the edge of Kaluts and ends in the central depression (Chaleh Markazi).

The Lut Desert is within the Lut Block, whose basement consists of pre-Jurassic metamorphic rocks (pelitic schist) as well as Jurassic sediments intruded by younger (Jurassic to Tertiary) granitic plutons and covered by Tertiary mafic to felsic volcanics (Arjmandzadeh and Santos, 2014; Tirrul et al., 1983; Yazdi et al., 2014). The region is active tectonically with many active faults (Boshrabadi et al., 2018).

A large portion of the region is too harsh for any plants, and has been referred to as aphytic (Mobayen, 1976) and devoid of life. However, recent work has described some of the extremophile microbial life existing in the soils and their abilities to withstand high temperatures,

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Sample locations and descriptions.

| Water Sampl | les | |
|--|---|---|
| Sample ID | Location | Description |
| St 10 | Lat 29° 34.831′ N Long 58° 58.428′E | Sample obtained by digging into the sand dune at the depth of 80 cm below the land surface (Ab-e Yalan). |
| St 19 | Lat 29° 48.780′ NLong 59° 11.675′E | Location known as "Logger Point". No Sign of surface water but dug into a brownish soil and reached water @ 90 cm depth. Image in Fig. S2. |
| St 21 | Lat 30° 04.394′ NLong 58° 55.298′E | Water off surface, flows for ~200 m and recharges (Sarcheshmeh-e Shoor Gaz) |
| St 22 | Lat 30° 04.144′ NLong 58° 56.153′E | Further downstream from #21, a very flat playa surrounded by scarpments. Known as Shoor Gaz Hamoun where surface discharge was sampled. |
| St 28 | Lat 30° 24.025′ N Long 58° 59.519′E | Low lying playa, Chale Malek-Mohammad sampled water from swamp area. Image in Fig. S2. |
| Soil Sample | s | |
| Sample ID | Location | Description |
| Sample ID | Location Lat 30° 58.20′ N Long 57 31.68′ E | Description Small depression, 70 km N of Shahdad with sparse Hammada salicornica shrubs |
| • | Lat 30° 58.20' N Long 57 31.68' E Lat 31° 0.05' N Long 57° 40.80' E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian |
| 2 5 8 | Lat 30° 58.20' N Long 57 31.68' E Lat 31° 0.05' N Long 57° 40.80' E Lat 30° 55.35' N Long 58° 22.33' E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian Mixture of Kalut soil profile and brown soil with some salt crystals near the Kal-e Lut |
| 2 5 8 9 | Lat 30° 58.20' N Long 57 31.68' E Lat 31° 0.05' N Long 57° 40.80' E Lat 30° 55.35' N Long 58° 22.33' E Lat 30° 53.90' N Long 58° 23.88' E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian Mixture of Kalut soil profile and brown soil with some salt crystals near the Kal-e Lut Brown soil from Lut Playa |
| 2 5 8 9 13 | Lat 30° 58.20′ N Long 57 31.68′ E Lat 31° 0.05′ N Long 57° 40.80′ E Lat 30° 55.35′ N Long 58° 22.33′ E Lat 30° 53.90′ N Long 58° 23.88′ E Lat 30° 34.82′ N Long 58° 35.62′ E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian Mixture of Kalut soil profile and brown soil with some salt crystals near the Kal-e Lut Brown soil from Lut Playa Sand from Kalute Abul hol |
| 2 5 8 9 13 18 | Lat 30° 58.20′ N Long 57 31.68′ E Lat 31° 0.05′ N Long 57° 40.80′ E Lat 30° 55.35′ N Long 58° 22.33′ E Lat 30° 53.90′ N Long 58° 23.88′ E Lat 30° 34.82′ N Long 58° 35.62′ E Lat 30° 6.63′ N Long 58° 43.57′ E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian Mixture of Kalut soil profile and brown soil with some salt crystals near the Kal-e Lut Brown soil from Lut Playa Sand from Kalute Abul hol Dagh (playa) of the eastern corner of Kalutes, scattered <i>Tamarix pycnocarpa</i> and <i>Suaeda fruticosa</i> |
| 2 5 8 9 13 18 20 | Lat 30° 58.20′ N Long 57 31.68′ E Lat 31° 0.05′ N Long 57° 40.80′ E Lat 30° 55.35′ N Long 58° 22.33′ E Lat 30° 53.90′ N Long 58° 23.88′ E Lat 30° 34.82′ N Long 58° 35.62′ E Lat 30° 6.63′ N Long 58° 43.57′ E Lat 30° 2.53′ N Long 58° 49.68′ E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian Mixture of Kalut soil profile and brown soil with some salt crystals near the Kal-e Lut Brown soil from Lut Playa Sand from Kalute Abul hol Dagh (playa) of the eastern corner of Kalutes, scattered <i>Tamarix pycnocarpa</i> and <i>Suaeda fruticosa</i> Sample from Shoorgaze Hamoun's puffy salt flat |
| 2 5 8 9 13 18 20 25 | Lat 30° 58.20′ N Long 57 31.68′ E Lat 31° 0.05′ N Long 57° 40.80′ E Lat 30° 55.35′ N Long 58° 22.33′ E Lat 30° 53.90′ N Long 58° 23.88′ E Lat 30° 34.82′ N Long 58° 35.62′ E Lat 30° 6.63′ N Long 58° 43.57′ E Lat 30° 2.53′ N Long 58° 49.68′ E Lat 29° 48.80′ N Long 59° 11.68′ E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian Mixture of Kalut soil profile and brown soil with some salt crystals near the Kal-e Lut Brown soil from Lut Playa Sand from Kalute Abul hol Dagh (playa) of the eastern corner of Kalutes, scattered <i>Tamarix pycnocarpa</i> and <i>Suaeda fruticosa</i> Sample from Shoorgaze Hamoun's puffy salt flat Location known as "2005 Logger Point". Sample contains high moisture. |
| 2 5 8 9 13 18 20 25 28 | Lat 30° 58.20′ N Long 57 31.68′ E Lat 31° 0.05′ N Long 57° 40.80′ E Lat 30° 55.35′ N Long 58° 22.33′ E Lat 30° 53.90′ N Long 58° 23.88′ E Lat 30° 34.82′ N Long 58° 35.62′ E Lat 30° 6.63′ N Long 58° 43.57′ E Lat 30° 2.53′ N Long 58° 49.68′ E Lat 29° 48.80′ N Long 59° 11.68′ E Lat 29° 45.58′ N Long 59° 3.11′ E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian Mixture of Kalut soil profile and brown soil with some salt crystals near the Kal-e Lut Brown soil from Lut Playa Sand from Kalute Abul hol Dagh (playa) of the eastern corner of Kalutes, scattered <i>Tamarix pycnocarpa</i> and <i>Suaeda fruticosa</i> Sample from Shoorgaze Hamoun's puffy salt flat Location known as "2005 Logger Point". Sample contains high moisture. Water runnel between Shoor Gaz and southern Lut, <i>Seidlitzia rosmarinus, Tamarix kermanensis</i> |
| 2 5 8 9 13 18 20 25 | Lat 30° 58.20′ N Long 57 31.68′ E Lat 31° 0.05′ N Long 57° 40.80′ E Lat 30° 55.35′ N Long 58° 22.33′ E Lat 30° 53.90′ N Long 58° 23.88′ E Lat 30° 34.82′ N Long 58° 35.62′ E Lat 30° 6.63′ N Long 58° 43.57′ E Lat 30° 2.53′ N Long 58° 49.68′ E Lat 29° 48.80′ N Long 59° 11.68′ E | Small depression, 70 km N of Shahdad with sparse <i>Hammada salicornica</i> shrubs Near saline river, Rude Shoore Gadom Berian Mixture of Kalut soil profile and brown soil with some salt crystals near the Kal-e Lut Brown soil from Lut Playa Sand from Kalute Abul hol Dagh (playa) of the eastern corner of Kalutes, scattered <i>Tamarix pycnocarpa</i> and <i>Suaeda fruticosa</i> Sample from Shoorgaze Hamoun's puffy salt flat Location known as "2005 Logger Point". Sample contains high moisture. |

Soil samples were taken from the top 10-cm of the soil profile and stored in plastic bags. Air-dried samples were passed through 2 mm sieve and diluted with distilled water following the method described by Bolukbasi et al. (2015). Soil conductivity and pH was measured using a pH/Conductivity meter (ORION STAR A215) by diluting samples with distilled water to 1:2.5 and 1:5, respectively. The percentage of gypsum content in the soil was determined gravimetrically by comparing samples dried at 60 °C and 105 °C (Porta et al., 1986). Total carbon (Ct) was measured by elemental analyzer following the Dumas method (Elementar C/N; VarioMax, Hanau, Germany). All soil measurements were conducted at Instituto Pirenaico de Ecología (IPE-CSIC), Spain.

UV light, and even radiation (Mazkour et al., 2017; Mohseni et al., 2014; Shirsalimian et al., 2017). Although previous studies have described the unusual desert geomorphology (Yazdi et al., 2014), there has been little attempt to understand other surficial and shallow subsurface processes occurring there. In addition, the recent expeditions discovered a shallow subsurface water system in at least a portion of this extremely hot place (Stone, 2016; Trescher, 2017). As a first attempt to better understand the biodiversity and describe the physical environment of the Lut Desert, and as part of a large scientific program (Adaptation and Function of Lut Desert Biodiversity (AFLDB) project), we present the first geochemical data on these waters, speculate on their origin and evolution, and compare them to what is known about other hyper-arid environments in Israel, China, Chile, Australia and the western United States.

2. Methods

The sampling coordinates and depths are given in Table 1, and sampling locations shown in Fig. 1. Surface water or shallow groundwater samples (~1 m depth) were taken by hand in pre-cleaned 120 ml widemouth LDPE bottles. In some cases, overlying sediment/soil was removed by shovel prior to sampling the water. The water samples were filtered through 0.4 µm membrane filters using precleaned polycarbonate filtration system into a series of smaller precleaned LDPE bottles and stored in the dark at ~4 °C until analysis. Major cations and anions were determined via ion chromatography after appropriate dilution with MilliQ[™] water (Lyons et al., 2005; Welch et al., 2010). Li and Sr were measured by ICP-OES and Ni, Rb, Ba and Pb measured by ICP-MS on aliquots diluted such that the total dissolved salts (TDS) were less than 1 g L^{-1} and acidified to 2% v/v HNO₃. Filtration blanks were also processed and analyzed as samples. Phosphate, $NO_3^- + NO_2^-$, NH_4^+ and H_4SiO_4 were measured using a Skalar San + + nutrient analyzer. The bicarbonate concentration was determined by

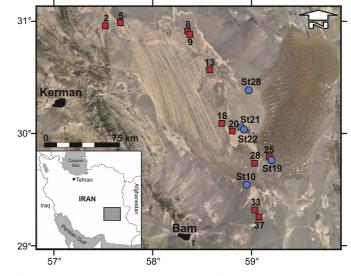


Fig. 1. Location of water (blue circles) and soil (red squares) sampling sites across the Lut Desert, east Iran. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

the difference in charge balance between the measured cations and anions (Lyons et al., 2012). The precision of all directly measured constituents was better than 10% (most were better than 5%), based on replicate analysis of samples and standards (Welch et al., 2010). Our previous work suggests that HCO3⁻ determined by charge balance difference are usually better than \pm 14% (Lyons et al., 2012). Water isotope composition, δD and $\delta^{18}O$, were determined using a Picarro water isotope analyzer (Carey et al., 2016).

Table 1

Table 2Water chemistry for Lut Dessert samples.

| | Cl | SO_4 | $NO_3 N$ | Br | F | HCO_3 |
|--|---|--|---|---------------------------------|-------------------|--------------------------------------|
| | mM | mM | mM | mM | mM | mM |
| St 10 | 284 | 40.6 | 0.866 | 0.099 | 0.035 | 75.5 |
| St 19 | 783 | 21.9 | 12.34 | 0.019 | na | 41.2 |
| St 21 | 634 | 32.7 | 8.72 | 0.019 | na | 36.9 |
| St 22 | 1874 | 28.9 | 30.85 | 0.027 | 0.032 | 263 |
| St 28 | 5373 | 5.3 | 82.83 | 0.052 | 0.119 | 66 |
| | Na mM | K mM | Mg mM | Ca mM | Li mM | Sr mM |
| St 10 | 394 | 2.01 | 10.8 | 12.3 | 0.718 | 0.187 |
| St 19 | 688 | 1.86 | 13.9 | 81.3 | 0.189 | 0.702 |
| St 21 | 596 | 1.54 | 22.3 | 51.7 | 0.225 | 0.506 |
| St 22 | 1910 | 6.77 | 65.3 | 94.5 | 0.647 | 1.11 |
| St 28 | 5250 | 8.86 | 89.3 | 353 | 2.18 | 3.27 |
| | D D0 | | NO | 0. | δ ¹⁸ 0 | <u></u> |
| | Ρ ΡΟ ₄ μΜ | N NH ₄ μM | NO ₂ + NO ₃ mM | Si μM | 8°°0 ‰ | δD ‰ |
| St 10 | 1.58 | 8.54 | 1.13 | 340 | -3.35 | -19.96 |
| St 19 | 0.53 | 21.4 | 12.3 | 282 | 5.64 | -14.16 |
| St 21 | 0.33 | 24.5 | 9.00 | 262 | 4.46 | -14.65 |
| 31 21 | 0.55 | | | | | 0.11 |
| St 21 St 22 | 1.04 | 46.0 | 35.1 | 279 | 6.35 | -9.11 |
| | | | 35.1 81.5 | 279 169 | 6.35 17.86 | -9.11 26.91 |
| St 22 St 28 | 1.04 2.56 | 46.0 97.5 | 81.5 | 169 | | 26.91 |
| St 22 | 1.04 | 46.0 | | | | |
| St 22 St 28 | 1.04 2.56 Ni | 46.0 97.5 Rb | 81.5 Ba | 169 Рb | | 26.91 TDS |
| St 22 St 28 | 1.04 2.56 Ni μg/l | 46.0 97.5 Rb μg/1 | 81.5 Βa μg/l | 169 Ρb μg/l | | 26.91 TDS g/kg |
| St 22 St 28 ICP MS St 10 | 1.04 2.56 Νi μg/1 24.5 | 46.0 97.5 Rb μg/1 43.6 | 81.5 Ba μg/l 125 | 169 Pb μg/l 1.5 | | 26.91 TDS g/kg 27.9 |
| St 22 St 28 ICP MS St 10 St 19 | 1.04 2.56 Νi μg/l 24.5 112 | 46.0 97.5 Rb μg/l 43.6 46.4 | 81.5 Ba μg/l 125 139 | 169 Pb μg/l 1.5 1.0 | | 26.91 TDS g/kg 27.9 50.8 |

3. Results and Discussion

The water geochemistry data and soil chemical parameters are shown in Tables 2 and 3, respectively. A number of general observations of these data can be made. All the waters are Na–Cl, but they are unlike seawater derived brines in that $Ca^{2+} > Mg^{2+}$ and $HCO_3^- > SO_4^{2-}$ (Fig. 2). The K concentrations are uniformly low relative to Na. Unlike some hypersaline waters in extremely hyper arid environments, the Li, Mg, and Br concentrations are relatively low (Long et al., 1992; Lyons and Welch, 1997; Vengosh, 2013). In fact, the increasing Cl:Br ratios with increasing Cl⁻ suggest extensive halite dissolution has helped increase the Cl concentration relative to Br in these waters (Fig. 3) (McCaffrey et al., 1987). With the possible exception of sample 10, the N:P ratios are extremely high, ~20,000 to 30,000:1, much higher than found in most natural waters (Berner and

| | Та | ble | 3 |
|--|----|-----|---|
|--|----|-----|---|

| Chemical properties | of soil | samples | from | Lut Desert. |
|---------------------|---------|---------|------|-------------|
|---------------------|---------|---------|------|-------------|

| Sample ID | pH | ECµS/cm | C_t % | GypsumWt % |
|-----------|------|---------|---------|------------|
| 2 | 9.12 | 859 | 0.95 | 8.92 |
| 5 | 8.57 | 836 | 1.16 | 17.59 |
| 8 | 7.92 | 4481 | 0.89 | 60.68 |
| 9 | 7.96 | 5780 | 1.12 | 71.00 |
| 13 | 9.18 | 1160 | 0.33 | 2.75 |
| 18 | 8.62 | 370 | 1.58 | 6.01 |
| 20 | 8.28 | 9210 | 1.05 | 50.79 |
| 25 | 8.89 | 1513 | 0.86 | 8.57 |
| 28 | 9.10 | 560 | 1.67 | 5.50 |
| 33 | 8.91 | 1298 | 1.92 | 7.43 |
| 37 | 7.59 | 2884 | 1.74 | 41.17 |

Berner, 2012; Schlesinger and Bernhardt, 2013). These high concentrations of $NO_3^- + NO_2^-$ indicate the accumulation of NO_3^- within the landscape, minimal biological uptake of N, and the lack of extensive denitrification within the shallow groundwater (Fig. 3).

The accumulation of soluble salts in a landscape occurs when evaporation exceeds precipitation and there is little to no flushing of the salts. Shallow saline groundwater can develop from a combination of two important processes: by meteoric waters being evaporated, hence concentrating the solutes along its flow path, and by the dissolution of previously accumulated salts. The former process can be traced by a change in water isotope signals (i.e. δD and $\delta^{18}O$) with salinity, while the latter through the observance of an increase in the ratios of certain solutes such as Cl:Br with salinity (Vengosh, 2013). As noted above and seen in Table 2, the Cl:Br of these waters increase with Cl⁻ (Fig. 3), suggesting strongly that the dissolution of previously deposited halite is a major control on the overall salinity of these waters. This appears to be particularly the case in the three most saline samples where the Cl:Na ratio is ~ 1 (Table 2, Fig. 3). This same phenomenon has been observed in other parts of Iran (Jalali, 2007; Nassery and Kayhomayoon, 2013).

We used the Pitzer database in PHREEQC (US Geological Survey, v 3.4.0, Parkhurst and Appelo, 2013) to calculate the saturation state of a number of common binary salts in these brines (Table 4). We were unable to measure pH in the field, so we have done these calculations using both pH 7 and 8, although only the CaCO₃ polymorphs and dolomite are greatly affected by the pH variations. The pH of the soil slurry measurements were all slightly alkaline (Table 3) with pH ranging from 7.59 to 9.18, thus only the modelling results for pH 8 are presented in Table 4. Calculations were also done at 25, 50 and 70 °C to estimate mineral saturation index (SI) over the range of temperatures expected in the field (Alavipanah et al., 2007; Rahimzadeh et al., 2009). The carbonate minerals were supersaturated in all the samples with greater saturation indices for samples with higher salinities (Tables 3 and 4). The saturation index for carbonate minerals increased with temperature due to their retrograde solubility, suggesting that increased temperature, as well as increased evaporation, should induce carbonate mineral precipitation from these saline brines. All waters were at, or very near, equilibrium with respect to gypsum and anhydrite (SI ranging from -0.64 to 0.34) with values generally increasing with increasing salinity. The calculated SI values decrease with temperature for gypsum, but increase for anhydrite, suggesting that the extreme temperatures in this environment favors the formation of anhydrite over gypsum. Most waters were also slightly supersaturated or very near equilibrium with respect to barite at 25 °C, except for the most saline water (sample 28), which was slightly undersaturated. Though saturation index decreases with increasing temperatures, suggesting that barite precipitates could start dissolving in the brines at in situ conditions. With the exception of sample 10, most of the brines were slightly supersaturated with respect to celestite (SrSO₄), and like barite, SI decreases with increasing temperature, suggesting that the temperature variations described in this environment could be important controls for the solubility of Ca, Sr, Ba, and SO₄ in these shallow brines. The soil data from the top 10 cm support these calculations in that the abundant total soil carbon content (C_{Total}) values range from ~1 to 2% suggesting significant amounts of CaCO₃. The gypsum concentrations are also very high but quite variable, with values between 2.75 and 71% of the total soil mass (Table 3). There is a correlation ($R^2 = 0.68$) between gypsum content of soil and EC suggesting evaporation has played a major role in chemical evolution of both soil and brine in the Lut Desert (Figs. 3 and 4, Fig. S3). Most of the waters were slightly undersaturated with respect to amorphous SiO2. Halite is undersaturated except in the most saline brine sample (Sample 28). These calculations, along with the soil data suggest that the precipitation of gypsum (CaSO₄•2H₂O) or anhydrite (CaSO₄), and the dissolution of previously deposited NaCl, are major controls on the Na⁺, Ca²⁺, SO₄²⁻ and Cl⁻ concentration in these waters and that the upper limit of the

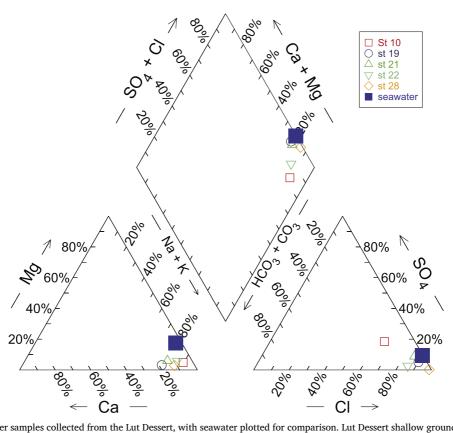


Fig. 2. Piper diagram of water samples collected from the Lut Dessert, with seawater plotted for comparison. Lut Dessert shallow groundwaters are all Na–Cl brines. Seawater data from Pilson (2012).

 H_4SiO_4 is probably controlled by amorphous SiO_2 precipitation. The latter hypothesis is also supported by the decrease of Si:Cl with increasing Cl concentration (Fig. 2), which indicates that Si is not behaving conservatively as the brine is concentrating. The low K

concentrations and the high Ca:Mg ratios may suggest that K^+ and Mg^{2+} concentrations are controlled by either adsorption of K and Mg onto silicate minerals, or authigenic clay mineral formation. Similar K loss has been observed along the flow path of shallow groundwaters in

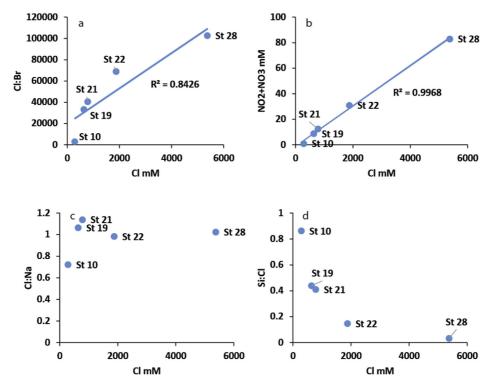


Fig. 3. Major ion ratio plots for water samples from the Lut Desert. a) Cl:Br vs Cl, b) NO3+NO2 vs Cl, c) Cl:Na vs Cl and d) Si:Cl vs Cl.

| Table 4 | |
|---|--|
| Saturation Indices of common binary salts calculated at 25, 50 and 70 $^\circ \! C$ and | |
| pH 8 for Lut Desert water samples. | |

Table 4

| Mineral | St 10 | St 19 | St 20 | St 21 | St 28 |
|----------------------|-------|-------|-------|-------|-------|
| рН 8 25°С | | | | | |
| Anhydrite | -0.64 | -0.31 | -0.28 | -0.23 | 0.26 |
| Gypsum | -0.3 | 0.01 | 0.05 | 0.05 | 0.2 |
| Barite | 0.66 | 0.25 | 0.18 | 0.1 | -0.56 |
| Celestite | -0.15 | -0.03 | 0.06 | 0.02 | 0.5 |
| Aragonite | 2 | 2.58 | 2.31 | 3.58 | 4.29 |
| Calcite | 2.28 | 2.87 | 2.59 | 3.87 | 4.57 |
| Dolomite | 4.55 | 5.05 | 4.91 | 7.55 | 8.79 |
| Halite | -2.86 | -2.17 | -2.33 | -1.24 | 0.57 |
| SiO ₂ (a) | -0.77 | -0.8 | -0.85 | -0.65 | -0.19 |
| рН 8 50°С | | | | | |
| Anhydrite | -0.36 | -0.04 | -0.01 | 0.05 | 0.32 |
| Gypsum | -0.31 | 0 | 0.04 | 0.04 | 0 |
| Barite | 0.41 | -0.03 | -0.08 | -0.19 | -0.87 |
| Celestite | -0.06 | 0.05 | 0.15 | 0.13 | 0.48 |
| Aragonite | 2.27 | 2.84 | 2.57 | 3.79 | 4.39 |
| Calcite | 2.6 | 3.18 | 2.9 | 4.13 | 4.72 |
| Dolomite | 5.22 | 5.69 | 5.55 | 8.06 | 9.06 |
| Halite | -2.9 | -2.21 | -2.37 | -1.27 | 0.48 |
| SiO ₂ (a) | -0.99 | -1.03 | -1.07 | -0.89 | -0.59 |
| рН 8 70°С | | | | | |
| Anhydrite | -0.14 | 0.17 | 0.21 | 0.25 | 0.34 |
| Gypsum | -0.28 | 0.01 | 0.05 | 0.04 | -0.17 |
| Barite | 0.25 | -0.21 | -0.26 | -0.4 | -1.09 |
| Celestite | 0 | 0.1 | 0.2 | 0.17 | 0.43 |
| Aragonite | 2.46 | 3.02 | 2.75 | 3.95 | 4.48 |
| Calcite | 2.8 | 3.37 | 3.09 | 4.29 | 4.83 |
| Dolomite | 5.53 | 5.99 | 5.85 | 8.28 | 9.14 |
| Halite | -2.92 | -2.23 | -2.39 | -1.28 | 0.41 |
| SiO ₂ (a) | -1.14 | -1.18 | -1.23 | -1.07 | -0.87 |

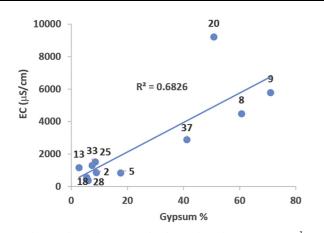


Fig. 4. The correlation between soil's electrical conductivity (μ S cm⁻¹) and gypsum content (%) in soil samples from Lut Desert.

the Atacama Desert (Magaritz et al., 1989). Except for sample 10, the Ca/Sr ratios are very similar to seawater (mean Ca/Sr 103 compared to seawater Ca/Sr of 113). Sample 10 more closely reflects the crustal ratio (of 66), indicating along with the highest H₄SiO₄ that this water reflects a chemical weathering component that becomes less significant at higher salinities. The concentrations of most trace elements detected (Li, Ni, Rb, Ba, Sr, Pb, as well as NH_4^+) generally correlate with salinity of the brines. The wetting and drying cycles are important in this system as demonstrated by both the very high Cl:Br ratios, and the mineral saturation calculations, which, as noted above, indicate that the dissolution of previously deposited NaCl is a very significant process. Drever and Smith (1978) have demonstrated that cyclic wetting and drying in arid climates can lead to brines depleted in SO₄²⁻, Mg²⁻ and Si relative to Cl⁻, similar to what we observe in these waters. This is due to the kinetics of dissolution of previously deposited salts with the most soluble salts redissolving more readily than the less soluble salts.

The stable isotopes of water are shown in Table 2 and plotted in Fig. 5A. The global meteoric water line as well as the local meteoric water lines (LMWL) for Tehran and snowmelt from the Zagros Mountains to the north and west of the Lut Desert are plotted for comparison (Mosaffa et al., 2015; Osati et al., 2014). The least saline sample (10) plots closest to the meteoric lines and is also similar to the modeled "isoscapes" precipitation value for this general region (Bowen and Revenaugh, 2003). Samples 21, 19 and 22 plot obliquely to the LMWLs (Fig. 5A) suggesting extensive evaporation, while sample 28, the most saline water, plots very far off the lines indicating massive evaporative loss. These data coupled with the Cl:Br values strongly indicate that both extensive evaporation and salt dissolution have played a role in producing these hypersaline waters.

Spatial distribution patterns of δ^{18} O and δ D values show significant depletion from station 28 to station 10 (Fig. 6, Fig. S4), where a one degree equatorward shift causes around 21 and 48‰ depletion in δ^{18} O and δ D values, respectively. The observed gradients in isotopic values of Lut water samples are too high to be explained by the latitude effect (*e.g.* Bershaw et al., 2012; Bowen and Wilkinson, 2002). The effect of altitude as well as the amount of precipitation (altitude and amount effects) cannot explain such a shift either, due to very low annual precipitation and low elevation gradient in Lut desert. A strong correlation (R² = 0.8) is observed in changes in δ^{18} O values with respect to total dissolved solid (TDS), Fig. 5B. This observation supports the idea that a high gradient shift in δ^{18} O and δ D values are related to the salinity of waters caused by evaporation processes (Conroy et al., 2014).

3.1. Implications for other hyper-arid environments

We have reviewed the available data from a number of other hyperarid regions, and the geochemistry of the soils/shallow groundwaters have great similarities. Shallow groundwaters in Death Valley/Mojave

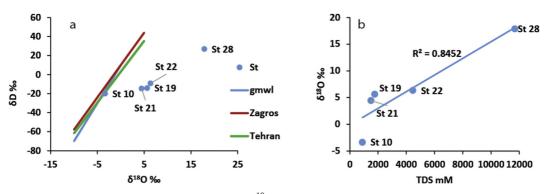


Fig. 5. A) Isotope data plots and 5B) the δ^{18} O-TDS relationship for Lut Desert water samples.

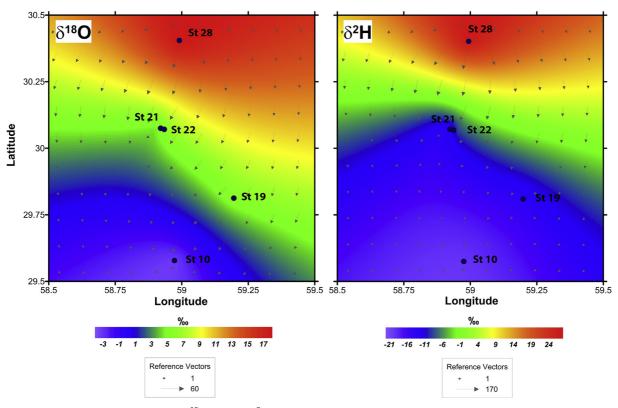


Fig. 6. Gradient of $\delta^{18}O$ (left) and δ^2H (right) in the Lut Desert. Arrows denote gradient vectors.

Desert, USA, the Atacama, Chile, and the Turpan Basin, NW China have high TDS, and are primarily Na-Cl waters, although some Na-SO4 waters also exist (Boutt et al., 2016; Chen et al., 2014; Thomas et al., 1996). These waters are evapoconcentrated meteoric waters that have also solubilized previously deposited evaporite minerals, such as gypsum, mirabilite and halite (Chen et al., 2014; Magaritz et al., 1989; Thomas et al. 1996). All of this information indicates that periodic wetting and the precipitation/dissolution cycling of soluble salts play a very important role in controlling the geochemistry of all these waters. The vadose zones of these environments, as well as the shallow groundwaters, can accumulate large amounts of very soluble ions, such as nitrate (Finstad et al., 2016; Lybrand et al., 2013; Qin et al., 2012). High $NO_3^- + NO_2^-$ concentrations in groundwater are observed in other hyper-saline systems such as the Negev Desert, and Central Australia (Barnes et al., 1992; Rosenthal et al., 1987). This NO₃⁻ is presumably deposited from the atmosphere and accumulates over time (Böhlke et al., 1997; Lyons et al., 2016) or was initially fixed biologically, oxidized and flushed from the unsaturated zone (Barnes et al., 1992; Rosenthal et al., 1987). Associated with the nitrate can be both perchlorate and iodate salts (Lybrand et al., 2016). We have observed another unidentified peak in the anion chromatograms which falls where the perchlorate anion would generally be seen. Perchlorate has been observed in other hyperarid regions in the world, including the Atacama and the McMurdo Dry Valleys of Antarctica (Ericksen, 1981; Jackson et al., 2015, 2012). In all these environments a high percentage of this nitrate originates from the atmosphere, suggesting, like in the Lut Desert, the role of biological processes in controlling N distribution is probably minor. Our data from the Lut, when compared to these other settings, suggest that the processes that control the geochemistry of these surficial and shallow subsurface environments are similar. The abundance of NO3⁻, and the potential presence of perchlorate, suggests little biological alteration of these very soluble salts.

The accumulation of these salts in soils and shallow groundwaters and the partitioning between the two are related to the stability of the landscape, the climate, and the landscape age, as previously noted by

Lybrand et al. (2016). Additions of water clearly can remove salts from the soils solubilizing and transporting them into the shallow groundwaters. Conversely, extensive hyper-aridity can lead to the precipitation of salts. Cation exchange and possibly authigenic aluminosilicate mineral formation in the shallow groundwaters can influence K concentrations, as we have observed in the Lut and others have observed in desert waters (Magaritz et al., 1989), as well as influence both the vertical and horizontal distribution of soluble binary salts in soils (Boutt et al., 2016; Finstad et al., 2016; Lybrand et al., 2013). The lack of halite saturation may also suggest that the basin is not hydrologically closed and water is being lost at depth through leakage as outlined in Wood and Sanford (1990) and Rosen (1994), as non-evaporative groundwater loss can result in steady state solute concentrations that do not approach more soluble salt deposition (Wood and Sanford, 1990). As noted above, the Lut Block is highly faulted, and tectonic controls on groundwater geochemistry in Iran have previously been noted (Masoodi and Rahimzadeh, 2018).

4. Conclusions

We present, to our knowledge, the first geochemical analysis of water from one of the hottest locations on the Earth, the Lut Desert, Iran. The data indicate that the salinity of these waters is produced by both evaporative processes and dissolution of previously deposited salts, and thus wetting and drying are very important factors in controlling brine geochemical evolution. Thermodynamic calculations indicate that waters are essentially in equilibrium with gypsum and are also highly supersaturated with respect to carbonate minerals. Chemical weathering of aluminosilicate minerals, in what we assume to be the fresher regions of the flow systems, produce high H_4SiO_4 concentrations that may be later controlled by equilibrium with amorphous SiO_2 precipitation as water is lost and brine becomes more saline. Soluble ions such as NO_3^- are found at very high concentrations in these waters suggesting little to no removal via biological processes.

environments are not evapoconcentrated to a great extent. The data presented within adds important new information on the geochemical evolution of hyper-arid environments from a system heretofore undescribed.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jaridenv.2020.104143.

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