Using Geochemical and Environmental Isotopes Composition to Study the Possible Leakage between Two Major Aquifers, Eastern Saudi Arabia

MAHMOUD S. ALYAMANI and MOHAMMED A. SHARAF
Faculty of Earth Sciences, King Abdulaziz University
P.O. Box 80206, Jeddah 21589, Saudi Arabia

Received: 27/11/2004 Revised: 22/2/2005 Accepted: 26/2/2005

ABSTRACT. Major ion analyses of 156 fresh and mineral groundwater samples from Umm Er Radhuma aquifer (UER) in eastern part of Saudi Arabia reveal a consistent pattern of variation in the sedimentary basin. The piezometric data shows a broad pattern of regional flow from west to east towards the Arabian Gulf coast. The groundwater chemistry is highly variable, where TDS ranged between 220 and 15815 mg/l; with an average of about 2257 mg/l. The chemical and isotopical compositions of the groundwater of UER aquifer show that the water is progressively enriched in Na⁺, Ca²⁺, Mg²⁺, Cl⁻ and SO₄²⁻ while isotopically (δ¹⁸O and δD) depleted in the direction of flow. The isotopic contents of the groundwater of UER, measurements of the piezometric level and stratification of groundwater chemistry with depth suggest that the UER aquifer receives a considerable amount of water leaking from the overlying Dammam Formation characterized by being younger and low in TDS. The groundwater chemistry in UER aquifer, therefore, is mainly controlled by mixing processes besides, the chemical reactions that take place between water and aquifer materials within the UER aquifer through its movement.

KEYWORDS: Groundwater chemistry, Leakage process, Sedimentary basin.

Introduction

The Umm Er Radhuma (UER) formation lies within latitude 28°41’N and longitude 44°41’E in the eastern province of Saudi Arabia (Fig. 1). It covers a surface area of about 160,000 km². Hydrogeologically and hydrochemically, the
Mahmoud S. Alyamani & Mohammed A. Sharaf

Table 1. Stratigraphic succession in the study area.

<table>
<thead>
<tr>
<th>Age</th>
<th>Formation</th>
<th>Generalized lithologic description</th>
<th>Thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cenozoic</td>
<td></td>
<td>Limestone, dolomite, marl and shale.</td>
<td>38 m</td>
</tr>
<tr>
<td>Tertiary</td>
<td>Dammam</td>
<td>Limestone, dolomite, marl and shale.</td>
<td></td>
</tr>
<tr>
<td>Eocene</td>
<td>Rus</td>
<td>Marl, chalky limestone and gypsum. Dominantly anhydrite in subsurface.</td>
<td>56 m</td>
</tr>
<tr>
<td>Paleocene</td>
<td>Umm-Er-Radhuma</td>
<td>Limestone, dolomitic limestone and dolomite.</td>
<td>243 m</td>
</tr>
<tr>
<td>(UER)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cretaceous</td>
<td>Aruma</td>
<td>Limestone: subordinate dolomite and shale. Lower part grades to sandstone in north-western and southern areas of outcrop.</td>
<td>142 m</td>
</tr>
</tbody>
</table>

The UER aquifer has been studied intensively since early 1970's (e.g. Watuki, 1968 and 1971; Sogreah, 1968; Italoconsult, 1969; Groundwater Development Consultant Company (GDC), 1979; BRGM, 1976 and 1979; Al-Sayari and Zotl, 1978; Al-Bassam, 1983, and 1987; Sharaf, 2001). It is considered a major groundwater reservoir of the Arabian Peninsula. Throughout the study area, the groundwater chemistry is highly variable. The available data concerning the chemical and isotopical composition of the groundwater indicated that an anomalous pattern existed. It shows that the groundwater down gradient becomes progressively enriched in major constituents, while its isotopic content is being depleted. Normally, it would be expected that the isotopic composition often followed similar trend that was observed in the chemical composition and to be enriched with heavy isotopes. The existence of this variation led us to investigate the groundwater conditions and define factor(s) that affect its observed chemistry and isotopic contents. The data concerning the hydrogeological and hydrochemical characteristics used by Sharaf (2001) was adopted in the present investigation.

Geology and Hydrogeology

The rock types, formed in a shallow sea, consist of a series of light colored, dense limestones, dolomitic limestones, and dolomite. Marl and shale are also found in the upper part of the formation in the central and southern areas. In the north, significant layers of anhydrite and some chert are found in the upper part. The stratigraphic succession is presented in (Table 1).
aquifer, the groundwater mainly moves through secondary porosity, and inter-
granular permeability is expected to be imprecise (Sharaf, 2001). The ground-
water flow in the study area is illustrated in (Fig. 2). A reasonably consistent set
of flow conditions can be noted by comparing the flow directions indicated by
arrows is mainly eastward down the regional dip, but detailed examination of
the data in the study area shows how the pattern of flow is probably controlled
by geological structures. For example, the tendency for groundwater to move in
northeastern direction is probably related to north-south regional fold (see Fig.
1). Groundwater occurs under semi-confined to confined conditions. The aver-
age transmissivity values of $7 \times 10^{-5}$ to 0.62 m$^2$ s$^{-1}$ while the average storativ-
ity of the aquifer is in the range of $10^{-5}$ and $10^{-3}$. The recharge of the aquifer
depends on direct infiltration of seasonal rainfall and by infiltration of runoff
from wadis; as is the case in other aquifers in Saudi Arabia. The average annual
rainfall is $> 100$ mm. The recharge is very limited, and is estimated that 4 to 8
mm infiltrate into the aquifer (BRGM, 1979). The age of the groundwater has
been determined to be 22,000 years (Al-Bassam, 1987).

Fig. 1. Location and general geology map.
Result and Discussion

Nearly 156 groundwater samples from UER aquifer were analyzed for major ions (e.g. Na\(^+\), K\(^+\), Ca\(^{2+}\), Mg\(^{2+}\), Cl\(^-\), HCO\(_3\)^- and SO\(_4\)^2-). Standard method (APH/AWWA/WPCF, 1989) was used. To obtain a better understanding of the groundwater behaviour in contact with different geological formations, the saturation indices of calcite, dolomite and gypsum minerals were calculated, using the hydrochemical program called “Phreeqc” (2001). In addition, 40 groundwater and 8 rainwater samples collected from the recharging zone of the aquifer were taken for stable isotopes analysis. The chemical composition of the groundwater from Dammam aquifer provided by Hassan (1998) was used for comparison purposes. Unfortunately, the saturation indices of the above mentioned minerals of the groundwater from the Dammam aquifer have not been calculated due to the absence of pH and groundwater temperature in the chemical analyses provided by the author. Ranges and mean of the major constituents are summarized in (Table 2).
The chemical analyses results of the groundwater of UER aquifer are highly variable. They show that cation composition varies between almost exclusively Ca\textsuperscript{2+} to dominantly Na\textsuperscript{+} and Ca\textsuperscript{2+} with relatively lesser amount of K\textsuperscript{+}. Among the anions, Cl\textsuperscript{−} and SO\textsubscript{4}\textsuperscript{2−} are dominant (Table 2). On the other hand, the groundwater of UER aquifer is supersaturated with respect to calcite and dolomite but with exception of a few samples, the groundwater is undersaturated with respect to gypsum (Table 2). The areal distribution maps of the major ions were constructed and shown in Fig. 3 to 5. Iso-maps for bicarbonate and potassium ions are rather difficult to draw and are not shown here, since neither constituent displayed the pronounced down gradient increase of the other constituents but rather appeared randomly distributed within the area. Figure 3(a) illustrates the areal distribution map of Ca\textsuperscript{2+} ion. It generally shows that Ca\textsuperscript{2+} concentrations increase from the outcrop to the coastal area, from less than 50 mg/l to more than 1500 mg/l, with no systematic variation observed in its concentration followed the groundwater movement pattern (Fig. 2). It rather shows that there are several tongues of Ca\textsuperscript{2+} contents extending from the south toward north direction. It covers a large area and its concentration varies between 200 to 600 mg/l. However, high contents of Ca\textsuperscript{2+} are found near the coastal area and reached up to 2000 mg/l. On the other hand, Mg\textsuperscript{2+} contents (Fig. 3b) are as low as 9 mg/l at the outcrop area and increases to values ex-

<table>
<thead>
<tr>
<th>Ion</th>
<th>Umm Er Radhuma (156)</th>
<th>Dammam** (75)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Min</td>
<td>Max</td>
</tr>
<tr>
<td>Ca\textsuperscript{2+}</td>
<td>35.0</td>
<td>1440.0</td>
</tr>
<tr>
<td>Mg\textsuperscript{2+}</td>
<td>7.0</td>
<td>456.0</td>
</tr>
<tr>
<td>Na\textsuperscript{+}</td>
<td>22.0</td>
<td>3882.0</td>
</tr>
<tr>
<td>HCO\textsubscript{3}\textsuperscript{−}</td>
<td>6.0</td>
<td>390.0</td>
</tr>
<tr>
<td>Cl\textsuperscript{−}</td>
<td>12.0</td>
<td>7891.0</td>
</tr>
<tr>
<td>K\textsuperscript{+}</td>
<td>0.0</td>
<td>84.0</td>
</tr>
<tr>
<td>S\textsubscript{4}\textsuperscript{2−}</td>
<td>13.0</td>
<td>2700.0</td>
</tr>
<tr>
<td>SI\textsubscript{calcite}</td>
<td>−1.10</td>
<td>2.06</td>
</tr>
<tr>
<td>SI\textsubscript{dolomite}</td>
<td>−0.06</td>
<td>4.74</td>
</tr>
<tr>
<td>SI\textsubscript{gypsum}</td>
<td>−2.37</td>
<td>0.08</td>
</tr>
<tr>
<td>TDS</td>
<td>220</td>
<td>15815</td>
</tr>
</tbody>
</table>

**After Hassan (1998), ( ) Number of samples, SI (Saturation Index).
Fig. 3. Areal distribution maps (a) calcium; and (b) magnesium.
ceeding 300 mg/l near the coastal region. Generally, Mg\(^{2+}\) distribution is similar to those observed for the Ca\(^{2+}\) concentration. The Na\(^{+}\) concentrations (Fig. 4a) increase from less than 25 mg/l at the outcrop to more than 3800 mg/l in the coastal area. The distribution map of Na\(^{+}\) shows that there is a zone of low Na\(^{+}\) in the central region and extending from the south toward north with maximum Na\(^{+}\) contents of less than 400 mg/l. Toward the coastal area and northeast region, the Na\(^{+}\) concentrations increased and reached up to 4000 mg/l. The distribution of Cl\(^{-}\) contents is shown in Fig. 4b. The Cl\(^{-}\) values range from less than 50 mg/l at the outcrop to more than 4000 mg/l near the coastal area. Although, Cl\(^{-}\) ion contents usually exceed Na\(^{+}\) ion concentrations, the Cl\(^{-}\) distribution follows the same pattern as that of Na\(^{+}\). Therefore, the same anomalies discussed for Na\(^{+}\) are prominent in the case of Cl\(^{-}\). The areal distribution of SO\(_{4}\)\(^{2-}\) ion is shown in Fig. 5. The SO\(_{4}\)\(^{2-}\) concentration range between less than 100 mg/l at the outcrop to more than 2000 mg/l in the northern central zone and near the coastal area. On the other hand, two areas can be distinguished with respect to SO\(_{4}\)\(^{2-}\) concentrations. In the northern half is enriched with SO\(_{4}\)\(^{2-}\) as the concentrations rise sharply to exceed 2000 mg/l, whereas in the southern part, the contents remained comparatively low for a considerable distance from the outcrop area. Beyond this distance, the SO\(_{4}\)\(^{2-}\) concentration starts increasing to reach a high value of more than 1800 mg/l in the southeast corner.

On the other hand, data for \(\delta D\) and \(\delta^{18}O\) were reported in the usual delta notation with SMOW as the standard (Graig, 1961). It shows that the \(\delta D\) values range from –55.4‰ to –2.11‰, and \(\delta^{18}O\) values range from –6.46‰ to –1.18‰ (Table 3). Areal distributions of \(\delta D\) and \(\delta^{18}O\) concentrations are shown in Fig. (6a) and (6b). They generally indicate that both elements are depleted downgradient rather than being enriched. Moreover, in the area to the west to Haradh village further south of the study area and southward, the groundwater isotopic contents display highly negative values, although, this area is characterized by an extremely arid conditions and lack of recharge (Al-Bassam, 1987).

As shown from the distribution maps of major ion contents no systematic variation in the major ion contents appears, and concurrent with groundwater flow direction, but rather several tongues of low concentrations occurred and almost prominent the middle zone and extend away from the outcrop towards the east, south and northeast. On the other hand, the isotopic contents of the groundwater almost show that the groundwater is depleted downgradient, which would not be expected but often being enriched with groundwater travel through its flow-path to east and north directions. Under this condition, it is believed that a strong potential factor may affect the groundwater chemistry in the area.

On the other hand, one important question to be considered is whether flow conditions that exist at present were the same over the last 22,000 years. Then
Fig. 4. Areal distribution maps (a) sodium; and (b) chloride.
why the groundwater down gradient is more saline and rather depleted in its isotopic composition? As the aquifer system is a relatively permeable and attains an active flow system (Sharaf, 2001), it is difficult to see why the groundwater in UER aquifer has not been completely flushed by low saline recharge water displacing preexisting formation water. On the other hand, the isotopic content of the groundwater is completely different from that found in the modern recharging water (Table 3), which is enriched in heavy isotopes. This may lead to suggest that the UER is probably receiving sufficient contribution of younger (lighter isotopic composition), and a relatively low saline water (Table 2) from the overlying Dammam aquifer through the thickening 56 m but fractured and fissured anhydrite and gypsum of the Rus Formation (Table 1), and the mixing process might be continuous as a result of leakage. This fact is substantiated by the following evidences which come from the chemical composition of the groundwater of UER aquifer.

The chemical composition of the groundwater of UER aquifer shows that a relatively high contents of SO\textsubscript{4}\textsuperscript{2–} ions in the aquifer mainly composed of limestone and dolomite. However, high concentrations of SO\textsubscript{4}\textsuperscript{2–} seem to have entered the solution through dissolving anhydrite and gypsum of the Rus Formation due to vertical flow of water from Dammam aquifer. This conclusion could be confirmed by plots of Ca\textsuperscript{2+} against HCO\textsubscript{3}–, SO\textsubscript{4}\textsuperscript{2–} and HCO\textsubscript{3}– + SO\textsubscript{4}\textsuperscript{2–} (Fig. 7). This figure indicated that neither limestone (CaCO\textsubscript{3}) of UER aquifer (Correlation Coefficient R\textsuperscript{2} = 0.001) nor gypsum (CaSO\textsubscript{4}.2H\textsubscript{2}O) and anhydrite
TABLE 3. Ranges of the isotopic contents of the groundwater of UER aquifer and rainwater.

<table>
<thead>
<tr>
<th>Element</th>
<th>Umm Er Radhuma (40)</th>
<th>Rainfall (8)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Min</td>
<td>Max</td>
</tr>
<tr>
<td>$\rho^{18}O$ (%o)</td>
<td>–6.46</td>
<td>–1.18</td>
</tr>
<tr>
<td>$\Delta D$ (%o)</td>
<td>–55.4</td>
<td>–2.11</td>
</tr>
</tbody>
</table>

( ) Number of samples.

(CaSO$_4$) ($R^2 = 0.75$) of the Rus Formation are by themselves control Ca$^{2+}$ concentrations, but together the carbonate and sulfate minerals dissolution do balance the concentration trend of Ca$^{2+}$ in the groundwater of UER aquifer ($R^2 = 0.84$). Another possible indication of leakage phenomena from the overlying aquifer can be depicted from Durov’s diagram (Fig. 8). This diagram indicates that most of groundwater samples plotted along the mixing and dissolution line, suggesting that mixing processes between low mineralized water (Dammam aquifer) with a relatively high saline water (UER aquifer). Figure 9, on the other hand, illustrated the relationship between Cl$^–$ and $\delta^{18}O$. It reflects that two groups can be distinguished. Group I contains 26 wells. This group has a relatively homogeneous chemical and isotopic composition. This group is probably affected by the groundwater leaked from the upper aquifer. Group II has a higher Cl concentration. This group might reflect the original chemical and isotopic compositions of the groundwater of UER aquifer. However, such variation in Cl$^–$ and $\delta^{18}O$ contents observed may indicate that stratification in the groundwater chemistry exist. This conclusion might be supported by the previous investigation carried out by Al-Bassam (1987). He outlined that the groundwater salinity increased with depth, and Na$^+$ and Cl$^–$ ions contents are increased. Whereas, Ca$^{2+}$, Mg$^{2+}$ and SO$_4^{2–}$ being decreased. His study might be considered as an indication confirmed the earlier conclusion that the UER aquifer has received sufficient contribution of younger (lighter isotopic composition), and a relatively low saline water enriched in Ca$^{2+}$ and SO$_4^{2–}$ contents from Dammam aquifer through the Rus Formation and being mixed with relatively high saline water of UER aquifer.

On the other hand, upward leakage from Aruma to the UER aquifer can be ruled out, because it does not agree with the hydrogeological data where the piezometric level of Aruma is lower than the UER piezometric level which suggests a downward leakage from the UER to Aruma aquifer.

It appears that leakage process probably operated for a long time, the flushing is incomplete and the zones of mixing, besides, the geochemical evolution processes between the groundwater and the aquifer materials still remain. More-
FIG. 6. Areal distribution maps; (a) δD, (b) δ¹⁸O (parts per mil relative to SMOW).
Fig. 7. Relationship of Ca$^{2+}$ with HCO$_3^-$, SO$_4^{2-}$, HCO$_3^- +$ SO$_4^{2-}$.
FIG. 8. Plotting of groundwater samples in Durov’s diagram.
over, the effect of vertical downward leakage may continue significantly as a consequence of the decline in the water levels in UER aquifer since the aquifer is being extensively developed for agricultural projects as well as for major domestic supplies in the area.

**Conclusion**

The groundwater in the UER aquifer is controlled by mixing processes due to downward leakage of younger water from the Dammam aquifer depleted in heavy isotopes and low in TDS. This fact is supported by the chemical and isotopic compositions of the groundwater. Furthermore, the water chemistry of the UER aquifer is also controlled by chemical reactions between water and the aquifer itself.

**References**


استخدام التركيب الكيميائي والنظائر البيئية لدراسة احتمال التسرب بين خزانين مائيين رئيسيين، شرق المملكة العربية السعودية

محمود سعيد اليماني، و محمد أمين شرف
كلية علوم الأرض، جامعة الملك عبد العزيز، جدة - المملكة العربية السعودية

الاستخلاص. تحليل العناصر الرئيسية لـ 16 عينة مياه جوفية متمتعة وعذبة من الحزام المائي أم الرضمة في الجزء الشرقي من المملكة العربية السعودية ساده فقط متغير في الحوض الرسوبى. تشير النماذج إلى أن الأملاح الصلبة المذابة تتراوح بين 20 و15815 ملجم/ل مع متوسط حوالي 22 ملجم/ل. التركيب الكيميائي وكذلك تركيب النظائر البيئية للمياه الجوفية من الحزام المائي أم الرضمة بين أن المياه غنية بعناصر الصوديوم والبوتاسيوم والنيتروجين والكلور والكبريت، بينما نزاريا شحينة بعنصر كبريت 18-18 والدكتر سيريان الماء. إن محتوى النظائر البيئية للمياه الجوفية من الحزام المائي أم الرضمة وقياسات مستوى الماء البيومترى والتغير الطيفي للمياه الجوفية مع العمق يقترح أن الخزان المائي أم الرضمة يمثل كميات مياه معقولة ترشح من مكون الدمام الذي يعلوه، والذي يتميز بأنه الأحدث وقلة في مجموع الأملاح الصلبة المذابة، وللذا فإن كيمياء المياه الجوفية للحزام المائي أم الرضمة يتمحول بها أساسا عمليات مائية حالية إلى جانب التفاعلات الكيميائية التي تحدث بين المياه الجوفية ومواد الخزان المائي داخل المتنين خلال حركتها.