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LEACHING OF RUSEIFA PHOSPHATES AND MAQARIN BITUMINOUS LIMESTONE AND ITS EFFECT ON THE QUALITY OF GROUNDWATER

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ملخص

لمت مقاربة كمية بعض المناصر الثقيلة في صحور الفوسفات من منطقة الرصفية، وصحور الحبر الربيبة من منطقة القارن مع كمية من ذلك المناصر في المياة الحوفية في هذه الصحور، ويبين من هذه الدراسات أن صحور الفوسفات هي المصدر الرئيسي لتلوث المياء الحوفية، أما في منطقة المقارن فقد ذلت الدراسة على أن المناصر الثقيلة بتركر في المياء ذات الفلوية العالية والتي تنتج عن ذوبان المعادن البابحة عن كلسنة الحجر الجيري، أما في الصحور الرئيسة عبر المتحولة قان محتوى المياة من المعاصر البديلة عادي

ABSTRACT

The amounts of trace elements in the circulating waters in Ruselfa phosphate rocks and the Maqarin bituminous limestone are compared to the amounts in groundwaters in these areas. Leaching of phosphate rocks with their high trace metal content is responsible for high trace element concentrations in the groundwaters of Ruselfa. In the Maqarin area, the leaching of the high temperature mineralization zone is responsible for the high pH waters enriched with base metals. The uncalcined bituminous limestone yielded normal pH waters without any indication of leaching. Mixing of the alkaline waters with the normal waters could give trace element values.

In the Ruseifa area, the groundwater is not recommended for drinking purposes, in accordance with WHO standards.

INTRODUCTION

Bituminous rocks of Upper Cretaceous and Eocene age and phosphate deposits of Upper Cretaceous age are widely distributed in Jordan. Important outcrops of bituminous limestone occur in the Yarmouk River and Lajjun areas. Phosphate deposits and occurrences are found at Ruseifa, El-Hasa, Suwelleh, Qatrana, Ma'an and Eshidiya. The bituminous limestone and phosphate rocks are particulary enriched with trace elements. Two types of localities were chosen to study the influence of leaching on the inflitrated water. The first locality is the bituminous limestone of the Maqarin area, 16 km N of Irbid (E 252, 250, N 237, 900); the second locality is the phosphate deposit in Ruseifa, 15 km NE of Amman (E 157, 900, N 298, 100, Palestine Grid) (Fig. 1). A comparison between trace elements in the sedimentary rocks and the groundwaters is made in this paper.

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GEOLOGIC SETTING

1. Geological Setting of the Ruselfa Area

The phoshpate in Ruseifa belongs to the Campanian-Maestrichtian age. It is overlain by the Chalk Marl Unit of Maestrichtian-Danian age, and underlain by the Silicified Limestone Unit of Campanian age. The thickness of the Phosphorite Unit ranges between 15 and 35m. A generalized section from Ruseifa is shown in Fig. 2. The section shows five phosphate beds. The lowest one has not been mined. The other four (numbers I to IV) vary between soft and consolidated phosphates. The phosphate beds are folded and faulted. The axes of the anticlines plunge towards the northwest.

2. Geological Setting of the Maqarin Area

The rock units are of Maestrichtain-Danian-Paleoence age and are equivalent to the Chalk Marl Unit. The thickness reaches 200m. The Maqarin area represents a SW-NE striking anticline which plunges with minor synclines trending NW-SF. Tensional faults of Neogene age trending NW-SE are also recorded (Wiesemann and Abdullatif, 1963). The exposed parts of the unit is shown in a generalized section in Fig. 3.

The section shows two major formations. The lowest is the bituminous limestone which is overlain by chalky limestone with chert intercalations. A metamorphosed zone occurs in the chalk limestone beds and is restricted to the lower and middle parts (Khoury and Nassir, 1982). The whole sequence is covered by Pleistocene basalt flows and soils (Wiesemann and Abdullatif, 1983).

SAMPLING TECHNIQUES

Eleven rock samples were chosen from the outcrops of the bituminous limestone in the Maqarin area along the southern slope of the Yarmouk River. Sixteen rock samples were also chosen from the four major phosphate beds in Ruseifa. Different types of water samples were also collected from both areas. Twenty-one groundwater locations in Ruseifa were sampled twice, in June 1981 and in March 1982. Nine locations within the phosphate deposit were chosen, in areas where water circulates through the phosphate rocks before it reaches the groundwater system. The other locations were from the surrounding area where no phosphate rocks are present. More than one hundred water samples were also collected and studied by Abdel-Jaber (1982).

ANALYTICAL TECHNIQUES

The rock samples were analysed for their mineral content using petrographical and X-ray diffraction techniques. Rock and water samples were also analysed for their trace metal content. The analyses were carried out in the Department of Geology and Mineralogy at the University of Jordan.

PETROGRAPHY AND MINERALOGY

Ruseifa Phosphates

Petrographic studies revealed the presence of different types of phospate rocks. These include cement-free phosphates, friable, calcareous phosphates, siliceous

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phosphates, calcareous phosphates, calcareous argillaceous phosphates, and argillaceous phosphates. Phosphates also occur in the from of fossil teeth and bones and in the form of pellets of different sizes (1 mm to 2 cm).

X-ray diffraction techniques confirmed the presence of fluorapatite $Ca_{1,n}(PO_4, CO_4)_{h} F_2$ and carbonate fluorapatite as the essential constituents. Other common minerals are calcite, quartz, mixed-layer lilite, and smectite of high expandability. Secondary uranium and vanadium minerals of metaautunite and metatyuyamunite are also present, filling fractures, and in the form of encrustations.

The tricalcium phosphate content in the deposits is variable, reaching a maximum (70%) in the soft phosphates. Other constituents are $CaCo_3(\sim 20^{a_0})$ and $SiO_3(\sim 8\%)$. Most of the phosphorus occurs in apatite $Ca_{10}(PO_4, CO_3)_6 F_2$. The structure of apatite is such that small amounts of VO_4 , As_3O_4 , and SO_4 a substitute for PO_4 whereas Na, Sr, U, Th, and the rare earths may substitute for Ca. Trace element compositions of the phosphate samples are shown in Table 1.

Magarin Bituminous Limestone.

Thin section study revealed that all the bituminous limestone is essentially biomicritic. The fossil remnants are essentially foraminiferal and comprise up to 40% of the constituents. Micritic calcite and organic matter reach 80%. Pyrite, quartz, hematite, and clays occur as accessory minerals. X-ray diffraction studies of the bituminous limestone indicate the presence of calcite as an essential constituent. Apatite, quartz, dolomite, kaolinite, mixed layer clays, illite, and gypsum occur in different proportions.

A complex series of minerals characterize the mineralized zone. Iwo assemblages are present (Khoury and Nassir, 1982):

- a. High temperature mineral assemblages which include recrystallized calcite, flourapatite, spurrite, diopside, wollastonite, and anorthite.
- b. Low temperature mineral assemblages which include tobermorite, afwillite, ettringite, barite, thaumasite, gypsum, and portlandite.

Trace elements are essentially concentrated and adsorbed in the organic matter of the bituminous limestone. The low temperature mineral assemblage is considered a good host for the trace elements. (Khoury and Nassir, 1982). The trace element composition of the rock samples from Magarin area is shown in Table 2.

HYDROLOGICAL CONDITION OF THE TWO AREAS

Ruselfa Area

The average precipitation in this area is 300 mm/yr. A relatively high portion of the precipitation infiltrates and escapes evaporation because of the highly fractured nature of the outcropping rocks. The Zarqa river, which drains the area west of Ruselfa, receives groundwater from the surrounding aquifers (Salameh, 1980). The Zarqa river in the Ruselfa area flows in the Massive Limestone, Silicified Limestone, and Phosphorite Units, which form the upper aquifer. The river in this portion is effluent in winter, and because of pumping, in summer it becomes in-

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fluent. The lower aquifer, locally designated as (A4), belongs to the Echinoidal Limestone Unit and is separated from the upper aquifer by a chalky marly sequence. Both aquifers are connected along the Zarqa river, which flows parallel to the Amman flexure in the southeast (Salameh, 1980).

Magarin Area

The average annual preciptation in the area is 450 mm/yr. The average annual air temperature is 20°C. Two main groundwater aquifers are present in the area (Abdel-Jaber, 1982). The first is a shallow, perched aquifer system in the Chalky Limestone Unit and is characterized by a temperature of less than 20°C. The second is a deeper aquifer system in the bituminous marl and is characterized by average temperatures of 27-28°C. Rainfall recharge is relatively limited due the semiaquiclude nature of the outcropping rocks (Salameh, 1980). Highly alkaline (pH=12.5) meteoric waters of calcium hydroxide type issue from the bituminous marl (Khoury and Nassir, 1982). Details of the description of the aquifers, their properties, and water behavior data in recharge and discharge are discussed by Salameh (1980) and Abdel-Jaber (1982).

HYDROCHEMICAL RESULTS

Ruselfa Area

The chemical analyses on water samples from the upper and lower aquifers indicated that the upper aquifer is an alkaline earth water type with prevailing bircabonate and chloride (Abdel-Jaber, 1982). The lower aquifer waters are of alkaline type with prevailing chloride. The T D S of both aquifers ranges from 500 to 650 ppm (Abdelnoor and Salameh, 1982).

The trace metal concentration of waters circulating through the phosphate rocks and from the surroundings is shown in Tables 3_a and 3_b . It is clear that the trace metal concentration increases manifold in the case of waters circulating through the phosphate rocks. Mo and V, for example, are below the detection limit in samples from the surrounding areas where waters circulate through non-phosphatic rocks. However the water which circulate in the phosphatic rocks show an average concentration of 14.7 ppb for Mo and 8.6 ppb for V. The circulating water passes through the phosphatic rocks to the main aquifer below, carrying the leached base metals.

Magarin Area

Different types of water occur in this area. An unusual highly alkaline water with a pH above 12.5 and a normal pH water (pH = 6.8) are very characteristic to the area (Khoury and Nassir, 1982). The high pH water is of alkaline earth type with prevailing hydroxides. The major cation is calcium (400 ppm); the major anion is hydroxide (300 ppm); Magnesium and bicarbonate are not detected. Sulfate anions (270 ppm) are second to the hydroxides (Abdel-Jaber, 1982; Khoury and Nassir; 1982, Barnes et al, 1982). The T D S of the water ranges between 500 and 1000 ppm. The trace element composition of the highly alkaline water is shown in Table 4a. The high pH water is characterized by its high Cr, Cd, Ni, Pb, Mo, and Co concentrations, which are 365, 8, 48, 65, 44, and 42 ppb respectively.

The normal pH waters issue from springs in the Chalk Marl Unit. The water is of alkaline earth type with prevailing bicarbonate. The TDS content is below 600 ppm

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(Abdel-Jaber, 1982). The trace metal composition of the normal pH water is shown in Table 4b. It is clear that the trace metal concentrations are low and do not exceed 10 ppb, except in the case of Pb and Ni where it reaches an average concentration of 22 and 16 ppb respectively.

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DISCUSSION

Comparison between Tables 3a and 3b indicates a clear increase in the trace metal concentration in the waters circulating in the phosphate rocks. This is a reflection of leaching of these rocks, since waters that circulate in non-phosphatic rocks show a low concentration of trace metals. Leaching of phosphate rocks containing most of the trace elements in the structure of apatite is indicated by the presence of secondary uranium and vanadium minerals (metaautunite and metatyuyamunite). Table 1 shows the trace element concentration in the phosphatic rocks. The circulating waters in the phosphatic rocks extract the trace elements from the apatite structure which is stable under alkaline conditions. The circulating waters in the non-phosphatic rocks shows normal values of trace element concentration.

The groundwater in the phosphates forms a lens-like body of chemically different water within the groundwater of the upper aquifer. The phosphate groundwater used to discharge into the Zarqa river. For some years, however, the groundwater level in the upper aquifer has been lowered by several meters as a result of overexploitation. This has caused the lens-like phosphate water to sink below the level of the Zarqa river. Accordingly, the phosphate water has joined the groundwater flow system, and the Zarqa river, which formerly played the role of an effluent stream, started to become partly influent in these reaches.

It is possible that the phosphate water, with its high trace metal content, will contaminate the waters extracted from wells in the surrounding areas.

Comparison between Tables 4a and 4b indicates that the normal pH water is not affected, since it circulates within the Chalk Marl Unit. The bituminous limestone beds do not affect these waters. The hydrochemistry and trace metal concentration are normal. The slightly high values shown in Table 4b for some trace metals such as Ni and Pb could be attributed to the mixing effect with the high pH waters. The high pH waters indicate intensive leaching from the bituminous and chalk limestone beds. The trace metal content is unusually high, and the water is unique in its calclum hydroxide nature. Table 2 shows that the bituminous limestone is enriched with trace metals which are absorbed essentially by the organic matter (Amerih, 1979). The formation of highly alkaline waters could be attributed to the presence of a high temperature mineralization zone in the Chalk Marl Unit. The high temperature mineralization can be explained by the spontaneous combustion model (Khoury and Nassir, 1982; Barnes et al, 1982). Late volcanic activity could have initiated combustion (Khoury & Nassir, 1982). Calcination of limestone leads to the formation of lime. Hydration of the lime would lead to the formation of portlandite, Ca (OH), which is highly soluble, and is responsible for the formation of highly alkaline waters. The circulation of the groundwater in the high temperature zone thus yields high pH waters. The combustion of the organic matter would liberate the heavy metals which could easily be concentrated within the high pH waters. The water which circulates through non-affected (non-mineralized) areas gives the normal pH

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water. Leaching without spontaneous combustion of the organic matter is not effective.

CONCLUSIONS AND RECOMMENDATIONS

- Leaching of the phosphatic rocks in Ruseifa area is responsible for high trace element concentration in the groundwaters underlying the phosphate deposits. This water is not recommended for drinking purposes, in accordance with WHO standards.
- 2. Leaching of the high temperature mineralization zone in the bituminous limestone is responsible for the formation of high alkaline water enriched with trace elements. The sedimentary rocks in the Chalk Marl Unit, including the non-mineralized bituminous limestone, yield unaffected water with normal pH and normal trace metal contents. Mixing with high pH waters give some high trace element values.
 - Future research is recommended to study all steps of dissolution processes in the phosphates and bituminous rocks.

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Chemical composition of some phosphate rocks of Ruseifa.*

mole	Fe %	Cr %	К%	Co %	Mo %	Sr %	V %	Ni %	Cd %	Cu %	Zn % 1		
1 mple 0.	Fe % 1.2803 1.3144 1.1151 4.2673 0.1833 1.4009 0.9735 0.9813 0.8738	Cr % 0.0748 0.1172 0.1122 0.1721 0.0748 0.1072 0.1222 0.1322 3 0.1397	0.200 0.213 0.333 0.300 0.133 0.300 0.217 0.200 0.133 0.600	0.1057 0.0857 0.1371 0.1086 0.0971 0.0714 0.0856 0.0914 0.1657 0.0714	0.1295 n.d. 0.0741 0.0741 0.0741 0.0926 - 0.1111 0.1111 0.2963	7.684 7.937 10.266 8.797 8.354 6.215 8.481 8.734 7.962 3 6.722	0.023 0.025 0.036 0.025 0.018 0.027 0.021 0.018 0.09 0.021	0.1172 0.1897 0.200 0.1931 0.0862 0.1586 0.1069 0.1207 0.1966 0.1345	0.0838 0.400 0.1714 0.0333 0.0524 0.1019 0.0276 0.0914 0.0829 0.1048 0.1086	0.092 0.019 0.023 0.034 0.037 0.025 0.025 0.026 0.039 0.028 0.030 0.036	0.4190 0.4266 0.4017 1.1596 0.5780 0.4669 0.6314 1.1249 0.7446 0.6266 1.0132	2.818 2.182 2.818 3.182 5.000 3.909 3.273 3.091 4.182 4.182 3.273	0.0207 0.0405 0.0414 0.0486 0.0387 0.0477 0.0135 0.0171 0.0207 0.0189 0.0531
0 1 2 13 14	1.4534 0.8109 1.091 0.7555 3.754 1.199	4 0.0898 9 0.1471 5 0.1740 8 0.221 4 0.212 0 0.234	0.600 0.233 0.133 0.100 0.100 0.400 4 0.167	0.0714 0.0800 0.0714 0.2428 0.097 0.065	0.074 0.0920 0.203 0.148 7 0.111 3 0.129	9.557 6 10.316 7 10.481 1 8.671 8.228 6 7.911	0.023 0.032 0.021 0.018 0.041 0.027	0.1724 0.759 0.1482 0.2069 0.2069 0.179	0.1086 0.0248 0.0238 0.100 0.085 3 0.2470	0.036 0.036 0.044 0.042 7 0.027 5 0.033	1.0132 1.1319 1.0257 1.0902 1.0972 0.9535	3.727 3.545 3.00 3.636 3.545	0.0081 0.0081 0.0261 0.0153 0.0126

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Samle	Co %	Cu %	Fe %	Pb %	Zn %	Cd %	Mn ∿	
2 3 4 5 6 6 7 8 9 10 11	0.0045 0.0056 0.0061 0.0086 0.0122 0.0041 0.0086 0.0097 0.0055 0.0050	0.0015 0.0015 0.0014 0.0012 0.0017 0.0009 0.0020 0.0021 0.0007 0.0001	0.9136 0.6780 0.8245 0.4307 0.6173 0.1350 0.4409 0.4080 7 0.3689 5 0.3170	0.0028 0.0076 0.0023 0.0125 0.0125 0.0028 0.0032 0.0016 0.0036 0.0028	0.0599 0.0624 0.1594 0.0569 0.1053 n.d. n.d. 0.0690 0.0323 3 0.0218	n.d. 0.0012 0.003 0.0012 0.0024 0.0011 0.0011 0.0005 n.d. n.d.	0.0075 0.0062 0.0294 0.0084 0.0060 0.0076 0.0074 0.0067 0.0073 0.0043	0.0193 0.0205 0.0189 0.0204 0.0238 0.0074 4.0.0209 7.0.0208 3.0.0156 3.0.0120

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Table 2: Trace elements content of some bituminous limestone samples from Maqarin.

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Analysis was carried out at the Department of Geology and Mineralogy, University of Jordan.

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Table 3a: Trace metal composition of water samples in contact with phosphate rocks (ppb)*. 21.

Analysis was carried out at the Department of Geology and Mineralogy, University of Joradn.

Location	Mn	Fe	Zn	Cd	Мо	Co	v	Ni	Cr	Pb	Cu	*	
Ruseifa well 18	7.3	52.0	13.4	9.0	5.7	9	3.8	18	00	38.6	10		
Ruseifa spring	10.8	70.5	44.9	9.3	7.6	21	7.9	21	3.0	37.6	8		
Ruseifa well 1	7.3	48.0	68.5	5.3	n.d.	25	n.d.	32	10	30.4	10		
Ruseifa well 2	7.3	50.3	61.0	5.5	n.d.	18	n.d.	31	10	44.8	10		
Ruteimh well	7.8	61.1	31.7	11.1	12.8	7	5.7	28	2	29.4	44		
Mititary School	6.0	152.6	147.7	7.4	13.2	31	6.3	37	12	34.5	20		
Zarqa well 16	28.3	660.7	119.9	14.5	26.3	13	17.2	98	18	99.4	22	10	
Zarqa well 14	25.6	149.4	65.8	10.1	21.1	23	11.1	57	18	45.8	11		
Zarqa well 14a	23.2	203.0	253.7	9.9	16.3	13	9.3	55	00	42.8	34		
Average	13.74	160.8	89.5	9.1	14.7	17.8	8.6	41.9	8.1	44.9	18.9		

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*

n.d. = not detected.

Location	Mn	Fe	Zn	Cd	Ni	Cr	Pb	Cu		
Tabarbor	5.2	41.2	23.7	3.9	3	2.2	0	21.1	21	
Culton 1	1	146	14	3	8	20	4	18	2	
Sukina 1	0.5	98	10	3.2	2	16	2	17	2	
Sukinia 4	1	181	11	3	4	18	2	14	2	
Ain Gnazai	2	437	23	3.3	7	5	8	19	7	
Ain Einimra	1	128	14	1.45	5	3	3	17	2	
Ras EIAIN	2	6.8	60	2	n.d.	21	n.d.	24	18	
Wadi ESSIT	5	119	18	3 7	12	12.3	2	n.d.	3	
Hisban	0	110	10	2.6	8	21	5	n.d.	5	
Um El Dannanii	4	148	14	1.6	11	12	4	n.d.	5	
Bahhath	n.d.	96	14	6.0	nd	21	3	29	n.d.	
Kheil	n.d.	212	18	0.0	7	31	2	22	8	
Naur	4	138	17	2	1	31	•			
Average	2.8	152	21.3	2.93	6.7	22.1	4.38	20.1	5.7	
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Table 3b: Trace metal composition of water samples in non-phosphate rocks (ppb)*

Analysis was carried out at the Department of Geology and Mineralogy, University of Jordan.
n.d. = not detected.

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Sampling	pH	TDS	Cu ppb	Mn ppb	Cr ppb	Ni ppb	Zn ppb	Pb ppb	FE ppb	Co ppb	Cd ppb	Mo ppb
site			7.40	6.24	479	25.2	0.03	43.9	117	44.3	6.11	31.5
Adit A6	12.5	1220	1.49	0.24	147	1999910101						76.0
(6 Samples)	12.5	1488	10.40	6.62	471	34.8	0.06	6.38	110	40.4	7.96	30.0
(5 Samples)	12.5	2450	24.92	16.72	292	70.5	0.11	81.5	137	44.3	9.16	59.3
(2 samples)	12.5	1572	14.88	16.18	256	59.7	0.10	69.2	96	36.8	9.76	46.4
AFS	14.5								110	11 5	8 75	43.5
Average	12.5	1683	14.4	11.44	364.5	47.6	0.08	64.6	115	41.5	0.25	

* Analysis was carried out at the Department of Geology and Mineralogy. University of Jordan.

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Table 4b: Trace Metal Composition of the Spring Water Magarin area*.

Location	pH	T.D.S	Cu	Mn	Cr	Ni	Zn	Pb	Fe	Co	Cd	
			ppm	ppb	ppb	ppb	ppb	ppm	ppb	ppb	ppb	ppb
Spring No. 4	6.9	746.55	5.32	17.81	7.22	15.65	0.01	18.60	0.063	9.60	2.34	
No. 9	6.8	573.02	5.53	6.30	3.44	12.99	0.02	21.70	0.123	13.29	1.64	
No. 2	6.8	484.69	4.68	20.27	3.44	13.65	0.02	n.d.	0.076	5.91	2.44	
Spring No. 3a	6.8	479.36	5.96	5.21	7.22	10.99	0.03	14.03	0.116	6.65	n.d.	
Spring No. 3b	6.8	326.85	3.39	7.22	15.57	8.97	0.03	12.31	0.396	10.13	1.55	
Quseir	8.5	589.86	13.65	25.02	3.09	15.55	0.09	23.81	0.371	n.d.	1.51	
Ghazalah	6.8	247.74	2.58	4.83	7.20	8.11	0.02	15.87	0.180	n.d.	1.00	
No. 7	6.8	450.76	6.46	3.78	6.18	29.65	0.03	19.16	0.086	9.94	1.79	
Ba'boul	6.8	606.40	8.07	25.08	19.27	7.41	0.02	27.6-	0.196	n.d.	1.79	
Sombal	6.8	444.34	6.68	4.85	4.00	31.17	0.04	17.03	0.080	n.d.	2.93	
Et Trab	6.8	502.86	6.93	5.12	2.74	12.17	0.02	12.27	0.052	5.94	0.97	
Quelbe	6.8	258.68	8.89	9.01	4.31	19.61	1.59	55.91	0.122	12.46	2.17	
Murdahia	6.8	796.57	8.67	20.23	12.00	19.77	0.03	24.48	0.266	18.41	2.28	
Average	6.8	500.6	6.69	11.9	7.14	15.8	0.03	21.9	0.163	10.25	1.9	

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* Analysis was carried out at the Department of Geology and Mineralogy, University of Jordan.

n.d. = not detected.

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