



Elemental Fractionation and Quantification of Metals from Uneaten Parts of Date Fruits (Phoenix Dactylifera L.)



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Abstract

Unconsumed parts of fruits can be a source of inorganic and organic elemental supplies to the biosphere, the geosphere and/or the hydrosphere that may induce changes in the chemical characteristics of the environment. The purpose of the present study is an investigation of fractionation of major and trace elements including the rare-earth elements (REEs) in the skin, the soft part, and the seed of the date fruit to evaluate the elemental amounts in uneaten parts of this fruit. The date seeds are concentrated in Fe, Ni, P, Zn, Mn, Co and REEs. Their soft parts can be sources of K, Sr, Rb, Zr, Ca and Mg, and the skin concentrates Cu, Si, Al, Cd, Ba, P, Th and Pb. About 40% of Si, 79% of Al, 26% of Mg and Ca, 21% of Fe, 30% of Mn, 15% of Ti and 24% of Na from whole fruit are located in the skin. The REEs are depleted in the soft part relative to the other ones. The seed skin has an ability to accumulate the light REEs (LREEs) relative to the middle REEs (MREEs) and heavy REEs (HREEs); no fractionation being visible in the soft part. The non-consumable parts of dates, such as the seeds and the skin may contribute to an REE enrichment of the ecosystem. About 0.14 to 1.1 mg of REEs can be extracted yearly per capita from uneaten date seeds with an overall interest in industrial applications and production of energy since availability of REEs is limited.

Keywords: Elemental fractionation; Trace; Rare-earth elements; Seed; Soft; Skin anomalies.

Introduction

To maintain life, a plant body possesses a variety of tissues and organs capable of division, differentiation and growth along defined processes. Food constituted of sugar, proteins (enzymes) and lipids manufactured by some tissues is transferred to other tissues for storage and use for the growth process with developments of organs with distinctive functions. For most plants with diversified organs (roots, stems, leaves, flowers, fruits and seeds), the functions are diversified too. For example, the main function of fruits is to protect seeds during the development and to attract birds, bees and other animals to eat the seeds and/or to carry them into new areas. During the growth of plants and the development of their fruits, three distinct parts develop: the exocarp (the outermost part), the mesocarp (that supports the fleshy middle part), and the endocarp (the innermost part with stony seed) [1]. The literature is replete with information on morphological and physiological constructs of a wide variety of fruits, each at different life stage from a nascent growth state to a mature or ripened state [2-5]. Organic compounds that constitute the various components of a fruit have also been extensively investigated [6-8]. Such organic compounds are basically made of

six essential elements that include C, H, N, O, P, and S and they constitute nearly 95% of the biomass on average.

Very significantly, all life forms also require various amounts of environmentally common inorganic ions such as K, Ca, Mg, Na, Si, and others, as well as traces including Fe, Mn, Co, Zn, Cu, Mo, and few others. Fruits are a main source of these major and trace elements that are involved in human physiology. As for human physiology, the main role of metals in plant growths is their effect on enzyme activity, which is primordial for activation of cellular processes and growth of plants. Metals are activators of many basic processes: assimilation and decomposition (polysaccharids to monosaccharids) for growth of fruits. Several previous studies have been reported on major elements chemistry of whole fruits [9-13]. However, little information is available on metals contents and their fractionation in various parts of the fruits.

Most of fruits and vegetables are not entirely consumed by humans [14] reported that many fruits and vegetables can generate up to 25% to 30% of waste materials, which are not further used [15] reported that apples, for instance, generate 10.9% of seeds and pulp as by-products, and 89.1% of final products during slicing, Papaya produces about 8.5% of peel waste, 6.5% of seeds, 32% of

unusable pulp and the peeling of mandarins generates about 16% of peels and 84% of finished product. The unconsumed parts such as seeds or skins may contain nutrients and organic molecules [16] and may lead to accumulation of nutrients in soils and oceans. These nutrients supplied to the biosphere, the geosphere and/or hydrosphere may induce changes in the chemical characteristics of the environment such as the chemical composition of surface waters, which may have a major impact on the natural systems linked to surface processes [17] showed that river chemistry is influenced in a major way by plant kingdom.

The present study is an investigation of both major and trace element data from different parts of a fruit; the selected fruit for this study being the date *Phoenix dactylifera* L since it is a very consumed fruit in many countries. Dates belong to fruits with a hardened endocarp called drupes. The hardened endocarp provides a physical barrier around the seed protecting it from disease and herbivory [18]. The date fruit is composed of a pericarp, mesocarp, endocarp and seed. The mesocarp represents the most part of the fruit pulp. The study is focused on the differentiation of a number of inorganic ions in edible, mature fruits (dates) and drupes of palm plants. Although this study does not specifically try to connect the array of distributions of the inorganic ions to their assimilation into organic macromolecules of various parts of ripened date fruits, the information has been gathered to capture information about the elemental roles in potential carbohydrate-fat relationships of seeds, which seem to indicate that fat contents in fat-storing seeds increase while carbohydrate contents decrease as seeds mature. The inorganic influences may be viewed from some other perspectives, such as their supplies from their ultimate root zone environments and net

growth of cell populations at ripened stages.

Material and Methods

For the present study, we took dates produced in local agricultural farms (Sultanate of Oman) from supermarkets. The different parts of the date fruit were separated into epicarp, mesocarp, seed skin and seed. These samples were washed twice with distilled water and with distilled-deionized water. The samples were wiped with clean tissue paper and cut into small pieces with a stainless-steel knife. The seeds were isolated and washed, dried and grounded into a fine powder. The freeze-dried fruits were digested using HNO_3 and HClO_4 .

The elemental contents of the different samples were determined on an inductively coupled plasma atomic emission spectrometer (ICP-AES of the Laboratoire d'Hydrologie et de Géochimie de la Surface de Strasbourg, France) for the major (Si, Al, Mg, Ca, Na, K and P) and some trace (Sr, Fe, Mn and Zn) elements, and on an inductively coupled plasma mass spectrometer (ICP-MS of the same laboratory) for the other trace elements (V, Cu, Ba, Cr, Co, Ni, Rb, As, Zr, Cd, U and Th) and the rare-earth elements (REEs). Repeated analysis of international standards on a weekly basis provided an analytical precision of +2.5% for the major elements, +5% for the trace elements and +10% for the REEs, on the basis of the analytical procedure [19].

Results

Concentrations of major and trace elements including rare earth elements (REEs) in the skin, soft part, seed skin and the seed of the date fruits are stored in Table 1.

Table 1: Concentration of major (in ppm) and trace elements (in ppb) in different parts of *Phoenix dactylifera* L.

ppm	Skin	Soft part	Seed skin	Seed
Si	68.412	58.772	23.619	22.829
Al	1922.747	81.680	278.535	144.481
Mg	1072.961	1477.199	694.758	987.433
Ca	944.206	1461.400	837.343	428.309
Fe	73.433	90.844	104.273	85.314
Mn	9.202	5.111	3.989	11.976
Ti	16.481	19.275	49.830	24.654
Na	1038.627	1042.729	1161.221	1129.623
K	6516.738	13034.111	6551.813	5639.425
P	1253.219	932.136	352.237	1414.004
ppb				
Sr	3714.163	15956.912	9637.343	1907.720
Zn	14051.502	11201.436	13563.375	14922.083
Cu	5579.399	4660.682	3475.763	3712.747
V	2635.193	473.968	1311.311	1050.628
Cr	1587.983	1816.876	3104.488	2606.822
Co	32.618	22.118	24.488	36.338
Ni	1424.893	892.639	1311.311	2030.162

Rb	7932.189	12086.176	4848.689	5015.368
Y	14.249	4.187	10.585	13.824
Zr	506.438	1161.221	725.961	327.828
Cd	8.841	6.241	6.952	6.004
Ba	830.901	727.540	786.786	688.043
La	32.017	10.269	32.309	30.176
Ce	85.665	37.364	97.163	103.878
Pr	6.266	1.817	6.478	7.425
Nd	23.863	6.241	21.724	26.384
Sm	4.292	1.027	3.555	4.740
Eu	0.858	0.158	0.790	1.027
Gd	4.120	1.027	3.397	4.582
Tb	0.515	0.158	0.395	0.632
Dy	3.004	0.711	2.133	3.239
Ho	0.515	0.158	0.395	0.711
Er	1.717	0.553	1.422	1.975
Tm	0.172	0.010	0.158	0.237
Yb	1.545	0.553	1.264	1.738
Lu	0.258	0.079	0.158	0.237
Pb	318.283	82.154	252.230	143.691
Th	15.365	4.503	5.135	6.478
U	5.923	1.343	5.846	3.713
TREEs	164.807	60.125	171.339	186.980

Major elements: Na, K, Mg, Ca, Al, Si, P, Ti

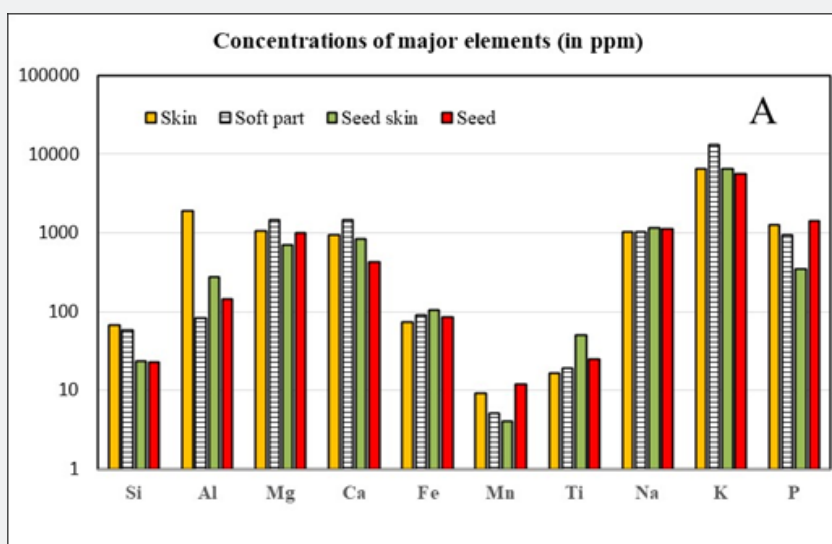


Figure1a: Concentration of major elements (in ppm) in different parts of Phoenix dactylifera L.

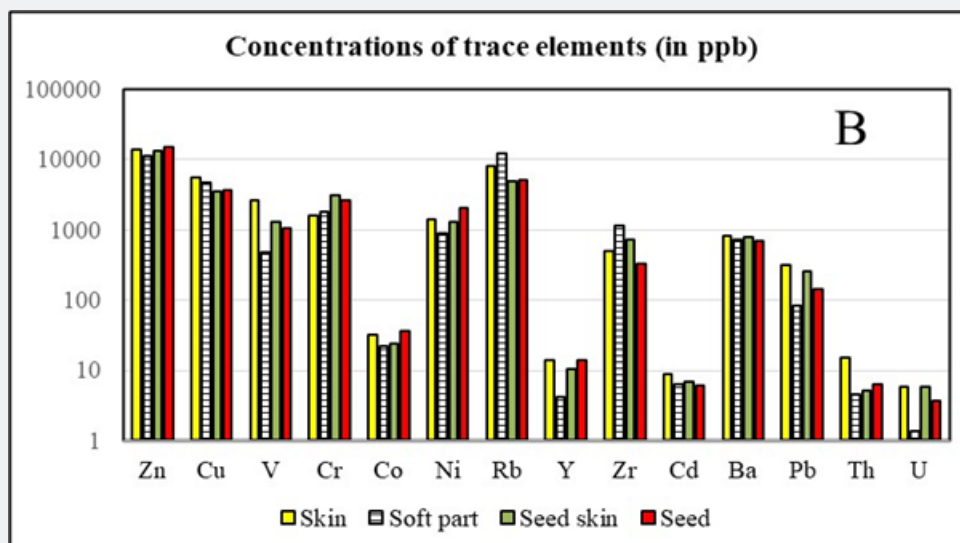


Figure1b: Concentration of trace elements (in ppb) in different parts of Phoenix dactylifera L..

The concentration of Na ranges from 1039 to 1161.2 ppm (average =1081 ppm), while that of Si ranges from 24 to 68 ppm (average = 23 ppm) and that of Al from 82 to 1923 ppm (average = 761 ppm). The concentration of Mg ranges from 695 to 1477 ppm (average = 1082), and that of Ca from 837 to 1461 ppm (average = 1081 ppm). That of Fe ranges from 91 to 104 ppm (average = 90 ppm), while that of K ranges from 6517 to 13034 ppm (average = 8701 ppm) and that of P from 352 to 1253 ppm (average = 846

ppm). The elemental distribution in the different parts of dates, including the skin, the soft part, the seed skin and the seeds range in the following order: for Si, skin>soft> seed skin=seed, for Al: skin>seed>soft, for Mg, soft>skin=seed>skin seed, for Fe: similar in these different parts, for Mn: seed >skin>soft>skin seed, for Ti: seed skin>seed>skin=soft; for Na, similar distribution in all parts, for K: soft>skin=seed=seed skin; and for P: seed>skin>soft>seed skin (Figure 1).

Transition metals

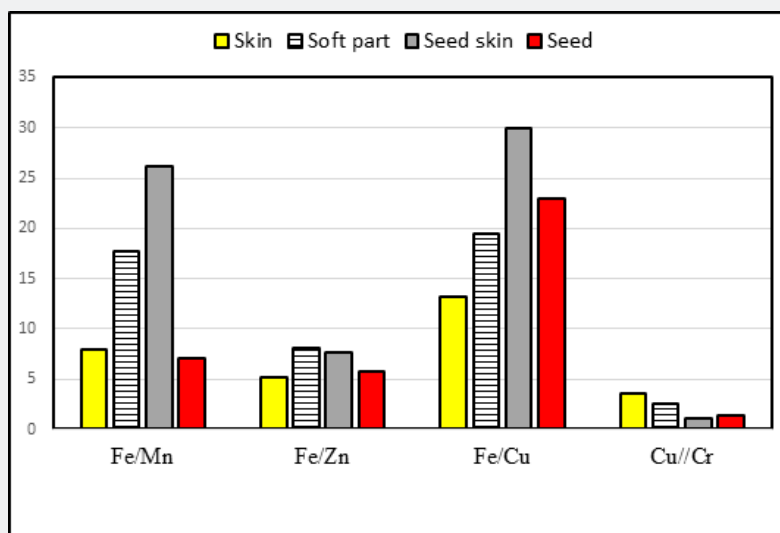


Figure2: Elemental ratios of trace elemental contents from different parts of Phoenix dactylifera L..

The total content of the metals Ni, Cu, V, Cr, Co, Zn, Mn, Fe, Ti and Zr is about 125 ppm in the skin, 135 ppm in the soft part, 147 ppm in the seed and 182 ppm in the seed skin. The Cu concentrations range from 3.7 to 5.6 ppm (average n= 4.4 ppm) and those of Zn range from 11.2 to 14.9 ppm (average = 13.4 ppm). Large differences can be seen for the different parts of the

date fruit (Table 1, Figure 2). For Sr: soft>seed skin>skin>seed; for Zn: seed>skin>soft>seed; for Cu: skin>soft>seed=seed skin; for V: skin>skin seed>seed>soft; for Cr: seed skin>seed>skin=soft, for Co: seed=skin>soft=skin seed; for Ni: seed=skin>soft=skin seed; for Rb: soft>skin>seed skin=seed; for Zr: soft>seed skin>skin>seed; for Cd: skin>skin seed>soft=seed; for Ba: similar contents; for Pb:

skin>seed skin>seed>soft; for Th: skin>seed>soft=seed skin ; for U: skin=seed skin>seed>soft. Among the different organs, the skin is especially enriched in Cu, Cd, Ba, Pb, Th and U. The depleted remaining elements are Sr and Cr.

Rare-earth elements

The total contents of REEs are about 164.8 ppb in the skin, 60.1 ppb in the soft part, 171.3 ppb in the skin of the seeds and 187.0 in the seeds. The contents are higher in the skin than in the soft part and higher in the seeds than in the skin of the seeds. Skins have a tendency to accumulate REEs in the opposite to the soft part.

Discussion

Distribution of elements between the skin, soft part, seed skin and the seed of the studied dates

Compared to results of [20], the date seeds yield similar ratios than the date fruit (about 6.5 to 11.3). Our results on Fe content fell within the range of 0.3–10.4 mg/100 g that were reported previously for date varieties from different countries. As stated above, K yields the highest concentrations among the different elements in the different parts of the date fruit. Similar results were observed by [21] for date seeds of 3 varieties of Moroccan dates (about 4153.3 ppm for seeds of the Boufgous Moroccan variety of dates). Another variety of Moroccan dates contains about 4 times more K than in the seed of the Dates studied here [21]. Beside K, the content of Ca, Mg, P and Na in the soft parts of the dates are also higher than the other elements. In the seed skin, Na is the second most concentrated element after K with significant concentrations compared to the other elements. In the seed, the abundant elements beside K, are P, Na, and Mg. Similar ranges were observed for Mg and Ca content in seeds of Omani dates, which are investigated here, compared to Moroccan dates [21]. The content of Na in seeds of the present study is higher than in the Moroccan dates.

Relative to previous studies on fruits, there is more Na than in the seeds of Baobab (*Adansonia digitata*) [22] but less K, Ca, Mg and Fe. Similar amounts of K and Fe were observed in the soft parts of the fruits from both studies. Compared to data of lupine seeds [23] there is more Fe and Zn and Cu, and less Mn in date seeds investigated during this study. Potassium has many functions in plants and fruits, such as stomatal regulation of transpiration and photosynthesis, enzyme activation and protein synthesis. It is also involved in photo-phosphorylation, transportation of photo assimilates from source tissues via the phloem to sink tissues [24–27].

Major elements

Comparison of elemental contents in the different organs shows that the most enriched elements in the date skin are Si, Al and P and the most depleted are Fe, Ti and Na. There is more Al and Si in the skin compared to the other parts, which suggests an

external supply of these elements probably by dust particles from atmosphere. Aluminum could be also due to internal distribution from roots being selectively transferred to the skin. The most enriched elements in the seed itself are Mn, K and P and the most depleted are Si and Ca, while the most enriched elements in the skin of the seeds are Fe, Ti, K, Al and Na and the most depleted are Si, Mg, Mn and P. The most enriched elements in the soft part of the dates are Mg, Ca and K and the most depleted are Al, Mn and P. The elements such as Ca and Mg are provided more from the soft part than from the other parts.

Trace elements

Most of the heavy metals are cofactors for several enzymes. In particular, Ni and Zn are present in a number of enzymes in plants and microorganisms and are used as co-factors for enzymes [28–36]. On the other hand, the abundance of elements in plants and fruits reflect their mobility from roots to the parts above ground. Inputs from atmosphere may be a secondary source of elements in plants. The release of elements in the rhizosphere and their transportation to stems and leaves or to fruits require enzymes with specific sites for specific metals.

Among the different compartments of the date fruit, the most enriched elements of the soft part are Sr, Rb and Zr, while V, Co, Ni, Pb, Th and U are the most depleted ones. The most enriched element in the skin of the seeds is Cr and the most depleted is Rb. The most enriched elements in the seeds are Zn and Ni and the most depleted are Sr, Zr, Cd and Ba. Compared to contents of Cu and Zn already analyzed in dates by [37], the different parts of the dates here have higher Cu and Zn contents, which is in the opposite to lower Pb in the analyzed date samples of this study. The distribution of Zn and Co are similar with higher concentration in the seed and less in the skin, Th and U having the same distribution. Elements such as Fe and Cr are more concentrated in the seed skin and less in the skin, which suggests a low mobility from the seed to the other parts. Iron may occur under ferrous and ferric forms; the ferrous form being more mobile than the ferric one. Such distribution suggests more oxidized conditions in the seed than in the other parts of the dates. The oxidized form of Cr is more mobile than the reduced one.

Fractionation of Cr, Mn, and Co more abundant in the seed indicates oxidized conditions, as well as in its skin. In the opposite, higher concentration of U, Th and Cu in the skin relative to the other parts suggests external sources of these elements. Similar results were reported by [38] for Cu and Pb, more abundant in the skin than in the pulp of peach fruits. For plums and marrows, the trace elements tested were higher in the pulp than in the skin. For both the K/Rb and Sr/Ca ratios (Table 2), the highest values are in the seed skin and the lowest are calculated for the skin (Figure 3). The K/Rb values reflect an organic effect more than an inorganic one. The lowest K/Rb ratio observed in the skin indicates an impact of silicate inputs, probably as atmospheric dust.

Table 2: Elemental ratios in different parts of *Phoenix dactylifera* L.

	Skin	Soft part	Seed skin	Seed
Fe/P	0.059	0.097	0.296	0.060
K/Rb	821.56	1078.43	1351.25	1124.43
Sr/Ca 10 ⁻³	3.93	10.92	11.51	4.45
U/Th	0.39	0.30	1.14	0.57
Fe/Mn	7.98	17.77	26.14	7.12
Fe/Zn	5.23	8.11	7.69	5.72
Fe/Cu	13.16	19.49	30.00	22.98
Cr/Cu	0.28	0.39	0.89	0.70
Zn/Cu	2.52	2.40	3.90	4.02
Co/Cr	0.02	0.01	0.01	0.01
Fe/Cr	46.24	50.00	33.59	32.73
Cr/Ni	1.11	2.04	2.37	1.28
Fe/Ni	51.54	101.77	79.52	42.02

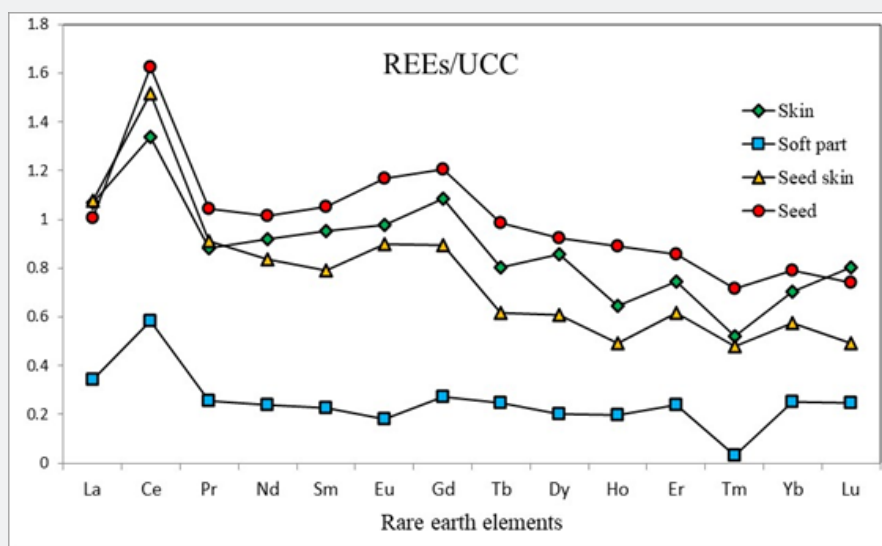


Figure3: Distribution patterns of REEs relative to the REE distribution in the continental crust.

As for Mn, Fe is essential in chlorophyll synthesis and P is essential for ATP elaboration from ADP. Similar changes of P and Mn in the fruit are in the opposite to Fe, generating Mn/Fe and P/Fe ratios that change in the same way. For both Mn and P, the transfer from the seed to the other parts is less significant than the transfer of Fe in the same conditions. Such relocation of Fe suggests the contribution of specific enzymes to facilitate the transfer from the seed. The ratios such as Fe/Cu and Fe/Mn change in similar way with high values in the seed skin; the ratios such as Zn/Ni, Fe/Zn and Fe/Cr (Table 2) showing the highest values in the soft part. The Zn/Cu ratio increases from seed to skin, which is to the opposite to decreasing Mn/Cu from seed to the other organs. The P/Fe and Mn/Fe ratios decrease in the soft part and in the seed skin, reflecting in turn a decrease of P and an increase of Fe in these parts.

In summary, about 40% of Si, 79% of Al, 26% of Mg and Ca, 21% of Fe, 30% of Mn, 15% of Ti and 24% of Na from the whole fruit, are located in the skin more than in the soft part except for

Mg and Ca and K with higher concentration in the soft part. For an average weight of 10g per date and based on the daily dietary invoke estimates of fruits: about 4 dates per day may provide 5 mg of Si, 80 mg of Al, 100 mg of Mg, 82 mg of Ca, 6.5 mg of Fe, 0.5 mg of Mn, 1.5 mg of Ti, 83 mg of Na, 88 mg of P, 0.8 mg of Sr, 1 mg of Zn and 0.4 mg of Cu.

Rare-earth elements (REEs)

Unlike the results of Bertoldi D. R, et al. [39], there are more REEs in the seed of *Vitis vinifera* L. and in its skin. Bertoldi D. R, et al. [39] reported more REEs not only in the seeds but also in the flesh parts of *Vitis vinifera*. As for the other metals, the REEs are provided by the rhizosphere. Their transportation requires ligands, which characteristics can be traced by metals. The distribution patterns of REEs can provide information about the type(s) of ligands used for transportation. It can also provide information about oxydation-reduction processes. Relative to crust, there is a similar distribution for the different parts: an

enrichment in LREEs (Figure 4) with a positive anomaly in Ce and a negative anomaly in Tm observed for the seeds and their skin.

The distribution of the REEs in the skin and the soft part is flat with the same anomalies.

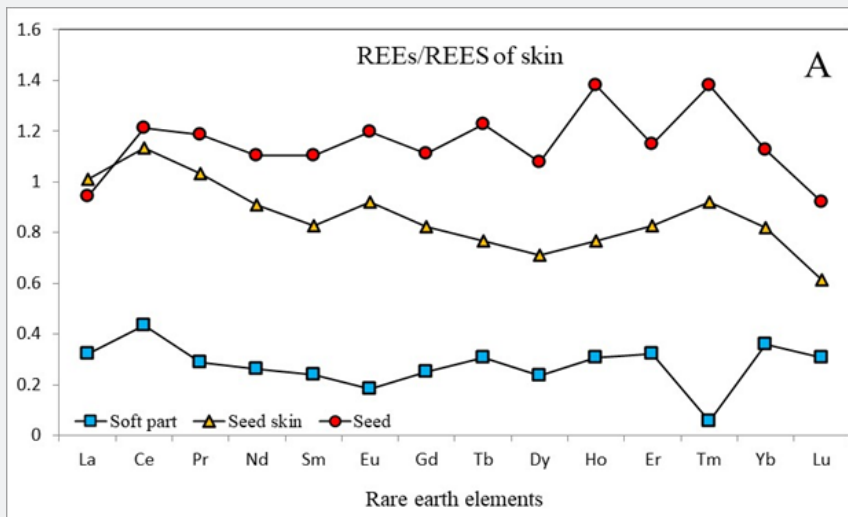


Figure4a: Distribution patterns of REEs relative to skin of Phoenix dactylifera L.

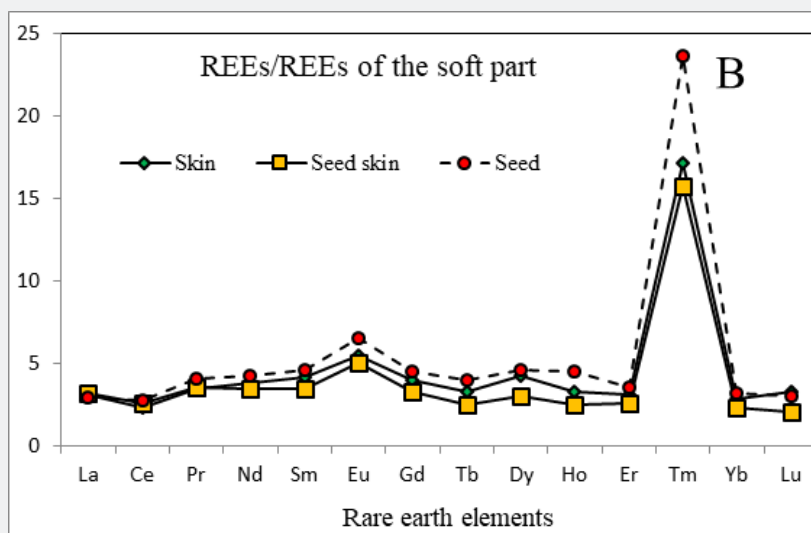


Figure4b: Distribution patterns of REEs relative to soft part of Phoenix dactylifera L.

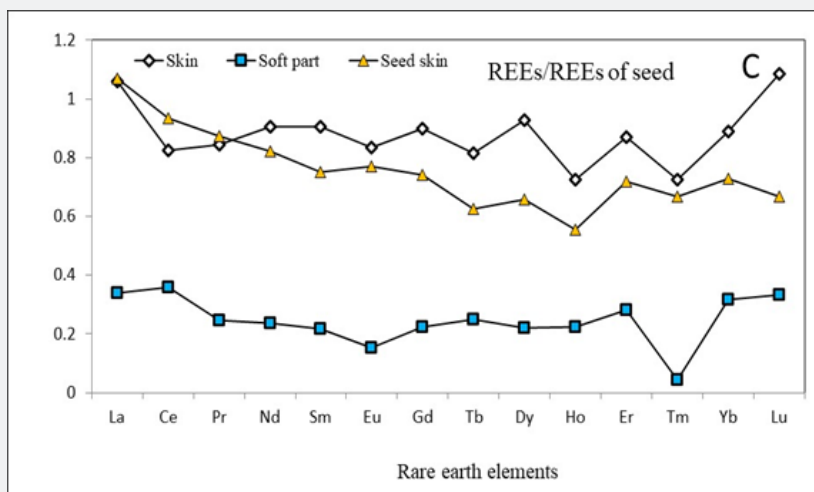


Figure4c: Distribution patterns of REEs relative to seed of *Phoenix dactylifera* L.

The enrichment in LREEs reflects a complexation with silicates. The soft part of the dates is characterized by a negative Tm anomaly and a positive Ce anomaly. Relative to skin, there is an enrichment of REEs in the seeds, a depletion of MREEs in the seed skin with a positive Ce anomaly, which indicates more Ce^{4+} than Ce^{3+} and an Eu positive anomaly suggesting more Eu^{3+} than Eu^{2+} . For the soft part, there is a slight depletion in MREEs with a Ce positive anomaly and slight negative Eu and Tm anomalies (Figure 4). As stated above, there is less P in the soft part and in the skin of the seed, which suggests that the depletion of MREEs in these two parts relative to the skin reflects a complexation of REEs with P in the skin. No correlation was observed between Fe and either the Ce or Eu anomalies, which suggests that the oxydation-reduction process is not the dominant process controlling the Ce concentration in the different parts. An enzyme action may have a control on the distribution of elements.

Relative to the soft part of the fruit, there is a slight enrichment in MREEs in the other parts, with a negative Ce anomaly and positive Eu and Tm anomalies (Figure 4). Relative to the seeds, the seed skin is characterized by depletion in MREEs and HREEs. The patterns are flat in the skin, flat in the soft part except for Ce with a positive anomaly and Eu with a negative anomaly (Figure 4). The slight depletion in MREEs characterizes the skin of the seeds and the soft part and may reflect less complexation with P in these two parts rather than in the seed. The Ce and Eu anomalies may reflect an oxydation—reduction or an enzyme effect. Since there is also a Tm anomaly, the enzyme effect is more credible. Enzymes are essential for chemical reactions and possess sites specific for elements. The soft part of the dates is depleted in REEs compared to the other parts. Unlike the soft part which is loaded with sugar and where there is little ability to complex REEs, the other parts are enriched in LREEs and depleted in MREEs and HREEs. For an average seed weight of about 0.5 g to 4 g and on the basis of a daily consumption of 4 dates, we observed that about 0.37 to 3 mg of REEs per day per capita would be provided by seeds that are never consumed. Yearly about 0.14 to 1.1 mg of REEs can be

provided per capita by the seeds, which are the uneaten part of dates. This result can be interested in industrial applications and production of energy since availability of REEs is limited.

Conclusion

We can learn from this study on various components of date fruits, that: (1) the skin is more enriched in Si, Al, Mn, Cd, Pb, Th and U than the other parts of the fruit, (2) the soft part is more enriched in Mg, Ca and Zr, (3) the seeds are more enriched in Mn, Ni, Co and REEs, (4) the seed of date is the most oxidized part. The REEs of the seeds and of the skin of the seeds are characterized by an enrichment in LREEs relative to crust with a positive anomaly in Ce and a negative anomaly in Tm that suggest a complexation with silicates. The REEs in the skin and the soft part are flat with the same anomalies.

The significant REE fractionation in the soft part outlines a slight depletion in MREEs relative to the skin of dates. For the skin of the dates, there is an enrichment in MREEs with a negative Ce anomaly and a positive anomaly in Eu and in Tm relative to the soft part. In the case of the seeds, there is an enrichment of REEs relative to skin, a slight enrichment in MREEs relative to the soft part. For 4 dates consumed per capita per day, about 0.14 to 1.1 mg of REEs can be yearly provided from uneaten seeds.

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Author Contributions

Khadija Semhi wrote the manuscript and contributed to the interpretation. Sam Chaudhuri contributed to elaboration of the project, to writing and interpretation.

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Data Availability Statement

The data supporting the outcome of this research work has been reported in this manuscript.

Conflicts of Interest

The authors declare no conflicts of interest.

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