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Abstract: Beryl occurs in the ancient Roman mines at Wadi Nugrus, South Eastern Desert of Egypt. It ranges from small crystals to 10 mm in size, and it varies in color, appearing as bright green, pale green, dark green and brown-green with biotite inclusions. The trace and minor elements were analyzed by the SIMS method. The two rims are richer in Cs, Na, Mg, Fe, Sc, V, Rb and H₂O than cores but are poor in Mn, Ca, Co, Sr and Li. The bright-green rim is richer than the pale-green one in Na, P, K, Ca, Fe, Rb, Cs and F, but poorer in Mg and Li. The alkaline elements (Cs, Na, Rb) and Fe correlate with the color zoning, and where beryl crystals have a maximum of these elements, the green color is strong and bright. The emerald of Wadi Nugrus has similarities with the geological setting of the Canadian emeralds. Emeralds occur along the contact zone between biotite schists, pegmatites and quartz veins. A large-scale interaction between Be-bearing magmatic fluids from granites and related pegmatites took place with hydrothermal fluids enriched in Cr, V, Sc, Mg and Ca after percolation through pre-existing serpentinite and talc carbonates, metagabbros and biotite schists and additional fluids bearing H₂O, NaCl and CO₂.

Keywords: beryl; SIMS method; trace element geochemistry; zoning; Wadi Nugrus; Egypt

1. Introduction

The demands for beryllium metal (Be), oxides and alloys are steadily increasing their importance in atomic energy programs. It is well known that the mineral beryl (Be₃Al₂Si₆O₁₈) is one of the main resources for beryllium worldwide. This could be related to its versatile application, especially in nuclear industries, and it is used as a basic material in space shuttles and nuclear reactors [1–3]. On the other hand, beryl has various colors and durability that have made it a fascinating gemstone from the ancient periods to recent times. The different varieties of beryl are distinguished as colorless to white (goshenite), pale green to dark green (emerald, contains traces of Cr^{3+} and V^{3+}), yellow or golden (heliodor, contains trace amounts of Fe³⁺), light blue (aquamarine, contains traces of Fe²⁺), dark blue beryl (maxixe, contains traces of Fe²⁺ and Fe³⁺), pink or rose (morganite, contains impurities of Mn²⁺) and red (bixbite, contains traces of Mn³⁺) [4–8].

Several occurrences were intensively explored for beryl all over the world. The paragenetic types of beryl mineral can be categorized from different geological settings worldwide according to [9–11] as follows: (1) highly evolved granites (S-type) and associated pegmatites [11–21]; (2) hydrothermal beryllium ore deposits related to metasomatized granites



Citation: Abdel Gawad, A.E.; Ene, A.; Skublov, S.G.; Gavrilchik, A.K.; Ali, M.A.; Ghoneim, M.M.; Nastavkin, A.V. Trace Element Geochemistry and Genesis of Beryl from Wadi Nugrus, South Eastern Desert, Egypt. *Minerals* 2022, 12, 206. https://doi.org/ 10.3390/min12020206

Academic Editor: Frederick Lin Sutherland

Received: 20 December 2021 Accepted: 2 February 2022 Published: 6 February 2022

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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). (e.g., greisen) [8,22–26]; (3) volcanogenic hosted beryllium deposits [14]; (4) emeraldbearing schist [3,8,17,18,27,28]; (5) carbonate-hosted beryllium ore deposit [6,14].

The Arabian Nubian Shield (ANS) has resulted from the collision during the Cryogenian– Ediacaran periods between East and West Gondwana [29]. Collision zones extend from the Middle East southward through Egypt, Sudan, Eritrea, Ethiopia, Somalia, Saudi Arabia, Yemen, Oman and Jordan (Figure 1a) and are dated from 700 and 580 Ma [30].

The ANS is a treasury of economic mineralization and could be categorized as follows: (1) the subduction stage dated ~870–635 Ma (first one), which is composed of island arc volcano–sedimentary associated with plutonic rocks [30]; (2) the continental collision stage dated ~630–580 Ma (second stage); and (3) the postcollision stage dated 580–540 Ma (third stage). The second and third stages are distinguished by the sedimentary basins' development that accompanies calc-alkaline to alkaline igneous intrusion [31–34]. The basement rocks of the Eastern Desert of Egypt represent the western part of the ANS [35] (Figure 1a,b).

Beryl was an important emerald resource worldwide during the Roman era, especially in the northeastern part of Africa. In Egypt, beryl was mined in the Eastern Desert since antiquity. Intensive mining explorations of beryl were made by Romans wh'o built settlements that are very close to the "Cleopatra's Mines" at Wadi Sikait in the vicinity of ancient emerald mining of Wadi Nugrus [36,37]. The Egyptian favorable environs of beryl in the Precambrian basement rocks can be categorized into two paragenetic types: (1) emerald in mica-schists and (2) beryl in granitoids [23,38–40].

However, granitoids bearing Be can be categorized into two types as follows: (1) peraluminous granites, Ta > Nb+Sn+Be±W-enriched, such as Li-rich albite granite (e.g., Nuweibi and Abu Dabbab areas), and (2) metasomatized granites, Nb >> Ta+Be+Sn+Mo+Wenriched, such as alkali feldspar granite (e.g., Homr Mukpid, Homr Akarim and Qash Amir areas) [23].

According to Abdalla and Mohamed [23], EPMA data showed that emeralds from mica schist at Wadi Sikait and Um Kabu (Egypt) are mainly composed of SiO_2 and Al_2O_3 with average values of 65.12 and 15.06 wt%, respectively. The trace and minor elements, including TiO₂, Cr₂O₃, FeO, MnO, NiO, MgO, CaO, Na₂O and K₂O, have average contents of 0.02, 0.13, 0.45, 0.03, 0.02, 2.33, 0.01, 1.8 and 0.04 wt%, respectively. The average water content is 2.38 wt%.

Shaw and Bunbury [41], based on EPMA data, stated that emeralds from Wadi Sikait (Egypt) were mainly composed of SiO₂ and Al₂O₃ with average values of 64.52, 13.52 and 13.1 wt%, respectively. In contrast, trace and minor elements including TiO₂, Cr₂O₃, FeO, MnO, MgO, CaO, Li₂O, Na₂O and K₂O have average values of 0.01, 0.13, 0.3, 0.02, 2.53, 0.03, 0.02, 1.61 and 0.04 wt%.

The genesis of green emeralds from Sikait, Zabara and Umm Kabo in the South Eastern Desert of Egypt is an important subject. Grundmann and Morteani [42] showed that emerald-bearing schists and quartz veins could be interpreted as a result of (a) the major shear zone of the Wadi Nugrus which controls an extensive alkali-metasomatism followed by postmagmatic fluids; (b) an alternative interaction between pegmatites and the pre-existing rocks varying from basic to ultrabasic rocks; and (c) syn- to post-tectonic regional metamorphism and Na-metasomatism.

As mentioned before, beryl mineralization occurs in different geological settings, but we focused on trace and minor element geochemistry of beryl from the ancient Roman sites. This work aims at investigating the zonation of beryl mineralization and the behavior of trace and minor elements from the marginal (rim) and central (core) zones, as well as beryl genesis in the Wadi Nugrus area, South Eastern Desert of Egypt.



Figure 1. (a) Geologic map showing the Arabian Nubian Shield (ANS); (b) geologic map showing the basement rocks of Neoproterozoic age in the Eastern Desert of Egypt [35]; (c) geological map of Wadi Nugrus, South Eastern Desert, Egypt, after [43–46].

2. Geologic Setting

The present work shows that the exposed basement rocks of Wadi Nugrus (Figure 1c) can be arranged from the oldest rock types to the youngest ones [43–46] as follows:

Biotite gneisses occur in the northern part of the mapped area (Figure 1c). These rocks are medium- to coarse-grained, greyish-white to pale pink colors, low to moderate relief. They are essentially composed of quartz, K-feldspar, plagioclase and biotite. Biotite gneisses are highly deformed, sheared, altered and affected by the deformational history of the pre-Pan-African gneisses and migmatites [43,47].

Mylonite (cataclastic) rocks occur in the northern part of the Wadi Nugrus area and are fine- to medium-grained, low in relief and capped by metasediments through thrust fault. They are composed of quartz, K-feldspar, plagioclase, muscovite and subordinate amounts of biotite. These rocks were highly deformed, altered and mylonitic fabric that resulted from ductile and brittle deformations. The studied area was dissected by shear zones having NNW–SSE, N–S and NE–SW trends [48,49]. Mylonites are considered as one of the important rocks bearing rare-metal mineralization in the studied area. They are enriched in uraninite, kasolite, uranophane, thorite, uranothorite, zircon, ishikawaite, columbite, fergusonite, xenotime, synchysite and cassiterite, as well as fluorite, apatite, barite and galena [47,50–52].

Serpentinites and talc carbonate (ultramafite) rocks are distributed as small masses, cavernous, fine- to medium-grained, and low to moderate in relief. They are pale gray to dark gray, violet-brown patches which characterize serpentinites and associated talc carbonates. These rocks are distinguished by their softness and are mainly composed of serpentinite, talc carbonates and magnesite minerals with chromite. The serpentinite rocks were thrust over metasediments, and the thrust fault strikes NE–SW and dips SE [48,49]. *Metagabbros (ophiolitic gabbros)* are widely distributed in the Wadi Nugrus as an elongated belt having NW–SE direction (Figure 1c). They are hard and medium- to coarsegrained and have black to dark gray colors. These rocks show low to moderate relief and are composed mainly of plagioclase, hornblende and pyroxene. The metagabbros are obducted on mylonites and intruded by the pegmatites.

Metasediments are represented by schists. They are highly foliated, lineated and finegrained; they have low to moderate relief, whitish-grey to dark grey colors and NW–SE and NE–SW trends [48,49]. They are essentially composed of hornblende schists, mica schists and graphite schists. These rocks were intruded by muscovite granite and pegmatites and represent roof pendants on top of them. Quartz veins crosscut these rocks (Figure 2a,b). Ancient Roman mines were used for green beryl exploration (Figure 2c,d). These mining processes resulted in huge mine dumps containing significant amounts of beryl.

Garnet muscovite granites occur as an elongate belt having NW–SE trend in the Um Solimate area. The granite has moderate to high relief and is highly fractured, jointed, cavernous and altered. It is distinguished by pink to reddish-pink colors; medium- to coarse-grained; and composed mainly of K-feldspar, plagioclase, quartz, muscovite and biotite, in addition to garnet. Um Solimate granites have been intruded into biotite gneisses, mylonites and metasediments and were crosscut by pegmatites and dikes.







(c)





(**d**)



Figure 2. (a) Quartz vein crosscut metasediments; (b,c) ancient Roman sites for beryl exploration; (d) green beryl (emerald) in quartz vein; (e) dark-green to brown-green emerald; (f) pale-green emerald in quartz.

Garnet muscovite granitic pegmatites vary from large to small masses that intruded all the pre-existing different basement rocks in the eastern part of Wadi Nugrus (Figure 1c). They occur as elongated belts having NW–SE trends and host xenoliths of different shapes and sizes. These rocks are very coarse-grained, composed mainly of megacrysts of Kfeldspar, quartz muscovite and biotite in addition to garnet. Pegmatites are enriched in rare-metal mineralization, especially Bi–Ni–Ag–Nb–Ta in addition to U and Th [46]. They were crosscut by basic dikes as well as quartz veins bearing tourmaline and beryl (Figures 1c and 2d–f). Greenish beryl crystals occur in the pegmatites, within the muscoviterich zones, especially outer to intermediate zones, and crystals reach up to 3 cm long (Figure 2d–f). Beryl also occurs as disseminated grains in quartz veins crosscut by metasediments and muscovite granite.

3. Materials and Methods

Pure beryl samples were obtained from the ancient mining dumps along the contact between metasediments and pegmatites of Wadi Nugrus, South Eastern Desert of Egypt (Figure 1c), using crude sieving followed by hand picking.

During the preparation of the standard epoxy mount (1-inch diameter) with minerals, the crystal was cut perpendicular to the prism faces (the elongation axis) in order to be able to study the growth zoning of beryl along the traverse from the center (core) to the edge (rims) of the crystal in a plane parallel to the basal pinacoid. The cross-section, probably, mainly characterizes the composition of the growth zones of the {10 1 0} prism face (Figure 3a).

The contents of trace and minor elements, water and volatiles in beryl were determined by secondary ion mass spectrometry (SIMS) using a Cameca IMS-4f ion microprobe at the Yaroslavl branch of the Institute of Physics and Technology (IPT) named after K.A. Valiev, Russian Academy of Sciences. The basics of the measurement technique corresponded to those reported in [53–56].



Figure 3. Natural zoned beryl crystal from the Wadi Nugrus, South Eastern Desert, Egypt, without gold coating (**a**) and transverse section of the studied beryl (with gold coating) and 25 analytical points (**b**).

The analyses were carried out in two steps using different protocols for the determination of volatiles (Cl, F, H) and light (B, Li) impurity elements and the main set (Na, Mg, P, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Ga, Rb, Cs). Primary O^{2-} ions were accelerated to about 14.5 keV and focused at the sample surface into a spot about $20-30 \ \mu\text{m}$ in diameter. The intensity of primary ion current was 5 nA (protocol «volatiles») and 1.5 nA (main protocol). Positive secondary ions were collected from an area of $10 \ \mu\text{m}$ (protocol «volatiles») or 25 $\ \mu\text{m}$ (main protocol) in diameter, limited by a field aperture. Molecular and ion clusters were energy-filtered using an offset voltage of $-100 \ \text{V}$, with an energy window of 50 eV. Three counting cycles were carried out with a discrete transition between mass peaks within a given set. The counting time was varied depending on signal intensity and was determined automatically by statistical control. The maximum counting time for any species in each cycle was 30 s.

The absolute concentrations of each element were calculated from the measured intensities of positive single-atom secondary ions (I_i), which were normalized to the intensity of secondary ${}^{30}\text{Si}^+$ ions (I_ ${}^{30}\text{Si}$), using the relative sensitivity factors K_i: C_i = I_i/I_ ${}^{30}\text{Si} \times \text{K}_i \times \text{C}_{\text{Si}}\text{O}_2$.

Calibration curves were based on the measurements of the set of well-characterized standard samples [57]. Quantification of phosphorus, scandium, iron, nickel and cobalt was carried out taking into account isobaric mass peak interference:

The phosphorous content was estimated using a stripping procedure. The contribution
of ³⁰Si¹H⁺ to the measured intensity of mass peak at 31 a.m.u. was determined from
intensity ²⁹Si⁺ and known abundances of natural silicon isotopes:

$$\label{eq:si1} \begin{split} ^{28}\mathrm{Si}^{1}\mathrm{H}^{+}/^{30}\mathrm{Si}^{+} &= \mathrm{I}(29 \ \mathrm{a.m.u.})/^{30}\mathrm{Si}^{+} - 1.517 \\ ^{30}\mathrm{Si}^{1}\mathrm{H}^{+}/^{30}\mathrm{Si}^{+} &= ^{28}\mathrm{Si}^{1}\mathrm{H}^{+}/^{30}\mathrm{Si}^{+}/29.875 \\ 31\mathrm{P}^{+}/^{30}\mathrm{Si}^{+} &= \mathrm{I}(31 \ \mathrm{a.m.u.})/^{30}\mathrm{Si}^{+} - {}^{30}\mathrm{Si}^{1}\mathrm{H}^{+}/^{30}\mathrm{Si}^{+} \end{split}$$

- 2. When quantifying scandium, the subtraction of ²⁹Si¹⁶O⁺ + ²⁸Si¹⁷O⁺ ion signal in the range of 45 a.e.m. required additional measurement of signal intensity at the 44th mass (²⁸Si¹⁶O⁺ + ⁴⁴Ca⁺). The contribution of ⁴⁴Ca⁺ was estimated by recalculating from measured ⁴²Ca⁺ intensity and known natural calcium isotope abundances [58].
- 3. The ⁵⁶Fe⁺ and ⁵⁹Co⁺ signal was corrected regarding a Si²⁺ cluster ion spectrum, assuming that the Si⁺/Si²⁺ intensity ratio for this matrix is known and varies slightly if the sample charging level is controlled.
- 4. The contribution of ⁴⁶Ti¹⁶O⁺ was taken into account when calculating nickel concentration by measuring the 62nd nickel isotope. TiO⁺ signal intensity was estimated by measuring ⁴⁷Ti⁺ ion current intensity, and ⁴⁷Ti⁺/⁴⁶Ti¹⁶O⁺ was measured in standard glasses. Because Ti concentration in beryl is low (15–34 ppm), the corrected Ni concentration is very close to the uncorrected one.

Ion probing of water required known approaches to decrease the background level. Before measurements, each sample was kept for at least 12 h in a mass spectrometer's analytical chamber, where high vacuum conditions were maintained. Analysis was preceded by the ion sputtering of conducting gold film and a surface pollutant layer at the area to be analyzed. Then the procedure of auto adjustment of the sample potential was applied. The static primary beam spot overlapped the field of view of the secondary ion optics (10 μ m in diameter) centered at the presputtered crater (~40 × 40 μ m). An anhydrous silicate (olivine) grain was introduced into each sample mount to measure the ¹H⁺/³⁰Si⁺ ion current ratio based on calibration relationships:

$$C[H_2O]/C[SiO_2] = (I(^{1}H^+) - I(^{1}H^+)bg)/I(^{30}Si^+) \times RSF \times K(SiO_2)$$

 $C[H_2O]$ and $C[SiO_2]$ are H_2O and SiO_2 concentrations in wt.%; $I(^1H^+)$, $I(^1H^+)$ bg and $I(^{30}Si^+)$ are measured secondary ion intensities in imp/s; $I(^1H^+)$ bg is the background signal intensity; and RSF is the relative sensitivity factor. The correction coefficient K(SiO₂) takes into account the dependence of RSF on SiO₂ concentration for which linear approximation was used:

$$K(SiO_2) = (1 - (SiO_2 - 50) \times 0.0185)$$

Calibrations were obtained using samples of natural and artificial glasses (like NIST glasses) (in all, 28 standard samples), covering a wide range of variation in SiO₂ (41 \div 77 wt%) and water (0.1 \div 8 wt.%) concentrations [59–63] (unpublished data of R.E. Bocharnikov). The results of calibration show that the maximum deviation from the reference value was no more than 15%, and the calculation error was 7%.

A similar approach was used for calculating fluorine and chlorine concentrations. The method for determining fluorine in silicate matrices is published in [64].

Standard glass NIST-610 [65] was used as a monitor before an analytical session. The trace and minor element detection limit ranges mainly from 0.005 to 0.010 ppm. The trace element measurement error did not exceed 10% for concentrations higher than 1 ppm and 20% for concentrations in the range of 0.1–1 ppm.

The analytical points (n = 25) are presented in Figure 3b, comprising the beryl sample's rim and core parts. The points are labeled as 1-5 (bright-green rim), 20-25 (pale-green rim), and 6-19 (green-brown) for the core.

4. Results

4.1. Characteristics of Beryl Mineralization

The beryl crystals from Wadi Nugrus are dominated by hexagonal prisms, elongate with marked zoning. The selected beryl crystal was characterized by transparent and green beryl (emerald) on a polished surface with a zonal change in color (Figure 3a). The color transparency and saturation vary from the bright-green rim (spots 1–5) on the left side of the beryl crystal, to the brown-green core (spots 6–19), to the pale-green rim (spots 20–25) on the right side that could reveal zoning for a number of trace and minor elements (Figure 3a,b and Table 1). The transparent area of the rim part on the left side through

the cross-section is most likely indicating a large number of gas–liquid inclusions [20]. The selected spots are listed on the profile are chosen in such a way to cover the different colors in the zoned beryl crystal from core to both rims. The direction of the profile itself is selected in order to avoid inclusions and cracks.

Table 1. Representative trace and minor element concentrations (ppm) in the analyzed beryl from Wadi Nugrus, South Eastern Desert, Egypt.

F1 (Analytical Spots																		
Element	1	2	3	4	5	20	21	22	23	24	25	Rim	1–5 *	Rim	20–25 *				
Na	18,205	17,248	15,722	14,727	16,942	15,642	15,158	14,938	16,399	15,505	14,978	16,5	569	15	,437				
Mg	14,423	15,054	14,439	13,574	16,077	15,307	15,069	15,081	15,627	15,177	15,410	14,7	714	15,279					
Р	123	253	322	233	295	385	54.7	266	237	158	7.47	24	5	185					
K	734	710	571	598	763	638	470	469	527	445	222	67	5	4	62				
Ca	327	477	379	388	557	491	315	334	389	383	219	42	6	3	555				
Sc	216	231	254	266	258	180	283	218	198	222	243	245		2	24				
11	29.0 295	24.0	23.3	22.3	21.7	17.2	16.5	15.7	20.1	21.4	15.3	24	.1	1	1./				
v Cr	285 760	370 885	418	406	403	332 806	403	393	393 772	394 852	424	37	0 /1	404 880					
Cr Mn	700 574	000 67.8	68 /	67.0	090 71 4	090 70.6	701 50 3	024 63 7	775.8	65 3	1255 54 7	66	1	64 9					
Fe	2199	2342	2081	2097	2567	1937	2083	2109	1800	1876	2078	22	.0 57	1980					
Co	1 23	1 43	1 29	1 05	1 43	1 18	0.99	1 08	1.31	1.08	0.79	1.2	<i>b</i> 9	1.07					
Ni	8.90	14.1	10.9	11.1	12.2	13.8	11.7	11.1	11.3	10.5	8.5	11	.4	11.07					
Ga	8.57	8.71	7.17	6.81	7.97	6.71	5.64	5.78	6.39	6.80	5.53	7.8	35	6.14					
Rb	27.0	25.6	21.8	20.3	24.7	21.2	17.6	21.0	20.0	18.4	18.5	23	.9	19.5					
Cs	194	234	246	282	238	107	229	132	168	195	145	23	9	163					
В	1.12	1.30	1.11	1.27	2.13	1.29	1.32	1.36	1.30	1.96	1.53	1.3	39	1.46					
Li	142	140	129	164	134	130	144	171	160	157	167	14	2	1	.55				
H_2O	41,144	43,805	38,415	42,073	43,985	41,321	41,217	44,387	38,865	43,454	38,646	41,8	384	41	,315				
Cl	1066	1137	1029	1126	1129	1025	955	1160	1133	1163	1183	10	97	1	103				
F	21.7	16.6	14.0	19.2	20.6	11.2	15.5	11.4	5.75	7.28	5.32	18	.4	9	.42				
Sr	2.28	4.25	3.22	3.15	4.69	4.29	1.18	2.43	4.37	3.81	0.16	3.5	52	2	.71				
					Analytical Spots														
El anna ana t																			
Element	6	7	8	9	10	11	12	13	14	15	16	17	18	19	Core 6–19 **				
Na	6	7	8	9	10	11	12	13	14	15	16	17 13 944	18	19	Core 6–19 **				
Na Mg	6 15,486 14 218	7 15,435 12 669	8 15,871 13,775	9 16,013 14,460	10 14,841 13,631	11 14,785 13,611	12 13,801 13,647	13 14,161 14,766	14 14,547 13,647	15 13,029 12 778	16 12,972 12 590	17 13,944 13 200	18 14,280 13 563	19 15,120 14 316	Core 6–19 ** 14,592				
Na Mg P	6 15,486 14,218 265	7 15,435 12,669 327	8 15,871 13,775 393	9 16,013 14,460 196	10 14,841 13,631 341	11 14,785 13,611 308	12 13,801 13,647 298	13 14,161 14,766 179	14 14,547 13,647 452	15 13,029 12,778 558	16 12,972 12,590 521	17 13,944 13,200 231	18 14,280 13,563 467	19 15,120 14,316 563	Core 6–19 ** 14,592 13,634 364				
Na Mg P K	6 15,486 14,218 265 721	7 15,435 12,669 327 781	8 15,871 13,775 393 622	9 16,013 14,460 196 415	10 14,841 13,631 341 598	11 14,785 13,611 308 559	12 13,801 13,647 298 895	13 14,161 14,766 179 389	14 14,547 13,647 452 405	15 13,029 12,778 558 415	16 12,972 12,590 521 376	17 13,944 13,200 231 442	18 14,280 13,563 467 504	19 15,120 14,316 563 527	Core 6–19 ** 14,592 13,634 364 546				
Na Mg P K Ca	6 15,486 14,218 265 721 518	7 15,435 12,669 327 781 537	8 15,871 13,775 393 622 502	9 16,013 14,460 196 415 581	10 14,841 13,631 341 598 432	11 14,785 13,611 308 559 379	12 13,801 13,647 298 895 444	13 14,161 14,766 179 389 464	14,547 13,647 452 405 526	15 13,029 12,778 558 415 482	16 12,972 12,590 521 376 418	17 13,944 13,200 231 442 386	18 14,280 13,563 467 504 394	19 15,120 14,316 563 527 487	Core 6–19 ** 14,592 13,634 364 546 468				
Na Mg P K Ca Sc	6 15,486 14,218 265 721 518 214	7 15,435 12,669 327 781 537 134	8 15,871 13,775 393 622 502 211	9 16,013 14,460 196 415 581 199	10 14,841 13,631 341 598 432 204	11 14,785 13,611 308 559 379 211	12 13,801 13,647 298 895 444 155	13 14,161 14,766 179 389 464 164	14 14,547 13,647 452 405 526 148	15 13,029 12,778 558 415 482 146	16 12,972 12,590 521 376 418 148	17 13,944 13,200 231 442 386 137	18 14,280 13,563 467 504 394 136	19 15,120 14,316 563 527 487 154	Core 6-19 ** 14,592 13,634 364 546 468 169				
Na Mg P K Ca Sc Ti	6 15,486 14,218 265 721 518 214 18.9	7 15,435 12,669 327 781 537 134 28.9	8 15,871 13,775 393 622 502 211 21.7	9 16,013 14,460 196 415 581 199 24.9	10 14,841 13,631 341 598 432 204 200	11 14,785 13,611 308 559 379 211 17.4	12 13,801 13,647 298 895 444 155 33.9	13 14,161 14,766 179 389 464 164 18.9	14 14,547 13,647 452 405 526 148 19.2	15 13,029 12,778 558 415 482 146 16.4	16 12,972 12,590 521 376 418 148 148 17.2	17 13,944 13,200 231 442 386 137 30.8	18 14,280 13,563 467 504 394 136 20.2	19 15,120 14,316 563 527 487 154 20.5	Core 6–19 ** 14,592 13,634 364 546 468 169 22.1				
Na Mg P K Ca Sc Ti V	6 15,486 14,218 265 721 518 214 18.9 336	7 15,435 12,669 327 781 537 134 28.9 262	8 15,871 13,775 393 622 502 211 21.7 349	9 16,013 14,460 196 415 581 199 24.9 337	10 14,841 13,631 341 598 432 204 20.0 333	11 14,785 13,611 308 559 379 211 17.4 355	12 13,801 13,647 298 895 444 155 33.9 299	13 14,161 14,766 179 389 464 164 18.9 336	14,547 13,647 452 405 526 148 19.2 316	15 13,029 12,778 558 415 482 146 16.4 307	16 12,972 12,590 521 376 418 148 148 17.2 294	17 13,944 13,200 231 442 386 137 30.8 288	18 14,280 13,563 467 504 394 136 20.2 297	19 15,120 14,316 563 527 487 154 20.5 322	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317				
Na Mg P K Ca Sc Ti V Cr	6 15,486 14,218 265 721 518 214 18.9 336 971	7 15,435 12,669 327 781 537 134 28.9 262 761	8 15,871 13,775 393 622 502 211 21.7 349 905	9 16,013 14,460 196 415 581 199 24.9 337 919	10 14,841 13,631 341 598 432 204 20.0 333 885	11 14,785 13,611 308 559 379 211 17.4 355 875	12 13,801 13,647 298 895 444 155 33.9 299 710	13 14,161 14,766 179 389 464 164 18.9 336 750	14 14,547 13,647 452 405 526 148 19.2 316 589	15 13,029 12,778 558 415 482 146 16.4 307 691	16 12,972 12,590 521 376 418 148 17.2 294 718	17 13,944 13,200 231 442 386 137 30.8 288 666	18 14,280 13,563 467 504 394 136 20.2 297 727 727	19 15,120 14,316 563 527 487 154 20.5 322 707	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 7777				
Na Mg P K Ca Sc Ti V Cr Mn	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 73.2	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 70.5	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 74.0	14 14,547 13,647 452 405 526 148 19.2 316 589 70.1	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6	16 12,972 12,590 521 376 418 148 17.2 294 718 65.6	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4				
Na Mg P K Ca Sc Ti V Cr Mn Fe	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 2038	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170	10 14,841 13,631 341 598 432 204 20,0 333 885 67,6 1874 2020	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 112	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671	14 14,547 13,647 452 405 526 148 19.2 316 589 70.1 1729	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694	16 12,972 12,590 521 376 418 148 17.2 294 718 65.6 165.6 165.6	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8 1653 120	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4 1853 854				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6 1874 0.99	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10 4	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 776	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671 3.53 125	14 14,547 13,647 452 405 526 148 19.2 316 589 70.1 1729 0.98 0.74	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694 1.00	16 12,972 12,590 521 376 418 148 148 17.2 294 718 65.6 1656 0.96	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.01	18 14,280 13,563 467 504 394 136 20,2 297 727 68.8 1653 1.20	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 19.7	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4 1853 1.34 21.0				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05 12.9 7.72	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21 9.89 2.56	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24 11.7 8 26	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32 46.6 8 20	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6 1874 0.99 10.0 7.05	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10.4 7.52	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 7.76 10 6	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671 3.53 125 6.46	14 14,547 13,647 452 405 526 148 19.2 316 589 70.1 1729 0.98 9.74 564	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694 1.00 10.4 5 85	16 12,972 12,590 521 376 418 148 148 17.2 294 718 65.6 1656 0.96 10.2 5 86	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.91 6 12	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8 1653 1.20 11.4 4 6 26	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 10.7 7 22	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4 1853 1.34 21.0 7.18				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni Ga Rb	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05 12.9 7.72 22.8	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21 9.89 7.56 18.9	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24 11.7 8.36 17 7	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32 46.6 8.29 12.8	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6 1874 0.99 10.0 7.05 19.2	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10.4 7.53 17 1	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 7.76 10.6 19.0	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671 3.53 125 6.46 16.8	14 14,547 13,647 452 405 526 148 19.2 316 589 70.1 1729 0.98 9.74 5.64 14.0	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694 1.00 10.4 5.85 16 4	16 12,972 12,590 521 376 418 148 148 17.2 294 718 65.6 1656 0.96 10.2 5.86 14.8	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.91 6.12 14 5	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8 1653 1.20 11.4 6.26 15.1	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 10.7 7.22 18.0	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4 1853 1.34 21.0 7.18 169				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni Ga Rb Cs	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05 12.9 7.72 22.8 142	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21 9.89 7.56 18.9 88 2	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24 11.7 8.36 17.7 100	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32 46.6 8.29 12.8 115	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6 1874 0.99 10.0 7.05 19.2 100	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10.4 7.53 17.1 97.0	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 7.76 10.6 19.0 82.0	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671 3.53 125 6.46 16.8 75 3	14 14,547 13,647 452 405 526 148 19.2 316 589 70.1 1729 0.98 9.74 5.64 14.0 80 5	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694 1.00 10.4 5.85 16.4 70.6	16 12,972 12,590 521 376 418 148 148 17.2 294 718 65.6 1656 0.96 10.2 5.86 14.8 75 4	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.91 6.12 14.5 78 5	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8 1653 1.20 11.4 6.26 15.1 91 2	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 10.7 7.22 18.0 84 9	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4 1853 1.34 21.0 7.18 16.9 91 5				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni Ga Rb Cs B	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05 12.9 7.72 22.8 142 1 30	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21 9.89 7.56 18.9 88.2 1.22	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24 11.7 8.36 17.7 100 1 17	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32 46.6 8.29 12.8 115 2.17	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6 1874 0.99 10.0 7.05 19.2 100 1.19	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10.4 7.53 17.1 97.0 0 91	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 7.76 10.6 19.0 82.0 160	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671 3.53 125 6.46 16.8 75.3 1.32	14 14,547 13,647 452 405 526 148 19.2 316 589 70.1 1729 0.98 9.74 5.64 14.0 80.5 116	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694 1.00 10.4 5.85 16.4 70.6 0 97	16 12,972 12,590 521 376 418 148 17.2 294 718 65.6 1656 0.96 10.2 5.86 14.8 75.4 119	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.91 6.12 14.5 78.5 110	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8 1653 1.20 11.4 6.26 15.1 91.2 1<75	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 10.7 7.22 18.0 84.9 0.89	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4 1853 1.34 21.0 7.18 16.9 91.5 1.28				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni Ga Rb Cs B Li	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05 12.9 7.72 22.8 142 1.30 138	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21 9.89 7.56 18.9 88.2 1.22 195	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24 11.7 8.36 17.7 100 1.17 165	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32 46.6 8.29 12.8 115 2.17 153	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6 1874 0.99 10.0 7.05 19.2 100 1.19 194	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10.4 7.53 17.1 97.0 0.91 156	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 7.76 10.6 19.0 82.0 1.60 151	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671 3.53 125 6.46 16.8 75.3 1.32 159	$\begin{array}{c} 14\\ 14,547\\ 13,647\\ 452\\ 405\\ 526\\ 148\\ 19.2\\ 316\\ 589\\ 70.1\\ 1729\\ 0.98\\ 9.74\\ 5.64\\ 14.0\\ 80.5\\ 1.16\\ 171\\ \end{array}$	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694 1.00 10.4 5.85 16.4 70.6 0.97 180	16 12,972 12,590 521 376 418 148 17.2 294 718 65.6 1656 0.96 10.2 5.86 14.8 75.4 1.19 163	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.91 6.12 14.5 78.5 1.10 177	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8 1653 1.20 11.4 6.26 15.1 91.2 1.75 187	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 10.7 7.22 18.0 84.9 0.89 167	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4 1853 1.34 21.0 7.18 16.9 91.5 1.28 168				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni Ga Rb Cs B Li H ₂ O	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05 12.9 7.72 22.8 142 1.30 138 39,370	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21 9.89 7.56 18.9 88.2 1.22 195 36,375	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24 11.7 8.36 17.7 100 1.17 165 38,005	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32 46.6 8.29 12.8 115 2.17 153 37,791	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6 1874 0.99 10.0 7.05 19.2 100 1.19 194 46,383	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10.4 7.53 17.1 97.0 0.91 156 37,725	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 7.76 10.6 19.0 82.0 1.60 151 41,383	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671 3.53 125 6.46 16.8 75.3 1.32 159 40,646	14 14,547 13,647 452 405 526 148 19.2 316 589 70.1 1729 0.98 9.74 5.64 14.0 80.5 1.16 171 38,748	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694 1.00 10.4 5.85 16.4 70.6 0.97 180 36,644	16 12,972 12,590 521 376 418 148 17.2 294 718 65.6 1656 0.96 10.2 5.86 14.8 75.4 1.19 163 38,850	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.91 6.12 14.5 78.5 1.10 177 38,579	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8 1653 1.20 11.4 6.26 15.1 91.2 1.75 187 42,093	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 10.7 7.22 18.0 84.9 0.89 167 39.262	Core 6-19 ** 14,592 13,634 364 546 468 169 22.1 317 777 70.4 1853 1.34 21.0 7.18 16.9 91.5 1.28 168 239,418				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni Ga Rb Cs B Li H ₂ O Cl	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05 12.9 7.72 22.8 142 1.30 138 39,370 1172	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21 9.89 7.56 18.9 88.2 1.22 195 36,375 1241	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24 11.7 8.36 17.7 100 1.17 165 38,005 1094	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32 46.6 8.29 12.8 115 2.17 153 37,791 950	10 14,841 13,631 341 598 432 204 20.0 333 885 67.6 1874 0.99 10.0 7.05 19.2 100 1.19 194 46,383 1117	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10.4 7.53 17.1 97.0 0.91 156 37,725 978	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 7.76 10.6 19.0 82.0 1.60 151 41,383 1052	13 14,161 14,766 179 389 464 164 18.9 336 750 74.0 2671 3.53 125 6.46 16.8 75.3 1.32 159 40,646 1058	$\begin{array}{c} 14\\ 14,547\\ 13,647\\ 452\\ 405\\ 526\\ 148\\ 19.2\\ 316\\ 589\\ 70.1\\ 1729\\ 0.98\\ 9.74\\ 5.64\\ 14.0\\ 80.5\\ 1.16\\ 171\\ 38,748\\ 1059 \end{array}$	15 13,029 12,778 558 415 482 146 16.4 307 691 67.6 1694 1.00 10.4 5.85 16.4 70.6 0.97 180 36,644 1120	16 12,972 12,590 521 376 418 148 17.2 294 718 65.6 1656 0.96 10.2 5.86 14.8 75.4 1.19 163 38,850 1138	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.91 6.12 14.5 78.5 1.10 177 38,579 1021	18 14,280 13,563 467 504 394 136 20.2 297 727 68.8 1653 1.20 11.4 6.26 15.1 91.2 1.75 187 42,093 1125	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 10.7 7.22 18.0 84.9 0.89 0.89 167 39,262 1048	Core 6-19 ** 14,592 13,634 364 546 468 169 22,1 317 777 70,4 1853 1.34 21.0 7.18 16.9 91.5 1.28 168 39,418 1084				
Na Mg P K Ca Sc Ti V Cr Mn Fe Co Ni Ga Rb Cs B Li H ₂ O Cl F	6 15,486 14,218 265 721 518 214 18.9 336 971 63.8 2284 1.05 12.9 7.72 22.8 142 1.30 138 39,370 1172 24.4	7 15,435 12,669 327 781 537 134 28.9 262 761 74.1 1657 1.21 9.89 7.56 18.9 88.2 1.22 195 36,375 1241 31.0	8 15,871 13,775 393 622 502 211 21.7 349 905 73.2 1775 1.24 11.7 8.36 17.7 100 1.17 165 38,005 1094 15.7	9 16,013 14,460 196 415 581 199 24.9 337 919 75.0 2170 2.32 46.6 8.29 12.8 115 2.17 153 37,791 950 95.7	$\begin{array}{c} 10\\ \hline 14,841\\ 13,631\\ 341\\ 598\\ 432\\ 204\\ 20.0\\ 333\\ 885\\ 67.6\\ 1874\\ 0.99\\ 10.0\\ 7.05\\ 19.2\\ 100\\ 1.19\\ 194\\ 46,383\\ 1117\\ 15.9 \end{array}$	11 14,785 13,611 308 559 379 211 17.4 355 875 68.9 1734 1.13 10.4 7.53 17.1 97.0 0.91 156 37,725 978 14.4	12 13,801 13,647 298 895 444 155 33.9 299 710 70.5 1788 1.12 7.76 10.6 19.0 82.0 1.60 151 41,383 1052 18.8	$\begin{array}{c} \textbf{13} \\ \hline 14,161 \\ 14,766 \\ 179 \\ 389 \\ 464 \\ 164 \\ 18.9 \\ 336 \\ 750 \\ 74.0 \\ 2671 \\ 3.53 \\ 125 \\ 6.46 \\ 16.8 \\ 75.3 \\ 1.32 \\ 159 \\ 40,646 \\ 1058 \\ 118.0 \end{array}$	$\begin{array}{c} 14\\ 14,547\\ 13,647\\ 452\\ 405\\ 526\\ 148\\ 19.2\\ 316\\ 589\\ 70.1\\ 1729\\ 0.98\\ 9.74\\ 5.64\\ 14.0\\ 80.5\\ 1.16\\ 171\\ 38,748\\ 1059\\ 9.10\\ \end{array}$	$\begin{array}{c} 15\\ 13,029\\ 12,778\\ 558\\ 415\\ 482\\ 146\\ 16.4\\ 307\\ 691\\ 67.6\\ 1694\\ 1.00\\ 10.4\\ 5.85\\ 16.4\\ 70.6\\ 0.97\\ 180\\ 36,644\\ 1120\\ 8.18 \end{array}$	16 12,972 12,590 521 376 418 148 17.2 294 718 65.6 1656 0.96 10.2 5.86 14.8 75.4 1.19 163 38,850 1138 131.7	17 13,944 13,200 231 442 386 137 30.8 288 666 69.8 1558 0.93 7.91 6.12 14.5 78.5 1.10 177 38,579 1021 11.8	$\begin{array}{c} 18\\ 14,280\\ 13,563\\ 467\\ 504\\ 394\\ 136\\ 20.2\\ 297\\ 727\\ 68.8\\ 1653\\ 1.20\\ 11.4\\ 6.26\\ 15.1\\ 91.2\\ 1.75\\ 187\\ 42,093\\ 1125\\ 36.4 \end{array}$	19 15,120 14,316 563 527 487 154 20.5 322 707 77.4 1700 1.11 10.7 7.22 18.0 84.9 0.89 0.89 167 39,262 1048 9.02	Core 6-19 ** 14,592 13,634 364 546 468 169 22,1 317 777 70,4 1853 1.34 21.0 7.18 16.9 91.5 1.28 168 39,418 1084 38.6				

* Mean for bright-green rim (spots 1–5) and mean for pale-green rim (spots 20–25). ** Mean for brown-green core (spots 6–19).

4.2. Compositional Zoning of Beryl

The concentrations of representative trace and minor elements (in ppm) in the analyzed beryl crystal from Wadi Nugrus, South Eastern Desert, Egypt, are given in Table 1, and their spatial distribution is represented in Figure 4a–v. The chemical zoning is visualized through profile from the bright-green rim, through the brown-green core, to the pale-green rim (spots 1–25) as mentioned (Figures 3 and 4, Table 1).



Figure 4. Zoning of the distribution of trace and minor elements (ppm) in the analyzed beryl from Wadi Nugrus, South Eastern Desert, Egypt. Zoning profiles for: (**a**) Mg in (ppm); (**b**) Fe in (ppm); (**c**) Cr in (ppm); (**d**) V in (ppm); (**e**) Na in (ppm); (**f**) Cs in (ppm); (**g**) Rb in (ppm); (**h**) Mn in (ppm); (**i**) Ti in (ppm); (**j**) Sc in (ppm); (**k**) Ca in (ppm); (**l**) K in (ppm); (**m**) Li in (ppm); (**n**) Ni in (ppm); (**o**) Ga in (ppm); (**p**) Co in (ppm); (**q**) P in (ppm); (**r**) Sr in (ppm); (**s**) H₂O in (ppm); (**t**) F in (ppm); (**u**) Cl in (ppm); (**v**) B in (ppm).

The distribution of trace and minor elements is consistent with the color zoning of beryl. The two rims show higher concentrations of Cs, Na, Mg, Fe, Sc, V and Rb than the core, but they contain low content of Mn, Ca and Li (Figure 4, Table 1).

Mg content in both rims and core of the crystal has nearly U-shaped zoning (Figure 4a). The average values of Mg are 14,714, 15,279 and 13,634 ppm in the bright-green rim, pale-green rim and brown-green core, respectively. Mg value is 16,077 ppm at spot 5 and 12,590 ppm at spot 16 (Figure 4a, Table 1).

Fe content shows average values of 2257, 1980 and 1853 ppm, corresponding to the bright-green rim, pale-green rim and brown-green core, respectively. In the core of the zoned beryl crystal, Fe content reaches 2671 ppm at spot 13 and decreases to 1558 ppm at spot 17, showing bell-shaped zoning (Figure 4b, Table 1). However, the distribution of Fe in both rims does not change in the same way dramatically when compared with Mg, which has U-shaped zoning (Figure 4b).

Cr is identified as one of the main chromophore elements and has nearly asymmetric U-shaped zoning, with average content values of 771, 880 and 777 ppm, corresponding to the bright-green rim, pale-green rim and brown-green core, respectively.

The core is characterized by its uniform distribution of these trace and minor elements (Figure 4d–g,j). The bright-green rim shows high average values of 16,569, 239, 23.9 and 245 ppm for Na, Cs, Rb and Sc, respectively, while the pale-green rim shows a high average value of 404 ppm for V (Table 1). The distributions of V, Na, Cs, Rb and Sc have U-shaped zoning with marked enrichment in both rims.

The distribution of Mn is characterized by a bell-shaped array with a markedly enriched core. The average values of Mn content are 66.0, 64.9 and 70.4 ppm in the bright-green rim, pale-green rim and brown-green core, respectively. The highest values are 77 and 76 ppm, recorded at spots 19 and 23 (Figure 4h, Table 1).

The distribution of Ti is very complex and does not show clear zoning (Figure 4i). The more transparent edge (bright-green rim) is characterized by a high average value of 24.1 ppm for Ti, while the average values 17.7 and 22.1 ppm of Ti concentrations correspond to the pale-green rim and brown-green core (Table 1).

The distribution of Ca, in general, varies within color zoning crystals with a markedly enriched core with bell-shaped features (Figure 4k). Ca contents are not stable, fluctuating with a marked spike in contents followed by a decrease towards the boundaries. Average contents of Ca are 426, 355 and 468 ppm, corresponding to the bright-green rim, pale-green rim and brown-green core, respectively (Table 1).

The distribution of K differs along the zoned beryl crystal (Figure 41). It shows the highest value (895 ppm) at spot 12, showing a bell shape, followed by low contents at the crystal boundaries. The average values of K are 675, 462 and 546 ppm in the bright-green rim, pale-green rim and brown-green core, respectively (Figure 41, Table 1).

The distribution of Li shows higher contents in the core than in both rims and is bellshaped. The averages of Li are 142, 155 and 168 ppm in the bright-green rim, pale-green rim and brown-green core, respectively (Figure 4m, Table 1).

The concentrations of Ni, Ga and Co are lower than 14 ppm (Figure 4n–p, Table 1), with the exception of two spots (9 and 14) having 47 and 125 ppm of Ni. The distribution of Ni and Co shows a bell shape, followed by a decrease at the crystal boundaries.

The distribution of P is similar to Ca, having a bell shape with a more enriched core (average 364 ppm) than the two rims (245 and 185 ppm for bright-green and pale-green rims, respectively) (Figure 4q, Table 1).

Sr is distinguished by low contents and has a marked bell-shaped distribution at spot 12 (19 ppm) followed by a decrease in concentration at rims (Figure 4r, Table 1).

The distribution of volatiles (H₂O, Cl and F) does not show defined zoning (Figure 4s–u). The water contents have average values of 41,884, 41,315 and 39,418 ppm for the bright-green rim, pale-green rim and brown-green core, respectively. The same level of water content is well observed at spot 10 (core) and reached 46,383 ppm, whereas the F and Cl contents at spot 10 show marked depletion (Figure 4t,u). The Cl contents generally show higher average values in both rims than in core: 1097, 1103 and 1084 ppm for the bright-green rim, pale-green rim and brown-green core, respectively (Figure 4u, Table 1). On the contrary, the average contents of F have the highest value (38.6 ppm) in the core, whereas 18.4 and 9.42 ppm values correspond to the bright-green rim and pale-green rim (Figure 4t, Table 1). B distribution shows low contents, and zoning features are not manifested (Figure 4v).

4.3. Binary Correlation Diagrams Using Trace and Minor Elements

Based on the plots of binary diagrams, we can say that the content of the elements Na, Mg and V versus Cs increases from the center of the crystal to its edge. In this case, the edge parts are depleted in Mn, Li and F versus Cs when compared with the central zone (Figure 5a–f). The plot of the binary Fe–Mg diagram shows a noticeable positive correlation with an obvious enrichment of the edge zone of the crystal (Figure 5g).



Figure 5. Binary diagrams show the correlation of trace and minor elements (ppm) in beryl from Wadi Nugrus, South Eastern Desert, Egypt. Symbols are the same as in all of the following diagrams: red square for bright-green rim (spots 1–5), black triangle for pale-green rim (spots 20–25) and blue lozenge for brown-green core (spots 6–19) of beryl crystal. (a) Na vs. Cs; (b) Mg vs. Cs; (c) V vs. Cs; (d) Mn vs. Cs; (e) Li vs. Cs; (f) F vs. Cs; (g) Fe vs. Mg binary diagrams.

4.4. Biotite Inclusions

There is a noticeable large amount of mica in the crystals, especially biotite, distributed evenly over the zones of crystal growth. The central zone is enriched with inclusions compared to edge zones. This distribution of biotite also affects the crystal's color and adds a brown tint to the central zone (Figure 3a).

The distribution of trace and minor elements shows an obvious higher content in the core, especially at spots B1, B2 and B3 with values of 7606, 2143 and 1553 ppm for K; 24,199, 18,072 and 18,270 ppm for Mg; 6076, 3062 and 3029 ppm for Fe; and 448, 158 and 108 ppm for Ti, than in other areas, which could be a good indicator of biotite inclusions (Figure 6).



Figure 6. K, Mg, Fe and Ti as good indicators of biotite inclusion through zoning profiles of beryl from Wadi Nugrus, South Eastern Desert, Egypt.

5. Discussion

5.1. Petrogenetic Aspects of Beryl

The garnet muscovite granite and related pegmatites affected the pre-existing ultramafites of serpentinite and talc carbonates, metagabbros and metasediments (schists) along their contacts, forming mineralized contact zones enriched in rare-metal mineralization. Pegmatites are late magmatic stages which are derived from garnet muscovite granite enriched in uraninite, coffinite, uranothorite, thorianite, zircon, monazite, columbite, microlite, uranopyrochlore, xenotime, barite, apatite, galena and fluorite, as well as noble metal mineralization such as argentite, native Ni and Bi with beryl and tourmaline [46].

The serpentinite and talc carbonates have MgO contents of 43.8–45.7 wt% with an average of 44.8 wt%, Cr contents of 2426–2709 ppm with an average 2648 ppm and V contents of 14–42 ppm with an average 29 ppm, whereas the metagabbros reach up to 9.3 wt%, 281 ppm and 201 ppm for MgO, Cr and V, respectively, at Sol Hamed, South Eastern Desert of Egypt [66,67]. On the other hand, Be reached up to 604 and 1700 ppm in granitic pegmatites (albite or sodic aplite dike) in the Wadi Sikait and Wadi Nugrus areas, South Eastern Desert of Egypt [44]. BeO in green beryl from quartz tourmaline veins and

from phlogopite schist rocks ranges from 13.36 to 13.83 wt% [27] in Kafubu, Zambia. Be could be related to metasomatic transfer via interaction of granitic pegmatite fluids with its contact ultramafic rocks [44]. At the contact between pegmatites enriched in Be and serpentinite and related talc carbonates enriched in Cr and V, the metasomatic fluids can dissolve quartz as well as Cr and V that could be remobilized from serpentinite bearing chromite, and Be could come from pegmatites to form green emerald.

The successive intrusion of muscovite granite could help to form the nature of the emerald from Wadi Nugrus, with pegmatites as well as quartz-bearing beryl arising from exsolution of a fluid/pegmatite enriched in volatile-rich magma bearing H₂O, NaCl and CO₂, which is similar in situation to aquamarine genesis in Canada [68,69] (Figure 7).



Figure 7. Genetic model (modified after Groat et al. [68] and Beal and Lentz [69]) for green beryl (emerald) along the mineralized contact between metasediments and pegmatites at Wadi Nugrus, South Eastern Desert of Egypt.

The gem zone of emerald in the study area is developed from fluids: fluid (1) enriched by Mg, Cr and V from serpentinite and related talc carbonate and metagabbros with fluid (2) enriched in Be as well as F, Li, W, B, Nb, Ta, Zr, U and Th from muscovite granite and their pegmatite. The abundance of beryl within the contact zones of pegmatites with metasediments suggests that beryl is formed from two heterogeneous natural fluid circulations. So, the emerald present within certain veins could be formed from a hydrothermal fluid enriched in the elements that are necessary for emerald formation from the muscovite granite and related pegmatites during progressive crystallization with the pre-existing basement rocks of the study area.

The emerald is associated with intensive fluid–rock interactions, including K- and Na-metasomatism of granitic pegmatites. This type of genetic pathway emerald is mostly common worldwide [70–73].

5.2. Trace and Minor Element Geochemistry as a Guide for Paragenetic Beryl Mineralization

The binary correlation diagram between the alkaline+light elements and transition elements + Ca + Mg shows a positive correlation trend in the analyzed beryl from browngreen core to bright-green rim. The bright-green rim shows higher contents in alkaline+light elements than do the other spots of brown-green core and pale-green rim. It obviously shows that each crystal zone's chemical composition has an individual trend (Figure 8).



Figure 8. Binary diagram shows the correlation between transition+Ca+Mg and alkaline+light elements in the analyzed zoning of beryl from Wadi Nugrus, South Eastern Desert, Egypt. Symbols are the same as in Figure 5: red square for bright-green rim (spots 1–5), black triangle for pale-green rim (spots 20–25) and blue lozenge for brown-green core (spots 6–19) of beryl crystal.

The Na/Li and Li/Cs relationship of alkalis in beryl could be a good indicator to reflect the geochemical features and genesis of mineral-forming fluid [13]. Neiva and Neiva [74] mention a decrease in both ratios during fractionation. Cs/Na, on the other hand, is a direct measure of fractionation [75] and should correlate negatively with Mg/Fe along with the fractionation trend. The binary diagram of Na/Li versus Li/Cs (Figure 9a) shows that higher Na/Li characterizes the bright-green rim with a near-constant Li/Cs value, whereas analyses show a slight increase in Li/Cs values with a decrease in Na/Li values characterizing the pale-green rim to the brown-green core. The Na/Li–Li/Cs diagram shows an obvious negative correlation towards the brown-green core (Figure 9a). During fluid evolution, Na and Cs increased from brown-green core to bright-green rim and may be responsible for the color zoning of the analyzed beryl. Na-metasomatism (albitization) could affect the high Na and Cs contents in rims [23,44].

The Mg/Fe ratio is an indicator for the rock's chemical composition in which the mineral-forming fluids have interacted, and the alkalis characterize the chemical composition of these fluids. Thus, the fluids could be evolved to different degrees that are responsible for the formation of beryl mineralization. The Mg/Fe ratio is high in the brown-green core with constant Cs/Na (Figure 9b). The binary diagram shows a marked negative correlation towards the bright-green rim (Figure 9b).

The binary diagram of the Cr/V ratio versus Fe is represented in Figure 9c. All the analyzed beryl shows that the Cr/V ratio varies between 1.51 and 2.91, while Fe varies between 1558 and 2670 ppm. The distribution of the analyzed beryl from Wadi Nugrus is similar to emerald from Afghanistan (Panjsher Valley), which is characterized by Cr/V ratios that vary between 0.83 and 2.64 and Fe values from 781 to 2530 ppm [18].

The emerald deposits of Swat Valley, Santa Terezinha de Goiás and Habachtal have formed during the interaction of metamorphic rocks with magmatic fluids, with high Cs concentration in emeralds from the Santa Terezinha. The binary diagram between Na/Li and Cs/Ga ratios [76] strengthens this hypothesis. Figure 10 shows that beryl from Wadi Nugrus, Egypt, occurs in the same chemical field as the emerald deposits from Swat Valley, Santa Terezinha de Goiás and Habachtal.



Figure 9. (a) Na/Li vs. Li/Cs ratios, (b) Mg/Fe vs. Cs/Na ratios and (c) Fe vs. Cr/V ratio plots in the analyzed beryl from Wadi Nugrus. Symbols are the same as in Figure 5: red square for bright-green rim (spots 1–5), black triangle for pale-green rim (spots 20–25) and blue lozenge for brown-green core (spots 6–19) of beryl crystal.



Figure 10. Na/Li vs. Cs/Ga binary discrimination diagram indicates the relationships between beryl from Wadi Nugrus, South Eastern Desert, Egypt, and different localities worldwide (Russia, Nigeria, Colombia, Santa Terezinha, Habachtal and Swat Valley) according to [76]. The diagram shows that the chemical ratios of Cs/Ga and Na/Li of emerald from Santa Terezinha de Goiás (Brazil), Swat Valley (Pakistan), and Habachtal (Austria) fall in the same field. Symbols are the same as Figure 5: red square for bright-green rim (spots 1–5), black triangle for pale-green rim (spots 20–25) and blue lozenge for brown-green core (spots 6–19) of beryl crystal.

The studied beryl is characterized by a high Cs/Ga ratio with a mean of 31.0, 12.9 and 26.8 for bright-green rim, brown-green core and pale-green rim, respectively. It is similar to Swat Valley, Santa Terezinha de Goiás and Habachtal emerald deposits, indicating appreciable Cs contents (pointing to a magmatic source) and a high Na/Li ratio related to higher Na concentration [58]. The analyzed beryl contains high mean H₂O contents of 4.19, 3.94 and 3.86 wt% for bright-green rim, brown-green core and pale-green rim, respectively. This feature of high mean H₂O content is similar to that determined for Habachtal (3.1 wt.%, n = 3), Santa Terezinha de Goiás (2.9 wt.%, n = 5) and Swat Valley (3.4 wt.%, n = 1) [17,70,77]. This could be probably a genetic Be–Cs source for emerald in these deposits, i.e., magmatic intrusions with huge fluid circulation indicative of metasomatism in a metamorphic environment (schist in Wadi Nugrus).

The Fe–Rb, Fe–Cs, Rb–Cs and Li–Fe discrimination diagrams (Figure 11) were used to differentiate between emeralds from Wadi Nugrus, South Eastern Desert, Egypt, and those worldwide [78]. The analyzed beryl spots from the Wadi Nugrus fall into the field of Russia (Urals) using Fe–Rb; straddle the boundary line between the fields of Afghanistan, Ethiopia and Russia (Urals) using Fe–Cs; and gravitate towards the fields of Russia (Urals), Brazil and Ethiopia using Rb–Cs and the fields of China and Afghanistan using Li–Fe.



Figure 11. Fe vs. Rb, Fe vs. Cs, Rb vs. Cs and Li vs. Fe binary discrimination diagrams show the relationships of trace and minor elements (ppm) between beryl from Wadi Nugrus, South Eastern Desert, Egypt, and beryl from various localities after Saeseaw et al. [78]. Compositional fields of beryl from various occurrences: 1—Colombia; 2—China; 3—Brazil; 4—Zambia; 5—Afghanistan; 6—Madagascar; 7—Ethiopia; 8—Russia (Urals). Symbols are the same as Figure 5: red square for bright-green rim (spots 1–5), black triangle for pale-green rim (spots 20–25) and blue lozenge for brown-green core (spots 6–19) of beryl crystal.



Figure 12a–c shows the elemental average content values of emerald from Wadi Nugrus (Egypt) and worldwide according to Aurisicchio et al. [58].

Figure 12. (a) Chromophore and transition element (CTE) vs. large ion lithophile element (LILE), (b) Cr vs. V and (c) B vs. Li binary diagrams show a comparison between beryl from Wadi Nugrus and beryl from other locations worldwide according to Aurisicchio et al. [58], using average trace and minor elements (ppm) of the analyzed emeralds.

These binary diagrams present the relations of the chromophore and transition elements (CTEs) (Cr and V) versus large ion lithophile elements (LILEs) (Rb, Ba, K, Sr, Cs), Cr versus V and B versus Li.

All emeralds from the studied area and worldwide have low concentrations of Ba, Sr and B. Emerald from Wadi Nugrus (Egypt) has an average value of 714 ppm for LILEs, which is higher than the values reported for emeralds originating from Colombia, Egypt (Sikait), Austria, Pakistan, Russia, Nigeria, Egypt (Zabara), Australia and Tanzania, according to Aurisicchio et al. [58] (Figure 12a). The CTEs in emerald from Wadi Nugrus (Egypt) show higher concentration (reaching 1149 ppm) than those from Mozambique, Austria, Zambia, Russia, Nigeria and Australia, according to Aurisicchio et al. [58] (Figure 12a). However, emeralds from Mozambique show the highest LILE content (with an average of 2436 ppm) and lower CTE content (with an average of 365 ppm). In contrast, emeralds from Pakistan display the highest CTE content (average 7068 ppm) and a lower content of LILEs (average 238 ppm) (Figure 12a).

The average contents of emerald from Wadi Nugrus (Egypt) show a higher concentration of V (349 ppm) and Cr (800 ppm) than those from Australia, Zambia, Mozambique and Russia, according to Aurisicchio et al. [58] (Figure 12b). On the other hand, emeralds from Pakistan present the highest Cr content (average 6635 ppm) and a low V content (average 433 ppm), while those from Colombia exhibit the highest V content (average 4543 ppm) and an average Cr content of 1591 ppm, according to Aurisicchio et al. [58] (Figure 12b).

Emeralds from Wadi Nugrus show a higher concentration of B (average 1.35 ppm) than those worldwide according to Aurisicchio et al. [58], except those from Egypt (Zabara) and Zambia with average B concentrations of 3.09 and 3.73 ppm, respectively (Figure 12c). Emeralds from Wadi Nugrus display a higher concentration of Li (average 160 ppm) than those from Egypt (Sikait), Egypt (Zabara), Madagascar, Nigeria, Tanzania, Afghanistan, Brazil and Colombia, according to Aurisicchio et al. [58]. Although emerald from Zimbabwe shows the highest Li content (average 983 ppm), it contains B in lower concentrations (average 0.46 ppm), according to Aurisicchio et al. [58] (Figure 12c).

6. Conclusions

- 1. Beryl from Wadi Nugrus shows high contents of large ion lithophile elements (LILEs), especially Rb, K and Cs, but it has low concentrations of Sr and B.
- Cs, Na, Mg, Fe, Sc, V, Rb and H₂O show a U-shaped pattern in the zoned emerald, indicating markedly enriched rims compared with the core, whereas Mn, Ca, Co, Sr and Li have bell-shaped zoning with markedly enriched core compared to both rims. On the other hand, Cr, Ti, K, Ni, Ga, F, Cl and B have no marked zoning (or very complex zoning).
- 3. The bright-green rim exhibits higher contents of Na, P, K, Ca, Fe, Rb, Cs and F than the pale-green rim but contains Mg and Li in low concentrations. The alkaline elements (Cs, Na, Rb) and Fe are responsible for the color zoning.
- 4. Granites and related pegmatites and aplite dikes are considered good sources for Be, which interacted with hydrothermal fluids enriched in Cr, V, Sc, Mg and Ca from the pre-existing ultramafic and mafic rocks in addition to fluids bearing water, NaCl and CO₂.
- 5. Emerald from Wadi Nugrus displays a higher concentration of LILEs than the emeralds from Colombia, Egypt (Sikait), Austria, Pakistan, Russia, Nigeria, Egypt (Zabara), Australia and Tanzania, while the chromophore and transition elements (CTEs) (Cr and V) have higher contents in the studied emerald than in emeralds from Mozambique, Austria, Zambia, Russia, Nigeria and Australia.

Author Contributions: Conceptualization, A.E.A.G., S.G.S. and M.A.A.; methodology, S.G.S. and A.K.G.; software, A.E.A.G., S.G.S. and A.K.G.; validation, A.E.A.G., S.G.S. and M.A.A.; formal analysis, S.G.S., A.E.A.G. and A.K.G.; investigation, A.E.A.G. and M.M.G.; resources, M.A.A. and M.M.G.; data curation, S.G.S., A.E.A.G. and A.K.G.; writing—original draft preparation, A.E.A.G.;

writing—review and editing, A.E.A.G., A.E., S.G.S., A.K.G., M.A.A. and M.M.G.; visualization, S.G.S. and A.E.A.G.; supervision, S.G.S., A.E., M.A.A. and A.V.N.; project administration, S.G.S., A.K.G., A.E., A.E.A.G., M.A.A. and M.M.G.; funding acquisition, A.E. All authors have read and agreed to the published version of the manuscript.

Funding: Research of beryl geochemistry was partly supported by a State contract of the Institute of Precambrian Geology and Geochronology of the Russian Academy of Sciences (No. FMUW-2022-0005). The article processing charge (APC) was covered by "Dunarea de Jos" University of Galati, Romania.

Data Availability Statement: Data are contained within the article.

Acknowledgments: We thank Evgeny Potapov and Sergey Simakin who assisted with measuring beryl trace and minor element compositions. The authors Antoaneta Ene and Ahmed Abdel Gawad acknowledge the support of "Dunarea de Jos" University of Galati, Romania, through grant No. RF3621/2021.

Conflicts of Interest: The authors declare no conflict of interest.

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