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Interpretation of sedimentary processes using heavy mineral unconstrained data

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Abstract: This work describes and interprets the presence of heavy minerals in the western Portuguese continental margin using a set of 78 bottom samples collected from 3 distinct areas of this margin: Porto, Aveiro and Nazaré canyon head areas. The main transparent heavy mineral suite (minerals with frequencies >10%), is composed by amphiboles (hornblende), mica (biotite), andalusite, tourmaline and garnet. A secondary suite (minerals with frequencies between 1 and 10%), is composed by pyroxene (enstatite, diopside and augite), staurolite, zircon and apatite. With very low frequency representing less than 1% we found rutile, olivine, kyanite, monazite, epidote, sphene, anatase, sillimanite and brookite. The main primary sources (igneous and metamorphic rocks) explain the presence of these minerals. However, the application of the principal component analysis, with a previous application of the centered log ratio transformation of the heavy mineral data, also stresses for the importance of the grain sorting as a process in controlling the heavy mineral occurrence. The importance of this process is mostly sustained by the distribution pattern of mica and of the most flattened amphibole grains in a way that these particles tend to have a hydraulic affinity to finer grained sediments.

Keywords: sand particles; geological sources; grain sorting

Graphical abstract. Main heavy minerals: Am-amphibole, Mi-mica, And-andalusite, To-tourmaline, Ga-garnet, Py-pyroxene, St-staurolite, Zi-zircon and Ap-apatite.

1. Introduction

Sedimentology encompasses several areas of expertise including the knowledge of the mineral composition as a tool to understand the sediment provenance [1], from where the heavy minerals emerge as one of the most widely-used techniques [2]. However, to achieve a good estimate of a heavy mineral population hundreds of these mineral particles must be identified and counted in each sample [3,4], which is a high time-consuming routine operation. Therefore, the study of these minerals should be considered a way to increase our knowledge about the sedimentary processes. Many works that use heavy minerals to interpret the sedimentary processes are based on quantified mineral abundances expressed by relative percentages that were computed from mineral counting values [5-7]. However, the interpretation of a heavy mineral suite present in a sedimentary deposit faces several drawbacks. Some of them are derived from natural causes that control the mineral occurrence patterns, such as, the mineralogical composition of the source region or the processes that operate during sedimentary cycles. Even when these drawbacks are known and controlled, we must count on the relationship between sediment grain size and the natural affinity of these minerals to appear more concentrated in certain grain sizes, due to the mineralogical characteristics of the primary source rocks and to the grain sorting process [8-10]. Another difficulty may arise when we apply standard methods of multivariate analysis to a mineral data set expressed in relative abundances ignoring the constant-sum constraint effect [11-12].

Using a set of 78 sea bottom sediment samples collected from the Porto, Aveiro and Nazaré canyon upper heads (Figure 1) we firstly aim to identify the transparent heavy mineral spectrum using the polarized light microscope and, in a complementary way, the electron microprobe analysis. Secondly, we intend to evaluate the relative importance of the main processes in controlling the mineral occurrence by the application of a multivariate statistical method (principal component analysis), avoiding the constant-sum constraint effect by the previous application of the centered log-ratio transformation [12]. Following this method of data analysis, we pretend to distinguish different mineral sources based on the mineralogical composition and on the main grain morphological features. We also intend to evaluate the importance of the grain sorting in the modification of the original mineral signatures of the sources.

Figure 1. Samples location according the 3 sampled areas: Porto, Aveiro and Nazaré canyon head areas.

2. Geological setting

Porto, Aveiro and Nazaré canyons are visible geomorphologic features of the WNW Portuguese continental margin on any bathymetric map (e.g. on the Google Ocean Map). However, while Porto and Aveiro canyons are considered as minor submarine valleys as they weakly indent the shelf [13], the Nazaré canyon is one of the largest canyons of the European Margin (170 km) because it cuts the entire width of the Portuguese Margin, from the Iberia Abyssal plain (at 5000 m depth) until the infralittoral zone off the Nazaré beach [14]. The geological nature of canyon heads surrounding area is also different: Porto canyon is craved in carbonated to detrital rocks highly dolomitized dating from Paleocene; Aveiro canyon is carved in biogenic and detrital limestones rocks dating from Neogenic and Eocenic; Nazaré canyon is craved on Mesozoic rocks [14,15].

The sedimentary cover of the referred canyon head surrounding areas is mainly made by sand with the presence of some other deposits richer in gravel or silt particles [16]. The Porto area reveals a higher grain size variability, ranging between sand and gravel at shallower depths (less than 100m) until fine sediment particles (silt and clay) well represented at middle shelf (Douro muddy deposit) and upper slope, where in some isolated spots of these finer particles can reach up to 70% of the sediment total weight [17]. The Aveiro area reveals a more homogenous sedimentary cover where the sand is the dominant textural type representing always more than 60% of the total sediment. In some small areas, between 100 and 150m depth, the gravel particles can represent up to $\frac{1}{3}$ of the total sediment. Finer sediments are only important in some small areas of the upper continental slope with almost 30% of the total sediment weight [16]. The shelf sedimentary cover near to the Nazaré canyon is in some locations dominated by coarse-grained particles (sandy gravel) namely at 40 to 80m depth. At those depths these particles constitute a sedimentary deposit with a geometry sub-parallel to the coast line orientation (paleo littorals). Fine and very fine sands are recorded in the inner shelf north of the canyon and close to its head. Additionally, two important muddy deposits areas are present in the middle shelf north and south of this canyon, at approximately 100 m depth [18,19].

3. Materials and Methods

The present work is based on a set of 78 bottom samples, collected from 3 different areas of the northern Portuguese continental margin, that match the Porto (30 samples), Aveiro (26 samples) and Nazaré (22 samples) submarine canyon upper heads (Figure 1). These samples were collected during several cruises during the decades of 1990/99 and 2000/09, using Smith-McIntyre grab on board hydrographical vessels (*Almeida Carvalho*, *NRP D. Carlos I*, *Andrómeda* and *Auriga*), within the scope of the Portuguese Instituto Hidrográfico program of cartography of the continental shelf sediments (SEPLAT), Sedimentary Dynamics of the Northern Portuguese Continental Shelf project (DISEPLA II), Hotspot Ecosystem Research on the Margins of European Seas project (HERMES) and on the Sedimentary Conduits of the West-Iberian Margin project (DEEPCO).

All the samples were first washed using hydrogen peroxide and distilled water to eliminate organic matter and marine salts. Grain-size analysis was done using the classic sieving method for sediments coarser than 0.5mm (at 0.5 ϕ interval) and the laser granulometer (Malvern 2000) for sediments finer than 0.5mm (1 ϕ). The textural statistical parameters (mean and sorting) were computed using the method of moment [20]. Heavy minerals were separated from medium (1 ϕ or 0.500mm) to very fine sand (4 ϕ or 0.063mm) using bromoform and then mounted in Canada balsam on glass slides. The required amount of heavy minerals to fill an area of 25x30mm of each slide without grain overlapping was obtained using a micro-splitter. An average of about 380 transparent heavy minerals per sample were counted under the petrographic microscope according to the line counting method [21]. Additionally, to confirm the identities of some specific heavy minerals less easily identified with a standard petrographic microscope (pyroxenes and olivines) some specific grain mounts were made using epoxy resin polished with *silicon carbide (sic)* and *diamond polishing in polishing cloths*. These grain mounts were then analyzed by an electron microprobe (JEOL Superprobe 733 at Lisbon University).

From the heavy mineral optical identification, we obtain a data matrix arranged in a way that each sample (observation) is a positive vector described by several proportional variables (minerals), as follows:

$$\begin{cases} x_i \geq 0, \text{ for all } i=1, \dots, D \\ \sum_{i=1}^D x_i = k, \quad k = 100\% \end{cases} \quad (1)$$

Where x_i denotes a value present in any matrix cell and k represents the counting results expressed in relative frequencies (sum of an entire line). This means that each sample is characterized by several minerals whose relative frequencies give rise to a constant sum of 100%. This inevitable arithmetical limitation implies that this data is constrained, in the sense that any relative value is not free to vary independently. This aspect is critical when we intend to analyze the data using common statistical methods designed for unconstrained data, as for example the PCA analysis [11,12]. To overcome this, we use the centred log-ratio transformation (*clr*) of our heavy mineral data [12]. Since this logarithmic transformation is incompatible with the presence of zeros (termed as “count zeroes”) we need to substitute those zeros. For this substitution we apply the Bayesian-multiplicative imputation method [22], which substitutes the zeroes with the concomitant preservation of the ratios between the non-zero parts [23]. This imputation method is implemented in the package *zCompositions* [24] running in R programming language. This imputation method was implemented using the mineral count data, that is, the data matrix containing the number of mineral grains counted in each sample, considering only the major transparent heavy mineral species, that is the suite composed by minerals with mean frequencies $\geq 1\%$ (Table S1). To replace the zeros of Table S1 the data (xlsx Excel format) is firstly converted into an RData file using the *CoDaPack* software [25]. After this transformation it is possible to apply the instructions for zeros replacement of the mineral count data matrix (Rcode S2). After running the R program, we obtain a new data matrix without zeros (Table S3). This new matrix was submitted to the *clr* transformation, according to the following formulas:

$$clr(x) = \left[\ln \frac{x_1}{g(x)}; \dots; \ln \frac{x_D}{g(x)} \right] \quad (2)$$

$$g(x) = \sqrt[D]{x_1 \dots x_D} \quad , \quad (3)$$

where $clr(x)$ it is the centered log-ratio transformation, $g(x)$ represents the geometric mean and x_D represents the individual relative frequency of each heavy mineral different from zero. The *clr* transformation can be done using the free software *CoDaPack*, that easily import the original data from an excel spreadsheet [25,26]. Alternatively, this transformation can also be done using an excel spreadsheet with the application of equations 2 and 3. The data matrix with the *clr* transformation can be seen on Table S4. This matrix is later used for the principal component analysis.

4. Results

4.1. Sediment texture (mean and sorting)

The mean grain size of the sampled sediments (correspondent to the 78 samples) lies between 2.08ϕ (fine sand; Aveiro canyon) and 3.96ϕ (very fine sand; Nazaré canyon) while the sorting lies between 1.58ϕ (poorly sorted; Nazaré canyon) and 2.01ϕ (very poorly sorted; Porto canyon). A more detailed analysis of the textural data reveals that the medium and fine sand are the dominant classes of the Porto and Aveiro canyon head areas and, in a different way, the Nazaré canyon head area denotes the presence of more heterometric sediment, from very coarse sand to medium silt (Table 1). The details of the grain size values of each sample analyzed can be seen on Table S5.

Table 1. Average values, maximum and minimum of mean and sorting. Values in ϕ units for the 3 canyon head areas (# = number of samples.)

Canyon	#	Mean (average)	Mean (maximum)	Mean (minimum)	Sorting (average)	Sorting (maximum)	Sorting (minimum)
Porto	30	2.26	5.24	1.12	2.01	3.24	0.82
Aveiro	26	2.08	3.65	0.70	1.88	2.76	0.81
Nazaré	22	3.96	6.28	-0.25	1.58	3.03	0.43

4.2. Heavy mineral identification (global results)

Under the petrographic microscope it was possible to identify the presence of 18 species that can be mentioned in descending order of their mean frequency in the 78 analyzed samples: amphibole (Am), mica (Mi), , andalusite (And), tourmaline (To), garnet (Ga), pyroxene (Py), staurolite (St), zircon (Zi), apatite (Ap), rutile (Ru), olivine (Ol), monazite (Mo), kyanite (Ky), epidote (Ep), sphene (Sp), anatase (Ana), silimanite (Si) and brookite (Br). For the 78 studied samples almost 30,000 transparent minerals were identified. The global results of the relative frequency of these minerals are represented on Table 2. These results (in count values) are displayed in Table S6 as a data matrix with 78 rows (samples) and 18 columns (minerals).

Table 2. Heavy mineral relative frequency considering all the analyzed samples (optical identification). Mean: mean frequency for each transparent heavy mineral (values in % referred to the total transparent heavy minerals). Max: higher frequency of each transparent heavy mineral. Min: lower frequency of each transparent heavy mineral. Heavy Minerals (HM): amphibole (Am), mica (Mi), andalusite (And), tourmaline (To), garnet (Ga), pyroxene (Py), staurolite (St), zircon (Zi), apatite (Ap), rutile (Ru), olivine (Ol), kyanite (Ky), monazite (Mo), epidote (Ep), sphene (Sp), anatase (Ana), silimanite (Si) and brookite (Br).

HM	Mean	Max	Min
Am	18.4	39.9	2.6
Mi	18.0	88.9	0.0
And	16.6	44.8	1.0
To	14.2	53.1	0.8
Ga	13.6	28.9	0.0
Py	6.1	38.0	0.0
St	5.2	13.3	0.0
Zi	2.9	11.5	0.0
Ap	1.0	8.4	0.0
Ru	0.8	4.0	0.0
Ol	0.6	3.6	0.0
Ky	0.6	2.4	0.0
Mo	0.5	3.7	0.0
Ep	0.4	3.4	0.0
Sp	0.3	2.4	0.0
Ana	0.3	2.7	0.0
Si	0.3	1.5	0.0
Br	0.2	1.7	0.0

The presence of Px and Ol minerals was further confirmed by the electronic microprobe analysis. This analysis validates the presence of 16 Py and 10 Ol grains. These grains are only present at Porto and Aveiro areas. Among the Py grains 6 are diopside, 6 are augite and the remaining 4 were classified as enstatite while the 10 Ol grains are forsterite (Table 3 and Figures 2 and 3).

Table 3. Mean chemical composition of the 16 Py and 10 Ol grains analysed. The numbers between brackets indicate the number of grains analysed.

	diopside (6)	augite (6)	enstatite (4)	forsterite (10)
SiO ₂	51.93	48.94	47.79	39.73
TiO ₂	0.37	0.82	0.06	0.02
Al ₂ O ₃	2.92	7.48	6.09	0.03
Cr ₂ O ₃	0.18	0.07	0.01	0.05
FeO	9.86	14.77	27.63	15.55
MnO	0.26	0.29	0.82	0.22
MgO	12.72	12.40	15.58	44.23
NiO	-	-	-	0.17
CaO	21.18	13.47	0.72	0.27
Na ₂ O	0.44	1.00	0.00	-
K ₂ O	0.00	0.15	0.01	-

Figure 2. Py grains composition in the system CaSiO₃-MgSiO₃-FeSiO₃. Dp – diopside, Hd – hedenbergite, Au – augite, En – enstatite, Fs - ferrosilite

Figure 3. Ol grains composition in the system Ca₂SiO₄-Mg₂SiO₄-Fe₂SiO₄ .Fo – forsterite, Fa – fayalite

4.3. Main heavy minerals

The main heavy mineral transparent suite is composed by Am, Mi, And, To, Ga, Py, St, Zi and Ap that correspond to the minerals with mean frequencies $\geq 1\%$ (Figure 4A). Nevertheless, each area has a specific main mineral signature. Porto canyon head is characterized by a main mineral suite composed by Am, Mi, And, To, Ga, Py, St, Zi and Ap (Figure 4B). Aveiro contains a most representative heavy suite made by Am, And, To, Ga, Py and St (Figure 4C). Nazaré area is dominated by the presence of Mi, Am, And, To, Ga and Zi (Figure 4D).

Figure 4A-4D. Heavy mineral transparent frequencies for all samples (A) and for each studied area (B to D). The box-plots represent the 25th and 75th quartiles. The horizontal line represents the 50th quartile (median). The small cross represents the mean value. The extremes of vertical line represent the maximum and minimum values.

4.4. Data manipulation and principal component analysis (PCA)

From Table 2 it is possible to conclude that the main heavy mineral assemblage is represented by the species from Mi to Ap, that is, the one's with a mean frequency equal or above 1 %. The other minerals (from Ru to Br) represent the accessory assemblage with extreme low frequencies. These accessory minerals have a random pattern of occurrence and in virtue of this they induce a considerable "noise" when we pretend to interpret the results. To avoid this "noise" the correct procedure is to eliminate the frequencies associated to these minerals and recalculate the other frequencies to 100%. The results obtained after this recalculation are represented on Table 4. These results (in relative frequencies) are displayed in detail in Table S1 as a data matrix with 78 rows (samples) and 9 columns (minerals).

Table 4. Main heavy mineral relative frequency (9 species) considering all the analyzed samples (optical identification). Mean: mean frequency for each transparent heavy mineral. Max: higher frequency of each transparent heavy mineral. Min: lower frequency of each transparent heavy mineral. Heavy Minerals (HM): amphibole (Am), mica (Mi), andalusite (And), tourmaline (To), garnet (Ga), pyroxene (Py), staurolite (St), zircon (Zi) and apatite (Ap).

HM	Mean	Max	Min
Am	19.3	41.3	2.6
Mi	18.2	89.5	0.0
And	17.4	48.2	1.0
To	14.9	53.2	0.8
Ga	14.3	30.4	0.0
Py	5.4	13.4	0.0
St	6.4	40.2	0.0
Zi	3.0	12.2	0.0
Ap	1.0	8.9	0.0

Using the CoDaPack we can make the *clr* transformation and the PCA analysis simultaneously. First, on the “File” menu we import the excel file with the data matrix of Table S3 and the CoDPack spreadsheet display the data according to the initial format of the excel spreadsheet (data matrix with no zeros). Then we choose on “Graphs” menu the “CLR biplot” option and select on the “Available Data” menu all the variables available as “Selected data”. To identify the sample groups, we must choose the option “ID” on “Groups” window. Following this operation, we obtain the “Biplot3d” diagram that can be viewed according the most important principal components (with the higher explained variance associated) with the inclusion of *clr* vectors for each heavy mineral. We can choose the most favorable orientation of the “Biplot3d” diagram to achieve the better visualization of these vectors on a 3D graph chart defined by the first three components that are simultaneously computed during this operation (Figure 5).

Figure 5. Biplot 3D *clr* diagram considering the first three principal components that explain more than 90 % of the data variance. Each color dot represents one sample. P (Porto; blue dots), A (Aveiro; red dots) and N (Nazaré; green dots). The most important *clr* vectors (longer vectors) are the ones of *clr.Mi*, *clr.Ap* and *clr.Py*.

As it was previously said the principal components are also extracted during the construction of the Biplot diagram and the results of the mineral loadings on the components that represent most of the explained variance are visible on Table 5 and Figure 6.

Table 5. Mineral loadings on the 8 extracted principal components (PC1 to PC8) that together explain 100% of variance. Var.: explained variance for each principal component. Σ Var: sum of the explained variance for each principal component.

	Am	Mi	And	To	Ga	Py	St	Zi	Ap	Var.	Σ Var.
PC1	0.05	0.79	0.05	0.03	-0.26	-0.53	-0.12	-0.07	0.05	53.56	53.56
PC2	-0.34	-0.30	0.01	0.02	0.00	-0.50	0.12	0.35	0.64	26.08	79.64
PC3	0.07	0.13	-0.15	-0.35	-0.19	0.43	-0.32	-0.28	0.66	11.78	91.42
PC4	-0.16	0.25	-0.40	-0.32	0.25	0.15	-0.27	0.68	-0.16	3.41	94.83
PC5	0.51	-0.25	0.36	-0.13	-0.51	-0.05	-0.29	0.43	-0.07	2.28	97.11
PC6	-0.55	0.14	0.07	0.25	-0.62	0.39	0.21	0.18	-0.07	1.60	98.71
PC7	0.24	0.04	-0.25	-0.52	-0.21	-0.05	0.75	0.03	-0.03	0.81	99.52
PC8	-0.35	0.09	0.71	-0.55	0.20	0.03	0.02	-0.06	-0.09	0.48	100.00

Figure 6. Plot of principal component loadings for first three components that together explain more than 90% of variance.

To avoid the effect of the Mi flakes that tend to be concentrated in the coarser part of the sediment grain size distribution curve due to their hydraulic equivalence [10] we proceed with a simplification of data matrix of Table S3. This simplification corresponds to the elimination of the Mi frequency choosing the operation of “Subcomposition/Closure” from “Data” menu of the CodaPack software. During this operation we select all the minerals of Table S3 (except Mi) and choose the option “Closure” equal to 1 in the “Options” menu. The projection of the Biplot diagram show a clear separation between the sampled areas according to the first 3 extracted components (Figure 7).

Figure 7. Biplot 3D *clr* diagram considering the first three principal components that explain about 90% of the data variance. Each color dot represents one sample. P (Porto; blue dots), A (Aveiro; red dots) and N (Nazaré; green dots). The most important *clr* vectors (longer vectors) are the ones of *clr.Am*, *clr.Ap* and *clr.Py*. This diagram results from the closure operation choose on the CodaPack software.

The principal components are also extracted during this operation and the results can be represented for a better visualization of the principal component loadings. The results show that the first 3 main components explain about 90% of the data variance composed by 8 initial variables or minerals (Table 6 and Figure 8).

Table 6. Mineral loadings on the 7 extracted principal components (PC1 to PC8) that together explain 100 % of variance. Var: explained variance for each principal component. Σ Var.: sum of the explained variance for each principal component.

	Am	And	To	Ga	Py	St	Zi	Ap	Var.	Σ Var.
PC1	0.16	-0.10	-0.10	0.15	0.75	-0.05	-0.26	-0.55	51.70	51.70
PC2	-0.32	-0.29	-0.38	0.04	0.45	-0.15	0.00	0.66	23.12	74.82
PC3	0.61	0.23	-0.02	-0.41	0.03	-0.29	-0.47	0.32	15.08	89.90
PC4	-0.36	0.07	0.32	0.10	-0.01	0.46	-0.72	0.14	4.83	94.73
PC5	0.32	-0.27	-0.18	0.77	-0.32	-0.16	-0.25	0.08	2.91	97.64
PC6	-0.28	0.13	0.59	0.18	0.08	-0.72	-0.01	0.03	1.41	99.05
PC7	0.24	-0.80	0.49	-0.22	0.03	0.11	0.07	0.07	0.95	100.00

Figure 8. Plot of principal component loadings for first three components that together explain about 90% of variance.

5. Discussion

5.1. Sedimentary processes

In the Porto area the main mineral association is composed by Am, Mi, And, To, Ga, Py, St, Zi, and Ap, where And is the mineral with the higher mean frequency. In the Aveiro area the main mineral association is composed by Am, And, To, Ga, Py and St, where Am is the mineral with the higher mean frequency. In the Nazaré area the main mineral association is composed by Mi, Am, And, To, Ga and St where Mi is the mineral with the higher mean frequency (Figures 4B to 4D). These mineral associations are the result of the interplay between the available sources and the grain physical sorting which are considered as key processes that determine the presence of heavy minerals in a detrital sedimentary deposit [2]. To know the relative importance of these processes we must provide significant insight into the structure of our data matrix. This was done by the application of the principal component analysis (PCA) where the original data was previously changed by the centered log-ratio transformation (*clr*) [7,11,12,27]. The 1st principal component explains a considerable proportion of the total variance (more than 50%; Table 5) and displays the opposite loadings of Mi relatively to Px being the other minerals almost neutral or with residual loadings (<0.3) (Figure 6). This component illustrates, above all, the hydraulic affinity of the platy mica grains from medium to very fine sand with finer particles. In fact, the presence of Mi according to the biplot diagram (Figure 5) is associated to the Nazaré area where several samples have a mean grain size of very fine sand to coarse silt (Table 1). This agrees with the knowledge of the hydraulic behavior of Mi grains in sand sediments known since the 1960's [28-32]. According to our data the high frequency of Mi (>40%) occurs only when the sediment has a mean grain size higher than 2.5 ϕ , that is, when the presence of fine to very coarse silt is very important. Moreover, when the Mi frequency is extremely high (Mi >60%) the sediment mean grain size is equal or higher than 3.5 ϕ (Figure 9). This agrees with the idea that Mi flakes are preferentially concentrated in the coarser part of the sediment tail because of their lamellar shape, that is, they are hydraulic equivalent to finer grained sediments [10].

Figure 9. Correlation between Mi frequency and sediment mean grain size. The higher frequency of Mi (>40%) occurs only when the sediment is finer than 2.5 ϕ .

The 2nd component (that accounts for about 26% of variance – Table 5) highlights the Py loading which is a common mineral found in the samples collected from Porto and Aveiro areas (Figures 4B, 4C and 6). Since this mineral is absent on the northern Portuguese river sediments [33] we hypothesize that this mineral can be sourced from basic igneous rocks located elsewhere in this continental shelf/upper slope. The results of the electron microprobe analyses show that these Py grains correspond to diopside and augite (clinopyroxenes) and enstatite (orthopyroxene) which supports the existence of this basic igneous source (Table 3 and Figure 2). The presence of Ol (forsterite) although being a mineral with a very low frequency (mean frequency of 0.6 % - Table 2), also supports the existence of this source (Table 3 and Figure 3). Together, these minerals appear always with angular forms or, in less frequent cases, with distinctive euhedral shapes (Figure 10).

Figure 10. Visual aspect of typical angular and euhedral grains of pyroxene (Py_1 to Py_4 are clinopyroxenes; Py_5 to Py_8 are orthopyroxenes) and olivine (Ol_1 to Ol_4). Py_4, Ol_1 to Ol_3 are euhedral grains.

Under the 3rd component (that accounts for about 12% of variance) it is visible the isolated loading of Ap (Figure 6). This loading is confirmed by the Biplot 3D diagrams of Figure 5 where *clr*Ap vector is linked to the samples collected from Porto area (blue dots). The presence of Ap grains is well documented on the Douro estuary sediment infilling [34] as well as in Minho, Lima and Douro rivers sediments [33]. The presence of these mineral in river and in estuary sediments is interpreted as the result of the erosion of the granitic and gneissic outcrops very common in these river basins [34-33]. So, from these results and considering that Porto samples are the most proximal ones from Minho, Lima and Douro river mouths, we can deduce that these outcrops are the main primary source of Ap.

If we suppress the Mi frequency (data matrix of Table S3) to avoid the statistical effect of the affinity between this phyllosilicate with finer grained sediments [10], the PCA results projected on the Biplot diagram show the existence of three main *clr* vectors correspondent to the minerals Ap, Py and Am. This Biplot diagram according to the 1st, 2nd and 3rd principal components shows that the *clr*.Ap vector is associated to the Porto samples, the *clr*.Py vector to the Aveiro ones and the *clr*.Am vector to Nazaré samples (Figure 7). Something that stands out greatly from these results is that the third component (accounting for about 15% of variance - Table 6) shows the opposition of Am loading to the ones of Zi and Ga (Figure 8). Whereas the reasoning for the contrasting hydraulic behavior between the platy grains of Mi and the other heavy minerals is well known [9,10,29], the apparent contrasting hydraulic behavior between Am grains and other denser minerals (like Zi and Ga) it is less commonly reported in the literature [33,35]. To investigate this behavior a quantitative shape analysis of 30 random chosen Am particles was done, using 10 Am particles of each studied area. These particles were characterized using the ratio between the short length (S) and the intermediate length (I), that is S/I. The S length corresponds to the grain thickness which is assumed to be the axis perpendicular to the glass slide plan of the heavy mineral preparation. The intermediate length (I) corresponds to the grain width assumed to be parallel to the glass slide plan and perpendicular to the maximum length of the mineral grain (in the same plane). These lengths were measured under the petrographic microscope as follows: The S length was estimated using the interference color chart [36], and the I length was measured using the graphic scale of the microscope eyepiece. The results show that the S/I ratio is higher on Am particles belonging to Porto and Aveiro areas (with S/I between 0.156 and 0.500), and lower on Am particles collected from the Nazaré area (with S/I between 0.055 and 0.150) (Figure 11). This means that most of Am particles collected from the Nazaré area have predominantly flattened shapes that make a visible contrast with the more equant ones found in Porto and Aveiro areas (Figure 12).

Figure 11. Values of the S/I ratio measured for 10 sets of 3 Am grains (set 1 to 10). P – Porto, A – Aveiro and N – Nazaré.

Figure 12. Visual catalogue of the most typical Am grains particles found on each studied area. The Am sets corresponds to the Am reference in Figure 11 (bar equal to 0.5mm).

It is well known that the mechanical breakdown of sand particles during river transport do not alter significantly the relative proportions of the mineral species available in sources areas [37-39] even when this transport is very long and takes place in high energetic and arid environments [40-41]. However, some mineral species like amphiboles are particularly susceptible to mechanical breakdown because of their brittle character and good cleavage [37]. For this specific mineral group, the mechanical abrasion understood as a process which reduces the grain size, by a combination of fracturing and rounding can be very important [2]. Additionally, the relative lower hardness of Am minerals ($H = 5$ to 6) in a combination with the presence of two well-developed sets of cleavage planes can contribute to the generation of a significant number of flattened particles during long transport paths and longtime of residence in high energy sedimentary environments. Moreover, we must consider the effect of physical grain sorting that takes place during transport and depositional stages that will eventually influence the proportion of flattened versus more equant Am grains. So, it is possible to hypothesize that the long transport path of the Am grains from their most important primary source area (e.g. Douro river basin; [33] until the Nazaré canyon head area (a pathway with more than 150 km along the inner continental shelf), can contribute to induce an efficient grain sorting where the most flattened Am particles are more frequently entrained and transported (in suspension) comparatively to the more equant ones. These flattened grains have high mobility and when they are sourced to the inner continental shelf they are easily entrained and transported as suspended load. Consequently, these particles constitute a natural tracer of sand transport along the inner continental shelf from the northern Portuguese river mouths (e.g. Douro river) until the Nazaré canyon head area. This interpretation agrees with the knowledge of the Nazaré canyon head area sedimentary dynamics: this area is presently receiving sediment particles derived from rivers input and littoral drift [16,42]. Using the *clr* vectors of the Ap, Py and Am minerals as the main vertices of a principal component ternary plot (available at CodaPack software) and choosing the centered operation, the separation between the 3 sample sets it is clearly visible (Figure 13). In this figure the 1st principal

component (about 52% of explained data variance - Table 6) it is the axis according to which the separation between the Porto and Aveiro samples is defined. In its turn, the 2nd component (about 23% of explained data variance - Table 6) it is the axis according to which the separation between Nazaré and the Porto/ Aveiro samples is defined.

Figure 13. Ternary plot showing the distribution pattern of the samples collected from the 3 studied areas (blue dots: Porto; red dots: Aveiro; green dots: Nazaré). The green line represents the first principal component axis (PC1) which explains about 52% of variance. The yellow line represents the second principal component (PC2) which explains about 23% of variance. Each vertex of the triangle represents the main vectors which length on Figure 7 is proportional to the variance of the correspondent mineral in the data set composed by 8 minerals and 78 samples (data set of Table S3 with the suppression of Mi and recalculation to 100% - closure operation). The triangle vertexes c(clo_Ap), c(clo_Am) and c(clo_Py) represent the mineral *clr* vectors of Figure 7 transformed by the closure (clo) and centred (c) operations.

5.2. Conceptual model for heavy mineral data interpretation

Although geological sources control much of the mineral variability, there are other important processes that must be considered. Among these processes we must essentially consider the physical grain sorting and, in some specific situations, the mechanical grain abrasion. As such, our interpretation considers two fundamental primary sources: a regional one related with the igneous and metamorphic rocks outcropping in the northwestern Portuguese river basins, and a local one correspondent to basic igneous rocks located elsewhere on the vicinity of the Porto and Aveiro head canyons. The main heavy mineral assemblage (as it is characterized on Figure 4A) it is primarily derived from that regional source as reported in previous studies [33,34,43]. However, based on the concepts about the sediment source and dispersal reconstructions [44,45], we must consider that our minerals could also be sourced from reworked sedimentary shelf deposits, namely the coarser ones present on this continental margin at depths between 60 and 100 m and genetically linked to ancient littorals (essentially relict sediments) [46,47]. The influence of this sedimentary source could be distinguished on the heavy mineral suite by the presence of rounded grains in opposition to the more angular ones linked to the disaggregation of igneous and metamorphic rocks. So, this means that the regional source includes the presence of a “first-cycle” grain mineral suite that is easily identified by the angular to very angular grain shapes and a “multiple-cycle” grain mineral suite easily recognized by the rounded to very rounded grain shapes (Figure 14). In its turn the influence of local source is also easily recognized by the presence of Py and Ol grains with angular to euhedral shapes (Figure 10).

Figure 14. Typical visual aspects of the main heavy mineral transparent suite according to Figure 4A. Minerals catalogued with number 1 correspond to the “first-cycle” grains with angular to sub-angular shapes (euhedral forms in some specific cases). Minerals catalogued with number 2 represent the “multi-cycle” grains with rounded to sub-rounded shapes. For Py (pyroxene) the type 2 (“multi-cycle”) it is not identified (No_Id).

The manipulation of the quantitative data (relative mineral frequency) through the PCA analysis allowed the evaluation of the real importance of the source influence versus grain sorting. For the Porto area it is clear a strong influence of the primary regional source. The vector *clrAp* of the Biplot diagram characterizes this source and most of Porto samples are projected around this vector (Figures 5 and 7). The influence of the local source is very strong on Aveiro area. The vector *clrPy* of the Biplot diagram represents this source and most of Aveiro samples are projected around this vector (Figures 5 and 7). In its turn, the Nazaré heavy mineral assemblage denotes the dominant influence of the regional source overlapped by the strong influence of the grain sorting (sorting controlled by grain shape differences). The presence of Mi grains and their specific hydraulic behavior explains the particularity of the Nazaré heavy mineral assemblage (Figure 9). The vector *clrMi* of the Biplot diagram represents the regional source modified by the grain sorting and most of Nazaré samples are projected around this vector (Figure 5). When the Mi values are suppressed the Am values take the place of the Mi ones in terms of statistical relevance on the PCA analysis. The vector *clrAm* of the Biplot diagram represents the regional source and most of Nazaré samples are projected around this vector (Figure 7). These results are related with the specific morphological characteristics of Am grains. Most of these grains found on Nazaré have predominantly flattened shapes in a visible contrast with the more equant ones found on Porto and Aveiro areas (Figures 11 and 12). This is indicative of the existence of a grain sorting controlled by shape that deeply affects the mineral spectrum found at Nazaré area. Figure 15 summarizes the interpretation of the heavy mineral occurrence pattern in the studied areas according to the sediment supplier and distribution processes (sources and physical grain sorting).

Figure 15. Conceptual model for the interpretation of the heavy minerals found in the 78 collected samples from Porto, Aveiro and Nazaré areas. The regional sources are responsible for two different kinds of supplied minerals. The “first-cycle” ones with angular to sub-angular shapes very similar to the ones found in northern Portuguese river sediments [33,34]. The “multi-cycle” ones with rounded to sub-rounded shapes sourced from ancient littoral zones present in middle shelf, south of Cávado river mouth [33]. Local sources are responsible for the presence of angular minerals sourced from proximal igneous basic rocks. Minho, Lima, Cávado, Ave, Douro, Vouga and Mondego represent the main rivers that flow into the Atlantic Ocean. The *clr* prefix represent the main mineral vectors with the higher percentage of associated variance (Ap–apatite, Py–pyroxene, Mi–mica and Am–amphibole).

6. Conclusions

This study identifies the fundamental processes that control the non-random heavy mineral occurrence patterns recognized by the analysis of 78 samples collected from 3 distinct areas of the northern Portuguese continental margin: Porto, Aveiro and Nazaré canyon head areas. Globally, the main heavy mineral assemblage is composed by amphibole (hornblende), mica (biotite), andalusite, tourmaline, garnet, pyroxene (diopside, augite and enstatite), staurolite, zircon and apatite. However, each studied area has a specific mineral signature. In the Porto area the high frequency of andalusite and apatite marks the specificity of the mineralogical signature. In the Aveiro area the high frequency of amphiboles and pyroxenes stands out as a distinctive mineral characteristic. In the Nazaré area the extreme high frequency of mica (biotite) shows the peculiarity of the mineralogical signature. These results based on the mineral relative frequency point towards to the primary source as a fundamental process in controlling heavy mineral variability. In truly, the main primary source rocks (of igneous and metamorphic nature) seems to be the most influent process in controlling the presence of the referred heavy minerals on the referred areas. When we apply the principal component analysis (PCA) complemented with the centered log ratio transformation (*clr*), the processes controlling heavy mineral occurrence are clearly discriminated and individualized. The results of this analytical procedure projected in a Biplot diagram show that the collected samples are clearly separated

according to their geographical location. Also, with this procedure it was possible to define 3 main centered log ratio vectors on a tridimensional diagram that explain the essential of data variance. The first vector is linked to apatite frequency and it has associated the Porto samples. The second one is linked to pyroxene frequency and it has associated the Aveiro samples. Finally, the third vector is linked to Mi and Am frequencies and it has associated the Nazaré samples. Also, the results show that the primary source rocks (of igneous and metamorphic nature) and the grain sorting are the main processes that control the heavy mineral occurrence. In the Porto and Aveiro areas the influence of the sedimentary source is dominant being characterized by the presence of “first -cycle” mineral grains (with angular to sub-angular shapes) and of “multi-cycle” mineral grains (with rounded to sub-rounded shapes). By the contrary, in the Nazaré area the effect of the grain sorting dominates the mineral assemblage which is characterized by the high frequency of mica and amphibole flattened grains.

Supplementary Materials: Table S1: main heavy mineral suite, R_code S2: code in R for the zero transformation, Table S3: data matrix without zeros, Table S4: data matrix with clr transformation, Table S5: sediment texture, Table S6: transparent heavy mineral suite.

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