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Marble Quarry Waste Rock Piles and Evaluation of Their Reprocessing Potential for Lime and Cement Production (Marble Zone, Alentejo, Portugal)

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Abstract

Approximately 80% to 90% of the total stone extracted by the ornamental stone industry in the Marble Zone (Alentejo, South Portugal) is wasted and discarded in the numerous quarry waste rock piles across the region. This enormous amount of marble waste, representing millions of tons of stone each year, mostly with an expected high chemical composition of calcium carbonate, has significant potential with regards to reprocessing and use in other industries. This study evaluates the potential viability of these marble waste piles for reprocessing. The methodological approach, designed in two phases, firstly selected potential sites and, secondly implemented a sampling plan to allow for the laboratory analyses. In the first phase, a total of 178 waste rock piles were identified, corresponding to an estimated 70mn tons of marble raw material. In the second phase, 30 selected piles were sampled and chemically analysed by calcimetry, XRF and flame emission spectrometry. The results show that 7 of the sampled piles present $CaCO_3$ content above 97%, and another 14 show contents between 95% - 97%. This indicates that the waste could be reprocessed to produce lime and/or cement (clay materials are found in the surrounding schist outcrops in the region). There is also the potential for aggregate production for civil engineering works.

Keywords: marble, waste reprocessing, lime production

1. Introduction

It isestimated that approximately 80% to 90% of the total stone extracted by the ornamental stone industry in the Marble Zone (Alentejo, South Portugal) is wasted and accumulated in the numerous waste rock piles of the region (Figure 1). This enormous amount of waste, corresponding to millions of tons of stone each year, is due to the combination of a variety of factors, such as:

1. high fracturing degree of the rock masses, which prevents the production of blocks with adequate trade sizes;
2. aesthetic aspects (presence of weathering features from oxidations, smudges, veins, etc.), which devalue the stone, to the point of making it unsuitable for ornamental use.

From the mineralogical point of view, marble, just like sedimentary limestone, is almost exclusively composed of calcite, i.e., chemically it is CaCO3 (calcium carbonate). Calcium carbonate is a raw material for many industries, from the above-mentioned ornamental stone industry, to others, such as construction and building materials, like gravel and other aggregates, cement, lime, fillers for paints, animal feed, paper, water treatment, etc. Considering these several potential uses, this study aims to evaluate the feasibility of an industrial use of marble waste produced by the ornamental stone industry in Alentejo, namely for lime and cement production. To attain this purpose, the study presents the methodology followed for the characterization of these waste piles in order to assess the potential for the reprocessing of this raw material.

Figure 1: Waste rock piles produced by the ornamental stone industry in the Marble Zone (Alentejo, South Portugal).

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2. Methodology

The methodology followed in this study is basically structured in two phases:

1. Phase 1- site selection
2. Phase 2- preparation of a sampling plan in order to allow the necessary laboratory analysis to characterize the material.

Phase 1, by using *Google Earth* images and integration in a GIS database (ESRI ArcGIS 9.3), led to the identification of a total of 178 waste rock piles (Figure 2),corresponding to a calculated total volume of approximately 41.7 million m^3 and a total tonnage of marble raw material close to 70 million tons (Table 1) [1–3].

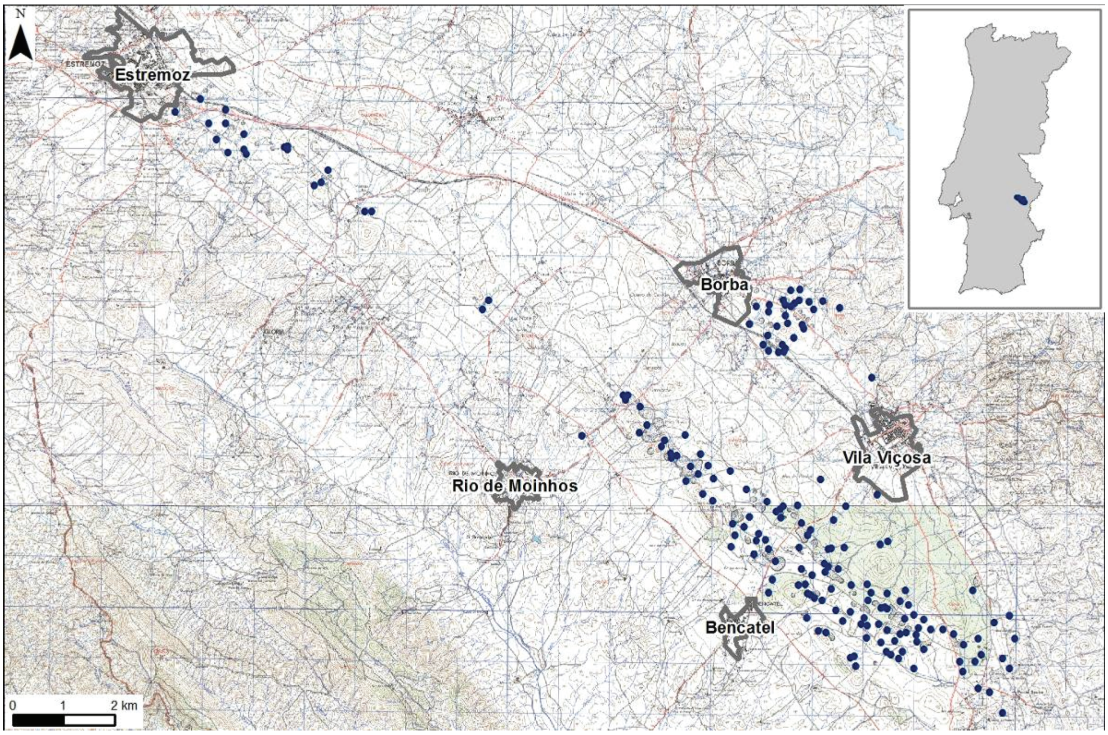

Figure 2: Location of the 178 waste rock piles (dots) identified using Google Earth images and integration in a GIS database.

Table 1: Total volume (V_T) and total solid volume (V_S), and respective total raw material volume (V_{RawM}) and tonnage (Q_{RawM}) calculated for the 178 waste rock piles.

Total Volume V_T	Solid Volume V_S	Raw Material	
		Volume V_{RawM}	Quantity Q_{RawM}
41.7*10^6 m^3	30.9*10^6 m^3	25.7*10^6 m^3	69.5*10^6 t

Based on the calculated quantity of raw material available to use, for example, in the production of lime, it is possible to estimate a total lifespan for each pile taking into consideration an average daily consumption of 300t, during 11 working months (335 days), as follows:

$$\text{Lifespan}_{years} = Q_{RawM} / 300*335 \quad (1)$$

Subsequently, Phase 2 delineated a sampling plan consisting of 30 selected waste piles geographically representative of the different marble types that are present in the Marble Zone region (Figure 3) and of the major extraction centres (Table 2): Cruz de Meninos (CM), Borba (B), Mouro-Vigária (MV), Lagoa (L) and Pardais (P). The total volume (V_T), solid volume (V_S), volume and quantity of raw material (V_{RawM} and Q_{RawM}) and the estimated lifespan (LS), according to equation (1), for each of the 30 selected piles are shown in Table 3.

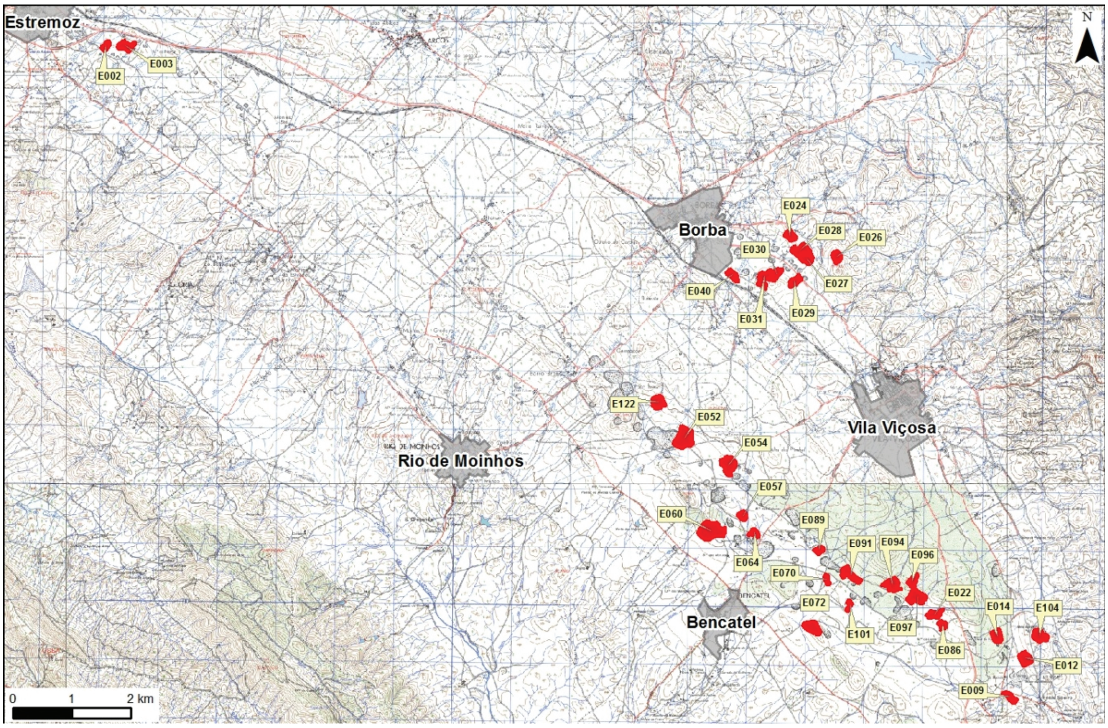

Figure 3: Location of the 30 selected waste rock piles (red shapes) representative of the different marble types that are present in the Marble Zone region.

Table 2: Number of waste rock piles selected from each sector and extraction centre in the Marble Zone.

Sector	Extraction Centre	N° of sampled Piles
Estremoz	Cruz de Meninos	2
Borba	Borba	8
Vila Viçosa	Mouro-Vigária	6
	Lagoa	10
	Pardais	4

Table 3: List of the 30 selected waste rock piles ranked from the largest to smallest in quantity of raw material (R – rank; EC – Extraction Centre; C – ID code)

R	EC	C	V_T (10^3 m^3)	V_S (10^3 m^3)	V_{RawM} (10^3 m^3)	Q_{RawM} (10^3 t)	LS (years)
1	MV	E060	4 066	3 012	2 510	6 776	67
2	MV	E052	3 066	2 271	1 892	5 109	51
3	MV	E054	1 646	1 219	1 016	2 743	27
4	L	E097	1 573	1 165	971	2 622	26
5	L	E072	1 121	830	692	1 868	19
6	B	E028	1 079	799	666	1 798	18
7	L	E094	1 009	747	623	1 682	17
8	P	E104	962	713	594	1 604	16
9	MV	E122	962	713	594	1 604	16
10	L	E091	914	677	564	1 523	15
11	B	E031	900	667	556	1 501	15
12	P	E012	823	610	508	1 372	14
13	CM	E003	752	557	464	1 253	12
14	P	E014	693	513	428	1 155	11
15	B	E027	642	476	396	1 070	10,6
16	L	E022	524	388	323	873	8,7
17	L	E096	485	359	299	809	8,0
18	B	E024	467	346	288	779	7,7
19	B	E029	467	346	288	779	7,7
20	B	E026	461	341	285	768	7,6
21	B	E030	453	336	280	755	7,5
22	B	E040	436	323	269	727	7,2
23	P	E009	351	260	216	585	5,8
24	MV	E057	303	224	187	504	5,0
25	MV	E064	271	201	167	452	4,5
26	CM	E002	257	190	158	428	4,3
27	L	E086	230	170	142	383	3,8
28	L	E089	205	151	126	341	3,4
29	L	E070	138	102	85	230	2,3
30	L	E101	129	95	79	214	2,1

3. Results

All samples collected from the 30 waste rock piles were chemically analysed by calcimetry, XRF (X-ray fluorescence) and FAES (flame atomic emission spectrometry) in order to determine contents in major and minor elements: Ca, Mg, Si, Al, Fe, Na, K, Ti, Mn; and also in TOC (total organic carbon), IR (insoluble residue) and LOI (loss on ignition).

A broad comparison of the obtained results in this study with those known of the 36 ornamental stone varieties produced in the region [4], does not show significant differences, thus corroborating that the major reason for all the stone waste being deposited in the waste piles is either due to insufficient block size in quarry extraction or to unsuitable aesthetic quality of the marble, rather than to poor raw material chemical characteristics, e.g. due to the presence of “contaminant” materials (such as schistous layers, basic igneous dykes, etc. that occur interspersed with the marble).

Table 4 shows the obtained results by grouping all samples from each extraction centre giving the respective mean and standard deviation (SD) values. Global mean, maximum, minimum and SD results for all 30 samples are also shown. According to literature review [e.g. [5, 6]], the major restraint for lime production, in terms of purity criteria of the raw material, mostly depends on the use that is intended. This means that, although there are reference values, essentially for carbonate content, they cannot be considered as mandatory limit values for the exclusion of a given sample. Still, generally, three ranges of carbonate content corresponding to the purity of the raw material can be considered: less than 95%; between 95% and 97%; and greater than 97%.

The chemical analyses of the sampled materials show that 7 piles present $CaCO_3$ content above 97% and another 14 show contents between 95% - 97%. This indicates a potential reprocessing use to produce lime and/or cement (clay materials are easily found in the surrounding schist outcrops in the region which, by the way, are source of local ceramic industry). The following figures (Figures 4 to 8 adapted from [3]) show the calcium carbonate content values obtained for the piles from each of the extraction centres analysed.

4. Final Considerations

This study has assessed the potential for reprocessing of the huge amount of waste produced by the ornamental stone industry in the Marble Zone of Alentejo, indicating as a feasible option the production of lime and/or cement although other uses may

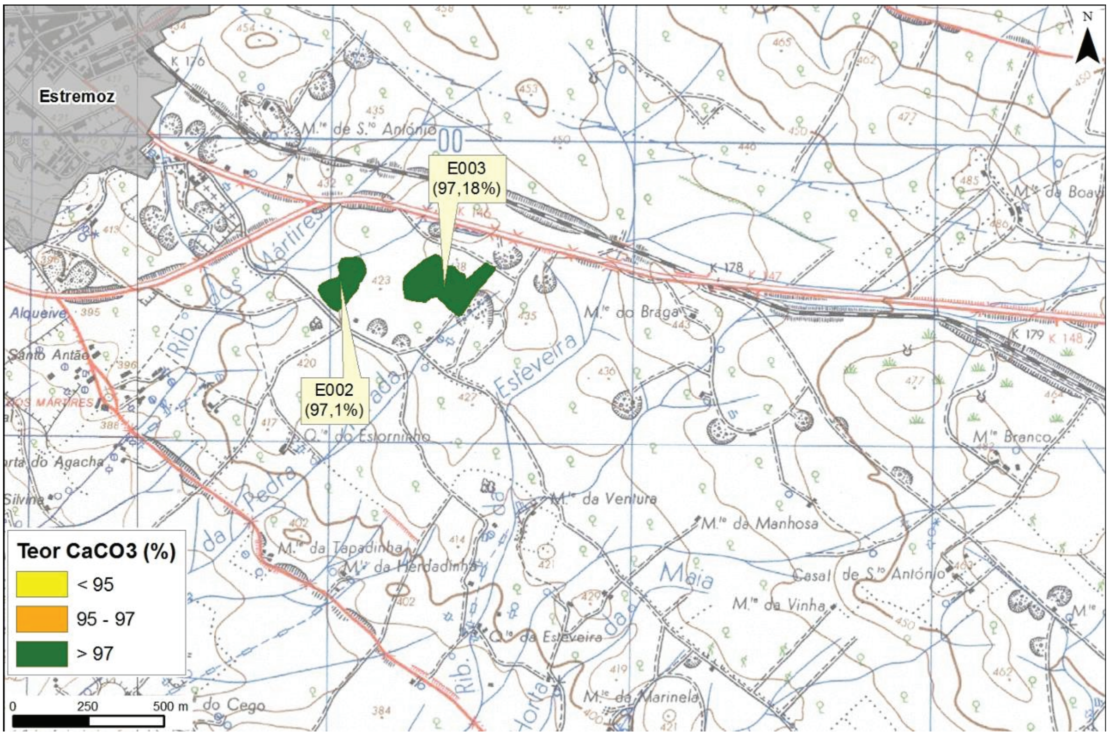

Figure 4: Calcium carbonate content for the waste rock piles from the Cruz de Meninos extraction centre.

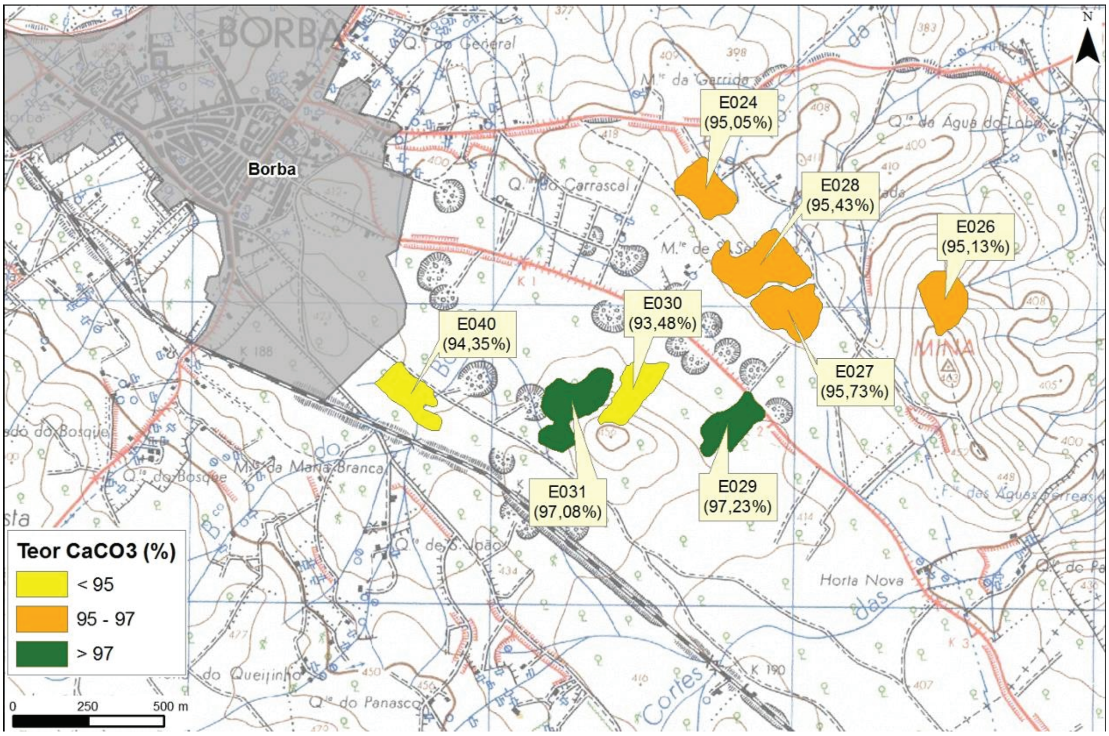

Figure 5: Calcium carbonate content for the waste rock piles from the Borba extraction centre.

point to aggregate production for civil engineering works, looking beyond the current «take and make waste» extractive model, towards a circular economy that focuses on positive society-wide benefits [7]. From the economic point of view, the main advantage

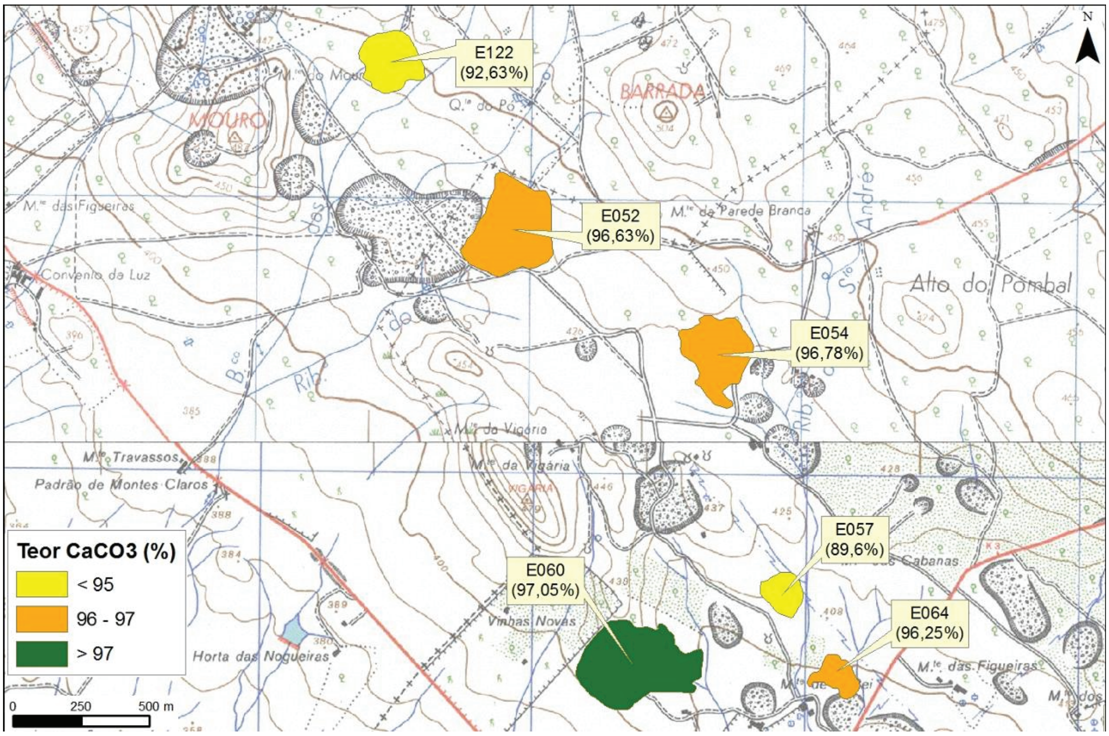

Figure 6: Calcium carbonate content for the waste rock piles from the Mouro-Vigária extraction centre.

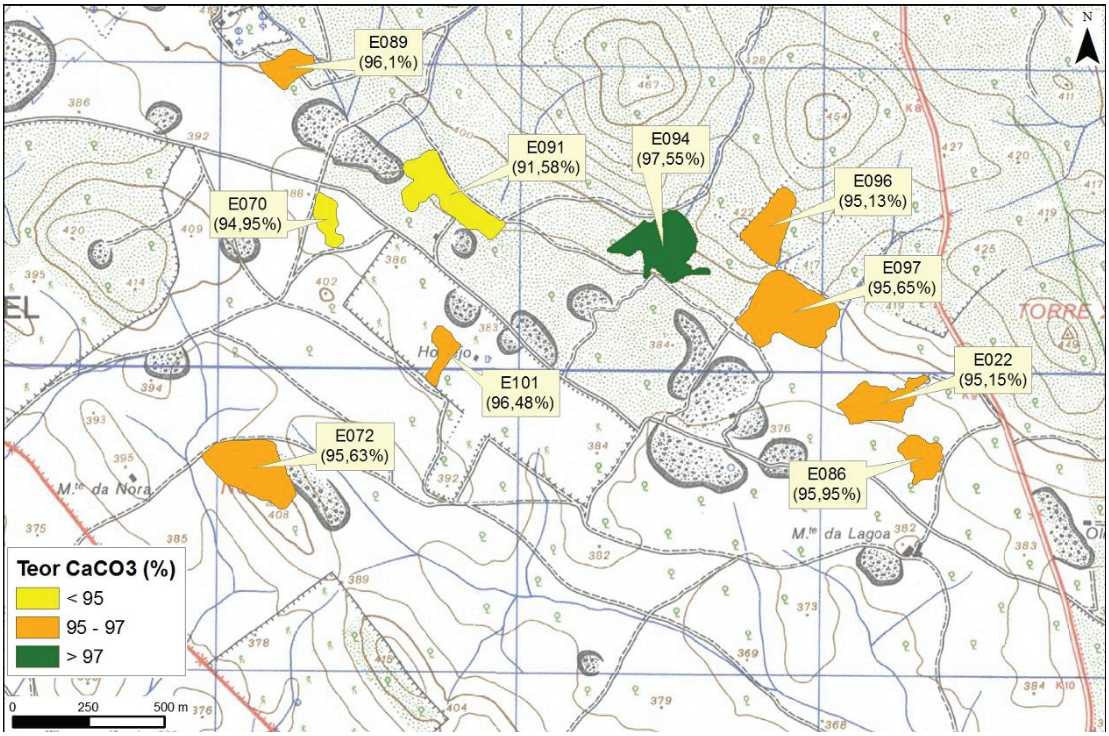

Figure 7: Calcium carbonate content for the waste rock piles from the Lagoa extraction centre.

associated to these reprocessing uses relates to the significant reduction of extraction costs.

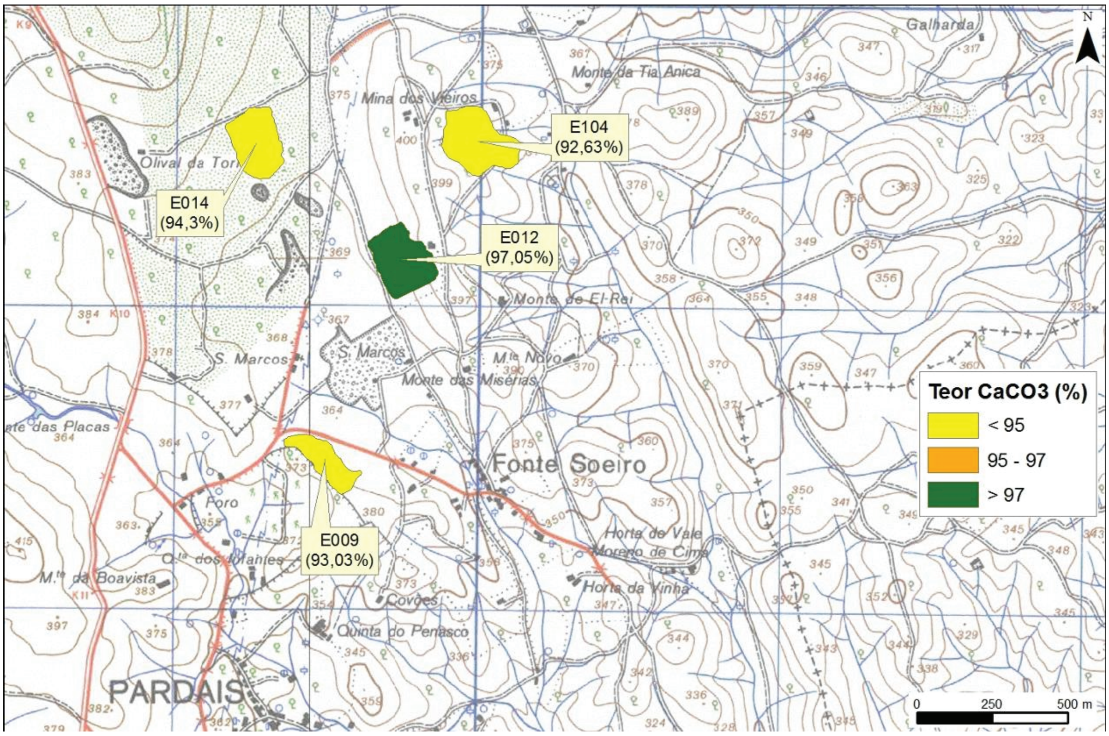

Figure 8: Calcium carbonate content for the waste rock piles from the Pardais extraction centre.

The potential use of marble waste originated from waste rock piles can also present relevant advantages to the environment, in a way that it can promote land reclamation by releasing land occupied by the piles and may help to avoid, or at least reduce, the perpetuation of land degradation by mining activities that often occur in environmentally sensitive designated areas (such as the National Parks of *Aire* and *Candeeiros* and of *Arrábida*).

Moreover, in relation to land planning and management plans in the exploitation areas, a few of the studied sites show conflicts with designated areas such as the REN and RAN (National Nature and Agriculture Reserves) which should be addressed in the future.

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Table 4: Obtained Global results (for the 30 samples) and grouped for each extraction centre (Cruz de Meninos, Borba, Mouro-Vigária, Lagoa and Pardais).

		Na_2O	MgO	Al_2O_3	SiO_2	K_2O	CaO	TiO_2	MnO	Fe_2O_3	IR	LOI	TOC	$CaCO_3$
		(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)	(%)
Global results	**Mean**	0.03	0.82	0.41	1.42	0.12	55.73	0.02	0.01	0.25	2.27	43.57	0.08	95.26
	Max.	0.08	1.97	0.82	2.42	0.23	58.20	0.11	0.06	0.40	4.16	52.82	0.22	97.55
	Min.	0.00	0.46	0.26	0.87	0.00	43.65	0.00	0.00	0.11	1.14	39.75	0.03	89.60
	SD	0.02	0.41	0.16	0.39	0.05	2.45	0.02	0.01	0.08	0.71	3.04	0.05	1.89
Cruz de Meninos	**Mean**	0.03	0.58	0.34	1.40	0.09	56.95	0.02	0.01	0.24	1.47	43.39	0.07	97.14
	SD	0.01	0.04	0.04	0.26	0.00	0.69	0.03	0.01	0.12	0.23	1.56	0.03	0.06
Borba	**Mean**	0.03	0.92	0.42	1.36	0.13	56.06	0.03	0.01	0.32	2.18	42.74	0.10	95.44
	SD	0.03	0.45	0.14	0.38	0.08	0.59	0.02	0.02	0.05	0.66	1.89	0.05	1.27
Mouro-Vigária	**Mean**	0.02	0.65	0.39	1.44	0.11	54.49	0.02	0.01	0.22	2.60	44.81	0.09	94.82
	SD	0.03	0.16	0.16	0.42	0.03	5.39	0.01	0.01	0.06	1.03	4.08	0.07	3.04
Lagoa	**Mean**	0.03	0.98	0.43	1.44	0.13	55.75	0.02	0.00	0.22	2.32	44.38	0.08	95.42
	SD	0.01	0.53	0.16	0.45	0.05	1.03	0.02	0.01	0.07	0.61	3.46	0.03	1.55
Pardais	**Mean**	0.02	0.62	0.43	1.51	0.12	56.24	0.03	0.01	0.21	2.23	41.41	0.07	94.25
	SD	0.00	0.12	0.26	0.44	0.06	0.91	0.06	0.02	0.11	0.46	1.80	0.03	2.00

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