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Managing Marble Quarry Waste: Opportunities and Challenges for Circular Economy Implementation

Antonio Tazzini [1], Francesca Gambino [1,*], Marco Casale [2] and Giovanna Antonella Dino [1]

[1] Earth Sciences Department, University of Torino, 10124 Turin, Italy; antonio.tazzini@unito.it (A.T.); giovanna.dino@unito.it (G.A.D.)
[2] Department of Management, University of Torino, 10124 Turin, Italy; ma.casale@unito.it
* Correspondence: francesca.gambino@unito.it

Abstract: Marble has been a prominent natural stone exploited since ancient times, commonly employed as a building material and ornamental stone. However, the disposal of waste generated from marble extraction, particularly fine sludge, poses significant environmental challenges for the dimensional stone industry. The difficulty in managing and recovering these materials, exacerbated by local regulations and the absence of suitable recovery protocols, often leads to landfilling. This issue is exemplified by the Carrara Marble Basin in the Apuan Alps (Tuscany, Italy), where more than half of the extracted marble remains as quarry waste or debris. Modern cutting technologies have intensified the production of finer materials and sawing residue, known as "*marmettola*", ranging in size from fine sand to silt. The disposal of these materials, commonly through landfilling or abandonment, has profound environmental repercussions. This research primarily aimed to carry out a preliminary physical, mineralogical, and morphological characterization of the fine waste and sludges generated from Carrara Marble exploitation. The findings reveal the high granulometric uniformity of the materials, and a nearly pure carbonate composition, suggesting potential for reuse in various industrial sectors such as paper, plastics, and pharmaceuticals production.

Keywords: ornamental stones; circular economy; marble sludge; waste recycling

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1. Introduction

The dimension stone industry has significant economic potential that has been steadily increasing over the years, in particular due to rapid expansion in the construction sector. As a natural material, stone has unique physical and mechanical properties, and therefore it is widely used as a raw material in construction, for sculptural masterpieces and architectural elements, representing the world's cultural heritage [1,2]. The extensive historical usage of dimension stones as construction materials underscores their enduring significance throughout mankind's existence. From the monumental pyramids of Egypt to the inspiring sculptures meticulously crafted by Michelangelo, diverse types of natural stones have been harnessed by human ingenuity for architectural endeavors [3]. The great heritage and historical values of ornamental and dimension stones ("Heritage Stones") in European cities, together with their high aesthetic value, make this market potentially very valuable.

According to statistics published in 2020 by countries with an active dimensional stone industry, including China, India, Turkey (which, together, make up around 60% of the world's quarry production), as well as Brazil, Iran (5.3%), Italy (3.8%), Spain, Egypt, Portugal and the USA (2–3%), the total amount of material extracted annually from dimensional stone quarries is about 316 Mt (gross quarrying production) [4–6], including marble, gneiss and granites, and other stones. Despite being seriously affected by the economic crisis, the dimension stone sector has continued to grow steadily on a global scale for 60 years. The growth in the use of dimension stone products has accelerated in recent years, with an annualized growth rate of 7.5% since 1986. This has been attributed to more frequent discoveries of new stone deposits, a growing awareness of stone culture (particularly in countries without a long tradition of stone use), and technical advances in extraction and beneficiation techniques. In terms of value, the dimension stone industry is significant, with the total value chain estimated at over USD 60 billion worldwide [7].

However, the production of dimension stones involves a great amount of waste (extractive waste), which poses several serious environmental problems, such as degradation and reduction in land quality, damage to biodiversity and destruction of habitats, water and air pollution (which in turn causes impacts on human health). Extractive waste (EW) is a material that must be removed either during extractive activity (such as topsoil, overburden, waste rock) or during the processing stages of industrial minerals, ores and rocks, and/or energetic materials (operating residues and tailings). EW production represents the second largest sector responsible for waste production in the EU, characterized by an annual production of about 622 million tons (26.6% of the total amount of waste production) [8]. The European Directive 2006/21/EC of 15 March 2006 [9] on EW management oversees the permitted conditions, storage, monitoring and control of the produced waste to ensure the protection of human health and the environment. Furthermore, EU policies aim to reduce the amount of waste disposed of in waste facilities and landfills by promoting waste recovery and recycling, also in the extractive industry (in line with circular economy principals).

In general, dimension stone production comprises three phases: exploration, quarrying and processing. Following the exploration phase, which includes volume estimation, stone characterization and environmental and economic studies, the subsequent quarrying stage involves the preparation of the excavation site, cutting and extracting large blocks of stone from the quarry, and then turning these blocks into smaller blocks for easy transportation to the processing plant [5]. During this phase, about 51% (161.5 Mt) of the total extracted material is regarded as solid waste from extraction operations, in the form of the following:

- Defective blocks which do not meet the quality standards, resulting from the presence of joints and cracks and poor choice of cutting geometry;
- Shapeless blocks, excessively irregular or characterized by inadequate volume for industrial-type processing;
- Offcuts, which occur when stone blocks with uneven surfaces are cut;
- Fine waste resulting from the mixing of water and dust generated during the cutting phase (from diamond wire cutting machine, toothed chainsaw, etc.).

At working plants, depending on the expected use, the incoming blocks may be cut into large, thick, flat slabs, typically squared, or into smaller tiles that will be sold as the final product. This process involves re-cutting the blocks that arrive from the quarry into regular blocks and then placing them in a cutting machine (such as a diamond gang saw machine). After the sawing process, the produced slabs are polished and smoothed, and the final cutting is performed. About 41% (154.5 Mt) of the regular blocks transported to dimension stone processing plants turns into waste during this phase, producing mainly fine materials (sawdust and residual sludge) that result from mixing water and dust produced during sawing, polishing and cutting. The total waste makes up about 71% of all the extracted material, with only 29% of that transformed into the final product [4,5].

The necessity to reduce the use of non-renewable natural resources and, at the same time, to minimize the negative impacts the on environment, has led to an increasingly high interest in recovering and recycling. EW from dimension stone exploitation (Figure 1) can potentially be recovered for different applications, such as the following:

- Aggregates, which are very important for the European industry because of their wide application fields such as aggregates for concrete used in the construction, building and infrastructure sector [10–13], railway ballast used in railway construction, bituminous aggregate for road construction, and aggregate for subfloor layers used in railway and road construction;
- Industrial minerals [14,15], crucial for a country's development across various sectors. They find applications in construction (clay for bricks, calcium carbonate for mortars

and gypsum for plasterboard), industries (feldspar, kaolin and quartz for ceramics and glass), and environmental use (such as clay/bentonite as a waterproof material for landfills);

- Artificial substrates (namely technosols) for environmental and quarry rehabilitation, employing quarry sludge (especially silicate sludge) with soil and/or organic waste materials such as composted municipal waste and green manure [16–18]. Other studies have investigated their employment as soil fertilizer, as a waterproof material for landfills, etc. [19];
- Geopolymers [20,21];
- Filler for industrial purposes, such as cement, mortar and concrete production [22–24];
- Raw materials and critical raw materials (RM/CRMs), crucial for the EU economy and industrial development. Ni, for example, is used for Ni-Cd batteries and in superalloys for electric power stations or for airplane turbines; Fe, employed in steel production, in the paint industry and for magnet production; Al in car and airplane production; Cu for electric devices and line production; together with some CRMs which can be associated with them (e.g., PGEs associated with Ni; Cd, Ge, Ga, In associated with Zn-Pb ore deposits), as well as REE, the importance of which has increased over the last few decades [25].

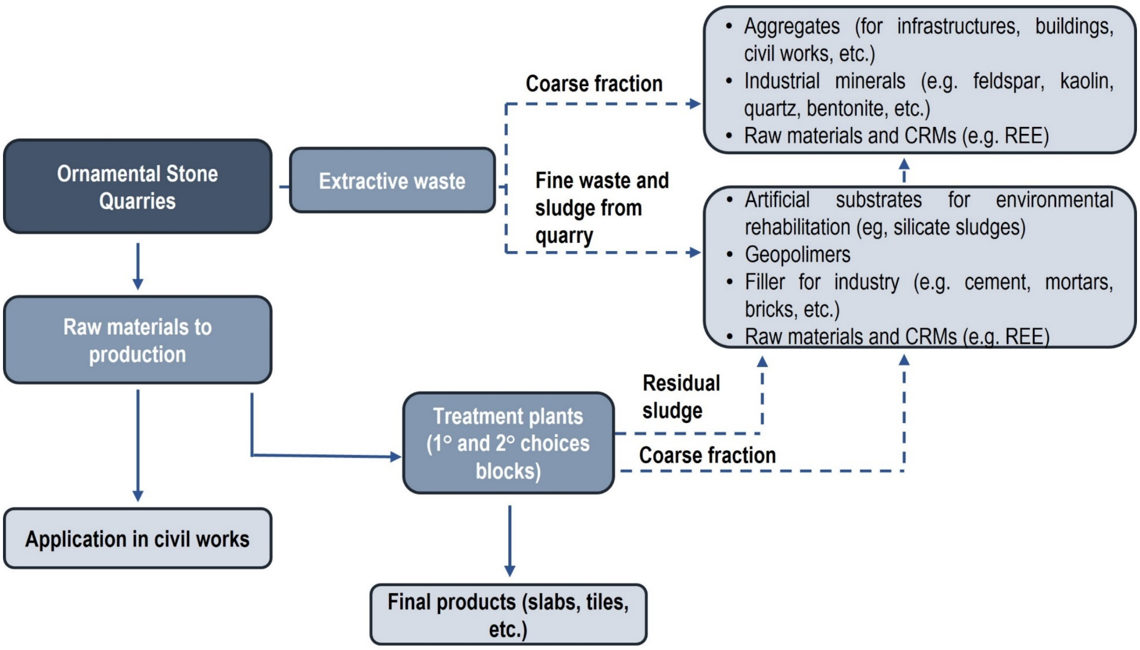


Figure 1. Simplified flow diagram about potential recovery of EW from dimension stones (modified from [26]).

Unlike any other extractive industry, marble exploitation produces large amounts of scraps/waste. The study from Mehta et al., 2020 [27] indicates that the level of waste generation is very high in marble industries, as, in the majority of them, less than or around 30–40% of the output is high-value products (in terms of fresh and high-value blocks) and more than 70% is waste (defective blocks and tiles, broken tiles and flakes, etc.), particularly in quarries using conventional techniques of blasting—no longer used in Italy—where the rocks are fractured. The fine waste produced during stone cutting activities and processing (including both the microfine dust/quarry sludge and the residual sludge from working operations) is the most difficult fraction to recover, due to its high management costs and lack of proper protocols to recover it, other than landfill [28]. The marble industry can produce about 30% of slurry waste (sludge) from quarry production, in relation to the volume of the excavated marble, 15–20% from processing activities, and about 5 to 7% resulting from polishing waste [27,29].

Fine fraction (sludge) from the dimension stone industry has, over time, been considered as real waste and therefore landfilled. This solution is no longer compatible with the increasing and pressing needs for environmental care and eco-sustainability.

The literature reviews concerning the recovery and use of marble sludge as secondary raw materials (SRMs) have mainly focused on applications in concrete [30–33], ceramics [34–36], mixed with lime and/or cement [37], other construction products [38,39] and as a reagent to reduce acid mine drainage phenomena [40]. Calcium carbonate ranks right at the top within these industries and in other areas because of its high-quality performance in the productive process and for the brightness it confers to the final products. Its usage in these sectors as mineral filler is dependent on its characteristics and market requirements (Tables 1–3) [41].

Table 1. Values for $CaCO_3$ filler requirements in paper industry [41]. GCC: ground calcium carbonate.

Parameter	Value
$CaCO_3$ [%]	>95
Brightness ISO [%]	80–96
Average particle size [µm]	<3
Particle size < 2 µm [%]	2–20
GCC solids content [%]	75–78

Table 2. Values for $CaCO_3$ filler requirements in rubber industry [41].

Parameter	Value
d_{50} [µm]	0.7–5.0
$CaCO_3$ [%]	98.5
SiO_2 [%]	0.1–0.4
Fe_2O_3 [%]	<0.08
Specific gravity [g/cm^3]	2.7
Particle size < 10.5 µm [%]	82–90

Table 3. Values for $CaCO_3$ filler requirements in tire industry [41].

Parameter	Value
Appearance	White to light grey powder
Specific gravity [g/cm^3]	2.7
Fineness—150 µm [%]	100
Fineness—45 µm [%]	>95
Fineness: Residue at 40 microns [%]	<0.5
Loss in mass at 105 °C [%]	<0.5
Loss on ignition [%]	43.50 ± 1.5
$CaCO_3$ [%]	>92
Cu [%]	<0.005
Mn [%]	<0.05
Particle size < 10.5 µm [%]	82–90

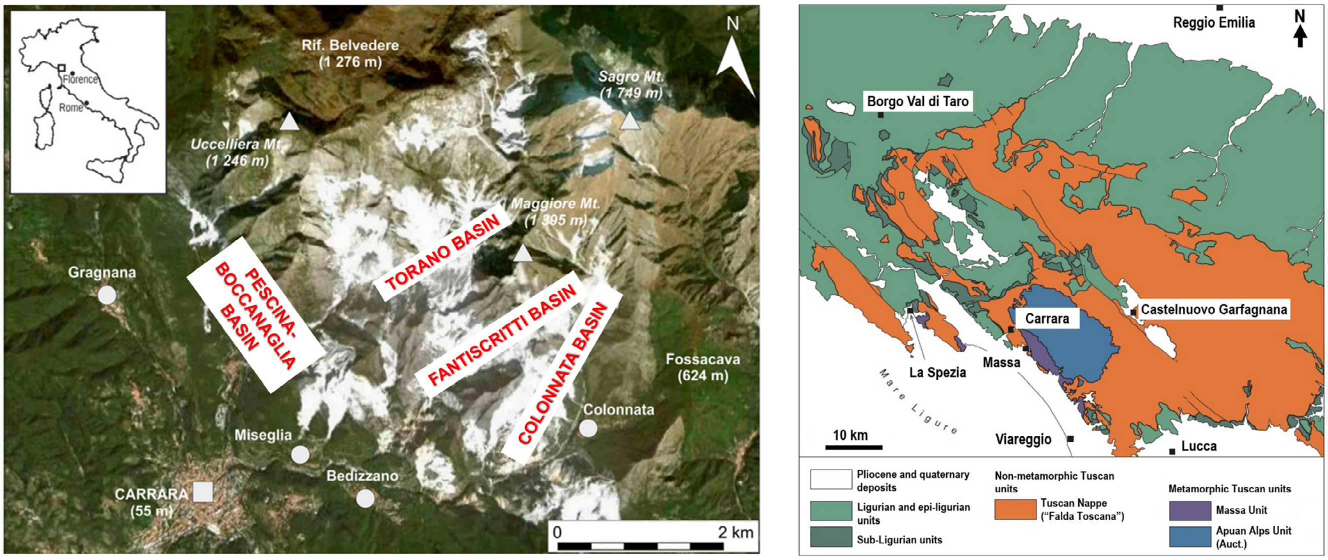
The effective management and recovery of dimension stone waste is crucial to improve environmental sustainability and resource efficiency in the extractive sector, in line with the sustainable mining approach. Indeed, by addressing this issue, it is possible to minimize the environmental impact, preserve natural resources for future generations and promote sustainable development.

The present paper focuses on the Carrara Marble Basin in the Apuan Alps, Italy. The first step was to evaluate some insights into the Carrara marble production context and quarry waste production, especially finer fractions. In phase two, the characteristics of marble sludge from primary and secondary operations in quarries were investigated, as well as sludge produced in working plants. Sludge materials were sampled accurately and in the most representative conditions to ensure a representative analysis (physical analysis and geotechnics, mineralogy and geochemistry).

2. The Carrara Marble Basin

2.1. Geological and Historical Setting

Situated in the northernmost reaches of Tuscany, Italy, the Carrara Marble Basin (CMB) is nestled within the picturesque Apuan Alps region and stands as one of the preeminent marble quarrying districts globally. Spanning approximately 375 square kilometers, this basin is intricately divided into four sub-basins, extending from the northwest to the south-east: "Pescina-Boccanaglia", "Torano", "Fantiscritti" and "Colonnata" (see Figure 2) [42]. Carrara marble, a widely revered dimension stone and proudly designated as a "Global Heritage Stone Resource" [43], constitutes a vital component of the Apuan Alps Unit (Paleozoic–Upper Oligocene). This geological formation emerged trough the metamorphism of Liassic platform deposits amidst the tumultuous Alpine orogeny, experiencing distinct temperature peaks and pressures ranging from 350 to 450 °C at 0.4–0.8 GPa [44–46].



(a) **(b)**

Figure 2. (**a**) Carrara Marble Basin: white areas evidence quarries and extractive waste (modified from [42]); (**b**) tectonic scheme of the northern Apennines (modified after [47]).

The extraction history of marble from the Carrara region extends beyond 2300 years, possibly making it the world's longest-standing extraction of a single mineral resource [48]. Using Etruscan evidence from certain areas (Versilia, Pisa and Volterra), along with specific analyses of funerary marbles along the Versilia River valley, it is possible to date the first use of Apuan marble to the pre-Roman period [49]. The material was transported downstream to Luni city and then along the river Tiber to the distribution quays in Rome. It is estimated that several thousand tons were exploited annually [50]. Marble exploitation stopped after the fall of the Roman Empire, but then started again during the Renaissance [51], especially due to Michelangelo's preference for Carrara white marble in his masterpieces. Marble exploitation saw a steady increase until the late 20th century, driven by technological advancements and growing international demand [52]. The use of explosive charges in shot holes, known as "Varata", from the 1800s often led to significant waste generation. The transportation of marble to and from the quarries was revolutionized by the "Marmifera" railway network in 1876, replaced by road vehicles in 1964 [49]. Another significant advancement was the introduction of helicoidal wire technology in Carrara quarries in the late 19th century, which improved cutting efficiency. The wire was pressed against the marble and made to slide by a pulley, quartz sand was introduced into the cut and conveyed by the wire to cut the marble. Water was used to remove sand and marble powder and to cool the tool [51]. From the 1990s, a surge in marble demand from foreign countries (such as China, Russia and the Emirates, above all) led to rapid quarrying growth. To meet these needs, diamond wire cutting machines and toothed chainsaws were introduced in Carrara quarries, enhancing production efficiency and product quality [46]. These innovations, along with the use of large front loaders and excavators, increased quarry production up to 1.5 Mt at the beginning of 2000; nowadays, this has decreased to about 1 Mt (referring to ornamental block production) [52,53].

Currently, Carrara still plays a key role in marble quarrying, hosting the largest number of quarries both at the provincial level (about 80 active quarries compared to those in the neighboring municipality of Massa, which are about half), and at the level of the entire Apuan territory (Garfagnana and Versilia basins). According to PABE ("Piani Attuativi dei Bacini Estrattivi delle Alpi Apuane" [54]), the total area currently occupied by the CMB accounts for about 16% of the municipality's total area, with a mix of small family-run quarries, which employ a limited number of workers (<5), and larger companies, such as cooperatives, employing from 10 up to 50 workers. However, the stone sector is predominantly composed of micro-enterprises (<10 employees), with only 9% classified as small businesses (from 10 to 50 workers) [54].

On the western side of the CMB, the Pescina-Boccanaglia basin, often grouped with the Torano basin due to its small size, hosts a limited number of quarries (<10). These two basins are renowned for producing marbles of the best qualities such as "Statuario", "Calacatta" (from Calacatta locality, north of Torano city), "Arabescato" and "Bianco Carrara" white marble. On the eastern side of the CMB, the Colonnata basin (where some of the marble sludge discussed in the following sections were sampled) serves as the primary production source of marble for the region, yielding prevalent white marble varieties such as "Ordinario" (extracted from over 50% of Carrara quarries and contributing to more than 50% of the total exploitation in the Apuan Alps region), "Venato" (the second most quarried marble variety, accounting for approximately 24.8%) and Bianco "Brouillé" (sub-variety of "Arabescato" marble). Dark-colored marble varieties like "Bardiglio" and "Nuvolato" are also extracted in Carrara quarries and make up about 3% of the total production. Marble varieties often take on specific local names based on their extraction locations [55].

Figure 3 illustrates the main varieties of marble extracted in the Apuan Alps region and within the CMB (from a macroscopic appearance), while principal data about Carrara marble characteristics (on average) are reported in Table 4.

Table 4. Carrara marble physical and mechanical characteristics (on average) [46].

Physical Characteristics	Value
Bulk density	2688 kg/m^3
Simple compression strength	1209 kg/cm^3
Compression strength after freezing	1181 kg/cm^3
Indirect tensile strength (Brazilian test)	174 kg/cm^3
Impact strength test	73.8 cm
Moisture absorption	0.16%

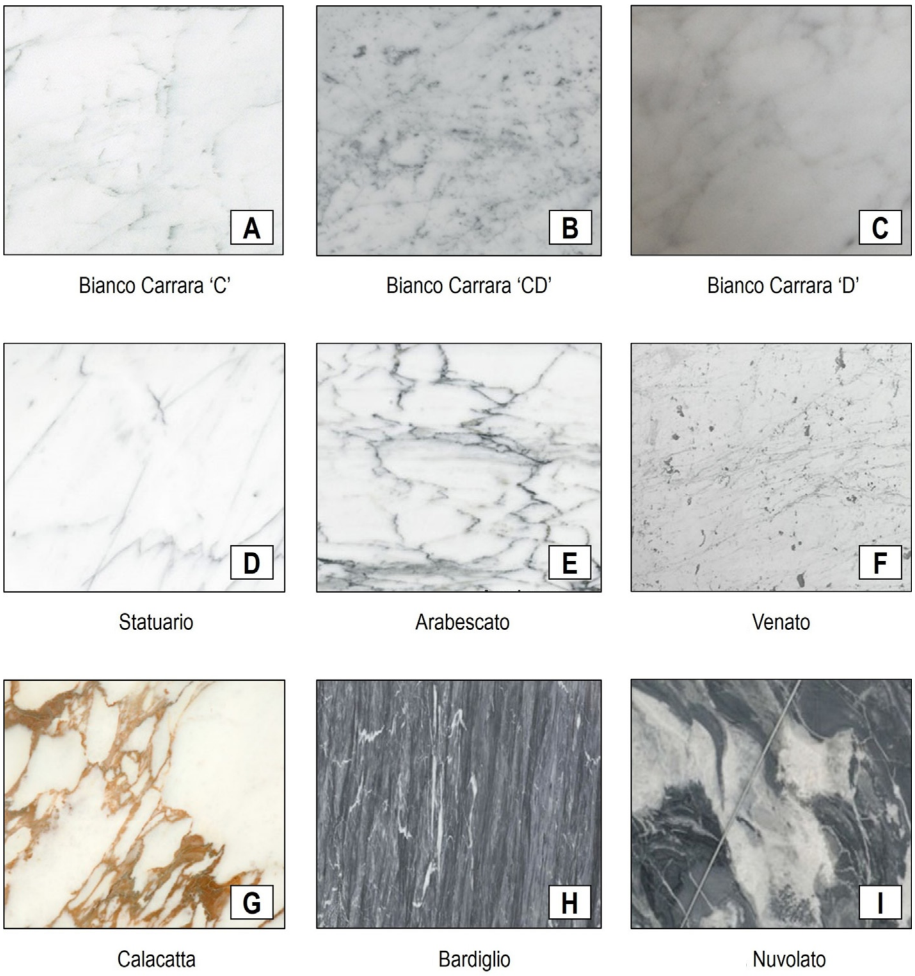


Figure 3. Macroscopic appearances of the main commercial varieties of marble from the Apuan Alps region. Bianco Carrara sub-varieties are distinguished according to the chromatic tone of the groundmass: 'C', 'CD' and 'D' (modified from Primavori, P. Available online: https://www.litosonline.com/en/article/different-varieties-marble-carrara, accessed on 2 April 2024).

2.2. Recent Developments and Technological Innovations in the CMB

During cutting and processing, natural stone like marble is subjected to various mechanical stresses in the production chain, such as sawing and cutting, calibrating, polishing and transport. Flexural loads, impacts, and vibrations are involved in affecting the strength of the weakness planes, which exist in a variety of forms, from large fractures, which affect the entire block, to smaller stylolites. In the past, the most common solution to the problem of fractures was to replace the damaged stone with a new one. This method could negatively affect the beauty or the strength of a structure, especially for historic buildings or sculptures [56].

As cited by Zanzi et al., 2023 [57], in recent years, the quarrying industry has experienced increasing demands also for non-destructive techniques to optimize stone production, such as sensing technologies for the mapping and characterization of rock fractures. For example, the GPR method (ground-penetrating radar) is a totally non-destructive sensing technique capable of mapping the fractures at different stages, such as during exploration and extraction in quarries, to optimize the production of building stones, reduce the production costs and waste, and for post-production demands to repair and restore the stones. New strategies try to repair the damaged stones in a fast and high-quality way in order to overcome the disadvantages of the old solutions. These methods apply different techniques, such as cement filling, the injection of epoxy and polyester resin, ultra-violet resin, and mastics [56,58]. The epoxy resin is composed of an epoxide component and a hardener or curing agent. Epoxy resins are thermosetting adhesive materials that are known for their strength and creep resistance and are widely used in the conservation and consolidation of a number of famous stone monuments in the world and in stones like granites and marbles [59]. Additionally, their resistance to mechanical stress and relatively high viscosity are two of the most significant characteristics of such resins. Furthermore, they can efficiently penetrate into porous and fractured zones, consolidating the weak parts of stones/blocks. All the aforementioned benefits explain why epoxy resin is an increasingly preferred choice to repair the fractures of stones and masonry structures [57].

Resining in the quarry of dimensional stone is often performed with epoxy resins that are injected between the stone and a layer of air-impermeable material previously attached to the bench; this layer of air-impermeable material has the function, through the aid of specific portable pumps, of placing the bench itself in a "vacuum". To apply the vacuum method, the block must first be washed, manually cleaned of any obstructions that might slow or prevent the resin from penetrating into it, and finally dried. With this procedure, the resin penetrates the various fractures and pores within the inner portion of the marble block, "welding" them together. In the CMB region, stone reinforcement technology began approximately 12 years ago, but even in the late 1990s, some companies in the Apuan Alps region conducted initial experiments with vacuum resin application [60]. In the CMB, where resin methods are used (Figure 4), the vacuum method is not often applied directly in the quarry, due to operating and environmental conditions that are not always favorable (it is mostly used downstream in working plants), so other methods of reinforcing blocks are used. Marble blocks can be reinforced in the four faces with wooden planks and epoxy resin so as to prevent their breakage during subsequent transport and sawing in the working plant, or they can be reinforced with a fiberglass mesh and epoxy resin on the external side (resin coating).

(a) **(b)**

Figure 4. Resining marble blocks inside the CMB; (**a**) resining a block directly in the quarry by external consolidation and "stitching" the fractures; (**b**) resining a block in working plants by the vacuum method (resin is also injected inside the block), applied before cutting into slabs, to obtain as many slabs as possible at the final stage.

Epoxy resin reinforcement in quarry has the following advantages:

- Reducing the amount of excavation required for the same amount of marble extracted;
- Reducing the amount of waste material such as rubble, flakes and fragments, which would be produced if a broken and less "healthy" block were to be cut, thus optimizing production and increasing the yield percentage in excavated blocks. In the Apuan Alps marble district, the average yield of a quarry is estimated to be between 20 percent and 50 percent in terms of marketable marble blocks [54,60]. Resin application in quarries has the potential to boost this percentage (although an accurate estimate is not easy

because it depends on the condition of the quarry—whether open-pit or tunnel—as well as the marble material and its state of fracturing);
- Increasing the safety of the operator in the quarry (a fragmented and unreinforced bench may not follow a predictable pattern during tipping);
- Making the transport of extracted blocks to processing plants safer.

It should be noted that these technologies are still considered experimental and are not yet widely adopted across all quarries in the Carrara region. Consequently, there is a lack of extensive literature or technical documentation supporting our dissertation.

In addition, the use of resins in quarries should be the subject of investigation, as the use of these materials can lead to environmental problems related to the use of potentially polluting chemicals. Indeed, bi-component epoxy resins, in which one of the substances used is often bisphenol A (BPA), which reacts with a second component, are often used for reinforcement and consolidation of stone blocks, both in the quarry and in working plants. If the epoxy resins are discharged into wastewater, BPA residues could leak into the environment. However, there are no reliable data on how BPA could leak out of the epoxy matrix and in what quantities. The amount of BPA residues in final applications depends on the type of epoxy resin used and also the process employed. In addition, BPA could be degraded once it enters the environment, further complicating estimates (https://epoxy-europe.eu/wp-content/uploads/2015/07/EPOXY-Leaflet_Safety_web.pdf, accessed on 2 April 2024).

Therefore, when using these technologies in the stone cutting industry, whether in the quarry or during processing in plants, care must be taken both during application to the blocks and afterwards, managing the possible residues generated as best as possible. Indeed, in the case of cutting fully resin-impregnated marble blocks, a mixture of dust, water and resin residues could be generated during the cutting process, which (as well as the other waste materials from quarries) may be dispersed into the environment and wastewater.

Consequently, for reuse and recycling objectives, they should be treated differently than "traditional" sludge generated in the quarries and/or working plants that do not use resins (for example, subjecting them to more in-depth chemical analysis). Targeted research is in progress to determine the specific impacts (economic and environmental) associated with the use of resins to consolidate both bench (at quarry yard) and blocks (at working plant).

2.3. Fine Waste and Sludge from Carrara Marble Basin (CMB)

As already introduced, the marble industry can produce marble waste at a proportion of about 30% (sludge) from quarry production, in relation to the volume of the excavated marble, 20–30% from dust produced during processing operations, and 5–7% resulting from polishing activities (in working plants) [27,29]. Consequently, as a result of the revenues generated by marble industries, about 200 Mt of marble waste from marble processing industries, as a waste powder or sludge, has been produced worldwide [61]. Such a volume of fine waste materials, if not managed well and (potentially) recovered/recycled can pose serious problems for the environment (i.e., on surface and underground water) and on the balance sheet of marble enterprises (due to the costs for their disposal. Such costs in Italy are about 30–35 EUR/t [28]).

Regarding waste production, the Carrara Marble Basin (CMB) quarries have generated an annual average of approximately 3 Mt/year over the past few decades, with figures showing a gradual decline from about 2.7 Mt in 2012 to 2.2 Mt by the end of 2017 [54]. This waste encompasses various materials, including coarse marble waste such as white flakes, dark flakes, defective-fractured marble blocks, and crushed slabs, as well as fine waste materials. In the CMB, two types of fine waste are generated. The first includes all the fine particles of rocks and soils generated during the excavation process to reach the marble deposit, commonly referred to as *terre* or *tout-venant*. Taking data from the Municipality of Carrara as a reference, it is estimated that in the period 2005–2016, a total of about 4 Mt of *terre* was extracted from the Carrara quarries, corresponding to an average of about 360.000 t/year [62]. More recent data indicate a decline in their production, from 223.276 t at the end of 2012 to 142.610 t at the end of 2017. Overall, these fine materials from excavation contribute to approximately 8% of the total quarry debris, with the remainder comprising white flakes (48.9%), dark flakes (40.6%) and defective blocks of marble, commonly referred to as "reefs" (1.8%).

The second type of fine waste generated in the CMB depends on the specific production context and marble processing technology employed; for example, fine waste generated during the extraction and processing of marble blocks in the quarry (quarry sludge), or those materials generated during the processing of blocks in working plants using diamond frame saws and multi-wire diamond machines ("sawdust sludge" or "residual sludge" [28]). Quarry sludge and residual sludge in the CMB, as well as throughout the Apuan Alps region, are commonly referred to as "*marmettola*".

In particular, with regard to fine waste produced in quarries (quarry sludge) we can identify two specific sub-categories depending on the extraction technology:

1. Fine waste produced by marble quarrying using a toothed chainsaw (Figure 5a) results in dry powder due to the absence of water during cutting operations, consisting mainly of very fine particles of rocks (varying in size from sand to silt) and sometimes, traces of oils and/or greases and hydrocarbons, accidentally leaking from the cutting machines or traces of metal resulting from the wear of cutting tools [63]. Also, as explained by [64,65], employing the toothed chainsaw generates a powder consisting of calcite with a grain size ranging from medium to fine sand (0.5–0.125 mm), consisting, microscopically, of sharp-edged granules and fracture surfaces concentrated along the cleavage planes. Finer granules (8–32 μm), probably generated by friction between the larger grains during cutting operations, can be recognized;
2. The use of water during excavation (as occurs during the use of diamond wire technologies) mainly causes the formation of a sludge, or mud (Figure 5b), consisting of a mixture of water and the finest fraction of cutting residue (<62.5 μm; silt), but usually presents with the same appearance microscopically as the particles produced with the previous technology.

(a) **(b)**

Figure 5. Examples of fine waste generated in Carrara marble quarrying operations; (**a**) marble quarry sludge from toothed chainsaw machine; (**b**) marble quarry sludge from diamond wire machine (available online: https://www.toscanachiantiambiente.it/la-marmettola-un-problema-ambientale-infinito-per-il-distretto-del-marmo/, accessed on 2 April 2024). Both wet and dry fine fractions can be produced; in particular, as for the area characterized by the presence of springs, the use of dry cutting is preferred, as it does not pollute water.

These materials must be managed as waste, and, consequently, the production volumes must be declared, annually, by the individual producing company, in the Single Environmental Declaration Form ("MUD"). Analysis of the MUD from companies in the Apuan Alps marble district suggests that most of the sludge production reported (about 200,000 t/year [63,66]) originates from working plants located downstream of the extraction sites.

In working plants, diamond blades are employed both in single- or mono-wire technology to square irregular blocks (largely employed for squaring both hard and soft stone blocks), and in multi-blade saw technology (the most commonly employed in Italian working plants to cut soft stone-like marbles). In the latter technology, a frame gives an oscillating movement to a set of blades, which are distanced according to the desired thickness of the slabs to be produced, and are used with water in order to cool the tools [67]. A detailed estimation of the quantities of marble sludge produced in quarries is not easy, as it depends not only on the volume produced, but also on the areal extent of all the cuts required to produce the blocks. In some cases, companies provide data on the thickness of the cuts, from which the volume of fine material produced per m^2 can be deduced, and on the cutting speed, from which the daily production (per chainsaw or diamond wire cutting machine) can be estimated. An acceptable estimate of marble quarry sludge production is assumed to be about 4–5% by volume of the total production of marble blocks, thus corresponding to about 46.000 t/year for the CMB alone [64,68].

The total fine waste produced in the quarry, including that from excavation (*terre* and *tout-venant*) and quarry sludge can thus be estimated to be around 0.5 Mt/year for the CMB.

Sludge from marble quarrying in the CMB is managed as (special) waste. If it is not reused in the quarry cycle, according to Italian Legislative Decree 117/08, it must be collected at the source for recovery and treatment (for instance, in the case of wet sludge, dehydrated), before being properly disposed of in designated landfills for inert waste, in accordance with Italian Legislation 152/2006 (CER-010413—waste from stone cutting and sawing). The sludge from marble quarrying in the CMB (and the Apuan region at large) is mostly collected (if the concentration of polluting elements in the waste does not exceed the limits set by Italian legislative Decree 152/2006) by authorized waste treatment companies and then sent to factories that use it in their own production stages as a neutralizing agent for acid effluents (for example, by mixing it with residues from the production of TiO_2) [69].

In the CMB, there is a large amount of marble sludge, along with fine materials from quarry coarse debris (*terre* and *tout-venant*) abandoned in the quarry areas [70]. It is evident that, without appropriate management, these waste materials (both *terre* and *tout-venant* from excavation and marble sludge), given the predominance of fine particles (silt class), can create a considerable negative impact on the environment, contaminating water and air, also with harmful effects on plants, animals and human health [71–74].

According to Aukour et al., 2008 [75], the existing marble establishments distributed in Zarqa Governorate (northwest of the capital city Amman, Jordan), violated the land use classification and municipality limits. According to the site characteristics during and after quarrying operations, marble production negatively and directly damaged the existing landform through the intrusion of urban development and the loss of attractive features such as vegetation and hills.

The major impact produced by marble manufacturing was the direct negative social impact. Particulate dust was the main environmental impact of marble production produced during the marble cutting phase. Dust is a generic term used to describe fine particles suspended in the atmosphere, produced during exploitation and also processing operations (disturbance of soil and/or rocks through mechanical action). The resulting dust particles from marble factories have high levels of toxic PM particles and exposure to it is a root cause of many fatal respiratory and carcinogenic diseases—such as nasal cancer, bronchitis, asthma and lung infection—in marble workers. Research conducted by Aukour et al., 2008 [75] showed that marble dust impacts were obvious in deteriorating adjacent soils, in which more than 5 cm accumulated at the surface, also affecting human, plant and animal growth in the surrounding community. In addition, in all of the marble production sites, as noted also by the local community, the noise resulting from traffic was much less than the noise produced during the cutting stage in marble production. Although the noise levels in the nearby residential area associated with the marble production companies remained below 60 dB (A) in both instances, the residents continued to be disrupted by the noise. Furthermore, the establishment of the marble infrastructure contributed to heightened traffic, particularly from heavy diesel vehicles making frequent trips—typically exceeding 10 trips per day based on the production volume of each plant.

A similar study was conducted in the Hayatabad residential area in Peshawar, Pakistan [76], that performed an air pollution analysis and air quality assessment. Based on the results, it was found that the air near the residential area had higher $PM_{2.5}$ and PM_{10} (lighter marble dust particles) levels than the World Health Organization (WHO)-recommended guidelines.

Furthermore, fine waste materials from marble quarrying can exert a polluting effect on water bodies through various combined effects [63]:

- Biological impact: Marble fine waste causes abrasion of the bodies and gills of macroinvertebrates and a reduction in food availability. As a result of the sedimentation of marble sludges, carbonate material is found on the riverbed, which hinders colonization, buries eggs and life stages attached to the substrate and destroys the diversity of microenvironments;
- Physical effects: marble sludge increases the degree of turbidity of river waters;
- Chemical effects: watercourses, deprived of the biological populations responsible for their self-purification, also carry a large part of the unchanged organic pollutant load.

Marble sludge, infiltrating by means of rainwater into underlying karst aquifers through solution cavities, is also responsible for the high vulnerability of groundwater bodies, which is particularly critical for carbonate rocks such as marble and meta-dolostone that make up the Apuan Alps region. One of the clearest examples of the impact of marble sludge on the Apuan Alps karst system are the springs located near the village of Equi Terme (Massa). These springs are fed by the northern sector of the Apuan Alps karst aquifer, which is mainly composed of marble and meta-dolostones, and is characterized by high and very rapid fluctuations in water flow. Marble quarrying is the main human activity occurring in the catchment area. During the dry season, the discharge of the springs can decrease down to 0.03–0.04 m^3/s, while during major floods, it increases to a total flow rate of 15–20 m^3/s. During floods and for a few days afterwards, the water becomes very turbid, but a slight whitish opalescence may persist for several days afterwards. During minor floods, the water remains clear until marble sludge (*marmettola*), washed out of the quarries and infiltrating the catchment area, reaches the spring through a network of highly conductive conduits [64].

Studies by Giannecchini et al., 2016 [42,77] have highlighted another substantial issue regarding fine marble waste, particularly that related to extractive waste facilities, where both fine particles and coarser fractions from pre-existing landfills, commonly referred to as "*ravaneti*" (particularly common and large in Apuan Alps quarries, see Figure 6), pose significant challenges. Indeed, large quantities of fine particles of marble sludge from quarrying activities, together with soils and *terre* from excavation, are dumped into them. Fine particles, since they show poor geotechnical properties, can penetrate into these old quarry dumps, worsening their stability conditions and often leading to instability phenomena, which can generate landslides and debris flows. The continuous discharge and accumulation of material, excavation, slope cutting or road construction often result in the overloading of the waste and/or exceedance of the friction angle, leading to slope instability. In addition, heavy rainfall, rainstorms and stream erosion are probably the most significant triggers of debris flows involving quarry waste, posing a threat to nearby populations, villages and infrastructure. Indeed, in 2003, a rainfall event of exceptional magnitude hit the CMB and caused an overflow of the main water collector of the marble basins of Carrara, the river Carrione (showed in Figure 6). The river overflow occurred due to the accumulation of material in the riverbed caused by the influx of substantial amounts of debris into the hydraulic network, resulting from landslides and the mobilization of *ravaneti*. This event led to significant damage, flooding numerous homes and factories and, unfortunately, resulting in the loss of one life.

Figure 6. (**a**) The Carrione river (in the municipality of Carrara) turns whitish after rain due to the presence of marble sludge (yellow arrow) coming from the quarry basins upstream; on the left, the blue arrow indicates clear water from an area without quarries (modified from [63]); (**b**) examples of quarry waste dumps (*ravaneti*) in the Carrara basin (modified after [77]).

The marble quarries in the Apuan Alps have been long and widely studied in many environmental respects, including marble formation volumes [78], rock mass stability [79] and restoration perspectives [80]. However, the real effects of marble sludge on groundwater, along with the processes governing its transport through karst aquifers, which are among the most important environmental issues to be considered, have not always been studied in detail.

Figure 7 shows an overview of the entire marble production line (from the first stage of quarrying to processing into finished products) and the relative types of waste produced during each stage.

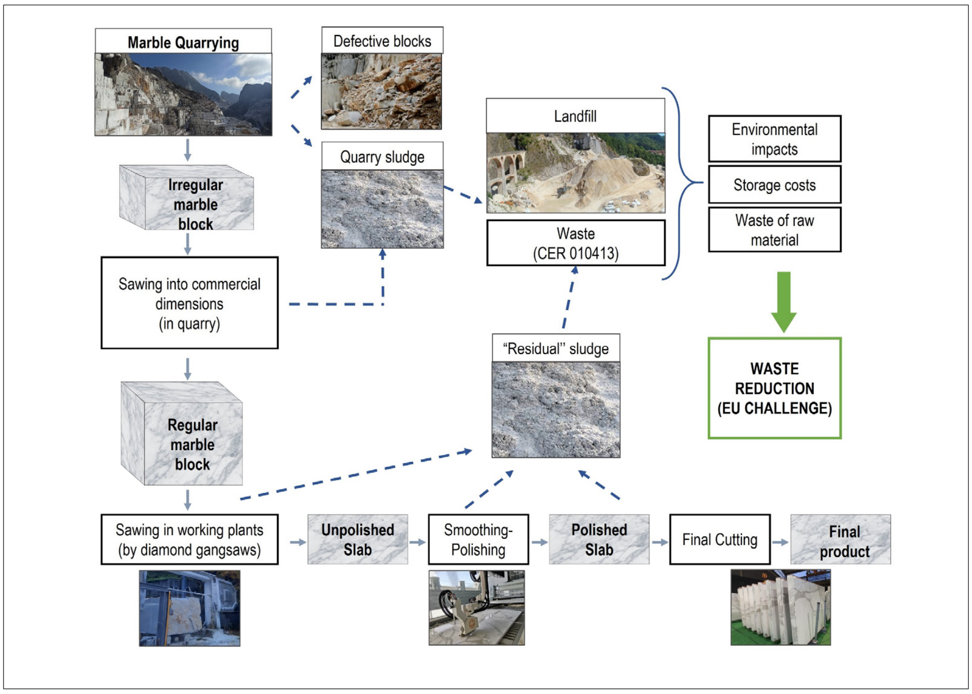


Figure 7. Schematic overview of marble quarrying and processing activities (continuous blue lines) with a focus on sludge (quarry sludge and residual sludge) generation (dotted blue lines) and management issues (green arrow).

3. Material and Methods

The first step of this research focused on the acquisition of general information about the marble extraction context, concerning primary ornamental materials production and data about quarrying waste. The data, collected by means of surveys in quarries of the CMB, include an overview of the CMB within its primary extraction basins, the main extraction technologies and production data for both primary ornamental blocks and quarrying waste, especially the finest fractions (the characteristics of which are described in Section 4.2) and their management strategies.

The second phase of the present research aimed to understand the characteristics of marble sludge produced in the CMB (both from quarries and from working plants, see Figure 8).

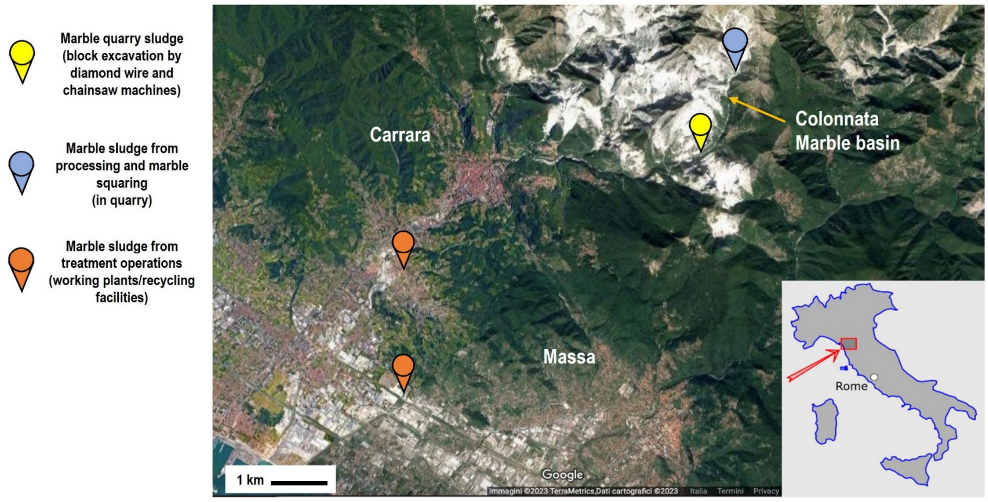


Figure 8. Sampling location of marble fine waste and sludge in CMB. The yellow arrow stands for the Colonnata marble basin, the easternmost of the 4 extractive basins of Carrara, while the red arrow indicates the location of CMB and Apuan Alps region in Italy.

The sampled waste materials (see Figure 9) include:

- Marble quarry sludge from primary marble excavation, mainly using diamond wire machines (wet sludge) and chainsaw cutters (microfine marble powder). The materials were sampled from a quarry from the Colonnata basin, and were collected immediately after the cutting phase, to preserve their original characteristics;
- Marble quarry sludge generated by processing the marble block once quarried (diamond wire squaring). The materials also come from a quarry in the Colonnata basin, and were sampled directly from sludge storage areas in the quarry area (e.g., settling tanks and/or filter bags);
- Marble sludge generated by marble squaring (also by diamond wire machines) in working plants (a necessary process for those blocks still retaining irregularities and requiring further resizing before the slab production phase with diamond frame-saws);
- Marble sludge deriving from treatment processes (filter-pressed sludge), both in working plants and recycling facilities in the Carrara area.

Of particular relevance for this research are the first two categories of waste materials (marble quarry sludges), crucial for the objectives of our work, focused on waste recovery/recycling and mitigating environmental impacts, starting from the very beginning stages of quarry cultivation and processing.

All the marble sludge sampled in the CMB are listed—depending on their origin (quarry or working plants) and production technology—in Table 5.

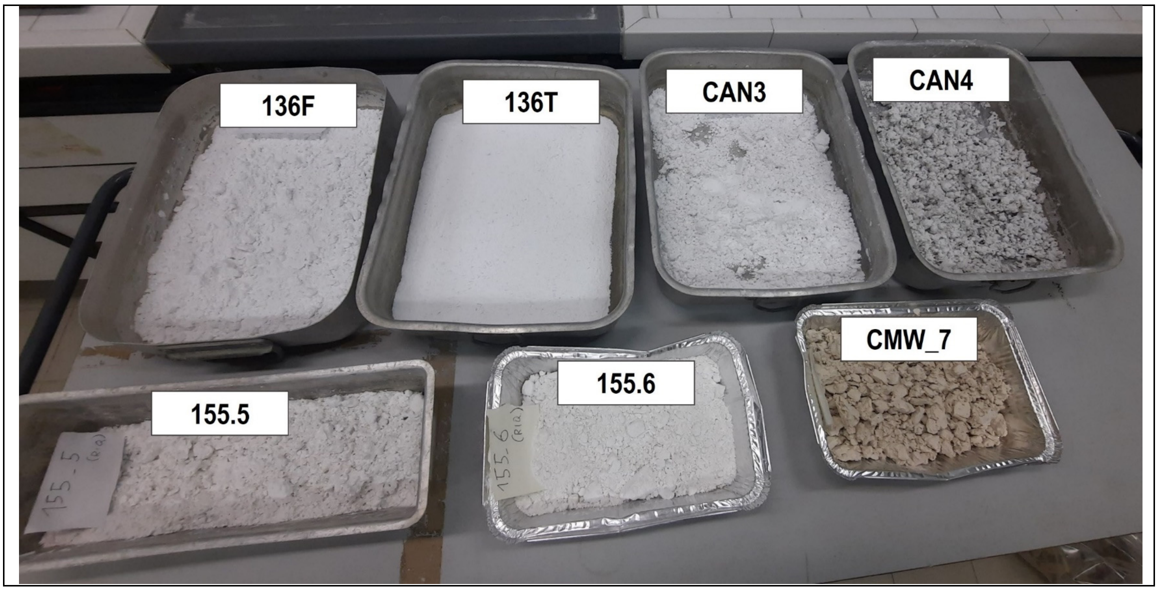


Figure 9. Representative samples of Carrara marble sludge in the laboratory.

Table 5. Summary table of marble sludge sampled in the CMB and its characteristics.

Sample n°	Type of Material (Marble Sludge)	Source/Location
136F	sludge from diamond wire machine cutting	quarry
136T	sludge from chainsaw machine cutting	quarry
CAN3	sludge from processing and marble squaring	working plant
CAN4	sludge from treatment operations (filter pressed/dewatered sludge)	working plant
155.5	sludge from processing and marble squaring	quarry
155.6	sludge from processing and marble squaring	quarry
CMW_7	sludge from treatment operations (filter-pressed/dewatered sludge)	working plant/recycling plant

At first, to find out the pollutant levels in the waste materials needed for proper landfill management, some of the marble sludge samples were sent to accredited labs for leaching tests, following the method EN 12457-2 [81] and UNI 10802 [82], in accordance with main Italian Legislation (Annex 3 D.M 5/02/1998, concerning the recovery of non-hazardous waste, then replaced by D.M. 5/04/2006 n.186).

After on-site sampling, laboratory analysis of marble sludge was carried out to provide insights into its physical, geotechnical, mineralogical and morphological characteristics. First, it was put in an oven to dry at a temperature of 100 ± 5 °C, to determine the water content. Also, before starting the analysis phase, a reduction in the samples (by coning and quartering method) was performed, to obtain representative sub-samples to a convenient size for testing.

The analytical procedure followed for the analyses and for obtaining the representative samples is schematized in Figure 10.

As for the physical–geotechnical characteristics, grain size analysis and Atterberg limit measurements were carried out. Given the very fine particles expected, grain size analysis was carried out by the wet method, according to UNI EN 933-1 [83] and UNI EN 933-2 [84], using ASTM sieves (interval 1–0.5–0.250–0.125–0.063 mm), together with the sedimentation method for particles < 0.063 mm. Atterberg limit measurements were performed on the particles < 0.475 mm for liquid limit and plastic limit determination, following ASTM D4318 [85].

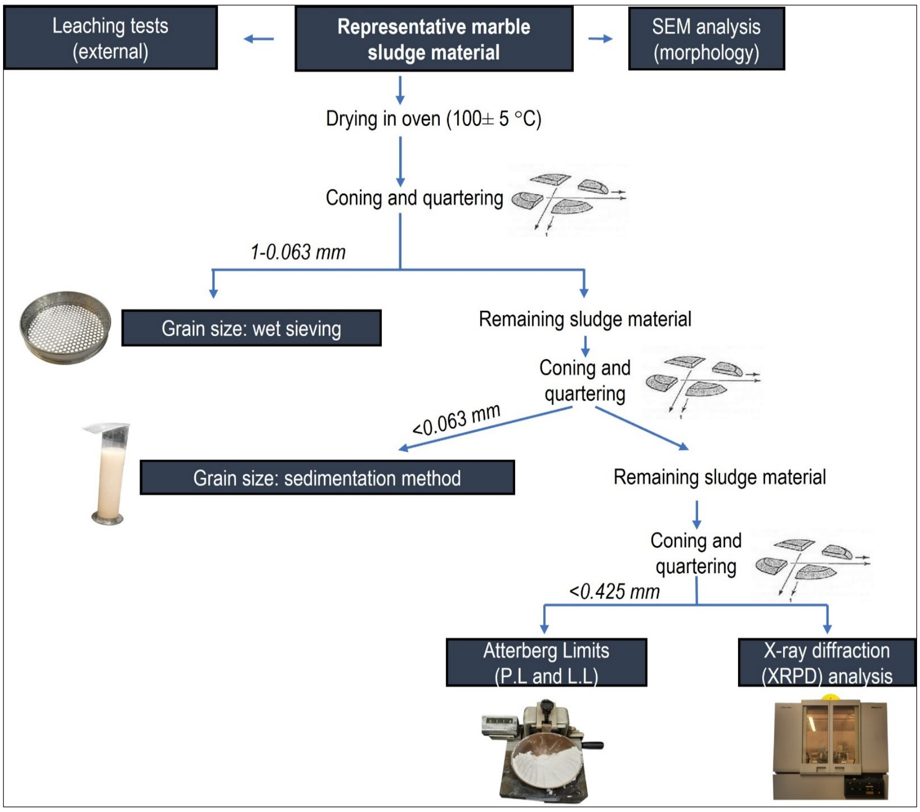


Figure 10. Analytical procedures on Carrara marble sludge samples.

Mineralogical analyses were carried out using X-ray powder diffraction (XRPD), a versatile, non-destructive analytical technique used to identify and qualitatively determine the crystalline phases present in powders and solid samples. The instrument used was a PANalytical X'Pert PRO PW3040/60 powder diffractometer (Department of Earth and Environmental Sciences, Milano, Italy), in Bragg–Brentano parafocalising geometry θ-θ, with a rotating sample holder (spinner). Semi-quantitative phase analysis was performed by the Reference Intensity Ratio (*RIR*) method. Qualitative phase analysis was performed with X'Pert High Score version 2.1 (2.1.0), using the database *ICDD-PDF2-2004.*

Morphological quality of the marble sludges particles was observed using the VEGA-TESCAN scanning electron microscope (SEM). Before analysis, samples had to be prepared so that they could be placed inside the chamber of the instrument. A small portion of the marble sludge sample was placed on special aluminum supports (stubs) using conductive tape, and then metal-coated (Au). Once prepared, all the samples on the stubs were mounted on a stage, inside the Electron Column apparatus ("stage carousel"), which allows, with simple movement, the examination of different samples. High-resolution topographic images were obtained using the VEGA TESCAN Essence software, using an SE (secondary electron) detector.

The operating conditions were as follows:

- Energy = 10 KeV;
- Beam current (BC) = 100 pA;
- Working distance (WD) = 6 mm;
- Scan speed = 4;
- Vacuum mode: HighVac (chamber pressure < 0.08 Pa).

The maps were acquired at three different magnifications. The first image corresponds to the lowest magnification (around 350×), while the second image was obtained at an intermediate magnification (500×). The third image is a detailed view, with variable parameters that differ from sample to sample.

4. Results

4.1. Preliminary Findings about CMB Extractive Industry (Marble and Waste Production)

The initial stage of the research involved data collection and contextual information regarding the CMB in the Apuan Alps region. This preparatory phase provided an overview of the extractive activities in the investigated area, in particular regarding the production of materials for ornamental purposes and both quarry waste and sludge, before proceeding to the analytical phase in the laboratory. In addition to the available literature (ISTAT reports, data from the Municipality of Carrara and the Tuscany Region), dialogue with employees of some quarry companies and visits to quarries operating in the CMB were of primary importance for this purpose.

Concerning marble production in the CMB, PRAER ("Piano Regionale delle Attività Estrattive" [86]), issued by the Tuscany Region in 2007, as reiterated by subsequent decrees of the Tuscany Region (e.g., "Piano Regionale Cave" in 2009 and Regional Law 35/2015), have set the minimum marble block quantity to be used exclusively for processing into blocks, slabs and other finished products as 25% (compared to the overall production).

The analysis of the data obtained from the Carrara municipal weighbridge highlights that quarry waste materials (shapeless blocks, *terre*, *tout-venant*, white and dark flakes) constitute over 70% of the total material extracted from marble quarries. In detail, looking at the data for the period 2012–2017, the average amount of waste and debris from the Carrara quarries was around 2.5 Mt/year, with an average waste percentage of 73.7% of the total material extracted. More recent data, specifically from 2020, show that the total material extracted from the CMB quarries was about 2.8 Mt. Out of this total, 690.400 t was identified as ornamental blocks, constituting an approximate yield of 25%. The remaining quantity, totaling around 2.1 Mt (approximately 75% of the total production), includes quarry extractive waste and debris. These percentages of quarry waste confirm the trend of previous years described above, and seem to be in accordance with PRAER regulations on block debris production volumes.

As for the market dynamics of the excavated marble blocks in the CMB and the entire Apuan basin, they are closely tied to the quality of the extracted marble and its distinct varieties. Particularly, premium marble varieties like "Statuario" or "Calacatta" are valued highly, reaching up to EUR 5.900/t for a first-choice block, EUR 2.600/ton for a semi-squared and defective block, and EUR 400/t for a shapeless block. These luxury goods are suppled to international markets, including countries such as Saudi Arabia, the USA, India, Australia and Brazil. In addition to these prestigious materials, a diverse range of moderately priced marbles are available in the area, including varieties like "Arabescato" and certain veined marbles like "Venato C" (about EUR 550/t for a first-choice block, with shapeless blocks of the same variety costing a few hundred EUR/t). The reference markets for these materials may be more localized, predominantly within Mediterranean countries such as Italy itself or even extending to China. These insights were further enriched by integrating data from the marble product sheet, which is a document provided by the Municipality of Carrara that details the primary selling prices for each marble variety.

4.2. Carrara Marble Sludge Characterization

This section aims to investigate deeper into the characteristics of the fine materials and quarry sludge generated in the CMB. The quantity of marble sludge generated from quarry operations has been estimated to be approximately 5% of the total extracted material, as previously described in Section 2.3. Therefore, the provided data align with the findings reported in recent studies [64,68].

The distinctions between sludge samples from quarrying activities and those from other treatments operations, such as dewatering and filter-pressed sludge, were primarily noticeable at a macroscopic level. Despite all having very fine grain sizes, the latter exhibited more grayish or brownish colors, while the quarry samples were all predominantly white. This color disparity was attributed, as revealed by subsequent mineralogical analyses, to the almost exclusive presence of calcium carbonate. Furthermore, some displayed a muddy appearance, especially those derived from diamond wire machine excavation and squaring operations (which imply the use of water), while others had a drier powder consistency. Overall, all the waste samples were found to be odorless in their solid state.

The results of the grain size analysis showed, for all the samples, a passing percentage of particles through a 0.063mm sieve of 80% (on average). More specifically, as observed from the particle size distribution, they fall almost entirely within the silt particle size fraction (<0.063 mm), with no clay particles < 0.002 mm (Figure 11). The calculated uniformity coefficient (d_{60}/d_{10}) is around 1, except for sample 136T (marble sludge from chainsaw machine cutting) which showed a coarser grain size than the others (silty sand) and a higher U (≈6). The results obtained from the grain size analyses are also in agreement with those obtained from the Atterberg limit tests (liquid limit and plastic limit), which describe a non-plastic behavior of the samples (P.I < 5), most likely due to the absence of clay particles < 0.002 mm (2 μm).

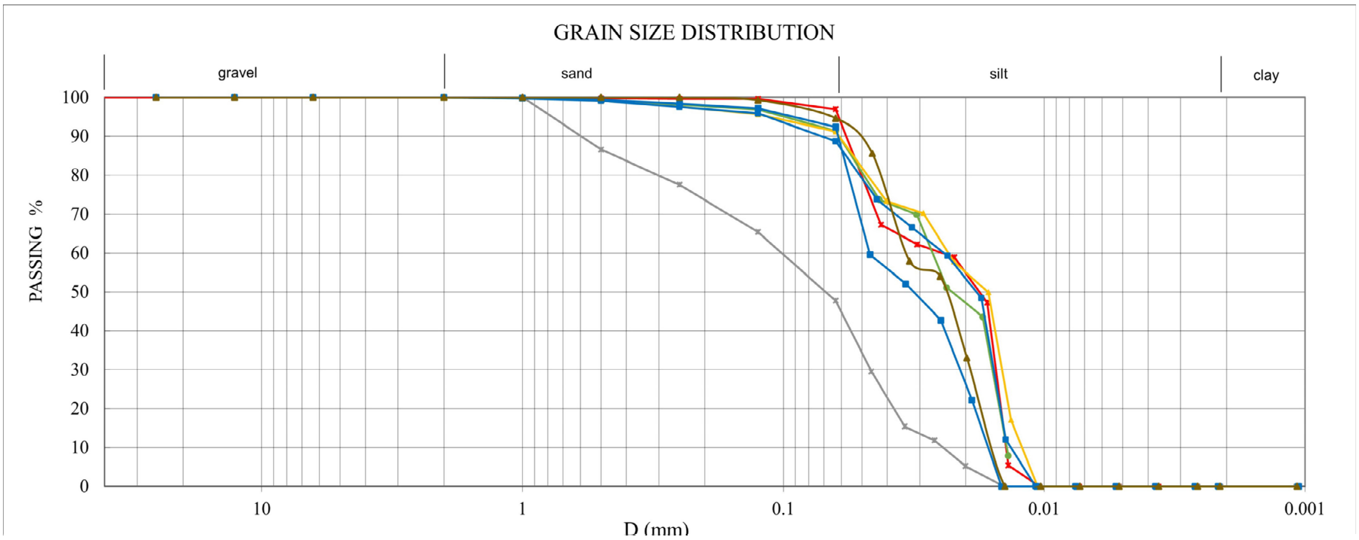


Figure 11. Grain size characteristics of Carrara marble sludge samples (grey line: marble sludge from chainsaw cutting–in quarry; sludge from diamond wire cutting–in quarry; blue lines: sludge from processing and marble squaring–in quarry; green line: sludge from processing and marble squaring–in working plant; red line; brown and yellow lines: filter pressed/dewatered sludge–in working plant and processing facilities).

X-ray diffraction analyses have shown that all the samples are composed almost entirely of calcium carbonate (>90% wt.), usually in the form of pure calcite or magnesian calcite (Figure 12a). Other components, even if in very low concentrations, are muscovite (varying around 5–10% wt.), dolomite (<1% wt.) and quartz (0.5–1% wt.). In Figure 12b–d, a comparison is presented between marble sludge obtained from a diamond wire machine (sample 136F) and chainsaw machine (sample 136T), both originating from primary excavation in the quarry. The calcite content is comparable in both samples, with a slightly higher concentration observed in the sludge derived from the chainsaw machine, and the same is seen with muscovite. On the other hand, the sample from the diamond wire cutting showed a higher dolomite content than that of the chainsaw sludge. In samples 155.5 and 155.6, originating from the squaring of marble from a quarry (also by a diamond wire machine), calcite remained the predominant component (>95% wt.), but dolomite was not noted. Traces of quartz were observed, though at very low concentrations (0.5–1% wt.). The marble sludge sample from the filter press treatment (dewatered sludge; CMW-7) exhibited higher mineralogical variability compared to the other samples. Apart from calcite (still the main component, at 91% wt.), dolomite (2% wt.), muscovite (1% wt.), and quartz (1% wt.) were detected.

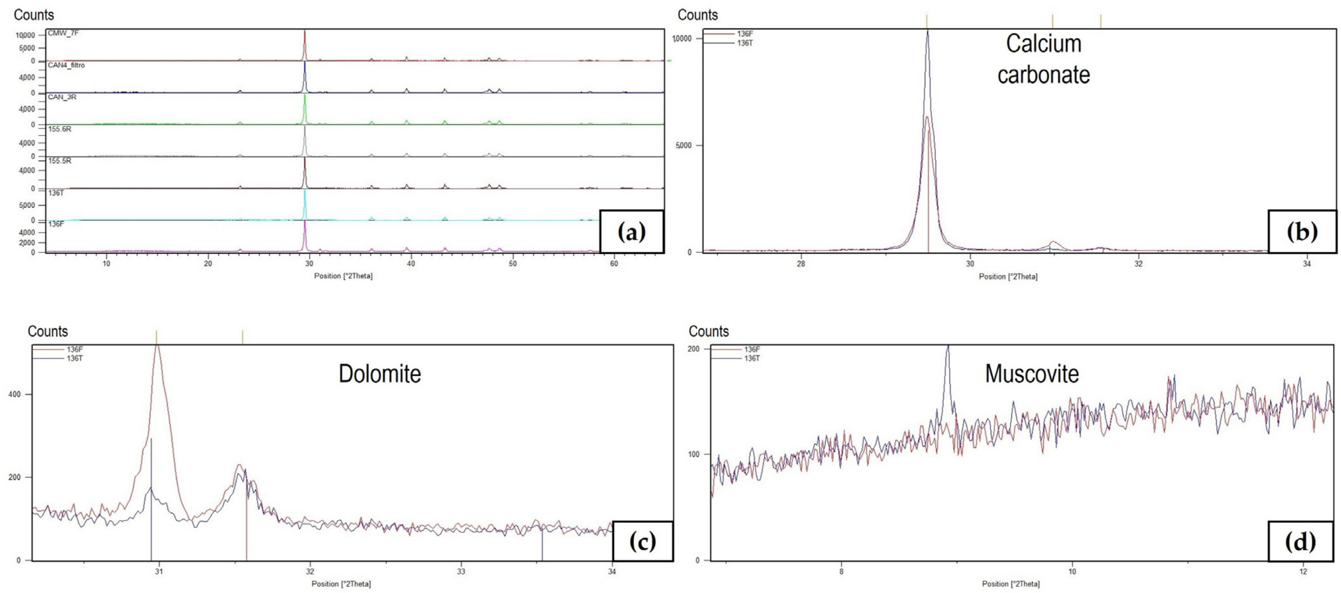


Figure 12. Mineralogical analyses of Carrara marble sludges examined in this study: (**a**) XRD patterns comparison of all sludge samples. From the top to the bottom: filter pressed/dewatered sludge–in working plant and processing facilities (red and blue lines); sludge from processing and marble squaring–in working plant (green line); sludge from processing and marble squaring–in quarry (grey and brown lines); marble sludge from chainsaw cutting–in quarry (light blue line), sludge from diamond wire cutting–in quarry (violet line); (**b**–**d**) XRD patterns comparison between marble quarry sludge from diamond wire cutting (red line) and chainsaw cutting (blue line).

SEM observations further confirmed that the Carrara marble sludge is composed entirely of calcium carbonate grains. Furthermore, SEM image processing revealed that most of the carbonate grains range between 8 and 10 μm up to 100–150 μm (fine to very fine sand and silt). Marble sludge produced by diamond wire machine cutting (Figure 13a,b) shows carbonate grains which range from a maximum of 100 μm to around 5 to 8 μm, with angular structures, sharp edges and different fractures along the crystal cleavage planes. Marble sludge from chainsaw machine cutting (Figure 13c,d) shows carbonate grains of the medium to fine sand range (up to 250–300 μm), which therefore are of a larger grain size than that of the sludge resulting from diamond wire cutting. Regarding the grain morphology, on the other hand, no major differences are noted compared to the diamond wire sludge, with calcium carbonate grains having an angular and fractured appearance.

Marble sludge from treatment operations (filter-pressed sludge in working plants and recycling plants) (Figure 13e–h) show similar characteristics to marble quarry sludge, such as the overall grain size and variability in clast sphericity and rounding. However, some of the clasts in these samples appeared to have less distinct fractures compared to other samples, where the granules seemed very "broken". Additionally, other than coarser grains, our observations revealed smaller particles and grains (around 8–10 μm) that were very irregular, forming some kind of aggregate or groundmass, and which were less pronounced in the other samples.

The results of the leaching test showed very low metal concentrations in the Carrara marble sludge (or not even detectable levels), and therefore, in any case, below the legal limits set out by Italian Legislative Decree 152/2006. The analysis of the carbonates (as $CaCO_3$) further confirms the almost pure composition of the Carrara marble sludge, with a $CaCO_3$ content always above 90% (%s.s.), especially for marble sludge from diamond wire machine and chainsaw machine cutting operations (Table 6). In these samples, the only components detected as being slightly above the limits seem to be Fluorides (F^-) and, in the case of the chainsaw quarry sludge, high values of hydrocarbons were found.

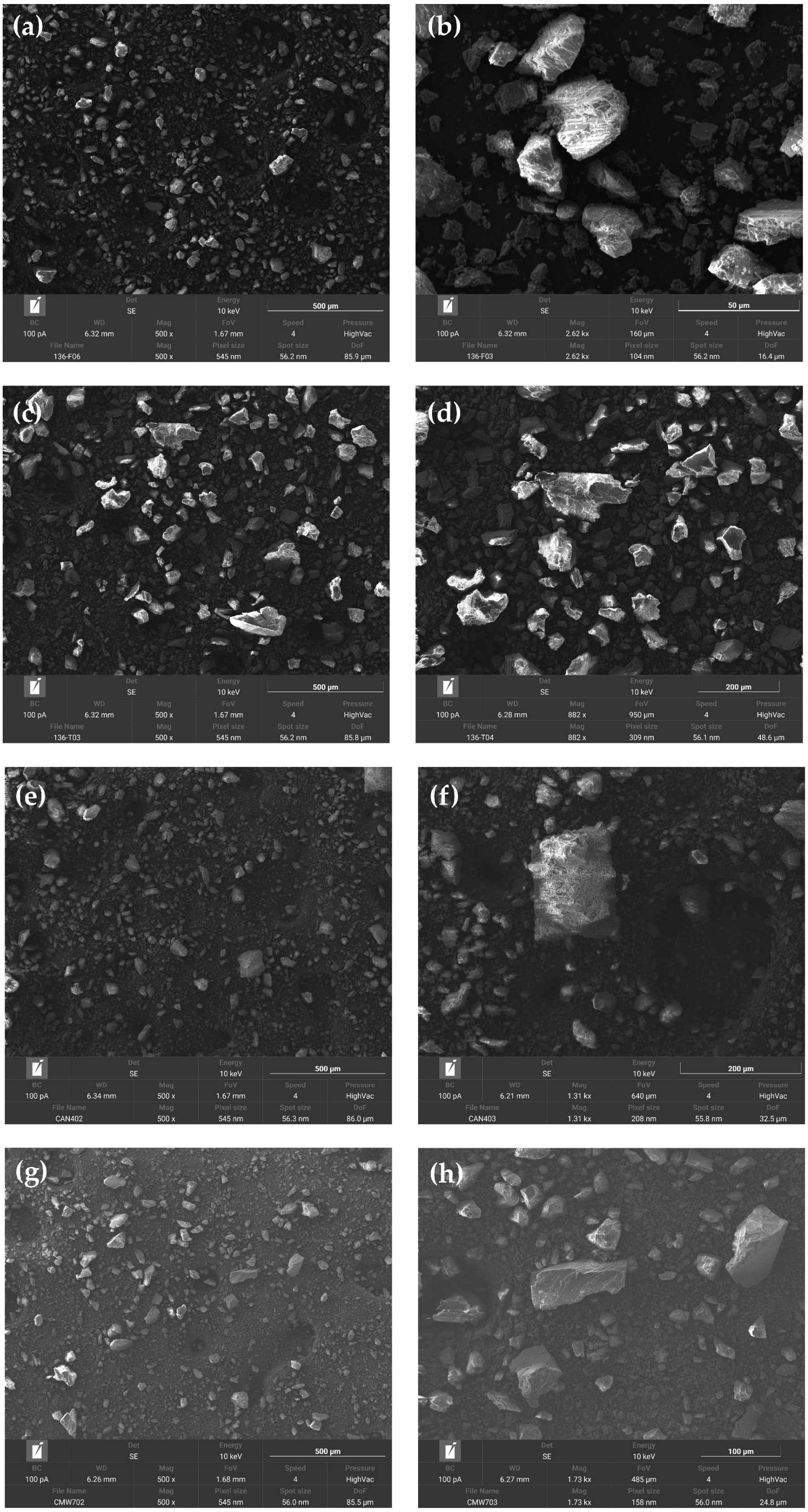


Figure 13. SEM images of calcium carbonate grains in the Carrara marble sludge: (**a**,**b**) marble quarry sludge from diamond wire machine cutting; (**c**,**d**) marble quarry sludge from chainsaw machine cutting; (**e**–**h**) sludge from treatment operations (filter-pressed sludge in working plant and recycling facilities).

Table 6. Leaching tests on Carrara marble quarry sludge. "NV" = not valuable because lower than quantification limit.

			Sample n°	**136F**	**136T**	**T**
				Marble Quarry Sludge (Diamond Wire Machine Cutting)	**Marble Quarry Sludge (Chainsaw Machine Cutting)**	**Marble Quarry Sludge (Backhoe Machine Cutting)** [1]
Parameter	**Mass Unit**	**Law Limit**	**Quantification Limit**	**Result**	**Result**	**Result**
Residue at 105 °C	%	-	-	79.2	100	100
pH	unit of pH	5.5–12	-	8.1	8.1	8.1
Carbonates ($CaCO_3$)	%s. s	>85	0.5	94	92	95
As	µg/L	<50	5	NV	NV	NV
Ba	µg/L	<1000	50	NV	NV	NV
Be	µg/L	<10	2	NV	NV	NV
Cd	µg/L	<5	1	NV	NV	NV
Co	µg/L	<250	10	NV	NV	NV
Tot. Cr.	µg/L	<50	5	NV	NV	NV
Ni	µg/L	<10	2	NV	NV	NV
Hg	µg/L	<1	0.5	NV	NV	NV
Pb	µg/L	<50	5	NV	NV	NV
Cu	mg/L	<50	5	NV	NV	13
Se	µg/L	<10	5	NV	NV	NV
V	µg/L	<250	10	NV	NV	NV
Zn	µg/L	<3000	10	NV	NV	NV
Cyanurium	ug/L	<50	10	NV	NV	NV
Chlorides (Cl^-)	mg/L	<100	10	NV	NV	NV
Fluoride (F^-)	mg/L	<1.5	0.2	0.39	0.44	0.47
Nitrate ($NO_3{}^-$)	mg/L	<50	5	NV	NV	NV
Sulphates ($SO_{4)}$)	mg/L	<250	10	10	14	23
Asbestos	mg/L	<30	Not applicable	NV	NV	NV
C5–C9 hydrocarbons	mg/kg	-	5	NV	NV	NV
C10–C40 hydrocarbons	mg/kg	-	50	NV	479	NV

[1] Sample "T" was investigated only for leaching test and not analyzed with other samples in this work.

Table 7 shows the results of the leaching test on Carrara marble sludge (sludge from processing activities such as marble squaring in the quarry).

Also in these case, no potentially polluting elements were found in significant concentrations (below legal limits or not detected). Of note, as in previous samples, basic pH values (around 9).

In this study, marble sludge from the exploitation and processing of Carrara marble (both in quarries and working plants) was analyzed to understand its characteristics and possibilities for reuse/recycling.

The results obtained from this work, including the physico–technical characteristics (grain size, Atterberg limits) and mineralogical compositions (XRD) of the waste samples, are summarized in Table 8.

Table 7. Leaching tests on Carrara marble processing sludge (also from quarry). "Lower than (<)" = the result is under the verified quantification limit.

Sample n°	155.5		155.6			
	Sludge from Processing and Marble Squaring (Quarry)				Sludge from Processing and Marble Squaring (Quarry)	
Parameter	**Mass Unit**	**Result**	**Parameter**	**Mass Unit**	**Law Limit**	**Result**
Dry residue at 105 °C	%	76.2	Dry residue at 105 °C	%	-	76.2
Fluoride (F^-)	mg/L	-	Fluoride (F^-)	mg/L	1.5	<0.50
Sulphates (SO_4)	mg/L	-	Sulphates (SO_4)	mg/L	250	6.2
Chlorides (Cl^-)	mg/L	-	Chlorides (Cl^-)	mg/L	100	10
Nitrate ($NO_3{}^-$)	mg/L	-	Nitrate ($NO_3{}^-$)	mg/L	50	<1
Al	mg/kg	<400	Al	-	-	-
As	mg/kg	<10	As	μg/L	50	<2.0
Ba	mg/kg	<40	Ba	mg/L	1	<0.020
Cd	mg/kg	<5.0	Cd	μg/L	5	<0.20
Co	mg/kg	-	Co	μg/L	250	<2
Tot. Cr.	mg/kg	<10	Cr.	μg/L	50	<2.0
Cr VI	mg/kg	<0.50	Cr VI	-	-	-
Fe	mg/kg	<400	Fe	-	-	-
Mn	mg/kg	<100	Mn	-	-	-
Hg	mg/kg	<5.0	Hg	μg/L	1	<0.10
Ni	mg/kg	<20	Ni	μg/L	10	<0.20
Pb	mg/kg	<50	Pb	μg/L	50	0.30
Cu	mg/kg	<10	Cu	mg/L	0.05	<0.0025
Sb	mg/kg	<50	Sb	-	-	-
Se	mg/kg	<50	Se	μg/L	10	0.26
Sn	mg/kg	<100	Sn	-	-	-
Tl	mg/kg	<100	Tl	-	-	-
Zn	mg/kg	<20	Zn	mg/L	3	0.0022
V	mg/kg	<100	V	μg/L	250	<2.0
Ca	mg/kg	260,000	Ca	-	-	-
Asbestos	-	-	Asbestos	mg/L	Not applicable	not detected
C5–C8 hydrocarbons	mg/kg	<5	C5–C8 hydrocarbons	-	-	-
C10–C40 hydrocarbons	mg/kg	1000	C10–C40 hydrocarbons	-	-	-
pH	unit of pH	9.6	pH	unit of pH	5.5–12	9.8

Table 8. Summary table of Carrara marble sludge characteristics obtained from this work. "n.f" = not found; "n.d" = not determined.

Sample n°.	136F	136T	CAN3	CAN4	155.5	155.6	CMW_7
	Marble Quarry Sludge (Diamond Wire Machine Cutting)	Marble Quarry Sludge (Chainsaw Machine Cutting)	Sludge from Processing and Marble Squaring (Working Plant)	Sludge from Filter Press Treatment (Working Plant)	Sludge from Processing and Marble Squaring (Quarry)	Sludge from Processing and Marble Squaring (Quarry)	Sludge from Filter Press (Working Plant/Recycling Facility)
water content %	15.5	1.9	22.2	19.9	16.9	2.7	20.0
% particles < 63 um (silt)	97.0	29.5	91.4	91.3	92.3	88.6	85.5
% particles < 2 um (clay)	n.f	n.f	n.f	n.f	n.f	n.f	n.f
Uniformity (U)	1.7	6.7	1.8	1.9	2.6	1.7	2.2
Grain size classification	silt	silty sand	weakly sandy silt	weakly sandy silt	weakly sandy silt	sandy silt	weakly sandy silt
L.L	n.d	n.d	n.d	n.d	20	23	20
P.L	n.d	n.d	n.d	n.d	18	20	18
P.I.	n.d	n.d	n.d	n.d	2	3	2
$CaCO_3$ % (from XRD)	96	92	96	83	>95	>95	91

5. Discussion and Conclusions

The present research serves as a thorough exploration into the current state of the art of the challenges and potential paths for more sustainable exploitation in a renowned marble district (such as Carrara), within the Apuan Alps region in Italy. The primary objective is to shed light on the promising aspects and challenges related to the fine waste generated in Carrara quarries, particularly microfine powder and sludge from primary marble extraction.

The results obtained from this research are promising, revealing that the fine marble sludge generated in Carrara quarries (*marmettola*) have very homogeneous grain sizes (silt) and are predominantly composed of $CaCO_3$ (90–95% by weight) with few other components (dolomite, quartz, muscovite), mainly inherited from the marble rock "at the source". Therefore, it has to be highlighted that the composition of the marble sludge studied in this work fits almost perfectly with the composition of the original marble. Also, the leaching tests conducted on the examined materials have shown that marble quarry sludge samples are virtually free of any level of contamination and hazardous substances with the exception of very few elements, such as hydrocarbons in the sludge sample from chainsaw quarrying (which probably leaked accidentally from the cutting machine itself). The latter sample also differs from the others in having a more heterogeneous and, on average, a coarser grain size (sand with silt), probably due to the characteristics of the tools used during the cutting of the marble bank (thickness of the chain saw blade of 3–4 cm), if compared to the diamond wire machine.

Overall, the marble sludge from Carrara quarries could potentially be reusable outside the quarry as nearly pure calcium carbonate "as such", without any further treatment (except for wet sludge that requires prior dehydration). This microfine sludge has promising characteristics (a high percentage of fine waste, low percentage of metals); thus, it could be reused in various industrial sectors that demand high-value micronized $CaCO_3$ products (e.g., filler in the manufacture of paper, paint, along with other chemical applications, like cosmetics and pharmaceuticals), as already demonstrated [6,35]. Additionally, there is the potential for its utilization in the construction sector for the manufacture of building products as substitutes for original materials, such as concrete, mortars (e.g., [63]), composite mixtures with lime and/or cement [37].

From an economic point of view, the implications of reusing marble waste are potentially significant and multifaceted. Utilizing marble sludge as a substitute for raw materials in various industrial sectors can lead to saved costs for manufacturers.

By using the sludge as a filler or component in products such as paper, paint, cosmetics and pharmaceuticals, companies may reduce the expenses associated with purchasing traditional raw materials. Quarry operators could potentially generate additional revenue streams by selling the marble sludge to industries looking for high-value micronized calcium carbonate products. This could serve as an alternative or supplementary source of income for quarry owners.

The availability of high-quality micronized calcium carbonate products derived from marble sludge could expand market opportunities for various industries. For example, manufacturers in the construction sector may find new applications for building products incorporating marble sludge, leading to the development of innovative products and potentially capturing new market segments, for example, as shown in a study by Dhoka [87]; "green concretes" which, employing waste produced by industries such as paper and pulp, mining waste, as well as marble dust from dimension stone, are therefore very often cheap to produce.

Concrete, indeed, is one of the most extensively used construction materials around the globe. The global production of concrete is about 12 billion tons a year, corresponding to almost 1 m^3 per person per year, causing it to become one of the largest users of the natural resources in the world. Currently, 4,100,000 thousand metric tons of cement is produced globally and this industry produces nearly 5% of the total man-made emissions of carbon dioxide [88,89]. A review conducted by Danish et al. [90] reported that the incorporation of marble fine waste in the preparation of concrete as a partial replacement of cement reduced the workability of cement and increased the water requirement, but on the other hand, the durability of the concrete exposed to an acidic atmosphere was increased. Finally, the use of stone waste (especially marble dust) reduced the overall construction cost and CO_2 emissions associated with concrete production.

Furthermore, selling fine fraction as a raw material in the abovementioned industrial fields allows for a reduction in the costs connected to "sludge" management and disposal. Indeed, reusing marble sludge instead of disposing of it as waste can have positive environmental implications. Minimizing waste generation reduces the need for landfill space and lessens the environmental impact associated with waste disposal. This could potentially lead to cost savings in waste management and contribute to a company's sustainability goals. The exploration of alternative uses for marble sludge may stimulate innovation and research in related fields. Companies and researchers may invest in developing new technologies and processes to optimize the utilization of marble sludge, leading to advancements in industrial practices and potentially creating intellectual property and competitive advantages.

Overall, the economic implications of reusing marble sludge from Carrara quarries are likely to be positive, encompassing cost savings, revenue generation, environmental benefits, market expansion, resource conservation and opportunities for innovation and research. Nevertheless, the extent of these implications will depend on various factors such as market demand, regulatory considerations, technological developments and the willingness of stakeholders to adopt sustainable practices.

However, our research has also highlighted several issues related to the marble industry that need to be taken into consideration.

The primary concern is related to the storage of the fine residues from the quarries, after the excavation stages and subsequent processing. In the CMB and, more broadly, throughout the Apuan marble district, it is common to observe quarry yards flooded with sludge and fine materials left from processing, often abandoned in the service area of the quarry and not collected adequately—such as directly vacuuming them after cutting and storing them in appropriate basins or filtering bags. The tanks designated for collecting sludge are commonly seen filled with various other materials. On one hand, this situation results in economic losses for companies. Failing to promptly collect these materials means missed opportunities for recovery and recycling, despite their promising characteristics, as highlighted in our research. Moreover, this behavior contradicts the principles of waste recycling outlined by the EU. Additionally, this practice gives rise to extensive issues impacting not only the environment, but also the local population. The direct effects on the environment, such as river turbidity and whitening, have made the local community particularly sensitive and critical, thus leading to the perception of every quarry-related activity as negative, especially in Carrara.

Furthermore, the negligent disposal of fine waste and sludge materials within the quarry area has implications for the company also. The presence of contaminants, notably hydrocarbons and oils derived from the frequent passage of vehicles (trucks, cars), as well as the operation of cutting machines such as toothed chainsaws, can lead to the potential pollution of the products, compromising their original composition (almost pure $CaCO_3$) and thus hinders their reuse as a secondary raw material (SRM) and forces their disposal as special waste with high economic and environmental burdens. Therefore, it is even more crucial to collect these materials "at the source".

Another source of concern is related to the sludge and residues from marble cutting in quarries that employ methods to reinforce defective marble benches and blocks, like resins (as already introduced in Section 2.2). These practices, while innovative and effective in improving cutting yield and enabling the excavation of defective blocks that would be otherwise unworkable through traditional technologies, may pose environmental issues. This is linked to the possible presence of potentially polluting chemicals (like bisphenol-A) within the resins themselves (for example, bi-component epoxy resins). These substances can be released directly into the environment or indirectly into the sawing fine residues, demanding additional attention compared to the other "traditional" fine sludge. To expand this topic, further investigations are needed, especially in quarries employing resin methods (not investigated in this research), to assess whether potentially pollutant fine sludge is being generated.

In conclusion, addressing the environmental impact of marble sludge in the CMB requires the immediate implementation of measures and best practices directly at the quarry site to mitigate these issues from the outset. Examples are as follows:

- Reduction in fine waste production through the use and implementation of dust extraction and storage facilities in the quarry, to be carried out immediately after the cutting phases;
- Monitoring the size, shape and chemical composition of inorganic particles that can be released in the air and also preventing the air dispersion of inorganic particles through irrigation;
- Attempting to implement and optimize new technologies of marble exploitation; for example, those for dry cutting, as already suggested in some studies [60]. Today, the machine used for dry cutting in CMB quarries is the diamond chainsaw, which produces residual materials with an irregular grain size, larger than that in the materials resulting from diamond wire cutting. Dry cutting with the latter is not possible today because, for safety reasons, the wire must be rubberized, and cutting without water would lead to the wire reaching temperatures incompatible with the rubber that covers it. In fact, water has a cooling and cleaning function. Dry cutting, therefore, could not only be functional in terms of saving water but also could be carried out with diamond wire in the production of higher-quality residual materials that can be immediately resold or reused;
- Improving waste management in the quarry, using proper temporary waste storage areas. In fact, as we described in this paper, the waste accumulated in the open areas of the quarry, whether it is marble sludge or *terre* and *tout-venant*, is subject to meteoric runoff, with the inevitable pollution of surface water, groundwater and springs. Therefore, the implementation of appropriate surface water regulation works, monitoring of groundwater levels, and quality monitoring of surface and groundwater is required;
- The collection and treatment of cutting wastewater. The turbid water is recycled and directed to a treatment plant (in a quarry) utilizing an immersion pump and bag filters, while marble sludge is periodically delivered to an authorized waste disposal company. Because the recycling of 100% of the water is not possible, and some of the water removed from production cycles is present in the waste fraction (sludge), it is urgent to ensure more efficient recycling, and develop more water treatment technologies (e.g., filter press technologies, decanters, etc.);
- Avoid dumping waste and debris on slopes inside the *ravaneti*, as described above. In fact, *ravaneti*, due to their high marble sludge and soil content, are also a very significant source of surface water, groundwater and spring pollution.

Positively reshaping the perception of these refined waste materials, especially in the CMB and throughout the entire Apuan territory, where this issue is still important, could turn them from perceived negatives into valuable resources, generating a positive impact on both the environment and society.

On a broader scale, the sustainability of the production of natural stones for building purposes, such as tiles and slabs, requires optimization of all phases of the production process, from exploitation to the final use. To ensure effective sustainability in the mining and extractive industry, it is imperative to address the following issues:

- **Planned management and organization of quarrying activity:** Start with a comprehensive understanding of mineral resources to plan land use and manage productivity effectively. A high level of knowledge about the deposit and its territory is indispensable for improving stone extraction;

- **Stone characterization:** The performance and productivity of a quarry is highly dependent on the type of machine used, which, in turn, is a function of rock characteristics. Machine specifications are generally readily known, but structural rock properties are not always readily available. The state of natural stress state and the fracturing of rock masses are key points in defining the possibility of extracting blocks of suitable size and volume. Structural anisotropy is a key control of the mechanical behavior of rocks under different environmental conditions, and the interaction between the crystal texture and its brittle mechanical behavior must be properly considered through appropriate laboratory tests. Indeed, if the extraction method causes significant fracturing of the stone blocks, alternative methods should be considered. It is therefore critical to conduct geotechnical investigations during the planning of work, combining the local structural features (for example, with rock fracture mapping) with the regional tectonic setting, to better understand the tectonic stress, local geological structures and morphology of the deposit. The workability of a rock is also linked to its textural characteristics and to its mineralogical composition. Therefore, the texture of the stone is another important factor to consider in the choice of cutting technology;
- **Reduction in quarry waste "at the source" and "best practices":** The ratio between the blocks above a minimum volume and the total rock extraction determines the quantity of the extractive waste. There is a need to reduce waste production within the quarry through the adoption of the best available exploitation techniques. At the same time, it is essential to focus on the collection of fine waste and sludge "at the source". As shown in this research, waste from Carrara marble exploitation may consist of pure $CaCO_3$ that is already pulverized, essentially being a semi-finished by-product for later applications as filler. The proper collection and management of waste in quarry areas, avoiding contamination with other materials, is a crucial step toward sustainable waste management and a circular economy in the extractive industry. In addition, the effective management of quarries and extractive waste (EW) requires collaboration between companies that invest in dimension stone quarries and working plants, and public bodies at different governance levels and scales (regional, national and international), involving stakeholders, citizens, research and education;
- **Characterization of quarry waste:** The characterization of extractive waste is important for understanding if the cutting method is efficient, and for understanding which treatment is most suitable or necessary to transform this material into by-products that are reusable in other processes. The systematic recovery of quarry waste could provide important benefits, reducing the economic and environmental costs related to their management and transport;
- **Research and promote reuse of rock wastes:** in addition to good practices and waste characterization, it is necessary to find "downstream" markets willing to accept these wastes as "new products." It is also necessary to inform and raise awareness of the need to accept and use products from the treatment of quarry waste (End of Waste Criteria);
- **Environmental compatibility and sustainability:** ensure the environmental compatibility and sustainability of mining and the extractive industry through the entire process: the evaluation of quarry planning, improvement of environmental performance during operations, and complete rehabilitation of the site at the end of operations;
- **Four helices approach** in managing issues connected to extractive waste management, and quarry management at large: cooperation and discussion among companies, public authorities, research centers and citizens.

Of particular interest is the exploration of multi-criteria methodologies, which assess the viability of EW facilities from a holistic perspective, encompassing economic considerations as well as the environmental and social dimensions [91].

EW recovery, encompassing recycling, reuse and reclamation, faces challenges due to data deficiencies and governance inadequacies. EU member states are required to maintain current inventories of closed EW facilities that cause or may cause potential threats to human health or the environment [92]. Further to this, despite the inventory of

EW facilities at risk on a local/national scale being generally rich in information (location, volume, characteristics, environmental impacts, company data, period of operation, etc.), at present, there is still no general and comprehensive European database with detailed information on the distribution of EW facilities or EW characteristics.

In recent years, efforts have been made to enhance the sustainable management of extractive activities, including quarries and mines. These actions focus on preventing and reducing the production of EW and promoting recovery through recycling, reuse or reclamation. Practical examples of sustainability initiatives already pursued by companies include efforts to promote the inclusion and socio-economic development of young people, enhance resilience to natural disasters, and contribute to the preservation and enrichment of cultural heritage. Additionally, they aim to ensure the safe disposal of EW in both the short and long term by analyzing the entire life cycle of the extractive industry (LCA) [93,94], employing decision support tools (DST) [91], and the application of best practices promoting the circular economy [95].

The *Best Available Techniques (BAT) Reference Document for the Management of Waste from Extractive Industries* provides up-to-date information and data on the management of extractive waste and is an important reference point for the EU to select extractive waste management activities based on site-specific characteristics and environmental risk assessments [96].

Indeed, risk management in the mining sector has greatly improved over the last 20 years with a number of different tools and techniques, and is an approach that assesses all risks associated with the construction, operation, and closure of the extractive industry facility [97]. While this approach can reduce environmental impacts, it may temporarily increase environmental effects due to reprocessing and weathering.

Therefore, careful planning and advanced technologies are vital to prevent pollutant release during new exploitation from EW facilities.

In conclusion, it is worth noting that ensuring true "sustainability" in the mining and extractive sector necessitates an interdisciplinary approach, which must encompass technical and environmental aspects, as well as social and economic considerations. In this framework, the exploitation and recovery of EW from ornamental stone quarries is in accordance with two modern and effective waste management strategies: the circular economy and sustainable mining.

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