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Asbestos and Asbestiform Minerals Transitioning from a Health Risk to an Earth Resource: A Case Study in the Serpentinites of the Pollino Ophiolite Massif (Calabria, Southern Italy)

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Abstract: Capturing atmospheric CO_2 and storing it in natural rock systems (i.e., alkaline earth silicate and hydroxide minerals) through carbonation reactions is a promising greenhouse gas mitigation research, and largely depends on the use of hydrated ultramafic rocks (serpentinites), which are widespread in orogenic belts around the globe. The Jurassic ophiolites in the Southern Apennines represent a natural analogue and a great opportunity for the mineralogical sequestration of CO_2. Serpentinites of the Pollino ophiolite Massif have been widely studied for their mineralogical, petrographic, and chemical characteristics, providing major findings on their impact on environmental and human health issues. Serpentinites here have been divided into cataclastic and massive types based on their structures and are characterized by pseudomorphic and vein textures. The mineralogy of the studied serpentinites consists of lizardite, antigorite, clino-crysotile, Cr-chlorite, magnetite, tremolite, actinolite, pyroxene and calcite, and the most commonly occurring serpentine minerals are polymorphs of serpentine including lizardite, clino-crysotile and antigorite. Systematic petrographic and mineralogical studies of serpentinites in ophiolites, such as those outcropping in the Southern Apennines can represent a starting point for the study of natural materials, which can be used for CO_2 storage and sequestration. A particular attention must be given to those carbonate phases, produced by carbonation processes to check whether the serpentinites of the Pollino Massif can be used for the induced mineral Carbon Capture Storage (CCS). If successful, this technique and the Pollino Massif serpentinites can become highly significant in safeguarding the health of our planet's climate.

Keywords: serpentinites; health risk; carbon storage and sequestration; carbonation reactions in silicate rocks; Pollino massif; southern Apennines; Calabrian ophiolites

1. Introduction

Carbon dioxide (CO_2) is a greenhouse gas and plays a major role in the regulation of Earth's temperature and affecting global warming patterns. Its concentration in the atmosphere has increased significantly over the past several decades [1–3] (Figure 1). Most carbon on Earth is bound in minerals and sequestered in deep-sea sediments, reminding us that increasing the uptake of CO_2 in minerals could reduce the greenhouse gas content in the atmosphere. Mineral carbonation is the process by which carbon dioxide (CO_2) reacts with minerals in rocks to form solid carbonates and represents the most environmentally and geologically stable method for carbon disposal [4]. This reaction can occur naturally over geological timescales or can be artificially accelerated for carbon capture and sequestration [5]. Moreover, anthropogenic gas emissions are projected to alter future climate with potentially non-trivial impacts [6], and the effects of increased CO_2 concentrations can be clearly documented through greenhouse effects, ocean surface acidification, and ecosystem fertilization [7,8].

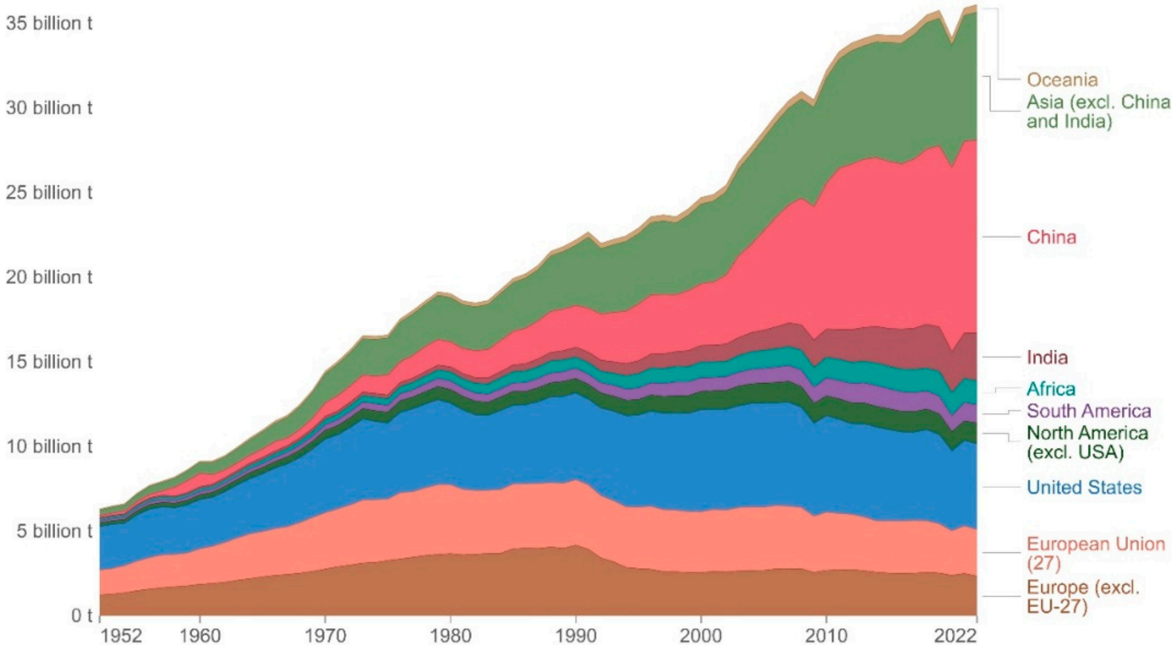

Figure 1. Annual CO_2 emissions by world region (modified after [3].

The observed increase in atmospheric carbon dioxide (CO_2) concentrations motivates the study of carbon capture and storage (CCS) as an important component of multilateral strategies to mitigate the risks of future climate change. Proposed reservoirs for CO_2 storage include terrestrial biomass, deep sea, saltwater aquifers, and minerals, and these reservoirs differ significantly in terms of the rate at which stored CO_2 leaks into the atmosphere. Characteristic storage periods increase from terrestrial pools (tens to hundreds of years), deep sea (hundreds of years), geological reservoirs (thousands of years), to thermodynamically stable minerals (> thousands of years) [9,10]. Carbonation is a process that can be performed on metal-rich, non-carbonate minerals (e.g., Mg_2SiO_4 i.e., Mg_2SiO_4 olivine, $Mg_3Si_2O_5(OH)_4$ serpentinite, $CaSiO_3$ wollastonite) to geologically and thermodynamically stable mineral carbonates (i.e., MgCO3 magnesite, $MgCa(CO_3)_2$ dolomite, $CaCO_3$ calcite, $FeCO_3$ siderite, $NaAl(CO_3)(OH)_2$ dawsonite) by induced exothermic alteration [11]. This technique attempts to mimic the natural low-temperature alteration (carbonation) of a wide range of silicate rocks that safely trap CO_2 over geological time [7,12,13]. Among various mineral species that can undergo carbonation reactions, Mg-rich minerals, as well as Ca-carbonates, are very interesting acting as important reservoirs of CO_2 since they are highly abundant at the Earth’s surface.

The mineral carbonation process is typically a reaction of olivine or serpentinite with carbon dioxide to form magnesite + quartz + water: $Mg_3Si_2O_5(OH)_4 + 3CO_2 = 3MgCO_3 + 2SiO_2 + 2H_2O$ [14]. The rate of CO_2 sequestration in nature is controlled primarily by reactive surface area, temperature, pH, and CO_2 partial pressure. There is an obvious advantage of in-situ carbonation since CO_2 is injected in-situ, with no need to mine, pulverize, or activate the serpentine rock. The ophiolites of the South Apennines (Italy) represent a natural analogue for in-situ mineralogical sequestration of CO_2. By binding the CO_2 in silicate rocks through mineral carbonation, it is possible to produce stable, also naturally occurring, carbonate rocks [15].

The typical procedure starts with a complete characterization of the mineralogy and composition of the rock, followed by laboratory and pilot-scale reactivity tests to determine the rate and extent of the carbonation reaction of a given rock [16,17]. In the case of field studies, once carbonation has started, the process is monitored to determine the extent of carbonation over time and to ensure long-term carbonate stability [7,18,19]. In the quarry site, the serpentinite rocks reacting with calcite in the presence of CO_2 could be a useful sink for carbonation processes and CO_2 storage.

Serpentinites cropping out in the Pollino mafic–ultramafic massif in the Calabria region of southern Italy have been studied extensively and analyzed for their mineralogical, petrographic, and chemical characteristics, which also provided relevant findings on their impact on environmental and human health [20–25].

Cataclastic serpentinites are more apt to be carbonated due to their texture with extensive fracturing of individual grains that increases the reactive surface area of minerals. Regarding future perspectives, it can be interesting to undertake a petrological study of the serpentinites of the Pietra Pica quarry with particular attention to the carbonate phases present and to the existence of talc, to check whether the serpentinites of the Pollino Massif can be used for the induced mineral Carbon Capture Storage.

2. Geology of the Pollino Ophiolite Massif

The Pollino Massif in the Southern Apennines (Italy) is located within the continental convergence zone between Eurasia and Africa and is composed mainly of Mesozoic and Tertiary magmatic and sedimentary rocks derived from the Ligurian ocean basin and the African passive margin. These oceanic and continental crustal rocks are overlain by Pliocene – Pleistocene terrestrial deposits [26] (Figure 2). Collectively, the Pollino Massif and the continental crustal rocks make up the Liguride Complex of the Apennines and the Calabria region [27–31].

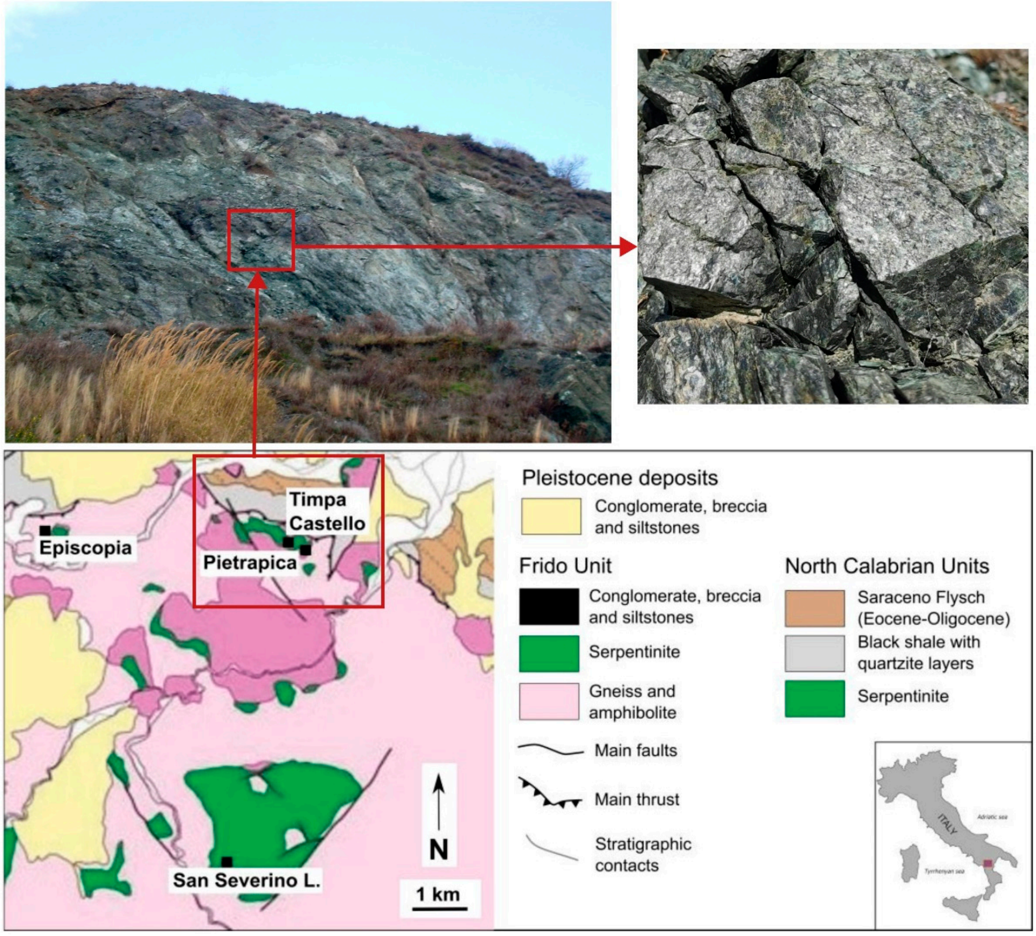

Figure 2. (a) Simplified geological map of the Liguride Complex in the Pollino Massif and location of the Pietrapica quarry site; (b) outcrop features of the Pietrapica serpentinite (modified after [32]).

The Pollino Massif (Jurassic palaeo – oceanic lithosphere) is part of the Frido tectonic unit [33], which includes a metasedimentary sequence (phyllite, meta-arenite, quartzite, and blocks of calcschist and metapelite; [34,35] and a Hess-type, incomplete ophiolite, composed of serpentinite, metabasalt, metagabbro, metapillow lavas, and dismembered metadolerite dikes [16,27,36]. The Pollino massif likely represents a fossil ocean–continent transition zone or a continental margin ophiolite [37,38], reminiscent of the modern Western Iberia rifted margin [39]. The second major tectonic entity in the Frido Unit is a non-metamorphic Calabro-Lucano Flysch or the North Calabria Unit. The Pollino Massif tectonically overlies the North Calabrian Unit and forms the structurally highest tectonic unit in the Liguride Complex.

Different interpretations have been proposed for the geological origin of the Liguride complex [40–42]. Bonardi et al. [28] have suggested that metapelitic rock assemblages, including both continental crustal and ophiolitic rocks, constitute a mélange zone (Episcopia – San Severino mélange). Other interpretations consider the Liguride complex as a stack of thrust sheets in an accretionary prism complex, characterized by variable lithologies and metamorphic overprints [30,34]. The rock units in the Frido Unit underwent HP-LT, blueschist facies metamorphism [43].

2. Materials and Methods

Serpentinite samples of the Frido Unit selected for this pilot study were collected at Pietrapica quarry, near the settlements of San Severino Lucano and Episcopia (Calabria-Lucanian boundary, Basilicata region, southern Italy), where outcrops of the Pollino massif serpentinites are widespread. Macroscopically, the analyzed serpentinites have been divided into two main types: (i) cataclastic types, showing fracturing and rigid-body rotation of mineral grains or aggregates; and (ii) massive types, consisting of fractured serpentinites embedded in cataclastic serpentinites. Recent petrographic and mineralogical studies have documented the occurrence of asbestos minerals in the area (e.g. [20–22,32,44,45].

Petrographic investigation of all the specimens was carried out by using a Nikon Alphaphot-2 YS2 optical microscope on thin sections of rock samples. Mineralogical analyses were performed on randomly oriented powder by using a Philips X'Pert 3040PW with CuKα radiation, 40 kV and 40 mA, 4 s per step, and step scan of 0.01 °2θ, in the Department of Sciences at University of Basilicata.

3. Results

The studied serpentinite rocks are characterized by pseudomorphic and vein textures [45,46]. The pseudomorphic texture (Figure 3a) represents mesh texture [47–49] in which serpentine and magnetite phases statically replace olivine crystals, and yellow brown bastite replace orthopyroxene. The vein texture is represented by different sub-millimetric veins intersecting each other and crisscrossing the pseudomorphic texture [45,46].

In the Pietrapica serpentinites, the protolith minerals are olivine, pyroxenes (either orthopyroxene and clinopyroxene) and spinel, whereas the secondary (i.e. metamorphic) minerals are serpentine, chlorite, magnetite, prehnite and amphibole. Olivine is locally recognizable in the core of serpentine + magnetite pseudomorphs in the mesh textures (Figure 3b). Orthopyroxene is observed as fresh porphyroclasts as well as bastite pseudomorphs, which retain the prismatic habit of the original orthopyroxene (Figure 3c); orthopyroxene also occurs as lamellae within clinopyroxene porphyroclasts. Clinopyroxene porphyroclasts are commonly rimmed by amphibole, which is interpreted as a tremolite-actinolite series based on optical characteristics (Figure 3d). Spinel forms red-brown coloured, xenomorphic, holly leaf-shaped porphyroclasts defined as typical of porphyroclastic upper mantle peridotites [50].

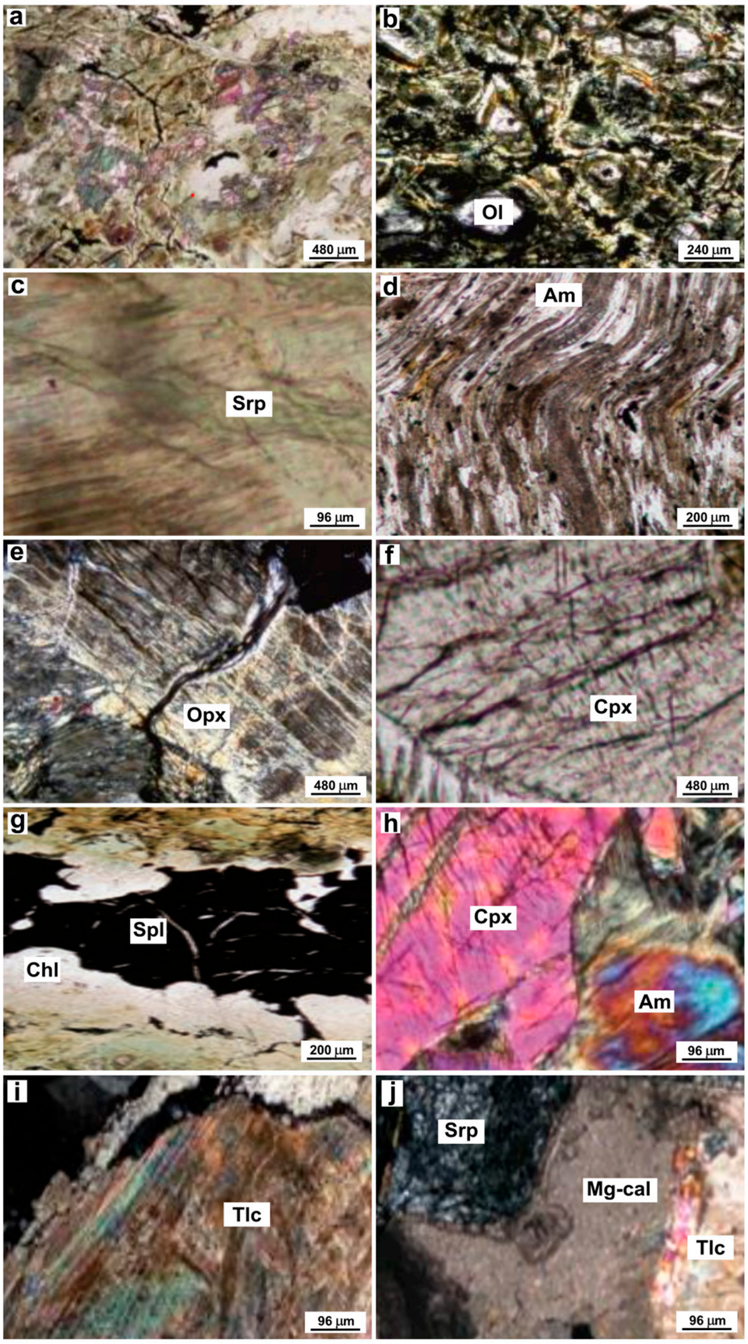

Figure 3. Representative photomicrographs of the Pietrapica serpentinite. (a) pseudomorphic textures, lower polarizer; (b) mesh texture with relicts of olivine crystals, crossed polarizers; (c) cross-cutting vein with serpentine fibres lower polarizer; (d) orthopyroxene deformed by kink fold band, lower polarizer; (e) orthopyroxene crystal, crossed polarizers; (f) relicts of clinopyroxene porphyroclasts 1N, 10X; (g) spinel porphyroclasts, lower polarizer; (h) clinopyroxene with tremolite rim (acicular amphibole) and chlorite; crossed polarizers; (i) Mg-carbonate with talc and serpentine 2X, 1X; (j) talc in carbonate vein with serpentine 2X, 1X. Legend: Ol: olivine; Srp: serpentine; Am: amphibole; Spl: spinel; Opx: orthopyroxene; Cpx: clinopyroxene; Tlc: talc. Mineral symbols are after [51].

Serpentinites are crosscut by several types of veins, filled with various serpentine minerals (Figure 3e). In some samples, kinked orthopyroxene is replaced by pseudomorphic amphiboles (Figure 3f), which are interpreted as belonging to the tremolite-actinolite series based on optical characteristics. Clinopyroxene porphyroclasts are sub-euhedral and commonly surrounded by amphibole, which belongs to the tremolite-actinolite series based on its optical characteristics (Figure

3g). Spinel forms red-brown coloured, xenomorphic, holly-leaf shaped porphyroclasts (Figure 3h). Talc is detected in some of the studied samples (Figure 3i) and is spatially associated with carbonate and serpentine veins (Figure 3j).

The results of our XRPD X-ray diffraction analysis have shown that the mineralogical phases of the serpentinite samples mainly include lizardite, antigorite, clino-crysotile, Cr-chlorite, magnetite, tremolite, actinolite, pyroxene and calcite. The most commonly occurring minerals are polymorphs of serpentine including lizardite (d = 7.27 Å), clino-crysotile (d = 7.32 Å), and antigorite (d = 7.23 Å). Amphibole minerals such as actinolite (d = 8.31 Å) and tremolite (d = 2.94 Å) were also detected in several samples (Figure 4). The presence of uvarovite in some samples is likely the result of a Ca-metasomatic process, in which the addition of Ca destabilizes the iron stored in the iron-bearing minerals of the serpentinites (such as magnetite) favouring its formation [52]. Uvarovite formation may also be related to degradation of ortho- and clinopyroxene phases in the rocks.

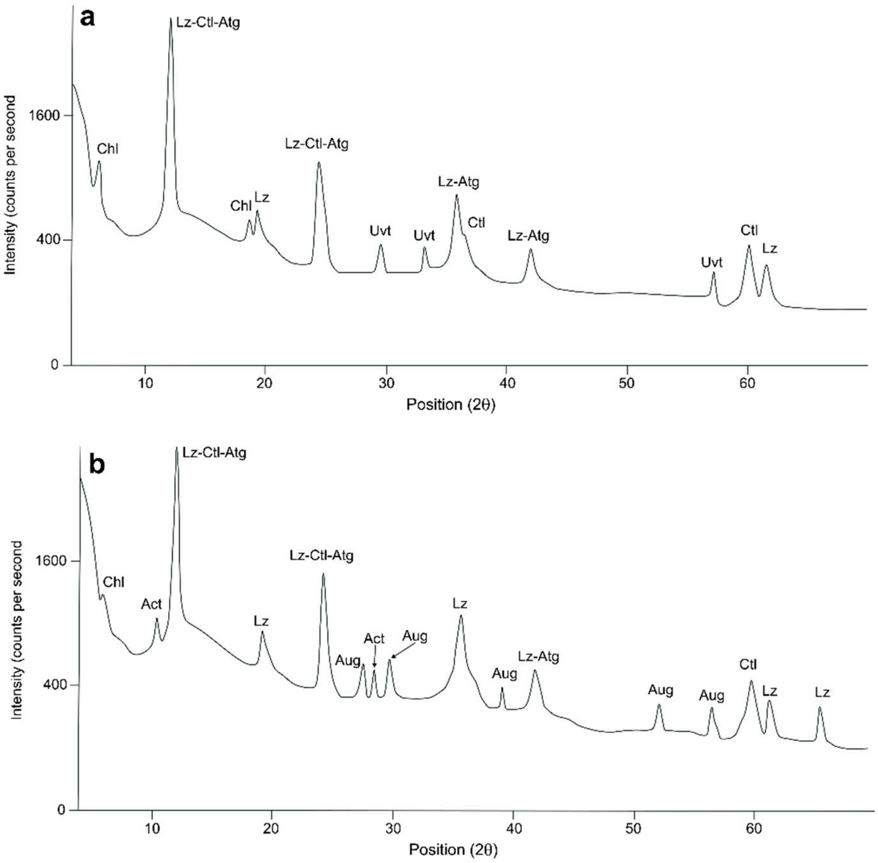

Figure 4. XRPD patterns of a massive (a) and cataclastic (b) serpentinite sample. Legend: Lz-Lizardite; Ctl-Chrysotile; Atg-Antigorite; Chl-Chlorite; Act: Actinolite; Aug-Augite; Uvt-Uvarovite.

4. Discussion

The petrographic and mineralogical study of serpentinites from the Frido Unit shows that Mg silicates, mainly serpentine and secondarily olivine, can be used for carbonation purposes. The use of serpentine rocks for CO_2 storage is an emerging technique in the field of carbon capture and storage (CCS). This technology aims to remove carbon dioxide (CO_2) produced by industries and power plants and deposit it underground, thereby reducing greenhouse gas emissions.

The objective of this preliminary work is to launch the idea for the suitability of natural analogue systems using different approaches in various sites in the Pollino area (Southern Apennines). Recent work has obtained precise estimates of the content of carbon-bearing minerals that host atmospheric CO_2. Wilson et al. [53] have shown that the amount of atmospheric CO_2 that has been bound in mineral form can be estimated from the Rietveld results for weight-percent abundance of hydrated magnesium carbonates. Wilson et al. [53] carried out a study on serpentinite-rich sterile samples from Clinton Creek and Cassiar (Australia) and it has been estimated that a total of 164,000 ± 16,400 tons of CO_2 are bound within the Clinton Creek sterile pile (applying a relative error of 10% corresponding to 4.36 wt. % of hydro magnesite). However, this high degree of accuracy is difficult to obtain with other quantitative phase analysis methods [53].

Large-scale application of mineral sequestration can represent high potential to fund remediation of mine sites and to provide reduction in greenhouse gas emissions. This refining technique could be applied to serpentine samples allowing for improved characterization of many tailings' materials and quantification of CO_2 sequestration. Recent work by [54] have shown the importance of mineralogical and petrographic studies to improve the mineral carbonation process. Initially, it was thought that the hypothesis that lizardite is more suitable for carbonation than antigorite was not correct, but it was, nevertheless, on the right track, because features that positively influence the suitability of a serpentinite rock for mineral carbonation are more common in lizardite than in antigorite. In contrast, amphiboles and pyroxenes have already been reported to be unsuitable for mineral carbonation [55,56].

Characteristics that influence the suitability of a serpentine such as mineralogical structure, parent rock, and subsequent rock transformation through metamorphosis must be considered when working with carbonation processes. These integrated studies can be useful for selecting injection sites on a field scale and potential additives or pre-treatment strategies to optimize carbonation reactions of Mg-silicate metals. Therefore, the petrographic and mineralogical study of natural occurring serpentinite rocks hosted in ophiolite sequences cropping out in the southern Apennine can serve as a starting point for the study and analysis of natural materials that can be used for CO_2 storage. It is important to continue research in this area at a detailed extent to find more efficient and sustainable ways to reduce carbon emissions and mitigate climate change.

5. Conclusions

The prospects of using serpentine rocks for CO_2 storage are promising. The market for CO_2 storage and sequestration that is developing in parallel with the European regulated market is an economically promising sector. The growth of this market is based on the prospects for the development of capture and storage technologies over the next decade. CO_2 storage in rocks, particularly in serpentine rocks, is important for several reasons:

- First, this technique allows some of the carbon dioxide in the atmosphere to be isolated and injected into deep reservoirs reducing the amount of CO_2 in the atmosphere, and hence helping to mitigate the greenhouse effect and global warming. CO_2 storage can facilitate the energy transition process.
- As we move toward cleaner energy sources, CO_2 storage can help manage residual CO_2 emissions.
- Deep reservoirs that were previously occupied by gas and/or oil can be used for CO_2 storage since these reservoirs are able to naturally contain gas and fluids, making them suitable for CO_2 storage.

Some projects are exploring the possibility of turning CO_2 into rock, a process that could offer a long-term and safe method of storage. However, it is important to note that these techniques are still under development and require further research to be implemented on a large scale. Each contribution aimed at providing detailed geological and petrographic features of natural outcrops may add a new piece of information towards sustainable solutions for greenhouse gas reduction.

Author Contributions: Conceptualization, Giovanna Rizzo, Yildirim Dilek and Rosalda Punturo; Data curation, Roberto Buccione; Formal analysis, Roberto Buccione and Marilena Dichicco; Methodology, Marilena Dichicco; Supervision, Giovanna Rizzo, Yildirim Dilek and Rosalda Punturo; Writing – original draft, Roberto Buccione; Writing – review & editing, Giovanna Rizzo.

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