The Availability of Limestone and Other Raw Materials for Ocean Alkalinity Enhancement

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Abstract The work assesses the availability and localizations of different raw materials suitable for ocean alkalinity enhancement (OAE), like limestone, olivine, magnesite and brucite, since several billion tons of rocky materials are needed to achieve meaningful results for carbon sequestration through OAE. Resources of carbonates are immense and widespread around all continents. Availability of pure carbonates is still very large (outcrop area 4.1 million km^2) and is not a constraint for the large-scale development of OAE. Outcrops of pure carbonates within 10 km from the coastline and below bare ground or scrub/shrub, preferred for the logistics of exploitation, account for about 70,000 km^2, and could provide about 5,000 Gt of limestone. These values increase by a factor of 3 and 8 within 50 and 100 km from the coastline, respectively. Potential resources of olivine, less easily identifiable from the geological data, are estimated in the order of a few hundred billion tons and could provide only a minor contribution to ocean-based carbon removal strategies. A comparison with the current level of world extraction of mineral raw materials is also provided. The annual production of limestone, estimated to be more than 6.6 Gt from deposits scattered all around the world, is about 9% of the world production of mineral raw materials (around 44 Gt yr^{-1}), and is of the same order of magnitude as coal (7.3 Gt yr^{-1}). The annual productions of magnesite (29 Mt yr^{-1}), olivine (8.4 Mt yr^{-1}) and brucite (1.5 Mt yr^{-1}) are two orders of magnitude lower.

Plain Language Summary The ongoing acidification of the oceans due to the increase of carbon dioxide (CO_2) concentration in the atmosphere and the consequent increased uptake by the sea can be tackled by adding alkaline compounds under controlled conditions. Since this promotes a number of reactions in the carbonate system which ultimately results in a drawdown of atmospheric CO_2, the enhancement of ocean alkalinity can also contribute to the global effort of limiting global warming, and it is widely studied in the literature among the CO_2 removal technologies. A limiting factor can be the worldwide availability of alkaline materials to be used for such a scope. The paper shows that the global availability of pure carbonates is vast, although unevenly distributed, and it could largely satisfy the requirement for large-scale development of ocean alkalinization. The required up-scaling of limestone production compared to the current level is much lower than what would be needed for other raw materials that could be used, such as olivine, magnesite and brucite. Moreover, a large fraction of pure limestone resources that could be exploitable is located nearby the coastline, in areas without or with low vegetation cover, which favors the logistics related to the exploitation.

1. Introduction

The increasing greenhouse gas (GHG) emissions into the atmosphere are heavily affecting the global climate and at the same time are acidifying the oceans (IPCC, 2019). In particular, the concentration of carbon dioxide (CO_2), the most important greenhouse gas (GHG) by radiative forcing, is significantly and continuously rising due to human activities such as the use of fossil fuels and ongoing deforestation (Friedlingstein et al., 2020). Further greenhouse gas (GHG) emissions and consequent warming are expected in the coming decades (IPCC, 2021). Rapid and far-reaching transitions in energy, land, urban infrastructure and industrial systems are then urgently needed to decrease the emissions in order to limit global warming (IPCC, 2018). Furthermore, in order to keep the increase of global temperature well below 2°C, as agreed with Article 2 of the Paris agreement, different approaches to reduce the carbon dioxide (CO_2) concentration in the atmosphere through so-called "negative emission technologies" (NETs) have been proposed, such as bioenergy with carbon capture and storage, afforestation and reforestation, land management to increase carbon in soils, enhanced weathering, direct carbon storage, ocean fertilization, and ocean alkalinization (EASAC, 2018; Minx et al., 2018). A portfolio of negative emission technologies (NETs) is needed to provide many Gt yr^{-1} of carbon removal, and as a consequence, the comparison of the different pros and cons, co-benefits and side effects of the different options is a developing

field of research. In particular, ocean alkalinity enhancement (OAE) and enhanced weathering have the potential additional benefit of counteracting ocean acidification, another consequence of the massive anthropogenic fossil fuel exploitation (Butenschön et al., 2021; Renforth & Henderson, 2017). Aim of this paper is then to estimate the potential worldwide availability of different minerals required for OAE.

In order to achieve meaningful results in removing atmospheric CO_2 and counteracting ocean acidification, large quantities of raw materials (limestone or other minerals and rocks) are needed, in the order of billions of tons per year. This poses a question on the actual availability of these materials and on the sustainability of the required upscaling in current extraction activities (NASEM, 2021). For this reason, this paper provides information about the global availability of deposits of calcium carbonate and its current extraction, as well as the current extraction of olivine, magnesite and brucite. In addition, for calcium carbonate and olivine, it also evaluates two factors for their potential exploitability, such as the distance of the potential deposits from the coastline and the soil use category in which they fall, considering that desert or sparsely vegetated soils in arid climates are easier to explore for deposits and there are less environmental concerns. The current level of worldwide extraction of mineral raw materials is also provided, to allow a comparison of the magnitude of current mining of minerals of interest for OAE and the exploitation of world mineral resources in the current economic production system.

### 1.1. Ocean Acidification, Weathering Processes and the Carbon Cycle

Carbon dioxide, being a soluble gas, equilibrates with seawater in the global oceans, increasing the concentration of $CO_2(aq)$ and bicarbonate ions (HCO_3^-), reducing at the same time the concentration of carbonate ions (CO_3^{2-}) in the surface waters. This causes the acidification of the oceans, and the perturbation of the $CaCO_3$ cycle, as well as impacts on marine organisms (Gattuso et al., 2015; Kroeker et al., 2013). Natural physical and biogeochemical processes act to counteract the variations of pH, at first on decadal to century timescales, through the weathering of carbonate rocks and the transport of CO_2-enriched surface waters down into the oceans. Then, ventilation of the deep ocean with CO_2-enriched and CO_3^{2-}-depleted water derived from the surface will reduce the stability of deposited calcium carbonate $(CaCO_3)$ in seafloor and will lead to increased rates of in situ carbonate mineral dissolution (Archer & Brovkin, 2008; Archer et al., 1998; Sarmiento et al., 1992). This restores the buffering capacity of the deep waters and the subsequent upwelling of these waters will then allow further CO_2 uptake from the atmosphere. In longer timescales, the residual atmospheric CO_2 perturbation is removed as a result of the response of terrestrial weathering of carbonate and silicate rocks, caused by a warmer and wetter climate resulting from elevated atmospheric CO_2 concentrations, drawing down more atmospheric CO_2. As a consequence, temperature and weathering return toward their unperturbed levels (Walker et al., 1981). A silicate weathering feedback acts to restore pCO_2 to some equilibrium value on timescales of around 100–1,000 kyr, setting the ultimate maximum duration of an anthropogenic carbon cycle perturbation (Archer & Brovkin, 2008; Lord et al., 2016).

1.2. Enhancement of Ocean Alkalinity

Given the importance of $CaCO_3$ dissolution in the carbon cycle, the artificial increase of ocean alkalinity via the dissolution of limestone-derived products (also known as ocean liming) has been proposed as a carbon dioxide removal technology (Kheshgi, 1995) and has gained attention due to its capability of addressing at the same time the issues of global warming (via increased uptake of atmospheric CO_2) and of ocean acidification (Renforth & Henderson, 2017). Harvey (2008) considered the direct application of limestone to the surface layer in regions where the depth of the boundary between supersaturated and unsaturated water is relatively shallow and where the upwelling velocity is large. Limestone dissolves and precipitates at a slow rate (about 1×10^{13} mol Ca/yr), but readily soluble calcium can be extracted from limestone in the form of lime (CaO) to produce calcium hydroxide (slaked lime). This requires a considerable amount of energy and causes CO_2 emissions that should be controlled (Caserini et al., 2019; Renforth et al., 2013).

$$CaCO_3 \rightarrow CaO + CO_2$$

$$CaO + H_2O \rightarrow Ca(OH)_2$$
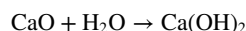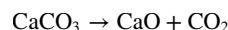
The principle behind ocean liming is based on the dissociation of $Ca(OH)_2$ and CO_2 to calcium ions and bicarbonate:

$$Ca(OH)_2 + 2CO_2 \leftrightarrow Ca^{2+} + 2HCO_3^-$$
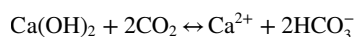
Hence, this reaction is theoretically capable of removing two moles of CO_2 per mole of added slaked lime and of increasing the alkalinity of the water body by two-mole equivalents. However, the actual efficiency of its implementation in removing atmospheric CO_2 is reduced depending on the environmental and dynamic conditions (Butenschön et al., 2021; Zeebe & Wolf-Gladrow, 2001). By assuming a conservative value of 1.4 mol of CO_2 absorbed per each mole of $Ca(OH)_2$ added (Keller et al., 2014), this implies that 1.62 tons of $CaCO_3$ are needed for each ton of CO_2 removed.

Weathering of silicates on land can remove CO_2 from the atmosphere; if these cations are transported to the ocean, they will increase the alkalinity and lead to an increase of (HCO_3^-) and CO_3^{2-} at the expenses of CO_2. The products of silicate weathering, therefore, are more effective in CO_2 removal if they reach the ocean than if they reprecipitate as secondary minerals on land. This is the reason why some authors proposed the dispersion in the ocean of silicate minerals such as olivine (magnesium-iron silicate mineral) to counteract ocean acidification with large-scale OAE (Albright & Cooley, 2019; Kohler et al., 2010). In the case of forsterite (Mg endmember), the occurring reaction is:

$$Mg_2SiO_4 + 4CO_2 + 4H_2O \rightarrow 2Mg^{2+} + 4HCO_3^- + H_4SiO_4$$
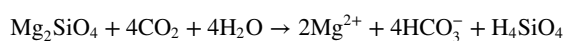
Hangx and Spiers (2009) and Kohler et al. (2010) emphasize that a large specific surface is needed for the process to be effective, which means that olivine grains must be in the order of 1–10 μm to achieve significant steady-state CO_2 uptake rates within a few decades. The quantity of CO_2 sequestered for every mole of olivine dissolved varies depending on the initial state of the seawater carbonate chemistry. The upper theoretical yield of the process is 1.25 g of CO_2 (0.34 g of C) mitigated per 1 g of olivine reacted, but in reality, the ratio of CO_2: olivine is closer to 1 g g^{-1} (Kohler et al., 2010). In case of olivine composition with magnesium and iron endmembers, CO_2 uptake is reduced. On the contrary, with pure fayalite (Fe_2SiO_4) what occurs is not useful for CO_2 removal, since it involves the release of Fe hydroxide, followed by a decrease in the supersaturation for aragonite and calcite compared to seawater, then stopping the CO_2 uptake. This is described by the following reaction:

$$Fe_2SiO_4 + {}^1\!/_2O_2 + 5H_2O \rightarrow 2Fe(OH)_3 + H_4SiO_4$$
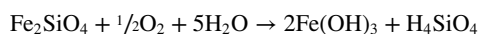
Forsterite-rich olivine has also been investigated as a feedstock for a coupled mineral carbonation ocean alkalinization process (Renforth & Kruger, 2013). According to this process, olivine could be fed into a (single-stage) mineral carbonation reactor at elevated pCO_2, producing magnesite ($MgCO_3$) and silica (mineral carbonation process). After dewatering, the precipitated magnesite could be fed into a slaking system to produce brucite ($Mg(OH)_2$), which could then be used for ocean liming, according to the reaction:

$$MgCO_3 + CO_2 + H_2O \rightarrow Mg^{2+} + 2HCO_3^-$$
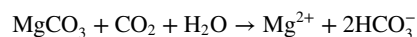
It's worth noting that $MgCO_3$ decarbonates at a lower temperature than $CaCO_3$, thus lowering the energy consumption for calcination.

Another option for OAE considers the direct extraction and dispersion in the ocean of brucite, following the reaction:

$$Mg(OH)_2 + 2CO_2 + H_2O \rightarrow Mg^{2+} + 2HCO_3^-$$
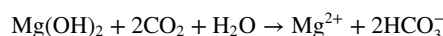
1.3. Carbonate Rocks

The chemical elements found in calcium carbonate are widely distributed. Calcium (Ca) is the fifth most common element in the Earth's crust (after oxygen, silicon, aluminum and iron) and had been extracted from early igneous rocks by the combined effects of erosion by the weather and corrosion by acidic gases (Oates, 1998). The combination of dissolved calcium ions and CO_2 resulted in the sedimentary deposition of calcium carbonate, which was subsequently converted into limestone rock. Calcium carbonate is deposited either by chemical precipitation, by biochemical processes and by organogenic sedimentation. Limestone is a widely distributed rock, comprising

about 15% of the sedimentary rocks on the surface of Earth and almost 10% of all the surface rocks of the planet (Parker, 1967).

Limestone and dolomite are the more common carbonate rocks used in the industrial sector (Kogel et al., 2006). Limestones are sedimentary rocks composed mostly of mineral calcite ($CaCO_3$), whereas dolomites are sedimentary rocks composed mostly of mineral dolomite ($CaMg(CO_3)_2$). Aragonite ($CaCO_3$), which is a pseudomorph of calcite, is economically important only in modern deposits such as those composed of oyster shells and oolites, thanks to the ease of extraction at the sandy beaches in tropical countries. Other commonly found carbonate minerals, associated with limestones and dolomites, comprise siderite ($FeCO_3$), ankerite ($Ca(Mg,Fe)(CO_3)_2$), and magnesite ($MgCO_3$). Dolomites are the result of a dolomitization process occurring during the diagenesis of limestone, where magnesium ions replace the calcium ions in the crystal lattice. The exchange takes place either directly during sedimentation, when the seawater is richer in magnesium than calcium, or considerably later during secondary dolomitization, when magnesium-rich water circulates through the finished rock (Oates, 2002).

Limestones are classified in several ways, based on geology, grain size, microstructure, texture and the principal impurities, but the classification most commonly used in the limestone industry is a practical chemical classification based on the carbonate content (Oates, 2002): limestone is divided in ultrahigh calcium limestone (>97.5% $CaCO_3$) and high-calcium limestone (95%–97.5% $CaCO_3$); high-purity carbonate rock is more than 95% combined $CaCO_3$ and $MgCO_3$, whereas high-magnesium dolomite is more than 43% $MgCO_3$ (theoretically pure dolomite is 45.7% $MgCO_3$, Oates, 2002). This classification is the most useful for the aim of this paper, since the chemical properties are the most relevant if the stone is calcinated, like in the production of lime.

Lime, chemically calcium oxide, is produced through the calcination of limestone or dolomite. High purity limestone is preferred for calcination, because during this process it loses 44% of its weight, thus the content of impurities will approximately double. The hydration of lime ("slaking") leads to calcium hydroxide (slaked lime).

Limestones and dolomites are characterized by a wide range of color, grain size and bed thickness, with common impurities being clay, sand, chert and organic matter. Different beds in a deposit can be mined for different purposes, for example, a high-calcium limestone for lime production and a dolomite section for crushed stone (Kogel et al., 2006). Overburden (a superficial bed of altered rock that can reach thicknesses of several meters) is generally present and must be stripped off; its nature and thickness can greatly affect the economics of the operation.

1.4. Olivine, Magnesite and Brucite

Mg-rich and Fe-rich olivine are frequently occurring in nature, in a range of mixtures between the end-members forsterite (Mg_2SiO_4) and fayalite (Fe_2SiO_4) (Deer et al., 2013), with pure forsterite that rarely exists in nature. The most common olivine of this solid solution series has a magnesium-iron ratio of 1.6:0.4, with the formula $Mg_{1.6}Fe^{2+}_{0.4}(SiO_4)$ (Kremer et al., 2019). The term olivine derives from the olive-green color of this intermediate composition. Olivine has two main occurrences: it can be found in dunite, which is an ultramafic rock that contains a minimum of 90% olivine with additional minerals, like ortho- and clinopyroxenes, as well as in serpentine group minerals as accessories such as magnesite, chromite, ilmenite, rutile, and magnetite. Additionally, in small volumes, plagioclase, garnet, and spinel can be found in dunites; olivine can also be found in serpentinite, which are metamorphic rocks composed mainly of the mineral serpentine $(Mg,Fe)3(Si_2O_5)(OH)_4$ along with other accessory minerals, which derives from alteration of olivine minerals in kimberlites, ophiolites, and gabbros that originally had large amounts of olivine (Harben & Smith, 2006).

There are many small deposits of olivine bearing rocks, but only a limited number of them are actually exploited because of the high fixed costs. Olivine bearing rocks are found within geological settings such as ultramafic intrusions, ophiolite complexes, rift zones/basalts of mid-ocean ridges and volcanic xenoliths (Kremer et al., 2019). The deposits large enough to be exploited are usually found in settings of ultramafic intrusions and ophiolite complexes. Ultramafic rocks such as dunite and peridotite contain much Mg and correspondingly contain the highest content of Mg-rich olivine (Deer et al., 2013; Krauskopf, 1982). The percentage of Mg in dunite-olivine varies from 88% to 93% in regular dunites to 80%–88% in Fe-rich dunites (Ackerman et al., 2009; Su et al., 2016).

Brucite, $Mg(OH)_2$, is an uncommon mineral found in metamorphic and ultramafic rocks. Having the highest magnesium content than any other raw material, it is a potential ore source for magnesium metal, which nowadays is in great demand (Simandl et al., 2007; USGS, 2019). Brucite can be found in different geological environments as in ultramafic rocks (Hora, 1998; Khan et al., 1971), in kimberlites (Malkov, 1974) and carbonatites (Lee et al., 2000). Most of the economic brucite deposits are hosted by marbles affected by high-temperature, low-pressure metamorphism: the highest grade, nearly monomineralic brucite deposits, are found in contact metamorphic aureoles, where the protolith consists of magnesite with little or no calcite, clays, chert or other mineral impurities (Simandl et al., 2007). Although ultramafic-hosted deposits were considered as potential sources of brucite in the past (Khan et al., 1971), the association of brucite with asbestos in these geological settings makes carbonate-hosted brucite deposits the recommended and preferred exploration targets.

Magnesite ($MgCO_3$), unlike brucite, is a relatively frequent mineral as a minor to trace component in rocks of most geological environments. Large monomineralic magnesite rock bodies are rather rare, in contrast to the frequency of the mineral magnesite, and can be found in different types of deposits associated with ultramafic rocks or stratabound in sedimentary environments. However, there are only two types of deposits that are of economic importance: the Veitsch type and the Kraubath type (Pohl, 1989, 1990; Schroll, 2002). The former represents the majority of the world's reserves of magnesite and is composed of large stratabound lenses within marine platform sediments with limestone and dolomites as country rocks. The latter comprises nearly monomineralic veins and stockworks of magnesite in ultramafic rocks being part of ophiolite suites or stratified basic intrusions; in this case, the country rocks are peridotite/dunite and serpentinite. Kraubath type magnesite is preferred for industrial use as it is purer and has less Ca and Fe content, although the difficulty of separating magnesite veins from the enclosing silicate host makes vein deposits more expensive to run (Abu-Jaber & Kimberley, 1992). Magnesite main use in industry is to provide magnesia (MgO) upon calcination.

2. Materials and Methods

2.1. Potential Availability

Data about carbonate rocks occurrence around the world have been extracted from the global lithological map database GLiM (Hartmann & Moosdorf, 2012) and elaborated through ArcGISPro2.8 software. GLiM is the most recent global lithological map and has a high resolution (map scale is on average 1:3,750,000, area-weighted), obtained by assembling 92 regional geological and lithological maps translated into lithological information with the help of regional literature, and is based on three pre-existing digital global lithological maps developed by Bluth & Kump (1991), Amiotte Suchet et al. (2003), and Durr et al. (2005).

The lithological classification is made up of three levels. The first one contains 16 lithological classes, comparable to previously applied definitions in global lithological maps. The additional two levels, which describe more specific rock attributes, contain 12 and 14 subclasses, respectively. In the GLiM, a full lithological classification consists therefore of three two-digit codes ("xxyyzz"). The first level class (xx) represents the dominant lithology, while the second (yy) and third (zz) optional level subclasses closer define the rocks and highlight local special attributes.

For the aim of this paper, two main categories of lithological units have been considered. Units denominated as xx = sc (carbonate sedimentary rocks) and yy = pu (pure carbonate, without subordinate siliciclastic sediments) are the most important for OAE. Units denominated xx = sc with yy different from pu, identifying carbonate-dominated units with siliciclastic sediments, comprise the totality of other carbonate units. Examples of sedimentary carbonate units are limestone, dolomite and marl.

It's worth noting that the presence of carbonaceous units does not necessarily mean that they can be exploited. In many areas, competition is intense for potential mineral lands for construction sites, recreation areas, nature preserves, and transport infrastructures. For this reason, the data set of worldwide natural protected areas has been considered (UNEP-WCMC, 2022).

Regarding olivine and brucite, the lithological map from Hartmann and Moosdorf (2012) provides the occurrences of major ultramafic rocks bodies and ophiolites, in which the main deposits of these commodities occur. The lithological units considered for reporting ophiolites and ultramafic rocks are denominated as xx = bp (basic plutonics) and as xx = mt (metamorphic rocks) with yy = am (mafic metamorphics mentioned) or yy = gr

(greenstone mentioned). These kinds of rock units represented on the lithological map are also the site of some large deposits for magnesite, although most of the deposits currently exploited are composed of large stratabound lenses within marine platform sediments with limestone and dolomites as country rocks, as stated previously.

However, due to the large scale of the map, some rock bodies that could be considered as suitable target for exploration are not reported, since they are too small (inaccuracies of up to 10 km can occur in the map); for this reason, the lithological map represents only a partial assessment of the potential sources of olivine, magnesite and brucite; data on the occurrence of ophiolites (important for olivine) has been integrated with data from different works published in literature (Harben & Smith, 2006; Kremer et al., 2019; Oze et al., 2007).

The availability of the raw materials considered in the paper is referred where possible in terms of resources and reserves. Resources are in situ concentrations of minerals or bodies of rocks that can become of potential economic interest due to the estimated tonnage and grade of mineralization. Reserves, on the other hand, are a sub-set of mineral resources that are derived from those by the application of pre-feasibility and feasibility studies to address all of the modifying factors in order to demonstrate that at the time of reporting, extraction could be economically justified (CRIRSCO, 2008).

Since the deposits close to the coastline are preferred for the logistics of exploitation for ocean liming, due to lower energy and cost penalty for transportation, a special focus has been dedicated to outcrops between 10, 50 and 100 km from the coastline, the latter taken from GISCO Eurostat 1:1 million scale (Eurostat, 2020). Although the overlapping between the lithological map and coastline is not perfect in some locations, the influence on the calculation is negligible.

The exploitation of rock materials induced serious environmental issues (Ganapathi & Phukan, 2020), such as land degradation, land occupation, dust pollution. Information on land use is of great interest for large-scale limestone mining, since environmental impacts and interferences with ecological systems are substantially lower if mining activities consider bare grounds, such as exposed rock soils or deserts, or soils with sparse cover of bushes, shrubs and tufts of grass. Detailed information on land use/land cover (10 classes at 10-m resolution) has been taken from ESRI, 2020 Land Cover (ESRI, 2020). Small outcrops with areas less than 1 km^2 has been neglected.

2.2. Worldwide Extraction Data

The assessment of worldwide production and use of limestone is difficult because no or scarce data are available for some major producing countries (i.e., China, Turkey and most of European countries) and different databases are used in other countries. Considering that limestone is an essential raw material for cement production, an assessment of limestone extraction could be derived from cement, lime and limestone production for some countries (Chilamkurthy et al., 2016; Peduzzi et al., 2013; Svendrup et al., 2017). Other data come from the statistic of "crushed stone", where limestone is aggregated in the production of sand and gravel as for USA data and European Union data (British Geological Survey, 2021; USGS, 2019).

With regards to magnesian minerals such as forsterite and magnesite, their production is well reported in the literature, since they are utilized in various industrial processes. Brucite production, on the other hand, has only recently begun to grow, since new industrial uses have been discovered (Lee, 2015; Simandl et al., 2007). However, most of the brucite used today has an artificial origin (Lee, 2015). Data about their deposit location and reserves were taken from different sources, such as Lee (2015), Wietlisbach (2018), Simandl et al. (2007), Kremer et al. (2019) and Wilson (2017).

Although the mineral raw materials of interest for OAE are limestone, magnesite, olivine, brucite, the world production of other industrial materials has been considered for comparison. Production data about iron and ferro-alloy metals, non-ferrous metals and minerals fuels can be found easily in literature, whereas for some industrial minerals only indirect estimates could be provided. Although sand and gravel are mined worldwide and are the largest volume of solid material extracted globally (Peduzzi et al., 2013), reliable data on their extraction are available only for some countries (Krausmann et al., 2009). This is because they are very common around the world, so their value is low, and furthermore because they are extracted for local use as close as possible to the place of use, mostly in small quarries. Sand and gravel extraction can also be estimated indirectly through the
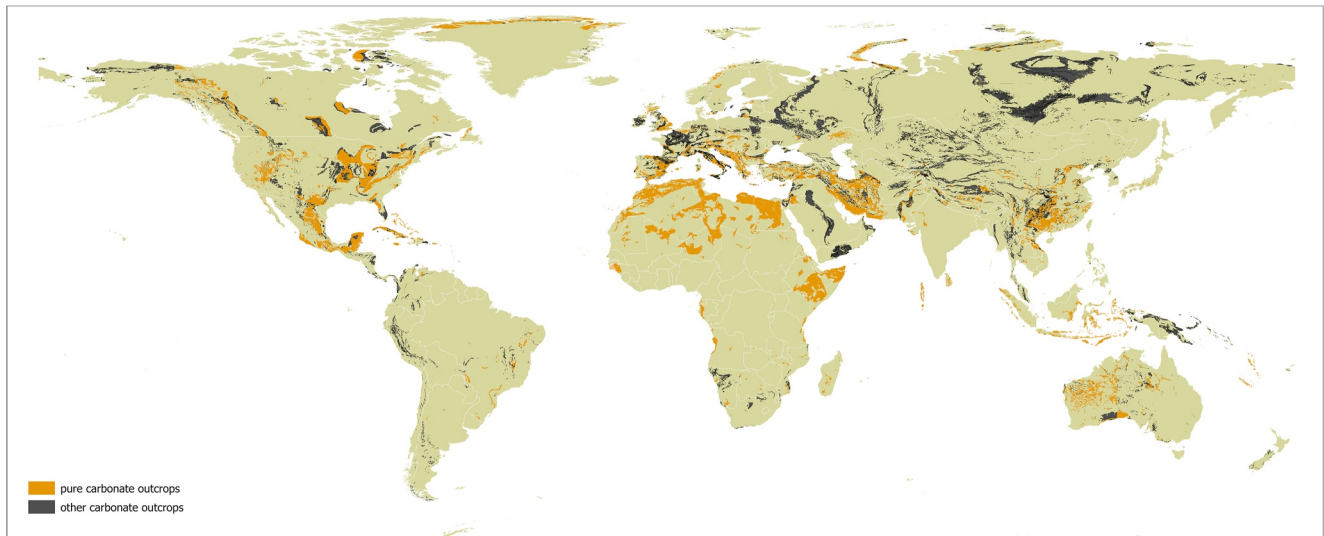
Figure 1. Pure carbonate and other carbonate rocks outcrops in the world (data from Hartmann & Moosdorf, 2012).

production of cement for concrete, assuming that four tons of sand and gravel are used for every ton of cement (Chilamkurthy et al., 2016).

3. Results and Discussion

3.1. Availability of Raw Materials for Ocean Liming

3.1.1. Carbonate Rocks

Figure 1 shows the map of the global distribution of pure carbonate and other carbonate units, whereas Figure 2 shows the percentage distribution of pure carbonate outcrops within different distances from the coastline, in different continents (country-level details are given in Table SM1 in Supporting Information S1). Total world pure carbonate outcrops area is about 4,1 million km^2. Large deposits within 50 km from the coastline are available in the Mediterranean area, in central and north Africa, in the Middle East, in South Asia and in Central America (Figure S1).

Figure 2. Pure carbonate outcrops at different distances from the coast.

Table 1
Pure Carbonate Units Outcrop Areas (10^3 km²) at the Global Level (Below 75° of Latitude) in Areas Within 10, 50 and 100 km From the Coastline, and Indicative Assessment of the Amount Extractable
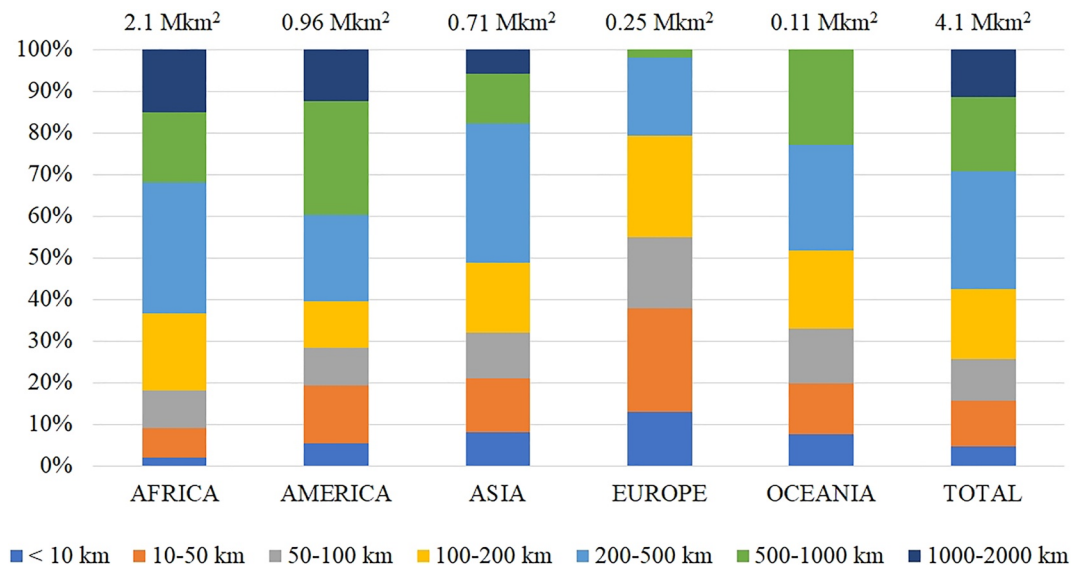
Rock	Soil use	Outcrop area (10^3 km²)			Weight—25 m depth (Gt)		
		<10 km	<50 km	<100 km	<10 km	<50 km	<100 km
Pure carbonate	Total	169	599	1,049	11,409	40,408	70,798
	of which below scrub/shrub[a]	*60*	*181*	*507*	*4,039*	*12,221*	*34,240*
	of which below bare ground[b]	*9.2*	*38*	*96*	*620*	*2,583*	*6,485*

[a]Scrub/shrub: moderate to sparse cover of bushes, shrubs and tufts of grass, savannas with very sparse grasses, trees or other plants. [b]Bare ground: exposed rock or soil, desert and sand dunes, dry salt flats/pans, dried lake beds, mines.

Considering that limestone formations are composed of beds or strata whose thickness can vary from a few centimeters to several meters, usually the total thickness of these formations is tens or hundreds of meters. Stone mining operations continue to develop at deeper depths, and excavation depth frequently reaches several dozens of meters (Newman et al., 2020). Since carbonate rock density is about 2.7 t/m³ (Manger, 1963), the potential resources of pure carbonate rocks are in the order of several tens of thousands of gigatons. Considering outcrops below 75° of latitude and within 10, 25 and 100 km from the coastline and assuming a conservative average excavation depth of around 25 m, the potential resources of pure carbonate are about 11,400, 40,400 and 70,800 Gt, respectively (Table 1).

Table 1 also shows that the amount of potential carbonate that could be mined in soils classified as scrub/shrub or bare ground within 10, 50 and 100 km from the coast, at latitude below 75°, are 69,200, 219,000 and 605,000 km², respectively, and could provide several thousand Gt of limestone. Details for the other soil use are given in Table SM2 in Supporting Information S1, whereas country data for soil use within 50 km from the coastline are reported in Table SM3 in Supporting Information S1. Iran and Somalia are the countries with the highest amount of pure carbonate close to the coastline, found almost entirely below scrub/shrub or bare ground (Table SM3 in Supporting Information S1).

The distance from the coast is just one of the main indicators of the potential exploitability of a limestone resource. Other factors, such as the proximity to transport infrastructure (ports, railways, roads), the availability and cost of renewable energy, or the presence of protected natural areas, are of great importance, and should be evaluated in a detailed analysis at the country level. Figure 3 shows that the existing sea ports are localized almost
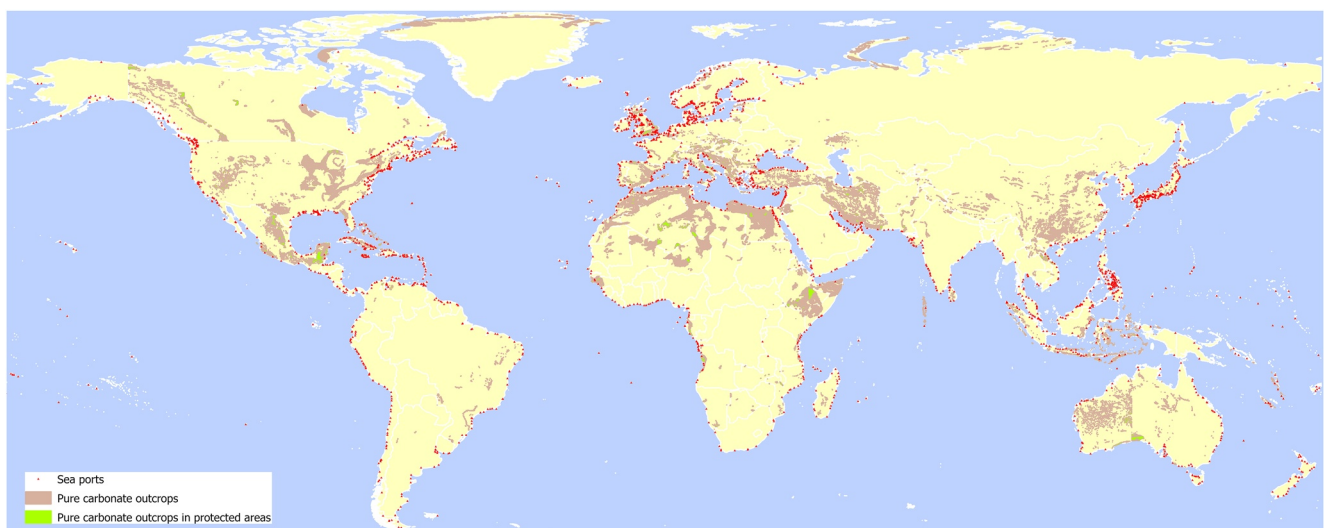

Figure 3. Pure carbonate outcrops, pure carbonate outcrops in protected areas and world sea ports (data from: Hartmann & Moosdorf, 2012; Ocha-HDX, 2022; UNEP-WCMC, 2022).
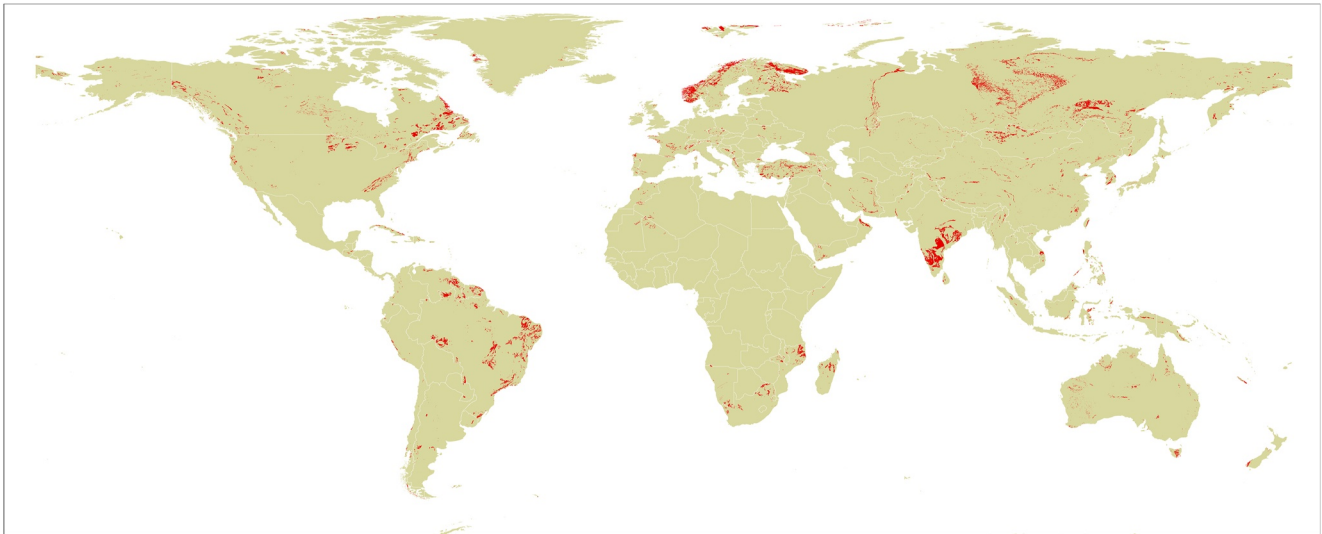
Figure 4. Ultramafic rocks and ophiolites (in red) in the world (data from Hartmann & Moosdorf, 2012).

along every coastline. Natural protected areas overlap with about 6% of the global limestone outcrops (0.324 M km^2).

3.1.2. Olivine, Magnesite and Brucite

Ultramafic and ophiolites rocks, which host the main olivine, brucite and magnesite deposits, are scattered all around the world (Oze et al., 2007). Large deposits have been found in northern Europe, along the east coast of the Adriatic Sea, in America along the Pacific Ocean coast, and in Asia, with the large ophiolites of Oman (the Samail ophiolite, 350 km long and 40 km wide) and of Papua New Guinea (200 × 50 km) (Kelemen & Matter, 2008).

Outcrop area of ultramafic and ophiolite rocks units reported in the global lithological map amount to 2,457·10^3 Mkm^2, with 125·10^3 and 193·10^3 Mkm^2, respectively within 10 and 50 km from the coastline. It is worth remembering that, as explained in Section 1.4, ultramafic rocks does not guarantee the presence of olivine-rich rocks, since many other rock types could be found in these environments; on the other side, some minor rock bodies are not visible due to the large scale of the map. The map of global outcrops of ultramafic rocks and ophiolites is reported in Figure 4.

With regards to olivine, reserves are estimated in a few tens of billion tons, while potential resources in a few hundreds of billion tons. The biggest reserves have been identified in west Norway, within the Fjordane Complex, about 2 billion tons (Harben & Smith, 2006). In Italy, olivine reserves are evaluated in 650 Mt, with mines located mainly in the Piedmont region. Other large deposits are the Horoman Hill in Japan, with reserves estimated at 100 Mt, and deposits in Xixia and Yubian Counties in China, with mineral reserves of 9 Mt. For the United States, large deposits are being identified in North Carolina, with the largest one containing an estimated 300 Mt of dunite, and in the Washington State, where potential huge resources of olivine were found at Twin Sister Mountain, with a dunite body of 200 Gt (Harben & Smith, 2006; Kremer et al., 2019; Moen, 1969).

For magnesite, the proven global reserves are estimated to be about 3.4 Gt, mainly in China, Russia and Turkey (Wietlisbach, 2018).

World brucite reserves and resources are impossible to estimate with any reasonable accuracy because there are many inconsistencies in the numbers reported in the literature (Simandl et al., 2007). A resources assessment of 71 million tons, concentrated in a few countries, has been provided by Wilson (2017). Deposits of economic significance of brucite can be found in Russia, China, North Korea and United States, with the biggest deposit being the Savkinsky deposit in Russia accounting alone for 25 Mt.

Table 2

World Production of Mineral Raw Materials of Interest/Not of Interest for Ocean Alkalinity Enhancement, in Mt yr^{-1}, in 2017 (Reichl et al., 2018; USGS, 2018)

Interest for OAE	Type of raw material	Production in 2017 (Mt yr^{-1})
Yes	Limestone	6,593
	Magnesite	29
	Olivine	8.4
	Brucite	1.5
	Total	6,631
No	Industrial minerals (sand, gravel, etc)	20,499
	Iron and ferro-alloy metals	1,639
	Non-ferrous metals	99
	Fossil fuels	14,664
	Total	36,902
Total world extraction of raw material		43,533

3.2. Mineral Raw Materials Production

Table 2 reports the main worldwide production in 2017 of different groups of mineral raw materials. The extraction of minerals of interest for OAE, 6.6 Gt yr^{-1}, accounts for about 15% of the total raw material world extraction (about 44 Gt yr^{-1}), where other major contributions (44%) come from industrial minerals (mainly sand and gravel) and fossil fuels (34%). The amount of coal (steam + coking + lignite) yearly extraction, 7.3 Gt yr^{-1} (Table SM4 in Supporting Information S1), is similar to that of limestone.

3.2.1. Limestone and Crushed Rocks

Crushed stones are used as construction material (i.e., for asphalt pavements) and as raw materials for cement production (in the case of limestone). Limestone is the most common crushed rock used worldwide, because of its relative softness that facilitates the excavation compared with other types of rock. In the United States, limestone adds to 70% of all crushed rocks production (USGS, 2019). Since no reliable data exist for many relevant countries, a conservative indirect assessment of limestone production is carried out based on the production of cement. An assessment of the global cement production is provided by USGS (2019) and amounts to 4.1 Gt yr^{-1} in 2017 (see Table SM5 in Supporting Information S1 for country data in the period 2011–2019). The global production of lime in 2017 amounts to 0.41 Gt yr^{-1} (see Table SM6 in Supporting Information S1 for country data in the period 2011–2019).

Available data on limestone production in 2017 in selected countries and the ratio between limestone and cement production is reported in Table SM7 in Supporting Information S1. Assuming, for a conservative estimate, a limestone/cement production ratio of 1.4, the average of the data available for 10 countries where both data were available, a global limestone production of around 6.6 Gt yr^{-1} is estimated. It should be noted that this is only a rough estimation, since cement can be made with marl substituting for limestone, and for some countries (i.e., United States) the limestone/cement ratio is far higher than the assumed value.

3.2.2. Olivine Production

Olivine global production for 2018 is assessed between 7.8 and 9.0 Mt yr^{-1} (Kremer et al., 2019). The two main producing countries are Norway, which has the largest commercial deposit in the world, and Japan. Other important producers in Europe are Greece, Spain and Turkey, with other countries outside Europe being China, Taiwan and Brazil (Table SM8 in Supporting Information S1).

3.2.3. Magnesite Production

The main magnesite producers are China, that with around 19 million tons in 2017 accounts for about 70% of the global production. China, Russia and Turkey together account for more than 80% of world production. The estimates of the global magnesite production vary widely, ranging from around 11 Mt in 2017 (Wietlisbach, 2018) to around 29 Mt for the same year (USGS, 2019), according to different data sources.

3.2.4. Brucite Production

With regards to brucite, it can be classified both as a magnesium metal ore or as an industrial mineral, but since there are no large, high grade, developed brucite deposits in production, natural brucite is not currently used as raw material for magnesium metal extraction (Simandl et al., 2007). Brucite current market is relatively small due to the rarity of high-grade deposits in comparison to other more common magnesium-bearing minerals, such as magnesite. The world brucite production ranges from 0.5 Mt yr^{-1} to 1.5 Mt yr^{-1} depending on the sources (Wietlisbach, 2018; Wilson, 2017).

3.3. Environmental Impacts and Risks

Although ocean alkalinization could be beneficial for the marine environment, different environmental issues should be considered.

Naturally occurring minerals like olivine or limestone for the production of lime are associated with various elements, depending on their source region. The main elements released through the use of lime, olivine or magnesite are Mg and Ca ions, along with minor elements like Fe and trace elements, that occur at relatively low concentrations in seawater, but their perturbation could be sufficient to have a toxic effect on sea organisms (Hartmann et al., 2013; Hauck et al., 2016; Moore et al., 2013).

Ca and Mg concentration changes in the ocean due to their addition on the surface will be small, due to the high background concentrations of these alkali metals in seawater (Bach et al., 2019), although the induced alkalinization could affect biological processes in some organisms, as shown in the paper by Cripps et al. (2013). With regards to Fe, whose concentration can be important in olivine, it has a low solubility in seawater (Boyd & Ellwood, 2010) so that a large fraction of the added Fe would presumably not become bioavailable, but if very large quantities of olivine are added to surface seawater, a corresponding quantity of Fe could become bioavailable (Hauck et al., 2016). Still regarding olivine, recent dissolution experiment by Montserrat et al. (2017) have shown that an increase in alkalinity is followed by an increase of dissolved Ni. Ni, although being a toxic metal for organisms, does not bio-accumulate in tissues, not affecting the food chain (Bach et al., 2019). However, Ni is likely one of the most important trace metal contaminants under olivine-based ocean alkalinization, but it is not the only one, with other potential trace metals (e.g., Cr, Cu, or Cd) (Simkin & Smith, 1970) that may bio-accumulate. The toxicity-related hypothesis above refers to scenarios where very high Ni and other trace elements concentrations occur. However, diluting the perturbation signal of potentially toxic elements in the oceans may make their concentration harmless for marine ecosystems (Bach et al., 2019).

About lime, which is made by calcining natural $CaCO_3$ rich source rocks with high purities, the majority of the remaining mass is composed of silicate and iron oxides. Therefore, Fe or Si addition will certainly be lower than from silicates such as dunite, mitigating the metal contamination risk (Bach et al., 2019; Choi et al., 2021).

Another aspect to be considered is the potential localized impact of spreading of mineral dissolution products on the ocean surface, since it could influence ocean biota in coastal and offshore regions. Logistically, minerals would most likely be added at discrete locations because an even distribution over entire ocean regions seems unfeasible (Kohler et al., 2013), potentially leading to hotspots of impact, depending on how minerals are added, on the type of material added, and on the attenuation/mixing in the surface ocean relative to dissolution rate of the mineral. Burt et al. (2021) show different carbon-uptake potentials in different regions of the world' oceans, mainly driven by the surface pattern of total alkalinity, but the limitation of the modeling experiments (i.e., little detail in the biological feedbacks of OAE), as well as differences with the results of previous works (Ilyina et al., 2013; Lenton et al., 2018), due to different modeling of the physical circulation and marine carbonate system states, suggest caution in defining single regions as most suitable for OAE.

A comprehensive evaluation of OAE and other carbon dioxide removal technologies requires a life-cycle approach that considers the entire supply chain, that is, the different technological options and energy sources available to produce slaked lime. The energy costs of OAE include mining, transformation, grinding, calcination, transportation on land and at sea, monitoring and verification, and for the renovation of existing bulk carrier ship or the construction of specialized vessels (Caserini et al., 2021). To date, a complete life-cycle assessment for OAE has not been conducted, and there are no empirical data for a scaled experiment or field trials with calculated energy budgets. According to NASEM (2021), this type of analysis is required and must be an element of future OAE research schemes.

4. Conclusions

The extraction and use of mineral resources have been a distinctive feature of human beings, have accompanied their civilization and have modified the Earth surface and the natural cycles. Today human activities are extracting about 44 billion tonnes of raw materials from the ground every year, for very different purposes. About one-third of them, 15 Gt, are mineral fossil fuels, used for energy generation or industrial production, and the

consequent CO_2 emissions are damaging the Earth's climate and are acidifying the oceans. The biogeochemical consequences of this interference are relevant.

The removal of CO_2 from the atmosphere and the contrast to ocean acidification through large scale ocean alkalinization require the extraction, transportation and dispersion of several gigatons of alkaline materials per year. For limestone, this corresponds globally to a limited upscale of the current production. In contrast, the increase of the extraction activities in some areas (i.e., near the coastlines), and of the calcination and slaking for the production of slaked lime, is much more relevant.

Although detailed local analyses are needed in order to evaluate the effective optimal excavation depth and the tonnage of the deposits, this work shows that pure limestone deposits close to the coasts could provide the required material for the alkalinization effort, with a limited upscaling compared to the current global extraction rate. Considering the area without or with low vegetation cover, those that can be a preferential target for geological exploration to assess the possibilities of large-scale exploitation are in north Africa, Iran and Australia. The potential resource of limestone in the area within 10 km from the coastline, in bare ground soil or covered with shrubs and scrubs is relevant, about 4,000 Gt, and rises to 15,000 and 40,000 Gt within 50 and 100 km from the coast, respectively.

Potential resources of olivine could be estimated in hundreds of billion tons, although a large fraction of these deposits is located far away from the coastline, raising the problem of transportation costs for their large-scale exploitation for ocean alkalinization. Regarding magnesite and brucite, their actual total production is orders of magnitude smaller than limestone, being a few tens of million tons compared to the billion tons of limestone. Their reserves are estimated to be around a few billion tons.

Another factor that should be taken into account is that the release of slaked lime in the ocean brings less environmental concerns than olivine, due to its lower content of potentially toxic elements.

The international effort to limit CO_2 emissions will lead to a substantial reduction in the enormous use of fossil fuels, in particular coal. Exploiting gigatons of limestone reserves per year for OAE could be a way to re-employ the equipment and manpower currently used in the coal sector, which currently extracts about 7,3 Gt yr^{-1}.

In conclusion, while the availability of limestone is not a constraint for the large-scale development of ocean alkalinization, the potential of the other raw materials such as olivine, brucite and magnesite is limited and could provide a minor contribution to ocean-based carbon removal strategies. Still, there is a strong need for optimizing the logistic of extraction and transportation activities at the global scale, as well to minimize the environmental impacts in order to fully deploy the potential benefit of ocean alkalinization for the sake of the climate and of the marine environment.

Data Availability Statement

Outcrops data of the global lithological map database are available in Hartmann and Moosdorf (2012). The shape file of the world coastlines and country borders has been taken from the EU Geographic Information System of the Commission (GISCO) portal (Eurostat, 2020). Data on land use/land cover has been taken from the ESRI, 2020 Land Cover (ESRI, 2020). The data set of worldwide natural protected areas in every continent is available in UNEP-WCMC (2022).

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