


## Article

# Potential of Tailing Deposits in Chile for the Sequestration of Carbon Dioxide Produced by Power Plants Using Ex-Situ Mineral Carbonation

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**Abstract:** In this study, the potential of copper tailing deposits in Chile for the sequestration of carbon dioxide (CO<sub>2</sub>) via ex-situ mineral carbonation integrating the recovery of valuable metals was assessed. An inventory of tailing deposits and CO<sub>2</sub> sources existing in Chile was constructed to determine the most suitable site for the installation of a future mineral carbonation plant and to evaluate the technical, economic, and environmental feasibility of CO<sub>2</sub> capture, separation, and transport from the source to the mineral carbonation plant. The data of the inventory of tailings deposits in Chile were obtained from the National Service of Geology and Mining. For the thermoelectric plants installed in Chile, data of energy production were obtained from the Energy National Commission. Through the use of the technique for order preference by similarity to ideal solution (TOPSIS) method and sensitivity analysis, the optimum location in the region of Antofagasta to install a mineral carbonation plant was identified. In addition, the results show that in the region of Antofagasta five tailing deposits have the potential to sequester between 66 to 99 Mt of CO<sub>2</sub>. Meanwhile, thermoelectric plants in 2018 produced about 9.4 Mt of CO<sub>2</sub> that is available to be sequestered, with a maximum generation potential of 21.9 Mt of CO<sub>2eq</sub> per year. The methodology and the study presented can be considered as a preliminary study to identify tailings that require further analysis.

**Keywords:** mine tailing deposits; CO<sub>2</sub> sequestration; mineral carbonation; mining waste



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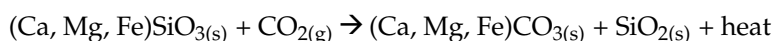
## 1. Introduction

The release of large amounts of greenhouse gases (GHGs) as a result of intensive human activities has contributed to global warming [1]. This warming occurs due to the presence of GHGs, which absorb the energy that would otherwise have escaped out into space, producing an increase in global temperature. Among the most important GHGs are CO<sub>2</sub>, CH<sub>4</sub>, N<sub>2</sub>O, SF<sub>6</sub>, hydrofluorocarbon (HFC), and perfluorocarbon (PFC), which were defined by the United Nations in the Kyoto Protocol. Of these gases, CO<sub>2</sub> has been reported to be the main substance of anthropogenic origin that increases the greenhouse effect [2]. For this reason, several technologies have been developed as measures to mitigate CO<sub>2</sub> emissions, either reducing anthropogenic CO<sub>2</sub> emissions or capturing and sequestering atmospheric CO<sub>2</sub> [3].

Within the technologies used for mitigation are the renewables energies, which are not completely free of emissions. Shrestha et al. [4] reported the average GHG emission rates estimated from several published studies on renewable energy processes. The emissions determined for the renewable energy sources in g CO<sub>2eq</sub>/kWh are: 86 for biomass, 71 for solar (solar photovoltaic), 67 for geothermal, 31 for wind, and 25 for hydroelectric. For

non-renewable energies (coal, oil, natural gas, and nuclear), average emissions are 1023, 780, 606, and 14 g CO<sub>2eq</sub>/kWh, respectively. Despite the increase in the use of renewable energies and the intensive search for better energy efficiency, fossil fuels are still preferred for energy generation due to their particular characteristics such as high energy density, easy use and storage, abundant supply, and, most attractive, their relatively low cost [5]. However, the combustion of fossil fuels releases large amounts of CO<sub>2</sub> to the atmosphere, accounting for up to 65% of the total gases expelled [6]. This situation will continue to increase globally, so supporting technologies for renewable energies have to be employed, such as CO<sub>2</sub> capture, either for long term storage or for reuse [7]. Within the technologies of carbon capture, use, and storage (CCUS), mineral carbonation (MC) is one of the most promising long term storage processes, since gaseous CO<sub>2</sub> is fixed as a solid, minimizing the risk of subsequent leaks into the atmosphere [8,9].

MC occurs when CO<sub>2</sub> reacts with oxides, hydroxides, or silicates of calcium (Ca), magnesium (Mg), and iron (Fe) to produce geologically stable carbonate species and silica. The following chemical reaction summarizes a family of reactions:



Usually, MC is an exothermic and spontaneous reaction in which conversion to carbonate is thermodynamically favored [10]. For example, for both Mg<sub>2</sub>SiO<sub>4(s)</sub> and CaSiO<sub>3(s)</sub> the reactions release 90 kJ/mol [5]. This process was proposed for the first time by Seifritz [11] to improve the natural silicate weathering process, which has occurred throughout the geological time scale.

The stability of the products of the above mineral carbonation reaction is explained by the carbon atom being at its lowest energy level in the carbonate, which provides a storage life of more than 100 thousand years [12]. This technology is also attractive because of the abundance in nature of this type of silicate, with the capacity to sequester all the CO<sub>2</sub> emitted by the processes of fossil fuel combustion [13]. Mineral carbonation is nothing more than a process of natural wear that occurs on geological scales, and therefore the process is extremely slow. Several studies have been presented to accelerate this process, including direct carbonation of the ore with CO<sub>2</sub> to indirect complex multi-stage processes [10,12,14]. The main interest of the studies in this field has been oriented to accelerate the mineral carbonation reaction speed in order to justify its industrial implementation. The simplest carbonation route in terms of process design and operation is the direct gas–solid route, although the reaction is very slow, even at high temperatures and pressures [15]. Figure 1 depicts the direct route compared with the indirect route. Indirect mineral carbonation is more favored through the indirect route since higher purity products are produced and the Ca and Mg conversion rate to carbonates is considerably higher [16].

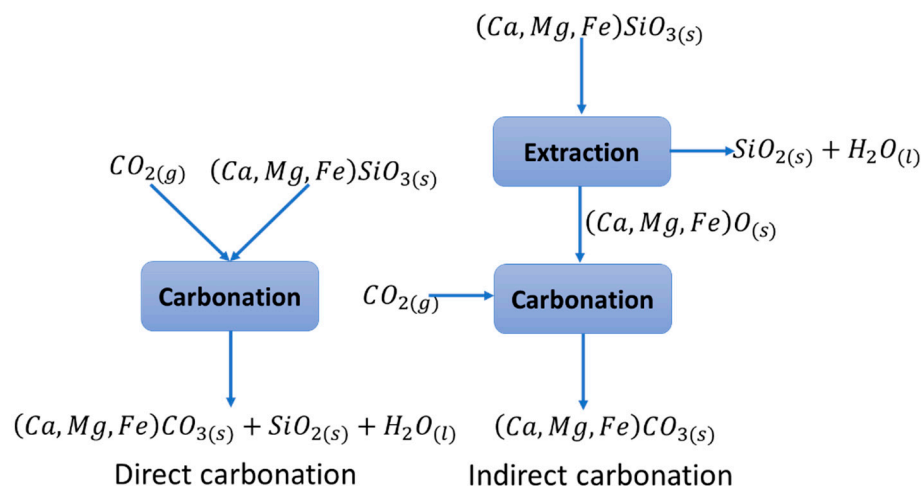


Figure 1. Direct and indirect carbonation gas–solid routes (adapted from Olajire [5]).

Nowadays, the use of mining waste for mineral carbonation has received considerable interest due to the high potential for commercialization towards mitigating climate change. For example, recently, it has been determined that the tailing of the Dumont Nickel Project of RNC Minerals can capture about 16% of the CO<sub>2</sub> annually emitted by their planned mining operation. In other words, the 15 Mt of tailings produced each year may sequester 21,000 tonnes of CO<sub>2</sub> per year by passive mineral carbonation [17]. Although CO<sub>2</sub> capture is not yet economically competitive with other technologies, new laboratory, pilot, and field-scale studies should help to reduce its costs, energy use, and environmental effects [18]. There are several reviews on MC in the literature. Li et al. [19] reviewed the integration of MC in the mining industry, including the modified passive mineral carbonation techniques in tailing facilities. Additionally, they analyzed the techno-economic assessments on existing integrated MC technologies. They concluded that value-added by-products would play an essential role in the future commercialization of this technology. More recently, Hills et al. [20] reviewed MC in mineral wastes with a focus on the manufacture of carbonate-cemented products. Since it is not economically profitable to extract calcium and magnesium to only carry out MC [21], it is necessary to give it a profit to promote the deployment of this technology, being able to achieve this through the recovery of valuable metals from mine tailings. Recently, Araya et al. [22,23] investigated the techno-economic feasibility of critical raw materials recovery from copper mine tailings in northern Chile. They obtained positive net present values for the production of rare earth oxides and vanadium pentoxide (V<sub>2</sub>O<sub>5</sub>). However, to analyze the potential of a tailings, it is necessary to evaluate different alternatives, including the extraction of valuable elements, elimination of toxic elements, new uses, among others. As in the initial phases of a study, not enough information is available to select technologies and carry out complete economic evaluations, the economic potential can be used to evaluate the economic performance of alternatives. For example, in the design of solids process the economic assessment between alternatives was performed at the initial step using the difference between the income from product sales and the cost of raw materials [24]. In another example, it was found that the income by sales can be an appropriate objective function to identify the optimal flotation circuit [25].

The main objective of this study was to analyze the potential of tailing deposits in Chile for the sequestration of CO<sub>2</sub> by ex-situ mineral carbonation integrating the recovery of valuable metals. To reach this main objective, an inventory of mine tailing deposits and CO<sub>2</sub> sources existing in Chile was constructed, and the most suitable sites for the installation of a future MC plant were identified. Additionally, the technical, economic and environmental feasibility of CO<sub>2</sub> capture, separation, and transport from the source site to the MC plant was discussed.

### 1.1. Mineral Carbonation

Mafic and ultramafic rocks are the most suitable tailing material for MC due to their high content of Ca, Mg, and Fe with low-grade potassium (K) and silica (SiO<sub>2</sub>), and because they are available in large amounts around the world [26]. These rocks were formed by mineral groups such as olivine (fayalite or Fe<sub>2</sub>SiO<sub>4</sub> and forsterite or Mg<sub>2</sub>SiO<sub>4</sub>), pyroxene (ferrosilite or FeSiO<sub>3</sub>, wollastonite or CaSiO<sub>3</sub>, and enstatite or MgSiO<sub>3</sub>), serpentine (Mg<sub>3</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub>), and Ca-plagioclase (anorthite or CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>). However, other minerals such as biotite, anorthoclase, anorthite, orthoclase, and hornblende have shown the ability of CO<sub>2</sub> capture [27,28]. These tailings can be treated using MC with either in-situ or ex-situ methods. The in-situ route refers to the process through which CO<sub>2</sub> is directly injected into the bedrock, either as a supercritical fluid or as an aqueous solution, at high concentration and high temperature [29], reaching reaction rates of a million times higher than those occurring in nature [30]. The constant monitoring of fugitive emissions is necessary by this route, due to the possibility of leakage of carbon dioxide into the atmosphere, as well as harmful impurities [31]. The ex-situ route refers to sending CO<sub>2</sub> and minerals to a chemical processing plant to induce MC. This involves a reduction in mineral size and the use of reagents (acid or base) in a multiple-stage, high-temperature, and high-pressure

process [32]. In ex-situ MC, industrial solid wastes can be used, which are generally alkaline, inorganic, and rich in calcium and magnesium [10,15]. The main advantages of using industrial solid wastes for MC are that they are reacted faster than minerals to this process, tend to have relatively open structures and larger reactive surface areas. Besides, they are available at low cost, with less pretreatment and less intensive energy conditions in their treatment required, and they can encapsulate certain toxic elements into the solid matrix [10].

Some of the industrial wastes that can be treated through MC are steel slag, cement kiln dust, recycled concrete aggregate, municipal solid waste, incineration ash, air pollution control residue, coal fly ashes, wood ash, red mud, paper mill waste ash, and mine tailings (MT). In recent years, several works have focused on the use of different types of mine tailings for ex-situ MC. For instance, Pasquier et al. [33] proposed an innovative approach for the production of pure  $\text{MgCO}_3$  through the direct capture of  $\text{CO}_2$  from flue gases and sequestration using serpentine mine tailings at moderate temperature and pressure conditions. Meyer et al. [34] studied the feasibility of using pyroxene-rich mine tailings for  $\text{CO}_2$  storage through the two-stage pH swing method. In the experiment at  $70^\circ\text{C}$  and a 2 M HCl solution, the overall conversion of cations was 30%, 9%, and 3% for Ca, Fe, and Mg, respectively. Ben-Ghacham et al. [27] studied the use of anorthosite mine tailings and waste concrete in ex situ MC under low pressure and ambient temperature for the aqueous route. They reached removals of 34% and 66% from inlet  $\text{CO}_2$  using anorthosite mine tailing and waste concrete, respectively. Boschi et al. [35] studied the spontaneous  $\text{CO}_2$  sequestration through serpentine carbonation at ambient temperature in a copper mine located in Italy.

### 1.2. Sources and Capture of Carbon Dioxide

$\text{CO}_2$  supply is another essential aspect to consider in MC processing of mine tailings. This gas is mainly produced from three sources: (1) industrial processes, (2) power production, and (3) fuel decarbonization. The largest  $\text{CO}_2$  sources are fossil fuels combustion power plants [36]. The advantage of sequestering  $\text{CO}_2$  emissions from power plants through MC has to do with the purity of the  $\text{CO}_2$  stream, due to the fact that impurities such as  $\text{NO}_x$  and  $\text{SO}_x$  do not have an impact on the carbonation process [37]. On the other hand, whether power plants are not located in the same site as the mine tailings, options for feedstock transport need to be evaluated [38]. Therefore, the evaluation of future MC projects should consider installing plants in sites with access to high  $\text{CO}_2$  supply. This evaluation plays an important role, since determining the most suitable site for the installation of a plant can minimize costs and harmful effects of the process on the environment [39]. For this purpose, mathematical and computational tools have been developed to support the evaluation of a finite number of decision alternatives, with the multi-criteria decision analysis methods (MCDA) being useful for selecting an optimum alternative, classifying alternatives, or ranking alternatives in a given preference order [40]. Specifically, the technique for order preference by similarity to ideal solution (TOPSIS) is a simple ranking method in its conception and application [41–44].

Within this context, the north of Chile appears to be a potential area for the installation of MC plants due to its extensive mining activity, mainly copper mining, located within the region of Antofagasta. This region produces more than half of the total amount of copper generated in Chile [45]. In 2017, 71% of the refined copper in Chile was produced using concentration mill processes (from primarily sulfide copper minerals), whereas 29% was produced via leach, solvent extraction, and electrowinning processes (for secondary sulfide and oxidized copper minerals) [46]. This trend will increase in the future due to the reduction of oxidized minerals and the release of primarily sulfide minerals [47]. Thus, an increase is expected in the amount of copper mine tailings generated by flotation and concentration processes, which, in Chile, are electrically supplied by the National Electricity System (SEN), formed by two previous suppliers: the Interconnected Central System (SIC) and the Interconnected System of the Great Northern Region (SING). In 2017, 59% of the electricity needs of the copper production industry was supplied by the SING system [46],

whose electricity is generated from fossil fuel sources. One objective of this study was to assess the potential of tailings deposits in Chile for sequestration of CO<sub>2</sub> by ex-situ MC, determining the most suitable site for installation of a MC plant, using the TOPSIS method.

## 2. Methodology

The methodology employed in this study consists of four main procedures: (1) construction of an inventory of the existing MT in Chile, (2) construction of an inventory of the thermoelectric power plants that provide energy to the mining plants, (3) application of the TOPSIS method, and (4) sensitivity analysis for site selection.

The purpose of constructing the MT inventory was to determine the quantity of CO<sub>2</sub> that can be sequestered using those tailings as feedstock for MC, and identifying the other valuable species that can be recovered from this tailing processing. The inventory of the thermoelectric power plants was constructed to quantify the CO<sub>2</sub> that is available for sequestration. Next, TOPSIS was applied for the selection of the best site for installing a Mineral Carbonation plant. Finally, sensitivity analysis was performed to determine the influence of the different criteria for selecting the most suitable site for the installation of a MC plant.

## 3. Collected Information

The information collected and the number of tailing deposits and power plants are described and discussed in this section.

### 3.1. Inventory of Tailing Deposits

The data on tailing deposits in Chile were obtained from the National Service of Geology and Mining (SERNAGEOMIN) [48], which is the technical governmental entity responsible for the information of geological resources of the country. The data are permanently updated since the first version of the data was published in 2017. For the first version, the source information was obtained from samples taken during two years, corresponding to three sample locations: the surface of the bucket; the surface of the dam walls; and the downstream soil sediments. After 2017, the geochemical study included depth drilling of some tailings deposits. The estimation of the CO<sub>2</sub> storage capacity of geological resources is the overriding factor to determine suitable CO<sub>2</sub> storage sites [49]. Over 740 tailings deposits were registered in Chile as of 2019, which are distributed between the regions of Tarapacá in the north and Aysén in the south. The largest number of tailings is found in the regions of Coquimbo, Atacama, and Valparaiso (52.0%, 21.5%, and 10.5%, respectively). The large number of tailings found in the Coquimbo region is due to the existence of many small mineral processing plants, which generate small tailing deposits. Regarding the operational status, 14.0% of the deposits are active, 23.4% are abandoned, and 62.6% are inactive. Active deposits are currently accumulating tailings; inactive deposits have reached the end of their useful life without executing legal closure but have a known responsible person; abandoned deposits have reached the end of their useful life, without executing legal closure and without a legal responsible person identified. In terms of volume, the 740 deposits have the capacity to contain a total volume of about 16,000 Mm<sup>3</sup>. Figure 2 depicts the tailing deposits in Chile by region, including active, inactive, and abandoned tailing deposits, whereas Table 1 summarizes the current and approved volume of active tailing deposits.

Since 27 active tailings contribute 92.7% of the overall current tonnage of the 740 deposits, we used the data of these 27 active tailings deposits to analyze their chemical compositions to obtain an average value for further studies. The average chemical composition of these 27 tailing deposits active is presented in Table 2. In Table 2, REE means rare earth elements; minors are trace elements (below 100 ppm); majors are rock forming elements, expressed as oxides; SD is the standard deviation. REE includes La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Sc, and Y; minors considers Co, Ni, Cr, Nb, Cs, Hf, Ta, Th, U, Mo, Sb, Sn, Ag, Cd, Bi, W, Au, and Hg; and majors corresponds to MnO, P<sub>2</sub>O<sub>5</sub>,

and SO<sub>3</sub>. Furthermore, these values make it possible to assess the potential ecological risks of tailing deposits, through the concentrations of As, Cd, Cr, Cu, Hg, Pb, Ni, Zn, and using as a base the methodology proposed by Lam et al. [50].

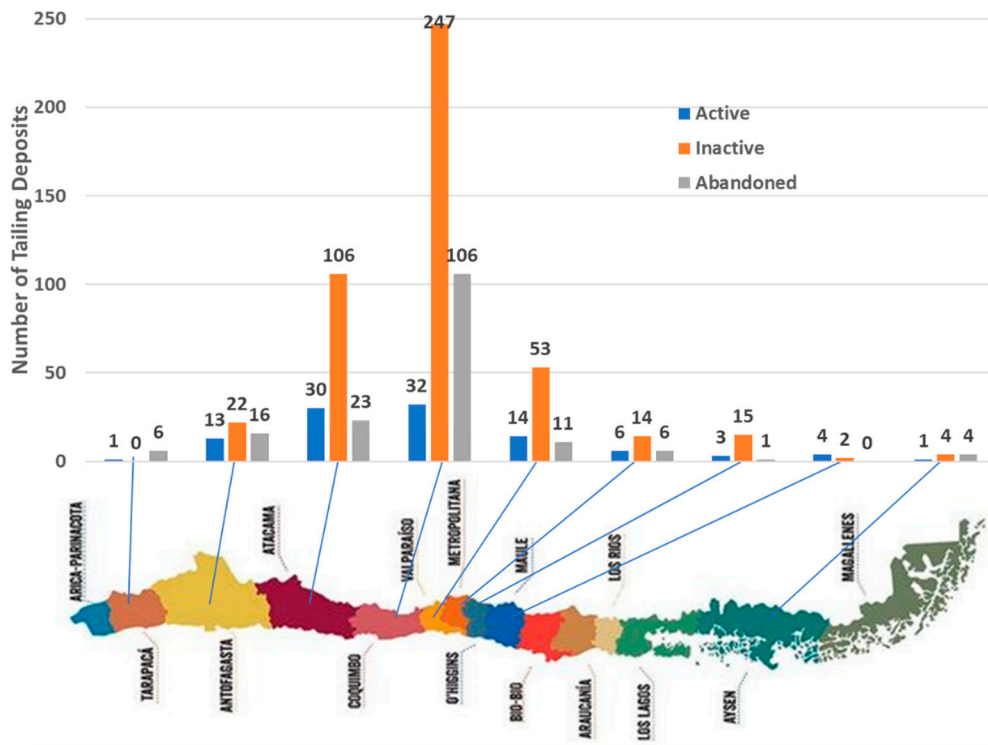


Figure 2. Number of tailing deposits in Chile 2019 by region.

Table 1. Current and approved volume of active tailing deposits. Vol., volume.

Mining Size	Amount (N°)	Current Vol. (Mm <sup>3</sup> )	(%)	Approved Vol. (Mm <sup>3</sup> )	(%)	Filling (%)
Small and medium	77	69	1.0	189	1.2	36.51
Large	27	6639	99.0	14,939	98.8	44.44
Total	104	6708	100.0	15,128	100.0	44.34

Table 2. Descriptive statistics of chemical composition and concentration (wt.%) of tailing deposits active in Chile: values calculated based on data. REE: rare earth elements; SD: standard deviation.

Descriptive Statistics	Cu	TiO <sub>2</sub>	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	Na <sub>2</sub> O	K <sub>2</sub> O	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Majors
Mean	0.098	0.565	9.570	3.326	3.159	2.645	3.424	14.497	56.949	5.332
Median	0.067	0.550	5.860	2.690	2.430	1.910	3.570	14.430	60.070	4.740
SD	0.087	0.189	8.170	2.348	1.973	2.733	1.339	3.034	9.728	3.666
Kurtosis	0.000	5.764	-0.792	1.241	0.942	20.187	-0.232	-0.610	-0.669	12.597
Skewness	0.000	1.474	0.870	0.977	1.132	3.887	-0.313	0.087	-0.467	3.102
Min	0.004	0.180	1.650	0.370	0.210	0.410	0.450	8.770	32.660	1.730
Max	0.367	1.400	27.530	11.020	9.260	18.350	6.010	21.450	73.570	23.870
Descriptive Statistics	V	Rb	Zn	Sr	Zr	Ba	Pb	As	REE	Minors
Mean	0.013	0.010	0.027	0.025	0.027	0.058	0.021	0.009	0.016	0.036
Median	0.013	0.008	0.010	0.015	0.026	0.053	0.006	0.002	0.016	0.034
SD	0.004	0.006	0.051	0.022	0.014	0.030	0.051	0.022	0.006	0.012
Kurtosis	0.000	0.000	0.001	0.000	0.000	0.000	0.002	0.001	0.000	0.000
Skewness	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Min	0.004	0.002	0.004	0.001	0.007	0.002	0.001	0.002	0.007	0.012
Max	0.027	0.033	0.262	0.076	0.061	0.125	0.259	0.106	0.037	0.082

Elements of interest with high concentrations are observed in Table 2, such as Cu, TiO<sub>2</sub>, Fe<sub>2</sub>O<sub>3</sub>, CaO, MgO, and REE. Elements such as V, Rb, Zn, Sr, Zr, and Ba are in percentages above 0.01%. Then, using these percentages as ore grade, that is, the ratio of contained valuable species per unit of ore (commonly expressed as a percentage, e.g., %Cu) and knowing the total tonnage of the tailings (Ton<sub>Total</sub>), the tonnage of valuable species to be recovered (Ton<sub>Val</sub>) was determined according to Equation (1).

$$\text{Ton}_{\text{Val}} = \text{Ton}_{\text{Total}} \times \text{ore grade} \quad (1)$$

Then, considering the species with high concentration and those that cannot be treated by MC (Cu, TiO<sub>2</sub>, and REE), the income potential generated by their recovery can be calculated according to Equation (2) considering the market values of 6504, 9100 and 279,388 USD/t for Cu, TiO<sub>2</sub>, and mean REE., respectively.

$$\text{Income potential} = (\text{Ton}_{\text{Cu}} \times \text{USD}/\text{t}_{\text{Cu}}) + (\text{Ton}_{\text{TiO}_2} \times \text{USD}/\text{t}_{\text{TiO}_2}) + (\text{Ton}_{\text{REE}} \times \text{USD}/\text{t}_{\text{REE}}) \quad (2)$$

When considering the species treatable by MC, the theoretical potential of sequestration can be estimated considering a maximum and minimum (Total CO<sub>2</sub> (min, max)). This is calculated using the maximum and minimum theoretical capacity of the sequestration of some minerals (CO<sub>2max.cap</sub>), which are given in g of CO<sub>2</sub> that can be sequestered for each 100 g of mineral feed. Based on the methodology proposed by Vogeli et al. [51], and assuming the mineralogical compositions from copper mine tailings in northern Chile are obtained using the quantitative evaluation of minerals by scanning electron microscopy (QEMSCAN) in the studies presented by Lam et al. [52] and Medina et al. [53], chlorite is the mineral with the highest capacity, at 39.6 g CO<sub>2</sub>/100 g chlorite, whereas muscovite has the lowest capacity, at 5.5 g CO<sub>2</sub>/100 g muscovite. Table 3 lists more details. The minerals indicated in Table 3 were obtained from Chilean tailings analysis, available in the literature. The references where they were identified correspond to magnetite, chlorite, albite, and muscovite [52–57], biotite [52,54,56,57], orthoclase [52–54,56], albite [52–54,57], anorthite [53,55], titanite, hornblende, and montmorillonite [52,57]. Additionally, amphibole-pyroxene was observed in Medina et al. [53] but was not considered in this study.

**Table 3.** Maximum theoretical mass of CO<sub>2</sub> in (g of CO<sub>2</sub>/100 g mineral) that can be sequestered by minerals.

Group	Mineral (100 g)	Direct Mineral Carbonation Reaction	CO <sub>2max.cap</sub> (g of CO <sub>2</sub> /100 g Mineral)
Oxides	Magnetite	$\text{Fe}_3\text{O}_4 + \text{CO}_2 + \text{H}_2\text{O} \rightarrow \text{FeCO}_3 + 2\text{FeOOH}$	19.0
Phyllosilicates	Biotite	$\text{KMg}_2\text{FeAlSi}_3\text{O}_{10}(\text{OH})_2 + 3\text{CO}_2 \rightarrow 2\text{MgCO}_3 + \text{FeCO}_3 + \text{KAlSi}_3\text{O}_8 + \text{H}_2\text{O}$	29.4
	Chlorite	$\text{Mg}_5\text{Al}_2\text{Si}_3\text{O}_{10}(\text{OH})_8 + 5\text{CO}_2 \rightarrow 5\text{MgCO}_3 + \text{Al}_2\text{O}_3 + 3\text{SiO}_2 + 4\text{H}_2\text{O}$	39.6
	Montmorillonite	$\text{Na}_{0.33}\text{Mg}_2\text{Si}_4\text{O}_{10}(\text{OH})_2\text{H}_2\text{O} + 2\text{CO}_2 \rightarrow 2\text{MgCO}_3 + 0.33\text{Na}_2\text{O} + 4\text{SiO}_2 + 2\text{H}_2\text{O}$	23.1
Tectosilicates	Muscovite	$2\text{KAl}_2(\text{AlSi}_3\text{O}_{10})(\text{OH})_2 + \text{CO}_2 \rightarrow \text{K}_2\text{CO}_3 + 3\text{Al}_2\text{O}_3 + 6\text{SiO}_2 + 2\text{H}_2\text{O}$	5.5
	Orthoclase	$2\text{KAlSi}_3\text{O}_8 + \text{CO}_2 \rightarrow \text{K}_2\text{CO}_3 + \text{Al}_2\text{O}_3 + 6\text{SiO}_2$	7.9
	Albite	$2\text{NaAlSi}_3\text{O}_8 + \text{CO}_2 \rightarrow \text{Na}_2\text{CO}_3 + \text{Al}_2\text{O}_3 + 6\text{SiO}_2$	8.4
Other silicates	Anorthite	$\text{CaAl}_2\text{Si}_2\text{O}_8 + \text{CO}_2 \rightarrow \text{CaCO}_3 + \text{Al}_2\text{O}_3 + 2\text{SiO}_2$	15.8
	Titanite	$\text{CaTiSiO}_5 + \text{CO}_2 \rightarrow \text{CaCO}_3 + \text{TiO}_2 + \text{SiO}_2$	22.5
	Hornblende	$\text{Ca}_2\text{Mg}_4\text{Al}(\text{Si}_7\text{Al})\text{O}_{22}(\text{OH})_2 + 6\text{CO}_2 \rightarrow 4\text{MgCO}_3 + 2\text{CaCO}_3 + \text{Al}_2\text{O}_3 + 7\text{SiO}_2 + \text{H}_2\text{O}$	32.4

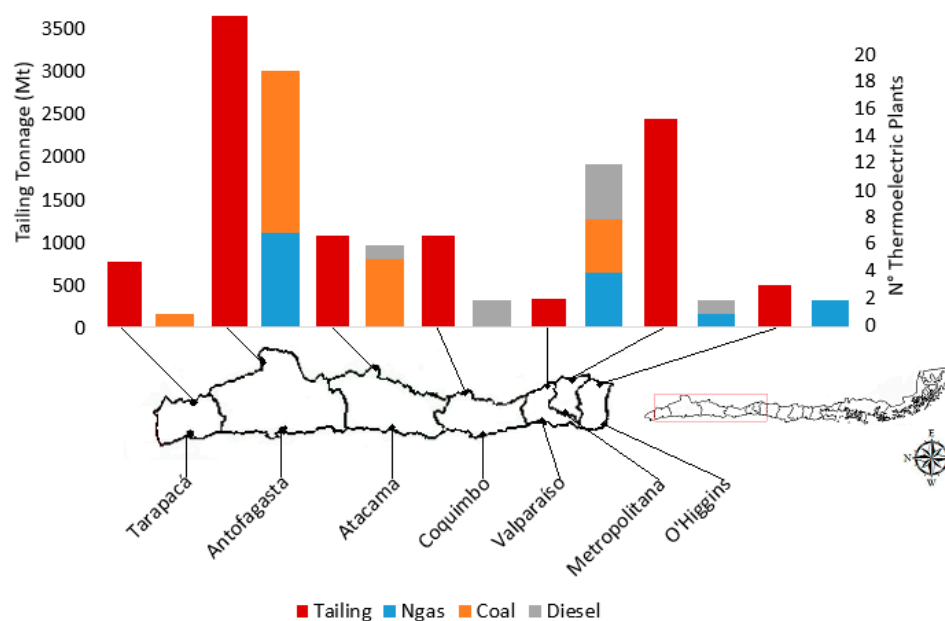
The mineralogical compositions of the 27 active tailings were not available, so it was assumed that all the tonnage values obtained from Equation (1) for the species of Na, K, Ca, Mg, and Fe are present as the minerals presented in Table 3 (Ton<sub>(Na,K,Ca,Mg,Fe)</sub>). Then, using Equation (3), the amount of CO<sub>2</sub> that can be sequestered by the 27 active tailings for each of the species (CO<sub>2SEQ</sub>) was determined.

$$\text{CO}_{2\text{SEQ}} = \text{Ton}_{(\text{Na,K,Ca,Mg,Fe})} \times \text{CO}_{2\text{max.cap}}/100 \quad (3)$$

Whether the minimum and maximum values of  $\text{CO}_2\text{SEQ}$  for each of the Na, K, Ca, Mg, and Fe species are considered, the minimum and maximum amount of  $\text{CO}_2$  that can be sequestered can be determined, as shown in Equation (4).

$$\text{Total CO}_{2(\text{min})} = (\text{CO}_{2\text{SEQ by Ca}})_{(\text{min})} + (\text{CO}_{2\text{SEQ by Mg}})_{(\text{min})} + (\text{CO}_{2\text{SEQ by Fe}})_{(\text{min})} \quad (4)$$

These 27 active tailings deposits are distributed between the Tarapacá and the O'Higgins regions (2000 km approx.). Considering this distance, a single MC plant to treat all the tailings would not be environmentally feasible due to the contamination produced by transporting only the tailings to the MC plant, which would considerably reduce the overall efficiency of the sequestration process, directly increasing the total cost of  $\text{CO}_2$  avoided (USD/t of  $\text{CO}_2$ ), which is already high. The idea was to integrate the MC plant, the tailings deposit, and the source from where the  $\text{CO}_2$  is obtained. The options for transporting  $\text{CO}_2$  were considered in the analysis, because the cost of  $\text{CO}_2$  transport, whether by trucks or pipes, increases with distance [51]. This aspect played an important role in the evaluation of the viability of the MC operation. The location of the 27 active tailing deposits is shown in Figure 3, which shows that the largest tonnage of active tailings, 37.1 wt.%, is concentrated in the Antofagasta region, as well as the largest number of thermoelectric plants with installed capacity greater than 100 MW. Therefore, the Antofagasta region is considered the best location for the installation of a MC plant.



**Figure 3.** Distribution of active tailing deposits and thermoelectric plants in Chile by region.

### 3.2. Inventory of Thermoelectric Power Plants

The inventory of thermoelectric power plants was constructed for the Antofagasta region, and energy production data were obtained from the National Energy Commission (CNE) [58], which is the governmental entity responsible for the management of the technical standards with which companies involved in the production, generation, transport, and distribution energy must comply. The entity responsible for overseeing emission standards of thermoelectric plants is the Superintendence of the Environment, which through the legal standard Supreme Decree (DS, from the Spanish Decreto Supremo) N°13/2011, establishes the maximum emission limits for the exhaust gas flow ( $\text{Nm}^3/\text{h}$ ) and defines the source term as the electric generation unit that is operating or declared under construction. The emission limits depend on the type of fuel and whether the plant is already in operation or is new. Table 4 shows these values by compounds. However, the DS N°13/2011, does not establish a limit for other pollutants, harmful heavy metals, and



CO<sub>2</sub> emissions; for the latter it only establishes its continuous monitoring. Moreover, it does not clearly specify what will be the penalties that will be applied to those plants that do not comply with the stated emission limits [59].

**Table 4.** Emission limit values in Chile, according DS N°13/2011 [59]. PM2.5: particulate matter 2.5.

State of the Plant	Fuel	PM2.5 (mg/Nm <sup>3</sup> )	SO <sub>2</sub> (mg/Nm <sup>3</sup> )	NO <sub>x</sub> (mg/Nm <sup>3</sup> )	Hg (mg/Nm <sup>3</sup> )
In operation	Solid	50	400	500	0.1
	Liquid	30	30	200	-
	Gas	-	-	50	-
New	Solid	30	200	200	0.1
	Liquid	30	10	120	-
	Gas	-	-	50	-

On the other hand, in the Antofagasta region, 19 thermoelectric plants have an installed capacity exceeding 100 MW, with a notable preference for the generation of energy by burning coal (Figure 3). In 12 plants, the installed capacity is about 2345 MW. The power generation efficiency (PGE) of each plant is determined according to Equation (5).

$$PGE = (ENE_{2018} / (\text{Cap} \times T)) \times 100 \quad (5)$$

In this equation, T is the number of operation hours when the plant is active throughout the year, Cap is the installed capacity of each power plants, measured in MW, and (ENE<sub>2018</sub>) is the total energy produced by each power plants during 2018, measured in MWh/year. The value of T is determined as 8760 h/year, assuming that the thermoelectric plants operates during all year without interruptions. This is carried out to calculate the maximum amount of energy generated by each plant, as well as the maximum quantity of GHG emitted per year (CO<sub>2eq,max</sub>). This number was obtained by multiplying the energy generation by the respective emission factor (EMF), according to Equation (6). The emission factors for each fuel area were taken from Shrestha et al. [4]: 1023 CO<sub>2eq</sub>/kWh for coal, 780 CO<sub>2eq</sub>/kWh for oil, and 606 g CO<sub>2eq</sub>/kWh for natural gas:

$$CO_{2eq,MAX} = \text{Cap} \times T \times \text{EMF} \quad (6)$$

To calculate the amount of CO<sub>2</sub> emitted, instead of the amount of CO<sub>2eq</sub> emitted, the composition of the exhaust gases of the thermoelectric plants that burn oil, coal, and natural gas obtained from Olajire [32], as well as the global warming potential (GWP) for each of the gases, is considered. The exhaust gases contain 54.91%, 59.94%, and 80.99% CO<sub>2</sub> for oil, coal, and natural gas, respectively. Then, the amount of CO<sub>2</sub> generated by the thermoelectric plants in Antofagasta during 2018 was determined using Equation (7).

$$CO_{2,2018} = ENE_{2018} \times \text{EMF} \times \%CO_2 / 100 \quad (7)$$

#### 4. Selection of Sites for MC

The TOPSIS method was used for the selection of the best site for installing a MC plant. Additionally, a sensitivity analysis was conducted to determine the influence of the different criteria on the selected site for the installation of the MC plant.

TOPSIS is a decision-making technique or MCDA, which, through a sequence of steps, is used to select the best alternative when comparing different criteria of a set of alternatives. Within these alternatives, none have exhibited superior performance for all criteria and an optimum alternative cannot be chosen [44]. The choice can be made by one or several experts, and, by using linguistic terms or numeric scale. However, its use and the selection of the criteria, the importance criteria weights, and the performance rating of each decision-maker must be chosen on a case-by-case basis. This technique was first proposed by Hwang and Yoon [60] on the basis that the best alternative must be located at

the shortest distance of an ideal value and simultaneously at the greatest distance of an anti-ideal value. These distances were calculated to determine the closeness coefficient of each alternative, according to the following steps:

Step 1: Assignment of the set of alternatives ( $a$ ), the criteria ( $i$ ), and criteria weights ( $w_i$ ). The performance of  $n$  alternatives  $a$  with respect to  $m$  criteria  $i$  are gathered in a decision matrix  $X = (x_{ia})$ , where  $i = 1, \dots, m$  and  $a = 1, \dots, n$ .

Step 2: The performance of the different criteria is normalized to enable comparison of the measurements with different units (e.g., km, USD, Mt). Here, several normalization methods can be used. In this work, the distributive normalization was employed, where the performance is divided by the square root of the sum of each squared element in a column:

$$r_{ia} = \frac{x_{ia}}{\sqrt{\sum_{a=1}^n x_{ia}^2}} \text{ where } i = 1, \dots, m; \text{ and } a = 1, \dots, n. \quad (8)$$

Step 3: The normalization values ( $r_{ia}$ ) are multiplied with their respective values of the criteria weights ( $w_i$ ) using Equation (9), and a weighted normalized decision matrix is generated.

$$V_{ia} = w_i \times r_{ia} \quad (9)$$

Step 4: The values of the weighted normalized decision matrix ( $V_{ia}$ ) are each compared with the ideal value ( $V_i^+$ ) and with the anti-ideal value ( $V_i^-$ ) of their respective criteria.  $V_i^+$  and  $V_i^-$  are obtained, when collecting the best and worst performance on each criterion of the normalized decision matrix, according to Equations (10) and (11), respectively.

$$A^+ = (V_1^+, \dots, V_m^+) \quad (10)$$

$$A^- = (V_1^-, \dots, V_m^-) \quad (11)$$

where  $V_i^+ = \max_a(V_{ia})$  if criterion  $i$  is to be maximized and  $V_i^- = \min_a(V_{ia})$  if criterion  $i$  is to be minimized.

Step 5: Calculate the distance between  $V_{ia}$  and  $V_i^+$  and between  $V_{ia}$  and  $V_i^-$  using Equations (12) and (13), respectively. We used a Euclidean distance, but another metric can be applied.

$$d_a^+ = \sqrt{\sum_i (V_i^+ - V_{ia})^2} \quad (12)$$

$$d_a^- = \sqrt{\sum_i (V_i^- - V_{ia})^2} \quad (13)$$

Step 6: The closeness coefficient ( $CC_a$ ) is determined for each of the alternatives using Equation (14).  $CC_a$  varies between 0 and 1, where the best results are closer to 1, and the worst results are closer to 0.

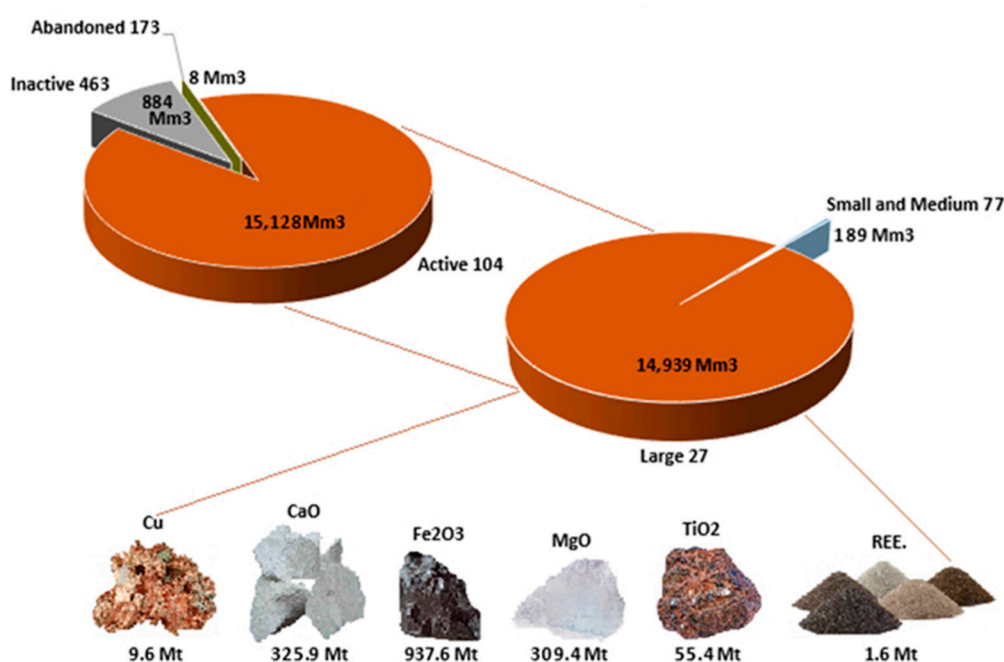
$$CC_a = \frac{d_a^-}{d_a^+ + d_a^-} \quad (14)$$

Step 7: The alternatives are ranked according to the  $CC_a$  in decreasing order. The alternative with the highest score is selected as the most suitable site for the installation of a MC plant.

Finally, the sensitivity analysis was performed to determine the sensitivity of the general decision to small changes in the criteria weights assigned during TOPSIS application. This was achieved by slightly varying the values of weights of each criterion and then observing its effect on the general decision.

## 5. Results and Discussion

Figure 4 summarizes the amounts of valuable species that could be recovered from the largest tailing deposits in Chile, considering only the species that occur in larger quantities. Recovering all the species would not be feasible due to their low concentration.



**Figure 4.** Valuable species tonnage that can be recovered from the largest tailing deposits in Chile.

The results indicated that 325.9 Mt of Ca, 937.6 Mt of Fe, and 309.4 Mt of Mg exists, which can be used as feedstock for MC. Additionally, there are 9.6 Mt of Cu, 55.4 Mt of TiO<sub>2</sub>, and 1.6 Mt of REE, contained in 27 of the total 740 deposits. On the other hand, Vogeli et al. [51] determined that the tailings rich in Ca and Mg produced by the main platinum mining industries in South Africa (Anglo American, Implats, Northam, and Lonmin) generated about 77.51 Mt/year of tailings between June 2009 and June 2010, which had the potential to theoretically sequester a total of about 13.94 Mt of CO<sub>2</sub>/year.

Chile generated around 318 Mt of tailings in 2012, about 511 Mt in 2016, and, by 2020, is expected to generate around 600 Mt [61]. Of the 27 active tailings deposits, 37.1% (wt.%) of the total tonnage are located in the Antofagasta region in six large-scale active deposits. The deposit of Laguna Seca has the largest sequestration potential of 44.09 Mt of CO<sub>2</sub> due to its large tonnage, whereas the Esperanza deposit can sequester the greatest amount of CO<sub>2</sub> per ton of tailings (0.044 t of CO<sub>2</sub>/t of tailings). The Mantos Blancos deposit can generate the highest income potential per ton of tailings (124.35 USD/t of tailing). The El Peñón deposit has the lowest sequestration potential with 0.46 Mt of CO<sub>2</sub>. Table 5 provides details of these results.

Five of the six deposits identified in Table 5 generate tailings from copper extraction, whereas El Peñón generates tailings from gold extraction. Additionally, the Mantos Blancos and Sierra Gorda deposits have high concentrations of REE (240 and 220 ppm, respectively), whereas the concentration in the rest of the world varies between 0.5 and 50 ppm. REE prices range between 5.5 to 1258 USD/kg for CeO<sub>2</sub> and Lu<sub>2</sub>O<sub>3</sub>, respectively. Esperanza, Laguna Seca, and Mantos Blancos deposits have titanium oxide grades over 5000 ppm, whereas Talabre deposit has a copper grade over 2000 ppm. In addition, Lam et al. [50] performed a study using the same data from the National Service of Geology and Mining [48], and they determined that none of these six tailings deposits in Antofagasta are located at less than 2 km from a water body and at less than 2 km from a populated area, so regarding their location, these would not pose a potential environmental risk. However, regarding their heavy metal contamination it is possible to assess the potential ecological risks, so the six tailings were evaluated for As, Cd, Hg, Pb, Cu, Ni, Zn, Cr. Table 6 provides these results, and details of their determination can be found in the Supplementary Materials. Metal such as Ni, Zn, Cu and Cr, do not represent an environmental risk, unlike Pb, Hg, Cd, and As, all of them are highly toxic and the last three are carcinogens. The Esperanza

deposit has the lowest risk index (RI), because none of its heavy metals are above the high level, followed by the Sierra Gorda deposit with one heavy metal at a very high level, the rest of the deposits have the RI at an extremely high level. El Peñon deposit has the highest RI, with this value far above the rest and with four heavy metals at extremely high levels, so this deposit for the authorities should represent a priority in its management.

**Table 5.** Potential of mass of CO<sub>2</sub> to be sequester and mean sequestration efficiency (MSE) with tailing deposits of the Antofagasta region.

Tailing Deposit	Current Ton (Mt)	Total CO <sub>2</sub> (Mt)		MSE	%			Income Potential
		CO <sub>2</sub> min	CO <sub>2</sub> max	(t of CO <sub>2</sub> /t of Tailings)	Cu	TiO <sub>2</sub>	REE	(USD/t of Tailings)
Mantos Blancos	131	3.22	4.59	0.030	0.193	0.500	0.024	124.35
Esperanza	240	8.41	12.62	0.044	0.040	0.590	0.015	97.24
Sierra Gorda	143	3.25	4.86	0.028	0.052	0.490	0.022	109.18
Talabre	1793	21.85	31.96	0.015	0.230	0.180	0.011	63.08
Laguna Seca	1302	28.95	44.09	0.028	0.103	0.525	0.016	99.64
El Peñon	21	0.31	0.46	0.019	0.048	0.280	0.017	75.71
Total	3630	66.01	98.57					

**Table 6.** Average heavy metal concentration values and potential ecological risk index for tailing deposits of the Antofagasta region (see last column for color definition).

Tailing Deposit	Ecological Risk Factor (E <sub>r</sub> ) (Dimensionless)								Potential Ecological Risk Index (ΣE <sub>r</sub> )	Contamination Level
	As	Cd	Hg	Pb	Cu	Ni	Zn	Cr	RI	
Mantos Blancos	40	223	5	89	72	10	1	5	444	Low
Esperanza	40	33	73	50	15	5	2	3	220	Middle
Sierra Gorda	40	140	11	56	19	5	1	3	276	High
Talabre	1562	65	143	470	85	9	2	2	2339	Very high
Laguna Seca	40	67	355	65	38	2	5	4	575	Extremely high
El Peñon	1953	327	596	1615	18	9	25	2	4545	

Regarding thermoelectric plants, Table 7 lists the existing plants with an installed capacity exceeding 100 MW (19 plants), of which the most are located in the district of Mejillones, followed by Tocopilla and then Taltal. The higher use of coal plants than natural gas plants was observed (PGE%), mainly due to the discontinuous supply of natural gas from Argentina, which is the fuel used by some of these plants.

However, the natural gas shipment of gas from Argentina to Chile has been restored, which provides Chile with new opportunities to decarbonize its energy matrix, as well as to increase the flexibility of its electrical system. The main results show that 19 plants together generated a total of 15,954 GWh in 2018, producing a total of 9.37 Mt of CO<sub>2</sub> that is available to be sequestered, with a maximum potential of 21.89 Mt of CO<sub>2</sub>eq per year. Finally, five large-scale active deposits have the potential to sequester about 66 Mt of CO<sub>2</sub>, considering the least favorable case (total CO<sub>2</sub>min). Only in the processing of the Talabre or Laguna Seca deposits using MC could practically all the CO<sub>2</sub> generated in 2018 by thermoelectric plants could be sequestered. This presents a considerable challenge, considering that the project in operation with the largest CO<sub>2</sub> capture capacity in the world is Century Plant, capturing 8.4 Mt of CO<sub>2</sub> per year, followed by Shute Creek Gas Processing Plant, capturing 7.0 Mt of CO<sub>2</sub> per year, both located in the USA [62].

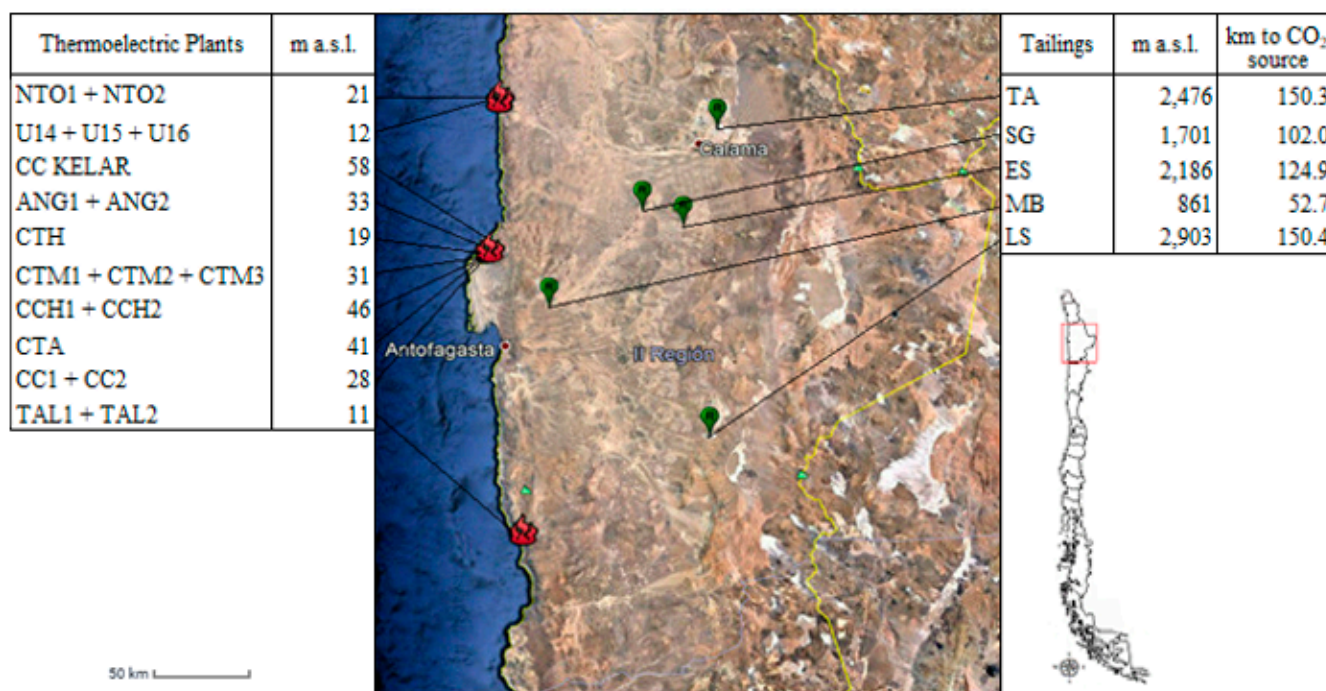
**Table 7.** Distribution, capacity, efficiency (PGE), and CO<sub>2</sub> to be sequestered of thermoelectric plants greater than 100 MW in Antofagasta region. NGas: natural gas; ANG1, ANG2: Angamos thermoelectric power plants; CC KELAR: Kelar thermoelectric power plant; CCH1, CCH2: Cochran thermoelectric power plants; CC1, CC2: Atacama thermoelectric power plants; CTA: Andina thermoelectric power plant; CTH: Hornitos thermoelectric power plant; CTM1–3: Mejillones thermoelectric power plants; NTO1, NTO2: Norgener thermoelectric power plants; TAL1, TAL2: Taltal thermoelectric power plants; U14–16: Tocopilla thermoelectric power plants.

Facility	Location	Source	Capacity (MW)	PGE (%)	ENE <sub>2018</sub> (GWh/year <sub>2018</sub> )	CO <sub>2eq</sub> .MAX (Mt/year)	CO <sub>2</sub> .2018 (Mt/year <sub>2018</sub> )
NTO1 + NTO2	Tocopilla	Coal	276.39	75.6	1831	1.48	1.12
U14–U16	Tocopilla	Coal/NGas	631.10	36.1	1997	3.00	1.10
CTM1–CTM3	Mejillones	Coal/NGas	583.29	23.4	1197	2.87	0.69
CTH	Mejillones	Coal	177.54	60.8	945	0.95	0.58
ANG1 + ANG2	Mejillones	Coal	558.20	75.3	3681	3.00	2.26
CCH1 + CCH2	Mejillones	Coal	549.72	68.0	3277	2.95	2.01
CTA	Mejillones	Coal	177.00	65.4	1015	0.95	0.62
CC KELAR	Mejillones	NGas	532.46	39.2	1829	2.29	0.90
CC1 + CC2	Mejillones	NGas	780.60	1.9	127	3.36	0.06
TAL1 + TAL2	Taltal	NGas	240.15	2.6	55	1.03	0.03
Total					15,954	21.89	9.37

To illustrate the challenge of integrating MC plants, the tailing deposit locations, and the thermoelectric plants, Figure 5, constructed using the software Google Earth, presents the geographical location of the 19 thermoelectric plants and five copper tailing deposits. In the Antofagasta region, the thermoelectric plants are located on the coast (red markers), 12 of which are located in a small area of the Mejillones Peninsula, whereas the tailing deposits are located in the coastal mountain (green markers). For tailing deposits, the distance to the nearest CO<sub>2</sub> source and the altitude were defined based on topography data from Google Earth [48]. The location of the MC plant should be closest to one of the tailing deposits since transporting the CO<sub>2</sub> to the mine site is preferable compared to transporting large amounts of ore to the CO<sub>2</sub> source site [38]. CO<sub>2</sub> transportation from the thermoelectric plants on the coast to the tailing deposits, where the MC plant should be constructed, could be performed by pipelines through a CO<sub>2</sub> distribution network. CO<sub>2</sub> pipelines could be installed close to water pipelines, which exist in this area and that send desalinated water from the reverse osmosis plants on the coast to the mining sites, in order to prorate the right of way cost and favor legal and engineering advantages. Pipeline transport is considered the best option when it comes to transporting CO<sub>2</sub> over long distances and in large volumes [63]. Particularly, in the U.S. over 6000 km of pipelines exist, intended for the CO<sub>2</sub> supply for enhanced oil recovery projects [64]. Pipeline transport is a mature technology, which is applied on a wide scale across the world. However, the cost of transporting CO<sub>2</sub> can play an important role considering the distances and the difference in level between the thermoelectric plants and the tailings deposits. The risks of this activity for the environment and population largely depend on local conditions, population density, and other risky activities that are carried out close to the pipelines. However, the mitigating of these risks is part of the existing knowledge [64]. Regarding CO<sub>2</sub> capture, aqueous monoethanolamine (MEA) is the most common absorbent used to CO<sub>2</sub> separation, due to its good rates of CO<sub>2</sub> mass transfer, is suitable for low CO<sub>2</sub> partial pressure conditions, is biodegradable and low cost [49]. Karimi et al. [65] determined the cost of CO<sub>2</sub> avoided is about 55.7 USD/t of CO<sub>2</sub> for a 150 MW post-combustion power plant using MEA. On the other hand, Valderrama et al. [66] indicated that ionic liquids represent an attractive form of capturing and transporting CO<sub>2</sub> for its sequestration in mine tailings. Additionally, Mena and Guirado [67] determined that the use of copper electrodes and ionic liquids to capture and transform CO<sub>2</sub> into value-added products is a suitable and greener alternative compared to other CCUS strategies. Regarding CO<sub>2</sub> sequestration, the use of mine tailings for MC eliminates the cost of mining, crushing, and

milling associated with primary mining, considerably reducing energy consumption and thus CO<sub>2</sub> emissions [5]. Moreover, it is possible to reduce the main cost of the ex-situ MC, when sharing the comminution cost with the flow sheet of valuable metal recovery. The whole process may be profitable through the valuable metal recovery, as well as carbon taxes. For example, the copper Chilean mining generates 3.0 tons of CO<sub>2eq</sub> per ton of copper produced, a value above the world average which is 2.6 [68], and also has a production cost of 217 cUSD/lb of Cu. So considering a market value of 296 cUSD/lb of Cu, a cost for CO<sub>2</sub> transport and storage of 228 USD/t of CO<sub>2</sub> captured [69], and 50% extra in the cost of production, due to the difficulty of extracting copper from tailings, the value of CO<sub>2</sub> taxes to be profitable in the whole process should be about USD 12 and 24 per ton of CO<sub>2eq</sub> for the Esperanza and Sierra Gorda deposits, respectively. Meanwhile, in China, Denmark, and the Netherlands, the value of CO<sub>2</sub> taxes is about USD 1.43, 14.3, and 25 per ton of CO<sub>2eq</sub>, respectively [70]. These values are a preliminary economic assessment that allows an analysis of the potential viability of a tailing project. After this assessment, a pre-feasibility study is necessary. This pre-feasibility study must look at the mineralogy, engineering, and economic factors, with a higher detail and precision level. The conclusions can change because there are uncertainties in the current analysis. For example, more negative conclusions will result if the assessment includes recoveries below 100% for copper and higher costs. On the contrary, a more optimistic position will be found if a higher copper price is considered (the current price is over 400 cUSD/lb Cu with which positive revenues are obtained), recovery of REE or TiO<sub>2</sub> is considered (positive revenues are obtained in this case), and reduction of the costs. If the milling costs are subtracted from the considered costs, the CO<sub>2</sub> taxes go down to USD 7 and 13 per ton of CO<sub>2eq</sub> for the Esperanza and Sierra Gorda deposits, respectively. Additionally, the copper industry faces a paradox in that copper is key to the overall global low carbon renewable energy challenge, but the supply chain delivering copper still has a high carbon footprint. Then, several copper producers seek to reduce the levels of CO<sub>2</sub> generation in order to differentiate their product from the conventional one to add value to their products. In addition, there is the possibility of obtaining commercial by-products as material for construction, road material, additives for cement, among others, which could reduce the total cost of CO<sub>2</sub> sequestration and reduce the amount of material that ends up in the tailing deposits [10,71]. However, since it is a mining operation, the potential contamination of soil, water, and air in surrounding areas exists, whereby the implementation of this technology must have a suitable protocol of management and control environment. All these topics are beyond the scope of this investigation, which is focused on determining the most suitable site for the installation of the MC plant.

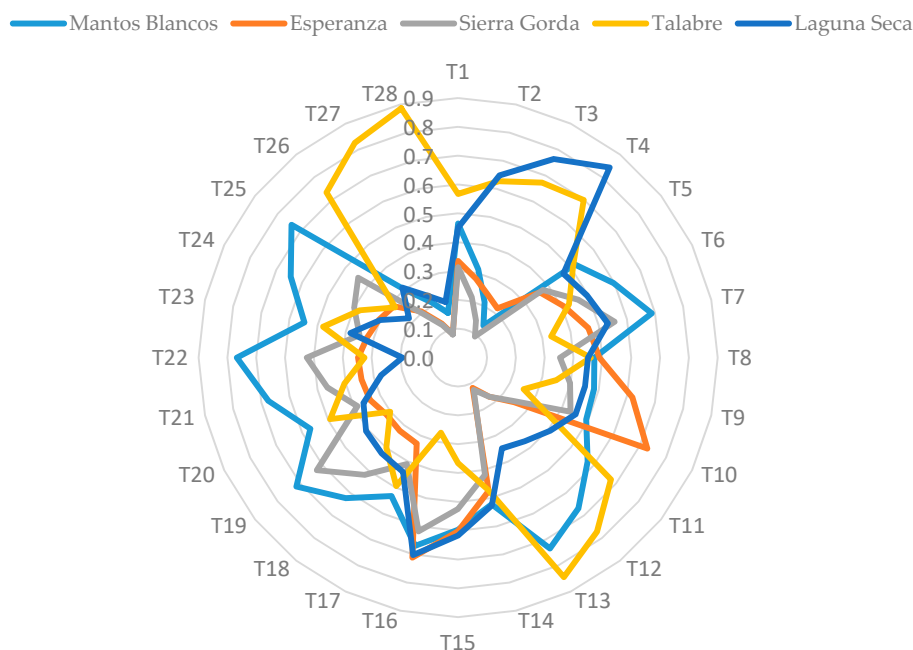
Regarding the TOPSIS method application, the following criteria were considered: (1) the average total amount of CO<sub>2</sub> that is able to be sequestered by the tailing, measured in Mt of CO<sub>2</sub> (C1); (2) income potential generated by the tailing in its processing, considering the sale of copper, titanium oxide, and REE, measured in USD/t of tailing (C2); (3) sequestration efficiency of the tailing, measured in t of CO<sub>2</sub>/t of tailing (C3); (4) copper grade, measured in % (C4); (5) titanium oxide grade, measured in % (C5); (6) rare earth grade, measured in % (C6); (7) altitude at which the tailings deposit is located, measured in meters above sea level (m a.s.l.) (C7); (8) distance to the nearest CO<sub>2</sub> source, measured in km (C8); and (9) potential ecological risk index, which is dimensionless (C9). Criteria C1 to C6 and C9 need to be maximized, whereas C7 and C8 need to be minimized. The  $CC_a$  obtained using TOPSIS were 0.46 for the Mantos Blanco (MB) deposit, 0.34 for ES, 0.31 for SG, 0.56 for Talabre (TA), and 0.45 for the Laguna Seca (LS) deposit. Details in the TOPSIS method results can be found in the Supplementary Materials. In the first instance (test T1), the highest value was obtained by the TA deposit. However, changes that may occur when the criteria weights change should also be analyzed for making the final decision.



**Figure 5.** Location of thermoelectric plants and tailing deposits in the Antofagasta region. TA: Talabre tailing deposit; SG: Sierra Gorda tailing deposit; ES: Esperanza tailing deposit; MB: Mantos Blanco tailing deposit; LS: Laguna Seca tailing deposit; m.a.s.l: meters above sea level.

### 6. Sensitivity Analysis

The results for the closeness coefficient ( $CC_a$ ) of several tests are presented in Table S8 and were obtained by varying the criteria weights. Tests T2 to T4 consider the variation of C1 with increments of 25.0%, 37.5% and 50.0% in its value. The results showed that the method is more sensitive to criteria C4 and C5 since they generate the largest variation in the numerical results and consequently in the ranking order. However, the most critical criteria are those related to altitude (C7) and to distance (C8). In the case of liquid transport by pipelines, the difference in costs between the pumps for transporting water and carbon dioxide is negligible [72]. In the case of water transport, Zhou and Tol [73] determined that 100 m of vertical water transporting consumes the same energy as 100 km of horizontal water transporting. This energy consumption would considerably reduce the global efficiency of sequestration due to the CO<sub>2</sub> generated by the transportation of CO<sub>2</sub> from the thermoelectric plant to the tailing deposits. The deposit with the optimal values for these criteria is MB. The MB deposit was most frequently ranked first, at 43% of the times, whereas the Sierra Gorda (SG) deposit was most frequently ranked last at 36% of the times. The results obtained from the 28 tests are presented in Figure 6, in which the results are presented in unit area. As observed, MB covered 36.10% of the total area, whereas TA, LS, SG, and Esperanza (ES) covered 34.18%, 25.91%, 17.42%, and 16.96%, respectively. Therefore, the final decision is that the MB deposit can be selected as the most suitable site for the installation of a MC plant in Chile.



**Figure 6.** Main results obtained from the technique for order preference by similarity to ideal solution (TOPSIS) method application.

## 7. Final Comments

Carbon capture and storage is a key technology for mitigating CO<sub>2</sub> emissions of anthropogenic origin, providing a method to achieve the transition from the use of energy produced by the burning of fossil fuels to increased usage of renewable energies to limit global warming below a threshold of 2 °C, according to the existing treaties to which Chile is signatory, such as the Kyoto Protocol, Montreal Protocol, Basel Convention, the United Nations Framework Convention on Climate Change, Antarctic Treaty, among others. Chile is sensitive to the effects of climate change throughout the country, starting from the south with glacier melting and rising sea levels, following in the center with lower rainfall and snow precipitation in the high mountains, and ending in the north with a higher frequency of dry days and longer heatwaves. In addition, it is seen that Chile has a regulatory framework that still has serious weaknesses in caring for the environment, in the case of thermoelectric plants without an emission limit for certain compounds, including CO<sub>2</sub>, while in the case of tailings deposits with scarce mineralogical data and lack of background metal concentrations data of soils, due to the large number of these.

However, in the north of Chile, the thermoelectric plants focused on producing electric power to support the demand required by the large amounts of mining in the region strongly depend on the burning of fossil fuels. The use of mining tailings for the sequestration of CO<sub>2</sub> produced by the power plants is proposed as a feasible solution to meet Chile's aim of decarbonizing its energy matrix, as well as the generation of economic value through the recovery of valuable metals. This technology can be applied to sequester emissions from power generating plants and to emissions produced by other industries, such as the production of cement or lime, which occur in the same area. The sequestration could be completed using mining tailings as well as storing CO<sub>2</sub> in a large number of saline aquifers existing in the region.

## 8. Conclusions

Following the discussion presented in this paper and the results presented in the preceding sections, the following main conclusions can be drawn:



1. Mineral carbonation is a feasible solution for tackling the problem of the deposition of tailings produced by the large copper industry developed in the north of Chile, as well as a strategy to mitigate the CO<sub>2</sub> emissions produced by many power plants that supply electric energy to the mining industry.
2. According to the proposed methodology, the region of Antofagasta is considered the best location for the installation of a future mineral carbonation plant as close to 3600 Mt of mine tailings are found in five large active deposits of tailings, which together have the potential to sequester between 66.0 to 98.6 Mt of CO<sub>2</sub>. In addition, 19 thermoelectric plants are in operation, which in 2018 produced about 9.4 Mt of CO<sub>2</sub> that is available to be sequestered, with a maximum generation potential of 21.9 Mt of CO<sub>2eq</sub> per year.
3. The results obtained when applying the method TOPSIS, in addition to the sensitivity analysis, showed that the mineral carbonation plant should be located close to the deposit of tailings from Mantos Blancos (MB), as it is located the shortest distance from the CO<sub>2</sub> source and at a lower altitude. The MB deposit has the highest concentration of Cu and REE, which generates the highest amount of income potential per ton of tailing (124.35 USD/t of tailings).
4. The recovery of valuable species, such as Cu, TiO<sub>2</sub>, and REE, must occur during the pretreatment stage of the tailings and before the precipitation of carbonates stage.
5. The results obtained are preliminary and require technical and economic studies of the tailings selected as the most promising to sequester CO<sub>2</sub>.

**Supplementary Materials:** The following are available online at <https://www.mdpi.com/2075-163X/11/3/320/s1>, Sensitivity analysis results.

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## Abbreviations

### Symbols

$\alpha$	Alternative
$CC_a$	Closeness coefficient
$i$	Criteria
$r_{ia}$	Normalization value
$T$	Number of operation hours throughout the year [8760/year]
$V_i^-$	Anti-ideal value
$V_i^+$	Ideal value

$V_{ia}$	Value of the weighted normalized decision matrix
$w_i$	Criteria weight
wt. %	Percentage by weight
$x_{ia}$	Decision matrix
<b>Abbreviations</b>	
Cap	Installed capacity of power plants [MW]
CO <sub>2</sub>	Carbon dioxide
m a.s.l.	Meters above sea level
NGas	Natural Gas
NO <sub>x</sub>	Nitrogen oxides
SO <sub>2</sub>	Sulfur dioxide
<b>Acronyms</b>	
ANG1, ANG2	Angamos thermoelectric power plants
CC KELAR	Kelar thermoelectric power plant
CCH1, CCH2	Cochrane thermoelectric power plants
CC1, CC2	Atacama thermoelectric power plants
CCUS	Carbon, Capture, Use, and Storage
CNE	Energy National Commission
CTA	Andina thermoelectric power plant
CTH	Hornitos thermoelectric power plant
CTM1, CTM2, CTM3	Mejillones thermoelectric power plants
DS	Supreme Decree (from the Spanish “Decreto Supremo”)
EMF	Emission Factor
ES	Esperanza tailing deposit
GHGs	Greenhouse Gases
GWP	Global Warming Potential
LS	Laguna Seca tailing deposit
MB	Mantos Blanco tailing deposit
MC	Mineral Carbonation
MCDA	Multi-Criteria Decision Analysis
MEA	Monoethanolamine
MSE	Mean Sequestration Efficiency [Mt of CO <sub>2</sub> /Mt of tailing]
MT	Mine Tailing
NTO1, NTO2	Norgener thermoelectric power plants
PGE	Power Generation Efficiency [%]
PM2.5	Particulate Matter 2.5 [ $\mu\text{m}$ ]
REE	Rare earth element
RI	Risk Index
SEN	National Electricity System
SERNAGEOMIN	National Service of Geology and Mining
SD	Standard Deviation
SG	Sierra Gorda tailing deposit
SIC	Interconnected Central System
SING	Interconnected System of the Great Northern region
TA	Talabre tailing deposit
TAL1, TAL2	Taltal thermoelectric power plants
TOPSIS	Technique for Order Preference by Similarity to Ideal Solution
US	United States
U14, U15, U16	Tocopilla thermoelectric power plants
<b>Sup. and sub-indexes</b>	
eq.	equivalent
eq.max.	equivalent maximum
max	maximum
max.cap	maximum capacity
min	minimum
SEQ	sequestered
Total	Total tonnage
Val	valuable specie
2018	year 2018

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