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#### **Supplementary material 1**

## **Techno-economic insights and deployment prospects of permanent carbon dioxide sequestration in solid carbonates**

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## **Note 1: In-situ mineralisation**

#### **General information**

A general estimation of the  $CO<sub>2</sub>$  sequestration capacity is using Eq. (1).

$$
G_{CO2} = A \cdot h \cdot \varphi \cdot E_{CO2}
$$

(1)

Wherein *GCO2* is the total sequestration potential, *A* is the area of the rock formation, h is the effective height of the formation,  $\varphi$  is the average porosity, and  $E_{CO2}$  is the storage efficiency. *ECO2* is defined as the CO<sup>2</sup> storage per pore volume and is generally 18.8- 48.7 kgCO $_2$ /m $^3$  [1] with the estimate provided by McGrail et al. [2] lying within that range. Oelkers et al. [3] summarise reported areas of 21 distinct onshore basaltic provinces as well as peridotite massifs and calculates the estimated  $CO<sub>2</sub>$  mineralisation potential therein using Eq. (1) with an estimated thickness of 500 m and a conservative *ECO<sup>2</sup>* of 10 kgCO<sub>2</sub>/m<sup>3</sup> and 30 kgCO<sub>2</sub>/m<sup>3</sup> for basalt and peridotite, respectively. Goldberg et al. [4] present a concise list of offshore  $MIN_{IN}$  sites with an overall expected total storage potential of 8238 - 41,191 GtC (29,657 - 148,288 GtCO2). Injection rates of about 0.3- 0.7 MtCO<sub>2</sub>/a per well are reported [5–7], whereas, at the Wallula basalt pilot project, only 14.6 ktCO<sub>2</sub>/a were injected per well [8]. In this study, an area demand of injection wells of about 28 km<sup>2</sup> per 1 MtCO<sub>2</sub>/a well array is assumed based on analyses of subsurface  $CO<sub>2</sub>$ plumes' radiuses in a recent study [9] and is in line with CO<sup>2</sup> storage models [8]. The specific area demand for MIN<sub>IN</sub> is assumed to be 28 km²/(MtCO $_2$ ⋅a) in this study. Kelemen et al. [10] assumed an effective area demand of  $62,500$  m<sup>2</sup> per well, capable of injecting 1-10 ktCO<sub>2</sub>/a, resulting in 6.25-62.5 km<sup>2</sup>/(MtCO<sub>2</sub>·a) for a hybrid DACCS and MIN<sub>IN</sub> site sequestering atmospheric  $CO<sub>2</sub>$  in peridotite. All MIN<sub>IN</sub> storage sites as well as their cumulative and annual potential are listed in the supplementary information 2. Because of significant uncertainties regarding the economic sequestration potential, a conservative case estimating only 10% of the theoretical storage potential to be technically and economically feasible is assumed. This assumption is yet to be confirmed by industrialscale MIN<sub>IN</sub> projects and bears therefore significant uncertainty.

#### **Cost assumptions**

Since data on economic parameters of MIN<sub>IN</sub> are still scarce, analogies from  $CO<sub>2</sub>$  injection in other subsurficial CO<sup>2</sup> sequestration sites must be employed. An average injection rate of 0.7 MtCO<sub>2</sub>/a is assumed at an average investment cost of 75 m€ per well [7] resulting in a CAPEX of 108.6 €/tCO2/a. For the CarbFix projected, cost of MIN<sub>IN</sub> of 20-30 USD/tCO<sup>2</sup> for an injection rate of 10-20 ktCO2/a are expected [11,12]. Kelemen et al.  $[10]$  note that MIN<sub>IN</sub> costs, based on cost estimations by Gunnarsson et al.  $[13]$ , 1040 USD/tCO<sup>2</sup> more compared to underground storage via injection of supercritical CO<sup>2</sup> in geological formations.

#### **Energy demand**

To compress CO<sub>2</sub> dissolved in freshwater for subsurface injection into suitable MIN<sub>IN</sub> sites, about 70 kWhel/tCO<sub>2</sub> are required [14]. The electricity demand is therefore mainly due to CO<sup>2</sup> compression and no heat demand is considered.

# **Note 2: Ex-situ mineralisation**

#### **General information**

Myers and Nakagaki [15] conducted a regional study on MINEX and concluded that Japan alone can achieve CDR at the rate of up to 7.6 GtCO $_2$ /a. Slag-based MIN<sub>EX</sub> is expected to enable cumulative MIN $_{EX}$  of 26.4-41.9 GtCO<sub>2</sub> between 2020 and 2100 [16]. Steel slag's high CaO and MgO content of about 37%<sub>wt</sub> and 9.1%<sub>wt</sub>, respectively, and the resulting weathering potential of around 384.7 kgCO<sub>2</sub>/t of slag, make it a valuable feedstock for MIN<sub>EX</sub>, with an expected global potential of 320-870 MtCO<sub>2</sub>/a in 2100 [17]. Renforth [17] notes that about 185 t of blast furnace slag and 117 t of steel slag are produced per tonne of crude steel. Production of one tonne of aluminium produces 3.45 t of bauxite residues, that can neutralise 44-66 kgCO<sub>2</sub>/t of bauxite residues [17]. About 115 kg of cement kiln dust are produced per tonne of cement clinker [17]. All these industrial solid wastes are suitable feedstocks for ex-situ  $CO<sub>2</sub>$  mineralisation. Pan et al. [18] also emphasise the potential for additional indirectly avoided CO<sup>2</sup> emissions by utilising carbonates as filler material in concrete blocks or cement mortars.

## **Cost assumptions**

Strunge et al. [19] examine a business case in the cement industry for  $MINEX$  using an integrated techno-economic assessment finding that  $CO<sub>2</sub>$  emissions can be reduced by partially substituting cement and filler material for concrete with silica and produced carbonates, respectively  $[20-22]$ . The opportunities of MIN $_{EX}$  for the cement industry, a hard-to-abate  $CO<sub>2</sub>$  emitter, was also studied by Ostovari et al. [23]. In a subsequent study, Ostovari et al. [24] modelled possible supply chains for the required feedstock, CO<sup>2</sup> source, source of renewable energy, and markets for the carbonates in the European context. Faber et al. [25] adapted learning rates of 10.55% to project future CAPEX of MINEX plants, based on estimations by Rubin et al. [26].

Gerdemann et al. [27] assumed a carbonation cost of 78-537 USD/tCO<sup>2</sup> depending on different feedstock, pre-treatment methods and regions in 2007. In 2013, Olajire et al. [28] assumed total cost of mineral carbonation of about  $105$  USD/tCO<sub>2</sub> avoided, while Geerlings and Zevenhoven [29] found a cost range of 15-100 USD/tCO<sub>2</sub> depending on the solid product value.

For feedstock rock mining, Beerling et al. [30] assumed a CAPEX of 6.0 €(tRock/a) and an OPEXfix of 4.6 €/tRock for an open-pit mine that has an ore output of 10,000 t/d and an economic lifetime of 10 years. The CAPEX estimates align with Goll et al. [31], who stated a CAPEX for open-pit mining of 1.8-7.1 €/tRock but a significantly higher OPEXfix of 12.7-27.9 €/tRock. Kelemen et al. [10] state costs for quarrying, crushing, and grinding of mine tailings at ~8.3 €/tRock. Strefler et al. [32] made an best estimate of 4.4 €/tRock for the CAPEX and of 22.2  $\epsilon$ /tRock for the OPEXfix of rock mining and grinding, ore processing, waste rock handling, and infrastructure development. The rock transportation cost are assumed to be  $4.4 \in H(\text{Rock-100 km})$  [32].

#### **Energy demand**

If energy demand is not further specified into electricity or heat, no such specification was provided by the respective reference.

Goll et al. [31] also assumed electricity demand of 27.8-83.3 kWh/tRock for rock mining and crushing, as well as 19.2-169.3 kWhel/tRock for grinding rock to 20 μm. Strefler et al. [32] gave a best estimate on the electricity demand of 19.4 kWhel/tRock, 55.6 kWhel/tRock, 127.8 kWhel/tRock, and 833.4 kWhel/tRock for rock grinding to 50 μm, 20 μm, 10 μm, and 2 μm, respectively. Also, an electricity demand of 2.8-8.3 kWhel/tRock for mining and crushing is assumed [32]. The practical minimum energy demand for rock mining including extraction and material handling is stated to be around 5.1 kWh/tRock [33]. Teir et al. [34] expect that for MINEX using serpentine  $44.4$ -52.8 kWhel/tCO<sub>2</sub> for grinding the rock to <74 μm are required.

Gerdemann et al. [27] found a total pre-treatment energy demand of 13-376 kWh/tRock, resulting in different mineralisation performances, which leads to a total energy demand of 429-2431 kWh/tCO2. Geerlings and Zevenhoven [29] state a range in energy demand of 694-2,777 kWh/tCO2, while Veetil and Hitch [35] assume a total energy demand of 470- 640 kWh/tCO<sub>2</sub> for MIN<sub>EX</sub>. Wang et al [36] states that MIN<sub>EX</sub> has a total energy demand including pre-treatment of 600-1200 kWh/tCO<sub>2</sub> depending on the type of feedstock, while it was stated that MIN<sub>EX</sub> using olivine requires  $447$  kWh/tCO<sub>2</sub> [37]. Zevenhoven et al. [38] simulated diverse MIN $_{EX}$  routes using serpentine and found an exergy demand of 720 kWhth/tCO<sub>2</sub> and 247 kWhel/tCO<sub>2</sub> for dry carbonation and 4,277 kWhth/tCO<sub>2</sub> for wet carbonation of CO<sup>2</sup> from flue gas of a lime kiln. Huijgen et al. [39] find a total electricity demand 403 kWhel/tCO2, 253 kWhel/tCO<sup>2</sup> thereof for grinding the feedstock and a total heat demand of 47 kWhth/tCO<sub>2</sub> for wollastonite in direct aqueous MIN $_{\text{EX}}$ . When steel slag was used as a feedstock, the electricity demand was 400 kWhel/ $tCO<sub>2</sub>$  and the heat demand was 354 kWht<sub>h</sub>/tCO<sub>2</sub> [39]. The feedstock material was ground to a particle size  $\leq$ 38 µm and the aqueous MIN<sub>EX</sub> was conducted at 200°C and 20 bara partial CO<sub>2</sub> pressure. Ostovari et al. [24] optimised a MINEX value chain on the European level by employing techno-economic parameters for three  $MINEx$  process configurations first introduced by Ostovari et al. [40]. Mineralising 1 tonne of CO<sub>2</sub> using 2 tonne of olivine in a reactor at 100 bar requires 103 kWh<sub>th</sub> and 689 kWh<sub>el</sub> [24,40,41]. When 2.55 tonne of serpentine are used to mineralise 1 tonne  $CO<sub>2</sub>$  at 115 bar 452 kWh<sub>th</sub> and 455 kWh<sub>el</sub> are required and for mineralising 1 tonne CO<sub>2</sub> using 4.4 tonne steel slag 407 kWhth and 592 kWhel are needed [24,27,39,40]. To derive these energy balances, Ostovari et al. [40] employed process simulations and assumed that 80% of sensible heat in treated feedstock can be recovered.

# **Note 3: Enhanced weathering**

## **Cost assumptions**

Strefler et al. [32] conclude that EW is a viable CDR option potentially capable of removing 95 GtCO<sub>2</sub>/a at cost of 50  $\epsilon$ /tCO<sub>2</sub> (60 USD/tCO<sub>2</sub>) and 4.9 GtCO<sub>2</sub>/a at cost of 167  $\epsilon$ /tCO<sub>2</sub> (200 USD/tCO2) for dunite and basalt, respectively. Beerling et al. [30] conclude that 0.5- 2.0 GtCO<sub>2</sub>/a can be removed from the atmosphere at cost of 67-150  $\epsilon$ /tCO<sub>2</sub> (80-180 USD/tCO2). Employing a land surface model simulating the effect of phosphorus release on ecosystem carbon sequestration, Goll et al. [31] find that cost of CDR on global hinterland alone through EW for removing  $0.2$ -2.5 GtCO<sub>2</sub>/a is 83-417  $\epsilon$ /tCO<sub>2</sub> (100- $500$  USD/ $tCO<sub>2</sub>$ ). The discrepancy to above mentioned studies is explained by the higher application cost on remote hinterland compared to agricultural land [31].

## **Energy demand**

Rock handling, i.e., mining, crushing and grinding for EW is assumed to be similar to the rock handling for feedstock preparation for  $MINEx$  (cf. sub-section 3.1.4 of the main study). However, while MINEX requires additional energy for reactor operation and thermal feedstock pre-treatment, EW's energy demand beyond feedstock preparation is limited to the requirements for rock transportation and spreading. In this work, energy demand for rock spreading is assumed to be negligible compared to the energy demand for long distance transportation.

## **Accounting for soil pH**

Soil pH affects the weathering rates of applied rock [32]. Figure 1 shows the average soil pH level aggregated for the nine major regions considered within the present study. The original data contains information about the soil pH level in 0.05-degree spatial resolution. As can be seen, global average data in the major regions ranges from below 5.7 to above 7.5. A simple, non-weighted, average is chosen, as the effect of soil characteristics on the efficacy of EW should be studied in high spatial resolution in future work. As elaborated by Strefler et al. [32], this can lead to significant uncertainty in the weathering rates of dunite and basalt.



Figure 1: Average soil pH in water in 0.05-degree spatial resolution (top) and averaged for nine major regions (bottom). Please note the different ranges of the colourbar for better visualisation. Data is taken from [42].

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