Assessing Carbon Capture and Carbonation in Recycled Concrete Aggregates: A Holistic Life Cycle Assessment Perspective with Simulation at Industrial Scale

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ABSTRACT

This work explores a life cycle assessment (LCA) concerning carbon capture and carbonation of recycled concrete aggregates (RCA), addressing the growing challenges posed by increased carbon emissions in power generation and ecological concerns tied to construction waste. We propose an integrated solution involving the capture of low-concentration carbon dioxide (CO_2) from NGCC power plants through aqueous ammonia absorption and sequestration by mineralization of RCA. The cradle-to-gate LCA focuses on an industrial-scale scenario for capturing 200 kilotonnes of CO_2 annually, with functional unit per tonne of carbonated RCA output. The findings reveal a net positive carbon abatement of 21.42 kg CO_2 eq., with heat production and electricity consumption being the major contributors to global warming potential (GWP). Within the spectrum of environmental impact categories under study, Marine Aquatic Ecotoxicity Potential (MAETP) exhibits a significant impact, primarily influenced by sulfur production. Human Toxicity Potential (HTP) follows as the second-worst contributor, with 96.80% of its impact attributed to heat and sulfur production. Subsequently, Acidification

Potential (AP) follows, arising from sulfur and sulfuric acid production. Sensitivity analysis and Monte Carlo simulation introduce a ±20% standard deviation to electricity and thermal energy consumption. Scenario analysis demonstrates a net carbon-positive abatement potential of 286.66 kg CO_2 eq. per tonne of CO_2 input. Employing carbon mineralization for CO_2 capture not only reduces emissions but also repurposes waste materials, providing a comprehensive and sustainable solution for effective CO_2 management, contributing to reduced sand mining, and offering cost advantages through the production of carbonated RCA.

1. Introduction

A collaborative effort among nations to mitigate the impacts of climate change, recognizing the pressing need to address imminent environmental challenges, is evident globally. Despite a lower average annual growth rate in the past decade (2010-2019) compared to the previous one, achieving the goal of limiting warming to 1.5°C by 2030 poses a formidable challenge [1]. This necessitates a concerted effort to transition to sustainable energy systems and achieve net-zero emissions by mid-century. Extensive research into carbon capture and storage (CCS) technology underscores its potential to significantly contribute to these climate change goals [2, 3]. CCS deployment offers solutions to various challenges associated with low-carbon energy, including emissions reduction in industries, addressing land availability issues, navigating siting restrictions, and enhancing social acceptance [4, 5]. Singapore, within this framework, has committed to reducing carbon emissions by 36% from 2005 levels by 2030 [6]. The nation pursues its climate objectives through sustainable development strategies and actively engages in research and development, particularly in technologies like carbon capture, utilization, and storage (CCUS).

CCUS, a multifaceted strategy designed to mitigate greenhouse gas emissions, primarily focuses on capturing carbon dioxide (CO_2) generated from industrial processes and power generation [7, 8]. This captured CO_2 can be repurposed for diverse applications, referred

to as Carbon Capture and Utilization (CCU), or securely stored underground to prevent its release into the atmosphere, known as CCS [8]. In the context of Singapore, the implementation of CCUS technologies is imperative due to the nation's substantial industrial emissions and the need for enhanced energy security in a resource-constrained setting [9].

Storage methods for CO_2 can be categorized into two main approaches: in-situ and ex-situ processing methods [10, 11]. In-situ carbonation involves the direct injection of CO_2 into geological formations to facilitate mineralization [12]. Initially introduced by Seifert in 1990 [13, 14], geological sequestration has become one of the most mature methods, reaching a relatively high level of maturity (TRL 7-9) [15, 16] . However, concerns regarding potential environmental risks, including CO_2 leakage, high initial costs, energy requirements, and monitoring challenges, have prompted exploration into alternative methods [17-19].

Ex-situ carbonation occurs above ground, involving the capture of CO_2 from industrial sources and its reaction with mined rocks, such as industrial waste materials and mining tailings [20, 21]. Accelerated mineral carbonation (AMC) mimics natural weathering processes to convert CO_2 into stable carbonates by reacting it with metal oxides or silicates, such as serpentine, olivine, limestone, wollastonite, and forsterite. Despite effectively sequestering CO_2, challenges such as slow reaction rates, high energy requirements, and limitations associated with mining natural minerals persist [15, 22].

As a result, there is a growing interest in exploring alternative sources, including alkaline solid wastes such as cement waste, steel slag, and incinerator bottom ash (IBA). These waste materials offer faster reactions, lower energy input, and higher efficiency in converting carbonates, presenting a viable solution for managing disposed recycled concrete aggregates (RCA) and IBA [15, 23-25]. This approach is particularly relevant in countries facing land resource shortages, such as Singapore [26].

Singapore encounters various challenges in embracing CCUS. The scarcity of land poses difficulties in identifying suitable sites for large-scale CCUS projects [26]. Moreover, the energy-intensive nature of CCUS processes, particularly low concentration CO_2 capture and compression, becomes complex in a country with limited domestic energy resources [26].

Within the realm of CCUS, mineral carbonation emerges as a promising pathway. This process involves the reaction of CO_2 with minerals to create stable carbonates, providing permanent and secure storage for captured CO_2 [26]. The production of these valuable materials, suitable for the building and construction sectors, reduces reliance on imports [26]. Additionally, the prospect of forming stable carbonates from waste materials aligns with circular economy principles, making it increasingly significant in advancing effective carbon capture and sustainable resource utilization.

Numerous studies have investigated the effects of carbonation treatment on concrete properties using various compositions of recycled aggregates, including RCA and recycled concrete aggregates-fine aggregates (RCA-FA), and have compared them with conventional concrete [27, 28]. For example, Sereng et al. conducted a study on accelerated carbonation, demonstrating a reduction in RCA porosity and water absorption coefficient by up to 67% [29]. Similarly, Zadeh et al. showed strength improvement, with a decrease of over two percentage points in aggregate crushing value (ACV) compared to the original RCA [30].

However, concerns have been raised regarding potential diminishing benefits [31, 32]. Notably, while carbonated RCA can enhance properties such as workability, mechanical strength, and durability of mortars and concrete, there is apprehension about negative effects on mechanical properties, particularly for RCA-FA derived from parent concrete made with fine aggregates (FA) blended cement. Studies indicate that carbonated RCA-FA exhibits inferior property values compared to other recycled aggregates [33]. Moreover, the overall effectiveness of carbonation in sequestering CO_2 may be compromised due to limited

carbonation depth resulting from less porous RCA, potentially impacting the mechanical properties of carbonated RCA [34].

Conducting a comprehensive Life Cycle Assessment (LCA) on CCUS is imperative to gain a thorough understanding of its environmental impacts. LCA evaluates the entire life cycle of CCUS technologies, identifying environmental hotspots, potential trade-offs, and overall sustainability [35, 36]. This approach facilitates informed decision-making, by enabling comparisons with alternative carbon mitigation strategies and promoting transparency [35, 36]. Additionally, LCA supports the assessment of resource and energy efficiency, contributing to the optimization and enhanced sustainability of CCUS. As a crucial tool, LCA ensures the environmental sustainability of CCUS and aligns with the imperative to address climate change responsibly and effectively.

Digulla et al. performed a comparative LCA of CO_2 utilization using industrial wastes as feedstock [37]. They incorporated information on mineralization routes from six sources into their LCA models [38-43]. Table 2 in the study by Digulla et al. presents a summary of the mineralization pathways assessed, encompassing details on feedstock and Technology Readiness Level (TRL) information [37]. It is observed that five out of six processes yield negative net emission values (-0.06 to -0.14 CO_2 eq. kg^{-1} feed) when sand is considered as a product [37].

Introducing a mineral carbonation process, we present an innovative approach to capture CO_2 emissions from natural gas combined cycle (NGCC) power plants. The process integrates aqueous ammonia absorption of CO_2 and the sequestration through RCA carbonation. Notably, the CO_2 concentration in NGCC flue gas (3-5%v/v) is lower than in coal-fired power plants (12-15%v/v) [44, 45]. Higher specific heat duty is required for lower CO_2 content owing to the reduced partial pressure of CO_2 in the flue gas exiting from NGCC power plants, resulting in a slowed-down absorption process [44]. Despite these challenges, exploring

mineral carbonation has merits. The production of carbonated RCA repurposes waste materials for construction and building, offering an alternative to sand for land reclamation and reducing the need for sand mining [26]. Carbonated RCA not only exhibits enhanced physical and mechanical properties; it also addresses environmental concerns.

Santolini et al. found that using recycled stabilized cement in rural pavement construction reduced environmental impacts in 17 out of 18 categories compared to natural gravel aggregate, with the exception of marine eutrophication [46]. Younes et al. demonstrated that increasing the use of RCA lowers environmental impacts [47]. Specifically, when comparing a concrete mixture containing 0% RCA to one with 100% RCA, reductions of 96.7%, 93.6%, and 71.9% were observed for land use, freshwater eutrophication, and climate change impacts, respectively [47].

The amount of CO_2 uptake by RCA is influenced by several factors. Gao et al. investigated the effect of moisture content in the reactor on the degree of carbonation, finding that a semi-wet process achieved a carbonation degree of 15.5% after 160 minutes, compared to approximately 10% after 170 minutes for the conventional wet process [48]. This finding is supported by Andreas et al., who observed that CO2 uptake by an RCA sample doubled with lower moisture content [49]. The composition of RCA also significantly impacts CO2 uptake. Jean et al. showed that RCA containing a sand fraction captures twice as much CO2 as natural aggregates [50]. While most studies focus on a gas-liquid-solid reaction, our process involves a liquid-solid reaction, where CO2 is first absorbed by an aqueous ammonia solution to form a carbonated ammonia solution, which then reacts with the solid RCA.

Pu et al. reported that incorporating carbonated RCA could significantly reduce energy requirements, environmental impacts, and associated costs compared to natural aggregate concrete [51]. Accelerated carbonation treatment exhibited substantial potential in mitigating environmental burdens, particularly concerning global warming potential [51]. Xing et al.

showcased that recycled aggregate concrete exhibits the least environmental impact among the three concrete types with identical mix designs [52]. CO_2 concrete excelled in global warming potential, attributed to the carbonation process's dual effect of sequestering CO_2 within recycled aggregate and enhancing concrete's mechanical properties [52]. Moreover, CO_2 concrete demonstrated reduced environmental footprint in most scenarios compared to virgin aggregate concrete, while maintaining equivalent compressive strength [52].

Our study aims to reduce landfill dependence, safeguard Singapore's land resources, and enhance environmental sustainability amidst urbanization and industrialization, which have led to increased construction and demolition waste generation [53, 54]. Efficient reuse of RCA can mitigate these challenges by reducing carbon emissions and construction costs while promoting sustainability in the construction sector [55]. We propose integrating RCA carbonation with CO_2 capture using an ammonia aqueous solution to enhance RCA properties and reduce reliance on natural minerals for CO_2 storage.

The integrated process of carbon capture and carbonation was optimized by modeling various scenarios and conditions based on experimental results. This optimized model was used to conduct an LCA to evaluate the carbon abatement potential and environmental performance across various impact categories. Our LCA encompasses the entire process from NGCC power plant flue gas extraction to the production of one tonne of carbonated RCA. It identifies key factors influencing the results and employs sensitivity analysis and Monte Carlo simulation to scrutinize key assumptions. Additionally, a scenario analysis adjusts the functional unit to per tonne of CO_2 input, providing a comprehensive examination of the carbon abatement potential in our mineral carbonation process.

2. Materials and Methods

The process described in this study aims to capture 200 kilotonnes of CO_2 annually from a NGCC power plant. Aspen Plus® V14 software was utilized to determine the mass balance and energy consumption of the process [56]. The United States' National Energy Technology Laboratory (NETL) extensively employed Aspen Plus to evaluate the cost and performance of carbon capture case studies for both pulverised coal (PC) and NGCC power plants [57]. Previous works by Bonalumi et al. [58] and Li et al. [59] have also employed Aspen Plus for techno-economic analyses. Given the similarity of our process to these examples, Aspen Plus® V14 software was chosen for modelling purposes [56].

Aspen Plus® V14 represents a versatile software solution offering numerous advantages for process simulation and optimization [56]. The software provides robust modelling capabilities, enabling accurate simulation of complex chemical processes. It features an interface to access advanced thermodynamic models and property databases, facilitating users in selecting appropriate models to suit their needs. Seamless integration with other AspenTech products streamlines comprehensive process simulation and optimization workflows [56]. The software simplifies the construction of process flowsheets and modeling of various unit operations through an extensive library of pre-built unit operations. Additionally, Aspen Plus® V14 includes powerful optimization tools to facilitate the optimization of process designs, operating conditions, and product quality to meet specific targets.

Our process was scaled to represent a pilot-scale CO_2 capture plant [60], operating 8000 hours per year. It comprises two sections: the absorber section and the solvent recovery/drying section, illustrated in Figure 1. In the absorber section, flue gas from the power plant stack contacts an aqueous ammonia solution in a counter-current packed absorber (AB01), forming a carbonated ammonium solution (CAS) containing various ionic species and complexes involving ammonium carbonate, bicarbonate and carbamate. The interactions described are

simulated utilizing the Electrolyte Non-Random Two Liquid (ELECNRTL) model, as detailed by Que and Chen [61]. Rate-based methodologies are employed in the absorber section to forecast these interactions. Our process integrates a water wash tower (WT01) for the recovery of ammonia gas released from the top section of the absorber. Additionally, an acid wash tower (WT02) is included to treat ammonia with a concentration of 2000 ppm, ensuring compliance with local regulations (43.07 ppm; 30 mg/Nm3). According to case studies conducted by the National Energy Technology Laboratory (NETL) in the United States, flue gas emitted from an NGCC power plant typically contains CO_2 at a concentration of 3-4% [57]. To adopt a conservative approach, this work assumes a CO_2 concentration of 3 vol%. A lower concentration is deemed conservative due to the increased volume of flue gas generated with lower CO_2 concentrations, necessitating larger equipment and resulting in a greater environmental footprint for flue gas processing [62]. The composition of the flue gas is provided in Table S3 of the Supporting Information (SI). The absorber operates under conditions of 20 °C and ambient pressure [58], while the wash towers and reactor operate at 25 °C and ambient pressure. The chemical composition of the RCA is assessed through X-ray fluorescence (XRF) analysis (refer to page 3 of SI). As per the Thermal Gravimetric Analysis (TGA) findings, the RCA utilized in the reaction demonstrates a maximum carbonation capacity of 10% by mass (refer to Table S2 on page 4 of SI). Consequently, the simulation adopts the chemical composition of the RCA as outlined in Table S1 of the SI and a 10% CO_2 carbonation capacity.

In the solvent recovery/drying phase, the carbonated RCA undergoes filtration (F01) and is subjected to two-stage drying (D01, D02) to retrieve the ammonia solvent for utilization in the absorber and wash tower (WT01). Ammonia, being the more volatile component, is reclaimed in the initial drying stage (D01) and subsequently returned to the absorber, while water is recuperated in the subsequent drying stage (D02) for application in the wash tower.

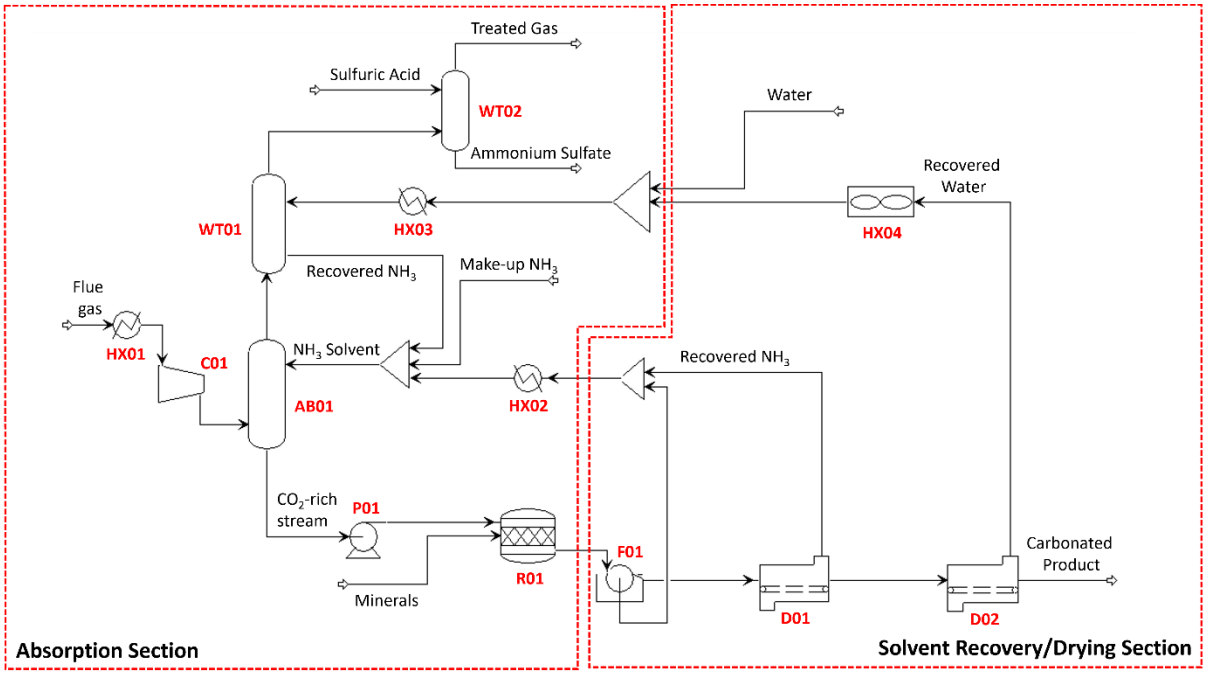

Figure 1. Schematic representation of our carbon capture process from cradle to gate, obtained using Aspen Plus® V14.

3. Life Cycle Assessment (LCA) Methodology

LCA methodology is executed following ISO 14040 [35], which offers fundamental principles and frameworks for conducting LCA. It is further supplemented by ISO 14044 [36], which outlines the requirements and guidelines. In our study, we adhered to these standards to ensure the robustness, comparability, and reliability of the LCA conducted. The LCA methodology involves a sequence of four steps: (1) defining the goal and scope, (2) conducting an inventory analysis, (3) performing an impact assessment, and (4) interpreting the results.

3.1.Goal and Scope Definition

The goal of the LCA is to assess the environmental impacts of the carbon capture process based on per tonne of carbonated RCA output. The scope, or the boundary of the LCA system, defines the extent of interactions between a process chain and the environment. Within this boundary, various stages of processing occur, resulting in the production of carbonated RCA. A "cradle-to-gate" system boundary is employed, as depicted in Figure 1, and it is modelled using Sphera® LCA for Experts (GaBi) version 10.7.1.28 software [63]. Detailed LCA model is available in the SI as Figure S1.

Before commencing an LCA study, it is imperative to establish the modelling parameters. Our process commences with the utilization of incineration flue gas from an NGCC power plant, and it concludes with the production of one tonne of carbonated RCA as the functional unit. The calculation of resource and energy consumption, along with their associated emissions, relies on mass allocation. The cut-off criterion is established at 0.10% of the reference flow, and all electricity requirements were met by the Singapore national electricity grid, which primarily consists of natural gas (94.3%), petroleum products (0.3%), coal (0.9%), and other sources (4.4%) [64]. This data reflects the electricity generation fuel mix for Singapore's electricity generation in June 2023. The 0.3% of petroleum products is

presumed to be divided between diesel (0.15%) and fuel oil (0.15%). The 4.4% from other sources is assumed to be entirely derived from waste-to-energy.

49.78% of the total heat production requirement, is recuperated from the flue gas cooler. This recovered thermal energy is then directed to fulfil a portion of the thermal energy demand for Dryers 1 and 2. The residual thermal energy necessary for the process is generated through an industrial furnace, which uses natural gas as the primary fossil fuel source.

The transportation and storage of chemicals in supply chain operations of our process are excluded from considerations for several reasons. First, supply chain operations have significant environmental impacts, and our paper aims to understand the true carbon uptake potential of our technology . Including transportation impacts could obscure this potential [65]. Secondly, our current work is at the simulation stage, and planning supply chain activities for chemicals is currently beyond the scope of this study. Additionally, Ang et al. have previously noted that the supply chain activities of chemical suppliers are not readily available for modeling needs [65].

Inventory Analysis

The life-cycle inventory (LCI) analysis process involves obtaining relevant input-output information or data related to the processes within the defined system boundary. A reliable LCI is crucial for achieving accurate LCA results. LCA modelling involves the essential selection of relevant data from Ecoinvent v3.9.1, as delineated in Table S7 of the SI [66]. Input-output data of our CO_2 mineralization technology are derived from the stream results generated by our simulation model in Aspen Plus® V14 [56]. Detailed information can be found in Tables S4 and S5 of the SI. The LCI analysis for the environmental profile of electricity generation in Singapore for June 2023 is established based on the methodology outlined in the study by Tan et al. [67]. The air emissions from cradle-to-gate for generating 1 MWh of electricity delivered

to the national grid, accounting for a 2.5% transmission loss, have been calculated and incorporated into our LCA model. Detailed specifics of this LCI are presented in Table S6 of the SI.

Impact Assessment

Numerous reputable life cycle impact assessment (LCIA) methodologies exist, such as CML 2001 – Aug. 2016, ReCiPe (Relevance, Endpoint, and Midpoint Assessment of Environmental Impacts), IMPACT 2002+, Eco-Indicator 99, ILCD (International Reference Life Cycle Data System), and TRACI (Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts). The CML 2001 methodology, developed by the Center of Environmental Science at Leiden University, is widely used in LCIA [68]. It assesses environmental implications concerning human health, ecosystem quality, and resource depletion throughout the life cycle of products or processes under investigation. Characterization factors are employed to quantify impacts based on their potential to harm human health, ecosystems, or resources [69]. ReCiPe integrates midpoint and endpoint methodologies, offering a holistic evaluation across multiple categories [70]. IMPACT 2002+ furnishes a scientifically robust framework and clear assessment guidelines, developed by the Swiss Federal Institute of Technology Lausanne [71]. Eco-Indicator 99 is optimal for health-focused studies due to its emphasis on human health impact assessment [72]. ILCD is esteemed for its credibility with standardized methodologies and guidelines endorsed by the European Commission [73]. TRACI, devised by the U.S. Environmental Protection Agency (EPA), specializes in assessing the environmental impacts of chemical emissions, particularly pertinent in sectors with substantial chemical usage [74]. In our study, we opted for CML 2001 – Aug. 2016 for impact assessment due to several reasons. Firstly, it is the predominant methodology in LCA studies for post-combustion CO_2 technologies [75-77]. Secondly, CML2001 – Aug. 2016 is primarily classified as a midpoint method, favoured for its simplified complexity, ease of quantification, data availability, scientific basis, consistency, and interpretability [68]. Further justifications for selecting CML2001 include its thorough assessment of environmental impacts, transparency, and clarity of methodology for facilitating

comprehension and interpretation of results [68]. It is a well-established, renowned methodology recognized for its rigour and comprehensiveness, and it is compatible with the Sphera® LCA for Experts (GaBi) version 10.7.1.28 software utilized in our research [63].

4. Results and Discussions

In this section, we assess and analyze the cradle-to-gate impacts per tonne of carbonated RCA output for global warming potential (kg CO_2 eq.), acidification potential (kg SO_2 eq.), eutrophication potential (kg PO_4 eq.), freshwater aquatic ecotoxicity potential (kg DCB eq.), human toxicity potential (kg DCB eq.), marine aquatic ecotoxicity (kg DCB eq.), photochemical ozone creation potential (kg C_2H_4 eq.), and terrestric ecotoxicity potential (kg DCB eq.). Abiotic Depletion Potential of Elements (ADPelements), Abiotic Depletion Potential of Fossil Fuels (ADPfossil fuels), and Ozone Layer Depletion (ODP) are excluded due to their absence from our CO_2 mineralization technology. Characterization factors are employed to transform the inventory analysis results into impact scores for the nominated eight environmental impact categories. These characterization factors sourced from the CML-IA database [69] have been integrated into the Sphera® LCA for Experts (GaBi) version 10.7.1.28 software [63] to facilitate a detailed and precise understanding of the relative environmental significance.

Global Warming Potential (GWP)

Referring to Figure 2, the greatest impact on Global Warming Potential (GWP) is attributed to heat production, accounting for 20.16 kg CO_2 eq. (50.85%). Subsequently, electricity consumption follows with 17.61 kg CO_2 eq. (44.41%), trailed by sulfur production at 1.82 kg CO_2 eq. (4.59%), and natural gas production and import from Indonesia (ID) at 5.98×10^{-2} kg CO_2 eq. (0.15%). According to our LCA results, the absorption and sequestration unit of our CO_2 mineralization technology can eliminate 61.07 kg CO_2 eq. The unit comprises an absorber and carbonation reactor. A total carbon abatement of 21.42 kg CO_2 eq. is achieved. In these unit processes, CO_2 and methane contribute to the kg CO_2 eq. The most significant contributor is heat production, primarily because CO_2 is the leading factor, accounting for 20.13 kg CO_2 eq.

Zhang et al. conducted a LCA study focusing on carbonation technologies applied to concrete debris across 14 diverse countries [78]. Their findings suggest that the net reduction in CO_2 emissions is relatively modest, with certain countries exhibiting a positive net emission, ranging from -4.3 to 15.7 kg CO_2 eq. per ton of RCA [78]. The authors underscore the critical importance of considering geographical factors when evaluating the feasibility of adopting mineral carbonation [78].

In a brief comparison between our study and that of Zhang et al., our technology demonstrates a higher total carbon abatement, indicating its potential as a CCUS technology [78]. However, it is important to note that our LCA did not include the RCA production process, as we utilize RCA obtained from a local construction company in Singapore. Furthermore, our study is situated in Singapore, which differs from the locations examined in Zhang et al [78]. Therefore, this comparison is preliminary and lacks precision due to variations in LCA scope.

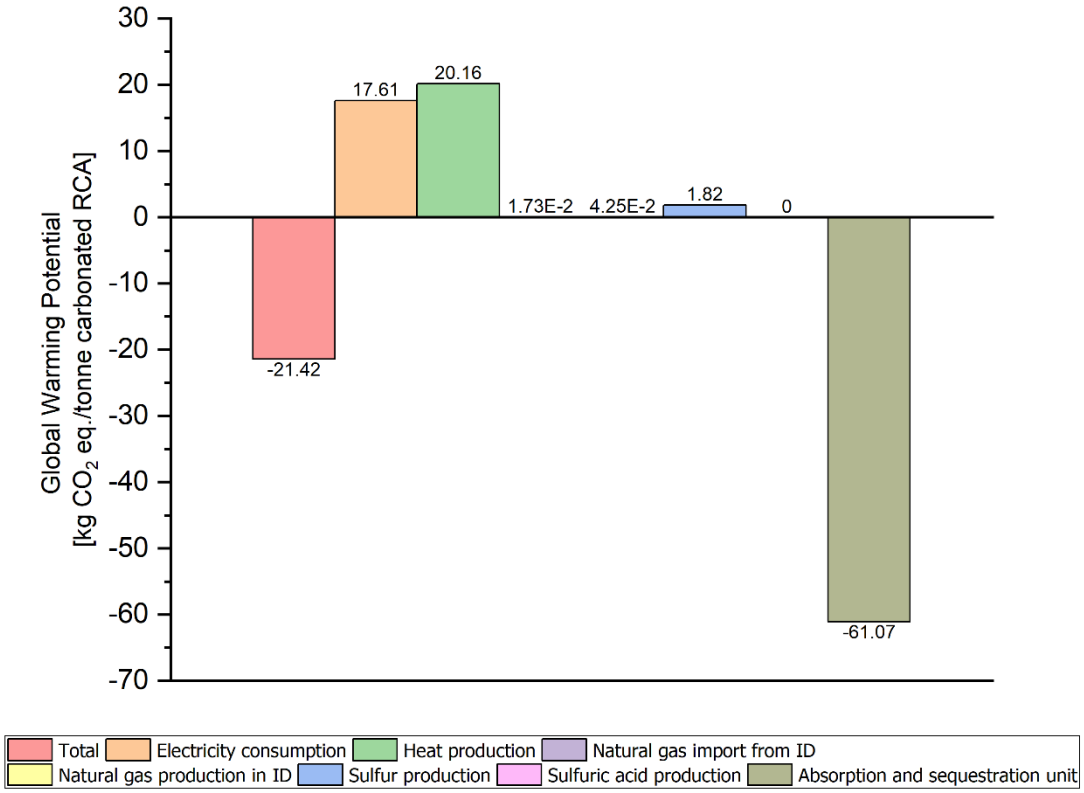

Figure 2. Global Warming Potential results for the production of one tonne carbonated RCA.

Acidification Potential (AP)

As depicted in Figure 3, the highest contribution to Acidification Potential (AP) comes from sulfuric acid production (0.35 kg SO_2 eq., 53.15%), with sulfur production (0.31 kg SO_2 eq., 46.85%) following as the next significant determinant. Contact Process is typically employed to produce sulfuric acid, with sulfur being one of the raw materials involved [79]. The necessity of sulfur and sulfuric acid production processes boils down to the requirement of an acid wash tower for lowering ammonia emissions to meet stringent discharge regulations [80]. Other contributions to AP comprise electricity consumption (1.99%), heat production (0.51%) and natural gas production in ID (0.01%). In this process, contaminants such as ammonia, nitrogen oxides, and sulfur dioxide can lead to AP. Sulfur dioxide is the primary air pollutant for AP in sulfuric acid production, sulfur production, heat production, and natural gas production and import from Indonesia. In contrast, nitrogen oxides constitute the primary contribution for AP in electricity production.

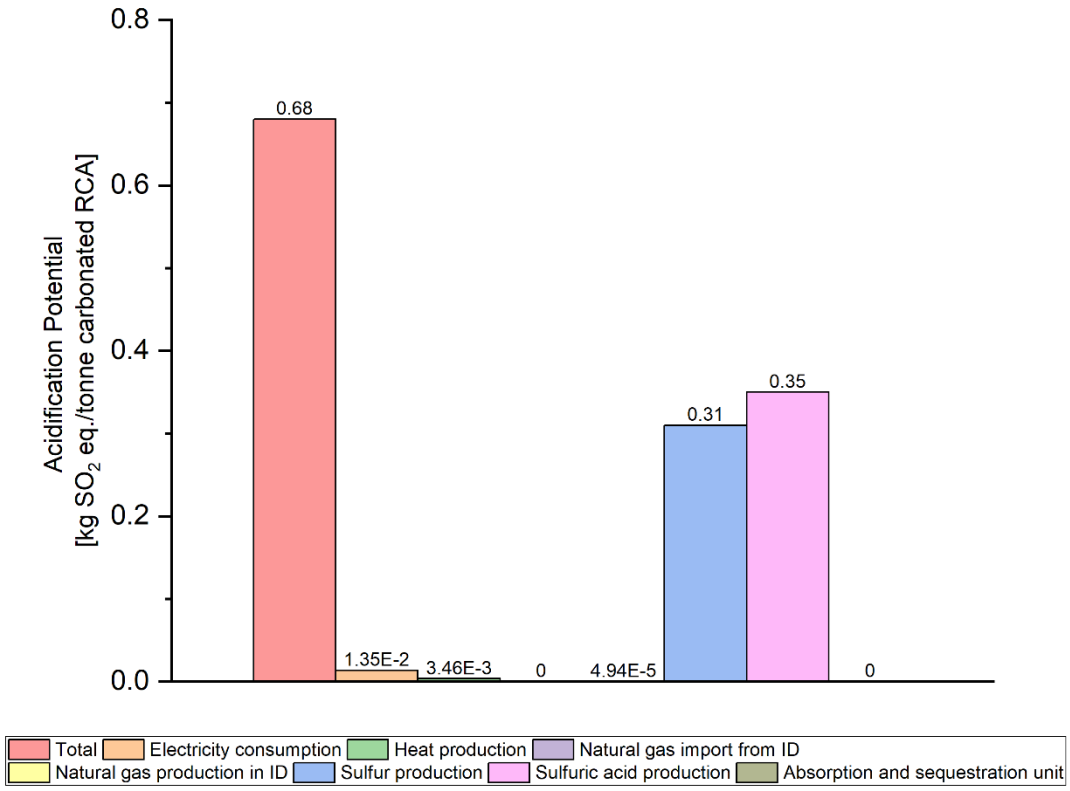

Figure 3. Acidification Potential results for the production of one tonne carbonated RCA.

Eutrophication Potential (EP)

According to Figure 4, the overall Eutrophication Potential (EP) contribution amounts to 4.16 × 10^{-3} kg PO_4 eq. Electricity consumption (3.02 × 10^{-3} kg PO_4 eq.) constitutes 72.48% of the EP contribution. Other notable contributors encompass heat production (8.46 × 10^{-4} kg PO_4 eq., 20.30%) and sulfur production (2.87 × 10^{-4} kg PO_4 eq., 6.90%). Air emissions in our process, such as ammonia, nitrogen oxides, and dinitrogen oxide, contribute to EP. EP is induced by chemical oxygen demand (COD), nitrate, nitrite, nitrogen, phosphorus, and ammonium. Nitrogen oxides constitute the key air pollutants contributing to EP in all categories, excluding natural gas import from Indonesia and the absorption and sequestration unit, as outlined in Figure 4.

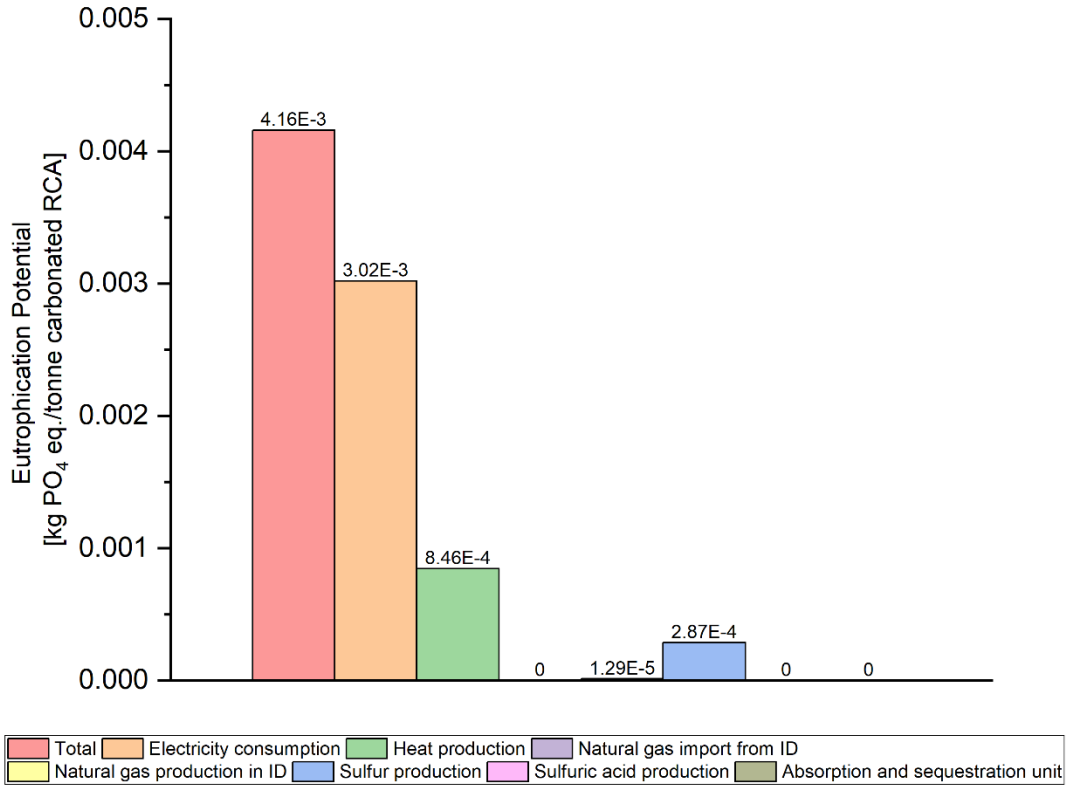

Figure 4. Eutrophication Potential results for the production of one tonne carbonated RCA.

Freshwater Aquatic Ecotoxicity Potential (FAETP)

The major portion of Freshwater Aquatic Ecotoxicity Potential (FAETP) is ascribed to sulfur production (3.67×10^{-2} kg DCB eq., 98.57%), as indicated in Figure 5. Other sources include heat production (5.25×10^{-4} kg DCB eq., 1.41%) and electricity consumption (7.03×10^{-6} kg DCB eq., 0.02%). The predominant factor contributing to FAETP is heavy metals. In sulfur production, the top three heavy metals released into the air are vanadium (2.92×10^{-2} kg DCB eq., 79.60%), nickel (6.33×10^{-3} kg DCB eq., 17.27%) and cobalt (3.01×10^{-4} kg DCB eq., 0.82%). In heat production, the major component is formaldehyde, a nonaromatic aldehyde.

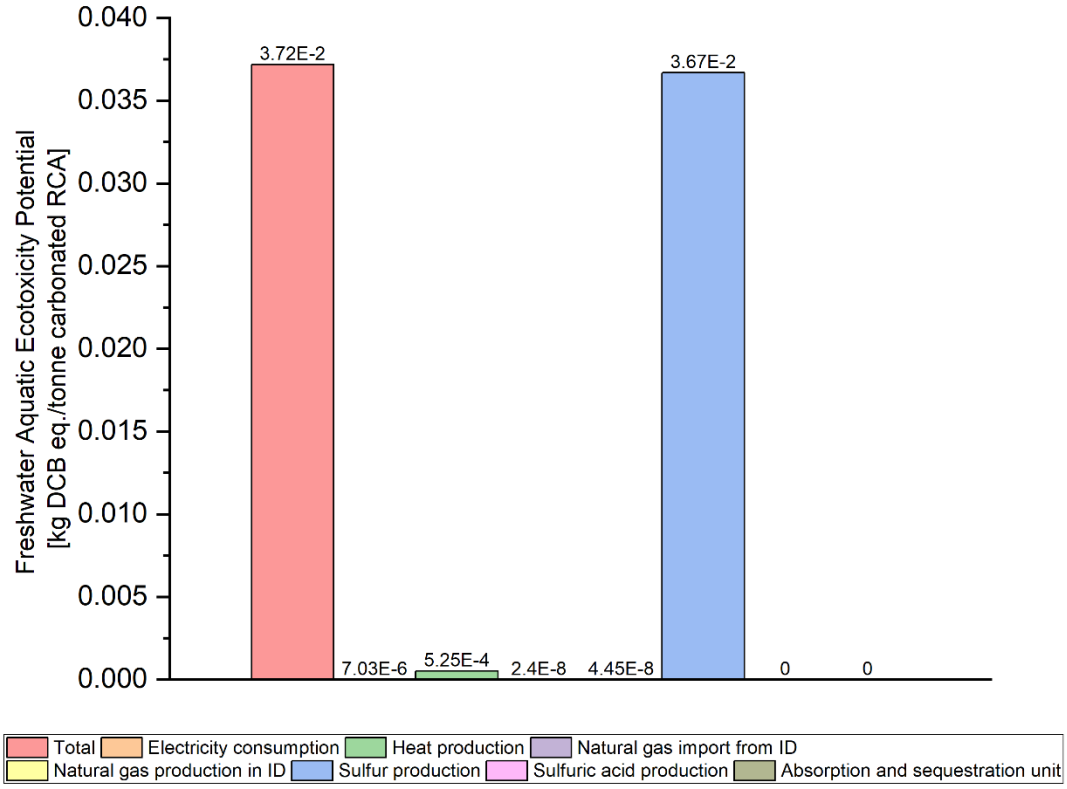

Figure 5. Freshwater Aquatic Ecotoxicity Potential results for the production of one tonne carbonated RCA.

Human Toxicity Potential (HTP)

Illustrated in Figure 6, Human Toxicity Potential (HTP) (1.73 kg DCB eq.) is derived from heat production (57.75%), sulfur production (39.05%), electricity consumption (1.62%), sulfuric acid production (1.57%) and natural gas production in ID (0.01%). Benzene, mercury, formaldehyde, toluene, and dioxins are instances of substances contributing to HTP in heat production. While heavy metals like nickel, vanadium, arsenic, cadmium, thallium, and cobalt collectively account for a significant contribution of 87.78% to sulfur production.

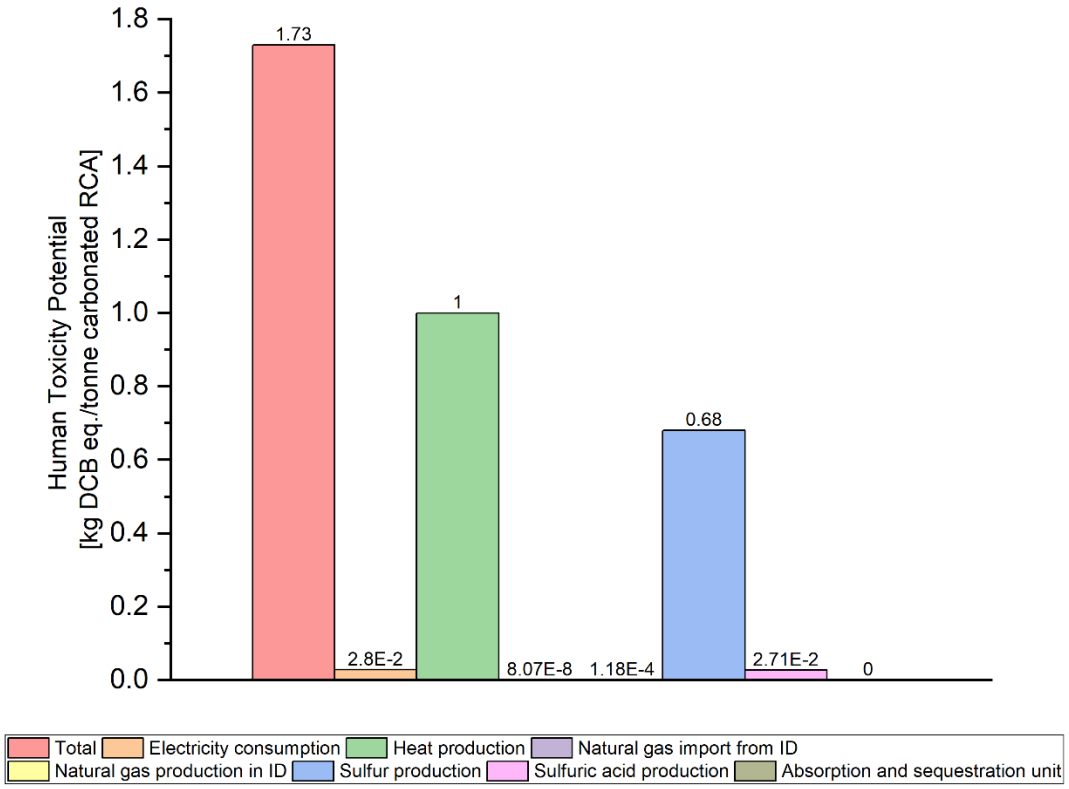

Figure 6. Human Toxicity Potential results for the production of one tonne carbonated RCA.

Marine Aquatic Ecotoxicity Potential (MAETP)

As shown in Figure 7, sulfur production contributes to nearly 100% of Marine Aquatic Ecotoxicity Potential (MAETP). Other factors such as heat production, natural gas production and import from ID, and electricity consumption have negligible impact on MAETP. In the context of sulfur production, vanadium is discharged into the air, making up 55.58% of MAETP. Subsequently, the inorganic release of hydrogen fluoride into the air adds 30.56% to MAETP. The third participant is the air emission of nickel (37.82 kg DCB eq.). In heat production, mercury is the predominant element driving MAETP.

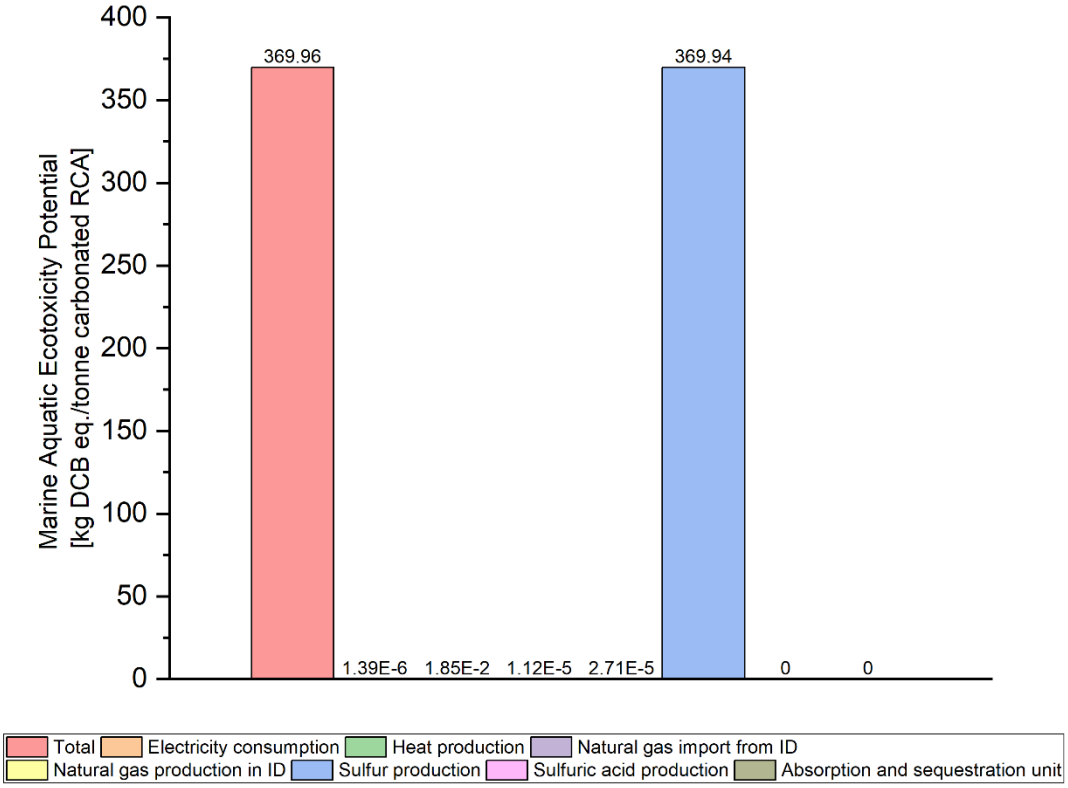

Figure 7. Marine Aquatic Ecotoxicity Potential results for the production of one tonne carbonated RCA.

Photochemical Ozone Creation Potential (POCP)

The bulk of the Photochemical Ozone Creation Potential (POCP) is associated with sulfuric acid and sulfur production, constituting 49.25% and 45.10%, as described in Figure 8, respectively. Other categories leading to POCP include electricity consumption (9.50×10^{-4} kg C_2H_4 eq., 3.45%), heat production (5.88×10^{-4} kg C_2H_4 eq., 2.14%), natural gas import (1.41×10^{-5} kg C_2H_4 eq., 0.05%) and production (3.71×10^{-6} kg C_2H_4 eq., 0.01%) from ID. The occurrence of POCP is induced by contaminants, including but not limited to carbon monoxide, methane, pentane, sulfur dioxide, and toluene. Sulfur dioxide is the major constituent responsible for the POCP in the unit processes, namely sulfuric acid and sulfur production, as indicated in our findings in Figure 8.

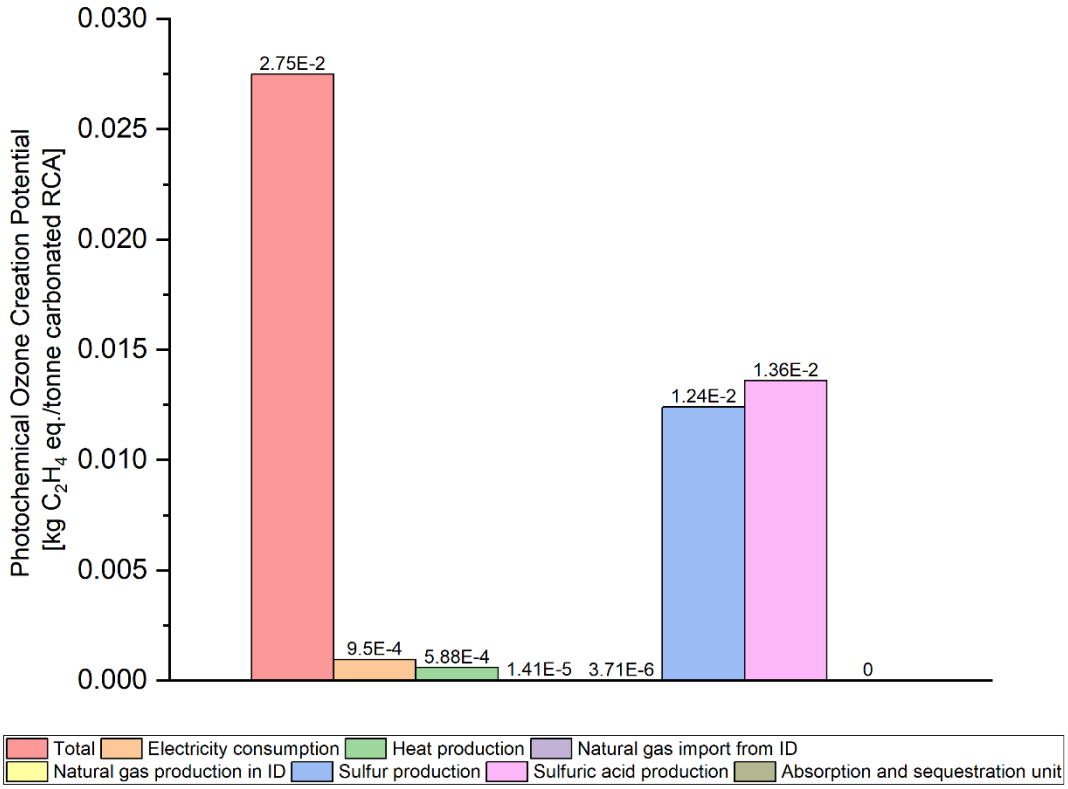

Figure 8. Photochemical Ozone Creation Potential results for the production of one tonne carbonated RCA.

Terrestric Ecotoxicity Potential (TETP)

Sulfur production contributes to a Terrestrial Ecotoxicity Potential (TETP) impact of 1.40 × 10^{-2} kg DCB eq., representing 97.62%. Heat production ranks second with an impact of 3.40 × 10-4 kg DCB eq., followed by electricity production (8 × 10-7 kg DCB eq., 0.01%). Natural gas production and import from ID have an inconsiderable impact on TETP. All these findings are detailed in Figure 9. Heavy metals are the sole determinants of TETP in sulfur production. The top three primary heavy metals are vanadium (1.12 × 10^{-2} kg DCB eq.), nickel (1.17 × 10^{-3} kg DCB eq.) and chromium (7.13 × 10^{-4} kg DCB eq.), collectively making up 93.38% of the TETP impact. In heat production, mercury stands out as the main constituent, contributing to 89.70% of the total. The second significant contribution comes from the air emission of formaldehyde (9.92%).

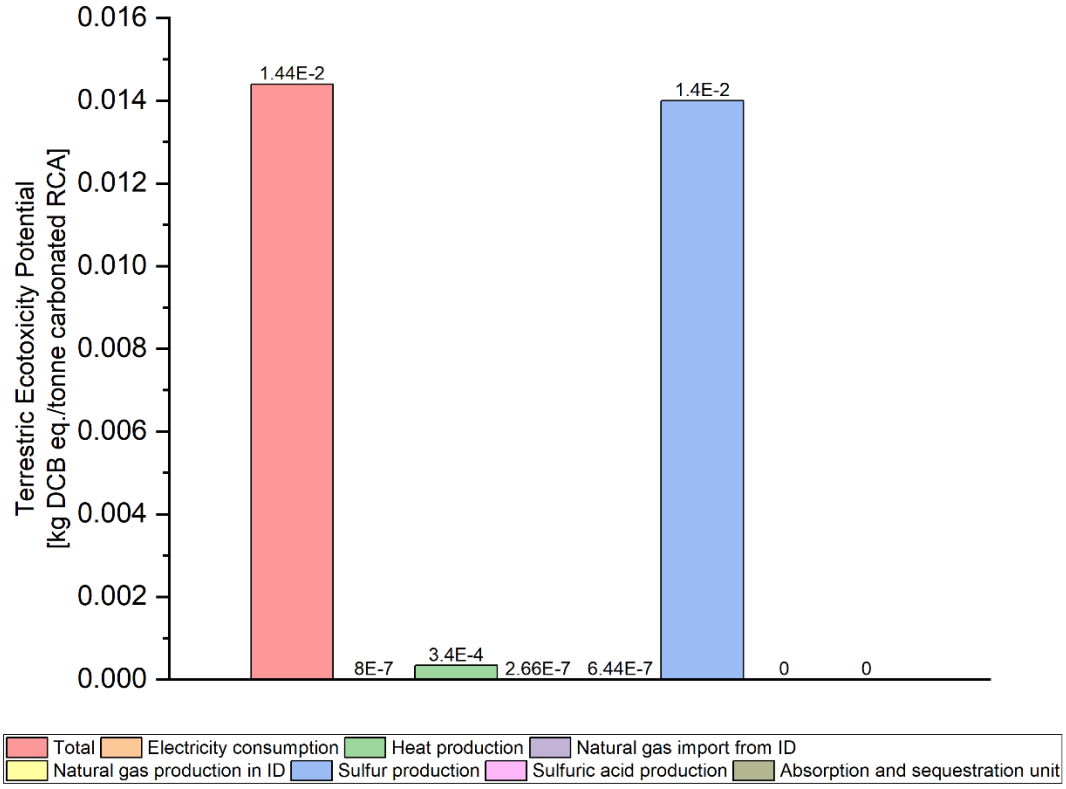

Figure 9. Terrestric Ecotoxicity Potential results for the production of one tonne carbonated RCA.

Sensitivity Analysis

Sensitivity analysis is conducted to comprehend the potential variability in LCA results and to identify the factors that significantly influence this variation [36, 81]. Sensitivity analysis is conducted utilizing the analyst feature provided in the Sphera® LCA for Experts (GaBi) version 10.7.1.28 software [63], with the resulting outputs presented in terms of percentage data variation. The sensitivity analysis for the selected eight environmental impact categories involves applying a standard deviation (SD) of ±20% [65] to the RCA mass, existing electricity and thermal energy consumption of the equipment in the process. No data variation was observed when applying a SD of ±20% to the RCA mass. This is likely attributable to the substantial excess of RCA in comparison to flue gas volume, resulting in an absence of observable change due to variations in RCA mass. The comprehensive breakdown of data variation, expressed as a percentage, for each piece of equipment and its total contribution based on electricity and thermal energy consumption is presented in Tables 1 and 2, respectively.

The most significant overall impact of data variation in electricity consumption on EP (±12.65%) is observed, followed by GWP (±12.43%), POCP (±0.60%), AP (±0.35%), and HTP (±0.28%). Solvent cooler makes the most substantial contribution to data variation in electricity consumption, while the pump has the lowest impact. This may be attributed to solvent cooler, which has the highest electricity consumption at 9.92×10^7 MJ per operating year compared to the other equipment. The pump, on the other hand, has the lowest electricity consumption at 1.58×10^5 MJ per operating year.

In the comparison of data variation in thermal energy consumption, HTP exhibits the most significant data variation of ±23.04%. Whereas AP shows the lowest variation at ±0.22%. Dryer 2 exhibits greater data variation in thermal energy consumption compared to Dryer 1, possibly attributed to its higher thermal energy requirement of 1.04×10^9 MJ in contrast to 7.86

$\times 10^8$ MJ per operating year. It is worth noting that FAETP, MAETP, and TETP are not influenced by a ±20% change in electricity consumption. Additionally, MAETP remains unaffected by alterations in thermal energy.

Table 1. Sensitivity analysis results with ±20% change in electricity consumption of the equipment.

Equipment	GWP (%)	AP (%)	EP (%)	HTP (%)	POCP (%)
Flue gas cooler	±0.07	NA	±0.07	NA	NA
Blower	±1.19	±0.03	±1.21	±0.03	±0.06
Pump	±0.01	NA	±0.01	NA	NA
Reactor	±0.22	±0.01	±0.23	±0.01	±0.01
Filter	±0.24	±0.01	±0.25	±0.01	±0.01
Condenser	±0.07	NA	±0.07	NA	NA
Wash tower cooler	±0.31	±0.01	±0.31	±0.01	±0.01
Solvent cooler	±6.85	±0.19	±6.97	±0.16	±0.33
Auxiliary	±3.46	±0.10	±3.52	±0.08	±0.17
TOTAL	**±12.43**	**±0.35**	**±12.65**	**±0.28**	**±0.60**

Table 2. Sensitivity analysis results with ±20% change in thermal energy consumption of the equipment.

Equipment	GWP (%)	AP (%)	EP (%)	FAETP (%)	HTP (%)	POCP (%)	TETP (%)
Dryer 1	±3.81	±0.10	±3.78	±0.24	±9.94	±0.39	±0.41
Dryer 2	±5.02	±0.13	±4.98	±0.32	±13.10	±0.51	±0.54
TOTAL	**±8.83**	**±0.22**	**±8.76**	**±0.56**	**±23.04**	**±0.90**	**±0.95**

Monte Carlo simulation

Monte Carlo simulation evaluates the uncertainty and variability in model outputs by randomly sampling input parameters from their probability distributions and conducting multiple iterations of the model [36]. In contrast to sensitivity analysis, which primarily examines the influence of individual input variables on the output, Monte Carlo simulation accounts for the collective effects of multiple input variables and their interrelations [82]. Monte Carlo simulation offers supplementary insights, including the probability distribution of model outputs, thereby enabling a more thorough comprehension of uncertainty and risk [81, 82]. Moreover, Monte Carlo simulation can integrate correlations among input variables, shedding light on how alterations in one variable may influence others and consequently impact the model outcomes [82].

In our Monte Carlo simulation, we conduct 10,000 iterations, adjusting electricity and thermal energy consumption by an SD of 20% [65], consistent with the settings used in the sensitivity analysis. The occurrences of various outputs produced by the Monte Carlo simulation are depicted as a normal distribution. The results of the Monte Carlo simulation, incorporating a ±20% change in both electricity and thermal energy consumption, are provided in Tables 3 and 4, respectively. Both results indicate that the mean value of GWP changes from the base case scenario, while the mean values of other environmental impact categories have slight or no changes. The analysis of Monte Carlo simulation results for a ±20% variation in electricity consumption reveals that EP is most significantly impacted, attributed to its largest standard deviation (SD) of 7.88%. Conversely, MAETP exhibits the least impact, given its SD of 4.09×10^{-8}%. In the analysis of Monte Carlo simulation results for a ±20% variation in thermal energy consumption, HTP registers the highest standard deviation (SD) at 16.50%, while MAETP shows the least impact with an SD of 1.42×10^{-3}%. The ±20% variation in electricity consumption has negligible impacts on FAETP, HTP, MAETP, and TETP, as the

values of the mean, 10%, 25%, 75%, and 90% remain unchanged. With a ±20% fluctuation in thermal energy consumption, only the values of MAETP remain unaffected.

Table 3. Monte Carlo simulation results with ±20% change in electricity consumption of the equipment.

Environmental Impact Categories	Mean	SD (%)	10% percentile	25% percentile	75% percentile	90% percentile
Global Warming Potential [kg CO_2 eq.]	2.27×10^{-1}	7.74	2.04×10^{-1}	2.15×10^{-1}	2.39×10^{-1}	2.49×10^{-1}
Acidification Potential [kg SO_2 eq.]	6.77×10^{-1}	2.16×10^{-1}	6.75×10^{-1}	6.76×10^{-1}	6.78×10^{-1}	6.79×10^{-1}
Eutrophication Potential [kg PO_4 eq.]	4.17×10^{-3}	7.88	3.75×10^{-3}	3.95×10^{-3}	4.39×10^{-3}	4.59×10^{-3}
Freshwater Aquatic Ecotoxicity Potential [kg DCB eq.]	3.72×10^{-2}	2.05×10^{-3}	3.72×10^{-2}	3.72×10^{-2}	3.72×10^{-2}	3.72×10^{-2}
Human Toxicity Potential [kg DCB eq.]	1.73	1.76×10^{-1}	1.73	1.73	1.73	1.73
Marine Aquatic Ecotoxicity Potential [kg DCB eq.]	3.70×10^{2}	4.09×10^{-8}	3.70×10^{2}	3.70×10^{2}	3.70×10^{2}	3.70×10^{2}
Photochemical Ozone Creation Potential [kg C_2H_4 eq.]	2.75×10^{-2}	3.75×10^{-1}	2.74×10^{-2}	2.75×10^{-2}	2.76×10^{-2}	2.77×10^{-2}
Terrestric Ecotoxicity Potential [kg DCB eq.]	1.44×10^{-2}	6.06×10^{-4}	1.44×10^{-2}	1.44×10^{-2}	1.44×10^{-2}	1.44×10^{-2}

Table 4. Monte Carlo simulation results with ±20% change in thermal energy consumption of the equipment.

Environmental Impact Categories	Mean	SD (%)	10% percentile	25% percentile	75% percentile	90% percentile
Global Warming Potential [kg CO_2 eq.]	2.27×10^{-1}	6.31	2.08×10^{-1}	2.17×10^{-1}	2.36×10^{-1}	2.45×10^{-1}
Acidification Potential [kg SO_2 eq.]	6.77×10^{-1}	1.58×10^{-1}	6.75×10^{-1}	6.76×10^{-1}	6.77×10^{-1}	6.78×10^{-1}
Eutrophication Potential [kg PO_4 eq.]	4.17×10^{-3}	6.26	3.83×10^{-3}	3.99×10^{-3}	4.34×10^{-3}	4.51×10^{-3}
Freshwater Aquatic Ecotoxicity Potential [kg DCB eq.]	3.72×10^{-2}	4.02×10^{-1}	3.70×10^{-2}	3.71×10^{-2}	3.73×10^{-2}	3.74×10^{-2}
Human Toxicity Potential [kg DCB eq.]	1.73	16.50	1.37	1.54	1.92	2.10
Marine Aquatic Ecotoxicity Potential [kg DCB eq.]	3.70×10^{2}	1.42×10^{-3}	3.70×10^{2}	3.70×10^{2}	3.70×10^{2}	3.70×10^{2}
Photochemical Ozone Creation Potential [kg C_2H_4 eq.]	2.75×10^{2}	6.45×10^{-1}	2.73×10^{-2}	2.74×10^{-2}	2.77×10^{-2}	2.78×10^{-2}
Terrestric Ecotoxicity Potential [kg DCB eq.]	1.44×10^{-2}	6.78×10^{-1}	1.42×10^{-2}	1.43×10^{-2}	1.44×10^{-2}	1.45×10^{-2}

Scenario analysis

Scenario analysis within LCA involves evaluating the environmental performance of a product, process, or system under varying conditions or assumptions [83]. It facilitates a thorough comprehension, informed decision-making, and comparison of alternative scenarios, aiding in the identification of sustainability improvement strategies by examining variations in different parameters [83]. In summary, scenario analysis investigates diverse future scenarios [83], sensitivity analysis identifies principal factors contributing to uncertainty [81], and Monte Carlo simulation quantifies the comprehensive uncertainty in model outputs [82]. Each method offers valuable insights into the reliability and robustness of model results. In the context of scenario analysis, we modified the functional unit to per tonne of CO_2 input to holistically study the carbon abatement potential of our mineral carbonation process.

The entire process results in 530.82 kg CO_2 eq. in GWP emissions, as illustrated in Figure 11. This total comprises heat production (269.91 kg CO_2 eq.), electricity consumption (235.74 kg CO_2 eq.), sulfur production (24.37 kg CO_2 eq.), natural gas production in ID (0.57 kg CO_2 eq.), and natural gas import from ID (0.23 kg CO_2 eq.). The absorption and sequestration unit can eliminate 81.75% of one tonne of CO_2 (817.47 kg CO_2 eq.). Considering this, our proposed technology achieves an overall capture efficiency of 286.66 kg CO_2 eq.

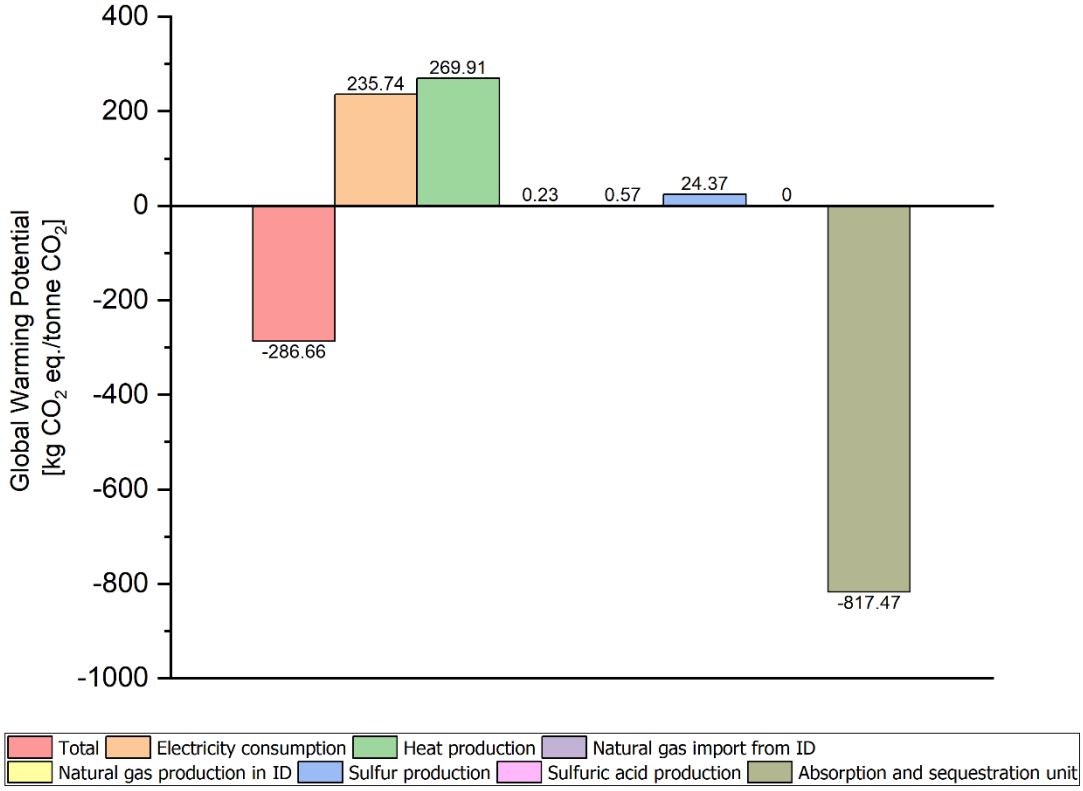

Figure 11. Global Warming Potential results based on input of one tonne CO_2.

5. Conclusion

In this study, we introduced a carbon mineralization technology targeting the capture and utilization of low-concentration CO_2 from the flue gas of NGCC power plants. The Materials and Methods section detailed the process designed to capture 200 kilotonnes of CO_2 per operating year. A cradle-to-gate LCA was conducted for this industrial-scale carbon mineralization technology, using a functional unit of per tonne of carbonated RCA output.

The results revealed a net positive carbon abatement of 21.42 kg CO_2 eq per tonne carbonated RCA output. Heat production and electricity consumption emerged as major hotspots contributing to carbon emissions. Among the eight studied environmental impact categories, MAETP demonstrated the most substantial environmental impact, with sulfur production identified as the key process. The top three air contaminants from sulfur production are vanadium, hydrogen fluoride, and nickel.

HTP was the second-worst contributor to environmental impact, with heat and sulfur production contributing to 96.80% of HTP. Heavy metals were identified as the primary contributors to HTP impact in our technology. Following HTP, AP had a significant environmental impact, driven by sulfur and sulfuric acid production for the acid wash tower, where sulfur dioxide was the primary air pollutant.

We conducted sensitivity analysis and Monte Carlo simulation by introducing a standard deviation of ±20% to the existing electricity and thermal energy consumption of the equipment to identify variations and uncertainties for process improvement. In sensitivity analysis, the solvent cooler exhibited the highest variation in electricity consumption across GWP, AP, EP, HTP, and POCP. Conversely, dryer 2 displayed the highest variation in thermal energy consumption across all environmental impact categories except MAETP, which remained unaffected by thermal energy changes. The mean value of GWP changed to 2.27×10^{-1} kg CO_2 eq. from the base case value of -21.42 kg CO_2 eq. in Monte Carlo simulation. Meanwhile, the Monte Carlo simulation results minimally or insignificantly impacted other environmental impact categories.

To gain deeper insights into the carbon abatement potential of our technology, we conducted a scenario analysis by adjusting the functional unit to per tonne of CO_2 input. Similarly, heat production and electricity consumption were identified as critical factors influencing GWP. Our system realized a net carbon-positive abatement potential of 286.66 kg CO_2 eq. per tonne of CO_2 input.

Enhancements to our carbon mineralization technology involve adoption of more energy-efficient and heat recovery equipment to reduce electricity and thermal energy requirements. The process can be further optimized by excluding the acid wash tower process, thereby eliminating the sulfur and sulfuric acid processes. This optimization can be achieved

by exploring the suitability of alternative equipment designed for removing ammonia to meet emission standards.

The prospect of employing carbon mineralization to capture and utilize CO_2 extracted from flue gas in NGCC power plant for carbonated RCA production presents a viable solution. This approach not only addresses Singapore's reliance on imported sand for land reclamation and construction but also positions carbon mineralization as a feasible choice for CCUS in Singapore. Regarding enhancement to our LCA, our future trajectory entails investigating a broader-scale scenario that extends beyond the capture of 200 kilotonnes of CO_2 annually. Additionally, we aim to investigate alternative ammonia treatment technologies to replace reliance on acid wash tower and integrate heat exchangers into our process for energy consumption reduction. Furthermore, to augment the comprehensiveness of our sustainability evaluation, we plan to assess the LCA of other CCUS technologies and perform techno-economic assessments.

CRediT authorship contribution statement

Pancy Ang: Life cycle assessment, data analysis, and writing – initial draft, visualization, review and editing. **Wayne Goh**: Process design, Aspen simulation, and data analysis. **Bu Jie**: Funding acquisition, supervision, and project administration. **Cheng Shuying:** Process design, Aspen simulation, data analysis, supervision and writing – review and editing.

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Declaration of competing interest

The authors declare that they have no known competing financial interests.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at DOI:

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