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Abstract

Ocean alkalinity enhancement is a promising carbon dioxide removal (CDR) approach, but scaling up to gigatonnes (Gt) of CO₂ per year will require safe, sustainable, and abundant alkaline feedstocks. Here, we propose the use of a relatively unexplored resource for OAE, namely naturally occurring sodium (bi)carbonates. We identified and mapped 109 such deposits globally, although quantitative resource information is available for only 16. Quantified deposits collectively contain >200 Gt of sodium (bi)carbonate-rich minerals and brines, dominated by trona (Na₂CO₃·NaHCO₃·2H₂O) and nahcolite (NaHCO₃) mainly concentrated in the USA, China, Turkey, and Kenya. We then assessed three OAE pathways using trona as a feedstock, i.e., 1) Mining, crushing, and ocean dispersal of trona (gross CDR capacity 0.16 tCO₂ t⁻¹); 2) Calcine trona with carbon capture and storage to produce soda ash (Na₂CO₃) (0.31 tCO₂ t⁻¹) prior to dispersal; and 3) Purification of soda ash via dissolution, crystallisation, and drying prior to dispersal. Using Green River, Wyoming, USA (~116 Gt of bedded trona) as a case study, life cycle assessment informed on the net-negativity of each pathway. Provided that mining and transportation have largely decarbonised, all pathways are net-negative (carbon penalties in the range 29–41%), with pathway 2 achieving the highest net CDR capacity (0.21 tCO₂ t⁻¹). Solution mining and reserves nearer the coastline can optimise net-negativity. Overall, naturally occurring sodium (bi)carbonate resources emerge as a promising future feedstock for soluble, safe, and scalable OAE.

Word count: ~7532

Keywords

Negative emissions technologies (NETs), climate change intervention, ocean acidification, evaporites, salt pans, alkaline/saline lakes, natron.

1. Introduction

Climate change, driven by rapidly rising atmospheric CO₂ concentrations, particularly since the mid-20th century [1,2], actively threatens global ecosystems, economies, and human wellbeing [3]. Even with the ambitious emissions reductions efforts that are currently pursued, limiting warming to well below 2°C, as specified in the Paris Agreement [4], will also require the deployment of large-scale carbon dioxide removal (CDR) [5]. CDR is increasingly recognised as a critical component in climate change intervention, enabling not only the removal of excess (anthropogenic origin) CO₂ from the atmosphere but also the offsetting of hard-to-abate emissions such as from transportation and land-use change [6,7].

However, the CDR landscape remains nascent, with most technologies being still at an early stage of development, and requiring rapid scale-up from the current ~1.3 million tonnes (Mt) of removals to the billion-tonne (gigatonne, Gt) scale as early as 2030 [8]. Among the many CDR approaches, ocean alkalinity enhancement (OAE), an umbrella term encompassing several technologies aiming at increasing the ocean's capacity to absorb and durably store CO₂, offers a promising pathway [9]. Specifically, through alkalinity addition to seawater, OAE accelerates the natural drawdown of atmospheric CO₂, facilitating its long-term (>10,000 years) sequestration, primarily as stable dissolved bicarbonate (HCO₃⁻) ions, while contributing, to some extent, to the mitigation of ocean acidification [10].

OAE can be implemented through a range of mineral-based and electrochemical pathways, each with distinct engineering restrictions and environmental challenges [11]. Mineral-based routes rely on abundant rocks, such as limestone or (ultra)mafic rocks, but these demand (substantial) processing to accelerate dissolution in seawater. For example, in coastal enhanced weathering, olivine-rich dunite is mined, crushed, and spread in coastal zones to accelerate CO₂ drawdown [12,13]. However, dissolution rates are slow, particularly at the particle sizes realistically encountered in coastal environments, while comminution to very small particle sizes comes with significant energy and environmental penalties, while heavy metals content such as nickel may also limit deployments capacities [14–16].

Limestone is another highly abundant rock resource, but it is not soluble in surface ocean waters and therefore requires pre-treatment by calcination or chemical dissolution in reactors before use [17–19]. In ocean liming, for instance, limestone is calcined and slaked to form hydrated lime (calcium hydroxide, Ca(OH)₂), which is then dispersed in surface waters to enhance CO₂ uptake [20]. While effective in principle, this pathway can cause local pH spikes, ecological stress, and runaway carbonate precipitation [21–23]. Other routes, such as electrochemical alkalinity generation which uses seawater/brines electrolysis or electro dialysis to generate alkalinity, namely aqueous sodium hydroxide (NaOH) [24–26], have been explored. However, current implementations remain at low technology readiness levels (TRLs), and depend on low-carbon electricity to achieve net CO₂ removal, while electrochemical methods also generate an acidic by-product stream which requires safe management [27]. These constraints motivate the search for alternative, lower-energy alkaline resources capable of supporting Gt-scale CDR, such as the synthesis of highly soluble hydrated carbonate minerals, e.g., ikaite (CaCO₃·6H₂O) [28].

An underexplored material for OAE is sodium carbonate (Na₂CO₃), which can be derived from naturally occurring sodium (bi)carbonate evaporites, containing minerals such as trona (Na₂CO₃·NaHCO₃·2H₂O), nahcolite (NaHCO₃), natron (Na₂CO₃·10H₂O), thermonatrite (Na₂CO₃·H₂O), and natrite (Na₂CO₃). Sodium carbonate dissolves readily in seawater, while, unlike (ultra)mafic rocks or lime, it does not release divalent cations (Ca²⁺, Mg²⁺) that are more prone to precipitation. Moreover, it appears that the use of Na₂CO₃ for OAE has so far been associated with relatively low ecological disturbances [29–31]. Despite these advantages, Na₂CO₃ has received comparatively little attention, largely because Khesghi's seminal paper on OAE suggested that naturally occurring Na₂CO₃ resources are too limited to support OAE at scale [32]. Their uneven, and often restricted, geographic distribution further adds to scalability concerns.

More recent work suggests that at least 95 natural sodium (bi)carbonate deposits have been identified globally, though many more remain unquantified [33,34]. Furthermore, a large proportion (>20%) of global soda ash (Na_2CO_3) demand, currently $\sim 75 \text{ Mt yr}^{-1}$, is met by the mining and processing of natural sodium (bi)carbonate evaporites ($\sim 15 \text{ Mt yr}^{-1}$), a large share of which is supplied from the Green River trona deposit in Wyoming [35]. (The remainder is met by synthetic soda ash produced via the energy intensive Solvay or the Hou processes). This raises a key question, could additional or underexplored deposits expand the global resource base, and can these feedstocks sustain OAE at the Gt-scale? Yet, even if sufficient resources exist, their net carbon performance must be carefully assessed, since raw trona (and the soda ash produced from it) offers lower CO_2 removal per unit of mass compared to materials such as lime or olivine [33], meaning there is a risk that upstream carbon emissions could be greater than the carbon benefit of OAE.

Therefore, to investigate the potential of naturally occurring sodium (bi)carbonate resources for OAE, we have mapped global trona, nahcolite, and other related minerals and brines, and quantified resources where possible. Using the life cycle assessment (LCA) methodology we then assessed the net-negativity of crushed trona and calcined trona (i.e. soda ash) using, as a case study, the existing Green River (Wyoming) trona mining and processing operation(s). Environmental hotspots were identified and avenues to optimise the net-negativity of these resources were proposed. Together, these analyses inform, for the first time, the sustainable scaling-up of naturally occurring sodium carbonate resources as a viable feedstock for climate change mitigation via OAE.

2. Materials and Methods

2.1 Mapping global sodium (bi)carbonate evaporite resources

An open access online database of minerals and their localities, deposits, and mines worldwide [36] was used as a basis for mapping sodium carbonate evaporite resources. The database was queried with exact mineral terms “trona”, “nahcolite”, “natron”, “thermonatrite”, “natrite”, and “wegscheiderite”. Search results were screened manually such that only substantial, naturally occurring deposits were included in the developed inventory. For example, soil accumulations [37] and anthropogenic efflorescences [38] were excluded.

The search was supplemented with additional queries via Scopus and Google Scholar for academic-identified natural sodium (bi)carbonate minerals, and further, by non-academic (government, media, and industry) sources. For these searches, we combined the mineral names listed above with broader terms like “evaporites”, “salt lakes”, “salt pans”, “saline lakes”, “alkaline lakes”, “brine lakes”, “salars” as well as “soda ash”, “sodium carbonate”, and “sodium bicarbonate”. In addition to geographic coordinates, data were collected on specific sodium (bi)carbonate mineralogy (see Table 1), as well as the other evaporite minerals present (e.g., halite (NaCl), mirabilite ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$)). Mixed alkali or alkaline-earth minerals such as shortite ($\text{Na}_2\text{Ca}_2(\text{CO}_3)_3 \cdot 2\text{H}_2\text{O}$), were not searched for specifically but were recorded when present. Wherever possible, resource/reserve estimates were included as well as current and historic production rates.

Finally, our review methodology for the global resource mapping was also extended to backward citation snowballing. Where possible, we traced resource estimates and supporting data back to their original sources to compare and verify the mass units used (short tons vs. metric tonnes) and avoid inconsistencies.

2.2 Conceptual and engineering pathways for OAE through sodium (bi)carbonate evaporites

In OAE, pre-processing is often essential to enhance the CO_2 removal potential of (mineral) feedstocks and/or improve their dissolution kinetics. Lee Pereira et al. provide a taxonomic framework for the different types of pre-processing [39], which includes: *enhanced weathering/direct addition*, where grinding is employed to increase surface areas and improve dissolution [15]; *ocean liming/heat transformation*, where minerals are subject to thermal

treatment, e.g., calcination of limestone (CaCO_3) to lime (CaO) with carbon capture and storage (CCS) [20]; *chemical pathways*, where minerals, e.g., CaCO_3 , are reacted with other chemicals, e.g., CO_2 and H_2O , to create more soluble forms of alkalinity [28,40]; and finally *electrochemical pathways*, e.g., electrochemical splitting of brines (rich in NaCl) towards NaOH production [24,26].

Sodium-based alkalinity has been widely examined for OAE, mainly through the electrochemical transformation route [24–26], although Na_2CO_3 has also played a role in a variety of seawater-based CO_2 capture technologies [41,42]. There are no fully-described pathways that propose directly or indirectly using naturally occurring solid minerals (e.g., evaporites) or brines of $\text{NaHCO}_3/\text{Na}_2\text{CO}_3$ for OAE. Thus, here we propose three main engineering pathways for OAE using trona which is dry-mined via the room-and-pillar method (i.e. the predominant method at the world’s largest trona mine located in Green River, Wyoming) (see **Figure 1**).

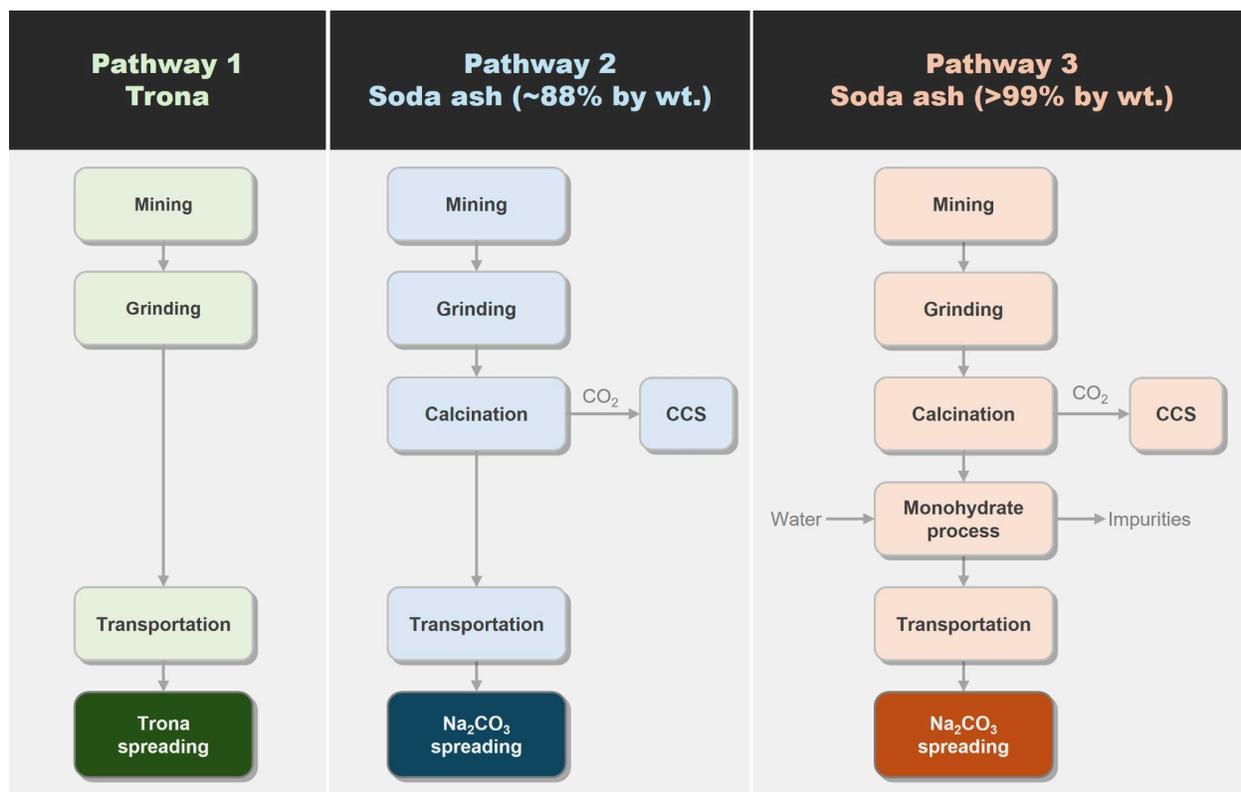
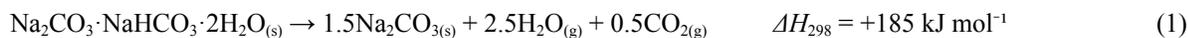


Figure 1 The proposed pathways for OAE using trona: 1. Trona is mined by the room-and-pillar method, crushed, transported, and dispersed directly into the ocean. 2. Crushed trona is calcined to produce soda ash, then dispersed into the ocean – the CO_2 released during trona calcination is captured and stored geologically (e.g., in saline aquifers). 3. Similarly, trona is calcined to soda ash but in this case is purified via the monohydrate process (dissolution, crystallisation, drying, etc.) before ocean spreading.

Specifically, pathway 1, which is the simplest, involves adding finely ground trona (<1 mm), or other Na_2CO_3 -rich evaporites such that they dissolve fully in well-mixed surface ocean waters. pathway 2 involves the grinding and thermal transformation (calcination) of crushed trona to soda ash (see Equation (1)) [43], followed by ocean spreading. Pathways 1 and 2 require that any impurities present in the mined ore are benign upon ocean addition, which appears to be the case in most identified resources (e.g., mainly halite, shortite) (see **Supplementary Table S1**).



If trona impurities are substantial (or not benign), then these can be removed by one of several purification routes. Pathway 3 explores this route whereby purification is achieved by the (energy-intensive) monohydrate process, in which calcined trona is first dissolved, then the insoluble impurities are removed by filtration and the pure sodium carbonate monohydrate ($\text{Na}_2\text{CO}_3 \cdot \text{H}_2\text{O}$) is precipitated out of the solution (via evaporative crystallisation) and then dried to anhydrous sodium carbonate. The produced dense soda ash, which is >99% pure, can be spread in surface ocean waters for OAE.

Although the monohydrate process is the current industrial standard for high-purity dense soda ash [44], such purity is not strictly necessary for OAE since common trona impurities are often environmentally benign (e.g., sand and clay), already abundant in seawater (e.g., halite, NaCl and mirabilite, $\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), or may even contribute additional alkalinity (e.g., shortite – $\text{Na}_2\text{Ca}_2(\text{CO}_3)_3 \cdot 2\text{H}_2\text{O}$). Thus, utilising low-purity Na_2CO_3 via pathway 2 offers distinct advantages compared to pathway 3 in terms of reduced energy and carbon footprint. However, pathway 3 represents an upper bound in processing intensity, allowing us to quantify the trade-offs between product purity and increased energy demand and upstream carbon emissions. Together, these three pathways cover a reasonable spectrum of processing intensity from simple grinding to thermal transformation with CCS, enabling the realistic assessment of OAE using naturally occurring sodium (bi)carbonates. Although it is likely other pathways for valorisation exist, such as electrochemical approaches [45,46], these are beyond the scope of this investigation and can be examined in future research. To ensure a fair comparison of the carbon penalties of the three pathways, the Green River trona mine and soda ash production facility in Wyoming (US) was used as the case study location, since it holds substantial resources (116 Gt) thus enabling future scaling.

The maximum theoretical CO_2 removal efficiencies for dissolution of trona and soda ash in seawater were determined using the multicomponent geochemical model code PHREEQC [47] with the database `lml.dat`. Seawater composition and temperature were taken as the annual mean values representative of the coastal regions near Portland, USA which is the assumed location for alkalinity dispersal [48]. The value for $\log_{10}K$ (K =equilibrium constant at 25 °C) for trona was obtained from Vanderzee (1982)[49].

2.3 Life cycle assessment

To assess the environmental sustainability, identify the main environmental hotspots, and optimise the net-negativity of each examined engineering pathway, a cradle-to-grave LCA methodology was employed, as detailed in ISO 14040 [50] and 14044 [51]. For the environmental modelling the software program SimaPro 10 was used, with the ecoinvent database (v3.12) being the preferred option to model the collected life cycle inventory (LCI) data. To provide insight about the carbon net-negativity of the considered pathways the single issue life cycle impact assessment (LCIA) method IPCC 2021 was used, considering the 100-year global warming potential (GWP100). All main processes, starting from trona mining, to alkalinity production, and finally its spreading for CO_2 drawdown, were included in their corresponding system boundaries. These main processes are shown in **Figure 1**. The collected LCI data were then used to estimate the life cycle carbon emissions (carbon footprint) of each engineering pathway for a gross removal of 1 tonne of CO_2 from the atmosphere, which is the functional unit herein. This then informed the carbon penalty of each pathway, i.e., the ratio of the total upstream carbon emissions to the gross CDR capacity. In essence, this represents the inefficiency of the system in carbon removal as it shows how much of the gross CDR capacity is 'lost' to upstream emissions. Given that the goal is to provide an initial understanding of the net-negativity and carbon hotspots of each examined OAE pathway, the LCI data collection stage focused on capturing the key primary data, such as electricity input for grinding and heat consumption for calcination when using the Green River trona mine as a case study. Further details on the collected LCI data and the assumptions and limitations of the LCA study are given in the **Supplementary Material (Appendix A and Table A1)**. Finally, the

intended audience of the LCA study includes the soda ash and CDR industries, OAE researchers, and decision- and policy-makers alike.

3. Results and discussion

3.1 Global sodium (bi)carbonate natural resource inventory

Our extensive literature survey identified 109 naturally occurring sodium (bi)carbonate deposits (see **Figure 2** and **Supplementary Table S1**), higher than previous estimates of 95 [34] and 66 [33]. These deposits are widely distributed across the globe but predominantly occur in the arid to semi-arid basins of the USA (e.g., in Wyoming, Nevada, and California), China (e.g., Inner Mongolia), and Africa. Out of these 109 occurrences, just 16 sites had reported resource estimates, and of these 16, only 4 provided reserve estimates. Yet, these 16 sites hold 208 Gt of various sodium (bi)carbonate-rich minerals (including brines), most of which are attributable to bedded trona ore (~124 Gt).

The world's largest known trona deposit occurs near Green River in the Green River Basin (southwestern Wyoming, USA). The Green River trona deposit was formed around 50 million years ago (Eocene), from a vast system of interconnected alkaline lakes [52]. Weathering of surrounding volcanic rocks released sodium (Na^+), which accumulated in this hydrologically-closed basin [53,54]. These lakes were enriched in dissolved inorganic carbon of magmatic and/or biogenic origin [55,56]. As evaporation progressed, trona became supersaturated and precipitated, forming thick, laterally extensive beds, interlayered with shale, and oil shale, now found at 180–900 m depth. The USGS estimates ~116 Gt (metric tonnes) of trona ore in 22 well-mapped lateral beds, with broader estimates of ~135 Gt [57–59]. Reserves are commonly reported as >36 Gt assuming conventional mining methods.

Within the broader Green River Formation, substantial amounts of nahcolite (NaHCO_3) are also found, specifically ~39 Gt of resources in the Piceance Creek Basin in Colorado [60]. The Piceance Creek Basin also hosts around 17 Gt of dawsonite ($\text{NaAlCO}_3(\text{OH})_2$) [61,62]. Another substantial resource was identified at Searles Lake, California, which holds over 800 Mt of soda ash equivalent reserves, primarily as sodium (bi)carbonate rich brines [61,63]. Smaller deposits exist at Owens Lake, California [64] and other alkaline lake environments (e.g., Big Soda Lake, Nevada and similar playa systems), however these collectively likely contribute less than a gigatonne of sodium (bi)carbonate mineral resources [34,65].

Outside of the US, major trona deposits occur in Turkey with trona resources in Beypazarı (~600 Mt) and Kazan (~1.6 Gt), with combined reserves of >900 Mt [66,67]. In China, natural soda ash production derives from several lacustrine basins, notably Alxa League (>1.5 Gt of sodium (bi)carbonates), Chaganor (Qagan Nur) Salt Lake (~11 Mt of sodium (bi)carbonate rich brines), Tongbai (~193 Mt, mainly nahcolite), and Wucheng (~37 Mt, trona/nahcolite) [61,68–70]. In Africa, the largest natural soda-ash resource is found at Lake Magadi (Kenya), which contains more

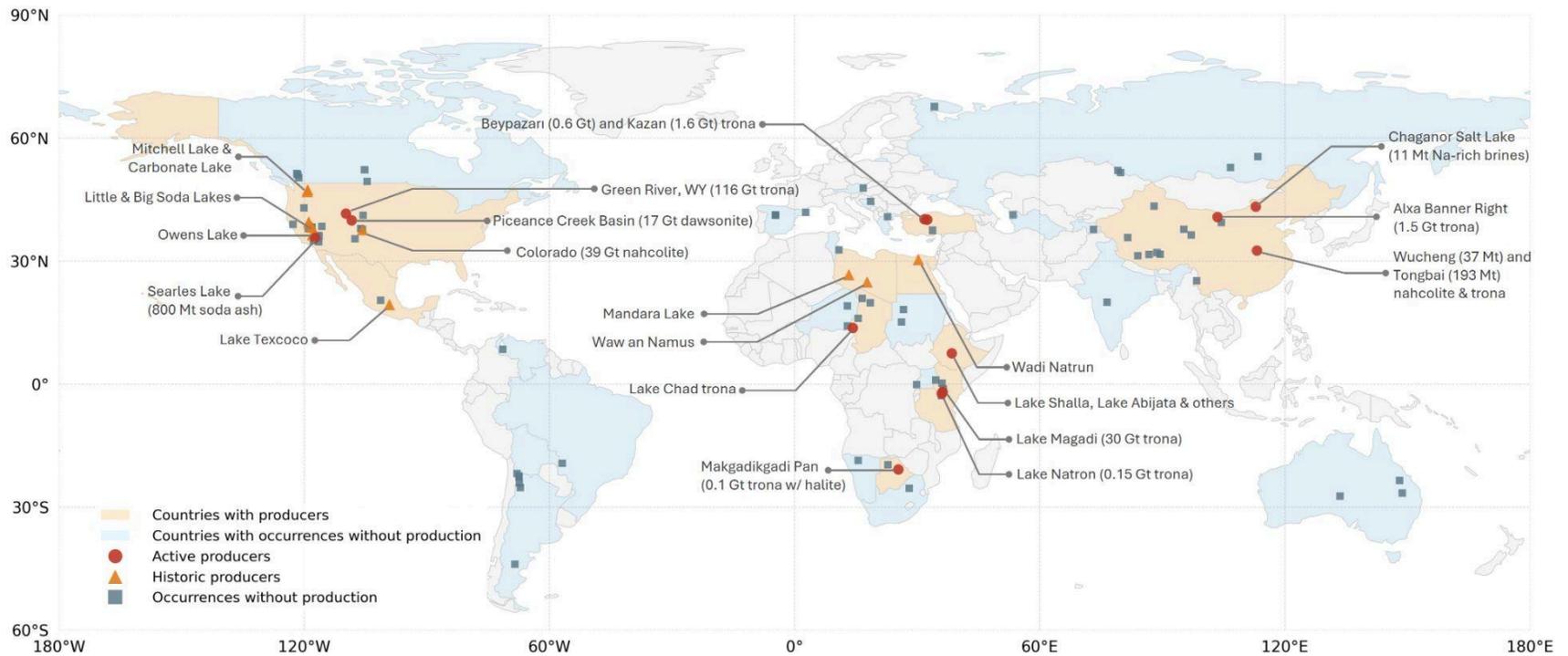


Figure 2 Locations and resource estimates (where applicable) of the world's natural sodium (bi)carbonate occurrences with historic, current, and potential future producers.

than 30 Gt of sodium (bi)carbonate resources (brines and trona crusts) [61,71–73]. Smaller occurrences (hundreds of megatonnes scale) also exist at Lake Natron, Tanzania, [74], Sua Pan, Makgadikgadi, Botswana, [75,76] and various sites in Chad [77,78] where trona and related evaporite minerals and brines, are present but remain less well quantified.

Of the sodium (bi)carbonate minerals targeted by our survey, trona ($\text{Na}_2\text{CO}_3 \cdot \text{NaHCO}_3 \cdot 2\text{H}_2\text{O}$) was the most frequently reported (87 occurrences), followed by natron ($\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$), thermonatrite ($\text{Na}_2\text{CO}_3 \cdot \text{H}_2\text{O}$), nahcolite (NaHCO_3), natrite (Na_2CO_3), and wegscheiderite ($\text{Na}_2\text{CO}_3 \cdot 3\text{NaHCO}_3$) (see **Figure 3** and **Supplementary Table S1**). Although not specifically targeted in our search, additional co-occurring sodium and (bi)carbonate-bearing minerals were also identified, including gaylussite ($\text{Na}_2\text{Ca}(\text{CO}_3)_2 \cdot 5\text{H}_2\text{O}$), burkeite ($\text{Na}_6(\text{CO}_3)(\text{SO}_4)_2$), pirssonite ($\text{Na}_2\text{Ca}(\text{CO}_3)_2 \cdot 2\text{H}_2\text{O}$), northupite ($\text{Na}_3\text{Mg}(\text{CO}_3)_2\text{Cl}$) among others. Besides these, other commonly recurring minerals included halite (NaCl), thenardite (Na_2SO_4), calcite (CaCO_3), mirabilite ($\text{Na}_2\text{SO}_4 \cdot 10\text{H}_2\text{O}$), and gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), etc.

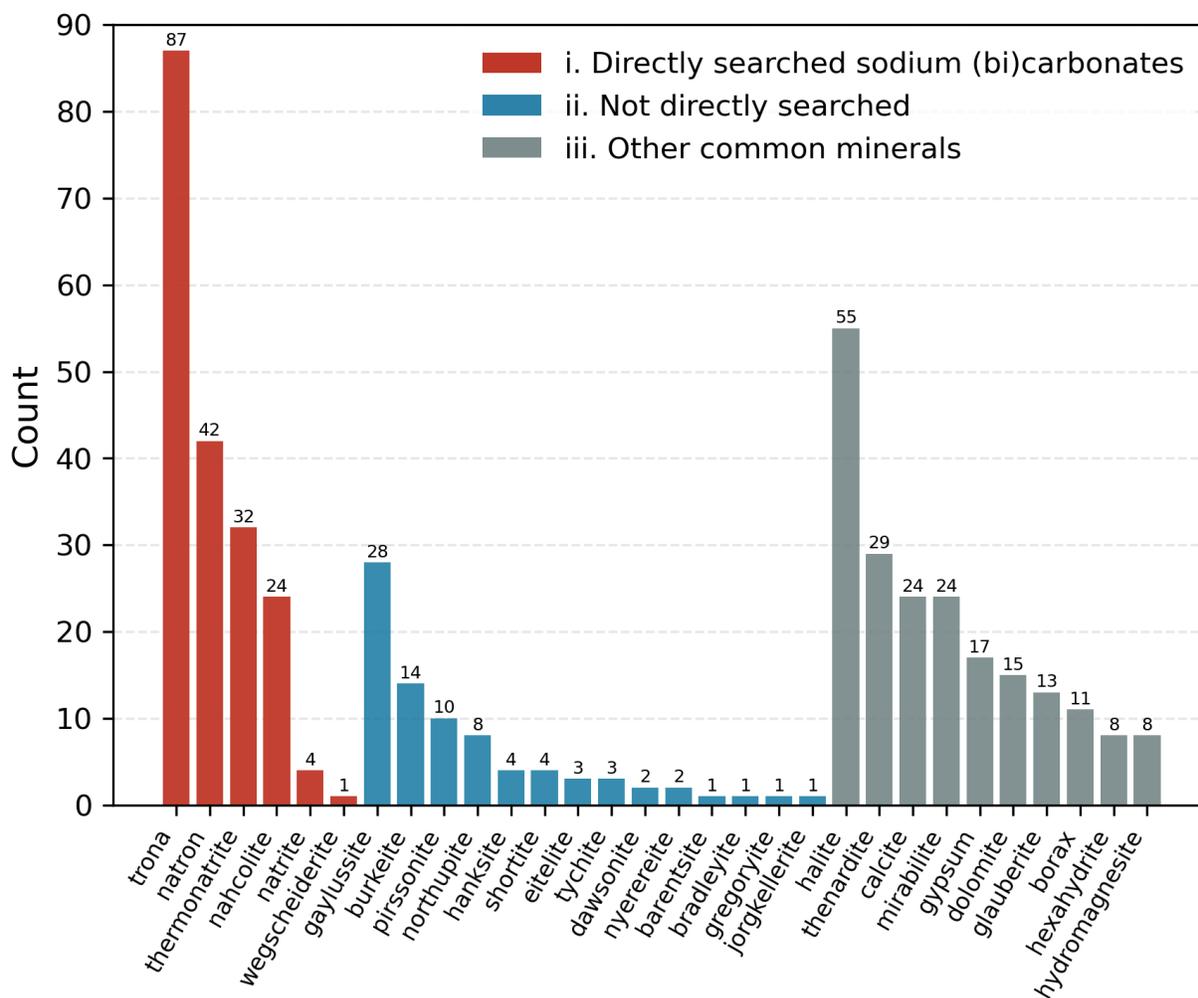


Figure 3 i. Sodium (bi)carbonate minerals which were the direct target of the literature search; ii. Sodium (bi)carbonate-type mineral occurrence count but which were not the direct target of the literature search; iii. Count for some of the commonly encountered minerals which are not sodium (bi)carbonates.

Impurity types and levels vary widely and influence each deposit's suitability for production, and also CDR – as they affect the gross CDR potential per mass of mined material. For example, of the 116 Gt of trona in Wyoming, 69 Gt are described as <2% “halite-free” while the remainder contains >2% halite [57,59,79]. Other impurities in Green River trona include 1–5% shortite, and minor calcite, dolomite, feldspar, sand, and organics [80,81]. Nahcolite-bearing oil shales from the Piceance Basin may contain up to 60 wt. % impurities, dominated by kerogen, clay minerals, quartz, and dolomite [81,82], while dawsonite in the Green River Formation typically occurs disseminated in shale and clay, representing less than 25 wt. % of the total rock [82,83].

More than 80% of the identified occurrences are associated with alkaline lacustrine environments. These include seasonally flooded/evaporative lakes such as Lake Magadi (Kenya) where sodium (bi)carbonate resources are found as buried subsurface layers, lake-bottom or shoreline crusts, as well as surface efflorescences, and dissolved within the aqueous phase [71] (see **Figure 4**). Similar minerals are also found in other lacustrine settings, including perennial or ephemeral alkaline lakes [84], playas or playa-lakes [85], salt pans [86], or paleolakes preserved as deeply buried lacustrine sequences in sedimentary basins (e.g., Green River Formation) [59]. Additional sodium (bi)carbonate-rich localities also occur in volcano (e.g., Trou au Natron, Emi Koussi, and Katwe Crater Lake) and meteor crater lakes [87], salt marshes (e.g., Rhodes salt marsh), hot springs and other hydrothermal systems [88], oases [89], natrocarbonatite volcanoes [90], salt mines [91,92], and subsurface brines [93].

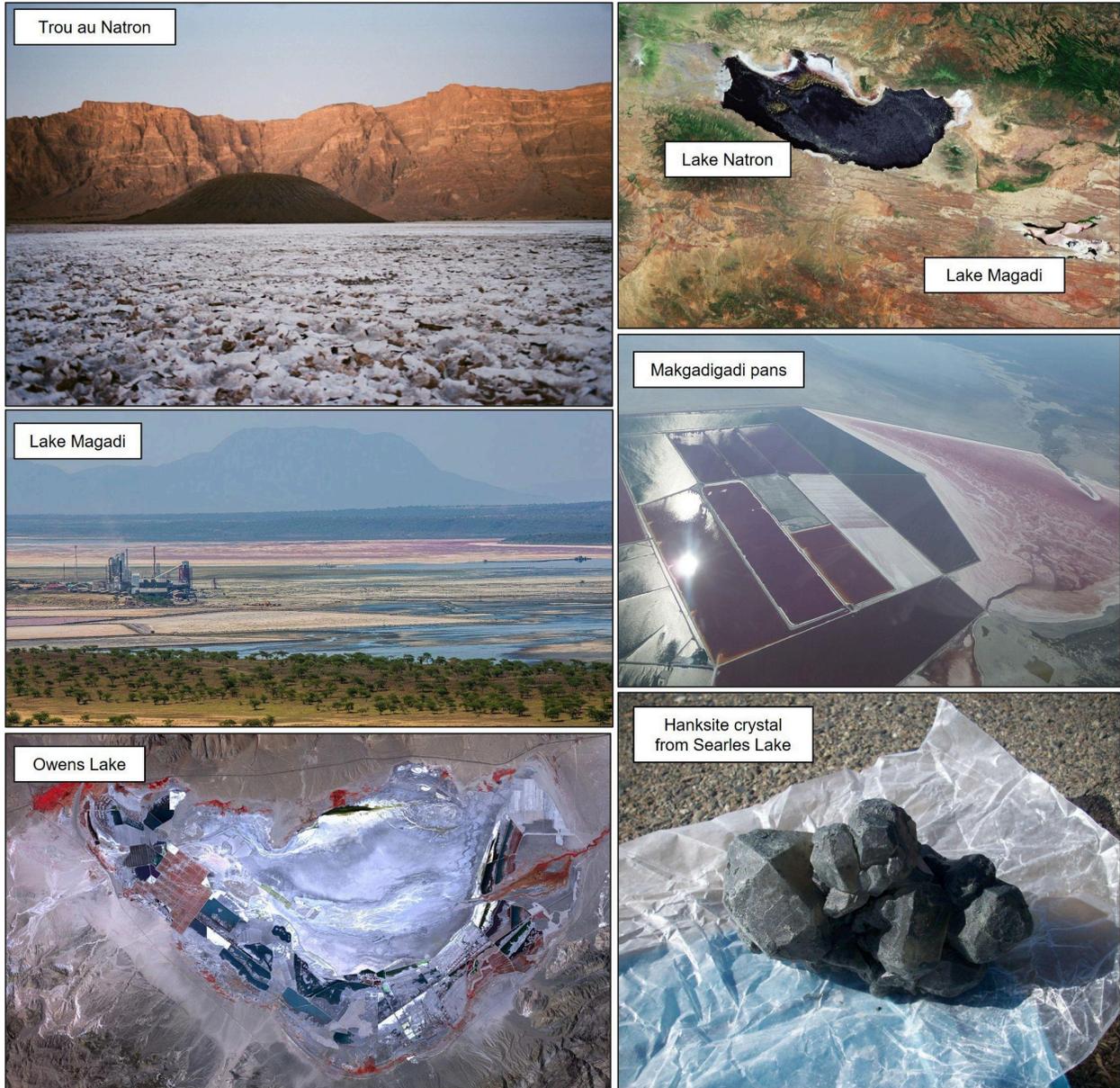


Figure 4 Trou au Natron – A volcano in northern Chad containing natron-rich evaporite minerals [94]; Lake Natron – A hypersaline soda lake in northern Tanzania where high evaporation produces extensive trona and natron crusts [95]; Lake Magadi – A Kenyan rift-valley soda lake known for large trona beds that support long-standing commercial soda ash production [96]; Makgadikgadi Pans – Large salt pans in Botswana that periodically flood and evaporate, depositing sodium carbonate minerals [97]; Owens Lake – a desiccated saline “soda” lakebed whose brines/salts are dominated by sodium carbonate and sodium sulfate, now managed with shallow flooding and other controls to suppress wind-blown dust emissions [98]; Hanksite ($\text{Na}_{22}\text{K}(\text{SO}_4)_9(\text{CO}_3)_2\text{Cl}$) crystal collected from the mud pile field trip on Searles Lake in Trona, California [99].

3.2 Review of current approaches for extraction and processing

Out of 109 occurrences, 12 sites have documented a history of extraction of natural sodium (bi)carbonate minerals (for various uses but mainly for soda ash), and a further 15 are currently active at various extraction rates (making a total of 27 current and historic producers – see **Figure 2** and **Supplementary Table S1**). The total rate of global natural soda ash production was determined to be $>24 \text{ Mt yr}^{-1}$ (not including $>2 \text{ Mt yr}^{-1}$ in the form of baking soda (NaHCO_3)) while capacity continues to grow, particularly in the US and China [100,101]. Natural sources thus contribute $\sim 1/3$ of the world's total soda ash production. **Appendix B** in the **Supplementary Material** provides more information regarding production rates by location and **Figure 4** gives some example sites of natural soda ash harvesting and production.

Trona, and other sodium (bi)carbonate rich materials, can be extracted and valorised via a variety of mining and processing methods. The dominant mining approach in Wyoming is the room-and-pillar method where recoveries are typically around 45% since pillars of trona must be left in place to support the mine structure leading to reserve estimates much lower than the total resources. Thus, of the estimated 116 Gt of trona in the Green River Basin, 36 Gt are reported as reserves i.e., economically recoverable assuming hard rock mining methods [35,57,58,102].

The solution mining method, common in Turkey, is increasingly introduced in Wyoming [103,104]. In solution mining, hot water, containing some dissolved Na_2CO_3 and NaOH , is injected into wells drilled into the trona-bearing formation [105–107]. The additional alkalinity is needed to prevent build-up of a passivating NaHCO_3 layer [105,108,109]. The ore dissolves in situ, and the trona-rich brine is then pumped to the surface for processing.

Solution mining offers several advantages over conventional mining, including increased extraction rates, improved safety (no personnel underground), lower labor intensity/cost, and reduced handling of insoluble impurities [110,111] and may in some cases offer increased recoveries by enabling access to deeper and/or thinner/irregular bodies of ore [57]. Nonetheless, the method requires large amounts of water and energy, and it carries challenges such as managing brine chemistry, and avoiding land subsidence as the ore body is leached. In some cases, both dry and solution mining can be used at one location [112,113].

Once the solid trona ore, or solution, has been extracted, it is typically processed to soda ash via the monohydrate process, which is the predominant method used at both the Green River and Beypazarı sites [44,114]. Briefly, crushed trona is calcined to drive off CO_2 and water and convert bicarbonate to carbonate, then dissolved in water and filtered to remove insoluble matter and the liquor is treated to remove organics. Then evaporative crystallisation to $\text{Na}_2\text{CO}_3 \cdot \text{H}_2\text{O}$ (monohydrate) follows, with centrifugation and drying to produce anhydrous soda ash. In the case where the trona arrives at the processing facility as a solution, the trona brine typically undergoes filtration followed by a steam stripping to decompose the bicarbonate portion, followed by monohydrate crystallisation.

Alternative processing routes for (solutions of) trona are also possible [115] but less common. In the decahydrate route for example, after steam stripping to decompose bicarbonate, sodium carbonate decahydrate ($\text{Na}_2\text{CO}_3 \cdot 10\text{H}_2\text{O}$) crystals can be precipitated by swinging the solution to lower temperature before further processing [116]. Whereas in the sesquicarbonate method, also used in some operations, purified trona solutions i.e., sodium sesquicarbonate ($\text{Na}_2\text{CO}_3 \cdot \text{NaHCO}_3 \cdot 2\text{H}_2\text{O}_{(\text{aq})}$) is precipitated via vacuum evaporative crystallisation and then calcined to light soda ash [117–119]. However, despite certain advantages, the monohydrate method is usually more cost effective than the other routes.

For alkaline lakes settings, which are typically less pure than solution-mined trona, such as those extracted at Lake Magadi, brines are pumped into solar ponds and concentrated via solar evaporation, precipitating out trona (alongside halite or other phases). The crystals are washed to remove some of the NaCl and then calcined to Na_2CO_3 . At other alkaline lake environments, such as Searles Lake, where the lake (and shallow subsurface) brines are more

complex (containing e.g., borates), these are typically processed using the fractional crystallisation method recovering multiple salts, with NaHCO_3 specifically being precipitated out by cold carbonation and then calcined to Na_2CO_3 [120,121].

Although most of current natural soda ash production is from deep bedded trona and surface lake brines, other sodium-rich carbonate and bicarbonate minerals could serve as feedstocks for OAE. For example, approximately 39 Gt of nahcolite (NaHCO_3) resources have been estimated in Colorado's Piceance Basin, with some minor production [60]. This could be thermally upgraded to Na_2CO_3 using analogous pathways to those already discussed here. However, the deposit is rich in oil shale (>50%), making extraction challenging and raising environmental concerns. Dawsonite ($\text{NaAlCO}_3(\text{OH})_2$) is another potential resource whereby it can be decomposed at $\sim 400^\circ\text{C}$ to yield NaAlO_2 , CO_2 , and H_2O [122], and the NaAlO_2 can subsequently be hydrolysed with hot water to produce NaOH and $\text{Al}(\text{OH})_3$ [123], making dawsonite a potential dual source of alkalinity and aluminium. Other sodium carbonate minerals such as shortite, eitelite, gaylussite, pirssonite, and northupite, could also be investigated, but are likely rarer and/or more complex to process, with lower solubility in seawater.

3.3 Gross CO_2 removal potential of sodium (bi)carbonate minerals by OAE

Table 1 summarises the theoretical (maximum) CDR potentials of several naturally occurring sodium (bi)carbonate minerals (e.g., natrite, thermonatrite, natron, nahcolite, and trona) when dissolved in seawater at 11°C , and equilibrated with air. For context, we also include aqueous sodium hydroxide ($\text{NaOH}_{(\text{aq})}$) which can be produced by other OAE technologies such as brine electro dialysis (~ 4 wt. %) and electrolysis (~ 30 wt. %) e.g., of desalination brines or seawater itself. CDR potentials for ocean liming via slaked lime ($\text{Ca}(\text{OH})_2$) and coastal enhanced weathering via olivine (e.g., here approximated as pure forsteritic olivine) are also given.

Most noticeably, the addition of nahcolite to the ocean cannot be considered CDR as it leads to a net release of 0.068 tonnes of CO_2 per tonne of applied mineral. Nahcolite dissolves according to:



To re-establish equilibrium within the seawater carbonate system, part of the added bicarbonate is converted via:



Thus, even though adding NaHCO_3 increases alkalinity and pH, it shifts the carbonate speciation toward $\text{CO}_{2(\text{aq})}$, leading to a net positive CO_2 flux to the atmosphere. By contrast, natrite (or soda ash when describing the industrial product), results in a CO_2 removal of $0.307 \text{ tCO}_2 \text{ t}^{-1}$, since a portion of the CO_3^{2-} reacts with $\text{CO}_{2(\text{aq})}$ and H_2O to form HCO_3^- . Trona, being composed of equal parts bicarbonate and carbonate, has a CO_2 removal intermediate to that of nahcolite and natrite ($0.156 \text{ tCO}_2 \text{ t}^{-1}$), consistent with its mixed stoichiometry.

Beyond simple equilibrium thermodynamics, other material properties will play a role in the rate at which the CDR potential is achieved (kinetics), and will include factors such as the type and quantity of impurities present (impurities may counter alkalinity generation in seawater e.g., by enable secondary mineral precipitation), solubility of the solids, particle size and density (e.g., light vs. dense soda ash). Where natural sodium (bi)carbonate minerals exhibit low intrinsic CDR potential or contain high levels of impurities, transformation and/or purification can increase their effectiveness. For example, soda ash/trona properties e.g., density, porosity, and particle size can be tailored through appropriate engineering. However, the rate and extent of CO_2 removal during OAE is only one part of the overall assessment. Valorisation and purification pathways, in particular calcination, are energy intensive and likely have high carbon footprints (upstream life cycle carbon emissions). Therefore, meaningful comparison of the different options requires evaluating their process emissions, and thus their net CDR performance, which is the focus of the next section.

Table 1 Maximum theoretical CO₂ removal efficiencies for different materials in seawater determined using the multicomponent geochemical model code PHREEQC [47]. Seawater composition and temperature were taken as the mean of annual data representative of the coastal regions near Portland, USA [48].

Chemical name	Chemical formula	Mineral name	Molar mass	Solubility in water (20°C)	Maximum theoretical CO ₂ removal in seawater		OAE technology
			g mol ⁻¹	g L ⁻¹	mol CO ₂ mol ⁻¹	tCO ₂ t ⁻¹	
Sodium sesquicarbonate dihydrate	Na ₂ CO ₃ ·NaHCO ₃ ·2H ₂ O	Trona	226.03	130	0.770	0.156	Pathway 1
Anhydrous sodium carbonate	Na ₂ CO ₃	Natrite	105.99	215	0.740	0.307	Pathway 2 & 3
Sodium carbonate monohydrate	Na ₂ CO ₃ ·H ₂ O	Thermonatrite	124.00	210–250	0.740	0.263	
Sodium carbonate decahydrate	Na ₂ CO ₃ ·10H ₂ O	Natron	286.14	210	0.740	0.114	
Sodium bicarbonate	NaHCO ₃	Nahcolite	84.01	99.6	-0.130	-0.068	
Sodium hydroxide (dry)	NaOH		40.00	>1000	0.870	0.958	
Sodium hydroxide (solution)	NaOH _(aq) (~4% by wt.)				0.870	0.038	Electrodialysis of NaCl
Sodium hydroxide (solution)	NaOH _(aq) (~30% by wt.)				0.870	0.287	Electrolysis of NaCl
Calcium hydroxide	Ca(OH) ₂	Portlandite	74.09	1.86	1.74	1.033	Ocean liming
Dimagnesium silicate	Mg ₂ SiO ₄	Forsterite	140.69	Insoluble	3.456	1.081	Coastal weathering

3.4 Life cycle assessment

The life cycle carbon emissions of the three examined pathways are shown in **Figure 5**, where it can be seen that all pathways are net-negative, with pathway 2 (294 kg CO₂ emitted per t of CO₂ removed or carbon penalty of 29.4%) having overall the lower carbon penalty and pathway 3 (406 kg emitted per t of CO₂ or 40.6% carbon penalty) the highest. We note that the results are dependent on significant strides in introducing large-scale methane flaring during mining, introducing CCS during trona calcination (pathway 2 and 3), and using electrical freight trains for transportation, all of which are not currently applied at the case study location. However, these remain realistic considerations as the industry is already moving towards such approaches. For example, the soda ash industry is already moving towards solution mining (currently accounting for 7% of the global production) which not only restricts methane fugitive emissions but is also less energy intensive with plans to reach net-zero by 2050 [124]. Furthermore, worldwide train infrastructure is switching to electrified rail lines, and, even though barriers persist, in the USA freight railway electrification is already under consideration [125]. Finally, for this technology to realise CDR, CCS at scale will be required, likely using oxy-fuel calciners, as is the case for other ocean CDR approaches that use thermal decomposition of carbonate minerals, such as ocean liming [18]. We note that as the industry moves towards solution mining the problem of methane fugitive emissions can be effectively minimised while CCS is already introduced at scale. With these considerations, the LCA modeling revealed that trona mining and transportation are the main carbon hotspots across the considered pathways. Specifically, the energy intensive underground mining contributed 35.1%, 32.6%, and 24.9% of the total life cycle emissions of pathway 1, 2, and 3 respectively. The large contribution in pathway 1 mainly traces back to the much larger mass of material needed for the gross removal of 1 t of atmospheric CO₂ (the functional unit of the LCA), i.e., 7.05 t of trona. On the other hand, mining's much smaller contribution (24.9%) in pathway 3 does trace back to the significantly lower mass that is needed to achieve this gross removal, together with the high total carbon footprint attributed to trona processing, which lowers mining's contribution to the total carbon footprint. Specifically, for pathway 2 and particularly pathway 3 direct (natural gas) and indirect (electricity) use of fossil fuels to drive the thermal decomposition processing greatly add to the corresponding carbon footprints (96.8 and 214 kg CO₂ respectively). Perhaps surprisingly, the least energy- and technologically-intensive approach, i.e., pathway 1, exhibited a higher carbon penalty than pathway 2 which entailed thermal decomposition. This is due to the fact that even though this pathway is relatively simple it requires significantly more mass per functional unit, due to the lower CO₂ removal potential of trona, which greatly increases the mining, transportation, and handling and spreading emissions.

Overall, these initial LCA results suggest the net-negativity of the process, but also the need to decarbonise transportation, electricity, and supply chains for this technology to be considered a meaningful CDR approach. There are also opportunities for further improving the net-negativity of the examined approaches. For example, methane flaring can greatly reduce mining's emissions [126], however, its use for heat production instead of flaring could also drastically reduce pathway's 2 and 3 carbon footprints. Furthermore, here, electricity from the current fossil fuel-dependent electricity grid was considered, implying that there is further room for improvement if renewable energy was used. Finally, although these routes may not currently be competitive with the monohydrate route, financial incentives to decarbonise the chemicals industry may shift the relative economics of different Na₂CO₃ production routes, as has been suggested for other sectors like mining and steel [127,128]. Indeed, methods which combined chlor-alkali electrolysis, direct air capture, and sodium carbonate crystallisation to produce synthetic carbon-negative soda ash could be applied to processing of trona [129]. Furthermore, Na₂CO₃ for OAE may be more viably produced with lower energy requirements via sesquicarbonate, decahydrate, or cold-carbonation routes, rather than the monohydrate route, given that OAE does not place strict requirements on the purity or other properties of the final product [130]. For example, dense soda ash (1000–1100 kg m⁻³ [131]), as produced by the monohydrate method, is typically needed for glassmaking. However, light soda ash (~500 kg m⁻³ [131]), produced by methods like the sesquicarbonate method, dissolves more quickly and may be better suited for OAE.

Sub-process Contribution to Total Carbon Penalty

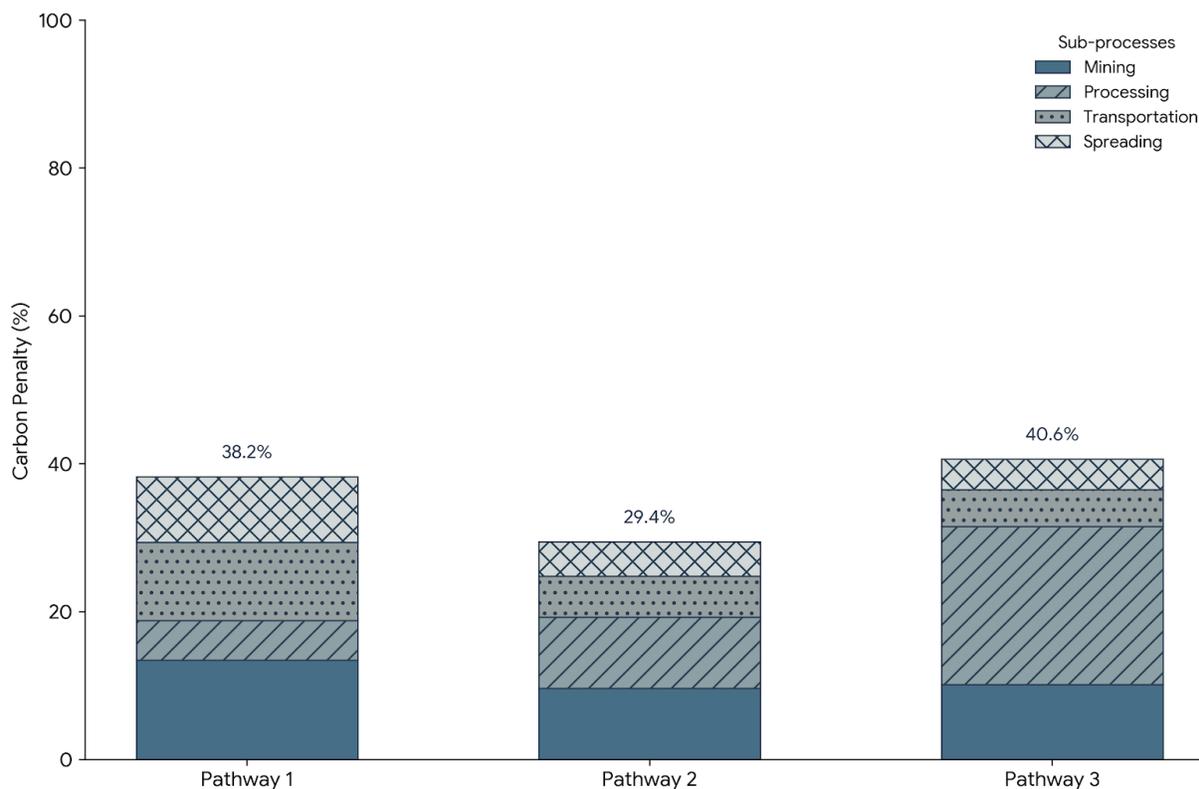


Figure 5 Carbon penalty for the three suggested OAE pathways (Pathway 1: direct trona addition (7.05 t per gross t CO₂ removed); Pathway 2: impure soda ash (3.68 t tCO₂⁻¹); Pathway 3: pure soda ash (3.23 t tCO₂⁻¹)) using trona from Green River, Wyoming. The carbon penalty is calculated as the total CO₂ emitted during mining, processing, transportation, and spreading, per gross ton of CO₂ removed via OAE.

3.5 Preliminary cost assessment

In the US, the average realised price of high-purity soda ash produced from naturally occurring deposits was ~\$208 t⁻¹ in 2024 [35], rising to ~\$266 t⁻¹ by mid-2025 [132]. (In contrast, in Europe, specifically Germany, where soda ash is typically produced synthetically via the Solvay process, prices were around \$356 t⁻¹ in 2025). Globally, reported prices span roughly \$170–370 t⁻¹ depending on region and product specification [133]. Production costs also vary with technology and location. In the US, conventional mining and processing of trona is estimated at ~\$100 t⁻¹ [133] and slightly lower in China [134]. In Turkey, where the solution-mining method dominates, costs are ~\$50 t⁻¹. (In Europe, Solvay-based production costs have been estimated at ~\$150 t⁻¹ [133]). The soda ash production costs detailed above equate to minimum costs for gross CO₂ removal on the order of \$70–500 tCO₂⁻¹.

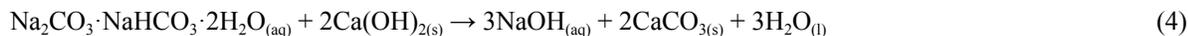
Although a detailed technoeconomic assessment is beyond the scope of this feasibility study, some indicative cost estimates are outlined. Assuming impurities in trona/soda ash have minimal environmental impact, it may be possible to bypass the energy-intensive monohydrate purification step entirely, substantially reducing costs and carbon penalties of pathway 1 and 2. Although solution mining was not considered herein, it may produce a trona-brine with fewer insoluble impurities than conventional room-and-pillar mining (which produces solid trona). This could enable production of higher purity of trona or soda ash while still avoiding the energy intensive monohydrate purification.

Using the solution-mining flowsheet of Nasün-Saygılı and Okutan (1996), two ocean alkalinity delivery options were compared: a dilute, carbonate-rich “crystalliser feed” liquor (28 wt% Na₂CO₃) and a solid Na₂CO₃·H₂O monohydrate (84.3 wt% Na₂CO₃). Because of the large water fraction in the liquor, the gross CO₂ removal per tonne delivered is much lower for liquor (0.08 tCO₂ t⁻¹ solution, or 0.22 tCO₂ t⁻¹ on a solids-equivalent basis) than for monohydrate crystals (0.35 tCO₂ t⁻¹), resulting in a mass-handling and transport penalty. A screening cost estimate for a 679,500 t yr⁻¹ plant (costs adapted from Nielsen 1983 and escalated to 2024 USD) finds downstream processing dominates capex/opex, motivating a “once-through” concept that avoids evaporation/crystallisation but adds pipeline/pumping costs, yielding a levelised cost trade-off between lower processing energy and higher water transport. Minimum cost estimates of <\$100–200 per gross tCO₂ removed are estimated (see **Figure D1**), and controlled primarily by the costs of sodium hydroxide used in solution mining. This order of magnitude is broadly comparable with reported cost ranges for other CDR technologies [135].

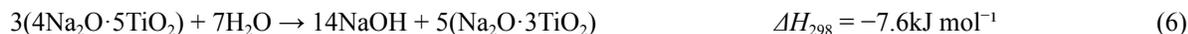
This work further indicated that trona-based OAE has potential using current supply chains, and can be cost-competitive provided upstream energy and process emissions remain controlled. Future opportunities can therefore focus on process/system choices that improve removal efficiencies or reduce life-cycle emissions. These opportunities fall into three overlapping categories: (i) lowering embodied energy and emissions by simplifying or bypassing purification steps (as shown with pathway 2), (ii) increasing delivered alkalinity (and hence CO₂ uptake) per tonne handled by shifting product form and reaction pathway (see efficiency values in Table 1), and (iii) reducing the carbon intensity of heat and power inputs.

3.6 Exploring other valorisation pathways for sodium (bi)carbonates

Some alternative processes for trona valorisation are proposed for exploration hereon. For example, soda ash could be converted to materials with greater CO₂ removal potential, notably NaOH (1 ton of dry NaOH can remove ~1 ton of CO₂ – see **Table 1**). One approach is to causticise dissolved trona using slaked lime:



Causticisation is already used in solution-mining operations, where NaOH is employed to raise brine alkalinity and facilitate dissolution of the bicarbonate fraction of trona [136,137]. However, this route generates CaCO₃, which must be regenerated in a kiln at high temperature. Moreover, the aqueous NaOH product would need to be transported to the ocean, something that is unlikely to be viable for sites further inland. Alternatively, solid (high purity) Na₂CO₃ could be transported to the coast and converted to NaOH in coastal plants via direct causticisation [138,139]. This transformation can be achieved using solid-state metal oxides e.g., the titanate cycle [139–142]:



NaOH can also be regenerated from Na₂CO₃ using oxides such as NaBO₂, Fe₂O₃, or Mn₂O₃ (and water). The Mn₂O₃-based approach is particularly interesting because reported calcination temperatures may be as low as ~600°C, potentially lowering thermal energy demand relative to titanates (~820°C) [143]. However, the viability of direct causticisation would likely depend on the cycling capacity over repeated cycles, something that is compounded by the build up of impurities.

Electrochemical approaches should also be investigated as alternatives to thermal trona valorisation. Sodium (bi)carbonate solutions can be converted to NaOH using electrolysis or bipolar membrane electrodialysis, producing NaOH and CO₂ directly at low to moderate temperatures [46,144–147]. In one electrodialysis configuration, Na⁺ is transported across a cation-exchange membrane to generate high-purity NaOH, while carbonate is acidified at the

anode, creating CO₂, and potentially other co-products such as H₂ and O₂ [45]. These routes could reduce thermal energy demand and provide a pathway to integrate renewable-electricity.

While NaOH offers higher unit CO₂ removal potential than Na₂CO₃ or trona, it also introduces substantial engineering and logistical challenges. An alternative research direction is the conversion of trona into solid compounds with higher alkalinity release potential, such as sodium ferrite (NaFeO₂) or sodium metasilicate (Na₂SiO₃). These may be synthesised by calcining finely crushed trona or sodium carbonate with Fe₂O₃ [148–150] or SiO₂ [151,152] at high temperature (>900 °C), e.g.:



When introduced to the ocean, NaFeO₂ hydrolyses, releasing alkalinity, e.g.,



However, at present, the environmental implications of introducing reactive sodium ferrites or silicates remain uncertain.

3.7 Scaling potential

Despite the scale and technological maturity of existing trona extraction and processing operations, such resources have been previously dismissed as too limited for large-scale OAE applications [32]. However, our results suggest that, although sodium (bi)carbonate evaporites are less abundant than other alkaline mineral resources such as limestone (CaCO₃) or ultramafic rocks (e.g., olivine ((Mg,Fe)₂SiO₄)), their global quantity still likely exceeds 200 Gt of resources.

Scaling to 1 GtCO₂ yr⁻¹ of CDR would require on the order of approximately 3.3 Gt yr⁻¹ of treated material (assuming 0.3 tCO₂ t⁻¹). Following a logistic scaling pathway (**Figure 6**, see Lee Pereira et al., 2025), and scaling-up and sustaining this industry for a century, would require cumulatively between 210 and 290 Gt of material. Whereas, scaling to 0.5 GtCO₂ yr⁻¹ would require between 110 and 150 Gt of material. A 200 Gt trona deposit equates to a 140 Gt of pure sodium carbonate. As such, currently identified resources could supply a 0.5 GtCO₂ yr⁻¹ OAE industry, scaling to 1 GtCO₂ yr⁻¹ would be possible if additional resources were identified.

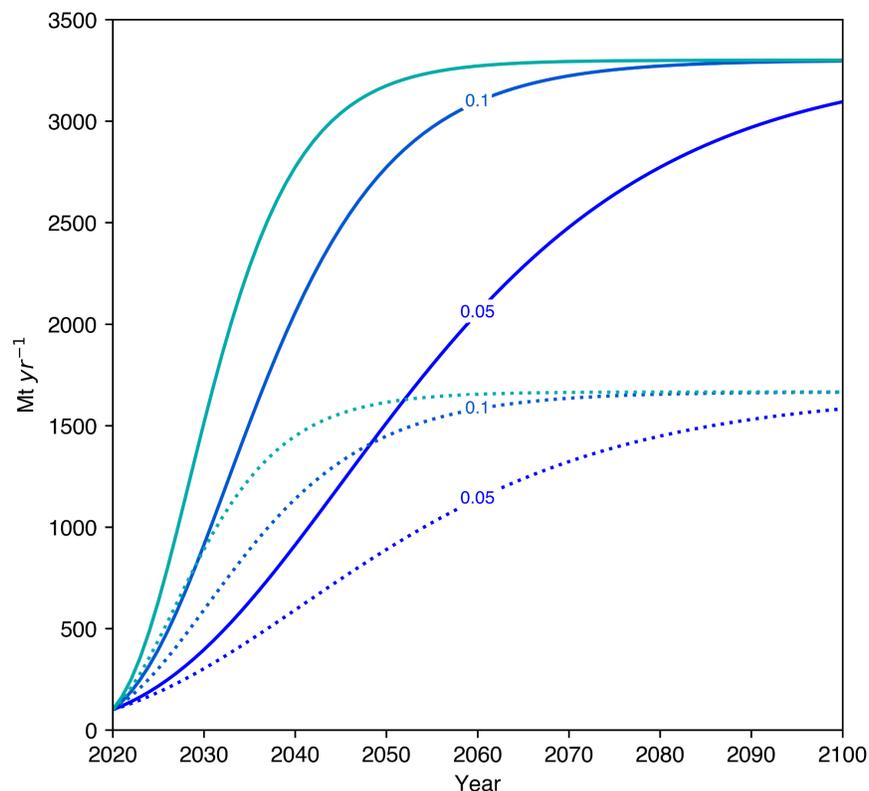


Figure 6 A scaling pathway for sodium carbonate ocean alkalinity enhancements for a removal of $0.5 \text{ GtCO}_2 \text{ yr}^{-1}$ (dotted lines) or $1 \text{ GtCO}_2 \text{ yr}^{-1}$ (solid lines). The colour of the lines denotes the upscaling rate (from 5–15%, labelled on the lines) following the approach outlined in Lee Pereira et al. (2025) [39].

Deposits likely remain undiscovered particularly in Africa and Australia [93,153–155], high-altitude regions in the Central Andes, Bolivia, and the Tibetan Plateau are also known to contain hundreds of saline and alkaline lakes [156], being plausible targets for further exploration. Indeed some deposits may have been overlooked for soda ash production due to high impurities contents. Such low grade ores could be useful in the context of OAE, where feedstock purity requirements (for salt) are not necessarily a strict limiting factor compared to common industrial use cases for soda ash. What is more, sodium (bi)carbonates produced from alkaline-lakes are sometimes replenished. For example, at Sua Pan (Makgadikgadi) in Botswana, where brines are extracted into evaporative ponds to produce NaCl and Na_2CO_3 , there is some evidence of replenishment thus complicating reserve estimates at such sites [157].

Regardless of the true magnitude of the resource base, key factors such as mineralogy and purity, accessibility, distance to the ocean (and associated transport type), available energy and material inputs for extraction and processing, as well as environmental constraints and public acceptance will ultimately determine whether natural sodium (bi)carbonate deposits are viable feedstocks for OAE. Our preliminary resource inventory and life-cycle assessment suggest that trona-based OAE could be both sustainable and scalable, but meaningful comparison across the many plausible feedstocks, process configurations, supply-chain routes, deployment locations, and energy-system contexts remains difficult without consistent, rigorous assessment. Addressing this requires comprehensive techno-economic analysis coupled with prospective life-cycle assessment under harmonised assumptions. However, relatively few OAE concepts have been evaluated in this way to date. Applying structured frameworks such as Lee Pereira et al. (2025) [39] would enable clearer benchmarking across OAE pathways and

against other carbon dioxide removal options, supporting more decision-relevant outcomes for investment, policy, and research.

4. Conclusion

This study identified naturally occurring sodium (bi)carbonate evaporites as a credible, underexplored feedstock class for OAE at a meaningful scale. Through a new global inventory, we map 109 occurrences worldwide, while noting that quantitative resource estimates remain available for only a minority of sites. Nevertheless, the compiled literature indicates a resource base of 124 Gt of bedded trona plus ~84 Gt of closely related sodium (bi)carbonate minerals (and brines), implying >200 Gt of potentially accessible material. Assuming all known global bedded trona resources (124 Gt) were added directly to the ocean (without generation of process emissions) a maximum of 18 Gt of CO₂ could be removed from the atmosphere. However, by valorising to Na₂CO₃ a total of 26 Gt could be removed from the atmosphere. Valorisation of global resources of nahcolite, dawsonite, and sodium (bi)carbonate brines, could add further removals at the Gt-scale. On top of this, our work suggests carbon penalties of between 30–40%. Thus, we find that sodium (bi)carbonate-based OAE represents a substantial CO₂ removal pathway, while likely offering relatively benign environmental impacts compared with alternatives such as ultramafic rock weathering. We therefore propose that direct addition of naturally occurring sodium (bi)carbonates and their valorised products be included in the current portfolio of OAE methods, alongside coastal enhanced weathering, ocean liming, and electrochemical approaches. However, robust prospective technoeconomic assessment is required to quantify cost trajectories, and compare these with alternative approaches. Finally, many alkaline-lake and evaporitic basin environments are ecologically sensitive and socially contested, such as Lake Magadi [158]. Thus, new projects using such resources should consider integrating early stakeholder engagement to secure public acceptance and social licence.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Authors credit statement

James Campbell: Conceptualisation; Data curation; Formal analysis; Investigation; Methodology; Validation; Visualisation; Writing – original draft; Writing – review & editing., Spyros Foteinis: Formal analysis; Investigation; Methodology; Validation; Writing – original draft; Writing – review & editing., Reinaldo Lee Pereira: Validation; Visualisation; Writing – review & editing., Mohamad Katish: Validation; Writing – review & editing., Phil Renforth: Formal analysis; Investigation; Methodology; Funding acquisition; Project administration; Validation; Writing – original draft, Writing – review & editing.

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Supplementary Material

Appendix A – Additional detail on LCA methodology

Green River trona is extracted using a room-and-pillar mining method with electrically powered continuous miners [1,2]. Each continuous miner is estimated to consume approximately 0.7 kWh per ton of trona mined, based on a power rating of 1,472 kW, a 70% load efficiency, and an operational rate of 500 t h⁻¹ [2,3]. Electric shuttle cars (170 kW, 15 t capacity) [4] transport the ore from the mining face to the main conveyor belt system (1,492 kW with a throughput of 900 t h⁻¹) which carries trona ore to the surface for processing [5]. Furthermore, room and pillar mining of trona is notorious for fugitive methane emissions, which greatly vary from 0.87 to 2.01 Mm³ CO₂e annually in the USA [6]. However, with proper management, such as methane flaring or valorisation for heat production, fugitive emissions can be minimised, e.g., the methane destruction efficiency of enclosed flaring is 99.5% [7]. Given that relevant LCI data for room and pillar trona mining were not identified in SimaPro databases, we updated and used as proxy LCI data for underground mining of hard coal, as this dataset not only captures associated energy inputs but also includes additional processes such as blasting and also the relevant machinery such as the conveyor belt system. Specifically, we used theecoinvent process “hard coal mine operation, underground ZA” which provides average inputs for board-and-pillar and longwall mining. Given that this process refers to a different spatial extent and material, the data that were not relevant such as the emissions from hard coal mining were excluded, while the relevant main inputs such as energy and water consumption were updated using data from the Western Electricity Coordinating Council (US-WECC), where Wyoming belongs to, while the 10% spoil (treated as shale) was also considered for the run-of-mine (ROM) trona. For the associated methane emissions we considered the higher end of the range for the Green River formation but considered that 95% of this is destroyed through flaring. This scenario is considered realistic as the industry is slowly moving away from the carbon intensive underground mining to the greener solution mining [8] which is also associated with significant reductions in methane emissions. The main inputs of this process are listed in **Table A1**.

The next stage common to all pathways is comminution. The energy input for loading the mined trona was taken from [9,10], while the crushing plant was assumed to be located within the trona mine, i.e., no transportation was ascribed to comminution. Furthermore, not all pathways employ the same particle size, so electricity consumption differs between pathways. Specifically, for Pathway 1 we consider a particle size of 50 µm, while for the two rest pathways the particle size was assumed to be 100 µm. (A trona particle diameter of <1 mm is typical for effective soda ash calcination in the monohydrate process [11]). However, for direct ocean spreading (Pathway 1) trona and soda ash solids must be sufficiently soluble in the surface ocean and thus smaller sizes are needed based on solubility [12], as is the case with ocean liming [10]. (A 1.5 mm Na₂CO₃ particle dissolved in well under a few minutes, with the dissolution rate increasing at higher temperatures and lower pH [13].) Here, the energy consumption to reach these sizes through comminution was estimated using Bond’s equation [14]. Since the Bond work index for trona is not reported, a conservative value of 11 kWh t⁻¹ was adopted, consistent with materials of similar hardness of other evaporites such as gypsum (~7 kWh t⁻¹, Mohs hardness ~ 2–2.5). For all pathways, the feed particle diameter was assumed to be 10 cm. Here, the energy input for comminution was considered as electricity from the US-WECC grid, along with proxy values for water use for cooling and dust suppression [15–17] and its pumping using a diesel driven pump. For Pathway 2 and 3, trona is assumed to be calcined in a kiln at 180°C following Equation (1) [18–21]. It was assumed that the heat to drive trona decomposition is provided by oxyfuel combustion of natural gas (lower heating value of 50 MJ kg⁻¹), which provides high purity carbon dioxide (CO₂) output that is required in carbon capture and storage (CCS) assumed herein. Specifically, the process of a natural gas-driven low-NO_x industrial furnace was updated to the case study conditions (US-WECC), while the O₂ input was assumed to originate from a cryogenic air separation unit (again using electricity from the US-WECC grid). After condensation of water vapour, a pure stream of CO₂ (>98%) is assumed to be obtained. A conservative CO₂ capture fraction of 0.95 was considered [22], assuming that the CO₂ emissions from natural gas burning are captured along with the emissions from the chemical decomposition of the trona matrix (Equation (1)). The CO₂ is

compressed to 110 bar, with an energy consumption of 120 kWh tCO₂⁻¹ [23] and subsequently is assumed to be transported approximately 100 km by pipeline (0.25 kWh km⁻¹ tCO₂⁻¹) [24] to the Sweetwater Carbon Storage hub where it is injected into deep saline aquifers [25]. For CO₂ capture and transportation to the storage site the energy input was considered as electricity from the US-WECC grid.

For the production of high-purity Na₂CO₃ considered in Pathway 3, we adopt a simplified version of the monohydrate process. Specifically, calcined trona is first dissolved in hot water at 60°C [26] to a concentration of 30% by wt. [27]. Insoluble impurities are removed by filtration. The dissolver temperature is maintained primarily by heat recovered from the hot solids and the exothermic heat of dissolution, with any additional requirement met by waste steam. The solution is fed into a multi-effect evaporative crystalliser operating at 100 °C (specific heat consumption of 3.2 GJ tNa₂CO₃⁻¹) where Na₂CO₃·H₂O crystals form [2]. Approximately 6% of the Na₂CO₃ is assumed lost in the purge stream containing soluble impurities such as NaCl [28]. The monohydrate crystals are then steam-dried in a fluidised bed at 100 °C to produce high purity (>99%) Na₂CO₃. All process heat and steam requirements are assumed to be supplied by oxy-fuel combustion of natural gas, with CO₂ capture and storage as described previously.

The solid products of Pathways 1 (trona, purity ~90%), 2 (Na₂CO₃, purity ~88%), and 3 (Na₂CO₃, purity >99%) are (loaded and) transported for approximately 1400 km by the existing rail link to Port of Vancouver (Washington, USA) [29]. Due to the long distance, our initial analysis revealed that for the process to be net-negative, and have a meaningful carbon dioxide removal (CDR) capacity, the existing diesel-powered freight trains should be replaced with electric trains. For this reason, the use of an electric train (hilly terrain and with 50% load factor i.e., empty return) was considered when operated with electricity from the local electricity mix (US-WECC). When the material reaches the port this is unloaded and loaded into ships, since here we conservatively assumed that ship spreading will be used regardless of the processing pathway using an approach similar to ocean liming [9,10]. Both the energy consumption for loading and for dockside operation was considered, while for spreading a bulk carrier ship was considered assuming a low discharge rate (25 kg s⁻¹ with a cruising speed of 25 km h⁻¹) [30]. Finally, maximum theoretical CO₂ removal efficiencies for different materials in seawater were determined using the multicomponent geochemical model code PHREEQC [31]. The seawater composition was taken as the mean of average yearly data representative of the coastal regions near Portland, USA [32].

Table A1 LCI data for three OAE pathways normalised per FU, i.e., the uptake of 1 t of CO₂ from the atmosphere when the material (trona or soda ash) is spread in the ocean.

Name	Unit	Pathway			Citations
		1	2	3	
<i>General inputs</i>					
Reactive material		trona	Na ₂ CO _{3(s)}	Na ₂ CO _{3(s)}	
Molar mass	g mol ⁻¹	226.03	105.99	105.99	
CO ₂ removal efficiency (pure)	t CO ₂ t ⁻¹	0.156	0.310	0.310	Determined in PHREEQC
Mass reactive material needed	t	6.41	3.23	3.23	
Particle size on spreading	µm	50	100	100	
Trona impurities total	% wt.	10	10	10	

Trona impurities (soluble)	% wt.	1	1	1	[33]
Trona impurities (insoluble)	% wt.	9	9	9	[33]
Purity of transported material	%	91	88	100	
Mass trona mined	t	7.05	5.04	5.30	

Mining and crushing

Loading	MJ	56.410	40.35	42.37	[10]
Blasting	kg	0.120	0.086	0.091	Ecoinvent
Conveyor belt	m	0.0176	0.0125	0.0132	Ecoinvent
Mine infrastructure (underground)	p	1.18E-07	8.42E-08	8.85E-08	Ecoinvent
Diesel	MJ	91.65	65.52	68.90	Ecoinvent
Electricity	kWh	103.64	74.09	77.91	Ecoinvent
Shale	t	0.78	0.55	0.58	Ecoinvent
Spoil treatment and handling	t	0.78	0.55	0.58	Ecoinvent
Methane emissions	kg	39.35	28.15	29.56	Own estimate
Grinding energy (Bond's equation)	kWh	83.46	41.65	43.73	Work index = 11 kWh t ⁻¹

Transformation

Heat consumption

Trona calcination (@180°C)	kWh	0	537.0	563.9	Own estimate
Evaporative crystalliser (steam)	kWh	0	0	987.53	Own estimate
Thermonatrite dehydration	kWh	0	0	471.74	Own estimate

Electricity consumption

CO ₂ compression	kWh	0	55.25	89.72	[23]
CO ₂ transport from Green River to Sweetwater	kWh	0	13.81	22.43	[24] Assumed 100 km distance

Materials consumption

O ₂ from ASU	t	0	0.16	0.61	Ecoinvent
Water for dissolution	m ³	0	0	8.83	Own estimate
Quicklime	kg	0	0	39.79	from SimaPro
Activated carbon	kg	0	0	0.65	[34]

Transportation and spreading

Load mass	t	7.05	3.68	3.23	
Distance (mine to port)	km	1416	1416	1416	Google Earth
Train loading	MJ	56.41	29.48	25.81	[10]
Freight transport	t km	9985	5217	4568	
Unloading/Ship loading	MJ	56.41	29.48	25.80	[10]
Dockside operation	MJ	133.97	70.00	61.29	[10]
Ship transport	t km	1.975	1.032	0.904	[30]
Ocean spreading	MJ	56.41	29.48	25.81	[30]

Appendix B – A brief history of sodium (bi)carbonate minerals extraction

Natural sodium carbonates have been used since ancient Egypt, as early as the 4th millennium BC. “Natron”, or more likely trona, was extracted from the shores and bottom crusts of saline lakes at Wādī al-Natrūn, northwest of Cairo (where the term “natron” is derived) [35]. The sodium (bi)carbonates were used as a desiccant and preservative (e.g., in mummification), among other uses. By the first century CE, minerals from lakes in these same regions were used by the Romans in glassmaking and medicine [36]. However, over the coming centuries, demand for glass grew to the point such that production from saline lakes was insufficient, thus triggering a switch to burning of Na-rich plant biomass (e.g., kelp) to supply the necessary Na_2CO_3 (the origin of the term “soda ash”).

Elsewhere in Africa, trona and related minerals were also extracted at Lake Natron, Tanzania and Lake Chad, (e.g., Niger) for use in cooking, including as an additive to porridge consumed by nursing mothers, which is believed to increase the quality and quantity of breast milk [37,38]. Similar uses of naturally occurring sodium (bi)carbonate minerals are documented in ancient Mesoamerican cultures. For example, the mineral “tequesquite”, which is a mixture of sodium (bi)carbonate and sodium chloride, was extracted from Lake Texcoco (a saline alkaline lake now covered by Mexico City), for applications like cooking and cleaning [39–41]. This same resource was exploited in 1942 when the Sosa Texcoco company was established to pump underlying brines into solar evaporation ponds, followed by carbonation and calcination to form soda ash, as well as using a lime cycle to produce caustic soda [42,43]. The plant produced as much as 200 kt of alkali products each year, mainly for soap-making, before eventually closing in 1987.

The first natural soda ash production in the US took place in 1868 at Little Soda Lake (and later Big Soda Lake) in Lahontan Valley, Nevada, producing a few hundred tonnes per year [44–46]. In 1887, a similar operation began at Owens Lake, California, where lake brines were pumped into evaporation ponds, enabling seasonal trona precipitation and harvesting [47–51]. Operational efficiency was later improved by carbonating the brines with CO_2 to precipitate NaHCO_3 , which was then calcined to soda ash [52]. Additional small-scale operations in the US occurred at Double Springs Marsh (Schurz, Nevada) (1886–1917), Union Pacific Lakes (near Laramie, Wyoming) (1895–NA), Carbonate Lake (Warden, Washington) (1927–1941), Mitchell Lake (Wilson Creek, Washington) (1936–1941) [47].

Smaller operations, including Owens Lake, eventually closed upon discovery in the 1930s of giant underground deposits of trona ore in the Green River Basin, Wyoming [53]. Cores taken during a period of extensive oil exploration revealed thick layers (up to ~11 m) of trona buried at 180–900 m depth with areas of 130–2200 km^2 [54,55]. Westvaco Chemical Corporation began mining in 1946 [56], and since then more than 600 Mt have since been extracted [47], in what is now the largest ongoing extraction site of naturally occurring sodium (bi)carbonate minerals. Aside from Wyoming trona, the only major US natural soda-ash operation based primarily on lake (and lake subsurface) brines is in Searles Lake, California, which began commercial soda ash production in 1931 and continues to operate today (Kostick, 1998).

Appendix C – Soda ash production by location

The largest producer of natural soda ash is the US at $\sim 12 \text{ Mt yr}^{-1}$, with the capacity to produce 14 Mt yr^{-1} [57]. In Green River Wyoming, total production by four different companies was $\sim 11\text{--}12 \text{ Mt yr}^{-1}$ of soda ash from deep trona mines (corresponding to $\sim 20 \text{ Mt yr}^{-1}$ of trona) [57]. In California, the only major lake-based soda ash operation (Searles Lake), produces $>1 \text{ Mt yr}^{-1}$ of soda ash, alongside various other salts including sodium bicarbonate, borax, sodium sulfate, and sodium chloride, by fractional crystallisation [58–61] but has recently announced plans to idle its operations due to the rising cost of energy [62]. Together these natural resources meet most of the US domestic demand for soda ash, in addition to substantial exports.

Turkey is another world-leading soda ash producer. Trona was identified during coal exploration in the Beypazari and Kazan basins near Ankara in the late 1970s and production was developed using solution mining whereby hot water is injected to dissolve trona and the resulting brines are processed to soda ash and baking soda [63]. In 2024, the Eti Soda plant at Beypazari (which entered full production in 2009) produced $1.9\text{--}2.0 \text{ Mt yr}^{-1}$ of natural soda ash and bicarbonate, while the Kazan Soda facility, commissioned in 2017, produced $>3 \text{ Mt yr}^{-1}$ with plans for further expansion [64].

In China, natural soda ash is now produced at a comparable scale to the US. The Berun project in Alxa Right Banner has the capacity to produce $\sim 7.8 \text{ Mt yr}^{-1}$ of soda ash and 2 Mt yr^{-1} of baking soda, with plans to increase capacity above 10 Mt yr^{-1} of soda ash [65]. Soda ash is also produced from trona/nahcolite mines in Wucheng and Tongbai (extraction rates unknown but likely at the kt yr^{-1} scale) [66] while sodium bicarbonate is produced ($\sim 0.3 \text{ Mt yr}^{-1}$) at Chaganur (aka Qagan Nur) Salt Lake. Soda ash from natural sources now contributes $\sim 16\%$ of China's total capacity, with the remaining capacity coming from synthetic production via the Solvay and Hou processes [57].

In Africa, production rates are more modest, Tata chemicals has produced 300 kt yr^{-1} of soda ash at their Lake Magadi site in Kenya [66], with plans to triple production [67]. Other sodium (bi)carbonate extraction sites include the Sua Pan which is one part of the Makgadikgadi Pan (Makgadikgadi Basin), Botswana (300 kt yr^{-1}) [68], Lake Chad (12 kt yr^{-1}) [69], Lake Abiyata and the surrounding lakes in Ethiopia (5 kt yr^{-1}) [70]. Other than one small project in Argentina, the Laguna Santa Maria Soda Ash Project [71] at the development stage, production in the rest of the world is small or unquantified.

Appendix D – Cost estimates for solution mining

Building on the solution-mining flowsheet of Nasün-Saygılı and Okutan (1996), we considered two delivery points for ocean alkalinity enhancement: (i) a solid monohydrate product, and (ii) a pre-crystallised carbonate-rich liquor produced upstream of evaporation/crystallisation. The reported speciation indicates that the solution-mining liquor (“crystalliser feed”) can be highly enriched in sodium carbonate (28.0 wt% Na₂CO₃; 0.7 wt% NaHCO₃; 71.3 wt% H₂O), whereas the downstream solid monohydrate contains 84.3 wt% Na₂CO₃ with 15.5 wt% structural water (**Table D1**). This compositional shift has a first-order impact on the gross CO₂ removal potential per tonne delivered: the dilute liquor corresponds to 0.08 tCO₂ per tonne of solution (but 0.22 tCO₂ per tonne of solids-equivalent, assuming conversion to monohydrate for consistent mass comparison), while the monohydrate crystals correspond to 0.35 tCO₂ per tonne. These values set the mass-handling and transport penalty associated with stopping earlier in the process.

To quantify the economic implications, we developed a screening cost estimate for a plant producing 679,500 t yr⁻¹ of sodium (bi)carbonate, adapting capital and operating cost line items from Nielsen (1983) and scaling to 2024 USD. Capital costs were escalated using a CEPCI ratio of 2.53 (1983 to 2024) and annualised using a 10% capital charge factor, giving a total installed capital of \$137.7M and annualised capital of \$13.8M yr⁻¹ (**Table D2**). Operating costs were inflated with an overall factor of 3.15 (1983 to 2024), yielding \$52.0M yr⁻¹ total operating cost (**Table D3**). Consistent with expectations for soda-ash style processing, the “processing” section dominates both capital and operating cost: solution-mining field items account for only \$2.85M of capital (~2%) and \$2.63M yr⁻¹ of operating cost (~5%), with the remainder driven by downstream processing (notably fuel and maintenance).

Because downstream processing is the principal cost hotspot, we also evaluated a once-through concept that intentionally eliminates evaporation/crystallisation and delivers the pre-crystallised liquor directly to the ocean. This case retains solution-mining costs and adds pipeline/pumping requirements using the assumptions in **Table D4** (e.g., water:solid mass ratio = 2, head loss 0.01 m km⁻¹ for a 0.5 m steel line, 70% pump efficiency, electricity at \$0.1 kWh⁻¹, and a pipeline cost of \$1.5M km⁻¹). Standard project factors (EPC and contingency) and an economic levelisation factor (1.2, consistent with an 8% discount rate and 25-year service life) were applied. **Figure D1** summarises the resulting levelised cost of gross CO₂ removal for the two delivery options, pre-crystallised liquor (blue) versus solid monohydrate (red), highlighting the trade-off between avoiding energy-intensive processing and accepting increased mass transport (water handling) when stopping earlier in the flowsheet.

Table D1 Sodium carbonate speciation through solution mining and processing adapted from Nasün-Saygili and Okutan (1996) [72]

Stage	Na ₂ CO ₃ (%)	NaHCO ₃ (%)	H ₂ O (%)	tCO ₂ t _{total} ⁻¹	tCO ₂ t _{solid} ⁻¹
Trona deposit	46.9	37.2	15.9	0.15	0.15
Crystalliser feed	28.0	0.7	71.3	0.08	0.22 ^a
Monohydrate crystals	84.3	0.2	15.5	0.35	0.35

a. assuming that the crystalliser feed would form monohydrate (15.5% H₂O), for consistent mass comparison with the monohydrate pathway

Table D2 Capital costs associated with solution mining and processing adapted from Nielsen (1983) [73] for an NaHCO₃ production capacity of 679,500 tonnes per year.

Cost item	\$ ₍₂₀₂₄₎ ^a
<i>Solution Mining</i>	
Casing Solution Wells	1,368,318
Casing Monitoring Wells	380,088
Well Head injectors	93,755
Well Head Producers	187,510
Piping - Trunk lines	301,537
Inj. and production lines	41,810
Contingency (20%)	474,604
<i>Processing</i>	
Major Equipment	35,474,913
Equipment labour	14,164,626
Bulk material labour	28,227,895
Access Road	228,053
Buildings	2,533,922
Service facilities	12,923,004
Engineering	8,463,301
Construction management	5,675,986
Contingency (25%)	27,112,969
Total Capital	137,652,292
Annualised Capital ^b	13,765,229

a. Capital costs were converted to present day values assuming a chemical engineering plant cost index ratio of 2.53 between 1983 and 2024.

b. Capital was annualised assuming a capital charge factor of 10%

Table D3 Operational costs associated with solution mining and processing adapted from Nielsen (1983) [73] for an NaHCO₃ production capacity of 679,500 tonnes per year.

Cost Item	\$ ₍₂₀₂₄₎ ^a
<i>Annual drilling</i>	
Drilling rig costs	1,039,500
Plant	30,240
Explosives	100,800
Cement	321,300
Drilling bits	90,720
Casing replacement	472,500
Large pipe relocating	507,150
Small pipe relocating	69,300
<i>Processing</i>	
Electricity	2,082,150
Fuel	20,475,000
Miscellaneous	315,000
Operating Labour	2,346,750
Supervisory Labour	352,800
Maintenance	10,080,000
Operating supplies	1,518,300
Laboratory charges	352,800
Taxes	2,529,450
Insurance	1,685,250
Overhead	7,667,100
Total Operational	52,036,110

a. Operational costs were scaled from 1983 to 2024 with an inflation factor of 3.15.

Table D4 Additional economic and process considerations for a once-through system for delivering pre-crystallised Na₂CO₃-rich liquid to the ocean.

Cost item	Value	Comment
<i>Alkaline liquid delivery</i>		
Water to solid mass ratio	2	[72]
Head loss within distribution pipe (m km ⁻¹)	0.01	For a diameter of 0.5 m [73] steel
Pump efficiency (%)	70	Assumed typical
Electricity cost (\$ kWh ⁻¹)	0.1	Conservative US costs for commercial water [74]
Cost of pipe (M\$ km ⁻¹)	1.5	
<i>Economic factors</i>		
Capital recovery	0.1	Assumed typical
Levelisation	1.2	Discount rate of 8% and a service-life of 25 years
Engineering procurement and contractor	0.1	Assumed typical
Contingency	0.2	Consistent with Nielsen (1983) [73]

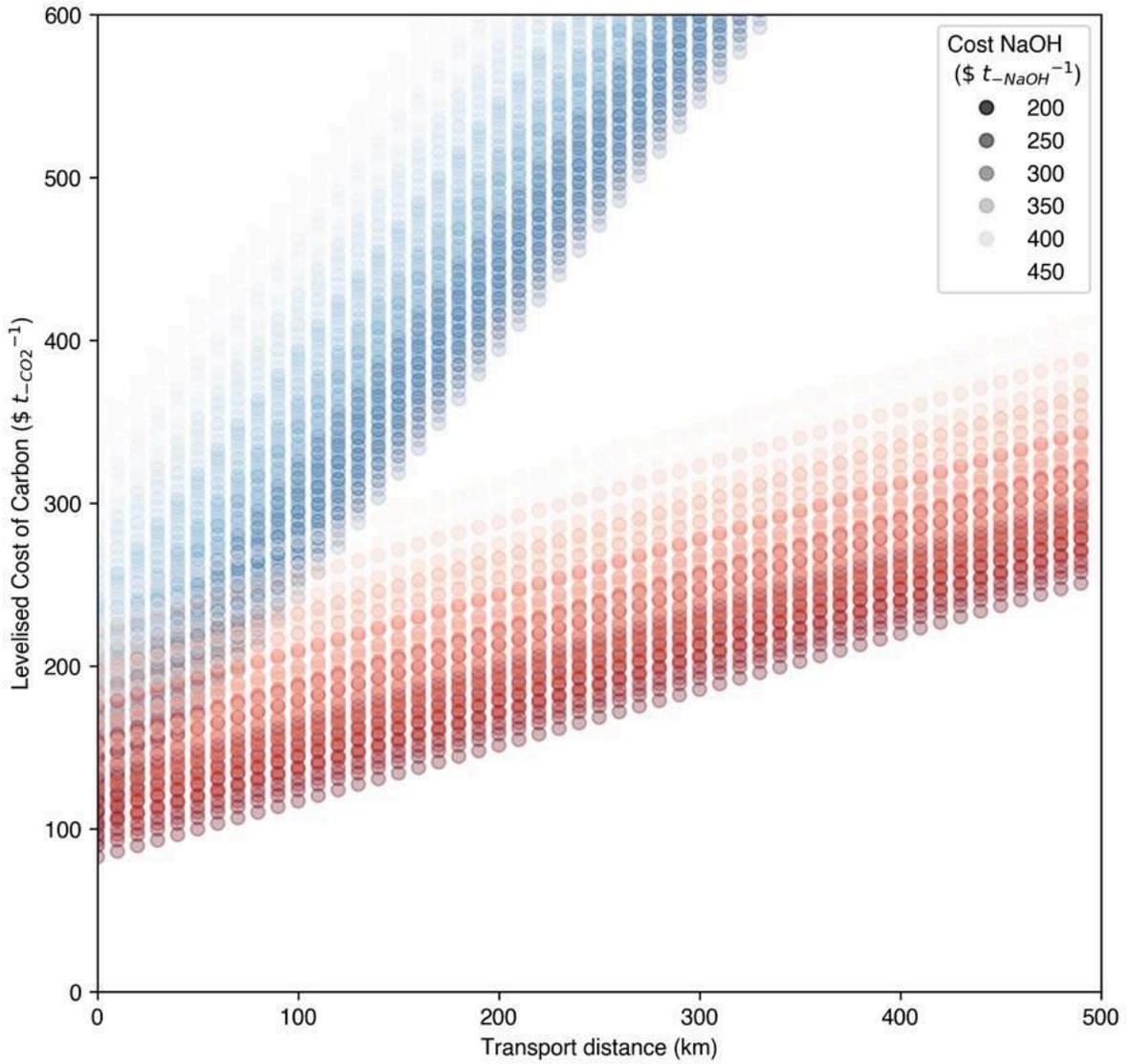


Figure D1 The levelised cost of gross CDR for a once-through system that dissolves trona (680 kt yr^{-1}) through solution mining and delivers the pre-crystallised liquid (blue) or solid monohydrate (red) to the ocean.

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