Environmental Science & Technology
Article
pubs.acs.org/est

CO_2 Mitigation Potential of Mineral Carbonation with Industrial Alkalinity Sources in the United States

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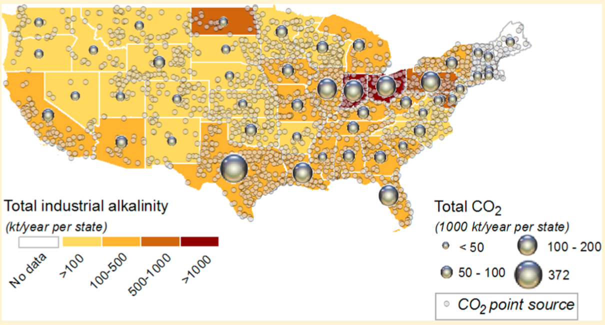

ABSTRACT: The availability of industrial alkalinity sources is investigated to determine their potential for the simultaneous capture and sequestration of CO_2 from point-source emissions in the United States. Industrial alkalinity sources investigated include fly ash, cement kiln dust, and iron and steel slag. Their feasibility for mineral carbonation is determined by their relative abundance for CO_2 reactivity and their proximity to point-source CO_2 emissions. In addition, the available aggregate markets are investigated as possible sinks for mineral carbonation products. We show that in the U.S., industrial alkaline byproducts have the potential to mitigate approximately 7.6 Mt CO_2/yr, of which 7.0 Mt CO_2/yr are CO_2 captured through mineral carbonation and 0.6 Mt CO_2/yr are CO_2 emissions avoided through reuse as synthetic aggregate (replacing sand and gravel). The emission reductions represent a small share (i.e., 0.1%) of total U.S. CO_2 emissions; however, industrial byproducts may represent comparatively low-cost methods for the advancement of mineral carbonation technologies, which may be extended to more abundant yet expensive natural alkalinity sources.

INTRODUCTION

Approximately 30 Gt of CO_2 are emitted in the atmosphere worldwide with approximately 6 Gt emitted from the United States alone.[1] It is widely accepted that a portfolio of solutions will be required for mitigation of CO_2 at scale.[2] The primary components of this portfolio include increasing fuel conversion efficiency, carbon capture and storage (CCS), renewables, and fuel switching.[3−6] Mineral carbonation falls under the CCS mitigation strategy as a form of CO_2 storage. Mineral carbonation refers to the reaction of CO_2 with alkali divalent cations (e.g., Ca^{2+} and Mg^{2+}) to produce carbonate minerals that are stable at atmospheric conditions. Both natural and industrial alkalinity sources exist and have been investigated for mineral carbonation. While naturally available alkalinity sources are abundant, their use as alkalinity resources is associated with high energy costs due to the mining and preprocessing (e.g., grinding) required.[7,8] Renforth et al. investigated industrial alkalinity source (e.g., aggregate and mine waste, construction and demolition waste, iron and steel slag, and fuel ash) availability for mineral carbonation, and estimated that the global production and sequestration potential are 7−17 Gt/yr and 67−1217 Mt-CO_2/yr, respectively; however, the sequestration potential results are based on the assumption that the divalent cation content of the material is completely converted to carbonate.[9] Previous work suggests that the carbonation reaction is kinetically limited by the dissolution step even for industrial residues; therefore, the design of carbonation systems requires accounting for kinetics in order to ensure adequate conversion rates.[10−12]

Despite the promise of mineral carbonation technologies, accurate accounting of alkalinity source production and sequestration potential has been limited due to lack of reliable alkalinity source availability/production data, variation in chemical and mineralogical content of alkalinity sources, and inconsistent methods of estimating potential. The present work focuses on industrial alkalinity sources, due to their availability and reactivity, and assesses the impact of using industrial alkalinity sources on mitigating CO_2, based upon alkalinity availability and geography. The location of resources is critical because transportation of CO_2 and/or alkalinity sources could result in significant energy consumption and cost.

Common industrial-sourced alkaline byproducts that have been investigated for mineral carbonation include coal fly ash (FA),[7] electric arc furnace (EAF) dust and steel-making slag (SS),[11,13−20] waste concrete[12,21] and cement kiln dust (CKD),[21−23] municipal solid waste incinerator (MSWI) ash,[24−29] asbestos mine tailings,[7,30] and bauxite residue.[29,31−37]

In addition to the potential CO_2 mitigation associated with mineral carbonation of industrial alkalinity sources, this process adds significant environmental benefit in the handling of industrial byproducts that may otherwise be considered as waste (or hazardous waste) materials.[27,28,38,39]

Received: February 10, 2013
Revised: June 4, 2013
Accepted: June 5, 2013
Published: June 5, 2013

dx.doi.org/10.1021/es4003982 | Environ. Sci. Technol. 2013, 47, 7548−7554

Due to the abundance of available alkalinity from coal-fired power, cement manufacturing, and steel production industries byproducts, the focus of the current study is the potential mitigation of CO_2 via carbonation using CKD, FA, and SS. It is worth highlighting that the available alkalinity is a function of the production rate and concentration of alkalinity in the alkalinity source; for instance, CKD and SS were selected despite relatively low production rates due to their high concentrations of CaO and/or MgO. In comparison, MSWI has a U.S. production rate similar to CKD and SS (8.5−9 Mt/yr), but was not included due to the variable composition and available alkalinity in MSWI.[27,28] Future work should include the investigation of additional promising industrial alkalinity sources such as MSWI.

Cement kiln dust is an alkali-rich dust produced during cement manufacturing at a ratio of approximately 0.15−0.20 tonne (t) CKD per t cement.[40] The typical weight percent ranges of calcium oxide (CaO) and magnesium oxide (MgO) in CKD are 38−50% and 0−2%, respectively.[41] Cement kiln dust is a fine-grained solid, with particle size on the order of micrometers,[22] and is an ideal source of alkalinity for mineral carbonation due to its composition and small particle size.[7] Huntzinger et al. investigated the carbonation of CKD at conditions of approximately 98% relative humidity, 25 °C, and 1 atm with a CO_2 partial pressure of 0.8 atm, and found that the degree of carbonation correlates directly with the mass fraction of calcium oxide and hydroxide content of the CKD.[22] The degree of carbonation at a given time, t, is defined as the mass of CO_2 taken up by the sample, $M_{CO2}(t)$, divided by the maximum theoretical carbonation of the sample. The average degree of carbonation was found to be approximately 77% over 8 days, with 90% of this carbonation occurring in less than 2 days.[22]

Fly ash is a residue generated from the combustion of coal, and is typically captured after coal combustion by air pollution control devices such as fabric filters or electrostatic precipitators. Fly ash comprises approximately 60% of all coal combustion waste. Fly ash is a complex, amorphous, and chemically heterogeneous material, and its physicochemical properties depend on the composition of the feed coal and the operating conditions of the coal-fired power plant. In the U.S., coal is ranked in one of four broad categories, listed in order of increasing purity: lignite, subbituminous, bituminous, and anthracite. While FA is often classified based on these ranks, it is important to note that the FA composition even within these categories varies greatly due to the chemical and physical heterogeneity of coal. In general, inorganic minerals comprise approximately 90−99% of fly ash, while organic compounds make up approximately 1−9%.[42] The inorganic minerals consist primarily of silicon dioxide (SiO_2) and CaO, along with other metal oxides such as Fe_2O_3 and MgO. The typical weight percent ranges of CaO and MgO in FA are 1−37% and 1−15%, respectively.[43−45] Typically, Ca-rich minerals are much less abundant than alumino-silicates and Fe-oxides in high-rank coals; however, in lower rank coals, Ca-rich minerals dominate the inorganic crystalline fraction of FA.[7] Fly ash can also be classified as Class F or Class C type. Class F type fly ash typically contains less than 20% CaO (lime) and is composed of more glassy silica and alumina compounds, while Class C fly ash is known to have self-cementing properties and contain more than 20% CaO. Montes-Hernandez et al. investigated the aqueous carbonation of FA and found that 82% of the FA CaO content is converted to $CaCO_3$ after reacting for 2 h at 30 or 60 °C.[46] The authors report that carbonation conversion is independent of initial CO_2 partial pressure, but did investigate high-pressure conditions ranging from approximately 10 to 39 atm of CO_2. Additional investigations have considered the carbonation of fly ash or fly ash−brine mixtures, and have also found that CaO present in FA is readily converted to $CaCO_3$.[43,47]

Steel slag is a byproduct of iron and steel manufacturing and includes the impurities separated from iron during ore smelting. Slag is comprised of a heterogeneous mixture of crystalline components, including iron oxides, calcium and magnesium hydroxides, oxides, silicates, and quartz.[48] Slag content varies depending on the ore and the smelting process; specifically, the type of furnace used, e.g., blast furnace (BF), basic oxygen furnace (BOF), EAF, and ladle furnace (LF). Data are rare on the actual average production of slag, as the amount of slag produced is not routinely measured. The amount of slag produced changes depending on the overall chemistry of the raw furnace feed, in particular the iron ore feed grade, and the type of furnace used. Approximately 0.2 t of steel slag is produced for each t of iron produced;[48] however, a significant portion of the slag is entrained metal and recovered during slag processing, and the amount of marketable slag remaining after entrained steel removal is usually equivalent to between 10% and 15% of the crude steel output.[49]

Steel slag is an ideal feedstock for mineral carbonation due to its high alkalinity and, more specifically, high Ca content. The typical weight percent ranges of CaO and MgO in SS are 32−58% and 3.9−10.0%, respectively.[15,48] Previous investigations suggest that SS is a viable feedstock for cost-effective CO_2 sequestration via mineral carbonation.[12,16,50] Bonenfant et al. investigated aqueous carbonation of EAF and LF slag suspensions and found sequestration capacities of 0.02 and 0.25 t-CO_2/t-slag, respectively.[15] Huijgen et al. investigated mineral carbonation of SS in aqueous suspensions and found that the reaction rate depends primarily on particle size and reaction temperature.[8] The authors report 74% extent reacted after 30 min for SS with particle size less than 38 μm at 19 bar and 100 °C. Stolaroff et al. estimated a SS carbonation potential of 0.27 t CO_2 sequestered per t SS, assuming that 75% of Ca content reacts with CO_2, with an estimated cost of \$8/t CO_2 sequestered.[6]

The carbonate produced through mineral carbonation has the potential beneficial reuse as synthetic aggregate. In addition to serving as a sink for CO_2, this synthetic aggregate has the potential cobenefit of preventing CO_2 emissions associated with mining aggregate. For this reason, the size of the aggregate market in the U.S. has been investigated since this market will inevitably serve as an upper limit of the CO_2 mitigation potential from reuse of mineral carbonation products. Natural aggregates are traditionally sourced from either crushed stone or sand and gravel, and the various aggregates can frequently be interchanged with one another. The 2010 market value of all natural aggregates was 17.5 billion dollars.[51] Natural aggregates are primarily used in the construction industry, and account for approximately half of U.S. mining industry output.[52] However, natural aggregates are not universally available and some areas lack quality and/or practically accessible natural aggregate. If the carbonate product could not be used in the aggregate industry, the carbonate product would have to be landfilled unless an alternative use could be found. In the current study, the only use considered for the carbonate product was the aggregate market.

To assess the CO_2 mitigation potential of mineral carbonation using industrial alkalinity sources, the current work determines the abundance and geographic location of industrial alkaline sources. In addition, the amount of synthetic aggregate that could potentially be produced from reaction of CO_2 with industrial alkalinity sources in the U.S. is compared on a state-by-state basis to the mined aggregate industry, to assess the extent to which synthetic aggregate could replace mined aggregate.

■ METHODOLOGY

The carbonation capacity for a given alkalinity source depends on the total alkalinity available, the reactivity of the alkaline components, the kinetics of the reaction, and the reaction conditions. In the present work, available alkalinity is used as a direct measure of the carbonation capacity of a given resource, and is a function of the maximum theoretical carbonation capacity of the resource and the expected extent reacted. It is important to note that the current work does not take into account the material or energy consumption of the carbonation processes; previous work suggests that the life-cycle CO_2 emissions associated with the process vary greatly depending on the alkalinity source and reaction conditions, but can be as low as 15% for CKD.[8] The maximum theoretical carbonation of a material is a measure of the alkalinity of the material; in the present work, alkalinity is defined to include Ca^{2+} and Mg^{2+}.[53] The expected extent reacted is based on previous reported values, explained in more detail for each resource below. Extent reacted is typically reported as a percentage of the maximum theoretical reacted. The available alkalinity is defined as the total Ca^{2+} and Mg^{2+} alkalinity of a resource multiplied by the percentage of expected extent reacted. Given the extremely simplified and generic metal oxide alkaline resource (MO), the carbonation reaction can be expressed as

$$MO + CO_2 \leftrightarrow MCO_3 + \text{heat} \qquad (R1)$$

with a 1:1 molar ratio of CO_2 to mineral oxide and to the carbonate product formed, such that $M = Ca^{2+}$ or Mg^{2+}.

U.S. maps of CO_2 emissions and alkalinity source, mined aggregate, and synthetic aggregate production were compiled using Geographic Information Systems (GIS) software ArcGIS.[54] The National Energy Technology Laboratory National Carbon Sequestration Database (NATCARB) provided CO_2 emissions by source for coal-fired power plants, cement kilns, and steel plants.[55] The production rate of FA, CKD, and SS alkalinity was estimated based on available data, as described below. Natural aggregate production by state was determined from the "Mineral Operations − Sand and gravel" and "Mineral Operations − Crushed Stone" of the National Atlas 2005 (map layers compiled by the Minerals Information Team of the USGS).[56,57]

The alkalinity sourced from fly ash was estimated based upon the type of coal burned. The coal types considered in this study include Appalachian Low-Sulfur bituminous, Appalachian Medium-Sulfur bituminous, Wyoming Powder River Basin subbituminous, Wyodak bituminous, North Dakota lignite, and Illinois #6 bituminous. Information regarding the U.S. power plant locations in addition to capacity and type of coal burned was determined from the U.S. Energy Information Administration data.[58] The coal composition, including ash content and distribution of calcium and magnesium oxides, differs among the various coal types and was determined from the internal fuel library of the Integrated Environmental Control Module (IECM) developed by Rubin and colleagues at Carnegie Mellon University.[59] For each of the coal types considered, the ratio of fly ash produced to CO_2 emitted was calculated from this software package based upon a 500-MW power plant. On average, power plants generate approximately 10−13 t per hour of ash, which is small in comparison to the approximate 435 t per hour of CO_2 generated. The rate of fly ash production by each power plant was calculated by multiplying the CO_2 emissions from the plant by the appropriate fly ash production-to-CO_2 emissions ratio. Based on previous work by Montes-Hernandez, the expected extent reacted for FA is assumed to be 82%.[46] For CKD and SS, the rate of alkalinity production was estimated based on the typical concentrations of calcium and magnesium oxides in the alkalinity source. The amount of CKD generated per source was calculated based on the CO_2 emissions of the source, a clinker-to-CO_2 production ratio of 1, and a CKD-to-clinker production ratio of 0.060, based on the assumption that a nonhazardous fuel kiln and dry process are used.[21,60,61] Based on data from Huntzinger et al., the expected extent reacted for CKD is assumed to be 77%.[22]

The amount of SS generated per source was calculated based on the CO_2 emissions of the source, a ratio of CO_2 emitted to steel produced of 0.64, and a ratio of steel produced to slag generated of 8.33.[15,62] Based on previous work by Huijgen et al., the expected extent reacted for SS is assumed to be 75%.[16]

■ RESULTS AND DISCUSSION

Annual Production of Alkalinity. To assess the potential for mineral carbonation using industrial alkalinity sources, the total U.S. production of CKD, FA, and SS were estimated, as shown in Table 1. Approximately 5.2 Mt of CKD are produced annually in the U.S. The SS production (after metal removal) in the U.S. is estimated to be 10.3 Mt/yr. U.S. coal-fired power plants produce approximately 52.8 Mt of FA annually.

Table 1. U.S. Production of CKD, FA, and SS and Associated Alkalinity

alkalinity source	references[a]	U.S. production (Mt/yr)	estimated uncertainty[b] (%)	associated alkalinity (Mt/yr)
cement kiln dust	21,60,61	5.2	±15	1.41−1.92
fly ash	58,59	52.8	±10	0.15−4.33
steel slag	15,62	10.3	±15	2.60−4.89

[a] Estimation based on alkalinity source production data and alkaline content data from the literature; details provided in Methodology. [b] Includes uncertainty of the NATCARB CO_2 emissions data, assumed to be 5%, and for CKD and SS uncertainty in production rate of alkalinity source per CO_2 emitted, assumed to be 10%, and for FA uncertainty in the IECM data, assumed to be 5%

The calculated production rates are in agreement with estimates reported in previous literature.[49,63,64] The resulting alkalinity production rates for CKD, FA, and SS are 1.67, 2.24, and 3.74 Mt/yr, respectively. It is important to note that the alkaline content (i.e., CaO and MgO) in CKD and SS is much greater than that in FA; therefore, the alkalinity production rates for CKD and SS are of the same order as for FA, despite their relatively low production compared to FA. Figure 1 shows the rate of Ca and Mg alkalinity production among the various alkalinity sources, based on the U.S. production rate, the concentration of Ca- or Mg-based alkalinity, and the expected

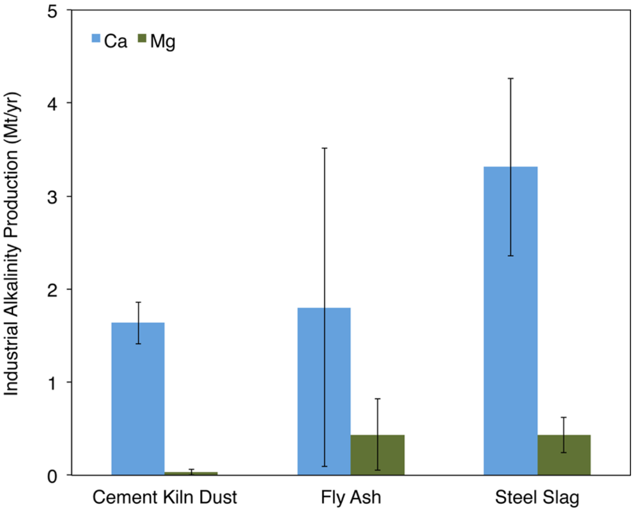

Figure 1. Industrial alkalinity production (Mt/yr) rates of Ca and Mg cations for U.S. industrial alkalinity sources.

extent reacted of each alkalinity source. For CKD and SS, the rate of alkalinity production was estimated based on the typical concentrations of Ca and Mg in the alkalinity source. For FA, the rate of alkalinity production was estimated based on the typical concentrations of Ca and Mg in each coal type. The error bars represent uncertainty based on the low and high values of Ca and Mg in the alkalinity source (see Introduction) and the estimated uncertainty in the alkalinity source production rate (see Table 1). Due to the high variability in FA composition, the uncertainty in FA-based alkalinity production is much greater than in SS or CKD. However, for all cases it is important to note that the low and high bounds represent extremes. For instance, the low bound of FA-based Ca production (0.1 Mt/yr) would occur only if all FA produced in the U.S. were low-Ca FA. FA and SS produce more than double the alkalinity that CKD produces, and approximately 88% of alkalinity available from industrial sources is Ca-based. In the case that carbonation reactions are optimized to improve the extent reacted, thereby increasing the available alkalinity from a given source, the alkalinity production for that source would increase. For instance, recent work by Chang et al. suggests that the extent reacted for SS could be as high as 89.4%, over 10% greater than the value assumed in the current work.[20]

The total Ca- and Mg- alkalinity available from CKD, FA, and SS is approximately 7.65 Mt/yr (6291 mol/s), with a mineral carbonation capacity of approximately 6.98 Mt-CO_2/yr. This value should be considered an upper limit for the CO_2 sequestration capacity of CKD, FA, and SS in the U.S., as it does not take into account the energy consumption or CO_2 emissions that would result from the carbonation process.

Geographic Availability of Carbonation Reactants. To determine the geographic relationship between CO_2 emissions sources and industrial alkalinity sources, the locations of industrial alkalinity sources have been mapped in relation to U.S. CO_2 emissions. Figure 2 provides the locations of stationary CO_2 emissions in the U.S. overlapped with circles representing CKD, FA, and SS production locations, scaled to the relative annual production.[55] States with no production of CKD, FA, and SS are Connecticut, Hawaii, Maine, New Hampshire, Rhode Island, and Vermont. It is important to note the difference in marker scales for the production of alkalinity sources versus CO_2; CO_2 production is roughly 3 orders of magnitude greater than alkalinity source production. As Figure 2 illustrates, because FA is a byproduct of burning fossil fuels and cement and iron and steel manufacturing are energy intensive processes, almost all locations where CKD, FA, or SS is produced also produce CO_2; therefore, it is unlikely that carbonation reactants would need to be transported great distances. The colocation of carbonation reactants is significant when considering the feasibility of mineral carbonation, since this would minimize the cost and energy consumption associated with material transport.

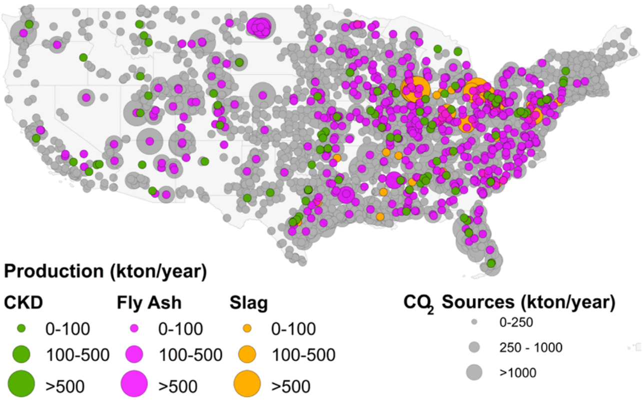

Figure 2. Industrial alkalinity sources and stationary CO_2 sources in the conterminous U.S. (CO_2 sources based on NatCarb, 2010).

Potential for Reuse of Carbonate As Synthetic Aggregate. The carbonate produced through mineral carbonation has the potential beneficial reuse as synthetic aggregate. The mined aggregate market serves as an upper limit of the CO_2 mitigation potential from reuse of mineral carbonation products as synthetic aggregate. The estimated annual outputs of crushed stone and sand and gravel produced for consumption in the U.S. in 2010 were 1.19 Gt and 820 Mt, respectively.[51] Absolute mined aggregate volumes are illustrated in Figure 3 by state, with the darker shades representing the higher production areas. Examination of Figure 3 reveals that Texas, Pennsylvania, and Missouri have the highest volumes of mined aggregate. In addition, the synthetic aggregate produced from the reaction of CO_2 (from point-source power plant emissions) with industrial-based alkalinity sources is presented

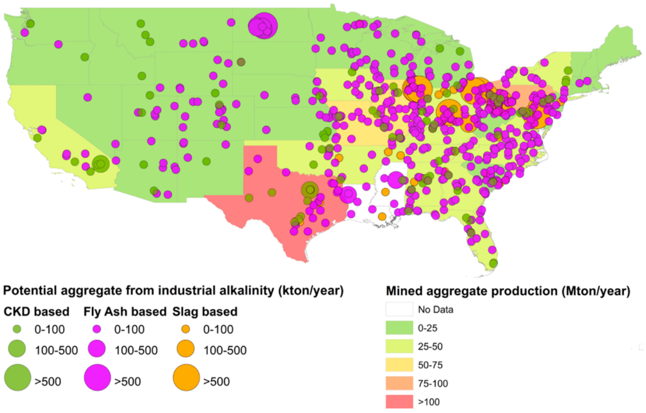

Figure 3. Comparison of potential synthetic aggregate production from mineral carbonation using industrial alkalinity sources to mined aggregate production in the conterminous U.S.

in Figure 3 and divided among SS, CKD, and FA sources. Given that the alkalinity source is the limiting resource for mineral carbonation, the location of the synthetic aggregate production was assumed to be that of the alkalinity source rather than the CO_2 source. From Figure 3 it becomes clear that the extent of synthetic aggregate production in total is an order of magnitude smaller than the mined aggregate volumes, and it can be assumed that most synthetic aggregate will find a market locally.

The potential total U.S. synthetic aggregate production using FA, CKD, and SS is estimated to be 20 Mt/yr, approximately 1.7% of total U.S. mined aggregate (i.e., 1.2 Gt/yr).[51] Figure 4 shows that the potential market share of synthetic aggregate from mineral carbonation using industrial-based waste products varies greatly in the U.S. The highest potential market share is found in North Dakota (171%). Table 2 shows a comparison of mined aggregate to synthetic aggregate for all of the states in which the synthetic aggregate could potentially replace more than 5% of the mined aggregate.

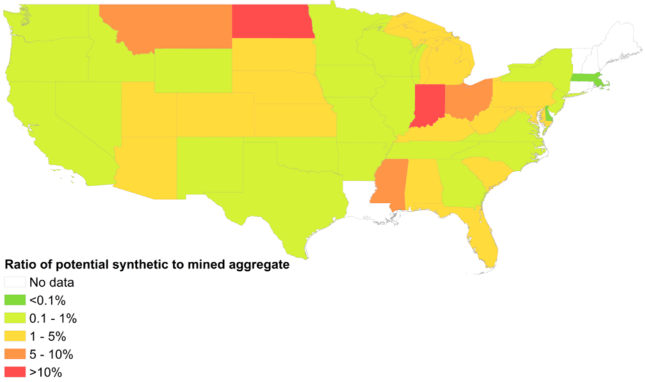

Figure 4. Ratio between potential synthetic aggregate production from mineral carbonation to mined aggregate production (i.e, the potential market share of synthetic aggregate) in the conterminous U.S.

Table 2. Top States of Synthetic Aggregate Replacement of Mined Aggregate

state	mined aggregate (kt/yr)	synthetic aggregate (kt/yr)	% synthetic of mined aggregate
North Dakota	985	1,687	171.3
Indiana	44,200	5,160	11.7
Mississippi	3130	278	8.9
Ohio	43,300	3,331	7.7
Montana	1,990	101	5.1

In addition to the direct CO_2 mitigation from mineral carbonation, it is interesting to also consider the CO_2 emissions associated with mining aggregate and the CO_2 mitigation potential of displacing mined with synthetic aggregate. The CO_2 emissions associated with the mining of crushed granite, crushed limestone, and industrial sand were determined based on the direct consumption of energy associated with mining.[10,36,44,65] Table 3 shows the CO_2 emissions per t of mined material, and the potential CO_2 emissions mitigation by replacing 1.7% of U.S. mined aggregate production with synthetic aggregate. The CO_2 mitigation potential of replacing mined aggregate with synthetic aggregate depends on the source of the mined aggregate, and the greatest impact would be attained by replacing sand and gravel-based aggregate given the high CO_2 emissions associated with sand and gravel mining.

Table 3. CO_2 Emissions and Mitigation Potential of Mined Aggregate

mined aggregate	CO_2 emissions (kg CO_2/t mined)	mitigated CO_2[a] (Mt CO_2/yr)
crushed granite	3.6	0.07
crushed limestone	4.5	0.09
sand and gravel	23.9	0.48

[a] Assuming 1.7% of U.S. mined crushed stone, crushed limestone, or sand and gravel aggregated replaced with synthetic aggregate.

This work shows that in the U.S., FA, CKD, and SS-based alkalinity has the potential to mitigate approximately 7.6 Mt CO_2/yr, of which 7.0 Mt CO_2/yr are CO_2 captured through mineralization and 0.6 Mt CO_2/yr are CO_2 emissions avoided through reuse of synthetic aggregate. This represents a small share (0.1%) of U.S. total CO_2 emissions; however, industrial byproducts may represent comparatively low-cost methods for the development of mineral carbonation technologies, which could be extended to abundant but more expensive natural alkalinity sources.

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the U.S. Department of Energy under Contract DE-AC36-08-GO28308 with the National Renewable Energy Laboratory for the Joint Institute for Strategic Energy Analysis. Additionally, we express our gratitude to Patricia Carbajales, Geospatial Manager at the Branner Earth Sciences Library and Map Collections, for her assistance with the GIS resources; Sam Krevor and Valentina Prigiobbe, who generously provided helpful advice and feedback; and Jasper Van der Bruggen, Noemi Alvarez, Wenny W. Ng, and Emilia Dicharry, for their assistance in making the maps used in this work.

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