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Building materials could store more than 16 billion tonnes of CO_2 annually

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Other Supplementary Material for this manuscript includes the following:

Data S1 to S8

Materials and Methods

S.1. Material mass consumed annually

To determine the carbon storage possible in building materials, first the mass of materials demanded annually were quantified (Supplementary data sheet 1):

- While there are many wood and wood-related products or outputs (including energy from wood resources) manufactured annually, here we only consider sawn wood and wood-based panels as wood products that would enter infrastructure systems. Based on data from the United Nations Food and Agricultural Organization, in 2016, these two wood categories amounted to approximately 824 million m^3 of material (*72*). While the densities of woods can vary, here we take a representative density of 1.5 g/cm^3 (*73*) to determine a mass of wood to infrastructure applications as 1.24 Gt.
- The United States Geological Survey (*74*) reported that 4.15 Gt of cement was produced globally in 2016.
- Aggregate demand is not well tracked (*75,70)*. However, using cement production, it can be estimated that there is approximately 30 Gt of concrete production per year (*74*, *75*, *76*, *77*). While there are several constituents that are used in the production of concrete, the most prevalent are hydraulic cement, water, and aggregates. Water demand is typically lower than cement demand, but it can be as high as cement demand per volume of concrete (*78*). Therefore, estimating the mass of cement production and a nearly equivalent water demand, we can determine a proxy for aggregates in concrete as being the remainder of the concrete. As such, we use these values to approximate aggregate production for use in infrastructure materials, such as concrete, as being approximately 21.7 Gt.
- Reported statistics indicate 335 Mt of plastics were produced in 2016 with an estimated 66 Mt of plastics going to building and construction uses (*79*).
- It is estimated that roughly 1.2 trillion bricks are produced each year (*80*); although, we note, data reporting for bricks is limited. Using this reported statistic and assuming that each brick weighs roughly 2 kg, global brick production is estimated here to be 2.4 Gt.
- Global asphalt production, amounting to 0.112 Gt was obtained from the United Nations Industrial Commodity 2016 Statistics (*81*). Given that asphalt bitumen typically comprises 5 wt% of asphalt pavement (*82*), the global production of aggregate for asphalt pavement production (2.1 Gt) was calculated.

S.2. Determining carbon storage potential

To determine the carbon storage potential of these building materials, their carbon content was determined (supplemental data sheet 3). Specific assumptions for each group of materials are outlined in the following subsections.

S.2.1 Theoretical storage potential per material

Cements. Here, we consider two carbonatable cements, carbonatable calcium silicate clinkers (CCSC) and magnesium oxides derived from magnesium silicates (MOMS). Through carbon mineralization, CCSC and MOMS are theoretically capable of sequestering roughly 0.2 and 0.5 kg CO_2 / kg cement respectively (*22*). We note, there are technical limitations for these cements

that may accrue at later life cycle stages: the pH of CO_2-cured cements is not high enough to protect steel from corrosion in reinforced concrete applications; the long-term durability of carbonatable cements requires further investigation prior to use; resource availability for CCSC is limited in many regions; an energy-efficient method for extracting MgO from relatively abundant magnesium silicate rocks has not yet been identified. Shifts in performance must be addressed and properly accounted for prior to utilization to ensure appropriate application.

Aggregates. Aggregates constitute the majority of concrete by mass, and sedimentary rocks are most typically used. Here we consider the use of synthetic rocks that have been formed via enhanced weathering to create pathways for accelerated carbon-uptake from the atmosphere, often resulting in materials with similar composition to the sedimentary rocks that are conventionally used (*83*). Although such carbonate-based aggregates are at varying technology readiness levels, we consistently model the CO_2 uptake potential based on the mass fraction of carbon in two carbonate minerals: $CaCO_3$ and $MgCO_3$. Based on the molecular masses of Ca, Mg, C, and O, we can estimate that carbon content is approximately 12% to 14% for aggregates. However, the most common means of extracting MgO and CaO that could then react with CO_2 under proper conditions to form $CaCO_3$ and $MgCO_3$ currently rely on decarbonation processes, which would lead to equivalent or greater emissions than the uptake potential (i.e. net emissions not net removal). However, other pathways exist, including mixing Mg- and Ca-rich industrial waste with a carbonate slurry (typically made from concentrated sources of CO_2 to achieve sufficient reaction times) to generate carbonate-based aggregates, thereby simultaneously valorizing waste materials and sequestering atmospheric CO_2 (*49*). Similarly, crushed concrete contains calcium hydroxide, which can react with CO_2 to form calcium carbonate ($Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O$) under appropriate conditions. Yet industrial wastes are currently not managed effectively, with roughly 50% of concrete waste going to landfills (*84*).

Biochar additives. While biogenic resources cannot easily be used at high levels within highly alkaline concrete, low levels of biogenic additives, particularly if they have undergone processing that improves dimensional stability and resilience in higher alkalinity, can act as a means of storing CO_2. Using biochar to store carbon in this way can valorize waste products such as agricultural byproducts. The carbon content of biochar can vary significantly based on the type of feedstock, therefore a ranges of 0.3 to 0.85 kg C / kg biochar (*85, 86*) is used. We analyze the potential if cement were replaced with up to 15 wt% of biochar, but note that replacement levels >5 wt% can lead to reductions in mechanical performance characteristics such as compressive strength (*87*), depending on characteristics of the biochar. Therefore, high levels of biochar replacement in concrete may only be possible for low load-bearing applications or when the biochar has desired properties.

Wood. Current use of wood as a building material accounts for roughly 2.2 Gt of stored CO_2. In addition to this photosynthetic carbon already being stored in the built environment, here we consider the effects of an increase in wood consumption by up to 20 wt% given that the demand

for industrial roundwood is expected to increase 25-40% by 2050 (*88*). Wood carbon content is variable depending on parameters such as species; however, most wood has a carbon content in the 40-60% weight range (*73*). We again highlight that forestry practices and end-of-life management are outside the scope of this work, but we note that poor management of biogenic resources can lead to undesired climate damages and should be addressed as an additional measure to limit emissions.

Bio-based Plastics. Plastics are not a large mass fraction of the built environment relative to materials like concrete and wood, but their use is gaining popularity, and pathways for creating carbon-storing plastics have been proposed. Consistent with other modeling assumptions, we calculate the carbon storage potential of plastics based on the mass fraction of carbon in the polymer, and this carbon is derived from atmospheric CO_2 via photosynthesis of biomass. The storage potential of bioplastics is sensitive to the type and source of biomass feedstock used given implications for land-use change emissions (*89*, *90*), which are outside the scope of our analysis. Plastic carbon content is also variable depending on backbone composition, additives, and other parameters, therefore a range of 40-90% is used. We also note that bio-based plastics now make-up ~1% of the plastic market, most of which are biodegradable and therefore not suitable for construction applications (*91*). Although production processes for more durable, nonbiodegradable bio-based plastics exist on a commercial scale, production costs are relatively high compared to petroleum-based plastics which has limited their growth rate, with only 0.2% of bioplastics being used in construction applications (*91*).

Brick. Brick, the oldest artificial building material, remains important, with roughly 1.2 trillion bricks produced annually (*80*). Recent studies have examined the potential for using agricultural wastes in brick (*92*). In this study, we assume up to 15 wt% of brick can be substituted with biomass fibers, a ratio which has been shown to maintain similar performance characteristics as traditional brick materials (*93, 94*). The carbon content of biomass fibers can vary significantly based on the type of feedstock, therefore a range of 0.4 to 0.6 kg C/kg (*95*) is used. The remaining mass (85%), is modelled as calcium carbonate formed via complete mineral carbonation of portlandite ($Ca(OH)_2$), similar to the processes described for crushed concrete. Although studies have examined the mechanical performance of portlandite brick, further research should investigate the mechanical performance of calcium carbonate brick, as well as the time scale required for complete carbonation.

Asphalt. Similar to concrete, asphalt pavement is a composite material that can be formed with several different carbon-storing materials. Here we consider the effects of using a carbon-storing binder and aggregates. We assume up to 100% replacement of traditional aggregate in asphalt pavement, with carbonate-based aggregate, with the aggregate uptake methods discussed above.

Bio-oil for asphalt. Asphalt pavement has two main components: (1) aggregate, which makes up roughly 95 wt% of asphalt, and (2) binder, asphalt bitumen, responsible for holding the

aggregate together. Asphalt bitumen is typically made from petroleum refining residues. Here we model the replacement of petroleum bitumen binders with bio-oil, a byproduct obtained from the thermal decomposition of biomass through pyrolysis or hydrothermal liquefication. Due to the similar chemical and rheological properties of bio-oil and asphalt binder, bio-oil can be used as a substitute or modifier for traditional petroleum-based asphalt binder. In addition to serving as a carbon storage method, the use of bio-oil instead of petroleum-based asphalt has been found to reduce energy requirements and costs due to lower required production temperatures (*96)*. The carbon content and performance characteristics of bio-oil can vary depending on its source (*97*). Here, as was done with our bio-based plastics models, CO_2 storage potential is based on carbon content. The carbon content of bio-oil for use in asphalt bitumen varies depending on the source, therefore literature on bio-oil production from agricultural by-products (*98*), wood waste (*99*), and food waste (*100*), was leveraged to obtain a range of carbon content (0.3 to 0.7 kg C/kg). We note, as with several of these carbon-storing technologies, there are currently reported performance differences from this biogenic resource. In general, it has been observed that the addition of bio-oil improves anti-aging and low-temperature performance but compromises high-temperature performance compared to traditional asphalt binder.

S.2.2. Method of calculation

For woods, plastics, aggregates, and bricks, stoichiometric relationships were used to determine the CDR potential. Namely, the following relationship was applied:

$$mass\ of\ CO_2\ stored = carbon\ content\ \times {}^{44}/_{12} \quad \text{S1}$$

The ratio of 44:12 is based on molecular masses of CO_2 and C respectively. We do not reflect additional emissions from the processes performed. We note the majority of processes require energy resources that have associated greenhouse gas (GHG) emissions. However, to estimate the CDR potential of building materials, we draw comparisons to the chemical-derived emissions of conventional materials. Using storage capacity, our findings suggest energy-derived emissions can still occur from the production of these CDR materials and still meet desired 1.5°C and 2°C warming targets with appropriate implementation of strategies.

Because conventional cement has minimal carbon content, we model three permutations on cementitious materials in this work. The first is based on a CCSC, for which the carbon storage potential has been modeled as 0.216 kg of CO_2 / kg of cement in previous work (*22*). The second is based on the carbonation of MOMS. In this second case, the carbon storage potential of 0.524 kg of CO_2 / kg of cement is modeled based on previous work (*22*). Finally, we consider the scenario in which 15% of cement is replaced by a high carbon content filler, such as biochar. In this third case, cement CDR potential is reduced by 15% and replaced by an 85% carbon-content biochar; the uptake potential of this biochar is determined following equation S1.

S.3. CO_2 emissions from materials production

To determine CO_2 emissions associated with individual materials studied herein, the fraction of these materials used in construction as reported by Hertwich (*12*) are used in most cases. However, for the quantity of cement and lime used in construction, estimates are made based on data from the United States Geological Survey (*101*); for the quantity of plastics used in construction, data are from (*102*).

The production of these materials results in CO_2 emissions throughout the supply chain, with the majority of emissions being a result of energy used in manufacturing and process-related emissions (i.e., those from chemical reaction processes or otherwise derived directly from the material resources). Similar to the scope of assessment considering bound carbon within the materials, process-related emissions are associated with the materials themselves, so we focus on these emissions. Process-related emissions from cement production result from decarbonization of limestone, amounting to roughly 0.52 kg CO_2/kg (*103*). Whereas process related emissions for aggregate production for concrete and asphalt pavement are assumed to be negligible, as the recovery of these aggregates should not lead to large flows of chemical-reaction derived emissions. The process-related emissions associated with plastic production vary significantly based on the type of polymer; however, it has been found that roughly 44% of cradle-to-gate impacts are attributed to emissions from petroleum refining (*104*); using this value, an average value of roughly 1 kg CO_2/kg plastic was used in this work. Chemical-derived emissions for brick production were assumed to be zero (*105*). Process related emissions for bitumen production (i.e., the asphalt binder) were assumed to be negligible given that it is a byproduct (rather than main product) of crude oil refining.

While cement has minimal carbon content, limestone (which does contain carbon) is regularly interground with cement or used as a mineral filler in concrete or mortar. The Global Cement and Concrete Association report the use limestone in cement blends at approximately 7% by weight in 216 (*106*). Higher interground carbon-based minerals or carbon-based filler assumed to be up to 15% has been proposed by the Portland Cement Association and used commonly in European countries as well as elsewhere for limestone (*107*). This difference between the potential use of an interground or filler carbon-based material suggest an additional 8% of such a resource. While the literature on the use of biochar in concrete remains nascent; here we will estimate the influence on carbon storage potential if the full 15% of this carbon-based resource were from biochar (*108*).

The utilization of biochar as a mineral filler itself is not critical; however, it provides a pathway for a high carbon uptake resource to be used in cement-based composites, potentially mitigating cement demand. In the production of biochar, biomass at its end-of-life is converted to biochar-based materials via pyrolysis (*15*). Biochar can be formed from a variety of biomass resources, such as agricultural crop residues. The use of these biochar resources can lead to net-negative emissions composites by overcoming emissions associated with production (*109*). Biochar carbon contents can vary based on crop, pyrolysis method, and other factors (*110*), but is approximated here as 30-85% based on a range of biomass resources reported by Lee et al. (*111*).

S.4. Sensitivity of uptake parameters

A sensitivity analysis was performed to examine the effects of variation in material carbon content and the effects of level of implementation of these carbon-sequestering materials on potential carbon storage at the global scale. To examine the sensitivity of each parameter individually, global carbon storage potential was calculated for a baseline scenario, in which all carbon contents and implementation levels were assumed to be the midpoint value of the minimum and maximum values (e.g., if the level of implementation can be anywhere from 0 to 100%, then 50% was chosen as the baseline). From here, several permutations were performed to examine the change in global CO_2 uptake when one individual parameter is changed to its minimum or maximum value. For wood, given that our model considers potential increases in consumption, rather than having one variable be "level of implementation" ranging from 0 to 100%, instead the sensitivity analysis examines the impact of varying increases in wood consumption from 0 to 20%. The results from this sensitivity assessment can be found in supplemental data sheet 5. A similar sensitivity analysis was done using heatmaps to examine the impact of CDR in each material on a per kg basis (see supplemental data sheet 5).

S.5. Resource availability assessment

A resource availability assessment was performed. Here, we focus on quantifying the resources required to fully implement these carbon-sequestering materials in construction applications and the quantity of resources currently available that can meet those demands. Noting that many strategies considered rely on utilization of readily available calcium or magnesium sources (e.g., those found in Earth's crust), the two main additional source groups of materials that were examined herein include (1) mineral wastes (to form carbonate-based aggregates and carbonatable cements) and (2) agricultural or woody residues as fillers or feedstocks for cement, bricks, plastic, and asphalt.

S.5.1. Availability of mineral wastes

Global annual production of mineral wastes, including red mud, blast furnace slag, steel slag, mine tailings, biomass ash, lignite ash, and hard coal ash, was obtained from the Renforth (*112*). In addition, the percentage of Ca and Mg in each waste stream was obtained from the literature (*112, 113*). Therefore, the total $CaCO_3$ and $MgCO_3$ that could be produced from these sources was calculated by first multiplying the mass of the mineral waste by the percent Ca and Mg content, and then multiplying that by the molar mass ratio of $MgCO_3$/MgO and $CaCO_3$/CaO. To determine the annual production of crushed concrete (which can also act as a carbonate-based aggregate source), the global average lifetime of residential (85 years), non-residential (50 years), and civil engineering (65 years) buildings, along with the global average market share of these concrete applications (30%, 32% and 37% respectively), was used to estimate the concrete stock reaching end-of-life in 2016 (*26*) as seen in equation S2.

$$Quantity\ of\ crushed\ concrete\ = \sum(P_{2016-t_{r,n,c}} * M_{r,n,c}) \qquad \text{S2}$$

Where P is the mass of concrete produced globally in the year 2016, $t_{r,n,c}$ and $M_{r,n,c}$ are the lifetimes and market shares of the residential, non-residential and commercial buildings, respectively.

Annual production of cement kiln dust (CKD) was determined using the CKD generation rate reported by the EPA (*114*), along with the global annual production of cement reported by the global cement and concrete association (GCCA) (*115*), these values can be found in supplemental data sheet 6.

S.5.2. Availability of agricultural and woody residue

Global annual production of crops and woody biomass was obtained from FAOStat (*116*), and the ratio of main crop to by-product is obtained from Shah *et al.* (*117*), and other sources (*118–120*). The moisture content and carbon content of the agricultural by-products was obtained from Phyliss 2 (*121*). The resulting carbon content available in agricultural residues was determined using equation S3.

$$\sum P_x * R_{x/y} * \left(1 - m_y\right) * C_y \qquad \text{S3}$$

Where P_x is the mass of crop x produced globally in a given year; $R_{x/y}$ is the ratio of main crop 'x' to by-product 'y'; m_y is the moisture content (%) of agricultural by-product 'y'; and C_y is the carbon content (in terms of dry weight %) of agricultural by-product 'y'. A similar approach was done for woody biomass; however, given that production quantities are reported in terms of volume, to determine the mass of woody residues, an average density of 0.29 tonne/m^3 was used, based on data from Phyliss 2 (*121*). The data for agricultural crop residues and woody biomass can be found in supplemental data sheet 6. To determine the availability of biochar, a similar equation is used (S4), where the dry mass of the residue is multiplied by the biochar yield (%). Biochar yield (B_y) for each residue was calculated using a pyrolysis model (*122*), assuming a 600°C-pyrolysis temperature, and utilizing average cellulose/hemicellulose/lignin and ash contents from Phyliss 2 (*121*).

$$\sum P_x * R_{x/y} * \left(1 - m_y\right) * B_y \qquad \text{S4}$$

The total resources required for global implementation of the carbon-sequestering materials examined herein was determined assuming a 1:1 replacement ratio of carbon (e.g., a bio-based plastic with a carbon content of 0.4 would require 0.4 kg of carbon from biomass). We note that, due to process inefficiencies, this conversion from feedstock to building materials might require higher levels of biomass/mineral resources. When assuming a 3:1 ratio of feedstock to product, we find that 100% replacement of plastics, brick, and asphalt binder in construction with bio-based counterparts would only require roughly 15% of global annual biomass residue production. However, we note that 72% of total biomass residues would need to be pyrolyzed to generate enough biochar for 15 wt% replacement of cement under this scenario. The results of this additional sensitivity analysis as well as the calculations for the resource availability assessment can be found in supplemental data sheet 6.

S.6. Comparison of global carbon storage potential to IPCC targets

In this study, total calculated carbon storage potential for the examined building materials is compared to the total carbon dioxide removal (CDR) needed to reach 1.5°C and 2°C targets as

reported in the IPCC sixth assessment report. For this analysis, we examine the minimum, median and maximum values as reported by the IPCC (see supplemental data sheet 7). To determine cumulative carbon storage by 2100, we assume a constant rate of material consumption (in line with quantities reported in supplemental data sheet 1) and assume full implementation of the carbon-storing technologies examined herein. A range of results is presented based on the minimum and maximum carbon contents of each material and their associated carbon storage potential. See supplemental data sheet 7 for these results. Further, a sensitivity analysis of these results is conducted, assuming a +/- 20% change in future material demand (see supplemental data sheet 8). Further, given that the production of plastics is expected to increase three-fold (46), an additional sensitivity analysis is done to examine the potential future contribution of bio-based plastics under this assumption.

Data S1. (separate file)
Global building material consumption and chemical emissions

Data S1. (separate file)
Technical replacement ability of carbon-storing materials in buildings

Data S3. (separate file)
Carbon dioxide storage potential for building materials

Data S4. (separate file)
Global carbon storage potential (minimum, average, and maximum values). Sensitivity of global carbon storage potential based on changes in material demand

Data S5. (separate file)
Sensitivity of assumptions on total global carbon storage potential and of the carbon storage potential per kg of material

Data S6. (separate file)
Resource availability assessment.

Data S7. (separate file)
Comparison of carbon storage potentials to IPCC targets (data for Figure 3).

Data S8. (separate file)
Sensitivity of carbon storage magnitudes compared to IPCC targets based on changes in material demand.

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