Supplemental Information

Cost-motivated pathways towards near-term decarbonization of the cement industry

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S.1. CO_2 Emission Calculations

As stated in the main text, we use average clinker fraction and kiln efficiencies reported in the Getting the Numbers Right (GNR) database to calculate cement emissions in our model.[1] In 2021, it was reported that the United States produced 73.6 Mt cement with an average of 89.44% clinker-to-cement mass ratio content in the cement.[1] The remaining 10.56% of cement is assumed to be gypsum, which serves to regulate the cement hardening time.[2,3] While these compositions are relevant for the baseline cement, it is also possible for alternative materials such as clay or LC[3] to replace clinker to limit CO_2 emissions from the calcination process.[1] This results in the following summation of cement materials that applies across all calculations used in this work:

$$1 = x_{\text{clinker}} + x_{clay} + x_{\text{gypsum}} \quad \text{(Eq. S1)}$$

Where x_{clinker} ($t_{clinker}$ / t_{cement}) is the mass fraction of clinker in cement, x_{clay} (t_{clay} / t_{cement}) is the mass fraction of clay in cement, and x_{gypsum} (t_{gypsum} / t_{cement}) is the mass fraction of gypsum in cement that is held equal to 0.1 for this study.

Given the cement produced, clinker content, and other raw material input properties, it is possible to calculate the process emissions via **Eq. S2**.

$$\xi_{\text{process}} = \frac{\left(\hat{m}_{\text{cement}}\right) M_{CO_2} f_{\text{lime}} x_{\text{clinker}}}{M_{\text{CaO}}} \quad \text{(Eq. S2)}$$

Where ξ_{process} (t_{CO_2} yr^{-1}) is the rate of process CO_2 emissions produced each year, $\hat{m}_{\text{cement}}$ (t_{cement} yr^{-1}) is the total mass of cement produced in a single year, M_{CO_2} (t_{CO_2} mol^{-1}) is the molecular weight of CO_2, M_{CaO} (t_{CaO} mol^{-1}) is the molecular weight of lime, f_{lime} (t_{CaO} $t_{clinker}^{-1}$) is the mass fraction of lime in clinker, and x_{clinker} ($t_{clinker}$ t_{cement}^{-1}) is the mass fraction of clinker in cement. The fraction of lime in clinker is typically 0.646.[4,5] As the clinker content is reduced, we can re-calculate these emissions using new x_{clinker} values. While this metric is reported in units of t_{CO_2} yr^{-1} for validation purposes, we can further normalize the emissions by dividing by the mass of cement produced each year to obtain emissions in t_{CO_2} t_{cement}^{-1}.

In addition to clinker emissions, the fuel emissions from heating the kiln to calcine the limestone (or clay if alternative SCMs are used) can be derived from reported emissions factors as in **Eq. S3**.

$$\xi_{\text{energy}} = \left(E_{\text{clinker}} x_{\text{clinker}} + E_{\text{clay}} x_{\text{clay}}\right)\left(\hat{m}_{\text{cement}}\right)\left(EF_{\text{fuel}}\right) \quad \text{(Eq. S3)}$$

Where ξ_{energy} (t_{CO_2} yr^{-1}) is the rate of CO_2 emissions associated with energy used per year, E_{clinker} (MJ $t_{clinker}^{-1}$) is the energy required to calcine the limestone to lime, E_{clay} (MJ t_{clay}^{-1}) is the energy required to calcine any clay used in the process, and EF_{fuel} (t_{CO_2} MJ^{-1}) is the emissions factor for the fuel of choice. To determine the energy required to calcine limestone and clay, we can refer to literature values that account for the inefficiencies of the kilns used in these processes. According to the GNR report, a short, dry kiln used in the limestone calcination process requires $E_{\text{clinker}} =$ 3.87 GJ $t_{clinker}^{-1}$.[1] Alternatively, calcining clay in a rotary

kiln requires $E_{clay} =$ 2.8 GJ t_{clay}^{-1} while calcining clay in a flash calciner requires $E_{clay} =$ 2 GJ t_{clay}^{-1}.[2] When considering alternative fuels, we assume that the same total energy is required to calcine clinker and/or clay and simply use the associated fuel emission factors to convert between the baseline and the alternative fuel. The emission factors of various fuels are included in **Table S1**. Additionally, for ease of costing, we also extracted relevant heating values and fuel costs in **Table S1**.

Table S1. Fuel metrics for all energy sources considered in this study. For this analysis, we assume the emission factor for coal is an average of the emissions factors for bituminous, subbituminous, and lignite coal. The emissions, heating values, and costs required for the fuels are well documented and widely available while those values for electricity are representative of predicted futuristic values such that all electricity comes from zero-emission sources.[6]

	Emissions Factor: EF ($t CO_2$ / GJ)[7]	**Heating Value: HV** (MJ / kg)	**Price:** C_{fuel} ($ / GJ)
Coal	0.0911	22.5[8]	3.77[9]
Natural Gas	0.0502	48.5[10]	1.90[11]
Petroleum Coke	0.0968	35[12]	1.94[13]
Biomass	0.0501	16.5[14]	4.66[15]
MSW*	0.0473	10[16]	7.40[17]
Electricity	0.0000	N/A	0.03 – 0.06 $/kWh

*MSW = Municipal solid waste

S.2. System Cost Expressions

In the main manuscript we outline that additional capital, operating, and capture costs must be included in estimating the total cost of cement production when alternative decarbonization retrofit options are pursued. The general cost expression (**Eq. 1**) can be further expanded as follows.

First, the capital costs for this system can be estimated from representative large-scale plants (**Table S2**). We can expand the capital costs according to **Eq. S4**.

$$C_{\text{CAPEX}} = C_{\text{CAPEX,Plant}} + C_{\text{CAPEX,LC}^3} + C_{\text{CAPEX, fuel}} + C_{\text{CAPEX, PSC}} \quad \text{(Eq. S4)}$$

Where $C_{\text{CAPEX,Plant}}$ ($\$\ t_{cement}^{-1}$) are the total costs to build the original cement plant and are constant across all scenarios studied, $C_{\text{CAPEX,LC}^3}$ ($\$\ t_{cement}^{-1}$) are the capital costs associated with altering the clinker fraction in the cement, $C_{\text{CAPEX, fuel}}$ ($\$\ t_{cement}^{-1}$) account for the increases in capital costs to change the fuel source, and $C_{\text{CAPEX, PSC}}$ ($\$\ t_{cement}^{-1}$) are the capital costs required to install PSC units. Because it is not possible to size a plant capable of producing all the U.S. cement demand, these capital costs are estimated for plants of a smaller cement production capacity and normalized by their individual production capacity. Specifically, the capital cost considered for this discussion are included in **Table S2** below.

Table S2. Capital cost breakdown for all baseline and additional technologies used in this work. Assumes a plant lifetime of 20 years.

	Capital Costs (M$)	**Plant Scale ($Mt_{cement}\ yr^{-1}$)**	**Amortized Capital Costs ($\$\ t_{cement}^{-1}$)**	**Reference**
Full Plant	341.0	0.99	17.29	*Thunder Said Energy*[18]
LC³ (flash calciner)	10.3	1	0.52	Scrivener et al.[2]
Fuel Mixture	11.3	2	0.28	CSI/ECRA-Technology Papers[19]
PSC	N/A	N/A	(41.85% of $60/tCO$_2$ is CAPEX for PSC)	Díaz-Herrera et al.[20]

In addition to the capital costs, we need to account for operating costs in estimating the total cost of the decarbonization technologies ($C_{\text{OPEX,fuel}}$ in $\$\ t_{cement}^{-1}$). We expand the operating costs as follows:

$$C_{\text{OPEX,fuel}} = C_{\text{fuel}}\left(\frac{1}{EF_{\text{fuel}}}\frac{\xi_{\text{energy}}}{\hat{m}_{\text{cement}}}\right) = C_{fuel}\left(E_{\text{clinker}}x_{\text{clinker}} + E_{\text{clay}}x_{\text{clay}}\right) \quad \text{(Eq. S5)}$$

Where C_{fuel} ($\$\ MJ^{-1}$) is the raw fuel cost to the cement manufacturer, EF_{fuel} (t_{CO_2} / MJ) is the emissions factor for the fuel of choice, ξ_{energy} ($t_{CO_2}\ yr^{-1}$) is the rate of CO_2 emissions associated with energy used per year, $\hat{m}_{\text{cement}}$ ($t_{cement}\ yr^{-1}$) is the total mass of cement produced in a single year, E_{clinker} (MJ / $t_{clinker}$) is the energy required to calcine the limestone to lime, E_{clay} (MJ / t_{clay}) is the energy required to calcine any clay used in the process, and all other variables are consistent with prior equations. Fuel costs in some cases were reported on a mass basis (i.e., $\$\ t_{fuel}^{-1}$), and thus to convert to an energy basis, we used the heating value

(HV, **Table S1**) to convert to a per energy basis. By costing and performing these calculations using knowledge of the energy required to calcine both limestone and clay, this automatically allows us to maintain a constant energy input but vary the mass of fuel needed to meet the energy needs.

In addition to energy costs, there are also raw material costs (Eq. S6):

$$C_{\text{OPEX,raw materials}} = C_{\text{limestone}}\left(\frac{M_{\text{limestone}}}{M_{lime}}\right)\left(\frac{\hat{m}_{\text{clinker}} f_{\text{lime}}}{\hat{m}_{\text{cement}}}\right) + C_{\text{LC}^3}\left(\frac{\hat{m}_{\text{LC}^3}}{\hat{m}_{\text{cement}}}\right) \quad \text{(Eq. S6)}$$

Where $C_{\text{OPEX,raw materials}}$ (\$ t_{cement}^{-1}) is the raw material operating costs of the system, $C_{\text{limestone}}$ (\$ $t_{limestone}^{-1}$) is the raw material costs for limestone, $M_{\text{limestone}}$ ($t_{limestone}\, mol_{limestone}^{-1}$) is the molecular weight of limestone, M_{lime} ($t_{lime}\, mol_{lime}^{-1}$) is the molecular weight of lime, $\hat{m}_{\text{clinker}}$ ($t_{clinker}$ yr^{-1}) is the total mass of clinker required in a single year, f_{lime} (t_{lime} $t_{clinker}^{-1}$) is the mass fraction of lime in clinker, $\hat{m}_{\text{cement}}$ (t_{cement} yr^{-1}) is the total mass of cement produced in a single year, C_{LC^3} (\$ $t_{LC^3}^{-1}$) is the raw material cost for alternative SCMs (LC^3), and $\hat{m}_{\text{LC}^3}$ (t_{LC^3} yr^{-1}) is the total mass of alternative SCMs (LC^3) required in a single year.

Finally, we must account for additional operating expenses associated with PSC and DAC. First we can express the total costs of PSC (Eq. S7 – Eq. S9) in order to further clarify trends observed in **Figure 2b**.

$$C_{\text{capture},PSC} = C_{CAPEX,PSC} + C_{OPEX,\,PSC} \quad \text{(Eq. S7)}$$

$$C_{CAPEX,PSC} = 0.4185\frac{C_{\text{PSC}}\xi_{\text{PSC, 90\%}}}{\hat{m}_{\text{cement}}} \quad \text{(Eq. S8)}$$

$$C_{OPEX,PSC} = \frac{C_{\text{PSC}}(1 - 0.4185)\xi_{\text{PSC}}}{\hat{m}_{\text{cement}}} \quad \text{(Eq. S9)}$$

Where $C_{\text{capture},PSC}$ (\$ t_{cement}^{-1}) is the total cost of implementing PSC, $C_{CAPEX,PSC}$ (\$ t_{cement}^{-1}) is the additional cost of cement production due to the capital costs associated with PSC, C_{PSC} (\$ tCO_2^{-1}) is the nominal cost of PSC at 90% removal, $\xi_{\text{PSC,90\%}}$ (tCO_2 yr^{-1}) is the total CO_2 captured via PSC in a year at the nominal 90% capture rate, $C_{OPEX,PSC}$ (\$ t_{cement}^{-1}) is the additional cost of cement production due to the operating costs associated with PSC, and ξ_{PSC} (tCO_2 yr^{-1}) is the total CO_2 captured via PSC in a year. The 0.4185 factor aligns with the PSC capital costs outlined in **Table S2** above. If we are solely capturing CO_2 via PSC and have a selected capture fraction (α_{PSC} (–)), the amount of CO_2 captured via PSC is equal to:

$$\xi_{\text{PSC}} = \xi_{\text{tot}}\alpha_{\text{PSC}} \quad \text{(Eq. S10)}$$

Where ξ_{tot} (tCO_2 yr^{-1}) are the total emissions remaining after any clinker or fuel replacements that need to be abated.

In the above expressions for determining the cost of PSC, we assume that regardless of the amount of CO_2 removed, the total capital costs associated with PSC will always apply, resulting in a constant value for $C_{CAPEX,PSC}$, resulting in a positive y-intercept value for the total costs as indicated in **Figure 2a** in the

main manuscript. Additionally, the operating costs will accumulate depending on the quantity of CO_2 removed, as given by a constant baseline cost of $C_{\mathrm{PSC}}(1-0.4185)$ in \$ tCO_2^{-1}. This constant rate of increase in costs manifests as a linear increase in costs for the metrics depicted in **Figure 2a** in the main manuscript.

Next, we can express the cost of DAC as in Eq. S11.

$$C_{\mathrm{capture},DAC} = C_{\mathrm{DAC}}\frac{\xi_{\mathrm{tot}}(1-\alpha_{\mathrm{PSC}})}{\hat{m}_{\mathrm{cement}}} \quad \text{(Eq. S11)}$$

Where C_{DAC} (\$ tCO_2^{-1}) is the cost of DAC and ξ_{DAC} (tCO_2 yr^{-1}) is the total CO_2 captured via DAC in a year.

The total CO_2 captured via DAC each year will account for the offset between the total CO_2 emitted from the cement process (either from process or energy emissions) and the CO_2 captured from PSC (if relevant). C_{PSC} and C_{DAC} are constant values and are calculated based on fully capturing the maximum amount of CO_2 allowed for the process of interest. Thus, if the amount of CO_2 captured via PSC is less than the rated capture amount, the actual cost of capture will be greater than C_{PSC}. In this study, our selected baseline cost for PSC is $C_{\mathrm{PSC}} =$ \$60 $^{t_{CO_2}\text{-}1}$ while that for DAC is $C_{DAC} =$ \$200 $^{t_{CO_2}\text{-}1}$.[21–24] The cost of PSC is varied to \$20 $^{t_{CO_2}\text{-}1}$ and \$40 $^{t_{CO_2}\text{-}1}$ in some of the studies to investigate the effects of lowering the price of capture.

In addition to these calculated costs, there are additional cement production costs that contribute to the final costs of plant operation including miscellaneous operating and electricity expenses.[18] Because it is difficult to estimate these based on throughput of the plant, we choose to estimate the costs in \$ t_{cement}^{-1} that each will contribute to the final baseline cost. Miscellaneous operating costs are estimated at \$20 t_{cement}^{-1} and the electricity costs are estimated at \$10 t_{cement}^{-1}. Given all these estimates, this leads to a cost breakdown for the baseline condition as in **Table S3**.

Table S3. Total cost breakdown of the baseline condition (i.e., 90% clinker, only coal fuel) for the cost of cement.

	Amortized Capital Costs (\$ t_{cement}^{-1})	**Fraction of Total Costs**
CAPEX	17.29	0.1731
Energy	13.14	0.1316
Raw Materials (limestone)	39.43	0.3949
Electricity	10	0.1001
Miscellaneous OPEX	20	0.2003
Total	99.86	1.00

To adjust the cost values outlined above from units of \$ t_{cement}^{-1} to units of \$ $^{t_{CO_2}\text{-}1}$, we simply apply the following equation:

$$\hat{C}_y = \frac{C_y \hat{m}_{\mathrm{cement}}}{\xi_{\mathrm{PSC}}} \quad \text{(Eq. S12)}$$

Where C_y (\$ t_{cement}^{-1}) is the total additional cost of pursuing "y" decarbonization method and $\hat{C}_y$ is the converted cost in units of \$ $t_{CO_2}^{-1}$. The subscript "y" can include any of the outlined decarbonization methods (i.e., clinker replacement, alternative fuels, PSC, or DAC). Thus, for processes with an upfront capital investment (i.e., PSC), when converting from \$ t_{cement}^{-1} to \$ $t_{CO_2}^{-1}$, this will result in a nonlinear cost curve as depicted in **Figure 2b**.

S.3. Additional Trends

Upon performing a brief sensitivity analysis of the cost of PSC and DAC CO_2 removal technologies, we produced the plot in **Figure S1**. This investigates the effects of lowering both the costs of PSC and DAC to 20 $t_{CO_2\text{-}1}$ and \$100 $t_{CO_2\text{-}1}$, respectively. We observe that the recommended decarbonization pathway (i.e., the options with the lowest total costs of production) remain the same as that observed in **Figure 3a**.

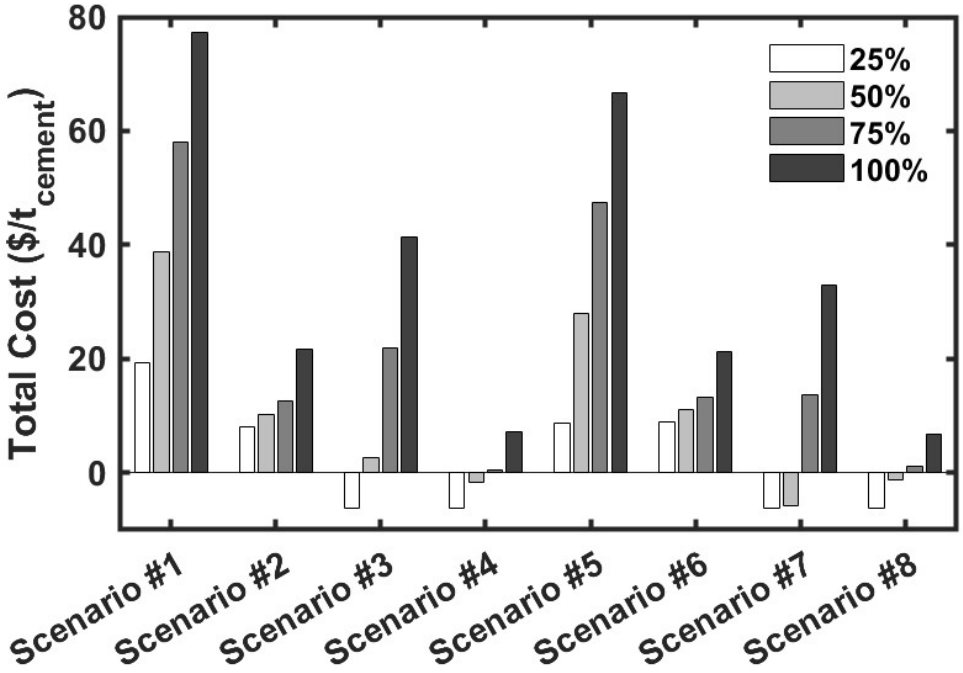

Figure S1. Total costs to a cement plant (\$ t_{cement}^{-1}) to achieve up to net-zero CO_2 removal assuming PSC = \$20 $t_{CO_2\text{-}1}$ and DAC = \$100 $t_{CO_2\text{-}1}$. The costs at 25%, 50%, 75%, and 100% removal are representative of actual costs that will be incurred by the cement manufacturing facility to achieve the desired CO_2 removal.

For a thorough inclusion of all analysis run in the main document, we included the full expansion of the cost curves associated with decarbonization Scenarios #1-8. This is presented in **Figure S2**.

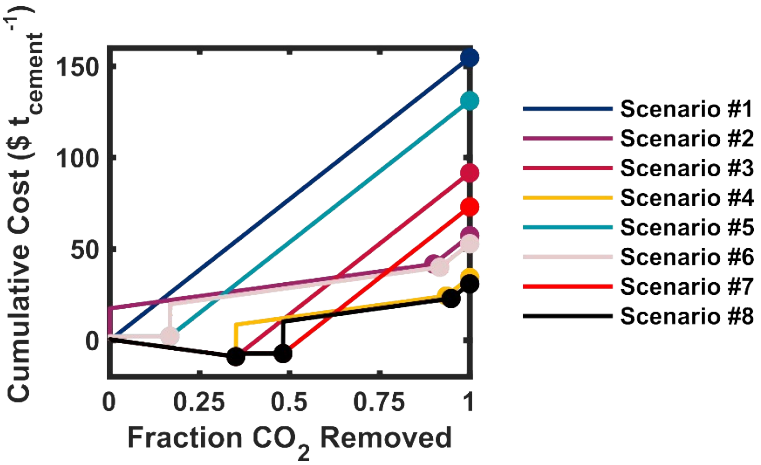

Figure S2. The total costs to a cement plant (\$ t_{cement}^{-1}) to achieve a fraction of CO_2 removal. This replicates Scenarios #4, 5, and 8 displayed in **Figure 3b**, however is expanded to include the trends of all scenarios.

In addition, we also investigated a scenario where the cost of DAC was lowered to \$100 $t_{CO_2}^{-1}$ while the cost of PSC was held at \$60 $t_{CO_2}^{-1}$. This reveals that the region above a removal fraction of 0.5 originally highlighted in **Figure 3b** is expanded due to the lower costs of DAC.

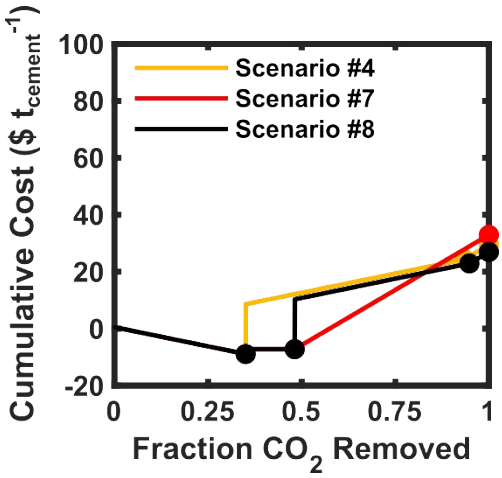

Figure S3. The total costs to a cement plant (\$ t_{cement}^{-1}) to achieve a fraction of CO_2 removal assuming PSC = \$60 $t_{CO_2}^{-1}$ and DAC = \$100 $t_{CO_2}^{-1}$.

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