

# 1 Environmental impacts of alternative cement binders

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8 **KEYWORDS.** Alternative cement; clinker; greenhouse gas emissions; environmental impacts;

9 mineral admixtures

10

11 **ABSTRACT.** Cement production is among the most difficult industrial activities to decarbonize.

12 Various measures have been proposed and explored to reduce its CO<sub>2</sub> emissions. Among these

13 measures, the substitution of portland cement (PC) clinker with alternative materials is arguably

14 the most effective, and consequently is an area of high research and commercial interest.

15 However, few studies have systematically quantified environmental impacts of alternative, i.e.,

16 non-PC, clinkers. Here, we quantify and compare environmental impacts arising from the

17 production of binders derived from several of the most commonly investigated alternative

18 cement systems. We show that binders derived from most of these alternative cements result in

19 lower greenhouse gas (GHG) emissions as well as other indicators of environmental impacts

20 relative to PC binder. The extent of these reductions varies as a function of energy requirements  
21 for production, process-related emissions from clinker formation, and raw materials demand.  
22 While utilization of alternative cements can be environmentally beneficial, similar reductions in  
23 GHG emissions can be achieved through use of partial replacement of PC with mineral  
24 admixtures. In this work, we quantitatively demonstrate the potential for alternative binders to  
25 mitigate environmental burdens and highlight the need to consider trade-offs among  
26 environmental impact categories when assessing these products.

27

## 28 **1. Introduction**

29 The production of portland cement (PC) has increased consistently throughout the past 100  
30 years, recently exceeding 4 billion tons annually <sup>1</sup>. This trend in production will likely continue  
31 into the future due to its essentially ubiquitous use in buildings and infrastructure <sup>2</sup>. Cement,  
32 including PC, is predominantly used in the production of concrete, the second most used material  
33 by mass after water <sup>3</sup>. Due to the massive scale of production, these materials directly account for  
34 7-8% of global anthropogenic CO<sub>2</sub> emissions <sup>4, 5</sup>, and a correspondingly notable climate change  
35 burden. With these substantial greenhouse gas (GHG) emissions coming from one material  
36 industry, mitigating these emissions has been a research focus for decades <sup>6</sup>. There are two  
37 primary sources for these emissions. The energy demand for cement production has alone  
38 contributed to 0.5-5% of annual global CO<sub>2</sub> emissions since the 1930s, and its relative  
39 contribution to annual CO<sub>2</sub> emissions has generally increasing over time <sup>7</sup>. However, the  
40 ‘process-based’ CO<sub>2</sub> emissions associated with the chemical conversion of CaCO<sub>3</sub> to CaO in a

41 cement kiln (i.e., calcination) currently leads to a higher proportion of CO<sub>2</sub> emissions than those  
42 related to its energy requirements <sup>8</sup>.

43 The relatively high proportion of process-based CO<sub>2</sub> emissions and the challenge to produce  
44 high technical performance cement at mass scale, has led to cement production being one of the  
45 most difficult to decarbonize industrial processes <sup>9</sup>. Roadmaps of technical measures to  
46 decarbonize cement production have been a key focus of technology driven climate mitigation  
47 literature. For example, the International Energy Agency (IEA) technology roadmap sets out a  
48 target to reduce total CO<sub>2</sub> emissions from the global cement industry by 24% by the year 2050 <sup>8</sup>.  
49 This roadmap assigns 37% of this reduction in CO<sub>2</sub> emissions to the use of alternative materials  
50 <sup>8</sup>. Several other technical measures are proposed to decarbonize this industry, including  
51 technology options like carbon capture and storage/utilization, and other measures like  
52 improving material efficiency (i.e., the utilization of less material for equivalent or improved  
53 performance, e.g., <sup>10</sup>). However, cement substitution (partial or full) is one of the most attractive  
54 technical measures: it is commonly implemented in industrial practice, and continues to remain  
55 central to industry and research efforts due to its relatively low cost and high potential to mitigate  
56 CO<sub>2</sub> emissions (e.g., <sup>11-14</sup>).

57 Despite the importance of cement substitution, there are presently few systematic  
58 environmental impact assessments of alternative cement binders. Here, we define an ‘alternative  
59 cement binder’ as a cement binder that can fully or partially replace conventional ordinary  
60 portland cement (OPC) or blended PC binders, but are not within established PC specifications  
61 <sup>15</sup>. There has been growing interest in the environmental impacts of alkali-activated materials <sup>16-</sup>  
62 <sup>18</sup>. However, here we focus on clinkered alternative cements, which undergo a pyroprocessing  
63 stage similar to OPC, yet are commonly perceived to have lower environmental impacts <sup>15</sup>. Such

64 alternative cements can contain similar or dissimilar clinker phases to conventional OPC, and  
65 these materials are capable of reacting (during curing) to produce a hardened binder. Differences  
66 in their chemical and mineral compositions relative to OPC can lead to reductions in thermal  
67 energy demand and process-based emissions, while retaining the potential for CO<sub>2</sub> uptake during  
68 curing. The chemistries of these cement systems are generally well-known (e.g., <sup>19-21</sup>), although  
69 understanding the effects of changing clinker composition on material properties in these  
70 systems remains more limited (e.g., <sup>22-24</sup>), as does a quantitative understanding of the  
71 environmental trade-offs from their production <sup>25, 26</sup>.

72 This study aims to quantify and compare the environmental burdens arising from the  
73 production of several alternative cement binders based on the following systems: reactive belite  
74 portland cement (RBPC); belite ye'elinite ferrite (BYF) cement; carbonatable calcium silicate  
75 cement (CCSC); magnesium oxide cement (MOM), and calcium sulfoaluminate-belite (C\$AB)  
76 cement. We compare each cement system to OPC, which is modeled here as 95% clinker and 5%  
77 gypsum, and to blended PC binders containing OPC and mineral admixtures at a 30 mass%  
78 replacement level. We perform this comparison using several environmental impact categories to  
79 avoid burden shifting and identify co-benefits among them.

80

## 81 **2. Materials and Methods**

### 82 **2.1. Scope of Assessment**

83 The goal of this study is to compare environmental burdens from the production of  
84 alternative cement binders to those arising from producing OPC binder. Since the technical  
85 performance of some of these alternative cement binders in mortar and concrete are not currently

86 fully characterized, our comparisons are based on a functional unit of the production of 1 kg of  
87 cement binder.

88 The scope of this work includes quantification of impacts from raw material acquisition  
89 through curing by means of carbonation or hydration (Fig. 1). Curing is incorporated into this  
90 assessment because it is anticipated to affect environmental impacts of the cement systems; the  
91 amount of water or CO<sub>2</sub> used in curing is calculated assuming a complete reaction of  
92 constituents takes place. We include blending with small quantities of gypsum as a mineral  
93 admixture, since this is typical practice to achieve desired mortar/concrete setting times;  
94 however, because the inclusion of other mineral admixtures in binders can vary widely, gypsum  
95 was the only mineral admixture assessed for the alternative cement systems. For the blended PC  
96 binder systems, the use of fly ash (FA), ground granulated blast furnace slag (GBS), natural  
97 pozzolans (NP), and calcined clay (CC) are considered as mineral admixtures, discussed in  
98 Section 2.3.7. While there is the potential to mitigate environmental impacts from more efficient  
99 utilization of cement at various stages of its life cycle, i.e., from raw material acquisition through  
100 disposal (e.g., <sup>5, 27, 28</sup>), this work focuses on potential mitigation during production. Therefore,  
101 this work involves a cradle-to-binder production gate assessment, meaning that carbonation by  
102 water-cured concrete during service, the roles of different cementitious properties on in-use  
103 performance, and the effects of other use and disposal related environmental impacts are  
104 excluded.

105 The environmental impact categories considered here were quantified using the 2008 United  
106 States version of the United States Environmental Protection Agency (EPA) Tool for Reduction  
107 and Assessment of Chemicals and Other Environmental Impacts (TRACI) <sup>29</sup>. In this method, ten  
108 environmental impact categories are considered. These include: (i) acidification; (ii)

109 eutrophication; (iii) global climate change; (iv) ozone depletion; (v) human health impacts from  
110 respiratory effects of particulate matter; (vi) human health impacts from cancer toxicity; (vii)  
111 human health impacts from non-cancer toxicity; (viii) eco-toxicity; (ix) photochemical smog  
112 formation; and (x) resource depletion, with a focus on fossil fuel use. Contributions to  
113 environmental impacts are discussed both in terms of: (a) origin – namely, process-based  
114 emissions, referring to those derived from chemical or production processes, and energy-based  
115 emissions, referring to those derived from energy utilization; and (b) production stage including  
116 raw material acquisition, transportation, energy (including both thermal and electricity),  
117 calcination, and curing.

118

## 119 **2.2. Assessing Clinker Phases**

### 120 **2.2.1. Chemical Compounds for Clinker Production**

121 Clinker is a calcined, quenched material that when ground finely can be used as a reactive  
122 constituent in cement. All cements considered here contain a clinker that has been pyroprocessed  
123 to gain reactivity. To quantify the energy requirement to carry out pyroprocessing for each  
124 cement system, i.e., the chemical reactions that take place in cement kilns at elevated  
125 temperatures, enthalpies of reactions for the different chemical conversions (raw materials to  
126 clinker phases; referred to herein as manufacturing enthalpy) were determined. We calculated  
127 these enthalpies of reactions, using enthalpy of formation data at 25°C, and based on common  
128 raw materials and clinker phase compositions for each cement system assessed (data are  
129 presented in Table 1; discussion of calculations are presented in the Supplementary Information).  
130 These calculations also quantified the masses of CO<sub>2</sub> emitted by these chemical reactions.

131 We limit our calculations to raw materials commonly used in clinker production based on  
132 similar assumptions in other work (e.g., <sup>21</sup>): CaCO<sub>3</sub> as the main calcium source (representing  
133 limestone); Al<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>(OH)<sub>4</sub> (representing clay), Al(OH)<sub>3</sub> (representing bauxite), and SiO<sub>2</sub>  
134 (representing siliceous rock) as the alumina and silica sources; and Fe<sub>2</sub>O<sub>3</sub> (representing iron ore)  
135 as the iron source. We used two sulfur sources for the ye'elimite clinker phase: CaSO<sub>4</sub>  
136 (anhydrite) and CaSO<sub>4</sub>.H<sub>2</sub>O (gypsum). These two sources lead to differences in enthalpies of  
137 reaction and CO<sub>2</sub> emissions for the production of ye'elimite due to water evaporation. Similarly,  
138 for the MgO clinkers, two sources of raw materials were considered. The first was MgCO<sub>3</sub>,  
139 which in its conversion to MgO, results in process-based CO<sub>2</sub> emissions. This system is hereafter  
140 referred to as MOMC. The other source considered was Mg<sub>2</sub>SiO<sub>4</sub>, which does not result in  
141 process-based CO<sub>2</sub> emissions to produce MgO (hereafter referred to as MOMS); however,  
142 efficient conversion methods are not currently known for this balance at industrial scale <sup>21</sup>,  
143 meaning that our results for this system should be considered as a theoretical exercise only.  
144

145 **Table 1.** Molecular masses and standard enthalpies of formation for clinker phases.

Molecular masses & standard enthalpies of formation	M (g/mol)	$\Delta H_f^0$ (kJ/mol)	$\Delta H_f^0$ (kJ/g)
Alite, (C <sub>3</sub> S)	228	-2931	-12.9
Belite, (C <sub>2</sub> S)	172	-2308	-13.4
Aluminate, (C <sub>3</sub> A)	270	-3561	-13.2
Ferrite, (C <sub>4</sub> AF)	486	-5080	-10.5
Quicklime, (C)	56	-635.1	-11.3
Wollastonite, (CS)	116	-1635	-14.1
Ye'elimite (C <sub>4</sub> A <sub>3</sub> \$) [from CaSO <sub>4</sub> .2H <sub>2</sub> O]	610	-8406	-13.8
Ye'elimite (C <sub>4</sub> A <sub>3</sub> \$) [from CaSO <sub>4</sub> ]	610	-8406	-13.8
Periclase (MgO) [from MgCO <sub>3</sub> ]	40	-601.6	-15.0
Periclase (MgO) [from Mg <sub>2</sub> SiO <sub>4</sub> ]	40	-601.6	-15.0
other (modeled as gypsum)	172	-2023	-11.8
Al <sub>2</sub> Si <sub>2</sub> O <sub>5</sub> (OH) <sub>4</sub> , kaolinite ('clay')	258	-4119	-16.0
MgCO <sub>3</sub> , magnesite	84	-1113	-13.3
Mg <sub>2</sub> SiO <sub>4</sub> , forsterite	140	-2173	-15.5
SiO <sub>2</sub> , quartz ('chert')	60	-910.7	-15.2

CaSO <sub>4</sub> ·2H <sub>2</sub> O, gypsum	172	-2023	-11.8
CaSO <sub>4</sub> , anhydrite	136	-1434	-10.5
Fe <sub>2</sub> O <sub>3</sub> , hematite ('iron ore')	160	-826.2	-5.2
CaCO <sub>3</sub> , calcite ('limestone')	100	-1207	-12.1

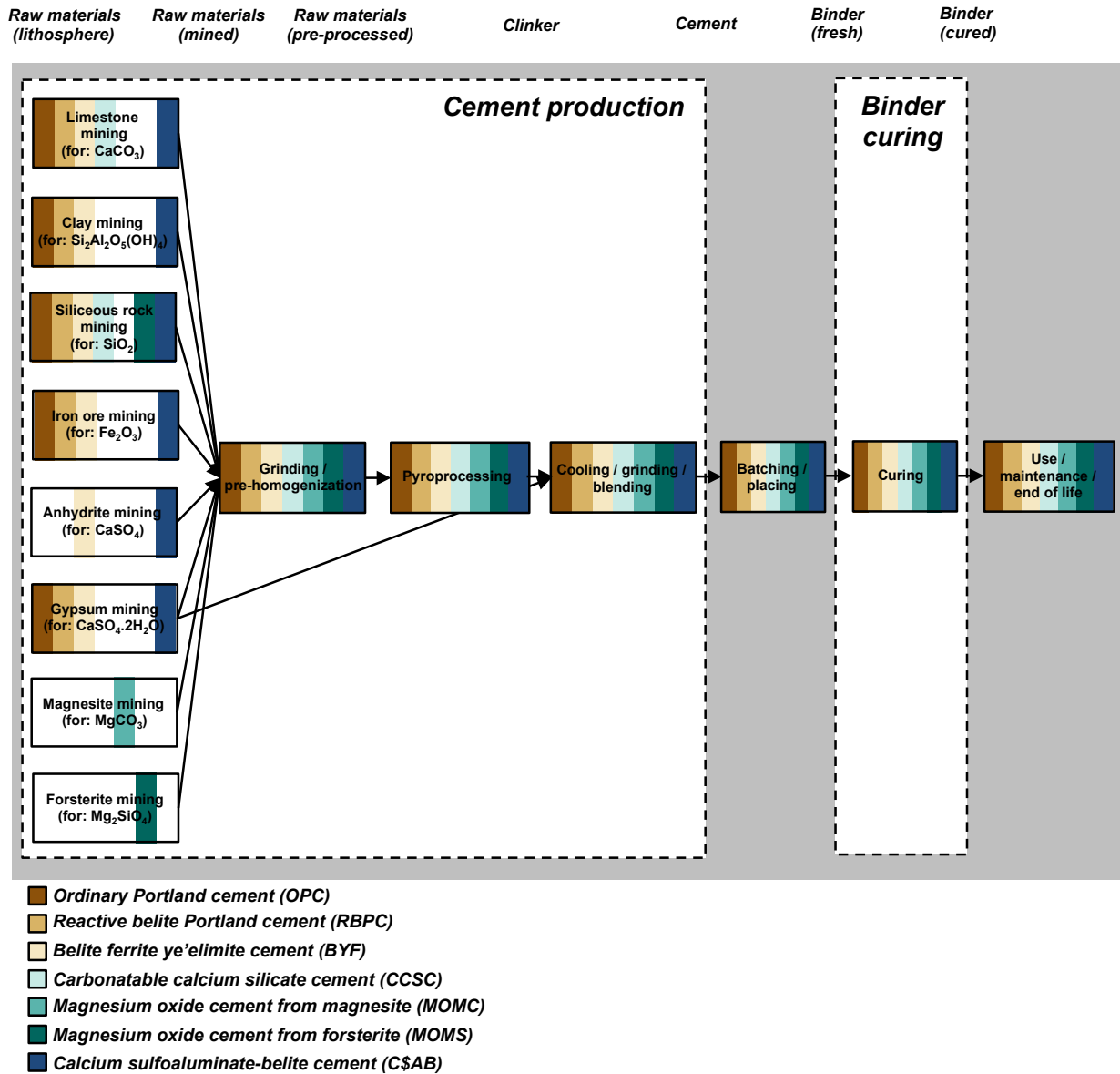
146 <sup>a</sup> Based on stoichiometric balances and enthalpies presented in the Supplementary Information.  
147 Note: These nomenclature are based on standard abbreviations listed in the Supplementary  
148 Information.

149

## 150 **2.2.2. Raw Materials for Clinker and Cement Production**

151 In order to form the necessary reactive phases in each clinker, raw materials must be used as  
152 discussed in Section 2.2.1. As noted previously, we modeled a setting control admixture as  
153 gypsum, which we used as an input to inter-grinding, which is a process that follows  
154 pyroprocessing (Fig. 1). To quantify environmental impacts for raw material acquisition and  
155 processing, data were based on models from Long Trail Sustainability's adaptation of the  
156ecoinvent database to U.S. production <sup>30</sup>.





157

158 **Figure 1.** Block flow diagrams showing the scope of assessment for each alternative cement  
 159 binder examined; transportation considered between raw material acquisition and cement  
 160 production; stages highlighted in gray were considered outside the scope of assessment.

161

### 162 2.2.3. Clinker Production Processes

163 While clinkering conditions vary (e.g., kiln efficiency), we assumed that cement production  
 164 occurs in Northern California to facilitate a consistent comparison among alternative cement

165 binders. In California, only kilns containing both preheater and precalciner stages are used <sup>31</sup>, so  
166 this type of kiln was modeled here. The fuel mix modeled was based on the United States  
167 average kiln fuel mix as reported by the United States Geological Survey for dry kilns <sup>1</sup>. In this  
168 work, we consider the lower enthalpy of production for the alternative clinkers to result in a  
169 percent reduction in total kiln thermal energy demand. Environmental impacts associated with  
170 electricity demand through pre-grinding and homogenization, kilning, cooling, and grinding  
171 were assessed based on electricity requirements from Marceacu *et al.* <sup>32</sup> and modeled  
172 consistently in each cement system. The electricity mix was modeled as the California electricity  
173 grid (based on California Energy Commission <sup>33</sup>).

174 All raw materials for the production of clinker were assumed to be transported a distance of  
175 500 km from the extraction site to the cement production site. This value exceeds common  
176 assumptions for transporting of limestone to cement kilns (e.g., approximately 100 km <sup>34</sup>) and  
177 the average of survey values for transportation distances for raw materials to cement kilns in the  
178 United States <sup>32</sup>. However, because some of the raw materials examined here are less widely  
179 available than limestone, a longer average transportation distance was assumed. Transportation  
180 of the FA, GBS, NP, and CC was based on their originating locations, as discussed in Section  
181 2.3.7, in some cases exceeding the values modeled for the raw materials. Transportation by rail  
182 was assumed, and a United States average freight train model was used <sup>30</sup>.

183 Beyond the aforementioned stages to produce cement systems, process-based emissions  
184 during cement kilning were assessed. These process-based emissions include CO<sub>2</sub> from the same  
185 chemical reactions that produce clinker phases, which were based on stoichiometric balances  
186 using the phases shown in Table 1.

### 187 **2.3. Cement Specific Production Considerations**

188 Considering the different types of cement systems, as well as sources and types of raw  
 189 materials utilized in their production, many permutations in composition and environmental  
 190 impacts can be expected. Therefore, we simplified our analysis using various assumptions for the  
 191 production processes and raw materials included. We discuss these assumptions in the  
 192 subsequent sections (Sections 2.3.1 to 2.3.7). The shorthand notation used to refer to each  
 193 cement system is presented in Table 2. Phase compositions of each alternative clinker-based  
 194 cement are shown in Table 3.

195 **Table 2.** Alternative cement nomenclature.

Cement type	Definition
OPC	Ordinary portland cement
RBPC	Reactive belite portland cement
BYF - HY	Belite ye'elimite ferrite cement, 46% belite, 35% ye'elimite
BYF - LY	Belite ye'elimite ferrite cement, 60% belite, 20% ye'elimite
C\$AB - HS	Calcium sulfoaluminate–belite cement, high sulfate content
C\$AB - LS	Calcium sulfoaluminate–belite cement, low sulfate content
CCSC	Carbonatable calcium silicate cement
MOMC	Magnesium oxide cement from MgCO <sub>3</sub>
MOMS	Magnesium oxide cement from Mg <sub>2</sub> SiO <sub>4</sub>
PC - FA	Blended PC cement with 30% fly ash by weight
PC - GBS	Blended PC cement with 30% ground granulated blast furnace slag by weight
PC - NP	Blended PC cement with 30% natural pozzolans by weight
PC - CC	Blended PC cement with 30% calcined clay by weight

196

197 **Table 3.** Alternative cement phase compositions.

clinker type	OPC	RBPC	BYF - HY	BYF - LY	C\$AB HS	C\$AB LS	CCSC	MOMC	MOMS
Alite, (C3S)	0.63	0.16	0	0	0	0	0	0	0
Belite, (C2S)	0.15	0.62	0.46	0.6	0.22	0.71	0	0	0
Aluminate, (C3A)	0.08	0.08	0	0	0	0	0	0	0
Ferrite, (C4AF)	0.09	0.09	0.17	0.17	0.03	0.07	0	0	0
Quicklime, (CaO)	0	0	0	0	0	0	0	0	0
Wollastonite, (CS)	0	0	0	0	0	0	1	0	0
Ye'elimite (C4A3\$)	0	0	0.35	0.2	0.65	0.15	0	0	0
Periclase (MgO) [from MgCO <sub>3</sub> ]	0	0	0	0	0	0	0	1	0
Periclase (MgO) [from Mg <sub>2</sub> SiO <sub>4</sub> ]	0	0	0	0	0	0	0	0	1
other (modeled as gypsum)	0.05	0.05	0.02	0.03	0.09	0.07	0	0	0

198 \* Approximations for mineral additives are based on <sup>23, 35-45</sup>.

199 **2.3.1. Ordinary Portland Cement**

200 The phase composition used for OPC results in a net reaction enthalpy of 1.67 MJ per kg of  
201 clinker produced. However, for the kilns modeled in this work (which typically operate under  
202 steady state conditions at  $\sim 1450$  °C), the energy required to produce OPC clinker is 3 MJ per kg  
203 (based on the efficiency of a preheater/precalciner kiln from <sup>46</sup>). In a cement plant, this remaining  
204 energy is either utilized elsewhere in the plant (i.e., heat integration) or wasted, such that the  
205 energy efficiency of clinker production can exceed 80% of the theoretical maximum <sup>47</sup>. Overall,  
206 these thermal energy requirements, and the fuel sources utilized to satisfy them, account for a  
207 significant portion of the GHG emissions arising from cement production. They have therefore  
208 been a key target area for mitigation strategies <sup>8</sup>. For the OPC phase composition considered  
209 here,  $\sim 0.5$  kg of process-based CO<sub>2</sub> is emitted per kg of cement produced (i.e., excluding CO<sub>2</sub>  
210 emissions from energy, transportation, and material acquisition).

211

### 212 **2.3.2. Reactive Belite Portland Cement**

213 In contrast to the OPC phase composition, RBPC predominantly contains belite, with alite as  
214 the main secondary phase (Table 3), and similar amounts of minor phases (typically 5-20  
215 mass%). This change in alite/belite composition ratio results in a reduction in net reaction  
216 enthalpy of 14%. We model this as a 14% reduction in the thermal energy requirements for the  
217 kiln. Additionally, this change reduces process-based CO<sub>2</sub> emissions by 6%.

218

### 219 **2.3.3. Belite Ye'elimite Ferrite Cement**

220 While phase compositions of all clinkers analyzed here normally show modest variation,  
221 BYF and C\$AB (Section 2.3.4) clinkers are known to have broader ranges of phase  
222 compositions. In order to capture this, we modeled two characteristic clinker phase compositions

223 for BYF clinker. The first composition contained a high amount (35 mass%) of ye'elimite,  
224 whereas the second contained less (20 mass%) of ye'elimite and more belite (60 mass%)<sup>21</sup>. As a  
225 result, the BYF cements assessed in this work require ~25% lower net reaction enthalpy than  
226 OPC, and emit 20-27% less process-based CO<sub>2</sub>.

227

#### 228 **2.3.4. Calcium Sulfoaluminate-belite Cement**

229 A similar approach to modeling C\$AB to BYF was used, since these clinkers also contain  
230 belite, ferrite, and ye'elimite, albeit in different ratios: the C\$AB considered herein contain 22-71  
231 mass% belite, 3-7 mass% ferrite, and 15-65 mass% ye'elimite. These materials contain lower  
232 ferrite and higher ye'elimite than the BYF clinkers assessed. We used clinker phase  
233 compositions from<sup>43</sup>. Our calculations show that the C\$AB cements assessed here require 27-  
234 37% lower net reaction enthalpy and emit 18-48% less process-based CO<sub>2</sub> than OPC clinker.

235

#### 236 **2.3.5. Carbonatable Calcium Silicate Cement**

237 CCSC cement was modeled as wollastonite (CaSiO<sub>3</sub>), based on similar assumptions in the  
238 literature (e.g.,<sup>21</sup>). Current mining of this mineral is limited to only a few countries<sup>48</sup>; therefore,  
239 here we modeled the production of wollastonite with raw material inputs of SiO<sub>2</sub> (siliceous rock)  
240 and CaCO<sub>3</sub> (limestone). We calculate a 54% lower net enthalpy of reaction, and 26% net lower  
241 process-based CO<sub>2</sub> emissions, than OPC. Additionally, because this material can be solidified  
242 through carbonation, the hardening process for this cement results in an uptake of carbon dioxide  
243 during curing (~0.2 kg of CO<sub>2</sub> per kg of cement<sup>42</sup>).

244

#### 245 **2.3.6. Magnesium Oxide Cement**

246 For the MgO based cements, as was noted previously, two sources of magnesium were  
247 considered: Mg<sub>2</sub>SiO<sub>4</sub> (forsterite, in the production of MOMS clinker) and MgCO<sub>3</sub> (magnesite, in  
248 the production of MOMC clinker). Mg<sub>2</sub>SiO<sub>4</sub> is common worldwide; however, technologies to  
249 efficiently convert Mg<sub>2</sub>SiO<sub>4</sub> to MgO do not currently exist <sup>21</sup>. The use of forsterite as a raw  
250 material for MgO-based cements leads to lower net reaction enthalpy and process-based CO<sub>2</sub>  
251 emissions than OPC clinker by 56% and 100%, respectively. Conversely, using MgCO<sub>3</sub> as the  
252 raw material source releases 2.1 times more process-based CO<sub>2</sub> emissions than for OPC clinker,  
253 as well as leading to a 76% higher net enthalpy of reaction. However, as with CCSC, MgO-based  
254 cement can be solidified through carbonation, and the process-based CO<sub>2</sub> that can be absorbed  
255 through this reaction can account for ~0.5 kg of CO<sub>2</sub> per kg of cement.

256

### 257 **2.3.7. Blended Portland Cement Systems**

258 Cements are typically blended with moderate fractions of mineral admixtures (either before  
259 or during batching), which contribute beneficially to the performance of concrete. Among the  
260 most commonly discussed mineral admixtures for their potential to mitigate GHG emissions  
261 from concrete production are FA, GBS, NP, and CC. To investigate the effects of their use on  
262 environmental impacts relative to the alternative cement systems (discussed above), we assessed  
263 blended PC binders comprising 30% replacement of these mineral admixtures for OPC. To  
264 model NP, the electricity demand from material acquisition was based on the University of  
265 California GreenConcrete Tool <sup>49</sup>, and because NP can be acquired in California, the State's  
266 electricity grid was used (from <sup>33</sup>). FA is not produced in significant quantities in California, so it  
267 was modeled as produced in Wyoming (energy demand for capture from <sup>49</sup>, electricity grid from  
268 <sup>50</sup>). Similarly, GBS is not produced in California, so it was modeled as produced in Pennsylvania

269 (energy demand from <sup>49</sup>, electricity grid from <sup>51</sup>). For this work, no impacts from production of  
270 the primary good, either coal-electricity or iron, were allocated to either the FA or the GBS.  
271 Finally, while CC is not produced in large quantities in California, there is potential for it to be  
272 produced locally in the near future, therefore, local production as assumed. For the production of  
273 CC, because kilning is required, the same electricity requirement as for clinker production was  
274 modeled (based on <sup>32</sup> and <sup>33</sup>). The kiln energy requirement, which is lower than that of  
275 conventional clinker, was based on <sup>52</sup>, using the same fuel mix as before (from<sup>1</sup>).

276

#### 277 **2.4. Sensitivity Analysis**

278 We performed a sensitivity analysis, with  $\pm 15\%$  change in model input parameter values  
279 (based on ranges used in assessment of alternative cements <sup>25, 26</sup>), to capture several effects: the  
280 manufacturing maturity of the alternative cements considered here, not all of which are currently  
281 used at industrial scale; and quantitative variability in the inventory used. This includes  
282 sensitivity of the inventory to raw materials selection. Here, raw materials for the ye'elimite,  
283 aluminate, and ferrite phases were modeled on the most prevalent or least expensive options,  
284 assuming that industrial production of cement systems requires inexpensive raw material  
285 acquisition. For this reason, the primary sulfur source for ye'elimite was modeled as gypsum,  
286 and the primary aluminum source for aluminate and ferrite was modeled as kaolinite (clay).  
287 However, it is possible to use other mineral resources, so in our sensitivity assessment we  
288 additionally considered anhydrite as a sulfur source and bauxite (modeled here as gibbsite) as an  
289 aluminum source. Our sensitivity analysis also considers kiln efficiency. Here, we assumed that a  
290 reduction in enthalpy of kiln product produced results in the same relative reduction in kiln  
291 thermal energy demand. As a sensitivity consideration, the inefficiency of the kiln, calculated as

292 the (absolute) difference between the kiln energy demand and the manufacturing enthalpy for  
293 OPC clinker, was considered to be the same for each cement system assessed. Therefore,  
294 differences in kiln energy represent changes in manufacturing enthalpy only.

295

### 296 **3. Results**

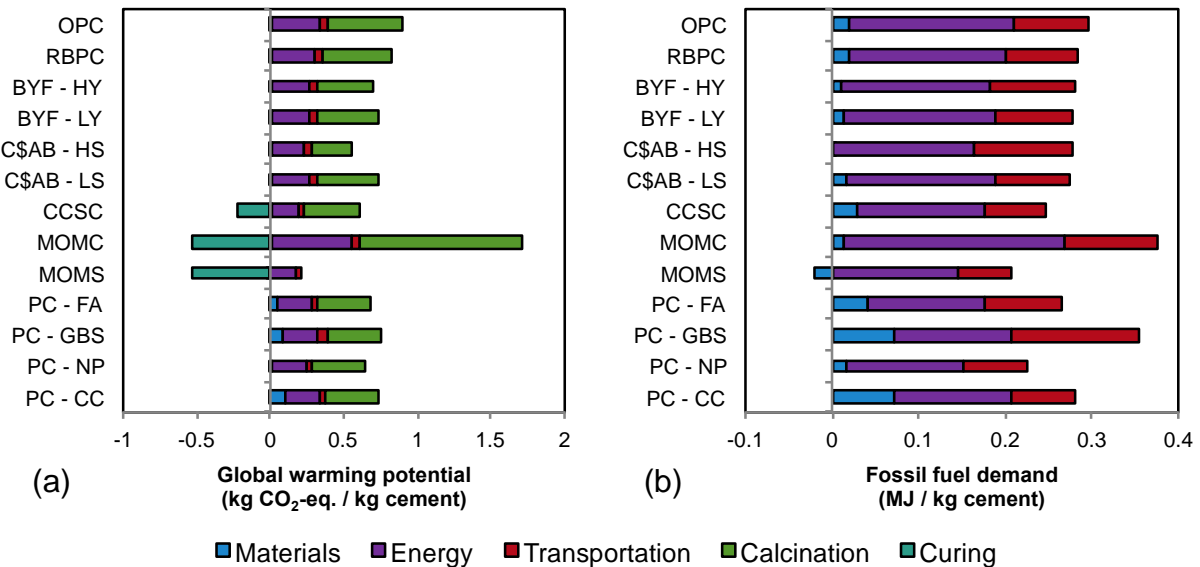
#### 297 **3.1. Greenhouse Gas Emissions**

298 Our results show that, with the exception of the MOMC system, all RBPC, BYF, CCSC,  
299 MOMS, and C\$AB binders modeled result in reductions of GHG emissions relative to OPC  
300 binder (Fig. 2). The lowest GHG emissions were determined for the MOMS system;  
301 substantially lower than any of the blended PC systems. We calculate negative GHG emissions  
302 for this binder (i.e., overall uptake of GHG emissions),  $-0.33$  kg CO<sub>2</sub>-eq. per kg cement;  
303 however, without CO<sub>2</sub> curing, MOMS results in  $0.19$  kg CO<sub>2</sub>-eq. per kg cement. This value, i.e.,  
304 cured with water rather than CO<sub>2</sub>, is still the lowest GHG emissions determined for the binders  
305 assessed here. The blended PC systems with 30% mineral admixture replacement result in 17 to  
306 29% reduction in GHG emissions, but all have higher emissions than the MOMS, CCSC, and  
307 high sulfate content C\$AB (C\$AB – HS) binders.

308 We calculate the highest GHG emissions for MOMC binder. This material emits  $1.2$  kg CO<sub>2</sub>-  
309 eq. per kg cement with CO<sub>2</sub> curing, and  $1.7$  kg CO<sub>2</sub>-eq. per kg cement without CO<sub>2</sub> curing.  
310 MOMC has the highest energy-derived GHG emissions as well as the highest process-based  
311 GHG emissions. This difference is noteworthy as both the highest and lowest GHG emissions  
312 modeled were for MgO-based cements, which highlights the significance of raw materials  
313 selection in developing alternative binders with lower GHG emissions.



314 With the exception of the MOMS system, which we modeled as having no process-based  
 315 CO<sub>2</sub> emissions, process-based emissions exceeded the energy-derived GHG emissions for OPC,  
 316 RBPC, CCSC, and MOMC binders, ranging from 48-96% of the total GHG emissions. For the  
 317 BYF and C\$AB binders, the energy-derived emissions had similar or higher contributions to  
 318 GHG emissions than process-based emissions, ranging from 34-46% of the total. Relatively low  
 319 GHG emissions were calculated for raw materials production (1-3% of total GHG emissions)  
 320 and transportation of materials (5-11% of total GHG emissions). Variations in transportation  
 321 emissions predominantly reflect the quantities of raw materials used.



322  
 323 **Figure 2.** (a) Global warming potential and (b) embodied energy (in terms of fossil fuel demand)  
 324 for ordinary portland cement and alternative cement binders.

325  
 326 **3.2. Energy Demand**

327 Similar to GHG emissions, the lowest fossil fuel demand was determined for the MOMS  
 328 binder (0.19 MJ per kg cement) due to the low net reaction enthalpy required (Fig. 2). The

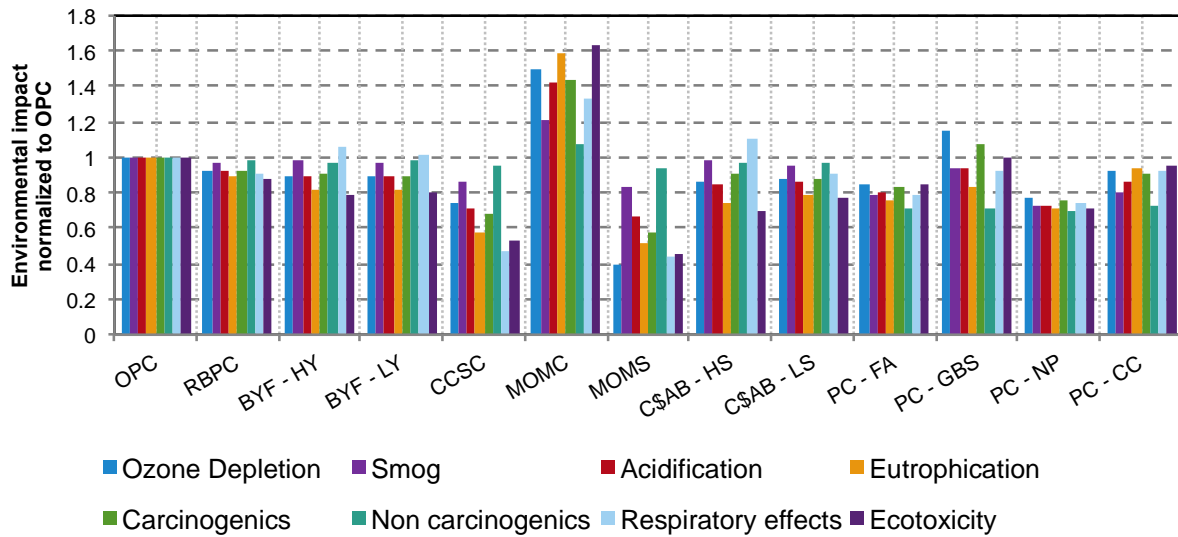
329 highest fossil fuel demand was determined for the MOMC binder (0.38 MJ per kg cement). The  
330 thermal energy and electricity required for raw material grinding, pre-homogenization, pyro-  
331 processing, cooling, and inter-grinding with mineral admixtures resulted in the majority of fossil  
332 fuel demand for each binder system (59-79% of the total fossil fuel demand). Raw materials  
333 production was responsible for less than 12% of the total fossil fuel demand, with the exception  
334 of the blended PC systems involving GBS and CC as raw material inputs – the relatively high  
335 polluting electricity grid for GBS and kiln energy for CC results in these blended PC systems  
336 having higher energy demand. Unlike GHG emissions, 24-35% of the fossil fuel demand arose  
337 from transportation of materials; again, variations predominantly reflect the different masses of  
338 raw materials transported.

339

### 340 **3.3. Other Environmental Impacts**

341 Most of the alternative binders assessed here show lower environmental impacts than OPC in  
342 impact categories other than climate change (Fig. 3). The production of MOMS binder  
343 consistently causes the lowest indicator values out of all the systems analyzed. Relative to OPC  
344 binder, impact category indicators for MOMS are: ozone depletion, 60% lower; smog formation,  
345 17% lower; acidification, 33% lower; eutrophication, 49% lower; carcinogenics, 42% lower;  
346 non-carcinogenics, 6% lower; respiratory effects, 56% lower; and eco-toxicity, 54% lower.  
347 Conversely, MOMC binder typically caused the highest indicator values for these environmental  
348 impact categories, exceeding the values for OPC binder in each category. With the exception of  
349 the MOMS and the CCSC, the remaining cement systems resulted in similar impacts to OPC  
350 outside of GHG emissions and fossil fuel demand. Some of these cement systems, such as the  
351 PC-NP binder, resulted in up to a 30% reduction in impact category indicator values, due to the

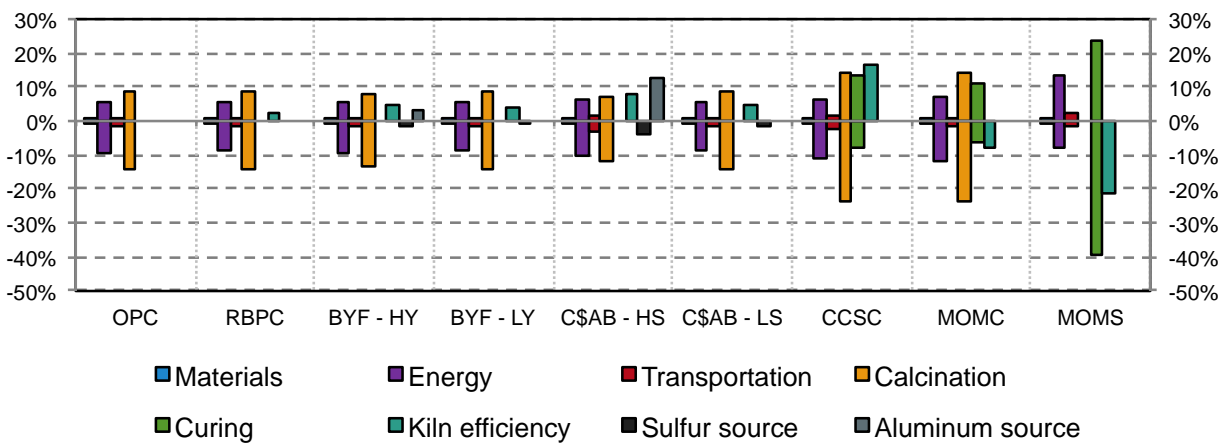
352 near negligible impacts of NP relative to OPC. For most of the cement systems, less than 15%  
 353 reduction in the remaining environmental impact categories was calculated. Notably, the cements  
 354 containing ye’elimité showed a potential to increase respiratory effects, ranging from no increase  
 355 to up to an 11% increase. The PC blended systems that had high transportation impacts or energy  
 356 for material production/acquisition also showed the potential for increases in environmental  
 357 impacts, namely, the PC-GBS system had higher ozone depletion and carcinogenic impacts than  
 358 OPC binder.



359  
 360 **Figure 3.** Environmental impacts associated with the production of ordinary portland cement and  
 361 alternative cement binders, including: (a) ozone depletion, smog, acidification, and  
 362 eutrophication, and (b) carcinogenics, non-carcinogenics, respiratory effects, and eco-toxicity.  
 363 The environmental impacts are normalized to the production of ordinary portland cement. Non-  
 364 normalized environmental factors are presented in the Supplementary Information.

365  
 366 **3.4. Sensitivity Analysis**

367 We present the sensitivity of GHG emissions to changes in model parameters in Fig. 4, with  
 368 the remaining sensitivity analysis results shown in the Supporting Information. Changing raw  
 369 material masses and transported distances of materials do not significantly affect the calculated  
 370 GHG emissions, for each binder system assessed here:  $\pm 15\%$  changes in these parameters cause  
 371 up to 2-3% change in overall GHG emissions. These parameters have less influence in systems  
 372 with relatively higher GHG emissions from fuels and calcination (e.g., MOMC).



373  
 374 **Figure 4.** Sensitivity analysis of global warming potential from ordinary portland cement and  
 375 alternative cement binders varying material resources, energy modeling assumptions, and input  
 376 parameters by  $\pm 15\%$ .

377  
 378 Conversely, changes to energy- and process-based emissions can significantly influence  
 379 overall binder production impacts. Changes of  $\pm 15\%$  in energy demand resulted in 5-13%  
 380 changes in overall GHG emissions across the binders assessed; changing kiln inefficiency leads  
 381 to greater impacts, by up to +21%. Changing process-based CO<sub>2</sub> emissions had a notable, but  
 382 lower influence, resulting in increases of up to 24% in GHG emissions across the binders  
 383 assessed. In these cases, the lower and higher limits correspond to MOMS (for which there are

384 no process-based CO<sub>2</sub> emissions) and MOMC (for which there are high process-based CO<sub>2</sub>  
385 emissions). For the cement systems with carbon curing, namely the CCSC, MOMS, and MOMC,  
386 there was a high sensitivity to variation in curing, namely 7-39% change in GHG emissions.  
387 Even with these variations, the lowest GHG emissions remain to be for the MOMS followed by  
388 the CCSC system. The use of anhydrite as a source of sulfur resulted in up to -4% change in  
389 GHG emissions and use of bauxite in addition to clay as an aluminum source resulted in up to a  
390 +12% change in GHG emissions. While these are smaller than the energy-related sensitivities for  
391 GHG emissions, the selections of sulfur and aluminum sources had a significant effect on other  
392 environmental impacts (see Supplementary Information). These findings again highlight the  
393 importance of raw materials selection on environmental impacts of cement binder production.

394

#### 395 **4. Discussion**

396 Deep decarbonization of the cement cycle requires a range of solutions throughout the life  
397 cycles of products such as buildings and infrastructure, both within cradle-to-binder production  
398 life cycle stages as analyzed here, but also in later life cycle stages<sup>53</sup>. Based upon our results, it  
399 is clear that the potential benefits of altering binder chemistry to most current alternatives will  
400 not be able to solely mitigate environmental impacts attributable to cement to the levels required  
401 by environmental targets (e.g.,<sup>8</sup>). However, smaller yet still ambitious goals to reduce cement  
402 cycle CO<sub>2</sub> emissions by targeting the binder production stage are possible. For example, the IEA  
403 has set forth goals to achieve a 24% reduction in CO<sub>2</sub> emissions from cement production from  
404 current levels by 2050<sup>8</sup>. The IEA proposes 37% of these emissions reductions to be achieved by  
405 lowering clinker content in cements beyond current levels by 2050<sup>8</sup>; however, this does not  
406 consider the use of lower-emissions clinkers. Here, we highlight that it is possible to reduce

407 environmental impacts by substitution of PC clinker demand with alternative clinkers. The IEA's  
408 24% CO<sub>2</sub> emissions reduction goal could be met by replacement of OPC (or the equivalent  
409 quantity of conventional clinker) by 43% CCSC (with CO<sub>2</sub> curing), 18% MOMS (with CO<sub>2</sub>  
410 curing, 31% without), or 63% C\$AB – HS. In order to achieve the same reductions in GHG  
411 emissions as modeled in the IEA roadmap <sup>8</sup> with substitution of OPC by mineral admixtures  
412 only, significantly greater levels than current use would be needed. Using the emissions from  
413 processing and transporting mineral admixtures, as applicable, from Section 2, increased use of  
414 FA would need to be 28%, use of GBS by 33%, use of NP by 24%, or CC by 37% beyond  
415 current levels. Therefore, substituting OPC and blended PC binders for alternative binders may  
416 aid in reaching these emissions goals.

417 We calculate that the IEA's 24% CO<sub>2</sub> emissions reduction goal may theoretically be achieved  
418 solely through increased use of some of these alternative binders, assuming that they all provide  
419 the same levels of service (i.e., technical performance here). It should be noted that  
420 supplementary cementitious materials are typically used to improve performance of cement-  
421 based systems. Some of the alternative cements explored in this work are currently being used at  
422 an industrial scale, namely the RBPC and C\$AB cements <sup>21</sup>. RBPC has shown to have the  
423 potential for low early strength, but high late strength and low heat of hydration making it  
424 especially desirable in mass concrete applications <sup>21</sup>. C\$AB cements have high early strength and  
425 several good durability properties; their potentially undesirable expansive behavior can be  
426 controlled <sup>43</sup>. There has been less study on the performance of the other alternative clinkers.  
427 Initial study has suggested that BYF cement systems have good sulfate resistance and  
428 dimensional stability, but little is known about other durability properties <sup>21</sup>. CCSC cement  
429 systems have shown to be able to attain good strengths when cured in CO<sub>2</sub> rich chambers and

430 have certain beneficial durability properties <sup>21</sup>. MgO-based cements have shown to have very  
431 low strength when cured via hydration; however, carbon-curing of these cement systems can lead  
432 to higher strength cements than conventional cements <sup>54</sup>. We note that CO<sub>2</sub>-cured cements are  
433 unsuitable for use in conventional steel reinforced concrete systems due to their lower pH.  
434 Because the effects on performance from use of some of the alternative binders presented in this  
435 work are not fully known, this presents an area for future study. Factors such as these highlight  
436 the importance of contextualizing our results within the life cycles of cement products.

437 In selecting potential material alternatives, mineral resource availability must be considered.  
438 The main raw materials required for the production of OPC are limestone and kaolin, which are  
439 considered to be abundant global resources <sup>21, 55, 56</sup>. These same raw materials are used for the  
440 production of RBPC and CCSC, although CCSC can also be made with the more rare  
441 wollastonite deposits <sup>21, 57</sup>. BYF and C\$AB cement systems predominately require limestone and  
442 clays, but they also need sulfur and alumina resources. If low levels of sulfur or alumina are  
443 needed, waste streams or impure resources may be utilized, but if resources like bauxite are  
444 necessary, issues may arise as these resources are more rare and are needed in other industries <sup>58</sup>.  
445 Such issues are not as pronounced for the acquisition of iron substrates for the ferrite phases, but  
446 these ores are still not as common as limestone <sup>59</sup>. For the MgO-based cement systems, there is  
447 theoretically enough magnesium resources globally, but energy-efficient methods to extract  
448 MgO remain limited <sup>21</sup>. With the exception of clays, the availabilities of many well-  
449 known/commonly used supplementary cementitious materials are limited <sup>60</sup>, meaning that  
450 substantially increasing the extent to which they are used is challenging. Furthermore, our results  
451 show that high polluting grids and high transportation distances of supplementary cementitious  
452 materials can outweigh their benefits in other environmental impact categories; notably, the PC–

453 GBS system exhibited higher carcinogenic and ozone depletion impacts. Therefore, the use of  
454 alternative cements, either alone or themselves blended with mineral admixtures, represent  
455 additional technically viable pathways to reducing GHG emissions in the cement cycle.

456

457

## 458 ASSOCIATED CONTENT

459 **Supporting Information.** A PDF file is supplied as Supporting Information. It includes  
460 Supplementary Methods, Supplementary Data Tables, Supplementary Figures, and  
461 Supplementary References.

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470 to the final version of the manuscript.

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#### 478 ABBREVIATIONS

479 PC, portland cement; OPC, ordinary portland cement; RBPC, reactive belite portland cement;  
480 BYF – HY, belite ye’elinite ferrite cement with high ye’elinite content; BYF – LY, belite  
481 ye’elinite ferrite cement with low ye’elinite content; C\$AB – HS, calcium sulfoaluminate–  
482 belite cement with high sulfate content; C\$AB – LS, calcium sulfoaluminate–belite cement with  
483 low sulfate content; CCSC, carbonatable calcium silicate cement; MOMC, magnesium oxide  
484 cement from magnesium carbonate; MOMS, magnesium oxide cement from magnesium silicate;  
485 FA, fly ash; GBS, ground granulated blast furnace slag; NP, natural pozzolan; CC, calcined clay;  
486 PC – FA, portland cement with fly ash; PC – GBS, portland cement with ground granulated blast  
487 furnace slag; PC – NP, portland cement with natural pozzolans; PC – CC, portland cement with  
488 calcined clay.

489

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