Substantial global carbon uptake by cement carbonation

Fengming Xi[1,2,3], Steven J. Davis[1,4], Philippe Ciais[5], Douglas Crawford-Brown[6], Dabo Guan[7], Claus Pade[8], Tiemao Shi[3], Mark Syddall[6], Jie Lv[9], Lanzhu Ji[1], Longfei Bing[1], Jiaoyue Wang[1], Wei Wei[10], Keun-Hyeok Yang[11], Björn Lagerblad[12], Isabel Galan[13], Carmen Andrade[14], Ying Zhang[15] and Zhu Liu[16,17]★

Calcination of carbonate rocks during the manufacture of cement produced 5% of global CO_2 emissions from all industrial process and fossil-fuel combustion in 2013[1,2]. Considerable attention has been paid to quantifying these industrial process emissions from cement production[2,3], but the natural reversal of the process—carbonation—has received little attention in carbon cycle studies. Here, we use new and existing data on cement materials during cement service life, demolition, and secondary use of concrete waste to estimate regional and global CO_2 uptake between 1930 and 2013 using an analytical model describing carbonation chemistry. We find that carbonation of cement materials over their life cycle represents a large and growing net sink of CO_2, increasing from 0.10 GtC yr^{-1} in 1998 to 0.25 GtC yr^{-1} in 2013. In total, we estimate that a cumulative amount of 4.5 GtC has been sequestered in carbonating cement materials from 1930 to 2013, offsetting 43% of the CO_2 emissions from production of cement over the same period, not including emissions associated with fossil use during cement production. We conclude that carbonation of cement products represents a substantial carbon sink that is not currently considered in emissions inventories[1,3,4].

A tremendous quantity of cement has been produced worldwide for the construction of buildings and infrastructure, namely: 76.2 billion tons of cement between 1930 and 2013, and 4.0 billion tons in 2013 alone[1]. When making cement, the high-temperature calcination of carbonate minerals (for example, limestone rocks) produces clinker (mainly calcium oxide), and CO_2 is released into the atmosphere from this process. These 'process' CO_2 emissions from cement production (as opposed to related emissions from fossil-fuel energy that may have been used during cement production) comprise approximately 90% of global CO_2 emissions from all industrial processes and 5% of global CO_2 emissions

from industrial processes and burning fossil fuels combined[2-4]. Cumulative cement process emissions are estimated to have released 38.2 Gt CO_2 from 1930 to 2013[2-4].

However, the calcium oxide in cement materials is not stable over time, and cement hydration products gradually reabsorb atmospheric CO_2 through a physiochemical process called carbonation[5-8]. Carbonation occurs when CO_2 diffuses into the pores of cement-based materials and reacts with hydrated products in the presence of pore water[8,9] (see Methods). The carbonation starts at the surface of the concrete or mortar and progressively moves inwards. Although carbonation reactions are known to civil engineers due to their effects on the strength and safety of structures[5,10], the resulting large-scale CO_2 uptake flux has not been quantified. In contrast to the instantaneous emissions of CO_2 during manufacture of cements, carbonation is a slow process that takes place throughout the entire life cycle of cement-based materials[5,11]. The CO_2 uptake through carbonation of cement materials is thus proportional to the time-integral of cement consumption. Previous studies have applied a life cycle assessment to estimate concrete carbon sequestration over 100–200-year timescales[5,11,12]. However, these studies were limited to concrete materials in specific regions, and did not account for CO_2 uptake in other types of cement materials found in built infrastructure: cement mortar, construction cement waste, and cement kiln dust worldwide.

Based on new data sets compiled from field surveys in China and a comprehensive synthesis of existing data and studies (see Methods), we modelled the global atmospheric CO_2 uptake by four different cement materials (concrete, mortar, construction cement waste, and cement kiln dust) between 1930 and 2013 in four regions (China, the US, Europe, and the rest of the world) and analysed the sensitivity of our uptake estimates to 26 different variables (see Methods).

[1]Institute of Applied Ecology, Chinese Academy of Sciences, Shenyang 110016, China. [2]Key Laboratory of Pollution Ecology and Environmental Engineering, Chinese Academy of Sciences, Shenyang 110016, China. [3]College of Architecture and Urban Planning, Shenyang Jianzhu University, Shenyang 110168, China. [4]University of California, Irvine, Department of Earth System Science, Irvine, California 92697, USA. [5]Laboratoire des Sciences du Climat et de l'Environnement, CEA-CNRS-UVSQ, CE Orme des 14 Merisiers, 91191 Gif sur Yvette Cedex, France. [6]Cambridge Centre for Climate Change Mitigation Research, Department of Land Economy, University of Cambridge, 19 Silver Street, Cambridge CB3 9EP, UK. [7]School of International Development, University of East Anglia, Norwich NR4 7TJ, UK. [8]Danish Technological Institute, Gregersensvej, 2630 Taastrup, Denmark. [9]College of Economy and Management, Shenyang Agricultural University, Shenyang 110000, China. [10]CAS Key Laboratory of Low-carbon Conversion Science and Engineering, Shanghai Advanced Research Institute, Chinese Academy of Sciences, Shanghai 201203, China. [11]Department of Plant Architectural Engineering, Kyonggi University, Gwanggyosan-ro 154-42, Yeongtong-gu, Suwon, Kyonggi-do 16227, South Korea. [12]Swedish Cement and Concrete Research Institute, CBI Betonginstitutet, SE-100 44 Stockholm, Sweden. [13]Department of Chemistry, University of Aberdeen, Aberdeen AB24 3UE, UK. [14]Eduardo Torroja Institute CSIC, Serrano Galvache 4, 28033 Madrid, Spain. [15]Shenyang Pharmaceutical University, Shenyang 110016, China. [16]Resnick Sustainability Institute, California Institute of Technology, Pasadena, California 91125, USA. [17]John F. Kennedy School of Government, Harvard University, Cambridge, Massachusetts 02138, USA. *e-mail: zhuliu@caltech.edu
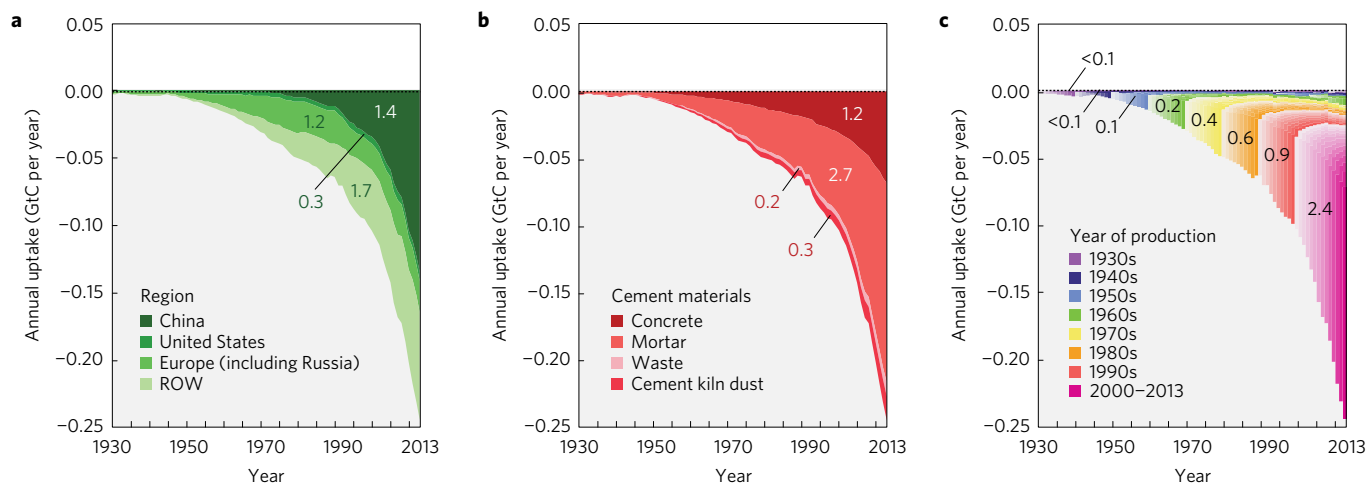
Figure 1 | Annual carbon sequestration by cement 1930–2013. a–c, Worldwide annual uptake of atmospheric CO_2 by cement, disaggregated by regions (**a**), by cement materials (**b**) and by years of which the cement produced (**c**). The numbers in each panel indicate the cumulative carbon sequestration (median values from our uncertainty analysis). ROW, rest of world.

Details of our calculations are available in the Methods. In summary, carbon sequestration from concrete was calculated from three stages in the life cycle of this material: service life, demolition, and secondary use of concrete waste[5]. In each case, we estimated exposed surface areas[5,10], thicknesses[10,13], exposure conditions including atmospheric CO_2 concentrations in different regions[5,9,14,15], and exposure time[5,16–18], then modelled carbon uptake by applying Fick's diffusion law[15] and concrete carbonation rate coefficients derived from both experimental measurements[5,18,19] and an extensive review of relevant literature[15,20]. The effect of different concrete strength classes, exposure conditions, additions, and coatings were explicitly modelled[5]. Exposure time in service life (t) was assumed to be the average building lifetime, ranging from 35 to 70 years[5,10–12], and carbon sequestration in demolition and secondary use stages was modelled assuming a spheric concrete shape for particles in waste[21], with carbonation fractions affected by waste concrete treatment methods, waste concrete particle size[13,22], and changing exposure conditions during phases of demolition and either reuse or disposal[13]. The carbon sequestration from mortar was calculated based on mortar utilization thickness[23] and annual carbonation depth using Fick's diffusion law. The carbon uptake from construction cement waste and cement kiln dust was estimated using the generation rate and carbonation fraction[24,25]. Model uncertainties and sensitivities to assumptions were evaluated by a Monte Carlo analysis that varies 26 individual parameters over 100,000 iterations (see Methods).

We find that a large fraction of global cement process CO_2 emissions, both cumulatively and annually in recent years, are reabsorbed by carbonation of cement materials. Figure 1a shows the annual carbon sequestration by cement materials between 1930 and 2013, disaggregated by world region. Based on our uncertainty analysis, we find a mean estimated global carbon uptake by all cement materials was 0.24 Gt C ($2\sigma = \pm10.0\%$) in 2013. Prior to 1982, the majority of sequestration occurred in Europe and the US, corresponding to the legacy carbon sink of cement building and infrastructure built during the 1940s and 1950s (Fig. 1a,c). Since 1994, cement materials used in China have absorbed more CO_2 than the other regions combined, due to its rapidly increasing cement production (Fig. 1a). Mortar cement consistently sequestered the most carbon, even though only ∼30% of cement is used in mortar (Fig. 1b). This is because mortar is frequently applied in thin decorative layers to the exterior of building structures, with higher exposure surface areas to atmospheric CO_2, and thus higher
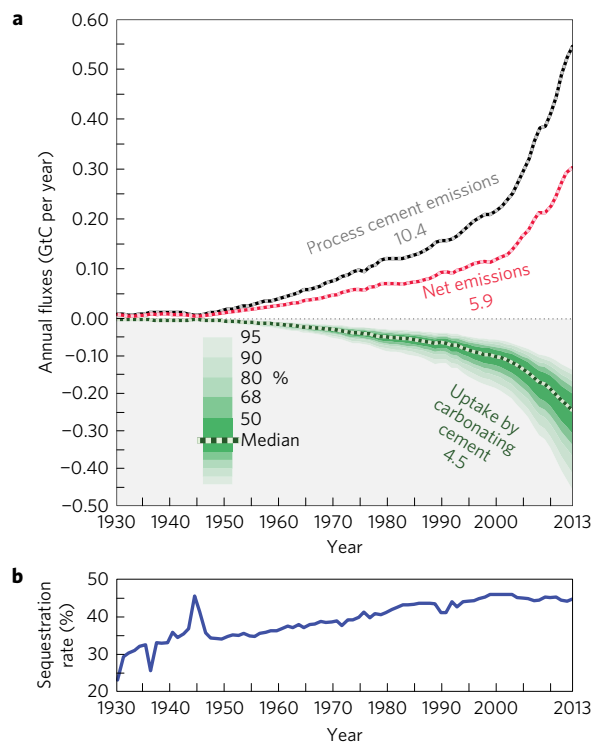

Figure 2 | Net cement emissions and annual sequestration rate 1930–2013. a, Between 1930 and 2013, 10.4 GtC was emitted by the cement industrial process[2] (dashed black line). Over the same period, however, carbonating cements absorbed 4.5 GtC (2.8–7.5 GtC, $p = 0.05$, green lines), or 43% of the cumulative cement emissions. **b**, Existing cement is thus a large and overlooked carbon sink, sequestering roughly 44% of cement emissions each year since 1980.

carbonation rate coefficients (see Supplementary Data)[23]. Despite a relatively smaller exposure area, and therefore lower carbonation rate coefficients, concrete is the second largest contributor to the carbon sink because ∼70% of all produced cement is used in concrete. Figure 1c shows the legacy effects of accumulating cement stocks; on average, between 2000 and 2013, 25.0% of the carbon sequestered each year was absorbed by cement materials produced
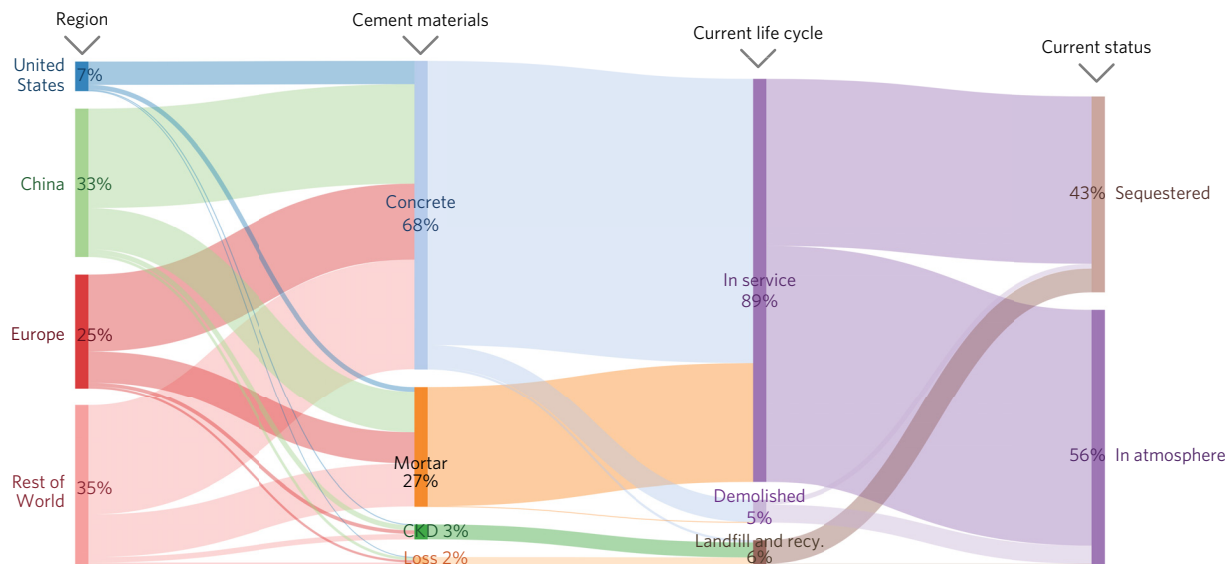
Figure 3 | Allocations of global historical cement process emissions 1930–2013. Between 1930 and 2013, 7%, 33%, 25% and 35% carbon dioxide emissions from cement production are from United States, China, Europe, and rest of world, respectively (Region). The emissions are 68% from concrete, 27% from mortar, 2% from loss cement in construction stage and 3% from CKD generation (Cement materials). The emissions are 89% in service life cement, 5% attributed to demolished cement, and 6% attributed to demolition cement landfill and recycling (Current life cycle). The emissions are 43% are sequestered by cement materials and 57% are remaining in atmosphere (Current status).

more than five years earlier and 14% produced more than ten years earlier. Demolition causes an increase in carbonation rates by exposing large and fresh surfaces. Because the average 35-year service lifetime of structures in China[16] is shorter than the average 65–70 years in the US[17] and Europe[5], the turnover of cement with respect to carbonation has been increasing over time, accelerating the uptake of CO_2 (Fig. 1c).

Figure 2 shows the net annual CO_2 emissions related to industrial process of cement production minus the estimated annual CO_2 sequestration from carbonation of cement materials. Between 1990 and 2013, the annual carbon uptake has been increasing by 5.8% per year on average, slightly faster than process cement emissions over the same period (5.4% per year; Fig. 2a). Using a bookkeeping model to estimate the carbon sink in established buildings and infrastructure each year, we estimate that a cumulative amount of 4.5 GtC (2.8–7.5, $p = 0.05$) has been sequestered by cement materials since 1930. The annual carbonation carbon sink increased from 0.10 GtC yr^{-1} in 1998 to 0.25 GtC yr^{-1} in 2013, which is consistent with previous estimations of carbon sequestration from cement-based materials (0.1–0.2 GtC yr^{-1}) from 1926 to 2008[7]. In total, we estimate that roughly 43% of the cumulative cement process emissions of CO_2 produced between 1930 and 2013 have been reabsorbed by carbonating cement materials, with an average of 44% of cement process emissions produced each year between 1980 and 2013 offset by the annual cement carbonation sink (Fig. 2b).

Figure 3 traces the cumulative cement process CO_2 emissions between 1930 and 2013 according to regional production and use of cement in different materials, and to the life cycle of each type of materials. In the case of concrete, an average of 16.1% of the initial emissions are absorbed during the service life of the material, with an additional 1.4% being absorbed during the demolition of cement structures and another 0.1% absorbed during the disposal or reuse of the concrete waste. In the case of mortar cement, an average of 97.9% of the annual initial emissions is absorbed during the material's service life, and the remaining 2.1% is absorbed in the demolition stage (Fig. 3). Given expected demolition, waste disposal, and reuse of cement materials from the large amount of concrete structures and infrastructure built in the past half-century,

and the still-increasing cement consumption in China and other developing countries, the carbon sink of cement materials can be anticipated to increase in the future.

Although the Intergovernmental Panel on Climate Change (IPCC) Guidelines for National Greenhouse Gas Inventories provides methods for quantifying CO_2 emissions during the cement production process, they do not consider carbon absorbed by carbonation of cement materials. Furthermore, the rate of sequestration by carbonating cement is increasing rapidly (by an average of 5.8% per year during the period 1990–2013) as the stock of cement buildings and infrastructure increases, ages and gets demolished and disposed. The overall size of the cement sink between 1930 and 2013 is significant for the global carbon cycle. We estimate that the global carbon uptake by carbonating cement materials in 2013 was approximately 2.5% of the global CO_2 emissions from all industrial processes and fossil-fuel combustion in the same year[2], which is equivalent to 22.7% of the average net global forest sink from 1990 to 2007[26]. The cement carbon sink of China alone in 2013 was about 0.14 GtC yr^{-1}, which accounts for 54% to 74% of the average net annual carbon sink in terrestrial ecosystems during the 1980s and 1990s[27].

It is well known that the weathering of carbonate and silicate materials removes CO_2 from the atmosphere on geologic timescales (10^4 years)[28]. However, the potential for removal by the weathering of cement materials has only recently been recognized[29]. Our results indicate that such enhanced weathering is already occurring on a large scale; existing cement stocks worldwide sequester approximately 1 billion tons of atmospheric CO_2 each year. Future emissions inventories and carbon budgets may be improved by including this cement sink. Moreover, efforts to mitigate CO_2 emissions should prioritize the reduction of fossil-fuel emissions over cement process emissions, given that produced cement entails creation of concomitant carbon sink. Indeed, if carbon capture and storage technology were applied to cement process emissions, the produced cements might represent a source of negative CO_2 emissions[30]. Finally, policymakers might productively investigate ways to increase the completeness and rate of carbonation of cement waste (for example, as a part of an enhanced weathering scheme)[31] to further reduce the climate impacts of cement emissions.

Methods

Methods, including statements of data availability and any associated accession codes and references, are available in the online version of this paper.

References

1. *Cement Statistics and Information* (USGS, 2015); http://minerals.usgs.gov/minerals/pubs/commodity/cement/index.html
2. Boden, T. A., Marland, G. & Andres, R. J. *Global, Regional, and National Fossil-Fuel CO_2 Emissions* (Carbon Dioxide Information Analysis Center, 2014).
3. *CO_2 Emissions From Fuel Combustion* (International Energy Agency, 2014).
4. Barcelo, L., Kline, J., Walenta, G. & Gartner, E. Cement and carbon emissions. *Mater. Struct.* **47,** 1055–1065 (2014).
5. Pade, C. & Guimaraes, M. The CO_2 uptake of concrete in a 100 year perspective. *Cement Concr. Res.* **37,** 1348–1356 (2007).
6. Huntzinger, D. N., Gierke, J. S., Kawatra, S. K., Eisele, T. C. & Sutter, L. L. Carbon dioxide sequestration in cement kiln dust through mineral carbonation. *Environ. Sci. Technol.* **43,** 1986–1992 (2009).
7. Renforth, P., Washbourne, C. L., Taylder, J. & Manning, D. Silicate production and availability for mineral carbonation. *Environ. Sci. Technol.* **45,** 2035–2041 (2011).
8. Johannesson, B. & Utgenannt, P. Microstructural changes caused by carbonation of cement mortar. *Cement Concr. Res.* **31,** 925–931 (2001).
9. Papadakis, V. G., Vayenas, C. G. & Fardis, M. N. Experimental investigation and mathematical modeling of the concrete carbonation problem. *Chem. Eng. Sci.* **46,** 1333–1338 (1991).
10. Gajda, J. *Absorption of Atmospheric Carbon Dioxide by Portland Cement Concrete* (Portland Cement Association, 2001).
11. Andersson, R., Fridh, K., Stripple, H. & Häglund, M. Calculating CO_2 uptake for existing concrete structures during and after service life. *Environ. Sci. Technol.* **47,** 11625–11633 (2013).
12. Dodoo, A., Gustavsson, L. & Sathre, R. Carbon implications of end-of-life management of building materials. *Resour. Conserv. Recy.* **53,** 276–286 (2009).
13. Engelsen, C. J., Mehus, J., Pade, C. & Sæther, D. H. *Carbon Dioxide Uptake in Demolished and Crushed Concrete* (Norwegian Building Research Institute, 2005).
14. Yoon, I.-S., Çopuroğlu, O. & Park, K.-B. Effect of global climatic change on carbonation progress of concrete. *Atmos. Environ.* **41,** 7274–7285 (2007).
15. Monteiro, I., Branco, F., Brito, J. d. & Neves, R. Statistical analysis of the carbonation coefficient in open air concrete structures. *Const. Build. Mater.* **29,** 263–269 (2012).
16. Hu, M., Bergsdal, H., van der Voet, E., Huppes, G. & Müller, D. B. Dynamics of urban and rural housing stocks in China. *Build. Res. Inf.* **38,** 301–317 (2010).
17. Kapur, A., Keoleian, G., Kendall, A. & Kesler, S. E. Dynamic modeling of In-Use cement stocks in the United States. *J. Ind. Ecol.* **12,** 539–556 (2008).
18. Yang, K.-H., Seo, E.-A. & Tae, S.-H. Carbonation and CO_2 uptake of concrete. *Environ. Impact Assess.* **46,** 43–52 (2014).
19. Galan, I., Andrade, C., Mora, P. & Sanjuan, M. A. Sequestration of CO_2 by concrete carbonation. *Environ. Sci. Technol.* **44,** 3181–3186 (2010).
20. Silva, A., Neves, R. & de Brito, J. Statistical modelling of carbonation in reinforced concrete. *Cement Concr. Compos.* **50,** 73–81 (2014).
21. Pommer, K., Pade, C., Institut, D. T. & Centre, N. I. *Guidelines: Uptake of Carbon Dioxide in the Life Cycle Inventory of Concrete* (Nordic Innovation Centre, 2006).
22. Kelly, T. D. *Crushed Cement Concrete Substitution for Construction Aggregates, a Materials Flow Analysis* (US Department of the Interior, US Geological Survey, 1998).
23. Lutz, H. & Bayer, R. *Ullmann's Encyclopedia of Industrial Chemistry* Vol. 11 (Wiley, 2010).
24. Bossink, B. & Brouwers, H. Construction waste: quantification and source evaluation. *J. Constr. Eng. Manage.* **122,** 55–60 (1996).
25. Khanna, O. S. *Characterization and Utilization of Cement Kiln Dusts (CKDs) as Partial Replacements of Portland Cement* (Univ. Toronto, 2009).
26. Pan, Y. *et al*. A large and persistent carbon sink in the world's forests. *Science* **333,** 988–993 (2011).
27. Piao, S. *et al*. The carbon balance of terrestrial ecosystems in China. *Nature* **458,** 1009–1013 (2009).
28. Berner, R. A., Lasaga, A. C. & Garrels, R. M. The carbon-silicate geochemical cycle and its effect on atmospheric carbon dioxide over the past 100 million years. *Am. J. Sci.* **283,** 641–683 (1983).
29. Rau, G. H., Knauss, K. G., Langer, W. H. & Caldeira, K. Reducing energy-related CO_2 emissions using accelerated weathering of limestone. *Energy* **32,** 1471–1477 (2007).
30. Smith, P. *et al*. Biophysical and economic limits to negative CO_2 emissions. *Nat. Clim. Change* **6,** 42–50 (2016).
31. Bobicki, E. R., Liu, Q., Xu, Z. & Zeng, H. Carbon capture and storage using alkaline industrial wastes. *Prog. Energy Combust. Sci.* **38,** 302–320 (2012).

Acknowledgements

Author contributions

F.X. and Z.L. designed the paper. F.X. conceived the research. F.X., C.P., T.S., J.W., K.H.Y., L.B., I.G., C.A. and P.C. provided the data from different countries and regions. T.S., F.X., J.W. and Y.Z. provided the survey statistics and experimental measurements data. S.J.D., F.X., Z.L., P.C., M.S. and D.C.B. performed the analysis. C.P., J.L., Z.L., L.J., P.C., K.H.Y., L.B., I.G. and Y.Z. provided the reference data. F.X., L.B., D.C.B., Z.L. M.S. and T.S. performed uncertainty analysis. S.J.D. and L.B. drew the figures. All authors contributed to writing the paper.
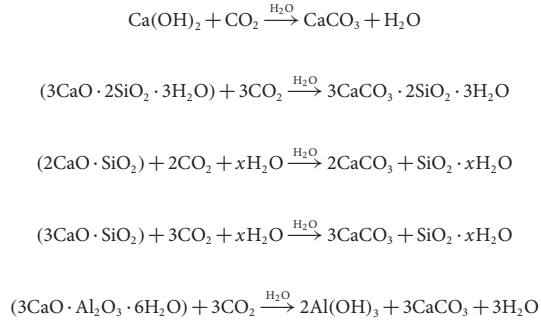
Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to Z.L.

Competing financial interests

The authors declare no competing financial interests.

Methods

Cement material carbonation. Civil engineers use the term 'carbonation' to describe a complicated physicochemical reaction between CO_2 and hydrated cement products in the presence of pore water, which ultimately sequesters carbon in cement material[15,32]. In solution of pore water, CO_2 reacts with $Ca(OH)_2$, and in turn reacts with calcium silicate, dicalcium silicate, tricalcium silicate, tricalcium aluminate and other hydrated products. The carbonation reactions start at the surface of the cement or concrete and moves inwards over time[10,33]. The main chemical reactions of carbonation are as follows:

$$Ca(OH)_2 + CO_2 \xrightarrow{H_2O} CaCO_3 + H_2O$$

$$(3CaO \cdot 2SiO_2 \cdot 3H_2O) + 3CO_2 \xrightarrow{H_2O} 3CaCO_3 \cdot 2SiO_2 \cdot 3H_2O$$

$$(2CaO \cdot SiO_2) + 2CO_2 + xH_2O \xrightarrow{H_2O} 2CaCO_3 + SiO_2 \cdot xH_2O$$

$$(3CaO \cdot SiO_2) + 3CO_2 + xH_2O \xrightarrow{H_2O} 3CaCO_3 + SiO_2 \cdot xH_2O$$

$$(3CaO \cdot Al_2O_3 \cdot 6H_2O) + 3CO_2 \xrightarrow{H_2O} 2Al(OH)_3 + 3CaCO_3 + 3H_2O$$

Process model of cement carbonation. A life cycle assessment (LCA) method is used to estimate carbon uptake by cement materials over time (see Supplementary Information). Total carbon uptake of cement (C_u) is calculated as

$$C_u = \sum Con + \sum Mor + \sum Waste + \sum CKD \qquad (1)$$

$\sum$ Con: carbon uptake by concrete cement.
$\sum$ Mor: carbon uptake by mortar cement.
$\sum$ Waste: carbon uptake by construction cement waste.
$\sum$ CKD: carbon uptake by cement kiln dust (CKD).

Carbon uptake by concrete cement. The concrete life cycle divided into three phases: service life (for example, in buildings), demolition and secondary use (including both disposal in a landfill and recycling)[5]. In this case, we calculate CO_2 uptake as

$$\sum Con = C_l^{t_l} + C_d^{t_d} + C_s^{t_s} \qquad (2)$$

$C_l^{t_l}$: carbon uptake during the service life.
$C_d^{t_d}$: carbon uptake during the demolition.
$C_s^{t_s}$: carbon uptake during the secondary use stage.

Service life. Concrete categories. We further break down cement utilization for different categories of concrete because the details of structure category are important for assessing strength class, cement content, exposure condition, exposed surface area, and service life[5,10,34–36].

Concrete strength classes. The strength classes of concretes are estimated based on the survey statistics and previous studies in the US[37,38], Europe and the rest of world[39] and Nordic countries[5].

Concrete cement content. The cement content for concrete (C_i) is the mass of cement used in one cubic metre of concrete (kg m^{-3})[34,37,39–43].

Exposure conditions, CO_2 concentrations, and additives. We estimate carbon uptake under five different categories of exposure conditions: exposed, sheltered, indoors, wet and buried[5]. Specifically, relative humidity, ambient CO_2 concentration[14,44] and additives have been shown to affect carbonation rate coefficients[9]. The range of applicable conditions are estimated based on the previously referenced, region-specific studies and survey statistics[5,9,10,14].

Coating and coverings. Application of surface coating and coverings such as paints can reduce the rates of cement carbonation by 10–30%[36,45]. Based on previous studies[46–50], we assess carbonation using carbonation correction coefficients meant to reflect the potential effects of coatings, including decreases in carbonation rates of up to 50% over the life cycle of concretes[51,52].

Concrete carbonation rates. Based on our estimates of concrete category, cement content, exposure conditions, additives and coatings, we use relevant concrete carbonation rate coefficients from various region-specific references[5,10,19]. We further calculated concrete carbonation rate coefficients by considering the impacts

of compressive strength class and exposure conditions (β_{csec})[12], cement additives (β_{ad})[36], CO_2 concentration (β_{CO_2})[9,14], and coating and cover (β_{CC})[47,53], according to ref. 5.

$$k_{li} = \beta_{csec} \times \beta_{ad} \times \beta_{CO_2} \times \beta_{cc} \qquad (3)$$

Service life duration. The concrete service life (t_l), the duration of the demolition stage (t_d), and the duration of the secondary use stage (t_s) are provided based on the previous, region-specific references[5,16–18,54].

Carbonation depth. The applicable carbonation rate coefficients and exposure times are used to calculate the carbonation depth (d_i) of concrete in each strength class and set of exposure conditions using Fick's diffusion law (equation (4))[5], where k_{li} is the carbonation rate coefficient of concrete in strength class i and t_l is the time of service life in years:

$$d_i = k_{li} \times \sqrt{t_l} \qquad (4)$$

Exposed surface area. The exposed surface area (A_i) of concrete in the US, China, Europe, and other countries based on average thickness of concrete structures are listed in the literature[5,10,21].

Volume of carbonated concrete in service life. The carbonated concrete volume V_i is calculated as

$$V_i = d_i \times A_i \qquad (5)$$

where A_i is exposed surface area and d_i represents the product of carbonation rate coefficient and carbonation depth for each concrete strength class i.

The carbonated cement in service life (W_{li}) can then be calculated as

$$W_{li} = \sum_{i=1}^{n} V_i \times C_i \qquad (6)$$

where C_i is the cement content of concrete in different strength classes (kg cement m^{-3})[34,37,39–43]. Next, we calculate the cumulative carbon uptake of carbonated concrete in service life ($C_l^{t_l}$)

$$C_l^{t_l} = W_l \times C_{clinker} \times f_{CaO} \times \gamma \times M_r \qquad (7)$$

where $C_{clinker}$ is clinker to cement ratio ranged from 75% to 97% according to IPCC guidelines of 1997 and 2006, f_{CaO} is the average CaO content of clinker in cement (65%, ranging from 60% to 67%)[55], γ is the proportion of CaO within fully carbonated cement that converts to $CaCO_3$ (0.80, ranging from 0.50 to 1.00; refs. 5,10–12,56,57), and M_r is the ratio of C element to CaO (a constant equal to the molar fraction in $(CO_2/CaO) \times (C/CO_2)$; 0.214)[5].

The variables $C_{clinker}$, f_{CaO} and M_r in the following equations (13), (21), (26), (29), (32) and (33) are the same as in (7).

Annual carbon uptake by concrete in service. Finally, we combine the results of the above calculations to calculate the annual carbon uptake in year t_l ($\Delta C_l^{t_l}$) as the cumulative carbon uptake in year t_l minus the cumulative carbon uptake in year $t_l - 1$:

$$\Delta C_l^{t_l} = \sum C_l^{t_l} - \sum C_l^{(t_l-1)} \qquad (8)$$

Demolition stage. During demolition, concrete structures are crushed into smaller pieces so that contained steel reinforcing can be recycled and the concrete can be more easily transported. The fate of demolition waste in different regions is taken from different sources in the literature. In China, sources suggest that more than 97% of concrete waste is landfilled, with less than 3% recycled[58]. In contrast, roughly 60% of concrete is recycled in the US, with the remaining 40% sent to landfills[17,22]. Recycling rates are even higher in Europe, with data showing that 61.1% recycled and only 38.9% was sent to landfill[5,21]. Other studies indicate that recycling rates in rest of world are quite low: about 25%[18,59].

Size and surface area of waste concrete pieces. The surface area of concrete pieces after demolition is difficult to estimate. We use available data to estimate a range and particle size distribution of different types of demolished concrete in each region[59,60].

Exposure time. We estimate the average exposure time (t_d) of concrete during the demolition stage is about 0.4 years in the whole world[5,11–13]. Almost all these demolished and crushed concrete pieces are exposed to open air; only very small proportions are stockpiled under shelter[12,59].

Carbonation of demolished concrete. We estimate the proportion (F_{di}) of concrete that will be carbonated during the demolition stage by assuming the shape of concrete particles and pieces is spherical[21]. The carbonation fraction is calculated according to particle size distributions and carbonation depths using Fick's diffusion law:

$$D_{0i} = 2d_{di} = 2k_{di} \times \sqrt{t_d} \qquad (9)$$

$$F_{di} = \begin{cases} 100\% - \int_a^b \frac{\pi}{6}(D - D_{0i})^3 / \int_a^b \frac{\pi}{6}D^3 \times 100\% & (a \geq D_{0i}) \\ 100\% - \int_{D_0}^b \frac{\pi}{6}(D - D_{0i})^3 / \int_a^b \frac{\pi}{6}D^3 \times 100\% & (a < D_{0i} < b) \\ 100\% & (b < D_{0i}) \end{cases} \qquad (10)$$

where F_{di} is the fraction of demolished concrete in strength class i that is carbonated, D_{0i} is the maximum diameter of particles that undergo full carbonation in strength class i, d_{di} the carbonation depth of particles in strength class i, k_{di} is the carbonation coefficient of concrete in strength class i in open air exposure conditions, t_d is the average time for the demolition stage, D is the diameter of demolished and crushed particles, a and b are the minimum and maximum diameter of crushed concrete particles in a given size distribution. All the particles less than D_{0i} will finish carbonation in t_d years or less, such that F_{di} will be 100%. For particle sizes larger than D_{0i}, F_{di} can be calculated by integration (equation (10)).

Using the fraction of concrete that will undergo carbonation for equation (10), we next calculate the mass of concrete cement carbonated during the demolition stage (W_d) as

$$W_{di} = (W_{ci} - W_{li}) \times F_{di} \qquad (11)$$

$$W_d = \sum_{i=1}^n W_{di} \qquad (12)$$

where W_{di} is the concrete cement carbonated during demolition for each concrete strength class i, W_{ci} is the cement consumed for each strength class i of concrete, W_{li} is the concrete cement carbonated during service life for each strength class i (W_{li} in equation (6)), F_{di} is the fraction of carbonated cement in concrete strength class i in the demolition stage, and W_d is the total mass of concrete cement carbonated in the demolition stage.

Total carbon uptake during demolition stage. Finally, we estimate total carbon uptake during the demolition stage ($C_d^{t_d}$) based on cement carbonated in demolition stage and carbonation fraction of differently treated concretes:

$$C_d^{t_d} = W_d \times C_{clinker} \times f_{CaO} \times \gamma \times M_r \qquad (13)$$

$C_d^{t_d}$ is carbon uptake of concrete cement during demolition and γ is the proportion of CaO within fully carbonated concrete that converts to $CaCO_3$. The other parameters are the same as in equation (7).

Secondary use stage. After demolition, concrete materials continue to absorb carbon dioxide during the secondary use stage. In sum, more than 91% of crushed concrete particles worldwide are buried, either in landfills or as part of their recycled use, such as for road base or backfill aggregates[13,18,22,58,59].

Carbonation depth in secondary use stages. The carbonation rate coefficients of waste concrete in the secondary use stage will be slow and decreasing due to the layer of carbonated cement (d_{di}) that was during the demolition stage[45] and the fact that most of the concrete is buried and not exposed to the air[5]. The total carbonation depth in the demolition stage and secondary use stage (d_{ti}) can be estimated by carbonated depth in the demolition stage (d_{di}) plus the new carbonation depth (d_{si}) during the secondary use stage. There is a time lag Δt_i for the same carbonation depth (d_{di}) from air exposure condition to buried condition as follows:

$$d_{di} = K_{di} \times \sqrt{t_{di}} = K_{si} \times \sqrt{t_{si}} \qquad (14)$$

$$k_{di} \times \sqrt{t_{di}} = k_{si} \times \sqrt{t_{si} + \Delta t_i} \qquad (15)$$

$$\Delta t_i = t_{di} \times \left(\frac{k_{di}^2}{k_{si}^2} - 1 \right) \qquad (16)$$

The total carbonation depth in the demolition and secondary use stages d_{ti} can then be calculated by

$$d_{ti} = d_{di} + d_{si} = k_{si} \times \sqrt{t_{si} + t_{di} + \Delta t_i} \qquad (17)$$

d_{di}: carbonation depth at the end of the demolition stage.
k_{di}: carbonation rate coefficient in the demolition stage (exposed to air).
t_{di}: carbonation time for existing carbonated depth d_{di} during the demolition stage.
k_{si}: carbonation rate coefficient of concrete particle in strength class i in the secondary use stage (buried condition).
t_{si}: carbonation time for d_{di} if waste concrete in the secondary use stage (buried condition).
Δt_i: time lag for the same carbonation depth (d_{di}) from buried condition to air exposure condition.
d_{ti}: total carbonation depth in the demolition stage and secondary use stage.

Fraction carbonized. The carbonation fraction of cement in concrete rubble (F_{si}) during the secondary use stage is calculated as:

$$D_{1i} = 2d_{ti} = 2K_{si} \times \sqrt{t_{si} + t_{di} + \Delta t_i} \qquad (18)$$

$$F_{si} = \begin{cases} 100\% - \int_a^b \frac{\pi}{6}(D - D_{ti})^3 / \int_a^b \frac{\pi}{6}D^3 \times 100\% - F_{di} & (a \geq D_{1i}) \\ 100\% - \int_{D1}^b \frac{\pi}{6}(D - D_{ti})^3 / \int_a^b \frac{\pi}{6}D^3 \times 100\% - F_{di} & (a < D_{1i} < b) \\ 100\% - F_{di} & (b < D_{1i}) \end{cases} \qquad (19)$$

where D_{1i} is the maximum diameter of particles that undergo full carbonation in strength class i in the demolition and secondary use stages, D is the diameter of demolished and crushed particles, F_{di} is the fraction of carbonated waste concrete particle in strength class i in the demolition stage, a and b are the minimum and maximum diameter of crushed concrete particles in a given size distribution. All the particles less than D_{1i} will finish carbonation in $t_{si} + t_{di} + \Delta t_i$ years, so there F_{si} is 100% - F_{di}. The value of F_{si} for particle size larger than D_{1i} can be calculated by integration.

Cumulative and annual carbon uptake during the secondary use stage. The cumulative and annual carbon uptake in the secondary use stage can be calculated by the following:

$$W_{si} = W_{ci} - W_{li} - W_{di} \qquad (20)$$

$$C_s^{t_s} = \left(\sum_{i=1}^n W_{si} \times F_{si} \right) \times C_{clinker} \times f_{CaO} \times \gamma \times M_r \qquad (21)$$

$$\Delta C_s^{t_s} = C_s^{t_s} - C_s^{(t_s-1)} \qquad (22)$$

where W_{si} is the weight of cement used for strength class i in the secondary use stage, W_{ci} the weight of cement used for strength class i in the building construction stage, W_{li} is the carbonated concrete cement in strength class i in the service stage, W_{di} is carbonated concrete cement for concrete strength class i in the demolition stage, F_{si} is the fraction of carbonated strength class i concrete cement in the treatment and secondary use stage, γ is proportion of CaO within fully carbonated cement that converts CaO to $CaCO_3$, $C_s^{t_s}$ is the cumulative carbon uptake in year t_s, and $C_s^{(t_s-1)}$ is the cumulative total carbon uptake in year $t_s - 1$, $\Delta C_s^{t_s}$ is annual carbon uptake in year t_s in the secondary use stage. The other parameters are the same as in equation (7).

Carbon uptake by mortar cement. **Cement utilization for mortars.** Cement mortar is used for rendering and plastering (that is, decorating), masonry (brick-laying), maintenance and repairing of concrete structures, and various other applications[23,61,62]. Most mortar is used for rendering, plastering and decorating[61].

The typical thickness of cement mortar utilization. Rendering and plastering mortar is usually applied in a thickness of 10–30 mm and decorating (finishing) mortar is typically much thinner, only 1–5 mm[23,61]. When used as tile adhesive or grout, mortar is typically applied in thicknesses of 15–30 mm and 3–30 mm, respectively[23]. For self-levelling under layers, thicknesses vary from 5 to 30 mm, and the thickness of mortar for screeds is 30–80 mm[61,62]. Most of these cement mortar thicknesses are about 20 mm[23,33,45]. The thickness of mortar for masonry is about 10 mm, except for a small proportion in 2–3 mm for very even blocks[23]. Mortar used for maintaining and repairing (that is, patching concrete structures and building surfaces) is applied similarly to rendering and adhesive uses, with a mean thickness of 25 mm.

Carbonation rate coefficients of cement mortar. Cement mortars have been shown to undergo carbonation at a faster rate than concrete[63,64]. The carbonation rate coefficients of cement mortar are between 6.1 mm yr$^{-1/2}$ and 36.8 mm yr$^{-1/2}$ in outdoor and indoor exposure conditions, respectively (in temperate climate conditions and according to our field survey and experiment data using the 1%

alcohol phenolphthalein solution). Carbonation depth will increase if the cement contains more additives[48]. In this study, we use an average carbonation rate for mortar of 19.6 mm yr$^{-1/2}$, but evaluate uptake assuming the full range 6.1 mm yr$^{-1/2}$ and 36.8 mm yr$^{-1/2}$.

Carbon uptake by mortar cements. We calculate annual carbon uptake based on the proportion of annual carbonation depth[65], and estimate carbon uptake as the sum of uptake by rendering and plastering mortar (C_{rpt}), uptake of masonry mortar (C_{rmt}), and uptake of maintaining and repairing mortar (C_{rmat}):

$$\sum Mor = C_{rpt} + C_{rmt} + C_{rmat} \qquad (23)$$

Annual carbonation of cement in mortar used for rendering, plastering, and decorating is calculated by

$$d_{rp} = K_m \times \sqrt{t} \qquad (24)$$

$$f_{rpt} = (d_{rpt} - d_{rp(t-1)})/d_{Trp} \times 100\% \qquad (25)$$

$$C_{rpt} = W_m \times r_{rp} \times f_{rpt} \times C_{clinker} \times f_{CaO} \times \gamma_1 \times M_r \qquad (26)$$

where d_{rp} is the carbonation depth of rendering mortar, K_m is the carbonation rate coefficient of cement mortar, d_{rpt} and $d_{rp(t-1)}$ are the carbonation depths in years t and $(t-1)$, respectively, d_{Trp} is the utilization thickness of rendering mortar, f_{rpt} is the annual carbonation percentage of cement mortar for rendering, C_{rpt} is the annual carbon uptake of carbonated mortar cement for rendering. γ_1 is the proportion of CaO within fully carbonated mortar cement that converts to CaCO$_3$, W_m is the cement for mortar and r_{rp} is the percentage of cement for rendering in mortar cement.

Carbon uptake of repairing and maintaining cement mortar. Annual carbon uptake of cement mortar for repairing and maintaining is calculated by

$$d_{rm} = K_m \times \sqrt{t} \qquad (27)$$

$$f_{rmt} = (d_{rmt} - d_{rm(t-1)})/d_{Trm} \times 100\% \qquad (28)$$

$$C_{rmt} = W_m \times r_{rr} \times f_{rmt} \times C_{clinker} \times f_{CaO} \times \gamma_1 \times M_r \qquad (29)$$

where d_{rm} is the carbonation depth of repairing and maintaining mortar, d_{rmt} and $d_{rm(t-1)}$ are the carbonation depths in years t and $(t-1)$, respectively. d_{Trm} is the utilization thickness of repairing and maintaining mortar. f_{rmt} is the annual carbonation percentage of cement for repairing and maintaining mortar. C_{rmt} is the annual carbon uptake of carbonated mortar cement for repairing and maintaining. r_{rr} is the percentage of cement for repairing and maintaining in mortar cement. γ_1 is the proportion of CaO within fully carbonated mortar cement that converts to CaCO$_3$.

The carbon uptake by masonry cement mortar can is calculated as

$$C_{rmat} = C_{mbt} + C_{mot} + C_{mnt} \qquad (30)$$

where C_{mbt} is carbon uptake by masonry mortar of walls with both sides rendered, C_{mot} is carbon uptake by masonry mortar of walls with one side rendered, and C_{mnt} is carbon uptake by masonry mortar of walls with no rendering. The carbon uptake calculation method of C_{mbt}, C_{mot}, and C_{mnt} is similar as that of rendering and plastering mortar by considering wall thickness and demolition effects.

Carbon uptake by cement in construction wastes. We estimate carbon uptake of construction waste[24,54] by

$$\sum_{waste} = C_{wastecon} + C_{wastemor} \qquad (31)$$

$$C_{wastecon} = \left(\sum_{i=1}^{n} W_{ci} \times f_{con} \times r_{cont} \right) \times C_{clinker} \times f_{CaO} \times \gamma \times M_r \qquad (32)$$

$$C_{wastemor} = \left(\sum_{i=1}^{n} W_{mi} \times f_{mor} \times r_{mor} \right) \times C_{clinker} \times f_{CaO} \times \gamma_1 \times M_r \qquad (33)$$

where $C_{wastecon}$ and $C_{wastemor}$ are carbon uptake by construction waste concrete and construction waste mortar, respectively. W_{ci} is cement used for concrete in strength class i, f_{con} is loss rate of cement for concrete in the construction stage[24,40], r_{cont} is annual carbonation fraction of construction waste concrete, γ is proportion of CaO within fully carbonated concrete that converts CaO to CaCO$_3$, W_{mi} is cement used for mortar in strength class i, f_{mor} is loss rate of cement for mortar[54,66], r_{mor} is annual

carbonation fraction of construction waste mortar. γ_1 is the proportion of CaO within fully carbonated mortar cement that converts to CaCO$_3$.

Carbon uptake by cement kiln dust. We estimate carbon uptake by CKD in different regions[67–69] of the world based on the cement production, CKD generation rate, and proportion of CKD treatment in landfill (Supplementary Data 4) as follows:

$$\sum_{CKD} = \left(\sum_{i=1}^{n} W_i \times C_{clinker} \times r_{CKD} \times r_{landfill} \right) \times f_{2CaO} \times \gamma_2 \times M_r \qquad (34)$$

where W_i is the cement production in region i, r_{CKD} is the CKD generation rate based on clinker[68], $r_{landfill}$ is proportion of CKD treatment in landfill, f_{2CaO} is CaO proportion in CKD[70], and γ_2 is the fraction of CaO within fully carbonated CKD that has been converted to CaCO$_3$.

Uncertainty analysis. We use a Monte Carlo method as recommended by the 2006 IPCC guidelines for National Greenhouse Gas Inventories to evaluate uncertainty of CO$_2$ removal due to cement material carbonation[71]. We identify 26 causes of uncertainties associated with carbon sequestration estimates, which we vary across wide ranges to estimate the implications for carbon uptake (see Supplementary Information). The mean value carbon uptake from global cement materials is 0.25 Gt C (2σ standard deviation of 10.03%) in 2013.

Data availability. The authors declare that the data supporting the findings of this study are available within the Article and its Supplementary Information files.

References

32. Papadakis, V. G., Vayenas, C. G. & Fardis, M. A reaction engineering approach to the problem of concrete carbonation. *AIChE J.* **35,** 1639–1650 (1989).
33. Huang, N., Chang, J. & Liang, M. Effect of plastering on the carbonation of a 35-year-old reinforced concrete building. *Const. Build. Mater.* **29,** 206–214 (2012).
34. Jonsson, G. & Wallevik, O. Information on the use of concrete in Denmark, Sweden, Norway and Iceland. *Icelandic Build. Res. Inst.* (2005).
35. Kelly, T. D. & Matos, G. R. *Historical Statistics for Mineral and Material Commodities in the United States* (United States Geological Survey Data, 2014); http://minerals.usgs.gov/minerals/pubs/historical-statistics/nickel-use.pdf
36. Lagerblad, B. *Carbon Dioxide Uptake During Concrete Life Cycle: State of the Art* (Swedish Cement and Concrete Research Institute, 2005).
37. Low, M.-S. *Material Flow Analysis of Concrete in the United States* (Massachusetts Institute of Technology, 2005).
38. Nisbet, M. A. *Environmental Life Cycle Inventory of Portland Cement Concrete* (Portland Cement Association Skokie, 2000).
39. *Ready-Mixed Concrete Industry Statistics 2001–2013* (European Ready Mixed Concrete Organization, 2014); http://www.ermco.eu
40. Zhou, H. *Construction and Installation Engineering Budget Manual* (China Machine Press, 2003).
41. Huang, Z. & Zhao, J. *Concrete Mix Proportion Quick Manual* (China Building Industry, 2001).
42. Pommer, K. & Pade, C. *Guidelines: Uptake of Carbon Dioxide in the Life Cycle Inventory of Concrete* (Nordic Innovation Centre, 2006).
43. *Concrete-Part 1. Specification, Performance, Production and Conformity* EN 206-1-2000 (European Committee for Standardization, 2001).
44. Talukdar, S., Banthia, N., Grace, J. & Cohen, S. Carbonation in concrete infrastructure in the context of global climate change: part 2–Canadian urban simulations. *Cement Concr. Compos.* **34,** 931–935 (2012).
45. Roy, S., Northwood, D. & Poh, K. Effect of plastering on the carbonation of a 19-year-old reinforced concrete building. *Constr. Build. Mater.* **10,** 267–272 (1996).
46. Klopfer, H. The carbonation of external concrete and how to combat it. *Bautenschutz Bausanieruniz* **3,** 86–97 (1978).
47. Browner, R. Design prediction of the life for reinforced concrete in marine and other chloride environments. *Durability Build. Mater.* **1,** 113–125 (1982).
48. Zhang, L. Carbonization delay coefficients research of concrete surface coverages [in Chinese]. *J. Xi'an Inst. Metall. Const. Eng.* **21,** 34–40 (1989).
49. Seneviratne, A. M. G., Sergi, G. & Page, C. L. Performance characteristics of surface coatings applied to concrete for control of reinforcement corrosion. *Constr. Build. Mater.* **14,** 55–59 (2000).
50. Zafeiropoulou, T., Rakanta, E. & Batis, G. Performance evaluation of organic coatings against corrosion in reinforced cement mortars. *Prog. Org. Coat.* **72,** 175–180 (2011).
51. Lo, T. Y., Liao, W., Wong, C. K. & Tang, W. Evaluation of carbonation resistance of paint coated concrete for buildings. *Constr. Build. Mater.* **107,** 299–306 (2016).

52. Moon, H. Y., Shin, D. G. & Choi, D. S. Evaluation of the durability of mortar and concrete applied with inorganic coating material and surface treatment system. *Constr. Build. Mater.* **21,** 362–369 (2007).
53. Papadakis, V. G. Effect of supplementary cementing materials on concrete resistance against carbonation and chloride ingress. *Cement Concr. Res.* **30,** 291–299 (2000).
54. Huang, T., Shi, F., Tanikawa, H., Fei, J. & Han, J. Materials demand and environmental impact of buildings construction and demolition in China based on dynamic material flow analysis. *Resour. Conserv. Recy.* **72,** 91–101 (2013).
55. Eggleston, S., Buendia, L. & Miwa, K. *2006 IPCC Guidelines for National Greenhouse Gas Inventories: Volume 3 Industrial Processes and Product Use* (Institute for Global Environmental Strategies, 2006).
56. Takano, H. & Matsunaga, T. CO_2 fixation by artificial weathering of waste concrete and coccolithophorid algae cultures. *Energy Convers. Manage.* **36,** 697–700 (1995).
57. Chang, C.-F. & Chen, J.-W. The experimental investigation of concrete carbonation depth. *Cement Concr. Res.* **36,** 1760–1767 (2006).
58. Lu, W. *Waste Recycling System Material Metabolism Analysis Model and Its Application* PhD thesis, Univ. Tsinghua (2010).
59. Kikuchi, T. & Kuroda, Y. Carbon dioxide uptake in demolished and crushed concrete. *J. Adv. Concr. Technol.* **9,** 115–124 (2011).
60. *Technical Specification for Application of Recycled Aggregate* JGJ/T 240-2011 (The Department of Housing and Urban–Rural Development, 2011).
61. Winter, C. & Plank, J. The European dry-mix mortar industry (Part 1). *ZKG Int.* **60,** 62–69 (2007).
62. *Beijing Local Standard - Technical Specifications for Application of Dry-mixed Mortar* DB11/T696-2009, J11582-12010 (Housing and Urban Rural Construction Standard Quota Division, 2010).
63. Sprung, S. & Kropp, J. *Ullmann's Encyclopedia of Industrial Chemistry* Vol. 9 (Wiley, 2010).
64. El-Turki, A., Carter, M. A., Wilson, M. A., Ball, R. J. & Allen, G. C. A microbalance study of the effects of hydraulicity and sand grain size on carbonation of lime and cement. *Const. Build. Mater.* **23,** 1423–1428 (2009).
65. Jo, Y. K. Basic properties of epoxy cement mortars without hardener after outdoor exposure. *Constr. Build. Mater.* **22,** 911–920 (2008).
66. Lu, W. *et al*. An empirical investigation of construction and demolition waste generation rates in Shenzhen city, South China. *Waste Manage.* **31,** 680–687 (2011).
67. Khanna, O. S. *Characterization and Utilization of Cement Kiln Dusts (CKDs) as Partial Replacements of Portland Cement* PhD thesis, Univ. Toronto (2009).
68. *Report to Congress on Cement Kiln Dust* (Office of Solid Waste and Emergency Response, 1993).
69. Hawkins, G. J., Bhatty, J. I. & O'Hare, A. T. in *Innovations in Portland Cement Manufacturing* (eds Bhatty, J. I., Miller, F. M. & Kosmatka, S. H.) 735–779 (Portland Cement Association, 2004).
70. Sreekrishnavilasam, A., King, S. & Santagata, M. Characterization of fresh and landfilled cement kiln dust for reuse in construction applications. *Eng. Geol.* **85,** 165–173 (2006).
71. Eggleston, S., Buendia, L., Miwa, K., Ngara, T. & Tanabe, K. *IPCC Guidelines for National Greenhouse Gas Inventories* (Institute for Global Environmental Strategies, 2006).