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Finding least-cost net-zero CO_{2e} strategies for the European cement industry using geospatial techno-economic modelling†

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Cement production is responsible for approximately 7% of anthropogenic CO_2-equivalent (CO_{2e}) emissions, while characterised by low margins and the highest carbon intensity of any industry per unit of revenue. Hence, economically viable decarbonisation strategies must be found. The costs of many emission reduction strategies depend on geographical factors, such as plant location and proximity to feedstock or on synergies with other cement producers. The current literature lacks quantification of least-cost decarbonisation strategies of a country or region's total cement sector, while taking stock of these geospatial differences. Here, we quantify which intervention ensembles could lead to least-cost, full decarbonisation of the European cement industry, for multiple European regions. We show that least-cost strategies include the use of calcined clay cements coupled with carbon capture and storage (CCS) from existing cement plants and direct air capture with carbon storage (DACCS) in locations close to CO_2 storage sites. We find that these strategies could cost €72–€75 per tonne of cement (t_{cement}^{-1}, up from €46–€51.5 t_{cement}^{-1}), which could be offset by future costs of cement production otherwise amounting to €105–€130 t_{cement}^{-1} taking the cost of CO_{2e} emission certificates into account. The analysis shows that for economically viable decarbonisation, collaborative and region-catered approaches become imperative, while supplementary cementitious materials including calcined clays have a key role.

Sustainability spotlight

Cement production, a major source of industrial emissions, urgently needs cost-effective emission reduction strategies. Decarbonising cement production is challenging, because it not only requires high temperatures but most of its CO_2 emissions stem from releasing CO_2 from limestone directly. While multiple different interventions for emission reduction have been suggested, many of them are going to work better in some locations than others. In this article we tackle the question, which mix of decarbonisation strategies are likely to be cheapest, taking multiple European regions as case studies and investigate how collaboration among cement producers can influence their costs to reach net-zero CO_{2e} emission by 2050. This issue aligns with SDG 9 (Industry, Innovation, and Infrastructure) and SDG 13 (Climate action).

Introduction

CO_2-equivalent (CO_{2e}) emissions must reach net-zero by 2050 (also referred to as deep/full decarbonisation) to limit global warming to 1.5 °C.[1] The cement industry alone is responsible for 7% of anthropogenic CO_{2e} emissions.[2,3] As the use of cement is essential for economic growth with a projected global market size of reaching 6.08 billion tonnes per year (a^{-1}) in 2026,[4] reducing cement's carbon intensity is vital. While alternative building materials (*e.g.*, wood) could be used in some cases, it seems unlikely that cement can be replaced entirely, and therefore zero-carbon production practices need to be developed and implemented.[5] Reducing CO_{2e} emissions in the cement industry is particularly challenging: cementitious products are characterised by high emissions and low margins,[6] leaving limited room for investments in decarbonisation methods, while the high process-inherent emissions reduce the technological options to produce "net-zero-CO_{2e}" cement.

Cement production involves limestone mining followed by calcination to produce clinker, before it is ground and blended with other materials (*e.g.*, gypsum) to reach specified cement

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properties. The calcination step is the main CO_2 contributor, stemming from two sources. It requires high temperatures (1400–1500 °C[5]), resulting in high energy-related emissions (from using fossil fuels such as coal), and causes CO_2 to be released from limestone itself as process-inherent emissions (eqn (1)), accounting for approximately 60% of cement's total CO_{2e} emissions.[7]

$$CaCO_3 \xrightarrow{1400-1500\ ^{\circ}C} CaO + CO_2\uparrow \quad (1)$$

Hence, to significantly reduce CO_{2e} emissions either the use of clinker must be reduced, or the produced CO_2 needs to be abated, *e.g.*, *via* CO_2 capture and storage (CCS). A review of the academic literature and policy reports suggests a multitude of potential decarbonisation strategies (Fig. 1).[6,8,10,16–18] As there are two major emissions sources (energy-related and process-inherent), it may be necessary to consider several complementary approaches for addressing each source.

Decreasing energy-related emissions can be accomplished by implementing energy efficiency measures and by fuel substitution. While substantial gains in efficiency have been made using these methods in recent years,[8] to further decrease energy-related emissions alternative fuels[6,8–11] or the adoption of process electrification[13] have been suggested.

To reduce process-inherent emissions, the cement industry has used clinker substitutes known as supplementary cementitious materials (SCMs). These SCMs can be either industrial by-products (such as steel slag and fly ash) or natural minerals (like limestone and natural pozzolans).[8,11,12,19,20] However, their use as a emission reduction measure is limited as they only substitute clinker partially and the availability of some industrial by-products is expected to decline in the future due to the implementation of more environmentally sustainable production processes in other industries.[21] Moreover, the use of SCMs has limitations related to the overall strength and workability of the cement (including increased water requirements and altered curing time).[12] Therefore, these measures alone will not suffice to mitigate all process-inherent emissions. Hence, the complementary use of emerging technologies such as alternative clinkers and novel SCMs (*e.g.*, calcined clay cements), carbon capture, utilisation (CCU, *e.g.*, CO_2 mineralisation), and storage (CCS)[10] as well as carbon dioxide removal (CDR, *e.g.*, direct air capture and storage (DACCS)), is essential.

A critical remaining question is how countries' cement sectors and individual cement plants can reach net-zero-CO_{2e}-emissions in a cost-effective manner? Cement plants are widely distributed across Europe (282 integrated cement plants located in Western Europe alone,[22] Fig. 2B), due to the low economic viability of transporting cement over long distances (90% of all cement is transported less than 281 km[29]). This also means that the economic viability of the aforementioned decarbonisation strategies depends heavily on plant location. For example, calcined clay cements require suitable kaolinite clays,[30–32] CO_2 mineralisation requires earth alkaline metal oxide containing minerals (such as olivine-bearing rocks which contain forsterite (Mg_2SiO_4 [23,33])) and carbon capture and storage requires suitable underground CO_2 storage sites (*e.g.*, depleted oil and gas fields[34]), each of which are found in different locations in Europe (Fig. 2A). Therefore, policy and industry decision-makers face the challenge of selecting suitable decarbonisation strategies for a wide range of cement plant locations to decarbonise the sector as a whole.

These strategies need to be based, among others, on comparative ex ante techno-economic assessments to evaluate the economic performance of each potential ensemble of decarbonisation interventions evaluated in a specific locational setting. However, techno-economic assessments that compare a suite of emission reduction technologies for the cement industry are sparse, while some generic analyses of individual technologies exist (*e.g.*, carbon capture,[35] calcined clay cements,[31] CO_2 mineralisation[36]). With CO_{2e} neutrality goals only one or two investment cycles away, policy and industry

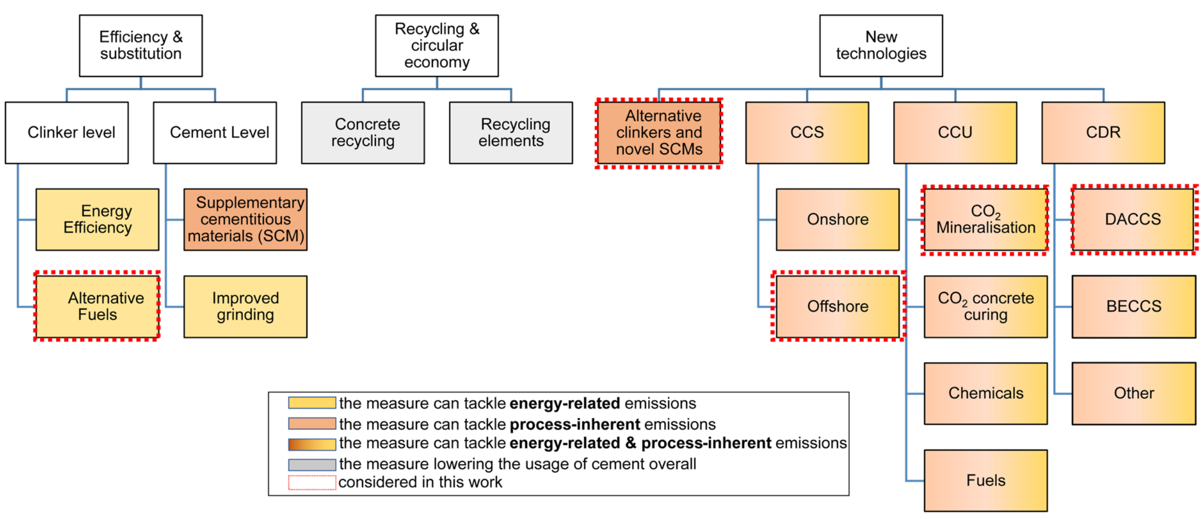

Fig. 1 Map of suggested interventions for emission reduction in cement production. Compiled from academic literature and policy advise reports[5,6,8–15] (Table S2†). Red-dotted interventions are investigated here. Classification of interventions based on Favier, *et al.*[8]

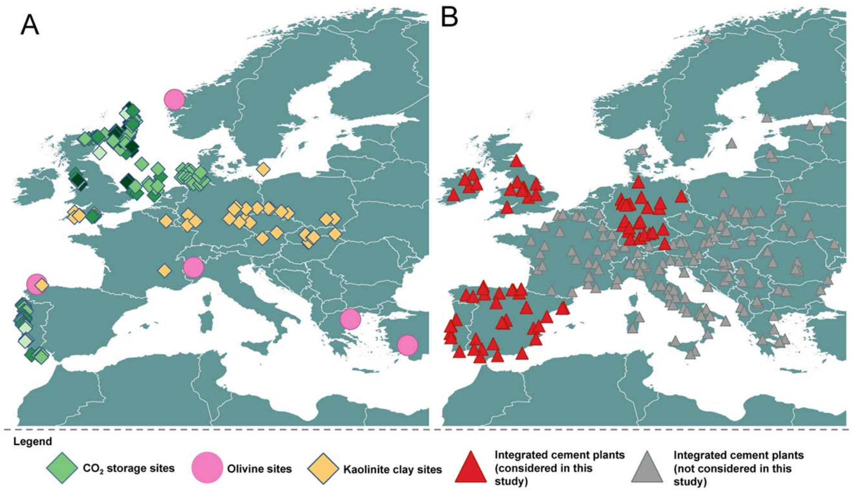

Fig. 2 Available locations for production of olivine-bearing rocks[23] and kaolinite clays[24–26] and for offshore CO_2 storage[27,28] of relevance for the here analysed regions (*i.e.*, Ireland, United Kingdom, Germany, Portugal and Spain) (A). Integrated cement plant locations in Europe[22] (B). Regions analysed in this study are marked in red.

decision-makers are in urgent need of detailed location-specific assessments, to help determine their technology and investment strategies.

To address this gap, we here present a techno-economic geospatial analysis for three selected case study regions (*i.e.*, United Kingdom and Ireland (UK&IRE), Germany (GER) as well as Portugal and Spain (PT&ES)) to cover the heterogeneity of regions across Europe. By providing a geospatial assessment of these strategies across various cement plants in Europe, we offer a novel framework for optimising decarbonisation efforts in the cement industry tailored to local differences, resources, and constraints, filling a current gap in the literature. We developed a mixed-integer linear programming model to determine least-cost emission reduction strategies for the entire industry in one region considering several selected interventions which either aim to reduce the clinker content, store CO_2 in cementitious products or capture CO_2 from the atmosphere. The goal of this study is to shed light on emission reduction strategies in the cement industry while considering synergies between cement plants as well as avoiding path dependencies (*i.e.*, earlier investment decisions affect, hinder future investments[37]). The key contributions of this study include:

• For the first time, least-cost, full cement decarbonisation strategies for selected European regions are presented, accounting for geospatial differences of individual cement plants.

• The value of collaboration between cement companies is quantified, showing the need for a concerted approach *versus* a single mover strategy.

• The value and effects of including selected abatement strategies is quantified, *e.g.*, of using CCS or calcined clays.

• Optimal investment sequences for the cement sector are modelled to avoid technology lock-ins that may lead to higher final costs.

• We show that collaborative, full, decarbonisation of cement sectors can be cost effective *viz-a-viz* expected CO_2 emissions taxes.

Methodology

To investigate net-zero CO_{2e} emission strategies for the cement industry, considering transport costs for feedstocks and synergies between decisions of multiple cement plants, we first conducted a literature review (Methodology – Literature review) and selected suggested interventions (Methodology – Selected Interventions), before estimating their costs at maturity (Methodology – Iterative learning for CO_2 emission reduction strategies). Second, we calculated the transport costs for feedstocks for different cement plant locations (Methodology – Transport of bulk materials) and fed both results (*i.e.*, cost estimates for interventions and transport costs) into a mixed-integer programming model (Methodology – Model structure). To investigate least-cost emission reduction strategies while avoiding path dependencies which would only allow certain interventions to be selected at a specific time, a solver first finds the least-cost strategies for reaching net-zero CO_{2e} emissions for an entire region (to be reached in the EU in 2050[38]) by implementing interventions at cement plants (*e.g.*, installing CO_2 capture) and/or constructing infrastructure (*e.g.*, CO_2 pipelines, CO_2 injection wells). After finding a least-cost solution for reaching net-zero, strategies for intermediary targets (*e.g.*, 50% emission reduction) were selected by the solver *via* back-casting (Methodology – Optimisation Framework). Because the solver finds a least-cost strategy for the entire region, in a last step we allocated costs for shared infrastructure to individual cement plants (Methodology – Post-processing).

Literature review

There are a multitude of strategies suggested for emissions reduction in the cement industry (Fig. 1). A holistic review of technology options and their emission reduction potentials was performed by Favier, *et al.*[8] Their review concluded that decarbonisation should be tackled in all stages of the value chain (from clinker, cement to concrete use). To reach deep decarbonisation multiple interventions might have to be implemented simultaneously, among them increasing energy efficiency and introducing alternative fuels, reducing clinker content in cement blends or the use of alternative clinkers as well as carbon capture, utilisation, and storage (CCUS). These suggestions were mostly in line with the roadmap by the European Cement Association,[10] which estimates the biggest share of emission reductions could be implement through CCUS, alternative fuels and *via* clinker substitutions. To investigate the costs of carbon capture at cement plants, Voldsund, *et al.*[35] provided a detailed techno-economic analysis showing that carbon capture not only can be costly (€42.4 € $t_{CO_2,avoided}^{-1}$ to €83.5 $t_{CO_2,avoided}^{-1}$), but also cannot be used to capture all emissions from a cement plant (*e.g.*, due to energy penalties). Hence, residual downstream emission will have to be captured elsewhere using carbon dioxide removal techniques, like direct air capture and storage. To this end, Young, *et al.*[39] provided a detailed assessment of different direct air capture technologies as well as their future costs. As clinker substitutes or alternative clinkers Favier, *et al.*[8] reviewed multiple different options, many of which are in early stages of development or for niche markets (*e.g.*, alkali activated systems,[8] magnesia cements,[8] CO_2 concrete curing in precast concrete production[20]) while the use of calcined clays was suggested of having the highest potential, which was confirmed by other researchers.[31,32,40] In Strunge *et al.*[36] among other researchers,[41,42] we previously showed that CO_2 mineralisation to produce SCMs, which both stores CO_2 permanently and replaces clinker can have similarly a large emission reduction potential. To investigate alternative fuels in the cement industry, Kusuma, *et al.*[43] reviewed multiple bio-based feedstocks such as wood pallets and suggested that a combination of biofuels with CCS might lead to high emission reductions through synergetic effects.

Selected interventions

To select potential interventions for this study, we first reviewed potential emission reduction strategies for the cement industry (Fig. 1) and selected strategies (interventions) with high emission reduction potential. We chose alternative fuels (biomass), calcined clay cements, CO_2 mineralisation, carbon capture and storage (CCS), and direct air capture and storage (DACCS). These interventions span the domain of decarbonisation options presented in Fig. 1, aside from recycling options.

The cement industry currently predominantly uses waste and industrial by-products[44] as alternative fuels, which show limited effectiveness due to their fossil carbon content (*i.e.*, fossil carbon is emitted during combustion of waste tyres, a common fuel replacement used).[10] Therefore, we only considered biofuels as wood pellets from North America as means to decrease energy-related emissions as the availability of biomass from Europe is limited (Note S1 and Table S1†).

We considered calcined clay cements, particularly using kaolinite clays ($Al_4[(OH)_8|Si_4O_{10}]$), as a means of reducing emissions by blending clinker (50%) calcined clay (30%), limestone (15%) and gypsum (5%), forming LC3 (Limestone Calcined Clay Cement).[31] This blend can significantly reduce emissions due to clinker replacement and lower calcination temperatures of clays, thus lower energy emissions per unit of cement produced[31,40,45] (Note S4†).

We considered CO_2 mineralisation as means of long-term CO_2 storage in cementitious products, where captured CO_2 reacts with minerals to create stable carbonates, producing supplementary cementitious materials as clinker replacements (we considered a substitution level of 30% clinker). As feedstock we here considered olivine-bearing rocks (*i.e.*, peridotite which contain forsterite (Mg_2SiO_4)), due to their availability and low costs[36] (Note S3†).

Due to its high emission reduction potential, we included CCS (*i.e.*, capturing CO_2 directly from cement plant flue gas and transporting it to geological storage sites). We only considered offshore CO_2 storage due to lower risk perceptions of lay people compared to onshore storage[46] (Note S2†).

To offset all residual emissions, thus allowing fully net-zero-CO_{2e} supply chains, we included DACCS for carbon dioxide removal from the atmosphere[47,48] (Note S5†).

As some combinations of interventions are possible (*e.g.*, CCS and CO_2 mineralisation), while others cannot be combined (*e.g.*, CO_2 mineralisation and calcined clay cements), we defined a set of interventions which can be considered by the solver. We here considered the following emission reduction interventions in the model:

(1) Conventional cement production + CCS.
(2) Conventional cement production + biofuels.
(3). Conventional cement production + biofuels + CCS.
(4) Conventional cement production + CO_2 capture and mineralisation.
(5) Reduced conventional cement production + CO_2 capture and mineralisation + biofuels.
(6) Reduced conventional cement production + CO_2 capture and mineralisation + biofuels + CCS.
(7) Reduced conventional cement production + CO_2 capture and mineralisation + CCS.
(8) Reduced conventional cement production + calcined clay.
(9) Reduced conventional cement production + calcined clay + biofuels.
(10) Reduced conventional cement production + calcined clay + CCS.
(11) Reduced conventional cement production + calcined clay + biofuels + CCS.
(12) Direct air capture and storage.

Iterative learning for CO_2 emission reduction strategies

Most of the here considered interventions/technologies are not yet mature. As technologies can be expected to decrease in cost through iterative learning, from the first plant (first-of-a-kind) to when *n* plants have been built (*N*th-of-a-kind), we used learning curves to estimate the cost of the interventions. Learning curves are essential tools in assessing the *N*th-of-a-kind cost of new technologies, particularly in fields like CCUS.[49] We followed the approach developed by Rubin *et al.*[50,51] where we first estimated first-of-a-kind costs by applying accurate contingencies, according to technology maturity (Table S7†) and then used leaning curves to approximate costs of *N*th-of-a-kind interventions (eqn (10) and (11). For CO_2 mineralisation, CO_2 capture and calcined clay production we followed Greig, *et al.*[52] and considered 20 built plants as technological maturity. For these interventions we used a learning rate of 10.55%, the mean of suggested learning rates suitable for CO_2 capture technologies Rubin, *et al.*[53] For direct air capture we used estimates for *N*th-of-a-kind plants from Young, *et al.*,[54] which also followed the same methodology. But here, instead of a fixed number of plants, we considered a total cumulative capacity of 1 Gt a^{-1} which is likely to be reached by 2050 [54] because many industries beside the cement industry will likely drive the investments and thus iterative learning of DAC technologies. For biofuel we did not consider any capital expenditures and hence do not consider technological learning. For CO_2 infrastructure (*i.e.*, pipelines, recompression stations between onshore and offshore pipelines) as well as CO_2 injection sites, we did not consider technological learning.

Case study regions

Due to limitations in data availability or quality as well as computational time to solve the model, we selected case study regions in Europe. To cover a wide range of the heterogeneity of regions across Europe, we chose United Kingdom and Ireland, Germany, and Portugal and Spain as case study regions (Table 1, Fig. S5 and Note S6†).

Transport of bulk materials

We evaluated the material transport for all case study regions (*i.e.*, United Kingdom & Ireland, Germany, and Portugal and Spain). We calculated the transport costs of biofuel, minerals, and clay for each region. The results show that large regional differences can be expected (Table 2 and Fig. S6†). Note that end-to-end transport costs were calculated for each feedstock to each single cement plant (see Bulk transport modelling).

Model structure

The constructed model consists of three components: (1) international bulk transport model (INTERNAT-BT), a model that computes transportation costs for various feedstocks (such as olivine-bearing rocks for CO_2 mineralisation), (2) industrial decarbonisation resource technology network model (INDiECAR-RTN), a mixed integer linear programming (MILP) model that allows us to build interventions using component models on existing cement plants and designs a CO_2 storage network (if needed), with the objective to minimise the total system cost (TSC) for a given emission reduction target and (3) cost allocator (COSTALLO), a post-processing model to allocate costs to each cement plant (Fig. 3). The techno-economic model uses cost functions and mass and energy balances for each unique intervention as inputs.

All models were developed using Python 3.9. To solve these complex problems, we used the commercially available solver CPLEX which uses a combination of algorithms (*i.e.*, simplex algorithms, primal-dual logarithmic barrier algorithms, a sifting algorithm) on a high-performance computing cluster. The solver finds quasi-optimal solutions given a set mixed integer programming (MIP) gap (*i.e.*, upper bound to solutions without constraints). The quasi-optimal solutions thus can slightly differ from run to run, given multiple solutions in a certain MIP gap exist.[56] For the majority of the runs, we specified a mip gap of 0.02% or a maximum runtime of 4 hours per run. Fig. 3 gives a high-level representation of the models used and their interactions.

Bulk transport modelling

The transportation cost for bulk goods varies with transportation distance and (combination of) available modalities. To find least cost transport options, we developed a transport model INTERNAT-BT, based on work by Collis and Schomäcker[57] and Benita, *et al.*[58] This model allows comparison of various transport mode combinations, including shipping, rail, and road, to identify the most cost-effective options. For its input data, the model relies on shapefiles representing European railways, sourced from Mapcruzin,[59] offshore shipping routes derived from automatic identification system data obtained from Halpern, *et al.*,[60] and port information sourced from Novikov.[61]

To determine the shortest transportation routes, the model initially creates simplified networks based on the provided

Table 1 Case study descriptions

	United Kingdom and Ireland	Germany	Portugal and Spain
Number of active integrated cement plants	16	31	34
Region characteristics	Much offshore CO_2 storage capacity at close proximity[27]	Limited offshore CO_2 storage capacity located only to the north[27,28,55]	Offshore CO_2 storage in Portugal available[27 a]
	No olivine-bearing rock deposits but kaolinite clays present in the south of England[26]	No olivine-bearing rock deposits available, but large amount of potential kaolinite clay deposits in Germany and surrounding countries (*i.e.*, Hungary, Czech Republic)[24]	Olivine-bearing rock deposits[23] and kaolinite clay deposits available in the north of Spain[25]

[a] Current databases only consider onshore CO_2 storage in Spain,[27] which we do not consider in this study (Note S2).

Table 2 Results of bulk transport calculations (see Bulk transport modelling for methods)

	United Kingdom and Ireland	Germany	Portugal and Spain
Biofuel	6–20 € t^{-1} (transport emissions: 136–183 $kg_{CO_{2e}}$ t^{-1})	17–48 € t^{-1} (transport emissions: 186–327 $kg_{CO_{2e}}$ t^{-1})	14–31 € t^{-1} (transport emissions: 150–200 $kg_{CO_{2e}}$ t^{-1})
Olivine-bearing rocks	5–13 € t^{-1} (transport emissions: 42–64 $kg_{CO_{2e}}$ t^{-1})	7–23 € t^{-1} (transport emissions: 16–85 $kg_{CO_{2e}}$ t^{-1})	3.3–22 € t^{-1} (transport emissions: 12–85 $kg_{CO_{2e}}$ t^{-1})
Clay	11–18 € t^{-1} (transport emissions: 34–80 $kg_{CO_{2e}}$ t^{-1})	1–13 € t^{-1} (transport emissions: 1–16 $kg_{CO_{2e}}$ t^{-1})	9–28 € t^{-1} (transport emissions: 28–104 $kg_{CO_{2e}}$ t^{-1})

shapefiles to reduce computational demands. It then employs the Dijkstra algorithm to compute the shortest paths. However, due to the significantly larger scale of the European road network compared to the rail network, which can result in computationally intensive network analysis, we implemented an application programming interface (API) integration with openstreetmaps.org (**http://openstreetmaps.org/**) (Fig. S2†). The cost of transportation ($C_{\text{transport}}$) for a specified mass flow of bulk material ($\dot{m}_{\text{bulk}}$) are subsequently calculated following eqn (2), considering multiple transport mode options represented by i choices and the calculated distances (dist) on each route as well as specific prices for transport (π).

$$C_{\text{bulk transport}} = \dot{m}_{\text{bulk}} \cdot \text{MIN}_{(i=1\ldots n)}[\pi_{\text{truck}} \cdot \sum \text{dist}_{\text{truck}} + \pi_{\text{train}} \cdot \sum \text{dist}_{\text{train}} + \pi_{\text{ship}} \cdot \sum \text{dist}_{\text{ship}}] \quad (2)$$

We calculated transport cost routes comparing 4 different possible combinations: (1) transport *via* ship, rail, and road, (2) transport *via* ship and road, (3) transport *via* rail and road, and (4) transport solely *via* road (Fig. S2†). To reduce the number of runs, we only calculated the least-cost transport routes for a given bulk material (*e.g.*, olivine bearing rock) by first finding the *n*-closest feedstock locations based on the linear distance between resource and the cement plant, with *n* reaching from 2–4 depending on the feedstock (*i.e.*, we limited finding the least-cost route for kaolinite clay for a given cement plant to the 4 closest clay sites and disregarded clay sites further away).

Depending on the selected modes of transport, the accuracy of the transport model differs due to variable data quality. The highest accuracy can be assumed for road transport as the route is calculated using an API integration to openstreetmaps.org (**http://openstreetmaps.org/**) (accuracy of 4–5*m* [62] reported). The used shapefiles for train and ship transport are shown in Fig. S10.† The least accurate transport cost calculation can be expected from ship transport as the shapefile uses existing transport data (with highest resolution in North America) and therefore some less-frequented routes may not be accurately captured. However, given the small costs of offshore shipping, it does not have a significant impact on results. For train transport the entire European railways network was considered. However, in reality freight trains may have to travel a longer distance as in some country's passenger trains may have priorities on certain routes.

Quantity of interest

As the quantity of interest for this geospatial-economic model, we here chose the change in levelised cost of product ΔLCOP (*i.e.*, added costs per tonne of cement $\dot{m}_{\text{cement}}$) in [€ t_{cement}^{-1}], which combines capital costs, here, total capital requirements (TCR) and operational expenditures (OpEx) for each cement plant location *g*. Note that we only consider ordinary Portland cement with a clinker factor of 100%, which can be used as is, or to formulate other standard cement blends.

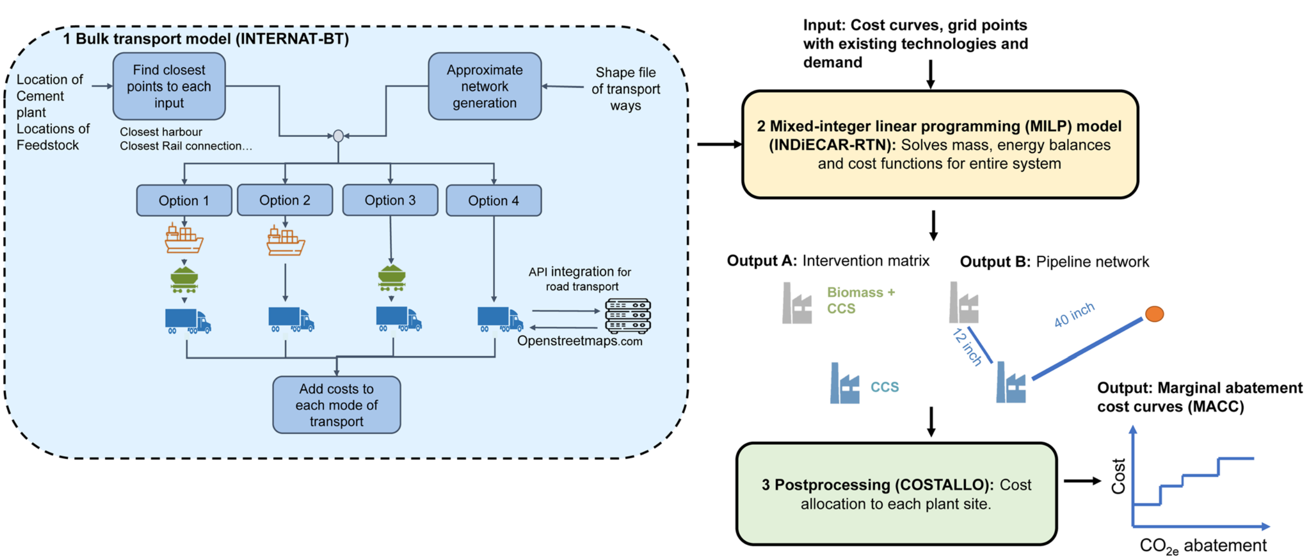

Fig. 3 Schematic representation of developed model. Additional details in Fig. S1–S4.†

To calculate ΔLCOP, we applied the methodology described earlier in Strunge *et al.*[36] which we developed in accordance to existing guidelines for techno-economic assessments in this field.[50,63,64] We discounted the capital costs (*i.e.*, TCR) using interest rate i and the plant's expected lifetime L.[65] The interventions can be built at individual cement plants, while CO_2 transport infrastructure, storage injection wells and direct air capture plants are investments shared by multiple plants. Therefore, they were allocated towards each cement plant location g that uses them (eqn (3) till eqn (8)).

$$\Delta \mathrm{LCOP}_{\mathrm{total},g} = \Delta \mathrm{LCOP}_{\mathrm{intervention},g} + \Delta \mathrm{LCOP}_{\mathrm{CO_2\ transport},g} + \Delta \mathrm{LCOP}_{\mathrm{CO_2\ storage},g} + \Delta \mathrm{LCOP}_{\mathrm{NET},g} \quad (3)$$

$$\Delta \mathrm{LCOP}_{\mathrm{intervention},g} = \frac{\alpha \cdot \mathrm{TCR}_{\mathrm{intervention},g} + \mathrm{OpEx}_{\mathrm{intervention},g}}{\dot{m}_{\mathrm{cement},g}} - \frac{\mathrm{OpEx}_{\mathrm{savings},g}}{\dot{m}_{\mathrm{cement},g}} \quad (4)$$

$$\Delta \mathrm{LCOP}_{\mathrm{CO_2\ transport},g} = \sum_{y_g} \left(\frac{\alpha \cdot \mathrm{TCR}_{\mathrm{pipeline},y} + \mathrm{OpEx}_{\mathrm{pipeline},y}}{\dot{m}_{\mathrm{CO_2 transported,pipeline},y}} \right) \cdot \left(\frac{\dot{m}_{\mathrm{CO_2 stored},g}}{\dot{m}_{\mathrm{CO_2 transported,pipeline},y}} \right) \cdot \left(\frac{\dot{m}_{\mathrm{CO_2 stored},g}}{\dot{m}_{\mathrm{cement},g}} \right) \quad (5)$$

$$\Delta \mathrm{LCOP}_{\mathrm{CO_2\ stroage},g} = \sum_{j} \frac{\alpha \cdot \mathrm{TCR}_{\mathrm{storage}} + \mathrm{OpEx}_{\mathrm{storage}}}{\dot{m}_{\mathrm{stored},j}} \cdot \left(\frac{\dot{m}_{\mathrm{CO_2 stored},g} + \dot{m}_{\mathrm{CO_2\ offset},g}}{\dot{m}_{\mathrm{cement},g}} \right) \quad (6)$$

$$\mathrm{LCOP}_{\mathrm{CDR},g} = \sum_{h} \frac{\alpha \cdot \mathrm{TCR}_{\mathrm{CDR},h} + \mathrm{OpEx}_{\mathrm{CDR},h}}{\dot{m}_{\mathrm{removed},h}} \cdot \left(\frac{\dot{m}_{\mathrm{CO_2 offset},g}}{\dot{m}_{\mathrm{cement},g}} \right) \quad (7)$$

$$\alpha = \left(\frac{i}{1-(1+i)^{-L}} \right) \quad (8)$$

Here, $\dot{m}_{\mathrm{cement},g}$ represents the cement plant's capacity in location g, y_g represents the set of pipeline segments CO_2 is transported through from location g to the storage site, $\dot{m}_{\mathrm{CO_2 transported,pipeline},y}$ describes the total amount of CO_2 transported in pipeline y, $\dot{m}_{\mathrm{CO_2 stored},g}$ describes the amount of CO_2 captured and stored from the cement plant in location g and $\dot{m}_{\mathrm{CO_2 offset},g}$ represents the amount of emitted CO_2 from a cement plant in location g that is offset using carbon dioxide removal technologies at location g or another location. $\mathrm{OpEx}_{\mathrm{savings},g}$ refers to operational expenditures that are saved compared to conventional production (*e.g.*, costs of energy, feedstock for clinker replacement by novel supplementary cementitious material).

We calculated the TCR for interventions using the total direct costs (TDC) as well as total overnight costs (TOC) for each technological intervention (eqn (9)).

$$\mathrm{TOC}_{\mathrm{intervention}} = \sum \mathrm{TDC} \cdot (1+f_{\mathrm{indirect}}) \cdot (1+f_{\mathrm{process}}) \cdot (1+f_{\mathrm{project}}) \cdot (1+f_{\mathrm{owner}}) \quad (9)$$

The factors $f_{\mathrm{indirect}}, f_{\mathrm{process}}, f_{\mathrm{project}}, f_{\mathrm{owner}}$ account for indirect costs, process contingencies, project contingences and owners' costs respectively. To calculate the TCR for an nth of a kind plant we used (eqn (10) and (11)).[50,66]

$$\mathrm{TCR}_{\mathrm{intervention}} = \left(\frac{\mathrm{TPC}}{\dot{m}_{\mathrm{cement}}} \right) \cdot N^{-E} \cdot \dot{m}_{\mathrm{cement}} \cdot (1+i)^{t_{\mathrm{construction}}} \quad (10)$$

$$E = \frac{\ln(1-\mathrm{LR})}{\ln(2)} \quad (11)$$

N characterizes the number of plants built, LR the learning rate, E the experience factor, i the interest during construction and $t_{\mathrm{construction}}$ the estimated time for construction in years. The OpEx were derived using mass and energy balances as a basis to calculate the costs of utilities and feedstocks, the location specific costs of material transport and the costs of labour (eqn (12)). Here, the amount of feedstock or utility needed is represented byw_i, $\pi_i$is the price for feedstock or utility for resource i and location g.

$$\mathrm{OpEx} = \sum w_i \pi_i + \mathrm{OpEx}_{\mathrm{fixed}} + \sum C_{\mathrm{bulk\ transport},i,g} \quad (12)$$

To scale the TCR for a given cement plant, we used a linear scaling approach, so it could be used in a linear programming formulation.

$$\mathrm{TCR}_{\mathrm{capacity\ new}} = \mathrm{TCR}_{\mathrm{capacity_{old}}} \cdot \left(\frac{\mathrm{capacity_{new}}}{\mathrm{capactiy_{old}}} \right) \quad (13)$$

To evaluate the CO_{2e} emissions offset by an intervention, we modified the approach used by Ostovari, *et al.*[67] In line with their methodology, we calculated the climate change impacts following the Intergovernmental Panel on Climate Change by using the European Commission's recommendations in the International Reference Life Cycle Data Handbook.[68]

For assessing the climate change effects of an intervention, we used the carbon footprinting method. We limited this study's scope to climate change impacts measured in CO_{2e} emissions, considering other impacts as beyond our study's techno-economic focus. In the emissions $e_{\mathrm{interventions}}$, we accounted for the emissions by the used feedstocks $e_{\mathrm{feedstocks}}$, their transport to the cement site $e_{\mathrm{transport}}$ (containing emissions from each used mode of transport), emission for electricity $e_{\mathrm{electricity}}$ and heat e_{heat} used in the process. From that burden we subtracted the emission reductions through CO_2 that is bound in the product or stored offshore, e_{stored}, and emissions that are avoided by replacing clinker production e_{replace} (eqn (14)).

$$e_{\mathrm{intervention}} = (e_{\mathrm{feedstocks}} + e_{\mathrm{transport}} + e_{\mathrm{el}} + e_{\mathrm{heat}}) - (e_{\mathrm{stored}} + e_{\mathrm{replace}}) \quad (14)$$

Optimisation framework

The optimisation framework INDIeCAR-RTN used in this work was built on a resource technology network (RTN) formulation by Sunny, *et al.*[69] based on Pantelides.[70] We described our adapted framework in Küng, *et al.*[71] In this RTN model we described a region using multiple grid cells, where each cell (g) represented a specific functional location (*i.e.*, cement plant site, potential CO_2 storage site, CO_2 terminal site to connect offshore pipelines to the onshore grid or trunkline location where shared pipelines networks can be pre-defined, Table S3†). The optimisation model's objective function was to minimise the total added levelised costs for cement production ($\Delta LCOP_{total}$) as a sum over the set of all cement plant locations (G_c) (eqn (15)) for a given emission reduction target.

$$\text{MIN}\sum_{g\in G_c}\text{LCOP}_{\text{total},g} \tag{15}$$

To solve this problem, we used a set of equations as constraints. The core element in the RTN formulation are sets of resources including cement, biofuel, olivine bearing rocks, emitted CO_2, captured, and liquidised CO_2 as well stored CO_2. The model must solve these mass balances for each grid cell and each resource. Beside the resources, the model contains interventions that can be installed at grid cell locations (*e.g.*, installing CO_2 capture at a cement plant to reduce the emitted CO_2 and convert it into captured CO_2 or installing injection wells to convert captured CO_2 into stored CO_2). Additionally, the model contains a range of infrastructure interventions to transport resources between grid cells, such as onshore CO_2 pipelines of different sizes. The general concept of the RTN formulation entails that all resources within each grid cell must be balanced, *i.e.*, production within the cell, flows to the cell *via* transport, local demand and outflows (eqn (16))

$$\begin{aligned}\varepsilon_{r,g,t} = \text{imp}_{r,g,t} - \text{dem}_{r,g,t} + \sum_{j}^{\text{intervention}}\left(\mu_{j,r}\cdot p_{j,g,t}\right) + \sum_{j}^{\text{Strg}}\left(\lambda_{j,\text{CO}_2,g,t}{}^{S}\right) \\ + \sum_{j}^{\text{CDR}}\left(\mu_{j,r}\cdot p_{j,g,t}\right) + \sum_{\hat{g}}^{G}\sum_{d}^{D}\left(q_{(\hat{g},g,r,d,t)} - q_{g,\hat{g},r,d,t}\right)\end{aligned} \tag{16}$$

Here, the term $\varepsilon_{g,r,t}$ denotes the emission rate of resource r in grid cell g at time t, $\text{imp}_{r,g,t}$ signifies the rate of importing resource r (importable resources are for example electricity and olivine bearing rocks which are imported from outside of the grid cell). The summation $\sum_{j}^{\text{intervention}}(\mu_{j,r}\cdot p_{j,g,t})$ describes the conversion rate of the resource for all installed interventions with $\mu_{j,r}$ being the conversion rate of resource r (*e.g.*, biofuel usage) by intervention j (*e.g.*, fuel switching to biofuel), while $p_{j,g,t}$ is the production rate of the (installed) intervention j (*e.g.*, output of cement plant) with a negative value indicating resource consumption and a positive value indicating resource production. The term $\sum_{j}^{\text{Strg}}(\lambda_{j,r,g,t}{}^{S})$ sums up all storage technologies in a grid cell where $\lambda_{j,r,g,t}^{S}$ delineates the rates of storing resource CO_2 using storage technology j, *i.e.*, injection wells. The term $\sum_{j}^{\text{CDR}}(\mu_{j,r}p_{j,g,t})$ sums up all carbon dioxide removal technologies, which can be installed at any grid cell and here consist of direct air capture technologies. Lastly, the final term considers the inflow and outflow of resources, denoted as $q_{\hat{g},g,r,d,t}$ and $q_{g,\hat{g},r,d,t}$ respectively, between grid cell g and its counterpart $\hat{g}$ through distribution installation d (*e.g.*, 9 inch onshore CO_2 pipeline).

In the RTN model, all resources must be balanced meaning the overall sum must be zero, except for CO_2. Excess CO_2 is released to the environment. Eqn (17) establishes the total net CO_2 emissions for the system over all grid cells (G) and major time frames (T).

$$m_{\text{CO}_2} = \sum_{g}^{G}\sum_{t}^{T}\varepsilon_{\text{CO}_2,g,t}\cdot\text{duration}_t \tag{17}$$

To solve case studies, we hence set a CO_2 reduction objective, which served as a constraint to the solver while the cement production of each plant needs to be fulfilled. Emissions can be reduced through either intervention, such as implementing carbon capture processes, or *via* installing CDR (here only direct air capture plants).

Each action, *e.g.*, choosing to build an intervention in a grid cell, comes with associated costs as total added levelised costs for cement production ($LCOP_g$). The solver's objective is to determine the optimal combination of technologies and infrastructure choices (integer decisions) that can meet the resource demands as constrained by eqn (16), comply with the CO_2 reduction constraint, and simultaneously minimise a cost-based objective function (eqn (15)).

The outcomes generated by this optimisation framework represent economically and environmentally optimal clusters of configurations that fulfil a predefined CO_2 or greenhouse gas (GHG) reduction target. To investigate optimal investment sequences while avoiding lock-in effects, we implemented a back-casting approach, where the solver starts with interventions selected for full decarbonisation and when given a decarbonisation goal (*e.g.*, 50%), it selects the least-cost options among these interventions.

Post-processing

Given that INDiECAR-RTN finds a least-cost solution for the entire system, we developed a post-processing model COSTALLO to allocate the costs towards the individual cement plants. Here, interventions built at a cement plant were directly allocated to the cement plant located in this grid cell (eqn (4)) and costs for CO_2 transport, storage and direct air capture (eqn (5)–(7)) were shared and only partially allocated towards each cement plant (Fig. S4†). While for CO_2 storage and direct air capture installation the costs were allocated considering a plant's captured CO_2 and offset CO_2 respectively, we used the Dijkstra algorithm to determine the route of captured CO_2 from each cement plant to a connected storage site. All pipeline investments were shared among all cement plants using a specific pipeline section on their route according to their amount of captured CO_2 and length travelled on the pipeline section. This approach also allowed us to introduce shared infrastructure with other industries (*i.e.*, trunk lines) and allocate the costs towards by the solver connected cement plants.

Results

We here present the results of the analysis across three case study regions (UK&IRE, GER and PT&ES), selected for their different levels of access to feedstocks and offshore CO_2 storage.

Full decarbonisation of the cement industry will require large investments in CO_2 transport and storage networks and carbon dioxide removal

The model results show that calcined clay cements coupled with CCS and DACCS are the least-cost ways to fully decarbonise the cement industry for all regions studied (Fig. 4), due to favourable economics for calcined clay cements (because of its low heat requirements, Note S4†) and comparably short transport distances onshore. High transport costs and emissions for both biofuels and CO_2 mineralisation (Table 2) limit their economic viability compared to the use of low-cost clays as clinker replacements in the cement industry. Notably, for one plant in the UK (*i.e.*, Wales), distant to the designed CO_2 transport network and proximity to kaolinite clay sites in South England (Fig. 2A) no CCS was chosen by the solver. Showing that in case of large distances to shared infrastructure, it can be least-cost to divert from using CCS as connecting pipelines would fully have to covered by this individual cement plant (Methodology – Post-processing).

Our model predicts the full decarbonisation of the different case study regions could be achieved with added levelised costs of (cement) production (ΔLCOP) of €75 per tonne of ordinary Portland cement (t_{cement}^{-1}) (UK&IRE), €71 t_{cement}^{-1} (GER), €74 t_{cement}^{-1} (PT&ES) compared to European average levelised cement costs of €46–€51.5 t_{cement}^{-1} (Fig. 4). This translates into annually added costs for the construction industry of €738M a^{-1} (UK&IRE), €2555M a^{-1} (GER) and €1529M a^{-1} (PT&ES) or 0.4% (UK&IRE), 0.7% (GER) and 0.9% (PT&ES) of the industry's annual production value.[72,73] Assuming the case studies are representative of the entire European Union and United Kingdom, added costs of approximately €19 billion annually could be expected, approximately 0.1% of their gross domestic product.[74,75] Most of the investment costs lie in direct air capture plants with €2322M (UK&IRE), €4820M (GER), €6933M (PT&ES) followed by CO_2 transport and storage infrastructure with €1524M (UK&IRE), €3302M (GER), €5035M (PT&ES) (Fig. 4). Direct air capture and CO_2 transport and storage accounted for 49–53% and 35–39% of total investment costs (which don't take place at cement production sites), while the interventions on the cement sites accounted for 12% of the total investment costs only. The high observed cost share for DACCS is commensurate with the observation that the other interventions are unable to fully decarbonise cement plants, even when implemented in tandem.

Collaboration between companies to design CO_2 storage transport and storage networks significantly reduces costs for the majority of plants

The above results were generated assuming full collaboration and coordination between cement producers in a country/region (Fig. 4). This is a strong assumption and to test its effect we quantified the difference in total decarbonisation cost if coordination only happens within, but not between holdings (the European cement market consists of a few large parent companies owning multiple plants in a region as well as smaller producers owning only a single or few plant(s) (Table S26†)). To that end, we ran the simulation again for each parent company, where a decarbonisation strategy must be found only taking the plants of one company into account (*e.g.*, when CO_2 transport and storage was selected, a pipeline network was optimised for all plants in a parent company's portfolio). The difference in costs between a collaborative approach (Fig. 4) and a non-collaborative approach for each plant is shown in Fig. 5. The results show that this non-collaborative, uncoordinated approach could yield significantly higher costs to reach full decarbonisation: additional annual costs – on top of the previously determined costs for decarbonisation in a collaborative approach (Fig. 4) – were calculated as €94M a^{-1} (UK&IRE), €159M a^{-1} (GER) and €116.5M a^{-1}(PT&ES) respectively. Per tonne of cement this translates into additional costs of 2.4€ t_{cement}^{-1} (PT&ES) to 6.1€ t_{cement}^{-1} (UK&IRE), Table S26.† While the presented costs can be seen as an upper bound as we assume an operation at full capacity with an average clinker factor of 0.737,[76] Fig. 5 illustrates that in this scenario these additional costs for decarbonisation will mostly have to be covered by a few companies, primarily by the companies that operate fewer plants. Additionally, in the case of central Spain the results illustrate that for some plants an uncoordinated approach could also lead to slightly lower costs for individual plants compared to a fully coordinated approach. This cost reduction was shown for some plants where the non-collaborative approach led to significantly smaller transport distances for CO_2 (*i.e.*, a direct pipeline was built to the shore where in the collaborative approach the pipeline had to be diverted to other cement plants).

Elaborate CO_2 transport networks are key to lower costs of reaching net-zero CO_{2e} emissions. Absence leads to biomass use

Least-costs strategies for full decarbonisation will need CO_2 transport and storage, and therefore elaborate CO_2 transport networks. The absence of such infrastructure will increase the costs of reaching full decarbonisation by another €9 t_{cement}^{-1} to €10 t_{cement}^{-1} (Fig. 6) translating into additionally added costs of €99M a^{-1} (UK&IRE), €284M a^{-1} (GER) and €186M a^{-1} (PT&ES) respectively. In the absences of CO_2 transport infrastructure, biofuel becomes a key strategy for many cement plants limited to transport costs (plants further inland do not select biofuel). While biofuel becomes a prevalent strategy in the absence of CO_2 transport infrastructure, the results show comparably small differences between plants using biofuel and the ones which do not. Note that in this study we solely considered biomass as fuel from North America, due to the limited availability of this resource in Europe (Table S1†), leading to comparably high transport costs as well high emissions (Table 2).

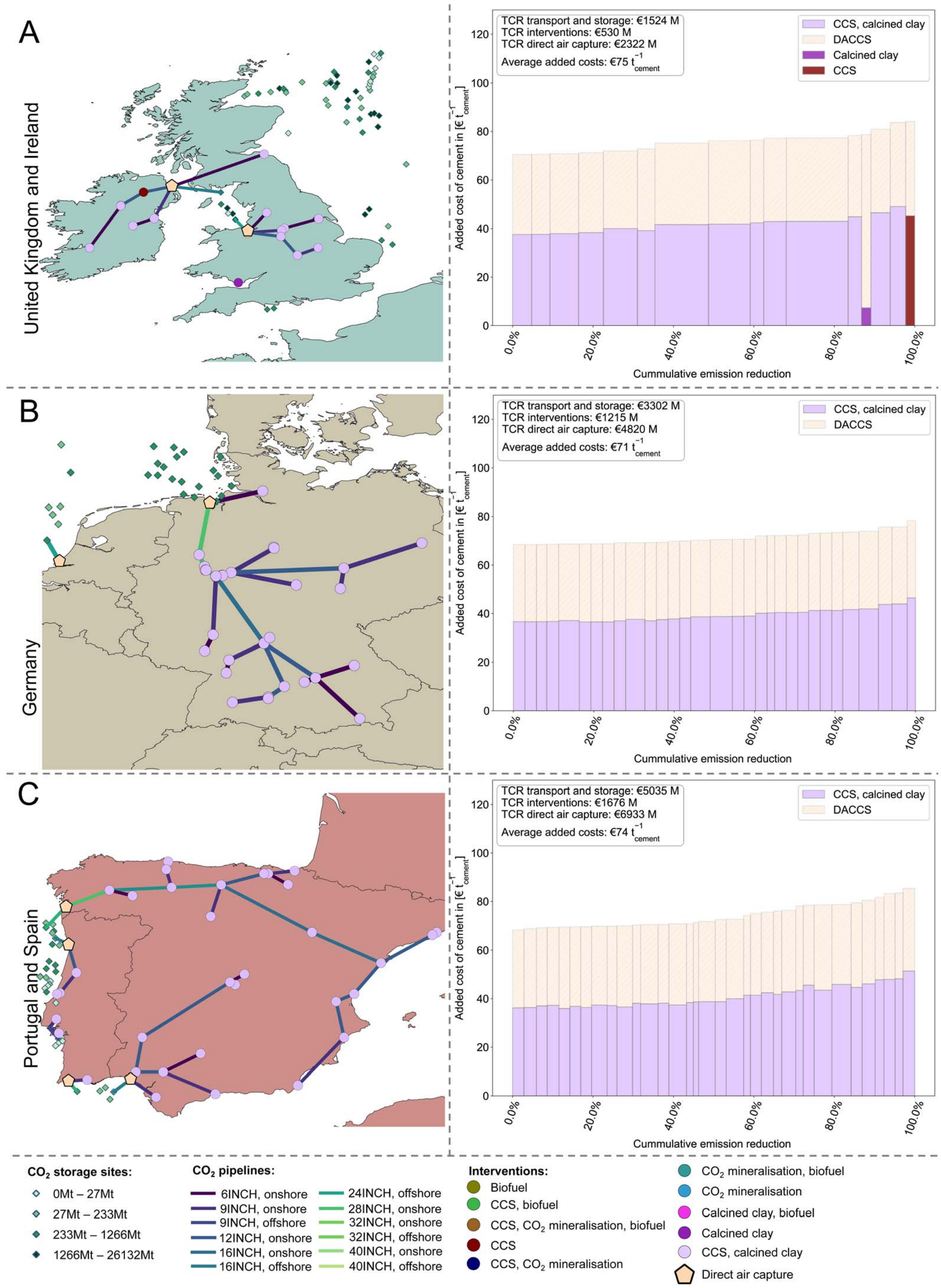

Fig. 4 Model results for reaching net-zero-CO_{2e}-emissions in (A) United Kingdom and Ireland, (B) Germany and (C) Portugal and Spain. Each circle represents one integrated cement plant and each orange pentagon a direct air capture location. Circle colours represent interventions. Line colours indicate CO_2 transport pipe thickness. Right panels show marginal CO_{2e} abatement cost curves for each region, each bar represents a single cement plant. Total capital requirements (TCR) describe the capital expenditures (CAPEX) of an investment. Assumptions and used calculations shown in Tables S6–S12, S14–S24.†

Calcined clay cements could become a breakthrough technology overall lowering the costs of full decarbonisation by €9–€15 per tonne of cement

As shown in (Fig. 4), calcined clay cements coupled with CCS could become a breakthrough technology for deep decarbonisation due to their low heat requirements. Scenarios excluding the use of calcined clay cements increase the estimated added levelised costs by €9 t_{cement}^{-1} to €15 t_{cement}^{-1} (compare Fig. 4–7). For plants located further inland, bio-fuel was not a least-cost intervention. Notably, for plants not using

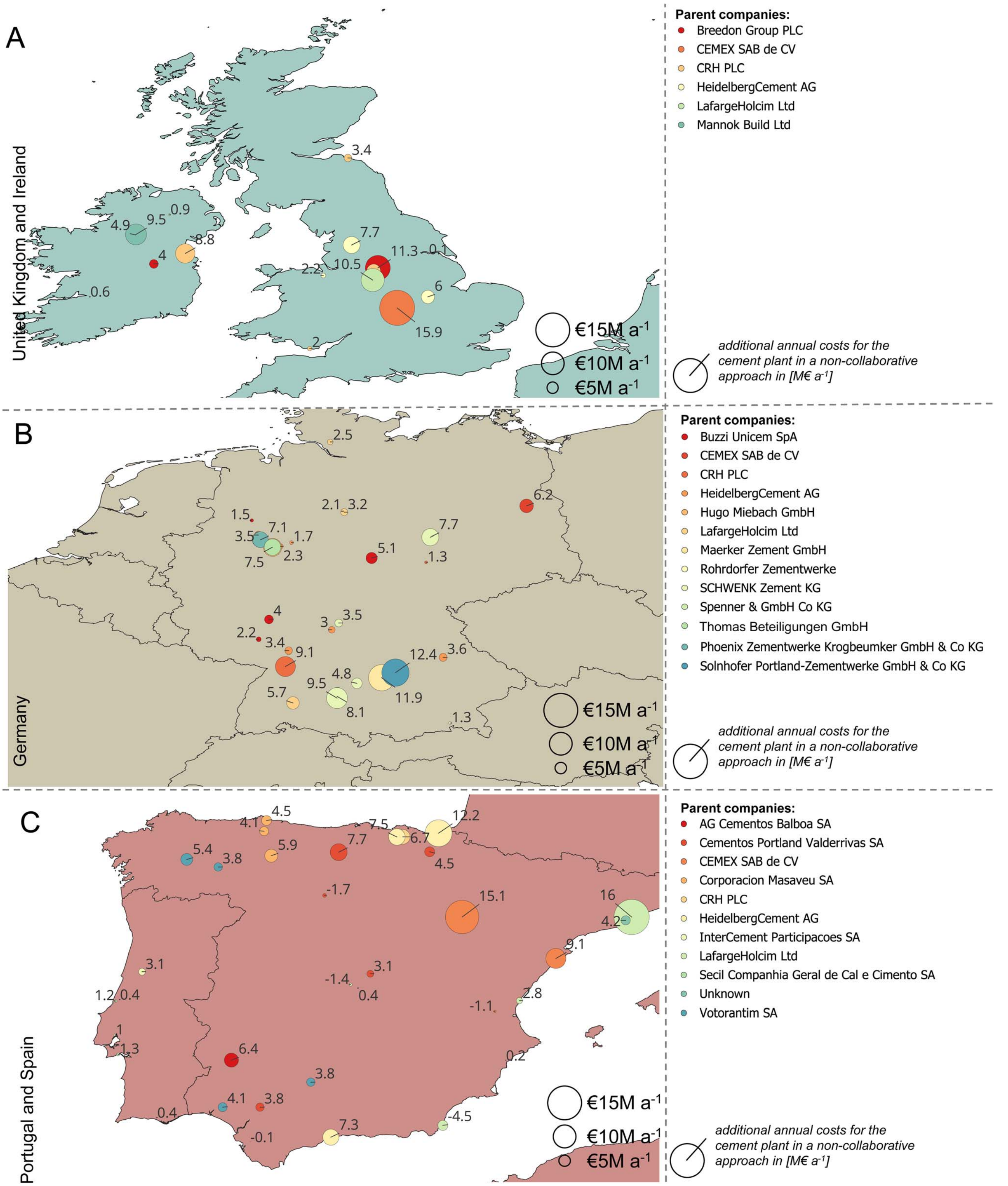

Fig. 5 Additional costs for reaching full decarbonisation in a non-collaborative approach compared to the coordinated approach. (A) United Kingdom and Ireland, (B) Germany and (C) Portugal and Spain. Model optimisations were done for each parent company separately. Each circle represents one integrated cement plant. The size of the circle indicates additional annual costs in million € per year (M€ a^{-1}) compared to the whole system optimisations in Fig. 4. The colour of each circle indicates the parent company taken from Tkachenko, *et al.*[22] Note, colours between panels (*e.g.*, A and B) may have been used multiple times for different parent companies. Detailed results shown in Table S26.† Note, to increase visibility pipelines are not shown here but individual pipeline networks were built by the solver for each parent company.

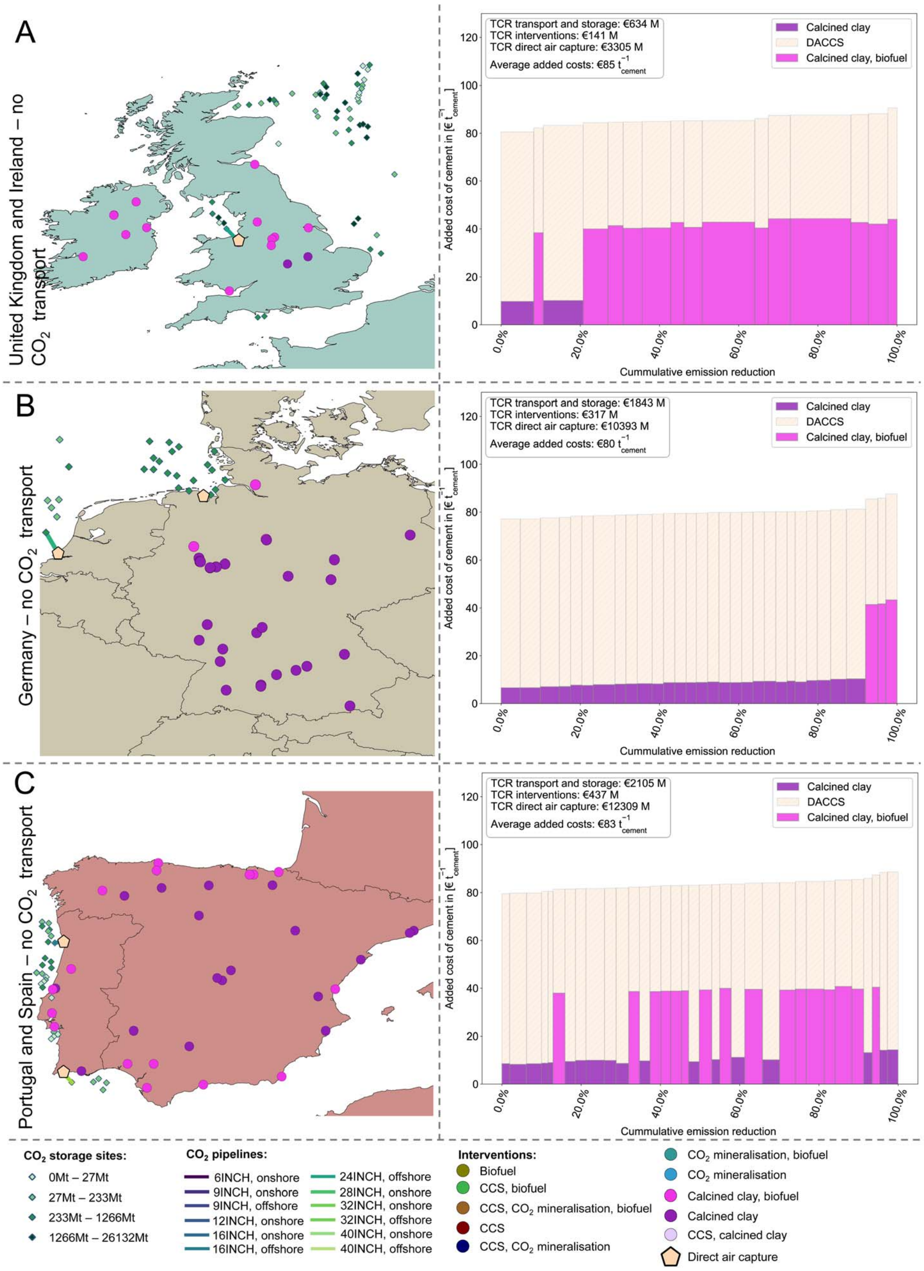

Fig. 6 Model results for reaching full decarbonisation in the absence of onshore CO_2 transport in (A) United Kingdom and Ireland, (B) Germany and (C) Portugal and Spain. Each circle represents one integrated cement plant and each orange pentagon a direct air capture location. Circle colours represent interventions. Line colours indicate CO_2 transport pipe thickness. Right panels show marginal CO_{2e} abatement cost curves for each region, each bar represents a single cement plant. Total capital requirements (TCR) describe the capital expenditures (CAPEX) of an investment Assumptions and used calculations shown in Tables S6–S12, S14–S24.†

biofuel, the majority of the decarbonisation costs lie in the costs for DACCS plants, which are not located at the cement plant itself. The results show that when CCS is combined with the use of biofuels, a significantly smaller share of the costs was used for DACCS because its high decarbonisation through storage of biogenic CO_2 leading to carbon dioxide removal from the atmosphere[5] (*i.e.*, when excluding emissions from transport of CO_2 and feedstocks CCS with biofuel leads to 91% emission from 850 $kg_{CO_{2e}}$ t_{cement}^{-1} to 77 $kg_{CO_{2e}}$ t_{cement}^{-1} or 93–94% emission reduction when coupled with CO_2 mineralisation or calcined clay cements, Fig. S8†). Note that this scenario also requires a CO_2 pipeline from Germany to Dutch offshore storage sites, resulting from increased CO_2 transport volumes.

Local resources (*e.g.*, olivine bearing rocks) can become key if widespread implementation of breakthrough technologies fail

In the absence of calcined clay cements (*e.g.*, due to low acceptance by the cement market), the use of CO_2

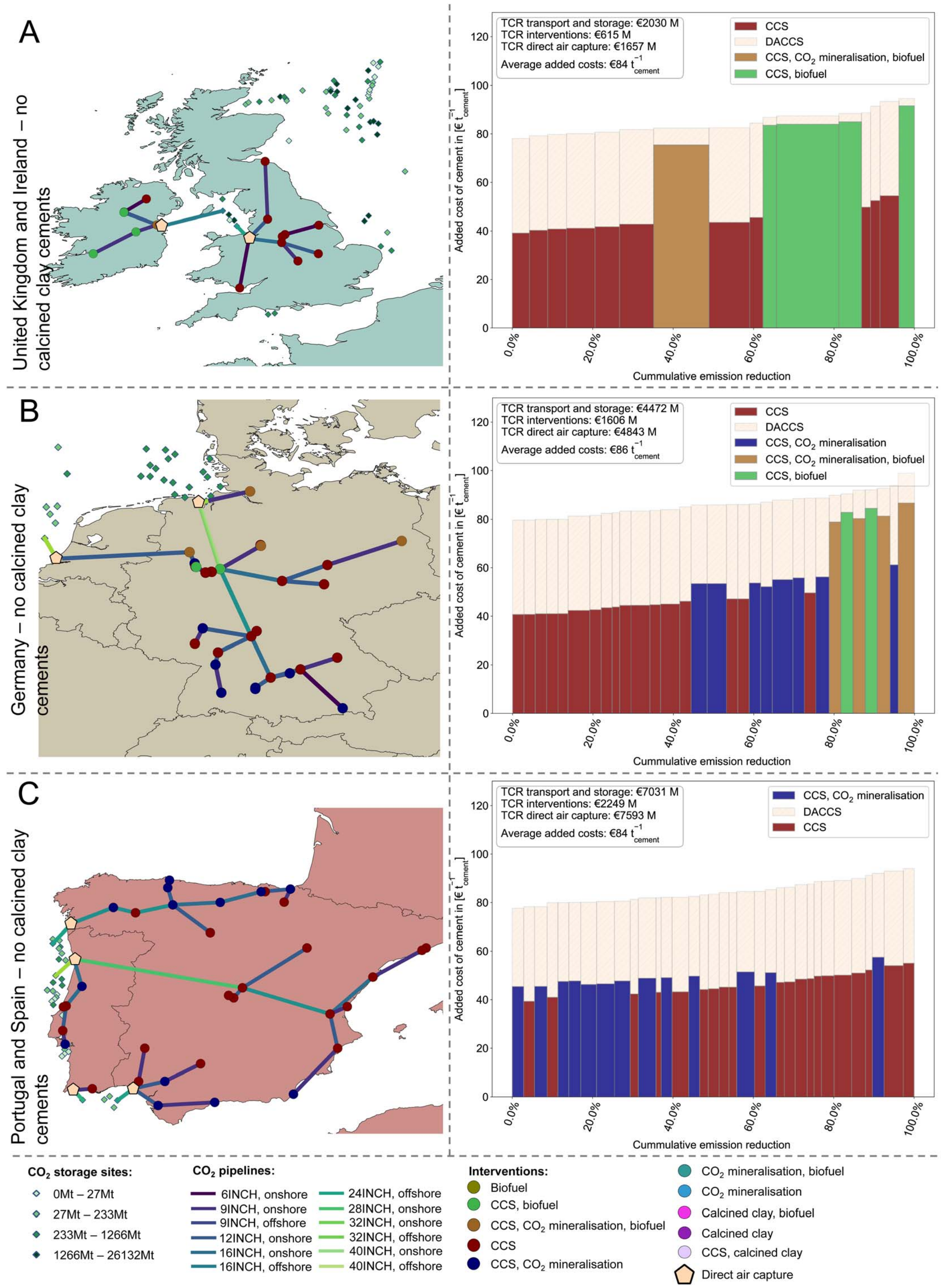

Fig. 7 Model results for reaching full decarbonisation without calcined clay cements in (A) United Kingdom and Ireland, (B) Germany and (C) Portugal and Spain. Each circle represents one integrated cement plant and each orange pentagon a direct air capture location. Circle colours represent interventions. Line colours indicate CO_2 transport pipe thickness. Right panels show marginal CO_{2e} abatement cost curves for each region, each bar represents a single cement plant. Total capital requirements (TCR) describe the capital expenditures (CAPEX) of an investment. Assumptions and used calculations shown in Tables S6–S12, S14–S24.†

mineralisation or biofuels become key strategies contingent on low cost of transport (*i.e.*, local availability of feedstock or access to offshore transport lead to significant differences in transport costs, Table 2). In Germany and Portugal and Spain, the use of CO_2 mineralisation becomes the main strategy after CCS (Fig. 7). In Spain, it is mostly plants located close to the coast that select this option, limited by high onshore transportation costs. In Germany, both in the north (close to the shore) and in the south (proximity to olivine-bearing rock deposits in northern Italy, Fig. 2) this intervention was chosen. Generally, in the absence of calcined clay options, a variety of different strategies are chosen and local differences play a larger role in selecting cost-optimal interventions (*e.g.*, proximity to feedstock or storage site).

Designing CO_2 transport networks should be coordinated with other industries which also plan to implement CCS

It is unlikely the cement industry will be responsible for constructing their own CO_2 transport infrastructure, this may rather become a commodity shared with other industries (*e.g.*, steel production, waste incineration). We investigated the use of predefined trunk lines which are only partially used by the cement industry. We modelled currently planned projects for the United Kingdom and Germany (Table S4†), to which the solver was allowed to build connections. The solutions obtained showed to be similar to those without trunk lines (compare Fig. 4–8), strengthening the robustness of the of the network designs obtained earlier. Note this analysis was only run for the British Isles and Ireland, and for Germany. To our knowledge there are no current offshore CO_2 transport plans for the Iberian Peninsula. The costs of decarbonising the cement industry may be fully offset by expected costs for greenhouse gas emission certificates, but first-of-a-kind investments remain to be overcome.

While these presented costs might appear high compared to current costs for cement production, they must be compared to future costs of unabated production including the societal costs of emitting CO_{2e}. Generally, future costs for cement production will significantly increase during the coming decades due to increasing costs of CO_2 emission allowances (*i.e.*, in the European Union the EU emission trading system (EU ETS) and in the United Kingdom the UK ETS), which aim to incentivise implementation of emission reduction strategies. In the EU and the UK as well in some other regions of the world (*i.e.*, California),

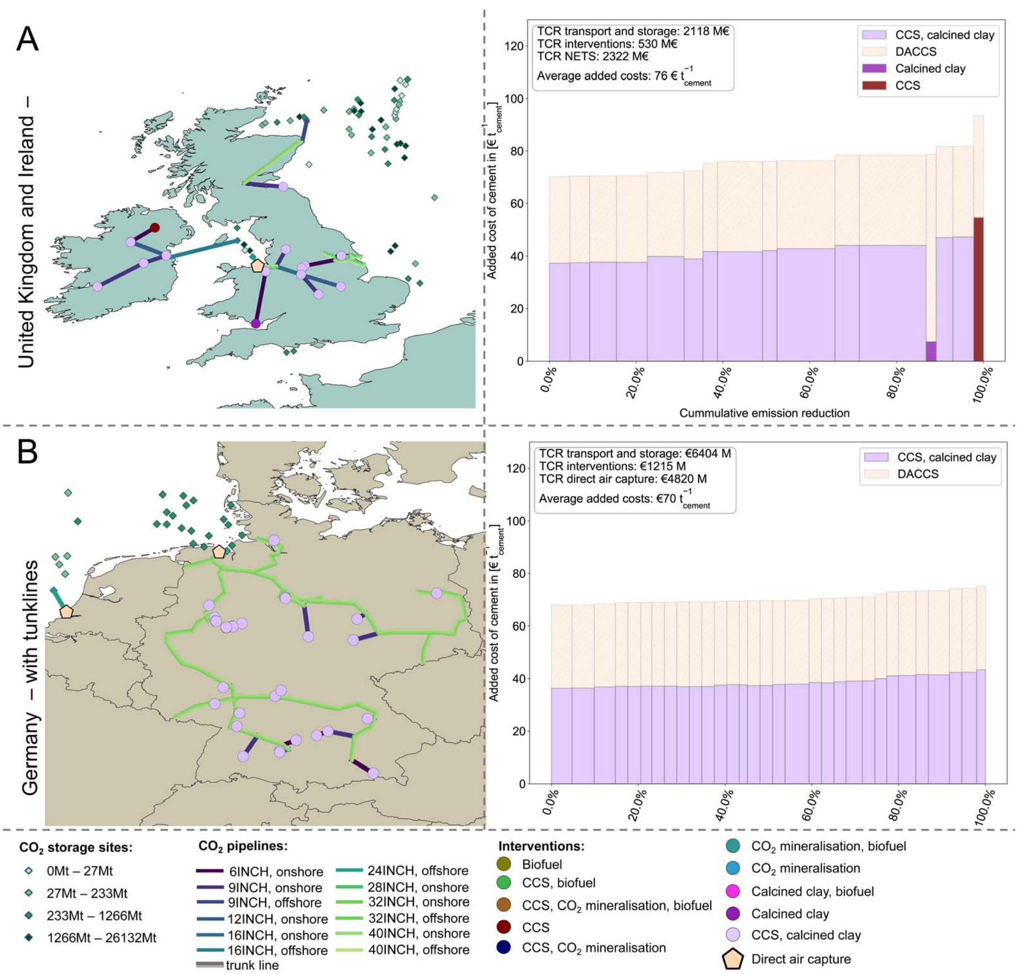

Fig. 8 Model results for reaching full decarbonisation considering currently planned CO_2 pipelines in (A) United Kingdom and Ireland, (B) Germany. Each circle represents one integrated cement plant and each orange pentagon direct air capture location. Circle colours represent interventions. Line colours indicate CO_2 transport pipe thickness. Line fading indicates a trunk line. Right panels show marginal CO_{2e} abatement cost curves for each region, each bar represents a single cement plant. Total capital requirements (TCR) describe the capital expenditures (CAPEX) of an investment. Assumptions and used calculations shown in Tables S4, S6–S12, S14–S24.†

CO_2 allowances use a "cap and trade" basis. Legislators establish a maximum limit (cap) on the total greenhouse gas emissions allowed for covered installations. This cap diminishes each year according to climate targets and the certificate prices are determined by the market.[77] Therefore, future costs of emitting CO_2 are likely to increase but depend on emission reduction of other plants (*i.e.*, demand and supply of certificates). Price estimates differ but are expected to lie between €130 $t_{CO_{2e}}^{-1}$ and €160 $t_{CO_{2e}}^{-1}$ in 2030 (€56 $t_{CO_{2e}}^{-1}$ and €111 $t_{CO_{2e}}^{-1}$ in 2025),[78] leading to added levelised costs for ordinary Portland cement production of €111 t_{cement}^{-1} to €136 t_{cement}^{-1}, significantly higher than all calculated costs in this study (Fig. 9A). While costs of certificates are likely to depend on market fluctuations and do not reflect the actual costs of emitting greenhouse gas emissions into the atmosphere, some suggest the consideration of social cost of carbon dioxide, which reflect the damages to society of emitting a tonne of CO_{2e} and is a key indicator for climate policy development.[79] Estimates for social cost for CO_{2e} vary from \$44 $t_{CO_{2e}}^{-1}$ to \$413 $t_{CO_{2e}}^{-1}$ (€40 $t_{CO_{2e}}^{-1}$ to €371 $t_{CO_{2e}}^{-1}$) with the mean being at \$185 $t_{CO_{2e}}^{-1}$ (€167 $t_{CO_{2e}}^{-1}$).[79] Considering the social costs for CO_{2e} emissions, cement production (ordinary Portland cement) hence would increase by €142 t_{cement}^{-1}, significantly outweighing the here calculated costs for full decarbonisation.

To compare costs of different technologies long term, technology maturity of the interventions must be considered. To this end, up to this point we only considered interventions at mature stage based on learning curve estimates[49] (Fig. 4–8). For interventions like carbon capture or CO_2 mineralisation we considered 20 plants[52] to be built with significantly higher costs to reach maturity, while for direct air capture a cumulative capacity of 1 Gt_{CO_2} must be installed to reach the costs used here[54] (Methodology – Iterative learning for CO_2 emission reduction strategies). We repeated the analysis shown in (Fig. 4), assuming each region would independently build first-of-a-kind (FOAK) plants, second-of-a-kind plants and so forth (Fig. 9B–D). This shows these first decarbonisation projects will be significantly more expensive. While ETS prices expected for 2030 will increase costs for cement production to a similar level as FOAK plants, the impact of current ETS prices at €70 $t_{CO_{2e}}^{-1}$ lead to added levelised costs for ordinary Portland cement production of €59 t_{cement}^{-1} in the model (excluding free allocation). Thus, current ETS prices may not incentivise cement plants to implement FOAK installations. This presents a timing mismatch between decarbonisation costs and credits, suggesting alternative support may be needed to kick-start cement's decarbonisation path. Additionally, the results show that even when multiple plants are built and maturity for interventions at

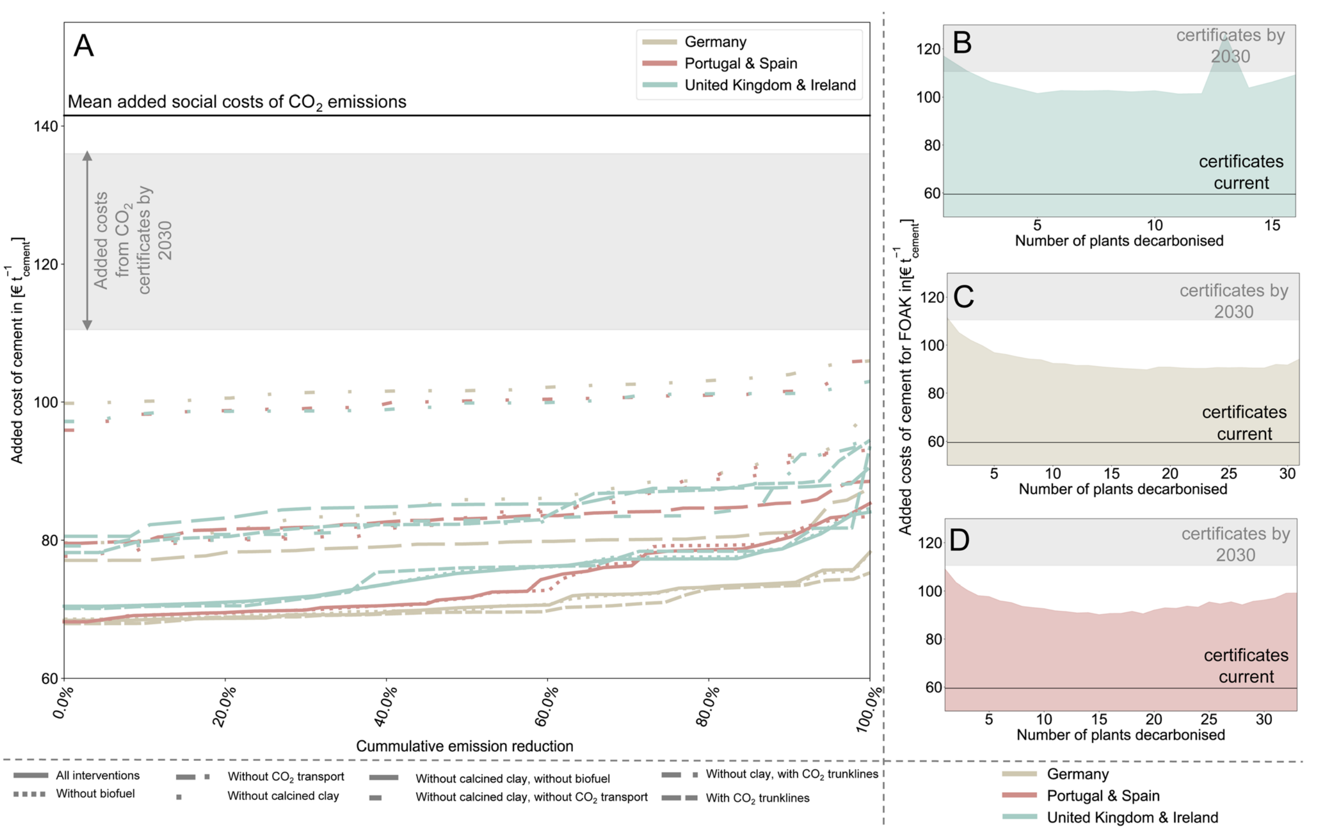

Fig. 9 (A) Comparison of results from different scenarios compared to added costs of cement production by CO_{2e} certificates (2030)[78] and social costs of CO_{2e} emissions,[79] with current emissions for clinker 850 $kg_{CO_{2e}}$ $t_{clinker}^{-1}$ [35] for ordinary Portland cement and no free-allocation of allowances, assuming mature technologies. Results for first-of-a-kind (FOAK) installations for (B) United Kingdom and Ireland, (C) Germany and (D) Portugal and Spain compared to added costs of cement production by CO_{2e} certificates in 2030 and current (2024). Note, FOAK installations were calculated using the interventions selected assuming technology maturity presented in Fig. 4. These strategies do not have to be least-cost for FOAK deployment.

the cement plant is reached, DAC costs remain high as the cement industry cannot bring down the costs alone (*e.g.*, decarbonising the German cement industry led to a cumulative DAC capacity of 8 Mt_{CO_2}).

First investments must be CO_2 infrastructure. Direct air capture plants will be built last

After determining which mix of interventions would be cost-optimal endpoint for fully decarbonised cement industry, we used a back-casting approach to determine how to reach full decarbonisation in the most economically viable way (*i.e.*, which investment is most economically viable to do first). The results show that first interventions at the cement plants themselves (*i.e.*, mostly CCS and calcined clay) are implemented before direct air capture technologies are built (Fig. 10). For reaching 50% emission reduction in all regions only partial CO_2 transport networks need to be built (but already sized to later accommodate the CO_2 volumes at the endpoint). Plants farther away from plant agglomerations (*e.g.*, those in Eastern Spain, East and Southeast Germany) are not connected to the pipeline network due to lack of synergies (*e.g.*, connecting a single plant further away from the storage site increases costs, compared to connecting multiple ones close by). Reaching 50% emissions reduction comes with an added levelised cost of cement production of €46 t_{cement}^{-1} (UK&IRE), €37 t_{cement}^{-1} (GER) and €43 t_{cement}^{-1} (PT&ES), accounting for 52% to 61% of the costs for full decarbonisation, again underlining the need for high upfront investments in times that ETS prices are at lower levels still. Reaching 75% emission reduction was achieved by connecting all plants to the CO_2 transport and storage network and only partially implementing direct air capture accounting for added levelised cost of cement production of €40 t_{cement}^{-1} to €50 t_{cement}^{-1}. Achieving 75% emission reduction only accounts for 56–68% of the total costs, compared to the 58–61% for 50% emission reduction, as most of the CO_2 transport infrastructure at this point has already been constructed (and is already included in the levelised costs of the 50% emission reduction scenario). The last 25% to reach full decarbonisation is then entirely achieved by implementing direct air capture plants and establishing additional storage sites (*i.e.*, CO_2 injection wells). Note, direct air capture technologies were also added for lower emission targets, but mostly as the cheapest option to reach residual emission reductions to a set target (*e.g.*, from 47 to 50%).

The obtained results are robust towards changes in feedstock and energy assumptions

As the outputs of ex ante modelling study, the results presented here contain significant uncertainties (*i.e.*, all technologies assessed here have not yet reached maturity requiring the use of assumptions and future costs of feedstocks and energy cannot be known at this time). To verify the robustness of the generated results, we performed an uncertainty analysis in the form of a one-at-a-time sensitivity analysis which we showed earlier to be a sufficient method for a computational expensive model that does not allow global uncertainty analysis.[80] We specified scenarios for feedstock prices with minimum and maximum estimates (*i.e.*, prices for biofuel, prices for olivine-bearing rocks, prices for kaolinite clays as well as prices for energy, Table S5†). The results suggest that the model is robust to changes in assumptions, as mostly the same strategies are chosen as for the base case (Tables S27–S32†). When all interventions are included, CCS coupled with calcined clay cements is chosen for almost all cement plants. However, high kaolinite clay prices lead to a divergence from this strategy (such as changing to CCS alone, CCS with biofuel or CCS with CO_2 mineralisation) (Tables S30 and S31†). Notably, when strategies are chosen using CCS or CO_2 mineralisation in the base case, the model's outcomes become much more sensitive to energy prices, as these measures use considerable amounts of electricity and heat to compress CO_2, facilitate reactions or separate products[36,81,82](Fig. 11). For calcined clay interventions, which use less energy than conventional cement, high energy costs therefore have a smaller and sometimes even negative impact (Table S13† and Fig. 11).

Discussions

The here presented analysis of decarbonising the cement industry across three European case study regions (*i.e.*, UK&IRE, GER, and PT&ES) showed multiple strategies toward achieving net-zero CO_{2e} emissions. The results highlighted the essential role of CCS and DACCS technologies alongside novel cement replacements such as calcined clay cements as the most cost-effective strategies. However, we here want to also highlight the complexities in *ex ante* modelling with significant uncertainties due to simplifications and unknowns and put the major findings in context.

CO_2 transport infrastructure planning

The results show that elaborate CO_2 transport networks as well as their swift implementation will be necessary to reach full decarbonisation in an economically viable way. The absence of such infrastructure could significantly increase the costs of achieving decarbonisation, emphasising the necessity for coordinated efforts and investments in transport networks shared among industries, regions and countries, which will require further efforts from industry and policy (*e.g.*, adapting legislation for transnational CO_2 transport such as the ratification of the London Protocol[83]). Arguably the CO_2 networks described here are unlikely to be realised in the presented form as many simplifications had to be made in the model (*e.g.*, only cement plants are included as CO_2 emitters, CO_2 pipelines only connect two grid cells following a straight line between cement plants, terminals etcetera, no exclusion zones are implemented). But the results show that collaborative planning for CO_2 transport networks with other cement plants and other industries intending to adopt CCS is imperative.

While others have modelled CO_2 transport networks for European industries in varying resolutions,[84–87] commonly based on emission datasets assuming a certain share of CO_2 capture at each plant level,[87] we here considered the cement

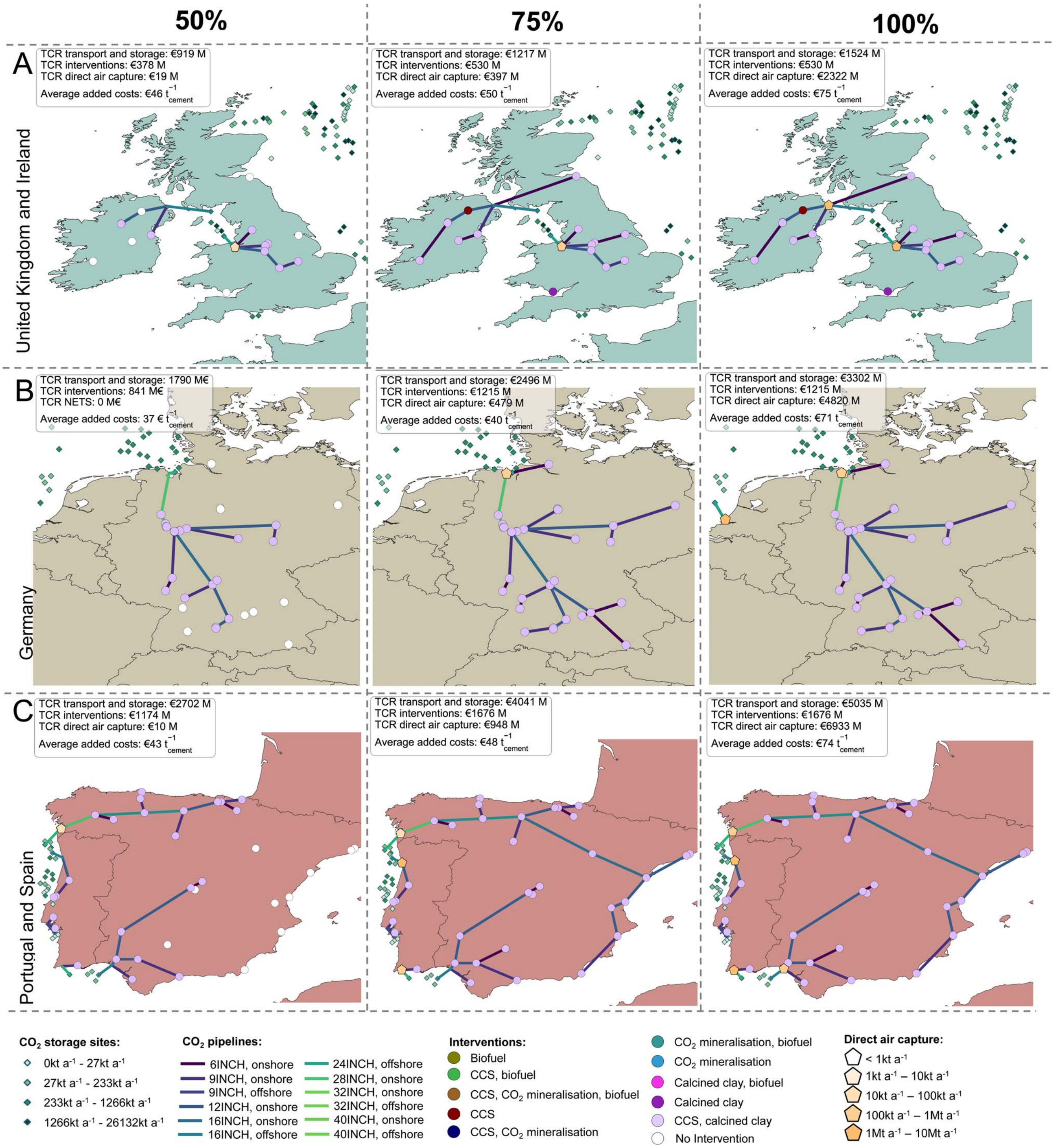

Fig. 10 Model results for reaching different decarbonisation goals (*i.e.*, 50%, 75% and 100%) in (A) United Kingdom and Ireland, (B) Germany and (C) Portugal and Spain. Each circle represents one integrated cement plant or direct air capture location. Circle colours represent interventions. Line colours indicate CO_2 transport pipe thickness. White circles are unabated cement plants further up the marginal cost curves. Total capital requirements (TCR) describe the capital expenditures (CAPEX) of an investment.

industry as a whole while including a range of different interventions (*i.e.*, biofuels, calcined clay cements) alongside CCS alone. This approach should be used in further research to model the interaction between decarbonisation strategies of multiple industries (*e.g.*, including the steel industry with their options for decarbonisation).

We showed that the pathway, in particular timing of the implementation of an intervention, towards decarbonisation will depend on the location. This could give some companies a competitive advantage over others. *E.g.*, a company owning multiple cement plants could leverage CO_2 certificates among their plants (which is allowed in the EU ETS[77]), while other

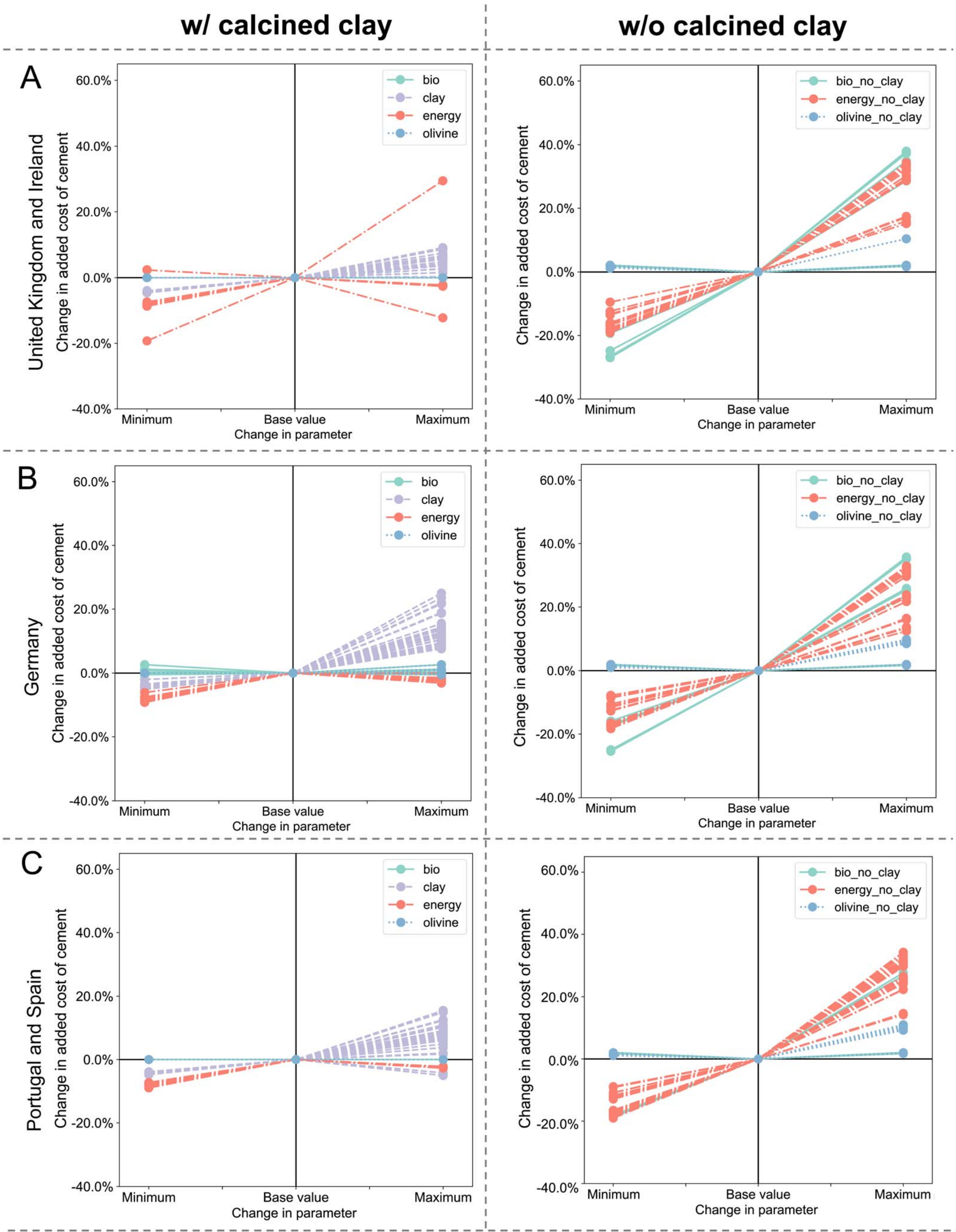

Fig. 11 Results of one-at-a-time sensitivity analysis. Each point represents the change of one cement plant in (A) United Kingdom and Ireland, (B) Germany and (C) Portugal and Spain allowing all interventions (w/calcined clay) and allowing all interventions except the ones containing calcined clay (w/o calcined clay). Ranges for parameters shown in Table S5.† Note, due to the nature of the mixed integer linear programming model and finding quasi-optimal solutions, the runs can never fully be repeated, leading to small variations as "noise" in the sensitivity analysis.

companies only owning few plants, will have to wait, *e.g.*, until CO_2 infrastructure is in place while being exposed to fluctuations in CO_2 certificate prices. This might require actions from legislature or collaboration between competing companies to level market opportunities.

Additionally, our results highlight the importance of the selection of CO_2 storage sites. Notably, all existing studies modelling CO_2 transport and storage networks in Europe use different assumptions for determining suitable CO_2 storage sites,[84–87] while some include onshore storage and others do not. We excluded onshore CO_2 storage due to higher risk perceptions by laypeople[46] and used data from European Comission – Joint Research Centre[27] to determine CO_2 storage sites which were published a decade ago and to this day provides the most comprehensive survey of potential storage sites within Europe, but still required a significant amount of assumptions to be made (especially for Germany where storage locations and sizes were not fully investigated) (Note S6†). This highlights that suitable storage sites both from a technological as well as societal perspective must be determined to allow modellers, industry and policy makers to plan reliable strategies towards net-zero CO_{2e} emissions. This is of particular importance for some countries such as Germany were the law must first be changed to allow CO_2 storage,[88] while in other countries the debate on CO_2 storage is only beginning to develop now.

Calcined clay cements

Our results underscore the potential of calcined clay cements in substantially reducing decarbonisation costs. These cements, due to their low heat requirements when coupled with traditional cement production with CCS, present a promising avenue, albeit contingent on market acceptance and availability of local resources. Despite the great economic potential of calcined clay cements shown in this study, current prices for kaolinite clays are significantly above the prices necessary to be economically viable (*i.e.*, in 2022 in the United States the average price for kaolinite clays sold was \$160 t_{clay}^{-1} (€150 t_{clay}^{-1}),[89] while we here assume a price of €30 t_{clay}^{-1} in the base case scenario (Table S5†)). Current markets for kaolinite clays include manufacturing ceramics, porcelain ware and floor or wall tiles due to its whitening properties[90] leading to a high cost and small volume markets (*e.g.*, in 2021 in Germany 864kt_{clay} and in the United Kingdom 735 kt_{clay} were produced[91] while 35 Mt_{cement} and 9 Mt_{cement} were produced respectively[92]). But clays for the cement industry might not have to exhibit these whitening properties and could be of lower grades. Nevertheless, clay reserves for lower grade clays in Europe outside of existing quarries may be less accessible,[20] where further investigations are needed (*i.e.*, for locations, capacities, extraction costs, clay compositions). Beyond the sourcing of raw materials, further research for the product applications is needed. The use of calcined clay in cement mixtures might lead to challenges due to high surface area alongside high water demands as well as colour control[20] which will have to be fully addressed before widespread market implementation, all the more reason to start investigating them now.

CO_2 mineralisation and biofuels

The absence of calcined clay cements (*e.g.*, due to lack of market adoption or lack of affordable feedstocks) could necessitate alternative strategies, such as CO_2 mineralisation or biofuels, particularly influenced by local resource availability and transportation costs, highlighting that flexibility in technological choices might become key in the regional context.

For CO_2 mineralisation, our results show that these strategies were chosen contingent to a plant's access to cheap transport or proximity to feedstock deposits. We solely considered olivine-bearing rocks as feedstocks used in a direct aqueous carbonation process, which we further developed to produce SCM for use in cement blends[36] (Note S3†). While we chose this process due to its lowest levelised costs of production compared to other processes[36] and the feedstock due to the large availability, there might be other processes and feedstocks to be considered. *E.g.*, serpentine-bearing rocks which might be available *e.g.*, in northern Europe as well as south England and south Spain,[26,41] albeit with higher energetic penalties for the carbonation reaction due to necessary heat activation.[33] Our results indicate that the main disadvantage of the considered CO_2 mineralisation process is in addition to limitations of feedstock transport, its comparably high energy demands (the process requires grinding to particle sizes under 10 μm, increased pressure of 100 bar and 190 °C to facilitate mineralisation).[36,93] Hence, other feedstocks like calcium oxide-rich industrial wastes (*e.g.*, steel slag, red mud, concrete and demolition waste) might be feasible as they could carbonate with lower energy penalties (*i.e.*, at lower temperatures, quicker residence times). But these calcium oxide-rich feedstocks are limited both by geographic availability of other industries (*i.e.*, steel plants[41]) and volume (*e.g.*, Germany produces 5.4 $Mt_{steel\ slag}$ a^{-1}[94] while producing 35 Mt_{cement} a^{-1}[92]). Thus, further geospatial studies will be necessary beyond the question of feedstocks for CO_2 mineralisation. The results show that CO_2 utilisation concepts could become a part of decarbonisation for some locations, particularly in combination with CCS. When CO_2 mineralisation concepts were chosen, it was due to their potential to permanently store CO_2. Other CO_2 utilisation options (*e.g.*, production of fuels), would not have been chosen by the solver as costly direct air capture plants would have to be built to capture CO_2 emissions created from the combustion at the end of their life cycles.

Similarly to CO_2 mineralisation, biofuels were predominately selected for plants with access to cheap ship transport (*i.e.*, Northern Germany) (Fig. 7). Although combining CCS with biofuels showed the highest emission reductions of all combinations through creating carbon dioxide removal from embodied biogenic CO_2 (excluding transport 55 $kg_{CO_{2e}}$ t_{cement}^{-1} to 77 $kg_{CO_{2e}}$ t_{cement}^{-1}, Fig. S8†), transport emissions (up to 327 $kg_{CO_{2e}}$ $t_{biofuel}^{-1}$ for south Germany, Table 2) and costs diminished the potential of using biofuels in many locations. Because none of the case study regions produces biofuels (*i.e.*, wood pellets) in sufficient amounts for the energy demands of its cement industry (*e.g.*, Germany, the Europe 3rd biggest producer of hardwood pellets could only satisfy 34% of its cement industries energy demand with hardwood pellets, Table S1†) we here only considered biofuels from North America, which has been suggested as a future potential source for biofuels in Europe.[95] Even including significant transport emissions from North America some plants (in particular in the United Kingdom and Portugal & Spain) still chose biofuels as a strategy. This indicates that in some regions with high biofuel production (*e.g.*, black forest in south Germany) the use of biofuels could become more attractive than shown here, contingent on low production costs to which the use of biofuel showed high sensitivity (Fig. 11). This should be addressed by future research. As biofuel production is limited in most countries (Table S1†) and might compete with other uses (*e.g.*, material use or food resources), many countries might define strategies for biofuel use in their sustainability roadmaps. For example, the German government has already announced that their national biomass strategy (yet to be published) will likely prioritise material use for biomass over energy use[96] and hence might limit the use of biofuels in the cement industry.

Direct air capture and carbon dioxide removal

Our results highlight the impact of direct air capture (as a model carbon dioxide removal technology) in deep decarbonisation strategies for the cement industry. We showed that if cement plants do not use biofuels, a significant share of their costs for deep decarbonisation will instead go to direct air capture and storage. While we showed that these strategies will be implemented last, due to their comparatively high costs (Fig. 10), in current industry roadmaps they play a minor role.[5,10]

Investment costs and first-of-a kind plants

While the cumulative investments calculated in our model appear substantial, the projected costs must be contextualised against potential future expenses incurred through greenhouse gas emission certificates. We acknowledge that all interventions have been considered with expected costs at technology maturity (*N*th-of-a-kind) and first-of-a-kind plant costs will thus first have to be overcome, which will necessitate support from governments or environmentally conscious customers (*e.g.*, green construction companies) as the ETS certificate market is not designed to reward companies carrying additional costs for first-of-a-kind investments.[97] The here presented costs reflect estimated long-term costs and thus are lower than recently presented studies[5] which estimate the cost based on current developments rather than at technology maturity.

Limitations

Inherently, the here developed model has limitations due to its ex ante nature (*i.e.*, accessing technologies which are not fully developed yet) as well as simplifications necessary to overcome computational limitations. First, we only considered one type of cement (ordinary Portland cement with simplified clinker factor of 1), while a multitude of different cement types are currently produced,[98] meaning this study neglected the use of clinker replacement in different cement types. But because all cement types contain ordinary Portland cement, most of the here derived conclusions are likely to hold true for other cement blends too. Similarly, we needed to limit the number of interventions studied due to computational constraints, while including interventions of different types (Fig. 1). While we acknowledge that other interventions may play an important role in decarbonisation for some cement plants or niche applications (such as alkali activated systems,[8] magnesia cements,[8] or CO_2 concrete curing in precast concrete production[20]), many of the learnings from this paper are transferable, as these concepts either aim to reduce the clinker content, store CO_2 in cementitious products or capture CO_2 from the atmosphere.

A further limitation of the here shown results lies in the availability of feedstocks. We did not consider capacities of potential feedstock sites for mineralisation, biofuels, or clays. While estimations suggest that potential capacities might be sufficient for widespread implementation of these concepts[24,33,41] detailed supply–demand matching would have to be done. Additionally, especially in the case of biofuels we did not consider environmental changes due to its harvest. The harvest of wood for wood pellet production can have significant impacts on the forest carbon and the environmental impact of biomass production is influenced by the tree's age when it is harvested, with break-even periods (until forest carbon is replaced) ranging up to multiple decades.[99,100] Thus, further research should be conducted to investigate which wood sources might be suitable for biofuel production in the cement industry and detailed life cycle assessments should be conducted.

A major challenge currently lies in the uncertainty of the decarbonisation strategies of other industries. Integrated assessment modelling should be conducted including other energy-intensive industries in particular as they also might use similar feedstocks for their decarbonisation strategies and consequently impact regional demand and costs (*e.g.*, other energy intensive industries might also consider biofuels).

Conclusions

For the first time, we presented a quantitative, spatially explicit analysis of cost-optimal strategies to transition the European cement industry towards net-zero CO_{2e} emissions, focusing on three regions in Europe. Our results demonstrated the feasibility and economic viability of achieving net-zero emissions through strategic interventions and technological advancements (*i.e.*, iterative learning), underpinning the notion that full decarbonisation of cement production is possible and economically sensible. However, our analysis underlined that these efforts require significant (upfront) investments that may need incentivising beyond CO_2 credits or taxes, collaboration, and rational sequencing of interventions. The main conclusions for policy and decision makers can be summarised as follows.

Conclusions for policy and decision makers

• Prioritise investments in carbon capture, transport, and storage technologies as they play a vital role in the industry's decarbonisation strategy. Especially CO_2 storage sites should be selected in conjunction with CO_2 transport infrastructure and should be developed swiftly.

• Collaboration and coordination between cement producers and with other industries to plan and implement CO_2 transport networks is essential for efficient and cost-effective decarbonisation. Moving alone will lead to substantially increased total costs of decarbonisation (added costs of up to €19 billion annually could be expected for the European Union and United Kingdom combined).

• Cement producers and governments must expedite the research into, and use of, calcined clay cements as a sustainable alternative, investigating local resource availability and market acceptance. Falling to include calcined clays may increase costs for deep decarbonisation by 12–21% or €9 to €15 t_{cement}^{-1}.

• Small cement producers or individual plant owners must explore options for synergies with other plants in their vicinity and develop strategies for emission reduction short to medium term (*e.g.*, through use of biofuels, alternative clinkers) until CO_2 transport infrastructure is built.

• Adapt decarbonisation strategies based on regional resources, transportation costs, and infrastructure availability (*i.e.*, implementing CO_2 mineralisation when CO_2 transport infrastructure is yet to be built).

• Facilitate initial investments in new technologies through subsidies, incentives, or joint investments as high costs for first-of-a-kind technologies must be overcome for favourable economics of deep decarbonisation.

• Develop location-dependent decarbonisation strategies, which should only include technologies that can still be used when full decarbonisation is reached to avoid malinvestments (especially important for CO_2 utilisation technologies without long-term CO_2 storage).

• Include carbon dioxide removal strategies (using DACCS or biofuel CCS) into decarbonisation strategies for the cement industry. It is virtually impossible to reach net-zero CO_{2e} cement production without this.

In summary, achieving net-zero CO_{2e} emissions in the cement industry is complex but feasible. It requires a multi-faceted approach involving technological innovation, strategic planning, and collaborative efforts across cement producers and other industries. The long-term economic benefits make a compelling case for industry and policy makers to commit to a swift transition in this sector.

Data availability

Code generated during this study has been published as Strunge[101] and can be accessed under: **https://doi.org/10.5281/zenodo.13737709**. The release also contains datasets of the all the results produced.

Author contributions

TS: conceptualisation, methodology, formal analysis, and writing—original draft. LK: methodology. NS: methodology of INDiECAR-RTN. NSh: conceptualisation and writing—review. PR: conceptualisation and writing—review. MV: conceptualisation, methodology, and writing—review.

Conflicts of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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