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Net-zero transition of the global chemical industry with CO₂-feedstock by 2050: feasible yet challenging†

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Carbon capture, utilization and storage (CCUS) have been projected by the power and industrial sectors to play a vital role towards net-zero greenhouse gas emissions. In this study, we aim to explore the feasibility of a global chemical industry that fully relies on CO2 as its carbon source in 2050. We project the global annual CO₂ demand as chemical feedstock to be 2.2-3.1 gigatonnes (Gt), well within the possible range of supply (5.2-13.9 Gt) from the power, cement, steel, and kraft pulp sectors. Hence, feedstock availability is not a constraint factor for the transition towards a fully CO2-based chemical industry on the global basis, with the exception of few regions that could face local supply shortages, such as the Middle East. We further conduct life cycle assessment to examine the environmental benefits on climate change and the trade-offs of particulate matter-related health impacts induced by carbon capture. We conclude that CO₂ captured from solid biomass-fired power plants and kraft pulp mills in Europe would have the least environmental and health impacts, and that India and China should prioritize low-impact regional electricity supply before a large-scale deployment of CCUS. Finally, two bottom-up case studies of China and the Middle East illustrate how the total regional environmental and health impacts from carbon capture can be minimized by optimizing its supply sources and transport, requiring cross-sectoral cooperation and early planning of infrastructure. Overall, capture and utilization of unabatable industrial waste CO2 as chemical feedstock can be a feasible way for the net-zero transition of the industry, while concerted efforts are yet needed to build up the carbon-capture-and-utilization value chain around the world.

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1. Introduction

While the chemical industry is an enabler for achieving the Sustainable Development Goals, ^{1,2} it is also the third-largest greenhouse gas (GHG) emitting sector and the largest consumer of fossil fuels among industrial sectors. ³ In 2019, in addition to 19.5 exajoule (EJ) of process energy consumption, the chemical industry was also responsible for 27.9 EJ of energy consumption in the form of feedstocks. ⁴ Thus, switching to renewable process energy alone is not sufficient to reach net-zero GHG emissions of the chemical industry. Replacing

fossil feedstocks with renewable or recycled ones is also essential for further minimizing the industry's GHG emissions and for its transition towards a circular economy.⁵

In recent years, carbon dioxide (CO₂)—the major GHG has been proposed as a candidate feedstock that may enable the aforementioned transition of the chemical industry. In particular, various carbon-utilization technologies have been and are being developed to produce commodity chemicals from CO₂, including through reduction and carboxylation reactions.6,7 A detailed analysis of potential CO2-based synthesis routes towards bulk and fine chemicals can be found in the work by Otto et al.8 Recent studies have increasingly focused on developing novel and cost-effective catalysts that enable high selectivity and conversion rates of desired products under mild reaction conditions. 6,9-14 In 2020, China launched a demonstration project that aims to produce 1000 tonnes of CO₂-derived methanol annually. 15 As such, CO₂ utilization as a renewable feedstock seemingly creates an ideal win-win situation that simultaneously combats global warming and enables a more sustainable chemical industry. However, to understand the feasibility of the concept, factors

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such as the regional supply security of CO2 need to be assessed against current and foreseeable mega-trends, which may sometimes work in an antagonistic manner.

34 gigatonnes (Gt) of CO₂ were emitted globally from fossil fuels in 2019, 60% of which came from point sources of the power generation and industrial sectors where many emissions are hard to abate. 16 Carbon capture has been projected to play a vital role in tackling these large-scale point emissions in most of the pathways towards limiting the temperature increase within 1.5 °C underlined by the Intergovernmental Panel on Climate Change (IPCC). 17 It has also been adopted as part of the long-term low emissions and development strategies by 83% of the national submissions under the Paris agreement. 18 Therefore, while the global capacity of carbon capture in operation is less than 40 megatonnes (Mt) by the end of 2021, 18 an accelerated deployment of carbon-capture projects may be expected, resulting in increases in CO₂ supply for industrial use.

Meanwhile, to avoid catastrophic consequences of global warming, industrial sectors are developing roadmaps towards net-zero GHG emissions by 2050, including using renewable energy sources, improving efficiency, and switching to innovative low-carbon production routes. 19-23 These trends imply that overall industrial CO2 emissions may drastically decrease in the future, resulting in reduced CO2 availability for carbon capture. In addition, there are other mechanisms for captured CO2, e.g., direct storage in geological reservoirs (known as carbon capture and storage, or CCS),24 and production of synthetic fuels.^{25,26} Each mechanism has its own advantages and disadvantages. For example, CCS could contribute to the longterm removal of CO₂ from the atmosphere, 27 but the regional storage capacity might remain a challenge, especially in India and China.²⁸ Public concerns, including safety considerations, are another obstacle yet to be addressed.²⁹ The deployment of these mechanisms could determine the local supply of captured CO2 for the chemical industry. To our knowledge, no detailed studies have addressed the future dynamics of regional CO2 supply-demand balance.

Furthermore, CO₂ capture is not free from environmental impacts. Chemical absorption with monoethanolamine (MEA), the currently most mature carbon-capture technology, is known for its high energy penalty, especially due to reboiler duty for solvent regeneration.^{24,30} Life cycle assessment (LCA) is a key method for evaluating the environmental benefits and trade-offs of carbon capture. Several LCA studies have been conducted for carbon capture in individual regions (e.g., Europe) and sectors (e.g., coal-fired power plants). 31-33 However, the environmental impacts of carbon capture depend on the regions and sectors from which CO2 is captured, as well as electricity and fuel mixes used to perform carbon capture, highlighting the need for cross-regional and cross-sectoral assessments. Von der Assen et al.34 have benchmarked the environmental impacts of carbon capture from different industrial sectors in Europe using LCA, including global warming impacts and fossil resources depletion. They concluded that CO₂ should first be captured from the sites

where CO₂ concentration in the flue gas is close to 100%, followed by paper mills and coal power plants. A follow-up study further pointed out that GHG savings can be significantly improved if the carbon-capture process uses renewable energy instead of the current electricity grid mix in Europe.35 However, both studies did not consider future global dynamics of the CO₂ supply-demand relationships, regions outside Europe, and other impacts such as particulate matter (PM)related health impacts, which are largely driven by fossilenergy use.36,37

Against these knowledge gaps, we endeavor in this study to provide insights into the strategic planning of future sustainable transitions of the global chemical industry, with a particular focus on exploring the supply-demand feasibility of a chemical industry in 2050 that fully relies on CO2 as its carbon source. We acknowledge that other potential net-zero transition pathways also exist for the chemical industry, e.g., through biomass valorization; these pathways will be assessed in our subsequent studies. Here, we start with the assessment of the future global and regional supply-demand balance of CO2, and then use LCA to examine the regional and sectoral savings on climate change impacts and trade-offs of PMrelated health impacts for capturing 1 kilogram (kg) of CO2. Based on the LCA results, we explore a regionalized sourcing strategy of CO2-feedstock, which is demonstrated by two bottom-up case studies, China and the Middle East. We further discuss our findings and their implications for future research and climate mitigation measures.

Methods

The study consists of three parts (see Fig. 1): (I) regional CO₂ supply-demand balance in 2050, (II) regionalized environmental and health impacts of CO2 capture from different sectors, and (III) case studies of regionalized CO2 sourcing strategies.

Global and regional supply-demand balance of CO₂

The potential regional CO₂ supply from the power and industrial sectors, and its demand as chemical feedstock in 2050 were projected under two CO2-emission scenarios that were intended to cover the possible range of uncertainties.38 The high-emission scenario was set according to the conservative scenarios defined by the International Energy Agency (IEA), such as the Stated Policies Scenario. The low-emission scenario was assumed to be the ambitious scenario that is compatible with IEA's Net Zero Emissions by 2050 Scenario, or equivalent. The detailed scenario settings of each sector are described in sections S1.1 and S1.2 in ESI.†

The following nine countries/regions of focus were chosen as they are the world's current largest CO2 emitters, the largest primary chemical producers, and/or under rapid development: China (CN), the United States of America (US), India (IN), the European Union (EU), Japan (JP), the Republic of Korea (KR), the Russian Federation (RU), Region Middle East (RME), and

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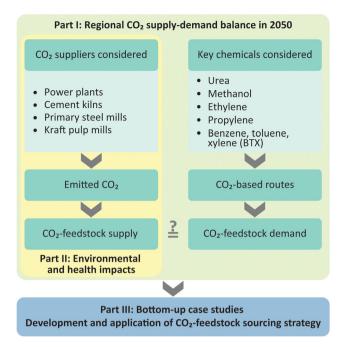


Fig. 1 Study overview.

Region Africa (RAF) (see Table S2 in ESI† for the region definitions). In 2019, these regions together account for 75% of the global CO₂ emissions¹⁶ and 90% of the global production of the seven base chemicals that are the current key building blocks for the bulk of the chemical industry (Table S7 in ESI†): ammonia, methanol, ethylene, propylene, and benzene, toluene and xylenes (BTX aromatics).3 All other countries were grouped together as "Rest of the World" (RoW).

2.1.1 Estimating CO₂ supply from the power and industrial sectors. For CO₂ supply, the power, steel, and cement sectors were selected, as they are the current largest fossil CO2 emission sources.³⁹ Moreover, the kraft pulp sector was included, which is the largest biogenic CO2 source according to a Europe-focused study. 40 In particular, large stationary sources that emit more than 0.1 Mt CO2 per year were considered. These include power plants (coal, natural gas, and solid biomass-fueled), cement kilns, primary steel mills, and kraft pulp mills. The primary steel mills include the blast furnacebasic oxygen furnace (BF-BOF) route, the direct reduced ironelectric arc furnace (DRI-EAF) route, and the smelting reduction-basic oxygen furnace (SR-BOF) route that is to be commercialized under the low-emission scenario in 2050.²¹

The sectoral CO₂ emissions were quantified by multiplying its projected product or energy output with the regionalized specific carbon emission intensity (which was determined by the production routes, type of fuels used, and energy efficiency). The production routes and type of fuels used are sector- and region-specific, with detailed descriptions in section S1.1 in ESI.† For energy efficiency, a global average of 30% improvement from industrial sectors was considered possible according to the United Nations Economic Commission for Europe. 41 For the high-emission scenario in

2050, we assumed that half of this energy-reduction potential would be realized, i.e., 15% improvement in energy efficiency in comparison to the 2019 baseline, as long as the improvement does not exceed the best-available technology (BAT) level identified in the literature. 22,42-44 For the low-emission scenario, we assumed that all manufacturing facilities would be operating at the energy efficiency of their BAT level.

For CO₂ capture, a capturing rate of 90% from flue gas was assumed as the technical limit for both scenarios, as it is the common industrial practice. 45-47 For power plants, cement kilns, and kraft pulp mills, capturing CO2 from all stacks was assumed; hence, the maximum CO2 capture capacity from these sources would be 90% of the total emissions.

In integrated steel mills, due to land constraints and economic factors, it would be difficult to capture CO2 from every stack. Thus, for BF-BOF steel mills, it was assumed that CO2 would only be captured at four types of stacks with the largest CO₂ emission rates (the blast furnace hot stove, on-site steam generation plant, coke oven, and lime kiln), representing 72% of their total CO₂ emissions.⁴⁷ Combined with the 90% capture limit from flue gas, a maximum of 65% of the total CO₂ emissions could be captured. For other types of steel mills, capturing 80% of their total emissions was assumed. 48

2.1.2 Estimating CO₂-feedstock demand from the chemical industry. In terms of demand, a future CO2-based chemical industry was assumed to be still based on the seven base chemicals as key building blocks. These base chemicals were assumed to be the key CO2 consumers, with the exception of ammonia. While ammonia does not contain carbon in its structure, urea-accounting for 55% of the downstream use of ammonia—is a direct CO2 consumer.

The potential CO₂-feedstock demand for a given chemical was calculated by multiplying the projected production volume of the chemical (in Mt per year) with the specific CO₂ demand in a given CO2-based production route (kg CO2 per kg chemical). The production routes and their corresponding specific CO₂ demand were assumed to be the same in both high- and low-emission scenarios. The global and regional production volumes of the seven key chemicals in 2050 under the two scenarios were projected using the growth rates estimated by IEA (Tables S33 and S34 in ESI†).3 The CO2-based production routes considered either are already commercialized (urea, ethylene, and propylene), or have a high technology readiness level (TRL) (methanol and BTX aromatics). 5,49 The specific CO2 demand was calculated through the stoichiometric balance of each production route, taking into account its corresponding conversion rate. Detailed descriptions of the CO2-based production routes for the seven key base chemicals can be found in section S1.2.2 in ESI.†

2.2 Environmental and health impacts of carbon capture

We assumed that 30 weight-% aqueous MEA would be applied to capture CO2 from all the sectors, as it is currently the most mature technology with successful retrofitting applications in various industries.²⁴ The captured CO₂ was assumed to be compressed to 110 bar as a general industrial practice for

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transportation.³³ The assessed impacts include climate change impacts, calculated with the global warming potential over 100 years (GWP100),⁵⁰ and PM-related health impacts.^{31,34} The selection of these two impacts is because they are among the most relevant impacts for combustion of fossil fuels, the main source of environmental and health impacts with regard to carbon capture.

LCA was then performed to quantify the impacts of CO₂ capture from different sectors. The functional unit was defined as 1 kg of captured CO2 compressed to 110 bar at the gate of carbon-capture facilities. Transportation was not considered in the global analysis, but its influence was evaluated in the detailed case studies below. Other utilization mechanisms of the captured CO2 were excluded in this study, as the focus is on assessing the availability of CO2 as feedstock and compare the impacts of different sources of CO2 with each other.

The impacts of capturing 1 kg CO2 were calculated as the difference of the impacts between each reference system (e.g., power plants, cement kilns, steel mills, and kraft pulp mills) with and without carbon capture (the only changing variable). MEA make-up is around 1-2 gram per kg CO₂ captured, and according to the previous studies, its climate change and other environmental impacts are generally negligible compared to the energy use for carbon capture. 31,33,51,52 Therefore, the impacts of MEA were not estimated in this study.

Energy use plays an important role in determining the environmental and health impacts of carbon capture. Capturing 1 kg CO₂ typically requires 3.6-4.0 megajoule (MJ) of low-pressure steam for MEA regeneration,24 in addition to around 0.17 kilowatt-hours (kW h) of electricity for pumping and CO₂ compression (Fig. S1 in ESI†).³³

More specifically, in the power plants and kraft pulp mills, steam and electricity supply for carbon capture were assumed to be extracted directly from the power generation units within the system boundary, without additional external energy supply. However, as net energy exporters, their net electricity output would be less. Due to the complexity to identify marginal electricity suppliers acknowledged by the previous studies, 34,53 in this study, additional electricity from the regional electricity grid was assumed to be required to compensate for this power loss, contributing to the environmental and health impacts of carbon capture.

For cement kilns and steel mills, waste heat was assumed to be available for utilization by the carbon-capture process. Therefore, the steam for MEA regeneration was supplied with a waste-heat boiler wherever possible, prior to using the same additional fuel mix as for the main production process (e.g., 100% coal in BF-BOF steel mills). As part of the sensitivity analysis, other scenarios were investigated where the steam was supplied with another natural gas boiler or using electrode vessels. The electricity needed for carbon capture was assumed to be supplied by the regional grid mix in 2019. Scenarios of the regional grid mix of electricity supply in 2050 based on IEA's World Energy $Outlook^{54}$ were also investigated as part of the sensitivity analysis. Detailed settings of the additional

energy requirement for carbon capture in different systems are provided in Table S37 in ESI.†

The "allocation, cut-off by classification" system model of ecoinvent 3.8 was used to calculate climate change impacts of regional electricity production from wind, hydro, geothermal, and nuclear power plants, as well as background processes of regional market fuel supply (coal, oil, natural gas, and biomass). Climate change and PM-related health impacts of per unit fuel combustion in the foreground processes of electricity and steam production were derived from the IPCC report⁵⁰ and Oberschelp et al., 36,39 respectively. Detailed descriptions of the calculation of impact factors can be found in sections S1.3.2 and S1.3.3 in ESI,† while a discussion of the inherent uncertainties of the approach is provided by Oberschelp et al. 36

2.3 Bottom-up case studies: development and application of CO₂-feedstock sourcing strategies in China and the Middle

China and the Middle East under the low-emission scenario in 2050 were selected as case studies to explore regionalized CO₂ sourcing strategies. They are the two largest producers of the base chemicals globally (Table S31 in ESI†). China is representative of regions with sufficient CO2 supply to their chemical manufacturing sites, whereas the Middle East represents places with insufficient in-region supply of CO₂-feedstock.

The geographical coordinates of existing large CO₂ point sources (i.e., fossil-fueled power plants, cement kilns, and primary steel mills), and chemical manufacturing sites of the seven key chemicals and their respective production capacities were collected. Pulp mills (estimated to be less than 2% of the total CO₂ supply in both regions in 2050) were excluded from the case studies due to data gaps. SR-BOF steel mills and biomass-fired power plants were also excluded, as their locations in 2050 remain unknown, because these technologies were assumed to grow in the coming decades. However, as biomass-fired power plants would be a major CO₂ supplier in China in 2050, a scenario that considers solid biomass-fired power plants as CO2 suppliers was investigated in the sensitivity analysis. To this end, to-be-eliminated coal-fired power plants, as described below, were assumed to be retrofitted into solid biomass-fired power plants to reduce stranded assets.⁵⁵

The projected regional CO₂ demand by the chemical industry and supply by the different sectors were allocated to all the existing sites based on their current production capacities, except for the coal power plants in China and the gas power plants in the Middle East as many production capacities were expected to be eliminated under the low-emission scenario. Therefore, for these power plants, a simplified elimination strategy based on technical attributes including the age, size, technology, and application of the power generation units was adapted and applied according to Cui et al. 56 It was also assumed that no extra capacity of these power plants would be added other than the ones that have already been announced, permitted, or are under construction (see section S1.4.2 in ESI† for details).

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Many chemical manufacturing sites are closely located in industrial parks. For a better map visualization, manufacturing sites within a 50-kilometer (km) circle were aggregated into chemical manufacturing clusters. The same aggregation was applied to each type of the $\rm CO_2$ sources. The direct distance between each chemical manufacturing cluster and each $\rm CO_2$ supply cluster within the region was calculated.

Transportation by trucks was assumed as a conservative assessment, as it represents the worst-case transportation scenario with the highest carbon footprint, and it does not require major infrastructure to be built. The dataset "transport, freight, lorry 16–32 metric ton, EURO3|RoW" in ecoinvent 3.8 was used to quantify the impacts of $\rm CO_2$ transportation by trucks. Scenarios with pipeline transportation were investigated as a best-case transportation scenario in the sensitivity analysis (described in section S1.4.4 in ESI†).

Based on the environmental and health impacts of carbon capture and truck transportation (calculation details in section S1.4.3 in ESI†), a linear model was developed to optimize the supply amount from each CO_2 supply cluster i to each chemical manufacturing cluster j ($x_{i,j}$) that would minimize the total feedstock-related GHG emissions or PM-related health impacts (see eqn (1)):

$$\min_{x_{ij}} \sum_{i} \sum_{j} (x_{ij} DIST_{ij} IMPACT_{ij}^{TR} + x_{ij} IMPACT_{i}^{CC})$$
 (1)

where $\mathrm{IMPACT}_{i,j}^{\mathrm{TR}}$ and $\mathrm{IMPACT}_{i}^{\mathrm{CC}}$ represent the impact intensity (climate change impacts or PM-related health impacts) of transportation and carbon capture, respectively, and $\mathrm{DIST}_{i,j}$ is the distance between i and j.

This objective function is subjected to the constraint of total CO_2 capture capacity of each supply cluster i (TS_i) and the total CO_2 demand by each chemical manufacturing cluster j (TD_j). If the regional supply would be able to cover its demand, the constraint functions are:

$$\sum_{j} x_{i,j} \le TS_i \forall i$$

$$\sum_{i} x_{i,j} \ge TD_j \forall j$$
(2)

where $x_{i,j}$ represents the CO₂ supply volume from supplier i to consumer j.

In the cases where regional supply would be insufficient to cover its demand and all regional supply would be used up, the constraints function become:

$$\sum_{j} x_{i,j} \ge TS_i \forall i$$

$$\sum_{j} x_{i,j} \le TD_j \forall j$$
(3)

3. Results

3.1 Global and regional supply-demand balance of CO₂

The global mass flow of CO_2 in 2050 (Fig. 2a) indicates that the CO_2 demand from the chemical industry (2.2–3.1 Gt CO_2 -

feedstock) can be well covered by the global supply capacity: 6.6–16.6 Gt of fossil and biogenic CO₂ are projected to be emitted from the power plants, cement kilns, steel mills, and kraft pulp mills globally under the two scenarios, of which 5.6–14.0 Gt could be captured (around 85% of the emissions). In other words, the demand from the chemical industry makes up 22% and 42% of the maximum CO₂ capture capacity under high- and low-emission scenarios, respectively. The power sector is the largest CO₂ emitter under both scenarios. However, unlike the high-emission scenario, under which coal-fired power plants contribute to almost one-third of the total CO₂ emissions, biomass-fired power plants are potentially the major CO₂ supplier under the low-emission scenario.

Meanwhile, the CO₂ supply-demand balance has a very strong regional pattern (Fig. 2b). Among the nine countries/regions investigated in detail, China and India have the largest CO₂-supply surplus under both the scenarios. In the EU, although fossil CO₂ emissions would decrease by almost 90% under the low-emission scenario in 2050 in comparison to 2019, biomass-fired power plants are projected to be rapidly deployed, emitting substantial amounts of biogenic CO₂. As a result, it is estimated to have a large CO₂-supply surplus of 0.45 Gt per year, next to China and India.

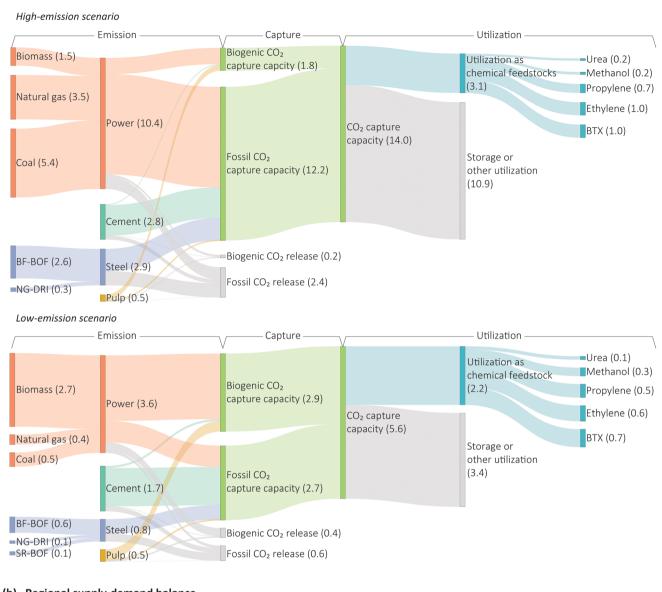
The Middle East region and the Republic of Korea, on the other hand, have relatively large chemical industries. However, CO₂ emissions under the low-emission scenario will drop drastically in comparison to 2019, mainly due to the decarbonization of the local electricity grid. Even if all CO₂-feedstocks within the region were supplied, there would likely still be a supply shortage. The insufficient supply would especially be noteworthy for the chemical industry in the Middle East under the low-emission scenario, where only 27% of the CO₂ demand from the chemical industry could be fulfilled with its intra-regional supply.

3.2 Environmental and health impacts of carbon capture

The GHG savings and PM-related health impacts of capturing 1 kg CO₂ depend on the sector from which carbon capture is performed. This is because of the different CO₂ concentrations in the flue gas and the different steam sources and electricity mixes used, as illustrated by the environmental-merit-order (EMO) curves in Fig. 3.

By capturing 1 kg $\rm CO_2$, the maximum GHG saving would be 1 kg $\rm CO_2$ -eq., assuming no additional energy input were required for carbon capture. Considering GHG emissions from the additional energy inputs, the actual GHG savings due to carbon capture from the different sectors would be between 0.6 and 0.9 kg $\rm CO_2$ -eq per kg $\rm CO_2$ captured on a global average. As a trade-off, between 1 and 5 × 10⁻⁸ disability adjusted life years (DALY) per kg $\rm CO_2$ captured PM-related health impacts would be attributed to carbon capture. If $\rm CO_2$ emissions in 2050 were captured to the full capacity, 12 000–33 000 DALYs would be lost every year because of PM-related health impacts, which corresponds 0.06–0.17% of the global total PM-related health impacts in 2019.⁵⁷

(a) CO₂ Mass flow (unit: Gigatonne)



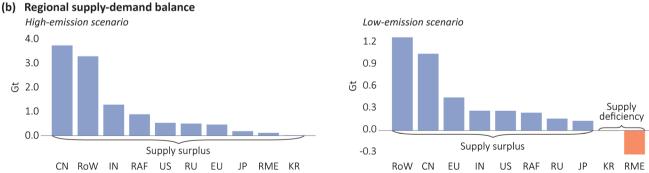


Fig. 2 (a) Estimated global CO₂ mass flow under the high- and low-emission scenarios (note the mass flows are differently scaled under the two scenarios). (b) Regional CO₂ supply-demand balance under the high- and low-emission scenarios. Abbreviations: CN, China; EU, the European Union; IN, India; JP, Japan; KR, the Republic of Korea; RAF, Region Africa; RME, Region Middle East; RU, Russian Federation; US: the United States of America; RoW: Rest of the World; BF-BOF, blast furnace-basic oxygen furnace; DRI-EAF, direct reduced iron-electric arc furnace; SR-BOF, smelting reduction-basic oxygen furnace.

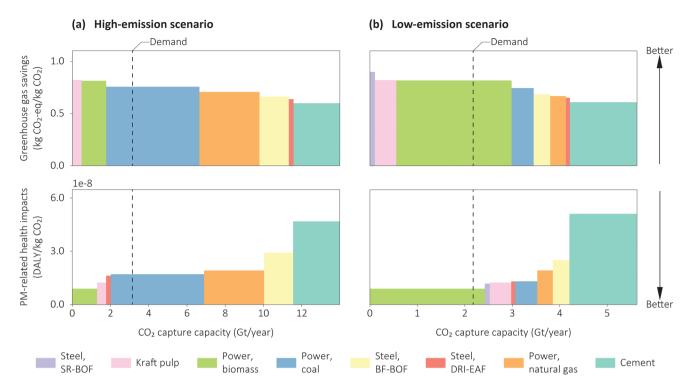


Fig. 3 Greenhouse gas savings (top) and PM-related health impacts (bottom) of 1 kg CO_2 captured from different industries, calculated with global average impact factors, plotted against the maximum global sectoral CO_2 supply capacity (the dashed line indicates the maximum CO_2 demand from the chemical industry) under (a) high-emission scenario and (b) low-emission scenario. Abbreviations: BF-BOF, blast furnace-basic oxygen furnace; DRI-EAF, direct reduced iron-electric arc furnace; SR-BOF, smelting reduction-basic oxygen furnace.

The GHG savings and PM-related health impacts of carbon capture are dominated by the amount and type of energy that needs to be additionally fed into the system. Generally speaking, higher CO_2 concentration in the flue gas would lead to a lower energy requirement for carbon capture. An extreme example is SR-BOF mills, where CO_2 concentration in the flue gas is close to 100%. In this case, only 0.15 kW h electricity per kg CO_2 captured is required for the compression. As a result, with 0.90 kg CO_2 -eq. GHG savings and 1.19 \times 10⁻⁸ DALY PM-related health impacts per kg of CO_2 captured, SR-BOF steel mills are one of the most preferred CO_2 source under the low-emission scenario. However, SR-BOF steel mills are only assumed to be in place under the low-emission scenario due to its low TRL today, and their maximum CO_2 supply capacity remains low.

Other preferred CO₂ suppliers with low carbon capture-related environmental and health impacts and large supply potential include biomass-fired power plants. This is because we assume that for power plants, the environmental and health impacts of carbon capture come from the regional electricity grid mix to compensate for the power loss due to carbon capture (see section 2.2). Biomass-fired power plants have higher CO₂ concentrations in the flue gas than natural gasfired power plants, leading to less power loss. In addition, biomass-fired power plants have a large share in the EU, where the electricity grid mix has one of the lowest impact factors, further reducing the impacts of carbon capture from them.

Fig. 4 shows that the impacts of carbon capture also have a strong regional pattern. This is mainly due to the different impact factors of the regional energy mix (Tables S45 and S46 in ESI†). Carbon capture in the EU can yield the most GHG savings with the lowest PM-related health impacts in most cases, thanks to its relatively clean electricity and fuel mix. Least savings in GHG emissions would be achieved by carbon capture in China and India as the result of their high dependency on coal in the fuel mix. Due to major advances in PMemission control in the Chinese power plants and the overall high PM emissions from a broad range of other sources in the region,³⁷ the carbon capture-induced relative PM-related health impacts in China are estimated to be comparable to other regions with lower pollution levels such as the EU. In contrast, in India, strict emission control of sulfur dioxide and nitrogen oxides in the industry37 is lacking, causing carbon capture also having the highest PM-related health impacts among the nine countries/regions investigated. These tendencies depend strongly on the regional background pollution and the emission regulations in 2050, and are thus subject to major uncertainties.

3.2.1 Sensitivity analysis: electricity and steam sources. Since the environmental and health impacts of carbon capture comes mainly from the associated energy consumption, the EMO curves vary when using different energy mixes (Fig. S2–S5 in ESI†). For electricity, besides the regional grid mix in 2019 as in the base case (Fig. 3 and 4), we also examined scen-

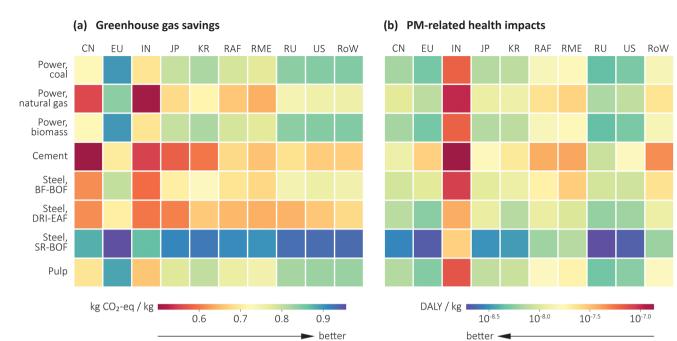


Fig. 4 Heat map of (a) greenhouse gas savings and (b) PM-related health impacts of capturing 1 kg CO₂, by supply sector and region. Abbreviations: CN, China; EU, the European Union; IN, India; JP, Japan; KR, the Republic of Korea; RAF, Region Africa; RME, Region Middle East; RU, Russian Federation; US: the United States of America; RoW: Rest of the World; BF-BOF, blast furnace-basic oxygen furnace; DRI-EAF, direct reduced iron-electric arc furnace; SR-BOF, smelting reduction-basic oxygen furnace.

arios with the projected regional grid mix in 2050 with and without full CCUS deployment. The impacts associated with carbon capture are lower when using the projected electricity grid mix in 2050, both with and without CCUS. The sectoral rankings in the EMO curves are not significantly affected, with the exceptions noted for the electricity grid mix with full CCUS development under the low-emission scenario. In this case, thanks to the carbon capture at biomass-fired power plants, higher electricity demand would lead to higher savings in GHG emissions, which is counter-intuitive. However, net-negative biomass electricity may not be an unlimited good due to the availability of suitable biomass, associated costs and other impacts of biomass-fired power plants, and all these factors should be assessed in future studies.

Besides using mixed-fuel boilers, the steam for MEA regeneration in cement kilns and steel mills can also be produced with natural gas boilers or electrode vessels. When natural gas is used as the fuel for steam generation, the ranking of cement kilns in EMO curves would be improved, especially when combined with the electricity grid mix in 2019. Scenarios of using electrode vessels to produce stream would have the lowest carbon footprint of all MEA regeneration options when the electricity grid mix is decarbonized.

3.3 Bottom-up case studies: optimizing the CO₂ supply to chemical manufacturing sites in China and the Middle East

China: The priority rankings of the four types of CO₂ sources in the EMO curve for China are the same, in terms of both GHG savings and PM-related health impacts: coal-fired power

plants are the most preferred suppliers, followed by BF-BOF steel mills, natural gas-fired power plants, and cement kilns (Fig. 5a and b).

However, in the supply-chain optimization where the impacts of truck transportation of CO2 are additionally considered, the results by minimizing the total GHG emissions (Fig. 5c) and by minimizing the total regional PM-related health impacts (Fig. 5d) are different. As a general pattern, the transportation distance is longer when minimizing the total regional climate change impacts (average transportation distance being 224 km) than when minimizing the total regional PM-related health impacts (average transportation distance being 182 km). For example, when minimizing the total regional PM-related health impacts, a local cement kiln would be chosen as the CO₂ supplier instead of a coal power plant if located more than 228 km away (break-even distance). When minimizing the total regional climate change impacts, this break-even distance becomes 1282 km. Overall, truck transportation accounts for 7-9% of the total climate change impacts and 32-38% of the total PM-related health impacts in the value chain of carbon capture and transportation (Table S53 in ESI†).

3.3.1 Sensitivity analysis 1: CO₂ transportation using pipelines. When pipelines were used for CO₂ transportation in China, the transportation distance would be longer in general (average distances being 227–233 km) because of the relative smaller impacts from pipeline transportation in comparison to truck transportation (Fig. S6 and Table S54 in ESI†). The coal power plants would still be more preferred than the cement kilns in terms of savings on climate change impacts

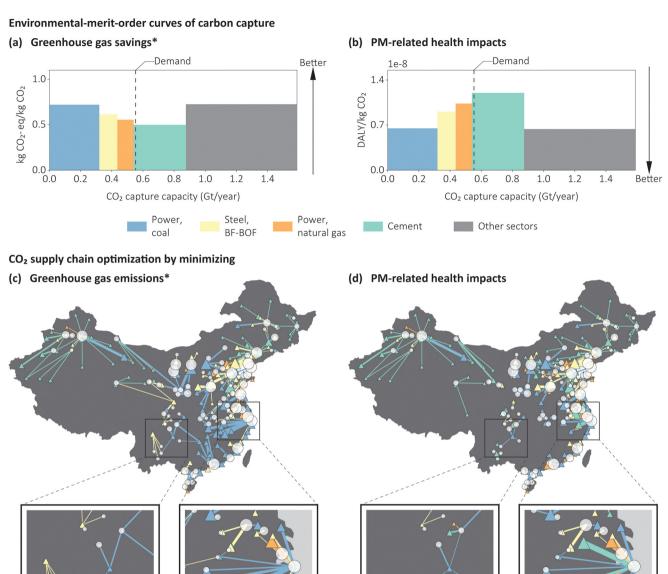


Fig. 5 (a) and (b): GHG savings and PM-related health impacts of carbon capture from different sectors in China. (c) and (d): CO₂ supply chain optimization in China under the low-emission scenario in 2050 by minimizing the total regional climate change impacts and the total regional PM-related health impacts. Zoomed-in areas represent examples of major differences between the two strategies. Abbreviations: BF-BOF, blast furnace-basic oxygen furnace.* GHG emissions of carbon capture = captured CO₂ amount – GHG savings.

▲ 1-5

0 1-5

— 1-5

▲ Power, natural gas

▲ <1

0 <1

— <1

Power, coal

even if they were located 3102 km farther than the cement kiln. This break-even distance would become 4802 km when considering the PM-related health impacts. As a result, total climate change impacts and total PM-related health impacts due to carbon capture and transportation in China would be

reduced by 6–9% and 34–36%, respectively, when pipelines were used for $\rm CO_2$ transportation in comparison to the basic scenario to fulfill the demand from the chemical industry.

▲ 5-10

O 5-10

5-10

△ Steel, BF-BOF

3.3.2 Sensitivity analysis 2: biomass-fired power plants as additional CO_2 suppliers. Biomass-fired power plants are

CO₂ supply (Mt)

Supplier sector

CO₂ demand (chemical industry) (Mt)

Transportation amount (Mt)

▲ >10

>10

▲ Cement

among the most preferred CO2 suppliers in China, with its low carbon capture-related impacts and large potential capture capacity under the low-emission scenario. Assuming truck transportation, when minimizing the total climate change impacts, all of the CO2 demand from the chemical industry would be supplied by nearby solid biomass- or coal-fired power plants, with an average transportation distance of 170 km (Fig. S7 and Table S55 in ESI†). When minimizing the total PM-related health impacts, more local suppliers of other types would be used and the average transportation distance would become 131 km. Still, solid biomass- and coal-fired power plants would account for 86% of the CO₂ supply to the chemical industry. Overall, when biomass-fired power plants were considered as additional CO2 suppliers to the chemical industry in China, total climate change impacts and total PMrelated health impacts due to carbon capture and transportation would be reduced by 18% and 24-28%, respectively.

The Middle East represents a region with insufficient CO2 supply under the low-emission scenario in 2050. Only 27% of the demand from the chemical industry could be fulfilled by carbon capture (Fig. 6a), even if it were deployed at the maximum capacity at all the natural gas-fired power plants, DRI-EAF steel mills, and cement kilns. Since all supply sources are utilized to the full capacity, the linear optimization model can only minimize the total transportation distance. As a result, the optimization results by minimizing the total GHG emissions and total PM-related health impacts are the same (Fig. 6b).

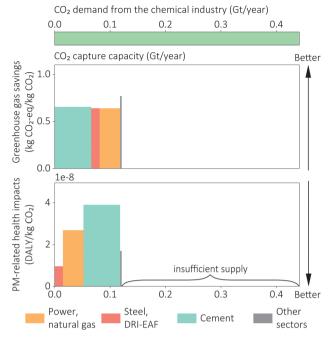
Biogenic electricity production is expected to be deployed in the Middle East in 2050, up to 59 terawatt-hour (TW h) under the low-emission scenario (see Table S15 in ESI†). Since there are only biogas-fired power plants but no solid biomassfired power plants in the region in 2019, we assumed the biogenic electricity in 2050 would also primarily come from biogas, which was excluded from the scope of carbon capture in this study due to the small average capacity of biogas-fired power plants (described in section S1.1.1 in ESI†). Even if we assume all 59 TW h biogenic electricity would come from solid biomass with another 50 Mt of CO₂ supply (as calculated with global average fuel efficiency under the low-emission scenario in 2050), only 40% of the feedstock demand from the chemical industry could be covered with local CO₂ supply.

4. Discussion

Feasibility of a CO₂-based chemical industry

The two scenarios explored in this study provide insights into the potential maximum and minimum range of CO₂ emissions in 2050. We estimate the global maximum CO2 capture capacity from power plants, cement kilns, steel mills, and kraft pulp mills in 2050 to be between 5.6 and 14.0 Gt. Our lower limit (from the low-emission scenario) is slightly lower than IEA's estimation in its net zero emissions scenario (7.6 Gt annual CO2 capture by 2050, including 1.0 Gt from direct air capture).⁵⁸ The main reason for the difference is that we do

(a) Environmental merit order curves of carbon capture



(b) CO₂ supply chain optimization

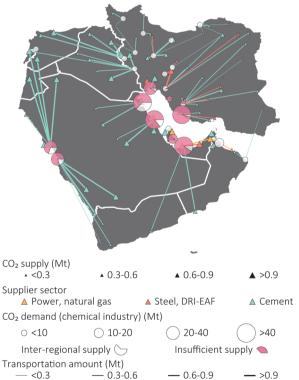


Fig. 6 (a) GHG savings and PM-related health impacts of carbon capture from different sectors in the Middle East. (b): CO2 supply chain optimization in the Middle East under the low-emission scenario in 2050. Abbreviations: DRI-EAF, direct reduced iron-electric arc furnace.* GHG emissions of carbon capture = captured CO2 amount - GHG savings.

not consider the chemical and refinery industries as potential CO₂ suppliers in this study, as our base assumption is net-zero emissions from the chemical industry.

We further investigate the $\rm CO_2$ -based production routes of the seven key chemicals that have either been commercialized or are with relatively high TRL as the key pathway to achieve net-zero GHG emissions of the chemical industry. As such, the maximum $\rm CO_2$ demand from the chemical industry in 2050 are estimated to be between 2.2 and 3.1 Gt. Our result is slightly lower than the estimated 2.8–3.7 Gt global $\rm CO_2$ demand from a $\rm CO_2$ -based chemical industry in 2030 by an early study, ⁴⁹ mainly because they assumed ammonia being also produced via a $\rm CO_2$ -based route.

Globally, our estimates suggest that the feedstock demand from the chemical industry can be well covered by the maximum CO₂ capture capacity. Hence, feedstock availability is not a constraint factor for the transition towards a fully CO₂-based chemical industry on the global basis. Despite the global abundancy of CO₂ supply, certain countries/regions such as the Republic of Korea and the Middle East might still face in-region supply shortage in 2050. A fully-CO₂ based chemical industry in those regions would require more costly options with higher environmental impacts, such as feedstock import from other regions and additional implementation of direct-air-capture facilities.³⁴

Furthermore, it is noted that the cost and stable availability of feedstocks usually play an important role in planning the locations of manufacturing sites for bulk chemicals. For example, the traditional petrochemical industry in the Middle East underwent rapid development in the past decades, due to the abundance of the low-cost feedstocks of oil and gas. However, with the intended transition of the chemical industry, the current feedstock advantage in the Middle East would diminish in the future. This indicates the CO₂-based chemical manufacturing sites might need to be re-positioned based on the regional feedstock supply-demand balance (see section 4.3 below).

4.2 Upscaling carbon-capture projects

While the maximum CO₂ capture capacity is not a constraint for the CO₂-based chemical industry on the global scale, the speed of actual deployment of carbon-capture projects can limit the large-scale CO₂ utilization. As of 2021, the global operational capacity of carbon capture is 0.04 Gt CO₂ per year. Is In order to satisfy the need from the chemical industry, the global capacity needs to increase by 55–78 times within the upcoming 30 years. The current main obstacles that limit large-scale deployment of carbon-capture projects in the short to medium term include the technical barrier to lower the energy penalty of carbon capture and the high financial cost. However, a previous study found that CCUS is more cost-effective than other carbon mitigation pathways in the long run, indicating the need for enabling policy to upscale carbon-capture projects.

Our study further demonstrates that EMO curves (Fig. 3) and impact heat maps (Fig. 4) are useful tools to decide which sectors and regions are to be prioritized for carbon-capture

projects. Our priority rankings in terms of GHG emission savings are comparable to the findings by Müller et al. for the EU, 35 although the carbon-capture method was not specified in their study. Carbon capture from solid biomass-fired power plants and kraft pulp mills in the EU would bring the highest savings on climate change impacts and incur the least PMrelated health impacts. Thus, future carbon-capture projects may first prioritize them by retrofitting these CO₂ point sources. Note that biomass-fired power plants and kraft pulp mills are usually not a major concern as GHG emitters, as they emit mainly biogenic CO2. However, as they are identified as the most preferred CO₂ supplier in this study, we highlight that these sectors should target negative CO₂ emissions by capturing biogenic CO2 to minimize the carbon capture-related impacts of the entire society. In the regions with their electricity grid mixes that have relatively high carbon footprint today, such as India and China, the benefits of GHG savings from carbon capture would be limited. This indicates the need for an energy transition as a prerequisite for making carbon capture more sustainable there.

Chemical absorption with MEA is included in this study as the main carbon-capture method due to its high TRL. It serves as a benchmark in terms of energy penalty and impacts for other capturing methods. Meanwhile, more advanced carbon-capture methods are being developed. For example, the new generation of alkanolamine solvents developed for chemical absorption of CO₂ are able to reduce the steam demand to 2.3 MJ kg⁻¹ CO₂ captured. Also, membrane-based CO₂ separation is being more widely studied due to its lower energy consumption than conventional carbon-capture methods. With the development of these new technologies, the GHG savings from carbon capture are expected to be higher than estimated in this study.

4.3 Towards net-zero emissions of the chemical industry

In addition to carbon capture, several other factors need to be considered in order to achieve a sustainable CO2-based chemical industry. For example, CO₂ transportation between various suppliers and chemical manufacturing sites needs to be carefully planned. Previous studies pointed out that pipeline transportation would be the most economically feasible regional transportation method when CCUS projects are up-scaled. 62 At the moment, the total length of CO₂ pipelines in operation worldwide is only around 8000 km, predominately in the United States for enhanced oil recovery. 63 However, in the China case study, we find that around 70 000 km (linear distance) pipeline network would need to be in place in order to achieve that all existing Chinese chemical manufacturing clusters receive their CO₂ supply via pipelines according to the optimization results (Fig. S6 in ESI†). The up-scaling of CO₂ pipeline requires cross-sectoral collaboration, especially regarding early-stage planning of point-to-point match of CO₂ suppliers and consumers.

In this study, we focus on the environmental and health impacts of carbon capture, without considering the impacts of converting CO_2 into chemicals. However, the latter is a necessary step to understand the overall environmental benefits and

trade-offs of a CO2-based chemical industry in comparison to the current petrochemical industry. For example, besides CO₂, hydrogen is also an important feedstock needed for a CO2based chemical industry. Based on the specific hydrogen demand in the individual synthesis routes considered (S1.2 in ESI†), the global hydrogen demand by a CO₂-based chemical industry in 2050 is estimated to be 344-501 Mt. This translates into 11-17 petawatt-hour (PW h) of electricity demand, taking into account recent research progress that has drastically decreased the electricity consumption of hydrogen production from >50 to 33 kW h kg⁻¹ H₂.⁶⁴ This electricity demand for hydrogen production corresponds to 50-72% of the world electricity final consumption in 2019.65 Recent LCA studies have demonstrated that CO2-based chemicals such as methanol and ethylene would have lower carbon footprint than their conventional counterparts only when using low-carbon hydrogen, such as green hydrogen from water electrolysis powered with zero- to low-carbon electricity. 66-69 Therefore, speedy upscaling of the capacity of zero- to low-carbon electricity is another pre-requisite for the transition to a fully CO2-based chemical industry.

In addition, development of low energy-consuming CO2based chemical production routes is essential. Lowering energy consumption can be achieved through advancements in catalyst development. For example, with mesoporous silver-catalyst, CO2 fixation of amines can be achieved under room temperature, significantly reducing the energy requirement for the synthesis of N-formylated amines. 14 In addition, research is ongoing to directly hydrogenate CO2 with hydrogen into ethylene, propylene, or BTX aromatics by using selective catalysts. 70,71 If such routes were commercialized and utilized instead of the conventional high-TRL manufacturing routes considered in this study, the total hydrogen demand from a global fully CO2-based chemical industry could be reduced by 35%.⁴⁹

In this study, we mainly consider the CO₂-based production routes for base chemicals, and assume the production of other chemicals would remain unchanged. However, we do acknowledge the rapidly evolving research of CO2-based production routes for other chemicals. For example, with metal nanoparticle catalysts or electrocatalysis, carbamate (a specialty chemical of high relevance in pharmaceutical and agrochemical industries) can be synthesized through direct incorporation of CO₂ at mild conditions. 6,72 In addition to the benefits of lowering energy consumption, this novel synthesis route can avoid the use of hazardous reagents as in the traditional production routes, fulfilling the green-chemistry concept.

In addition, in regions with potential supply constraints of CO2-feedstock or renewable electricity, CO2-based chemical production should be first considered for chemicals with the largest savings on climate change impacts when comparing with the corresponding petrochemicals. A full cradle-to-gate comparative LCA would be necessary as the next step to understand which types of reactions should be prioritized for CO₂ utilization as feedstock.

We acknowledge that many other strategies are being investigated by the chemical industry for its transition to net zero

emissions, including carbon capture at existing chemical manufacturing sites for storage (the CCS pathway), a circular economy with enhanced plastics recycling (the CE pathway), and utilizing biomass as feedstock (the biomass pathway). With the CCS pathway, existing chemical manufacturing sites would be able to continue using their current technology and infrastructure, avoiding the costly stranding of their existing assets. 11 However, net-zero emissions can be hardly achieved with CCS alone because the multiple CO₂ emission points on a chemical manufacturing complex with varied flow rates and CO2 concentrations make it difficult to capture all emissions from every single stack. In addition, the chemical industry will still be highly dependent on fossil fuels with the CCS pathway.

The CE pathway provides synergies to deal with plastic waste and reduce the virgin feedstock demand for plastic production. However, the benefits of plastics recycling might be curtailed by the low collection rates,73 limited utilization options for secondary plastics,74 and hazardous additives used in the plastic production.⁷⁵ The theoretical maximum material recycling rate (combined for mechanical and chemical recycling) was assessed to be 70% across all plastic types, 76 vet the best rate of plastic recycling for material use achieved so far is only 24% in Germany in 2019.⁷⁷ In addition, the material recycling of other chemical products such as thermosets is even more difficult. Therefore, the role of a circular economy in reducing demand of virgin feedstocks in the chemical industry is projected to remain limited in the foreseeable future.

With the biomass pathway, the electricity demand from plastic production can be reduced by more than 90% in comparison to utilizing CO2 as the feedstock, but at the expense of more than 40 EJ biomass demand. 76 The availability of future biomass supply from energy crops, wastes and residues, and forestry for material and energy use is highly uncertain with estimations between 107 and 1723 EJ, particularly due to different critical assumptions in crop yield, future population and diet shift.⁷⁸ Depending on the type of biomass utilized, various environmental impacts might be incurred as a consequence, such as biodiversity loss, ocean acidification, or eutrophication. 11,79 Therefore, a thorough analysis of using different biomass types as chemical feedstocks is required, taking into account the future biomass availability, TRL of biomass conversion technologies, and environmental impacts from feedstock acquisition and conversion.

Taking into account the pros and cons, previous and our studies suggest that a combination of CCUS and the aforementioned strategies would be needed for transitioning the chemical industry towards net-zero emissions.⁷⁶ More in-depth analysis of the three different pathways above and possible combinations, together with CCUS, are warranted to develop the best global and regional net-zero-emission strategies for the chemical industry.

4.4 Towards net-zero emissions of the whole society

This study helps to set priorities regarding where to start capturing CO₂. To keep the temperature increase within 1.5 °C, it is imperative to capture CO_2 emissions in almost all sectors and regions in 2050, provided the additional environmental and health impacts associated with carbon capture being not prohibitive.⁵⁸ Once reaching such full carbon capture, from the perspective of the whole society, new chemical manufacturing sites can be planned at locations with easy access to CO_2 -feedstock and renewable electricity, regardless of the CO_2 supplying sectors.

Besides the chemical industry, other sectors also rely on CO_2 utilization as a potential low-carbon pathway. For instance, a recent study estimated the potential demand of CO_2 from the fuel sector to be between 0.9 and 3.1 Gt per year in 2050. Because However, when used as fuel for transportation, CO_2 is released immediately upon combustion. As such, CO_2 emissions are only delayed by days or months, and it is difficult to capture the emitted CO_2 again. Future studies on global and regional CO_2 supply-demand balance need to take its potential utilization in other sectors into consideration as well. Temporally dynamic cradle-to-grave LCAs need to be conducted to compare different potential CO_2 utilization pathways to decide which pathway should be implemented and prioritized to maximize the environmental benefits of carbon capture and utilization.

A successful CCU value chain would need early-stage planning that involves the collaboration across sectors to solve potential conflicts of interest of different stakeholders. One open debate is regarding how to allocate the credits (e.g., GHG savings from carbon capture) and burdens (e.g., PM-related health impacts) related to carbon capture between the $\rm CO_2$ emitter (e.g., electricity) and the $\rm CO_2$ consumer (e.g., $\rm CO_2$ -based chemicals). 35,81,82 In this study, we quantify the total GHG savings and PM-related health impacts from carbon capture from the point view of the society as a whole, and hence, do not need to perform allocation between the $\rm CO_2$ emitters and consumers.

For future LCA of CO₂-based chemical products, we propose to continue considering CO2 as a waste because the main product system (e.g., power plants) would pay for carbon capture as an emission abatement method. In this case, the main product system should get all the credits and burdens related to carbon capture, and the chemical industry gets burden-free CO₂ supply from the power or industrial sectors. This is a different approach from Müller et al., 35 where they considered CO2 as a valuable product, and hence, CO2-feedstock should be credited for the avoided CO2 emissions using the substitution approach. Their approach means that the power plants or cement kilns cannot reduce or avoid CO2 emissions with the CCU (since the credit of the reduction goes to the user of CO₂). However, we would argue that the CCS approach is a case of waste treatment, and the main product would be credited for CO₂ removal. Similarly, CCUS could be considered to be a waste treatment method so that it will not make a difference for the main product system in terms of environmental and health impacts whether the captured CO₂ is for storage or utilization. In other words, our recommendation is to ensure the methodological consistency of allocation in LCA among different downstream mechanisms of carbon capture. In addition, to reach net-zero emissions of the whole society, more CO₂ needs to be captured than demanded from the chemical industry. In this sense, it should not make a difference for the chemical industry which industrial CO₂ source they utilize.

Furthermore, some researchers argue that a focus on CO₂ utilization instead of storage may cause costly distractions from climate change mitigation to reach net-zero GHG emissions because it does not guarantee the net removal of CO₂. Storage capacity that CCS faces, especially in India and China. Additionally, increased implementation of CCU and circular chemical use can help the chemical industry to move away from the reliance on fossil fuel.

4.5 Uncertainties and sensitivity analysis

The uncertainty of CO_2 supply and demand in 2050 is addressed by using a worst-case high-emission scenario and a best-case low-emission scenario to capture the possible range. Under the both scenarios, we find that the CO_2 -feedstock demand from the chemical industry can be well covered by the CO_2 supply on the global average.

The environmental and health impacts of carbon capture are sensitive to the choice of electricity and fuel mixes. When the electricity grid mix were decarbonized, using steam generated in electrode vessels for MEA regeneration would provide the largest GHG savings. However, with the current electricity grid mix, natural gas boiler would be recommended for steam generation on the global average because of its lower impacts.

Power systems, such as coal-fired power plants, may also choose to keep the power output from the primary unit constant. In that case, the energy for carbon capture would need to be supplied by additional fuel input into the system. The CCS facility at the Petra Nova power plant in the US (currently suspended), for example, built a 75-megawatt auxiliary gas turbine to supply electricity and steam for the capture unit. ⁸⁴ However, since CO₂ from the gas turbine is not captured, the overall rate of CO₂ emissions with the Petra Nova model was found to be more than twice as high as with the Boundary Dam model that does not rely on additional fuel source for the energy supply to the capture unit. ⁸⁵

When planning future point-to-point CO_2 supply chain, many of the potential emission sources in 2050 could still be unknown, especially for solid biomass-fired power plants and SR-BOF steel mills that are currently not widely deployed. In the sensitivity analysis in the China case study, we assume the to-be-eliminated coal power plants are retrofitted into solid biomass-fired power plants. It might also be a sensible decision to build solid biomass-fired power plants and SR-BOF steel mills as CO_2 suppliers adjacent to chemical manufacturing sites to minimize the cost and impacts of CO_2 transportation.

5. Implications

Paper

Building on the findings, we would like to highlight the following options as future actions to ensure an effective and efficient systemic low-carbon transition of the chemical industry and the entire society.

- (1) Feedstock availability should be taken into account when planning future chemical manufacturing sites. Manufacturing sites of bulk chemicals are usually centralized in regions with easy and low-cost access to feedstocks. With the feedstock transition of the chemical industry from oil and gas to CO₂, the scale of the chemical industry in regions such as the Middle East would be expected to shrink due to insufficient local feedstock supply. This could be less ideal for the region's economic development, and corresponding measures need to be considered to mitigate such negative impacts (e.g., by transitioning from large-scale production of low-price base chemicals to smaller-scale production of high-price chemicals). On the contrary, the development of the chemical industry in China was constrained by the availability of oil and gas in the past. With the highest CO₂ supply potential, the chemical industry there could be further developed. Before such expansion takes place, it is key to ensure the availability lowimpact electricity grid mix in China.
- (2) Low-impact energy mix is fundamental before the large-scale deployment of CCUS projects. Enormous heat and electricity demand is expected from the CCUS value chain: from CO₂ capture and compression, to transportation, and to produce hydrogen as co-reactants. Therefore, to ensure the largest savings on climate change impacts with the least additional other impacts, it is essential to prioritize low-impact energy mix. This implies that the immediate priority for China and India should be to increase the share of renewable energy in their electricity grid mix to make the CCUS value chain more sustainable.
- (3) Strategic cross-sectoral planning is key to minimizing the total environmental impact for the whole society to achieve netzero GHG emissions. In recent years, all sectors are developing their own net-zero-emission pathways. For example, many coalfired power plants are considering being retrofitted into solid biomass-fired power plants, as a pathway to reduce their greenhouse gas emissions while avoiding the need of carbon capture. ⁸⁶ However, our analysis reveals that larger cross-sectoral co-benefits from carbon capture can possibly be achieved and should be strived for. Among others, solid biomass-fired power plants should be prioritized for CCUS projects because of its lower carbon capture-related environmental impacts. We call on policy makers and industrial stakeholders to establish open and transparent dialogues and benefits-sharing mechanisms to foster such cross-sectoral collaborations.

Author contributions

J. H. conceived the idea, collected and processed data, and wrote the manuscript. Z. W. conceived the idea, assisted with

research, and edited the manuscript. C. O. modelled the regionalized PM-related health impacts, and assisted with data collection and manuscript editing. G. G.-G. assisted with research and manuscript editing. S. H. supervised the project, assisted with research and manuscript writing.

Conflicts of interest

The authors declare no conflict of interest.

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References

- 1 S. A. Matlin, G. Mehta, H. Hopf and A. Krief, *Nat. Chem.*, 2015, 7, 941–943.
- 2 WBCSD, Chemical Sector SDG Roadmap, World Business Council for Sustainable Development, Geneva, Switzerland, 2018.
- 3 IEA, *The future of petrochemicals*, International Energy Agency, Paris, France, 2018.
- 4 IEA, World energy statistics and balances, International Energy Agency, 2021.
- 5 A. M. Bazzanella and F. Ausfelder, *Low carbon energy and feedstock for the European chemical industry*, DECHEMA, Frankfurt am Main, Germany, 2017.
- 6 S. Chongdar, S. Bhattacharjee, S. Azad, S. Samui, S. Dutta, R. Bal and A. Bhaumik, ACS Appl. Mater. Interfaces, 2021, 13, 40157–40171.
- 7 E. Alper and O. Y. Orhan, Petroleum, 2017, 3, 109-126.
- 8 A. Otto, T. Grube, S. Schiebahn and D. Stolten, *Energy Environ. Sci.*, 2015, **8**, 3283–3297.
- 9 S. C. Peter, ACS Energy Lett., 2018, 3, 1557-1561.
- 10 A. Mustafa, B. G. Lougou, Y. Shuai, Z. J. Wang and H. P. Tan, J. Energy Chem., 2020, 49, 96–123.
- 11 P. Gabrielli, M. Gazzani and M. Mazzotti, *Ind. Eng. Chem. Res.*, 2020, **59**, 7033–7045.
- E. Lam, K. Larmier, P. Wolf, S. Tada, O. V. Safonova and C. Coperet, J. Am. Chem. Soc., 2018, 140, 10530–10535.
- 13 O. Martin, A. J. Martin, C. Mondelli, S. Mitchell, T. F. Segawa, R. Hauert, C. Drouilly, D. Curulla-Ferre and J. Perez-Ramirez, *Angew. Chem.*, *Int. Ed.*, 2016, 55, 6261–6265.
- 14 S. Chongdar, S. Bhattacharjee, S. Azad, R. Bal and A. Bhaumik, *Mol. Catal.*, 2021, **516**, 111978.
- 15 Chinese Academy of Science, "Liquid Sunshine" Enlightens New Way of Green Energy, https://english.cas.

- cn/newsroom/research_news/chem/202011/t20201102_247124. shtml, (accessed April, 2022).
- 16 IEA, Data and statistics, https://www.iea.org/data-and-statistics/data-browser/?country=WORLD&fuel=CO2%20emissions&indicator=CO2BySector, (accessed Dec. 15, 2021).
- 17 J. Rogelj, D. Shindell, K. Jiang, S. Fifita, P. Forster, V. Ginzburg, C. Handa, H. Kheshgi, S. Kobayashi, E. Kriegler, L. Mundaca, R. Séférian and M. V. Vilariño, in An IPCC Special Report on the impacts of global warming of 1.5 °C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the global response to the threat of climate change, sustainable development, and efforts to eradicate poverty, 2018.
- 18 Global CCS Institute, *Global Status of CCS: 2021*, Melbourne, Australia, 2021.
- 19 EUROFER, Low carbon roadmap pathways to a CO₂ neutral European steel industry, European Steel Association, Brussels, Belgium, 2019.
- 20 IEA, *Technology roadmap: low-carbon transition in the cement industry*, International Energy Agency, Paris, France, 2018.
- 21 IEA, Iron and steel technology roadmap: towards more sustainable steelmaking, International Energy Agency, Paris, France, 2020.
- 22 N. A. Ryan, S. A. Miller, S. J. Skerlos and D. R. Cooper, Environ. Sci. Technol., 2020, 54, 14598–14608.
- 23 CEMBUREAU, Cementing the European green deal: reaching climate neutrality along the cement and concrete value chain by 2050, Brussels, Belgium, 2019.
- 24 M. Bui, C. S. Adjiman, A. Bardow, E. J. Anthony, A. Boston, S. Brown, P. S. Fennell, S. Fuss, A. Galindo, L. A. Hackett, J. P. Hallett, H. J. Herzog, G. Jackson, J. Kemper, S. Krevor, G. C. Maitland, M. Matuszewski, I. S. Metcalfe, C. Petit, G. Puxty, J. Reimer, D. M. Reiner, E. S. Rubin, S. A. Scott, N. Shah, B. Smit, J. P. M. Trusler, P. Webley, J. Wilcox and N. Mac Dowell, *Energy Environ. Sci.*, 2018, 11, 1062–1176.
- 25 G. R. M. Dowson and P. Styring, Front. Energy Res., 2017, 5, 26.
- 26 G. Y. Zang, P. P. Sun, E. Yoo, A. Elgowainy, A. Bafana, U. Lee, M. Wang and S. Supekar, *Environ. Sci. Technol.*, 2021, 55, 7595–7604.
- 27 T. Bruhn, H. Naims and B. Olfe-Krautlein, *Environ. Sci. Policy*, 2016, **60**, 38–43.
- 28 J. Lane, C. Greig and A. Garnett, *Nat. Clim. Change*, 2021, 11, 925–936.
- 29 P. Tcvetkov, A. Cherepovitsyn and S. Fedoseev, *Heliyon*, 2019, 5, e02845.
- 30 P. A. Webley and D. Danaci, in *Rsc Energy Environ S*, ed. M. Bui and N. Mac Dowell, The Royal Society of Chemistry, Croydon, UK, 2020, ch. 5, pp. 106–167.
- 31 D. A. Chisalita, L. Petrescu, P. Cobden, H. A. J. van Dijk, A. M. Cormos and C. C. Cormos, *J. Cleaner Prod.*, 2019, 211, 1015–1025.
- 32 L. Giordano, D. Roizard and E. Favre, *Int. J. Greenhouse Gas Control*, 2018, **68**, 146–163.
- 33 K. Volkart, C. Bauer and C. Boulet, *Int. J. Greenhouse Gas Control*, 2013, **16**, 91–106.

- 34 N. von der Assen, L. J. Muller, A. Steingrube, P. Voll and A. Bardow, *Environ. Sci. Technol.*, 2016, **50**, 1093–1101
- 35 L. J. Müller, A. Katelhon, S. Bringezu, S. McCoy, S. Suh, R. Edwards, V. Sick, S. Kaiser, R. Cuellar-Franca, A. El Khamlichi, J. H. Lee, N. von der Assen and A. Bardow, *Energy Environ. Sci.*, 2020, 13, 2979–2992.
- 36 C. Oberschelp, S. Pfister and S. Hellweg, *Environ. Sci. Technol.*, 2020, 54, 16028–16038.
- 37 C. Oberschelp, S. Pfister, C. E. Raptis and S. Hellweg, *Nat. Sustainability*, 2019, 2, 113–121.
- 38 US Environmental, Protection Agency (EPA), Uncertainty and Variability, https://www.epa.gov/expobox/uncertainty-and-variability, (accessed April, 2022).
- 39 C. Oberschelp, S. Pfister and S. Hellweg, under review.
- 40 L. Rosa, D. L. Sanchez and M. Mazzotti, *Energy Environ. Sci.*, 2021, 14, 3086–3097.
- 41 UNECE, Draft industrial energy efficiency action plan, and assessment of the role of the United Nations Economic Commission for Europe in delivering on it, United Nations Economic Commission for Europe, Geneva, Switzerland, 2022.
- 42 F. Schorcht, I. Kourti, B. M. Scalet, S. Roudier and L. D. Sancho, *Best available techniques (BAT) reference document for the production of cement, lime and magnesium oxide*, The Joint Research Centre of the European Commission (JRC), Luxembourg, Luxembourg, 2013.
- 43 T. Lecomte, J. Ferrería de la Fuente, F. Neuwahl, M. Canova, A. Pinasseau, I. Jankov, T. Brinkmann, S. Roudier and L. Delgado Sancho, Best Available Techniques (BAT) reference document for large combustion plants, The Joint Research Centre of the European Commission (JRC), Luxembourg, Luxembourg, 2017.
- 44 A. Hasanbeigi and C. Springer, *How clean is the U.S. steel industry? An international benchmarking of energy and CO₂ intensities*, Global Efficiency Intelligence, San Francisco, CA, USA, 2019.
- 45 IEAGHG, Techno-economic evaluation of retrofitting CCS in a market pulp and integrated pulp and board mill, IEA Greenhouse Gas R&D Programme, Cheltenham, UK, 2016.
- 46 IEAGHG, Towards zero emissions: CCS in power plants using higher capture rates or biomass, IEA Greenhouse Gas R&D Programme, Cheltenham, UK, 2019.
- 47 IEAGHG, Iron and steel CCS study (techno-economics integrated steel mill), IEA Greenhouse Gas R&D Programme, Cheltenham, UK, 2013.
- 48 K. Meijer, *HIsarna developing a sustainable steel production process*, Tata Steel, 2018.
- 49 A. Kätelhön, R. Meys, S. Deutz, S. Suh and A. Bardow, *Proc. Natl. Acad. Sci. U. S. A.*, 2019, **116**, 11187–11194.
- 50 IPCC, *IPCC guidelines for national greenhouse gas inventories*, International Panel on Climate Change, Hayama, Japan, 2006.
- 51 Quantis, D3.6 Life Cycle Assessment, Project LEILAC, 2021.
- 52 D. Garcia-Gusano, D. Garrain, I. Herrera, H. Cabal and Y. Lechon, *J. Cleaner Prod.*, 2015, **104**, 328–338.

53 H. Lund, B. V. Mathiesen, P. Christensen J. H. Schmidt, Int. J. Life Cycle Assess., 2010, 15, 260-271.

- 54 IEA, World energy outlook 2021, International Energy Agency, Paris, France, 2021.
- 55 Y. Lu, F. Cohen, S. M. Smith and A. Pfeiffer, Nat. Commun., 2022, 13, 806.
- 56 R. Y. Cui, N. Hultman, D. Cui, H. McJeon, S. Yu, M. R. Edwards, A. Sen, K. Song, C. Bowman, L. Clarke, J. Kang, J. Lou, F. Yang, J. Yuan, W. Zhang and M. Zhu, Nat. Commun., 2021, 12, 1468.
- 57 Institute for Health Metrics and Evaluation (IHME), GBD Compare, IHME, University of Washington, Seattle, WA, USA, 2022.
- 58 IEA, Net zero by 2050: A roadmap for the global energy sector, International Energy Agency, Paris, France, 2021.
- 59 S. Budinis, S. Krevor, N. Mac Dowell, N. Brandon and A. Hawkes, Energy Strategy Rev., 2018, 22, 61–81.
- 60 D. J. Heldebrant and J. Kothandaraman, in Rsc Energy Environ S, ed. M. Bui and N. Mac Dowell, The Royal Society of Chemistry, Croydon, UK, 2020, ch. 3, pp. 36-68.
- 61 R. Khalilpour, K. Mumford, H. B. Zhai, A. Abbas, G. Stevens and E. S. Rubin, J. Cleaner Prod., 2015, 103, 286-300.
- 62 IPCC, IPCC special report on carbon capture and storage, International Panel on Climate Change, Cambridge, United Kingdom and New York, NY, USA, 2005.
- 63 National Petroleum Council, Meeting the dual challenge: a roadmap to at-scale deployment of carbon capture, use, and storage, Washington, D.C., USA, 2019.
- 64 F. Sun, J. S. Qin, Z. Y. Wang, M. Z. Yu, X. H. Wu, X. M. Sun and J. S. Qiu, Nat. Commun., 2021, 12, 4182.
- 65 IEA, Electricity Information: Overview, International Energy Agency, 2021.
- 66 J. Artz, T. E. Muller, K. Thenert, J. Kleinekorte, R. Meys, A. Sternberg, A. Bardow and W. Leitner, Chem. Rev., 2018, **118**, 434–504.
- 67 Y. Khojasteh-Salkuyeh, O. Ashrafi, E. Mostafavi and P. Navarri, J. CO2 Util., 2021, 50, 101608.
- 68 J. Fernández-González, M. Rumayor, A. Domínguez-Ramos and A. Irabien, Ind. Eng. Chem. Res., 2022, 6163-6172.

- 69 S. K. Nabil, S. McCoy and M. G. Kibria, Green Chem., 2021, 23, 867-880.
- 70 Y. Ni, Z. Chen, Y. Fu, Y. Liu, W. Zhu and Z. Liu, Nat. Commun., 2018, 9, 3457.
- 71 J. J. Gao, C. M. Jia and B. Liu, Catal. Sci. Technol., 2017, 7, 5602-5607.
- 72 T. K. Xiong, X. Q. Zhou, M. Zhang, H. T. Tang, Y. M. Pan and Y. Liang, Green Chem., 2021, 23, 4328-4332.
- 73 W. d'Ambrières, Field Actions, Sci. Rep., 2019, 12-21.
- 74 M. Klotz, M. Haupt and S. Hellweg, Waste Manage., 2022, 141, 251-270.
- 75 H. Wiesinger, Z. Y. Wang and S. Hellweg, Environ. Sci. Technol., 2021, 55, 9339-9351.
- 76 R. Meys, A. Katelhon, M. Bachmann, B. Winter, C. Zibunas, S. Suh and A. Bardow, Science, 2021, 374, 71-76.
- 77 C. Lindner, J. Schmitt and J. Hein, Material flow diagram of plastics in Germany in 2019 (in German), CONVERSIO, Mainaschaff, Germany, 2020.
- 78 R. Slade, A. Bauen and R. Gross, Nat. Clim. Change, 2014, 4, 99-105.
- 79 S. Walker and R. Rothman, I. Cleaner Prod., 2020, 261, 121158.
- 80 C. Hepburn, E. Adlen, J. Beddington, E. A. Carter, S. Fuss, N. Mac Dowell, J. C. Minx, P. Smith and C. K. Williams, Nature, 2019, 575, 87-97.
- 81 N. von der Assen, J. Jung and A. Bardow, Energy Environ. Sci., 2013, 6, 2721-2734.
- 82 N. von der Assen, P. Voll, M. Peters and A. Bardow, Chem. Soc. Rev., 2014, 43, 7982-7994.
- 83 N. Mac Dowell, P. S. Fennell, N. Shah and G. C. Maitland, Nat. Clim. Change, 2017, 7, 243-249.
- 84 Scottish Carbon Capture & Storage (SCCS), Petra Nova carbon capture project details, https://www.geos.ed.ac.uk/ sccs/project-info/4, (accessed March, 2022).
- 85 H. C. Mantripragada, H. B. Zhai and E. S. Rubin, Int. J. Greenhouse Gas Control, 2019, 82, 59-68.
- 86 U.S. Department of Agriculture, Coal-power plants rejuvenated with biomass: an economic, social, and environmentally sustainable transition to clean power, Washington, D.C., USA, 2017.