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Environmental impacts of carbon capture, transport, and storage supply chains: Status and the way forward

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ABSTRACT

Carbon capture, transport, and storage (CCTS) enables the decarbonization of industrial emitters. CCTS is regarded as crucial in reaching net-zero emission targets but currently stands far behind the required scale. CCTS deployment for point sources may be accelerated by CCTS chains relying on currently available technology, called pioneering supply chains. In particular, transporting CO_2 in standard containers can be implemented without new transport infrastructure. Pioneering CCTS chains must not cause more emissions than they store to successfully avoid CO_2 emissions. Using life cycle assessment, we show that pioneering CCTS chains emit less CO_2 than they store permanently, demonstrating that CCTS can already today avoid 50 to 70% of point source GHG emissions. This evidence proves robust against uncertainties based on the scarce operational experience in CCTS. Our environmental assessment shows that increasing the capture rate above the assumed 90% is a main lever to increase emissions avoidance of the CCTS chains above 80%. Capturing and transporting the CO_2 causes large shares of the chain's global warming impact as they rely on fossil fuels. Reducing GHG emission intensity of energy supply and switching to pipeline-based transport can reduce global warming and other environmental impacts compared to pioneering CCTS chains. Our analysis shows that pioneering chains can accelerate infrastructure scale-up while successfully storing CO_2 from point sources.

1. Introduction

Reaching net-zero carbon emissions is expected to require significant capacities of carbon capture and storage (CCS) to decarbonize hard-to-abate industry sectors and generate negative greenhouse gas (GHG) emissions to offset residual emissions (Galán-Martín et al., 2021; IPCC, 2022). Around 8 billion tons of carbon dioxide ($\mathrm{Gt\,CO_2}$) need to be permanently stored per year by 2050 to comply with 1.5 °C warming scenarios (IRENA, 2021). Currently, deployment levels are lower by a factor of 200, with global annual storage of around $0.04\,\mathrm{Gt\,CO_2}$. Of the annually stored volume, a substantial fraction is used in enhanced oil recovery (Lyons et al., 2021) to increase crude oil extraction instead of avoiding CO_2 emissions (U.S. Department of Energy, 2023). Closing the gap between existing and required CO_2 management infrastructure requires a fast scale-up (Lyons et al., 2021).

Hard-to-abate point source emissions can be avoided by capturing CO_2 from the flue gas and storing it permanently (Galán-Martín et al., 2021). The IEA defines point sources as hard-to-abate if they emit process emissions or require high-temperature heat (IEA, 2020). Capture from hard-to-abate point sources has lower specific energy demands than direct air capture (IRENA, 2021), which may still be necessary for additional carbon dioxide removal (IPCC, 2022). For point source emitters (e.g., industrial plants), CCS infrastructure usually consists of CO_2 capture and conditioning at the emitter site, transport from the point of capture to the storage site, and geological storage in deep saline aquifers, depleted oil and gas fields, or basaltic formations (IPCC, 2005; Snæbjörnsdóttir et al., 2020; Becattini et al., 2022). CO_2 point sources and sinks are often separated by long distances, possibly even located in different countries (Morbee et al., 2012; Becattini et al., 2022; ETH Zürich, 2022).

Transport plays a key role when designing CO_2 supply chains, bridging the distance and ultimately enabling sequestration. To highlight the

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equal importance of transport, we thereafter refer to such systems as carbon capture, transport, and storage (CCTS) supply chains.

In Europe, the CCTS infrastructure is in a very early stage with only eleven operating facilities, seven of which are at pilot or demonstration scale (Global CCS Institute, 2023). In particular, no large-scale transport infrastructure is available in Europe, in contrast, for example, to the existing pipelines installed in the United States (IPCC, 2005; IEA, 2022). However, a pipeline network requires large upfront investments (Becattini et al., 2022), and stakeholders will likely only build a network if two preconditions are met: (1) enough CO_2 is captured to utilize the capacity of the pipeline and (2) storage sites offer the corresponding capacity to suppliers. From an emitter perspective, capturing CO_2 only seems reasonable if transport to the storage site is available (Reyes-Lúa and Jordal, 2021). The dilemma between installing capture units, developing storage sites, and expanding the transport infrastructure can be seen as a three-sided “chicken-and-egg” problem (Schlund et al., 2022).

The dilemma can be partly resolved by government-funded projects or financial incentives to store CO_2 (Martin-Roberts et al., 2021; Schlund et al., 2022). Another solution to the dilemma may come from the technological perspective, where “ready-to-use” transport options establish transport routes before a pipeline network. Transport options are considered to be ready-to-use if they are currently on a high technology readiness level and available for rent or purchase on the market. Transport and other technologies fulfilling this requirement are called “pioneering technologies” in the present study, and CCTS systems relying on them are called “pioneering CCTS supply chains”.

As CCTS supply chains aim to reduce GHG emissions, their life-cycle GHG emissions should not exceed the amount of stored CO_2. To know whether CCTS reduces the global warming impact of a point source, rigorous accounting of emissions through life cycle assessment (LCA) is essential. To detect burden shifting, other environmental impacts should be investigated (Müller et al., 2020). Many previous LCA studies on the topic of CCTS chains have established their environmental viability, i.e., that more CO_2 is stored than is emitted: Early work reported that adding CCTS could reduce the life-cycle GHG emissions of cement plants up to 78% and up to 92% for various fossil power plants (Volkart et al., 2013). Similar reductions of global warming impact were found when assessing other point sources. For example, CCTS at waste-to-energy (WtE) plants reduces their GHG emissions significantly and can even produce negative emissions (Pour et al., 2018; Bisinella et al., 2022).

Although the potential of CCTS to reduce life-cycle GHG emissions has been shown for various applications, most assessments have focused on fully-developed CO_2 transport infrastructures, mostly via pipelines, without assessing the transition to such infrastructure within the context of the aforementioned “chicken-and-egg” problem (Koornneef et al., 2008; Volkart et al., 2013; Negri et al., 2021; Terlouw et al., 2021; Bauer et al., 2022). The widespread assumption of existing pipelines results in a relatively small global warming impact connected to transport. Thus, its climate impact has often been reported together with permanent storage and estimated between 1 and 10% (Zapp et al., 2012). While these assessments quantify the impacts of large-scale, and mostly pipeline-dependent CCTS chains, their conclusions do not hold for a CCTS system deployed today. Installing pipelines is not a solution for the near term as it is challenging, not only in the densely populated areas of Europe (Wuppertal Institut, 2018) but also in more remote regions such as Alberta in Canada (Enhance Energy Inc., 2015). Considerations of nature conservation areas, acceptance in local communities, and acquiring the approval of land-owners may extend already long planning and construction times (Enhance Energy Inc., 2015; Wuppertal Institut, 2018). Repurposing existing natural gas pipelines may offer an alternative to constructing new ones. However, due to differences in operating pressure and corrosion resistance, the re-employment for CO_2 transport requires extensive maintenance and is not a short-term solution (Onyebuchi et al., 2018). Ostovari et al. (2022) present an alternative scenario to the pipeline assumption: The authors have considered the potentially problematic installation of pipelines in their study and assessed truck-based transport as an alternative. Their results show that truck transport over 325 km reduces the GHG mitigation potential of their CCTS chains by 1.5%. However, their assessment focuses on producing stable carbonates as a product for which only limited demand and no large-scale industrial plants exist (Ostovari et al., 2022). Other authors not considering pipelines resort to large-scale, dedicated CO_2-ships as alternatives (Bisinella et al., 2022), which have not been demonstrated yet (IEA, 2022).

The distances considered in many studies also reflect a more mature landscape of CCTS in Europe: The assumed transport distances are considerably smaller than in most announced projects. The IPCC considers a transport distance of 300 km to be economically reasonable (IPCC, 2005). In line with the IPCC recommendation, Wei et al. (2021) suggested a global layout for CCTS in line with a 2 °C climate target, where 80% of all CO_2 sources worldwide can be connected to a storage site within 300 km (Wei et al., 2021). Other authors use the same order of magnitude for transport distances (Volkart et al., 2013; Bauer et al., 2022). However, early CCTS projects will likely need longer source-to-sink distances due to the currently limited options for permanently storing CO_2 underground and the heterogeneous distribution of CO_2 sources and sinks (Morbee et al., 2012; d’Amore and Bezzo, 2017; d’Amore et al., 2021; Becattini et al., 2022). Authors investigating specific case studies, e.g., Bisinella et al. (2022), report significantly longer transport distances due to the lack of feasible storage sites within the 300 km suggested by the IPCC (2005). We therefore see the need to quantify the environmental impacts of pioneering CCTS chains, to kick-start developing and deploying a global CCTS infrastructure.

To this aim, we assess four pioneering CCTS chains relying entirely on currently available technologies and covering large distances in distinct locations across Europe. We cover a variety of hard-to-abate sectors by considering two cement, one WtE, and one pulp & paper plant. All considered technologies, the emitter locations for capture, and the storage site have been selected based on the precondition that they are currently available and interested in implementing CCTS operations (ACCSESS, 2022; ETH Zürich, 2022). Following the definition of *pioneering technologies* above, the transport is assumed to be based on standard containers. Standard containers for transporting liquefied CO_2 are widely used throughout the food and beverage industry and thus readily available (Meeberg Group, 2022). For point sources that aspire to establish CCTS chains soon, the most straightforward approach is a single-source single-sink supply chain directly connecting the capture and the storage sites (Morbee et al., 2012). The present study considers single-source single-sink supply chains, evaluates the ability of each chain to avoid CO_2 emissions from point sources, and quantifies its respective CO_2 avoidance efficiency. To limit burden shifting, impacts in other environmental areas are evaluated. By analyzing multiple CCTS chains under uncertainties due to their limited operational experience, we can resolve the challenges of deploying CCTS chains with different transportation types, point sources, and European locations.

2. Pioneering chains

The four CCTS pioneering chains are described in the following and result from a techno-economic assessment of several transport chain options. A shortest-path algorithm determined the minimum-cost route and transport option (Oeuvray et al., 2022).

The system boundary of each CCTS chain includes five main stages (Fig. 1): (1) the capture unit, (2) the conditioning unit, (3) temporary storage at the emitter site, (4) transport from the point source to the storage site *Northern Lights*, and (5) permanent storage in the Northern Lights offshore storage. The working principle and underlying assumptions of the capture and conditioning stages do not differ between the four chains, and they are once described in Section 2.1. The same applies to the geological storage described in Section 2.3.

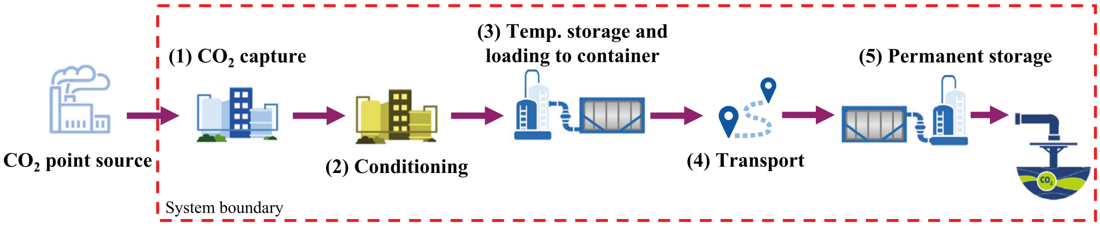

Fig. 1. Schematic of employed system boundary for pioneering CCTS chains with permanent storage in offshore aquifers.

2.1. Carbon capture and conditioning at the emitter sites

The design of the capture and conditioning units follows the same principles for all four chains, although all capture units differ slightly as they are adapted for each emitter to minimize the energy consumption per unit of CO_2 captured. Rigorous mass and energy balances are obtained through process simulations for the CO_2 capture and the CO_2 conditioning processes.

For each point source, the flue gas containing CO_2 enters the system boundary after being emitted and before flowing into the capture unit (Fig. 1). The CCTS chains use the amine-based capture process configuration described by Voldsund et al. (2019). The study also describes the oxyfuel process, offering best energy performance, and calcium-looping technologies with the highest emission abatement potential. However, the retrofitability is highest for post-combustion systems (Voldsund et al., 2019). The focus of the study are pioneering CCTS chains that offer the least complex retrofit to minimize production disruption due to an already high investment. With extensive experience in installing post-combustion-based CO_2 capture units (IEA, 2023), the amine-based technology is considered the most suitable for pioneering CCTS chains. The capture stage uses an aqueous 2-amino-2-methyl-1-propanol (AMP) solvent promoted with piperazine (PZ). The AMP-PZ mixture is shown to be more energy efficient than monoethanolamine, the solvent often used as reference (Feron et al., 2020; Biermann et al., 2022). A 90% capture rate serves as a conservative estimate already achieved in commercial-scale projects (Kennedy, 2020) and is widely accepted as a benchmark (Bui et al., 2018).

A standard absorber-stripper process configuration is considered, following the work of Voldsund et al. (2019). The flue gas is sent to the bottom of the absorber column, flowing counter-current to the solvent, into which the CO_2 dissolves physically and then reacts with the amines. The CO_2-depleted gas passes through a water wash section at the top of the absorber column to recover solvent and thus reduce emissions. The CO_2-rich solvent is sent to the top of the stripper (desorber column), where steam generated in a reboiler regenerates the solvent and produces purified CO_2.

The purified CO_2 leaves the top of the stripper and is sent to conditioning, while the lean amine solvent is recycled to the absorber column. A lean-rich heat exchanger preheats the CO_2-rich solvent stream from the absorber using the heat in the CO_2-lean solvent stream from the desorber. The chosen solvent composition is 33 wt% AMP - 12 wt% PZ, while the mass ratio of lean solvent and gas flow rate is between 2.2 and 2.9, depending on the optimal value identified for each pioneering chain. The geometrical sizes of the absorber and desorber are selected as a compromise between energy efficiency and capital expenditures while complying with operational constraints (e.g., avoiding flooding).

The AMP-PZ capture process is simulated in Aspen Plus V10 with a rate-based modeling approach. The model is tuned to represent the behavior of the selected solvent, including selected interaction coefficients, equilibrium constants, and kinetic parameters. The model is validated against experimental data within the range of solvent operating temperature reported by Brúder et al. (2011). The capture process simulation provides energy and mass flows for the LCA, including solvent makeup, heating, cooling, and electricity demands.

The standard configuration of all capture units in this study uses a power-and-heat co-generation unit powered by natural gas to supply heat. Despite their carbon footprint, fossil fuels are chosen for energy supply since the technology is readily available and can be deployed instantaneously. Heat supply solutions with lower GHG intensity, such as biomass boilers, are investigated as alternative heat supply scenarios in Section 4.2. The CO_2 stream leaves the capture unit at ambient temperature and pressure to be compressed and liquefied in the conditioning stage.

The capture unit already provides a high-concentration CO_2 stream (~98 mol% CO_2). After conditioning, the CO_2 stream fulfills the purity requirements of the Northern Lights project for transport and permanent storage (Equinor, 2019a; Northern Lights JV DA, 2022). The conditioning process includes a multi-stage CO_2 compression train, followed by liquefaction with a refrigeration cycle that uses ammonia (NH_3) as a working fluid, as described by Deng et al. (2019). The CO_2 leaves the conditioning stage in liquid form at 15 bar.

The conditioning process is simulated in Aspen HYSYS V10. The Peng-Robinson equation of state is used to calculate the thermodynamic properties and vapor–liquid equilibrium of CO_2 and CO_2 mixtures. The process design is optimized by means of an internal SQP-based optimization utility to lower the overall specific energy consumption of the system while meeting the CO_2 pressure and purity specifications. Conditioning removes potentially harmful impurities and liquefies the CO_2. The critical impurities to handle are water and oxygen. Water is removed between the compression stages with knock-out drums, and the $\leq$30 ppmv specification is assured using a Temperature Swing Adsorption dehydration unit downstream of the compression train. The $\leq$10 ppmv specification for oxygen is reached by purging a small portion of uncondensed gas after the recirculation flash, which also causes a small fraction of CO_2 to be vented to the environment. The simulation results provide data on the purge stream flow rate and composition, as well as cooling water and electricity requirements to the LCA.

The capture and conditioning simulations also provide the basis for the equipment size, material, and area requirements. The steel demands for the process equipment are obtained from Aspen Process Economic Analyzer. The total steel weight used as input to the LCA considers redundancy for equipment such as pumps and heat exchangers. The concrete demand is estimated considering the total equipment area requirement.

As pipelines that can transport large amounts of CO_2 in a continuous flow have yet to be built in Europe, batch-wise transport is assumed. Due to the discontinuous nature of batch-wise transport, temporary storage tanks are required for buffering liquefied CO_2 at each point source. From the storage tanks, the CO_2 is discharged to a standard container whenever one is available on site. Details regarding the temporary storage can be found in the Supplementary Information.

2.2. Point sources and transport

The four assessed point sources consist of two cement plants, one in Germany (*HM Hannover*) and one in Poland (*HM Górażdże*), a pulp and paper plant in Sweden (*SE Skutskär*), and a WtE plant in Switzerland (*KVA Linth*). The setup of each route from the point sources

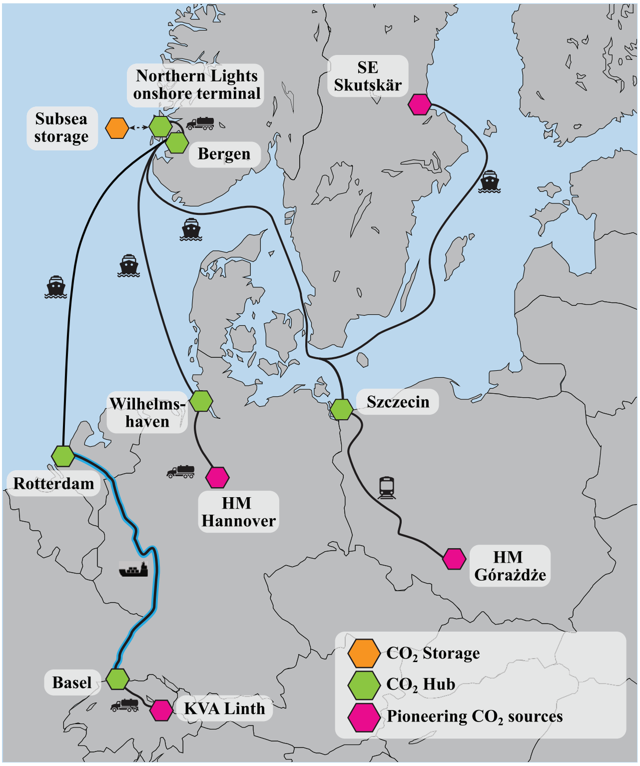

Fig. 2. Map of the four pioneering CCTS chains from the point sources to the storage site. The individual transport modes are depicted, but distances are only approximately to scale. Illustration: SINTEF.

to permanent storage differs between the emitters (Fig. 2). All emitters and the relevant details of the transport stage are briefly described in the following in order of increasing distance to the storage site:

The cement plant *HM Hannover* in Germany, owned by the company Heidelberg Materials, produces GHG emissions from burning fuel for the heat supply to the process. Additionally, the process itself releases CO_2 during the clinker-forming reaction, contributing approximately 60% to the total direct CO_2 emissions of a cement plant (Verein Deutscher Zementwerke e.V., 2021). Clinker is the cement's base material, which will continue causing CO_2 emissions, even with a decarbonized heat supply. The process cannot be fully decarbonized; thus, CCTS is inevitable for net-zero cement production (Fennell et al., 2022). As the majority of CO_2 emissions result from the process itself and not from energy use, the cement sector is unlikely to substantially reduce CO_2 emissions without relying on CCTS systems (Fennell et al., 2022). Hence, the cement sector is included in the scope of our analysis. The cement plant in Górażdże, called *HM Górażdże*, also belongs to Heidelberg Materials. It has the same working principle and thus represents the same type of CO_2 sources as the plant in Hannover but allows us to resolve the impact of different transport modes and countries.

The Swedish pulp and paper plant *SE Skutskär* produces pulp from wood in an energy-intensive process called the "kraft pulp" process (Suhr et al., 2015). Kraft pulping emits CO_2 from several sources: First, heat is required in recovery boilers where processing chemicals are recovered. The recovery boilers are fired with wood residues or biogenic fuels generated from the pulp mill process. Second, another recovery process uses lime and forms calcium carbonate. Regenerating the calcium carbonate back to lime requires high-temperature heat and emits CO_2 from the carbonate. Detailed descriptions of the kraft pulp process can be found in the literature (Bajpai, 2018).

The waste-to-energy plant *KVA Linth* in Switzerland serves several purposes: (1) municipal solid waste is collected from the region surrounding the plant and incinerated to reduce its volume for landfill; (2) the incineration drives a steam cycle to generate electricity that is fed into the power grid, and heat, which is fed into a district heating system. An additional CCTS supply chain is assessed from a second WtE plant (*KVA Hagenholz*) in Zurich, Switzerland. As KVA Hagenholz is located within 50 km of KVA Linth, the differences in the supply chain setup are minor, and thus, its description and results are only presented in the Supplementary Material, D.

As the transport will be realized in the short term, we assume using standard CO_2 containers for the entire route. The transport vehicles are established ships, barges, trains, and trucks used for standardized cargo containers, and thus, the vehicles can be purchased or rented from existing manufacturers at any time. Each container can carry approximately 20 t of CO_2. While some dedicated transportation vessels exist for CO_2, such as trucks (Asco Carbon Dioxide LTD, 2021) and railway carriages (VTG, 2023), containers have certain advantages: They are available immediately as they have been used in the food and beverage industry for many years. The infrastructure to transfer standardized shipping containers from one transport vehicle to another is widely available in harbors and other freight transportation hubs. No additional infrastructure must be built for intermediary storage or transfer of CO_2 along the transport route as the containers serve as intermediate storage. The existing facilities for container handling can be used, facilitating a sooner start of operation of the transport chain and less upfront investment. Therefore, temporary storage tanks are installed only at the point source site but not at subsequent transfer locations to minimize the installation effort required. After unloading, all containers must be returned to the point source, to fill them again. Therefore, the described chains transport containers in both directions, once filled with CO_2 and once almost empty. Return transport uses the identical vessels and routes as outward transport.

The transport routes are chosen from a selection of potential transport routes that are expected to operate within one decade. The cost-optimal routes from each point source are determined via a minimum cost path algorithm based on a techno-economic assessment of the transport modes and their operation (Oeuvray et al., 2022). The chosen transport modes, fuels, transshipment locations, and exact distances are reported in Table 1. The transport routes from all point sources to the storage site in Norway are shown in Fig. 2. The transport modes are highlighted, and the approximate routing of the sections is shown.

2.3. Geological storage

The only storage facility currently in Europe that accepts large quantities of CO_2 from external sources is the Northern Lights project in Norway (Northern Lights JV DA, 2022). Other storage sites are actively being developed but are expected to start their operations well after the Northern Lights site (Adomaitis and Kartit, 2023). Thus, all assessed pioneering CCTS chains deliver their CO_2 to Northern Lights. The Northern Lights project will receive CO_2 from suppliers at the onshore terminal on the coast of Norway. Completion and start of injection are expected for 2024 when the first injection phase commences. The first phase of injection foresees 1.5 Mt of CO_2 injected per year (Northern Lights JV DA, 2022). Phase one is also considered for the pioneering chains; thus, all environmental impacts of Northern Lights are allocated to the respective injection capacity. The cumulative amount of CO_2 from the pioneering CCTS chains exceeds the injection capacity of Northern Lights in the first phase. However, it is assumed that the pioneering CCTS chains may trigger the next phase of the Northern Lights project with its expansion to 5 Mt/y injection capacity (Reyes-Lúa et al., 2021; Northern Lights JV DA, 2022)

After arrival, the containers are unloaded, and the liquid CO_2 is pumped into temporary storage tanks that are part of the onshore terminal (Equinor, 2019b). From the onshore facility, the CO_2 flows through an offshore pipeline to the subsea facility, where the CO_2 is injected into the geological formation. Final storage depth is at around 2660 m (Equinor, 2019b; Northern Lights JV DA, 2022). The previously

Table 1
Distance and fuel type of the transport sections to the Northern Lights onshore facility of all CCTS value chains. Note that the truck is distinguished between the Norwegian one from Bergen to Northern Lights (denoted “NO”) and the one in the country of the point source if present (without label). The transport routes are chosen for the lowest cost among a selection of feasible routes.

		Starting point (distance per section in km)			
Vessel[a]	**Fuel**	**HM Hannover**	**HM Górażdże**	**SE Skutskär**	**KVA Linth**
Truck	Diesel	Hannover (253)			KVA Linth (144)
Train	Electricity		Górażdże (453)		
Barge	Diesel				Basel (853)
Ship	Ship oil	Wilhelmshaven (800)	Szczecin (1118)	Skutskär (1790)	Rotterdam (993)
Truck (NO)	Diesel	Bergen (51)	Bergen (51)	Bergen (51)	Bergen (51)
Total distance in km		1104	1571	1841	2041

[a] All vessels are container-bearing vehicles.

developed CO_2 injection sites in the North Sea, Sleipner, and Snøhvit, showed no migration of CO_2 from the storage during their operation (Furre et al., 2019). For the Northern Lights storage, similar behavior of the CO_2 is assumed, and leakage is thus considered to be negligible. Therefore, the CO_2 is assumed to be permanently stored and does not leave the system boundary after its injection into the underground.

3. Life cycle assessment of pioneering CCTS chains

The life cycle assessment (LCA) conducted for the four pioneering chains follows the principles and rules of the International Organization for Standardization (ISO) (DIN EN ISO 14040:2021-02, 2021; DIN EN ISO 14044:2021-02, 2021). According to the standards ISO 14040:2021 and ISO 14044:2021, an LCA is divided into four phases: goal and scope definition, life cycle inventory analysis, life cycle impact assessment (LCIA), and interpretation.

3.1. Goal and scope definition

The LCA of the pioneering CCTS chains in Europe determines whether they avoid more GHG emissions through capture and sequestration than they cause during their life cycle from cradle to grave. The CO_2 avoidance efficiency quantifies the environmental performance of the CCTS chains. The CO_2 avoidance efficiency η_{avoid} is defined as the net amount of CO_2 the CCTS chain avoids per amount of CO_2 stored when considering all life-cycle GHG emissions of the value chain:

$$\eta_{\text{avoid}} = \frac{m_{\text{CO}_2,\text{stored}} - CC_{\text{chain}} - m_{\text{CO}_2,\text{uncaptured}}}{m_{\text{CO}_2,\text{stored}}} \qquad (1)$$

with m_{CO_2} being the mass of CO_2 stored and uncaptured respectively. CC_{chain} is the climate change impact of the CCTS chain. Net avoidance of CO_2 emissions requires a positive CO_2 avoidance efficiency. Note that the CO_2 avoidance efficiency uses the amount of CO_2 stored as reference. The avoidance efficiency is more conservative than the carbon capture efficiency sometimes employed in literature (e.g., Deutz and Bardow (2021)) by treating the uncaptured CO_2 as GHG emissions (see discussion in Section 5).

The LCA aims at supporting CCTS-related decision-making regarding the near-term realization of CCTS chains. Since the CCTS chains not only release GHG emissions but also affect the environment in further ways, this study adopts LCA principles to assess additional categories in which impacts could arise to discuss their causes and potential mitigation options. With little experience deploying CCTS chains, the eventual design and operation are uncertain. The effect of the design and operational uncertainty on the environmental impacts is studied with a sensitivity analysis.

The LCA is conducted as a cradle-to-grave analysis of the CO_2, including all involved processes and equipment. A point source’s flue gas is considered the “cradle” of the CO_2. Permanent geological storage is the “grave” of the CO_2 stream. Viewing the chain as a process to treat the CO_2 also suggests using the functional unit tonne of CO_2. Specifically, we base all flows and impacts on the amount of CO_2 that is permanently stored underground, i.e., the functional unit is 1 *tonne of CO_2 stored* ($t_{\text{CO}_2\text{–stored}}$). Such a functional unit allows the comparison of the four evaluated chains even though their emitters serve different purposes.

In LCA, the background system includes the processes not modeled within the system boundary. For most of the background system, the LCA study relies on the Ecoinvent database, version 3.8, system model *cut-off by classification*. The *cut-off* system model is recommended by Ecoinvent for use in systems where recycling processes are expected to play a minor role (Wernet et al., 2016). Chemicals used in the production processes of the CO_2 capture solvents are taken from the CarbonMinds Database (Stellner et al., 2022), as they were not available from Ecoinvent.

The foreground system of the LCA includes the processes specifically modeled for the CCTS value chains. As described in Section 2.1, the capture and conditioning models stem from process simulations conducted within the research project ACCSESS (2022) and are built on literature (Deng et al., 2019; Voldsund et al., 2019; Biermann et al., 2022). The transport chains are based on background models from Ecoinvent (Wernet et al., 2016) and industry information regarding the containers (Meeberg Group, 2022). The permanent storage model follows the impact assessment of Northern Lights (Equinor, 2019b). More details regarding the inventories of the foreground models are included in the Supplementary Information A and B.

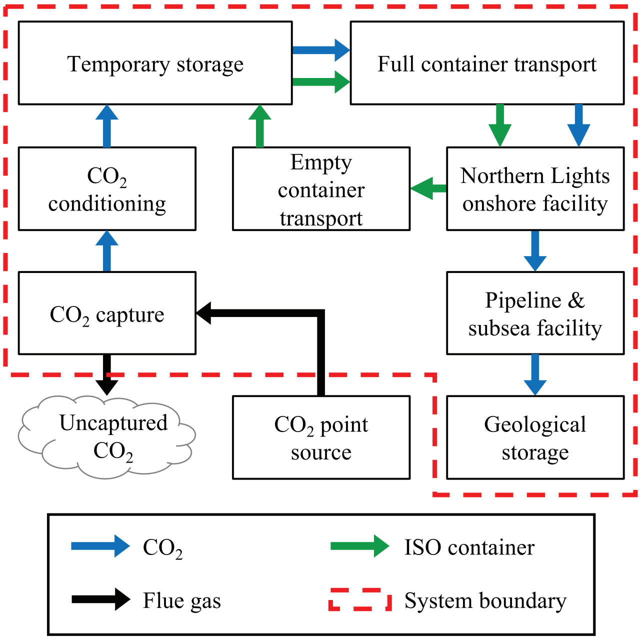

Fig. 3. System boundary of the life cycle assessment and all included processes for a general pioneering CCTS chain irrespective of the point source.

Table 2
Parameters of the sensitivity analysis and their range for optimistic and pessimistic cases. Details regarding the determination of optimistic and pessimistic ranges are included in the Supplementary Information A.5.

Category	Varied parameters	Optimistic	Base case	Pessimistic
Electricity	Demand of all stages except transport	−15%	–	+20%
Capture	Heat demand	−15%	–	+20%
	AMP demand	−20%	–	+30%
	PZ demand	−20%	–	+30%
AMP production	Material demand per kg AMP produced[a]			
	Hydrogen	0.068 kg	0.071 kg	0.085 kg
	2-Nitropropane	1 kg	1.1 kg	1.56 kg
	Formaldehyde	0.34 kg	0.37 kg	0.53 kg
	Energy demand per kg AMP produced[a]			
	Steam	0 MJ	7.7 MJ	21.7 MJ
	Electricity	0 MJ	0.6 MJ	1.58 MJ
	Fuel	0 MJ	0.15 MJ	0.65 MJ
Temporary storage	Buffer time	3 days	5 days	7 days
Transport	CO_2 remaining in container	0%	4%	10%
	Fuel/electricity demand	−14%	0%	+17%
Leakage	CO_2 leakage from conditioning	0%	0.33% – 1.61%	2%
	CO_2 venting from containers per 1000 km	0%	0%	2.5%

[a] All three subparameters are varied together in one sensitivity.

Following the recommendation by the European Commission (2021), we use the "Environmental Footprint" version 3.0 (EF 3.0) environmental impact categories. The global warming impact is calculated based on the global warming potential of an elementary flow over 100 years, the time horizon the European Commission recommends (European Commission, 2021). For impacts beyond climate change, an additional 15 categories are included in the analysis. All impact categories are categorized into three levels of recommendation: Level I categories have the best quality and are considered "recommended and satisfactory", level II categories are "recommended but in need of some improvements", and level III impacts are "recommended, but to be applied with caution" (EC-JRC-IES, 2011). No process included in the foreground model of the CCTS chains has a multi-product output. CO_2 is considered to be a product without economic value and thus is not attributed any burden from the point source. The point source itself is excluded entirely from the system boundary of each CCTS chain (Fig. 3), and all of its impacts are attributed to the commercial product. Therefore, multi-functionality is not an issue in this analysis.

3.2. Sensitivity analysis

The effect of parametric uncertainty on the LCA results is evaluated in a univariate sensitivity analysis, where upper and lower bounds are identified for the most relevant data and propagated through the LCA model. The analysis serves two purposes: First, the relevance of individual parameters for the LCA results is assessed. The outcome shows which parameters greatly affect the environmental impacts and should be investigated for further improvement. Second, the design and operation of pioneering CCTS chains are still uncertain as very little operational experience exists, and this uncertainty propagates to their environmental impacts. The combined effect of all uncertain parameters is used to estimate the overall uncertainty. The overall uncertainty is represented in a best- and worst-case scenario. As the uncertain parameters are interdependent, the best- and worst-case scenarios are not simply the sum of uncertainties. The scenarios represent the parameters' combined effect and allow for deriving an upper and lower bound of the environmental impacts.

For the sensitivity analysis, parameters that are expected to have a large impact on environmental performance or high uncertainty are chosen. Specific uncertainty distributions for the parameters are not yet available due to the limited operational experience. Therefore, base case, optimistic, and pessimistic values are defined for each parameter (Table 2) to showcase the full range of potential impacts. The ranges for optimistic and pessimistic values are determined based on known uncertainties where possible or chosen following best practices for evaluating low-maturity technologies (Christensen and Dysert, 2005; Langhorst et al., 2022). The Supplementary Information contains details about the uncertainty estimation.

Permanent storage is expected to have little effect on the global warming impact of the CCTS chain, as has been shown in previous publications (Volkart et al., 2013; Terlouw et al., 2021). Thus, its electricity demand is the only included uncertainty. For the production of AMP, all material and energy demands are aggregated into one material and one energy cluster, respectively, which is varied as a whole. The parameters and their sensitivity ranges are shown in Table 2.

The parameter ranges result in 11 independently varied parameters, each with an optimistic and pessimistic scenario, used to assess each parameter's impact on global warming impact. The overall uncertainty of the parameters is assessed by integrating the optimistic and pessimistic values of all parameters into one best- and one worst-case scenario, respectively. Such aggregated scenarios allow for deriving an upper and lower bound of the environmental impacts.

4. Results

For CCTS supply chains, the global warming impact is arguably the most important impact category. The global warming impact is presented in Section 4.1 followed by an analysis of the impact of energy inputs to the chain in Section 4.2. Impact categories besides climate change are shown in Section 4.3, and lastly, Section 4.4 presents the sensitivity analysis.

4.1. Global warming impact and the importance of transport in pioneering CCTS chains

Fig. 4 shows the contributions to the global warming impact for all pioneering CCTS chains. The chains are sorted by increasing transport distance. For all chains, and all uncertain scenarios, the CCTS chains emit fewer GHGs than they store, demonstrating the climate benefit of pioneering CCTS chains. For the 4 pioneering chains, the CO_2 avoidance efficiencies lie between 50 and 70%. The avoidance efficiency describes the net amount of CO_2 that is avoided (cf. Eq. (1)): Per 1000 kg of CO_2 stored, between 500 and 700 kg are effectively avoided compared to the point source's emissions without the CCTS chain. Therefore, all four chains fulfill their purpose of avoiding CO_2 emissions.

The chain from SE Skutskär causes the least GHG emissions with an overall global warming impact of 295 $\mathrm{kg_{CO_2\text{-}eq.}/t_{CO_2\text{-}stored}}$. The most carbon-intensive chain is the one from HM Górażdże with life-cycle GHG emissions of $503\,\mathrm{kg_{CO_2-eq.}/t_{CO_2-stored}}$. A smaller difference exists between the GHG emissions of HM Hannover and KVA Linth with $365\,\mathrm{kg_{CO_2-eq.}/t_{CO_2-stored}}$ and $378\,\mathrm{kg_{CO_2-eq.}/t_{CO_2-stored}}$, respectively. KVA Hagenholz causes $376\,\mathrm{kg_{CO_2-eq.}/t_{CO_2-stored}}$ of life-cycle GHG emissions (cf. Supplementary Information D.2).

The worst-case of the CCTS chains (cf. Table 2) shows that the global warming impact may be considerably higher than the base case for all chains. However, even with the uncertainty, all chains still avoid CO_2, although with lower efficiencies. A detailed assessment of the sensitivity study follows in Section 4.4.

The uncaptured CO_2, i.e., the 10% of CO_2 remaining in the flue gas, also contributes to the total global warming impact of the CCTS chains. Attributing the uncaptured CO_2 to the emissions from the CCTS chain is a conservative choice. As discussed in Section 3.1, the incoming CO_2 has no impacts as a waste stream. Thus, all emissions of the point source are attributed to its product. The uncaptured CO_2 leaving the capture unit with the remaining flue gas should, therefore, also be attributed to the point source rather than the capture unit. Otherwise, double counting of emissions may occur. However, allocating the uncaptured CO_2 to the capture unit is conservative as our study does not discuss the point source.

The conservative assumption has a distinct advantage: the effort of increasing the capture rate directly improves the performance of the CCTS chain (Brandl et al., 2021), therefore leading to an incentive for higher capture rates. Still, capture rates other than 90% are not in the scope of the present study. Consequently, if the uncaptured CO_2 is not accounted for in the avoidance efficiency, no statement about the degree of decarbonization of a point source can be made. Besides double counting, attributing the uncaptured CO_2 to the CCTS chain instead of the point source may also lead to responsibility issues: When the CCTS chain operator gets attributed all impacts from the flue gas, the point source has no incentive to minimize its emissions. Therefore, the presented allocation method only allows to derive conclusions for the CCTS chains and not for the point source.

The described issue of allocating the impact of uncaptured CO_2 arises due to the choice of CO_2 as the functional unit. The issue is not reported in the literature that uses the point source's product as the functional unit, e.g., Volkart et al. (2013) and Bisinella et al. (2022). In the present study, the uncaptured CO_2 is included, but where interesting, the CCTS value chain's GHG emissions without the uncaptured CO_2 are additionally discussed. For example, when excluding the uncaptured CO_2, the global warming impact of the CCTS chains reduces by 22 to 38%, ranging from $182\,\mathrm{kg_{CO_2-eq.}/t_{CO_2-stored}}$ for SE Skutskär to $391\,\mathrm{kg_{CO_2-eq.}/t_{CO_2-stored}}$ for HM Górażdże.

The results highlight the importance of the capture rate for deeply decarbonizing point source emitters. As the uncaptured CO_2 emissions are in the same order of magnitude as the emissions from the most influential CCTS stages (cf. Fig. 4), the capture rate has a similar influence as, e.g., the energy demand of the capture unit.

The capture process, without uncaptured CO_2, causes between 25 and 33% of the climate impact of the full chain. As they are already discussed above, the uncaptured CO_2 emissions are excluded here. The direct and indirect emissions from the natural gas co-generation are responsible for over 70% of the capture stage's impact. Thus, although the CCTS chains show a positive CO_2 avoidance efficiency, the conservative choice of heat from natural gas drastically decreases the efficiency. Due to the large heat demand of the capture unit, the chain efficiency strongly depends on the GHG intensity of the heat supply; the effect of a lower CO_2-intensity of heat is discussed in more detail in Section 4.2.

The remaining emissions of the capture unit are predominantly caused by the chemicals required in the capture process: AMP and PZ are required for stocking the process before start-up and due to degradation of the chemicals during operation. The production of AMP is GHG intensive; thus, a large make-up of AMP due to degradation negatively affects the global warming impact of the capture unit. However, the results apply only to the studied solvent-based capture process and heat supply and might thus differ for other capture processes. Construction, deconstruction, and waste treatment show a minor global warming impact in comparison.

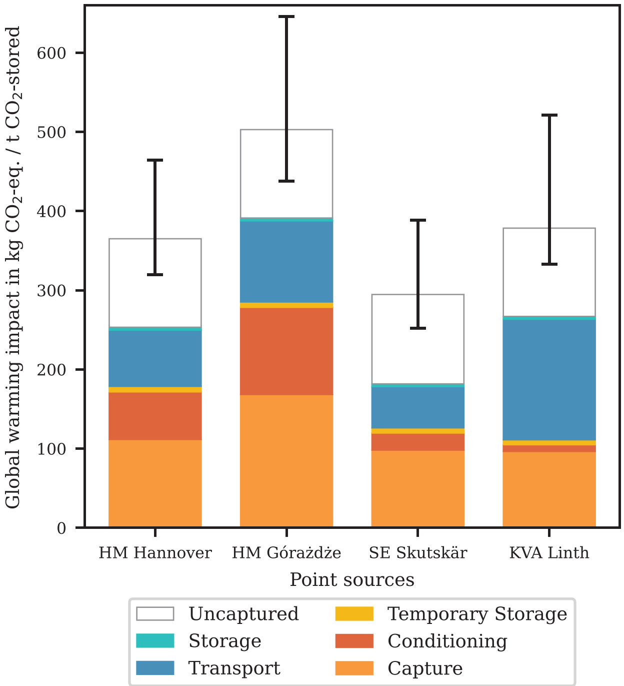

Fig. 4. Global warming impact of permanently storing 1 t of CO_2 for the four investigated pioneering chains in the base case (cf. Table 2), split along the CCTS stages. The chains are sorted by increasing transport distance. The error bars identify the minimum and maximum values of the global warming impact when considering the best- and worst-case scenarios defined in Section 3.2, respectively.

With relative contributions of 16 and 22% for the chains from HM Hannover and HM Górażdże, respectively, conditioning contributes substantially to their total impact. In the chains from KVA Linth and SE Skutskär, conditioning contributes only 2 and 7%, respectively.

The global warming impact of the conditioning unit is dominated by the GHG intensity of the electricity supply. Both the compression and the refrigeration parts of the conditioning are powered by electricity. Poland's power production relies mainly on fossil fuels, including over 70% produced from hard coal or lignite (Ember, 2022a). Consequently, Polish electricity has direct emissions more than 2 times higher than the EU-27 average (Ember, 2022c). Therefore, the conditioning at HM Górażdże shows the highest climate impact out of all assessed locations. In contrast, the Swiss and Swedish grid mixes are among the least carbon-intensive in Europe (Ember, 2022c), such that their conditioning units do not contribute significantly to the global warming impact of the full CCTS chains. Similar to the heat supply, the GHG emissions of the electricity supply influence the overall chain efficiency and are evaluated further in Section 4.2.

Previous studies mostly conclude that transport has only a minor global warming impact compared to other parts of the chain (Volkart et al., 2013; Bisinella et al., 2022). In contrast, the relevance of transport in the pioneering chains is substantial for all chains, ranging between 52 and $152\,\mathrm{kg_{CO_2-eq.}/t_{CO_2-stored}}$ in absolute and between 18 and 40% in relative contribution. Two main reasons are responsible for the large difference in environmental impacts to previous studies: First, the transport chain relies on pioneering technologies instead of pipelines. Most pioneering transport modes rely on the combustion of fossil fuels, having large direct emissions during their use phase.

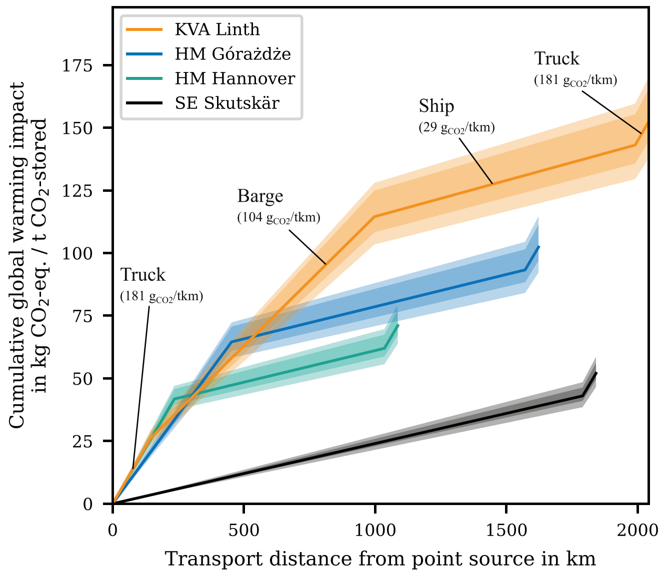

Fig. 5. Cumulative global warming impact for the transport distance of all chains. The shaded areas show the uncertainty from both transport-related parameter ranges in Table 2. The outer shade represents uncertainty from fuel demand and the inner shade from CO_2 remaining in the container. For KVA Linth, the transport modes including their specific carbon-intensity in $g_{CO_2-eq.}$/tkm are given.

The direct emissions of transport may decrease with future transport technologies, e.g., by using ammonia or hydrogen as low-carbon fuels (Al-Breiki and Bicer, 2021). Second, transport distances are several times larger than what is expected in most future scenarios where transport infrastructure and storage sites are widely available.

Often, 300 km or less is discussed in the literature as consensus value (cf. Section 1) (IPCC, 2005; Volkart et al., 2013; Wei et al., 2021). In contrast, the studied pioneering chains have total distances ranging from 1104 to 2041 km, exceeding the IPCC recommendation for economic chains by a factor of almost 7 (IPCC, 2005). Combined, the two effects well explain the large impact of the transport stage.

Even when considering only the pioneering chains, the impact of their transport stages differs strongly: The deviations stem from the transport modes employed for each chain and the differences in transport distance. Fig. 5 shows the cumulative global warming impact per tonne of CO_2 as a function of the source-to-sink distance. The impact of container production is distributed among all transport modes by their respective distance.

The transport chain with the highest absolute global warming impact, originating from KVA Linth, shows a 2.6 times higher impact than the lowest one, which starts at SE Skutskär. However, their transport distances differ only by 11%: 2041 km from KVA Linth and 1841 km from SE Skutskär. The greater dependence on onshore transport of the chain from KVA Linth causes a higher global warming impact. Onshore transport from KVA Linth cannot benefit from container ships' large economies of scale. Consequently, the consumption of fossil fuels per transported weight is higher for onshore than for offshore transport and causes more global warming impact per tonne-kilometer. The effect of direct emissions of onshore transport is evident in Fig. 5 through the steeper slopes of the first two sections, truck and river barge, in the KVA Linth transport chain compared to the ship section. A closer distance to shore favors the respective point sources as large container ships cause less impact than onshore transport modes. In general, the three employed onshore transport modes, truck, train, and barge, in this order, show the largest climate impact per kilometer.

The power supply becomes important for electrified transport: The train from HM Górażdże runs on electricity from the Polish grid, which is largely based on fossil fuels. The emissions of the fossil power plants thus indirectly determine the train's global warming impact per distance, which is almost as large as for the diesel-powered truck adopted for HM Hannover and KVA Linth. A lower carbon-intensity of electricity would reduce the life-cycle GHG emissions of the train section.

Storing CO_2 results in a small climate impact (below 2% for all chains), which is in line with publications that reported minor contributions (Volkart et al., 2013; Bisinella et al., 2021). Noteworthy, temporary storage even causes slightly more global warming (below 3% of total climate impact) than permanent storage, although the latter is arguably more complex and requires more infrastructure. Although the Northern Lights onshore facility also includes small storage tanks, their volume is smaller than the point source temporary storage and is allocated to much larger volumes of processed CO_2. Therefore, the Northern Lights facilities, including storage tanks, cause less global warming impact per tonne of CO_2 stored than the temporary storage installed at the emitter sites.

4.2. GHG intensity of heat and electricity

Heat and electricity are the two major energy inputs for capture, conditioning, and temporary storage at the point source. For capture, the heat is produced from natural gas in the base case resulting in a high GHG impact for the capture unit, irrespective of location. As conditioning is a power-intensive process, the electricity grid mix highly influences its global warming impact. Temporary storage requires electricity, too, although the amount is small compared to conditioning. Heat and electricity from low-carbon solutions can effectively reduce the global warming impact of the CCTS chain. This section evaluates the effect using the point source HM Hannover as an example, since the GHG intensity of Germany's electricity supply is closer to the average European grid mix than at the other locations (Ember, 2022c). The Supplementary Information provides the results for the three remaining pioneering point sources.

Fig. 6 shows the global warming impact of capture, conditioning, and temporary storage, henceforth called "point source stages", at HM Hannover for varying GHG intensities of heat and electricity. The base case with the German electricity grid mix (521 $g_{CO_2-eq.}/\,kWh_{el}$, Wernet et al., 2016) and heat supplied by a natural gas co-generation unit (29 $g_{CO_2-eq.}/MJ_{th}$, Wernet et al., 2016) causes 290 $kg_{CO_2-eq.}/t_{CO_2-stored}$ (red cross in Fig. 6). Switching to biomass-based heat from wood could reduce the climate impact by around 21%. Reaching the same level of global warming impact with another electricity mix would require grid emissions of 94 $g_{CO_2-eq.}/kWh_{el}$, approximately an 80% reduction from current levels.

Decarbonizing heat and electricity both offer meaningful ways to reduce the CO_2-intensity of the point source stages. Heat decarbonization would be more effective than electricity decarbonization: The highest yearly GHG reduction rate of the German electricity mix since 1990 was 13% (Umweltbundesamt, 2022). Assuming a continuous GHG reduction of 13% per year, it would still take more than 13 years to reach the required CO_2-intensity. Decarbonizing the electricity grid does not offer a fast route to reducing the global warming impact of the point source CCTS stages. The finding holds for the capture technology chosen for the pioneering CCTS chains, as amine-based capture requires a lot of heat.

Additional options to reduce GHG emissions from heat supply exist, e.g., (1) the use of heat pumps and (2) capturing the CO_2 from the flue gas of the natural gas co-generation unit. A heat pump links the GHG emissions of heat and power directly to the CO_2-intensity of the electricity grid. With the current grid mix in Germany, however, a heat pump does not reduce the global warming impact compared to using a biomass boiler: GHG emissions increase by approximately 30 to 50% for heat pumps with a coefficient of power (COP) between 5 and 7 (Wernet et al., 2016). However, heat pumps can provide low-carbon heat in a

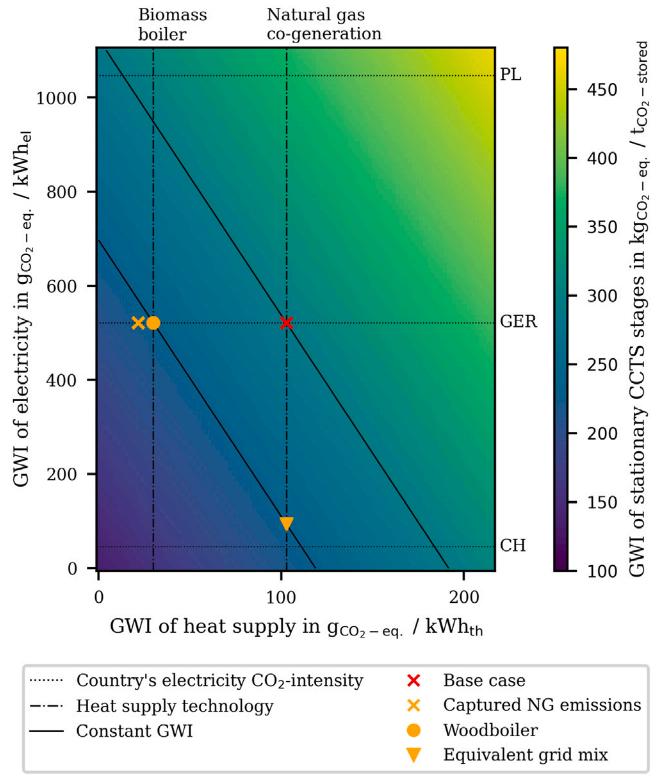

Fig. 6. Effect of GHG intensity of heat and electricity on the global warming impact of CO_2 point source stages for HM Hannover. The values of heat from woody biomass, natural gas co-generation, and captured emissions from natural gas are drawn into the diagram. For selected countries, the GHG intensity of their electricity grid mix is indicated as well (PL: Poland, GER: Germany, CH: Switzerland).

future decarbonized electricity grid in Germany. In countries with GHG intensity of electricity below approximately $145\,g_{CO_2-eq.}/kWh_{el}$, heat pumps (assuming a COP of 7) already have a lower climate impact than heat from a biomass boiler.

Capturing the CO_2 from the flue gas of the natural gas co-generation unit can also reduce the direct CO_2 emissions of the heat supply. At the cement plant in Hannover, this additional capture reduces GHG emissions of the point source stages by 37% (cf. Fig. 6) and emissions of the whole CCTS chain by 18% compared to the base case, assuming all equipment and utilities to scale linearly with CCTS supply chain capacity.

If existing on-site heat generation is available, e.g., at WtE plants, the existing heat supply might (partially) satisfy the heat demand of the capture unit (Pour et al., 2018). However, the heat integration requires additional constructions at the point source and potential process adjustments. More importantly, the heat often already satisfies a heat demand (e.g., district heating). In this case, another heat source would have to substitute the heat supplied to the capture process to continue supplying the original demand. Following the substitution principle of LCA, the environmental impacts of the substitution process are attributed to the CCTS chain. Therefore, the heat supply's impacts depend on the choice of the substitution process. To avoid introducing additional subjectivity into the LCA, the heat integration option is excluded from this study, and we assume that the point source's operation is not impacted by adding CCTS.

However, low-carbon power will eventually be required to reach CCTS chain efficiencies above 90%: Even with fully decarbonized heat, the point source stages of HM Hannover still cause $94\,kg_{CO_2-eq.}/t_{CO_2-stored}$ given the current CO_2-intensity of electricity in Germany. In such a scenario, the electricity demand of compression and liquefaction becomes the determining factor of the global warming impact. Therefore, only heat and electricity supply decarbonization together can provide CCTS chain efficiencies above 90%.

4.3. Other environmental impact categories

The environmental impacts of the pioneering CCTS chains beyond their effect on global warming are analyzed via 16 impact categories in total. The impact categories are based on the Environmental Footprint guidelines, version 3.0 (EF 3.0) (European Commission, 2021) described in Section 3.1. The impacts of the pioneering chains are normalized to the KVA Linth point source, i.e., the chain with the longest transport distance. The results of the chains are compared to each other to identify common trends and differences between locations and chain designs. Fig. 7 presents the impacts of each chain relative to the KVA Linth impacts. The impacts are ranked based on their level of recommendation as stated in the ILCD Handbook by the European Union (EC-JRC-IES, 2011).

The chain from HM Górażdże shows the largest impact among the pioneering chains in 7 out of 16 categories. KVA Linth has the highest impact in 6 categories. Impacts from HM Hannover and SE Skutskär are the largest in two and one categories, respectively. When ranking the chains by the number of categories in which they cause the highest impact, the order is the same as for global warming impact: SE Skutskär only causes a larger impact than the other chains in the category *ionizing radiation: human health* caused by nuclear power in the Swedish electricity mix (Statistics Sweden, 2022). This impact is also found for the chain from KVA Linth as the Swiss power system relies on around 28% nuclear power (Ember, 2022b). Similarly, the large impacts of the HM Hannover and HM Górażdże chains in the category *eutrophication: freshwater* are caused by the respective electricity mix, based on lignite power plants (Statistisches Bundesamt, 2022; Ember, 2022a). Their impacts are around 4 to 8 times higher than those of KVA Linth and represent the only large relative deviation between the four CCTS chains.

For KVA Linth, the transport section dominates the impact categories, which causes more than half of the impact in 11 of the 16 categories. The diesel and ship fuels that are already the main contributor to the transport chain's climate impact also cause the majority of *ozone depletion*, *photochemical ozone formation*, *acidification*, *eutrophication: terrestrial*, and *eutrophication: marine*. These impacts result from the direct emissions from combusting the fuels, such as nitrogen oxides and sulfur dioxide, and the indirect emissions of the production and processing of petroleum. For the category *land use*, the large impacts of transport are not related to fossil fuels: Land use is caused by the trucks and barges relying on human-made transportation networks, i.e., roads and canals, that occupy large areas. Other transport-related impacts appear in the *particulate matter formation* and *human toxicity: non-carcinogenic* categories through particles resulting from wear on tires, brakes, and the road. The transport stage has significant LCA impacts for all chains, but most for KVA Linth, due to its higher reliance on onshore transport.

Besides transport, only the capture stage contributes large shares in multiple impact categories. The largest source of these impacts is the natural gas used for the heat supply. Its production, transport, and combustion cause high shares of impacts on *ozone depletion* and depletion of *energy resources: non-renewables*. Temporary storage only significantly affects the impact in *human toxicity: carcinogenic* and *material resources: metals/minerals* through steel production.

The impact of injecting CO_2 into geological storage is only a notable factor in the category of *water use*. The electricity consumption of the onshore facility and the injection process is based on the Norwegian electricity grid mix. While the power grid has a low GHG intensity, it is primarily powered by hydro energy (Statistics Norway, 2022), which has a relatively large impact on *water use*. However, the impact category *water use* has high uncertainty and is therefore categorized as a level III recommendation. It should be noted that the category

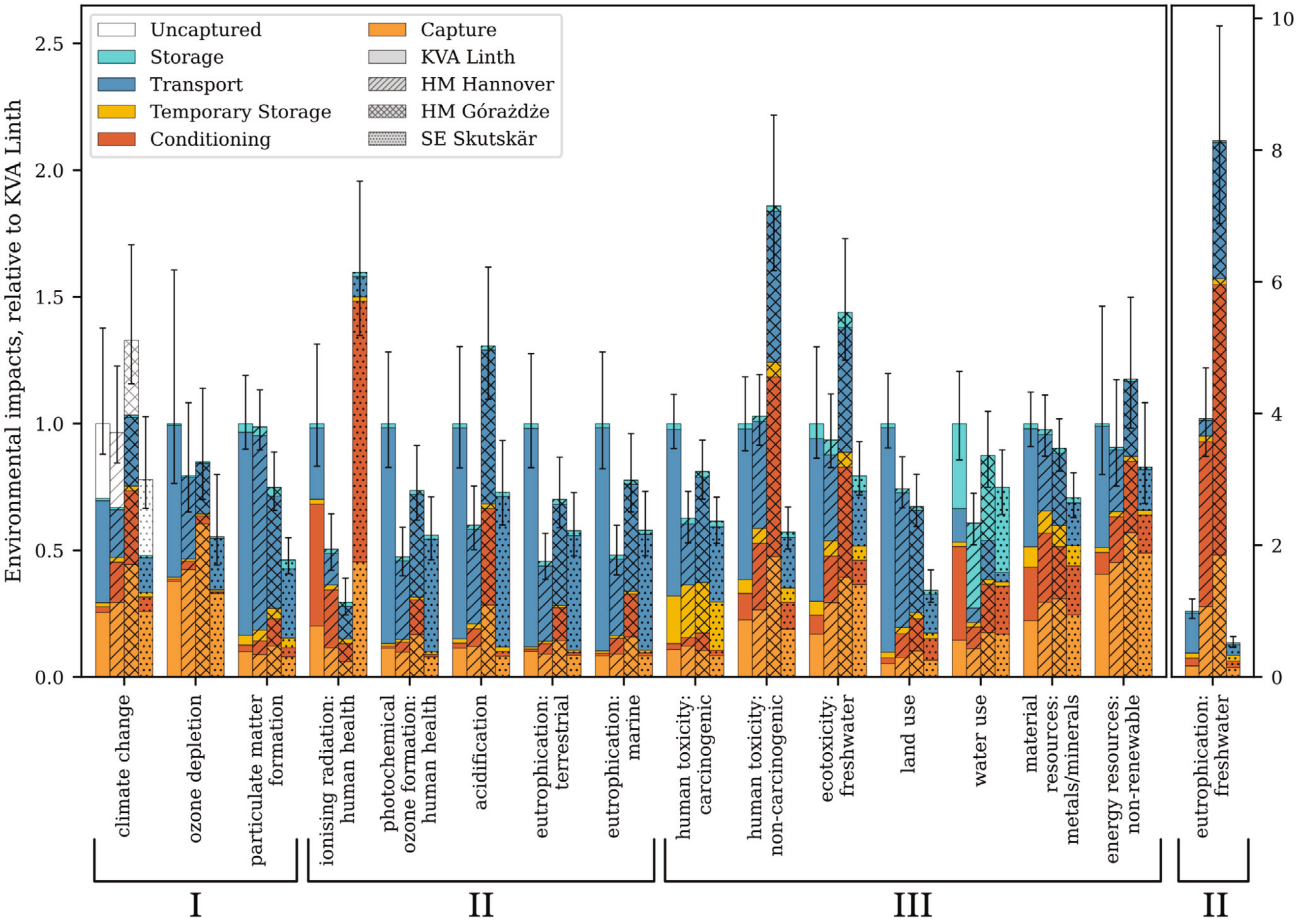

Fig. 7. Environmental impact of the four chains in all assessed impact categories normalized to the impact of KVA Linth. Impacts are split along the stages of the chain and ordered according to their recommendation level (I, II, or III) (European Commission, 2021). The category 'Eutrophication: freshwater' is shown with a separate axis due to its proportions. The error bars represent the cumulative uncertainty stemming from the best- and worst-case parameter ranges identified in Section 3.2.

has been overestimated in previous versions of the Environmental Footprints (European Commission, 2017). In the other categories, the effect of temporary and geological storage seems similarly insignificant as in the climate change category.

Overall, many impact categories closely match the results of global warming impact in comparison between chains and CCTS stages. The primary underlying cause for impacts is fossil fuel mining and combustion, followed by the extraction of resources such as steel.

4.4. Sensitivity analysis

The sensitivity of the results with respect to the parameters introduced in Section 3.2 is evaluated in the univariate sensitivity and uncertainty analysis. Fig. 8 shows the absolute change in global warming impact for all CCTS chains via tornado plots. Following the magnitude of their impact on the chains, the parameter sequence in Fig. 8 comprises a priority list of which uncertainties are relevant in pioneering CCTS chains.

Following that prioritization, CO_2 leakage from the transport step is the most important uncertainty for all analyzed chains. The leakage increases the impact from the base case by 28 to 53 $\mathrm{kg}_{\mathrm{CO_2-eq.}}/\mathrm{t}_{\mathrm{CO_2-stored}}$, and the leakage-induced impact depends only on the absolute transport distance from the point source (cf. Table 1). These results match previous studies, which show that leakage is an influential factor for the global warming impact of transport and storage (Terlouw et al., 2021).

Leakage from conditioning also worsens the climate impact of a chain. Like transport leakage, conditioning leakage counts towards direct emissions from the chains but also reduces the amount of CO_2 stored. When CO_2 is lost to the environment after liquefaction, the impacts of capturing and conditioning still contribute to the global warming impact, although the CO_2 cannot be stored anymore. The chain's total global warming impact is distributed over less CO_2 stored due to the lost CO_2.

Capture is an important factor, with its main impact coming from the heat supply, which is determined by the specific demand for heat and its GHG intensity. If capture requires 20% more heat, the resulting global warming impact increases by 17 to 24 $\mathrm{kg}_{\mathrm{CO_2-eq.}}/\mathrm{t}_{\mathrm{CO_2-stored}}$. However, these results are based on using natural gas co-generation for the heat supply, and less GHG intensive heat would reduce the impact of uncertain heat demand in the capture process.

Similar to the heat demand, the impact of uncertainty in the power demand correlates with the GHG intensity of the used electricity. However, the heat supply shows only minor regional differences in GHG emissions, while discrepancies are large between different regions characterized by different electricity GHG emissions. For example, at KVA Linth, the 20% increase of electricity demand causes an almost negligible increase of less than 1.5 $\mathrm{kg}_{\mathrm{CO_2-eq.}}/\mathrm{t}_{\mathrm{CO_2-stored}}$ on the global warming impact. The chain from HM Górażdże, highly affected by the GHG intensive Polish electricity, shows a 20 times larger increase for the same uncertainty in power demand.

Although all parameters have been varied independently of each other, some are not completely independent: The amount of CO_2 remaining in the container on the return journey affects the transport leakage from venting during the outward leg. Transport leakage stems from pressure increase inside the container, which depends on the heat transfer from the environment to the liquid CO_2. If the CO_2 temperature increase is minor such that the pressure does not exceed the container's limits, transport leakage is negligible. A low temperature can be achieved by keeping some CO_2 in the container, such as in the pessimistic case of the "CO_2 in container" parameter (cf. Table 2). In such a case, the loaded CO_2 does not have to cool down the container when being loaded and remains longer at the conditioning pressure. Both venting and keeping CO_2 in the container cause additional emissions and create a trade-off. Under the assumed uncertainty ranges, keeping some CO_2 in the containers is preferable over accepting venting during transport. However, this finding assumes that leakage is completely prevented when 10% of the CO_2 remains in the container.

The parameters related to the consumption and production of AMP and PZ affect the resulting climate impact the least despite their high uncertainty (cf. "AMP production" in Table 2) due to the limited experience with CO_2 capture.

Overall, the sensitivity analysis identifies critical uncertain parameters for the global warming impact of the CCTS chains: Leakage is the most critical, also due to its twofold effect on the carbon footprint. Besides leakage, energy-related uncertainty in all stages greatly influences the global warming impact of pioneering CCTS supply chains.

5. Discussion

While pioneering chains reduce GHG emissions, their efficiencies are lower than previously analyzed CCTS chains in literature (Volkart et al., 2013; Pour et al., 2018; Bisinella et al., 2022): The achieved efficiencies in the base case, including uncaptured CO_2, range from 50% for the chain from HM Górażdże to 70% for the chain from SE Skutskär. Transport based on containers is one of the main drivers of the global warming impact as it contributes between 26 and 57% of the chain's impact when disregarding the uncaptured CO_2. With containers, economies of scale are limited as the number of containers increases linearly with the amount of captured CO_2, which likely creates issues when fulfilling larger sequestration targets. For those amounts of CO_2, a pipeline network seems inevitable (Becattini et al., 2022; IEA, 2022). Such a network may also resolve scalability issues and the dependency of the CO_2 transport on fossil fuels, depending on the electricity source used for pumping the CO_2 through the pipeline. An intermediate step between container-based transport and pipelines may be dedicated CO_2 transport vehicles, such as tanker barges or rail tank cars for trains (IEA, 2022).

Despite these disadvantages, containers appear to be a viable solution for the near term to break the three-sided "chicken-and-egg" problem from the transport side. The logistics benefit from existing infrastructure at transfer hubs such as harbors, and the availability enables stakeholders to act immediately. Therefore, the containers may not be the preferable long-term solution for reaching large-scale CO_2 sequestration targets, but a bridging solution to accelerate the deployment of CCTS infrastructures.

The transport stage is dominant for the chain's impact on climate change and other categories. Many negative impacts arise from unabated and unfiltered direct emissions from fossil fuels, releasing particles and oxides into the atmosphere. Replacing those fuels is of great importance when trying to reduce the overall environmental impact of the chains. However, while reducing global warming impact, the alternatives to fossil fuels may cause burden shifting to other environmental impacts, which needs to be assessed carefully (Yang et al., 2012). Furthermore, replacing fossil fuels will be challenging in the short term. For onshore transport, electrified railway transport represents an alternative in regions with high shares of renewable energies. However, if no railway infrastructure exists at a particular point source, the CO_2 still needs to be transported to a nearby cargo railway station. Trucks and barges may use electricity (Volvo Trucks, 2023) or low-carbon fuels (Hand, 2023) to lower their emissions, and technology advancements for the decarbonization of maritime shipping are an ongoing research topic (Mallouppas and Yfantis, 2021). However, the availability of such transport vehicles and fuels at scale is currently unclear (Van Grinsven et al., 2021). Relying on recently developed transport modes comes with difficulties, adding to the logistical challenge of establishing the CO_2 chain itself. Assessing transport modes with low technology readiness levels (TRL) from an LCA perspective introduces additional uncertainties (Zimmermann et al., 2022). Integrating future technologies in the form of a prospective LCA (Yousefzadeh and Lloyd, 2021) may offer more insights into the trade-off between installing inferior technologies now or low-carbon solutions in the future.

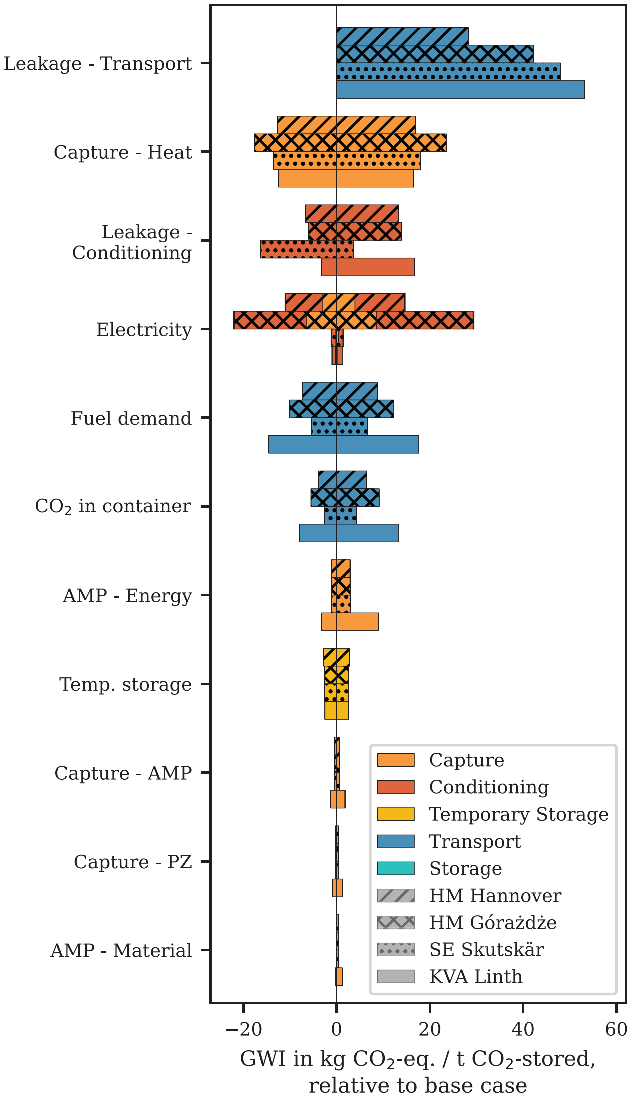

Fig. 8. Impact of sensitivity parameters from Table 2 on the carbon footprint for all CCTS supply chains. The parameters are sorted in descending order of their average additional global warming impact of the pessimistic case.

The safety of transporting CO_2 is discussed in the literature, primarily focusing on transport via pipelines or tanker ships (IPCC, 2005; Di Biagio et al., 2017; Sleiti and Al-Ammari, 2022). However, safety risks also exist for pioneering chains where rapid losses of CO_2 could occur, e.g., in a road accident. Although a sudden CO_2 release is possible during container-based transport, the risk of resulting asphyxiation or intoxication is expected to be negligible due to the limited quantity ($20\,t_{CO_2}$) of CO_2 in each container. The risk may increase where the topography allows the CO_2 to accumulate or if the wind speed is low (Mazzoldi et al., 2008). Ruptures of CO_2 tanks due to the high pressure pose a safety risk besides asphyxiation (Clayton and Griffin, 1994) that needs to be addressed in container design. However, the assumed transport modes are deployed today in the food and beverage industry, and their safety has therefore been assessed and demonstrated. While the additional transport deployed for CCTS would increase the volumes of CO_2 transported, a detailed assessment of the safety risks is not part of this study and will need to be investigated.

When aiming for environmentally-optimal CCTS chains, pipelines are the preferable solution for the transport stage (Gabrielli et al., 2022; IEA, 2022). Pipelines can better handle large volumes (IEA, 2022) and exploit economies of scale. Due to their lower climate impact, pipelines increase the CO_2 avoidance efficiency of CCTS chains. Consequently, less CO_2 must be stored to reach sequestration targets. Until pipelines are deployed, the transport will substantially contribute to the CCTS chains' environmental impact but still enables the avoidance of CO_2 emissions.

Besides the transport stage, considerable improvement potential lies in the energy supply of the CO_2 capture unit. Replacing natural gas co-generation units with wood-fired biomass boilers significantly reduces the carbon footprint of the capture stage. At certain emitter types, e.g., pulp & paper plants, the use of biomass seems more realistic than natural gas, even today, because such plants already process large amounts of wood. However, additional intricacies and challenges appear when switching to biomass, namely, availability issues and correct calculation of the emission balances (Singh et al., 2021; European Commission, 2022; Navare et al., 2022). Wood can serve as long-lived storage of biogenic emissions, and the longer the embedded emissions are stored, the better the effect on global warming (Navare et al., 2022). Such effects must be considered when assessing the environmental impact of biofuels. Point sources emitting biogenic CO_2 also bear the potential to produce negative emissions (Fuss et al., 2018; Rosa et al., 2021). However, negative emissions can only be claimed if the emission counting follows rigorous accounting principles (Tanzer and Ramírez, 2019).

The univariate sensitivity analysis highlights the importance of leakage not only in the transport but also in the conditioning stage: even small absolute amounts of lost CO_2 have a considerable effect. The outcome of the leakage analysis extends to all stages and highlights the importance of having a CO_2-tight CCTS chain. Leakage is a parameter ideally under the control of a point source or CCTS chain operator, and from a technological perspective, avoiding leakage does not depend on external stakeholders. Other contribution factors, for example, the CO_2-intensity of electricity or the production of chemicals, often depend on the respective location or external stakeholders and can only be influenced with enormous additional efforts.

The capture rate is also independent of external stakeholders. Although increasing the capture rate does not directly influence the global warming impact of the actual CCTS chain, it improves the CO_2 avoidance efficiency. On a national or global scale, higher capture rates will also reduce the amount of carbon dioxide removal required to achieve net-zero targets (Dods et al., 2021). The cost increase from increased capture rates above 90% is expected to be less expensive than carbon dioxide removal technologies (Danaci et al., 2021). Therefore, it is important to include the capture rate in the design considerations of the CCTS chain. However, LCAs allocating the uncaptured CO_2 to the CCTS chain must be aware of the potential double counting: Since our analysis assumes that incoming waste CO_2 has no impacts, its GHG emissions are thus assigned to the CO_2 point source. By conservatively treating uncaptured CO_2 as GHG emissions of the CCTS chain, these emissions are double counted. Neglecting the uncaptured CO_2 in Eq. (1) would increase the maximum CO_2 avoidance efficiency of the pioneering CCTS chains from 70 to 81%. This issue shows that proper regulation is required for allocating the uncaptured CO_2 in CCTS chains. As capture rates above 90% are an ongoing research topic (Danaci et al., 2021; Dods et al., 2021), CCTS chains in the future may utilize higher capture rates than analyzed in this study.

Overall, the results of the uncertainty analysis (cf. Fig. 4) show the robustness of the supply chains against uncertainty. Although the CO_2 avoidance efficiency may decrease in the worst-case scenario, all chains still avoid CO_2 emissions. However, the uncertainty analysis does not allow conclusions regarding the probability of the best- and worst-case scenarios. Probability distributions of the parameters included in the sensitivity analysis are not yet available. With more experience in CCTS chains, a Monte-Carlo style uncertainty analysis will provide more insight into the expected variation of environmental impacts and the likelihood of the presented best and worst cases.

6. Conclusions

The developed LCA shows that implementing CCTS chains using existing technologies successfully mitigates greenhouse gas emissions from all assessed point sources, even under worst-case assumptions. The CO_2 avoidance efficiency ranges from 50 to 70%, i.e., storing 1 t of CO_2 avoids 500 to 700 $\mathrm{kg}_{CO_2\text{-eq.}}$ of GHG emissions in the base case. Even under the considered uncertainty, the chains can avoid CO_2 emissions from all point sources, while revealing possible improvements to reach higher efficiencies. A major target identified for improving efficiency is the transport stage for pioneering CCTS chains. Its high global warming impact contrasts the results of previous LCA studies, which have not relied on ready-to-use technologies. The use of fossil fuels and small economies of scale lead to high impacts in global warming and other LCA categories. Switching to pipeline transport may lower global warming and other impacts simultaneously.

Besides transport, CO_2 capture is a major source of GHG emissions in the CCTS chains, mainly from heat supplied through natural gas. For locations with CO_2-intensive electricity, the conditioning unit causes additional high impacts. A sensitivity analysis shows that the improvement potential by reducing the GHG intensity of heat is large.

The capture rate plays an integral role in the CCTS chain's CO_2 avoidance efficiency, which is reduced by the uncaptured CO_2. However, including the uncaptured CO_2 in the chains' emissions potentially leads to counting the respective global warming impact twice: Once as part of the point source emissions and once in the CCTS chain. When excluding the uncaptured CO_2, the CO_2 avoidance efficiency of the pioneering chains increases by 11 percentage points and reaches 61 to 81% CO_2 avoided, highlighting the influence of the capture rate.

The capture and transport stages are major contributors also to impact categories besides climate change. For most categories, fossil fuel combustion for propulsion and heat causes the impact. The remaining categories are highly affected by the power generation technologies of the locations' respective power grids.

Due to limited experience with implementing CCTS chains, uncertainty is high. Individual parameters increase the chains' global warming impact by up to 16% or reduce them by up to 6%. Leakage of CO_2 is found to have a large impact on the GHG emissions of the whole chain. Besides leakage, uncertain energy demands have a large absolute effect if the energy supply has high specific GHG emissions.

The assessed CCTS chains effectively avoid CO_2, even though they rely on pioneering technologies and considerable amounts of fossil fuels. Therefore, pioneering CCTS chains may offer a solution to bridging the gap until a less carbon-intensive infrastructure, including high-capacity pipelines, is available.

CRediT authorship contribution statement

Johannes Burger: Conceptualization, Data curation, Investigation, Methodology, Validation, Visualization, Writing – original draft. **Julian Nöhl:** Data curation, Investigation, Methodology, Validation, Writing – review & editing. **Jan Seiler:** Conceptualization, Methodology,

Supervision, Writing – review & editing. **Paolo Gabrielli:** Conceptualization, Supervision, Writing – review & editing. **Pauline Oeuvray:** Methodology, Validation, Writing – review & editing. **Viola Becattini:** Conceptualization, Project administration. **Adriana Reyes-Lúa:** Methodology, Writing – original draft. **Luca Riboldi:** Methodology, Writing – review & editing. **Giovanni Sansavini:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing. **André Bardow:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The models used to create the life cycle assessment are included in the supplementary information. All required data can be licensed from Ecoinvent and CarbonMinds.

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Appendix A. Supplementary data

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.ijggc.2023.104039.

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