



# Measuring the global warming potential of polygeneration in coal-based hydrogen systems

Diego Hincapié-Ossa<sup>a</sup>, Daniel B. Gingerich<sup>a,b,c,d,\*</sup>

<sup>a</sup> Department of Civil, Environmental and Geodetic Engineering, The Ohio State University, Columbus, OH, 43210, USA

<sup>b</sup> Department of Integrated Systems Engineering, The Ohio State University, 43210, Columbus, OH, USA

<sup>c</sup> Environmental Science Graduate Program, The Ohio State University, 43210, Columbus, OH, USA

<sup>d</sup> The Sustainability Institute, The Ohio State University, 43210, Columbus, OH, USA

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## ABSTRACT

In this paper, we evaluate the potential for *polygeneration* to reduce the climate impacts of a coal-based H<sub>2</sub> system via a series of single-impact life-cycle assessments (LCAs) studying the global-warming potential of H<sub>2</sub>-fuel, electricity, and ammonia production. This allows us to determine carbon capture, usage, and storage (CCUS) requirements to match the relevant benchmark emissions and the potential of polygeneration to reduce requirements. We find that implementing polygeneration substantially reduces CCUS energy requirements: while H<sub>2</sub>,<sub>TH</sub> sole production requires very efficient CCUS systems (6% energy penalty) to outperform uncontrolled natural gas, combining polygeneration with a less efficient CCUS (23% penalty) produces low-carbon H<sub>2</sub> and NH<sub>3</sub> (<2.5 kg<sub>CO2e</sub>/kg each) for certain product mixes. Our results show that a system with CCUS with 26% penalty (consuming 400 kWh/TON<sub>CO2</sub>) can outperform the benchmarks if used to generate electricity at least 50% of the time. This work demonstrates the benefits of flexible LCA approaches in multi-sectoral problems.

## 1. Introduction

There is a widely recognized need to reduce the use of materials to produce the goods society relies upon to meet economic and sustainability goals [1]. Efficiency-focused strategies, like *polygeneration*, can contribute to these goals by optimizing resource and flow allocations within product systems. Polygeneration converts single-product systems into *multifunctional systems* with more than one useful output - often increasing efficiency and reducing waste (e.g., using bagasse in a sugar mill to economically produce bioethanol) and its growing interest is creating new literature and practical applications [2].

One promising technology to contribute to the goal of more sustainably meeting society's energy needs is the use of hydrogen (H<sub>2</sub>) as a decarbonized energy carrier [3]. Hydrogen fuel offers high energy density on a mass basis and emits zero carbon when combusted [4]. However, despite these advantages, the current demand for H<sub>2</sub> fuel remains negligible (at less than half of a percent of global use in 2021). Challenges across the entire supply chain including low round-trip system efficiency [5], low energy per unit of volume [6], high variability in local conditions for storage [7], higher leakages [8], and higher perceived risks compared to other thermal fuels [9] are disadvantages in

producing H<sub>2</sub> as a fuel commodity for use off the production site [10]. Consequently, most H<sub>2</sub> is used for on-site produced feedstocks in the chemical industry or oil refineries, comprising 52% and 43% of the demand, respectively [11]. While industrial use of H<sub>2</sub> fuel is expected to increase by 20% by 2030 [11], achieving this will likely require government interventions to promote deployment [11–13].

Currently cost-competitive methods for H<sub>2</sub> production have substantial climate impacts. In 2021, H<sub>2</sub> production by steam methane reformation (SMR), coal gasification, and refinery by-production accounted for almost all H<sub>2</sub> worldwide (62%, 19%, and 18%, respectively) with minimal production using renewable electricity or carbon capture, utilization, and storage (CCUS) [11]. Although there is a large difference between the median climate impact values reported in the literature for uncontrolled SMR and gasification (13 and 22 kg<sub>CO2e</sub>/kg<sub>H2</sub>, respectively) compared with production via wind- and solar-driven electrolysis (3 and 2 kg<sub>CO2e</sub>/kg<sub>H2</sub>, respectively) [14], cost differences continue to explain the choice in production methods. The levelized cost of H<sub>2</sub> from SMR is less than half that of gasification (1.1 vs 2.6 \$/kg<sub>H2</sub>) [15] and about one-fifth that of renewable-powered electrolysis (~5 \$/kg<sub>H2</sub>) [16] - which is also highly sensitive to electricity prices [6,17–20]. As for lower-carbon fossil options, CCUS-controlled

\* Corresponding author. 470 Hitchcock Hall, 2070 Neil Ave, 43210, Columbus, OH, USA.

E-mail address: [gingerich.62@osu.edu](mailto:gingerich.62@osu.edu) (D.B. Gingerich).

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SMR ( $4.6\text{kgCO}_2\text{e/kgH}_2$ ) and gasification ( $4.1\text{kgCO}_2\text{e/kgH}_2$ ) are reported to be currently less costly ( $\sim 1.6$  and  $\sim 3.1$   $\$/\text{kgH}_2$ , respectively) [15, 21–23] and also can produce larger amounts of  $\text{H}_2$  than electrolysis with smaller land footprints as well [24].

Producing cost-competitive low-carbon  $\text{H}_2$  at scale currently involves high technoeconomic uncertainty and notable environmental trade-offs. Although there is a mature body of literature on the topic, cost and performance assessments of SMR and gasification with CCUS remain highly sensitive to uncertain technoeconomic parameters [25–27]. Moreover, some studies warn about assessments using overly optimistic assumptions [28,29]. To date CCUS outside of the oil and gas sector has not been deployed at scale and still requires additional demonstration to reduce uncertainty. In addition to economic factors, studies have found environmental trade-offs from  $\text{H}_2$  production pathways with low global warming potential (GWP). Gasification and SMR with CCUS have notably high impacts in acidification potential [30–32] while solar-driven electrolysis produces significantly larger terrestrial ecotoxicity potential and both wind- and solar-driven electrolysis have substantially greater freshwater and marine ecotoxicity, even compared to uncontrolled fossil- $\text{H}_2$  [33].

Developing a strategy to produce lower-carbon coal-based  $\text{H}_2$  and integrating it with agro-chemical activities can advance sustainability in different ways. Fundamentally, displacing conventional fossil fuels with low-carbon  $\text{H}_2$  will reduce greenhouse gas emissions (GHG). Furthermore, leveraging conventional energy activities to promote emerging low-carbon technologies increases acceptability [34] and could temporarily alleviate economic burdens in impacted communities during low-carbon transitions. For instance, as communities that rely on coal-sector employment tend to be more socioeconomically vulnerable [35], a coal-based  $\text{H}_2$  decarbonization pathway can help increase adaptive capacity if the financial benefits are invested in creating human or technological capital for a non-fossil based economy [36]. Additionally, integrating ammonia ( $\text{NH}_3$ ) in these polygeneration systems extends those potential social benefits to the agricultural sector [37]. Finally, this integration also improves the economic viability of  $\text{H}_2$  as there is an existing demand and infrastructure for  $\text{NH}_3$  and electricity that can be used to create opportunities for learning to address barriers for  $\text{H}_2$  fuel deployment.

Despite the potential for multifunctional coal-based  $\text{H}_2$  systems and numerous single-product technical analyses, there is a lack of life-cycle environmental assessment literature on polygeneration systems. Of possible coal-based  $\text{H}_2$  production technologies, Integrated Gasification Combined Cycles (IGCC) systems are the best characterized. The IGCC process transforms solid carbon-based fuels into synthetic fuel gases (i. e., gasification) [38] to produce electricity as final output with high energy efficiencies (e.g., dry-feed entrained gasifiers achieve 50.9% on LHV basis) [39] and relatively lower plant emissions compared to solid fuel combustion ( $720$  vs  $777\text{g/kWh}_{\text{Net}}$ ) [40]. Additionally, this process operates at conditions favorable for efficient CCUS implementation [41] with emissions as low as  $\sim 100\text{g/kWh}_{\text{Net}}$  [40]. Numerous technical [42–46] and financial [40,47–57] studies have reported costs and stack  $\text{CO}_2$  emissions of IGCC systems using the common dry-feed entrained *Shell gasifier*, while only a handful have studied the life-cycle environmental impacts in single-output electricity production [58,59].

In contrast to life-cycle  $\text{H}_2$ -based electricity production analysis, there are numerous published life-cycle assessments (LCA) studying climate impacts on single-output, coal-derived  $\text{H}_2$  as a thermal fuel [15, 20,20,21,24,33,58,60–68], but few have analyzed gasification in multifunctional systems [69]. This is representative of multifunctional energy systems in the technology assessment literature broadly. In their 2017 review of the polygeneration literature, Jana and De [2] note that technical and economic assessments substantially outnumber environmental ones. Critically, from the environmental studies they report, before the Jana and De review, only one was an LCA addressing polygeneration [70], a disparity given the abundant literature on multi-sectoral integration of energy systems around  $\text{H}_2$  products [71–73].

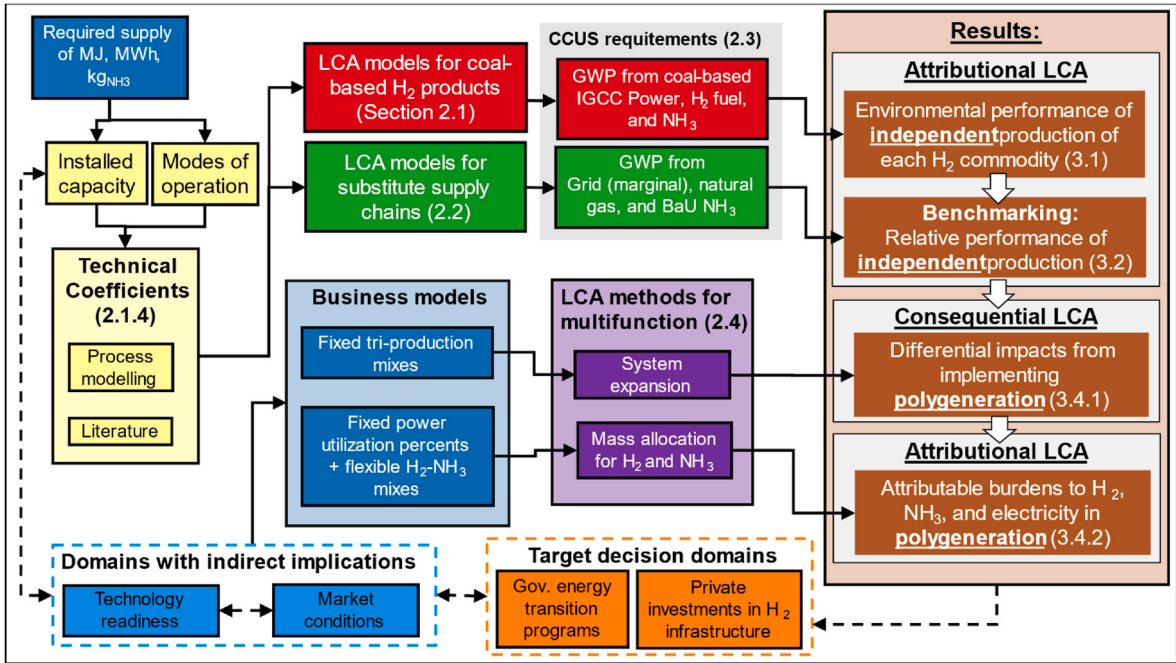
The need for improved methods for assessing multifunctional systems can partly explain this gap. Life-cycle assessments are a well-established environmental characterization method, with standardized approaches established in ISO 14040 and 14044 [74,75], that enable analysis of polygeneration systems. However, despite its maturity, LCA practitioners have criticized the standards for several reasons, including their guidelines for multifunctional systems. The standards set a hierarchy to address multifunctional systems by avoiding *allocation* “wherever possible” and prioritizing techniques like *system expansion* [74]. This hierarchy has drawn criticism [76,77] as some practitioners stress that it is excessively prescriptive [78], excludes valuable existing techniques [79], and contradicts the principle that study design should follow LCA goal and scope [80]. Others highlight the failure of the standards to adequately distinguish between *attributorial* (to determine the burdens that are associated to a product and its lifecycle) and *consequential* (to determine the consequences of a decision) [81–84] studies – an often critical distinction in method choice. Since the publication of the Jana et al. review, the number of  $\text{H}_2$  polygeneration LCAs has increased – many of which do not strictly adhere to standards [85–89]. These studies either used more than one method to address multifunctionality [85,86] or elected to implement non-preferred allocation methods like economic [87], input-based [88], and time [89].

In recent  $\text{H}_2$  LCA studies, scholars and practitioners have opted for more flexible goal-and-scope-based studies with focus on methodological questions, however multifunctionality and uncertainty analysis are dominated by particular approaches. In a systematic review of  $\text{H}_2$  LCAs, Puig-Samper et al. [14] found that attributorial approaches significantly outnumber LCAs explicitly adopting consequential approaches [14] with a growing number of works that compare allocation methods [90, 91] and techniques [92]. Substitution techniques dominate multifunctionality studies in production stages [93–95] with only one study employing system expansion [90]. Meanwhile, more than half of  $\text{H}_2$  LCAs that address uncertainty use scenario-based approaches with only one study using Monte Carlo simulations [96]. Our work contributes to diversifying the growing  $\text{H}_2$  LCA literature with a multifunctional approach that includes a consequential LCA, an attributorial study that combines allocation and system expansion, and uncertainty analysis using Monte Carlo simulations.

In this paper, we demonstrate the potential advantages of a flexible use of LCA techniques for multifunctional systems over strict adherence to standards by exploring the relative climate benefits of *gasification-based tri-production*. We do so by tailoring an LCA framework through a series of attributorial and consequential approaches to address a specific question about  $\text{H}_2$  impacts as part of a polygeneration strategy: “can polygeneration contribute to the reduction of the climate impacts of coal-based  $\text{H}_2$  fuel if a combined production process with electricity and ammonia is implemented?” Using this framework, we determine the CCUS requirements for three products ( $\text{H}_2$  produced and used as fuel, an input for  $\text{NH}_3$ , and electric power) to match the life-cycle climate impact of existing, independently-produced benchmark processes and then investigate the extent that a combined production of a basket of these three products contributes to lowering required CCUS standards compared to making these products independently. Finally, we analyze what proportions of impacts are attributable to each product to contextualize the burdens of the  $\text{H}_2$  fuel product and find competitive environmental advantages in polygeneration relative to single-product systems. Our flexible LCA approach provides valuable insights on the benefits of polygeneration that overly prescriptive adherence to LCA standards precludes.

## 2. Methods

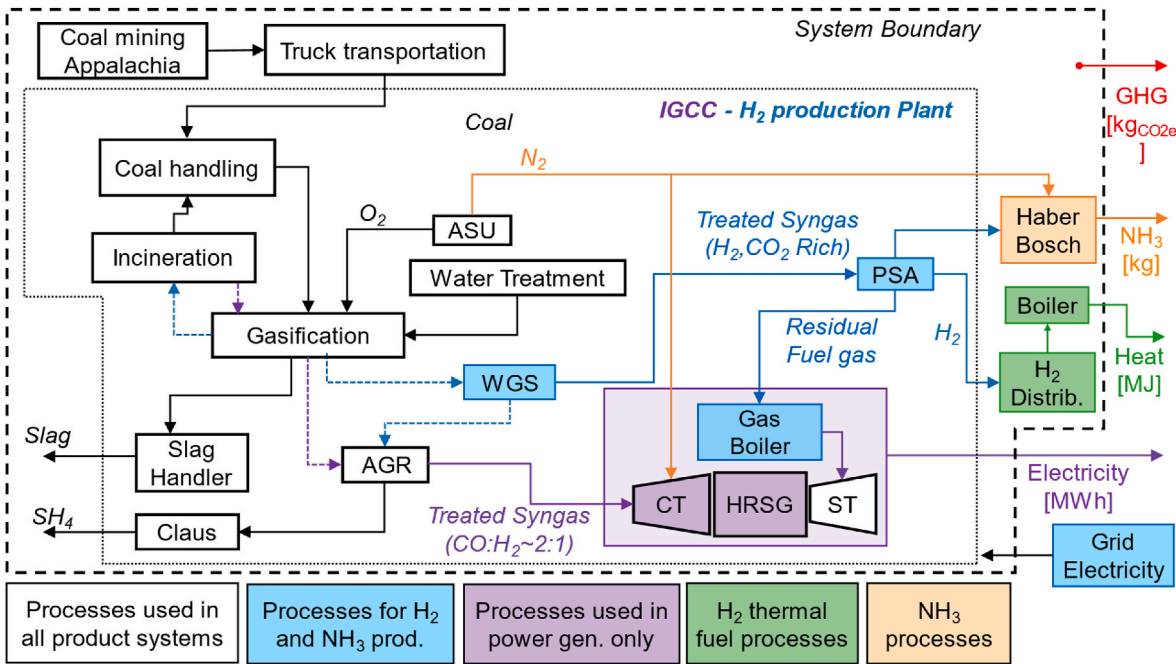
We designed a three-fold LCA-based framework to assess the climate impacts of a coal-based  $\text{H}_2$  polygeneration schema (Fig. 1). First, using an attributorial approach, we determine a life-cycle climate impact (measured using GWP) baseline for the impacts that the coal-based  $\text{H}_2$



**Fig. 1.** LCA framework to analyze climate impacts from coal-based H<sub>2</sub> products and differential impacts of implementing polygeneration. We calculate GWP impacts from the H<sub>2</sub>-based supply chain, compare those impacts with existing products (substitute supply chains) and analyze the performance of combined production of power, fuel, and ammonia using multifunctional LCA techniques. The expected domains of influence are represented with dotted lines. Numbers in parenthesis denote the section of the main manuscript.

products generate (electricity, H<sub>2</sub> as fuel, and NH<sub>3</sub>) and compare these impacts to business-as-usual (BaU) benchmarks of substitute commodities (i.e., the regional grid, combusted natural gas, and NH<sub>3</sub> supplied in Ohio) allowing us to calculate the CCUS efficiency requirements for coal-based production to reduce climate impacts. Second, using a consequential LCA approach, we evaluate strategic (macro) decisions of implementing polygeneration systems with capacity to deliver different

baskets of products. Third, we again use an attributional LCA approach to evaluate the burdens of each product in the polygeneration system. Our third analysis also allows us to assess operational (micro) decisions about varying the proportion in the basket of products.



**Fig. 2.** Coal-based H<sub>2</sub> supply chain with three product systems and system boundary. The system includes the upstream processes and the required services needed to supply coal as feedstock, the IGCC and H<sub>2</sub> production plant, NH<sub>3</sub> synthesis (via the Haber-Bosch process), H<sub>2</sub> distribution and boiler. White boxes represent process units that are used in all the three different product systems and colored boxes serve one or two product systems.

## 2.1. LCA of coal-based $H_2$ (gasification-based) supply chains

### 2.1.1. Process description

Our model quantifies climate impacts occurring from the material extraction phase to the transformation steps of gasification, power generation, and  $NH_3$  synthesis. We modeled three coal-based product systems using a cradle-to-gate approach (upstream processes and the transformation in the gasification, power, and ammonia plants). The upstream processes include the extractive activities (e.g., fuels and feedstock production) to run the transformation processes and we set the downstream boundary on the product once it is made available for utilization. The three products from this system include: (1)  $H_2$  fuel at 99.9% purity at 6.5 MPa (commercial conditions for pipeline transportation) [15], (2) alternating current electricity at the power plant, and (3)  $NH_3$  for fertilizer production. The core of the three product systems is the *dry entrained bed gasification (Shell gasifier)* based on the technical assumptions presented in the Supporting Information (SI) Section 1 (design assumptions and unit processes description), with the core assumptions based on the NETL baselines for fossil- $H_2$  electricity [40] and  $H_2$  production [15]. In Fig. 2, we represent the three product-systems including the main unit processes and the system boundaries.

### 2.1.2. Goal and scope

Our LCA aims to determine if and how producing three  $H_2$  commodities via a polygeneration process can improve environmental performance relative to a dedicated, sole-product processes. The objectives of our study are twofold: 1) to assess the consequences of polygeneration implementation as an alternative to develop the production of three coal-based  $H_2$  commodities and 2) to determine what proportion of the burdens from that polygeneration system are attributable to  $H_2$  fuel. We intend for these results to help policymakers in analyzing alternatives involving coal-based  $H_2$  production and understanding the potential of efficiency-based approaches like polygeneration in scaling new technologies, such as low-carbon  $H_2$  production infrastructure.

### 2.1.3. Defining a baseline: system boundaries and key assumptions for an attributional LCA

To address the LCA objectives, we establish a baseline of impacts from the sole production of the three coal-based  $H_2$  commodities. We modeled industrial-size facilities using commercial technologies (i.e., power generation in an IGCC plant with capacity  $640MW_{Net}$  [40],  $H_2$  production at 650 T/day [15], and  $NH_3$  production 2000 T/day [97]) under favorable operational conditions in Ohio – a location where annual hydrogen demand for industry and refineries is projected to grow to more than 600,000 tonnes by 2050 [98]. This assumes our product systems operate at a maximum production rate, so blends correspond to production over periods of time (e.g., annual or seasonal) which allow us to use average production levels without further partial operation assumptions (e.g., we do not consider efficiency losses due to simultaneous production). We first built the LCA models in OpenLCA [99] to quantify life-cycle 100-year-horizon climate impacts from sole sourcing each commodity. Our functional units (FUs) for the sole-production processes are  $1MJ_{Th}$  of fuels,  $1kWh_{net}$  of electricity, and  $1kg_{NH_3}$  of fertilizer. The scope of our work is a cradle-to-gate analysis, including upstream processes, but not impacts associated with distribution and use phases (i.e., heat or  $NH_3$  distribution to final customer or electricity delivery to the grid). We assumed operation with bituminous coal sourced from Appalachia and delivery of finished products within Ohio. Additional details on the unit processes (and data sources) serving each product system can be found in Table S1 in SI Section 2.

### 2.1.4. Inventory

We used literature data to characterize the systems in our process-based LCA. Much of our model leverages data from the NETL fossil energy plant baselines [40] and comparison of state-of-the art fossil-based

$H_2$  production [15] reports. In addition, we used other peer-reviewed and grey literature detailing the dry-fed, entrained-bed Shell Gasifier to inform the model. Since the core of our analysis and the bulk of the emissions are caused in the gasification plant stage, we built our inventory for foreground processes based on flow sheets of unit processes reported in the literature as we did not find in LCA databases available data for specific product units in the gasification production system at the required conditions. Our data on background processes comes from a combination of literature values and dedicated LCA databases. We prioritized data from Ohio or the United States Midwest (for geographic representativeness) or from the past 15 years (to reflect the maturity of the Shell Gasifier system). Where high-quality or sufficient literature data was unavailable, we supplemented it using energy/mass balances and reaction kinetics, (e.g., we used stoichiometric  $NH_3$ - $H_2$  ratio and overall process efficiencies in the literature to characterize the Haber-Bosch process). Finally, we iteratively modeled and selected the processes and parameters, excluding flows and processes that accounted for less than 2% of impact (e.g.,  $N_2O$  contributions as GHG and  $H_2$  transportation from the gasification facility to the Haber-Bosch plant for  $NH_3$  production).

### 2.1.5. Modeling uncertainty

We incorporated uncertainty into our LCA model via Monte Carlo simulations (1000 runs per model) with triangular distributions using the median literature value as the mode and the maximum and minimum values as the extreme parameters. To build the distributions, we prioritized collecting data for the same processes and conditions. When data from identical processes were not available, we harmonized sources to use the same assumptions - guaranteeing consistency. For example, we accepted references for different high-rank bituminous coals as the literature reports less than 5% efficiency variation of the Shell gasification process using those types of coals with and without CCUS [46, 53].

## 2.2. Benchmark process impacts

The comparison benchmarks for coal-based  $H_2$  are the GWPs of the BaU substitute commodities in Ohio. The power generation benchmark corresponds to the on-peak marginal unit of the PJM system (i.e., median value of  $689g_{CO_2e}/kWh_{net}$ ) [100]. The uncontrolled natural gas supply chain includes emissions, losses, and energetic inputs for extraction in the Appalachian and Gulf of Mexico regions and combustion in industrial boilers (i.e.,  $108g_{CO_2e}/MJ$ , median) [8,29,101]. Finally, we modeled the effects from SMR-produced  $NH_3$  using data for a mix of state-, national- and international-supply chains that likely serve the United States market (i.e.,  $2.6kg_{CO_2e}/kg_{NH_3}$ ) [102].

## 2.3. Calculating carbon capture requirements

Carbon capture systems require a significant amount of energy to operate, creating a trade-off between capturing  $CO_2$  and second-order emissions due to energy consumption [103]. We analyze this trade-off using switchover analysis to identify the required CCUS energy efficiency for the production of the coal-based  $H_2$  basket to have the same climate impact as their respective BaU benchmark. The relationship in Eq. (1) shows the conditions where the coal-based  $H_2$  basket has equal or better environmental performance compared to the benchmark, where  $E_{BAU,i}$  represents the GWP intensity for a FU of product  $i$  (in  $[kg_{CO_2e}]$ ),  $E_{PG,i}$  is the GWP produced by the coal-based  $H_2$  system for product  $i$  (in  $[kg_{CO_2e}]$ ). The gross carbon captured is the product of the capture effectiveness,  $\eta_{CCUS}$  (in percent), that represents the ratio of  $CO_2$  captured, and the stack emissions,  $SP_{G,i}$  (in  $[kg_{CO_2}]$ ), for each product  $i$ . The second-order emissions are associated with energy used in the production process and is the product of the gross carbon captured, the emissions intensity of the power used for the process,  $G_{kWh}$  (in  $[kg_{CO_2,emitted}/kWh]$ ), and the energy efficiency of the capture system,  $f_{CCUS}$ ,



measured in  $[kWh/Ton_{CO_2,captured}]$ .

$$E_{BAU,i} \geq E_{PG,i} - \eta_{CCUS} S_{PG,i} + (\eta_{CCUS} S_{PG,i}) (G_{kWh} f_{CCUS}) \quad (\text{Eq. 1})$$

We vary  $f_{CCUS}$  to find the switchover point (i.e., where the two sides of Eq. (1) are equal). In our main study cases, energy efficiency,  $f_{CCUS}$ , is either 350 and 400 kWh/TON<sub>CO<sub>2</sub></sub>, corresponding to energy penalties of 23% and 26%, medium and high energy consumption values reported in the CCUS literature [29,104,105]. Although CCUS systems typically require heat and electrical energy inputs, we represent energy consumption as one value (measured in  $[kWh]$ ) that comprises both energy requirements. This reduces model complexity and to present results as percent of energy penalty relative to plant generation capacity. We assumed a capture effectiveness,  $\eta_{CCUS}$ , of 85%, a conservative value reported in the literature for CCUS in IGCC systems [26,27,40,106].

## 2.4. Assessing impacts in multifunctional H<sub>2</sub> systems

While the ISO LCA standards specify a preference between techniques to address multifunctionality, our work includes both system expansion and physical allocation, to address different questions about implementation alternatives of polygeneration. We use system expansion (Case I) to analyze consequences from structural decisions about production capacity or contractual set-ups of polygeneration at given fixed production schedules (e.g., plants with power purchase agreements with fixed energy, H<sub>2</sub>, and NH<sub>3</sub> commitments). We use a physical allocation strategy (Case II) to determine attributions to each product. Running Case II for different product mixes also let us analyze impact changes associated with variation in operation, assuming flexible operation is possible (e.g., a plant without long-term production contracts for the three products). Based on the nature of the products and to address the attributional question, we designed a novel allocation technique that we call “*adapted mass allocation*” that combines physical allocation with *system expansion*.

### 2.4.1. Case I: using system expansion to analyze consequences from structural decisions

With system expansion, we analyze the impacts from designing and implementing polygeneration systems with defined product bundles. Our approach explores not only the case in which the plant only generates power as we studied six different IGCC use levels. This product bundle includes 91,800 tonnes<sub>NH<sub>3</sub></sub>/year of NH<sub>3</sub> (given the estimated growth of 15% of the NH<sub>3</sub> production in Ohio by 2050) [98,107], while utilizing the IGCC capacity at 0% (i.e., no electricity generation), 15%, 33%, 50%, 67%, and 85% of the time with the balance of the H<sub>2</sub> feedstock sold as fuel.

Each of these operation regimes represents a different product bundle. We model the amount of electricity production,  $P_{Elec,net}$  (in [MWh]), as a function of the system’s capacity factor,  $C_f$ , the capacity of the IGCC plant,  $I_{IGCC}$  (in [MW]), the capacity for H<sub>2</sub> production,  $I_{H_2}$  (in [T/h]), and the percent utilization for power generation,  $U_{IGCC}$  (Eq. (2)). Complementing this, Eq. (3) represents the relationship of the H<sub>2</sub> produced and sold as thermal fuel,  $P_{H_2,Th}$  ([kg]), with the amount used to produce ammonia,  $P_{H_2,NH_3}$  ([kg]). With those equations and other production efficiency variables (i.e., the stoichiometric H<sub>2</sub>–NH<sub>3</sub> relationship and H<sub>2</sub> lower heating value), we derive impacts per functional unit for each production mix under the NH<sub>3</sub> production level. For a detailed example of this calculation, see SI Section 3.

$$P_{Elec,net} = C_f I_{IGCC} U_{IGCC} \quad (\text{Eq. 2})$$

$$P_{H_2,Th} = C_f I_{H_2} (1 - U_{IGCC}) - P_{H_2,NH_3} \quad (\text{Eq. 3})$$

### 2.4.2. Case II: allocating impacts in a basket of material and non-material products

We use allocation to determine to what extent impacts are attributable to H<sub>2</sub>-fuel within the basket of products. Since the ISO LCA

standards suggest that preferred allocation methods should reflect “underlying physical relationships” [75], we implement mass-based allocation, as mass is a property of a substance independent of use, making it a meaningful for both fuels and chemicals. Other common allocation methods, like energy or exergy allocation, require additional assumptions (e.g. that NH<sub>3</sub> thermal energy content appropriately represents its use as fertilizer). However, under conventional mass allocation schemes, electricity cannot have allocated impacts as it does not have mass.

To address this challenge, we designed and tested three alternative partition methods and selected the most suitable one to model our main case. These three methods include: i) allocation based on the mass of the H<sub>2</sub> used to produce each commodity, ii) allocation based on the mass of the material products (H<sub>2</sub> and NH<sub>3</sub>) with impacts allocated to electricity based on the mass of the used H<sub>2</sub>, and iii) an adapted mass allocation strategy in which non-material product (electricity) are subtracted from the total impact with remaining impacts allocated based on product mass, then we separately compare IGCC electricity production with the BaU benchmark (the electricity grid).

We present *iii* in the main manuscript as it is uniquely suited for our application. It is the only method that reflects a true proportional relationship between the allocation factor and the quantity of all the products. This method also fits the plausible scenario in which the system produces a fixed amount of electricity as a determinant product (e.g., a plant under a power purchase agreement) and the rest of the schedule is flexible to produce varying proportions of H<sub>2</sub> and NH<sub>3</sub>. Moreover, with this method, we only assign impacts based on mass to the material products and avoid attributing GHG to electricity based on mass which would be an unnatural choice given it is a non-material product. This leverages system-expansion logic, similar to the allocation at the point of substitution (APOS) technique [108] that also expands the analysis beyond the separation point. However, APOS is not suited to our application as it is designed for analyzing the conversion of a waste by-product to a saleable product whose impacts should be partially allocated to the main product. In contrast, each product in our basket has a standalone utility and could be a determining product if the application requires it.

Furthermore, despite their practical advantages, there are conceptual inaccuracies in the first two partition methods that might distort allocation. Those methods maintain a complete partition logic and are, therefore, more explainable: we assign shared impacts based on some quantification of mass of delivered product or used in the process. Method *i* has the additional advantage that it corresponds to a simpler subdivision, a preferred technique per the ISO standards [74,75] while methods *ii* and *iii* are more complex. Although the three methods can be meaningful, methods *i* and *ii* are inaccurate as they assign emissions to electricity using the H<sub>2</sub> input mass flow as a proxy, disregarding the thermal energy of the CO co-combust in the IGCC (as the syngas fuel is not pure H<sub>2</sub>), distorting mass flows. Despite this, we present results for methods *i* and *ii* as a sensitivity test in SI Section 4 and Fig. S3 in which we obtained higher impacts allocated to H<sub>2</sub> fuel. Finally, our allocation analysis also allows us to study impact variations of operational (micro) changes to the basket of products. In Case II, we model fixed utilization factors for power generation as defined in Case I, while varying the H<sub>2</sub> - NH<sub>3</sub> mix. For a given power utilization factor  $U_{IGCC}$ , we generated emission curves for the entire range of H<sub>2</sub>–NH<sub>3</sub> co-production balances during the remaining operation time. To properly assign impacts, we identified the emissions from processes exclusively used to produce NH<sub>3</sub> (i.e., processes related to the Haber-Bosch) and allocated them solely to that product which further makes our technique conceptually different from APOS.

### 3. Results

#### 3.1. Lifecycle climate impacts from coal-based $H_2$ products

Production emissions are the main source of climate impacts for each coal-based  $H_2$  product system (right bar in Fig. 3). For uncontrolled IGCC, Fig. 3A shows that the direct  $CO_2$  stack emissions account for 81% of the  $1068g_{CO_2e}/kWh_{net}$  of the expected impacts for power production, followed by methane from coal mining (12%) and other  $CO_2$  sources (6%). Similarly,  $CO_2$  emissions from  $H_2$  feedstock production create the largest impacts in  $NH_3$  synthesis system, 70% of the expected  $4.6kg_{CO_2e}/kg_{NH_3}$ , while  $CO_2$  from electricity use and methane from  $H_2$  production generate additional 13% each (Fig. 3B). In the thermal fuel product system,  $CO_2$  emissions from the  $H_2$  production plant creates the largest climate impacts - causing 65% of the  $216g_{CO_2e}/MJ$  (Fig. 3C). This is followed by emissions from coal extraction (14%) and emissions from electricity production (12%). Notably,  $CO_2$  emissions contribute about 85% of the GWP in all three product systems. Furthermore, the expected values from our analysis are close to literature reported values (SI Section 5, Fig. S2 and Table S2).

Our results have substantial uncertainty due to the underlying wide ranges of parameters taken from the literature. The uncertainty shown by the error bars in Fig. 3 is driven by a few parameters (e.g., electricity consumption in gasification auxiliaries like Air Separation and Acid Gas Removal) have distributions that vary by more than one order of magnitude. Uncertainty in these parameters in one-at-a-time analyses produce negligible ( $<2.7\%$ ) variations, indicating that the combination of extreme cases leads to outliers.

#### 3.2. Relative performance of the independent production vs the BaU benchmarks

All the  $H_2$ -based commodities in our analysis have significantly larger impacts than their BaU benchmarks in sole-production without CCUS. Fig. 3A and C shows that IGCC power and  $H_2$  fuel production are deterministically dominated by their BaU comparisons. The expected climate impacts from the  $H_2$  fuel production case are twice that of the natural gas supply chain ( $216$  vs  $110g_{CO_2}/MJ$ ), while uncontrolled IGCC power generation causes 55% higher GWP than the marginal generator ( $1068$  vs  $689g_{CO_2}/kWh_{net}$ ). The  $NH_3$  case also shows a significant difference (Fig. 3B), with the coal-based  $NH_3$  system causing 88% more GHG emissions than the BaU option ( $4.6$  vs  $2.6kg_{CO_2}/kg_{NH_3}$ ). For all three products, most emissions are smokestack emissions, so CCUS could address the largest source of GWP. Distributional statistics from the Monte Carlo simulations are tabulated in SI Section 6, Table S3.

Without CCUS, power generation has the smallest difference from its BaU benchmark, the regional grid (Fig. 3A). The largest difference between these two products occurs during production:  $CO_2$  emissions from the IGCC stack are 47% higher than the grid on-peak marginal generator ( $868$  vs  $460g_{CO_2e}/kWh_{net}$ ). However, grid electricity has 14% more upstream impacts than IGCC ( $229$  vs  $201g_{CO_2e}/kWh_{net}$ ).

The  $H_2$  fuel production and  $NH_3$  synthesis processes perform significantly worse relative to their benchmarks, natural gas and BaU  $NH_3$  supply. In the  $NH_3$  case, the gasification process causes 50% more GHG than  $NH_3$  supply based on SMR ( $4.6$  vs  $2.6kg_{CO_2e}/kg_{NH_3}$ ) (Fig. 3B). For the  $H_{2,th}$  case (Fig. 3C), the  $CO_2$ -free combustion of  $H_2$  does not compensate for production phase differences as gasification generates more than four times the GWP attributable to natural gas supply processes ( $144$  vs  $35g_{CO_2e}/MJ$ ) and upstream processes add to this difference ( $75$  vs  $9g_{CO_2e}/MJ$ ). The emissions from the natural gas combustion stage are substantially smaller than the accumulated difference for production and distribution with the  $H_{2,th}$  fuel product ( $64$  vs  $175g_{CO_2e}/MJ$ ).

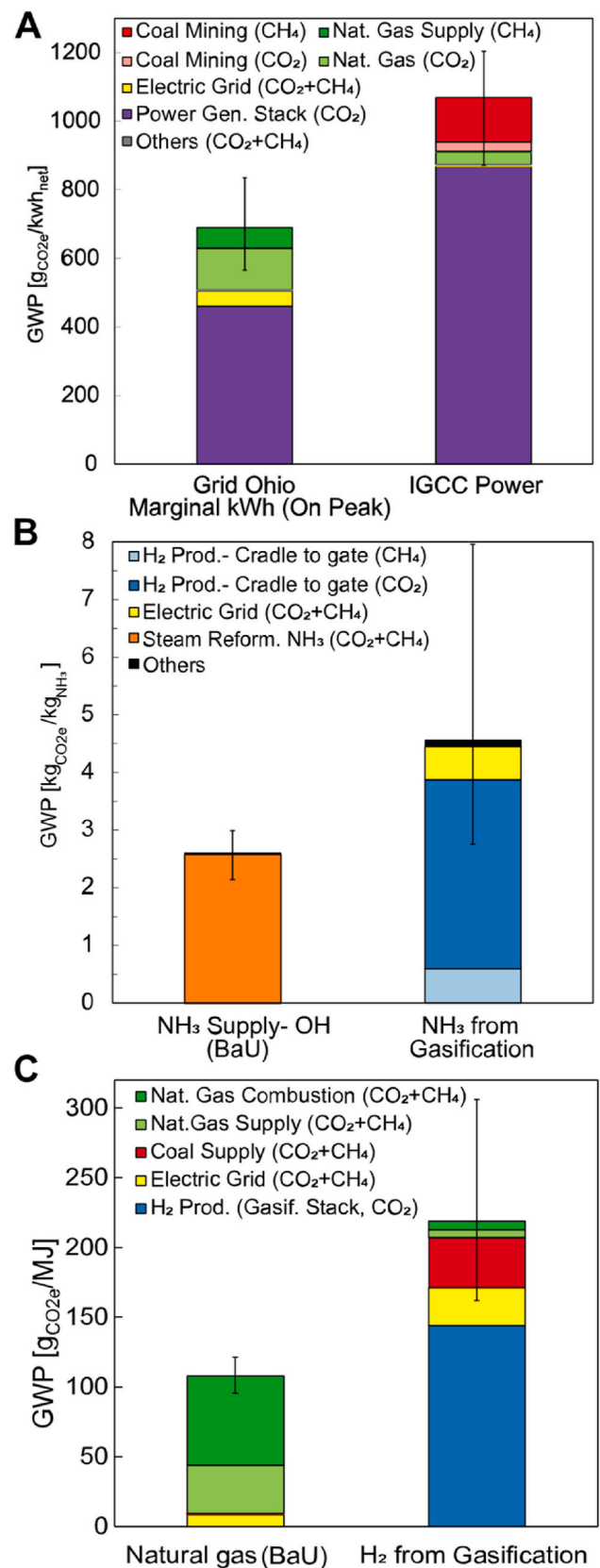


Fig. 3. GWP comparison with BaU benchmarks of coal-based  $H_2$  A) Power Generation, B)  $H_2$  fuel, and C)  $NH_3$  production. The BaU supply outperformed all the gasification-based products without CCUS deployment. N.B. the units for panel (B) are in kilograms, rather than grams.

### 3.3. Calculating carbon capture requirements

Gasification-based products with larger differences relative to their benchmarks require more efficient CCUS systems. In Fig. 4, the IGCC process line plot intersects at a higher CCUS power consumption level, which indicates that this process needs a less-efficient CCUS (one that consumes a larger amount of energy per tonne captured) to become GHG-competitive than other processes. This result aligns with the differences between products that are shown in Fig. 3 when we see that IGCC required a reduction in its carbon intensity by 35.5% to be GHG-competitive with its benchmark (vs a 43.5% reduction required by  $\text{NH}_3$  and 50.5% by  $\text{H}_2$  to match the impacts of their respective benchmarks).

While producing  $\text{H}_2$  fuel or  $\text{NH}_3$  within a sole-function system requires substantial reductions in CCUS energy use from existing technologies (to 94 and 189  $\text{kWh}/\text{TON}_{\text{CO}_2}$ , respectively), the same is not necessarily true for IGCC systems. In simulations incorporating a CCUS system, we determine that a CCUS process with a power consumption as high as 566  $\text{kWh}/\text{TON}_{\text{CO}_2}$  allows power generation via an IGCC system to have less life-cycle climate impact than the marginal kWh of the grid (intersection between the purple line and the dotted line in Fig. 4). That allowable energy penalty is 37% of the IGCC net generation for break-even climate impacts which is a significantly higher specific consumption than literature values for IGCC plants of 19% [40], 21.3% [105], 14–25% [104]. Counterintuitively, this result shows that a less efficient CCUS, even one that consumes more energy than current technology, will still lead the IGCC to have less climate impact than the selected benchmark (the electricity grid). In addition to needing to capture less  $\text{CO}_2$  than other products, the reason for this reduced efficiency requirement is because the CCUS is powered by the lower-carbon IGCC system (with a lower  $G_{\text{kWh}}$ ) itself and not by the more carbon-intense electricity on the grid.

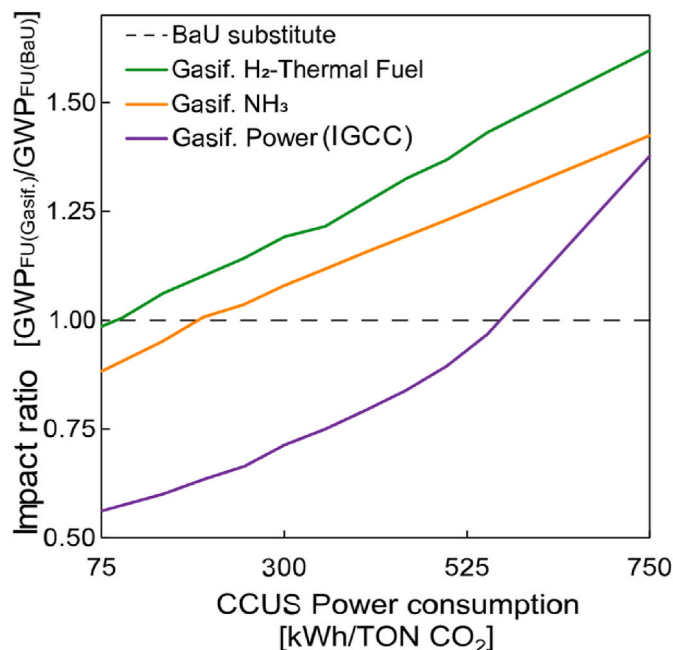


Fig. 4. Climate impacts of gasification-based sole-production of three products vs BaU benchmarks as function of CCUS energy requirements. The horizontal axis represents the CCUS energy efficiency (where less efficient systems have higher power consumption to capture one tonne of  $\text{CO}_2$ ) and the vertical axis is the ratio of impacts for the gasification-based product with their benchmark (a ratio higher than 1 corresponds to having higher impacts than the benchmark).

### 3.4. Combined tri-production effect on CCUS requirements

#### 3.4.1. System expansion: differential impacts from implementing polygeneration of power- $\text{H}_2$ - $\text{NH}_3$

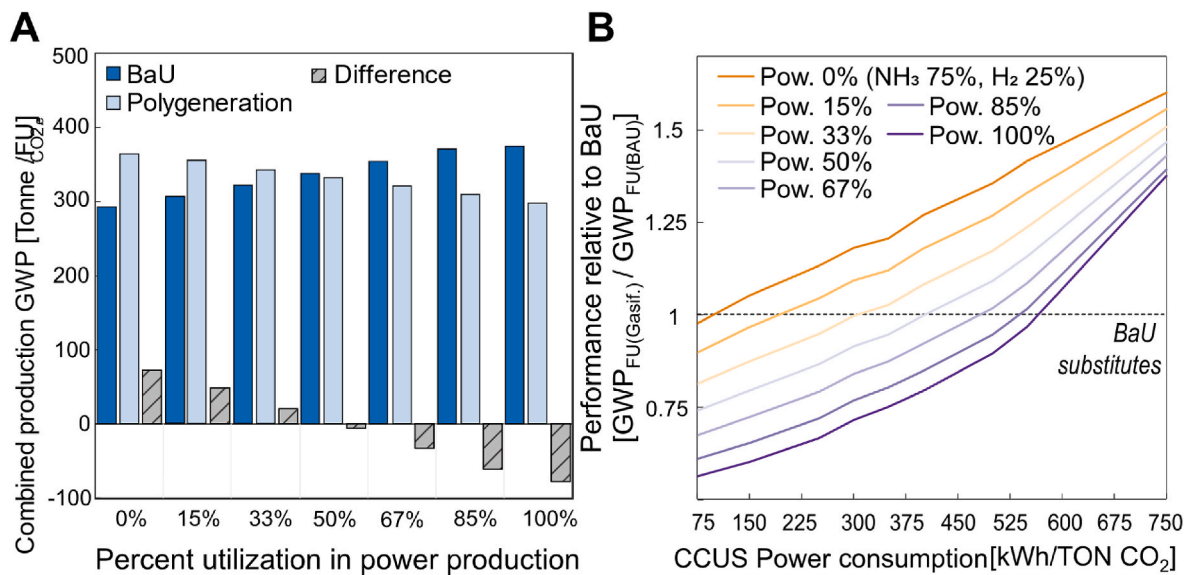
Implementing CCUS with certain product combinations leads to polygeneration that is GHG-competitive compared with BAU benchmarks (regional grid, SMR-ammonia, and natural gas). We analyzed different polygeneration product mixes with a CCUS that consumes 400  $\text{kWh}/\text{TON}_{\text{CO}_2}$ , an estimate based on current systems [40,104,105] corresponding to a 26% energy penalty ( $\text{CCUS}_{26\%}$ ), as derived in SI Section 7. At higher utilizations for power production, the emission reduction increases. The inflection point for this system is close to 50% utilization for power, as generating electricity with at least 50% of the system operation leads to a small reduction in the overall impacts compared to the basket of products from the BaU systems (indicated with negative grey hatched bars in Fig. 5A). We see that utilizing the plant 67% of the time for power generation with  $\text{CCUS}_{26\%}$  causes 13% lower climate impacts from the gasification tri-production and reductions of 20% are possible with 85% IGCC utilization.

The line plots in Fig. 5B show that the CCUS energy efficiency breakpoint for the tri-generation system to match the benchmark bundle (i.e., where the relative performance of a system is equal to 1), depending on the operation regime, is between 566 (for 100% use as IGCC represented as the darkest purple line) and 99  $\text{kWh}/\text{TON}_{\text{CO}_2}$  (for full operation to produce  $\text{H}_2$  and  $\text{NH}_3$ , or the darkest orange line). The latter means that the BaU product basket is comparable to the equivalent basket produced by the polygeneration system with CCUS consuming 99  $\text{kWh}/\text{TON}_{\text{CO}_2}$  with 0% IGCC utilization. This means that without IGCC electricity production, the CCUS process needs to be very energy efficient to make the polygeneration system GHG-competitive compared to the benchmarks. When we increase the IGCC utilization to 33% and 50%, less energy-efficient CCUS is sufficient to match the performance of the corresponding BaU baskets. In those cases, CCUS efficiencies as high as 300 and 400  $\text{kWh}/\text{TON}_{\text{CO}_2}$  levels, respectively, lead the system to outperform the benchmarks. In other words, polygeneration with higher power generation can use lower-efficiency CCUS systems yet achieve the goal of matching the benchmark emission intensities. Therefore, the higher use of IGCC shifts the curve to the right (requiring lower CCUS efficiency) as the FU of the expanded system has a greater proportion of electricity, the best performing product.

#### 3.4.2. Mass allocation: attributable climate impacts to $\text{H}_2$ fuel and net difference

We found large operation ranges where  $\text{H}_2$  outperforms natural gas, but this does not necessarily mean that the overall impacts from the polygeneration basket are lower than BaU. In the  $\text{CCUS}_{23\%}$  case,  $\text{H}_2$  fuel causes lower GHG than natural gas in wide ranges (green and blue line segments in Fig. 6A or regions where the solid green line is below the dotted green line in Fig. 6B and C). However, we found that at low power production, polygeneration-based  $\text{NH}_3$  causes significantly larger GHGs than the BaU case for almost any mix of  $\text{NH}_3$  and  $\text{H}_2$ . This tradeoff is visible in Fig. 6B where the solid orange line is above the benchmark dotted line in almost the entire range of possible mixes - especially in mixes where  $\text{H}_2$  has relatively lower impacts than natural gas. However, as in the system expansion case, we found that producing more power increases the ranges at which the three products simultaneously outperform the BaU benchmarks. These ranges correspond to the blue segments in Fig. 6A for polygeneration with  $\text{CCUS}_{23\%}$  (350  $\text{kWh}/\text{TON}_{\text{CO}_2}$ ). Critically, with 85% power utilization, all the  $\text{H}_{2,\text{th}}$ - $\text{NH}_3$  mixes (using the remaining 15% of produced  $\text{H}_2$ ) lead to a GHG-competitive basket (rightmost blue segment in ternary Fig. 6A plot and Fig. 6C). For comparison purposes, we report additional results for CCUS with 36% and 10% energy penalty in SI Sections 8 and 9 and Figs. S3 and S4.





**Fig. 5.** Performance of the combined tri-production for different business models for power generation. A. Tri-production with CCUS assuming an energy consumption of 400 kWh/TON<sub>CO<sub>2</sub></sub> (an energy penalty of 26%). B. Comparison for different power utilization factors with respective BaU benchmark. Higher power utilization requires less efficient CCUS to outperform the BaU cases (allows for higher consumption per captured tonne of CO<sub>2</sub>).

## 4. Discussion

### 4.1. Polygeneration for CCUS deployment in fossil-derived H<sub>2</sub> systems

Polygeneration aims to make systems more efficient by reducing the use of primary resources and non-shared infrastructure via integrated supply chains to produce multiple products. However, possible improvements are constrained by technological limits. In our case, a tri-production strategy reduces climate impacts, leveraging the relatively better performance of one product – power generation via IGCC (without CCUS). This demonstrates that polygeneration can ease adoption of feasible CCUS (Figs. 5 and 6, and Fig. S4).

By demonstrating the GWP benefits of combining polygeneration and CCUS, our work demonstrates a path for emerging technologies to cross the technological *valley of death*, or the phase of technology maturation in which an emerging technology moves from bench-scale (with R&D funding subsidies to support its deployment) to demonstration-scale and commercially successful products. We find technical benefits from connecting an emerging technology with an integrated, multisectoral business strategy, a critical strategy to overcome this valley [109–113]. In fact, we show that CCUS is already a technological alternative that can enable fossil-derived H<sub>2</sub> with similar level of impacts than current commodity alternatives.

Implementing current CCUS technologies can lead power generation via IGCC in sole-production modes to emit less GHGs than the current electricity grid – and may help reduce the environmental impact of electricity generation in renewable-constrained areas. However, implementing CCUS is insufficient for sole-production modes of coal-based H<sub>2</sub> and NH<sub>3</sub> to match benchmark emission levels. While some work [29] warns that the high uncertainty surrounding CCUS energy penalties, IGCC is consistently found to enable relatively efficient CO<sub>2</sub> capture given its higher stack-flow GHG concentrations [26,41,104,105]. Consistent with this, we find that even under conservative estimates of high energy penalties (i.e., CCUS<sub>36%</sub> consuming 550 kWh/TON<sub>CO<sub>2</sub></sub> removed), IGCC with CCUS leads to lower GHG emissions than the grid mix over its life-cycle (Fig. 3) while H<sub>2</sub> fuel and NH<sub>3</sub> production would require improved capture systems with lower energy penalties to outperform their benchmarks (i.e. CCUS<sub>6%</sub> consuming 94 kWh/TON<sub>CO<sub>2</sub></sub> and CCUS<sub>12.4%</sub> requiring 189 kWh/TON<sub>CO<sub>2</sub></sub>, respectively). Thus, the implementation of current CCUS technologies in polygeneration systems

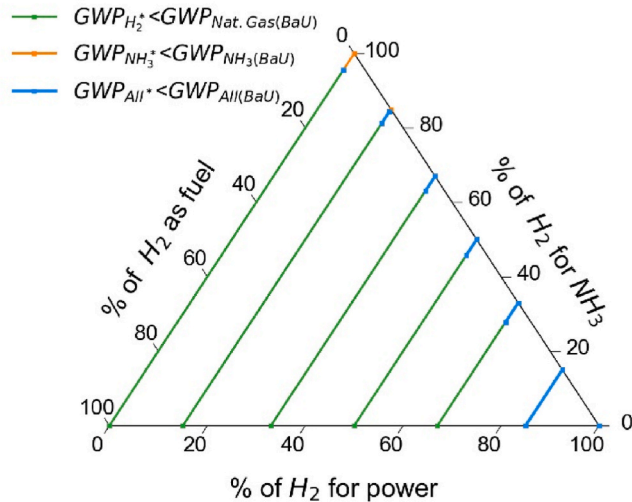
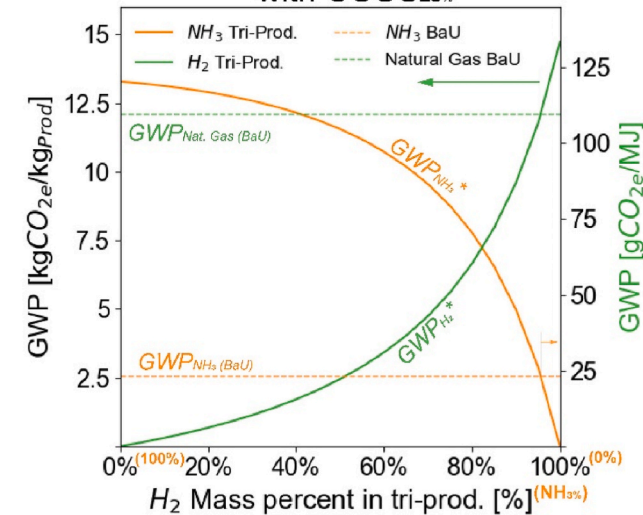
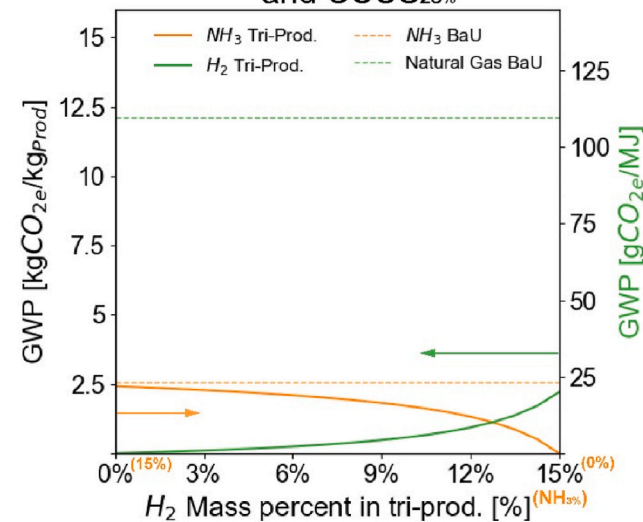
can enable H<sub>2</sub> and NH<sub>3</sub> production with similar emissions as uncontrolled natural gas and SMR-based ammonia synthesis.

Fundamentally, the relatively better performance of IGCC electricity production can enable GHG-competitive coal-based H<sub>2</sub> and NH<sub>3</sub> production without requiring significant CCUS efficiency improvements. In a polygeneration system using 15% of its capacity to produce power, gasification with CCUS<sub>23%</sub> (consuming 350 kWh/TON<sub>CO<sub>2</sub></sub>) allows for GHG-competitive tri-production for at least some H<sub>2</sub>–NH<sub>3</sub> mixes (Fig. 6A). And with 85% power utilization, any mix of H<sub>2</sub> fuel and NH<sub>3</sub> synthesis from the remaining 15% creates lower climate impacts than the BaU product basket (Fig. 6C). Moreover, under 85% utilization, the resulting H<sub>2</sub> fuel could be considered low-carbon given current international standards (3.0 kg CO<sub>2e</sub>/kgH<sub>2</sub> per EU, 2.4 per UK, and 4 per USA standards) [14,114,115]. Critically, we show that an inefficient CCUS with 36% penalty in polygeneration (SI Section 8 Fig. S3) can still enable lower climate impacts than a system with an extremely efficient CCUS without polygeneration (SI Section 9 and Fig. S4B for a CCUS with 10% energy penalty).

Despite finding that polygeneration-schemes can produce products below benchmarks, we stress that this does not imply that our system is climate (or environmentally) benign. The BaU benchmarks represent products *as produced today* and do not represent a performance target for zero-carbon futures (absent compensatory carbon-negative technology deployment). Also, our study is limited to GHG assessment and does not consider other environmental impacts that can be significant in coal-based H<sub>2</sub> production. For example, water consumption and eutrophication are reported to be significantly higher in IGCC with CCUS than in uncontrolled natural gas fueled power [40]. However, our goal was not to comprehensively assess a broad set of environmental impacts, but to develop and demonstrate the benefits from polygeneration in a particular impact. Our LCA framework could be expanded to include other impact categories. Furthermore, the economic viability of CCUS systems continues to be uncertain and depends on the specific applications [27], operation modes [116], and require further policy support to be achieved widespread [12]. Fundamentally, the main implications of our work do not lie with identifying the benefits of the specific coal-based H<sub>2</sub> tri-generation case study, but in the demonstration of the potential benefits of combining polygeneration with technological improvement.

Our approach of analyzing the benefits from combining technological improvement with efficiency-seeking strategies can be applied to



**A** Tri-Prod and CCUS<sub>23%</sub>\* Vs BaU**B** H<sub>2</sub>-NH<sub>3</sub> Co-Prod. (No power) with CCUS<sub>23%</sub>**C** Tri-Prod with 85% power utilization and CCUS<sub>23%</sub>

(caption on next column)

**Fig. 6.** GWP from the combined tri-production with CCUS<sub>23%</sub> compared to the BaU cases, using mass allocation. Panel A shows the relative climate impacts for ternary mixes; each extreme point in the triangle represents 100% utilization to produce only one product and inner points represent product mixes. Panels B and C show variations in mass production of the two material products (H<sub>2</sub> and NH<sub>3</sub>) for different cases. Panel B shows a system with CCUS<sub>23%</sub> with no power production and Panel C shows a system with CCUS<sub>23%</sub> and 85% power utilization. In the ternary plot in Panel A colors show comparisons with the combination of the BaU benchmarks. Producing power during 85% of the time (Panel C and the iso-power line at 85% in Panel A) leads to all the three products from the tri-production to outperform the respective BaU substitute benchmark for the full range of H<sub>2</sub> - NH<sub>3</sub> mix (0–15%) production. In the emission curves (B–C) The arrows show the regions at which production of NH<sub>3</sub> (orange) and H<sub>2</sub> (green) from the tri-production generates less impacts than their respective BaU benchmark. Names marked with star (\*) represent impacts from the tri-production system. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

other emerging products and technologies. The market penetration and growth of installed capacity to synthesize these products, even in integrated systems, are crucial for the technological learning needed for them to mature and scale up. We show that technological improvement, when complemented with strategies to increase efficiency, can optimize the benefits from the current state of those technologies. This supports the idea that policies focused on improving industrial resource efficiency (e.g., promoting deployment of sector-coupling or incentivizing poly-generation in supply chains) can push emerging products into the market, based on quantifiable environmental impact reductions – even before they can generate those reductions in standalone deployment.

#### 4.2. Reconciling system expansion and physical allocation in an LCA for multifunctional systems

Enabling assessment of system-scale environmental benefits from multifunctional systems is crucial to justify investment in polygeneration. Polygeneration systems are expected to increase in the future [2, 71,117] partly motivated by their environmental performance benefits relative to single-product systems [2,118–120]. Standardized assessment techniques, like LCA, have a critical role in quantifying and communicating those benefits. Furthermore, systems-thinking approaches that analyze intersectoral interactions are necessary to screen, evaluate and optimize the design of new polygeneration solutions [121]. With our framework and case study, we contribute to the goal of improving the way the LCA community assesses the environmental performance of complex systems.

Our work shows that a tailored design of consequential and attributional LCA using different techniques to address multifunctionality can facilitate analysis of polygeneration systems, demonstrating the value of a flexible interpretation of ISO requirements for LCA studies (when not used for comparative, commercial purposes). With a series of attributional and consequential LCAs we analyze the consequences of implementing a coal-based H<sub>2</sub> polygeneration system and determine the impacts associated to one of the resulting products (H<sub>2</sub>-fuel) with a hybrid allocation methodology. This hybrid approach suits the analysis of a basket of products comprising material (e.g., H<sub>2</sub> and NH<sub>3</sub>) and non-material products (e.g., electricity), as we isolate the impacts of the non-material product and use a mass-allocation method to allocate impacts only to the material products. This subtraction is also adequate to represent the case where we have a determining product (electricity) and two dependent products in order to avoid economic allocation, which would require assumptions about future prices. Finally, that hybrid approach also allows to assess operational decisions that do not require structural changes in the system, without necessarily becoming a consequential analysis.

Our approach fits the nature of our system and the goal of our work

as it enabled us to appropriately represent the polygeneration system and our set of research questions related to 1) consequences of implementing polygeneration and 2) attribution of impacts to  $H_2$  fuel. This method is consistent with our goal and scope but is not compliant with the ISO 14040 and 14044 standards, as the standards recommend the user to select only one technique, preferably system expansion rather than allocation – regardless of the study goal. We structure our framework following the salient recommendation in the LCA literature of distinguishing between consequential and attributional LCAs and building the methodology over that basis [81–84]. After making this distinction, selecting system expansion to address the consequential question and allocation for the attributional was a logical methodological decision. However, our method exemplifies how a prescriptive selection of techniques continues to be potentially inconvenient even after the distinction between consequential and attributional is made: in our hybrid “adjusted allocation” approach we combine system expansion and allocation to match the physics and the business strategy in the polygeneration application we are studying. A more conventional partition method (either following the ISO standards or accepting a strict dichotomy between LCA categories and, as widely recommended in the literature, rejecting the use of substitution techniques in an attributional approach) [83,84] would require additional assumptions about a basket comprised of material and non-material products.

In our method, we also reconcile the idea that attributional LCAs can inform decision-makers about consequences from operational decisions furthering the idea of a spectrum between consequential and attributional LCAs rather than a discrete categorization. A secondary result from our attributional analysis allows us to study consequences from operational (i.e., micro) decisions. Using attributional LCA and allocation to assess short-term decisions is recommended in the International Reference Life Cycle Data System (ILCD) handbook [122], but that approach has been criticized [123]. In our case, we categorize this part of our analysis as attributional because the question in our partition analysis is about determining the impacts attributable to  $H_2$  fuel. However, by running a series of attributional analyses for different  $H_2$ – $NH_3$  proportions we identified overall changes in GHG impacts for different production points. These changes can be interpreted as consequences from varying production, allowing our analysis to inform operational decisions. Since both results (i.e., the attributional impacts of  $H_2$  and the differential overall impacts from decisions in the implementation of the polygeneration system) are relevant to our study objectives, we place our work somewhere in a ‘more continuous spectrum of approaches’ [124] rather than in a single category. However, if we must classify this work, it is attributional, given our initial objective and research question.

Our work contributes to the broader literature advocating for a flexible use of LCA methods and highlights a variety of inconsistencies and ambiguities in current standards [76–80,84], especially when dealing with multifunctionality. We recommend revisions to the ISO standards and relevant guidance that includes interpreting the most prescriptive sections as orienting principles rather than strict requirements when LCA is done for non-commercial purposes. In general, we emphasize that the goal and scope of the LCA should drive methodological decisions to better reflect the nature of the system. In particular, when addressing multifunctionality, we suggest choosing techniques that allow the modeler to represent the system under operational and economic conditions the analyst wants to study. Doing so, as done in this study, not only accurately represented a particular business model, but was necessary to identify operational conditions under which the system has improved environmental performance.

## 5. Conclusion

In our work, we provide evidence that the viability of coal-based  $H_2$  to reduce emissions compared to current products can in part be addressed by polygeneration. We demonstrated that the technological

requirements of CCUS in a combined, tri-production system are significantly lower compared to the requirements of independently producing  $H_2$ ,  $NH_3$ , and power. Fundamentally, current CCUS technologies are not sufficient for the standalone production of coal-derived  $H_{2,Th}$  and  $NH_3$  to reach the goal of producing lower life-cycle GHG emissions than uncontrolled natural gas and SMR  $NH_3$ , but that goal is achievable in a polygeneration set-up with the same CCUS technologies. Although our work shows the potential relative climate benefits of deploying polygeneration in a coal-based  $H_2$  production, a comprehensive assessment of environmental impacts is not in the scope of this work. A study measuring other indicators is therefore recommended to more broadly characterize system environmental performance.

To quantify the GWP impacts, we developed a framework that shows the potential to extend the use of LCA to analyze an alternative approach to deploying gasification technologies in integrated product systems. The results of our work provide useful insights about potential environmental benefits of a tri-production system built around  $H_2$ . By drawing an extended system boundary that includes multiple supply chains, we explored a wider set of environmental consequences from decisions about the system. We encourage LCA practitioners and technology analysts to take advantage of the system-thinking quality inherent to LCAs by analyzing emerging products in coupled applications.

Critically, our method demonstrates the utility of a flexible LCA approach for multifunctional product systems that address increasingly complex problems in industrial ecology. In our case, the combination of system expansion and allocation shows the potential of these techniques to help address different research questions when justified by the goal and nature of the LCA. We further contribute to the growing body of work [76–80,84] calling for LCA standards to provide orienting principles rather than prescriptive rules in the selection of multifunctional analytical techniques. We show that aligning methods selection with the goal definition and after an appropriate categorization (i.e., *attributional* or *consequential*) supports the definition of creative and consistent frameworks that allow addressing complex questions of multifunctional systems. However, we show that strict adherence to rules for technique selection, or adherence to a strict categorization without informed nuances may hinder the creativity necessary to address the study’s goal. Ultimately, our methods reject firm rules that constrain novel and useful analytical frameworks for quantitative life-cycle thinking. Addressing these issues with current LCA approaches will be essential in studying the technology for a more efficient, sustainable future.

## CRedit authorship contribution statement

**Diego Hincapié-Ossa:** Writing – original draft, Visualization, Methodology, Formal analysis, Data curation. **Daniel B. Gingerich:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Funding acquisition, Conceptualization.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Diego Hincapié-Ossa reports financial support was provided by National Science Foundation. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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expressed in this material are those of the authors and do not necessarily reflect the views of the National Science Foundation.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ijhydene.2024.12.356>.

Symbols

$H_2$ :	Hydrogen
$NH_3$ :	Ammonia
LCA:	Life cycle assessment
GHG:	Green House gas
GWP:	Global warming potential
CCUS:	Carbon capture, utilization, and storage
SMR:	Steam methane reformation
IGCC:	Integrated gasification combined cycle
BaU:	Business as usual
WGS:	Water gas shift
AGR:	Acid gas Removal
ASU:	Air separation unit
PSA:	Pressure Swing Adsorber
HRSRG:	Heat recovery steam generator
CT:	Combustion Turbine
ST:	Steam Turbine
CO:	Carbon Monoxide
T:	Metric tonnes
FU:	Functional unit
E:	Emissions of GHG
K:	Carbon captured
S:	Stack emissions
I:	Installed capacity
$\eta_{CCUS}$ :	CCUS capture efficiency (Fraction of the CO <sub>2</sub> stack flow that is captured. The used value is 0.85)
$f_{CCUS}$ :	CCUS energy efficiency (energy consumption per tonne of CO <sub>2</sub> captured. This quantity is variable in our switchover analysis).
G:	GHG emission intensity by unit of power
$Q_{CCUS}$ :	GHG emission intensity by unit of carbon captured in the CCUS
P:	Production
U:	Utilization factor (% of time)
$C_f$ :	Capacity factor (% of installed capacity units)
TRL:	Technology Readiness Index
<b>Subscripts:</b>	
$i$	Index for the different products
CCUS	Metric or process associated with the carbon capture, and utilization process
IGCC	Metric or process associated with the integrated gasification combined cycle process
PG	Metric or process associated with the polygeneration system
BAU	Metric or process associated with business-as-usual systems
##%	Subscripts as percentages reflect the energy penalty in the CCUS process
Th	Thermal
Elec	Electrical

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