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Integrating Hybrid Life Cycle Assessment with Multiobjective Optimization: A Modeling Framework

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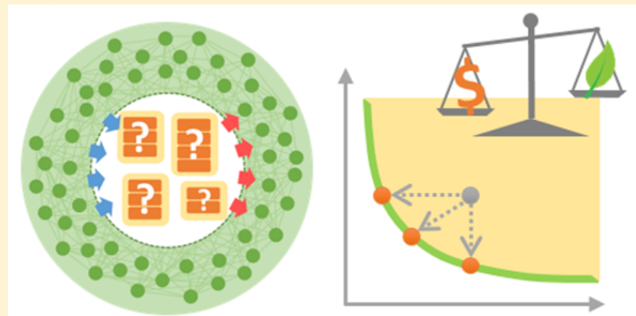
Supporting Information



ABSTRACT: By combining life cycle assessment (LCA) with multiobjective optimization (MOO), the life cycle optimization (LCO) framework holds the promise not only to evaluate the environmental impacts for a given product but also to compare different alternatives and identify both ecologically and economically better decisions. Despite the recent methodological developments in LCA, most LCO applications are developed upon process-based LCA, which results in system boundary truncation and underestimation of the true impact. In this study, we propose a comprehensive LCO framework that seamlessly integrates MOO with integrated hybrid LCA. It quantifies both direct and indirect environmental impacts and incorporates them into the decision making process in addition to the more traditional economic criteria. The proposed LCO framework is demonstrated through an application on sustainable design of a potential bioethanol supply chain in the UK. Results indicate that the proposed hybrid LCO framework identifies a considerable amount of indirect greenhouse gas emissions (up to 58.4%) that are essentially ignored in process-based LCO. Among the biomass feedstock options considered, using woody biomass for bioethanol production would be the most preferable choice from a climate perspective, while the mixed use of wheat and wheat straw as feedstocks would be the most cost-effective one.

1. INTRODUCTION

Life cycle assessment (LCA) is a well-recognized tool for evaluating the environmental impacts throughout a product's life cycle.[1] From cradle to grave, a product's life cycle includes sourcing of raw materials, logistics, production and use phases, and end-of-life disposal.[2] LCA has been widely used to develop sustainability strategies in both the public and private sectors.[3] Many companies and researchers use LCA to compare the sustainability performances of different alternatives and to provide guidance in long-term planning and policy making (e.g., Eco-Efficiency Analysis by BASF).[2,4] The typical procedure is enumerating all potential alternatives, performing LCA for each alternative, comparing their indicators of sustainability (e.g., global warming potential[5] and Eco-Indicator 99[6]), and then making the choice (e.g., selection of manufacturing technology and feedstock) according to designated sustainability criteria.[7] However, this approach becomes intractable when a large or even infinite number of alternatives are involved.[8] Aiming to tackle this challenge, life cycle optimization (LCO) framework was introduced, and the importance of a more extensive use of computational optimization tools in environmentally conscious decision making was established.[9] Optimization allows us to incorporate all potential decisions (e.g., selection of manufacturing technology, choice of plant location, and capacity of manufacturing process) into a mathematical model that consists of multiple objectives (e.g., cost, profit, cumulative energy consumption, emissions, and water consumption) and constraints (e.g., mass balance, stoichiometry, product specification, and resource availability).[10−12] Furthermore, the technique significantly facilitates the search for the desired sustainable solution by employing optimization algorithms.[13−16] Therefore, the combination of LCA with multiobjective optimization (MOO) allows one to conduct a thorough analysis of all potential alternatives and automatically identify both ecologically and economically better decisions within the LCO framework.[17,18]

Three common LCA methodologies can be potentially employed in an LCO study, namely process-based, input-output (IO)-based, and hybrid LCA.[19−21] Most LCO applications in the literature are developed upon the traditional process-based LCA for life cycle inventory (LCI) compilation.[22] The initiative to incorporate LCA objectives in process design and improvement was taken by Fava[23] and Azapagic.[24,25] Following their idea, many researchers have therefore undertaken process-based LCO studies aiming to simultaneously optimize both environmental and economic performances for systems at various scales.[26−29] Although process-based LCA provides more accurate and detailed process information, this

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"bottom-up" method results in system boundary truncation and underestimation of the true impact.[21] Since a large portion of the life cycle is neglected due to the truncated system boundary, any decisions made in process-based LCO studies are based on incomplete life cycle information and may not be truly optimal.[22] In contrast, IO-based LCA is a "top-down" method that relies on coarse and simplified models derived from highly aggregated empirical data.[30] It utilizes regional/national economic input-output (EIO) data coupled with averaged sectoral environmental impact factors.[31] While IO-based LCA has an expanded life cycle boundary, it lacks details at the process scale. Therefore, LCO studies based on IO-based LCA method are restricted to macroscopic analysis at national or global levels.[32,33] Hybrid LCA has been proposed to achieve a systematically complete LCA system and retain process specificity.[21] It combines the strengths of both process-based LCA and IO-based LCA and addresses their respective shortcomings, thus enabling us to quantify both direct and indirect environmental impacts in a detailed and comprehensive manner.[19,20,34]

So far, sustainability studies using hybrid LCA focused solely on analysis with static LCI data. Consequently, all processes and exchanges in the system are predetermined and fixed.[22] However, processes to be deployed in practice are influenced by many factors, for example, availability of feedstock, acceptance by market, economies of scale, and access to transportation modes.[35] Different decisions in various parts of a system can lead to distinct LCIs. In traditional process-based LCO studies, such changes are captured using fundamental process and supply chain models, but these benefits do not automatically extend to hybrid LCO. In a hybrid LCO study, the interactions do not merely exist within the process system boundary, but also among the numerous industrial sectors in the economy and between the processes and sectors across the process system boundary.

Very few sustainability studies have attempted to integrate all-encompassing flexible LCIs with MOO. As a pioneer in this area, Bakshi et al.[22,36] proposed a "Process-to-Planet" modeling framework that applies hybrid LCA to sustainable process design problems from an LCA perspective. They developed an integrated matrix formulation that incorporates models at the equipment, value chain, and economy scales in a consistent manner. A case study on bioethanol plant design was also provided. Our goal is to develop a general hybrid LCO modeling framework that allows for thorough analysis of all alternatives and assists in optimal decision-making based on a complete life cycle system. This work occupies a niche between the works that combine MOO with process-based LCA and the works that use single-objective optimization with hybrid LCA. With the aid of mathematical programming methods, the proposed framework is applied to a supply chain design problem, which can be closely related to existing works on process design and process improvement problems.[24,36−41] We employ integrated hybrid LCA method for the LCA component in our LCO framework, which is regarded as the state of the art in LCA and has a consistent and robust mathematical framework.[42] Overview of integrated hybrid LCA along with detailed mathematical models of the hybrid LCO framework are presented in the next section. We present an application on sustainable design of a potential bioethanol supply chain in the UK based on a multiregional input−output (MRIO) model.[43] Decisions such as facility location, technology selection, production planning, and logistics are optimized by simultaneously minimizing the total project cost and minimizing the sum of direct and indirect life cycle greenhouse gas (GHG) emissions. To the best of our knowledge, this is the first time that a hybrid LCO study has been conducted for sustainable supply chain design.

2. MATERIALS AND METHODS

2.1. Integrated Hybrid LCA. Since its definition by Suh[34] and Suh and Huppes,[21] many researchers have contributed to the development and application of integrated hybrid LCA.[42−44] On one hand, integrated hybrid LCA uses process-based LCA methodology to capture key life cycle processes. On the other hand, it complements the truncated process system boundary with IO-based LCA methods that include the macroeconomic system within which the processes operate. The resulting hybrid LCI, therefore, retains the level of detail and specificity from process-based LCA and has the completeness of an economy-wide system boundary from IO-based LCA.[21] Exchanges within the process system boundary are represented by a process matrix that describes the inputs of goods to processes in various physical units. Exchanges within the economy are represented by the direct requirements matrix that describes interdependencies among various industrial sectors in monetary units at a highly aggregated level.[45] The direct requirements matrix can be derived from regional/national EIO models consisting of a transaction matrix, a value added matrix, and a final demand vector.[46,47] Throughout the rest of this article, we will refer to the two systems above as the process system and the IO system, respectively. Exchanges across the process and IO systems are captured in upstream and downstream cutoffs matrices.[34] Upstream cutoff flows are inputs to the process system that are produced by industrial sectors in the IO system. These flows are typically specified in monetary units. Downstream cutoff flows are outputs from the process system that are consumed by industrial sectors in the IO system. These flows are typically specified in various physical units. Market price data are used to convert physical units of flows originating from the process system and monetary units of expenditures originating from the IO system. Let m be the number of processes/goods in the process system and n be the number of industrial sectors in the IO system. Using a matrix notation, the mathematical basis for integrated hybrid LCA is given by

$$\text{full environmental impact} = [e_p \quad e_{io}]\begin{bmatrix} A_p & -C_d \\ -C_u & I - A_{io} \end{bmatrix}^{-1}\begin{bmatrix} y \\ 0 \end{bmatrix} \quad (1)$$

where A_p is a square matrix representing the process inventory with dimension $m \times m$; A_{io} is the direct requirements matrix with dimension $n \times n$; I is an identity matrix with dimension $n \times n$; C_u is a matrix representation of upstream cutoffs to the process system with dimension $n \times m$; C_d is a matrix representation of downstream cutoffs to the process system with dimension $m \times n$; e_p is the process inventory environmental extension vector with dimension $1 \times m$; e_{io} is the IO environmental extension vector with dimension $1 \times n$; $[y \quad 0]^T$ is the functional unit column with dimension $(m + n) \times 1$, where all entries are 0 except for final products from the process system y.

Using a single direct requirements matrix for A_{io} is known as the industry-industry approach. This approach does not account for sectors that produce more than one commodity. In addition, supply and use tables (SUTs) can be used to

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construct A_{io}, which is known as the commodity-industry approach that provides greater flexibility in dealing with multiproduct processes.[34,42,43] Furthermore, MRIO model can be used for A_{io} to represent the global economy. Typically, a two-region model is formulated including the region where our processes operate and the other region called Rest of the World (ROW).[42,43,48] The negative sign assigned to C_u and C_d indicate the direction of cutoff flows across the process system boundary. Detailed procedures for constructing C_u and C_d along with techniques to avoid double counting can be found in the literature.[43,44,49−51]

For interested readers, we have proposed an illustrative example that compares the results of process-based LCA and integrated hybrid LCA. The problem is adapted from the classic toaster example by Suh.[34] Through the illustrative example, we demonstrate that ignoring the indirect impacts from the IO system may cause a considerable underestimation of the true impacts. A hybrid LCA model based on complete life cycle information is considered more appropriate for decision making.

2.2. Integrated Hybrid LCO. Consistent with the structure of integrated hybrid LCA, the proposed integrated hybrid LCO model also consists of four parts: process system, IO system, upstream cutoffs, and downstream cutoffs. As illustrated in Figure 1, the outer layer represents the IO system, and the

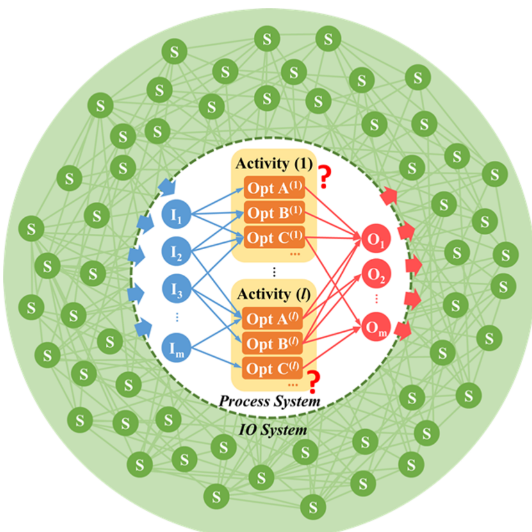

Figure 1. Illustration of Hybrid LCO framework.

inner layer represents the process system. The dashed circle represents the incomplete process system boundary, across which exchanges between the two systems are depicted. Upstream cutoffs are denoted by the thick arrows originating from the IO system on the left. Downstream cutoffs are denoted by the thick arrows originating from the process system on the right. Integration of the four parts thus provides us with a precise and comprehensive framework for decision making. Unlike the static LCI in LCA studies, all four parts of the proposed integrated hybrid LCO model are flexible. Different decisions made regarding the deployment of processes can lead to varying hybrid LCIs. This relationship is modeled by "activities" in the process system, which is defined as a flexible process that involves decision making. For example, a transportation activity can involve the selection of transportation modes and choice of shipping routes. Decisions made in activities directly influence the process LCI, and indirectly affect exchanges between the process and IO systems and exchanges among different industrial sectors in the IO system. Once the decisions in all activities have been made the hybrid LCI would be fixed, and the full environmental impact could be measured based on this hybrid LCI. The goal of this framework is to use mathematical programming methods to help make optimal decisions in terms of designated sustainability objectives while satisfying all specified constraints. This is achieved by combining integrated hybrid LCA with MOO, which allows simultaneous optimization under multiple sustainability objectives. In contrast to single-objective optimization, MOO leads to a series of Pareto-optimal solutions rather than a single optimal solution. These solutions possess the property that none of their objectives can be unilaterally improved without worsening at least one of the other objectives. In this regard, all of these solutions are optimal but emphasize different criteria.[52,53] A Pareto frontier can be obtained by plotting all Pareto-optimal solutions. Solutions on one side of the Pareto frontier are suboptimal, and solutions on the other side are unattainable. Therefore, solutions on the frontier indicate the best one can achieve in a sustainable design or improvement problem. It is worth noting that we can set the baseline to zero for design problems since the system is nonexistent before the design is implemented. In contrast, the choice of a baseline is critical to the improvement problems, where the alternative solutions must be compared with the original system to evaluate the impact of changes.

Instead of building our modeling framework using matrix formulation from an LCA perspective, we devise a general optimization model that seamlessly integrates the process system, IO system, and upstream and downstream cutoffs. This model allows us to describe complex systems with mathematical equations and parameters and to represent important decisions with different types of variables. As shown below, the environmental and economic objectives are given by (2) and (3); the process system is modeled by (4); the upstream cutoffs are calculated by (5); the IO system is modeled by (6); and the downstream cutoffs are calculated by (7). For clarification, all variables are denoted by upper-case letters and all indices and parameters with fixed values are denoted by lower-case letters.

$$\min \mathrm{Obj}_{\mathrm{env}} = \sum_m e_m^p Q_m + \sum_n e_n^{io} P_n \tag{2}$$

$$\min \mathrm{Obj}_{\mathrm{econ}} = \sum_l g_l(X_l, Y_l) \tag{3}$$

$$\text{s.t.} \quad Q_m = \sum_l f_{l,m}(X_l, Y_l), \ \forall \ m \tag{4}$$

$$U_n = \sum_m c_{n,m} p_m Q_m, \ \forall \ n, \ m \in In \tag{5}$$

$$P_n - \sum_{n'} a_{n,n'} P_{n'} \geq U_n, \ \forall \ n \tag{6}$$

$$Q_m \geq r_m + \sum_n d_{m,n} P_n, \ \forall \ m \in \mathrm{Out} \tag{7}$$

In the above integrated hybrid LCO model, we index the goods/processes in the process system by m and the industrial sectors in the IO system by n. The various activities within the process system are indexed by l. The environmental objective (2) minimizes the sum of direct and indirect environmental

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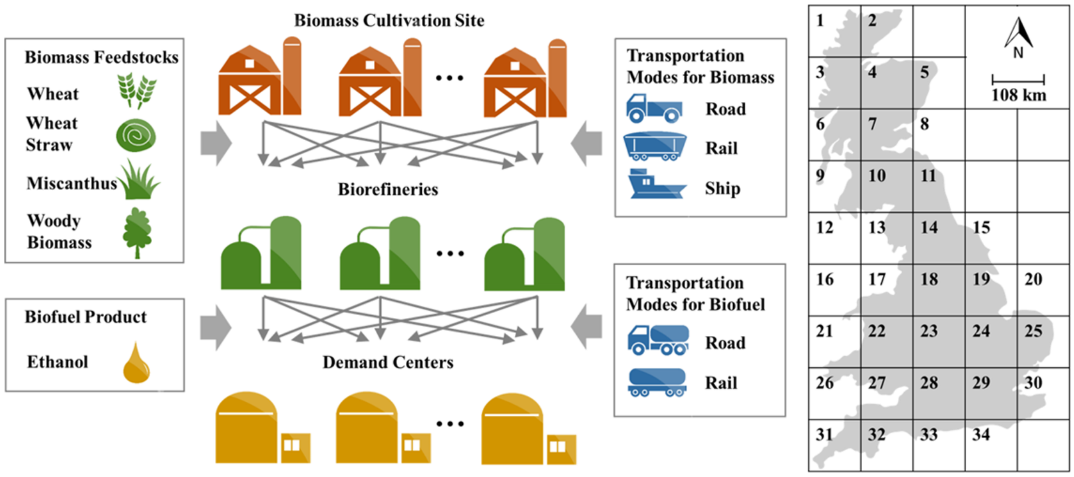

Figure 2. Superstructure of the UK advanced biofuel supply chain with the UK map discretized into 34 square cells.

impacts from the process and IO systems, where e_m^p and e_n^{io} are the environmental impact factors of process m and industrial sector n, respectively. Q_m is the total net input/output of good/process m from all activities in the process system. P_n is the total output of sector n. The economic objective (3) minimizes the total project cost, including the costs from all activities. $g_l(\cdot)$ is the cost evaluation function for activity l, which depends on the value of continuous variables X_l and integer variables Y_l. Continuous variables X_l can be used to model decisions on quantities of raw material acquisition and product sales, transportation flows, capacity of manufacturing processes, level of inventory, etc. Integer variables Y_l can be used to model decisions on the selection of facility location, manufacturing technology, capacity level, transportation mode, etc. Constraint (4) indicates that the process LCIs are dependent on the decisions in all activities, where $f_{l,m}(\cdot)$ stands for the mapping between process m and the decisions in activity l. Constraint (5) quantifies the upstream cutoffs originating from the IO system to the process system. We use $m \in In$ to denote the subset of goods m that are inputs to the process system. U_n is the total exchange of commodity from sector n to the process system. $c_{n,m}$ is the upstream technical coefficient, of which detailed derivation procedures are documented by Wiedmann et al.[43] and Acquaye et al.[44] p_m is the unit price of good m used to convert physical unit flows into equivalent expenditures. Constraint (6) indicates that the total output of each sector minus the direct requirements within the economy should satisfy the requirements by the process system, where $a_{n,n'}$ is the IO technical coefficients. It is worth noting that Leontief inverse[45] is not required because the sectoral output is explicitly denoted by $P_{n'}$ and optimization algorithms can directly handle the linear eq 6. Constraint (7) indicates that outputs produced from the process system must satisfy the external demand plus the consumption by various industrial sectors in the IO system. We use $m \in Out$ to denote the subset of products that are outputs from the process system. r_m is the external demand for product m. $d_{m,n}$ is the downstream technical coefficient, of which a detailed derivation can be found in the works by Suh.[34,50]

The nature of the resulting optimization problems depends on a number of factors. First, if any of the functions in $f_{l,m}(\cdot)$ or $g_l(\cdot)$ is nonlinear, the resulting optimization problem is a nonlinear program. Second, if any integer variables Y_l is involved, the resulting optimization problem is a mixed-integer program. Third, different applications typically lead to different optimization problems. For example, supply chain optimization often leads to mixed-integer programs while process design usually leads to nonlinear programs.[13] Solution of different types of mathematical programs requires corresponding optimization algorithms and solvers. A number of MOO techniques can be used to simultaneously optimize multiple objectives.[54] In this work, we choose the ε-constraint method for MOO, which is efficient in implementation.[55]

2.3. Uncertainty Analysis. LCA studies are conducted by an LCA analyst to support the decision maker in making sound choices. Therefore, it is critical to improve how uncertainty and variability is communicated in an LCA. As suggested by Herrmann et al.,[56] the expected uncertainty of an LCA statement (i.e., the answer to an LCA question or inquiry) is dependent on (1) the budget constraints for the LCA analyst, which is decided by the decision maker; (2) the size of the LCA space; and (3) the capability of the analyst. Assuming that the budget constraints and the capability of the analyst are constant, Hermann et al.[56] proposed a taxonomy to scale the expected level of uncertainty in LCAs. This taxonomy operates in six dimensions, and each dimension is read as a switch with two possible settings—either left or right. Specifically, the six dimensions and available settings are (1) tangibility—tangible (T) vs intangible (I); (2) repetitivity—single period (S) vs multiple periods (M); (3) scale—micro (i) vs macro (a); (4) time—retrospective (R) vs prospective (P); (5) change—baseline (B) vs change (C); and (6) value—physical (Y) vs value (V). By definition, the left settings corresponds to a lower uncertainty. As more dimensions are set to the right position of the switch, the expected inherent uncertainty increases. In contrast to most ex-post approaches that occurs after the LCA,[57−61] this taxonomy can be applied ex ante to an LCA study. We adopt this approach of uncertainty analysis in our hybrid LCO framework. Once the goal and scope of a hybrid LCO study is determined, we can classify the study using the taxonomy by Hermann et al.[56] to help better understand, rank and hence confront uncertainty for LCO.

3. RESULTS AND DISCUSSION

3.1. Problem Statement. We demonstrate the proposed integrated hybrid LCO framework with an application on sustainable design of a UK advanced biofuel supply chain in this section. Faced with increasing concerns on GHG emissions and

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energy security, the UK and the wider EU community have been setting out long-term strategies to promote biofuel production to substitute traditional fossil fuels.[62,63] To the best of our knowledge, existing hybrid LCA studies do not consider practical factors such as biomass availability, biorefinery capacity, and geographical variations, whereas existing sustainable biofuel supply chain optimization models that do consider these factors are built exclusively on process-based LCA.[8,64,65] We develop a first-of-its-kind hybrid LCO model for biofuel supply chain applications.

The techno-economic supply chain model is adopted from that by Akgul et al.,[66] of which the superstructure is given in Figure 2. The biofuel supply chain consists of three stages, namely biomass cultivation sites, biorefineries, and demand centers. Biomass feedstocks are acquired from biomass cultivation sites, where four types of biomass feedstocks are considered, namely wheat, wheat straw, miscanthus, and woody biomass (short rotation coppice). We assume that the annual biomass availability is stable and the soil has been in production throughout the planning horizon. Note that if the land has been fallow for a long time, tilling to prepare it for cultivation may release a pulse of CO_2.[67] Biomass feedstocks are converted into bioethanol using a biochemical route in biorefineries, where a pretreatment process (ammonia fiber explosion) is employed, after which lignocellulose can be hydrolyzed and then fermented. We consider four plant capacity levels with different capital costs. Depending on the biomass feedstock used in biorefineries, the conversion rate and operational cost vary. For the convenience of our data integration procedure, all monetary values are adjusted to the year of 2011 for inflation and other relevant financial corrections. Conversion to monetary values representing other years can be performed by using appropriate inflation indices (e.g., consumer price index). Credits of coproducts are implicitly accounted for in the parameters on operational cost. Bioethanol produced in biorefineries are sold to demand centers. We adopt one of the demand scenarios in the paper by Akgul et al.,[66] in which a total consumption rate of 2802 ton bioethanol/day are distributed to six demand centers in the UK. Three transportation modes are considered for shipping biomass feedstocks and bioethanol, namely road, rail, and ship. Specifically, the road and rail modes are for intercell transportation within the UK, the road mode is for local transportation within a cell, and the ship mode is for transoceanic transportation of imported biomass from foreign suppliers. As shown in Figure 2, we follow the approach by Akgul et al.[66] to discretize the UK map into 34 square cells, each with dimensions of 108 × 108 km. These cells are the potential locations of biomass cultivation sites, biorefineries, and demand centers. One dummy cell is considered to represent a foreign wheat supplier. All input data on biomass cultivation, bioethanol production, and transportation are given in the Supporting Information, which are taken from Akgul et al.[66]

A process system and an IO system constitute a complete life cycle boundary of this problem. The flexible hybrid LCI in this model is derived based on Ecoinvent v2.2[68] and the MRIO model in Wiedmann et al.[43] A total of 40 relevant processes are considered in the process system, each corresponding to an entry in the Ecoinvent database. The list of these 40 processes is presented in the Supporting Information, including chemicals, biomass, transport services, etc. Note that the CO_2 sequestration effect during biomass cultivation may vary depending on whether the soil had been in production previously or fallow.

Following the MRIO approach by Wiedmann and his coworkers, the IO system is built based on four tables, namely supply and use tables for the UK, and supply and use tables for the ROW. Each table contains 224 sectors/commodities, including mining, grain farming, power generation etc. Consequently, the resulting compound IO matrix has a dimension of 896 × 896 (896 = 4 × 224). The structure of the compound matrix was provided in the Supporting Information of the work by Wiedmann et al.[43] Following the approach by Wiedmann et al.,[43] upstream technical coefficients in the model are derived by modifying the corresponding IO technical coefficients. Sectoral inputs already considered in the process system are nullified to avoid double counting. All unit prices for converting physical unit inputs to equivalent expenditures are taken from the original MRIO model.[43] The downstream cut-offs are neglected as suggested by a number of researchers.[44,49−51] Six groups of GHGs in the Kyoto Protocol are considered, namely CO_2, CH_4, N_2O, HFCs, PFCs, and SF_6.[43] The GWP damage model[5] is used to generate an aggregated indicator in the unit of kg CO_2 equivalent. GHG emissions factors for all processes in the process system are obtained from Ecoinvent v2.2,[68] and GWP factors for all industrial sectors in the IO system are obtained from the original MRIO model.[43] Note that indirect changes or intangible effects, such as indirect land use changes (ILUC), are not considered in this problem for the simplicity of demonstration. However, the proposed hybrid LCO framework is general that such impacts can be easily incorporated by considering additional parameters, variables, and equations in the optimization model.

We consider an environmental objective and an economic objective in this integrated hybrid LCO model. The environmental objective is to minimize the full life cycle GHG emissions resulting from all supply chain activities, including emissions from both process and IO systems. Note that there are other important environmental- and policy-relevant impact indicators than GHG emissions, including acidification, nitrification, resource depletion, respiratory inorganics, land use, human toxicity, etc. We consider one environmental objective for the simplicity of demonstration, whereas additional impact indicators can be easily incorporated in the hybrid LCO framework via MOO. The economic objective is to minimize the total project cost, including the investment cost, production cost, transportation cost, and import cost. This cost is assumed to be borne by the biofuel manufacturer and does not include externalities. The aim is to simultaneously minimize the economic and environmental objectives by optimizing the following decisions:

- Biomass acquisition rate of each type of biomass feedstocks from biomass cultivation sites and foreign suppliers;
- Selection of locations and capacity levels for biorefineries;
- Production rate of bioethanol and consumption rate of biomass at biorefineries;
- Selection of transportation modes and shipping routes for biomass and bioethanol;
- Transportation flows of biomass feedstocks and bioethanol between all facilities.

The resulting optimization problem is formulated as a bicriterion mixed-integer linear programming (MILP) problem. A detailed model formulation is provided in the Supporting

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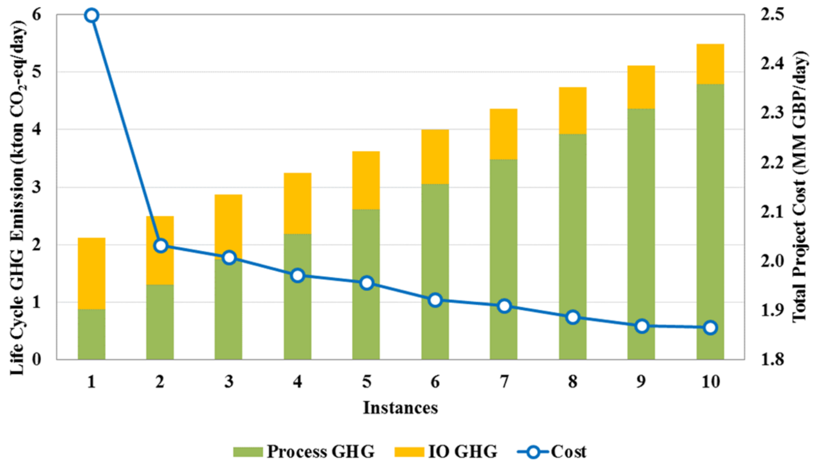

Figure 3. Pareto profile with the total project cost and life cycle GHG emissions for 10 instances.

Information. The MILP problems were solved by the solver CPLEX.

According to the taxonomy by Herrmann et al.,[56] this hybrid LCO study is classified as ISa-PCY. The study is classified as (1) "intangible (I)" because the actual correlations among supply, production and demand can be very complicated (e.g., competition with petrochemical fuels, legal reasons, CO_2 emission taxes) and might not be accurately captured in the model; (2) "single period (S)" because the annual planning model considers only a single period; (3) "macro (a)" because the supply chain under study is at a national scale; (4) "prospective (P)" because the construction and operation of this supply chain will take place in the future; (5) "change (C)" because the development of this supply chain will change the renewable energy supply; and (6) "physical (Y)" because the facility locations as well as the quantities of materials, energy and GHG emissions are specified in the model.

3.2. Optimization Results. Following the ε-constraint method for MOO, we have solved 10 cost-minimization instances with the cap on total life cycle GHG emissions set at 10 different values evenly distributed between its minimum and maximum values. Since this is a design problem, we consider a baseline with zero life cycle GHG emissions and zero total project cost. As shown by Figure 3, the LCO results indicate significant trade-offs between the economic and environmental objectives. The stacked column chart shows the breakdowns of process and IO life cycle GHG emissions of all instances. The line chart shows the total project costs of all instances and represents the Pareto frontier. As the cap on the total life cycle GHG emissions reduces from 5488 to 2128 ton CO_2-equivalent/day, the total project cost climbs from £ 1.87 to £ 2.50 MM/day. We observe that the total project cost increases rapidly as the cap on full life cycle GHG emissions goes below approximately 2500 ton CO_2-equivalent/day. All solutions above the Pareto frontier are suboptimal, and all solutions below this frontier are unattainable. While all solutions on the Pareto frontier are optimal, solutions on the left emphasize more on GHG mitigation, and solutions on the right tend to achieve a more cost-effective supply chain design. Specifically, the point at the upper left corner (Instance 1) has the lowest full life cycle GHG emissions, so it is considered as the most preferable solution from a climate perspective; the point at the lower right corner (Instance 10) has the lowest total project cost, so it is considered as the most cost-effective solution. One can choose any preferred optimal solution on the Pareto frontier. It is also worth noting that the ratio of process GHG emissions over IO GHG emissions varies over the 10 instances, as shown by the stacked columns. At Instance 1, the process GHG emissions contribute 41.6% of the full life cycle GHG emissions, and the IO GHG emissions contribute 58.4%. In contrast, at Instance 10, the process GHG emissions contribute 87.2% of the full life cycle emissions, and the IO GHG emissions contribute 12.8%. This difference is due to the different decisions made in the supply chain system, and we will provide detailed discussions on these two extreme solutions in the following paragraphs.

Results of Instance 1 are obtained by minimizing the full life cycle GHG emissions without setting any budget on the project cost. The corresponding supply chain configuration is given in Figure 4a. A total of six biorefineries are built in the same cells

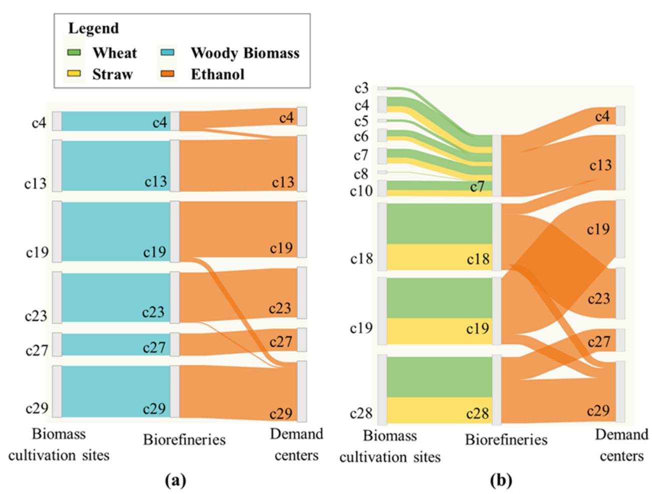

Figure 4. Optimal supply chain configuration and material flows between the facilities for (a) the solution with minimum full life cycle GHG emissions, and (b) the solution with minimum total project cost.

as the locations of demand centers. Two biorefineries are at capacity level 1, three at capacity level 3, and one at capacity level 4. The daily demand of 2802 tons of bioethanol is exclusively produced from 11 924 tons of woody biomass. This result indicates that producing bioethanol from woody biomass is the option that leads to the fewest life cycle GHG emissions. All woody biomass is acquired locally in the same cells as the locations of biorefineries in order to avoid long-distance

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biomass transportation. Rail is the preferred mode of transportation for shipping bioethanol from biorefineries to demand centers because of the lower unit transportation GHG footprint compared to road transport. The total project cost of this solution levelized on a daily basis is £ 2.50 MM/day. The major cost comes from production, which accounts for 77% of the total project cost; the investment cost levelized on a daily basis accounts for 16%; and the sum of local and inter-region transportation costs collectively accounts for 7%. The sum of direct and indirect GHG emissions of this solution is 2128 ton CO_2-equivalent/day. Life cycle GHG emissions profiles are given in the Supporting Information, where we summarize the GHG emissions from each process in the process system, and each industrial sector in the UK IO system and ROW IO system. In the process system, the major contributor is the production of ammonia, which results in 387 ton CO_2-equivalent/day. The conversion of woody biomass to bioethanol requires significant use of ammonia during biomass pretreatment, and the ammonia manufacturing process is energy-intensive. The second largest contributor in the process system is from acquisition of the biomass feedstock—woody biomass, which results in 133 ton CO_2-equivalent/day. These GHG emissions are primarily due to chemical and energy use in the cultivation, collection and preparation phases of the woody biomass feedstock. In the IO system, the major contributor is the sector of natural gas and services in the ROW, which results in 205 ton CO_2-equivalent/day, reflecting the fact that imported natural gas plays a significant role in the UK's energy supply. The second largest contributor in the IO system is the sector of electricity production from coal in the UK, which results in 147 ton CO_2-equivalent/day, reflecting the fact that majority of the power supply in the UK comes from coal-fired power plants.

On the opposite side of the solution above, results of Instance 10 are obtained by minimizing the total project cost without setting any cap on the life cycle GHG emissions. The corresponding supply chain design is shown in Figure 4b. A total of four biorefineries are built, and all biorefineries are at capacity level 4 in order to take advantage of economies of scale. The daily demand of 2802 tons of bioethanol is produced from 5701 tons of wheat and 3589 tons of wheat straw. In this solution, wheat is chosen as the major biomass feedstock as it has the lowest unit production cost compared to other types of biomass feedstocks. Since the availability of domestic wheat does not suffice the total requirement for bioethanol production, wheat straw is chosen as the secondary biomass feedstock. Wheat straw is a coproduct of wheat acquisition, thus having a lower acquisition cost than miscanthus and woody biomass. As shown in Figure 4b, three of the biorefineries source their biomass feedstocks from local biomass cultivation sites. The only exception is the biorefinery in cell 7, which obtains most biomass feedstocks from biomass cultivation sites in the surrounding cells. Rail is the preferred transportation mode for shipping both biomass and bioethanol, because it has a lower unit transportation cost compared to road transport. The total project cost of this solution levelized on a daily basis is £ 1.87 MM/day. The production cost accounts for 70% of the total project cost; the investment cost accounts for 18%; and the transportation cost accounts for 12%. The ratio of investment cost over production cost is increased compared to Instance 1, because of the deployment of larger-size biorefineries that benefit from economies of scale. The transportation cost is 5% higher than that at Instance 1 due to the long-distance transportation of wheat and wheat straw at Instance 10. The full life cycle GHG emissions of this solution is 5488 ton CO_2-equivalent/day, which is more than twice of that of Instance 1. Life cycle GHG emissions profiles are given in the Supporting Information. In the process system, the major contributor is acquisition of wheat, which results in 2929 ton CO_2-equivalent/day. The second largest contributor is acquisition of wheat straw, which results in 728 ton CO_2-equivalent/day. Both biomass feedstocks require significant use of energy, water, and fertilizers during the cultivation and acquisition phase. In the IO system, the major contributor is the sector of electricity production from coal in the UK, which results in 145 ton CO_2-equivalent/day, and the second largest contributor is the sector of natural gas and services in the ROW, which results in 94 ton CO_2-equivalent/day. This distribution of emissions is similar to that of the previous solution, reflecting that natural gas and power are the major sources of GHG emissions.

3.3. Comparison with Other Studies. We briefly review the hybrid LCA results on bioethanol production from other studies in this section. To facilitate the comparison, we convert all life cycle GHG emissions data to a unit of energy, that is, GJ bioethanol. The specific energy of bioethanol is assumed to be 23.4 MJ/kg and the energy density is 18.4 MJ/L.[69] According to these assumptions, the lowest life cycle GHG emissions in our LCO results is 32.45 kg CO_2 equivalent/GJ at Instance 1, and the highest life cycle GHG emissions is 83.69 kg CO_2 equivalent/GJ at Instance 10.

Bright et al.[70] undertook an environmental assessment of wood-based biofuel production to evaluate the GHG mitigation potentials under different consumption scenarios in Norway using a hybrid biregion LCA method. The reported full life cycle GHG emissions are 21 and 27 kg CO_2 equivalent/GJ for bioethanol production via thermochemical conversion and biochemical conversion, respectively.

Acquaye et al.[42] calculated the life cycle GHG emissions of biodiesel and bioethanol produced from various biomass feedstocks in UK using a hybrid MRIO LCA framework. The reported sum of process and IO GHG emissions are 25.1, 29.1, and 72.9 kg CO_2 equivalent/GJ for bioethanol produced from sugar cane, sugar beet, and corn, respectively. Specifically, the IO GHG emissions account for 36.7%, 8.6%, and 3.6% of the full life cycle GHG emissions for bioethanol produced sugar cane, sugar beet, and corn, respectively.

Palma-Rojas et al.[71] evaluated the energy use, life cycle GHG emissions, and employment impact of bioethanol production from bagasse in Brazil using the integrated hybrid LCA method. The reported sum of process and IO GHG emissions is 60.0 kg CO_2 equivalent/GJ, where the IO GHG emissions account for 49.3% of the full life cycle GHG emissions.

In summary, the full life cycle GHG emissions values obtained from our hybrid LCO study are on the same order of magnitude as the literature values reviewed above. Differences in values are due to the choice of data set and biomass feedstocks. Consistent with our hybrid LCO results, these literature values also suggest that the unit life cycle emissions as well as the ratio of process emissions over IO emissions vary significantly by the biomass feedstocks used for bioethanol production.

3.4. Limitations. A limitation of the proposed hybrid LCO framework is that it has been tested on only a limited number of case studies. More examples are needed to demonstrate the applicability of the proposed framework. Another limitation lies

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in that hybrid LCO study requires much more efforts in data collection and data processing compared to traditional process-based LCO. The policy relevance of the optimization results is dependent on the quality of the data used.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.5b04279.

Detailed mathematical model formulation, notations and input/output data for the case study (PDF) (XLSX)

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Notes

The authors declare no competing financial interest.

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