A bi-criterion optimization approach for the design and planning of hydrogen supply chains for vehicle use

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Abstract

In this paper, we address the design of hydrogen supply chains for vehicle use with economic and environmental concerns. Given a set of available technologies to produce, store and deliver hydrogen, the problem consists of determining the optimal design of the production-distribution network capable of satisfying a predefined hydrogen demand. The design task is formulated as a bi-criterion MILP, which simultaneously accounts for the minimization of cost and environmental impact. The environmental impact is measured through the contribution to climate change made by the hydrogen network operation. The emissions considered in the analysis are those associated with the entire life cycle of the process, and are quantified according to the principles of Life Cycle Assessment (LCA). To expedite the search of the Pareto solutions of the problem, we introduce a bi-level algorithm that exploits its specific structure. A case study that addresses the optimal design of the supply chain capable of fulfilling the expected future hydrogen demand in Great Britain is introduced to illustrate the capabilities of the proposed approach.

Keywords: Supply chain management, optimization, sustainability, hydrogen, life cycle assessment.

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Introduction

The use of hydrogen as an alternative fuel and energy carrier is receiving increasing interest as the environmental impact of hydrocarbons becomes more evident. Hydrogen can be produced safely, is environmentally friendly and has many potential energy uses, including powering road vehicles and aircrafts, and heating homes and offices. In particular, its use as a transportation fuel in fuel cell vehicles offers potentially attractive advantages over existing energy sources, particularly regarding emissions of greenhouse gases over the entire life cycle.[1]

Nowadays, most of the hydrogen is produced in petroleum refineries or in the chemical industry, mostly via steam reforming. The hydrogen obtained is usually consumed on-site rather than sold on the market, and is mainly used as a feedstock for petroleum refining and for the manufacture of ammonia fertilizer, plastic, resins, solvents, and other industrial commodities. Little hydrogen is currently utilized as an energy source, or as an energy carrier to transport energy from the production to the consumption sites. Specifically, only about 5 % of hydrogen is considered as "marketable" and delivered elsewhere as a liquid or gas by truck or pipeline.[2] Thus, there is still a large gap to achieve the transition from the current fossil-based economy towards a new one based on hydrogen.

The adoption of hydrogen in the current energy system depends to a large extent on the ability to solve the technological problems posed by the aforementioned transition process. In this regard, one of the key issues that still remains open is how to determine the optimal structure of the network capable of fulfilling the growing hydrogen demand in the existing markets. In this context, minimizing exclusively the total cost may lead to solutions that do not fully exploit the environmental benefits of switching to a more sustainable energy system. To avoid this situation, the design task must be posed as a multi-criteria decision-making problem, which allows for the simultaneous assessment of environmental and economic concerns at the early stages of the process development.[1] Unfortunately, little research has been conducted to date in this area.

The aim of the present work is to address the environmentally conscious design of hydrogen networks. The design task is formulated as a bi-criterion mixed integer linear program (mo-MILP) that accounts for the simultaneous minimization of total cost and environmental impact. A tailor-made decomposition strategy that reduces the computational burden of the model by exploiting its mathematical structure is also presented. The capabilities of the proposed approach are illustrated through a case study based on a real scenario, for which the set of Pareto efficient solutions are calculated.

The paper is organized as follows. In first place, a literature review of the topic is presented. A formal definition of the problem under study is next given, along with the associated mathematical formulation. The following section introduces the decomposition strategy employed to expedite the search of the Pareto solutions of the problem. The capabilities of the proposed modeling framework and solution strategy are next illustrated through a case study, and the conclusions of the work are finally drawn.

Literature review

Driven by environmental concerns and energy security, the idea of a "hydrogen economy" has been gaining followers not only in the scientific and engineering areas but also in politics and businesses.[3] Several converging forces explain the interest in hydrogen. First, the technological advances made in different fields such as fuel cells, which are regarded as the potential successors to batteries, power plants, and internal combustion engines. Secondly, the growing competition in the energy industry. Finally, in third place but maybe of greater importance, are the energy-related problems such as the energy security, air pollution, and climate change, which question the sustainability of the current energy system. In this regard, hydrogen would play a fundamental role in reducing worldwide CO_2 emissions, thus contributing to avoid global warming.

To promote the hydrocarbon economy, government and industry are encouraging the use of gasoline and methanol as sources of hydrogen. A cleaner path based on obtaining hydrogen from natural gas and renewable energy, or using the fuel directly on vehicles, has received much less support, in part because the cost of building a hydrogen infrastructure is widely viewed as prohibitively high. However, several recent studies indicate that the transition towards a hydrogen energy system may be much cleaner and far less expensive than expected.[2]

Iceland was one of the first pioneers in studying the feasibility and advantages of a hydrogen economy, announcing in 1999 its intention to become the world's first hydrogen society.[4] Hawaii and the South Pacific island of Vanuatu, have also promoted the hydrogen economy, whereas in China, the use of polygeneration using coal as a feedstock may become an economic source of hydrogen.[5,6]

In the private sector, over 100 companies are seeking to commercialize fuel cells for a broad range of applications. Hydrogen is being researched for direct use in automobiles and planes and the most important suppliers of energy and automobile manufacturers are creating divisions of hydrogen and making significant investments in this area.[7]

Unfortunately, despite the potential benefits, and the previous attempts towards a hydrogen economy, its full realization faces a number of social, technical and economic obstacles. Hydrogen still needs to be included into the energy policies and strategies of administration, which tends to preserve the hydrocarbon-based statu quo. Furthermore, today's energy market leads to artificially low fossil fuel prices and encourages the production and use of those fuels, making it difficult for hydrogen and fuel cells to compete with the deep-rooted gasoline-run internal combustion engines and coal-fired power plants.[8] Ogden and her colleagues[9] point out that environmental considerations will be a key issue in the hydrogen transition that is expected to take place in the automotive industry. In this regard, the adoption of strong policies such as zero-emission targets or fiscal incentives will be a critical determinant of the success of fuel cell vehicles.

One of the most significant obstacles to achieve the hydrogen transition is the general perception that a hydrogen supply chain (SC) - the system for producing, storing, and delivering the gas - would be prohibitory expensive to build, in comparison with a system based on gasoline. Thus, automotive and energy companies are investing millions of dollars in the development of reformer and vehicle technologies to obtain and use hydrogen, keeping the current petroleum-based infrastructure unchanged.[10] Along these lines, Jensen and Ross[11] state that the widespread introduction of hydrogen into car fleets faces three main technical challenges. The first is integrating small, inexpensive, and efficient fuel cells into the vehicles. The second is the improvement of storage technologies. Finally, the third and most important one in terms of economic and environmental impact, is developing an efficient infrastructure for producing and delivering hydrogen.

Much of the early work in the area of hydrogen supply chains has been promoted by US legislative pressures, like was done in California in order to improve air quality. In this context, Ogden et al.[9,12] examined different short-term options for producing and delivering compressed gaseous hydrogen through commercially available technologies for production, storage, and distribution. For a longer term, Ogden[13] studied other centralized methods for hydrogen production, including gasification of biomass, coal or municipal solid waste, or electrolysis based on wind, solar and nuclear energy.

Furthermore, the design of hydrogen supply chains has also attracted increasing attention in the UK. Guy[14] examined the development of a logistic infrastructure in London and the Southeast, whereas Joffe and coworkers[15] presented a methodology for modeling a hydrogen infrastructure for refueling buses in London and an analysis of the technical issues for installing a hydrogen facility. The authors concluded that the endeavor is both feasible and economically attractive, despite some technical challenges that will be overcome in the near future as technology progresses.

The performance of different pathway options to produce and deliver hydrogen has also been assessed in other comparative studies.[16,17] Although these approaches provide valuable insights into the hydrogen infrastructure, they usually restrict the analysis to a reduced number of options, which represents a major limitation. The alternative to these methods is to develop mathematical programming models capable of generating and assessing in a systematic way a very large number of process alternatives.

In one of the first works in this area, Van Den Heever and Grossmann[18] discussed the integration of production planning and scheduling for the optimization of a hydrogen supply network. This work addressed the operational level of an existing network but did not take into account design aspects.

Hugo et al.[1] introduced a generic optimization-based model for the optimal design and planning of hydrogen infrastructures. This model utilized formal optimization techniques to assess diverse process alternatives in terms of investment and environmental impact. The model applied an end-of-pipe approach that aimed at reducing the well-to-wheel greenhouse gas emissions.

More recently, Almansoori and Shah[19] presented a mathematical programming approach to design and operate a future British hydrogen supply chain.

Despite the studies and projects devoted to hydrogen, most of them focus on a particular component of the hydrogen supply chain, such as technologies for production, storage or distribution, instead of adopting a systems approach for designing and operating the whole infrastructure as a single entity.[19] These simplifications aim at reducing the complexity of the problem, the size of which grows exponentially as one enlarges the scope of the analysis to include other echelons of the hydrogen supply chain. Moreover, another disadvantage of the aforementioned works is that the joint analysis of the technical feasibility, economic and environmental issues is usually omitted.

With the aim to overcome these limitations, this work presents a holistic approach to address the optimal design of hydrogen supply chains with environmental and economic concerns. Our method relies on the combined use of mathematical programming and LCA principles, which enables the automatic generation and assessment of process alternatives that may lead to significant environmental and economic benefits. The approach presented is complemented with an efficient solution method that makes it possible to address large-scale problems arising in real-world situations.

Problem statement

The design problem addressed in this article has as objective to determine the configuration of a three-echelon hydrogen network for vehicle use (production-storage-market) with the goal of minimizing the cost and environmental impact.

The structure of the three-echelon SC taken as reference in this work is depicted in Figure 1. This network includes a set of plants, where hydrogen is produced, and a set of storage facilities, where hydrogen is stored before being delivered to the final customers. We assume that the overall region of interest (e.g., a country, a continent, etc.) is divided into a set of grid squares of equal size that correspond to different subregions of the original region of interest. The SC entities can then be located in any of these grids, each of which is characterized by a given hydrogen demand. Hence, the set of grids of the problem along with the associated geographical distribution of the demand must be provided as input data by the decision maker. The environmentally conscious network design problem can therefore be stated as follows.

Given are a fixed time horizon, the demand of hydrogen in each grid and time period, investment costs and capacity limitations of plants and storage facilities, costs associated with the network operation (raw materials, operating, transportation and inventory costs), environmental data (emissions associated with the network operation and damage model) and interest rate.

The goal is to determine the SC configuration along with the planning decisions that simultaneously minimize the total cost and environmental impact. The major decisions include:

- Structural decisions: number, type of process technology, location and capacity of plants and storage facilities; number and type of transportation units (e.g, tanker trucks, railway tube cars, etc.) and transportation links to be established between the SC entities.
- Planning decisions for each time period: production rates at the plants; inventory levels at the storage facilities; flows of hydrogen between plants and storage facilities and sales of products.

The mathematical formulation proposed to address this problem is described in the following section.

Mathematical model

The model presented is inspired on the work of Almansoori and Shah[19] , in which the authors proposed a steady state "snapshot" formulation of a hydrogen network that considered a time-invariant demand. Specifically, our model modifies and extends the original one in order to account for the evolution of the network over time and a time-variant demand. Furthermore, an additional feature of our formulation is the inclusion of environmental concerns that are considered along with the traditional economic objective. This consideration has led to a bi-criterion decision-making problem, the solution of which comprises a set of Pareto optimal points that trade-off cost and environmental impact. Our model includes four main sets of equations that are next described in detail.

Mass balance constraints

The mass balance must be satisfied in every grid and time period. Thus, for every hydrogen form i, the initial inventory kept in a grid (S_{igt-1}) plus the amount produced (PR_{igpt}) and the input flow rate ($Q_{ig'glt}$), must equal the final inventory (S_{igt}) plus the amount delivered to the customers (D_{igt}) and the output flow rate ($Q_{igg'lt}$).

$$\sum_{s \in SI(i)} S_{igst-1} + \sum_{p} PR_{igpt} + \sum_{g' \neq g} \sum_{l} Q_{ig'glt} = \sum_{s \in SI(i)} S_{igst} + D_{igt} + \sum_{g' \neq g} \sum_{l} Q_{igg'lt} \qquad \forall i, g, t \tag{1}$$

In this equation, $SI(i)$ represents the set of technologies that can be used to store product form i. Furthermore, the total amount of hydrogen in any form i consumed in grid g in period t must be lower than the hydrogen demand in that location ($\overline{D_{gt}}$) and greater than the minimum demand satisfaction level ($dsat$):

$$\overline{D_{gt}} dsat \leq \sum_{i} D_{igt} \leq \overline{D_{gt}} \qquad \forall g, t \tag{2}$$

Capacity constraints

Plants

The capacity of each plant technology p that produces product form i at grid g in period t is represented by a continuous variable denoted by C^{PL}_{gpt}. Equation 3 constraints the total production rate of technology p (PR_{igpt}) to be lower than the existing capacity and higher than a minimum desired percentage, τ, of the available installed capacity:

$$\tau C^{PL}_{gpt} \leq \sum_{i} PR_{igpt} \leq C^{PL}_{gpt} \qquad \forall g, p, t \tag{3}$$

The capacity of technology p at grid g in any time period t is calculated from the existing capacity at the end of the previous period plus the expansion in capacity carried out in t:

$$C^{PL}_{gpt} = C^{PL}_{gpt-1} + CE^{PL}_{gpt} \qquad \forall g, p, t \tag{4}$$

In this equation, CE^{PL}_{gpt} represents the expansion in capacity of plant technology p executed in period t at grid g.

Equation 5 is applied to bound the capacity expansions within lower and upper limits. These limits are calculated from the number of plants installed in the grid (N^{PL}_{gpt}) and the minimum and maximum capacities associated with each technology p ($\underline{PC^{PL}_{p}}$ and $\overline{PC^{PL}_{p}}$, respectively).

$$\underline{PC^{PL}_{p}} N^{PL}_{gpt} \leq CE^{PL}_{gpt} \leq \overline{PC^{PL}_{p}} N^{PL}_{gpt} \qquad \forall g, p, t \tag{5}$$

Storage facilities

The storage capacity of product form i during period t in grid g associated with technology s is represented by the continuous variable C^{ST}_{gst}. Equation 6 forces the total inventory of product in form i kept at the end of any period t at the storage facilities of type s installed in grid g to be lower than the available capacity.

$$\sum_{i \in IS(s)} S_{igst} \leq C^{ST}_{gst} \qquad \forall g, s, t \tag{6}$$

In this equation, $IS(s)$ denotes the set of product forms i that can be stored by technology s. Furthermore, the amount of hydrogen delivered by the storage facility to the customers is constrained by its capacity. Thus, this work considers that the capacity required to handle a given amount of hydrogen, assuming regular shipment and delivery schedule, is twice the average storage inventory level kept at the storage facility.[20] During steady-state operation, the average inventory of a product form i in grid g, is determined from the amount delivered to customers (D_{igt}) and the storage period θ. This storage period is introduced to cover fluctuations in both supply and demand as well as plant interruptions:[19]

$$2\left(\theta D_{igt}\right) \leq \sum_{s \in SI(i)} C^{ST}_{gst} \qquad \forall i, g, t \tag{7}$$

Finally, the capacity of the storage technology at any time period is determined from the previous one and the expansion in capacity executed in the same period (CE^{ST}_{gst}):

$$C^{ST}_{gst} = C^{ST}_{gst-1} + CE^{ST}_{gst} \qquad \forall g, s, t \tag{8}$$

Similarly, as with the manufacturing plants, the value of CE^{ST}_{gst} is bounded within lower and upper limits. These bounds are given by the number of storage facilities installed in the grid (N^{ST}_{gst}) and the corresponding minimum and maximum storage capacities associated with each storage technology s ($\underline{SC^{ST}_{s}}$ and $\overline{SC^{ST}_{s}}$, respectively), as stated in Eq. 9:

$$\underline{SC^{ST}_{s}} N^{ST}_{gst} \leq CE^{ST}_{gst} \leq \overline{SC^{ST}_{s}} N^{ST}_{gst} \qquad \forall g, s, t \tag{9}$$

Transportation constraints

In this block of equations, we make use of the binary variable $X_{gg'lt}$, which takes a value of one if a transportation link of type l (i.e., tanker trucks, railway tuber cars, etc.) is established between grids g and g' in time period t, and zero otherwise. The definition of such a variable is enforced via Eq. 10.

$$\underline{QC_{lgg'}} X_{gg'lt} \leq \sum_{i} Q_{igg'lt} \leq \overline{QC_{lgg'}} X_{gg'lt} \qquad \forall g, g'(g \neq g'), t \qquad \forall l \in LI(i) \tag{10}$$

Note that a zero value of the aforementioned binary variable prevents the flow of those materials that can be transported via technology l ($l \in LI(i)$) from taking place, whereas a value of one allows the transport flows within some lower ($\underline{QC_{lgg'}}$) and upper limits ($\overline{QC_{lgg'}}$). Furthermore, a grid can either import or export hydrogen, but not both at the same time. This is because if a grid can only satisfy its needs by importing from other grids, it would not make sense for that grid to export to other grids:

$$X_{gg'lt} + X_{g'glt} \leq 1 \qquad \forall g, g'(g \neq g'), l, t \tag{11}$$

Objective function

The model must optimize the economic and environmental performance of the network. The economic objective is represented by the total discounted cost, whereas the environmental impact is measured through its contribution to climate change.

Total cost

The total cost (TDC) is calculated as the summation of the discounted costs associated with each time period:

$$TDC = \sum_t \frac{TC_t}{(1+ir)^{t-1}} \tag{12}$$

In this equation, ir represents the interest rate and TC_t is the total amount of money spent in period t, which includes the capital (FCC_t, TCC_t) as well as operating costs (FOC_t, TOC_t) given by the production, storage and transportation facilities of the network:

$$TC_t = FCC_t + TCC_t + FOC_t + TOC_t \qquad \forall t \tag{13}$$

The calculation of each of these terms is described in detail in the next sections.

Facility capital cost

The facility capital cost in period t (FCC_t) is determined from the capacity expansions made in the manufacturing plants and storage facilities during that period:

$$FCC_t = \sum_g \sum_p \left(\alpha^{PL}_{gpt} N^{PL}_{gpt} + \beta^{PL}_{gpt} CE^{PL}_{gpt}\right) + \sum_g \sum_s \left(\alpha^{ST}_{gst} N^{ST}_{gst} + \beta^{ST}_{gst} CE^{ST}_{gst}\right) \forall t \tag{14}$$

The parameters α^{PL}_{gpt}, β^{PL}_{gpt} and α^{ST}_{gst}, β^{ST}_{gst} are the fixed and variable investment terms corresponding to plants and storage facilities, respectively. These parameters reflect the concept of economies of scale.

Transportation capital cost

The transportation capital cost, which includes the cost of the trucks and railcars required to satisfy the demand, is calculated via constraint 15:

$$TCC_t = \sum_l N^{TR}_{lt} \cdot cc_{lt} \qquad \forall t \tag{15}$$

Here, cc_{lt} represents the capital cost associated with transport mode l in period t, whereas N^{TR}_{lt} is an integer variable that denotes the total number of transportation units of type l purchased in period t that transport product i (i.e., $l \in LI(i)$). The average number of trucks and/or railcars required to satisfy a certain flow between different grids is computed from (1) the flow rate of products between the grids ($Q_{igg'lt}$); (2) the transportation mode availability (av_l); (3) the capacity of a transport container ($tcap_l$); (4) the average distance traveled between the grids ($distance_{gg'}$); (5) the average speed ($speed_l$); and (6) the loading/unloading time ($lutime_l$), as stated in Eq. 16:

$$\sum_{t' \leq t} N^{TR}_{lt'} \geq \sum_{i \in IL(l)} \sum_g \sum_{g' \neq g} \sum_t \frac{Q_{igg'lt}}{av_l \, tcap_l} \left(\frac{2 \, distance_{gg'}}{speed_l} + lutime_l \right) \qquad \forall l \tag{16}$$

Note that the total number of transportation units available in any period t includes the ones purchased in the same period t as well as those acquired in previous periods. Therefore, the left hand side of the inequality in Eq. 16 represents the summation of all the transportation units purchased in all the time periods t' up to the actual period t (i.e., $t' \leq t$). In this equation, $IL(l)$ denotes the set of product forms i that can be transported by transport mode l. For the sake of simplicity, this work assumes that each transportation facility can only operate between two predefined grids. Thus, in constraint 16, the distance between grids g and g' ($distance_{lgg'}$) is multiplied by two to account for the return journey of the trucks/railcars.

Facility operating cost

This term is obtained by multiplying the unit production and storage costs (upc_{ipt} and usc_{ist}, respectively) by the corresponding production rates and average inventory levels:

$$FOC_t = \sum_i \sum_g \sum_p upc_{igpt} PR_{igpt} + \sum_i \sum_g \sum_{s \in SI(i)} usc_{igst} \left(\theta D_{igt} \right) \qquad \forall t \tag{17}$$

Transportation operating cost

The total operating cost associated with the transportation tasks carried out in period t (TOC_t) includes the fuel (FC_t), labor (LC_t), maintenance (MC_t) and general costs (GC_t):

$$TOC_t = FC_t + LC_t + MC_t + GC_t, \qquad \forall t \tag{18}$$

The fuel cost is a function of the fuel price ($fuelp_{lt}$) and fuel usage:

$$FC_t = \sum_i \sum_g \sum_{g' \neq g} \sum_{l \in LI(i)} fuelp_{lt} \frac{2 distance_{gg'} \, Q_{igg'lt}}{fuelc_l \, tcap_l} \qquad \forall t \tag{19}$$

In Eq. 19, the fractional term represents the fuel usage, and is determined from the total distance traveled in a trip ($2\,distance_{lgg'}$), the fuel consumption of transport mode l ($fuelc_l$) and the number of trips made per period of time ($\frac{Q_{igg'lt}}{tcap_l}$). Furthermore, as shown in Eq. 20, the labor transportation cost is a function of the driver wage ($wage_l$) and total delivery time (term inside the brackets):

$$LC_t = \sum_i \sum_g \sum_{g' \neq g} \sum_{l \in LI(i)} wage_{lt} \left[\frac{Q_{igg'lt}}{tcap_{il}} \left(\frac{2\,distance_{lgg'}}{speed_l} + lutime_l \right) \right] \qquad \forall t \qquad (20)$$

The maintenance cost accounts for the general maintenance of the transportation systems and is a function of the cost per unit of distance traveled (cud_l) and total distance driven:

$$MC_t = \sum_i \sum_g \sum_{g' \neq g} \sum_{l \in LI(i)} cud_l \left(\frac{2\,distance_{gg'l}\, Q_{igg'lt}}{tcap_l} \right) \qquad \forall t \qquad (21)$$

Finally, the general cost includes the transportation insurance, license and registration, and outstanding finances. It can be determined from the unit general expenses (ge_{lt}) and number of transportation units, as follows:

$$GC_t = \sum_l \sum_{t' \leq t} ge_{lt} N^{TR}_{lt'} \qquad \forall t \qquad (22)$$

Environmental impact assessment: application of LCA principles

An imperative reason for pressing on the hydrogen alternative is the risk of climate change. Thus, in this work, the environmental performance of the network is measured by its contribution to climate change, which nowadays represents one of the major environmental concerns. Specifically, such a contribution is assessed by following the principles of Life Cycle Assessment (LCA), in a similar way as was done before by the authors.[21–23] The two major advantages of this strategy are that: (1) it allows to cover the entire life cycle of the product, process or activity being assessed, and (2) it includes a damage model that links the emissions released and waste generated with the corresponding environmental damage (i.e., contribution to climate change).

More precisely, this work makes use of the Eco-indicator 99 framework, which includes the most recent advances made in LCA. The Eco-indicator 99 allows for the computation of eleven impact categories, which are further aggregated into a single metric (i.e., Eco-indicator 99) that supports objective environmental assessments.

The goal of this work is to explore the environmental benefits, in terms of overall contribution to climate change, of adopting a hydrogen economy. Therefore, instead of calculating the Eco-indicator 99 itself, we focus our attention on only one of its impact categories: damage to human health caused by climate change. The computation of this metric follows the first three LCA phases: goal and scope definition, inventory analysis and impact assessment. The remaining phase - LCA interpretation - is addressed by including the impact assessment results into a bi-criterion optimization framework. These phases are described in detail in the next sections.

Goal and scope definition In this phase, the system boundaries and the impact categories are identified. In our specific case, the environmental analysis is restricted to the domain of the hydrogen network. Thus, we perform a "cradle-to-gate" analysis that embraces all the logistic activities of the network, starting from the extraction of raw materials and ending with the delivery of hydrogen to customers. With regard to the impact categories, we only evaluate the damage to human health caused by climate change.

Inventory analysis The second phase of LCA provides the inputs and outputs of materials and energy associated with the process (Life Cycle Inventory), which are required to calculate the environmental impact.

In the context of hydrogen networks, the environmental burdens are caused by the extraction of raw materials, the manufacturing and storage tasks, and the transportation of materials between grids. Mathematically, the inventory of emissions due to the operation of the network can be expressed as a function of some continuous decision variables of the model. Specifically, for each chemical b, they can be calculated from the production rates at the plants (PR_{igpt}), and the transport flows ($Q_{igg'lt}$), as stated in Eq. 23.

$$LCI_b = \sum_i \sum_g \sum_p \sum_t PR_{igpt}(\omega_{bp}^{PR} + \omega_{bi}^{ST}) + \sum_i \sum_g \sum_{g' \neq g} \sum_{l \in LI(i)} \sum_t Q_{igg'lt}\omega_b^{TR} \qquad \forall b \tag{23}$$

The first term of Eq. 23 represents the emissions associated with the manufacturing and storage tasks. The manufacturing tasks include the extraction of raw materials, the generation of the necessary utilities (i.e., steam, electricity, etc.) and the direct emissions from the main processes. The emissions associated with the storage tasks are due to the generation of the energy consumed in the compression of hydrogen. Finally, the second term of Eq. 23 considers the emissions of the transportation tasks.

In Eq. 23, ω_{bp}^{PR}, ω_{bi}^{ST}, and ω_b^{TR} denote the life cycle inventory entries (i.e., emissions released) associated with chemical b per reference flow of activity. In the production and storage of hydrogen, the reference flow is one unit of main product produced/stored. In the transportation tasks, the reference flow is one unit of mass transported one unit of distance.

Impact assessment In this stage the process data are translated into environmental information. As was mentioned before, we only consider the damage to human health caused by climate change. This metric is specified in Disability Adjusted Life Years (DALYs). A damage of one means that one life year of one individual is lost, or one person suffers four years from a disability with a weigh of 0.25.

Mathematically, the damage caused is calculated from the life cycle inventory and the corresponding damage factor (υ_b), as stated in Eq. 24.

$$DAM = \sum_{b} \upsilon_b LCI_b \tag{24}$$

The damage factor represents the link between the results of the inventory phase and the damage in the corresponding impact category. For the human health damage category, the damage model includes: (1) a fate analysis, to link any emission, which is expressed in terms of mass, to a temporary change in concentration; (2) an exposure analysis, to link this temporary concentration to a dose; (3) an effect analysis, to link the dose to a number of health effects; (4) a damage analysis to translate the health effects into Disability Adjusted Life Years (DALYs).

Interpretation Finally, in the fourth phase the results are analyzed and a set of conclusions or recommendations for the system are formulated. In this regard, the final goal of LCA is to provide criteria and quantitative measures for comparing different process operation and design alternatives. One of the main shortcomings of LCA is that it lacks a systematic way of generating such alternatives and identify the best ones in terms of environmental performance. To circumvent these limitations, in this paper we follow a combined approach that consists of coupling LCA and optimization tools within a single decision-making framework.[22,24,25] Thus, in our work the preferences are articulated in the post-optimal analysis of the Pareto optimal solutions. This approach provides further insights into the design problem and allows for a better understanding of the inherent trade-off between economic and environmental criteria.

Other constraints: bounds on integer variables

The total number of production plants, storage facilities and transportation units can be constrained to be lower than certain upper limits (UB^{PL}_{gp}, UB^{ST}_{gst} and UB^{TR}_{l}, respectively). This can be easily done by adding constraints 25 to 27:

$$N^{PL}_{gpt} \leq UB^{PL}_{gpt} \qquad \forall g, p, t \tag{25}$$

$$N^{ST}_{gst} \leq UB^{ST}_{gst} \qquad \forall g, s, t \tag{26}$$

$$N^{TR}_{lt} \leq UB^{TR}_{lt} \qquad \forall l, t \tag{27}$$

Solution strategy

The overall bi-criterion MILP can be expressed as follows:

$$\begin{aligned} \min_{x,X,N} \quad & (TDC(x,X,N), DAM(x,X,N)) \\ \text{s.t.} \quad & \text{constraints 1 to 27} \\ & x \in \mathbb{R}, X \in \{0,1\}, \; N \in \mathbb{N} \end{aligned}$$

Here, x denotes the continuous variables of the problem (capacity expansions, production rates, inventory levels and materials flows), X represents the binary variables (i.e., establishment of transportation links), and N are the integer variables denoting the number of plants, storage facilities and transportation units of each type selected.

For the calculation of the Pareto set of a multi-objective problem, two main methods exist in the literature. These are the weighted-sum method and the ϵ-constraint method.[26] The weighted-sum method, is only rigorous for the case of convex problems, whereas the ϵ-constraint method is also rigorous for the nonconvex case. The identification of the noninferior solutions can also be formulated as a parametric programming problem,[27] and solved with tailor made solution algorithms[22] for parametric programming. This last approach does not perform well when the combinatorial complexity of the model is very high, which turns out to be our case.

Thus, in this work, the Pareto solutions of the problem are computed via the ϵ-constraint method, which entails solving a set of instances of problem (M) corresponding to different values of the auxiliary parameter ϵ:

$$\begin{aligned} \text{(M)} \quad \min_{x,X,N} \quad & (TDC(x,X,N)) \\ \text{s.t.} \quad & \text{constraints 1 to 27} \\ & DAM(x,X,N) \leq \epsilon \\ & \underline{\epsilon} \leq \epsilon \leq \overline{\epsilon} \\ & x \in \mathbb{R}, X \in \{0,1\}, \; N \in \mathbb{N} \end{aligned}$$

where the lower and upper limits within which the epsilon parameter must fall (i.e., $\epsilon \in [\underline{\epsilon}, \overline{\epsilon}]$) are obtained from the optimization of each separate scalar objective:

$$\begin{aligned} \text{(M1a)} \quad (\bar{x}, \bar{X}, \bar{N}) = \arg \min_{x,X,N} \quad & (DAM(x,X,N)) \\ \text{s.t.} \quad & \text{constraints 1 to 27} \\ & x \in \mathbb{R}, X \in \{0,1\}, \; N \in \mathbb{N} \end{aligned}$$

which defines $\underline{\epsilon} = DAM(\bar{x}, \bar{X}, \bar{N})$ and

$$\begin{aligned} \text{(M1b)} \quad (\bar{x}, \bar{X}, \bar{N}) = \arg \min_{x,X,N} \quad & (TDC(x,X,N)) \\ \text{s.t.} \quad & \text{constraints 1 to 27} \\ & x \in \mathbb{R}, X \in \{0,1\}, \; N \in \mathbb{N} \end{aligned}$$

which defines $\bar{\epsilon} = DAM(\bar{x}, \bar{X}, \bar{N})$.

To expedite the calculation of the Pareto solutions, we next present a bi-level algorithm that exploits the specific structure of the model. Our solution procedure relies on hierarchically decomposing the monolithic formulation (M) into two levels, an upper level master problem (UP) and a lower level slave problem (LO), between which the algorithm iterates until a termination criterion is satisfied.[28]

Bi-level algorithm

The proposed decomposition algorithm (see Figure 2) solves an upper level master problem (UP), which is a specific relaxation of problem (M), to obtain a lower bound for the cost. In the master problem, the integer variables that represent the production plants and storage facilities are removed, whereas the ones associated with the transportation units are relaxed. Furthermore, a set of auxiliary binary variables are added to represent the selection of a specific technology over the entire planning horizon. Problem (UP) is therefore combinatorially less complex than the original model (M), and provides as output the subset of manufacturing, storage and transport technologies to be selected.

The lower level planning problem (LO) is solved for the selected set of technologies, yielding an upper bound to the cost for any feasible solution of (UP). Since only a subset of processes is selected in (UP), model (LO) contains fewer integer variables and is not as combinatorially complex as problem (M).

We note that for both problems (UP) and (LO), the equations for all time periods are included for all the processes that are considered. The computational expense is lowered by reducing the number of integer variables in each level. The problems are solved iteratively by adding integer and logic cuts until the bounds converge.

Upper level problem

The upper level master problem (UP) is obtained from model (M) as follows. The integer variables N_{gpt}^{PL}, N_{gst}^{ST} are removed, whereas variable N_{lt}^{TR} is relaxed into the continuous variable RN_{lt}^{TR}. Equations 5 and 9, which impose capacity limitations based on the number of installed facilities, are also removed. Constraint 14 is reformulated as follows:

$$\begin{aligned} FCC_t = & \sum_g \sum_p \left(\alpha_{gpt}^{PL} \frac{CE_{gpt}^{PL}}{PC_p^{PL}} + \beta_{gpt}^{PL} CE_{gpt}^{PL} \right) + \\ & \sum_g \sum_s \left(\alpha_{gst}^{ST} \frac{CE_{gst}^{ST}}{SC_s^{ST}} + \beta_{gst}^{ST} CE_{gst}^{ST} \right) \qquad \forall t \end{aligned} \tag{28}$$

Finally, three new sets of binary variables are added to the model: Z_{gp}^{PL}, Z_{gs}^{ST} and Z_{l}^{TR}. These binary variables take a value of one if the corresponding technology, either manufacturing process p, storage facility s or transportation mode l, are selected in grid g, and zero otherwise. The definition of these variables is enforced via the following constraints:

$$\frac{CE_{gpt}^{PL}}{\overline{PC}_{p}^{PL}} \leq Z_{gp}^{PL} UB_{gpt}^{PL} \qquad \forall g, p, t \tag{29}$$

$$\frac{CE_{gst}^{ST}}{\overline{SC}_{s}^{ST}} \leq Z_{gs}^{ST} UB_{gst}^{ST} \qquad \forall g, s, t \tag{30}$$

$$RN_{lt}^{TR} \leq Z_{l}^{TR} UB_{lt}^{TR} \qquad \forall l, t \tag{31}$$

As can be observed, if a technology is not selected (i.e., Z equals zero), the associated production rates, inventory levels and transportation units are forced to zero. On the other hand, when the binary variables take a value of one, the technologies are allowed to operate within specific upper bounds (UB_{gpt}^{PL}, UB_{gst}^{ST} and UB_{lt}^{TR}, respectively). Note that these bounds, which were also used in Eqs. 25 to 27, represent the maximum number of production plants, storage facilities and transportation units that can be selected.

The upper level problem can therefore be expressed as follows:

$$\begin{aligned}
\text{(UP)} \quad \min_{x,X,RN,Z} \quad & TDC(x, X, RN, Z) \\
\text{s.t.} \quad & \text{constraints 1 to 4, 6 to 8, 10 to 31} \\
& DAM(x, X, N) \leq \epsilon \\
& \underline{\epsilon} \leq \epsilon \leq \overline{\epsilon} \\
& x, RN \in \mathbb{R}\,,\ X, Z \in \{0, 1\}
\end{aligned}$$

where Z denotes the new set of auxiliary binary variables. Note that problem (UP) has $|g|^2 \cdot |l| \cdot |t| + |g| \cdot |p| + |g| \cdot |s| + |l|$ binary variables, whereas problem (M) has $|g|^2 \cdot |l| \cdot |t|$ binary variables and $|t| \cdot (|g| \cdot |p| + |g| \cdot |s| + |l|)$ integer variables. Thus, (UP) is combinatorially less complex than (M).

Model (UP) is a relaxation of problem (M) that has the following property (see the proof in the Appendix):

Property 1. *Problem (UP) provides a lower bound to the solution of problem (M).*

Lower level problem

The lower level is represented by the original MILP model (M), which is solved for only a subset of technologies (i.e., manufacturing plants, storage and transport facilities) predicted at the upper level. The main motivation for this procedure is that the number of integer variables, and, hence, the size of the lower level, is reduced by excluding the technologies that were not selected by the upper level problem through the auxiliary binary variables Z obtained at iteration r. This is accomplished by adding the following inequalities:

$$N_{gpt}^{PL} \leq \overline{Z_{gp}^{PL}} UB_{gpt}^{PL} \qquad \forall g, p, t \tag{32}$$

$$N_{gst}^{ST} \leq \overline{Z_{gs}^{ST}} UB_{gst}^{ST} \qquad \forall g, s, t \tag{33}$$

$$N_{lt}^{TR} \leq \overline{Z_{l}^{TR}} UB_{lt}^{TR} \qquad \forall l, t \tag{34}$$

where $\overline{Z_{gp}^{PL}}$, $\overline{Z_{gs}^{ST}}$ and $\overline{Z_{il}^{TR}}$ denote the optimal values of the auxiliary binary variables of the master problem. Note that constraints 32 to 34 force the integer variables of (M) to take a zero value when the corresponding auxiliary binary variables are zero, and are inactive otherwise.

The overall lower level problem can therefore be formally stated as follows:

$$\begin{aligned} \text{(LO)} \quad \min_{x,X,N} \quad & TDC(x, X, N) \\ \text{s.t.} \quad & \text{constraints 1 to 27, 32 to 34} \\ & DAM(x, X, N) \leq \epsilon \\ & \underline{\epsilon} \leq \epsilon \leq \overline{\epsilon} \\ & x \in \mathbb{R}\,,\ X \in \{0, 1\}\,,\ N \in \mathbb{N} \end{aligned}$$

Hence, the lower level problem (LO) is simply defined by adding to problem (M) the inequalities 32 to 34. Note that model (LO) yields a valid upper bound to (M), because its search space is contained in the domain of (M). Thus, any feasible solution of (LO) is also a feasible solution of (M). In our algorithm, the upper-level (UP) and the lower-level (LO) problems are solved iteratively until the bounds of each level converge within a specified tolerance.

Integer and logic cuts

The upper level problem must be resolved at each iteration in order to provide new solutions for the lower level. This procedure is repeated until the termination criterion is satisfied. To expedite the calculation of the master problem, we make use of integer and logic cuts. The integer cuts are employed to exclude those solutions explored so far. These cuts are mathematically expressed as follows:[29]

$$\sum_{m \in W_1^r} Z_m - \sum_{m \in W_0^r} Z_m \leq |W_1^r| - 1 \qquad \forall r \tag{35}$$

where $W_1^r = \{m | \overline{Z_m^r} = 1\}$ and $W_0^r = \{m | \overline{Z_m^r} = 0\}$, with $\overline{Z_m^r}$ being the value of the m component of the vector of binary variables in the optimal solution computed in iteration r. Note that W_1^r and W_0^r are both obtained from the optimal solution of the upper level problem in iteration r.

Furthermore, to reduce the search space and the number of iterations in the decomposition procedure, we employ logic cuts that allow to eliminate suboptimal alternatives. More precisely, we make use of subset cuts, which are motivated by the cuts proposed by Iyer and Grossmann.[28] The following property establishes the basis for the derivation of such cuts (the proof can be found in the Appendix):

Property 2. *Let $W_1^r = \{m|\overline{Z_m^r} = 1\}$ and $W_0^r = \{m|\overline{Z_m^r} = 0\}$ correspond to the optimal solution of (UP) in iteration r. For all iterations $s > r$, if W_1^r is feasible in (LO), then any solution $W_1^s \subset W_1^r$ will result in a solution of (LO) such that $TDC_{LO}^s \geq TDC_{LO}^r$.*

The cut for precluding subsets can be logically written as:

$$\left(\bigvee_{m \in W_0^r} Z_m \right) \vee Z_n \qquad \forall r \qquad \forall n \in W_1^r \tag{36}$$

This logic expression can be mathematically translated into the following constraint:

$$\sum_{m \in W_0^r} Z_m + Z_n \geq 1 \qquad \forall r \qquad \forall n \in W_1^r \tag{37}$$

A description on how to derive cuts from logical inference clauses is given by Raman and Grossmann.[30]

Algorithmic Steps

The detailed steps of the proposed decomposition strategy, which are applied for every selected value of ϵ, are as follows:

1. Set iteration count $r = 0$, upper bound $UB = \infty$, lower bound $LB = -\infty$, and tolerance error $= tol$.
2. Set $r = r + 1$. Solve the MILP master problem (UP):
   - If problem (UP) is infeasible, then stop.
   - Otherwise, set the current lower bound to: $LB = LB^r$, and define:

$$W_1^r = \{m|\overline{Z_m^r} = 1\} \tag{38}$$

$$W_0^r = \{m|\overline{Z_m^r} = 0\} \tag{39}$$

   where LB^r is the objective function value associated with the optimal solution of (UP) in iteration r, and $\overline{Z^r}$ represents the vector of binary variables for the same solution.

3. For fixed $\overline{Z^r}$, solve the lower level problem (LO) to obtain a capacity expansion plan and an upper bound to the cost.

   - If problem (LO) is infeasible, then add the following integer cut to (UP) and go to step 2:

$$\sum_{m \in W_1^r} Z_m - \sum_{m \in W_0^r} Z_m \leq |W_1^r| - 1 \tag{40}$$

   - Otherwise, update the current upper bound as follows: $UB = \min_r \{UB^r\}$, where UB^r represents the objective function value associated with the optimal solution of (LO) in iteration r.

4. Check the convergence criteria:

   - If $\frac{UB-LB}{UB} \leq tol$, then stop. The solution corresponding to UB (i.e., the solution of model (LO) in the iteration with minimum cost) satisfies the termination criterion (i.e., it can be regarded as optimal within the predefined optimality gap).
   - Otherwise, define the set of integer and logic cuts that will be added to problem (UP) in the next iteration:

$$\sum_{m \in W_1^r} Z_m - \sum_{m \in W_0^r} Z_m \leq |W_1^r| - 1 \tag{41}$$

$$\sum_{m \in W_0^r} Z_m + Z_n \geq 1 \qquad \forall n \in W_1^r \tag{42}$$

   and go to step 2.

Remarks

- The proposed decomposition algorithm provides the global optimal solution to the original mixed-integer linear programming (MILP) model in a finite number of iterations, since the number of possible production, storage and transport technologies is finite.

- The integer and logic cuts in Eqs. 41 and 42 are added cumulatively at each iteration to the upper-level model (UP), which leads to an increase in its size.

- The master problem can be tightened by adding the following constraints:

$$\tau \underline{PC_p^{PL}} Z_{gp}^{PL} \leq \sum_t PR_{igpt} \qquad \forall i, g, p \tag{43}$$

$$Z_{gs}^{ST} \leq \sum_t \frac{CE_{gst}^{ST}}{\overline{SC_s^{ST}}} \qquad \forall g, s \tag{44}$$

$$Z_l^{TR} \leq \sum_t RN_{lt} \qquad \forall l \tag{45}$$

These equations impose lower limits on the production rates, expansions in the capacity of the storage facilities and number of transportation units if the corresponding processes are selected, and are inactive otherwise. The use of such inequalities leads to tighter bounds, which in turn reduces the computational burden of the algorithm.

- To reduce the number of iterations when there is slow convergence between the lower and upper bounds, a nonzero optimality tolerance *tol* can be specified in step 1 (e.g., 1%- 5%). Note that, it might be difficult to close the gap between the bounds in large problems. However, even in those cases, the proposed procedure might be able to generate good feasible solutions whose global optimality can be guaranteed within the difference of the bounds.

Case study

The capabilities of our modeling framework and solution strategy are illustrated through a case study based on a real-world scenario. Due to space limitations, a brief overview of this example is next provided. A more detailed description can be found in the work of Almansoori and Shah.[19]

We address the optimal design of a hydrogen SC for vehicle use in UK. A superstructure of technologies is postulated in which all different alternatives for production, storage and transportation of hydrogen are embedded (see Figure 3). This superstructure also considers the possibility of establishing these technologies in a set of regions or grids distributed all over the country. Each of these regions has an associated hydrogen demand to be fulfilled either locally or by importing hydrogen from other grids.

The goal of the model is to determine the optimal set of production, storage and transportation technologies along with their location in order to minimize the associated environmental impact and total cost. The specific process alternatives considered in the study are given bellow:

- Production technology: steam methane reforming (SMR), coal gasification and biomass gasification.
- Storage technology: liquid hydrogen storage and compressed gas storage.
- Transportation technology: liquid hydrogen (LH_2) tanker truck, liquid hydrogen railway tank car, compressed-gaseous hydrogen (CH_2) tube trailer, compressed-gaseous hydrogen railway tube car.

All the data of the problem, which was collected from a variety of sources, can be found in the work of Almansoori and Shah.[19] A preprocessing step was performed to reduce the computational burden of the original model by merging adjacent regions with low demands into aggregated grids. The new grids along with the associated demand are shown in Figure 4 and Table 1. The upper bounds on the number of plants, storage facilities and transportation units were 20, 250 and 50, respectively. We also assume that no plants can be established in grids 1 and 22 (i.e., the associated upper bounds are zero). The fixed and variable investment terms associated with the production technologies and warehouses, which are given in Tables 2 and 3, were determined from the investment costs corresponding to the minimum and maximum capacity solutions. Specifically, the cost of the maximum capacity solution was taken from the work of Almansoori and Shah,[19] whereas that corresponding to the minimum capacity alternative was estimated from the former one via the *six-tenth* rule. The interest rate, the minimum demand satisfaction level and the minimum desired percentage, τ, of the available installed capacity were set to 10%, 90% and 25%, respectively. It was also assumed that raw materials are available at the same price in all the grids.

The data of the emission inventories associated with the operation of the hydrogen network (i.e., extraction of raw materials, production, storage and transportation of hydrogen) were estimated from different sources in the literature.[3,31–33] This information was complemented with that given by the Ecoinvent database,[34] from which the emissions associated with the energy generation and transportation tasks were retrieved. Table 4 summarizes the main results of the LCA analysis carried out to determine the environmental impact of the production, storage and transportation tasks. In our study, the parameters of the damage model that translates the emissions released into the damage to human health caused by climate change were taken from the Eco-indicator 99 methodology,[35] assuming the average weighting set and the Hierarchist perspective.

Computational performance

We first solved several problems of different levels of complexity based on the number of time periods, each of which has a length of one year. The goal was to illustrate the performance of the algorithm as compared to the full-space method. For the sake of simplicity, the ϵ parameter was set to ∞ in all the cases, which is equivalent to minimizing the cost as a single objective.

All the problems were implemented in GAMS[36] and solved in the full-space using the CPLEX 9.0 solver. Tables 5 and 6 shows the problem sizes and solution times for the proposed decomposition algorithm and the full-space method, both solved for a 1% optimality tolerance. Note that in the case of the decomposition strategy, this gap represents the difference between the solutions of the higher level and lower level problems, and not the optimality gap with which the sub-problems were solved. In all the problems solved, the optimality gap was reached in only one iteration.

As can be observed, the proposed approach shows better numerical performance than the full-space method. For small problems ($t \leq 2$), the full-space method is almost as efficient as the decomposition strategy, since the number of integer variables is very small. On the other hand, as the size of the problem increases the differences in CPU time are more significant. Specifically, for $t \geq 3$, the decomposition strategy provides near optimal solutions (i.e., solutions with an optimality gap of 1%) in CPU times that are approximately one order of magnitude lower than those reported by the full-space approach. As can be observed, the higher level problem can be solved very quickly, whereas the lower level problem is the bottleneck of the proposed method. Note that in the latter formulation the number of integer variables is significantly decreased in comparison with the full space model. This greatly reduces the combinatorial complexity of the problem, and, thus, its computational burden.

Pareto solutions and discussion

Having proved the application and computational effectiveness of the proposed algorithm, our approach was next used to generate the complete Pareto set associated with one of the previous examples. Specifically, we solved the problem with a 5-year time horizon (i.e, the case when t=*5*). The upper bound on the number of transportation units was set to 300, whereas the remaining data were the same as in the previous case.

The values of $\underline{\epsilon}$ and $\overline{\epsilon}$ that define the interval within which the value of DAM must fall were firstly calculated by maximizing both objectives separately. The interval $[\underline{\epsilon}, \overline{\epsilon}]$ was next partitioned into 50 subintervals of equal length, and model (M) was then calculated for every possible value of ϵ. The proposed algorithm was solved with an optimality gap (i.e., tolerance) of 1 %. The problem size is given in Table 5. The total CPU time required to generate the Pareto solutions was 6,857 CPU seconds.

Figure 5 shows the Pareto solutions obtained by following the proposed procedure. Each point of the Pareto set entails a specific SC structure and a set of planning decisions. Note that a natural trade-off exists between total cost and environmental impact, since a reduction in the latter metric can only be achieved by compromising the cost of the network. Furthermore, five different structural alternatives (A,B,C,D and E) were identified in the Pareto set. In the first design A (i.e., minimum cost solution), hydrogen is produced via steam reforming and it is transported and stored as a liquid. In the alternative B, hydrogen is generated from steam as well as biomass gasification, and it is also handled as a liquid. Finally, in designs C,D and E, hydrogen is produced exclusively from biomass. These last alternatives differ in the technology employed to transport and store hydrogen. In solution C, compressed hydrogen gas is selected, whereas in option D, part of the hydrogen is transported and stored as a liquid and another part as a gas. In the solution that causes the least environmental impact (E), only compressed hydrogen gas is used. As can be seen, the Pareto curve is rather smooth in the region that goes from A to C, whereas from C to E the slope increases drastically and the shape of the curve becomes sharp.

In light of these results, one can conclude that the best manner to achieve significant environmental savings without compromising too much the total cost of the network is to replace steam reforming by biomass gasification (structural solution B). On the other hand, substituting compressed gas hydrogen by liquid hydrogen is not a good choice, since this alternative significantly increases the total cost of the hydrogen SC without reducing to a large extent the associated environmental impact. For instance, the environmental impact of solution C is $6 \cdot 10^5$ DALYs lower than that of A, whereas its cost doubles approximately that of A . On the other hand, the shift from C to E leads to a reduction of only $1 \cdot 10^4$ DALYs in the environmental impact, but increases the cost by a factor of 6, when compared to solution C, and by a factor of 10, when compared to A.

Figures 6 and 7 depict the SC configurations of the extreme solutions (the minimum cost and environmental impact alternatives). The figure also provides the number and type of production plants and storage facilities established in each grid along with the associated transportation links between them. It can be observed that the regions of UK with high population density, mainly Manchester and London (grids 10 and 17, respectively), have higher number of production plants and storage facilities than those with low hydrogen demand. The figures also show that there is at least one storage facility installed in every grid, while the same is not true for the manufacturing plants. Particularly, no manufacturing plant is built in grids 1 and 22, in which the demand is satisfied by importing hydrogen from other neighboring grids. On the other hand, it is necessary to install storage facilities in these grids, since they are still required to deliver hydrogen to the final customers.

It can also be found that the minimum environmental impact solution has more production plants than the minimum cost one (i.e., 51 vs 36). Furthermore, in the former solution 97.66% of the overall demand is fulfilled by local production, whereas in the latter this percentage drops to 92.20%. These results indicate that the former alternative represents a slightly more decentralized network (i.e., more plants are established). This is because the establishment of more production facilities reduces the flow of materials between grids and consequently the emissions associated with the transportation tasks. On the other hand, this policy also increases the capital cost of the SC, and for this reason is not adopted to the same extent in the most profitable solution.

Note also that in the minimum cost solution, hydrogen is stored and transported as a liquid, whereas in the minimum environmental impact alternative, compressed hydrogen is employed. This result may seem surprising, since the capacity of the transport container is about 20 times higher for liquid hydrogen than for compressed gas. This may eventually lead to an increase in the number of trips required between production plants and storage facilities and therefore to higher emissions of green house gases. Note, however, that the energy associated with the liquefaction of hydrogen is much higher than that associated with the generation of compressed hydrogen gas. As commented before, the transportation tasks are minimized in the more sustainable solution by building the plants as close as possible to the markets. Hence, in practice, the latter effect (i.e., energy required for compressing hydrogen) compensates the former one (i.e., emissions of the transportation tasks) from the environmental point of view. Unfortunately, from an economic perspective, this policy is not appealing at all, since replacing liquid hydrogen by compressed gas hydrogen leads to very large capital costs.

The extreme solutions also differ in the manufacturing technology employed to produce hydrogen (i.e., steam reforming for the minimum cost alternative and biomass gasification in the more sustainable one). These results are due to the low cost of the steam methane reforming and low environmental impact of the gasification of biomass.

Figure 8 shows the contribution of the different sources of impact to the total environmental damage for the extreme Pareto solutions. In the minimum cost solution, the main source of impact is the production of hydrogen. In the minimum environmental impact solution, the main positive contribution to the total impact is the compression of hydrogen. Note that in the latter case the production of hydrogen has a negative sign. This is because producing hydrogen from biomass has the environmental advantage of reducing the CO_2 emissions over the entire life cycle of the process.[3,32,33] Particularly, in this solution it occurs that the carbon sequestration capacity of the biomass feedstock (i.e., wood) compensates the other emissions of the process. As can be seen, in both cases the impact due to the transportation and storage tasks is rather small in comparison with that associated with the generation of hydrogen. These results are in consonance with those shown in Figure 5, and explain why substituting the original storage and transportation technologies by others that cause less impact does not lead to a significant reduction of the overall environmental damage.

A breakdown of the total cost of each extreme solution is given in Figure 9. As can be observed, in both cases the main contributor to the total cost is the storage capital cost. Furthermore, in the minimum impact solution these costs are approximately one order of magnitude larger than in the minimum cost alternative, and so is the total discounted cost of the network. This is because the unit storage cost for liquid hydrogen is much lower than that associated with a pressure vessel (18 \$ kg^{-1} vs. 281 \$ kg^{-1}).[31] These data also explain the sharp change in the shape of the Pareto curve that occurs when switching from liquid hydrogen to compressed gas hydrogen. On the other hand, the transportation cost represents the smallest contribution to the total cost, mainly because both networks have a high level of decentralization (above 90%). Note that these results agree with those obtained by Almansoori and Shah.[19]

Conclusions

This work has addressed the optimal design and planning of sustainable hydrogen supply chains for vehicle use. The design task was formulated as a bi-criterion mixed integer linear programming (MILP) problem that seeks to minimize cost and environmental impact. The environmental impact was holistically measured over the entire life cycle of the process by applying the Eco-indicator 99 methodology, which follows the principles of LCA. A decomposition strategy that exploits the mathematical structure of the model was also introduced to expedite its solution.

The capabilities of the proposed modeling framework and solution strategy were shown through a case study based on a real scenario. On the computational side, our solution technique proved to be approximately one order of magnitude faster than the full-space method. Furthermore, the Pareto solutions calculated by our algorithm provided valuable insights into the design problem and suggested process alternatives that may lead to significant environmental improvements. Specifically, from the obtained results it can be concluded that important reductions in the contribution to global warming can be achieved by replacing steam reforming by biomass gasification. Furthermore, we also observed that decentralized hydrogen networks in which the transportation tasks are minimized lead to lower environmental impacts. These recommendations are intended to guide decision-makers towards the adoption of more sustainable alternatives.

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Notation

Indices

b	environmental burdens
i	hydrogen form
g	grid zones
l	transportation mode
p	manufacturing technologies
s	storage technologies
t	time period

Sets

$IL(l)$	set of hydrogen forms that can be transported via transportation mode l
$IS(s)$	set of hydrogen forms that can be stored via technology s
$LI(i)$	set of transportation modes that can transport hydrogen form i
$SI(i)$	set of storage technologies that can store hydrogen form i

Parameters

av_l	availability of transportation mode l
cc_{lt}	capital cost of transport mode l in period t
cud_{lt}	maintenance cost of transportation mode l in period t per unit of distance traveled
$\overline{D_{gt}}$	total demand of hydrogen in grid g in period t
$distance_{gg'}$	average distance traveled between grids g and g'
$dsat$	demand satisfaction level to be fulfilled
$fuelc_l$	fuel consumption of transportation mode l
$fuelp_{lt}$	price of the fuel consumed by transportation mode l in period t
ge_{lt}	general expenses of transportation mode l in period t
ir	interest rate
$lutime_l$	loading/unloading time of transportation mode l
$\overline{PC_p^{PL}}$	upper bound on the capacity expansion of manufacturing technology p
$\underline{PC_p^{PL}}$	lower bound on the capacity expansion of manufacturing technology p
$\overline{QC_{gg'l}}$	upper bound on the flow of materials between grids g and g' via transportation model l
$\underline{QC_{gg'l}}$	lower bound on the flow of materials between grids g and g' via transportation model l
$\overline{SC_s^{ST}}$	upper bound on the capacity expansion of storage technology s
$\underline{SC_s^{ST}}$	lower bound on the capacity expansion of storage technology s
$speed_l$	average speed of transportation mode l
$tcap_l$	capacity of transport mode l
upc_{igpt}	unit production cost of hydrogen form i produced via technology p in grid g in period t
usc_{igst}	unit storage cost of hydrogen form i stored via technology s in grid g in period t
$wage_{lt}$	driver wage of transportation mode l in period t

α_{gpt}^{PL}	fixed investment term associated with manufacturing technology p installed in grid g in period t
α_{gst}^{ST}	fixed investment term associated with storage technology s installed in grid g in period t
β_{gpt}^{PL}	variable investment term associated with manufacturing technology p installed in grid g in period t
β_{gst}^{ST}	variable investment term associated with storage technology s installed in grid g in period t
ω_{bp}^{PR}	emissions of chemical b associated with the production of one unit of hydrogen via technology p
ω_{bi}^{ST}	emissions of chemical b associated with the compression of one unit of hydrogen into physical form i
ω_{b}^{TR}	emissions of chemical b per unit of mass transported one unit of distance
θ	average storage period
τ	minimum desired percentage of the capacity that must be utilized

Variables

C_{gpt}^{PL}	capacity of manufacturing technology p in grid g in period t
C_{gst}^{ST}	capacity of storage technology s in grid g in period t
CE_{gpt}^{PL}	capacity expansion of manufacturing technology p in grid g in period t
CE_{gst}^{ST}	capacity expansion of storage technology s in grid g in period t
D_{igt}	amount of hydrogen form i distributed in grid g in period t
FC_t	fuel cost in period t
FCC_t	facility capital cost in period t
FOC_t	facility operating cost in period t
GC_t	general cost in period t
LC_t	labor cost in period t
MC_t	maintenance cost in period t
N_{gpt}^{PL}	number of plants of type p installed in grid g in period t (integer variable)
N_{gst}^{ST}	number of storage facilities of type s installed in grid g in period t (integer variable)
N_{lt}^{TR}	number of transportation units of type l purchased in period t (integer variable)
PR_{igpt}	production of hydrogen mode i via technology p in period t in grid g
$Q_{igg'lt}$	flow of hydrogen mode i via transportation mode l between grids g and g' in period t
S_{igst}	amount of hydrogen in physical form i stored via technology s in grid g in period t
TC_t	total amount of money spent in period t
TCC_t	total transportation capital cost in period t
TDC	total discounted cost
TMC_{lt}	transportation capital cost of mode l in period t
TOC_t	transportation operating cost in period t
$X_{gg'lt}$	binary variable (1 if a link between grids g and g' using transportation technology l is established, 0 otherwise)

Appendix A: Proofs

For convenience in the presentation of the proofs, the objective functions of problems (M) and (UP) are denoted by $f(\cdot)$ and $g(\cdot)$, respectively.

Proof of Property 1. Let $\overline{p}$ be the solution of model (UP) that defines its optimal objective $g(\overline{p})$. Assume that there exists a feasible solution $\overline{q} = (\overline{x}, \overline{X}, \overline{N})$ of (M) such that $f(\overline{q}) < g(\overline{p})$. Now, consider the point $\overline{t} = (\overline{x}, \overline{X}, \overline{RN}, \overline{Z})$, which has the same vectors of continuous and binary variables $\overline{x}$ and $\overline{X}$ as $\overline{q}$, and in which the values of $\overline{RN}$ and $\overline{Z}$ are defined as follows:

$$\overline{RN^{TR}_{lt}} = \overline{N^{TR}_{lt}} \qquad \forall l, t$$

$$\overline{Z^{PL}_{gp}} = \begin{cases} 0 & \text{if } \overline{N^{PL}_{gpt}} = 0 \, \forall t \\ 1 & \text{otherwise} \end{cases} \quad \forall g, p$$

$$\overline{Z^{ST}_{gs}} = \begin{cases} 0 & \text{if } \overline{N^{ST}_{gst}} = 0 \, \forall t \\ 1 & \text{otherwise} \end{cases} \quad \forall g, s$$

$$\overline{Z^{TR}_{l}} = \begin{cases} 0 & \text{if } \overline{N^{TR}_{lt}} = 0 \, \forall t \\ 1 & \text{otherwise} \end{cases} \quad \forall l$$

Clearly, $\overline{t}$ is feasible in (UP). Now, from constraints 5 and 9 in (M), we have that:

$$\frac{\overline{CE^{PL}_{gpt}}}{\overline{PC^{PL}_{p}}} \leq \overline{N^{PL}_{gpt}} \qquad \forall g, p, t$$

$$\frac{\overline{CE^{ST}_{gst}}}{\overline{SC^{ST}_{s}}} \leq \overline{N^{ST}_{gst}} \qquad \forall g, s, t$$

which implies that the facility capital cost associated with solution $\overline{t}$ is lower than or equal to that corresponding to $\overline{q}$. Since all the remaining terms of $f(\cdot)$ and $g(\cdot)$ are the same, we conclude that $g(\overline{t}) \leq f(\overline{q}) < g(\overline{p})$. This contradicts that $\overline{p}$ is the optimal solution of (UP).$\square$

Proof of Property 2. Consider the lower level problems associated with W_1^r and W_1^s, which are denoted by $(\text{LO})^r$ and $(\text{LO})^s$, respectively. From the fact that W_1^s is a subset of W_1^r, it follows that the right hand side of Eqs. 32, 33 and 34 in $(\text{LO})^s$ must be lower or equal than that in $(\text{LO})^r$. Since all the remaining equations are the same in both formulations, and Eqs. 32, 33 and 34 are tighter in $(\text{LO})^s$, we conclude that $(\text{LO})^r$ is a relaxation of $(\text{LO})^s$, and therefore yields a valid lower bound on its optimal solution, which implies that $TDC^r_{LO} \leq TDC^s_{LO}$. $\square$

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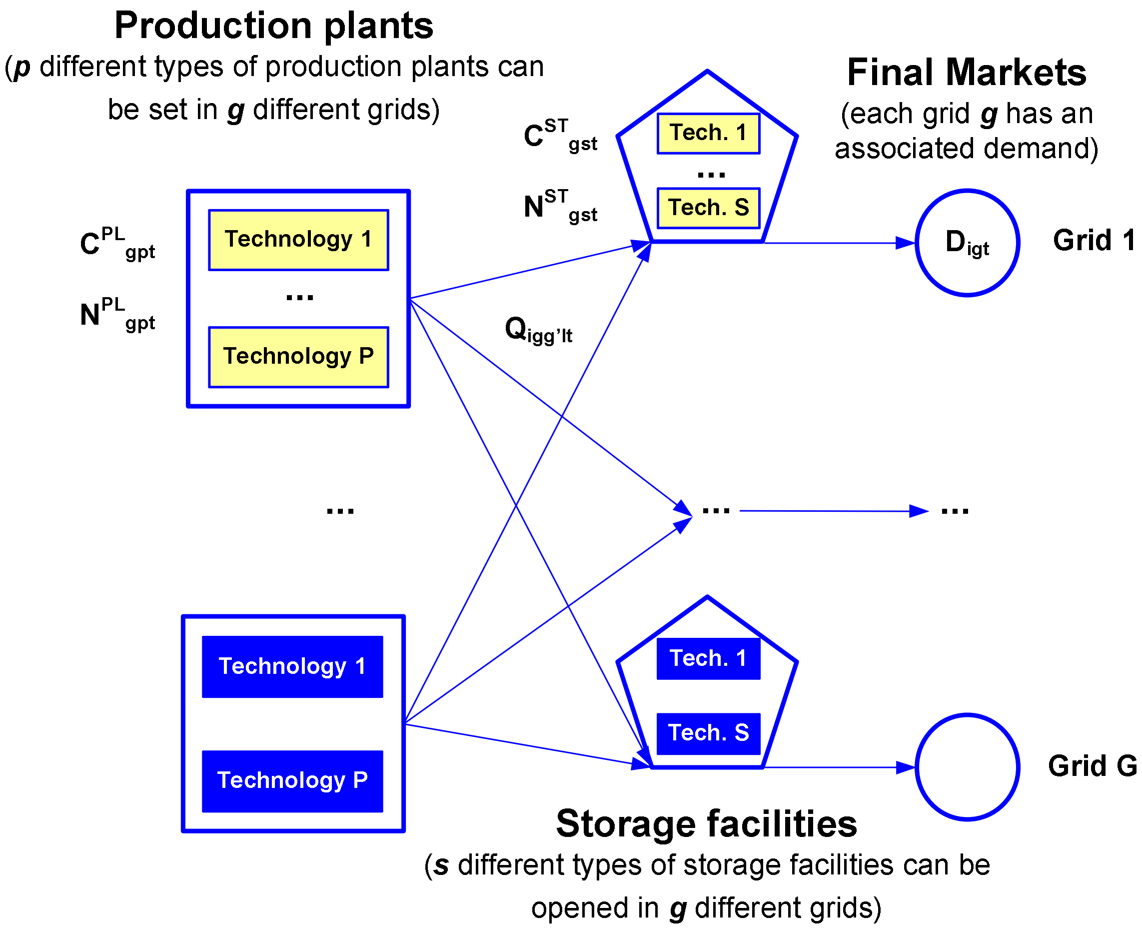

Figure 1: Three-echelon supply chain taken as reference.

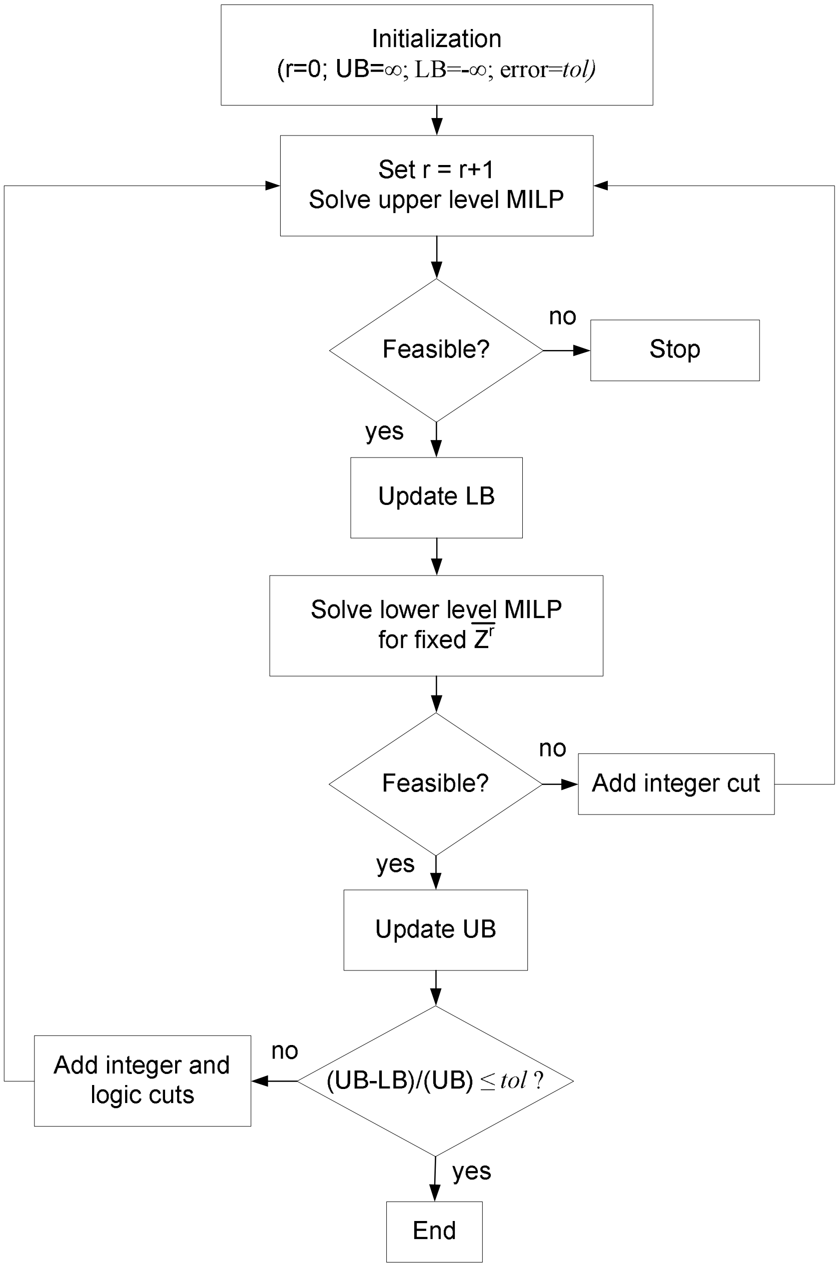

Figure 2: Flowchart for the bilevel decomposition algorithm.

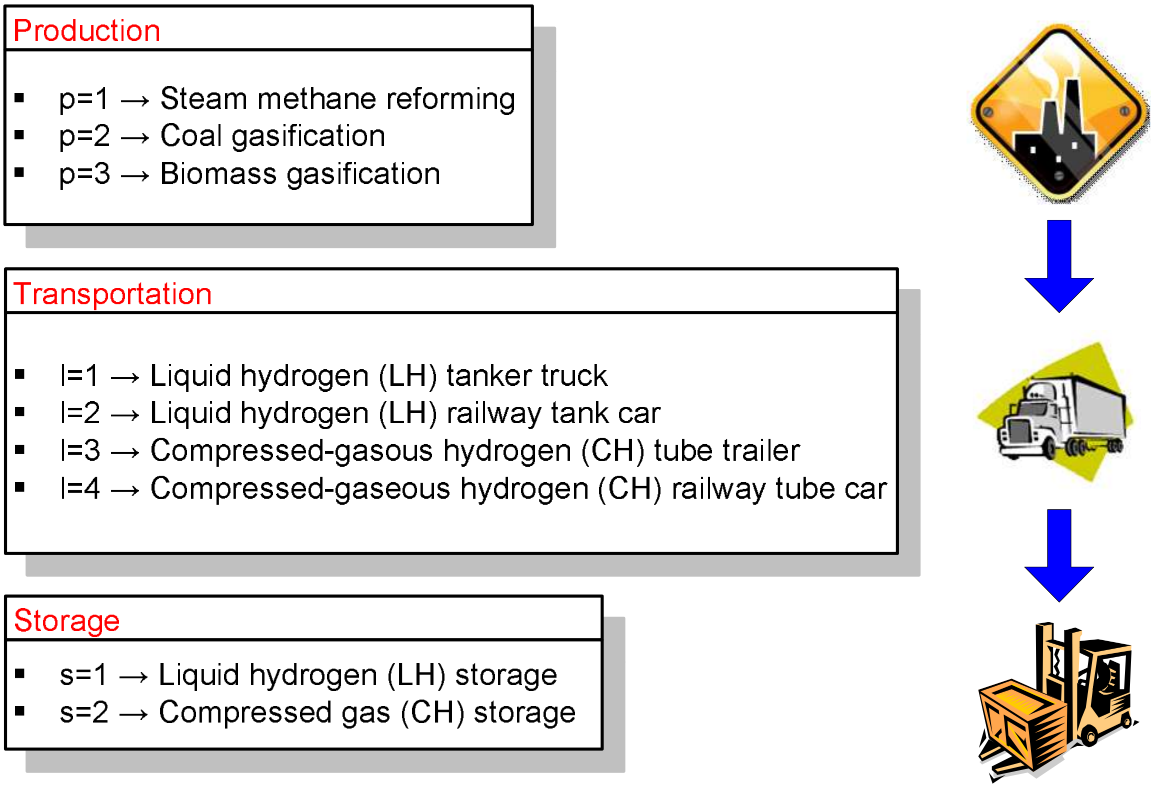

Figure 3: Set of process alternatives of the case study.

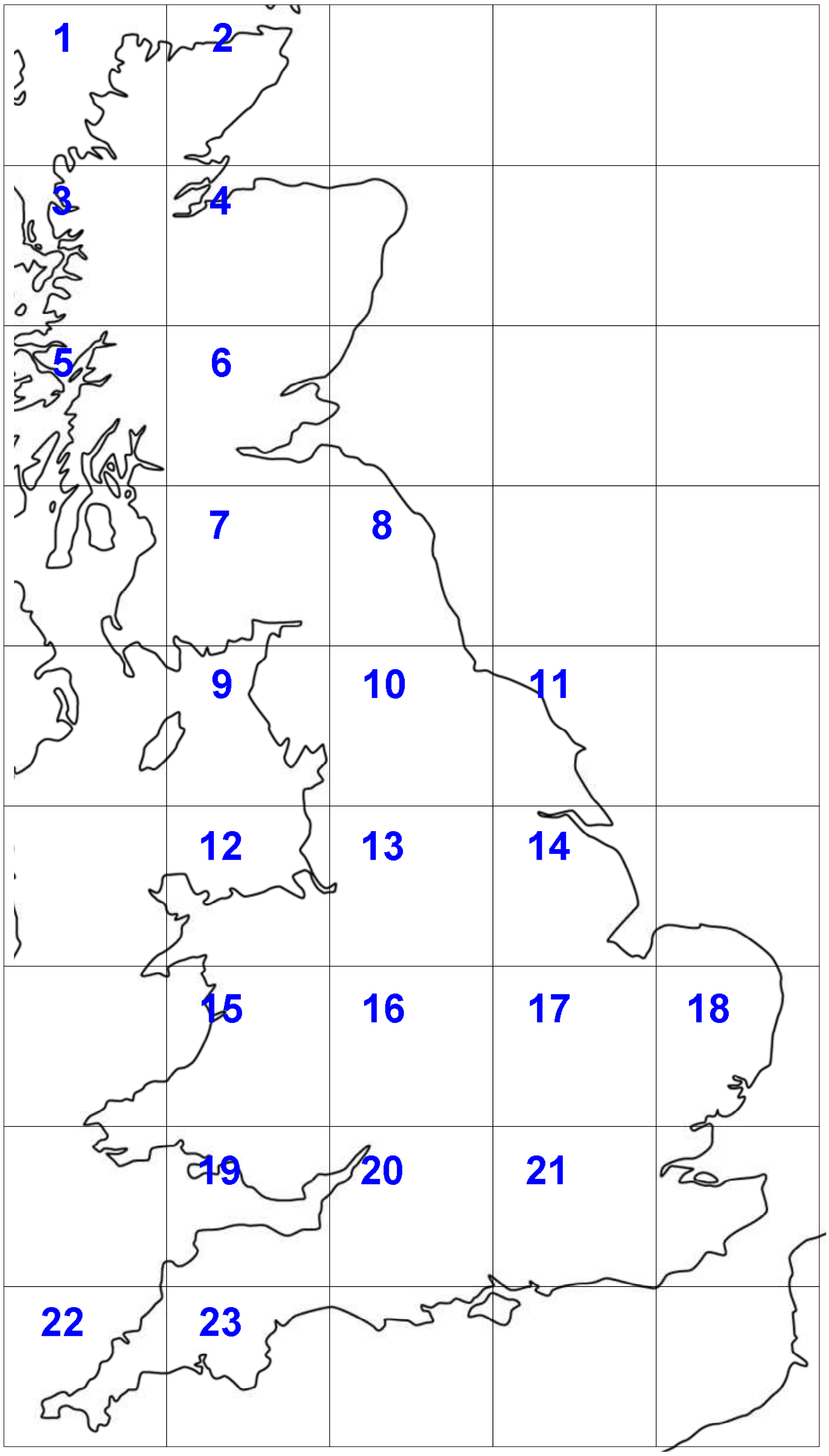

Figure 4: Set of grids (potential locations) for the SC entities in the case study.

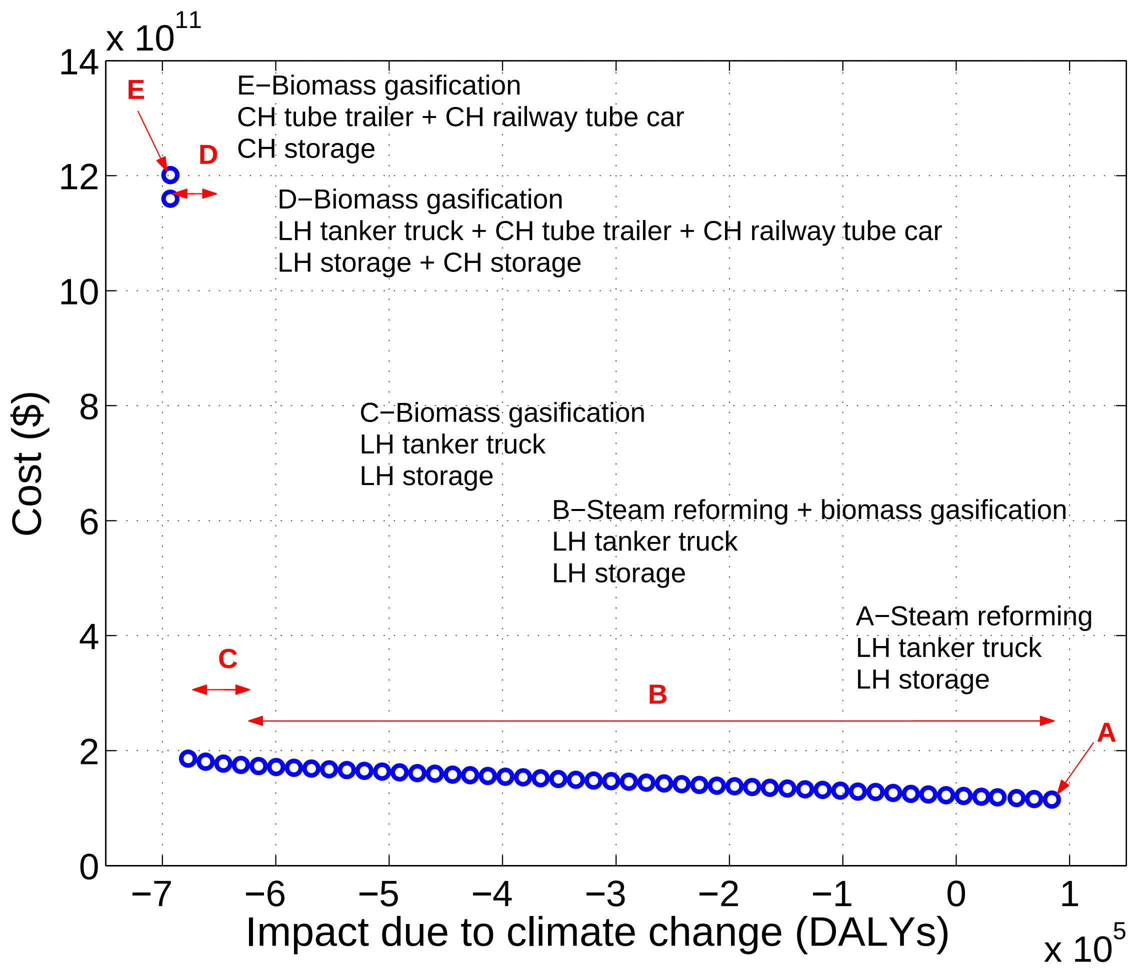

Figure 5: Pareto set.

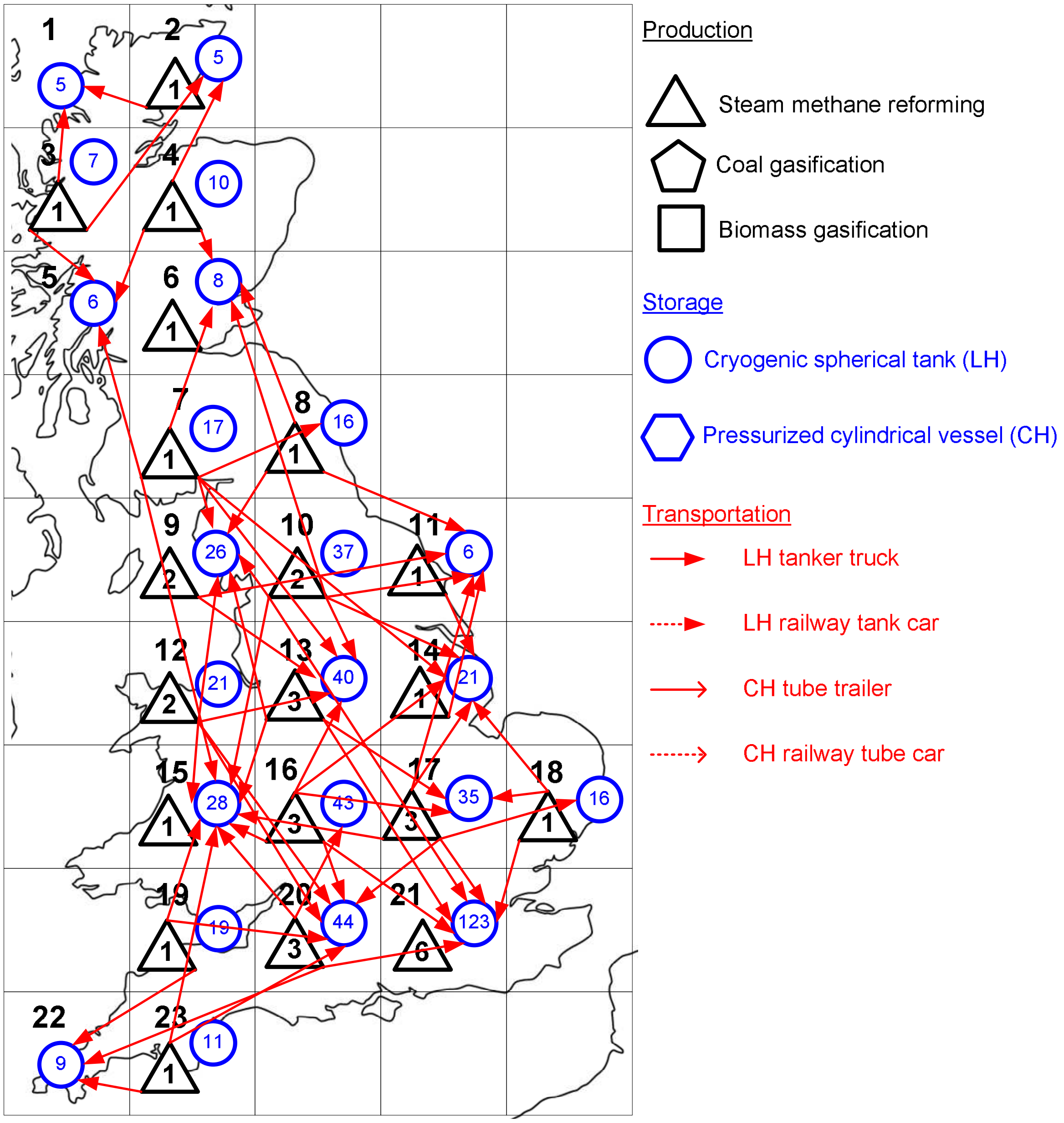

Figure 6: Minimum cost solution.

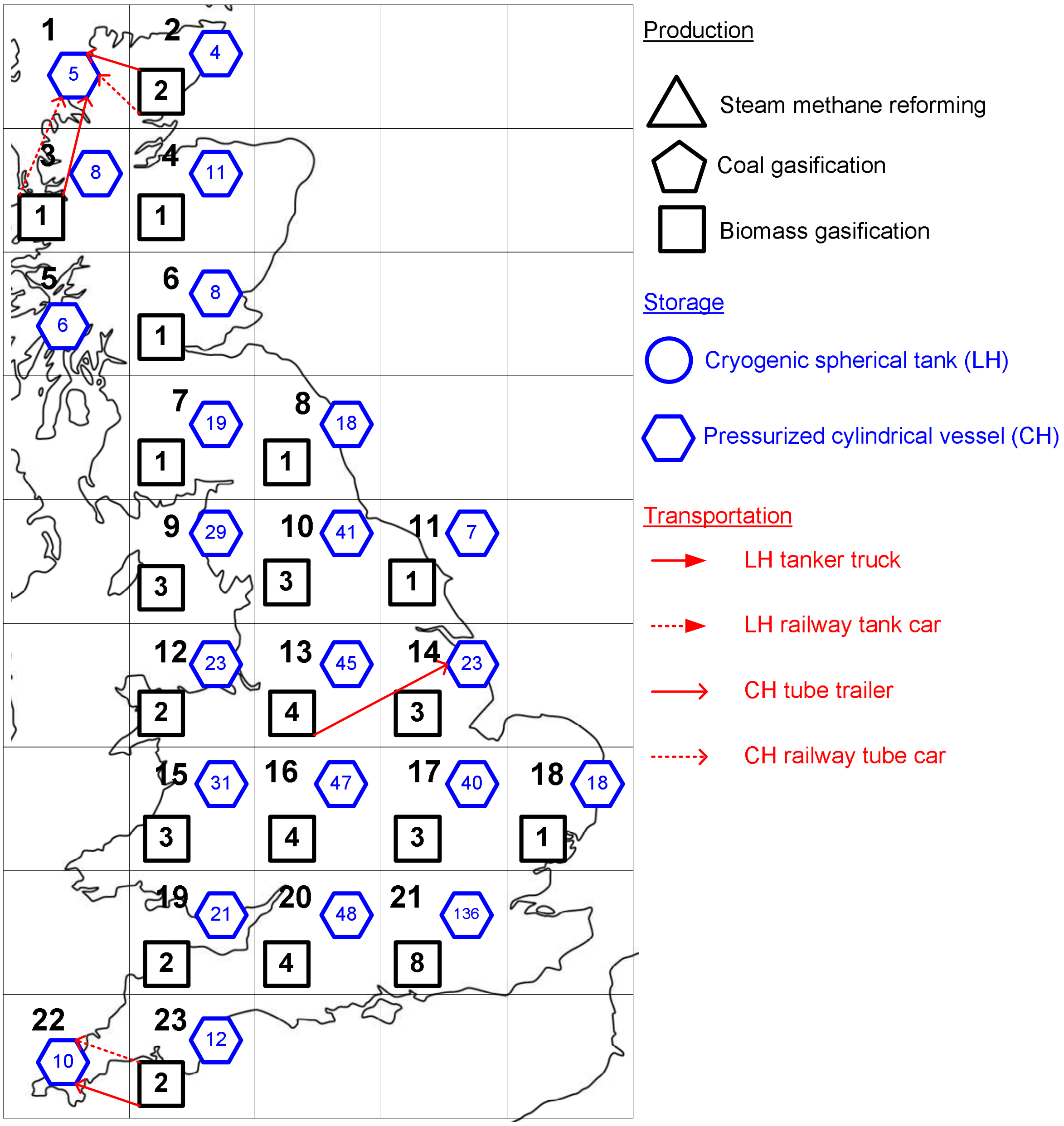

Figure 7: Minimum environmental impact solution.

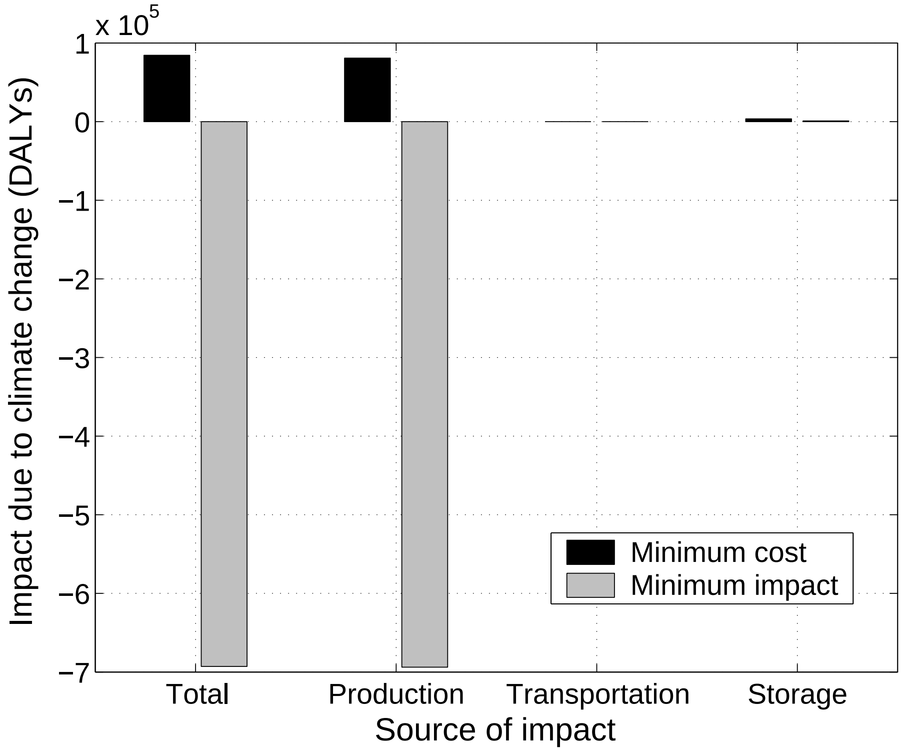

Figure 8: Main sources of impact.

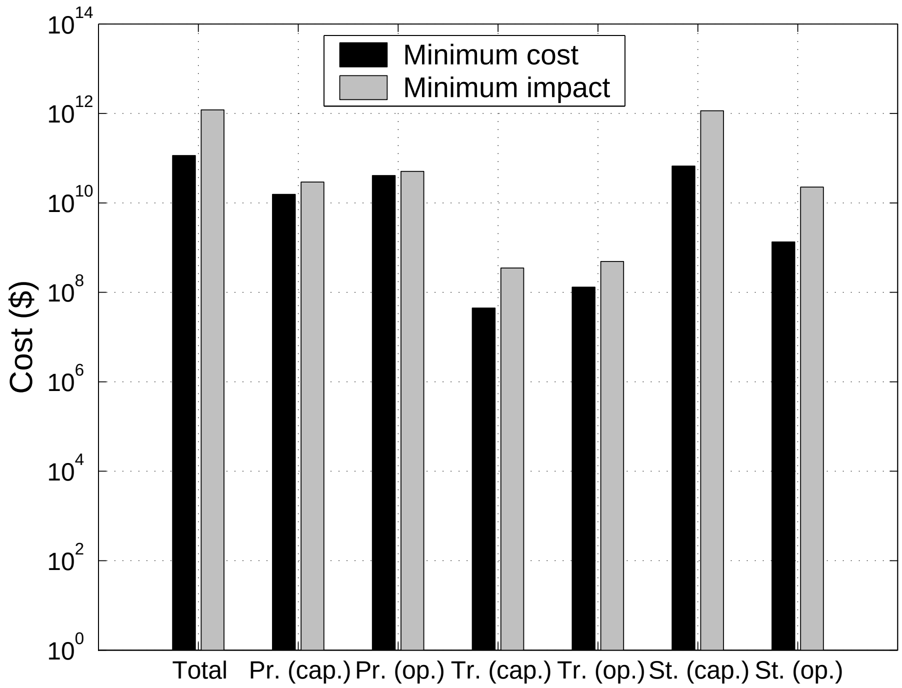

Figure 9: Breakdown of total cost into production (Pr.), transportation (Tr.) and storage (St.) capital (cap.) and operating (op.) costs for the extreme Pareto solutions.

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Table 1: Demand for t=1 (assume an annual increase of 5 %)

Grid	Hydrogen demand (ton/yr)
1	510
2	400
3	790
4	1,195
5	650
6	900
7	2,005
8	1,925
9	3,220
10	4,510
11	715
12	2,565
13	4,985
14	2,485
15	3,435
16	5,235
17	4,305
18	1,985
19	2,285
20	5,320
21	15,255
22	1,040
23	1,260

Table 2: Fixed and variable capital investment terms for plants for t=1 (assume an annual increase of 5 %)

	α^{PL}_{gpt} (\$)		β^{PL}_{gpt} (\$/kg)	
	LH_2	CH_2	LH_2	CH_2
Steam reforming	4.22×10^7	2.99×10^7	2.81	1.99
Coal gasification	7.55×10^7	6.08×10^7	5.04	4.05
Biomass gasification	1.11×10^8	7.15×10^7	7.42	4.77

Table 3: Fixed and variable capital investment terms for storage facilities for t=1 (assume an annual increase of 5 %)

	α^{ST}_{gst} (\$)	β^{ST}_{gst} (\$/kg)
Cryogenic spherical tank (LH_2)	9.05×10^6	209.17
Pressurized cylindrical vessel (CH_2)	1.40×10^8	3,247.25

Table 4: Environmental impact data

Technology	Impact due to climate change
Steam reforming (DALYs $\cdot$ kg^{-1})	3.34×10^{-6}
Coal gasification (DALYs $\cdot$ kg^{-1})	6.07×10^{-5}
Biomass gasification (DALYs $\cdot$ kg^{-1})	-2.58×10^{-5}
Cryogenic spherical tank (LH_2) (DALYs $\cdot$ kg^{-1})	1.44×10^{-7}
Pressurized cylindrical vessel (CH_2) (DALYs $\cdot$ kg^{-1})	3.20×10^{-8}
Tanker truck (LH_2) (DALYs $\cdot$ ton^{-1} $\cdot$ km^{-1})	3.49×10^{-8}
Tube trailer (CH_2) (DALYs $\cdot$ ton^{-1} $\cdot$ km^{-1})	7.77×10^{-8}
Railway tank car (LH_2) (DALYs $\cdot$ ton^{-1} $\cdot$ km^{-1})	7.85×10^{-9}
Railway tube car (CH_2) (DALYs $\cdot$ ton^{-1} $\cdot$ km^{-1})	1.20×10^{-8}

Table 5: Computational results for $t \leq 5$

	Variables[a]					
	binary	discrete	continuous	equations[a]	time (s)	cost (\$)[b]
t=1						
Full space	2,116	188	2,545	7,103	0.23	7.5308×10^{10}
Bilevel					0.29	7.5308×10^{10}
LB	2,308	-	2,545	7,111		
UB	2,116	45	2,545	7,103		
t=2						
Full space	4,232	376	5,084	14,200	1.55	8.5393×10^{10}
Bilevel					0.77	8.5314×10^{10}
LB	4,424	-	5,088	14,166		
UB	4,232	90	5,084	14,200		
t=3						
Full space	6,348	564	7,623	21,297	29.36	9.5180×10^{10}
Bilevel					11.34	9.5073×10^{10}
LB	6,540	-	7,631	21,221		
UB	6,348	135	7,623	21,297		
t=4						
Full space	8,464	752	10,162	28,394	191.75	1.0493×10^{11}
Bilevel					21.54	1.0507×10^{11}
LB	8,656	-	10,174	28,276		
UB	8,464	180	10,162	28,394		
t=5						
Full space	10,580	940	12,701	35,491	255.97	1.1492×10^{11}
Bilevel					44.16	1.1468×10^{11}
LB	10,772	-	12,717	35,331		
UB	10,580	225	12,701	35,491		

[a]Variables and equations in the first iteration
[b]Objective function value with 1% optimality gap

Table 6: Computational results for $t \geq 6$

	Variables[a]					
	binary	discrete	continuous	equations[a]	time (s)	cost ($)[b]
t=6						
Full space	12,696	1,128	15,240	42,588	523.32	1.24137×10^{11}
Bilevel					44.39	1.24227×10^{11}
LB	12,888	-	15,260	42,386		
UB	12,696	270	16,098	42,588		
t=7						
Full space	14,812	1,316	17,779	49,685	1,558.95	1.34135×10^{11}
Bilevel					55.22	1.33602×10^{11}
LB	15,004	-	17,803	49,441		
UB	14,812	315	18,780	49,685		
t=8						
Full space	16,928	1,504	20,318	56,782	795.90	1.42362×10^{11}
Bilevel					180.84	1.42449×10^{11}
LB	17,120	-	20,346	56,496		
UB	16,928	360	21,462	56,782		
t=9						
Full space	19,044	1,692	22,857	63,879	1,239.45	1.51041×10^{11}
Bilevel					185.33	1.51059×10^{11}
LB	19,236	-	22,889	63,551		
UB	19,044	405	24,144	63,879		
t=10						
Full space	21,160	1,880	25,396	70,976	7,195.72	1.59968×10^{11}
Bilevel					443.42	1.59467×10^{11}
LB	21,352	-	25,432	70,606		
UB	21,160	450	26,826	70,976		

[a]Variables and equations in the first iteration
[b]Objective function value with 1% optimality gap

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