Australia-Japan telecoupling of wind power-based green ammonia for passenger transportation: Efficiency, impacts, and sustainability

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ABSTRACT

Ammonia is a renewable energy medium appropriate for distant trading; therefore, many countries and companies have formulated ambitious strategies to develop energy transitions to use green ammonia for transportation systems. However, the associated social, economic and environmental impacts, and the overall viability and sustainability of these transitions are still a mystery, because of the lack of sufficiently complicated evaluations. To fill this gap, an integrated life cycle assessment and emergy evaluation (LCA-EME) method was developed and applied to synthesize, compare and recognize the hotspot nodes of resource depletion, emissions and impacts, and to quantify the exploitation and utilization efficiencies, environmental loadings and sustainability of the Australia-Japan telecoupling of wind power-based ammonia for electric vehicles (EV) and hydrogen fuel cell vehicles (FCV) used for passenger transportation, compared with two fossil fuel-based EV transportation systems. The results revealed that the transition to ammonia-based fuels can reduce nonrenewable energy consumption by >29.64% and Greenhouse Gas (GHG) emissions by >10.00%; however, the demand for emergy resources >2.03 times and biotic endpoint impacts >1.56 times, both of which mainly occurred in the sending subsystem of the telecoupling interaction. The results highlighted the necessity of internalizing the 'external' resource stress and its biotic impacts, increasing the utilization efficiency and the recycling rate of minerals and fresh water, and decreasing the endpoint impacts to guarantee the sustainability of the telecoupled energy transitions. Integrated LCA-EME was confirmed as a valuable tool for handling complex, multi-nodal nexus problems of telecoupling, which is widely needed for energy transition strategy making.

1. Introduction

Ammonia has the highest volumetric hydrogen content of any liquid fuel (including gasoline, liquefied natural gas, ethanol and even liquid hydrogen) [1], and its high energy density is accompanied by a lower economic cost per unit of energy production, as well as greatly

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simplified operations for its handling, distribution, and storage [2]. In addition, ammonia can be produced from water vapor and nitrogen through an electrochemical synthesis process [3], and it can be used as a feedstock for electricity generation or to power fuel cells by directly supplying it to a solid oxide fuel cell (SOFC) [4], or by cracking it to hydrogen as the energy feedstock for proton exchange membrane fuel cells (PEMFC) [5]. All these characteristics make ammonia a potential track for the development of a green energy economy in areas with abundant renewable energy resources, like Australia [6,7], and it offers a vast potential for the storage as well as the telecoupling of renewable energy between regions with high renewable energy intensity and regions lean in renewable resources, like Japan [8].

Considering that 18.6% of total Greenhouse Gas (GHG) emissions in Japan in 2019 were contributed by the transportation sector, in which emissions from automobiles accounted for 86.1% [9], the Japanese government set out the Japan Revitalization Strategy that aims to increase the percentage of new energy vehicles (NEVs) to be 50%–70% of new car sales by 2030. Furthermore, they expect all passenger vehicles will be replaced by NEVs by 2050 [10]. In addition, to promote national energy security, Japan has a strategy to cut its dependence on fossil fuels and to increase the diversity of its energy mix, since the transportation sector contributes 23.3% of Japan's national total energy consumption [11].

Many Life Cycle Assessment (LCA) studies have explored the attractive characteristics of green ammonia in decreasing fossil fuel consumption and lowering GHG emissions, especially using wind power-based ammonia [12], because wind power has been the fastest developing fraction of worldwide electricity over the past three decades [13]. Increasing the quantity of green ammonia/hydrogen-based NEVs will also help accomplish this purpose [14]. All of these results, make importing green ammonia for NEV transportation, a likely ideal option for Japan to meet all of the above demands. However, fossil fuel is not the only cost that needs to be considered to determine the viability and sustainability of ammonia/hydrogen as the basis for the vehicle transport sector. Similarly, GHG emissions are not the only environmental impact that needs to be considered in making energy transition strategies [15]. For example, the criticality of rare minerals (like cobalt, lithium, *etc.*) and their impacts on the viability of wind power-based green ammonia and the use of NEVs are both receiving more attention [16,17]. Less fossil fuel consumption does not always mean a high energy efficiency. For example, the well-to-pump energy efficiency of photovoltaic FCV is only 80% that of conventional gasoline vehicles [14]. Furthermore, using ammonia for power generation could lead to increased regional $PM_{2.5}$ concentrations in the atmosphere, which will cause a higher rate of human health impacts [18]. Thus, all the associated environmental and energy impacts of green ammonia interactions in the two telecoupled countries or regions (*i.e.*, the sending and receiving subsystems) and in the spillover subsystem (areas outside the two telecoupled countries, but affected by the telecoupling actions) are still not clear. In addition, the processing efficiencies of these 'options' are still a mystery due to the lack of a sufficiently complicated evaluation, and therefore, there is a question about the overall viability and sustainability of these energy transitions.

Although LCA is widely applied for quantifying resource utilization and the environmental impacts of alternatives under research and development (R&D), it has also been widely criticized from many points of view, *e.g.,* the use of subjective boundaries [19], the lack of uncertainty analysis [20], and the disparity of dimensions and amphibolous conclusions derived from the analysis [21]. It is still a problem within LCA to deal with varying environmental impacts quantified in different and incomparable units, *e.g.,* global warming potential (GWP) measured in grams of CO_2 eq., and terrestrial acidification potential (TAP) in grams of SO_2 eq. *etc*. The LCA results for a suite of alternatives are often combinations of pros and cons; *i.e.,* the excellent emission reduction ability of one alterative is often accompanied by increasing environmental impacts of some other aspects as a tradeoff. Many multi-criteria

decision analysis (MCDA) methods, like the Analytic Hierarchy Process (AHP), Technique for Order Preference by Similarity to an Ideal Solution (TOPSIS) and Preference Ranking Organization Methods for Enrichment Evaluations (PROMETHEE) *etc.*, can be applied to solve this problem, but the "objective weighting" scoring systems employed in these tools can sometimes be extremely biased. Thus, weighting is not allowed by ISO14044 in comparative assertions disclosed to the public [22]. Consequently, after a complicated quantification analysis and a long list of specific ecological-economic characteristics are considered, amphibolous conclusions or no conclusion about the holistic superiority of different options are found in many LCA studies. For example, it was disclosed that the forest biomass-based EV and FCV transports result in a reduction of net GHG emissions and freshwater eutrophication (FE), but also caused an increase photochemical oxidation [23]. A widely accepted method to portray a comprehensive picture of the sustainability of alternatives considering the tradeoffs among resource depletions, environmental impacts and other ecological economic characteristics is still lacking, and is urgently needed for strategy making at different regions/scales [24,25].

Emergy, defined as the total amount of a specific kind of energy (*e.g.*, solar joules) that was required for producing a product, such as an energy, material or service flow [26], provided a solution for comprehensively visualising the multiple tradeoffs from the perspective of the biophysical donor value of the output. Through over three decades of accumulated studies, the Unit Emergy Values (UEVs, *i.e.*, the emergy per unit output) for many kinds of energies, materials and services have been determined and are available in the literature and online (http://www.emergysociety.com/emergy-society-database/). Simultaneously, emergy evaluation (EME) has become a well-tested tool for integrated evaluation of ecological economic systems and processes, including new energy production systems [27,28], trading and consumption systems and processes carried out within a larger life cycle perspective [29,30]. From an opposing perspective, EME has been criticized for a suite of perceived shortages, *e.g.*, the lack of utility-side considerations, especially the environmental impact of emissions [31], the use of subjective system boundaries [32], and the lack of uncertainty analysis [24,33].

Some researchers recognized that the joint use of LCA and EME can complement the deficiencies of both, and provide better insights on the complexity of system performance, *i.e.*, combining the biophysical donor-side emergy cost evaluation and the downstream user-side environmental impacts [34]. Based on this idea, a few studies have taken a new synthesized approach linking these two environmental evaluation methods [35–37]. However, LCA and EME were used in parallel or hybrid mode in most of these studies, which just provide complementary results from different viewpoints, *i.e.*, donor-vs. user-perspectives. A few studies tried to integrate LCA into EME, by considering the impacts of emissions in the emergy index systems [38,39], but a widely accepted methodological framework is still lacking [32]. Another important issue with regard to the integration of LCA and EME is to quantify and decrease the uncertainty of UEVs by proposing optional frameworks for UEV calculation based on life cycle inventory (LCI), which has been used and modified widely in conventional LCA cases [40–42]. In addition to this innovation, the treatment of uncertainty is still needed to increase the reliability and credibility of both LCA and EME on multi-scales from data acquisition to model simulation [43].

The telecoupling framework [44] was first applied in evaluating the environmental impacts and sustainability of transiting energy use from fossil fuels to renewable energies, *e.g.*, green ammonia, in this study. Two green ammonia scenarios and two fossil fuel scenarios were set up and classified into 3 subsystems, respectively, *i.e.*, the sending subsystem, the receiving subsystem, and the spillover subsystem. An integrated LCA-EME was launched for a more complex quantification and evaluation of the energy-water-material-emission-impact-sustainability multi-nodal nexus issues of the energy transition at both system and subsystem levels. Both the mid-point potential impacts of emissions and

the biotic endpoint impacts, including but not limited to global warming and human health effects, were quantified and brought into the emergy indices for evaluating efficiency and sustainability. The uncertainties of the LCI database transferred to the evaluation results (for resource depletion, emissions, impacts, efficiency, and sustainability) were quantified and considered in statistical comparisons among the scenarios at both system and subsystem levels.

2. Scenarios and methods

2.1. Scenarios and life cycle inventory

Australia is the largest provider of both coal and natural gas used in Japan. Simultaneously, with abundant land and renewable energy resources, including wind power resources, Australia is one of the largest potential green ammonia suppliers for East Asian countries, especially Japan [45]. Thus, we assumed that all the coal, natural gas and green ammonia evaluated in this study were mined (coal and NG) or produced by wind power (assuming that chlor-alkali electrolysis was employed to produce hydrogen from water, and the electrochemical synthesis process was used to produce ammonia using hydrogen and diatomic nitrogen) in Australia, and then shipped (at low temperature and high pressure for NG and green ammonia, and normal temperature and pressure for coal) from Sydney to Tokyo (8700 km) for use in power or hydrogen production in Japan. The coal and NG mined in and imported from Australia were transformed to electricity in thermal power plants in Tokyo, and then sent to supply electricity to EVs for passenger transport through power charging stations all over Japan (Scenarios 1 and 2, S1 [coal EV] and S2 [NG EV]) (Fig. 1 (1) & (2), Supplementary Table 1). Two different wind power-based green ammonia utilization routes were considered. The first is turning green ammonia into electricity through a solid oxide fuel cell (SOFC, 180 kW), and then supplying the electricity to EVs through power charging stations (Scenario 3, S3 [ammonia EV], Fig. 1 (3), Supplementary Table 1). The other route

is cracking green ammonia into hydrogen through a proton exchange membrane fuel cell (PEMFC), and then supplying the hydrogen to FCVs (Scenario 4, S4 [ammonia FCV] (Fig. 1 (4), Supplementary Table 1).

The functional unit of 1 km transport carried by a passenger EV or FCV in Japan was set as the reference for related inputs to and output and emissions from each subsystem. Each of the four scenarios (S1, S2, S3, S4) were further classified into four processing steps for life cycle inventory (LCI) and further LCA-EME (Fig. 1), i.e., the energy feedstocks (coal, NG, and wind power-based green ammonia) production in Australia (SN-1[a], in the sending subsystem) (Footnote: [a] SN-1, scenario N step 1), shipping energy stocks overseas from Sydney to Tokyo (SN-2, for the spillover subsystem), power or hydrogen production in Japan (SN-3, for the receiving subsystem) and running a passenger EV or FCV in Japan (SN-4, also for the receiving subsystem). The target factors for each processing step were quantified according to the relevant publications [46–48] (Supplementary Table 1).

The LCI was conducted on SimaPro 8.5.2.0 for the averages and variances of all life cycle energy and material inputs to and emissions from each processing step of the four scenarios.

2.2. Integrated LCA and EME methods

All raw data (averages and variances) were converted to emergy by multiplying by the relevant published UEVs [26,50–54] which were all converted to 1.20E+25 seJ/yr planetary solar equivalent baseline [55], if they were not being published in that way before (Supplementary Tables 2 and 3).

2.2.1. Emergy inputs and resource depletions

All energy and material inputs or depletions were counted as inputs. Consequently, the depletion of resource availability (RA) was not counted in the environmental impact to avoid double counting.

To fill the needs of index calculations, the inputs were classified into local free renewable inputs (R, including geothermal and biomass
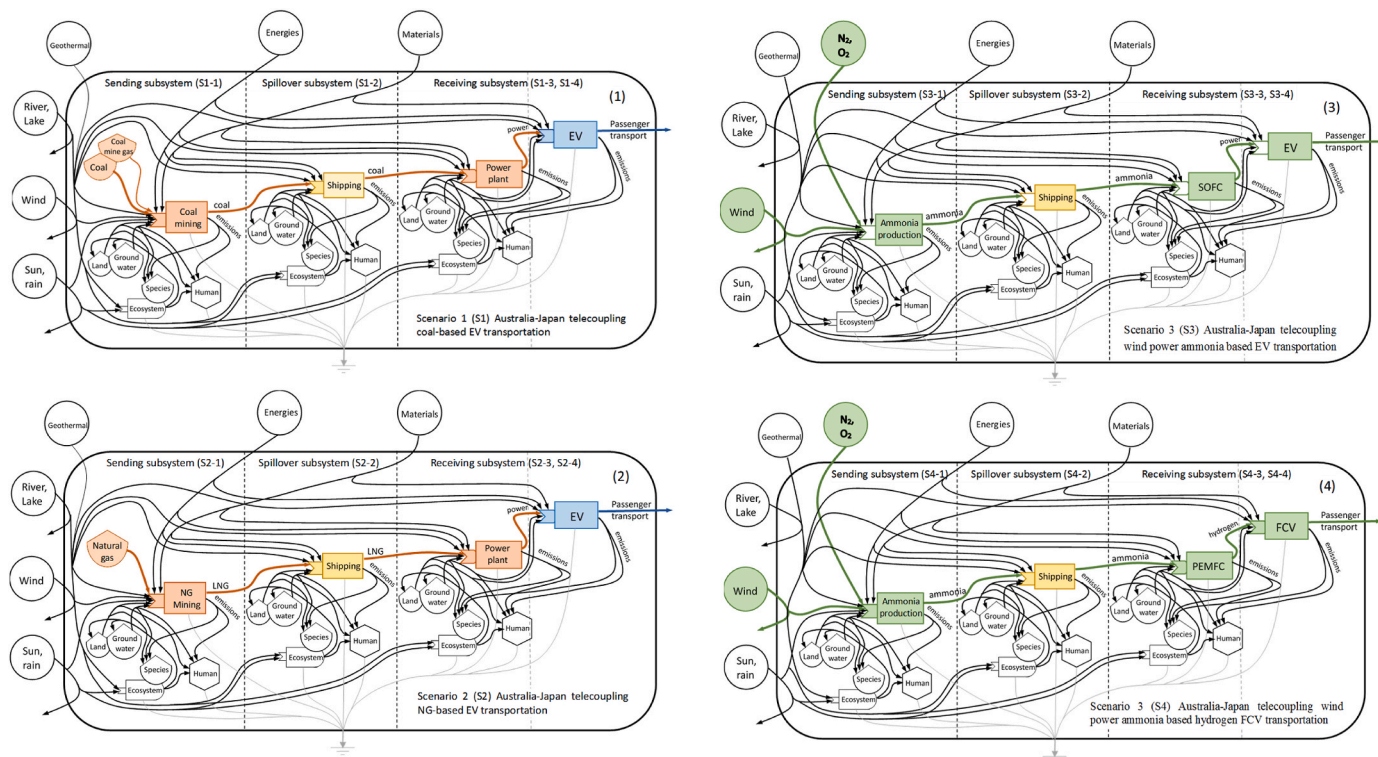

Fig. 1. Conceptual Energy Systems Language [49] diagram of the four telecoupling scenarios. (1) Energy system diagram of the coal-based EV scenario (S1). (2) Energy system diagram of the NG-based EV scenario (S2). (3) Energy system diagram of the wind power-based green ammonia EV scenario (S3). (4) Energy system diagram of the wind power-based green ammonia hydrogen FCV scenario (S4).

energy, solar radiation, wind kinetic energy, river and lake water, nitrogen, oxygen, and the global average R density over the occupied land area ($8.05E+10$ sej/m^2/yr)[b] (Footnote: [b] based on $1.20E+25$ sej/yr baseline, and the area of land on Earth is $1.49E+14$ m^2), local free non-renewable inputs (N, including ground water, besides coal and coal mine gas for S1-1 and NG for S2-1), and purchased inputs (F, including all other energy and mineral inputs).

2.2.2. Emergy emissions and impacts

The emissions and their biotic endpoint impacts were quantified following the classification framework of ReCiPe 2016 [56], but their relative magnitudes were based on an emergy evaluation.

2.2.2.1. Emissions and potential impacts.

Fourteen kinds of emissions were counted and organized based on the attention given to their recognized impacts, and the availability of UEVs. The GHG emissions of CO_2, CH_4, N_2O to the air affect global warming (GW), the emissions of NH_3, NOx, SO_2, SOx to the air affect terrestrial acidification (TA), the emissions of $POCl_3$, PCl_5, PCl_3 to water, and P to water and soil affect freshwater eutrophication (FE), and the emissions of N, NH_4^+ and NO_3^- to water affect marine eutrophication (ME).

The potential impacts of these emissions were quantified on the same uniform basis, as emergy intensity, by multiplying by their UEVs which were calculated based on their biogeochemical cycles during the Pre-industrial Era [50] (Supplementary Table 2). The potential impacts of these emissions were classified into the above four "midpoint" impact categories (*i.e.*, GW, TA, FE and ME), although what was quantified is the potential impacts of these emissions, which is not limited only to the categories in which they were classified. Consequently, the potential impacts of all these emissions can be directly summed up and compared for a holistic picture, but this synthesis ignores the interactions among them.

2.2.2.2. Biotic impacts.

Two endpoint impacts were considered to capture the biotic impacts in the areas of protection (AoP) due to the emission of pollutants from and the resource stress caused by the 4 scenarios, following the classification of ReCiPe 2016 [56], *i.e.*, the damage to human health (HH) and ecosystem quality (EQ), respectively, indicated by the Disability-Adjusted Life Years (DALY) and the Potentially Disappeared Fraction of species (PDF) [57,58], which were quantified by multiplying with the relative midpoint to endpoint factors for the hierarchic perspectives (Supplementary Table 3). Besides the above four midpoint environmental impact categories, water stress (WS) and land occupation (LO) were also counted as the key factors behind malnutrition and the loss of biodiversity in this study, according to ReCiPe 2016 (Supplementary Table 3).

The annual emergy flows per person in Australia and Japan in 2016, were calculated based on the National Environmental Accounting Database (NEAD, https://cep.ees.ufl.edu/nead/) for national emergy consumption in 2008, and the National Yearbook (http://www.stat.go.jp/english/data/nenkan/67nenkan/index.html) for real GDP and population in 2016, based on the observation that a long-term linear correlation exists between real GDP and total national emergy consumption [51]. As a result, $1.71E+17$ sej/capita/yr in Australia in 2016 was used for the sending subsystem in the four scenarios (fuel mining and production in Australia, SN-1); while $6.02E+16$ sej/capita/yr in Japan in 2016 was used for the receiving subsystem (the power or hydrogen production and consumption in Japan, SN-3 and SN-4). The mean value of the emergy flow per capita per year between Australia and Japan, $1.16E+17$ sej/capita/yr, was applied to the spillover subsystem (the international shipping between Australia and Japan, SN-2).

There are $1.95E+06$ species living on the land, and in freshwater, and in the marine system as assumed in ReCiPe 2016 [56]. Based on the $1.20E+25$ seJ/yr planetary emergy baseline [55], this gives $6.15E+18$ sej/species/yr as an estimate of the cost for maintaining one species for

one year.

2.2.3. EME indices

All four scenarios were assumed to be steady state systems or processes that exist without changes in their internal storages. Consequently, the emergy outputs from the four energy systems or scenarios were equal to their corresponding inputs, according to the emergy algebra [26].

Besides the classical emergy indices, *i.e.*, Unit Emergy Value (UEV), Emergy Yield Ratio (EYR) [26], Environmental Loading Ratio (ELR) and Emergy Sustainability Index (ESI) [59], two new emergy indices were calculated by bringing the emissions and the biotic endpoint impacts of resource depletions and emissions into consideration.

All emissions are essentially wasted inputs. More emissions mean lower utilization efficiency of the resource inputs, besides causing a higher potential for environmental impacts per unit input, which is captured by the Emergy Waste Ratio (EWR).

$$EWR = \frac{W}{R + N + F} \tag{1}$$

$$W = \sum_{i=1}^{n} GW_i + \sum_{j=1}^{m} TA_j + \sum_{k}^{o} FE_k + \sum_{l}^{p} ME_l \tag{2}$$

R is the local free renewable inputs, while N is the local free non-renewable inputs, F is purchased inputs. W is the total emergy of the emissions, while GW_i is the *i*th GW emission, TA_j is the *j*th TA emission, FE_k is the *k*th FE emission, and ME_l is the *l*th ME emission.

The biotic endpoint impacts of emissions and resource depletions are quantified by the emergy loss considering human and other species regenerations, these can be measured, respectively, by corresponding changes to DALY and PDF. The larger the endpoint impact per unit output, the higher the confirmed biotic endpoint impacts of the system or process under study, as indicated by the Biotic Impact Ratio (BIR).

$$BIR = \frac{BI}{R + N + F} \tag{3}$$

$$BI = \sum_{i=1}^{n} DALY_i + \sum_{j=1}^{m} PDF_j \tag{4}$$

BI is a measure of the emergy-based biotic endpoint impacts. $DALY_i$ is the Disability Adjusted Life Years impacts of the *i*th emission/resource depletion, and PDF_j is the Potentially Disappeared Fraction of species impact of the *j*th emission/resource depletion.

All the above indices were calculated for each telecoupling scenario as a whole system, and its three subsystems (sending, receiving and spillover subsystems), considering the spatial impacts.

2.2.4. Uncertainty quantification and statistical analysis

The uncertainties of each input and emission obtained from SimaPro 8.5.2.0 was given by the variables' averages and standard deviations. After being converted to emergy through their relative UEVs, all standard deviations of inputs and emissions were propagated into the sub-total and total uncertainties of the emergy inputs, emissions and output through using the first-order Taylor series expansion method [33,60], without making assumptions about the unknown probability distributions in determining the uncertainties. However, the first-order Taylor series expansion method is not applicable to quantify the uncertainties of the indices which were not simply sums of categories. Thus, an assumption has to be made at this stage for simulating the propagated uncertainties in emergy indices, because there is no way to determine the actual distributions of each subtotal and total emergy input, emissions and output. The uncertainties of the emergy indices, *i.e.*, EYR, ELRs, ESIs, EWR and BIR, were quantified by performing a Monte Carlo simulation 10,000 times, and by assigning a normal distribution to all input, emission and impact categories to meet the prerequisites of

further statistical analysis, One-way Analysis of Variance (ANOVA). The uncertainties of the categorized inputs, emissions, impacts and emergy indices were quantified by the percent of the standard deviation (SD%) used to clarify the hotspots of uncertainties in this study (Supplementary Table 4).

ANOVA with Tukey's honest significant difference (HSD) test was performed to compare the inputs, emissions, impacts, and emergy indices among the four scenarios, based on performing a Monte Carlo simulation 100 times, in which a normal distribution was required for and assigned to all items under analysis. Data analyses were carried out by R version 3.3.1, with the statistically significant differences being set at $P < 0.05$. All the differences (higher or lower) being mentioned in the following sections were significant differences, and only results with significant differences were further compared quantitatively between the averages.

3. Results and discussion

3.1. Efficiency, impacts and sustainability of the telecoupled energy transition at the system level

Fossil fuel shortage and global warming caused by energy sourced GHG emissions are key driving forces for the development of new energy vehicles, and, especially, green ammonia as a carbon-free fuel/hydrogen carrier [1]. Our results show that replacing fossil fuel (coal and NG) with wind power-based green ammonia as the energy feedstock for EV and hydrogen FCV transportation are good solutions to address the two problems above with significant reductions in both nonrenewable energy depletion (>29.64%, Fig. 2. (1)) and GHG emissions (>10.00%, Fig. 2. (2)), which are results consistent with many previous studies [12, 14].

However, the production, transportation, conversion and utilization processes of energies, including green ammonia, also profoundly impact our socio-environmental systems across multiple dimensions, and these impacts are not simply limited to fossil fuel consumption and GHG

emissions [61]. Our results showed that, as a tradeoff for the reductions in non-renewable energy consumption and GHG emissions, the demands of the two green ammonia scenarios for renewable energies (>2.38 times), materials (>2.42 times), water (>2.08 times) and land (>1.52 times) were all significantly higher than that of the two fossil fuel scenarios (Fig. 2. (1)). In addition, the TA emission from S4 was higher (>8.61%) than the two fossil fuel scenarios (Fig. 2. (2)). Finally, compared with the two fossil fuel scenarios, both of the two green ammonia scenarios had over 2.03 times the total emergy demand (Fig. 2. (1)), accompanied by over 1.56 times the biotic endpoint impacts (Fig. 2. (3)), without a reduction in the complex emissions (Fig. 2. (2)), which were generally considered important for their impacts in GW, TA, FE and ME.

With a much higher renewable fraction of emergy input (>52.67%), both the EYR and ELR of the two green ammonia scenarios (S3 and S4) were superior to the values of the two fossil fuel scenarios (*i.e.,* the EYR was higher and the ELR was lower). Finally, the two green ammonia scenarios had similar ESIs, which were over 5.43 times that of the two fossil fuel scenarios (Fig. 2. (4)). Besides, the EWRs and BIRs of S3 and S4 were, respectively, less than 0.85 and 0.83 times that of the two fossil fuel scenarios.

Minerals are the largest emergy input category to all four energy telecoupling scenarios, which accounted 62.12%, 64.86%, 78.54% and 75.46% of emergy inputs to S1, S2, S3 and S4, respectively (Fig. 2. (1), Fig. 3). Specific material input structural analysis showed that over 96% of the mineral inputs to S1 and S2 were consumed at the EV steps (S1-4 and S2-4), while over 67% and 32% of the mineral inputs to S3 and S4 were consumed in the green ammonia production step (S3-1 and S4-1) and by the NEVs (EV and FCV) steps (S3-4 and S4-4), respectively (Fig. 3, Supplementary Fig. 1). This observation quantitatively explains why so many material-energy nexus studies have been launched to address problems at these two hot-spots. Previous studies have been focused on the supply-demand of rare materials and metals needed for wind turbine manufacture, fuel cells, and EV batteries [62,63], the catalysts for both green ammonia synthesis [64] and cracking [65], and
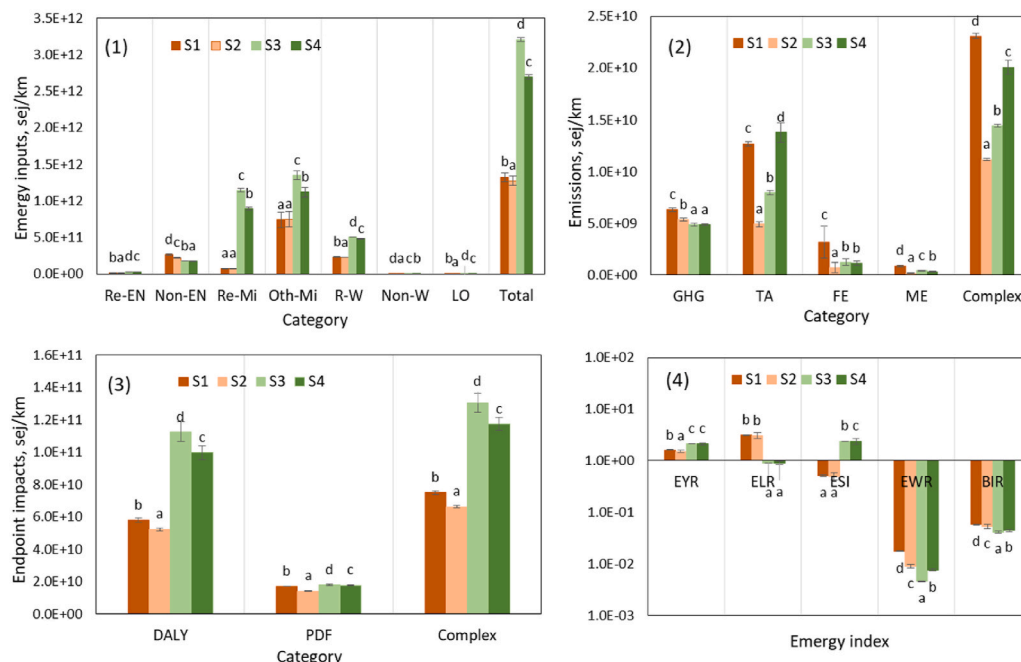

Fig. 2. Emergy inputs, emissions, impacts and sustainability of the four telecoupling scenarios. (1) Emergy inputs to the four telecoupling scenarios. (2) Emergy emissions from the four telecoupling scenarios. (3) Emergy-based biotic endpoint impacts of the four telecoupling scenarios. (4) Emergy indices of the four telecoupling scenarios (dimensionless). Note: 1. Re-EN, renewable energy input; Non-EN, nonrenewable energy input; Re-Mi, renewable mineral input; Oth-Mi, other mineral input; R–W, renewable water input; Non-W, nonrenewable water input; LO, land occupation; Total, total input. 2. GW, global warming emissions; TA, terrestrial acidification emissions; FE, freshwater eutrophication emissions; ME, marine eutrophication emissions; Complex, complex emissions; 3. DALY, disability-adjusted life years; PDF, potentially disappeared fraction of species; Complex, complex biotic endpoint impacts; 4. EYR, emergy yield ratio; ELR, environmental loading ratio; ESI, emergy sustainability index; EWR, emergy waste ratio; BIR, biotic impact ratio. 5. The different lowercase letters indicate significant differences at P <

0.05 level among different scenarios at each telecoupling subsystem and the whole system or process level. The following figures use the same definitions for the comparison of scenarios.
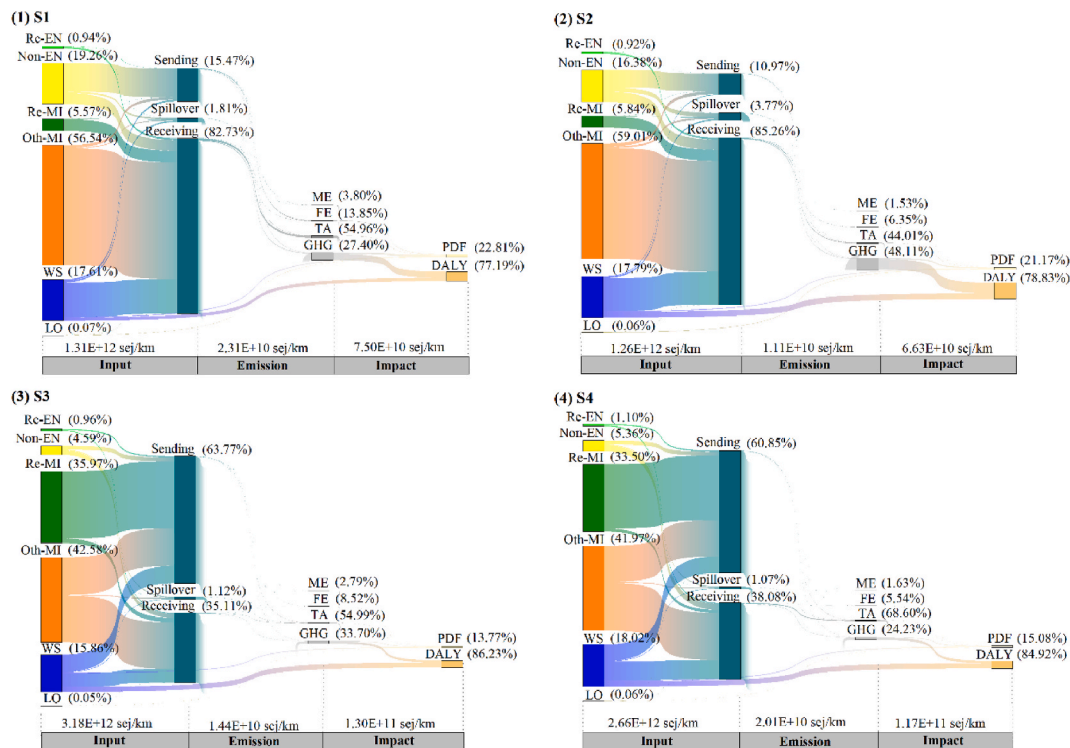
Fig. 3. Structure of emergy-based inputs, emissions, and biotic impacts of the four telecoupling scenarios. (1) Structure of inputs, emissions, and biotic impacts of the coal-EV scenario (S1). (2) Structure of inputs, emissions, and biotic impacts of the NG-EV scenario (S2). (3) Structure of inputs, emissions, and biotic impacts of the wind power-based green ammonia EV scenario (S3). (4) Structure of inputs, emissions, and biotic impacts of the wind power-based green ammonia hydrogen-FCV scenario (S4).

the impacts of material structure on vehicle energy efficiency [16] and fuel economy [66]. In recent years, the recycling problems of the large amount of non-biodegradable materials, such as in lithium-ion batteries [67] and wind turbine blades [68] at the end-of-life (EOL) stage are receiving more attention. Besides nitrogen, our results showed that the fractions of gravel, sodium chloride, barite and calcite each accounted for more than 5% of the mineral inputs to wind power-based green ammonia production. This was also true for the gravel, manganese, iron and cadmium inputs to EVs, and the gravel, iron and cadmium inputs to FCVs (Supplementary Fig. 2). Thus, the security of supplements and the utilization efficiency and recycling of these minerals also need to be specifically considered for material security and sustainability of the two green ammonia scenarios.

Many key global sustainability challenges, *e.g.*, energy scarcity, water depletion, and environmental impacts are complicated and closely intertwined [69]. Water and energy use are tightly interdependent with each other, and both are critical resources. Consequently, the nexus of energy and water has been studied in the past decade at both macro and micro scales, and these concerns were gradually expanded in recent years to include land [70], food [71], and emissions (especially GHGs) [25,72]. Many studies have explored that a large amount of virtual water is needed for the processes of extracting, transporting and processing fossil fuels, power plant cooling, and irrigating biofuel feedstock plants [73]. It was showed that shifting to low-carbon renewable energies, like wind and solar power, could potentially decrease the pressure on water, and reduce carbon emissions to some degree [74]. However, different opinions are not lacking either. For example, a hybrid input-output and process analysis showed that industrial water use induced by the infrastructure of a solar power plant had more than twice the lifecycle freshwater use of a coal-fired power plant [75].

Our results showed that the emergy inputs of water to S2, S3 and S4 were all significantly higher than that of energy, and the emergy inputs of water to both the two green ammonia scenarios were over 2.08 times

that of the two fossil fuel scenarios (Fig. 2. (1)). Furthermore, water stress replaced GHG emissions to become the largest biotic endpoint impacts factor for both of the two green ammonia scenarios (>68.49% and 57.21%, respectively, for S3 and S4. Fig. 3 (3) and (4)). Thus, the hidden role of virtual water behind wind power-based green ammonia/ hydrogen NEVs life cycle needs to be fully considered. How to increase the utilization efficiency and recycling rate of fresh water are essential for the viability of both wind power-based green ammonia/hydrogen NEVs development, as well as for regional human health security and biodiversity conservation.

GHGs reduction is one of the main targets of the renewable energy transition, including the development of green ammonia/hydrogen, a carbon-free fuel. However, our results found that GHG emissions was the largest emergy-based emission category for S2, while TA (emissions of NOx, NH$_3$, SO$_2$, SOx to air) was the largest emergy-based emission category for S1, S3 and S4, and the second largest emergy emission category for S2 (Fig. 2. (2)). Although GHG emissions from the two green ammonia scenarios were significantly lower than that of the two fossil fuel scenarios (S1 and S2), their TA, FE and ME emissions were all significantly higher than that of the NG scenario (S2). Finally, the complex of four types of emissions of the two green ammonia scenarios fell in between that of the two fossil fuel scenarios (Fig. 2. (2)). From the point of view of elemental analysis, the energy transition from S1 and S2 to S3 and S4 could bring a reduction in carbon emissions, but not in the nitrogen and sulfur produced (Supplementary Fig. 3). The nitrogen emissions, especially nitrous oxides (NOx) generated during green ammonia combustion has been widely acknowledged as a key environmental risk of ammonia utilization as a carbon-free fuel [76]. Our results showed that impacts from the emergy-based emissions of sulfur (SO$_2$) from the two green ammonia scenarios were over 1.20 times higher than that of the nitrogen species (NH$_3$, NOx, N$_2$O) emitted. The sulfur emissions from S4 were the highest among the four scenarios, which was 1.15, 3.01 and 1.93 times that of the sulfur emitted by S1, S2

and S3, respectively (Supplementary Fig. 3). It has been shown that, by the 1980s, the global cycles of balances for nitrogen, carbon and sulfur, had moved 2.5%, 5% and 71% away from their dynamic equilibrium that existed in the Preindustrial Era, circa 1850 [50]. As a result, apparently the global cycle of sulfur should be managed much more carefully than that of carbon and nitrogen.

3.2. Efficiency, impacts and sustainability of telecoupled energy transitions at the subsystem level

How to deal with the spatial mismatches between availability and demand for minerals, water and energy has been identified as a tough challenge for societies at both the global [77] and regional levels [78]. Economic globalization greatly enhanced the interactions among distant countries and regions through the flows of information and people, as well as by the intensive trade in goods and energies. These telescoping interactions filled the need for re-matching the spatially mismatched supply-demand of the above access, but this situation has been accompanied by many complicated social economic and environmental impacts. The telecoupling framework is a universal and hierarchical paradigm for examining these interactions, and it has assisted in gaining a better understanding of these complex social-ecological interactions [44]. Consequently, the telecoupling framework has been broadly implemented in diverse disciplines at the regional to international scales [79]. However, most of the previous telecoupling studies were focused on fossil fuel, electricity, and biofuel systems and trades [80].

Under the telecoupling framework, the LCA-EME results explored that transiting from fossil fuels to wind power-based green ammonia/hydrogen through a telecoupling between Australia and Japan will bring different impacts to the sending (Australia), receiving (Japan) and spillover (transport outside Australia and Japan) subsystems, with respect to resource depletion and in the expected environmental and social impacts.

In the receiving subsystem, Japan, both of the two green ammonia scenarios (S3 and S4) are better choices than the two fossil fuel scenarios (S1 and S2). For example, the green ammonia hydrogen FCV scenario (S4) had the lowest total emergy cost (Fig. 4. (1)), ME emissions (Fig. 4. (2)), Environmental Loading Ratio (ELR), and the highest Emergy Yield Ratio (EYR) and Emergy Sustainability Index (ESI, Fig. 4. (4)) among the four scenarios examined for the receiving subsystem. Furthermore, the green ammonia EV scenario (S3) had the lowest GHGs and complex

emissions (Fig. 4. (2)), and biotic endpoint impacts (Fig. 4. (3)) for the receiving subsystem, accompanied with the lowest Emergy Waste Ratio (EWR) and Biotic Impact Ratio (BIR, Fig. 4. (4)).

A different story was observed for the sending subsystem, Australia. In the sending subsystem (Fig. 5. (1)), both S3 and S4 required over 8.00 times the total emergy demand of the two fossil fuel scenarios (S1 and S2). This was due to the significantly higher input of minerals (>66.23 times), water (>21.70 times) and land occupation (>3.28 times) of S3 and S4 in Australia, besides an over 16.64 times increase in renewable energy consumption (Fig. 5. (1)). This trend was not accompanied by a significant reduction in the complex emissions (Fig. 5. (2)), but showed an over 3.62 times increase in the complex biotic endpoint impacts (Fig. 5. (3)). Specifically, S3 had total emergy demand (Fig. 5. (1)), complex emissions (Fig. 5. (2)), and biotic endpoint impacts (Fig. 5. (3)) all significantly higher than the other three scenarios, while S2 (NG based EV) had all of the above items significantly lower than the other three scenarios. However, both S3 and S4 had all their EYRs (>2.88 times), ELRs (<0.03 times), EWRs (<0.18 times) and BIRs (<0.45 times) better than the values of those indices for S1 and S2 (Fig. 5. (4)), due to a relatively higher input fraction of renewable natural resources. Finally, the sending subsystem of S3 and S4 had similar ESIs (about 6.32), which were over 8.24 times that of S1 and S2, but were accompanied by a much lower emergy production/conversion efficiency, as indicated by their significantly higher total emergy inputs (equal to the UEVs in this study, >8.00 times). The UEV of S3 was 9.99, 14.61 and 1.25 times that of S1, S2 and S4, respectively, for the sending subsystem (Fig. 5. (1)).

The spatial rematch of the supply-demand for energies, minerals and waters is the fundamental reason behind the above uneven impacts, *i.e.*, over 82% of the emergy inputs to S1 and S2 were consumed by the receiving subsystem (Fig. 3. (1) and (2)), while over 60.19% of the emergy inputs to S3 and S4 were consumed by the sending subsystems (Fig. 3. (3) and (4)).

As is widely known, Japan is an energy and mineral scarce country, and it has also been found to be a water scarce country, because it was one of the major international gross virtual water importers during the period 1996–2010 [81]. In contrast, Australia, as a water rich nation, was one of the key virtual water exporters, especially to Japan [82]. The results of this study showed that the water inputs to the sending subsystems of both S3 and S4 were over 21.70 times that of S1 and S2 (Fig. 5. (1)). Thus, the telecoupling transition between Australia and Japan from coal and NG to wind power-based green ammonia will
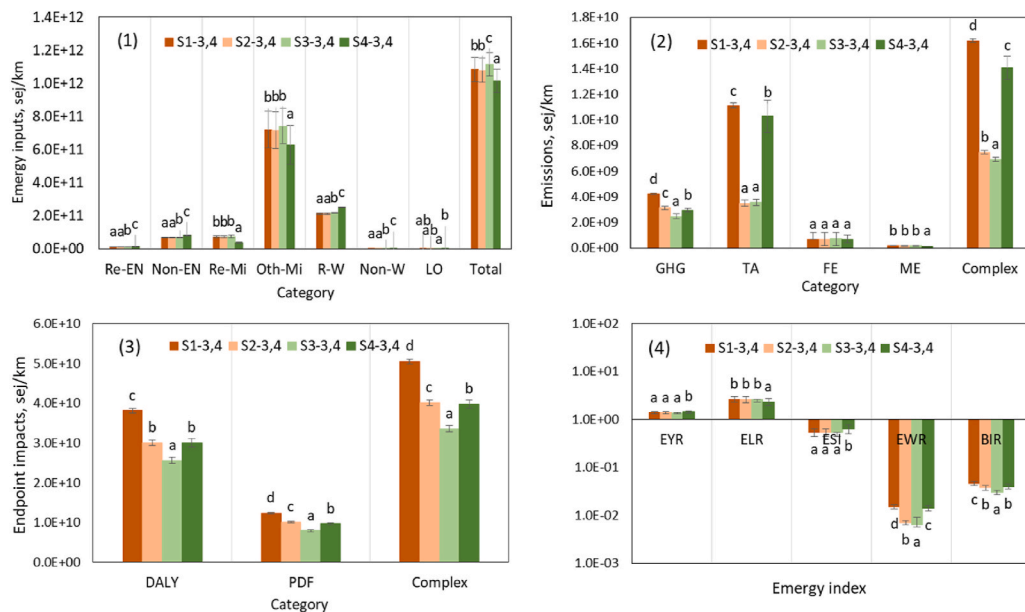

Fig. 4. Emergy inputs, emissions, impacts and sustainability of the receiving subsystems of the four telecoupling scenarios. (1) Emergy inputs to the receiving subsystems of the four telecoupling scenarios. (2) Emergy emissions from the receiving subsystems of the four telecoupling scenarios. (3) Emergy based biotic endpoint impacts of the receiving subsystems of the four telecoupling scenarios. (4) Emergy indices of the receiving subsystems of the four telecoupling scenarios (dimensionless).
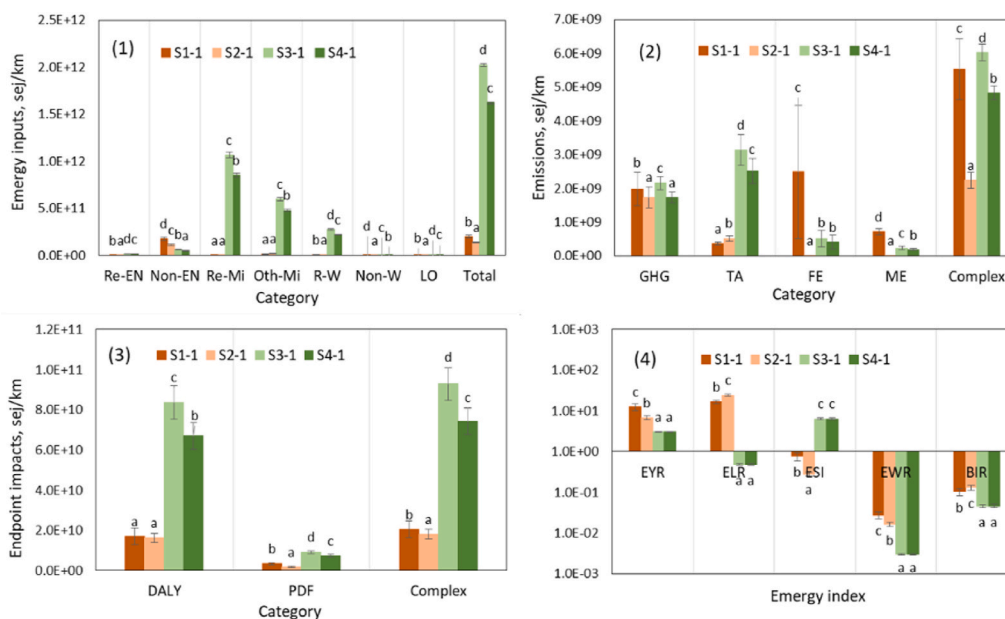
Fig. 5. Emergy inputs, emissions, impacts and sustainability of the sending subsystems of the four telecoupling scenarios. (1) Emergy inputs to the sending subsystems of the four telecoupling scenarios. (2) Emergy emissions from the sending subsystems of the four telecoupling scenarios. (3) Emergy based biotic endpoint impacts of the sending subsystems of the four telecoupling scenarios. (4) Emergy indices of the sending subsystems of the four telecoupling scenarios (dimensionless).

further strengthen the partnership between these two countries, through increasing the virtual water flow from Australia to Japan (Fig. 3).

"How to internalize the 'external' resource depletion and the accompanying biotic endpoint impacts caused by the telecoupling interactions in the sending subsystem?" will be one of the fundamental issues that must be resolved to guarantee and stabilize these telecoupling energy transitions. These transitions directly affect the energy and water security of Japan, and the conservation of resources, human health, and biodiversity in Australia. Other social, economic and environmental issues are also important, *e.g.*, the technological innovation needed for improving the efficiency of electrochemical synthesis of ammonia, the cracking/combustion of ammonia, the utilities and the final recycling rate of minerals are also key to the long-term success of these energy conversions [1].

Full detection of the effects of telecoupling activities on the spillover subsystems is also essential to guarantee global sustainability goals [83]. Our results showed that, in the spillover subsystem, both of the two green ammonia scenarios had their total emergy demands (Fig. 6. (1)), and their EYR, ELR, ESI and EWR indices (Fig. 6. (4)) fall in between those of the two fossil fuel scenarios. This pattern was accompanied by significantly lower BIR, <0.65 times that of the two fossil fuel scenarios (Fig. 6 (4)). The complex emissions and the biotic endpoint impacts of S4 were significantly less than that of the other three scenarios (Fig. 6. (3) and (4)). Thus, in the spillover/transportation subsystem, the energy transition from fossil fuel to wind power-based green ammonia for hydrogen FCVs is an ideal choice for decreasing the complex emissions, the endpoint impacts and the sustainability of the spillover system. Our results confirmed the opinion that a higher energy density means a

Fig. 6. Emergy inputs, emissions, impacts and sustainability of the spillover subsystems of the four telecoupling scenarios. (1) Emergy inputs to the spillover subsystems of the four telecoupling scenarios. (2) Emergy emissions from the spillover subsystems of the four telecoupling scenarios. (3) Emergy-based biotic endpoint impacts of the spillover subsystems of the four telecoupling scenarios. (4) Emergy indices of the spillover subsystems of the four telecoupling scenarios (dimensionless).

lower transportation cost ratio [84]. Thousands of tons of liquid ammonia are transported by ships, rail and road tankers in many parts of the world through existing transporting facilities, with increasing attention being is paid to security strategies to prevent leakage [85]. Thus, there are now existing facilities and improving security strategies for ammonia transport.

3.3. LCA-EME based multi-nodal nexus evaluation

Our results confirmed that complex "water-energy-material-emission-impact nexus" (WEMEIN) thinking is needed to guarantee the sustainability of the telecoupled energy transition [61]. This multi-nodal nexus can capture greater reality than two or three-nodal nexus studies, but it also means the results will be more complicated and harder to integrate [69]. From the aspect of bio-physical donor values, emergy evaluation is a valuable tool in handling the complicated multi-nodal nexus problems of telecoupling activities as illustrated by WEMEIN problems, because it can synthesize inputs, emissions and impacts that are generally quantified in different units in a single measure. Tracing WEMEIN flows through Energy Systems Language network diagrams (Fig. 1) is helpful in recognizing resource depletion hotspots and potential environmental impacts [86]. The currently developing emergy and LCI databases and LCA software (*e.g.* NEAD, Ecoinvent, ELCD, GaBi, CLCD, ReCiPe *etc.*) dramatically improve the accuracy of the coupled LCA-EME on both the input-side compiling resource depletions and the output-side quantifying emissions and impacts, which provided a much more realistic detection of flows for structuring sustainable resource management strategies [39,40], *e.g.*, the biotic endpoint impacts and the accompanying social and economic impacts of resource stress and land occupation in different nations and regions can be quantified [56].

The Unit Emergy Value (UEV), defined as the amount of emergy required to make one unit of a given product or service is a fundamental conversion factor needed for all emergy syntheses [26]. Taking advantage of the superior LCI databases to improve the accuracy of the UEV calculations is one of the important issues in LCA-EME studies [41]. Besides being a basic conversion factor, the UEV is also a fundamental emergy indicator, inversely related to the efficiency of the production system or process [87]. A product's UEV does not have to be consistent with that of its EYR, which is an indicator of the system's ability to use 'free' local resources and the competitiveness of those resources in the economic market. In this study, the total emergy input to each of the scenarios is equal to the UEV for 1 km EV or hydrogen FCV passenger transport in Japan, which was set as the uniform output from all the four scenarios. Combined with a detailed analysis of inputs, emissions, and impacts, the UEV discloses an essential characteristic of the four systems and processes under study, which cannot be discovered from the three most commonly reported emergy indices, the EYR, ELR and ESI alone.

Another long-term important issue in the field of emergy studies, is "How to account for the emergy of emissions/wastes and bring them into the framework of the sustainability evaluation?". So far, the most widely accepted and applied method is to calculate the additional ecosystem services (*e.g.*, from wind and water) required for diluting emissions as an indicator of the environmental impacts [88]. This method is similar to the biogeochemical cycle methods applied in this study [50], thus, no social or economic impacts were considered in this approach. To fill this gap, measures of the integrated environmental and social economic impacts were also estimated, as effects from the receiving perspective, specifically focused on the impacts on human health and biodiversity loss.

The impacts of all four telecoupling scenarios in the DALY indicated human health (HH) losses were over 3.38 times the ecosystem quality (EQ) loss indicated by the PDF. One of the main hidden reasons for this result is that the regeneration of human contributions to the system are associated with social and economic factors, in addition to the biogeochemical input that is the sole cost for other species. It was found that, up to the 1980s, the global emergy base of the world had increased to

5.58 times that of the renewable emergy inflows of the Earth [50]. The higher the socioeconomic development and the higher socioeconomic welfare per person, the higher the DALY impacts. Thus, uneven socioeconomic development is another factor that could affect the HH impacts. In this study, in 2016, the emergy flow per capita in Australia (1.71E+17 sej/capita/yr) was 1.94 times that in Japan (6.02E+16 sej/capita/yr). In the two fossil fuel scenarios, over 57.50% DALY impacts and over 71.90% PDF impacts were located in the receiving subsystem (Japan, Fig. 3. (1) and (2)); while for the two green ammonia scenarios, over 67.26% DALY impacts and over 41.83% PDF impacts happened in the sending subsystem (Australia, Fig. 3. (3) and (4)). As a result, the emergy-based DALY impacts were 3.38 and 3.72 times the PDF impacts for the two fossil fuel scenarios, while they were 6.26 and 5.63 times the PDF impacts for the two green ammonia scenarios (Fig. 2. (3)). It was found that in 2008, the emergy flows per capita in Australia and Japan were around 4.3 and 1.5 times that of the median value for 99 national economies (3.71E+16 sej/yr/cap) [89]. From this point of view, importing wind power-based green ammonia as an energy feedstock for NEVs from underdeveloped regions that have abundant wind energy resources, existing green power facilities, and cheap labor might be a better option for nations and regions lacking renewable energy resources.

Many scientists think that the ecosystem services needed for emission/waste load dilution and buffering should be counted as a further cost in the total emergy budget of a process or system [39,90]. However, the extra emergy needed from the ecosystem is not actually available, which is why emissions lead to impacts. In addition, emissions do not come from nothing, but occur because inputs are not being fully utilized, which means, taking the emergy of emissions/wastes as an extra input causes a double counting problem. From a different perspective, the endpoint impacts could be taken as an extra cost without bringing in double counting problems. However, it was found that this consideration did not bring a different result in this study from that found by the classical EYR, ELR and ESI analysis (Supplementary Fig. 4), with the endpoint impacts in only HH and EQ being quantified.

3.4. Scenario and data uncertainties

The sources of uncertainty in LCA and EME studies are generally classified into two categories, *i.e.*, scenario/modeling and parameter/data uncertainties [43]. For scenario/modeling, certain complex aspects of the system, such as the transportation distances of ammonia in Australia and Japan were not counted in this study, considering that the terrestrial transportation distances were much shorter than the international shipping distance, *i.e.* (8700 km from Sydney to Tokyo).

One of the other main missing parts of this study is the labor cost of the economic analyses, which are of broad interest and have been evaluated by many publications. For example, it was found that both wind power hydrogen [91] and ammonia-hydrogen [5] are cost competitive in niche applications. In addition, the price of wind power is expected to further decline 37–49% by 2050 [92], while the cost of wind power-based hydrogen production is expected to decrease to around 1 USD/kg hydrogen by 2050 [93]. All these results explain why so many economies at different scales from nations to companies are now invested in demonstrating or developing new technologies or business cases for ammonia-based energy [1]. This is also why we believe it is an urgent affair to clarify the complex environmental impacts of these activities booming behind this attractive economic blueprint.

Considering the parametric uncertainty of LCIs, the hotspots of uncertainty were mainly located at the coal mining process in Australia (the sending subsystem of S1), and the FE and ME emissions (fresh water and marine eutrophication) for all four telecoupling scenarios. As a result, the propagated variations of the total FE and ME emissions indicated by their variance as a percent of the standard deviation, SD% (Supplementary Table 4), for all four scenarios were higher than 18% and 6%, respectively. Besides, the propagated variations of the emergy

indices of the sending subsystem (S1) were all higher than 7% (Supplementary Table 4). Fortunately, the relatively high LCI data variations of FE and ME emissions did not disturb the exploration of the significant one-way AONVA results among the four scenarios (Fig. 3. (2)). Similarly, significant one-way ANOVA results were found for most of the inputs, emissions, impacts and indices between the subsystems of S1 and the other three Scenarios too. No significant differences were found for the input of both renewable and other minerals used in the sending subsystems of S1 and S2, although the average differences were 75% and 28%, respectively (Fig. 5. (1)). The reason behind these results is the relatively higher variations of the LCI data for the sending subsystems of S1 and S2, which were over 24% and 12%, respectively (Supplementary Table 4). Thus, further LCA study is needed for the coal and NG mining systems in Australia, considering that coal and natural gas (NG) are still the second and third largest fractions in global primary energy consumption, and Australia is the largest global producer and exporter of coal and NG [13].

4. Conclusion

Complex water-energy-material-emission-impact nexus (WEMEIN) thinking is needed to guarantee the sustainability of energy transitions. Replacing fossil fuel (coal and NG) with wind power-based green ammonia as the energy feedstock for EV and hydrogen FCV can bring a significant reduction in fossil fuel depletion (>29.64%) and GHG emissions (>10.00%) accompanied with increased energy efficiency. However, as a tradeoff, the total emergy demands of the two green ammonia scenarios were over 2.03 times that of the two fossil fuel scenarios, accompanied by an over 1.56 times increase in the biotic endpoint impacts.

The unevenly distributed multi-nodal nexus benefits/impacts of telecoupled energy transitions need to be quantified and shared among all involved subsystems, especially the sending and receiving subsystems. Changing the telecoupled energy between Australia and Japan from fossil fuel to wind power-based green ammonia can bring a significant reduction in ME and GHG emissions and in the biotic endpoint impacts in the receiving subsystem, besides a significant decrease of GHG emissions in the spillover subsystem, but it causes, respectively, an over 8.00 and 3.62 times increase in the total emergy cost and biotic endpoint impacts in the sending subsystem.

The integrated LCA-EME method was confirmed as a valuable tool under development for handling the complex WEMEIN problems. This method provides a mechanism to synthesize, compare and recognize the hotspot nodes of resource depletions, emissions and impacts occurring within the nexus, and to quantify the exploitation and utilization efficiencies, environmental loadings and sustainability of energy transitions at different scales, not limited to ammonia/hydrogen issues alone.

Credit author contribution statement

Hongfang Lu and Bin-Le Lin designed the study and collected the data. Hongfang Lu developed the integrated LCA-EME method, analyzed the results and wrote the manuscript with contributions from all co-authors.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2022.112884.

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