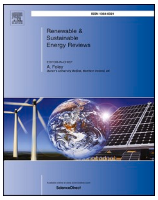
Renewable and Sustainable Energy Reviews 149 (2021) 111417

Contents lists available at ScienceDirect

Renewable and Sustainable Energy Reviews

journal homepage: www.elsevier.com/locate/rser



Can crop residues provide fuel for future transport? Limited global residue bioethanol potentials and large associated land, water and carbon footprints

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ARTICLE INFO

Keywords:
Bioethanol
Biofuel
Bioenergy
Advanced biofuel
Land footprint
Water footprint
Carbon footprint
Water-energy-food nexus
Bio-based economy

ABSTRACT

Bioethanol production from non-crop based lignocellulosic material has reached the commercial scale and is advocated as a possible solution to decarbonize the transport sector. This study evaluates how much presently used transport related fossil fuels can be replaced with lignocellulosic bioethanol using crop residues, calculates greenhouse gas emission savings, and determines lignocellulosic bioethanol's land, water, and carbon footprints. We estimate global bioethanol production potential from 123 crop residues in 192 countries and 20 territories under different environmental constraints (optimistic and realistic sustainable potentials) versus no constraints (theoretical potential) on residue availability. Previous studies on global bioethanol production potential from lignocellulosic material focused on one or few biomass feedstocks, and excluded (un)constrained residue availability scenarios. Our results suggest the global net lignocellulosic bioethanol output ranges from 7.1 to 34.0 EJ per annum replacing between 7% and 31% of oil products for transport yielding relative emission savings of 338 megatonne (Mt; 70%) to 1836 Mt (79%). Emission savings range from 4% to 23% of total transport emissions in the realistic sustainable versus theoretical potential. Land, water and carbon footprints of net bioethanol vary between potentials, countries/territories, and feedstocks, but overall exceed footprints of conventional bioethanol. Averaged footprints range between 0.14 and 0.24 m^2 land per megajoule (MJ^{-1}), 74–120 L water MJ^{-1}, and 28–44 g CO_2 equivalent MJ^{-1}, with smaller footprints in the theoretical potential caused by the exclusion of secondary residues and low price of alternative biomass chains in the sustainable potential.

1. Introduction

The climate as we know it is changing linked to the cumulative emission of greenhouse gases (GHGs) from human activities [1]. Energy-related fugitive emissions from fuels and fuel combustion are responsible for about 74% of global anthropogenic GHG emissions (in 2017) [2]. Energy-related global CO2 emissions reached around 33 gigatonnes in 2019 that was 10 gigatonnes more than in 2000 [3]. Therefore, climate policies prioritize actions to reduce energy related emissions with transition to renewable energy, defined as any form of energy obtained from biological, solar or geophysical sources (e.g., hydropower, wind, waves and tides, geothermal) that is replenishable at a rate exceeding or matching its rate of use [4]. In 2017, the share of renewables was almost 14% of the world's total primary energy supply with bioenergy responsible for 68% of renewables [5].

Bioenergy is energy derived from various biomass feedstocks [4]. Biomass can be a biological material generated from recently living or living organisms [6]. Compared to the variable renewable sources such as solar and wind technologies, bioenergy is more reliable and easier to dispatch when needed [4,7], because unlike solar and wind energy it can be stored like fossil energy sources. Most of today's bioenergy is in the form of traditional bioenergy, i.e. solid biofuels/charcoal that is used for

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Abbreviations: CF, Carbon Footprint; CH_4, Methane; CO_2eq, CO_2 Equivalent; EJ, Exajoule; EUR, Euro; GHG, Greenhouse Gas; GJ, Gigajoule; GWh, gigawatt-hour; Kcal, kilocalorie; kWh, Kilowatt-Hour; LF, Land Footprint; LHV, lower heating value; MJ, Megajoule; Mt, Mega tonne; Mtoe, mega tonnes of oil equivalent; MW, Megawatt; NPP, Net Primary Production; PV, Photovoltaic; RPR, Residue Production Ratio; RSR, Residue to Surface Ratio; USD, United States Dollar; WF, Water Footprint.

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[1] Deceased November 18, 2019.

https://doi.org/10.1016/j.rser.2021.111417
Received 18 September 2020; Received in revised form 8 June 2021; Accepted 21 June 2021

residential heating and cooking in developing countries [5]. In western countries, bioenergy in the liquid and gaseous form is more valuable as it can substitute, among other things, for conventional transport fuels [8] that are responsible for a quarter of global GHG emissions from fuel combustion [2].

Liquid biofuels are promoted as low carbon alternative fuels that can help decarbonize the transport sector. Liquid biofuels include different generations. First generation or conventional biofuels are produced mainly from food crops [9,10] or their substrates such as sugar, vegetable oils, seeds, starch, grains, etc. [11]. Second generation biofuels are produced from non-food biomass (i.e., lignocellulosic feedstock like bioenergy crops, residues and wastes) [10–12]. Third generation biofuels are produced from algae [13] while fourth generation can be electrofuels and photobiological solar fuels [12]. Often, non-conventional biofuels are simply referred to as advanced biofuels [14,15]. Advanced biofuels production is favored over conventional biofuels because conventional liquid biofuels production entails large water [16,17] and land [18,19] requirements, competes with food production and can affect food prices [20–25]. Advanced liquid biofuels produced from non-edible feedstock can avoid issues associated with conventional biofuels [9,14,24,26], making advanced liquid biofuels produced from residues and wastes an attractive option.

In theory, a diverse range of transport biofuels, from jet fuel to hydrogen is possible [27]. However, the practical advanced biofuel production is currently limited [28–31]. Advanced bioethanol from lignocellulosic feedstock is one of the advanced biofuels that can be currently produced at the commercial scale [31,32]. The forest/agricultural residues are examples of lignocellulosic biomass that are rich in carbohydrates that can be used to produce biofuels [33]. Such residues are currently utilized for other purposes and type of competing uses vary spatially and between different sources of biomass [34].

Many studies estimated the global bioenergy resource potentials in terms of primary energy and came to different results based on assumptions, methodologies, datasets, time-frames, etc. One crucial criterion that to a large extent determines the approach and methodology is the type of bioenergy potential that can be differentiated into theoretical, technical, sustainable, market, etc., based on the nature of restrictions that limits bioenergy availability [35]. For instance, IIASA's Global Energy Assessment estimates the current theoretical global bioenergy potential, that is all of the aboveground net primary production (NPP), and places the estimate at 1126 EJ (EJ) year^{-1} and the practical potential (excluding NPP used for fiber, feed, and food) at 793 EJ year^{-1} based on gross calorific value [6]. Fischer and Schrattenholzer [36] estimated the economic bioenergy potential (based on economic criteria) in 2050 and placed the number at 350–450 EJ. The latter included crop residues, forest products, grassland biomass, animal and municipal wastes. Beringer et al. [37] estimated global bioenergy potential in 2050 under different land availability scenarios and reported the range as 130–270 EJ from all biomass source combinations. Searle and Malins [38] estimated the sustainable bioenergy potential in 2050 (from dedicated energy crops, forestry residues, crop residues and wastes) and placed the number at 60–120 EJ year^{-1} as primary energy. A decreasing availability is reported in these studies going from theoretical to other types of potentials and temporal decline in availability caused by the land use changes [36–38] (e.g., increasing area of cultivated land to meet food demand of a growing population).

Some studies estimated global bioenergy potential in terms of energy carriers such as liquid biofuels. Davis et al. [39] reported the sustainable bioethanol production potential using lignocellulosic biomass from Agave and concluded that 6.1 billion liters of bioethanol can be produced annually. Abbas and Ansumali [40] estimated the theoretical bioethanol potential from rice husk and reported the range of 20.9–24.3 billion liters per annum. However, no study has looked at the global total liquid biofuel/bioethanol theoretical and sustainable potentials per crop residue and across countries. This is important, however, considering that many low-carbon energy scenarios and roadmaps assume that advanced bioenergy, including advanced bioethanol, will play a large role. For instance, the IEA's Bioenergy roadmap estimates the advanced bioenergy demand at 25 EJ by 2060 [41]. Similarly, International Renewable Energy Agency's (IRENA) REMap Scenario assumes that about 22% (~22 EJ) of transport energy consumption comes from liquid biofuels and biogas, with advanced biofuels making up half of all liquid biofuels [42]. To inform the energy transition discourse and transition planning, it is relevant then to evaluate the total theoretical liquid biofuel potential, the sustainable potential, and to determine the environmental footprint family (land, water, and carbon) of liquid biofuel per crop residue and across countries.

The environmental footprint family is an umbrella term that encompasses different footprint concepts [43–46]. In this study, by environmental footprint family we consider the land, water and carbon footprints. Environmental footprints serve as indicators of the pressure of an activity or a product on the environment and help to understand impacts of this pressure [46]. A single footprint indicator focuses on a particular environmental concern [46], but a footprint family composed of two or more individual footprints can help assess broader environmental impacts by complementing each other [43–45]. Only few studies have assessed a combination of two or more footprints of conventional [18] and advanced biofuels [13], but the footprint assessment of advanced biofuels for theoretical versus sustainable potential, across countries and for different feedstocks is missing.

This study evaluates how much of the transport related fossil fuels can be replaced with lignocellulosic bioethanol using crop residues, calculates the scale of GHG emission savings that this replacement can bring, and determines land and water resources that are required for producing the lignocellulosic bioethanol per crop and country. Specific steps to accomplish this objective are (1) to estimate the global lignocellulosic bioethanol production potential from agricultural residues with and without setting residue availability restrictions; and (2) assess the environmental footprint (i.e., land, water, and carbon) of a residue-based bioethanol per crop, country and estimated potential. The results can help us evaluate and compare the GHG emission factor of lignocellulosic bioethanol for different crop residues and across different countries while also comparing their sustainability from the land and water perspectives.

2. Methods

2.1. Theoretical potential vs optimistic sustainable potential vs realistic sustainable potential

The current study calculates the theoretical, optimistic sustainable, and realistic sustainable bioethanol potentials. The theoretical potential is the least restrictive and assumes that all biomass (from crop residues) is available to be used for bioenergy [4,47,48] while the optimistic sustainable and realistic sustainable potentials have restrictions on the amount of biomass collection considering the technical and environmental implications [35,47]. Technical restrictions relate to the current technical possibilities taking into account competition with existing residue uses (e.g., feed, food, and fibre) [35,47], while the environmental restrictions relate to mitigating adverse impacts on water, soil or biodiversity [35,48].

Theoretical potential assumes complete collection of the primary (aboveground residue) and secondary residue. The *optimistic* sustainable potential assumes the residue availability is limited to 50%, which is consistent with the assumption made by Scarlat et al. [49], Kadam and McMillan [50]. The realistic sustainable potential assumes the residue availability is limited to 25%, which is consistent with the assumption made in the report by the International Energy Agency [51] (and only slightly higher than the 20% sustainable average assumed by Searle and Malins [38]). Residues generated from orchards are assumed non-competitive and the availability is constant across scenarios.

The range of residue availability in the sustainable potential variants is in line with previous studies (Supplemental Information; SI Table A1). Breakdown of proportion of residue remaining in the field due to environmental vs technical considerations are 30% and 20% in the *optimistic* sustainable potential, and 50% and 25% in the *realistic* sustainable potential, respectively.

2.2. Overview of bioethanol production

Production of bioethanol can be divided into the agriculture stage and the conversion stage (Fig. 1). The agriculture stage generates two types of biomass outputs at the site: (1) the main product/economic yield, and (2) primary residue. In addition, the processing of the main product can generate a secondary residue off-site (i.e. food processing, fiber crop byproducts) and for simplicity of presentation, it is added to the agriculture stage. Examples of primary and secondary residues are summarized in Table 1. This study focuses on the primary agricultural residue (generated in the field during harvesting) as well as the secondary residue (generated at the processing sites) but excludes the tertiary residue (post-consumer residues) [9] to reduce uncertainty. Specifically, two main reasons for excluding the tertiary residue are: (i) lack of reliable data as the amount of organic waste depends on variables like the consumption pattern, and economic development [52]; and (ii) that many regional and country policies aim to minimize food losses (i.e., tertiary residues) and organic waste is progressively being collected to convert to compost.

The conversion stage is assumed to generate two types of outputs: (1) bioethanol, and (2) electricity. Co-produced electricity is counted as output only if it is in excess of electricity required to satisfy biorefineries own needs. This surplus electricity can be sold to the grid [53] and as such become an economically valued by-product of the biorefinery processes.

Availability of secondary residue for bioethanol is relatively more difficult to assess than the primary residue. Technical, economic and environmental factors [61], such as the difficulty assessing the fraction that has existing uses [9,62–64] (i.e., corncobs are already used to produce chemical materials or rural energy while the fraction of rice husk and bagasse is already used for power generation), and complexity of logistical constraints of transporting them to biorefineries [65] are some of the obvious reasons. Even so, the secondary residues can play an important role because of being concentrated at a specific location, avoid issues with their disposal [62], and having a stable year-round supply [9].

Secondary residue (i.e., bran or hulls from cereals; molasses from sugar crops; bran from pulses; shells from nuts; cake from oilseeds; fruit pulp from fruits) is estimated for crops with reported conversion factors by FAO [66]. All secondary residue is assumed available for bioethanol production in the theoretical bioethanol potential estimation, but unavailable under the sustainable potential restrictions because of competition with existing uses and logistical constraints.

Table 1
Primary and secondary residue examples for different plant categories. Compiled using data from TARA [54], Ryan and Openshaw [55], Koopmans and Koppejan [56], Jólli and Giljum [57], Offermann et al. [58], Zafar [59], and Vassilev et al. [60].

Plant variety	Primary residue	Secondary residue
Field and seed crops (e.g., cereals, sugar crops, roots and tubers, pulses, temporary oil and fiber crops, etc.)	Leaves, pods, stalks, straw, stems, tops	Bran, cob, husk, molasses, bagasse, pomace, shell, peelings, presscake
Vegetable crops (e.g., cabbages, green beans, tomatoes, carrots, etc.)	Foliage, leaves, shoots, stems	Peelings, pods
Orchard and vineyard crops (e.g., fruits, nuts, grapes, gooseberries, etc.)	Pruning, fronds, stems, shoots, leaves, old plants	Fiber, shells, bunches, copra, pulp, peelings, rind, sepal, tops, husks, fruit stems

2.3. Environmental footprint calculations overview

Land, water, and carbon footprints are frequently used environmental footprints and together form the environmental footprint family [44,45] (it can include other footprints [43] which are not included in this study). Land footprint refers to appropriation of land as a resource [46]. Water footprint refers to consumptive freshwater appropriation in the supply chain of a product or an activity and includes three components: a blue component (consumption of surface and groundwater), a green component (consumption of precipitation water), and a grey component (water required to assimilate pollution to accepted water quality standards) [67]. Carbon footprint refers to supply chain GHG emissions of a product [46]. Calculation boundaries for individual footprints assumed in this study are presented in Fig. 2.

Environmental footprints per unit of bioethanol's stored net energy are calculated in five steps. The first step calculates water, energy and carbon (CO_2 equivalent [CO_2eq] emissions) intensity of inputs. The second step calculates gross bioethanol and gross electricity production per crop, and per country. The third step determines the value fraction of bioethanol compared to the outputs of agriculture and conversion stages. The fourth step calculates the net bioethanol output by subtracting energy equivalents embedded in inputs during the agriculture stage from the gross bioethanol's stored energy. The fifth step calculates

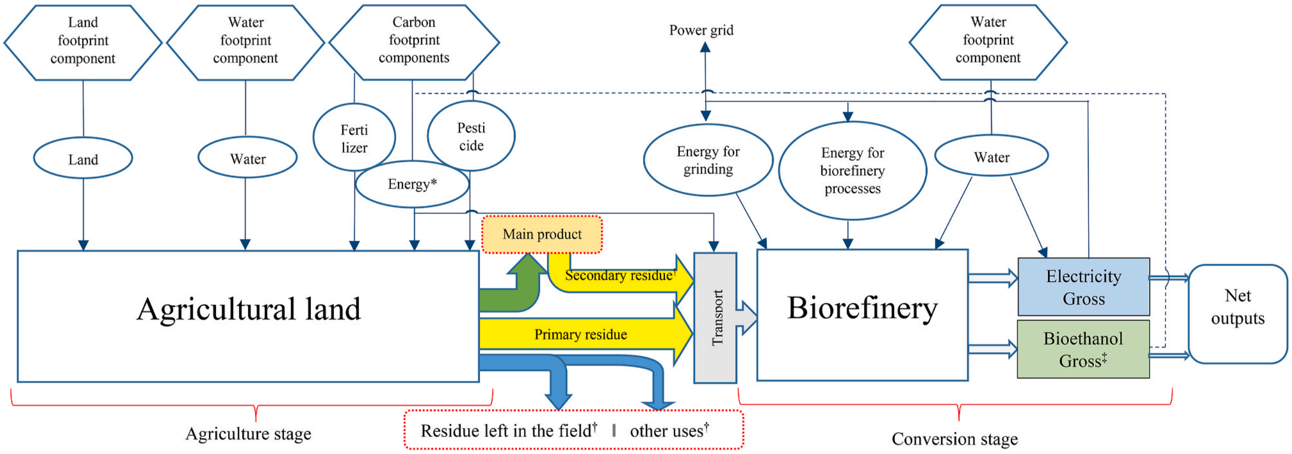

Fig. 1. Schematization of lignocellulosic bioethanol production. Note: * energy component includes diesel and grid electricity use; † availability of secondary residue, residue left in the field and residue allocated for other uses differs between scenarios; ‡ gross to net distinction is shown to clarify energy balance but has no effect on the carbon footprint calculations.

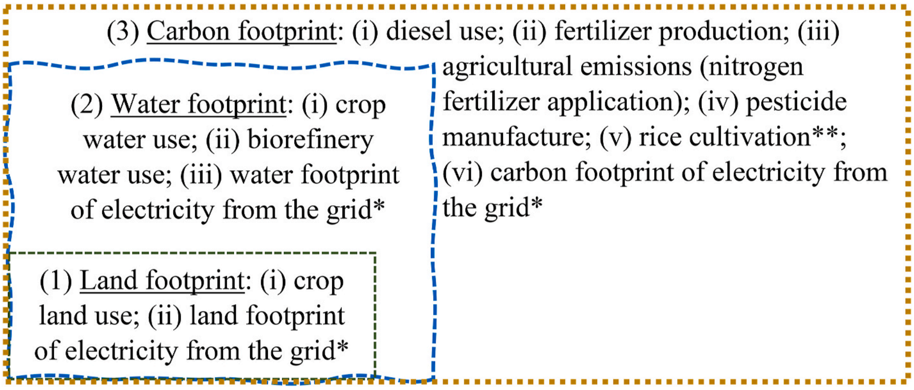

Fig. 2. Calculation boundaries for land, water, and carbon footprint assumed in this study. Note: * refers to indirect footprints linked to electricity from the grid for crops with insufficient co-generated electricity; ** specific to rice.

the land, water, and carbon footprints per bioethanol's stored net energy per crop, and per country.

2.3.1. *Step one - resource inputs*

The environmental footprint family related categories of inputs are summarized in Table A2. The land footprint of bioethanol production is linked to: (1) agricultural land use and (2) land footprint of the power grid. The crop harvested area from FAOSTAT [68] is used as the basis of the agricultural land use. The land footprint of the power grid is calculated by multiplying the land footprint of electricity generated from a particular source (except for the ambiguous "other renewables" category) [69] by its percent contribution to the global electricity generation mix in 2018 [70] and summing the results. The "other renewables" category is assumed as a mix of geothermal, solar thermal, and biomass. The land footprint of "other renewables" is calculated as the average footprint of its composites.

The water footprint of bioethanol is linked to: (1) crop water use; (2) biorefinery's water use; and (3) water footprint of electricity (from the power grid). Ten year average crop water use data is obtained from Mekonnen and Hoekstra [71], and consists of blue, green, and grey components. Water use in biorefinery refers to evaporative losses [53] of blue water. The water footprint of the power grid is calculated by multiplying the median net water footprint of electricity generated from a particular energy source [72] by its percent contribution to the global electricity generation mix in 2018 [70] and summing the results. The water footprint of "other renewables" is calculated as the median of the average water footprints of its composites.

The carbon footprint of bioethanol is linked to (1) diesel use; (2) fertilizer production; (3) agricultural emissions related to synthetic (nitrogen) fertilizer application; (4) pesticide manufacture; (5) rice cultivation (only applicable to rice that emits methane gas during growth); and (6) the power grid. For the carbon footprint calculations, the background processes (e.g., diesel production, tractor manufacture), direct and indirect emissions from land-use change are not considered. Moreover, consistent with the IPCC assumptions [73], the CO_2 emissions from annual biomass combustion are assumed zero as the equivalent amount is sequestered during their growth. Similarly, the primary residues from perennial orchards (pruning, leaves, stems; see Table 1) are assumed to represent short-term carbon cycle and have zero CO_2 emissions. Diesel use for ploughing (21 cm), sowing, fertilizing pesticide spraying, mowing (the residue), bailing and handling (the residue) [74] is converted to the energy equivalent using the lower heating value (LHV) of diesel calculated from IEA and OECD [75] and then to CO_2eq emissions using diesel fuel's emission factor [76]. Diesel is also used to transport residues and consistent with previous studies [48,77–79] a transportation distance is limited to 50 km from field to the biorefinery, where increasing distance can lead to higher transportation costs and GHG emissions [41]. Considering the complexity of interaction between the dry matter, moisture content, truck payload constraints and bulk and solid density [80], two different transportation energy requirements (megajoule [MJ] tonne^{-1} km^{-1}) are assumed for primary vs secondary residues [81]. The moisture content is an important factor that can affect transport, handling and processing, and residue storage [82]. The carbon footprint of transportation is calculated by multiplying available primary and secondary residue fresh weight (calculated in Step 2 below) by their respective energy requirements assuming a distance of 50 km and using diesel's emission factor.

Carbon emissions from fertilizer production are calculated as a product of a five year averaged nutrient nitrogen applied in a specific country [68] and the average CO_2eq emissions (for ammonium nitrate, calcium ammonium nitrate, ammonium nitrosulphate, and calcium nitrate) per weight of a product at the plant gate [83]. Agricultural emissions (direct and indirect nitrous oxide) related to nitrogen fertilizer application are calculated using the tier 1 methodology [84] where the nitrogen input is a five year averaged nutrient nitrogen applied in a specific country [68]. Results are converted to CO_2eq emissions using the 100 year global warming potential of nitrous oxide [1]. The carbon footprint of pesticide manufacturing is calculated as a product of the weighted average pesticide manufacturing energy with the related emission factor from Audsley et al. [85] and the five year average pesticide application rate in a given country [68]. To calculate the emission from rice cultivation, the FAOSTAT [68] implied emission factor of 140 kg methane (CH_4) per hectare (ha^{-1}) is multiplied by the crop harvested area.

Country specific, nationwide grid emission factors for 94 countries are compiled from IGES [86], Koffi et al. [87], and OECD [88]. For countries with missing emission factors, a global average emission factor was calculated across the 94 countries and used instead. Electricity is used for grinding wet residues and for biorefinery processes. Many different interconnected factors influence lignocellulosic biomass recalcitrance that can be structural or chemical in nature [89]. Among them the particle size is a key parameter influencing cellulose hydrolysis potential as it can influence the accessible surface area and accessible pore volume that are important for enzymatic hydrolysis [89]. The study adopted energy required for grinding to the size of 0.5 mm at the biorefinery from Ref. [90]. Electricity use in a biorefinery is linked to the bioethanol production and consistent with a previous report [53] assumed 1.03 kW-hour (kWh) per liter of ethanol.

2.3.2. *Step two – gross bioethanol and gross electricity production*

Gross bioethanol production, *BE* (tonnes), depends on: (a) the amount of residue (dry weight); (b) cellulose and hemicellulose content of a particular residue; (c) cellulose and hemicellulose conversion and recovery efficiencies; (d) ethanol stoichiometric yield per unit of mass; and (d) glucose and xylose fermentation efficiencies as:

$$BE = R \times Carb \times CR_{eff} \times SY_{BE} \times F_{eff} \tag{1}$$

where R is dry residue in tonnes; $Carb$ refers to residue cellulose/hemicellulose content (%); CR_{eff} is the conversion and recovery efficiency of cellulose/hemicellulose from Table 2 (%); SY_{BE} is the stoichiometric ethanol yield per unit of sugar; and F_{eff} is the fermentation efficiency of glucose and xylose sugars (%). Table 2 shows the residue to gross bioethanol conversion calculation parameters from Badger [91].

The key data for calculating the residue availability is the FAOSTAT [68] database covering 160 primary crops (not all of them are included in this study). For 27 crops (e.g., mushrooms, berries, spices, fiber crops), the residue production ratio (RPR) and residue to surface ratio (RSR) necessary to determine the amount of residue per crop were not available and therefore these crops were excluded (SI, Table A3). RPR refers to a ratio of crop residue to crop main produce [61,92] that is used in converting tonnes of crop production to tonnes of residue production (appropriate for field crops that are more homogenous). RSR refers to the ratio of residue to surface area [92] that is used to convert area of production to tonnes of residue production (appropriate for orchards that are more variable). Further 10 crops were excluded from the analysis due to the absence of price data, bringing the final number of crop specific residues utilized in this study to 123, spanning an area across 192 countries and 20 territories (territory here is used in a broad sense to refer to areas that are not fully independent states or are disputed regardless of the legal status, e.g., overseas region, department, collectivity, etc.). The area covered by the crops included in this study envelopes 98.8% of the total primary crop area reported by FAOSTAT, thus the amount of available residue and the global bioethanol production potential calculated in this study covers most of the global potential.

Residue productions are calculated using (i) RSR compiled from literature for orchards (SI, Table A4); (ii) RPR compiled from Fischer et al. [61], Eisentraut [9], and Terrapon-Pfaff [93] for field crops; and (iii) using the FAO [66] conversion factors for the production of secondary residue (SI, Table A3). Specifically, primary residue for field crops is obtained by multiplying the five year average primary crop production data from FAOSTAT [68] by the crop specific RPR. Primary residue for orchards is obtained by multiplying the five year average crop harvested area data from FAOSTAT [68] by their respective RSR values compiled from literature [56,94–112].

Secondary residues are obtained by multiplying the five year average crop production data from FAOSTAT [68] by the FAO [66] conversion factors using the following assumptions. Among different by-products only the by-product of the main production route (i.e., flour production by-product rather than alcohol production by-product of wheat processing) is assumed for each of the crops using the world average number when possible (i.e., wheat bran range is 10–26% while the world average is 18%). Conversion factors specifying secondary residue production is available for 45 of 123 crops utilized in this study.

Moisture content of biomass used at biorefinery can be high [113], but the ethanol yield depends on the dry residue weight. Wet residue is converted to dry weight as:

$$DW_i = HW_i \times Dmc_i \tag{2}$$

where DW_i is the dry weight of residue feedstock for crop i (in tonnes); HW_i is harvested moisture weight of residue feedstock for crop i (in tonnes); and Dmc_i is the dry matter content of the harvested residue feedstock for crop i (%).

Moisture content of primary field residues is obtained from Fischer et al. [61] and Eisentraut [9]. When a moisture content of orchard residue is not given, the fresh matter of orchard residues is assumed 40%, which is consistent with assumptions in previous studies [49,92,96,97,111,114]. For ease of calculations, all orchard residues are normalized to 40% moisture content to calculate energy required for transportation and to 0% moisture content to calculate bioethanol production using equation (2). Fresh matter of secondary residue is assumed 15% with the exception of pulp and molasses. For the latter, reported total soluble sugar yields (%) [115] are converted to gross ethanol assuming 50% cellulose specific calculation parameters and 50% hemicellulose specific calculation parameters. Owing to this general assumption, the ethanol yield from molasses is likely under- or overestimated.

Cellulose, hemicellulose and lignin are the three major components of lignocellulosic biomass [89,116,117]. Among the three components, hemicellulose is the easiest to hydrolyse, followed by cellulose [89,118], and both can be converted to sugars through enzymatic or chemical methods [115,119]. Crop specific compositions of residue feedstock in primary residues is compiled from the Phyllis2 database (25 residues) [120], Reddy and Yang (1 residue) [121], and Hassan et al. [122] (1 residue). For the remaining 96 cases an average biomass composition is utilized (SI, Table A3). Ratios between cellulose, hemicellulose and lignin can vary based on factors such as the harvesting season, culture conditions, and age [122]. The average biomass composition for this study is calculated using data from: Milbrandt and Overend [123], Menon and Rao [20], and Redin et al. [124]. Composition of secondary residue feedstock is compiled from the Phyllis2 database (13 residues) [120], Hassan et al. [122] (5 residues), Zabed et al. [115] (6 residues), and for the remaining 21 cases an average biomass composition is utilized.

Electricity co-generation in a biorefinery is calculated using the heat content of post-processed residue sludge. Specifically, assumed heat of combustions are 7.1 kilocalorie (kcal) g^{-1} (29.7 MJ kg^{-1}) for lignin, 4.2 kcal g^{-1} (17.6 MJ kg^{-1}) for cellulose [125], and 17.0 MJ kg^{-1} for hemicellulose [126]. The lignin content of residue is assumed to remain intact in the post-processed sludge as it can hinder hydrolysis [24,33,89,119] and undesirable for bioethanol production. In addition, considering reported conversion and recovery efficiencies (Table 2), 24% of cellulose and 4% of hemicellulose is assumed to remain in the post-processed residue sludge. Consistent with Humbird et al. [53], thermal conversion efficiency of post-processed residue sludge to steam is assumed at 80% and steam to electricity at 31%, yielding the post-processed sludge to electricity efficiency of 24.8%. Electricity generation is thus calculated as:

$$E_i = ((L_i \times 29.706) + (C_i \times 17.573) + (H_i \times 17)) \times 0.248 \tag{4}$$

where E_i is the electricity output (MJ) from post-processed residue sludge for residue i; L_i is the lignin content (kg) of post-processed residue sludge for residue$i$; C_i is the cellulose content (kg) of post-processed residue sludge for residue$i$; and H_i is the hemicellulose content (kg) of post-processed residue sludge for residue$i$.

2.3.3. Step three - value fraction calculations

The value fraction of bioethanol compared to the total outputs is calculated differently for theoretical vs sustainable potential variants and for different energy inputs (energy used directly for ethanol production vs energy for general agricultural uses). Specifically, the agriculture stage outputs in the theoretical potential are: crop, bioethanol,

Table 2

Residue to gross bioethanol conversion calculation parameters. Source: Badger [91].

Carbohydrate	Content	Conversion and recovery efficiency	Glucose fermentation efficiency	Xylose fermentation efficiency	Ethanol stoichiometric yield
Cellulose	Varies (Table SI A3)	0.76	0.75		0.51
Hemicellulose		0.96		0.5	

and electricity (if there is net electricity output, see step four below). The agriculture stage outputs in the two sustainable potential variants are: crop, bioethanol, electricity (if there is net electricity output), and the alternative biomass delivery chain (e.g., fodder, bedding, construction, mushroom industry, power station) - for non-orchards/vineyard crop residues (orchard and vineyard residues are assumed non-competitive [41]). When energy is used directly for ethanol production the outputs are bioethanol and electricity while when energy is used for general agricultural activities the outputs are the same as the agriculture stage outputs. The conversion stage outputs are: bioethanol and electricity (if there is net electricity output). The general formula for the value fraction calculation is:

$$f_v(BE) = \frac{price(BE) \times w(BE)}{\sum_{p=1}^{3}((price(p) \times w(p))} \tag{5}$$

where the value fraction of bioethanol is $f_v(BE)$, $price(BE)$ is the price of bioethanol (United States Dollars [USD] MJ^{-1}), and $w(BE)$ is the quantity of gross bioethanol (MJ). Denominator is the total value of output products calculated as price times quantity. Five year average crop prices in USD per unit of mass are obtained from FAOSTAT [68]. When prices are available only in standard local currency (SLC), they are converted to USD using the exchange rate from FAOSTAT [68]. In the absence of a country specific crop price a global average crop price is calculated and utilized. The price of electricity in USD for individual countries for 2018 is obtained from the World Bank [127], and when the price for a particular country is not reported a global average price is substituted. The CBOT ethanol price is calculated as average of closing prices in 2018 from Nasdaq [128]. The market price for the alternative biomass delivery chain is uncertain and depends on many factors (i.e., type of residue [129]; costs of collection and transport [130]; etc.). Some recent studies such as the FAO & EBRD study [131] had used the residue collection price as the primary indicator of the residue price. In this study the price of the alternative biomass delivery chain (price for the actual resource paid to farmers) is assumed 6 USD fresh tonne^{-1}, which is in line with 6 euros tonne^{-1} paid in Europe [130] and 5 USD ton^{-1} paid in the USA [132] (US ton is 0.91 metric tonne^{-1}). Temporal variation in straw prices is assumed irrelevant as a Danish study projected less than a 10% increase between 2012 and 2022 under three different scenarios [78].

2.3.4. *Step four – net bioethanol calculation*

Gross bioethanol output per harvested area is converted to net bioethanol output by subtracting the energy embedded in inputs. Specifically, energy embedded in the fertilizer production, pesticide production, energy used directly for ethanol production, energy used for general agricultural activities for residue i in a given country (step 1 above) is multiplied by the value fraction of bioethanol from step three and subtracted from gross bioethanol output for residue i in a given country calculated in step two.

Net electricity output (or net required input from the grid) is calculated by subtracting (i) electricity use in biorefineries and (ii) electricity use for residue grinding from (iii) the gross electricity output (step 2). Specifically, electricity use in biorefineries is calculated by multiplying gross bioethanol output for residue i in a given country by electricity used per volume of gross ethanol (step 1). Electricity use for residue grinding is calculated by multiplying the amount of residue (fresh weight) for residue i (step 2) by 40 kWh tonne^{-1}. The LHV of ethanol is taken from USDOE [133]. The ethanol density is taken from Humbird et al. [53].

2.3.5. *Step 5 – environmental footprints, land, water and carbon footprints, per unit of net bioethanol*

The land footprint per unit of net bioethanol is calculated as the sum of the land footprint from the agriculture stage and the land footprint of the grid (if electricity is required from the grid). The land footprint of the agriculture stage is calculated as: the value fraction of gross bioethanol (step two) divided by the net bioethanol output per harvested area (step four). The land footprint of electricity from the grid is calculated by dividing the land footprint of net electricity required from the grid by the net ethanol output.

The water footprint per unit of net bioethanol is calculated as the sum of the (i) water input of the agriculture stage multiplied by the value fraction of bioethanol, (ii) water input of the conversion stage multiplied by the value fraction of bioethanol, and (iii) water footprint of the power grid (if required), divided by the net bioethanol output from step four. The value fraction of bioethanol differs between different potentials, crops, countries, and between the agriculture versus the conversion stages (see step three).

The carbon footprint per unit of net bioethanol is calculated in three steps. The first step multiplies the respective emissions by the value fraction of bioethanol. The second step divides each category of emissions by the net bioethanol output. The third step sums the results. Specific categories of emissions are: (i) emissions from using diesel; (ii) emissions from fertilizer production; (iii) agricultural emissions (nitrogen fertilizer application); (iv) emissions from pesticide manufacture; (v) emissions from rice cultivation; and (vi) emissions related to the power grid (if required).

3. Results

3.1. *Global crop residue, bioethanol and bioelectricity production potentials*

Global total annual net bioethanol production potential from the 123 crop residues calculated in this study ranges from 34 EJ under no restrictions for residue collection (theoretical potential) to 7 EJ when the primary residue collection is restricted to a maximum of 25% (realistic sustainable potential; Fig. 3). Depending on the country conditions and the potential-specific assumptions, not all crop residues can produce positive net bioethanol as the energy inputs for production can exceed the energy output and are not counted in this total. The difference between the gross and net bioethanol output is smaller in the theoretical potential, 9%, than in the optimistic sustainable potential, 11%, and the realistic sustainable potential 13%, reduction. The reason for this is that energy inputs for general agricultural activities remain the same between the theoretical potential and the two sustainable potential variants despite declining bioethanol output (linked to reduced primary residue availability and exclusion of secondary residue for bioethanol production in the sustainable potential) requiring a relatively larger share of the gross bioethanol output to be used in the production processes.

In practical terms, the global total net bioethanol produced from 123

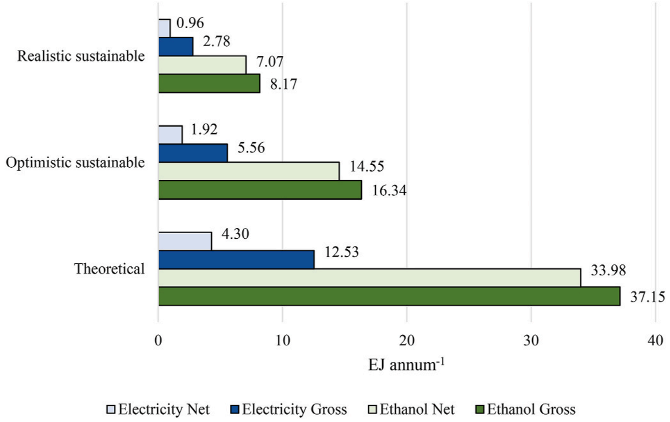

Fig. 3. Total annual global bioethanol and electricity production capacity (EJ annum^{-1}) under three different potential variants.

crop residues can only replace between 7 and 13% (realistic sustainable potential and optimistic sustainable potentials, respectively) to 31% (theoretical potential) of 2589 mega tonnes of oil equivalents (Mtoe) oil products [134] (108 EJ year^{-1}) currently consumed in the world's transport sector (2017 numbers). Bioethanol mix with oil products is common, although using a blend of bioethanol with gasoline exceeding 10% (by volume) can pose infrastructure compatibility issues [27].

At the regional level, only Africa could produce enough net bioethanol (at the theoretical potential) from local crop residues to replace oil products consumed in the transport sector (Fig. 4). However, in terms of optimistic sustainable or realistic sustainable potentials, Africa can replace 54% or 26% of oil products use in Africa's transport sector, respectively. Central & South America have the second largest capacity in terms of producing net bioethanol from local crop residues to replace its transport sector's demand for oil products, from 83% at the theoretical potential to 16% at the realistic sustainable potential. Asia Pacific and Eurasia, are placed in the middle of the regional ranking and can produce net bioethanol that could replace from a high of 46% and 39% (theoretical potential) to a likely range of 9% and 8% (realistic sustainable potential), respectively. Europe and North America have relatively smaller capacity to replace their demand for oil products as they can replace from a high of 23% and 19% (theoretical potential) to a low of 5% and 4% (realistic sustainable potential), respectively. The Middle East has the least potential from a high of 5% (theoretical potential) to a low of 1%.

The net total annual co-generated electricity ranges from a maximum of 4.3 E J (theoretical potential) to 1–1.9 E J in the realistic and optimistic sustainable potential, respectively (Fig. 3). The gross total annual co-generated electricity is much higher, but most of it is required as input in the bioethanol production. The difference between the gross and net electricity output changes little between the theoretical and sustainable potentials, i.e., 66% reduction in the theoretical potential and 65% reduction in the sustainable potential.

Maize and rice paddy residues contribute the most to the bioethanol output under all potentials (Fig. 5). However, soybeans residue is expected to produce more net bioethanol than wheat residue in the theoretical potential while the reverse is true in the two sustainable potential variants. The reason may be that soybean yields relatively larger secondary residues compared to wheat which becomes unavailable under the two sustainable potential variants. Similarly, oil palm fruit residue is one of the top 10 net bioethanol producing feedstocks in the theoretical potential, however, it falls to 25th place in the realistic sustainable potential. The percent contribution of different crop residue categories to the total differs very little between the optimistic sustainable potential and the realistic sustainable potential (not shown).

At a more aggregate level, cereal residues account for half of the total net bioethanol regardless of the potential (Figure A1). Fruit residues account for 19% of net bioethanol in the optimistic sustainable and realistic sustainable potentials and only 16% in the theoretical potential. Oil crops is the next largest contributing crop group that accounts for 19% of net bioethanol in the theoretical potential and 14% in the realistic sustainable potentials. Fibres, stimulants, vegetables, and pulses account for 3% of net bioethanol in each of the potentials. Sugar crops account for 3% of net bioethanol in the theoretical potential and only 2% in the optimistic and realistic sustainable potentials. Root crops account for about 1% of net bioethanol in each of the potentials while crops in the "other" group (i.e., tobacco, natural rubber) and nuts account for less than 0.5% of net bioethanol in each of the potentials.

Countries with the largest net bioethanol production potentials are: China (mainland), the USA, India, and Brazil (Fig. 6). These are countries with large crop production and consequently large residue availability. The relative ranking of the major 8 countries does not change between the different potentials with minor reordering occurring thereafter. The percent contribution of different countries varies little between the realistic sustainable potential and the optimistic sustainable potential, although the ordering of some countries changes (e.g., Côte d'Ivoire and Canada are the top 10th and 11th in the realistic sustainable potential while in the optimistic sustainable potential their rankings switch places).

Countries with the largest bioethanol output are the large producers of maize, rice paddy, soybeans, wheat, and bananas. While the share of maize residue in net bioethanol output is largest in the USA (52–57% theoretical vs realistic sustainable potential) and China (28–29%), rice paddy residue contributes the most in India (31%, no difference between the theoretical and realistic sustainable potentials) and China (25%). Soybeans is the largest contributor in Brazil (45% in theoretical vs 40% in realistic sustainable potential) and the USA (30% in theoretical vs 24% in the sustainable potential). Wheat residue is a large contributor in India (14%) and China (12%), while banana residues are responsible for 12–14% of bioethanol output in India and 8–10% in Brazil in the theoretical vs realistic sustainable potential, respectively.

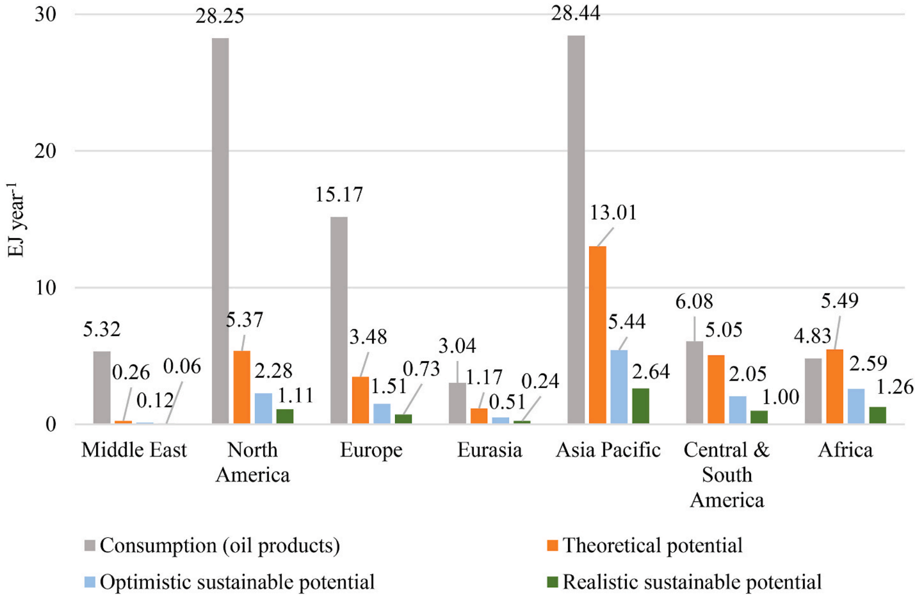

Fig. 4. Transport sector's annual consumption of oil products per world region (year: 2017) and the annual net bioethanol production potential from local feedstock. Regions are presented in the ascending order (from left to right) according to their capacity to replace oil products with the net bioethanol. *Note*: world regions are compiled according to the International Energy Agency's country classifications to match transport sector's oil products consumption data and cover 163 countries (out of 212 countries & territories included in this study).

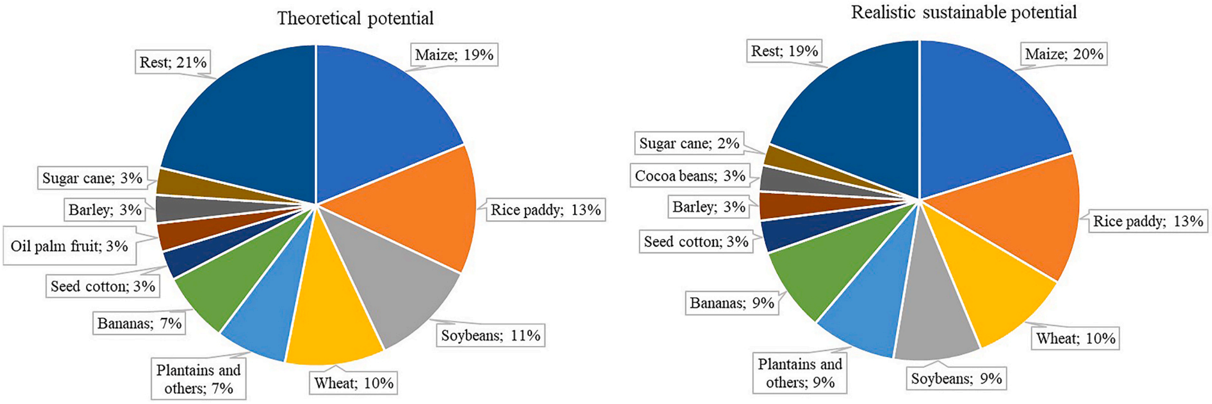

Fig. 5. Contribution of different crop residue categories to the global total annual net bioethanol production potential.

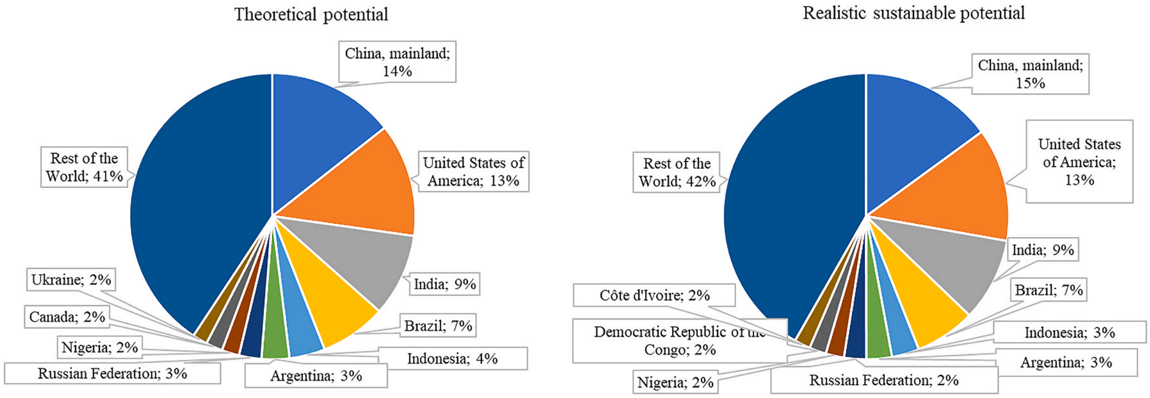

Fig. 6. Contribution of different countries to the total annual global bioethanol production potential.

3.2. *Land, water and carbon footprints of net bioethanol*

Regional average environmental footprints of net bioethanol reveal differences across world regions (Table 3). Asia Pacific has the smallest average land footprint (LF) per unit of net bioethanol production regardless of potentials and the smallest average water footprint (WF) in the optimistic and realistic sustainable potentials. Asia Pacific also has the largest average carbon footprint (CF) regardless of potentials. Eurasia has the largest average land and water footprints. North America has the smallest average CF per unit of net bioethanol in the theoretical potential. Europe has the smallest average water footprint (WF) per unit of net bioethanol production in the theoretical potential.

The global average environmental footprint per unit of net bioethanol production across the main crops is presented in Table 4. For all three footprints (LF, WF and CF), a generally increasing trend can be observed going from theoretical to realistic sustainable potential, caused mainly by: (a) exclusion of secondary residues in the two sustainable potential variants that lowers the volume of bioethanol output per unit of land while crop water uses and the GHG emissions (fertilizer, pesticide, indirect machinery; methane from rice cultivation) stay the same; and (b) low price of alternative biomass chains in the two sustainable potentials compared to the price of bioethanol that increases resources allocations to bioethanol.

The global average environmental footprint of net bioethanol varies greatly between crops. The global average LF of net bioethanol ranges from a low of 0.006 m^2 MJ^{-1} in the theoretical potential (0.007-m^2 MJ^{-1} in optimistic and realistic sustainable potentials) to a high of 1.03 m^2 MJ^{-1} in the theoretical potential (1.81–3.42 m^2 MJ^{-1} in the optimistic and realistic sustainable potentials, respectively). The global average WF of net bioethanol ranges from a low of 4.9 L MJ^{-1} to 609 L MJ^{-1} in the theoretical potential and from a 5.7 L MJ^{-1} to 1559 L MJ^{-1} in the realistic sustainable potential. The global average CF of net

Table 3
Production-weighted average environmental footprints of net bioethanol production across world regions.

Region*	LF (m^2 MJ^{-1})			WF (L MJ^{-1})			CF (g MJ^{-1})		
	Theor.	Optimist.	Realist.	Theor.	Optimist.	Realist.	Theor.	Optimist.	Realist.
Africa	0.12	0.15	0.18	62	80	99	8	10	12
Asia Pacific	0.08	0.10	0.12	53	63	71	21	27	30
Central & South America	0.08	0.11	0.13	52	68	80	8	10	13
Eurasia	0.27	0.36	0.44	105	143	175	13	16	21
Europe	0.11	0.15	0.18	48	63	74	14	17	21
Middle East	0.14	0.17	0.19	82	96	107	14	15	19
North America	0.09	0.12	0.15	50	68	81	8	10	13

Note: * world regions are compiled according to the International Energy Agency's country classifications and cover 163 countries (out of 212 countries & territories included in this study).

Table 4
Global average environmental footprints of net bioethanol production across the main crop residue categories. LF in m^2 MJ^{-1}; WF in L MJ^{-1}; CF in g CO_2eq MJ^{-1}.

	Global average														
	Across all crop residues			Maize residue			Rice paddy residue			Wheat residue			Soybean residue		
	LF	WF	CF	LF	WF	CF	LF	WF	CF	LF	WF	CF	LF	WF	CF
Theoretical	0.14	74.3	28.4	0.09	51.3	8.1	0.07	45.1	34.3	0.17	82.2	11.7	0.10	51.1	7.6
Optimistic sustainable	0.19	96.9	34.4	0.12	65.8	10.3	0.08	55.5	42.7	0.22	105.6	15.4	0.15	73.4	11.2
Realistic sustainable	0.24	119.8	44.4	0.14	77.0	12.6	0.09	61.2	48.1	0.26	123.4	19.6	0.17	85.3	13.9

bioethanol ranges from a low of 1.6–2.0 g CO_2eq MJ^{-1} (theoretical vs realistic sustainable potential, respectively) to a high of 597 g CO_2eq MJ^{-1} in the theoretical potential and 1146 g CO_2eq MJ^{-1} in the realistic sustainable potential.

Tradeoffs between different footprints becomes clearer when compared at the more aggregate crop groups level (Table 5). Specifically, the smallest global average LF per unit of net bioethanol can be obtained by using residue from “fruits” in the theoretical potential. Using residue from “fruits” also leads to the smallest CF per unit of net bioethanol in the optimistic and realistic sustainable potentials. However, the smallest global average CF per unit of net bioethanol in the theoretical potential can be obtained by using residue from stimulants. In contrast, using residue of root crops leads to the largest CF per unit of net bioethanol regardless of the potential. Using residue from the “other” group leads to the largest WF per unit of net bioethanol regardless of potentials and using residue from vegetables leads to the smallest WF regardless of potential.

Total CF of the annual global net bioethanol production from the 123 crop residues calculated in this study ranges from 474.4 Mt in the theoretical potential to 250.7–143.3 Mt in the optimistic sustainable potential and realistic sustainable potential, respectively. Replacing oil products consumed in the transport sector (assuming the reported carbon intensity of road transport of 68.0 g CO_2 MJ^{-1}) [134], with lignocellulosic bioethanol can lead to the relative CO_2 emission savings in the range of 70–79% (Table 6). CF is composed of many components that makes generalizations difficult, but even so, when electricity is required from the grid (often for crop residues with high moisture content that do not co-generate much electricity, i.e., roots, vegetables; Table 5), the CF of bioethanol increases. In some instance, the CF of bioethanol exceeds the carbon intensity of road transport. Removing countries with an average CF of greater than 68.0 g CO_2 MJ^{-1} has small effect on the relative emission savings, where only in case of realistic sustainable potential, exclusion of countries with large CF leads to slight increase of relative emission savings from 70% to 71%.

4. Discussion

For the first time, this study estimates the bioethanol production potential from 123 crop residues (primary and secondary) across 192 countries and 20 territories and assuming different potentials that are linked to different constraints on crop residue availability.

4.1. Overview of lignocellulosic bioethanol potential

The annual global total lignocellulosic bioethanol yield in terms of net energy calculated in this study, ranges from a maximum of 34 EJ to a likely range of 14.6 to 7.1 E J per annum. Even at the lowest end of this range, the output energy exceeds current (i.e., 2018) global total liquid biofuel production of 5729 GW-hour (GWh, approximately 0.02 EJ) [135] by orders of magnitude. Net co-generated electricity yield of 1.0–4.3 E J means adding renewable electricity equaling 50–200% of the 2018’s electricity supply from solar photovoltaics (PV; 1.98 E J) [135]. However, despite such an impressive potential stored in crop residues, there are number of drawbacks for them to have an expected impact with the five main impediments being: (a) output energy can replace only a fraction of that consumed in the transport sector, far less than the scale of biofuel use envisioned by prominent energy scenarios and with even smaller GHG emission reductions; (b) time is of essence where every year counts for climate action, but the current gap between actual production and production at the scales calculated in this paper remains gargantuan; (c) the environmental footprint family of solar PV and solar PV derived hydrogen is smaller than the environmental footprint of net bioethanol production; (d) a lack of policy coherence among different sectors (e.g., agriculture, transportation, energy) can inhibit that the land, water and carbon footprints of lignocellulosic bioethanol remain small in a long run; and (e) stable feedstock supply can be difficult to maintain with growing demand for biomass resources supporting bio-based economies. The subsequent sections address these five impediments.

(a) Lignocellulosic bioethanol – non-viable drain?

We found that energy stored in the net lignocellulosic bioethanol produced from 123 crops can replace between 7 and 31% of energy consumed in the transport sector from oil products and reduce transport sector’s emissions from a maximum of 23% to a likely range of 4–9%. In some countries, the transport sector’s annual consumption of oil is so large that the global total bioethanol produced from 123 crops around the globe is not likely (i.e., optimistic sustainable potential and realistic sustainable potential outputs) to be sufficient to satisfy their individual

Table 5
Global average environmental footprint of net bioethanol production per crop groups.

Crop groups	LF (m^2 MJ^{-1})			WF (L MJ^{-1})			CF (g CO_2eq MJ^{-1})		
	Theor.	Optimist.	Realist.	Theor.	Optimist.	Realist.	Theor.	Optimist.	Realist.
Cereal	0.13	0.17	0.19	62.6	81.5	95.5	16.6	21.0	25.1
Fibres	0.07	0.09	0.10	50.8	60.8	68.0	7.1	8.8	10.6
Fruits	0.02	0.02	0.03	17.5	25.1	34.1	4.6	3.1	3.4
Nuts	0.14	0.18	0.20	120.7	157.2	173.0	6.3	8.7	10.7
Oil crops	0.11	0.16	0.19	62.1	81.9	93.9	7.5	11.7	14.7
Others	0.13	0.14	0.15	165.3	179.9	194.7	9.4	10.9	13.7
Pulses	0.23	0.29	0.34	77.8	99.6	117.7	13.6	18.6	24.9
Roots	0.06	0.06	0.07	45.9	48.8	53.2	133.1	142.8	161.3
Stimulants	0.08	0.12	0.16	86.0	124.0	163.4	4.0	5.1	6.5
Sugar crops	0.06	0.07	0.07	66.1	78.0	84.6	23.1	26.8	28.9
Vegetables	0.02	0.02	0.02	15.3	15.8	16.4	37.5	39.4	43.4

Table 6
Comparing total and relative emission savings in the transport sector by replacing oil products with the lignocellulosic bioethanol potential calculated in this study.

Potential	Emissions (Mt CO_2)			Emission savings (%)	
	Net bioethanol production	Corresponding emissions from oil products*	Transport sector's total emissions with replacement (actual 2017 emissions – 8040 Mt CO_2)	Relative emission savings	Total emission savings in the transport sector
Theoretical	474.4	2310.7	6203.7	79%	23%
Optimistic sustainable	250.7	989.5	7301.1	75%	9%
Realistic sustainable	143.3	481	7702.3	70%	4%

Note: * the current global average carbon intensity of road transport is assumed 68 g CO_2 MJ^{-1} [134].

country-scale or regional demands. For instance, in none of the global regions, the net bioethanol production from local feedstock in the optimistic or realistic sustainable potentials is insufficient to replace dependence on oil products. Not only that, even using the global total annual bioethanol production capacity under optimistic sustainable potential (16.3 EJ as gross and 14.6 EJ as net) and realistic sustainable potential (8.2 EJ as gross and 7.1 E as net) is not enough to satisfy the United States of America (23.8 EJ) or the People's Republic of China's (21.5 EJ) [134] consumption of oil products in the transport sector.

Replacing oil products with net bioethanol results in small emission savings to have a large effect on climate goals. Assuming that the carbon intensity of whole transport sector is the same as that of road transport and using the total available net bioethanol to replace equivalent amounts of energy used in the transport sector with crop/country specific CF, reduces total sector's emissions (i.e., 8040 Mt of CO_2 in 2017) [134] by 23% (theoretical potential) to 4–9% (sustainable realistic and optimistic variants, respectively). Realistically, such reduction can offset the share of global transport emissions that is growing, nearly 1.9% annually since 2000 (up to 2019) [136], but has little effect on drastically decarbonizing the transport sector to meet the climate goals without demotorization and expansion of electric [137] and hydrogen based vehicles [138].

Biofuel targets envisioned in some prominent energy scenarios are not likely to be met without a large contribution of conventional biofuels. For instance, IEA's 2DS envisions some 30% (30 EJ) coming from biofuels by 2060 [41] while IRENA's REMap Scenario assumes 22% (22 EJ) of transport energy coming from biofuels by 2050 [42]. Findings of this study suggest that the likely (sustainable) net bioethanol production potential will be optimistically around 15 EJ (but realistically around 7.1 E J) that is far less than the assumed biofuel values and meeting such targets would involve, to a large degree, continued dependence on conventional biofuels or a shift to electricity or hydrogen from sun or wind. An additional reason is that despite having a broad spectrum of possible advanced biofuel technologies such as Fatty Acid Methyl Ester (FAME), Hydrotreated Vegetable Oils (HVO), synthetic fuels via gasification, pyrolysis oils and others (that may be even cheaper to produce than lignocellulosic bioethanol) [41] such technologies nevertheless mostly utilize and hence compete for the same feedstock (i.e., agricultural residues). Moreover, output of some advanced biofuel technologies is not necessarily compatible with existing infrastructure (e.g., pyrolysis oils) [27,139], and often regarded as conventional biofuel technology rather than advanced (e.g., FAME using palm oil as feedstock) [139].

Present day biofuel production is mostly conventional. In 2017, biofuels accounted for 92% of renewable energy use in the transport sector [31]. While lignocellulosic bioethanol production was virtually non-existent, conventional biofuel production reached 143 billion liters in 2017 [31]. IEA estimates [31] that the advanced biofuels production will reach 1.4 billion (0.9 Mtoe or 0.038 E J) by 2023 with cellulosic bioethanol making up 60% or 0.54 Mtoe (0.023 E J). In other words, the scale of advanced biofuel production in 2023 will reach 1% compared to the scale of conventional biofuels produced in 2017. Although some 70 small-scale advanced biofuel plants are dispersed around the world, they add relatively little to the production compared to the commercial-scale plants [31]. An estimated number of large-scale advanced lignocellulosic bioethanol plants in 2018 is reported between 5 (being constructed) [31] and 12 (being operational) [139]. This falls far too short of the scale required to reach the production volumes calculated in this study.

(b) Mind the gap?

The speed of making the energy transition to reach the climate goals is a key element for consideration [140]. The advanced biofuels demand of 25 EJ by 2060 estimated by the IEA's Bioenergy Roadmap requires building about 4300 large scale plants (200 MW capacity operating 8000 h $year^{-1}$) [41]. Similarly, the advanced biofuel demand of 16 EJ to meet 18% of transport sector's final energy consumption by 2050 requires building 80–100 refineries annually costing about 20 billion USD per year [139]. However, investment trends in advanced biofuels production are declining since 2011, and did not even reach one billion USD in 2017 or 2018 [139]. Most of the announced and under-construction advanced biofuel plants are located in developed countries such as the USA (27%), in European countries (32%), that are responsible for some 60% of global share while India (18%), and Canada (5%) are the only other major sovereign players with the rest of the world responsible for less than 20% [31].

As with all innovations, the biofuel technology cannot spread simultaneously and the diffusion would cause temporal and spatial differences [141]. However, the spatial and temporal difference in the speed of scaling up the technology to reach the level of production potential calculated in this study may come too late to support the climate goals. Without efficient measures to reduce our GHGs by 2030, the global warming is likely to reach 1.5 C° that increases climate-related risks for human and natural systems [137].

While the early adopters of advanced biofuel technology are likely to be developed countries with sufficient investment funds, concentrating investments in developed countries alone would not yield the necessary result. Currently, the final energy consumption in non-OECD transport sector is growing faster than in OECD countries (i.e., 5.7% vs 2.4% from 2015 to 2017) [134] and is expected to increase by 77% from 2018 to 2050 while the OECD energy consumption in the transport sector during that period declines by 1% [142]. Given the much higher price tag of advanced bioethanol (29–44 euro [EUR] per gigajoule [GJ^{-1}]) compared to fossil fuel (8–14 EUR GJ^{-1}) or even conventional bioethanol prices (which varies from 13 to 15 EUR GJ^{-1} in the USA to 18–29 EUR GJ^{-1} in the Netherlands) [41] developing countries are not likely to make the transition to using lignocellulosic bioethanol a priority. Moreover, many countries are not on track to meeting various sustainable development goals [143] and tackling numerous societal challenges simultaneously with limited funds would likely prevent countries from overspending on advanced biofuels even when the price of a tradeoff is safety of the planet and humanity. Filling the gap between almost non-existent lignocellulosic bioethanol production of now to at least 7.1 E J per annum would optimistically take decades to fill.

(c) Mind the environmental footprint: lignocellulosic bioethanol vs solar PV electricity and hydrogen

In comparison with the environmental footprint of solar PV electricity and solar PV derived hydrogen, the environmental footprint family of lignocellulosic bioethanol is larger. Certainly, the lignocellulosic bioethanol's footprint calculated in this study is based on the value of bioethanol over the price of all co-products (i.e., crops, co-produced electricity, and the alternative biomass supply chain) and is thus sensitive to price fluctuations. To better understand the influence of price fluctuations on the value fraction calculations and on the results of this study, we conducted a sensitivity analysis by considering four alternatives with different price changes: (i) bioethanol price increased by 25% and the price of all co-products decreased by 25%, (ii) bioethanol price decreased by 25% and the price of all co-products increased by 25%, (iii) bioethanol price increased by 25% and the price of all co-products remains unchanged, and (iv) bioethanol price decreased by 25% and the price of all co-products remains unchanged.

In alternatives one and two the numerator and the denominator of the value fraction change in opposite directions, which has a greater effect on the results than changing only the price of bioethanol in the numerator in alternatives three and four (Tables A5-A8). Logically, increasing price of bioethanol with or without decreasing prices of other products used in value fraction calculations (i.e., alternatives one and three) results in a decrease in net bioethanol output but an increase in land, water and carbon footprints, irrespective of the type of potential. The explanation being that the increase in price of bioethanol increases the value fraction of bioethanol and hence larger shares of land, water, and carbon inputs are attributed to the bioethanol, increasing footprints. Also, an increase in the value fraction of bioethanol entails that a larger share of energy inputs during the production are attributed to the bioethanol, which decreases the net bioethanol output. With the same logic, when the price of bioethanol decrease with or without decreasing prices of other products used in value fraction calculations (i.e., alternatives two and four), the net bioethanol output increases and the footprints decrease.

Changing prices in the four alternatives affects the environmental footprints and the net bioethanol output differently across the potentials and crop groups. Even so, the extent of changes in footprints for the same crop group within the same potential are generally similar. For example, alternative one increases the LF of bioethanol from the residues of cereals by 43%, 57%, or 65% (for the theoretical, optimistic, and realistic potentials respectively). Similarly, the WF of bioethanol from cereal residues increases by 44%, 58% or 66% (for the three potentials, respectively), while the CF increases by 46%, 57%, or 61% (Table A5).

Although, the results of this study are sensitive to changes in the price of bioethanol, electricity, crops and alternative biomass supply chains, alternative allocation principles not based on economic values also introduce challenges. For example, one alternative was to use a physical allocation based on mass. However, even the physical allocation faces challenges, such as: (i) electricity, a co-product of biorefinery cannot be measured in mass units; and (ii) data on the actual moisture content of residue becomes crucial (e.g., moisture content of the same residue for feed vs for combustion would be different) but is difficult if not impossible to obtain.

In comparative terms, the environmental footprint of lignocellulosic bioethanol is larger than the environmental footprint of conventional bioethanol or solar PV electricity or solar PV derived hydrogen. For instance, the global average LF of lignocellulosic bioethanol (Table 4) is orders of magnitude larger than the LF of solar PV electricity (0.001 m^2 MJ^{-1}) and solar PV derived hydrogen (0.002 m^2 MJ^{-1}) [144]. Similarly, the global average LF of net lignocellulosic bioethanol production across all crops (0.14–0.24 m^2 MJ^{-1}, theoretical vs realistic sustainable potential) is still larger than the LF of conventional bioethanol from sugar beet or sugarcane (0.07 m^2 MJ^{-1}) [18].

The WF comparison between lignocellulosic bioethanol and conventional bioethanol depends on the feedstock but the WF of lignocellulosic bioethanol is larger than the WF of solar PV electricity and solar PV derived hydrogen. The global average WF of conventional bioethanol at 57 L MJ^{-1} (sugar beet feedstock) [18] is smaller than the average WF of lignocellulosic bioethanol, 74–120 L MJ^{-1} across all crops (in theoretical vs realistic sustainable potential).

The average CF of lignocellulosic bioethanol falls below 10 g CO_2eq MJ^{-1} (across all potentials) only when residues from "fruits" or "stimulants" are used as feedstock whereas the carbon footprint of solar PV (harmonized across studies) is approximately 5.6–6.9 g CO_2eq MJ^{-1} [145]. The average CF of conventional bioethanol from sugarcane and sugar beet (14–19 g CO_2eq MJ^{-1}) [18] is also smaller than the average CF of lignocellulosic bioethanol calculated across all crop residues (28–44 CO_2eq MJ^{-1} theoretical vs realistic sustainable potential).

Decisions on which alternative fuels and vehicles to promote in decarbonizing the transport sector should be made in consideration of differences between environmental footprints of alternatives fuels/electricity and local land and water resource availability and scarcity, and account for the water-energy-food nexus.

(d) Walk the line: nexus thinking is vital

Long-term sustainability of advanced bioethanol production can benefit from a coherence between different sectors such as the agriculture, energy and transport sector. As the findings of this study suggest the net bioethanol production is context specific and can be favorable from one resource perspective while simultaneously being unfavorable from another, depending on the crop and region. The most striking example of tradeoffs being in the Asia Pacific region, where the land and water footprints per unit of net bioethanol output are the smallest but the average carbon footprint is the largest compared with the other world's regions (in the sustainable potentials, Table 3). Interactions between different resource sectors or the nexus [146] thinking can help to reduce land, water and carbon footprints of lignocellulosic bioethanol because the tradeoff or synergy between different footprints, can be managed. For instance, expansion of green electricity can help lower the CF of the grid and reduce the CF of net bioethanol for feedstocks that require additional electricity input from the grid.

In another example, farmers will not grow relevant crops (i.e., high bioethanol yielding residues) unless there is a stable demand while in the absence of relevant crop cultivation in the area, project developers cannot count it as a guaranteed feedstock supply [139]. In such cases, incentivizing farmers to grow specific crops (agriculture domain) before the demand is firmly established can help meet the transport and energy targets. In a similar way, agricultural policies can help maintain seasonal price variations to a minimum by ensuring sufficient supply chain's capacity to dry and store the residue feedstock [139]. Price of feedstock has a large effect on the price of lignocellulosic bioethanol and thus site selection to ensure low-cost supply of feedstock is important [41]. Price of feedstock accounts for 70–90% of total production cost for conventional vs 35–50% for lignocellulosic bioethanol [139].

Nexus approach can also help harmonize policies to avoid cross-sectoral damages. For instance, Brazil is conducting trials of new sugar cane varieties that can yield higher biomass without compromising sugar yield [31]. By focusing on maximizing bioethanol yield per unit of land (sugarcane can be used for conventional and lignocellulosic bioethanol production) to meet transport targets, such approach can damage the agriculture in the long term. Moreover, monoculture raises sustainability concerns [25,139,147], so if crops are to be rotated, then the land, water and carbon footprint of advanced bioethanol will change at least on yearly basis. In a flip side, existing or future undesirable agricultural policies such as subsidies can mask the real cost of biofuels in terms of resource uses and even lead to depletion of scarce local resources. Water is commonly subsidized despite being a scarce and energy intensive resource [143] (e.g., in parts of Uzbekistan, water is lifted to crop fields located at heights of up to 200 m by means of pumps

[148]).

(e) Too much but not enough

Biomass has many existing and emerging uses that makes the future availability of residue for biofuel production uncertain. Increasingly, biomass is used to produce monomers and polymers to replace petroleum based resources [149,150]. Under a business as usual scenario, the biomass use for chemicals and plastics is expected to grow several folds by 2050 [151]. When coupled with the increase in demand for traditional biomass uses such as for animal feed and traditional bioenergy, the total biomass demand is expected to grow 50% by 2050 under business as usual scenario [151]. As climate change is likely to affect food security and water supply [137], the crop cultivation pattern may change suddenly causing shortage of residue for bioethanol production. Some studies [152] suggest that among the competing demands for biomass, biofuel production has the least priority and if faced with a choice of residue for fuel vs residue for feed and products, the price of feedstock may be not the only determining factor. In the US, most (if not all) of the advanced biofuel plants failed because of putting a plug on state or federal support [139]. In the absence of state's support and with uncertain future of residue supply, making an investment in new technology calls for a thorough techno-economic assessment considering all risks.

In the future, transportation will use a mixture of fuels and regions will differ in their mixtures. Some regions have better options for PV, for example, and might produce hydrogen for trade and others have better options for lignocellulosic bioethanol, because of their agricultural potentials. The global lignocellulosic bioethanol production potential is limited and some studies suggested that even the fossil fuels (e.g., crude oil, natural gas) cannot meet the increasing future transport energy demand [153]. Bridging the increasing gap between transport energy demand and supply requires diversification of renewable transport fuels, like a rapid growth of electric and hydrogen vehicles and associated solar PV and renewable hydrogen production systems.

5. Conclusion

This paper estimates the theoretical and two variants of sustainable lignocellulosic bioethanol production potentials using 123 crop residues in 192 countries and 20 territories. Land, water and carbon footprints per unit of net bioethanol output are calculated per crop residue and country/territory. The energy output is differentiated between gross and net by subtracting energy inputs into bioethanol production.

The results combined with the current state of advanced bioethanol production and supply of biomass leads to two major conclusions. First, lignocellulosic bioethanol production can replace only a small share of energy from oil products consumed in the transport sector. Lignocellulosic bioethanol production based on sustainable utilization of crop residues can replace only a small share of transport sector's demand for oil products – 7–13%. Even the theoretical utilization of all crop residues can replace less than one third of transport sector's demand for oil products – 31%. At the regional scale, replacing oil products used in the transport sector with net bioethanol produced from local feedstock is not possible anywhere when considering the environmental constraints.

Second, the environmental footprint of advanced bioethanol varies based on crops, countries and the potentials, but overall exceeds the footprints of conventional bioethanol, solar PV electricity, and solar PV derived hydrogen. In terms of crop groups, "fruits" generally lead to smallest carbon footprint per unit of net bioethanol in the sustainable potentials while "vegetables" lead to smallest land and water footprints. It is more difficult to generalize a crop group with the largest footprints. That is because there are LF, WF and CF tradeoffs among different crop groups. In terms of country groups, Asia Pacific has the smallest LF and WF but also the largest CF. North America has the smallest CF in the theoretical potential, while Africa has the smallest CF in the optimistic and realistic sustainable potentials. Finally, the footprints become larger moving from theoretical to optimistic sustainable and realistic sustainable potentials: sustainability of agricultural management does go at the expense of environmental resource efficiency of bioethanol production from residues.

The gap between current bioethanol production capacity and the required capacity to match the level of production calculated in this study is large and may require decades to fill while the future supply of residue and the environmental footprint of future lignocellulosic bioethanol production remains uncertain. Even when the theoretical production capacity can be reached, if we rely on current modes of transport with internal combustion engines and feed them as much as possible with crop residue based lignocellulosic bioethanol we will remain heavily dependent on fossil fuels. Therefore reducing the motorization rate and shifts in modes of transport, e.g. towards a fleet of electric or hydrogen based vehicles, are necessary to decarbonize the transport sector.

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.

Acknowledgements

Authors B.H., J.F.S., and M.S.K, gratefully acknowledge funding by the European Union's Horizon 2020 research and innovation programme under Grant Agreement No. 689669 (MAGIC).

This work reflects the author's views only; the funding agencies are not responsible for any use that may be made of the information it contains.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2021.111417.

Author contribution

Bunyod Holmatov: Conceptualization, Methodology, Formal analysis, Writing - original draft, Review & Editing, Visualization; Joep F. Schyns: Methodology, Software, Writing - review & editing; Maarten S. Krol: Conceptualization, Methodology, Writing - review & editing, Supervision; Winnie P. Gerbens-Leenes: Methodology, Writing - review & editing; Arjen Y. Hoekstra: Conceptualization, Methodology, Supervision.

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