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Biofuels, Electrofuels, Electric or Hydrogen?: A review of current and emerging Sustainable Aviation Systems

Pimchanok Su-ungkavatin[1*], Ligia Tiruta-Barna[1], Lorie Hamelin[1]

[1] Toulouse Biotechnology Institute (TBI), INSA, INRAE UMR792, and CNRS UMR5504, Federal University of Toulouse, 135 Avenue de Rangueil, F-31077, Toulouse, France

* Corresponding author e-mail address: su-ungka@insa-toulouse.fr

Abstract

Climate neutrality is becoming a core long-term competitiveness asset within the aviation industry, as demonstrated by the several innovations and targets set within that sector, prior to and especially after the COVID-19 crisis. Ambitious timelines are set, involving important investment decisions to be taken in a 5-years horizon time. Here, we provide an in-depth review of alternative technologies for sustainable aviation revealed to date, which we classified into four main categories, namely i) biofuels, ii) electrofuels, iii) electric (battery-based), and iv) hydrogen aviation. Nine biofuel and nine electrofuel pathways were reviewed, for which we supply the detailed process flow picturing all input, output, and co-products generated. The market uptake and use of these co-products was also investigated, along with the overall international regulations and targets for future aviation. As most of the inventoried pathways require hydrogen, we further reviewed six existing and emerging carbon-free hydrogen production technologies. Our review also details the five key battery technologies available (lithium-ion, advanced lithium-ion, solid-state battery, lithium-sulfur, lithium-air) for aviation. A semi-quantitative ranking covering environmental-, economic-, and technological performance indicators has been established to guide the selection of promising routes. The possible configuration schemes for electric propulsion systems are documented and classified as: i) battery-based, ii) fuel cell-based and iii) turboelectric configurations. Our review studied these four categories of sustainable aviation systems as modular technologies, yet these still have to be used in a hybridized fashion with conventional fossil-based kerosene. This is among others due to an aromatics content below the standardized requirements for biofuels and electrofuels, to a too low energy storage capacity in the case of batteries, or a sub-optimal gas turbine engine in the case of cryogenic hydrogen. Yet, we found that the latter was the only available option, based on the current and emerging technologies reviewed, for long-range aviation completely decoupled of fossil-based hydrocarbon fuels. The various challenges and opportunities associated with all these technologies are summarized in this study.

Keywords: Batteries; Environmental performance; Kerosene; Regulatory frameworks; Sustainable Aviation Fuels (SAF); Fuel cells

1. Introduction

With ca. 920 Mt carbon dioxide (CO_2) emissions per year (year 2019; [1]), the global aviation industry (commercial, private and military) represented approximately 2.5% of all-human induced CO_2 emission (37 Gt in 2019; [2]) and accounts for ca. 12% of emissions from all transport sources [3]. Despite growing public concerns on the contribution of the flying industry to climate change [4–7], the aviation traffic was, prior to the COVID-19 outbreak, forecasted to grow 4.3% per annum and the number of passengers to increase by as much as 20 trillion revenue passenger kilometers (RPK) by 2038 [8,9], compensating most gains obtained from years of fuel consumption efficiency improvements (the global amount of fuel burned per passenger dropped by 23% between 2005 and 2017, from 4.4 L per 100 km to 3.4 L per 100 km; [10]). The aviation sector itself acknowledges the need of finding sustainable alternatives to fossil fuels. For instance, the International Civil Aviation Organization (ICAO) launched the CORSIA initiative (Carbon Offsetting and Reduction Scheme for International Aviation) in 2016, with the vision to achieve CO_2 emission mitigation for the aviation sector corresponding to a carbon (C) neutral growth by the year 2020 (CNG2020 target). It thereby became the first economic sector to adopt a global, universal, and binding system to control its GHG emissions. In addition, the International Air Transport Association (IATA) and the Air Transport Action Group (ATAG) have set CO_2 emission reduction goals of 50% for 2050 (relative to 2005 levels).

The mitigation of the climate impact of aviation is thus the main driver of the present work. Here, we focus on commercial aviation (passengers and cargo), as this represents 88% of the CO_2 emissions from global aviation ([11]; military, private, and other flights, e.g. emergency, represent the remaining 12%). Commercial aviation relies essentially on airplanes covered by the European Certification Specification 25 (CS-25) [12] and excludes air taxis [13]. From here onwards, the term "aviation" will thus specifically refer to commercial aviation. Reducing CO_2 (and overall GHG) from aviation may be achieved either by a decreased demand (itself spurred by e.g., shifts in transportation habits), or by technological means (e.g., enhancing fuel consumption efficiency, developing alternative energy sourcing).This study focuses on the latter strategy, i.e. the potential of alternative energy sources to enhance the environmental performance of aviation in comparison to flying on fossil-based kerosene. Four large families of emerging alternative energy sources for fueling future aviation can be highlighted: biofuels, electrofuels (liquid hydrocarbons produced through an electricity input), electricity (battery-based), and hydrogen. These all involve several variants and are here referred to as "sustainable aviation" (SA) systems, though the term sustainability remains to be assessed as later detailed.One commonly used denomination is "sustainable aviation fuels" (SAF), but we refrained from using it for two reasons. First, this term is typically used to represent biofuels (e.g. [14–17]), although the recent ReFuelEU aviation initiative and its regulation proposal have explicitly expanded it to electrofuels (also designated as synthetic fuels) [18–20]. Second, some of the emerging technologies (e.g., batteries) do not fit within the concept of fuel, hence we here propose the more inclusive "sustainable aviation" terminology instead.

In the literature, several terminologies are used to designate aviation fuels, such as jet fuels or kerosene. In industry, it is referred to as Jet A or Jet A-1 (among all differences between these, the most notable one relates to their freezing point; Jet A -40°C (233K), Jet A-1 47°C (226K); [21,22]). Through this study, the term kerosene (C_8-C_{16} hydrocarbons) is used to refer to liquid aviation fuels in general, whether these are fossil-based or not. Fossil-free aviation fuels deriving from bio-based feedstock (denoted as bio-kerosene or biofuels; the latter term is used herein) are drawing great interest in achieving GHG emission reduction targets for the aviation industry [23]. Today, biofuels generated from different technology pathways must be certified by the American Society for Testing and Materials International (ASTM) (ASTM D7566 – Standard Specification for Aviation Turbine Fuel Containing Synthesized Hydrocarbons) or equivalent standards (e.g., Standard 90-091 of United Kingdom's Ministry of Defense; [24]) before it can be used in commercial aircraft. So far, there are only six certified pathways for biofuels, namely the Fischer-Tropsch (FT) process, hydroprocessed esters and fatty acids (HEFA), direct sugar to hydrocarbon (DSHC), alcohol-to-jet (ATJ; with isobutanol or ethanol intermediates), and catalytic hydrothermolysis jet (CHJ). Some of these pathways involve variants (e.g. FT with added aromatics). Here, sugar and syngas fermentation (followed by upgrading of the produced alcohol) are considered separately instead of being aggregated under the umbrella term "ATJ", as often found in the literature (e.g. [16,25,26]). This is done to reflect the drastically different technologies and overall process flow involved in both cases, among others.

Besides biofuels, additional alternative kerosene for fossil-free aviation include electrofuels (sometimes known as synthetic fuels or powerfuels or power-to-liquid). Aviation electrofuels require a source of carbon and hydrogen (H_2) to generate hydrocarbon liquid fuels having properties similar to those of fossil-based kerosene. Hydrogen may be produced via water-splitting technologies including approaches such as water electrolysis [27], thermochemistry [28], or bio-photolysis [29]. Carbon may stem from biomass-free options such as direct capture of carbon dioxide (CO_2) from the atmosphere (typically termed Direct Air Capture, DAC) [30,31]. The carbon source may also arise from biogenic carbon through the use of syngas stemming from biomass gasification. The syngas may also stem from high-temperature co-electrolysis (using water and CO_2 as input, the latter being biomass-derived or not) [32]. Carbon may also stem from CO_2 captured from a point source (industrial process) [33–35]. Liquid fuels are then produced through the FT process [32] or methanol (CH_3OH) synthesis [36,37], although only the former has been certified by ASTM [36,37]. At present, both electrofuels and biofuels can be used as drop-in fuels, i.e. they are, up to a certain blending limit, interchangeable with conventional kerosene, and do not require adaptation of the fuel distribution network or the engine.

So far, there are four main categories of alternative kerosene. They include i) Synthetic paraffinic kerosene (SPK) largely composed of normal- and iso-paraffins (e.g., obtained with the FT, HEFA, SuF, SF pathways), ii) Synthetic aromatic kerosene (SAK), primarily consisting of aromatics designed as blend components (e.g. produced in the APR pathway), iii) Synthetic iso-paraffin (SIP) consisting essentially of farnesane ($C_{15}H_{32}$), and iv) Synthetic kerosene (SK) consisting of a composition similar to fossil-based kerosene (e.g. produced in the CHJ pathway; a new SK fuel from this pathway (CHJ-SK) has just been certified in 2020) [16,38]). SPK is however the predominant type of alternative kerosene generated from the currently certified pathways [26].

These four types of alternative kerosene are reported to provide cleaner combustion, with reductions of soot; [39,40], particulate matter (PM); [41], and sulfur emissions [42], among others because of the lower aromatic content in the fuel (when not zero). On the other hand, a low aromatic content has the disadvantage to cause shrinkage of some elastomers in the engine seal [43,44]. Reflecting this risk, ASTM D7566 requires a minimum aromatics content of 8% by volume, and as a result alternative fuels have been used as blends with conventional kerosene, up to a certain blending limit varying between 10% (DSHC) and 50% (all other certified processes) of the total fuel volume [17,26,43]. For longer-term solutions, state-of-the-art engines (with novel sealing materials) are being developed to be compatible with unblended aviation biofuels and electrofuels, as tested and announced by aircraft and engine original equipment manufacturers (OEM), namely Airbus [45], Boeing [46], Rolls-Royce [47], Deutsche Aircraft [48].

Through this review, electric aviation refers to the use of batteries as energy sourcing to power aircrafts (whether for propulsion, on-board operating systems, or both in a hybridized mode). One challenge this poses is the development of energy-dense batteries as light and compact as possible, which is challenging with today's lithium-ion (Li-ion) battery technology (around 300-400 Wh kg^{-1}; [49]), relative to Jet A-1 (approx. 12,000 Wh kg^{-1}; [21]). Lithium-based batteries (with the introduction of novel electrodes/electrolytes such as advanced lithium-ion batteries (Adv. Li-ion), solid-state lithium-ion (SSB), or lithium-sulfur (Li-S)), are foreseen as the most promising approaches for mass sensitive applications like aviation, owing to their higher specific energy density [50,51]. However, these advanced Li-based batteries are still in the early stages of research, and many challenges need to be tackled before their mass production. Current Li-ion batteries have nevertheless been applied as a sole energy source for small aircraft capable of carrying less than 20 passengers per flight missions (e.g. Eviation's Alice aircraft; [52], Bye Aerospace's eFlyers aircraft; [53], Pipistrel's Velis Electro; [54], Heart Aerospace's ES-19; [55]). For larger aircraft, electric aviation requires to be combined with another energy source such as conventional (or alternative) kerosene, an approach commonly known as hybridization [56].

Additionally, hydrogen (both gaseous and cryogenic forms) [57,58] is currently researched as an alternative option for use in commercial flights. In the perspective this hydrogen is produced through water-splitting approaches, most of the associated environmental impacts are reported to be related to the type of electricity used [59–61]. Because of the low volumetric energy density of H_2, the heavy on-board system infrastructure required to accommodate H_2 weakens its benefits, as in the case of batteries [62,63]. Moreover, additional external infrastructures are required in both cases, including a charging station for the case of batteries, a H_2 refueling station for hydrogen, while redesigned airframes making the plane lighter or improving its aerodynamic performances are optional. Unlike electric aviation, H_2 systems require a refueling time comparable to conventional (and alternative) kerosene [64].

These two approaches (electric and hydrogen) are not associated with CO_2 emissions during the flight. Although the principle of zero tailpipe emissions applies to 100% electric (battery-based) aviation, the pollution occurring during batteries manufacturing (and recycling) is not zero and needs, as for all upstream activities of all other SA approaches, to be addressed as well [65]. Similarly, the emissions generated during the production of H_2 may be important, especially if stemming from conventional steam reforming of natural gas instead of water electrolysis or other carbon-free approaches [61]. Water vapor and nitrogen oxides (NO_x) are tailpipe pollutants from H_2 aviation and are considered as predominant climate forcers [57,66,67]. They are potential precursors, along with soot (emitted with liquid kerosene), for contrail and cirrus cloud formations, depending upon the ambient atmospheric conditions, which in turns affects the amount of heat trapped within the technosphere [63,65,68,69]. The formation of NO_x (also responsible for tropospheric ozone formation) from H_2 combustion is reported to be lower compared to the NO_x emissions from kerosene combustion [57,63].

There is, thus, a variety of technical possibilities towards a more sustainable aviation sector, and each involve a variety of options in terms of feedstock. While some options can act as drop-in fuels, others require additional infrastructure, and all options affect the environment (not only climate) in different ways. It thus remains unclear, in the perspective of long-term investment decisions towards tomorrow's aviation, which option leads to the highest overall environmental performance, and even whether some of the options are truly sustainable and environmentally superior to fossil-based kerosene. Clear requirements defining sustainability for aviation do exist, among others through the sustainability criteria elaborated in CORSIA by ICAO [70] and to some extent in the Recast Renewable Energy Directive (RED II [71]) for biofuels, electrofuels as well as (renewable) hydrogen feedstock, although these are not aligned with one another [25]. It should be noted that the former criteria go well beyond the sole GHG reductions, also including criteria on enhancing water quality and availability, or soil health, for example.

Nevertheless, to quantify sustainability, Life Cycle Assessment (LCA) is acknowledged as a central tool for environmental management and decision support, in the European Union in particular [72]. Because it can address impacts in a full system perspective, LCA has grown to become one of the key tools for assisting the direction of the so-called "green transition". Yet, to perform such analysis, vital and timely for the case of aviation, the very first step is to have the full overview of the process flow of the alternatives under assessment, including the overview of the types of co-products being generated. It is our claim that such overview is not yet in place for all alternative SA options simultaneously, being one possible reason why no LCA comparing all four-alternative exist at the moment. Although reviews have been published to document the production process, and, to some extent, the sustainability aspects of alternatives to fossil kerosene, these reviews focused mainly on only one of the SA categories distinguished herein. Aviation biofuels have been the most widely reviewed. Notable reviews are the ones of [73,74], where the conversion processes are extensively described, with key technological advances and challenges. In addition to that, economic and environmental aspects of biofuel pathways are comprehensively discussed by [75] and [44]. Similarly, an overview of the state-of-the-art implementation of biofuels within the aviation sector was presented by [76]. Although not as extensively reviewed, the main synthesis pathways for a variety of electrofuels were qualitatively discussed in [77], with regards to the required physical and chemical properties of these fuels when used in airplanes. The studies of [78] as well as [79], albeit not focusing on the aviation sector per se, carried out an economic analysis for a variety of electrofuels documenting several technologies and variations in plant size. However, the electrofuels investigated in these studies can be considered only as intermediates for aviation fuel production. The recent work of [80], on the other hand, represents a notable advance where a general overview of both carbon-free and biofuels options is described. Albeit valuable, the study of Bauen [80] remains largely qualitative and does not detail the technical conversion pathways and processes involved, and completely excludes electrofuels. Moreover, all of the above-mentioned studies, except [44,75], completely overlook the co-products generated along the production process. Yet, these do generate market interactions that in turn are part of the overall economic and environmental performance of these new alternatives, and accordingly must be understood.

In the perspective of bridging these gaps, and in the vision of supplying a harmonized comparative background to assess the environmental consequences of current and emerging options for fueling the aviation sector, the present study presents a comprehensive review intending to supply a holistic understanding in the field of sustainable aviation . Concretely, this review intends to gather and discuss information and data relevant and necessary for comparative environmental assessments (in particular LCA) of emerging candidate options for sustainable aviation.

The review is organized into thirteen sections. The underlying methodological approach used for the review is first described (section 2). The review starts with detailing the biofuels, electrofuels, electric (battery-based) and hydrogen families and options within each (sections 3, 4, 6, and 7, respectively) as well as the co-products obtained in the liquid fuel pathways (biofuel and electrofuel families) and their potential uses as alternatives to fossil-based products (section 5). A semi-quantitative ranking is proposed (section 8), covering environmental, economic, and technological performance indicators, in order to provide a preliminary screening of the environmental performance potential associated with each approach in the near- (2030) and long-term (2050) timescales. In addition to the four dominant SA approaches that are the object of this study, other marginal alternative energy sourcing are reported in section 9.

The electric propulsion systems (i.e., those involving electric motors; either battery- or fuel-based) are discussed in section 10. Section 11 discusses the development of regulatory frameworks and legislations related to clean aviation. Finally, sections 12 and 13 address the challenges and prospects for the deployment ofSA, with emphasis on the availability and sustainability of raw materials and the scientific and technological advances needed.

2. Methodological approach

Overall, approximately 600 records, including scientific literature, patents, (company) reports, conference proceedings, and company websites, were extensively screened and reviewed. Efforts were made to ensure that the most recent data from announcements and literature were used. However, in the very process of writing this article, some of these announcements have changed (e.g., the cancellation of the Airbus E-Fan X project in April 2020), and although we strived to keep all the information presented herein up-to-date, it can happen that some will no longer apply following the publication of this article.

The scientific literature search was performed through two key search engines, namely Google Scholar and Web of Sciences, while the Google Search engine was used for screening the grey literature. A variety of generic keywords such as "aviation decarbonization," "alternative aviation fuels," "sustainable aviation regulation" have first been used, while snowballing strategies [81] were applied to capture additional content. A few variations of the abovementioned keywords have also been used to capture additional records. This procedure was repeated for each specific topic addressed in this review.

Moreover, although we strive to explain all key terminology used within the manuscript (e.g., what the notion of electrofuels exactly encompasses), a glossary is provided (SM2) to clarify all terms and acronyms used. Throughout the manuscript, we report the units as supplied in the original references but provide the conversion in units of the International System of Units (SI).

3. Aviation biofuels

Because of their maturity and compatibility with the existing fueling infrastructure, biofuels is the most dominant SA family.

Three large biofuel categories are here distinguished according to the feedstock used: carbohydrate-rich, lignocellulosic and oil-based biomass. These will be used within this section to organize the description of each biofuel.

Fig. 1 presents, in relation with these three categories, an overview of nine different biofuels pathways for which known investments in terms of capacity have been made.

Among the nine pathways covered in Fig. 1, six are already certified by the ASTM D7566. The remaining three are either under the process of certification (APR, IH^2; integrated hydropyrolysis and hydroconversion) or under development (HDCJ pathway). Some pathways are well developed, documented, and applied in large-scale production (pilot/demonstration plants), while some, such as the CHJ and IH^2 pathways, are less extensively documented. These are briefly described herein, and additional insights on, e.g., the biofuels properties, blending ratios, or the leading industrial producers to date are available in supplementary material 1 (SM1) (Tables S1-S2).

Low readiness level emerging pathways are disregarded from this section because they are scarcely documented, including the one described by e.g. [82], where volatile fatty acids (VFAs) from anaerobic digestion of residual biomass are processed for catalytic upgrading to kerosene-ranged paraffins [82]. It also includes the emerging methanol-based pathway to produce kerosene described in e.g. [83].

3.1 Biofuel pathways involving carbohydrate-rich feedstock

This category comprises three pathways (Fig. 1). They greatly rely upon carbohydrate-rich feedstock such as sugarcane, sorghum, maize dextrose, maize, etc., but can also be used with less dense carbohydrate sources such as molasses. Simple mechanical pretreatment for particle size reduction (milling, chopping, grinding, etc.) may be performed for sugar-rich feedstock (e.g., sugarcane, maize, fodder beet). For lignocellulosic-rich feedstock (e.g., straw, woody crops, and/or residues), mechanical pretreatment is followed by delignification. In this step, the lignin fraction is typically separated from cellulose and hemicellulose by alkaline deacetylation, followed by vacuum filtration [75,84]. The remaining lignin may be utilized in various applications, such as the generation of heat and electricity or the production of syngas (through gasification) for liquid fuel production [85,86]. The cellulose and hemicellulose fractions are further processed with enzymatic or acid hydrolysis approaches to recover C_5 and C_6 sugars. As a result, a hydrolysate is produced and further used as input for the pathways described in this section, namely the aqueous phase reforming (APR), DSHC, and SuF pathways.

3.1.1 Aqueous phase reforming (APR)

In this pathway, the generated hydrolysate (Fig. 2) first undergoes a purification and concentration process. In this step, numerous techniques may be applied, for instance, alkaline solvents for ash and residual lignin removal and filtration for insoluble solid residues removal [74,85]. Depending on the initial feedstock characteristics, the sugar-rich hydrolysate produced in the concentration step undergoes an hydrotreating process to convert sugars and organic acids through hydrogenation (forming polyhydric alcohols) and/or hydrogenolysis (forming shorter-chain compounds) [86,87]. Oxygen is then removed from the obtained molecules through two successive steps: the APR followed by a condensation process.

In the APR process, the aqueous phase is reacted in the presence of a catalyst under a large variety of operating conditions. A temperature range of 175–300°C (448-573K) and pressure range of 10-90 bar (1-9 MPa) are necessary [86] when catalysts such as Cu, Re, Ni, Fe, Co, Ru, Pd, Rh, or Pt are used [85]. Examples of specific operating conditions reported in the literature are summarized in SM1. The reactions happening within this process strongly depend upon the specific configuration and typically include dehydrogenation of alcohols, hydrogenation of carbonyls, deoxygenation, hydrogenolysis and cyclization. As a result of this step, the water-soluble oxygenated compounds are converted into a liquid mixture (APR liquid), which is a complex mixture of hydrocarbons and undesired oxygenated hydrocarbons (e.g. alcohols, aldehydes, ketones). These may be separated at this stage or fed directly to the subsequent condensation step [87] as illustrated in Fig. 2. A gas phase rich in H_2, CO_2, CO and light alkanes is also produced (Fig. 2), which can be directly used for heat and power production [84], among other uses. Alternatively, the H_2 produced in the gas stream could be separated and recirculated to the process [84,88], prior to burning the light alkanes (C_1-C_4) for heat and electricity [84,85,88]. The exact mixture of both gas and liquid output is rather difficult to predict given the complex mixture of hydrocarbons and oxygenated hydrocarbons involved [89].

Through condensation reactions, the liquid mixture from the APR process is upgraded to longer-chain hydrocarbons. For example, base condensation is applied for the production of gasoline and kerosene. The liquid condensate may, prior to distillation, undergo hydrotreating according to the desired hydrocarbon fuels output. The resulting products are polyhydric alcohols or shorter-chain compounds depending on the hydrotreating approaches used. Dehydration of alcohols into alkanes and oligomerization (using solid phosphoric acid or zeolite as catalyst) is used for kerosene production [86]. Hydrogenation and/or isomerization might also be required to ensure conformity with the specific market requirements (e.g. the mandatory ASTM D1655 for aviation fuel blends to be used in commercial aviation) [85]. As illustrated in Fig. 2, these processes can be combined in different configurations and with different operating conditions to fit with the desired final product. Finally, the generated products are subsequently condensed and distillated to produce the desired aviation fuels, here as hydrodeoxygenation synthetic aromatic kerosene (HDO-SAK) or hydrodeoxygenation synthetic kerosene (cycloparaffin-rich fuel) (HDO-SK), along with hydrocarbons co-products including naphtha, and diesel [84,86]. Currently, both HDO-SK and HDO-SAK are under consideration for ASTM certification [90].

3.1.2 Direct sugar to hydrocarbon (DSHC)

The DSHC pathway allows to produce aviation biofuels without an alcohol intermediate (Fig. 3). The sugars derived from the biomass feedstock input are converted to $C_{15}H_{24}$ (denoted as isoprenoid farnesene) through fermentation, subsequently hydrogenated to farnesane ($C_{15}H_{32}$), which can be used as aviation fuel [91,92]. The joint venture between Amyris and Total is the main global developer of the DSHC pathway with carbohydrate-rich feedstock such as maize, sugar beet, sugarcane [93]. These companies engineered microorganisms capable of fermenting both C_5 and C_6 sugars [94,95]. Accordingly, lignocellulosic-rich substrates can also be used as a feedstock in this pathway. Additional examples of industrial partnerships developed to produce aviation biofuels via the DSHC pathway are described in SM1.

The fermentation process with the engineered yeasts takes place at operating temperatures of 30-34°C (303-304K) [94]. A liquid/solid centrifugation process separates the yeast cells and fermentation broth. The supernatant, consisting of farnesene oil, farnesene emulsion and fermentation broth, is collected for further purification. Within the purification process, the collected supernatant is heated in the de-emulsification unit (65-70°C; 338-343K) with the addition of surfactant and is transferred to liquid/liquid centrifugation in order to separate the oil and aqueous phases [94]. The distillation stage separates the contaminants into a heavy fraction containing triglyceride, monoglyceride and salts, and a light fraction of the distilled farnesene. Distilled farnesene then undergoes hydrogenation in the presence of catalysts such as Ni, Pd, Ru, Pt, Mo, Zn, etc. [96]. The purification step may be combined with hydroprocessing in downstream operations for achieving high recovery efficiency (97% reported) [94] (not shown in Fig. 3). The farnesane produced can be used as diesel fuel for terrestrial transport or as aviation kerosene, although the former tends to be preferred due to the low blending ratio allowed for the kerosene obtained from this pathway (10% by volume; Table S1). Alternatively, the farnesene can be chemically converted to produce a variety of products including fragrances, flavors, cosmetics, lubricants, etc. The co-products derived from the separation and purification step (Fig. 3) are reported to be used in anaerobic digestion for biogas production [95], where the biogas is subsequently led to a steam methane reforming (SMR) process in order to produce part of the hydrogen needed for the hydrogenation step.

3.1.3 Sugar fermentation with alcohol upgrading (SuF)

This pathway is based upon the fermentation of the hydrolysate derived from carbohydrate-rich biomass (e.g. molasses, sugarcane, sweet sorghum, sugar beets, food waste, inedible fodder maize) or lignocellulosic-based biomass (e.g. cereal or rice straw) to produce alcohols [97–99]. The generated alcohol, typically ethanol or isobutanol [100,101], is then further upgraded to aviation biofuel through a series of four key processes (Fig. 4) described below (fermentation generating an ethanol or isobutanol intermediate, dehydration, oligomerization, hydrogenation). Both alcohol intermediates are certified by ASTM [16]. The solid residue (often referred to as distiller grains) not converted to alcohol is generally dried and sold as a protein-rich ingredient for animal feed [102–104]. This pathway is often aggregated with the SF pathway (3.2.1.1) under the umbrella of "alcohol-to-jet (ATJ)", and the biofuels production with those pathways are referred to as ATJ-SPK.

In the fermentation step of SuF, bacteria, in particular Clostridia and a modified strain of *E. coli*, are commonly used for isobutanol production through the acetone-butanol-ethanol (ABE) fermentation process [105], albeit other microorganisms are also possible in the presence of yeast *S. cerevisiae* [106] or engineered yeast (details in SM1). For ethanol, *S. cerevisiae* yeasts are typically used [107]. Fermentation temperatures from 20°C (293K) to 95°C (368K) are reported, depending upon the microorganisms used [102,106,108,109]. After the fermentation, the broth containing the desired alcohols, microorganisms and other organic compounds is separated into an alcoholic fraction and distiller grains (by e.g. membrane separations, distillation, solid/liquid separation, etc.) [102]. The alcohol obtained from the fermentation stage will typically not be pure but consists of a mixture of C_2-C_6 alcohols including ethanol, propanol, butanol, isobutanol and pentanol [102].

In the case of the isobutanol intermediate pathway, the produced isobutanol is typically converted into isobutenes in the presence of dehydration catalysts such as inorganic strong acids, metal oxides, zeolites, acidic resins, etc. at operating temperatures ranging from 250-350°C (523-623K) [98,110] (dehydration stage; Fig. 4). Isobutene monomers are oligomerized in the presence of acid- or metal-based catalysts, which results in a liquid mixture of longer chains alkenes (C_{10}-C_{16}) and shorter chains (C_4-C_8) ones (oligomerization stage; Fig. 4). Shorter chains alkenes are separated and recycled to the oligomerization unit [98]. The heavier olefin fractions (C_{10}-C_{16}) are then fed to the hydrogenation process (Fig. 4) [111]. The hydrogenated hydrocarbons are distilled into distinct fractions, namely the ATJ-SPKalong with an isooctane co-product [44]. A variation has been proposed where the heavier olefin fractions are, prior to hydrogenation and fractionation, enriched in aromatics (C_8-C_{16}; content of ca. 20 vol.% [38]) in order to get a fuel without blending limits, leading to the production of so-called synthetic kerosene with aromatics [26]. This is promoted by among others Byogy and Swedish Biofuels and is under ASTM certification process [90,112]. However, at the light of the available information [113,114], it remains unclear whether the upstream part of this process involve direct biomass fermentation as described herein, or the fermentation of gasified biomass (i.e. syngas), which is further described in section 3.2.1.1. Additional details on the SuF process are presented in SM1. The process with the ethanol intermediate is similar; oxygen is removed via a catalytic dehydration process, producing ethylene, then turned into linear or non-linear (branched) α-olefins through a catalytic oligomerization process, in turn hydrogenated to produce paraffins (C_9-C_{16} alkanes) [115].

3.2 Pathways handling residual and lignocellulosic biomass feedstock

Some pathways can be considered as specifically targeting residual (often lignocellulosic) biomasses such as primary forestry residues, crop residues, municipal solid waste (MSW), etc., although these can also technically be used in the previously described biofuels pathways if subjected to pre-treatments. These low-value residues have attracted tremendous attention due to their potential to avoid the competition with food production [116], and their potential important bio-physical availability [117–119]. The pathways described herein use thermochemical conversion processes to convert lignocellulosic biomasses into kerosene. The vision is whether to first convert residual biomasses to gas, and then convert this gas into liquid fuels through so-called Gas-to-Liquid (GtL) processes, or to convert the biomass into a bio-oil to be further processed to kerosene.

3.2.1 Biomass-to-gas via gasification: a prerequisite to several biofuels and electrofuels pathways

Gasification and anaerobic digestion are the two most known technologies to convert biomasses into gas. While the former targets biomasses with dry matter content of at least 65%, the latter is typically considered for wet biomasses (dry matter content below 30%) [120]. The exact amount of carbon from the biomass that will convert to gas, as well as the gas composition itself, will heavily depend upon the technology, process conditions and biomass composition. Anaerobic digestion can be expected to convert ca. 60% of the biomass carbon into biogas [121] while gasification is expected to convert at least 75% of the carbon into syngas [122]. The present review will, based on the reviewed pathways, only focus on gasification, as none consider anaerobic digestion (for kerosene production).

Prior to gasification, biomass must be pretreated into fine particles (80-100 mm) by mechanical techniques (including chopping, grinding) in order to enhance the efficiency of moisture removal, which in turns facilitates the biomass conversion to syngas [123–125]. Gasification typically occurs at temperatures of 600-1,000°C (873-1,273K) or even higher with controlled amount of oxidizing agent such as air, steam, oxygen, their mixture or supercritical water [123–125]. Supplying the appropriate amount of oxidizer is an important parameter for a high syngas production and for limiting the amount of by-products generation (e.g. char) [124]. During the gasification process, carbonaceous materials are transformed through several reactions including drying, pyrolysis (PL), combustion and reduction of feedstock to produce syngas, water vapor, tar, and a solid co-product denoted as char [126].

Syngas, based on the experimental data compiled by [122] for fixed bed gasifiers, is a gaseous mixture of H_2 (10-20%), CO (15-23%), CO_2 (8-18%), CH_4 (1-4%) and N_2 (42-60%). Syngas impurities are composed of tar particles, nitrogenous compounds (NH_3, HCN), sulfur compounds (H_2S, COS, CS_2), hydrogen halides (HCl, HF), and trace metals (Na, K) [127]. Hence, syngas needs to be further conditioned to reform tar contaminants, and remove particulate matters and acid gases prior to utilization in downstream gas-to-liquid processes (Figs. 5 and 6). Depending on the gas-to-liquid process to be used, additional steps to adjust for the H_2/CO ratio of the cleaned syngas may be required. While it could be avoided entirely for the SF pathway, the FT typically requires adjusting the H_2/CO ratio to ~2.0 to avoid methane formation. In addition, syngas must be strictly purified from acid gas and ash to avoid catalyst deactivation during the FT step as well as downstream corrosion [124]. Details on gasification and syngas conditioning and cleanup processes are presented in SM1.

3.2.1.1 Gas-to-liquid: Syngas fermentation and alcohol upgrading (SF)

The syngas deriving from waste gases from industrial processes and/or gasified biomass can be converted into various intermediate products (e.g. alcohols, organic acids) through fermentation process (Fig. 5) after cleanup/conditioning as above described. Different microorganisms can be used to obtain a panel of co-products [128]. Typically, the objective is to convert syngas into ethanol, which is subsequently upgraded into kerosene.

Microorganisms such as acetogenic *Clostridium* spp. convert syngas into several intermediates including ethanol, 2,3-butanediol (2,3-BDO), and acetic acid [109,129,130]. Optimum fermentation conditions intensively depend upon the microorganisms used [131], however, pressures higher than ambient pressure are preferred for enhancing the syngas transfer between the gas and the liquid phase [132]. After the fermentation, the broth including co-products and microbial biomass is fractionated through several processes depending on the co-products formed. For example, distillation is typically used for ethanol recovery [130], while simulated moving bed chromatography may be used for 2,3-BDO recovery [133].

The fermentation residue, comprising microbial biomass and other organic fractions (also known as distillation grains), is filtered [134]. The filtered liquid stream, consisting of soluble nutrient mixtures, can be further recycled back to the fermentation reactor while the filtrated insoluble sludge is typically sent to anaerobic digestion. The resulting biogas can be used for internal steam and power generation [130,133].

Ethanol may be upgraded to kerosene through a variety of processes. First, ethanol is converted to ethylene in the presence of catalysts such as γ-alumina (Al_2O_3), transition metal oxides, or zeolites at operating temperatures of 320-500°C (593-773K) [135]. Oligomerization then converts the produced ethylene to linear long-chain olefins at operating temperatures of 100-300°C (373-573K), also in the presence of catalysts [136]. Nickel complexes are commonly used as catalysts for industrial ethylene oligomerization (e.g. used in the Shell Higher Olefin Process; SHOP) [136,137]. Aluminum-based catalysts (tri-ethyl-aluminum) generate, through the Gulf and Ethyl process, linear α-olefins and by-products (alkanes and branched α-olefins) [137].

The longer-chain olefins generated are afterwards hydrogenated into alkanes. Copper, zinc chromite or sulfide are utilized as catalysts for high-pressure hydrogenation with temperature ranges of 150 and 200°C (423 and 473K) and under a pressure of 200-350 bar (20-35 MPa) [137]. Hydrogenated hydrocarbons are further distilled to recover the C_8-C_{16} fraction known as ATJ-SPK. In addition, naphtha and diesel can be obtained as co-products [133].

The SF pathway, when deriving from lignocellulosic biomass, has an overall reported energy efficiency of 57%, while the FT pathway has an overall reported energy efficiency of 45% [138].

The fermentation pathway provides numerous advantages such as operating conditions near the ambient temperature and pressure [139]. However, the main challenge with the SF pathway is the low solubility of syngas in the fermentation medium, which limits the mass transfer to the liquid phase, in turns resulting in the generation of untargeted (or less desired) products [140,141].

3.2.1.2 Gas-to-Liquid: Fischer-Tropsch (FT) process

The FT technology has been extensively used for the production of synthetic liquid fuels and chemicals (e.g. diesel; [21]; kerosene; [142,143]; naphtha; [144]). FT consists of a series of catalytic processes (Fig. 6) which convert purified syngas (mostly composed of H_2 and CO) into liquid fuels. During the FT synthesis, the purified syngas is passed over catalysts in specific process conditions to form a variety of hydrocarbons (ranging from gases to waxes) following three main reactions [145]:

$$2H_2 + CO \rightarrow (CH_2) + H_2O \qquad (1)$$

$$(2n+1)H_2 + nCO \rightarrow C_nH_{2n+2} + nH_2O \qquad (2)$$

$$(2n)H_2 + nCO \rightarrow C_nH_{2n} + nH_2O \qquad (3)$$

The hydrocarbon product composition is strongly influenced by the operating temperature, pressure, syngas composition, and used catalyst [44,147–150]. The FT process can be categorized into high temperature (HTFT) and low temperature (LTFT). Gasoline, solvent oil and olefins as shorter-hydrocarbon compounds can be generated through HTFT process with operating temperatures of 310-340°C (583-613K). LTFT will typically involve operating temperature of 210-260°C (483-533K) and generate an hydrocarbon mixture consisting of ca. 50% solid wax, the remaining consisting of a liquid phase containing aromatics and cycloparaffins [143].The wax can later be processed to produce naphtha, kerosene and diesel, among others [124,150]. It should be noted that the wax is sometimes defined as C_{20} [151], C_{21} [152], C_{22} [153] or C_{23} [143] and heavier fractions. Common catalysts used are transition metals like Fe, Co, Ni and Ru. Fe and Co are widely used catalysts, with distinct selectivity. For example, Fe-based LTFT generates higher olefins concentration as Co-based LTFT, but with lower hydrocarbon conversion and more CO_2 production. Cobalt-based catalyst is often preferred for GtL FT due to its high activity and selectivity to linear paraffins [44,154–156].

There are three fractions generated from the FT process: gaseous, liquid and wax phases. The gaseous phase represents unconverted syngas and contains CO, H_2, CO_2, and potentially N_2. It can either be fractionated for H_2 recovery or be recycled back to the FT unit in order to maximize kerosene yield [154], while a fraction (ca. 10% of the syngas) will end as off-gas (Fig. 6). The liquid hydrocarbons from FT undergo an hydrotreating process including deoxygenation, decarboxylation and decarbonylation in the presence of supported base metal- or supported noble metal- catalysts [142]. Additional hydrogenation may be applied for transforming olefins and residual oxygenated molecules into saturated hydrocarbons. Wax ($C_{20\text{-}23+}$) produced in the FT unit are transformed to smaller molecules by hydrocracking followed by isomerization into branched hydrocarbons. Obtained hydrocarbons with different lengths are distilled to produce naphtha, kerosene, diesel and lubricants [108,143,157].

This technology allows the use of lignin deriving from lignocellulosic biomass as primary feedstock for bio-based kerosene production (SPK). The SPK derived from the FT process, denoted as FT-SPK, consists of a high proportion of n- and iso-paraffins with a maximum of 15% by weight of cycloparaffins resulting in a high cetane number, high specific energy, and high thermal stability. FT-SPK provides a high quality kerosene in the absence of sulfur, nitrogen, and other impurities [43,87]. FT-SPK with the addition of aromatics (denoted as SPK/A) is another FT-SPK variation, certified in 2015 [26]. This variant is here considered within the overall umbrella of FT-SPK.

3.2.2 Hydrotreated depolymerized cellulosic jet (HDCJ)

Residual biomass can be converted into liquid fuels via a pyrolysis (PL) process, denoted as HDCJ-PL, or via hydrothermal liquefaction (HTL) for wet substrates, denoted as HDCJ-HTL (Figs. 1 and 7). Oily feedstock (e.g. waste cooking oil) can also be used directly (i.e. straight to the separation/upgrading process of Fig. 7).

The PL process converts dry biomass into bio-oil, gases, and biochar, the proportion and composition depending on the process conditions and the nature of the feedstock [158]. Biomass feedstock, finely ground (< 5 mm), is dried to achieve less than 10% moisture content by weight, often seen as mandatory for the PL process, especially in the perspective of quality bio-oil production intended for use as a transport fuel [159–161]. Fast PL is conducted at relatively high temperatures (450-550°C; 723-823K) and ambient pressure with short residence time of the gas phase (typically less than 2s) [162]. Fast PL is proposed for maximizing the production of liquid hydrocarbons (bio-oil) while lower temperatures PL mainly produce a solid product known as biochar [163]. In the HTL process, residual biomass reacts with water at temperature ranging from 200-450°C (473-723K) and pressures of 50-280 bar (5-28 MPa) [164] with or without a catalyst. This process is suitable for the conversion of relatively wet biomasses (5-35% dry matter content) into bio-crude oil; drying pretreatment processes are therefore not necessary [162,164–167]. During the HTL process, bio-oil is produced through multiple reactions such as hydrolysis, dehydration, decarboxylation, condensation, cyclisation or, polymerization. The produced bio-oil contains lower oxygen (in oxygenated compounds) and moisture content with higher heating value in comparison to the PL bio-oil [92,168,169]. Moreover, PL bio-oil has higher acidity and weaker thermal stability relative to the HTL bio-oil [169]; therefore it requires more extensive upgrading processes. More details about the HDCJ processes and examples of industrial developments can be retrieved in SM1. During the upgrading process of the bio-oil to kerosene (Fig. 7), distillation, centrifugation and extraction are preliminary processes for the fractionation of the bio-oil into a gaseous phase, a liquid bio-crude oil phase (the main output), an aqueous phase and solid residue (e.g. tar). Subsequently, other upgrading processes are performed, for example, emulsification, catalytic cracking and/or, steam reforming [170]. The liquid bio-crude oil phase is further refined by hydrotreating to increase its alkane composition, by reduction of heteroatom-containing molecules and by hydrogenation of unsaturated molecules. Hydrotreated hydrocarbons are distilled into liquid fuels including naphtha, kerosene and diesel.

3.2.3 Integrated hydropyrolysis and hydroconversion (IH^2)

The integrated hydropyrolysis and hydroconversion (IH^2) pathway is characterized by a series of thermochemical processes, enabling the conversion of lignocellulosic biomass into hydrocarbon fuels, namely gasoline, kerosene, and diesel ranged hydrocarbons [171]. Four key processes are involved: pretreatment, hydropyrolysis, hydroconversion, and steam reforming (also known as hydrogen manufacturing unit) [171] (Fig. 8).

Lignocellulosic biomass is first passed to the pretreatment unit (including size reduction and drying). Pretreated biomass is sent to the catalytic hydropyrolysis, operated at temperatures of 350-450°C (623-723K), where it is reacted with hydrogen at an incoming pressure of 20-35 bar (2-3.5 MPa) in the presence of a catalyst. As a result, it is converted to deoxygenated vapors along with light gases vapors (C_1-C_3 hydrocarbons, and CO), and char [172,173]. This process minimizes the undesirable properties of standard pyrolysis bio-oil, namely a high acid number, low heating value, high oxygen content, low liquid fuels yields, etc., by adding hydrogen to the pyrolysis process [172]. Char is continuously removed by cyclones, followed by a hot filter [172]. Light gases are further converted to hydrogen (to be reused in the process) through a steam reformer [26,174]. Char, on the other hand, is passed to the hydroconversion reactor. It operates at temperatures 340-400°C (613-673K), at essentially the same pressure as the hydropyrolysis, with a catalyst that induces hydrodeoxygenation and hydrogenation to take place [174,175]. Within this unit, deoxygenated vapors are converted to hydrocarbon vapors (of gasoline, diesel, kerosene) through a hydrotreating processes (e.g. hydrodeoxygenation, hydrogenation). They are subsequently recovered through condensation (e.g. via a distillation column) to liquid hydrocarbon fuels [172,175].

Currently, this pathway is going through the ASTM certification process [26,90]. IH^2 could be a blending component owing to its high aromatic content (up to 92 vol.% when from woody biomass; [174]), which is above the maximum limit for aviation kerosene (25 vol.%) [38]. They can be additionally added to SPK (low/lacking aromatics) [174].

3.3 Pathways involving oil feedstock

Oil feedstock may be derived from many sources, for example, non-edible oil crops (e.g. camelina, jatropha, carinata, pongamia, pennycress) or oleochemical wastes (e.g. waste cooking oils; WCO, waste animal fats, greases stemming from municipal waste facilities). Crops are converted to oil through several processes such as, mechanical processes (e.g. chopping, pressing, chipping), chemical extraction with solvent and/or enzymatic methods [176].

3.3.1 Hydroprocessed esters and fatty acids (HEFA)

Triglycerides containing saturated and/or unsaturated fatty acids can be converted to liquid hydrocarbons (naphtha, kerosene, diesel) by hydroprocessing under various conditions [161,177,178], as illustrated in Fig. 9. In the literature, this is typically referred to as hydroprocessed esters and fatty acids (HEFA) when related to terrestrial biofuels production, and as hydroprocessed renewable jet (HRJ) when related to aviation fuels. In term of process pathway, these terminologies are here considered as equivalent.

Waste fats and oils with different degrees of unsaturation are firstly hydrogenated to saturate the double bonds leading to the production of propane along with free fatty acids. The hydrogenated fatty acids are subsequently converted into straight chain hydrocarbons through hydrotreating processes including deoxygenation, decarboxylation and decarbonylation with the formation of H_2O, CO_2 and CO, respectively [73,75,179]. Subsequently, the hydrocarbons produced are transformed by isomerization and hydrocracking reactions. Isomerization converts straight chain hydrocarbons into highly branched alkanes exhibiting a low freezing point, a desired property as a blending component. Long chain hydrocarbons are broken through an hydrocracking process to desirable carbon length compounds (e.g. in the range of C_8 to C_{16} aviation kerosene) [180]. The product is distilled into naphtha, kerosene and diesel [181,182].

Several parameters have an impact on product generation during the HEFA process, for instance, the type of catalyst used, the fatty acids profile of the feedstock used and the operating conditions [181,183,184]. A variety of research and industrial experiments resulting in the production of aviation biofuels with the HEFA pathway under various conditions are summarized in SM1.

The HEFA pathway is considered as a relatively mature technology at commercial scale. However, the availability and the cost of the sustainable oleochemical feedstock, in particular if waste-based (as in e.g., [185,186]) or relying on limited marginal lands (e.g., [187–190]) to grow the feedstock, may become a limiting factor in the perspective of increased biofuels demand in the future. The HEFA pathway with the variant of using third-generation feedstock (e.g. oils stemming from microalgae *Botryococcus braunii*, denoted as Bb oil) has been recently certified (ASTM approved for 10% maximum blend in 2020) [191]. This pathway produces biofuels known as hydroprocessed hydrocarbons HEFA-SPK (HH HEFA-SPK).

High freeze point HEFA-SK (HFP HEFA-SK, also known as HEFA+) is another variation of already certified HEFA, nearing final ASTM approval [90,112]. The production cost is expected to be reduced as involving lower requirements for the upgrading process (e.g. isomerization) relative to HEFA-SPK [26]. This is among others due to the longer length of the molecule chain, varying from C_{15} to C_{18} [26,192,193]

3.3.2 Catalytic hydrothermolysis jet (CHJ)

The CHJ pathway, similarly to the HTL process, is based on high temperature water chemistry (hydrothermal processes), converting oil-based feedstock into a mixture of straight, branched and cyclic hydrocarbons as shown in Fig. 10. The process is composed of four steps including hydrothermal pre-conditioning, catalytic hydrothermolysis (CH), upgrading through hydrotreatment and products fractionation.

During the pre-conditioning stage, the oil-based material is cracked resulting in the production of free fatty acids with the removal of heteroatoms (S, N, metals, etc.) in the presence of steam and catalyst under operating temperatures of 150-300°C (423-573K) and pressures of 5-50 bar (0.5-5 MPa) [194]. These generated fatty acids are converted in a CH unit at elevated temperature (240-450°C; 513-723K) and pressure (15-250 bar; 1.5-25 MPa) [194]. Within the CH unit, numerous reactions take place including cracking, hydrolysis, decarboxylation, isomerization, and cyclization to produce a mixture of paraffin and cyclic hydrocarbons. In a nutshell, the outputs of the CH unit consist of an organic phase and an aqueous phase. The aqueous phase is composed mostly of low molecular weight carboxylic acids (C_2-C_5), glycerol, and some of small polar molecules [194]. They are transformed through decarboxylation and dehydration into alkene products. These intermediates could be upgraded to aviation biofuels through alcohol recovery stage (oligomerization, hydrogenation and distillation) as described above. The organic phase, referred to as CHJ bio-crude, is further decarboxylated, hydrogenated, and finally distilled into several product fractions including naphtha, kerosene (denoted as catalytic hydrothermolysis jet-synthesized kerosene; CHJ-SK) and diesel [195].

Aviation biofuels obtained with this process contain high density aromatics, iso-paraffins and cycloparaffins [194,196]. Additional details on the CHJ pathway and its industrial developments are presented in SM1.

4. Aviation liquid electrofuels

Electrofuels (power-to-liquid) allow, pushed to its extreme, to decouple the production of kerosene from the demand of biomass, relying on water-based hydrogen (H_2), and atmospheric CO_2 [19,197]. Through hydrogen produced from water electrolysis, electrofuels involve the storage of electrical energy within chemical bonds in the form of liquid fuels, these providing more energy density and lower aircraft mass, compared to emerging electric (battery) aviation and hydrogen aviation.

Clear climate benefits have been documented when renewable sources of energy (whether electricity or heat) are employed to produce the required H_2 [186–188; SM2].

4.1 Hydrogen supply

There are several pathways to produce hydrogen with different resources, e.g. from fossil fuel resources through steam reforming and/or partial oxidation, from non-fossil resources through biomass gasification or fermentation, and from water-splitting technologies [201–205]. Today, around one-third of global hydrogen supply is obtained as a by-product from industrial processes, (e.g. chlorine production from electrolysis of brine; [206–208]). Here, the focus is on hydrogen production decoupled from a carbon source, and as the main product driving the production process.

Water is a promising resource generating growing interest for hydrogen production. Several pathways are possible, such as electrochemistry (including water electrolysis and photo electrolysis) [209,210], thermochemistry [211], or biological water-splitting such as bio-photolysis [29,204] as summarized in Fig. 12. The latter, along with photo-electrolysis, have however not been considered any further herein, being still in early development stages.

Water electrolysis, in particular if powered with fluctuating power in excess of demand, has attracted growing attention as a process to generate so-called green hydrogen [203,212]. There are different water electrolysis technologies available such as alkaline electrolysis [213], proton/polymer exchange membrane (PEM) electrolysis [214], or solid oxide electrolysis (SOE) [215,216] (details on these technologies are presented in SM1). However, these technologies remain relatively expensive, and their profitability is bound to low renewable energy prices [58,62]. Alkaline electrolysis, for example, provides two to four times the production cost of the conventional steam reforming of natural gas [62,217,218], as further detailed in SM1.

4.2 Carbon sourcing

Carbon sources include (i) biomass (undergoing gasification), (ii) fossil or biogenic carbon stemming from industrial activities captured at a point source (e.g. biofuel/bioethanol production, waste gases from steel production and cement industry, from biogas upgrading process, mine gases, etc.; here denoted as point source CO_2) and (iii) atmospheric carbon captured by DAC (Fig. 11) [130,197]. As DAC, point source carbon also requires a capture process, as further described in SM1. It should be noted that biomass here also acts as source of H_2.

There are two major technical approaches for DAC. One is based on the absorption of CO_2 using low-toxicity solvents such as water (through a scrubbing process) and alkaline aqueous solutions (NaOH, $Ca(OH)_2$ KOH) with a CO_2 strong affinity. In addition to water and alkaline solutions, amino acid salts, ammonia, polyglycol ether and ionic liquids can also be used as solvent for CO_2 extraction [219]. The solvent-based technology is mature and is already applied in large-scale plant, however, the solvent regeneration is a high energy-consuming process. The second approach utilizes an alkaline carbonate bonded to a mesoporous solid support in which the sorbents can be easily regenerated. The most promising technology is the use of supported amine materials; this solid sorbent-based approach revealed higher sorption performance with higher capacities and selectivity, and lower heating requirement compared to the liquid sorbent-based method [31,220]. However, the tradeoff is the high operational expenditure resulting from sorbent degradation [220].

On the other hand, capturing technologies for industrial point sources are already well addressed and deployed (details in SM1). CO_2 concentrations vary greatly according to the origin of the point source, ranging from 15-30 vol.% for the cement industry, to up to 90-100 vol.% for natural gas processing [221,222]. The capture cost is around ten times less than the DAC technology cost, and is mainly associated to the cost of energy needed [30,78,221,223].

4.3 Liquid hydrocarbon synthesis

In this step, hydrogen is combined with CO_2 to produce syngas. This happens with a reverse water gas shift reaction (i.e. reaction (6) below, from right to left) in the direct electrolysis pathway (Fig. 11a) or through a co-electrolysis process where water thermal splitting and reverse water gas shift reactions jointly occur, thereby converting steam and CO_2 into syngas [224] (Fig. 11b). This is in particular possible with high temperature electrolysis using SOE (600-

1,000°C; 873-1,273K) [224]. Co-electrolysis provides high conversion and energy efficiencies utilizing the industrial waste heat derived from other industrial processes such as the FT synthesis [75,216]. The reverse water gas shift reaction implies operating conditions of varying from ca. 300--800°C (573-1,073K) depending on the use of a catalyst (typically copper-based; [225–228]) as well as the operating conditions (e.g., syngas compositions; [228]).

As shown in (Fig. 11c), a third option is to thermochemically combine H_2 (from H_2O) and CO_2 for syngas production through the direct use of concentrated solar radiation as energy source. Nuclear and geothermal resources are also possible [229]. Here, the H_2O and CO_2 conversion into syngas is carried out by multi-step thermochemical cycles such as cerium-chlorine, copper-chlorine, sulfur-iodine, iron-chlorine, etc. [202,229–231]. This was demonstrated in the SOLARJET project [202], with a 4 kW solar reactor prototype. The upscaling of this solar thermochemical reactor (50 kW) is being performed within the SUN-to-LIQUID project [205,232], where syngas is to be produced from concentrated solar energy.

To produce liquid fuels, the syngas is further used in either the FT or methanol synthesis process, as depicted in Figs. 11a-c (and Table S3). For the FT route, the syngas-to-kerosene conversion is exactly as descried for biofuels (Fig. 6).

In the methanol route, syngas is converted at temperatures of 150-300°C (423-573K) and under pressures in the range of 10-100 bar (1-10 MPa) in the presence of copper-based catalysts (e.g. Cu/ZnO) [233]. The hydrogenation of CO/CO_2 can be described by the following reactions [234]:

$$CO + 2H_2 \rightarrow CH_3OH \qquad \Delta H_{298K} = -91 \text{ kJ mol}^{-1} \qquad (4)$$

$$CO_2 + 3H_2 \rightarrow CH_3OH + H_2O \qquad \Delta H_{298K} = -50 \text{ kJ mol}^{-1} \qquad (5)$$

The water gas shift reaction occurs simultaneously according to the following reaction [234]:

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad \Delta H_{298K} = -41 \text{ kJ mol}^{-1} \qquad (6)$$

Subsequently, the methanol is condensed and separated by distillation. It can then be processed to the desired chemicals and fuels [233,234]. This conversion and upgrading of methanol to desired fuels and chemicals comprises several processes depending on the preferred target product. For instance, methanol may be used for olefin synthesis (an alkene intermediate to produce kerosene) via di-methyl ether, oligomerization and hydrotreating [37,233,235]. The methanol generated from syngas could also serve for the production of gasoline (denoted as methanol-to-gasoline), as currently done in commercial plants, for example ExxonMobil [236]. However, no aviation electrofuels have yet been produced via the methanol pathway [37]. On the other hand, FT-SPK has already been tested, and approved by ASTM D7566 as a blending constituent. To the best of our knowledge, the first electrofuel industrial plant (Power-to-Liquid for the production of e-kerosene) was officially opened at the beginning of October 2021 in Werlte (Emsland, Germany) [237]. The Lufthansa Group announced, in October 2021, purchases of at least 25,000 L (ca. 25 m^3) annually over the next five years [238].

5. Co-products generation in liquid fuels production pathways (biofuels and electrofuels)

The technologies previously described (sections 3 and 4, Figs. 2-11) generate, besides the desired fuels, multiple co-products, no matter which route is used. As such, biofuel/electrofuel production pathways are to be seen as refineries rather than mere kerosene suppliers (Table S2). Their co-products include liquid fuels (other than kerosene), chemicals, animal feed, etc. and their generation depends on the specific technologies and operating conditions being used within a given conversion route as illustrated in Table 1. In some cases, it may also depend upon market conditions, where the production chain can flexibly be adjusted towards enhanced production of the most valuable product. These co-products represent an additional market or value generation opportunity for the production plant. These co-products are here grouped in three major categories: chemicals, liquid fuels, and other products.

5.1 Chemicals

Various chemical compounds are generated as co-products during production processes. Those that have been reported by biofuel producers are detailed here.

5.1.1 Propane

Propane (C_3H_8) is formed as a co-product in the FT and HEFA pathway (Figs. 6 and 9, respectively). Propane is used in a variety of applications, for instance as a fuel for commercial boilers, camping stoves, heating animal houses (e.g. piglet nursery) in livestock production, etc. It can also be used as refrigerant [239,240]. Propane is generally pressurized and stored as liquid in storage vessels or tanks. Due to its high energy density and high quality combustion characteristics, propane is also used as alternative vehicle fuel for internal combustion engine [241,242]. Propane is categorized as one of the bulk components of liquefied petroleum gas (LPG) in combination with other gases such as butane, isobutene, isopentane. Currently, propane is typically generated as a co-product of natural gas processing and petroleum refinery [137]. The cost of propane thus corresponds with fossil fuels resources. The global propane market has been estimated to reach \$84 billion (with 200 million tonnes in its production) by 2030 [243].

5.1.2 Naphtha

Naphtha is a mixture of liquid hydrocarbons comprising carbon compounds ranging from C_5-C_9 [244]. Naphtha is the main combustible component of both gasoline and kerosene. It has a great potential for diverse industrial purposes including plastic production or its use as a cleaning extraction or dilution agent [245,246]. Naphtha is traditionally generated as a co-product in fractional distillation processes from the petrochemical industry, including the production of (fossil-based) kerosene for aviation fuel. The market price of naphtha is thus closely tight to the price of crude oil [247].

Biofuel pathways involve the production of naphtha as a co-product (including FT, HDCJ, and HEFA; Figs. 6, 7, and 9, respectively). It can be recirculated within the process, for instance in the FT pathway it can be fed into the partial oxidation unit and reformed as syngas feedstock to produce a greater amount of aviation fuel [248]. Additional examples of such bio-based naphtha use are described in SM1.

5.1.3 2,3-Butanediol (optional)

2,3-BDO ($C_4H_{10}O_2$) is a bulk commodity chemical. It is seen as a promising fuel additive or gasoline blendstock for enhancing the octane number [249]. 2,3-BDO is readily convertible to butadiene, butane, methyl-ethyl ketone (MEK) which could be used as intermediates in a variety of product manufactures. In particular, MEK is used in several applications such as solvents in surface coating, printing inks, dewaxing agent, liquid fuel additive, indirect food additive for adhesives and polymers [250]. According to [249,251,252], 2,3-BDO will reach a global market around \$220 million by the year 2027.

2,3-BDO is usually produced on the industrial scale by conventional chemical (or synthetic) methods [253,254]. In the production of aviation biofuel, 2,3-BDO is involved in the SF pathway (Fig. 5). It is not directly generated (hence the optional label above), but can be recovered providing adjustments to the product separation step prior to ethanol production, if favorable market conditions makes it desirable [249]. For recovery, the fermentation broth including liquid mixture or a mixed alcohol stream containing 2,3-BDO would be processed with separation techniques such as fractional distillation, evaporation, pervaporation, adsorption [249]. For instance, LanzaTech has patented the production of 2,3-BDO from CO-rich industrial waste gases (from the steel industry) by fermentation (Table S2). Their commercial ethanol/2,3-BDO production plant has a production capacity of 30-50 million gallons (ca. 0.11-0.19 million m^3) and costed \$75-125 million. In partnership with Orochem Technologies (USA), LanzaTech seeks to economically convert its 2,3-BDO into MEK or 1,3-butadiene through thermocatalytic processes [249].

5.1.4 Isobutene (optional)

Isobutene (C_4H_8) may be optionally recovered in the SuF pathway. It is produced after dehydration of alcohols, just prior the oligomerization process (Fig. 4). If the market conditions are favorable to isobutene, a choice could be made to stop the process after the dehydration process for a certain proportion of the stream, and not continue further to kerosene production. Isobutene production thus implies less bio-based kerosene production; hence its “optional” label.

Isobutene is used as a building block in the manufacturing of several industrial products namely fuel additive, butyl rubber, plastic and lubricants, domestic gas, chemicals and cosmetics, etc. Isobutene can be reacted with methanol leading to methyl tert-butyl ether (MTBE) or with ethanol leading to the production of the gasoline additive ethyl tert-butyl ether (ETBE), used as an anti-knocking agent for the automotive industry [255]. The isobutene polymerization generates butyl rubber that can be used as precursor in several products such as window seals, bottle stoppers, protective gloves, etc. Isobutene is also one of the main constituents of LPG. Moreover, isooctane (a gasoline blendstock) could be generated by dimerization of isobutene [255].

Currently, isobutene as a key chemical building block, is massively obtained from petrochemical sources. Being a major precursor in various industrial applications with continuous demand, its market is worth $25 billion with 15 million tonnes produced annually and used as cosmetic ingredients and specialty fuels [256].

5.2 Liquid fuels

5.2.1 Isooctane

Isooctane is a co-product produced in the SuF pathway (Fig. 4), during the oligomerization/hydrogenation steps. Isooctane represents a large share of gasoline composition and has properties (e.g. high energy content, high antiknock quality) that make it suitable as a blending component for the production of premium-grade fuels [257]. Isooctane is typically produced through the dimerization of isobutene and isopentene generated as by-products from steam cracking of naphtha and light gas oil in refineries, dimer separation and hydrogenation in conventional industrial processes [257]. Isooctane is further used as a feedstock to produce gasoline, in a process where it is blended with naphtha [258].

The major market for the isooctane co-product generated in the production of aviation biofuels appears to be its use as drop-in blending component for the automotive industry, with a global market reaching $99 billion in 2019 [259]. Yet, new markets are also emerging. For instance, high purity solvents or specialty fuels used for racing and classic cars [260].

5.2.2 Gasoline

Gasoline is a refined product of petroleum consisting of hydrocarbon mixtures, additives and blending agents. The gasoline composition strongly depends on various parameters such as the crude oil sources, the refinery process available and product specification defined by octane rating [261]. Gasoline may be generated in particular in the SuF (Fig. 4) and FT (Figs. 6 and 11) pathways. In the latter, gasoline is not directly produced as a co-product, but can be produced from the isooctane generated as explained above. Similarly, gasoline (C_5-C_{12} hydrocarbon ranged; [262]) can also be produced from the APR, FT, and CHJ pathways (Figs. 2, 6, 10, respectively); it is not directly visible in the figures, but is captured within the naphtha fraction. The fraction generated strongly depends upon the operating conditions, which in turn can, to some extent, be adjusted according to the market value of bio-based kerosene, diesel and gasoline. For instance, in the production of aviation biofuel through LTFT process, the proportions of gasoline observed correspond to approximately 10-15% of product distillation output [263]. Additional information about renewable gasoline is described in SM1.

5.2.3 Diesel

Diesel is a key fuel powering compression ignition engine. As for gasoline, it is derived from petroleum refining, and its exact composition is influenced by market demands and prices.

Diesel with carbon distribution ranging C_{10}-C_{20} is produced in aviation biofuel pathways such as SF, FT (Figs. 2, 5, and 7-10) and in considerable volumes. It is optionally produced in the SF pathway, derived from ethanol upgrading processes. Furthermore, diesel is also generated as the co-product from the FT process where the volume produced is also depended upon the operating conditions [263]. The main market for the diesel co-product appears to be as a renewable fuel for terrestrial transport as reflected by several recent examples of purchasing agreements and collaboration deals detailed in SM1.

5.3 Others

5.3.1Waxes

Waxes are generated as a co-product in the FT pathway (Figs. 6 and 11). They consist primarily of straight chain alkanes ($C_{20\text{-}23+}$) which are typically not used in fuel refinery due to their physical properties with i.e. high melting point, low viscosity and hardness. During the LTFT route, the heavy fraction of the FT syncrude accounts for 20-30% weight of total hydrocarbons. The molecular mass of the wax fraction generated is higher relative to the wax produced in the HTFT process [263]. These wax fractions can be cracked into lower molecular weight compounds appropriate for use as liquid fuels or may be sold as precursor of a variety of products. These heavy alkanes have a high potential commercial value due to their competitive prices and versatility in both industrial and medical applications including petroleum jellies, lithium grease, engine oil, industrial gear oil, industrial cleaners, adhesives, etc. [264]. Examples of wax purchase agreements are listed in SM1.

5.3.2 High-protein animal feed

An animal feed co-product is generated in the SuF pathway (Fig. 4), where the protein-rich solid residues derived from the fermentation broth can be further processed. Upon drying, these are often referred to as distiller dried grains, and typically have a high concentration of crude protein with an amino acid profile suitable for animal feed [265]. Ten pounds (around 4.5 kg) of animal feed can be produced from one gallon (ca. 0.0038 m^3) of aviation biofuel [266]. As in the case of the distiller dried grains co-generated with bio-based alcohol (e.g. ethanol) production, the access to the feed market is real and already in place [267].

Table 1 Summary of the co-products generated in the various liquid biofuels production pathways for aviation

Co-product	**Conversion pathways**	**Production status**	**Market uptake and Displacement**	**Co-product ratio**[a] (kg MJ^{-1} kerosene) (% mass)[b]
Chemicals				
Propane	FT HEFA	Generated from triglyceride hydrogenation	• One of the main LPG constituents • Used as fuel in numerous applications (e.g. commercial boiler, burner, etc.) • Displaces fossil-based propane from petrochemical sources (natural gas processing, crude oil refinery)	0.0003[1,12] (1.1%) 0.004[2,3] (9.7%)
Naphtha	FT HDCJ HEFA	Generated along with aviation biofuel (final production step)	• Gasoline blending component • Precursors for plastics manufacturing • Can be recycled in the FT unit to produce additional aviation fuel • Displaces fossil-based naphtha from petrochemical sources	0.0004[1,12](1.3%) 0.019[4] (27.0%) 0.005[2,3] (12.5%)
2,3-butanediol (2,3-BDO)	SF	Generated along with ethanol, may be recovered through a separation process before the ethanol production (optional)	• Conversion to various precursors namely MEK, 1,3-butadiene, etc. • Displaces 2,3-BDO from chemical engineering (or synthetic) methods	0.013[5] (if they are recovered from the fermentation process) (33.0%)
Isobutene	SuF	Derived from isobutanol, which can be further processed the dehydration (optional)	• Precursor for numerous products such as butyl rubber, plastics, isooctane, etc. • One of the main LPG constituents • Dimerization for isooctane production • Polymerization in butyl rubber production • Displaces isobutene from petrochemical sources	Unclear, because isobutene is an intermediate that can either be processed (fully or partly) to kerosene or not depending on the market context (flexible production)

Liquid fuels				
Isooctane	SuF	• Generated along with aviation biofuel (final production step)	• (Premium-grade) gasoline production • Displaces fossil-based isooctane from petrochemical sources	0.002[6,7] (7.5%)
Gasoline	APR	• Derived from isobutanol in the SuF process, which can be further upgraded to isooctane (optional) • Generated along with aviation fuel (final production step)	• Transportation fuel • Displaces fossil-based gasoline derived from petrochemical source	0.003[8] (8.0%)
	FT			0.004[a,1,12] (14.7%)
	IH^2			0.086[9] (72.2%)
	CHJ			0.016[10] (28.1%)
Diesel	APR	• Generated along with aviation fuel from APR, FT and HEFA processes • Derived from isobutanol in the SuF process, which can be further upgraded to diesel (optional)	• Transportation fuel • Displaces fossil-based diesel derived from petrochemical sources	0.012[8] (32.0%)
	SF			0.003[5] (8.7%)
	HDCJ			0.028[4] (40.1%)
	HEFA			0.009[2,3] (21.1%)
	IH^2			0.010[9] (8.3%)
	CHJ			0.018[10] (31.0%)
Others				
Waxes	FT	• Generate along with aviation biofuel (final production step)	• Raw materials for various products such as petroleum jellies, adhesive, etc. • Displaces petrochemical-based waxes	0.001[1,12] (2.9%)
High-protein animal feed	SuF	• Derived from fermentation residues and may require further processing such as evaporation/drying	• High protein and nutrient concentrations derived from dried distiller grains • Displaces marginal carbohydrate, protein and lipid sources (maize, soybean meals and palm oil) in animal feed	0.037[11] (59.4%)

Note. [a] Co-product ratios presented are extracted from the available literature in which the operating conditions intend to maximize kerosene production. This could not be done for the advanced IH^2 process, which is currently essentially used for gasoline production as demonstrated in the study of [174]. [b] The mass (%) of the co-products are provided in parenthesis.This includes the % (mass) out of all products (including kerosene): therefore the total is lower than 100%. The calculation procedure can be retrieved in SM2. The considered lower heating value (LHV) of kerosene is 43.0 MJ kg^{-1}.

References: [1] [143]; [2] [268]; [3] [269] ; [4] [270]; [5] [133]; [6] [271]; [7] [272]; [8] [273]; [9] [174]; [10] [274]; [11] [266]; [12] [275]

6. Electric (battery-based) aviation

Batteries can serve as the energy sourcing for either propulsion (as further explained in section 10.1) or non-propulsion systems (overall electricity within the aircraft). The latter is often referred to as the “more electric” architecture, and is applied in aircraft to power services such as the flight control system and cabin environmental control system, or the auxiliary power unit (APU) [49,276,277]. “More electric” architectures were implemented in the Airbus A350 and Boeing 787 [51,276].

Albeit battery is one of the most promising energy sourcing (for propulsion) in terms of climate impacts (further discussed in section 8), there are several limitations to overcome for a widespread implementation within aviation. First, they cover essentially short-haul flights due to the limited energy storage capacity. Table 2 details the emerging lithium-based battery technologies for aviation applications [277]. The present lithium-ion batteries (Li-ion) have a maximum specific energy of 400 Wh kg^{-1} (1.44 MJ kg^{-1}) [49,278], which is relatively low in comparison with conventional kerosene, having a specific energy density of ca. 12,000 Wh kg^{-1} (43.2 MJ kg^{-1}). This introduces a crucial mass penalty factor to consider [277,279,280], and affects the final supplied service (e.g. reduction of the number of seats and thus passengers carried, a lower distance travelled; [56,279]).

To overcome this challenge, different innovations are in place. The main focus has been on the use of different materials for the cathode/anode, and on the use of different electrolytes (Table 2). This has led to the development of high performance batteries including advanced lithium metal/silicon anode (Adv. Li-ion) [49], solid-state electrolyte battery (SSB) [278,281], lithium-sulfur (Li-S) [282,283] and lithium-air (Li-air) batteries [284,285]. Moreover, nickel-rich NCM (Nickel-Cobalt-Manganese)-811 cathodes [49,286], alternative anode material to graphite (e.g. silicon, lithium metal) [287], approximate solid-state electrolytes in SSB [49], prevention of polysulfide shuttle effects in Li-S batteries [288,289] and improvement of moisture sensitivity in Li-air batteries [285,290] are additional avenues being investigated.

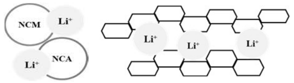
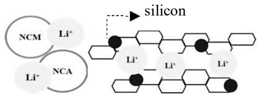
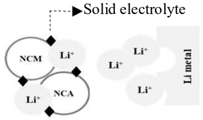
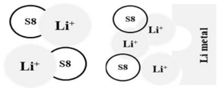
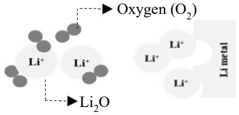
Other advances for batteries have been introduced, for instance with systems based on sodium, magnesium, zinc, and aluminum [291], albeit, still in the early stage of research and development. These can be potential candidates to substitute the lithium, cobalt, natural graphite, silicon metals used as the major materials for the electrodes (defined as EU critical raw materials; CRM [292]) [293]

Another concern applying to battery electric aviation relates to safety, among other associated with overheating of the battery, which can cause failure of thermal runaway, known to be a key cause of undesired incidents [294,295]. For example, the fire incident of the Boeing 787 Dreamliner in 2013 was reported to be due to the failure of the Li-ion batteries, powering the aircraft APU [296]. Therefore, the development of high-performance, long-lived batteries with thermal stability is one of the numerous technological challenges to be solved before broad deployment [286].

One other important issue relates to the charging time of batteries and the possibility to recycle them after their limited lifetime (>1,000 cycles for current Li-ion batteries; [49]). The battery swapping station model, where batteries are not charged but replaced by charged batteries during the turnaround phase of the plane, is one option that has been considered to overcome the charging time issue [297,298], however, this option implies high investment costs because of the great number of batteries involved.

Although battery recycling has advantages over the use of virgin metals (e.g., need for less energy for extraction operations, avoid emissions from batteries disposal) [299], several challenges remain to be alleviated, related to a variety of factors. The efficiencies of the collection and dismantling methods (e.g., difficulties in the dissociating used graphite from others) and the appropriate recycling technologies (e.g., for the developing sustainable methods, for high purity requirements such as 99.9% carbon purity for the graphite; [300]) are some examples [299,301,302].

Table 2 Battery technology outlook for aviation

		Lithium-ion (Li-ion)	**Advanced Li-ion**	**Solid-state battery**	**Lithium-sulfur (Li-S)**	**Lithium-air (Li-air)**
						
Battery cell chemistry	Cathode (+)	Lithium metal oxide such as LFP, LMO, Li-NMC Li-NCA[3]	Lithium metal oxide with Ni-rich fraction (e.g. Li-NMC811)[4,5,6]	Lithium metal oxide With Ni-rich fraction (e.g. Li-NMC811)[4,5,6]	Sulfur Graphene Acetylene black[6,7,8]	Porous carbon (e.g. carbon nanotube)[10]
	Anode (-)	Graphite (with silicon)	Lithium metal or Silicon[5,11]	Graphite Silicon[3]	Lithium metal Graphite[5,12]	Lithium metal[5,12,13]
	Electrolyte	Organic liquid (e.g. lithium salt-LiPF$_6$)[3]	Organic liquid (e.g. lithium salt-LiPF$_6$)[3]	Inorganic solid (e.g. Li$_{10}$SnP$_2$S$_{12}$) Organic solid (e.g. polycarbonate)[5]	Organic liquid (e.g. LiN(SO$_2$CF$_3$)$_2$)[3,5]	Organic liquid (e.g. LiPF$_6$, LiCO$_4$) Aqueous solution (e.g. H$_2$SO$_4$)[10]
	Binder	PAA PVDF[6,8]	PVDF[6,8]	PVDF[6]	PEI PVP[8]	PAA[8]
	Conductive additive	Carbon black[3,6]	Carbon black[3,6]	Carbon black[3,6]	Not specified	Not specified
Cell properties	Gravimetric (Wh kg^{-1})[1] (MJ kg^{-1})	300[14] (1.08)	450[14] (1.62)	400-500[15] (1.44-1.80)	300-400[14] (1.08-1.44)	1,350[9] (4.86)
	Volumetric (Wh L^{-1})[1] (MJ L^{-1})	700[14] (2.52)	1,200[14] (4.32)	Not specified	400[14] (1.44)	Not specified
	Life-time (cycle)[2]	1,000-2,000[16-17]	1,000[18]	500[15]-1,000[14]	100[14]	Not specified
	Nominal cell voltage (V)	3.7[14]	3.8 (Li-metal anode)[14]	3.8[15]	2.1[14]	2.9[14]
Status	Development status	Commercial scale (TRL 9)[14,16]	Demonstration scale (TRL 7)[14,16]	Technology validation Small-scale prototype (TRL 3-4)[14,16]	Small-scale prototype (TRL 4)[14,16]	Technology concept (TRL 1-2)[14,16]

[1] Energy density is reported at the cell level derived from published papers and reports which were available at the time of writing. The SI unit of energy (MJ) is presented in parenthesis. [2] Lifetime of battery is represented for 100% Depth of Discharge (DoD). TRL displayed has been retrieved from the references as indicated in the table.

Sources: [3][299]; [4][303]; [5][286]; [6][37]; [7][304]; [8][305]; [9][306]; [10][307]; [11][308]; [12][309]; [13][310]; [14][49]; [15][311]; [16][278]; [17][56]; [18] [312] LFP: Lithium Ferro Phosphate; LMO: Lithium Manganese Oxide; NCA: Nickel-Cobalt-Aluminum; NMC: Nickel-Manganese-Cobalt-; PAA: Polyacrylic acid; PEI: Polyethyleneimine; PVDF: Polyvinylidene fluoride; PVP: Polyvinyl pyrrolidone; TRL: Technology Readiness Level.

7. Hydrogen aviation

The energy density of hydrogen, in terms of energy-to-weight ratio (MJ kg^{-1}) is three-fold that of conventional kerosene, rendering it suitable for longer flights in large planes supporting high payload capacity. Despite being lighter, hydrogen, in its cryogenic liquid form, has an energy-to-volume ratio (MJ L^{-1}) four-fold lower than fossil kerosene, which implies obvious storage challenges [313]. The different pathways for hydrogen production through water-splitting technologies are presented in Fig. 12.

Cryogenic hydrogen (referred as liquid hydrogen; LH_2) requires cryogenic storage (-253°C; 20K) to maintain hydrogen in a liquid form [314]. This is due to hydrogen's high specific volume at standard atmospheric pressure and temperature.

Hydrogen propulsion system have been demonstrated whether in combustion mode or fuel cell mode (the latter is detailed in section 10.2.1). The Russian manufacturer Tupolev manufactured, in 1989, the first hydrogen aircraft using both gaseous H_2 (from an LH_2 tank), and liquefied natural gas (LNG) fuel in a gas turbine engine to propel the Tupolev 155 (Tu-155) [315]. The direct use of H_2 in gas turbines and piston engines generally involve that LH_2 is pumped from the tank to a heat exchanger, where LH_2 is heated to gaseous form prior to being injected into the combustion chamber of the engine [316]. The A380 MSN1 is an additional undertaking demonstrator to be powered by the H_2 combustion mode, which is expected to fly by 2026 [317]. Those require, besides the gas turbine with modified components (such as combustion chamber, fuel injector), a cryogenic tank with insulation system, additional on-board infrastructure such as high-pressure pumps, and a heat exchanger suitable to H_2 combustion characteristics (e.g., a tendency to flashback, a high flame temperature) [62,313,316,318], although scarcely detailed. Hydrogen turbine engines with low-NO_x emissions are already expected as the next improvement of this technology [58,62,63]. Boeing Phantom Eye is another example of an unmanned drone powered by hydrogen combustion in piston engines [319]. Other hydrogen aircraft, and projects can be retrieved in SM1.

Cryogenic LH_2 has a propensity to leak due to hydrogen's high boil-off rate and small size molecule. Because of this, the use of H_2 raises important concerns about safety, and involves stringent procedures and requirements with regards to safety regulation certification, at airports, production and storage sites, transportation, distribution as well as for the aircraft operation, to avoid and control these leakages in case of occurrence [58,313,320]. It is even suggested to build airports where only H_2 would be used for all airport vehicles used for ground operations [321]. Despite this, H_2 has some advantages safety-wise, over conventional kerosene, such as lower flammability (more H_2 leak than kerosene leak is required prior to fire hazards, among others due H_2 rapid dispersion in air), and a higher auto-ignition temperature (550°C; 823K for H_2 versus 300°C; 573K for conventional kerosene), leading to a lower risk of spontaneous ignition of the fuel [320–322].

The key challenge of hydrogen as aviation fuel relates to the mass penalty induced by the onboard storage system. The mass penalty derived from the H_2 storage system is, at current technological state, reported to be twice as much as the conventional aviation fuel-based APU, when the compressed H_2 system is applied [58,63].

A light cryogenic storage tank with cooling system advancement is necessary for tackling this challenge [62]. On top of this, the fuselage may need a further redesign to accommodate the cryogenic hydrogen tank. The Airbus ZEROe project for instance announced the use of a blended wing body (BWB), as one type of aircraft allowing to efficiently accommodate hydrogen [323].

8. Semi-quantitative comparison and prioritization in the perspective of environmental mitigation

The strengths, challenges and prospects of the four SA families studied herein are summarized in Fig. 13. At the light of Fig. 13, it however remains unclear which pathways are the most promising from a technical, economic and environmental point of view.

To this end, a semi-quantitative multi-criteria decision matrix has been established to enable a relative prioritization of the various types of SAapproaches elaborated to this point (Table 3), inspired by the approach used in [324]. It is meant as a tool to pre-screen the global performance, and is by no means intended to replace state-of-the art quantitative assessment. It proposes three performance criteria (Table 3) seen as key forsustainable aviation: technology status, market prospects, and environmental performance, reflecting that short- and longer-term environmental mitigation are deeply intertwined with broader market considerations. For each of these critical criteria, sub-criteria were elaborated, to which a score (from 0 to 3) is given, on the basis of quantitative or qualitative descriptions (Table 3). The tool is distinctively applied on two timescales: near-term future (~2030) and long-term (~2050). These 10-20 years' time scopes are used as judged representative to reflect the time elapsed between two generations of technologies [325]. Several supporting references, including literature, scientific papers, patents, industrial/research organization reports, announcements, were accessed to assign scores for all criteria, and all pathways. This is thoroughly documented in SM2.

Figure 14 summarizes, for all pathways considered herein, the score obtained for some of the key quantitative performance indicators of Table 3, namely fossil fuel reduction, GHG reduction and technology readiness level (TRL), distinguished according to the time scope. Accordingly, 11 groups of pathways can be distinguished (Fig 14). Pathways of group (1) are, according to the criteria selected in Fig. 14, the most promising, implying zero fossil fuels requirement and high (>60%) GHG reductions expected in comparison to fossil kerosene. It involves hydrogen (produced with alkaline or PEM electrolysis) as well as one biofuel and electrofuel pathway. The former is highlighted as particularly interesting due to its higher TRL. On the other hand, pathways of group (11), involving Li-ion and Advanced Li-on batteries used in hybridized mode (i.e. used for propulsion) are shown as the least interesting, involving a high fossil fuels share and less than 40% expected GHG reductions.

Despite useful, Figure 14 does not allow to reflect all criteria, especially the qualitative ones, of Table 3. In a second stage, the Analytical Hierarchy Process (AHP) protocol proposed by [326] has therefore been used to assign weights to the different sub-criteria through the application of weighting factors between 0 and 1 to each sub-criteria, the total being 1 (Table 3). This is of course subjective, but the vision, for the screening exercise to be useful, is to supply a weight choice that is transparently argued (SM2). For instance, here, the sub-criteria “requirement of fossil fuels” is the one with the highest weight for both the short- and long-term, keeping a maximum of fossil carbon in the ground being seen as a key driver for the development of SA at the first place. Final weighted scores are calculated by multiplying the weighting factors by the sub-criteria assigned scores, as detailed in SM2. The resulting final weighted scores are presented in Figs. 15 and 16, for near- and long-term, respectively, with a breakdown per sub-criteria.

As a result (Figs. 15-16), the most promising biofuels technologies uncovered for the near-term (Fig. 15) are HEFA (oleochemical wastes) and BGFT (biomass gasification and the FT process, with residual lignocellulosic biomass), significantly standing out from the other biofuel technologies. This result for (waste-derived) HEFA is explained by two key advantages of this pathway in comparison to the other options addressed: it benefits from existing capacity and it is not directly requiring additional arable land demand. For BGFT (based on lignocellulosic biomass), this decoupling from additional land demand is also important, along with its expected lower well-to-wake GHG emissions ([327,328]; SM2). Based on available literature, a well-to-wake reduction of GHG emissions of 91% was considered for BGFT (in comparison to fossil fuels), which is much higher than for all other biofuels (SM2). Yet, this result is to be considered with caution, as essentially reflecting methodological choices attributing GHG credits to energy-dense co-products. For electrofuels, only one technology stands out, namely FT using H_2 from alkaline water electrolysis and carbon from DAC (irrespective of the exact DAC technology), due to its expected lower well-to-wake GHG emissions. For electric (battery-based) systems, current technology for lithium-ion battery is highlighted with a higher weighted score, essentially benefiting from its greater maturity. For H_2 systems, alkaline water electrolysis significantly stands out from the other H_2 production technologies assessed, benefiting from its greater existing capacity (and the weight assigned to this criterion in the near-term).

For the long-term (Fig. 16), the results are similar for electrofuels, and biofuels, except that here, the BGFT lignocellulosic biomass clearly stands out from all other biofuels, essentially because of its expected lower well-to-wake GHG advantage, weighted higher for the long-term (SM2). This explanation also applies for the electrofuels result. The alkaline water electrolysis is again shown as the most interesting technology for H_2 production, but the difference with other technologies is less pronounced than in the short-term. For electric (battery), however, the lithium-air battery is highlighted as the most interesting technology due to its anticipated higher energy density (see Table 2) and hence ability to carry more passengers with a longer distance traveled [56] or to the use less fossil kerosene supplement in a hybrid mode. Higher weights are put on these sub-criteria, namely the requirement of fossil fuel, GHG emissions reduction, in comparison to the short-term.

Table 3 Semi-quantitative criteria for prioritizing sustainable aviation approaches[a]

Overall criteria	**Weight (0-1)[b]**		**Sub-criteria**	**Score**				**Comments**
	2030	**2050**		**0**	**1**	**2**	**3**	
Technology current status	0.19	0.25	Requirement of fossil fuels	100% fossil fuel required	> 50% fossil fuel required	≤ 50% fossil fuel required	Not required or Required for takeoff only	Based on known standards for aviation (e.g. ASTM D7566, etc.)
	0.12	n/c	Technology readiness level (TRL)	0-2	3-4	5-7	8-9	The general principles used for TRL assessment can be retrieved in SM2.
	0.12	n/c	Existing capacity (× 10^{9} MJ y^{-1})	<1	1.00-4.99	5.00-9.99	> 10	Current production capacity
	0.01	0.02	Energy density (MJ kg^{-1})	<20	20-30	31-40	> 40	43 MJ kg^{-1} is considered as a default value for conventional kerosene [143]. The ranges proposed herein are based on this.
Market prospects	0.03	0.04	Commodity co-products (with offsetting production costs) (€ kg^{-1})	< 0.10	0.10-0.49	0.50-1.0	> 1.0	The average jet fuel price in 2020 was around 0.60 € kg^{-1} [329]. We also consider that aviation kerosene has a fuel density of 0.785 kg L^{-1} [143].
	0.08	0.06	Passenger carrying capacity (seat)	Not for carrying passenger	1-50	51-100	> 100	Electric battery aviation is evaluated based on [56,325]. Hydrogen aircraft are evaluated based on [63].
	0.08	0.06	Flight range (km)	<500	500-2,000	2,001-5,000	> 5,000	Electric battery aviation is evaluated based on [56,330]. Hydrogen aircraft are evaluated based on [63].
	0.04	0.03	Cost competitiveness in low-fossil carbons future (€cent MJ^{-1})	>5.00	3.01-5.00	1.31-3.00	<1.30	Conventional jet fuel price 1.30 €cent MJ^{-1} [329].
Environmental performance	0.12	0.17	GHG emission reduction in comparison to fossil kerosene, as reported by studies	<40%	40-60%	60-80%	>80%	The Well-to-Wake GHG emission of conventional jet fuel is considered to be 87.5 gCO$_2$-e MJ^{-1} [331].
	0.02	0.08	Energy demand to produce the fuel (MJ MJ^{-1} fuel produced or battery)	> 10.0	5.01-10.0	2.0-5.0	< 1.99	
	0.12	0.17	Land requirement	Growing crops with a high impact on the food/feed competition	High share of residual biomass (wastes), but crops may be involved	Biomass share overall low, and from residual resources only	No biomass needed	Relates to feedstock sourcing only
	0.05	0.12	Sustainable availability of feedstock	Highly limited availability Feedstock otherwise used for food and feed	Limited availability: Feedstock otherwise not used for food/feed (Non-edible crops)	Limited availability but independent from land (e.g. residual, waste) included in the EC critical raw materials (CRMs) list 2020 [292]	Abundant availability (e.g. solar energy, CO$_2$ from the air)	
	0.02	0.01	Water demand (L H$_2$O MJ^{-1} fuel produced or battery)	>100	50.01-99.99	10.01-50	< 10	Considering the overall water requirement during theirproduction stage.
	1.00	**1.00**						

[a] n/c: not considered. The reference data and rationale used for the scores attributed herein are further detailed in SM2; [b] The weight of each sub-criteria has been calculated through the Analytical Hierarchy Process (AHP) as described in [326]

9. Other emerging energy sourcing for aviation not considered herein

9.1 Liquefied natural gas (LNG) based aviation

The use of LNG (i.e. liquefied methane) as a fuel is highly researched and discussed for the maritime sector ([332,333]), and to some extent the aviation sector (e.g. Savion aerospace; [334]), though it is still very marginal. One driver for the development of this technology is the price of fuels; low natural gas price relative to fossil kerosene may favor the LNG. It is further suggested that LNG aviation fuels could reduce CO_2 tailpipe emissions of approximately 20-25% relative to conventional kerosene, for the same energy content [313,335]. Although LNG is presently derived from natural gas, it could stem from biomethane generated through anaerobic digestion [121], biomass gasification [336], or power-to-methane routes decoupled from biomass [337]. It also has a gravimetric energy density slightly higher than conventional kerosene (around 48-54 MJ kg^{-1}) [335,336].

LNG for aviation, however, still involve a number of challenges. First, it implies, as for LH_2, cryogenic form (-162°C; 111K), and thus a certain mass penalty [313,338,339]. It also implies infrastructure not already in place (e.g. refueling system, LNG storage tanks, liquefaction facilities) [335,338]. Importantly, eventual leakages of LNG is another factor to be considered, as methane has a global warming potential (GWP_{100}) around 28 times the one of CO_2 [340].

As LH_2, LNG is compatible with whether combustion mode or fuel cell mode [341]. The former has been show-cased in gas turbines (e.g., TU-155 aircraft [315]) as well as within the Advanced Hybrid Engines for Aircraft Development (AHEAD) project [342]. The combustion mode implies that modification and/or change in propulsion systems are required, including engines, injectors and heat exchangers, able to use this fuel [343,344]. The fuel cell mode has been showcased in a solid oxide fuel cell (SOFC) within the Subsonic Ultra Green Aircraft Research (SUGAR) project [341]. To make it viable on long-range aircraft, additional research on advanced materials for fuel cells is however needed [341].

9.2 Ammonia based aviation

The use of liquefied ammonia (NH_3) for aviation has been discussed and considered within some research and industrial communities [345,346]. For instance, ammonia-powered aircraft are being developed within the Zero-carbon Ammonia-Powered Turboelectric propulsion project (ZAPturbo) [347]. In comparison to LH_2, it involves lower mass penalties owing to the superior density of liquid ammonia (730 kg m^{-3}; [343]). Ammonia requires refrigeration to -33°C (240K) under atmospheric pressure, this implies prior refrigeration when on ground, and is maintained by the outside temperature during cruise altitude (at ca. 8-12 km; [348,349]) through heat exchanger systems [346]. Refrigerated ammonia can be stored in the wings like in the case of conventional kerosene [350].

However, most ammonia is currently produced from hydrogen through steam reforming of natural gas, and N_2 is derived from air separation through the energy-intensive Haber-Bosch process [343,351]. Alternatives using renewable H_2 (derived from water electrolysis rather than natural gas; also referred to as the 2nd generation path) [352,353], or electrochemical ammonia synthesis as an alternative to the Haber-Bosch process (electrochemical N_2 reduction reaction, NRR, known as the 3rd generation path) [351,353,354] are being developed. However, at present, the production cost of renewable ammonia is roughly twice the price of conventional ammonia. Its production cost is essentially shaped by the price of renewable electricity [355]. In addition, ammonia has a low gravimetric energy density (18.6 MJ kg^{-1}; [343,347]), this means that ammonia-powered aircraft would have a shorter flight range relative to (conventional) kerosene and hydrogen [356], considering a similar payload mass. Moreover, an enhanced use of atmospheric N_2 to produce (additional) ammonia implies an enhanced risk for contributing to excess reactive nitrogen in the environment, known to cause a range of negative environmental, socio-economic, and health consequences [353,357], besides being one of the exceeded planetary boundaries [358].

Ammonia per se has a high ignition temperature, and low flow velocity [357,359]. Therefore, a mixture with other reactive molecules (e.g. H_2), has been proposed as one option to improve combustion properties ([353]). In this endeavor, it is proposed to add a cracking unit to the aircraft, where part of the ammonia dissociates into nitrogen and hydrogen, resulting in a mixture of NH_3-H_2-N_2 [347,357]. This feature closely makes it compatible with the current (hydrocarbon-based) gas turbine engines [347,353,357], and is being developed by e.g. Raytheon Technologies Research Center [347] and Reaction Engines Ltd. [345]. Ammonia can also be employed in the fuel cell mode [360], but no showcasing of this has been found. Water vapor, nitrogen, and nitrogen oxides (NO_x) are considered as the only tailpipe emissions of ammonia [356]. To minimize the effects derived from NO_x formation, mitigation technologies such as a low-NO_x combustors have been proposed [347]. Additionally, new materials for storage tanks (in particular for on-board NH_3) are required to safely deal with the corrosive properties of NH_3 [353]. Ammonia's toxicity (inhalation hazards) is another issue to be tackled and would imply mandatory specific training before handling [353,357].

9.3 Solar aviation

Solar energy generates growing attention for aviation [361,362], due to its unlimited availability and envisioned sustainability. Solar-powered aircraft for passenger-carrying purposes however faces many challenges. First, solar panels are limited in their capturing efficiency (around 20%; [363,364]). Even if an increase in efficiency (reaching 100%) has been achieved ([362]), the generated power might not be sufficient to deal with the payload a passenger aircraft implies. In addition, because of low efficiency power devices of solar-powered aircraft (e.g., photovoltaic cells, rechargeable batteries; [364]), the maximum speed would not reach beyond 50 miles hr^{-1} (160 km hr^{-1}) (commercial aircraft traveling at about 600 miles hr^{-1}; 965 km hr^{-1}) [362]. The high variability of photon capturing angles is also reported to result in lower power generated (relative to the expected maximum power) [362,364]. Other issues are reported, for instance, the vulnerability of solar panel materials to adverse weather conditions, or the need for large wingspans. Nevertheless, solar power is successfully applied and deployed for unmanned aircraft system/unmanned aerial vehicle (UAS/UAV) as a satellite service (additional detail in SM1).

The deployment of solar aviation for carrying passengers therefore appears as an unlikely option till the first half of the century, but it may be considered in hybridization with the other SA systems discussed within the present study.

9.4 Others

Finally, additional initiatives, albeit marginal such as methanol [365,366], and ethanol [367,368]direct use, do exist and have not been covered within this review due to a lack of available documentation about these.

10. Electric propulsion systems for aviation

Electric propulsion (to be distinguished from battery-electric) represents configuration schemes involving an electric motor to supply mechanical power to the aircraft propulsors (which could be propellers or fans) (Fig. 17). These configuration schemes differ with regards to whether they imply or not (i) energy storage from batteries; (ii) a (H_2) gas turbine fueled by H_2 or liquid hydrocarbons and (iii) fuel cells fueled by H_2 or liquid hydrocarbons and supplying electricity to electric motors.

Advancing the propulsion efficiency is, after the energy sourcing itself, one key lever considered to improve the overall environmental performance (including noise reduction) of future aviation. Throughout this section (and in Fig. 17), the term propulsor represents devices generating thrust either by a propeller in the turboprop or by fans in the turbofans. It also encompasses the generation of shaft power in the case of turboshaft engines. The term "(H_2) gas turbine" collectively refers to the turboshaft, turboprop, and turbofan engines. Converter refers to the devices converting the voltage of the electrical power source, sometimes denoted as power electronic [276], and considers that one type of converter is an inverter, which converts direct current (DC) to alternating current (AC).

To represent the proportion of electric power in total power, two parameters are used [51,56], respectively degree of energy hybridization (H_E) and degree of power hybridization (H_P). H_E and H_P vary from 0 to 1 depending on the conceptual designs [51,369]. H_E is zero when there are no energy storage devices involved (no batteries), and 1 when the power is solely supplied via batteries. H_P is 1 when only electric motors are supplying the required mechanical power, and 0 if no electric motors are involved in supplying the required power.

10.1 Battery-dependent electric propulsion configurations

Electric aircraft relying on batteries have gained substantial interest in recent years. Batteries can serve as the energy sourcing for propulsion either as standalone ("all electric" concept; Fig. 17a) or in combination with other fuels ("hybrid electric" concept; Figs. 17b-c).

The all electric concept is also denoted as "full electric" (e.g. in [370–372]). An all electric configuration (Fig. 17a) involves that (H_P=1) because the propulsor is powered by an electric motor only but also involves that (H_E=1) because it relies solely on batteries without any hydrocarbon-based fuels or H_2 [51].

The concept of "hybrid electric" (Figs. 17b-c) implies, on top of the electric motor, the use of a (H_2) gas turbine fueled by liquid hydrocarbons or H_2 to generate additional mechanical power. As AC power is required for most of the electric motors that can be used in aviation, converters are required [373]. The parallel and series hybrid electric concepts are the two main configurations that have been tested (Figs. 17b-c), however, a series-parallel hybrid scheme has also been demonstrated (not depicted in Fig. 17) [373,374]. The parallel hybrid electric configuration (Fig. 17b) involves (H_2) gas turbines and electric motors to generate mechanical power from fuels combustion and batteries, respectively, [375]. Hence a mechanical transmission (typically gearbox) is required to integrate and control these two mechanical power sources prior to the propulsor [373,376,377]. In the series hybrid electric configuration, the generator powered by the (H_2) gas turbine produces electricity for both the electric motor(s) (which is directly linked to the propulsor; $H_P = 1$), and to charge the battery onboard ($0 < H_E < 1$) [51,371,378] (Fig. 17c).

Several projects have been launched to demonstrate the feasibility of these battery dependent configurations) (Fig. 18). For example, the startup Eviation (Israel) launched Alice, a nine-passenger all electric aircraft with an autonomy range of 650 miles (1,046 km), expected to be in service by 2024 [379]. The aircraft relies on a battery that can be fully charged within 70 minutes and is expected to be FAA-certified (FAA: USA Federal Aviation Administration) by 2024 [379]. However, a fire incident has occurred during the Alice ground test in January 2020 caused by a fault with a ground-based battery system [380]. The company magniX (Canada) designed and demonstrated a high-power density electric propulsion system with the world's first fully electric commercial seaplane, tested on a flight in December 2019 (6-passenger aircraft; flight range of 1,000 km) [381,382]. In May 2020, magniX together with Harbour Air successfully tested an all electric powered system carrying up to 9 passengers for a reported flight range of ca. 160 km (e-Caravan; retrofitted from Ceravan 208) [383,384]. It is announced to be the largest all electric aircraft with a successful test flight (in the middle of 2020). In Europe, EasyJet and Wright Electric are developing since 2017 an all electric aircraft (186 seats) and announced it will start using electric aircraft to cover short-haul routes by 2030 [385,386]. Additional details on these emerging developments along with specifications on these technologies are presented in SM1.

10.2 Electric propulsion independent from batteries

Electric propulsion configurations independent from the presence of batteries include: Fuel cell electric and Turboelectric systems.

10.2.1 Fuel cell electric propulsion (may involve batteries as backup)

A fuel cell propulsion system could be employed in the mode of all electric (in Fig. 17d), or hybrid electric (electric motor and (H_2) gas turbine; Figs. 17e-f) [373,387], where hydrocarbon-based fuels or H_2 are converted to electricity that in turns powers an electric motor involving a modification of the propulsion system [62].

Fuel cells serve as energy converters, not as energy storage as in the case of batteries. Fuel cells can continuously produce electricity as long as fuels (whether hydrocarbon-based fuels or H_2) are fed to the fuel cell. The existing fuel cell technologies, for the automobile sector and stationary power applications, include the proton exchange membrane fuel cells (PEMFC) and SOFC , depending on the electrolyte employed, operating conditions, and fuels compatibility [387,388]. Fuel cells can be considered for the propulsion system in aviation, providing an increase in their specific power (targeting 2 kW kg^{-1}) has been achieved [57,63,388]. The electricity produced in excess of what is needed for propelling the aircraft can additionally be stored in batteries as a backup energy source, not directly involved in the propulsion system [62] (Figs. 17d-f). Fuel cell propulsion also provide the advantage of short refueling time relative to batteries with fewer risks of reducing the lifetime [58].

Hydrogen fuel cell designs for aircraft are being developed, for instance by Airbus in ZEROe concepts (hybrid hydrogen propulsion system). These aircraft are expected to enter service (with a carrying capacity of 100-200 passengers) by 2035 [323]. Furthermore, the first commercial H_2 fuel cell aircraft (with a carrying capacity of 10-20 passengers, and maximal distance traveled up to 555 km), was announced by ZeroAvia to be ready by the year 2024.

Unlike the H_2 combustion in gas engines and piston engines (Fig. 17i) where air is involved in the combustion process (thus involving nitrogen outputs such as nitrogen oxides; NO_x); the only two by-products from H_2 fuel cell systems are water vapor and a small amount of heat. This, however, involves non-negligible cooling requirements [58,63]. Additionally, fuel cell propulsion is unlikely to be competitive for heavy payload and long distances, with four times the mass of current aircraft engines to generate the same power output [58,63].

Various projects have been intensively studied in developing and improving hydrogen fuel cell technologies (Fig. 17), as detailed in SM1. Hybridization with fuel cell and battery propulsion is another alternative configuration (not depicted herein) [389].

10.2.2 Turboelectric propulsion (no batteries)

Turboelectric propulsion (Figs. 17g and h) is another configuration for electric powertrain systems, and fully relies on hydrocarbon fuel or H_2 as its energy source. Here, no energy storage (e.g. batteries) is involved [369,370,378]. A (H_2) gas turbine drives the generator that powers electric motors for thrust or shaft power generation. In the full turboelectric system (hence, $H_E = 0$, $H_P = 1$; Fig. 17g) [51,390], the generic (H_2) gas turbine term denotes a turboshaft engine [391]. In the partial turboelectric concept (Fig. 17h), the generated mechanical power from the (H_2) gas turbine is partially delivered to the generator ultimately feeding the electric motor(s), and the remaining power is delivered directly to the other propulsors [51,279,370]. In this case (Fig. 17g), H_E would be zero as there are no energy storage devices involved, while H_P would be between 0 and 1 because both a (H_2) gas turbine and an electric motor are used to supply energy to the propulsor. In this particular case, however, a clear standard for attributing the appropriate non-zero and non-one hybridization value is still missing [51,369].

Albeit the propulsive efficiency of electrically-driven propulsors can improve, the integration of the electric system introduces mass penalties (although lighter than the all electric configuration as it does not involve batteries) [279,391–394]. To enhance the overall performance in terms of fuel burnt and mass, this configuration concept has been integrated with optimized aircraft design, for instance in NASA's STARC-ABL aircraft design (Single-aisle Turboelectric AiRCraft with an Aft Boundary - Layer propulsors) [394]. This allowed a 12% reduction of fuel burnt compared to the conventional aircraft concept (conventional kerosene configuration (Fig. 17j) and airframe design) [394]. The Wright ECO-150R aircraft capable of carrying up to 150 seats (flight range of ~ 3,000 km) initially demonstrated ca. 44% fuel burn reduction in comparison to the conventional configuration [378]. NASA's N3-X aircraft is another turboelectric concept with a blended wing body in which electric motors are used to drive several distributed electric fans, where the effective bypass ratio (BPR) is increased while reducing the fan pressure ratio [393]. It demonstrated to provide ca. 63% energy and 90% NO_x reductions [395]. This architecture has been proposed as the upcoming technology to meet environmental goals [370,396], where hydrocarbon-based fuels (biofuels/electrofuels) or H_2 can be used instead of fossil kerosene.

Other projects on electric propulsion systems are demonstrated in Fig. 18 and can be further retrieved in SM1.

11. Regulatory frameworks/ Sustainability policies

Policy and regulatory frameworks are often pinpointed as essential to encourage the deployment of SA, and to ensure this deployment is sustainable [397–399]. Alternative fuels for aviation have been considered in numerous launched/ongoing regulatory framework and sustainable policy initiatives. These essentially apply to liquid biofuels and to some extent electrofuels, owing to their technological maturity. As of today, the use of biofuels nevertheless remains minimal, with less than 1% of total aviation fuel demand [25,400,401] (electrofuels are not yet widely available for commercial use).

A handful of frameworks/policies have also been paying attention to other emerging alternatives to power aircraft, including electric (battery) aviation and hydrogen (H_2) systems. The regulatory context on alternatives implemented at the point of writing is summarized herein, with a greater focus on schemes affecting the EU countries.

11.1 European Union's sustainable & smart mobility strategy

The EU's sustainable & smart mobility strategy (part of the EU Green Deal) has been adopted by the European Commission on December 9th, 2020. Being a strategy, it is not binding, but it aims to ensure that the EU's transport systems are able to achieve the climate targets for 2030, and the 2050 climate neutrality [25], and describes the Commission's current vision to reach these goals. A package of milestones for all transport modes are proposed, covering also the aviation sector [402]. For instance, the production and deployment of biofuels and electrofuels (labelled as SAF in the strategy) are highly encouraged at Union airports to reduce aircraft's emissions in line with the ReFuelEU Aviation proposal (section 11.5). Alternative propulsion technologies powered by electricity or hydrogen are also required to achieve zero (carbon-) emissions. Other measures, such as improving the air traffic management (ATM) efficiency, developing an environmental label program, and revising the EU Emission Trading Scheme (EU ETS) with respect to aviation (section 11.4) are also mentioned [403].

11.2 'Fit for 55' package

The European Commission has proposed, as announced in the European Green Deal communication [403], to increase the 2030 GHG emission reduction target (at overall EU level) from 40% to 55%, relative to 1990 levels. To this end, the European climate-, energy- and transport-related legislation is being revised under the so-called Fit for 55 package [404], being a set of proposals to align the EU legislation with the new target. The most relevant points of this package for aviation include: i) the amendment to the Renewable Energy Directive II (RED II) (section 11.3); ii) the revision of the EU ETS (section 11.4), ii) the ReFuel EU Aviation proposal (section 11.5), and iii) the revision of the Energy Taxation Directive (section 11.6).

11.3 EU Renewable Energy Directive (RED)

The EU Renewable Energy Directive (RED) adopted in 2009 established an overall policy framework for the production and promotion of energy from renewable sources. It is a binding regulation for EU Member States. The RED requires all EU countries to ensure a share of at least 10% of final energy consumed in transportation stems from renewable sources by 2020 [405]. The RED (or RED I) target, however, does not fully include the aviation sector, by limiting, in the calculation of the gross final energy consumed by a Member State, the aviation share to maximum of 6.18% (4.12% in the specific case of Malta and Cyprus).

The RED was further amended with the Directive 2015/1513 to, among others, recognize the opportunity offered by biofuels/electrofuels to enhance the consumption of fuels not derived from fossil resources within the aviation sector. This translated in the so-called 'voluntary opt-in' [406]. The Member States could implement this opt-in differently: in form of a certificate system for fuel suppliers, or as a tax exemption for reaching the 10% of final energy consumption from renewable sources by 2020 [407].

The revised RED for the period 2021-2030, denoted as RED II, requires a minimum share of 14% of final energy consumption in transport sector to be derived from renewable energy by 2030, and is set as an obligation for fuel suppliers [71]. It also sets additional sustainability criteria, namely requirements associated to specific GHG emission savings, specific shares of advanced fuels to incorporate (in 2022, 2025, 2030), criteria for feedstock sourced form forest biomass (e.g., harvesting with legal permits, maintaining soil quality and biodiversity during the harvesting process), and the consideration of indirect land use changes (ILUC) to limit the use of high ILUC-risk feedstock from land with high carbon stocks [71,408,409], among others. The new directive further introduces a slight incentive for using non-food biomass for aviation and maritime fuels production through a multiplication factor of 1.2 (of their energy content) in the calculation of renewable energy not stemming from feedstock intended for food and feed consumption. As the original RED, RED II has no specific target for the aviation sector.

Key provisions affecting the aviation sector have been suggested in an amendment to RED II proposed by the Commission in July 2021 [410]. This includes a reduction of the minimum share of advanced fuels for 2025 and 2030, and the introduction of a new sub-category of renewable fuel, namely renewable fuels of non-biological origin (RFNBOs; including electrofuels, renewable hydrogen, renewable electricity). Besides the minimal share of advanced fuels to be supplied to the overall transport sector, the amendment proposes to also ensure a minimal share of 2.6% of RFNBO by 2030.

11.4 EU Emission Trading System (EU ETS)

The EU ETS is one of the EU's policy instrument to mitigate climate change [411], used to implement the current 40% GHG emission reduction target for 2030. It is a mandatory cap-and-trade system and covers approximately 40% of total EU GHG emissions from industrial activities, including the aviation and maritime sectors. The EU ETS aims at achieving economy-wide emission reduction targets through tradable allowances putting a price on carbon emissions.

Currently, for the aviation sector, the EU ETS covers only CO_2 tailpipe emissions. The flights operated within the European Economic Area (EEA) as intra-EEA flights (arriving at and departing from EU airports) are presently accounted in the EU ETS, while flights to and from non-EEA countries are exempted until the end of 2023 [18,412]. Overall, emissions allowances to airline operators covered by the EU ETS, from 2013-2020, are distributed as follows: 82% are granted as free allowances allocated on the basis of airlines' efficiency in transporting passengers and cargo while 15% are auctioned, for a price reaching ca. €25 for one tonne of CO_2 at the end of 2019 [413] up to about €85 in May 2021 [414]. The remaining 3% are reserved for new entrants (those starting activities after 2010) and fast-growing airlines (>18% average tonne km annual growth between 2010-2014) [415]. The cap for free allowances is annually limited to 97% of the average aviation emissions for the years 2004-2006 (approx. 210-220 million tonnes CO_2) [416,417]. It was further reduced to 95% for 2013-2020. From 2021 onwards, the cap is to be decreased by an annual linear reduction factor (LRF) of 2.2% [418,419]. For CO_2 emissions exceeding the cap, the aircraft operators have to purchase EU emission unit allowances (EUAs) at auction, or from other sectors, leading to additional costs for the airlines. CO_2 emissions are significantly dependent upon the carried weight, namely the revenue tonne kilometer (RTK) for cargo flight, the RPK for passenger-carrying flight. The EU allowances are used to fund emission-saving projects in lower-income countries, such as the development of innovative renewable energy technologies or, modernization in power sector and energy system [398,420].

The revision proposed for the EU ETS in the framework of the Fit for 55 package includes the following points relevant for aviation: (i) Free allowance distribution will be phased out progressively from 2024 (by: 25%, 50%, and 75%, in 2024, 2025 and 2026, respectively), for a complete phase-out from 2027 onwards [18]; (ii) Starting 2025, the application of a LRF of 4.2% (instead of 2.2%) is proposed to meet the more stringent 2030 emission target (revision proposed by the Fit for 55 package) [237,401,415]; (iii) The allowances will be capped at current level (instead of an average historical amount); (iv) Flights between the EU's outermost regions and flight to Switzerland and the United Kingdom (UK) will be included in the EU ETS; (v) International flights (involving CORSIA participating states; 11.10) departing from or arriving at an airport inside the EEA (extra-EEA flight) will be also be integrated [421,422].

Currently, the uptake of SAF can be an incentive for airline operators as this system counts biofuels/electrofuels (to the extent it complies with the sustainability criteria defined in the RED) as having zero emissions (no allowances required to be surrendered) [25]. However, this approach is not economically incentivized to encourage the use of SAF as their prices has remained higher than the CO_2's price (the price of at least €160 per tonne of CO_2 emissions is required). This is required to report, of the EU ETS, based on SAF purchasing records [25].

11.5 The ReFuelEU Aviation proposal

The ReFuelEU Aviation proposal is a proposal for a regulation to boost the supply and demand for SAF in the EU, by 2050. This proposal has been launched as a part of the ‘Fit for 55’ package, and was released in July 2021 after several consultation rounds and an impact assessment of the sector [25]. In the context of ReFuelEU, the term SAF specifically covers RED-compliant (section 11.3) biofuels including advanced biofuels (e.g. agricultural and forestry biomass; listed in Annex IX Part A of RED II), biofuels produced from feedstock listed in RED II Annex IX, Part B (waste oils and fats), and electrofuels (equivalent to the term synthetic fuels) [71].

The key highlight of the current proposal version is an obligation for fuel suppliers to ensure that the aviation fuel made available at EU airports contains a minimum share of SAF (from 2 vol.% in 2023 to 63 vol.% in 2050), including a minimum share of electrofuels (0.7 vol.% of overall SAF in 2030 to 28 vol.% in 2050) [25]. The European Parliament has recently endorsed the ReFuel EU proposal, and proposed to raise the SAF share from 63% to 85% by 2050 with the inclusion of electricity and hydrogen in the overall (sustainable) aviation fuel mix [423].

However, until 2035, fuel suppliers will be allowed to supply this minimum share of SAF to airportsthrough the trading of SAF certificates obtained by overachievers (suppliers with an excess of SAF) [424]. To mitigate the effects of tankering (the use, prior to enter the EU, of more fuel than required for the flight in order to reduce the amount of SAF to be used, and thus save on fuel costs), ReFuelEU proposes a requirement that EU and non-EU airlines, for all flights departing from the EU, tank at least the whole required amount for their subsequent flight. The proposal also includes an obligation for EU airports to provide the infrastructure required, including hydrogen refueling, electric recharging, to reach the above-mentioned SAF supply targets. Additionally, a Sustainable Aviation Fund is proposed by the Parliament in order to facilitate the establishment of cleaner aviation, including an investment support in sustainable fuels, and in research on innovative propulsion systems [423].

Non-compliance penalties according to the Commission’s criteria will be proposed by the Member State authorities and are to be communicated to the Commission by the end of 2023 [424].

11.6 The Energy Taxation Directive (ETD)

The Energy Taxation Directive is a European directive in order to support the EU’s climate and energy policy frameworks through a taxation system of energy products used as motor fuels, heating fuels, and electricity [425,426].

A tax exemption for (conventional) aviation fuels granted by the ETD for intra-EEA flights to be abolished in the revision of ETD, as part of the ‘Fit for 55’ package. The revised ETD will raise the minimum tax rate for fossil kerosene for intra-EEA flights: starting at zero from 2023 onwards and to be increased gradually (annual rate of 10%) over a transition period of ten years, until the rate of €10.75 GJ^{-1} is reached [425,426].

The revised taxation for aviation fuels proposes to consider energy efficiency and environmental impacts [25]. Therefore, the reduction of tax rates or exemption for SAF as well as renewable hydrogen (i.e., minimum tax rate of zero) contribute to make these fuels more economically interesting to airlines, relative to fossil jet fuels, and foster their uptake [25,427]. This strategy would work coherently with the ReFuelEU Aviation proposal to encourage the necessary investment in SAF production and their deployments. These minimum tax rates would not be applicable for pleasure flights and business flights [427].

11.7 EU climate law

In the framework of the European Green Deal, the EU launched its first proposal for a Climate Law [428]. This law makes it legally binding for the EU to achieve a balance between GHG emissions and emissions removals (so-called neutrality) by 2050. The current proposal of the Climate Law covers all GHGs. As it stands now, there are no clear measures specifically applying to the aviation sector. The updated GHG reduction target (details in section 11.2) along with the climate neutrality target by 2050 is currently in the process of being enshrined into the EU Climate Law [415,428]. For aviation, one consequence of the EU Climate Law could be the quicker deployment of DAC technologies for electrofuels and carbon-free technologies (e.g. batteries, hydrogen), on the premise that decarbonized renewable energy can be used.

11.8 The European Advanced Biofuels Flightpath Initiative (EABFI)

The EABFI was launched in 2011 as a partnership between the European Commission and major European stakeholders including airlines and biofuel producers. The objective is to promote the commercialization of biofuels in terms of production, storage and distribution in an endeavor to support the European Commission's ambition to reach energy security [429]. Concretely, this translates in the objective of reaching 2 million tonnes of biofuel consumption per annum by 2020 through the construction of advanced biofuels production plants in Europe. The EABFI is a shared and voluntary commitment to promote the biofuel deployment through appropriate financial mechanisms [429]. However, its 2020 goal has not been met. The EABFI is working on an updated roadmap towards 2030 [429,430].

11.9 The French sustainable aviation targets as an example of national initiative

France was one of the first country to announce the ambition of making its aviation industry "the cleanest in the world".

At the end of 2017, France planned to facilitate the production, distribution and deployment for aviation biofuels, corresponding to a "Commitment to Green Growth" with five industrial partnerships including Air France, Airbus, Safran, Total and Suez Environment [431]. The intention, as stated in the French "National Low Carbon Strategy" of March 2019 is the deployment of 2% and 5% biofuels of the expected gross demand of the aviation fuel in 2025 and 2030, respectively [432]. Accordingly, aviation biofuels should be produced from resources listed in Annex IX of the EU RED II [71]. The HEFA from WCO, being a mature technology, is the pathway that received the most focus. By 2050, it is intended that alternative liquid fuels from other advanced pathways (both aviation biofuels and electrofuels) substitute 50% of conventional kerosene [432].

In addition, in early 2020, French government officials announced a 15 billion euros recovery plan for the aeronautical industry, including 1.5 billion euros aid to support research and development into cleaner aviation technologies, for instance, the improvement of engine efficiency with a 30% reduction in fuel consumption for the early 2030s, relative to the current engine, or the development of carbon-emission free aircraft whether electric or hydrogen powered by 2035 [433–435]. Investments to transit towards electric and hydrogen aviation have been announced, along with investments in the advancement of biofuel/electrofuel production pathways in order to reduce GHG emissions [433].

11.10 Carbon Offsetting and Reduction Scheme for International Aviation (CORSIA)

The Carbon Offsetting and Reduction Scheme for International Aviation (CORSIA) was launched in 2016 by ICAO in the endeavor of meeting international goals in terms of GHG mitigation. The aviation industry organizations including IATA and ATAG have set goals of CO_2 emission reductions by 50% in 2050 (relative to 2005 level). It covers all states that are ICAO members. CORSIA aims to offset the emission from international aviation that is not covered under the Paris Agreement. The aircraft operators with emissions greater than 10,000 tonnes CO_2 from the use of aircraft with a maximum certified take-off mass greater than 5,700 kg need to prepare emission monitoring plans and emission reports for all their international flights on an annual basis, from 1 January 2019.

The total CO_2 emissions of each operator are required to calculate in order to demonstrate the offsetting requirements [436,437]. The aircraft operators will then be required to purchase emission units labelled as "carbon offsets" from the Aviation Carbon Exchange (ACE), in order to offset the CO_2 emissions exceeding the defined baseline [438]. These carbon offsets are credits in certified projects that reduce carbon emissions.

Aircraft technology development, operational improvement and deployment of biofuels can be used to achieve their CO_2 offsetting requirements. The sustainability criteria for alternative aviation fuels has been developed under CORSIA Eligible Fuels, which will support the maximum use of biofuels and long-term investment in their productions [439].

ICAO's CORSIA is being implemented since 2019. The compensation phase thus begins in 2021. This market-based measure encouraged the airlines and other aircraft operators to use SAF. CORSIA SAF are defined in CORSIA

Eligible Fuels [70] (SAF in CORSIA corresponds to biofuels in this review). The objective of CORSIA is to reach carbon neutral growth onwards in the aviation sector, in an endeavor to stabilize the net CO_2 emissions from international aviation [70]. CORSIA is composed of three implementation phases: the pilot phase (2021-2023), a first phase (2024-2026) and a second phase (2027-2035). Year 2019 is considered as the baseline emissions for the pilot phase instead of the first proposed baseline of average 2019-2020 due to the pandemic's impact on the international aviation emissions. The baseline for the first-phase is still under discussion [421,440]. During the pilot and first phases, the offsetting requirements will be applicable for ICAO member states that have volunteered to participate in the scheme. A total of 81 states have officially participated in the pilot phase, representing approximately 76% of international aviation activities in terms of RTK [398]. The second phase is legally binding for all ICAO member states, with the exception of least developed countries and, states with small share of international traffic (less than 0.5% of air traffic), unless they volunteer to participate.

ICAO has launched a detailed requirement for the monitoring, reporting and verification (MRV) of emissions for the CORSIA scheme [439]. It proposes default life cycle assessment (LCA) emission values for five (biomass-based) certified production pathways, these being backed up by a detailed methodological study [439]. However, the calculated default life cycle emission for what CORSIA defines as lower carbon aviation fuels (here referred as electrofuels) and the latest certified CHJ pathway have not been announced at the time of writing.

12. Challenges for Sustainable Aviation (SA) systems

Some challenges for the use of the SA approaches detailed in this review, in the future low GHG emissions, are summarized in Fig. 13 and discussed below.

12.1 Availability of sustainable biomass feedstock

In the perspective of a sustainable transition towards GHG neutral economies, the procurement of sustainable feedstock not inducing additional arable land demand is a key concern when it comes to biofuels [441], including for aviation biofuels [330,398]. In this context, residual biomasses generated increased attention as they can be decoupled from the need for additional arable land (e.g. [119,442,443]). Residual biomasses have the potential to feed the future low fossil carbon aviation, and several examples have been documented and even show-cased (Table 4). Numerous aviation biofuel producers such as LanzaTech or Neste have adapted their technologies to flexibly incorporate residual biomasses (Table S2).

Table 4 Documented examples of residual biomasses used in the production of aviation fuels

Feedstock	**Residues/wastes**	**Conversion pathway**	**Comments**	**References**
Carbohydrate	Molasses	SuF	• Co-product from sugar production, albeit already sold as an ingredient for the feed industry	[97]
	Food waste	SuF	• Organic waste from food processing industries, restaurants, and household	[99,266]
	MSW[1]	FT, IH[2]	• Organic portion of household waste	[156,174]
	Anaerobic sludge (digestate)	HDCJ	• Digestate from anaerobic digesters • The digestate is used as an input for bio-crude oil production by HTL[1], subsequently upgraded to kerosene	[165]
	Algae biomass	HDCJ	• Grown in wastewater • For bio-crude oil production by HTL, subsequently upgraded to kerosene	[444]
	Swine manure	HDCJ	• For bio-crude oil production by fast pyrolysis, subsequently upgraded to kerosene	[159]
Lignocellulose	Forestry waste	SuF, IH[2] FT	• Pretreatment requirement • Gasification and syngas conditioning requirements	[98,157,174,266]
	Wheat/rice straw	SuF DSHC	• Pretreatment requirement • Pretreatment and hydrolysis requirements	[266,445]

	Maize stover	SF, FT DSHC APR	• Gasification and syngas conditioning requirements • Pretreatment and hydrolysis requirements • Pretreatment and hydrolysis requirement	[91,132,446]
	Woody chips	SF, FT DSHC APR	• Gasification and syngas conditioning requirements • Pretreatment and hydrolysis requirement • Pretreatment and hydrolysis requirement	[95,132,447]
	Bagasse	SF, DSHC	• Gasification and syngas conditioning requirements • Pretreatment and hydrolysis requirements	[95,130]
	Eucalyptus tips	FT	• Gasification and syngas conditioning requirements	[147]
	Wood sawdust	HDCJ	• For bio-crude oil production by HTL, subsequently upgraded to kerosene	[168]
	Maize stalk	HDCJ	• Pretreatment requirement • For bio-crude oil production by HTL, subsequently upgraded to kerosene	[166]
	Straw stalk	HDCJ	• For bio-crude oil production by fast pyrolysis, subsequently upgraded to kerosene	[448]
Oil/Fat	Waste cooking oil (WCO)	HEFA CHJ	• Collected from restaurants, food processing industries	[196,449,450]
	Non-edible sunflower oil	HEFA	• Extracted from sunflower residual wastes	[184]
	Tall oil pitch	HEFA	• Residue from the distillation of tall oil	[248]
	Animal fats	HEFA	• Slaughterhouse waste	[248]
	Palm fatty acid distillate	HEFA	• Co-product of palm oil production	[248]
	Brown grease	CHJ	• Derived from grease trap waste	[196]

Note. [1] MSW: Municipal Solid Waste; HTL: Hydrothermal liquefaction

A key question remains whether the residual biomass potential is large enough to supply the demand. We estimated, on the basis of the ICAO projections [451], for 2045, a global aviation fuel demand of 16.73 EJ y^{-1} (ca. 390 Mt kerosene y^{-1}). In comparison, it was 7.62 EJ y^{-1} in 2019 (details in SM2). In the hypothetical case that this future demand should be fully supplied by a biofuel pathway (i.e. disregarding the ASTM D7566 standard on aromatics), we evaluated, considering the FT-SPK pathway as a technology capable to process most residual biomasses, that at least 46 EJ y^{-1} of biomass feedstock is required (assuming 0.183 kg biomass (dry matter) MJ^{-1} FT-SPK and 15 GJ t^{-1} biomass; details in SM2). This alone represents ca. 23%of the global residual biomass potential (being at maximum ca. 200 EJ y^{-1} based on the meta-study of [117]). Yet, the transition towards GHG neutral economies implies other demands for this limited potential. One example is the plastic sector; based on a forecasted demand of 1,124 Mt plastics for 2050 from [452], and considering a biomass:bioplastic ratio of 1.26 [453], a rough biomass demand of ca. 20 EJ y^{-1} can be derived if future plastics are to be bio-based (details in SM1). Similarly, supplying 100% bio-based maritime fuels would, considering the 2050 projected demand of 9.50 EJ y^{-1} [332,333], also require at least 20 EJ biomass y^{-1} (estimation based on supplying 9.50 EJ y^{-1} with 100% bio-based LNG, itself deriving from the anaerobic digestion of straw biomass; details in SM1). This rough yet illustrative back of the envelope estimation of the productions illustrates the eventual pressure on the residual biomass potential, where only 3 demands already mobilizes ca. half of the potential.

Further, while some biofuel pathways (e.g., the FT and IH^2) can use most types of residual biomasses, other pathways are less flexible, such as the HEFA pathway requiring oil feedstock. At present, aviation biofuels are mainly obtained from the HEFA pathway [248]. This pathway has the advantage of having a greater energy yield in comparison to other biofuels pathways (an overall energy efficiency of ca. 75% for HEFA in comparison to 40% for FT-SPK; [44,454]). The current production capacity of HEFA is reaching ca. 100,000 tonnes SPK y^{-1} (0.1 Mt SPK y^{-1}) (in 2019; reported by Neste) derives from dedicated oilseed crops and a variety of oil/fat residues [248]. One question is to which extent this new capacity will be based upon waste feedstock versus dedicated oilseed crops. Moreover, basing new investment strategies upon food waste poses the risk of rebound effects encouraging whether the generation of waste, or inducing unforeseen additional demand for the most competitive oil feedstock (often identified as palm oil, e.g. [443]) if no waste oil can be supplied. Globally, the potential of waste fats, oils, and greases has been estimated to ca. 1 EJ y^{-1} [455]. The current 100,000 tonnes HEFA-SPK y^{-1} capacity mobilizes ca. 0.02 EJ waste oil and fat. For illustration purposes, supplying the 2045 aviation demand of 16.73 EJ y^{-1} by 100% HEFA-SPK would require 77.41 EJ waste oil and fats (details in SM1). Despite being an unrealistic scenario, it illustrates that the current potential could only supply 1% of the future demand. On the other hand, competing bioeconomy sectors (e.g. bio-based polypropylene; [456]) also aspire to use these fat waste resources.

12.2 Uncertain deployment of DAC

Liquid fuel pathways (biofuels, electrofuels) have the advantage of not requiring heavy infrastructure changes in comparison to the other options discussed in this review. These imply two sources of carbon: biomass and captured-C, either from the atmosphere through DAC or from industrial point sources. While the former and the latter are limited, the potential of DAC is theoretically very large, to the extent the technology is deployed.

There are currently 15 DAC plants operating worldwide, capturing more than 9,000 tonnes CO_2 y^{-1} (0.009 Mt CO_2 y^{-1}) [30,223]. To produce 16.73 EJ (2045 demand) of electrofuels (FT pathway), approximately 1,200 Mt CO_2 captured would be needed, based on the stoichiometry described in the FT reaction (details in SM2). Keith et al.[30] provide the design and engineering costs for a plausible advanced DAC plant to be implemented at industrial scale, capturing 1 Mt CO_2 y^{-1} when operated at full capacity. Taking the plant of Keith et al. as a basis, it implies that ca. 1,200 DAC plants would need to operate to supply the carbon needed for future aviation demand by biomass-free FT electrofuels. Yet, the costs of DAC [457] are often pinpointed as a barrier for massive deployment [458,459], along with uncertainties on the markets for CO_2 to ensure a revenue offsetting the costs of capture. Keith et al. [30] report, for the 1 Mt CO_2 y^{-1} industrial plant they describe, current levelized costs ranging from US$ 94-232 t^{-1} CO_2 (range reflecting different technology choices), while other studies report that levelized costs below €50 t^{-1} CO_2 are achievable by mid-century [460,461]. For comparison, the first commercial-scale DAC plant built in 2017 (with storage of the captured carbon) costed US$ 600 t^{-1} CO_2, foreseen to decrease to US$ 200 t^{-1} CO_2 as additional plants are built [462]. It is further argued that DAC costs are minor when reported to a country gross domestic product (GDP) [463], or to global GDP (an emergency massive DAC deployment would imply an investment of 1.2-1.9% global annual GDP; [464]).

Nevertheless, DAC technologies are improving and maturing, reflecting among others their vital role in stabilizing warming at 1.5°C above pre-industrial levels [465–467]. On-going reported improvements include new contactors and tower designs [457,468], optimized operating conditions (e.g. kinetics stability, process stability; [468]), new materials development (e.g. composites of potassium carbonate and γ-Al_2O_3; [457], amine-oxide hybrid materials; [469]), or alternative regeneration processes (e.g. electrochemistry; [470]).

Assuming a favorable techno-economic environment, the potential of DAC is theoretically unlimited. Potentials up to 40 Gt CO_2 y^{-1} (40,000 Mt CO_2 y^{-1}) by the end of the century have been reported [471] for global warming mitigation, albeit the meta-study of Fuss et al. [462] suggests a potential limited to 0.5-5 Gt CO_2 y^{-1} (500-5,000 Mt CO_2 y^{-1}). For comparison, the global annual CO_2 fossil emissions were ca. 36 Gt CO_2 (36,000 Mt CO_2) in 2019 [472].

However, it must be highlighted that the driver for DAC is the perspective of inducing so-called negative emissions, i.e., a net long-term sequestration (or removal) of atmospheric CO_2. The use of the captured CO_2 for fuels, as proposed for aviation electrofuels, is therefore deriving the technology from its original purpose.

12.3 Need for sustainable hydrogen

All pathways described herein involve hydrogen, with the exception of electric (battery-based) SA , unless part of a hybridized system involving a share of hydrogen fuel or liquid biofuels or electrofuels. Hydrogen is used whether for hydrogenation of aviation biofuels (Figs. 2-10), electrofuels (Fig. 11), as well as for fossil-based kerosene production, although more H_2 is used for biofuels than fossil kerosene (3 to 75-fold more, depending on the pathway; [473]). Yet, some alternatives will require significantly more H_2, namely electrofuels and LH_2 pathways.

Electrofuels, in particular, require H_2 for the reverse water gas shift reaction, where 12 moles of CO_2 are reacted with 12 moles of H_2 to produce 12 moles of CO (Eq. 3 and its adjustment in SM1). This CO is then reacted with another

25 moles of H_2 in order to generate for 1 mole of $C_{12}H_{26}$ fuel via the FT process. This involves that approximately 169 Mt H_2 is required to produce 16.73 EJ (2045 demand) with this pathway (details in SM2), involving 1,520 Mt of water (equivalent to 1.52 billion m^3). This clearly exceeds the current capacity of H_2 produced by water electrolysis (around 2% global H_2 production), which amounts to 1.4 Mt H_2 y^{-1} [473]. Furthermore, the amount of water needed is not negligible either, representing ca. 10% of Australia's freshwater withdrawals (ca. 15.95 billion m^3 in the year 2017; [474]). This also applies for electricity, as detailed in 12.4.

Albeit not discussed in this study, hydrogen can also stem from biomass-based hydrocarbons through a gasification process, a process that can also be applied to fossil resources (e.g. coal) [475]. This, however, implies an additional demand for limited biomass and land resources as discussed in 12.1. Renewable H_2 can also be produced through biogas/biomethane reformation (instead of natural gas) [63], which again implies increasing the pressure on biomass residues to produce the biogas.

Hydrogen fuels (LH_2), on the other hand, are not dependent upon a carbon intermediate, and therefore require slightly less hydrogen when reported by MJ of fuel (details in SM2).

12.4 Need for sustainable electricity

Several of the emerging pathways presented in this review involve important electricity requirements, in particular for electrofuels. Electricity consumption for DAC alone may need between 0.23 and 0.37 kWh kg^{-1} CO_2 captured (0.83 and 1.33MJ kg^{-1} CO_2 captured) [30,31,476]. Water electrolysis, on the other hand, requires 30-80 kWh kg^{-1} H_2 (around 110-290 MJ kg^{-1} H_2) (Table S4), depending on the technologies. To produce the 16.73 EJ demand in 2045 with current technologies for electrofuels, an estimated range of 5,900-10,000 TWh electricity (ca. 21-36 EJ) is needed (i.e. 0.55-0.74 kWh electricity MJ^{-1} electrofuel produced; 1.81-2.66 MJ MJ^{-1} electrofuel produced), depending on the DAC and electrolysis technology considered (detailed calculations in SM2). This illustrates, for electricity alone, that 25%-140% more energy is required compared to what is obtained from the produced electrofuel, which only makes sense in the perspective of harnessing the full potential of fluctuating decarbonized energy sources. Independently of the electrolysis-DAC technological combination selected, water electrolysis always represents ca. 90% of the consumption; the electricity consumption for DAC thus appears negligible in comparison.

The 0.55-0.74 kWh MJ^{-1} electrofuel derived herein lies in the wide range of estimates found in the literature (0.59-1.00 kWh MJ^{-1} electrofuel; 2.12-3.60 MJ MJ^{-1} electrofuel, details in SM2). It should also be highlighted that 5,900-10,000 TWh electricity y^{-1} (21-36 EJ electricity y^{-1}), i.e. the electricity requirement to produce 16.73 EJ electrofuels, is not a negligible quantity. To put it into perspective, the global electricity consumption in 2019 was about 23,000 TWh (ca. 82.8 EJ) [477]. In other words, the equivalent of 25%-43% of today's global electricity consumption would be required to supply the electricity needed just for producing the H_2 necessary for a demand of 16.73 EJ of electrofuels per year. Similarly, supplying 16.73 EJ of fuels with LH_2 would require only ca. 5,500-9,000 TWh electricity (20-32 EJ electricity), including the liquefaction requirements for ground storage (details in SM1 and SM2). It should be highlighted that in both cases, the lower end of interval is due to the use of SOE technology, based on high-temperature water splitting rather than electricity. This technology requires, per unit of H_2 produced, ca. 40% less electricity than alkaline electrolysis, but 40% more heat (SM2).

12.5 Limits of current quantification methods for environmental impacts

To compare the different emerging technologies covered in this study, it must be ensured that the same service is supplied (the amounts of passengers and freight transported over a given distance and time). These may require different number of aircraft (e.g. smaller electric planes), type of aircraft (e.g. to accommodate onboard hydrogen storage systems [57]); infrastructure (e.g. charging), blending with fossil kerosene, etc. Current comparative studies assessing the environmental impacts of alternative fuels often only compare impacts per MJ fuel (e.g. [478–480]), which cannot be applied for SA that are not purely based upon liquid fuels. Further, the requirements for blending with fossil kerosene are typically completely ignored (e.g. [478,479,481]).

Another key issue is the evaluation of the global warming potential related to the non-CO_2 emissions generated during propulsion. Pollutants such as NO_x and particulate matter (including black carbon) serve as precursors for contrail formation, an important climate forcer [68,348]. These are generally ignored in current life cycle assessment studies (e.g., [115,327,482]). Yet, Lee et al. [348] illustrated that the greatest share of cumulative radiative forcing from global aviation between 2000 and 2018 is due to these non-CO_2 effects, and in particular on the radiative forcing effect of NO_x, and to a smaller extent to the aviation induced cloudiness (including cirrus induced by condensation contrails). In the perspective of life cycle assessments, this implies two issues. First, there is a need to quantify these emissions, and second to translate these into warming effect (and eventual other environmental impacts), often through characterization factors.

To date, the most robust method available for the former is to use emission factors relating to the amount of fuel use (e.g., in g of pollutant per g of fuel burnt), which can be found in the literature for all substances including contrails [348,483–485]. Yet, non-CO_2 emissions are not necessarily due to the fuel itself, and are also highly dependent upon the location where they are emitted. Emissions of NO_x, for instance, can be considered as independent from the fuel chemical composition [486], stemming from the nitrogen in the air. Similarly, aviation induced cloudiness is highly influenced by the water vapor saturation of the air where the emission takes place, and by the aerosols (e.g., sulfate, soot) emitted during combustion.

Moreover, there is no consensus on the characterization factors to use to translate NO_x, induced cloudiness, water vapor, black carbon and sulfate to a warming effect. Although recent studies tend to refer to the factors proposed by Lee et al. [348], the differences in proposed factors are really large. For aircraft NO_x for example, the effect will depend not only on the emission altitude but also on the latitude as discussed by Fuglestvedt et al. [483]. A GWP_{100} between -2.1 to 71 can be obtained following the modeling approach. Köhler et al. [487] proposed a GWP_{100} of 75 at the global scale with important variations according to the region of emission. Other methods have also been proposed to translate these non-CO_2 substances into a warming effect, such as the global temperature change potential (GTP), a measure of the temperature change (rather than heat absorbed) over a given time period due to the emission of a gas, in comparison to CO_2 [483,488,489]. Step-change emission approaches (e.g., GWP*, Combined GTP; CGTP) [340] which are applied to a change in emission rate rather than a change in emission amount (as for GWP and GTP) [490], should also be highlighted as existing alternative approaches.

Accounting for non-CO_2 climate forcers will impact the hydrogen-based SA approaches. The hydrogen used in H_2 internal combustion engines or fuel cells, can generate approximately 2.6 times more water emissions relative to conventional kerosene [58,491]. Lee et al. [348] proposed a GWP_{100} of 0.06 for water vapor. Despite useful, this may however not entirely reflect the exact effect that massively deploying hydrogen aviation would have on the radiative equilibrium of water vapor in the low stratosphere [492].

Finally, reductions of soot and particulate matter (PM) have been reported for alternative liquid fuels, in particular biofuels (e.g., HEFA-SPK, FT-SPK) [39,40,486]. This reduction could be explained by the low levels of aromatic content in biofuels [40,486,493]. Some studies, however, consider these differences as negligible (e.g., [327]), while others shed light on the high level of uncertainty still existing regarding these measurements in test rig [486,494]. It thus remains unclear whether biofuels (and electrofuels) really induce a reduction in particulate matters, and the magnitude such reduction could have.

12.6 Other impacts

Noise is another impact of aviation especially for residential developments around airports. Novel propulsion systems including battery-electric and hydrogen aviation are foreseen to mitigate and minimize noise impact [58,495,496], facilitating their use in densely populated areas [330,496]. The electric propulsion system, magniX eBeaver aircraft, for instance, has been reported to reduce the average noise level by about 22% (~66 dBA) relative to standard Beaver with piston engines (~85 dBA) [497].

Electric (battery) and H_2 aviation will induce the need for additional infrastructure, among other to charge batteries or store hydrogen. The recharging time, which is mainly dependent upon the charging point (e.g. power outlet capacity) and the battery capacity, is one key challenge of electric aviation to meet desired flight turnaround times.

Similarly, hydrogen refueling stations should be developed with important flow rates to maintain flight turnaround times and prevent hydrogen boil-off issues [58,63]. According to [387], onsite hydrogen production (through water electrolysis) may be possible in a foreseeable future to produce the needed hydrogen on-demand. Cryogenic storage (whether on ground or onboard), which implies the storage of liquefied hydrogen below -253°C (20K) in a double-walled vessel with vacuum insulation [57], implies the need for reliable components including valves, and pumps allowing to ensure these cryogenic conditions as well as safety requirements [498].

13. Prospects for Sustainable Aviation (SA) systems

Despite the potential of the four large SAapproaches described herein, there are a number of issues to be addressed prior to broad adoption. This section provides the prospects of their applications for sustainable aviation in the upcoming future. Other sustainable routes are also discussed.

13.1 Future aspects for unblended biofuels/electrofuels

All pathways documented in this study require blending with fossil carbon with exception of hydrogen aviation and 100% electric concepts (either 100% battery as shown in Fig. 17a, or hybrid LH_2 or fuel cell with battery, as in Figs. 17b to 17d). The prospects for alternative hydrocarbon-based fuels (biofuels/electrofuels) is summarized herein.

One of the key limiting factors of liquid fuels free of fossil carbon is their low fuel density (below ASTM's minimum specific requirement of 775 kg m^{-3} [499]) and their low (<8%) aromatics content [44,500], which shrinks engine seals. The latter effect is notably observed with the commonly used nitrile rubber material [499,501]. To overcome this, a certain research focus has been placed on the development of new sealing materials preventing leakages [499,502].

The use of 100% alternative liquid fuels is also being investigated through the possibility of combining SAK with HEFA-SPK [503]. SAK is produced through the APR pathway (Fig. 2) by converting oxygenated compounds to aromatics in the presence of heterogeneous catalysts such as the aluminosilicate zeolite ZSM-5 [504], a pathway currently under the ASTM approval process [26].

Besides aromatics, cycloparaffins (e.g. cyclohexane, cyclooctane), at a concentration above 30 vol.%, have been shown to supply suitable volume swelling properties [505,506]. They provide increased fuel density, similar to the function of aromatics. Furthermore, they have high gravimetric heat of combustion with the respect to aromatics, resulting in reduction of particulate matter emissions from fuels containing aromatics [21,506]. These compounds, that could be blended in the fuel, have been produced through multiple pathways and from numerous renewable feedstock, for instance through hydrodeoxygenation of lignocellulosic biomass [507,508]. This approach can be introduced as near-term solution for current aircraft, before new engines capable of handling unblended SPK are widely available.

Another investigated solution is, as earlier reported, the development of alternative fuels identical to conventional kerosene (synthetic kerosene; SK) that could be used without blending. This includes the recently certified CHJ-SK (in February 2020; Fig. 10), as well as some biofuels currently under certification (e.g. ATJ-SK from the SuF pathway, $HEFA^+$) [26,38]. It is important to note that there is currently no guideline for blending more than one alternative fuel with conventional kerosene (or several alternative fuels without conventional kerosene) within the same hydrant system [509].

Finally, new engine generations capable of using low aromatic fuels may be another solution. For example, the D328neo aircraft manufactured by Deutsche Aircraft Inc. has been publicized in September 2021, to be compatible with unblended alternative fuels by the use of new sealants, surface protection measures, and materials. It is a sub 50 seater regional commuter expected to start its test flight in 2024 and to enter into service by the end of 2025 [48,510].

13.2 Electric propulsion development

Motors and generators are key elements for parallel hybrid electric and turboelectric configurations [388] (Fig. 17), with additional converters in the case of electric configurations, whether from battery or fuel cell. Further research for the high power-to-weight ratio of electric components is essential to be able to fit with the aircraft weight and volume constraints [390]. A thermal management system, in particular the removal of the generated waste heat, is another key challenge for further research [388]. Alongside, certification standards must be developed to assure the safety and use of batteries (or fuel cells).

High power density fuel cells need to be developed to attain the power requested in propulsion system, and to improve the weight and volume constraints in commercial aircraft. Current power density of fuel cells is ca. 0.75 kW kg^{-1}, whereas 2 kW kg^{-1} would be required for commercial aircraft [57,63,387].

Another key development is the optimization of the aircraft configuration, in order to optimally integrate the electrical propulsion system and batteries into the aircraft [277], which in turn is dependent on the electric propulsion configuration (Fig. 17). This is also studied in combination with alternative design approaches to improve the overall propulsion efficiency (e.g. boundary layer ingestion, wingtip propulsors, BWB aircraft) [511].

13.3 Further improvement of hydrogen aviation

Albeit hydrogen can be used as standalone fuel, the integration of either fuel cells or hydrogen combustion engines (whether gas turbines or piston engines) in airplanes remains an engineering challenge. Among others, novel airframes such as the BWB design may present the advantage of having extra volume to accommodate onboard LH_2 storage systems [58,63,313]. In addition to engine developments, airframe designs are required for accommodating cryogenic hydrogen storage, which is beyond the current aircraft capacities. Hydrogen tanks can be placed either inside or outside the fuselage (referred to as integral or non-integral, respectively) [58,512]. Drag penalty can be reduced with the integral method [58] (hydrogen storage inside the fuselage), which may have an impact on overhead luggage storage for short-to-medium ranged airplanes [498]. Placed outside the fuselage, the passenger-carrying capacity remains unchanged, however, the drag is likely to be increased.

Development in new materials (e.g. polymer matrix composites; [513]) for hydrogen tanks is needed in order to make these as light as possible. With the newly developed materials, the gravimetric energy density is expected to reach 10-21 kWh kg^{-1} (around 36-76 MJ kg^{-1}) of empty storage system [62,63]. If these values are achieved, the system becomes highly competitive with conventional storage systems (ca. 8.9 kWh kg^{-1}; equivalent to ca. 32 MJ kg^{-1}) [514]. Additionally, developments to improve the insulation system for cryogenic LH_2 allowing to minimize boil-off losses (e.g. vaporization) has been demonstrated [512].

As for electric (battery) aviation, developments are needed to optimize the integration of propulsion system and hydrogen storage system, as well as for certifications and regulations ensuring safety through the overall supply chain.

The optimal hybridization between fuel cells and hydrogen combustion engines at the different phases of the flight (i.e. takeoff and climb phase powered by hydrogen turbines; cruise powered by fuel cells) is also being investigated in order to reduce fuel consumption and NO_x emissions [63].

Similarly, improvements are being performed on hydrogen internal combustion system to reduce NO_x emission. For instance, technological and environmental improvements have been obtained with a micro-mix combustor [63,396] or lean direct injection [515].

13.4 Other innovations towards sustainable aviation

One notable innovation is the recent AHEAD project, proposing a Multi-Fuel Blended Wing Body (MFBWB) aircraft with hybrid turbofan engines, capable of using both a cryogenic fuel (LH_2 and LNG are specifically mentioned), and a liquid fuel such as biofuel or conventional kerosene. While the BWB is expected to overcome the mass issues related to the cryogenic system, the multi-fuels approach is proposed in order to significantly reduce the overall emissions from engines [342]. A reduction of 50% CO_2 emission (in LNG/conventional kerosene mode) was shown compared to a traditional Boeing 777-200 LR model using fossil kerosene [313,342]. Emissions of NO_x, soot, and CO were also mitigated by ca. 80% relative to the baseline B777-200 ER aircraft [342].

The Taxibot is another example of innovation to reduce the environmental impacts of the aviation sector. It is a pilot-controlled towing vehicle applied for assisting taxiing-aircraft between the terminals and the runway and vice versa while the aircraft's engines are switched off [516,517]. It is an emerging pushback approach to minimize aircraft fuel consumption (by 50 to 85% reduction) and noise (by 60% reduction) while taxiing [58,516]. Approximately 20 million tonnes of CO_2 emission of the world's airliners are estimated to be reduced during the taxi phase of flights if the TaxiBot is adopted [516]. The Taxibot is in operation at Schiphol [518], Delhi and Bangalore airports [519], and serves Lufthansa [517]. Currently, it is powered whether by electric or diesel engines [518], but the vision is to power them with 100% batteries or hydrogen in the future [21,520].

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.

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Author contributions

Pimchanok Su-ungkavatin: Conceptualization, Visualization, Data curation, Formal analysis, Investigation, Methodology, Original draft preparation, Review & editing. **Ligia Tiruta-Barna:** Supervision, Writing – review & editing. **Lorie Hamelin:** Conceptualization, Funding acquisition, Investigation, Resources, Supervision, Writing – Review & editing.

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