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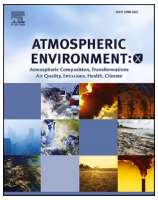

Hydroprocessing of fossil fuel-based aviation kerosene – Technology options and climate impact mitigation potentials

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ABSTRACT

Aviation contributes about 4 % of global anthropogenic climate forcing primarily by contrails, CO_2 and NO_x emissions. Renewably sourced aviation kerosene can help to reduce the climate impact from CO_2 and from contrails, but so far, its production capacities are very small. Hence, the climate impact of using fossil fuel-based kerosene with a hydrogen content increased by hydroprocessing as short term mitigation measure is studied here. Therefore, the change in net energy forcing (ΔEF_{net}) in 2019 is calculated as the sum of the change in contrail energy forcing ($\Delta EF_{contrail}$) and additional CO_2 emissions ($\Delta EF_{hydroprocessing}$) from aviation kerosene hydroprocessing ($\Delta EF_{net} = \Delta EF_{contrail} + \Delta EF_{hydroprocessing}$). The results show that hydroprocessed aviation kerosene can reduce the net energy forcing EF_{net} by about 33 % with $\Delta EF_{hydroprocessing}$ penalty of 5 %-points. Increasing the hydroprocessing severity increases the relative climate benefit, which is only slightly affected by the emissions factor for hydroprocessing or the choice of the time horizon. Data limitations about fuel composition and its effect on contrails and climate cause considerable uncertainties and the fuel's compliance with specification standards needs consideration. This study on the climate effect of hydroprocessed fossil kerosene can help to assess near-term measures to reduce the climate impact from aviation.

1. Introduction

Today, the contribution of the air transport sector to global anthropogenic climate forcing is estimated to be 3.5–4 %. This climate impact is caused by the release of greenhouse gases (GHG), mainly CO_2, and several other effects not directly related to CO_2. CO_2 presently accounts for roughly one-third of aviation's effective radiative forcing (ERF) (Lee et al., 2009, 2021), the remaining two-thirds consist of contrail effects, indirect effects of NO_x emissions, and direct and indirect aerosol effects (by decreasing order of magnitude) (Lee et al., 2021). The ERF of contrails has been estimated as 58 (17–98) mW/m^2, corresponding to 57 % of the overall aviation-related ERF until 2018 (Lee et al., 2009, 2021).

In the decades before the Covid-19 pandemic, the global air traffic volume grew by 3–4 %/year. Despite the disruptions due to the pandemic (Schumann et al., 2021a), a return to pre-pandemic growth rates is expected in the years to come (ICAO). Expected growth rates of international commercial air traffic by around 4 %/year do overcompensate reductions of passenger-specific fuel consumption by about 1.3 %/year (Grewe et al., 2021). As a result, if these rates remain constant, global aviation growth will continue to outpace efficiency gains; i.e., the worldwide commercial aviation kerosene consumption and associated effects on global climate are very likely to increase. Studies expect that the global aviation's CO_2 emissions might more than double within the next 20 years to values clearly above 2 Gt of CO_2 in 2050 (Grewe et al., 2021). Since non-CO_2 effects are a result of aviation kerosene combustion, they very likely also increase in the future; if no measures for their mitigation are taken (Grewe et al., 2021). Thus, the air transport sector urgently needs to lower both, its CO_2 and non-CO_2 related climate impact in order to reduce its overall climate impact effectively and promptly.

Initially, contrails form when the exhaust emitted by aviation kerosene combustion cools down and condenses mainly on the particles emitted from the turbines. The droplets formed subsequently freeze into ice crystals if temperatures are sufficiently low. Depending on temperature and humidity of the surrounding atmosphere, contrails can be short lived, but in ice supersaturated air they can persist for several hours (Vázquez-Navarro et al., 2015; Schumann et al., 2017) and potentially spread over large areas, thereby lose their linear shape and

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develop into contrail cirrus. Finally, the contrail cirrus dissolves because its ice crystals sublimate (Urbanek et al., 2017), e.g., by sedimentation into warmer and dryer air masses.

Whether or not a contrail forms can be determined by the Schmidt-Appleman criterion (SAC). It defines a threshold temperature below which contrail formation occurs (Appleman, 1953; Schumann, 1996; Bräuer et al., 2021a). This temperature depends on ambient conditions (pressure, humidity) and aircraft properties (Voigt et al., 2010, 2011; Jeßberger et al., 2013) including propulsive efficiency, released combustion heat, and water vapor emissions due to aviation kerosene combustion (Bock and Burkhardt, 2016). On a microphysical level, contrail formation is characterized by the condensation of gases (mainly water vapor) onto aerosol particles. Water droplets form on non-volatile and ultrafine aqueous aerosol particles in the cooling engine exhaust. For the soot-rich emission regime of present aircraft (10^{14} to 10^{15} particles/kg_{fuel}), condensation on soot particles is the dominant contrail formation process. In this regime, ice crystal numbers are nearly proportional to the number of emitted soot particles (Kärcher, 2018; Kleine et al., 2018; Märkl et al., 2023). However, for temperatures close to the SAC criterion only a fraction of soot particles is activated and the aircraft's aerodynamic vortices additionally reduce the fraction of soot particles activated for ice nucleation (Voigt et al., 2011; Kleine et al., 2018; Schumann et al., 2012; Unterstrasser, 2016). Various experimental measurements have shown that within the soot-rich regime, a reduction in soot particle emission numbers yields fewer but larger initial ice crystals (Bräuer et al., 2021a, 2021b; Voigt et al., 2021; Moore et al., 2017).

Aromatic aviation kerosene components are efficient soot precursors due to their tightly bound ring structure and a lower hydrogen content resulting from conjugated double bonds (Moore et al., 2017; Schripp et al., 2022). Therefore, reducing the aromatic content of aviation kerosene lowers soot emission numbers which in turn causes fewer but larger initial ice crystals (Voigt et al., 2021). Due to their increased weight they sediment faster into warmer and/or drier air layers (Kärcher, 2018). Eventually, this results in faster sublimation, a shorter contrail lifetime, and hence a reduced climate impact (Teoh et al., 2022a). Simultaneously, reducing the aromatics content of a particular fuel increases its overall hydrogen content. The consequence is that the water emissions from combustion are rising and thus slightly increase contrail occurrence (Teoh et al., 2022a). Simultaneously, the increased hydrogen content (and also the reduced sulfur content) increases the fuel's mass-specific energy content and slightly decreases mass-specific CO_2 emissions.

Accordingly, reducing the aromatics content of aviation kerosene poses a potential fuel-related measure to lower the contrail climate impact. Renewably sourced aviation kerosene[1] usually exhibits a much reduced aromatics content compared to fossil fuel-based kerosene. Flight experiments demonstrated that their use reduces soot particle number emissions (Bräuer et al., 2021a; Voigt et al., 2021; Moore et al., 2017; Brem et al., 2015; Schripp et al., 2018) and thus subsequently the contrail climate impact (Burkhardt et al., 2018; Bock and Burkhardt, 2019).

However, more than 99 % of the global aviation kerosene supply is presently produced from fossil fuel sources. While roughly 311 Mt/year of fossil fuel-based kerosene are consumed, production volumes of renewably sourced kerosene are estimated at around 0.2 Mt/year (IEA. Renewables 2022, 2022; WEF and CST). Even when considering recent initiatives to ramp up the production of renewably sourced kerosene, most studies indicate significant market shares of renewably sourced kerosene (>10 %) not before 2030 (IEA, 2021; Staples et al., 2018; Kieckhäfer et al., 2018).

For fossil fuel-based kerosene, hydrothermal processing can reduce their aromatics content considerably and the technology is available at industrial scale (Brink, 2020). Mild hydroprocessing primarily involves the removal of sulfur and (under certain conditions) the saturation of aromatics into cycloalkanes. Harsher process conditions also result in breaking of aromatic rings into acyclic alkanes. In refineries currently operated, the hydrogen required for hydroprocessing is often provided by steam methane reforming (SMR) of natural gas and sometimes via petrol coke gasification (Hsu and Robinson, 2017; Midilli et al., 2021). These pathways cause substantial CO_2 emissions, which incur an additional climate impact. Thus, the resulting net effect between reduced contrail climate impact and an increased CO_2 climate impact from aviation kerosene hydroprocessing needs to be evaluated carefully, as it might result in an overall increased climate impact. While few studies exist about hydroprocessed fossil-based kerosene (Faber et al., 2022), no study could be found which quantifies the associated climate impact mitigation potential.

This paper aims to analyse the net climate-impact of hydroprocessing of aviation kerosene from various types of crude oil with the goal to reduce the climate impact from contrails. Notably, this affects the climate impact from aviation's CO_2 emissions only marginally, but it might provide a fast and feasible option to reduce the overall climate impact from current aviation kerosene use. Only technological options being available in the short term (up to 5 years) are selected for this assessment, to evaluate and assess a short-term mitigation option.

Section 2.1 examines relevant changes of aviation kerosene composition by hydroprocessing. The contrail formation process and the cause-and-effect chain for contrail climate impacts and associated methods and data for its estimation are presented in section 2.2. Based on this, the climate impact reduction potential is analysed and implications on specification compliance are discussed (section 3). Finally, the results are summarized and potential further research areas are highlighted.

2. Methods and data

This study evaluates aviation kerosene processing strategies differing in processing intensity (Fig. 1). The central criterion for the assessment is the resulting change in net energy forcing, defined as amount of energy added to the Earth-atmosphere system. The choice of any metric to compare the climate impacts of CO_2 emissions and contrails is inherently limited; namely by large uncertainties in the contrail efficacy (Lee et al., 2021, 2023; Teoh et al., 2023; Schumann and Mayer, 2017; Hansen et al., 2005; Ponater et al., 2005; Rap et al., 2010), differences in time horizons considered and lacking consensus on a common metric (Teoh et al., 2020a). The choice of energy forcing as climate metric follows previous studies, which also compare a reduction in contrail climate impacts and additional CO_2 emissions (Teoh et al., 2020a, 2020b, 2022a). The climate penalty of additional aviation kerosene hydroprocessing ($\Delta EF_{hydroprocessing}$) and the climate benefit of less climate effective contrails are considered ($\Delta EF_{contrail}$). Based hereupon, the net change in energy forcing (ΔEF_{net}) is derived as the sum of the change in energy forcing from additional CO_2 emissions due to mild and harsh hydroprocessing ($\Delta EF_{hydroprocessing}$) and the change in energy forcing from contrails ($\Delta EF_{contrail}$) in 2019.

2.1. Aviation kerosene processing

The contrail climate impact can be reduced by reducing soot precursors in aviation kerosene. The sooting tendency decreases roughly from poly-to mono-cyclic aromatics via cyclo- and iso-alkanes to unbranched alkanes (Schripp et al., 2018, 2022). In this order, the respective hydrogen content of the aviation kerosene components increases. Thus, the hydrogen content is often used as proxy for the sooting tendency of aviation kerosene during combustion in aircraft turbines. Various in-flight and on-ground measurement campaigns support the

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[1] Such fuels are often referred to as "sustainable aviation fuel" or "SAF". However, this term is not used here, as the fuel's renewable origin does not guarantee the fulfilment of other sustainability aspects (e.g., biodiversity or social criteria).

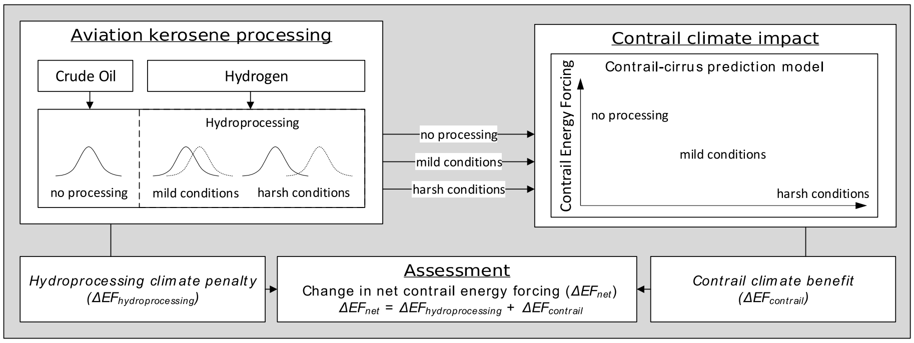

Fig. 1. Methodological approach: The assessment is based on the change in net energy forcing (ΔEF_{net}), which weighs the climate penalty of additional aviation kerosene hydroprocessing ($\Delta EF_{hydroprocessing}$) with the contrail climate benefit ($\Delta EF_{contrail}$) resulting from an increased fuel hydrogen content.

validity and the limitations of this proxy (Bräuer et al., 2021a; Märkl et al., 2023; Voigt et al., 2021; Brem et al., 2015; Schripp et al., 2018). The hydrogen content of aviation kerosene can be increased by hydroprocessing. This is a commonly used process used for fuel finishing within modern refineries (Hsu and Robinson, 2017). Hydrogen is added to the fossil straight-run kerosene derived from crude oil under elevated pressure and temperature conditions and reacts with heteroatoms (e.g., sulfur) and – depending on process conditions – saturates aromatics or even cracks cyclic components to alkanes (Hsu and Robinson, 2017; Coker, 2018).

2.1.1. Effect on aviation kerosene properties

The composition of crude oil and its aviation kerosene fraction varies greatly, typically depending on the crude oil's respective reservoir. The existing studies on the distribution of hydrogen contents of conventional aviation kerosene (Zschocke et al., 2017; Edwards, 2020; PQIS, 2011) indicate a mean hydrogen content of 14.1 m-% with a single standard deviation $\sigma = 0.2$ m-% points and a sample size $n = 57$. Here, only large, international airports are considered. These are typically supplied by pipelines and operate large fuel storage tanks resulting in an averaging tendency for the kerosene properties. The maximum permissible sulfur content of aviation kerosene is 0.3 m-% (ASTM D1655, 2022).

Hydroprocessing at comparatively mild processing conditions (typically 270–340 °C, 15–30 bar (Hsu and Robinson, 2017), "hydrotreatment") involves the removal of contaminants such as e.g., sulfur and the saturation of unsaturated components, such as alkenes. The saturation of aromatics into cycloalkanes can also be achieved within the upper temperature and pressure range, but typically aromatics will not be cracked to acyclic alkanes. Reactions by decreasing ease of reaction are heteroatom removal (mainly sulfur), a saturation of olefins, and then saturation of aromatics. Accordingly, for a substantial reduction in aromatics, the majority of the sulfur in the feed will be removed (Hsu and Robinson, 2017). The hydrogen demand is estimated based on the stoichiometric reactions for removing all sulfur in the feed and a conversion of 50 % of the feed's aromatics content into cycloalkanes. A conversion to acyclic alkanes is rather unlikely due to the mild process conditions during such a hydrotreatment. Based on these assumptions, the hydrogen content of the conventional aviation kerosene can increase by 0.3 m-% points (cf. Supporting Info)

In contrast, substantially harsher process conditions (typically 370–510 °C, 140–170 bar (Hsu and Robinson, 2017), "hydrocracking") even enable cracking of aromatics to acyclic alkanes. Typical reactions for such conditions are (again by decreasing ease of reaction) heteroatom removal, olefin saturation followed by the saturation of rings, cracking of cycloalkanes, and finally, the cracking of acyclic alkanes (Scherzer and Gruia, 1996). It is also assumed that all sulfur is removed and now 80 % of the aromatics in the feed are saturated and broken to acyclic alkanes. Hence, the hydrogen content of the conventional aviation kerosene can increase by about 0.7 mass-% points (cf. Supporting Info)

Aviation kerosene specifications limit the sulfur content of conventional aviation kerosene to 0.3 m-% (ASTM D1655, 2022). However, studies on distributions of aviation kerosene show that the majority of fossil-fuel based aviation kerosene contains not more than 0.15 m-% sulfur (Zschocke et al.; PQIS. Petroleum Quality Information System). To indicate the full value range, the sulfur content in this study is varied uniformly from 0.0 to 0.3 m-%. Typically, sulfur occurs in the form of heteroatoms. During hydroprocessing, these heteroatoms are saturated by hydrogen and release sulfur as hydrogen sulfide (H_2S).

2.1.2. Climate impacts of aviation kerosene processing

Presently, most hydrogen is produced from steam methane reforming (SMR) and coal/coke/petrol coke gasification (8.2 Exa-Joule (EJ) and 3.2 EJ in 2019, respectively (IEA, 2019)). A minor amount is produced from water electrolysis (less than 0.1 EJ (IEA, 2019)). The first two options result in substantial GHG emissions (about 10 kg CO_2/kg_{H2} for steam methane reforming (SMR) and about 20 kg CO_2/kg_{H2} for petrol coke gasification (Gaillot et al., 2023). In case of electrolysis, the lifecycle CO_2 emissions strongly depend on emissions from the provision of the electricity used for water splitting. In principle, if the entire power for electrolysis is provided from renewable sources, hydrogen from water electrolysis could be produced with almost zero CO_2 emissions. Thus, the emissions factor for hydrogen provision is varied between 0 kg CO_2/kg_{H2} and 20 kg CO_2/kg_{H2}. Hydroprocessing is an exothermic process (Hsu and Robinson, 2017), CO_2 emissions from potential heat demands or benefits from heat integration are not considered in this study.

The energy forcing of the additional CO_2 emissions from hydroprocessing is calculated as a measure for the climate penalty of aviation kerosene processing. The energy forcing is calculated based on the absolute global warming potential (AGWP) of a carbon dioxide (CO_2) pulse emission for time horizons (TH) of 20, 50 and 100 years (Teoh et al., 2022a)(Eq. (1)).

$$CO_2\ EF\ [J] = \int_0^{TH} RF_{CO2}(t)\ dt \times S_{earth}$$

$$\approx AGWP_{CO2,TH} \times m_{CO2} \times S_{earth} \qquad \text{(Eq. 1)}$$

$AGWP_{CO2,TH}$ is the mass-specific, absolute global warming potential of CO_2 (2.39×10^{-14}, 5.35×10^{-14}, 8.8×10^{-14} W yr kg^{-1} m^{-2} for a 20/50/100 year time horizon (Joos et al., 2013; Gaillot et al., 2023), respectively). S_{earth} refers to the earth's surface (5.101×10^{14} m^2) (NASA, 2023). m_{CO2} denotes the mass of CO_2 emitted by the additional hydroprocessing and is calculated as the product of the flight dataset's fuel consumption (cf. section 2.2, Supporting Info), the fuel mass-specific amount of hydrogen required for hydroprocessing and the emissions factor for hydrogen provision. Accordingly, m_{CO2} corresponds to the amount of CO_2, which would have been released if all fuel used in the flight dataset considered here would have been hydroprocessed. The increased hydrogen content results in an increasing gravimetric energy content of the aviation kerosene and thus reduces the flight dataset's fuel consumption. Accordingly, the amount of fuel to be hydroprocessed decreases and also CO_2 emissions from kerosene combustion decrease. While the first effect is taken into account, the reduction in CO_2 emissions from aviation kerosene combustion is not taken into account here. As the reduction in combustion-related CO_2 would (partly) compensate for the additional emissions from hydroprocessing, the change in net energy forcing (ΔEF_{net}) can be considered as conservative estimate (cf. Supporting Info). The use of flight data from the year 2019 eliminates uncertainties from air traffic development scenarios as well as uncertainties from assumptions on future meteorological conditions. The inter-annual variability of the simulation results is discussed in section 3.1.4.

2.2. Contrail climate impact

The contrail climate impact is estimated using the so-called contrail cirrus prediction model (CoCiP) from its open source distribution pycontrails/CoCiP version 48.1 (Teoh et al., 2020a, 2022a, 2022b, 2023; Schumann, 2012; Shapiro et al., 2023). This model simulates the life cycle of contrail segments formed along individual flight trajectories (Schumann, 2012).

In the contrail cirrus prediction (CoCiP) model, when consecutive waypoints meet the Schmidt-Appleman criterion (SAC), contrail formation is assumed. The soot emissions number influences the initial number of ice crystals within the contrail. However, a lower boundary is introduced at 10^{13} kg^{-1} to consider ambient aerosols and organic particles as ice nuclei (Kärcher, 2018). Additionally, the ambient temperature affects the initial number of ice crystals, as well as the soot activation rate (Bräuer et al., 2021a) and the fraction of ice particles that survive the wake vortex phase (Schumann, 2012). Contrail segments enduring the wake vortex phase are simulated in the model using time steps of 1 800 s until they reach their end-of-life conditions; i.e., the contrail ice crystal number decreases below the background ice nuclei concentration ($<10^3$ m^{-3}), the contrail's optical depth $\tau_{contrail}$ is less than 10^{-6}, or the lifetime surpasses a maximum of 24 h (Schumann, 2012). The contrail cirrus prediction (CoCiP) model calculates the local contrail radiative forcing (RF') for each waypoint, which represents the change in radiative flux over the contrail area (Teoh et al., 2022a). The contrail energy forcing is calculated as the product of the contrail segment RF', length, and width integrated over the contrail segment's lifetime (Teoh et al., 2020a, 2022b; Schumann and Heymsfield, 2017). The efficacy of the contrail radiative forcing is assumed to be unity, neglecting secondary effects such as the reduction of natural cloudiness as a result of contrail formation (Lee et al., 2021). Such secondary effects are beyond the scope of this study, the sensitivity of the results to various efficacies is shown in the Supporting Info.

The simulations cover 797,602 departing flights from the five largest European airports in the year 2019 by passenger numbers (Airports Council International), namely London-Heathrow, Paris Charles-de-Gaulle, Amsterdam Schiphol, Frankfurt International and Madrid-Barajas. For the uncertainty assessment, selected months from the years 2018 and 2023 are compared against the same four months related to the year 2019. Flight trajectories are loaded from the OpenSky Automatic Dependent Surveillance Broadcast (ADS-B) database (Schäfer et al.). Aircraft performance is based on the EUROCONTROL Base of Aircraft Data Family 3.15 (EUROCONTROLa; EUROCONTROLb) and the International Civil Aviation Authority (ICAO) Emission Databank (EDB) (EASA). Meteorological data is used from the European Centre for Medium-Range Weather Forecast fifth generation high-resolution reanalysis (ECWMF ERA5) (ECMWF) (0.25° × 0.25° horizontal resolution for 37 pressure levels and at a temporal resolution of 1 h). The ERA5 humidity fields are adjusted to in-situ observations using the so-called method of exponential scaling with latitude correction (Teoh et al., 2023). These datasets and methods have been extensively documented (Teoh et al., 2020a, 2022a, 2022b, 2023; Schumann, 2012; Schumann et al., 2021b). The average hydrogen content of all fuel supplied to the abovementioned airports is increased gradually in eight increments from 14.1 m-% up to 14.9 m-% and the fuel's hydrogen content-based soot particle emission index is calculated (Teoh et al., 2022a). The resulting contrail energy forcing is weighted against the energy forcing of the additional CO_2 emissions from additional hydroprocessing of aviation kerosene. The resulting net climate impact is evaluated for a range of hydrogen provision methods (petrol coke gasification, steam methane reforming, water electrolysis) and time horizons (20, 50 and 100 years). By default, the effect of fuel sulfur changes is not considered in the calculation of the contrail energy forcing. Several studies (Märkl et al., 2023; Voigt et al., 2021; Kärcher et al., 2015; Moore et al., 2015; Miake-Lye et al., 1998; Schumann et al., 2002; Petzold et al., 2005) suggest that a lower fuel sulfur content might further reduce the ice nucleation efficiency of the soot particles and would subsequently also lower the contrail's energy forcing. Accordingly, the simulations can also be considered conservative in this regard. Furthermore, the emission of sulfate aerosols has a direct and indirect (via aerosol cloud-interaction) climate impact. As both, direct and indirect climate impacts are still associated with large uncertainties (Lee et al., 2021), robust conclusions on the net climate impact of a kerosene sulfur reduction cannot be drawn so far and this effect is excluded here.

3. Results and discussion

3.1. Results

The measures studied here do not tackle CO_2 emissions from the combustion of fossil fuel-based kerosene. Therefore, from a mid-to long-term perspective, switching towards renewably sourced kerosene will be necessary, particularly because of the long atmospheric lifetime of CO_2. For the timespan while the availability of renewably sourced kerosene is limited, the climate effects of hydroprocessing fossil fuel-based kerosene are assessed here.

3.1.1. Variation of crude oil

Table 1 summarizes hydrogen demand and resulting CO_2 emissions for different aviation kerosene processing strategies and hydrogen sources. Sulfur contents at the specification limit can increase overall hydrogen demand by 2.5–5 %.

Fig. 2 compares the energy forcing of CO_2 emissions from hydroprocessing ($EF_{hydroprocessing}$, ◇) with the associated contrail climate impact ($EF_{contrail}$, ○) for each day in the year 2019 assuming hydrogen from steam methane reforming (SMR) and a time horizon of 50 years. Additionally, the mean net energy forcing per day is depicted (EF_{net}, ✕) and Fig. 3 compares the relative changes.

The daily (intra-annual) variation in contrail energy forcing is shown to indicate the daily variations in $\Delta EF_{contrail}$. $\Delta EF_{contrail}$ ranges from individual days with a cooling climate impact to days with $EF_{contrail}$ being five or more times higher than the mean. The mean EF_{net} per day from contrails is $< 5{*}10^{16}$ J, while some of the daily variations significantly exceed this value. $EF_{hydroprocessing}$ is significantly smaller, the additional CO_2 emissions due to hydroprocessing reduce the climate benefit from a reduction in energy forcing from contrails non-visibly in this figure representation. The largest changes in EF_{net} are caused by the reduced soot emissions and ice crystal number in contrails from the hydroprocessed fuel. The climate impact of hydroprocessing increases for harsher process conditions, since they result in an increased hydrogen demand (and thus $EF_{hydroprocessing}$). For the hydrogenation of aromatics to cycloalkanes (mainly mild hydroprocessing), less hydrogen is required than for hydrocracking to acyclic alkanes (mainly harsh hydroprocessing). In principle, higher sulfur contents increase the resulting climate impact, as the removal of sulfur incurs an additional hydrogen demand. However, against the daily variation in contrail energy forcing, this effect is barely noticeable. Due to the large daily variation of the contrail climate impact, in the following sections the

Table 1
Hydrogen demand of each aviation kerosene processing strategy and estimates for the resulting CO_2 emissions.

Type of hydroprocessing	H_2 demand [$kg_{H2}/t_{kerosene}$]			Additional CO_2 emissions [$kg_{CO2}/t_{kerosene}$] ([g_{CO2}/MJ])		
	Total[a]	aromatics conversion[a]	sulfur removal[a]	high[a]	medium[a]	low[a]
Reference case	0	0	0	0 (0)	0 (0)	0 (0)
Mild hydroprocessing	4–4.2	4	0–0.2	80–84 (1.82–1.91)	40–42 (0.91–0.95)	0 (0)
Harsh hydroprocessing	8–8.2	8	0–0.2	160–164 (3.64–3.73)	80–82 (1.82–1.86)	0 (0)

[a] Rounded figures.

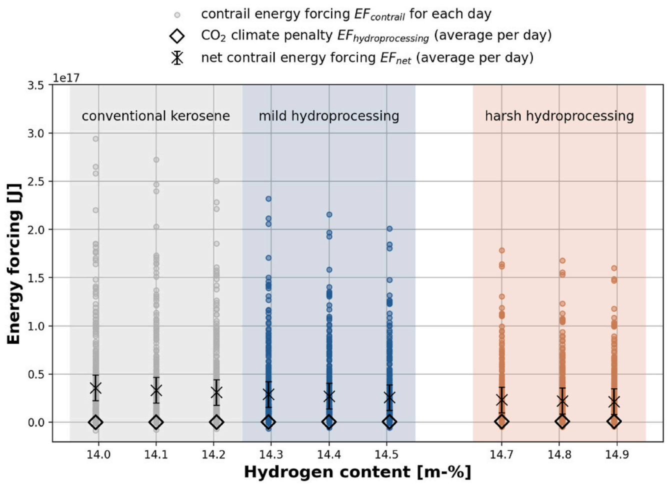

Fig. 2. Energy forcing of CO_2 emissions from hydroprocessing ($EF_{hydroprocessing}$, ◇) per day, contrail energy forcing for each simulated day ($EF_{contrail}$, ○) and mean net energy forcing per day in 2019 (EF_{net}, ×), for increasing fuel hydrogen content (**m-%** mass-percentage); error bars indicate the double standard deviation from the mean) assuming medium hydrogen provision emissions and a 50-year time horizon.

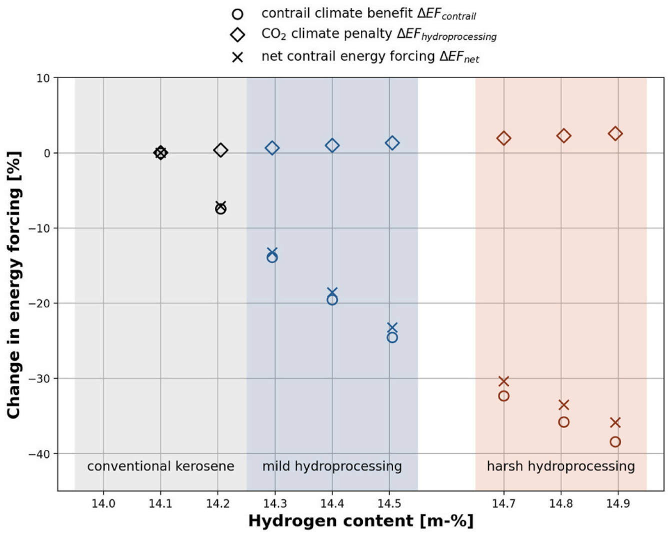

Fig. 3. Change in energy forcing for increasing kerosene hydrogen content in **m-%** mass-percentage by CO_2 emissions from hydroprocessing ($\Delta EF_{hydroprocessing}$, ◇), by contrails ($\Delta EF_{contrail}$, ○) and the resulting net effect (ΔEF_{net}, ×) for a 50-year time horizon.

annual sums of the hydroprocessing and contrail climate impact for the year 2019 are used and will be compared to other years later on (section 3.1.4).

Fig. 5 shows the annual change in $\Delta EF_{contrail}$, $\Delta EF_{hydroprocessing}$ and the resulting ΔEF_{net} for increasing hydrogen content by increased hydroprocessing for the flight dataset from the five largest European airports in 2019. The mean hydrogen content of fossil kerosene (cf. section 2.1.1) of 14.1 m-% is chosen as reference (no relative change in energy forcing). In general, the reduction in contrail energy forcing intensifies with increasing hydroprocessing intensity. The climate impact reduction does not increase linearly, but rather weakens in particular for high hydrogen contents, due to the increasing climate penalty of increasingly intense hydroprocessing. Additionally, contrail occurrence slightly increases with increasing hydrogen content (Teoh et al., 2022a) and thus further dampens the gain in climate impact reduction. This is shown by a decreased reduction rate of $\Delta EF_{contrail}$ for higher hydrogen contents of the harshly hydroprocessed fuels and the higher $\Delta EF_{hydroprocessing}$ for higher hydroprocessing intensities, both lead to a decrease in the sensitivity of the ΔEF_{net} with higher hydroprocessing intensity. Furthermore, high sulfur contents marginally increase hydroprocessing emissions. Accordingly, kerosene from crude oil with low aromatics and low sulfur content would be preferable from a climate perspective. The contrail climate benefit $\Delta EF_{contrail}$ increases significantly from mild (~20 %) to harsh processing conditions (~38 %). The hydroprocessing climate penalty $\Delta EF_{hydroprocessing}$ is small and also increases from mild (~1 %) to harsh conditions (~3 %), and leads to a net energy forcing reduction potential of hydroprocessing ΔEF_{net} of about 18 % for mild and about 33 % for harsh processing conditions.

3.1.2. *Variation of greenhouse gas (GHG) emissions intensity of hydrogen provision*

Fig. 4 shows the variation of net energy forcing for different hydrogen provision pathways. The highest reduction potentials (~35 % reduction) are achieved in the case of low hydrogen provision emissions (▽), which could potentially be realized by using renewably sourced hydrogen. Next is the medium emissions factor (×, ~33 % reduction), while the lowest potential exists for high specific emissions (△, ~31 % reduction).

The differences in hydrogen provision pathways can be explained by their different degrees of emission intensity. In case of the (idealized) assumption of hydrogen provision with no CO_2 emissions (e.g., by electrolysis powered by renewable energy sources), the net climate impact reduction ΔEF_{net} corresponds to the reduction in contrail climate impact $\Delta EF_{contrail}$ and increases by about 3 % points compared to a medium emissions factor for hydroprocessing (~10 kg_{CO2}/kg_{H2}). For high emission intensities (~20 kg_{CO2}/kg_{H2}) the climate impact reduction is slightly dampened (~3 % points) by the CO_2 emissions $\Delta EF_{hydroprocessing}$. Note that a potential climate impact reduction from lower in-flight CO_2 emissions due to an increased gravimetric energy content is excluded in this study (cf. 2.1.2). Hypothetically, net CO_2 emissions might even decrease when considering both, the increased gravimetric energy content and hydroprocessing with sufficiently low

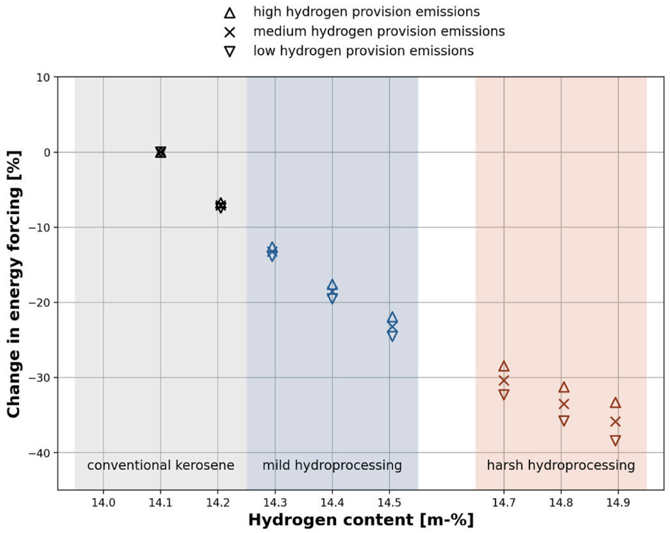

Fig. 4. Change in energy forcing (ΔEF_{net}) versus hydrogen content (**m-%** mass-percentage) for hydrogen provided with high (△), medium (×) and low CO_2 emissions (▽) for a 50-year time horizon.

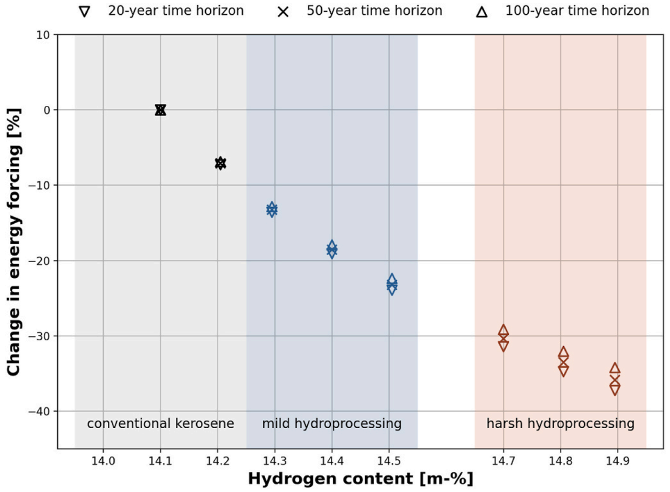

Fig. 5. Change in energy forcing for a time horizon of 20 years (▽), 50 years (×) and 100 years (△) assuming medium hydrogen provision emissions (**m-%** mass-percentage).

CO_2 emissions.

Consequentially, the lower the CO_2 emissions from hydrogen provision, the greater the climate-impact mitigation potential. However, even the use of hydrogen from processes causing high CO_2 emissions would still enable a reduction in net energy forcing by more than 30 % for harsh hydroprocessing.

3.1.3. Variation of the time horizon

Fig. 5 shows the net energy forcing for a time-horizon of 20 years (▽), 50 years (×) and 100 years (△). A medium emissions intensity of hydrogen provision is assumed (10 kg_{CO2}/kg_{H2}). The net climate impact decreases with increasing processing intensity for all time horizons considered. The reduction potential for a 20 year time horizon is slightly larger than the 50 year and 100 year perspective, but this choice of the time horizon has only a small effect on the relative changes in ΔEF_{net}.

The lifetime of a contrail lies within several hours, which is very short compared to all time-horizons considered. Hence, variations in the contrail-related energy forcing are not altered by the different time horizons. As a long-lived greenhouse-gas, CO_2 has an atmospheric lifetime from decades to centuries. A longer time horizon considers a longer effect duration of CO_2 emissions and thus increases the energy forcing from a CO_2 pulse emission. Therefore, the time horizon affects the contribution of CO_2 emissions from hydroprocessing towards the net energy forcing. However, since the climate impact of CO_2 emissions from hydroprocessing is quite small compared to the mean contrail climate impact, a large reduction in climate impact can still be observed even for adverse cases with high emission intensities. Thus, Fig. 5 clearly shows that not only for short-time frames (i.e., 20 years), but also in the longer-term (here 100 years) hydroprocessing of fossil fuel-based kerosene yields additional climate impact reductions, which are most pronounced for harsh processing conditions.

3.1.4. Uncertainty assessment

Fig. 6 shows net energy forcing per month for the different hydroprocessing strategies for 2018 (⬡), 2019 (□) and 2023 (⋄) to investigate the impact from different weather systems and air traffic volumes. To enhance computational efficiency, January, April, July and October were selected as representative months for each season. Trajectory data for October 2023 were not available in the OpenSky database and are thus not taken into account. In Fig. 6, the left ordinate shows the net energy forcing per month in absolute terms and the right ordinate shows the relative change in net energy forcing.

In absolute terms, the net energy forcing shows a clear inter-annual variability (e.g., ~20 % for 14.1 m-% hydrogen content between 2023 and 2019). Both, inter-annual weather variability and different air traffic volumes contribute to this variation.

In relative terms, however, the inter-annual variability is substantially reduced to a few percentage points. By using the reference case (hydrogen content 14.1 m-%) as baseline, the effects of different air traffic volumes or inter-annual weather variability can largely be excluded.

In conclusion, the absolute reduction in energy forcing for the hydroprocessing intensities varies with the overall climate impact of contrails among different years. However, the relative potential to mitigate the net contrail climate impact for a particular year appears to be rather consistent inter-annually. It amounts to about 18 % for mild hydroprocessing and about 33% for harsh hydroprocessing.

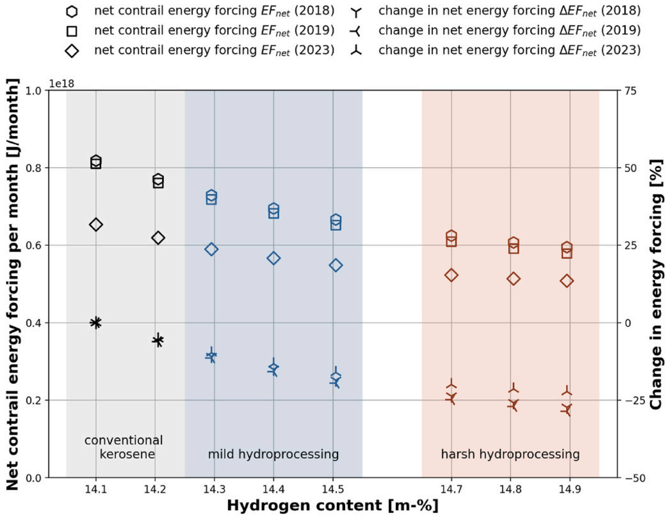

Fig. 6. Net energy forcing in absolute (left ordinate, empty symbols) and relative terms (right ordinate, star symbols) for January, April, July and October and different years assuming medium hydrogen provision emissions and a 50-year time horizon (**m-%** mass-percentage).

3.2. Discussion

3.2.1. Climate impact reduction potential

The climate benefits of reducing the contrail energy forcing by means of additional fuel processing outweigh the climate penalty of CO_2 emissions from aviation kerosene hydroprocessing. Based on the taken assumptions, this is not only the case for hydrogen provision with high CO_2 emissions, but also when longer timeframes (up to 100 years) are considered for the climate impacts.

The influential factors on climate impact (fuel hydrogen, and aromatic content, CO_2 emissions from hydrogen provision) could either be influenced by a particularly favourable crude oil (with high hydrogen and thus low aromatics content) and/or hydroprocessing based on hydrogen produced from renewably sourced energy. The development of future crude oil properties is primarily driven by overall crude oil demand for which aviation kerosene demand plays a minor role only. Thus, the future development of overall crude oil demand and its associated properties is difficult to predict (Hsu and Robinson, 2017). Simultaneously, renewably sourced hydrogen is increasingly considered as an important energy carrier for the future (Sens et al., 2022; DNV, 2022). Accordingly, using hydrogen with low lifecycle CO_2 emissions seems to be more realistic than a dedicated sourcing of favourable crude oils.

Hydroprocessing of aviation kerosene creates several secondary effects, not considered here. The increase in hydrogen content also increases the gravimetric energy density of the "low-contrail" fuel. However, this change in fuel consumption also reduces the CO_2 related climate impact of fuel combustion. This effect would counterbalance the CO_2 climate impact of hydroprocessing. Since this study assumes stoichiometric reactions for fuel processing and a broadly aggregated combustion model, a valid comparison of both effects requires further research. Additionally, the reduced aromatics content results in fewer soot particle emissions, which in turn reduces air pollution and thus improves air quality in particular at airports.

The kerosene's sulfur content does not only affect the hydrogen required for hydroprocessing but potentially also the number of soot particles activated into ice and hence the contrail climate impact. Various studies (Märkl et al., 2023; Voigt et al., 2021; Moore et al., 2015; Miake-Lye et al., 1998; Schumann et al., 2002; Petzold et al., 2005) describe the fuel sulfur content as contributing factor towards soot particle activation. For modern jet turbines with soot emissions in the low-soot regime, the activation of sulfur aerosols might gain importance for contrail formation (Märkl et al., 2023). In terms of emissions, lower sulfur emissions will improve air quality at airports and reduce the acid rain potential.

Previous studies investigated costs and environmental benefits of removing naphtalenes from aviation kerosene (Barrett and Speth, 2021; MathPro Inc, 2023) and estimate a smaller impact on reducing soot emissions and thus subsequent climate impact. As these studies only consider the removal of multi-cyclic aromatics, their measures are not as severe as those discussed here.

Some limitations remain inherent to this study, such as a more specific refinery configuration, detailed aviation kerosene composition after hydroprocessing and the influence of individual fuel components (e.g., cycloalkanes) on contrail formation. Due to the limited availability of aviation kerosene properties, the distribution of aviation kerosene hydrogen content is based here on 57 samples from the World Fuel Survey (Edwards). In light of the strong regional variations of contrail climate impacts, a regionally differentiated study with a larger dataset might allow for more detailed results. Straight-run kerosene differs largely from vacuum-gas oil (the usual feed of a hydrocracker). Hence, it is questionable if both could be processed simultaneously in the same processing unit. A more detailed simulation study of the process would require considering interdependencies with other products and detailed processing flows. Here, the effects of individual aviation kerosene components (e.g., aromatics or cycloalkanes) are aggregated by using their hydrogen content. More detailed combustion chemistry models and measurements of effects on contrail formation are necessary to validate these initial findings.

3.2.2. Specification compliance

Standardized fuel specifications define the permissible chemical and physical properties of aviation kerosene. Mainly, these are laid out in the specification ASTM D1655 (ASTM D1655, 2022); other specification standards exist in some countries (e.g., Russia and China). Their requirements, properties, and limitations are – in the case of commercial aviation – similar to those defined by ASTM (Rumizen et al., 2018). Renewably sourced kerosene must comply with the ASTM-standard D7566, which includes a lower limit of 8 vol-% aromatics content for renewably sourced kerosene. This limit has been introduced because aromatics can play an important role in sealing tightness and fuel quantity measurements among others (Zschocke et al.; Pechstein et al., 2018). Especially legacy aircraft are considered susceptive to a risk of e.g., fuel leakages due to deteriorated seal tightness after prolonged exposition with aromatics free aviation kerosene. For these reasons, compliance with the prescribed aromatics content is a critical issue also for hydroprocessed fossil-based kerosene, even though the specification for fossil-based kerosene, ASTM D1655 does not include a lower limit for the aromatics content.

Table 2 shows the change in aromatics content for the different aviation kerosene processing strategies and the resulting net energy forcing reduction (for medium hydrogen provision emissions, 50 year time horizon). Mild hydroprocessing achieves an aromatics content in the range of the ASTM-standard D7566 lower limit and in turn a reduction in net energy forcing by about 18 %. Harsher process conditions would remove aromatics beyond the minimum content, but also allow for a larger net energy forcing reduction of about 33 %.

Based on these values, mild hydroprocessing appears rather unproblematic from a specification compliance perspective. Harsh hydroprocessing at global scale would thus most likely require additional developments of the current fuel specifications. (Kramer et al., 2022)

Thus, in the short term, the aromatics content of aviation kerosene could be reduced within the current limits, i.e., down to 8 vol-%. This aviation kerosene could be used in all aircraft, even in older legacy aircraft with sensitive sealing materials. The drawback of this option is a limited net energy forcing reduction at about 18 % (Table 2). As most present-day refineries already use mild hydroprocessing ("hydrotreatment"), e.g., for road transport fuel desulfurization, this option might even incur comparatively low refinery modifications.

The climate mitigation potentials of harsh hydroprocessing would most likely require additional measures to ensure safe operation also for legacy aircraft. These measures could involve the development of an additional fuel standard for low contrail kerosene. Newly produced aircraft could potentially be certified for this standard. Some of the currently used aircraft could potentially be modified, e.g., by replacing particular fuel sealings or sensors with components compatible with such a low contrail kerosene. Moreover, it should be evaluated which legacy aircraft cannot be modified and would not be compatible with such a new fuel standard.

The increased hydrogen content due to fuel processing strategies

Table 2
Hydrogen and aromatics content and associated energy forcing reduction (medium hydrogen provision emissions, 50 year time horizon).

Processing case	Hydrogen content [m-%]	Aromatics [m-%] (incl. Napthalene)	Net energy forcing reduction [%]
Reference case	~14.1	~16.0	0
Mild hydroprocessing	~14.4	~8.0	~18
Harsh hydroprocessing	~14.8	~3.2	~33

alters further chemical and physical properties besides the aromatics content of the aviation kerosene produced (e.g., density). However, the relationships between aviation kerosene composition and resulting properties are very complex (Hall et al., 2021; Cookson et al., 1987), hence a comprehensive study of all specification-related aviation kerosene properties would be required to ensure safe operation of air transport. This would most likely involve the analysis of physical volumes of hydroprocessed fossil-fuel based kerosene and is thus far beyond the scope of this study (Kramer et al., 2022; Hall et al., 2021).

Additionally, a reduced aromatics content of fossil-based kerosene might incur challenges for its blending with renewably sourced kerosene, since the latter typically does barely contain any aromatics. As soon as the aromatics content of the blend decreases below 8 %, it would fall out of the limits of the current specification. The adjustment of both specifications, ASTM D1655 and ASTM D7566 as described above and an increasing fleet penetration of modern aircraft might alleviate this constraint.

3.2.3. *Conclusion*

Based on these results, increasing the hydrogen content of aviation kerosene, and thus reducing sulfur and aromatics content, allows for a short-term reduction in aviation's contrail-climate impact. Increased hydroprocessing intensity improves the mitigation potential but also raises associated uncertainties. The results discussed above show a reduction in net energy forcing between about 18 % (mild conditions) and about 33 % (harsh conditions) seems achievable by hydroprocessing for the case of medium emissions from hydrogen provision. The climate impact reduction potential is influenced by the unprocessed kerosene fraction's hydrogen and aromatics content and the CO_2 emissions intensity of hydrogen provision. The use of hydrogen with a low emissions intensity (e.g., from renewable sources) seems to be a promising option to further reduce the climate penalty of fuel processing.

A potential future option to balance the climate impact reduction and additional CO_2 emissions more efficiently would be to use such a "low-contrail" fuel only on flights where contrails form with a warming climate impact. As only a small percentage of the flights are responsible for a large share of the contrail climate impact, this might reduce the required amount of aviation kerosene to be treated (Teoh et al., 2020a, 2022a, 2022b); e.g., for the North Atlantic flight corridor, around 3–12 % of the flights cause around 80 % of the contrail climate impact (Teoh et al., 2022b). Supplying low-contrail kerosene (regardless of its fossil/renewable origin) specifically to the most climate-relevant flights would necessitate to segregate logistics infrastructures to accommodate for two different aviation kerosene types. Associated costs and emissions and their trade-off with additional climate benefits could be another area for further research.

The extent to which the benefits estimated in this analysis are further constrained, e.g., by the effects of individual aviation kerosene components on contrail formation or constraints from aviation kerosene specifications, is yet to be determined.

While this study provides a first insight into the potential of hydroprocessing of fossil-based kerosene to reduce aviation's climate impact, given the above-mentioned uncertainties, many open questions remain and provide potential for surprises. Therefore, before additional hydroprocessing of kerosene can be implemented as climate mitigation measure, specific uncertainties inherent to this study need to be reduced.

For instance, more precise hydroprocessing models might provide more detailed data on the resulting aviation kerosene composition and thus on specification compliance. For harsh hydroprocessing, compliance with current fuel specifications appears to be a critical issue especially for legacy aircraft. The abovementioned safety concerns have to be addressed and fuel specifications need to evolve accordingly. Moreover, comprehensive experimental data and detailed studies on the emission properties of such a hydroprocessed, "low-contrail" fuel would be required to provide insights into potential aircraft and infrastructure modifications, as well as on the magnitude of the climate impact reduction. More detailed combustion models and an extended suite of ground and cruise measurements of the effects of the hydroprocessed, "low-contrail" fuel on contrail formation are required to evaluate these initial findings. Additionally, a study of the associated cost of hydroprocessing, e.g., by increased hydrogen demand and switching to renewably sourced hydrogen would be interesting from an economic perspective. This particularly holds true for the cost as function of hydrogen content, since presumably cost are over-proportionately higher for removing all aromatics compared to a partial reduction in aromatics content by milder hydroprocessing. Based on this relationship, marginal abatement cost for hydroprocessing of fossil-based kerosene could be determined and allow for a comparison with other aviation climate impact mitigation measures.

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CRediT authorship contribution statement

Gunnar Quante: Writing – original draft, Methodology, Investigation, Conceptualization. **Steffen Voß:** Writing – review & editing, Methodology. **Nils Bullerdiek:** Writing – review & editing. **Christiane Voigt:** Writing – review & editing, Supervision, Methodology, Conceptualization. **Martin Kaltschmitt:** Writing – review & editing, Supervision, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.aeaoa.2024.100259.

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