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Uncertainties in mitigating aviation non-CO_2 emissions for climate and air quality using hydrocarbon fuels†

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The uncertainties over the effects of aviation non-CO_2 emissions on climate and air quality are assessed in the context of potential mitigation measures for liquid hydrocarbon fuels. Aviation non-CO_2 emissions that affect climate include nitrogen oxides (NO_x), aerosol particles (soot and sulphur-based), and water vapour. Water vapour and aerosols have small direct radiative effects but are also involved in the formation of contrails and contrail cirrus, currently, the largest non-CO_2 effect on climate. These non-CO_2 effects on climate are quantified with low confidence, compared to that of CO_2, which is quantified with high confidence. The sign of the NO_x radiative effects may change from positive to negative. The effects of soot and sulphur emissions on cloudiness are very poorly understood and studies indicate forcings that range from large negative through to small positive. NO_x and soot emissions can be reduced through changes in combustion technology but have tradeoffs with each other and CO_2. Soot can also be reduced through reduced aromatic content of fuels. In all cases, there are complex choices to be made because of tradeoffs between species, and CO_2. Contrail cirrus and soot aerosol–cloud interactions potentially have opposing signs but are both related to soot emissions (at present) and need to be considered together in mitigation strategies. Because of the uncertainties and tradeoffs involved, it is problematic to recommend definitive courses of action on aviation non-CO_2 emissions since they may be of limited effect or have unintended consequences. Aviation's non-CO_2 effects on climate are short-term, as opposed to those of CO_2, which last millennia. If aviation is to contribute towards restricting anthropogenic surface warming to 1.5 or 2 °C then reduction of emissions of CO_2 from fossil fuels remains the top priority. In terms of air quality, the situation is more straightforward with emissions standards being set by the International Civil Aviation Organization for NO_x and non-volatile particulate matter (and other minor species), which need to be complied with.

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Environmental significance

The emissions of aviation have effects on both climate and air quality. For climate, this includes both the long-lived greenhouse gas, CO_2 and non-CO_2 emissions result in a range of effects on climate. For air quality, the non-CO_2 emissions may impact upon human health, particularly those of NO_x and ultrafine particles. The sector has grown strongly in recent decades, and post the COVID-19 pandemic, is set to grow strongly again. This paper assesses the effects on air quality and climate in the context of potential mitigation options and 'net zero', for conventional liquid hydrocarbon fuels. Many of the effects are only quantified with limited scientific certainty and can have complex technological or operational tradeoffs that require careful analysis in order that perverse outcomes are avoided, and strategic investment decisions are based on a solid evidence base. Some of the proposed solutions do not always consider these issues carefully enough and we draw together the different lines of evidence to show where improvements to basic science knowledge are required.

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1 Technical summary

Aviation started to affect climate through both its CO_2 and non-CO_2 emissions from around 1940. Effects on climate are quantified by the metric 'effective radiative forcing (ERF) of climate' (which measures the way these effects perturb the earth-atmosphere energy balance relative to pre-industrialization). Aviation is currently calculated to represent about 3.5% of the total current radiative impact on climate,

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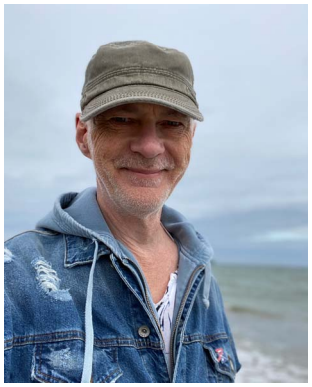
approximately 66% of which is currently attributed, with considerable uncertainty, to non-CO_2 emissions. Aviation non-CO_2 emissions, particularly NO_x and particulate matter, also have effects on local air quality, impacting human health.

In the last two decades, air traffic has grown strongly, and approximately 50% of the total historical emissions of CO_2 from the sector occurred over this period. This is important since CO_2 accumulates in the atmosphere. As the effects of the COVID-19 pandemic decline, it is expected that air traffic will gradually recover to growth rates similar to pre-pandemic levels.

Changes in aviation technology influence the composition of the global fleet slowly because of the 20 to 30 year timeframe of an individual aircraft's lifetime, the approximately 10 year development timescale of a new aircraft, and the in-production timescale of a decade or more. To meet internationally agreed targets requires rapid reductions of emissions of CO_2 to 'net zero' by 2050, and thereafter the active removal of atmospheric CO_2 to limit increases in global mean surface temperatures to 1.5 °C. This urgent requirement to reduce global CO_2 emissions means that the aviation sector faces serious challenges to contribute to the required emissions reductions.

A number of ways forward are being discussed that mostly involve changes in fuel to lower fossil carbon intensity substitutes, or alternative types of fuel, *e.g.*, liquid hydrogen. Compensation for continued fossil CO_2 emissions with equal removal and permanent storage of CO_2 emissions is also



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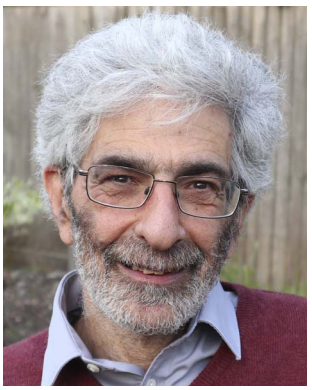

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a possibility but does not address non-CO_2 emissions, without additional actions. Currently, the fleet relies on liquid hydrocarbon fossil fuels and a number (currently 9) of drop-in replacement fuels that are certified for usage at up to 50% blends but are not available at either volume or price that enables significant substitution at present.

One of the major concerns over aviation's current and future effects on climate is related to its non-CO_2 emissions. The non-CO_2 emissions that can affect climate include oxides of nitrogen (NO_x), soot (small particles of unburned carbon), water vapour, and sulphur dioxide (SO_2). These emissions affect atmospheric composition and changes in cloudiness, which in turn affect the radiative balance of the atmosphere. The largest 'best estimate' non-CO_2 effects from aviation on climate are currently understood to be the formation of contrail cirrus and effects on atmospheric chemistry from emissions of NO_x. Contrail cirrus clouds are formed from the initial emission of water vapour condensing on co-emitted soot particles in cold ice-supersaturated regions of the atmosphere. The net radiative effect of contrails and contrail cirrus is warming from the residual of longwave warming, offset by short-wave cooling in the daytime (which is strongest at dawn/dusk). Emissions of NO_x from aircraft engines result in the formation of tropospheric ozone (O_3, a greenhouse gas) but also result in the destruction of ambient methane (CH_4), a greenhouse gas emitted from other sources, with a net positive forcing (*i.e.*, warming) from the balance of O_3 production and CH_4 destruction and its associated other cooling effects.

Non-CO_2 emissions of NO_x and particles are of ongoing concern to effects on human health. The World Health Organization has recently (2021) revised its guidelines of recommended limits and exposure of ambient air quality (over prior 2005 recommendations) for nitrogen dioxide (NO_2) from 40 μg m^{-3} as an annual mean to 10 μg m^{-3} and for PM_{10} (particulate matter with a diameter of 10 microns or less) from 20 μg m^{-3} as an annual mean to 15 μg m^{-3}. Shorter-term concentration recommendations have also changed. In the UK and Europe, current legislation limits annual average concentrations of NO_2 to 40 μg m^{-3} and PM_{10} to 40 μg m^{-3} for the protection of human health. USA National Air Quality Standards (NAAQS) are more complex, but the equivalent 'primary' standards (for the protection of human health) are 53 parts per billion (ppb) NO_2, or 101 μg m^{-3} as an annual average, and separate standards for PM_{10} of a 24 hour average of 150 μg m^{-3} and $PM_{2.5}$ of 12 μg m^{-3} as an annual average. Of significance to UK domestic air quality policy was the attribution of air pollution for the first time as a contributory factor to the death of a 9-year-old child in 2020 by a coroner.[1]

The purpose of this assessment is to provide an authoritative and up-to-date statement on the consensus science relating to aviation's non-CO_2 effects on the atmosphere, both in terms of climate and air quality, and how these may change in the future. The effects of both potential future technologies and fuels (liquid hydrocarbon type 'sustainable aviation fuels' – SAFs) are considered, as some of the non-CO_2 emissions are different. We do not consider changes to fuels that are nonconventional (*e.g.*, liquid cryogenic hydrogen or electric power sources) nor do we consider in any detail, putative high-flying supersonic aircraft. Neither do we consider the effect of climate change on aviation operations.

Considering future changes in atmospheric effects are vital in taking decisions on future technology development pathways, particularly under circumstances when reducing non-CO_2 emissions, or effects, may be at the cost of either extra CO_2 emitted or a loss of full opportunity to reduce aircraft CO_2 emissions. We find the following:

- The reduction of CO_2 remains a top priority because of its well-understood, powerful, and persistent effect on climate. Emissions of CO_2 accumulate in the atmosphere and the residual (about 20%) of any CO_2 emission remains in the atmosphere for millennia (high confidence).[2]
- Reductions of aircraft NO_x emissions through international regulation by the International Civil Aviation Organization (ICAO) have been driven historically by concerns over air quality in relation to human health in and around airports. Ambient air quality is regulated *via* national ambient air quality standards. Compliance with international regulations for aircraft NO_x emissions therefore remains a priority, as is also the case for emissions of non-volatile particulate emissions (nvPM)[3] from aircraft engines. Efforts to reduce NO_x emissions potentially bring about conflicts with improvements on specific fuel consumption, and the resultant incremental reductions in CO_2 emissions (high confidence).
- The design route to lowering fuel consumption, and hence CO_2 emissions, is to have a higher overall engine pressure ratio and higher turbine entry temperature. This tends to result in greater emissions of NO_x unless additional efforts are made to reduce them. Understanding the effect of tradeoffs between reduced emissions of NO_x and potentially increased emissions (or more limited reductions) of CO_2 can only be achieved with atmospheric science concepts. For example, in one modelling study, it was shown that a 43% reduction in NO_x with a corresponding increase in CO_2 emissions by 2% would bring about no net benefit to climate (low to medium confidence).
- Venting of engine emission oil may contribute to volatile particle emissions, depending on the design of the engine and the venting mechanism. There is some concern that these emissions may contribute to volatile organic particles, which could have harmful health effects in the vicinity of airports, and it has also been suggested that such particles may play a potential role in ice nucleation in relation to contrail formation in the soot-poor regime (very low confidence).
- The net effect of aircraft engine NO_x emissions on ERF remains uncertain because of their complex interaction with atmospheric chemistry, which is non-linear and highly dependent on other sources of emissions. The outcome of this is that there is no unique radiative forcing effect per unit NO_x emission (on a global scale). Also, the effect of aircraft NO_x emissions on ozone varies strongly with location, altitude, and time of emission. Furthermore, limited studies show that projected future reductions in other NO_x surface sources under strong climate protection scenarios for 1.5° (scenario SSP1.9) and 2° (scenario SSP2.6) can shift the balance of the net radiative effect of aircraft NO_x (the residual of positive and negative ERFs) from

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positive to being negative in the future, even with increasing global aviation NO_x emissions. The temperature effects of these changes are poorly understood, because of the complexities of a negative forcing a global scale and a positive forcing in the northern hemisphere, but available evidence indicates that a net negative NO_x global-mean ERF could still result in northern hemisphere warming (low confidence).

• The above factors make decisions on NO_x reductions that impact on fuel efficiency difficult. Externally-driven decisions on future international (ICAO) regulatory requirements will have a strong impact on developmental and manufacturing decisions but changes that would increase CO_2 emissions are to be avoided, since the climate outcome of a NO_x–CO_2 tradeoff is uncertain and not easy to estimate, since any extra CO_2 emission causes additional warming (high confidence).

• The formation of contrail cirrus clouds by aviation is estimated to be the largest non-CO_2 ERF, at present, but with large uncertainties. While ERF is a better measure of temperature response than RF, it is not necessarily complete and the climate sensitivity to contrail cirrus may be smaller than that of CO_2.

• Contrail cirrus is currently driven by the persistence of initial ice crystals formed from water vapour condensing on soot emissions under threshold temperature conditions ($\leq$233 K) and ice-supersaturated conditions at cruise altitudes. The radiative effect varies strongly with location, altitude, and time of emissions (from positive to negative). When contrails form in ice-supersaturated regions, they can persist and spread and grow into an extensive contrail cirrus cloud coverage. Using conventional fossil fuel, reductions of soot number emissions down to a threshold of around 10^{14} per kg fuel can reduce the number of ice crystals (medium confidence), which is modelled to reduce the ERF of contrail cirrus (very low confidence). At lower number concentrations of soot per kg fuel, separate microphysical process modelling indicates that at temperatures well below (12 K) threshold formation conditions, nucleated ice crystal numbers can increase, owing to enhanced activation of ultrafine aqueous particles, or start to level-off (non-linearly) at temperature conditions close to threshold formation conditions (very low confidence). The equivalent detailed behaviour for low aromatic content, low sulphur 'SAF' has not been modelled or assessed.

• Soot emissions can be reduced by both technological means through changes in combustion technology and adoption of low aromatic fuels (high confidence). Lowering the aromatic content of fuels results in reduced soot mass and number emissions (high confidence). Some current combustor technologies, developed originally to reduce NO_x emissions, are not always so effective in reducing soot emissions. For some combustor technologies there is an inherent trade-off between the conditions to reduce NO_x and soot. Pure bio-based and synthetic SAF have low aromatic content and zero sulphur content. Such fuels would reduce soot number concentrations in the plume (high confidence) and may reduce contrail ERF (very low confidence). Currently, modelling the effect of reducing the aromatic content of kerosene fuel through, *e.g.*, increased use of SAF, on contrail ERF is incomplete and highly parameterized. Such modelling does not address the role of aqueous ultrafine particles at low soot regimes and the co-benefit of SAF on reducing contrail ERF should be interpreted as tentative and as yet unproven.

• In global climate models, soot particles, once processed through contrails have been assumed to initiate heterogeneous nucleation of ice crystals at lower supersaturations than homogeneous nucleation. The resulting aerosol–cloud interaction with cirrus may cause a negative forcing, or possibly a small positive forcing (very low confidence), depending upon modelling assumptions. The effects of soot on contrail cirrus properties are normally modelled separately to these aerosol–cloud interactions but aircraft soot emissions underly both; both effects should be considered together when considering mitigation of soot emissions, by either technological means or altering fuel composition.

• Currently, when considered together, the present-day effects of contrail cirrus (assessed with low confidence) and aerosol–cloud interactions (assessed with very low confidence) could have a net positive or net negative forcing.

• Navigational avoidance of cold ice-supersaturated regions (largely during the night) has been suggested as a means of reducing the occurrence of persistent contrails. There is currently very low confidence in this mitigation measure. This is because of the challenges in making accurate predictions of persistent contrails on the time and space scales required for operational implementation on an individual flight basis of such a mitigation measure, and lack of data/evaluation of reductions in contrail cirrus ERF *versus* potential CO_2 ERF increases from changing flight levels/routes under such an approach. Moreover, the fundamental premise of contrail cirrus ERF being of importance to mitigate is not yet established.

• The use of SAF has been suggested to result in reductions in contrail cirrus forcing (very low confidence) although the evidence for this is weak (with conflicting modelling results), whereas no effect on NO_x emissions is envisaged from the use of SAF (medium confidence). The effects of SAF on aerosol–cloud interactions of soot and sulphur are potentially important (introduction of SAF may result in the removal of a negative forcing in both cases) but are still highly uncertain.

• Continued use of fossil fuel for aviation, combined with permanent removals of CO_2 to compensate for aviation emissions would bring about no non-CO_2 benefits (high confidence) other than those from increased fuel efficiency (with a caveat on tradeoffs), unless additional actions were taken. These could include the active removal of aromatic compounds from the fuel, which is likely to result in an energy cost and therefore increased CO_2 emission at the fuel refinery operation, estimated to be 97 kg CO_2 per tonne of kerosene, an additional 3% CO_2 from the fuel burned (very low confidence).

• Compliance with international regulations of NO_x and nvPM emissions is essential. It would be prudent to pursue technological approaches that limited the NO_x-fuel burn tradeoff as much as possible so that efforts to reduce CO_2 are not thwarted by NO_x emission reduction requirements.

• In terms of transitioning either to SAF or potentially conventional kerosene with reduced amounts of aromatics, it is important to pursue elastomer technologies for fuel systems

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that do not rely on aromatic content of fuels for sealing (high confidence).

2 Background and introduction

Aviation is responsible for a range of emissions that affect climate that can be quantified in terms of its 'Effective Radiative Forcing' (ERF, in watts per square metre, see Section 4), for which a positive forcing implies warming, and a negative one, cooling:

- Emissions of CO_2 result in a positive ERF.
- Emissions of nitrogen oxides (NO_x, where NO_x = NO + NO_2)[4] result in the formation of tropospheric ozone (O_3), an important greenhouse gas, *via* atmospheric chemistry, with a positive ERF.
- Emissions of NO_x also result in the destruction of ambient methane (CH_4), again *via* atmospheric chemistry, which is accompanied by a parallel, decadal loss of tropospheric O_3 and a reduction in stratospheric water vapour, with a negative ERF.
- Emissions of water vapour into the stratosphere result in a positive ERF.
- Emissions of sulphur dioxide (SO_2) arising from sulphur in the fuel, which is oxidized to form sulphate particles, resulting in a negative (termed "aerosol-radiation interaction") ERF.
- Emissions of soot particles result in a positive aerosol-radiation interaction ERF.
- The formation of persistent linear contrails that may develop into contrail cirrus clouds (depending upon atmospheric conditions) results in both positive and negative ERF effects but overall, cause a positive ERF effect.
- Sulphate and soot emissions may also interact with low and high-level clouds (termed "aerosol–cloud interactions"), respectively, causing ERFs of highly uncertain magnitude, likely to be negative in the case of sulphate, and of uncertain sign in the case of soot. The effects are also dependent on the background aerosol.

Aviation is calculated to represent about 3.5% of the total (2018) anthropogenic radiative impact on climate, approximately 66% of which is currently attributable, with considerable uncertainty, to non-CO_2 emissions.[6] This ERF from aviation CO_2 + non-CO_2 ERF has been separately calculated to represent approximately 4% of the contribution to global mean surface temperature increase since pre-industrialization, or approximately 0.04 ± 0.02 °C to 2019.[7]

The most recent assessment of aviation ERF,[6] is shown in Fig. 1. The details of many facets of this figure, including the ERF metric used, are discussed in Sections 4–8.

Some non-CO_2 emissions from aircraft engines, namely NO_x, nvPM, carbon monoxide (CO) and unburned hydrocarbons (HC)

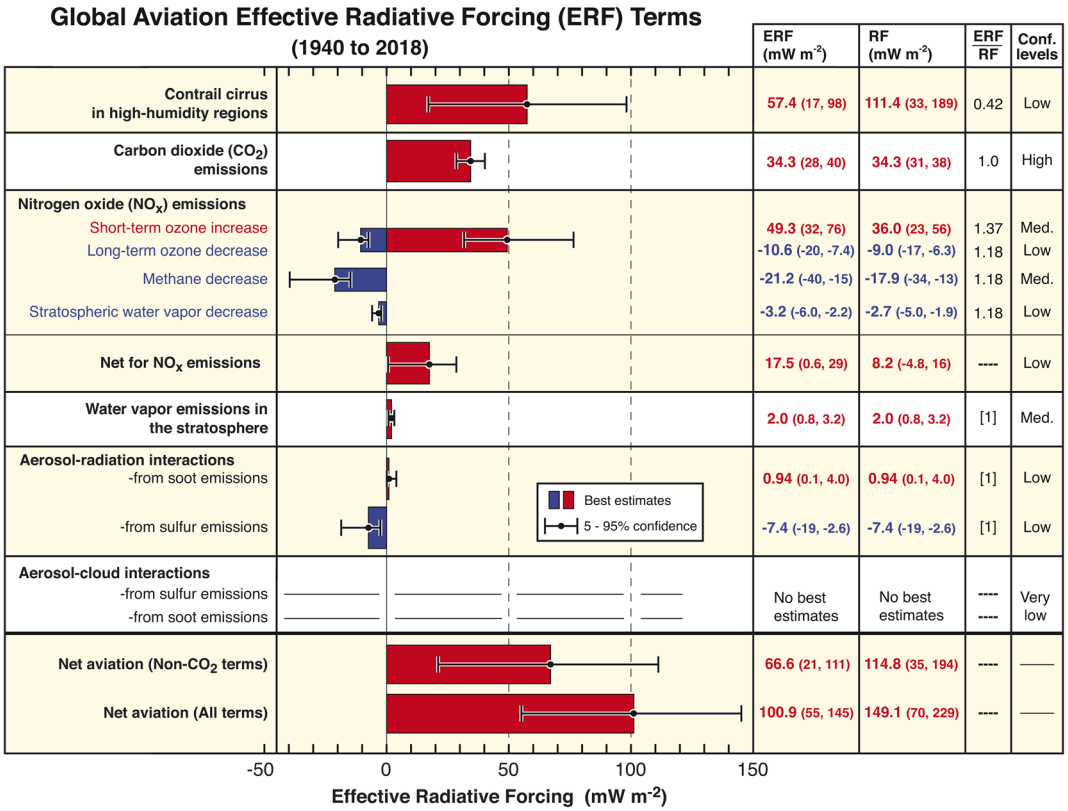

Fig. 1 Best-estimates for effective radiative forcing (ERF) terms from global aviation from 1940 to 2018. The bars and whiskers show ERF best estimates and the 5–95% confidence intervals, respectively. Red bars indicate positive terms and blue bars indicate negative terms. Numerical ERF and RF values are given in the columns with 5–95% confidence intervals along with ERF/RF ratios and confidence levels. RF values are multiplied by the respective ERF/RF ratio to yield ERF values. ERF/RF values designated as [1] indicate that no ERF/RF estimate is available yet. Taken from Lee *et al.* (2021).[6]

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are regulated under international agreements made at ICAO[8] and implemented through national legislation. These regulations are primarily intended for the protection of human health through ambient air quality standards throughout the Landing-Take-Off cycle (LTO), which is defined by the emissions up to 3000 feet.

During the coming years, decisions will need to be made as to the relative level of investment in the various technological innovations presented within the pathway to net zero. Solutions including hydrogen-fuelled aircraft, Sustainable Aviation Fuel (SAF)[9] utilization in conventional aircraft, fuel cells and electrification of aircraft, will need to be assessed in a holistic manner. Decarbonization and the reduction of non-CO_2 emissions both represent opportunities and should both be considered in strategic comparisons for investment.

This present study summarizes the current state of knowledge on aviation non-CO_2 emissions in relation to current fossil fuels and potential future liquid SAF substitute fuels; it identifies key concepts, uncertainties, and where the science base needs to be improved.

In Section 2, the policy, societal and policy background is described and some key concepts around 'net zero' are outlined. In Section 3, the background to the regulatory landscape is set out, along with future developments, and how they might relate to aircraft engine emission regulations, along with other international initiatives to reach net zero within the aviation sector. In Section 4, aviation emissions and their effect on the atmosphere are described in terms of ERF. In Section 5, specific issues around air quality are examined; the impacts on human health, contributions of aircraft to local quality, and the contribution of cruise-level emissions to ground-level concentrations. In Section 6, the linkages between approaches to reducing CO_2 *via* changes in fuels and non-CO_2 effects are outlined. In Section 7, the concept of CO_2 emissions equivalences between CO_2 and non-CO_2 emissions are further outlined and how these metric types colour the perception of the importance of non-CO_2 *vs.* CO_2 emissions. Also, consideration is given to how perceptions on 'equivalence' would change, if the commonly accepted policy metric (the 100 year Global Warming Potential; GWP_{100}), its time horizon, or values change. In Section 8, some of the trade-offs between engine technology development in terms of fuel efficiency described in Section 3, and non-CO_2 effects are explored, and the key uncertainties identified. What all the foregoing might mean for strategic decision-making is discussed in Section 9, and lastly, in Section 10, proposed research requirements to make progress on key uncertainties are briefly highlighted.

In this overview, the following aspects were out of scope:

- Noise impacts of aviation.
- Substances not emitted by conventional jet fuel powered aircraft engine exhausts.
- Comparisons between conventional jet fuel powered aircraft and hydrogen-fuelled aircraft, fuel cells and electrified aircraft.
- Putative fleets of high-altitude supersonic aircraft.
- Technology design iteration costs to achieve compliance with a change in regulation *e.g.*, emission limit changes.
- The impact of climate change on aviation activities.
- Costs.

3 The climate crisis and 'net zero' driver

3.1 Policy developments

Concern about the prospect and adverse impacts of global warming date back to the 1970s, with early forecasts of the climate response to continued fossil fuel dependent economic activity proving remarkably accurate. Despite declining surface temperatures at the time and some suggestions of an oncoming ice age, Nordhaus (1977),[12] drawing on Manabe and Wetherald (1967),[13] predicted a human-induced increase in global temperatures starting in the late 1970s and exceeding 0.6 °C over the next 40 years. This is exactly what has occurred. Total human-induced warming since the late 19th century, conventionally accepted as representative of pre-industrial conditions (*i.e.*, 1850–1900), reached 1.07 °C (best estimate, range 0.8 to 1.3 °C) in the decade 2010–2019 (Intergovernmental Panel on Climate Change, IPCC, 2021)[14] and accelerated to over 0.2 °C increase per decade (IPCC, 2018).[15] At the current rate, warming would reach 1.5 °C above pre-industrial shortly after 2030, and 2 °C shortly after mid-century.

The fact that this warming was both foreseeable and foreseen is not simply of academic interest to fossil fuel dependent industries, since it increases the downside risk of continuing with business-as-usual, opening the possibility of relatively rapid shifts in public and legislative opinion affecting their business model. Moreover, the resultant warming raises the question of responsibility for historical emissions, loss, and damage.[16]

Scientific understanding of the importance of different radiative forcing mechanisms has evolved significantly since the 1980s but understanding of the global temperature response to those forcings has only undergone minor revisions. Concern, however, has increased with improved knowledge of the impacts of climate change at different warming levels (*e.g.*, IPCC, 2022 AR6 WGII),[17,18] and increased evidence of extreme events being linked to increases in greenhouse gases. Until 2009, the consensus working interpretation of what the 1992 United Nations Framework Convention on Climate Change (UNFCCC) referred to as "dangerous anthropogenic interference in the climate system" was a global warming of greater than 2 °C.

A 2 °C limit was mentioned in the 2009 Copenhagen Accord and 2010 Cancun Agreement, but not formally agreed. Following the IPCC 5th Assessment Report and Structured Expert Dialogue, and under intense pressure from a coalition of vulnerable countries including small island states, the Paris Agreement in 2015 adopted the much more ambitious goal of "holding the increase in the global average temperature to well below 2 °C above pre-industrial levels and pursuing efforts to limit the temperature increase to 1.5 °C".

Under the Paris Agreement, the UNFCCC explicitly requested a report from the IPCC into the impacts of a global warming of 1.5 °C and associated mitigation pathways, recognizing the lack of research available in 2015 into these very ambitious mitigation goals. The resulting Special Report on Global Warming of 1.5 °C (SR1.5; IPCC, 2018)[15] found "robust differences" between

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a climate at 1.5 °C *versus* one at 2 °C of warming (SR1.5, Chapter 3, 3.3.1), with "climate-related risks to health, livelihoods, food security, water supply, human security, and economic growth … projected to increase with global warming of 1.5 °C and increase further with 2 °C." (SR1.5 SPM, B5) The finding that additional warming beyond 1.5 °C has clear and demonstrable impacts fuelled intense interest in the question of Loss and Damage, the UNFCCC's mechanism for addressing unavoidable climate-related harm, as well as the possibility of civil liability should, as seems likely, temperatures exceed 1.5 °C: again, this presents a down-side risk to fossil-fuel-dependent companies and industries.

In the first major update to the UNFCCC process after Paris, the 2021 Glasgow Climate Pact strengthened the language around the Paris temperatures goals, including "resolves to pursue efforts to limit the temperature increase to 1.5 °C". With the exception of 2016, which may have been an emissions accounting effect, and the COVID pandemic, anthropogenic emissions of the main greenhouse gases, CO_2, methane, and nitrous oxide,[19] have continued to rise; the latest World Meteorological Organization's Greenhouse Gas Bulletin reporting increases of all these long-lived greenhouse gases.[23] This consistent pattern of ever-more ambitious temperature targets and continued failure to achieve coordinated emission reductions highlights an increasing risk of sudden shifts in the regulatory and investment climate as tension mounts between demands for and failure to deliver climate action, suggesting a high premium on flexible response strategies that can be scaled rapidly.

3.2 'Net zero' in the Paris Agreement process and how it relates to aviation

Despite limited progress on emission reductions, considerable progress has been made over the past decade in acknowledging what needs to be done to limit global warming, and the need for net zero emissions. Article 4 of the Paris Agreement states: "In order to achieve the long-term temperature goal set out in Article 2, Parties aim … to achieve a balance between anthropogenic emissions by sources and removals by sinks of greenhouse gases in the second half of this century." The Glasgow Climate Pact, paragraph 17, introduced the phrase 'net zero', while also highlighting the distinct requirements on CO_2 and other greenhouse gases: "limiting global warming to 1.5 °C requires rapid, deep and sustained reductions in global greenhouse gas emissions, including reducing global carbon dioxide emissions by 45 per cent by 2030 relative to the 2010 level and to net zero around mid-century, as well as deep reductions in other greenhouse gases." Net zero is generally accepted in the UNFCCC process to refer to what it will take to achieve the "…balance…" referred to in the Paris Agreement but remains ambiguous in important respects, particularly in relation to aviation.[24]

The IPCC AR6 defines net zero CO_2 emissions as "(the)…* condition in which anthropogenic carbon dioxide (CO_2) emissions are balanced globally by anthropogenic CO_2 removals over a specified period" (AR6, Glossary) and makes clear in the definition of land-use change that "anthropogenic removals" are those that occur as a direct result of human activity (SR1.5 Glossary). Processes such as the additional uptake of atmospheric CO_2 by vegetation due to the 'CO_2 fertilization' effect resulting from past emissions are not considered 'removals' by the IPCC, although they are considered removals in national inventories and in the voluntary carbon markets if they occur on 'managed lands'.[25,26]

While the definition of a removal is a matter of convention, it is important to note that the scientific studies that established that achieving net zero CO_2 emissions would be sufficient to halt the increase in CO_2-induced warming—providing the basis for Article 4 of the Paris Agreement—used the IPCC definition. If all CO_2 uptake on managed lands is reclassified as a negative emission and hence used to offset ongoing fossil fuel emissions, and a much higher proportion of the land surface is reclassified as managed (which is also occurring in national inventories), then achieving nominal net zero CO_2 emissions would no longer be sufficient to halt global warming.[27]

This classification of CO_2 'removals' from managed lands as being available to offset fossil fuel emissions is accepted under the UNFCCC.[25] However, this confuses and conflates the short-term C cycle with the long-term cycle, since managed land CO_2 'removals' (itself, a result of past CO_2 emissions and the fertilization effect) is already accounted for in C-cycle models. Such accounting is not the scientific definition of net zero, as used by the IPCC,[27] and doing so risks double counting.[25,26]

This point is important for any mitigation strategy that relies upon removal of CO_2 into the biosphere (through, for example, afforestation) to compensate for continued emissions from fossil fuels. At present, accounting for carbon uptake in 'nature-based solutions' does not, in general, distinguish deliberate ('active') from inadvertent ('passive') consequences of human activity; this distinction is essential to deliver the desired climate outcome. For net zero CO_2 emissions to be consistent with halting warming, only CO_2 removals that are the direct consequences of ongoing human activity must be included in aggregate human-induced emissions. It is important to distinguish between 'active' carbon uptake and 'passive' uptake that results from processes such as plants growing faster because of higher atmospheric levels of CO_2 due to past emissions; otherwise, the assumption that net zero emissions will be sufficient to halt global warming is not valid. For some forms of nature-based solutions, making this distinction may even be impossible.

The IPCC AR6 defines net zero greenhouse gas emissions as a "Condition in which metric-weighted anthropogenic greenhouse gas (GHG) emissions are balanced by metric-weighted anthropogenic GHG removals over a specified period." (IPCC, 2021, AR6 Glossary)[14] and notes "The quantification of net zero GHG emissions depends on the GHG emission metric chosen to compare emissions and removals of different gases, as well as the time horizon chosen for that metric.", where "…metric…" refers to the 'exchange rate' used to compare emissions of different gases (see Section 7). The IPCC does not recommend any specific metric, but the UNFCCC requires that emissions are reported using the GWP_{100} metric, and this is the metric used to define net zero emissions. Importantly for aviation, neither the

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Paris Agreement nor the Glasgow Climate Pact make specific reference to non-greenhouse-gas climate forcers, such as contrail cirrus, but as long as these individually have a warming effect, they might be regarded as implicitly included since they play a net warming role, under Article 2, although not identified under Article 4. The IPCC and UNFCCC are much more circumspect about including cooling aerosols in the definition of net zero because of the potential implications for solar geoengineering.

The IPCC defines climate neutrality (in SR1.5) as "a state in which human activities result in no net effect on the climate system" but avoided using the term in the AR6 WGI (IPCC, 2021)[14] because "the concept is diffuse … and not readily quantified" (IPCC, 2022, AR6 WGIII, Cross Chapter Box 3).[28] While definitions remain contested, the impact of different climate drivers on global average surface temperature, which largely determines climate impacts and is the focus of international climate agreements, is well understood. The IPCC SR1.5 summarized: "Reaching and sustaining net-zero global anthropogenic CO_2 emissions and declining net non-CO_2 radiative forcing would halt anthropogenic global warming on multi-decadal timescales." (SR1.5 SPM, A2.2). For aviation, with its current significant non-CO_2 radiative forcing impact, the joint condition is important: to halt further aviation-induced warming, both CO_2 emissions need to be reduced to net zero and net positive non-CO_2 radiative forcing needs to decline, with the necessary rate of decline implicitly reported by the IPCC and confirmed in various papers, being approximately 3% per decade.

A challenge for aviation is that, while immediate climate targets are typically set in terms of greenhouse gas emissions aggregated using the GWP_{100} metric, the long-term goal of the Paris Agreement is set in terms of global temperature; when short-lived climate forcers such as contrail cirrus are concerned, emissions aggregated using GWP_{100} do not actually reflect impact on global temperature.[29] The discrepancies are non-negligible. As stated in the IPCC AR6 WGI[14] (Chapter 7, Section 7.6.1.4)., "expressing methane emissions as CO_2-equivalent using GWP100 overstates the effect of constant methane emissions on global temperature by a factor of 3 to 4 (Lynch *et al.*, 2020),[30] while understating the effect of any new methane emission source by a factor of 4 to 5 over the 20 years following the introduction of the new source (Lynch *et al.*, 2020, their Fig. 4)."[31] Allen *et al.* (2022)[27] demonstrate that this statement also applies to any SLCF. At present, it is impossible to predict exactly how this slightly incoherent situation will be resolved. It may be that governments will persist with the status quo and simply accept that emissions targets do not reflect warming outcomes. Alternatively, in future, for example as emissions begin to decline and these discrepancies become more obvious, governments may decide to reframe incentives based on impact on global temperature rather than impact on aggregate GWP_{100} emissions. Such a change would increase the incentives for immediate reductions in short-lived climate forcers, and increase penalties for any increases, by correctly reflecting their impact on global temperature, while at the same time recognizing that the warming impact of constant ongoing SLCF emissions has been overstated relative to that of CO_2. In the meantime, it is essential to quantify the impact of decisions and pathways on both emissions aggregated using GWP_{100} and global temperature. So-called 'warming equivalent' emissions support this dual reporting, is discussed further in Section 7.

However, there is acknowledged ambiguity in the text of the Paris Agreement over the emissions under consideration, and the consequential definition of 'net zero'.[32] How 'net zero' relates to aviation is also complicated by its non-CO_2 emissions, and the fact that international aviation emissions are not explicitly identified in the agreement and fall outside of Nationally Determined Contributions (NDCs), whereas the Kyoto Protocol gave specific responsibility for international aviation emissions to ICAO.[24]

4 Combustion, emissions, and fuels

4.1 The existing and anticipated regulatory and policy framework

Engine emissions standards for international aviation are set by ICAO's Committee on Aviation Environmental Protection (ICAO-CAEP).[33] These standards limit the LTO emissions of NO_x, CO, HC and nvPM (number and mass) from aircraft engines.[34] It is worth noting here that emissions of CO_2 are dealt with at the aircraft level rather than the engine level, since the same engine can be fitted to varying sizes of aircraft, and the emission of CO_2 is dependent upon the total fuel burn, which, in turn is dependent upon the aircraft technology, size, and mass.[35] In recent years, the significance of HC and CO emissions from aircraft engines has decreased owing to much lower emission rates from improved combustion. As other sources of emissions are reduced at ground level, these emissions may become more important in the future.

The focus at CAEP in recent years has been on updating the NO_x regulations and developing new regulations for nvPM mass and number. The motivation for ICAO-CAEP engine emissions standards is to improve local air quality in and around airports and remains a concern for policy makers (see Section 5). There is additional concern over the climate effects of nvPM and NO_x emitted at altitude (as discussed in Section 4). As a preliminary hypothesis, it is assumed that reducing emissions of these pollutants during the LTO cycle will also reduce them at altitude, an assumption that is under constant review at the CAEP technical working groups.[36]

The regulatory emission standard for NO_x is denoted 'Dp/Foo', which is the mass, in grams (Dp), of NO_x emitted during the reference LTO cycle, divided by the rated output (Foo) of the engine (kN thrust). The NO_x regulation (Dp/foo) has an allowance for increased overall pressure ratio (OPR) of the engine (see Fig. 2).

For nvPM there are two regulatory emission standards. One is the maximum nvPM mass concentration during the LTO cycle (the CAEP/10 nvPM standard), which replaces the old smoke number standard based on visibility of the exhaust. The other is LTO nvPM mass and nvPM number standards denoted as total nvPM mass (in mg) and particle number emitted during the LTO cycle per rated kN thrust (the CAEP/11 nvPM standards).

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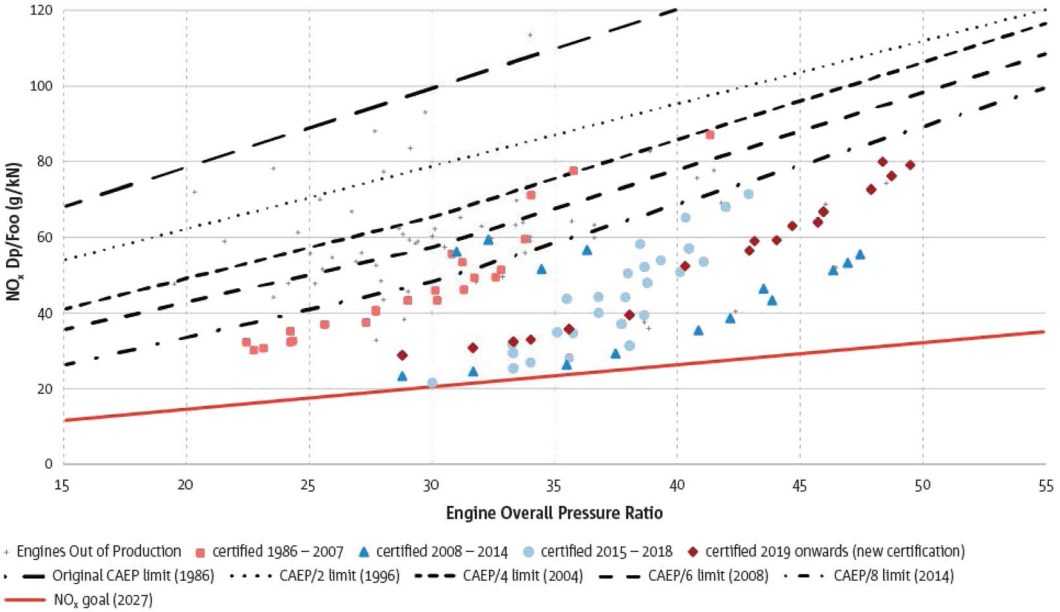

Fig. 2 Engine NO_x emissions certification data[39] showing the metric as ordinate and the pressure ratio as abscissa. The lines represent the various CAEP standards and goals.

The nvPM standards do not have an allowance for OPR but do include an allowance for the smaller rated thrust engines (below 200 kN for in production engines and 150 kN for new type engines) in recognition of the difficulty of scaling relevant technologies to smaller engines.

There is strong pressure to reduce the direct operating costs of aircraft through improved fuel efficiency, and this has largely driven the direction of aircraft engine technology development towards large turbo-fan engines, with the consequence of increased temperatures and pressures at the combustor inlet. As noted above, the NO_x regulation (Dp/Foo) has an allowance for increased OPR of engines in recognition of this. The trend for increases in OPR, means that the total amount of NO_x emitted per unit rated thrust (see Fig. 2) and per LTO cycle is still increasing. Fig. 2 shows that most engines since 2000 have been comfortably below the regulation level for NO_x. There is work currently underway in the relevant CAEP emissions/technical working group (WG3) to consider future emissions reductions through the regulatory standards (known as 'stringencies', within CAEP) that would lower the regulatory levels for NO_x after 2025. Similarly, changes to nvPM regulations post-2025 are being considered.

For NO_x, work is currently being undertaken within CAEP to consider a potential new metric that does not make an OPR allowance. The driving impetus behind this is two-fold. Firstly, increasing emissions of NO_x during LTO may be a concern for local air quality around airports. Secondly, some research suggests that emissions of NO_x at altitude may affect surface air quality, although the significance of such attribution is uncertain (see Section 5 and Appendix 1). Moreover, it has been long-established through modelling that cruise NO_x emissions can enhance tropospheric O_3 formation, affecting atmospheric chemistry and climate (see Section 4). Because of these considerations, the potential need for an additional emission reporting point beyond the conventional four, that better represents cruise conditions, is being considered.

The impact of nvPM on local air quality continues to be of concern although the relative contribution from aviation emissions to ambient levels of particulate matter is highly airport/location-specific and depends largely on background emissions. Combustion emissions of nvPM from modern engines (jet engines or road vehicles) tend to be smaller than an equivalent diameter of 50 nm, whereas most monitoring of particulates is as a mass concentration of particles less than 2.5 μm. As a result, ambient air quality regulations and most monitoring data do not reflect the contribution made by these very small particles (see Section 5). The (indirect) contribution of nvPM during cruise to contrail formation is also a consideration in future policy and may provide added motivation for reviewing the regulatory levels of the nvPM engine emission standards.

Potential tightening of the regulatory levels for the NO_x and nvPM mass/number standards is being considered for the next CAEP cycle[37] depending on the outcomes of the ongoing technical work at CAEP at the time of writing[38] and the combustion technology progress.

4.2 Combustion technology, fuel composition and emissions

The regulatory consideration of aircraft non-CO_2 emissions is currently dominated by NO_x and nvPM. These emissions are determined by the temperature and pressure of air entering the combustor, the temperature leaving the combustor, the design of the combustor and the nature of the fuel used. The difference between entry and exit temperature depends on the fuel–air

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ratio and it increases as the thrust of the engine is increased. To design a more efficient engine it is necessary to increase both the overall pressure ratio and the combustor exit temperature and in any likely scenario the incentive to increase both will remain.

In all combustors there is more air than is required for stoichiometry; stoichiometric combustion would result in temperatures of about 2600 K whereas even the latest turbines are limited to about 2000 K. The formation of NO_x is slow, compared with other chemical reactions in the combustor, but it rises rapidly above 1700 K. The formation of NO_x therefore requires regions of high temperature where the mixture is maintained with excess oxygen. It is necessary to distribute the fuel widely in the air, ideally to form a near-uniform mixture; non-uniform pockets can approach a stoichiometric mixture which will reach very high temperatures.

nvPM are formed in the fuel rich regions of the combustor and to remove them it is necessary to maintain the mixture at high temperature for as long as possible to burn up the particles of carbon. Except for the requirement for good mixing of the fuel with the air, the requirements for low nvPM are therefore in conflict with those for low NO_x.

The design and development of combustors is very costly and time consuming, taking perhaps two decades from start to finish. Manufacturers therefore like to continue to use the same combustor, or style of combustor from one engine to another. There are two basic types of combustors in use now, the Rich-Burn, Rapid Quench, Lean-Burn (RQL) and the lean-burn type (see Fig. 3).

In the RQL combustor, the fuel initially burns in a rich region which is rapidly diluted ('quenched') to a lean mixture; in the rich region there is insufficient oxygen to form NO_x and in the diluted region the temperature is intended to be low enough to avoid the formation of NO_x. Only in the brief period when the mixture passes through stoichiometric is NO_x formation appreciable; the process relies on the chemical reaction to form NO_x being relatively slow compared with the other major chemical reactions. The production of nvPM, occurs in the fuel-rich regions of the combustion system and the particles are then mostly consumed in the leaner regions of the combustor. Burning up the nvPM requires the temperatures in the latter regions of the combustor to be high, which is in direct conflict with the requirements for minimizing NO_x production. The mixing is never wholly complete and the level of NO_x and nvPM depends on how uniform the lean region is. However, this inherent trade-off in the RQL combustors, which were originally conceived for NO_x reduction, means that the controlling nvPM at the same time as reducing NO_x provides design challenges.

In the lean-burn combustor, excess air is distributed around the fuel injection so that the mixture is always lean, except for the pilot injector, and temperatures are held well below stoichiometric. When the combustor exit temperature becomes high, approaching 2000 K, the time for full combustion also gives time for NO_x formation and NO_x levels appear to rise above the best RQL combustors. The lean-burn combustor also needs a pilot stage for use at low power (*i.e.*, low fuel-flow) conditions. Except during pilot-only operation, downstream

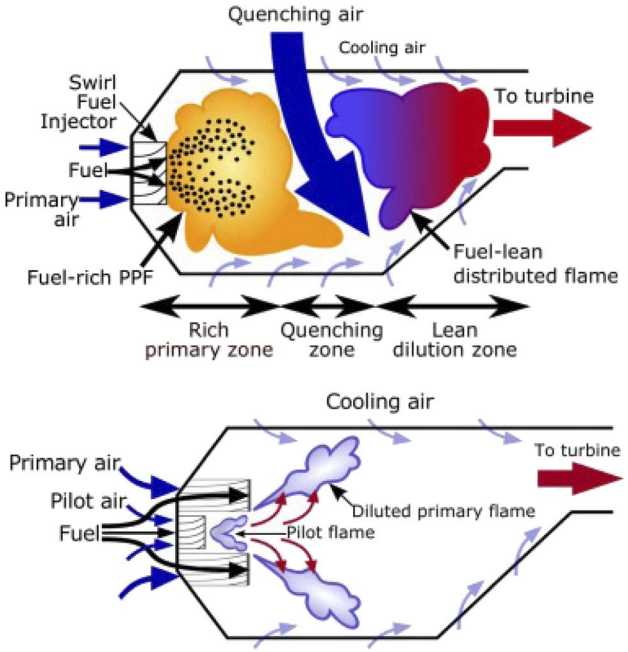

Fig. 3 Sketches of combustion chambers from Bergthorson and Thomson (2015),[40] showing RQL (top) and TAPS lean burn (bottom) combustors.

lean-burn regions promote burn-out of any particles formed by the pilot, so that exit levels of nvPM should be low. GE have installed lean-burn ('TAPS') combustors (see Fig. 3) in some recent engines and, very low levels of NO_x are found for low thrust versions of engines. However, the NO_x rises rapidly as the thrust, and turbine entry temperature, rises. Recent ground-based certification measurement data confirm that nvPM mass and number levels are low for these combustors.

At present, RQL combustion systems are more widely used and based on the physical understanding of the processes in the combustor, the tradeoff between NO_x and nvPM should be more significant than for lean burn engines.[41]

Water injection has also been suggested as a means to limiting NO_x emissions.[42] Water ingestion into the air stream ahead of the combustor will lower the temperature of the air entering the combustor and will therefore reduce the formation of NO_x. Water ingestion can also lead to an increase in engine efficiency and thrust. During the take-off cycle, when NO_x emissions tend to be important, it is a possible remedy, but with considerable extra cost and complexity to the powerplant and additional issues associated with operations.

A more recently investigated unregulated emission that may play a role in overall particle emissions is that of engine lubrication oil venting. There is a device to separate lubricating oil from the air which gets entrained – the bearing chambers allow air to enter them so that oil does not spill out in the gas path of the engine. The separator has a breather pipe to take away the air and the end of the pipe needs to be a low-pressure region, venting to atmosphere. The vent can be in many places, sometimes in the bypass duct and sometimes on the outside of the

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nacelle. One solution is a vent pipe in the centre of the core jet flow leaving the engine. The oil mist will get drawn into the plume, and thence into the vortex.

4.3 Critical decisions in future technology development and possibilities

The issues associated with emissions of NO_x and nvPM on air quality near airports are relatively straightforward in that there are emissions regulations and ambient local air quality standards to be complied with. Attributing measured levels of the pollutants to aircraft emissions is much more difficult, and attribution can only be determined with dispersion models (see Section 5) since there is no unique tracer associated with aircraft emissions.

For the effect of emissions on climate, the issues are far more complex and continue to be assessed (see Section 4). In the era of net zero, the reduction and elimination of fossil-based CO_2 emissions is the primary policy requirement associated with climate protection. How the aviation sector responds to this will have consequences for its non-CO_2 effects, and therefore policies to reduce emissions of NO_x and nvPM. If the drive for CO_2 reductions includes large and widespread uptake of SAF, then nvPM emissions will be reduced as a potential co-benefit even without combustion technology developments. This is because SAF can have lower or even zero amounts of sulphur and aromatic hydrocarbons, and soot is preferentially formed by the combustion of aromatics.

NO_x emissions would not be impacted by SAF fuel types and if the effects of NO_x on both climate and local air quality continue to be of concern, there would therefore be continued pressure to use combustor technology to reduce its levels. In this SAF-led CO_2-reduction scenario, the fuel impacts on decreasing nvPM would provide a wider design space for the combustion engineers to focus on NO_x reduction technology. However, the tension between OPR and turbine entry temperature, to reduce fuel-burn and CO_2, and NO_x would still present a challenge.

If direct air capture (DAC) of CO_2 with permanent removal becomes viable (see Section 6.2), fossil-based kerosene may continue to be used. In that case, the present issues of NO_x and nvPM would remain and the policy pressure to control both would presumably make cleaner combustion produced by better technology a priority. In the case of continued use of fossil-based kerosene, the lean-burn and more advanced RQL would continue to be developed.

5 Aviation emissions and effects on the atmosphere–climate

5.1 Aviation effective radiative forcing and its relation to current and historical aircraft emissions

The most frequently used measure of the climate importance of different aviation-induced atmospheric effects (CO_2, contrails, *etc.*) is radiative forcing (RF) which has units of watts per square metre (W m^{-2}). It characterizes the size of the perturbation to the planetary radiation budget due to the effect, relative to some prior period, typically pre-industrialization. The climate system then responds to RF leading to, for example, surface temperature change (a positive RF leads to a warming, and *vice versa*). The concept is discussed at length in IPCC assessments.[31,43]

RF enables a comparison of the size of different climate-change drivers, *e.g.*, greenhouse gases, and also both within and between sectors. RF is proportional to the equilibrium surface temperature change relative to (say) pre-industrial temperature ($T - T_{pi}$), that would result if that RF was applied for many decades, so that:

$$T - T_{pi} \approx RF/\lambda$$

where the constant of proportionality, λ, is the equilibrium climate feedback parameter in W m^{-2} K^{-1}. The actual value of λ is a chronic uncertainty in climate science (see also Box 7.1 of Förster *et al.*, 2021).[31] Hence it is more straightforward to compare RF than temperature change between studies which may use a different value of λ. A similar expression can be used to represent temperature changes in response to a constant forcing over shorter time periods, but with a different constant of proportionality.

It is important to note that this expression is valid when applied to global mean forcing and temperature response. The local temperature response is dependent to only a limited extent on the geographical pattern of RF (see also Section 4.5.1), but it is significantly dependent on climate feedback mechanisms which are, for example, responsible for the so-called 'Arctic Amplification' of the pattern of climate change whereby the Arctic warms at roughly twice the rate of the global-mean warming.[44]

There are additional reasons for using RF rather than surface temperature in comparisons. If state-of-the-art Earth System Models (ESMs) were used to characterize the surface temperature impact of aviation climate-change drivers, there would be many difficulties. ESMs simulate many interactions in the climate system: between weather systems of different spatial scales, and between the atmosphere, oceans, and land surface. By their nature, they therefore simulate climate 'noise'—that is, unforced natural climate variability. Distinguishing the temperature signal of relatively small forcings, *e.g.*, aviation contrails, from this noise is difficult, requiring long integrations of computationally intensive ESMs and/or performing experiments with artificially inflated perturbations. The computational expense also means that radiative calculations in ESMs are of reduced complexity and may not include all relevant processes. Also, it is difficult to perform sensitivity experiments in ESMs to fully capture the effect of uncertainties in the drivers of climate change.

Since the early uses of RFs, conceptual refinements in its definition have improved its use as a comparative measure, so that the global-average temperature change from, for example, a 1 W m^{-2} perturbation due to contrails, is more similar to that resulting from 1 W m^{-2} due to aviation-induced CO_2 changes. Recent IPCC assessments[31,43] have adopted effective RF (ERF) as the preferred measure, as it incorporates adjustments resulting from an RF (*e.g.*, in cloudiness) that occur on a more rapid

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timescale than resulting surface temperature changes (which occur over periods of decades), see Fig. 4. ERF is still a concept in development; in general, rapid adjustments can only be calculated by using an ESM, and so are subject to the difficulties noted above, and these adjustments can also vary significantly between different ESMs. Diagnosing ERFs from ESM integrations suffers from some of the same problems as mentioned in the previous paragraph for diagnosing temperature change. The ERFs have to clearly emerge from the ESMs climate 'noise'. The reduced complexity of radiative transfer processes needed in ESMs may mean they do not fully represent all climate forcing mechanisms. Moreover, even for ERF, studies find that λ is not identical across all forcing agents.[43,46]

For some climate change mechanisms, notably CO_2, the difference between RF and ERF is small (order 10%). For others, notably contrails, results from the rather few available ESM experiments are not in full agreement, but a consensus has emerged that the ERF is around half the RF; this adds additional uncertainty to characterization of contrail climate effects. We adopt ERF as the chosen metric.

Two other aspects of RF and ERF must be emphasized:

• First, although rooted in observational science and evaluated where possible by comparison with observations, RFs are largely a model-based construct. Quoted uncertainties emerge from a mixture of modelling sensitivity experiments, comparison between similar calculations performed by different groups (and more limited benchmark calculations) and expert judgement on the importance of poorly constrained processes.

• Second, RF is a measure of the effect of changes in, for example, CO_2 or contrails, between two times. Because there were no significant aviation emissions prior to 1940, our chosen metric is the change between 1940 and the present-day (defined here as 2018). It is, therefore, a snapshot at 2018 (with reference to Fig. 1) and it is important to recognize that the timescale of emissions driving ERF depends on the atmospheric residence time of the particular emission. For CO_2, a significant fraction ($\approx$80%) of emitted CO_2 effectively remains in the atmosphere for decades to centuries, with around 20% persisting for more than 10 000 years; it is a persistent, or cumulative pollutant. Therefore, the CO_2 RF reported for 2018 (Fig. 1) includes the effect of emissions dating back to the early days of aviation. And even if CO_2 emissions remained constant, CO_2 concentrations and the resulting RF would continue to grow (see Section 2.2 and Fig. 5).

By extreme contrast, typical persistent contrail lifetimes are several hours, and so the RF for 2018 (Fig. 1) results from very recent aviation; contrails resulting from emissions even a few weeks earlier do not influence ERF. In contrast to CO_2, if contrail-formation rates remained constant, then the RF associated with them would also remain constant, as is illustrated in Fig. 5, which illustrates the change in non-CO_2 RF and CO_2 RF under a hypothetical constant emission scenario. This particularly highlights the fact that the Radiative Forcing Index (the ratio of total aviation RF to the aviation CO_2 RF) is not a fixed constant but depends on the emissions scenario. Fig. 6 illustrates the warming impact of aviation[7] resulting from different growth scenarios, accounting for both CO_2 and non-CO_2 forcing. It shows how the fractional contribution of non-CO_2 forcings to aviation's total radiative forcing and hence warming impact (contrast the length of the green and purple bars on the right-hand side of this figure) is strongly scenario-dependent, and depends on the rate of growth of aviation, even assuming no change in fuel composition. In Section 7, we describe the full impact of aviation in terms of 'warming-equivalent' CO_2 emissions, meaning the CO_2 emissions that would have the same impact on global temperatures over these multi-decade

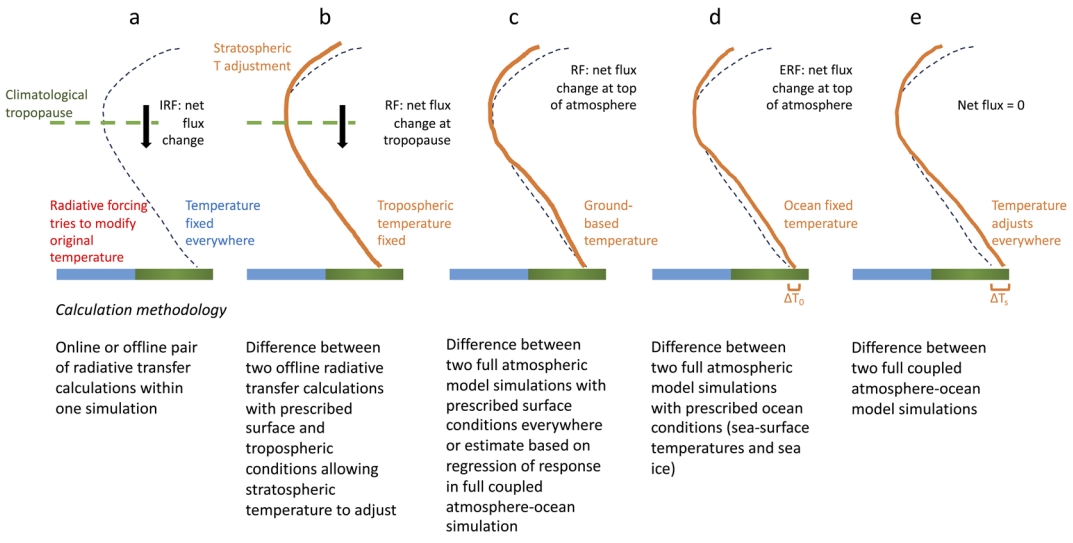

Fig. 4 Schema comparing (a) instantaneous RF (IRF), (b) RF, which allows stratospheric temperature to adjust, (c) flux change when the surface temperature is fixed over the whole Earth (a method of calculating ERF), (d) the ERF calculated allowing atmospheric and land temperature to adjust while ocean conditions are fixed and (e) the equilibrium response to the climate forcing agent. The methodology for calculation of each type of forcing is also outlined. ΔT_0 represents the land temperature response, while ΔT_s is the full surface temperature response. Based on Myhre *et al.* 2013,[31] their Fig. 8.1.

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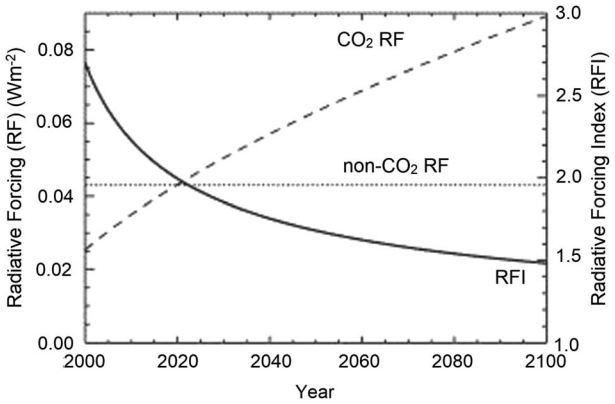

Fig. 5 The development of aviation radiative forcing for hypothetical constant CO_2 and non-CO_2 emissions at 2000 levels, showing that a non-CO_2 forcing, such as from contrails would equilibrate, whereas CO_2 emissions continue to accumulate in the atmosphere and therefore the radiative forcing attributable to CO_2 continues to increase. The RFI (radiative forcing index) on the right-hand axis is the ratio of the total (non-CO_2 plus CO_2) forcing divided by the CO_2 forcing which can be seen to fall continuously over the time period of the plot (Förster *et al.*, 2006).[47]

timescales. The impact of these differing lifetimes is important not only for understanding RF but also in the design of other CO_2-equivalent (CO_2-e) metrics to compare aviation climate effects including the Global Warming Potential (Section 7).

5.2 Atmospheric lifetimes of aviation non-CO_2 forcing agents

A further important distinction, partly related to differing lifetimes, is the large dependence of non-CO_2 forcings on the location and, sometimes, time of year or even the time of day of aviation emissions. Because of the long lifetime of CO_2, its climate effect is assumed to be independent of the location of emissions; the timescales for the mixing of CO_2 both vertically and horizontally are small compared to its long lifetime.

Contrails: persistent contrails and contrail cirrus require specific atmospheric conditions. First, the Schmidt-Appleton criterion must be satisfied,[48] where the mixing of engine exhaust air with environment air leads to saturation and short-lived contrails. This depends on the temperature and humidity of both the exhaust and the environment, and hence depends on the engine type and settings, and the fuel used. Second, for short-lived contrails to persist, the environmental air has to be 'ice-supersaturated'; this means there is enough water vapour in the atmosphere to condense to form ice clouds, but there are insufficient 'nuclei' for the vapour to condense on. Ice-supersaturated regions (ISSRs) are relatively rare (typically 10–15% of the time over the UK), and only exist in certain weather conditions, and can be very patchy in both vertical and horizontal extent, typically a few hundred metres in depth and 100–300 km in horizontal extent.[49] Moreover, these ISSRs have their own internal variability.[50] Using 1 Hz (~200 m horizontal resolution) aircraft-based observations, Diao *et al.*, (2014)[51]

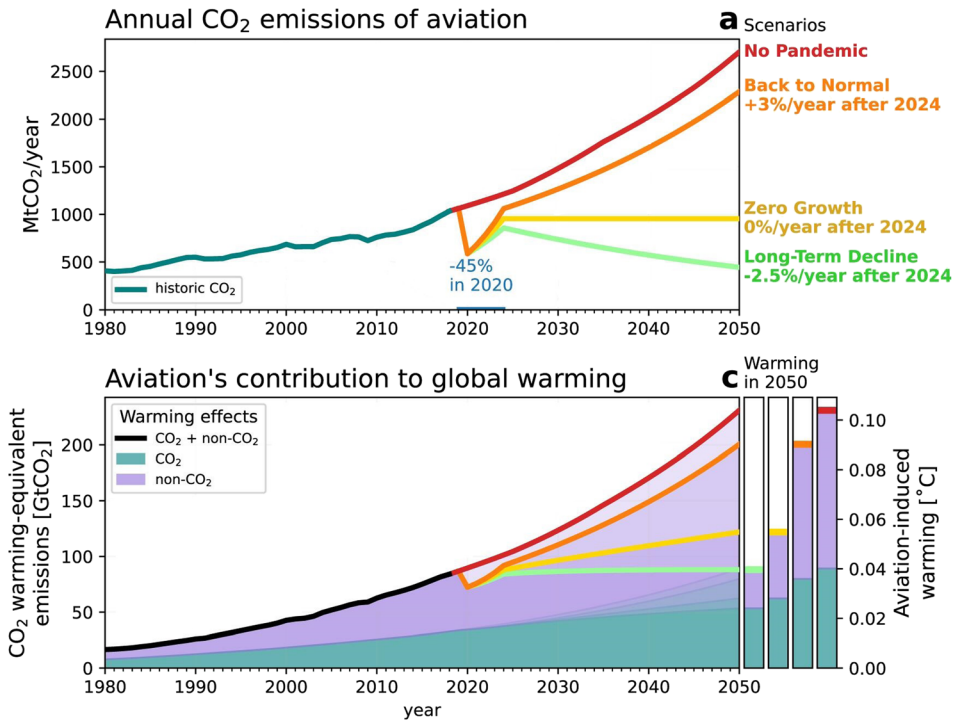

Fig. 6 Aviation's contribution to global warming to 2050 using a simple equation relating forcing to temperature following four scenarios: no pandemic, back to normal, zero long-term growth, and long-term decline (upper panel). The lower panel shows the cumulative warming-equivalent emissions of CO_2 and non-CO_2 effects of aviation since 1940 and the corresponding aviation-induced global warming. Scenarios are colour-coded as in the upper panel (figure adapted from Klöwer *et al.*, 2021).[7] This figure helps to illustrate how the fraction of non-CO_2 warming equivalent emissions to the total changes over time, according to the CO_2 emissions scenario (see especially the right-hand side of lower panel).

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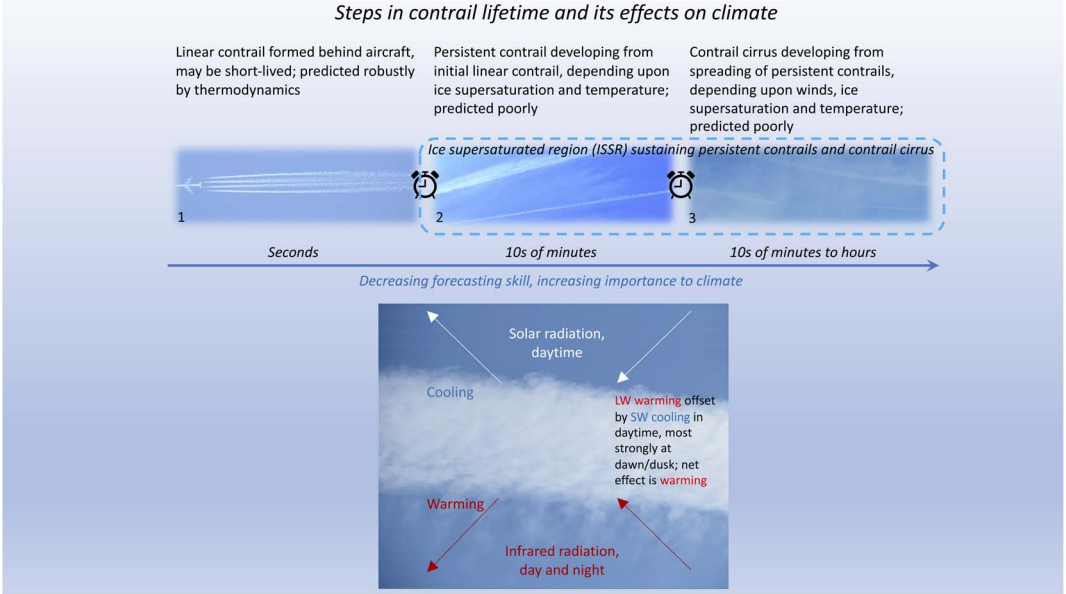

Fig. 7 The different timescales of contrail formation and potential transition to persistent contrails forming contrail cirrus. As shown in the lower panel, the net forcing is the residual of short wave and long wave radiation processes, which is time and space variant. In general contrails tend to have a net negative forcing during the day, and a net positive forcing at night, which is considered to dominate the overall time-integrated forcing (based on Shine and Lee, 2021).[57]

showed that the horizontal scale of ISS has mean and median lengths at ~3 km and 1 km, respectively, *i.e.*, a more heterogeneous spatial structure of ISS conditions than previously reported, of an ~150 km median length.[52] Tan *et al.* (2016)[50] point out (that) "The influences of these microscale ISS, which are on the subgrid scales of most climate model simulations (~10–100 km), have yet to be quantified."

An issue relevant to contrail-avoidance strategies is that not all weather forecast models include the atmospheric processes necessary to represent ISSRs (*e.g.*, the effects of gravity waves);[53] even if they do, it does not mean they can be forecast with sufficient accuracy,[54,55] or resolution.[50] Amongst other things, the meteorological forecast models require good observations (or technically, a good assimilation)[56] of the atmospheric conditions to initiate the forecasts. Some aspects of the timescales of formation and forcing response are shown in Fig. 7.

A further significant complication with calculating contrail RF is that contrails reflect incoming solar radiation (causing a negative forcing) but also absorb thermal-infrared radiation emitted by the underlying surface and atmosphere (causing a positive forcing); the net forcing is a relatively small residual of these two terms.[58] While there is high confidence that the global-mean contrail forcing is positive, the net forcing is positive at night, predominantly negative at dawn/dusk (when the solar zenith angle is large), and there is a near balance between positive and negative forcing during the day, which can result in either a net positive, or negative, depending on the particular conditions. Overlaid on the physical conditions are variable patterns of diurnal traffic amounts.[59] This depends not only on the location of the aircraft emission but also on the duration of the contrail. The net forcing of an individual contrail could change due its advection by the wind into regions where the balance is different, or due to the change of the position of the sun in the sky.

A further difficulty with contrails is that the condensation of water vapour in ISSRs can affect natural cirrus clouds that would otherwise have formed in the absence of contrails.[60] This complication is in principle accounted for by the rapid adjustments that are part of the ERF definition, but present evidence indicates that different climate models would likely simulate these adjustments in different ways. The effect of such adjustments will be missing from all studies that calculate RF rather than ERF.

Results from a recent state-of-the-art global model calculation[61] of the contrail cover and RF (not the ERF) using a 2006 flight inventory are shown in Fig. 8. Local 'hot spots' where the net RF reaches 1 W m^{-2} can be seen in regions of high air traffic over Europe and eastern North America. Averaged over the globe, the net RF is about 20 times smaller (44 mW m^{-2}), made up of a longwave RF of 85 mW m^{-2} and a shortwave RF of −41 mW m^{-2}.

5.2.1 NO_x emissions. In a similar, but arguably less extreme way, the radiative forcing by NO_x emissions depends on atmospheric conditions. Emissions of NO_x from aircraft result in changes in atmospheric composition through chemical reactions.[62,63] *Via* these reactions, emissions of NO_x result in an increase in tropospheric ozone (resulting in a positive forcing), and a decrease in ambient CH_4 (emitted from other surface sources), since OH is formed, the principal sink term of CH_4 (ultimately forming CO_2 and water vapour), which can be accounted as a 'negative forcing'. Secondly, the increase in OH, reduces methane's lifetime, so less of it reaches the

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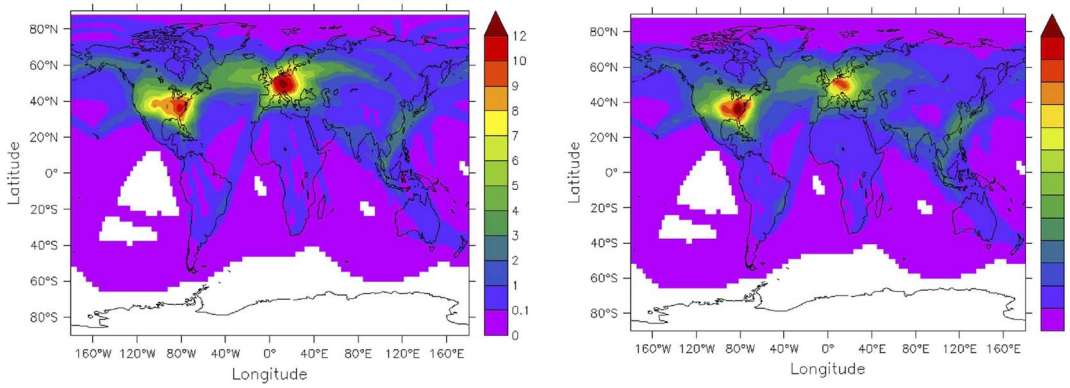

Fig. 8 Model simulation of the global distribution of contrail cirrus coverage associated with a visible optical thickness ≥0.05 in percent (left hand panel) and the net radiative forcing due to contrail cirrus in mW m^{-2} (right hand panel). Taken from Fig. 5 of Bier and Burkhardt (2022).[61]

stratosphere and hence there is less production of stratospheric water vapour from methane oxidation and a small reduction in background tropospheric ozone, since CH_4 provides peroxy radicals involved in the catalytic production of ozone.

Multiple modelling studies have demonstrated that the net forcing depends on the latitude, altitude, and time of year of emission as well as the emission amount.[64–69] This is largely because of variations in the chemical and photochemical conditions of the air. Fig. 9 shows spatial maps of aircraft emissions, ozone perturbation and short-term ozone radiative forcing, which exhibit different characteristics. The NO_x emissions reflect the main routes and frequency of emissions. While the ozone pattern

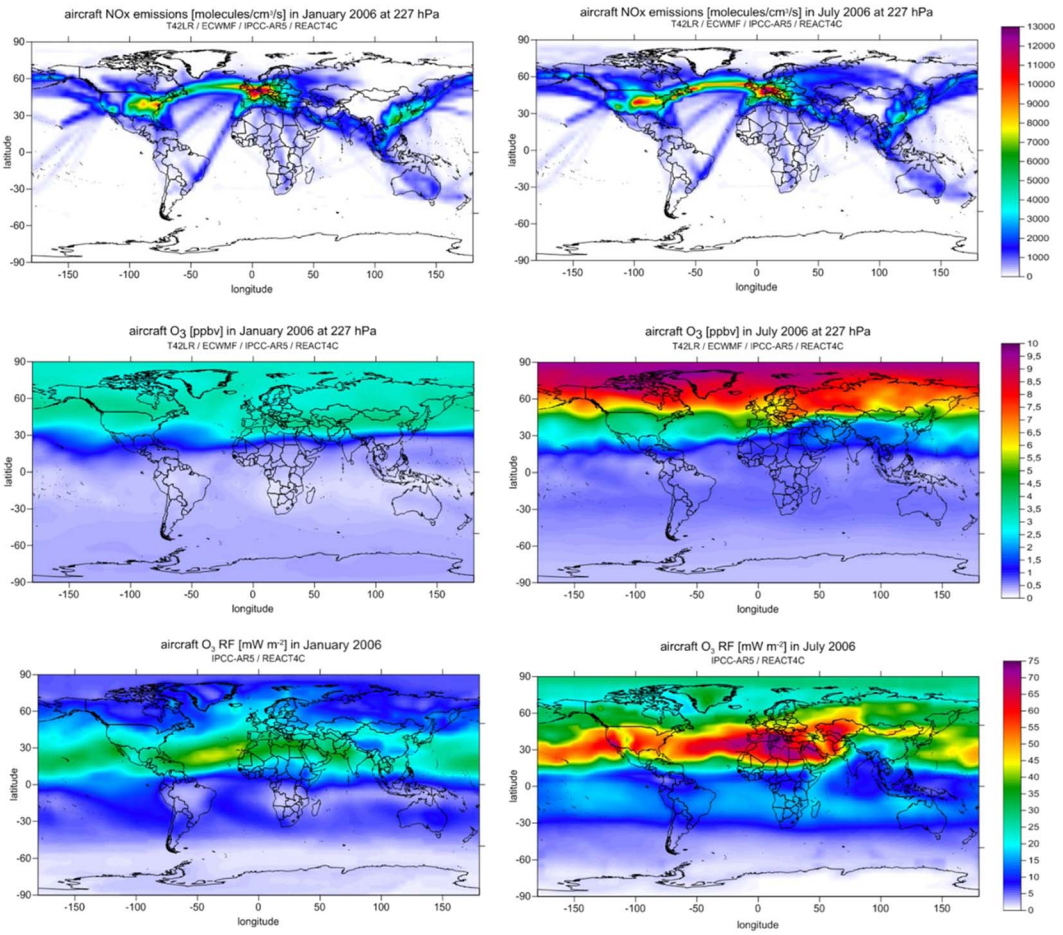

Fig. 9 Simulations of global aircraft NO_x emissions in 2006, January (upper left-hand panel), July (upper right-hand panel), ozone concentrations attributable to the aircraft NO_x at 227 hPa (approximately 11 km) (middle panels) in ppb, and the consequential radiative forcing (lower panels) (data replotted from simulations described by Skowron *et al.*, 2015).[69]

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broadly reflects the NO_x emission pattern, it is much more spread out and reflects both the distribution of emissions, ozone lifetime of several weeks, and the ozone-production sensitivity of the atmosphere. Fig. 9 (lower panel) shows the resultant pattern of radiative forcing from the ozone perturbation, which is shifted southwards. Both the distribution of ozone (ppb) and its sensitivity to aircraft NO_x (ppt) can be seen in Fig. 10, which shows 2D averaged 'slices' of the atmosphere (latitude by height) of aircraft

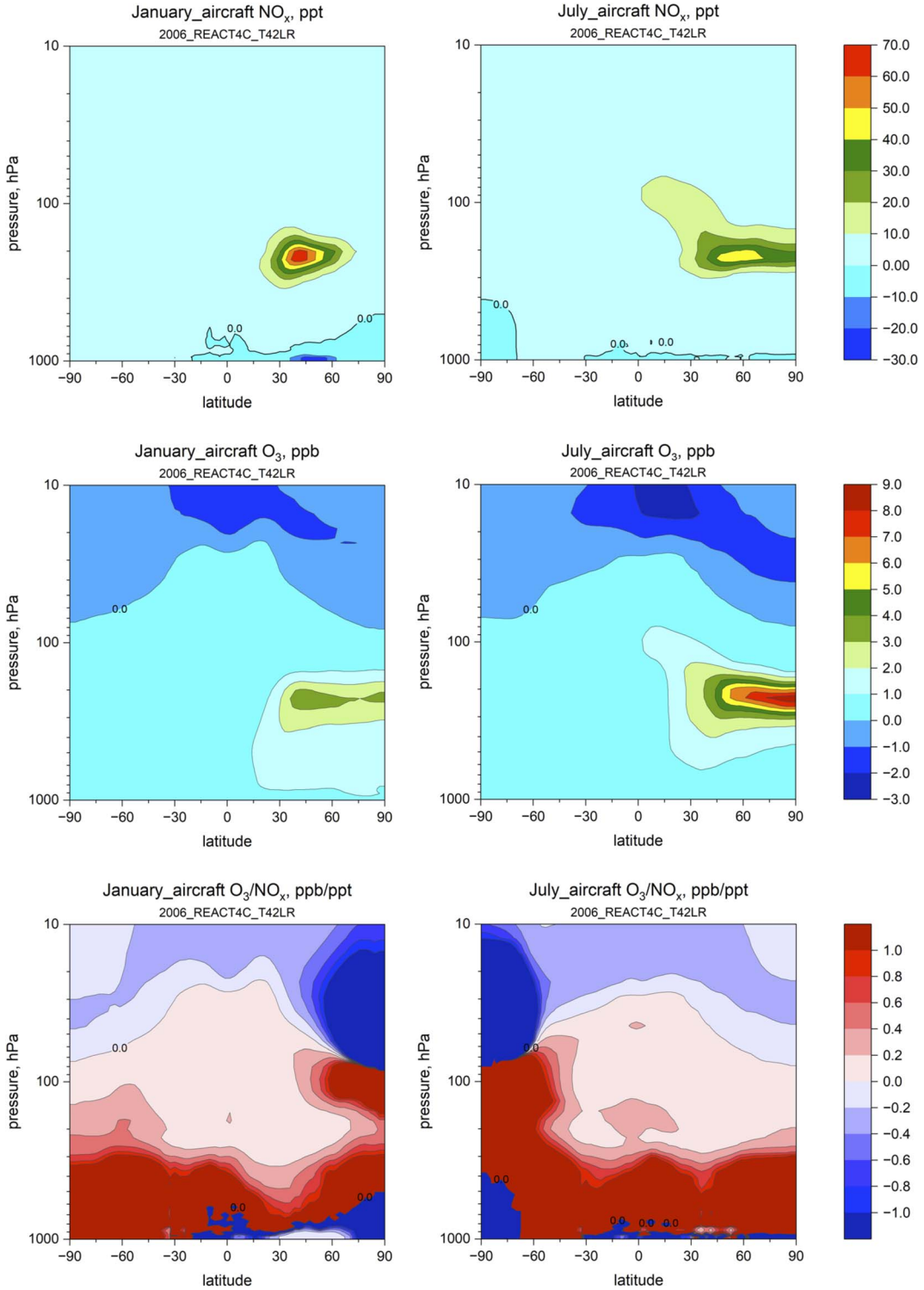

Fig. 10 Two-dimensional (latitude by height) averaged NO_x concentrations (VMR – volume mixing ratios) from aircraft (upper panels, left–right, Jan and July) in ppt, the resultant ozone concentration perturbations (middle panels) and the sensitivity of ozone to NO_x (ozone ppb/NO_x ppt). Data from Skowron *et al.* (2015)[69] simulations.

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NO_x and ozone perturbation concentrations, and the ratio of O_3/NO_x. The ratio plots (lower panels of Fig. 10) show that the sensitivity of the atmosphere in terms of ozone production per unit NO_x is not evenly distributed because of sensitivity to photochemistry and background concentrations of other precursors, and the magnitude of the aircraft NO_x itself (see Lee *et al.*, 2010, Section 5.3).[63] Superimposed on the ozone perturbation is the latitudinal sensitivity to net ozone (short wave + long wave) forcing, centred at the equatorial tropopause, as shown in Fig. 11 (taken from Rap *et al.*, 2015).[70] As with contrails, it is not only when and where the emission occurs, but the weather conditions at the time of emission and specifically the subsequent trajectory of the air as it is blown by the winds.[71] In general, air advected towards the equator will lead to a greater RF, both because the photochemical reactions become more rapid and because a given ozone perturbation leads to a greater forcing per unit ozone change (see above and *e.g.* ref. 65 and 66). For related reasons, a given NO_x emission in northern summer leads to a greater ozone forcing than a wintertime emission (see Fig. 9).

However, a complication with NO_x emissions, especially in the use of CO_2-e metrics such as GWPs and Global Temperature-change Potentials (GTPs) is that the effect (shown as positive in Fig. 12) depends strongly on the time horizon of the emission. The positive forcing due to initial formation of ozone is gradually counteracted by the negative forcing resulting from reduction of methane (and the consequent reduction in ozone) leading to a net negative GTP for time horizons between approximately 20 and 60 years, increasing to small positive/close to zero values at around 100 years, plus.

5.2.2 H_2O emissions. Fig. 1 shows that the direct radiative forcing of water vapour emissions (distinct from their effect on contrails) is currently assessed to be small. A major determinant of this effect is the height of emission relative to the tropopause. The residence time of water vapour is short (a few days) when emitted into the troposphere and lower stratosphere where current subsonic aircraft fly, but months to years when emitted higher into the stratosphere at typical supersonic cruise altitudes of 20 km, plus.[72]

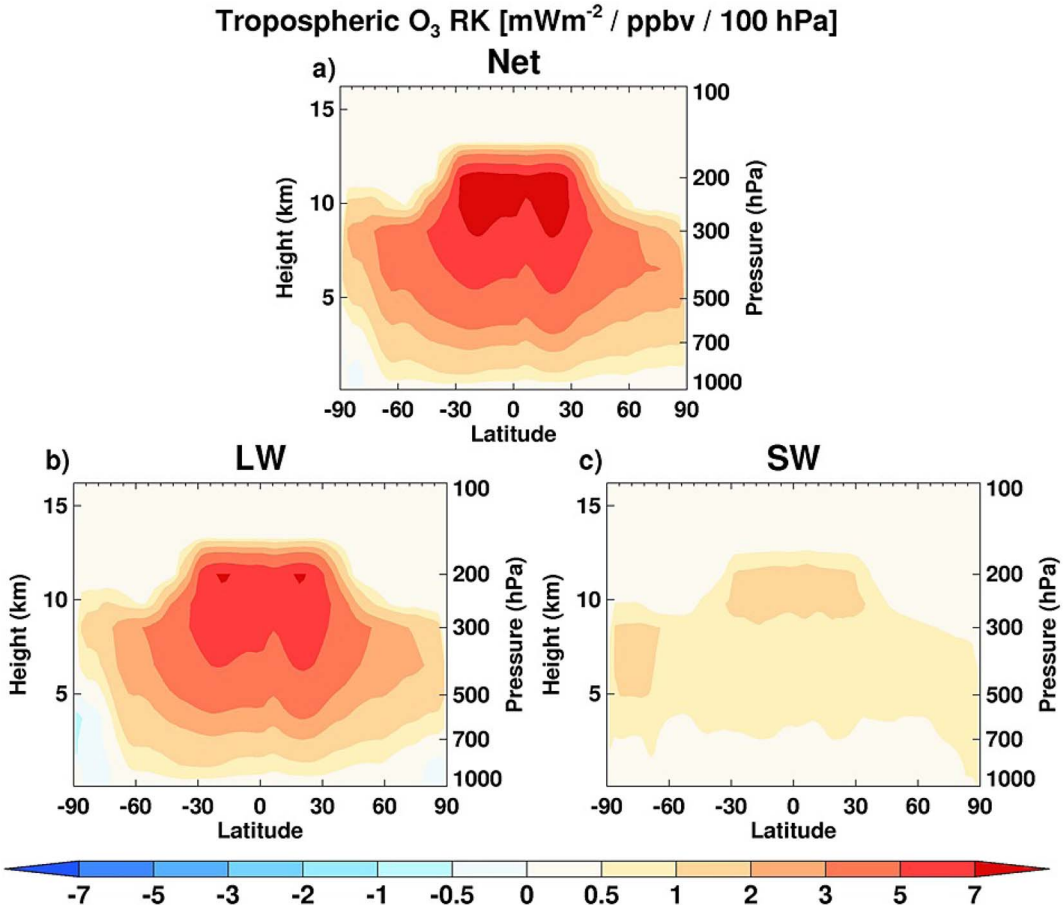

Fig. 11 Annual zonal mean tropospheric ozone radiative kernel (RK)[75] under all-sky conditions in units of mW m^2 ppb^{-1}/100 hPa for (a) net (LW + SW), (b) LW, and (c) SW. SW (short-wave) refers to the RF resulting from changes in the absorbed solar-radiation by the Earth-atmosphere system, which is mostly incident in the wavelength range 0.2 to 4 μm. LW (longwave) refers to changes in the outgoing longwave radiation at the top of the atmosphere; LW radiation (mostly in the wavelength range 4 to 400 μm sometimes) originates from emission by the Earth and its atmosphere and is modulated by absorption by atmospheric constituents. Radiative forcing is the sum of the individual SW and LW RFs. Note that this illustrates the general radiative response from tropospheric ozone, formed from precursor emissions of NO_x, CO, CH_4, HCs and is not an aviation perturbation. From Rap *et al.* (2015).[70]

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5.3 Uncertainties and confidence levels in the aviation radiative terms

5.3.1 Uncertainty assessment. The overall ERF for aviation has recently been re-assessed and calculated for 2018 as part of a large international collaborative exercise;[6] these values were adopted as the basis of the assessment of aviation non-CO_2 forcings in IPCC AR6 WGI[78] and are shown in Fig. 1. The CO_2 term was derived from the mean of three simple carbon-cycle models that determine the accumulation rate of CO_2 in the atmosphere from anthropogenic emissions, and the non-CO_2 terms were either assessed from literature values or recalculated, being careful to normalize the forcings to assumed emission rates and emission indices to the same year (see details and ESI of Lee *et al.*, 2021).[6]

Although uncertainty bars for the individual forcing terms were given by Lee *et al.* (2021),[6] it is important to understand the necessary differences in approach to each term. So, for example, for the 'net NO_x' term, the combination of the positive ERF due to the short-term ozone response and for the longer-term methane (and associated stratospheric water vapour and longer-term ozone) negative ERF response, the uncertainties were based on a statistical analysis of model simulations (around 50, from a smaller number of models). However, the calculated spread of the model results does not necessarily capture uncertainties in underlying processes, so the calculated statistical variation is likely to be an underestimate of the overall uncertainties.

Moreover, while there is a good statistical sample of NO_x model results of perturbation and RFs, the RF to ERF adjustment is based upon only one coupled climate model simulation,[79] which is a highly unsatisfactory situation. In contrast, the contrail cirrus results were from only 3 model systems and 4 sets of results—the uncertainties were estimated based largely on expert judgement regarding the underlying processes.

So, for example, in the case of the underlying terms to the net-NO_x forcing, many model results were available and the uncertainty was the statistical uncertainty; in contrast, the uncertainties calculated for the contrail + contrail cirrus term of Lee *et al.* (2021)[6] were partially the result of expert judgement over processes represented in only three models.

In terms of these assessed uncertainties, they were combined in a Monte Carlo analysis, to represent the relative uncertainties of CO_2 and non-CO_2 terms. Fig. 13 shows that the non-CO_2 terms (combined) contribute 8 times more uncertainty than the CO_2 forcing term. Consequently, the uncertainty in the overall aviation ERF term is dominated by the non-CO_2 uncertainties.

The uncertainties in the science are also represented, in part, by the 'confidence levels' assigned to the forcing levels, using the 'matrix method' of the IPCC,[80] in which agreement (low, medium, high) and evidence (limited, medium, robust) are combined in a semi-subjective manner (see Tables 4a and b in Lee *et al.*, 2021).[6]

5.3.2 Contrails and contrail cirrus uncertainties. The uncertainties of contrails and contrail cirrus were described in detail in the ESI of Lee *et al.* (2021).[6] In assessing confidence levels, the contrail cirrus term had robust evidence for the phenomenon (visual and satellite observations), however the low confidence was driven by the small number of models and

Box 1: The COVID-19 pandemic and aviation contrails and cloudiness – what did we learn?

Several studies have attempted to use the COVID-caused reduction in air traffic to further understand contrail cirrus, and their properties and radiative forcing. Some of these have used cloudiness observations which potentially give new insight into the effect of contrails (and the COVID-related reductions). We argue that modelling studies of the effect of air traffic reductions essentially just reflect the current understanding of contrail forcing already encapsulated in Fig. 1 and thus add limited new insight.

Digby *et al.* (2021)[81] used satellite-derived data of cirrus cloud fractions, comparing data from 2003–2020 with data for the March–April–May period of 2020. The deviations of the 2020 (global-mean) cloud fractions were not deemed significant (at the 5%) level, given the relatively high interannual variability. This study, as do others, highlights a difficulty that weather conditions (for example, over Europe) in April 2020 were in any case anomalous, which makes it particularly hard to detect COVID-related changes. *Via* a statistical analysis, Digby *et al.* (2021)[81] were unable to detect a robust response of cirrus cloud fractions to aviation changes, which was interpreted as indicating that this change is smaller than expected from model-studies of contrail cirrus occurrence. They conclude with (in their words), a back-of-the-envelope estimate of the implications for (pre-COVID) contrail ERF, finding it to be 8 mW m^{-2}; this is only 10–20% of the Lee *et al.* (2021)[6] best estimate shown in Fig. 1 (although uncertainty ranges overlap).

Quaas *et al.* (2021)[111] used the same satellite data as Digby *et al.* (2021),[81] but focused on the 27–68°N region, separating this into sub-regions based on the change in air traffic in spring 2020; they compare this with data for the 2011–2019 period. In regions with the largest air traffic reduction, they found a 9 ± 1.5% reduction in cirrus fraction (*i.e.*, the cirrus fractional coverage decreased from about 32% to 29%), and a much more uncertain reduction in cirrus infrared emissivity. They used these observed reductions to estimate a (pre-COVID) global contrail-cirrus RF, *via* radiative transfer modelling, and concluded that the global-mean RF is 61 ± 39 mW m^{-2}; this is about one-half of the Lee *et al.* (2021)[6] RF value in Fig. 1, which they attributed to a lower contrail-cirrus coverage. Accounting for the ERF/RF ratio given in Fig. 1, this forcing is about 3 times higher than Digby *et al.*'s estimate, but with a substantial overlap in the uncertainty ranges.

Duda *et al.* (2023)[112] used data from the same satellites, focusing on the conterminous United States (CONUS), north Atlantic and north-east Pacific oceans, specifically to analyse linear contrail coverage, and their optical properties (derived in April and May 2020 with the same periods in 2018 and 2019). These observations are then used to compute the subsequent RF impact. Duda *et al.* (2023)[112] also used numerical weather reanalysis data to examine the extent to which changes in atmospheric conditions between these years may have affected the potential to form linear contrails and concluded that most (typically 70%) of the change in linear contrails was due to the change in air traffic rather than atmospheric conditions. They find that the linear contrail cover decrease depended on month, time of day and location, but in 2020 it was typically around 60% of that found in 2018 and 2019 (air traffic over CONUS was estimated to be 50% of that in 2018/2019 over CONUS and 20% of that over the north Atlantic). Changes in contrail optical properties were more modest (on average visible optical depths were 9% larger, but this depended on region and the comparison year), indicating that contrail cover was more affected than the contrail properties. The calculated changes in net RF again depended on month, time of day and location but typical values in 2020 were 50–70% of those in 2018/2019

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over the ocean areas and around 80% over CONUS. Duda *et al.* note that this decrease is much larger than found by Quaas *et al.* (2021)[111] but this was likely because Duda *et al.* specifically target linear contrails, whereas Quaas *et al.* (2021)[111] targeted thin cirrus of which a relatively smally proportion is due to aviation. Thus, this paper confirms that the impact of COVID is detectable in satellite observations of linear contrail cover which leads to a significant decrease in calculated RF; however, the relationship between changes in air traffic, contrail cover and RF is not a straightforward linear one.

In a model-based study, Gettelman *et al.* (2021)[113] consider the ERF impacts of COVID-related reductions in aviation during 2020. Their baseline ERF (62 ± 59) mW m^{-2} is close to the central value given in Fig. 1. Interestingly the impact of the COVID-related reductions on ERF, as they were most marked between March and August 2020, were judged insignificant. This was because of the degree of compensation between the solar (cooling) and thermal infrared (warming) components of contrail forcing (see Section 4.2) and the fact that the flight reductions were most marked in the northern hemisphere. Contrail (solar) cooling is largest in summertime, whilst the contrail (thermal infrared) warming is fairly constant through the year. The timing of the COVID-induced changes in 2020 meant a relatively large reduction of the solar cooling (due to fewer contrails) that essentially cancelled out the reduced thermal infrared warming.

Schumann *et al.* (2021)[114] (see also Voigt *et al.*, 2022)[115] focused on an analysis of geostationary satellite data over Europe, comparing March–August 2020 with March–August 2019. They noted that weather-induced differences were larger than aviation-induced differences. They claimed detection of an aviation signal but noted that "a quantitative assessment of the contrail model validity is beyond the information content of the data". Schumann *et al.* (2021)[116] present a model-based view of the event over the same area, finding that the model-based contrail-cirrus cover reduced from 4.6% to 1.4% between 2019 and 2020; this paper does not include any observational support for this value, but the 3% reduction is comparable to the value found by Quaas *et al.* (2021).[111]

Teoh *et al.* (2022)[117] performed a model-based study (using similar methodology to the Schumann *et al.* (2021)[116] modelling study over Europe) focusing on the North Atlantic region (defined as 40–75°N and 50–10°W). They examined the forcing during a COVID period defined as April 2020 to March 2021, calculating a reduction in contrail coverage from 0.4% to 0.14%. As with Gettelman *et al.* (2021)[113] they find compensating impacts in the solar and thermal infrared components, but nevertheless find a strong reduction in both RF and ERF (ERF was not explicitly modelled) which was reduced to one-third of their pre-COVID values. Given the different time period and region, it is hard to directly compare results, although Teoh *et al.* (2022)[117] assert that their results are consistent with Gettelman *et al.* (2021)[113] despite that paper finding little change in ERF.

Meijer *et al.* (2022)[118] applied machine learning to geostationary satellite over the contiguous USA, to derive contrail-cirrus coverage, contrasting data from 2018 and 2019 with 2020. For 2020 as a whole, they found a 22% reduction in contrail cirrus coverage in 2020 from the 2018–2019 total coverage of 0.26%, for a 36% reduction in distance flown above 8 km; the changes were most marked in April 2020, when contrail cover approximately halved. They also highlight the non-linear relationship between the two quantities, likely due to meteorological variability. Between 2018 and 2019 distance flown increased but contrail cover decreased. They did not attempt to quantify the impact of these changes on radiative forcing estimates.

Li and Groß (2021)[119] (see also Voigt *et al.*, 2022)[115] used lidar-based measurements of cirrus properties from the Cloud–Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) satellite, which compared data from 2014–2019 with March and April 2020, focusing mostly on Europe. The use of "pre-COVID" data over more years than in Schumann *et al.* (2021)[114] partially mitigates the role of meteorology variations in causing the Spring 2020 differences. Over Europe, mean cirrus thicknesses were lower (1.2 km *versus* the average 1.4 km) in March 2020, but this difference was much less marked in April; cirrus occurrence was also lower in April 2020. Similar results were found for the USA but less marked over China. This paper did not attempt to quantify the impact of these changes on radiative forcing estimates.

Zhu *et al.* (2022)[120] also used global CALIPSO data to infer how ice crystal number concentration in cirrus clouds changed during the COVID outbreak. They found a significant increase in northern hemisphere mid-latitudes, which they interpreted as an aerosol–cloud effect driven by reductions in contrail-processed aviation soot (see Section 4.3). They then used these observations in a modelling study of the resulting radiative impact of these changes. The global-mean radiative forcing, attributed to the 73% reduction in flight mileage was calculated to be 21 ± 27 mW m^{-2}. Although this is presented as a positive radiative forcing, we interpret it as a reduction of the negative radiative forcing of -140 ± 70 mW m^{-2} previously reported by Zhu and Penner (2020)[103] for aviation soot emissions; we note that a 73% reduction in soot emissions resulted in only a 15% reduction in the negative forcing, indicating a strong non-linearity in the role of aviation soot. Zhu *et al.* (2022)[120] acknowledge numerous uncertainties, including their attribution of the observed changes to aviation-emitted soot.

Taken together, these works demonstrate the difficulties of separating out the effect the COVID-related change in air traffic on cirrus clouds from the expected year-to-year variation in these clouds; those studies that exploit longer periods of data should perhaps be given greater weight. These difficulties may more relate to the limitations in current observing systems, and the fact that contrail cirrus changes are inferred from cirrus changes. Those studies that focus on more limited regions (either Europe or the USA) give evidence of a reduction in cirrus-cloud cover in Spring 2020, and an indication that those clouds may be thinner. The two observational studies that attempt to link their results to (pre-COVID) contrail cirrus RF or ERF indicate much smaller values than in Fig. 1; this is mostly due to reductions in the estimated contrail cirrus cover, but the uncertainties in these estimates overlap. Given that the RF is not an observed variable, and cirrus properties beyond the fractional coverage influence RF, any conclusions drawn from these studies must remain tentative.

the inherent poor quantification of processes. Moreover, the net forcing is the result of a small residual of long wave and short wave forcings, which have inherent uncertainties.

More recently, Digby *et al.* (2021)[81] show through an analysis of satellite observations that cirrus cloud did not exhibit a detectable global response to the dramatic aviation reductions of spring 2020. This was interpreted as evidence that model-based estimates may overestimate aviation induced cloudiness (see also Box 1). Although the phenomenon of contrails and contrail cirrus is clearly observable, the size of the radiative effect is still under discussion. In Box 1, the rather unique situation of the COVID pandemic and the dramatic downturn in air traffic is examined in terms of what changes in contrails and cloudiness could be observed.

5.3.3 Net-NO$_x$ uncertainties. For the net NO$_x$ forcing, the main short-term ozone and long-term methane responses had a 'medium' confidence level assigned, based on observations of the trends in overall trace gases and laboratory measurements of chemical kinetics, driving the atmospheric chemistry simulated in the models. The overall 'low' confidence level was driven by the necessity of combining the 4 individual forcing terms and the reliance on modelling for the cooling terms of methane reduction, water vapour and long-term ozone response. Thus, although 'uncertainties' have been represented as well as possible for aviation-related forcings, they are not equal either in terms of representation or content, and therefore need to be understood, when mitigation measures are considered.

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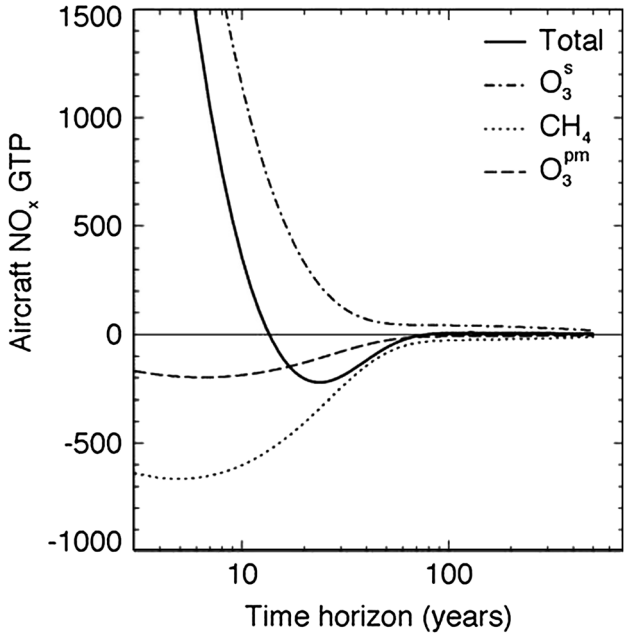

Fig. 12 Temporal variation of an aircraft NO_x GTP ('total'), and sub-components of the short-term ozone temperature response (O_3^s), the long-term negative methane temperature response (CH_4) and the long-term ozone temperature response, or ozone primary mode (O_3^{pm}). The GTP is the ratio of the temperature change due to pulse emission of NO_x (expressed in mass of N) to the temperature change due to an equal mass emission of CO_2 at a given time horizon. From Fuglestvedt *et al.* (2010).[77]

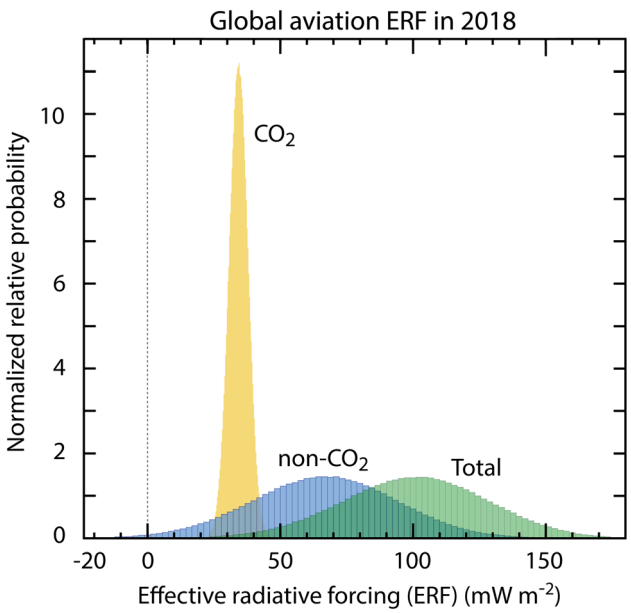

Fig. 13 Probability distribution functions (PDFs) for aviation ERFs in 2018 based on the results in Fig. 1. PDFs are shown separately for CO_2, the sum of non-CO_2 terms, and the net aviation ERF. Since the area of each distribution is normalized to the same value, relative probabilities can be compared. Uncertainties are expressed by a distribution about the best-estimate value that is normal for CO_2 and contrail cirrus, and lognormal for all other components. From Lee *et al.* (2021).[6]

An additional source of uncertainty in the basic effect of aircraft NO_x emissions on ozone is the issue of scale. In modelling NO_x emissions, it is widely assumed that the emissions are instantaneously available, or diluted, at the grid scale of the model and are not modified by plume-scale interactions. Plume-scale chemistry of aircraft emissions at altitude has been studied for over two decades (*e.g.* ref. 82 and 83) and reviewed recently by Tait *et al.* (2022)[84] but is not widely incorporated into modelling because of the co-dependency of the plume and the background. In essence, the oxidation of S and N species is accelerated by the direct formation of OH in the combustor and turbine section of the engine, and some small emission of OH has been modelled to remain.[82,83] This will oxidise a small amount of the NO_x emissions directly to HONO and HNO_3 (and S species to H_2SO_4). These higher oxidized states of N effectively remove the NO_x from the subsequent larger scale cycling of NO_x and HO_x that is involved in the formation of ozone. Two basic problems remain: verification of OH levels at the engine exit (it is very difficult to measure,[85] particularly in-flight) and the incorporation of plume-scale chemical interactions into global chemical transport models. A number of efforts have been made at this over the years, to calculate what are called 'effective NO_x emissions'.[82,86–90] However, it has proven difficult to successfully incorporate interactive multiscale modelling, since what happens in the plume depends on the background and *vice versa*.

A further source of uncertainty in the NO_x system is the formation of nitrate particles and their role in radiative forcing, which is generally thought to represent a negative term.[91]

The above influences of 'effective NO_x emissions' and the formation of nitrate particles would both tend to reduce the net NO_x ERF. Both issues require further work to resolve them.

5.3.4 Aerosol–cloud interactions uncertainties. The aerosol–cloud interactions of both soot and sulphur species were not assigned any best estimate in the assessment of Lee *et al.* (2021)[6] (see Fig. 1). It is likely that the small SO_2 emission from global aviation (0.2 Tg S per year, assuming an average fuel sulphur content of 600 ppm), will result in a negative aerosol–cloud forcing,[6] although of uncertain magnitude. In terms of mitigation, through the potential use of 100% zero-sulphur content fuels such as bio-SAF or renewable synthetic fuels, this negative forcing would simply be removed.

In terms of the aerosol–cloud interactions of soot, soot particles are not efficient ice nucleating particles (INPs) for cold mixed-phase clouds >235 K (ref. 92) but there is evidence that at very cold temperatures soot that has been processed by contrails can be an INP through the pore condensation and freezing mechanism.[93,94]

Aircraft emitted soot, not processed by contrails do not act as cloud INP. Once soot particles have been processed through contrail ice crystal formation and subsequently sublimated, they become more ice-active through the pore condensation and freezing mechanism.[95] While aging due to ozone oxidation potentially renders aviation soot particles better INPs, the coating of soot particles by sulphuric acid during this aging[96] deactivates their ice-forming ability. Based on laboratory measurements, Gao *et al.* (2022)[97] have argued that in the case of aviation soot particles, this aging is in competition with the

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pore-condensation-freezing process, so that it seems likely that aged sulphate-coated particles do not make good INPs for cirrus clouds.[98,99]

The size and sign of this soot-induced forcing has been subject to modelling investigation for some time. Some authors finding a large negative forcing;[100–103] the most recent of these studies found a forcing of −140 mW m^{-2}. Others have found a small forcing.[104–106] Since soot heterogeneous nucleation can occur at lower ice supersaturation thresholds than soluble aerosol homogeneous formation,[107] which is normally dominant in an unpolluted atmosphere, water vapour may condense preferentially on the soot particles; this results in a smaller number of larger ice particles relative to the unpolluted case. These larger particles are more readily removed from the cloud by gravitational settling, so that the cloud contains less condensed water in the ice phase. The clouds are then less reflective (a positive shortwave RF), but also less able to absorb longwave radiation (a negative longwave RF). In Zhu and Penner's (2020)[103] calculations, the shortwave RF (350 mW m^{-2}) is more than offset by the longwave RF (−490 mW m^{-2}) yielding their net RF of −140 mW m^{-2}. The parameterizations used in the models to represent the effect have been found to be sensitive to a number of factors; the vertical velocity of updrafts, the ice activities and number concentrations of different INP types (*i.e.*, mineral dust, and secondary organic aerosols [SOA] – largely from non-aviation sources *e.g.*, Zhu and Penner, 2020).[103] The sensitivities to assumed supersaturation thresholds and the relative amounts of aviation soot available as INP were explored by Righi *et al.* (2021);[108] the resultant forcings ranged from approximately −35 mW m^{-2} to +13 mW m^{-2} for 2014 air traffic. Using an alternative approach, Kärcher *et al.* (2021)[98] used a cirrus cloud column model coupled with laboratory measurements and found that the activated fraction of INPs was <1%, such that soot perturbed visible optical depths were not significantly different from homogeneously formed cirrus, depending on assumptions regarding the poorly constrained number-size distribution of processed aviation soot particles. The authors suggested that this provided a constraint for the overall forcing from this term and that current global models may overestimate the magnitude of this forcing. In a follow-up study, the soot impact on cirrus was studied more realistically including competing effects of mineral dust particles (known to be efficient INPs), and variations in updrafts speeds and INP abundance.[109]

The key points here are that the aerosol–cloud interaction of aviation soot in global climate modelling studies remains unresolved, and that the underlying emission, soot, is common to both contrail cirrus (an accepted significant but uncertain positive net forcing) and aerosol–cloud interactions with cirrus. Until the latter forcing is better understood, any efforts to reduce soot emissions (with the prime purpose of reducing contrail cirrus forcing, either through operational means or changes in fuel), will, on current understanding, have a net uncertain climate outcome. Moreover, modelling has shown that reducing soot emissions significantly without inhibiting volatile plume particle formation will not prevent contrail formation.[110] The degree to which SAF may form (non-sulphur) volatile particles is unknown.

5.4 Changes in understanding between recent assessments of non-CO_2 forcings

To get a sense of the evolution of understanding in the past decade, the assessments[121] of Lee *et al.* (2009: 'L09')[123] and Lee *et al.* (2021: 'L21')[6] are briefly compared. The earlier assessment was a rather simple scaling exercise of earlier data from Sausen *et al.* (2005)[124] and IPCC (1999)[20] whereas the later L21 assessment was based on a much more extensive literature base normalized from the underlying emissions originally used, with some additional original calculations. Fig. 14 shows a comparison of the two assessments for the same base year of 2005 (the base year of L09).[125] A major difference between the two assessments was that L21 used ERF for the aviation forcing terms for which estimates were available. Fig. 14 shows both the RF *vs.* RF, and RF *vs.* ERF comparisons. The major changes resulted from improved scientific understanding. Some are due to changes in understanding of individual aviation RFs, but a major change is due the wider adoption of ERF as the forcing metric of choice.

Between the L09 and L21 assessments, there have been more modelling studies of RF which have been able to exploit improved understanding of contrail properties from *in situ* observations of contrail cirrus; this led to an improvement in the assessed level of scientific understanding (LOSU – an analogue to the Confidence Levels used in L21) from "very low" to "low". The L21 2005 best estimate of RF for contrail cirrus increased by about a factor of 2, but when the improved (but still incomplete) understanding of rapid adjustments is considered (which means that the ERF is about 0.42 × RF), the 2005 L21 ERF is very similar to the L09 2005 RF estimate. This still exceeds the L21 CO_2 ERF but the stated uncertainty in the 2005 contrail cirrus ERF is 6 times larger than the L21 CO_2 uncertainty.

The degree of compensation between the positive and negative forcing components due to NO_x has a large influence on the net-NO_x forcing. As discussed in L21, in comparing different studies, it is important to account for the fact that studies with a large ozone forcing tend to have a large (negative) methane forcing. Although the 2005 net NO_x RF was approximately halved by the L21 assessment over L09, this has been compensated by incorporating rapid adjustments, which means that the L21 ERF is similar to that of L09 for 2005. However, this comparison is complicated by the fact that the additional negative forcings associated with CH_4 destruction (reductions in stratospheric water vapour, and the long-term reduction of background O_3) were not accounted for in L09. The net-NO_x Level of Scientific Understand (LOSU) has decreased from "medium-low" to "low", partly because of the change in the degree of compensation and partly because of uncertainties associated with the rapid adjustments, which are based upon only one modelling study.[79]

The LOSU in water vapour is improved from "low" in L09 to "medium" in L21 because more detailed studies are now available.[126] In L09, the upper limit of the stated RF uncertainty was close to the best estimate of CO_2 forcing. This reduction in

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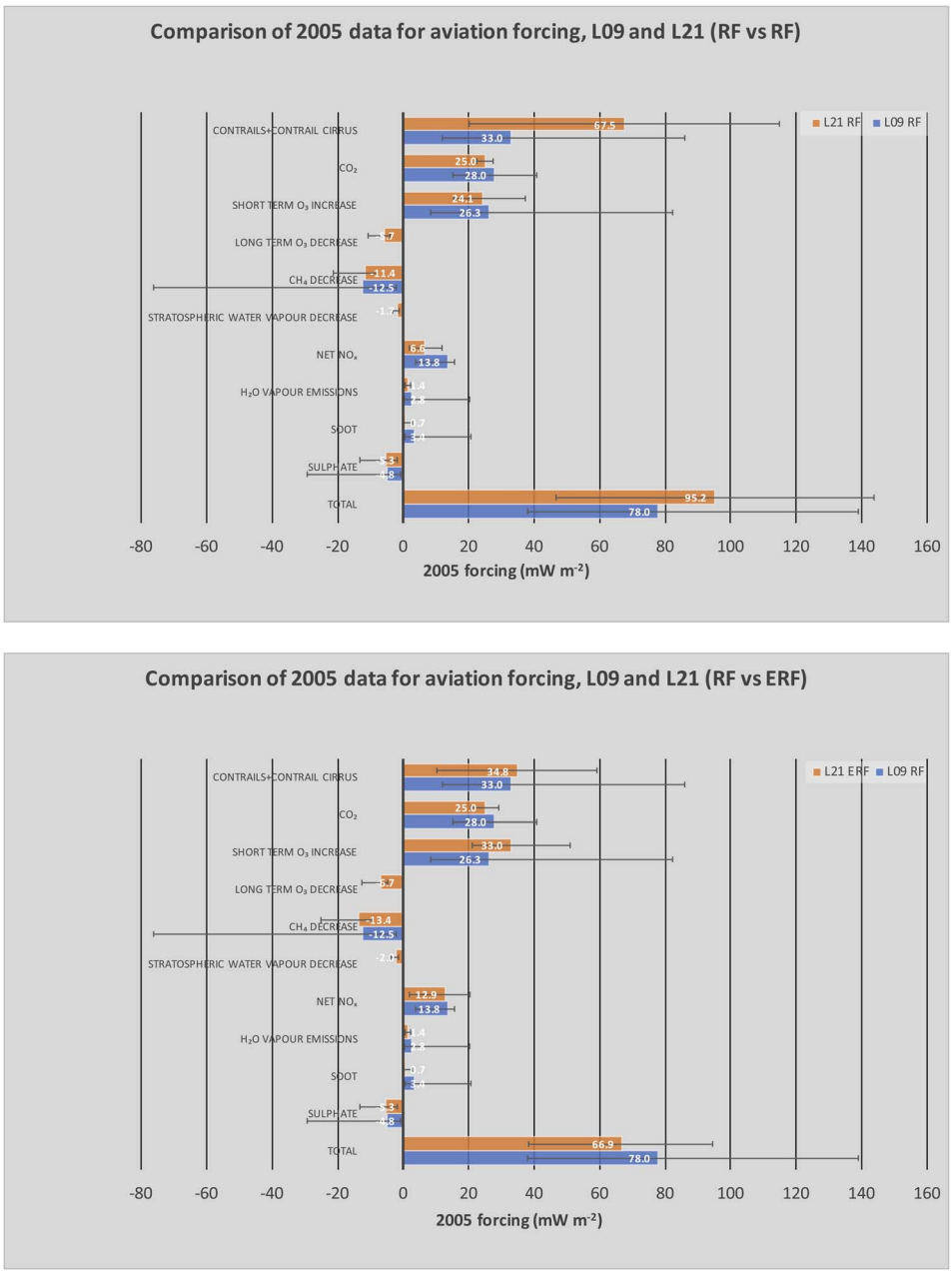

Fig. 14 A comparison of aviation RF assessments from Lee *et al.* (2009)[119] and Lee *et al.* (2021)[6] (L09 (blue), L21 (orange)) in terms of RF and ERF for the year 2005 (the base year of L09). Features to be noted are that the earlier L09 assessment of the net NO_x term did not include some of the secondary negative terms associated with CH_4 destruction, and that the confidence intervals used in L09 were 10%, 90% whereas in L21 they were 5%, 95%. Also, some of the uncertainty distributions changed from (assumed) log normal in L09 to discrete probability distribution functions in L21, based on a much more extensive database of calculations from the literature.

uncertainty renders water vapour to be one of the smallest forcings of those considered by L21.

The aerosol-radiation interaction is now assessed to be more strongly negative because the cooling influence of sulphate now dominates over the warming influence of soot. The best estimate forcings for both soot and sulphur remain small, but the absolute size of the uncertainties in sulphur forcing remain large. L09 did not include aerosol–cloud interactions. The increased number of studies justified its inclusion in L21, but the disagreement amongst them meant that no best estimate was provided.

Although the net aviation 2005 RF increased by 22% between L09 and L21, the inclusion of rapid adjustments reduces the difference when ERF is considered; the L21 2005 ERF is about 14% smaller than the L09 2005 RF.

5.5 How things may change in the future

The uncertainties calculated by Lee *et al.* (2021)[6] strictly only apply to the present day forcings that they present. The uncertainties in future forcings are subject to additional uncertainties, principally from future volumes of air traffic,

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emissions, and their geographical distributions, but also from different chemical background atmospheres that either alter the forcing response (for example, changes in temperature and humidity affecting CO_2 forcing), or the background atmospheric chemical composition (in the case of NO_x) such that the forcing per unit emission can change over time, or even switch sign. In addition, changes in the future climate itself (in terms of *e.g.*, temperature, humidity) may also have impacts on the contrail cirrus forcing response.[127,128]

These future uncertainties become of particular importance when considering (climate) mitigation responses for aviation and any implicit technology responses, with their associated investment costs.

A frequent misunderstanding is that the present-day large proportion of non-CO_2 forcing to total forcing is a fixed fraction. As shown in Fig. 5, for an idealized scenario of constant emissions, non-CO_2 forcing remains approximately constant, whereas the CO_2 forcing increases over time. The present-day large fraction of non-CO_2 forcing is the result of the strong rate of change in recent years of fuel consumption of ~4% per year. This results in a strong and immediate increase in the rate of non-CO_2 forcing, whereas the CO_2 forcing responds more slowly, as a result of the cumulative emissions. This is further illustrated in the future scenarios of Klöwer *et al.* (2021)[7] (Fig. 6) who showed the varying proportions of non-CO_2 to total temperature response for increasing, constant, and declining fuel usage and CO_2 emissions. The present-day large fraction of non-CO_2 forcing results in the implication that non-CO_2 emissions are important to mitigate, whereas the behaviour of the forcings in relation to fuel usage and type, along with the non-linearity of forcing per unit emission, suggests that more careful thought is required, which is explained in the following sections. In this section, we focus on the implications of changing composition of aircraft emissions; however, it is important to stress that any change in aviation growth rates would itself impact the relative contribution of non-CO_2 to CO_2 forcing, even with no change in composition, because of the contrasting lifetimes of CO_2 and non-CO_2 forcing agents in the atmosphere. This issue is discussed further in Section 8.

5.5.1 Aircraft NO_x emissions in the future and their relationship to other future background emissions. The contribution of aircraft NO_x emissions to the formation of tropospheric ozone is only one of many anthropogenic sources; other emissions of carbon monoxide, methane, and non-methane hydrocarbons play a role in ozone formation. Therefore, the effect of an emission of aircraft NO_x depends on other sources. This was illustrated by Skowron *et al.* (2021)[129] who showed that the net-NO_x aircraft forcing term varied for the same aircraft emissions, depending on the background emissions. This was because the 'cleaner' atmosphere resulted in more ozone per unit emission of aviation NO_x, but counteracting this, the increased (aviation) NO_x emissions implicit in the future scenarios also resulted in a stronger CH_4 destruction rate, such that the resultant negative forcing outweighed the positive forcing from short-term ozone. Both these effects of variability of net NO_x RF with a fixed aircraft NO_x emission and the potential switching of net NO_x RF from positive to

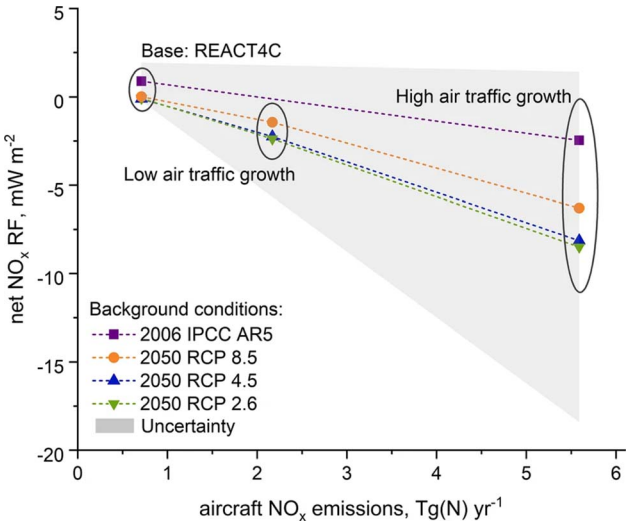

Fig. 15 Aviation net NO_x radiative forcing (RF) (the sum of the short-term positive O_3 RF perturbation and the negative RF terms caused by a reduction in CH_4 lifetime), by aviation NO_x emission rate according to a range of background emission scenarios. Aviation net NO_x RF systematically decreases with increasing NO_x emissions from aviation, showing a variation according to the background surface emissions, with high mitigation (RCP 2.6) having a smaller (or 'more negative' at larger emission rates) aviation net NO_x RF than, lower mitigation scenarios (RCP 4.5, RCP 8.5) for the same aviation NO_x emission. Overall uncertainties are indicated by the grey shading, which is one standard deviation (68% confidence interval). From Skowron *et al.* (2021).[129]

negative, are shown in Fig. 15, taken from Skowron *et al.* (2021).[129] Other more recent results[91] broadly confirm these conclusions.

Thus, it is important to recognize that in the future, aviation NO_x emissions may have a net negative ERF, in contrast to the present day. The net global and hemispherical temperature effect of the combined positive and negative forcings needs more work to understand. The local temperature response is not necessarily correlated or collocated with the local forcing,[130,131] since the pattern of temperature response at the earth's surface is predominantly driven by internal climate feedbacks.[132,133] Moreover, Shindell *et al.* (2015)[132] found an enhanced temperature response from inhomogeneous forcings relative to equivalent global mean forcing by well-mixed greenhouse gases at the global mean level. More specifically in the context of NO_x, Shine *et al.* (2005),[134] for surface NO_x emissions, and Lund *et al.* (2012),[135] for aviation emissions, examined the geographical distributions of surface temperature responses in climate models. Lund *et al.* (2012)[135] found that the ozone-methane compensation in global-mean RF does not properly reflect the temperature impact, which showed ozone-driven warming in the northern hemisphere but much reduced warming (and, in one climate model, cooling) in the southern hemisphere, where the effect of methane destruction could dominate over ozone. Given that in Skowron *et al.*'s (2021)[129] future aviation case, the net future forcing is negative driven by a reduction in a well-mixed GHG, CH_4, and that the positive forcing from O_3 occurs mostly in the northern mid-latitudes (see, *e.g.*, Fig. 9 for representative present-day

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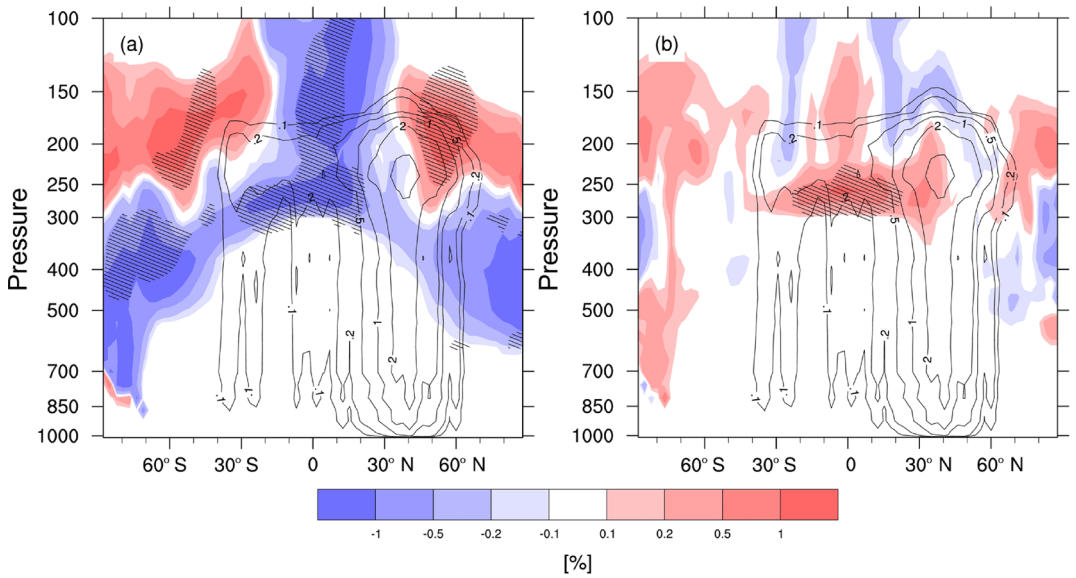

Fig. 16 Changes in the formation probability of persistent contrail formation between 2006–2050 attributable to climate change (a) and improved propulsion efficiency (b) in one Earth System Model (Bock and Burkhardt, 2019).[128] The black contours indicate projected 2050 annual flight distances (in 10^8 km), as a function of latitude and pressures, the hatched areas indicate differences of statistical significance.

conditions), understanding the temperature response on regional and hemispherical scales needs further work. The important point is that current understanding indicates that, for NO_x, the net global-mean RF does not adequately reflect its importance for regional surface temperature response, with implications for CO_2-e metric design.

5.5.2 Contrails in a changing climate. Future projections of contrail radiative forcing are dependent on changes in the volume of air traffic, its geographical and vertical distribution, propulsion efficiency and soot emissions. They are, in addition, dependent on changes in climate, which can impact on temperature and humidity in the upper troposphere, which in turn dictates not only whether the Schmidt-Appleman criterion is met, but also on the probability of ISSR occurrence.

In one modelling study[128] a distinct change in the probability of persistent contrail formation was found (Fig. 16), with decreased probability at cruise altitudes in the tropics and increases in mid to high latitudes due to climate change (Bock and Burkhardt, 2019, their Fig. 5).[128] In their simulations, the global-mean contrail forcing due to climate change was only minimally different in 2050 compared to 2006, due to compensatory changes in different locations. The general pattern of decreased formation probability at cruise altitudes (around 250 hPa) in the tropics and increased probability in mid- and high latitudes is robust across a number of different Earth System Models but with differences in the strength of the effect and details of the pattern.[127] Hence, whether the degree of compensation found by Bock and Burkhardt (2019)[128] would be found in other ESMs is an open question, as is the dependence on the trajectory of future climate change. All such studies are dependent on assumptions regarding future changes in air traffic distribution, engine characteristics and fuels.

6 Aviation emissions and effects on the atmosphere–air quality

6.1 Aviation-air quality context

Air quality in the vicinity of airports is subject to European and international regulations that set limits on NO_2 concentration and particles (classed as 'particulate matter' with a corresponding mean aerodynamic diameter of 10 or 2.5 micrometers–PM_{10}, $PM_{2.5}$). The ICAO LTO cycle emissions regulations are focused upon air quality requirements (see Section 3.1), but emissions during other phases of the flight greatly exceed the quantities during LTO and are now attracting more attention. Ambient air quality regulations for NO_2 and PM_{10} and $PM_{2.5}$ and ozone are in place in the UK and Europe under Council Directive 85/203/EEC. The World Health Organization (WHO) also recommend limit values for NO_2, $PM_{2.5}$ and PM and ozone.[136]

The regulations for ambient air quality—often referred to as 'air quality standards' vary across regions of the world and are subject to change. We outline some of the regulations pertinent to the UK, the US, and the WHO recommendations, which have significant influence on national and international policy.

In general, air quality standards refer to ambient measurements of NO_2, and particulate matter, with an upper limit size in terms of diameter (10 or 2.5 microns). It is important to differentiate air quality standards from aviation emission regulations, which refer to NO_x, summing the NO + NO_2, as it is assumed that all the NO will be rapidly converted to NO_2, and 'nvPM' — whereas ambient PM is 'total', *i.e.*, the volatile plus non-volatile fractions. In terms of aircraft emissions, the volatile *vs.* non-volatile partitioning is operationally defined by the measurement system, for which the sample is heated to 350 °C. In practice, this means that the non-volatile fraction is largely black and organic carbon, and the volatile fraction consists of

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oxidized S compounds (sulphuric acid and its salts), potentially nitrates, and semi volatile organic compounds that form particles. Typically, both fractions of aircraft PM emissions are in the sub 100 nm range and are often referred to as 'ultrafine particles (UFPs)'; the non-volatiles occur in this range (Aitkin mode) and the volatile particles tend to be smaller, at <10 nm (nucleation mode). Since the volatile fraction of aircraft PM emissions are strongly fuel-composition dependent, they are, at present, not characterized in the emissions regulations. Nonetheless, the volatile fraction will contribute to UFPs in the ambient atmosphere in the vicinity of airports.

Ambient concentrations of NO_2 have been considered of potential harm to human health in terms of respiratory impacts and asthma,[137] although the evidence for the effects on premature mortality is compromised by the epidemiology of co-emitted particulate pollutants.[138] Indeed, the UK Committee on the Medical Effects of Air Pollutants in their most recent report on NO_2 failed to reach agreement that there was sufficient evidence to infer a causal association between long-term average ambient concentrations of NO_2 and risk of death, concluding that some of the aspects of the epidemiological evidence reviewed, weakened the case for a causal link between long-term exposure to NO_2 and all-cause mortality.[139] Nevertheless, the WHO recently based its updated recommendation[140] of a maximum annual average level of 10 $\mu g\ m^{-3}$ to protect human health on three major systematic reviews and meta-analyses.[141–143]

Ozone is a secondary pollutant formed by a series of chemical reactions in the presence of sunlight. Photochemical reactions of NO_x and volatile organic compounds (VOCs) originating largely from combustion processes, govern the concentration of ground-level O_3 in the atmosphere. Highest levels of ozone generally occur during the summer months and downwind of the combustion emission sources.

Particles are thought to adversely affect human health and the evidence for this is considered robust. It is thought that ultra-fine particles (UFPs) of 100 nm aerodynamic diameter, or less are particularly harmful.[144] These can penetrate deep into the lungs and may enter the bloodstream. There are some workplace regulations concerning UFPs but, as noted above, the current ambient regulation limits are expressed as $PM_{2.5}$ and PM_{10}. The $PM_{2.5}$ and PM_{10} mass concentration measurements are for total PM and reflect the quantity of UFP in ambient air rather poorly as these smaller particles make up a small part of the PM mass. For aircraft engine emissions of nvPM, which tend towards particles of around 50 nm or less (the particle mass of a 50 nm particle is approximately one millionth that of a 5 μm particle), they are likely to be a very small contributor to these ambient mass measurements.

Aircraft engine emissions of particles are operationally defined by the measurement system as 'volatile' and 'non-volatile' fractions. At present (assuming the usage of fossil fuel with S present at an average level of 600 ppm), the vPM primarily comprises sulphate and organic particles, and the nvPM, soot particles. Soot particles are largely unburned carbon which is formed in the combustor of aircraft engines combined with some organic carbon, and therefore considered 'primary' emissions (*i.e.*, directly emitted). Sulphur present in the fuel is oxidized on combustion to form sulphur dioxide gas, which is subsequently oxidized in the plume and atmosphere to form sulphate particles. A small fraction, around 2%, of the sulphur is oxidized to sulphate within the engine and emitted as 'primary' gas-phase sulphuric acid. Other volatile particles can exist from the formation of nitrate particles from emissions of NO_x and organic compounds to form particulate organic matter (POM). However, while the evidence for the potential formation of nitrate and POM is robust from knowledge of atmospheric processes, the quantification of these from aircraft emissions is poor.

The size and number of primary soot particles are a function of the engine technology and the thrust setting whereas the emission of SO_2 scales linearly with fuel sulphur content, and therefore with fuel usage. The size and number concentration of secondary sulphate particles is a function of atmospheric processes. While the volatile and non-volatile fractions of particles are often considered separately, they are in fact, mixed. For example, soot can form the 'core' particle and be coated with sulphate. Moreover, the toxic feature of nvPM is generally the coating of the carbon with organic combustion produces, such as polycylic aromatic hydrocarbons (PAH).

Both volatile and non-volatile particles are ultimately removed from the atmosphere by the processes of chemical transformation, or wet and dry deposition to the earth's surface. The impact of aircraft emissions on air quality is usually considered from the perspective of the LTO cycle, which is quantified to 3000 feet above ground level.

It is worth noting that the current ambient regulations, as mass-based concentration limits, are for total PM (vPM and nvPM) and the epidemiological evidence upon which the regulations are based does not distinguish between vPM and nvPM or their chemical composition. Furthermore, the use of the $PM_{2.5}$ mass concentration metric for ambient regulation is very biased towards the larger particle sizes. As noted above, toxicology, however, points to very small particles (<100 nm) being more harmful and that the toxic organic chemicals caused by incomplete combustion very likely coating or being carried by small soot particles are the real cause of harm. Therefore, soot and soot-coated particles from combustion sources are likely to be more toxic than the sulphate aerosol component. Based on emerging and future toxicology and air pollution health impact research, future ambient regulations may change to include, for example, particle number concentrations and/or chemical composition parameters. However, this is not imminent and $PM_{2.5}$ and PM_{10} remain the current regulated metrics for ambient particulate air quality purposes.

One of the potential sources of particle emissions from aircraft is the engine lubricating oil.[145–148] The oil is emitted through the breather vent or unintentionally as the result of worn seals. It has been suggested that UFPs may be emitted from these systems as condensable organic compounds.[148] These particles remain poorly characterized and understood in terms of verifying their source and composition, and effects on human health.

6.2 Pollutants in the vicinity of airports

LTO emissions from aircraft have an impact on the ground-level concentrations of pollutants including NO_x and particles. Ultra-

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Box 2: The health impacts of aviation emissions

Ambient air quality standards for NO_2 have been set in many countries but the question over its health impacts remains somewhat open. NO_x is emitted by road vehicles, and it is well documented that there are serious health consequences of living in heavily polluted areas, particularly for children, and that NO_x tends to be present when pollution is high. In the UK, the Committee on the Medical Effects of Air Pollutants (COMEAP) considered this and reported in 2018 (ref. 139) but could not arrive at an agreed consensus. Attention was drawn to reviews of the USEPA (2016)[137] and Health Canada (2016),[165] which concluded that the evidence for long-term NO_2 concentrations and total mortality was suggestive, but not sufficient, to infer a causal relationship. It is widely recognized that NO_2 acts, in part, as a marker of traffic-related pollutants including ultrafine particles. COMEAP's own assessment of the evidence[138] concluded that "it would be sensible to regard NO_2 as causing some of the health impact found to be associated with it in epidemiological studies". Atkinson *et al.* (2018)[166] undertook a meta-analysis of 48 studies and concluded: "We therefore consider that as the evidence stands at present, the causal basis for estimating the burden of NO_2 on mortality and loss of life expectancy remains weak". However, despite the lack of clear causality, the World Health Organization has recently produced guidelines[136] calling for a reduction in levels of pollutants, including a lowering of the allowable levels of NO_2. During the 2010 eruptions of Eyjafjallajökull, when aviation was stopped over much of Europe, the opportunity was taken to compare the levels of NO_x around the perimeters of a number of European airports.[156] This report concluded that "the contribution of air traffic to local air quality in the vicinity of airports is very small".[167]

The issues associated with particulate matter are rather different. The evidence upon which the current understanding is based in part is the US 'Six-Cities' study[163] in which levels of: total particles, $PM_{2.5}$ (fine particles of 2.5 μm and less), sulphur dioxide, sulphate particles, aerosol acidity, and ozone were studied. The dependence between health outcomes and sulphate and $PM_{2.5}$ was almost linear. This has come to be associated with causality. In fact, the emissions of sulphate and the relatively large (2.5 μm) particles probably occurred alongside the potentially far more damaging ultrafine particles resulting from combustion, which were not characterized, but modern measurement techniques can now show are present. Volatile (vPM) in the aircraft plume consists largely of sulphate with subsequent condensation of sulphuric acid with water, the sulphur coming from the fuel. The vPM particles are small, less than 20 nm and aircraft engine nvPM is also small, of the order of 30 nm or less.

Whilst the collection of $PM_{2.5}$ will capture the particles at, for example, 25 nm, the mass of a 2.5 μm particle will be about a million times larger. The monitoring of $PM_{2.5}$ by mass concentration will therefore be dominated by the larger, heavier particles and will not reflect the actual contribution of ultrafine particles from jet engine (or modern diesel) exhaust which are better represented by particle number concentration measurements. Better instrumentation including particle size and number measurement has now allowed much smaller particles to be detected and there is evidence that particles of order 30 nm are carried into the alveoli of the lung and able to pass through cell walls. If the particles were pure carbon, it is unclear what harm this would do, but there is clear evidence that the solid particles are carriers of other products of combustion, such as polycyclic aromatic hydrocarbons (PAHs), some of which are known to be toxic and/or carcinogenic.

It is clear that a toxicological basis is needed to give a firm basis for any causality attribution from correlation. There is extensive literature on the effects of particulates. A small sample describing some of the effects is: Oberdörster *et al.* (2005), Cassee *et al.* (2013), Robinson *et al.* (2017), Holme *et al.* (2019), and Kelly and Fussell (2020).[168–172] The carbon forming the core of the particulate does not seem to have an unequivocally harmful effect at the concentrations of concern, but the material coating the particle is the principal cause of harm. Robinson *et al.* (2017)[170] showed that diesel exhaust particles produce a strong reaction in the vagus nerve of laboratory animals (*in vivo* and *in vitro*) and in humans; if, however, the particles are first washed, the cells do not respond. The use of correlations between health impacts and $PM_{2.5}$ (or larger, PM_{10}) has given rise to the assumption that all particulate matter less than 2.5 μm in size is equally damaging to health, regardless of size or chemical composition. The Six-Cities and other studies, *e.g.*, Ostro (2004)[173] have shown that particulate concentrations are correlated with ill health. This has been used with work to show that NO_x released at cruise can be brought to the surface contributing to ground level concentration of $PM_{2.5}$ by reacting with NH_3 to form NH_4NO_3 (a reversible reaction). On the assumption that all particulates are harmful, large annual rates of death attributable to cruise emissions have been estimated[159] conversely, because the concentration response functions are mass-based, the nvPM, which better represent fine particles of soot appear to have a very small effect. Toxicology would indicate that the reverse situation could be true, and that the S and N particles are less harmful. A better understanding of the likely nature of harm from exhaust particulates (ultrafine particles on order 20 nm coated in toxic substances) is vitally important when gathering evidence for emission regulations.

In summary, combustion products can be harmful to health, but correlations attributing the harm to specific constituents are confounded by the emissions containing a mixture of particulates and NO_x, and the particulates being of different size and composition. The toxicological evidence points to NO_x and larger non-combustion particles being relatively less harmful to health, but nvPM potentially very harmful because of the ability of ultrafine particles to penetrate into many organs of the body carrying with them toxic organic compounds. Recent work on the toxicological effects of aircraft emissions has pointed to this.[154,174]

fine particles (<100 nm in aerodynamic diameter) are of particular concern, since these maybe harmful to health (see Bendtsen *et al.*, 2021 for a review),[149] and many measurement campaigns have confirmed their presence around airports.[150–153] Specific studies of health effects of aircraft emissions (as opposed to UFPs in general) are rather few but indicate that there may be demonstrable effects on bronchial cells.[154] Emissions of organic compounds (which may contribute to secondary ultrafine particles) remain poorly characterized, particularly in terms of speciation[63] but recent more complex measurements have made important steps to identify such species.[155]

Concentrations of these pollutants may be determined from short-term measurement campaigns, long-term monitoring stations or from air quality dispersion modelling exercises.

Measurement campaigns can provide a snapshot of the aircraft contribution, whilst monitoring data provide longer term trends of the concentrations, allowing comparison with regulatory limits. However, it is often challenging to attribute the source of the contribution for the resulting PM, due to other non-aircraft airport-related sources. It should be noted that long term PM monitoring is most often undertaken for $PM_{2.5}/PM_{10}$, whilst UFP measurements at airports tend to be single short campaigns.[151] Measurement campaigns of UFPs using particle number measurements tend to pick up airport (including aircraft) activity in a way that $PM_{2.5}$ monitoring does not. Several campaigns have observed increased UFP concentrations in the vicinity of airports.[156] However, until there are standardized UFP measurements taken over longer periods, these UFP campaigns provide only a snapshot, and without ambient air quality regulations in place for particle number/UFP concentrations it is not possible to assess the impacts of the measured concentrations on local air quality. The potential health impacts of UFP from combustion sources such as aircraft emissions is an issue

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that will require review in the context of emerging research and potential future regulation.

In terms of modelling, LTO PM and NO_x concentrations are usually calculated with a local scale dispersion model. A local scale dispersion model requires detailed input and includes airport specific information such as location of runway, topography and aircraft movement journals and relevant taxi times. In terms of local scale modelling, the concentration results are highly localized since dispersion characteristics are location-specific, with the atmospheric stability criteria, deposition rate and removal processes highly dependent on local meteorological conditions and topography. The key point is that the aviation contribution to concentrations is inherently unverifiable, since no unique 'marker' or signature to aircraft emissions is available. Local scale modelling requires a knowledge of all sources in the area of interest, so the only comparison that is possible, is that of total concentrations with measured ambient concentrations.

Generally, some impacts from NO_x emissions of large airports, including aircraft but also airside surface traffic, can be identified in the vicinity of the airport by dispersion modelling and measured concentrations. For example, modelled annual mean NO_2 air pollution near Heathrow Airport (based on measurements made during 2016) is comparable to levels found in the urban area of London but lower than those of central London.[157] Ozone is a pollutant that is also of concern in terms of health impacts, and vegetation damage but requires a detailed representation of atmospheric chemistry in a dispersion/regional model.

Dispersion modelling studies generally do not show a noticeable contribution to $PM_{2.5}$ concentrations around airports (which is consistent with the relative size of combustion primary particulate emissions, nvPM) for example at London Heathrow.[157]

6.3 The contribution of cruise emissions to ground-level concentrations

Issues associated with aviation emissions have, by convention, been split up into those from the LTO cycle, with ICAO emission regulations applying to them to a defined height of 3000 feet, and those above *i.e.*, non-LTO including those mainly at cruise but also including climb and descent.

LTO emissions from aircraft engines are regulated and considered in terms of their contribution to air quality degradation, and those at cruise conventionally associated with climate impacts (*e.g.*, NO_x, soot/sulphate particles, contrails and contrail cirrus cloudiness enhancement) but not separately regulated. The broad assumption is that reductions in emissions in the LTO cycle, as mandated to meet the CAEP regulations, will also reduce the emissions during climb and cruise phases. Over the last 18 years, there has been a growing body of literature, starting with Tarrasón *et al.* (2004)[158] looking at the possibility that emissions at cruise (or non-LTO) may also contribute to degradation in air quality near the earth's surface, specifically ozone concentrations and $PM_{2.5}$ with a number of studies showing that the modelled contribution to ground level $PM_{2.5}$ from non-LTO aviation emissions exceeds that from LTO emissions A review of this literature is included as an Annex (see Appendix 1). Two main effects have been examined in a range of studies; the effects of non-LTO emissions of NO_x on ground level ozone concentration, a photochemical pollutant that has effects on human health and plants, and the contribution to ground level $PM_{2.5}$ concentrations. The modelled contribution to ground level $PM_{2.5}$ is mostly from secondary aerosol particulate matter with only a small, or negligible contribution from aviation soot or black carbon emissions (nvPM). For example, Barrett *et al.* (2010)[159] estimated that, on a global basis, 99% of population-weighted ground level, aircraft-attributable $PM_{2.5}$ is secondary sulfate-ammonium-nitrate aerosol and only 1% is primary particulate matter. The additional aerosol particles ($PM_{2.5}$) at ground level from non-LTO aviation emissions are generally modelled through a complex set of atmospheric interactions and chemical reactions at the global scale, with some at the regional and local scale.

Non-LTO emission effects on ground-level air quality are inherently far more uncertain than the LTO fraction, as the emissions and transport through the atmosphere can only be modelled (with global models) and not measured. The modelled impact of non-LTO emissions at ground level is entirely dependent on chemical interactions including with other background pollutants and removal processes such as wet and dry deposition. Transport processes (large-scale and convective) and their parameterization are also important in the redistribution of pollutants. These are all highly complex processes that are often simplified in models ('parameterized'), which are illustrated schematically in Fig. 17.

Vennam *et al.* (2017)[160] used a regional scale model to estimate the impacts of full-flight aircraft emissions on air quality, looking specifically at the sensitivity to horizontal grid resolution. A comparison of a 108×108 km^2 scale with 36×36 km^2 scale resolution gave approximately 70 times and 13 times larger aviation impacts for O_3 and $PM_{2.5}$ with the coarser resolution. These differences are mainly due to the inability of the coarse resolution simulation to capture nonlinearities in chemical processes near airport locations and other urban areas.

The modelled interactions include aircraft-attributable HNO_3 and H_2SO_4, from NO_x and SO_x, respectively, and background NH_3. The presence of NH_3, emitted from surface (largely agricultural) sources can enhance the formation of nitrate particles, in the absence of H_2SO_4. The aviation induced aerosol increase near the ground is highly dependent on background NH_3 concentrations whose current range of uncertainty is large. Several studies also predict an increase in surface level O_3 due to the NO_x emissions from non-LTO emissions, with higher contributions modelled during the winter months when ambient levels are lower.[161]

A range of global/regional scale models show similar qualitative contributions from non-LTO emissions to ground level $PM_{2.5}$ and O_3 but there are large differences in the scale of estimated absolute perturbations of $PM_{2.5}$ and O_3 and perhaps more importantly, there are differences in terms of the significance associated with these modelled perturbations.

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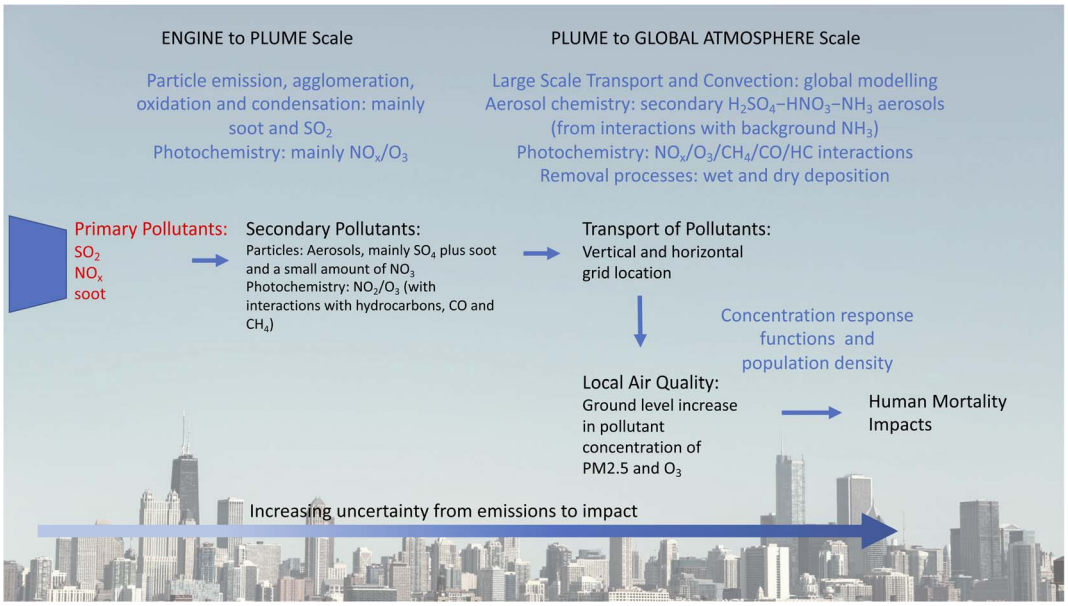

Fig. 17 Schema of processes that lead to potential changes in mortality from aircraft cruise emissions.

Some studies[159] use the modelled aviation-induced concentrations to combine with population density and Concentration Response Functions (CRF) to predict additional mortality rates associated with these aircraft emission perturbations. It is assumed that these particles will carry a burden of morbidity and mortality based on a concentration without regard for their chemical composition and it is assumed that there is no threshold concentration for the effects on health.[162] The application of CRF to estimate potential excess mortality is driven by the underlying population densities and the results are also very sensitive to the CRF used. Other researchers considered that the marginal increases in $PM_{2.5}$ (and ozone) were not sufficiently substantial (<1% of background) to be meaningful considering the uncertainty in state-of-the-art models.[164]

In conclusion, non-LTO emissions in a range of modelling studies are shown to contribute to ground level $PM_{2.5}$ and O_3 concentrations but the scale of the contribution when compared to background levels is small and the significance of the contribution to local air quality and then to human health is uncertain. Effects of aircraft emissions on surface air quality and health during non-LTO and LTO phases will continue to advance and should remain under review.

7 Non-CO_2 effects and their relationships to future fuels

7.1 The potential impacts of future non-fossil kerosene type fuels

In terms of liquid kerosene-type fuels, there is a general desire to move away from fossil fuels to fuels that have a lower (fossil) carbon footprint, which are often drop-in bio-based or synthetic fuels, termed 'Sustainable Aviation Fuels', or SAFs. The issue of defining 'sustainable' and what an acceptable 'lower carbon footprint' is, are issues that are not addressed here and can be contentious,[175] other than the assumption that the lifecycle C is less than that of fossil kerosene. Nonetheless, such fuels would have implications for aviation non-CO_2 emissions that are either independent or co-dependent on combustion technology and developments.

Sulphur compounds are present in aviation fuel at ppm levels,[176] with the regulatory limit being specified by UK Defence Standard 91–91 and in the US by ASTM D1655 at 3000 parts per million by mass (ppm). In practice, levels are found at around 600–800 ppm.[176] It should be noted that fuel survey (composition) results are less available than they were over two decades ago, when fuel S content was routinely monitored and reported.[20]

The presence of suIphur in fossil kerosene fuels results in the emission of sulphur dioxide (SO_2) and a small fraction of directly oxidised S^{VI} as sulphuric acid (H_2SO_4), at approximately 2% ± 0.5%.[177] The SO_2 oxidises in the plume relatively slowly *via* OH to form sulphuric acid, which forms part of the aerosol. These aerosols can have a small direct negative (cooling) RF, reflecting solar radiation back to space. In addition, these sulphate particles and the primary emission of SO_2 can be removed slowly from cruise altitudes (where the bulk of the emission/fuel burn occurs, globally) and affect lower-level warm liquid clouds, enhancing cloud droplet density and decreasing mean droplet size (as do all S emissions from surface sources), which is potentially a significant effect (negative tens of milliwatts per square metre),[104,178,179] relative to other aviation radiative effects. However, this effect is very poorly quantified, and Lee *et al.* (2021)[6] were unable to give a best estimate of this forcing in their assessment. Biofuels and synthetic fuels in their pure form contain very low levels or zero S, so very simply, as these fuels are potentially used with increasing volume/proportion to the global kerosene usage, the associated cooling effects would quickly disappear.

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For soot particles, the situation is more complex than for S. Soot is a result of the combustion of kerosene and is determined by the combustion conditions (the efficiency or 'cleanness' of combustion) and the chemical composition of the fuel. Thus, there are two routes by which soot particle emissions may be reduced. The technological combustion route has been demonstrated with the development of lean-burn engines (see Section 3.2). However, the soot particle number emission index (per kg fuel), is not the sole determinant of the formation of ice crystals. While both technological improvements and reduced fuel aromatic content may reduce soot particle number emissions, modelling indicates that there is a level below which further reductions in soot may not reduce ice crystal numbers further, and even potentially increase them, at temperatures well below contrail formation threshold temperatures because of enhanced activation of ultrafine aqueous particles. This has been shown in theoretical modelling,[110] see Fig. 18.

The significance of smaller soot number concentrations ($<10^{14}$ kg per fuel) at colder temperatures is clear, in that combustion technology improvements to reduce soot particle numbers will not necessarily reduce ice crystal numbers at these colder temperatures, and rather the opposite results, that it increases ice crystal number, since fewer soot particles are available for activation.

Many measurements of SAF emissions at both ground and more recently a few at cruise altitudes have shown that soot number concentration is related to the aromatic content of the fuel. Fossil kerosene contains generally around 20% aromatics by volume, whereas this is much lower or zero for pure SAF fuels. For emissions from pure SAF combustion[180] in ground conditions/test rigs, soot is greatly reduced, but not to zero. Emissions from certified SAF/fossil kerosene blends also show reductions in soot number concentration and recently, ice crystal concentration.[181] There have been a number of measurement campaigns that have quantified reductions in soot emission behind aircraft engines at the ground, using SAF compared with conventional kerosene.[182–186]

The observed reductions in ice crystal number coincident with reduced soot number concentration emissions with SAF[181] confirms earlier theoretical studies that pointed out the theoretical relationship between soot number and ice crystal number,[187–190] and the potential outcome and co-benefit of reducing CO_2 and potentially contrail forcing with increased usage of SAF.[191,192] The relationship between ice crystal number concentration (N_{ice}) and RF is shown in a global model simulation from Burkhardt *et al.* (2018)[191] in Fig. 19. These model results assume a linear relationship between soot number and N_{ice} and it is not explicitly modelled. Moreover, it is critical to note that this modelling applies only to a 'high soot regime' of $>10^{14}$ soot particles kg per fuel, and not 'low soot regimes', since the model of Burkhardt *et al.* (2018)[191] does not consider ultrafine aqueous particles.[110] A later development of the model[192] incorporates a parameterization of N_{ice} from soot,[189] but the authors caution that the role of ultrafine aqueous particles in N_{ice} at low soot regimes is not represented. Other modelling of the effect of low-soot biofuels[193] shows rather conflicting results that imply 67 to 75% reductions in soot result in a net change in contrail RF of −4% to +18%, depending on ice crystal habit (shape) assumed. Bier and Burkhardt (2022) used a updated N_{ice} formation scheme[192] and in addition,

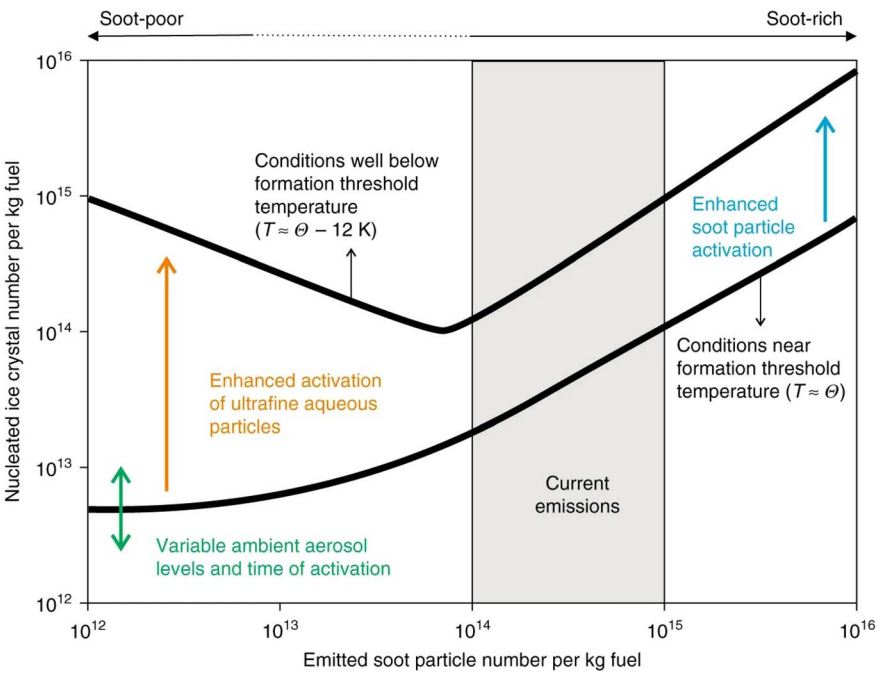

Fig. 18 Modelled ice crystal number emission index (per kilogram of fuel burnt) in the jet regime as a function of the number emission index of emitted soot particles for two temperature conditions, one close to the contrail formation threshold temperature of approximately 225 K, and a temperature 12 K below this value. At lower ambient temperatures, the microphysical mechanism of particle activation shifts in favour of ultrafine aqueous particles involving S and volatile organics. Taken from Kärcher (2018).[110]

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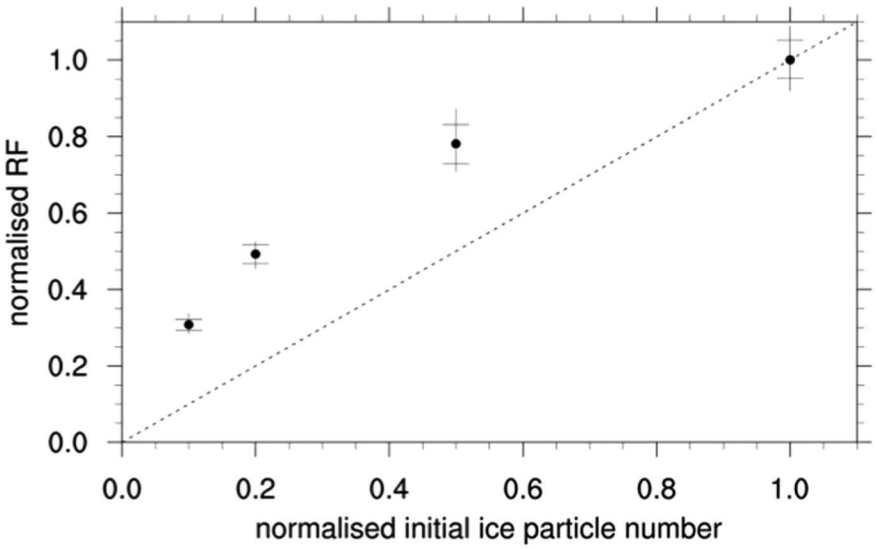

Fig. 19 Normalized contrail cirrus RF *vs.* normalized initial ice particle number in plume, showing that an approximate 80% reduction in ice particles results in a 50% reduction in RF (taken from Fig. 1(f) of Burkhardt *et al.*, 2018).[191]

incorporated a wake-vortex loss parameterization. With this model configuration, Bier and Burkhardt (2022)[61] found a somewhat smaller change of −41% in RF for an 80% reduction in soot than did Burkhardt *et al.* (2018)[191] (*cf.* approximately −50%, noting the linear assumption of N_{ice} with soot numbers, assumed in Burkhardt *et al.*, 2018).[191]

As far as we are aware, there are only 2 global climate models that incorporate a description of the contrail cirrus formation process and its subsequent radiative forcing (*e.g.*, Bier and Burkhardt *et al.*, 2022 and prior publications;[61] Chen and Gettelman, 2013).[194] Thus, the modelled reductions in RF from reduced soot number emissions from usage of SAF described by Burkhardt *et al.* (2018)[192] and Bier and Burkhardt (2022)[61] should be interpreted as indicative only, and subject to change, should more models and calculations become available. Moreover, the results are applicable for the 'soot-rich' regime only, *i.e.*, $>10^{14}$ particles per kg fuel. The processes involved from changed fuel composition (particularly SAF) through to contrail forcing are incompletely modelled and heavily parameterized. Thus, any 'co-benefit' of SAF contrail forcing remains somewhat speculative until all processes are included, including the role of ultrafine aqueous particles at low soot regimes.

In summary, a change in fossil fuel to a SAF-type liquid kerosene blend is likely to result in reduced S emissions. This, in turn, will potentially remove uncertain negative ERFs from sulphate aerosol-radiation interaction. Blends of SAF also result in lower aromatic fuel content, which will result in reduced soot number concentrations. This will be beneficial to air quality in the vicinity of airports. The climate effects of reduced aromatics and soot numbers from SAF is less certain, but early observations indicate reduced ice crystal numbers, as predicted by microphysical modelling of plumes, and one model shows reduced contrail cirrus RF from reduced soot from SAF usage. The same overall outcome of reduced sulphur and aromatics can also be achieved by hydrotreating jet fuel. Faber *et al.* (2022)[195] estimate that to reduce aromatic content of fossil kerosene would require an additional 97 kg of CO_2 per tonne of kerosene at a cost of €10 per tonne kerosene (2021 prices).

The technological requirement to run low aromatic content fuel in aircraft fuel supply systems will represent a partial barrier to SAF fuels of >50% content (the currently allowable limit), since elastomers in the fuel supply system will need to be developed and changed for these purposes.[196]

7.2 The potential impacts of continued usage of fossil kerosene fuel

One of the possibilities, or scenarios, of future aviation is the continued usage of fossil kerosene but with permanent equivalent removal of CO_2 from the atmosphere. This topic has had little or no discussion in the aviation-related literature, and much of the literature in terms of CO_2 mitigation of aviation fossil fuel is focussed on substitution, or alternatives, to liquid hydrocarbons as a fuel type, *e.g.*, liquid hydrogen.[197] The development of biofuels has been under discussion for over a decade, and more recently, the manufacture of synthetic fuels for aviation has been under discussion,[198] under a number of potential (sustainable) production pathways,[199] including powering the chemical engineering process from sunlight and using air as a carbon source.[200] The largest barriers to producing synthetic fuels are the costs and feasibility of powering the process at scale by renewable energy and powering and removing ambient CO_2 by direct air capture (DAC). In essence, the process of production of synthetic fuels is the 'reverse' of combustion, by which a carbon source (*e.g.*, CO_2) is combined with a hydrogen source (*e.g.*, water) to produce kerosene. The energetic requirements are considerable.

Given the evident difficulties of producing enough SAF at scale, economically, a possibility might be an alternative of

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continuing to use fossil kerosene but to use a DAC process (when developed for use at scale) to capture and permanently store the equivalent amount of CO_2. This could potentially cut the production costs dramatically, although increases in fuel prices would still be a likely outcome. There are a variety of issues to be acknowledged and considered (although not addressed here since they are beyond the paper's scope): public acceptability, continued usage of fossil fuel, risks involved in 'putting off' mitigation rather than pursuing multiple lines of technology and policy, the as-yet unknown constraints of renewable energy and DAC availability, the continued availability of the kerosene fraction at sufficient demand volumes under a 'low carbon/reduced fossil fuel usage' scenario, and lastly, non-CO_2 aspects of aviation effects on climate.

In terms of non-CO_2 effects, if such a scenario of continued fossil fuel usage were pursued, it is clear that non-CO_2 effects would not be mitigated over today's level of effect (which is proportional to the growth rate of aviation fuel usage). The magnitude of the net NO_x effect in the future, remains unclear, even to its sign (positive/negative), regardless of (liquid) fuel pathways taken because of its dependence on future background emissions (see Section 4.3). It has been argued that SAF represents a significant opportunity to co-mitigate contrails and contrail cirrus with reduced aromatic content fuels.[201] Equally, it would be possible to hydrotreat fossil fuels to remove the aromatic components and S. This would have an associated energy expenditure (see Section 6.1)[195] but would nonetheless be possible. Both proposals remain problematic and uncertain, since the contrail-forming potential in the low soot emission regime ($<10^{14}$ particles per kg fuel) at low temperatures is unknown although modelling suggests an increase.[110] Hydrotreating fossil fuel to mitigate contrails is also problematic, since as per the 'SAF case', the contrail-forming potential in the low soot emission regime ($<10^{14}$ particles per kg fuel) at low temperatures is unknown and in addition increased (system) CO_2 emissions need to be considered. Even if the increased energy requirement to hydrotreat the fossil fuel was from renewable sources, this would neglect the question of whether such renewable energy was better utilized in abating fossil CO_2 emissions, elsewhere in the energy system. Usage of such hydrotreated, reduced aromatic content fuels (and SAF-type fuels) would also depend on the ability of current engine/fuel technologies to adapt to such fuels, in terms of fuel line integrity and elastomer sealing.[202]

8 Measuring mitigation–emissions equivalency metrics

Emissions are conventionally expressed in corporate reporting, and national reporting to the UNFCCC, in terms of an aggregate 'carbon footprint' that combines CO_2 and non-CO_2 effects into a single 'CO_2-equivalent' (CO_2-e) emissions using the 100 year global warming potential metric (GWP_{100}).[203] Other emission metrics, such as the Global Temperature change Potential (GTP)[31,206] are also in use, and the metric used along with the time horizon are a matter of user choice.[77] Because of the current large contribution of SLCFs, such as contrail cirrus, to aviation's climate impact, the 'apparent' amount of aviation CO_2-e emissions varies strongly according to both choice of metric and time horizon. These choices strongly influence the perceived importance of total (CO_2 + non-CO_2) aviation emissions relative to other sectors (for example, the impact of energy, dominated by CO_2, is not as sensitive to CO_2-e metric choice). Problems with CO_2-e emissions become particularly evident when SLCF emissions are constant or falling.[207]

Fig. 20 shows that the perceived non-CO_2 emissions as CO_2-e are highly variable, by a factor of approximately 20 across a time horizon (TH) of *e.g.*, 100 years, depending upon the metric used. For example, for SLCFs like contrail cirrus, the annual rate of CO_2-e emissions is given by the time-averaged radiative forcing due to that forcer divided by the 100 years absolute global warming potential ($AGWP_{100}$) of CO_2. Representing SLCFs with CO_2-e understates their immediate (sub-decadal) impact on global temperatures but overstates their impact on longer (century) timescales. Motivated by the urgency of the climate issue, some argue for metrics that emphasize more immediate impacts, such as GWP_{20}. Since the $AGWP_{20}$ of CO_2 is almost a factor of 4 smaller than its $AGWP_{100}$, using GWP_{20} in place of GWP_{100} more than doubles nominal total aviation emissions, leading to considerable confusion over the actual impact of aviation emissions relative to other sectors and the perception that there is no non-arbitrary way of calculating aviation's climate impact.

This perception is incorrect: the impact of aviation emissions on global temperature is well understood (albeit quantitatively uncertain) and not dependent on arbitrary metric choices. It can be expressed as the amount of CO_2 that would have the same impact on global temperature as aggregate aviation emissions over a given period, accounting for their amount and composition and (crucially) how aviation emissions are changing over time. This quantity, known as 'CO_2-warming-equivalent' emissions, can be calculated in a number of ways, including using a full-scale climate-carbon-cycle model,[208] but a number of approximations have been introduced over recent years, including GWP*—GWP 'star'[207,209,210] and the combined global warming/temperature-change potentials (CGWP & CGTP).[211] All give broadly similar results. The bar on the right of the Fig. 20 shows CO_2-warming-equivalent emissions corresponding to aviation emissions in 2018 calculated using GWP*.[212] However, it must be stressed that the Fig. 20 CO_2-warming equivalent emissions are specific to aviation emissions in 2018 (and their historical variation). As discussed by Lee *et al.* (2021),[6] this equivalence would be quite different in future scenarios that differ significantly from the current trends in aviation emissions.

While there is still no consensus on what metric is 'best' for quantifying sector-level emissions, it is widely agreed that specifying aggregate cumulative and short-lived climate forcing agents separately in both emissions reporting and target-setting is essential for transparency, since it allows their combined impact on global temperature to be calculated.[213] In

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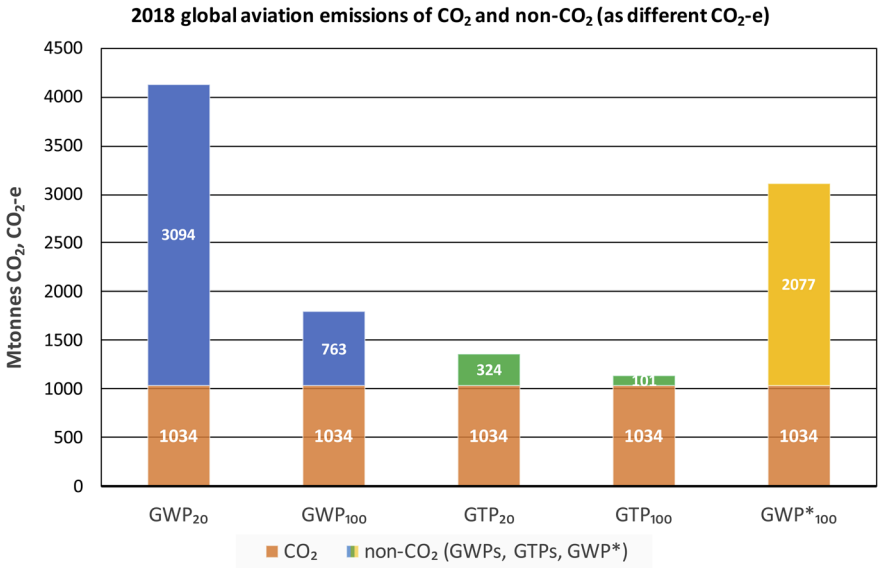

Fig. 20 Emissions from global aviation in 2018, expressed as Mtonnes of CO_2 and apparent emissions of non-CO_2, expressed as various metrics and time horizons. The CO_2 emissions (orange) for 2018 are evidently invariant, whereas the non-CO_2-e emission magnitude varies with metric (blue, GWP; green GTP, yellow GWP*) and time horizon. Data from Lee *et al.* (2021).[6]

contrast, the temperature impact of aggregate CO_2-equivalent emissions using GWP_{100} (or any other traditional metric) is ambiguous.

9 Tradeoffs between emissions and their effects

9.1 Trading between long-lived greenhouse gases and short-lived climate forcers – the issues

One of the fundamental problems with non-CO_2 mitigation for aviation is that there can be trade-offs between either technologies, operations, or policies that target non-CO_2 effects, which result in increased CO_2 emissions. This leads to a question of "is it of net benefit?", which is very difficult to answer, since it invokes a metric-based comparison of the non-CO_2 radiative effects with the CO_2 radiative effect. As has been outlined in Section 7, this is at the heart of the CO_2 equivalence emission calculations; although warming-equivalent emissions can be estimated (acknowledging the forcing uncertainties) for current emissions, the relationship between warming due to non-CO_2 emissions and CO_2 emissions could alter dramatically in future. The issue comes down to temporal behaviour of short-lived climate forcers, such as net NO_x, and contrails, being compared with a long-lived greenhouse gas such as CO_2.

In the case of NO_x, this is largely envisaged to be a technological trade-off, whereby improved fuel efficiency can make reductions of NO_x more difficult (see Section 3). This is at the core of the allowance of increased NO_x emissions for higher OPR engines in the CAEP regulations, as shown in Fig. 2. This issue is sometimes posed in the opposite sense, *e.g.*, "an x% reduction in NO_x would be possible but may result in a y% fuel (CO_2) penalty – will this yield a net benefit?".

For contrail formation, the potential trade-off is usually posed from it being a consequence of an operational measure, since no technological measures appear to be available, other than potentially reducing the aromatic content of fossil fuel, which will have a refinery (lifecycle) cost of increased CO_2, of 97 kg CO_2 per tonne of kerosene, according to one estimate.[195] Increased fuel efficiency in large modern turbofan engines is thought to have resulted in cooler exhaust through improved propulsive efficiency, resulting in contrail formation over slightly greater vertical extents of the atmosphere.[214] Avoidance of contrails can in theory be done by changing trajectory over an assumed ideal for fuel efficiency, usually in the vertical extent of a changed cruise altitude, but for a potential CO_2 penalty from non-fuel-optimal cruise altitudes.

The perhaps single exception of a candidate for a 'win–win', is the increased usage of SAF—targeting a reduction in life-cycle fossil CO_2 emissions—for which initial results indicate a reduction in contrail visible optical depth and forcing from contrail cirrus (see Section 6) in the soot-rich regime ($>10^{14}$ particles per kg fuel). However, while there is limited observational evidence for reduced ice crystal number from the use of SAF fuel low in aromatics,[181] the basis for estimating a consequential reduction in forcing is weak, being based on only one model, and there is the significant uncertainty of what happens in the low soot regime (see Fig. 18), with even unintended outcomes possible of increased contrails.

9.2 NO_x *vs.* CO_2

Higher combustion temperatures and combustor inlet pressures have gone hand-in-hand with fuel efficiency improvements of many combustion systems. This tends to increase the propensity for the engine system to produce NO_x, formed from the combination of atmospheric nitrogen and oxygen. This is

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generally mitigated by different combustor technology approaches, as outlined in Section 3.

Very little work studying the atmospheric implications of such trade-offs between NO_x and CO_2 has been undertaken. Freeman *et al.* (2018)[215] formulated a simple first approach to a hypothetical case of a 20% reduction in NO_x implying in a 2% increase in fuel consumption[20] by running a constant emissions scenario and examining the RF and temperature response differences after 100 years. For a 2% fuel penalty, NO_x emissions needed to be reduced by >43% to realize an overall benefit. Conversely, to ensure that the fuel penalty for a 20% NO_x emission reduction did not increase overall forcing, a 0.5% increase in CO_2 was found to be the 'break even' point (no benefit or disbenefit).

9.3 NO_x *vs.* nvPM

Production of NO_x in the combustor generally occurs under hot combustor conditions (with a high fuel-to-air ratio) and high temperatures. NO_x formation has slight preference towards the lean side of stoichiometric conditions, as oxygen ions are the rate limiting step in creation of NO_x. The reaction for NO_x production tends to be relatively slow compared to other reactions in the combustor but the rate of NO_x production increases significantly at higher temperatures. Particle formation tends to increase when the mixture is fuel-rich (*i.e.*, a higher fuel to air ratio) in the primary zone of the combustor. In addition, burning off any particles formed in the combustor requires high temperatures on the lean side of stoichiometric conditions. Designing an efficient, reliable combustor to minimize both NO_x and nvPM can thus present some basic technology trade-offs.

Technology that has been developed to minimize NO_x production includes two basic types: the lean-burn combustor and the RQL combustor (see Section 3).

The lean-burn type minimizes NO_x production by ensuring while the mixture is always lean, the temperature is kept well below that needed for effective NO_x production, which in turn can require a longer time for full combustion. It is a delicate balance between time and temperature, as when exit temperatures become high (approaching 2000 K) the time required for full combustion can mean that there is sufficient time for NO_x to form and the NO_x levels can start to rise to levels above those found in RQL combustors. Overall, though, the lean burn conditions tend to minimize particle formation with lower NO_x emissions.

In RQL combustors, the fuel initially burns rich where there is insufficient oxygen to form NO_x, then the mixture is rapidly 'quenched' to lean conditions. There is only a brief period when NO_x formation is appreciable and as the formation of NO_x is relatively slow, the short time at these conditions is relied upon to minimize production. With RQL combustion the production of nvPM (*i.e.*, soot) tends to occur in the fuel rich regions of the combustion system and the particles are then consumed in the leaner regions of the combustor. However, as noted in Section 3.2 the mixing is never wholly complete and the level of NO_x and nvPM depends on how uniform the different regions are. This inherent trade-off in the RQL combustors, which were originally conceived for NO_x reduction, means that the controlling nvPM at the same time as reducing NO_x provides some challenges to the design.

Although these qualitative directional trade-offs in combustion design for NO_x and nvPM emissions are well-known there is a lack of available quantitative data on these trade-offs, and no scientific assessments have been performed to our knowledge on, *e.g.*, net NO_x ERF *vs.* contrail ERF. Such a comparison would not have the difficulty of comparing warming over very different timescales, *e.g.*, a SLCF with CO_2; however, it would necessitate the comparison of two very poorly quantified SLCF ERFs.

9.4 Contrail avoidance *vs.* CO_2

Section 4.2 and Fig. 21 highlight the significant difficulties in the putative efforts to reduce net aviation forcing by navigational avoidance of contrails. There are two distinct but equally important aspects to the problem. The first problem is that navigational avoidance is heavily dependent on the quality of meteorological forecasts of ice-supersaturated regions which have been shown to be inadequate for this purpose in several studies.[54,55] Assuming that navigational avoidance diverts aircraft from their minimal fuel route (and hence minimum CO_2 route), then false alarms (contrails predicted but they did not occur) indicate an unnecessary diversion. False negatives (contrails not predicted but they did occur) mean a missed opportunity. Robust statistics on these are needed to assess whether diversions would have avoided contrails, and these are not yet available. A worst case would be a false alarm on the original route but a false negative on the diverted route.

The second problem is that navigational avoidance is a necessary but not a sufficient condition in strategies to reduce aviation climate effects. Even if contrails are successfully avoided, asserting that any potentially increased CO_2 emissions are justified is required. Again, there are multiple issues. One is that, even if the forcing of the time- and location-specific avoided contrail was known, any gain would depend on the metric chosen to compare contrail and CO_2 climate impacts. For metrics such as the GWP or GTP, use of a longer time horizon would in general make it harder to justify diversion. Another is that in general the forcing due to a contrail (avoided or caused) is subject to multiple uncertainties (as is clear from Fig. 1) and so assessing the size of a diversion that is justified becomes difficult; contrail forcings cannot be observed with any accuracy (the avoided forcing is even harder to assess!) so there is a heavy reliance on model estimates. In addition, the assessment has to be done for the specific case of the forcing caused by the contrail (or avoided) rather than using global mean forcing estimates such as those shown in Fig. 1; that localized forcing will depend on many parameters, including time of day and location and how these change over the lifetime of the contrail. A further issue is that any assessment should consider other non-CO_2 forcings; even if the contrail plus CO_2 forcing is reduced, this is no guarantee that the net aviation forcing is reduced.

9.5 Contrail reduction *via* fuel composition *vs.* CO_2

It is now well established through testing of emissions characteristics at ground and cruise levels that usage of liquid

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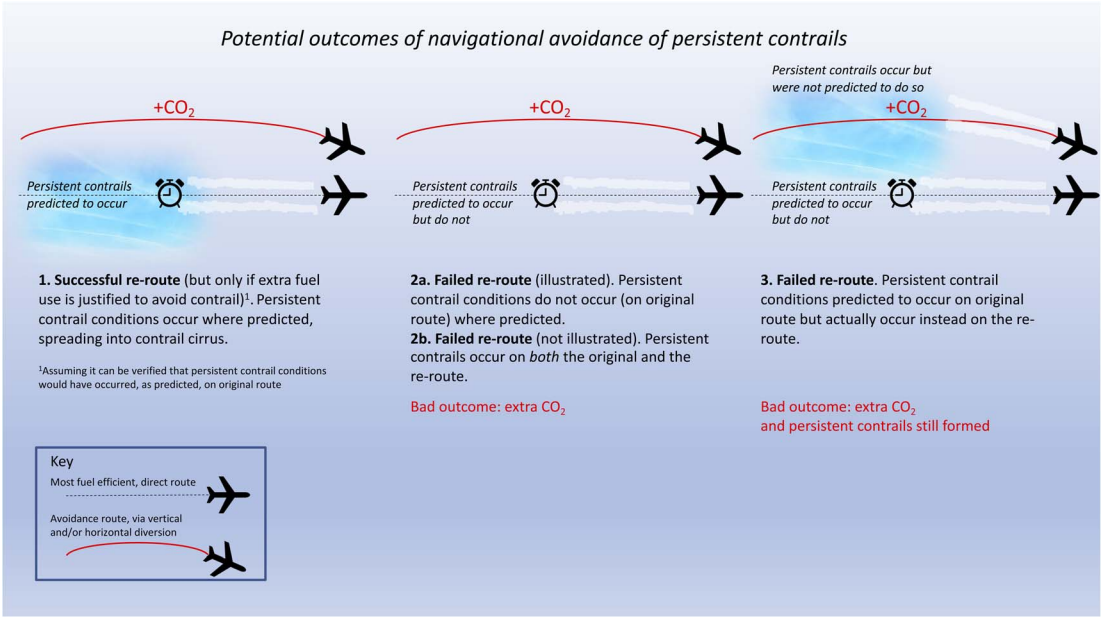

Fig. 21 Potential outcomes of navigational avoidance of persistent contrails showing a successful re-route and failed outcomes because of failure to predict ISSR correctly. From Shine and Lee (2021).[216]

kerosene-like SAF, intended to reduce life cycle emissions of fossil CO_2, with a reduced aromatic content, consequentially has lower emissions of soot particles.[184,217,218] Recent measurements have confirmed that the ice particle number concentration is also reduced with the usage of SAF.[181,219]

Modelling studies have suggested that SAF, with its lower aromatic content, produces fewer but larger ice crystals that sediment and sublimate more quickly and thus have a lower radiative forcing;[190–192,220,221] see also Fig. 19. Assuming lower life-cycle emissions of CO_2 in the SAF deployed, this could potentially be a 'win–win' situation. However, this remains somewhat speculative with contrasting results[193] who authors suggest that SAF usage results in a larger RF for contrails, and an incomplete representation of particle microphysics in such modelling remains problematic, since ultrafine aqueous particles in the low soot regime may increase contrails.

More recently, it has been suggested that such SAF (which, at present is limited in quantity) might be deployed more effectively in combination with a prediction of contrails, such as outlined in Section 8.3. Such an approach may be effective[201] but the approach would need to solve the ongoing difficulties of predicting ISSRs and the assumed benefit of SAF on contrail forcing needs to be unambiguously demonstrated, which is unclear because of the lack of explicit consideration of volatile particles at low soot numbers (see Kärcher, 2018).[110]

Given that it has been shown that a reduction in the aromatic content of fuel (as demonstrated by usage of SAF) can reduce soot number and ice crystal number (as per measurements) and potentially RF (as per modelling), it might also be suggested that a reduction in aromatic content of fossil fuel also represents a more immediate mitigation opportunity, to reduce the contrail RF. Aromatics are present at approximately 20–25% of fuel, by volume, and have been found to prevent shrinkage of elastomer seals in aircraft fuel systems. If new future elastomers can safely withstand lower aromatic levels,[222] then lowering them may ultimately reduce contrail RF.

Faber *et al.* (2022)[195] made a first assessment of potential energy costs at the refinery to reduce aromatic content of aviation kerosene and calculated an increase of 97 kg CO_2 per tonne of fuel produced, an additional 3% of the fuel burned. Thus, such an approach would still invoke a 'tradeoff' situation requiring CO_2-e with all its concomitant problems, in which it would be necessary to ensure that there was a net climate benefit, rather than disbenefit. An alternative might be to ensure that the processing of the fuel was from renewable energy sources. While this might be preferable, it does not remove the necessity of a tradeoff assessment, since if the renewable energy available were alternatively used for a direct CO_2 emission reduction, this would have a far more certain outcome.

10 Strategic decision-making and investment in relation to non-CO_2 emissions

One of the main recurrent themes emerging from this assessment is that because of the linkages between aviation non-CO_2 and CO_2 emissions, they cannot be considered in isolation, since any mitigation efforts on non-CO_2 may have consequences for CO_2 emissions, and similarly between non-CO_2 emissions.

Non-CO_2 emissions, other than NO_x, are potentially modified (to a greater or lesser extent) using different future liquid SAF-type drop-in replacement fuel-types, although it will be

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some time before these are produced at the scale required for commercial aviation. Although not considered here, liquid hydrogen as a power source, would have a larger direct water vapour emission, and although there are no or few combustion particles (soot), it is likely that the contrails formed will have different characteristics in terms of density, lifetime and radiative effects.[79] Emissions of NO_x are thought to be similar to liquid hydrocarbon fuels, since this is a function of combustion, not the fuel source.

10.1 Non-CO_2 emissions under a scenario of continued usage of fossil fuel

Under assumed continued use of fossil-based kerosene, the main non-CO_2 environmental pressures are on emissions of NO_x, nvPM (soot) and to a much lesser extent, SO_2 (in terms of affecting local air quality). NO_x and nvPM are regulated by ICAO-CAEP and are therefore subject to international regulatory pressures.

The case for further NO_x regulation within ICAO is based upon the requirement to protect local air quality, since it is the levels of NO_2 in ambient air that are the subject of local/national regulations, to which many sources contribute, including aircraft during the LTO cycle in and around airports. In addition, there is some limited evidence that cruise emissions may contribute to air quality degradation at the ground although this is subject to significant modelling uncertainties (see Appendix 1). Ambient air quality regulations for NO_2 (and in some cases, NO_x) are based on uncertain epidemiology which may have historically conflated the effect of ultrafine particles, which tend to be co-emitted alongside NO_x for many sources. The evidence for toxicological effects of NO_2 appears to be rather weak. However, there is no sign of ambient air quality regulations being relaxed in the foreseeable future and, if anything, the recent revision of recommended limits downwards by the WHO[140] by a factor of 4 (as an annual average) are likely to increase (inter)national regulatory pressures, so ICAO-CAEP NO_x emission regulations remain an issue for manufacturers to comply with and for them to take a precautionary view on possible future standard changes. For the climate effect of NO_x emissions, this remains a highly uncertain term, with the models and the science requiring further refinement to quantify the present-day and future effect. That there is net present-day positive ERF from aircraft NO_x emissions should not be taken to be a time-independent conclusion, with two studies finding that the net radiative effect could be negative in the future, depending on the magnitude of both the aircraft emissions, and other anthropogenic surface precursor emission sources of ozone. Cruise NO_x levels are not currently regulated for climate protection purposes, although the inclusion of a representative certification point for this phase of flight is under discussion at CAEP.

For nvPM (the regulatory measure at CAEP, whereas here we have more broadly used the term 'soot'), once again, the primary purpose of the CAEP regulations is to protect human health from LTO emissions, and similar to NO_x, ambient air quality standards are the driving force, although these tend to be specified in terms of a mass-basis of $PM_{2.5}$ or PM_{10}. Since most aircraft primary emissions of nvPM/soot are in the ultrafine particle size range, *i.e.*, 100 nm or less in aerodynamic diameter, they are unlikely to contribute greatly on a mass-basis, although the CAEP emission regulations are for mass and particle number. There is more certainty in the toxicological literature that ultrafine particles can be harmful for human health, so the evidence base for emission regulation is perhaps more robust than for NO_2. The key difference between the two ambient air quality type regulations is that the measurement/assessment basis for ultrafine particles is rather poor, being mostly mass-based, rather than number-based.

In terms of climate, the role of soot particles in contrail formation is relatively well understood, and the effect of emissions reduction (number concentration) on ice crystal formation has been observed in a few cases.[181,219] However, the specific quantification of the climate effect of their reduction is poorly quantified, since it depends on modelling rather than measurement.[191] Soot particles from aircraft engines may also play a role in the postulated modification of cirrus[110] although the magnitude and sign of the radiative effect is still under debate.[98,101,108] It is well understood that soot emissions can be reduced by changes in combustor technology and also by reduction of the aromatic content of the fuel.[181,195]

Sulphur dioxide emissions are a function of the composition of the fuel used and the current upper value as stipulated by Defence Standard 91–91 and ASTMD1655 is 3000 ppm (by mass). Levels of S are generally considered to be at around 600 ppm,[176] although the database for this assessment is very sparse. Sulphur dioxide is the primary pollutant resulting from sulphur in the fuel in aircraft exhaust (by about 98%)[177] and oxidises relatively slowly (typically, 1% per hour) in the atmosphere to form sulphate particles. Thus, for LTO emissions, the additional mass-based particulate burden of the atmosphere from aircraft exhaust does not occur in the vicinity of the airport. The contribution of sulphate to ground-based concentrations of particles has been discussed (see Appendix 1), and in terms of mass, greatly exceeds that of nvPM allegedly contributing to premature mortalities. In terms of climate, S does not contribute significantly to contrail formation, soot particles being the primary condensation particle, but there is the potential for it to form a small but significant fraction of UFPs in the early plume stage. At current levels of soot emissions, these do not play an active role in ice crystal formation but there exists the possibility that with reduced soot emission number, at less than 10^{14} particles per kg fuel, UFPs could increase the ice crystal number concentration at temperatures well below the threshold of 233 K for contrail formation.[110] Sulphur and soot effects are often considered to be independent of each other but the potential interaction of particle microphysics (sulphuric acid around soot particles) in ice crystal formation should not be ignored.

Sulphate particles are responsible for a small negative ERF due to direct aerosol-radiation interaction (see Fig. 1) and the potential for aircraft sulphate to contribute to increased droplet number density in lower-altitude water clouds, with a potentially significant negative ERF, to 10s of mW m^{-2} (see Fig. 5 of Lee *et al.*, 2021, and references therein)[6] but with no best estimate available, indicating the very low confidence level in this term.

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10.2 Non-CO_2 emissions under a scenario of SAF usage

Here, as outlined in Section 1, we interpret a liquid kerosene-like sustainable aviation fuel, or 'SAF' as being a drop-in liquid hydrocarbon fuel that has a lower fossil carbon footprint (on a lifecycle basis) than kerosene. This might be certain types of biofuels or synthetic fuels produced from renewable electricity to produce hydrogen, which is combined with a carbon source, such as CO_2 directly captured from the atmosphere. The merits, energy requirements, and costs of such fuels are not considered here, rather their consequences for non-CO_2 emissions.

The usage of SAF will impact on non-CO_2 emissions of water vapour, SO_2, and nvPM. SAF tends to have a lower overall molecular weight with a greater H/C ratio than conventional fossil-based kerosene; this gives a slightly increased energy density but also greater water vapour emissions. However, this is of the order 5% or less and the existing direct ERF term of water vapour (see Fig. 1) is small, some 2% of the total. The initial water vapour emission index also affects the thermodynamics of initial contrail formation, according to the Schmidt-Appleman criterion[48] but the effect of this apparently small change has not been assessed.

SAF also contains very low or zero S in biofuels[176] and in synthetic fuels. In terms of air quality, this will likely reduce the mass of secondary S-based particulates formed from aviation emissions, which represent in excess of 95% by mass according to Barrett *et al.* (2010).[159] In terms of climate, low/zero S will remove the small negative direct aerosol-radiation ERF, and the much less certain S aerosol–cloud interaction forcing.

The usage of SAF perhaps most significantly reduces soot emissions by mass and number. It was outlined in Section 6 that aromatic compounds found in fossil fuel are largely responsible for soot emissions from aircraft gas turbines, and that measurements of soot number in the exhaust of SAF-fuelled engines show much lowered concentrations of soot number[181,184,223] and ice crystal number concentrations.[181,219] These measurements confirm earlier modelling studies,[187,189,190,220] which were used in global modelling studies that suggest reductions of soot and ice crystal number from SAF reduce the RF, and hence likely the ERF, of contrail cirrus of contrail cirrus.[191,192] However, this generalization only applies to the soot rich regime of $>10^{14}$ particles per kg fuel; under this threshold, the outcome is uncertain.

10.3 Synthesis

In terms of continued fossil fuel usage, reductions in NO_x will need to be necessary, if ICAO-CAEP regulations are revised. There is a precedent for these reductions and a reduction ('stringency') has not been introduced since 2010 (CAEP/8). Emissions of nvPM ('soot') may also be reduced *via* a further updated ICAO-CAEP regulation, although it is noted that reductions in aromatic content of fuel *via* SAF replacement could reduce emissions of soot significantly. Whether this results in a net benefit has not been evaluated.

If, and when SAF, becomes more widely available, this will have the consequence of reducing nvPM (soot) emissions. This may take some pressure off combustion technology requirements. To the best understanding, uptake of SAF will not affect NO_x emissions and there will therefore still be a requirement to comply with ICAO-CAEP regulations.

11 Future directions and research needs

11.1 Future research – aviation and climate

It is clear that there is an appetite amongst some stakeholders for non-CO_2 mitigation of aviation effects on climate, but we have serious reservations over recommending definitive courses of action until there is better quantification of the actual effects, and further studies of the tradeoffs between non-CO_2 reductions *vs.* potential CO_2 increases. It is realised that this represents a serious barrier to technology development and policy making but there are no short cuts, and the underlying danger is of either nugatory and expensive efforts which are not easily reversed or making matters worse in terms of the total climate effect of aviation.

Future research for climate effects is categorized in two ways: firstly, a basic improvement to the various ERF terms; secondly, the application of better quantified ERF terms in determining mitigation outcomes, and optimization of mitigation to reduce overall effects on climate. There are many details that could be highlighted but here a high-level approach is taken to identify where the main problems lie.

Contrails and contrail cirrus: this is currently understood to be the largest non-CO_2 ERF term for present-day aviation but still remains poorly quantified by a limited number of models. The uncertainties are large because of a lack of physical understanding and quantification of basic processes and their dependencies, including both cloud physics important in the formation and persistent of contrails, and radiative processes, required to quantify the ERF of cirrus. Moreover, there are only a small number of global models with which the ERF is quantified (which have a variable range of complexity and completeness in terms of representation of processes from fuel composition through to forcing) and a poor observational basis for verification. More global modelling efforts of ERF are required, along with the actual climate response (in terms of ΔT), and further analyses of observational data should be made. More understanding is required of the effect of adopting new fuels on contrail formation and representation of missing processes is important. In the meantime, the effect of SAF on contrail forcing remains unresolved. Organic particles from lubricating oils have also been suggested to play a potential role in particle emissions and ice nucleation[224] and this should be investigated.

Net-NO_x effect: while there are many models that have been used to assess this, the larger sample size should not be misunderstood as fully constraining the uncertainties, since the models are similar. There are still basic properties that are poorly quantified, such as the 'effective' NO_x emissions, since small-scale and relatively fast conversion processes that occur in the plume are not generally considered in global modelling.

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Since the background and plume processes are co-dependent, this makes a solution to this modelling problem challenging. Plume scale effects need to be incorporated. Direct observational verification of the formation of short-term ozone has not so far been possible because of the small size of the perturbation relative to the background. Individual processes, such as chemical kinetics/reaction rates are observable in the laboratory. Virtually all the modelling has been of RF (stratospherically adjusted) and only one attempt at modelling the efficacies of the NO_x perturbations (utilized to give an ERF) has so far been reported. The ERF and climate response of ozone and methane effects urgently needs addressing, since the ERF adjustments are relatively large 1.37 for short-term ozone, 1.18 for methane, and associated processes.[6] The role of nitrate particle formation in quantifying the ERF remains open and should be researched further. The regional temperature effects of opposing signs (and quite different geographical distributions) of forcing from methane and ozone, especially under future conditions of a negative net global-mean forcing, need to be investigated.

Aerosol–cloud interactions of soot and sulphur species: the size and sign of these effects has remained elusive for the past 15 years with still no clear convergence of results. The sign of the S aerosol cloud interaction is likely to be negative but of unclear magnitude. Although there is some convergence that the sign of the aerosol–cloud interactions of soot are negative, there is still vigorous debate in the literature over its magnitude, and sensitivity studies are still able to show positive forcings. Improvements to global models and how basic processes are parameterized are needed, alongside laboratory measurement of aerosol properties to provide reliable parameters for the models.[225] Moreover, the linkage between contrail cirrus models and the effects of soot in aerosol–cloud interactions needs to be made. As mitigation of one (through reduced soot emissions) has consequences for the other.

Improvements to understanding mitigation options: aviation non-CO_2 climate effects depend sensitively on where and when emissions occur, and so a 'one-size fits all' approach to mitigation may be inappropriate. Nonetheless, some progress might be made relatively quickly, albeit that outstanding uncertainties from the basic understanding of ERF terms will persist until improved and constrain the study of mitigation options.

For potential operational mitigation options, the research requirements are much greater, since basic properties of the atmosphere (*e.g.*, ISSR occurrence and its prediction) are not well quantified and need to be improved before such mitigation can be contemplated for operationalization.

11.2 Future research – aviation and air quality

For air quality effects of aircraft emissions, the challenges have some similarities but also differences to climate effects.

Quantification of emissions: NO_x and nvPM emissions are characterized carefully through the emissions regulatory process at ICAO-CAEP. These measurements relate to the LTO cycle, and there are outstanding issues and uncertainties to converting them to non-LTO (principally cruise) emissions, which need to be resolved. The role of lubricating oil in potential particle emissions requires further work.

Source attribution, LTO cycle: calculation of the contribution of aircraft sources to local air concentrations of pollutants is only possible *via* modelling. Different countries and regions may (or may not) have their own approved or recommended practices to modelling this in terms of local air quality. The adequacy of such modelling is beyond the scope of this present document.

Source attribution, cruise-to-ground: there is also a potential contribution of non-LTO emissions to ground-level air quality. This is very poorly quantified, generally by global models that may not be entirely suited to such an assessment purpose because of their spatial scale and parameterization of complex atmospheric processes that re-distribute high-level emissions to the earth's surface. Also, the basic assessment approach is one of particle mass affecting health, which is highly questionable and may imply targeting the wrong emission.

Aircraft emissions and human health impacts: while there are unambiguous air quality standards for gaseous and particulate pollutants, and regulations that apply in various states and regions of the world, the actual contribution of pollutants to harming human health is not well-defined. This is a matter for fundamental research into toxicology and dose–response functions, since much of the research is based on epidemiology which may have multiple causal factors. It is clear, however, that UFPs represent a serious and confirmed threat to human health.

Author contributions

David Lee: funding acquisition, project administration, conceptualization, investigation, methodology, writing – original draft, writing – review and editing. Myles Allen: conceptualization, investigation, methodology, writing – original draft, writing – review and editing. Nicholas Cumpsty: conceptualization, investigation, methodology, writing – original draft, writing – review and editing. Bethan Owen: conceptualization, investigation, methodology, writing – original draft, writing – review and editing. Keith P. Shine: conceptualization, investigation, methodology, writing – original draft, writing – review and editing. Agnieszka Skowron: conceptualization, investigation, methodology, writing – original draft, writing – review and editing.

Conflicts of interest

There are no conflicts to declare.

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1 https://www.theguardian.com/environment/2020/dec/16/girls-death-contributed-to-by-air-pollution-coroner-rules-in-landmark-case.

2 In the summary, and elsewhere, we adopt the framework of the Intergovernmental Panel on Climate Change to assess 'confidence levels' (Mastrandrea *et al.*, 2011).[80]

3 Non-volatile particulate matter (nvPM) is a regulatory definition, which has strict conditions of measurement under ICAO Annexe 16. In the atmosphere, this equates to 'soot', which is defined here as a mix of black and organic carbon. Soot is emitted in aircraft engine exhaust as very small particles, mostly under 100 nm in diameter. In the literature, 'black carbon' (or BC) is sometimes used interchangeably with soot. Here, 'soot' is preferred since it encompasses organic compounds as well.

4 Most of the primary emission of NO_x is as NO. It is quickly converted to NO_2 by ozone (O_3) in the atmosphere. A small fraction of the NO_x may be a primary emission of NO_2 at higher power settings, although the fraction can be large at idle (Wormhoudt *et al.*, 2007)[5]

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