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Identifying the ozone-neutral aircraft cruise altitude

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HIGHLIGHTS

- We use adjoint methods to calculate ozone loss resulting from flights at any altitude.
- We find that ozone depletion from aircraft depends non-linearly on flight altitude.
- Supersonic aircraft are likely to cause significant ozone depletion.
- Flight at 13–14 km altitude could be "ozone neutral".
- Inclusion of sulfur in fuel may increase this altitude by 0.5–1.0 km.

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ABSTRACT

Depletion of stratospheric ozone, and the associated increase in population exposure to UV radiation, is an environmental consequence of high-altitude, supersonic aviation. Assessments of the impacts of emissions from subsonic aircraft – which fly at lower altitudes – have instead shown that they produce a net increase, rather than decrease, in global net ozone, suggesting the existence of an intermediate "column ozone neutral" cruise altitude. Knowing this altitude and its variation with factors such as latitude, season, and fuel composition could provide a pathway towards reducing the environmental impacts of aviation, but would require a prohibitive number of atmospheric simulations. We instead use the newly developed GEOS-Chem tropospheric-stratospheric adjoint to identify the location of the column ozone-neutral aircraft cruise altitude as a function of these factors. We show that, although the mean ozone neutral altitude is at 13.5 km globally, this varies from 14.6 km to 12.5 km between the equator and 60°N. This altitude varies by less than a kilometer between seasons, but the net depletion resulting from flying at greater altitudes varies by a factor of two. We also find that eliminating fuel sulfur would result in a neutral altitude 0.5–1.0 km greater than when conventional jet fuel is burned. Our results imply that a low Mach number supersonic aircraft burning low-sulfur fuel (e.g. biofuels) may be able to achieve net zero global ozone change. However, for a fleet to achieve ozone neutrality will require careful consideration of the non-linear variation in sensitivity with altitude and latitude.

1. Introduction

Previous supersonic aircraft design, or newly proposed designs by organizations such as Boom, are designed to cruise at altitudes in the range of 14–20 km (Aerion, 2020; Berton et al., 2020; Boom, 2020; Spike, 2020). Studies of the emissions impacts from aircraft at such altitudes have generally found a net decrease in column ozone - a greenhouse gas and air pollutant (Cunnold et al., 1977; Grewe et al., 2007; Kawa, 1999; Kinnison et al., 2001; Tie et al., 1994; Cunnold et al., 1977; Grewe et al., 2007; Kawa, 1999; Kinnison et al., 2001; Tie et al., 1994) – with even greater depletion projected for hypersonic aircraft flying at altitudes of up to 30–40 km (Kinnison et al., 2020). The decrease in global column ozone from supersonic aviation emissions is associated with an increase in population exposure to solar UV

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radiation, which damages the DNA and is a leading cause of melanoma (van Dijk et al., 2013).

Commercial activities from subsonic aircraft, cruising at altitudes between 10 and 12 km, cause ~4% of total anthropogenic radiative forcing and ~16,000 premature mortalities per year due to degraded air quality (Eastham and Barrett, 2016; Grobler et al., 2019; Lee et al., 2020; Quadros et al., 2020). High-altitude emissions from commercial aircraft activities are also the only significant anthropogenic emissions at altitude, leading to different effects than those of other sectors. Emissions of nitrogen oxides (NO_x) from subsonic civil aircraft at typical cruise altitudes of 10–12 km, for example, have an atmospheric lifetime of 10 days compared to a few hours for the same emissions at the surface (Jaeglé et al., 1998). Although NO_x emissions in most tropospheric locations will result in production of ozone, the lower temperatures, drier conditions, and longer residence times of the upper troposphere result in increased ozone production efficiency. NO_x emissions from subsonic aircraft therefore result in three times more ozone per unit mass when compared to near-surface emissions (Gilmore et al., 2013; Jaeglé et al., 1998). Although ozone is destroyed by NO_x from subsonic aircraft in the lower stratosphere, the amount of stratospheric ozone depletion is negligible compared to the increase in the troposphere (Köhler et al., 2013), thus leading to a net positive increase in column ozone from subsonic aviation emissions.

These studies raise the possibility of a net column ozone-neutral cruise altitude. Emissions from aircraft flying at this neutral altitude would induce no net change in column ozone, with the stratospheric ozone loss equaling the increase in ozone at lower altitudes. A column-ozone neutral altitude may represent a cruise altitude for aircraft at which some impacts – such as the risk of increased skin cancer rates or UV-related crop damage – can be minimized. Emissions at this altitude would not address other environmental damages such as changes in surface air quality or potential changes in stratospheric temperatures and circulation (Son et al., 2008; Xie et al., 2016). Furthermore, knowing the "ozone-neutral" cruise altitude might provide a step along the path towards limiting the total environmental impact of aviation.

To date the column ozone-neutral altitude has not been robustly quantified, and has at best been found only for a specific aircraft and route distribution (e.g. (Zhang et al., 2021)). Based on the estimated impacts of supersonic and subsonic aviation, it is likely to vary with factors such as the composition of aircraft emissions, the latitude of flight, and even the season. Quantifying the dependence of global ozone depletion on these factors can inform the design and regulation of future aircraft including both subsonic and supersonic civil airliners. However, most efforts to quantify ozone changes in response to aviation emissions have used computationally expensive chemistry transport models and multi-year integrations to account for the long stratospheric timescales (Zhang et al., 2021). Identifying the conditions which would result in net ozone neutrality as a function of space, time, and exhaust composition is therefore computationally intractable using conventional forward modeling approaches.

An alternative is to use adjoint-based methods (Hakami et al., 2007; Menut, 2003; Menut et al., 2000; Sandu et al., 2005). These calculate the sensitivity of global quantities with respect to emissions at any location in a single simulation. Such approaches have been used to constrain emission sources from measurements (Chai et al., 2009; Kopacz et al., 2010; Zhu et al., 2013) and to quantify the relationship between emissions and surface air quality (Ashok and Barrett, 2016; Gilmore et al., 2013; Henze et al., 2009; Koo et al., 2013; Schmidt, 2003). However, due to the long transport lifetimes and lack of removal processes in the stratosphere, stratospheric adjoint models demand more robust numerical stability, significantly more data storage, and longer (multi--year) integrations.

We extend the adjoint of the tropospheric-stratospheric chemistry-transport model (CTM) GEOS-Chem to enable multi-year adjoint simulations. A continuous nine-year simulation is performed to quantify the net change in global column ozone which results from aircraft emissions occurring at any latitude, longitude, altitude, and season. We then average results from the final five years of these sensitivities to identify the long-term ozone-neutral cruise altitude as a function of latitude and time of year. Finally, using a plume chemistry model to account for local-scale processes (Fritz et al., 2020), we quantify the relationship between the engine NO_x emission index (in grams of NO_x as NO_2 per kg of fuel burned), fuel sulfur content, latitude, altitude, and ozone depletion.

2. Method

Adjoint sensitivities are obtained using the GEOS-Chem adjoint model, for which we implement the Unified Chemistry eXtension (UCX) (Eastham et al., 2014), enabling modeling of stratospheric chemistry and feedbacks to the troposphere. The GEOS-Chem adjoint model consists of a base model and a differentiated model, both of which have been updated with the UCX. In addition to stratospheric chemistry based on NASA's Global Modeling Initiative mechanism, the UCX adds long-lived species such as N_2O and CFCs to GEOS-Chem, along with prognostic calculation of stratospheric aerosols, water vapor, and methane.

Previous assessments of the atmospheric impacts of supersonic aviation have found a high sensitivity to water vapor emissions in the stratosphere, where water vapor is rare and promotes ozone depletion (Kawa, 1999). The GEOS-Chem UCX prescribes tropospheric water vapor concentrations based on relative humidity fields from archived meteorology, while stratospheric water vapor is allowed to evolve and chemically react (Eastham et al., 2014). However, this leads to an excess inflow of water vapor in the stratosphere through the tropical tropopause. We thus add a new treatment for stratospheric water vapor which consists of prescribing an annual cycle for the water vapor mixing ratio at the tropical tropopause. A sinusoidal annual cycle in the water vapor mixing ratio is applied between 3.0 and 4.8 ppmv in the 30°S - 30°N latitude band. Evaluation of the base GEOS-Chem model was performed by Speth et al. (2021) and suggested that the model is capable of reproducing observed magnitudes and seasonal cycles in key stratospheric species, although vertical mixing may be underestimated. A brief summary of this analysis is also provided in the Supplementary Information.

Aviation emissions are computed from the Aviation Emission Inventory Code (Simone et al., 2013) and updated monthly. These aviation emissions inventories include aircraft fuel burn, NO_x, CO, and hydrocarbons emissions. SO_2 and H_2SO_4 aerosol emissions are calculated by assuming 600 parts sulfur per million parts fuel, by mass, and a 2% conversion rate of SO_2 to H_2SO_4. A constant emission index of 20 mg/kg_{fuel} is used for organic carbon. Water vapor emissions are computed assuming an emission index of 1.23 kg/kg_{fuel}.

All GEOS-Chem UCX adjoint simulations use the GEOS-5 meteorological product. Unless otherwise specified, we compute nine years of adjoint sensitivities, using meteorology for 2004 to 2013. The mean, long-term sensitivity of ozone to emissions is then calculated as the average of the "last" five years of this nine-year period. Since the adjoint integrates these sensitivities backwards in time, this corresponds to the sensitivity of the atmosphere to emissions occurring in the period 2004–2008 (inclusive).

The resulting adjoint sensitivities express the change in global mean column ozone to a change in emissions as a function of location, time and species. We focus on column ozone since previous scientific studies that quantified the role of (subsonic or supersonic) aviation emissions on ozone have expressed ozone responses expressed in terms of ozone column (Cunnold et al., 1977; Eastham and Barrett, 2016; Kinnison et al., 2020; Speth et al., 2021; Cunnold et al., 1977; Eastham and Barrett, 2016; Kinnison et al., 2020; Speth et al., 2021). Additionally, changes in column ozone directly affects surface UV exposure and a column ozone-neutral altitude directly translates to a surface UV flux-neutral altitude. To evaluate sensitivities to aviation fuel burn, we scale the sensitivities with uniform emission indices, representative of

the aviation industry. We use a constant CO emission index of 2 g/kg_{fuel}, representative of cruise conditions. Similarly, we keep the black carbon and organic carbon emission indices fixed to 20 mg/kg_{fuel}. We assume a water vapor emission index of 1.23 kg/kg_{fuel}. Aviation fuel naturally contains sulfur and, for this paper, we use, unless otherwise specified, a baseline fuel sulfur content of 600 parts per million (ppm) on an elemental sulfur mass basis. However, we note that fuel sulfur is subject to variations. Unless otherwise specified, we keep the nitrogen emission index at 15 g(NO_2)/kg_{fuel}, representative of subsonic aviation operations, based on estimates from the global aviation inventory from the Federal Aviation Administration (FAA) Aviation Environmental Design Tool (AEDT) for 2015.

In this paper, we focus primarily on the impact of nitrogen oxides, sulfur emissions on ozone, while also including contributions from water vapor, carbon monoxide, and black and organic carbon. Carbon and methane atmospheric cycles have previously been shown to be significant to both subsonic and supersonic aviation emissions impacts, moderating the tropospheric ozone response (Khodayari et al., 2015; Speth et al., 2021). However, for this work we use prescribed surface methane mixing ratios, and thus do not consider this feedback. An additional caveat is that, although the GEOS-Chem UCX adjoint includes photolysis calculations, it does not currently allow sensitivities to propagate through changes in photolysis rates. This means that feedbacks such as the ozone "self-healing" effect in response to an increase in NO_x beyond baseline levels will not be captured in the calculated sensitivity of stratospheric ozone to NO_x emissions.

We estimate the column ozone-neutral altitude as the location where the sum of the adjoint sensitivities, weighted by the fleet-wide emission indices, reaches zero:

$$\Sigma \frac{\partial J}{\partial c_i}(z_{neutral}) \times EI_i = 0$$

The share of each aircraft emission component to the total environmental impact of aviation emissions is computed as

$$|\frac{\partial J}{\partial c_i}| \times EI_i \Big/ \Sigma(|\frac{\partial J}{\partial c_i}| \times EI_i)$$

where $\frac{\partial J}{\partial c_i}$ represents the sensitivities of column ozone to a change in concentration in species c_i and EI_i represents the emission index of species i in g/kg_{fuel}. To estimate the component-wise share, we use the absolute value of the adjoint sensitivities to prevent the denominator from reaching zero. For instance, this means that an overall net zero contribution, consisting of e.g. −10 mDU from sulfur and +10 mDU from NO_x, will yield a 50% relative contribution from both species.

We evaluate the contribution of fuel sulfur to the environmental impact of aviation emissions as:

$$(|\frac{\partial J}{\partial c_{SO_2}}| \times EI_{SO_2} + |\frac{\partial J}{\partial c_{H_2SO_4}}| \times EI_{H_2SO_4}) \Big/ \Sigma(|\frac{\partial J}{\partial c_i}| \times EI_i). ,$$

The plume-scale conversions are derived using an aircraft plume chemistry model (Fritz et al., 2020). We compute the remaining fraction of NO_x after 24 h, as well as the amount of produced nitrogen reservoir species as a function of the NO_x and sulfur emission indices. The conversion between SO_2 and sulfate aerosols is also estimated from the same simulations. The meteorological conditions are kept constant throughout all of the plume model simulations.

We then estimate the column ozone-neutral altitude, but this time, accounting for plume-scale changes in NO_x, HNO_2, HNO_3, HNO_4, N_2O_5 and peroxyacetyl nitrate (PAN), and SO_2 and H_2SO_4.

3. Results

Unless otherwise stated, we assume a fuel sulfur content of 600 ppm and fleet average NO_x emission indices of 15 g NO_x per kg of fuel, consistent with current estimates for the subsonic aircraft fleet (Lee et al., 2020). Averaging globally, we show that aircraft emissions are column ozone-neutral in the 12–15 km altitude range (Fig. 1A). Below this altitude, aviation emissions lead to a net increase in column ozone, with the highest sensitivities in the less polluted Southern Hemisphere of up to 6.0 mDU/(Tg_{fuel}/year). At higher altitudes, aircraft engine emissions instead lead to increasing ozone depletion, reaching −120 mDU/(Tg_{fuel}/year) at 20 km.

Our results show seasonal variations in the sensitivities of total ozone column to aviation emissions. Fig. 1A shows the sensitivity of global mean column ozone to aircraft emissions occurring between 30°N and 60°N, a latitude band which includes 67% of year-2005 aircraft emissions (variations in sensitivity by longitude are negligible) (Simone et al., 2013). At 11 km, the sensitivity of column ozone to emissions varies by season, being maximized in April and October and minimized during January and July. At higher altitudes, these sensitivities increase in magnitude and reverse in sign. We also find that local sensitivities are instead minimized in April and maximized in October. For instance, emissions from an aircraft cruising in the Northern Hemisphere at 15 km in April will lead to double the ozone depletion reaching −20 mDU/(Tg_{fuel}/year), compared to a similar flight in October which will cause −10 mDU/(Tg_{fuel}/year). If the flight instead occurs at 20 km, the seasonal pattern remains the same but the sensitivities are a factor of nine greater.

The response of global mean ozone to emissions also varies outside of this latitude band. Fig. 1B shows zonally-averaged adjoint sensitivities for the same period. Aircraft emissions in the less polluted Southern Hemisphere between 8 and 12 km altitude lead to twice the amount of ozone produced compared to the same emissions in the Northern hemisphere. The increased sensitivity in the Southern hemisphere is likely due to the lower background concentrations of NO_x and volatile organic compounds (VOCs). The ozone production efficiency at upper tropospheric altitudes is negatively correlated with background NO_x concentrations, implying that emissions of aircraft NO_x into the cleaner Southern hemisphere will result in more ozone production (Holmes et al., 2011). Above 15 km, this hemispheric asymmetry is no longer present. We instead identify a difference between impacts resulting from emissions in the tropics (30°S - 30°N) and those at higher latitudes. For example, at 22 km altitude, the net ozone depletion per unit fuel burn in the tropics is −250 mDU/(Tg_{fuel}/year) compared to an average of −150 mDU/(Tg_{fuel}/year) at higher latitudes. This suggests that high Mach number supersonic aviation, which cruises at higher altitude, will cause greater ozone depletion if flights take place in the tropics, but that the opposite may be true for subsonic or low Mach number supersonic flights.

Panels A and B of Fig. 1 show that sensitivities are in the range of −2 to +6 mDU/(Tg_{fuel}/year) for altitudes between 8 and 14 km, compared to −2 to −300 mDU/(Tg_{fuel}/year) in the 14–25 km altitude band. The larger magnitude at higher altitudes is consistent with the greater role played by chemical catalytic cycles in the stratosphere. In the 14–20 km altitude range, the sensitivities double every 2 km, increasing from −15 to −125 mDU/(Tg_{fuel}/year). Between 20 and 25 km, the magnitude of the sensitivity increases linearly, with a change in magnitude of 35 mDU/(Tg_{fuel}/year) for every additional km.

Fig. 1C shows the sensitivity of global column ozone to each chemical component of aviation emissions. The results are re-interpreted in Fig. 1D as the percentage share contributed by each emission component to the total aviation emissions-induced response. As described in Section 2, the percentage share is here defined in absolute terms, such that two individual contributions of −10 and + 10 mDU, would each contribute 50% of the aggregated response. We find that NO_x emissions below 15 km produce a net increase in ozone, but produce a net decrease in ozone above this point. They are also responsible for 80% of all ozone changes for cruise altitudes below 12 km or above 20 km.

From Fig. 1, we find that the ozone-neutral cruise altitude varies with latitude. At Northern high and mid-latitudes, the neutral altitude is approximately constant at 13 km. This behavior differs from the tropics

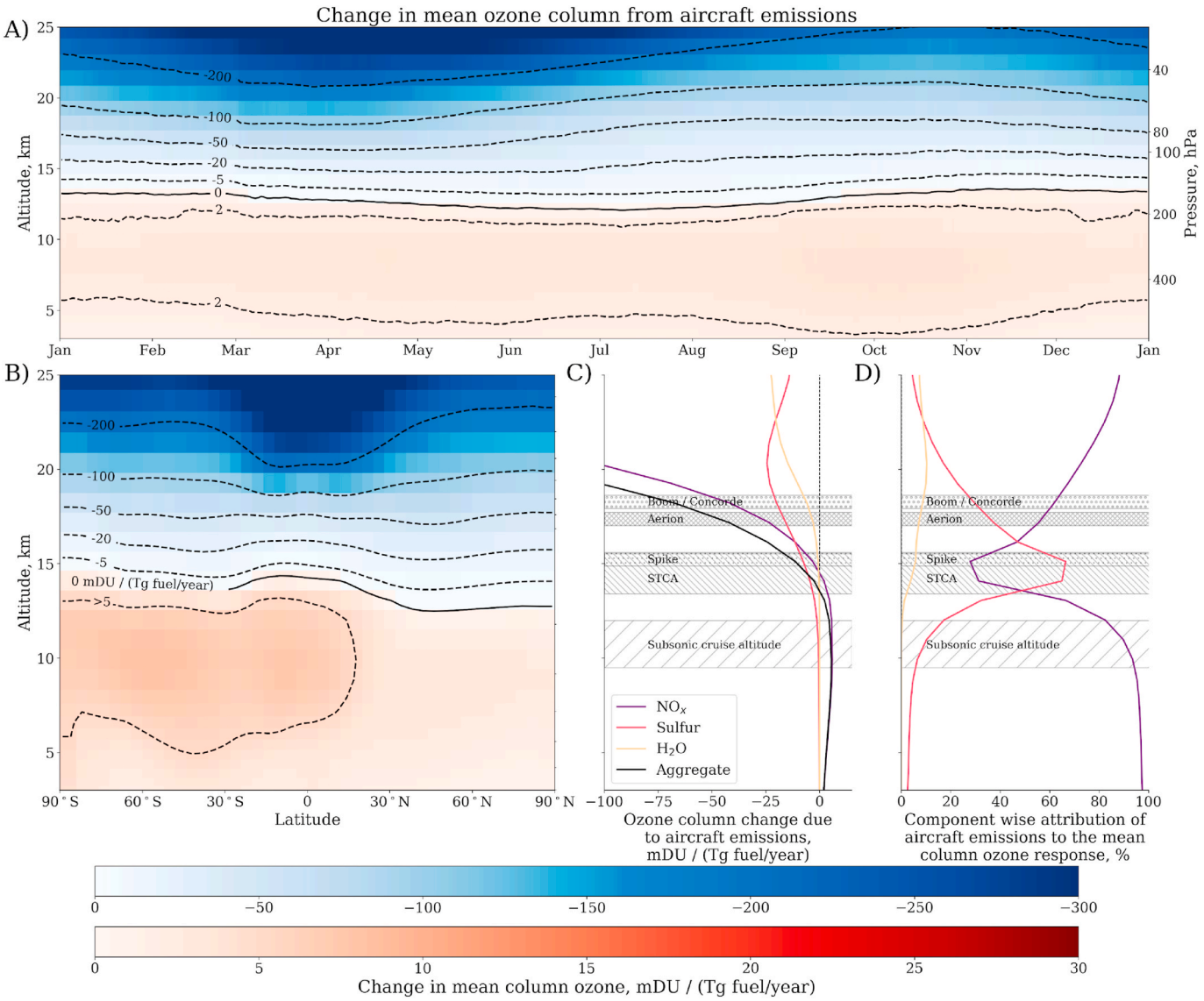

Fig. 1. Sensitivity of nine-year average ozone column to aviation fuel burn calculated with the GEOS Chem UCX adjoint model. The displayed sensitivities are averaged over the last five years of a nine-year run, allowing for four years of spin-up. The contour lines correspond to isolines of the sensitivity of column ozone to aviation fuel burn, which is also represented with color shading. The column ozone-neutral altitude is displayed as a continuous line. Other isolines are displayed with dashes. Panel A: Annually-averaged sensitivity weighted by aircraft emission indices in the 30°N-60°N latitude band. Panel B: Zonally-averaged sensitivity weighted by aircraft emission indices. Panel C: Contribution of each emission component to the total emission-weighted sensitivity of mean column ozone. Different cruising altitudes of subsonic and supersonic aircraft designs are displayed. Panel D: Component wise attribution of aircraft emissions expressed as a percentage. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

where the ozone-neutral altitude varies between 13.0 km and 14.3 km, at approximately 30°N and 10°S, respectively. In the Southern Hemisphere, the average neutral altitude is 14 km, with a global maximum at the South Pole at 14.6 km. The South Pole maximum and higher Southern Hemisphere neutral altitude are linked to positive time-averaged sensitivities of column ozone to NO_x emissions in the 12–16 km range over Antarctica. NO_x emissions over the South Pole convert active chlorine into chlorine reservoir species, thus lowering the potential stratospheric ozone depletion during austral spring. We show that the 30°N-60°N averaged sensitivities peak in October in the troposphere reaching 4 mDU/(Tg_{fuel}/year) at an altitude of 8 km, consistent with previous studies (Gilmore et al., 2013). In this latitude band, the ozone-neutral altitude varies between 12.0 and 13.6 km, for July and November, respectively. For comparison, typical subsonic aircraft cruise at altitudes of 10–12 km (Barrett et al., 2010).

Fig. 2 shows how the globally-averaged ozone-neutral cruise altitude varies as a function of both NO_x emission index and fuel sulfur content. Fig. 2 displays the average ozone-neutral cruise altitude globally (Panel A) and in the 30°N-60°N latitude band (Panel B). We identify that the column ozone-neutral altitude ranges between 10.7 and 14.5 km, and 9.7 and 13.7 when averaged globally, and in the 30°N-60°N latitude band respectively. These altitude ranges account for plume-scale processes.

4. Discussion

These results enable us to identify the ozone neutral cruise altitude not only for the current aircraft fleet as a whole, but also how this altitude varies as a function of the location and exhaust composition of a specific flight. We find that increases in NO_x emissions result in a higher ozone-neutral altitude. This is explained by the fact that the ozone-neutral altitude is located below the altitude at which the sensitivity

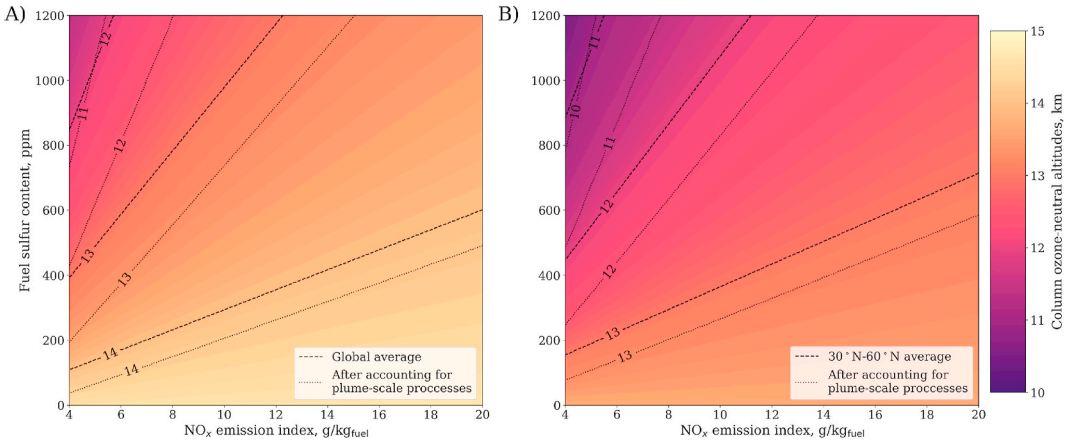

Fig. 2. Column ozone-neutral altitudes as a function of the fleet-wide NO_x emission index and fuel sulfur content. The black dashed lines in Panel A represent contour levels of the globally-averaged column ozone-neutral altitude, while, for Panel B, the averaging is performed in the 30°N-60°N, where most of aviation's fuel is burned. For both panels, the dotted lines represent the same isolines after accounting for plume-scale processes.

to NO_x emissions reverses from positive to negative. Additionally, we show that an increase in sulfur emissions leads to a reduction in the neutral cruise altitude with larger reductions at low fleet-wide NO_x engine emission indices.

Given the non-linear dependence of the sensitivities on cruising altitude, achieving ozone neutrality for an entire aircraft fleet requires care. Indeed, assuming a spread in the cruising altitude centered on the ozone neutral altitude is not sufficient, as an aircraft flying (e.g. 500 m) above the column ozone-neutral altitude leads to a greater magnitude change in ozone compared to that of a similar flight flying (e.g. 500 m) below this neutral altitude.

At all altitudes, sulfur emissions lead to net ozone depletion, but this depletion is greater at altitudes above 12 km. We find that sulfur emissions cause the largest net ozone impact of any component when emitted in the 13–16 km altitude band, where NO_x emissions impacts are transitioning from net ozone production to net ozone depletion. Above 16 km, NO_x emissions again become the largest contributor to net ozone change. HO_x-driven ozone depletion through water vapor emissions also becomes significant in this region, with its contribution to ozone changes reaching 10% at 20 km, consistent with previous reports on the effects of high-altitude emissions (Grewe et al., 2007).

While aviation-induced ozone impacts affect spatial scales that span thousands of kilometers (Barrett et al., 2010), processes occurring in aircraft plumes - which are defined by spatial scales well below those resolved by global models - have been found to significantly affect the long-term ozone production from aviation emissions (Cameron et al., 2013; Fritz et al., 2020; Paoli et al., 2011). As mentioned in Section 2, we account for these effects using a plume-scale emissions model. If these plume scale processes are neglected, a significant positive bias is introduced into the estimation of the ozone neutral cruise altitude. We find that this would result in the ozone neutral cruise altitude being overestimated by between 0.3 and 1.0 km, at NO_x emission indices of 15 and 5 g/kg_{fuel} respectively. This bias in the neutral altitude is similar in magnitude for both the global average and the 30°N-60°N average.

Based on the results presented in Fig. 2, we identify that – considering the global average – flights between 13.4 and 13.6 km altitude are column ozone-neutral assuming the baseline exhaust composition described in Section 2. Specifically, the global net change in column ozone is less than 1.7×10^{-4}%/(Tg_{fuel}/year). In comparison, previous estimates have found that subsonic aviation increases column ozone by 3.5×10^{-3}%/(Tg_{fuel}/year) (Weisenstein et al., 1998). If we consider only emissions in the 30°N-60°N latitude band, the neutral altitude is approximately 1 km lower than the global average.

If zero- or near-zero-sulfur fuel is considered, the global mean ozone neutral cruise altitude increases from 13.4 – 13.6 km to 14.0–14.5 km. Once sulfur is removed, the ozone neutral altitude becomes nearly independent of the NO_x emission index. However, if fuel sulfur is still present, then reducing the NO_x emission index instead decreases the ozone neutral cruise altitude. For current fuel sulfur contents, reduction in the fleet-wide NO_x emission index from 15 $g(NO_x)/kg_{fuel}$ to 6 $g(NO_x)/kg_{fuel}$ would reduce the ozone neutral cruise altitude by ~0.5 km. Water vapor emissions do not significantly affect the column ozone-neutral altitude, as their contributions only exceed 5% of the aggregated response above 15 km.

Current fuel sulfur content is subject to significant variability and uncertainty. Assuming that the average fuel sulfur content has an uncertainty range of 300–800 ppm, we estimate the globally-averaged neutral altitude range under uncertainty to be 13.3 and 14.1 km. In the 30°N-60°N latitude band, this uncertainty range becomes 12.4–13.2 km, with a mean value of 12.8 km.

Finally, future changes in stratospheric chlorine and sulfur loading are expected to alter the magnitude of the ozone response to high-altitude emissions (Brasseur and Granier, 1992; Weisenstein et al., 1998). In this study, we do not consider the role of future stratospheric chlorine and sulfur concentrations on the column ozone-neutral altitude. Other potential sources of disagreement are the ozone "self-healing" effect, long-term methane feedbacks, and the potential interaction of supersonic aircraft emissions with ice clouds, especially in the context of contrails and polar stratospheric clouds. Such interactions should be considered in future work.

5. Conclusion

The majority of existing subsonic, medium-to-long-haul commercial airliners are designed to cruise at altitudes of 10–12 km, balancing competing factors. For supersonic aircraft higher cruise altitudes are needed, increasing with Mach number (speed) - the Mach 2 Concorde cruised at an altitude of around 18 km. Our results suggest that low Mach number supersonic aircraft may be able to achieve ozone-neutral cruise around 14 km, with the exact altitude depending on the fuel sulfur content and/or the NO_x emission index of the aircraft engines. Given the spread in the cruising altitude of an entire fleet, the fleet-averaged cruise altitude would be lower than the column ozone-neutral altitude.

NO_x emission indices have been slowly increasing over the past decade or so, due in part to rising engine overall pressure ratios and turbine inlet temperatures (Lee et al., 2020). Assuming no reduction in fuel sulfur content, we find that further increases in the NO_x emission index will increase the magnitude of aviation's impacts on column ozone and increase the altitude at which aviation would become ozone neutral. However, if efforts to design ultra-low-NO_x engines succeed, ozone

neutral cruise may become possible at altitudes as low as 12 km.

However, this neglects the role of sulfur. Current trends in aviation are towards ultra-low sulfur fuel, either through desulfurization of conventional jet fuel or through naturally zero-sulfur options such as biofuels. We show that this will increase the net ozone production associated with aviation at any altitude, effectively increasing the altitude at which ozone-neutral cruise can be achieved by up to 1.0 km. Desulfurization will also leave NO_x as the only significant contributor to aviation's ozone impacts, as the contribution of water vapor emissions to column ozone is only significant above 15 km. This means that while future changes in the NO_x emissions index would modify the magnitude of aviation's impacts on ozone, it would cause no significant change in the ozone-neutral cruise altitude.

In 2015, commercial aircraft burned around 280 Tg of jet fuel, around 90% during cruise (Lee et al., 2020). If supersonic aircraft are successfully reintroduced to the fleet, it is expected that – compared to modern subsonic aircraft - they will require more fuel to be burned at higher altitudes to achieve the same flight distance. Our results provide the information needed to include ozone depletion when evaluating the costs and benefits of new aircraft designs, whether subsonic or supersonic, and how these impacts will be modified by future changes in fuel composition or engine emissions factors.

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CRediT authorship contribution statement

Thibaud M. Fritz: Formal analysis, Methodology, Software, Validation, Visualization, Writing – original draft, Writing – review & editing, Investigation, Data curation. **Irene C. Dedoussi:** Methodology, Software, Writing – review & editing. **Sebastian D. Eastham:** Methodology, Validation, Writing – review & editing, Supervision, Conceptualization, Project administration. **Raymond L. Speth:** Writing – review & editing, Supervision, Conceptualization, Funding acquisition, Project administration. **Daven K. Henze:** Methodology, Software, Writing – review & editing. **Steven R.H. Barrett:** Resources, Project administration, Supervision, Funding acquisition, Conceptualization, Writing – review & editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2022.119057.

Abbreviations

CTM	Chemistry Transport Model
UCX	Unified Chemistry eXtension

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