Temporal and spatial variability in the aviation NO$_x$-related O$_3$ impact

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Temporal and spatial variability in the aviation NO\textsubscript{x}-related O\textsubscript{3} impact

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Abstract

Aviation NO\textsubscript{x} emissions promote tropospheric ozone formation, which is linked to climate warming and adverse health effects. Modeling studies have quantified the relative impact of aviation NO\textsubscript{x} on O\textsubscript{3} in large geographic regions. As these studies have applied forward modeling techniques, it has not been possible to attribute O\textsubscript{3} formation to individual flights. Here we apply the adjoint of the global chemistry–transport model GEOS-Chem to assess the temporal and spatial variability in O\textsubscript{3} production due to aviation NO\textsubscript{x} emissions, which is the first application of an adjoint to this problem. We find that total aviation NO\textsubscript{x} emitted in October causes 40\% more O\textsubscript{3} than in April and that Pacific aviation emissions could cause 4–5 times more tropospheric O\textsubscript{3} per unit NO\textsubscript{x} than European or North American emissions. Using this sensitivity approach, the O\textsubscript{3} burden attributable to 83\,000 unique scheduled civil flights is computed individually. We find that the ten highest total O\textsubscript{3}-producing flights have origins or destinations in New Zealand or Australia. The top ranked O\textsubscript{3}-producing flights normalized by fuel burn cause 157 times more normalized O\textsubscript{3} formation than the bottom ranked ones. These results show significant spatial and temporal heterogeneity in environmental impacts of aviation NO\textsubscript{x} emissions.

Keywords: aviation, ozone, adjoint

1. Introduction

As the demand for aviation continues to increase, a more complete understanding of the impact of aircraft emissions on the environment is required to make informed aviation policy, design and operational decisions. Aircraft emissions have two primary environmental impacts. First, emissions in the lower as well as upper troposphere increase the concentration of ground-level particulate matter and ozone (Barrett et al 2010), which can lead to detrimental human health impacts (Pope 2002, Laden et al 2006). Second, aircraft emissions have a range of climate impacts, as has been reviewed by Lee et al (2010). This has motivated the development of policy, technology and operational initiatives to reduce the environmental impacts of aviation (FAA 2011, Mahashabde et al 2011).

In particular, the atmospheric impact of aviation NO\textsubscript{x} emissions has been a focus of previous atmospheric chemistry–transport modeling studies (Lee et al 2010). The introduction of additional NO\textsubscript{x}, especially at high altitudes, leads directly to the formation of ozone (O\textsubscript{3}), one of the naturally occurring greenhouse gases in the atmosphere. The direct ozone radiative forcing (RF) of aviation is approximately equal to (within ∼6\%) the CO\textsubscript{2} RF of aviation in 2005. We note, however, that the latter RF is due to the whole history of aviation emissions given the long atmospheric lifetime of CO\textsubscript{2}, while the direct O\textsubscript{3} RF is only a function of the past ∼2 months of emissions (Stevenson and Derwent 2009). Aircraft NO\textsubscript{x} emissions also increase the oxidative capacity of the atmosphere due to an increase
in the OH production rate (Kohler 2010). Increasing OH concentrations lead to a decrease in methane lifetime, which subsequently results in a decadal loss in O$_3$, Lee et al (2010) estimates the total RF due to aircraft NO$_x$ emissions at +0.0138 W m$^{-2}$, where approximately half of the direct (i.e. short-term) ozone RF of +0.0263 W m$^{-2}$ is offset by longer time scale impacts. This letter examines direct O$_3$ production due to aviation NO$_x$—which is potentially amenable to operational or routing mitigation measures—but we note that the longer-term effects are also important and there are significant uncertainties in these (Holmes et al 2011).

Aircraft NO$_x$ emissions, as compared to other anthropogenic sources, are important because the production efficiency of ozone at cruise altitudes (9–12 km) is higher than the production efficiency at ground level (Stevenson et al 2004, Kohler et al 2008), and the RF efficiency (forcing per molecule) at cruise altitudes is relatively high (Naik et al 2005). Despite the reasonably well understood chemistry of aviation-attributable O$_3$ decreasing the ozone impact of aviation is complicated by the relatively short ozone lifetimes and the dependency of ozone production and destruction on the local chemical state of the atmosphere and local transport characteristics. Thus, decreasing NO$_x$ emissions in one particular region may not be as effective in reducing aviation’s climate impact as decreasing emissions elsewhere, making the optimal mitigation strategy unclear.

Previous (perturbation) studies have focused on the differences in regional impacts of ground-level anthropogenic NO$_x$ emissions, with Naik et al (2005) concluding that anthropogenic NO$_x$ reductions in Southeast Asia have the largest impact on total and upper-tropospheric ozone concentrations. Fry et al (2012) obtained similar results, but also concluded that reductions in NO$_x$ emissions create atmospheric warming due to the recovery of atmospheric methane given reductions in OH concentrations. Aircraft NO$_x$ focused studies, such as those by Stevenson et al (2004) and Kohler et al (2008), were bulk region perturbation studies. Stevenson et al (2004) examined the size and duration of the O$_3$ and OH perturbation given aircraft NO$_x$ emissions at different times of year, while Kohler et al (2008) perturbed aircraft NO$_x$ emissions within several altitude bins and observed their relative impacts. A more recent perturbation study by Kohler et al (2012) showed that forcing attributable to short-term O$_3$ peaks near the equator when compared to higher latitudes and also determined that the net forcing due to NO$_x$ emissions is positive (i.e. warming).

These perturbation studies were based on taking the difference of forward atmospheric chemistry–transport simulations. Such approaches have the benefit of both being relatively straightforward to implement and show the spatially varying impact of emissions, but only work for perturbations of sufficient size to avoid subtractive errors. A perturbation approach also requires a simulation for every location and time of interest, making the assessment of more than a few locations or times computationally intractable. Here we apply an adjoint sensitivity approach, which results in the sensitivity of an objective function (tropospheric O$_3$) to NO$_x$ emissions at all locations and times from one simulation. The only previous application of an adjoint sensitivity approach to aviation NO$_x$–O$_3$ is given in table 1 of the supporting information of Bowman and Henze (2012), who applied adjoint modeling to determine the contribution of several sectors to the total O$_3$ instantaneous radiative effect for August 2006; such an approach has not been used in the current context before.

The tropospheric O$_3$ impact of individual flights has not previously been computed. While operational strategies to mitigate contrails from aviation have previously been investigated, this has not been possible for aviation-attributable O$_3$ as this requires quantification of impacts at the resolution of a single flight. The aim of this letter is therefore to quantify the O$_3$ impact of individual civil aviation flights and to quantify the spatial and temporal variability in the impact of aviation NO$_x$ emissions. We also assess the primary chemical production and loss pathways relevant to O$_3$ and compare aggregate results to previous work. This represents new understanding of the variability in the atmospheric impacts of flights and a step towards estimating the O$_3$-related benefits of flight-level operational optimization.

2. Methodology

This section describes the modeling techniques applied to determine the spatial and temporal heterogeneity in the aviation-attributable O$_3$ and the methodology for computing the per flight ozone impacts.

2.1. GEOS-Chem and the GEOS-Chem adjoint

GEOS-Chem was used in this analysis (Bey et al 2001). GEOS-Chem is a global chemistry–transport model that includes transport, wet and dry deposition, and gas and aerosol phase chemistry. Gas phase chemistry is solved using the kinetic pre-processor tool (KPP) developed by Damian et al (2002). All simulations use a 4° × 5° horizontal grid with 47 vertical layers, which includes pressure levels up to 0.010 hPa. GEOS-Chem is primarily a tropospheric model (i.e. a full chemistry simulation is performed up to the tropopause) where linearized ozone chemistry is used within the stratosphere. Each simulation in this study is for 16 months, where the first four months are used as spin-up.

The GEOS-Chem adjoint model was developed by Henze et al (2007). The model was first used for aviation in particular to determine the impact of aircraft emissions on surface air quality by Koo et al (2013). Adjoint models, in general, are applied in sensitivity analysis when the (typically single) output quantity of interest is set and the sensitivity of that output to (many) model input parameters is desired. The adjoint approach avoids an ensemble of forward model runs in calculating sensitivities, where only one ‘backwards’ integrating simulation is required for each output of interest. This is of particular use in studying the impact of aviation given that we are generally interested in total global or regional (e.g. North America, Asia) impacts, where every emissions grid cell is an input. The fundamentals of this approach are described in Errico (1997) for the application of adjoint models in meteorological study.
Anthropogenic emissions inputs for GEOS-Chem are detailed in van Donkelaar et al. (2008). Aircraft emissions are derived from the methodology presented in Barrett et al. (2010) and use the 2005 civil aviation emissions inventory by Simone et al. (2013). NO\textsubscript{e} emissions are given on an NO\textsubscript{2} mass basis where emissions are partitioned by mole fraction. Cruise emissions are partitioned as 90% NO, 9% NO\textsubscript{2}, and 1% HONO (Barrett et al. 2010). Landing/take-off (LTO) emissions are partitioned as 76% NO, 23% NO\textsubscript{2}, and 1% HONO (Barrett et al. 2010). Full-flight emissions (i.e. LTO and cruise emissions) are used in this analysis, where total aircraft NO\textsubscript{e} emissions amount to 2.66 Tg(NO\textsubscript{2}), annually. We also note that chemistry within aircraft exhaust plumes is neglected, where emissions are assumed to be instantaneously diluted into the local grid cell.

2.2. Sensitivities to aircraft emissions

The quantity of interest is the annually averaged O\textsubscript{3} perturbation due to aircraft NO\textsubscript{e} emissions, which is given by

\[
\delta O_3 = \frac{1}{T} \sum_{i=1}^{N_{LAT}} \sum_{j=1}^{N_{LON}} \sum_{k=1}^{N_{TROP}} \sum_{t=1}^{T} \left[ \frac{\partial J}{\partial E_{NOe}(i,j,k,t)} \right] \times E_{NOe}(i,j,k,t),
\]

(2.1)

where \(E_{NOe}\) is the time-varying three-dimensional emissions matrix, \(T\) is the number of time steps within the (one year) period over which O\textsubscript{3} impacts are averaged, \(N_{LAT}\) is the number of latitude grid cells, \(N_{LON}\) is the number of longitude grid cells, \(N_{TROP}\) is the number of troposphere altitude layers, and \(\partial J/\partial E_{NOe}\) is the adjoint sensitivity with objective function

\[
J = \sum_{i=1}^{N_{LAT}} \sum_{j=1}^{N_{LON}} \sum_{k=1}^{N_{TROP}} \sum_{t=1}^{T} M_{O_3}(i,j,k,t),
\]

(2.2)

with \(M_{O_3}\) being the mass of O\textsubscript{3} in a model grid cell. The adjoint sensitivity \(\partial J/\partial E_{NOe}\) — a four-dimensional matrix — is therefore the total subsequent change in tropospheric O\textsubscript{3} burden given 1 kg of NO\textsubscript{e} emitted at a location \(i, j, k\) and time \(t\).

Applying first-order sensitivities in estimating impacts assumes a linear relationship between the specie of interest and aircraft emissions at the level of the perturbation being considered. This assumption has been shown to be valid for the O\textsubscript{3} response to total aircraft NO\textsubscript{e} for a range of emissions perturbations in magnitude and altitude (Kohler et al. 2008). We infer that the linear approximation is at least as valid for individual flights as for all aviation NO\textsubscript{e}. Diurnal variations in sensitivities are not considered in this analysis as the daily variations in ozone production from aircraft NO\textsubscript{e} emissions was <2% of the seasonal variation based on a spectral (FFT) analysis of aircraft NO\textsubscript{e} emissions-weighted sensitivity data. This is not necessarily true of ground-level sensitivities.

2.3. Chemical pathways analysis

In order to determine the primary drivers behind the temporal as well as spatial patterns of the calculated ozone sensitivities, a simplified analysis of the underlying chemistry in the forward simulations is performed. The time rate of change for any given specie within a chemical kinetic system is the difference between the total production and loss rates. Each production and loss rate in turn is a function of the product of the rate coefficient and the relevant species concentrations (Seinfeld and Pandis 2006). Solving chemical kinetics represents a challenge given the stiffness of the system (i.e. orders of magnitude differences in reaction rate time scales), where implicit methods are used to avoid small time steps (Hairer and Wanner 2002). Here, a simplified approach is taken to quantify each pathway’s contribution. The total ozone rate of change is disaggregated into its most significant production and loss pathways (significant being ~5% of the total rate within the chemistry time step).

Once four-dimensional sensitivity data have been calculated, the first-order atmospheric response of O\textsubscript{3} concentrations to aircraft NO\textsubscript{e} emissions can be estimated. The aircraft emissions model by Simone et al. (2013) generates three-dimensional emissions matrices for each aircraft type flown between a pair of airports. Following the generation of the gridded emissions data for a particular scenario, the first-order response can then be determined by taking the inner product (i.e. component-wise multiplication and summation) between

Table 1. Pathways of O\textsubscript{3} production and loss considered in the chemical pathway analysis.

<table>
<thead>
<tr>
<th>Pathway</th>
<th>Reaction</th>
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<tbody>
<tr>
<td>(1)</td>
<td>O + O\textsubscript{2} + M \rightarrow O\textsubscript{3} + M</td>
</tr>
<tr>
<td>(2)</td>
<td>O\textsubscript{3} + NO \rightarrow NO\textsubscript{2} + O\textsubscript{2}</td>
</tr>
<tr>
<td>(3)</td>
<td>O\textsubscript{3} + OH \rightarrow HO\textsubscript{2} + O\textsubscript{2}</td>
</tr>
<tr>
<td>(4)</td>
<td>O\textsubscript{3} + HO\textsubscript{2} \rightarrow OH + 2O\textsubscript{2}</td>
</tr>
<tr>
<td>(5)</td>
<td>O\textsubscript{3} + NO\textsubscript{2} \rightarrow NO\textsubscript{2} + O\textsubscript{2}</td>
</tr>
<tr>
<td>(6)</td>
<td>O\textsubscript{3} + h\nu \rightarrow O\textsubscript{2} + O(\text{I}D)</td>
</tr>
<tr>
<td>(7)</td>
<td>Dry deposition</td>
</tr>
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</table>
the sensitivity (for a particular day of the year) and emissions matrices, as indicated in equation (2.1). This process, which is computationally inexpensive compared to a complete forward model simulation, allows for the rapid calculation of the atmospheric impacts associated with each flight.

3. Results

This section presents the results from the adjoint sensitivity simulations using the cost function definition in equation (2.2) and the chemical pathway analysis. A forward model analysis was performed with and without aircraft emissions to compare GEOS-Chem with previous aviation NO\textsubscript{x} studies. GEOS-Chem predicts an annually averaged O\textsubscript{3} perturbation of 0.8 DU as compared to 0.9 DU estimated by Kohler et al (2008). Using the GEOS-Chem adjoint-calculated ozone sensitivities multiplied by total aviation NO\textsubscript{x} emissions results in an annually averaged O\textsubscript{3} perturbation that is 10% lower than the forward model result, consistent with the weak nonlinearity found in previous studies.

3.1. Sensitivity of ozone burden to aircraft emissions

From the GEOS-Chem adjoint model, the sensitivity of total tropospheric O\textsubscript{3} to NO\textsubscript{x} emissions is calculated. Figure 1 shows the average effect of 1 kg of aircraft NO\textsubscript{x} emitted as a function of time of year on the annual average tropospheric O\textsubscript{3} burden in kg (black). This was computed by taking the aviation NO\textsubscript{x} emissions-weighted average of the sensitivity matrix for each hour of the year (then averaging over one year). The results show that aviation NO\textsubscript{x} emissions in October cause 40% more annually averaged tropospheric O\textsubscript{3} than emissions in April. Stevenson et al (2004) also showed a peak ozone perturbation given an aircraft NO\textsubscript{x} perturbation in October.

Figure 2 shows the sensitivity of annually averaged tropospheric O\textsubscript{3} to NO\textsubscript{x} emissions at cruise altitudes (9–12 km average), where the sensitivity has been averaged over a year. The peak sensitivity is over the Pacific at 4\textdegree S, 170\textdegree E (~1000 km northeast of the Solomon Islands). In this location an emission of 1 kg of aircraft NO\textsubscript{x} would result in a 15 kg increase in O\textsubscript{3} burden averaged over one year. This is 5.1 times higher than the sensitivity in Europe and 3.7 times higher than North America. This peak in sensitivity in the equatorial region is consistent with recent forward modeling results by Kohler et al (2012). It is also consistent with the results presented in Stevenson and Derwent (2009) which showed peak short-term O\textsubscript{3} integrated RF impacts in the remote Pacific region. High sensitivities were associated with NO\textsubscript{x}-scarce regions (e.g. remote Pacific), as was the case in this analysis. We note from figure 2 that the ‘center of mass’ of the sensitivity is south of the equator, indicative of the weighting between both NO\textsubscript{x} scarcity and solar zenith angle in ozone sensitivity.

The chemical pathway analysis is used to investigate these temporal and spatial variations. The rate of change of aviation-attributable ozone is approximated as

\[
\frac{d(\delta O_3)}{dr} \approx \frac{d(O_3)}{dr} \bigg|_{av} - \frac{d(O_3)}{dr} \bigg|_{w/o \ av},
\]

where av denotes the rate of change of O\textsubscript{3} with aviation, w/o av denotes the rate of change without aviation, and \(\delta O_3\) denotes the O\textsubscript{3} perturbation attributable to aircraft emissions.

The pathways in figure 3 account for 86% of total net rate of change of aviation-attributable O\textsubscript{3}, while all the pathways considered in the analysis overestimate net production by 3%. The ‘Production’ pathway is the net O\textsubscript{3} production resulting from the direct generation of O\textsubscript{3} by O + O\textsubscript{2} (where atomic oxygen is generated by way of NO\textsubscript{2} photolysis) minus the destruction due to ozone’s reaction with NO (which tracks well with the rate of the HO\textsubscript{2} + NO reaction).
Figure 3. Change in aviation-only O\textsubscript{3} production and loss pathways for (a) aviation emissions in October relative to April and (b) cruise altitude NO\textsubscript{x} emissions near the Solomon Islands relative to Europe.

Figure 3(a) shows the primary pathways that drive the temporal pattern in the averaged sensitivity when comparing pathway rates in October (peak sensitivity) to April (minimum sensitivity). We note that ‘April’ in figure 3 is an average across the first and last month’s sensitivities given that they are not exactly cyclical (figure 1). It can be seen that while overall production rates in October are higher, chemical and photolytic loss rates are also larger in magnitude, thus the higher production rate is offset by the higher loss rates, which is chemically dominated by ozone’s reaction with OH. Figure 3(a) shows that while lifetimes in April may be longer (i.e. loss rates are lower), this does not correspond to peak ozone sensitivity to aircraft NO\textsubscript{x} emissions. The low loss rates in the winter months, however, do correspond with the peak in the aviation-attributable O\textsubscript{3} perturbation.

Figure 3(b) shows the relative change in each pathway for the location of peak sensitivity in the Pacific near the Solomon Islands relative to the average for Europe. The spatial difference is driven by decreased total production (photolysis) as well as increased O\textsubscript{3} + HO\textsubscript{2} loss, decreased O\textsubscript{3} + OH loss, and increased photolytic loss. This follows from the NO\textsubscript{x}-scarce environment in the remote Pacific. Loss due to HO\textsubscript{2} is more prevalent in this region given that NO\textsubscript{x} is not present to cycle HO\textsubscript{2} to OH, resulting in a lower O\textsubscript{3}+OH loss rate. Given that the overall production rate is also lower (i.e. HO\textsubscript{2} + NO is not as significant), this suggests that O\textsubscript{3} production is NO limited, thus resulting in higher ozone sensitivities in this region. Note that the positive O\textsubscript{3} + HO\textsubscript{2} loss rate indicates a decrease in ozone loss by way of HO\textsubscript{2} when aviation emissions are introduced. In addition, pathway rates are higher in the case of figure 3(b) as only chemistry at cruise altitudes is being considered, where rates are in general higher for aviation-attributable ozone at this altitude range as opposed to production or loss rates in the lower troposphere.

As fully spatially resolved sensitivity data is generated (having been averaged in time), ground-level sensitivities are also compared to previous studies that investigated the impact of changing ground-level anthropogenic NO\textsubscript{x} emissions. Our results generally agree with the previous studies, indicating that sensitivities are highest in Southeast Asia while the overall temporal patterns show consistently high sensitivities in the boreal summer and early autumn months and minimum sensitivities in the winter. Naik et al (2005) estimated a factor of 9 difference between the magnitude of sensitivities for Southeast Asia relative to North America for global O\textsubscript{3} due to changes in regional NO\textsubscript{x} emissions, whereas a factor of ~2 is found here when calculating spatially averaged sensitivities. The magnitudes of the sensitivity values, however, approximately correspond to the sensitivity values calculated by Fry et al (2012) where ozone sensitivities are on the order of 1 kg O\textsubscript{3}/kg NO\textsubscript{x}.

3.2. Comparison to ozone production efficiency

The sensitivity results presented in figure 2 also provide an alternative measure to the more common ozone production efficiency (OPE). The concept of OPE was introduced by Liu et al (1987). In this analysis, however, we use the definition presented in Seinfeld and Pandis (2006) and define OPE (in a simplified atmosphere) as

\[
\text{OPE} = \frac{k_{\text{HO}_2 + \text{NO}}[\text{HO}_2][\text{NO}]}{k_{\text{NO}_2 + \text{OH}}[\text{NO}_2][\text{OH}]}.
\]

Equation (3.2) thus defines OPE to be the ratio of the time rate of O\textsubscript{3} production (approximated by the rate of the additional production of NO\textsubscript{2}) to the time rate of NO\textsubscript{3} removal (i.e. HNO\textsubscript{3} production). This definition does not capture the impact of O\textsubscript{3} removal pathways, but rather is a reflection of the likelihood of NO\textsubscript{3} to promote O\textsubscript{3} production rather than exit the ozone cycle through nitric acid production. In addition, OPE is an instantaneous measure of the production tendency of NO\textsubscript{3}, where longer-term atmospheric impacts are not captured. The adjoint sensitivity, however, represents the total expected net production of O\textsubscript{3} and its lifetime induced by a small increment in NO\textsubscript{x} concentration at a particular time and location in the atmosphere. Figure 1 shows OPE plotted (in blue) versus the previously calculated ozone sensitivity.

The respective peaks of OPE and O\textsubscript{3} sensitivity do not coincide. OPE peaks in the (boreal) summer months where photolysis of NO\textsubscript{2} is most active, preceding the sensitivity
peak by 2–3 months. When comparing OPE and the ozone sensitivities to the chemical pathway analysis, we see that the OPE tracks closely with the total O$_3$ sensitivity to the chemical pathway. When comparing OPE and the ozone contribution of each flight to the total ozone perturbation, we see that the peak ozone sensitivity to aircraft NO$_x$ emissions is by large aircraft flying short distances such as from Mauritius to Saint Denis (islands near Madagascar) rank highly using this normalization. In addition, flights, whether ranked by total impact or FOF, have origins or destinations in Australia or New Zealand. Within the top five, however, only one flight appears for both normalized (Auckland to Seoul). The highest O$_3$ impact flight on a fuel burn-normalized basis results in approximately 157 times more O$_3$ than the lowest impact flight. However, because fuel burn also varies by two orders of magnitude across flights, the highest impact flights result in four or five orders of magnitude greater O$_3$ production than the lowest impact flights.

Table 2 also shows that the flights with the highest total impact do not necessarily correlate to the flights with the highest impact on a fuel burn basis. Dividing by fuel burn roughly normalizes the flight by distance for a comparably sized aircraft. Thus, flights that remain within the Australia–Southeast Asia area result in large FOFs, while a flight such as Auckland to Los Angeles generates a lower FOF since a portion of the flight is spent outside this high sensitivity zone.

Table 3 shows the annually averaged O$_3$ impact for the top five flights normalized by seat-km. Results are dominated by large aircraft flying short distances such as from Mauritius to Saint Denis. Spatially, the majority of these flights occur in South America or Southeast Asia, although many of the flights from Mauritius to Saint Denis (islands near Madagascar) rank highly using this normalization.

We find that the top (500, 5000, 50 000) unique flights ranked by O$_3$ impact create (11.3, 47.7, 93.7)% of the total O$_3$ impact while constituting (5, 36, 91)% of the seat-km and fuel burn. In addition, all flights, whether ranked by total impact or FOF, have origins or destinations in Australia or New Zealand. Within the top five, however, only one flight appears for both normalizations (Auckland to Seoul). The highest O$_3$ impact flight on a fuel burn-normalized basis results in approximately 157 times more O$_3$ than the lowest impact flight. However, because fuel burn also varies by two orders of magnitude across flights, the highest impact flights result in four orders of magnitude greater O$_3$ production than the lowest impact flights.

Table 2. Ranking of top five flights by O$_3$ impact for two different metrics. The first section shows ranking by total impact, i.e. the contribution of each flight to the total ozone perturbation. The second section shows ranking by FOF, i.e. ozone impact normalized by fuel burn and scaled by the lowest impact flight. Return trips were not counted (only the higher of a return-trip pair are counted).

<table>
<thead>
<tr>
<th>Rank</th>
<th>Origin</th>
<th>Destination</th>
<th>Aircraft</th>
<th>Total O$_3$ (kg)</th>
<th>FOF</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Auckland, New Zealand</td>
<td>Seoul, South Korea</td>
<td>B777</td>
<td>22 900</td>
<td>157</td>
</tr>
<tr>
<td>2</td>
<td>Sydney, Australia</td>
<td>Seoul, South Korea</td>
<td>B777</td>
<td>18 200</td>
<td>146</td>
</tr>
<tr>
<td>3</td>
<td>Sydney, Australia</td>
<td>Bangkok, Thailand</td>
<td>B777</td>
<td>17 600</td>
<td>144</td>
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<tr>
<td>4</td>
<td>Brisbane, Australia</td>
<td>Bangkok, Thailand</td>
<td>B777</td>
<td>14 900</td>
<td>139</td>
</tr>
<tr>
<td>5</td>
<td>Christchurch, New Zealand</td>
<td>Seoul, South Korea</td>
<td>B777</td>
<td>19 700</td>
<td>139</td>
</tr>
</tbody>
</table>

Table 3. Total ozone impact normalized by seat-km.

<table>
<thead>
<tr>
<th>Rank</th>
<th>Origin</th>
<th>Destination</th>
<th>Aircraft</th>
<th>kg/st-km</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Mauritius</td>
<td>Saint Denis</td>
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<td>Montevideo</td>
<td>Buenos Aires</td>
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<td>Saint Denis</td>
<td>A340</td>
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<td>0.012</td>
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<td>Rio De Janeiro</td>
<td>Sao Paulo</td>
<td>B777</td>
<td>0.011</td>
</tr>
</tbody>
</table>
(0.6, 6, 60)% of the unique flights. Thus the first (500, 5000, 50,000) unique flights are (18.8, 7.8, 1.6) times as effective per seat-km at creating \( \text{O}_3 \) than the average flight per seat-km.

5. Discussion

Previously, perturbation studies have been used to investigate the impact of aviation emissions in different locations and times, particularly with respect to the impact of \( \text{NO}_x \) emissions on tropospheric ozone burdens. These studies have been largely restricted to bulk regions or particular injection times due to limitations inherent in forward sensitivity analyses in this context. In this study, an adjoint approach is used to determine the full four-dimensional sensitivity of tropospheric ozone to \( \text{NO}_x \) emissions using the GEOS-Chem adjoint model. Using this approach per flight impacts were determined.

While ozone production related to aviation peaks in the boreal summer, aviation \( \text{NO}_x \) emissions in the autumn are most effective at increasing the annual averaged tropospheric \( \text{O}_3 \) burden. This occurs at approximately the midpoint between the peak in ozone production and the peak in ozone lifetime associated with aviation \( \text{NO}_x \) emissions. On a spatial basis, cruise altitude \( \text{NO}_x \) emissions near the Solomon Islands would contribute 5.1 times more to the annual averaged tropospheric \( \text{O}_3 \) burden than emissions over Europe. More importantly, sensitivities in Southeast Asia—where aviation is growing significantly faster than the global average—are more than double those in North America and Europe. This implies that the marginal impact of aviation growth in future will be greater than the historical growth that has been concentrated in North America and Europe.

Our analysis showed that the highest total ozone perturbations were caused by individual flights with origins and destinations in Australia or New Zealand, which was also true if results were normalized by total fuel burn. Using this metric, the most impactful flight (of the 83,000 unique flights considered) creates 157 times more ozone per kg of fuel burned than the minimum. These flights also correspond to areas of high ozone sensitivity to \( \text{NO}_x \) emissions and flight paths largely contained within high sensitivity regions. We also find that a disproportionate fraction of the direct ozone impact from aviation can be attributed to a relatively small fraction of flights. The strength of the spatial and temporal variability in ozone production due to aviation \( \text{NO}_x \) emissions raises the possibility of time and location dependent mitigation measures, which may be facilitated by the four-dimensional sensitivity data created in this analysis. We note other atmospheric impacts associated with aircraft emissions also need to be considered and will be the subject of future work. As Stevenson and Derwent (2009) showed, the longer-term \( \text{CH}_4 \) climate response may outweigh the near-term \( \text{O}_3 \) perturbation due to aircraft emissions.

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