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# Trading Off Aircraft Fuel Burn and NO<sub>x</sub> Emissions for Optimal Climate Policy

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ABSTRACT. Aviation emits pollutants that affect climate, including CO<sub>2</sub> and NO<sub>x</sub>; NO<sub>x</sub> indirectly so, through the formation of tropospheric ozone and reduction of ambient methane. To improve the fuel performance of engines, combustor temperatures and pressures often increase, increasing NO<sub>x</sub> emissions. Conversely, combustor modifications to reduce NO<sub>x</sub> may increase CO<sub>2</sub>. Hence, a technology tradeoff exists, which also translates to a tradeoff between short lived climate forcers and a long-lived greenhouse gas, CO<sub>2</sub>. Moreover, the NO<sub>x</sub>-O<sub>3</sub>-CH<sub>4</sub> system responds in a non-linear manner, according to both aviation emissions and background NO<sub>x</sub>. A simple climate model was modified to incorporate non-linearities parameterized from a complex chemistry model. Case studies showed that for a scenario of a 20% reduction in NO<sub>x</sub> emissions the consequential CO<sub>2</sub> penalty of 2% actually increased the total radiative forcing (RF). For a 2% fuel penalty, NO<sub>x</sub> emissions needed to be reduced by >43% to realize an overall benefit.

Conversely, to ensure the fuel penalty for a 20% NO<sub>x</sub> emission reduction did not increase overall forcing, a 0.5% increase in CO<sub>2</sub> was found to be the ‘break even’ point. The timescales of the climate effects of NO<sub>x</sub> and CO<sub>2</sub> are quite different, necessitating careful analysis of proposed emissions tradeoffs.

## INTRODUCTION

Aviation is essential to international travel, and is a growing industry, with passenger traffic increasing at an average of 5.3% per year since 2000. It releases anthropogenic emissions in a physically and chemically complex region of the atmosphere. Aviation emissions consist primarily of carbon dioxide (CO<sub>2</sub>), nitrogen oxides (NO<sub>x</sub>), sulfur oxides (SO<sub>x</sub>) and soot or ‘black carbon’ emissions, and small amounts of water vapour<sup>1-3</sup>. The climate impacts of aviation NO<sub>x</sub> emissions are complex, since they affect the climate by contributing a positive radiative forcing (RF) through the promotion of tropospheric ozone formation and a negative RF by reducing methane lifetime. There are additional negative RF effects from the CH<sub>4</sub> lifetime reduction through small reductions in background O<sub>3</sub> and stratospheric water vapour<sup>4</sup>, although the balance is a net positive forcing<sup>2,5</sup>. At ground level, aviation NO<sub>x</sub> is also considered an air pollutant due to its role in ozone production.

In 1981, ICAO adopted a first certification standard to control aircraft NO<sub>x</sub> emissions in response to concerns over the effect of NO<sub>x</sub> emissions on surface air quality. As further NO<sub>x</sub> stringency assessments were undertaken it became apparent that the engine modifications necessary to reduce NO<sub>x</sub> resulted in a fuel burn penalty, and therefore a CO<sub>2</sub> penalty. Hence, it was realized that a tradeoff existed between the two pollutants<sup>6-8</sup>.

A further issue arises over the timescale of the perturbations to the atmosphere; aviation NO<sub>x</sub> emissions and their associated impacts on ozone and methane contribute a short-lived climate forcing to the atmosphere, whereas CO<sub>2</sub> release has an impact on a much longer timescale. In order to understand the environmental consequences of the technology tradeoff, it is necessary to model the climate impacts in some way for both NO<sub>x</sub> and CO<sub>2</sub> perturbations over longer timescales. Most studies of the radiative impact of aviation consider either present-day forcing, or a scenario of e.g 2050 emissions<sup>1, 9-11</sup>. Here, we focus on the very long term as this is not normally considered and only a few studies deal with this<sup>12-14</sup>. The long-term is important as it affects the choice of the mitigation options outlined here, i.e. the long-term impact of a small increase in CO<sub>2</sub> emissions that accumulate vs shorter-term effects that reduce forcing.

Adding to the complexity of this, the NO<sub>x</sub>-O<sub>3</sub>-CH<sub>4</sub> atmospheric system is known to be non-linear, sensitive to both the perturbing emissions being studied (i.e. aviation) and the NO<sub>x</sub> levels of the background atmosphere<sup>15-17</sup>. Such calculations are normally conducted with complex 3D models of the atmosphere that account for this with a sophisticated chemical scheme. The reduction in CH<sub>4</sub> lifetime, is normally calculated offline by a simplified parameterization since CH<sub>4</sub> has a lifetime of approximately 10 – 12 years. However, model simulations for periods of around 100 years are necessary to account for a significant fraction of the CO<sub>2</sub> emissions, usually done in simplified climate models (SCMs). Previously, small perturbations of the NO<sub>x</sub> system have been treated as linear<sup>12</sup> (e.g. Sausen and Schumann, 2000) in SCMs. Since this is known to induce inaccuracies into the computations, a new non-linear parameterization of a SCM was derived from a more complex atmospheric chemistry model, MOZART-3, to model the longer-term effects of aviation NO<sub>x</sub> emissions.

Having demonstrated and incorporated a suitable non-linear NO<sub>x</sub> scheme into a SCM, a series of model runs were designed in order to study the tradeoff between aviation NO<sub>x</sub> and CO<sub>2</sub> emissions over a 100 year period. Through changes in aircraft engine design and emissions characteristics, the relative emissions of NO<sub>x</sub> and CO<sub>2</sub> can be tuned to address specific mitigation targets. From the perspective of climate change mitigation, the model runs investigate the amount of NO<sub>x</sub> reduction needed to account for any increases in CO<sub>2</sub> emissions and also, how much additional CO<sub>2</sub> can be emitted before additional forcing is incurred, should NO<sub>x</sub> emissions be reduced by a set amount, in this case -20%.

The model runs also assess the impact of the background NO<sub>x</sub> emission on the sign of the NO<sub>x</sub> RF and how this impacts on a tradeoff scenario, therefore two different background NO<sub>x</sub> levels are investigated, one to represent a near present day atmospheric composition and one to represent a background atmosphere where significant surface NO<sub>x</sub> emissions reduction has taken place.

## METHODS

*Overall simulations design and modeling tools.* Comparing emissions and their climate effects in some form of emission equivalence is a complex subject itself<sup>18</sup>. However, in this study, the tradeoff question can be posed in a simple way in the sense of variation of RF and change in global mean surface temperature ( $\Delta T$ ) after 100 years for constant emissions conditions over some defined base case. First, the global CTM (chemistry transport model) MOZART was used to investigate the linearity of the NO<sub>x</sub>-O<sub>3</sub> and NO<sub>x</sub>-CH<sub>4</sub> relationships in response to different background conditions. The results of those model runs were then used to create a new non-linear NO<sub>x</sub> parameterization to be used in a tradeoff study.

87 The tradeoffs simulations performed with the SCM represent a parametric study, where all  
88 variables are kept constant over time, beginning with a constant amount of fuel use per year. This  
89 was to gauge the response of the system to a simple (constant) input, rather than being a scenario  
90 study of actual projections. The constant value of fuel use was ~250 Tg per year, the  
91 observational fleet value at 2012 (International Energy Agency data), background CO<sub>2</sub> was kept  
92 constant at 404 ppm, the background value as of March 2016, thus removing the transient nature  
93 of CO<sub>2</sub> modeling - in order to remain consistent with the constant NO<sub>x</sub> background used in the  
94 CTM runs outlined below. The global fleet emissions index for NO<sub>x</sub> (EINO<sub>x</sub> in g NO<sub>2</sub>/g fuel  
95 burned) was kept constant at 13, a representative fleet average. Aviation CO<sub>2</sub> and NO<sub>x</sub> emissions  
96 were fixed over an arbitrary 100 year simulation at ~790 Tg CO<sub>2</sub> (kerosene to CO<sub>2</sub> conversion of  
97 3.16) and 3.24 Tg NO<sub>2</sub> (0.98 Tg N) per year respectively as a result of the constant fuel use. This  
98 scenario represented the ‘base case’ where the total RF was taken to be the sum of the net NO<sub>x</sub>  
99 and CO<sub>2</sub> radiative forcings. Note that no ‘history’ of CO<sub>2</sub> emissions prior to the start year was  
100 incorporated. The base case was then perturbed, the constant fuel value was changed to reflect a  
101 percentage increase or decrease in CO<sub>2</sub> and NO<sub>x</sub> emissions, while still remaining constant over  
102 time. A common scenario from the literature suggested that a 2% fuel penalty could be incurred  
103 when NO<sub>x</sub> emissions were reduced by 20% - owing to engine modification<sup>7,19,20</sup> - to determine  
104 whether a net RF benefit was realized or not. The model runs then followed a logical path of  
105 determining how much NO<sub>x</sub> reduction is in fact necessary to counteract the additional 2% CO<sub>2</sub>  
106 emissions, i.e. ‘breaking even’, while ensuring that overall RF does not exceed that of the base  
107 case. It is then investigated, were the situation to be reversed and NO<sub>x</sub> reduction was held at -  
108 20% below the base case, how much of CO<sub>2</sub>/fuel penalty is allowed before forcing goes above

that of the base case. Sensitivity simulations were also run to understand the consequences of high and low NO<sub>x</sub> background emissions.

Two basic modeling tools were necessary – a sophisticated 3D CTM of the global atmosphere ('MOZART' v3) and a simple climate model (SCM)(LinClim). MOZART was used to fully represent the impacts of changing aircraft NO<sub>x</sub> emissions at varying levels and backgrounds<sup>21</sup>, the results of which were used to formulate a simplified parameterization in the LinClim SCM ('LinClim', based on Sausen and Schumann, 2000), which simulated both net NO<sub>x</sub> and CO<sub>2</sub> radiative impacts. These modeling tools are described below.

*Three-dimensional global chemical transport model – MOZART.* The 3D CTM MOZART (Model for OZone And Related Tracers), version 3, was used to simulate the ozone burden and methane lifetime change resulting from aviation NO<sub>x</sub> emissions in this study. MOZART-3 was evaluated by Kinnison et al (2007) and has been applied in several atmospheric studies<sup>22-25</sup>. The European Centre for Medium Range Weather Forecasts (ECMWF) ERA-Interim reanalysis data for 2006 provided the meteorological fields that drive the transport of chemicals within MOZART. The background emissions necessary for MOZART<sup>26</sup> represent the year 2000 and were originally compiled for the IPCC AR5 report. The background data are made up of surface emissions of anthropogenic activity and biomass burning, and the European Union project POET (Precursors of Ozone and their Effects on Troposphere) supply the biogenic surface emissions<sup>27</sup>. The choice of meteorology data driving the model will affect the calculations of the NO<sub>x</sub>/O<sub>3</sub>/CH<sub>4</sub> impacts. Kinnison et al., (2007), when evaluating MOZART3 model performance against observations of various chemical species, noted better agreement when similar ECMWF reanalysis data were used vs other dynamical data. MOZART3 was also driven with ECHAM/5 GCM data as a test, the results from which are given in the SI. Inter-model variability is another

132 source of uncertainty in CTM modeling, in Søvde et al., (2014) MOZART3 is tested against  
133 other models in its ability to model NO<sub>x</sub> emissions<sup>29</sup>, the results of that analysis are extended in  
134 the SI, to show the variability of aviation NO<sub>x</sub> responses in a small subset of CTMs and how  
135 MOZART compares to other models.

136 The aim of the CTM simulations was to model how the atmosphere reacts to the release of  
137 varying levels of aviation and background NO<sub>x</sub> emissions. Although it is known that aviation  
138 NO<sub>x</sub> increases tropospheric ozone burden and reduces methane lifetime, the question arises as to  
139 when this relationship becomes non-linear. The SCM LinClim previously incorporated a linear  
140 scheme for NO<sub>x</sub> - O<sub>3</sub> and NO<sub>x</sub> - CH<sub>4</sub>, such that the purpose of running iterative simulations with  
141 MOZART was to determine whether a new non-linear parameterization of LinClim could be  
142 formulated, and also determine the sensitivity of this non-linear response to different background  
143 NO<sub>x</sub> conditions.

144 For each simulation run on MOZART, the model was run without aircraft emissions, referred to  
145 as the ‘reference run’ and then again with aircraft emissions, referred to as the ‘perturbation run’.  
146 Each of these runs is preceded by a ‘spin up year’, which used the same meteorology, and  
147 describes the time taken by the model for the atmospheric constituents to reach equilibrium. The  
148 reference run is then subtracted from the perturbation run and the difference plotted, thus  
149 showing the impact of aviation on the atmosphere. The variables for the perturbations runs are a  
150 series of increasing aviation emissions, each of which was run in two different background  
151 atmospheric NO<sub>x</sub> states, described below. The spin up and either reference or perturbation run  
152 constitutes a total run time of two years, which is sufficient to show the tropospheric ozone  
153 response to aviation NO<sub>x</sub> emissions<sup>28</sup> and the perturbations to methane lifetime are corrected to  
154 account for its longer lifetime as described in the supplementary material.



155 Ozone and methane are modeled in MOZART-3 using a constant background NO<sub>x</sub> level.  
156 Therefore, to investigate the impact of a changing background atmosphere, two different  
157 background atmospheric NO<sub>x</sub> scenarios (global value and spatial pattern) are used which replace  
158 those from the original background emissions inventory. The values of background NO<sub>x</sub>  
159 emissions used here are 20.76 Tg N yr<sup>-1</sup> and 44.75 Tg N yr<sup>-1</sup> and were taken from the  
160 Representative Concentration Pathways (RCPs) to represent low and high levels of NO<sub>x</sub> in the  
161 background atmosphere. The low NO<sub>x</sub> background comes from RCP3 in the year 2100 and the  
162 high from RCP8 in the year 2020 (see SI, Figure S1). These two values were chosen to represent  
163 the highest and lowest projected range of possible background NO<sub>x</sub> levels over the next 100  
164 years in accordance with the RCP scenarios, thus the results are bounded in that particular range.

165 The aviation scenarios run on MOZART-3 were generated using the REACT4C aircraft  
166 emissions data set<sup>29</sup> as a starting point (from the European project – Reducing Emissions from  
167 Aviation by Changing Trajectories for the benefit of Climate – ‘REACT4C’). The REACT4C  
168 data were then multiplied by different factors to create several aviation emissions scenarios of  
169 increased aviation activity (all with the same spatial pattern). Aviation emissions are expected to  
170 grow more strongly in some regions than others, particularly the Far East/China, differential  
171 growth may affect the balance of the O<sub>3</sub>/CH<sub>4</sub> perturbation. However, this effect has been found  
172 to be small, of the order <3% (see SI). The REACT4C emission scenarios were then modeled  
173 with MOZART-3 using both the low NO<sub>x</sub> and high NO<sub>x</sub> background atmospheres, described  
174 above.

175 Emissions of aircraft NO<sub>x</sub> were calculated to be approximately 0.7 Tg N yr<sup>-1</sup> in the REACT4C  
176 aviation emissions scenario<sup>29</sup> (2006). Emissions scenarios indicate that these emissions may  
177 increase by 2050, over the range 0.8 – 5 Tg N yr<sup>-1</sup> <sup>1, 9, 30-32</sup>. The MOZART CTM was used in a

series of 10 simple computer simulations, scaling up the REACT4C aviation emissions over a ‘realistic’ range of emissions through to beyond those currently anticipated. In addition, 7 further simulations at larger incremental changes ( $> 7 \text{ Tg N yr}^{-1}$ ) were run well beyond what might be considered ‘realistic’ in order that the non-linearity of the response of the system could be evaluated. In total, 17 simulations were run for the ‘high’ background  $\text{NO}_x$  emissions and a further 17 simulations for the ‘low’ background  $\text{NO}_x$  emissions.

In order to develop a new parameterization, the RF of all the effects of aviation  $\text{NO}_x$  emissions release were calculated which, in this study, comprise of short term ozone, methane, long term ozone and stratospheric water vapor (SWV). We acknowledge the effects of aerosols in terms of their overall radiative impact (direct and indirect) of aviation<sup>33</sup>. Their impact on the  $\text{NO}_x$ - $\text{O}_3$ - $\text{CH}_4$  systems is still not well established. Pitari et al. (2015, 2016) find a small effect that reduces the net  $\text{NO}_x$  effect (it being a balance of positive and negative terms) in the aerosol providing a surface for  $\text{NO}_x \rightarrow \text{HNO}_3$  conversion<sup>34-35</sup>. MOZART3 does not include these terms and more work is needed to better establish this effect. Short term ozone RF was calculated using monthly mean ozone fields from MOZART and the Edwards-Slingo radiative transfer model, therefore the relationship between ozone burden and RF is linear (see SI) that also includes a stratospheric adjustment calculation (see SI), methane RF was calculated using the methodology of Hansen et al., (1988)<sup>36</sup>. The use of the ES code also introduces further uncertainties (see SI). The long-term ozone and SWV effects are taken to be 0.5 times the methane forcing (uncertainty 60%) and 0.15 times the methane forcing (uncertainty 71.43%) respectively based on Myhre et al., (2013)<sup>4,37</sup>(one should note that the uncertainties provided here are for global averages, not specifically aviation perturbations).

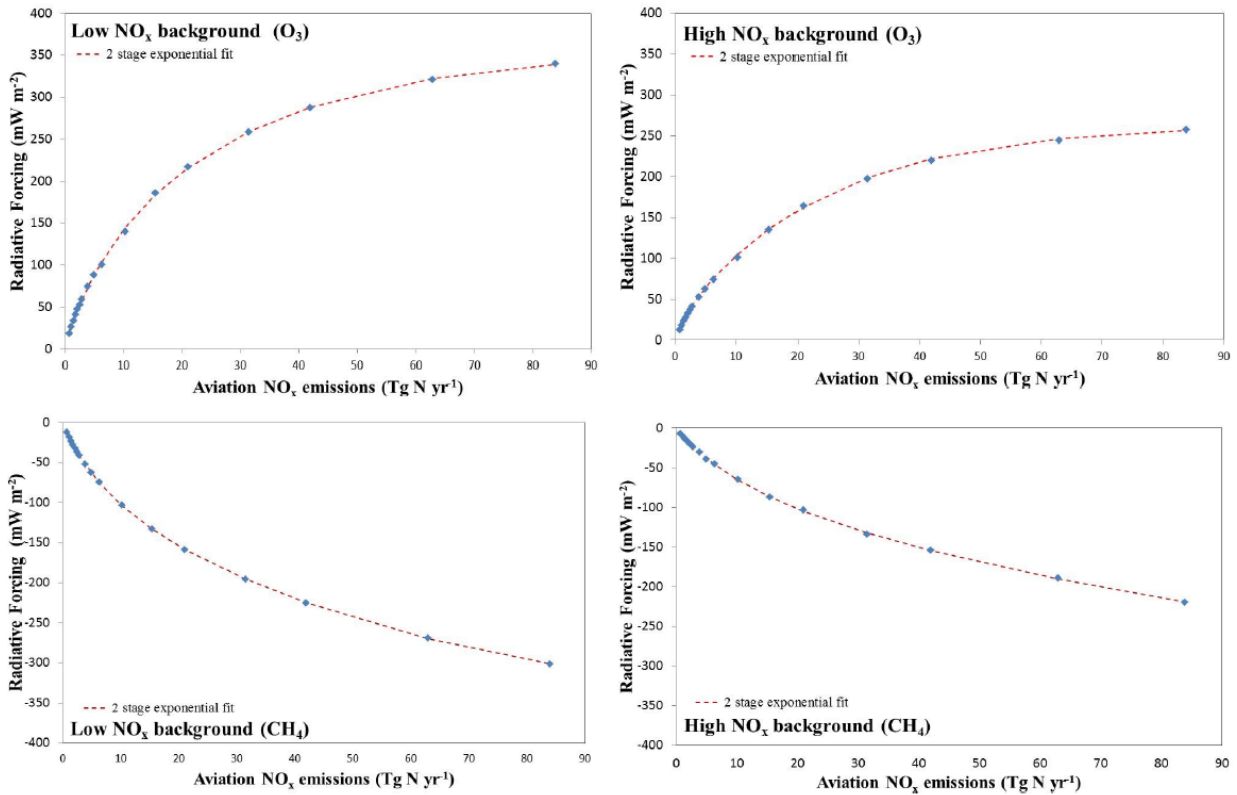
*The Simplified Climate Model, 'LinClim'.* LinClim was used to investigate tradeoffs in the climate response between aviation NO<sub>x</sub> and CO<sub>2</sub> emissions, simulations need to be performed over the longer term. CTMs are computationally very expensive and demanding to run, particularly when complex chemistry is involved. Simple climate models provide a way to simulate future RF responses, from which climate temperature responses can be calculated while running quickly and inexpensively. This type of model can run climate simulations of long duration – up to hundreds of years – using input values of CO<sub>2</sub> and other long-lived greenhouse gases generated from full general circulation model simulations and impulse response functions<sup>12</sup>.

LinClim is a linear climate response model that has been tailored specifically to aviation and includes all the effects of aviation as outlined by the IPCC (1999)<sup>1,38</sup>. The 'linearity' implied in its name assumes that RF and temperature responses are small enough, and can therefore be treated as linear subtractions/additions. Global aviation fuel burn is the input for LinClim and from this, LinClim calculates the resulting emissions of CO<sub>2</sub> and NO<sub>x</sub> using emissions indices. For CO<sub>2</sub>, this is simple, for every 1 kg of fuel burned, 3.16 kg of CO<sub>2</sub> is emitted. CO<sub>2</sub> concentration is then calculated using the impulse response function (IRF) from Hasselmann et al., (1997)<sup>39</sup>. The current carbon cycle in LinClim is based on the Maier-Reimer and Hasselmann (1987)<sup>40</sup> model and the CO<sub>2</sub> RF is calculated with the function used in IPCC AR4<sup>41</sup>. For NO<sub>x</sub>, the emission index (EINO<sub>x</sub>) of the global fleet is required. The current parameterization in LinClim for calculating ozone and methane RF assumes a linear relationship between aviation NO<sub>x</sub> emissions and the resulting ozone and methane RF changes. Therefore, a new parameterization, created using the results from the MOZART runs described above, was used to calculate the RF from aviation NO<sub>x</sub> emissions. This RF value was then used as an input to

223 LinClim and the corresponding temperature response from aviation net NO<sub>x</sub> RF was calculated.  
224 The temperature response formulation is based on the method described in Hasselmann et al.  
225 (1993)<sup>42</sup>. The calculated temperature response is also dependent on the climate sensitivity  
226 parameter and the lifetime of the temperature perturbation. These are tuned to LinClim's 'parent'  
227 Atmosphere-Ocean General Circulation Models (AOGCMs). In this study, LinClim was tuned to  
228 19 different parent models and the median temperature response was taken.

## 229 RESULTS

230 *Effects of aviation NO<sub>x</sub> emissions on ozone and methane abundances.* The results of the  
231 MOZART runs show that as aviation NO<sub>x</sub> emissions increase, so does the associated global  
232 ozone burden and RF (Figure 1; Figure S2). This relationship is approximately linear up to ~2 Tg  
233 N yr<sup>-1</sup> of aviation NO<sub>x</sub> emissions and shows clear non-linearity thereafter in both the low NO<sub>x</sub>  
234 and high NO<sub>x</sub> background atmospheric states. At values of aviation NO<sub>x</sub> emissions greater than  
235 ~2 Tg N yr<sup>-1</sup> ozone formation per NO<sub>x</sub> molecule reduces as aviation emissions increase,  
236 reflecting the non-linearity of the NO<sub>x</sub>-O<sub>3</sub> system<sup>15-17</sup>.



**Figure 1.** The radiative forcing resulting from ozone burden (Tg O<sub>3</sub>) (upper panels) and methane lifetime change (years) (lower panels) due to aviation NO<sub>x</sub> emissions in the low (left hand panels) and high (right hand panel) NO<sub>x</sub> atmospheric background states. Each point represents one of the emissions scenarios run on MOZART described in the text. The trend line shows a two-stage exponential fit of the data, which was used to create a new net NO<sub>x</sub> RF parameterization.

Aviation NO<sub>x</sub> emissions result in an enhancement of OH abundance, which in turn reduces methane lifetime since OH is its principle sink term ( $\text{CH}_4 + \text{OH} \rightarrow \text{CH}_3 + \text{H}_2\text{O}$ ). The change in methane lifetime and reduction in atmospheric abundance associated with the release of aviation NO<sub>x</sub> thus produces a negative RF. Similar to the NO<sub>x</sub> – O<sub>3</sub> relationship, the relationship between aviation NO<sub>x</sub> emissions and methane lifetime reduction (and therefore associated RF) is

approximately linear until aviation NO<sub>x</sub> emissions reach ~2 Tg N yr<sup>-1</sup> (Figure 1; Figure S2) and becomes non-linear thereafter.

The effects of aviation NO<sub>x</sub> emissions on methane lifetime differ depending on the state of the background into which the emissions are released. The lifetime of methane is reduced substantially more (per NO<sub>x</sub> molecule) in the low NO<sub>x</sub> background scenario than the high NO<sub>x</sub> (by an average of 50% over the range of NO<sub>x</sub> emission values used here). The low NO<sub>x</sub> background enables greater formation rates of ozone as described above, which in turn results in an increased concentration of OH and therefore greater decreases in methane lifetime.

The emissions of NO<sub>x</sub> used in this study represent ‘realistic’ values (the highest density of data points in Figure 1 and data shown in Figure S2), through to anticipated ranges of values in future scenarios, to values which are far beyond those expected. However, the purpose of using such values is two-fold; firstly, to demonstrate that the response with a complex global CTM is able to show the expected non-linear response and secondly, to determine at what point the production of O<sub>3</sub> starts to saturate. Clearly, even within the range of emissions suggested in the literature (up to ~5 Tg N yr<sup>-1</sup>), a linear response is not expected, and such a response in a simplified model would over-estimate RF and therefore temperature responses.

It has been established that the responses of ozone and methane to aviation NO<sub>x</sub> emissions are not linear and thus, cannot be treated as such in a parameterization for a simple climate model. The results presented in Figure 1 (and Figure S2) quantify the range over which the linear relationship of NO<sub>x</sub> emissions to ozone burden and methane lifetime change is valid. It is shown that both the NO<sub>x</sub> – O<sub>3</sub> and NO<sub>x</sub> – CH<sub>4</sub> regimes are linear up to ~2 Tg N yr<sup>-1</sup> of aviation NO<sub>x</sub> emissions and therefore, a linear regression is appropriate, however linearity ceases after 2 Tg N

yr<sup>-1</sup> and the data are better represented by exponential fitting. These fit coefficients (Table SI1) can be used to calculate the RF of ozone and methane perturbations resulting from aviation NO<sub>x</sub> emissions in studies using SCMs such as LinClim.

Using the constant emissions scenario described in the methods, and keeping the EINO<sub>x</sub> constant at 13 g NO<sub>2</sub>/kg fuel (3.9 g N/kg), the new parameterization was used to calculate the total forcing from aviation NO<sub>x</sub> emissions over 100 years (Table 1). The results show that in these simplified cases, the background atmosphere determines the sign of the net NO<sub>x</sub> forcing from aviation emissions. In the high NO<sub>x</sub> background, aviation NO<sub>x</sub> emissions contribute a positive net forcing or warming, however, in the low NO<sub>x</sub> background, aviation NO<sub>x</sub> emissions contribute a negative net forcing, or cooling. The difference in sign is due to the fact that in lower NO<sub>x</sub> backgrounds, more OH is available for methane removal, therefore it is enhanced over ozone production in the low NO<sub>x</sub> background, compared with the high NO<sub>x</sub> background where ozone production dominates, resulting in an overall positive net forcing from NO<sub>x</sub>. As the long-term ozone effect and SWV perturbation are calculated from the methane forcing, their contribution enhances the negative forcing in the low NO<sub>x</sub> environment.

Table 1 also gives comparative data on the net NO<sub>x</sub> forcing from LinClim's linear parameterization and the new non-linear parameterization. While the methane forcing is comparable between the two methods, the ozone forcing is overestimated by the linearized form of LinClim. Although this comparison uses low NO<sub>x</sub> values, which fall within the 'linear range' of the NO<sub>x</sub>-O<sub>3</sub>-CH<sub>4</sub> system, the system is still inherently non-linear, and therefore the non-linear regime developed here does give slightly different results.

**Table 1.** Radiative forcing ( $\text{mW m}^{-2}$ ) resulting from aviation  $\text{NO}_x$  calculated using LinClim and the new non-linear parameterizations described in the text, when the same fuel scenario is used – as described in ‘methods’.

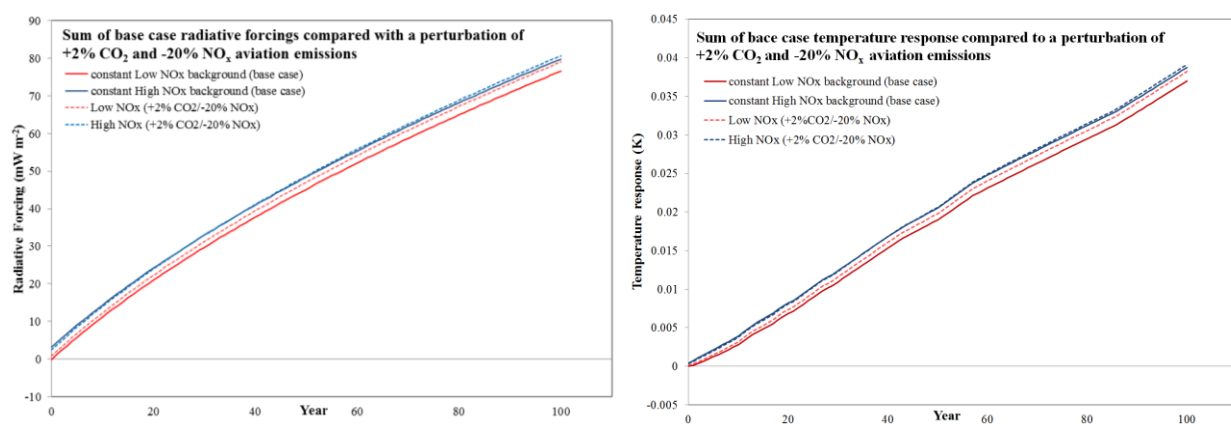
Calculation used/forcing	Short term $\text{O}_3$	Methane	Long term $\text{O}_3$	SWV	Total $\text{NO}_x$ RF
<b>Non-linear</b> (low $\text{NO}_x$ background)	26.80	-17.13	-8.56	-2.57	-1.468
<b>Non-linear</b> (high $\text{NO}_x$ background)	18.09	-9.91	-4.95	-1.48	1.745
<b>Linear</b> (Linclim)	28.74	-13.61	-6.80	-2.04	6.279

*Tradeoff model runs using a simple climate model*

Throughout these model runs, two base case scenarios were considered (Figure 2); total aviation forcing was taken as the  $\text{CO}_2$  plus net  $\text{NO}_x$  forcing, one scenario using the low  $\text{NO}_x$  background and one the high  $\text{NO}_x$  background and the  $\text{CO}_2$  background was set at a constant value of 404 ppm throughout (2016 value, as explained in the methods section). When the base case is perturbed by reducing  $\text{NO}_x$  emissions by 20% and increasing  $\text{CO}_2$  emissions by 2%, total aviation forcing increases by 3.87% for the low background  $\text{NO}_x$  case, and 0.55% for the high background  $\text{NO}_x$  case after 50 years, and by 3.1% and 1.12% after 100 years (low, high  $\text{NO}_x$  backgrounds respectively). This demonstrates that for an ambition that reduces the  $\text{NO}_x$  emissions by 20%, the resultant 2% increase in  $\text{CO}_2$  emissions (Figure 2) means that the total effect is greater than the base case – potentially inadvertently having an adverse effect on climate rather than an intended benefit. Therefore, the next step was to determine exactly how much  $\text{NO}_x$  reduction is required to reduce the total aviation forcing to below that of the base case when  $\text{CO}_2$



emissions are assumed to increase by 2% because of the technology tradeoffs. Emissions of NO<sub>x</sub> were incrementally reduced until the total forcing was the same as the base case. For the high background NO<sub>x</sub> case, aviation NO<sub>x</sub> would need to be reduced by 43% to ‘break even’ in terms of RF after 100 years, or by 38% in terms of temperature response after 100 years. The temperature response is lower due to the thermal inertia of the climate system, since the system has an additional response time over RF.



**Figure 2.** The sum of aviation NO<sub>x</sub> and CO<sub>2</sub> RF (left) and associated temperature response (right) as a result of the constant base case emissions and the initial perturbation case of -20% NO<sub>x</sub>, +2% CO<sub>2</sub>, both described in the text, over 100 years.

For the low background NO<sub>x</sub> case, the results are more complex – net NO<sub>x</sub> emissions provide a negative RF, since methane removal dominates over ozone production. This means that any reduction in aviation NO<sub>x</sub> emissions in the low NO<sub>x</sub> background reduced the negative forcing, leading to an overall greater forcing. Therefore, the only way to reduce overall forcing from aviation when CO<sub>2</sub> emissions are increased by 2% is to, rather counter-intuitively, *increase* aviation NO<sub>x</sub> emissions. This provides an additional negative forcing to counteract the additional positive forcing from CO<sub>2</sub>. This is a somewhat unrealistic case in that the CO<sub>2</sub> penalty would

presumably not be incurred. It was found that aviation NO<sub>x</sub> emissions had to be increased by 37% to counteract the additional RF provided by the 2% increase in CO<sub>2</sub> and reduce the overall forcing to below that of the base case after 100 years (Table 2), and by 33% to reduce the associated temperature response (Table 3). However, what this case does show is that the overall impact in terms of RF and temperature does not depend solely on the technology tradeoffs, but also on the background atmosphere.

The next model runs assume that NO<sub>x</sub> reduction is held at 20% below the base case and it was determined how much of a CO<sub>2</sub> penalty is permitted before total forcing increases above that of the base case. It was calculated that CO<sub>2</sub> can only be allowed to increase by 0.5% over the base case without incurring a forcing or temperature penalty over 100 years in the high NO<sub>x</sub> background. Thus, for this case it can be interpreted that any CO<sub>2</sub> penalty less than 0.5% will yield a net climate benefit. In the low NO<sub>x</sub> background, any reduction in NO<sub>x</sub> emissions causes an increase in overall forcing as described above. Therefore, in this scenario, the forcing is increased over the base case by reducing NO<sub>x</sub> by 20% before any CO<sub>2</sub> increase is considered. Thus, it was determined that, should NO<sub>x</sub> emissions be reduced by 20% in the low NO<sub>x</sub> background, CO<sub>2</sub> emissions would also have to be reduced by 1.5% to counteract the additional forcing and temperature change incurred by the reduction in NO<sub>x</sub> emissions over 100 years (Tables 2 and 3).

**Table 2.** The percentage difference in RF for each perturbation case as compared to the base case.

	High NO <sub>x</sub> background			Low NO <sub>x</sub> background		
		50 year end point	100 year end point		50 year end point	100 year end point
	Model run	% diff from BC	% diff from BC	Model run	% diff from BC	% diff from BC
CO <sub>2</sub> held at +2% from BC	-25% NO <sub>x</sub>	0.16%	0.89%	+21% NO <sub>x</sub>	0.08%	0.87%
	-26% NO <sub>x</sub>	0.08%	0.84%	+22% NO <sub>x</sub>	-0.02%	0.81%
	-27% NO <sub>x</sub>	0.0017%	0.79%	+23% NO <sub>x</sub>	-0.11%	0.76%
	-28% NO <sub>x</sub>	-0.08%	0.74%	+25% NO <sub>x</sub>	-0.30%	0.65%
	-30% NO <sub>x</sub>	-0.25%	0.64%	+30% NO <sub>x</sub>	-0.77%	0.38%
	-40% NO <sub>x</sub>	-1.13%	0.12%	+32% NO <sub>x</sub>	-0.96%	0.27%
	-41% NO <sub>x</sub>	-1.22%	0.06%	+33% NO <sub>x</sub>	-1.05%	0.22%
	-42% NO <sub>x</sub>	-1.31%	0.01%	+34% NO <sub>x</sub>	-1.15%	0.16%
	-43% NO <sub>x</sub>	-1.41%	-0.05%	+35% NO <sub>x</sub>	-1.24%	0.11%
	-44% NO <sub>x</sub>	-1.51%	-0.10%	+36% NO <sub>x</sub>	-1.33%	0.052%
	-45% NO <sub>x</sub>	-1.60%	-0.16%	+37% NO <sub>x</sub>	-1.43%	-0.0019%
NO <sub>x</sub> held at -20% from BC	+0.5% CO <sub>2</sub>	-0.91%	-0.35%	-2% CO <sub>2</sub>	-0.12%	-0.91%
	+1% CO <sub>2</sub>	-0.42%	0.14%	-1.5% CO <sub>2</sub>	0.39%	-0.52%
	+2% CO <sub>2</sub>	0.55%	1.12%	-1% CO <sub>2</sub>	0.90%	0.12%

**Table 3.** The percentage difference in temperature change for each perturbations case as compared to the base case.

	High NO <sub>x</sub> background			Low NO <sub>x</sub> background		
		50 year end point	100 year end point		50 year end point	100 year end point
	Model run	% diff from BC	% diff from BC	Model run	% diff from BC	% diff from BC
CO <sub>2</sub> held at +2% from BC	-20% NO <sub>x</sub>	0.35%	1.01%	+15% NO <sub>x</sub>	0.44%	1.09%
	-23% NO <sub>x</sub>	0.09%	0.85%	+18% NO <sub>x</sub>	0.11%	0.90%
	-24% NO <sub>x</sub>	-0.004%	0.80%	+19% NO <sub>x</sub>	0.005%	0.84%
	-25% NO <sub>x</sub>	-0.09%	0.75%	+20% NO <sub>x</sub>	-0.10%	0.78%
	-38% NO <sub>x</sub>	-1.36%	0.0016%	+30% NO <sub>x</sub>	-1.19%	0.16%
	-40% NO <sub>x</sub>	-1.57%	-0.12%	+32% NO <sub>x</sub>	-1.41%	0.04%
	-41% NO <sub>x</sub>	-1.68%	-0.18%	+33% NO <sub>x</sub>	-1.52%	-0.02%
NO <sub>x</sub> held at -20% from BC	+0.5% CO <sub>2</sub>	-1.11%	-0.46%	-2% CO <sub>2</sub>	0.16%	-0.76%
	+1% CO <sub>2</sub>	-0.62%	0.03%	-1.5% CO <sub>2</sub>	0.67%	-0.52%
	+2% CO <sub>2</sub>	0.35%	1.01%	-1% CO <sub>2</sub>	1.19%	0.27%

## DISCUSSION

The results presented here provide important insights for industrial technology development and policy-making, regarding tradeoffs between different aviation emissions species. It has been found that, while there is a tradeoff between aviation NO<sub>x</sub> and CO<sub>2</sub> emissions, in terms of climate change, CO<sub>2</sub> emissions still provide the majority of the forcing from aviation and a

smaller change in its emission affects the total forcing much more than an equivalent change in  $\text{NO}_x$  emission. The balance of the previously well-known positive RF from ozone, and the counterbalancing negative RF from reduction in methane lifetime has changed with the more recent assessment of the additional negative RF terms from SWV reduction<sup>4</sup>, and reduction in longer-term ozone<sup>43</sup>. One must also consider the role of aviation  $\text{NO}_x$  as a polluter at ground level, and during the landing-take off cycle, hence why its reduction from aircraft emissions is desirable.

In terms of a tradeoff between different emissions, one must cautiously consider where the benefit would lie in reducing one species at the expense of another. Regarding the common scenario proposed in the literature, that a reduction of  $\text{NO}_x$  by 20% incurring a fuel penalty of 2%, while that would reduce pollution from  $\text{NO}_x$  at ground level, it was shown to be worse overall in terms of total climate impact, as the additional  $\text{CO}_2$  forcing from the fuel increase was not counteracted by the reduction in  $\text{NO}_x$  emissions. In terms of the ambition of achieving a climate benefit from  $\text{NO}_x$  emission reductions, we show that a fuel increase should probably be avoided and our test case (20%  $\text{NO}_x$  emission reduction) showed that even an increase of 0.5% fuel would yield no net climate benefit. Either much stronger  $\text{NO}_x$  emission reductions would be necessary, or a condition that no fuel penalty is incurred is the best option. In any case, we show that a careful environmental assessment is required. Even the cases described here may be considered simplistic in terms of realism, but serve as an initial quantitative assessment of tradeoffs which has so far, been absent.

Another important consideration highlighted in this study is the effect of the background atmosphere. If background/surface NO<sub>x</sub> emissions were to decrease, which may be likely as industries aim to cut air pollution at ground level, the net forcing from aviation NO<sub>x</sub> emissions could result in a negative forcing, thus, aviation NO<sub>x</sub> mitigation would not be at all beneficial in terms of climate: however, it is likely that there will be an ongoing requirement to reduce NO<sub>x</sub> emissions at ground-level in order to reduce air pollution impacts on human health. Thus, further consideration of scenarios and test cases should be given to future work to properly assess air quality and climate impacts.

The complex interactions that have been demonstrated here show that scientific assessment and advice can assist in technology development and policy related to aircraft impacts, but it needs to be done with great care – moreover, the interactions between motivations for improving air quality and climate would benefit from extending the results to simple cost-benefit analyses. Currently, only cost-effectiveness analyses are considered in regulatory development within ICAO (International Civil Aviation Organization). As with any atmospheric modeling study, attention must be paid to the uncertainties surrounding computer simulations, the data used and the analysis of the results.

412 ASSOCIATED CONTENT

413 **Supporting Information.** – RF calculations, CH<sub>4</sub> corrections, extra information for CTM, RCP  
414 explanations

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416

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422 **Author Contributions**

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425 **Notes**

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427

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#### ABBREVIATIONS

CO<sub>2</sub> Carbon dioxide  
GHGs Greenhouse gases  
ICAO International Civil Aviation Organization  
IPCC Intergovernmental Panel on Climate Change  
NO<sub>x</sub> Nitrogen oxides (NO + NO<sub>2</sub>)  
RF Radiative Forcing

#### REFERENCES

1. Penner, J., Lister, D. H., Griggs, D. J., Dokken, D. J., McFarland, M. Eds. *Aviation and the global atmosphere. Prepared in collaboration with the Scientific Assessment Panel to the Montreal Protocol on Substances that Deplete the Ozone Layer*; Intergovernmental Panel on Climate Change. Cambridge University Press; UK., 1999.
2. Lee, D. S.; Fahey, D. W.; Forster, P. M.; Newton, P. J.; Wit, R. N. C.; Lim, L. L.; Owen, B.; Sausen, R. Aviation and global climate change in the 21<sup>st</sup> century. *Atmospheric Environment*, **2009**, 43, 3520 – 3537, DOI:10.1016/j.atmosenv.2009.04.024.
3. Lee, D. S.; Pitari, G.; Grewe, V.; Gierens, K.; Penner, J. E.; Petzold, A.; Prather, M. J.; Shumann, U.; Bais, A.; Bernsten, T.; Iachetti, D.; Lim, L. L.; Sausen, R. Transport impacts on atmosphere and climate: Aviation. *Atmospheric Environment*, **2010**, 44, DOI 10.1016/j.atmosenv.2009.06.005.



4. Myhre, G.; Nilsen, J. S.; Gulstad, L.; Shine, K. P.; Rognerud, B.; Isaksen, I. S. A. Radiative forcing due to stratospheric water vapor from CH<sub>4</sub> oxidation. *Geophys. Res. Lett.* **2007**, 34, L01807.
5. Myhre, G.; Shine, K. P.; Rädel, G.; Gauss, M.; Isaksen, I. S. A.; Tang Q.; Prather M. J.; Williams, J. E.; van Velthoven, P.; Dessens, O.; Koffi, B.; Szopa, S.; Hoor, P.; Grewe, V.; Borken-Kleefeld, J.; Berntsen, T. K.; Fuglestvedt, J. S. Radiative forcing due to changes in ozone and methane caused by the transport sector. *Atmospheric Environment*, **2011**, 45, DOI 10.1016/j.atmosenv.2010.10.001.
6. Lewis, J. S.; et al. Aircraft technology and its relation to emissions. In *Aviation and the Global Atmosphere*; Penner, J. E., Lister, D. J., Griggs, D. J., Dokken, D. J., McFarland, M. Eds.; Intergovernmental Panel on Climate Change, Cambridge University Press, Cambridge 1999; pp 373.
7. Faber, J.; Greenwood, D.; Lee, D. S.; Mann, M.; Mendes de Leon, P.; Nelissen, D.; Owen, B.; Ralph, M.; Tilston, J.; van Velzen, A.; van de Vreede, G. Lower NO<sub>x</sub> at higher altitudes. Policies to reduce the climate impact of aviation NO<sub>x</sub> emission, CE-Delft, Delft, The Netherlands, 2008.
8. Kyprianidis, K. G.; Dahlquist, E. On the trade-off between aviation NO<sub>x</sub> and energy efficiency. *Appl. Energy*, **2017**, 185, DOI 10.1016/j.apenergy.2015.12.055.
9. Owen, B.; Lee, D. S.; Lim, L. L. Flying into the Future: aviation emission scenarios to 2050. *Env. Sci. Technol.* **2010**, 44, 2255–2260, DOI 10.1021/es902530z.

10. Flemming, G; Ziegler, U. Environmental and Economic Assessment of NO<sub>x</sub> Stringency Scenarios, Aircraft Technology Improvements. ICAO Environmental Report 2010. ICAO, Montreal, Canada, 2010.
11. Flemming, G; Ziegler, U. Environmental Trends in Aviation to 2050. ICAO Environmental Report 2013, Montreal, Canada, 2013.
12. Sausen, R.; Schumann, U. Estimates of the climate response to aircraft CO<sub>2</sub> and NO<sub>x</sub> emissions scenarios. *Climatic Change*, **2000**, 44, 27 – 58, DOI 10.1023/A:1005579306109.
13. Khodayari, A.; Wuebbles, D. J.; Olsen, S. C.; Fuglestedt, J. S.; Berntsen, T.; Lund, M. T.; Waitz, I.; Wolfe, P.; Forster, P. M.; Meinhausen, M.; Lee, D. S.; Lim, L. L. Intercomparison of the capabilities of simplified climate models to project the effects of aviation CO<sub>2</sub> on climate. *Atmospheric Environment*, **2013**, 75, 321 – 328, DOI 10.1016/j.atmosenv.2013.03.055.
14. Lund, M. T.; Aamaas, B.; Berntsen, T.; Bock, L.; Burkhardt, U.; Fuglestedt, J. S.; Shine, K. P. Emission metrics for quantifying regional climate impacts of aviation. *Earth. Syst. Dynam.*, **2017**, 8, 547 – 563, DOI 10.5194/esd-8-547-2017.
15. Isaksen, I. S. A.; Hov, O.; Hesstvedt, E. Ozone generation over rural areas. *Environ. Sci. Technol.* **1978**, 12, DOI:10.1021/es60147a011.
16. Berntsen, T. K.; Isaksen, I. S. A. Effects of lightning and convection on changes in tropospheric ozone due to NO<sub>x</sub> emissions from aircraft. *Tellus*, **1999**, 51B, 766 – 788, DOI 10.3402/tellusb.v51i4.16484.

17. Stevenson, D. S.; Derwent, R. G. Does the location of aircraft nitrogen oxide emissions affect their climate impact? *Geophys. Res. Letts.* **2009**, 36, DOI: 10.1029/2009GL039422.
18. Fuglestvedt, J. S.; Shine, K. P.; Berntsen, T.; Cook, J.; Lee, D. S.; Stenke, A.; Skeie, R. B.; Velders, G. J. M.; Waitz, I. A. Transport impacts of atmosphere and climate: Metrics. *Atmospheric Environment*, **2010**, 44, DOI 10.1016/j.atmosenv.2009.04.044.
19. ICAO (2010) ICAO environmental report 2010, Montreal, Canada.
20. Newton, P. Long-term Technology Goals for CAEP. Presented at the ICAO Colloquium on Aviation Emissions with Exhibition, 14 – 16 May, 2007.
21. Kinnison, D. E.; Brasseur, G. P.; Walters, S.; Garcia, R. R.; Marsh, D. R.; Sassi, F.; Harvey, V. L.; Randall, C. E.; Emmons, L.; Lamarque, J. F.; Hess, P.; Orlando, J. J.; Tie, X. X.; Randel, W.; Pan, L. L.; Gettleman, A.; Granier, C.; Diehl, T.; Niemeier, U.; Simmons, A. J. Sensitivity of chemical tracers to meteorological parameters in the MOZART-3 chemical transport model. *J. Geophys. Res.* **2007**, 112, DOI 10.1029/2006JD007879.
22. Sassi, F.; Kinnison, D. E.; Boville, B. A.; Garcia, R. R.; Roble, R. Effect of El Nino – southern oscillation on the dynamical, thermal and chemical structure of the middle atmosphere. *J. Geophys. Res.* **2004**, 109, D17108, DOI 10.1029/2003JD004434.
23. Liu, Y.; Liu, C. X.; Wang, H. P.; Tie, X. X.; Gao, S. T.; Kinnison, D.; Brasseur, G. Atmospheric tracers during the 2003-2004 stratospheric warming event and impact of ozone intrusions in the troposphere. *Atmos. Chem. Phys.* **2009**, 9, 2157 – 2170 DOI 10.1007/s11274-015-1903-5.

24. Skowron, A.; Lee, D. S.; De León, R. R. The assessment of the impact of aviation NO<sub>x</sub> on ozone and other radiative forcing responses – The importance of representing cruise altitudes accurately. *Atmospheric Environment*, **2013**, 74, 159 – 168, DOI 10.1016/j.atmosenv.2013.03.034.
25. Skowron, A.; Lee, D. S.; De León, R. R. Variation of radiative forcings and global warming potentials from regional aviation NO<sub>x</sub> emissions. *Atmospheric Environment*, **2015**, 104, 69 – 78, DOI 10.1016/l.atmosenv.2014.12.043.
26. Lamarque, J.-F.; Bond, T. C.; Eyring, V.; Grainer, C.; Heil, A.; Kilmont, Z.; Lee, D.; Liousse, C.; Mieville, A.; Owen, B.; Schultz, M. G.; Shindell, D.; Smith, S. J.; Stehfest, E.; Van Aardenne, J.; Cooper, O. R.; Kainuma, M.; Mahowald, N.; McConnell, J. R.; Naik, V.; Riahi, K.; van Vuuren, D. P. Historical (1850 – 2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: methodology and application. *Atmos. Chem. Phys.* **2010**, 10, 7017 – 7039, DOI 10.5194/acp-10-7017-2010.
27. Granier, C.; Guenther, A.; Lamarque, J. F.; Mieville, A.; Muller, J. F.; Olivier, J.; Orlando, J.; Peters, G.; Petron, G.; Tyndall, G.; Wallens, S. POET, a database of surface emissions of ozone precursors. 2005 (available at [http://eccad.sedoo.fr/eccad\\_extract\\_interface/JSF.jsf](http://eccad.sedoo.fr/eccad_extract_interface/JSF.jsf)).
28. Skowron, A. The impact of emissions of nitrogen oxides from aviation on tropospheric chemistry – the counterbalancing roles of ozone and methane. Ph.D. Thesis, Manchester Metropolitan University, Manchester, UK, 2013.
29. Søvde, O. A.; Matthes, S.; Skowron, A.; Iachetti, D.; Lim, L.; Owen, B.; Hodnebrog, Ø.; Di Genova, G.; Pitari, G.; Lee, D. S.; Myhre, G.; Isaksen, I. S. A. Aircraft emissions

mitigation by changing route altitude: A multi-model estimate of aircraft NO<sub>x</sub> emission impact on O<sub>3</sub> photochemistry. *Atmospheric Environment*, **2014**, 95, 468 – 479, DOI 10.1016/j.atmosenv.2014.06.049.

30. Olsen, S. C.; Wuebbles, D. J.; Owen, B. Comparison of global 3-D aviation emissions datasets. *Atmos. Chem. Phys.* **2013**, 13, 429 – 441, DOI 10.5194/acp-13-429-2013.

31. Khodayari, A.; Olse, S. C.; Wuebbles, D. J. Evaluation of aviation NO<sub>x</sub> induced radiative forcings for 2005 and 2050. *Atmospheric Environment*, **2014**, 91, 95 – 103, DOI 10.1016/j.atmosenv.2014.03.044.

32. Yan, F.; Winijkul, E.; Streets, D. G.; Lu, Z.; Bond, T. C.; Zhang, Y. Global emission projections for the transportation sector using dynamic technology modelling. *Atmos. Chem. Phys.* **2014**, 14, 5709 – 5733, DOI 10.5194/acp-14-5709-2014.

33. Gettelman, A.; Chen, C. The climate impact of aviation aerosols. *Geophys. Res. Lett.*, **2013**, 40, 2785 – 2789, DOI 10.1002/grl.50520.

34. Pitari, G.; Iachetti, D.; Di Genova, G.; De Luca, N.; Søvde, O. A.; Hodnebrog, Ø.; Lee, D. S.; Lim, L. L. Impact of coupled NO<sub>x</sub>/aerosol aircraft emissions on ozone photochemistry and radiative forcing. *Atmosphere*, **2015**, 6 751 – 782, DOI 10.3390/atmos6060751.

558

559 35. Pitari, G.; Cionni, I.; Di Genova, G.; Søvde, O. A.; Lim, L. Radiative forcing from aircraft  
560 emissions of NO<sub>x</sub>: model calculations with CH<sub>4</sub> surface flux boundary condition.  
561 *Meteorologische Zeitschrift*, **2016**, 26 (6), 663 – 687, DOI 10.1127/metz/2016/0776.

562 36. Hansen, J.; Fung, I.; Lacis, A.; Rind, D.; Lebedeff, S.; Ruedy, R.; Russell, G. Global  
563 climate changes as forecast by Goddard Institute for Space Studies three-dimensional  
564 model. *Journal of Geophysical Research*, **1988**, 93 (D8), 9341 – 9364, DOI  
565 10.1029/JD093iD08p09341.

566

567 37. Myhre, G.; Shindell, D.; Bréon, F-M.; Collins, W.; Fuglestvedt, J.; Koch, D.; Lamarque, J-  
568 F.; Lee, D.; Mendoza, B.; Nakajima, T.; Robock., A.; Stephens, G.; Takemura, T.; Zhang,  
569 H. Anthropogenic and Natural Radiative Forcing. In *Climate Change 2013: The Physical  
570 Science Basis. Contribution of working Group I to the Fifth Assessment report of the  
571 Intergovernmental Panel on Climate Change*. Stocker, T. F., Qin, D., Plattner, G-K.,  
572 Tignor, M., Allen, S. K., Baschung, J., Nauels, A., Xia, Y., Bex, V., Midgley, P. M., Eds.;  
573 Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA,  
574 2013.

575 38. Lim, L.; Lee, D. S.; Sausen, R.; Ponater, M. Quantifying the effects of aviation on radiative  
576 forcing and temperature with a climate response model. *Proceedings of the TAC-  
577 Conference*, June 26 – 29, 2006, Oxford, UK.

39. Hasselmann, K.; Hasselmann, S.; Giering, R.; Ocana, V.; VonStorch, H. Sensitivity study of optimal CO<sub>2</sub> emission paths using a simplified structural integrated assessment model (SIAM). *Climatic Change*, **1997**, 37, 345 – 386, DOI 10.1023/A:1005339625015.
40. Maier-Reimer, E.; Hasselmann, K. Transport & storage of CO<sub>2</sub> in the ocean – an inorganic ocean-circulation carbon cycle model. *Climate Dynamics*, **1987**, 2, 63 – 90, DOI 10.1007/BF01054491.
41. Solomon, S.; et al. Technical Summary. In: *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*; Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., Miller, H. L., Eds.; Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA.
42. Hasselmann, K.; Sausen, R.; Maier-Reimer, E.; Voss, R. On the cold start problem in transit simulations with coupled atmosphere-ocean models. *Climate Dynamics*, **1993**, 9 (2), 53 – 61, DOI 10.1007/BF00210008.
43. Holmes C. D.; Tang, Q.; Prather, M. J. Uncertainties in climate assessment for the case of aviation NO. *Proc. Natl. Acad. Sci. USA*. **2011**, 108, DOI 10.1073/pnas.1101458108.