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Trading Off Aircraft Fuel Burn and NO_x Emissions for Optimal Climate Policy

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10 ABSTRACT. Aviation emits pollutants that affect climate, including CO₂ and NO_x; NO_x 11 indirectly so, through the formation of tropospheric ozone and reduction of ambient methane. To 12 improve the fuel performance of engines, combustor temperatures and pressures often increase, 13 increasing NO_x emissions. Conversely, combustor modifications to reduce NO_x may increase 14 CO₂. Hence, a technology tradeoff exists, which also translates to a tradeoff between short lived 15 climate forcers and a long-lived greenhouse gas, CO₂. Moreover, the NO_x-O₃-CH₄ system 16 responds in a non-linear manner, according to both aviation emissions and background NO_x. A 17 simple climate model was modified to incorporate non-linearities parameterized from a complex 18 chemistry model. Case studies showed that for a scenario of a 20% reduction in NO_x emissions 19 the consequential CO₂ penalty of 2% actually increased the total radiative forcing (RF). For a 2% 20 fuel penalty, NO_x emissions needed to be reduced by >43% to realize an overall benefit.

Conversely, to ensure the fuel penalty for a 20% NO_x emission reduction did not increase overall forcing, a 0.5% increase in CO_2 was found to be the 'break even' point. The timescales of the climate effects of NO_x and CO_2 are quite different, necessitating careful analysis of proposed emissions tradeoffs.

25

26 INTRODUCTION

27 Aviation is essential to international travel, and is a growing industry, with passenger traffic 28 increasing at an average of 5.3% per year since 2000. It releases anthropogenic emissions in a 29 physically and chemically complex region of the atmosphere. Aviation emissions consist 30 primarily of carbon dioxide (CO₂), nitrogen oxides (NO_x), sulfur oxides (SO_x) and soot or 'black carbon' emissions, and small amounts of water vapour¹⁻³. The climate impacts of aviation NO_x 31 emissions are complex, since they affect the climate by contributing a positive radiative forcing 32 33 (RF) through the promotion of tropospheric ozone formation and a negative RF by reducing 34 methane lifetime. There are additional negative RF effects from the CH₄ lifetime reduction 35 through small reductions in background O₃ and stratospheric water vapour⁴, although the balance is a net positive forcing 2,5 . At ground level, aviation NO_x is also considered an air pollutant due 36 to its role in ozone production. 37

In 1981, ICAO adopted a first certification standard to control aircraft NO_x emissions in response to concerns over the effect of NO_x emissions on surface air quality. As further NO_x stringency assessments were undertaken it became apparent that the engine modifications necessary to reduce NO_x resulted in a fuel burn penalty, and therefore a CO₂ penalty. Hence, it was realized that a tradeoff existed between the two pollutants⁶⁻⁸. 43 A further issue arises over the timescale of the perturbations to the atmosphere; aviation NO_x 44 emissions and their associated impacts on ozone and methane contribute a short-lived climate 45 forcing to the atmosphere, whereas CO₂ release has an impact on a much longer timescale. In 46 order to understand the environmental consequences of the technology tradeoff, it is necessary to 47 model the climate impacts in some way for both NO_x and CO₂ perturbations over longer 48 timescales. Most studies of the radiative impact of aviation consider either present-day forcing, or a scenario of e.g 2050 emissions^{1, 9-11}. Here, we focus on the very long term as this is not 49 normally considered and only a few studies deal with this¹²⁻¹⁴. The long-term is important as it 50 51 affects the choice of the mitigation options outlined here, i.e. the long-term impact of a small 52 increase in CO₂ emissions that accumulate vs shorter-term effects that reduce forcing.

Adding to the complexity of this, the NO_x-O₃-CH₄ atmospheric system is known to be non-53 54 linear, sensitive to both the perturbing emissions being studied (i.e. aviation) and the NO_x levels of the background atmosphere¹⁵⁻¹⁷. Such calculations are normally conducted with complex 3D 55 56 models of the atmosphere that account for this with a sophisticated chemical scheme. The 57 reduction in CH₄ lifetime, is normally calculated offline by a simplified parameterization since 58 CH_4 has a lifetime of approximately 10 - 12 years. However, model simulations for periods of 59 around 100 years are necessary to account for a significant fraction of the CO₂ emissions, usually 60 done in simplified climate models (SCMs). Previously, small perturbations of the NO_x system have been treated as linear¹² (e.g. Sausen and Schumann, 2000) in SCMs. Since this is known to 61 62 induce inaccuracies into the computations, a new non-linear parameterization of a SCM was 63 derived from a more complex atmospheric chemistry model, MOZART-3, to model the longer-64 term effects of aviation NO_x emissions.

65	Having demonstrated and incorporated a suitable non-linear NO _x scheme into a SCM, a series of
66	model runs were designed in order to study the tradeoff between aviation NO_{x} and CO_{2}
67	emissions over a 100 year period. Through changes in aircraft engine design and emissions
68	characteristics, the relative emissions of NO_x and CO_2 can be tuned to address specific mitigation
69	targets. From the perspective of climate change mitigation, the model runs investigate the
70	amount of NO_x reduction needed to account for any increases in CO_2 emissions and also, how
71	much additional CO_2 can be emitted before additional forcing is incurred, should NO_x emissions
72	be reduced by a set amount, in this case -20%.
73	The model runs also assess the impact of the background NO _x emission on the sign of the NO _x
74	RF and how this impacts on a tradeoff scenario, therefore two different backgrounds NO _x levels

are investigated, one to represent a near present day atmospheric composition and one to
 represent a background atmosphere where significant surface NO_x emissions reduction has taken
 place.

78 METHODS

79 Overall simulations design and modeling tools. Comparing emissions and their climate effects in some form of emission equivalence is a complex subject itself¹⁸. However, in this study, the 80 81 tradeoff question can be posed in a simple way in the sense of variation of RF and change in 82 global mean surface temperature (ΔT) after 100 years for constant emissions conditions over 83 some defined base case. First, the global CTM (chemistry transport model) MOZART was used 84 to investigate the linearity of the NO_x-O₃ and NO_x- CH₄ relationships in response to different 85 background conditions. The results of those model runs were then used to create a new non-86 linear NO_x 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 CO₂ was kept 92 constant at 404 ppm, the background value as of March 2016, thus removing the transient nature 93 of CO_2 modeling - in order to remain consistent with the constant NO_x background used in the 94 CTM runs outlined below. The global fleet emissions index for NO_x (EINO_x in g NO₂/g fuel 95 burned) was kept constant at 13, a representative fleet average. Aviation CO₂ and NO_x emissions 96 were fixed over an arbitrary 100 year simulation at \sim 790 Tg CO₂ (kerosene to CO₂ conversion of 97 3.16) and 3.24 Tg NO₂ (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_x 99 and CO₂ radiative forcings. Note that no 'history' of CO₂ 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_2 and NO_x emissions, while still remaining constant over 102 time. A common scenario from the literature suggested that a 2% fuel penalty could be incurred when NO_x emissions were reduced by 20% - owing to engine modification^{7,19,20} - to determine 103 104 whether a net RF benefit was realized or not. The model runs then followed a logical path of 105 determining how much NO_x reduction is in fact necessary to counteract the additional 2% CO₂ 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_x reduction was held at -108 20% below the base case, how much of CO_2 /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_x background emissions.

111 Two basic modeling tools were necessary – a sophisticated 3D CTM of the global atmosphere 112 ('MOZART' v3) and a simple climate model (SCM)(LinClim). MOZART was used to fully 113 represent the impacts of changing aircraft NO_x emissions at varying levels and backgrounds²¹, 114 the results of which were used to formulate a simplified parameterization in the LinClim SCM 115 ('LinClim', based on Sausen and Schumann, 2000), which simulated both net NO_x and CO₂ 116 radiative impacts. These modeling tools are described below.

117 Three-dimensional global chemical transport model – MOZART. The 3D CTM MOZART

118 (Model for OZone And Related Tracers), version 3, was used to simulate the ozone burden and 119 methane lifetime change resulting from aviation NO_x emissions in this study. MOZART-3 was evaluated by Kinnison et al (2007) and has been applied in several atmospheric studies²²⁻²⁵. The 120 121 European Centre for Medium Range Weather Forecasts (ECMWF) ERA-Interim reanalysis data 122 for 2006 provided the meteorological fields that drive the transport of chemicals within MOZART. The background emissions necessary for MOZART²⁶ represent the year 2000 and 123 124 were originally compiled for the IPCC AR5 report. The background data are made up of surface 125 emissions of anthropogenic activity and biomass burning, and the European Union project POET 126 (Precursors of Ozone and their Effects on Troposphere) supply the biogenic surface emissions²⁷. 127 The choice of meteorology data driving the model will affect the calculations of the NO_x/O₃/CH₄ 128 impacts. Kinnison et al., (2007), when evaluating MOZART3 model performance against 129 observations of various chemical species, noted better agreement when similar ECMWF re-130 analysis data were used vs other dynamical data. MOZART3 was also driven with ECHAM/5 131 GCM data as a test, the results from which are given in the SI. Inter-model variability is another

source of uncertainty in CTM modeling, in Søvde et al., (2014) MOZART3 is tested against
other models in its ability to model NO_x emissions²⁹, the results of that analysis are extended in
the SI, to show the variability of aviation NO_x responses in a small subset of CTMs and how
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_x emissions. Although it is known that aviation 138 NO_x 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 $NO_x - O_3$ and $NO_x - CH_4$, 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_x 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_x 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 response to aviation NO_x emissions²⁸ and the perturbations to methane lifetime are corrected to 153 154 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_x level.
156	Therefore, to investigate the impact of a changing background atmosphere, two different
157	background atmospheric NO _x scenarios (global value and spatial pattern) are used which replace
158	those from the original background emissions inventory. The values of background NO _x
159	emissions used here are 20.76 Tg N yr ⁻¹ and 44.75 Tg N yr ⁻¹ and were taken from the
160	Representative Concentration Pathways (RCPs) to represent low and high levels of NOx in the
161	background atmosphere. The low NO_x 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 _x levels over the next 100
164	years in accordance with the RCP scenarios, thus the results are bounded in that particular range.
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Emissions of aircraft NO_x were calculated to be approximately 0.7 Tg N yr⁻¹ in the REACT4C aviation emissions scenario²⁹ (2006). Emissions scenarios indicate that these emissions may increase by 2050, over the range 0.8 - 5 Tg N yr⁻¹ ^{1, 9, 30-32}. The MOZART CTM was used in a

178 series of 10 simple computer simulations, scaling up the REACT4C aviation emissions over a 179 'realistic' range of emissions through to beyond those currently anticipated. In addition, 7 further 180 simulations at larger incremental changes (> 7 Tg N yr⁻¹) were run well beyond what might be 181 considered 'realistic' in order that the non-linearity of the response of the system could be 182 evaluated. In total, 17 simulations were run for the 'high' background NO_x emissions and a 183 further 17 simulations for the 'low' background NO_x emissions.

184 In order to develop a new parameterization, the RF of all the effects of aviation NO_x emissions 185 release were calculated which, in this study, comprise of short term ozone, methane, long term 186 ozone and stratospheric water vapor (SWV). We acknowledge the effects of aerosols in terms of their overall radiative impact (direct and indirect) of aviation³³. Their impact on the NO_x-O₃-CH₄ 187 188 systems is still not well established. Pitari et al. (2015, 2016) find a small effect that reduces the 189 net NO_x effect (it being a balance of positive and negative terms) in the aerosol providing a surface for NO_x \rightarrow HNO₃ conversion³⁴⁻³⁵. MOZART3 does not include these terms and more 190 191 work is needed to better establish this effect. Short term ozone RF was calculated using monthly 192 mean ozone fields from MOZART and the Edwards-Slingo radiative transfer model, therefore 193 the relationship between ozone burden and RF is linear (see SI) that also includes a stratospheric 194 adjustment calculation (see SI), methane RF was calculated using the methodology of Hansen et al., (1988)³⁶. The use of the ES code also introduces further uncertainties (see SI). The long-term 195 196 ozone and SWV effects are taken to be 0.5 times the methane forcing (uncertainty 60%) and 0.15 197 times the methane forcing (uncertainty 71.43%) respectively based on Myhre et al., $(2013)^{4,37}$ (one should note that the uncertainties provided here are for global averages, not 198 199 specifically aviation perturbations).

200 The Simplified Climate Model, 'LinClim'. LinClim was used to investigate tradeoffs in the 201 climate response between aviation NO_x and CO_2 emissions, simulations need to be performed 202 over the longer term. CTMs are computationally very expensive and demanding to run, 203 particularly when complex chemistry is involved. Simple climate models provide a way to 204 simulate future RF responses, from which climate temperature responses can be calculated while 205 running quickly and inexpensively. This type of model can run climate simulations of long 206 duration - up to hundreds of years - using input values of CO_2 and other long-lived greenhouse 207 gases generated from full general circulation model simulations and impulse response functions¹². 208

209 LinClim is a linear climate response model that has been tailored specifically to aviation and 210 includes all the effects of aviation as outlined by the IPCC (1999)^{1,38}. The 'linearity' implied in 211 its name assumes that RF and temperature responses are small enough, and can therefore be 212 treated as linear subtractions/additions. Global aviation fuel burn is the input for LinClim and 213 from this, LinClim calculates the resulting emissions of CO₂ and NO_x using emissions indices. 214 For CO₂, this is simple, for every 1 kg of fuel burned, 3.16 kg of CO₂ is emitted. CO₂ 215 concentration is then calculated using the impulse response function (IRF) from Hasselmann et al., (1997)³⁹. The current carbon cycle in LinClim is based on the Maier-Reimer and Hasselmann 216 $(1987)^{40}$ model and the CO₂ RF is calculated with the function used in IPCC AR4⁴¹. For NO_x, 217 218 the emission index ($EINO_x$) of the global fleet is required. The current parameterization in 219 LinClim for calculating ozone and methane RF assumes a linear relationship between aviation 220 NO_x emissions and the resulting ozone and methane RF changes. Therefore, a new 221 parameterization, created using the results from the MOZART runs described above, was used to 222 calculate the RF from aviation NO_x emissions. This RF value was then used as an input to

223	LinClim and the corresponding temperature response from aviation net NO _x RF was calculated.
224	The temperature response formulation is based on the method described in Hasselmann et al.
225	$(1993)^{42}$. 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_x emissions on ozone and methane abundances. The results of the
231	MOZART runs show that as aviation NO _x emissions increase, so does the associated global
232	ozone burden and RF (Figure 1; Figure S2). This relationship is approximately linear up to \sim 2 Tg
233	N yr ⁻¹ of aviation NO_x emissions and shows clear non-linearity thereafter in both the low NO_x
234	and high NO _x background atmospheric states. At values of aviation NO _x emissions greater than
235	\sim 2 Tg N yr ⁻¹ ozone formation per NO _x molecule reduces as aviation emissions increase,
236	reflecting the non-linearity of the NO _x -O ₃ system ¹⁵⁻¹⁷ .





Figure 1. The radiative forcing resulting from ozone burden (Tg O₃) (upper panels) and methane lifetime change (years) (lower panels) due to aviation NO_x emissions in the low (left hand panels) and high (right hand panel) NO_x 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_x RF parameterization.

244

Aviation NO_x emissions result in an enhancement of OH abundance, which in turn reduces methane lifetime since OH is its principle sink term (CH₄ + OH \rightarrow CH₃ + H₂O). The change in methane lifetime and reduction in atmospheric abundance associated with the release of aviation NO_x thus produces a negative RF. Similar to the NO_x – O₃ relationship, the relationship between aviation NO_x emissions and methane lifetime reduction (and therefore associated RF) is approximately linear until aviation NO_x emissions reach ~2 Tg N yr⁻¹ (Figure 1; Figure S2) and becomes non-linear thereafter.

252 The effects of aviation NO_x emissions on methane lifetime differ depending on the state of the 253 background into which the emissions are released. The lifetime of methane is reduced 254 substantially more (per NO_x molecule) in the low NO_x background scenario than the high NO_x 255 (by an average of 50% over the range of NO_x emission values used here). The low NO_x 256 background enables greater formation rates of ozone as described above, which in turn results in 257 an increased concentration of OH and therefore greater decreases in methane lifetime. 258 The emissions of NO_x used in this study represent 'realistic' values (the highest density of data 259 points in Figure 1 and data shown in Figure S2), through to anticipated ranges of values in future 260 scenarios, to values which are far beyond those expected. However, the purpose of using such 261 values is two-fold; firstly, to demonstrate that the response with a complex global CTM is able to 262 show the expected non-linear response and secondly, to determine at what point the production 263 of O₃ starts to saturate. Clearly, even within the range of emissions suggested in the literature (up to ~5 Tg N yr⁻¹), a linear response is not expected, and such a response in a simplified model 264 265 would over-estimate RF and therefore temperature responses.

It has been established that the responses of ozone and methane to aviation NO_x 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_x emissions to ozone burden and methane lifetime change is valid. It is shown that both the NO_x – O₃ and NO_x – CH₄ regimes are linear up to ~2 Tg N yr⁻¹ of aviation NO_x emissions and therefore, a linear regression is appropriate, however linearity ceases after 2 Tg N

272 yr⁻¹ and the data are better represented by exponential fitting. These fit coefficients (Table SI1)
273 can be used to calculate the RF of ozone and methane perturbations resulting from aviation NO_x
274 emissions in studies using SCMs such as LinClim.

275

276 Using the constant emissions scenario described in the methods, and keeping the EINO_x constant 277 at 13 g NO₂/kg fuel (3.9 g N/kg), the new parameterization was used to calculate the total forcing from aviation NO_x emissions over 100 years (Table 1). The results show that in these simplified 278 279 cases, the background atmosphere determines the sign of the net NO_x forcing from aviation 280 emissions. In the high NO_x background, aviation NO_x emissions contribute a positive net forcing 281 or warming, however, in the low NO_x background, aviation NO_x emissions contribute a negative 282 net forcing, or cooling. The difference in sign is due to the fact that in lower NO_x backgrounds, 283 more OH is available for methane removal, therefore it is enhanced over ozone production in the 284 low NO_x background, compared with the high NO_x background where ozone production 285 dominates, resulting in an overall positive net forcing from NO_x . As the long-term ozone effect 286 and SWV perturbation are calculated from the methane forcing, their contribution enhances the 287 negative forcing in the low NO_x environment.

288 Table 1 also gives comparative data on the net NO_x forcing from LinClim's linear

289 parameterization and the new non-linear parameterization. While the methane forcing is

290 comparable between the two methods, the ozone forcing is overestimated by the linearized form

291 of LinClim. Although this comparison uses low NO_x values, which fall within the 'linear range'

292 of the NO_x-O₃-CH₄ system, they system is still inherently non-linear, and therefore the non-

293 linear regime developed here does give slightly different results.

Table 1. Radiative forcing (mW m⁻²) resulting from aviation 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 O ₃	Methane	Long term O ₃	SWV	Total NO _x RF
Non-linear (low NOx background)	26.80	-17.13	-8.56	-2.57	-1.468
Non-linear (high NOx background)	18.09	-9.91	-4.95	-1.48	1.745
Linear (Linclim)	28.74	-13.61	-6.80	-2.04	6.279

297

298

299 Tradeoff model runs using a simple climate model

300 Throughout these model runs, two base case scenarios were considered (Figure 2); total aviation 301 forcing was taken as the CO₂ plus net NO_x forcing, one scenario using the low NO_x background 302 and one the high NO_x background and the CO₂ background was set at a constant value of 404 303 ppm throughout (2016 value, as explained in the methods section). When the base case is 304 perturbed by reducing NO_x emissions by 20% and increasing CO₂ emissions by 2%, total 305 aviation forcing increases by 3.87% for the low background NO_x case, and 0.55% for the high 306 background NO_x case after 50 years, and by 3.1% and 1.12% after 100 years (low, high NO_x 307 backgrounds respectively). This demonstrates that for an ambition that reduces the NO_x 308 emissions by 20%, the resultant 2% increase in CO₂ emissions (Figure 2) means that the total 309 effect is greater than the base case – potentially inadvertently having an adverse effect on climate 310 rather than an intended benefit. Therefore, the next step was to determine exactly how much NO_x 311 reduction is required to reduce the total aviation forcing to below that of the base case when CO_2

emissions are assumed to increase by 2% because of the technology tradeoffs. Emissions of NO_x were incrementally reduced until the total forcing was the same as the base case. For the high background NO_x case, aviation NO_x 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_x and CO_2 RF (left) and associated temperature response (right) as a result of the constant base case emissions and the initial perturbation case of -20% NO_x , +2% CO₂, both described in the text, over 100 years.

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For the low background NO_x case, the results are more complex – net NO_x emissions provide a negative RF, since methane removal dominates over ozone production. This means that any reduction in aviation NO_x emissions in the low NO_x background reduced the negative forcing, leading to an overall greater forcing. Therefore, the only way to reduce overall forcing from aviation when CO₂ emissions are increased by 2% is to, rather counter-intuitively, *increase* aviation NO_x emissions. This provides an additional negative forcing to counteract the additional positive forcing from CO₂. This is a somewhat unrealistic case in that the CO₂ penalty would presumably not be incurred. It was found that aviation NO_x emissions had to be increased by 37% to counteract the additional RF provided by the 2% increase in CO_2 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.

336 The next model runs assume that NO_x reduction is held at 20% below the base case and it was 337 determined how much of a CO₂ penalty is permitted before total forcing increases above that of 338 the base case. It was calculated that CO₂ can only be allowed to increase by 0.5% over the base 339 case without incurring a forcing or temperature penalty over 100 years in the high NO_x 340 background. Thus, for this case in can be interpreted that any CO₂ penalty less than 0.5% will 341 yield a net climate benefit. In the low NO_x background, any reduction in NO_x emissions causes 342 an increase in overall forcing as described above. Therefore, in this scenario, the forcing is 343 increased over the base case by reducing NO_x by 20% before any CO₂ increase is considered. 344 Thus, it was determined that, should NO_x emissions be reduced by 20% in the low NO_x 345 background, CO₂ emissions would also have to be reduced by 1.5% to counteract the additional 346 forcing and temperature change incurred by the reduction in NO_x emissions over 100 years 347 (Tables 2 and 3).

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	High NO _x background			Low NO _x background		
		50 year end	100 year end		50 year end	100 year end
		point	point		point	point
	Model run	% diff from BC	% diff from BC	Model run	% diff from BC	% diff from BC
	-25% NO _x	0.16%	0.89%	+21% NO _x	0.08%	0.87%
	-26% NO _x	0.08%	0.84%	+22% NOx	-0.02%	0.81%
	-27% NOx	0.0017%	0.79%	+23% NOx	-0.11%	0.76%
	-28% NOx	-0.08%	0.74%	+25% NOx	-0.30%	0.65%
from BC	-30% NO _x	-0.25%	0.64%	+30% NO _x	-0.77%	0.38%
at +2% i	-40% NOx	-1.13%	0.12%	+32% NO _x	-0.96%	0.27%
O ₂ held	-41% NO _x	-1.22%	0.06%	+33% NO _x	-1.05%	0.22%
Ō	-42% NO _x	-1.31%	0.01%	+34% NO _x	-1.15%	0.16%
	-43% NO _x	-1.41%	-0.05%	+35% NO _x	-1.24%	0.11%
	-44% NOx	-1.51%	-0.10%	+36% NOx	-1.33%	0.052%
	-45% NO _x	-1.60%	-0.16%	+37% NO _x	-1.43%	-0.0019%
% from	+0.5% CO ₂	-0.91%	-0.35%	-2% CO2	-0.12%	-0.91%
d at -209 BC	+1% CO2	-0.42%	0.14%	-1.5% CO2	0.39%	-0.52%
NO _x hel	+2% CO2	0.55%	1.12%	-1% CO2	0.90%	0.12%

353 Table 2. The percentage difference in RF for each perturbation case as compared to the base354 case.

	High NO _x bac	kground		Low NO _x bac	kground	
		50 year end	100 year end		50 year end	100 year end
		point	point		point	point
	Model run	% diff from BC	% diff from BC	Model run	% diff from BC	% diff from BC
	-20% NO _x	0.35%	1.01%	+15% NO _x	0.44%	1.09%
	-23% NOx	0.09%	0.85%	+18% NO _x	0.11%	0.90%
om BC	-24% NOx	-0.004%	0.80%	+19% NO _x	0.005%	0.84%
ıt +2% fı	-25% NOx	-0.09%	0.75%	+20% NO _x	-0.10%	0.78%
02 held a	-38% NO _x	-1.36%	0.0016%	+30% NO _x	-1.19%	0.16%
S	-40% NO _x	-1.57%	-0.12%	+32% NO _x	-1.41%	0.04%
	-41% NOx	-1.68%	-0.18%	+33% NOx	-1.52%	-0.02%
from	+0.5% CO ₂	-1.11%	-0.46%	-2% CO ₂	0.16%	-0.76%
l at -20% BC	+1% CO2	-0.62%	0.03%	-1.5% CO2	0.67%	-0.52%
NO _x helc	+2% CO ₂	0.35%	1.01%	-1% CO2	1.19%	0.27%

358 Table 3. The percentage difference in temperature change for each perturbations case as359 compared to the base case.

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363 DISCUSSION

364 The results presented here provide important insights for industrial technology development and

- 365 policy-making, regarding tradeoffs between different aviation emissions species. It has been
- 366 found that, while there is a tradeoff between aviation NO_x and CO₂ emissions, in terms of
- 367 climate change, CO₂ emissions still provide the majority of the forcing from aviation and a

368 smaller change in its emission affects the total forcing much more than an equivalent change in 369 NO_x emission. The balance of the previously well-known positive RF from ozone, and the 370 counterbalancing negative RF from reduction in methane lifetime has changed with the more 371 recent assessment of the additional negative RF terms from SWV reduction⁴, and reduction in 372 longer-term ozone⁴³. One must also consider the role of aviation NO_x as a polluter at ground 373 level, and during the landing-take off cycle, hence why its reduction from aircraft emissions is 374 desirable.

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376 In terms of a tradeoff between different emissions, one must cautiously consider where the 377 benefit would lie in reducing one species at the expense of another. Regarding the common 378 scenario proposed in the literature, that a reduction of NO_x by 20% incurring a fuel penalty of 379 2%, while that would reduce pollution from NO_x at ground level, it was shown to be worse 380 overall in terms of total climate impact, as the additional CO₂ forcing from the fuel increase was 381 not counteracted by the reduction in NO_x emissions. In terms of the ambition of achieving a 382 climate benefit from NO_x emission reductions, we show that a fuel increase should probably be 383 avoided and our test case (20% NO_x emission reduction) showed that even an increase of 0.5%384 fuel would yield no net climate benefit. Either much stronger NO_x emission reductions would be 385 necessary, or a condition that no fuel penalty is incurred is the best option. In any case, we show 386 that a careful environmental assessment is required. Even the cases described here may be 387 considered simplistic in terms of realism, but serve as an initial quantitative assessment of 388 tradeoffs which has so far, been absent.

390 Another important consideration highlighted in this study is the effect of the background 391 atmosphere. If background/surface NO_x emissions were to decrease, which may be likely as 392 industries aim to cut air pollution at ground level, the net forcing from aviation NO_x emissions 393 could result in a negative forcing, thus, aviation NO_x mitigation would not be at all beneficial in 394 terms of climate: however, it is likely that there will be an ongoing requirement to reduce NO_x 395 emissions at ground-level in order to reduce air pollution impacts on human health. Thus, further 396 consideration of scenarios and test cases should be given to future work to properly assess air 397 quality and climate impacts.

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399 The complex interactions that have been demonstrated here show that scientific assessment and 400 advice can assist in technology development and policy related to aircraft impacts, but it needs to 401 be done with great care – moreover, the interactions between motivations for improving air 402 quality and climate would benefit from extending the results to simple cost-benefit analyses. 403 Currently, only cost-effectiveness analyses are considered in regulatory development within 404 ICAO (International Civil Aviation Organization). As with any atmospheric modeling study, 405 attention must be paid to the uncertainties surrounding computer simulations, the data used and 406 the analysis of the results.

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412 ASSOCIATED CONTENT

- 413 Supporting Information. RF calculations, CH₄ corrections, extra information for CTM, RCP
- 414 explanations
- 415
- 416
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422 Author Contributions

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433	ABBR	EVIATIONS
434	$\rm CO_2$	Carbon dioxide
435	GHGs	Greenhouse gases
436	ICAO	International Civil Aviation Organization
437	IPCC	Intergovernmental Panel on Climate Change
438	NO_x	Nitrogen oxides (NO + NO ₂)
439	RF	Radiative Forcing
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441	REFEI	RENCES
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