

# 1 The assessment of the impact of aviation 2 NO<sub>x</sub> on ozone and other radiative forcing 3 responses – the importance of representing 4 cruise altitudes accurately

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6 A. Skowron, D. S. Lee, R. R. De León

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8 <sup>1</sup>Dalton Research Institute, Manchester Metropolitan University, John Dalton  
9 Building, Chester Street, Manchester M1 5GD, United Kingdom.

10

11 **Abstract:** Aviation emissions of NO<sub>x</sub> result in the formation of tropospheric  
12 ozone (warming) and destruction of a small amount of methane (cooling),  
13 positive and negative radiative forcing effects. In addition, the reduction of  
14 methane results in a small long-term reduction in tropospheric ozone (cooling)  
15 and, in addition, a long-term reduction in water vapour in the stratosphere  
16 (cooling) from reduced oxidation of methane, both negative radiative forcing  
17 impacts. Taking all these radiative effects together, aircraft NO<sub>x</sub> is still thought to  
18 result in a positive (warming) radiative effect under constant emissions  
19 assumptions. Previously, comparative modelling studies have focussed on the  
20 variability between models, using the same emissions database. In this study, we  
21 rather quantify the variability and uncertainty arising from different estimations  
22 of present-day aircraft NO<sub>x</sub> emissions. Six different aircraft NO<sub>x</sub> emissions  
23 inventories were used in the global chemical transport model, MOZART v3. The  
24 inventories were normalized to give the same global emission of NO<sub>x</sub> in order to  
25 remove one element of uncertainty. Emissions differed in the normalized cases  
26 by 23% at cruise altitudes (283 hPa – 200 hPa, where the bulk of emission  
27 occurs, globally). However, the resultant short-term ozone chemical  
28 perturbation varied by 15% between the different inventories. Once all the  
29 effects that give rise to positive and negative radiative impacts were accounted  
30 for, the variability of net radiative forcing impacts was 94%. Using these  
31 radiative effects to formulate a net aviation NO<sub>x</sub> Global Warming Potential (GWP)

32 for a 100-year time horizon resulted in GWPs ranging from 60 to 4, over an order  
33 of magnitude. It is concluded that the detailed placement of emissions at  
34 chemically sensitive cruise altitudes strongly affects the assessment of the total  
35 radiative impact, introducing a hitherto previously unidentified large fraction of  
36 the uncertainty of impacts between different modelling assessments. It is  
37 recommended that future formulations of aircraft NO<sub>x</sub> emissions focus efforts on  
38 the detailed and accurate placement of emissions at cruise altitudes to reduce  
39 the uncertainty in future assessments of aviation NO<sub>x</sub> impacts.

40

41

42 *Keywords:* Aviation, radiative forcing, emissions, nitrogen oxides, ozone response

43 **1 Introduction**

44

45 The impact of aviation NO<sub>x</sub> emissions on the production of tropospheric ozone  
46 (O<sub>3</sub>) has been investigated since the early 1970s (Hidalgo and Crutzen, 1977).  
47 Several thematic research programmes in the US and Europe investigated  
48 aircraft NO<sub>x</sub> effects on tropospheric chemistry in the 1980s and 1990s (see Lee  
49 et al., 2010 for a summary).

50

51 Despite the length of time over which this effect has been investigated, it still  
52 represents an active research area. One particular milestone was the IPCC  
53 (1999) Special Report on 'Aviation and the Global Atmosphere', which  
54 summarized results of a number of 3D global chemical transport models (CTMs),  
55 which were relatively newly developed over 2D models. The IPCC (1999)  
56 highlighted the finding that whilst NO<sub>x</sub> emissions from the existing fleet of  
57 subsonic aircraft resulted in a small increase in tropospheric O<sub>3</sub>, there was also a  
58 small but significant reduction in ambient CH<sub>4</sub> (for an equilibrium calculation of  
59 constant emissions), since CH<sub>4</sub> has a lifetime of approximately 8–12 years and  
60 takes some time to respond to an additional NO<sub>x</sub> increase. The IPCC thus  
61 identified a positive ozone radiative forcing (RF) and a negative RF associated  
62 with a NO<sub>x</sub> increase from aircraft.

63

64 Wild et al. (2001) also identified that with this long-term CH<sub>4</sub> decrease, a small  
65 decrease in O<sub>3</sub> also resulted (again, for equilibrium conditions). However, it has  
66 taken some time to realise that this O<sub>3</sub> decrease could be significant over the  
67 longer time-period and that the overall RF response from aircraft NO<sub>x</sub> arises  
68 from one positive and two negative RF responses. More recently, Myhre et al.  
69 (2011) summarized a number of model responses to an aircraft NO<sub>x</sub> increase and  
70 also highlighted a fourth RF response in that a decrease in CH<sub>4</sub> also ultimately  
71 resulted in a small negative RF response from water vapour in the stratosphere.  
72 Any CH<sub>4</sub> response takes decades to come to an equilibrium response and its  
73 mixing time means that it can enter the stratosphere where it can be oxidised to  
74 water vapour. The water vapour results in a positive forcing in the stratosphere,

75 so that any reduction in CH<sub>4</sub> will result in a reduction in water vapour forcing in  
76 the stratosphere and can therefore be viewed as a negative RF from aviation NO<sub>x</sub>.  
77

78 The complexity of NO<sub>x</sub>-O<sub>3</sub>-CH<sub>4</sub> system is intensified not only through different  
79 timescales of responses of its components (positive forcing is short-term,  
80 negative responses are long-term), but also by differences of their spatial  
81 extents: while short-term O<sub>3</sub> enhancement is regional, the CH<sub>4</sub>, CH<sub>4</sub>-induced O<sub>3</sub>  
82 and stratospheric water vapour (SWV) act on a global scale.

83

84 Many studies have been published over the past 20 years assessing the impact of  
85 aviation NO<sub>x</sub> emissions on tropospheric chemistry and RF (see Lee et al., 2010  
86 for a recent review, along with Myhre et al., 2011). Among them, the  
87 investigations regarding the impact of aircraft NO<sub>x</sub> from modified cruise altitudes  
88 also exist (Gauss et al., 2006, Frömming et al., 2012). However, these studies are  
89 not always straightforward to compare, since the models have had varying  
90 degrees of complexity in terms of completeness of representation of  
91 tropospheric and stratospheric chemistry, horizontal and vertical resolution.

92

93 There are numbers of tools which are utilized in order to place on a common  
94 scale different climate impacts. The most traditional are radiative forcing (RF)  
95 and global warming potential (GWP). Radiative forcing (RF) is an accepted  
96 measure of the strength of the perturbation of Earth-atmosphere system caused  
97 by natural agents and human activity. The global warming potential (GWP) is a  
98 ratio of the RF from the emission of a species relative to that of CO<sub>2</sub> for a nominal  
99 kg release of both gases, integrated over given time horizon. While radiative  
100 forcing is a backward looking measure, the global warming potential through its  
101 relativity is one step further in the cause-effect chain.

102

103 In this paper, we revisit the NO<sub>x</sub> impact on chemical composition of the  
104 troposphere using a 3D CTM, MOZART v3 (Kinnison et al., 2007) and quantify  
105 the individual RF responses from short-term O<sub>3</sub> increases, long-term CH<sub>4</sub> and O<sub>3</sub>  
106 decreases, and CH<sub>4</sub> feedback effects on stratospheric water vapour. Most  
107 importantly, the impact of using different emission inventories is investigated in

108 this work, since all previous comparative studies have utilised different models.  
109 Here, we take a novel approach of using a single model with different  
110 (normalized) emissions that have slightly different assumptions and therefore  
111 vertical distributions of emissions in order to better understand the impact of  
112 height distribution of aircraft NO<sub>x</sub> emissions and their representation in  
113 emission inventories.

114

## 115 **2 Methodology and experimental design**

116

### 117 *2.1 Aircraft emission inventories*

118

119 The aircraft inventory datasets are usually generated from an aircraft movement  
120 database, the characteristic of a global fleet in terms of type of aircraft and  
121 engines, fuel-flow model, calculation of emissions at vertical scale from fuel flow,  
122 landing and take-off emissions (LTO). The aircraft movement databases are a  
123 mixture of a flight plan data, flight operation data, radar data, Official Aviation  
124 Guide (OAG) data ([www.oag.com](http://www.oag.com)) and idealized great circle routes analysis. A  
125 comprehensive comparison of global aviation inventories was recently  
126 presented by Olsen et al. (2013).

127 Here, six different aircraft inventories were investigated:

128

129 1. AEDT (Aviation Environmental Tool) for the year 2006 (Wilkerson et al.,  
130 2010). The global aircraft data was provided by Volpe National Transportation  
131 Systems Centre. The aircraft fuel burn and emissions were estimated based on  
132 an individual flight by flight analysis. This inventory is based on radar data for  
133 Europe and North America, which account for 70-80% of global aircraft  
134 movements, and for the remaining flight movements the OAG data were used.

135

136 2. AEM (Advanced Emission Model) for the year 2006  
137 ([www.eurocontrol.int/services/advanced-emission-model](http://www.eurocontrol.int/services/advanced-emission-model)). AEM is a stand-  
138 alone system (developed and maintained by EUROCONTROL), which calculates  
139 aviation emissions and fuel burn. It uses a few basic databases: aircraft, aircraft

140 engines, fuel burn rates and emissions indices. AEM is aimed to analyse the flight  
141 profile data, on a flight by flight bases, for different air traffic scenarios.

142

143 3. AERO2K inventory for the year 2002 (Eyers et al., 2005) was developed  
144 under the EC 5th Framework Programme. The dataset is based on a radar  
145 tracked flight data for North America and Europe. Data for the rest of the world  
146 are covered by scheduled flights data from Back Aviation database (Back, 2002)  
147 and by routing information. Forty representative aircraft types were applied in  
148 order to calculate the fuel burn and emissions for each flight using means from  
149 the PIANO ([www.piano.aero](http://www.piano.aero)) aircraft performance model.

150

151 4. REACT4C (EC 7th Framework Programme Reducing Emissions from  
152 Aviation by Changing Trajectories For the benefits of Climate) for the year 2006  
153 ([www.react4c.eu](http://www.react4c.eu)). The input data are the CAEP-8 comprehensive set of aircraft  
154 movements, which are individual movements for 6 weeks of the year, scaled to a  
155 full year's movements. The air traffic movements are from radar data for flights  
156 for Europe and North America and the remaining global flight movements are  
157 taken from OAG. The models used to generate this inventory are: the fuel-flow  
158 model PIANO (Project Interactive Analysis and Optimization model) and global  
159 emissions model FAST (The Future Aviation Scenario Tool) (Owen et al., 2010),  
160 similarly as for QUANTIFY and TRADEOFF data presented below.

161

162 5. QUANTIFY (Quantifying the Climate Impact of Global and European  
163 Transport Systems) for a year 2000 (Owen et al., 2010). The inventory consists  
164 of OAG data for scheduled flights and AERO2K's traffic for non-scheduled aircraft  
165 movements. The QUANTIFY dataset, once released, was scaled to the  
166 International Energy Aviation (IEA) aviation fuel burn total for year 2000.

167

168 6. TRADEOFF for the year 1992 (Gauss et al., 2006). The dataset was created  
169 based on a flight track data from the EUROCONTROL and FAA from the year  
170 1991/1992. Four months of aircraft movements: July 1991, October 1991,  
171 January 1992 and April 1992 were scaled to a full year's movement. The global  
172 aircraft movement data are a combination of air traffic control and scheduled

173 data. Sixteen civil aircraft-engine type combinations represent the global fleet of  
174 aircraft.

175 Aircraft inventories used in this study are three dimensional gridded datasets,  
176 with a 1°x1° horizontal resolution and a vertical resolution which varies from  
177 1km through 610m to 500 ft. An overview of the characteristics of each  
178 inventory is presented in Table 1. Military emissions have not been taken into  
179 account in this study.

180

181 Each dataset represents different years of emissions, which results in different  
182 amounts of burned fuel ranging from 210 Tg/yr for AEDT (2006) to 114 Tg/yr  
183 for TRADEOFF (1992) which affects the emitted NO<sub>x</sub>. In order to exclude the  
184 differences in the amount of injected NO<sub>x</sub> and consequently its impact on O<sub>3</sub>  
185 response, the NO<sub>x</sub> emissions of each inventory were scaled to the same global  
186 total as the REACT4C , which is 2.33 Tg (NO<sub>2</sub>)/yr.

187

## 188 *2.2 Global chemical transport model of the atmosphere*

189

190 The Model for Ozone and Related Tracers, version 3 (MOZART-3) was used in  
191 this study. It is a 3D Chemistry Transport Model (CTM) comprehensively  
192 evaluated by Kinnison et al. (2007) and extensively used for different  
193 applications, e.g. impact of El Niño and La Niña events on dynamical, thermal and  
194 chemical structure of the middle atmosphere (Sassi et al., 2004), distribution of  
195 stratospheric O<sub>3</sub> and downward O<sub>3</sub> transport in the UTLS region during the  
196 sudden stratospheric warming event in January 2004 (Liu et al., 2009), forecast  
197 analysis of the ozone hole over Antarctica in 2008 (Flemming et al.,2011),  
198 evaluation of Ozone Depletion Potentials for n-propyl bromide (Wuebbles et al.,  
199 2011).

200

201 MOZART-3 is built on the framework of the transport model MATCH (Model for  
202 Atmospheric Transport and Chemistry) (Rasch et al., 1997) and accounts for  
203 advection (flux-form semi-Lagrangian scheme of Lin and Rood (1996));  
204 convection (shallow and mid-level convection scheme of Hack et al. (1994) and

205 deep convective transport of Zhang and MacFarlane (1995)); boundary layer  
206 exchanges (Holstag and Boville, 1993) and wet and dry deposition (Brausser et  
207 al., (1998) and Müller (1992), respectively).

208

209 MOZART-3 represents detailed chemical and physical processes from the  
210 troposphere through the stratosphere. The chemical mechanism consists of 108  
211 species, 218 gas-phase reactions, 71 photolytic reactions (including the  
212 photochemical reactions associated with organic halogen compounds) and 17  
213 heterogeneous reactions. The kinetic and photochemical data is from NASA/JPL  
214 (Sanders et al., 2006).

215

216 The anthropogenic (non-aviation) and biomass burning emissions are taken  
217 from Lamarque et al. (2010) and represent the year 2000. The biogenic surface  
218 emissions are taken from the European Union project POET (Precursors of Ozone  
219 and their Effects on Troposphere) (Granier et al., 2005).

220

221 The horizontal resolution applied in this study is T42 ( $\sim 2.8^\circ \times 2.8^\circ$ ) and the  
222 vertical domain extends from the surface to 0.1hPa with 60 hybrid layers (Figure  
223 1). The transport of chemical compounds is driven by the meteorological fields  
224 from European Centre for Medium Range Weather Forecasting (ECMWF),  
225 reanalysis ERA-Interim for the year 2000 (Simmons et al., 2007).

226

227 Seven experiments were performed, one reference (no-aircraft) run and six  
228 perturbation (aircraft) simulations, each starting in January 2000 and finishing  
229 in December 2000; each simulation was preceded by one year spin-up. The  
230 aircraft perturbation is derived by extracting the difference between aircraft and  
231 no-aircraft experiments. The calculations of O<sub>3</sub> change and CH<sub>4</sub> lifetime change,  
232 along with RFs, covers the surface-1 hPa domain. Since our experiments are  
233 performed for 2 years, the magnitude of aircraft stratospheric response is not  
234 fully representative. The O<sub>3</sub> depletion due to aircraft NO<sub>x</sub> emissions, from current  
235 fleets, is relatively small (-0.01 DU), thus it does not affect O<sub>3</sub> changes  
236 significantly. However, the O<sub>3</sub> column change, presented in this paper is  
237 overestimated by 5.1% for surface-1 hPa domain and the short-term O<sub>3</sub> RF is



238 underestimated by 0.6%. The CH<sub>4</sub> lifetime reduction and its negative RF are  
239 overestimated by 0.2%.

240

241 *2.3 Radiative forcings and global warming potentials calculations*

242

243 The short-term O<sub>3</sub> radiative forcings are calculated off-line using the Edwards –  
244 Slingo radiation code (Edwards and Slingo, 1996). This comprehensive radiative  
245 transfer model was developed in the UK Meteorological Office and is based on  
246 the two-stream equations in both the long-wave and short-wave spectral  
247 regions. Cloud treatment is based on averaged ISCCP D2 data (Rossow and  
248 Schiffer, 1999). Climatological fields of temperature and specific humidity are  
249 based on ERA-Interim data (Simmons et al., 2007). The calculations were  
250 performed on monthly O<sub>3</sub> MOZART-3 output. To account for a stratospheric  
251 adjustment a 20% reduction was applied to the O<sub>3</sub> RF, following the work of  
252 Stevenson et al. (1998).

253

254 A one year CTM simulation is not long enough to calculate the change in CH<sub>4</sub>  
255 concentration, as it takes decades for CH<sub>4</sub> to come into equilibrium with the  
256 perturbed OH fields. That is why to obtain the steady state concentrations of CH<sub>4</sub>  
257 in the perturbation runs the change in CH<sub>4</sub> lifetime owing to reaction with OH  
258 was calculated for each inventory, which then, based on Fuglestvedt et al. (1999),  
259 was multiplied by the reference CH<sub>4</sub> concentration and a feedback factor of 1.4  
260 (Prather et al., 2001) to reflect the effect of changes of CH<sub>4</sub> on its own lifetime:  
261  $[\text{CH}_4]_{\text{ss}} = [\text{CH}_4]_{\text{ref}} * (1 + 1.4 * \Delta\alpha_0 / \alpha_{\text{ref}})$ , where  $\Delta\alpha_0 = \alpha_{\text{per}} - \alpha_{\text{ref}}$  and  $[\text{CH}_4]_{\text{ref}}$  is a  
262 reference run concentration.

263 The RF of CH<sub>4</sub> is calculated using a simplified expression defined in Ramaswamy  
264 et al. (2001). The impact of methane change on stratospheric water vapour  
265 (SWV) is also included and as described in Myhre et al. (2007) the RF of SWV is  
266 assumed to be 0.15 times that of methane RF. The CH<sub>4</sub>-induced O<sub>3</sub> is computed  
267 based on an assumption that 10% increase of CH<sub>4</sub> leads to 0.64 DU increase of  
268 ozone (Prather et al., 2001) and this ozone has a specific RF of 42 mW m<sup>-2</sup> DU<sup>-1</sup>  
269 (Ramaswamy et al., 2001).

270 The temporal evolution of net RF following the NO<sub>x</sub> emission is required in order  
271 to calculate GWP. It can be assumed that the constant one year emission is a step  
272 emission followed by a decay of the resulting forcing from the end of the year  
273 onwards. The GWP calculations are based on a methodology described by  
274 Fuglestvedt et al., (2010). The primary-mode lifetime is taken into account, while  
275 the long-term effects (CH<sub>4</sub> with SWV and CH<sub>4</sub>-induced O<sub>3</sub>) are integrated over  
276 time horizons.

277

### 278 **3 Results**

279

#### 280 *3.1 Aircraft NO<sub>x</sub> emissions*

281

282 Since various methodologies have been applied to derive different inventories,  
283 the distribution of NO<sub>x</sub> emissions reveals some discrepancies. The geographical  
284 distribution shows rather common picture (Figure 2). The image of altitudinal  
285 spacing is not so unique (Figure 3).

286

287 The Northern Hemisphere is the main location of aircraft NO<sub>x</sub> emissions:  
288 emissions in 30-60N and 0-30N regions constitute 65% and 24%, respectively,  
289 for AEDT, AEM and REACT4C inventories; AERO2K, QUANTIFY and TRADEOFF  
290 have slightly more emissions in mid northern latitudes (68%) and less over the  
291 northern tropical region (21%). Most of aircraft NO<sub>x</sub> emissions occur over North  
292 America, Europe and South-East Asia. AERO2K, QUANTIFY and TRADEOFF have  
293 a bit lower emissions, by ~4%, in 60-120E region and a bit more, by ~4%, in  
294 120-60W region than AEDT, AEM and REACT4C.

295

296 The largest part of the NO<sub>x</sub> emissions are injected between 9 and 12 km for most  
297 of the inventories, only AEDT and AEM have more aircraft NO<sub>x</sub> over 12 km than  
298 the other inventories. AERO2K's NO<sub>x</sub> emissions at cruise altitudes constitute only  
299 43% of its total aircraft NO<sub>x</sub> emissions (Figure 3), which, when compared with  
300 57% of QUANTIFY, 58% of AEM, 59% of REACT4C and TRADEOFF and 63% of  
301 AEDT, is quite low. The 'missing' ~10% is hidden under AERO2K's relatively high  
302 NO<sub>x</sub> emissions at mid-altitudes, which is 34%, while for all the rest of inventories

303 it is around 25%. The difference in the vertical structure of NO<sub>x</sub> emissions  
304 between AERO2K and the other datasets is significant.

305 The original aircraft emission data, with their regular vertical gridding (500ft,  
306 610m or 1km) are interpolated by MOZART-3 to its irregular (with hybrid sigma  
307 layers every ~1 km in the upper troposphere and lower stratosphere (UTLS)  
308 region) vertical spacing (Figure 1). Figure 4 shows the vertical distribution of  
309 aircraft NO<sub>x</sub> emissions in MOZART-3 for the six aircraft inventories. Each dataset  
310 represents the same amount of global total aircraft NO<sub>x</sub> (2.33 Tg NO<sub>2</sub>). The initial  
311 resolution of dataset plays a significant role when it is redistributed into the  
312 lowest CTM's vertical layers. Taking into account that the vertical resolution in  
313 MOZART-3 near surface is high (~45 m) the datasets with higher resolution  
314 (AEM and AERO2K) have more aircraft NO<sub>x</sub> emissions near ground (1000-950  
315 hPa).

316 In MOZART-3 most of aircraft NO<sub>x</sub> emissions are injected in the 283-200 hPa  
317 region, where the emissions differ by 23% when the greatest (TRADEOFF) and  
318 the smallest (AERO2K) numbers are taken into account. The peak of aircraft NO<sub>x</sub>  
319 emissions is observed at 227 hPa, with the greatest values occurring for  
320 REACT4C and TRADEOFF. AEDT and AEM have more emissions at 200 hPa and  
321 at higher altitudes, than other inventories, which raises the possibility of more  
322 efficient accumulation of N molecules (Seinfeld and Pandis, 2006).

323

### 324 *3.2 Chemical perturbation*

325

326 The response of the NO<sub>x</sub>-O<sub>3</sub>-CH<sub>4</sub> system affected by aviation NO<sub>x</sub> emissions is  
327 presented in Figure 5. The positive peak of NO<sub>x</sub> response is observed at 227 hPa  
328 for all inventories, except for AEDT and AEM, which have their maximum one  
329 level higher, at 201 hPa. This suggests that potential of NO<sub>x</sub> perturbation is  
330 greater for higher NO<sub>x</sub> emissions. The greatest NO<sub>x</sub> response is observed for  
331 AEDT and the smallest for AERO2K, which consequently affects the O<sub>3</sub>  
332 perturbation, which follows the same pattern of differences between datasets in  
333 terms of emissions. The largest O<sub>3</sub> response takes place at 227 hPa level for all

334 inventories. The enhanced O<sub>3</sub> changes the oxidizing capacity of the troposphere.  
335 In general, aircraft NO<sub>x</sub> perturbs the OH/HO<sub>2</sub> ratio: increases OH and decreases  
336 HO<sub>2</sub>. The positive OH response is observed through all tropospheric domain, the  
337 negative HO<sub>2</sub> response is observed mainly at flight altitudes. While the impact of  
338 AERO2K inventory on NO<sub>x</sub> and O<sub>3</sub> in UTLS region is relatively weak, it is  
339 responsible for greater aircraft OH in mid-altitudes and consequently CO and  
340 CH<sub>4</sub> changes, than other datasets. The more efficient CO oxidation results in a  
341 greater AERO2K's HO<sub>2</sub> perturbation in mid-altitudes compared to other  
342 inventories.

343

344 The same amount of emitted NO<sub>x</sub>, but different vertical distributions of NO<sub>x</sub>  
345 emissions lead to significant differences in short-term O<sub>3</sub> response between  
346 inventories. Table 2 gives global and annual means of total column O<sub>3</sub> change (in  
347 DU) and ozone production efficiency values for six different inventories. The  
348 greatest column change is observed for AEDT (0.56 DU) and AEM (0.54 DU) and  
349 the smallest is for AERO2K (0.48 DU). Also QUANTIFY shows a relatively lower  
350 O<sub>3</sub> perturbation (0.50 DU) compared with the other FAST inventories REACT4C  
351 and TRADEOFF (0.52 DU).

352 The O<sub>3</sub> production in the troposphere has been shown to be sensitive to the  
353 height of the initial precursor emissions (Köhler et al., 2008). This is indirectly  
354 observed in our results, where more molecules of ozone are produced per  
355 molecule of N emitted for inventories with NO<sub>x</sub> emissions at higher altitudes, it  
356 being 30 for AEDT, 29 for AEM, 28 for REACT4C, 27 for QUANTIFY and  
357 TRADEOFF and 25 for AERO2K (Table 2). This shows that a lower potential, by  
358 15%, is observed for AERO2K inventory compared to AEDT dataset in terms of  
359 ozone production, which is consistent with the spread of O<sub>3</sub> column change.

360

361 The methane lifetime due to destruction by OH in a reference case was observed  
362 as 8.88 years. In contrast to the O<sub>3</sub> responses, the CH<sub>4</sub> lifetime reductions are  
363 observed to be quite uniform among different inventories (Table 3) ranging from  
364 -0.074 years for AERO2K and TRADEOFF, -0.073 years for REACT4C to -0.070  
365 years for AEDT.

366

367 3.3 *Radiative forcing and global warming potential for aviation NO<sub>x</sub> emissions*

368

369 The latitudinal distributions of short-term O<sub>3</sub> RF for the six inventories are  
370 shown in Figure 6. The pattern for each inventory shows similar characteristics  
371 and it is consistent with zonal-mean distributions of NO<sub>x</sub> emissions (Figure 2)  
372 (both rather short-lived), with dominating role of short-term O<sub>3</sub> RF over the  
373 Northern Hemisphere. While agreement in the resultant short-term O<sub>3</sub> RF  
374 between inventories at high southern and northern latitudes is observed the  
375 tropical region (30°S-40°N) shows discrepancies. The largest spread in the  
376 short-term O<sub>3</sub> RF between inventories occurs over northern tropical belt (15-  
377 30°N), where locally the standard deviation reaches 3.0-3.5 mWm<sup>-2</sup>, over Middle  
378 East, Pacific and North Africa.

379

380 Table 4 presents the global and annual mean RF (mWm<sup>-2</sup>) for short-term O<sub>3</sub>,  
381 CH<sub>4</sub>-induced O<sub>3</sub>, CH<sub>4</sub>, and SWV for a series of inventories. The standard deviation  
382 of short-term O<sub>3</sub> values is 1.0 mWm<sup>-2</sup> (with AEDT and AERO2K resulting in  
383 highest and lowest numbers, 14.3 and 11.5 mWm<sup>-2</sup>, respectively). The CH<sub>4</sub>  
384 responses are much more consistent, the standard deviation is 0.2 mWm<sup>-2</sup> (with  
385 values ranging from -7.1 mWm<sup>-2</sup> for AERO2K and TRADEOFF and -6.7 mWm<sup>-2</sup> for  
386 AEDT). The net aircraft NO<sub>x</sub> RF values ranges from 3.6 mWm<sup>-2</sup> for AEDT, 2.3  
387 mWm<sup>-2</sup> for REACT4C to 0.2 mWm<sup>-2</sup> for AERO2K, with 1.2 mWm<sup>-2</sup> standard  
388 deviation.

389

390 Myhre et al. (2011) reported GWP values for aviation NO<sub>x</sub> emissions using the  
391 same aircraft emissions, the same experimental design and a range of five  
392 models. The differences in their results constitute a good insight into  
393 uncertainties which arise from usage of different global chemistry models. In  
394 contrast, the spread in results which are presented in this study gives a measure  
395 of differences that arise from usage of different aircraft inventories. The  
396 resulting GWP values for three time horizons (20, 100 and 500 years) are given  
397 in Table 5. The values show significant differences, which are enhanced with  
398 larger time horizons; however, the sign of calculated responses shows a  
399 consistently net positive value. The largest differences come from the AEDT and

400 AERO2K inventories, being 57%, 93% different for 20, 100 time horizons  
401 respectively. The increase of discrepancy with larger time horizons can be  
402 explained by CH<sub>4</sub>, as its response 'remains' for a few decades after NO<sub>x</sub> emission.  
403 The GWP reduction between a 20 year time horizon and a 100 year horizon is  
404 larger for inventories where the CH<sub>4</sub> lifetime reduction is more enhanced, e.g. it  
405 is 82% for AEDT, 83% for AEM, 85% for REACT4C, 87% for QUANTIFY and  
406 TRADEOFF, 97% for AERO2K. Due to relatively short lifetimes of the net NO<sub>x</sub>  
407 components the GWP (H=500) differs between inventories only by the CO<sub>2</sub>  
408 integral in the denominator.

409

#### 410 **4 Discussion**

411

412 Figure 7 shows the vertical profiles of the relative difference of NO<sub>x</sub>, O<sub>3</sub>, OH and  
413 HO<sub>2</sub> responses to that of the AEDT inventory (chosen simply as it gives the  
414 largest overall response). The aircraft NO<sub>x</sub> perturbation for AEDT occurring at  
415 227 hPa is about 25% greater than that for AERO2K. The difference increases  
416 with higher altitudes and constitutes 55% at 100 hPa. On the contrary, AERO2K's  
417 NO<sub>x</sub> significantly exceeds that of AEDT at mid altitudes, where the difference  
418 reaches 500% at 762 hPa. The response of the O<sub>3</sub> for these discrepancies is not  
419 the same. It is observed that AERO2K's O<sub>3</sub> response indeed dominates in the low-  
420 troposphere region (1000-600 hPa) but only by about 4%, whereas AEDT's  
421 dominance at cruise altitudes reaches 25% and 50% at 100 hPa. The same  
422 pattern is observed for all inventories, just the scale of differences is not so well  
423 pronounced. The linear correlation between additional NO<sub>x</sub> and O<sub>3</sub> response is  
424 observed in UTLS region, being the strongest at 227 hPa and becoming weaker at  
425 higher altitudes, for AERO2K the ratio of O<sub>3</sub> to NO<sub>x</sub> is 1 at 227 hPa and 0.9 at  
426 100hPa (e.g. for AEM it is 1 at 227 hPa and 0.7 at 100 hPa, for REACT4C it is 1.5  
427 at 227 hPa and 0.8 at 100 hPa).

428

429 Interestingly, the greatest O<sub>3</sub> aircraft perturbation did not introduce the  
430 strongest CH<sub>4</sub> reduction, as might be expected from the chemistry. The  
431 altitudinal distribution of emissions can hide the explanation: a significant  
432 fraction of AERO2K's NO<sub>x</sub> emissions occurs in the mid-troposphere. The ~4%'s

433 dominance of AERO2K's O<sub>3</sub> in 900-700 hPa region significantly changes the  
434 oxidizing capacity of the low troposphere (there is more OH by about 30% than  
435 for AEDT). Annual mean concentrations of OH and CH<sub>4</sub> and temperature are  
436 greater at lower altitudes which catalyse OH production and CH<sub>4</sub> destruction.

437

438 Another implication of AERO2K's enhancement of O<sub>3</sub> concentrations at lower  
439 altitudes is that this O<sub>3</sub> is not as radiatively efficient as O<sub>3</sub> at higher altitudes  
440 (Lacis et al., (1990), Köhler et al., (2008)).

441

442 It is worth to mention, that not only the height is important, also the  
443 geographical distribution of aircraft NO<sub>x</sub> emissions plays a certain role in terms  
444 of O<sub>3</sub> RF response. The NO<sub>x</sub> emissions from low latitudes have a greater impact  
445 on climate forcings than the NO<sub>x</sub> emissions from high latitudes (Berntsen et al.,  
446 (2005), Köhler et al., (2012)) The AERO2K dataset has lower, compared to other  
447 inventories, NO<sub>x</sub> emissions at cruise altitudes over East Asia and over the Pacific  
448 (not shown here), which was also noted by Olsen et al. (2012). Even though  
449 certain regional differences in the distribution of aircraft NO<sub>x</sub> emissions occur,  
450 they are not as powerful as altitudinal discrepancies in terms of O<sub>3</sub> production  
451 (based on work to be published).

452

453 Figure 8 shows a short-term O<sub>3</sub> RF and net NO<sub>x</sub> RF responses normalized to 1 Tg  
454 of emitted N reported for a number of model simulations (Stordal et al. (2006),  
455 Hoor et al. (2009), Myhre et al. (2011), Hodnebrog et al. (2011, 2012)). The  
456 inter-model mean O<sub>3</sub> and NO<sub>x</sub> RF values are 22.2 and 5.0 mW m<sup>-2</sup>/Tg(N) yr<sup>-1</sup>,  
457 respectively. While for short-term O<sub>3</sub> responses most of the model results are  
458 placed within a one standard deviation range, the net NO<sub>x</sub> RF values constitute a  
459 more diverse picture. This can be explained by the inter-study differences in the  
460 components taken into account for net NO<sub>x</sub> calculation. Myhre et al. (2011)  
461 pointed out that the ratio of the CH<sub>4</sub> lifetime change to the O<sub>3</sub> column change is  
462 very specific for each model, which also influence the net NO<sub>x</sub> numbers. The  
463 results produced by MOZART-3 are in good agreement with other studies.

464

465 The spread in RF values (20% for O<sub>3</sub> RF and 94% for net NO<sub>x</sub> RF) between six  
466 aircraft inventories is of a smaller magnitude to that of uncertainties between  
467 models; however, it is still significant. Myhre et al. (2011) reported a 36% spread  
468 in O<sub>3</sub> RF values and 54% in net NO<sub>x</sub> RF values between a set of five different  
469 models (note that Myhre et al. (2011) account for the time-history emissions  
470 (Grewe and Stenke, 2008) in their long term effects); Hoor et al. (2009) reported  
471 a 64% spread in O<sub>3</sub> RF and 89% in net NO<sub>x</sub> (net is without SWV) values between  
472 five different models and Stordal et al. (2006) showed a 33% spread in O<sub>3</sub> RF and  
473 59% in net NO<sub>x</sub> RF (net is a sum of short-term O<sub>3</sub> and CH<sub>4</sub> only) values between  
474 three different models. This places the discrepancies between different  
475 inventories on the same scale of importance as inter-model differences.

476

## 477 **5 Conclusions**

478

479 The coupled NO<sub>x</sub>-O<sub>3</sub>-CH<sub>4</sub> system, as affected by aviation NO<sub>x</sub> emissions, results in  
480 a regional short-term O<sub>3</sub> positive radiative forcing and a global long-term O<sub>3</sub>, CH<sub>4</sub>  
481 and SWV negative responses. Nonetheless the overall radiative forcing induced  
482 by current day emissions of aviation NO<sub>x</sub> from 3D CTM MOZART-3 is positive as  
483 shown by this study, ranging from 3.6 to 0.2 mW m<sup>-2</sup>.

484

485 By using one model (3D CTM MOZART-3) and a series of aircraft inventories  
486 (AEDT, AEM, AERO2K, REACT4C, QUANTIFY, TRADEOFF) scaled to the same  
487 global total (2.33 Tg (NO<sub>2</sub>)/yr), the sensitivity of O<sub>3</sub> response due to the  
488 discrepancies in vertical distribution of aircraft NO<sub>x</sub> emissions was investigated.  
489 It is observed that the differences in the vertical distribution of aircraft NO<sub>x</sub>  
490 emissions between inventories, with AEDT and AERO2K being the most  
491 different, strongly influence the aircraft short-term O<sub>3</sub> response and  
492 consequently the net NO<sub>x</sub> effect.

493

494 The aviation impact on RF per unit emission of NO<sub>x</sub> differs from inventory to  
495 inventory and ranges from 5.2 to 0.3 mW m<sup>-2</sup>/Tg(N) yr<sup>-1</sup> (for AEDT and AERO2K,  
496 respectively). The responses of short-term O<sub>3</sub> RF range from 20.5 to 16.5 mW m<sup>-2</sup>  
497 <sup>2</sup>/Tg(N) yr<sup>-1</sup> (for AEDT and AERO2K, respectively). It is observed that both, the



498 O<sub>3</sub> and net NO<sub>x</sub> RF are greater for inventories with higher peak of perturbation of  
499 NO<sub>x</sub> concentrations, which is a direct result of the amount of emitted NO<sub>x</sub> at  
500 higher altitudes.

501

502 The spread between aircraft short-term O<sub>3</sub> RF (20%) and aircraft net NO<sub>x</sub> RF (94  
503 %) values, which emerges from usage of different aircraft inventories should not  
504 be neglected as it constitutes a significant range of uncertainty. Careful attention  
505 should be paid to formulating aircraft emission inventories where precise cruise  
506 altitudes are defined.

507

508

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517

518

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