The assessment of the impact of aviation NO_X on ozone and other radiative forcing responses – the importance of representing cruise altitudes accurately

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Abstract: Aviation emissions of NO_x result in the formation of tropospheric ozone (warming) and destruction of a small amount of methane (cooling), positive and negative radiative forcing effects. In addition, the reduction of methane results in a small long-term reduction in tropospheric ozone (cooling) and, in addition, a long-term reduction in water vapour in the stratosphere (cooling) from reduced oxidation of methane, both negative radiative forcing impacts. Taking all these radiative effects together, aircraft NO_x is still thought to result in a positive (warming) radiative effect under constant emissions assumptions. Previously, comparative modelling studies have focussed on the variability between models, using the same emissions database. In this study, we rather quantify the variability and uncertainty arising from different estimations of present-day aircraft NO_x emissions. Six different aircraft NO_x emissions inventories were used in the global chemical transport model, MOZART v3. The inventories were normalized to give the same global emission of NO_x in order to remove one element of uncertainty. Emissions differed in the normalized cases by 23% at cruise altitudes (283 hPa – 200 hPa, where the bulk of emission occurs, globally). However, the resultant short-term ozone chemical perturbation varied by 15% between the different inventories. Once all the effects that give rise to positive and negative radiative impacts were accounted for, the variability of net radiative forcing impacts was 94%. Using these radiative effects to formulate a net aviation NO_x Global Warming Potential (GWP)

for a 100-year time horizon resulted in GWPs ranging from 60 to 4, over an order of magnitude. It is concluded that the detailed placement of emissions at chemically sensitive cruise altitudes strongly affects the assessment of the total radiative impact, introducing a hitherto previously unidentified large fraction of the uncertainty of impacts between different modelling assessments. It is recommended that future formulations of aircraft NO_x emissions focus efforts on the detailed and accurate placement of emissions at cruise altitudes to reduce the uncertainty in future assessments of aviation NO_x impacts.

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

1 Introduction

The impact of aviation NO_x emissions on the production of tropospheric ozone (O_3) has been investigated since the early 1970s (Hidalgo and Crutzen, 1977). Several thematic research programmes in the US and Europe investigated aircraft NO_x effects on tropospheric chemistry in the 1980s and 1990s (see Lee et al., 2010 for a summary).

Despite the length of time over which this effect has been investigated, it still represents an active research area. One particular milestone was the IPCC (1999) Special Report on 'Aviation and the Global Atmosphere', which summarized results of a number of 3D global chemical transport models (CTMs), which were relatively newly developed over 2D models. The IPCC (1999) highlighted the finding that whilst NO_x emissions from the existing fleet of subsonic aircraft resulted in a small increase in tropospheric O_3, there was also a small but significant reduction in ambient CH_4 (for an equilibrium calculation of constant emissions), since CH_4 has a lifetime of approximately 8–12 years and takes some time to respond to an additional NO_x increase. The IPCC thus identified a positive ozone radiative forcing (RF) and a negative RF associated with a NO_x increase from aircraft.

Wild et al. (2001) also identified that with this long-term CH_4 decrease, a small decrease in O_3 also resulted (again, for equilibrium conditions). However, it has taken some time to realise that this O_3 decrease could be significant over the longer time-period and that the overall RF response from aircraft NO_x arises from one positive and two negative RF responses. More recently, Myhre et al. (2011) summarized a number of model responses to an aircraft NO_x increase and also highlighted a fourth RF response in that a decrease in CH_4 also ultimately resulted in a small negative RF response from water vapour in the stratosphere. Any CH_4 response takes decades to come to an equilibrium response and its mixing time means that it can enter the stratosphere where it can be oxidised to water vapour. The water vapour results in a positive forcing in the stratosphere,

so that any reduction in CH_4 will result in a reduction in water vapour forcing in the stratosphere and can therefore be viewed as a negative RF from aviation NO_x.

The complexity of NO_x-O_3-CH_4 system is intensified not only through different timescales of responses of its components (positive forcing is short-term, negative responses are long-term), but also by differences of their spatial extents: while short-term O_3 enhancement is regional, the CH_4, CH_4-induced O_3 and stratospheric water vapour (SWV) act on a global scale.

Many studies have been published over the past 20 years assessing the impact of aviation NO_x emissions on tropospheric chemistry and RF (see Lee et al., 2010 for a recent review, along with Myhre et al., 2011). Among them, the investigations regarding the impact of aircraft NO_x from modified cruise altitudes also exist (Gauss et al., 2006, Frömming et al., 2012). However, these studies are not always straightforward to compare, since the models have had varying degrees of complexity in terms of completeness of representation of tropospheric and stratospheric chemistry, horizontal and vertical resolution.

There are numbers of tools which are utilized in order to place on a common scale different climate impacts. The most traditional are radiative forcing (RF) and global warming potential (GWP). Radiative forcing (RF) is an accepted measure of the strength of the perturbation of Earth-atmosphere system caused by natural agents and human activity. The global warming potential (GWP) is a ratio of the RF from the emission of a species relative to that of CO_2 for a nominal kg release of both gases, integrated over given time horizon. While radiative forcing is a backward looking measure, the global warming potential through its relativity is one step further in the cause-effect chain.

In this paper, we revisit the NO_x impact on chemical composition of the troposphere using a 3D CTM, MOZART v3 (Kinnison et al., 2007) and quantify the individual RF responses from short-term O_3 increases, long-term CH_4 and O_3 decreases, and CH_4 feedback effects on stratospheric water vapour. Most importantly, the impact of using different emission inventories is investigated in

this work, since all previous comparative studies have utilised different models. Here, we take a novel approach of using a single model with different (normalized) emissions that have slightly different assumptions and therefore vertical distributions of emissions in order to better understand the impact of height distribution of aircraft NO_x emissions and their representation in emission inventories.

2 Methodology and experimental design

2.1 Aircraft emission inventories

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

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

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

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

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

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

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

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

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

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

Each dataset represents different years of emissions, which results in different amounts of burned fuel ranging from 210 Tg/yr for AEDT (2006) to 114 Tg/yr for TRADEOFF (1992) which affects the emitted NO_x. In order to exclude the differences in the amount of injected NO_x and consequently its impact on O_3 response, the NO_x emissions of each inventory were scaled to the same global total as the REACT4C , which is 2.33 Tg (NO_2)/yr.

2.2 Global chemical transport model of the atmosphere

The Model for Ozone and Related Tracers, version 3 (MOZART-3) was used in this study. It is a 3D Chemistry Transport Model (CTM) comprehensively evaluated by Kinnison et al. (2007) and extensively used for different applications, e.g. impact of El Niño and La Niña events on dynamical, thermal and chemical structure of the middle atmosphere (Sassi et al., 2004), distribution of stratospheric O_3 and downward O_3 transport in the UTLS region during the sudden stratospheric warming event in January 2004 (Liu et al., 2009), forecast analysis of the ozone hole over Antarctica in 2008 (Flemming et al.,2011), evaluation of Ozone Depletion Potentials for n-propyl bromide (Wuebbles et al., 2011).

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

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

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

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

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

Seven experiments were performed, one reference (no-aircraft) run and six perturbation (aircraft) simulations, each starting in January 2000 and finishing in December 2000; each simulation was preceded by one year spin-up. The aircraft perturbation is derived by extracting the difference between aircraft and no-aircraft experiments. The calculations of O_3 change and CH_4 lifetime change, along with RFs, covers the surface-1 hPa domain. Since our experiments are performed for 2 years, the magnitude of aircraft stratospheric response is not fully representative. The O_3 depletion due to aircraft NO_x emissions, from current fleets, is relatively small (-0.01 DU), thus it does not affect O_3 changes significantly. However, the O_3 column change, presented in this paper is overestimated by 5.1% for surface-1 hPa domain and the short-term O_3 RF is

underestimated by 0.6%. The CH_4 lifetime reduction and its negative RF are overestimated by 0.2%.

2.3 Radiative forcings and global warming potentials calculations

The short-term O_3 radiative forcings are calculated off-line using the Edwards – Slingo radiation code (Edwards and Slingo, 1996). This comprehensive radiative transfer model was developed in the UK Meteorological Office and is based on the two-stream equations in both the long-wave and short-wave spectral regions. Cloud treatment is based on averaged ISCCP D2 data (Rossow and Schiffer, 1999). Climatological fields of temperature and specific humidity are based on ERA-Interim data (Simmons et al., 2007). The calculations were performed on monthly O_3 MOZART-3 output. To account for a stratospheric adjustment a 20% reduction was applied to the O_3 RF, following the work of Stevenson et al. (1998).

A one year CTM simulation is not long enough to calculate the change in CH_4 concentration, as it takes decades for CH_4 to come into equilibrium with the perturbed OH fields. That is why to obtain the steady state concentrations of CH_4 in the perturbation runs the change in CH_4 lifetime owing to reaction with OH was calculated for each inventory, which then, based on Fuglestvedt et al. (1999), was multiplied by the reference CH_4 concentration and a feedback factor of 1.4 (Prather et al., 2001) to reflect the effect of changes of CH_4 on its own lifetime: $[CH_4]_{ss} = [CH_4]_{ref} * (1 + 1.4 * \Delta\alpha_0/\alpha_{ref})$, where $\Delta\alpha_0 = \alpha_{per} - \alpha_{ref}$ and $[CH_4]_{ref}$ is a reference run concentration.

The RF of CH_4 is calculated using a simplified expression defined in Ramaswamy et al. (2001). The impact of methane change on stratospheric water vapour (SWV) is also included and as described in Myhre et al. (2007) the RF of SWV is assumed to be 0.15 times that of methane RF. The CH_4-induced O_3 is computed based on an assumption that 10% increase of CH_4 leads to 0.64 DU increase of ozone (Prather et al., 2001) and this ozone has a specific RF of 42 mW m^{-2} DU^{-1} (Ramaswamy et al., 2001).

The temporal evolution of net RF following the NO_x emission is required in order to calculate GWP. It can be assumed that the constant one year emission is a step emission followed by a decay of the resulting forcing from the end of the year onwards. The GWP calculations are based on a methodology described by Fuglestvedt et al., (2010). The primary-mode lifetime is taken into account, while the long-term effects (CH_4 with SWV and CH_4-induced O_3) are integrated over time horizons.

3 Results

3.1 Aircraft NO_x emissions

Since various methodologies have been applied to derive different inventories, the distribution of NO_x emissions reveals some discrepancies. The geographical distribution shows rather common picture (Figure 2). The image of altitudinal spacing is not so unique (Figure 3).

The Northern Hemisphere is the main location of aircraft NO_x emissions: emissions in 30-60N and 0-30N regions constitute 65% and 24%, respectively, for AEDT, AEM and REACT4C inventories; AERO2K, QUANTIFY and TRADEOFF have slightly more emissions in mid northern latitudes (68%) and less over the northern tropical region (21%). Most of aircraft NO_x emissions occur over North America, Europe and South-East Asia. AERO2K, QUANTIFY and TRADEOFF have a bit lower emissions, by ~4%, in 60-120E region and a bit more, by ~4%, in 120-60W region than AEDT, AEM and REACT4C.

The largest part of the NO_x emissions are injected between 9 and 12 km for most of the inventories, only AEDT and AEM have more aircraft NO_x over 12 km than the other inventories. AERO2K's NO_x emissions at cruise altitudes constitute only 43% of its total aircraft NO_x emissions (Figure 3), which, when compared with 57% of QUANTIFY, 58% of AEM, 59% of REACT4C and TRADEOFF and 63% of AEDT, is quite low. The 'missing' ~10% is hidden under AERO2K's relatively high NO_x emissions at mid-altitudes, which is 34%, while for all the rest of inventories

it is around 25%. The difference in the vertical structure of NO_x emissions between AERO2K and the other datasets is significant.

The original aircraft emission data, with their regular vertical gridding (500ft, 610m or 1km) are interpolated by MOZART-3 to its irregular (with hybrid sigma layers every ~1 km in the upper troposphere and lower stratosphere (UTLS) region) vertical spacing (Figure 1). Figure 4 shows the vertical distribution of aircraft NO_x emissions in MOZART-3 for the six aircraft inventories. Each dataset represents the same amount of global total aircraft NO_x (2.33 Tg NO_2). The initial resolution of dataset plays a significant role when it is redistributed into the lowest CTM's vertical layers. Taking into account that the vertical resolution in MOZART-3 near surface is high (~45 m) the datasets with higher resolution (AEM and AERO2K) have more aircraft NO_x emissions near ground (1000-950 hPa).

In MOZART-3 most of aircraft NO_x emissions are injected in the 283-200 hPa region, where the emissions differ by 23% when the greatest (TRADEOFF) and the smallest (AERO2K) numbers are taken into account. The peak of aircraft NO_x emissions is observed at 227 hPa, with the greatest values occurring for REACT4C and TRADEOFF. AEDT and AEM have more emissions at 200 hPa and at higher altitudes, than other inventories, which raises the possibility of more efficient accumulation of N molecules (Seinfeld and Pandis, 2006).

3.2 Chemical perturbation

The response of the NO_x-O_3-CH_4 system affected by aviation NO_x emissions is presented in Figure 5. The positive peak of NO_x response is observed at 227 hPa for all inventories, except for AEDT and AEM, which have their maximum one level higher, at 201 hPa. This suggests that potential of NO_x perturbation is greater for higher NO_x emissions. The greatest NO_x response is observed for AEDT and the smallest for AERO2K, which consequently affects the O_3 perturbation, which follows the same pattern of differences between datasets in terms of emissions. The largest O_3 response takes place at 227 hPa level for all

inventories. The enhanced O_3 changes the oxidizing capacity of the troposphere. In general, aircraft NO_x perturbs the OH/HO_2 ratio: increases OH and decreases HO_2. The positive OH response is observed through all tropospheric domain, the negative HO_2 response is observed mainly at flight altitudes. While the impact of AERO2K inventory on NO_x and O_3 in UTLS region is relatively weak, it is responsible for greater aircraft OH in mid-altitudes and consequently CO and CH_4 changes, than other datasets. The more efficient CO oxidation results in a greater AERO2K's HO_2 perturbation in mid-altitudes compared to other inventories.

The same amount of emitted NO_x, but different vertical distributions of NO_x emissions lead to significant differences in short-term O_3 response between inventories. Table 2 gives global and annual means of total column O_3 change (in DU) and ozone production efficiency values for six different inventories. The greatest column change is observed for AEDT (0.56 DU) and AEM (0.54 DU) and the smallest is for AERO2K (0.48 DU). Also QUANTIFY shows a relatively lower O_3 perturbation (0.50 DU) compared with the other FAST inventories REACT4C and TRADEOFF (0.52 DU).

The O_3 production in the troposphere has been shown to be sensitive to the height of the initial precursor emissions (Köhler et al., 2008). This is indirectly observed in our results, where more molecules of ozone are produced per molecule of N emitted for inventories with NO_x emissions at higher altitudes, it being 30 for AEDT, 29 for AEM, 28 for REACT4C, 27 for QUANTIFY and TRADEOFF and 25 for AERO2K (Table 2). This shows that a lower potential, by 15%, is observed for AERO2K inventory compared to AEDT dataset in terms of ozone production, which is consistent with the spread of O_3 column change.

The methane lifetime due to destruction by OH in a reference case was observed as 8.88 years. In contrast to the O_3 responses, the CH_4 lifetime reductions are observed to be quite uniform among different inventories (Table 3) ranging from -0.074 years for AERO2K and TRADEOFF, -0.073 years for REACT4C to -0.070 years for AEDT.

3.3 Radiative forcing and global warming potential for aviation NO_x emissions

The latitudinal distributions of short-term O_3 RF for the six inventories are shown in Figure 6. The pattern for each inventory shows similar characteristics and it is consistent with zonal-mean distributions of NO_x emissions (Figure 2) (both rather short-lived), with dominating role of short-term O_3 RF over the Northern Hemisphere. While agreement in the resultant short-term O_3 RF between inventories at high southern and northern latitudes is observed the tropical region (30^0S-40^0N) shows discrepancies. The largest spread in the short-term O_3 RF between inventories occurs over northern tropical belt (15-30^0N), where locally the standard deviation reaches 3.0-3.5 mWm^{-2}, over Middle East, Pacific and North Africa.

Table 4 presents the global and annual mean RF (mWm^{-2}) for short-term O_3, CH_4–induced O_3, CH_4, and SWV for a series of inventories. The standard deviation of short-term O_3 values is 1.0 mWm^{-2} (with AEDT and AERO2K resulting in highest and lowest numbers, 14.3 and 11.5 mWm^{-2}, respectively). The CH_4 responses are much more consistent, the standard deviation is 0.2 mWm^{-2} (with values ranging from -7.1 mWm^{-2} for AERO2K and TRADEOFF and -6.7 mWm^{-2} for AEDT). The net aircraft NO_x RF values ranges from 3.6 mWm^{-2} for AEDT, 2.3 mWm^{-2} for REACT4C to 0.2 mWm^{-2} for AERO2K, with 1.2 mWm^{-2} standard deviation.

Myhre et al. (2011) reported GWP values for aviation NO_x emissions using the same aircraft emissions, the same experimental design and a range of five models. The differences in their results constitute a good insight into uncertainties which arise from usage of different global chemistry models. In contrast, the spread in results which are presented in this study gives a measure of differences that arise from usage of different aircraft inventories. The resulting GWP values for three time horizons (20, 100 and 500 years) are given in Table 5. The values show significant differences, which are enhanced with larger time horizons; however, the sign of calculated responses shows a consistently net positive value. The largest differences come from the AEDT and

AERO2K inventories, being 57%, 93% different for 20, 100 time horizons respectively. The increase of discrepancy with larger time horizons can be explained by CH_4, as its response 'remains' for a few decades after NO_x emission. The GWP reduction between a 20 year time horizon and a 100 year horizon is larger for inventories where the CH_4 lifetime reduction is more enhanced, e.g. it is 82% for AEDT, 83% for AEM, 85% for REACT4C, 87% for QUANTIFY and TRADEOFF, 97% for AERO2K. Due to relatively short lifetimes of the net NO_x components the GWP (H=500) differs between inventories only by the CO_2 integral in the denominator.

4 Discussion

Figure 7 shows the vertical profiles of the relative difference of NO_x, O_3, OH and HO_2 responses to that of the AEDT inventory (chosen simply as it gives the largest overall response). The aircraft NO_x perturbation for AEDT occurring at 227 hPa is about 25% greater than that for AERO2K. The difference increases with higher altitudes and constitutes 55% at 100 hPa. On the contrary, AERO2K's NO_x significantly exceeds that of AEDT at mid altitudes, where the difference reaches 500% at 762 hPa. The response of the O_3 for these discrepancies is not the same. It is observed that AERO2K's O_3 response indeed dominates in the low-troposphere region (1000-600 hPa) but only by about 4%, whereas AEDT's dominance at cruise altitudes reaches 25% and 50% at 100 hPa. The same pattern is observed for all inventories, just the scale of differences is not so well pronounced. The linear correlation between additional NO_x and O_3 response is observed in UTLS region, being the strongest at 227 hPa and becoming weaker at higher altitudes, for AERO2K the ratio of O_3 to NO_x is 1 at 227 hPa and 0.9 at 100hPa (e.g. for AEM it is 1 at 227 hPa and 0.7 at 100 hPa, for REACT4C it is 1.5 at 227 hPa and 0.8 at 100 hPa).

Interestingly, the greatest O_3 aircraft perturbation did not introduce the strongest CH_4 reduction, as might be expected from the chemistry. The altitudinal distribution of emissions can hide the explanation: a significant fraction of AERO2K's NO_x emissions occurs in the mid-troposphere. The ~4%'s

dominance of AERO2K's O_3 in 900-700 hPa region significantly changes the oxidizing capacity of the low troposphere (there is more OH by about 30% than for AEDT). Annual mean concentrations of OH and CH_4 and temperature are greater at lower altitudes which catalyse OH production and CH_4 destruction.

Another implication of AERO2K's enhancement of O_3 concentrations at lower altitudes is that this O_3 is not as radiatively efficient as O_3 at higher altitudes (Lacis et al., (1990), Köhler et al., (2008)).

It is worth to mention, that not only the height is important, also the geographical distribution of aircraft NO_x emissions plays a certain role in terms of O_3 RF response. The NO_x emissions from low latitudes have a greater impact on climate forcings than the NO_x emissions from high latitudes (Berntsen et al., (2005), Köhler et al., (2012)) The AERO2K dataset has lower, compared to other inventories, NO_x emissions at cruise altitudes over East Asia and over the Pacific (not shown here), which was also noted by Olsen et al. (2012). Even though certain regional differences in the distribution of aircraft NO_x emissions occur, they are not as powerful as altitudinal discrepancies in terms of O_3 production (based on work to be published).

Figure 8 shows a short-term O_3 RF and net NO_x RF responses normalized to 1 Tg of emitted N reported for a number of model simulations (Stordal et al. (2006), Hoor et al. (2009), Myhre et al. (2011), Hodnebrog et al. (2011, 2012)). The inter-model mean O_3 and NO_x RF values are 22.2 and 5.0 mW m^{-2}/Tg(N) yr^{-1}, respectively. While for short-term O_3 responses most of the model results are placed within a one standard deviation range, the net NO_x RF values constitute a more diverse picture. This can be explained by the inter-study differences in the components taken into account for net NO_x calculation. Myhre et al. (2011) pointed out that the ratio of the CH_4 lifetime change to the O_3 column change is very specific for each model, which also influence the net NO_x numbers. The results produced by MOZART-3 are in good agreement with other studies.

The spread in RF values (20% for O_3 RF and 94% for net NO_x RF) between six aircraft inventories is of a smaller magnitude to that of uncertainties between models; however, it is still significant. Myhre et al. (2011) reported a 36% spread in O_3 RF values and 54% in net NO_x RF values between a set of five different models (note that Myhre et al. (2011) account for the time-history emissions (Grewe and Stenke, 2008) in their long term effects); Hoor et al. (2009) reported a 64% spread in O_3 RF and 89% in net NO_x (net is without SWV) values between five different models and Stordal et al. (2006) showed a 33% spread in O_3 RF and 59% in net NO_x RF (net is a sum of short-term O_3 and CH_4 only) values between three different models. This places the discrepancies between different inventories on the same scale of importance as inter-model differences.

5 Conclusions

The coupled NO_x-O_3-CH_4 system, as affected by aviation NO_x emissions, results in a regional short-term O_3 positive radiative forcing and a global long-term O_3, CH_4 and SWV negative responses. Nonetheless the overall radiative forcing induced by current day emissions of aviation NO_x from 3D CTM MOZART-3 is positive as shown by this study, ranging from 3.6 to 0.2 mW m^{-2}.

By using one model (3D CTM MOZART-3) and a series of aircraft inventories (AEDT, AEM, AERO2K, REACT4C, QUANTIFY, TRADEOFF) scaled to the same global total (2.33 Tg (NO_2)/yr), the sensitivity of O_3 response due to the discrepancies in vertical distribution of aircraft NO_x emissions was investigated. It is observed that the differences in the vertical distribution of aircraft NO_x emissions between inventories, with AEDT and AERO2K being the most different, strongly influence the aircraft short-term O_3 response and consequently the net NO_x effect.

The aviation impact on RF per unit emission of NO_x differs from inventory to inventory and ranges from 5.2 to 0.3 mW m^{-2}/Tg(N) yr^{-1} (for AEDT and AERO2K, respectively). The responses of short-term O_3 RF range from 20.5 to 16.5 mW m^{-2}/Tg(N) yr^{-1} (for AEDT and AERO2K, respectively). It is observed that both, the

498 O_3 and net NO_x RF are greater for inventories with higher peak of perturbation of
499 NO_x concentrations, which is a direct result of the amount of emitted NO_x at
500 higher altitudes.
501
502 The spread between aircraft short-term O_3 RF (20%) and aircraft net NO_x 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
509 **Acknowledgements:** Two anonymous reviewers are thanked for their constructive
510 comments. We thank Bethan Owen, Ling Lim and Emily Gray for help in understanding
511 the aircraft inventory peculiarities and Ole Amund Søvde for helpful remarks on the
512 manuscript. The CAEP/MDG, EUROCONTROL
513 (http://www.inventair.eurocontrol.int/home) and projects: AERO2K
514 (http://cate.mmu.ac.uk/aero2k.asp), REACT4C (http://www.react4c.eu), QUANTIFY
515 (http://www.pa.op.dlr.de/quantify), TRADEOFF (http://www.iac.ethz.ch/tradeoff) are
516 thanked for aircraft datasets. This study was funded by the UK Department of Transport.
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