Aircraft emission mitigation by changing route altitude: A multi-model estimate of aircraft NO$_x$ emission impact on O$_3$ photochemistry

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**Abstract**

The atmospheric impact of aircraft NO$_x$ emissions are studied using updated aircraft inventories for the year 2006, in order to estimate the photochemistry-related mitigation potential of shifting cruise altitudes higher or lower by 2000 ft. Applying three chemistry-transport models (CTM) and two climate-chemistry models (CCM) in CTM mode, all including detailed tropospheric and stratospheric chemistry, we estimate the short-lived radiative forcing (RF) from O$_3$ to range between 16.4 and 23.5 mW m$^{-2}$, with a mean value of 19.5 mW m$^{-2}$. Including the long-lived RF caused by changes in CH$_4$, the total NO$_x$-related RF is estimated to about 5 mW m$^{-2}$, ranging 1–8 mW m$^{-2}$. Cruising at 2000 ft higher altitude increases the total RF due to aircraft NO$_x$ emissions by 2 ± 1 mW m$^{-2}$, while cruising at 2000 ft lower altitude reduces RF by 2 ± 1 mW m$^{-2}$. This change is mainly controlled by short-lived O$_3$ and show that chemical NO$_x$ impact of contrail avoiding measures is likely small.

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**Graphical Abstract**

Aircraft emissions (Tg/year)

**Highlights**

- Multi-model study on the aircraft NO$_x$-related effect on the atmosphere.
- New inventory for aircraft NO$_x$ emissions, representative for year 2006.
- The total RF due to NO$_x$ emissions is found to be about 5 mW m$^{-2}$.
- Shifting cruise altitudes up or down by 2000 ft changes RF by about 2 mW m$^{-2}$.
- Contrail-avoiding measures will likely give small NO$_x$ impact.

**1. Introduction**

In this multi-model study we assess how conceptual changes to aircraft flight altitude can alter the impact of aircraft NO$_x$ emissions on atmospheric gas phase chemistry. Aircraft NO$_x$ emissions have
long been known to affect atmospheric O₃ (e.g. Johnstone, 1971; Hesstvedt, 1974; Hidalgo and Crutzen, 1977), and research on the radiative impacts of aircraft emissions were recently summarised by Lee et al. (2010), later to be revisited by Myhre et al. (2011). Aircraft engines produce different types of exhaust gases and particles. Of gases, NOₓ and H₂O are most important for the reactive gas phase chemistry, affecting O₃ production and loss through the O₃–NOₓ chemistry (Johnstone, 1971; Hidalgo and Crutzen, 1977; Johnson et al., 1992; Schumann, 1997; Dameris et al., 1998; Grewe et al., 2002; Gaus et al., 2006). Emitted particles may affect the atmospheric composition by providing surfaces for heterogeneous reactions (e.g. Lee et al., 2010). Aircraft emissions also produce contrail cirrus (e.g. Burkhardt and Kärcher, 2011; Yi et al., 2012) and CO₂. Such changes will affect the radiative balance, imposing a reactive forcing (RF). However, our focus is on reactive gas-phase chemistry only, more specifically on the effect of NOₓ emissions from aircraft.

The atmospheric impact of emissions of short-lived gases have in general been found to depend strongly on where they are emitted (Berntsen et al., 2006), including aircraft NOₓ emissions (Stevenson and Derwent, 2009; Köhler et al., 2012), in compliance with earlier work showing that radiative forcing from O₃ is strongest in the troposphere and lower stratosphere (UTLS) due to lower temperatures and clean background atmosphere (e.g. Wang and Sze, 1980; Lacis et al., 1990; Hansen et al., 1997; Forster and Shine, 1997). Could relatively simple mitigation strategies for aircraft emissions reduce the atmospheric impact? In light of this question, we assess here two mitigation options using idealised emission inventories in sophisticated climate-transport models (CTM).

During the EU project Reducing Emissions from Aviation by Changing Trajectories for the benefit of Climate (REACT4C), three sets of aircraft emission inventories were produced, to assess the NOₓ impact of shifting aircraft cruise altitudes by one flight level, i.e. representative for typical contrail-avoiding measures.

Related studies were carried out in the project TradeOff (Gauss et al., 2006; Stordal et al., 2006), and recently Frömming et al. (2012) re-investigated TradeOff inventories. Related studies have also been carried out for a possible future stratospheric fleet (Grewe et al., 2007; Pitari et al., 2008; Søvde et al., 2007). Here, we use 3 CTMs and 2 climate-chemistry models (CCM) run in CTM mode. Important improvements from previous studies are the multi-model approach, update to more recent aircraft traffic inventories, increased model resolution, and that all models comprise detailed chemistry in the troposphere and the stratosphere, fully covering the UTLS where most aircraft have their cruise altitude and hence main emissions.

The models are described in Section 2, and the aircraft emission inventories in Section 3. Atmospheric impacts are presented in Section 4 and the changes in RF in Section 5. We summarise the study in Section 6.

2. The models

Five different models are used in this study: MOZART-3, ULAQ-CTM, EMAC QCTM, Oslo CTM2 and Oslo CTM3. A short description of the models and their basic set-ups are presented next. In Supplementary Material (SM) Sect. S1, a model overview table is presented and additional information on annual emission numbers is given. We note that our models are spun up for at least 4 years, which we have found sufficient for this study. Except for ULAQ-CTM, the models are driven by or nudged to real meteorology from the European Centre for Medium-Range Weather Forecasts (ECMWF), as will be explained.

2.1. MOZART-3 CTM

The Model for OZone and Related Tracers, version 3 (MOZART-3) is a three dimensional CTM, comprehensively evaluated by Kinnison et al. (2007) and extensively used for different application studies (Gettelman et al., 2004; Park et al., 2004; Sassi et al., 2004; Liu et al., 2009; Wuebbles et al., 2011; Skowron et al., 2013). The horizontal resolution is T42 (≈ 2.8° × 2.8°) and the vertical domain extends from surface to 0.1 hPa with 60 hybrid layers. Vertical resolution is 700–900 m at aircraft cruise altitudes (250–200 hPa). The transport of chemical compounds as well as hydrological cycle is driven by the meteorological fields from ECMWF Interim 6-h reanalysis (ERA-Interim).

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, convection, boundary layer exchanges and wet removal and dry deposition. Advection of tracers is performed following the numerically fast flux-form semi-Lagrangian scheme of Lin and Rood (1996). Convective mass fluxes are re-diagnosed by MATCH, using the shallow and mid-level convection scheme of Hack (1994) and deep convective transport formulation of Zhang and McFarlane (1995). Related studies were carried out in the project TradeOff (Gauss et al., 2007; Pitari et al., 2008; Søvde et al., 2007). Here, we use 3 CTMs and 2 climate-chemistry models (CCM) run in CTM mode. Important improvements from previous studies are the multi-model approach, update to more recent aircraft traffic inventories, increased model resolution, and that all models comprise detailed chemistry in the troposphere and the stratosphere, fully covering the UTLS where most aircraft have their cruise altitude and hence main emissions.

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2.2. EMAC QCTM

The EMAC QCTM is the global ECHAM/MESSy Atmospheric Chemistry (EMAC) run in a quasi-CTM mode. EMAC is a numerical chemistry and climate simulation system that includes submodels describing tropospheric and middle atmosphere processes and their interaction with oceans, land and human influences (Jöckel et al., 2010). It uses the second version of the Modular Earth Sub-model System (MESSy2, Jöckel et al., 2010). The core atmospheric model is the 5th generation European Centre Hamburg general circulation model (ECHAM5, Roeckner et al., 2003, 2006). Free tropospheric dynamics (up to about 200 hPa) are nudged towards the analysed ECMWF meteorology, and in this study aviation emission interact only on chemical processes, and not with model dynamics (Deckert et al., 2011). A horizontal resolution of T42 with 90 vertical hybrid pressure levels up to 0.01 hPa is used. Vertical resolution is ~660 m at aircraft cruise altitudes.

This results in realistic atmospheric dynamics, and clouds are treated in a standard ECHAM scheme (Sundqvist, 1978; Lohmann and Roeckner, 1996). An aerosol climatology (Tarlé et al., 1984) is used for the calculation of the radiation field and separate climatologies are used in the troposphere (Kerkweg, 2005) and stratosphere for H₂SO₄ (Stratospheric Aerosol and Gas Experiment – SAGE) to provide aerosol surfaces for heterogeneous chemistry. Gas phase chemistry is calculated with the MECCA1 chemistry submodel (Sander et al., 2005) consistently from the surface to the
stratosphere. The applied chemical mechanism includes full stratospheric complexity, but neglects the sulphur and halogen families in the troposphere. It has been evaluated by Jöckel et al. (2006).

Anthropogenic emissions are taken from Lamarque et al. (2010), while biomass burning is from GFEDv3.1. Biomass burning is distributed in one level only (140 m thick). Lightning NO$_x$ is distributed horizontally following Grewe et al. (2001), and vertically following Pickering et al. (1998). A base value of 5 Tg (N) yr$^{-1}$ is applied, for which the meteorological conditions may impose a 5–10% variation on annual basis. Total emissions are listed in SM Sect. S1, where surface NO$_x$ also includes a soil source. Other emissions depending on meteorological conditions are the soil source with a base value of 7 Tg (N) yr$^{-1}$, and the isoprene emissions having a base value of 315 Tg (C) yr$^{-1}$. Also for these the annual variation caused by meteorology is 5–10%.

2.3. ULAQ-CTM

The University of L’Aquila model is a global scale climate-chemistry coupled model (ULAQ-CCM, Pitari et al., 2002; Eyring et al., 2006; Morgenstern et al., 2010) extending from the surface to the mesosphere (0.04 hPa) and operated here in CTM mode. Dynamical data, i.e. velocity stream-function and velocity potential, are provided by the background GCM run in a reference case, with no feedbacks of aviation induced changes. Horizontally, a T21 spectral resolution is used in a 6$^\circ$ $\times$ 5$^\circ$ grid. Vertically the model spans 126 log-pressure levels (~560 m spacing).

The ULAQ-CCM has been fully described in Pitari et al. (2002) and also in Eyring et al. (2006) and Morgenstern et al. (2010) for the SPARC-CCMVal model inter-comparison and validation campaigns. Since then, some important updates have been made in the model: (a) increase of horizontal and vertical resolution to T21 and 126 layers as described above; (b) inclusion of a numerical code for the formation of upper tropospheric cirrus cloud ice particles (Kärcher and Lohmann, 2002); (c) update to Sander et al. (2011) recommendations for cross sections of species, and the parameterisation of Minschwaner et al. (1993) for the Schumann–Runge bands, based on fixed-temperature opacity distribution function formulation; (d) upgrade of the radiative transfer code for calculations of photolysis, solar heating rates and top-of-atmosphere radiative forcing. The oceanic surface temperature is assimilated from the Hadley Centre for Climatic Prediction and Research; the parameterisation of periodic natural forcings (solar cycle, quasi-biennial oscillation) is included on-line.

The chemistry module (Pitari et al., 2002) is organised with long-lived and surface-flux species CH$_4$, N$_2$O, CFCs, HCFCs, CO, NMVOC, NO$_x$ and with all medium and short-lived species grouped in the families O$_x$, NO$_x$, HO$_x$, CHO, Cl$_x$, Br$_x$, SO$_x$ and aerosols. In total there are 40 transported species and 26 species at photochemical equilibrium. For aerosols there are 57 size categories. The model includes the major components of stratospheric and tropospheric aerosols (sulphate, carbonaceous, soil dust, sea salt, PSCs). Aircraft NO$_x$ emissions are emitted as the NO$_x$ family species, and partitioned by applying photochemical equilibria between nitrogen species.

The updated radiative transfer module operating on-line in the ULAQ-CCM, is a two-stream delta-Eddington approximation model (Toon et al., 1989). It is used for chemical species photolysis rate calculations in ultra-violet (UV) to visible (VIS) wavelengths and for solar heating rates and radiative forcing in UV-VIS-near-infrared bands. The ULAQ model calculations of photolysis rates and surface and top-of-atmosphere radiative fluxes have been validated in the framework of SPARC and AEROCOM inter-comparison campaigns (Chipperfield et al., 2013; Randles et al., 2013).

2.4. Oslo CTM3

The Oslo CTM3 (Søvde et al., 2012) is a three dimensional off-line CTM. It is a recent upgrade of Oslo CTM2 (see Section 2.5). The improvements from Oslo CTM2 to CTM3 includes improved transport scheme parameterisation, wet scavenging processes, lightning parameterisation and calculations of photodissociation rates, as described by Søvde et al. (2012). The model is driven by 3-h forecasts generated by the Integrated Forecast System (IFS) of the ECMWF, cycle 36r1. Horizontal resolution is T42, and vertically the model domain spans 60 layers between the surface and 0.1 hPa. Vertical resolution is 700–900 m at aircraft cruise altitudes.

How the forecast data are put together is explained by Søvde et al. (2012). Advection of chemical species is carried out using the improved second order moments scheme (Prather et al., 2008; Søvde et al., 2012). Convective transport of tracers is based on the convective upward flux from the ECMWF model, allowing for entrainment or detrainment of tracers depending on the changes in the upward flux. Detrainment rates are read from the meteorological data, possibly increasing or decreasing the mixing with ambient air as the plume rises (Søvde et al., 2012). Turbulent mixing in the boundary layer is treated according to the Holtslag K-profile scheme (Holtslag et al., 1990).

Oslo CTM3 comprises comprehensive tropospheric chemistry, accounting for the most important parts of the O$_3$–NO$_x$–hydrocarbon chemistry cycle (Berntsen and Isaksen, 1997), the tropospheric sulphur cycle (Berglen et al., 2004), and also comprehensive stratospheric chemistry as explained in Søvde et al. (2008). Also included are tropospheric nitrate aerosols and sea salt aerosols. For the chemistry calculations, the quasi steady state approximation (QSSA) chemistry solver (Hesstvedt et al., 1978) is used.

Surface emissions are taken from RETRO (RETRo Emissions, 2006), except for biomass burning, which is taken from the Global Fires Emission Database version 3 (GFEDv3). Due to a conversion error, the Oslo CTM3 (and Oslo CTM2) assumed GFEDv3 NO$_x$ to be NO$_2$ instead of NO$_x$, giving about 3 Tg (N) yr$^{-1}$ instead of the correct 4.7 Tg (N) yr$^{-1}$. We have not had the possibility to re-run the simulations with correct value, but assume increasing surface NO$_x$ emissions by 3–4% will not change the impact of aircraft emissions noticeably. Emissions of NO$_x$ from lightning is described by Søvde et al. (2012), where the emissions are distributed through the year based on the amount of convection in the meteorological data.

2.5. Oslo CTM2

The Oslo CTM2 (Søvde et al., 2008) is the previous version of Oslo CTM3. It is widely used in previous studies (e.g. Berntsen and Isaksen, 1997; Berglen et al., 2004; Isaksen et al., 2005; Søvde et al., 2007, 2011). The chemistry schemes in Oslo CTM2 and Oslo CTM3 are the same; tropospheric and stratospheric chemistry is included, as is the tropospheric sulphur scheme. Due to computational constraints, CTM2 does not include tropospheric nitrate aerosols and sea salt aerosols in this study. The model resolution is the same as for CTM3, as are the meteorological data used to drive the model.

Compared to CTM3, the CTM2 advection of chemical species is calculated by the older second-order moment method (Prather, 1986). Convective transport is similar to CTM3, but does not use detrainment rates from meteorological data, thus having a somewhat more effective transport to higher altitudes (Søvde et al., 2012). Turbulent mixing is treated as in CTM3, according to the Holtslag K-profile scheme (Holtslag et al., 1990).

Surface emissions are the same as for CTM3. NO$_x$ emissions from lightning differ from CTM3, and are coupled on-line to the
convection in the model using the parameterisation proposed by Price et al. (1997), along with their seasonal variation, as described by Berntsen and Isaksen (1999). In this work, we have updated the vertical distribution of lightning NO\textsubscript{x} emissions to be the same as in Oslo CTM3. The old treatment often placed lightning NO\textsubscript{x} at too high altitudes, even into the stratosphere. Effectively, the new treatment reduces UTLS NO\textsubscript{x}, giving larger impact of the aircraft NO\textsubscript{x} emissions. This update is further discussed in SM Sect. S3.

3. Aircraft emissions

REACT4C aircraft emissions inventories are produced using the FAST global aircraft movements and emissions model, one of the models approved for use by the International Civil Aviation Organization (ICAO)'s Committee on Aviation Environmental Protection (CAEP) Modelling and Database Group (MDG) (ICAO, 2013). FAST has been previously used in various applications such as the impacts of emissions trends (Lee et al., 2009; Owen et al., 2010; Olsen et al., 2013) and NO\textsubscript{x} impacts on atmospheric chemistry (Gauss et al., 2006; Skowron et al., 2013).

The base case inventory is generated from 6 weeks of flight movements data from the year 2006, derived for the CAEP Round 8 (CAEP/8) MDG work programme (ICAO/CAEP, 2009), and found to be representative when extrapolated to an annual basis. Movement data are compiled from radar data of North American and European airspace, which covers ~80% of global civil aviation traffic. The remaining movements from the rest of the world are obtained from the Official Airline Guide (OAG). All flights were assumed to follow great circle trajectories, but with distance (and consequently fuel) correction applied using an empirical formula derived during the CAEP/8 Goals Modeling Work (ICAO/CAEP, 2009). This great circle correction is used to estimate actual distance and fuel burnt. The aircraft emissions are modelled using 42 aircraft types representative of the 2006 global fleet.

Annually averaged inventories are provided on a $1^\circ \times 1^\circ$ horizontal spacing and 610 m vertical spacing (2000 ft, one flight level), assuming standard atmosphere conditions. The latitude-height and horizontal distributions are shown in SM Fig. SF1.

Two possible mitigation inventories are produced, shifting cruise altitudes up and down 2000 ft, respectively, for aircraft types contributing >1% to the base case global fuel and distance flown (21 aircraft types). Fig. 1 shows global numbers of fuel (top) and NO\textsubscript{x} (bottom) for each vertical level of the inventories. While distance flown is approximately 38.9 billion km in all inventories, fuel use is 178.3, 176.9 and 180.7 Tg yr\textsuperscript{-1} in bases case, +2000 ft and –2000 ft, respectively. For NO\textsubscript{x}, the corresponding numbers are 0.710, 0.716 and 0.710 Tg (N) yr\textsuperscript{-1}. The respective increase (1.3%) and decrease (0.8%) in fuel burnt observed for upward and downward shifts are to be expected, as generally the fuel efficiency increases with higher altitude. The opposite decrease (0.01%) and increase (0.8%) in NO\textsubscript{x} emissions occur due to a higher NO\textsubscript{x} emission index (EI) at higher altitudes, while a downward shift to a lesser extent reduces EI(NO\textsubscript{x}).

4. Chemical impact

Before assessing the impact of the mitigation inventories, we quantify the overall chemical responses of aircraft NO\textsubscript{x} emissions by comparing the base case simulations (BASE, BC for short) to simulations without aircraft emissions (NOAIR, NA). The mitigation option of flying higher (PLUS, PL) and lower (MINUS, MI) are then compared to BASE. Radiative forcing is discussed in Section 5.

Several studies have estimated the effect of aircraft NO\textsubscript{x} emissions in the past, as summarised by Holmes et al. (2011). Both 5% perturbations (e.g. Hoor et al., 2009; Hodnebrog et al., 2012) and 100% perturbations (e.g., Gauss et al., 2006) have been applied. The latter gives the overall effect of aircraft, without considering compensating effects from other emission sectors due to chemical non-linearity (Grewe et al., 2010): A change in one NO\textsubscript{x} emission
source (e.g. road traffic) may alter the effect on O3 caused by another source (e.g. industrial emissions). For aircraft emissions, a possible compensating NOx source could be lightning, and to assess how O3 changes induced by lightning NOx are modified by aircraft emissions, a tagging method has to be applied (Grewe et al., 2010; Grewe, 2013). Non-linear chemistry depends on the background of NOx and hydrocarbons (Grewe et al., 2002; Köhler et al., 2008), but is often assumed to increase with larger changes in NOx. Changing the geographic location may contribute to larger non-linearities (Köhler et al., 2008; Stevenson and Derwent, 2009). Grewe et al. (2002) found that aircraft NOx produces O3 more efficiently at lower altitudes, but is counterbalanced by a shorter NOx lifetime due to increased wet removal of HNO3. Our vertical PLUS/MINUS inventories do not change the geographical emission patterns significantly, suggesting their non-linear impact will be relatively small. Non-linear effects in winter are expected to be smaller due to less sunlight. A thorough assessment of the non-linear chemistry following a vertical shift in cruise altitudes has been outside the scope of this work. Our main focus is to identify mitigation possibilities of flying higher or lower, so we apply the 100% perturbation method also for the comparison of BASE against NOAIR, even though this may mask non-linearities.

Generally, NOx contributes to production of O3 in the troposphere and to destruction of O3 in the stratosphere (e.g. Grewe et al., 2002). Because of different transport schemes in the models, the amount of NOx transported to the stratosphere will differ. We will come back to this in the next sections.

A change in NOx sets up a perturbation in OH which again changes the greenhouse gas CH4. For a NOx increase, OH generally also increases, thereby reducing CH4, whose main loss channel is through OH. A CH4 perturbation has a long atmospheric lifetime (~12 years) and sets up a perturbation in O3 having the same lifetime, usually referred to as the primary mode O3. These long-lived changes in CH4 and O3 have the same sign. However, our model runs do not capture the long-lived changes in CH4 and O3, because we use fixed concentrations of CH4 at the model surface. This is a well-known approach to reach chemical steady-state more quickly because the other species have shorter lifetimes than CH4. While the method does not represent the “true” chemical steady-state because the long-lived effects of a CH4 perturbation are missing, the method is still applicable because the radiative effects of CH4 and the primary mode O3 can be easily calculated from CH4 lifetime changes. As will be explained in Section 5, this means that CH4 can be allowed to differ slightly between the models.

To match the inventory year, the models also apply meteorological conditions for the year 2006, either driven directly (MOZART-3, Oslo CTM2, Oslo CTM3) or nudged (EMAC QCTM) by ECMWF meteorology, or as in ULAQ-CTM by using its own GCM winds. A month-by-month comparison may thus be less representative, so we present Jun-Jul-Aug (JJA) and Dec-Jan-Feb (DJF) seasonal averages for all models.

4.1. Base case aircraft NOx emissions

Comparing BASE–NOAIR simulations we show the JJA zonal mean impact on O3 from all models (Fig. 2), where maximum impact range from 4.8 ppb to 8.8 ppb. Winter time (DJF, Fig. 3) impact is smaller due to less photochemical activity, ranging from 3.4 ppb to 4.4 ppb. These impacts are consistent with earlier results (e.g., Hoor et al., 2009). Four features stand out in Figs. 2 and 3, namely an upper stratosphere reduction in MOZART-3 and ULAQ-CTM; larger vertical extent of the JJA ∆O3 response in ULAQ-CTM; low DJF impact on O3 at high latitudes in ULAQ-CTM; and a positive ∆O3 at ~20 hPa in Oslo CTM2, EMAC-QCTM and Oslo CTM3. MOZART-3 and ULAQ-CTM seem to transport aircraft NOx to higher altitudes (SM Figs. SF2 and SF3), indicating more vigorous vertical transport in extra-tropic cruise altitude regions. MOZART-3 has the highest ∆O3, and although being more confined in the vertical at high latitudes, low-latitude ∆O3 reach higher altitudes, suggesting faster upward transport in the tropics. The small DJF high latitude ∆O3 in ULAQ-CTM can be explained by little photochemical activity and more effective downward transport, or possibly by a strong polar stratospheric vortex. Finally, the Oslo CTM2, EMAC-QCTM and Oslo CTM3 increase in O3 at ~20 hPa is caused by reduced back-scattering of UV; more UV is absorbed at cruise altitudes. This is masked by the stratospheric NOx increase in the other models.
Except for being transported to high altitudes in MOZART-3 and ULAQ-CTM, the modelled NO$_x$ = NO + NO$_2$ is largely consistent between the models, with JJA maxima of about 47–64 ppb, except ULAQ-CTM which has about twice this value. In winter the models agree better on maximum values (36–70 ppb), which is shifted to mid-latitudes due to lack of sunlight at high latitudes. In general, the models agree on tropospheric NO$_2$ and the inter-hemispheric transport. Similar features are found in NO$_3$ and all non-HNO$_3$ nitrogen species (SM Figs. SF4–SF7). Larger impacts are accompanied by higher background values of NO$_x$ and NO$_2$ (SM Figs. SF10–SF17), indicating slower removal of atmospheric NO$_x$ in ULAQ-CTM than in the other models. Higher background NO$_x$ could also result in a smaller O$_3$ production efficiency.

Model differences in O$_3$ are thus mainly due to differences in transport or wet scavenging of NO$_x$ or O$_3$ itself. These processes are important for determining the background distributions. The transport differences between the models should be sorted out in the future; it has unfortunately been out of scope of this work. Oslo CTM2 has typically lower background NO$_x$, than Oslo CTM3, thereby having a larger impact of aircraft NO$_x$ on O$_3$ (SM Figs. SF10–SF12 and Figs. SF14–SF16). Such changes are e.g. due to less wet removal of HNO$_3$, or slightly higher winter time NO$_x$ emissions from lightning (Søvde et al., 2012), and should be expected to differ in the other models also.

All models except ULAQ-CTM have the largest impact on NO$_x$ and NO$_x$ in winter when aircraft at high latitudes fly more frequently in the stratosphere due to a lower tropopause, and hence the aircraft emissions are less subject to tropospheric wet removal. The reverse seasonal cycle in ULAQ-CTM is probably due to more removal of HNO$_3$ on ice cirrus during winter, but less wet scavenging during summer.

4.2. Plus 2000 ft aircraft NO$_x$ emissions

The O$_3$ impact of increasing cruise altitude by 2000 ft is shown in Figs. 4 and 5, for JJA and DJF, respectively. In the region of maximum impact, i.e. 30°N–90°N at ~200 hPa, the JJA ΔO$_3$ is about 2 ppb compared to the BASE case. DJF ΔO$_3$ is expectedly smaller due to reduced photochemical activity, giving increases of up to 0.4–1.2 ppb compared to BASE. Otherwise, DJF and JJA show similar features. Oslo CTM2 and Oslo CTM3 have the largest winter time impact on ΔO$_3$, probably due to the lower background NO$_x$ giving more effective O$_3$ production.

Compared to BASE, all models show O$_3$ reductions at higher stratospheric altitudes, with largest impacts for the models having largest stratospheric impact in BASE–NOAIR. An upward shift of cruise altitude reduces tropospheric NO$_x$, and thus slightly O$_3$. This reduction is largest in ULAQ-CTM where it also stretches equatorwards. Oslo CTM2 and CTM3, however, have no visible tropospheric reduction in the JJA zonal mean. On average, our models show an O$_3$ column increase of ~0.03 DU, consistent with previous studies (Gauss et al., 2006; Fröming et al., 2012). An increase is also consistent with Köhler et al. (2008).

In the region of maximum aircraft impact, a dipole structure can be seen in ULAQ-CTM (30°N, 275 hPa, Fig. 4), showing up more clearly in ΔNO$_x$ and Δ(NO$_x$–HNO$_3$) (SM Figs. SF18–SF21). The ΔNO$_x$ dipole is also seen in MOZART-3 and EMAC QCTM in JJA and in all models in DJF. The winter time dipole structures show up more clearly because more emissions are emitted in the stratosphere where the stable air acts to keep the sharp gradients in the vertical. This also applies for the mentioned dipole structures for O$_3$. Although the dipole is tilted differently in ULAQ-CTM, all models are generally consistent when it comes to changes in reactive nitrogen species.

Cruise region NOx changes vary from ~4% to ~10% in summer and ~10% to ~30% in winter (SM Figs. SF24–SF25), suggesting that non-linearities play a small role in summer but may be important in winter. We also note that the small winter ΔNO$_x$ in ULAQ-CTM (SM Figs. SF23 and SF5) is probably due to more efficient HNO$_3$ loss on cirrus ice in winter.

4.3. Minus 2000 ft aircraft emissions

Lowering the cruise altitude by 2000 ft reduces the impact on O$_3$ (Figs. 6 and 7). In the region of maximum impact, summer time ΔO$_3$ is reduced by up to 1–2 ppb compared to the BASE case. This reduction is smaller in winter due to less photochemical activity.

In general, this response is almost opposite of PLUS–BASE. The models show an increase in upper-stratospheric O$_3$, most pronounced in the models where BASE–NOAIR has the strongest stratospheric impact. A slight increase in O$_3$ in the lower
troposphere is seen, least pronounced in Oslo CTM3. The total O3 column is reduced by ~0.02 DU, consistent with previous studies (Gauss et al., 2006; Köhler et al., 2008; Fromming et al., 2012). Corresponding \( \Delta \text{NO}_x \), \( \Delta (\text{NO}_2 - \text{HNO}_3) \) and \( \Delta \text{NO}_y \) are shown in the SM Figs. SF26–SF33; largely showing opposite features compared to PLUS–BASE.

5. Radiative forcing estimates

Short-lived O3 radiative forcings (RFs) have been calculated for all simulations performed in this study, using two different radiative transfer models (RTM) in off-line simulations, namely the Oslo and ULAQ radiative models. We discuss these results before describing the CH4 related forcing. The Oslo RTM uses separate radiative transfer codes for long-wave and short-wave radiation (Myhre et al., 2011), more specifically a broad band code (Myhre and Stordal, 1997) and the multi-stream DISORT code (Stamnes et al., 1988), respectively. Stratospheric temperature adjustment is included in the simulations, to follow the standard definition of radiative forcing (Forster et al., 2007). See SM Sect. S4 for more details.

The ULAQ radiative transfer module is the same as used in the ULAQ-CCM (Section 2.3), and has been validated in the framework of SPARC and AEROCOM inter-comparison campaigns (Chipperfield et al., 2013; Randles et al., 2013). It applies a two-stream delta-Eddington approximation model in the solar spectrum (Randles et al., 2013), and a two-stream delta-Eddington broad band correlated-k distribution model in the long-wave planetary spectrum (Chou et al., 2000). Stratospheric temperature adjustment is performed in the Fixed Dynamic Heating (FDH) approximation (Ramanathan and Dickinson, 1979). See SM Sect. S4 for more details.

Fig. 4. Change in O3 (ppb) due to lifting aircraft emissions by 2000 ft, calculated as the difference between the PLUS minus BASE simulations, for the JJA season. Contours are shown for every 0.2 ppb.

Fig. 5. Change in O3 (ppb) due to lifting aircraft emissions by 2000 ft, calculated as the difference between the PLUS minus BASE simulations, for the DJF season. Contours are shown for every 0.2 ppb.
Annual RFs for short-lived O₃ are listed in Table 1 for all model runs, separated into short wave forcing (SW), long wave forcing adjusted for stratospheric temperature change (LW) and net forcing (Net), showing that the two RTMs agree fairly well, especially for LW. For the BASE–NOAIR, our models produce net annual RF ranging from 16.4 mW m⁻² to 23.5 mW m⁻², with a mean of ~19.5 mW m⁻². MOZART-3 has the smallest RF due to the smallest SW contribution, even though its LW forcing is slightly higher than for ULAQ-CTM. Oslo CTM2 and CTM3 have higher RFs, and differ internally by about 1.3 mW m⁻², mainly caused by LW. Net RF between the RTMs differs by about 0–5%, while LW about 5–15% and SW 15–20%. This is in line with Stevenson et al. (2013).

Annual mean RF differences between the models mainly originate during winter time, while in summer the models agree better (SM Sect. 5.5). Noticeable lower winter time RFs in ULAQ-CTM and partly in MOZART-3 correspond well with their smaller tropical O₃ perturbations in the zonal means, and can also be seen in the horizontal RF distribution (SM Sect. 5.5). This strengthens the indication that ULAQ-CTM and to some extent MOZART-3 have stronger tropical upwelling than the other models. We also note that relative differences in monthly O₃ RF between the RTMs are similar to their annual mean differences.

Scaling to 1 Tg (N) yr⁻¹ emissions, we find the short-lived O₃ RF from aircraft to be 27.4 mW m⁻², comparing well with the 27.3 ± 9.7 mW m⁻² of Holmes et al. (2011). Our results are somewhat higher than the QUANTIFY studies (Myhre et al., 2011; Hodnebrog et al., 2012) giving ~21 mW m⁻², i.e. we have a higher sensitivity per emission. However, because of the differences in spatial and temporal distribution of REACT4C and QUANTIFY emissions, such a direct comparison should be carried out with care (Skowron et al., 2013).

Fig. 6. Change in O₃ (ppb) due to lowering aircraft emissions by 2000 ft, calculated as the difference between the MINUS minus BASE simulations, for the JJA season. Contours are shown for every 0.2 ppb.

Fig. 7. Change in O₃ (ppb) due to lowering aircraft emissions by 2000 ft, calculated as the difference between the MINUS minus BASE simulations, for the DJF season. Contours are shown for every 0.2 ppb.
A 2000 ft higher cruise altitude produces a small RF increase ranging from about 0.5 mW m$^{-2}$ to 2.9 mW m$^{-2}$, with smallest impact in ULAQ-CTM and largest in Oslo CTM2. The largest changes are caused by increased LW, most pronounced during summer months, and the amount of RF largely follows the $\Delta O_3$ in Figs. 4 and 5. Thus the RF differences are related to the differences in transport and background NO$_2$, between the models; emitting more often in the stratosphere, the compensating negative RF at high altitudes is increased.

Flying at altitudes 2000 ft lower consistently changes the sign of LW RF, although absolute values are similar to PLUS vs BASE. A change in sign is also seen for SW RF, except for MOZART-3 where SW RF is zero or slightly positive. The largest contribution to forcing changes is still from LW, and the net RF ranges from about $-0.5$ to $-2.4$ mW m$^{-2}$, compared to the BASE case. As for the PLUS case, CTM2 produces the largest change, closely followed by CTM3. Roughly halving their values, we find the RF from MOZART-3 and EMAC QCTM, and about another halving we get the RF from ULAQ-CTM. The downward shift of aircraft NO$_2$ emissions reduces upper-stratospheric O$_3$ destruction, and also puts a larger fraction of the changes due to different aircraft emission inventories, along with changes due to different aircraft emission inventories, all calculated from modelled monthly means of OH, CH$_4$ and temperature. We find that BASE–NOAIR produces an RF for CH$_4$ ranging from $-10.7$ mW m$^{-2}$ to $-7.1$ mW m$^{-2}$ (Table 3). Increasing cruise altitudes leads to 0–0.2 mW m$^{-2}$ increase in this RF, while decreasing cruise altitudes decrease it by 0.1–0.4 mW m$^{-2}$.

Table 1

<table>
<thead>
<tr>
<th>Model</th>
<th>MOZART-3</th>
<th>ULAQ-CTM</th>
<th>EMAC QCTM</th>
<th>Oslo CTM2</th>
<th>Oslo CTM3</th>
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<tr>
<td>SW</td>
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<td>4.51</td>
<td>4.74</td>
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<tr>
<td>LW</td>
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<td>4.24</td>
<td>3.88</td>
<td>3.97</td>
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<tr>
<td>Net</td>
<td>12.9</td>
<td>12.5</td>
<td>14.0</td>
<td>16.4</td>
<td>17.1</td>
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<tr>
<td>SW–NOAIR</td>
<td>3.52</td>
<td>4.93</td>
<td>4.51</td>
<td>4.74</td>
<td>5.28</td>
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<tr>
<td>LW–NOAIR</td>
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<td>4.24</td>
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<td>12.5</td>
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Table 2

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<td>0.0010</td>
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<tr>
<td>Oslo CTM3</td>
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<td>-0.087</td>
<td>0.0000</td>
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</table>

will change stratospheric water vapour and give an additional RF of 15% of CH$_4$ RF (Myhre et al., 2007).

The total annual RFs due to our 0.71 Tg (N) yr$^{-1}$ aircraft NO$_x$ emissions are listed in Table 4, giving mean RF of $-5$ mW m$^{-2}$, with a range of 1–8 mW m$^{-2}$. Cruising at the 2000 ft higher altitude increases this RF by about 2 ± 1 mW m$^{-2}$, while cruising at the 2000 ft lower altitude reduces it by 2 ± 1 mW m$^{-2}$.

6. Summary and conclusions

This work comprises a multi-model assessment of the aircraft NO$_x$ emission impact on the atmosphere for the contrail-avoiding mitigation possibilities of shifting the main cruise altitudes up or down one flight level. Updated aircraft emission inventories based on year 2006 aircraft movements have been applied, and the models comprise comprehensive tropospheric and stratospheric chemistry to fully cover the UTLS.

The chemical O$_3$ perturbation and sensitivity to the altitude of aircraft NO$_x$ emissions are consistent with earlier studies (e.g., Gauss et al., 2006; Köhler et al., 2008; Frömming et al., 2012).

We find the short-lived O$_3$ changes due to 0.71 Tg (N) yr$^{-1}$ of aircraft NO$_x$ emissions to cause an RF of 19 ± 3 mW m$^{-2}$, or 27 ± 5 mW m$^{-2}$ when scaled to 1 Tg (N) yr$^{-1}$, matching Holmes et al. (2011) well. Our RF from CH$_4$ is in the lower range of Holmes et al. (2011), while RF from primary mode O$_3$ is somewhat higher due to new IPCC recommendations; previously the RF of the primary mode was estimated to be about 30% of the CH$_4$ RF, but has now been updated to 50% (Myhre et al., 2013).

Accounting for CH$_4$ related changes we find a total RF due to aircraft NO$_x$ of about 5 mW m$^{-2}$, with a range of 1–8 mW m$^{-2}$. The lower end of this range is due to one model giving a stronger effect on CH$_4$, and without this model the range would be about 4–8 mW m$^{-2}$, with a mean of 6 mW m$^{-2}$.

Shifting cruise altitudes, and hence aircraft NO$_x$ emissions, 2000 ft upwards increases the total RF due to gaseous photochemistry by about 2 ± 1 mW m$^{-2}$. Similarly, a downward shift of 2000 ft reduces the RF by about 2 ± 1 mW m$^{-2}$. Scaled to 1 Tg (N) yr$^{-1}$, this amounts to about 2.5 ± 1.5 mW m$^{-2}$. These changes are mainly driven by short-lived O$_3$, of which they represent about 10%.

Table 3

<table>
<thead>
<tr>
<th>Model</th>
<th>BC–NA</th>
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<th>MI BC</th>
</tr>
</thead>
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<tr>
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<td>EMAC QCTM</td>
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Table 4

<table>
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<td>MOZART-3</td>
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<tr>
<td>ULAQ-CTM</td>
<td>-7.198</td>
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<td>EMAC QCTM</td>
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<tr>
<td>Oslo CTM2</td>
<td>-8.970</td>
</tr>
<tr>
<td>Oslo CTM3</td>
<td>-9.398</td>
</tr>
</tbody>
</table>

RF long-lived O$_3$ = 0.5 RF(CH$_4$)

RF stratospheric H$_2$O = 0.15 RF(CH$_4$)
We have not assessed the potential of aircraft aerosol emissions to affect the radiative budget. Aerosols may absorb or scatter light, and also have an impact on NOx by providing surfaces for conversion to HNO3. The latter effect is small and reduces the aircraft-induced O3 by ~10% (Iachetti et al., 2014, personal communication, in preparation).

Burkhardt and Kärcher (2011) found AIC RF to be about 31 mW m−2, and Myhre et al. (2013) suggest 50 mW m−2. While Forringer et al. (2012) and Fichter et al. (2005) studied cruise altitude shifts and found linear contrail RF changes smaller or however, that CO2 is still important for the total climate impact of starting the work on MOZART-3.

number ACP8-GA-2009-233772. We acknowledge Emily Gray for results.

A contrail study could in principle be set up for our inventories, but this has been outside the scope of our work. Also, there are very few models calculating aerosol and cirrus effects; building a multi-model comparison for these would increase the confidence in their results.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.06.049.

References


