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Aircraft pollution: a futuristic view

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Abstract

Impacts of NO_x, H_2O and aerosol emissions from a projected 2050 aircraft fleet, provided in the EU project SCENIC, are investigated using the Oslo CTM2, a 3-D chemical transport model including comprehensive chemistry for the stratosphere and the troposphere. The aircraft emission scenarios comprise emissions from subsonic and supersonic aircraft. The increases in NO_y due to emissions from the mixed fleet are comparable for subsonic (at 11–12 km) and supersonic (at 18–20 km) aircraft, with annual zonal means of 1.35 ppbv and 0.83 ppbv, respectively. H_2O increases are also comparable at these altitudes: 630 and 599 ppbv, respectively. The aircraft emissions increase tropospheric ozone by about 10 ppbv in the Northern Hemisphere due to increased ozone production, mainly because of subsonic aircraft. Supersonic aircraft contribute to a reduction of stratospheric ozone due to increased ozone loss at higher altitudes. In the Northern Hemisphere the reduction is about 39 ppbv, but also in the Southern Hemisphere a 22 ppbv stratospheric decrease is modeled due to transport of supersonic aircraft emissions and ozone depleted air. The total ozone column is increased in lower and Northern mid-latitudes, otherwise the increase of ozone loss contributes to a decrease of the total ozone column. Two exceptions are the Northern Hemispheric spring, where the ozone loss increase is small due to transport processes, and tropical latitudes during summer where the effect of subsonic aircraft is low due to a high tropopause. Aerosol particles emitted by aircraft reduce both aircraft and background NO_x, more than counterweighting the effect of NO_x and H_2O aircraft emissions in the stratosphere. Above about 20 km altitude, the NO_x (and thus ozone loss) reduction is large enough to give an increase in ozone due to aircraft emissions. This effect is comparable in the Northern and Southern Hemisphere. At 11–20 km altitude, however, ozone production is reduced due to less NO_x. Also $ClONO_2$ is increased at this altitude due to enhanced heterogeneous reactions (lowered HCl), and ClO is increased due to less NO_x, further enhancing ozone loss in this region. This results in a 14 ppbv further reduction of ozone. Mainly, this results in an increase of the total ozone column

due to a decrease in ozone loss caused by the NO_x cycle (at the highest altitudes). At the lowermost latitudes, the reduced loss due to the NO_x cycle is small. However, ozone production at lower altitudes is reduced and the loss due to ClO is increased, giving a decrease in the total ozone column. Also, at high latitudes during spring the heterogeneous chemistry is more efficient on PSCs, increasing the ozone loss.

1 Introduction

Air traffic is continually increasing. According to the Global Market Forecast 2004–2023 (Airbus, 2004), revenue passenger kilometres will increase by 5.3% annually in this period, while freight tonne kilometres (airfreight) will increase by 5.9% annually. Although less fuel-efficient aircraft eventually will be replaced with new aircraft, the amount of burnt fuel will increase. A further increase in air traffic is assumed to take place after 2020, and by 2050 new high speed civil transport aircraft (so-called supersonic aircraft) are suggested to be developed to meet the demands for faster transport. Subsonic aircraft fly slower than the speed of sound, and at altitudes lower than about 12 km, while supersonic aircraft fly faster than the speed of sound and at altitudes up to almost 20 km. In the EU project SCENIC (Scenario of aircraft Emissions and Impact studies on Chemistry and climate, Rogers, 2005), several possible aircraft scenarios for 2050 were developed by Airbus, covering both commercial and non-commercial traffic.

Cruising at 16–18 km altitude, supersonic aircraft are mostly located well above the tropopause, implying that aircraft will emit directly into the stratosphere – a region of more efficient accumulation of pollution. Subsonic traffic cruise at 8 to 12 km, so their emissions often take place in the lower stratosphere at high latitudes, especially during winter when the tropopause is low.

Emissions from aircraft include ozone precursors (e.g. NO_x) as well as aerosol particles, which all affect atmospheric chemistry, e.g. ozone production and destruction. Also components important for climate, such as H_2O and CO_2 are emitted from aircraft. Several studies focusing on aircraft NO_x emissions have been carried out (Hidalgo and

Crutzen, 1977, Johnson et al., 1992, Schumann, 1997, Dameris et al., 1998, Grewe et al., 2002, Gauss et al., 2006), most of them addressing the impact of subsonic aircraft. The atmospheric impact of aircraft emissions has also been thoroughly addressed by e.g. Fabian and Krcher (1997), Brasseur et al. (1998), Isaksen (2003). This study is an extension of previous studies done within the EU projects TRADEOFF, CRYOPLANE and SCENIC. We focus on the atmospheric impact of a future fleet comprising sub- and supersonic aircraft (referred to as "mixed fleet"). In Sect. 2 the model setup and the emission scenarios are described, and in Sect. 3 the atmospheric impact of NO_x and H_2O aircraft emissions is presented. The impact of aerosol emissions from a mixed fleet is studied in Sect. 4.

2 Model setup and aircraft emissions

A new version of the Oslo CTM2 global chemical transport model (described by Søvde et al., 2007[1], and briefly by Gauss et al., 2006) is applied. For transport processes the highly accurate and non-diffusive Second Order Moments scheme (Prather, 1986) is applied for advection, the Tiedke mass flux scheme (Tiedke, 1989) is used for deep convection, and boundary layer mixing is treated according to the Holtslag K-profile scheme (Holtslag et al., 1990). The model includes comprehensive schemes for tropospheric and stratospheric chemistry, and is designed to study the upper tropopsphere and lower stratosphere (UTLS), the region covering the cruising altitudes of aircraft. The numerical integration of chemical kinetics is done applying the Quasi Steady State Approximation (QSSA) (Hesstvedt et al., 1978), using three different integration methods depending on the chemical lifetime of the species. In particular, the new model version also includes a new heterogeneous chemistry scheme. The spectral resolution applied is T21 (5.6×5.6 degrees) in 40 vertical layers, stretching from the surface to

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2hPa (pressure center of the top model layer at 10 hPa).

The natural and industrial emissions are taken from the EU project POET (Precursors of Ozone and their Effects in the Troposphere) (POET report, 1999). The model is set up for simulating the year 2050 according to the SCENIC project (Rogers, 2005), where the industrial emissions of NO_x, CO and volatile organic compounds (VOC) are scaled up from the year 2000 by 2.2, 1.53 and 1.55, respectively. As the initial chemical conditions, we use a steady-state atmospheric composition from a previous 2050 study, including lowered CFC loading (according to World Meteorological Organization, 2003), and higher methane (scaled up to ~2549 ppbv at the surface) and N_2O (~372 ppbv at the surface) loadings. Also emissions of stratospheric species were set to 2050 level, as were the model upper boundary conditions for all species (taken from the Oslo 2-D model described by Stordal et al., 1985). Otherwise, natural emissions are set to the level of year 2000. In the Oslo CTM2, we calculate stratospheric water vapor by assuming that the H_2 content in water vapor, methane and hydrogen gas is constant ($H_{sum} = H_2O + 2CH_4 + H_2$). H_{sum} is set to 8.598 ppmv excluding aircraft emissions of H_2O. The meteorological data is taken from the European Centre for Medium-Range Weather Forecasts (EMCWF) Integrated Forecast System for the year 2000.

Starting from the steady-state conditions from a previous 2050 study, a spin-up of 7 years without aircraft emissions was performed, followed by additional six-year runs including each of the aircraft emission scenarios. The aircraft emission inventories studied in this work are listed in Table 1, and are given in 1x1 horizontal resolution and 305 m vertical resolution. These data are interpolated into the Oslo CTM2 grid each model time step.

Based on a predicted increase in non-commercial as well as commercial aircraft traffic, emissions from the most likely mixed fleet is given in MIX. Aircraft emissions in this study include NO_x and H_2O, omitting other products of fuel combustion such as CO_2, CO, different hydrocarbons and sulfur oxides; although CO_2 is an important greenhouse gas, the lifetime is too long to distinguish the aircraft effect from other sources, and increases of the other combustion products are low compared to the background

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concentrations, hence they are expected to have a small atmospheric impact (Fabian and Krcher, 1997).

NO_x in the troposphere produces ozone in the presence of peroxy radicals and sunlight, while in the stratosphere it causes ozone depletion through the catalytic NO_x cycle. H_2O is an important source of the OH radical ($H_2O + O^1D \rightarrow 2OH$), which contributes to ozone loss. In this study we focus on the total atmospheric impact of NO_x and H_2O emissions. Due to the high background concentration of H_2O in the troposphere, the effects of H_2O emissions in the troposphere are negligible. However, in the stratosphere H_2O is very low, and therefore aircraft emissions have a larger effect on the OH formation.

Scenario AERO is similar to MIX, except that aircraft aerosol emissions are included. Aerosols offer surfaces for heterogeneous chemistry, which is important for NO_x chemistry. The surface area density (SAD) of these aerosols was included in the model as a transported tracer with SAD-emissions calculated according to Danilin et al. (1998),

$$SAD = \frac{3\mu_f \rho_a \xi EI(S) \mu_{H_2SO_4}}{r \rho_s w_{H_2SO_4} \mu_S} \quad (1)$$

where ρ_s=1.5 gcm^{-3} is the density of sulfur, ρ_a is the density of air, $w_{H_2SO_4}$=50% is the weight percent of sulfuric acid and μ_S and $\mu_{H_2SO_4}$ are the molecular weights of sulfur and sulfuric acid, respectively. The particle radius is assumed to be r=10 nm, and the emission index of sulfur is EI(S)=4×10^{-4}. The fuel mixing ratio (μ_f) is calculated from the MIX NO_x emissions applying the EI(NO_x) for supersonic aircraft (4.6 g/kg). Even though subsonic aircraft have a higher emission index for NO_x and may result in a too low SAD value for the subsonic flight levels, the application of the chosen emission index is justified by the uncertainty of EI(S) and the conversion factor (ξ=0.05) of sulfur to sulfate, and the fact that the particles will be most important in the stratosphere. To simulate washout of emitted species, a method suggested by Danilin et al. (1998) is used, where emitted gases and aerosols are removed with e-folding time of 5 days below 400 hPa. In the top model layer, an e-folding time of 1 year is applied to simulate

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transport and mixing upwards. In this study, sedimentation of aircraft aerosols is not considered, but will be discussed in Sect. 5. Aircraft aerosol emissions are superimposed on the Oslo CTM2 background aerosols, which are taken from a monthly zonal mean SAD climatology derived from various satellite measurements, primarily SAGE II according to Thomason et al. (1997), and data for the year 1999 is applied. A sensitivity study of the MIX scenario without background aerosols (noBACK) is also carried out.

3 Impact of emissions from a mixed fleet

Subsonic and supersonic aircraft cruise at different altitudes. The subsonic fleet is larger than the supersonic fleet, giving the largest contribution to aircraft NO_x emissions. This is clearly seen from Fig. 1, showing the annual zonal mean change in reactive nitrogen (NO_y) with maximum increase at 11km, and a secondary maximum at about 18 km. Clearly, the secondary maximum is due to supersonic aircraft, while the 11 km maximum is due to subsonic aircraft. The seasonal variation of the monthly zonal mean maximum due to subsonic aircraft ranges from 1.1 in September to 1.9 ppbv in May (Fig. 2). During winter and spring, a larger fraction is emitted into the stratosphere than during summer months due to a lower tropopause, increasing the impact of subsonic aircraft. The higher tropopause in summer is accompanied by relatively strong convection, mixing and washout at mid-latitudes, leading to a reduction in the NO_y increases in the mid-latitude troposphere. This causes the maximum impact of aircraft emissions to shift northward (Kraabøl et al., 2002). A similar latitude shift of the maximum value is reported by Gauss et al. (2003).

Although the contribution of supersonic aircraft to the zonal mean change is much smaller than for subsonic aircraft, the maximum NO_y changes due to supersonic aircraft are locally comparable with subsonic aircraft emissions, especially in the summer months. Supersonic flights are confined mostly to the North Atlantic corridor, and this small zonal extent results in the slightly lower zonal mean maximum shown in Fig. 1.

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Aircraft emissions of H_2O contribute to an annual zonal mean increase of up to 624 ppbv above the background H_2O (Fig. 3), ranging from 609 to 833 ppbv in the monthly mean. The maximum change in monthly zonal mean H_2O due to aircraft is mainly located at 11km altitude, and is largest during summer when the maximum shifts to higher altitudes. As seen before, this shift is caused by higher tropopause levels extending transport and washout processes to higher altitudes.

The annual zonal mean effect of aircraft emissions (NO_x and H_2O) on ozone is given in Fig. 4, showing a maximum of 10ppbv at 10km and a minimum of –39 ppbv at 24 km. From month to month, these values vary from 7.2 to 18.4 ppbv (5.5 to 8.9%) and from –48.8 to –31.4 ppbv (–1.8 to –0.8%), respectively (Fig. 5). The maximum variation largely follows the variation of the NO_y maximum, but at an altitude of 9–10 km. This means that the maximum ozone production is located at a lower altitude than the maximum of emissions. Unlike NO_y and NO_x, the maximum ozone increase is shifted northwards for a longer time period in spring and summer, partly due to a higher tropopause and more washout processes at mid-latitudes. Also important for this shift is the increased amount of sunlight at high latitudes during spring and summer, causing more ozone production even for moderate NO_x increases.

NO_x contributes to ozone production in the presence of sunlight and peroxy radicals. At some altitude (the turn-over point), the amount of these radicals is too low for ozone production, and the catalytic NO_x cycle takes over, reducing ozone. The zero line of Fig. 4 does not represent the turn-over point, since transport processes also affect this figure: Downward transport of ozone depleted air will lower the altitude of the zero line. H_2O emissions, however, contribute to ozone destruction at all altitudes through the OH chemistry. Looking at Fig. 5, the monthly variation of maximum ozone depletion due to aircraft resembles the variation of maximum production. The maximum depletion is located at about 30–40 N, around 24km, except during summer months, when a shifting northwards is seen, accompanied by a lowering of the maximum depletion to about 20 km altitude. This shift is due to stratospheric transport processes and more sunlight at higher latitudes. Being located above the NO_x and NO_y maxima, the max-

imum ozone depletion is caused by upward transport of NO_x and H_2O, followed by more effective depletion at higher altitudes. Most of this ozone depletion is due to supersonic aircraft; only a small fraction originates from the upward transport of subsonic aircraft emissions. Mixing of tropospheric (ozone increased) and stratospheric (depleted ozone) air in a mixed fleet scenario will therefore lower the tropospheric ozone increase due to subsonic aircraft.

Interhemispheric transport of stratospheric aircraft emissions is clearly seen in Figs. 1 and 3, in contrast to transport of subsonic aircraft emissions at lower altitudes, which is negligible (not shown). The high altitude transport will affect ozone in the Southern Hemisphere although emissions occur in the Northern Hemisphere.

When affecting ozone, aircraft emissions may also alter the total ozone column. The changes in daily zonal averages of the total ozone columns for the last two simulated years are shown in Fig. 6, ranging from –1.56 DU to 1.65 DU. The increase in the total column is located in the area of highest flight density. At higher latitudes, however, the supersonic aircraft contribute more to ozone loss, showing a net decrease in the total column, except during spring and summer months where the ozone production maximum is shifted northwards, overriding the ozone loss at higher altitudes. The ozone decrease in the Southern Hemisphere is mainly due to cross-hemispheric transport of emissions from supersonic aircraft, and to some extent transport of ozone depleted air. From August to October, there is also a decrease in total ozone in the tropical region. This is because the tropospheric ozone production due to aircraft emissions is small due to a high tropopause and increased washout of NO_y during these months, causing the stratospheric depletion to dominate.

A sensitivity study omitting aircraft emissions of H_2O (scenario noH2O) reveals that ozone changes due to aircraft emissions are mainly caused by NO_x. This is carried out looking at the MIX – noH2O difference. Emissions of H_2O give maximum ozone losses ranging from 6.7 to 11.4 ppbv in the monthly zonal mean (not shown). The maximum effect of H_2O on ozone is generally located at 25–30 N and 24 km height with values ranging from 6 to 7 ppbv. However, the largest decrease in ozone is found in October–

November due to PSC chemistry taking place at about 18 km at the South Pole. The ozone reduction due to aircraft emissions of H_2O is most prominent in the stratosphere, although there is some ozone reduction caused by H_2O in the troposphere. At the highest latitudes around 24 km altitude and during summer, H_2O emissions slightly enhance heterogeneous chemistry on background aerosols, reducing NO_x and thereby increasing ozone.

4 Impact of aerosol emissions from a mixed fleet

Heterogeneous reactions may take place on sulfate aerosols converting NO_x into HNO_3 – reactions being most effective at low temperatures. Depending on the altitude of this conversion, the resulting reductions of NO_x will either reduce the production of ozone in the troposphere and lowermost stratosphere, or reduce the loss at higher altitudes. However, the formation of PSCs and chlorine activation may be enhanced by a reduction of NO_x.

Calculating the aircraft aerosol SAD emissions based on the method of Danilin et al. (1998), we find an annual zonal average SAD maximum of 4.7 $\mu m^2/cm^3$ (Fig. 7), the monthly zonal mean maximum varying steadily through the year (Fig. 8), ranging from 3.8 $\mu m^2/cm^3$ in September to 6.2 $\mu m^2/cm^3$ in May. As for NO_y emissions, the largest values are found in the Northern Hemispheric winter and spring at about 47 N and 11 km height, also shifting slightly to the north during summer and autumn because of the meteorological conditions. Although the maximum value decreases during summer conditions, the surface area density values in the stratosphere increase due to upward transport. The tropospheric reduction of NO_x due to aerosol emissions is largest in spring, while the stratospheric reduction is largest in autumn. Hence, the largest reduction of ozone production due to aerosol emissions from aircraft occurs during spring, while the largest reduction of ozone loss at higher altitudes takes place in autumn.

The heterogeneous conversion of NO_x to HNO_3 will also occur on background aerosols, similarly affecting the production and loss of ozone. In Fig. 9 we see the

annual zonal mean effect of aircraft aerosol emissions on ozone (AERO – MIX), superimposed on the effect of background aerosols (MIX – noBACK), for latitudes between 30 and 60 degrees in (a) the Northern and (b) Southern hemisphere. Most prominent is the increase in ozone between 18 and 30 km due to ozone loss reduction, where the effect of aircraft aerosols is comparable with the background aerosols. For altitudes between 12 and 20 km aircraft aerosols contribute to an ozone reduction through reduced production and a slight increase in ozone loss due to more PSCs or supercooled ternary solutions (STS).

The amount of PSCs is dependent on HNO_3 and H_2SO_4, where the latter is calculated from the amount of background aerosols available (described by Søvde et al., 2007[1]). Thus an increase in HNO_3 and H_2SO_4 due to aircraft aerosol emissions leads to a slight increase of PSCs and therefore an increase in ozone loss. Also, a reduction in NO_x will result in more active chlorine available for depleting ozone. These effects are apparent at altitudes between 12 and 20 km.

The maximum increase of ozone for a mixed fleet including aerosol emissions (Fig. 9), has a larger magnitude than the maximum decrease due to NO_x and H_2O emissions only (Fig. 4). Aircraft aerosols give a reduction of NO_x through heterogeneous reactions, reducing the loss of ozone due to the NO_x cycle. The increase in aerosol SAD also intensify the heterogeneous conversions, lowering the concentration of HCl and thus the heterogeneous loss of $ClONO_2$. In this way more NO_x is bound up in $ClONO_2$, while also the ClO is increased, since less NO_2 is available for forming $ClONO_2$. The same applies for $BrO/BrONO_2$. Both NO_x and ClO are responsible for ozone loss, so by looking at Fig. 10 we see that above 18–20 km the NO_x cycle is more important for ozone than the ClO – giving an increase of ozone due to reduced NO_x. From 18–20 km and down to about 11–12 km (lower at the Southern Hemisphere), the turn-over point where NO_x starts to produce ozone will be reached. However, enhaned ClO and BrO increase the ozone loss in addition to the reduced ozone production due to lower NO_x. Below 11–12 km the NO_x reduction due to aerosols is not large enough to overcome the aircraft emissions of NO_x, resulting in increased ozone production

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there (Fig. 10). Therefore, a mixed fleet with aerosol emissions results in an increase in the daily zonal mean total column at most latitudes (Fig. 11; AERO minus noAC). At the lowermost latitudes through the year, and at the highest latitudes during summer, the reduced ozone production and enhanced loss due to ClO cause a decrease in ozone. The ozone column reduction at high latitudes during spring is due to increased heterogeneous chemistry on PSCs and therefore increased ozone loss due to ClO.

Separating the effect of aircraft aerosols, by comparing the AERO scenario to the mixed fleet scenario (MIX), aircraft aerosols reduces the total ozone column in spring, and increase it in autumn (Fig. 12). Again, the decrease is due to reduced ozone production below the turn-over point (about 20 km), combined with increased ozone loss by ClO and due to heterogeneous reactions on PSCs/STS. The increase is due to ozone loss decrease at higher altitudes (Fig. 13). The decrease in ozone column near the North Pole during May through June is due to more ozone loss below 20 km (due to ClO increase and reduced ozone production) than the ozone loss is decreased above 20 km (due to NO_x). Aerosols are also transported into the Southern Hemisphere, readily reducing NO_x there as well, giving similar results, however smaller in magnitude. The cross-hemispheric transport is most effective above 18km, mainly resulting in reduced ozone loss. However, ClO is also increased here, to some extent enhancing the ozone loss below 18 km. The ozone loss is also increased in spring when heterogeneous chemistry on PSCs/STS is enhanced, increasing the Ozone Hole depth.

A more intriguing effect is a slight increase in stratospheric H_2O due to aircraft aerosols (not shown). The annual zonal mean maximum increase in H_2O after six years of aircraft emissions is 7.3 ppbv (6.2 to 8.4 ppbv in the monthly zonal mean). This is due to heterogeneous reactions on the aerosols, either producing H_2O directly, or increasing the amount of chlorine which may react with methane: Because the stratospheric H_2O in the model is calculated assuming constant total hydrogen, a decrease in methane will increase the amount of H_2O. Further tests to separate these effects are left for another study since we have shown that the change in ozone due to aircraft emissions is mostly due to NO_x.

5 Discussion and conclusions

We have studied a projected future aircraft fleet for the year 2050, presenting the effects of NO_x, H_2O and particle emissions from aircraft. Emphasis is put on the uppermost troposphere and lowermost stratosphere with respect to a stratospheric aircraft fleet.

An increase in subsonic aircraft increases tropospheric and lower stratospheric ozone in agreement with previous studies, although with a larger impact (7 to 18 ppbv ozone increase in the monthly zonal mean) compared to studies of the present fleet (3 to 8ppbv, Gauss et al., 2006). Supersonic aircraft will contribute to ozone depletion in the stratosphere (31 to 49 ppbv in the monthly zonal mean), so that the mixed fleet reduces the total ozone column. In the Southern Hemisphere the effect on ozone is due to cross-hemispheric transport of aircraft emissions and ozone depleted air, and is therefore located at high altitudes, reducing the total ozone column. At northern high latitudes, aircraft emissions cause an increase in the total ozone column in spring and a decrease in autumn.

The maximum contribution of H_2O emissions from a mixed fleet to stratospheric ozone depletion ranges from 6.7 to 11.4 ppbv in the monthly zonal mean, where the largest effect is an indirect effect through heterogeneous chemistry in October and November. In the troposphere there is a slight decrease in ozone due to H_2O, some of which may also be due to transport of ozone depleted air from above when including H_2O emissions.

When introducing aircraft aerosol emissions, both background and emitted NO_x are reduced due to heterogeneous chemistry. Also HCl is reduced due to this, increasing $ClONO_2$ and ClO. The latter is also increased due to less NO_x. In this way ozone loss due to NO_x is reduced (above 20 km), while ozone loss due to ClO is enhanced in the region between 11 and 20 km (and a little higher at the lowermost latitudes). Depending on the location of emitted aerosols, one of these processes may dominate over the other, contributing to an increase (less NO_x) or reduction (more ClO) of the total ozone column. To some extent aircraft aerosols will also reduce tropospheric NO_x

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and therefore ozone production, contributing to a reduction of the total ozone column. The total ozone column is further decreased by an increase in ozone loss due to PSCs: an increase in the surface area density of aerosols converts more NO_x to HNO_3 and the reduction of NO_x activates more chlorine, increasing ozone loss due to PSCs. During summer and autumn, however, ozone loss reduction at higher altitudes due to aircraft aerosols is most prominent, giving an increase of the total ozone column.

It is clear that NO_x and H_2O emissions from supersonic aircraft contribute to a decrease in total ozone, while lower flying subsonic aircraft contribute to an increase in total ozone. Ozone loss south of 30 S is due to cross-hemispheric transport of supersonic aircraft emissions and to some extent ozone depleted air. North of 45 N ozone loss dominates over ozone production at lower altitudes, except during spring and early summer when transport processes and the amount of sunlight increase the ozone production. Given aerosol emissions from aircraft, stratospheric NO_x is reduced by more than what is emitted from aircraft, effectively increasing the ozone column by lowering the ozone loss.

It should be noted that in this study we have assumed no sedimentation of aircraft aerosols. This is probably not a realistic assumption, and if included, the effect of the calculated aerosol surface area density would be smaller. It is then possible that the ozone increase due to reduced NO_x would be smaller, and the ozone decrease due to ClO unchanged, possibly resulting in a decrease in the total ozone column. Nevertheless, aircraft aerosol emissions seem to be important when assessing the impact of aircraft in the stratosphere, where the ozone loss due to NO_x is reduced, but where heterogeneous reactions indirectly increase the ozone loss due to ClO and BrO.

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Table 1. The aircraft scenarios used in this study.

Scenario	Fleet	NO_x [Tg/year]	H_2O [Tg/year]	Aerosol emissions
noAC	No aircraft	0	0	no
MIX	Sub- and supersonic (mixed) fleet	7.445	901	no
AERO	MIX with aerosol emissions	as MIX	as MIX	yes
noH2O	MIX without H_2O emissions	as MIX	0	no
noBACK	MIX without background aerosols	as MIX	as MIX	no

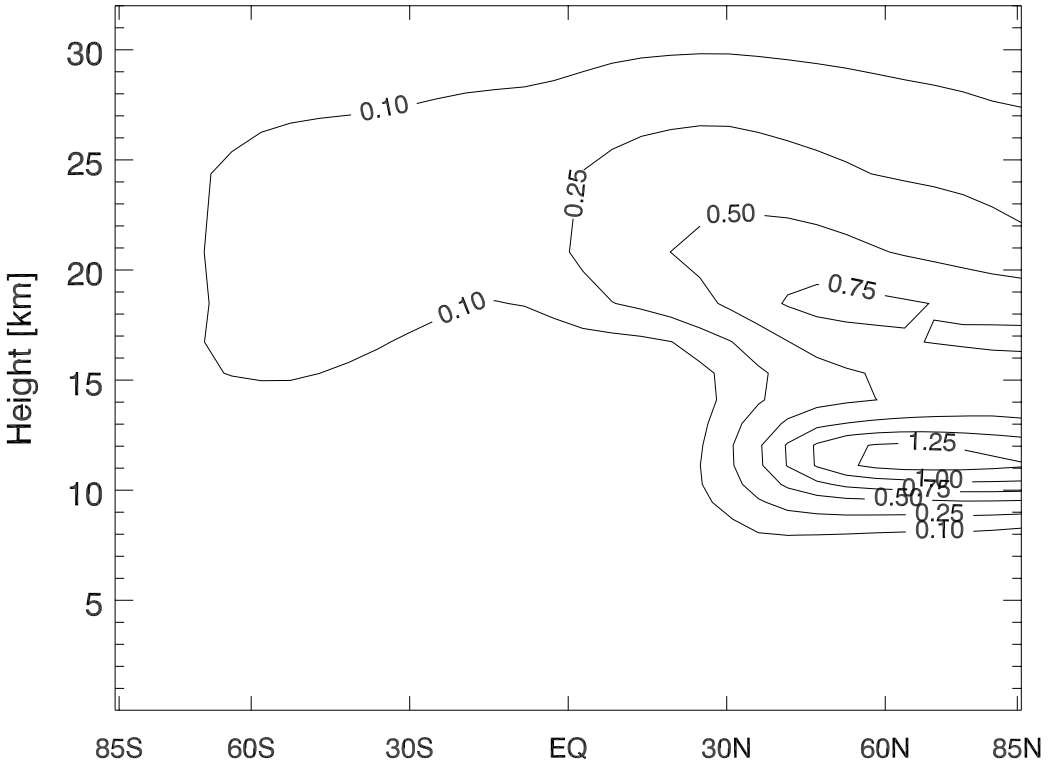

Fig. 1. Effect of mixed fleet aircraft emissions on NO_y, annual zonal mean (MIX minus noAC). Max: 1.35 ppbv.

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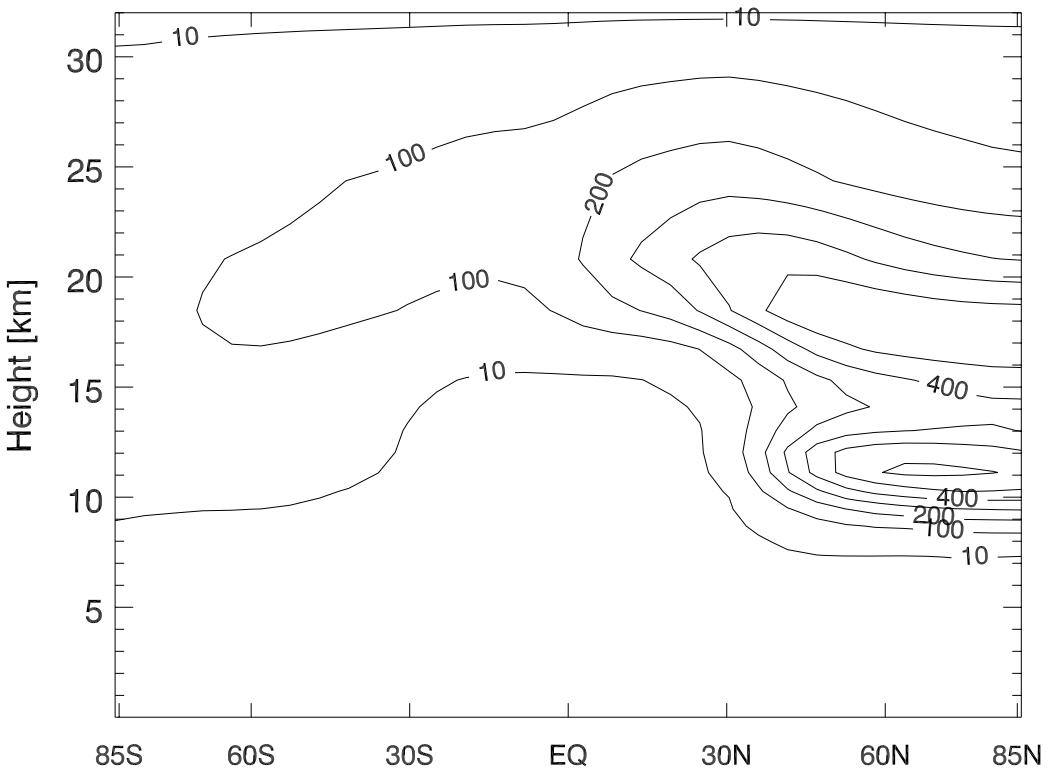

Fig. 2. Latitude and magnitude variation of the monthly mean maximum effect of aircraft emissions on NO_y (MIX minus noAC): 1.1 to 1.9 ppbv.

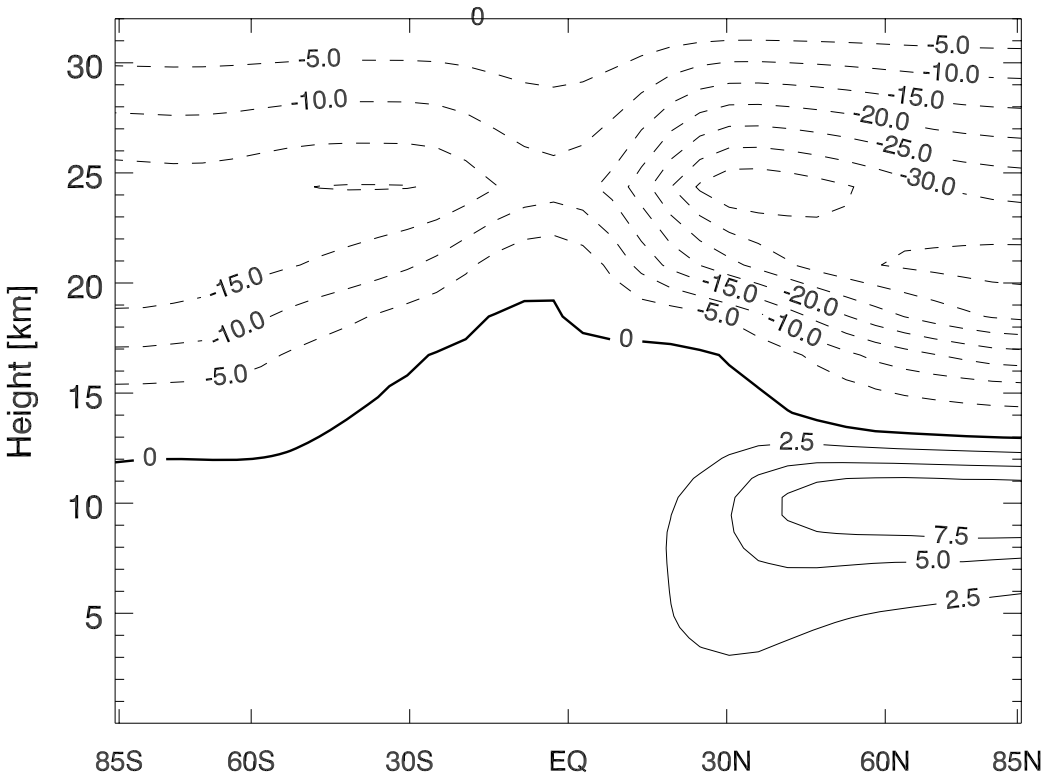

Fig. 3. Effect of aircraft emissions on H_2O, annual zonal mean (MIX minus noAC). Max: 629.69 ppbv.

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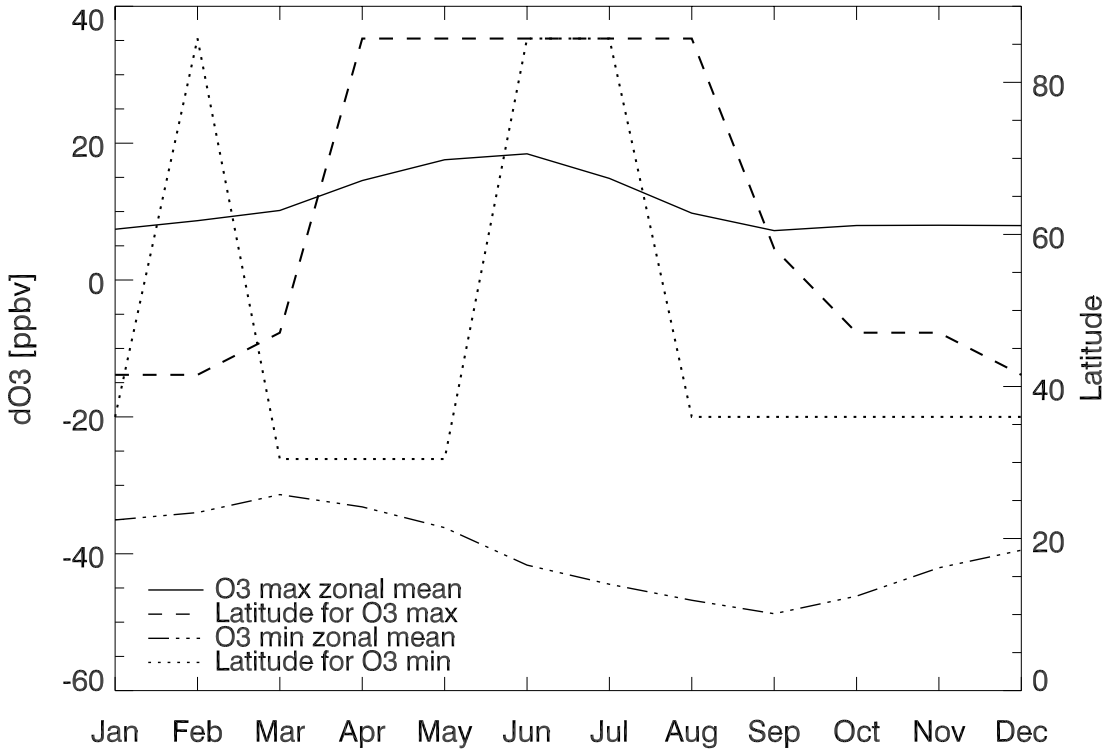

Fig. 4. Effect of aircraft emissions on O_3, annual zonal mean (MIX minus noAC). Min: –39.24 ppbv, max: 9.96 ppbv.

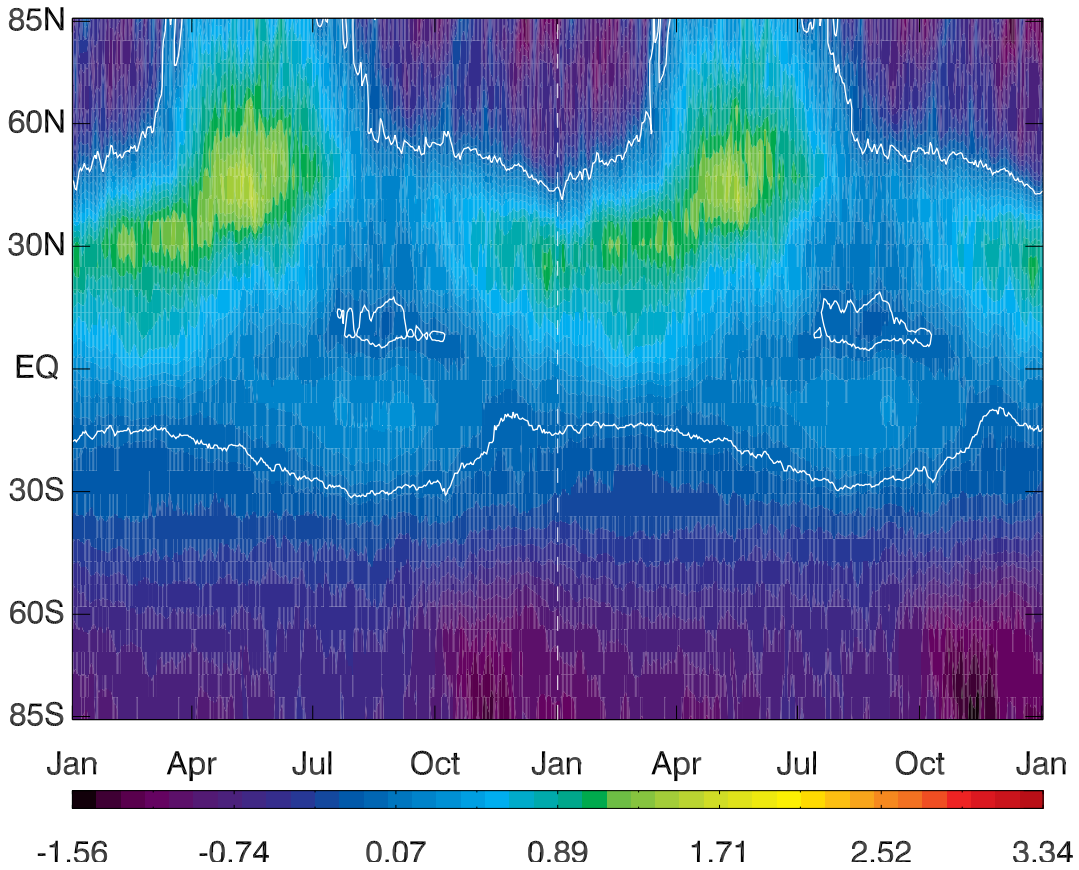

Fig. 5. Magnitude and latitude variation of the monthly zonal mean maximum/minimum effect of aircraft emissions on O_3 (MIX minus noAC), maximum ranging from 7.2 to 18.4 ppbv (due to subsonic aircraft) and minimum from –48.8 to –31.4 ppbv (due to supersonic aircraft).

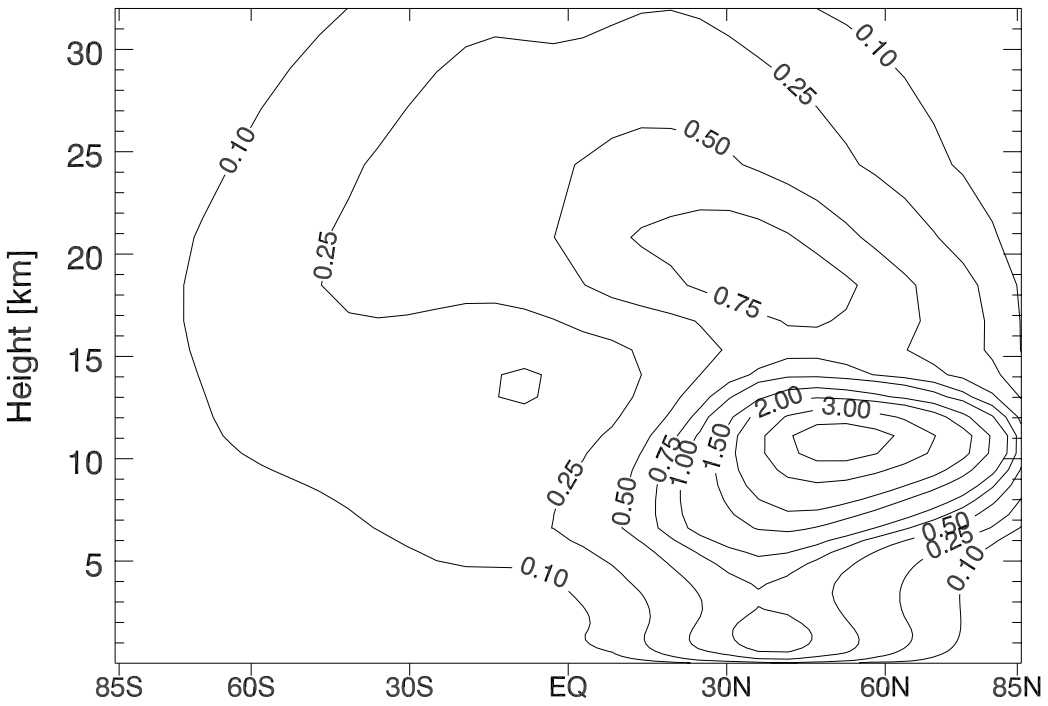

Fig. 6. Changes in the daily zonal mean total ozone column [Dobson Units] due to aircraft emissions (MIX minus noAC) for the last two years of simulation, ranging from –1.56 DU to 1.65 DU.

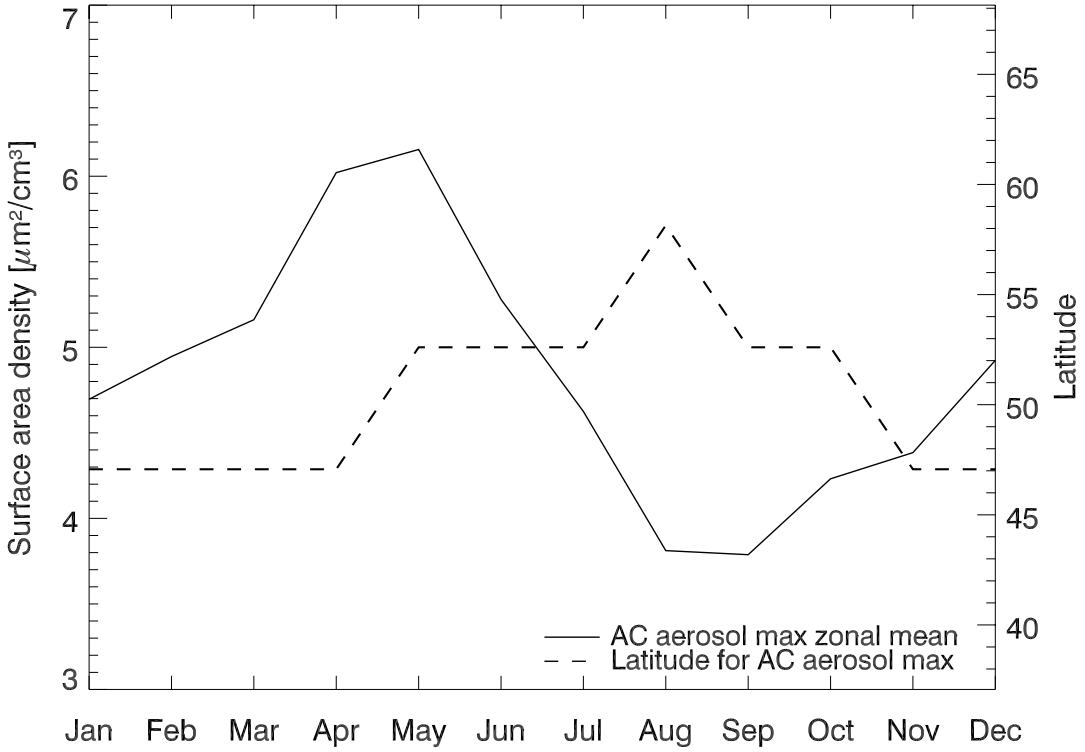

Fig. 7. Annual zonal mean of aircraft aerosol surface area density from the mixed fleet. Max: 4.7 $\mu m^2 cm^{-3}$.

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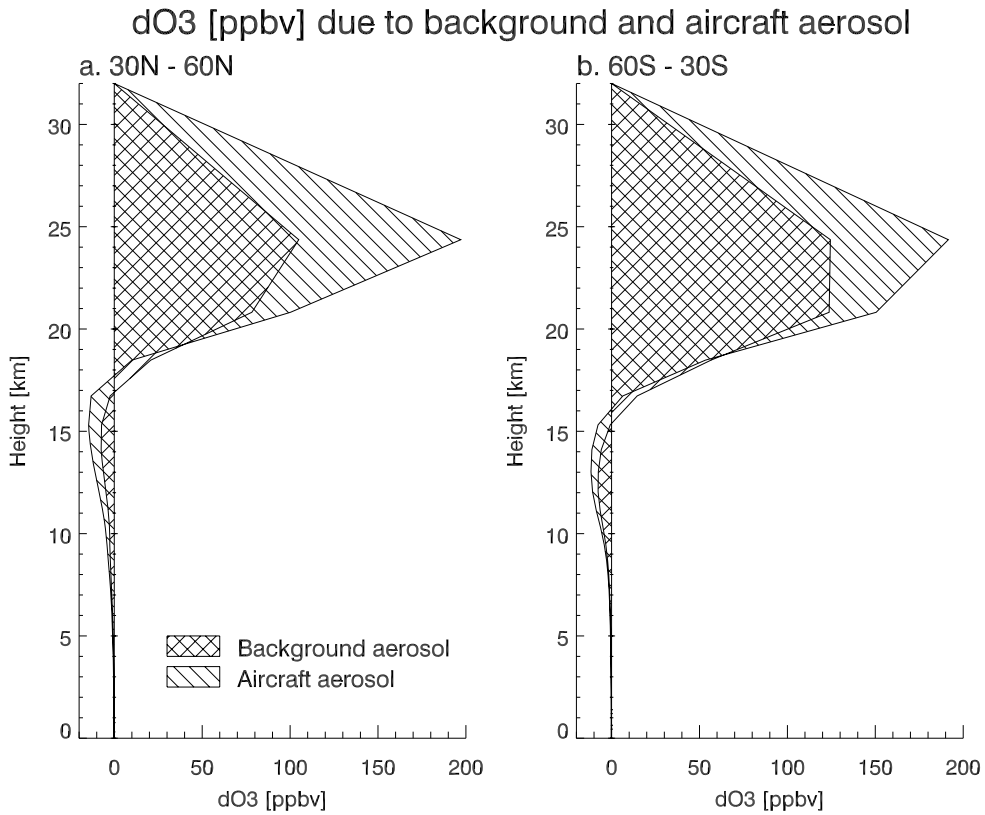

Fig. 8. Variation of the magnitude and latitude of the monthly zonal mean maximum of aircraft aerosol surface area density.

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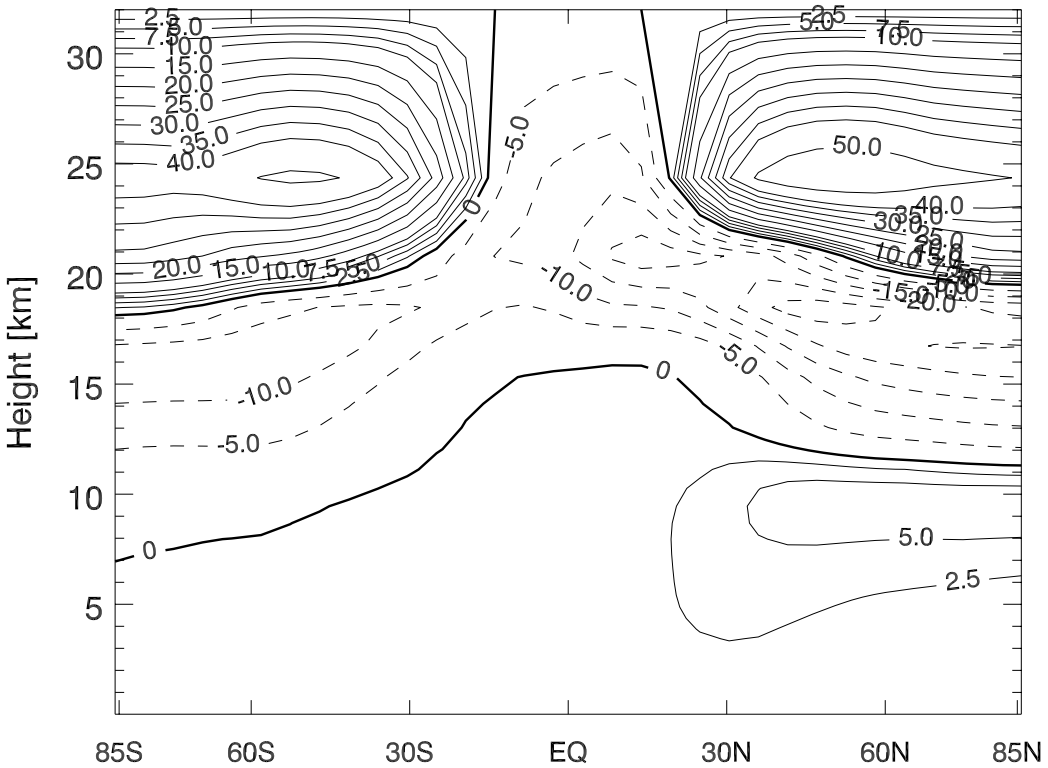

Fig. 9. Annual zonal mean effect of background aerosol on O_3 (cross-hatched) and the contribution from aircraft aerosol emissions (hatched). a: 30 N–60 N, b: 60 S–30 S.

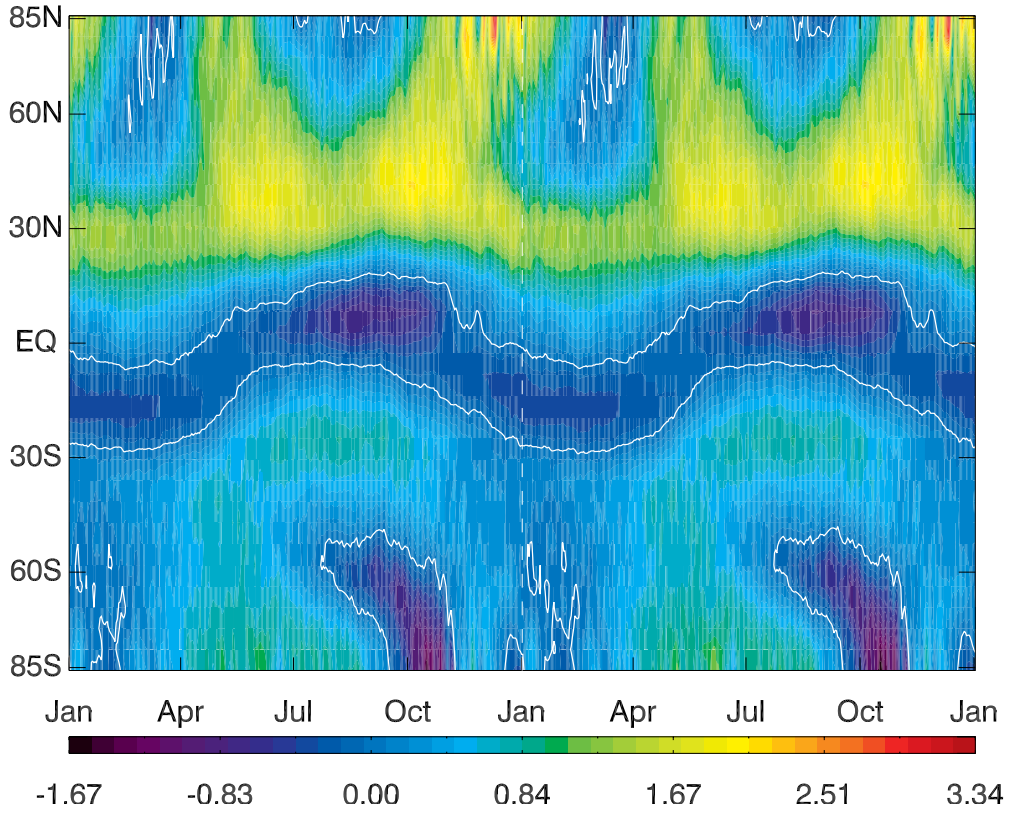

Fig. 10. Annual zonal mean effect of a mixed fleet with aerosol emissions on O_3 (AERO minus noAC).

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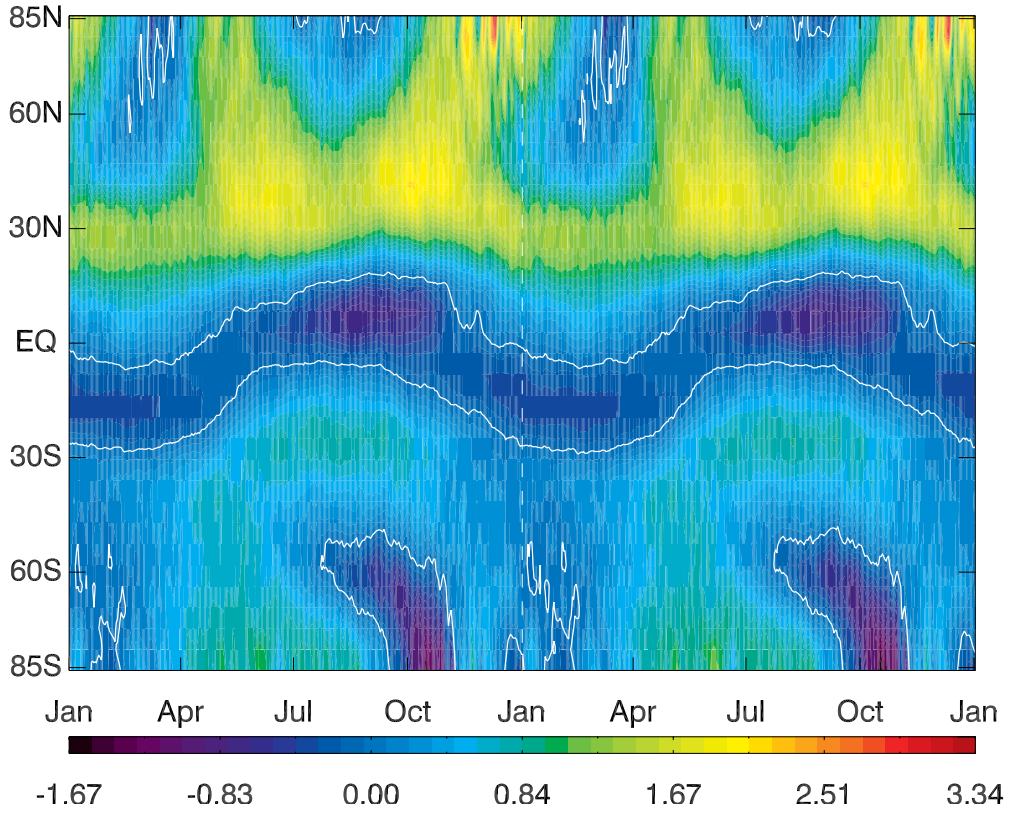

Fig. 11. Changes in the daily zonal mean total ozone column [Dobson Units] due to the mixed fleet aircraft emissions including aerosol emissions (AERO minus noAC), ranging from –1.67 DU to 3.34 DU.

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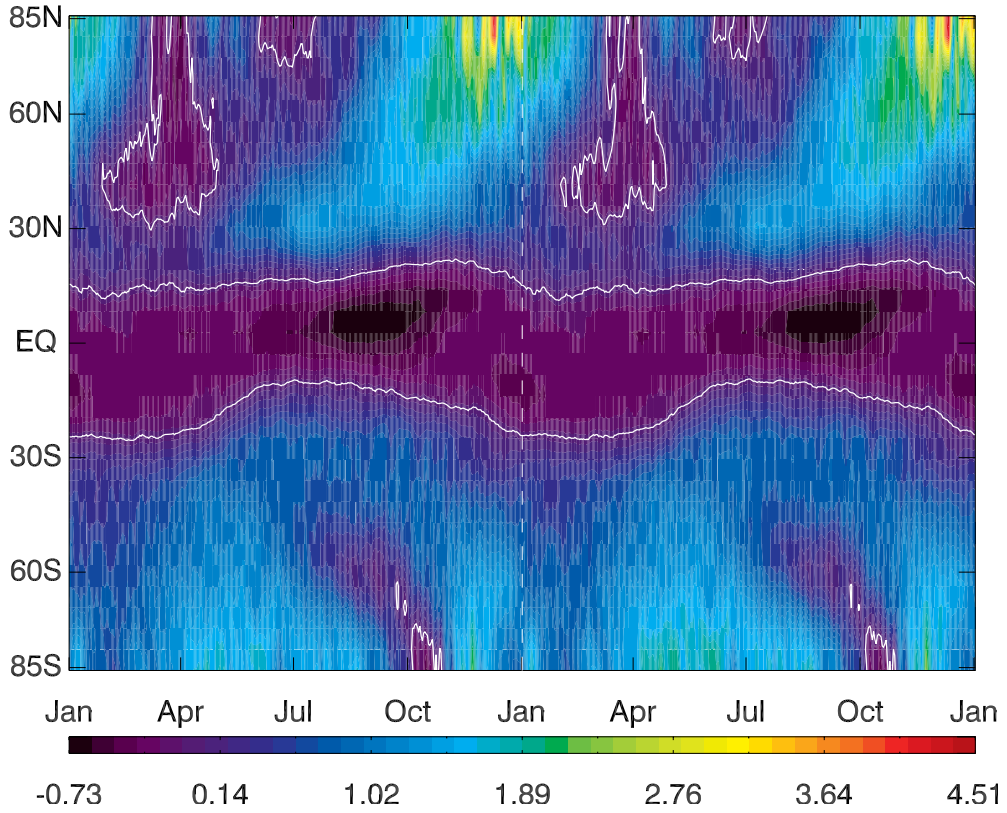

Fig. 12. Changes in the daily zonal mean total ozone column [Dobson Units] due to the mixed fleet aerosol emissions (AERO minus MIX).

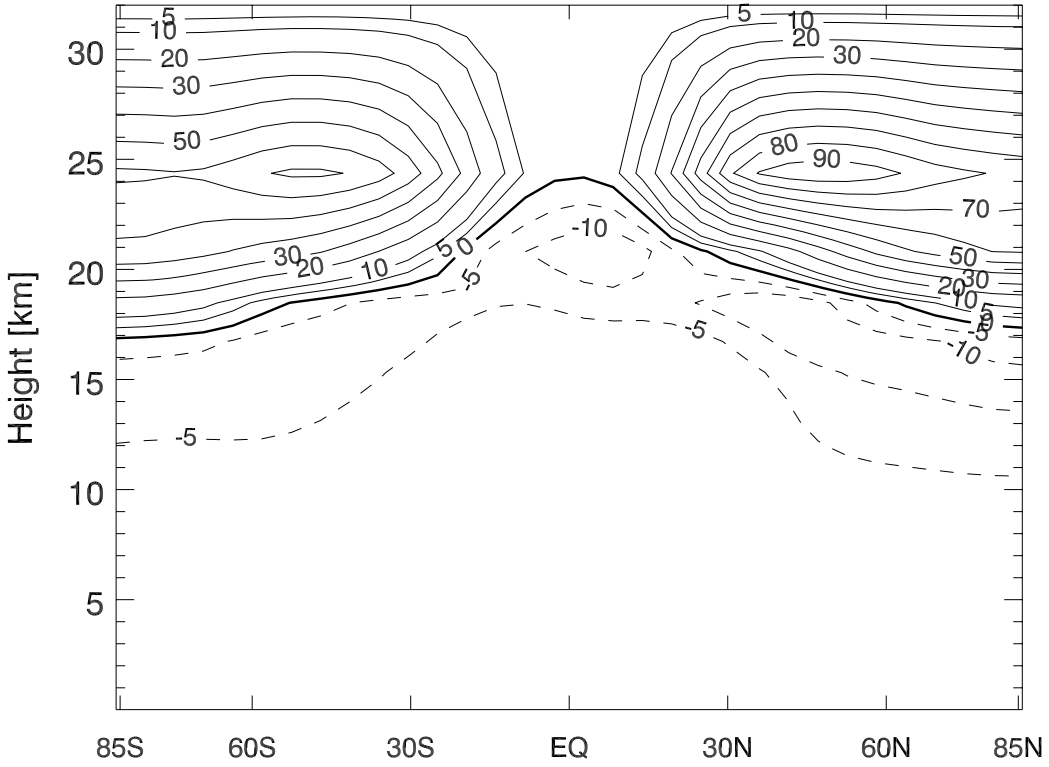

Fig. 13. Annual zonal mean effect of aircraft aerosol emissions on O_3 (AERO minus MIX).

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