The origin of ozone

V. Grewe

Atmos. Chem. Phys. Discuss., 5, 9641–9668, 2005
www.atmos-chem-phys.org/acpd/5/9641/
SRef-ID: 1680-7375/acpd/2005-5-9641
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

The origin of ozone

V. Grewe

Institut für Physik der Atmosphäre, DLR-Oberpfaffenhofen, Wessling, Germany

Received: 14 July 2005 – Accepted: 31 July 2005 – Published: 7 October 2005

Correspondence to: V. Grewe (volker.grewe@dlr.de)

Abstract

Highest ozone production rates can be found at around 30 km in the tropical stratosphere, leading to ozone mixing ratios of about 10 ppmv. Those stratospheric air masses are then transported to extra-tropical latitudes via the Brewer-Dobson Circulation. This is thought to be the main mechanism, how mid- and high latitude ozone is generated. By applying the climate-chemistry model E39/C, this view is investigated in more detail. The origin of ozone in the troposphere and stratosphere is analyzed, by incorporating a diagnostic ("marked ozone origin tracers") into the model, which allows to identify the origin of ozone. In most regions the local simulated ozone concentration is dominated by local ozone production, i.e. less than 50% of the ozone at higher stratospheric latitudes is produced in the tropics, which conflicts with the idea that the tropics are the global source for stratospheric ozone. Although episodic stratospheric intrusions occur basically everywhere, the main ozone stratosphere-to-troposphere exchange is connected to exchange processes at the sub-tropical jet-stream. The tropospheric influx of ozone amounts to 420 Tg per year, and originates on the Northern Hemisphere from the extra-tropical stratosphere, whereas on the Southern Hemisphere a re-circulation of tropical tropospheric ozone contributes most to the influx of ozone into the troposphere. In the model, the upper tropopause of both hemispherers is clearly dominated by tropical tropospheric ozone (40%–50%) except for Northern Summer, where the tropospheric contribution (from the tropics as well as from the Northern Hemisphere) does not exceed 20%.

1. Introduction

The ozone distribution in the lower stratosphere is largely controlled by tropical ozone production and below 30 km by large-scale transport, i.e. the Brewer-Dobson-Circulation, especially in winter (World Meteorological Organization (WMO), 1999). Air masses are uplifted in the tropics, transported then to the winter pole and subside

ACPD

5, 9641–9668, 2005

during winter. However, transport time-scales and the stratospheric age of air exceeds two years (Hall and Plumb, 1994) meaning that the seasonal cycle has to be taking into account. Early studies of the meridional circulation (Dunkerton, 1978) in a Lagrangian view suggest that the air masses are rising in the tropics, transported to higher latitudes, but than are swapped northward and southward according to the direction of the Brewer-Dobson Circulation during the course of year. This makes the differentiation between dynamical and chemical implications for ozone more complicated, since the chemical life-time of ozone highly depends on the time of year and on latitude. Furthermore, it has to be noted that wave-breaking in the lowermost stratosphere results in transport of tropical air masses to higher latitudes through the sub-tropical barrier even at lower stratospheric altitudes. These so-called streamers were frequently observed (e.g. Offermann et al., 1999) and transport low ozone air masses to higher latitudes in all seasons, depending on strength of the storm tracks (Eyring et al., 2003). It has been speculated that this process may have an important contribution to the mid-latitude ozone concentration at the tropopause altitudes (Grewe et al., 2004). The WMO report (1999) summarizes this by pointing out that the classical view of the Brewer-Dobson-Circulation has been significantly refined in recent years, which has consequences on the ozone controlling processes. And it is questionable, whether the tropical stratospheric region is the dominant source of ozone, even at higher latitudes.

This paper aims at separating the importance of large-scale transport from the importance of chemistry for different regions taking into account their variations during the course of the year. The basic question to be answered is: Taking the ozone concentration at any given point, in which regions is this ozone produced? This has an implication on estimates of trends, because, if we know the contributions from different regions to a local ozone concentration, we can estimate the impact of variations or trends in large-scale transport on the local ozone concentration, i.e. on its trends.

To answer those questions the fully coupled climate-chemistry model E39/C is applied, which includes both troposphere and stratosphere dynamics and chemistry. A number of diagnostic ozone tracers are included, which label ozone molecules accord-

ing to their production region. The model and this technique is described in the following two sections. Section 4 addresses the origin of the stratospheric ozone, whereas Sect. 5 describes the origin of ozone transported into the troposphere. Section 6 discussed the production and transport of tropospheric ozone. In Sect. 7 a comparison to observational data is provided including a discussion of the implication on the findings.

2. Model description

For the determination of the origin of ozone, the coupled chemistry-climate model E39/C is applied, which consists of the spectral atmospheric general circulation model ECHAM4.L39(DLR) (E39) and the chemistry module CHEM (C). E39 is based on the climate model ECHAM4 (Roeckner et al., 1996) with increased vertical resolution from 19 to 39 levels and the top layer centered at 10 hPa (Land et al., 1999). In this study, the model was applied with a horizontal resolution of T30, i.e. a corresponding grid size of 3.75°×3.75°. Water vapour, cloud water and chemical species are advected by a so-called semi-Lagrangian scheme. E39 includes parameterization schemes for small scale physical processes, like convection or cloud formation. The model has been used in a variety of tracer transport studies (Land et al., 2002; Rogers et al., 2002; Timmreck et al., 1999).

The chemistry module CHEM (Steil et al., 1998) is based on the family concept. It contains the most relevant chemical processes for describing the tropospheric background NO_x-CH_4-CO-HO_x-O_3 chemistry as well as the stratospheric homogeneous and heterogeneous ozone chemistry. CHEM includes 37 chemical species of which 9 are explicitly transported and the others are grouped to 3 families. 107 photochemical reactions and 4 heterogeneous reactions on polar stratospheric clouds (PSCs) and on sulphate aerosols are considered in CHEM, however not yet bromine chemistry. Mixing ratios of methane (CH_4), nitrous oxide (N_2O) and carbon monoxide (CO) are prescribed at the surface. Nitrogen oxide emissions at the surface (natural and anthropogenic sources) and from aircraft are considered. Lightning NO_x emissions

ACPD
5, 9641–9668, 2005

are calculated interactively depending on the mass fluxes in deep convective clouds according to Grewe et al. (2001).

Model climatologies and detailed descriptions are given in Hein et al. (2001) and Grewe et al. (2001). The model has been applied for various chemistry-transport and climate-chemistry investigations (Schnadt et al., 2002; Grewe et al., 2002; Dameris et al., 2005) and contributed to model inter-comparisons (Austin et al., 2003). Recently, Dameris et al. (2005) showed that ozone variations during the last 40 years can be realistically reproduced by employing realistic external forcings (e.g. volcanic eruptions, El Niño, Quasi-Biennial Oscillation).

For this investigation a 5 year run has been performed applying constant boundary conditions for 1990 and including additional ozone origin tracers (see below).

3. Methodology

As a first step, areas of interest for the ozone origin are defined (Table. 1). The nine areas (3 latitude bands and 3 height regions) are illustrated in Fig. 1. The areas are chosen, so that the main ozone production area (tropical mid stratosphere) can be separated from extra-tropical regions and that the tropospheric ozone production areas can be separated from the stratosphere.

In a second step, an ozone tracer ($^{i}O_3$, with i index for the region) is assigned to each of the nine regions. Each tracer experiences ozone production only in that region, but ozone destruction in the whole model domain, according its concentration:

$$\frac{d\,{}^{i}O_3}{dt} = {}^{i}O_3^{Prod} - O_3^{Loss}\frac{{}^{i}O_3}{O_3}, \quad (1)$$

with ${}^{i}O_3^{Prod}$ and O_3^{Loss} the ozone production and loss in ($m^3/m^3/s$), derived from the modelled ozone O_3 in (m^3/m^3), with the restriction that ${}^{i}O_3^{Prod}$ equals to zero for each grid point outside the region i. It can easily be shown that $\sum_i {}^{i}O_3 = O_3$ for all time

ACPD
5, 9641–9668, 2005

steps and grid points, if this equation holds for the initial conditions and that for all other initial conditions $\sum_i {}^{i}O_3$ converges exponentially, proofing the convergence of the methodology. At any grid point ozone originates therefore by $\frac{{}^{i}O_3}{O_3} \times 100\%$ from region i.

This analysis technique has been implemented into the climate-chemistry model E39/C (Hein et al., 2001) and applied to a model simulation, which is identical to Grewe et al. (2001). Figure 2 shows the results for January, the combined effect of chemistry and transport, occurring until January and leading to the simulated ozone concentration in January. For the region TRMS (mid of the top row) it can clearly be seen that ozone is produced in that area and how it is destroyed along its way through the atmosphere. Only roughly 1 to 5 ppbv out of 6–9 ppmv, i.e. less than 0.1%, is reaching the ground on the Northern Hemisphere. It does not imply that these ozone molecules are actually produced in January, indeed more likely during summer, i.e. that they are a reminiscence from summertime ozone production, which is then transported slowly into the troposphere.

4. Stratospheric ozone origin

Figures 2 and 3 show the evolution of the nine diagnostic ozone tracers for January and July, respectively. The first impression is that each ozone tracer has its peak value in its region of origin, which directly follows from Eq. (1). In the tropics at 10 to 30 hPa, ozone is chemically controlled, which implies that local ozone production plays the major role. However, at mid and higher latitudes the picture is reversed. Between 30° N and 60° N in January and between 30° S and 45° S in July, i.e. on the winter hemispheres, the tropical ozone (TRMS) dominates over local ozone production. This can be made clearer by colour coding the regions of origin and re-arranging them in the order of their importance (Fig. 4): first (second, third) column shows the main (second, third) contributing region, which region is indicated by the colour code (Fig. 1)

and the isolines indicate how much the region contributes to the ozone amount (in percentage). For example, in January at 20 hPa an 40° N, i.e. in the region NHMS, the main contributor to the ozone concentration is the ozone produced in the tropics (light green, TRMS) with 70%, followed by ozone produced in the NMHS region (pink) by around 30%. Other contributors are almost unimportant with values less than ≈2% (e.g. dark green, TRLS). However, in the summer hemispheres ozone chemistry is fast enough, i.e. the life-time is small enough, that the local ozone production dominates over transport from the tropics. This means that the seasonal cycle is an important factor for regenerating stratospheric ozone at mid and high latitudes.

This seasonal cycle is illustrated in Fig. 5a for the main contributor to ozone at 50 hPa, 60° N. There, ozone originates mainly from the tropical mid stratosphere between February and July, with peak values of 2.2 to 2.7 ppmv in March and April, well-known as the spring-time mid-latitude ozone maximum. These high values are then reduced by roughly 50% to 60% until October, reaching then values of about 1 to 1.2 ppmv. Ozone produced in the extra-tropics decreases from higher values in winter (2.3 to 2.5 ppmv) down to about 1.4 ppmv in June, indicating that local ozone production dominates over transport from the tropics between August and February. In the beginning of the winter season (November) the mid-latitude ozone concentration originates by approximately 1/3 from the tropics and 2/3 from local ozone production during summer and autumn. Contributions from other regions are almost unimportant.

At polar latitudes (90° N; Fig. 5b), the picture is somewhat different. In the beginning of the winter, ozone originates mainly from local (NHMS) ozone production with around 3 ppmv, which is then chemically destroyed, reaching values in February and March of about 1.5 to 2.7 ppmv, showing a loss of about 0.3 to 1.5 ppmv. The large inter-annual variability is consistent with observational data and covers the observed range (Müller et al., 1996; Rex et al., 2004). It shows the ability of the model E39/C to simulate the variability of the stratospheric chemical and dynamical processes. Ozone originating from the tropics has an increasing importance at high latitudes from January (≈1 ppmv) to April (up to 3 ppmv). To better illustrate the chemical regime and to better separate

dynamical from chemical processes, Fig. 5c shows the seasonal cycle for 90° N equivalent latitude, i.e. for the center of the polar vortex, instead of the North Pole like in Fig. 5b. In the center of the vortex, the increase of tropical ozone is delayed compared to the North Pole, and the high TRMS ozone values of up to 3 ppmv are only reached at the end of the winter, when the vortex breaks up and mixing of vortex and non-vortex air is increased. Wintertime ozone supply from the tropics is increasing very differently from year to year between roughly 1 and 2 ppmv, which is a consequence of the different simulated norther winter situations. It also partially masks the chemically ozone loss of 0.3 to 1.5 ppmv in the model, so that the ozone depletion is correctly simulated, but the total ozone decline ends up to be underestimated. Ozone produced in the TRMS region declines then quite rapidly from March onwards mirroring the decrease of the ozone life-time of roughly 2 months, which is in accordance with previous findings by Stenke and Grewe (2004). They showed that the life-time of a spring-time chemically induced ozone perturbation (in this case caused by ozone mini-holes) vanishes totally at the end of summer. Fioletov and Shepherd (2003) found similar lifetimes for Northern Hemisphere mid-latitude ozone. They showed that ozone anomalies in late spring are correlated with summertime ozone. The correlation still holds until beginning of autumn, with low values however. Deviations in March of ±30 DU decrease to ±20 DU in June and ±5 DU in September, based on TOMS and SBUV data from 1979 to 2000. This would suggest an e-folding time of around 3 months, somehow longer than simulated, though the approach differs.

At lower stratosphere tropical altitudes (100–60 hPa, Fig. 5d) ozone chemistry is slow and ozone is dominated equally by chemistry and transport (Brasseur and Solomon, 1986). Since the meridional circulation has also a seasonal cycle, which peaks in February (Reithmeier, 2001), the impact of tropospheric ozone (TRTS) peaks with some delay in April. The contribution from NHMS region has a quite strong seasonal cycle, peaking in June, when it contributes with similar amount to local ozone like the local ozone production. This reflects the Brewer-Dobson circulation, which transports ozone towards the winter poles, i.e. ozone produced at mid and high latitudes during

March to June is transported to lower latitudes. In total, up to 0.5 ppmv (Figs. 2 and 3) can be produced locally and transported to higher latitudes via streamer events (Eyring et al., 2003). This is an important contribution to ozone at the tropopause, especially in the Southern Hemisphere, where this is the second important contributor with roughly 30% (Fig. 4).

In general, the seasonal cycles are showing little inter-annual variability, except for Northern Hemisphere winter episodes. However, this may be somehow underestimated by the model set-up, since not the whole natural variability is reproduced in the applied model set-up. The inclusion of external forcings enhance the variability (Dameris et al., 2005).

5. Stratosphere troposphere exchange

The large-scale stratosphere troposphere exchange of air mass occurs predominantly in conjunction with the subtropical jet stream and the polar jet stream (Holton et al., 1995). Figure 6a shows the simulated mass exchange calculated with the method of Wei (1987). The values are in good agreement with those derived with the same methodology based on meteorological data from the European Centre of Medium-Range Weather Forecasts (Grewe and Dameris, 1996). The simulated stratosphere-troposphere exchange occurs clearly in conjunction with the sub-tropical jet-stream, peaking on either hemisphere in winter and spring. Since the location of the jet stream moves to higher latitudes in summer in either hemisphere, the air mass exchange peaks also at higher latitudes.

The respective ozone fluxes are shown in Fig. 6b. A total annual mean ozone flux into the troposphere of 420 TgO_3/a (range: 346–474) is simulated, which is on the lower side of estimates derived from observations of 450 (range: 200–870) TgO_3/a (Murphy and Fahey, 1994), 510 (range: 450–590) TgO_3/a (Gettleman et al., 1997) and 500 (range: 479–523) TgO_3/a (Olsen et al., 2004). However, compared with other modelling studies, ranging from 391 to 1440 TgO_3/a (IPCC, 2001), and taking into

account the estimate uncertainties and ranges, the simulated ozone influx of 420 Tg/a seems to be reasonable. The overall pattern of the ozone flux is similar to the mass fluxes. However, the seasonal cycle of ozone in the lower stratosphere has an impact on the absolute values, e.g. leading to higher ozone fluxes at around 30° N during spring (MAM) compared to winter (DJF).

Figure 7 shows where the exchanged ozone originates from for the Northern (a) and Southern (b) Hemisphere. In the Northern Hemisphere the annual influx is dominated by ozone produced in the NHMS region. It shows a quite pronounced seasonal cycle, with lowest values in winter, because of the low ozone production rates and the stronger impact from the tropics, since the ozone life time is increased, leading to farer-reaching dispersion of tropical ozone. For the same reason ozone from the TRMS region has a minimum in summer-time. Almost the same amount of ozone mixed into the troposphere originates from the tropical mid stratosphere (TRMS) and from the tropical troposphere (TRTS). Latter represents a re-circulation through the lowermost stratosphere.

Exactly this mechanism leads to the most important contribution to the ozone influx in the Southern Hemisphere (Fig. 7b). As in the Northern Hemisphere, the seasonal cycle of the ozone mixed into the troposphere originating form SHMS is reversed compared to those from the tropics (TRTS, TRLS, and TRMS) again for the same reasons.

6. Tropospheric ozone origin

In the troposphere the origin of ozone at low altitudes is mainly dominated by local ozone production (Fig. 4), except for the ozone in the southern extra-tropics during winter, where the transport of tropical ozone is over-compensating the local ozone production. In the tropics, ozone chemistry is fast enough throughout the year so that the local ozone production is always more important than ozone transport from higher latitudes or from the stratosphere. Interestingly, largest contributions to the local ozone concentration in the upper troposphere of both hemispheres are coming from

tropical ozone (TRTS) throughout the whole year, except for northern summer (Fig. 4, left), where ozone from lower altitudes dominates. At lower altitudes, tropical ozone is even second important in the extra-tropics, probably it is mixed down from the upper troposphere to the lower troposphere, since low level transport would be combined with a much shorter ozone lifetime.

Ozone, which is produced in the stratosphere (i.e. not in NHTS, TRTS, and SHTS), has an important contribution to ozone in the troposphere, in the order of 30% in the winter hemisphere and 20% in the summer hemisphere (Fig. 8). However, on the Northern Hemisphere the contribution of ozone produced in the stratosphere to the tropospheric ozone budget is systematically higher (around 5%) than in the Southern Hemisphere, for the respective season, which largely is a consequence of the higher ozone exchange rates in the Northern Hemisphere compared to the Southern Hemisphere (Fig. 6). Ozone in the tropical troposphere clearly has a lower contribution from ozone produced in the stratosphere because of faster chemistry. The areas with stronger ozone influx (Fig. 7) clearly show an enhanced stratospheric contribution. The origin of the ozone produced in the stratosphere and found in the troposphere can be identified in Fig. 4. In the tropics, ozone mainly originates from SHMS and TRMS in January and from NHMS in July. In the extra-tropics, the most important region are SHMS for the Southern Hemisphere and NHMS for the Northern Hemisphere, but not the region TRMS.

Since non-methane-hydrocarbon chemistry is not included in the model, the ozone lifetime is overestimated by the model, which likely leads to too high values of stratospheric ozone found in the troposphere, especially close to the Earth's surface. Lamarque et al. (2005) applied the MOZART-2 model, which includes non-methane-hydrocarbon chemistry, found a contribution of stratospheric ozone to the surface ozone concentration of up to 10% in the Northern Hemisphere and up to 20% in the Southern Hemisphere, for 1990 conditions. Whereas in this study the contributions are as much as 20% for the Southern Hemisphere and around 25% in the Northern Hemisphere, which partly (besides differences in emission datasets) can be explained

by the enlarged ozone lifetime in the E39/C model near the surface.

7. Discussion

In order to determine the origin of ozone, a CCM was applied and coupled to a simple diagnostic. That implies that the quality of the results are largely determined by the quality of the model and the diagnostic.

First, the diagnostic, which marks ozone molecules according to their region of production (see Sect. 3) has a very low potential error. Since ozone transport is linear and chemistry is not affected, the only potential error source is numerical diffusion occurring in the transport scheme. Those can arise, when large gradients occur. However, the relatively high vertical resolution at tropopause regions of about 700 m, is sufficient to correctly maintain gradients especially at tropopause altitudes, where the ozone gradient maximizes (Grewe et al., 2002). Therefore, a significant impact is not expected.

Second, the models ability to simulated atmospheric transport and chemistry, and its interactions has to be good enough to come to reliable conclusions. Land et al. (1999) and Austin et al. (2003) showed that the meridional circulation is in reasonable agreement with observations. Dameris et al. (2005) showed that the stratospheric dynamics in terms of stratospheric wind and temperature fields are in agreement with NCEP data, except for the Southern Hemisphere, where the temperatures are too low and the polar vortex is too stable. In addition the interaction of dynamics and chemistry is well represented, with a high variability of ozone during Northern Hemisphere winter and spring season. However, a closer look clearly reveals some drawbacks. For example, the vertical ascent in the tropical stratosphere is too fast, instead with 0.4–0.5 mm/s (Stenke, 2005) compared to around 0.3 mm/s from observational data (Rosenlof, 1995), which impacts the tape recorder effect and water vapour is lifted too fast.

Further, the stratosphere is characterized by several barriers to transport. The subtropical barrier, is clearly visible in, e.g. simulated N_2O fields, and also the frequency

of quasi-horizontal exchange processes, is realistically reproduced in terms of location, seasonal cycle and absolute frequency (Eyring et al., 2003). However, the polar night jet, as a barrier to transport is too leaky. Although a clear gradient is maintained throughout the winter, subtropical airmasses are mixed into the polar vortex as shown in Sect. 4, masking partially the polar chemical ozone loss. This on the other hand, demonstrates the ability of the applied methodology to identify model deficits.

Connected to the overestimated tropical updraft, the turnaround times for stratospheric air masses, i.e. the age of air is underestimated, leading to values around 2.5 to 3 years at higher latitudes, 25 km compared to 3.5 years from observational data (Hall and Plumb, 1994). This is partially connected to the upper boundary in the applied model system, which is centered at 10 hPa and forces the upper branch of the Brewer Dobson Circulation into this layer. On the other hand, the interaction of chemistry and transport in the extra-tropical stratosphere, seems to be realistically simulated, since the turn-around time of a stratospheric ozone perturbations is realistically simulated (see Sect. 4).

Anyway, the discussed model deficiencies point at the same direction, the transport of ozone, produced at tropical stratospheric latitudes to higher latitudes is overestimated in the model compared to observational data. This even implies that the one finding of this paper, namely that less than 50% of the ozone found at higher stratospheric latitudes is actually produced in the tropics, is even an upper boundary, because longer transport time-scales would decrease that contribution.

The evaluation of the contribution of ozone produced in the stratosphere to the tropospheric ozone budget is a challenge. In Sect. 5 it has been shown that the exchange of ozone into the troposphere is reasonably simulated, at least within the ranges of uncertainty. How much this ozone produced in the stratosphere contributes to the tropospheric ozone budget is then a combination of the tropospheric mixing and chemical ozone lifetime in the troposphere. At the surface, observational data suggest that the stratospheric contribution amounts to 14% at Areskutan, Sweden, with peak values up to 50% (Bazhanov and Rodhe, 1997). Model studies suggest a maximum contribution

to near surface ozone ranging from 5% (Follows and Austin, 1992) to 10% (Southern Hemisphere) and 20% (Northern Hemisphere) (Lamarque et al., 2005), which is less than in found in this study (15%–30%) and may partially be explained by missing NMHC chemistry, which would increase tropospheric HO_2 concentration and therefore reduce the ozone lifetime (Roelofs and Lelieveld, 2000). Another model study suggest that the stratospheric ozone may contribute to the tropospheric ozone column by 30% to 70% (de Laat et al., 2005), which seems to be more in agreement with our findings and indicates that the ozone lifetime may be overestimated especially at lower altitudes. Marcy et al. (2004) gave estimates of the stratospheric contribution to upper tropospheric ozone based of very precise HCl measurements, which correlate to stratospheric ozone. They concluded that 20% to 80% of the uppertropospheric ozone originates from the stratosphere in the altitude range of roughly 10 to 15 km at 31° N–33° N) Taking into account the large variability in the region of the subtropical jet, where the measurements were performed, those values are well covered by the model. However, a more climatological basis would be needed to sufficiently evaluate the model.

8. Conclusions

In this study, the origin of ozone has been analyzed, by applying the climate-chemistry model E39/C and including regionally marked ozone tracers. This methodology allows to attribute the ozone concentration at a given point of the atmosphere to some regions, where these ozone molecules were generated, i.e. where its origin is located.

Although highest local ozone production rates can be found in the tropics at around 30 km, this region does not dominate the extra-tropical stratospheric ozone distribution. At higher latitudes the local ozone production rates are considerably smaller, but the destruction rates are also smaller and, especially in summer-time, photochemical ozone production is the dominant ozone source. Estimates of the stratospheric ozone lifetime in northern mid and high latitudes has been found to be broadly in agree-

ment with observational data based on total ozone column measurements (Fioletov and Shepherd, 2003). However, there are some indications that the modelled lifetime may to too large.

During winter and spring polar ozone, i.e. ozone within the polar vortex, predominantly originated from NHMS ozone production during the preceding summer and autumn. This ozone mixing ratios of about 3 ppmv are then depleted during winter and spring to values as low as 1.5 ppmv, i.e. an ozone destruction of up to 50% is found. Ozone supply from the tropics (TRMS) is increased during that time, partially masking the chemical ozone depletion, which may be a result of the location of the uppermost model layer at 30 km. Here it is worthnoty to point out that this model deficiency could only be identified through the applied methodology.

Since the Northern Hemisphere has a large dynamical variability during winter and spring, this results in large interannual variations of ozone, which is also reflected in the exchange of stratospheric ozone into the troposphere. The total influx of ozone into the troposphere of around 420 TgO_3 per year has been found to agree fairly well with estimates derived from observations and other model studies. In the Northern Hemisphere the region NHMS dominates this influx with contributions from TRMS (tropical mid-stratosphere) and TRTS (tropical troposphere). The latter means a recycling of tropospheric ozone through the lower-most stratosphere. This recycling process is found to be the dominant source for ozone mixed into the troposphere in the Southern Hemisphere.

Surface ozone is found to be produced locally by around 75% and 85% in winter and summer, respectively, which may be underestimated due to the lack of NMHC chemistry in E39/C. However, other model studies (Lamarque et al., 2005) suggest that the differences are not large and may be in the order of 10%.

In the future, this approach can help to analyze long-term simulations (Dameris et al., 2005) and to attribute changes in ozone to processes or to climate variability patterns. It would also be a reasonable tool to identify model to model differences, since it provides a possibility to separate transport and chemical processes.

Acknowledgements. This study was sponsored by the EU-Projects SCENIC and TROCCINOX. The simulations were performed at the Deutsches Klimarechenzentrum GmbH (DKRZ). I would like to thank M. Dameris for helpful comments.

References

Austin, J., Shindell, D., Beagley, S., Brühl, C., Dameris, M., Manzini, E., Nagashima, T., Newman, P., Pawson, S., Pitari, G., Rozanov, E., Schnadt, C., and Shepherd, T.: Uncertainties and assessments of chemistry-climate models of the stratosphere, Atmos. Chem. Phys., 3, 1–27, 2003,
SRef-ID: 1680-7324/acp/2003-3-1. 9645, 9652

Bazhanov, V. and Rodhe, H.: Tropospheric ozone at the swedish mountain site Areskutan: budget and trends, J. Atmos. Chem., 28, 61–76, 1997. 9653

Brasseur, G. and Solomon, S.: Aronomy of the middle atmosphere, Reidel Publishing Company, Netherlands, 1986. 9648

Dameris, M., Grewe, V., Ponater, M., Deckert, R., Eyring, V., Mager, F., Matthes, S., Schnadt, C., Stenke, A., Steil, B., Brühl, C., and Giorgetta, M.: Long-term changes and variability in a transient simulation with a chemistry-climate model employing realistic forcing, Atmos. Chem. Phys. Discuss., 5, 2297–2353, 2005,
SRef-ID: 1680-7375/acpd/2005-5-2297. 9645, 9649, 9652, 9655

de Laat, A., Aben, I., and Roelofs, G.-J.: A model perspective on total tropospheric O_3 column variability and implications for satellite observations, J. Geophys. Res, 110, doi:10.1029/2004JD005 264, 2005. 9654

Dunkerton, T.: On the mean meridional mass motions of the stratosphere and mesosphere, J. Atmos. Sci., 35, 2325–2333, 1978. 9643

Eyring, V., Dameris, M., Grewe, V., Langbein, I., and Kouker, W.: Climatologies of subtropical mixing derived from 3d models, Atmos. Chem. Phys., 3, 1007–1021, 2003,
SRef-ID: 1680-7324/acp/2003-3-1007. 9643, 9649, 9653

Fioletov, V. and Shepherd, T.: Seasonal persistence of midlatitude total ozone anomalies, Geophys. Res. Lett., 30, doi:10.1029/2002GL016 739, 2003. 9648, 9655

Follows, M. and Austin, J.: A zonal average model of the stratospheric contributions to the tropospheric ozone budget, J. Geophys. Res., 97, 18 047–18 060, 1992. 9654

Gettleman, A., Holton, J., and Rosenlof, K.: Mass fluxes of O_3, CH_4, N_2O, and CF_2Cl_2 in the lower stratosphere calculated from observational data, J. Geophys. Res., 102, 19 149–19 159, 1997. 9649

Grewe, V. and Dameris, M.: Calculating the global mass exchange between stratosphere and troposphere, Ann. Geophys., 14, 431–442, 1996, SRef-ID: 1432-0576/ag/1996-14-431. 9649, 9666

Grewe, V., Brunner, D., Dameris, M., Grenfell, J., Hein, R., Shindell, D., and Staehelin, J.: Origin and variability of upper tropospheric nitrogen oxides and ozone at northern mid-latitudes, Atmos. Environ., 35, 3421–3433, 2001. 9645, 9646

Grewe, V., Dameris, M., Fichter, C., and Sausen, R.: Impact of aircraft NO_x emissions. Part 1: Interactively coupled climate-chemistry simulations and sensitivities to climate-chemistry feedback, lightning and model resolution, Meteorol. Z., 3, 177–186, 2002. 9645, 9652

Grewe, V., Shindell, D., and Eyring, V.: The impact of horizontal transport on the chemical composition in the tropopause region: lightning NO_x and streamers, Adv. Sp. Res., 33, 1058–1061, 2004. 9643

Hall, T. and Plumb, R.: Age as a diagnostic for stratospheric transport, J. Geophys. Res., 99, 1059–1070, 1994. 9643, 9653

Hein, R., Dameris, M., Schnadt, C., Land, C., Grewe, V., Köhler, I., Ponater, M., Sausen, R., Steil, B., Landgraf, J., and Brühl, C.: Results of an interactively coupled atmospheric chemistry-general circulation model: Comparison with observations, Ann. Geophys., 19, 435–457, 2001, SRef-ID: 1432-0576/ag/2001-19-435. 9645, 9646

Holton, J., Haynes, P., McIntyre, M., Douglas, A., Rood, R., and Pfister, L.: Stratosphere-troposphere exchange, Rev. Geophys., 33, 403–439, 1995. 9649

IPCC, Climate Change 2001 – THe scientific basis, Contributions of working group I to the Third Assessment Report of the Intergovernmental Panel of Climate Change (IPCC), Intergovernmental Panel on Climate Change, Cambridge University Press, New York, NY, USA, 2001. 9649

Lamarque, J.-F., Hess, P., Emmons, L., Buja, L., Washington, W., and Granier, C.: Tropospheric ozone evolution between 1890 and 1990, J. Geophys. Res., 110, D08 304, doi:10.1029/2004JD005 537, 2005. 9651, 9654, 9655

Land, C., Ponater, M., Sausen, R., and Roeckner, E.: The echam4.l39(dlr) atmosphere gcm, technical description and climatology, DLR-Forschungsbericht, 1991-31, 45 pp., ISSN 1434-

8454, Deutsches Zentrum für Luft- und Raumfahrt, Köln, Germany, 1999. 9644, 9652
Land, C., Feichter, J., and Sausen, R.: Impact of vertical resolution on the transport of passive tracers in the echam4 model, Tellus, 54, 344–360, 2002. 9644
Marcy, T., Fahey, D., Gao, R., Popp, P., Richard, E., Thompson, T., Rosenlof, K., Ray, E., Salawith, R., Atherton, C., Bergmann, D., Ridley, B., Weinheimer, A., Loewenstein, M., Weinstock, E., and Mahoney, M.: Quantifying stratospheric ozone in the upper troposphere with in situ measurements of hcl, Science, 304, 261–265, 2004. 9654
Müller, R., Crutzen, P., Grooss, J.-U., Brühl, C., III, J. R., and Tuck, A.: Chlorine activation and ozone depletion in the arctic vortex: Observations by halogen occultation experiment on the upper atmosphere research satellite, J. Geophys. Res, 101, 12 531–12 554, 1996. 9647
Murphy, D. and Fahey, D.: An estimate of the flux of of stratospheric reactive nitrogen and ozone into the troposphere, J. Geophys. Res., 99, 5325–5332, 1994. 9649
Offermann, D., Grossmann, K., Barthol, P., Knieling, P., Riese, M., and Trant, R.: The CRyogenic Infrared Spectrometers and Telescopes for the Atmosphere (CRISTA) experiment and middle atmosphere variability, J. Geophys. Res, 104, 16 311–16 325, 1999. 9643
Olsen, M., Schoeberl, M., and Douglass, A.: Stratosphere-troposphere exchange of mass and ozone, J. Geophys. Res, 109, doi:10.1029/2004JD005 186, 2004. 9649
Reithmeier, C.: Untersuchungen zum globalen Spurenstofftransport und Stratosphren-Troposphren Austausch mit dem Lagrangeschen Modell ECHAM4/ATTILA, DLR-Forschungsbericht, 2001-09, 178 pp., ISSN 1434-8454, Deutsches Zentrum für Luft- und Raumfahrt, Köln, Germany, 2001. 9648
Rex, M., Salawitch, R., von der Gathen, P., Harris, N. R. P., Chipperfield, M., and Naujokat, B.: Arctic ozone loss and climate change, 2004. 9647
Roeckner, E., Arpe, K., Bengtsson, L., Christoph, M., Claussen, M., Dümenil, L., Esch, M., Giorgetta, M., Schlese, U., and Schulzweida, U.: The atmospheric general circulation model echam4: Model description and simulation of present-day climate, MPI-Report, 218, Max-Planck-Institut, Hamburg, Germany, 1996. 9644
Roelofs, G.-J. and Lelieveld, J.: Tropospheric ozone siulation with a chemistry-general circulation model: Influence of higher hydrocarbon chemistry, J. Geophys. Res, 105, 22 697–22 712, 2000. 9654
Rogers, H., Teyssedere, H., Pitari, G., van Velthoven, V. G. P., and Sundet, J.: Model intercomparison of the transport of aircraft-like emissions from sub- and supersonic aircraft, Meteorol. Z., 11, 151–159, 2002. 9644

Rosenlof, K.: Seasonal cycle of the residual mean meridional circulation in the stratosphere, J. Geophys. Res., 100, 5173–5191, 1995. 9652

Schnadt, C., Dameris, M., Ponater, M., Hein, R., Grewe, V., and Steil, B.: Interaction of atmospheric chemistry and climate and its impact on stratospheric ozone, Clim. Dynamics, 18, 501–517, 2002. 9645

Steil, B., Dameris, M., Brühl, C., Crutzen, P., Grewe, V., Ponater, M., and Sausen, R.: Development of a chemistry module for gcms: First results of a multiannual integration, Ann. Geophys., 16, 205–228, 1998, SRef-ID: 1432-0576/ag/1998-16-205. 9644

Stenke, A.: Stratosphärischer Wasserdampf in einem gekoppelten Klima-Chemie Modell: Simulation, Trends und Bedeutung für die Ozonchemie, Ph.D. thesis, Ludwig-Maximilians Universität München, 2005. 9652

Stenke, A. and Grewe, V.: Impact of dynamically induced ozone mini-hole events on psc formation and chemical ozone destruction, Advances in Space Research, 33, 1062–1067, 2004. 9648

Timmreck, C., Graf, H.-F., and Feichter, H.: Simulation of Mt. Pinatubo volcanic aerosol with the Hamburg climate model echam4, Theor. Appl. Climatol., 62, 85–108, 1999. 9644

Wei, M.-Y.: A new formula on the exchange of mass and trace constituents between the stratosphere and troposphere, Q.J.R. Meteorol. Soc., 120, 3079–3086, 1987. 9649

World Meteorological Organization (WMO):, Scientific Assessment of ozone depletion: 1998, WMO Rep. 44, Geneva, 1999. 9642

Table 1. Areas of interest for the ozone origin. NH, TR, and SH describes Northern Hemisphere, Tropics and Southern Hemisphere. TS, LS, and MS denotes troposphere, lower stratosphere and mid stratosphere.

	Latitude	Pressure (hPa)
NHTS	90° N–30° N	1000–200
TRTS	30° N–30° S	1000–100
SHTS	30° S–90° S	1000–200
NHLS	90° N–30° N	200–100
TRLS	30° N–30° S	100–40
SHLS	30° S–90° S	200–100
NHMS	90° N–30° N	100–10
TRMS	30° N–30° S	40–10
SHMS	30° S–90° S	100–10

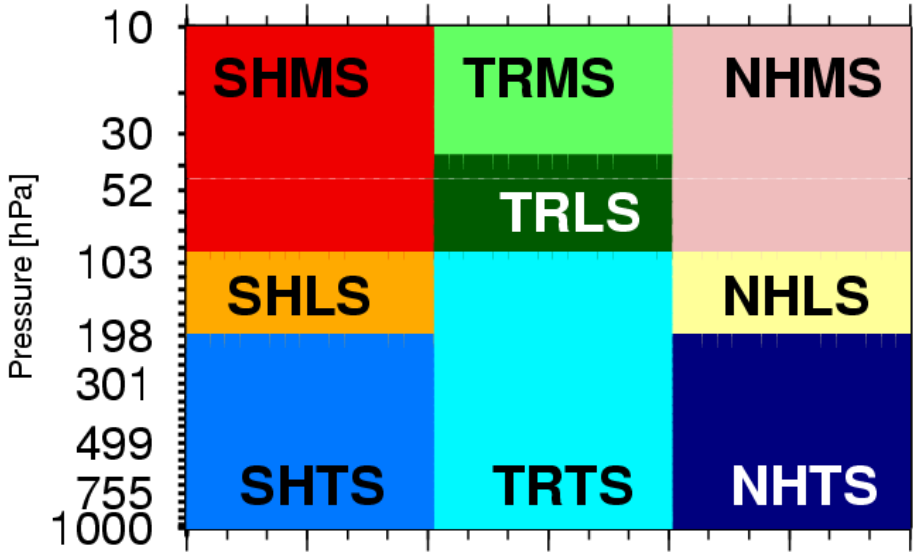

Fig. 1. Nine areas of ozone origin. The color index is also used for Figs. 4, 5 and 7. See also Table 1.

ACPD
5, 9641–9668, 2005

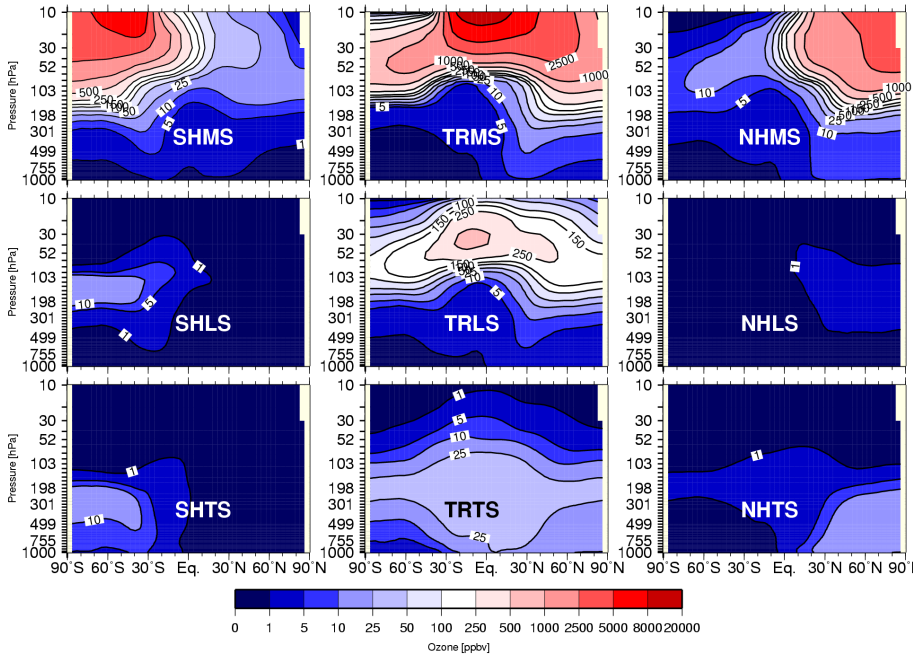

Fig. 2. Contributions of the nine ozone production regions to the total ozone content (ppbv) for January.

ACPD
5, 9641–9668, 2005

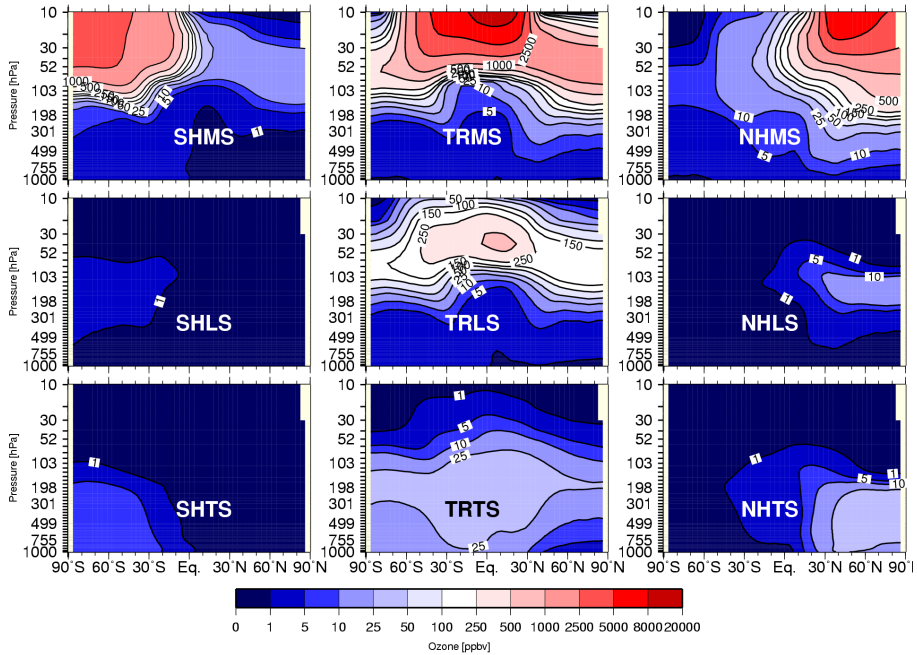

Fig. 3. Contributions of the nine ozone production regions to the total ozone content (ppbv) for July.

ACPD
5, 9641–9668, 2005

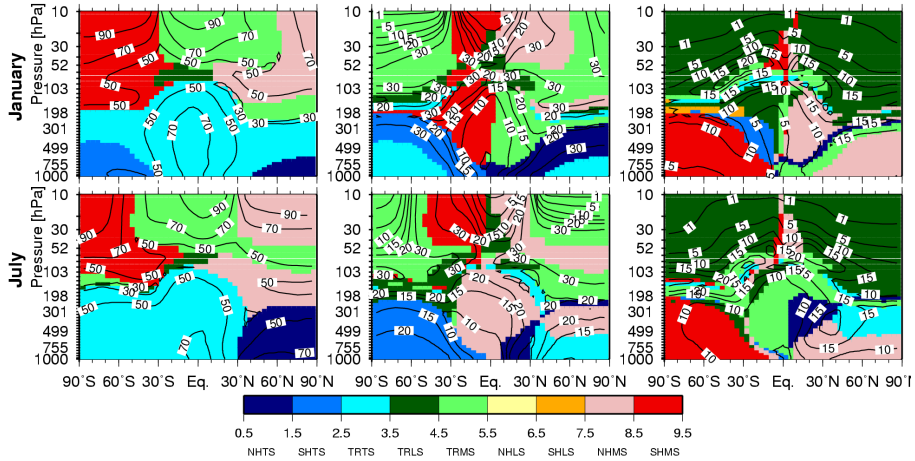

Fig. 4. Decomposition of the zonal mean ozone content. Main contribution (first column), second (second column) and third (third column) important contribution to the ozone content for January (top) and July (bottom). The color code (see Fig. 1) indicates the origin, the isolines its mixing ratio (ppbv). Isolines are only meaningful within one color area.

ACPD
5, 9641–9668, 2005

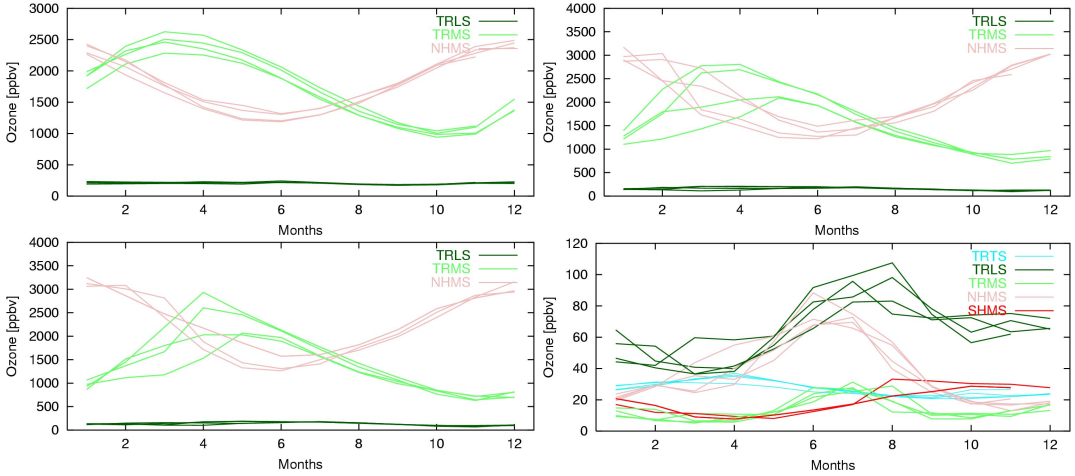

Fig. 5. Seasonal cycle of the ozone mixing ratios (ppbv) from various origin at 60° N and 50 hPa (first row), 90° N and 50 hPa (second row),90° N equivalent latitude and 50 hPa (third row),and the Equator at 76 hPa (bottom row). Four consecutive cycles are presented.

ACPD
5, 9641–9668, 2005

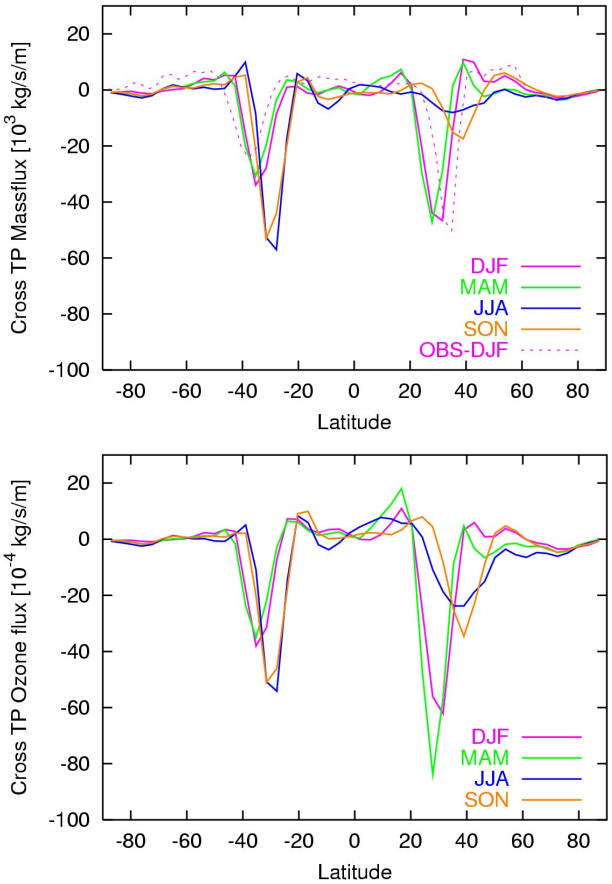

Fig. 6. Zonal mean air mass **(a)** and ozone **(b)** exchange (kg/m/s) through the thermal tropopause for the 4 seasons. The black line indicates DJF mass exchange derived from ERA-15 data, taken from (Grewe and Dameris, 1996).

ACPD
5, 9641–9668, 2005

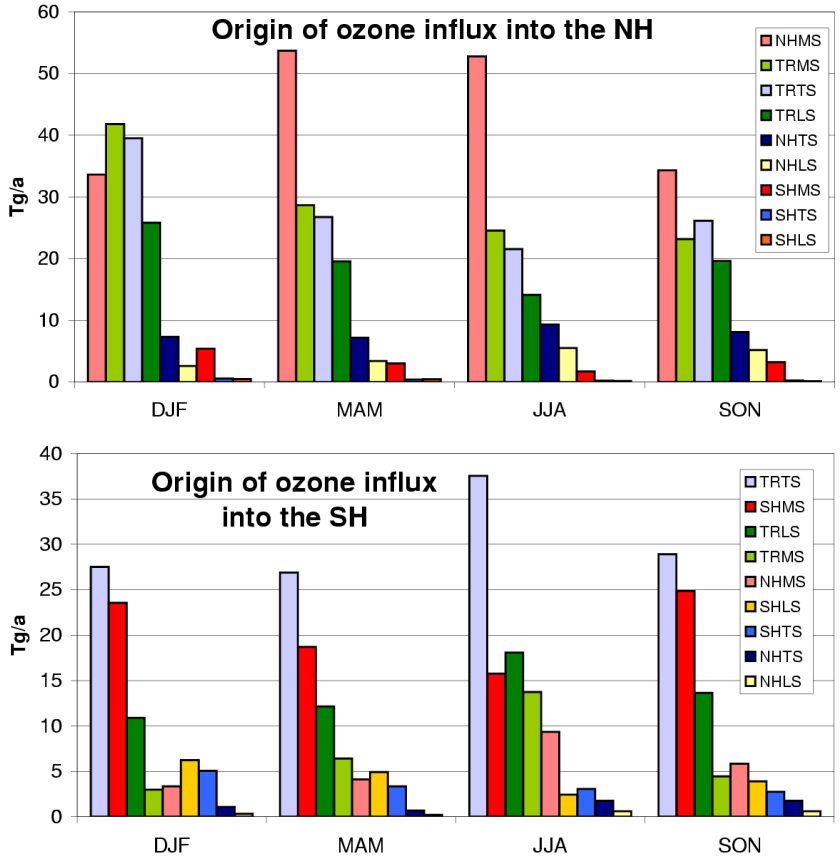

Fig. 7. Ozone influx from the stratosphere into the troposphere (Tg/a) for the Northern Hemisphere (top) and the Southern Hemisphere (bottom) splitted up into the regions of ozone origin. The regions are in the order of their importance regarding the total annual influx.

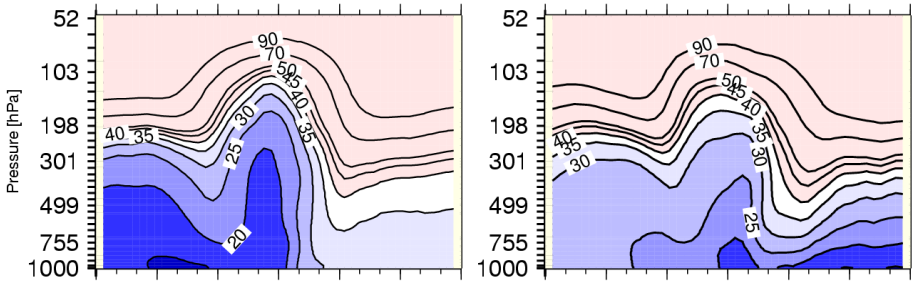

Fig. 8. Contribution (%) of stratospheric ozone to the total ozone for January (left) and July (right).

ACPD
5, 9641–9668, 2005