Dynamic-chemical coupling of the upper troposphere and lower stratosphere region

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Abstract. The importance of the interaction of chemistry and dynamics in the upper troposphere and lower stratosphere for chemical species like ozone is investigated using two chemistry-climate models. Species emitted in the upper troposphere, like NO_x (=$NO+NO_2$) by lightning or aircraft, have the chance to be transported into the lowermost stratosphere. Trajectory calculations suggest that the main transport pathway runs via the Inter Tropical Convergence Zone, across the tropical tropopause and then to higher latitudes, i.e. into the lowermost stratosphere. Longer lifetimes of NO_x in the lower stratosphere yield an accumulation of NO_x there, which feeds back on upper troposphere chemistry. This effect has been estimated for lightning NO_x emissions and reveals a contribution of at least 25% to 40% to the total northern hemisphere mid-latitude lightning increase of either NO_x and ozone.

1. Introduction

Increasing attention has been paid to effects of aircraft emissions [*Brasseur et al.*, 1998; *IPCC*, 1999] and lightning emissions [e.g. *Stockwell et al.*, 1999; *Grewe et al.*, 2000] on the chemical atmospheric composition. Lightning and aircraft are the only two direct NO_x (=$NO+NO_2$) emitters in the upper troposphere. Their impact on climate via production of ozone and destruction of CH_4 has been discussed on the basis of model simulations [*IPCC*, 1999]. Most of these models include detailed troposphere chemistry and some kind of upper boundary condition between 50 and 100 hPa to represent stratospheric chemistry. However, the impact of an upper boundary condition at these altitudes on estimates regarding the upper troposphere has not yet been investigated.

Holton et al. [1995] presented principal transport patterns concerning stratosphere-troposphere exchange: Isentropic mixing from the upper tropical troposphere to the mid-latitude lowermost stratosphere and large-scale

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tropical ascent and isentropic transport to higher latitudes are the main transport mechanisms by which upper troposphere tropical air masses may influence the lower stratosphere at higher latitudes. Estimation of the effects of emissions in the upper troposphere are strongly influenced by these transport pathways, since emissions of NO_x may accumulate in the lowermost stratosphere, where its lifetime greatly exceeds its tropospheric lifetime. The present paper aims at investigating the effect of a coupling of troposphere and stratosphere chemistry and dynamics. Using the Lagrangian transport algorithm ATTILA [*Reithmeier*, 2000] trajectory calculations provide information about the pathways of species emitted in the upper troposphere. The coupled chemistry-climate model ECHAM4.L39(DLR)/CHEM of the troposphere and lower stratosphere and the troposphere GISS chemistry-climate model will be used to investigate the importance of troposphere-stratosphere coupling, exemplary for lightning emissions.

2. Brief model descriptions

The Lagrangian transport scheme ATTILA [*Reithmeier*, 2000] has been developed on the basis of the chemistry transport model STOCHEM [*Collins et al.*, 1997]. It has been coupled to ECHAM4, in a way that full information is provided at every time-step. It includes state-of-the-art parameterizations for turbulent mixing within the boundary layer [*Maryon et al.*, 1991], diffusion [*Walton et al.*, 1988], and convection [*Brinkop and Sausen*, 1997].

The coupled climate-chemistry model ECHAM4.L39(DLR)/CHEM [*Hein et al.*, 2000; *Land*, 1999] is based on the spectral general circulation model (GCM) ECHAM4 [*Roeckner et al.*, 1996] with increased vertical resolution (700 m at tropopause altitudes). The chemical module [*Steil et al.*, 1998] has already been coupled to ECHAM3 and used in a variety of studies regarding the tropospheric and stratospheric chemistry [e.g. *Grewe et al.*, 1999]. It includes stratospheric heterogeneous and homogeneous ozone chemistry and tropospheric NO_x-CH_4-CO-OH-O_3-chemistry with 107 photochemical reactions and 37 species.

The GISS II' ('two prime') GCM [*Hansen et al.*, 1997] was developed from the GISS GCM version II. GISS II' has 9 'sigma' coordinate levels leading to a vertical resolution at the tropopause of 3 to 4 km. A chemistry module has been coupled to the GCM including 52 reactions of 24 species for tropospheric chemistry [*Shindell et al.*, 2000]. It describes mainly the same chemistry like CHEM, but also takes into account N_2O_5 hydrolysis on sulfate aerosol. Ozone, NO_x, and NO_y are prescribed

at 110 hPa.

Peroxyacetyl nitrate (PAN) and explicit non-methane hydrocarbon (NMHC) chemistry is omitted in both models. Surface NO_x emissions of 33.1 and 38.1 TgN per year, and aircraft NO_x emissions of 0.56 and 0.51 TgN per year are included in the ECHAM4.L39(DLR)/CHEM and the GISS model, respectively. The lightning parameterizations generate NO_x interactively with the convection scheme [*Price et al.,* 1994].

The two climate-chemistry models differ mainly in the vertical resolution, the treatment of stratospheric chemistry and the overall transport characteristics, and less in the description of tropospheric chemistry.

3. Transport of species emitted in the upper troposphere

To investigate the transport pathways of emitted species the Lagrangian transport scheme ATTILA has been applied on the basis of an ECHAM4 present-day climate simulation. Two different idealized emission sources and regions have been defined. An aircraft source (A) and a tropical lightning source (L), which both are located in the upper troposphere (150-300 hPa) but at different latitudes, i.e. 20°N-50°N and 20°S-20°N, respectively. For each region 10,000 air parcels have been released. After one year of integration 85 to 90% (Table 1) of the air parcels are in the troposphere (T) and 4 to 5% in the lowermost stratosphere (LMS). Since the regions themselves are of very different volume and mass, a mass related comparison offers the best basis for the interpretation of the results concerning the importance of upper troposphere emissions for these regions. The density of air parcels originating from the source regions A and L and the contribution to the total mass in the lowermost stratosphere is around 60% of the corresponding tropospheric contributions (Table 1). This clearly reveals the importance of the coupling of the lowermost stratosphere and upper troposphere for investigations concerning upper troposphere emissions.

Figure 1 shows how air parcels are transported from the two sources regions A and L to the lowermost stratosphere. Most of these air parcels (more than 85% for the aircraft source and more than 70% for the lightning source) are first transported downwards and then, at the Inter Tropical Convergence Zone (ITCZ), upwards into the lower tropical stratosphere. Meridional transport moves those air parcels to the lowermost stratosphere of both hemispheres. Therefore species emitted in the upper troposphere are mainly entering the lowermost stratosphere via transport across the tropical

Table 1

Figure 1

tropopause. They may then even return to the upper troposphere by a downward flux from the stratosphere to the troposphere in the extra-tropics.

4. Impact of lightning NO_x emissions on the chemistry

Considering these transport pathways, the effect of lightning NO_x emissions is studied with the ECHAM4.L39(DLR)/CHEM model and the GISS model, which have upper boundaries for NO_y and NO_x at 10 hPa and 110 hPa, respectively. Additionally, ECHAM4.L39(DLR)/CHEM has been applied with an upper boundary at 80 hPa, with values for NO_y derived from the simulation with the 10 hPa upper boundary. For each of those three models, 2 simulations for the year 1990 were performed: one with and one without emissions of NO_x from lightning. A 5 year simulation length is considered after a spin up time. Figure 2 views the results for nitrous oxide and ozone. It clearly shows that ECHAM4.L39(DLR)/CHEM allows for an accumulation of NO_x in the upper troposphere and lower stratosphere. Maximum relative changes are found in the tropical stratosphere and upper troposphere, where NO_x is more than four times higher than in the experiment without the NO_x lightning source. Ozone is increased by 70% in the upper tropical troposphere and also shows increases at higher latitudes. Introducing an upper boundary at 80 hPa strongly reduces the effect of lightning emissions. The maximum tropospheric NO_x increase is still at 200 hPa, but strongly reduced. However, below that region only small differences occur between the 2 simulations. Nitrogen oxides at southern and northern mid and high latitudes are much less influenced by lightning. The GISS model, which has the NO_x upper boundary at 110 hPa, shows a similar pattern of the lightning induced NO_x changes as the ECHAM4.L39(DLR)/CHEM model with the lowered upper boundary condition. In the tropics relative changes due to lightning are 50% less. However, absolute changes are similar. The increase at higher northern latitudes mainly results from lightning occurring in the extra-tropics. Ozone changes are only found in the tropics, peaking in the middle troposphere.

The differences in the calculated impact of the lightning NO_x emissions can largely be attributed to the suppressed transport mechanisms, as shown in the previous section. The contribution of this troposphere-stratosphere dynamical-chemical coupling is estimated in Table 2. In the extra-tropical upper troposphere (225 hPa to 350 hPa) this contribution to the enhancement of the NO_x and NO_y due to lightning accounts

Figure 2

Table 2

for roughly 25% to 35% and 25% to 40%, respectively. Its contribution to an ozone increase is about 35% to 40% in that region. These values are much lower in the tropical region.

More than a third of the increase of ozone due to lightning at northern mid and high latitudes can therefore be attributed to tropical NO_x emissions, which is 10% of the NO_x concentration there. In the southern hemisphere it is even 15%, since the background NO_x concentration is smaller, but the coupling mechanism effects uniformly both hemispheres.

Taking into account that the lowered upper boundary (80 hPa) still allows some transport from the tropical troposphere to the lowermost stratosphere, these estimates can only be lower limits for the ECHAM4.L39(DLR)/CHEM. This is supported by the GISS model, which allows no such transports and shows no significant ozone changes at mid latitudes.

5. Discussion and Conclusion

Applying a Lagrangian trajectory model, we showed that species emitted in the northern upper troposphere, e.g. NO_x by aircraft or lightning, have the chance to reach the lowermost stratosphere of both hemispheres. The species are transported downwards and southwards to the tropics and then in the ITCZ into the tropical stratosphere and afterwards to the lowermost stratosphere. Almost all trajectories from the northern upper troposphere to the lowermost stratosphere showed this pathway. For the trajectory simulations, no sinks have been taken into account, since we only focused on the principal transport pathways. The dynamical-chemical simulations, which include sinks and sources for nitrogen species, clearly show the long range impacts. This transport pathways give a reasonable explanation of how species released by aircraft, mainly flying on the northern hemisphere, affect the southern lowermost stratosphere, as shown with aviation fuel tracer simulations [*Danilin et al.*, 1998] and aircraft NO_x simulations [*Grewe et al.*, 1999].

By lowering the upper boundary from 10 hPa to 80 hPa, the effect of this transport mechanism on the chemical impact of lightning has been investigated. It clearly shows a strong impact on upper troposphere NO_x and ozone in the extra-tropical upper troposphere, indicating that the coupling of the upper troposphere with the lower stratosphere is essential for estimates of the chemical impacts of emissions, released in the upper troposphere. Since the trajectories pass the lower troposphere, the coupling mechanism is also relevant for surface emissions, at least in the tropics.

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Figure 1. Composites of trajectories which start in the source regions A (left) and L (right), then are transported to the tropical (20°S to 20°N) troposphere (air pressure more than 400 hPa) and then end in the lowermost stratosphere. Mean trajectories are calculated for each of these three paths and afterwards combined to a single trajectory. The composites include 70%, 100%, 71%, and 67% of all trajectories from the source region A to NLMS, A to SLMS, L to NLMS, and L to SLMS, respectively (see Table 1).

Figure 1. Composites of trajectories which start in the source regions A (left) and L (right), then are transported to the tropical (20°S to 20°N) troposphere (air pressure more than 400 hPa) and then end in the lowermost stratosphere. Mean trajectories are calculated for each of these three paths and afterwards combined to a single trajectory. The composites include 70%, 100%, 71%, and 67% of all trajectories from the source region A to NLMS, A to SLMS, L to NLMS, and L to SLMS, respectively (see Table 1).

Figure 2. Annual mean relative changes [%] of NO_x (left) and ozone (right) due to lightning NO_x emissions. Results are shown for the ECHAM4.L39(DLR)/CHEM models with an upper boundary at 10 hPa (top) and 80 hPa (middle) and for the GISS model (bottom) with an upper boundary at 110 hPa. Changes are calculated relative to a simulation without lightning emissions. All ECHAM4.L39(DLR)/CHEM results are significant at a 99% level. In the GISS model, the light and dark shading indicates the 95% and 99% significant levels (t-test). The tropical changes of NO_x due to lightning in the both models with a low upper boundary have similar absolute changes.

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Table 1. Share of particles [%], its density [air parcels per air mass], and share of mass within the target regions [%], which are transported from the source region A (150-300 hPa, 20°N-50°N) and L (150-300 hPa, 20°S-20°N) to the stratosphere 'S' ($\leq$ 150 hPa), northern lower-most stratosphere 'NLMS' (150-250 hPa, 20°N-90°N), southern lower-most stratosphere 'SLMS' (150 - 250 hPa, 20°S-90°S), and the troposphere 'T' (rest).

		Region	T	NLMS	SLMS	S
Share of	[%]	A	87.9	2.3	2.0	7.8
particles	[%]	L	86.0	2.5	2.5	9.0
Particle	$[(10^{16}\text{kg})^{-1}]$	A	21	14	12	10
density	$[(10^{16}\text{kg})^{-1}]$	L	21	14	14	10
Mass	[%]	A	3.6	2.3	2.0	1.7
fraction	[%]	L	5.6	3.8	3.9	3.1

Table 2. Relative contribution [%] of the troposphere-stratosphere coupling to the lightning induced enhancement of NO_x, NO_y, and ozone in the upper troposphere (225-350 hPa). The contribution [%] of the troposphere-stratosphere coupling relative to the background concentration is given in brackets.

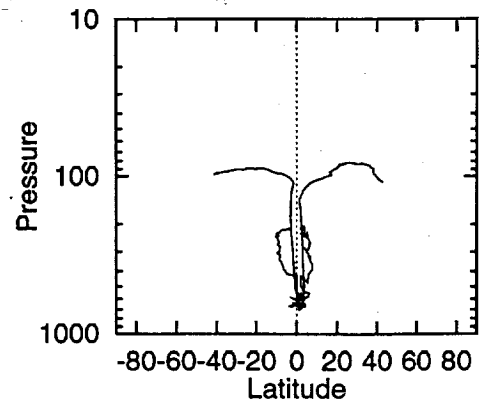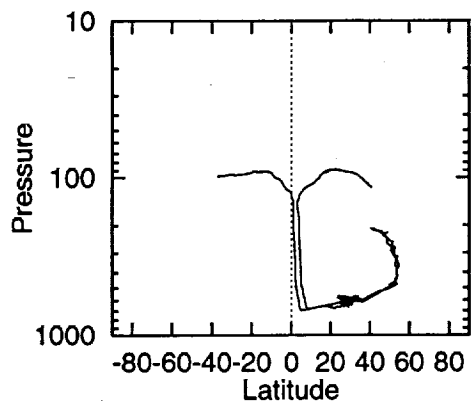|	90°N -60°N	60°N -30°N	30°N -30°S	30°S -60°S	60°S -90°S
NO_x	34 (10)	26 (10)	5 (4)	19 (14)	23 (14)
NO_y	39 (8)	26 (9)	8 (7)	22 (15)	26 (14)
O_3	41 (6)	34 (7)	17 (8)	28 (11)	29 (10)

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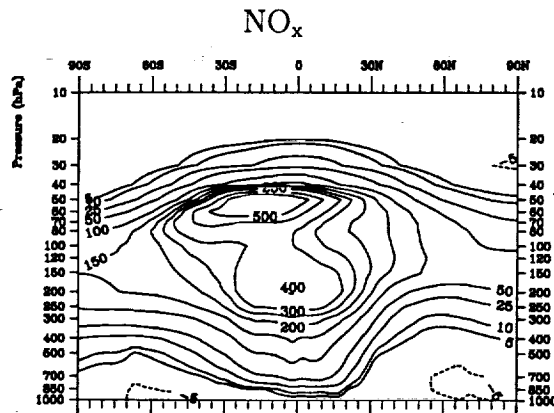
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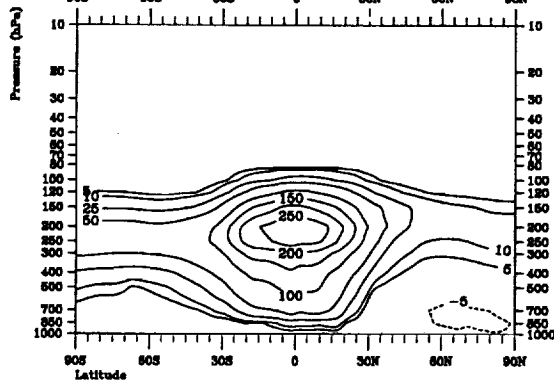
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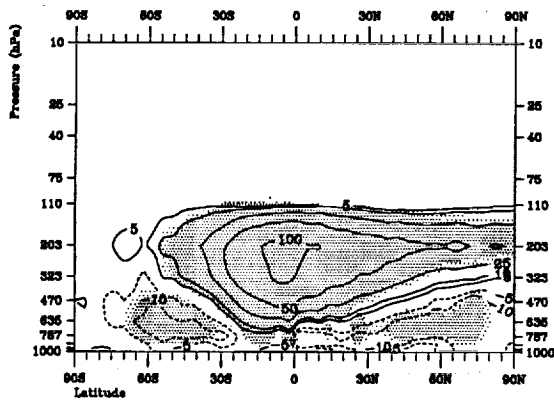
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