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# Carbon monoxide as a tracer for tropical troposphere to stratosphere transport in the Chemical Lagrangian Model of the Stratosphere (CLaMS)

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#### **Abstract**

Variations in the mixing ratio of trace gases of tropospheric origin entering the stratosphere in the tropics are of interest for assessing both troposphere to stratosphere transport fluxes in the tropics and the impact on the composition of the tropical lower stratosphere of quasi-horizontal in-mixing into the tropical tropopause layer from the mid-latitude stratosphere. Here, we present a simplified chemistry scheme for the Chemical Lagrangian Model of the Stratosphere (CLaMS) for the simulation, at comparatively low numerical cost, of CO, ozone, and long-lived trace substances (CH<sub>4</sub>, N<sub>2</sub>O<sub>1</sub> CCl<sub>2</sub>F, and CO<sub>2</sub>) in the lower tropical stratosphere. The boundary conditions at the ground are represented for the long-lived trace substances CH<sub>4</sub>, N<sub>2</sub>O, CCl<sub>2</sub>F, and CO<sub>2</sub> based on ground-based measurements. The boundary condition for CO in the free troposphere is deduced from MOPITT measurements. We find that the zonally averaged tropical CO anomaly patterns simulated by this model version of CLaMS are in good agreement with observations. The introduction of a new scheme in the ECMWF integrated forecast system (Tompkins et al., 2007) for the ice supersaturation after September 2006, results in a somewhat less good agreement between observed and simulated CO patterns in the tropical lower stratosphere after this date.

#### 1 Introduction

Carbon monoxide (CO) has sources at the Earth's surface mostly from incomplete combustion of fossil fuels and from biomass burning. CO is also chemically produced in the troposphere by the oxidation of methane and non-methane hydrocarbons and is removed from the atmosphere by oxidation with OH, leading to a lifetime ranging between several weeks to a few months in the lower atmosphere; the life time in the stratosphere is a few months (Schoeberl et al., 2006). The CO concentration in the troposphere is further determined by transport processes. Because of its relatively long lifetime CO is useful as an indicator of long-range transport of polluted air-masses. In particular,

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because of the low background mixing ratios of CO in the lower stratosphere, CO is an important tracer of troposphere to stratosphere transport, during strong convective activity (Ricaud et al., 2007), within the Asian monsoon circulation (Park et al., 2007, 2008), and during slow ascent and mixing at the bottom of the tropical tropopause layer (TTL) (Konopka et al., 2007; Schoeberl et al., 2006; Fueglistaler et al., 2009; Homan et al., 2010). Moreover, it has been observed that seasonal variations in the mixing ratio of CO entering the stratosphere at the bottom of the TTL are carried upward with the rising air in the tropics with the signal being observable throughout the tropical lower stratosphere (Schoeberl et al., 2006; Randel et al., 2007; Liu et al., 2007), a phenomenon commonly referred to as "atmospheric tape recorder" (Mote et al., 1996).

Here we describe an extension of the Chemical Lagrangian Model of the Stratosphere (CLaMS) (McKenna et al., 2002b,a) that allows multi-annual simulations of CO in the tropical upper troposphere and lower stratosphere to be made for investigations of phenomena like the atmospheric tape recorder in CO. CLaMS was originally designed as a model for stratospheric polar chemical ozone loss (McKenna et al., 2002a; Grooß et al., 2002; Konopka et al., 2004; Grooß and Müller, 2007) and was recently extended to the analysis of transport processes in the tropopause region, both in the extratropics (Pan et al., 2006; Vogel et al., 2011) and in the tropics (Konopka et al., 2007, 2010). This version of CLaMS was recently successfully applied to describe the irregular tape-recorder pattern discovered for HCN (Pumphrey et al., 2008; Pommrich et al., 2010).

The model concept used here is rather different from chemical models of the stratosphere and troposphere that employ a comprehensive description of the tropospheric chemistry and a wide range of emissions of trace substances at the ground (e.g., Jöckel et al., 2006; Garcia et al., 2007; Schultz et al., 2008; Morgenstern et al., 2009). In contrast, we make use of CO measurements from the Measurements Of Pollution In The Troposphere (MOPITT) satellite experiment (Deeter et al., 2003) for formulating a lower boundary condition for CO for multi-annual simulations of upper tropospheric and lower stratospheric transport of CO in the tropics. Together with boundary conditions at the

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ground for the long-lived species  $CH_4$ ,  $N_2O$ ,  $CCI_3F$ , and  $CO_2$  and a simplified chemistry scheme (with prescribed radical concentrations) this allows successful simulations of CO in the lower tropical stratosphere to be made at comparatively low numerical cost. We present simulations of CO using the CLaMS chemical transport model over the time period from October 2001 to the end of 2008 for the tropical lower stratosphere and compare the results with both remote sensing and in-situ measurements. We find that this model version of CLaMS allows a successful simulation of the CO anomaly pattern in the lower tropical stratosphere.

### 2 Measurements of tropical CO

We use several sources for tropical CO data, both for prescribing boundary conditions for a multi-annual CLaMS simulation and for evaluating the results of the simulation. We use data from the MOPITT instrument, a nadir looking satellite experiment (Deeter et al., 2003), from the Microwave Limb Sounder (MLS), a microwave limb viewing instrument (Waters et al., 2006) and in-situ measurements from the Cryogenically Operated Laser Diode (COLD) spectrometer (Viciani et al., 2008) onboard the high-flying research aircraft Geophysica.

The MOPITT remote sensing instrument was launched aboard the EOS Terra satellite on 18 December 1999; routine measurements began in March 2000. Terra is in a sun-synchronous orbit with an inclination of approximately 98.2°. MOPITT views the Earth over all latitudes with a pixel size of 22 × 22 km and a cross-track swath that provides a near-global measurement of the distribution of CO every three days. Operational MOPITT CO products in version 3 are based on 4.7 µm thermal-channel radiances. The information content in MOPITT retrievals is quantified through the Degrees of Freedom for Signal (DFS) which indicates the number of independent pieces of information in the retrieved profile (Deeter et al., 2004). In the tropics and the mid-latitudes the DFS is in general greater then 1, which give some profile shape information, while a DFS of 1 indicates that the profile contains as much information as the column. The

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maximum sensitivity is located around ~ 5 km, 500 hPa (Deeter et al., 2003). Here we use the level 2 data of the version 3 retrievals of MOPITT CO that are extensively validated (e.g., Emmons et al., 2004; Emmons et al., 2007; Luo et al., 2007; Emmons et al., 2009, and references therein). Since 2009 the version 4 of MOPITT CO data exists, described by Deeter et al. (2010).

MLS is one of four instruments on the EOS Aura satellite which was launched on 15 July 2004. MLS is viewing the Earth's limb in the orbit plane of the Aura spacecraft. Observations range from 82° S to 82° N every day and are spaced 140 km apart along the ground track. MLS detects thermally emitted radiation in several bands of the sub-millimeter spectral region yielding profiles of temperature, geopotential height and the mixing ratio of more than fourteen chemical species. The mixing ratio of CO is obtained from measurements of the spectral line at 230 GHz. Here we use version 2.2 retrievals. The MLS retrieval technique is described in detail by Livesey et al. (2006) and the validation of the 2.2 CO data product described by Pumphrey et al. (2007) and Livesey et al. (2008).

The COLD spectrometer is a Tunable Diode Laser (TDL) instrument designed for operation on a high-altitude aircraft for the measurement of CO (Viciani et al., 2008). The absolute in-flight accuracy for CO is  $\sim 6\%-9\%$ . The precision is given by the value of either 1% of the mixing ratio or 7–8 ppb, depending on which value is the largest. The time resolution of the data is 4 s which corresponds to a spatial resolution of about 800 m for an average airplane cruising speed of 750 km h<sup>-1</sup> (Viciani et al., 2008).

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### 3.1 The Chemical Lagrangian Model of the Stratosphere (CLaMS)

### 3.1.1 CLaMS transport

CLaMS is a chemical transport model designed to simulate chemistry, advection, and mixing in the stratosphere and upper troposphere (McKenna et al., 2002a,b; Konopka et al., 2007, 2010). Because of the Lagrangian representation of transport, with the intensity of mixing in model transport being driven by the strength of flow deformation, the model should be particularly well suited for the simulation of tracer transport in the vicinity of strong transport barriers and the associated tracer gradients. The specific model setup employed here follows closely the setup described by Konopka et al. (2007, 2010). The horizontal resolution is 100 km and the vertical resolution in the tropospause region about 400 m. Multi-annual, global CLaMS simulations of the whole troposphere and stratosphere (from the ground up to the stratopause around 2500 K potential temperature) cover the time period from October 2001 to the end of 2008.

The model employs a hybrid coordinate ( $\zeta$ ) that corresponds to isentropic coordinates above  $p_r = 100 \, \text{hPa}$  and smoothly transforms to a pressure based vertical coordinate below  $p_r$  (Mahowald et al., 2002). The hybrid coordinate  $\zeta$  is defined as

$$\zeta(p) = f(\eta)\theta(p,T(p)), \quad \eta = \frac{p}{p_0}$$
(1)

with

$$f(\eta) = \begin{cases} \sin\left(\frac{\pi}{2} \frac{1-\eta}{1-\eta_r}\right) & \eta > \eta_r \\ 1 & \eta \le \eta_r, \quad \eta_r = \frac{\rho_r}{\rho_0}, \quad \rho_0 = 1000 \text{hPa} \end{cases}$$
 (2)

The use of this coordinate allows the tropospheric upwelling as it is represented in the pressure tendencies  $(\dot{p}=\omega)$  of the ECMWF operational analysis to be represented

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in the CLaMS transport (see Konopka et al., 2007, for details). However, a parametrization of subgrid scale convectively driven vertical transport, as it is present for example in the ECMWF model, is not included in this model version of CLaMS. Furthermore, in order to obtain mass-conserving transport on an annual scale the calculated vertical velocities  $\dot{\zeta}$  are corrected (Rosenlof, 1995; Konopka et al., 2010). In this way zonally and annually averaged total mass fluxes vanish across each  $\zeta(\lambda)$  isoline ( $\lambda$ , latitude).

### 3.1.2 Simplified chemistry scheme

The CLaMS model contains a detailed comprehensive stratospheric chemistry scheme including catalytic ozone loss cycles as well as heterogeneous chemistry on the surfaces of stratospheric aerosol particles and polar stratospheric clouds (McKenna et al., 2002a; Wegner et al., 2011). Here, the focus is on a limited set of trace species in the TTL, in particular on CO. Therefore, a strongly simplified chemistry is implemented in this model version, which is designed to describe reasonably well the first order chemical loss of long-lived tracers, ozone production and chemical loss in the TTL, and the production and loss of stratospheric CO. There are four long lived tracers considered in the simplified chemistry scheme, namely CH<sub>4</sub>, N<sub>2</sub>O, CCl<sub>3</sub>F, and CO<sub>2</sub>. Ploeger et al. (2011) recently showed that the O<sub>3</sub> budget in the upper TTL and lower stratosphere is well represented employing this simplified chemistry scheme. The reactions

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Rate constants are deduced from current recommendations (Sander et al., 2006). The photolysis rates are calculated as diurnal averages using the CLaMS photolysis code (Becker et al., 2000). Products other than CO,  $O_3$  or the four considered long-lived tracers are ignored in this approach.

Reaction (R9) constitutes the rate limiting step of the  $HO_x$  catalytic ozone loss cycle (Reactions R9–R10) in which two ozone molecules are destroyed ( $2O_3 \rightarrow 3O_2$ ). Thus, Reaction (R9) is assigned twice the recommended value to represent the ozone loss through this cycle and in turn, the rate of Reaction (R10) is set to zero. By this approach and by accounting for photochemical production of ozone through the photolysis of molecular oxygen Reaction (R8), the most important reactions that determine ozone concentrations in the lowermost stratosphere are represented in the model.

The concentrations of the radicals CI,  $O(^1D)$ , OH, and  $HO_2$  are prescribed as tabulated values as a function of latitude and pressure level for each month. There are two options. First, values for these radicals are deduced from diurnal averages of the Mainz 2-D photochemical model (Grooß, 1996). Second, concentrations of methane and ozone are taken from the HALOE climatology (Grooß and Russell, 2005) and the

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radical concentrations are then calculated from a simulation of the box model version. of CLaMS including the complete chemistry scheme (McKenna et al., 2002b) for each pressure level, longitude and month. These tabulated climatologies of Cl. O(1)D. OH and HO<sub>2</sub> are available as electronical supplement of this paper, as they are considered as the most reliable. However, the model results shown here are based on diurnal averages of the Mainz 2-D photochemical model.

### Boundary conditions for CO and long-lived tracers at the top and bottom of the model domain

The model setup presented here is aimed at using CO as a tracer of tropospheric air transported into the TTL, rather than investigating CO in the boundary layer and the detailed impact of tropospheric sources. Therefore, CO is prescribed in the free troposphere based on MOPITT measurements. We use the MOPITT measurements at 500 hPa, where MOPITT provides reliable measurements (Deeter et al., 2004; Emmons et al., 2004). Although reported at 500 hPa, due to the rather wide averaging kernels of MOPITT (Deeter et al., 2004), these values are influenced by the CO values in a thick layer (~ 200-700 hPa) in the free troposphere. MOPITT only reaches global coverage after three days and in addition the density of measurements is often limited by cloud occurrence. Therefore, we use trajectory calculations from the MOPITT measurement locations during a five day period, forward and backward, to 12:00 UTC on the central day of the five day period. The data are binned into a 2° × 6° latitude/longitude grid and then averaged on each grid point location weighted by the distance to the corresponding grid point location to construct the boundary condition for the central day. An example of the CO field on a regular grid derived in this way for 2 January 2008 is shown in Fig. 1.

The model levels below 500 hPa are set to the CO values determined through the trajectory procedure described above, except in the lowest model level, where CO measurements from the NOAA/CMDL ground-based measurement network are prescribed **GMDD** 

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(Novelli et al., 2003)<sup>1</sup>. The CO fields deduced in this way, however, underestimate the CO total column measured by MOPITT. Thus, the prescribed CO values are corrected (scaled up by 20%) so that the CO column in the model agrees more closely with the MOPITT CO column measurement. CO at the upper boundary ( $\theta = 2500 \, \text{K}$ ) is set 5 according to the Mainz-2D model (Grooß, 1996).

For the long lived tracers, CO<sub>2</sub>, CH<sub>4</sub>, CCl<sub>3</sub>F, and N<sub>2</sub>O boundary conditions are prescribed as mixing ratios in the lowest model level. For CO<sub>2</sub> and CH<sub>4</sub> the measurements from the NOAA/CMDL ground-based measurement network are used (Masarie and Tans, 1995; Novelli et al., 2003).

CCI<sub>3</sub>F and N<sub>2</sub>O are derived from the in situ CATS instruments (CATS-Chromatograph for Atmospheric Trace Species) being in continuous operation at six NOAA baseline observatories since 1999<sup>2</sup>. At the upper boundary CCl<sub>3</sub>F is set to zero, whereas the HALOE climatology (Grooß and Russell, 2005) and the Mainz-2D model (Grooß, 1996) are used for CH<sub>4</sub> and N<sub>2</sub>O, respectively.

In addition, for the interpretation of the data and for the definition of the upper boundary of CO<sub>2</sub>, the mean age  $\Gamma(\lambda,\phi)(\lambda,\text{latitude},\phi,\text{longitude})$  is used. The mean age  $\Gamma$  is determined by considering a linear increasing tracer in the boundary layer.  $\Gamma$  is calculated as the time lag, at a given time, between the stratospheric tracer value and the tracer value in the boundary layer (Waugh and Hall, 2002).  $\Gamma$  at  $\zeta = 2500 \,\mathrm{K}$  is derived from MIPAS observations of SF<sub>6</sub> (Stiller et al., 2008). The upper boundary of  $CO_2$  is defined through:  $[CO_2](\lambda, \phi, t) = \langle CO_2 \rangle (t - \Gamma(\lambda, \phi))$ , where  $\langle CO_2 \rangle$  denotes the mean value of CO<sub>2</sub> in the tropics (zonally averaged in the latitude range ±30°) calculated from the NOAA/CMDL ground-based measurement network. Thus, at the upper boundary, the CO<sub>2</sub> distribution is derived from that in the tropical boundary layer that was observed in the past quantified in terms of the mean age.

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<sup>1</sup>see also http://www.esrl.noaa.gov/gmd/ccgg/globalview/co/co\_intro.html

<sup>&</sup>lt;sup>2</sup>http://www.esrl.noaa.gov/gmd/hats/insitu/cats/

The volume mixing ratio of ozone at the surface is set to zero, between the surface and  $\theta = 500\,\mathrm{K}$  the volume mixing ratio of ozone is calculated based on the simplified chemistry scheme described earlier in this paper, above  $\theta = 500\,\mathrm{K}$  ozone is prescribed from the HALOE climatology (Grooß and Russell, 2005).

#### 4 Results

### 4.1 Comparison of simulated CO with measurements

#### 4.1.1 MOPITT measurements

In Fig. 2, we show a comparison of tropical CLaMS CO simulations with MOPITT measurements. CLaMS values are convoluted with the MOPITT averaging kernels and, like the MOPITT measurements, zonally averaged over the latitude band  $\pm 15^{\circ}$ . There is in general good agreement, particularly the seasonal variation of tropical CO is well represented. At 500 hPa ( $\zeta\approx 250\,\text{K}$ ) the average CO mixing ratios are close to the MOPITT measurements, at the levels above correspondence between the simulations and the measurements is less good. After end of 2006 there are bigger differences between the maxima (even at 250 K) than in the years before, the correspondence for the lower values is equally good, as before 2006; the problems after 2006 are probably due to the introduction of a new ice supersaturation scheme in the ECMWF integrated forecast system (Tompkins et al., 2007) in September 2006. We discuss this point in the next section.

In Fig. 3, we show examples of a comparison of the results of the CLaMS simulations of CO anomaly with MOPITT measurements on the model surface  $\zeta=350 \, \mathrm{K}$ ;  $\sim 150 \, \mathrm{hPa}$ , close to the mean convective outflow level in the tropics. CLaMS values are convoluted with the MOPITT averaging kernels for an appropriate comparison. The overall agreement is satisfying for all the years. Differences occur mainly in the tropics. However, it is not surprising that the model shows a deficit in the tropics, because the

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upward transport is represented in the model solely by the large-scale vertical winds  $(\omega = \dot{p})$  which do not contain information of sub-grid scale, convectively driven vertical transport.

#### 4.1.2 MLS measurements

An important test for the ability of a model to describe transport and chemistry of longer lived tropospheric tracers like CO in the tropical upper troposphere and lower stratosphere is the temporal development of anomaly signals in the air ascending in the tropics, commonly referred to as "atmospheric tape recorder" (Mote et al., 1996; Schoeberl et al., 2006). For a successful reproduction of the CO tape recorder pattern, a model needs to be able to describe correctly the seasonality of CO in the upper troposphere, the transport within the troposphere to the bottom of the TTL, where the seasonal CO pattern is imprinted on the ascending air (or where, in other words, the "tape head is written"). Further, the model needs to be driven with the appropriate vertical velocities for the upward transport in the tropical lower stratosphere without the anomaly pattern being destroyed by an excessive vertical or horizontal diffusion or incorrect chemistry. Finally, the extent of horizontal in-mixing into the TTL, which affects tape recorder patterns (Konopka et al., 2010) needs to be correctly described.

In Fig. 4, the simulated anomaly pattern of tropical, zonally averaged CO is shown, i.e. the CO "tape recorder". For an appropriate comparison, the CLaMS values were vertically smoothed with a top hat function (with a width of ~3 km) to approximately take into account the field of view of the MLS instrument. The simulation successfully reproduced the observed approximately one-year tape recorder pattern of CO (Schoeberl et al., 2006; Liu et al., 2007). Further, the onset of enhanced as well as of reduced CO volume mixing ratios are well described. However, after September 2006, the agreement between the CLaMS simulations of the CO anomaly pattern and the MLS CO observations is less good than before. In September 2006 a new ice supersaturation scheme was introduced in the ECMWF integrated forecast system (Tompkins et al., 2007). Therefore, we suggest that the impact of the new ice supersaturation

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scheme on temperatures in the tropical lower stratosphere is responsible for the less good agreement after this date. Due to the introduction of the new scheme, the water vapour in the lower stratosphere increases in the operational analyses of ECMWF and, as a consequence, the air temperature increases. Higher temperatures in the TTL mean smaller values of the diabatic heating due to the long-wave radiation derived in CLaMS from the Morcrette scheme under clear sky conditions (Morcrette, 1991). Consequently, tropical upwelling in the TTL decreases. We suggest that these lower vertical velocities are responsible for the underestimation of the CO concentrations in the lower tropical stratosphere after September 2006.

#### 4.1.3 COLD measurements

Because of their high spatial and temporal resolution, the in-situ measurements of CO from the COLD instrument allow the quality of the vertical structure of CO in the CLaMS simulations to be evaluated. In Figs. 5 and 6, we compare the vertical CO profile measured by COLD during two flights of the TROCCINOX campaign. The flight on 5 February 2005 was planned in order to sample air masses influenced by both, fresh and aged convection while the flight of 15 February 2005 was a survey flight of the TTL aimed at sampling air at background conditions. The comparison for the flight on 5 February shows that in the CLaMS simulation, the low stratospheric mixing ratios of CO at altitudes greater than about 380 K are reproduced reasonably well. However, the strong enhancement of CO at altitudes below, with peak values of CO of about 130 ppb at 340-380 K are substantially underestimated. We attribute the strong CO enhancement at 340-380 K to air that is lofted by convection to these altitudes and thereafter injected into the upper troposphere in the convective outflow region. Indeed, while young air masses, with a simulated mean age of air of about half a month, are observed at ≈ 350 K the simulated mean age of air is not compatible with fresh convection. Consequently, we interpret the observed model underestimate of CO as being caused by the lack of a convective parametrization in CLaMS. The model produces some signature of convectively driven upward transport as a peak in the CO distribu**GMDD** 

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tion at about 330 K that coincides with very young air masses, recently being lofted from the ground. Thus, the ECMWF vertical winds ( $\dot{p}$ ) employed in the CLaMS simulation discussed here contain some signature of the convective activity that was sampled during the Geophysica flight on 5 February 2005, albeit a much weaker one than in reality.

The reproduction by the CLaMS simulations of CO mixing ratios measured on 15 February is much more successful than the simulation for 5 February. The agreement is generally reasonable with both the decline of CO with altitude above about 350 K and the enhancement in the upper troposphere being reproduced in the simulation. There is some indication of an underestimate of the peak CO in the convective outflow region, but much less pronounced than for the flight on 5 February. Therefore, we conclude that the lack of a representation of subgrid scale convectively driven upward transport of trace species in the troposphere in CLaMS does not substantially impact the simulation of CO for tropical background conditions. For a successful representation of the impact on CO of individual convective events, such as observed for the flight on 5 February 2005, a model resolution at the spatial scales of convective events is necessary.

#### 5 Conclusions

We presented a simplified chemistry scheme for the chemical transport model CLaMS for the simulation of CO, ozone, and long-lived trace substances (CH $_4$ , N $_2$ O, CCl $_3$ F, and CO $_2$ ) in the lower tropical stratosphere at comparatively low numerical cost. The boundary conditions at the ground are represented for the long-lived trace substances CH $_4$ , N $_2$ O, CCl $_3$ F, and CO $_2$  based on ground-based measurements. The boundary conditions for CO in the free troposphere are based on MOPITT measurements. The focus of this work is on the quality of the simulated CO fields (see Ploeger et al., 2011, for a discussion of the simulated ozone budget in the TTL). Comparison with in-situ measured CO profiles shows good agreement for background conditions, while

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signatures of recent convective events are underestimated. We conclude that the lack of a parametrization of convectively driven upward transport in the tropical troposphere does not substantially impact the quality of the simulation of CO for background conditions in the tropical troposphere. Comparison of tropical CLaMS CO simulations with MOPITT measurements in the upper troposphere (500–250 hPa) show overall good agreement. However, this agreement, in particular for the winter maximum CO values, becomes worse after the end of 2006. We attribute the worse performance of the model under these conditions to the introduction of a new supersaturation scheme of the ECMWF (Tompkins et al., 2007) which leads to higher water vapour concentrations in the tropical tropopause region, thus to higher temperatures and, consequently, to lower tropical ascent rates. Further, we find that the zonally averaged tropical CO anomaly patterns simulated by this model version of CLaMS are in good agreements with observations.

Supplementary material related to this article is available online at: http://www.geosci-model-dev-discuss.net/4/1185/2011/gmdd-4-1185-2011-supplement.zip.

Acknowledgements. The authors thank both the MOPITT and the MLS team for providing such great research products publically available. We thank Felix Ploeger for helpful comments on the manuscript. TROCCINOX was partially funded by the Commission of the European Community under the contract EVK2-CT-2001-00122 and other TROCCINOX partners.

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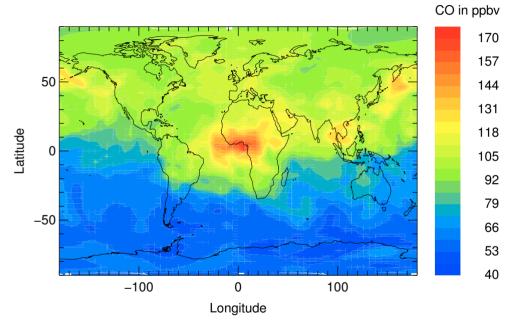
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**Fig. 1.** Mixing ratios of CO on the 500 hPa level on 2 January 2008, as derived from five days of MOPITT measurements by a trajectory technique. (See text for details).

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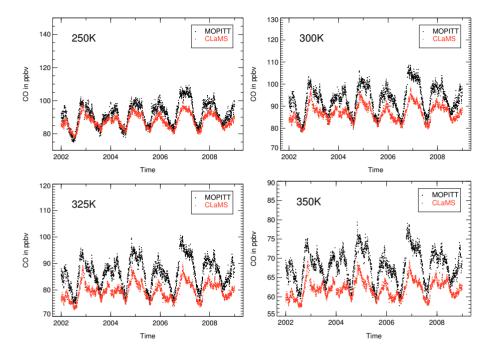
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**Fig. 2.** Comparison of tropical CLaMS CO simulations (red) with MOPITT measurements (black) from the beginning of 2002 to the end of 2008. For an appropriate comparison the CLaMS values are convoluted with the MOPITT averaging kernels and, like the MOPITT measurements, zonally averaged over the latitude band of  $\pm 15^{\circ}$ . The comparison is done for the CLaMS model levels  $\xi = 250\,\mathrm{K}$ , 300 K, 325 K, and 350 K corresponding to pressure levels of 500 hPa, 350 hPa, 250 hPa, and 150 hPa, respectively. The shift between the red and the black lines indicates the model bias.

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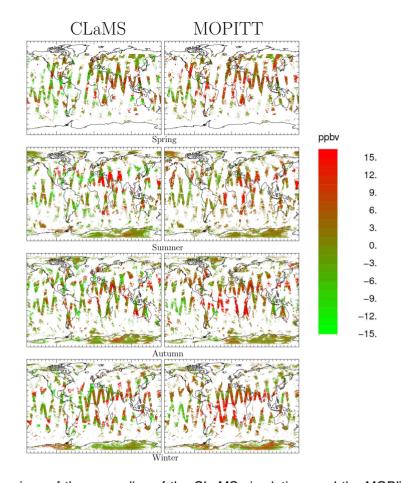
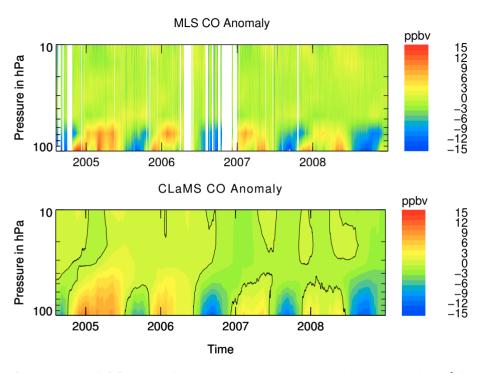


Fig. 3. Comparison of the anomalies of the CLaMS simulations and the MOPITT measurements of CO on the model surface  $\xi = 350 \,\mathrm{K}$ ; 150 hPa by showing the differences between the values and the respective zonal mean for the four seasons. For an appropriate comparison, the CLaMS values were convoluted with the MOPITT averaging kernels.



**Fig. 4.** Comparison of CO anomalies, zonally averaged in the latitude band  $\pm 15^{\circ}$  from MLS measurements (top panel) and the CLaMS simulation (bottom panel) for the time period 8 August 2004 to 29 December 2008. The CLaMS values were vertically smoothed with a top hat function (with a width of  $\sim 3$  km) to approximately take into account the field of view of the MLS instrument. The black lines indicates where the anomaly is zero.

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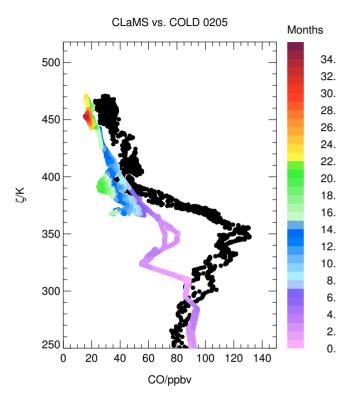




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**Fig. 5.** Comparison of the simulation of CO from CLaMS (coloured) with in-situ measurements from the COLD instrument (black) on the Geophysica on 5 February 2005. During this flight events of fast high-reaching convection were observed. The x-axis shows the volume mixing ratio of CO in ppbv, the y-axis reflects the height using the height coordinate in the model. The mean age of air calculated by the model is colour-coded and given in months.

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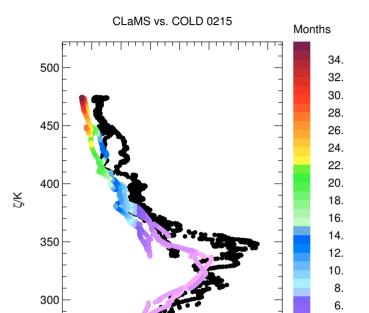
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**Fig. 6.** As Fig. 5, but for 15 February 2005. This flight was planned as a survey flight to measure TTL conditions at background conditions.

80

CO/ppbv

100 120 140

60

250

0

20

40

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