CLOUD RESOLVING STUDIES OF TRACE GAS SCAVENGING AND PHOTOCHEMISTRY

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1. Introduction

A cloud resolving online chemistry transport model was developed based on WRF and has been used to study the transport of trace gases, the scavenging of soluble trace gases, and influences of lightning produced nitrogen oxides (NO_x=NO+NO_2) on local photochemistry. This presentation mainly focuses on the influence of the retention coefficient (i.e. the fraction of a dissolved trace gas which is retained in hydrometeors during freezing) on the vertical re-distribution of soluble trace gases by deep convective clouds. A few previous model studies (Crutzen and Lawrence 2000; Mari et al. 2000; Barth et al. 2001; Yin et al. 2002) have suggested that even highly soluble trace gases can reach the upper troposphere if they are ejected from freezing hydrometeors at high altitudes. Here, results from cloud system resolving model runs (in which deep convection is initiated by small random perturbations and so-called ”large scale forcings” are prescribed) for a tropical oceanic (TOGA-COARE) and a mid-latitude continental case (ARM) are compared to runs in which a bubble was used to initiate deep convection (STERAO, ARM). Furthermore, the modeled ozone budget in the TOGA-COARE/CEPEX region (located in the tropical West Pacific) including influences of lightning NO_x is briefly discussed.

2. Model Description and Setup

A modified height coordinate prototype version of WRF (Skamaraock et al. 2001) is used in this study (see Salzmann et al. 2004; Salzmann 2005, for details). For soluble trace gases the uptake by, sedimentation together with, and mass transfer between different model categories of hydrometeors in the liquid or ice phase are parameterized (similar to Barth et al. 2001). Direct exchange between the ice phase and the gas phase is only considered for HNO_3 and H_2O_2, but not for the idealized tracers discussed in the next section. The tracer mass transfer between different hydrometeor categories is assumed to be proportional by mass to the hydrometeor mass transfer. The retention coefficient is assumed to be independent of the freezing process (e.g. dry or wet growth riming, homogeneous freezing) and is applied whenever tracer mass is transferred from the liquid to the ice phase. For the study of reactive trace gases, a CO-CH_4-HO_x-NO_x “background” chemistry and a simple lightning NO_x parameterization have been implemented. In Section 3, solely results from 3-D runs with periodic lateral boundary conditions are presented. The horizontal domain size used in these runs is 278x278 km^2. For TOGA COARE (19–26 December 1992) and ARM-A (26 June 1997 23:30 UTC – 30 June 1997 23:30 UTC) results from multi-day runs with prescribed “large scale forcings” are presented. In these runs very small (maximum 0.1 g kg^{-1}) water vapor perturbations are applied during the first 2.5 h of the simulations (prior to the onset of deep convection). The STERAO run is initialized with three positively buoyant thermals as in Barth et al. (2001). Furthermore, a run was initialized on 29 June 1997, 23:30 UTC with meteorological profiles from ARM-A in which a positively buoyant ($\Delta\theta$=5 K) thermal with radius r=20 km and height z_o=1800 m was used to initiate deep convection. In the multi-day runs in Section 3, the tracer fields are reset to their (horizontally homogeneous) initial values every 24 h after an initial offset of 12 h. In Section 4, results from 2-D runs with specified boundary conditions for trace gases (derived from a global chemistry transport model) are presented. The domain length used in these runs is 500 km and the mean ascent in the TOGA COARE region is taken into account for calculating the vertical tracer advection. The horizontal resolution is 2 km and the timestep is 5 s in all runs. The vertical resolution is 350 m in the TOGA COARE runs and variable grid spacings with increasing resolution towards the Earth’s surface are used in the ARM and the STERAO runs.

3. Retention Coefficient and Vertical Transport

Fig. 1 shows horizontally averaged mixing ratio profiles calculated for two different initial profiles (T1 and T2) for the TOGA COARE case. The tracers with initial profiles T1 are initially located in the lower troposphere, while the

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Table 1: Ratios α=$\overline{\mu}_s/\overline{\mu}_i$, where $\overline{\mu}_s$ and $\overline{\mu_i}$ are the average mixing ratios in the upper troposphere of highly soluble and insoluble tracers, respectively, for two different initial profiles (T1 and T2) for TOGA COARE (T.C.), ARM A, and STERAO.

	T.C. 2.5 h	T.C. 12 h	T.C. 24 h	ARM 2.5 h	ARM 12 h	ARM 24 h	STERAO	ARM BUB
α_r[1] T1	$2.5 \cdot 10^{-4}$	$2.1 \cdot 10^{-4}$	$1.2 \cdot 10^{-4}$	$9.0 \cdot 10^{-4}$	$1.2 \cdot 10^{-3}$	$1.0 \cdot 10^{-3}$	$1.8 \cdot 10^{-2}$	$1.3 \cdot 10^{-2}$
α_e T1	$1.4 \cdot 10^{-2}$	$1.0 \cdot 10^{-2}$	$9.7 \cdot 10^{-3}$	$4.1 \cdot 10^{-2}$	$6.6 \cdot 10^{-2}$	$8.4 \cdot 10^{-2}$	0.90	0.46
α_r T2	0.56	0.52	0.32	0.77	0.67	0.48	0.88	0.96
α_e T2	0.70	0.67	0.48	0.83	0.76	0.61	0.98	0.99

[1] Soluble tracers are either assumed to be completely retained (α_r) or completely rejected (α_e).

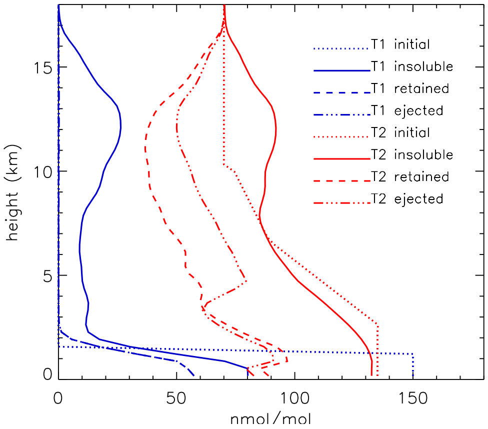

Figure 1: Initial tracer profiles and horizontally averaged mixing ratios for the TOGA COARE case 12 h after the beginning of each 24 h time slice (average for 6 time slices).

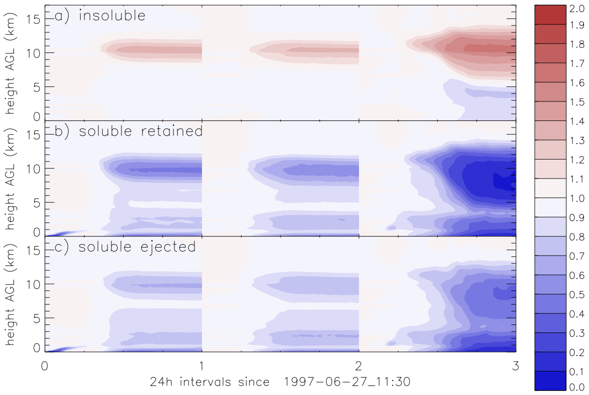

Figure 2: Time series of the ratio β of modelled averaged mixing ratio to initial mixing ratio for tracers with initial profile T2 during the ARM-A episode (except first and last 12 hours).

initial profile of T2 is a CO profile which has been used by Barth et al. (2001) in their pioneering cloud resolving model study of soluble tracer transport during STERAO. The tracers have been assumed to be either insoluble, highly soluble and completely retained during hydrometeor freezing, or highly soluble and completely ejected during hydrometeor freezing. While the insoluble tracers are efficiently transported to the upper troposphere, scavenging prevents efficient transport for the soluble tracers independent of the retention coefficient (compare also Fig. 2). Highly soluble tracers with initial profile T1 are efficiently scavenged already below the 0°C level. The strong sensitivity of tracers with initial profle T2 to the retention cofficient suggests that the retention coefficient plays a large role for the scavenging of highly soluble trace gases with a (chemical) source in the upper troposphere (e.g. H_2O_2).

Based on the ratios α=$\overline{\mu}_s/\overline{\mu}_i$ of soluble to insoluble tracer average mixing ratios in the upper troposphere after modelled deep convection, Barth et al. (2001) have suggested that global models such as the one used by Crutzen and Lawrence (2000) may underestimate the transport of highly soluble tracers to the upper troposphere. Crutzen and Lawrence (2000), however, investigated the transport of soluble tracers with a surface source (similar to T1), while the initial profile specified by Barth et al. (2001) is identical to T2. Based on Fig. 1 and Table 1, one can attribute the difference noted by Barth et al. (2001) to the use of different initial/boundary conditions in the two studies. Table 1 shows ratios α for the TOGA COARE and ARM multiday runs based on averages over the output times 2.5 h, 12 h, and 24 h after each re-initialization. Furthermore, the table shows the ratios at the end of the simulation (after 2.5 h) for the STERAO and the ARM A “bubble” (ARM BUB) case which were initialized with positively bouyant thermals as described in the previous section. Note that especially for T2 the ratios generally depend on domain size, since they depend on the ratio of cloudy area to cloud free area. For T1 α is small except for α_e in the STERAO and the ARM BUB case, i.e. if large scale forcings are applied together with small random perturbations, retained as well as rejected highly soluble tracers are not efficiently transported from the boundary layer, neither for the tropical oceanic case nor for the mid-latitude continental case, which is characterized by more vigorous deep convection. The relative difference between α_r and α_e in Table 1 is, however, large, indicating that the retention coefficient may be important for highly soluble gases for which even inefficient transport could play a role. Relatively large average upper tropospheric mix-

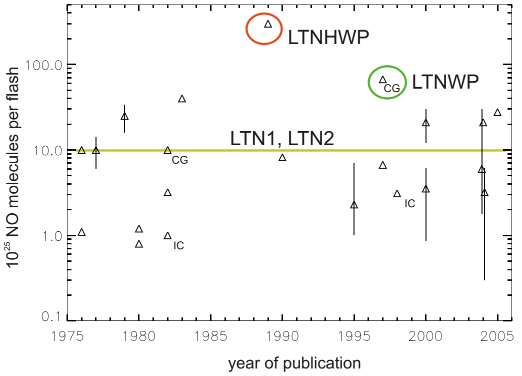

Figure 3: Literature values for the number of NO molecules produced per flash. For references see Salzmann (2005).

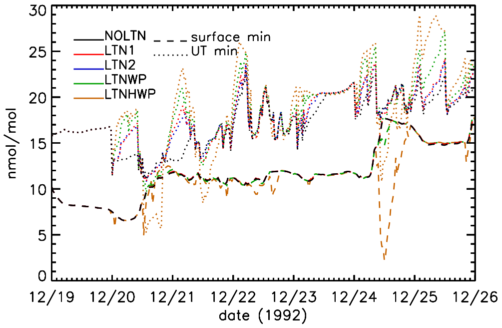

Figure 4: Time series of the minimum ozone mixing ratios in the lowest model layer (surface layer) and of the minimum ozone mixing ratios in the upper troposphere (UT) for various lightning NO_x sensitivity runs.

ing ratios of highly soluble non-retained tracers were only found for cases in which deep convection was initialized by bubbles (which is consistent with results from earlier studies using cloud resolving models). Given the apparent dependence of the results on the model setup, one could argue that in the future more studies with different approaches (especially with a more realistic initiation of deep convection, e.g. considering effects of orography) are needed. Although neither of the techniques for initiating deep convection used here is perfect, this study indicates that cloud resolving models using bubbles to initiate deep convection may possibly have over-estimated the influence of the retention coefficient on the vertical transport of highly soluble tracers.

4. The Ozone Budget in the TOGA-COARE/CEPEX Region and Influences of Lightning NO_x

Chemical loss of ozone in the marine boundary layer and upward transport of ozone poor air by deep convection have been suggested as a possible explanation for record low ozone mixing ratios in the upper troposphere observed during CEPEX (e.g. Kley et al. 1996). However, comprehensive global and cloud resolving models have failed to reproduce these extremely low ozone mixing ratios in various studies. An exception is the study by Wang and Prinn (2000). Based on results from a sophisticated cloud resolving model including chemistry, Wang and Prinn suggested the production of NO_x by lightning and the subsequent ozone titration as a possible cause for the near zero ozone mixing ratios. Their simulations did not show the vertical transport of ozone poor air to the upper troposphere to significantly decrease ozone concentrations in the upper troposphere. Instead, they found the downwards transport of ozone rich air induced by deep convection to lead to increases of the average ozone mixing ratios in the upper troposphere.

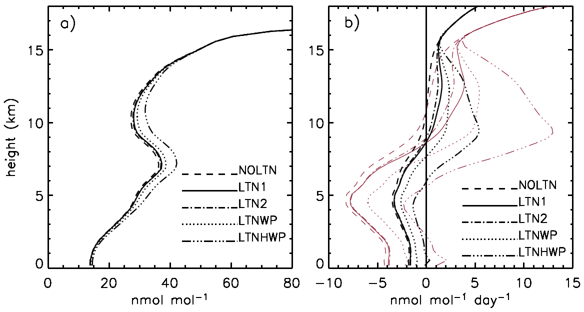

Figure 5: (a) Domain and time averaged ozone volume mixing ratios for solar zenith angle SZA$<$70° (b) Net chemistry tendency: diurnal averages (black) and averages for SZA$<$70°.

Here, results from multi-day sensitivity runs with specified boundary conditions for trace gases and various assumptions regarding flash rates and NO_x production per flash are presented (see Salzmann 2005, for a detailed description of the runs). In the LTNWP and the LTNHWP runs, the same numbers of molecules per flash have been adopted as in Wang and Prinn (2000), i.e. $300{\cdot}10^{25}$, and $67{\cdot}10^{25}$ molecules per cloud to ground (CG) flash, respectively. The numbers of CG flashes per day per km domain length in the present study, are, however, about a factor three lower than in the study of Wang and Prinn. In the LTN1 and the LTN2 run, a significantly lower NO production per CG flash has been adopted, which is closer to typical values from the literature ($10{\cdot}10^{25}$ molecules, see Fig. 3). Reductions of ozone mixing ratios which could explain the observed near zero ozone mixing ratios (in agreement with Plate 6 of Wang and Prinn 2000) have only been calculated for the LTNHWP run (Fig. 4). The number of molecules per flash used in the LTHNWP run has, however, been judged to be unrealistically high in a number of more recent studies. The domain averaged ozone mixing ratios are found to increase with increasing lightning NO_x production in the present study (Fig. 5). Wang and Prinn

(2000), on the other hand, found the average ozone mixing ratios to decrease with increased lightning NO_x production based on a sensitivity run for a single storm in which they calculated higher flash rates and assumed the extremely high NO production of $300{\cdot}10^{25}$ molecules per flash. Small scale downwards transport of ozone in thin filaments is simulated in association with with the rear inflow into deep convective systems (see snapshot in Fig. 6). Upwards transport of O_3-poor air from the lower troposphere, however, on average dominates in the upper troposphere, where vertical advection is a negative contribution to the model calculated ozone budget (Fig 7). This contrasts the result of Wang and Prinn, who in their simulations found downwards transport to cause increased ozone mixing ratios in the upper troposphere. Wang and Prinn, however, prescribed a relatively "flat" ozone profile (i.e. a profile with a small vertical gradient in the troposphere) as their initial condition. We argue that near zero ozone values observed during CEPEX were not caused by lightning NO_x production since significant local decreases in ozone are only computed when an unrealistically high number of NO_x molecules produced per flash is prescribed. Instead, we suggest that the ozone profiles observed during CEPEX may have been strognly influenced by the deep convection and the larger scale flow associated with the intraseasonal oscillation. Due to constraints in computing resources, domain sizes of cloud resolving models have been too small in order to simulate the chemical loss of ozone in the marine boundary layer over larger areas and, consequently, longer times. As faster computers become increasingly available, 3-D cloud resolving model studies with larger model domains will certainly help us to better understand the processes influencing the atmospheric composition in and downwind of regions with frequent deep convection.

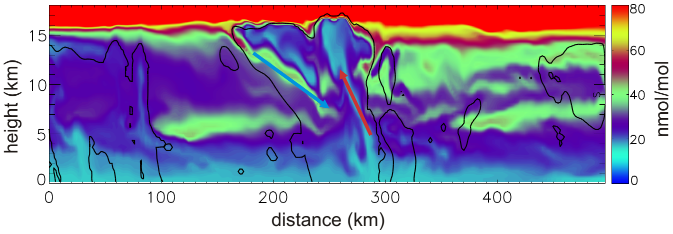

Figure 6: Ozone volume mixing ratios (shaded) and total hydrometeor mixing ratio contour q_t=0.01 g kg^{-1}. Arrows indicate ozone poor air transported upwards and ozone rich air transported downwards.

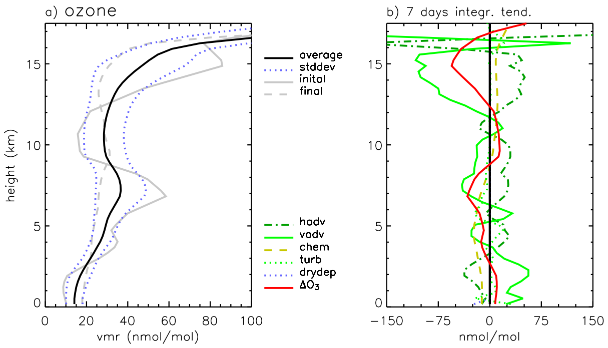

Figure 7: (a) Time and domain averaged O_3 mixing ratios ± standard deviation, initial and final O_3 mixing ratio profiles. (b) Ozone budget (time integrated and domain averaged tendencies divided by the average air density) for horizontal and vertical advection (hadv, vadv), chemistry (chem), turbulent diffusion (turb), and dry deposition (drydep), and the difference between the initial and the final profile (ΔO_3).

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