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# Chemistry-transport modeling of the satellite observed distribution of tropical tropospheric ozone

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## Abstract

We have compared the 14-year record of satellite derived tropical tropospheric ozone columns (TTOC) from the NIMBUS-7 Total Ozone Mapping Spectrometer (TOMS) to TTOC calculated by a chemistry-transport model (CTM). An objective measure of error, based on the zonal distribution of TTOC in the tropics, is applied to perform this comparison systematically. In addition, the sensitivity of the model to several key processes in the tropics is quantified to select directions for future improvements. The comparison indicates a widespread, systematic ( $\sim 20\%$ ) underestimate of TTOC over the tropical Atlantic Ocean, which maximizes during austral spring. This 'Atlantic mismatch' is largely due to a misrepresentation of seasonally recurring processes in the model, while minor differences between model and observations over the tropical Pacific Ocean are mostly due to uncaptured interannual variability. Although chemical processes determine the TTOC extent, dynamical processes dominate the TTOC distribution, as the use of actual meteorology pertaining to the year of observations always leads to a better agreement with TTOC observations than using a random year or a climatology. The modeled TTOC is remarkably insensitive to many model parameters due to efficient feedbacks in the ozone budget. Nevertheless, the simulations would profit from an improved biomass burning calendar, as well as from an increase in  $\text{NO}_x$  abundances in free tropospheric biomass burning plumes. The use of multi-year satellite derived tropospheric data to systematically test and improve a CTM is a promising new addition to existing methods of model validation, and is a first step to integrate tropospheric satellite observations into global ozone modeling studies.

## 1. Introduction

The availability of trace gas measurements in the tropics has increased considerably by a large number of intensive campaigns (LBA, PEM, TRACE, INDOEX, SAFARI) and the expansion of the number of measurement stations in the tropics. The interest in

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the tropics is based on the active photochemistry, rapid convective mixing along with lightning discharge, high humidity, widespread biomass burning, and growing industrial emissions. All these processes influence the concentration of oxidants such as ozone and the hydroxyl radical. The latter plays a crucial role in the removal of biogenic emissions and anthropogenic pollution from the atmosphere.

Observations from satellites play an increasingly important role in monitoring of the chemical composition of the troposphere. The Total Ozone Mapping Spectrometer (TOMS) instrument aboard the NIMBUS-7 and Earth Probe (EP) Satellites (McPeters and Labow, 1996) has been used to retrieve tropospheric column ozone (TTOC) (Fishman and Larsen, 1987; Kim et al., 1996; Ziemke et al., 1998; Hudson and Thompson, 1998), while the GOME instrument aboard ERS-2 has been used to retrieve columns of tropospheric NO<sub>2</sub>, SO<sub>2</sub> (Eisinger and Burrows, 1998; Burrows et al., 1999), and BrO (Richter et al., 1998). In the near future, SCIAMACHY (Burrows and Chance, 1992) will expand this suite of trace gases with N<sub>2</sub>O, CO<sub>2</sub>, CO and CH<sub>4</sub>. These last two trace gases are also the focal point of the MOPITT instrument (Pan et al., 1998) aboard the EOS-TERRA satellite, launched in 2000. The most important advantages of satellite observations are the global coverage and the continuous monitoring on time scales from days to years, and sometimes even decades. For historically data sparse regions such as the tropics, this will contribute significantly to our understanding of atmospheric processes and help detect changes.

In addition, satellite measurements can easily be combined with chemistry-transport models (CTM) and global circulation models (GCM), since spatial and temporal scales are often comparable. There are several ways in which these properties can be exploited in a quantitative way. This includes for instance assimilation of satellite measurements into models to constrain chemistry calculations (e.g. Jeuken et al., 1999) and provide boundary conditions. Also, satellite measurements can be used as additional constraints in inversion problems, where the global distribution of a tracer is used to infer the distribution and magnitude of its sources and sinks (e.g. Houweling et al., 1999). Vice versa, model calculations can be used in satellite retrieval algorithms to

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provide a ‘first guess’ field or to provide missing parameters needed in the retrieval (e.g. [Chance et al., 2000](#)). The growing amount of satellite observations and expansion of remotely sensed trace species will intensify the use of these observations in modeling studies.

5 From the several tropospheric satellite observations available at the moment, the TTOC record derived from the NIMBUS-7 observations is the most extensive. It spans the complete NIMBUS-7 observational period (1979–1992) on a high spatial ( $2 \times 1$  degrees) and temporal (twice monthly) resolution. This has enabled analysis of seasonal cycles ([Martin et al., 2000](#)), trends ([Thompson and Hudson, 1999](#)) and inter-annual variability ([Ziemke et al., 1999](#)) in tropical ozone. Moreover, this dataset has shown to be very useful in studying both local scale features such as the effect of El Niño-Southern Oscillation (ENSO) related fires over Indonesia ([Chandra et al., 1998](#); [Thompson et al., 2001](#)), as well as large scale phenomena such as the evolution of the South Atlantic ozone maximum ([Thompson and Hudson, 1999](#)) and the changing Walker circulation during ENSO ([Ziemke et al., 1999](#)). Nevertheless, most modeling studies have only used the TTOC data for a qualitative comparison, while ozone sondes ([Logan, 1999](#)) were used for a quantitative comparison. Here, we explore how satellite derived TTOC can be used to validate and improve our CTM.

15 Previously, we have tested our model against in situ data from sondes and ground-based stations ([Lelieveld and Dentener, 2000](#); [Marufu et al., 2000](#); [Peters et al., 2001](#)). Comparison to the climatology of [Logan \(1999\)](#) revealed an apparent problem in the representation of the zonal ozone distribution in the tropics, which shows a maximum of TTOC over the Atlantic Ocean and a minimum over the Pacific Ocean. This distribution is often called the ‘zonal wave one pattern’ ([Kim et al., 1996](#)), and is a typical example of a large scale feature in tropical ozone that can be well studied with a combination of global models and satellite observations. Therefore, we present a quantitative comparison with TTOC derived with the Modified Residual (MR) algorithm of [Hudson and Thompson \(1998\)](#). We have developed a measure of error to consistently perform this comparison. This measure of error allows us to recognize systematic shortcom-

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ings in our model on seasonal and interannual time scales, and separate the influence of meteorology and photochemistry on our model results. Moreover, it helps to identify processes that should be considered for improvement and allows us to quantify the sensitivity of our model results to several processes such as biomass burning and lightning NO<sub>x</sub> emissions. It is important to stress that these techniques are not used to ‘tune’ the model to observations. The processes that are most likely to cause the observed differences should be carefully studied in future work, preferably in a three dimensional perspective using ozone sondes. Our work presented here should be seen as an exploration of model sensitivities providing guidance to actual improvements.

In Sect. 2, we introduce the CTM that was used in this study, and we discuss the chemistry scheme. The TTOC observations used are the subject of Sect. 3. We introduce a measure of error to compare model calculations of TTOC with satellite observations in Sect. 4. In Sects. 5, 6 and 7 we present our modeling results, focused on the role of seasonal and interannual variability, meteorology and photochemistry, respectively. In Sect. 8 we explore the models sensitivity to the treatment of biomass burning emissions and lightning NO<sub>x</sub> emissions, which is illustrated with several ozone sondes (Sect. 9). Finally, we discuss our results in Sect. 10, and give a short summary in Sect. 11.

## 2. Model description

We use the chemical tracer transport model version 3 (TM3) to simulate transport and photochemistry of 48 trace gases within a global domain. The TM3 model has a horizontal resolution of 5° × 3.75° (lon × lat) and uses 19 layers with a hybrid of sigma-pressure coordinates in the vertical, with the model top at 10 hPa. Approximately five layers constitute the boundary layer, nine the free troposphere, and five the lower and middle stratosphere. Transport in the off-line model is prescribed using ECMWF data (Gibson et al., 1997). The model uses six-hourly, 3-dimensional fields of geopotential height, horizontal mass-fluxes, temperature, specific humidity and cloud information

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(liquid water and ice water content), as well as 2D information on cloud cover and precipitation (large-scale and convective). The subgrid scale transport by turbulence and convection is calculated from archived diffusion and entrainment/detrainment coefficients from the ECMWF model using a modified scheme by [Louis \(1979\)](#) and [Tiedtke \(1989\)](#), respectively. All the input fields are extrapolated from the ECMWF archived resolution to the TM3 model resolution. The results presented in Sect. 5 and 6 were obtained using the multi-year (1979–1993) simulation by [Lelieveld and Dentener \(2000\)](#), which was also used in [Peters et al. \(2001\)](#). The configuration of the TM3 model in the sensitivity studies (Sect. 8) is identical, and pertain to the year 1992. However, the model currently runs on a new SGI-3800 massive parallel system from the Dutch National Supercomputing Facility (NCF). A short list of processes in the model and references to more elaborate discussion of these processes is given in Table 1. We will limit ourselves to a description of the processes explicitly relevant for this study, being the emissions from biomass burning, the photochemistry of ozone and its precursors, and the lightning  $\text{NO}_x$  parameterization.

## 2.1. Biomass burning emissions

The emissions of biomass burning in the model were originally constructed from two datasets. The EDGAR emission database ([Olivier et al., 1996](#)) prescribes the total amount of CO, HCHO,  $\text{NO}_x$ , and NMHC emitted yearly. This database is based on estimates for the year 1990 that were obtained from various sources on a resolution of  $1^\circ \times 1^\circ$ . For the temporal distribution of these emissions, the model relies on the [Hao and Liu \(1994\)](#) fire calendar, which contains monthly fractions of annual amounts of biomass burned on a  $5^\circ \times 5^\circ$  resolution. For the sensitivity studies presented in Sect. 8, we have replaced the fire calendar of [Hao and Liu \(1994\)](#) with one based on fire counts from the AVHRR satellite ([Flasse and Ceccato, 1996](#)), which contains 10-day composites of global fire activity for 1992–1993. The observations were used to construct a time distribution function by scaling the monthly total of counts by the yearly total of counts, yielding a fraction of fires occurring each month. Thus, the yearly totals

remain the same, but the emissions maximize in the month with the largest number of fires.

It is important to note that this approach is meant to provide a simple alternative to the use of the [Hao and Liu \(1994\)](#) database, suitable for a model sensitivity analysis.

In our algorithm, we do not account for the size of the fires that were observed; large fires and small fires contribute equally to our calendar, while in reality the amount of trace gases released to the atmosphere can differ strongly. Our algorithm will therefore overestimate the influence of these small fires and, since the total emissions are constant, underestimate the influence of large fires. Also, our algorithm does not include the smoldering phase of the fires, which can continue for several weeks especially after forest fires (M.O. Andreae and P. Merlet, Emission of trace gases and aerosols from biomass burning, Global Biogeochemical Cycles, accepted, 2000). Due to the neglect of the smoldering phase, the timing of smoldering-related emissions (such as CO, NH<sub>3</sub>) is biased towards the moment of detection of the fire, i.e. the flaming phase. Nevertheless, our alternative fire calendar suffices for a sensitivity analysis, whereas real satellite derived emission estimates require a more sophisticated approach ([Pinty et al., 2000](#)) (B. Duncan, R. Martin, A. Staudt, R. Yevich and J. Logan: Interannual and Seasonal Variability of Biomass Burning Emissions Constrained by Remote-Sensed Observations, manuscript in preparation, 2001).

## 2.2. Photochemistry of ozone

The photochemistry scheme in the TM3 model is based on the CBM-4 mechanism of [Gery et al. \(1989\)](#). It includes 38 trace gases and 110 gas phase reactions. The CBM-4 mechanism was initially intended to simulate ozone photochemistry over urban areas with high NO<sub>x</sub> abundances. Therefore, [Houweling et al. \(1998\)](#) modified the scheme such that low NO<sub>x</sub> conditions are also well captured. This has improved ozone photochemistry over more remote areas, where NO<sub>x</sub> concentrations are much lower. The CBM-4 mechanism is based on the family concept, where trace gases of common origin with similar lifetimes and reactive properties are lumped into a family, which has

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the common characteristics of all the species it is meant to represent. This concept and the consequences for the photochemistry are described in more detail in the work of [Gery et al. \(1989\)](#), [Duncan and Chameides \(1998\)](#), [Houweling et al. \(1998\)](#) and [Pöschl et al. \(2000\)](#).

5 Precursors for ozone production that are emitted from biomass burning include  $\text{NO}_x$ , CO, formaldehyde and acetaldehyde. Other NMHC compounds that are known to originate from biomass burning (e.g. acetone, acetonitrile, ethene) are represented through the emissions of several lumped species (parafinic and olefinic NMHC's, methylglyoxal). The scheme includes the photochemistry of peroxy-acetyl nitrate (PAN), which  
10 is an important secondary product from the oxidation of acetaldehyde and contributes significantly to the free tropospheric nitrogen budget ([Singh et al., 1995](#)). Dry deposition is an important sink for  $\text{NO}_x$ ,  $\text{HNO}_3$ , PAN, formaldehyde and acetaldehyde, while wet deposition through scavenging by clouds removes predominantly  $\text{HNO}_3$ , formaldehyde and  $\text{H}_2\text{O}_2$ . The chemical mechanism includes the heterogeneous conversion of  
15  $\text{NO}_x$  to  $\text{HNO}_3$  on sulfate aerosols according to [Dentener and Crutzen \(1993\)](#), as well as a description of sulfur chemistry ([Jeuken, 2000](#)).

### 2.3. Lightning $\text{NO}_x$ emissions

The lightning  $\text{NO}_x$  emissions in the model are based on the widely used parameterization of [Price and Rind \(1992\)](#). The spatial distribution of lightning, as well as the  
20 intensity, is calculated using the heights of cloud tops in systems of deep convection. To discriminate between the efficiency of lightning  $\text{NO}_x$  production over the oceans relative to land, we have assumed a ratio of 1:10 in production efficiency, following [Levy et al. \(1996\)](#). The lightning  $\text{NO}_x$  production efficiency in cloud-to-ground discharges has been scaled to account for the higher efficiency in high latitudes ([Mackerras and Darveniza, 1994](#)). The  $\text{NO}_x$  emissions are uniformly distributed between cloud base and  
25 cloud top, proportional to air-density. By scaling the lightning  $\text{NO}_x$  emissions with meteorological parameters, interannual variability in the model meteorology is also reflected in interannual variability in the lightning  $\text{NO}_x$  emissions. The total annual simulated

lightning NO<sub>x</sub> emissions over the period 1979–1993 are  $4.6 \pm 0.3 \text{ TgN yr}^{-1}$ .

In our sensitivity experiments, we have used the vertical lightning NO<sub>x</sub> distribution profiles as given in [Pickering et al. \(1998\)](#). In this work, the vertical distribution depends on the location (tropics or extra-tropics), and surface characteristics (land or sea). Also, we have implemented a lightning NO<sub>x</sub> parameterization developed by [Meijer et al. \(2001\)](#). This follows the principles of [Price and Rind \(1992\)](#), however, the occurrence and intensity of lightning NO<sub>x</sub> emissions is coupled to the convective precipitation in the model. This was shown to correlate well with space-based observations of lightning flash rates over Europe, and was tested against NO<sub>x</sub> observations in the extra-tropics. The main difference between the two approaches is the fraction of the emissions occurring in the tropics. In the new parameterization, this fraction decreases from 80% to 70%, which agrees better with independent estimates ([Levy et al., 1996](#); [Price and Rind, 1992](#)).

#### 2.4. Labeled tracers

To attribute photochemically produced ozone to the different processes in our model, we applied a tracer labeling technique. The assumption that ozone formation in the troposphere is NO<sub>x</sub>-controlled ([Crutzen, 1988](#)) implies that NO-to-NO<sub>2</sub> conversion by peroxy-radicals can be used to diagnose ozone formation. Thereto, we have introduced labeled tracers of NO<sub>x</sub>, the most important NO<sub>y</sub> species, and ozone. The NO<sub>x</sub> tracers are labeled according to their source category, representing emissions from lightning, biomass burning, nonagricultural soils, and industrial sources. When a labeled NO molecule is converted to NO<sub>2</sub> by a peroxy radical, the subsequent photolysis of NO<sub>2</sub> yields an ozone molecule labeled accordingly. The major reaction pathways associated with NO<sub>x</sub>/NO<sub>y</sub> conversions are labeled as well, to account for recycling of NO<sub>x</sub> from its reservoir species. Furthermore, tropospheric ozone introduced through stratosphere-troposphere exchange (STE) is labeled. The labeled tracers participate in all other processes open to their regular, non-labeled counterparts. This approach was tested

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and discussed in [Lelieveld and Dentener \(2000\)](#).

### 3. Observations

We use the NIMBUS-7 TTOC record (1979–1992) to detect systematic flaws in our model and to gain more insight in the sensitivity of our model calculated ozone. The observations were derived from the TOMS total ozone observations with the Modified-Residual (MR) method ([Hudson and Thompson, 1998](#)). The TTOC record provides twice monthly fields of TTOC, covering all the tropical latitudes where a zonal wave one pattern in total column ozone is observed. The horizontal resolution of the dataset is  $2^\circ$  longitude  $\times$   $1^\circ$  latitude ( $230 \times 115$  km at the equator), and through averaging over several orbital passes of the satellite, a picture of the full zonal distribution of TTOC is obtained in several days. Through averaging the observations over 15-day periods, the noise in the dataset is strongly reduced. The data is available on the internet through <http://metosrv2.umd.edu/~tropo/>.

The TTOC record has been validated extensively against available ozone sondes in the tropics, and generally a high correlation is obtained ([Thompson and Hudson, 1999](#)). This validation could only be performed for ozone sonde stations that were situated in the Southern Hemisphere tropics, since the only station in the Northern Hemisphere tropics (Paramaribo, Suriname, 55W, 5N) was not in operation before September 1999. In the MR retrieval algorithm of the TTOC, sonde observations from selected stations in the SH are used to obtain a-priori information on the zonal distribution of ozone, which is thereafter projected onto each grid point of the total column ozone observations to yield the TTOC.

The most important uncertainties in the observations of TTOC are related to the assumed tropopause height as well as the insensitivity of the TOMS instrument to tropospheric ozone near the surface. The instrument is progressively insensitive to the signal of ozone originating from altitudes below the stratospheric ozone maximum. The algorithms that were developed to circumvent these problems are explained in detail in

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Hudson and Thompson (1998). The one sigma standard deviation of the TTOC product is reported as 5.3 DU, and is mainly due to the averaging of the ozone sondes over two weeks, which are used to determine the tropospheric background ozone abundance in the MR algorithm. The standard deviation is expected to increase with distance from these stations, and is probably larger in the NH than in the SH, since the underlying zonal wave one pattern used in the retrieval was based on SH ozone sondes. However, since there is no objective way to define the standard deviations at other locations, we will assume that  $\sigma = 5.3$  DU over the whole domain.

Alternatively, ozone sondes and in situ measurements from campaigns such as TRACE-A, PEM, NOXAR, or MOZAIC could be used to test our models performance in the tropics. The data acquired in these campaigns generally include detailed vertical information on several trace species and meteorological parameters. However, there is an important trade off to consider that can be seen as a choice between the amount of detail of information and the areal and temporal coverage. The individual campaigns usually cover periods of several weeks in a specific year, and focus on a limited geographical region. Especially near the surface, they are often influenced by local pollution or particular small scale meteorological conditions (e.g. sea-breeze, orographic flows). This is especially disadvantageous when comparing to global models that generally apply coarse resolutions and parameterizations not designed to capture these small scales. Ozone sondes and in situ measurements are much better suited to study smaller temporal and spatial scales, or the effect of a single process (e.g. biomass burning or convection). These data would therefore form an ideal basis to improve possible model shortcomings, which is the next step in our research. However, in this work we take advantage of the global, multi-year TTOC record to firstly isolate the regions, seasons, and processes that require improvement.

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#### 4. Measure of error

Figure 1 (top) shows the observed TTOC distribution in September 1992, when biomass burning emissions were particularly strong. The South Atlantic ozone maximum clearly stands out. The TTOC maximum exceeds 50 DU, gradually decreasing with distance from the Atlantic, reaching minimum values between 10 and 20 DU over the Pacific Ocean. The comparison with model simulation of TTOC for the same year and month in Fig. 1 (bottom) shows distinct differences. The absolute values of the TTOC often differ by more than the one sigma (5.3 DU) standard deviation of the measurements, and many smaller scale features are not reproduced. The discrepancies are largest over the Atlantic Ocean, where a systematic mismatch is seen between observations and model calculations. In contrast, the model calculations reproduce the observations quite well over the Pacific Ocean. This is consistent with earlier results (Peters et al., 2001), based on a comparison with ozone sondes from Samoa, Ascension and Natal.

Beside this 'Atlantic mismatch', there are two other important features that appear from Fig. 1. Firstly, the amount of detail in the observations is larger. This can be partly attributed to the higher resolution of the observations, but it is also caused by a lack of variation in the monthly emission data. To account for the different resolutions of the datasets, we interpolated the model results to the  $2^\circ \times 1^\circ$  degree resolution of the TTOC observations and we applied a zonal running average to the TTOC observations, with a window size equal to the zonal model grid-size (5° degrees). This filters out strong local features in the observations at resolutions smaller than the model grid, which cannot be reproduced by the model.

Secondly, it is important to note that variations in the meridional direction are small compared to the large zonal variation. Since the area depicted in Fig. 1 encompasses only 5–6 model grid boxes in the meridional direction (appr. 15° degrees), the model can not reproduce fine scale structures in the meridional direction. Therefore, the next step in our comparison is to take the meridional average of the respective datasets at

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each longitude, which yields the zonal distribution of TTOC.

#### 4.1. Zonal TTOC distribution

Figure 2 shows the derived zonal TTOC distribution in September 1992 for both datasets. The latitudinal extent of 15 degrees is a typical value for the MR TTOC product. The figure clearly shows the zonal extent of the Atlantic mismatch, characterized by model calculated TTOC values that are consistently lower than the observations over the 90 W to 90 E region. Although this mismatch maximizes each year in September, other months show a similar problem. The TTOC distribution depicted in Fig. 2 is chosen as the focal point of our comparison. Conveniently, the TTOC retrieval algorithm uses the meridional extent of the zonal wave one pattern to distinguish tropical air masses from extra-tropical air masses. Thus, this pattern is always present in the observations, and points that do not qualify as ‘tropical’ in the retrieval algorithm are not included in the comparison to our model calculations.

To make an objective comparison between the observed and modeled zonal TTOC distributions, a mathematical measure of error is introduced, which consists of two components. First, we define  $\epsilon_1$ , which is simply the correlation ( $r$ ) between the two lines in Fig. 2. The parameter  $\epsilon_1$  indicates how well the horizontal gradients in the zonal TTOC distribution are reproduced, and how well transitions in TTOC induced by the land-sea distribution for instance are captured. The shape of this pattern is influenced amongst other factors by the location of the Inter Tropical Convergence Zone (ITCZ), the presence of biomass burning plumes, or the occurrence of large scale convection and associated lightning. Increasing values of  $\epsilon_1$  denote a better representation of these zonal gradients. It is important to note that  $\epsilon_1$  is not influenced by the absolute value of TTOC, and its value will increase only when the zonal gradients in the model calculations better fit the observations. This is advantageous because it excludes the errors in the measurements and model that are introduced through uncertainties in the tropopause height. On the other hand, the parameter  $\epsilon_1$  cannot be used to assess the absolute fit between the observations and the calculations.

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Therefore, we also introduce a second parameter,  $\epsilon_2$ , that is defined as the root mean squared (RMS) deviation between observations and measurements, scaled by the standard deviation of the observations:

$$\epsilon_2(t) = \frac{1}{N_{lon}} \sum_{lon} \sqrt{\frac{(\overline{TOMS(t)} - \overline{TM3(t)})^2}{\sigma_{TOMS}^2}} \quad (1)$$

5 Where  $\overline{TOMS(t)}$  is the observed zonal distribution of TTOC for each month ( $t$ ), and  $\overline{TM3(t)}$  is the model calculated zonal distribution of TTOC for each month. The value of  $\epsilon_2$  will decrease as the discrepancy between observations and calculations decreases.

The number of longitudes used in the summation can vary depending on the scale we want to look at. For instance, in the analysis of the variability (Sect. 5), we analyze  
 10 14-year time series of  $\epsilon_2$  at each location ( $N_{lon} = 1$ ,  $t = 168$ ), while for the analysis of our sensitivity runs (Sect. 8) we subdivide the zonal domain into only two regions; a Pacific region ( $N_{lon} = 90$ ,  $t = 168$ ) and an Atlantic region ( $N_{lon} = 90$ ,  $t = 168$ ). Together,  $\epsilon_1$  and  $\epsilon_2$  enable us to quickly and objectively quantify the agreement with the TTOC observations, and intercompare the results of different model simulations.

## 15 5. Comparison of TTOC

As a first step, we calculate the values of  $\epsilon_2$  for each longitude and each month ( $t = 168$ ) of our multi-year simulation. The result is shown in Fig. 3, where a negative sign indicates that the model calculated values are higher than the observations. It shows that the values of  $\epsilon_2$  range from 0–2  $\sigma$  (appr. 0–10 DU) over the tropical Pacific,  
 20 to more than 3  $\sigma$  (appr. 16 DU) over the tropical Atlantic. The largest underestimates occur during August through November. The model calculated values of TTOC are generally lower than the observations. The only time the model slightly overpredicts the TTOC values is in December–January–February over the Pacific Ocean. The consistent underestimate of TTOC could indicate a model shortcoming, but could also

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indicate problems with the TTOC data. We will discuss this further in Sect. 10.

It is clear from Fig. 3 that the Atlantic mismatch is the most serious problem in our comparison, and that it is an annually recurring feature. In contrast, high values of  $\epsilon_2$  over the Pacific Ocean appear for several years, but not in all. To separate these systematic differences from the ones that occur occasionally, we have decomposed each time series of  $\epsilon_2$  into a seasonal cycle, and a component of interannual variability. The seasonal cycle of  $\epsilon_2$  is shown in Fig. 4. The figure confirms that most of the Atlantic mismatch is caused by seasonal differences. On average, the seasonal cycle contains 75% of the variance of the  $\epsilon_2$  time series over this region. In contrast, the seasonal cycle of  $\epsilon_2$  over the Pacific Ocean explains less than 50% of the original variance. From this, we can conclude that the most important model improvements should focus on the seasonality of TTOC over the Atlantic Ocean. Possibly, the Pacific region would profit from such improvements as well, since the two regions are coupled through transport.

## 6. Influence of transport

The coupling between the regions through transport is not the same in each year due to the occurrence of anomalous meteorological conditions such as those induced by ENSO. Through the use of ECMWF wind fields for each year, we have included this transport variability, and Peters et al. (2001) showed that the effect of ENSO on TTOC is present in our multi-year simulation. In this Section, we will apply our measure of error to quantify the influence of transport on our results.

We compare each year of TTOC observations with each year of our multi-year simulation. In our multi-year simulation, we have 15 years of calculated TTOC, which differ in meteorology (including lightning), stratospheric upper boundary conditions for ozone, and the anthropogenic emissions which include trends over the 1979–1993 period. In this section, we use yearly averaged values for 1979–1992 of the monthly calculated  $\epsilon_1$  to assess the performance of our model. Since  $\epsilon_1$  is independent of the absolute values of TTOC, the influence of increasing emissions in the multi-year simulation is

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largely excluded in this comparison. Thus, any year-to-year difference in  $\epsilon_1$  is almost entirely due to changes in meteorology.

For each year of the TOMS observations (1979–1992,  $n = 14$ ), we have calculated 14 values of  $\epsilon_1$ ; one value for each year of simulations (1979–1992,  $m = 14$ ). These individual values of  $\epsilon_1$  denote the success in simulating the observed zonal distribution of TTOC, given the specific meteorological conditions of each of the 14 simulated years. The average  $\epsilon_1$  of these individual years would represent the value when a random year of meteorology is applied, while there is one value of  $\epsilon_1$  that corresponds to the correct meteorology matching the year of TTOC observations.

Figure 5 shows the multi-year average value (open diamonds) and range of interannual variation (bars) for each of the 14 years TTOC observations, as well as the values of  $\epsilon_1$  for the model year corresponding to the year of observations (filled diamonds). The simulations performed with actual meteorology consistently perform better than average. In fact, the simulation with actual meteorology generally ranks higher than any other year. This is due to the fact that meteorological conditions determine to a large extent the location of local ozone maxima, as well as the zonal gradients in TTOC. Although the differences between a correlation of 0.7 and 0.84 might seem small, the fact that these differences show up in a yearly and meridionally averaged TTOC distribution indicates that the individual differences are in fact quite large.

From the figure one can also see that the 14-year average value of  $\epsilon_1$  varies from year-to-year, as does the range. This shows that some years of observations are more difficult to model than others presumably because their specific conditions are more difficult to capture. This is obvious for 1982/83, when El Niño conditions prevailed. While the simulation with actual meteorology performs well, some other years show a poor correlation. This indicates that in such a year, the zonal TTOC distribution can not be modeled very well if one does not consider the specific meteorological conditions during ENSO. The fact that the maximum of  $\epsilon_1$  for 1983 is the lowest in the whole series shows that other processes than transport that change during ENSO also influence the shape of the zonal TTOC distribution. Such processes include the

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occurrence of wildfires following anomalous droughts. Although this was shown to have a large impact on TTOC (Chandra et al., 1998; Thompson et al., 2001), it was not included in our multi-year model simulations. Adding this variability in emissions to the model is a difficult challenge, for which tools are currently being developed [B. Duncan, R. Martin, A. Staudt, R. Yevich and J. Logan: Interannual and Seasonal Variability of Biomass Burning Emissions Constrained by Remote-Sensed Observations, manuscript in preparation, 2001].

## 7. Role of photochemistry

Beside the previously mentioned wildfires, other sources contribute to the photochemical production of ozone in the troposphere, and thus partly determine the zonal distribution of TTOC as depicted in Fig. 2. By using the labeled tracers (see Sect. 2.4), we have calculated the contribution of several tropospheric ozone sources to the modeled TTOC distribution. The processes that were thus separated are photochemical ozone production from lightning, industrial, biomass burning, and soil NO<sub>x</sub> emissions, as well as the contribution from the stratospheric influx of ozone. Figure 6 shows the contribution of these processes to the modeled zonal TTOC distribution in September 1992.

Ozone production from industrial NO<sub>x</sub> emissions contributes several DU to the modeled ozone concentration. There is only very little difference in zonal direction, because the most important source is located in the NH extra-tropics. Transport in longitudinal direction by the powerful jet streams of the NH and SH occurs more rapidly than meridional transport to the tropics and across the ITCZ, leading to a largely zonally invariant distribution in the tropics. Lightning NO<sub>x</sub> is a strong local source of NO<sub>x</sub>, which adds 5–10 DU of TTOC to most columns. Also here, the contribution is almost zonally invariant, although the influence is slightly larger over the Atlantic Ocean than over the Pacific. An almost equal contribution to TTOC comes from stratosphere-troposphere exchange. The contribution consists mostly of stratospheric ozone that crosses the

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tropopause in the sub-tropics, and is subsequently transported to the tropics. This source is generally closer to the tropics than industrial  $\text{NO}_x$  emissions and the zonal distribution shows more variation. The strongest contributors to the ‘wave one’ character of tropical TTOC are soil  $\text{NO}_x$  emissions and biomass burning  $\text{NO}_x$  emissions. These sources clearly contribute strongly to the Atlantic Ocean maximum of TTOC, while exerting only little influence on the tropical Pacific region.

The seasonality of ozone from biomass burning (not shown) is very similar to the seasonality of  $\epsilon_2$  in Fig. 4. In contrast, ozone produced from lightning  $\text{NO}_x$  emissions displays its maximum in April–May–June when the ITCZ crosses the equator going to the NH, while ozone produced from soil emissions is almost constant throughout the year. This again suggests that biomass burning could be partly responsible for the observed Atlantic mismatch. To see which part of our treatment of biomass burning could be the cause of the seasonal ozone underestimate, we have performed a sensitivity analysis.

## 8. Sensitivity analysis

Beside our treatment of biomass burning emissions, the sensitivity analysis includes simulations to investigate uncertainties in the lightning  $\text{NO}_x$  emission parameterization. These two processes occur predominantly in the tropics, in contrast to industrial emissions, soil emissions and STE. Although the seasonality and zonal distribution of lightning  $\text{NO}_x$  emissions do not match the pattern of  $\epsilon_2$  (Fig. 4), its large contribution to the ozone budget and large uncertainties in magnitude and distribution justifies a more detailed look at its parameterization.

### 8.1. Strategy

To ensure an objective intercomparison between different model simulations, we now define  $\epsilon_2$  for two different regions. The first region encompasses all longitudes between

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90 W and 90 E ( $N_{lon} = 90$ ), and the other region encompasses the longitudes outside this area (180 W–90 W and 90 E–180 E,  $N_{lon} = 90$ ). Since the good performance of the model over the Pacific Ocean was determined independently by comparison with ozone sondes (Peters et al., 2001), our strategy to determine whether a simulation constitutes an improvement over our base run is: (1) ensure that  $\epsilon_2$  for the Pacific fit does not increase; (2) achieve a decrease of  $\epsilon_2$  for the Atlantic mismatch, and (3) try to increase the value of  $\epsilon_1$ . Only when these three criteria are met, a systematic improvement of modeled TTOC has been achieved. This has turned out to be a very strict constraint due to the large number of observations and the scale of the phenomenon. All the sensitivity simulations pertain to the year 1992.

Table 2 presents an overview of all the sensitivity simulations that were performed, and their success in meeting the criteria described earlier. Yearly average values of  $\epsilon_1$  and  $\epsilon_2$  for the four simulations that met the criteria are given in parenthesis. The base simulation is the model as used in Peters et al. (2001) as well as in the previous sections of this work. We will discuss the specifics of four of the sensitivity simulations first, while the experiments that did not qualify as improvements are addressed later.

Three of the possible improvements are related to biomass burning. Simulations were performed with an increased injection height of biomass burning products, double  $\text{NO}_x$  emissions from biomass burning, and an alternative fire calendar, respectively. These experiments increase the amount of  $\text{NO}_x$  in the free troposphere over the tropical Atlantic, and thereby increase the photochemical production of ozone. The amount of  $\text{NO}_x$  reaching the free troposphere is determined by the complex interaction between emissions, deposition to the surface, oxidation by OH, and convective venting from the planetary boundary layer (PBL).

The efficiency by which an  $\text{NO}_x$  molecule contributes to ozone formation depends to a large extent on its lifetime, i.e., its ability to reach the free troposphere and avoid dry deposition. Due to the dry conditions prevailing during biomass burning events, strong convective systems can be assumed to be absent near large fires. Moreover,  $\text{NO}_x$  has a lifetime of only several days in the PBL, and in its oxidized form ( $\text{HNO}_3$ ) surface

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deposition is even faster and rainout during convective transport occurs quickly. Therefore, the interaction between meteorology and emissions largely determines whether a  $\text{NO}_x$  molecule will reach the free troposphere, or will be oxidized and/or deposited in the PBL. Our three successful experiments influence this interaction in three different ways.

5 Firstly, we reduce rapid deposition of  $\text{NO}_x$  in the PBL and increase the concentration in the free troposphere by distributing the biomass burning  $\text{NO}_x$  emissions over the lower 6 km of the troposphere. Obviously this is arbitrary, but the fact that this leads to an improvement shows that an improvement would be realized if more biomass  
10 burning  $\text{NO}_x$  were transported to the free troposphere in our model. This could indicate a lack of transport of biomass burning products from the planetary boundary layer (PBL), which is plausible since this parameterization depends on 6-hourly updated diffusion parameters, that can not adequately capture the diurnal growth and decline of the PBL. Using  $\text{Rn}^{222}$ , [Dentener et al. \(1999\)](#) already noticed that our model appears  
15 to underestimate convective periods over the African continent. It could also indicate that biomass burning smoke plumes may reach the free troposphere from their own buoyancy, which is not included in our model. Finally, [Yienger and Levy \(1995\)](#) suggested that post-burning soil emissions of  $\text{NO}_x$  could contribute to the emission budget. These emissions could easily reach the free troposphere under unstable or convective  
20 conditions occurring after the fire activity has ceased.

In addition, the interaction between transport and biomass burning in the model could be improved through implementation of a more realistic fire calendar. As a result, the location and occurrence of convection would become more consistent with the emissions. The convective vertical exchange of  $\text{NO}_x$  decreases in July and August, and increases by 15% in September and October. This leads to more ozone production  
25 in the free troposphere during these months. This agrees well with the findings of [Cooke et al. \(1996\)](#) and [Galanter et al. \(2000\)](#), who applied more detailed alternative fire calendars to point out shortcomings in the inventory of [Hao and Liu \(1994\)](#). The fact that  $\epsilon_1$  increases significantly indicates that the interaction between biomass burning

and convection has a strong influence on the zonal TTOC gradients.

The third important parameter in the description of biomass burning is the emission magnitude. A doubling of the emissions of  $\text{NO}_x$  from biomass burning also increases the flux of  $\text{NO}_x$  from the planetary boundary layer. Table 3 shows the consequences for the tropospheric ozone budget. The increase in production of ozone is largely balanced by an increase in destruction, as well as by an increase in deposition and an increase in advection of ozone out of the Atlantic region. The net effect of a doubling of the  $\text{NO}_x$  emissions is therefore a 2 Tg increase in ozone, which is only 5% of the tropospheric burden. The effect on the TTOC columns is similar (appr. 2 DU). The effect of increased emissions is more pronounced when it is coupled to the alternative fire calendar. In fact, this simulation shows the highest sensitivity. This suggests that the correct representation of the occurrence and magnitude of biomass burning is an important prerequisite to model TTOC, in agreement with Galanter et al. (2000).

An alternative method to increase the concentration of  $\text{NO}_x$  in the free troposphere is through lightning  $\text{NO}_x$  emissions. Our current emission magnitude of  $4.6 \text{ TgN yr}^{-1}$  is in the lower part of the range of Inter Governmental Panel on Climate Change (IPCC) estimates ( $4\text{--}20 \text{ TgN yr}^{-1}$  (Prather et al., 2001)), and a doubling would still be within the uncertainty limits. Table 3 shows that this would yield an improvement in our comparison to TTOC. However, this would also strongly affect the concentrations of ozone in the extra-tropics, leading to a large overestimate of ozone at locations that were previously modeled quite well. A factor of two increase in emissions therefore seems unlikely, unless one would consider a latitude dependent increase of lightning  $\text{NO}_x$  emissions towards the tropics. The relatively large sensitivity to the lightning  $\text{NO}_x$  emissions nevertheless makes this an important process in our simulations and further studies are called for.

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## 9. Vertical ozone profiles

To illustrate the results of our sensitivity experiments we present a comparison with selected vertical ozone profiles from several tropical stations. These stations are part of the Southern Hemisphere Additional Ozone Soundings (SHADOZ) program, which encompasses 11 ozone sonde stations throughout the tropics. The details of the different stations and a comparison to TOMS data and ground-based measurements can be found in: The 1998–2000 SHADOZ (Southern Hemisphere Additional Ozonesondes) Tropical Ozone Climatology: Comparisons with TOMS and Ground-based Measurements, A. M. Thompson et al., J. Geophysical Research – Atmospheres, submitted manuscript, 2001.

Figure 7 shows six observed (black solid lines) vertical ozone profiles from selected stations and dates from the year 1998, as well as several model calculated profiles from 1998. These modeled profiles show results from a standard model simulation (blue dashed lines), a simulation with the meteorology of 1997 applied (red dashed lines), and a simulation with additional biomass burning  $\text{NO}_x$  emissions and an alternative fire calendar (green dashed lines).

In many cases, the base simulation captures the general shape of the vertical ozone profile quite well (a, b, c, d, and f), while the simulation with wrong meteorology does not (a, b, d, and f). This again illustrates the importance of large-scale transport in modeling tropospheric ozone, which we also concluded from our comparison to TTOC in Sect. 6. As expected, adjusting the emission parameterization most strongly influences the stations over the Atlantic Ocean and African continent (c, d, e, and f). What is most important to note from the comparison however, is that many features of the individual sonde profiles are not reproduced by the model. Adjusting the biomass burning emission parameterization, or any of the other parameters we tested, does not appear to lead to systematic improvements.

There are several reasons for this. First of all, we mention the strong damping present in the ozone production, which is obvious from the small differences between

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the blue and green lines in Fig. 7 and from Table 3. Generally, the ozone mixing ratios increase by approximately 0–5 ppbv at all altitudes, leading to an increase of several Dobson Units when vertically integrated. This difference is small compared to the observed underestimate of TTOC. Second, the comparison presented is obviously hampered by the representation of different spatial scales. When we try to improve our model through changes in the parameterizations, the response seems to be strongest on the large scales which are dominated by transport, while the smaller scales remain largely unaffected. Since these smaller scales are dominant in the ozone sondes, intercomparing the different model runs does not convey much information on the performance of the models parameterizations. In fact, it is impossible to judge from Fig. 7 which simulation performs better; green or blue?

The comparison illustrates that individual sondes are not the best product to use when testing a CTM. As was recognized previously by Logan (1999), climatologies of ozone observations are more useful in this respect, even though they lack interannual variability and the density of the measurement network is quite low. We show that the use of TTOC offers a suitable alternative, where the lack of vertical information is ‘compensated’ by the geographical coverage, the frequency of observations, and the length of the observational record.

## 10. Discussion

Our model evaluation depends to some extent on the accuracy of the TTOC data as well as their assumed standard deviations. The meteorological variability in the model and in the satellite TTOC dataset is very similar, which provides some confidence in the retrieval algorithm applied to produce this dataset. Even though the MR retrieval applies sonde observations from two specific years (1990–1991) to produce the entire 1979–1992 record, the details of individual years seem to have been retained well. Nevertheless, the accuracy and the standard deviations of the TTOC observations are not yet well quantified because the ozone sondes do not provide an absolute reference

due to the limited coverage. Moreover, the retrieval algorithm used to derive TTOC is under continuous review and many improvements and updates were made over the past years, and this will probably continue in the next years. Such updates will change the absolute values of TTOC, and thus influence our results.

5 To illustrate possible effects, we calculated the values of  $\epsilon_1$  and  $\epsilon_2$  after applying a uniform negative 5 DU offset to the TTOC measurements. This brought model and measurements almost within one-sigma, and was an even more effective way to reduce the underestimate of TTOC than any of our sensitivity simulations. However, a 5 DU offset in TTOC cannot be justified, and the dependence of  $\epsilon_2$  on the accuracy of  
10 the measured TTOC columns does not invalidate our approach. If the TTOC measurements change, for instance due to an update in the retrieval algorithm,  $\epsilon_2$  can easily be re-defined to focus on regions that consequently appear most problematic, and then applied to the sensitivity runs to isolate possible solutions. Nevertheless, a thorough validation of the retrieved TTOC data using ozone sondes, as well as a more detailed  
15 quantification of the uncertainties would add to the usefulness of these data in global modeling studies.

The fact that a 5 DU reduction in the observations would have greater impact than any of our sensitivity simulations further illustrates the robustness of ozone to perturbations. This is also obvious from the small ranges in  $\epsilon_1$  and  $\epsilon_2$  in Table 2. Although  
20 the change in forcing is often quite extreme (factor of two), the response of ozone is only small. Table 3 illustrates that this nonlinear, damped response is mostly due to strong feedbacks in the chemistry, as well as in the deposition working against any change in the concentration of ozone. These feedbacks are especially important in the tropics, since both photochemistry and uptake of ozone by vegetation depend strongly  
25 on the sunlight intensity. It is also the non-linearity of the system that requires CTMs to use sophisticated mathematical solvers for the chemistry with small time steps to solve the system of partial differential equations (PDEs), which generally consumes a large amount of computer resources.

An effective way to reduce the demand of computer resources is by linearizing the

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chemistry. The solution of the system of PDEs is then pre-calculated for many different boundary conditions and initial conditions, and the solution of the system at each time step is retrieved from an archive of solutions matching the forcing. Klonecki and Levy (1997) have shown that this can be done in a way that retains many of the feedbacks, and their model performs well in simulating tropical ozone, CO and NO<sub>x</sub>. Moreover, the method of Klonecki and Levy (1997) seems to produce more ozone over the tropical Atlantic and display greater sensitivity to forcing (see also Galanter et al., 2000), which is difficult to reconcile with our results. We speculate that by linearizing the chemistry, the sensitivity of these models will be larger than in models that calculate the solution of the PDEs at each time step.

Our results show that the meteorology particularly strongly influences the distribution of ozone. It is striking that the range of values of  $\epsilon_1$  in Fig. 5 is much larger than the range in Table 2. Apparently, zonal gradients in TTOC are largely determined by transport. Using actual meteorology has clear advantages over “climatological” meteorology, because the interannual variability is large, also addressed by Peters et al. (2001). Although the differences in correlation might seem quite low, it is important to realize that we are comparing vertically integrated, monthly mean, meridionally averaged ozone patterns. The fact that improvements can be achieved indicates that the differences associated with transport are an important aspect of the models performance. Nevertheless, many models still rely on climatological wind fields, or apply one year of meteorological analyses to simulate any year of interest. This approach will yield results that are best compared to climatologies of observations, and are much less suited to study multi-year time series, or instantaneous distributions of species with lifetimes that are smaller than, or comparable to, the time scale of transport.

The requirements of the model concerning meteorology are very demanding. Not only does large scale transport determine the shape of the zonal TTOC distribution, meteorology also introduces interannual variability, and its interaction with biomass burning emissions is one of the most sensitive model parameters. However, meteorological conditions in the tropics are complex, and their representation in numerical

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weather prediction models such as the ECMWF model suffers from a lack of in-situ measurements that are used as constraints. Especially subgrid scale vertical mixing processes are difficult to model. This includes the vertical transport of biomass burning plumes. Mauzerall et al. (1998) estimated that the majority of ozone production takes place in these plumes. It thus seems obvious that besides improvements of our knowledge on the occurrence and intensity of fires, further improvements should focus on the sub-grid representation of turbulent and convective transport of biomass burning plumes.

Our study shows most convincingly that it is very unlikely that a single factor affecting ozone is responsible for the observed differences between model and observations. Given the sensitivities as presented in Table 2, a combination of processes would be required to bring observations and model within one-sigma. This fact, together with the strongly damped response of ozone to the applied forcing strongly complicates attempts to infer information on model parameters (e.g. lightning NO<sub>x</sub> and biomass burning emissions) from the tropical distribution of TTOC. The simultaneous use of global CO and NO<sub>x</sub> observations, for instance, could be useful in this respect, as these could help to constrain the emissions. Ongoing developments in remote sensing might make these products available on spatial and temporal scales suitable for the approach presented in this work.

## 11. Conclusions

We have combined satellite observations of TTOC with model calculations to test the performance of our model and prioritize directions for improvement. Such a systematic comparison of model calculations with satellite observations has not been performed before, and it yields interesting insights in problems and possibilities associated with the use of increasingly available space-borne tropospheric observations. Through a large number of sensitivity runs, we were able to quantify the influence of meteorology, as well as biomass burning and lightning NO<sub>x</sub> emissions on the simulation of TTOC.

Our most important conclusions are:

- With a standard set of emissions such as in [Houweling et al. \(1998\)](#); [Lelieveld and Dentener \(2000\)](#); [Peters et al. \(2001\)](#) the Atlantic TTOC maximum is not sufficiently captured, whereas the interannual variability over the Pacific Ocean is represented quite well.
- Using actual meteorology in a model is the most important prerequisite to reproduce the horizontal and vertical distribution of tropical tropospheric ozone.
- Efficient feedbacks in the tropical ozone budget strongly damp the response of ozone to changes in precursor emissions, which makes ozone an unsuitable tracer to improve the representation of precursor emissions in global models.
- Systematic improvements in our model can be achieved through a better representation of the biomass burning timing and magnitude, including their interannual variability.
- Quantitative use of satellite derived TTOC to test a global model is a promising approach, that would strongly profit from a further reduction of the uncertainties in TTOC data.

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**Table 1.** Processes included in the model and references to the original work

Process	Based on	Ref. <sup>a</sup>
Advection	<a href="#">Russel and Lerner (1981)</a>	<a href="#">Dentener et al. (1999)</a>
Convection (subgrid)	<a href="#">Tiedtke (1989)</a>	<a href="#">Dentener et al. (1999)</a>
Turbulent mixing (subgrid)	<a href="#">Louis (1979)</a>	<a href="#">Dentener et al. (1999)</a>
Stratospheric boundary		<a href="#">Lelieveld and Dentener (2000)</a>
Photolysis	<a href="#">Landgraf and Crutzen (1998)</a> <a href="#">Krol and van Weele (1997)</a>	
Dry deposition	<a href="#">Wesely (1989)</a>	<a href="#">Ganzeveld et al. (1998)</a>
Wet deposition	<a href="#">Guelle et al. (1998)</a>	<a href="#">Jeuken (2000)</a>
Biomass burning emissions <sup>b</sup>	<a href="#">Hao and Liu (1994)</a>	<a href="#">Marufu et al. (2000)</a>
Lightning NO <sub>x</sub> emissions <sup>b</sup>	<a href="#">Price and Rind (1992)</a> <a href="#">Levy et al. (1996)</a>	<a href="#">Meijer et al. (2001)</a>
Other emissions	<a href="#">Olivier et al. (1996)</a>	<a href="#">Lelieveld and Dentener (2000)</a>
Chemistry, CBM-IV <sup>b</sup>	<a href="#">Gery et al. (1989)</a> <a href="#">Houweling et al. (1998)</a>	<a href="#">Houweling et al. (1998)</a>

<sup>a</sup> This column refers to previous work with the TM3 model.

<sup>b</sup> This process is discussed in Section 2.

**Table 2.** Experiments conducted and their success in meeting the criteria described in Section 8.1. Positive (negative) signs indicate an improvement (deterioration) of the parameter  $\epsilon_1$  or  $\epsilon_2$  compared to our base simulation, the letter m denotes a marginal change. Values for experiments that qualify as improvements are given in parenthesis

Experiment	$\epsilon_1$	$\epsilon_{2,Pac}$	$\epsilon_{2,Atl}$
Base	0.84	0.8	1.8
<i>Biomass burning</i>			
2 × NO <sub>x</sub>	+ (0.85)	+ (0.7)	+ (1.6)
2 × CO	m	+	+
2 × NMHC	m	+	+
2 × J(HNO <sub>3</sub> )	-	-	+
Injection height	+ (0.85)	+ (0.7)	+ (1.5)
Alternative calendar	+ (0.85)	+ (0.8)	+ (1.7)
<i>Lightning</i>			
2 × NO <sub>x</sub> lightning	+ (0.85)	+ (0.5)	+ (1.1)
5 × NO <sub>x</sub> oceans	-	+	+
Pickering profiles	-	+	-
Coupling to conv. prec.	-	-	-
land/ocean ratio 2:1	-	+	-
<i>Other</i>			
P(O <sub>3</sub> ) from J(O <sub>2</sub> )	-	-	+
150% subsidence	-	-	+
150% convection	m	-	-
increased model resolution	m	+	m
5 DU offset	m (0.84)	+ (0.6)	+ (1.0)

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**Table 3.** Budgets of ozone in the base run, and in a simulation with double  $\text{NO}_x$  emissions from biomass burning. The last column shows the difference between both simulations. Numbers are tropospheric integrals over the tropical Atlantic region and adjacent continents [80W-40E,12S-12N]<sup>a</sup> for the year 1992

	Base (A)	double $\text{NO}_x$ (B)	B-A
Chem. production	1095	1200	+105
NO+HO <sub>2</sub> (%)	62	62	0
NO+CH <sub>3</sub> O <sub>2</sub> (%)	16	16	0
NO+XO <sub>2</sub> (%)	17	17	0
NO+CH <sub>3</sub> COO <sub>2</sub> (%)	5	5	0
Chem.destruction	951	1019	+68
O <sub>3</sub> + hv (%)	59	58	-1
O <sub>3</sub> + OH (%)	10	10	0
O <sub>3</sub> + HO <sub>2</sub> (%)	25	26	+1
O <sub>3</sub> + HC (%) <sup>b</sup>	6	6	0
Net chemistry	+144	+181	+37
Advection	26	7	-19
Dry deposition	166	182	+16
STE <sup>c</sup>	0	0	0
Total	+4	+6	+2
Tropospheric burden	40	42	+2

<sup>a</sup> Units are  $\text{TgO}_3 \text{ yr}^{-1}$ , unless percentage explicitly stated.

<sup>b</sup> HC denotes isoprene, ethylene and olefinic hydrocarbons.

<sup>c</sup> direct STE in this region is negligible.

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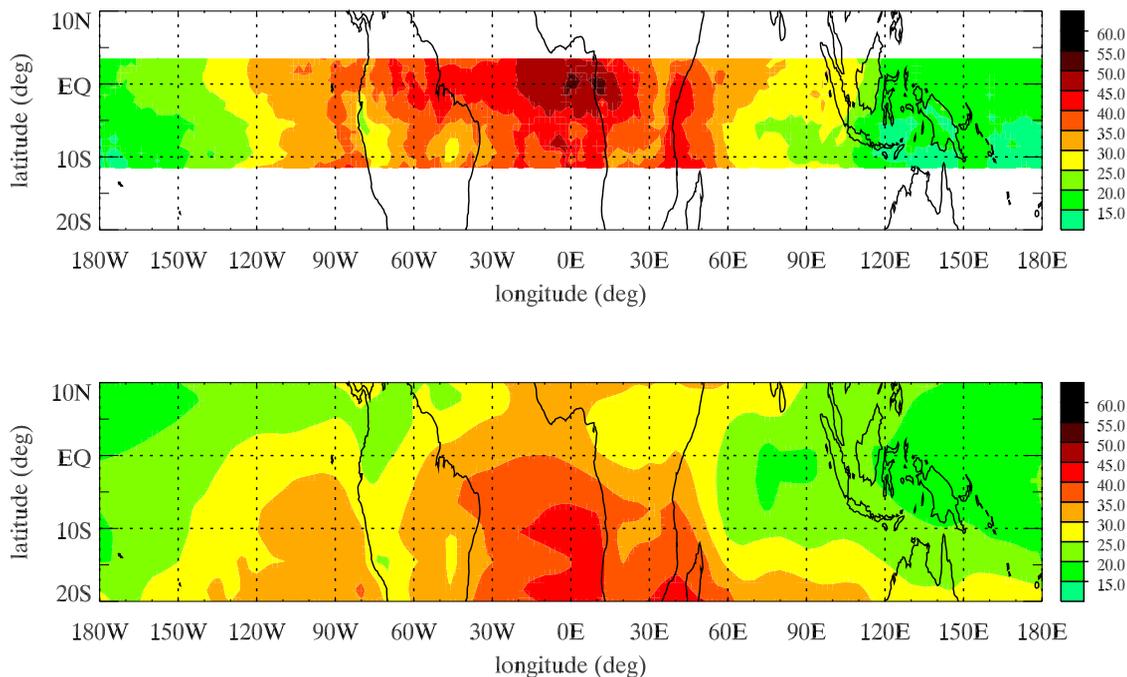
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**Fig. 1.** Observed (top) and model calculated (bottom) distribution of TTOC in September 1992. Note the widespread underestimate of TTOC over the tropical Atlantic Ocean and adjacent continents.

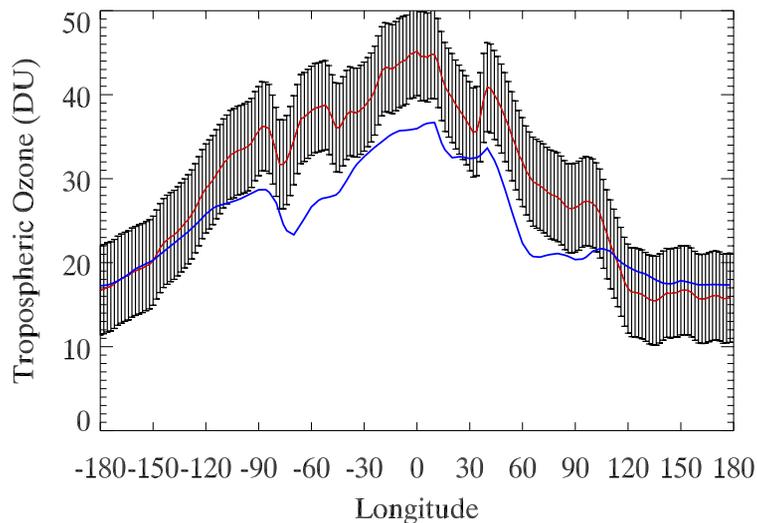
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**Fig. 2.** The meridionally averaged zonal TTOC distribution in model calculated (blue) and satellite retrieved (red) TTOC for September 1992. This distribution is used to define a measure of error according to Eq. (1). The shaded lines denote one-sigma standard deviation (5.3 DU)

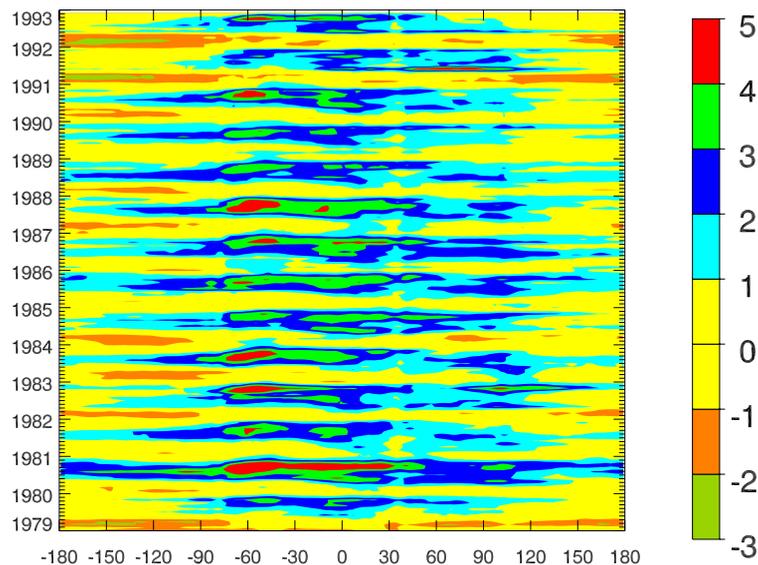
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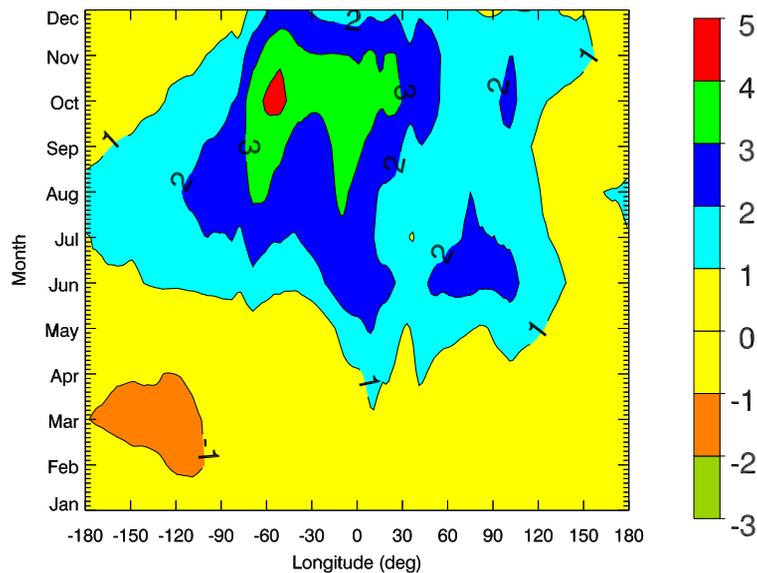
**Fig. 3.** Hövmoller plot of  $\epsilon_2$  as a function of longitude for the period 1979–1992. A value of 1 denotes a difference of 5.3 DU ( $1-\sigma$ ) between model and observations. The  $+/-$  signs were added to the figure to signify model underestimates or overestimates respectively.

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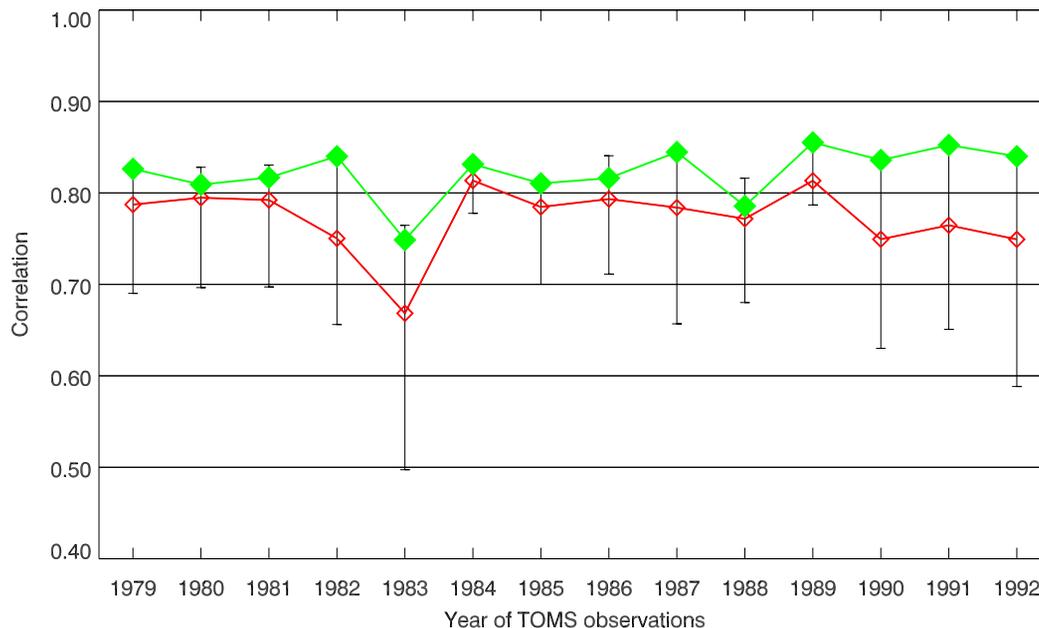
**Fig. 4.** 14-Year average seasonal cycle of  $\epsilon_2$  as a function of longitude. A value of 1 denotes a difference of 5.3 DU ( $1-\sigma$ ) between model and observations. The  $+/-$  signs were added to the figure to signify model underestimates or overestimates respectively.

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**Fig. 5.** Correlation coefficient ( $\epsilon_1$ ) between the zonal TTOC distribution in observations and simulations. For each year of TTOC observations (1979–1992) correlations were calculated with each year of a multi-year simulation with our CTM (1979–1992). The bars denote the range of correlations, the red diamonds are the averages. This value is representative for a simulation with ‘climatological’ meteorology. The green diamonds signify the correlation when the correct meteorology of the year of observations is applied to the model.

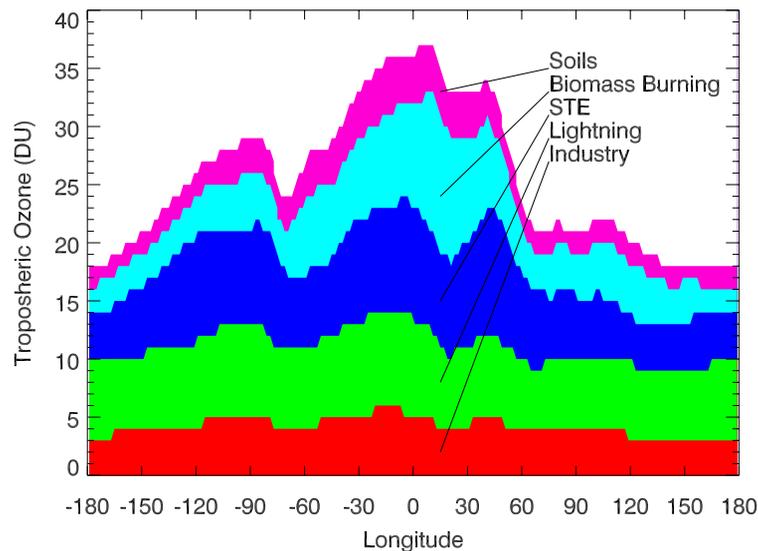
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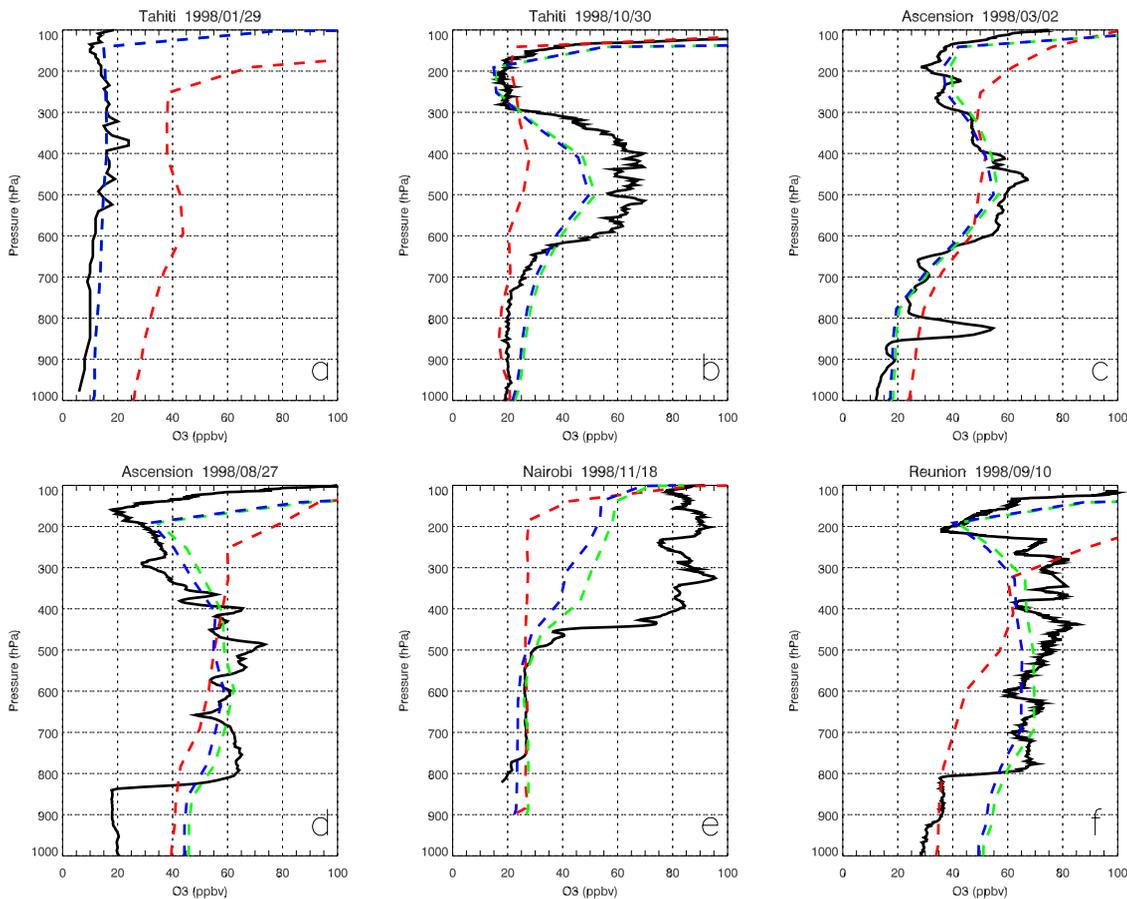
**Fig. 6.** Absolute contribution of several processes to the modeled TTOC distribution. Processes include photochemical production of ozone following  $\text{NO}_x$  emissions from soils, lightning, biomass burning and industrial sources, as well as ozone from stratosphere-troposphere exchange. Values for September 1992.

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**Fig. 7.** Selected sondes from six SHADOZ stations in 1998. The profiles show observations (black), a standard model run for 1998 (blue dashed), a model run with meteorology from 1997 (red dashed), and a model run with additional  $\text{NO}_x$  emissions and a new fire calendar (green dashed).

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