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The influence of biogenic emissions from Africa on tropical tropospheric ozone during 2006: a global modeling study

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The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

We have performed simulations using a 3-D global chemistry-transport model to investigate the influence that biogenic emissions from the African continent exert on the composition of the tropopause in the tropical region. For this purpose we have applied two recently developed biogenic emission inventories provided for use in large-scale global models (Granier et al., 2005; Lathi  re et al., 2006) whose seasonality and temporal distribution for isoprene, biogenic NO and biogenic volatile organic compounds is markedly different. The use of the climatological values for biogenic emissions provided by Lathi  re et al. (2006) results in an increase in the amount of nitrogen sequestered into longer lived reservoir compounds which contributes to the reduction in tropospheric ozone burden in the tropics. The associated re-partitioning of nitrogen between PAN, HNO₃ and organic nitrates also results in a ~5% increase in the loss of nitrogen by wet deposition. At a global scale there is a reduction in the oxidizing capacity of the model atmosphere which increases the atmospheric lifetimes of CH₄ and CO by ~1.5% and ~4%, respectively. By the use of sensitivity studies we show that the release of NO from soils in Africa accounts for between ~5–45% of tropospheric ozone in the African troposphere, ~10% in the upper troposphere and between ~5–20% of the tropical tropospheric ozone column over the tropical Atlantic Ocean. The subsequent reduction in OH over the source regions allows enhanced transport of CO out of the region. For biogenic volatile organic C1 to C3 species released from Africa the effects on tropical tropospheric ozone are rather limited, although this source contributes to the global burden of VOC by between ~2–4% and has a large influence on the organic composition of the troposphere over the tropical Atlantic Ocean. Comparisons against a range of different measurements indicate that applying the climatology of Lathi  re et al. (2006) improves the performance of TM4 for 2006 in the tropics.

ACPD

9, 10367–10427, 2009

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



1 Introduction

Biogenic emissions of trace gas species from vegetation and soils are considered to be an important emission source that can significantly affect the composition of the troposphere for many tropical regions (e.g. Aghedo et al., 2007). Although isoprene is the most dominant biogenic emission (Guenther et al., 2006) recent field measurements have shown that a diverse range of Biogenic Volatile Organic Compounds (BVOC's) are released at lower rates from a range of vegetation types (e.g. Villanueva-Fierro et al., 2004). Therefore, to be able to differentiate the contributions from anthropogenic and natural activity on tropospheric ozone in remote regions necessitates that accurate estimates related to the global fluxes of such BVOC's are available.

The most important biogenic emission for tropospheric ozone production is the direct emission of nitric oxide (NO) from recently wetted-soils. This is now widely recognized as being an important natural source of reactive nitrogen at remote locations in the tropics (e.g. Yienger and Levy, 1995). The source strength is dependent on certain meteorological and geophysical variables such as recent precipitation history, soil type and temperature (Ludwig et al., 2001; Delon et al., 2009). The optimal combination of these parameters results in intense pulses of NO being emitted for short periods of time after any rainfall event (Davidson, 1992; Stewart et al., 2008). Such events are often followed by periods of relative inactivity where the emission of NO is much lower. This imposes a seasonal cycle on the emission between the dry and wet seasons, where biogenic NO_x emissions exhibit only moderate inter-annual variability compared to biomass burning emissions (Van der Werf et al., 2006; Van der A et al., 2008). For sparsely populated regions such as Equatorial Africa, these emissions have the potential to influence the background levels of tropospheric ozone (O₃) and, thus, the atmospheric lifetimes of other important trace gas species such as CO and CH₄ via perturbations in [OH], especially considering that a large fraction of the total global oxidation of such trace species occurs in the tropics.

Further information regarding both the scale and importance of such biogenic emis-

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



sions has recently been forthcoming by the analysis of observations made from a host of earth-orbiting satellites. For instance, Jaeglé et al. (2004) used NO₂ columns retrieved from the Global Ozone Monitoring Experiment (GOME), in tandem with independent fire observations, to investigate the spatial and seasonal variation of the release of NO from soils. They conclude that such soil emissions account for ~40% of surface NO_x in Africa, affecting 3×10⁶ km² of sub-Saharan Africa during the wet season. Another prominent example is the decadal trend analysis conducted by Van der A et al. (2008) using a composite of data from various satellite instruments, where they conclude that NO_x emissions from soils are the dominant sources of nitrogen oxides in northern Africa (between ~12–35° N). Moreover, the maximum emissions occur in the summer months of each respective hemisphere, which is somewhat anti-correlated with the release of NO_x from biomass burning events. For BVOC's the detection of surface concentrations from satellites is problematic due to the weak spectral absorption features exhibited by most compounds in the spectral windows typically used for such measurements and the relatively low concentrations (typically less than a nmol/mol). Although HCHO can be retrieved from such satellite observations (e.g. Palmer et al., 2003), the predominant fraction in remote regions is assumed to come from the oxidation of isoprene (C₅H₈) rather than as a result of a direct biogenic emission.

The large surface area over which soil NO emissions are important means that any model used to investigate the composition of the troposphere in the tropics needs to include an accurate inventory. To date, the predominant emission inventory used in large scale global 3-D Chemistry Transport Models to account for the emissions of NO from soils is that provided by Yienger and Levy (1995), which results in a global estimate of 5.5 TgN/year. This is somewhat less than the annual emission flux estimated by Jaegle et al. (2004) of 7.3 TgN/year, which is more consistent with the inventory of Davidson and Kingeree (1997) of 7.9 TgN/year. For the biogenic VOC's there are only a few estimates currently available, which are widely used throughout the modeling community. For instance the release of biogenic CO is typically set equal to the values taken from Müller and Brasseur (1995), regardless of the emission inventory, whereas

The influence of biogenic emissions from Africa

J. E. Williams et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

for the C2 compounds the values of Müller (1992) are still commonly used.

A large source of error when calculating such global biogenic emission estimates occurs during the process of upscaling, which can be partly overcome by the use of more sophisticated biogeochemical models that utilize dynamic vegetation data. Such models account for the correct distribution, plant biophysics, rate of growth, variations in leaf area index (LAI) and differences in vegetation type (e.g. Potter et al., 1996; Lathi  re et al., 2006), which all influence the magnitude and seasonality of regional biogenic emissions. More sophisticated interactive approaches have also become available (e.g. Ganzeveld et al., 2002) which update the algorithm of Yienger and Levy (1995) in order to account for the subscale chemical processes and mixing which occur under/through the tree canopy. By coupling other meteorological parameters such as precipitation rates into the calculation of the NO emissions, the inter-annual variability can be captured with more accuracy. Finally, Delon et al. (2007) have proposed a method based on a neural-network approach that reacts towards climatic variables on very short time-scales. They have recently coupled this to a meso-scale model for studying an event in Equatorial Africa observed during the West African Monsoon (WAM) as part of the African Monsoon Multidisciplinary Experiment (AMMA) (Delon et al., 2008; Stewart et al., 2008). This resulted in increases in tropospheric O₃ in the lowest few kilometers of the atmosphere compared to simulations using the inventory of Yienger and Levy (1995). For BVOC's a new set of estimates has recently become available for trace species such as HCHO and CH₃COOH (Lathi  re et al., 2006). Moreover, there are also corresponding estimates for isoprene and NO, which ensures consistency with the method from which they are calculated.

In this study we investigate the influence of NO and BVOC emissions from soils and vegetation on the composition of the troposphere above Equatorial Africa, and surrounding regions affected by outflow, as part of the AMMA integrated research project (www.amma-eu.org; Redelsperger et al., 2006). For this purpose we use the recent inventory for biogenic emissions derived from multi-annual simulations between 1983 and 1995 from the ORCHIDEE model (Lathi  re et al., 2006) provided as part of the

The influence of biogenic emissions from Africa

J. E. Williams et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

RETRO emission database (<http://retro.enes.org>). This is compared to the alternative estimates made available in the Present and future surface Emissions of atmospheric compounds (POET) database (Granier et al., 2005) to quantify the differences introduced into large-scale simulations of the global troposphere.

In Sect. 2 we give details concerning the model description and the sensitivity studies that have been performed. In Sect. 3 we will highlight the differences in the seasonality and magnitude between the two emission inventories for the African continent. In Sect. 4 we will show the effect of both soil NO_x and other BVOC's on tropospheric ozone and the associated NO_x reservoir species PAN and HNO₃. In Sect. 5 we will then investigate the effect on the oxidative capacity of the global troposphere by analysis of the chemical budget terms. In Sect. 6 we will then assess the performance of the model against a suite of measurements taken in and around Equatorial Africa, and finally, we will present our conclusions in Sect. 7.

2 Model description

The global CTM used in this study is the TM4 model, which has recently participated in a host of multi-model intercomparison exercises (e.g. Van Noije et al., 2006; Stevenson et al., 2006). The model version is identical to that described in Van Noije et al. (2006) except that the $\gamma_{\text{N}_2\text{O}_5}$ value has been reduced to 0.02 in line with the recommendations of Evans and Jacob (2005) and the rate co-efficient data has been updated according to the latest recommendations (Atkinson et al., 2006; Sander et al., 2006). Moreover, the photolysis rates have also been re-assessed using a chemical box-model, and the scaling parameters have been updated for the photolysis of the lumped organic nitrate species and methylglyoxal. The effects of this are discussed in depth in Williams and van Noije (2008). TM4 is driven by ECMWF meteorological data originally provided on 91 vertical layers but subsequently coarsened onto the TM4 resolution of 34 layers, where the vertical resolution is typically 1 km in the tropical upper troposphere/lower-stratosphere. The horizontal resolution used throughout was 3° longitude×2° latitude.

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Since TM4 does not include stratospheric chemistry, the total overhead ozone column was nudged towards the monthly mean averages from the Ozone Monitoring Experiment (OMI) using the climatology of Fortuin and Kelder (1998) to define the stratospheric profile.

5 For the anthropogenic emissions we adopt the sector segregated estimates for the year 2000 made available through the RETRO website (<http://retro.enes.org>). These are based on the TNO Emission Assessment Model (TEAM) and are segregated with respect to the sector from which the emission originates. The emissions from all sectors except transport are injected between 0–200 m above the ground. These are supplemented with inventories for NH_3 and SO_2 taken from the EDGAR-HYDE version 1.3
10 (Van Aardenne et al., 2001). For ship emissions we adopt the estimates from Endresen et al. (2003) and for aircraft emissions those from the AEROCOM project (Lee et al., 2002; Grewe et al., 2002). For biomass burning emissions we use the monthly aggregated estimates from the GFEDv2 database (Van der Werf et al., 2006) and apply the
15 injection heights given in Lavoué et al. (2000). These biomass burning emissions are injected at a fixed rate throughout each day of the month. For lightning NO_x we apply the parameterization of Meijer et al. (2001) which gives a global lightning source of $\sim 5.9 \text{ TgN}$ for 2006, which is slightly higher than the estimate of $\sim 5 \text{ Tg}$, but well within the current uncertainty range (Schumann and Huntrieser, 2007). For all runs we do not
20 include specific emissions for CH_4 but impose a latitudinal gradient at the surface. It should be noted that non-eruptive volcanic SO_2 emissions are taken from Andres and Kasgnoc (1998).

In order to investigate the influence of the different biogenic inventories we define a number of different experiments where we vary the biogenic emission fluxes applied
25 in the model. For the baseline simulation (hereafter referred to as BASE) we use the biogenic emission files supplied from the POET project (Granier et al., 2005), which provide values for biogenic NO , CO , CH_3OH , C_2H_4 , C_2H_6 , C_3H_8 , C_3H_6 , acetone, isoprene and the monoterpenes. For the simulation using the 12 year climatology of Lathiére et al. (2006) from 1983–1995 (hereafter referred to as LATH) we replace the

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



specific inventories for NO, CH₃OH, acetone, isoprene and the monoterpenes. In the tropics (20° N–20° S) we distribute the emission of isoprene over the first two model layers and impose a diurnal variation on the emission to account for the effects of sunlight and temperature. For the other BVOC's we adopt the POET biogenic inventories for CO, C₂H₄, C₂H₆, C₃H₈ and C₃H₆ and supplement them with additional ORCHIDEE inventories for CH₃CHO, CH₃COOH, and HCHO.

The TM4 model uses the modified CBM4 chemical mechanism of Houweling et al. (1998), which represents C2 and higher organics as a set of lumped organic species based on the reactivity of the characteristic functional groups. Table 1 shows how each of the biogenic species described above is introduced into TM4 and summarises the degree of commonality between each of these runs with respect to biogenic carbon emissions. The speciation into CBM lumped species was done according to the details given in Yarwood et al. (2005). Here explicit species are partitioned according to the functional groups which they contained (e.g.) PAR for paraffinic bonds and OLE for olefinic bonds. A main difference between the inventories is that no ALD2, OLE or HCHO is introduced from biogenic sources in the BASE run.

To determine the influence of NO emitted from soil in Africa we define a sensitivity study where the soil NO flux is removed for the African Continent (hereafter referred to as NOSOIL). To investigate the cumulative effect of the BVOC's we define a sensitivity study where we remove all biogenic emissions of organic compounds over Africa, except those for isoprene and the monoterpenes (hereafter referred to as NOBIO). The emission fluxes for these two species are so large that we feel it would be unrealistic to completely remove them and the effects of isoprene are not the focus of these studies. Such effects have been covered previously in the literature (e.g. von Kuhlmann et al., 2004) therefore we place the focus on the other biogenic carbon emissions available in the inventory, which are not included in many models.

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3 Differences in the emission inventories for Africa

In this section we investigate the differences in the biogenic emission inventories for both the BASE and LATH runs. For this study we principally focus on Africa (defined here as between 20° W–40° E, 40° S–40° N) and adopt four latitudinal zones in Africa similar to those defined in the AMMA Multi-model Intercomparison Project (AMMA-MIP, Williams et al., 2009), these being: Sahara (20–40° N), Sahel (10–20° N), Guinea (0–10° N) and southern Africa (40° S–0° N). This corresponds to land surface areas of $8.59 \times 10^6 \text{ km}^2$, $6.55 \times 10^6 \text{ km}^2$, $5.63 \times 10^6 \text{ km}^2$ and $8.32 \times 10^6 \text{ km}^2$, respectively.

Figures 1 to 3 show direct comparisons of the monthly emission fluxes for biogenic NO, isoprene and organic carbon, respectively, for all four latitudinal zones. It can be seen that for both the NO and BVOC monthly emissions the seasonal distribution differs markedly between the inventories. Table 2 compares the resulting annually integrated anthropogenic, biomass burning and biogenic emission totals for each latitudinal zone in order to quantify the importance of the biogenic emission for each zone. For biogenic emissions of NO most latitude zones exhibit a contribution that exceeds that from anthropogenic activity. The exception is the Sahara region where the dry conditions and cities in the southern Mediterranean result in a higher contribution from anthropogenic emissions. Moreover, the differences in the annually integrated fluxes between the inventory of Yienger and Levy (1995) and Lathi  re et al. (2006) are relatively small for all regions, although the seasonality is much larger in Lathi  re et al. (2006) as shown in Fig. 1. There are also differences with respect to longitude that are not highlighted by the latitudinal comparison. For instance, during February there is more surface NO_x near the west coast of Africa around 10° S in the Lathi  re et al. (2006) inventory. Such differences will become apparent when performing a comparison of model output from the various simulations with ground measurements (see Sect. 6). For CO, where the same biogenic inventory is used in both simulations, the contribution from biogenic activity is the lowest of all emission sources, although still not negligible, reaching ~50% of the anthropogenic contribution for the Guinea region. For isoprene, the POET inven-

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



tory has higher annual emissions for all regions, where the differences are especially large for the Sahel and southern Africa. This can be partially explained by considering the origin of the estimates in both inventories. In the POET inventory the estimates from Guenther et al. (1995) are employed whereas in Lathi  re et al. (2006) the variation in the distribution of vegetation due to deforestation and climatic change between 1983 and 1995 are accounted for, which has the potential to reduce the annual global emission of isoprene substantially (Ganzeveld and Lelieveld, 2004). However, the largest differences between the emission inventories are related to the other BVOC's, where differences of >200% occur in the monthly fluxes, although in terms of total biogenic carbon this is somewhat negated by the increased isoprene emissions in the POET inventory from each region.

4 The Seasonal differences in tropospheric O₃ and NO_x over Africa

Once emitted, NO is oxidized to NO₂ via the reaction with HO₂, CH₃O₂ or O₃. This additional NO₂ can then be either rapidly photolysed (producing O₃) or react with reactive free-radical species such as OH, HO₂ or CH₃C(O)O₂ to form more stable reservoir species for reactive nitrogen, namely HNO₃, HNO₄ or PAN. A further nitrogen reservoir is included in TM4 in the form of a lumped organic nitrate (hereafter referred to as ORGNTR). However, for brevity we exclude it from the subsequent comparisons shown in the figures below and simply make a few comments on the subsequent changes as a result of adopting the various inventories. Such reservoir species have longer atmospheric lifetimes than NO₂ and, once formed, maybe transported away from the main source regions by convective and advective transport processes. Upon decomposition, photolytic destruction or oxidation these species may then re-release reactive nitrogen far away from the source regions. Moreover, during the wet season a certain fraction is removed via heterogeneous scavenging (e.g.) transfer of HNO₃ and ORGNTR into cloud droplets/wet aerosol followed by wet deposition. The additional BVOC in the LATH inventory also alters the HO_x budget by increasing the fraction of OH scavenged

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



by organic trace species (see Sect. 5).

In order to investigate the differences in the distribution and resident concentration of tropospheric O_3 and associated NO_x reservoirs between each of the emission inventories used in this study we have defined a number of 2-D cross-sections which reflect the “zonality” of the African region similar to that used in AMMA-MIP project (Williams et al., 2009). A subset of the chemical species is averaged over a 9° longitude range between $12\text{--}21^\circ$ E for a latitude range of 20° S to 40° N, which is chosen as it encompasses a wide range of land types. The values have been written out by TM4 every three hours for all vertical levels which allows various comparisons to be performed offline for the whole of the region using different averaging frequencies (i.e.) daily, weekly or monthly. Here we wish to look at the seasonal behaviour and therefore use seasonal averages of the data.

Figure 4 shows the distribution of tropospheric O_3 as a seasonal mean calculated from the 2-D cross section for the LATH simulation. Plots are shown for all seasons, namely December-January-February (DJF), March-April-May (MAM), June-July-August (JJA) and September-October-November (SON) of 2006, along with the corresponding percentage differences when compared to the BASE simulation. For the lower troposphere the highest $[\text{O}_3]$ occurs between $\pm 10^\circ$ latitude during the seasons DJF and JJA, where there is a shift from the Northern Hemisphere (hereafter referred to as NH) to the Southern Hemisphere (hereafter referred to as SH) in the course of the year. The maximum $[\text{O}_3]$ values of ~ 100 ppbv occur near regions influenced by intense biomass burning activity. Later in Sect. 6, we will show that TM4 generally has a tendency to over-estimate surface O_3 when compared to different observational datasets. The differences in $[\text{O}_3]$ when compared with the BASE run show regional similarities with those that occur between the soil NO_x emission inventories as shown in Fig. 1, with maximum differences occurring in southern Africa during season DJF. For periods where there are similar regional biogenic NO_x emissions (e.g. southern Africa during MAM) there is also a signature present from the higher isoprene emissions present in the BASE run. This contributes to the depletion of O_3 during the night, where the

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



magnitude of differences typically equals a few percent. For the Sahel region, which is devoid of any significant isoprene emission in the 2-D transect (not shown), there are persistent decreases for the LATH run in the lower troposphere throughout the year. This occurs even though there is a moderate “peak” in soil NO for season JJA as shown in Fig. 1. The reason is that there are temporal (longitudinal) differences in the release of NO from soils within the latitude band (not shown). For the upper troposphere (hereafter referred to as UT) there are generally decreases of <5%, apart from regions directly influenced by convective updraft from the lower layers. These decreases at higher altitudes pertain to higher [O₃] values, which results in the LATH run having a lower tropospheric O₃ burden than the BASE run when integrating the differences throughout the tropospheric column (see Sect. 5). In general, when compared to aircraft measurements, TM4 shows a tendency to under predict UT [O₃] for certain seasons (see Sect. 6). These decreases in tropospheric [O₃] extend out over the tropical Atlantic Ocean. This induces changes in the tropical tropospheric ozone column (TTOC) simulated in the model, which is defined as the integrated ozone column below 200 hPa as used in Valks et al. (2003). A signature of the enhanced seasonality present in the Lathi  re et al. (2006) emission inventory for soil NO_x can also be seen in the differences in the TTOC as far as South America (e.g.) increases of ~10% occur for JJA between 0–20   N (not shown). This has the potential to make improvements to previous comparisons of the TTOC between 3-D CTM’s and satellites, such as those presented in Peters et al. (2002) and Valks et al. (2003).

Figure 5 shows a similar figure for PAN. For seasons DJF and JJA the tropospheric distribution is quite similar to O₃, where maximal [PAN] occurs near regions of intense biomass burning activity as a result of the enhanced NO_x emissions reacting with isoprene oxidation products. As a result there is also a signature in [PAN] in the lower levels from regions exhibiting high isoprene emissions for other seasons (e.g. 5–10   N during MAM). Analysing the chemical budget shows that the dominant source of the CH₃C(O)O₂ radical for the tropics in the modified CBM4 mechanism is via the oxidation of aldehydes, whose main source is from the oxidation of the other lumped organic

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



species (Houweling et al., 1998). This explains why the highest [PAN] generally occurs between 10° S–10° N over densely forested regions and the lowest [PAN] (shown as blue areas in Fig. 5) occurs over the Sahel. Once formed, a significant fraction is subsequently transported via convective mixing throughout the entire troposphere, which ultimately dissociates releasing NO₂. Comparing the differences between the simulations shows that for the NH there are generally increases in [PAN] in the LATH run, especially for season JJA. As a result [PAN] increases of ~10–20% also occur in the troposphere of the tropical Atlantic Ocean, where there is a reduction in [NO_x] by a few percent (not shown). For latitudes >20° N a significant fraction is advected into northern Africa from southern Europe as a result of the higher isoprene emissions that are introduced into southern Europe in the Lathi  re et al. (2006) inventory. Another competing effect is an increase in the partitioning of NO₂ into HNO₃ as a result of the elevated [OH] formed via the photolysis of the enhanced [O₃] in the LATH run (see below). This repartitioning contributes to the decreases in [PAN] which are shown for the UT above southern Africa during seasons DJF and SON. The increases in the UT in the NH are again due to elevated [PAN] being advected into the region from outside of Africa.

Finally Fig. 6 shows the corresponding plot for HNO₃, which is the most abundant NO_x reservoir in the model with a burden that is ~5 times that of the other nitrogen reservoirs in the tropics (see Sect. 5). The formation of HNO₃ is linked to the photolysis of O₃, which explains the strong similarities that occur between the increases shown in Figs. 4 and 6. Such a dependency can also be seen as a seasonal cycle when analyzing ground based measurements, as we show later in Sect. 6. The highest [HNO₃] values again occur near the regions affected by biomass burning emissions, where maximal concentrations of between 8–10 ppbv occur. Comparing the corresponding seasonal distribution of [PAN] it can be seen that, when integrating along the transect, more reactive nitrogen is stored in the form of HNO₃ as a result of this enhanced production of HNO₃ in the lower troposphere. This increases the loss of nitrogen via wet deposition due to the higher wet scavenging efficiency. Moreover, this finding holds

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



for most chemical mechanisms that include explicit NO_x chemistry involving these two main nitrogen reservoir species.

The final nitrogen reservoir included in the chemical scheme is the ORGNTR species, which is predominantly formed by the reaction of NO with the XO2N alkyl-nitrate operator included in the modified CBM4 mechanism (Houweling et al., 1998). The main source of the XO2N operator is the oxidation of either isoprene or the lumped organic species, PAR, by OH. Here the additional emissions of BVOC in the LATH run increase the annual net production of XO2N by $\sim 60\%$ in the tropics, resulting in an annual increase in [ORGNTR] by $\sim 20\%$. The atmospheric lifetime of ORGNTR is comparable to that of PAN allowing it to be transported outside of the main source regions, where increases of $\sim 10\%$ occur over the tropical Atlantic Ocean.

For the NOSOIL and NOBIO sensitivity studies we briefly discuss the corresponding differences as compared to the LATH run. Figure 7 shows the effect of removing the biogenic emission of NO from soils in Africa on tropospheric ozone for the 2-D-cross section (corresponding with Fig. 4). For the Sahel, Guinea and southern Africa regions, which exhibit the highest biogenic NO emissions (see Fig. 1), $\sim 5\text{--}45\%$ of the lower tropospheric ozone in the LATH run is formed as a direct result of NO which is emitted from soils. However, the latitude at which the largest seasonal difference occurs does not strictly follow the seasonal distribution shown in Fig. 1 due to the variation in the regional contribution to NO_x from biomass burning. The reduction in $[\text{O}_3]$ due to the absence of soil NO emissions severely depletes [OH] by upto $\sim 60\%$ at the surface, which significantly reduces the oxidizing capacity of the tropical troposphere (see Sect. 5). Again, the maxima in the seasonal differences in [OH] occur at similar locations as the maxima in the seasonal differences in $[\text{O}_3]$. For [PAN] and $[\text{HNO}_3]$ there are also significant reductions, with differences of up to $\sim 70\%$ for both across the entire latitude range (not shown). This subsequently dampens the amount of reactive nitrogen reaching the UT thus decreasing the in situ formation of O_3 in the UT by $\sim 5\text{--}7\%$. A negative feedback mechanism exists in the sense that the reduction in [PAN] is in part due to the suppressed isoprene oxidation due to the lower [OH]. This provides

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



a marginal offset to the reduction in $[O_3]$ due to lower isoprene by reducing the fraction of reactive nitrogen partitioned in PAN.

In Figs. 8 and 9 we investigate the differences introduced by the NOSOIL run concerning the long-range transport of tropospheric O_3 for the tropical regions surrounding Africa. For this purpose we show longitudinal cross-sections of averages between 0–20° N and 0–20° S for the months of June and December. These are the months when the soil NO_x emissions are the most significant in Guinea and southern Africa, respectively (see Fig. 1). The percentage differences shown for the NH tropics (June) and SH tropics (December) reveal that tropospheric O_3 is reduced by as much as ~15–35% over a large area above Africa, which supports the conclusion drawn from analysing the 2-D transect. This subsequently results in less O_3 being transported out across the ocean reducing the TTOC in the marine boundary layer by between 5–10% above the Atlantic Ocean. This strong influence has the potential to improve on previous comparisons of the total tropical ozone column between the TM model and the TTOC retrieved from satellites (e.g. Valks et al., 2003). There are also decreases in the in situ production efficiency, with ~10–15% less [PAN] and [ORGNTR] being transported out of the region, thus reducing the availability of $[NO_x]$ in the free troposphere (not shown). For $[HNO_3]$ there are large reductions of between 30–60% (not shown) meaning that soil NO_x emissions from Africa are a dominating source term for this reservoir species via the transport of important pre-cursors.

For the NOBIO sensitivity studies the effect on all three nitrogen reservoirs is minimal for the chosen latitudinal transect (not shown), as well as for tropospheric O_3 where differences of $\pm 1\%$ occur. However, the additional BVOC loading in the LATH run results in a reduction in $[OH]$ of ~5% throughout the troposphere, resulting in similar differences in $[HNO_3]$ (although this is dependent on the season). The reduction in the biogenic emission of ALD2 and PAR causes a reduction in both PAN and ORGNTR due to the perturbations introduced for the radical pre-cursors of both species. The westerly transport of BVOC's out of the region results in large decreases in the VOC concentrations in troposphere over the tropical Atlantic Ocean (~20–50% ALD2 and PAR, ~5%

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



OLE). Therefore, although the effects of biogenic VOC's on tropospheric O₃ formation are rather small compared to the influence of biogenic NO, a strong contribution is made to carbon loading in the Atlantic region as discussed in Sect. 5. It should also be noted that the emission of species such as CH₃OH are not included in this study.

5 Effect on the chemical budgets of tropospheric O₃ and nitrogen reservoirs

In this section we examine how the chemical budget for tropospheric O₃ differs between the sensitivity studies. Then we interpret the changes in the oxidation capacity of the atmosphere by examining the changes in the oxidation of both CH₄ and CO. In addition, we also quantify the differences in the partitioning of reactive nitrogen between the three most important reservoirs in the modified CBM4 mechanism discussed in Sect. 4. For this purpose we define the tropics as being the latitude band from 34° S–34° N in order to limit the influence from southern Europe and to include the southern coast of Africa. The budget analysis is calculated for all longitudes in order to account for the differences in the chemical processes which occur in regions affected by advection of trace species and chemical pre-cursors out of Africa as shown in Figs. 8 and 9, and that the POET inventory was applied globally in the BASE run. For the global budgets we adopt the methodology that is similar to that used in the multi-model intercomparison for large-scale 3-D CTM's performed by Stevenson et al. (2006). In our calculation we define the chemical production as the sum the reactions of NO+HO₂, NO+CH₃O₂ and NO+XO₂ (a lumped peroxy species) which are the main reactions which convert NO into NO₂. For the chemical destruction we account for the loss of O₃ by photolysis, OH, HO₂, ethene, olefins and isoprene. The stratosphere-troposphere component is calculated from the difference of the production and destruction terms whilst accounting for the dry deposition component. For the effects on the oxidative capacity of the troposphere given in Table 5 the procedure is identical to that used in Shindell et al. (2006).

Table 3 lists the chemical production and destruction terms for tropospheric O₃ in the

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



tropical troposphere integrated over the entire year. The corresponding changes with respect to the global burden and tropospheric lifetime of O_3 (τ_{O_3}) are given in Table 4. Comparing the chemical terms for production and destruction in Table 3 for both the BASE and LATH simulations shows that the BASE simulation is more chemically active which results in a higher tropical O_3 burden at the end of a year, although τ_{O_3} remains relatively unchanged. This increase in the production of O_3 in the BASE simulation is due to a decrease in the fraction of reactive nitrogen sequestered into longer-lived reservoir species when compared to the LATH simulation, as shown in Sect. 4. This results in a $\sim 5\%$ increase in both the wet and dry deposition terms of HNO_3 for the tropics in the LATH simulation, where the annual biogenic NO_x emission is approximately equal between both emission inventories (see Table 1).

In the NOSOIL simulation there is a $\sim 4.5\%$ reduction in the production of O_3 in the tropics, which subsequently reduces the O_3 burden in the tropics by $\sim 2\%$ (or $\sim 3\text{ Tg } O_3$). This translates into a $\sim 3.5\%$ reduction in the global production of O_3 , corresponding to a $\sim 1.6\%$ reduction in the annually integrated total global burden, due to the relative importance of the tropics towards global O_3 chemistry. This is smaller than the $\sim 5\%$ contribution found in the study of Aghedo et al. (2007) for a multi-annual simulation conducted between 1997–2002 using the ECHAM5-MOZ model adopting different emission inventories. In the NOBIO simulation there are only small differences in tropospheric O_3 introduced when compared to the LATH simulation, thus the direct emissions of C1-C3 organic compounds such as CO and HCHO from Africa do not seem to contribute much to the regional O_3 budget. However, it is most likely that the total biogenic carbon flux for C1-C3 species should be higher to account for the large number of species missing from the emission inventories (see discussion concerning CH_3OH below).

The two most dominant sink reactions for OH in the troposphere involve the oxidation of CH_4 and CO, which are both abundant trace gas species with relatively long lifetimes. Moreover, in the troposphere their depletion is almost exclusively limited to the reaction with OH, meaning that they act as a valid diagnostic for the integrated

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



changes associated with the HO_x budget and the rate at which they are depleted can be used to detect changes in the oxidative capacity of the global troposphere. Comparing the total global destruction of CH_4 between the BASE and LATH simulations shows that $\sim 2\%$ less CH_4 is oxidized during 2006 in the LATH run as a result of suppressed $[\text{OH}]$, which translates as an increase in the tropospheric lifetime of CH_4 (τ_{CH_4}) by 0.15 years ($\sim 1.5\%$). For CO the corresponding increase in the tropospheric lifetime (τ_{CO}) is a few days ($\sim 4\%$). For the NOSOIL simulation there is a further decrease in the rate at which CH_4 and CO are oxidized compared to the LATH simulation, which is in line with the decreases in tropospheric O_3 discussed in Sect. 4. Again, this introduces increases in the tropospheric lifetimes of CH_4 and CO due to the perturbation in the HO_x budget. For CO the fraction which can be transported away from the main sources regions increases as a result of the further increase in τ_{CO} , where $\sim 50\%$ of CO released globally from biomass burning originates in Africa (Jain, 2007). Previous studies have shown that CO from Africa can be transported significant distances throughout the SH (e.g. Edwards et al., 2006). To quantify the influence of soil NO_x on the transport of CO out of Africa we utilise another feature of the CTM experiments, which was the inclusion of a set of tagged tracers for CO. For each of the four regions defined in Sect. 3 we included a chemically active tracer whose concentration was set equal to the cumulative emission of CO from biogenic, anthropogenic and biomass burning sources. Comparing the tropospheric burdens for the tagged CO tracers between the LATH and NOSOIL simulations shows that the global burden increases by between 6–10%. Therefore, for years where there is little rainfall, and thus suppressed soil NO_x , and a corresponding increase in the incidence of wildfires, the CO which is released biomass burning activity from Africa has a significantly longer lifetime.

For the NOBIO simulation there is only a marginal increase in the chemical reactivity when compared to the LATH simulation, although BVOC's from Africa are important for the organic carbon loading of the tropical Atlantic. Comparing the tagged tracer global burdens for CO shows that there are reductions of $\sim 15\%$ as a result of removing the biogenic emission of CO. This leads to a reduction of $\sim 4.4 \text{ Tg CO}$ in the global

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



troposphere, which is $\sim 1.5\%$ of the total global burden for CO in the LATH simulation. Moreover, this reduces τ_{CH_4} by $\sim 1.2\%$ compared to the LATH simulation. Two additional emission fluxes are provided in the emission inventory of Lathi  re et al. (2006) which were not included in this experiment, these being for HCOOH and CH₃OH. Both HCOOH and CH₃OH are fairly soluble and thus undergo some loss by wet deposition, although for CH₃OH the global flux is estimated to be between 123–343 CH₃OH Tg yr^{−1} (Millet et al., 2008), with a large fraction coming from biogenic and oceanic sources. If included in the modified CBM4 mechanism CH₃OH would be oxidized by OH and produce additional HCHO, with a net conversion of OH into HO₂ (Tie et al., 2003). As a result CH₃OH is thought to be a significant global source of both HCHO (Millet et al., 2006) and, ultimately, CO (Duncan et al., 2007), which will have feedbacks via the additional scavenging of OH. For a more information regarding the global effects of this biogenic emission the reader is referred to the study by Tie et al. (2003).

Finally, Table 6 lists both the global and tropical burdens for the nitrogen and carbon budgets, in order to assess the importance of the African continent on the global troposphere. Comparing the BASE and LATH simulations shows that, for the LATH simulation, the repartitioning of nitrogen results in a higher fraction being stored in the longer lived reservoir species PAN and ORGNTR, which increases in the amount of nitrogen sequestered by $\sim 3\%$ for the global troposphere. This goes some way to explaining the reduction in the global production of O₃ shown in Table 3. For isoprene, there is also a $\sim 3\%$ reduction in the global burden in the LATH simulation, which again has implications for the night-time destruction of O₃ and the production of (e.g.) ALD2 and OLE. This also compensates for the missing biogenic HCHO in the BASE simulation. For the organic carbon flux there is an increase of $\sim 44\%$ in C-C bonds (PAR) the LATH simulation as a direct result of the increase in the global biogenic carbon flux. Moreover, the global burden of ALD2 also increases $\sim 6\%$, which subsequently increases the fraction of nitrogen stored as PAN as a result of an increase in the precursors.

Analysing the corresponding changes for the NOSOIL simulation shows the largest

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



changes are for PAN and ORGNTR, with the global burden of HNO_3 remaining relatively constant. This is a result of the reduction in $[\text{OH}]$ which reduces the oxidation of isoprene by $\sim 8\%$ ($\sim 10\%$ in the tropics), and PAR by $\sim 2\%$ ($\sim 5\%$ in the tropics). Both of these are pre-cursors of ALD2, which causes a reduction in the fraction of nitrogen stored as PAN and ORGNTR. This acts as a small compensating effect for the lack of NO from soils with respect to the availability of reactive nitrogen. For the NOBIO simulation a comparison of the global burdens shows that emissions of BVOC's from Africa account for $\sim 1.5\%$ of ALD2, $\sim 2.5\%$ of OLE and $\sim 5\%$ of PAR in the global troposphere ($\sim 10\%$, $\sim 4\%$ and $\sim 14\%$ in the tropics, respectively). This is a direct effect rather than from oxidation products of other species as shown by the equal burdens in isoprene. This also has consequences for the NO_x budget as shown by the $\sim 2.5\%$ increase in the global burden of ORGNTR, although the influence on tropospheric O_3 is marginal as shown above.

6 Comparisons with observations

In this section we assess how the TM4 model performs for the African Continent by comparing co-located model output with a host of different measurement data. Given that the differences between the NOBIO run and LATH run were found to be rather small in terms of differences in the tropospheric O_3 burden we omit this sensitivity test from the following comparisons for brevity and in order to focus the discussion on the most important differences.

Figure 10 shows comparisons of tropospheric $[\text{O}_3]$ against the MOZAIC data sampled during the take-off and landing of passenger aircraft from Windhoek in Namibia (22.5°S , 17.5°E) for all seasons during 2006. The sampling frequency is approximately once every two days resulting in a statistically robust dataset. Moreover, profiles are taken during both daytime and nighttime therefore capturing the diurnal variation that occurs for tropospheric O_3 . In general, although the agreement is rather good for both the BASE and LATH simulations, the LATH simulation does result in a moderately bet-

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



ter agreement for the mid- to UT using the current experimental setup, although this is dependent on the particular season (compare JJA vs. SON). The largest deviation from the observed average occurs during the SON season, where all simulations underestimate values by $\sim 10\text{--}15\%$ in the mid- to UT. In order to investigate this further an additional sensitivity test was performed where a daily cycle was imposed on the daily emission of trace species from biomass burning, as observed using satellites (e.g. Boersma et al., 2008), and the injection heights were increased to 2 km in the tropics following observations by Labonne et al. (2007). However, this only resulted in a relatively insignificant improvement in the agreement for season SON, suggesting that this is not the principal cause.

At the surface the model values tend to be over-estimated by $\sim 50\%$ throughout the entire year, where the stratospheric contribution is shown by means of a chemically active O_3 tracer for stratospheric O_3 similar to that described in Lelieveld and Dentener (2000). For JJA this accounts for $\sim 20\%$ of the tropospheric O_3 profile measured at Windhoek as shown in Fig. 10c, although this does not impact on the quality of the comparison at the surface as seen by comparing the other seasons. For the NISOIL run (shown as the black line) the profile tends to underestimate the values of the seasonal average obtained from the MOZAIC measurements throughout the year, especially for season DJF where the emission of NO from soil are maximal in this region of Africa (see Fig. 1). For this season soil NO_x accounts for between $\sim 10\text{--}15\%$ of the TTOC over Africa near the airport (see Sect. 4).

Figure 11 shows a similar comparison in the UT against the MOZAIC dataset, this time exploiting the measurements taken at the cruise altitude made by the passenger aircraft traveling to and from Frankfurt, Germany. Here we choose to show comparisons for March, June, September and December 2006 to highlight the monthly variability in both the measurements and the simulations. The contributions due to stratospheric intrusions are again shown using the chemically active O_3 tracer in order to assess whether the in situ formation of O_3 in the troposphere is the dominating term. The observations show the meridional gradient which has been previously identified by

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



both Sauvage et al. (2007) and Maunois et al. (2008), who have derived a gradient in the UT $[O_3]$ of 1.13 ± 0.17 ppbv/deg latitude and 0.94 ± 0.18 ppbv/deg latitude, respectively. The extent of the agreement with the measurements varies between the different simulations, although the largest deviation is again that for the NOSOIL run indicating that biogenic NO_x is important for $[O_3]$ at altitudes well above the boundary layer. This agrees with the conclusions of a similar sensitivity study performed by Maunois et al. (2008), who used a mesoscale model for July 2006 along with the MOZAIC data over Africa, although here we extend the domain of the comparison to 20° S and show various months during the course of the year. The contribution from STE is quite marked in the extra-tropics and generally results in a good agreement at latitudes $>10^\circ$ N. However, although the vertical resolution of TM4 is defined by the vertical levels available in the ECMWF meteorological fields in the tropical tropopause region it still maybe insufficient to capture all intrusions from the stratosphere. Moreover, the climatological boundary conditions which we adopt (Fortuin and Kelder, 1998) could also impose their own signature. None of the simulations can capture the increases in UT O_3 observed directly over regions where intense biomass burning activity occurs (e.g. $0-20^\circ$ N in March and $0-20^\circ$ S in September). It is interesting to see that for $0-10^\circ$ N (June) and $10-20^\circ$ S (December) there is an appreciable difference between the BASE and LATH profiles. Considering that the biomass burning inventories are identical in both simulations, this must be related to differences in the biogenic NO_x inventories. This shows either that the biogenic NO source strength maybe too high in the LATH inventory for December in southern Africa during 2006 or that the convective transport from the boundary layer is too efficient.

In order to assess the performance of the parameterization for convective transport directly below the flight transect we include some passive tracers in the simulations. Figure 12 shows the corresponding distribution of the four convective tracers for the corresponding months shown in Fig. 11. Here the concentrations are fixed at 100 pptv below 850 hPa and each tracer given a fixed atmospheric lifetime of 20 days. The area over which each tracer is defined is equal to that used for the segregated emission

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



totals shown in Fig. 1. At the cruise altitudes concentrations are <10 pptv for all of the months shown, indicating that the air is aged by several days before it reaches the UT. This allows time for the photochemical production of O_3 to occur at lower altitudes, which is then uplifted to the flight altitude. In general, the strength of the convective transport is stronger from southern Africa and Guinea than the other regions. For these two regions the maximal emission of pre-cursors from biomass burning occurs between July–September and December–January, respectively, where the magnitude of the annually integrated emission fluxes for Guinea are twice those for southern Africa (not shown). Bian et al. (2007) have shown that the variability between different biomass burning emission datasets is larger for southern Africa than for northern Africa as a result of the differences in the intensities between fires in each region. From Fig. 11 it can be seen that for March, June and September the model under predicts the MOZAIC measurements for certain latitudes. When accounting for the regional differences in the release of soil NO and the seasonality in the biomass burning activity, this suggests that the strength of the convective activity in the model from southern Africa is too weak during June and September, whilst being too strong in December. Moreover, possible deficiencies in the quality of the emissions for southern Africa could also play a role. Finally, the agreement between the model and MOZAIC measurements between 0 – 10° N is rather good during December but gets worse for March, where the contribution to the biomass burning emission from all regions become similar, except for Sahara (not shown). Considering that the contribution from southern Africa increases during March (see Fig. 12) implies that the emission flux in southern Africa is too low.

Figure 13 shows a comparison of co-located ozone profiles for seasons DJF and JJA with measurements from both the Shadoz network (Thompson et al., 2003) and those specifically measured during the AMMA project (Thouret et al., 2009). Here only model profiles for the specific measurement dates are averaged to ensure that a consistent comparison is performed. The quality of the comparison at each site is somewhat dependent on the season as would be expected considering the seasonality of the regional emission sources. The differences between the BASE and LATH

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



simulations appear to be quite small in spite of the variations in the seasonality of the emissions, although at Cotonou (6.6° N, 2.2° E) there is a noticeable difference in the mid-troposphere during season DJF. This lack of difference between emission inventories is surprising considering the comparisons shown in Fig. 4 and suggests that the temporal differences near the sampling sites are not so large. The worst agreement occurs for Cotonou as a result of exceptional values of [O₃] which were measured around 700–800 hPa at the site during December and which were attributed to an unknown local source (Minga et al., 2009). Although there are instances where TM4 performs rather well (e.g. Irene during JJA) the model does not have the ability to capture the “peaks” in tropospheric O₃ that occur between 700–800 hPa (e.g. Cotonou during DJF). These lower-tropospheric maxima have been ascribed to air masses transported into the region by (e.g.) the Harmattan and African Easterly Jet (AEJ), and thus have different chemical histories (Sauvage et al., 2005). The quality of the comparison shows that under the current configuration the model has difficulty in being able to capture the vertical fine structure observed in the sondes, where Sauvage et al. (2005) have shown that, in principle, the ECMWF meteorological fields can capture such events. For the UT, the differences are relatively large, which is not consistent with the comparisons made against the MOZAIC measurements shown in Fig. 10. This suggests that the low sampling frequency for each season, and the large variability that occurs between such samples (as shown by the 1- σ error bars) hinders the quality of the comparisons. Moreover, the use of homogenous monthly fluxes for pre-cursor emissions from all sources also introduces inaccuracies concerning the variability observed in the model. Some improvement could possibly be made to the simulations by adopting the 8-day biomass burning emission files available from the GFEDv2 inventory.

Again the NOSOIL run exhibits the worst overall agreement of all simulations for all sites, although in some circumstances, when the regional soil NO emissions are low, the differences between model profiles can be rather small (e.g. Nairobi during DJF). This supports the conclusion that the effect of soil NO_x on tropospheric O₃ formation is not strictly limited to the boundary layer when integrated over time. Moreover, this also

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



shows that a global CTM can capture the same features as those simulated by meso-scale models (Saunois et al., 2008; Delon et al., 2008), whose run times are restricted to a number of days. When comparing results from the additional sensitivity test with the daily cycle imposed on the biomass burning emissions and increased injection heights the resulting model profiles exhibited a few ppbv less in both the mid- and upper troposphere, thus degrading the quality of the comparisons. Another possible cause for the disagreement is that the convective parameterization does not provide enough convective mixing of pre-cursors and/or O₃ out of the boundary layer, as shown for the MOZAIC in-flight measurements, although regional emission sources are likely to have much more influence for the sonde measurements in the lower levels.

Figure 14 shows a comparison of interpolated model output against a composite of the tropospheric O₃ measurements taken along the flight tracks of the French and German Falcon and the British BAe146, all of which participated in the AMMA measurement campaign during July and August 2006. With the exception of the M55-Geophysica measurements, the observations are identical to those shown in Janicot et al. (2008). In the lower troposphere the observations show that [O₃] typically varies between 30–40 ppbv, although there are instances where the variation is much greater. In the upper troposphere there is a wide variation of values ranging between 30–80 ppbv, although the median is around 50–55 ppbv. Some extreme events are also captured around 5° N in the mid-troposphere where [O₃] reaches between 80–100 ppbv (red/black points in Fig. 13a). In general all of the simulations fail to emulate the correct variability shown in the observations. Moreover, there are values of ~50 ppbv in the lower troposphere around 12° N in all simulations. The similarity between the LATH and NOSOIL simulations shows that the cause of these high values is not the peak in NO emissions from soils between 0–10° N (see Fig. 1) but possibly transport of O₃ from other regions. Conversely, the highest values which occur in the lower troposphere around 5° N are also not captured by the model in any of the simulations.

For the upper troposphere it can be seen that the BASE simulation has moderately higher [O₃] but still underestimates the high values observed up the column and at

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



around ~ 250 hPa. For the NOSOIL simulation, some of the low $[\text{O}_3]$ which occurs around 15°N are also seen in the measurements indicating that some variability in soil NO emissions must occur over the measurement period. This is most probably due to variations in the amount of soil NO_x released throughout the month as observed by Stewart et al. (2008), which is not accounted for in the model as a result of using a fixed emission flux for the entire month from the climatology. Short periods of enhanced NO emissions occur after periods of intense rainfall and the representation of this in any model is important for capturing such events (Adon et al., 2009; Delon et al., 2009) and could also explain why TM4 fails to capture the higher $[\text{O}_3]$ events. Therefore, it seems necessary to attempt to incorporate this into any future inventory biogenic emission inventory. Considering that the inter-annual variability in rainfall is relatively high during the WAM (e.g. Long et al., 2000), it maybe worthwhile to try and segregate biogenic emissions into a weekly frequency to try and improve on the current climatology.

Figure 15a–c shows comparisons of the monthly means for surface O_3 , NO_2 and HNO_3 from the model simulations with the corresponding monthly means from measurements taken at a selection of measurement sites from the IDAF network (IGAC/DEBTS/Africa International Global Atmospheric Chemistry/Deposition of Biogeochemically Important Trace Species/Africa), respectively. Further details on the IDAF measurement program can be found at <http://obs-mip.fr/idaf>. These sites are principally located between 0 – 15°N and $\pm 10^\circ$ longitude and are, therefore, representative of the great ecosystems of Western Equatorial Africa (Martins et al., 2007; Galy-Lacaux et al., 2009). For some months, no measurements are available at certain sites (which are represented as a zero value in the Figures). In general the surface O_3 measured across all sites ranges between 10 – 30 ppbv, where higher values are typically occur during the summer months. When comparing both the BASE and LATH simulations the model consistently over predicts surface $[\text{O}_3]$ by between ~ 50 – 100% , although the seasonal cycle is captured to some degree. The BASE simulation typically shows higher values than the LATH simulation, which agrees with the comparisons shown in Figs. 10–12. Similar plots to those shown in Sect. 4 for a chemically

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



active tracer included to assess the influence of stratospheric O_3 indicates that there is a $\sim 2\text{--}5$ ppbv contribution at the surface near the measurement sites (not shown), which accounts for $\sim 10\text{--}15\%$ of the surface values. The worst agreement is shown for Zoétélé (3.1°N , 11.6°E), which is situated in a tropical forest. At this location a contributing factor to the low values in the measurements is that tropical forests are considered to be a substantial sink as a result of all the different strata of vegetation under the canopy contributing to both the deposition and chemical destruction (Cros et al., 1992). Such detail is obviously not included in a global model, where the disagreement is compounded by the fact that the area contained in a typical grid-cell in the model covers a range of different vegetation and land types. In the NOSOIL run at some measurement sites there is a clear decrease in $[O_3]$ of up to $\sim 50\%$ during certain months. This is a cumulative effect of a reduction in the local production and that transported over the measurement site from other regions (cf. Figs. 8 and 9). However, in many instances removing the soil NO_x has little effect on $[O_3]$ (e.g. Zoétélé for all seasons). Considering that tropospheric ozone has a typical lifetime of ~ 23 days in these runs (see Sect. 6), the majority must be formed from other sources, notably biomass burning, as found in Adon et al. (2009) (see below).

For $[NO_2]$, two maxima are typically visible during the year for the dry savannah, as determined from an analysis of the decadel dataset measured at both Banizoumbou and Katibougou (10.5°N , 7.3°W) by Adon et al. (2009). The first occurs during May–June at the beginning of the rainy season, which is attributed to soil NO_x emissions, and the second related to a biomass burning source. For the wet savanna and forested sites (i.e. Djougou and Zoétélé) the seasonal cycle is less marked thus a regional biomass burning source is thought to be more important. When comparing the model monthly means with the measurements it can be seen that the model exhibits higher $[NO_2]$ during winter for both the BASE and LATH runs, whereas for the summer the agreement is much better. Moreover, the temporal differences between both emission inventories become more obvious, where the distribution of NO_x in the POET inventory results in more local emissions near the measurement sites. This

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



generally results in an over-prediction compared to the LATH inventory, indicating that both the timing and temporal distribution is better in the LATH run (see comparison for Katibougou, Mali in Fig. 15b). The NOSOIL run shows that the seasonal cycle at Banizoumbou is almost entirely due to soil NO_x , whereas at Katibougou there is a greater contribution due to biomass burning. That said, the emission of NO from soils appears to be too high at Banizoumbou (as shown by the comparison of the LATH run during winter). At Katibougou the model over-predicts $[\text{NO}_2]$, for the earlier months, whereas the agreement is good for April–May and the end of the year. At this site the biomass burning contribution helps to capture the correct seasonal variability.

For HNO_3 the detection limit of the measurement technique is ~ 0.1 ppbv, thus for many of the months the $[\text{HNO}_3]$ measured is at, or below, this threshold. The model generally over predicts the formation and/or the lifetime of HNO_3 during winter, whilst under predicting during the summer. Therefore some degree of a signature is present from the $[\text{NO}_2]$ comparison. For summer, the conversion of reactive NO_x into HNO_3 is not optimal in spite of the high $[\text{O}_3]$ (and thus $[\text{OH}]$), which should enhance the formation rate of HNO_3 . Unfortunately no corresponding data is available for either PAN or organic nitrates, which would be instructive in terms of determining whether the concentrations of these species are also under predicted (i.e.) whether there is too little nitrogen in the system as a result of the low NO_x emissions for this season. Moreover, this would also allow an assessment of whether the cause is either a deficiency associated with the chemical mechanism (such as the recently proposed production of HNO_3 involving NO and HO_2 , Butkovskaya et al., 2007), an over estimation of the deposition flux, a missing source term or whether the emission of NO_x is too high for this year from either biomass burning or biogenic sources. For the NOSOIL run the corresponding comparisons show that this source accounts for $\sim 50\%$ of the local production during winter and over 75% during the summer, as would be expected considering the $[\text{NO}_2]$ comparisons.

The influence of biogenic emissions from Africa

J. E. Williams et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

7 Conclusions and outlook

In this study we have applied two recently developed inventories for biogenic emissions (provided as part of the POET and RETRO databases) in a state-of-the-art CTM for the purpose of investigating the effect on the composition of the tropical troposphere over Africa. We have found that the seasonality and temporal distribution of isoprene, biogenic volatile organic carbon and NO emissions are quite different, which introduces differences with respect to both regional and global $[O_3]$, the fraction of reactive nitrogen sequestered into reservoir compounds and the total carbon loading in the tropical troposphere. In terms of the chemical budget, the POET biogenic emission inventory results in a more chemically active troposphere, where the atmospheric lifetimes of CH_4 and CO decrease by $\sim 1.5\%$ and $\sim 4\%$, respectively. For the Lathi  re et al. (2006) inventory, there is an increase in the total nitrogen lost by wet deposition of $\sim 5\%$ which contributes to a lower tropospheric ozone burden. This also introduces differences in the tropical tropospheric ozone column that is calculated between both inventories, which has the potential to improve comparisons between CTM's and satellite instruments (e.g. Valks et al., 2003). For the biogenic volatile organics, there is a $\sim 3\%$ reduction in the global isoprene burden in the inventory of Lathi  re et al. (2006), along with increases in alkanes, aldehydes and olefins of between $\sim 1.5\text{--}5\%$ as a result of the additional direct emissions. When comparing simulations against a suite of in situ measurements the inventory of Lathi  re et al. (2006) results in a better agreement in the majority of cases.

Sensitivity studies have shown that the emission of NO from soils in Africa are responsible for between $\sim 5\text{--}45\%$ of tropospheric $[O_3]$ over equatorial Africa. This influences the oxidative capacity of the African troposphere in the region, which has implications regarding the export of trace species via the perturbations to the atmospheric lifetimes of abundant trace species. Therefore, CTM studies which focus on quantifying the long-range transport of trace gases should ensure that they adopt recent biogenic emission inventories to account for changes in local climate and/or land-use. For bio-

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



genic volatile organic compounds the effects on tropospheric $[O_3]$ seem to be rather minimal. The use of tagged tracers shows that biogenic CO emissions are responsible for ~15% of the global burden of CO of the direct emissions in Africa. Moreover, analyzing the chemical budget shows that the biogenic emissions from the African Continent are responsible for ~1.5% of ALD2, ~2.5% of OLE and ~5% of PAR in the global troposphere. This has an influence on the oxidative capacity via the scavenging of OH, especially near the source regions.

Although the climatology for biogenic emissions provided by Lathi  re et al. (2006) improves the overall performance of TM4 in the tropics when compared with observations, there are still problems in that the variability seen by the measurements are often not captured by the simulations. This implies that more detail should be introduced regarding the temporal variability of such emissions when performing simulations for various time periods. For offline CTM modeling the incorporation of an online algorithm, such as that described in Ganzeveld and Lelieveld (2002), may not be feasible considering that constraints are often imposed on the complexity of any large-scale model in order to achieve realistic runtimes. Therefore, a step forward for offline modeling would be to extend the time period for which ORCHIDEE output is available in order to account for the further deforestation and desertification which has occurred in the last decade. Moreover, rather than assembling a climatology the emission fluxes could be supplied as monthly segregated emission estimates for each year similar to that provided for biomass burning emissions (e.g. Van der Werf et al., 2006). By using meteorological data such as precipitation rates the degree of inter-annual variability could be improved which would improve large-scale simulations.

For future climates the emission of NO from soils will most likely increase by ~20% by 2100 to $10.5 \text{ Tg N yr}^{-1}$ as suggested in Hauglustaine et al. (2005) as a result of the temperature increases and changes in vegetation which are predicted to occur in tropical regions. Associated changes will also occur for the BVOC's, where the total global carbon flux is predicted to double. This will most likely increase the weekly variability in tropospheric ozone production over Africa during the Wet Season where

The influence of biogenic emissions from Africa

J. E. Williams et al.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I ◀](#)[▶ I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

more intensive rainfall is imposed on a drier background environment introducing significant seasonal differences with respect to tropospheric ozone over Africa. Our studies also indicate that this will also affect the tropical marine environment over the Atlantic, where increases in the background VOC levels can be expected along with possible reductions in ozone thus increasing atmospheric lifetimes. The simultaneous increases in tropospheric water vapour may moderate this to some extent, although increased cloud cover would further dampen OH production. Our studies also indicate that the long-range transport of CO out of the region would also be enhanced which would have implications for back ground levels in the SH.

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J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

Table 1. Details concerning the chemical speciation of the organic biogenic emissions used in both the BASE and LATH simulations. The emission flux for CH₃OH is used to account for emissions of CH₃CH₂OH, where the flux is set at 10% of the CH₃OH emissions. For species with (+) the same emission inventory is used in both simulations.

Organic Species	BASE	LATH	Speciation in the modified CBM4 mechanism
CO ⁺	yes	yes	CO
HCHO	no	yes	HCHO
CH ₃ OH	yes	yes	0.1*PAR
C ₂ H ₄	yes	yes	ETH
C ₂ H ₆ ⁺	yes	yes	2*PAR
C ₃ H ₈ ⁺	yes	yes	3*PAR
C ₃ H ₆ ⁺	yes	yes	PAR+2*OLE
CH ₃ CHO	no	yes	ALD2
HCOOH	no	yes	N/A
CH ₃ COOH	no	yes	0.5*PAR
Acetone	yes	yes	3*PAR
Isoprene	yes	yes	ISOP
Monoterpenes	yes	yes	ISOP

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

Table 2. Emission totals for the contributions to NO_x, CO, isoprene and organic carbon from (in listed order) anthropogenic activity, biomass burning and biogenic activity for each latitudinal zone. For the biogenic component integrated totals are shown for both the LATH and BASE simulations, respectively, along with resulting percentage difference for each of the regions. For biogenic CO the same inventory is used in both runs. For the NOSOIL simulations emissions with (#) are removed and for the NOBIO simulation emissions with (*) are removed. Summing these segregated emission totals gives 6.91 (6.91) TgN yr⁻¹, 250.15 TgCO yr⁻¹, 112.07 (137.62) TgC yr⁻¹ (isoprene) and 20.34 (11.27) TgC yr⁻¹ (BVOC's), where the BASE sums are given in parenthesis except for CO.

Trace species	Sahara (20–40° N)	Sahel (10–20° N)	Guinea (0–10° N)	Southern Africa (40° S–0° N)
NO _x (TgN/yr)	1.12	1.13×10 ⁻¹	2.30×10 ⁻¹	6.96×10 ⁻¹
	2.81×10 ⁻³	2.40×10 ⁻¹	1.01	1.19
	2.01×10 ⁻¹ (#)	5.75×10 ⁻¹ (#)	7.64×10 ⁻¹ (#)	7.64×10 ⁻¹ (#)
	2.18×10 ⁻¹	6.12×10 ⁻¹	7.74×10 ⁻¹	7.02×10 ⁻¹
	(-7.5%)	(-6.3%)	(-1.3%)	(+8.0%)
CO (TgCO/yr)	13.83	14.90	23.10	18.46
	1.64×10 ⁻¹	13.77	65.37	70.43
	3.09	4.43	10.59	12.02
	(N/A)	(N/A)	(N/A)	(N/A)
	3.46	16.10	51.45	41.06
Isoprene (TgC/yr)	3.58	20.54	56.99	56.51
	(-3.4%)	(-27.6%)	(-9.7%)	(-27.3%)
Organic Carbon (TgC/yr)	2.48	1.07	1.97	1.40
	4.24×10 ⁻³	3.57×10 ⁻¹	1.87	7.83×10 ⁻¹
	5.05×10 ⁻¹ (*)	1.25(*)	4.84(*)	3.81(*)
	1.47×10 ⁻¹	1.46×10 ⁻¹	3.59×10 ⁻¹	6.86×10 ⁻¹
	(+343%)	(+858%)	(+1348%)	(+556%)

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

Table 3. The changes in the budgets and burdens for tropospheric ozone in the tropical region from the four simulations used in this study as integrated for 2006. The methodology is similar to that described in the multi-model intercomparison of Stevenson et al. (2006). The mean annual stratosphere-troposphere exchange equals $\sim 295 \pm 2 \text{ Tg O}_3 \text{ yr}^{-1}$.

	Chemical O ₃ production (Tg O ₃ yr ⁻¹)	Chemical O ₃ destruction (Tg O ₃ yr ⁻¹)	Dry Deposition (Tg O ₃ yr ⁻¹)	τ_{O_3} (days)	Tropo Burden (Tg)
BASE	3137	3050	380	21.1	176
LATH	3099	3011	382	21.0	173
NOSOIL	2961	2900	361	21.4	170
NOBIO	3102	3012	381	21.1	174

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

Table 4. The changes in the global budgets and burdens for tropospheric ozone from the four simulations used in this study as integrated for 2006. The methodology is taken from the multi-model intercomparison of Stevenson et al. (2006), where the troposphere is all grid cells with $[O_3] < 150$ ppbv. The annual stratosphere-troposphere exchange is equal to $\sim 493 \text{ Tg O}_3 \text{ yr}^{-1}$ for all simulations.

Simulation	Chemical O ₃ production (Tg O ₃ yr ⁻¹)	Chemical O ₃ destruction (Tg O ₃ yr ⁻¹)	Dry Deposition (Tg O ₃ yr ⁻¹)	τ_{O_3} (days)	Tropo Burden (Tg)
BASE	4330	4160	662	23.8	315
LATH	4294	4122	664	23.8	312
NOSOIL	4143	3394	642	24.2	307
NOBIO	4292	4121	663	23.8	312

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

Table 5. The integrated annual changes in the integrated global budgets, lifetimes and burdens for tropospheric CH₄ and CO from the four simulations used in this study. The methodology is taken from the multi-model intercomparison of Stevenson et al. (2006), where the troposphere is all grid cells with [O₃] < 150 ppbv.

Simulation	OH+CH ₄ destruction (Tg CH ₄ yr ⁻¹)	Tropospheric CH ₄ burden (Tg CH ₄)	τ_{CH_4} (Years)	Tropo CO burden (Tg CO)	τ_{CO} (Months)
BASE	474	4271	9.15	340	1.81
LATH	465	4273	9.31	346	1.88
NOSOIL	449	4274	9.61	354	1.96
NOBIO	472	4273	9.20	337	1.86

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

Table 6. The changes to the global and tropical tropospheric burdens for the three main reservoir species that contain reactive nitrogen oxides, isoprene and other BVOC's to which the additional emissions from the Lathi  re et al. (2006) inventory contribute. All budget terms are given in either TgN yr^{-1} or TgC yr^{-1} , with the tropical burdens being shown in Paranthesis.

	BASE	LATH	NOSOIL	NOBIO
HNO_3	0.660 (0.505)	0.657 (0.502)	0.656 (0.502)	0.657 (0.502)
PAN	0.191 (0.101)	0.203 (0.099)	0.199 (0.095)	0.201 (0.097)
ORGNTR	0.183 (0.134)	0.207 (0.147)	0.198 (0.139)	0.202 (0.143)
ISOP	0.431 (0.403)	0.418 (0.376)	0.455 (0.413)	0.418 (0.375)
HCHO	0.349 (0.279)	0.339 (0.263)	0.335 (0.260)	0.337 (0.262)
PAR	3.320 (0.970)	4.780 (1.719)	4.883 (1.803)	4.603 (1.568)
ALD2	0.327 (0.173)	0.349 (0.170)	0.341 (0.163)	0.344 (0.166)
OLE	0.313 (0.172)	0.332 (0.222)	0.344 (0.233)	0.323 (0.213)

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I ◀](#)[▶ I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

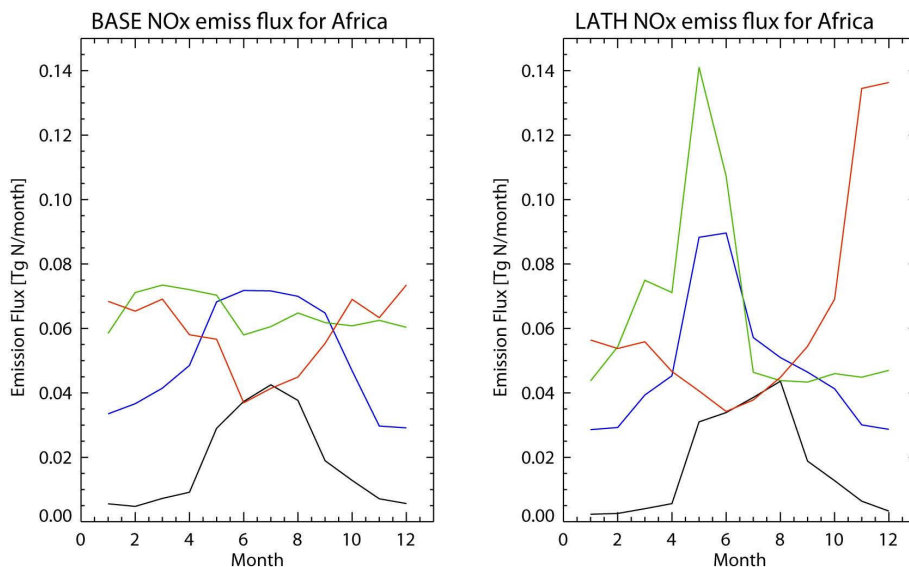


Fig. 1. A comparison of the monthly emission flux of NO from soils in TgN as provided in **(a)** Yienger and Levy (1995) and **(b)** Lathi  re et al. (2006). The key for the latitudinal regions is: (black) Saharan, (blue) Sahel, (green) Guinea and (red) southern Africa.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I ◀](#)[▶ I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

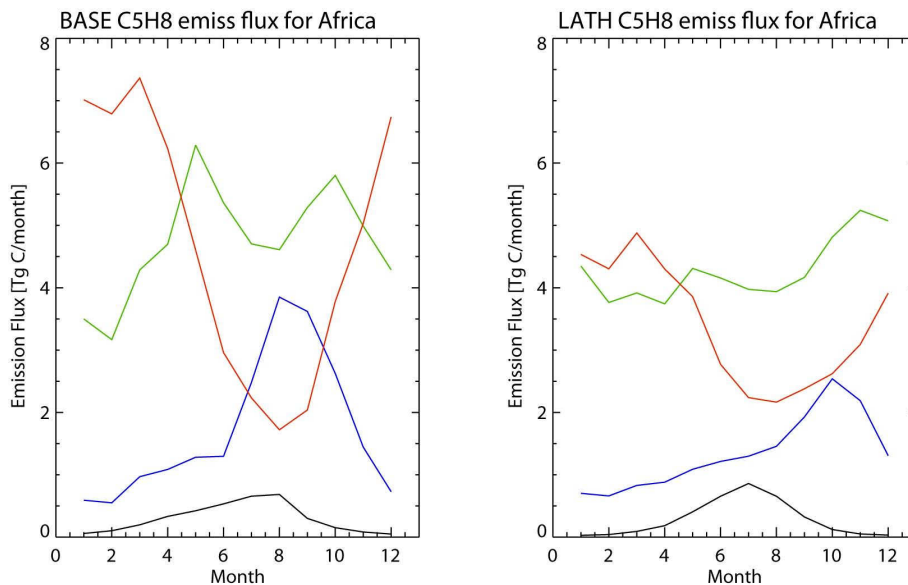


Fig. 2. A comparison of the monthly emission flux of Isoprene in TgC as provided in **(a)** the POET inventory and **(b)** Lathi  re et al. (2006). The key for the latitudinal regions is: (black) Saharan, (blue) Sahel, (green) Guinea and (red) southern Africa.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I ◀](#)[▶ I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

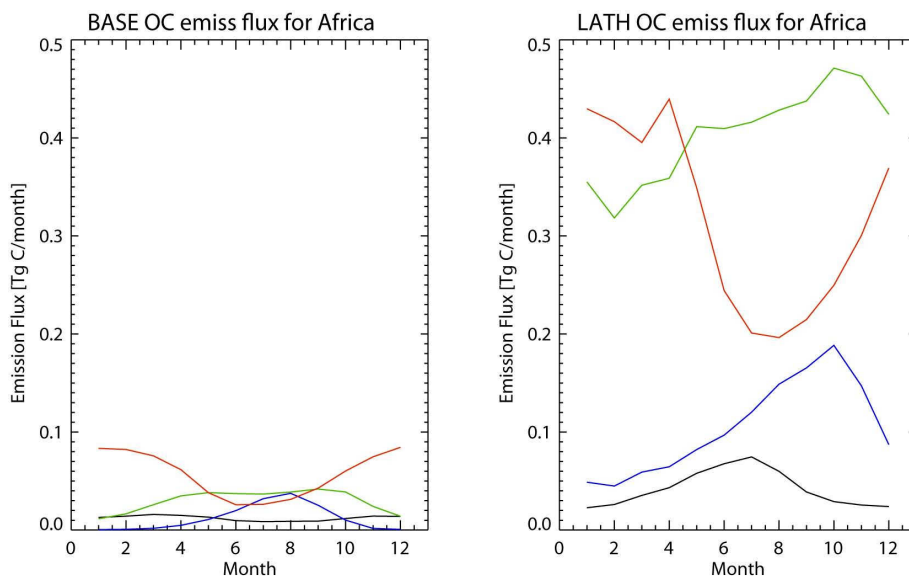


Fig. 3. A comparison of the monthly emission flux of organic carbon from vegetation in TgC as provided in **(a)** the POET inventory and **(b)** Lathière et al. (2006). The key for the latitudinal regions is: (black) Saharan, (blue) Sahel, (green) Guinea and (red) southern Africa.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

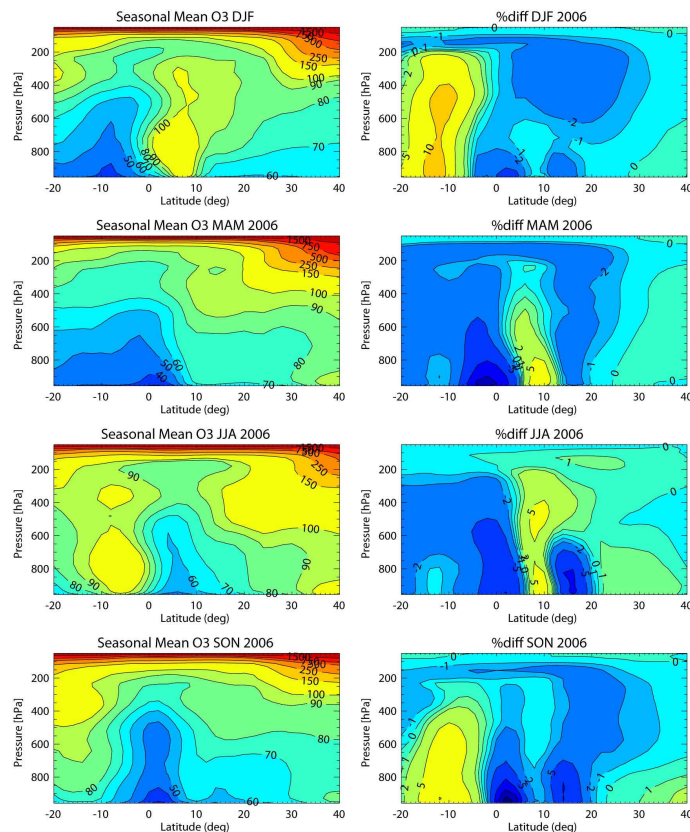


Fig. 4. A comparison of the seasonal means of tropospheric ozone in ppbv as averaged between 12–21° E for **(a)** DJF, **(b)** MAM, **(c)** JJA and **(d)** SON. The corresponding percentage change between the BASE and LATH simulations is shown for each season, where the difference shown is from $(\text{LATH-BASE})/\text{LATH} \times 100\%$.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

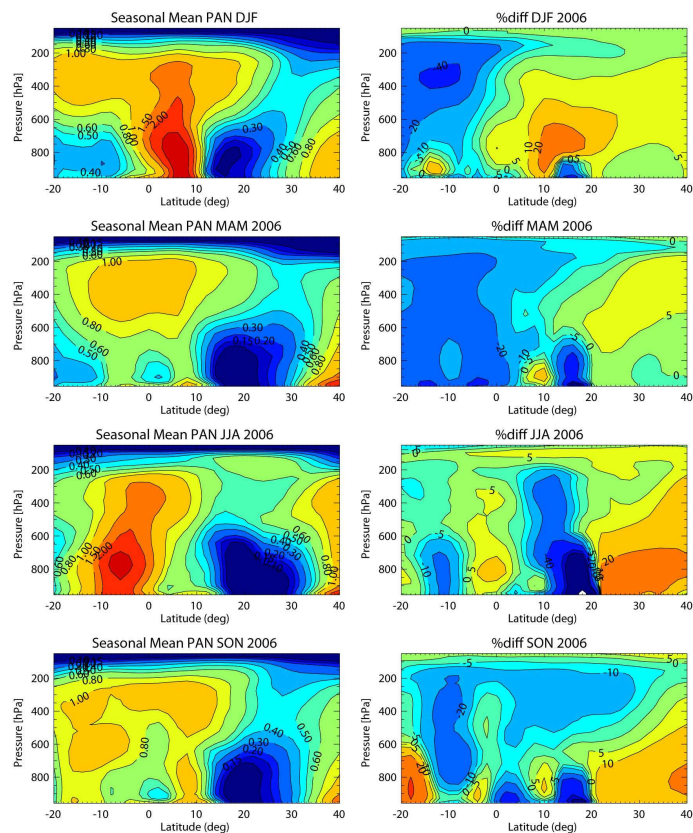


Fig. 5. As for Fig. 4 except for PAN.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

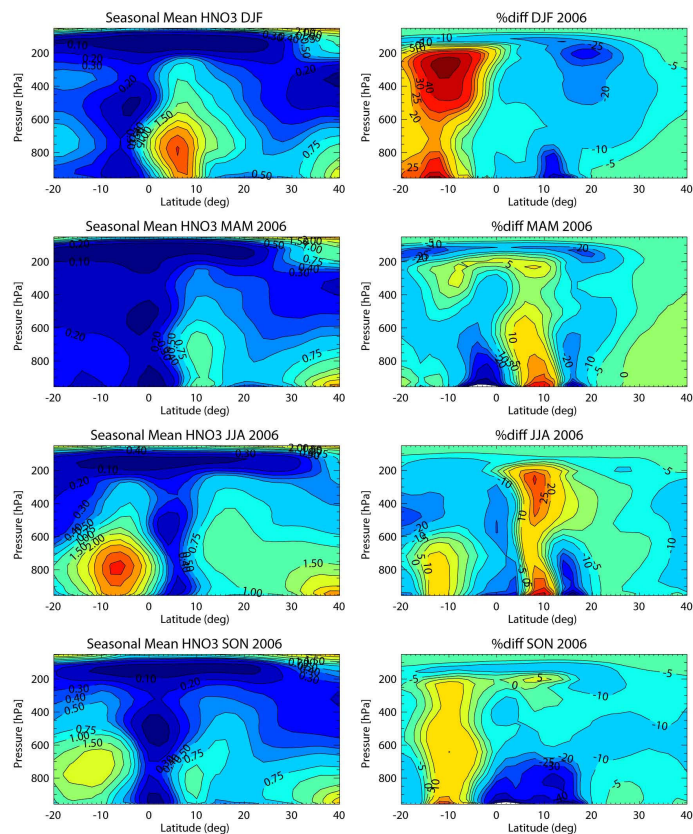


Fig. 6. As for Fig. 4 except for HNO₃.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

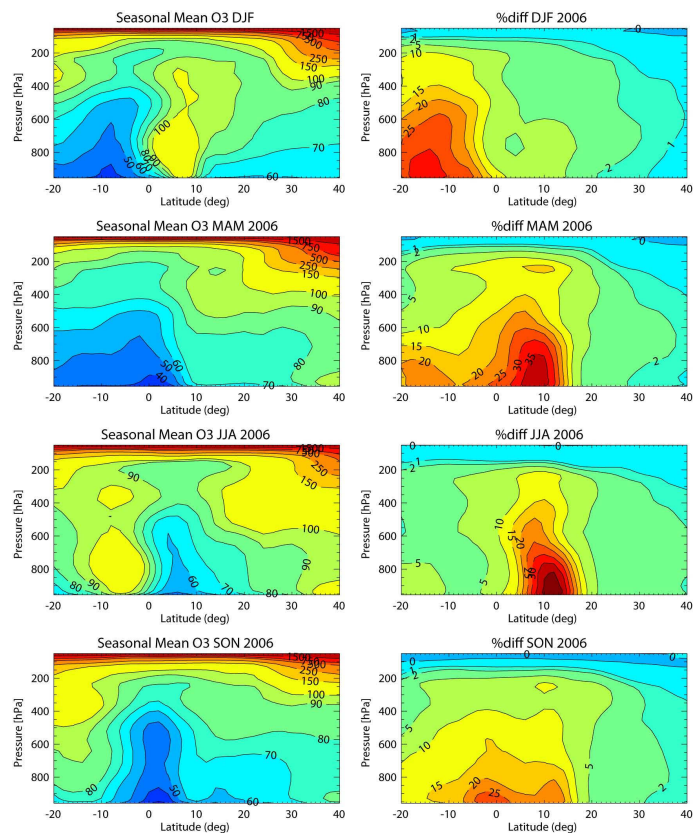


Fig. 7. As for Fig. 4 except for the sensitivity test NOSOIL that excludes emissions of NO from soils in Africa.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

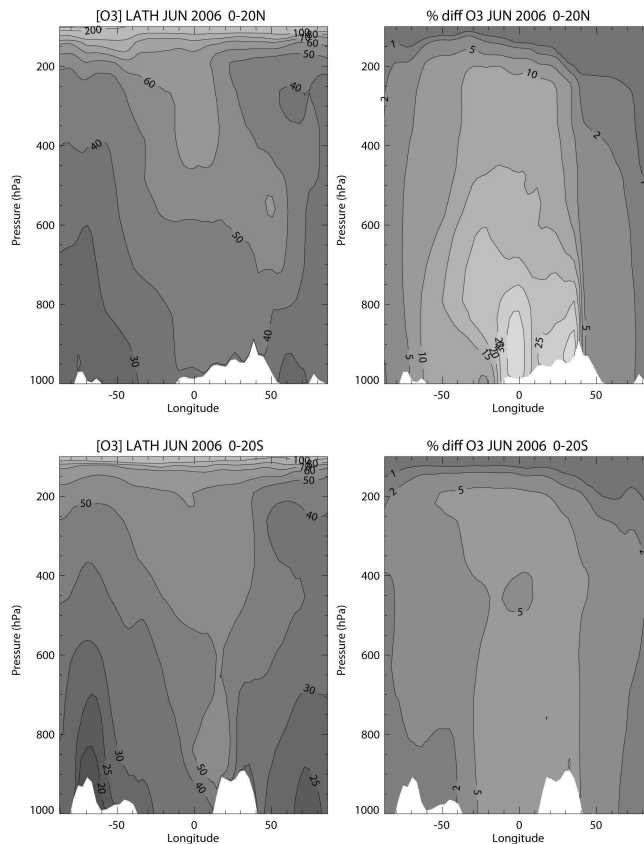


Fig. 8. The concentration of tropospheric O₃ in the LATH simulation and the percentage differences when compared to the NOSOIL simulation as averaged between 0–20° N (top row) and 0–20° S (bottom row) during June 2006. The percentage difference is shown as $(\text{LATH} - \text{NOSOIL}) / \text{LATH} \times 100\%$. For this month the major soil NO_x source is located in the Guinea region.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

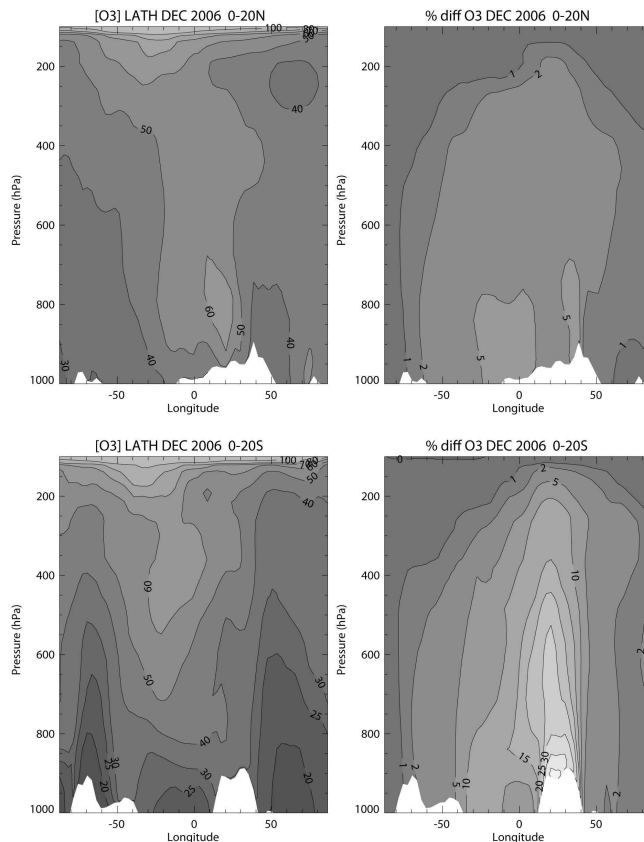


Fig. 9. As for Fig. 8 except for December 2006. For this month the major soil NO_x source is located in the southern Africa region.

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

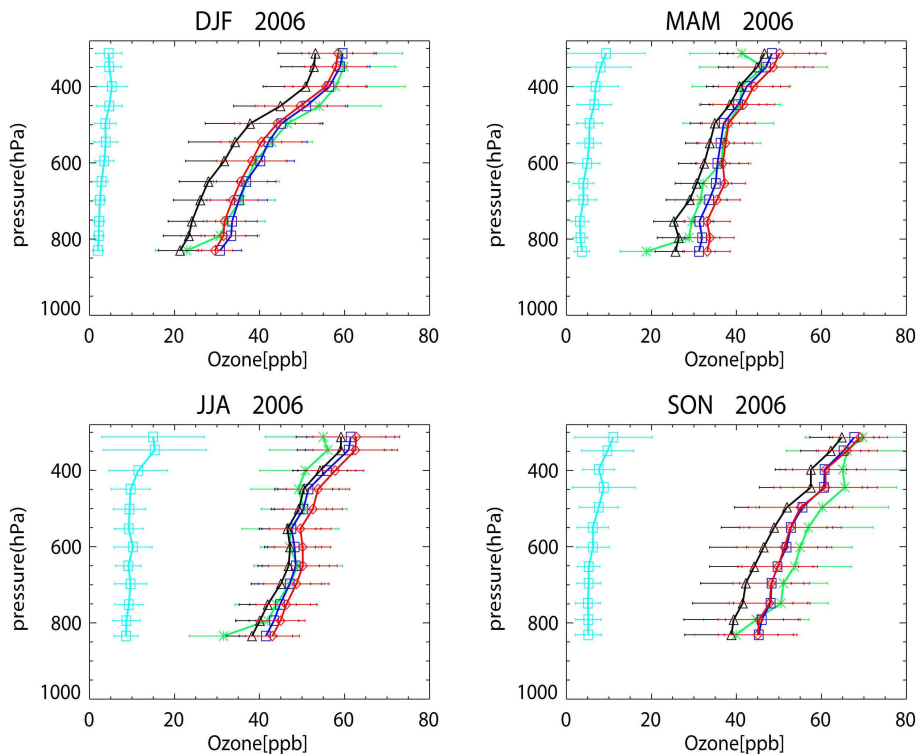


Fig. 10. Comparisons of co-located tropospheric ozone profiles for all seasons compared to MOZAIC take-off and landing measurements from Windhoek in Namibia (22.5° S, 17.5° E). The results from three simulations are shown: (red) BASE, (dark blue) LATH and (black) NOSOIL, and compared against an average of the available measurements (green). The contribution due to stratospheric ozone intrusions is also shown (light blue) using a chemically active ozone tracer, as taken from the LATH run. The error bars represent the 1- σ deviation from the mean.

The influence of biogenic emissions from Africa

J. E. Williams et al.

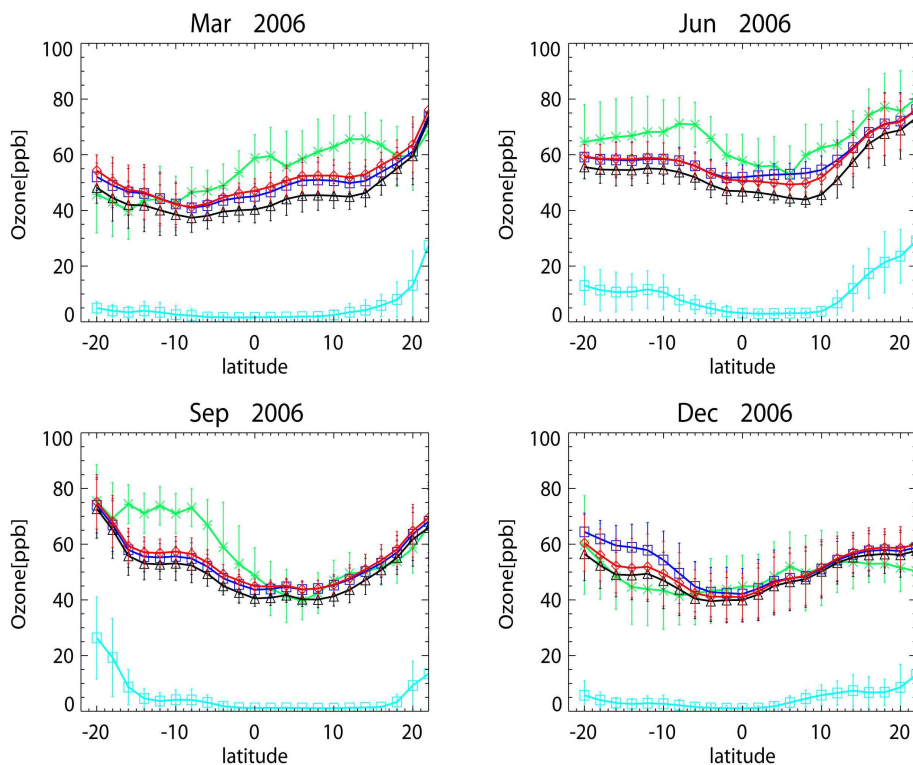


Fig. 11. Comparisons of upper tropospheric ozone as compared to MOZAIC in-flight measurements from Windhoek in Namibia (22.5° S, 17.5° E) to Frankfurt in Germany (50.0° N, 8.6° E). The colour key is identical to that used in Fig. 10. The error bars indicate the 1- σ deviation from the mean.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I◀](#)[▶I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

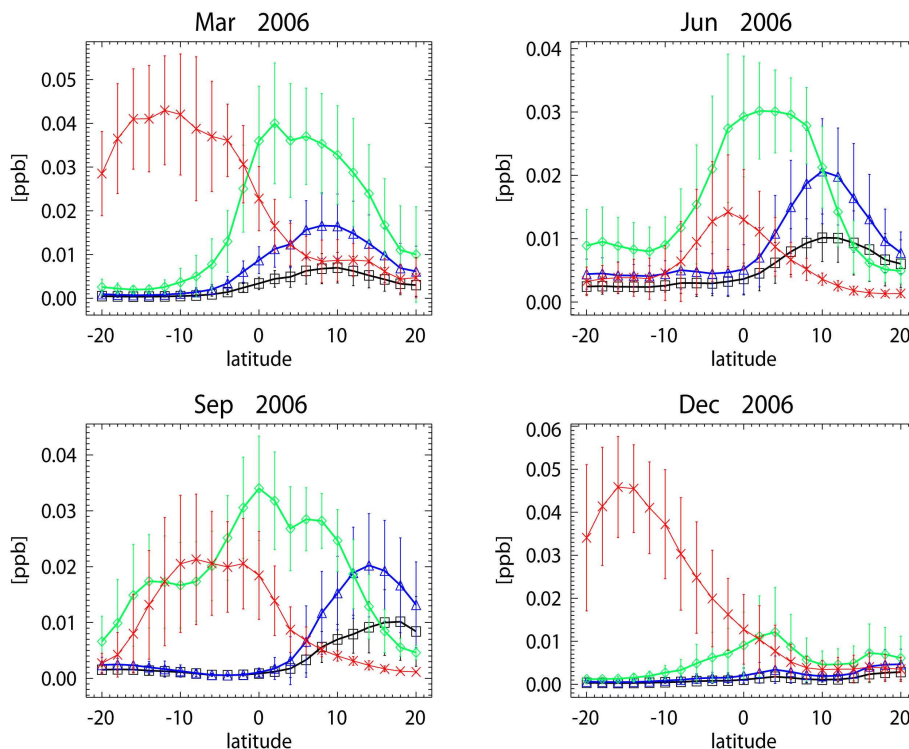


Fig. 12. Comparisons of the four passive tracers used to quantify the strength of convective activity below the MOZAIC flight tracks from Windhoek in Namibia (22.5° S, 17.5° E) to Frankfurt in Germany (50.0° N, 8.6° E). The colour key for the four passive tracer species are as follows: (black) Saharan, (blue) Sahel, (green) Guinea and (red) southern Africa. The error bars indicate the 1- σ deviation from the mean.

The influence of biogenic emissions from Africa

J. E. Williams et al.

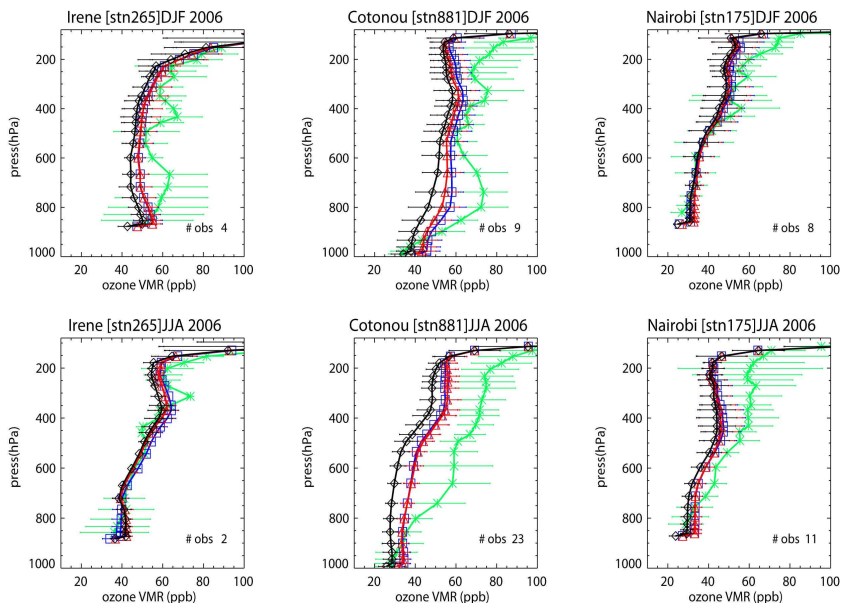


Fig. 13. Comparisons of co-located tropospheric ozone profiles for seasons DJF (top) and JJA (bottom). The soundings shown are taken from (left to right) Irene (25.9° S, 28.2° E), Cotonou (6.6° N, 2.2° E) and Nairobi (1.3° S, 36.8° E), taken from the SHADOZ database (Thompson et al., 2003). The results from three simulations are shown: (red) BASE, (blue) LATH and (black) NOSOIL, and compared against an average of the available measurements (green). The number of observations used for each comparison is shown in the upper left corner of each plot and the error bars indicate the 1- σ deviation from the mean.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

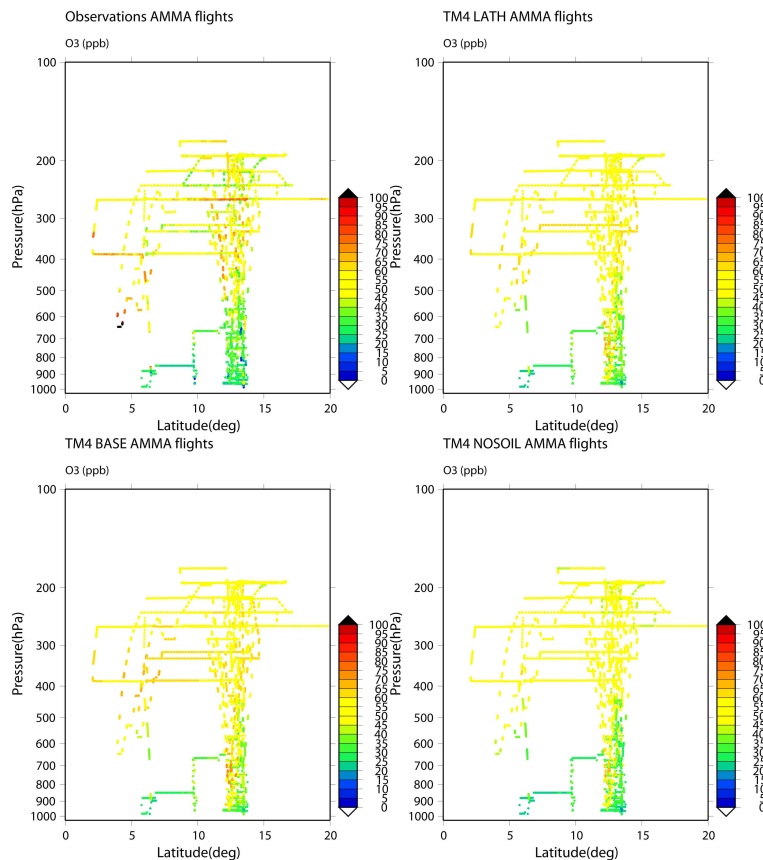


Fig. 14. A comparison of composite O_3 measurements taken above Africa against model output for the various simulations. The figures show the composite of observations for tropospheric O_3 from the various aircraft (top left), the LATH run (top right), the BASE run (bottom left) and the NOSOIL run (bottom right).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

The influence of biogenic emissions from Africa

J. E. Williams et al.

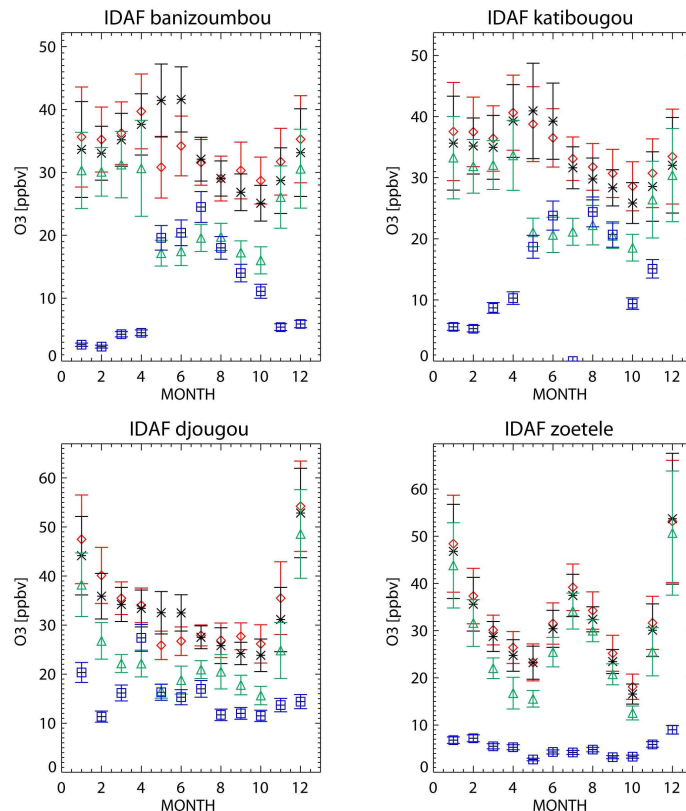
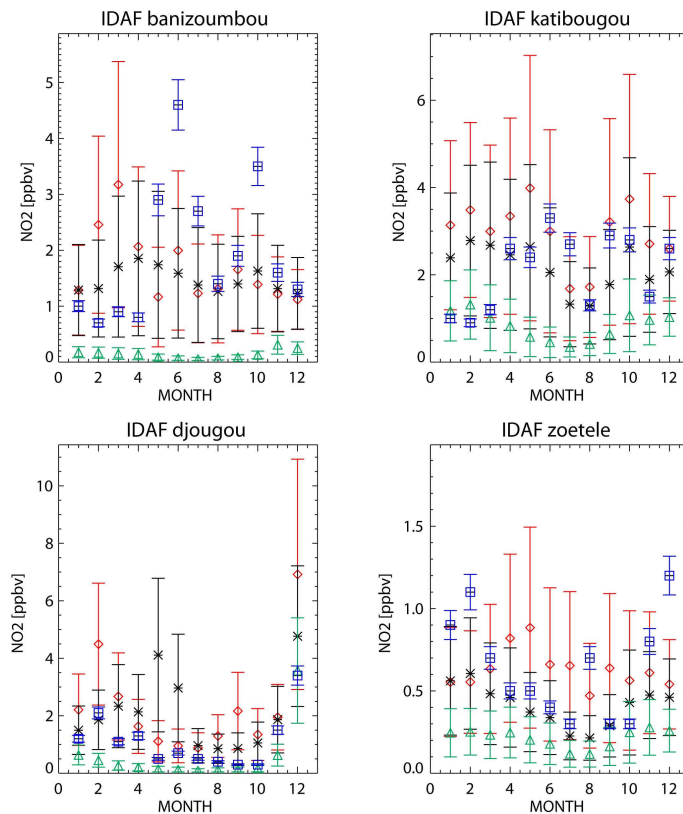


Fig. 15a. Comparisons of monthly mean values for surface O_3 at a selection of IDAF measurement sites, namely: Banizoumbou, Niger (13.3° N, 2.4° E), Katibougou, Mali (10.5° N, 7.3° W), Djougou (9.4° N, 1.6° E), Benin and Zoétéle (3.1° N, 11.5° E), Cameroon. Model results from all simulations are shown: where ($\diamond$) BASE, (*) LATH, (Δ) NOSOIL and ($\square$) are the measurements. The errors on the measurements are taken from Adon et al. (2009) and for the model is 1- σ values are calculated from the 3 hourly output.

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[I ◀](#)[▶ I](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

**The influence of
biogenic emissions
from Africa**

J. E. Williams et al.

**Fig. 15b.** As for Fig. 13a except for NO_2 .

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

I◀

▶I

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



The influence of biogenic emissions from Africa

J. E. Williams et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

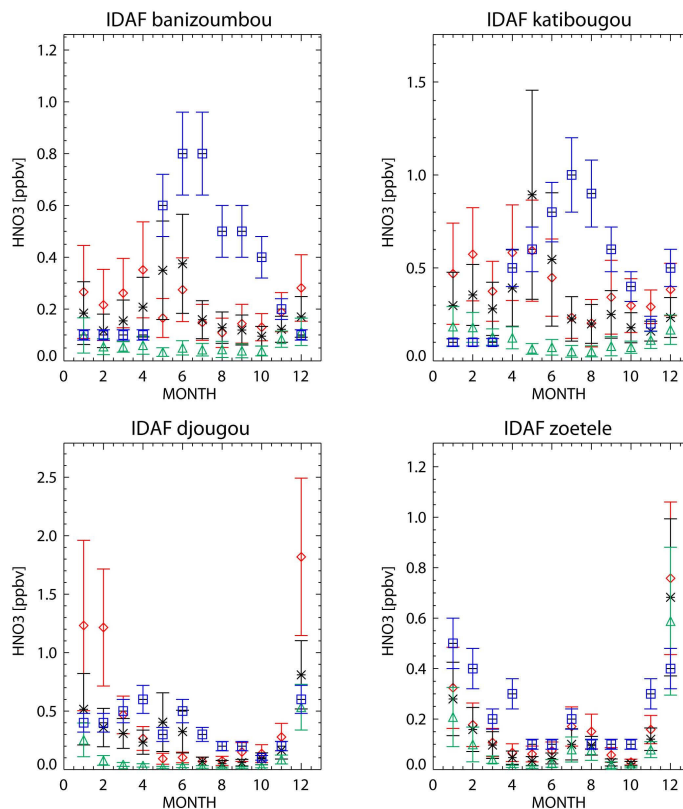


Fig. 15c. As for Fig. 13a except for HNO_3 . The detection limit for $[\text{HNO}_3]$ is 0.1 ppbv.