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**Convection impacts  
on tropospheric  
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R. M. Doherty et al.

# **Influence of convective transport on tropospheric ozone and its precursors in a chemistry-climate model**

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

EGU

## Abstract

The impact of convection on tropospheric  $O_3$  and its precursors has been examined in a coupled chemistry-climate model. There are two ways that convection affects  $O_3$ . First, convection affects  $O_3$  by vertical mixing of  $O_3$  itself. Convection lifts lower tropospheric air to regions where the ozone lifetime is longer, whilst mass-balance subsidence mixes  $O_3$ -rich upper tropospheric (UT) air downwards to regions where the  $O_3$  lifetime is shorter. This tends to decrease UT ozone and the overall tropospheric column of  $O_3$ . Secondly, convection affects  $O_3$  by vertical mixing of ozone precursors. This affects  $O_3$  chemical production and destruction. Convection transports isoprene and its degradation products to the UT where they interact with lightning  $NO_x$  to produce PAN, at the expense of  $NO_x$ . The combined effect of  $NO_x$  to PAN conversions and downward transport of lightning  $NO_x$  results in UT  $NO_x$  decreases. Convective lofting of  $NO_x$  from surface sources appears relatively unimportant. Despite UT  $NO_x$  decreases, UT  $O_3$  production increases as a result of UT  $HO_x$  increases driven by isoprene oxidation chemistry. However, UT  $O_3$  tends to decrease, as the effect of convective overturning of  $O_3$  itself dominates over changes in  $O_3$  chemistry. The changes in tropical UT  $O_3$  are transported polewards resulting in a 15% decrease in the global tropospheric  $O_3$  burden. These results contrast with an earlier study that uses a model of similar chemical complexity. Differences in chemistry schemes – in particular isoprene-driven changes, as well as differences in convection schemes themselves, are the most likely causes of such discrepancies. Further modelling studies are needed to constrain this uncertainty range.

## 1. Introduction

The parameterisation of sub-grid scale convection is known to be a major uncertainty in both chemistry transport models (CTMs) and general circulation models (GCMs). A substantial element of divergence amongst CTM simulations arises from differences

## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

**Convection impacts  
on tropospheric  
ozone and its  
precursors**

R. M. Doherty et al.

in convection parameterisation schemes, which have a particularly large influence on the distributions of short-lived species such as  $\text{NO}_x$  ( $\text{NO}+\text{NO}_2$ ),  $\text{HO}_x$  ( $\text{OH}+\text{HO}_2$ ), and ozone ( $\text{O}_3$ ) (Prather and Jacob, 1997; Collins et al., 1999; Prather et al., 2001; Gauss et al., 2003). Future emissions projections (e.g., Nakicenovic et al., 2000) indicate strong growth of ozone precursor emissions in the tropics – how these are processed by convection will be an important determinant of future ozone concentrations and the oxidising capacity of the atmosphere. GCM simulations project future regional changes in the distribution of convection over many parts of the globe (Cubasch et al., 2001). Convection is therefore important in the context of understanding how future climate change may affect tropospheric chemistry, via changes in both vertical mixing and the distribution of lightning  $\text{NO}_x$  (e.g., Stevenson et al., 2005). Quantifying uncertainties on the impact of deep convection on tropospheric trace gas abundances and distributions are also necessary for assessing how shifts in convection on interannual (and longer) timescales associated with the El Niño Southern Oscillation (ENSO) modulate tropospheric chemistry variability (e.g., Peters et al., 2001).

Since  $\text{O}_3$ , its lifetime and its production efficiency per  $\text{NO}_x$  molecule all typically increase with height, deep convection strongly affects  $\text{O}_3$  and its precursors throughout the tropical tropospheric column (TTC). Considering ozone in isolation, increased vertical mixing tends to lower TTC  $\text{O}_3$ , as  $\text{O}_3$ -poor air mixes upwards, to regions where its lifetime is longer, whilst  $\text{O}_3$ -rich air mixes downwards through mass-balance subsidence, to regions where its lifetime is shorter (e.g., Lelieveld and Crutzen, 1994; Lawrence et al., 2003). However, the effects of convection on ozone precursors also need to be considered. Where there are surface  $\text{O}_3$  precursor sources, especially short-lived ones such as  $\text{NO}_x$  and isoprene ( $\text{C}_5\text{H}_8$ ), convection significantly increases these precursor concentrations in the mid- and upper-troposphere (MT and UT) at the expense of lower troposphere (LT). Collins et al. (1999) and von Kuhlmann et al. (2004) identify convective uplifting of isoprene and its degradation products as a major source of tropical UT  $\text{HO}_x$ . Lawrence et al. (2003) find that lofting of surface  $\text{NO}_x$  is a significant driver of increases in  $\text{O}_3$  production over much of the tropospheric column.

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

---

**Convection impacts  
on tropospheric  
ozone and its  
precursors**R. M. Doherty et al.

---

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

Lightning in the tropics is a major  $\text{NO}_x$  source directly associated with convection, with most  $\text{NO}_x$  added to the UT (Pickering et al., 1998). Convection redistributes lightning  $\text{NO}_x$  emissions downwards at the expense of the UT. There are also potential interactions between isoprene and lightning emissions (e.g., von Kuhlmann et al., 2004).

5 An important isoprene degradation product is the peroxy acetyl radical ( $\text{CH}_3\text{COO}_2$ ), which can affect  $\text{NO}_x$  by promoting PAN formation over land areas where isoprene and lightning  $\text{NO}_x$  emissions are co-located. The net impact of convective mixing is thus sensitive to the profile of  $\text{O}_3$  and its precursors prior to convection, and is a complex balance between transport and a variety of chemical effects. Lawrence et al. (2003)  
10 found the effect of convective changes in precursor emissions on the tropospheric  $\text{O}_3$  burden to be more important than the convective redistribution of  $\text{O}_3$ .

In this paper, the role of convective mixing in determining the distribution of  $\text{O}_3$  and its precursors is investigated using the STOCHEM-HadAM3 coupled chemistry-climate model. Section 2 describes the model and the experiments. Section 3 describes the  
15 influence of convection on  $\text{O}_3$  and its precursors, as well as on  $\text{O}_3$  production and destruction for tropical land and ocean separately. Changes to the global  $\text{O}_3$  budget are then outlined. Section 4 discusses the results of this paper compared to those from other studies, highlights model differences and uncertainties; Sect. 5 presents conclusions.

## 20 2. Model and experiments

The STOCHEM-HadAM3 coupled CTM-GCM model has been described in detail in previous studies (Sanderson et al., 2003a, b; Stevenson et al., 2004, and references therein), so a limited description of relevant model processes is presented here. HadAM3 (Pope et al., 2000) is the atmospheric component of the HadCM3  
25 atmosphere-ocean general circulation model (GCM). In this study, HadAM3 was driven by prescribed monthly sea-surface temperatures (SSTs) for 1980–2002 and was run at standard resolution of  $3.75^\circ$  longitude by  $2.5^\circ$  latitude and 19 vertical levels. These

levels are concentrated at the surface and extend to 10 hPa. Meteorological fields are passed to the CTM every 3 h.

STOCHEM is a Lagrangian tropospheric CTM, first described by Collins et al. (1997), with subsequent major updates to the chemistry (Collins et al., 1999), convection (Collins et al., 2002) and deposition schemes (Sanderson et al., 2003a). Its atmosphere is divided into 50 000 equal mass air parcels that are advected by HadAM3 winds. Air parcels are mapped onto a 5° longitude by 5° latitude horizontal grid with 9 equally spaced vertical levels, which extends from the surface to ~100 hPa. Stratospheric ozone influx is calculated from a present-day stratospheric ozone climatology (Li and Shine 1995) distributed into the troposphere by HadAM3 vertical winds at 100 hPa. Stratospheric input of HNO<sub>3</sub> is modelled similarly assuming a fixed N:O<sub>3</sub> ratio of 1:1000 (Murphy and Fahey, 1994). In this study, the chemical fields from STOCHEM are not fed back into the radiation scheme in HadAM3.

STOCHEM uses the Collins et al. (2002) Lagrangian convective mixing scheme. This mixing scheme uses 3-D convective updraught mass fluxes generated from HadAM3 (Gregory et al., 1997). It represents rapid convective updraughts, entrainment and detrainment of air parcels, and slower mass balance subsidence of larger regions of surrounding air. This scheme simulates radon (<sup>222</sup>Rn) profiles that compare favourably with observations (Collins et al., 2002). The generation of lightning NO<sub>x</sub> is linked to the convection scheme following Price et al. (1997). Convective precipitation from HadAM3 is used to identify lightning occurrence, and cloud height and thickness used to calculate the number of flashes for marine and continental clouds. The profiles of Pickering et al. (1998) are used to vertically distribute the lightning NO<sub>x</sub> emissions for three regimes: midlatitude continental, tropical continental and tropical marine; midlatitude marine regions are treated like tropical marine regions. For these three regimes most lightning NO<sub>x</sub> (55–75% depending on regime) is emitted in the UT above 8 km. Although 20% of lightning NO<sub>x</sub> mass is emitted in the lower troposphere (LT) between 0–1 km in midlatitude continental regions, this amount is relatively small compared to surface anthropogenic and natural (soils and biomass burning) NO<sub>x</sub> emissions over

---

## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.

---

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

these regions (Labrador et al., 2004). Using this lightning  $\text{NO}_x$  scheme and these distribution profiles the ratio of lightning  $\text{NO}_x$  emissions in the UT over tropical land regions compared to oceanic regions is about 20:1 in these model simulations.

Two experiments were performed for the period 1980–2000: a “control” experiment with normal convective mixing of ozone and its precursors, and a “convection off” experiment with no convective mixing of ozone or its precursors, although convection in the climate model – HadAM3 and lightning  $\text{NO}_x$  emissions still occur. Also, water vapour comes from HadAM3 and does not change between the two experiments. The effect of convective mixing of trace gases can be investigated from these experiments.

Anthropogenic and natural emissions of global trace gases in both experiments are identical. Anthropogenic emissions come from the SRES A2 scenario (Nakienovi et al., 2000) for the present-day period. Note that emissions from all the different SRES scenarios are similar over this period. Natural emissions are as given in Stevenson et al. (2004). For analysis, a chemical tropopause is defined using the 150 ppbv ozone isopleth, the definition used by Prather et al. (2001).

### 3. Results

The main region of deep convection, with strong up-draughts that reach the tropopause, occurs in the tropics mainly along the inter-tropical convergence zone (ITCZ) (Fig. 1). Shallower convection that extends up to the mid-troposphere also occurs in the tropics and in mid-latitudes typically associated with frontal activity (Fig. 1).

#### 3.1. Influence of convection over tropical land and ocean

Figure 2 depicts annual zonal mean tropical ozone and  $\text{NO}_x$  concentrations for tropical land and ocean regions separately (Fig. 2a–d) for the control experiment (i.e. normal convective mixing) and the impact of switching on convective mixing (Fig. 2e–h). Both species are generally higher over land, where the main  $\text{NO}_x$  surface and UT lightning

## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

⏪

⏩

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

---

**Convection impacts  
on tropospheric  
ozone and its  
precursors**R. M. Doherty et al.

---

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

sources are located. The ozone lifetime and production efficiency increase with height – these factors, in conjunction with the stratospheric source of ozone – produce the ozone gradient from the surface (25–35 ppb over land, 10–25 ppb over oceans) to the UT (40–60 ppb over land, 30–55 ppb over oceans) (Fig. 2a–b). The vertical distribution of sources gives  $\text{NO}_x$  over land its typical ‘C’ shaped profile (Fig. 2c–d; above 100 ppt in the LT and UT). Over oceans, most of the UT  $\text{NO}_x$  arises from advection of UT land lightning  $\text{NO}_x$  emissions (with a small contribution from lightning over the oceans, Sect. 2) and the  $\text{NO}_x$  lifetime controls the vertical profile (5–20 ppt at the surface, 70–100 ppt in the UT). The shorter  $\text{NO}_x$  lifetime compared to ozone results in a greater  $\text{NO}_x$  land-sea contrast relative to ozone.

Both land and ocean regions show large decreases in UT  $\text{O}_3$  (10–30 ppb) and  $\text{NO}_x$  (100–300 ppt over land; 50–200 ppt over the ocean) due to convection (Fig. 2e–h). LT  $\text{O}_3$  and  $\text{NO}_x$  also decrease over land regions (1–3 ppb and 10–100 ppt respectively, Fig. 2e, g), but show divergent behaviour over the ocean, where  $\text{O}_3$  increases slightly (0–3 ppb), whilst  $\text{NO}_x$  decreases slightly (5–10 ppt), (Fig. 2f, h). Conversely, MT  $\text{O}_3$  and  $\text{NO}_x$  generally increase over land (by 0–5 ppb and 2–50 ppt, Fig. 2e, g), and over the oceans north of 10° S. Over the remote oceans, south of 10° S, MT  $\text{O}_3$  decreases (0–5 ppb) and MT  $\text{NO}_x$  increases slightly (0–5 ppt), (Fig. 2f, h).

Figure 3 displays vertical changes in tropical ozone chemical production and destruction when convective mixing is switched on. Since NO is the main species involved in  $\text{O}_3$  production (through the  $\text{NO}+\text{HO}_2$  reaction), the changes in ozone chemical production with convective mixing are generally similar to that of  $\text{NO}_x$  changes (compare Fig. 3a–b, Fig. 2g–h). However there are some differences. In particular, UT  $\text{O}_3$  production increases over land, whereas  $\text{NO}_x$  decreases.  $\text{O}_3$  chemical destruction is primarily determined by water vapour concentrations and the  $\text{O}_3$  vertical distribution (mainly through the  $\text{H}_2\text{O}+\text{O}(^1\text{D})$  reaction). As water vapour concentrations do not change between experiments, the changes in  $\text{O}_3$  chemical destruction are similar to the changes in ozone (compare Fig. 3c–d, Fig. 2e–f), although again there are some differences. For example, UT  $\text{O}_3$  chemical destruction increases slightly over equatorial land and



in some parts of the MT over ocean south of 10° S, despite decreases of O<sub>3</sub> in these regions.

The effects of convection on NO<sub>x</sub> can partly be understood in terms of a flattening of the 'C' shaped profile over land, and advection of this effect over the oceans. This occurs as UT NO<sub>x</sub> from lightning emissions is displaced to lower altitudes, and surface NO<sub>x</sub> emissions are lifted to higher altitudes, which results in higher NO<sub>x</sub> concentrations in the MT (Fig. 2g–h). However, the UT NO<sub>x</sub> decreases over land are larger by an order of magnitude, suggesting that mixing of NO<sub>x</sub> is not the only factor influencing NO<sub>x</sub> concentrations (Fig. 2g–h). As discussed in Sect. 1, convection also affects non-methane hydrocarbons (NMHCs) including isoprene, lifting these gases and their degradation products into the UT (Collins et al., 1999). Isoprene has a very short lifetime (<1 h; Seinfeld and Pandis, 1997), and is emitted in substantial quantities from tropical vegetation (Guenther et al., 1995; Sanderson et al., 2003b). The natural source of isoprene is much larger than the mainly anthropogenic sources of other NMHCs in the tropics. Here, the prevalence of isoprene, together with its short lifetime, mean that its distribution is strongly affected by convection. Figure 4a shows that convection increases MT and UT isoprene at the expense of surface; these changes are largest over equatorial land. Convective lifting of isoprene brings the peroxy acetyl radical (CH<sub>3</sub>COO<sub>2</sub>), an isoprene degradation product, into contact with lightning NO<sub>x</sub>. This leads to significant PAN formation in the cold UT over equatorial land at the expense of NO<sub>x</sub> (Fig. 4c, Fig. 2g), and this effect is advected across the oceans (Fig. 4d). PAN acts as a reservoir for NO<sub>x</sub>, and its breakdown to release stored NO<sub>x</sub> is strongly temperature dependent – in the cold UT PAN is relatively stable and has a lifetime of the order of months. Thus the impact of convection on NO<sub>x</sub> is through convective transport of NO<sub>x</sub> itself in combination with isoprene-PAN-NO<sub>x</sub> chemistry in the UT.

For O<sub>3</sub>, as discussed above for NO<sub>x</sub>, upward transport of LT air over land and ocean displaces UT ozone-rich air, which subsides downwards. Over land, similar to NO<sub>x</sub>, O<sub>3</sub> is lifted away from its source region and the MT has higher O<sub>3</sub> concentrations. Therefore vertical mixing of the ozone profile itself as well as changes in NO<sub>x</sub> distribution are

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## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.

---

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

**Convection impacts  
on tropospheric  
ozone and its  
precursors**

R. M. Doherty et al.

responsible for the changes in the  $O_3$  distribution over polluted land. However, changes in UT  $NO_x$  over land contrast with changes in UT ozone production over land. Despite a large reduction in UT  $NO_x$  (Fig. 2g) with convective mixing, UT ozone production increases (Fig. 3a). The role of isoprene is again important here, as its oxidation generates peroxy radicals ( $HO_2$  and  $RO_2$ ), which are required for  $O_3$  production. Ozone production in the tropical UT tends to fall due to the reduction in UT  $NO_x$ , but this effect is more than offset by an increase in UT  $HO_2$  produced from isoprene (Fig. 4e, f). As  $HO_2$  also destroys  $O_3$ , the  $HO_2$  increases explain the small equatorial land UT ozone destruction increases (Fig. 3c). Despite an overall increase in net chemical production in the UT, ozone decreases. This indicates that vertical transport of ozone itself is more important than changes in chemistry in determining the outcome of convective mixing in the UT. Over the remote southern oceans, MT  $O_3$  decreases and surface  $O_3$  increases in contrast to  $NO_x$  and ozone production changes. This also implies that over remote tropical locations vertical transport of ozone itself is more important than changes in  $NO_x$  chemistry.

Convection therefore influences modelled  $O_3$  distributions by a combination of vertical transport of  $O_3$ , as well as by affecting the vertical distribution of its precursors, especially  $NO_x$  and  $HO_2$  and the resultant chemical production of  $O_3$ . Vertical transport appears to be the dominant process in clean air, over the remote oceans in the MT and LT, and in polluted land regions in the UT. Elsewhere both convective transport and chemistry changes contribute significantly to the overall impact of convection on the  $O_3$  distribution.

To assess the influence of vertical transport on ozone distribution the relationship between the ozone gradient in the convection off experiment and the change in TC ozone due to convection was examined over the tropics (Fig. 5). A strong correlation was found ( $r=0.7$ ). For each 10 ppb steepening in the ozone gradient between the UT ( $\sim 150$  hPa) and surface, convection reduces TC ozone by 1.5 DU (Fig. 5). The relationship is stronger if the change in UT ozone alone is considered ( $r=0.9$ , not shown). This also suggests that vertical transport of ozone plays a major role in the resultant

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

O<sub>3</sub> distribution after convection, particularly in the UT.

### 3.2. Influence of convection in the extra-tropics

Deep convective mixing in the tropics has a global effect on O<sub>3</sub> distributions as a result of hemispheric transport (Fig. 6a). UT ozone not only decreases in the tropics but also decreases by similar amounts in the midlatitudes and polar regions (10–30 ppb). Since convection is relatively shallow and less intense in the midlatitudes (Fig. 1) compared to the tropics, the extra-tropical O<sub>3</sub> decreases can be inferred to mainly emanate from the tropical UT reduction, whereby there is less tropical UT ozone available to be transported polewards. Slightly lower O<sub>3</sub> stratospheric influx (Table 1) may also contribute to lower extra-tropical UT O<sub>3</sub>. Convective mixing also influences NO<sub>x</sub> distributions well outside the tropics (Fig. 6b). The changes are highly skewed towards the northern hemisphere reflecting the large surface NO<sub>x</sub> emissions in this region. Tropical UT NO<sub>x</sub> decreases (100–200 ppt) are transported towards the midlatitudes; however the strong negative anomaly changes to a positive anomaly polewards of about 45°, especially in the Northern Hemisphere. This reflects the shorter NO<sub>x</sub> lifetime compared to O<sub>3</sub> and the larger hemispheric differences in the levels of NO<sub>x</sub>.

In northern midlatitudes, convection generally reduces surface O<sub>3</sub> and NO<sub>x</sub> (except for O<sub>3</sub> around 60° N), mainly reflecting enhanced venting of polluted continental NO<sub>x</sub> and ozone-rich air by shallow convection (Fig. 6a, b). The increase of O<sub>3</sub> accompanied by a large decrease of NO<sub>x</sub> (100–200 ppt) around 60° N is the titration effect. This effect occurs under very high NO<sub>x</sub> levels predominantly in winter (low UV levels for NO<sub>2</sub> photodestruction) whereby NO reacts with ozone to form NO<sub>2</sub>. Thus with convective mixing, surface air low in ozone and high in NO<sub>x</sub> is replaced by air with higher ozone and lower NO<sub>x</sub> concentrations. In the polar regions, large-scale subsidence of UT air, that is less rich in ozone due to hemispheric transport, results in lower surface ozone. At northern polar latitudes, where winter NO<sub>x</sub> levels are high, weak mixing lifts NO<sub>x</sub> upwards from the LT and MT. This effect combined with mid-latitudinal anomaly transport generates negative LT/MT NO<sub>x</sub> anomalies in the northern pole (Fig. 6b). In

## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

---

**Convection impacts  
on tropospheric  
ozone and its  
precursors**

---

R. M. Doherty et al.

---

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

EGU

the southern midlatitudes as in the southern tropics, where precursor emissions are lesser, surface  $O_3$  increases and  $NO_x$  decreases slightly.

MT  $O_3$  increases in the northern mid-latitudes up to around  $40^\circ N$ , as in the northern tropics, but decreases elsewhere in the midlatitudes and polar regions as large scale subsidence of large UT anomalies dominates the vertical  $O_3$  distribution (Fig. 6a). Shallow convection produces strong MT  $NO_x$  increases (10–50 ppt) in the northern midlatitudes at the expense of the surface (Fig. 6b). These mid-latitudinal MT  $NO_x$  increases are transported to the remote troposphere.

In the column average, convective overturning leads to TC  $O_3$  decreases almost everywhere (Fig. 6c). Decreases are largest (up to 8 DU) over the clean southern ocean regions, stemming from hemispheric transport of lesser amounts of tropical UT  $O_3$ . Over equatorial land regions, where MT  $O_3$  increases are greatest, TC  $O_3$  decreases are smallest (0–2 DU) and in small areas above strong  $NO_x$  source regions (e.g., S.E. Asia) TC  $O_3$  increases slightly (0–1 DU). Tropospheric column-average  $NO_x$  also decreases (strongly) over tropical land regions (50–100 ppt, Fig. 6d) and outflow oceanic regions. The northern mid-latitudes show increases over land and Pacific regions with the largest changes (10–50 pptv, Fig. 6d) over western Eurasia, reflecting upward mixing in shallow convective regions.

Overall, for  $O_3$  deep convective mixing in the tropics is the most important driver of global changes in  $O_3$ , whereby tropical changes are propagated polewards. However, deep convection in the tropics and mid-latitude shallow convection are both important drivers of global  $NO_x$  changes.

### 3.3. Influence of convection on the global $O_3$ budget

Global budgets for  $O_3$  with and without convective mixing are calculated in Table 1. Chemical production increases globally by 3.8%, mainly due to tropical and northern mid-latitudinal MT  $NO_x$  increases (Figs. 6b, 3a). Global chemical destruction also increases (by 3.0%) as a result of MT ozone increases (Fig. 6a, 3c). Changes in chemical production and destruction are small outside the tropics and northern mid-latitudes and

---

**Convection impacts  
on tropospheric  
ozone and its  
precursors**R. M. Doherty et al.

---

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

tropics, respectively (not shown). Because net chemical production is the small residual of two large budget terms it has a relatively large increase of 11.3%. Dry deposition (which increases with surface ozone) globally increases by 3.5%. Stratospheric ozone influx decreases by 5.6% with convective mixing. Although the relative changes in dry deposition and stratospheric influx are modest, the absolute changes are small. Despite increased global net chemical production due to tropical MT increases, the global ozone burden decreases by 14.8%, mainly due to UT decreases outside of the tropics. This illustrates the very strong role of hemispheric transport in modifying ozone concentrations in areas where net chemical production of ozone is low. Likewise, the ozone lifetime decreases globally from 22.4 to 18.5 days (17.3%). The global NO<sub>x</sub> burden also decreases globally by 22.3% and PAN increases by a similar amount (not shown).

#### 4. Discussion

Lelieveld and Crutzen (1994) calculate a 20% decrease in the global ozone burden due to convective mixing, using a global model with no NMHC chemistry. In their model, they found that the effects of mixing O<sub>3</sub> over-ride any chemical effects due to mixing of O<sub>3</sub> precursors. More recently, Lawrence et al. (2003), using a global model with detailed NMHC chemistry (similar to that used in this study), found a global ozone burden increase of 12% from convective mixing. In their study, Lawrence et al. (2003) found the effects of changes in precursor chemistry due to mixing (in particular lifting of surface NO<sub>x</sub> emissions) to outweigh the effect of vertical mixing of O<sub>3</sub> itself. In our study, O<sub>3</sub> decreases by 15% due to convective mixing, and the effect of O<sub>3</sub> vertical transport outweighs the effect of precursor mixing changes (Table 1; Fig. 6a). One similarity between the Lawrence et al. (2003) and this study is that MT O<sub>3</sub> increases due to mixing of precursor NO<sub>x</sub> emissions. Also, in the Lawrence et al. (2003) study, the changes in tropical UT O<sub>3</sub> are small, thus the effect of hemispheric transport will be less apparent than in this study.

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**Convection impacts  
on tropospheric  
ozone and its  
precursors**R. M. Doherty et al.

---

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

Lawrence et al. (2003) also report ozone budgets for their simulations. In common with this study, they find that switching on convection results in an increase in production of  $O_3$ , although they find a larger percentage increase (Table 1). Another similarity is that convection reduces the ozone lifetime; however the impact on lifetime is much stronger in our study. As a consequence, the tropospheric ozone burden falls in our study, whereas Lawrence et al. (2003) find an increase. The reduction in lifetime in our results arises mainly due to a reduction in UT  $O_3$  concentrations. In our model these UT  $O_3$  reductions are driven mainly by the vertical mixing of ozone (Fig. 5), as they occur despite increases in land UT chemical production (Fig. 3a). An interesting observation is that land UT chemical production increases despite large reductions in  $NO_x$  (Figs. 3a, 2g) – this indicates that  $HO_x$  increases (Fig. 4e, f) outweigh the  $NO_x$  decreases (Fig. g, h). In the Lawrence et al. (2003) study UT  $O_3$  increases, which suggests that UT  $O_3$  production increases are greater than in our study.

A comparison of the emissions used in the two studies shows that NO surface, aircraft and ship emissions are similar but lightning  $NO_x$  emissions are higher by 2.1 TgN/yr (~40%) in our study; NMHCs generally are also higher by 5–20 Tg/yr, and isoprene is higher by ~225 Tg/yr (~65%) compared to the MATCH model (von Kuhlmann et al., 2003). The higher isoprene and lightning  $NO_x$  emissions in this study may result in greater amounts of PAN formation and consequently a larger UT  $NO_x$  reduction, although higher isoprene also suggests higher UT  $HO_x$ . Emissions of both lightning  $NO_x$  and isoprene from vegetation carry high levels of uncertainty, and it is unclear whether the higher or lower levels of emissions more closely resemble the real world. Both these natural sources are sensitive to climate, and thus likely to respond to changes in climate (Stevenson et al., 2005).

In addition, there may be significant model differences in these two studies in the isoprene degradation schemes which affects the amount of  $NO_x$  converted to PAN and amount of  $HO_x$  produced, as well as possible differences in locations of lightning  $NO_x$  emissions. The isoprene-lightning  $NO_x$  interaction in this study leads to significant PAN formation, locking up UT  $NO_x$ , and reducing the ability of the UT to produce

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**Convection impacts  
on tropospheric  
ozone and its  
precursors**R. M. Doherty et al.

---

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

O<sub>3</sub>. This mechanism may be much less important in the MATCH-MPIC model used by Lawrence et al. (2003), resulting in larger increases in O<sub>3</sub> production with convection. In STOCHEM, this mechanism limits the effectiveness of convection in promoting chemical production of UT ozone, and consequently the impact of vertical O<sub>3</sub> mixing is the process of overriding importance. In a model sensitivity study, von Kuhlmann et al. (2004) found that isoprene and lightning emissions magnitudes, as well as the details of isoprene degradation schemes, all had important influences on tropical ozone. Differences in isoprene and lightning emissions magnitudes and locations as well as different isoprene chemical schemes may well contribute to differences in ozone sensitivity to convection. Further sensitivity studies are needed to clarify how different emissions and isoprene schemes influence tropospheric O<sub>3</sub> response to convective mixing. If the isoprene emissions or isoprene-lightning interactions are too strong in the STOCHEM model, this may explain its tendency to underestimate O<sub>3</sub> in some parts of the free troposphere; although model estimates are within the uncertainty range of the observations (Stevenson et al., 2004; Dentener et al., 2004). Further validation work is also needed in this area.

Vertical resolution and upper boundary conditions for O<sub>3</sub>, as discussed by Lawrence et al. (2003) could be another source of different responses with convective mixing. However, although the vertical grid resolution is lower in STOCHEM than in the MATCH-MPIC model the Lagrangian sampling in STOCHEM is at roughly the same resolution (50,000 air parcels compared to 64×32×20=40 960 grid cells). Differences in upper boundary conditions may also have some influence.

Finally, differences in the convection schemes themselves (Prather et al., 2001) may be an important, if not the most important cause, of the different effects of convective mixing on the O<sub>3</sub> burden in the two studies. In particular, differences in the efficiency of vertical transport of NO<sub>x</sub> and O<sub>3</sub> out of the boundary layer (Beekman et al., 2003), as well as differences in representing the relevant convective processes (e.g. updraughts, entrainment/detrainment, mass-balance subsidence) may be crucial.



## 5. Conclusions

Model experiments have been performed to examine the role of deep convective mixing in determining the distributions of tropospheric O<sub>3</sub> and its precursors. In contrast to results from an earlier study with a model of similar chemical complexity (Lawrence et al., 2003), convective mixing in this study reduces the O<sub>3</sub> burden, due to vertical O<sub>3</sub> redistribution and associated lifetime changes. Convection of ozone precursors has several competing effects on O<sub>3</sub> chemical production. Over tropical land, convection lofts surface sources of NO<sub>x</sub> and NMHCs in particular isoprene, tending to increase their concentrations in the MT and UT at the expense of the LT. However, the subsidence associated with convection mixes downwards UT lightning NO<sub>x</sub>. UT NO<sub>x</sub> is also converted to PAN by the introduction of peroxy acetyl radicals, an oxidation product of isoprene. The overall effect is a reduction in UT NO<sub>x</sub>, together with an increase in UT HO<sub>x</sub>; the net effect is a small increase in ozone production. However, this increase in O<sub>3</sub> production is more than counteracted by the effects of convective mixing of ozone itself, and UT O<sub>3</sub> reduces. The features generated over the tropical land are advected over the tropical oceans, although over the remote oceans, the impact of local vertical mixing becomes more dominant. The changes in tropical UT O<sub>3</sub> are transported polewards resulting in a 15% decrease in the global ozone burden despite a 4% increase in chemical production. This apparent contradiction arises because the mean tropospheric ozone lifetime reduces substantially (by 17%); this is mainly because less O<sub>3</sub> resides in the UT, where its lifetime is longest. Differences between this study and that of Lawrence et al. (2003) arise from a combination of variations in chemistry and convection schemes, with possibly slightly differing treatments of lightning NO<sub>x</sub> and vegetation isoprene emissions proving important. With such divergent results from two apparently similar models (in terms of resolution and complexity), and the importance of deep convective mixing processes in altering the state of the atmosphere on interannual and longer timescales, further studies with other models are needed to constrain this range of uncertainty.

### Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion



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## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

---

**Convection impacts  
on tropospheric  
ozone and its  
precursors**

---

R. M. Doherty et al.

---

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

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**Convection impacts  
on tropospheric  
ozone and its  
precursors**

R. M. Doherty et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.

**Table 1.** 20-year average O<sub>3</sub> budget changes (control – convection off).

Budget term (Tg (O <sub>3</sub> )yr <sup>-1</sup> )	20-year average control (convection on)	20-year average convection off	% change with convective mixing	Lawrence et al. (2003) % change	Lelieveld and Crutzen (1994) % change
O <sub>3</sub> chemical production	4757	4582	+3.8	+16.7	+14
O <sub>3</sub> chemical destruction	4236	4114	+3.0	+15.0	+22
O <sub>3</sub> net chemical production	521	468	+11.3	+81.8	-25
O <sub>3</sub> dry deposition	881	851	+3.5	+7.0	+15
O <sub>3</sub> stratospheric influx	361	383	-5.6	-2.1	
O <sub>3</sub> burden (Tg O <sub>3</sub> )	263	309	-14.8	+11.9	-20
O <sub>3</sub> lifetime (days)	18.5	22.4	-17.3	-2.5	
NO <sub>x</sub> burden (Tg N)	0.156	0.200	-22.3	+14	-30

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

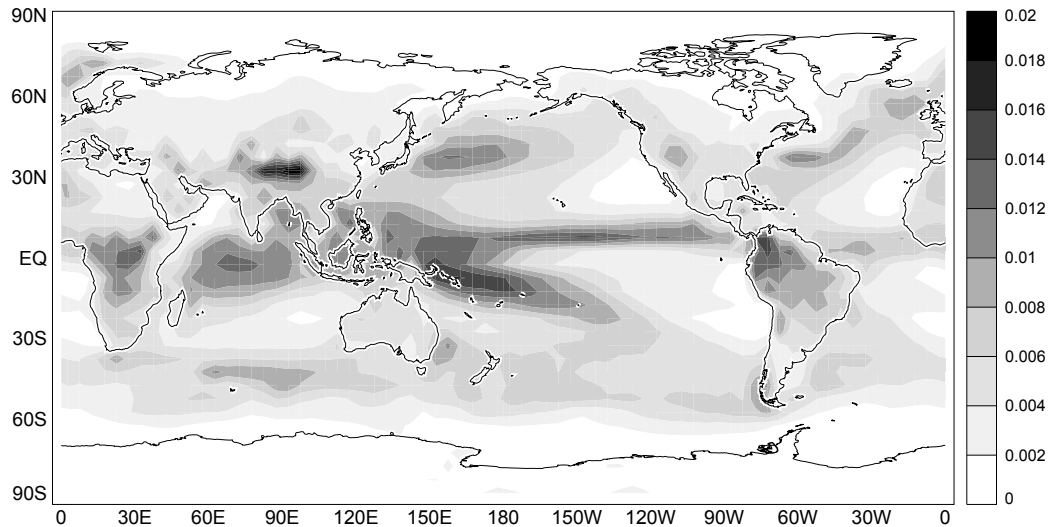
Full Screen / Esc

Print Version

Interactive Discussion

**Convection impacts  
on tropospheric  
ozone and its  
precursors**

R. M. Doherty et al.

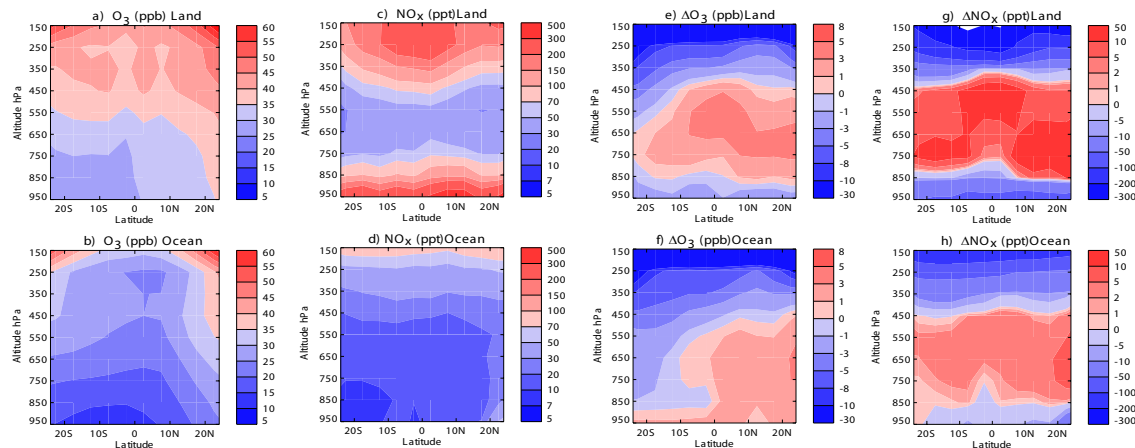


**Fig. 1.** 20-year average convective updraught flux ( $\text{kg s}^{-1}$ ) at 600 hPa (MT) from the control experiment. Generally only in the tropics do updraught fluxes reach 150 hPa (not shown).

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

## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.



**Fig. 2.** 20-year average zonal mean tropical land and ocean (a, b)  $O_3$  (ppb) and (c, d)  $NO_x$  (ppt) for the control experiment and 20-year average zonal mean tropical land and ocean (e, f)  $O_3$  and (g, h)  $NO_x$  differences control-convection off.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

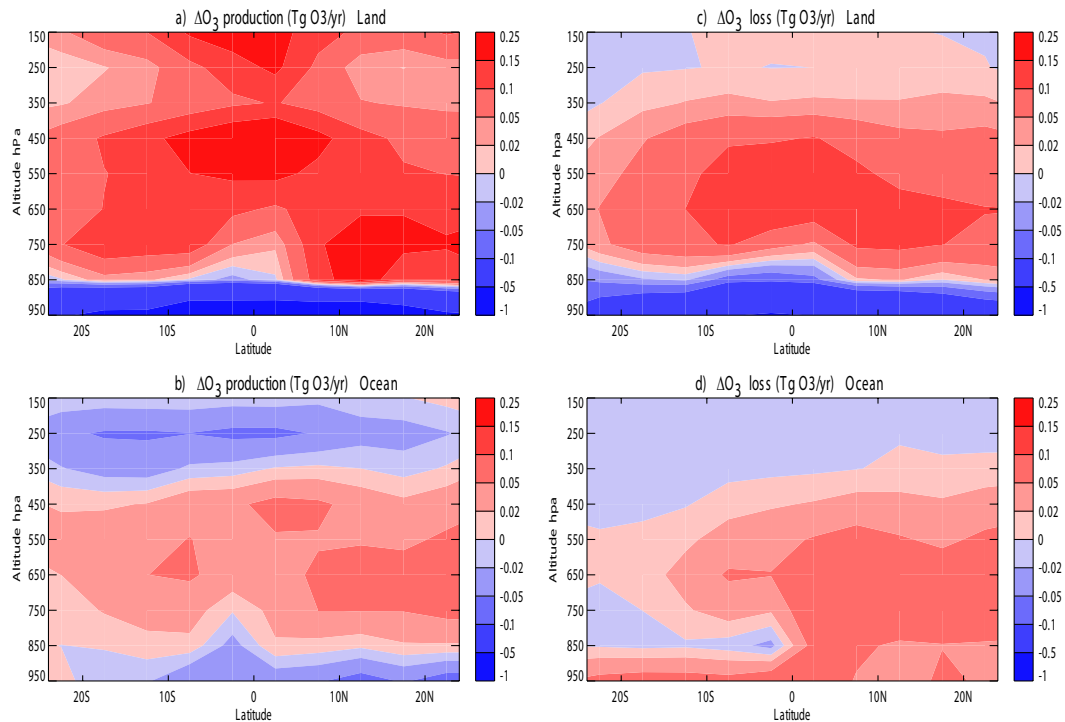
Full Screen / Esc

Print Version

Interactive Discussion

**Convection impacts  
on tropospheric  
ozone and its  
precursors**

R. M. Doherty et al.

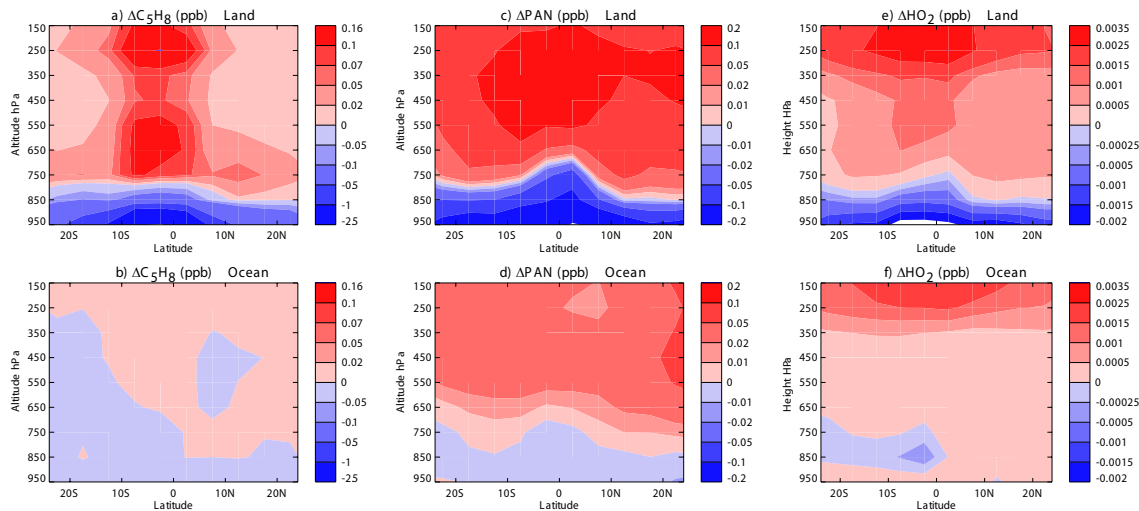


**Fig. 3.** 20-year average zonal mean tropical land and ocean ozone (a, b) production and (c, d) loss differences (Tg O<sub>3</sub>) control-convection off.

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

## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.



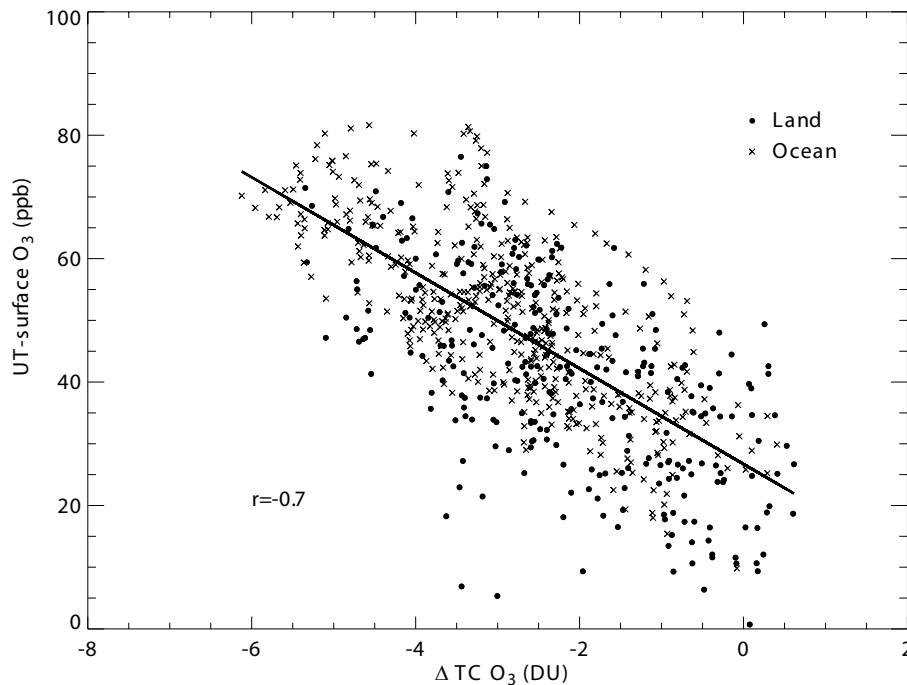
**Fig. 4.** 20-year average zonal mean tropical land and ocean (a, b) isoprene, (c, d) PAN and (e, f) HO<sub>2</sub> differences (ppb) control-convection off.

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



**Convection impacts  
on tropospheric  
ozone and its  
precursors**

R. M. Doherty et al.

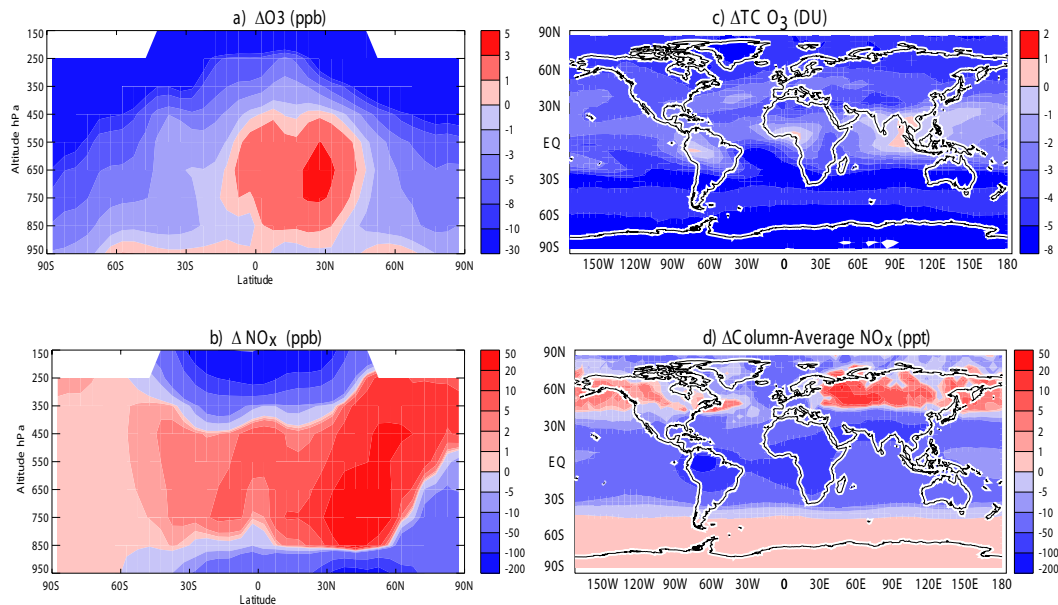


**Fig. 5.** 20-year average change in TC O<sub>3</sub> (DU) with convective mixing (control-convection off) vs. 20-year average UT-surface O<sub>3</sub> (ppb) (convection off experiment) for combined tropical land and ocean grid boxes.

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

## Convection impacts on tropospheric ozone and its precursors

R. M. Doherty et al.



**Fig. 6.** 20-year average global zonal mean change in **(a)** O<sub>3</sub> (ppb) and **(b)** NO<sub>x</sub> (ppt) (control – convection off), and 20-year global average **(c)** TC O<sub>3</sub> change (DU) **(d)** column-average NO<sub>x</sub> change (ppt) (control – convection off).

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

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