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# Tropospheric ozone variations governed by changes in stratospheric circulation

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**The downward transport of stratospheric ozone is an important natural source of tropospheric ozone, particularly in the upper troposphere, where changes in ozone have their largest radiative effect[1]. Stratospheric circulation is projected to intensify over the coming century, which could lead to an increase in the flux of ozone from the stratosphere to the troposphere[2–4]. However, large uncertainties in the stratospheric contribution to trends and variability in tropospheric ozone levels[5–7] make it difficult to reliably project future changes in tropospheric ozone[8]. Here, we use satellite measurements of stratospheric water vapour and tropospheric ozone levels collected between 2005 and 2010 to assess the effect of changes in stratospheric circulation, driven by El Niño/Southern Oscillation and the stratospheric Quasi-Biennial Oscillation, on tropospheric ozone levels. We find that interannual variations in the strength of the stratospheric circulation of around 40%—comparable to the mean change in stratospheric circulation projected this century[2]—lead to changes in tropospheric ozone levels in the northern mid-latitudes of around 2%, approximately half of the interannual variability. Assuming that the observed response of tropospheric ozone levels to interannual variations in circulation is a good predictor of its equilibrium response, we suggest that the projected intensification of the stratospheric circulation over the coming century could lead to small but important increases in tropospheric ozone levels.**

Modelling studies and observational analyses, the latter generally based on sparse data over limited regions, suggest that the stratosphere may play an important role in modulating tropospheric ozone abundances[5–7], but the magnitude of the stratospheric contribution and its importance in the tropospheric ozone budget are poorly constrained. Coupled chemistry–climate models (CCMs) consistently predict increases in the stratospheric circulation over the next century[2–4], with corresponding circulation-driven increases in the stratosphere-to-troposphere (STE) ozone flux of 20–30% from 1965 to 2095[2]. Most of these CCMs, however, lack comprehensive tropospheric chemistry and cannot be used to assess the impact of changes in the STE ozone flux on tropospheric ozone. A few tropospheric CCMs, which have poorly resolved stratospheres and often use prescribed stratospheric ozone, have examined the role of the stratosphere in future tropospheric ozone trends. They produce a much larger range of climate-driven STE ozone flux increase (25–80%) than stratospheric CCMs, and the magnitude of the change in STE ozone flux in some cases determines whether mid-latitude tropospheric ozone increases or decreases given other climate- and emissions-driven changes in the budget[9–11]. Estimates of the tropospheric ozone response to changes in the STE ozone flux are thus critical to developing effective air quality policies.

Six years of ozone and water vapour measurements from the Tropospheric Emission Spectrometer (TES) and Microwave Limb Sounder (MLS) onboard NASA's Aura satellite (Supplementary Information 1) reveal that dynamical variability driven by El Niño/Southern Oscillation (ENSO) and/or the stratospheric Quasi-Biennial Oscillation (QBO) provides a 'natural experiment' that allows us to quantify the impact of changes in the stratospheric circulation on tropospheric ozone. As discussed below, ENSO and the QBO are highly correlated during the observational period, making it impossible to separate the contribution of the two modes to observed circulation changes. Previous studies have shown that both ENSO and the QBO modulate the stratospheric circulation[12–14] and ozone[15–17], and, according to models, the STE ozone flux[18–20]. Other analyses have assessed variability in the stratospheric circulation and STE ozone flux using MLS measurements[21,22], without examining the origins or tropospheric effects of this variability. This paper provides an observation-based end-to-end connection of these elements and an assessment of their impact on tropospheric ozone.

As shown below and illustrated in Fig. 1, El Niño/easterly shear QBO (Fig. 1a,b) act to strengthen the stratospheric overturning circulation (particularly during winter, via mechanisms described in Supplementary Information 2) (Fig. 1c) and hence increase transport of air from the ozone maximum poleward and downward to mid-latitudes (Fig. 1d), which increases the STE ozone flux and thus tropospheric ozone (Fig. 1e). In contrast, La Niña/westerly shear QBO is associated with a weakened circulation and decreases in the STE flux and ozone. For the purpose of examining the tropospheric impact of changes in stratospheric transport, El Niño/easterly shear QBO therefore provides an analogue to climate change (with caveats discussed below) in which increased greenhouse gases (GHGs) are predicted to strengthen the circulation, leading to increased STE ozone flux[2,3].

Figure 2a shows the 2005–2010 time series of the Multivariate ENSO Index[23] and a QBO shear index calculated from the difference in the Singapore zonal wind at 50 hPa and 25 hPa (U50 − U25; ref. 24), along with deseasonalized zonal mean anomalies in stratospheric tropical upwelling derived from MLS water vapour measurements[21] (Fig. 2b; Supplementary Information 3), northern mid-latitude lower stratospheric ozone from MLS (Fig. 2c), and northern mid-latitude mid-tropospheric ozone from TES (Fig. 2d). We focus on the Northern Hemisphere because variability in lower

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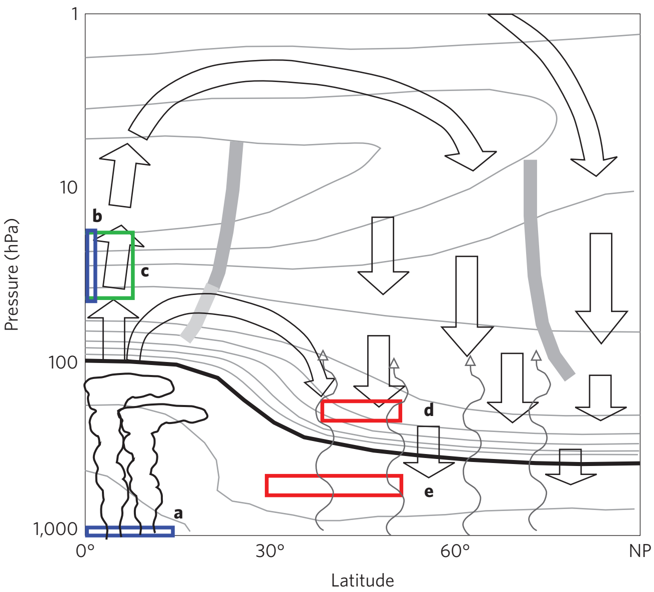


**Figure 1 | Schematic of processes responsible for observed interannual variability in stratospheric and tropospheric ozone and the location of observations/diagnostics.** Thin grey lines represent ozone isopleths, thin wavy arrows planetary-scale waves (Supplementary Information 2), and large arrows stratospheric circulation for Northern Hemisphere winter. The thick black line indicates the tropopause, and thick grey lines mixing barriers in the subtropics and at the polar vortex edge, with lighter grey indicating weaker transport barriers. **a**,**b**, The blue boxes (**a**) and (**b**) show regions of the Multivariate ENSO index[22] and QBO shear index, respectively. **c**, The green box shows the location of water vapour observations used to diagnose tropical upwelling. **d**,**e**, The red boxes (**d**) and (**e**) show the locations of mid-latitude lower stratospheric and mid-tropospheric ozone observations, respectively.

stratospheric ozone is approximately three times larger there than in the Southern Hemisphere for this period.

The tropical upwelling anomalies indicate variability in the 8° S–8° N Brewer–Dobson circulation of $\sim\pm40\%$, which is representative of but probably larger than the variability over the broader upwelling region (Supplementary Information 3). The upwelling anomalies are highly correlated with both ENSO and QBO shear indices from the prior month (Fig. 2e,f). Although ENSO and the QBO are not always in phase, they are highly correlated from 2005 to 2010 ($R = 0.67$). Recent work has shown that the QBO nonlinearly modulates ENSO variability in wave activity[25,26], such that the interplay between the two modes in affecting the circulation can be quite complex.

Ozone in the mid-latitude lower stratosphere is primarily controlled by transport. The 'lower branch' of the stratospheric circulation carries air directly from the tropics to mid-latitudes just above the tropical tropopause, whereas the 'upper branch' carries air through the ozone maximum in the middle stratosphere[27] (Fig. 1). Observed anomalies in northern mid-latitude lower stratospheric ozone (Fig. 2c) are correlated with anomalies in middle stratospheric tropical upwelling (Fig. 2b). However, owing to variations in the timing and magnitude of tropical upwelling anomalies over the 56–26 hPa range, upwelling at a single pressure level (56 hPa) provides the highest correlation with ozone, with a lag of two months between a change in upwelling and the ozone response (Fig. 2g). Physically, it is clear that the ozone variations must primarily reflect variations in the upper branch of the circulation because the lower branch carries air with tropical ozone abundances, which are much lower than those in mid-latitudes and therefore cannot be responsible for ozone increases observed during El Niño/easterly shear QBO years, and because observed ENSO/QBO anomalies in tropical lower stratospheric ozone are opposite in sign to those in mid-latitudes (Fig. 3).

The time series of deseasonalized zonal mean anomalies in Northern Hemisphere mid-tropospheric ozone (Fig. 2d) exhibits a positive trend not seen in the lower stratosphere (Fig. 2c). Figure 2h shows the relationship between anomalies in Northern Hemisphere lower stratospheric ozone (Fig. 2c) and detrended anomalies in mid-tropospheric ozone (Fig. 2d) one month later. Ozone is averaged over a larger latitude range in the troposphere (30° N–50° N) than in the stratosphere (40° N–50° N) because the anomalies slope equatorwards with increasing pressure (Fig. 3). Correlations between stratospheric and tropospheric ozone anomalies have been shown to indicate transport of ozone from the stratosphere to the troposphere[6] (Supplementary Information 4). The correlation shown in Fig. 2h thus indicates that changes in stratospheric ozone driven by variations in the large-scale circulation account for ~16% of the variability of zonal mean ozone in the northern mid-latitude mid-troposphere based on $R^2$. Furthermore, the slope of the regression line (0.075 with a bootstrap standard error of $\pm0.026$) indicates that a 25% increase in stratospheric ozone results in an increase of ~2% in tropospheric ozone (equal to ~1/2 the total tropospheric ozone variability of $\pm4\%$), consistent with a recent modelling study[20] (Supplementary Information 5). Given the spatial heterogeneity of the STE flux[20], regional impacts of STE flux variations on tropospheric ozone may be much larger than those seen in the zonal mean.

Figure 3 shows the contrast between mean ozone anomalies during La Niña/westerly shear QBO (Fig. 3a) and El Niño/easterly shear QBO (Fig. 3b) conditions. In the tropical upper troposphere, the ozone anomalies reflect changes in tropical convection associated with ENSO. During El Niño, the decrease in upper tropospheric ozone associated with increased convection over the central/eastern Pacific exceeds the increase in upper tropospheric ozone arising from decreased convection over the western Pacific[15], resulting in a negative zonal mean ozone anomaly. The situation is reversed during La Niña, producing a positive zonal mean ozone anomaly. At $p < 100$ hPa, the ozone anomalies reflect changes in tropical upwelling, with stronger (weaker) upwelling bringing up more (less) low-ozone air from the tropical upper troposphere. The mid-latitude anomaly is out of phase with the tropical anomaly throughout the entire Northern Hemisphere middle and lower stratosphere, and the largest mid-latitude tropospheric ozone anomalies are found near the subtropical jet (30° N–40° N), where most transport from the stratosphere to the troposphere occurs[20]. Southern Hemisphere ozone anomalies are discussed in Supplementary Information 6.

This analysis represents a new and unique way of investigating the relationship between stratospheric and tropospheric ozone and provides the first empirical estimate of the response of tropospheric ozone to changes in the stratospheric circulation. These observations introduce stringent constraints for CCMs and can be used to diagnose model errors in stratospheric and tropospheric ozone variability and the relationship between ENSO/QBO, the stratospheric circulation, and stratospheric and tropospheric ozone. The 20–30% 1965–2095 increase in STE ozone flux found in CCMs is associated with increases in tropical upwelling that are similar in magnitude to the circulation change associated with ENSO/QBO (Supplementary Information 7). The Community Atmosphere Model with Chemistry (CAM-Chem)[28] has until recently been one of very few CCMs with a well-resolved stratosphere and comprehensive tropospheric chemistry, both of which are required to assess the impact of changes in stratospheric transport on tropospheric ozone. Analysis of a modified CCM-Validation activity REF-B2 simulation[2] with constant ozone precursors (Supplementary Information 8) shows a

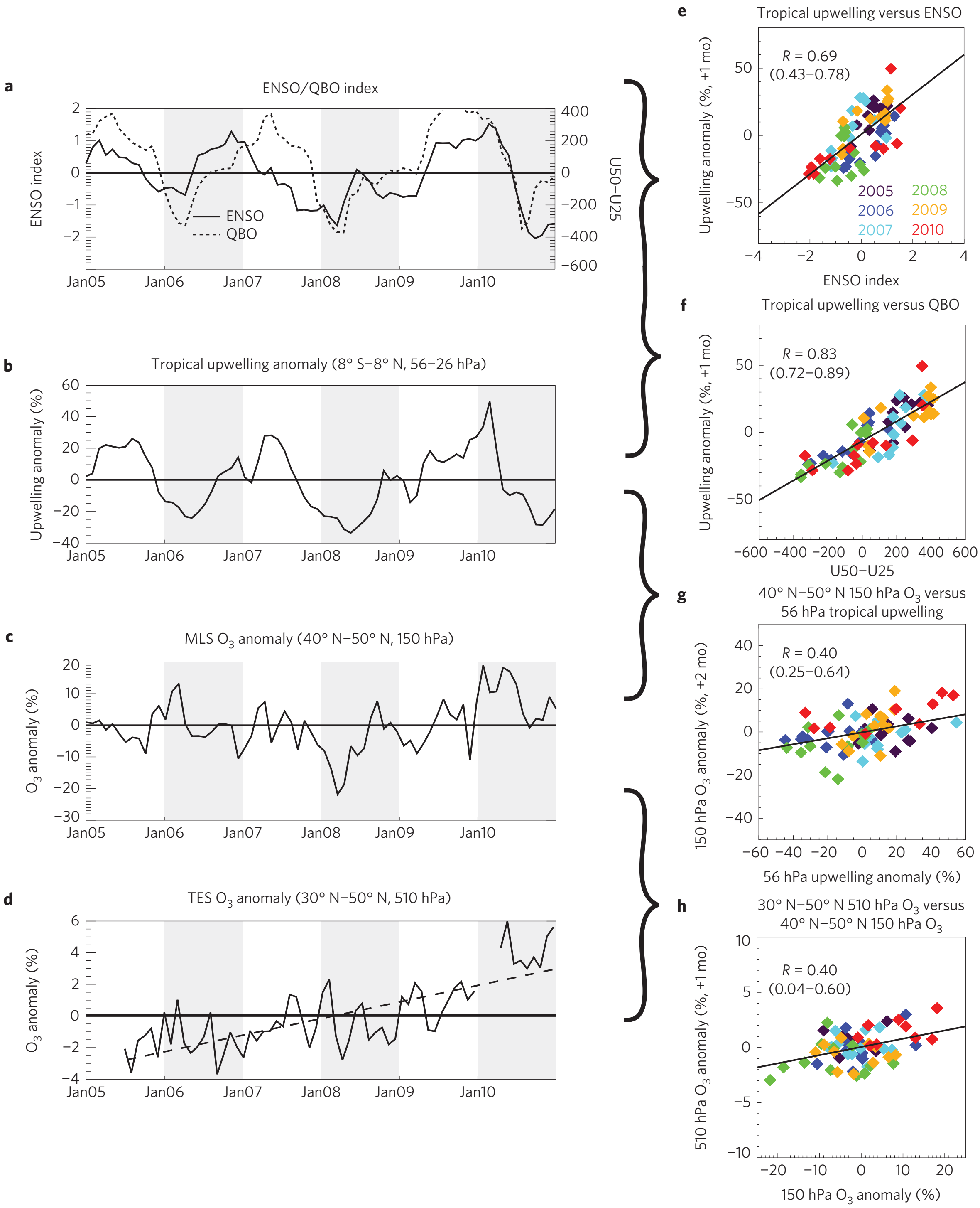


**Figure 2 | Time series of and relationships between ENSO/QBO, stratospheric circulation and ozone. a**, Time series of Multivariate ENSO Index[22] (solid line) and QBO shear index (dashed line). **b–d**, Time series of deseasonalized monthly mean zonal mean anomalies for 8° S–8° N 56–26 hPa tropical upwelling from MLS water vapour[20] (**b**), MLS 40° N–50° N 150 hPa ozone (**c**), and TES 30° N–50° N, 510 hPa ozone (**d**). Grey shading indicates even years. Dashed line in **d** shows the linear trend in TES ozone. **e**,**f**, Scatter plots of monthly mean tropical upwelling anomalies versus ENSO index (**e**) and QBO index (**f**) one month prior. **g**, Scatter plot of MLS 150 hPa ozone anomalies versus 56 hPa tropical upwelling anomalies two months prior. **h**, Scatter plot of TES 510 hPa detrended ozone anomalies versus MLS 150 hPa ozone anomalies one month prior. Black lines show the least-squares fit to the data; colours indicate year of anomalies on the *x* axis of each plot as given in **e**. Correlation coefficients and 95% bootstrap confidence intervals (in parentheses) are provided.

similar-to-observed relationship between stratospheric and tropospheric ozone anomalies for the recent past (1980–2000) ($R = 0.43$, with a 25% change in 40° N–50° N lower stratospheric ozone resulting in a 2% change in 30° N–50° N tropospheric ozone). The mean increase in northern mid-latitude lower stratospheric ozone from the recent past (1990–2000) to the future (2090–2100) is 24%, and the total climate-driven mean increase in northern mid-latitude mid-tropospheric ozone, which we expect to be

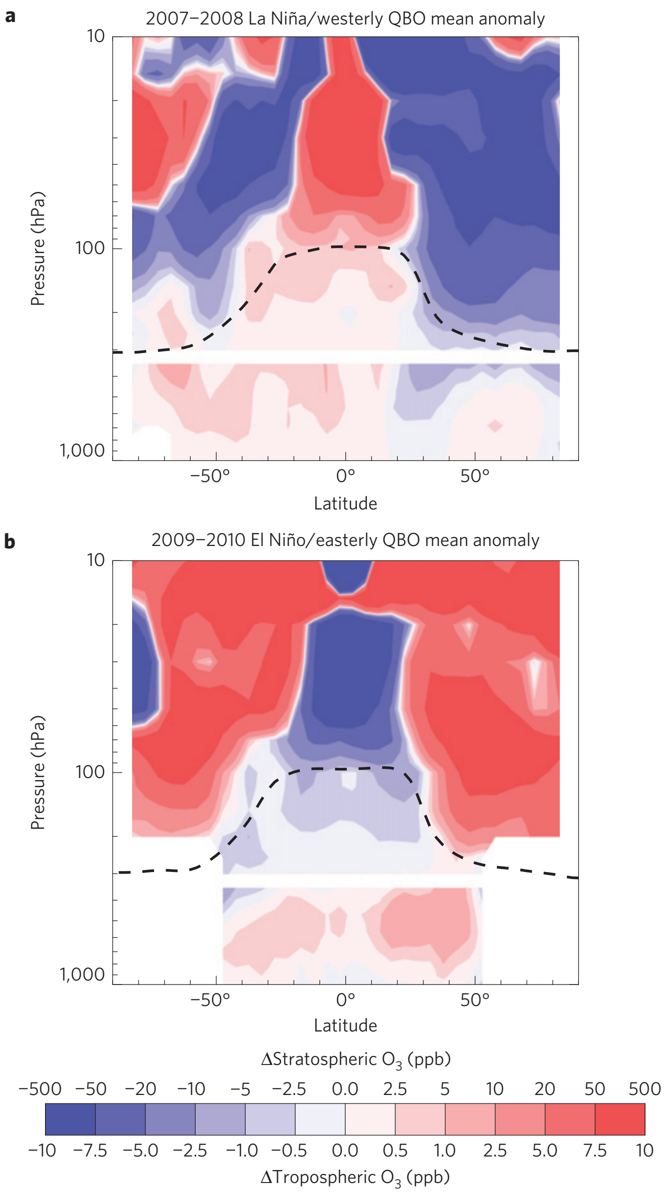


**Figure 3 | Latitude–pressure cross sections of ozone during La Niña/westerly shear QBO and El Niño/easterly shear QBO. a**, Mean deseasonalized zonal mean ozone anomalies (ppb) averaged over the time period of the mid-latitude lower stratospheric ozone anomalies associated with the strongest La Niña/westerly shear QBO in the record (November 2007–August 2008). For $p \leq 200$ hPa, we use the mean of the TES and MLS anomaly time series; for $p > 200$ hPa only TES data are used. Anomaly values for $p < 300$ hPa are shown on top of the colour bar, values for $p > 300$ hPa are shown below. The dashed line shows the mean tropopause location for the same period from the National Center for Environmental Prediction (NCEP) National Center for Atmospheric Research (NCAR) reanalysis[30]. **b**, Same as **a** but for the strongest El Niño/easterly shear QBO (June 2009–August 2010).

dominated by increases in temperature and the STE ozone flux (Supplementary Information 8), is 5%. This is similar in magnitude to the change in tropospheric ozone predicted by both the modelled and observed relationship between stratospheric and tropospheric ozone anomalies and about one fifth of the increase in tropospheric ozone seen in response to a similar change in stratospheric ozone in a model without tropospheric chemistry[4], results from which have been improperly used to estimate an expected 30% increase in tropospheric ozone due to future changes in stratospheric transport[7].

A suite of CCMs with comprehensive troposphere–stratosphere dynamics and chemistry is required to robustly determine whether model differences in the equilibrium response of tropospheric ozone to long-term changes in stratospheric ozone are correlated to differences in modelled ENSO/QBO-driven interannual variability, such that the observations presented here can provide constraints on long-term changes in mean tropospheric ozone. The mechanisms by which ENSO, the QBO, and increased GHGs alter the stratospheric circulation differ[12,14,29]. However, mechanistic differences may not be important vis-à-vis quantifying the impact of circulation changes on tropospheric ozone because the upper branch of the circulation is affected (and thus changes in the mass flux are similarly related to changes in ozone transport) for both ENSO/QBO (as shown by this work) and GHGs (refs 2–4). Our observations establish the relationship between stratospheric circulation changes and tropospheric ozone for the current climate and, combined with the CAM-Chem results, suggest that GHG-induced changes in the stratospheric circulation will lead to increases in mid-tropospheric ozone that, although modest, represent a significant fraction of the total ozone increase associated with climate change.

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## Author contributions

J.L.N. led the analysis and wrote the paper. T.F. calculated the tropical upwelling rates from MLS water vapour measurements. G.L.M., M.L.S. and N.J.L. provided expertise on the use of MLS data, and J.W. provided expertise on the use of TES data. All authors contributed comments on the manuscript.

## Additional information

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## Competing financial interests

The authors declare no competing financial interests.