

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Stratospheric impact on tropospheric ozone variability and trends: 1990–2009

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Received: 22 June 2011 – Accepted: 7 July 2011 – Published: 11 August 2011

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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We evaluate the influence of stratospheric ozone on the interannual variability and trends in tropospheric ozone from 30–90° N between 1990 and 2009 using ozone measurements and a global chemical transport model (the Community Atmospheric Model with chemistry) with input meteorology from the National Center for Environmental Prediction. The model simulation uses constant interannual emissions. Both the model and measurements indicate that on large spatial scales stratospheric interannual ozone variability drives significant tropospheric variability and contributes to long-term tropospheric ozone trends. To diagnose the measured variability we utilized measurements from ozonesondes and the Measurements of OZone and water vapour by in-service Airbus airCRAFT programme (MOZAIC) north of 30° N. We identify a regionally robust 150 hPa ozone signal from measurements over Canadian, Northern European and Central European regions and at 500 hPa over Canadian, Northern European and Eastern US regions. Averaged over these regions, the 150 hPa interannual ozone variability explains 69 % of the interannual variability at 500 hPa. The simulated stratospheric signal explains 81 % of the simulated variability over these same regions. Simulated and measured ozone are significantly correlated over these regions and the simulation suggests that the ozone record over these regions is representative of the overall hemispheric 500 hPa ozone record from 30–90° N. The measured 500 hPa trends averaged over these three regions between 1990 and 2000 and 1990 and 2009 are $0.73 (\pm 0.51) \text{ ppbv yr}^{-1}$ and $0.27 (\pm 0.19) \text{ ppbv yr}^{-1}$, respectively. The simulated trends in 1990–2000 and 1990–2009 are $0.29 \pm 0.10 \text{ ppbv yr}^{-1}$ and $0.13 \pm 0.05 \text{ ppbv yr}^{-1}$, respectively; however, these trends are substantially larger when the model is sampled for missing data exactly as the measurements are. Simulated stratospheric ozone accounts for 79 % of the simulated 500 hPa trend between 1990 and 2000 and 100 % of the simulated trend between 1990 and 2009. Due to the importance of local meteorology and emissions at the surface it is difficult to isolate the stratospheric component of measured surface ozone variability. Overall when averaged between 30–90° N simu-

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lated surface interannual ozone trends are $0.18 \text{ ppbv yr}^{-1}$ and $0.07 \text{ ppbv yr}^{-1}$ between 1990 and 1999, and between 1990 and 2009, respectively. We have identified a number of surface sites where the measured interannual ozone variability is correlated with the 150 hPa ozone signal. Most notably these sites include the high mountain sites over Europe and Macehead, Ireland. Over Macehead the measured 150 hPa ozone signal explains 40 % of the interannual variability of the unfiltered measured ozone record. The simulated and measured ozone are highly correlated over Macehead. The Macehead measured and simulated unfiltered ozone trends between 1990 and 2000 are $0.28 (\pm 0.33)$ and $0.17 (\pm 0.13) \text{ ppbv yr}^{-1}$ respectively; between 1990 and 2009 the measured and simulated trends are $0.18 (\pm 0.11)$ and $0.08 (\pm 0.06) \text{ ppbv yr}^{-1}$, respectively. Increases in the simulated stratospheric ozone component accounts for 53 % and 75 % of the overall modeled trend for the two periods at Macehead.

1 Introduction

Tropospheric ozone is photochemically produced in situ from ozone precursor emissions or transported from the stratosphere. Tropospheric ozone impacts air quality and human health (Bell et al., 2004), atmospheric radiative forcing (Forster and Shine, 1997), and ecosystem productivity (UNEP, 2006) with resulting impacts on food (Chameides et al., 1994) and climate (Sitch et al., 2007). It also modifies the “oxidizing capacity” of the troposphere, impacting the lifetime and radiative forcing of methane (Fiore et al., 2002). Using conservative ozone-mortality epidemiologic relationships (Bell et al., 2004), even one ppbv of ozone, approximately 20 % of the surface ozone attributed to Stratosphere-Troposphere Exchange (STE) in the Northern Hemisphere (NH) mid- and high latitudes (Hess and Lamarque, 2007) may have relatively large consequences on human health (West et al., 2007).

Early semi-quantitative ozone measurements at the end of the 19th century (Volz and Kley, 1988; Marenco et al., 1994) suggest that ozone has increased 3–4 fold since the preindustrial period. Model simulations (Forster et al., 2007), although deficient in

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some details, have been able to qualitatively capture much of this remarkable ozone increase, an increase driven by the increased emissions of ozone precursors associated with industrialization. However, an explanation for the interannual trends in tropospheric ozone over the last few decades has proven more elusive despite the regulatory need to establish clear links between how changes in ozone precursor emissions and changes in climate result in changes in tropospheric ozone. The difficulty is in part due to the fact that the ozone variability results from complex and mutually dependent interactions between meteorology, ozone photochemistry, and STE. Here we examine interannual tropospheric ozone variability in the NH midlatitudes between 1990 and 2009 using analyzed ozone measurements from the stratosphere and troposphere in conjunction with a model simulation. We show that in recent years changes in the transport of ozone from the stratosphere to the troposphere can explain a considerable fraction of the interannual variability in tropospheric ozone; furthermore we show that in many locations this increased transport results in a positive trend in tropospheric ozone.

In the last two decades positive ozone trends have been found in locations both representative of the free troposphere (Tarasick et al., 2005; Zbinden et al., 2006; Thouret et al., 2006; Ordonez et al., 2007; Cooper et al., 2010) and also at surface locations with implications for air quality (Jaffe et al., 2003; Simmonds et al., 2004; Parrish et al., 2004; Carslaw, 2005; Derwent et al., 2007; Jaffe and Ray, 2007; Parrish et al., 2009); however, the trend is not positive at all locations (Oltmans et al., 2006; Lelieveld et al., 2004). Over the west coast of the US the trends have been qualitatively associated with increased Asian emissions of ozone precursors (Cooper et al., 2010). More puzzling, however, are ozone trends at sites further removed from the rapid growth in ozone precursor emissions over Asia. At Macehead Ireland between 1987 and 2003 baseline ozone trends are 0.49 ± 0.19 ppbv yr⁻¹ (Simmonds et al., 2004), although ozone appears to have leveled off in recent years (Derwent et al., 2007) yielding a 20-yr trend of 0.31 ± 0.12 ppbv yr⁻¹ between 1987 and 2007. Zbinden et al. (2006) and Thouret et al. (2006) have established trends in the tropospheric ozone column and

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network; Terao et al. (2008) found regionally specific vertical stratosphere-troposphere correlations both in measured ozonesonde data and in a model simulation; Thouret et al. (2006) found correlated anomalies in the lower stratosphere and upper troposphere from MOZAIC data; Ordonez et al. (2007) found high correlations between ozone measured at high alpine sites over Europe and lower stratospheric ozone at nearby ozonesonde stations. A number of modeling studies have also noted correlations between stratosphere-troposphere exchange and El Nino Southern Oscillation (Zeng and Pyle, 2005; Voulgarakis et al., 2011) and the Arctic Oscillation (Hess and Lamarque, 2007). From 2001–2005, Hsu and Prather (2009) find that the root mean square variability in ozone due to STE is 8 % and 13 % of the net ozone flux in the NH and Southern Hemisphere (SH), respectively.

Here we show the importance of STE in driving tropospheric ozone variability and trends on large scales using both measurements and model simulations. Between 1990 and 2009 increased STE has contributed to the positive ozone trends found in many locations. We show that this has had large impacts on the Macehead Ireland ozone record, a site frequently used to argue that baseline ozone is increasing. Section 2 describes our methodology and includes a description of the measurements and model utilized in this study. Section 3 gives an overview of simulated results. The data analysis of ozone trends and variability from the lower stratosphere to the surface is given in Sect. 4. Section 5 evaluates the model simulation against the available data. Here we also investigate the components of the large 1998–1999 ozone anomaly measured at many NH locations. Discussion and conclusions are given in Sect. 6.

2 Methodology

Measurements and model simulations of ozone are analyzed from 1990 through 2009 north of 30° N to examine the importance of the stratosphere on the tropospheric ozone budget. The measured datasets, their analysis and details on the model simulations are given in this section.

2.1 Data utilized

Several different types of measurements are used to quantify the trend and variability in ozone (Table 1) including measurements from ozonesondes and from the Measurements of Ozone and water vapour by in-service Airbus airCraft programme (MOZAIC) (Marengo et al., 1998). A limited number of surface measurements are also analyzed, although surface measurements are not emphasized in this paper. Only measurement platforms with relatively frequent, consistent and long-term measurements were selected for analysis. In addition we required that most of the base period from 1995 through 2005 be regularly sampled.

The ozonesonde data was obtained from the Ozone and Ultraviolet Data Center Environment Canada. Two types of ozonesonde are used: the electrochemical concentration cell (ECC) and the Brewer Mast (BM) bubbler. Our analysis relies on only one of the techniques at each site. Thus, while the Payerne ozonesondes switched from BM to ECC in 2002, we only analyze the record prior to 2002. The ozone record at Uccle is not included as the ozonesonde switched type in 1997, within the middle of the base period and immediately prior to the large changes in analyzed ozone (e.g., see Fig. 3). Most ozonesonde profiles are modified by a correction factor (CF) to account for discrepancies between the ozonesonde measurements and measured total ozone. However, the use of a correction factor is controversial within the troposphere (SPARC, 1988). As in Logan (1994, 1999) the CF for each sonde profile is used to provide a filter for the quality of the data. Sampling frequency for the ozonesondes was never more than 16 per month over any particular site. Detailed discussion on the ozonesonde measurements is given in SPARC (1988). The ozonesonde measurements were analyzed at 150 hPa and 500 hPa. Eighteen ozonesonde sites were selected for our data analysis (Table 1).

In addition to the ozonesonde measurements we have utilized three regional clusters of vertical ozone profiles from take-off and landing measured during the MOZAIC programme. Their selection was determined so as to include the most visited regions and

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the impact of the stratosphere than the signal at any individual measurement site. The regions are selected based on the geographic location of the measurement sites, coherency of the measurements between the different measurement sites and number of measurements. (v) For some analysis the individual regional signals are averaged to give our best representation of the overall ozone signal from 30–90° N

The individual sites within a region are averaged using either the ozone AAMD or the normalized ozone AAMD. Averaging using the normalized AAMD allows us to construct a regional signal where the variability at each station is given equal weight. Simply averaging the ozone deviations themselves is likely to weight the overall regional record towards those stations with more ozone. Each measurement location has its own mean ozone concentration, bias and standard deviation.

We consider the ozone record to be regionally robust within those regions that (i) have at least two independent measurement sites and (ii) the measurements at the different sites within each region have a high degree of correlation. Even within the regions that satisfy these general criteria we consider the ozone signal to be robust only for those years for which we can determine the annually averaged concentration at two or more measurement sites. We consider the annual signal to be sufficiently determined if ozone concentrations are reported for 10 out of 12 months.

2.2 Chemical transport model

The three-dimension chemistry transport model used in these simulations is the Community Atmosphere Model with chemistry (CAM-chem), in which chemistry from the Model of Ozone and Related Tracers version 4 (MOZART-4) (Emmons et al., 2010a) has been imported into the Community Atmosphere Model (Collins et al., 2006). In these simulations CAM-chem is driven by the meteorology from the National Center for Environmental Prediction/National Center for Atmospheric Research (NCEP/NCAR) reanalysis (Kalnay et al., 1996). CAM-chem uses the same procedure for inputting meteorological fields as has been long established through the Model of Atmospheric Transport and Chemistry (MATCH) (Mahowald et al., 1997; Rasch et al., 1997). The

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same algorithm has been incorporated into the MOZART-4 model. As the transport algorithms in CAM-chem are similar those that have been incorporated into MOZART-4, simulating chemistry within the offline version of CAM-chem represents a rather modest extension to that which has been used for years within MOZART. Recently, the MOZART model has been evaluated both globally and locally (Emmons et al., 2010a; Emmons et al., 2010b). Results from CAM-chem driven by offline meteorology have been used and evaluated in an international assessment of source sink relationships for the Hemispheric Transport of Air Pollution (HTAP) assessment study (Fiore et al., 2009; UNCEC, 2007; Sanderson et al., 2008; Shindell et al., 2008; Anenberg et al., 2009; Reidmiller et al., 2009). In particular Jonson et al. (2010) compared simulated ozone in CAM-chem against ozonesonde measurements. CAM-chem, driven by NCEP/NCAR reanalysis meteorology, has also been shown to simulate the interannual ozone variability associated with the Arctic Oscillation, which in many locations is driven by the interannual variability in STE (Hess and Lamarque, 2007). Tropospheric emissions are interannually constant allowing no interannual variability in ozone precursor emissions. Emissions are largely based on the POET emission inventory and are reported in Fiore et al. (2009). The model simulations has two years of spinup prior to 1990.

A unique tagging methodology is used to calculate the portion of tropospheric ozone that can be attributed to transport from the stratosphere. In this methodology emitted odd nitrogen is labeled and subsequently tracked as the nitrogen is physically transported and chemically modified (Hess and Lamarque, 2007). In these simulations we tag all sources of tropospheric odd nitrogen emissions including both natural and anthropogenic emissions. Tropospheric photochemical ozone is defined as that portion of ozone produced through the tropospheric photochemical reactions involving the tagged nitrogen species. Tropospheric photochemical ozone is also destroyed through the simulated photochemistry. By tagging tropospheric photochemical ozone we explicitly quantify the portion of ozone in the troposphere that originates from the stratosphere: it is simply the difference between the total simulated ozone and tropospheric

photochemical ozone. Within dilute biomass burning plumes far from local emission sources, this method of tagging ozone concentrations is equivalent to that estimated from turning off the NO_x emissions in the first place (Pfister et al., 2006).

Stratospheric ozone is parameterized using a methodology based on that used in McLinden et al. (2000). In the original methodology the production (Tg/yr) of a synthetic ozone (Synoz) tracer is specified within the equatorial stratosphere to equal the estimated cross-tropopause stratospheric flux of ozone to the troposphere (McLinden et al., 2000). Within the troposphere Synoz is destroyed. Tropospheric ozone is simply set to the concentration of Synoz at the tropopause. Here, instead of specifying the stratospheric production of Synoz, we specify the concentration of Synoz within the stratospheric source region. The concentrations are obtained from a previous equilibrated simulation where the production of synoz was specified. In a test year, either specifying the concentration of Synoz or specifying the production of Synoz resulted in the same distributions of both ozone and Synoz. However, specifying the concentration of Synoz allows the interannual variability due to variations in the atmospheric circulation to be simulated: by specifying the concentration there is no constraint on the amount of Synoz that can be transported from the stratosphere to the troposphere. In the current configuration CAM-chem does not does capture the variability due interannual variability in stratospheric chemistry.

3 Simulation overview: average ozone variability north of 30°N

Tropospheric ozone, its component produced through NO_x catalyzed tropospheric chemistry, and its component transported from the stratosphere, are calculated in CAM-chem between 1990 and 2009. Figure 1 gives the time evolution of the ozone AAMD for these various ozone components. The components are calculated north 30°N for the ozone column below 300 hPa, the 500 hPa ozone concentration concentration and ozone column integrated over the first 100 hPa above the surface.

The time evolution of each ozone component is remarkably similar irrespective of

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the level it is calculated on. The stratospheric component of ozone increases throughout the record; however, after approximately 1999 the increase is compensated by a decrease in the tropospheric photochemical component of ozone. As a result tropospheric ozone increases rather rapidly between 1990 and 1999, reaches a peak in 1999 and then levels off. An analysis of these results allows us to reach the following conclusions. (i) There is a remarkable degree of vertical homogeneity throughout the troposphere for ozone variations calculated on large spatial scales and long timescales. This suggests these variations are communicated throughout the ozone column. (ii) The simulated interannual variability of NH ozone north 30° N is largely explained by its stratospheric component. The latter explains over 81 % of the variance of the area-averaged ozone at 500 hPa and 77 % of the area-averaged variance in the surface layer. (iii) Much less variability is explained by the stratospheric contribution when sampled at the particular measurement sites given in Table 1. On average the stratosphere explains only 50 % of the variance when evaluated at any particular site at 500 hPa and 25 % of the variance at any particular site at the surface. The simulation suggests that averaging over many sites isolates the common source of variability due to the stratosphere. An explanation of the variability at any single site must invoke more complex mechanisms. (iv) Ozone averaged over all the selected 500 hPa and surface sites in Table 1 gives a good representation of the overall variations in ozone north of 30° N. The correlation between simulated ozone averaged over all the surface sites in Table 1 and surface ozone averaged north of 30° N is 0.78; the respective correlation for the 500 hPa sites is 0.97. Thus averaging ozone over all these sites should be a good proxy for the overall hemispheric ozone variability. (v) Ozone averaged regionally over the individual sites within a region (see Table 1) also gives a good representation of the overall variations in ozone north of 30° N at 500 hPa with the exception of Japan. The 500 hPa simulated ozone variability averaged north of 30° N explains only 32 % of the regional variability over the Japanese measurement sites; otherwise the overall averaged 500 hPa ozone variations explain more than 77 % of the variability for the Canadian, Northern European and the US regions. Over Japan ozone variabil-

ity is strongly impacted by the summer monsoon (Zbinden et al., 2006) especially at the southern sites. We conclude the variability over Japan is not representative of the large-scale variability. (vi) The simulated ozone trends north of 30° N are 0.25 ppbv yr⁻¹ at 500 hPa between 1990 and 2000, and 0.11 ppbv yr⁻¹ between 1990 and 2009. The corresponding trends at the surface are 0.18 ppbv yr⁻¹ and 0.07 ppbv yr⁻¹, respectively.

We were initially concerned that the increase in the stratospheric portion of ozone throughout the record (Fig. 1) could be attributed to a poor initialization of Synoz. That is, if the initial simulated concentrations of Synoz were not in equilibrium, then an increase in the stratospheric portion of ozone could not necessarily be ascribed circulation changes. It might be simply due to an adjustment of Synoz towards equilibrium. To test this hypothesis we ran a number of sensitivity tests. In these sensitivity tests we ran a number of additional simulations of three periods: (A) 1988–1989, (B) 1989–1990 and (C) 2003–2004. These periods were chosen as representative years towards the beginning of the simulation period when one might expect the simulation to be the most out of equilibrium. In these sensitivity tests we repeated the meteorology from the original simulation year, but allowed the chemistry to evolve; thus, the initial chemical condition for each successive simulation were taken as the final condition from the previous simulation of that period. We iterated this process four times over period (A), 3 times over period (B) and 2 times over period (C). While Fig. 2 suggests the simulation rapidly reaches equilibrium after only one iteration, it was instructive to use multiple iterations for selected periods. If the stratospheric portion of the ozone were increasing because Synoz was out of equilibrium then we would expect that Synoz and the stratospheric portion of ozone would continue to increase even if the meteorology were interannually constant. However, the sensitivity tests suggest this is not the case (Fig. 2). Generally we found the portion of stratospheric ozone in the troposphere to be rather insensitive to the initial conditions: for each successive simulation over the three periods the ozone concentration on December 31 is almost identical to that in the previous simulation. Thus on the timescale of a year the stratospheric ozone within the troposphere is strongly dependent on the meteorology, and nearly independent of

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the initial conditions (within the range of initial conditions examined). This is particularly striking for the 1988–1989 period when the stratospheric ozone concentration is clearly out of balance during the initial simulation. However, within a year (i.e., 31 December 1988), the stratospheric portion of ozone rapidly equilibrates to a value that appears relatively insensitive to the initial chemical conditions.

4 Ozone variability – the observational record

In this section we examine the observation record of ozone variability from the lower stratosphere to the surface for the selected measurement sites (see Table 1) north of 30° N, between 1990 and 2009.

4.1 Stratospheric regional ozone record

We find the six sites in Canada, the five sites within Central Europe, and the three sites within Northern Europe to have regionally robust ozone records and include them in our analysis of the large-scale ozone changes at 150 hPa. Over the Eastern US and over Southern Europe only one ozonesonde measurement is available, precluding us from being able to make a regionally robust assessment of ozone variations in those regions. The correlation amongst the three Japanese sites at 150 hPa is small and insignificant (see Supplement, Table S1). We were concerned that these sites were not correlated because of a tendency for some of the sites to occasionally sample tropospheric air masses at 150 hPa, especially the more southern sites and during the summer months. However, increasing the altitude of the sampling, or examining the correlation only during the winter months did not increase the correlations between these sites. Therefore, we conclude that it is unlikely that these sites are not correlated due to tropospheric contamination. The fact that these sites are at a generally low latitude range (32°–43° N) and span a wide latitude range may be a contributing factor to their low correlations. In summary, we could not isolate a robust regionally coherent signal

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in the lower stratosphere over Japan. The six sites in Canada are positively correlated, with most of the correlations significant at the 95 % level; within the Central European and Northern European regions most of the sites are also highly correlated with each other (see Supplement, Table S1). This suggests combining the measurements within each of these regions gives a regional representation of the ozone variability. We also note that the ozone records are positively correlated at sites across these regions, in many cases with a high degree of significance (see Supplement, Table S1).

The overall stratospheric signal is taken as the average of the normalized ozone AAMD at 150 hPa from the three regionally robust signals: Canada, Northern Europe and Central Europe (Fig. 3a). The correlation between the regional ozone records for the Canadian, Northern European and Central European sites are highly significant (greater than 99 % using a 2-sided Student's T test). All regions, even those without robust signals have an ozone maximum in 1998–1999 timeframe and a pronounced dip in 2000 (Fig. 3a). An analysis of MOZAIC upper tropospheric and lower stratospheric measurements over Iceland, the Eastern US and Europe also clearly indicates a pronounced lower stratospheric ozone anomaly during 1998–1999 timeframe (Thouret et al., 2006). With the exception of Japan the long timescale variability in all regions is also similar: the ozone levels from 2001–2009 are uniformly higher than those prior to 1997, with generally increasing ozone from the early 1990s through most of the first decade of the 21st century. The regional signal from Canada and Northern Europe shows a very high degree of correlation from year to year. The signal from central Europe, while showing the same the large amplitude variability as the Canadian and Northern European signals deviates somewhat in the smaller amplitude variability. This is particularly evident after 1999. The interannual variability in the lower stratospheric ozone record can be attributed to the interplay between stratospheric dynamics and photochemistry (Shepherd, 2008). The lower stratospheric ozone minimum at 150 hPa in 1992 and 1993 (Fig. 3a) can be attributed, at least in part, to the Mt. Pinatubo eruption in June 1991 (WMO, 2003) although circulation anomalies may also play a role (Hadjinicolaou et al., 1997).

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4.2 Tropospheric regional ozone record

The normalized measured ozone AAMD for various regions at 500 hPa (see Table 1) are given in Figs. 4 and 5. The correlation between the interannual ozone variations at the 18 ozonesonde sites and at the 3 MOZAIC clusters is given in the Supplement (Table S2). Of the six regions we use only Northern Europe, the Eastern US, and Canada to construct an overall record of the 500 hPa ozone variation north of 30° N (Fig. 3b). We exclude the following regions from our overall analysis: Southern Europe because this region contains only one ozonesonde site; Central Europe because we could not isolate a regionally representative robust signal over this region; and Japan, because both the simulation and the measurements suggest the ozone variations over Japan are not well correlated with either the other measurement sites or with the hemispheric averaged ozone variations (see Fig. 4). Further discussion of the regional records is given below.

All regions show a pronounced ozone peak in 1998–1999, and a minimum in 2000 and 1997, although the minimum over Japan is rather broad (see Fig. 3b; Fig. 4 for selected regional records). Using MOZAIC-derived measurements of tropospheric ozone column Zbinden et al. (2006) also noted the 1998–1999 ozone anomaly extended to three continents. The measurements over Northern Europe and Canada show a pronounced ozone dip between 1991 and 1995; Japan does not show this dip and the ozonesonde record over the Eastern US only extends back to 1995. Oltmans et al. (1998) relates this ozone minimum at high latitudes to the impact of Pinatubo.

Within each region the individual measurement sites over the Eastern US, Northern Europe and Canada are highly correlated within their regions (see Fig. 4 and the Supplement, Table S2). This is particularly noteworthy for the Canadian sites, as these encompass a broad range of latitudes and longitudes (Table 1), from 60°–114° W and 54°–80° N. The correlation between the ozone record for the individual Canadian stations are all highly significant except between Goosebay and Resolute (see Supplement, Table S2).

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The analysis over Central Europe bears somewhat more discussion as the measurement network over Europe is particularly dense, and it is frequently used to evaluate model simulations (see Fig. 5). The ozonesonde sites within this region are generally positively correlated, but not significantly so, with comparatively large differences in the ozone record between the different sites (see Supplement, Table S2). Particularly striking is the fact that the correlation between the European ozonesonde sites and the MOZAIC cluster is generally low and frequently negative. A thorough discussion of differences in the MOZAIC and ozonesonde measurement can be found in Schnadt Poberaj et al. (2009), but the MOZAIC record should give more accurate measurements. The central European ozonesonde sites and MOZAIC are all geographically close: the spatial differences between the measurement sites are unlikely to explain their differences (Schnadt Poberaj et al., 2009). The number of profiles (14 326, or 88/month on average) in the European MOZAIC cluster during the 1994–2007 measurement period is much larger than the number of ozonesondes profiles at any particular site. For example at Hohenpeissenberg, the ozonesonde location with the most frequent measurements, 1663 profiles (10/month on average) were taken over the same period. However, the difference in measurement frequency does not seem to explain the discrepancy between the ozonesonde and the MOZAIC measurements. To show this we have constructed a record of the MOZAIC measurements taken within a one-day interval of the ozonesonde measurements at Hohenpeissenberg (not shown). Subsampling the MOZAIC record at the frequency of the ozonesonde measurements does not substantially alter the MOZAIC record (not shown) nor does it bring the two sets of measurements into better agreement. We have included one surface station, the Jungfraujoch, as a comparison in Fig. 5. While the Jungfraujoch site (3580 m) is somewhat lower than 500 hPa, the measurements at Jungfraujoch are highly correlated with the 500 hPa MOZAIC measurements over Europe ($R = 0.72$). They also highly correlated with the ozonesonde record over all of the Northern Europe sites, all of the individual Canadian sites and the MOZAIC measurements over the US (see Supplement, Table S2). The Jungfraujoch measurements are not significantly corre-

lated with any of the European ozonesonde measurement stations during the period 1990–2009.

Nevertheless, we can find the average regional ozone change over Central Europe by simply averaging all the individual ozonesonde profiles and the MOZAIC cluster (Fig. 5). This regionally averaged record is generally consistent with that found in other regions: there is an ozone minimum near 1993, 1997 and 2000, with an ozone maximum in 1999 and a broad maximum in 2003–2005. An ozone maximum from 1994 into 1996 seems to be reflected in most of the station records during this period. However, while the MOZAIC and JungfrauJoch measurements suggest the amplitude of this maximum is less than that of the 1998–1999 maximum, many of the ozonesonde sites suggest this earlier peak is the stronger of the two (i.e., Payerne, Lindenberg, Hohenpeissenberg, Debilt).

4.3 Surface ozone record

As discussed above in Sect. 3, simulated ozone suggests that the variability of surface ozone is sensitive to stratospheric input. Rapid mixing along poleward and upward sloping isentropic surfaces (see Bowman and Carrie, 2002) connects surface ozone changes with upper and middle tropospheric changes thousands of kilometers to the north. In addition, Tang et al. (2011) suggests convection may transport stratospheric ozone to the surface during NH summer. Nevertheless, at the surface local sources of ozone precursor emissions are likely to exert a profound local control on the variability, linking the variability to local-to regional emission sources and local meteorological variability. Furthermore the surface concentration of ozone transported from the stratosphere is considerably less than at 500 hPa (see Table 2). Thus any stratospheric signal at the surface will likely be difficult to detect. This is consistent with our analysis in Sect. 3 suggesting that the stratosphere has a rather small role in determining the variability of ozone at any particular surface site.

It is outside the scope of this paper to make a thorough evaluation of the impact of the stratosphere on surface sites. What we have done however is to find a number of

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surface sites that clearly show the 1998–1999 ozone anomaly (see Fig. 6). Many surface sites do not show this major ozone anomaly, in particular the many surface sites over the interior western US analyzed by Jaffe and Ray (2007). A list of the particular surface sites analyzed here is given in Table 1. These sites have diverse geographic locations and altitudes between sea level and 3580 m. With the exception of the Egbert site over Southeastern Canada, the correlation amongst these surface sites tends to be highly significant and great than 0.45 (see Supplement, Table S3). We show only one elevated site over Central Europe, the Jungfraujoch; however, the interannual correlation of ozone measured at all the elevated surface sites we examined in Central Europe (Jungfraujoch, Payerne, Rigi and Sonnblich) is highly significant (not shown). The correlation between each of these sites and Macehead (at sea level) is also highly significant, suggesting a common cause of variance with incoming air over the Atlantic. We discuss the Macehead site measurements in more detail in Sect. 5.1. Ryori, the Japanese surface site is significantly correlated with the European sites examined (see Supplement, Table S3). The Canadian site (Egbert) is positively correlated with all the other sites, although only significantly at Jungfraujoch. The ozone evolution at these sites has many of the characteristics noted previously at 500 and 150 hPa: ozone minima near 1993, ozone maxima in 1999 and generally elevated ozone levels after 1999. It is important to emphasize that we have not completed a comprehensive analysis of all surface sites, we have merely selected a few with a common signal.

4.4 Summary: the impact of the stratosphere on the troposphere

The record of normalized ozone AAMD at 500 hPa for the various examined regions is given in Fig. 3b. In addition this figure shows the overall signal averaged over those regions with a robust and representative signal (Canada, the Eastern US and Northern Europe). The similarity between ozone averaged over those regions with a robust record at 150 hPa and at 500 hPa is clearly evident: both levels have an ozone minimum in 1993, 1997 and 2000 and a maximum in 1999. We note, however, that the measured peaks in 2002 and 2004 in the overall 150 hPa ozone record are not echoed in the

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5 tropospheric record, while the 500 hPa peak in overall tropospheric ozone record in 2003 is evident in the Central European stratospheric record, but not in the Canadian or Northern European records. The common variability between the stratosphere and troposphere appears to be largely due to the impact of large-scale events: the relatively small ozone concentrations in 1993, the high concentrations in 1998–1999, and the overall increase of ozone from the 1990s to the first decade of the 21st century.

10 The average measured overall stratospheric signal (averaged over the Canadian, Northern European and Central European regions) explains 69 % of the averaged measured overall tropospheric variability at 500 hPa (averaged over the Canadian, Northern European and Eastern US regions); when examined over each of these tropospheric regions individually the overall averaged stratospheric signal explains less of the variability, between 42 and 56 % (although still significant at the 99 % level).. By averaging the 500 hPa ozone signal over distinct regions we increase the common variance and minimize the regional impacts. This acts to isolate the long-timescale, large spatial scale processes.

15 The measurements only sample the stratosphere and troposphere over a few distinct regions. To what extent are these regions and their correlation indicative of the simulated hemispheric-wide correlation (i.e., 30–90° N) between tropospheric ozone and its stratospheric component? Without additional data it is impossible to say definitively. In the stratosphere, the analyzed data come from a geographically wide range of measurements, spanning from Edmonton at 114° W and 54° N to Sodankyla at 27° E and 67° N. The correlation between the Edmonton and Sodankyla timeseries is 0.76 (Supplement, Table S2) and the correlation between the different stratospheric regions with robust data is always greater than 0.70. With the high correlation between distinct regional signals, the vertical stability of the stratosphere, and the relatively high wind speeds it might be expected that the measured stratospheric ozone anomalies are likely to be homogenized over hemispheric spatial scales. The argument that the stratospheric signal impacts the troposphere on hemispheric scales is harder to make from the measurements. Unfortunately, the Japanese tropospheric measurements do

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not appear to be representative of the larger scale tropospheric signal. In addition, the regional variability is larger in the troposphere than the stratosphere. Thus we must largely rely on the model simulation to conclude that stratospheric ozone variability impacts the troposphere on hemispheric scales. The correlation between the regional records from the regions with a robust signal at 500 hPa (the Eastern US, Canadian and Northern European regions) are in the range 0.4–0.5. The simulated correlations between these regions are in the range 0.79–0.95, significantly larger than the measured correlations. This suggests the impact of measurement noise and/or the fact that a substantial portion of regional tropospheric variability is not simulated. As discussed in connection with Fig. 1 the simulation suggests that 81 % of the 30–90° N averaged 500 hPa ozone variability is explained by the simulated stratospheric ozone component. Furthermore we find that simulated regionally averaged ozone is representative of the large-scale 30–90° N variability.

Terao et al. (2008) examined the vertical correlation of monthly ozone anomaly deviations throughout the NH. They found the vertical correlations between locations within the troposphere are geographically indistinct. This suggests rapid tropospheric mixing and is consistent with our results in Fig. 1. However, Terao et al. (2008) found the correlations between the stratosphere and troposphere to have a distinct geographical signature. It is perhaps not too surprising that on monthly timescales regional changes in the jet streams or position of cyclones may have vertically deep impacts on ozone transport within well-defined regions. In this paper, however, we are looking for correlations between stratospheric and tropospheric ozone on interannual timescales and on large spatial scales. The longer timescales examined in this study would tend to mix local ozone anomalies over a larger region. Some evidence for this can be found in Hsu and Prather (2009): within a few months they show the more localized ozone anomalies associated with stratosphere-troposphere exchange are well mixed latitudinally.

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5 Simulated interannual ozone variability

5 Simulated and measured ozone variability, trends and correlations are given in Table 2. As stratospheric ozone is parameterized in our model we have restricted our analysis to the troposphere. Calculated correlations between the model and measurements
10 (Table 2) exclude the period between 1991 and 1995 due to the influence of Pinatubo at 500 hPa, but include this period at the surface. The impact of Mt. Pinatubo on surface ozone concentrations does not appear to be large (Fig. 6). The experimental design is only intended to capture ozone variability due to meteorological influences and thus will not capture the chemical impacts of the Mt. Pinatubo eruption. However,
15 in evaluating the measured and modeled ozone trends we do include the Mt. Pinatubo period. The measured trends are likely to be somewhat impacted by Mt. Pinatubo; however, as the analysis period begins prior to Mt. Pinatubo the impact will not play a major role in the trend calculation. Model-measurement comparisons at surface sites are difficult, as the model does not simulate heterogeneities in local emissions and meteorology. At the high altitude station of the Jungfraujoch we sample the model at the elevation of the site and not in the simulated surface layer as the model is not able to resolve the actual topography. This type of sampling does not account for the fact that under certain meteorological conditions elevated sites do not measure free-tropospheric air. At Macehead we present both filtered and unfiltered measurements
20 of ozone, where the filtered measurements are those with trajectories from the clean air sector over the Atlantic Basin (Derwent et al., 2007). The filtered record is designed to represent the baseline conditions at Macehead (Derwent et al., 2007). The monthly averaged monthly model output makes it impossible to filter the simulated record for the baseline conditions at Macehead. The model output should be compared to the unfiltered record, although it is noteworthy that the baseline and unfiltered ozone records at Macehead are highly correlated.
25

Simulated ozone, assuming constant emissions, generally overestimates the measurements slightly at 500 hPa; at the surface sites the overestimate tends to be more

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substantial (Table 2). The simulated stratospheric ozone ranges from over 20 ppbv at the 500 hPa sites to approximately 1–4 ppbv at the lower altitude surface sites. The simulated ozone variance is significantly less than that observed. Note that while the interannual variability in emissions is constant, Pozzoli et al. (2011) suggest emission variability contributes little to the large-scale ozone variability. The variability of the stratospheric portion of simulated ozone accounts for well over 50 % of the total variability for all the 500 hPa regions, as well as at Jungfraujoch and at Macehead.

The simulated ozone is significantly correlated with the measured ozone record in those regions with a regionally robust measured signal at 500 hPa, i.e. Canada ($R = 0.64$), Eastern US ($R = 0.70$) and Northern Europe ($R = 0.55$) (Table 2 and Fig. 7). Note, that the observed ozone minimum in 1993–1994 attributed to Pinatubo is not captured in the simulation (Fig. 7). Over Canada, Northern Europe and the Eastern US the simulation is particularly good, although simulated ozone tends to be too low near the beginning of the period. Nevertheless, the simulation clearly captures the sharp increase in ozone between 1995 and 1999, the 1998–1999 ozone maximum, and the flattening of the ozone trend subsequent to 1999. The simulated ozone maximum in 2001 in Canada, the Eastern US, Northern and Central Europe is weakly seen in the Canadian measurements, but not in the other regions; the measured maximum in 2003 over Northern Europe and the Canada is not particularly well captured in the simulation. Averaged over the Canadian, US and Northern European regions at 500 hPa simulated stratospheric ozone explains 81 % of the ozone variability; ozone averaged over the regions with a robust measured signal at 150 hPa explains 69 % of the measured troposphere variability over these same three regions.

Over Central Europe the simulated and measured ozone are not correlated, although it is important to note that the measurement record over this region is not robust. The discrepancy between the simulation and the measurements over Central Europe is particularly poor prior to 1991, although only two ozonesonde records are analyzed prior to 1991 at this time, with a large discrepancy between the Jungfraujoch measurements and those from the ozonesondes (Fig. 5). However, even over Central Europe a

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number of the measured peak ozone events are captured by the simulation including 1996, 1999, 2004 and 2006, although there are some discrepancies in amplitude and timing.

The model simulation over Japan does not capture the measured interannual variability. The measurement sites over Japan, as previously seen from observations and the model simulation, are not representative of the northern mid-latitude ozone variability.

Simulated ozone is significantly correlated with the measurements at two out of the four surface sites examined, i.e., Macehead ($R = 0.66$) and Jungfraujoch ($R = 0.55$). The measured ozone records at other high altitude measurement sites over Europe are similar to that at the Jungfraujoch (Ordonez et al., 2007) and are not further analyzed here. The modeled and measured ozone record at all four surface sites is given in Fig. 8. For all the surface sites examined both the model and measurements show a generally increasing ozone trend throughout the 1990s. In 1999 both model and measurements have an ozone maximum, although the simulated maximum is not particularly pronounced. Both simulation and measurements show a maximum in the 2003–2004 timeframe. Over Europe the 2003 heat wave likely contributes to this maximum. Simulated ozone generally shows more normalized variability amongst these surface measurement sites than observed, with pronounced ozone minimums in 1998 and 2006. The correlation between the average simulated and measured ozone normalized AAMD over these four surface sites is highly significant (0.66).

The simulated and measured trends at 500 hPa over the two periods are all positive in each of the regions examined, but their significance is very much regionally dependent. However, the 500 hPa measured and simulated ozone trends averaged over the regions with a robust measured signal (Canada, Northern Europe and the Eastern US) are more consistent. When averaged over these regions between 1990 and 2000 or between 1990 and 2009 the measured and simulated trends are highly significant, reasonably consistent with each other and large (see Robust Signal, Table 2). Here the simulated trends are filtered as the measured trends: regions without

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two or more valid measurements on a particular date are not included in the calculation. When the simulated data is not filtered (i.e., all the data is used) the simulated trends decrease substantially; 0.29 ± 0.10 ppbv yr⁻¹ between 1990 and 2000 (with 73 % of the trend due increases in stratospheric ozone), and 0.13 ± 0.05 ppbv yr⁻¹ from 1990–2009 (with 100 % of the trend explainable by increases in stratospheric ozone). These trends should be compared to the simulated 500 hPa ozone trend north of 30° N noted in Sect. 3 ($.25$ ppbv yr⁻¹ and 0.11 ppbv yr⁻¹ during 1990–2000 and 1990–2009, respectively). Note, in any case, that both simulated and observed trends at 500 hpa have decreased during the last ten years.

At the surface measured trends are everywhere positive and significant except for the unfiltered trend at Macehead between 1990 and 2000, and the long-term trend at Egbert. The simulated surface trend is positive everywhere, but only significant at Macehead and Jungfrauoch. At the latter two locations most of the simulated trend can be traced to increases in stratospheric ozone. In general the simulated surface trends tend to underestimate the measured trends. The simulated and analyzed record at Macehead is examined in more detail in the section below.

5.1 Record at Macehead

The ozone record at Macehead has been discussed in detail in the literature and is often cited as evidence that “background” ozone trends are increasing (e.g., Parrish et al., 2009). Here we examine the simulated and measured ozone at Macehead more specifically. Figure 9 shows the measured and simulated ozone signal at Macehead. Simulated ozone is highly correlated with both the filtered and unfiltered measurement records, although the correlation with unfiltered ozone is somewhat higher (Table 2). Simulated ozone captures the 1999, 2004 ozone peaks and the 2001 and 2005 minimums, but does not capture a number of the details of the measured record between 1994 and 1997. The ozone change at Macehead can be equally well fit by assuming a linear trend (Table 2) or by assuming an ozone jump between 1997 and 1999, with low ozone values prior to this time and higher values subsequently. Ozone in the

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filtered measurement record, unfiltered measurement record and model simulation is, respectively, on average 4.1, 1.9 and 1.0 ppbv higher subsequent to 1999 than prior to 1997.

The correlations between the overall stratospheric ozone signal (Fig. 3a) and measured baseline ozone, unfiltered ozone and simulated surface ozone at Macehead (Fig. 6 and 9) are 0.73, 0.63 and 0.52, respectively. The simulated correlation between ozone and its stratospheric component at Macehead is 0.58. Each of these correlations is significant at greater than 98 %, using a 2-sided Student's T test. The Macehead ozone record is also highly correlated with ozone measured over the elevated surface sites over Europe (see Supplement, Table S3). It has been previously suggested that stratospheric variations are responsible for the ozone trends at the elevated surface sites of Jungfraujoch and Zugspitze (Ordonez et al., 2007). Eighty percent of the simulated jump in ozone between the period prior to 1997 and subsequent to 1999 is due to increases in the stratospheric component of ozone. This measurement analysis and model simulation strongly suggests that the stratosphere exerts a strong source of variability at both Macehead and the elevated surface sites over Europe.

The increasing surface ozone trends at Macehead Ireland (Simmonds et al., 2004; Carslaw, 2005; Derwent et al., 2007) have been difficult to explain as this site is far removed from Asia, with its large increases in emissions. Instead Macehead should be more directly impacted by emission decreases over the US and Europe. Calculations and simulations (Lamarque et al., 2010; Fiore et al., 2009) suggest global emission changes are not sufficient to explain the measured trends at Macehead. We find that the simulated trend at Macehead between 1990 and 2009 is $0.08 \text{ ppbv yr}^{-1}$, with 75 % of the trend due to increases in the stratospheric component of ozone. Combining this simulated trend (only due to changes in circulation) with the estimated trend at Macehead due to changes in emissions gives a trend reasonably consistent with that observed. A tagged tracer of ozone produced from 6 Tg N of NO_x emitted from East Asia (where the East Asian region is taken as defined in Fiore et al. (2009)) gives 1.3 ppbv

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of ozone over Macehead. If we assume Asian emissions of NO_x roughly double between 1990 and 2010 with emissions in 1990 of 5.5 Tg N/yr (Klimont et al., 2009), the resulting linear sensitivity gives an ozone increase of $0.06 \text{ ppbv yr}^{-1}$ over Macehead between 1990 and 2010. However, decreases in European and North American emissions will result in a compensating reduction of ozone at Macehead. Fiore et al. (2009) estimates emission decreases over Europe act to reduce ozone over Europe by $.03\text{--}.05 \text{ ppbv yr}^{-1}$. Assuming 30 % of the airmasses that reach Macehead are transported from Continental Europe (D. Derwent, personal communication, 2010). gives an estimated ozone trend at Macehead of minus 0.012 ppbv/yr (30 % of $.04 \text{ ppbv yr}^{-1}$) due to European emission reductions. Decreases in North American emissions will likely also cause ozone reductions at Macehead, but we do not have a good quantitative estimate of the amount. The resulting estimated ozone trend at Macehead due to Asian emission increases and European emission decreases is $0.048 \text{ ppbv yr}^{-1}$ between 1990 and 2009, far less than the $0.18 \text{ ppbv yr}^{-1}$ trend observed in the unfiltered ozone record. However, if we include the simulated trend due to meteorological changes ($0.08 \text{ ppbv yr}^{-1}$) we obtain an overall trend of $0.128 \text{ ppbv yr}^{-1}$. While significantly underestimating the observed trend ($0.18 \text{ ppbv yr}^{-1}$), the estimated trend is quite close given the crudeness of the calculation.

5.2 The 1998–1999 ozone anomaly

A large ozone anomaly in 1998–1999 has been previously noted in the upper troposphere and lower stratosphere (Thouret et al., 2006). The anomaly in total column tropospheric ozone has been traced to anthropogenic and stratospheric ozone perturbations (Koumoutsaris et al., 2008), while Voulgarakis et al. (2011) relates it to changes in stratospheric ozone flux following an El Nino event. In this study this anomaly is evident over all regions analyzed from the lower stratosphere to the surface. This ozone anomaly is prominent at most longitudes at 50° N and 500 hPa during the period from 1998–1999 (Fig. 10). Our analysis and a more local analysis of the simulated anomaly over Europe (Koumoutsaris et al., 2008) suggests this anomaly can be attributed to

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model simulation. However, in other locations combining the measurements resulted in a consistent and representative ozone signal including: Canada, Northern Europe and Central Europe at 150 hPa; Canada, Northern Europe and the Eastern US at 500 hPa.

The analysis presented here allows us to draw the following conclusions:

1. Ozone variability in the NH lower stratosphere (at 150 hPa and north of 30° N) between 1990 and 2009 is dominated by very low ozone concentrations associated with the Pinatubo eruption in 1993 and very high concentrations in 1998–1999. In general the ozone concentrations between 1998 and 2009 are higher than those between 1990 and 1998. Interannual variations in ozone as measured over Canada and Northern and Central Europe are highly correlated between 1990 and 2009, although there is some regional discrepancy in the smaller amplitude anomalies. We argue that the stratospheric ozone variability as measured over these latter regions is likely to be representative of the overall stratospheric signal north of 30° N. However, we have been unable to show this conclusively from the measurements.
2. The measured 500 hPa ozone signal annually averaged over Canada, Northern Europe and the Eastern US is strongly correlated with the overall measured stratospheric ozone signal, where the latter signal is determined by averaging those regions with robust regional stratospheric records (Canada, Central and Northern Europe). The measured stratospheric signal explains 69 % of the averaged tropospheric variability at 500 hPa over these three regions; simulated stratospheric ozone explains 81 % of the ozone variability over the same regions. The ozone signal between these regions is highly correlated in both the model and measurements at 500 hPa. Outside these three regions we could not determine from the measurements alone whether the stratospheric ozone signal explains a significant fraction of the 500 hPa tropospheric ozone variability north of 30° N. Unfortunately, ozone at the 500 hPa Japanese ozondesonde sites is not correlated with the ozone record over the other sites in the model or in the measurements,

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- nor is it representative of the overall simulated 500 hPa signal. However, the simulated variability within the Canadian, Northern European and the Eastern US regions is representative of the overall 500 hPa ozone variability north of 30° N. We also note that over these three regions the model and measurements are significantly correlated. Finally, Zbinden et al. (2006) found that during the years of the MOZAIC ozone record (1994–2006) total tropospheric ozone over Japan, the Eastern US and Europe are highly correlated, suggesting a hemispheric wide signal.
3. Due to strong vertical mixing there can be little doubt that the stratospheric signal detected at 500 hPa also impacts surface sites. However, at the surface the influence of local emission sources and meteorology, combined with the smaller concentration of stratospheric ozone, is likely to mask the stratospheric signal. The simulation suggests that the stratospheric signal explains 77 % of the area-averaged variance at the surface north of 30° N, but only 25 % of the variance at any particular site. While we did not extensively investigate the surface ozone record, we found a number of surface sites with a record similar to that at 500 hPa. Most notably the baseline ozone record at the Macehead site is highly correlated with the stratospheric ozone signal ($R = 0.73$), with the record at the Jungfraujoch site ($R = 0.83$), as well as at other high altitude surface ozone sites over Europe (not shown). Ordonez et al. (2007) has previously shown that the high altitude ozone record over Europe is significantly correlated with stratospheric ozone. Both the model simulations and the measurements suggest a strong stratospheric influence at the Macehead site.
4. The strong correlation between lower stratospheric (150 hPa) and middle tropospheric (500 hPa) ozone suggests stratospheric ozone trends strongly impacts tropospheric ozone trends. This correlation appears to be particularly driven by large-scale events including the Mt. Pinatubo eruption and the 1998–1999 ozone anomaly. The strong correlation between the stratosphere and tropo-

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sphere is consistent with large tropospheric ozone increases during the 1990s followed by a leveling off of ozone levels after 1999. The 500 hPa measured and simulated ozone trends averaged over the tropospheric regions with robust measurements between 1990 and 2000 are highly significant. The measured and simulated trends between 1990 and 2000 are $0.73 (\pm 0.51) \text{ ppbv yr}^{-1}$ and $0.50 (\pm 0.11) \text{ ppbv yr}^{-1}$, respectively; between 1990 and 2009 the measured and simulated trends are $0.27 (\pm 0.19)$ and $.14 (\pm 0.10) \text{ ppbv yr}^{-1}$, respectively. However, when the simulated regional trends are not filtered as the measurements, the simulated trends are substantially smaller: $0.29 \pm 0.10 \text{ ppbv yr}^{-1}$ and $0.13 \pm 0.05 \text{ ppbv yr}^{-1}$ in 1990–2000 and 1990–2009, respectively. These trends are similar to the simulated trends averaged from $30\text{--}90^\circ \text{ N}$ at 500 hPa: $0.25 \text{ ppbv yr}^{-1}$ at 500 hPa between 1990 and 2000, and $.11 \text{ ppbv yr}^{-1}$ between 1990 and 2009.

5. The strong correlation between lower stratospheric ozone and surface ozone suggests an important influence of stratospheric ozone trends on surface trends. This is particularly noteworthy at the Macehead measurement site and at the Jungfraujoch site (as shown by Ordonez et al., 2007). Measured trends at Jungfraujoch are similar to those measured at Macehead within the 95% confidence interval. The correlations between the overall measured stratospheric ozone signal and measured baseline ozone, measured unfiltered ozone and simulated surface ozone at Macehead are 0.73, 0.63 and 0.52, respectively. The simulated correlation between ozone and its stratospheric component at Macehead is 0.58. We find that the simulated trend at Macehead between 1990 and 2009 is $0.08 \text{ ppbv yr}^{-1}$, with 75% of the trend due to increases in the stratospheric component of ozone. The corresponding measured trend of unfiltered ozone is $0.18 \text{ ppbv yr}^{-1}$. Considering emission increases over Asia, we find the simulated and measured trends are roughly consistent.

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6. Simulated ozone suggests that when averaged over large regions most of the interannual ozone variability is due to the stratospheric component. While emission variability is not included in these runs, the simulations of (Pozzoli et al., 2011) suggest it contributes little to the large-scale ozone variability.

7. The model simulation suggests the input of stratospheric ozone has been continually increasing between 1990 and 2009 north of 30° N. Between 2000 and 2009 this increase is balanced by a decrease in ozone generated in the troposphere by photochemical reactions. This has led to rather dramatic ozone increases prior to 1999, and then to ozone leveling off, as has been observed over many of the regions examined. However, reanalysis winds are likely to poorly capture the residual circulation and its changes (A. Douglas, personal communication, 2011). That said the simulation does capture a number of the large-scale features observed in the measurement analysis, including the 1998–1999 ozone anomaly.

8. The simulation tends to underestimate the measured ozone variance and trends. There are a number of possible explanations for this: the simulation does not capture the very low ozone values correlated with the Pinatubo eruption and it is unlikely to fully capture stratospheric circulation anomalies due to the few stratospheric levels in the input meteorological dataset. A better quantification of the trends that can be attributed to the stratosphere will require more sophisticated simulations including better representations of stratospheric chemistry and atmospheric circulation.

In conclusion the model simulations and the measurements are consistent with interannual stratospheric ozone variability on large spatial scales driving tropospheric variability on large scales. This variability extends to the surface. The results are also consistent with non-negligible tropospheric ozone trends, both at the surface and in the free troposphere, being driven through variations in stratospheric ozone input between 1990 and 2009.

Supplement related to this article is available online at:
[http://www.atmos-chem-phys-discuss.net/11/22719/2011/
acpd-11-22719-2011-supplement.pdf](http://www.atmos-chem-phys-discuss.net/11/22719/2011/acpd-11-22719-2011-supplement.pdf).

Acknowledgements. The ozonesonde data was provided from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) operated by Environment Canada, Toronto, Ontario, Canada under the auspices of the World Meteorological Organization. The data from the surface sites were downloaded from the World Data Centre for Greenhouse Gases, and were contributed by Federal Environment Agency Austria, Swiss Federal Laboratories for Materials Testing and Research, Japan Meteorological Agency, Environment Canada. Authors greatly acknowledge the European Commission and INSU-CNRS for funding as the Airbus support and the following airliners Lufthansa, Air France, Austrian and former Sabena who have been carrying the MOZAIIC instrumentation free of charge since 1994. We would also like to thank Richard Derwent for help obtaining the data at Macehead, Ireland. This work was supported by the NSF grant AGS-1042787.

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Table 1. Measurement sites used for analysis within this paper.

Station	Region ¹	Type ²	Lon	Lat	Elev	Years ³	Use ⁴
Alert	Canada	Sonde	62° W	82° N	NA	1990–2008	ST, T
Churchill	Canada	Sonde	94° W	59° N	NA	1990–2008	ST, T
Edmonton	Canada	Sonde	114° W	54° N	NA	1990–2008	ST, T
Eureka	Canada	Sonde	85° W	80° N	NA	1992–2008	ST, T
Goosebay	Canada	Sonde	60° W	53° N	NA	1990–2008	ST, T
Resolute	Canada	Sonde	95° W	75° N	NA	1990–2008	ST, T
Egbert	Canada	Surface	80° W	44° N	253m	1990–2003	SF
Kagoshima	Japan	Sonde	131° E	32° N	NA	1990–2005	T
Sapporo	Japan	Sonde	141° E	43° N	NA	1990–2008	T
Tateno	Japan	Sonde	140° E	36° N	NA	1990–2008	T
MOZAIC ⁵	Japan	Air	NA	NA	NA	1994–2006	T
Ryori	Japan	Surface	142° E	39° N	260 m	1990–2008	SF
Wallops	US	Sonde	76° W	38° N	NA	1990–2008	T
MOZAIC ⁶	US	Air	NA	NA	NA	1994–2006	T
Ny Alesund	N. Eur	Sonde	12° E	79° N	NA	1990–2006	ST, T
Scoresbysund	N. Eur	Sonde	22° W	70° N	NA	1993–2003	ST, T
Sodankyla	N. Eur	Sonde	27° E	67° N	NA	1990–2009	ST, T
Debilt	C. Eur	Sonde	5° E	52° N	NA	1994–2002	ST, T
Hohenp ⁷	C. Eur	Sonde	11° E	48° N	NA	1990–2008	ST, T
Legionowo	C. Eur	Sonde	21° E	52° N	NA	1993–2008	ST, T
Lindenberg	C. Eur	Sonde	14° E	52° N	NA	1992–2008	ST, T
Payerne	C. Eur	Sonde	8° E	47° N	NA	1992–2002	ST, T
MOZAIC ⁸	C. Eur	Air	NA	NA	NA	1994–2008	ST, T
Jungfrauoch	C. Eur	Surface	8° E	47° N	3580 m	1990–2008	SF
Macehead ⁹	C. Eur	Surface	10° E	53° N	10 m	1990–2006	SF
Madrid	S. Eur	Sonde	4° W	40° N	NA	1994–2008	ST, T

¹ Region the measurement station is classified into, where N. Eur refers to North Europe, S. Eur refers to South Europe and C. Eur refers to Central Europe.

² Type of platform where Sonde refers to ozonesonde, Air refers to aircraft measurements and Surface refers to surface measurements. ³ Refers to the measurement years utilized in this study. ⁴ Refers to the location where the measurements are used to analyze ozone trends: ST refers to stratosphere, T refers to troposphere and SF refers to surface. ⁵ Time sorted MOZAIC ozone profiles over Tokyo, Osaka and Nagoya (3094 profiles). ⁶ Time sorted MOZAIC profiles for New York, Boston and Washington (5052 profiles). ⁷ Hohenpeissenberg ⁸ Time sorted MOZAIC profiles for Frankfurt and Munich (14326 profiles)

⁹ At the Macehead site we use both baseline data (Derwent et al., 2007) where the ozone record has been selected for incoming air over the Atlantic and the unfiltered samples which include occasional flow from Europe.

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Table 2. Measured and simulated ozone for various 500 hPa regions and surface sites as defined in Table 1. Measured sites/regions are given in bold, simulated sites/regions are given in italics. Value in parenthesis in columns 3–6 are for the stratospheric portion of simulated ozone; values in parenthesis in column 7 gives the significance of the model-measurement correlation. Model and measurement statistics are only calculated over dates for which 500 hPa regions have two or more measurements. Annually averaged ozone is used to compute the table statistics.

	Region/Site	Mean (ppbv)	Standard Deviation (ppbv)	Trend ¹ 1990–2000 (ppbv yr ⁻¹)	Trend ¹ 1990–2009 (ppbv yr ⁻¹)	Correlation ² Obs./ Model
500 hPa	Canada	54.5	3.1	0.66±0.66	0.34 ±0.19	0.64 (99%)
	<i>Canada</i>	57.7 (23.9)	0.94 (1.2)	0.28±0.09 (0.25)	0.13±0.06 (0.19)	
	C. Europe ³	59.1	1.3	0.09±0.44	0.02±0.12	0.09 (NS)
	<i>C. Europe</i>	61.2 (22.3)	0.75 (0.60)	0.22±0.14 (0.07)	0.10±0.05 (0.07)	
	N. Europe ⁴	58.1	2.6	0.61±0.66	0.33±0.26	0.55 (95%)
	<i>N. Europe</i>	57.9 (21.5)	0.8 (0.76)	0.27±0.18 (0.18)	0.14±0.08 (0.14)	
	Japan	58.3	2.3	0.45±0.59	0.18±0.22	0.18 (NS)
	<i>Japan</i>	62.8 (24.3)	0.89 (0.76)	0.17±0.26 (0.08)	0.07±0.08 (0.09)	
	E. US ⁵	57.9	2.4	1.6±1.9	0.45±0.51	0.70 (98%)
	<i>E. US</i>	62.5 (24.2)	0.82 (0.79)	0.41±0.78 (0.27)	0.12±0.19 (0.11)	
	ROBUST SIGNAL ⁶	56.2	2.7	0.73±0.51	0.27±0.19	0.67 (99%)
<i>ROBUST SIGNAL</i>	58.5 (23.1)	1.3 (1.2)	0.50±0.11 (0.24) ⁷	0.14±0.10 (0.21) ⁸		
Surface sites	Macehead (B⁹)	40.0	2.1	0.56±0.36	0.37±0.13	0.66 (99%)
	Macehead (All¹⁰)	34.6	1.4	0.28±0.33	0.18 ±0.11	0.76 (99%)
	<i>Macehead</i>	41.6 (4.9)	0.71 (0.51)	0.17±0.13 (0.09)	0.08 ±0.06 (0.06)	
	Jungfraujoch	51.3	2.6	0.76±0.52	0.32 ±0.18	0.55 (97%)
	<i>Jungfraujoch</i>	57.4 (17.5)	0.76 (.73)	0.16 (.11)	0.10±0.05 (0.10)	
	Egbert	29.4	1.5	0.36±0.30	0.17±0.23	0.0 (NS)
	<i>Egbert</i>	36.8 (1.7)	0.78 (.11)	0.07±0.19 (0.01)	0.09±0.09 (0.01)	
	Ryori	38.4	1.8	0.54±0.43	0.21±0.15	-0.18 (NS)
	<i>Ryori</i>	42.2 (4.0)	1.1 (.28)	0.13±0.4 (0.07)	0.01±.11 (0.04)	

¹ Trend range is given at the 95% significance level. Bold red gives those trends significantly different from zero at the 95% confidence level. ² Correlation is of annually averaged values. Regional correlations only include those periods when the regional measurement signal is robust: that is independent measurements exist at two or more sites within the region. Correlations exclude the period impacted by Mt. Pinatubo, 1991–1995 at 500 hPa (see text). Significance of model-measured correlation as evaluated with a 2-sided Student's T test is given in parenthesis. Bold red gives values significant at the 95% level. ³ Central Europe ⁴ Northern Europe ⁵ Eastern US ⁶ Signal averaged over those regions where the regional ozone signal is robust: Canada, Northern Europe and the Eastern US ⁷ Using all simulated data the trends are 0.29 ±0.10 (0.23)ppbv yr⁻¹ ⁸ Using all simulated data the trends are 0.13±0.05 (0.17)ppbv yr⁻¹ ⁹ Filtered baseline ozone ¹⁰ Unfiltered ozone.



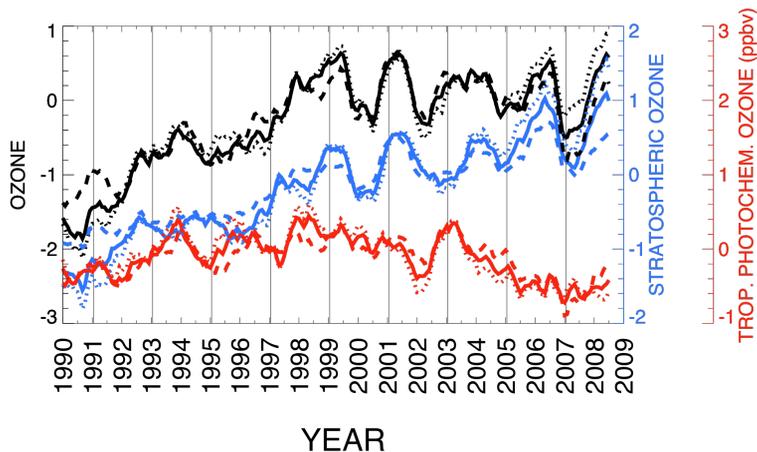


Fig. 1. 12-month running mean of simulated ozone: tropospheric photochemically produced ozone (red), ozone transported from the stratosphere (blue) and their sum (black). For each component ozone is plotted as the annual average of monthly deviations from the 1995–2005 ozone component for model gridpoints north of 30° N. Solid gives the total ozone integrated below 300 hPa, dotted the ozone at the 500 model level and dashed the surface ozone (averaged over the first 100 hPa). All vertical scales are given in ppbv.

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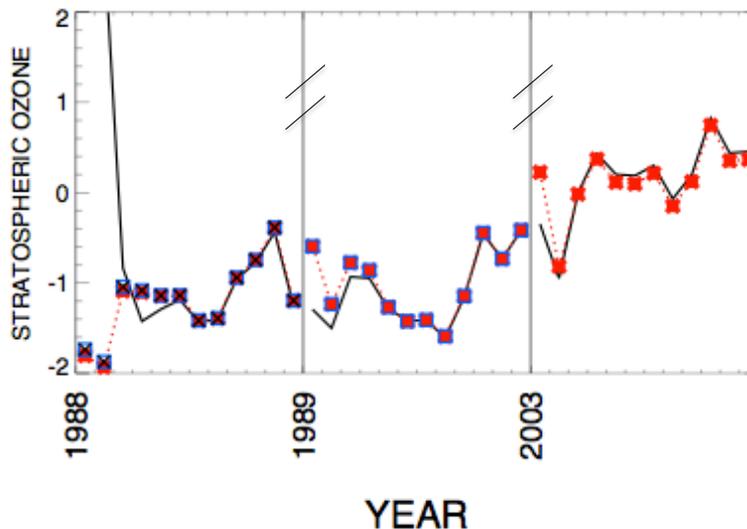


Fig. 2. Monthly averaged simulated stratospheric ozone component north of 30° N and integrated below 300 hPa for a series of 4 iterative simulations of the period 1988–1989, 3 iterative simulations of the period 1989–1990 and two iterative simulations of the period 2003–2004 (see text for details). Black line in each case gives the original and base simulation; dotted red line with red asterisks give the second simulation; blue boxes give the third simulation and crosses give the fourth simulation. The initial conditions for each simulation are taken as the final conditions from the previous simulation. In each case ozone is plotted as the monthly deviation (ppbv) from the 1995–2005 monthly mean component from the base simulation.

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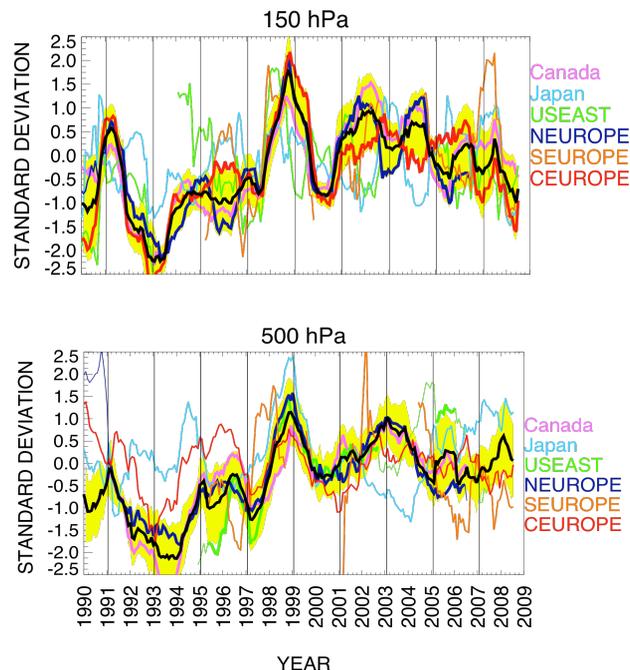


Fig. 3. 12-month running mean of regionally averaged (a) ozonesonde measurements at 150 hPa and (b) ozonesondes and MOZAIC measurements at 500 hPa. Ozone is plotted as the normalized annually averaged monthly deviation with respect to the 1995–2005 period for each region as given in Table 1. Thick lines indicate those regions with robust and representative signals: Canada, Northern Europe and Central Europe at 150 hPa; Canada, Northern Europe and the Eastern US at 500 hPa. An overall ozone average is taken about the regionally representative signals (black thick line) with the yellow shaded region representing two standard deviations of the individual records that comprise the overall signal.

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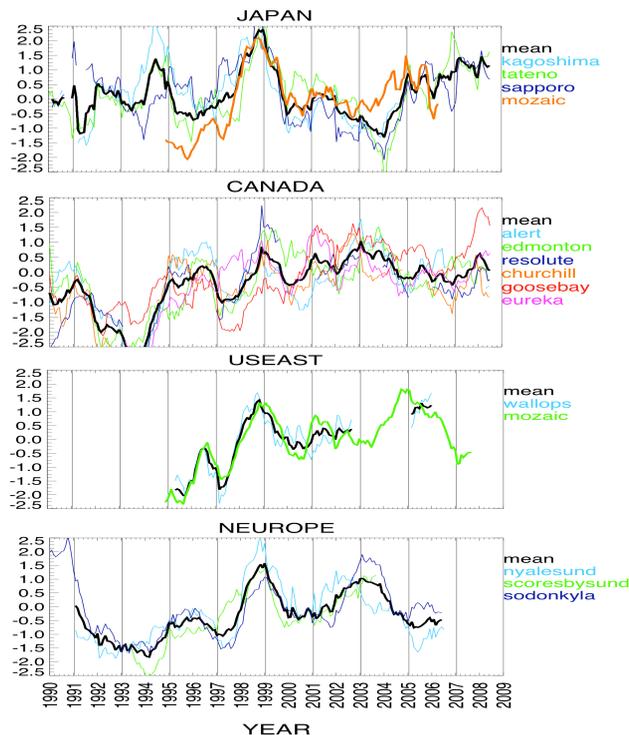


Fig. 4. 12-month running mean of 500 hPa analyzed ozone measurements for the following regions: Japan, Canada, US East and Northern Europe. Measurements from each individual ozonesonde site and MOZAIC are shown. Over each region the mean of the measurements is given in black when sufficient measurements are available for at least two measurement sites to report an annually averaged concentration. Ozone is plotted as the normalized annually averaged monthly deviation with respect to the 1995–2005 period.

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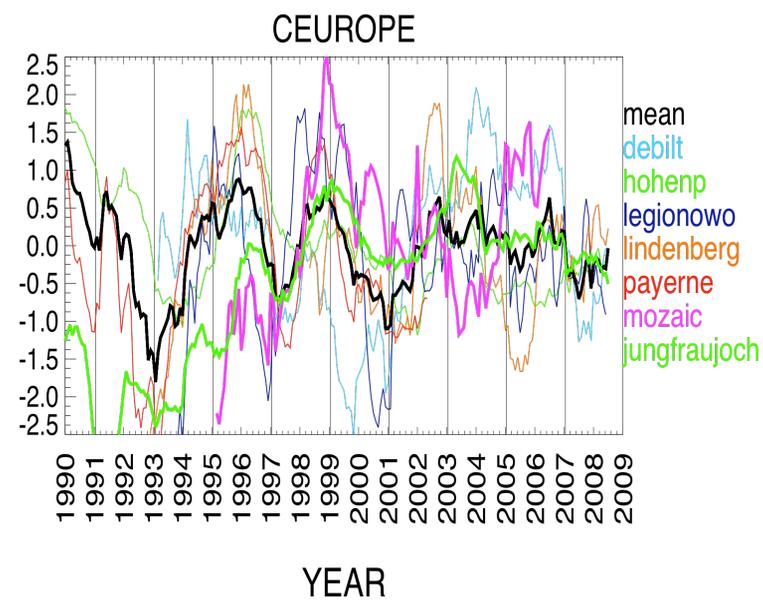


Fig. 5. As in Fig. 4 but for the central European region. Surface measurements at Jungfraujoch are shown for comparison.

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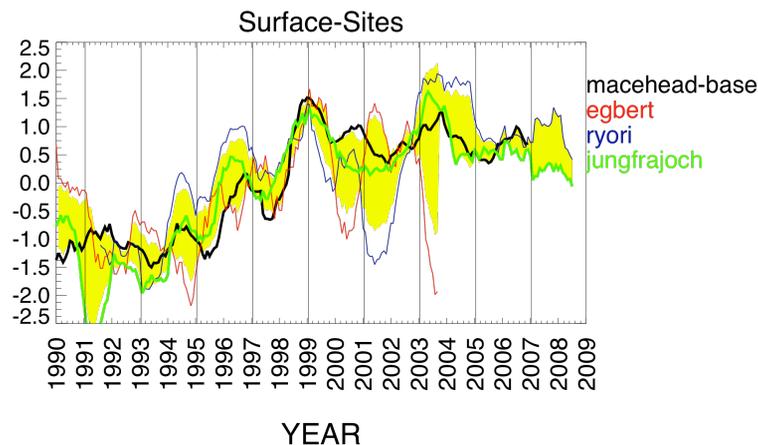


Fig. 6. 12-month running mean of the surface observational ozone record at: Macehead Ireland (10 m) ; Egbert Canada (253m); Ryori, Japan (260 m), and the Jungfraujoch Switzerland (3580 m). The Macehead data is shown filtered for clean air (Derwent et al., 2007). Ozone is plotted as the normalized annually averaged monthly deviation with respect to the 1995–2005 period. Yellow gives the regional variability about the measured record. The width of the yellow region represents two standard deviations of the regional records about their overall mean.

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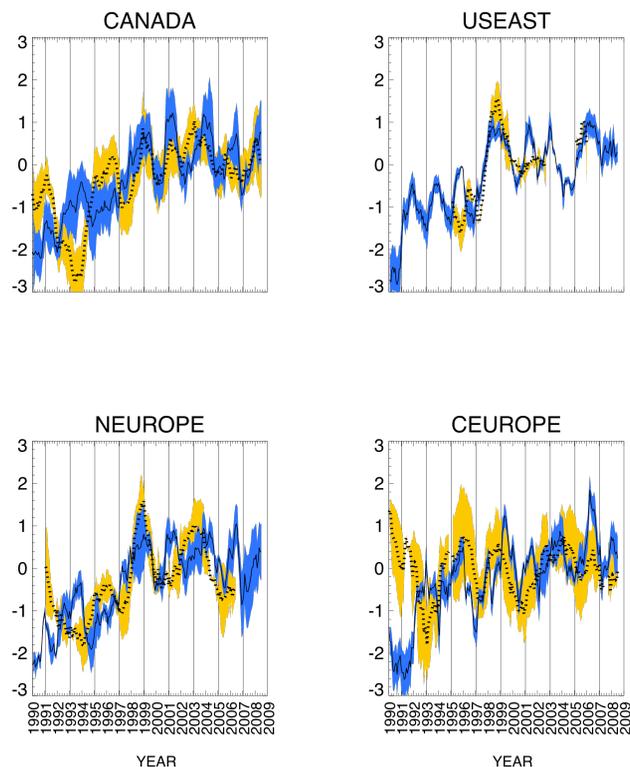


Fig. 7. 12-month running mean of 500 hPa simulated and measured ozone record for various regions. Ozone is plotted as the normalized annually averaged monthly deviation with respect to the 1995–2005 measured or simulated ozone. The width of the yellow (blue) shaded region represents two standard deviations of measured (simulated) ozone over the various measurement sites within each region at a given time. The dashed (solid) line gives the mean of the measurements (simulation) within the given region at any time.

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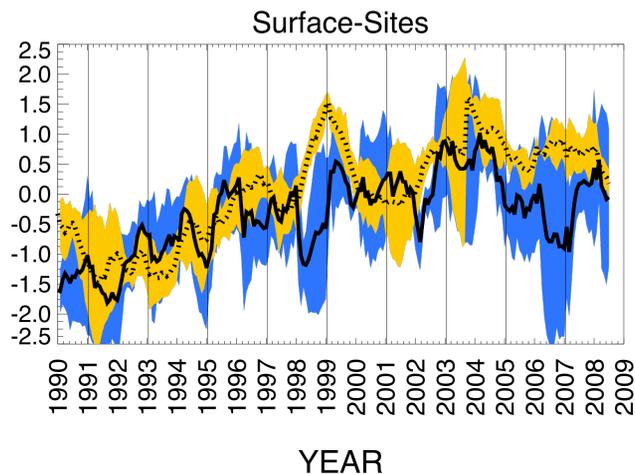


Fig. 8. 12-month running mean of simulated and analyzed ozone at the four analyzed surface sites. Ozone is plotted as the normalized annually averaged monthly deviation with respect to the 1995–2005 measured or simulated ozone. The width of the yellow (blue) shaded region represents two standard deviations of measured (simulated) ozone over the various measurement sites. The dashed (solid) line gives the mean of the measurements (simulation) at any time.

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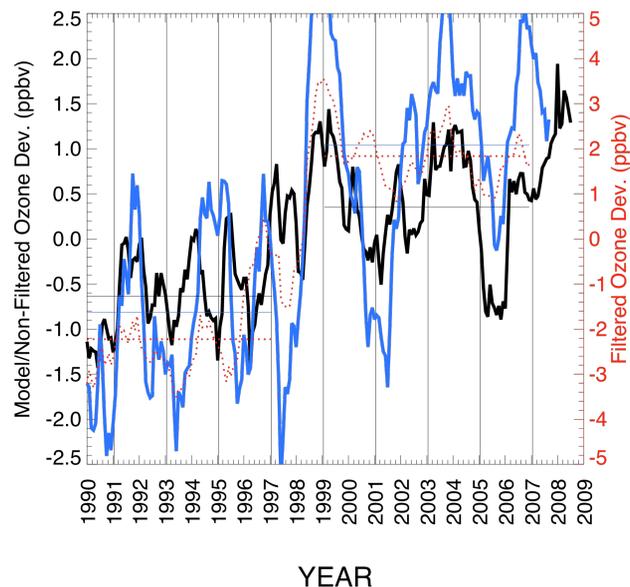


Fig. 9. 12-month running mean of measured and simulated ozone at Macehead Ireland. Ozone is plotted in terms of the annually averaged monthly deviation (ppbv) with respect to the 1995–2005 period. Simulated ozone (black), unfiltered measured ozone (blue) and filtered baseline measured ozone (red dotted) are shown. Scale for simulated and unfiltered measured ozone on left, for filtered measured ozone on the right. Horizontal lines give mean ozone values from 1989–1997, and from 1999–2007 for each of the ozone records.

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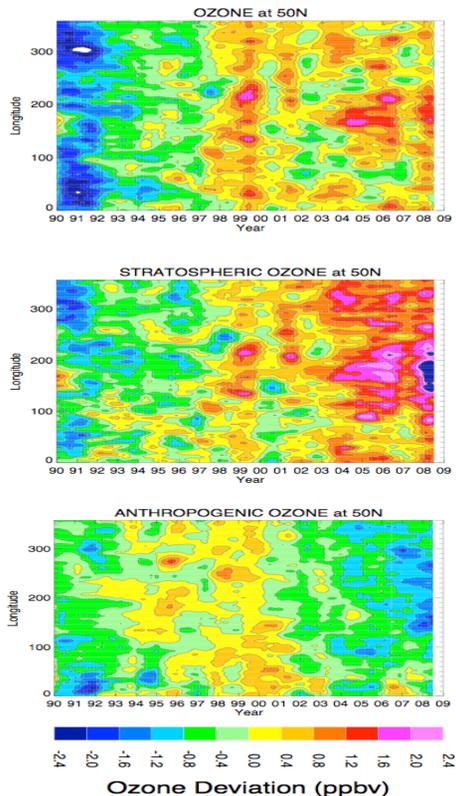


Fig. 10. Simulated smoothed 12 month running mean of ozone at 50° N and 500 hPa. Ozone is plotted in terms of the annually averaged monthly deviation (ppbv) with respect to the 1995-2005 period. Top panel shows total ozone, middle panel shows stratospheric ozone and bottom panel shows ozone produced from tropospheric photochemical reactions.

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