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Reassessment of causes of ozone column variability following the eruption of Mount Pinatubo using a nudged CCM

P. Telford[1], **P. Braesicke**[1], **O. Morgenstern**[1,2], **and J. Pyle**[1]

[1]NCAS-Chemistry-Climate, University of Cambridge, Cambridge, UK
[2]now at now at National Institute of Water and Atmospheric Research, Private Bag 50061, Omakau, Central Otago 9352, New Zealand

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Correspondence to: P. Telford (paul.telford@atm.ch.cam.ac.uk)

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Abstract

The eruption of Mount Pinatubo produced the largest loading of stratospheric sulphate aerosol in the twentieth century. This heated the tropical lower stratosphere, affecting stratospheric circulation, and provided enhanced surface area for heterogeneous chemistry. These factors combined to produce record low values of extra-polar total ozone column. Though well studied, there remains some uncertainty about the attribution of this low ozone, with contributions from both chemical and dynamical effects. We take a complementary approach to previous studies, nudging the temperature and horizontal winds in the new UKCA CCM to reproduce the atmospheric response and assess the impact on global total ozone. We then combine model runs and observations to attribute the variability to chemical and dynamical effects. To estimate the effects of increased heterogeneous chemistry we compare runs with volcanically enhanced and background surface aerosol density, noting that this causes depletion of global ozone peaking at about 7 DU in early 1993, in good agreement with observations. We subtract this effect from the observed variability and attribute the remaining variability to dynamical effects. We see that the remaining variability is dominated by the QBO. In addition to global averages we examine tropical and mid-latitude ozone, diagnosing contributions from El Niño in the tropics and some dynamically driven low ozone in northern mid-latitudes, which we see as possible evidence of changes in the QBO. We conclude that, on a global scale, the record lows of extra-polar ozone are produced by the increased heterogeneous chemistry, although there is evidence for dynamics having producing low ozone in certain regions, such as northern mid-latitudes.

1 Introduction

The eruption of Mount Pinatubo injected 20 Tg of SO_2 into the stratosphere which was, on the time-scales of weeks, converted into sulphate aerosol. At the time of maximum aerosol loading there was 30 Tg of sulphate aerosol in the stratosphere (Bluth et al.,

1992), the highest loading in the twentieth century (Robock, 2000). Following the eruption low values of ozone were observed in the lower stratosphere, particularly in winter and spring (Hofmann et al., 1994; McGee et al., 1994; Randel et al., 1995). Angell (1997) calculated reductions in the total ozone column ranging from 2% in the tropics to 7% in mid-latitudes. Ozone depletion in the aerosol cloud was higher, reaching around 20% (Grant, 1992, 1996).

Two processes involving the aerosol are implicated in the ozone reduction. The direct chemical effect was a result of the aerosol surface acting as sites for heterogeneous chemical reactions (Solomon, 1999). This locked NO_x into inactive HNO_3 (Solomon, 1999; Fahey et al., 1993), liberating chlorine from reservoirs to active species, enabling the catalytic destruction of ozone (Robock, 2000). There was also an indirect effect as a result of the aerosol altering the temperature and dynamics of the stratosphere by absorbing radiation, heating the lower stratosphere. The temperature increases in the lower and middle stratosphere were significant, with peak increases at 30 hPa of around 3.5 K (Labitzke and McCormick, 1992). This positive anomaly gradually decreased over the course of 1992 as the tropical aerosol was dispersed (McCormick et al., 1995). The heating of the stratosphere also caused increased tropical up-welling, raising air from regions with low ozone, reducing the ozone column (McCormick et al., 1995). The associated subsidence at high latitudes brought ozone down, potentially masking some of the destruction by chemical processes (Robock, 2000).

The ozone column in the Pinatubo period was also influenced by the phase of the Quasi-Biennial Oscillation (QBO) and the phase of the El Niño Southern Oscillation (ENSO). The QBO is a downward propagating variation of tropical zonal winds with a varying periodicity of around 28 months. It modulates the tropical up-welling and extra-tropical wave propagation, both affecting ozone concentration (Baldwin et al., 2001). The heating of the lower stratosphere by the aerosols produced by Pinatubo affected the QBO, "locking" the phase of the QBO for several months (Thomas et al., 2009). The years 1991 and 1992 had pronounced El Niño conditions as indicated by the multivariate ENSO index (Wolter and Timlin, 1993). El Niño affects tropical ozone,

by changing circulation patterns and cooling the stratosphere. Much of the variation relates to changes in longitudinal patterns and is removed by considering zonal means, but there is a residual variation in the tropical UTLS (Shiotani and Hasebe, 1994; Logan et al., 2003; Zeng and Pyle, 2005; Pyle et al., 2005). In the extra-tropics Randel and Cobb (1994) note some variation but confined to the Pacific and Shibata and Deushi (2008) model variability in profiles, though much seems to cancel when ozone columns are considered.

The large perturbation to the Earth's climate produced by the eruption has been the subject of many model studies, either focussing on the period itself (Bekki and Pyle, 1994; Rosenfield et al., 1997; Al-Saadi et al., 2001; Thomas et al., 2008, 2009) or as part of simulations of recent ozone variations (Chipperfield, 1999; Dameris et al., 2005; Stolarski et al., 2006; Fleming et al., 2007; Shibata and Deushi, 2008).

Detailed studies of the ozone chemistry around the time of Pinatubo (Solomon et al., 1996; Rosenfield et al., 1997; Chipperfield, 1999) indicated that the main impact arose from the increase in surface aerosol density. Zonally averaged at 43° N these studies modelled depletion of 2–3%, with larger depletion, up to 10%, at higher latitudes in spring. Solomon et al. (1996) attributed this to changes in the NO_x/NO_y abundance in the lower stratosphere enhancing the ClO/Cl_y ratio. They also noted that this depletion underestimated that observed by 50% and attributed the remainder to dynamical causes. Rosenfield et al. (1997) examined the impact of changes in aerosol heating rates on ozone columns. They diagnosed depletion in the tropics, around 2%, immediately after the eruption with a small increase later in mid-latitudes. They also noted that changes in photolysis rates arising from increased optical depths had very little impact. The importance of dynamical effects was raised by Hadjinicolaou et al. (1997) who used a CTM forced by ECMWF analyses in the 1990s. Ignoring the chemical effects of the eruption their model still produced low ozone after the eruption. However they focussed their study on northern mid-latitudes.

The impacts of the eruption have also been considered in studies of recent variability of ozone, in addition to the QBO, Cl loading and solar variability (Dameris et al.,

2005; Stolarski et al., 2006; Fleming et al., 2007; Shibata and Deushi, 2008). Stolarski et al. (2006) used a statistical model to determine the importance of these different terms in modelled and observed ozone. They showed that the eruption led to a peak reduction in global ozone[1] of 6.7±1.1 DU, with the largest effect observed in the northern mid-latitudes. Their modelled response is smaller (4.0±1.1 DU) and predominantly in southern mid-latitudes. Fleming et al. (2007) note similar features, observing little effect in southern mid-latitudes in the data in contrast to significant modelled effects there. Dameris et al. (2005) looked at the impact of the eruption as part of a long term CCM study. They modelled a 20% increase in the vertical ascent rate leading to a 5% decrease in column ozone in the tropics.

Studies of the effects of Mt. Pinatubo have acquired new relevance after Crutzen (2006) and Wigley (2006) suggested that stratospheric sulphate aerosols could be used to mitigate global warming. Trenberth and Dai (2007) noted the disruption of the global hydrological cycle caused by the eruption. Tilmes et al. (2008) used a simple empirical method to look at the effect on polar ozone whilst Robock (2008) considered other widespread impacts on regional climate, the biosphere and ozone depletion and, in a more detailed study (Robock et al., 2008), predicted benefits to preserving arctic ice, but warn of possible disruptions to the African and Asian monsoons. Understanding the causes of ozone variability after the Pinatubo eruption could help inform judgements about the impacts of this geo-engineering.

Whilst the observation of low total ozone and the fact that the increased surface aerosol density caused ozone depletion are well established, the exact link of the observed low ozone to the increased surface aerosol density and other factors, such as dynamical variability, is still uncertain. Differing conclusions regarding the relative importance of chemistry and transport have been reached. It is important to resolve these uncertainties. For instance, as the concentrations of stratospheric halogens decrease,

[1]More precisely quasi-global ozone (60° N to 60° S), as used by Chipperfield et al. (2006) with high latitudes excluded due to unavailability of continuous ozone measurements during winter.

the impact of increased surface aerosol density, from volcanoes or geo-engineering schemes, on ozone is expected to decrease. However dynamical factors such as changes in stratospheric circulation, may still produce low ozone even with reduced halogen concentrations.

We use a new, complementary, approach to investigate the relative importance of different impacts of the eruption. This approach uses a nudged CCM combining many of the best aspects of the existing CTM and CCMs. We use the information of the meteorological analyses, as do CTMs, but in a framework with consistent model physics, including vertical wind as a prognostic variable in the CCM. The ability to reproduce the observed meteorology allows us to combine observations and model runs, with and without the increased surface aerosol density, to probe the different impacts of the eruption.

2 Set up

2.1 Model

We use a nudged (Telford et al., 2008) version of the UKCA model (Morgenstern et al., 2008a). It is based upon the Met Office's Unified Model (MetUM), with the chemistry provided by the stratospheric version of UKCA (Morgenstern et al., 2008b).

The configuration used here has

- a horizontal resolution of 3.75°×2.5° in longitude and latitude.
- 60 hybrid height levels in the vertical, from the surface up to a height of 84 km.
- a dynamical time-step of 30 min.

The sea surface temperatures and sea ice coverage are prescribed from the HadISST dataset (Rayner et al., 2003). The technique of nudging is used to reproduce the

atmospheric conditions over the period studied. This constrains the model horizontal winds and temperatures to ERA-40 re-analysis data (Uppala et al., 2005).

The surface aerosol density (SAD) used to calculate heterogeneous chemical reactions is prescribed from a data-set created at NASA[2] principally from the SAGE II instrument (Thomason et al., 1997), augmented by a small amount of data from the SAM II instrument (McCormick et al., 1979). The optical depth is prescribed from the data-set of Sato et al. (1993).

2.2 Data

The TOMS[3]/SBUV dataset provides the total ozone data from which we obtain information on variability during the 1990s (Bodeker et al., 2005). We attribute the causes of this variability by comparing to known drivers of variability, such as the QBO and ENSO. We use the zonal wind at 50 hPa over Singapore as a proxy index for the QBO (Naujokat, 1986). Information about the phase of the ENSO is taken from the Multivariate Ensemble Index (MEI) of Wolter and Timlin (1993).

2.3 Methodology

Due to the incomplete coverage of the TOMS/SBUV data-set we exclude all data from latitudes polewards of 60°. Averaging the remaining data produces a quasi-global (60° N to 60° S) total column ozone average, which we denote as "global ozone". In addition we average the total ozone column between 10° N to 10° S to obtain "tropical ozone" and between 30° to 60° S and N to obtain southern and northern "mid-latitude ozone", respectively. We process these averages to remove the simpler variability. This processing is done in three stages. First the linear trend is removed, then we reduce the annual cycle by removing a sinusoidal best fit, before finally the remainder is smoothed using a 6 month running average.

[2]Data-set prepared by D. Considine.

[3]http://jwocky.gsfc.nasa.gov

We performed three model runs (Table 1). Run A utilised all aspects of the model, nudging to ERA-40 and prescribing the SAD, to simulate the effects of the eruption. To isolate the impact of heterogeneous chemistry Run B was made with the SAD fixed to 1990 values, which is taken to represent background levels. This run is intended to represent the situation without the Pinatubo aerosol, although it does include the associated dynamical changes from nudging. It is initialised from Run A at the start of January 1991. Run C was made without nudging to investigate how well the free running model performs.

First, we compare the global total ozone data between Runs A, B and C to assess whether the runs provide a reasonable representation of the observed variability. We define the chemical variability of ozone to be that caused by the changes in SAD after the eruption. This is evaluated from the differences in the ozone column between Runs A (O_3^A) and B (O_3^B). Runs A and B are nudged to the same meteorological fields so that the difference in ozone between the two runs can be attributed to the change in SAD. This modelled chemical variability ($\Delta O_3^{chem} \equiv O_3^A - O_3^B$) is then subtracted from the observed ozone record (ΔO_3^{obs}). The remaining ozone residual ($\Delta O_3^{dyn} \equiv \Delta O_3^{obs} - \Delta O_3^{chem}$) should then correspond to the dynamical variability if we have successfully removed the chemical variability. We repeat the procedure for tropical and northern and southern mid-latitude ozone.

3 Results

Firstly we compare the global average ozone column between the model runs and the unprocessed data to assess the model's ability to reproduce the observed variability. We proceed to apply the methods described in Sect. 2.3 to distinguish chemical and dynamical changes for global (Sect. 3.2), tropical (Sect. 3.3) and mid-latitude ozone (Sect. 3.4).

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3.1 Validation of UKCA global ozone column

Figure 1 shows the global average ozone column in the data and the model runs. For consistency the model ozone is treated identically to the data, with latitudes polewards of 60° being excluded. As the global average ozone column in all model runs shows a slight high bias, we compare deviations from the mean rather than average values.

The combination of meteorology nudged towards re-analysis data and a simple prescription of increased surface aerosol density used in Run A produces a good representation of global ozone variability. Given that TOMS/SBUV ozone data is used in the ERA-40 assimilation over this period (Dethof and Holm, 2004) this good correspondence may be expected even though we neglect other factors, such as changes in chlorine loading and solar UV flux. This may reflect that these factors do not vary much over this period (Eyring et al., 2006; Fleming et al., 2007), or simply that the factors included in our model suffice.

Run B, with background SAD values, is able to reproduce much of the observed variability. However, as expected, it overestimates ozone in the years immediately after the eruption. The free running model, Run C, is also able to capture many of the features of the ozone variability, with some decrease after the eruption and large ozone loss in 1994/95. However there are periods, such as 1993 and 1997, where significant differences can be seen. One explanation for these discrepancies is that the phase and periodicity of the modelled QBO do not match those observed (Morgenstern et al., 2008b).

3.2 Attribution of variability in global ozone

We apply the method described in Sect. 2.3 to evaluate the chemical effect (ΔO_3^{chem}) and the residual, dynamical variability (ΔO_3^{dyn}). These are plotted in Fig. 2. In addition we regress the residual ozone variability to our QBO proxy.

The chemical effect of the eruption is clear, with ozone depletion increasing as the sulphate cloud spreads. This loss peaks at 7.2 DU around the start of 1993, then de-

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creases in magnitude as the sulphate is removed from the stratosphere. By fitting this variation using a linear increase then a linear decrease we note that the initial ozone depletion was at a rate of 5.7±0.2 DU/year with the recovery at a rate of 1.8±0.1 DU/year. The magnitude and timing of the effect is in good agreement with that obtained by statistical methods, with Stolarski et al. (2006) determining the effect to peak in 1993 at 6.7±1.1 DU.

To consider our sensitivity to the prescribed SAD we have used an alternative data-set from the CCMVal project based on that compiled by SPARC (Thomason and Peter, 2006). This data-set has finer vertical and latitudinal resolution, and if the model is sensitive to the exact distribution of SAD could produce different values of total ozone. The magnitude of the ozone loss using this data-set is only 0.2 DU smaller, indicating that with such large increases in SAD, its exact treatment is relatively unimportant, which may well explain why our coarsely gridded SAD data is able to reproduce the observed ozone depletion. The timing is slightly different, with the rate of ozone recovery being slightly slower. The initial increase caused by heterogeneous chemistry shown in Fig. 2 is also less pronounced, which is attributed to the smaller vertical extent of the aerosol having a smaller impact on NO_x chemistry.

We subtract the chemical variation (ΔO_3^{chem}) from the observed variability as described in Sect. 2.3 to obtain the residual, dynamical variability (ΔO_3^{dyn}). This residual exhibits a quasi biennial variation. We superimpose our QBO proxy index, the 50 hPa zonal wind at Singapore, with a 7 month time lag to account for time to propagate to the extra-tropics. The dynamical variability and our QBO proxy correlate well, with a correlation coefficient greater than 0.8. A similar correlation is obtained if a lag of 8 months rather than 7 is used. Evidently the remaining variability is attributable predominantly (more than 65%) to the QBO. The presence of this correlation also increases our confidence in the reliability of the subtraction of the modelled chemical variability, as any residual chemical signal would be unlikely to correlate with the QBO. By using time lagged correlations we also conclude that there is no evidence that the ENSO has any large impact on the remaining variability.

Comparing these effects we conclude that the main cause of the observed low global ozone after Pinatubo was the increased surface aerosol density. However dynamics may have a greater effect on regional scales. To investigate whether this is the case we examine three regions separately: the tropics and northern and southern mid-latitudes.

3.3 Tropical ozone column

The QBO is a tropical phenomenon. In light of its importance in the global residual it is interesting to see its role in the modelled tropical ozone column. So we repeat the procedure used to study the global ozone for the tropics. The resulting contributions from chemical and dynamical factors are shown in Fig. 3.

The contribution from the increased SAD is smaller, peaking at around −3 DU, than in the global case, but the rates of change are similar, decreasing at 5.1±0.2 DU/year before recovering at 1.9±0.1 DU/year.

As in the global case the residual, "dynamical", variability correlates well with our QBO proxy index, with a correlation coefficient greater than 0.8. The lag used is different, and the proxy is inverted, which reflects the different influence the QBO has on tropical and global ozone. After removing the variability associated with the QBO we still see low ozone at the time of the Pinatubo eruption. The reduction, around 5 DU, occurs before the eruption and coincides with the 1991/1992 El Niño. When we correlate this variability with our ENSO proxy we produce a correlation coefficient of 0.45, significant at the 95% level. The ability to produce this correlation without a lag suggests a more immediate impact of ENSO on tropical ozone column. We see no evidence of unusually low ozone in the dynamical variation after the eruption, although this may be due to our technique being insensitive to the small changes or because this signal is inseparable from the QBO variability. The significant correlation between our dynamical change and our physical proxies increases confidence that our technique still performs well even on regional scales.

In the tropics the chemical depletion is smaller than the dynamical variability, which is still attributable to the QBO, although with a significant contribution from ENSO. We

now investigate the mid-latitudes where both the chemical effects are larger and where previous studies (Solomon et al., 1996; Hadjinicolaou et al., 1997) have argued for an important dynamical contribution to ozone variability.

3.4 Mid-latitude ozone columns

The mid-latitudes are the regions where large chemical ozone depletion (Chipperfield, 1999) and dynamically induced low ozone (Hadjinicolaou et al., 1997) have been modelled after the eruption. To investigate whether we see such effects we repeat the procedure used to study the global and tropical ozone for the northern and southern mid-latitudes. The resulting contributions from chemical and dynamical factors are shown in Fig. 4.

Ozone depletion due to increased aerosol surface is larger than that at lower latitudes, peaking around 10 DU in both the Northern and Southern Hemispheres (Fig. 4). The variation of the depletion over time is less smooth, showing a large annual cycle, with the heterogeneous depletion peaking in spring. The magnitude of the depletion is similar in both hemispheres, in agreement with the studies of Fleming et al. (2007) and Stolarski et al. (2006). When we subtract the chemical variability from the observations to produce the "dynamical" variability we see differences between the northern and southern mid-latitudes. This is in agreement with the observed ozone variability, as seen by Bodeker et al. (2001), Stolarski et al. (2006) and Fleming et al. (2007), which had lower values in northern mid-latitudes than in southern.

In the Southern Hemisphere the amplitude of ozone inter-annual variability is constant throughout the period. We note that away from the eruption there appears to be some correlation with our QBO proxy, with none in the years immediately after the eruption. In the Northern Hemisphere the variability looks similar to that in the Southern in the early and later years. However in 1993 and 1995 we diagnose low ozone, which appears to correlate well with our QBO proxy.

This additional dynamically driven low ozone in the northern mid-latitudes can explain the discrepancy observed between the chemical impact and observations

by Solomon et al. (1996) and others. In northern mid-latitudes the dynamically induced low ozone is also considerable larger than the chemical depletion, in agreement with the results of Hadjinicolaou et al. (1997). Interestingly during the years after the eruption better correlation is obtained using a lag of 5 months, as opposed to 7 months in the other years, suggesting a qualitative difference in the relationship between the QBO and mid-latitude ozone after the eruption of Mt. Pinatubo. This may be due to impacts of the eruption on the QBO, such as phase-locking, as seen by Thomas et al. (2009) and others.

4 Conclusions

We use the new UKCA CCM to study the ozone variability around the time of the eruption of Mount Pinatubo. We "nudge" the model to reproduce the observed dynamical variability and investigate the impacts of increased heterogeneous chemistry by comparing results of runs with and without increased surface aerosol density. We subtract this effect from the observed variability to obtain the impacts of "dynamical" variability. The record low global ozone is mainly a result of the increased heterogeneous chemistry, but augmented by some, regional, dynamical effects, most notably in northern mid-latitudes.

The increased heterogeneous chemistry causes global ozone depletion, peaking at around 7 DU in early 1993, in good agreement with observations. The global dynamical variability is smaller than the chemical depletion and, by comparing with a QBO proxy, can be attributed to being predominantly caused by the QBO. On global scales we see no evidence of contributions from ENSO or direct dynamical impacts of the eruption. In the tropics the chemical depletion is smaller than the dynamical variability, which is still attributable to the QBO, although with a significant contribution from ENSO.

In both northern and southern mid-latitudes the heterogeneous chemistry causes ozone depletion, peaking around 10 DU in late 1992 and early 1993. In addition there is dynamical variability which correlates with the QBO, though this variation differs be-

tween the north and south. In the north the dynamical driven variability is greater, producing reductions in ozone columns even larger than the chemical depletion, in agreement with Hadjinicolaou et al. (1997). We speculate that this low ozone arises from qualitative differences in the QBO after the eruption.

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Table 1. Summary of runs made in the model.

Run name	Dynamics	Surface Aerosol
Best Guess (A)	nudged (1988–1998)	"pinatubo" (1988–1998)
Background (B)	nudged (1991–1998)	background (1990)
Free (C)	free (1988–1998)	"pinatubo" (1988–1998)

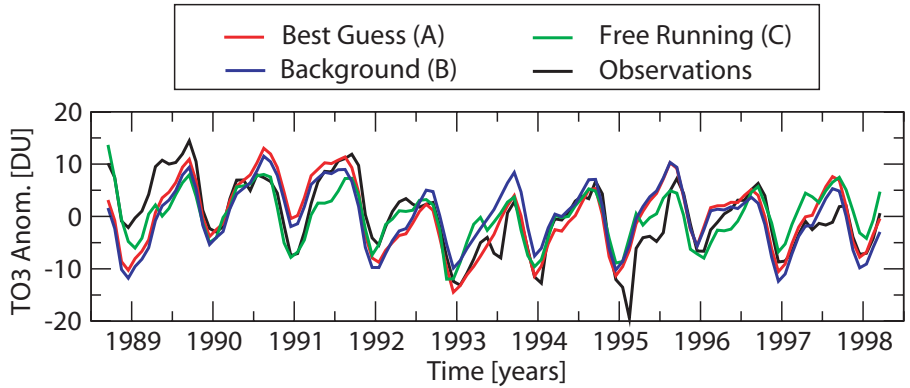

Fig. 1. Average global ozone column anomaly as a function of time in the data and in the three model runs.

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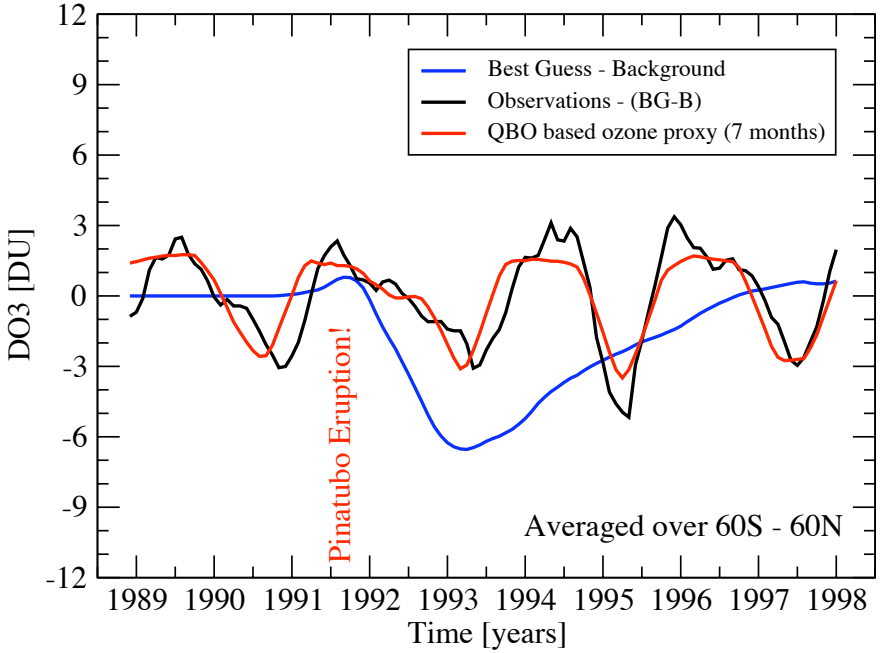

Fig. 2. Contribution to global ozone variability from chemical (ΔO_3^{chem}, blue line) and from dynamical effects (ΔO_3^{dyn}, black line). The chemical effects are evaluated from the differences between the best guess run (Run A, BG) and that with background SAD (Run B). The dynamical effects are evaluated by subtracting this difference (BG-B) from the observations. A regression of ΔO_3^{dyn} to the QBO proxy, with a lag of 7 months, is superimposed for comparison (red line).

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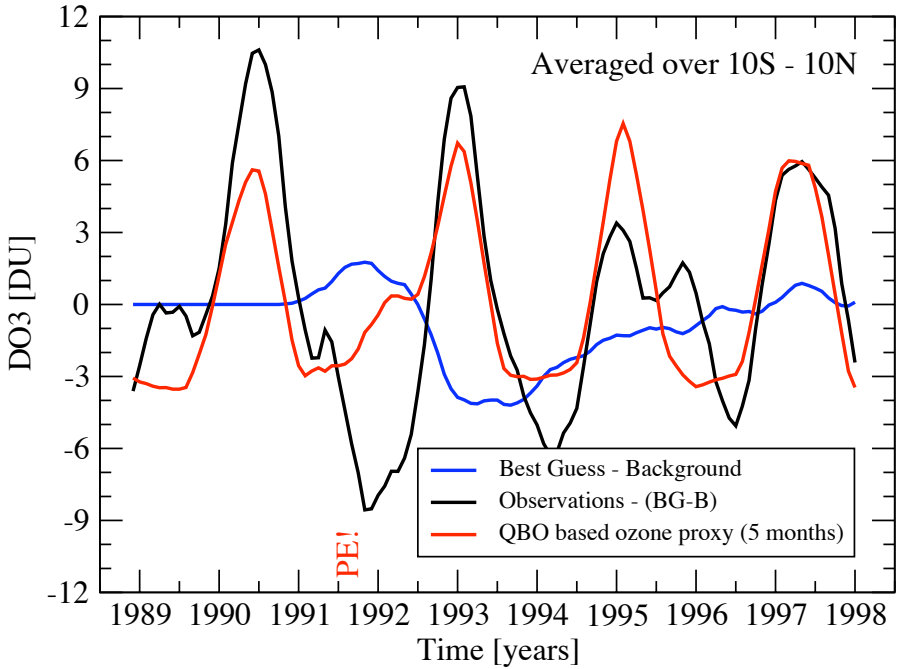

Fig. 3. Contribution to tropical (10° S–10° N) variability from chemical (ΔO_3^{chem}, blue line) and from dynamical effects (ΔO_3^{dyn}, black line). The chemical effects are evaluated from the differences between the best guess run (Run A, BG) and that with background SAD (Run B). The dynamical effects are evaluated by subtracting this difference (BGB) from the observations. A regression of ΔO_3^{dyn} to the QBO proxy, with a lag of 5 months, is superimposed for comparison (red line). The timing of the Pinatubo eruption is marked by "PE".

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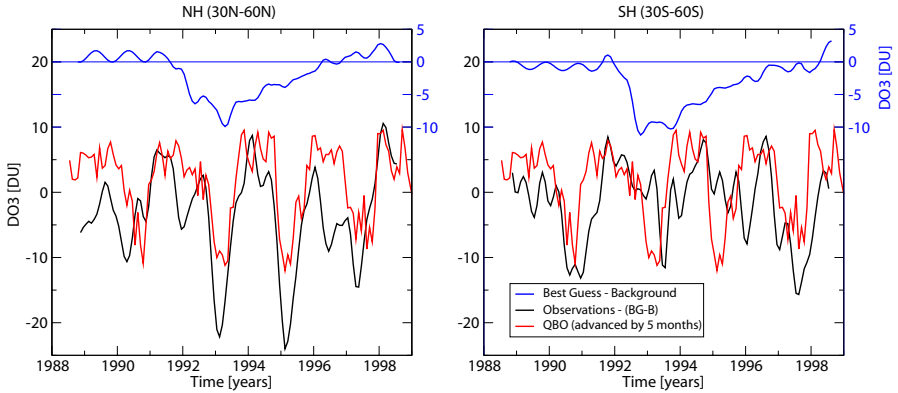

Fig. 4. Contribution to northern (left) and southern (right) mid-latitudes (30°–60°) ozone variability from chemical (ΔO_3^{chem}, blue line) and from dynamical effects (ΔO_3^{dyn}, black line). The chemical effects are evaluated from the differences between the best guess run (Run A, BG) and that with background SAD (Run B). The dynamical effects are evaluated by subtracting this difference (BGB) from the observations. A regression of ΔO_3^{dyn} to the QBO proxy, with a lag of 5 months, is superimposed for comparison (red line).