INTERANNUAL VARIATIONS AND TRENDS OF TOTAL OZONE AT NORTHERN MIDLATITUDES: CORRELATION WITH STRATOSPHERIC EP FLUX AND POTENTIAL VORTICITY

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1. INTRODUCTION

It is well established that column ozone amounts at middle latitudes have been decreasing for several decades (WMO, 1999; 2003). Analyses of ozonesonde data have shown that the implied ozone decreases have occurred primarily in the lower stratosphere (~ 15 to 24 km altitude) (Logan, 1994; SPARC, 1998). However, a detailed attribution of these decreases to specific causal mechanisms has not yet been achieved (WMO, 2003). Recently, it has become clear that negative ozone trends at northern middle latitudes are not actually linear: While linear trends were approximately constant and strongly negative over the period from 1980 to 1993, a negligible or slightly positive trend has prevailed at many locations since that time (e.g., Fioletov et al., 2002; Steinbrecht et al., 2003). Thus, any complete interpretation of negative ozone trends at northern middle latitudes must explain not only the overall ozone decline but also the non-linear trends.

Possible causal mechanisms for lower stratospheric ozone trends and interannual variability at northern middle latitudes can be divided into two general classes that are not fully independent of one another: Chemical dynamical. Chemical and dynamical. Chemical and processes that may contribute to midlatitude ozone decreases are related to the increase in anthropogenic halogens that is known to be the main cause of ozone declines in the polar regions and in the upper stratosphere. These include direct mechanisms (chlorine activation on liquid aerosol and ice particles; e.g., Solomon et al. (1996; 1998) and indirect mechanisms (export of ozone-depleted or chlorineenriched air from the polar vortex; e.g., Knudsen and Grooβ, 2000; Hauchecorne et al., 2002; Chipperfield, 2003). However, chemically induced ozone losses can have dynamical feedbacks. For example, increased ozone losses in polar regions can lead to radiatively reduced temperatures, which can then accelerate the circumpolar winter vortex. On the other hand, dynamical changes can influence ozone through both chemical feedbacks and direct transport. An example of chemical feedbacks is the increased Arctic chemical ozone loss that occurs during dynamically quiet conditions when the vortex is cold and isolated

(Weber et al., 2003; Rex et al., 2004). These coupled feedbacks make it difficult to determine the relative contributions of dynamics and chemistry to ozone loss.

As reviewed by Holton et al. (1995), the wintertime Brewer-Dobson circulation, which transports ozone to middle and higher latitudes in winter and spring, is driven in part by extratropical wave forcing. (It is also driven by the seasonal cycle in diabatic heating; see, e.g., Garcia, 1987.) Evidence for a contribution to midlatitude ozone trends from a long-term weakening of the Brewer-Dobson circulation was first pointed out by Fusco and Salby (1999). This trend component represents a change in the overall abundance of ozone in the stratosphere forced by a combination of dynamical transport and photochemical production and loss. Based on correlative and regression relationships using Eliassen-Palm (EP) planetary wave flux as a measure of the wave forcing, Randel et al. (2002) have estimated that this mechanism may have caused 20 to 30% of the column ozone trend at northern midlatitudes over the 1979 to 2000 period.

At northern midlatitudes in winter and spring, it is well known that both poleward and equatorward synopticscale wave breaking events occur in the lower stratosphere (e.g., Nakamura, 1994; Peters and Waugh, 1996). In the N. H., anticyclonic poleward wave events are characterized by northward deformations of the subtropical tropopause resulting in protrusions of ozonepoor, low potential vorticity (PV) air that extend from the tropical upper troposphere into the midlatitude lowermost stratosphere (cf. Holton et al., 1995). These poleward events are often complemented by equatorward wave events at other longitudes that generate tongues of ozone-rich stratospheric air extending into the tropical upper troposphere (Waugh and Polvani, 2000). The resulting meandering boundary between low ozone regions and higher ozone regions at northern midlatitudes has been referred to as the "subtropical front" by Hudson et al. (2003).

Poleward wave events produce corresponding poleward displacements of the mean subtropical jet and midlatitude storm tracks; they are most common during winters with an enhanced polar vortex (positive Northern Annular Mode, AO, and NAO indices). During these winters, the zonal wind field at midlatitudes tends to be less cyclonic implying greater numbers of anticyclonic wave events. When the waves break (McIntyre and Palmer, 1983; 1984), cut-off anticyclones often form and irreversible exchanges of air occur between the

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stratosphere and troposphere (see, e.g., Hintsa et al., 1998). However, even if the waves do not break, the monthly zonal mean ozone column at a given latitude can still be significantly modified by one or more of these non-linear events. In some cases, the ozone column is further reduced locally during poleward wave breaking events by southward excursions of the polar vortex and by upward deflection of the zonal flow over the associated upper tropospheric anticyclone, causing extreme ozone minima, sometimes referred to as ozone ``miniholes'' (e.g., Petzoldt, 1999; James et al., 2000; Hood et al., 2001; Semane et al., 2002).

There is evidence that long-term increases in poleward transient Rossby wave events in the subtropical lower stratosphere have contributed to midlatitude zonal mean ozone trends since 1979 (Hood et al., 1997; 1999; McCormack and Hood, 1997; Reid et al., 2000; Appenzeller et al., 2000; Orsolini and Limpasuvan, 2001; Br\"onnimann and Hood, 2003). These increases in poleward wave events have occurred during a period of generally increasing AO index (i.e., increased negative anomalies of pressure over the polar cap and positive anomalies at low latitudes; see Figure 1c of Gillett et al., 2002). On the basis of regression relationships between column ozone and Ertel's PV, it has been estimated that increases in poleward wave breaking frequency could explain as much as 40\% of the zonal mean column ozone trend at northern midlatitudes during the month of February over the 1979 to 1998 time period (Hood et al., 1999). Recent analyses of ozonesonde data also support the hypothesis that especially low monthly mean ozone amounts in the midlatitude lowermost stratosphere over Europe in winter tend to occur during years characterized by extreme ozone minima associated with poleward breaking Rossby waves (e.g., Logan and Megretskaia, 2004).

In this paper, empirical (correlative and regression) methods are applied to investigate further the two primary dynamical mechanisms that have been proposed as significant contributors to column ozone trends at northern midlatitudes. For a more complete description, the reader is referred to Hood and Soukharev (2005). A preprint of the latter paper is available by anonymous ftp at <u>ftp.lpl.arizona.edu</u> (cd to /pub/lpl/lon/stratosphere)..

2. OZONE INTERANNUAL VARIABILITY AND TRENDS AT NORTHERN MIDLATITUDES

We consider here an updated version (Version 8) of the merged satellite column ozone data set prepared originally at the NASA Goddard Space Flight Center (Stolarski et al., 2000). This data set combines the Nimbus 7 and Earth Probe TOMS data, the Nimbus 7 Solar Backscattered Ultraviolet (SBUV), and the NOAA 9, 11, and 14 SBUV/2 data. It is available at http://code916.gsfc.nasa.gov/Data_services/merged/.

The data set is more continuous with fewer gaps (for

example, the SBUV data were used to fill in a period during 1995 and 1996 when there were no TOMS measurements) and has been adjusted using operational overlap periods to minimize differences in calibration between the various instruments. It covers the period 1979 to 2002 (24 years).

In order to provide a brief overview of long-term variations in the merged TOMS - SBUV record, a standard multiple regression statistical model was including terms representing variability applied associated with the equatorial quasi-biennial wind oscillation (QBO), the 11-year solar cycle, and assumed linear trends. A contour map of the resulting monthly linear trend regression coefficients (in per cent per decade) is shown in Figure 1. Shaded areas indicate where trends are significant at the 2σ (95\% confidence) level. For recent detailed analyses of total ozone interannual variability and trends, see Fioletov et al. (2002) and Steinbrecht et al. (2003). An examination of Figure 1 shows that, in the N.H., maximum linear trends occur at northern midlatitudes (about 40N to 45N) in winter (February). Trends are also relatively large in the Arctic spring but, because of lack of backscattered sunlight during the polar night, these trends are not clearly shown in Figure 1.



Figure 1. Linear column ozone trends based on a standard multiple regression statistical analysis of the merged TOMS/SBUV data record as described by Stolarski et al. (2000). Shaded areas are significant at the 95% confidence level.

To illustrate the interannual and decadal variability of ozone for the month and latitudes where ozone linear trends are largest, Figure 2 plots monthly zonal mean ozone for February at latitudes ranging from 30N to 60N. Interannual variability associated with the QBO is evident. In addition, a relatively large minimum occurs in 1993; this minimum follows the June 15, 1991 eruption of Mt. Pinatubo and is caused mainly by the chemical and/or dynamical effects of this aerosol injection event (Hadjinicolaou et al., 1997; Stenchikov et al., 2002; Rozanov et al., 2002). At the latitudes plotted in Figure 2, the modified circulation (positive phase of the AO in winter during the QBO west phase) is a partial cause of the negative ozone anomaly in 1993 (G. Stenchikov, private communication, 2004). At 40N where the trend maximizes, the trend is mostly linear but is less steep (the linear trend coefficient becomes less negative)

toward the end of the record. At 60N, however, the last part of the record has no detectable trend and a possible slight upturn appears during the last seven years. As noted, for example, by Fioletov et al. (2002), the latter upturn appears to occur too rapidly to represent a recovery from chemically induced ozone depletion associated with reduced anthropogenic halogen emissions. Rather, natural climate variability with its effects on stratospheric circulation is a more likely explanation..



Figure 2. Time series of merged TOMS/SBUV ozone monthly zonal means for February at a series of latitudes in the Northern Hemisphere.

3. OZONE CHANGES CORRELATED WITH STRATOSPHERIC EP FLUX

Fusco and Salby (1999) first demonstrated a correlation between hemispherically averaged upward EP flux at 100 hPa during January and the increase of extratropically averaged total ozone during the same month for the 1979 to 1993 time period. Randel et al. (2002) extended their analysis to 2000 and found similar correlations between upward EP flux in January (averaged over the 40N to 70N latitude band) and the change in ozone during the same month at northern midlatitudes. The 40N to 70N band was used by them because EP flux maximizes within this latitude range and nearly identical results were obtained using averages over the entire hemisphere. This is considered to be a reasonable measure of the strength of the overall Brewer-Dobson circulation, which influences ozone at all latitudes in the winter hemisphere. We note that it may also be a useful proxy for wave breaking and, hence, guasi-horizontal mixing, another ozone transport mechanism, at midlatitudes.

Here we extend by two years the analysis of Randel et al. (2002) and investigate further the application of EP flux data to predict interannual variability of northern midlatitude ozone in winter. Specifically, values of the monthlly averaged meridional eddy heat flux, {v'T', which is approximately proportional to the vertical component of the EP flux, are calculated at 100 hPa based on National Centers for Environmental Prediction (NCEP) reanalysis data (Kalnay et al., 1996).

For the purpose of estimating the contribution of EP flux variations to interannual midlatitude ozone variations in winter, regression relationships between monthly mean EP flux and the column ozone tendency were first derived (Hood et al., 2005; available by anonymous ftp as noted earlier). Figure 3 compares the observed zonal mean ozone time series for February at 45N with a model time series calculated by integrating over time the ozone tendency estimated from the eddy heat flux data of January and February of each year. Specifically, following Randel et al. (2002), the ozone anomalies are calculated from

$$O_3(t) = \int_{JF} (\Delta O_3 / \Delta t) dt$$
 (1)

where monthly $(\Delta O_3/\Delta t)$ is estimated from observed monthly averages of daily anomalies in v'T' using empirical correlations between these two quantities. Again following Randel et al. (2002), the ozone anomalies are assumed to be zero at the beginning of January for each year. The predicted ozone anomalies are then added to the long-term mean ozone value for February to produce the time series shown in the figure. Both the observed ozone (solid line) and the modelpredicted ozone (dashed line) exhibit a downward trend. The overall correlation coefficient is R = 0.43 implying that approximately 20\% of the interannual variability is explained by the model based on EP flux anomalies. If both time series are detrended, the correlation coefficient is reduced further to 0.31.



Figure 3. Comparison of observed and empirically estimated column ozone when only the mean 40N to 70N EP flux is considered as a predictor variable. For the empirically estimated time series, the ozone anomaly on Jan. 1 of each year was assumed to be zero.

4. OZONE CHANGES CORRELATED WITH ERTEL'S POTENTIAL VORTICITY

To estimate the contribution of local synoptic wave forcing to midlatitude ozone trends, one may employ as an effective dynamical proxy Ertel's PV on isentropic surfaces that connect the tropical upper troposphere with the midlatitude lowermost stratosphere (Hood et al., It has been established empirically that 1999). maximum correlations (~ 0.6) between PV and total ozone at northern midlatitudes occur above the mean tropopause at pressures of 200 - 300 hPa, corresponding to potential temperature surfaces in the range of ~ 330 to 350 K (Vaughan and Price, 1991; see especially Figure 3 of Ziemke et al., 1997). Ertel's PV, which for inviscid, adiabatic motion is conserved, has several advantages for the purpose of estimating the influence of short-term dynamical processes on column ozone trends. First, the appropriate isentropic surfaces (330 - 350 K) lie at a significantly lower level (200 - 250 hPa or 10 - 12 km altitude at middle latitudes) than where decadal lower stratospheric temperature trends are observed. Therefore, long-term differences in this quantity primarily reflect differences in dynamical transport occurring on short time scales during different winters, e.g., differences in the number and amplitudes of poleward synoptic wave events. Since ozone is also approximately conserved on lower stratospheric isentropic surfaces on synoptic wave time scales, differences in PV can be used as a first estimate for the change in ozone to be expected from transport effects alone. Second, comparisons of daily and monthly PV maps with column ozone maps allow an identification of the basic physical cause of the associated ozone trend contributions. As shown, for example, by Hood et al. (1997), extreme ozone minima, occurring with increased frequency or amplitude during a given month, can significantly reduce the monthly zonal mean column ozone value for that year, thereby influencing ozone interannual variability and trends.

Zonally averaged column ozone and 350 K PV are significantly correlated at northern midlatitudes during the winter season (Hood et al., 1999; Hood and Soukharev, 2005). Although depending significantly on latitude, correlation coefficients are generally somewhat higher than those obtained for ozone tendencies versus EP flux and are significant during more months (November through March) at northern midlatitudes. observed interannual PV variations are Since the dominantly caused by year-to-year differences in local synoptic wave forcing, these correlations allow the possibility of estimating the resulting contribution to column ozone interannual variability and trends. It would then be possible to add this contribution to that of non-local planetary wave forcing of the Brewer-Dobson circulation in order to estimate the total contribution of these two dynamical processes to column ozone trends. However, a prerequisite for such an application is that the interannual zonal mean PV variations should be independent of interannual EP flux variations, i.e., ozone changes caused by changes in the Brewer-Dobson circulation should be separate and independent of ozone changes caused by year-to-year differences in local synoptic wave forcing. Consistent with this expectation, 350 K PV and 100 hPa 40N to 70N average eddy heat flux are not significantly correlated at all latitudes of interest during the northern winter (see Hood and Soukharev, 2005).

In the same format as Figure 3, Figure 4 shows a comparison of observed zonal mean column ozone at 45N in February to values predicted by regression relationships between 350 K PV and column ozone at this latitude and for this month. Specifically, the interannual zonal mean ozone anomalies ΔO_3 were calculated from

$$\Delta O_3 = m \Delta P V_{350} \tag{2}$$

where m is the longitudinally averaged sensitivity and ΔPV_{350} is the zonal mean deviation of the 350 K PV at the same latitude during the same month. The overall correlation coefficient (0.55) is somewhat higher than that obtained using the EP flux data (Figure 3) and indicates that the regression model can explain approximately 30\% of the interannual variability. It should be noted that a regression model analogous to (2) can also be applied to individual longitudes using sensitivities and PV deviations for each longitude; the resulting predicted ozone anomalies can then be averaged over longitude. However, results do not differ significantly from that obtained using (2) with zonally averaged sensitivities and zonal mean PV deviations.



Figure 4. Comparison of observed and empirically estimated column ozone at 45N for the month of February when only 350K PV is considered as a predictor variable.

Finally, since the 350 K PV and 100 hPa 40N to 70N mean EP fluxes are not significantly correlated, the contributions predicted by (1) and (2) can be combined linearly to yield an estimate for the total contribution of these two dynamical mechanisms to ozone interannual variability and trends. Results are shown in Figures 5 and 6 for February and March at 45N. (Note that the March contribution from changes in the Brewer-Dobson circulation are calculated from (1) by integrating over the January to March period.) The correlation coefficients (0.67 and 0.65) are higher than obtained using either the EP flux data alone (equation 1) or the 350 K PV data alone (equation 2). Almost half of the interannual variability is explained by the combined regression

model. At 50N, the correlation coefficients are as high as 0.70 in February and March. Figure 6 also provides some evidence that the upturn in column ozone amounts that is seen for some months at northern midlatitudes beginning in about 1993 is at least partially simulated by the dynamical transport empirical model. The ozone increase from 1994 to 2002 is approximately simulated.



Figure 5. Comparison of observed and empirically estimated column ozone at 45N for the month of February when both EP flux averaged over 40N to 70N and 350K PV are considered as predictor variables.



Figure 6. Same as Figure 5 but for the month of March.

A major feature in the observed ozone time series that is not adequately simulated by the dynamical transport empirical model is the depth of the 1993 ozone minimum. As noted above, this extreme minimum closely follows the Mt. Pinatubo volcanic aerosol injection event originating in June, 1991 and is caused mainly by the resulting effects on stratospheric circulation at N.H. midlatitudes (along with a direct chemical contribution). In addition, the minimum is especially large in 1993 because it occurred at a time when the equatorial wind QBO was in its westerly phase. At such times, an induced meridional circulation anomaly exists characterized by adiabatic ascent in the extratropics and descent in the tropics (e.g., Plumb and Bell, 1982). Since the dynamical transport model does not explicitly include the QBO, it is possible that the increased extratropical ascent at this time combined with direct chemical losses on Pinatubo aerosol are sufficient to explain the depth of the anomaly. A more detailed examination of this minimum should be the subject of future work. It may be possible, for example, to apply a regression model that also includes a QBO term to improve upon the simulations shown in Figure

12. However, such a modification would be allowed only if it can be shown that the PV and EP flux interannual variations are not significantly correlated with the QBO.

A similar analysis can be applied to show that about half of the average linear ozone trend at northern midlatitudes over the 1979 to 2002 period can be attributed to a combination of these two dynamical transport mechanisms (Hood and Soukharev, 2005).

5. CONCLUSIONS AND DISCUSSION

In this paper, an attempt has been made to apply empirical (correlative and regression) methods in order to estimate the contribution of two dynamical processes to column ozone interannual variability at northern midlatitudes. Both non-local EP flux variations (implying variations in the Brewer-Dobson circulation) and local PV variations (implying local synoptic wave forcing) were found to

contribute significantly to observed trends and interannual variability. The former process is estimated to contribute 18 to 25\% to observed maximum negative ozone trends in February near 40N to 50N while the latter process is estimated to contribute 27 to 31% to N.H. EP flux variations and local these trends. midlatitude PV variations are not statistically correlated with one another so it has been assumed here that their contributions can be linearly summed. Therefore, the total contribution of these processes to northern midlatitude ozone trends in winter is estimated to be approximately 50%. This empirical estimate is in reasonable agreement with the chemical transport model experiments of Hadjinicolaou et al. (2002). As seen most clearly in Figure 6, the combined empirical model is also able to simulate approximately the change in trend that has occurred since 1993-95 for some months at some latitudes. As reviewed in the Introduction, decreases in EP flux wave forcing and increases in the frequency of anticyclonic, poleward breaking Rossby waves that lead to PV decreases at northern midlatitudes in winter are both consistent with observed long-term net increases in polar vortex strength and longevity as well as increases in the AO and NAO indices.

The cause(s), natural or anthropogenic, of observed decadal changes in stratospheric circulation remain unclear. Observed long-term circulation changes in the Southern Hemisphere may have been produced by polar ozone depletion in the Antarctic spring (Thompson and Solomon, 2002; Gillett and Thompson, 2003) and/or increased greenhouse gas (GHG) forcing (Marshall et al., 2004). A similar influence of ozone depletion in the N.H., in the direction of enhancing the AO and NAO indices, is also expected; however, the reduced amplitude of Arctic ozone depletion makes detection of this influence more difficult in the N.H. against the background of natural variability (e.g., Graf et al., 1998; Gillett et al., 2002). Several authors have reported numerical model simulations indicating that increased GHG emissions, which heat the tropical upper troposphere and cool the polar lower stratosphere in

late fall and early winter, could produce a long-term strengthening of the northern polar vortex (e.g., Perlwitz and Graf, 1995; Kodera et al., 1996; Shindell et al., 1998; 1999; 2001; Perlwitz et al., 2000). In addition, long-term trends in the polar vortex and tropospheric AO could be enhanced further by GHG forced decreases in the tropospheric meridional temperature gradient (Stenchikov et al., 2002). The latter is caused by polar amplification of warming resulting from albedo-snow-sea effects and lead to a decrease in wave activity flux into the stratosphere, allowing the polar vortex to increase in strength. On the other hand, other model simulations of the effect of {\it future} increases in GHG forcing predict an increase in overall generation of planetary waves leading to a weaker N.H. polar vortex (for a review, see Austin et al. (2003). Therefore, it must be concluded that the origin of observed changes in stratospheric circulation over the past few decades is not yet well understood.

Acknowledgments. The TOMS / SBUV ozone processing team (led by R. Stolarski and R. McPeters) at the NASA Goddard Space Flight Center provided the column ozone data used in this work. The NCEP/NCAR Reanalysis data team at the NCEP Climate prediction Center provided the data used to calculate EP fluxes and 350 K potential vorticity. Work leading to this paper was supported by a grant from the NASA Atmospheric Chemistry Modeling and Analysis Program.

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