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## TOTAL OZONE TRENDS DEDUCED FROM NIMBUS 7 TOMS DATA

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**Abstract.** The Total Ozone Mapping Spectrometer (TOMS) on the Nimbus 7 satellite has been measuring the total column amount of ozone over the globe for more than 11 years. Recent improvements in the data analysis have led to a technique for determining and removing drift in the calibration such that the data at the end of the record are precise to  $\pm 1.3\%$  ( $2\sigma$ ) relative to the data at the beginning of the record. A statistical model, including terms for seasonal variation, linear trend, quasi-biennial oscillation, solar cycle and second-order autoregressive noise has been fit to the TOMS time series of total ozone data. The linear trend obtained when this statistical model is fit to the TOMS data averaged between 65N and 65S latitudes is  $-0.26 \pm 0.14\%$  /year or  $-3\%$  over the 11.6 year time period from November, 1978 through May, 1990. The trend is near zero ( $0.0002 \pm 0.2\%$  /year) at the equator and increases towards both poles. At 50N the annually averaged trend is  $-0.5 \pm .21\%$  /year. The 50N trend over the 11.6 year time period shows a strong seasonal variation from more than  $-0.8\%$  /year in winter and early spring (February and March) to about  $-0.2\%$  /year in summer (July and August).

## Introduction

Decreases in the total amount of stratospheric ozone have been predicted to occur as a result of human activities, especially the release of chlorofluorocarbons into the atmosphere (Molina and Rowland, 1974). These predictions remained in the realm of theory until the discovery of rapid ozone decreases in the springtime Antarctic ozone hole (Farman, et al., 1985) and reports of a wintertime decrease in ozone at northern mid latitudes (WMO, 1990).

This paper reports trends derived from the data of the Total Ozone Mapping Spectrometer (TOMS) which has been measuring the total column content of ozone from the time of its launch, on the Nimbus 7 spacecraft, in late October of 1978 until the present time. TOMS is an Ebert-Fastie monochromator that scans across the nadir track to provide a global map of the deduced ozone each day. It measures the ultraviolet albedo of the Earth at 6 wavelengths. Four of these (312.5, 317.5, 331.2, and 339.8 nm) are used in pairs for the measurement of total ozone in combination with surface reflectivity measurements at 360 and 380 nm.

The main disadvantage of TOMS, or any other satellite,

is the difficulty in maintaining calibration over time. A new method has been developed to determine the calibration drift of the TOMS instrument (Herman, et al., 1991) based on a requirement of internal consistency in ozone measured with different wavelength pairs. This method produces a calibrated total ozone data set which is independent of the Dobson network. The reprocessed data, called version 6, are now available from the National Space Science Data Center. Evaluation of the propagation of errors through the calibration and analysis indicates that the ozone measurements are precise to  $\pm 1.3\%$  ( $2\sigma$ ) at the end of the record relative to the beginning of the record (Herman, et al., 1991). This is confirmed by comparison to the World Standard Dobson Instrument (#83) during satellite overpasses, and by comparison to a composite of stations in the Dobson network (McPeters and Komhyr, 1991). The present paper examines the trends determined from TOMS as a function of latitude and season.

## Statistical Model

Ozone trends, as well as solar-cycle and quasi-biennial oscillation (QBO) signals have been estimated from the TOMS data by regression analysis. The regression model used is

$$O_3(t) = \mu + \alpha \cdot trend + \beta \cdot QBO + \gamma \cdot Solar + noise \quad (1)$$

where  $\mu$ ,  $\alpha$ ,  $\beta$ , and  $\gamma$  are constants to be estimated.

To characterize the noise in the ozone data, (1) was first fitted separately for each week of the year. At most latitudes, the noise series showed second order autoregressive structure, although in some cases the second order term was not statistically significant. To allow for the possibility that the autoregressive structure itself had seasonal structure, both the first and second order autoregressive terms were assumed to be of the form of a constant plus an annual cycle.

With the statistical structure of the noise determined, the regression model, (1) was refit to the full data set by generalized least squares. The seasonal model consisted of a constant plus annual variation and 3 harmonics, 6 month, 4 month and 3 month. The trend model consisted of a linear trend throughout the data record, modulated by annual and semi-annual terms. The quasi-biennial oscillation model consisted of a coefficient multiplied by the lagged smoothed Singapore 30 mbar winds (obtained from J. Angell, personal communication), where the coefficient had a term to allow for annual variation. Finally, the solar model consisted of a coefficient multiplied by the smoothed 10.7 cm solar radio flux measured at Ottawa (from the U.S. Department of Commerce reports on Solar Geophysical Data), where the coefficient again allowed for annual variation. In making inferences about the parameters in these various submodels, the parameters of the noise model were regarded as known.

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### Global Average Results

The TOMS data can be integrated over the globe to obtain a global average time series. Because TOMS does not make measurements in the polar night, the area-weighted average between the latitudes of 65S and 65N, where year round measurements are available, has been calculated. The areal coverage is 90% of the globe, but does not include the Antarctic ozone hole region. Fitting the statistical model of equation (1) to this time series leads the following parameters:

Trend =  $-0.26 \pm 0.14$  %/year or -3% over the 11.6 year data record

Solar =  $3.7 \pm 0.6$  DU/100 units of F10.7 or 1.5% over the solar cycle

QBO =  $-0.4 \pm 0.1$  DU/10 knots of 30 mbar wind or 1% over a QBO cycle.

The units of the solar cycle and quasi-biennial oscillation (QBO) terms are explained in the section on the latitude dependent trends. The solar and QBO terms are both consistent with the findings of the Ozone Trends Panel (WMO, 1990). The error estimate for the trend is a  $2\sigma$  root sum of squares (rss) estimate of the combined  $\pm 0.13$  %/year instrumental trend error and the  $\pm 0.04$  %/year statistical error for the model fit.

### Latitude and Seasonal Dependence

The linear trend term, expressed in %/year, derived from the statistical model is shown in Figure 1 as a function of latitude from 60S to 60N. The error bars shown are the rss sum of the  $2\sigma$  statistical uncertainty from the model described above and the absolute error of  $\pm 1.3\%$  over the 11-year record. The observed trend is not statistically different from zero from 20S to 20N. Models of the fluorocar-

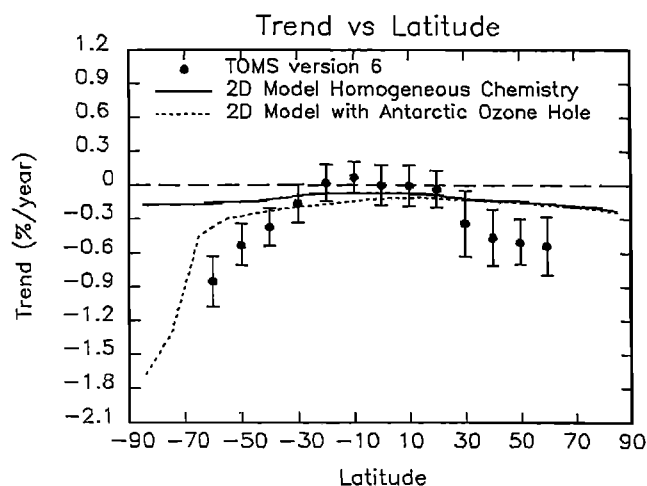


Fig. 1. Seasonally-averaged ozone trend term, deduced using a statistical model, as a function of latitude. Error bars represent  $2\sigma$  statistical errors plus an estimated instrumental trend uncertainty of  $\pm 1.3\%$  ( $2\sigma$ ) over 11 years. Solid line shows a 2D model calculation with only homogeneous gas-phase chemistry. Dashed line shows a similar 2D model with an imposed ozone hole in the Antarctic spring.

bon effect on ozone for gas-phase homogeneous chemistry predict about 1% change in tropical ozone during this period. A typical example is shown in Figure 1 by the solid line (Jackman, personal communication using the model described by Douglass, et al., 1989). If the trend were truly as near zero as shown for 20S to 10N, then we would have to conclude that the models overestimate the effect of pure gas-phase chlorine chemistry. However, the error bars shown on the figure lead to the conclusion that the observed data are consistent with model predictions. The TOMS data do not have sufficient trend accuracy in the 11.6 years to resolve the expected change of less than 1%.

Poleward of 20 degrees in both hemispheres, the TOMS data show statistically significant trends. By 60S the negative trend is nearly 0.9 %/year and is increasing with latitude as the region of the Antarctic ozone hole is approached. A reasonable interpretation is that this is due to mixing of ozone poor air from the Antarctic ozone hole region with air at lower latitudes, causing an ozone dilution effect. A statistically significant trend is also seen through northern midlatitudes. The statistical uncertainties are larger because of the greater degree of meteorological variability. The negative trend is about 0.4 to 0.5 %/year from 40 to 60N. It is possible that this change is due to the effects of polar chemical processing in the Arctic vortex.

To examine the question of Arctic processing more closely, Figure 2 shows the deduced trend as a function of latitude and season. A region of zero or slightly positive trend is seen around the equator. The trend becomes increasingly negative towards the south with a seasonal

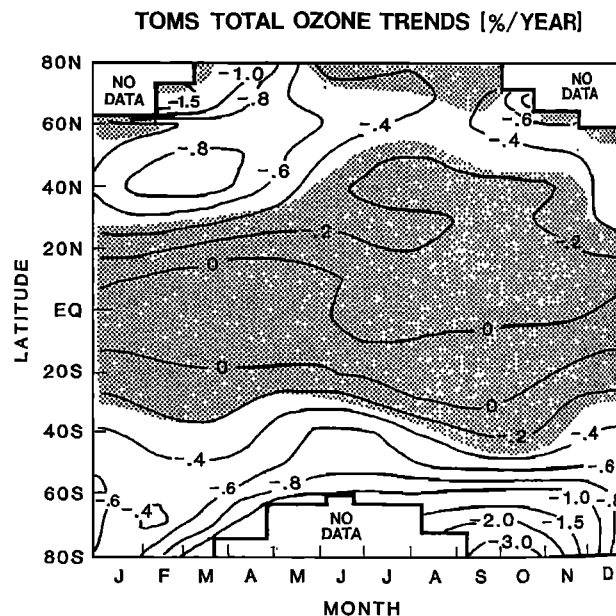


Fig. 2. Trend term as a function of latitude and season from the statistical model. Seasonal variation is fit with only 2 harmonics, annual and semi-annual. The shaded area indicates where the trends are not statistically different from zero at the  $2\sigma$  level. Significance was determined by rss sum of absolute and statistical error estimates. Poleward of the heavy solid line is polar night where no TOMS measurements are possible.

maximum in October. The northern hemisphere shows a larger midlatitude seasonal variation with a maximum in winter, consistent with the previous findings of Bojkov et al. (1990). There are a number of quantitative differences with the results of Bojkov et al., based on the analysis of data from ground-based Dobson spectrophotometers through the end of 1986. Their winter trends ranged from about -0.1 to -0.3 %/year, increasing toward the pole. The TOMS data show a much larger trend, reaching just over -0.8 %/year, with a maximum just north of 40N.

Also indicated in Figure 2 are the statistical significance of the derived trends. The shading indicates regions where the trend is not statistically different from zero at the  $2\sigma$  level. The unshaded regions are those with statistically significant trends. The entire region south of about 40S shows statistically significant trends. In the north, the winter trends are statistically significant from about 30 to 60N. In the spring, the region of significance extends to 80N. During summer and fall it shrinks to a 10 degree band centered between 50 and 60N.

Figure 3 shows a more direct comparison of the northern and southern hemisphere trends as a function of time of year. In the top panel, 60N is compared to 60S (shifted 6 months) with  $2\sigma$  error bars indicated. The seasonal pattern in each case is the same, with larger trends at 60S. The bottom panel shows the same results for 40N and 40S. The seasonal behavior is different in the two hemispheres, with 40N showing the large winter, early spring trend, and a more or less annual component, while 40S shows a semianual component with much less overall seasonal difference. The winter trend at 40N is larger than that at 40S by more than the combined  $2\sigma$  error bars.

There is a small region between 70 and 80N in Figure 2 where large trends are seen just after polar night in late February and early March. Although these meet the test

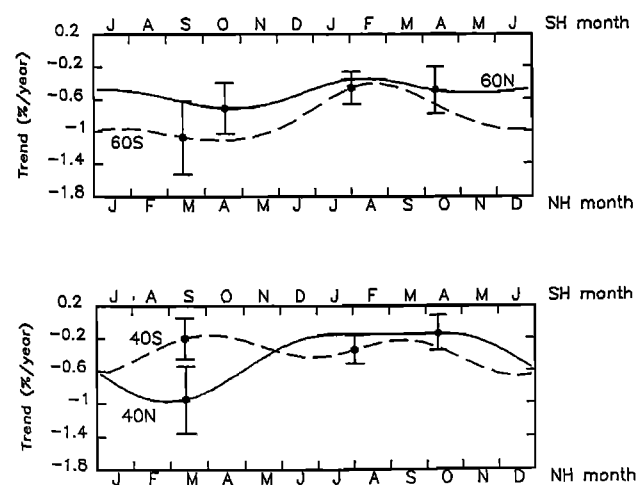


Fig. 3. Comparison of the northern and southern hemisphere trend vs. time of year for a) 60 degrees latitude and b) 40 degrees latitude. Error bars indicated include only the  $2\sigma$  statistical errors. Southern hemisphere data have been shifted by 6 months relative to the northern hemisphere; the months for the southern hemisphere are indicated at the top of each graph while those for the northern hemisphere are indicated at the bottom.

of statistical significance, they are heavily influenced by extremely low 1990 data. The low springtime 1990 ozone data could be a manifestation of interannual variability in stratospheric dynamics, in that the character of the spatial distribution of total ozone is quite different from previous years. It also has been suggested that the 1990 spring data at high northern latitudes may be influenced by the large solar proton events of late 1989 (Reid, et al., 1991). The statistical model has also been fit without using any 1990 data. The midlatitude peak at 40N is essentially unchanged, but the high latitude peak disappears.

A possible explanation for the larger trends in the northern midlatitudes, seen in this work, is that the data extends through mid 1990 while the Bojkov et al. results were based on a trend assumed to begin in 1970 and to extend only through 1986. Bojkov, et al. give the average trend over 17 years, while the TOMS data give the average trend for 1979 through mid 1990. If the negative trend is caused by homogeneous gas-phase chemistry of the chlorine from fluorocarbons, then it would be expected to increase slightly with time. If the trend is being caused by heterogeneous reactions modifying the 'normal' gas-phase chemistry within the Arctic vortex, then a significant quadratic term in total chlorine concentration would be expected through the chlorine dimer term (Molina and Molina, 1987) leading to an acceleration of the trend.

Another complicating factor in the comparison of the TOMS results with those of the Dobson stations is that tropospheric ozone is increasing in the northern hemisphere, partially offsetting a decrease in the stratospheric ozone layer. TOMS does not measure lower tropospheric ozone with full sensitivity (Klenk, et al., 1982) and will slightly overestimate the trend in the northern hemisphere. A very conservative estimate of the possible error can be made. The largest trend in tropospheric ozone has been measured in a series of balloon measurements at Payerne, Switzerland (Stachelin and Schmid, 1991). They show that tropospheric ozone in February increased from 19.9 DU to 29.4 DU between 1979 and 1988. The TOMS zonal mean ozone change at 45N in February (see Figure 2) is a decrease of about 9% or 35 DU. If TOMS measured only half of the 9.5 DU tropospheric increase, then the true trend would have been a decline of 8% or 30 DU, a 1% underestimate of the trend. A similar analysis produces less than a half percent trend error for summer conditions. Since hemispheric changes in tropospheric ozone are almost certainly less than that measured in northern Europe, the northern hemispheric trend error in TOMS due to its reduced sensitivity to tropospheric ozone should be no more than half a percent. The tropospheric trend error for the equatorial zone and southern hemisphere is negligible.

An interesting feature of the data in Figure 2 is that the northern hemisphere winter trend reaches as far south as 30N. This may be the result of polar processing and the transport of that air to mid latitudes as the vortex is distorted by wave activity, or as it breaks up. Air which has had its active chlorine content increased as a result of heterogeneous processes would be transported away from the pole and result in a negative gradient in the amount of active chlorine away from the pole. The sunlight available to drive the chlorine destruction process increases away from the pole, and the overlap with the active chlorine concen-

tration could lead to a maximum destruction away from the pole. While this is qualitatively reasonable, there is no quantitative model to show that it is consistent with measured trends, nor is there yet a consensus within the scientific community about how much chemically perturbed air can be transported to mid latitudes during the winter (see e.g. Tuck, 1989; Schoeberl and Hartmann, 1991; Hartmann et al., 1989). Another possibility is that the heterogeneous reaction of  $\text{N}_2\text{O}_5$  with  $\text{H}_2\text{O}$  on background sulfuric acid aerosol perturbs the  $\text{NO}_x$  and hence chlorine chemistry (see e.g. Brasseur et al., 1990).

### Conclusions

TOMS data for the 11 year 7 month period from November, 1978 through May, 1990 show a statistically significant trend in the global average (65S to 65N latitude). The latitude dependence of this trend, near zero at the equator and increasing towards both poles, indicates the likelihood that the origin of the trend is a high latitude process. The southern hemisphere trend is larger and is consistent with dilution effects from the Antarctic ozone hole. The northern hemisphere trend is also significantly larger than expected from homogeneous gas phase chlorine chemistry. The northern mid latitude ozone trend shows a significant seasonal cycle with a maximum in late winter (February and March). The maximum is located near 40 degrees latitude and reaches greater than 0.8 %/year of ozone decrease. Because the observed depletions reach 0.6 %/year in early January at latitudes below 50N, caution must be used in interpreting these results as due to Arctic heterogeneous chemistry. Detailed modeling studies and further measurements of the species involved in the chlorine-ozone chemistry will be needed to determine the mechanism of the observed decrease.

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