Trends in the vertical distribution of ozone: a comparison of two analyses of ozonesonde data.

J. A. Logan¹, I. A. Megretskaia¹, A. J. Miller², G. C. Tiao³, D. Choi³, L. Zhang³, R. Stolarski⁴, G. J. Labow⁵, S. M. Hollandsworth⁶, G. E. Bodeker⁷, H. Claude⁸, D. DeMuer⁹, J. B. Kerr¹⁰, D. W. Tarasick¹⁰, S. J. Oltmans¹¹, B. Johnson¹¹, F. Schmidlin¹², J. Staehelin¹³, P. Viatte¹⁴, and O. Uchino¹⁵.

- 1. Department of Earth and Planetary Sciences, Harvard University, Cambridge, Massachusetts
- 2. Climate Prediction Center, National Weather Service, NOAA, Washington, D.C.
- 3. Graduate School of Business, University of Chicago, Illinois
- 4. NASA Goddard Space Flight Center, Greenbelt, Maryland
- 5. Raytheon-STX, Lanham, Maryland
- 6. Steven Myers and Associates Corp., Vienna, Virginia
- 7. National Institute of Water and Atmospheric Research, Lauder, New Zealand
- 8. Deutscher Wetterdienst, Observatorium Hohenpeissenberg, Germany
- 9. Meteorological Institute of Belgium, Brussels
- 10. Atmospheric Environment Service, Downsview, Ontario, Canada
- 11. Climate Monitoring and Diagnostic Laboratory, NOAA, Boulder, Colorado
- 12. NASA Wallops, Wallops Island, Virginia
- 13. Institute for Atmospheric Sciences, Swiss Federal Institute of Technology, Zurich
- 14. Aerological Station, Swiss Meteorological Institute, Payerne
- 15. Atmospheric Environment Division, Japan Meteorological Agency, Tokyo

Submitted to the Journal of Geophysical Research, September, 1998.

Revised Manuscript, April 27, 1999.

Abstract.

We present the results of two independent analyses of trends in the vertical distribution of ozone. For most of the ozonesonde stations we use data that were recently reevaluated and reprocessed to improve their quality and internal consistency. The two analyses give similar results for trends in ozone. We attribute differences in results primarily to differences in data selection criteria, rather than in statistical trend models. We find significant decreases in stratospheric ozone at all stations in middle and high latitudes of the northern hemisphere from 1970 to 1996, with the largest decreases located between 12 and 21 km, and trends of -3 to -10 %/decade near 17 km. The decreases are largest at the Canadian and the most northerly Japanese station, and are smallest at the European stations, and at Wallops Island, U.S.A. The mean mid-latitude trend is largest, -7 %/decade, from 12 to 18 km for 1970-96, and the decrease in ozone is significant from 10.5 to 25 km. For 1980-96, the decrease is more negative by 1-2 %/decade, with a maximum trend of -9%/decade in the lowermost stratosphere. The trends vary seasonally from about 10 to 17 km, with largest ozone decreases in winter and spring. Trends in tropospheric ozone are highly variable and depend on region. There are decreases or zero trends at the Canadian stations for 1970-96, and decreases of -2 to -8 %/decade for the mid-troposphere for 1980-96; the three European stations show increases for 1970-96, but trends are close to zero for two stations for 1980-96 and positive for one; there are increases in ozone for the three Japanese stations for 1970-96, but trends are either positive or zero for 1980-96. The U.S. stations show zero or slightly negative trends in tropospheric ozone after 1980. It is not possible to define reliably a mean tropospheric ozone trend for northern mid-latitudes, given the small number of stations and the large variability in trends. The integrated column trends derived from the sonde data are consistent with trends derived from both surface based and satellite measurements of the ozone column.

1. Introduction.

Accurate knowledge of trends in the vertical distribution of ozone is needed to evaluate current understanding of the processes responsible for decreases in the ozone column, and processes responsible for changes in tropospheric ozone. The vertical profile of ozone loss determines how global stratospheric temperatures will be affected by ozone depletion and how surface temperatures will respond to changes in the entire profile [e.g., Ramaswamy et al., 1996; Hansen et al., 1997; Miller et al., 1992]. The primary sources of information on profile trends are ozonesondes, the Stratospheric Aerosol and Gas Experiment (SAGE), Solar Backscattered Ultraviolet (SBUV) instruments, and the Umkehr technique [e.g., Logan, 1994; Wang et al., 1996; Hollandsworth et al., 1995; Miller et al., 1997; Harris et al., 1997]. Ozonesondes provide the only information on the vertical distribution of ozone in the troposphere and lower stratosphere below 20 km, and they provide the most reliable information on trends below 30 km prior to the satellite measurements that started in 1979. Lidars are now providing measurements of the ozone profile in the stratosphere and/or troposphere at a few stations, but long term records are lacking [WMO, 1998].

As part of the continuing efforts by the international community to make more reliable estimates of ozone trends, those responsible for the ozonesonde observations undertook a review of the historical data and made them available for trend analysis under the general organizational umbrella of the World Climate Research Programme, Stratospheric Processes and their Role in Climate (SPARC). The data were examined and re-analyzed to provide, as much as possible, consistency and continuity in the data. The reevaluated data will be archived at the World Ozone and Ultraviolet Data Center (WOUDC). A major goal of this exercise was to improve the quality of data that could be used in an international assessment of trends in the vertical distribution of ozone conducted in 1997 [WMO, 1998]. The re-evaluated sonde data, along with SAGE, SBUV, Umkehr, and TOMS (Total Ozone Mapping Spectrometer) data were used as part of this assessment. Results are presented in four papers. Here, we describe the results of two independent analyses of the sonde data. Cunnold et al. [1999] describe trends in the lower stratosphere derived from SAGE I and II (Version 5.96) data, and Newchurch et al. [1999] describe the trends in the upper stratosphere derived from Umkehr, SAGE, and SBUV. A comparison of trends derived from all the techniques and an analysis of their consistency with column ozone trends is given in Randel et al. [1999].

Consideration of the possible sources of error inherent in the ozonesonde system led to two different approaches to data selection prior to trend determination, as described in detail in Section 2. One, used by Tiao, Choi, Zhang, and Miller, employs strict data selection criteria [Miller et al., 1995] and a statistical model that includes an autoregressive error analysis; this will be referred to as the Tiao et al. analysis. The other approach, adopted by Logan and Megretskaia, uses a less stringent data filter [Logan, 1994], and a similar statistical model but without an autoregressive error analysis; this will be referred to as the LM analysis. The trend models are described in Section 3, results from the two analyses are compared in Section 4, and the results are discussed in Section 5.

2. Data Selection and Analysis.

2.1 The ozonesonde data.

Re-evaluated data were provided for three stations in Europe, Hohenpeissenberg, Payerne and Uccle; three in Japan, Sapporo, Tateno, and Kagoshima; Boulder, Colorado, and Hilo, Hawaii, in the United States; and Lauder, New Zealand (Table 1). The data for Payerne are undergoing further homogenization [Stubi et al., 1998], and an interim data set was provided. Data for four Canadian stations and for Wallops Island, U.S., were taken from the World Ozone and Ultraviolet Data Center (WOUDC) in July 1997: these data were not reevaluated as part of the SPARC activity, but the Wallops Island data were reprocessed as described below. The data from each sonde program were processed according to the best judgement of those responsible for the program, so that procedures were not necessarily identical [WMO, 1998]. Data from Aspendale/Laverton in Australia were not used, as problems have been identified with the data and they are currently undergoing reanalysis [Atkinson, pers. comm., 1997]. Data from Natal, Brazil were not used because of sparse data for 1993-96 [Kirchhoff, pers. comm., 1997]; trends

- 4 -

for the earlier data are given in Logan [1994].

Trends analyses of the various types of data used in the SPARC activity were conducted on the vertical co-ordinate fundamental to the measurement technique, which is pressure for the sondes [WMO, 1998]. The sonde data were provided in a variety of formats. These were processed into a common format which gives the column of ozone in Dobson units in 33 equally spaced layers in log pressure from 1000 to 6.3 hPa (30 layers up to 10 hPa). (1 Dobson unit (DU) = 2.69 x 10¹⁶ molecules ozone cm⁻²). The vertical resolution, ~1 km, was chosen to be similar to that of SAGE data.

The characteristics of the major types of ozonesondes are described in WMO [1998]; this report also provides details about the sonde programs and data reduction algorithms, and discusses issues of data quality in depth [see also WMO, 1995; Reid et al., 1996; Logan, 1994; Tiao et al., 1986]. The Canadian stations used Brewer Mast (BM) sondes until 1980, and electrochemical concentration cell (ECC) sondes thereafter. The three European stations use BM sondes, the Japanese stations use their own type of sonde, type KC, and the other stations use ECC sondes. The initial goal of the sonde programs started in the 1960s was to investigate the distribution of stratospheric ozone, and the effects of large scale synoptic influences on lower stratospheric ozone. The data were not used to examine ozone trends until the early 1980s [e.g., Angell and Korshover, 1983].

Most stations making sonde measurements scale the ozone profile to an independent measurement of the ozone column made by a Dobson or Brewer spectrophotometer, and provide the scaling, or correction factor (CF), with the data [WMO, 1995, 1998]. The practice of scaling to the ozone column arose because BM sondes underestimate stratospheric ozone by 10-25% [Dutsch, 1966]. There are difficulties associated with this practice, as the use of a scaling factor may distort the shape of the vertical profile, and any errors in the column measurements are carried over into the sonde results [e.g., Hilsenrath et al. 1986; WMO, 1998]. It has been suggested that the scaling procedure is not appropriate for the tropospheric part of the profile. The SPARC review of data quality concluded that the scaling procedure should be applied to the whole

- 5 -

profile, as this issue is still in an exploratory stage [WMO, 1998]. The correction factors are provided with the sonde data, and the user can thus choose whether or not to use the data scaled to the ozone column. Previous studies have shown that trends in ozone are similar whether or not the data are scaled to the ozone column, as long as there is no trend in the correction factors at a particular station [Logan, 1985, 1994]. The CFs are commonly used as a quality check [e.g., Tiao et al., 1986, Logan, 1994].

The scaling procedure requires a homogeneous data set for the ozone column and an estimate of the amount of ozone above the altitude reached by the balloon (usually between 20 and 7 hPa). The column measurements in turn depend on the absorption coefficients for ozone. The coefficients recommended by the World Meteorological Organization (WMO) for reducing the spectrophotometer data changed in 1992 to those measured by Bass and Paur [1985]. The sonde data from Europe, Japan, and the United States were provided for the SPARC activity scaled to column measurements on the Bass-Paur scale; the Lauder data were provided unscaled, but correction factors were given with the data [Bodeker et al., 1998]. The data for Wallops Island were reprocessed to scale the profiles to ozone column data on the Bass-Paur scale [Oltmans et al., 1998]. The Canadian ECC data at WOUDC were already scaled to column measurements on the Bass-Paur scale [Tarasick et al., 1995]; the BM data are scaled to column data on the older Vigroux scale, and were adjusted to the Bass-Paur scale (multiplied by 0.9743) for the analyses shown here. There are concerns about the quality of the BM data for these stations [WMO, 1998]. The Canadian stations followed operating procedures recommended by the manufacturer, while the European stations developed their own procedures [Claude et al., 1988] which are considered now to be more reliable [WMO, 1998].

The standard procedure for estimating ozone above the balloon burst is to assume a constant mixing ratio based on the value at or near the top of the profile. A new method using an SBUV climatology is now available; it gives mean correction factors that are larger by about 3.5% for Boulder and 5% for Hilo [McPeters et al., 1997]. The new method was used for the Wallops Island, Boulder, Hilo, and Lauder data [Oltmans et al., 1998]. The SBUV climatology

- 6 -

was used for Payerne soundings that ended between 30 and 17 hPa, while for those that reached above 17 hPa a constant mixing ratio was assumed. Between 94 and 100% of the acceptable soundings (according to the CF criteria used by LM) from the reevaluated stations and from Wallops Island reach 20 hPa, allowing a good estimate to be made of the ozone column above burst altitude (Table 2). About 80% of the Canadian sondes reach 20 hPa, except for Resolute (~70%). The Canadian ECC profiles that fail to reach 17 hPa are not scaled to the ozone column, and a value of 1.0 is given for the correction factor in the WOUDC archive. For the older BM data, a mean default value is used for the CF if the column is not measured; otherwise the ozone values would be too low by 20-25%. For Resolute (75°N), almost all the winter data are scaled to the default CF because of the lack of column measurements.

Intercomparisons of BM and ECC sondes in the 1970s and early 1980s showed that ECC sondes measured more ozone in the troposphere than BM sondes by about 15 to 20% (with a range of 7-38%), while differences in the stratosphere are less than 5%, as long as the profiles are first scaled to the ozone column [e.g., Logan, 1994; WMO, 1995, 1998]. The change from BM to ECC sondes at the Canadian stations around 1980 introduced a shift in tropospheric ozone values that must be accounted for in deriving trends. Tiao et al. [1986] introduced an intervention term in their statistical trend model to account for any shift in ozone, and this approach was adopted here by both groups. The intervention term specifies the time of the change in instrument type, and the magnitude of any shift in the data is computed as part of the statistical model fit. The magnitude of the intervention term thus derived is similar to the differences between BM and ECC sondes found in the intercomparisons, and varies among stations [Tiao et al., 1986]. ECC sondes were used exclusively in Canada after 1980 (September 1980 for Goose Bay), so no intervention term in necessary in deriving trends for 1980-96.

The correction factor was used as a selection criterion in the trend analyses. Time series for the correction factors for selected stations are shown in Figure 1; those for the other stations used here are shown in WMO [1998]. Mean values of the correction factor are typically about 1.0 for ECC and KC sondes and 1.25 for BM sondes, except for Hohenpeissenberg, where the

- 7 -

mean CF is about 1.1 [Logan, 1994]. Reasons for the different mean correction factors for the three BM stations are not fully understood. The jump in the correction factors at Uccle in 1989 is caused by a change in the way they are calculated. The standard procedure was used before 1989, and since then they have been determined in the laboratory, by taking the ratio of the ozone concentration from a calibrated source, and from the ozone sensor of the sonde. The pump efficiency has also been adjusted. These changes, motivated by a decrease in quality of the pumps supplied with the BM sondes in 1989, are described in DeBacker et al. [1998a,b] and WMO [1998]. At Payerne, the preflight protocol of Hohenpeissenberg was introduced in 1983, and in 1984 changes were made in how the pump flow was measured. Since 1993, several changes in sonde preparation and calibration were introduced [WMO, 1998]. These changes seem to have caused lower correction factors, particularly in 1993. The jump in the correction factors at Goose Bay in 1980 is caused by the change from BM to ECC sondes. At Hohenpeissenberg, the radiosonde type and interface were changed in August, 1995, and any possible effects on ozone data are being investigated; there was no dramatic effect on the CFs.

2.2 Data Selection Criteria.

Logan and Megretskaia (LM) required that the correction factors were in the range 0.9-1.35 for BM sondes (except for Hohenpeissenberg where 0.9-1.2 was used), and in the range 0.8-1.2 for ECC and KC sondes [cf Logan, 1994]. Tiao et al. used the CF criteria of 0.9-1.2 for BM and 0.9-1.15 for ECC and KC sondes, as in their earlier work [Miller et al., 1995]. The fraction of soundings that met these two sets of criteria are given in Table 2. Tiao et al. required also that there was an ozone column measurement for the day of the sounding, and that the balloon burst was above 16 hPa; these requirements are to ensure that a correction factor can be calculated, and that a good estimate can be made of the ozone profile above burst altitude.

LM used the data as provided to the SPARC working group, which meant that profiles for all stations except Lauder were scaled to ozone column measurements. Tiao et al. removed the scaling to the ozone column from the sonde data by dividing each profile by the correction factor. If there is a trend in the CF this can result in different trends being derived for the ozone

- 8 -

profile, while if there is no trend, removing the scaling has little effect on the trends [e.g., Logan, 1985, 1994; Miller et al., 1995].

LM analyzed the trends for 33 layers and for 11 layers obtained by adding the ozone columns in 3 adjacent layers; results for 11 layers are shown here, as results for 33 layers did not appear to provide more useful information on trends, and the error estimates are similar (Figure 2). Tiao et al. aggregated the 33 layers into 15 Umkehr layers that they have used in prior work [Tiao et al., 1986; Miller et al., 1995].

Tiao et al. omit a significantly larger fraction of the soundings from trend analysis than LM because of their stricter selection requirements, as shown in Table 2; the last column gives the fraction of soundings retained by Tiao et al. and the third column the fraction retained by LM. Both groups use the same CF criteria, 0.9-1.2, for Hohenpeissenberg and retain ~90% of the soundings. In the worst cases, the BM data from Canada and Payerne, Tiao et al. use only 25-44% of the profiles, while LM use 70-90%; for the BM stations, a larger fraction of soundings are omitted from the earlier part of the record, because of the downward trend in the correction factors. For Wallops Island, the Canadian ECC soundings, and the Japanese stations, Tiao et al. retain about 45-65% of the profiles (except for Churchill, 31%), while LM retain about 85-95%. Another consequence of the stricter data selection criteria of Tiao et al. is that there are more months without any data in the time series used for trend analysis, as well as fewer soundings contributing to each monthly mean, as shown in Table 3. This is particularly true for the first 5 years at Payerne, Churchill, and the Japanese stations. The main cause of data loss for the Tiao et al. analysis is the stricter correction factor criteria; for the Canadian stations the requirement of the balloon reaching 16 hPa also causes significant data loss.

Trends in the correction factor using the Tiao et al. data selection criteria are given in Table 4. There are small but significant trends in the CF at several stations, mostly in the range -1 to -4%/decade. For both Uccle and Payerne, the stations with the largest trends, the trend is caused in large part by the changes in the typical magnitude of the CF in recent years (Figure 1).

3. Statistical Trend Models.

Logan and Megretskaia model (LM). The model includes monthly means, four seasonal trends, a lagged QBO and a solar dependence. The monthly means are weighted by the inverse of the interannual monthly variance (e.g., the variance for the January means, etc), in an iterative procedure; after fitting the model with the observed variances, the weights are recalculated from the residuals for the fit, to remove the influence of the trend on the variances. The latter step is repeated another time to obtain the final weights for the model. The model does not account for autoregression. An intervention term is included in the model for the four Canadian stations at all pressure levels, at the date of the change from BM to ECC sondes; a similar term is included at Payerne for the tropospheric levels in April 1977 to account for the change in the time of soundings from ~1600 to 0930 [Logan, 1985]. The QBO time series used is the 30 hPa wind speed for Singapore and the F10.7 cm solar radio flux at Ottawa is used for the solar cycle dependence. Time lags were determined for the QBO as described in Logan [1994].

Tiao et al. model. The model includes monthly means, four seasonal trends, a lagged QBO, a solar dependence, and an intervention term as above for the Canadian stations and for Payerne. The noise term is modelled as a first order autoregressive model, with different variances in different seasons. In practice, the model is first run with no seasonal weighting. From the residuals of the fit, the seasonal weights are calculated as the square roots of the estimated variances for different seasons. (The estimated variance is given by the sum of squares of residuals divided by the corresponding number of data points.) These weights are then used in a weighted regression, and the procedure repeated to convergence; the convergence criteria is that the change in the sum of squares of the residuals is less than 0.001. The QBO time series is the average of 50 hPa winds at Singapore, Balboa, and Ascension Island. The model assumes zero trend before Jan. 1 1970. Outliers are removed from the analysis; these are defined as points whose residuals are more than three standard deviations away from the model fit.

The models are similar in that they fit 12 monthly means and 4 seasonal trends, and allow for the dependence of ozone on the QBO and solar flux. Difference between the models are the inclusion of autoregression, the assumption of zero trend before 1970, the removal of outliers,

and the use of seasonal weighting (LM use monthly weighting) in the Tiao et al. model. The weighting in the Tiao et al. model is iterated to a convergence criteria, rather than a fixed number of times (LM). Eight of the stations have data before 1970 (Table 1). The vertical distribution of trends are compared primarily as relative trends (e.g., percent per decade), the conventional way of showing ozone profile trends. Note that each group used different layers and that percentage trends were computed relative to a different reference. The LM trends are given relative to the mean of the time series for which the trend is calculated, and the Tiao et al. trends relative to the seasonal intercept in 1970 (or beginning of series if later) adjusted for solar effects and intervention if used. A disadvantage of using the beginning of the time period as a base is that it is not necessarily well-defined by observations once a significant fraction of the data have been omitted by the selection criteria. Absolute trends in $DU \text{ km}^{-1}$ and column trends in the troposphere (1000-250 hPa) and stratosphere (250-16 hPa) are shown, so that the results can be compared in the same units (DU) for the same columns. It would not produce self-consistent results for relative trends to reference the results from the two groups to the same values, since the data treatment procedures of LM and Tiao et al. cause the time series for a given station and level to be different, and the original 33 levels are then aggregated into different layer thicknesses.

Trends were computed for 1970 (or the start of data if after 1970) to 1996 by both groups and for 1980-96 by LM. Both groups computed the annual trends as the average of the seasonal trends and used the covariance matrix of seasonal trends to calculate the standard error of the annual trend.

4. Results.

4.1 Lower Stratosphere.

Time series.

Figure 3 (upper panel) shows monthly mean values of ozone near 90 hPa for three stations, using the LM selection criteria; the right hand panels show the time series of deseasonalized monthly means. Similar results for the Tiao et al. selection criteria are shown in the lower panel

of Figure 3. Measurements are made weekly in Canada and 2-3 times a week at the European stations; measurements were made about weekly in Japan before 1975 and after 1989, with infrequent data and few summer measurements in between (Table 3). The frequency of measurements is reflected in the variability in the monthly values. The stricter criteria used by Tiao et al. introduces more gaps in the time series at most stations, as shown in Table 3, and may also increase the variance in the monthly means as they are derived from fewer soundings. Time series up to 1991 to 1993 for are shown in Logan [1994] for most sonde stations using WOUDC data.

The three European stations are the closest together of the sonde locations. There do not appear to be any major biases in ozone concentrations near 90 hPa, except for the early 1970s where values at Hohenpeissenberg are higher than those at Uccle and Payerne (Figure 4).

Trends for 1970-96.

Figure 5 shows seasonal trends in the vertical distribution of ozone for 1970-96, while Figure 6a shows annual trends, all in percent per decade. The solid lines are the results of LM and the dotted lines those of Tiao et al. Results are shown for pressure levels centered below 13 hPa, ~29 km. There are concerns about the quality of the non-ECC data above ~28 km [WMO, 1998], so the trends in the top level should be viewed with caution for these stations.

The seasonal trends computed by each group usually agree within $\pm 3\%$ /decade for most stations, and in the stratosphere almost all trends agree within their standard errors. Agreement is worst for Churchill, with differences of $\pm 10\%$ /decade in winter and summer; this station suffers from serious data loss with the selection criteria of Tiao et al.. Annual mean trends agree within 2%/decade in the stratosphere, with the exception of results from Payerne and Uccle: as discussed in Section 2.2, these stations have the largest trends in correction factors for 1970-96.

Figure 6b shows the annual trends in absolute units (DU/km/decade). This illustrates the contribution of different altitudes to the column trend in ozone. This comparison avoids the problem introduced by percentage trends being referenced to the mean at different times. Figure 6b shows that the two groups obtain similar trend results except for Uccle and Payerne above 50

hPa. The dashed line in Figure 6b shows trends computed with the LM model, but with the data selection criteria of Tiao et al. and with their practice of removing the scaling to the ozone column. When the two groups treat the data in the same way before deriving trends, their results are similar at most altitudes at most stations (compare dotted and dashed lines), and the major discrepancies at Uccle and Payerne are removed.

In order to separate the effects of data selection and of removing the scaling to the ozone column, trends were calculated with the LM model using the Tiao et al. data selection criteria, with and without dividing by the correction factor. Results are shown in Figure 7 (in percent per decade); the difference between the solid line and the dashed line is caused by data selection criteria, and the difference between the dashed line and the dotted line is caused by removing the scaling to the ozone column (i.e., dividing by the CF).

The different trends in Figure 7 are due primarily to the data selection criteria for stations with no trend in the correction factor, such as Sapporo and Kagoshima. Conversely, the different trends are due primarily to the trend in the correction factors for Hohenpeissenberg, where the selection criteria were almost the same (see Table 4). The effect on trends of dividing by the correction factor is largest (2-3 %/decade) for Uccle, Payerne, and Wallops Island, the stations with the largest trend in the correction factors. For the other stations, the effect of dividing by the correction factor is <2 %/decade. Dividing by the CF generally shifts the profile to smaller or larger trends depending on the magnitude of the trend in the CF. Changing the data selection criteria sometimes changes the shape of the vertical profile for the trend; effects are usually largest in the lowermost stratosphere where ozone is most variable. Changing the data selection criteria causes differences of less than 2.5 %/decade in annual trends for most stations, but differences are as large as 5-7%/decade for some pressure levels at Wallops Island, Churchill, and Edmonton. Differences in seasonal trends (not shown) are somewhat larger than differences in annual trends, but most are also within 2.5 %/decade. In addition to changing the magnitude of the trends, the stricter data selection criteria increase the standard errors of the trends, as may be seen in Figure 7.

Figure 8 shows the column trend for 250 to 16 hPa in DU per decade. Trends derived by LM for stratospheric ozone are generally more negative than those derived by Tiao et al.; however, when the LM model is run with the data treatment of Tiao et al., there is much less difference compared to the results of Tiao et al. The implication of these results and those in Figure 6b is that the treatment of data prior to trend analysis can have more effect on derived trends than the details of the statistical model. The inclusion of autocorrelation in the Tiao et al. model gives standard errors that appear only slightly larger than those in the LM model for Hohenpeissenberg (see Figure 5), where the data selection criteria were almost the same.

Trends in ozone are largest in the lower stratosphere, with maximum annual trends of -3 to -10 %/decade (Figure 9). Statistically significant decreases in ozone are found by both groups at all the stations analyzed here for 1970-96. The ozone decrease is located between about 30 hPa and the tropopause. These results are similar to those shown in previous analyses of WOUDC data that ended a few years earlier [Logan, 1994; Miller et al., 1995; WMO, 1995; Harris et al., 1997]. However, the decreases in ozone for 1970-91 were not statistically significant for all stations [Logan, 1994]. Bojkov and Fioletov [1997] analyzed WOUDC data for Hohenpeissenberg and Edmonton relative to the tropopause height, and found significant decreases 1-2 km above the tropopause.

The trends from the different stations form a reasonably compact band from 125 to 30 hPa, as shown in Figure 9. Both groups find largest decreases at the Canadian stations and Sapporo, Japan, and smallest decreases in ozone at the European stations and Wallops Island. The increase in ozone above 50 hPa at Uccle (Tiao et al. analysis) is an anomaly, likely caused by removing the scaling to the ozone column; the CFs showed showed a large jump in 1989 (Figure 1). Both negative and positive trends are seen near 13 hPa (29.5 km), but these results may be less reliable than those at 20 hPa (27 km) and below.

The mean trend for northern mid-latitudes (36°-59°N) is shown in Figure 10. There are significant decreases in ozone from 250 to 30 hPa, and both groups find a mean trend of -7 %/decade from 200 to 80 hPa. The trend from Tiao et al. is about 1 %/decade less negative than

- 14 -

the trend from LM for 80 to 15 hPa. This may be caused in part by the removal of the corrections factors in the Tiao et al. analysis; the mean trend for the CFs for the stations in Figure 10 is -1 %/decade (Table 4). Referencing the trend to 1970 (Tiao et al.) rather than the mean over 1970-96 (LM) may also give less negative trends. The error shown for each layer is the usual standard error for the sample mean of the nine annual trends: this error was larger for all layers than the root mean square error of the standard errors of the annual trend estimates for the nine individual stations.

Figure 11 shows the ensemble of seasonal trends for 36°-59° N (LM results). The narrowest band of trends is found in spring near the ozone maximum, where it is easiest to measure ozone. There is a seasonal variation in the trends that appears to depend on region, as disussed by Logan [1994] and Bojkov and Fioletov [1997]. Ozone decreases are largest in spring and summer at the Canadian stations, and in winter and spring at the European stations, as shown in Figure 12. There is no significant decrease in ozone below 90 hPa in summer and autumn at the European stations, while the decrease persists to 200 hPa in spring. The decreases in ozone are larger for the Canadian than for the European stations, and persist to 200 hPa and below in all seasons. The Canadian trends for 1970-96 are subject to some uncertainty, given the concerns about the BM data for the 1970s [WMO, 1998]. Sapporo and Tateno in Japan also show largest decreases in winter and spring.

Figure 13 shows the composite seasonal behavior for stations located between 36° and 53° N. Both groups find strong seasonality in the trends from about 300 to 90 hPa, but the seasonal patterns are somewhat different. Both groups show largest losses in ozone in spring and smallest losses in autumn. Tiao et al. find the ozone decrease in winter is almost as large as in spring, while LM find the winter decrease is smaller than in spring.

Comparison of trends for 1980-96 with 1970-96.

Annual trends for 1980 to 1996 are shown in Figure 14, while the mean trend for 36°-59° N is shown in Figure 15 (solid line). The results are compared to the trend for 1970-96 (dashed line). The decrease in ozone in the lower stratosphere is larger for the period 1980-96 than for

the period 1970-96 for about half the stations. The mean trend is more negative by 1-2%/decade, with a maximum trend in the lower stratosphere of -9 %/decade. The errors in the seasonal trends are larger for the shorter period. The summer trends for the Japanese stations have extremely large errors because of the paucity of data in much of the 1980s (Table 3) and these contribute to the large errors in the annual trends. The errors in the mean trend for 1980-96 are somewhat larger than those for 1970-96.

Figure 14 includes results for three relatively new stations. There are significant decreases in ozone at Boulder from 80 to 20 hPa, while Hilo shows a significant decrease only at 80 hPa; it shows a significant increase at 15 hPa. There are no significant trends in the lower stratosphere at Lauder (1986-96), but there is an increase of ~5%/decade at 30 hPa, similar to that reported by Bodeker et al. [1998]. The data for Lauder were not scaled to the ozone column, and there is a small but significant trend in the correction factor (Table 4). For the Lauder data scaled to the ozone column, trends in the lower stratosphere are close to zero and are insignificant except at 30 hPa. Bodeker et al. [1998] present a detailed analysis of trends in the Lauder data, and Cunnold et al. [1999] discuss the Lauder data in the context of TOMS and SAGE data for southern mid-latitudes. Although the sonde data suggest an increase in ozone after 1986, the column data from Lauder, as well as TOMS and SAGE data, show a decrease in stratospheric ozone since 1979 [Bodeker, pers. comm., 1999; Cunnold et al. 1999].

Differences in trends derived from SPARC and WOUDC data.

Tiao et al. applied their trend model to the SPARC data and to the data archived at WOUDC for Hohenpeissenberg, Payerne, and the Japanese stations. Results from the two data sets are fairly similar for the stratosphere. The WOUDC data give slightly more negative trends than the SPARC data for the lower stratosphere at Hohenpeissenberg in all seasons. The results for Payerne and the Japanese stations are similar for both data sets from about 80 to 20 hPa.

4.2 Troposphere

Time series.

Figure 16 shows monthly mean values of ozone near 500 hPa for three stations. The data from Goose Bay are from BM sondes before August 1980, and ECC sondes thereafter. Both statistical models used an intervention term to account for any shift in ozone values at the time of the instrument change.

Ozone values at Hohenpeissenberg are systematically higher than those at Payerne from about 1978 to 1990, while there is less bias before 1978, as shown in Figure 17 [cf Logan, 1994]. Values at Uccle are higher than those at Payerne and at Hohenpeissenberg up to about 1986. These biases result in different trends for the European stations. The data for Payerne are provisional, as discussed in WMO [1998], and may be revised further; there are particular concerns about the consistency of the tropospheric data in the 1980s and for 1990-93.

Trends for 1970-96.

There are significant spatial variations in the magnitude and sign of the trends in tropospheric ozone (Figures 5, 6, and 11), with increases found at the European and Japanese stations, and decreases, or no significant trend at the Canadian stations. The increases over Europe and at Kagoshima are significant between the surface and 300 hPa, while the increases at Sapporo and Tateno are significant only up to 500 hPa. The tropospheric trends do not vary significantly with season (e.g., Figures 5 and 12). The Tiao et al. trends are more positive than the LM trends for the European stations and Wallops Island, less negative for the Canadian stations, and about the same for the Japanese stations. Tiao et al. find increases of 6-25 %/decade for the European stations, while LM find increases of 5-15 %/decade. This difference is caused in part by referencing the trend to the beginning rather than the middle of the record. At the lowest layer at Hohenpeissenberg, for example, the Tiao et al. trend is 16 %/decade, and the LM trend 10 %/decade; however, the Tiao et al. trend would be only 12 %/decade, if referenced to the same value as the LM trend. Similarly, the Tiao et al. trend in the lowest layer at Wallops Island would be reduced from ~3 %/decade to 1.2 %/decade (similar to the LM trend) if referenced to the same value as the LM trend. The results from the two groups for Hohenpeissenberg and Payerne for the absolute trend (Figure 6b) and for the tropospheric column trend (Figure 18) are

- 17 -

in better agreement than the results for the percentage trends, also indicating that the reference point makes an important difference to the latter. The two groups find similar results for the column trends for 1000-250 hPa for most stations, and using the LM model with the data treatment of Tiao et al. generally improves the agreement.

The ozone increases in Europe and Japan from LM for 1970-96 are somewhat less than those derived for 1970-91 from data archived at WOUDC [Akimoto et al., 1993; Logan, 1994]. This is caused by the up and down nature of the ozone variations in the last few years (see Figure 16). Bojkov and Fioletov [1997] find the increase at Hohenpeissenberg to be significant 1 km below the tropopause. Logan [1994] concluded that there was no evidence for a long term increase in ozone at the Canadian stations by comparing data for the first 5 years with data for 1987-91, given the different responses of BM and ECC sondes. This contradicted the results of Wang et al. [1993] who reported an increase of 10 %/decade for the Canadian stations. In this work, LM find long term decreases for the Canadian stations (-2 to -9 %/decade), while Tiao et al. find similar decreases or no significant trend. Oltmans et al. [1998] find an increase of 15 %/decade for Hohenpeissenberg for 1968-95, using a least squares fit to annual mean values. They also find no trend at Wallops Island and results very similar to those in Figure 6 for Tateno. They selected these stations for the consistency of their record, and used the data at WOUDC.

Trends for 1980-96.

Trends for the many of the stations for 1980-96 are substantially different from those for 1970-96 (Figure 14). Of the European stations, Uccle shows no trend in ozone, Hohenpeissenberg has a slight negative trend in the middle troposphere, and only Payerne shows a positive trend, ~10 %/decade. There are concerns about the consistency of some of the tropospheric data for Payerne as discussed above. There is no trend in ozone at Tateno, while Sapporo and Kagoshima have increases of 5-15 %/decade, not all of which are significant. The many gaps in the data record for these stations in the early 1980s give rise to large errors in the summer and annual trends as discussed above. The Canadian stations have decreases of -2 to -8 %/decade, and these are more reliable than the results for 1970-96 (ECC sondes only). Previous analyses of

the Canadian ECC data also gave decreases [Logan, 1994, Tarasick et al., 1995; Oltmans et al., 1998]. Oltmans et al. [1998] find no significant trend in ozone for Hohenpeissenberg, Wallops Island, Boulder, Tateno, and Hilo for 1979-95 in the middle troposphere, in agreement with the results in Figure 14. The increases in ozone in the late 1960s and 1970s seem to have levelled off at several of the sonde stations, and also at remote surface sites, as discussed earlier by Logan [1994] and by Oltmans et al. [1998]. The mean trend for the northern mid-latitudes is zero, 4%/decade less than the mean trend for 1970-96 (Figure 15). The decrease is caused by the less positive trend for the later period at the European stations and Tateno. The concept of a average mid-latitude trend is less appropriate for the troposphere than for the stratosphere. The locations of the sonde stations in remote regions of Canada, and more polluted regions of Europe and Asia, may lead to different regional influences on tropospheric ozone from trends in emissions of NO_x and CO, changes in stratospheric input of ozone, and changes in weather [e.g., Logan, 1994]. There are not enough stations to define a realistic statistical average of tropospheric trends even for northern mid-latitudes.

Differences in trends derived from SPARC and WOUDC data.

Tiao et al. derive similar tropospheric trends using the SPARC and WOUDC data for Hohenpeissenberg and the Japanese stations. The results for the two data sets are significantly different for Payerne, with much larger tropospheric increases derived for the WOUDC data than for the data used here (e.g., 4.3 %/decade vs. 2.4 %/decade at 600 hPa). The WOUDC archive contained erroneously high values for ozone in early 1990s, until these data were later withdrawn. Miller et al. [1995] reported anomalously high trends for Payerne, based on the WOUDC data. The cause of the unrealistic values for Payerne in the early 1990s was an electronics problem that occurred when the type of meteorological sonde was changed. The data were subsequently corrected [Stubi et al. 1998], and provided for the SPARC analysis. Further revisions are expected after more work on homogenizing the Payerne record.

4.3 Comparison of sonde, Dobson, and TOMS trends at the sonde locations.

Column trends were derived from the sonde trends (in DU) by integrating from the surface to ~16 hPa, omitting trends derived from the less reliable data near the top of the soundings. These are compared to trends derived from column data obtained with Dobson or Brewer instruments on the same day as the soundings. (The column data were sometimes unavailable for the Canadian ECC sondes and Wallops Island, and are generally unavailable for Resolute and Churchill in winter). The trends in the column data were obtained using the LM model, omitting measurements on days that did not meet the CF criteria for sondes used by LM. Figure 19 compares these column trends and the integrated sonde trends for both groups. The LM results agree somewhat better with the column trends for the European stations, while the Tiao et al. results underestimate the column loss more than the LM results. This is likely caused by the removal of the scaling to the ozone column in the Tiao et al. analysis; there is a negative trend in the CFs at each European station. For the Canadian and Japanese stations, there is no systematic bias between the two sets of results with respect to the column trends. At the Canadian stations, an intervention term was used in the statistical models independently at each level, so it is less likely that the integral of the sonde trends will equal the column trends. If there is a decrease in ozone above 16 hPa, the integrated sonde trends should be less negative than the column trends, which is sometimes but not always the case. The SAGE data for 1979-96 indicate that the seasonal trends in the ozone column above 16 hPa for 40°-50°N are -2 to -4 DU/decade [W. Randel, personal communication, 1998].

The column trends derived from TOMS by Hollandsworth for Nov. 1978 to Oct. 1994 [WMO, 1998] are compared to the integrated sonde trends and Dobson/Brewer column trends in Figure 20. The three trends agree within their standard errors; agreement is best for the European stations, Boulder and Tateno. The TOMS data confirm that the percent decrease in ozone (not shown) is largest in spring and summer at the Canadian stations (except Goose Bay with largest losses in autumn) and in winter and spring at the European stations. Figure 20 is not the best way to evaluate the differences between the ground based and TOMS ozone columns, as it uses the ground based column data only on the days when sondes were available; its purpose is

the comparison with the sonde data.

5. Discussion and Conclusions.

5.1 Analysis methodologies.

Results of the two analyses of trends in stratospheric ozone give fairly similar profiles for ozone loss, especially when viewed as annual trends (Figures 6 and 9) or as an average over several stations (Figure 10). Annual trends derived by the two groups agree within 2%/decade, and agree within their standard errors with the exception of Uccle and Payerne. There are larger differences in details of the seasonal trends at individual stations, as discussed above.

The two groups selected the data for analysis in different ways, LM used the data scaled to the ozone column, Tiao et al. did not, and they used different statistical models. The differences in the latter are the assumption of zero trend prior to 1970, the inclusion of autocorrelation, the treatment of outliers, and the method of weighting, including the iterative procedure. All can contribute to differences in results, although the first does not apply to Churchill, Edmonton, and Wallops Island which have no data prior to 1970, nor to Goose Bay where none of the 1969 data meet the Tiao et al. selection criteria. The zero trend assumption was designed to mimic the effect of chlorine on stratospheric ozone and is not appropriate for the troposphere, where increases in NO_x are thought to be the primary cause of trends in ozone. Running the LM model with the data treatment of Tiao et al. indicated that the primary reason for different results of the two analyses appears to be the treatment of data prior to trend analysis.

It is important that the reference point for calculating percentage trends be given, since it influences the magnitude of the relative trends. The use of the beginning of the time series rather than the mean can make deceasing trends appear less negative and increasing trends appear more positive, with the effect being largest for largest trends. For sparse time series, the reference point may be less well defined if it is based on the intercept of the fit rather than on the mean.

Which trend results for 1970-96 are likely to be more reliable? Here we offer some comments and make suggestions for future work. First we note that each group chose a set of criteria for treating the data, and a trend model that they thought to be defensible. As we have shown above, the results are robust although there are differences in detail.

The major difference between the two analyses is the data selection criteria. LM use less strict criteria, with the goal of maximizing the amount of data of reasonable quality available for analysis. The criteria of Tiao et al. are designed to maximize the quality of data used in trend analysis, but the end result is the loss of 55% of the Payerne data, 60-75% of the Canadian BM data, and 35-55% of the Uccle, Wallops Island, Canadian ECC, and Japanese data (70% for Churchill) (Table 2). Gaps are introduced in the time series, and they become noisier.

It is recommended in WMO [1998] that the sonde data should be scaled to the ozone column for derivation of reliable stratospheric data; Tiao et al. selected their data treatment prior to this recommendation, and removing the scaling to the ozone column in their standard model is a new approach for them. Their previous analyses [Tiao et al., 1986; Miller et al., 1995] have used the data scaled to the ozone column, and they have criticized the approach of removing the scaling [Tiao et al., 1986]. Logan [1985, 1994] and Miller et al. [1995] have shown trends with and without dividing by the correction factor, to isolate its effect; the trends appear more reliable when the data are scaled to the ozone column, because of the trends in the correction factors at some stations. Payerne and Uccle have jumps in the correction factors at the time of procedural changes or changes in the algorithm used to derive the ozone profile, as discussed above (see also WMO [1998]). Any offsets or trends in the correction factor make it inappropriate to remove the scaling to the ozone column before deriving trends (unless an intervention term is included in the statistical model to account for the offset). For the stations analyzed here, the effects are largest for Uccle, Payerne and Wallops Island; dividing by the CFs makes the trends less negative by 2-3%/decade for 1970-96, and for the case of Uccle, makes the stratospheric trends appear as outliers compared to the other stations. The effects are potentially larger for 1980-96 where the trends in the CF are somewhat larger for several stations, but Tiao et al. did not analyze the data for this period.

Tiao et al. [1990] and Weatherhead et al. [1998] show the possible importance of including

- 22 -

autoregressive errors within the statistical trend estimation procedure. This appears to have a relatively minor effect on errors derived for trends at Hohenpeissenberg, where the data selection was nearly the same (Figure 6). The treatment of the data prior to deriving trends has a larger impact on trends and associated errors. The errors on the trends derived with the LM model using the Tiao et al. data selection criteria are larger than those with the LM criteria (Figure 7) as the time series are noisier. The errors derived by Tiao et al. are generally larger than those derived by LM (Figure 6b), but the reasons for this are unclear, given the differences in data selection and in statistical models. The effect on sonde trends and errors of autoregression, removal of outliers, and treatment of weighting is under further investigation by Tiao et al., and will be reported elsewhere.

The optimal selection of sonde data for trend analysis is clearly a subject of debate, given the different approaches adopted by two groups working independently with the same data. If one wishes to maximize both data quality and quantity, advantages of both approaches could be blended, e.g., the less restrictive correction factors used by the LM group, requiring that the balloon reach 20 hPa (less strict than 16 hPa), and requiring a measurement of the ozone column (as required by Tiao et al.), and keeping the normalization to the ozone column. This set of criteria would require a check on the profile measurement using another technique, yet not exclude so much data. With the proposed criteria, 80-93% of the reevaluated data sets and of the Wallops Island data would be retained (Table 2). For the Canadian stations, 45-65% of the BM data and 40-80% of the ECC data would be retained. Using these conditions in the LM model, the results are almost identical to those in Figure 5 and 6 (LM results) for the European stations and Sapporo, and very similar to those for the other stations, with largest differences (1-3 %/decade) in the lower stratosphere for the Canadian stations.

Reevaluation could clearly improve the Canadian ECC data set. About 20% of the soundings fail to reach 20 hPa and many ozone column measurements are missing. Wallops Island has a similar problem with missing ozone column data. Reprocessing of the data after 1980 using TOMS data to derive ozone columns would solve the latter problem. The SBUV profile

- 23 -

climatology could be used to derive the top of the profile for soundings that reach 20 hPa, allowing a correction factor to be derived [McPeters et al., 1997].

5.2 Trend Results.

Stratospheric trends. The two analyses shown here demonstrate that there is a statistically significant decrease in ozone in the northern mid-latitudes between 250 and 30 hPa (10.5-25 km) from 1970 to 1996, and that the largest decrease, -7 %/decade, is located between 200 and 80 hPa (11.5-18 km). All stations show significant decreases in ozone, with a range of -3 to -10 %/decade near 100 hPa (17 km). The decreases are largest at the Canadian stations and the most northerly Japanese station, and are smallest at the European stations and Wallops Island. For trends starting in 1980, the decrease in ozone is more negative by 1-2 %/decade, with a maximum trend in the lowermost stratosphere of -9 %/decade. The trends in ozone derived from ozonesondes for 40°-53°N are in excellent agreement with trends derived from SAGE I and II from 15 to 28 km [Cunnold et al., 1999].

The seasonal variation in the trends is located primarily in the lowermost stratosphere, between about 250 and 90 hPa (10-17) km, with little seasonality in the trends above 20 km. The seasonal variation depends on region, with largest decreases in winter and spring at the European stations, and at the two most northerly Japanese stations. There is no significant decrease below 90 hPa at the European station in summer and autumn, while the decrease persists to 200 hPa in spring. Decreases are largest in spring and summer at the Canadian stations, and persist to 200 hPa and below in all seasons. The results of the two analyses are most different for the Canadian seasonal trends. The Canadian Brewer-Mast data are of questionable reliability [WMO, 1998], so the trend results are less reliable than those for other stations. The seasonal trends for the ECC Canadian data starting in 1980 indicate largest decreases in spring (not shown).

Tropospheric trends. Trends in ozone are highly variable, and depend on region. There are decreases or insignificant trend at the Canadian stations for 1970-96, and decreases of -2 to -8 %/decade in the mid-troposphere for 1980-96. The European stations show increases of 5-25

- 24 -

%/decade which are significant from the surface to 300 hPa (9 km) for 1970-96; trends are close to zero for two stations after 1980, and only Payerne gives an increase, 10 %/decade for 1980-96. The Payerne data are subject to revision. The increases at the Japanese stations are largest near the surface, 10-15 %/decade for 1970-96, and decrease with increasing altitude in the troposphere. They are insignificant by 9 km at the two northerly stations, and by 12 km at Kagoshima (32° N). Tateno, the Japanese station with most data, shows no trend in ozone for 1980-96, while Sapporo and Kagoshima give increases that are not always significant. There is no significant trend in ozone at the U.S. stations, Wallops Island (for both periods), or Boulder and Hilo for the later data. The variability in tropospheric trends combined with the small number of mid-latitude stations makes it impossible to define reliably a mean tropospheric trend.

Consistency of sonde and column trends. The integrated column trend derived from the sonde data should be consistent with the Dobson and Brewer column data, since the individual soundings are scaled to the column measurement, at least for the LM analysis. The column trends derived from both sonde analyses agree with the column trends for 1970-96 for most stations and seasons. The column trends for 1980-96 from the LM analysis agree also with the column trends derived from ground-based and TOMS measurements.

5.3 Implications of this study for future profile measurements and analyses.

One of the primary motivations for continuing to measure the vertical profile of ozone at the sonde stations analyzed here is to monitor changes in the vertical distribution of ozone. Considerable effort and expense is put into obtaining these data, yet a large fraction of the soundings are rejected in many trend analyses, depending on the data selection criteria chosen. The majority of stations make measurements once a week (Table 3). With only four potential measurements to characterize ozone in a given month, we can ill afford to have these data rejected in subsequent analysis. The quality of data at the long-term stations needs be assured. If a particular sounding does not pass an acceptable criterion with respect to the CF, or with respect to other measures of quality, an additional sounding could be flown. It appears that a consensus is required as to what constitutes an acceptable sounding. The dialogue needs to continue also on

- 25 -

treatment of sonde data prior to trend analysis.

Acknowledgements.

--

The work performed at Harvard University was funded with support from the National Air and Space Administration, grants NAGW-2632, and NAG1-1909, and the National Science Foundation, grant ATM-9320778. The work at the University of Chicago was supported by the National Oceanic and Atmospheric Administration under the Office of Global Programs.

References.

Akimoto, H., H. Nakane, and Y. Matsumoto, The chemistry of oxidant generation: tropospheric ozone increase in Japan, in *Chemistry of the Atmosphere, The Impact on Global Change*, ed. J.W. Birks, J. G. Calvert, R. E. Sievers, pub. American Chemical Society, Washington, DC, 1993.

Angell, J.K. and J. Korshover. Global variation in total ozone and layer mean ozone: an update through 1981. *J. Climate and Appl. Met.* 22, 1611-1626, 1983.

Attmanspacher, W., and H. U. Dutsch, International ozonesonde intercomparison at the Observatory Hohenpeissenberg, January 19 to February 5, 1970, *Ber. Dtsch. Wetterdienstes*, *16*(120), 1970.

Bass A. M., and R. J. Paur, The ultraviolet cross-sections of ozone: I. The measurements, in *Atmospheric ozone*, ed. C. S. Zeferos and A. Ghazi, Reidel, Dordrecht, Boston, Lancaster, pp 606-610, 1985.

Bodeker, G. E., I. S. Boyd, and W. A. Matthews, Trends and variability in vertical ozone and temperature profiles measured by ozonesondes at Lauder, New Zealand: 1986-1996, *J. Geophys. Res.* 103, 28661-28681, 1998.

Bojkov, R. D., V. E. Fioletov, Change of the lower stratospheric ozone over Europe and Canada, *J. Geophys. Res.*, *102*, 1337-1347, 1997.

Claude, H., R. Hartmannsgruber, and U. Kohler, Measurement of atmospheric ozone profiles using the Brewer/Mast sonde, WMO Global Ozone Research and Monitoring Project, Report No. 17, WMO/TD No. 179, 1988.

Cunnold, D. M., H. J. Wang, L. Thomason, J. Zawodny, J. A. Logan, and I. A. Megretskaia, SAGE (v.5.96) ozone trends in the lower stratosphere, *J. Geophys. Res.*, submitted, 1998.

De Backer, H., D. De Muer, E. Schoubs, and M Allaart, A new pump correction for Brewer-Mast ozonesondes, Proc. 18th Quadrennial Ozone Symposium, Ed. R. Bojkov and G. Visconti, Parco Scientifo and Tecnologico D'Abruzzo, Italy, Vol.2, pp. 891-894, 1998a.

De Backer, H., D. De Muer, and G. De Sadelaer, Comparison of ozone profiles obtained with Brewer-Mast and Z-ECC sensors during simultaneous ascents, *J. Geophys. Res.*, 103, 19641-19648, 1998b.

De Muer, D., and H. De Backer, Influence of sulfur dioxide trends on Dobson measurements and on electrochemical ozone soundings, Atmospheric ozone conference, Tromso 28-29 June 1993, SPIE proceedings series, Vol. 2047, 18-26, 1994.

Dutsch, H. U., Two years of regular ozone soundings over Boulder, Colorado, National Center for Atmospheric Research, 1966.

Hansen, J., M. Sato, and R. Ruedy, Radiative forcing and climate response, *J. Geophys. Res.*, *102*, 6831-6864, 1997.

Harris, N. R. P., G. Ancellet, L. Bishop, D. J. Hoffman, J. B. Kerr, R. D. McPeters, M. Prendez,W. J. Randel, J. Staehelin, B. H. Subbaraya, A. Volz-Thomas, J. Zawodny, and C. S. Zerefos,Trends in stratospheric and free tropospheric ozone, *J. Geophys. Res.*, 102, 1571-1590, 1997.

Hilsenrath, E., et al., Results from the balloon ozone intercomparison campaign (BOIC), J. Geophys. Res., 91, 13137-13152, 1986.

Hollandsworth, S.M., R.D. McPeters, L.E. Flynn, W. Planet, A.J. Miller, and S. Chandra, Ozone trends deduced from combined Nimbus 7 SBUV and NOAA 11 SBUV/2 data, *Geophys. Res. Lett.*, 22, 905-908, 1995.

Logan, J.A., Trends in the vertical distribution of ozone: An analysis of ozonesonde data, J. *Geophys. Res.*, 99, 25553-25585, 1994.

McPeters, R. D., G. J. Labow, B. J. Johnson, A satellite-derived ozone climatology for balloonsonde estimation of total column ozone, *J. Geophys. Res.*, *102*, 8875-8885, 1997.

Miller, A. J., L. E. Flynn, S. M. Hollandsworth, J. J. Luisi, I. V. Petropavlovskikh, G. C. Tiao, G. C. Reinsel, D. J. Wuebbles, J. Kerr, R. M. Nagatani, L. Bishop, C. Jackman, Information content of Umkehr and solar backscattered ultraviolet (SBUV) 2 satellite data for ozone trends and solar responses in the stratosphere, *J. Geophys. Res.*, *102*, 9257-19263, 1997.

Miller, A. J., R. M. Nagatani, G. C. Tiao, X. F. Niu, G. C. Reinsel, D. Wuebbles, and K. Grant, Comparisons of observed ozone and temperature trends in the lower stratosphere. Geophys. Res. Lett., 19, 929-932, 1992.

Miller, A. J., G. C. Tiao, G.C. Reinsel, D. Wuebbles, L. Bishop, J. Kerr, R.M. Nagatani, J.J. deLuisi, and C. L. Mateer, Comparisons of observed ozone trends in the stratosphere though examination of Umkehr and balloon ozonesonde data, *J. Geophys. Res.*, 100, 11,209-11218, 1995.

Newchurch, M. J., and 19 co-authors, Upper stratospheric ozone trends, 1979-1996, *J. Geophys, Res.*, to be submitted, 1998.

Oltmans, S. J. and 16 others, Trends of ozone in the troposphere, *Geophys. Res. Lett.* 25, 139-142, 1998.

Ramaswamy V., M. D. Schwarzkopf, and W. J. Randel, Fingerprint of ozone depletion in the spatial and temporal pattern of recent lower-stratospheric cooling, *Nature*, 382, 616-618, 1996.

Randel, W., R. Stolarski, D. Cunnold, J. A. Logan, and M. J. Newchurch, Trends in the vertical distribution of ozone, *Science*, to be submitted, 1998.

Stubi, R., V. Bugnion, M. Giroud, P. Jeannet, P. Viatte and B. Hoegger, Long term ozone balloon sounding series at Payerne: Homogenization methods and problems, Proc. 18th Quadrennial Ozone Symposium, Ed. R. Bojkov and G. Visconti, Parco Scientifo and Tecnologico D'Abruzzo, Italy, in press, 1998.

Tarasick, D. W., D. I. Wardle, J. B. Kerr, J. J. Bellefleur, and J. Davis, Tropospheric ozone trends over Canada: 1980-1993, *Geophys. Res. Lett.*, 22, 409-412, 1995.

Tiao, G.C., G.C. Reinsel, J.H. Pedrick, G.M. Allenby, C.L. Mateer, A.J. Miller, and J.J. DeLuisi, A statistical analysis of ozonesonde data, *J. Geophys. Res.*, *91*, 13,121-13,136, 1986.

Tiao, G.C., G.C. Reinsel, D. Xu,J.H. Pedrick, X. Zhu, A.J. Miller, J.J. DeLuisi, C. L. Mateer, and D.J. Wuebbles, Effects of autocorrelation and temporal sampling schemes on estimates of trend and spatial correlation, *J. Geophys. Res.*, *95*, 20507-20517, 1990.

Wang, W-C., Y-C. Zhuang, and R. D. Bojkov, Climate implications of observed changes in ozone vertical distributions at middle and high latitudes of the northern hemisphere, *Geophys. Res. Lett.*, 20, 1567-1570, 1993.

Wang H. J., D. M. Cunnold, X. Bao, A critical analysis of Stratospheric Aerosol and Gas Experiment ozone trends, *J. Geophys. Res. 101*, 12495-12514, 1996.

Weatherhead, E.C., and 12 others, Factors affecting the detection of trends: Statistical considerations and applications to environmental data, *J. Geophys. Res.*, *103*, 17149-17161, 1998.

World Meteorological Organization, Scientific Assessment of Ozone Depletion: 1994, Global Ozone Research and Monitoring Project - Report No. 37, Geneva, Switzerland, 1995.

World Meteorological Organization, Assessment of Trends in the Vertical Distribution of ozone, SPARC Report No. 1, Global Ozone Research and Monitoring Project Report, No. 43, World Meteorological Organization, 1998.

ID	Station	Lat.	Long.	Туре	Period
SPARC	C DATA				
53	Uccle	51	4	BM	1/69-12/96
99	Hohenpeissenberg	48	11	BM	11/66-12/96
156	Payerne	47	7	BM	11/66-12/96
12	Sapporo	43	141	KC	12/68-12/96
67	Boulder	40	-105	ECC	3/79-12/96
14	Tateno	36	140	KC	11/68-12/96
7	Kagoshima	32	131	KC	1/69-12/96
109	Hilo	20	-155	ECC	9/82-12/96
256	Lauder	-45	170	ECC	8/86-12/96
WOUD	C DATA				
24	Resolute	75	-95	BM	1/66-11/79
				ECC	12/79-2/96
77	Churchill	59	-94	BM	10/73-8/79
				ECC	9/79-12/96
21	Edmonton	53	-114	BM	10/72-8/79
				ECC	9/79-12/96
76	Goose Bay	53	-60	BM	6/69-8/80
	·			ECC	9/80-12/96
107	Wallops Is.	38	-76	ECC	5/70-5/95

Table 1: Sonde data used in the analysis.

Adapted from WMO [1998].

--

Brewer Mast data	Years	0.9-1.35	0.9-1.35	0.9-1.35	0.9-1.2	0.9-1.2	0.9-1.2
			20 hPa	20 hPa		16 hPa	16 hPa
Station				Column			Column
Uccle	70-96	0.87	0.84	0.84	0.58	0.54	0.54
Hohenpeissenberg	70-96	-	-	-	0.93	0.91	0.91
Payerne	70-96	0.83	0.80	0.80	0.47	0.44	0.44
Resolute	70-79	0.91	0.60	0.46	0.75	0.39	-
Churchill	73-79	0.69	0.49	0.49	0.44	0.25	0.25
Edmonton	72-79	0.78	0.60	0.59	0.53	0.34	0.34
Goose Bay	70-80	0.81	0.65	0.64	0.42	0.32	0.32
ECC data	Years	0.8-1.2	0.8-1.2	0.8-1.2	0.9-1.15	0.9-1.15	0.9-1.15
200 0000		0.0 1.2	20 hPa	20 hPa	017 1110	16 hPa	16hPa
Station				Column			Column
Resolute	80-96	0.96	0.71	0.47	0.90	0.59	-
Churchill	80-06	0.97	0.80	0.41	0.89	0.67	0.31
Edmonton	80-96	0.95	0.84	0.78	0.84	0.68	0.63
Goose Bay	80-96	0.95	0.75	0.61	0.87	0.63	0.50
Sapporo	70-96	0.88	0.83	0.83	0.67	0.60	0.60
Tateno	70-96	0.90	0.85	0.85	0.65	0.59	0.59
Kagoshima	70-96	0.85	0.81	0.81	0.53	0.46	0.46
Wallops	70-96	0.98	0.93	0.80	0.88	0.77	0.66
Boulder	80-96	0.94	0.92	0.92	0.91	0.88	0.88
Hilo	82-96	0.89	0.88	0.88	0.87	0.85	0.85
Lauder	86-96	0.92	0.87	0.87	0.92	0.84	0.84

Table 2: Fraction of soundings that meet various criteria.

The third column gives the fraction of soundings that met the CF criteria required for the LM analysis (except for Hohenpeissenberg); the fourth column gives the fraction that also reach 20 hPa and the fifth column the fraction that also have an ozone column measurement. The sixth column gives the fraction that meet the Tiao et al. CF criteria, the seventh column gives the fraction that also reach 16 hPa, and the eighth column gives the fraction that also have an ozone column measurement. For Boulder, Hilo, Lauder and the Japanese stations, no CF is given if there in no ozone column so the soundings fail the CF criteria. For the BM Canadian stations, a default CF is given which fails the Tiao et al. CF criteria except for Resolute but meets the CF criteria. Resolute is not required to have an ozone column in winter. Wallops Island was given a default CF of 1.0 when no correction factor was available.

Station	Period	Original	LM	Tiao et al.
Uccle	1/70-6/89	9.1(0.85)	7.6(0.83)	3.8(0.71)
	7/89-12/96	11.0(1.0)	10.6(1.0)	9.6(1.0)
Hohenpeissenberg	1/70-12/77	3.5(1.0)	3.0(0.97)	2.9(0.97)
	1/78-12/96	9.9(1.0)	9.4(1.0)	9.2(1.0)
Payerne	1/70-12/75	8.2(0.97)	5.1(0.94)	1.7(0.65)
	1/76-12/96	10.9(1.0)	9.5(1.0)	5.7(0.95)
Resolute	1/70-12/96	3.3(0.97)	3.1(0.94)	2.2(0.75)
Churchill	10/73-12/96	3.3(0.96)	3.0(0.95)	1.8(0.5)
Edmonton	1/73-12/96	3.5(0.99)	3.1(0.98)	2.1(0.85)
Goose Bay	1/70-12/96	3.7(0.98)	3.3(0.96)	2.1(0.72)
Sapporo	1/70-12/74	2.6(0.98)	2.3(0.97)	1.9(0.83)
	1/75-6/89	1.0(0.49)	1.0(0.44)	1.0(0.27)
	7/89-12/96	3.2(0.97)	2.8(0.95)	2.2(0.85)
Tateno	1/70-12/74	2.4(0.78)	2.3(0.75)	1.6(0.64)
	1/75-6/89	1.7(0.64)	1.6(0.63)	1.4(0.55)
	7/89-12/96	4.2(1.0)	3.8(1.0)	2.6(0.91)
Kagoshima	1/70-12/74	2.3(0.95)	2.0(0.92)	1.3(0.60)
	1/75-6/89	1.0(0.51)	1.0(0.47)	1.0(0.23)
	7/89-12/96	3.0(0.95)	2.6(0.92)	1.8(0.78)
Wallops Island	5/70-4/95	2.5(0.78)	2.5(0.78)	2.0(0.65)
Boulder ¹	1/80-12/84	1.5(0.68)	1.5(0.68)	1.3(0.67)
	1/85-12/96	3.8(0.87)	3.6(0.86)	3.5(0.85)
Hilo ¹	9/82-12/96	3.3(0.91)	3.0(0.88)	2.9(0.88)
Lauder ¹	8/86-12/96	5.7(1.0)	5.5(0.96)	5.0(0.96)

Table 3: Numbers of soundings each month before and after data selection.

The table gives the mean number of soundings per month, for months that have at least

--

--

one sounding, followed in parentheses by the fraction of months for which there were soundings in the given period. The statistic given is the trimmed mean, which omits the lowest and highest 5% of the numbers; this was used because occasional months have a large atypical number of soundings. The column "Original" is the data before any selection criteria were applied, the next column is for the LM selection criteria, and the last column for the Tiao et al. selection criteria.

1. Tiao et al. did not analyze trends for these stations.

70-96	80-96
-2.6±0.6	-3.7±1.3
-1.9±0.5	-3.0±0.8
-1.3±0.5	NS
NS	NS
-1.3±0.9	-3.3±1.7
NS	NS
-2.0 ± 1.1	-2.5±1.8
-	-3.7±1.2
-	-3.7±1.3 ^a
-	-1.5 ± 1.5^{b}
70-79	80-96
-2.3±2.5	NS
NS	-2.3±2.4
5.1±6.0	-2.7±1.4
NS	2.0±1.7
	70-96 -2.6±0.6 -1.9±0.5 -1.3±0.5 NS -1.3±0.9 NS -2.0±1.1 - - 70-79 -2.3±2.5 NS 5.1±6.0 NS

Table 4. Trend in correction factors (%/decade).

The trend in the correction factor was calculated using a least squares fit to monthly mean values; two standard errors are given. NS indicates that the trends are statistically insignificant, and most of these are smaller than 1%/decade; values are given for trends that are significant or are close to significant. The Tiao et al. data selection criteria were used, i.e, the sonde reached 16 hPa, there was an ozone column measurement (except for Resolute), and the CF was within the range 0.9-1.2 (BM) and 0.9-1.15 (ECC). Trends are given separately for the two types of sondes for the Canadian stations, and for the two analysis periods at the other stations. Adapted from [WMO, 1998].

a. Trend for 1982-96

b. Trend for 1986-96

Figure Captions.

Figure 1. Correction factors for selected sonde stations. Adapted from WMO [1998].

Figure 2. Annual trends in the vertical distribution of ozone for 1970-96 for Hohenpeissenberg, in percent per decade. The dashed line shows trends computed by LM for the ozone column in 33 layers equally spaced in log presure from 1000 to 6.3 hPa. The solid line shows results for 11 layers obtained by summing the ozone content in 3 consecutive layers. Results are shown below 10 hPa.

Figure 3. Time series of monthly mean values for ozone in DU and deseasonalized monthly means for selected stations. The correction factors used by LM were applied for the top panels (a), and the data selection criteria used by Tiao et al. for the lower panels (b) (see text). Values are shown for one of the 33 levels near 90 hPa, and the same relative scale is used for both sets of means. Adapted from WMO [1998].

Figure 4. Difference of monthly mean values for the three European stations near 90 hPa. The correction factors used by LM were applied (see text). Adapted from WMO [1998].

Figure 5. Seasonal trends in the vertical distribution of ozone for 1970-96. The results of LM are shown by the solid line and the Tiao et al. results by the dotted line. Two standard errors are shown. Trends are plotted at the midpoint of the pressure levels used in each analysis. Updated from WMO [1998].

Figure 6a. Annual trends in the vertical distribution of ozone for 1970-96. The results of LM are shown by the solid line and the Tiao et al. results by the dotted line. Two standard errors are shown. Updated from WMO [1998].

Figure 6b. Annual trends in the vertical distribution of ozone for 1970-96 in DU/km/decade. Pressure was converted to altitude using the U.S. Standard Atmosphere.

The results of LM are shown by the solid line and the Tiao et al. results by the dotted line. Two standard errors are shown. The dashed line shows trends derived with the LM model run with the Tiao et al. data treatment (their selection criteria, and with the data divided by the CF). Updated from WMO [1998].

Figure 7. Sensitivity of annual trends in the vertical distribution of ozone to data treatment prior to trend analysis. All trends were derived with the LM model. The solid line shows results for the LM data selection criteria; the dotted line shows results with the Tiao et al. data selection criteria, and with the data divided by the CF); the dashed line shows results with the Tiao et al. data selection criteria, but without dividing by the CF. Updated from WMO [1998].

Figure 8. Column trend in ozone in DU/decade from 250 to 16 hPa for 1970-96. The triangles are results from LM, the crosses those from Tiao et al., and the circles are results for the LM model, with the data treatment of Tiao et al. (their data selection, and with the data divided by the CF). Updated from WMO [1998].

Figure 9. Annual trends for individual sonde stations located between 36° and 59° N, superimposed. Updated from WMO [1998].

Figure 10. Mean annual trend for the sonde stations located between 36° and 59° N. The solid line shows the LM results, the dashed line the Tiao et al. results. Two standard errors are shown; these were calculated as the usual standard error for the sample mean of the nine annual trends (one third of the standard deviation of the nine trend estimates), for each pressure level. Updated from WMO [1998].

Figure 11. Seasonal mean trends for the sonde stations located between 36° and 59° N, superimposed. LM results. Updated from WMO [1998].

Figure 12. Seasonal mean profiles for three European stations, 48°-51°N (left) and for

three Canadian stations, 53°-59° N (right). LM results. Updated from WMO [1998].

Figure 13. Seasonal mean profiles for stations located between 36° and 53°N. LM results, left panel, and Tiao et al. results, right panel. Updated from WMO [1998].

Figure 14. Annual trends for 1980-96 (solid lines) compared to trends for 1970-96 (dashed lines) where available. The Lauder trends are for 1986-96, and the Hilo trends for 1982-96. LM results. Updated from WMO [1998]. Updated from WMO [1998].

Figure 15. Mean annual trend for the sonde stations located between 36° and 59°N (excluding Boulder), for 1980-96 (solid line) compared to the mean trend for 1970-96 (dotted line). LM results. Two standard errors are show, calculated as in Figure 10. Updated from WMO [1998].

Figure 16. Time series of monthly mean values for ozone in DU Left) and deseasonalized monthly means (right) for selected stations. The correction factors used by LM were applied (see text). Values are shown for one of the 33 levels near 500 hPa, and the same relative scale in used for both sets of means. From WMO [1998].

Figure 17. Difference of monthly mean values for the three European stations near 500 hPa. The correction factors used by LM were applied (see text). From WMO [1998].

Figure 18. Column trend in ozone in DU/decade from the lowest layer to 250 hPa. The triangles are results from LM, the crosses those from Tiao et al., and the circles are results for the LM model, with the data treatment of Tiao et al. (their data selection, and with the data divided by the CF). Updated from WMO [1998].

Figure 19. Comparison of column trends for ozone for 1970-96. The circles show the trend in the overhead ozone column measured on the same days as the sondes used in the LM analysis, computed with the LM model. The crosses show the integrated sonde trend up to 16 hPa, computed with the LM model, and the triangles show the integrated sonde

trend up to 16 hPa computed with the Tiao et al. model. Two standard errors are shown. The errors for the sonde data are only approximate, as they do not account for any correlation between ozone at one layer and the next, and the Tiao et al. errors are smaller in part because they are for 12 layers rather than 9. The three results for each season are offset for clarity. Updated from WMO [1998].

Figure 20. Comparison of column trends for ozone for 1980-96. Circles and crosses show the column and integrated sonde trends defined as in Figure 20. The sonde results are the LM analysis. The triangles show the trend in TOMS data for Nov. 1978 to Oct. 1994, computed by Hollandsworth. The three results for each season are offset for clarity. Updated from WMO [1998].



Correction Factors For Individual Soundings







Trends for 70 - 96



Trends for 70 - 96



Trends for 70 - 96







Annual trends for 70 - 96.



250-16 hPa













Annual trends.







Figure 16. Time series of monthly mean values for ozone in DU and deseasonalized monthly means for selected stations. The correction factors used by the Harvard group were applied (see text). Values are shown for one of the 33 levels near 500 mbar, and the same relative scale in used for both sets of means.



501.22 hPa

1000-250 hPa





