

Trends in the Vertical Distribution of Ozone: A Comparison of Two Analyses of Ozone Sonde Data

by J. A. Logan, et al.

A group of scientists did a year-long study of the observed changes in the amount of ozone as a function of altitude. The study was carried out under the auspices of the GCRP Stratospheric Processes As Related to Climate (SPARC) program and the International Ozone Commission (IOC). One part of this study was the evaluation of trends. This chapter, which was chaired by myself and Bill Randel of NCAR, will result in four papers to be submitted for publication.

11/1/98  
410-827

This paper is the first of those to come from the SPARC/IOC report. It presents analyses from nine ozone sonde stations which have records of about 30 years in length. These stations are located in Japan, Europe, and North America. A number of conclusions can be reached from these data:

- 1) All stations show a significant negative trend for ozone in the lower stratosphere.
- 2) The ozone trends show a seasonal variation which is mostly confined to the altitudes between 10 and 18 km.
- 3) The seasonal variation in the trend has a maximum negative trend in the winter and spring, but the details are different in each of the regions of the globe measured.
- 4) Tropospheric ozone trends are extremely variable such that it is not possible to use the information from nine stations to form a meaningful global or hemispheric average.



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

J. A. Logan<sup>1</sup>, I. A. Megretskaya<sup>1</sup>, A. J. Miller<sup>2</sup>, G. C. Tiao<sup>3</sup>, D. Choi<sup>3</sup>, L. Zhang<sup>3</sup>, L. Bishop<sup>4</sup>,  
R. Stolarski<sup>5</sup>, G. J. Labow<sup>6</sup>, S. M. Hollandsworth<sup>7</sup>, G. E. Bodeker<sup>8</sup>, H. Claude<sup>9</sup>, D. DeMuer<sup>10</sup>,  
J. B. Kerr<sup>11</sup>, D. W. Tarasick<sup>11</sup>, S. J. Oltmans<sup>12</sup>, B. Johnson<sup>12</sup>, F. Schmidlin<sup>13</sup>, J. Staehelin<sup>14</sup>, P.  
Viatte<sup>15</sup>, and O. Uchino<sup>16</sup>.

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. Allied Signal Inc, Buffalo, New York
5. NASA Goddard Space Flight Center, Greenbelt, Maryland
6. Raytheon-STX, Lanham, Maryland
7. Space Applications Corporation, Vienna, Virginia
8. National Institute of Water and Atmospheric Research, Lauder, New Zealand
9. Deutscher Wetterdienst, Observatorium Hohenpeissenberg, Germany
10. Meteorological Institute of Belgium, Brussels
11. Atmospheric Environment Service, Downsview, Ontario, Canada
12. Climate Monitoring and Diagnostic Laboratory, NOAA, Boulder, Colorado
13. NASA Wallops, Wallops Island, Virginia
14. Institute for Atmospheric Sciences, Swiss Federal Institute of Technology, Zurich
15. Aerological Station, Swiss Meteorological Institute, Payerne
16. Atmospheric Environment Division, Japan Meteorological Agency, Tokyo

September, 1998.

Submitted to the Journal of Geophysical Research.



**Abstract.**

We present the results of two independent analyses of ozonesonde measurements of the vertical profile of ozone. For most of the ozonesonde stations we use data that were recently reprocessed and reevaluated 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 and in utilization of data correction factors, 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 17.5 km for 1970-96. 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 12 to 17.5 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 for 0-27 km. Cunnold et al. [1998] describe trends in the lower

stratosphere derived from SAGE I and II (Version 5.96) data, and Newchurch et al. [1998] 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. [1998].

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 Megretskaja, 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 for the analyses here. Data for four Canadian stations and for Wallops Island 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. 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 so few sondes were flown in 1993-6 [Kirchhoff, pers. comm., 1997]; trends for the earlier data are given in Logan [1994]. The sonde data were provided in a variety of formats. To facilitate trend analysis, 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 mbar (30 layers up to 10 mbar). The vertical resolution, ~1 km, was chosen to be similar to that of SAGE data.

A detailed discussion of the characteristics of the three types of ozonesondes currently in use is given in WMO [1998]; this report also provides details about the sonde programs, and discusses issues of data quality in depth [see also WMO, 1995]. The Canadian stations used Brewer Mast sondes until 1979-80, and electrochemical concentration cell (ECC) sondes thereafter. The three European stations use Brewer Mast sondes, the Japanese stations use their own type of electrochemical 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].

It is common practice that the integrated ozone column derived from the sonde profile measurement is scaled to an independent measurement of the ozone column made by a Dobson or Brewer spectrophotometer, and the scaling, or correction factor (CF), is used as a criterion for judging the quality of the data. The spectrophotometer results, in turn, depend on the absorption coefficients for ozone used to derive the column measurement. The set of coefficients recommended by the World Meteorological Organization for reducing the spectrophotometer data changed in 1992 to those measured by Bass and Paur [1985]. The reevaluated sonde data used here were all scaled to ozone column measurements on the Bass-Paur scale, with the exception of the data from Lauder which were provided unscaled, but with the CF. The data for Wallops Island from WOUDC were reprocessed so that each profile is scaled to reevaluated ozone column data on the Bass-Paur scale [Oltmans et al., 1998]. For the Canadian data archived at WOUDC, the ECC data are scaled to ozone column measurements on the Bass-Paur scale, but the older BM data are scaled to column data on the older Vigroux scale [Tarasick et al., 1995]. In order to make a more homogeneous data set, we multiplied the Canadian Brewer Mast data by



0.9743 to put them on the Bass-Paur scale. There are concerns about the quality of the Brewer Mast data for these stations, as the operating procedures did not meet standards employed by the three European stations [Claude et al., 1988], and they are considered less reliable for determination of trends than the European data [WMO, 1998].

Scaling the sonde profile to the column measurement requires an estimate to be made of the ozone amount above balloon burst, usually between 20 and 7 mbar. It has been standard practice to use the mixing ratio at or near the top of the profile to estimate the ozone column above, although a new method, using an SBUV climatology to estimate the top of the profile is now available [McPeters et al., 1997]; the latter method was used for the Wallops Island, Boulder, Hilo, and Lauder data [Oltmans et al., 1998]. The SBUV climatology was used for Payerne soundings that ended between 30 and 17 mbar, while for those that reached above 17 mbar 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 mbar, allowing a good estimate to be made of the ozone column above burst altitude (Table 2). About 80% of the Canadian sondes reach 20 mbar, except for Resolute, where about 70% reach 20 mbar. The Canadian ECC profiles that fail to reach 17 mbar 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 no ozone column is measured; for the high latitude station Resolute, almost all the winter data are scaled to the default CF because of the lack of column measurements.

There is a significant difference in tropospheric ozone values measured by BM and ECC sondes that must be accounted for in deriving trends for the Canadian stations. Intercomparisons 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%) [Logan, 1994], as discussed in WMO [1995; 1998]. Tiao et al. [1986] used an intervention term in their statistical trend model at the time when BM sondes were replaced with ECC sondes, and this approach was adopted here by both groups. The magnitude of the intervention term is similar to the differences

between ECC and BM sondes found in the intercomparisons, and varies among stations [Tiao et al., 1986]. For trends starting in 1980 (September 1980 for Goose Bay), the Canadian data are obtained exclusively with ECC sondes, so no intervention term is necessary.

## 2.2 Data Selection Criteria.

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 in this analysis 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 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. [1998] 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.

The selection criteria for the CFs used by LM were the same as those of Logan [1994], 0.9-1.35 for BM sondes except for Hohenpeissenberg, where 0.9-1.2 was used, and 0.8-1.2 for ECC and KC sondes. 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. also required that there was an ozone

column reading for the day of the sounding, and that the balloon burst was above 16 mbar: these requirements are to ensure that a good estimate can be made of the ozone profile above burst altitude, and that the profile measurement is consistent with the column measurement. Tiao et al. remove 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 profile [e.g., Logan, 1985, 1994; Miller et al., 1995].

LM analyzed the trends for 33 layers and for 11 layers obtained by summing 3 consecutive layers; only the latter results are shown here, as results for 33 layers did not appear to provide more useful information on trends than the 11 layers, and the error estimates are similar (Figure 2). Tiao et al. aggregated the 33 layers into 15 Umkehr layers that they have used in their previous analyses [Tiao et al., 1986; Miller et al., 1995].

The stricter requirements used by Tiao et al. mean that they omit a significantly larger fraction of the soundings than LM, particularly for the BM stations, as shown in Table 2. The third column gives the fraction of soundings retained in the LM analysis, and the last column the fraction retained in the Tiao et al. analysis. Hohenpeissenberg is the exception, since both groups use the same CF criteria, 0.9-1.2, and retain ~90% of the soundings. In the worst cases, Payerne and the Canadian BM data, only 25-44% of the soundings are used by Tiao et al., compared to 70-90% by LM; for the BM stations, a larger fraction of soundings are omitted from the earlier part of the record than from the later part, because of the downward trend in the correction factors. For the Canadian ECC soundings, Wallops Island, and the Japanese stations, Tiao et al. retain about 45-65% (except for Churchill, 31%), while LM retain about 85-95%. 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 mbar also causes significant data loss.

Trends in the correction factor using the Tiao et al. data selection criteria are given in Table 3. 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 Megretskaja 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, 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 mbar wind speed for Singapore and the F10.7 cm solar radio flux at Ottawa is used for the solar cycle dependence. 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 residual noise is modelled as a first order autoregressive model, with different variances in different seasons. The model is first run with no seasonal weighting, and the final seasonal weighting is determined in an iterative procedure. The QBO time series is the average of 50 mbar 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 predetermined 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  $\text{DU km}^{-1}$  and column trends in the troposphere (1000-250 mbar) and stratosphere (250-16 mbar) are shown, so that the results can be compared in the same units (DU) for the same columns.

Trends were computed for 1970 (or the beginning of the record 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 four seasonal trends. The covariance matrix of seasonal trends was used to calculate the standard error of the annual trend.

## **4. Results.**

### **4.1 Lower Stratosphere.**

#### *Time series.*

Monthly mean values of ozone near 90 mbar are shown in Figure 3 (upper panel) for selected stations, using the LM selection criteria; the right hand panels show the time series of monthly anomalies. Figure 3 (lower panel) shows the time series using the Tiao et al. selection criteria. Measurements are made 2-3 times a week at the European stations and weekly in Canada; at the Japanese stations measurements were made weekly at the beginning and end of the time series, with infrequent data and few summer measurements in between. The frequency of measurements is reflected in the variability in the monthly values. The effect of the stricter Tiao et al. criteria is to introduce more gaps in the time series, and to increase the variance in the monthly anomalies. Time series up to ~1993 for all sonde stations (WOUDC data) are shown in

Logan [1994].

Uccle, Hohenpeissenberg, and Payerne are the closest together of the sonde locations. There do not appear to be any major biases in ozone values near 90 mbar, except for the early 1970s where values at Hohenpeissenberg tend to exceed those at Uccle, and to a lesser extent, those at Payerne (Figure 4).

*Trends for 1970-96.*

Seasonal trends in the vertical distribution of ozone are shown in Figure 5 for 1970-96, in percent per decade; the solid lines are those from LM and the dotted lines those from Tiao et al. Annual trends are shown in Figure 6. The sonde data are expected to be reliable for trend determination up to 27 km, according to the recommendations in WMO [1998]. Results are shown for pressure levels centered below 13 mbar, ~29 km, so results for the top level should be considered less reliable than for the other levels.

The seasonal trends determined by the two groups usually agree within  $\pm 3\%$ /decade for most stations, and almost all trends agree within their standard errors in the stratosphere. Agreement is worst for Churchill, with differences of  $\pm 10\%$ /decade in summer and winter; this station suffers from serious data loss with the Tiao et al. data selection criteria. The annual mean trends (Figure 6a) agree within  $2\%$ /decade in the stratosphere, with the exception of results from Uccle and Payerne; these are the two stations with the largest trends in the correction factors for 1970-96, as discussed in Section 2.2.

The annual trends are shown in absolute units (DU/km/decade) in Figure 6b. This comparison avoids the problem of the percentage trends being referenced to the mean at different times; pressure was converted to altitude using the U.S. Standard Atmosphere. This figure, like Figure 6a, shows that the two groups obtain similar results for the ozone trends except for Payerne and Uccle above 50 mbar. The dashed line in Figure 6b shows the absolute trends computed with the LM model, but with the data treatment of Tiao et al.; their CF criteria, the requirement that the balloon reaches 16 mbar and that there is a measurement of the ozone column, and with the data divided by the correction factor. The results from the two groups are

similar at most altitudes at most stations when they use the same data treatment (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 dividing by the correction factor, trends were calculated with the LM model using the Tiao et al. data selection criteria, with and without removing the scaling to the ozone column. Results are shown in Figure 7 (in percent per decade) where 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 dividing by the correction factor.

For stations with no trend in the correction factor, Sapporo and Kagoshima, the different trends in Figure 7 are due primarily to the data selection criteria, while for Hohenpeissenberg, where the selection criteria were almost the same, the different trends are due primarily to the trend in the correction factors (see Table 3). 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 remaining stations, the effect of dividing by the correction factor is <2 %/decade. Dividing by the CF generally shifts the profile to larger or smaller trends depending on the magnitude of the trend in the CF. Changing the data selection criteria sometimes changes the shape of the trend vertical profile, with largest effects on trends generally 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 parts of the trend profile at Wallops Island, Churchill, and Edmonton (Figure 7). 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 results as the column trend (in DU per decade) for 250 to 16 mbar; this comparison also avoids the problems of the absolute trends being computed on different layer thicknesses and percentage trends being referenced to the mean at different times. The LM stan-

standard model generally gives more negative trends for stratospheric ozone than the Tiao et al. model; however, when the LM model is run with the data treatment of Tiao et al., there is much less difference compared to the Tiao et al. results. The results in Figure 6b and 8 imply 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). There are statistically significant decreases in ozone at all the stations analyzed here. The decrease in ozone is located between about 30 mbar and the tropopause. These results are similar to those shown in Logan [1994], Miller et al. [1995], WMO [1995], and Harris et al. [1997] for analyses of WOUDC data that ended a few years earlier. Bojkov and Fioletov [1997] analyzed the Hohenpeissenberg and Edmonton data at WOUDC relative to the tropopause height, and found significant decreases only 1-2 km above the tropopause.

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

The mean trend in ozone for the sonde stations located from 36° to 59° N is shown in Figure 10. Both groups find a mean trend of -7 %/decade for 100 to 200 mbar. From 80 to 15 mbar, the mean trend from Tiao et al. is about 1 %/decade less negative than the mean trend from LM. 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 3). 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 standard error of the nine annual



trends; this error was larger for all layers than the root mean square error of the annual trend errors for the individual stations.

The ensemble of seasonal trends for 36°-59° N from the LM model is shown in Figure 11. 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 discussed previously by Logan [1994] and Bojkov and Fioletov [1997]. Decreases in ozone are largest in spring and winter at the European stations, and in spring and summer at the Canadian stations, as shown in Figure 12. There is no significant decrease in ozone below 90 mbar at the European stations in summer and autumn, while the decrease persists in spring to 200 mbar. The decreases in ozone are larger for the Canadian stations than for the European stations, and persist to 200 mbar and below in all seasons. Given the concerns about the Brewer Mast data for the Canadian stations [WMO, 1998] these results are subject to some uncertainty. The Japanese stations Sapporo and Tateno also show largest decreases in winter and spring.

The composite seasonal behavior of the eight stations located between 36° and 53° N is shown in Figure 13. Both groups find strong seasonality in the trends from about 300 to 90 mbar, but the seasonal patterns are somewhat different. Both groups find the largest losses in ozone in spring and the smallest losses in autumn in this region. For the Tiao et al. results, the losses in winter are almost as large as in spring, while for the LM results, the winter losses are smaller than those 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). In each figure the results are compared to the trend for 1970-96 (dashed line). For about half the stations, the decrease in ozone in the lower stratosphere is larger for the period 1980-96 than for the period 1970-96. The mean trend is more negative by 1-2.5%/decade, with a maximum trend in the lower stratosphere of -10 %/decade. The errors in the seasonal trends are larger for the shorter period, because of the short length of the time series. For the Japanese stations the summer trends have extremely large errors because

of the paucity of data in much of the 1980s and these contribute to the large errors in the annual trends. The errors in the mean trend for 1980-96 are larger than the errors in the mean trend for 1970-96, because of these factors.

Figure 14 includes results for three stations that started operation after the 1970s. Boulder (1980-96) shows significant decreases in ozone from 80 to 20 mbar, while Hilo (1982-96) shows a significant decrease only at 80 mbar; it shows a significant increase at 15 mbar. The data from Lauder (1986-96) do not show any significant trends in the lower stratosphere, but show an increase of  $\sim 5\%$ /decade at 30 mbar, similar to that reported by Bodeker et al. [1998]. The re-evaluated sonde data were not scaled to the correction factor, and there is a small but significant trend in the correction factor (Table 3). For the Lauder data scaled to the correction factor, trends in the lower stratosphere are close to zero and are insignificant except at 30 mbar. Bodeker et al. [1998] present a detailed analysis of trends in the Lauder data.

#### *Differences in trends derived from SPARC and WOUDC data.*

Tiao et al. have applied the same trend model to the re-evaluated SPARC data and to the data archived at WOUDC prior to re-evaluation for Hohenpeissenberg, Payerne, and the three Japanese stations, for 1970-96. Results from the two data sets for each station are fairly similar with the exception of results for Payerne in the troposphere, discussed below. The WOUDC data give slightly more negative trends than the SPARC data for the lower stratosphere at Hohenpeissenberg in all seasons. For the Japanese stations and Payerne the results are similar for both data sets from about 80 to 20 mbar.

## 4.2 Troposphere

### *Time series.*

Monthly mean values of ozone near 500 mbar are shown in Figure 16 for selected stations; the right hand panels show the monthly anomalies. The data from Goose Bay are for BM sondes prior to August 1980, and ECC sondes thereafter. An intervention term was used in both statistical models to account for the jump, which intercomparisons from the 1970s and early 1980s sug-

gest to be about 15-20%. Difference time series for the three European stations are shown in Figure 17 for 500 mbar. There are systematic biases for ozone values in the mid-troposphere in the SPARC data sets, as were found in the WOUDC data for Hohenpeissenberg and Payerne [Logan, 1994]. For the reevaluated data, ozone values at Hohenpeissenberg are systematically higher than those at Payerne from about 1978 to 1990, while there is less bias before 1978. Values at Uccle are higher than those at Payerne and at Hohenpeissenberg up to about 1986. These biases result in different trends for the three stations. The data used here for Payerne are provisional, as discussed in WMO [1998], and are likely to be revised further. There are particular concerns about the consistency of the tropospheric data for Payerne in the 1980s and for 1990-93.

*Trends for 1970-96.*

The most obvious feature of the tropospheric trends shown in Figures 5, 6, and 11 is that there are significant spatial variations in the magnitude and sign of the trends, with decreases, or no significant trend at the Canadian stations and increases at the European and Japanese stations. The increases over Europe and at Kagoshima are significant from the surface up to 300 mbar, while the increases at Sapporo and Tateno are significant only at up to 500 mbar. There is not a significant seasonal variation in the tropospheric trends (e.g., Figures 5 and 12). The Tiao et al. trends are less negative for the Canadian stations, more positive for the European stations and Wallops Island, and about the same for the Japanese stations, compared to the LM results. LM find increases of 5-15 %/decade for the European stations, while Tiao et al. find increases of 6-25 %/decade. Part of this difference is caused by referencing the trend to the beginning rather than the middle of the record. For example, at the lowest layer at Hohenpeissenberg, the Tiao et al. trend is 16 %/decade, while the LM trend is 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 about 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 Payerne and Hohenpeissenberg for the absolute trend (in DU

$\text{km}^{-1}\text{decade}^{-1}$ , Figure 6b) and for the tropospheric column trend (in DU/decade, (Figure 18) are in closer agreement than the results for the percentage profile trends, another indication that the reference point makes an important difference to the results in  $\%/decade$ . The results of the two groups for the column trends for 1000-250 mbar are similar for most stations, and using the Tiao et al. data treatment with the LM model generally improves the agreement.

The ozone increases in Europe and Japan from LM for 1970-96 are somewhat less than those reported for 1970-91 for the WOUDC data [Akimoto et al. 1993; Logan, 1994]. This is caused by the relatively flat values of ozone in the last few years (see Figure 16). Bojkov and Fioletov [1997] find the increase at Hohenpeissenberg to be significant only 1 km below the tropopause. By comparing data for the first 5 years from the Canadian stations with data for 1987-91 Logan [1994] concluded that there was no evidence for a long term increase in ozone at the Canadian stations, given the different responses of BM and ECC sondes. This result contradicted the earlier analysis of Wang et al. [1993] who reported an increase of 10  $\%/decade$  for all the Canadian stations. Using an intervention in the statistical model to treat the change in sonde type, LM find long term decreases for all the Canadian stations (-2 to -9  $\%/decade$ ), while Tiao et al. find similar decreases or no significant trend at the same stations. Oltmans et al. [1998] report 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 analyzed only these three stations, selected for the consistency of their record, and used the data at WOUDC.

#### *Trends for 1980-96.*

There is a major change in many of the trends for the period 1980-96 compared to 1970-96 (Figure 14). For the European stations, only Payerne shows a positive trend, of  $\sim 10 \%/decade$ , while Uccle shows no change in ozone, and Hohenpeissenberg has a slight negative trend in the middle troposphere. There are concerns about the consistency of the tropospheric data for Payerne in the 1980s and early 1990s. Tateno also shows no change in ozone, while Sapporo and Kagoshima have increases of 5-15  $\%/decade$ , not all of which are significant. There are many

gaps in the data record for these two stations in the early 1980s, particularly for summer. These gaps give rise to large errors in the summer trends, which contribute to the large errors in the annual trends. The Canadian stations show decreases of -2 to -8 %/decade, and these are more reliable than the results for 1970-96 since ECC sondes were used for the whole record. Previous analyses of the Canadian ECC data also showed decreases [Logan, 1994, Tarasick et al., 1995; Oltmans et al., 1998]. Oltmans et al. [1998] find no significant trend in ozone for Hohenpeissenberg, Boulder, Wallops Island, Tateno, and Hilo for 1979-95 in the middle troposphere, in agreement with the results in Figure 14. The increases in ozone apparent in the late 1960s and 1970s appear to have levelled off at several of the sonde stations, and also at remote surface sites, as discussed also by Logan [1994] and by Oltmans et al. [1998]. The mean trend for the stations from 36°-59° N is zero, 3.5 %/decade less than the mean trend for 1970-96 (Figure 15). The change is caused by the less positive trend for the later period at the European stations and Tateno. The concept of a mean 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<sub>x</sub> and from changes in stratospheric input of ozone [e.g., Logan, 1994]. There are not enough stations to form a true statistical average of tropospheric trends, even for northern mid-latitudes.

*Differences in trends derived from SPARC and WOUDC data.*

Tiao et al. find similar tropospheric trends using the SPARC and WOUDC data for Hohenpeissenberg, Sapporo, Tateno and Kagoshima. The results for the two data sets are dramatically different for Payerne, with much larger tropospheric increases derived for the WOUDC data than for the data used here (Figure 19). The history of problems with the Payerne data is documented in WMO [1998], and 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 data archived at WOUDC at that time. 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 for the early 1990s were subsequently corrected [Stubi et al. 1998], and provided for the SPARC analysis. Further revisions are expected in the Payerne data after more work on homogenizing the record.

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

The sonde trends in DU were integrated from the lowest layer to the layer with its top boundary near 16 mbar, omitting trends derived from the less reliable data obtained near the top of the soundings. These are compared to trends derived from the Dobson (or Brewer) column data that were obtained on the same day as the sonde measurements. (The column data were sometimes unavailable for the Canadian ECC sondes and Wallops Island, and are mostly unavailable for Resolute and Churchill in winter). The trends in the column data were derived using the LM model used for sonde data, omitting measurements on days that did not meet the CF criteria for sondes used by LM. Figure 20 compares the column trends and the integrated sonde trends, the latter for both the LM and Tiao et al. results. The LM results are in somewhat better agreement 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 CFs 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 Dobson 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 Dobson trends. If there is ozone loss above 16 mbar, the integrated sonde trends should be less negative than the Dobson 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 mbar for 40°-50°N are -2 to -4 DU/decade [W. Randel, personal communication, 1998].

Figure 21 shows the column trends derived from TOMS by Hollandsworth for Nov, 1978 to Oct. 1994 [WMO, 1998], compared to the integrated sonde trends and Dobson column trends for 1980-1996. The three trends agree within their standard errors, but the 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 21 is not an ideal comparison of the ground based and TOMS ozone columns, since it uses the ground based column data only on the days when there was an ozone sounding; 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, treated the normalization to the column measurements differently, and used different statistical models. The differences in the trend models are the assumption of zero trend prior to 1970, the inclusion of autocorrelation, and the removal of outliers in the model of Tiao et al., and the method of weighting, including the iterative procedure. All these 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  $\text{NO}_x$  are thought to be the primary cause of trends in ozone. The comparisons of absolute trends and of column trends indicates that the primary reason for different results of the two analyses appears to be the treatment of data prior to trend analysis; results expressed in this form offer the advantage of not being influenced by the different reference point for computing percent trends.

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 decreasing 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? Rather than make a judgement, we offer some comments. 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. They do not require the soundings to reach a certain height because over 94% of the soundings at the reevaluated stations and at Wallops Island reach 20 mbar. For the soundings from reevaluated stations an ozone column measurement is given whenever a correction factor is given. 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). The main cause of data loss is the stricter correction factor criteria, although the other requirements cause significant data loss for the Canadian stations and Wallops Island. The effect is that 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 dividing the data by the correction factor for 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 dividing the data by the correction factor [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 correction factor, 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 correction factor before deriving trends. 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 autoregressive errors within the statistical trend estimation process. 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 mbar (less strict than 16 mbar), 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 mbar 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 climatology could be used to derive the top of the profile for soundings that reach 20 mbar, 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 mid-latitude lower stratosphere in the northern hemisphere from 1970 to 1996, and that the largest decreases are located between 200 and 50 mbar (12-21 km). All stations show significant decreases in ozone, with a range of -3 to -10 %/decade near 100 mbar (17 km). The decreases are largest at the Canadian stations and Sapporo (the most northerly Japanese station), and are smallest at the European stations and Wallops Island in both analyses. For the mid-latitude stations, the mean trend is significant from 200 to 30 mbar (12-24 km) and is largest, -7 %/decade, from 200-80 mbar (12-17.5 km). 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 seasonal variation in the trends is located primarily in the lowermost stratosphere, between about 12 and 17.5 km. There is little seasonal variation 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 mbar at the European station in summer and autumn, while the decrease persists to 200 mbar in spring. Decreases are largest in spring and summer at the Canadian stations, and persist to 200 mbar 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 no significant trend at the Canadian stations for 1970-96; for the more reliable ECC data after 1980, there are decreases of -2 to -8 %/decade in the mid-troposphere. The European stations show increases of 5-25 %/decade which are significant from the surface to 300 mbar (9 km) for 1970-96; there is no significant trend for Uccle and a marginally significant decrease for Hohenpeissenberg for trends starting in 1980. Only Payerne gives an increase, 10 %/decade for 1980-96, and this is a provisional data set 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 (30° 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 American 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 reliably define 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 also agree 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. 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 to 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 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.

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.*, in press, 1998.

Bojkov, R. D., V. E. Fioletev, 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., J. J. Wang, L. Thomason, J. Zawodny, and J. A. Logan, SAGE (v.5.96) ozone trends in the lower stratosphere, *J. Geophys. Res.*, to be 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 Scientifico and Tecnologico D'Abruzzo, Italy, in press, 1998.

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.

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.

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 balloon-sonde 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*, 9257-9263, 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 through 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 Scientifico 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.*, 10<sup>2</sup>, 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 Association, Global Ozone Research and Monitoring Project Report, No. 43, World Meteorological Organization, 1998.



Table 1: Sonde data used in the analysis.

ID	Station	Lat.	Long.	Type	Period
<b>SPARC DATA</b>					
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
<b>WOUDC DATA</b>					
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 2: Fraction of soundings that meet various criteria.

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
Station			20 mbar	20 mbar		16 mbar	16 mbar
				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
Station			20 mbar	20 mbar		16 mbar	16 mbar
				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

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 mbar 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 mbar, 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 is 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 used by LM; for the ECC soundings, the default CF is 1.0, which 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.

Table 3. Trend in correction factors (%/decade).

Period	70-96	80-96
Uccle	-2.6±0.6	-3.7±1.3
Payerne	-1.9±0.5	-3.0±0.8
Hohenpeissenberg	-1.3±0.5	NS
Sapporo	NS	NS
Tateno	-1.3±0.9	-3.3±1.7
Kagoshima	NS	NS
Wallops Is.	-2.0±1.1	-2.5±1.8
Boulder	-	-3.7±1.2
Hilo	-	-3.7±1.3 <sup>a</sup>
Lauder	-	-1.5±1.5 <sup>b</sup>

Period	70-79	80-96
Resolute	-2.3±2.5	NS
Churchill	NS	-2.3±2.4
Edmonton	5.1±6.0	-2.7±1.4
Goose Bay	NS	2.0±1.7

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 mbar, 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.

- a. Trend for 1982-96
- b. Trend for 1986-96

Figure Captions.

Figure 1. Correction factors for selected sonde stations.

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 pressure from 1000 to 6.3 mbar. The solid line shows results for 11 layers obtained by summing the ozone content in 3 consecutive layers. Results are shown below 10 mbar.

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 mbar, and the same relative scale is used for both sets of means.

Figure 4. Difference of monthly mean values for the three European stations near 90 mbar. The correction factors used by LM were applied (see text).

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.

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.

Figure 6b. Annual trends in the vertical distribution of ozone for 1970-96 in DU/km/decade. 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 were

derived with the LM model run with the Tiao et al. data treatment (their selection criteria, and with the data divided by the CF).

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.

Figure 8. Column trend in ozone in DU/decade from 250 to 16 mbar 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).

Figure 9. Annual trends for individual sonde stations located between 36° and 59° N, superimposed.

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 standard error of the nine trend values at each pressure level.

Figure 11. Seasonal mean trends for the sonde stations located between 36° and 59° N, superimposed. LM results.

Figure 12. Seasonal mean profiles for three European stations, 48°-51°N (left) and for three Canadian stations, 53°-59° N (right). LM results.

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

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.

Figure 15. Mean annual trend for the sonde stations located between  $36^{\circ}$  and  $59^{\circ}$ N, for 1980-96 (solid line) compared to the mean trend for 1970-96 (dotted line). LM results. Two standard errors are shown; these were calculated as the standard error of the nine trend values at each pressure level.

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 mbar, and the same relative scale is used for both sets of means.

Figure 17. Difference of monthly mean values for the three European stations near 500 mbar. The correction factors used by LM were applied (see text).

Figure 18. Column trend in ozone in DU/decade from the lowest layer to 250 mbar. 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).

Figure 19. Comparison of seasonal trends for (i) sonde data re-evaluated for this study (solid lines) and (ii) for sonde data archived at WOUDC (dotted lines), for Payerne. The same data selection criteria were applied to both sets of data. Tiao et al. results.

Figure 20. 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 mbar, computed with the LM model, and the triangles show the integrated sonde

trend up to 16 mbar 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.

Figure 21. 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.

Fig. 1

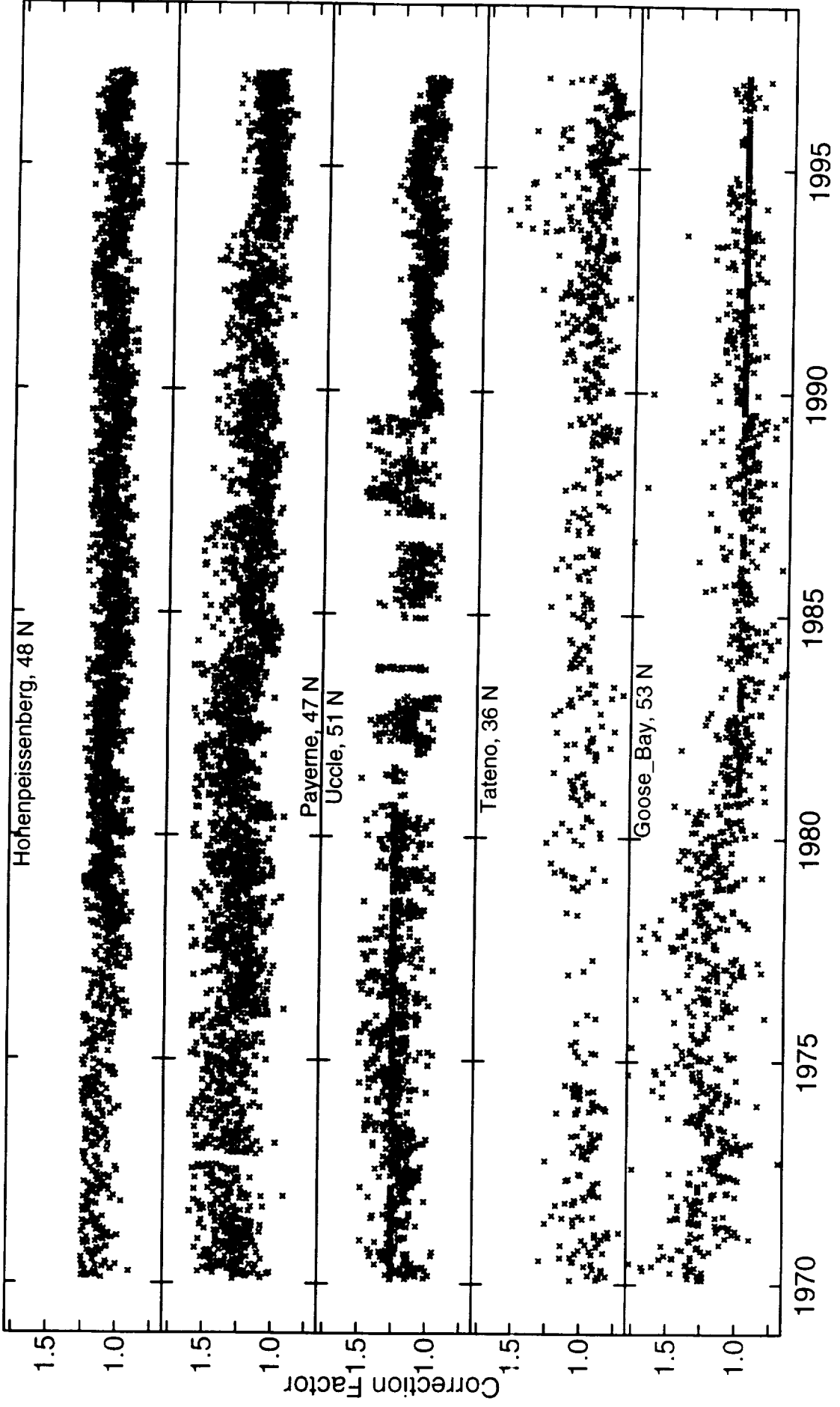
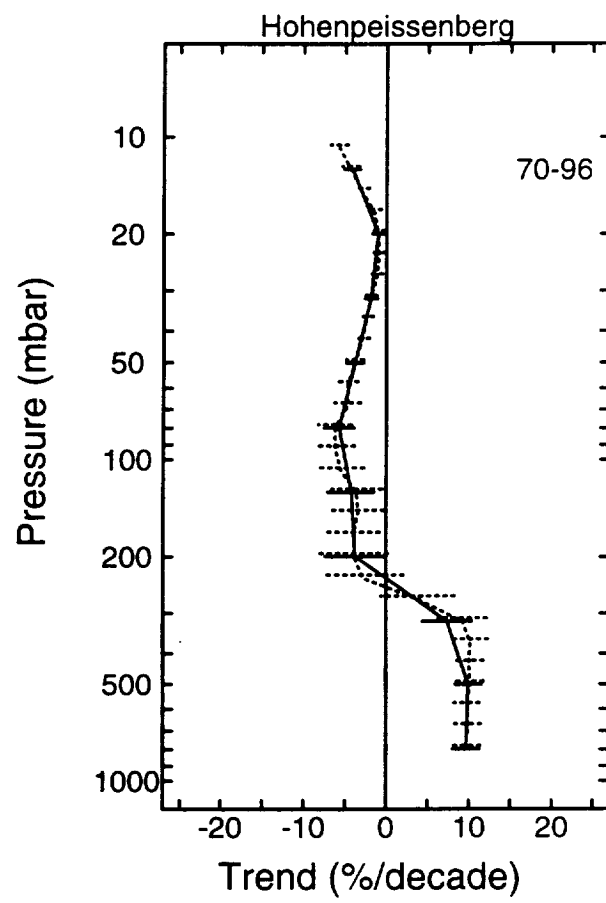
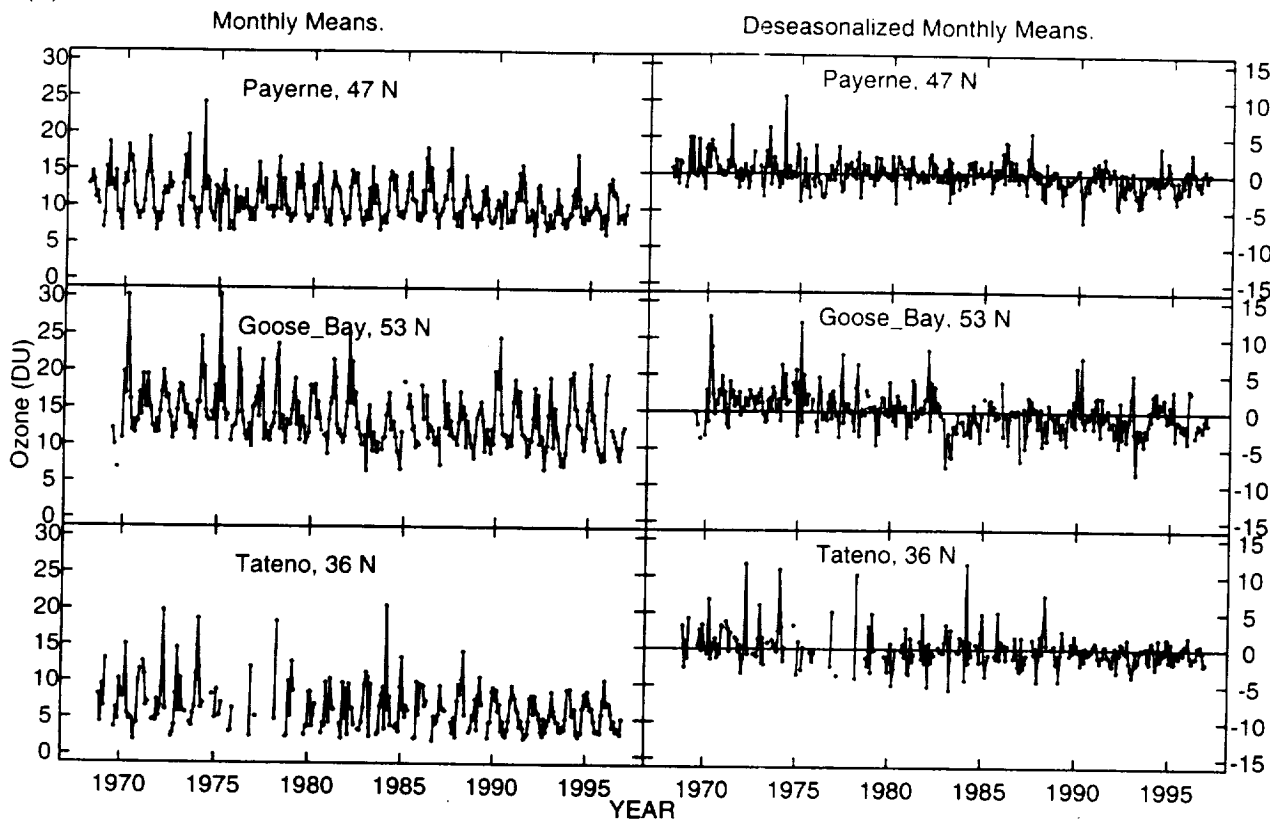




Fig. 2



(a)



(b)

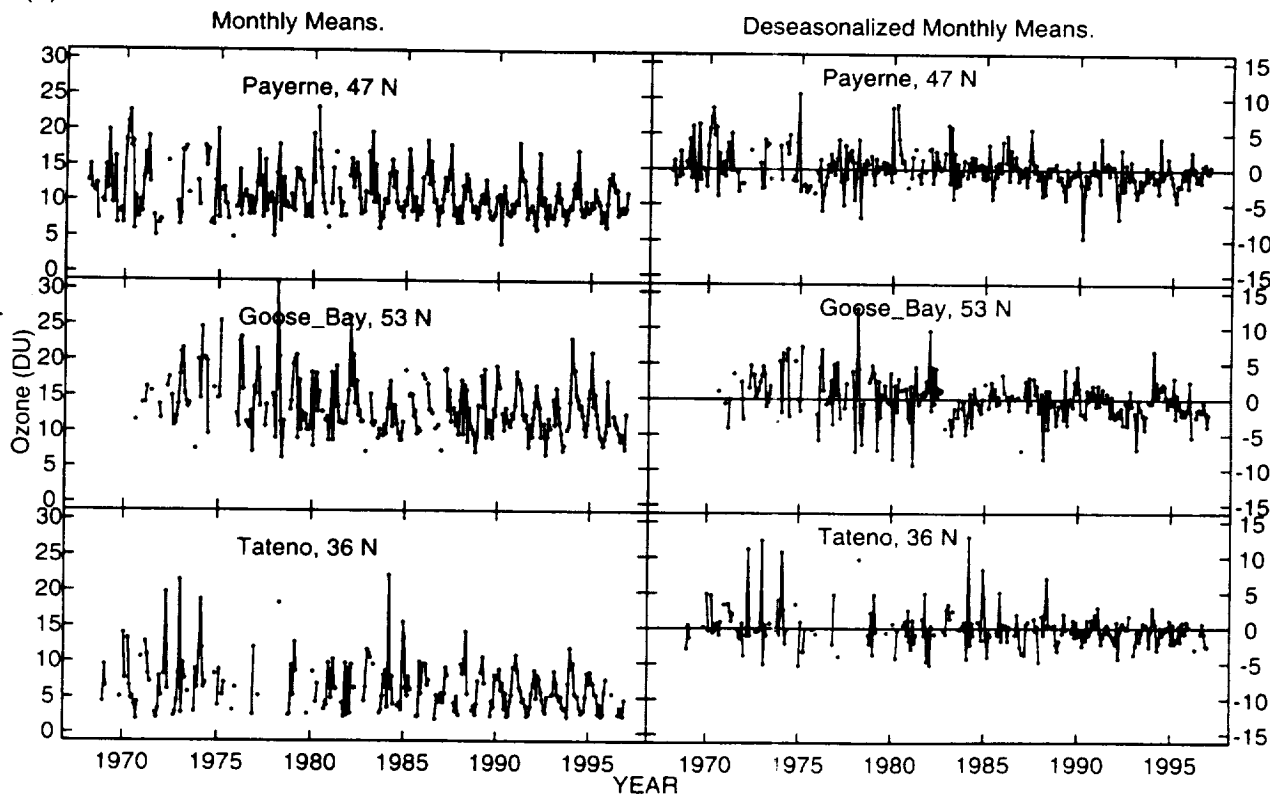
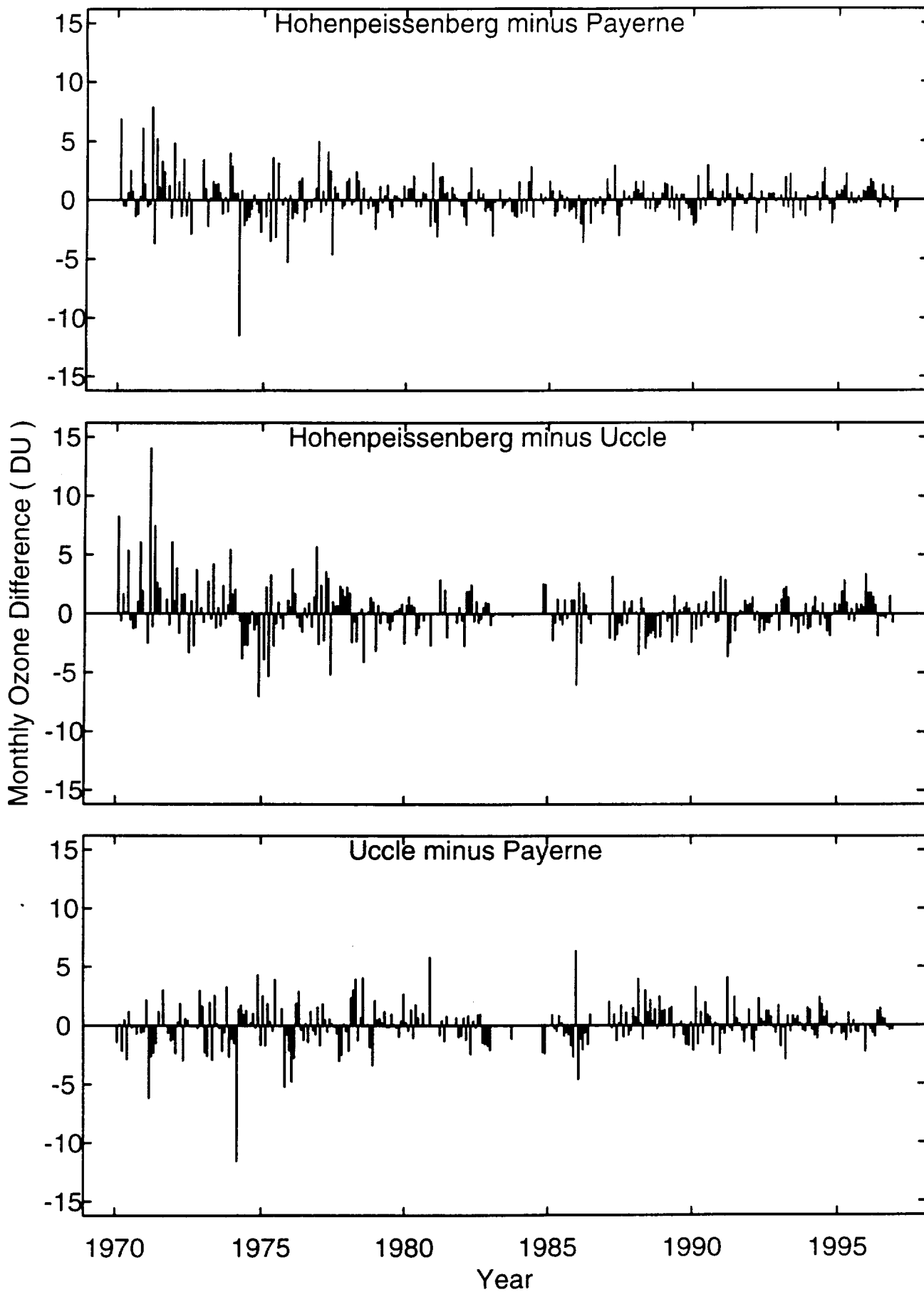
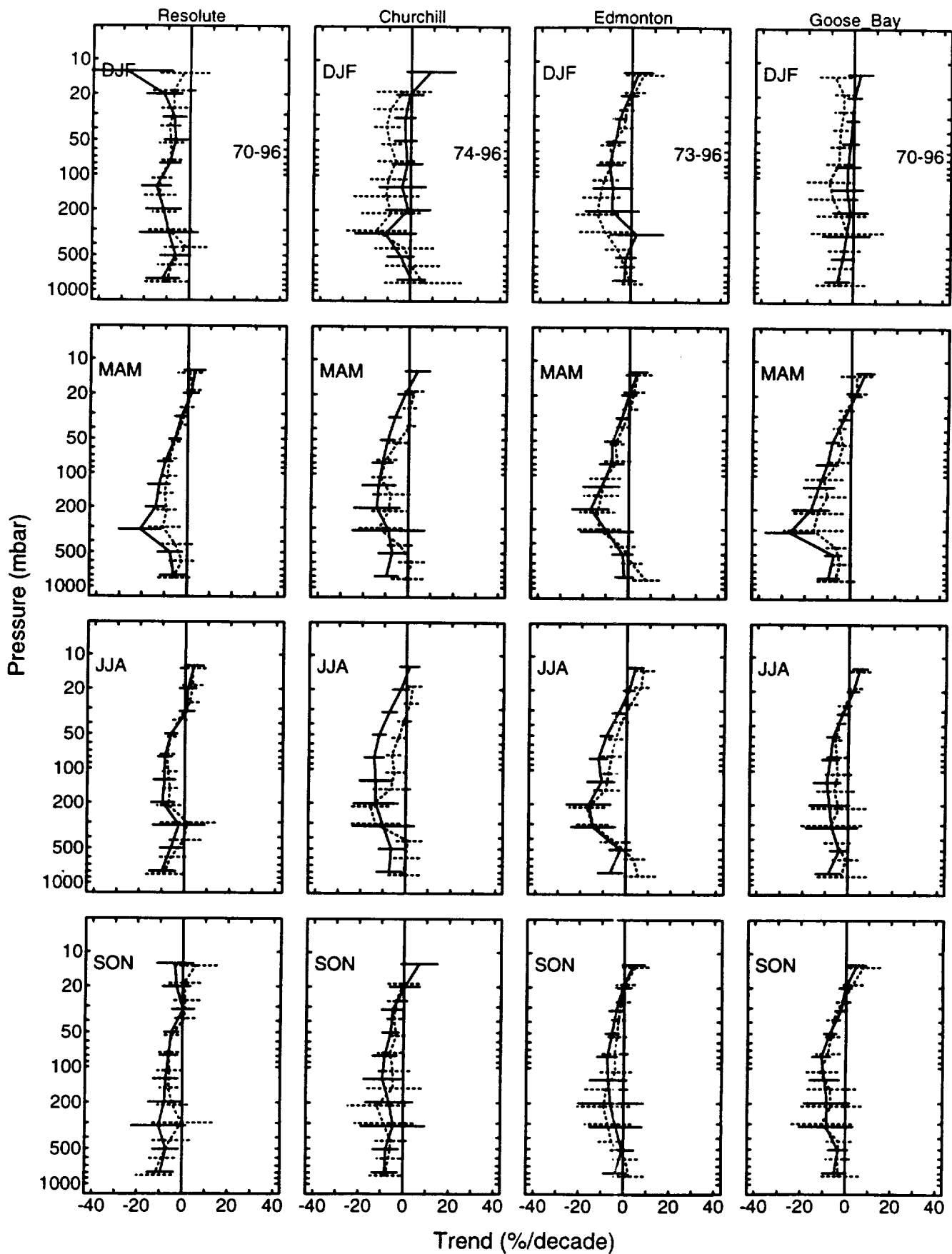
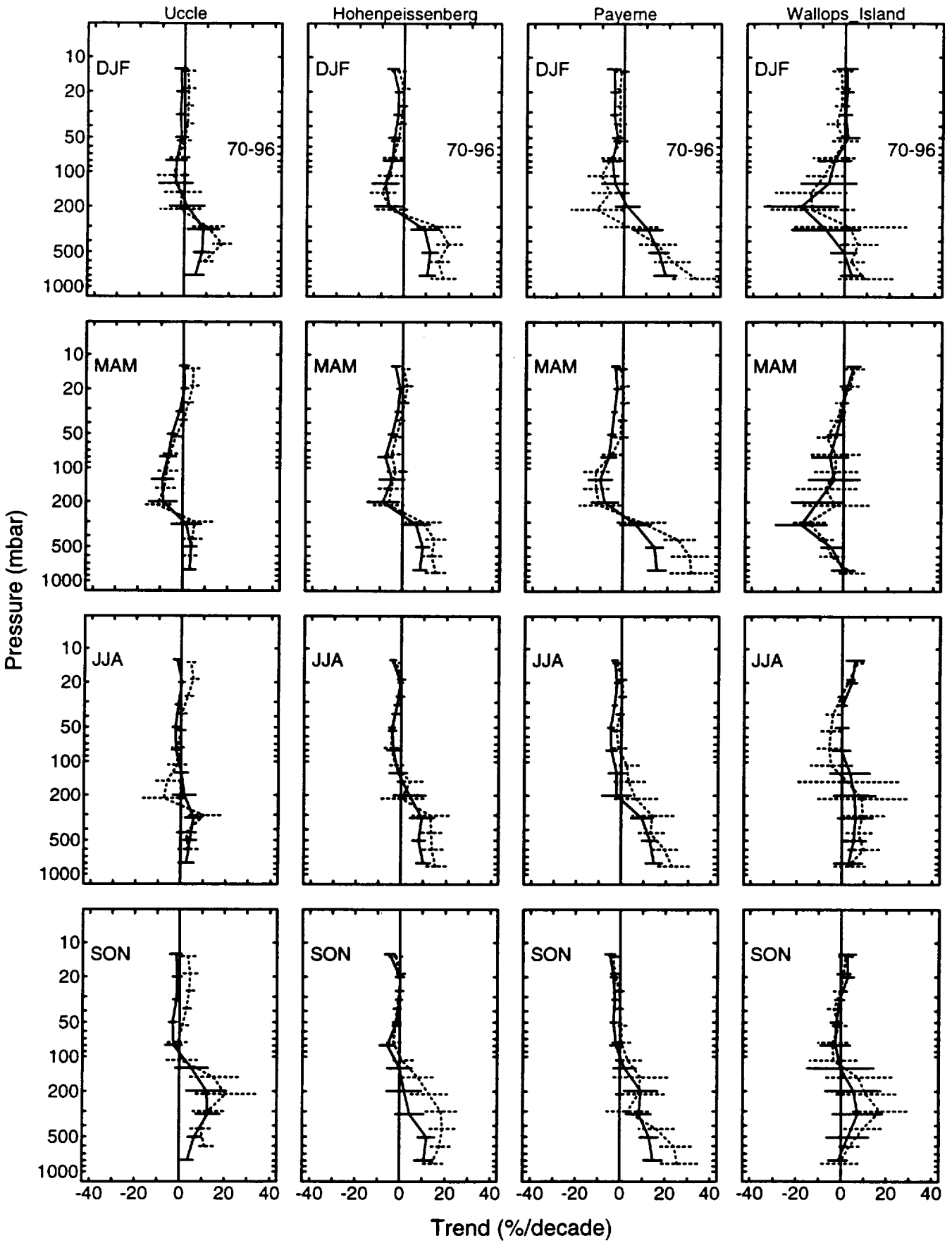


Fig. 4







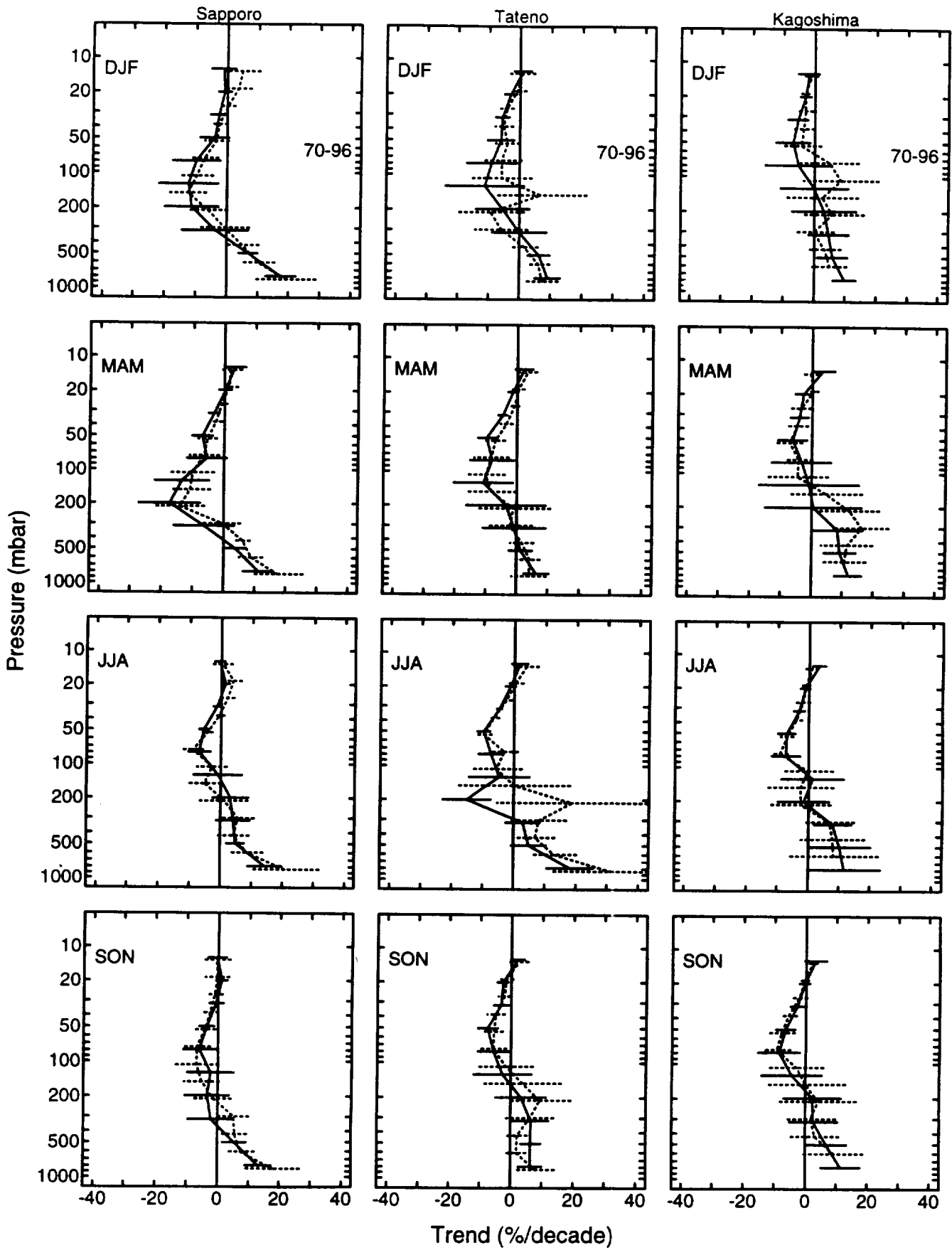


Fig. 6a.

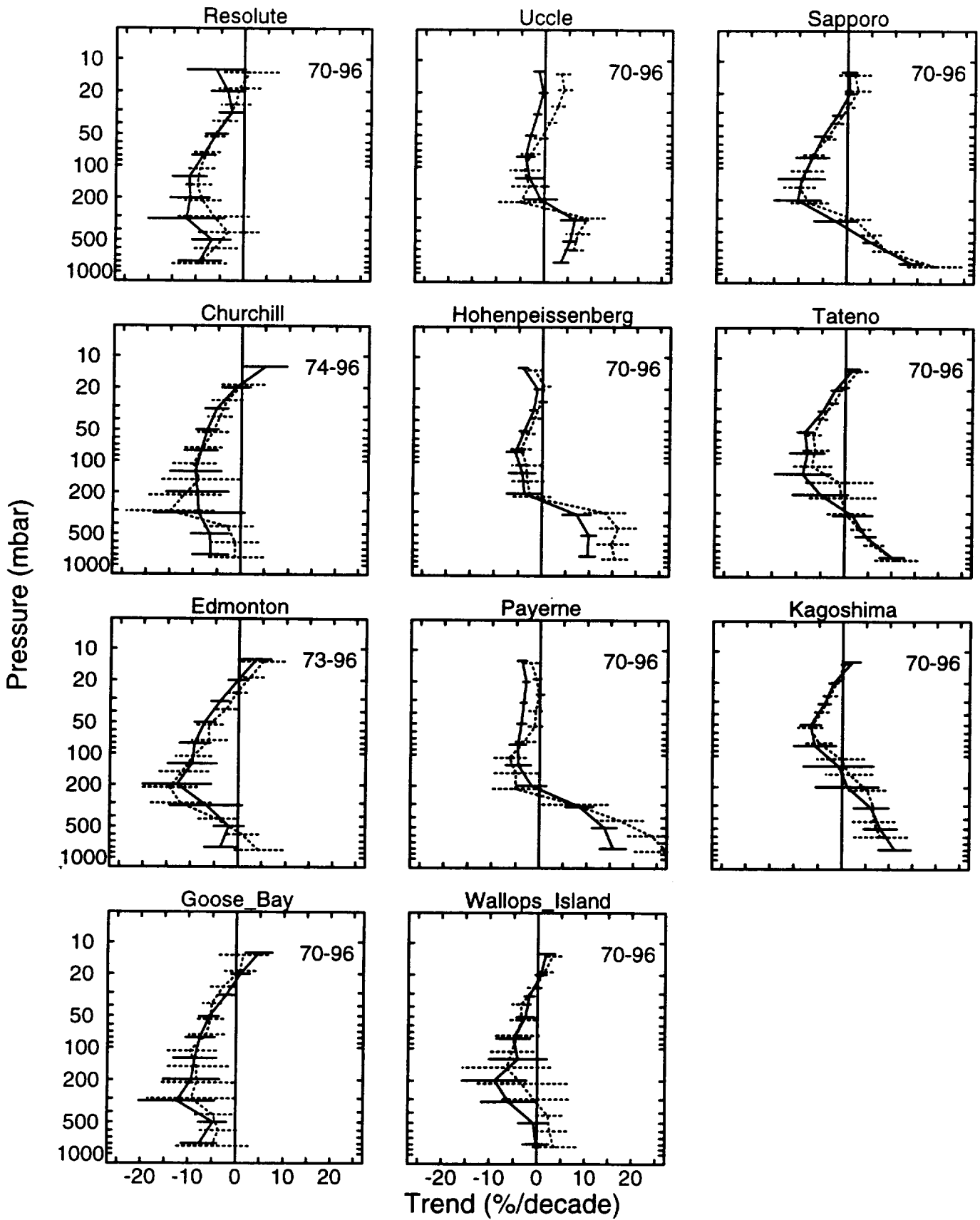


Fig 6b

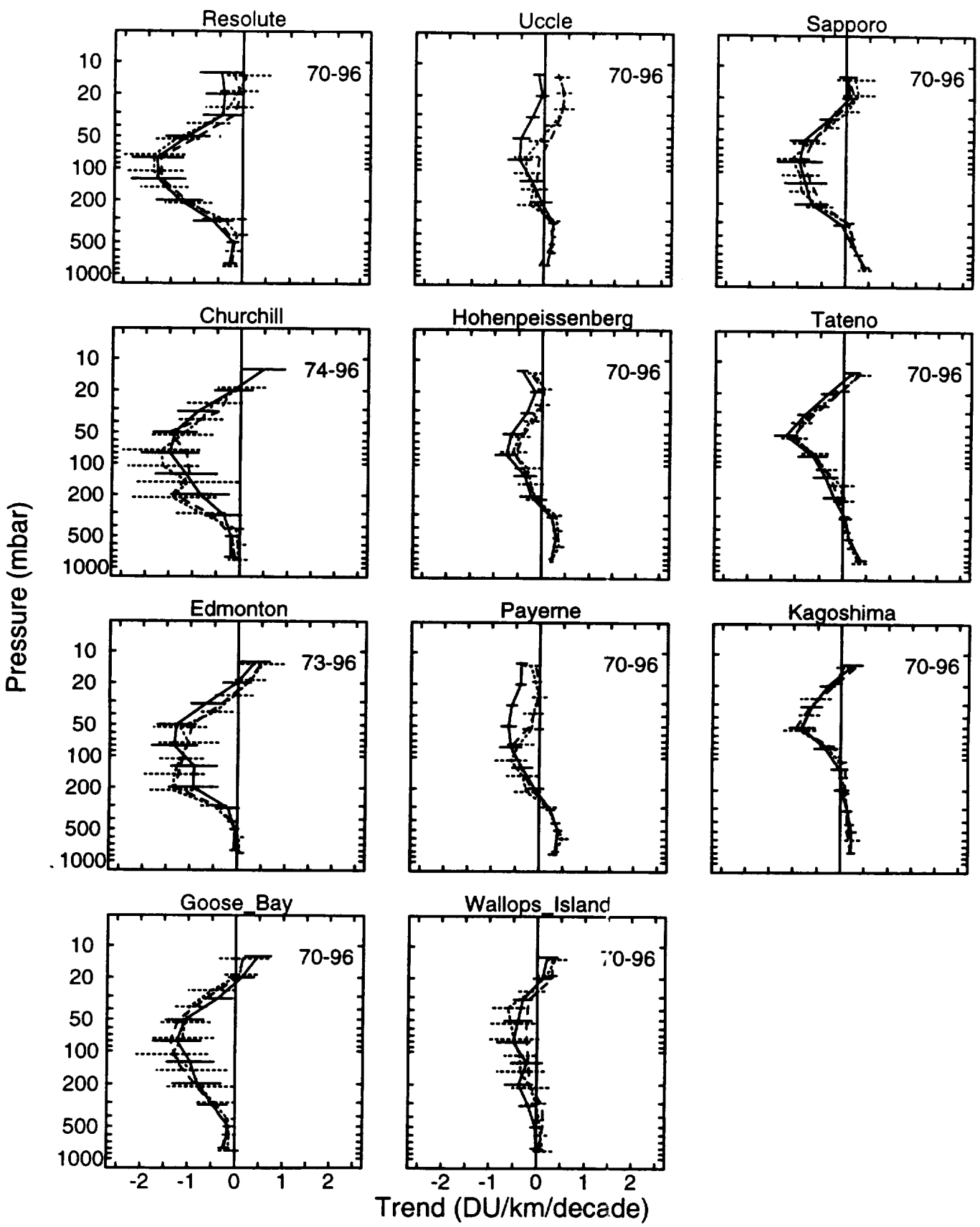
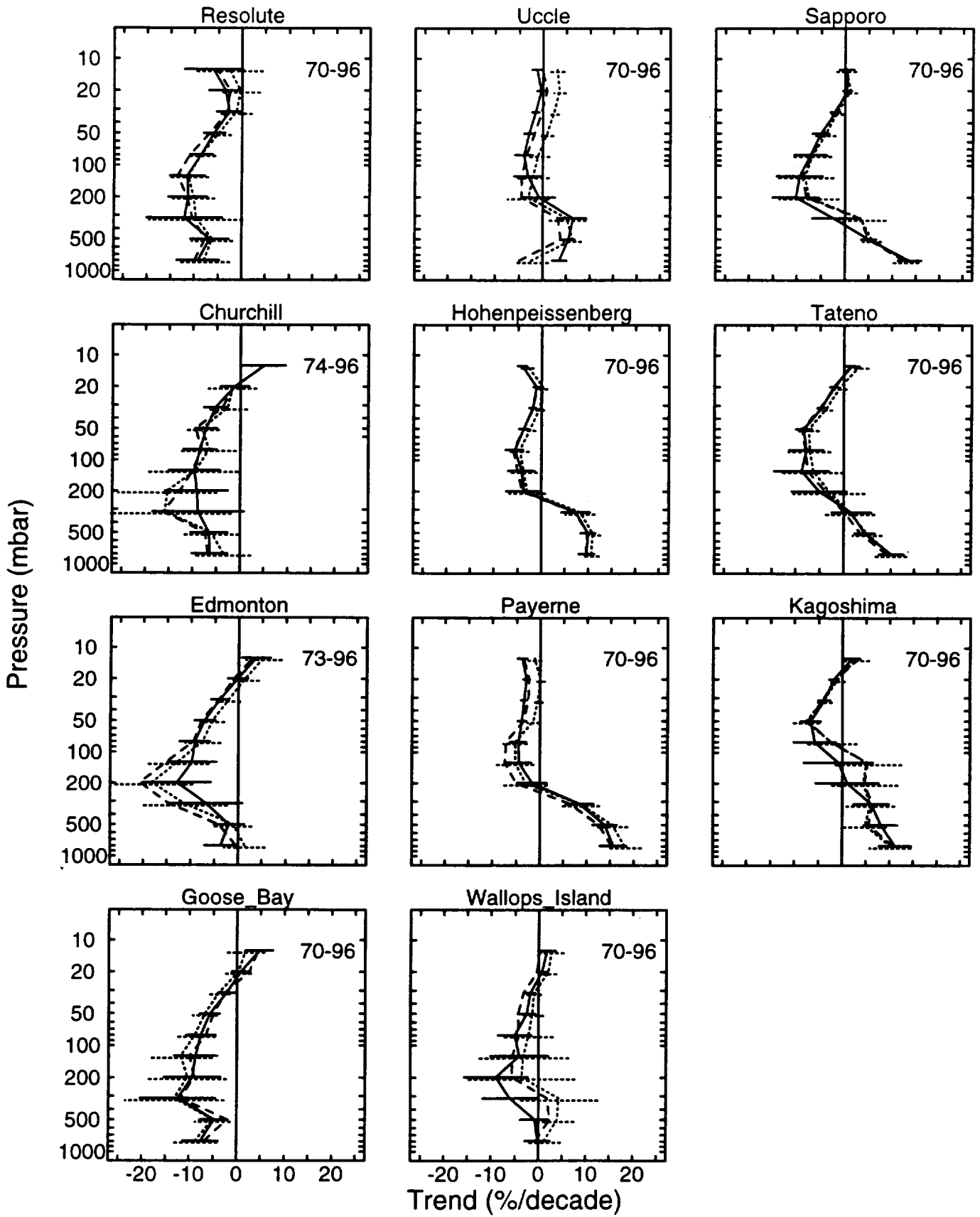


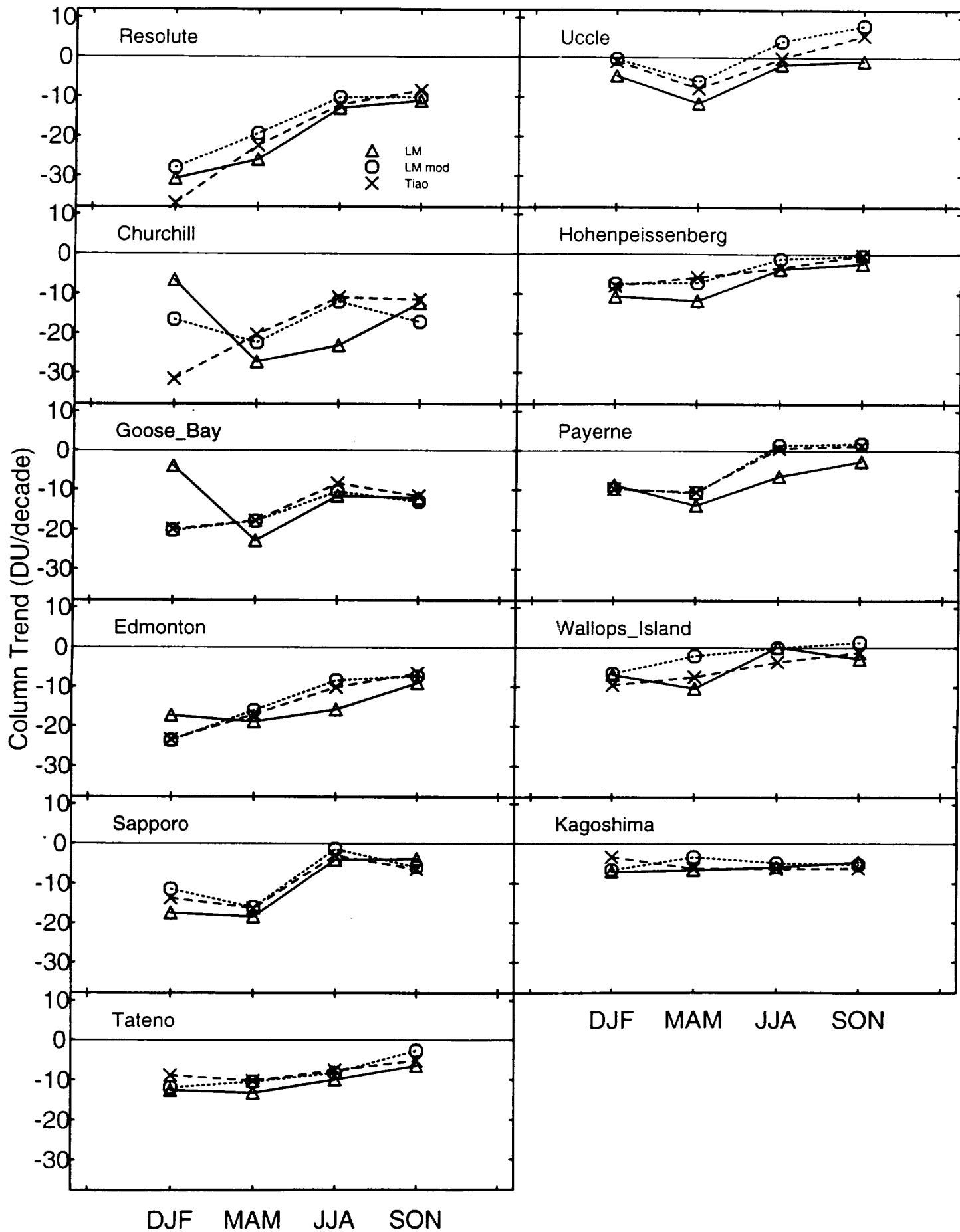


Fig. 7



250-16 mbar

Fig. 8



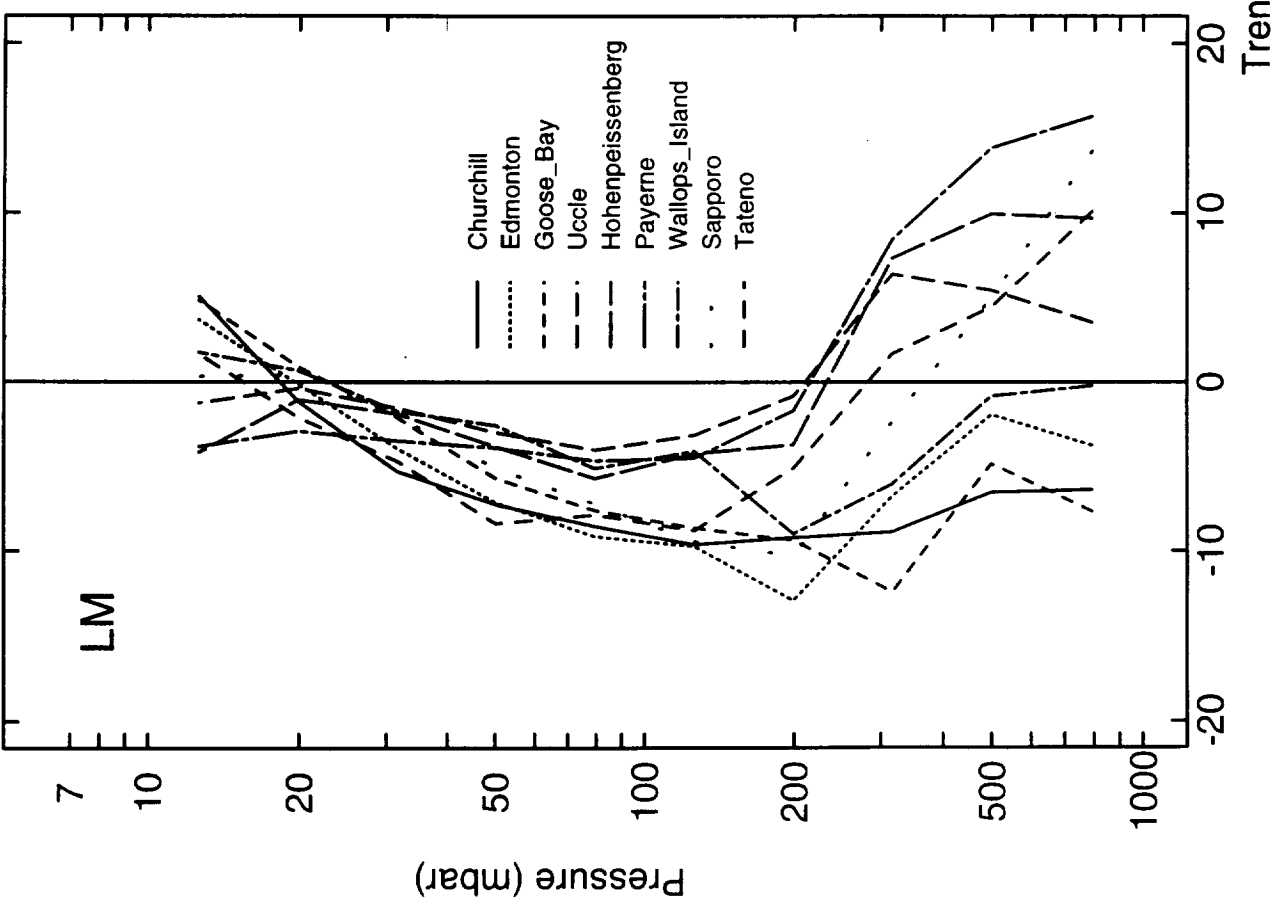
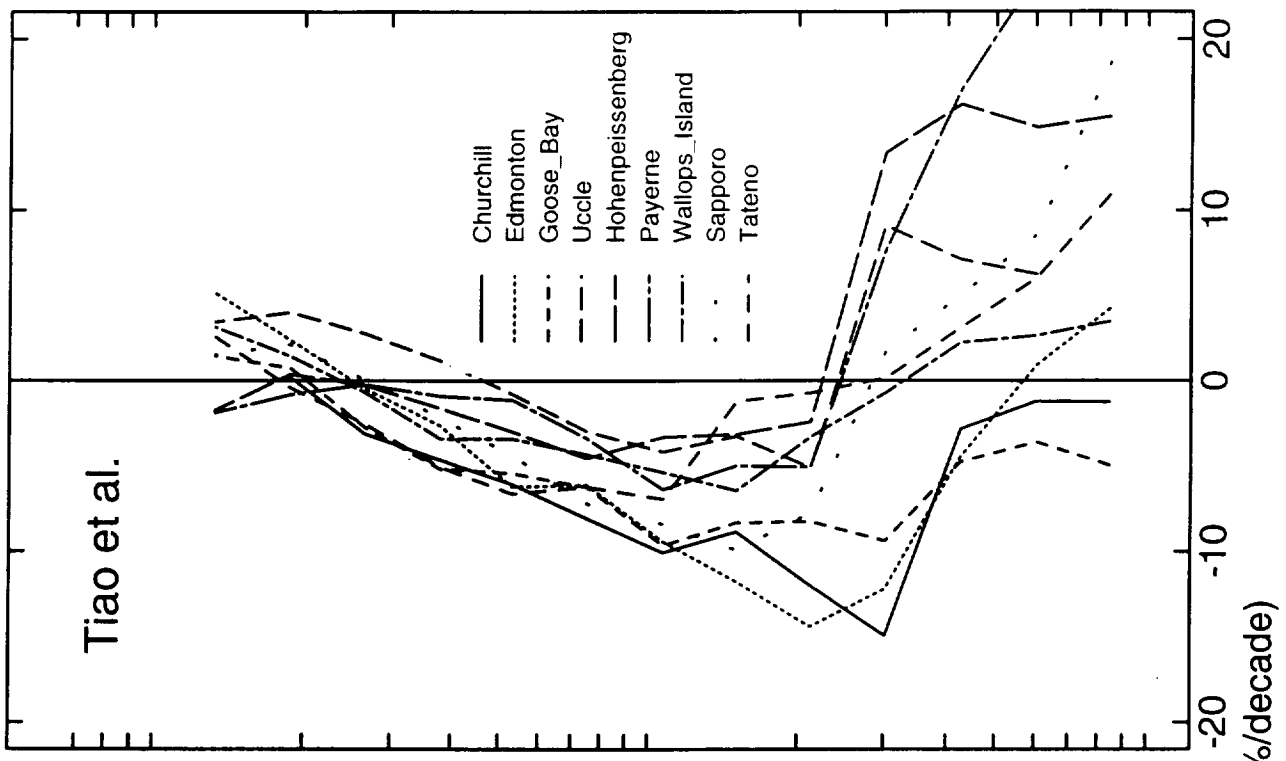


Fig. 10

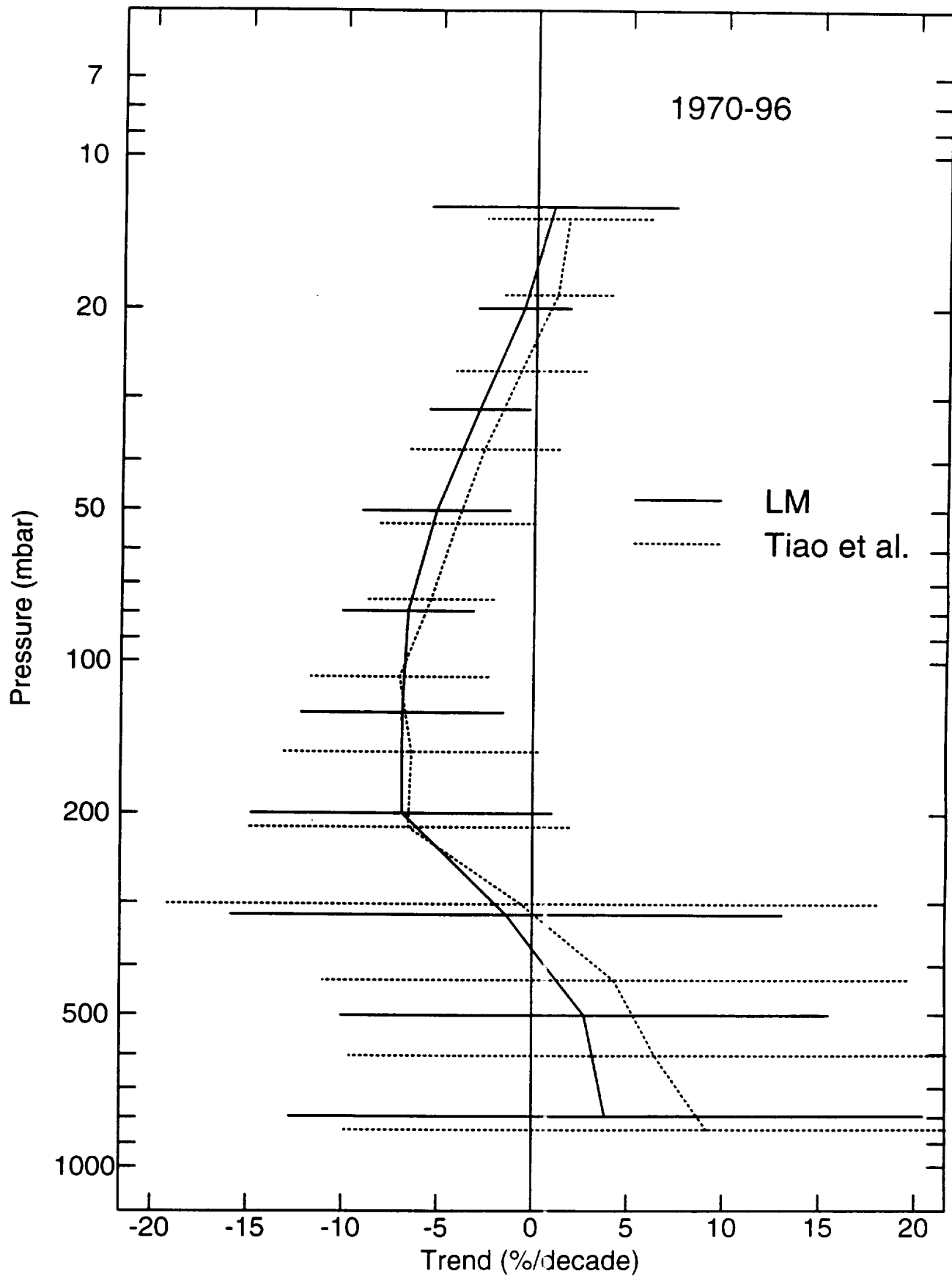


Fig. 11

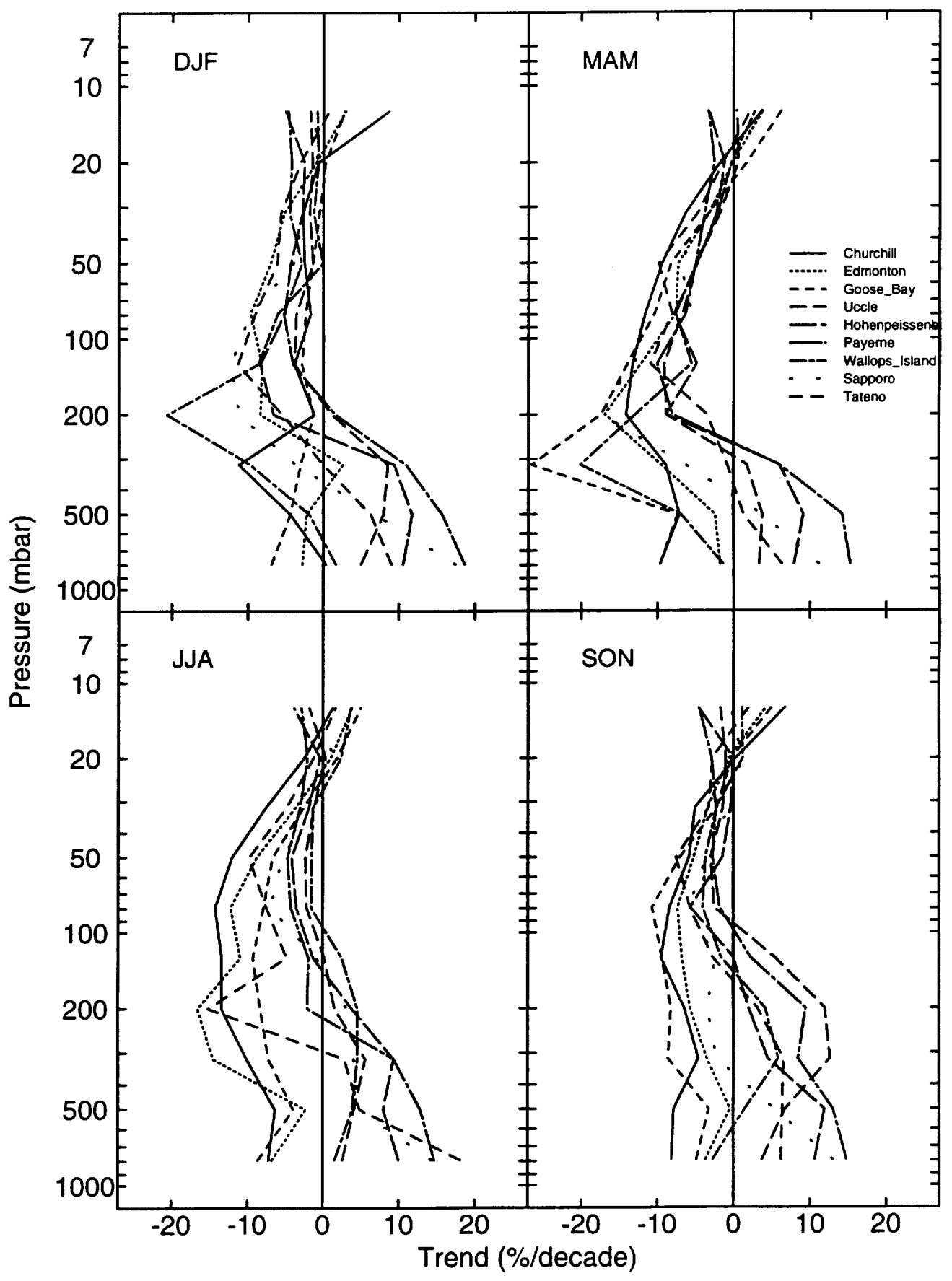


Fig. 12

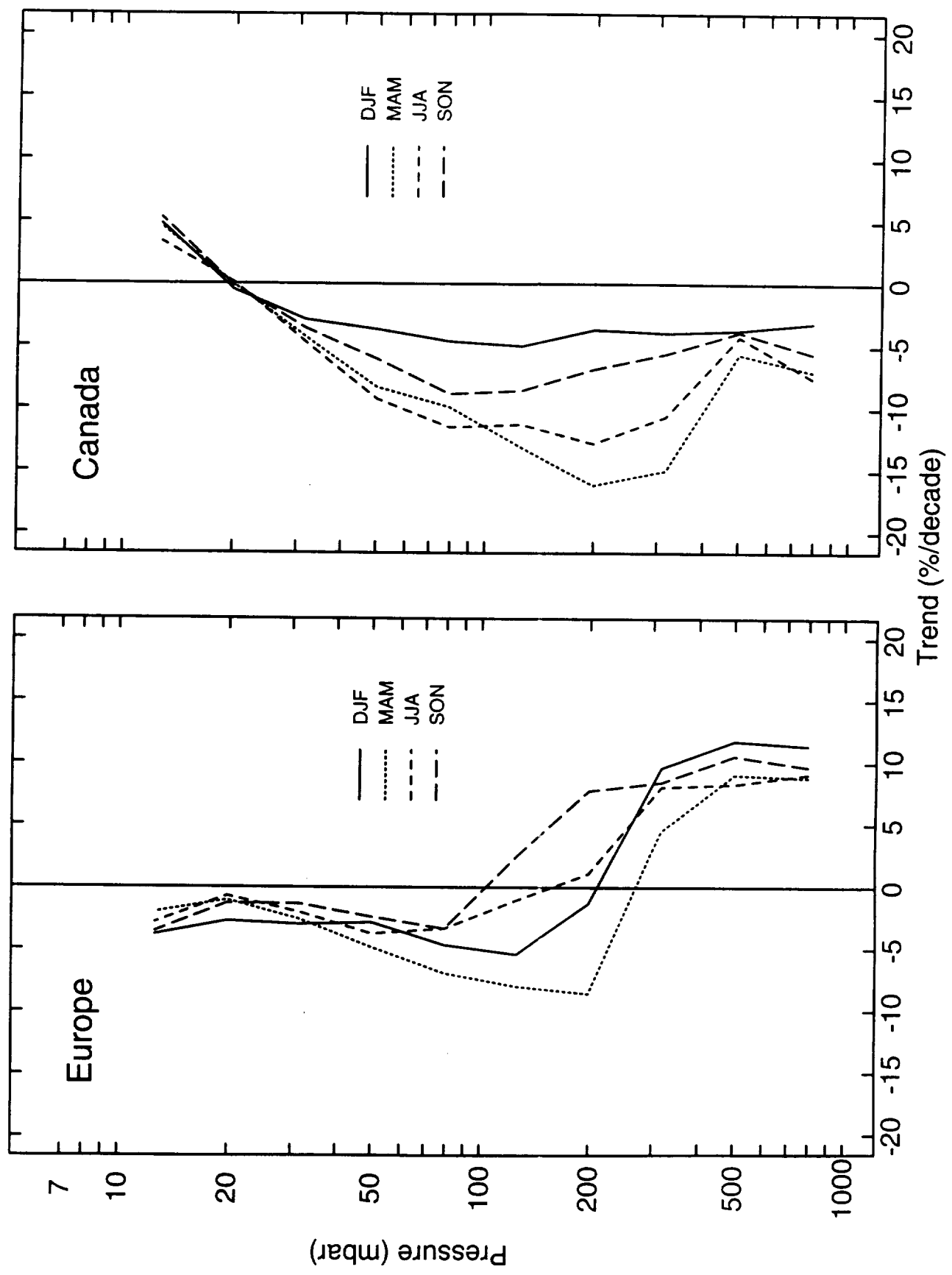


Fig. 13

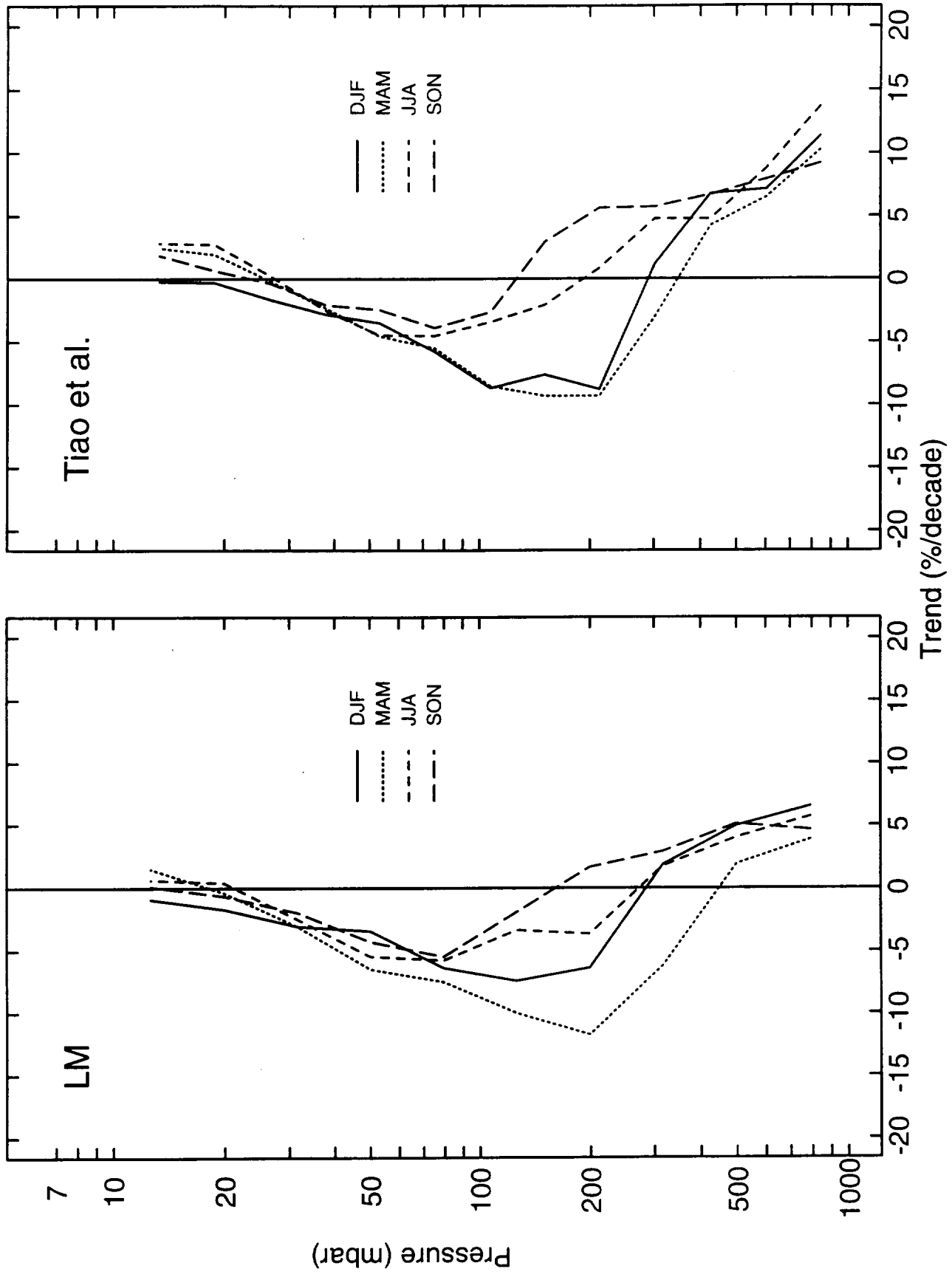


Fig. 11

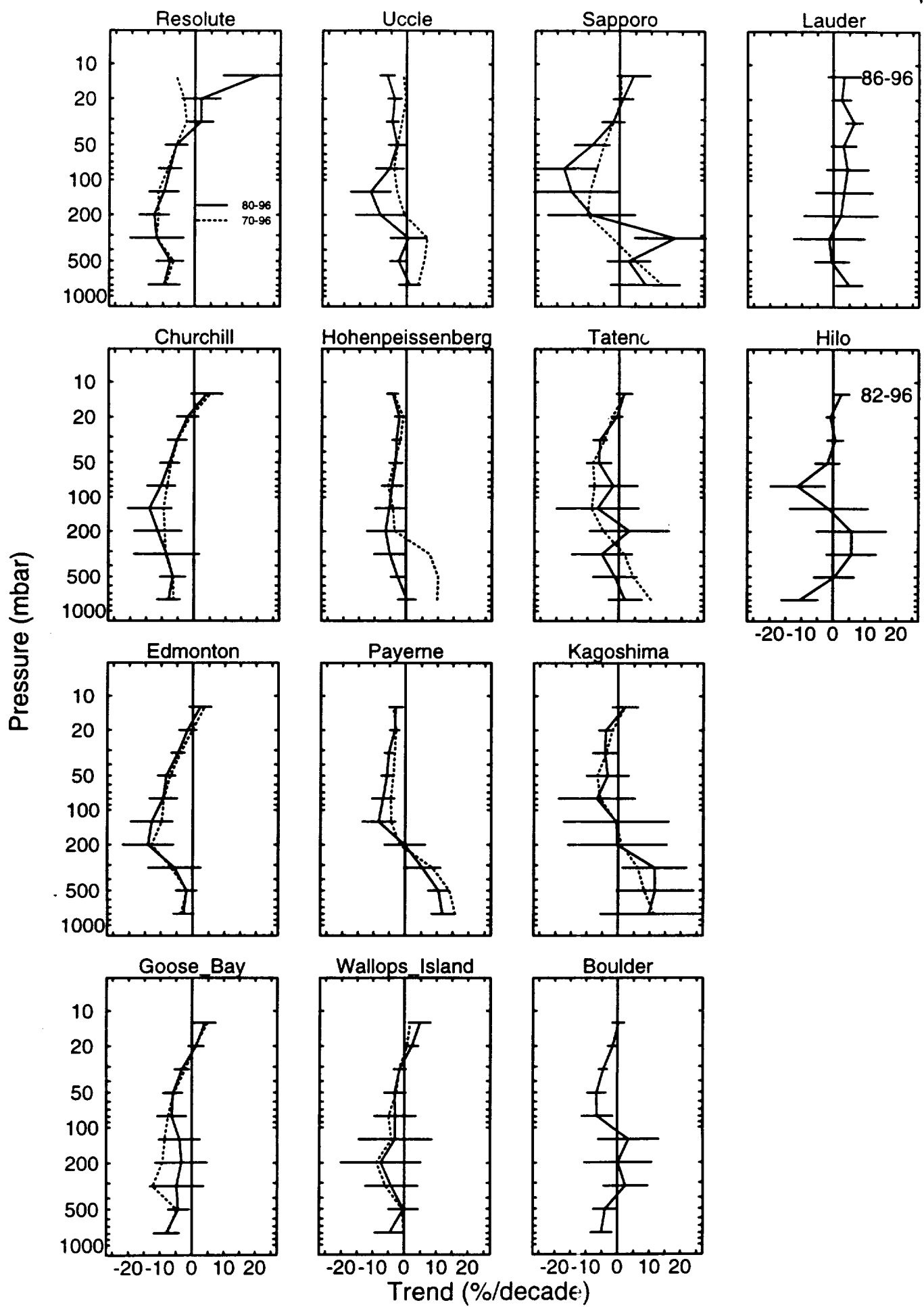




Fig. 15

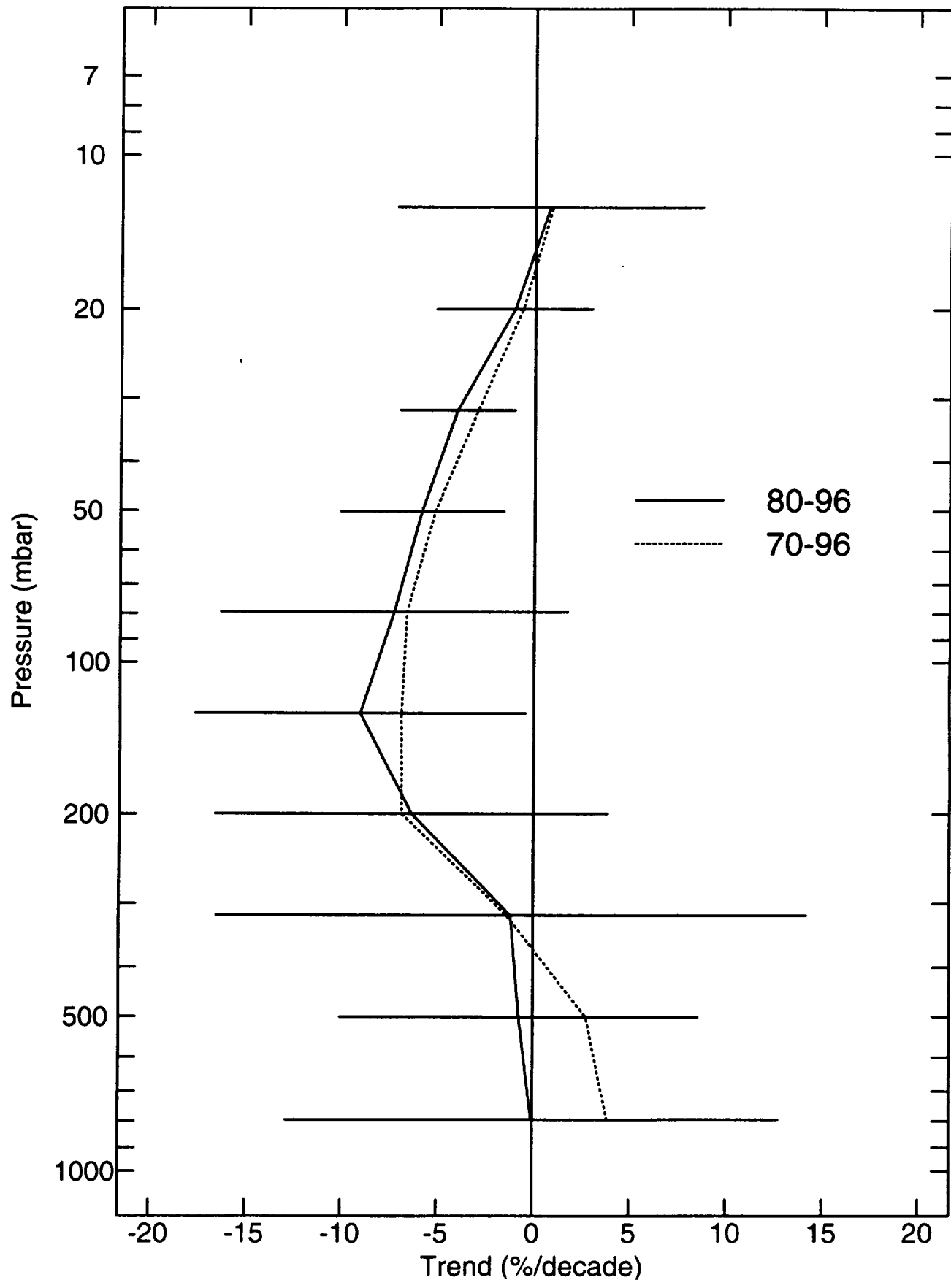
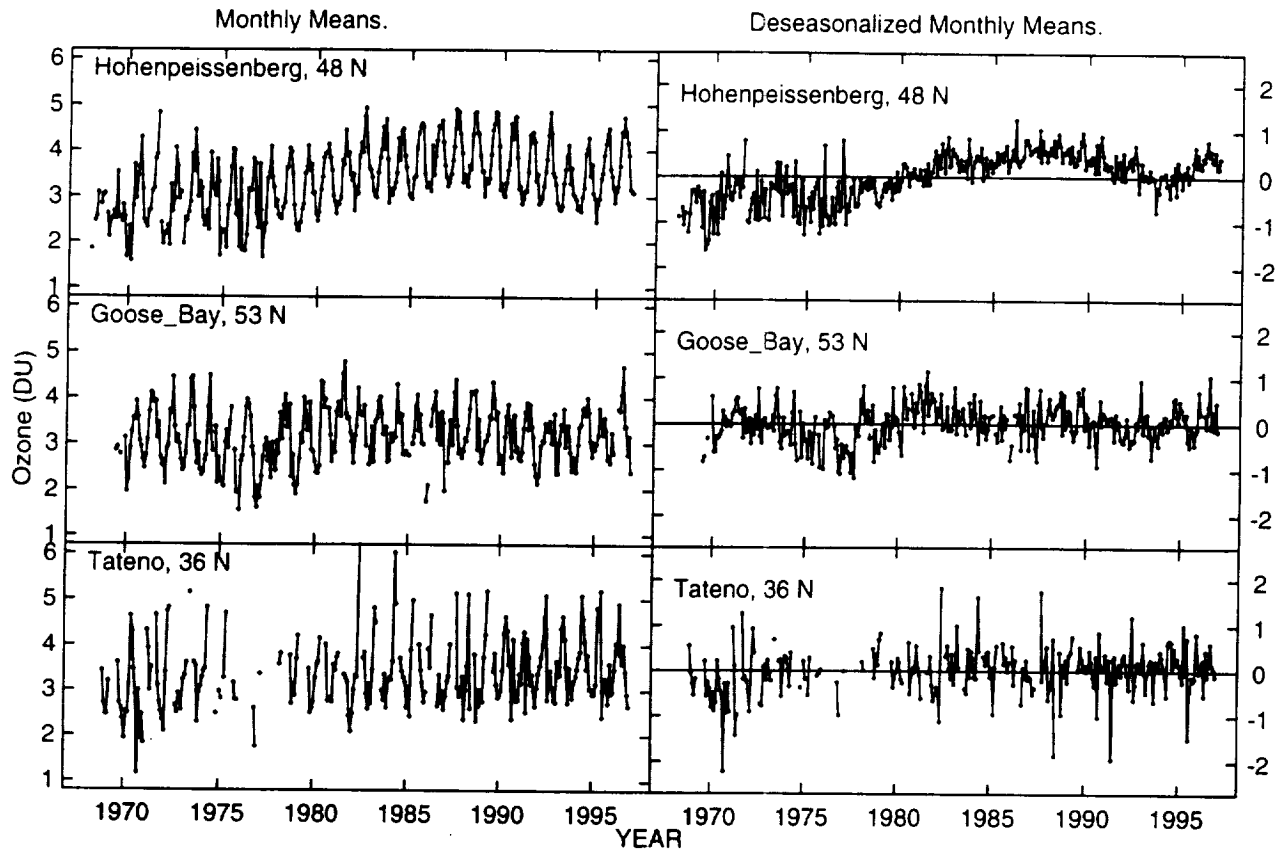


Fig. 16



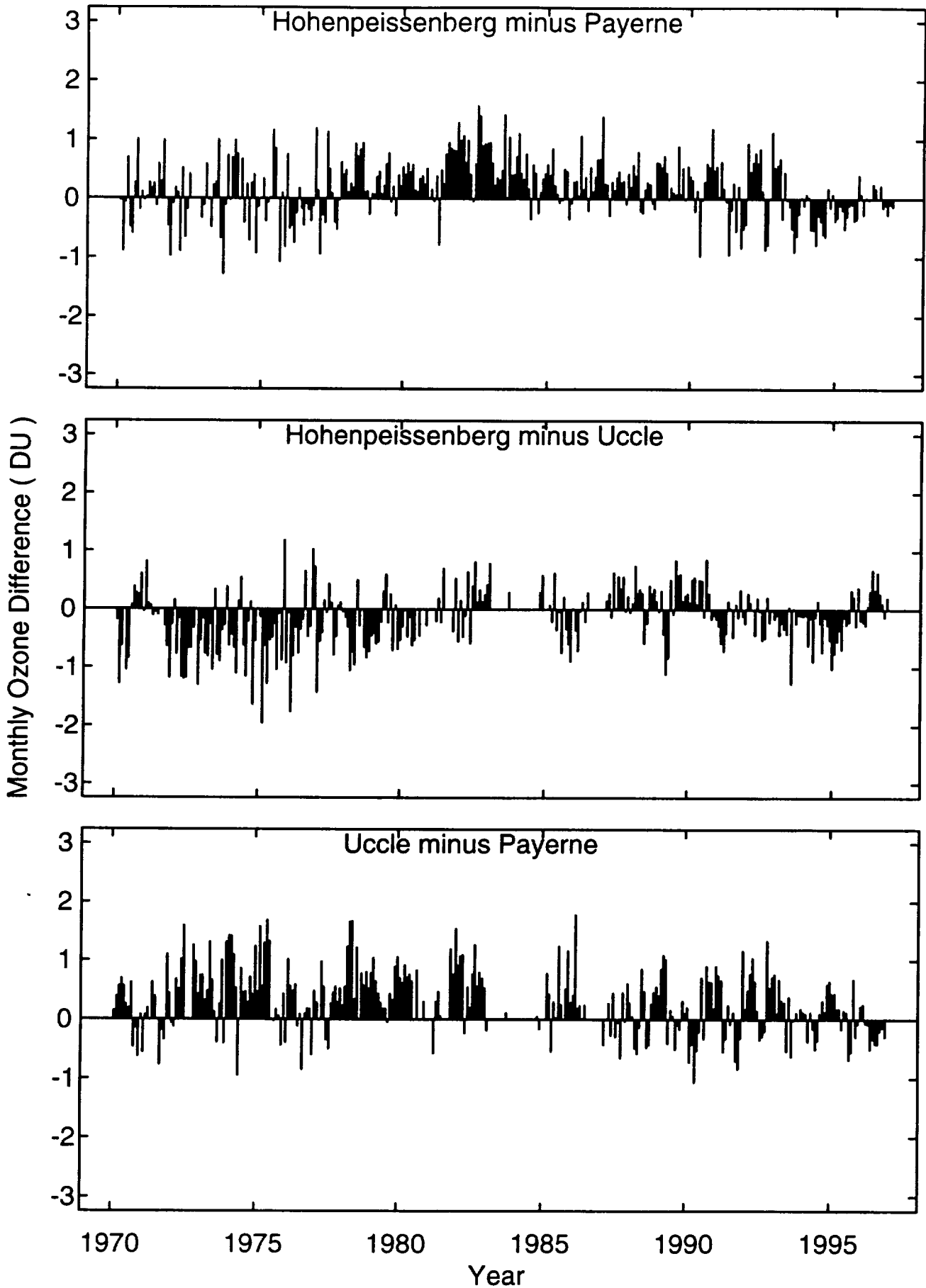


Fig. 18

1000-250 mbar

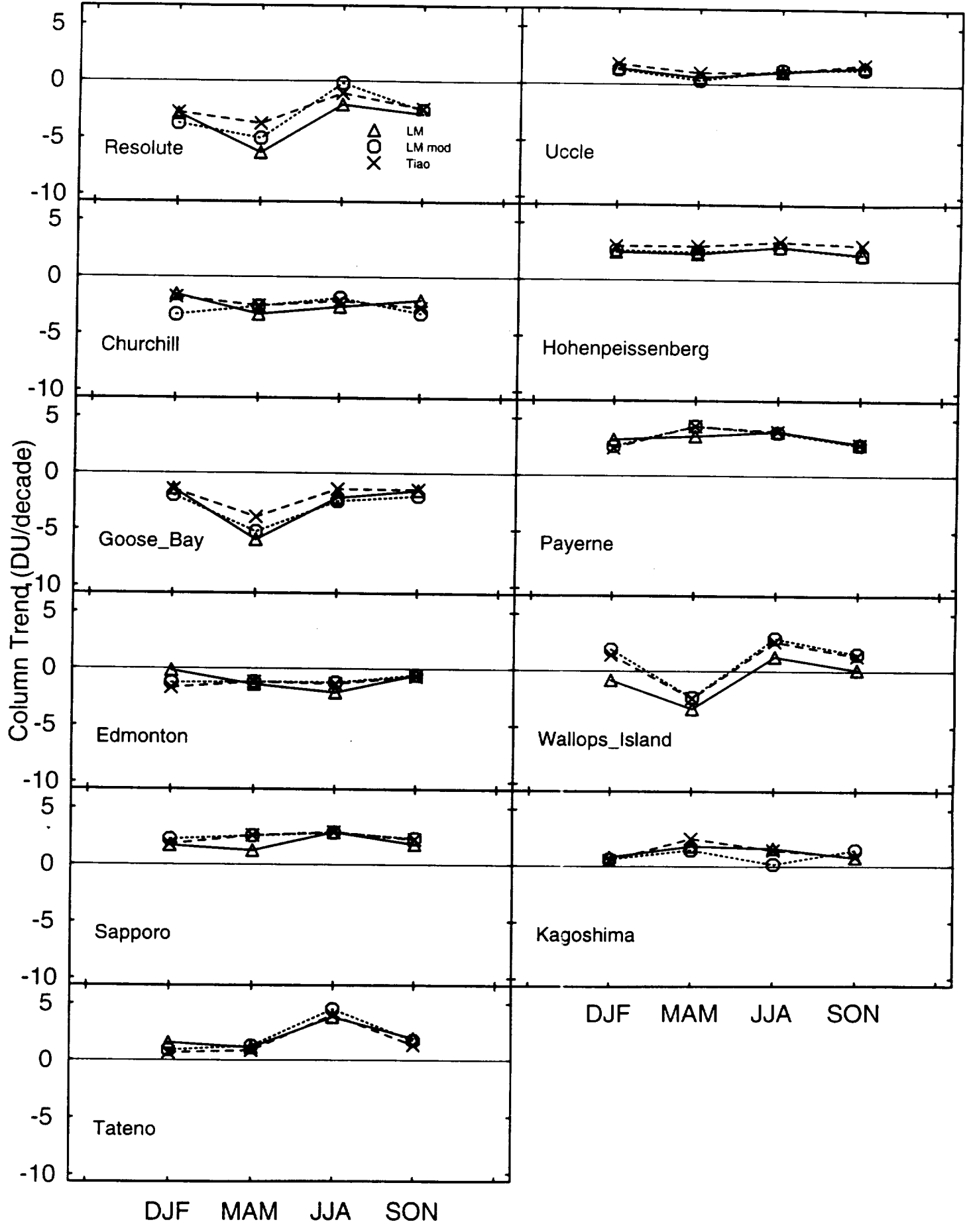


Fig. 19

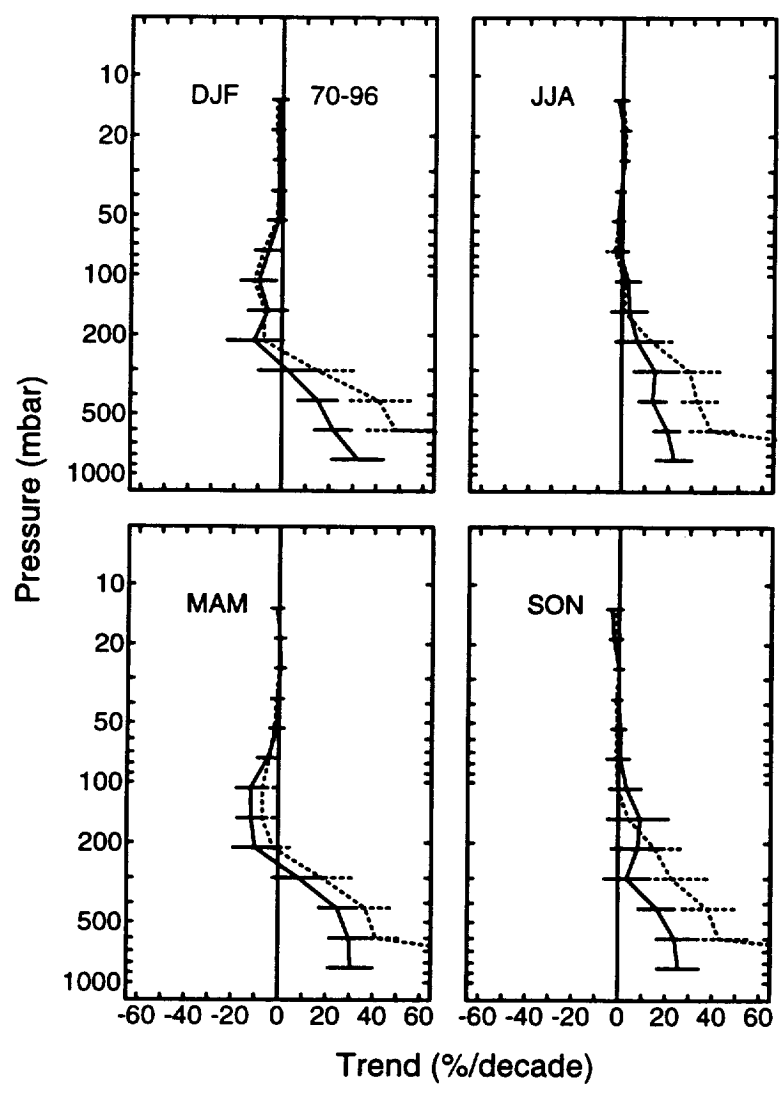


Fig. 20

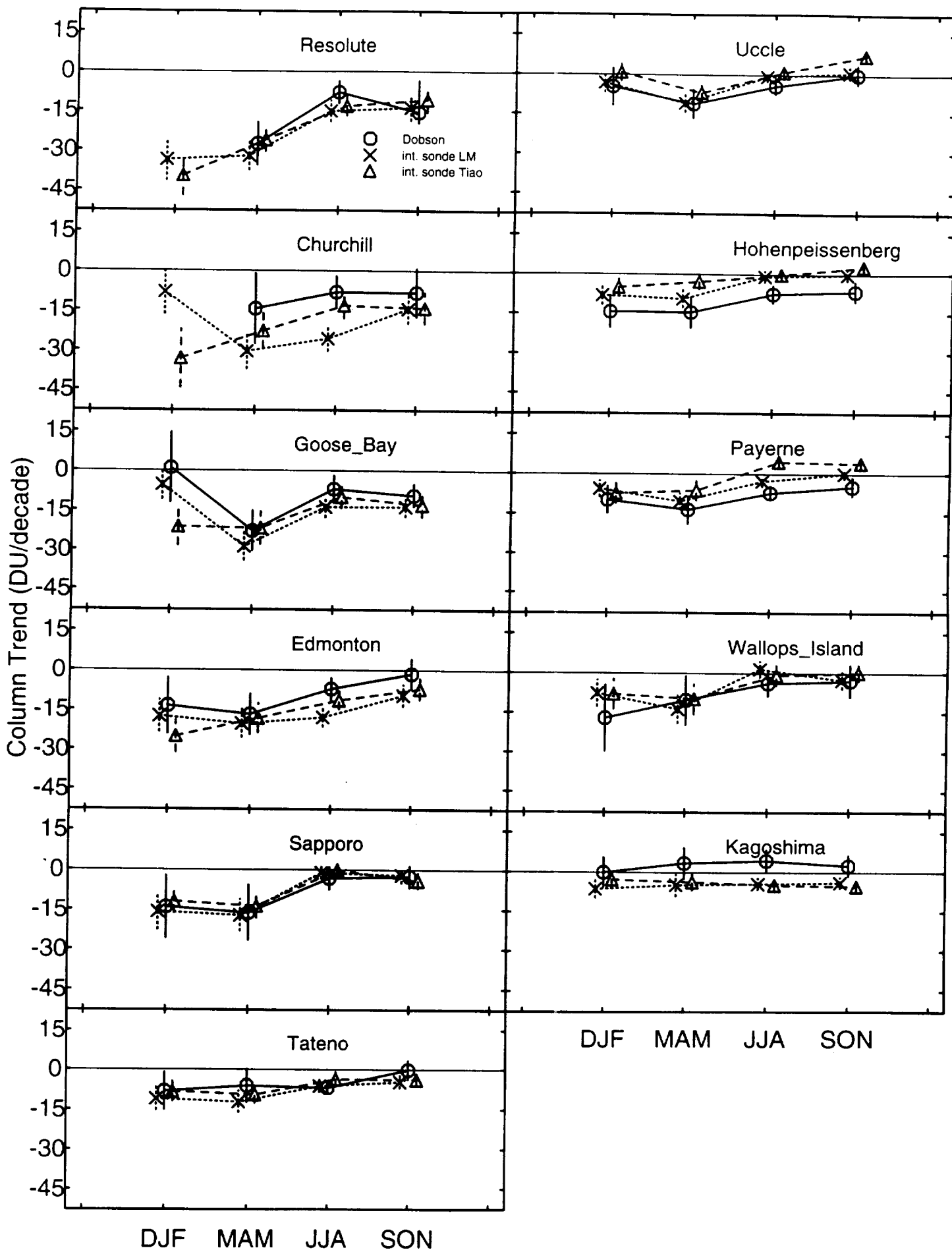


Fig. 21

