

## Comparisons of observed ozone trends in the stratosphere through examination of Umkehr and balloon ozonesonde data

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**Abstract.** During the past several years, several authors have published results of the annual and seasonal trends depicted in the total ozone data from both satellite and ground-based observations. The examination of the vertical profile data available from the balloon ozonesonde and Umkehr observations, however, has been generally restricted to limited periods and to nonseasonal trend calculations. Within this study, we have examined the nonseasonal and the seasonal trend behavior of the ozone profile data from both ozonesonde and Umkehr measurements in a consistent manner, covering the same extended time period, 1968–1991, thus providing the first overall comparison of results. Our results reaffirm the observation of significant negative ozone trends in both the lower stratosphere (15–20 km), about  $-6\%$  per decade, and upper stratosphere (35–50 km), about  $-6\%$  per decade, separated by a nodal point in the region of 25–30 km. The upper stratosphere decrease is, apparently, associated with the classic gas phase chemical effect of the chlorofluorocarbons, whereas the cause of the lower stratospheric decline is still under investigation, but may well be associated with the chlorine and bromine chemistry in this region.

### 1. Introduction

Within the continuing response to the concerns of possible stratospheric ozone depletion associated with the release of chlorofluorocarbons (CFCs) and halons into the stratosphere, one fundamental question is the comparison of the satellite versus ground-based profile measuring systems. On the one hand, the ground-based data tend to begin in the mid-1960s, but are spatially limited, whereas the satellite data are more global, but tend to begin in the late 1970s. Thus each system is complementary to our understanding of the overall changes of ozone with time. Many articles and publications have been presented in recent times, offering results of various parts of the total information. *Reinsel et al.* [1987] and *Bloomfield et al.* [1982], for example, performed statistical analyses of the Umkehr observations covering the period from the mid-1960s to the early 1980s, whereas more recent trend calculations have focused on the period from about 1978 [e.g., *DeLuisi et al.*, 1989; *Reinsel et al.*, 1989; *World Meteorological Organization (WMO)*, 1988, 1989] for comparison with the available satellite data. Overall, the results have been quite consistent, indicating a significant

ozone decrease in the upper stratosphere of about  $-3$  to  $-5\%$  per decade.

More recently, *Mateer and DeLuisi* [1992] have developed a new algorithm for the estimation of the Umkehr vertical ozone profile that makes use of improved ozone absorption coefficients and their temperature dependence. This algorithm has been implemented at the World Ozone Data Centre (Toronto) since 1993. Thus we are presented with an opportunity to reevaluate the Umkehr ozone profile trends, utilizing the results from the updated algorithm as well as extending the data set as far back in time as warranted by the available data to account for aerosol effects [*DeLuisi*, 1979].

In addition to the Umkehr observations, the balloon ozonesonde data also extend back to the late 1960s. These ozonesonde data are extremely complementary to the Umkehr data in that they overlap in the lower stratosphere. *Tiao et al.* [1986] presented a detailed statistical trend analysis of monthly averages of balloon ozonesonde data through 1982, and their analysis was updated using data through 1986 in the WMO ozone assessment report [*WMO*, 1989] with very similar results. Basically, they found a significant ozone decrease in the lower stratosphere (approximately between 13 and 22 km) of about  $-6\%$  per decade, peaking at about 20 km. Furthermore, *Miller et al.* [1992] examined the relationship of this trend to that of rawinsonde temperatures in the same altitude region. They found a statistically significant negative trend in the observed temperatures that was in substantive agreement with that computed via a radiative model utilizing the observed ozone decreases. In the lower troposphere the evidence was for positive ozone change, although except for the lowest layer, the results were not statistically different from zero [*Angell*, 1986; *Logan*, 1985; *Bojkov*, 1988]. We note, furthermore, that *Bojkov* [1988] examined the long-term ozone records of surface observations and found increases of about 10% per decade. *London and Liu* [1992], in addition, found signifi-

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cantly positive trends in the midtroposphere as well as significantly negative trends in the lower stratosphere.

We will present the results of extending the trend analysis of the Umkehr and balloonsonde data from 1968 to June 1991, with particular emphasis on the following points: (1) discussion of the trend effects as a function of season and (2) comparison of the Umkehr and balloonsonde data in the overlap region.

## 2. Statistical Methodology

The seasonal, trend, autocorrelation, and other effects in the ozone data are accounted for by the use of the regression time series model given below [e.g., *Reinsel et al.*, 1987, 1994a, b; *Tiao et al.*, 1986]. To investigate the seasonal nature of the ozone trends, the model used is

$$Y_t = \sum_{m=1}^{12} \mu_m I_{mt} + \sum_{s=1}^4 \omega_s I_{st} X_t + \gamma_1 Z_{1t} + \gamma_2 Z_{2t} + \delta U_t + N_t \quad (1)$$

where  $Y_t$  is the monthly average value of the ozone amount for month  $t$ ;  $I_{mt}$ ,  $m = 1, \dots, 12$ , denotes an indicator series for the  $m$ th month of the year, which equals 1 if  $t$  corresponds to month  $m$  of the year and zero otherwise;  $\mu_m$  denotes the mean ozone amount in month  $m$ ;  $I_{st}$ ,  $s = 1, \dots, 4$ , denotes an indicator series for the  $s$ th season of the year, which equals 1 if  $t$  corresponds to season  $s$  of the year and zero otherwise;  $X_t$  denotes a linear trend function starting in January 1970;  $\omega_s$  denotes the trend or change (in Dobson units per year) starting in January 1970 for season  $s$ ;  $Z_{1t}$  denotes the  $f_{10.7\text{cm}}$  solar flux series;  $Z_{2t}$  represents ancillary data that may be included within the analysis, such as aerosol optical depth;  $\delta U_t$  is an intervention level shift term; and  $N_t$  is a residual noise series modeled as a first-order autoregressive (AR(1)) model,

$$N_t = \phi N_{t-1} + \varepsilon_t \quad (2)$$

where  $\varepsilon_t$  is a series of white noise.

The intervention level shift term is included, when necessary, to account for discontinuities in the observed data that result from factors such as changes in instrumentation or movement of station location. It is represented by a time series,  $U_t$ , consisting of zeros up to the discontinuity and 1s afterward, and the statistical procedure estimates the magnitude  $\delta$  of the shift.

The seasonal trend estimates of the data obtained from model (1) correspond to four seasons of the year, which we have grouped as winter (December–February), spring (March–May), summer (June–August), and fall (September–November). The four seasonal trend values are expressed in percent per decade, obtained by dividing the seasonal trend estimate (in Dobson units) by the average of the mean ozone amount within the season, and then scaling to percent per decade. These percentage trends will be referred to as the seasonal trend estimates. Similarly, an annual trend estimate is calculated from the four seasonal trend estimates, also scaled to percent per decade.

## 3. Ozonesonde Data and Trend Results

A detailed statistical trend analysis of monthly averages of balloon ozonesonde readings from 1970 through 1982 was

**Table 1.** Ozonesonde-Umkehr Layers Utilized in Analysis

Layer	Upper Boundary, mbar	Approximate Upper Boundary, km
1A	716	2.9
1B	507	5.5
1C	359	8.0
1D	253	10.3
2A	179	12.5
2B	127	14.7
3A	90	16.9
3B	63	19.1
4A	45	21.3
4B	32	23.6
5A	22	25.8
5B	16	28.1
6A	11	30.5
6B	8	32.8
7	4	37.5
8	2	42.7
9	1	48.1

presented by *Tiao et al.* [1986]. This analysis was updated in the *WMO* [1989] ozone assessment report, using data through 1986. The two trend analyses show very similar results even though the extended period encompassed the El Chichon volcanic eruption and the significant El Nino event of 1982. For ease of comparison with other available ozone profile data, such as Umkehr, the ozonesonde soundings were converted into fractional Umkehr layers, which are presented in Table 1. The ozonesonde stations, data spans, and number of points are given in Table 2. We note, also, that *London and Liu* [1992] and *Logan* [1994] present an excellent summary of the data quality and quantity of the ozonesondes.

At the Canadian stations within the period 1979–1980, the sites changed instrumentation from the Brewer-Mast (BM) to the electrochemical concentration cell (ECC), requiring an intervention term in the analysis as described above. Consideration of the data at these sites, however, revealed that a significant problem ensued when the intervention term was applied, in that the trends in total ozone did not tend to agree with the trends summed over all layers. This is now considered to be the result of applying the correction factor (the ratio of a total ozone measurement to the summation of the ozonesonde data) equally at all layers for each instrument [e.g., *Hilsenrath et al.*, 1986; *WMO*, 1993; *Logan*, 1994]. Therefore, for this study we have analyzed the data both in its published corrected form and in an uncorrected form, wherein we have divided each profile by the appropriate correction factor. For this analysis, only data with correction factors within 0.9–1.2 for the BM sonde and 0.9–1.15 for the ECC and Japanese sondes are utilized to help maintain the overall quality of the data with time. One effect of this limitation, of course, is that we tend to lose data for the analysis. We then extended the analysis for the BM sonde to include upper limit correction factors 1.3, 1.4, and 1.5 but found no apparent effect on the results. Thus we present our results only for the original limits cited above. At Payerne, an intervention term was included only in layers 1A–1D to account for a change in balloon release time in 1977. Finally, as we examined the results, it became increasingly clear that a division of the seasonal trend analysis into

**Table 2.** Ozonesonde Stations and Data Spans

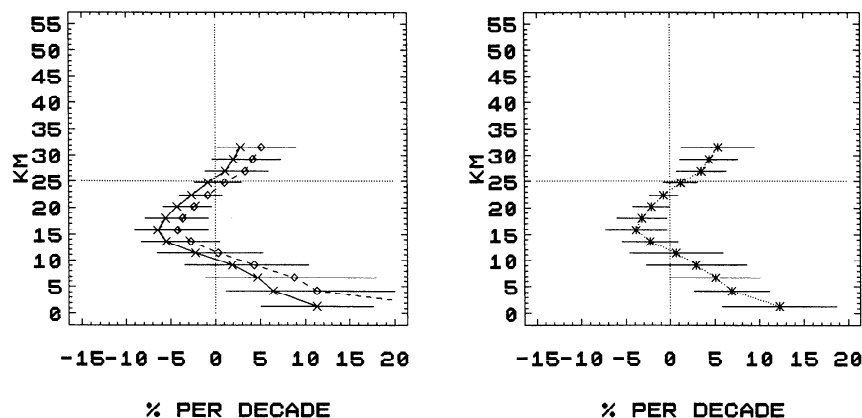
Station	Latitude, deg N	Data Span	Dates of Intervention	Points
Churchill	59	Sept. 1973 to June 1991	Sept. 1979	273
Edmonton	54	Oct. 1970 to June 1991	Sept. 1979	369
Goose	53	Dec. 1970 to June 1991	Sept. 1980	392
Lindenberg	52	Jan. 1975 to April 1991		467
Hohenpeissenberg	48	May 1968 to May 1991		1896
Payerne	47	Aug. 1968 to Dec. 1990		1383
Sapporo	43	Dec. 1968 to June 1991		168
Wallops Island	38	May 1970 to June 1991		337
Tateno	36	Nov. 1968 to June 1991		238
Kagoshima	32	Dec. 1968 to June 1991		125

an extended period, 1968 through June 1991, and a recent period, 1977 through June 1991, was not practical, in that the shorter record became too noisy. Therefore, we present the seasonal results only for the lengthier period. As a major element of this study is the comparison of the balloon ozonesonde data with that of the Umkehr, we restrict this study to stations in the midlatitudes of the northern hemisphere, which constitutes the major overlap area. Consequently, we do not consider results for the two southern hemisphere stations and Resolute. Finally, we note that during the last several years, major efforts have been initiated to reanalyze the available data in a coherent manner. This study incorporates all total ozone data that have been reprocessed and archived in the World Ozone Data Centre, Toronto, as of April 1994.

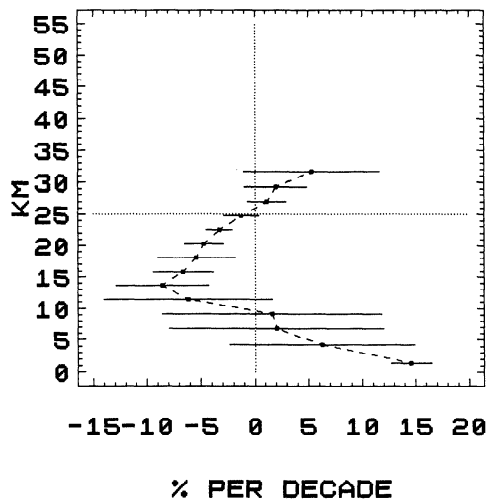
Within Figure 1 (left panel) we display the results of the annual trend analysis based on the nonseasonal trend form of (1) for the databases with and without the correction factors. This figure displays the average trend estimates over the 10 northern midlatitude stations for each layer, with associated 95% confidence limits. We see that the shapes of the two average trend curves are very similar and that the negative trend in the 15- to 20-km region is significant in both. In addition, the trend values of the corrected data are slightly more negative than those for the uncorrected data, indicating that, overall, there is a trend in the correction factors applied to the ozonesonde data. This is discussed in more detail by Logan [1994]. This suggests a change in the ozonesonde

accuracy with time that will require further analysis. As part of this, we note that above the region of ozone decrease, the trends become positive, and are statistically significant above about 25 km. This feature, however, is clouded by the fact that this is the region where the pump efficiency corrections are added and both this and the absolute altitude are affected by the accuracy and changes in the pressure sensors utilized [e.g., WMO, 1993]. Consequently, we add on the diagram a dotted line at 25 km to delineate that the trends above this level may be instrumentally induced and are, perhaps, not to be trusted. One consequence of this observed increase in the upper levels is that it may impact the other layers through the correction factors. If the sonde values in the upper levels are artificially increasing, this would be reflected in the computed total ozone, which would then require a negative trend in the correction factors. This, in turn, redistributes the change throughout the profile.

In the troposphere, the data also show an increase over time that appears to be significant in the lower layers. However, in the lowest layer the values for the uncorrected data go off scale to over 40% per decade. This is mainly due to the observations at Payerne, where the annual trend value is 150% per decade  $\pm 13\%$  even after accounting for the change in balloon launch time and is caused by rapidly increasing ozone values in the region up to 10 km during the last 5 years of data record. These data have been examined by Staehelin and Schmid [1991], and it is not clear as to the cause of this discrepancy in trends in layer 1 between



**Figure 1.** Annual ozone trend for the period 1968–1991 from balloonsondes. (Left) Crosses, database with correction factors applied; diamonds, without correction factors applied. (Right) Asterisks represent database without corrections and with Payerne deleted. Error bars represent 95% confidence limits.



**Figure 2.** Annual ozone trend for period 1977–1991 from balloonsondes. Database with correction factors applied.

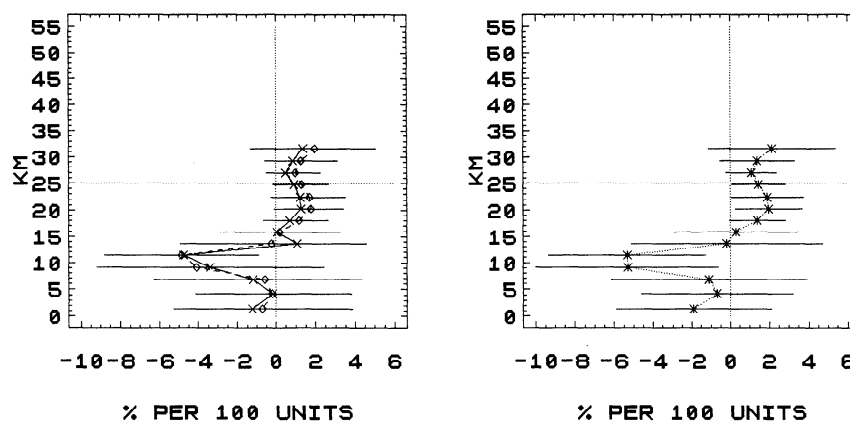
Payerne and most other ozonesonde stations. In view of this feature, however, we have computed the average trends in, yet, a third manner, which is without the corrections and with Payerne removed. These results are depicted in Figure 1 (right panel). Again, the shape of the trend profile with altitude is very similar to the previous results, but the values in the lowest layer are reduced to about the 12% per decade level. In any case, as has been discussed by Logan [1985, 1994] and Tiao *et al.* [1986], we must be careful not to overstate the results to the entire globe without additional evidence. In particular, Logan [1994] discusses the increases in the troposphere within the context of the ozonesonde quality and sensitivity to changes in sulfur dioxide as well as the chemical production of tropospheric ozone. In this regard, the general pattern of ozone loss in the lower stratosphere is supported by results from the stratospheric aerosol and gas experiment [McCormick *et al.*, 1992; WMO, 1989] for a shorter period, and further analysis is required on the magnitude and extent of the effect.

As a further consideration, we have examined the annual trend results for the 10 stations with correction factors applied for the later period, 1977–1991. The results are presented within Figure 2. From this diagram we see that the

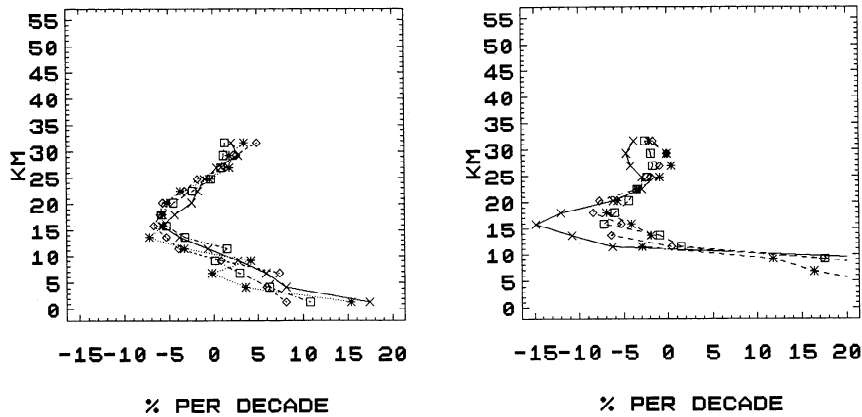
most recent trends are very similar to the longer-term trend, but are somewhat more negative in the upper troposphere–lower stratosphere (7–17 km) and slightly more positive in the lowest layer. The error bars of the trend estimates in Figures 1 and 2 are quite large, however, and show that the results are not significantly different in the statistical sense. Nonetheless, these results do support the total ozone trend observations [WMO, 1989] in which the total ozone trends are observed to have become progressively more negative with time. Our results are in general agreement with those of London and Liu [1992].

Finally, as noted above, we have included the effects of the solar variation into the computations by the inclusion of the  $f_{10.7cm}$  solar flux term in model (1). The results of the estimated solar coefficients averaged over the stations for the same three databases of Figure 1 are plotted in Figure 3. Here, several interesting elements appear. Overall, there is some evidence of a mild solar effect in the lower stratosphere (17–25 km), where the coefficients are positive, of the order of 1–2% per 100 solar flux units, but generally the estimates are only barely statistically significant in this region. Also, within the lower troposphere the results are statistically not significant. However, within the 10- to 12-km region, we see strong negative coefficients that appear significant. Clearly, this cannot be attributed to an actual solar association and must be examined further. From the perspective of a possible influence of the “solar effect” on the trend calculations, we note that the unusual perturbation in the solar coefficient estimates at 10–12 km is several kilometers below the significant negative trend estimates in ozone.

Within Figure 4 (left panel) we indicate the average seasonal trends for the 10 northern mid-latitude ozonesonde stations for the standard case with the ozonesonde data corrected. Examination of this figure indicates that, in general, there is little or no indication of any substantive seasonal effect in the trends averaged over the stations. Logan [1994], however, points out that the European stations do indicate a seasonal effect, with the largest trend in spring and smallest in summer, which is in agreement with the total ozone data from both the total ozone mapping spectrometer (TOMS) and Dobson instruments [Stolarski *et al.*, 1992; WMO, 1992]. We interpret our results to mean that the more generally limited number and quality issues of the ozonesonde observations makes them insufficient to resolve



**Figure 3.** Same as Figure 1 but for coefficients of  $f_{10.7}$  effect.



**Figure 4.** Seasonal trend analysis for period 1968–1991 from balloonsondes. (Left) All stations with corrections applied. (Right) Hohenpeissenberg and Payerne combined with corrections applied. Seasons expressed as winter (crosses), spring (diamonds), summer (asterisks), and fall (squares).

the seasonal trend features. To test this hypothesis, we calculated the seasonal trends for the two stations with the greatest number of data points (Table 2), Hohenpeissenberg and Payerne, and the results are portrayed in Figure 4 (right panel). In this figure we see that the greatest depletion in the region from 10 to 20 km occurs in the winter with spring, just barely greater than the other seasons.

Comparing our results by station to those of Logan [1994], we find that for Hohenpeissenberg we are quite consistent in the stratosphere but that we tend to be more positive in the lower layers of the troposphere. For Payerne, our computations show somewhat greater negative trends for winter and summer in the 15- to 20-km region, but are otherwise consistent.

We also note in Figure 4 (right panel) that the trend results go off scale in the lowest layers. For layer 1A the winter, spring, summer, and fall results at Hohenpeissenberg and Payerne are indicated in Table 3.

#### 4. Umkehr Data and Trend Results

A recent development of the Umkehr measurement system is the new data inversion algorithm presented by Mateer and DeLuisi [1992]. This revised inversion algorithm, amongst other elements, utilizes the new ozone absorption coefficients of Bass and Paur, which results in a change in the scale of measurement and their temperature dependence. The revised algorithm does not alleviate the requirement for consideration of the stratospheric aerosol effect, though, and this will be discussed below. While Mateer and DeLuisi do not consider the data in Umkehr layers 3 and 4 (about 15–25 km) to be totally independent for trend analysis, we have retained these layers within our statistical examination, as they overlap the balloonsonde region of interest. The major

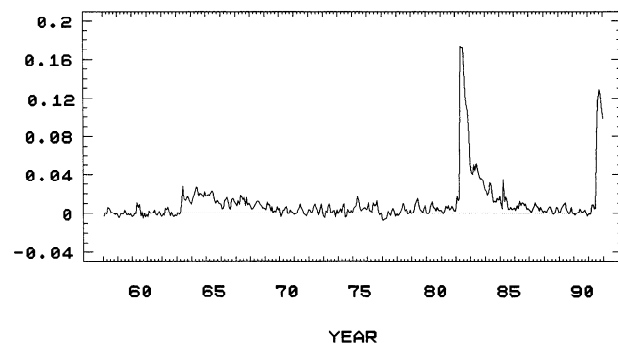
issue is how much information is available in each layer versus how much is inferred. For example, if the ozone profile is directly retrieved above 20 km along with the total ozone, can we infer the nature of the ozone changes below 20 km? DeLuisi *et al.* [1994] have compared information from the Umkehr and solar backscattered ultraviolet (SBUV) satellite instruments and indicated that agreement exists down to about layer 3, but below that, substantial differences occur.

With respect to the impact of stratospheric aerosols on the data, DeLuisi *et al.* [1989] and Mateer and DeLuisi [1992] have shown that the Umkehr observations are very susceptible to contamination from stratospheric aerosol loading and that the computation of trends must take this into consideration. As an example, Mateer and DeLuisi [1992] have demonstrated that the errors in the Umkehr retrieval for layer 8 (about 40 km) are of the order of  $-6\%$  per 0.01 optical thickness and the sign of the error reverses at the lower layers. To help place this effect in perspective, Figure 5 shows the stratospheric optical thickness as a function of time as determined from the Mauna Loa Observatory measurements of transmissivity via the following relationship:

$$T = e^{-\tau}$$

$$\tau = \tau_{\text{background}} + \tau_{\text{stratospheric aerosols}}$$

$$\tau_{\text{stratospheric aerosols}} = \ln(T_{\text{background}}/T)$$



**Figure 5.** Time series of stratospheric optical thickness at Mauna Loa, Hawaii.

**Table 3.** Seasonal Trends in Layer 1A for Hohenpeissenberg and Payerne

	Winter	Spring	Summer	Fall
Hohenpeissenberg	50.3	25.8	24.0	29.7
Payerne	25.3	21.5	23.2	25.0

Units are percent per decade.

**Table 4.** Summary of Umkehr Data

Station	Latitude, deg N	Data Span	Dates of Intervention	Points
Edmonton*	54	May 1969 to Feb. 1988		305
Belsk	52	Feb. 1968 to June 1991		984
Arosa	47	Jan. 1968 to Dec. 1990		3328
Haute Province	44	Sept. 1983 to June 1991		1228
Sapporo*	43	April 1968 to June 1991	March 1976, March 1985, June 1989	629
Boulder	40	Feb. 1978 to June 1991		1107
Lisbon	39	Jan. 1968 to June 1991	Jan. 1978, Jan. 1988	1381
Tateno*	36	Jan. 1968 to June 1991	Jan. 1976	2412
Kagoshima*	32	Feb. 1968 to May 1991	Jan. 1977, Jan. 1984, Jan. 1986, Dec. 1989	602
Cairo	30	Jan. 1978 to May 1991		387
New Delhi	29	Aug. 1974 to June 1991		522

where  $T$  is the transmission from the sun photometer (300–2500 nm) and  $T_{\text{background}} = 0.933$ , the average  $T$  from 1958–1962 and 1969–1973.

As aerosol optical depth measurements are not available near 320 nm, it becomes necessary to infer the optical depth at this wavelength from measurements at other wavelengths. In particular, the extensive set of measurements on atmospheric transmission of solar radiation (300–2500 nm) is believed to correspond to an effective wavelength near 700 nm [e.g., *DeLuisi et al.*, 1989]. Although we would require further information on particle size to associate the changes at 700 nm to those at 320 nm, the close relation between atmospheric transmission and the optical depth of stratospheric aerosols suggests that it is reasonable to consider the transmission data as a proxy for the timing of the effect of the aerosols on the Umkehr record. This would certainly not be true for individual days when the aerosol patchiness would greatly affect the results, but does appear to be true when considering the large-scale effects of El Chichon in 1982 and Mount Pinatubo in 1991 [e.g., *McCormick et al.*, 1984; *Trepte et al.*, 1993]. We see from Figure 5 that the aerosol optical depth perturbations from Mount Agung in 1963 were of the order of 0.02 and that the more recent eruptions of El Chichon and Mount Pinatubo were more than a factor of 5 greater. Thus, any consideration of the trend results from the Umkehr observations must include the effects of the long-lived stratospheric aerosols. In particular, we note that the effects of El Chichon and Pinatubo are an order of magnitude larger than the others and at these levels of aerosol loading, nonlinear effects may enter into the effect on Umkehr measurements. Therefore, we remove from consideration those data between November 1982 and June 1983 and do not extend the analysis into the Pinatubo period.

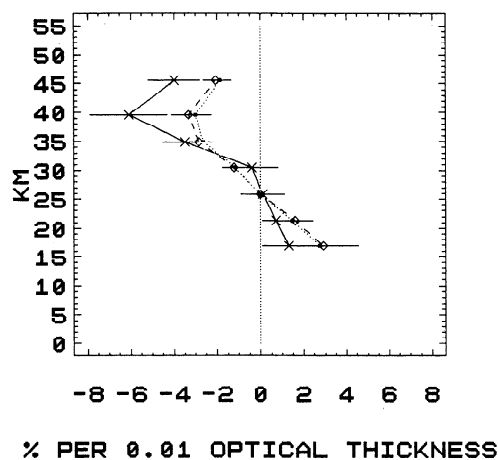
The database for the Umkehr analysis is presented in Table 4. The data are essentially limited to the midlatitudes of the northern hemisphere. To be consistent with the philosophy of the ozonesonde-Umkehr comparisons expressed earlier, we do not include Poona, at 19°N latitude, within this study. As for the situation with the ozonesondes, interventions were deemed necessary for several stations to account for instrument changes, etc. We also note that the data considered within this study reflect the revisions as of April 1994. The data from Arosa, however, are a special situation. In March 1994 we were informed of a substantial revision to the Arosa data that resulted in substantive

changes in the total ozone values during our period of interest from 1978. However, these data were noted as provisional, and it was stated that further, minor revisions were forthcoming. On this basis, we examined the trends determined from the revised Arosa data against those determined from the data in the actual archive. We found that the trend effects of the revisions on the Umkehr data were relatively minor and when averaged within an overall regional or latitudinal band were even less pronounced. Consequently, we have utilized only the original archived Arosa data within this study.

For these 11 stations the trend analyses were determined utilizing (1) with the  $\gamma_2 Z_{2t}$  term included to account for the effect of stratospheric aerosol on the measurements. The major issue in doing this is the establishment of the proper database for the aerosol content. For example, the database from Mauna Loa Observatory is continuous within the time frame of the Umkehr observations, but is located near the southern boundary and may not be the best representative of the Umkehr sites. *DeLuisi et al.* [1989] have considered this problem and have developed a methodology that combines available lidar data at Hampton, Virginia, and other stations along with the SAGE II observations with data from Mauna Loa. Basically, we have established four time series for the aerosol term, one each for 20°N, 30°N, 40°N, and 50°N.

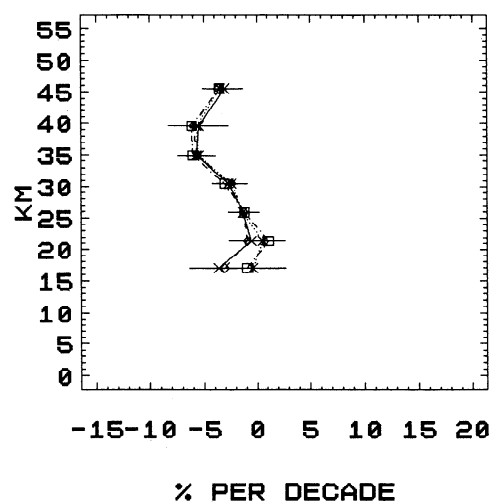
Trend results have been obtained using the new inversion algorithm for the Umkehr data for the period from January 1968 through June 1991. The overall estimates for trend, solar cycle, and aerosol effects were obtained by combining estimation results over the 11 Umkehr stations. As the consideration of the aerosol error can be a major effect in the estimation of the ozone trends, we examine first the results of the optical thickness coefficient estimations within Figure 6. This diagram depicts the results of the *Mateer and DeLuisi* [1992] model calculations along with our coefficient estimates for both the 1968–1991 and the 1977–1991 data series. We see that the coefficient estimates for the two time periods are consistent among themselves and are quite similar in shape to the model calculated values. Our current estimates are slightly more positive than the theoretical values of *Mateer and DeLuisi* [1992]. These results suggest that we have incorporated the aerosol impact on the Umkehr data in a reasonable manner and that we may have some confidence in the solar coefficient estimates.

Within Figure 7 we depict the annual trend and solar



**Figure 6.** Umkehr aerosol error. Theoretical value (crosses) and computed values for 1968–1991 (diamonds) and 1977–1991 (small squares).

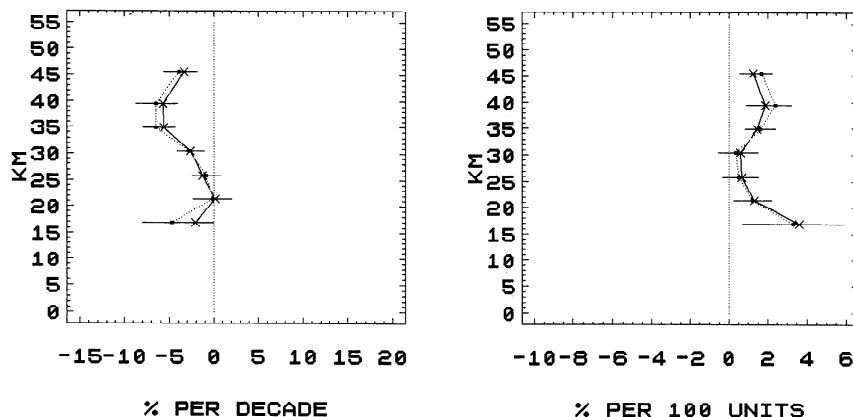
coefficient estimates for the 1968–1991 and the 1977–1991 time periods, respectively. In general, the results are quite similar, and the values are not statistically different between the two data periods. For the trends, the values are about  $-6\%$  per decade in layers 8 and 7 (35–40 km). The negative trend decreased to near zero down to layer 4 (21 km) and then increased again in layer 3 (17 km). Examining the trend estimates more closely, we see that the later period indicates slightly more negative trends than the longer period in both the upper and lower layers, and remain the same (near zero) for the layers between. While the results within layer 3, showing about twice the negative trend for the shorter period, may be driven by the general total ozone results within the algorithm, the upper layer findings are a new result. These must be considered within the total context of theory and observation of the chemical and meteorological elements. With respect to the solar effects, Figure 7 (right panel), the coefficients for the shorter period are slightly more positive than for the overall period in the two topmost layers, but not below. One additional point that should be made is that the new Umkehr algorithm has the tendency to lower the altitude of the maximum effect in the upper layers. That is, from the previous algorithm the tendency was for



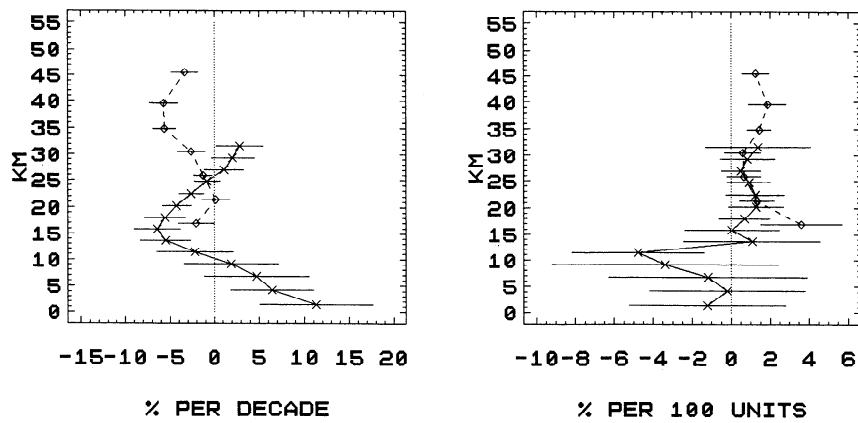
**Figure 8.** Same as Figure 4, but for Umkehr data from 1968–1991.

the trend results to have the largest negative values in layer 9 (45 km) with the largest error bars. The new results show curvature at the upper layers [see *Reinsel et al.*, 1994a, b], with the largest trends in layers 8 and 7 (35–40 km). A similar effect is noted for the solar coefficients. These findings are in better agreement with the solar effects expected from theory [Wuebbles *et al.*, 1991]. We note, also, that within layer 3 (17 km) the solar coefficients indicate a relative maximum that will be discussed below within the context of comparison with the ozonesonde results.

Trend results have also been obtained for the longer data period based on the seasonal trend analysis model, and the results are indicated in Figure 8. The overall seasonal trend estimates were obtained for each of the four seasons and each Umkehr layer by combining trend estimation results over the 11 Umkehr stations. For this data period there is little evidence of any seasonal differences in the trends for Umkehr layers 5–9 (25–45 km). There are slightly more negative trends in the winter and spring seasons than the summer and fall seasons in layer 4 (21 km), and a moderate trend difference, of about  $-2.5\%$  per decade, between those seasons in layer 3 (17 km). This may well be reflective of the



**Figure 7.** (Left) Annual ozone trend and (Right) coefficient of  $f_{10.7}$  effect from Umkehr data for periods 1968–1991 (crosses) and 1977–1991 (small squares).



**Figure 9.** (Left) Ozone annual trend and (Right) coefficient of  $f_{10.7}$  effect for period 1968–1991 from ozonesondes (crosses) and Umkehr observations (diamonds). Ozonesonde database with correction factors applied.

effects of the total ozone on the vertical profile included within the Umkehr algorithm.

## 5. Summary

Within the previous sections, we have discussed the status of the ozone balloonsonde and Umkehr trend computations separately. In this summary, we will put the pieces together and demonstrate where the two data sets agree and disagree. For this purpose we will focus on the results from (1) Umkehr 1968–1991 and (2) ozonesonde with correction factors and with Payerne included, 1968–1991.

In Figure 9 we display the results for the annual trends and solar effects. Examining the trend results, we see that in the lower stratosphere the Umkehr and ozonesonde results cross each other. The ozonesonde trends are more positive above 25 km and more negative below, with the results marginally statistically significant. As indicated previously, the topmost levels of the balloonsonde data tend to be the most suspect, as are the lower layers of the Umkehr retrievals. One possible explanation for the Umkehr results is that the algorithm is placing too much of the total ozone change, including the troposphere, into the lower stratosphere, and this must be further evaluated. With respect to the differences in the region of 30 km, this should be a region of considerable information for the Umkehr observations. Recent work by R. Hudson et al. (personal communication, 1995) examines similar data over Europe and determines that the upper level increase is not found in the ozonesonde data from Hohenpeissenberg.

With respect to the solar effect, we see from Figure 9 (right panel) that in the overlap region the Umkehr coefficients agree quite well with those from the ozonesondes except in layer 3 (17 km), where the Umkehr values are higher and indicate a relative maximum. As the error bars are quite large in this region, however, the results are not significantly different.

With respect to the seasonal aspects of the trends, the ozonesonde data set did not provide an indication of a seasonal variation. As analysis of both the Dobson and TOMS satellite total ozone observations clearly depict a seasonal effect, we surmise that the current ground-based profile observations have insufficient signal-to-noise ratios to

discern this effect and that we would require many more observations than available. As a test of this hypothesis, examination of the data from Hohenpeissenberg and Payerne, which have the greatest number of observations, did indicate a stronger decline in winter. In the case of the Umkehr observations, the seasonal variation is depicted in layers 3 and 4 (17–21 km).

Finally, for both the ozonesonde and Umkehr data, the trend calculations appear to be slightly more negative for the 1977 to June 1991 period than for the 1968 to June 1991 period for the regions 7–17 km and above 35 km. The results are not statistically different at each layer but, overall, provide an “indication” that the trend has increased during the latter period.

In summary, the extended updates and comparisons presented within this study reaffirm the previous results of significant negative ozone trends in the lower stratosphere, about  $-6\%$  per decade, and in the upper stratosphere, about  $-6\%$  per decade, separated by a nodal point in the region of 25–30 km. The upper stratosphere decrease is, apparently, associated with the classic gas phase chemical effect of the CFCs, whereas the cause of the lower stratospheric decline is still under investigation, but may well be associated with the chlorine and bromine chemistry in this region.

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## References

- Angell, J. K., Annual and seasonal global temperature changes in the troposphere and low stratosphere, 1960–85, *Mon. Weather Rev.*, *114*, 1922–1930, 1986.
- Bloomfield, P., M. L. Thompson, and S. Zeger, A statistical analysis of Umkehr measurements of 32–46 km ozone, *J. Appl. Meteorol.*, *21*, 1828–1837, 1982.



- Bojkov, R. D., Ozone changes at the surface and in the free troposphere, in *Tropospheric Ozone* edited by I. S. A. Isaksen, pp. 83–96, D. Reidel, Norwell, Mass., 1988.
- DeLuisi, J. J., Umkehr vertical ozone profile errors caused by the presence of stratospheric aerosols, *J. Geophys. Res.*, **84**, 1766–1770, 1979.
- DeLuisi, J. J., D. A. Longenecker, C. L. Mateer, and D. J. Wuebbles, An analysis of northern middle-latitude Umkehr measurements corrected for stratospheric aerosols for 1979–1986, *J. Geophys. Res.*, **94**, 9837–9846, 1989.
- DeLuisi, J. J., C. L. Mateer, D. Theisen, P. K. Bhartia, D. Longenecker, and B. Chu, Northern middle-latitude ozone profile features and trends observed by SBUV and Umkehr, 1979–1990, *J. Geophys. Res.*, **99**, 18,901–18,908, 1994.
- Hilsenrath, E., et al., Results from the Balloon Ozone Intercomparison Campaign (BOIC), *J. Geophys. Res.*, **91**, 13,137–13,152, 1986.
- Logan, J. A., Tropospheric ozone: Seasonal behavior, trends, and anthropogenic influence, *J. Geophys. Res.*, **90**, 10,463–10,482, 1985.
- Logan, J. A., Trends in the vertical distribution of ozone: An analysis of ozonesonde data, *J. Geophys. Res.*, **99**, 25,553–25,585, 1994.
- London, J., and S. C. Liu, Long-term tropospheric and lower stratospheric ozone variations from ozonesonde observations, *J. Atmos. Terr. Phys.*, **54**, 599–625, 1992.
- Mateer, C. L., and J. J. DeLuisi, A new Umkehr inversion algorithm, *J. Atmos. Terr. Phys.*, **54**, 537–556, 1992.
- McCormick, M. P., T. J. Swisler, W. H. Fuller, W. H. Hunt, and M. T. Osborn, Airborne and ground-based lidar measurements of the El Chichon stratospheric aerosol from 90°N to 56°S, *Geofis. Int.*, **23**(2), 187–221, 1984.
- McCormick, M. P., R. E. Veiga, and W. P. Chu, Stratospheric ozone profile and total ozone trends derived from the SAGE I and SAGE II data, *Geophys. Res. Lett.*, **19**, 269–272, 1992.
- 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.
- Reinsel, G. C., G. C. Tiao, A. J. Miller, D. J. Wuebbles, P. S. Connell, C. L. Mateer, and J. J. DeLuisi, Statistical analysis of total ozone and stratospheric Umkehr data for trends and solar cycle relationship, *J. Geophys. Res.*, **92**, 2201–2209, 1987.
- Reinsel, G. C., G. C. Tiao, J. J. DeLuisi, S. Basu, and K. Carriere, Trend analysis of aerosol-corrected Umkehr ozone profile data through 1987, *J. Geophys. Res.*, **94**, 16,373–16,386, 1989.
- Reinsel, G. C., G. C. Tiao, D. J. Wuebbles, J. B. Kerr, A. J. Miller, R. M. Nagatani, L. Bishop, and L. H. Ying, Seasonal trend analysis of published ground-based and TOMS total ozone data through 1991, *J. Geophys. Res.*, **99**, 5449–5464, 1994a.
- Reinsel, G. C., W. K. Tam, and L. H. Ying, Comparison of trend analyses for Umkehr data using new and previous inversion algorithms, *Geophys. Res. Lett.*, **21**, 1007–1010, 1994b.
- Staehelin, J., and W. Schmid, Trend analysis of tropospheric ozone concentrations utilizing the 20-year data set of ozone balloon soundings over Payerne, *Atmos. Environ., Part A*, **25**, 1739–1749, 1991.
- Stolarski, R., R. Bojkov, L. Bishop, C. Zerefos, J. Staehelin, and J. Zawodny, Measured trends in stratospheric ozone, *Science*, **256**, 342–349, 1992.
- Tiao, G. C., G. C. Reinsel, J. H. Pedrick, G. M. Allenby, C. L. Mateer, A. J. Miller, and J. J. DeLuisi, A statistical trend analysis of ozonesonde data, *J. Geophys. Res.*, **91**, 13,121–13,136, 1986.
- Trepte, C. R., R. E. Veiga, and M. P. McCormick, The poleward dispersal of Mount Pinatubo volcanic aerosol, *J. Geophys. Res.*, **98**, 18,563–18,573, 1993.
- World Meteorological Organization, Report of the International Ozone Trends Panel 1988, Global Ozone Research and Monitoring Project, *Rep. 18*, Geneva, 1988.
- World Meteorological Organization, Scientific assessment of stratospheric ozone: 1989, Global Ozone Research and Monitoring Project, *Rep. 20*, Geneva, 1989.
- World Meteorological Organization, Scientific assessment of ozone depletion: 1991, Global Ozone Research and Monitoring Project, *Rep. 25*, Geneva, 1992.
- World Meteorological Organization, Third WMO intercomparison of the ozonesondes used in the global ozone observing system, Global Ozone Research and Monitoring Project, *Rep. 27*, Geneva, 1993.
- Wuebbles, D. J., D. E. Kinnison, K. E. Grant, and J. Lean, Effect of solar flux variations and trace gas emissions on recent trends in stratospheric ozone and temperature, *J. Geomagn. Geoelectr.*, **43**, 709–718, 1991.
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