Interannual Changes in the Atmospheric Ozone Derived from Ground-Based Measurements

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(Received September 8, 1992; Revised December 28, 1992)

Abstract

Total ozone data (from ground-based measurements) for 3 zones (north polar, north temperate, tropics) and for the world as a whole, as well as the ozone content in troposphere and stratosphere layers over the north polar and north temperate regions have been analyzed to detect long-term variations (trends) and short-term variations (related to the QBO, ENSO, and the 11-year solar cycle). Various multiple regression models (adjusted for the trends, interannual quasi-periodical variations, and the serial correlations) have been used to delineate the anthropogenic effects in the atmospheric ozone.

An apparent downward trend in the total column amount of ozone over mid and high latitude areas of the northern hemisphere, as well as over the tropic region, and the globe as a whole has been found during the period 1970-1990 in all seasons. The rate of ozone loss depends on the season (in the extratropical zones) and shows an increase with latitude. The trends in the layer-mean ozone at mid and high latitudes vary with altitude, *i.e.*, tropospheric ozone increases whereas stratospheric ozone decreases. The negative trends in total ozone over the northern extratropical regions are caused by the ozone depletion in the lower stratosphere (16-24 km layer).

Latitudinal changes in the 'natural' interannual variations in ozone, related to the QBO, ENSO, and the long-term solar forcing, have been discussed. We have found that the magnitude of the 'natural' variations in total ozone may depend both on the phase of the QBO (east, west), ENSO (warm, cold) and on the strength of the QBO, ENSO, and the long-term solar activity. The long-term changes in the atmospheric dynamics seem to influence the observed trends in the atmospheric ozone only slightly.

1. Introduction

The first paper dealing with the total ozone trends appeared in 1971 (Komhyr *et al.*, 1971). In that study an increase of about 6% during the decade 1961 to 1969 was found in the total

ozone values over North America. London and Kelley (1974) reported an upward trend of total ozone in the northern hemisphere and no statistically significant trend in the southern hemisphere during the 1960s. Johnston *et al.*, (1973) suggested that the positive trend over the northern hemisphere was caused by the recovery of the atmosphere from the effects of the atmospheric nuclear tests that were banned in 1962.

The hypothesis (Molina and Rowland, 1974)

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about the catalytic destruction of ozone by man-made chlorofluoromethanes stimulated a number of studies of long-term changes in total ozone. In all the studies on total ozone trends that were published thereafter and up 1988, no significant negative trend in total ozone on a hemispherical scale was found (e.g., Oehlert, 1986). In recent statistical trend analyses (e.g., Bojkov *et al.*, 1990; World Meteorological Organization (WMO), 1990; Stolarski *et al.*, 1992), a significant downward trend was calculated at high and mid latitudes in both hemispheres during the last two decades. Findings from the Total Ozone Mapping Spectrophotometer (TOMS) on the Nimbus 7 satellite are in concurrence with the trends derived from the ground-based network (Stolarski *et al.*, 1991).

The trend analyses of the ozone profile measurements, made by ozonesondes over northern mid latitudes, show tropospheric ozone increases and stratospheric ozone decreases with a peak loss in the lower stratosphere (15–20 km), (e.g., Stolarski *et al.*, 1992). Trends in the ozone profile from satellite measurements (combined SAGE 1 and SAGE 2 data) confirmed that the large ozone decreases have occurred in the lower stratosphere (below 20 km) in the region where the partial pressure of ozone is high (McCormick *et al.*, 1992).

The main reason for the fact that the recent trend estimates are different from those calculated in earlier studies is an increase of the rate of ozone loss since the late 1970s (Stolarski *et al.*, 1992; Krzyścin, 1992). Outside the tropics, the trend estimates for the last decade are larger than expected from calculations with increasing fluorocarbons and normal gas-phase photochemistry (e.g., Stolarski *et al.*, 1992). The heterogeneous chemistry on the surfaces of sulfate aerosols in a likely cause of ozone loss outside the tropics (Brasseur *et al.*, 1990). In the polar regions (especially over the southern hemisphere), the ozone loss is enhanced by heterogeneous chemical reactions taking place on the surfaces of particles in Polar Stratospheric Clouds (e.g., Solomon *et al.*, 1986).

The relatively large natural variability of the atmospheric ozone makes the trend detec-

tion difficult. There are indications of variations of total ozone induced by the 11-year solar cycle as well as by interannual circulation fluctuations (as the Quasi Biennial Oscillation-QBO and the nearly quadrennial El Niño Southern Oscillations-ENSO).

Recently, in a standard multiple regression model two methods have been used to account for the interannual natural variations in the atmospheric ozone. The multiple regression model with a linear trend assumption after 1970 (so-called 'ramp trend model') has been used widely in ozone trend studies, e.g., WMO (1990), McCormick *et al.* (1992), Stolarski *et al.* (1992), Krzyścin (1992).

In the first method of parameterization of the interannual changes in the atmospheric ozone, these changes are assumed to be proportional to indices describing factors forcing the year-to-year atmosphere variations. The following indices have been considered: sunspot number (or 10.7 cm solar radio flux), zonal wind at 50 hPa over the equator region, southern oscillation index (SOI)-normalized pressure difference between Darwin and Tahiti. In the second method, the interannual variations in atmospheric ozone are modelled as a sum of sinusoids with the periods obtained from the spectrum analysis of the ozone time series. The first method was used in many trend analyses (e.g., Bojkov *et al.*, 1990; WMO, 1990; Stolarski *et al.*, 1991; Stolarski *et al.*, 1992), but the second one only in a few (Degórska *et al.*, 1988; Krzyścin, 1992).

In the past statistical studies, the man-made effects in the atmospheric ozone were not delineated precisely. They focused either on the long-term changes (trends during the last two decades) or the short-term changes (changes with a time scale up to about one decade). In this paper, the interannual variations of the atmospheric ozone are examined to disclose the relation between the man-made and the 'natural' changes in the ozone. To achieve this goal, the two basic multiple regression models (see the passage above for the models' descriptions) have been compared. Moreover, the parameterization of the quasi-periodical variations (or periodical variations) in total

ozone has been improved taking into account the interactions between factors forcing the interannual variations in ozone.

2. The ozone data base

NASA/WMO International Trends Panel-1988 (WMO, 1990) have found strong incongruities in the published data of many ground-based ozone stations due to instrument calibrations. The corrections for these calibrations had not been applied to the past data. For several Dobson stations, a provisionally revised data set was prepared by the Panel with the corrections applied to the monthly averages of total ozone. The ozone data from only a few stations were reevaluated on a reading-by-reading basis (by station personnel). These include Belsk, Hradec Hralove, Uccle, Sapporo, Tateno, Kagoshima, Naha. However, we have no access to the newest data from Hradec Hralove and Uccle.

In our analysis the aggregate time series, combined from deseasonalized total ozone monthly means at individual stations (the stations used are listed in Table 1), have been examined to detect the interannual variations of the atmospheric ozone. Deseasonalization was done by subtracting from monthly mean values the long-term monthly mean pertaining to the same calendar month. The method of constructing the aggregate time series is as follows:

$$O_{3i}(t_j) = \frac{1}{n} \sum_{m=1}^n \frac{O_{3im}(t_j) - O_{3im}^*(j)}{O_{3im}^*(j)}, \quad [1]$$

where $O_{3i}(t_j)$ is the deseasonalized monthly mean of total ozone in region i in month t_j , $O_{3im}(t_j)$ the monthly mean of total ozone in month t_j at station m belonging to region i , $O_{3im}^*(j)$ the long-term monthly means of total ozone in calendar month j at station m , n the number of stations in the region i , t_j the number of months since January 1957 ($t_j=1$ for January 1957).

In the tropic and north temperate regions, the aggregate time series have been constructed using provisionally revised data updated through December 1986 (these data have been taken from WMO, 1990) and the data published

by the World Ozone Data Center (WODC) for the period January 1987 through December 1990. The carefully revised data (reevaluation was made on a reading-by-reading basis) for the whole period analyzed — January 1957 through December 1990 (if possible) — have been considered for only a few stations.

In the north polar region, there are only a few Dobson stations having provisionally revised data. Therefore, the aggregate time series has been combined from provisionally revised and original data before December 1986 (see Table 1 for the names of stations). Moreover, three polar filter ozonometer stations (located in Russia) have been used to obtain the aggregate time series. It is well known that the quality of the filter ozonometer is poor. Since 1973 the modified version of the filter ozonometer has replaced the old version, but still the quality of this instrument is low compared to the Dobson spectrophotometer. However, since 1973 the filter ozonometers data have been considered to enlarge the data base.

It should be kept in mind that the composite time series derived by averaging various types of ozone data is only an approximation of real data. It seems that errors in the composite time series caused by the inhomogeneity of individual stations data, have been smoothed partially by the averaging procedure [1].

Global total ozone and the profile ozone data (from ozonesonde) are taken from the GEDEX CD-ROM. The NASA Earth Science and Applications Division initiated in 1991 the preparation of an extended data base for the global change research program called the Greenhouse Effect Detection Experiment (GEDEX). The data have been stored on CD-ROM.

The atmospheric ozone data for the GEDEX CD-ROM were prepared by J. K. Angell in the following way: seasonal total ozone deviations from long-term seasonal means at individual Dobson stations had been averaged to form seasonal deviations for the north polar, north temperate, tropical, south temperate, and south polar regions in the period 1958–1990 (for the names of the stations see Table 1 and Table 2 of Angell, 1989). The

Table 1 Ground-based ozone stations in the tropic region, north temperate and north polar regions, which have been used to form the combined time series of zonally averaged total ozone.

Station	Country	Latitude	Longitude	Data Start
TROPIC REGION				
1 Samoa	U.S.A.	14°15' S	170°34' W	1/76
2 Huancayo	Peru	12°03' S	75°19' W	2/64
3 Mauna Loa	U.S.A.	19°32' N	155°35' W	1/64
4 Naha	Japan	26°12' N	127°41' E	4/74
NORTH TEMPERATE REGION				
1 Lerwick	U.K.	60°08' N	1°11' W	1/57
2 Leningrad	Russia	59°58' N	30°18' E	8/68
3 Churchill	Canada	58°45' N	94°04' W	1/65
4 Edmonton	Canada	53°34' N	113°31' W	7/57
5 Belsk	Poland	51°50' N	20°47' E	3/63
6 Bracknell	U.K.	51°23' N	0°47' W	5/69
7 Uccle	Belgium	50°48' N	4°21' E	2/71
8 Hradec Kralove	Czechoslovakia	50°11' N	15°15' E	1/62
9 Hohenpeissenberg	Germany	47°48' N	11°11' E	1/67
10 Caribou	U.S.A.	46°52' N	68°01' W	6/62
11 Arosa	Switzerland	46°46' N	9°40' E	1/57
12 Bismark	U.S.A.	46°46' N	100°45' W	1/63
13 Toronto	Canada	43°47' N	79°28' W	1/60
14 Sapporo	Japan	43°03' N	141°20' E	2/58
15 Rome	Italy	42°05' N	12°13' E	1/57
16 Boulder	U.S.A.	40°01' N	105°15' W	1/64
17 Cagliari	Italy	39°15' N	9°03' E	1/57
18 Wallop Island	U.S.A.	37°15' N	75°31' W	1/70
19 Nashville	U.S.A.	36°15' N	86°34' W	1/63
20 Tateno	Japan	36°03' N	140°08' E	6/57
21 Kagoshima	Japan	31°38' N	130°36' E	2/58
22 Quetta	Pakistan	30°11' N	66°57' E	8/69
23 Cairo	Egypt	30°05' N	31°17' E	11/74
NORTH POLAR REGION				
1 Resolute	Canada	74°43' N	94°59' W	7/57
2 Dikson Island	Russia	73°39' N	80°14' E	7/57*
3 Barrow	U.S.A.	71°19' N	156°36' W	8/73
4 Tromso	Norway	69°39' N	18°57' E	7/57
5 Murmansk	Russia	68°58' N	33°03' E	10/61*
6 Reykjavik	Iceland	64°08' N	21°54' W	2/75**
7 Yakustk	Russia	62°55' N	129°45' E	9/61*
8 Lerwick	U.K.	60°08' N	1°11' W	1/57**
9 Leningrad	Russia	59°58' N	30°18' E	8/68**
10 Churchill	Canada	58°45' N	94°04' W	1/65**

* Filter ozonometer stations

** Dobson stations with re-evaluated data in the North Polar Region

Table 2 Trends in total ozone (year-round, winter, and summer trends) during the period 1970–1990 (in % per decade) for the north polar and north temperate regions, tropic region, and the world as a whole. The trend estimates have been derived by the multiple regression models Ia, Ib, IIa, IIb, (for models' description see section 4). The errors of the trend estimates (in parentheses) are at 2σ level.

Region	TREND IN TOTAL OZONE IN THE PERIOD 1970-1990 (% PER DECADE)											
	Model I						Model II					
	Version Ia			Version Ib			Version IIa			Version IIb		
	__Year__	__Winter__	__Summer__	__Year__	__Winter__	__Summer__	__Year__	__Winter__	__Summer__	__Year__	__Winter__	__Summer__
Tropics	-0.97(0.28)	-1.21(0.60)	-1.05(0.36)	-1.04(0.27)	-1.29(0.60)	-1.06(0.33)	-0.94(0.31)	-1.14(0.65)	-1.05(0.39)	-0.70(0.27)	-0.76(0.57)	-0.83(0.34)
North Temperate	-1.70(0.37)	-2.50(0.88)	-0.97(0.40)	-1.64(0.31)	-3.03(0.74)	-0.85(0.34)	-1.67(0.38)	-2.36(0.90)	-1.10(0.42)	-1.85(0.35)	-3.19(0.80)	-1.15(0.35)
North Polar	-2.57(0.68)	-3.16(1.60)	-1.90(0.71)	-2.46(0.56)	-3.57(1.34)	-1.94(0.58)	-3.02(0.70)	-3.61(1.63)	-2.33(0.74)	-2.56(0.62)	-3.67(1.43)	-1.77(0.61)
World	-1.77(0.31)	-1.76(0.56)	-1.56(0.52)	-1.50(0.28)	-1.53(0.54)	-1.33(0.47)	-2.00(0.31)	-2.00(0.58)	-1.77(0.54)	-2.00(0.35)	-2.01(0.66)	-1.77(0.57)

boundaries between the zones had been delineated by the Tropics of Cancer and Capricorn, and the Arctic and Antarctic Circles.

Seasonal deviations for the world in period 1958–1990 had been calculated by means of a 1, 3, 4, 3, and 1 weighting of north polar, north temperate, tropical, south temperate, and south polar climatic-zone deviations (according to relative areal extent of the zones), respectively. Because the original data (published by WODC) from many stations had been averaged together to prepare the time series of the global total ozone, it seems that this time series is not sensitive to the errors caused by recalibrations of Dobson and filter spectrophotometers. For more details of the data preparation, see Angell (1987, 1988).

The ozonesonde data (updated on GEDEX CD-ROM) have been examined to disclose the atmosphere layers which are responsible for trends in total ozone. Because of the influence of the El Chichon eruption in 1982 on ozone values calculated by the Umkehr method (e.g., DeLuisi, 1979; Reinsel *et al.*, 1984) the Umkehr ozone profiles are not examined here.

Seasonal ozone deviations (in the period 1968–1989) from the long-term seasonal means in 2–8 km, 8–16 km, 16–24 km, 24–32 km layers were stored on the CD-ROM by J. K. Angell for the regions: north polar, north temperate, south temperate, and south polar.

Until recently, almost all the ozonesonde stations were located in the north temperate zone. The GEDEX data are based on the results taken at: Wallops Island, Churchill, Edmonton, and Goose Bay in North America, Hohenpeissenberg, Lindenberg, Berlin, Lisbon, Payerne, Prague, Legionowo, and Uccle in Europe, Kagoshima, Sapporo, and Tateno in Japan. Some comments on the data handling can be found in Angell (1987, 1988). For other regions, the data are based on the results taken at one station only. Therefore, the representativeness of the data seems to be limited. However, we decided to examine the results of the sonde measurements taken at Resolute (75° N, 95° W) because of the growing interest of the ozone changes in the north polar region, which are expected to be influenced by the same ozone

destructive processes causing the Antarctic ozone hole.

3. Sources of the quasi-periodical variations in the atmospheric ozone

The interannual quasi-periodical variations in the atmospheric ozone are induced partially by the 11-year solar cycle, the QBO winds in the stratosphere over the equatorial region, and sea surface temperature (SST) fluctuations in the eastern equatorial Pacific. There are, of course, other fluctuations of ozone the sources of which are not yet fully recognized. Some of these fluctuations seem to be related to the variations in the temperature and chemical composition of the lower stratosphere.

The relation between total ozone and sunspot number has been discussed for about 30 years (for review, see Angell, 1989). During the 11-year cycle (from solar activity maximum to minimum) ozone declines about 1–3% (e.g., Oehlert, 1986; Angell, 1989; WMO, 1990). In Fig. 1a, we present 10.7cm solar radio flux (SRF) time series (used as a proxy index of the long-term solar activity) which has been considered in our trend models. In Fig. 1b, the energy spectrum of this series is shown. As it was expected, a distinct maximum at around 11 years has been found.

A near 2-year oscillation of the zonal wind in the tropical stratosphere was discovered in 1961 (Reed *et al.*, 1961; Ebdon and Veryard, 1961). A QBO induced variation in total ozone was reported in 1962 (Funk and Garnham, 1962). The results of studies since then (e.g., Angell and Korshover, 1973; Oltmans and London, 1982; WMO, 1990) indicate that the changes in total ozone (in the period between the east and west wind maxima) are a few percent of long-term mean ozone values and show latitudinal dependence.

The 50 hPa wind at Balboa (9° N, 80° W) has been chosen as a representative of the QBO wind in the tropical stratosphere. In Fig. 2a, the wind is shown for the period 1957–1990 and in Fig. 2b a spectrum analysis of the QBO wind at Balboa over the whole available data period (1951–1990) is presented. The dominant oscilla-

Fig. 1 Smoothed time series of 10.7 cm solar radio flux at Ottawa, 1957-1990 (Fig. 1a) and its power spectrum (Fig. 1b) taken from the whole period of the observations, 1948-1990. The time series is plotted as monthly means that have been smoothed by taking the 12-month running mean. The extremes in power spectrum are marked by the letters and the corresponding wave periods (in years). The power spectrum is shown in arbitrary units.

Fig. 2 Smoothed time series of 50 hPa Balboa wind, 1957-1990 (Fig. 2a) and its power spectrum (Fig. 2b) taken from the whole data period, 1951-1990. The time series is plotted as monthly means (in knots) that have been smoothed by taking 1-4-6-4-1 weighting. The extremes in power spectrum are marked by letters and the corresponding wave periods (in months). The power spectrum is shown in arbitrary units.

tion has been found at around 28 months (this period of the QBO is similar to that reported by Reed *et al.*, 1961). The secondary peaks (33 and 24 months) suggest that the QBO oscillations are not regular. Oltmans and London (1982) reported that during 1951–1978 the QBO period obtained from the wind measurements at Balboa varied from 22 to 35 months.

There has been evidence that relations between the 11-year solar cycle and stratospheric temperature (e.g., Labitzke, 1987) as well as between 11-year solar cycle and total ozone (Angell, 1989) change according to the phase of the QBO. For example, Labitzke (1987) reported that when equatorial stratospheric winds are westerly the low stratospheric temperature in the northern polar region is accompanied by the low level of long-term solar activity. Changes in the atmospheric dynamics, induced by the interaction between the QBO and the 11-year solar cycle (see, Kodera, 1991 and Kodera *et al.*, 1991 for a possible physical mechanism of the solar/QBO modulations) should influence ozone transport. Therefore, it was not a surprise that Angell (1989) found indications that the relation between total ozone and sunspot number may depend on the QBO phase. In section 4 we

present a new version of the multiple regression model, which accounts for the ozone changes due to interactions between the ozone forcing factors.

An impact of SST in the eastern equatorial Pacific on the atmospheric ozone has been widely discussed during the past few years. Statistical analyses show (Komhyr *et al.*, 1988 and 1991; Angell, 1990) that the total ozone variations at the South Pole appeared to be related to both QBO and SST. The ozone changes in the tropics, caused by both QBO and ENSO, have been discussed by Zerefos *et al.* (1992) and Shiotani (1992). The possibility of a large change in mid latitudinal total ozone due to strong warming in the eastern equatorial Pacific (like that during 1982/1983 ENSO event) was discussed by Bojkov (1987) and Zerefos *et al.* (1992).

In our analyses we use the Southern Oscillations Index—SOI (the normalized surface pressure difference between Tahiti and Darwin) to parameterize the ENSO event impact on the atmospheric ozone. The anticorrelation between eastern equatorial Pacific SST and SOI is well known (e.g., Quinn and Burt, 1972). In Fig. 3a, we present the SOI time series for the period 1957–1990, and in Fig. 3b the power

Fig. 3 Smoothed time series of Southern Oscillation Index, 1957–1990 (Fig. 3a) and its power spectrum (Fig. 3b) taken from the whole data period 1866–1990. The time series is plotted as monthly means that have been smoothed by taking the 12-month running mean. The extremes in power spectrum are marked by letters and the corresponding wave periods (in years). The power spectrum is shown in arbitrary units.

spectrum of the SOI oscillations obtained from all the available data (1866–1991). From Fig. 3b we can infer that the ENSO consists of various scale fluctuations, between two and 20 years, the nearly 4-year component being the most conspicuous among them.

All pictures presented in this chapter have been drawn using the GEDEX CD-ROM data. Spectral analyses of these data have been made using the maximum entropy method, for details of which see Ulrych and Bishop (1975).

4. The multiple regression models

The basic idea of the regression models presented in this paper is similar to that used by Reinsel *et al.* (1981), *i.e.*, variations in the atmospheric ozone are divided into parts to account for the overall mean ozone level, linear trend, long-term quasi periodical variations (with the periods above 1 year) and the noise (assumed to have autoregressive structure). Then, the following multiple regression model has been used to parameterize variations in the aggregate time series of the atmospheric ozone:

$$O_3(t) = \mu + \alpha * t + \beta * LTV(t) + Noise(t), \quad [2]$$

where $O_3(t)$ is the ozone amount in month t or in season t , t the number of months since the data beginning ($t=1$ at the beginning of data, $t=408$ at the end of the data constructed on a monthly basis), μ the overall mean level of ozone (changing with month or season), α the trend coefficients (depending on calendar month or season), β a collection of coefficients characterizing an impact of the interannual quasi-periodical variations in the atmosphere (assigned here as $LTV(t)$) on the atmospheric ozone, and $Noise(t)$ the noise term in month (or season) t (combined with the random noise and noise correlated with the noise term in months or seasons $t-1, t-2, \dots$).

In fact, the ozone data are divided into 12 groups (according to the calendar month) by model [2]. The groups are not treated separately during the calculation of the model's coefficients because of the assumed serial correlation in the ozone data (see construction of the

noise term of model [2]).

The deseasonalized monthly means of total ozone in the north polar, north temperate, and tropical regions show a weak tendency to be correlated for lags greater than 2 months. Therefore the noise term in [2] is modelled as a second order autoregressive process:

$$Noise(t) = \gamma_1 * Noise(t-1) + \gamma_2 * Noise(t-2) + R(t), \quad [3]$$

where γ_1 and γ_2 are autoregression coefficients (depending on calendar month), and $R(t)$ the residual part of the noise (which should represent a random noise with normal distribution and zero mean).

The ozonesonde and global total ozone data (updated on a seasonal basis) appeared to be correlated with the lag of one season. Therefore, the first order autoregression model has been used to parameterize the noise term ($\gamma_2=0$). All the model's coefficients ($\mu, \alpha, \beta, \gamma_1, \gamma_2$) have been calculated by the least squares method.

The trend model developed by the International Trends Panel (WMO, 1990) can be treated now as a basic model for ozone trend detection because of its use in many analyses. Therefore, we should point the differences of our model from the Trends Panel model. These are as follows:

- a) μ and α have been assumed to be different in the periods 1957–1969 (or 1958–1969 in the case of global total ozone) and 1970–1990 to account for the effects of atmospheric nuclear tests in the early 1960s. The ozone trends seem to be positive in the first period due to a recovery of the atmosphere from the effects of the atmospheric nuclear tests. The panel's model was based on the assumption that the trends had begun in 1970 (so called 'ramp trend model') and before 1970 the long-term changes in total ozone had been forced by nitrogen oxides coming to the atmosphere due to nuclear explosions. The panel's model used a proxy variable derived from 2-D photochemical model to account for the nitrogen oxides impact on total ozone during that period.

b)- first order autoregressive coefficients (γ_i) have seasonal structure (depending on month or season). It provides a possibility that in months, when a dynamic regime of the atmosphere ozone changes, the lagged autocorrelation coefficients in the ozone time series are smaller than in months belonging to the same regime. Autoregressive coefficients in the Trends Panel model had no seasonal structure.

c)- $\beta * LTV$ term has been modelled in the following ways:

Model Ia

$$\beta * LTV(t) = \sum_{i=1}^k \left\{ a_i * \cos\left(\frac{2\pi}{T_i} t\right) + b_i * \sin\left(\frac{2\pi}{T_i} t\right) \right\}, \quad [4]$$

where T_i are periods of the ozone fluctuations obtained from the spectral analyses (above 1 year), a_i and b_i coefficients to be determined by the least squares fit, and k the number of distinct peaks in the spectrum of the ozone fluctuations.

Model IIa

$$\beta * LTV(t) = \delta_1 * QBO(t) + \delta_2 * SOI(t) + \delta_3 * SUN(t), \quad [5]$$

where $QBO(t)$, $SOI(t)$, $SUN(t)$ represent time series of the 50 hPa zonal wind at Balboa, SOI, and 10.7 cm SRF, respectively (see Fig. 1a, Fig. 2a, Fig. 3a). The magnitudes of the QBO, ENSO, and the 11-year solar cycle impact on the atmospheric ozone (δ_i coefficients) have been determined statistically (by the least squares method). In the Trends Panel model, $\beta * LTV(t)$ consisted of the components describing the QBO and the solar effects in ozone (the ENSO effects were omitted).

d)- to describe interactions between the ozone forcing factors (*i.e.*, between waves in Model Ia, or between $QBO(t)$, $SOI(t)$, $SUN(t)$ in Model IIa) we modify the $\beta * LTV(t)$ term of

model [2] as follows:

Model Ib

$$\begin{aligned} \beta * LTV(t) = & \sum_{i=1}^k \left\{ a_i \cos\left(\frac{2\pi}{T_i} t\right) + b_i \sin\left(\frac{2\pi}{T_i} t\right) \right\} \\ & + \sum_{i=1}^{k-1} \sum_{j>i}^k \left\{ c_{ij} \cos\left(\frac{2\pi}{T_i} t\right) * \cos\left(\frac{2\pi}{T_j} t\right) \right. \\ & + d_{ij} \cos\left(\frac{2\pi}{T_i} t\right) * \sin\left(\frac{2\pi}{T_j} t\right) \\ & + e_{ij} \sin\left(\frac{2\pi}{T_i} t\right) * \cos\left(\frac{2\pi}{T_j} t\right) \\ & \left. + f_{ij} \sin\left(\frac{2\pi}{T_i} t\right) * \sin\left(\frac{2\pi}{T_j} t\right) \right\}, \quad [6] \end{aligned}$$

where c_{ij} , d_{ij} , e_{ij} , f_{ij} are interaction coefficients.

Model IIB

$$\begin{aligned} \beta * LTV(t) = & \delta_1 * QBO(t + \Delta_{QBO}) \\ & + \delta_2 * SOI(t + \Delta_{SOI}) \\ & + \delta_3 * SUN(t + \Delta_{SUN}) \\ & + \delta_{12} * QBO(t) * SOI(t) \\ & + \delta_{13} * QBO(t) * SUN(t) \\ & + \delta_{23} * SOI(t) * SUN(t), \quad [7] \end{aligned}$$

where δ_{12} , δ_{13} , δ_{23} are interaction coefficients, and Δ_{QBO} , Δ_{SOI} , Δ_{SUN} time lags relative to the forcing functions $QBO(t)$, $SOI(t)$, $SUN(t)$, respectively.

Model Ib differs from Model Ia not only because interaction terms are added to $\beta * LTV(t)$, but also periods T_i have been assumed to be unknown (*i.e.*, to be determined by the least squares method) at the first calculation step of Model Ib. Coefficients a_i , b_i , c_{ij} , d_{ij} , e_{ij} , f_{ij} are to be calculated together with T_i at the same step of the least squares procedure. Therefore, Model Ib becomes nonlinear. Assumption of a linear trend in the atmospheric ozone remains unchanged. Likewise, Model IIB is not linear, because the lags Δ_{QBO} , Δ_{SOI} , Δ_{SUN} will have to be calculated together with magnitudes δ_i . The nonlinear least squares fit proce-

ture, called *Levenberg-Marquardt* method has been used to find the model's constants (for details see Press *et al.*, 1990). To start this procedure, initial values of the model's constants should be selected. The 'first guess' coefficients have been obtained from the results of Model Ia (or Model IIa), while the other constants (*i.e.*, interaction coefficients and lags) are set to be 0. The results of Model Ib and IIb appear insensitive to the initial values of the coefficients. However, a calculation time of the model depends on the selected initial set of the coefficients rather heavily.

5. Trends in total ozone

Figs. 4a–4b present smoothed time series of total ozone in the following regions: north polar, north temperate, tropics, and the world as a whole. Figs. 5a–5b show the corresponding spectra calculated by the maximum entropy method. Long-term variations in total ozone can be inferred from the dashed lines in Figs. 4a–4d. These lines have been derived as a sum of sinusoidal waves (filtered out from total ozone time series by spectrum analysis) with periods above 12 years plus the linear trends. Figs. 4a–4d show basically (see dashed lines) the increase in total ozone before 1970 and decrease after 1970. This is in accordance with the assumption (used in Model Ia, Ib, IIa, IIb) of the different trends before and after 1970. In all regions, the ozone loss since 1970 has been found. Global total ozone data show a large ozone depletion since the late 1970s, probably as a result of the deep 'hole' over the Antarctic.

It should be noted that signs of a recovery of the global atmospheric ozone have been seen since 1986/1987. The reprocessed TOMS data (version 6) show also a global recovery of the average ozone amount since 1986 (Herman *et al.*, 1991). Comparing the patterns of the long-term variations in total ozone (dashed line in Fig. 4d) in the late 1980s and the early 1980s, we see that the recovery has been initialized after the sharp ozone loss in the 1982/1983 and 1985/1986. It is extremely difficult to say whether the global recovery really exists, because the large ozone drop between the late 1970s and 1985 was

formed partially due to deep dip in total ozone during the period 1982/1983 (result of eruption of El Chichon and a strong ENSO warm event) and 1985 (result of special atmosphere circulation, see Bojkov, 1987).

In Table 2, the results of trend analysis are presented. Statistically significant trends have been revealed in all analyzed regions and seasons during the period 1970–1990. The trends show latitudinal variations, *i.e.*, the rate of ozone loss increases with latitude.

Our trend estimates correspond closely to the pattern of ozone loss found by Stolarski *et al.* (1992). For example, they calculated the long-term trends, derived from ground-based total ozone data, for the band 26° – 64° N as $-1.8 \pm 0.4\%$ per decade over the whole year, $-2.7 \pm 0.6\%$ per decade in winter, and $-1.3 \pm 0.4\%$ per decade in summer.

We performed a trend analysis for the world and the north polar region using the original data published by WODC. Therefore, the data are not adjusted for the instruments' re-calibrations, and the results obtained for these regions should be accepted with caution. The trends for these regions were not calculated by Stolarski *et al.* (1992) because the revised total ozone data were not available. However, we think that the results of the trend analysis for these regions have been affected by the data inhomogeneities only slightly. The errors related to the changes of the instrument's calibration level have been smoothed partially by the averaging procedure used to form the aggregate time series.

It should be noted that the trends in global ozone show weaker seasonal variations compared to those over the north temperate and north polar regions. This implies the possibility of the trend variations on a hemispherical scale. The relatively small trends in the summer/autumn season in the northern hemisphere (NH) have met a large ozone loss in the southern hemisphere (SH) during winter/spring in SH. We have found that the global total ozone loss reaches a maximum in NH autumn (September through November), *i.e.*, $-2.28 \pm 0.68\%$ per decade (Model Ia) or $-2.16 \pm 0.66\%$ per decade (Model IIa) probably due to the

Fig. 4 Variations of monthly total ozone percentage deviations from the long-term monthly means (smoothed by taking the 12-month running mean) for the north polar (Fig. 4a), north temperate (Fig. 4b), the tropic region (Fig. 4c), and the world as a whole (Fig. 4d). The dashed lines represent the long-term variations in total ozone derived as a sum of the trend estimates and the Fourier components with periods greater than 12 years.

Fig. 5 Power spectra of the time series presented in Fig. 4. Figs. 5a, 5b, 5c, 5d correspond to Figs. 4a, 4b, 4c, 4d, respectively. The peaks in spectra are marked by letters and values of wave period (in months).

appearance of the Antarctic Ozone Hole. Unfortunately, the limited number of Dobson stations in SH makes the trend analysis of zonally averaged total ozone unreliable there.

The various trend models presented in this paper provide almost the same trend estimates. The differences between the trends are not statistically significant at 2σ level. Then, in the case when only a trend estimate of the atmospheric total ozone is under consideration, it seems useless to focus on details of the interactions between the ozone forcing factors (as it has been done in Models Ib and IIb).

6. Total ozone changes induced by the QBO, ENSO, and long-term solar activity

The spectrum analysis of total ozone (Figs. 5a-5d) shows peaks in the spectra correspond-

ing roughly to those of the spectra of the QBO, ENSO, and 10.7 cm SRF. The QBO variations in total ozone are present in all analyzed regions, but unexpected strong variations with periods of 20-23 months are also found. The peaks in total ozone spectra corresponding to the strongest peaks in the ENSO and in 10.7 cm SRF spectra are slightly shifted. Therefore, this suggests the possibility that the changes in total ozone forced by the QBO, ENSO, and the 11-year solar cycle are modulated partially by interactions between the ozone forcing factors. Model Ib and IIb provide a tool to examine how the interdependence between the ozone forcing factors manifests in the atmospheric ozone (for further discussion see section 7). Table 3 shows the magnitudes of ozone changes induced by the QBO, ENSO, and 10.7 cm SRF (coefficients δ_i derived from Model IIa).

Table 3 The magnitudes of the total ozone changes induced by the QBO, ENSO, and the long-term solar activity variations (coefficients δ_i of Model IIa) in the tropic region, north temperate and north polar regions, as well as for the world as a whole. The units of the QBO, ENSO, and solar coefficients are %/knot, %/(1 SOI unit), %/($10^{-23} \text{ Js}^{-1} \text{ m}^{-2} \text{ Hz}^{-1}$), respectively. Estimated 2σ limits are shown in parentheses.

REGION	Magnitudes of the ozone changes		
	QBO	SOI	SUN
TROPICS	0.035 (0.010)	0.569(0.183)	0.010(0.004)
NORTH TEMPERATE	-0.027 (0.010)	-0.136(0.194)	0.007(0.003)
NORTH POLAR	-0.057 (0.018)	-0.320(0.363)	0.003(0.006)
WORLD	0.001 (0.010)	-0.008(0.190)	0.015(0.003)

6.1. The QBO effects in total ozone

To calculate the ozone changes between east and west phases of the QBO, coefficient δ_1 should have been multiplied by 50. This is the average change in speed (in knots) of the QBO wind (measured at Balboa) between the west (westerly wind at 50hPa over the equatorial region) and the east phase of the QBO.

The zonally averaged total ozone variations induced by the QBO are statistically significant. The largest manifestation of the QBO effects in total ozone has been found in the north polar region, *i.e.*, about 3% increases between west and east phase. In the region 30° – 60°N and the tropics the amplitude of the QBO is smaller, *i.e.*, about 1.8% decrease in the tropics and 1.3% increase in the north temperate region between the west and the east phase. These estimates agree with the QBO changes in total ozone recently calculated for: the north polar, north temperate, and tropics regions by Angell (1989). The QBO effects in global total ozone (calculated by Model IIa) are much smaller than in the zonally averaged total ozone, *i.e.*, the change between west and east phase of the QBO is found to be not larger than 0.5% (this estimation is within the range of the 2σ error, and the magnitude itself is not statistically significant). However, both Models Ia and

Ib reveal statistically significant (at 2σ level) wave in global total ozone with a 28-month period and an amplitude of 0.4%.

6.2. The ENSO effects in total ozone

The ENSO effects in total ozone, calculated by Model IIa, have been found statistically significant in the tropics only. Using the value presented in Table 3, the scale of ozone decreases in the tropics between the change from cold to warm phase of the Southern Oscillation, which took place in the period 1982/1983 (very strong the ENSO warm event), can be estimated as 1.5% of the long-term mean of equatorial total ozone. This is approximately equal to the depletion in equatorial total ozone due to the ENSO found by Shiotani (1992).

For the north temperate region, a spectrum analysis of the aggregate time series of total ozone has shown a distinct peak at 43 month that corresponds to the largest peak of SOI spectrum exactly. The amplitude of 43 month wave in total ozone, as calculated by Models Ia and Ib, is about 0.8% (statistically significant at 2σ level). However, Model IIa has not been able to detect the ENSO variations in total ozone in this region. The following explanations seem to be possible:

- almost 4 year disturbances in the atmospheric dynamics over the north temperate regions are not related to the ENSO,
- changes in total ozone depend on the strength of the warm (or cold) phase of ENSO, so weak or moderate ENSO may influence total ozone in a different way than the strong one,
- the ENSO impact on the atmospheric ozone is modified by both the QBO and the long-term solar activity (or other unknown yet factors).

We show that Model IIb has provided opportunities to discuss the last two of the above-mentioned options. For further discussion, see section 7.

In the extratropical regions (north polar, north temperate) and the globe as a whole, the ENSO induced disturbances in total ozone have not been found highly statistically significant (at 2σ level). However, in the north polar and

north temperate regions, the ENSO induced disturbances have been found statistically significant at 1σ level.

In the global total ozone data, the ENSO related variations have not been revealed. Because the ENSO variations in tropical total ozone have been found highly statistically significant, lack of the ENSO signal in the global total ozone data implies that the phase of the ENSO induced disturbances in the extratropical total ozone is probably opposite to that in tropical total ozone, *i.e.*, an overall anticorrelation exists between SOI and ozone there. This relation is not highly statistically significant because in-phase SOI-ozone relation seems to exist during a very strong ENSO warm event (for more details see section 7).

6.3. The solar effects in total ozone

Long-term changes in solar activity are manifested in global total ozone as well as in the zonally averaged data for the north temperate and the tropic regions. To obtain the change in total ozone between the extremes of the long-term sun activity, coefficients δ_3 (Table 3) should be multiplied by 100 (or 150 in the case when the solar activity maximum is especially high as in 1957/1958). Table 3 shows that an increase in solar activity between the minimum and maximum of solar activity is accompanied by an increase of 1.5–2% in global total ozone. Slightly lower values have been found for the tropic region (1–1.5%) and for the north temperate region (0.7–1%). The magnitudes of the total ozone deviations (significant at 2σ level) induced by 10.7 cm SRF are similar to those reported by other authors (e.g., Angell, 1989). Surprisingly, changes in total ozone over the north polar region (obtained from Model IIa) have not been found related to the 11-year solar cycle probably due to large natural ozone variations masking the relation between total ozone and solar activity.

Likewise, difficulty to find a correlation between sunspot number and total ozone values (the seasonal means of ozone and the 3-month averages of the sunspot number were compared) over the north polar region was reported earlier by Angell (1989). However, we will able

to show a strong correlation between total ozone and solar activity there using Model IIb. It should be noted here, that the calculated changes in total ozone are associated with long-term changes in UV radiation flux reaching the upper atmosphere, and other components of solar variability (like particle fluxes) have not been parameterized by Model IIa and Model IIb.

6.4. Time lag between the forcing factors and their response in total ozone

To describe the relation between the total ozone variations and a forcing factor, both the magnitude of the induced total ozone variations and a phase shift (time-lag relative to forcing factor) should be considered. Model IIb provides this possibility and the time-lag is calculated precisely as one of the coefficients estimated by the least squares fit. In the past ozone trend models, the time-lag was assumed to be known as an input value of the trend model (e.g., the Trends Panel model, WMO, 1990) and probably it had a little influence on the estimated trends. However, it seems that the precision of the time-lag estimate becomes more important when the short-term changes in total ozone are under consideration. Table 4 shows the time-lags (in months) between the total ozone variations and the corresponding forcing factors. For a better comparison with the earlier findings, the time-lag of the QBO disturbances in total ozone is calculated as a time difference between the maximum of the QBO wind and the maximum in the QBO induced ozone. For other forcing factors, the time-lag refers to the time differences between the ozone forcing factors and its maximum response (positive or negative) in total ozone. The negative value of time-lag means that the induced variations in total ozone follow the forcing factor.

For the tropic and the north temperate regions, the results presented in Table 4 could be compared with the recent results obtained by Zerefos *et al.* (1992). Using the regression formulas (the regression formulas, *i.e.*, equation (1) and (2) of Zerefos *et al.* (1992), enable us to calculate the QBO phase in total ozone at a

Table 4 Time lags (in months) between the induced total ozone variations (by the QBO, ENSO, and long-term solar activity) and ozone forcing factors for the tropic region, north polar and north temperate regions, as well as for the world. The negative lag means that the induced total ozone variations follow the forcing factor. These estimations have been derived by Model IIb and the errors of the estimates (in parentheses) are at 2σ level.

REGION	TIME LAG: TOTAL OZONE/FORCING FACTORS (in months)		
	QBO	SOI	SUN
TROPICS	-4 (2)	-5 (4)	-5 (4)
NORTH TEMPERATE	-11 (2)	-1 (1)	-8 (8)
NORTH POLAR	-14 (1)	1 (1)	9 (6)
WORLD	-	-	0 (2)

given latitude) and locations of four Dobson stations in the tropics (see Table 1 for the names of these stations) we estimate the average time-lag of the QBO in the tropical total ozone as about -3 months, which is close to our estimate by Model IIb. Likewise, using the same procedure we estimate that the time-lag of the QBO in total ozone over the north temperate region as -10 months, which is somewhat lower than our estimates based on Model IIb (*i.e.*, -11 months).

At high latitudes (the north polar region) the time lag is about -14 month. Comparing the values of the QBO time-lag in the tropic, north temperate, and north polar regions we can confirm the value of the phase progression of the QBO signal in total ozone found by Zerefos *et al.* (1992) (about 2 months per 10 degrees of latitude).

The phase of the QBO and ENSO in global total ozone are not presented in Table 4 because the magnitudes of these variations in total ozone have been found very small (not statistically significant even at 1σ level).

The time-lag of ENSO in total ozone at low latitudes has been found to be about a half year. The ENSO in total ozone at mid and high latitudes seems to be out of phase with the SOI

because the magnitude of the ENSO variations in total ozone is negative there (see, Table 3). For the extratropical regions the time lag of ENSO cannot be calculated precisely because the magnitude of the ENSO variations in total ozone is not statistically significant at 2σ level. It should be noted here that, in the past, estimates of phase of the ENSO variations in total ozone have not been done by careful statistical methods. Shiotani (1992) and Zerefos *et al.* (1992) suggested that a maximum sign of the ENSO in total ozone at low latitudes appears after a few months after a maximum phase of an ENSO event (defined as a minimum value of SOI).

The 11-year variations in total ozone are nearly in phase with the long-term changes of 10.7 cm SRF. This finding was reported by many authors (*e.g.*, Angell and Korshover, 1976; Angell, 1989; WMO, 1990). However, we have found a small phase shift between the solar induced variations in total ozone and the 10.7 cm SRF, which seems to have latitudinal dependence. In the tropic region, the 11-year variations in total ozone follow the forcing factor (the time lag is about 2 seasons). In northern mid latitudes, there is possibility of the zero lag. In the north polar region, the solar induced variations in total ozone precede the long-term variations in the 10.7 cm SRF (the time lag is about 3 seasons). The solar induced variations in global total ozone have been found exactly in phase with the long-term variations in the 10.7 cm SRF. Therefore, the zonal differences in the phase shift probably cancel out in the global total ozone data. Our findings should be treated with caution, because there is a likelihood that the latitudinal changes in the phase shift (between the solar induced variations in total ozone and the long-term solar activity) may be generated erroneously, *i.e.*, due to extremely low ozone values in the early 1960s, which appeared a few years before the minimum in the solar activity due to nuclear bomb tests, and the long-lasting (about 2 years) protracted maximum in solar activity during 1968/1969 without a sharp maximum.

7. Total ozone changes due to the interactions between the forcing factors

Recently discovered interdependence between solar activity and the tropospheric-lower stratospheric circulation according to the phase of the equatorial QBO (Labitzke, 1987; Labitzke and van Loon, 1988, 1989) implies a possible relation between atmospheric ozone and solar activity depending on the phase of the QBO. Recently, global changes in the atmospheric dynamics induced both by the phase of the QBO (west, east) and the phase of the long-term solar activity (low or high solar activity) have been discussed (Kodera, 1991). Angell (1989) reported that in the tropics most of the relation between sunspot number and total ozone was associated with the east QBO winds, whereas in the north temperate zone the relation was statistically significant only during the period of the west QBO winds.

On the basis of the recent findings, we think it is reasonable to include the solar/QBO interaction term into the ozone trend model. For the other interaction terms (QBO/ENSO, and ENSO/solar) we have no support from the atmospheric circulation studies. The specifics of the atmospheric circulation related to the QBO/ENSO and solar/ENSO interactions have not yet been discussed. However, the possibility of associations among solar flux, QBO, ENSO, and the climate changes has been pointed by Barnston and Livezey (1991).

Model IIb provides the possibility of gaining more details about the relation between year-to-year changes in the atmospheric ozone and the ozone forcing factors. In this model, the changes in total ozone induced directly by the forcing factors (zonal wind at 50 hPa in the equatorial region, SOI, 10.7 cm SRF) as well as the changes related to interactions between the factors (*i.e.*, these changes in the atmospheric ozone are assumed to depend on products between the ozone forcing factors) are parameterized taking the QBO phase into account. This means that the coefficients δ_i and δ_{ij} are grouped according to the east and the west phase of the QBO. Table 5 shows the magnitudes of changes in total ozone (derived from Model IIb) forced by the QBO, ENSO, and the long-term solar activity. Model IIb allows the magnitudes to be different in the east and the west phase of the QBO.

The long-term solar induced variations in total ozone have been found statistically significant in almost all the regions for both phases of the QBO. The assumption that the ozone forcing factors depend on the QBO phase enables us to find the solar induced variations in total ozone at high latitudes (the north polar region), *i.e.*, about 2-3% ozone drop between the maximum and the minimum of solar activity.

Comparing the magnitudes of the solar induced variations in total ozone during the west and the east phase of the QBO, the largest

Table 5 As in Table 3 except that the results have been obtained by Model IIb, in which the magnitudes of the changes in total ozone, related to the forcing factors (the QBO, ENSO, solar cycle), have been grouped according to the QBO phase.

REGION	MAGNITUDES OF THE TOTAL OZONE CHANGES					
	Q B O		E N S O		S U N	
	WEST	EAST	WEST	EAST	WEST	EAST
TROPICS	0.045(0.038)	0.025(0.010)	-0.243(.584)	0.446(.327)	0.010(0.006)	0.016(0.004)
NORTH TEMPERATE	-0.025(0.020)	-0.028(0.013)	0.125(.590)	-1.462(.803)	-0.004(0.005)	0.008(0.004)
NORTH POLAR	-0.310(0.206)	-0.126(0.039)	-0.207(1.67)	-1.709(1.47)	0.018(0.010)	0.017(0.006)
WORLD	-0.001(0.032)	0.010(0.020)	0.232(.939)	-0.225(.447)	0.016(0.008)	0.015(0.005)

difference has been revealed in the north temperate region. In this region, the solar induced variations in total ozone are manifest during the east phase of the QBO, whereas in the west phase of the QBO the ozone-sun relation is not statistically significant. In other regions, the differences between the magnitudes of the solar induced variations during the east and the west phase of the QBO are too small to decide on the solar impact on total ozone in accordance with the QBO phase. However, it should be noted that the QBO phase has especially weak influence on the global total ozone (see almost the same magnitudes of the solar induced variations in the global total ozone in both phases of the QBO). The results presented in Table 5 suggest the possibility that the long-term solar impact on total ozone depends on latitude, *i.e.*, the highest is in the north polar region, the lowest at northern mid latitudes.

In the northern hemisphere, the relation between the long-term solar activity and total ozone is more manifest in the east phase of the QBO. It seems that in the southern hemisphere, most of the relation should be associated with the west phase of the QBO, because the solar induced variations in the global total ozone do not show dependence on the QBO phase.

The QBO effects have been disclosed in the zonally averaged total ozone during both phases of the QBO. In the north temperate region, the magnitude of the QBO forced variations in total ozone calculated at the westerly wind maxima (the wind speed about 20 knots) seems to be about half of that at the easterly wind maxima (the wind speed about 40 knots). Almost the same magnitudes of the QBO disturbances in total ozone at peaks of the equatorial 50 hPa zonal wind have been disclosed over the tropic and north polar regions.

Table 5 shows the possibility of large QBO variations in total ozone over the north polar region, *i.e.*, about 5% increase and decrease at the peak during the east and the west phase of the QBO, respectively. Closely similar values for the QBO induced disturbances in total ozone over the Arctic (*i.e.*, during winter, 8% increase in the east phase and 7% decrease in the west phase) were reported by Bojkov (1988) from the

revised total ozone data.

The ENSO induced variations in total ozone have been revealed in zonally averaged data during the east phase of the QBO. In the tropics, the ENSO in total ozone and SOI are in phase, whereas in the north temperate and polar regions an out-of phase relation is found.

A strong correlation between total ozone at low latitudes and ENSO has been recently reported (Shiotani, 1992; Zerefos *et al.*, 1992). At higher latitudes, there is suggestion (Bojkov, 1987) of a possible in phase relation between SOI and total ozone following only the very large ENSO events similar to that in 1982–1983. During the rest of the period an anticorrelation exists, which is statistically insignificant (this has been confirmed also by the results derived from Model IIa, see Table 3).

Bojkov (1987), using ozonesonde data in Europe, suggested that SOI-ozone relation depends on the QBO, *i.e.*, during the periods when the QBO and SOI were in phase (e.g., the 1982/1983 ENSO event) the ozone deficiency period was especially prolonged in the lower stratosphere. Therefore, it seems that to explain the relation ENSO-total ozone in the extratropical regions interactions between the ozone forcing factors should be considered.

Figs. 6a, 7a, 8a show the 'natural' variations (sum of the QBO, ENSO, solar induced variations in total ozone) derived from Model IIa (the ozone changes do not depend on interactions between the ozone forcing factors) and from Model IIb (the interactions between the factors are included). In addition, Table 6 shows the interaction coefficients between the ozone forcing factors grouped according to the east and the west phase of the QBO. To estimate the quality of fit by Model IIb, in Figs. 6b, 7b, 8b we present the time series of the smoothed (by the 12-month running mean) model's noise, *i.e.*, $Noise(t)$ term of [2]. These time series represent the interannual variations in total ozone not explained by the QBO, ENSO, and the solar forcing. The smoothing procedure, applied to $Noise(t)$ term, enables us to partially suppress the random noise (a part of $Noise(t)$, see formula [3]). Then, the time series presented in Figs. 6b, 7b, and 8b reveal the

interannual variations in total ozone induced by other unknown factors probably related to the changes in the atmospheric dynamics.

The statistically significant (at 2σ level) interaction coefficient QBO-ENSO has been revealed in the tropic region during the west

phase of the QBO (see Table 6). However, the differences between Model IIa and IIb (see Fig. 6a) have been found quite small; for example during the 1982/1983 ENSO the 'natural' variations in total ozone by Model IIb have appeared to be only 0.5% lower than those calculated

Fig. 6 Time series of the 'natural' variations of the tropical total ozone (sum of the variations induced by the QBO, ENSO, and solar activity cycle, Fig. 6a) and time series of the 'natural' variations of total ozone not explained by the multiple regression models (*i.e.*, the model's noise, Fig. 6b). The dashed curve (Fig. 6a) and continuous curves (Figs. 6a and 6b) represent the results of Model IIa (model without the interaction terms) and Model IIb (model with the interaction terms), respectively. The curves are plotted as monthly means that have been smoothed by the 12-month running means. The straight lines (bold continuous in Fig. 6a, and dashed in Fig. 6b) represent the linear best fit for the period 1970-1990.

Fig. 7 The same as Fig. 6 except that the results are for the north temperate region.

Fig. 8 The same as Fig. 6 except that the results are for the north polar region.

Table 6 The magnitudes of the changes in total ozone (grouped according to the QBO phase) induced by the QBO/ENSO, QBO/solar, and ENSO/solar interactions for the tropic region, north temperate and north polar regions, as well as for the world. The units for the QBO/ENSO, QBO/solar, ENSO/solar interaction coefficients (see columns of this table) are $\% / \{\text{knot} \cdot (1 \text{ SOI unit})\}$, $\% / \{\text{knot} \cdot (10^{-23} \text{ Js}^{-1} \text{ m}^{-2} \text{ Hz}^{-1})\}$, $\% / \{(1 \text{ SOI unit}) \cdot (10^{-23} \text{ Js}^{-1} \text{ m}^{-2} \text{ Hz}^{-1})\}$, respectively. The results have been obtained by Model IIb and estimated 2σ limits are shown in parentheses. The statistically significant results are underlined.

REGION	MAGNITUDES OF THE TOTAL OZONE CHANGES					
	QBO/ENSO		QBO/SUN		ENSO/SUN	
	WEST	EAST	WEST	EAST	WEST	EAST
TROPICS	<u>0.138</u> (0.058)	-0.002(0.014)	-0.003(.0006)	<u>.0002</u> (.0001)	-0.0059(.0069)	.0018(.0041)
NORTH TEMPERATE	-0.008(0.067)	-0.011(0.017)	<u>.0008</u> (.0004)	.0001(.0002)	<u>.0094</u> (.0062)	<u>.0138</u> (.0064)
NORTH POLAR	-0.039(0.130)	-0.008(0.031)	.0014(.0017)	<u>.0007</u> (.0003)	.0057(.0131)	<u>.0185</u> (.0114)
WORLD	0.001(0.007)	0.004(0.019)	.0001(.0006)	-.0000(.0002)	.0006(.0089)	.0010(.0060)

from Model IIa. Fig. 6b shows that Model IIb explained almost all the variations of total ozone during the 1982/1983 ENSO (reported as very strong, see Fig. 3a).

In the tropic region, the QBO-solar interaction has been revealed statistically significant during the east phase of QBO. If the easterly wind maximum meets a maximum of solar activity, this interaction will result in about 1% loss in total ozone. In northern mid latitudes,

the QBO-solar and ENSO-solar interactions during the west phase of QBO and the ENSO-solar interaction during the east phase have been found statistically significant. In the north polar region, ENSO-solar and QBO-solar interactions have been revealed during the east phase of the QBO. Fig. 7a shows that the interactions between the ozone forcing factors could yield an additional ozone decrease of about 1% in the north temperate region during the 1982/

1983 ENSO. Fig. 7b shows that during that period the ozone amount was about 1% lower than that predicted by Model I Ib.

The large ozone decline over the extratropical region during the period 1982–1983 cannot be attributed to the west phase of the QBO and the strong ENSO warm event only. The eruption of El Chichon in Mexico in April 1982 seems to have had an impact on total ozone at northern hemisphere latitudes in winter 1982/1983 and spring 1983 (e.g., Bojkov, 1987). The manifestation of the large volcanic eruption in total ozone was discussed extensively during the last decade (for review, see Chandra and Stolarski, 1991). We estimate the El Chichon impact on total ozone at northern mid and high latitudes was quite small (see the ozone deficit of up to 1% in 1982/1983 in Figs. 7b and 8b). Our result agrees with the findings of Bojkov (1987) and Chandra and Stolarski (1991). However, there have been several papers that indicated the large impact of the volcanic eruption on total ozone. For example, Pitari *et al.* (1991) reported about 5% decline in total ozone during winter 1982/1983 and spring 1983 over the north temperate region (based on a 2D model) due to heterogeneous reactions on both background and volcanic aerosols.

In sections 6 and 7 we discussed the impact of the QBO, ENSO, and solar activity on the total ozone amount. However, there are, of course, several total ozone deficiencies which are not related to these factors. The fluctuations in the temperature and chemical composition of the lower stratosphere seem to force changes in total ozone also and disturb the relations between the quasi-periodical interannual variations in total ozone and the forcing factors.

8. The 'natural' trends in total ozone

In the previous sections, we examine long-term variations (trends) and short-term variations (with a time scale up to about one decade) in the atmospheric ozone. The interannual changes in the atmospheric ozone have been estimated from the multiple regression models. Two families of the multiple regression models

have been used. They have been selected according to the method of parameterization of the quasi-periodical variations in the atmospheric ozone. The main differences between the two families of the models are as follows:

- in the first family, the ozone forcing factors are not defined explicitly (the spectrum analysis reveals the dominant waves in atmospheric ozone), but their effects on total ozone are expressed as a sum of sinusoids,
- in the second family, the ozone forcing factors are selected a priori and the induced changes in the atmospheric ozone must follow the time variations in the forcing factor (a time-lag between the forcing factor and the induced variations in ozone is calculated by Model I Ib).

Therefore, according to the second family of the models, the longest time scale of the induced quasi-periodical disturbances in the atmospheric ozone is probably about 11 years. We have found periodical disturbances with a time scale larger than 11 years in the forcing factors (SOI and 10.7 cm SRF). However, it seems that their impact on the atmospheric ozone is rather small.

The intriguing question of a scale of the 'natural' trends in total ozone is discussed in this section. The 'natural' trend means linear changes in the atmospheric ozone due to the long-term changes (with a time scale larger than a decade) in the ozone forcing factors not related to human activity. We have focused mainly on the variations in the atmospheric ozone forced by the QBO, ENSO, and the long-term changes in solar activity. Figs. 6a, 7a, and 8a show time series comprising all the natural variations in total ozone parameterized by Model I Ia and I Ib, *i.e.*, these time series have been constructed taking into account all terms of the basic model [2] excluding the trends and noise term. The straight lines fitted to the modelled 'natural' variations in the period 1970–1990 (bold lines) express a scale of long-term changes in the ozone due to the long-term changes in the ozone forcing factors (for example, see Fig. 1 for the positive trend in the solar activity in the period 1970–1990, which

influences the positive trend in total ozone; this trend is not accounted for by term α of Model [2]).

The rate of ozone changes calculated from the slope of the line has been found quite small (about 0.1% per decade decrease in the tropics and north polar region, and about 0.1% increase in the north temperate region). However, there are other important factors having a large impact on the ozone variations (e.g., temperature, chemical composition of the lower stratosphere). In Figs. 6b, 7b, and 8b the interannual changes in the total ozone not explained by Model IIb are presented. These changes have been calculated from the smoothed (by the 12-month running mean) noise of the model and they may not be necessarily trendless. The least squares procedure is based on the assumption that only the trend in $R(t)$ term of Model [2] (see expression [3]) is equal to zero because it represents the random residuals from the trend model.

The changes of total ozone are strongly influenced by variations of the ozone amount in the lower stratosphere (the 16–26 km layer, where the partial pressure of ozone reaches its maximum) where dynamical processes govern the ozone distribution. Therefore, it is possible that the total ozone changes, seen in Figs. 6b, 7b, and 8b, are a result of the changes in the atmospheric dynamics not related to the QBO, ENSO, and the solar induced changes in the dynamics. However, the long-term changes in the smoothed model's noise (see dashed lines in Figs. 6b, 7b, and 8b) have been found quite small (of the order 0.1% per decade, not 2σ statistically significant) compared to the trend estimates (presented in Table 2).

There is a possibility that the trends calculated in section 5 are partially the result of secular changes (with a time scale comparable to or larger than the analyzed data period) in the atmospheric dynamics and the chemical composition of the atmosphere. These effects in the atmospheric ozone are difficult to delineate because variations in the atmospheric ozone induced by secular changes in the atmosphere may be superimposed on the anthropogenic effects in total ozone (related to the man-made

changes in the chemical composition of the atmosphere). However, the spectrum analysis reveals only waves in the atmospheric ozone corresponding roughly to the QBO, ENSO, and the long-term solar forcing. In the spectra (see Figs. 5a, 5b, 5c, 5d), there are no signs of the variations with a time scale comparable with the data period or larger (the maximum entropy method of spectrum analysis provides the possibility of revealing such long-term wavy disturbances). Therefore, it seems that during the last two decades the impact of the secular changes in the atmosphere on trends in total ozone is limited. Then, the changes in the chemical composition of the atmosphere (mostly caused by human activities, e.g., increasing contamination of the atmosphere by the CFCs) are probably a main source of the observed trends in the atmospheric ozone.

Komhyr *et al.* (1991) suggested that the downward trend in ozone since the early 1970s may have been at least partially induced by the changes in the strength of the maximum easterly wind at 50 hPa over the equatorial region. They found that the maximum speed of the easterlies decreased with time at the rate of 0.82% per year during 1962–1975, and in the period 1976–1988 increased at the rate of 0.36% per year. In contrast, during 1962–1988, the maximum westerly wind speed remained relatively constant. Using the rate reported by Komhyr *et al.* (1991) and the results of Model IIb (Table 5) we estimate that the increase in the maximum speed of the easterlies forced even an increase in total ozone over the north polar region at a rate not higher than 0.2% per decade. This region was chosen because the magnitude of the QBO in total ozone is largest at northern high latitudes. The trend induced by the long-term changes in the equatorial stratosphere is quite small compared to the observed trends. Therefore, it seems that long-term changes in the tropic atmosphere dynamics may make only a small contribution to the observed trends.

9. Interannual changes in layer-mean ozone in the north polar and north temperate region

All results presented in this section are based on the layer-mean ozone seasonal means updated on the GEDEX CD-ROM. Figs. 9a, 10a, 11a, 12a show the ozone content in 2–8 km, 8–16 km, 16–24 km, 24–32 km layers in the north temperate region during the period 1968–1989, respectively. To reveal long-term changes in the layer-mean total ozone we construct time series (see dashed lines in Figs. 9a–12a) combining the trend estimates and long-term Fourier components (components with periods longer

than 12 years).

An increase in tropospheric ozone (layer-mean ozone in 2–8 km) and a decrease in the ozone amounts in the lower stratosphere (16–24 km) can be inferred directly from Figs. 9a and 11a without the help of any trend model. Comparing Figs. 9a–12a with Fig. 4b we see that the layer-mean ozone time series show individual characteristics, *i.e.*, do not necessarily follow the shape of the total ozone time series. For example, the large drop in total ozone in 1985 is not seen in the 16–24 km layer (but seen in the 24–32 km layer), the large ozone decline in 1982/1983 is not observed in layers 2–8 km and 24–32 km, *etc.*

Fig. 9 Variations of seasonal layer-mean ozone percentage deviations from the long-term seasonal means for the north temperate region in the 2–8 km layer (continuous curve in Fig. 9a) and the corresponding power spectrum (continuous line in Fig. 9b). The dashed curve (Fig. 9a) shows the long-term variations in time series, calculated in the same way as those in Fig. 4. Statistically significant peaks of the spectrum (at 2σ level) are marked by letters and wave period (in years).

Fig. 10 The same as Fig. 9 except that the results are for the 8–16 km layer.

Fig. 11 The same as Fig. 9 except that the results are for the 16-24 km layer.

Fig. 12 The same as Fig. 9 except that the results are for the 24-32 km layer.

The spectra of layer-mean ozone are presented in Figs. 9b-12b. Peaks in the spectra in layers 8-16 km and 24-32 km correspond roughly to the QBO and ENSO periodicities. In the layer 2-8 km, only the solar induced variations in the layer-mean ozone seem to exist. Table 7 shows the magnitudes (based on Model IIa) of the changes in layer-mean ozone at the northern mid latitudes induced by the forcing factors. The QBO related changes are statistically significant in the upper troposphere and lower stratosphere (8-16 km and 16-24 km layers). Surprisingly, the solar induced variations have been revealed in the tropospheric layer only.

Angell (1989) found that the correlation between sunspot number and sonde-derived ozone in the northern temperate region is statistically insignificant in all layers, even in 16-24 km and 24-32 km layers (these layers contain

70% of the atmospheric total ozone). The reason why the solar induced variations could not be revealed from the sonde-derived ozone is probably the following, the layer-mean ozone is more variable than the total ozone amount due to forcing (depending on altitude) caused both by dynamics and chemical composition of the atmosphere (therefore, errors of the statistical estimates are large).

Table 8 shows the results of the trend analyses by Models Ia and IIa for the north temperate region, and north polar region (data from Resolute station only). Models Ib and IIb provided almost similar results and the interaction coefficients between the waves (Model Ib) and the ozone forcing factors (Model IIb) have not been found 2σ statistically significant. Therefore, the results of Models Ib and IIb are not presented here. As it was expected, the trends show dependence on altitude. The posi-

tive trends have been disclosed in the 2-8 km layer in the north temperate region and at Resolute. The trends in this layer show variations with season, *i.e.*, in the north temperate region trends in warm seasons (summer,

Table 7 The magnitudes of the total ozone changes induced by the QBO, ENSO, and the long-term solar activity variations for the north temperate layer-mean ozone (2-8 km, 8-16 km, 16-24 km, 24-32 km layers). These results have been derived by Model IIa using the sonde-derived ozone for the period 1968-1989. The errors of the estimates (in parentheses) are at 2σ level. The statistically significant results are underlined.

LAYER	Magnitudes of the ozone changes		
	QBO	SOI	SUN
2-8 km	-0.058(0.064)	-0.379(1.147)	<u>0.029(0.023)</u>
8-16 km	<u>-0.136(0.116)</u>	-1.853(2.123)	-0.013(0.041)
16-24 km	<u>-0.068(0.065)</u>	-0.686(1.137)	-0.002(0.025)
24-32 km	-0.001(0.039)	0.061(0.703)	-0.002(0.012)

autumn) are higher than in cold seasons (winter, spring). The opposite seasonal variations in the tropospheric ozone trend have been found at Resolute. In the 16-24 km layer, the negative trend has been disclosed in both regions in all seasons. The ozone loss is higher at Resolute than in the north temperate region (about 2 times) with a distinct maximum in winter and a minimum in autumn. In the 16-24 km layer, the seasonal trend variations in the north temperate region are much smaller than those at Resolute. A decreasing tendency in the atmospheric ozone has been revealed also in the upper troposphere and lower stratosphere (the 8-16 km layer, see the year-round and summer trends) over Resolute, whereas in the north temperate zone the trends have been found statistically insignificant there and the trend in autumn shows even an increasing tendency. In layer 24-32 km, the negative trend has been disclosed in the year-round data for the north temperate region and in the winter data taken at Resolute. The large errors of the seasonal trend estimates for the north temperate region and Resolute do not allow us to make a conclu-

Table 8 Trends (year-round, and seasonal) in the layer-mean ozone (in % per year) during the period 1970-1989 for the north temperate and the north polar region. The following layers have been considered: 2-8 km, 8-16 km, 16-24 km, 24-32 km. These results have been derived by Model Ia and Model IIa using the sonde-derived ozone for the period 1968-1989. The errors of the trend estimates (in parentheses) are at 2σ level.

-----TRENDS IN LAYER MEAN OZONE - (% per year)-----										
LAYER	MODEL Ia					MODEL IIa				
	YEAR-ROUND	DEC-FEB	MAR-MAY	JUN-AUG	SEP-NOV	YEAR-ROUND	DEC-FEB	MAR-MAY	JUN-AUG	SEP-OCT
NORTH TEMPERATE REGION										
2-8 km	1.13 (0.15)	1.07(0.28)	0.72(0.20)	1.32(0.30)	1.42(0.32)	1.01 (0.17)	0.97(0.32)	0.60(0.21)	1.19(0.34)	1.30(0.36)
8-16 km	0.11(0.22)	-0.07(0.46)	-0.37(0.48)	0.33(0.39)	0.55(0.37)	0.12 (0.27)	-0.09(0.55)	-0.26(0.56)	0.29(0.47)	0.54(0.45)
16-24 km	-0.45(0.14)	-0.39(0.34)	-0.52(0.29)	-0.45(0.24)	-0.43(0.21)	-0.44 (0.15)	-0.39(0.37)	-0.45(0.30)	-0.46(0.26)	-0.46(0.23)
24-32 km	-0.08(0.07)	-0.11(0.16)	-0.05(0.15)	-0.11(0.13)	-0.07(0.09)	-0.08 (0.09)	-0.10(0.20)	-0.06(0.18)	-0.10(0.16)	-0.06(0.10)
NORTH POLAR REGION										
2-8 km	0.85 (0.32)	1.05(0.78)	1.08(0.57)	0.66(0.44)	0.61(0.60)	0.39 (0.30)	0.51(0.84)	0.60(0.61)	0.25(0.49)	0.18(0.66)
8-16 km	-0.37 (0.30)	-0.55(0.85)	-0.37(0.62)	-0.49(0.45)	-0.09(0.37)	-0.36 (0.32)	-0.53(0.91)	-0.25(0.63)	-0.53(0.42)	-0.11(0.39)
16-24 km	-0.85 (0.15)	-1.11(0.36)	-0.78(0.30)	-0.84(0.22)	-0.69(0.30)	-0.82 (0.19)	-1.07(0.44)	-0.74(0.35)	-0.81(0.27)	-0.64(0.36)
24-32 km	-0.09 (0.20)	-0.52(0.43)	0.11(0.34)	0.16(0.30)	-0.10(0.46)	-0.02 (0.21)	-0.39(0.45)	0.05(0.34)	0.25(0.31)	0.00(0.47)

sive statement about seasonal variations in the ozone trends in the middle stratosphere.

The seasonal trend variations in layer-mean ozone should be treated with special caution because the errors of the trend estimates are relatively large. Furthermore, as discussed by Bojkov and Reinsel (1985), there are long standing problems with the tropospheric ozonesonde measurements. The results of measurements in 2–8 km and 8–16 km layers are less reliable than those in 16–24 km and 24–32 km layers.

Trend analysis of total ozone data for the north temperate region shows (Table 2) that the rate of ozone loss is about -1.7% /decade. In the north temperate region, the trend in total ozone, composed as a weighted sum (weighted according to each layer contribution to total ozone amount, *i.e.*, 5%, 15%, 35%, 35% for 2–8 km, 8–16 km, 16–24 km, 24–32 km, respectively, Angell, 1987) of the statistically significant trend estimates in layer-mean ozone, can be calculated as $-1.44 \pm 0.2\%$ per decade. The 2σ error of this estimate has been obtained assuming that the trends in layer-mean ozone are uncorrelated. The loss in total ozone at Resolute, derived from the trend estimates in the layer-mean ozone, has been calculated as $-3.4 \pm 0.4\%$ per decade. This value is slightly larger than the estimates of trend in total ozone over the north polar region (see, Table 2).

The correspondence between the trend estimates of total ozone obtained from the total ozone data and from the sonde-derived ozone raises a confidence in the quality and representativeness of the sonde-derived ozone as to be good enough for a trend analysis.

Results of the trend analysis in layer-mean ozone over the north temperate region presented in this section can be compared with the recent trend estimates of sonde-derived ozone updated through 1991 (Stolarski *et al.*, 1992). They calculated trends in the ozone profile for the following northern mid latitude stations: Churchill, Edmonton, and Goose Bay in Canada, Tateno in Japan, and Hohenpeissenberg in Germany. Tropospheric ozone increases (ranging from 5% to 20% per decade) and stratospheric ozone decreases (ranging from 5% to

15% per decade) were reported. Moreover, the trends in the ozone profile, based on the satellite measurements (SAGE I/II ozone data from the period 1979–1991), have revealed the rate of the ozone loss comparable with ours trend estimates, *i.e.*, using the results of the trend analysis in the SAGE data, presented by McCormick *et al.* (1992), the ozone loss in the 17–25 km layer (averaged over the northern mid latitudes), can be estimated roughly as about -0.4% per year, whereas the ozone loss in 25–35 km seems to be very small and statistically insignificant.

10. The 'ozone hole' over the Arctic

Resolute, located at 75°N , is a station nearest the North Pole with a long and continuous ozone record. Therefore, it is interesting to make a comparison between the variations of sonde-derived ozone at Resolute and the variations in the Antarctic ozone.

The springtime Antarctic ozone depletion was first reported in 1984 (Chubachi, 1984). There has been a large decrease in the abundance of spring-time ozone since the late 1970s. The TOMS observations of Antarctic ozone show the springtime (September–November) peak rate of ozone decline of about 30% per decade a much lower rate of ozone loss in summer (December–February), *i.e.*, about 10% per decade. Measurements of HCl, ClONO₂ and ClO in the Antarctic corroborate the view that heterogeneous chemical reactions taking place on the surfaces of particles in Polar Stratospheric Clouds (PSCs) release reactive chlorine species from the normally chemically inactive reservoir forms of chlorine, such as HCl and ClONO₂, leading to great enhancement of ozone destruction by the chlorine catalytic chain. The ozone hole over the Antarctic appears in spring after the polar night. When the sun rises above the horizon, solar UV causes abrupt release of chlorine from the compounds formed during the polar night due to the heterogeneous chemical reactions in PSC.

Analyzing the sonde-derived ozone at Resolute, we have found that the dramatic ozone loss is not observed in the Arctic. A maximum

loss in total ozone (about 5% per decade) was discovered in winter (not in spring) and the largest change in the seasonal ozone trends between autumn and winter (not between winter and spring). Therefore, it is suggested that the effects of PSCs on the ozone atmospheric ozone are probably limited in the Arctic.

Findings of the Airborne Arctic Stratospheric Expedition (e.g., Turco *et al.*, 1990) and the balloon campaign at Kiruna (Hofmann *et al.*, 1989; Kondo *et al.*, 1990) indicate that, in spite of evidence of the presence of PSCs and an enhanced level of chemically active chlorine inside the polar vortex, both the magnitude and the extent of these phenomena are reduced relative to the Antarctic. Then, what is the cause of the winter ozone depletion in the Arctic? An effective ozone depletion mechanism involving heterogeneous chemistry on sulfate aerosols has recently been examined by Brasseur *et al.* (1990). These reactions result in enhanced abundance of ClO, which triggers ozone depletion by the chlorine catalytic chain. Observations indicate the presence of reactive chlorine in the stratosphere (with a maximum in winter) over the northern mid and high latitudes at amounts greater than gas-phase chemistry alone could produce (King *et al.*, 1991; Toohey *et al.*, 1991). Then, the heterogeneous chemistry on sulfate aerosols is a likely cause of ozone depletion at mid and high latitudes, but the details of the heterogeneous chemistry still need to be investigated.

11. Summary and conclusion

In this paper, the total ozone data as well as the sonde-derived ozone have been considered. The changes in total ozone have been obtained from the composite time series combining individual station data (monthly or seasonal deviations from the long-term mean level). The composite time series of total ozone, updated on a monthly basis for the period 1957–1990, have been prepared for the following regions: the tropic, north temperate, and north polar regions. The global total ozone data, updated on GEDEX CD as seasonal deviations from the long-term seasonal means, have

been examined for the detection of year-to-year changes in total ozone in the period 1958–1990. The ozone profile data (seasonal deviations) consist of the zonally averaged ozonesonde data over the north temperate region and the results of ozonesonde measurements taken at Resolute (this station represents the north polar region). For these regions, the GEDEX CD provides the seasonal deviations of the layer-mean ozone in the period 1968–1989 in the following layers: 2–8 km, 8–16 km, 16–24 km, and 24–32 km.

The trends and the magnitudes of ozone deviations influenced by the 'natural' variations in the atmosphere (related to the QBO, ENSO, long-term solar activity cycle) have been calculated by means of the least squares fit of the multiple regression model to the observed data. Our basic improvement on the multiple regression model, developed by Reinsel *et al.* (1981), lies in details of the parameterization of the 'natural' changes in the atmospheric ozone. Various multiple regression models have been considered in accordance with the method of this parameterization.

An assumption that the part of ozone variations may be influenced by the interactions between the ozone forcing factors (or dominant wavy disturbances in the atmospheric ozone) allows us to construct a new version of the multiple regression model accounting for these effects (see Models Ib and IIb). In this case, the nonlinear least squares method is used to estimate the magnitude of the year-to-year changes in atmospheric ozone. Moreover, Model IIb provides the possibility of examining how the induced variations in the atmospheric ozone are influenced by the QBO phase. The following are our main findings:

- 1) a downward trend (the winter, summer, and year-round trend) in total ozone has been revealed over zones (the north polar, north temperate, and tropic) and over the world as a whole in the period 1970–1990,
- 2) the decrease in total ozone outside the tropic region depends on the season and in the northern hemisphere a maximum is reached in winter (December–March),
- 3) the seasonal trend variations in the global

total ozone are much smaller than those in the extratropical regions, the maximum ozone loss occurred in northern hemispheric autumn probably due to the Antarctic ozone hole,

- 4) the rate of total ozone loss increases with latitude,
- 5) in the northern hemisphere, outside the tropics, the ozone amount in the lower stratosphere (layer 16–24 km) shows a strong decreasing tendency, whereas in the troposphere an increase in ozone has been revealed,
- 6) the trends in the layer-mean ozone show seasonal variations. In the troposphere, larger ozone increases are found in summer and autumn over the north temperate region, but in winter at Resolute. In the lower stratosphere, the rate of ozone loss reaches its maximum in winter (at Resolute) and spring (the north temperate zone),
- 7) in the lower stratosphere over Resolute, the maximum ozone decline was discovered during, not after, the polar night (as in the case of the Antarctic ozone hole) and the minimum in autumn (not during polar night). Therefore, it seems that the polar stratospheric clouds slightly influence the long-term changes in the atmospheric ozone over the Arctic,
- 8) the results of the trend analyses do not depend on the model, *i.e.*, Models Ia, Ib, IIa, and IIb provided almost the same trend estimates (within the range of 2σ errors). Therefore, the small differences between the estimated trends raise a confidence in the trend models,
- 9) the spectrum analysis of total ozone data shows peaks in the spectrum which seem to be a result of the QBO, ENSO, and the long-term solar activity impact on the total ozone amount,
- 10) the peaks in the spectra corresponding to the quasi-quadrennial and the 11-year oscillations in total ozone are somewhat shifted according to the expected peaks (expected from the spectrum analyses of the ENSO, and 10.7 cm SRF). Then, there is the likelihood that this shift is a manifestation of interactions between the ozone forcing factors. Model IIb reveals that these interactions are in some cases statistically significant, whereas the wave-to-wave interactions (Model Ib) are found statistically insignificant,
- 11) the QBO variations in total ozone show latitudinal variations. In the tropic region, the QBO in total ozone and zonal wind at 50 hPa over the equator are nearly in phase. In the north polar and north temperate regions, an out-of-phase relation exists,
- 12) in the north polar region, the time-lag between the QBO disturbances in total ozone and zonal equatorial wind at 50 hPa (about 14 months) is greater than in the north temperate (about 11 months) and the tropic region (about 3–4 months). This shows the phase propagation of the QBO disturbances in total ozone,
- 13) the QBO amplitude in total ozone varies with latitude. The highest is in the north polar region (especially large if the ozone data are grouped according to the QBO phase). The lowest is in the north temperate region,
- 14) in the north polar and tropic regions, the QBO amplitude in total ozone is almost the same during the east and the west phase of the QBO, whereas in the north temperate region the amplitude is greater in the east phase,
- 15) the QBO disturbances in the layer-mean ozone are revealed in 8–16 km and 16–24 km layers in the north temperate region,
- 16) the ENSO induced variations in total ozone are in phase with SOI in the tropic region. These variations are better expressed in the east phase of the QBO. There is a time lag of a few months between the ENSO in total ozone and the SOI there,
- 17) in the north temperate and north polar regions, the relation between the ENSO disturbances in total ozone and SOI depends on the interactions between the ozone forcing factors. For example, if the large ENSO event (similar to that in 1982/1983) meets the west phase of the QBO and the period of high solar activity, the large ozone depletion will occur in the north temperate region,

- 18) the differences between observed total ozone and the modelled total ozone (by Model IIb) in the north temperate region in the period 1982–1983 imply that the eruption of El Chichon in May 1982 in Mexico had a small effect on total ozone in winter 1982/1983 and spring 1983,
- 19) the QBO and ENSO variations in global total ozone are rather weak. They are not disclosed by Models IIa and IIb. Model Ia estimates about 0.8% ozone increases in total global ozone between the west and the east phase of the QBO,
- 20) the long-term variations in solar activity influence zonally averaged total ozone as well as the global total ozone. The relation between the solar induced disturbances in total ozone and 10.7cm SRF is almost in phase. However, the time lag between the solar induced disturbances and the 10.7 cm SRF shows dependence on latitude. In the north polar region, the solar induced variations precede the 10.7 cm SRF, in the north temperate region the relation is almost in phase, in the tropics the solar induced variations follow the 10.7 cm SRF. Global total ozone data show that the relation is exactly in phase,
- 21) the magnitude of solar induced variations in total ozone depends on latitude, the largest is in the north polar region (derived from Model IIb, when the ozone forcing factors are grouped according to the QBO phase), the smallest is in the north temperate region during the west phase of the QBO (the relation between total ozone and the solar activity is not statistically significant there). In the tropics, the higher magnitude of the solar induced variations is found in the east phase of the QBO. The QBO phase impact on the magnitude of the solar induced variations in global total ozone has not been revealed.

The long-term 'natural' variations in total ozone (caused by the long-term changes in the QBO, ENSO, and solar activity as well as in other yet unknown factors) have been found almost trendless in the period 1970–1990 (see Figs. 6, 7 and 8). In addition, the long-period waves in the atmospheric ozone, which may

have influenced the ozone trends, have not been revealed by the spectrum analysis. Then, we can conclude that the decrease in the atmospheric ozone found during the last two decades is the result of human activities. The present study indicates that the 'natural' variations in the atmospheric ozone consist of a number of components (originating mostly from disturbances of the ozone transport mechanisms), among which the interactions between the ozone forcing factors (the QBO, ENSO, and long-term solar activity) should be considered in the statistical analyses also.

Acknowledgments: —The author would like to thank R. A. Shiffer for providing the GEDEX data. The author acknowledges the thoughtful suggestions made by two anonymous reviewers, which helped to improve the paper. This work was supported by the Science and Technology Agency of Japan Fellowship.

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地上測定から導かれた大気オゾンの年々変動

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北極域、北半球温帯域、熱帯域の3つの帯域、および全球に関して、地上測定によるオゾン全量値、ならびに北極域、北半球温帯域の対流圏と成層圏のオゾン量を、長期変動（トレンド）と短期変動（QBO, ENSO, それに長周期の太陽活動に関連したもの）の検出を目的として解析した。大気オゾンの人為的変動を明らかにするために、各種の多重回帰モデル（トレンド、年々の準周期的変動、また連続相関に関する）について調べた。

1970年から1990年の期間の各季節に対し、北半球の中高緯度帯、熱帯域、さらに全球について、明らかなオゾン全量の下降トレンドが見られた。オゾン消失の割合は、熱帯外の地域では季節に依存し、緯度が高くなるにつれ増加している。

中高緯度帯の層毎の平均オゾンのトレンドは、高度により一様ではない。すなわち対流圏オゾンは増加を示し、成層圏オゾンは減少を示している。北半球の熱帯域外のオゾン全量についての負のトレンドは下部成層圏（16-24 km 層）でのオゾン破壊によって起こされていることが示された。

QBO, ENSO, そして太陽活動に関係した自然要因のオゾンの年々変動の緯度変化を論じた。オゾン全量の自然変動の大きさは、QBO (東風と西風), ENSO (warm と cold) の位相, および QBO, ENSO, そして長周期太陽活動の強さの両者に依存している可能性が見いだされた。大気力学の長期変動が大気オゾンのトレンドに影響を与えていることの可能性については、わずかであることがわかった。