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Tropospheric ozone annual variation and possible troposphere-stratosphere coupling in the Arctic and Antarctic as derived from ozone soundings at Resolute and Amundsen-Scott stations

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ABSTRACT

The tropospheric ozone annual variation in the northern and southern polar regions is analyzed from ozone sounding data obtained at Resolute (74.41° N, 94.59° W) during a 15-year period (1974–1988) and Amundsen-Scott (South Pole) during a 7-year period (1967–1971, 1986–1987). Tropospheric ozone is always less abundant in the southern than in the northern polar region. The difference is greatest in spring in the tropopause layer where the Arctic ozone mixing ratio can be 5 × as large as the mixing ratio in Antarctica. The phase of ozone annual variation above Resolute changes (increases) gradually from the stratosphere across the tropopause to the middle troposphere. Unlike this, the phase of the Antarctic ozone annual harmonic has a discontinuity in the layer of the changing tropopause level, so that the annual harmonic in the upper troposphere, lower stratosphere is 4-to-5 months out of phase (earlier) to that above and beneath. Above both the Arctic and Antarctic stations, the ozone mixing ratio and its vertical gradient evolve in a similar manner in the wide layer from the lower stratosphere to the middle troposphere. This likely points out that ozone in this layer is controlled from above. An indication of the stratospheric-tropospheric ozone exchange above Resolute is noted from mid-winter to spring. The analysis of columnar tropospheric ozone changes gives an lower estimate of the cross-tropopause ozone flux up to $5 \cdot 10^{10}$ mol cm^{-2} s^{-1}. Above the South Pole, the cross-tropopause ozone flux is not usually large. However, in winter, when the tropopause is not strongly pronounced, it can rise to $4.5 \cdot 10^{10}$ mol cm^{-2} s^{-1}, but does not penetrate into the middle troposphere. There is also some evidence that early in the spring, when the stratospheric ozone "hole" is developed, the stratospheric-tropospheric exchange conducts the influence of the "hole" into the upper troposphere, where the integrated ozone destruction is estimated to be $8 \cdot 10^{10}$ mol cm^{-2} s^{-1}. Correlation analysis gives no ozone-tropopause correlation in the Antarctic in winter, while in other seasons as well as during all seasons in the Arctic, there are negative correlation peaks just above the tropopause.

1. Introduction

The ozone balance in the troposphere is influenced by various and complicated dynamical and photochemical processes. The importance of a photochemical source of tropospheric ozone is now well-recognized on both the regional and global scales, see, e.g., Crutzen (1988). Another significant source of tropospheric ozone is ozone inflow from the stratosphere. The relative rôle of the two main sources is not well-known, but there are good reasons to assume that in the polar regions, the photochemical source is less important that at low latitudes, though even in Antarctica, we cannot exclude a long-range transport of pollutants or their inflow from the stratosphere. There are various mechanisms of stratosphere-troposphere exchange, see, e.g., Reiter (1975), Shapiro (1980). There is also

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experimental evidence of ozone exchange episodes in the Arctic (Shapiro et al., 1987; Oltmans et al., 1989) and, in a more exotic case, in the Antarctic (Robinson et al., 1983). Gregory et al. (1989) measured tropospheric ozone in Antarctica aboard an aircraft in spring and found high ozone concentration in the upper troposphere near or underneath the ozone "hole". However the ozone-rich air seldom penetrated below ca. 9 km altitude.

Mechanisms which govern the ozone evolution are displayed particularly in its annual course. At present, the most familiar features of Antarctic ozone are annual variations of total and stratospheric ozone, arising through interest for the ozone "hole" (see, e.g., Solomon (1988)), and surface ozone (see, e.g., Oltmans and Komhyr (1976)). Features of Antarctic ozone annual evolution in the troposphere are less-known. A recent analysis of ozonesonde data made by Gruzdev and Sitnov (1991) has revealed some important differences in the annual behaviour of tropospheric ozone in the northern and southern polar regions. The main object of this paper is to present a more comprehensive analysis of tropospheric ozone annual variations in the Arctic and Antarctic and to study mechanisms responsible for some observed features. The data base is ozonesonde measurements at the stations of Resolute (74.43°N, 94.59°W) over a period of 15 years (1974–1988) and Amundsen-Scott (the South Pole, 2800 m above sea level) over a period of 7 years (1967–1971, 1986–1987). The data were obtained from archives entitled Ozone Data for the World, published by the WMO Ozone Data Center, Downsview, Canada. Before analysis, the data were transformed to an altitude scale and then interpolated to a vertical grid in 1-km steps. The vertical gradients at altitudes away from the earth surface were calculated for 1-km thickness layers, while the calculation of the near-surface gradients used the surface point and the nearest other point in the sounding data.

2. Ozone annual variation

2.1. *Analysis of altitude-time cross sections*

Fig. 1a shows the altitude-time distribution for the monthly-mean ozone mixing ratio above the South Pole, while Fig. 1b shows the mixing ratio difference for the two stations (Amundsen-Scott minus Resolute) divided by the mixing ratios at Amundsen-Scott. The monthly means are com-

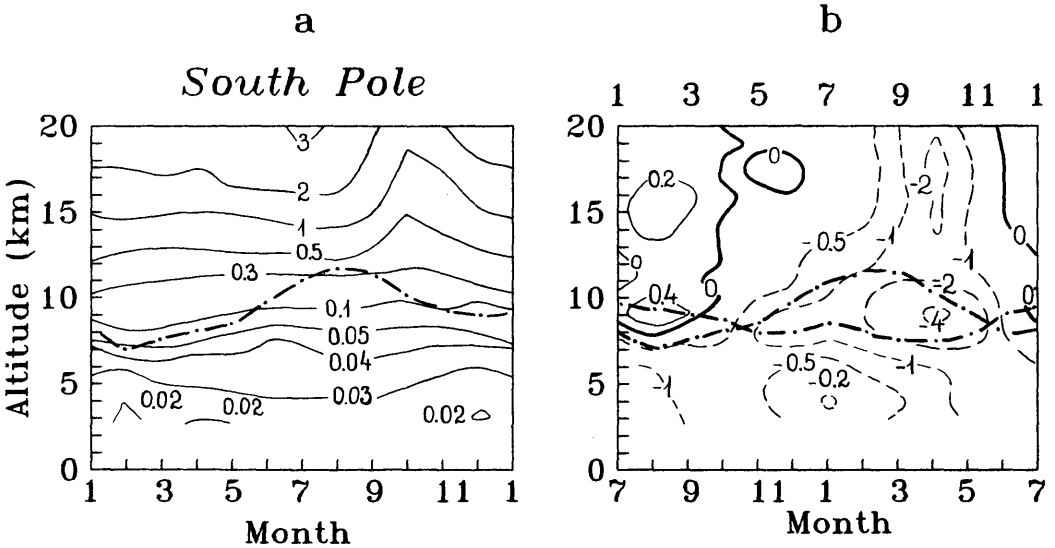

Fig. 1. Altitude-time cross-sections of (a) the monthly-mean ozone mixing ratios (ppmv) above Amundsen-Scott and (b) Amundsen-Scott minus Resolute ozone mixing ratios (with a half-year shift in the Resolute data), divided by Amundsen-Scott mixing ratios. The upper abscissa of (b) corresponds to Amundsen-Scott, while the lower abscissa corresponds to Resolute. Dashed-dotted lines denote the tropopause.

puted by averaging all the profiles in the month. In Fig. 1b, Resolute data are shifted by 6 months. Different types of ozone annual evolution in different parts of the Antarctic troposphere and stratosphere are evident in Fig. 1a. In particular, the effect of the ozone "hole" is displayed in the stratosphere for September to November. According to Fig. 1b, tropospheric ozone is always less abundant in the southern than in northern polar region. The difference is greatest in the tropopause layer in spring-time, where the Arctic ozone mixing ratio in October can be 5 × larger than in Antarctica. In the middle and lower troposphere, the difference is larger in summer and smaller in winter. In the stratosphere, the difference is also mostly negative, with the largest values in the area of the ozone "hole" (14–19 km). A relatively small excess of ozone in the Antarctic stratosphere can be seen late in summer.

To analyze variations and periodicities, it is

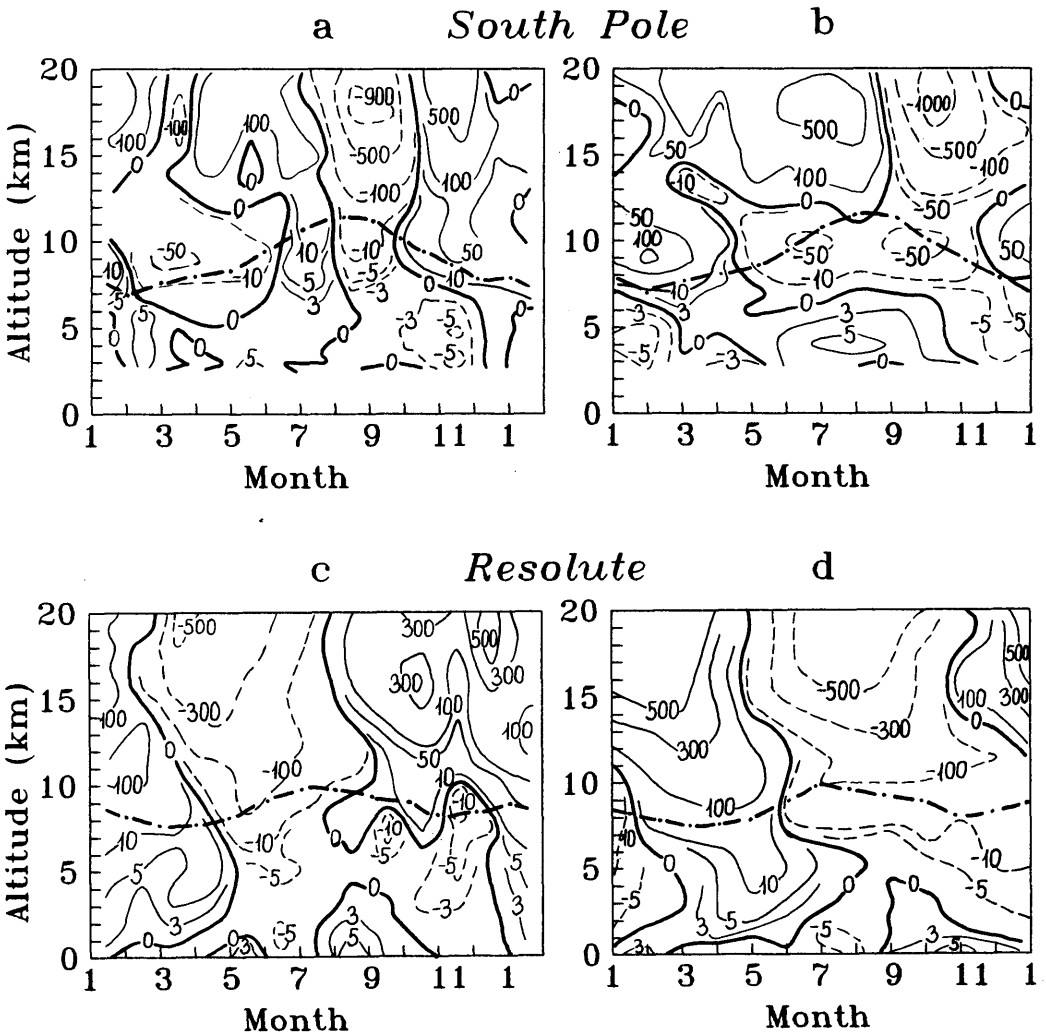

Fig. 2. (a) Month-to-month changes in ozone mixing ratios (ppbv) and (b) deviations from long-term annual-mean ozone mixing ratio (ppbv) for Amundsen-Scott. (c, d) As in (a, b) but for Resolute. Dashed-dotted lines denote the tropopause.

more convenient and useful to study the space-time dynamics of some characteristics of variability (Gruzdev and Sitnov, 1991; Gruzdev et al., 1991). Fig. 2 shows ozone month-to-month changes and deviations of ozone monthly-means from the long-term annual-means above Amundsen-Scott and Resolute. Month-to-month changes are the differences between the monthly means for two neighbour months (latter-month value minus former-month value). The ozone annual evolution above Resolute reveals certain peculiarities in the boundary layer, in the troposphere above the boundary layer, and in the stratosphere (Figs. 2c, d). Various phases of the annual variation (extreme values, local annual mean values) are usually reached first in the stratosphere and surface layer; in the mid-troposphere, these regimes are reached 3 to 6 months later (see zero isolines). Coherent patterns of isolines in the stratosphere, middle troposphere above Resolute in winter and spring point out the stratosphere-troposphere coupling during these seasons. There should be noted an abrupt and simultaneous increase of ozone mixing ratio almost in the entire troposphere in winter and early spring (Fig. 2c), which most likely reflects direct stratospheric ozone intrusions into the troposphere. Such intrusions have been shown to occur during this season (Shapiro et al., 1987; Oltmans et al., 1989).

The content and time variability of ozone in the surface and boundary layer of the Arctic atmosphere are largely controlled by synoptic and photochemical processes (Kelley, 1973; Barrie et al., 1988). One must note the decrease in surface ozone early in spring (Fig. 2c) and the long delay (as much as 6 months) in reaching the annual mean ozone content in the middle troposphere (Fig. 2d). The decrease is connected probably with spring ozone destruction occurring under the Arctic near-surface temperature inversion as the Sun rises (Barrie et al., 1988).

The relationship between tropospheric and lower stratospheric ozone above the South Pole changes during the year (Figs. 2a, b). Besides, Fig. 3 shows the month-to-month ozone mixing ratio changes for two separate periods of observations: before (1967–1971) and during (1986–1987) the years when the ozone "hole" becomes well developed. Note the winter tropopause is much higher for the latter period. The characteristic features of ozone annual variation above the South Pole are the isolated regimes of variability in different atmospheric layers in some seasons. The behaviour of ozone in the wide neighbourhood of the tropopause (i.e., in the lower stratosphere and upper troposphere) is often different from the ozone evolution above and beneath. In winter and spring, when the tropopause is high and often

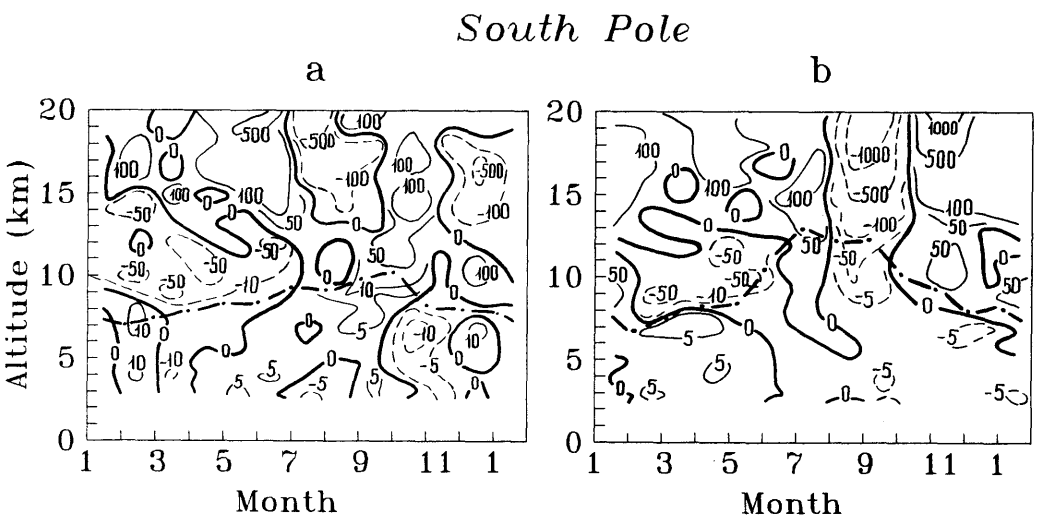

Fig. 3. Month-to-month ozone changes (ppbv) above Amundsen-Scott for (a) 1967–1971 and (b) 1986–1987. Dashed-dotted lines denote the tropopause.

disappears (Makhover, 1983), ozone in the lower stratosphere and upper troposphere changes in a coherent way (Fig. 2a and 3a, b). Note that the ozone annual variations in the upper and lower parts of the Antarctic troposphere are asynchronous (Fig. 2b).

Synchronous ozone changes in the troposphere and stratosphere above Amundsen-Scott during winter-early spring (Figs. 2a, 4) imply stratosphere-troposphere coupling, probably due to a subsidence motion which most strongly occurs during the cold period (Voskresensky and Lisakov, 1976). The tropopause therefore does not preclude such motions. However, even for the isolated winter Antarctic atmosphere, we cannot completely ignore the mean horizontal motions, because of asymmetry of the Antarctic continent (Lisakov, 1976; Voskresensky and Lisakov, 1976).

Fig. 2a, b and 3 also show that the regime of the ozone "hole", as well as the regimes of the winter (Fig. 2a) and spring (Fig. 3a) ozone increases, penetrate into the upper troposphere down to 7–8 km (see also Section 3). At least two mechanisms of this tropospheric ozone decrease should be examined. The first implies the reduced vertical ozone flux, because of low ozone in the "hole" and diminished ozone vertical gradient. The second involves the transport from the stratosphere into the upper troposphere of ozone-destroying pollutants which may continue to influence ozone in the troposphere.

It should be noted that the ozone mixing ratios and their vertical gradients evolve in a similar way in the wide layer from the lower stratosphere to the middle troposphere over the both stations. This may occur if the ozone content of this layer is controlled from above. Therefore, in a rough sense, the increase (decrease) in the above ozone abundance would accordingly increase (decrease) the vertical gradient. If ozone is controlled from below, the relationship between ozone content and its vertical gradient should be the opposite. The exact mechanisms of such relationships cannot be pointed here out. These may be, for example, vertical motions,

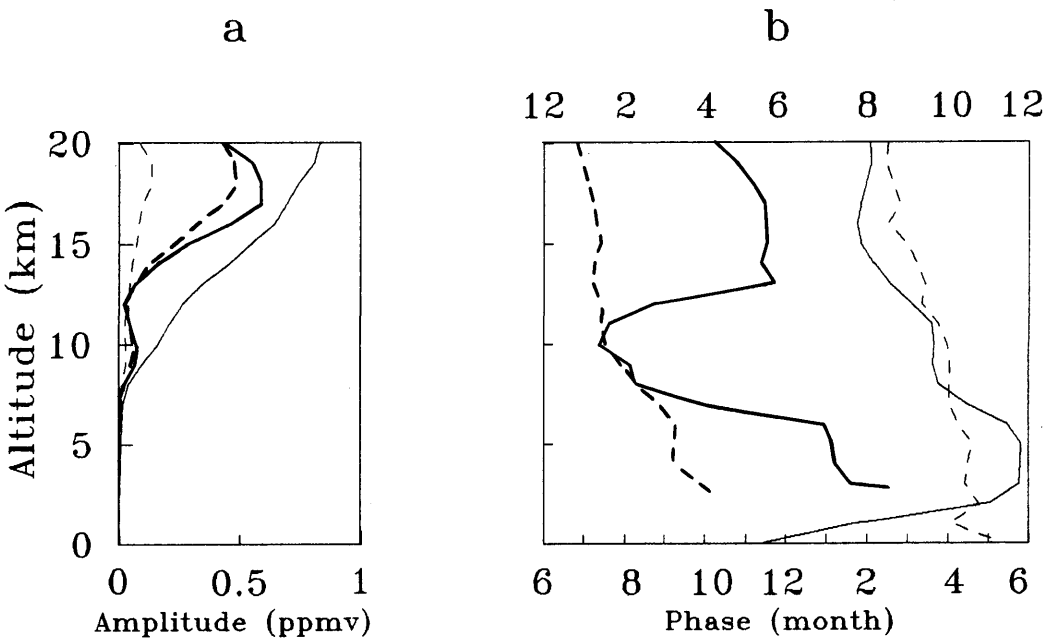

Fig. 4. (a) Amplitude and (b) phase (time of the maximum) of the annual (full curves) and semi-annual (dashed curves) harmonics as a function of altitude for Amundsen-Scott (thick curves) and Resolute (thin curves) ozone mixing ratio. The upper abscissa of Fig. 5b corresponds to Amundsen-Scott, while the lower abscissa corresponds to Resolute.

or, in a more general case, altitude-dependent divergence or convergence of ozone fluxes, both vertical and horizontal.

2.2. *Harmonic analysis of ozone annual variation*

The features of ozone annual variation in different layers of the stratosphere and troposphere are also revealed by harmonic analysis and shown in Fig. 4. The amplitude of the annual harmonic of Resolute's ozone exceeds the amplitude of the semi-annual harmonic in almost the entire sounded atmosphere except the upper part of the boundary layer (Fig. 4a). The semi-annual harmonic also increases in the troposphere. In

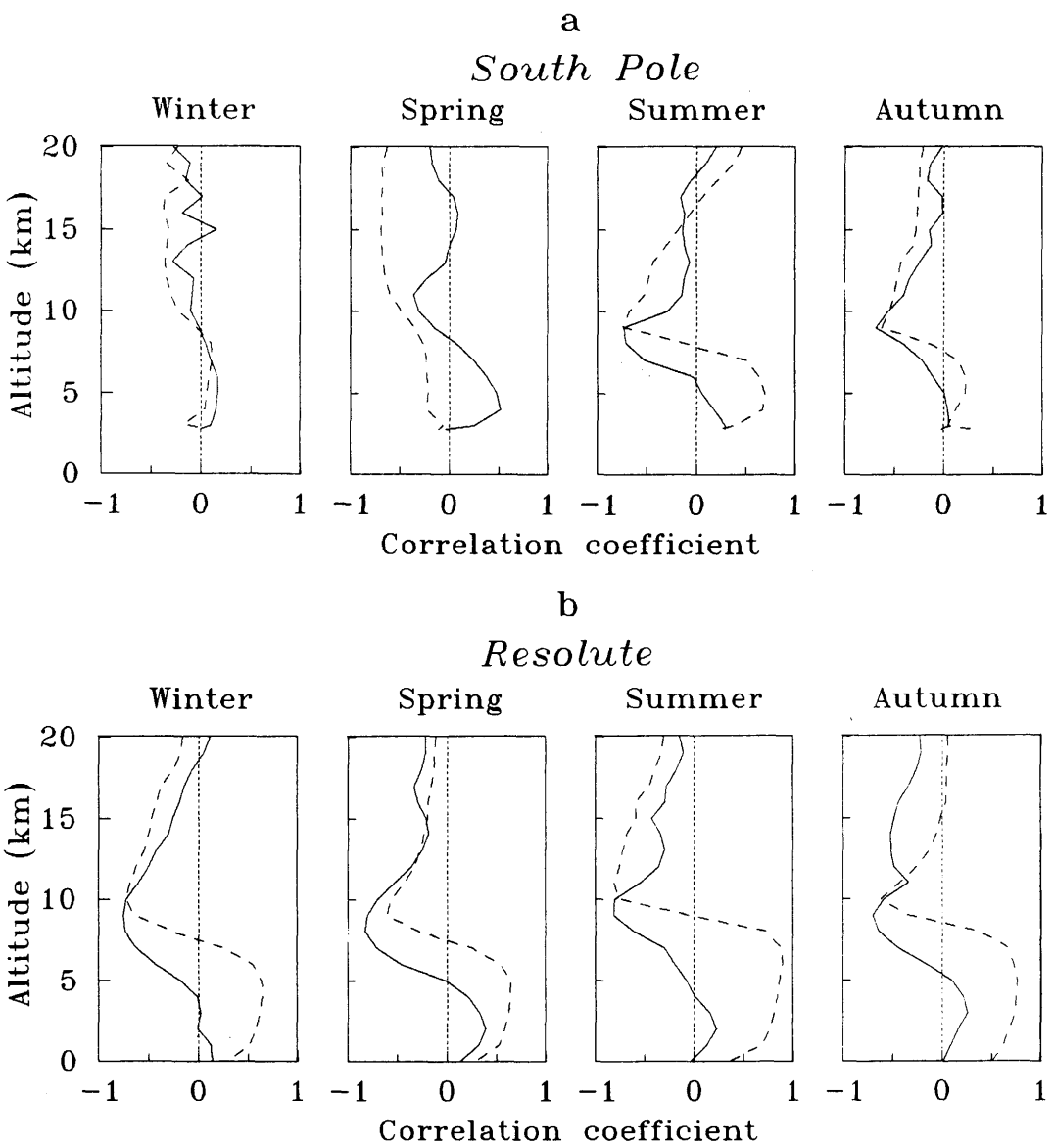

Fig. 5. Winter, spring, summer and autumn ozone-tropopause (full curves) and temperature-tropopause (dashed curves) correlation coefficients for (a) Amundsen-Scott and (b) Resolute data.

contrast, the annual and semi-annual harmonics of Antarctic ozone contribute nearly equally to the ozone annual variation, with the annual harmonic prevailing in the lower troposphere and in the tropopause layer. The amplitudes have two local maxima: at the tropopause level and in the layer where the stratospheric ozone "hole" is observed.

The maximum of the ozone annual harmonic for Resolute is achieved first in the surface layer and then in the stratosphere, from whence this phase spreads gradually to the middle troposphere (Fig. 4b). The maximum phase of the semi-annual harmonic has a more monotonic altitude distribution. Above the boundary layer, it approaches the annual curve, thus reinforcing the maximum and weakening the minimum of ozone annual variation. Note that no strong peculiarity is evident in the phase curve at the Arctic tropopause level; only some steepening occurs there.

Unlike in the Arctic, the phase of the Antarctic ozone annual harmonic has a discontinuity in the tropopause layer, so that the annual harmonic in the upper troposphere, lower stratosphere is 4-to-5 months out of phase (earlier) compared to that above and beneath. The semi-annual maximum phase gradually increases downward from high altitudes, being in phase with the annual maximum phase in the tropopause layer (thus reinforcing the annual maximum) and approximately out of phase above and beneath the tropopause layer (thus weakening the annual maximum and reinforcing the minimum). Therefore, in most part of the Antarctic atmosphere, except in the wide tropopause layer, the negative annual deviations are more prominent than the positive ones.

2.3. Ozone-tropopause correlation

It was seen from the preceding analysis that ozone evolution in certain parts of the troposphere is influenced by atmospheric processes which occur at and in the neighbourhood of the tropopause. Ozone-tropopause correlation can therefore give additional information about these processes. Fig. 5 shows how the Arctic ozone mixing ratio and temperature correlate with the tropopause height during different seasons. In all the seasons, except winter in Antarctica, there is good negative correlation with statistically significant (at 1% or 5% level) peaks just above the appropriate mean tropopause level. Summer and autumn correlation coefficients for the Arctic and Antarctic regions have similar altitude dependence both for ozone and temperature; moreover their near-tropopause peaks have similar values of -0.7–0.8. The same large anti-correlation is also seen in winter and spring for the Resolute ozone curves. In contrast, above the South Pole, near-tropopause ozone does not correlate with the tropopause height in winter and correlates only slightly in spring. This apparently reflects the fact that the tropopause often disappears or is not pronounced in winter and early in spring, and air can freely flow across it. It should also be noted that the near-tropopause winter correlation peak (and to a less degree, the spring correlation peak) for the Arctic region is considerably wider than the summer and autumn peaks, which reaffirms the winter stratosphere-troposphere coupling seen in Fig. 2c.

In the lower troposphere, the ozone-tropopause correlation is usually slightly positive for the both stations, with the largest value of 0.5 in spring for the Antarctic station.

The largest contributions to the correlations shown in Fig. 5 are made by seasonal and inter-annual variability (see Gruzdev and Sitnov, 1991).

3. Estimation of ozone fluxes across the tropopause

The analysis in Section 2 has shown the important rôle of troposphere-stratosphere coupling in the annual variation of tropospheric ozone. In order to get quantitative estimates of ozone fluxes across the tropopause, let us consider the transfer equation for ozone:

$$\frac{\partial n}{\partial t} + \nabla f = q, \tag{1}$$

where n is an ozone concentration, f is an ozone flux, t is time, ∇ is the divergence operator, and q is an internal (photochemical) source. Introducing a "slow" time, T, with a scale greater than a month, then we can write:

$$\frac{\Delta \bar{n}}{\Delta T} + \nabla \bar{f} = \bar{q}, \tag{2}$$

where $(\bar{})$ is a monthly-mean, and $\Delta T = 1$ month. We can now average the finite-difference equation (2) over many years and get the equation of the

same shape with ($^-$) now denoting a many-year monthly-mean.

Let X be the mean total ozone content in the troposphere:

$$X = \int_{z_0}^{z_T} \bar{n}\, dz,$$

where z_0 is the surface level and z_T is the tropopause level. Then, we can write:

$$\frac{\Delta X}{\Delta T} = \int_{z_0}^{z_T} \frac{\Delta \bar{n}}{\Delta T} dz + n(z_T) \frac{\Delta z_T}{\Delta T}. \quad (3)$$

Substituting $\Delta\bar{n}/\Delta T$ from (2) into (3), decomposing $\bar{f}$ into horizontal, $\bar{f}_h$, and vertical, $\bar{f}_v$, fluxes and introducing

$$Q = \int_{z_0}^{z_T} \bar{q}\, dz, \qquad F_h = \int_{z_0}^{z_T} \bar{f}_h,$$

we get:

$$S \equiv -\{\bar{f}_v(z_T) + \nabla F_h - Q\} = \frac{\Delta X}{\Delta T} - n(z_T)\frac{\Delta z_T}{\Delta T} - \bar{f}_v(z_0). \quad (4)$$

S contains the vertical ozone flux at the tropopause level, $\bar{f}_v(z_T)$, the integral photochemical source, Q, and the divergence of an integral horizontal ozone flux, ∇F_h. We may believe that the advective term, ∇F_h, is not so large that it need be considered, because of a partial mutual compensation of random synoptic-scale motions when averaged over time. However this term can still be sufficiently large, so that it cannot be eliminated from (4), because of: (i) the possible existence of a time-mean circulation, (ii) a possible correlation between the fluctuations of horizontal wind and spatial ozone inhomogeneities. The photochemical term, Q, is thought to be small and not essential in winter, but essential in other seasons. Moreover, near the tropopause, q has to be negative, as ozone there is more abundant as a rule than is needed for a photochemical equilibrium. Thus, in a common case we can write:

$$S = f_T - \nabla F_h + Q, \quad (5)$$

where the flux, $f_T \equiv -\bar{f}_v(z_T)$, is directed from the stratosphere to troposphere. If ∇F_h is eliminated, S gives a lower estimate of ozone flux, f_T, (because $Q < 0$) when $S > 0$, and does not give any estimate of flux when $S < 0$; vice versa, when $S < 0$, we get a lower estimate of the photochemical sink, Q.

The surface ozone flux, $\bar{f}_v(z_0)$, on the right-hand side of eq. (4) is governed by the ozone destruction at the Earth's surface:

$$\bar{f}_v(z_0) = -w_d n \varepsilon, \quad (6)$$

where w_d denotes the destruction rate, and ε is a surface factor indicating the fraction of ozone

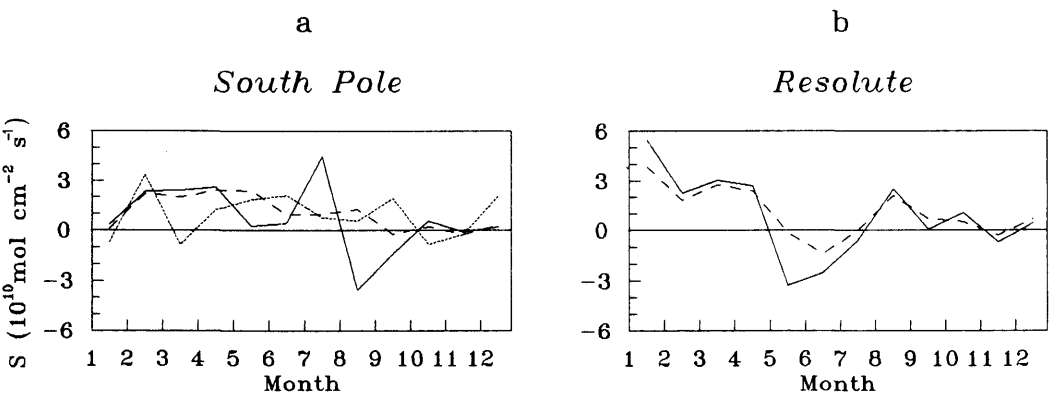

Fig. 6. (a) Sum factor S (see eqs. (4), (5)) as a function of time for Amundsen-Scott ozone data: for the 1986–1987 period of observations, with the upper level condition taken at the mean tropopause height (full curve); for two periods: 1986–1987 (dashed curve) and 1967–1971 (dotted curve), with the upper level condition taken at 7-km height. (b) Sum factor S for Resolute data (1974–1988), with the upper level condition taken at the mean tropopause height (full curve) and 7-km height (dashed curve).

molecules in contact with the surface. Fabian (1973) gives $w_d = 0.02$ cm s^{-1} for snow and ice surfaces, and $\varepsilon \approx 0.9$ for horizontal wind velocities $u \geqslant 3$ m s^{-1} above an ice surface. Therefore we get from (6):

$$\bar{f}_v(z_0) = -0.018\bar{n}(z_0). \quad (7)$$

Eqs. (4)–(7) give a basis for calculations of the sum factor, S, and estimation of the ozone flux, f_T, across the mean tropopause level. The S-value may be considered as a lower estimate of the cross-tropopause ozone flux in cold (e.g., winter) seasons when photochemical processes seem to weaken. However, S tells nothing about the cross-tropopause flux when photochemistry dominates in ozone changes. In this case, the S-value may be considered as a lower estimate of the integrated photochemical sink of tropospheric ozone, if photochemical processes result in ozone destruction.

Fig. 6b shows two variants of the monthly dependent distribution of S for Resolute, with z_T equal to the mean tropopause height (full curve) and with $z_T = 7$ km = const. (dashed curve). S has high positive values in winter and spring and significant negative values in early summer. In autumn and early winter, the S-value is not so large. Taking into account the above discussion, values of about $5.5 \cdot 10^{10}$ and $3 \cdot 10^{10}$ mol cm^{-2} s^{-1} can be lower estimates of cross-tropopause ozone fluxes in mid-winter and spring. The lower value of the integrated strength of a photochemical sink of tropospheric ozone is estimated to be $3 \cdot 10^{10}$ mol cm^{-2} s^{-1} early in summer. It should be noted that the positive S-values at the tropopause and constant 7-km level are close to each other. This suggests that the troposphere-stratosphere coupling above Resolute affects mid-tropospheric ozone (see also Fig. 2c). The estimated Arctic winter-spring cross-tropopause ozone fluxes are smaller than those extrapolated by Fabian and Pruchniewicz (1977) ($7 \cdot 10^{10}$ mol cm^{-2} s^{-1}) but are close to those used by Levy et al. (1985) in a model study ($5 \cdot 10^{10}$ mol cm^{-2} s^{-1}).

Similarly to Fig. 6b, two curves in Fig. 6a show the S-values for the 1986–1987 period of observations at Amundsen-Scott, with z_T equal to the tropopause height (full curve) and with $z_T = 7$ km (dashed curve). The third, dotted, curve in Fig. 6a shows the S-value for the 1967–1971 period of observations at Amundsen-Scott, with $z_T = 7$ km. The cross-tropopause flux of ozone above the South Pole in 1986–1987 is largest in winter when it reaches a value at least as large as $4.5 \cdot 10^{10}$ mol cm^{-2} s^{-1}. Early in spring, when the tropopause is still high and feebly marked and when the stratospheric ozone "hole" is developed (Fig. 2a, b and 3b), S in Fig. 6a takes the largest negative value of $-3.5 \cdot 10^{10}$ mol cm^{-2} s^{-1}. Assuming that the cross-tropopause ozone flux is maintained at the former value, the integrated tropospheric ozone photochemical sink can be estimated to be at least as large as $8 \cdot 10^{10}$ mol cm^{-2} s^{-1}. In contrast to the Arctic region, the ozone fluxes from the stratosphere above the South Pole do not usually penetrate deep into the troposphere as can be observed from the different behaviour of the full and dashed curves in Fig. 6a. Moreover, the photochemical processes in the troposphere beneath the ozone "hole" are also limited to the upper troposphere. This again illustrates the mentioned breakdown of the stratosphere-troposphere coupling in the Antarctic middle troposphere.

The mean cross-tropopause ozone flux above the South Pole during 1967–1971 (not shown) differs from that during 1986–1987, not only because of differences in ozone variability during these periods, but also because of large differences in the tropopause heights seen in Fig. 3a, b. The mean fluxes across the constant 7-km level are more suitable for comparison. The common features of the dashed and dotted curves in Fig. 6a are the largest S-values in autumn and early winter and small, near-zero, S-values in spring.

5. Conclusions

The preceding analysis has revealed essential differences in tropospheric ozone annual variations above Resolute and Amundsen-Scott stations, which are apparent, particularly, in different manifestations of stratospheric-tropospheric ozone exchange. The stratosphere-troposphere coupling over both the Arctic and Antarctic stations is most obvious during winter and early spring. In the Arctic, it even affects lower troposphere ozone. Unlike in the Arctic, the stratosphere-troposphere coupling above the South Pole is limited to the upper troposphere.

The results of these analyses for only two stations cannot be fully extended to the whole polar

regions, especially in the Arctic. This deficiency is hard to overcome because of the lack of other polar ozone recording stations with sufficiently long, and full years of measurements. It is likely, however, that qualitative and some quantitative ozone annual variation characteristics derived in this paper are to be found over large regions of the Arctic and Antarctic. This is so, because such characteristics reflect, to a large part, features of the global atmospheric circulation in these regions.

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