Ozone and atmospheric transport processes

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ABSTRACT

A systematic program to measure vertical ozone distribution was established by the Air Force Cambridge Research Laboratories in January 1963. High resolution measurements with the Regener chemiluminescent ozonesonde have been obtained from a network of twelve stations in North America, which extends from the Canal Zone to the Arctic region. Processed data from the first two years of network operation were used to analyze the broad-scale characteristics of the ozone distribution.

Detailed meridional cross sections of ozone mixing ratio were constructed and compared with simultaneous distributions of other atmospheric tracers including potential vorticity and radioactive debris. The striking consistency in the behavior of these tracers, each having quite different sources and sinks, suggests that the extratropical lower stratosphere is strongly stratified but on the average remains well mixed along surfaces which have a significant slope downward toward higher latitudes. To a good first approximation in all seasons, trace substances in this region of the atmosphere tend toward a uniform distribution along surfaces of constant potential vorticity. Direct computations of the temporal eddy ozone flux indicate that the northward transport across middle latitudes over North America occurs predominately at the base of the stratosphere near the tropopause. The average flux strength observed in the summer and fall seasons is less than one-third of the average winter and spring transport.

1. Introduction

The development of an effective balloon-borne ozonesonde by V. REGENER (1960, 1964) has led to a systematic program for the measurement and analysis of the vertical ozone distribution. A network of ozonesonde stations, as shown in Fig. 1, was established in January 1963 by the Air Force Cambridge Research Laboratories with the cooperation of the operational weather services of the United States and Canada and several universities in the United States. The widespread network extends from the Canal Zone at 9° N to Fairbanks, Alaska, and Thule, Greenland, in the polar region. In general, sounding balloons equipped with Regener instruments have been launched at 1200 UT each Wednesday at each participating station. The program also has included special periods of more frequent ozone observations at selected stations and has been further supplemented by ozone and other meteorological measurements made with a high-altitude research aircraft.

Although the experimental ozone network is sparse in relation to the synoptic-scale changes in the ozone distribution, the resultant data provide an opportunity to study the over-all structure and behavior of an atmospheric tracer constituent with a completeness heretofore unattainable. This paper presents a preliminary analysis of the broad-scale ozone distribution as derived from the relatively large number of high-resolution vertical profile measurements made during the first two years of the program. The distributions of ozone and other tracer quantities are compared and discussed in relation to large scale mixing processes in the atmosphere.

2. Ozone measurements

The dry-chemical method of ozone measurement as described by REGENER (1964) is based on the luminescent reaction of ozone with an active substance consisting primarily of silica-gel and an organic dye, rhodamine B. Under controlled aspiration of ambient air, the photon yield of the disk is proportional to the ozone density. The emitted light intensity is detected by a photomultiplier tube and the resultant

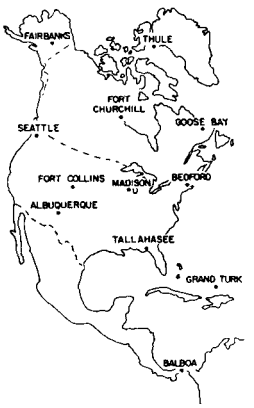

FIG. 1. North American ozonesonde network.

current is fed into the transmitter circuitry of a radiosonde, which is flown in combination with the ozone instrument. Commutator sequencing allots approximately equal time to ozone and meteorological data over a 15-second cycling interval. The response of the sensor to changes in ozone concentration is instantaneous, so that extremely high resolution is achieved in the vertical distribution measurements.

For absolute measurement of ozone, the chemiluminescent sonde must be calibrated. Calibration is achieved in the network program through reference to the total amount of ozone in a vertical column overhead as measured by an ozone spectrophotometer (DOBSON, 1937). The observed concentration of ozone at all levels for an individual sounding is adjusted by a single factor given by the ratio of the total amount measured by a spectrophotometer to the vertically integrated amount measured by the ozonesonde. Also a uniform correction (see HERING & BORDEN, 1965) was applied for a systematic change in sensor sensitivity during ascent in soundings made during the period September 1963 to May 1964.

The absolute accuracy of the ozonesonde data is difficult to assess. The errors inherent in the observations of the vertical distribution include the errors in the measurements of total amount. For example, systematic differences of about six per cent exist between spectrophotometer observations made simultaneously using different pairs of wave-length intervals. Estimates of the relative accuracy of the chemiluminescent-type ozonesonde data have been made through analysis of serial ascents and by comparison (see HERING & DUTSCH, 1965) with vertical distribution measurements using the Brewer-Mast electrochemical ozonesonde. These results indicate that the standard error of relative partial pressure is about 10 nanobars (mb $\times$ 10^{-6}).

3. Characteristics of the vertical ozone distribution

The data for all individual ascents made in the network program are being published in a series of reports (HERING, 1964; HERING & BORDEN, 1964) which are available upon request from the AFCRL. These reports also provide a description of the data processing procedures followed and contain climatological summaries of the ozone data.

An ozonesonde ascent that is typical of those made in the tropical region is shown in Fig. 2. The basic features of the Canal Zone profiles remain essentially the same during all months of the year. Low concentrations prevail throughout the deep and well-mixed troposphere, although minor increases in ozone amount are frequently observed in the upper troposphere in association with occurrences of a more stable temperature structure in the region immediately below the tropopause. The very prominent increase in ozone with increasing height in the lower stratosphere consistently begins at the cold tropical tropopause, and the layer of maximum partial pressure is almost invariably centered in the 25 to 27 km altitude range. Perturbations in the ozone and temperature structure in the tropical stratosphere are characteristically of small vertical scale and are small in amplitude.

Also shown in Fig. 2 is an estimate of the photochemical equilibrium distribution of ozone adapted from the computations of LONDON & PRABHAKARA (1962). The deep production layer is centered near 30 km in the equatorial region. The ozone concentration above the source region is controlled essentially by photochemistry (see, for example, CRAIG 1950). However, below about 25 km ozone is a relatively stable constituent until it is eventually transported down to the surface level where it is destroyed by reaction with ground cover, aerosols and pollutants. Although many uncertainties remain

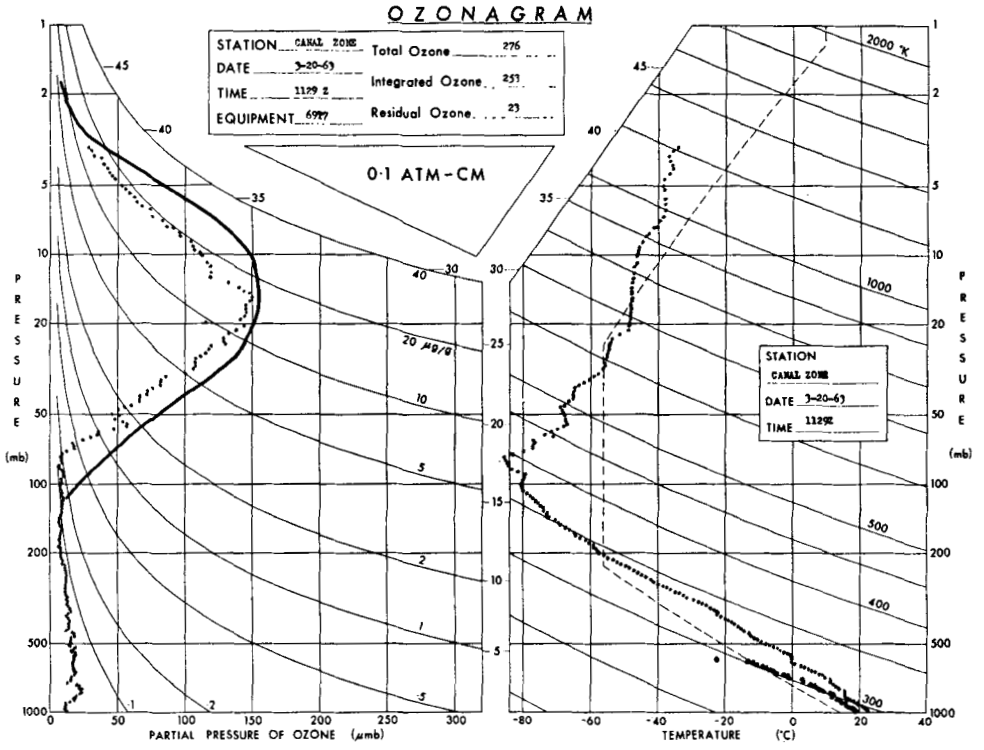

FIG. 2. Ozone, temperature and dew point profiles made at Albrook AFB, Canal Zone, on 20 March 1963. Continuous curve on left diagram is an estimate of the photochemical equilibrium ozone distribution (adapted from LONDON & PROBHAKARA, 1962).

in the photochemical computations, it is seen that the observed distribution in the equatorial region conforms in general with the theoretical equilibrium profile. The deficit of observed ozone with respect to equilibrium amounts clearly must be accounted for by a net transport downward and toward higher latitudes.

In striking contrast to equatorial ozone behavior, the ascents made in the middle and high latitudes show large synoptic-scale and seasonal variations so that it is difficult to define a typical sounding. The Thule, Greenland, ascent shown in Fig. 3, which was made on the same day as the example Canal Zone ascent, closely resembles the average spring profile for the polar region. Large concentrations relative to photochemical equilibrium amounts exist in the lower stratosphere. Although large fluctuations occur in association with the development and motion of large scale disturbances, the maximum partial pressure is usually found near the 16 to 18-km altitude range.

4. Analysis of the broad-scale ozone distribution

It is important to remember in the following discussion that it is the ozone mixing ratio (not density and not partial pressure) which is conservative following the air motion. For example, vertical displacements occurring in the 22 to 30 km altitude stratum with an ozone distribution similar to that shown in Fig. 3 would result in only small local changes in ozone, since the vertical gradient of ozone mixing ratio is small. On the other hand, ozone would be a very sensitive indicator of vertical motion on this day and location in the altitude interval 8 to 20 km, where the relative change in mixing ratio with height is large.

The average seasonal distributions of ozone mixing ratio along a cross section extending from the Canal Zone to Thule were constructed from a total of 421 ozonesonde ascents obtained in 1963 and 1964. Fig. 4 shows the average

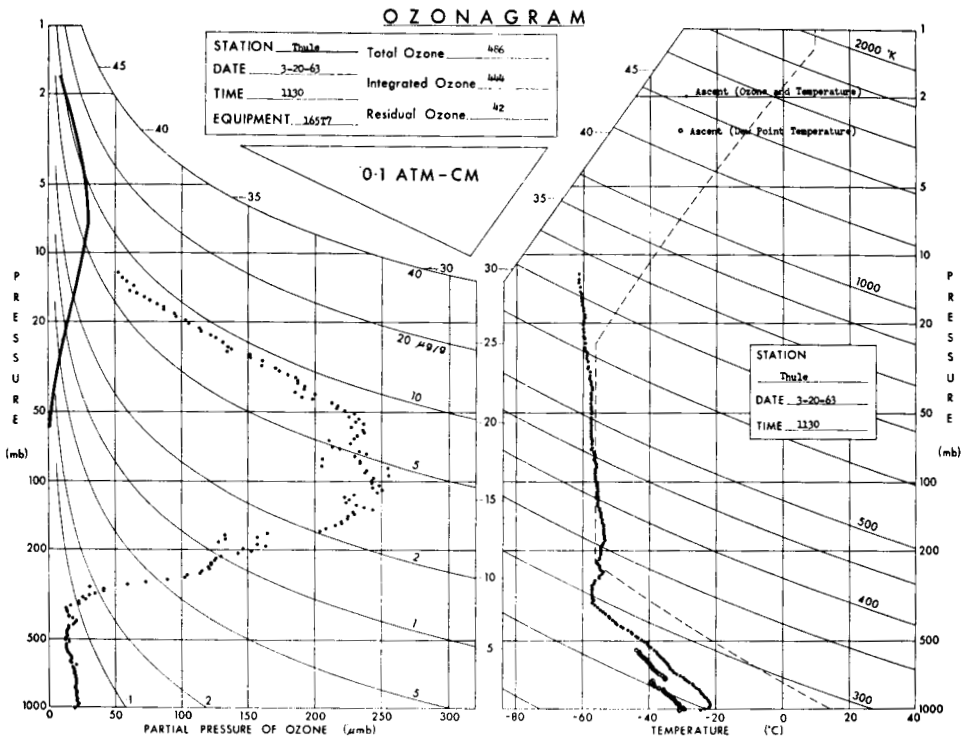

FIG. 3. Ozonagram for 20 March 1963 ascent at Thule AFB, Greenland. (See Fig. 2 for legend.)

ozone cross section for the spring, which is the season of maximum ozone. The average tropospheric ozone amount is quite low (near 0.05 ppm) at all latitudes, with a small but significant maximum at 30 to 40° N. Average values in the stratosphere range from 0.3 ppm near the tropopause to over 13 ppm at 30 km in the equatorial source region. The average mixing ratio surfaces in the lower stratosphere have a prominent slope downward toward the north in middle latitudes. The meridional gradient of mixing ratio decreases with increasing height and reverses direction at about 25 km. Near 30 km, the average ozone mixing ratio at the equator is almost twice as large as the polar region values.

As shown in Fig. 5, the mean mixing ratio pattern in the fall (minimum ozone season) is

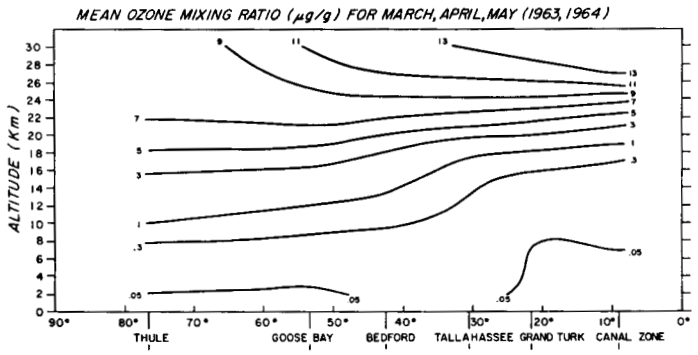

FIG. 4. Average ozone mass mixing ratio distribution for spring 1963–1964. Values are in μg/g or ppm.

quite similar to the spring pattern, although lower average amounts are observed throughout the cross section. The largest vertical displacements with season in the mean ozone surfaces occur in the lower stratosphere in association with the seasonal variations in tropopause position. The seasonal decrease in total ozone mass from spring to fall that is represented by the patterns shown in Fig. 5 is 17 per cent, and the change for that portion of the cross section north of 45° N is 25 per cent.

For a more complete understanding of the circulation processes which maintain the ozone distribution, it is instructive to compare the observed structure with the distributions of other atmospheric tracer quantities. One such quantity, potential vorticity, can be determined from conventional radiosonde observations and has been used extensively (REED, 1955; DANIELSON, 1959; STALEY, 1962) for the analysis of atmospheric circulation dynamics. (Potential vorticity $= -g\eta(\partial\theta/\partial p)$, where g is gravitational acceleration, η is the absolute vorticity computed on an isentropic surface and $\partial\theta/\partial p$ is the vertical gradient of potential temperature with respect to pressure.) Potential vorticity is a conservative property following the air motion (see STALEY, 1962) except for changes due to friction and changes due to a vertical gradient of diabatic heating. Fig. 6 shows a comparison of the average distributions of ozone and potential vorticity along 75° W for February–March

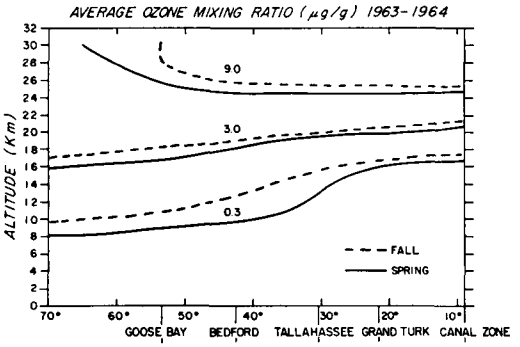

FIG. 5. Comparison of 2-year (1963–1964) average mixing ratio cross sections for the spring and fall seasons.

1963. A striking similarity is noted in the patterns of these tracer quantities in the lower stratosphere poleward of about 20° N and below 20 km. Comparisons made for other months indicate that the correspondence in the average distributions in this region is maintained in all seasons, although the ozone amount associated with a given value of potential vorticity is somewhat higher in the spring than in the fall.

This evidence supports the hypothesis (see GODSON, 1960, FEELEY & SPAR, 1960, and NEWELL, 1961) that the flux of tracer substances is governed largely by eddy transport mechanisms. Furthermore, the correspondence in the ozone and potential vorticity patterns suggests that the eddy mixing occurs preferen-

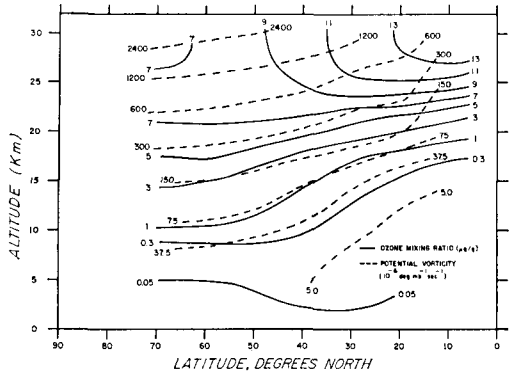

Fig. 6

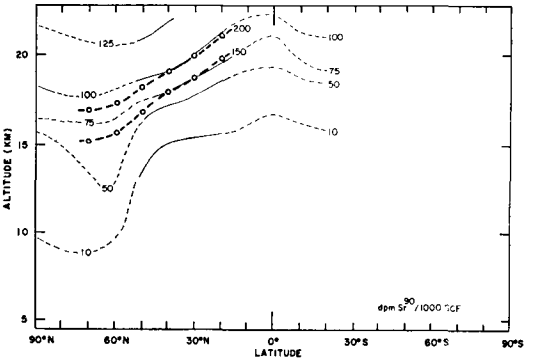

Fig. 7

FIG. 6. Average ozone and potential vorticity distributions for February–March 1963. ——, ozone mixing ratio (μg/g); – – –, potential vorticity (10^{-6} deg mb^{-1} sec^{-1}).

FIG. 7. Comparison of the mean distribution of Strontium-90 in the Star Dust sampling corridor, June–September (from FRIEND *et al.*, 1962) and the average potential vorticity distribution (heavy lines with circles).

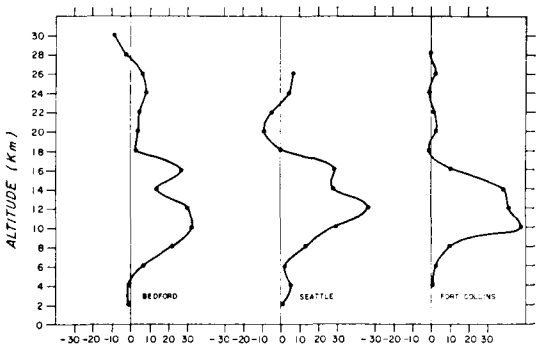

Fig. 8

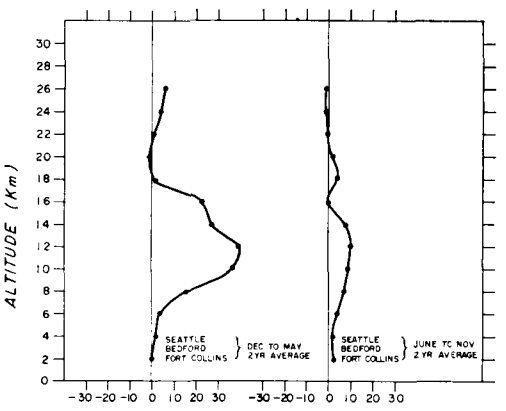

Fig. 9

FIG. 8. Two-year average transport of ozone by transient eddies from December to May 1962–1964. Values are in 10^{-9} gm sec^{-1} cm^{-2}.

FIG. 9. Transient eddy ozone transport, 1962–1964, in 10^{-9} gm sec^{-1} cm^{-2}.

tially along the surfaces of constant potential vorticity. Good agreement in the tracer pattern configurations is observed below 20 km in middle and high latitudes, where large scale disturbances primarily of tropospheric origin are observed. Poor pattern correspondence is noted in low latitudes, where an essentially zonal flow regime is maintained, and in the upper stratosphere above the region normally influenced by tropospheric wave development.

A similar mixing behavior was noted by FEELEY & SPAR (1960) and NEWELL (1963) from an analysis of the spread of a specific radioactive nuclide, W^{185}, introduced into the equatorial stratosphere during the 1958 nuclear test series. The test debris was observed to mix northward and downward along surface having a slope greater than that of the mean potential temperature surfaces, which are less steeply inclined than the average potential vorticity surfaces. Further evidence of selective mixing along potential vorticity surfaces is shown in Fig. 7. The rough comparison of the average distribution of potential vorticity with the quasi-steady state distribution of Sr^{90} in the summer of 1961 near the end of the relatively long moratorium period reveals good pattern correspondence in the extra-tropical lower stratosphere. Thus, the overall consistency in the behavior of various atmospheric tracers in this region, each having different sources and sinks, suggests that all tracer substances tend toward a uniform distribution along the mean potential vorticity surfaces.

5. Preliminary calculations of ozone transport

Estimates of the downward ozone flux to the earth's surface by REGENER (1957) and indirect analyses of the ozone budget (see, for example, JUNGE, 1962) show that a very substantial amount of ozone is transported annually from the source region in the equatorial stratosphere northward and downward to the sink in the surface boundary layer. Rough calculations of the horizontal eddy transport of ozone have been made by NEWELL (1961, 1964) assuming that the ozone density in the lower stratosphere is proportional to the observed total ozone amount. These results show both northward and southward ozone transport by the transient or temporal eddies over broad regions of the hemisphere at 100 mb. However, a substantial net ozone transport northward by the temporal eddies was computed for the hemisphere across middle and high latitudes in both the winter and summer half years. Similarly, his indirect computations of the standing wave component indicated a consistent northward flux in all seasons.

Although the ozonesonde network data are

limited to only the North American portion of the hemisphere, it is of interest to examine the results of direct computations of the transient eddy flux from the simultaneous measurements of the wind and ozone profiles. Fig. 8 shows the 2-year average values calculated for the winter and spring seasons for three stations near 40° N, where the data are most plentiful. The profiles for the winter half year consistently demonstrate that the maximum northward transport by the temporal eddies across middle latitudes over North America occurs at the base of the stratosphere near the tropopause. The indicated eddy flux diminishes sharply above 16 km to rather small values. Fig. 9 shows the 2-year average temporal eddy flux for the winter and summer half years as given by the combined data sample from the 3 midlatitude stations. Although the average transport remains northward at all levels up to 22 km, the over-all flux strength is much weaker during the less vigorous and less disturbed circulation regime in the summer and fall seasons. If the indicated transports are indeed representative of the average hemispheric flux, the total northward ozone transport across middle latitudes would be 1.4×10^9 tons for the cold half year and 0.4×10^9 tons for the warm half year. The calculated annual ozone transport of 1.8×10^9 tons is very close to the average burden of total ozone in the northern hemisphere as given by an analysis of ground-based spectrophotometer measurements (LONDON, 1962).

Acknowledgments

The ozonesonde network has been sustained by the combined efforts of a large number of participants representing many organizations (see HERING, 1964), who have carried out the program with exceptional understanding, care and efficiency. Additional thanks are due to my colleague, Thomas R. Borden, Jr., for his invaluable assistance in the data processing and computational phases of the work, and to C. N. Touart and R. M. Myers for many helpful comments and suggestions about the interpretation of the experimental data. Thanks are also due to Dr. M. L. Barad for reviewing this paper.

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ОЗОН И ПРОЦЕССЫ ПЕРЕНОСА В АТМОСФЕРЕ

В январе 1963 года в Кембриджской военно-воздушной исследовательской лаборатории были начаты систематические измерения вертикального распределения озона. Система из двенадцати станций, расположенных в Северной Америке от Панамского канала до Арктических областей, производила измерения высокой точности с помощью химико-люминисцентного озонного зонда Регенера. Полученные в течение первых двух лет данные использованы для анализа крупномасштабных характеристик распределения озона. Построена детальная карта распределения в меридиональной плоскости процентного содержания озона и произведено сравнение с аналогичными одновременными картами распределения других атмосферных характеристик, включая потенциальный вихрь и радиоактивные осадки. Удивительная последовательность в поведении этих карт, каждая из которых имеет совершенно различные источники и стоки, наводит на мысль, что внеэкваториальное понижение стратосферы сильно стратифицировано, но в среднем остается хорошо перемешанным вдоль поверхностей, имеющих значительный наклон вниз по направлению к высоким широтам. В первом приближении, годном для всех сезонов, распределение вещества (линий постоянных значений характеристик) в этой области атмосферы приближается к равномерному вдоль поверхностей постоянных значений потенциального вихря. Непосредственное вычисление вязкого потока озона показывает, что перенос озона в северном полушарии через средние широты над Северной Америкой происходит в основном в нижних слоях стратосферы. Средняя интенсивность потока, наблюдаемая летом и осенью, меньше чем одна треть среднего зимнего и весеннего потока.