Vertical and Horizontal Fluxes of Ozone at the Tropopause from the First Year of GASP Data

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ABSTRACT

Ozone measurements taken from commercial airliners (GASP data) are used to estimate the vertical and horizontal fluxes of ozone near the tropopause. The annual average flux of O_3 into the troposphere at 30–50°N is 7.8×10^{10} molecules cm^{-2} s^{-1}, which is nearly the same as indirect estimates based on surface O_3 data, thus supporting the hypothesis that the amount of ozone in the troposphere is essentially controlled by injection from the stratosphere. The present GASP estimates of the total flux of O_3 into the troposphere verify the model results of Cunnold *et al.* (1975), although the distribution of flux between mean motions and diffusion is different and thus suggests that models with coarse horizontal resolution must continue to parameterize much vertical transport by diffusion coefficients. A significant variation in the east-west spatial autocorrelation function of O_3 near the tropopause is found to be about 1900 km. Monthly estimates of the horizontal transient eddy flux of ozone are generally smaller than seasonal or yearly results based on ozonesonde data. This is perhaps because the present estimates are made over monthly periods to reduce the influence of correlation between the annual variations in ozone and meridional wind. The available data support the hypothesis that transient eddy fluxes of O_3 have large longitudinal variations.

1. Introduction

The transport of ozone by atmospheric motions is an important link in the global ozone cycle (Dütsch, 1974), but few direct estimates of ozone transport are available. Recent interest in possible anthropogenic effects on the ozone layer has led to many efforts to model those effects. To assess the reliability of a model, its ability to reproduce the observed ozone cycle must be tested. Although there are some indirect estimates of exchange across the tropopause (Fabian and Pruchniewicz, 1976; Reiter, 1975), there have been no direct measurements of the vertical ozone flux, and direct estimates of the horizontal ozone flux are limited to a few ozonesonde stations (Hering, 1966; Dütsch and Favarger, 1969; Hutchings and Farkas, 1971; Pittock, 1968).

The purpose of this paper is to present the first direct estimates of the vertical flux of O_3 through the tropopause. Also given are monthly estimates of the horizontal flux of O_3 near the tropopause, based on the best geographic data coverage available so far. While the data set used here spans only 13 months, it serves to establish the seasonal patterns.

2. Data

All ozone data used here are from the Global Atmospheric Sampling Program (GASP) measurements. GASP is an ongoing effort to measure ozone and other atmospheric variables with instruments aboard commercial airliners. Details on instrumentation, routes, etc., can be found in Holdeman *et al.* (1976), and references therein. A case study of one series of flights is given by Falconer and Holdeman (1976).

A monthly summary of the amount of data and the limits of its geographical distribution is given in Table 1. Although there were a few flights around the world or into the Southern Hemisphere, the bulk of the flights were within the contiguous United States, from the mainland to Hawaii, and from the United States to Europe. The *in situ* O_3 mixing ratio, measured by an ultraviolet absorption photometer, is reported every 5 min (i.e., about every 75 km) during flight above 6 km, but about four observations per hour are missed because the instrument is in a calibration mode. Most of the observations are taken between 10 and 12 km altitude.

Flight level pressure, temperature and winds from the aircraft system are reported with each O_3 observation. Supplementary parameters were computed for each O_3 observation from the NMC Northern Hemisphere grids of isobaric height fields and tropopause pressure fields, which are available at 0000 and 1200 GMT. The map time nearest the mid-time of each flight was used to compute several parameters, including aircraft altitude, geostrophic winds, tropopause separation pressure ($P_{\text{aircraft}}-P_{\text{tropo}}$), and the algebraic sign of the vertical velocity from the diagnostic omega equation. Linear interpolation along isobaric surfaces

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TABLE 1. Summary of GASP data.

Month	Total flights	Total observations	Latitude range	Longitude range
1975				
Mar	57	1263	9°N–61°N	180°E–180°W
Apr	26	554	19°N–47°N	75°W–159°W
May	66	1625	23°S–47°N	45°W–114°E
Jun	35	908	19°N–45°N	84°W–159°W
Jul	3	78	21°N–41°N	84°W–159°W
Aug	16	434	21°N–47°N	84°W–159°W
Sep	23	579	21°N–43°N	75°W–159°W
Oct	25	716	21°N–43°N	75°W–159°W
Dec	10	326	21°N–45°N	72°W–156°W
1976				
Jan	36	1119	9°N–61°N	180°E–180°W
Feb	54	1435	33°S–43°N	72°W–114°E
Mar	39	1057	9°N–57°N	180°E–180°W

and with height was used to estimate these parameters at the aircraft's location for each ozone observation. Complete details are given in Nastrom (1977).

In an effort to establish confidence in the GASP data, mean O_3 values from GASP are compared with those from North American ozonesondes (from Wilcox *et al.*, 1975) in Fig. 1. As most of the GASP data at 40–50°N were taken over North America, differences due to longitude should be small. March data from 1975 and 1976 have been averaged although individual values are also shown. Linear interpolation has been used for November's absent data, and a 1–2–1 smoothing has been applied. The GASP data appear to provide mean values comparable to those from ozonesondes, as the small discrepancies found in Fig. 1 will likely be resolved with more years of data.

3. Results

a. *Vertical ozone flux*

In an effort to estimate stratospheric-tropospheric exchange from the present data, all O_3 observations taken within 50 hPa of the tropopause and north of 30°N were sorted according to the sign of the associated vertical motion estimated by the omega equation from the height fields. The use of the layer tropo$\pm$50 hPa is admittedly arbitrary, but not critical because similar flux estimates result when the layers tropo to tropo$-$100 and tropo to tropo$+$100 are used. The mean ozone in each motion group is given in Table 2, where it will be noted that the O_3 associated with downward motion is always greater than that associated with upward motion. Assuming no net mass transfer across the tropopause, this implies there is a net downward flux of O_3, but to estimate the magnitude of the flux the mean magnitude of the vertical velocity at the tropopause is needed.

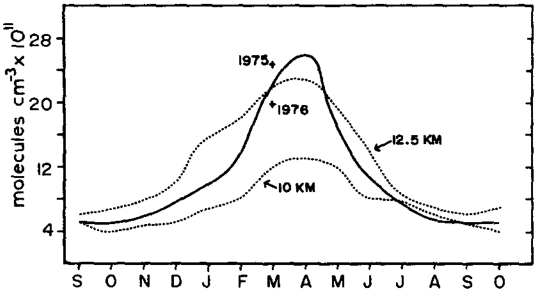

FIG. 1. Annual variation of ozone at 11–12 km, 36–42°N for March 1975 through March 1976 (solid line). The average of all March data was used, but individual year's averages are depicted by crosses. A 1–2–1 smoothing has been applied. Dotted lines are ozonesonde means at 40°N from Wilcox *et al.* (1975).

Case studies of the vertical velocity field suggest that near the tropopause its mean magnitude is a few tenths of a centimeter per second (Palmén and Newton, 1969), but detailed statistics for the Northern Hemisphere do not appear to be available. In the statistical study by Angell (1975), based on Southern Hemisphere EOLE data, the cumulative frequency distribution 50% line is at 0.5 cm s^{-1}. Angell's results show a small variation with season, but that is neglected here as his model is probably valid only for guidance, e.g., it assumes a constant yearly temperature lapse rate. The flux of O_3 across the tropopause, based on $|\bar{w}|=0.5$ cm s^{-1}, is given in Table 2. The confidence limits in Table 2 are

TABLE 2. Ozone mixing ratio, tropo$-$50 hPa to tropo$+$50 hPa, sorted according to the sign of w. Only data north of 30°N are used here. The number of observations is given in parentheses. The diffusive flux is based on $K=3\times10^3$ cm^2 s^{-1}. See text.

	Mean ozone (ppbv)		Mean air density (10^{-4} g cm^{-3})	Net flux based on $\lvert\bar{w}\rvert=0.5$ cm s^{-1} (10^{10} molecules cm^{-2} s^{-1})	Diffusive flux (10^{10} molecules cm^{-2} s^{-1})
	Upward motion	Downward motion			
Winter (D,J,F)	68.0 (887)	79.6 (907)	3.75	9.0±2.5	0.24
Spring (M,A,M)	214.7 (769)	227.3 (758)	3.63	9.5±4.9	0.25
Summer (J,J,A)	143.5 (126)	155.0 (126)	3.30	7.9±8.2	0.13
Autumn (S,O,N)	73.5 (231)	80.6 (151)	3.19	4.7±3.3	0.11
			Average	7.8	0.18

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the root-sum-squares of the standard errors of the mean of the two motion groups each season. The average yearly value, 7.8×10^{10} molecules cm^{-2} s^{-1}, compares well with the results of Fabian and Pruchniewicz (1976) who estimate the flux to be 7.9 and 8.6 units at 35 and 45°N, respectively, based on surface ozone measurements. This very close agreement supports the hypothesis that the amount of O_3 in the troposphere is essentially controlled by injection from the stratosphere.

The vertical ozone transport estimates presented above reflect only the transport by motions whose wavelength is longer than about 700 km, i.e., twice the spacing of the NMC grid at mid-latitudes. The transport of O_3 by disturbances $\lesssim$700 km can be estimated by assuming the flux is the product of an eddy diffusion coefficient and the gradient of O_3 across the tropopause. The diffusion coefficient at the tropopause used by Cunnold *et al.* (1975), 3×10^3 cm^2 s^{-1}, is adopted here, and the gradient of O_3 is estimated by finite differences of mean values of layers 50 hPa thick and centered 25 hPa above and below the tropopause. The resulting estimates of the diffusive flux (Table 2) are only about 3% as large as the corresponding fluxes by large-scale motions. The diffusive flux in winter is based on layer mean values centered 75 and 25 hPa above the tropopause for two reasons. The vertical gradient of O_3 changes rapidly near the tropopause. Also, the NMC tropopause model used after 15 December 1975 apparently yields consistently high estimates of the tropopause pressure (Holdeman *et al.*, 1976).

It is interesting to compare the present estimates of ozone transport across the tropopause with the model results of Cunnold *et al.* (1975), keeping in mind that the latitude band 30–50°N may only poorly represent global mean values and that the results in Table 2 do not include transport by zonal mean motions. Using 10 km as the mean global tropopause height, the transport by large-scale eddies is 31.4 metric tons s^{-1}, and that by diffusion is 0.7 ton s^{-1}. Cunnold *et al.*, give corresponding values of 27 and 5 tons s^{-1}, respectively. Thus, although the total flux is the same (perhaps fortuitously) it is distributed differently. This may be due to their model's truncation at zonal wavenumber 6, for significant O_3 variations near the tropopause are associated with wavelengths near 1900 km (wavenumber 16 at 40°N), as shown in Fig. 2 below. This suggests that if dynamical models are truncated at a low wavenumber, the proper flux of O_3 into the tropospheric sink must be accommodated by parameterized diffusion.

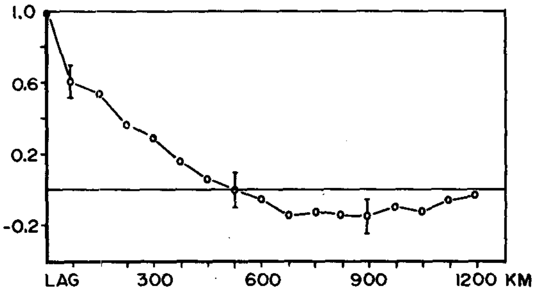

FIG. 2. Distance-lagged autocorrelation coefficients of ozone along east-west flight legs, based on 33 flights. See text.

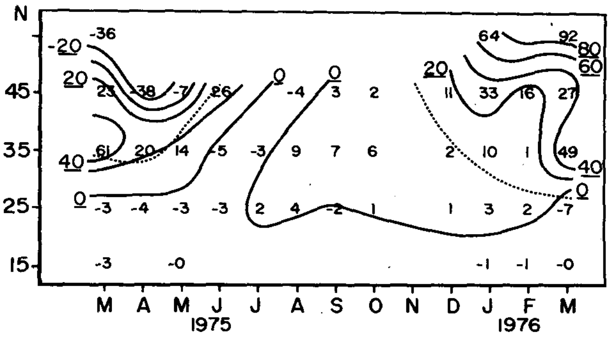

FIG. 3. Northward flux of ozone by transient eddies at 11–12 km by month (units 10^{-9} g cm^{-2} s^{-1}). The dotted lines are mean tropopause location. Isopleth labels have been underlined.

Fig. 2 shows the average spatial autocorrelation function of O_3 based on 33 individual flight legs. The flight legs were chosen to be at a constant pressure level throughout, at least 1200 km long, and were oriented nearly east-west. Over half of the data were spaced at 5-min (75 km) intervals, did not cross the tropopause, and were turbulence free. The vertical bars in Fig. 2 extend one standard error of the mean either side of the mean. The curve in Fig. 2 can be crudely approximated as the product of an exponential decay (i.e., red-noise persistence) and a cosine variation with half-wavelength near 950 km (wavelength near 1900 km). Note that red-noise persistence is characteristic of all atmospheric variables, and that 1900 km is about the distance across intense troughs or ridges.

The results given in Fig. 2 may be of interest to those analyzing other types of O_3 data. It is well-known from sampling theory that measurements should be taken at twice the highest frequency of variability to be resolved. Thus, O_3 measurements near the tropopause should be spaced no more than 950 km in the east-west direction or significant aliasing will occur. Because total ozone is highly correlated with the 100 mb height, it probably has a similar scale of preferred variability. In that case, widely spaced data (e.g., the Nimbus 4 orbits which are about 2000 km apart at 45°N) may be useful only for making zonal or monthly means because synoptic analyses may be severely aliased. There do not seem to have been any studies on this problem.

b. Horizontal ozone flux

Estimates of the ozone flux by transient eddies are given in Figs. 3 and 4. Largest fluxes are generally found in the stratosphere during late winter, although negative values occurred in March–May 1975 at mid-latitudes. The latter fact is contrary to expectations,

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TABLE 3. Flux of ozone by transient addies at 40–50°N and 11–12 km with data divided into 60° longitude sets. The number of observations in each case is given in parentheses. Units are 10^{-9} g cm^{-2} s^{-1}.

	60°E–0	0–60°W	60–120°W	120–180°W	180–120°E
March 1975	−0.1 (2)	12.0 (8)	6.6 (21)	30.2 (27)	—
March 1976	−4.6 (9)	24.5 (29)	8.0 (54)	30.5 (30)	−48.4 (18)
Combined March	−7.8 (11)	28.1 (37)	8.2 (75)	40.1 (57)	−48.4 (18)

as studies of the transient eddy flux based on ozonesonde data (e.g., Hering, 1966; Hutchings and Farkas, 1971) have found positive fluxes throughout the lower stratosphere. This seeming discrepancy may be related to sampling deficiencies or 1975 may have been a very unusual year. A further explanation is the differing length of period over which the transients are computed. Hering combined all data over half-year periods and Hutchings and Farkas combined all data regardless of season, while in the present study monthly periods have been used. The correlation of the annual cycles in ozone and meridional wind thus contributes very little to the present monthly flux estimates. To illustrate the effect of using differing time periods for defining "transient" motions, imagine the meridional wind and the O_3 amount to change from month to month, but to have a constant value within each month. If one then computes transient eddy fluxes over monthly intervals, the results would be zero, but if periods longer than a month were used to compute fluxes, large results would be obtained. The annual cycle in meridional wind at a given location arises primarily from the growth and collapse of standing spatial waves. Standing waves induce a flux of O_3 only if O_3 also has a standing wave pattern, but the magnitude, or even the algebraic sign, of the standing eddy flux cannot be determined from single station data. Because the annual cycles in ozone and meridional wind at one location are not truly transients in the desired sense, they must be removed before computing transient eddy fluxes.

Indeed, the "transient" eddy flux at 11–12 km at 40–50°N based on all GASP data from December–May is 49.5×10^{-9} g cm^{-2} s^{-1}, and in June–October it is 2.3 units, in good agreement with Hering's values (40 and 8 units, respectively), while the average monthly fluxes are 9.3 and 5.3 units, respectively. The difference between the seasonal (49.5, 2.3) and monthly average (9.3, 5.3) values is accounted for by the correlation of the monthly means of ozone and meridional wind.

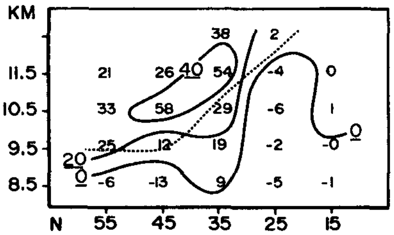

FIG. 4. Northward flux of ozone by transient eddies for combined 1975 and 1976 March data (units 10^{-9} g cm^{-2} s^{-1}). The dotted line is mean tropopause location, and isopleth labels have been underlined.

Although the influence of seasonal variations is minimized in Figs. 3 and 4, these results have noteworthy shortcomings. They may contain a contribution from possible standing eddy fluxes because all data have been used in this first effort, regardless of longitude. However, most data are taken from 65° to 120°W, so these results are not true zonal mean values, and there is no reason to expect zonal symmetry of transient eddy fluxes. In fact, because synoptic disturbances are known to have preferred tracks, distinct asymmetry of the transient eddy flux should be expected. This hypothesis is supported by the limited data during March in Table 3, where transient eddy fluxes at 40–50°N are given for 60° longitude zones. As additional data become available, the zonal variations of eddy flux can be studied in more detail.

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