Seasonal Variability of Middle Latitude Ozone in the Lowermost Stratosphere Derived from Probability Distribution Functions

M C Cerniglia, A R Douglass, R B Rood, L C Sparling, J E Nielsen

Abstract. A fundamental requirement for a model to be useful for assessment of the environmental impact of aircraft exhaust is realistic transport in the lowermost stratosphere. Model transport in this region can be examined and diagnosed through comparisons with observations. Here we present ozone climatologies derived from several years of ozonesonde observations and from observations made by the Halogen Occultation Experiment (HALOE) on the Upper Atmosphere Research Satellite (UARS). To investigate the influence of synoptic waves and tropopause height on the middleworld ozone distribution, the observations are separated into two air mass classes based on Ertel's potential vorticity at 300 hPa. High (low) potential vorticity at 300 hPa suggests that the tropopause is low (high). Conditional probability distribution functions (PDFs) are calculated for these two classes at a series of potential temperature surfaces spanning the domain from the midlatitude tropopause to surfaces higher than the mean tropical tropopause (~380K). The climatologies are similar for the two data sources despite differences in spatial and temporal sampling. The observed PDFs are compared with PDFs calculated for an ozone simulation which uses the NASA Goddard chemistry and transport model driven by assimilated wind fields. Results suggest that the model maintains an appropriate mix of stratospheric and tropospheric air in the vicinity of the midlatitude tropopause and lowermost stratosphere. The model also simulates realistic annual cycles.

Introduction

A detailed understanding of transport and constituent distributions in the lower stratosphere is necessary to reduce the uncertainty in the assessment of aircraft emissions on atmospheric chemistry. The current subsonic aircraft fleet generally flies at 8-13 kilometers near the midlatitude tropopause and often flies in the lower stratosphere at high latitudes. The proposed supersonic aircraft fleet will cruise at approximately 21 kilometers and will traverse the midlatitude upper troposphere and lower stratosphere flight corridors during takeoff and landing. This deposition of supersonic exhaust into the lower stratosphere will add to the slow downward transport (Holton et al., 1995) of supersonic cruise altitude emissions from the overworld. Transport processes in this region are inherently complex and difficult to simulate on short and long time scales. Assessment models must demonstrate appropriate transport and mixing in the lowermost stratosphere (LMS) while maintaining proper constituent gradients. The ability of global models to properly represent the observed transport processes of the LMS must be directly addressed.

Ozone serves as an ideal tracer to investigate model transport in the LMS due to its long lifetime and the availability of observations (e.g. ozonesondes) for model validation. In recent years, HALOE has measured ozone profiles down to the tropopause (ref). Ozone profiles from balloon sondes and HALOE show a high degree of variability in the LMS. We have developed a method to characterize the ozone distribution in the LMS as a means of determining whether or not model transport in this region is realistic. Climatologies are established using observed midlatitude ozone profiles from six years of ozonesonde data and over four years of HALOE Level 2 data. The climatologies are compared by constructing PDFs from each climatology which span the domain of the LMS. Similar analysis is performed on output from a 1997 CTM ozone simulation. Direct comparisons are made between the observations and modeled ozone. By applying constraints on the calculations of the PDFs, additional information can be ascertained about the impact of planetary and synoptic variability. We infer the effects of transport and mixing processes that dominate certain vertical regimes of the atmosphere from the shapes and chemical distinctness of the histograms (Sparling, 1998).

Much of our present understanding of constituent distribution in the upper troposphere and lower stratosphere comes from the study of stratosphere-troposphere exchange. Past studies have focused on the role of synoptic and sub-synoptic scales that directly perturb the tropopause. Attention has been placed on transport mechanisms in the upper tropopause in the vicinity of tropopause folds (e.g. Danielsen et al., 1970; Shapiro 1980; Lamarque and Hess, 1994; Langford et al., 1996; Beekmann et al., 1997) or associated with cutoff lows (e.g. Bamber et al., 1984; Vaughan and Price, 1989; Price, 1990; Price and Vaughan, 1993; Ancellet et al., 1994). The global scale dynamical approach to cross tropopause transport was addressed in a review by Holton et al. (1995). This general mean circulation view of stratosphere-troposphere exchange is based on tropospheric waves that propagate into the overworld and dissipate to form a westward wave-induced zonal force. This force ultimately generates a flow towards the high latitudes, followed by subsidence through the LMS and transport into the extratropical troposphere. Air is then drawn into lower latitudes where it is transported across the tropical tropopause. The "downward-sideways control" discussed by Holton et al. (1995) does not require specific attention to synoptic-scale disturbances. Holton et al. (1995) points out that transport issues connected with the evaluation of the impact of aircraft emissions are not completely addressed by this framework of stratosphere-troposphere exchange and local processes must be considered.

Previous modeling efforts of stratosphere-troposphere exchange involve the use of meteorological fields from general circulation models and data assimilation systems. These include the direct investigation of the meteorological analysis (Mote et al., 1994; Cox et al., 1995; Dunkerton, 1995; Appenzeller et al., 1996a) or its indirect use in contour advection and trajectory calculations (Ancellet et al., 1994; Newman and Schoeberl, 1994; Appenzeller et al., 1996b). Mesoscale (Lamarque and Hess, 1994; Ebel et al., 1996) and global model studies (Chen, 1995; Rood et al., 1997; Gettelman, 1998) usually focus on specific case studies or seasonal transport simulations.

In the work presented here, we deviate from the standard descriptions of observations and model output and emphasize the use of PDFs for the statistical illustration of data and model calculations. Sparling (1998) investigated a variety of stratospheric transport and mixing processes with the use of PDFs.MORE Sparling (1998) also commented on the importance of assessment models to capture statistical features generated from observations. The results of our PDF analysis suggests good agreement between the midlatitude ozonesonde and HALOE data despite several biases. The model maintains a proper vertical mix of stratospheric and tropospheric air in the LMS and tropopause region. The CTM experiment displays realistic seasonal ozone cycles that vary with altitude, exhibits planetary and synoptic wave dependance and suggests realistic horizontal and vertical transport and mixing. Such climatological comparisons provide a robust evaluation of the model's ability to simulate the tracer transport in the LMS as opposed to individual cases studies of transport events. The climatology also captures the cumulative effects of more frequent smaller sub-synoptic scale events.

How well does the CTM simulate the mix of stratospheric and tropospheric air observed in the lower stratosphere, the height of synoptic variability, and the seasonal changes in ozone and tropopause height suggested by the data? What can we infer about the distribution of aircraft effluent based on this analysis? In the followings sections, we will addresses these questions as well as characterize ozone transport in the midlatitude lower stratosphere. The following section provides a description of the ozone datasets used in this study. The next section provides a brief discussion about the CTM and the ozone simulation of 1997. The analysis of the data and model output is then presented followed by a discussion of the results.

Ozone Datasets

Profile data from fifteen ozonesonde stations from 1991 through 1996 are used to establish an observed ozone climatology. The vertical resolutions of the sonde profiles are on the order of a few hundred meters in the lower stratosphere. The accuracy of the observations is ~5% in the lower stratosphere and ~10% in the troposphere (REF). The principle behind the ozonesonde measurements is the output current from an electro-chemical concentration cell produced by a reaction of potassium iodide with local ozone. *In situ* measurements of temperature and pressure are used to correct for operational variations of the sonde's air pump. Data used in this study is sampled from the 42 to 59N latitude band. This is representative of the extratropical midlatitudes and includes the regions of maximum tropopause fold and cutoff low activity (ref Ebel et al., 1996, Beekmann et al., 1997; Price and Vaughan, 19xx). There are inherent sampling problems associated with the sonde stations. The stations are not uniformly distributed and their sampling frequency varies. The balloon launch frequency of the European stations is much greater then those in Asia or North America. The proximity of the ozonesonde sites to the geographical storm tracks, monsoon regions, and other areas of persistent and anomalous meteorological features can introduce significant sampling biases. Beekmann et al., (1997) investigated tropopause folding activity using 10 years of the European Center for Medium range Weather Forecasting analysis and determined that regions in Europe and east of North America and Asia were areas of maximum folding activity. Ebel et al. (1996) presented a similar climatology with slightly greater folding activity at midlatitudes occurring over the Pacific Ocean. Ebel et al. (1996) extended the analysis to include global cutoff low activity and showed that the geographical preference for their formation was analogous to the folds with higher frequencies over landmasses (also see Price and Vaughan, 1992). Monthly analysis of the Goddard Earth Observing System Data Assimilation System (GEOS-DAS) mean PV fields at 46N and at 300 hPa was performed to identify prominent storm tracks and regions of breaking and standing waves. Results suggested that longitudinal distributions of anomalous PV were consistent with the climatologies of Beekmann et al., (1997), Ebel et al. (1996) and Price and Vaughan (1992). Though the locations of the ozonesonde sites and their sampling frequency present some degree of sampling bias, we show in later sections that the profile sampling is not corrupted by their geographical proximity to persistent weather patterns.

Level 2 HALOE ozone profile data from 1994 through the spring of 1998 are used to construct a second observed climatology. HALOE makes a total of approximately 30 sunrise and sunset occulatation measurements at two latitudes per day. For the purpose of our analysis, we utilize data from the 42 to 59N latitude band. Ozone data gathered in the lower stratosphere prior to 1994 was significantly effected by Pinatubo aerosols that prevented accurate observation of constituents in the lower stratosphere and are omitted in this study. The vertical resolution of HALOE is approximately 2 kilometers and the horizontal footprint is approximately 120 kilometers ?? HALOE ozone channel mixing ratio error estimates are ~10% in the mid-stratosphere and ~30% near 100 hPa (ref Russel et al., JGR 1993). A quality control procedure was implemented in our analysis to determine the lowest reliable height of each sampled profile. This is based on the standard deviation of the data. This procedure indicates that the lower observation limit of HALOE is approximately the tropopause level. These lowest observation heights are not significantly effected by convective clouds patterns situated over landmasses. DISCUSS HALOE over sampling at turnaround and relative longitudinal sampling symmetry.

As part of the UARS ozone channel validation, Bruhl et al., (1996) made comparisons of 100 HALOE and ozonesonde profiles and reported that there was excellent agreement in most cases. Root-mean-square differences appeared largest in winter and spring due to increase atmospheric variability during these two seasons. Bruhl et al., (1996) made a direct study of 30 summer and winter sonde profiles from HALOE (mean latitude of 47.7N) and Hohenpeissenberg (47.8N, 10E). The agreement was fair in the lower stratosphere though percent differences increased quickly below 100 hPa. Temporal and spatial variability and Pinatubo aerosol contamination represented a large fraction of their error estimate with the midlatitude correlative measurements.

Model and Experimental Setup

The CTM is described by Douglass et al. (1997). The model utilizes off-line assimilated winds obtained from the GEOS-DAS. The model uses a 2.5 longitude by 2.0 latitude horizontal grid and a 29 level hybrid sigma-pressure vertical coordinate. The vertical domain of the model includes 11 sigma levels from the ground to 130 hPa followed by 17 isobaric levels to 0.1 hPa. The vertical resolution in the lower stratosphere is approximately 1 kilometer. Transport is performed using the flux-form semi-Lagrangian advection routine of Lin and Rood (1996). The one year ozone simulation includes gas phase production and loss chemistry (Jackman et al.,1996) which is updated bi-monthly. An additional global tropospheric loss with an e-folding time of seven days is applied to sigma levels below one scale height. The time step is 900 seconds. The simulation runs from December 26, 1996 through the end of 1997. The initialization is derived from the procedure described by Lary et al. (1995); its implementation in the CTM is discussed by Douglass et al. (1997).

DISCUSS BIAS !!

The use of assimilated data for transport minimizes uncertainties associated with the calculations important for assessment studies. The assimilated wind fields are derived from observations, therefore placement and relative intensity of synoptic scale events which contribute to interactions between the upper troposphere and lower stratosphere are properly simulated. Rood et al. (1992, 1997) presented studies of observed and modeled ozone variability connected with planetary and synoptic scale transport mechanisms. Cerniglia et al. (1995) investigated the transport associated with a polar cutoff low during a persistent blocking event. Cerniglia et al. (1995) showed the cutoff low was responsible for advecting material from the base of the Arctic vortex into the upper troposphere and midlatitudes. Douglass et al. (1996) presented results of a full year ozone simulation and showed that, upon completion of this integration, the synoptic scale features displayed by modeled column ozone were very similar to TOMS observations. The model maintained proper horizontal and vertical ozone gradients in the lower stratosphere. Douglass et al. (1996) concluded that recent improvements in the assimilation system lead to a more realistic residual circulation, realistic zonal mean values of total ozone, and a good representation of synoptic variability at mid- to high-latitudes. DOUGLASS1998??Strahan et al. (1998) used the CTM to simulate the propagation of the phase and amplitude of the tropospheric CO2 seasonal cycle. This signal was used to diagnose transport in the upper troposphere and lower stratosphere. Strahan et al. (1998) showed that the CTM simulated realistic separation of upper tropospheric and lower stratospheric air, proper horizontal transport of the CO2 signal out of the tropical troposphere into the midlatitude stratosphere, and that there was a realistic tropopause barrier. Evaluation of model performance with observations shows the ability of the CTM and assimilated winds to simulate constituent variability for time scales of days to season to interannual.

Analysis of Observations and Model Output

Figure 1a shows a time series of ozone profiles taken from the Hohenpeissenberg station (47.8N, 11E) during 1997. The balloon launch dates are marked with a "+" below the abscissa. The LMS is indentified in Figures 1 a,b by the 380K potential temperature surface and the 2 pvu dynamical tropopause (48N, 11E) both highlighted in white. The "standing mix" of many local ozone maxima and minima is displayed by this cross section and this variability is representative of other northern midlatitude stations. The ozonesonde observations indicate that the midlatitude upper troposphere and LMS is a complex well mixed region subjected to horizontal and vertical advection of stratospheric and tropospheric air. The mechanisms responsible for this mixing are the associated with large scale eddy transport. The synoptic structure displayed in Figure 1a shows the greatest activity during DJF (December-January-February) and MAM (March-April-May). Sporadic low ozone structures (blue and green images) typical of the upper troposphere are representative of ozone advection to isentropic levels near 400K and above. These usually correspond to the effects of breaking Rossby waves that transport subtropical ozone northward. Occurring more frequently are local ozone maxima (red and orange features) present at levels near 350K that are characteristic of stratospheric air typically found at higher altitudes. These observed perturbations generally extend from the middle stratosphere to the lowermost stratosphere. These recurrent transport events are due to meridional advection of air associated with large scale eddies and waves such as upper tropospheric cyclones, blocking patterns, vortex fragments and filamentation. These events are important mechanisms for cross-tropopause exchange and downward transport from the overworld. (ref). MORE on vert transport.

The annual ozone cycle and the seasonal motion of the tropopause is evident in Figure 1a. A substantial transition in tropopause height occurs at the end of July and into August. Figure 1a shows that during the mid-summer and fall, the upper troposphere and LMS possess the lowest seasonal ozone mixing ratios. This is a result of the poleward and upward migration of low subtropical ozone (light and dark blue images) to midlatitudes connected with the seasonal lifting of the tropopause, convection, and increased meridional winds????. Animations of 1997 column ozone data from the Earth Probe Total Ozone Mapping Sectrometer (EP-TOMS refs) suggests that the more intence events (blue images that extend into the overworld during August and September) are linked to transport of low polar ozone past the Hohenpeissenberg station. These transport mechanisms are different than those that occur in winter and spring. During fall and early winter, the tropopause returns to nominal levels and diabatic decent and synoptic activity significantly increases. The downward transport of high ozone mixing ratios leads to stronger gradients in the overworld.

Throughout the year, different mechanisms are responsible for maintaining the observed "standing mix" and seasonal ozone cycles in the upper troposphere and LMS. The midlatitude transport features displayed in Figure 1a are consistent with the seasonal nature of stratospheric-tropospheric exchange and breathing of the LMS. Appenzeller et al., (1996) used a mass continuity model based on pressure variations of the tropopause and the 380K isentropic surface to calculate the seasonal mass of the LMS and net mass transport across the tropopause. Appenzeller et al., (1996) determined that the northern LMS has a maximum mass in midwinter and a minimum in late summer. This is consistent with the highlighted LMS in Figure 1. For the evaluation of the impact of aircraft effluent, it is not adequate to merely simulate the horizontal and time evolution of the tropopause. It is also important to capture the vertical structure of the tracer distribution relative to the height of the tropopause. Figure 1b shows the corresponding time series taken from 1997 CTM simulation. Model output at (48N, 11E) is plotted at the same frequency as the Hohenpeissenberg sonde launches. Despite the high ozone bias that develops, the CTM captures the dynamical variability observed in the middle world and overworld. The model produces seasonal cycles similar to the sondes including the transition to lower ozone partial pressures that occurs in mid-summer and extends into late fall. The confused vertical gradients seen in the data is approximately reproduced however the CTM generates stronger gradients later in the simulation. Note that the Hohenpeissenberg data displayed in Figure 1a is shown at full vertical resolution which is about a few hundred meters; the CTM vertical resolution is nearly a kilometer within this potential temperature range. Rood et al. (1992) made a similar comparison between ozonesonde data collected at Lerwick and model output during the late January 1989 minihole event. Rood et al. (1992) noted the model's inability to simulate the confused structure seen in the upper troposphere and lower stratosphere and the limited extent of synoptic scale perturbations into the stratosphere. Continuous improvements of the CTM transport scheme and with the GEOS-DAS assimilation has lead to a much improved representation of transport in the upper troposphere and stratosphere (see Douglass et al., 1996).

Ertel's potential vorticity (PV) is a measure of horizontal and vertical stability.NEED LEADIN It provides a good diagnostic for a range of dynamical processes. In the absence of diabatic effects, stratospheric PV can be treated as a tracer and it behaves in a manner similar to ozone (refs, Danielsen, 1968,1970; Beekman et al., 1994). Strong gradients of isentropic PV identify semipermeable boundaries between regions of dissimilar character (Hoskins, 1985). For example, on planetary scales, the middle latitude surf zone is bounded by the PV barriers of the subtropical jet stream and the stratospheric polar vortex (ref). These boundaries or transport barriers can easily be located on isentropic maps of PV and other dynamical tracers in the lower stratosphere. Synoptic scale events are evident as regions of high or low PV bounded by strong gradients (e.g. cutoff lows and highs, cyclones, filaments; see Hoskin 1985).

As in the horizontal framework of PV, vertical transport barriers exist that separate air of different makeup. The tropopause separates the low tropospheric values of PV and ozone from higher values found in the stratosphere. The thermal definition of the tropopause is the lowest height at which the temperature lapse rate decreases to 2K per kilometer or less for a depth of at least 2 km (ref WMO). The tropopause can also be located by strong PV gradients and correspond to an isosurface of approximately 2 pvu (1 pvu = 10^{-6} m^2 s^{-1} K kg^{-1}). REF Gettleman. This is an acceptable dynamical definition of the midlatitude tropopause. Strong gradients in ozone can be use to identify the interface between the stratosphere and troposphere (see Figure 1). Bethan et al., (1996) made a comparison of the ozone and thermal definitions of the tropopause using sonde data from four European stations. Bethan et al., (1996) concluded that it was possible to define the tropopause in terms of ozone mixing ratio by locating the sharp gradient in ozone; 98% of the investigated profiles revealed that the ozone tropopause was, on average, 780 meters below the thermal tropopause.

The primary applications of PV in this analysis is to determine air mass identity and to characterize the magnitude and frequency of midlatitude wave events. Figure 2 shows contour maps of isobaric PV at 300 hPa for representative days of MAM (April 1, 1997) and JJA (July 12, 1997). The PV distributions for DJF and SON are similar to MAM and are omitted. The 300 hPa surface is chosen because it approximates the height of the annual midlatitude tropopause (see Morgan and Nielsen-Gammon, 1998). The bold 2 pvu contour marks the intersection with the dynamical tropopause. We can see from Figure 2a that MAM has a number of strong events that coincide with high PV values (darker color images). The majority of the systems transverse the midlatitudes. Note the large meridional PV gradient associated with the Arctic vortex and the succession of high and low PV air masses present between 35 and 55N latitude in Figure 2a. These midlatitude cyclonic scales constantly distort and displace the local tropopause and perturb the lower stratosphere. Examples of these include cyclones and cutoff lows. They generate large scale eddy transport near the tropospheric jet stream and are responsible for the advection that generates the fall and winter "standing mix" displayed in Figure 1a. Systems of this nature usual propagate strongly into the stratosphere as they transfer material across the tropopause. Further mixing is linked to sub-scale processes and smaller eddies (e.g. tropopause folds, filaments, and turbulence mixing near the jet stream). NEED refs.

Adjacent to Figures 2a is the PDF of the longitudinal PV distribution of the 46N latitude circle for April 1st. DISCUSS HOW PDFS ARE CALCULATED. The PV histogram shows a broad PV range with a relatively large variance that is representative of MAM. The low end of the PV distribution (most probable PV values < 1 pvu) corresponds to low subtropical PV air that is advected to midlatitudes during cyclonic events and Rossby wave breaking. At the other end of the spread, and with less probability of occurrence, are PV values in excess of 4 pvu. These high PV values are linked to strong episodic events that transport air from the high latitude PV polar reservoir. Figure 2a reveals most midlatitude high PV systems remain attached to this reservoir. If they detach, the high PV blobs often rejoin their PV source region at a later time which suggest that most of their transport is reversible. A similar phenomenon occurs with low PV air masses that break free from the subtropical barrier and are transported irreversibly to higher latitudes while others return reversibly to subtropical latitudes (Rood et al., 1992b; Rood et al., 1997). Systems that break free from their source regions and ultimately dissipate at midlatitudes are often responsible for irreversible mixing of stratospheric and tropospheric air. Through some stratospheric-tropospheric exchange may be associated with these systems, the magnitude and efficiency of cross tropopause exchange varies and the overall impact on the annual ozone budget has not been fully quantified (ref). Transport is strongly related to transience, with irreversibility arising from either diabatic or nonlinear processes (see Rood et al., 1997). Cyclones that occur during winter and spring are effective for large scale meridional advection of air, stratospheric-tropospheric exchange, and overworld-middleworld transport.

During mid-summer the tropopause is raised at mid- and high- latitudes as a result of the convective lifting associated with increased solar heating (Appenzeller et al.,1996; Schubert and Munteanu, 1988). This seasonal motion of the tropopause is accompanied by an upward and poleward migration of subtropical air to the midlatitudes. This is evident in the seasonal transition displayed in Figures 2a and 2b which shows the majority of high PV air present in MAM is replaced with the low PV air during JJA. Along with this migration is the transport of low ozone mixing ratios to higher latitudes and altitudes. There are fewer synoptic systems and their corresponding PV values are less. These systems tend to be isolated from a high latitude PV source. Planetary wave events diminish. Referring back to Figure 1a, we note a sharp decrease in ozone maxima in the LMS after June and continued upward movement of the tropopause through the fall. Though the frequency of cutoff low events (Price and Vaughn, 1992) and stratospheric ozone intrusion activity (Van Haver et al., 1996) increases somewhat, there is little horizontal advection associated with these systems. The Asian and Mexican monsoons playWHEN? a large role in horizontal nearly isentropic mass exchange from the troposphere into the stratosphere and the poleward migration of subtropical air (Chen, 1995; other refs). A representative PV distribution of these transport mechanisms is presented along side Figure 2b. The histogram reveals a distribution that is sharply peaked at low PV values of less than 1 pvu. The small variance is representative of the large scale migration of low PV air from the subtropics as well as a significant decline in strong PV events. Though we have only discussed daily snapshots here, monthly averages of PV at 46N and the corresponding PDFs are quite similar to those displayed in Figure 2. DISCUSS dunkerton and chens papers.

PV anomalies depress the tropopause height in regions of high cyclonic vorticity and lift the tropopause in regions of low anticyclonic vorticity. This is a result of vorticity stretching and compression (Price, 1990, Price and Vaughan, 1993). Baroclinic waves and eddies that lower the tropopause height will replace ozone-poor tropospheric air with ozone-rich stratospheric air and vice versa. By evaluating the differences in lower stratospheric ozone concentrations associated with profiles sampled from high and low tropopause air masses, the accumulative impact of wave events on ozone and tracer transport in the LMS can be characterized. In this analysis we focus on these two classes of profiles to investigate the influence of planetary and synoptic scales on ozone transport in the lower stratosphere. By calculating two conditional ozone PDFs rather than a single non-conditional PDF (with no air mass classification), we effectively separate the dynamical variability associated with higher PV anomalies. This relationship provides a statistical method to test the ability of a global model to simulate the observed ozone behavior. It allows a direct comparison between different ozone observation platforms, namely the ozondsondes and HALOE. This allows us to study the effects of planetary and synoptic scale events on model generated and observed constituent variability. The separation or difference of the most probable ozone value from each class of PDFs is representative of the climatological impact of planetary and synoptic events.

Ozone transport in the lower stratosphere is examined through a statistical analysis of the observed climatologies. These procedures are also applied to a 1997 CTM ozone simulation. In the following analysis we are concerned with that part of the observed and modeled ozone profile that resides in the upper troposphere and LMS. This region will be directly effected by synoptic perturbations on the local tropopause. Conditional ozone PDFs are found at isentropic surfaces which span the lower stratosphere for the four seasons. The conditionality is based on the profile's air mass classification near the tropopause level. PV at 300 hPa at the location of each ozone profile is used to determine if the profile resides in a high or low tropopause air mass. This criteria for a high tropopause air mass is a PV value of less then 2 pvu and equal to or greater then 2 pvu for a low tropopause air mass. The Figure 2a PV map at 300 hPa locates examples of horizontal regions of a low tropopause air mass (marked with "L") and a high tropopause air mass (marked with a "H"). Ozone profiles sampled from a high (low) tropopause air mass will exhibit a subtropical (polar) character. The extent to which a synoptic event penetrates into the stratosphere will have a direct impact on profile shape at higher altitudes.

The ozone PDFs are constructed by binning the data and model output by potential temperature and partial pressure. Temperatures from the National Center for Environmental Protection analysis are provided with the level 2 HALOE data below 29 kilometers. Ozonesonde temperatures are measured *in situ*. Temperatures corresponding to the gridded CTM output are from GEOS-DAS analysis. These temperatures are used to calculate the potential temperatures. The observations are interpolated to the pressure levels of the CTM which is approximately 1 kilometer in the lower stratosphere. Area weighted ozone data and model output are organized into 10 Kelvin by 10 nanobar bins. The probability that an ozone value will occur on a certain potential temperature surfaces is found by calculating the number of observations in a particular ozone bin divided by the total number of observations or points for that isentropic level. This fraction represents this probability in percent. Due to the limited amount of HALOE data, CTM and ozonesonde points are randomly rejected so that the total number of points for a given isentropic surface are comparable to HALOE.

Conditional PDFs from HALOE, the ozonesondes, and the CTM on the 350K potential temperature surface are presented in Figure 3. The solid (dashed) line histogram represents ozone values for the high (low) tropopause air mass classes. Hereafter these will be referred to as "high tropopause PDF" and "low tropopause PDF" respectively. The 350K isentrope is representative of the "standing mix" and seasonal variability the exists in the midlatitude LMS (see Figure 1a). The emphasis here is on the separation of the most probable ozone values from both classes. This separation is a measure of the impact of the wave events and seasonal tropopause height on profile shape. The structure and separation of the high and low tropopause PDFs suggests that the observations and the CTM display a mix of stratospheric and tropospheric air which varies seasonally as wave activity and tropopause height fluctuates. Figure 3 also shows that the CTM demonstrates a seasonal ozone cycle similar to the observations (COMMENT on CTM SON PDF, assm has large residual circ, synoptic events vertically advected large o3 gradient).

Figure 1a shows the influence of wave activity and tropopause motion on the seasonal structure of the lower stratospheric "standing mix". This is inferred by the seasonal changes in the most probable ozone values, the difference of these values, the variances, and the distribution tails. Referring to Figure 3 and to the ozone distribution at 350K in Figure 1a, the influence of weather patterns becomes apparent. The synoptic systems that correspond to high PV anomalies and lower the tropopause display a high most probable ozone value compared to the high tropopause PDF. Low PV anomalies and the low PV background associated with a high tropopause height generate a small most probable ozone value. The highest ozone mixing ratios plotted in Figure 1a occur in late February and spring. The strong transport events, associated with the advection of high ozone concentrations, are described in the Figure 2a PV PDF with high values of ~4 to 6 pvu. DISCUSS delta most prob o3.

The weather systems associated with the late summer and early fall flow patterns are responsible for the shapes of the JJA and SON PDFs shown in Figure 3. The JJA histograms show the greatest seasonal separation of the most probable ozone values for the high and low tropopause PDFs. The SON histograms show separations analogous to winter and spring however the mechanisms responsible for the distinctness are different. The high tropopause PDFs of JJA and SON are a consequence of the increase in tropopause height and poleward movement of low subtropical ozone into midlatitudes. Note that the variance in the SON high tropopause PDF is less then that of JJA. Figure 1a suggests that the tropopause does not begin to lift significantly until late July and begins to descend to nominal levels in November. The associated poleward movement of low subtropical ozone to midlatitudes follows this migration of tropopause height which generates these PDF variances. (EXPLAIN WHY) The JJA low tropopause PDF is a result of the stronger PV events that persist into June. The intensity of these synoptic events diminishes as the summer progresses into fall and their ability to produce significant horizontal advection decreases. This is evident in the observed SON low tropopause histograms which correspond to smaller ozone partial pressures compared to JJA. Synoptic systems that occur during these periods (e.g. cutoff lows and stratospheric ozone intrusions) exhibit mostly vertical transport of slightly higher ozone mixing ratios from the lower stratosphere into the upper troposphere. The Figure 1a time series shows that the ozone gradients between 400 and 450K are very weak during fall. Hence, these weak gradients (corresponding low ozone compared to June) are transported downward with synoptic events and with the increased downward circulation from the overworld. Monthly PV and ozone PDFs (not shown) give further insight into these seasonal differences.NEED TO MAKE THIS MORE CLEAR.

The analysis shows that the largest most probable ozone values at the 350K surface for both PDF classes occurs during spring and the smallest most probable ozone values are found during fall. This is consistent with known stratospheric-tropospheric activity and seasonal mass changes of the LMS. The net mass transport across the tropopause has a maximum in late spring and a minimum in fall (Appenzeller et al., 1996). The separation in the high and low PDF most probable values is a direct measure of the impact of wave events and seasonal tropopause height on profiles structure at 350K. However, during late summer and early fall there is significant poleward and upward migration of the tropopause which transport low subtropical ozone northward to higher altitudes. Figure 3 displays good agreement in the shape of the observed ozone PDFs at 350K throughout the year. The separation of most probable ozone values and variances are very similar as well. DISCUSS #s. The largest dissimilarity exists in DJF and may be a result of the level of the tropopause and the lower observation limit of the HALOE instrument. (DISCUSS PERCENT OF PROFILE DATA IN TROP & STRAT AND POSSIBLE IMPACT ON DIFF W/OBS PDFs). put these fractions on the plot.

There is good agreement with the CTM ozone seasonal PDFs compared to the observations. The largest discrepancy is seen during SON. This can be partially explained by the large ozone gradient (Figure 1b) that the model develops during fall and December. The strong downward transport produced by the GEOS-DAS circulation transport this gradient to lower levels. This gives rise to the higher most probable CTM ozone value in SON and the greater variances displayed by the model PDFs. Note that the simulated

December 1997 ozone is combined with January and February 1997 ozone used to calculate the DJF histograms. Though the simulation does not display a high ozone bias until two months of integration, the high bias of December corrupts the true nature of the modeled transport exhibited by the DJF histogram.

To examine the seasonal cycle of ozone as a function of height, PDFs are constructed without classifying profiles based on air mass identity. Monthly PDFs are calculated from the observed climatologies and from the 1997 CTM simulation. Since the amount of data varies significantly from month to month, the statistics are not as robust as with the seasonal PDFs. However it allows for a comparison with known seasonal ozone trends and it tests the models ability to capture the height dependant seasonal ozone variability. From the monthly ozone PDFs (not shown), the most probable ozone value is ascertained by averaging the most and second most probable ozone values. This is done for the HALOE, ozonesondes, and for the model output. Figure 4 shows a surface plot of most probable monthly ozone as a function of potential temperature and time. The lower height limit is set to 340K to be consistent with the approximate lower observation range of HALOE. The maximum values of the z-axes are not constant. This view of most probable ozone values suggests reasonable agreement between the two observation platforms. There are similarities with the CTM results. These seasonal cycles are consistent with the Pan et al. (1997) analysis of LMS ozone taken from the SAGE II satellite.

The surface plots of Figure 4 suggests that the midlatitude stratosphere is broken into two vertical regimes. The lower regime has an upper limit of approximately 390K. Within this vertical domain, ozone transport is controlled by planetary and synoptic wave activity and the seasonal motion of the tropopause. Maximum amplitudes are occur in the spring with significantly smaller amplitudes during late summer and early fall (REF pubs on o3 seasonal cycles). Above 390K the upper transport regime reveals a similar seasonal cycle but the apparent change in amplitude from spring to late summer and early fall is much less. This upper domain has a stratospheric character where transport is controlled by the diabatic residual circulation. The characterization of these two vertical transport domains is consistent with Holton (et al., 1995) though the PDF analysis can not distinguish between quasi-isentropic transport between the subtropics and midlatitudes from horizontal and vertical transport associated with planetary and synoptic waves. MORE on penetration of events into lower stratosphere.

The CTM surface plot of Figure 4 implies that the model captures the main features displayed by the data. The CTM surface plot is smoother and the separation of the two vertical regimes is not as sharp compared to the observations. However this is expected by examination of Figures 1a and 1b. The time series of Figure 1a shows a very chaotic lower stratospheric mix of high stratospheric ozone with low ozone values characteristic of the upper troposphere. The observed vertical gradients have a confused nature. Figure 1b reveals similar patterns but the vertical gradients are more monotone with less fine scale structure. The simulation does show the same degree of penetration of synoptic event into the stratosphere as represented by the data. DISCUSS significance of variance, tails and extreme events.

Discussion

Holton et al. (1995) noted that the transport of aircraft emissions near the tropopause is not completely addressed by the global circulation of nonlocal-control within the overworld. It is the local transport processes that occur at the synoptic scale which link the large scale subsidence from the overworld and middle world to two-way exchange with the troposphere. Appenzeller et al. (1996) stated that the net flow across the tropopause is the difference in the inflow from the overworld and the change in mass of the LMS. Though Appenzeller et al. (1996) did not include any two-way exchange between stratosphere and the troposphere, the bulk of stratospheric material enters the troposphere in an irreversible manner. In order for irreversible stratospheric-tropospheric exchange to occur, the destruction or creation of PV must take place (refs). Non-conservation of PV is usually linked to diabatic processes such as latent heat, radiative effects, and turbulent mixing. Irreversibility is also linked to wave amplification and decay time, horizontal velocity, and the magnitude and duration of diabatic heating processes. The ozone PDF analysis displayed in Figure 3 gives some qualitative insight into the question of irreversibility. Variances of the high and low tropopause ozone PDFs indicate the degree of variability of planetary and synoptic systems. Small fluctuations near the most probable ozone values of both PDFs correspond to reversible processes which commonly occur at the tropopause level. The outer tails of the PDFs (low ozone tail of the high tropopause PDF and the high ozone tail of the low tropopause PDF) represent the less frequent extreme events associated with strong PV anomalies. The chemical similarity (measured by ozone partial pressure) between the high and low tropopause air mass can be evaluated by considering the area of overlap of the respective PDFs. If there was no common area, then the air masses are chemically distinct. If there is complete overlap, then the air masses cannot by chemically distinguished. Such is the case at higher altitudes (midlatutude potential temperatures ~> 450K) where the impact of synoptic events becomes negligible. These parameters present a means by which transport processes, described by different observation platforms and global models, can be compared (see Sparling, 1998).

Total ozone is a good indicator of tropopause and PV variability. Schubert and Munteanu (1988) calculated high correlations (> 0.6) of synoptic-scale total ozone and tropopause pressure fluctuations in midlatitudes. The quantitatively analysis of medium scale disturbances in the southern hemisphere and their influence on total ozone distribution were documented by Schoeberl and Krueger (1983). Vaughan and Price (1991) presented a theoretical relationship between vorticity and total column ozone and concluded that distortions in total ozone were dependant on the depths and amplitudes of vorticity anomalies. Intense PV anomalies can penetrate strongly into the lower stratosphere. These episodic events can reach pressure levels as low as 30 hPa (ref). Since time scales for exchange between the LMS and overworld are slower then for cross-tropopause transport (ref Appenzeller), vertical mixing induced by these strong event are of particular importance. During winter months, most synoptic systems (e.g. cutoff lows and streamers) that migrate from high- to mid- latitudes are connected to the high PV source reservoir located at polar latitudes. Systems of this nature can be directly tied to transport out of the base of the polar vortices (Rood et al., 1992; Cerniglia et al., 1995). They have direct implications on transport of stratospheric aircraft emissions to lower levels. REFER TO fig1.

The depth to which midlatitude synoptic events penetrate into the lower stratosphere can be investigated by calculating height dependant correlations between column ozone and local PV. A direct comparison between Earth Probe Total Ozone Mapping Sectrometer (EP-TOMS refs) data and modeled column ozone is made by examining the vertical structure and seasonal dependance of these correlations. Figure 5 shows seasonal variations of these correlations for 1997. The EP-TOMS data is horizontally interpolated to the same horizontal grid as the PV analysis and CTM column ozone. Linear correlation coefficients of total ozone and local PV are found every other day for all longitudes lying along latitude circles within the 42 to 59N deg band. A cosine weighted average is applied to the correlations. The result is squared and is plotted in the height time series of Figure 5. There is good agreement with correlations calculated from the EP-TOMS data (Figure 5a) and the CTM output (Figure 5b). Though the magnitude of the correlations are generally lower for the data, the seasonal cycles are similar and the extent of the high correlations into the lower stratosphere agree qualitatively. The strongest correlations are found in late winter and early spring when planetary and synoptic waves are most intense. A secondary maximum in the correlations is seen in fall and early winter. During summer, the minimum correlations are calculated and these occur at the lowest annual level. DISCUSS lower correlations w/toms due to use of strat pv. The seasonal dependance of these strong episodic events and the height to which they perturb into the lower stratosphere displayed in Figure 5 is inferred from Figure 1. The ozone time series shows strong events penetrating to altitudes consistently exceeding 460K throughout the winter and spring months.

-discuss possible interannual variability w/1997 compared to climatologies. refer to coys paper for 1997.
-discuss pan et al., 1997.

Conclusions

The ozonesonde data reveals that the tropopause region and the lowermost stratosphere (LMS) is a complex area of horizontal and vertical mixing of stratospheric and tropospheric air. Mixing in these regions is associated with material advection induced by planetary and synoptic scale eddies (Shepherd, 1996 and references therein). In this study, probability distribution functions (PDFs) are used to investigate the impact of planetary and synoptic waves on tracer transport and mixing in the LMS. Midlatitude ozone climatologies are established from six years of balloon sonde data and over four years of HALOE Level 2 data. Conditional ozone PDFs are constructed from the two observed climatologies and from a 1997 CTM ozone simulation. The PDFs characterize ozone distribution on potential temperature surfaces that span the lower stratosphere. The PDF analysis provides a diagnostic to study the CTM's ability to simulate the observed ozone distribution in the midlatitude middleworld and overworld. The PDF representation provides a diagnostic framework in which to interpret measurements and compare them with models and independent observations (see Sparling, 1998). The model results show good agreement with the sondes and HALOE data. The CTM maintains a reasonable mixture of stratospheric and tropospheric air in the vicinity of the tropopause and in the LMS. The ozone PDF analysis suggests that the simulation exhibits proper transport and mixing and captures the seasonal ozone cycle. The planetary and synoptic scale variability generated by the GEOS-DAS winds and displayed by the CTM suggests that there is appropriate wave activity linking the downward transport of material from the overworld to complex large scale mixing in the lowermost stratosphere and ultimate transport with the troposphere. The model represents the effects of day to day cross tropopause transport as well as middleworld-overworld exchange (Chen, 1995; Holton et al., 1995) which occurs on a seasonal time scale.

The ozone PDF analysis suggests that the stratosphere is separated into two vertical regimes separated by the 390K potential temperature surface. The upper region is driven by the diabatic residual circulation and extratropical wave-driven pumping. The lower regime is controlled by seasonal planetary and synoptic wave variability and tropopause motion. The analysis of the lower regime shows that the seasonal variation in most probable ozone has a large spring maximum and a summer minimum. This is consistent with the annual stratosphere-troposphere cycle (refs).

Midlatitude observations from 1997 EP-TOMS and modeled column ozone are used to calculate height dependant correlations with local PV. There is good agreement between the data and the CTM correlations. The highest correlations occur during spring with a secondary maximum in winter. The minimum correlations take place during summer. The correlations deduced from the CTM qualitatively agree with the EP-TOMS with respect to the height of maximum correlation and the seasonal nature of the correlations. High springtime correlations are seen in the low- to mid- stratosphere associated with intense synoptic events that penetrate strongly into the stratosphere. This is consistent with the PV PDFs from Figure 2. [MORE ON the perturbation of the LS by synoptic events.]

Global assessment models must include a proper characterization of transport in the vicinity of the tropopause and LMS. The tropopause region and the LMS includes the cruise altitude of subsonic aircraft. The upper troposphere and LMS should be thought of as a mixture of stratospheric and tropospheric air rather then two distinct bodies separated by a semi-permeable boundary. MORE discuss implications wrt aircraft exhaust.

Acknowledgements

We acknowledge the Goddard Ozone Processing Team for supplying EP-TOMS data and the Data Assimilation Office for providing the meteorological analysis.

References

Ancellet, G., M. Beekmann, and A. Papayannis, Impact of a cutoff low development on downward transport of ozone in the troposphere, J. Geophys. Res., 99, 3451-3468, 1994.

Bamber, D. J., P. G. W. Healey, B. M. R. Jones, S. A. Penkett, A. F. Tuck, and G. Vaughan, Vertical profiles of tropospheric gases: Chemical consequences of stratospheric intrusions, Atmos. Environ., 18, 1759-1766, 1984.

Beekman, M., et al., Regional and global tropopause fold occurrence and related ozone flux across the tropopause, J. Atm. Chem., 28, 29-44,1997.

Bruhl, C. et al., Halogen Occultation experiment ozone channel validation, J. Geophys. Res., 101, 10,217-10,240, 1996.

Cerniglia, M. C., R. B. Rood, and A. R. Douglass, Three-dimensional simulation of the influence of a cutoff low on the distribution of northern hemisphere processed air in late January 1992, J. Geophys. Res., 100, 16,431-16,443, 1995.

Danielsen, E. F., Stratospheric-tropospheric exchange based on radioactivity, ozone and potential vorticity, J. Atmos. Sci., 25, 502-518, 1968.

Danielsen, E., R. Bleck, J. Shedlovsky, A. Wartburg, P. Haagenson, and W. Pollock, Observed distribution of radioactivity, ozone, and potential vorticity associated with tropopause folding, J. Geophys. Res., 75, 2353-2361, 1970.

Douglass, A., R. Rood, C. Weaver, M. Cerniglia, and K. Brueske, Implications of three-dimensional tracer studies for two-dimensional assessments of the impact of supersonic aircraft on stratospheric ozone, J. Geophys. Res., 98, 8949-8963, 1993.

Douglass, A. R., C. J. Weaver, R. B. Rood, and L. Coy, A three-dimensional simulation of the ozone annual cycle using winds from a data assimilation system, J. Geophys. Res., 101, 1463-1474, 1996.

Ebel, A., H. Elbern, J. Hendricks, and R. Meyer, Stratosphere-troposphere exchange and its impact on the structure of the lower stratosphere, J. Geomag. Geoelectr., 48,135-144,1996.

Langford, A. O., C. D. Masters, M. H. Proffitt, E.-Y. Hsie, and A. F. Tuck, Ozone measurements in a tropopause fold associated with a cut-off low system, Geophys. Res. Lett., Vol. 23, 2501-2504, 1996.

Lamarque, J. F., and P. G. Hess, Cross-tropopause mass exchange and potential vorticity budget in a simulated tropopause folding, J. Atmos. Sci., 51, 2246-2269, 1994.

Lin, S. J., and R. B. Rood, Multidimensional flux form semi-Lagrangian transport schemes, Mon. Wea. Rev., in press, 1996.

Morgan, M. C., and J. W. Nielsen-Gammon, Using tropopause maps to diagnose weather systems, Mon. Wea. Rev., 126, 2555-2579, 1998.

Price, J. D., Stratospheric-tropospheric exchange in cut-off-low systems, Ph.D. thesis, Univ. Coll. of Wales, Aberystwyth, 1990.

Price, J., and G. Vaughan, The potential for stratosphere-troposphere exchange in cut-off-low systems, Q. J. R. Meteorol. Soc., 119, 343-365, 1993.

Rood, R. B., A. R. Douglass, and C. J. Weaver, Tracer exchange between tropics and middle latitudes, Geophys. Res. Lett., 19, 805-808, 1992b.

Schoeberl, M. R., and A. J. Krueger, Medium Scale Disturbances in total ozone during southern hemisphere summer, Bull. Am. Meteorol. Soc., 64, 1358-1365, 1983.

Schubert, S. D., and M. -J. Munteanu, An analysis of tropopause pressure and total ozone correlations, Mon. Wea. Rev., 116, 569-582, 1988.

Schubert, S. D., R. B. Rood, and J. Pfaendtner, An assimilated dataset for earth science applications, Bull. Am. Meteorol. Soc., 74, 2331-2342, 1993.

Shapiro, M. A., Turbulent mixing within tropopause folds as a mechanism for the exchange of chemical constituents between the stratosphere and the troposphere, J. Atmos. Sci., 37, 994-1004, 1980.

Shepherd, T. G., Transport and mixing in the lowermost stratosphere, First SPARC General Assembly, Melbourne, 1996.

Sparling, L. C., Statistical perspectives on stratospheric transport, J. Geophys. Res., in manuscript, 1998.

Van Haver, P., D. De Muer, M. Beekmann, and C. Mancier, Climatology of tropopause folds at midlatitudes, Geophys. Res. Lett., 23, 1033-1036, 1996.

Vaughan, G., and J. D. Price, Ozone transport into the troposphere in a cut-off low event, Ozone in the Atmosphere, R. D. Bojkov and P. Fabian (eds.), pp.415-418, A. Deepak, Hampton, Va., 1989.

Figure Captions