

A NEW PERSPECTIVE OF STRATOSPHERE–TROPOSPHERE EXCHANGE

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Consideration of the time scales and vertical extent of stratosphere–troposphere exchange events reveals new insight into this phenomenon and its impact on surface ozone.

Stratosphere–troposphere exchange (STE) is important for the chemical composition of both the lowermost stratosphere (LS) and the troposphere. Modifications thereof in a changing climate may significantly affect stratospheric ozone depletion (Butchart and Scaife 2001) and the oxidizing capacity of the troposphere (Lelieveld and Dentener 2000). However, STE is still poorly understood and inadequately quantified, due to the involvement of physical and dynamical processes on local to global scales (Holton et al. 1995), and conceptual problems.

On a long-term and global scale, and in the zonally averaged sense (Brewer 1949), there is slow as-

cent from the troposphere to the stratosphere in the Tropics (Plumb 1996; Mote et al. 1996), quasi-isentropic transport to the extratropics in the stratosphere (Waugh 1996), and downward flow from the stratosphere to the troposphere in middle and higher latitudes (Fig. 1). This circulation is related to the dissipation of extratropical planetary and gravity waves in the stratosphere (Haynes et al. 1991). Based upon calculations of the monthly averaged hemispheric mass flux into the LS (Rosenlof 1995) and of the mass of the LS, the monthly mean net mass flux across the tropopause can be evaluated (Appenzeller et al. 1996).

This two-dimensional picture may suggest that STE in the extratropics is a continuous downward flow. However, actual STE is highly episodic, associated with strong mesoscale perturbations of the tropopause (Appenzeller and Davies 1992), and occurs in both directions. In situ and remote sensing observations have produced evidence for the existence of layers of originally tropospheric air in the midlatitude LS (Hintsala et al. 1998; Vaughan and Timmis 1998), and stratospheric intrusions into the troposphere (Danielsen 1968; Stohl and Trickl 1999).

In order to assess the impact of STE on atmospheric chemistry, STE mass fluxes and geographical distributions of STE must be calculated from global meteorological data. Traditionally Eulerian diagnos-

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DOI: 10.1175/BAMS-84-11-1565

In final form 20 February 2003
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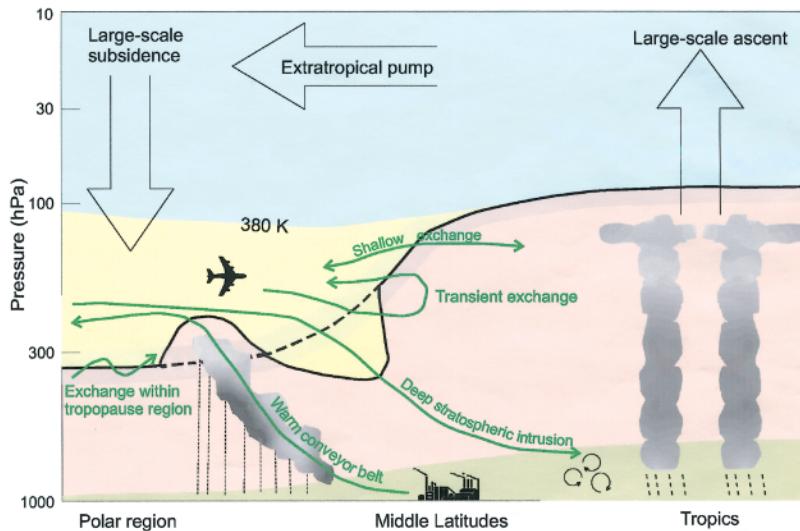


FIG. 1. Global aspects of STE from Holton et al. (1995), with our Lagrangian STE concept superimposed. The average position of the tropopause is shown by the thick black line, with shaded regions on either side representing the tropopause region. The blue region is the “overworld,” in which isentropes (above the 380-K isentropic surface) lie entirely in the stratosphere, the yellow region is the lowermost stratosphere where isentropic surfaces cross the tropopause, the pink region is the free troposphere, and the brown region is the ABL. Broad arrows show transport by the global-scale circulation. Green trajectories illustrate our new concept. Pink and yellow bulges near the warm conveyor belt and the deep stratospheric intrusion indicate strong perturbation of the tropopause from its average position (dashed). Note that the pressure is not to scale.

tics (Wei 1987) have been used that estimate the air motion relative to the (moving) tropopause. However, existing Eulerian estimates of STE are highly sensitive to intrinsic parameters (Wirth and Egger 1999; Gettelman and Sobel 2000) and are in poor agreement with each other (recent global estimates differ by up to a factor of 2 at least; cf. Gettelman and Sobel 2000).

Even accurately calculated cross-tropopause mass fluxes are insufficient to characterize relevant aspects of STE. In particular it is of major importance to also consider the pathways of exchange of the air parcels and to determine their vertical penetration and residence time in the troposphere and LS (Stohl 2001; Wernli and Bourqui 2002). Such an approach was emphasized in the framework of the Influence of Stratosphere–Troposphere Exchange in a Changing Climate on Atmospheric Transport and Oxidation Capacity (STACCATO), a project involving 13 research groups, which was funded by the European Union. The project’s detailed results are published in a special section of the *Journal of Geophysical Research* (2003, Vol. 108, no. D12). Here, we present an introduction to the new Lagrangian concepts developed during STACCATO and discuss their implications.

The pathways and time scales of STE can best be investigated with a Lagrangian approach (cf. Fig. 1). For instance, STE limited to the tropopause transition region (defined here as the layer having potential vorticity between 1 and 4 pvu) has moderate impact on atmospheric chemistry (because differences in the chemical composition within such a shallow layer are relatively small for many substances). The same holds for transient exchange of stratospheric air (i.e., air that returns to the LS after residing only a short time in the troposphere), which has little opportunity to mix with tropospheric air. On the other hand, deep exchange events that transport air irreversibly from the potentially polluted atmospheric boundary layer (ABL) upward into the LS (see Fig. 2), or vice versa, within a short time interval, have a much greater impact. Note also that rapid deep descent of stratospheric air can be associated with severe weather

(Browning and Reynolds 1994; Browning and Golding 1995; Goering et al. 2001) and cause ozone peaks at the surface (Stohl et al. 2000). Furthermore, due to the nonlinear dependence of chemical processes on mixing (Esler et al. 2001), atmospheric composition is affected differently by deep versus shallow exchange events, because only deep events can bring together air masses of strongly different composition that can subsequently mix. Thus, if the subset of deep exchange events has characteristics (geographical location, seasonal cycle) that are different from the whole set of exchange events, existing STE estimates, which are dominated by shallow events (those that do not extend beyond the tropopause transition region), will not characterize the effects of deep STE correctly.

In this study, two Lagrangian models driven with reanalysis data from the European Centre for Medium-Range Weather Forecasts (ECMWF) are used to establish two complementary 15-yr climatologies of STE. The two approaches differ and their results allow different aspects of STE to be highlighted. The overall spatial patterns of STE fluxes of the two climatologies have been intercompared and agree

well. Here, we shall concentrate on the time scales and the depths of STE events. The term “exchange” in STE refers to two-way transport. To clearly distinguish between upward and downward cross-tropopause transport, upward transport will be referred to as troposphere-to-stratosphere transport (TST), and downward transport as stratosphere-to-troposphere transport (STT). After a careful discussion of the characteristics of deep exchange events, the impact of STT events on the seasonality of tropospheric ozone will be examined, in particular, to investigate whether deep STT may be responsible for the late springtime peak in lower-tropospheric ozone concentrations typically observed at remote sites in middle and high latitudes (Monks 2000).

DATA AND MODELS. The 15-yr ECMWF reanalysis dataset (ERA-15; Gibson et al. 1997) is a comprehensive global meteorological dataset covering the

period 1979–93 with a 6-hourly analysis frequency. Thirty-one model levels are available in the vertical direction, of which the highest lies at 10 hPa, and a $1^\circ \times 1^\circ$ resolution is provided in the horizontal, derived from spectral fields with a resolution of T106. This dataset constitutes a sophisticated “mixture” of all globally available meteorological observations and state-of-the-art short-term numerical forecasts.

FLEXPART is a Lagrangian particle model that calculates the transport and dispersion of nonreactive particles and includes parameterizations for turbulence and convection and is driven, in this experiment, by the ERA-15 model-level data. FLEXPART has been validated with data from large-scale tracer experiments (Stohl et al. 1998) and performs well in comparison with other models (Meloan et al. 2003). It has been used for studying the intercontinental transport of ozone (Stohl and Trickl 1999), the advection of Canadian forest fire emissions to Europe (Forster et al. 2001), and the dispersion of aircraft emissions in the stratosphere (Forster et al. 2003), for example.

At the beginning of the simulation 500,000 particles, each carrying an equal fraction of the total atmospheric mass, were distributed homogeneously throughout the atmosphere and were then tracked over the course of 15 yr. They were free to wander across hemispheres and between the stratosphere and troposphere. Only small deviations from a correct atmospheric mass distribution were observed, even after 15 yr of simulation. Each particle carried several clocks. For instance, a stratospheric clock that was set to zero in the stratosphere started running when the particle entered the troposphere and was reset to zero upon the particle’s re-entry into the stratosphere. The tropopause is defined in FLEXPART as a dynamical tropopause poleward of 30° , represented by the potential vorticity (PV) surface of 2 pvu

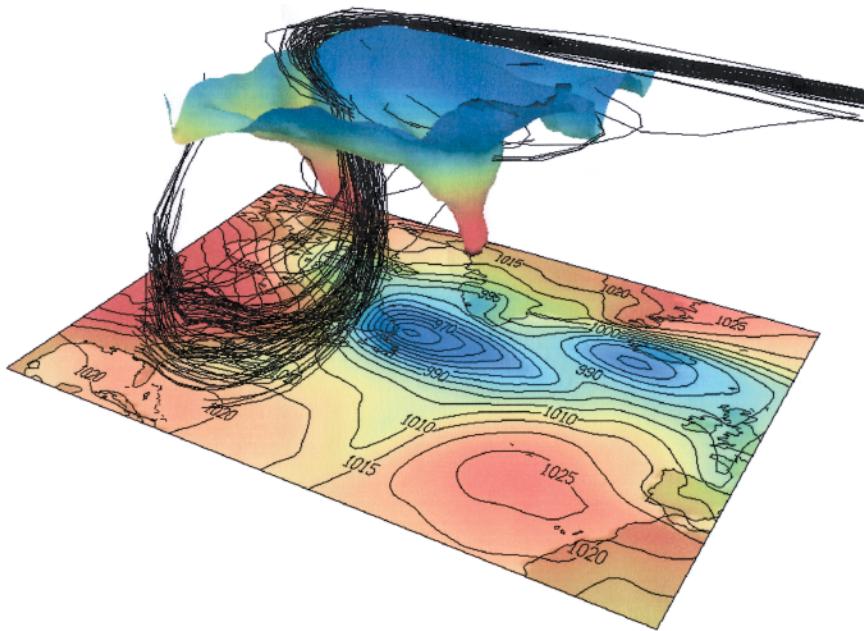


FIG. 2. Illustration of a deep exchange event. This event, occurring on 25 Feb 1982, transported potentially polluted ABL air from the North American east coast into the LS within 4 days. The depicted region covers the North Atlantic ocean between North America, Greenland, and the European west coast (bright red colors for land areas). The rapid ascent of the exchange air parcels (trajectories are shown as black lines) occurs in the warm sector of an intense surface low pressure system situated in between New Foundland and Greenland (surface colors indicate sea level pressure, low values are blue). The tropopause crossing happens in a region characterized by an elevated, cold tropopause (three-dimensional strongly deformed surface, colored according to temperature; blue is for low temperatures). In the LS the air parcels turn anticyclonically and move rapidly eastward. The moist ascent of the air parcel trajectories in the troposphere is typical for the so-called warm conveyor belt flows associated with extratropical cyclones and is responsible for the major portion of the system’s cloud and precipitation structures.

(where $1 \text{ pvu} = 10^{-6} \text{ K m}^2 \text{ kg}^{-1} \text{ s}^{-1}$) and a thermal tropopause equatorward of 20° , which is the lower boundary of a layer with a thickness of 2 km, in which the temperature gradient is lower than 2 K km^{-1} (Hoinka 1997). Between these latitudes, the two definitions are linearly interpolated.

Other clocks were employed in FLEXPART to characterize tropospheric air and ABL air. The clocks were used to establish age spectra of both the mass fluxes across several control surfaces and the mixing ratios of ABL, tropospheric, and stratospheric air. Fifteen age classes, ranging from 6 to > 1 yr, were used. More details on how FLEXPART was set up for this experiment are given by James et al. (2003a). For more general information on FLEXPART, see information online at www.forst.tu-muenchen.de/EXT/LST/METEOR/stohl/flexpart.html.

The Lagrangian analysis tool (LAGRANTO) calculates three-dimensional kinematic trajectories and tracks their several physical and dynamical parameters (including PV) (Wernli and Davies 1997). It has been compared to other frequently used trajectory packages (Stohl et al. 2001) and applied to different aspects of tropospheric and stratospheric dynamics. These include, for instance, the objective analysis of conveyor beltlike moist airflows (Wernli 1997) and the systematic investigation of stratosphere ozone profiles in midlatitudes (Koch et al. 2002).

The setup for the STE calculations is described by Wernli and Bourqui (2002) and Sprenger and Wernli (2003). The same tropopause definition is used as in FLEXPART. For the 15-yr period, 700,000 trajectories were calculated daily, starting on a regular grid covering the Northern Hemispheric tropopause region. The temporal evolution of PV along the trajectories was used to identify significant cross-tropopause exchange events. “Deep exchange” refers to the category of exchange events where the trajectories’ largest pressure value exceeds 700 hPa.

The FLEXPART and LAGRANTO models differ in a number of ways, such that their STE results complement each other well. LAGRANTO traces STE trajectories for up to 4 days only, whereas the particle clocks in FLEXPART count indefinitely and, thus, can also account for very long residence times, limited only by the total length of the integration. On the other hand, LAGRANTO stores indefinitely the exact paths of all air mass trajectories that cross the tropopause, whereas FLEXPART only stores a derivative of this information relating to mass fluxes across prespecified surfaces and concentration field snapshots in conjunction with tracer age. In particular, the “origin–destination” concept of STE trajectories avail-

able in LAGRANTO on a 3D-grid is not possible with FLEXPART.

RESULTS. *The residence time of STE events.* First, we present evidence that most of the STE events are rather transient. With FLEXPART, we calculated the age spectra of the return flux of stratospheric air into the LS, defined as the time stratospheric air spent in the troposphere before returning to the LS, and, vice versa, for tropospheric air. There are large upward ($372.3 \times 10^{17} \text{ kg yr}^{-1}$) and downward ($376.7 \times 10^{17} \text{ kg yr}^{-1}$) mass fluxes in the extratropical Northern Hemisphere. Thus, the comparatively small annual net mass flux ($4.4 \times 10^{17} \text{ kg yr}^{-1}$) is downward, in agreement with previous budget estimates (Appenzeller et al. 1996; see also James et al. 2003a, for a detailed comparison). According to FLEXPART the residence time of more than 90% of the STT (TST) particles in the troposphere (stratosphere) is less than 6 h, before returning to the stratosphere (troposphere). STT (TST) particles that stayed more than 4 days in the troposphere (stratosphere) contribute only $7.8 (4.2) \times 10^{17} \text{ kg yr}^{-1}$, or approximately 1%–2% to the total FLEXPART fluxes. Despite their low magnitude, these exchange events are very important, allowing sufficient time for mixing and being transported farthest away from their source. In contrast, the majority of the exchange events with short residence times are not transported far from the tropopause. Their quantification is highly sensitive to model parameterizations, input data, interpolation errors, etc., and, therefore, contains a lot of noise. However, with our Lagrangian tools, we can separate out the more significant exchange events by specifying an appropriate minimum residence time of STT (TST) particles in the troposphere (stratosphere), whereas Eulerian methods consider all events the same, thus, suffering more from high levels of noise (e.g., Gettelman and Sobel 2000).

Characteristics of deep exchange events. Next we focus only on air parcels with residence times greater than 4 days and investigate the geographical and seasonal variability of deep exchange events that are associated with rapid transport (within 1–4 days) between the low troposphere (below 700 hPa) and the LS. Results from the LAGRANTO calculations for the winter season reveal distinct patterns of low-tropospheric “origins” of deep cross-tropopause transport into the LS and “destinations” of deep exchange in the reverse direction (Fig. 3a). Preferred low-tropospheric source regions for rapid transport into the LS (green contours) are the entrance regions of the midlatitude

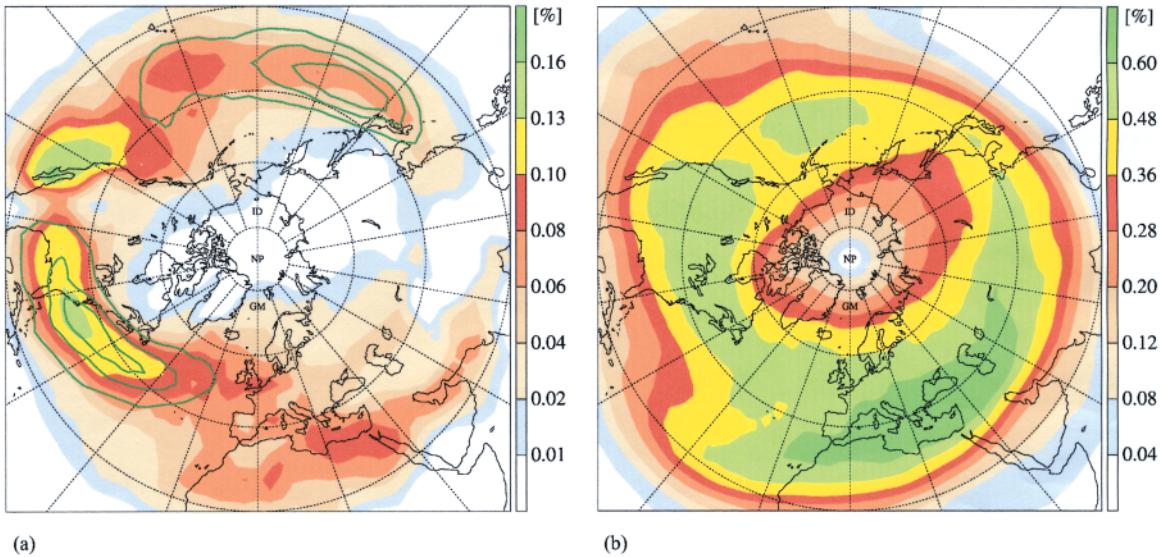


FIG. 3. Winter (Dec–Feb) climatology (1979–93). (a) Low-tropospheric “destinations” of deep stratospheric intrusions (in colors) and “origins” of deep cross-tropopause transport into the LS (green lines for 0.04%, 0.08%, and 0.12%). Values are in % and indicate the probability that a low-tropospheric air parcel ($p > 700$ hPa) was transported downward from the LS during the previous 4 days or will be transported upward into the LS during the next 4 days. (b) The upper-tropospheric “destinations” ($p < 500$ hPa) of shallow stratospheric intrusions.

North Pacific and Atlantic storm tracks. More specifically, they are located over the Pacific between Japan and the date line, and over the eastern United States and western Atlantic. This indicates that the high-emission areas in Japan and the eastern United States are the most important source regions for rapid transport of pollutants into the LS during winter (cf. the example shown in Fig. 2). The reason for this spatial distribution is the high frequency of warm conveyor belts (Browning 1990; Wernli and Davies 1997) associated with extratropical cyclones in these regions (Stohl 2001; Eckhardt et al. 2003). Similarly, there are preferred areas toward the end of the Pacific storm track and the beginning of the Atlantic storm track where deep downward exchange influences the low troposphere (color shading in Fig. 3a) along the U.S. west coast, in the western North Atlantic (a preferred region for cyclone formation), and (albeit weaker) in the Mediterranean region. It is in these areas that stratospheric intrusions are most likely to impact directly on the surface O_3 budget. In contrast, total STT that includes, and is dominated by, shallow events shows much less geographical variations: Fig. 3b shows the frequency distribution of STT air parcels in the upper troposphere ($p < 500$ hPa) and reveals a maximum in the latitude band from 30° to 60° N with comparatively little zonal variability.

Furthermore, the seasonality of deep STT in the extratropics differs strongly from that of the total STT.

First, the amplitude of the seasonal cycle of downward exchange (integrated over the Northern Hemisphere) is much more pronounced for deep than for shallow events. It has a distinct winter maximum and summer minimum; for instance, in May (July) deep STT amounts to 40% (10%) of the January value (Sprenger and Wernli 2003). Second, deep STT affects most areas in the Northern Hemisphere less frequently during spring than winter, except for the U.S. West Coast and northeastern China.

Implications for surface ozone. An ozone maximum is observed at many background surface measurement stations in the Northern Hemisphere in spring, typically in late April or May (Monks 2000; Harris et al. 1998). A qualitative comparison of the seasonal cycles of the surface ozone mixing ratios with those of net cross-tropopause fluxes of ozone, which both show a spring maximum, has often led to the conclusion that surface ozone and STT are strongly related to each other. However, this argument is based on the assumption that net cross-tropopause fluxes represent the influence of STT at the surface, which is not the case (as indicated by the discussions above and confirmed by the FLEXPART analyses). Indeed, recent chemistry transport model calculations suggest a very small contribution of stratospheric ozone at the surface. For instance, Fusco and Logan (2003) estimate that approximately 30%–50% of the ozone in the up-

per troposphere originates in the stratosphere throughout the year, whereas only 10% of the surface ozone is of stratospheric origin in winter and spring, and even less in summer. We find that these large vertical differences in the stratospheric contribution to tropospheric ozone are partly due to the relative scarcity of deep STT events compared to the full set of STT events. Furthermore we argue that it is difficult to explain a springtime maximum of surface ozone with a stratospheric source, even on the basis of tropospheric dynamics alone, because the frequency of deep STT events decreases strongly from winter to spring (Fig. 4).

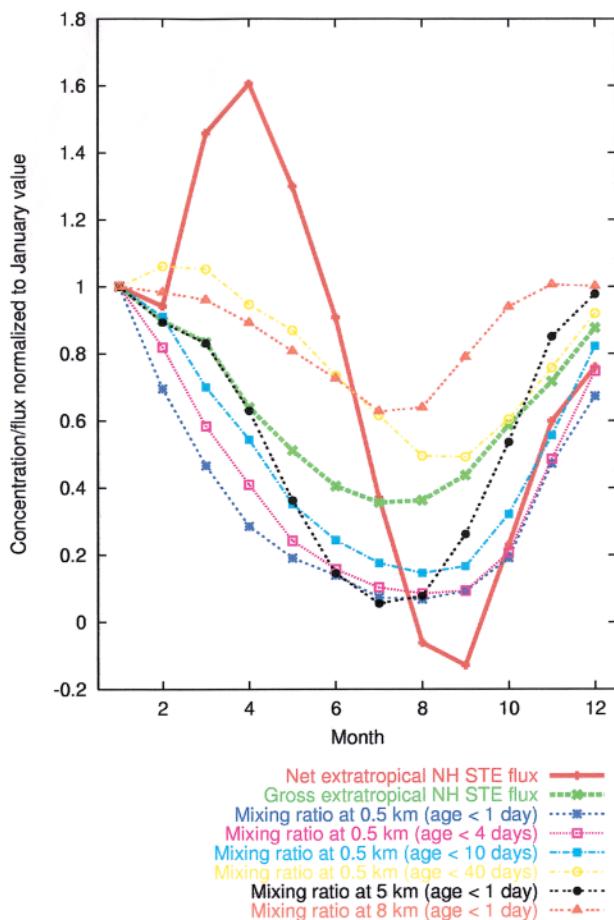


FIG. 4. Seasonality of STE. The figure shows the average (1979–93) annual variations of the net and gross downward extratropical (poleward of 21°N) fluxes of air across the tropopause, of the mixing ratios of stratospheric air at 500 m MSL for four different residence times (ages) in the troposphere, and of the mixing ratios of stratospheric air with tropospheric residence time of less than 1 day at 5 and 8 km, respectively. All values have been divided by the respective Jan value and are in relative units. Mixing ratios are averaged over the latitude belt 45°–60°N.

In agreement with other studies (Appenzeller et al. 1996) the FLEXPART results show the largest extratropical net cross-tropopause mass flux in April, but the gross downward STT flux already maximizes in January and drops by 50% by May (Fig. 4). Close to the surface, the mixing ratio of stratospheric air that recently entered the troposphere decreases even more between January and May. For STT with tropospheric residence times less than 1 day (10 days) our calculations indicate an 80% (65%) drop in surface mixing ratios between January and May, and a further decrease toward summer. This is in agreement with a strongly reduced frequency of observed deep stratospheric intrusions during summer (Stohl et al. 2000). The seasonal variation becomes less pronounced only for residence times longer than 20 days. However, at higher levels (5–8 km, cf. Fig. 4), the annual cycle is much weaker than at the surface, even for short tropospheric residence times.

To investigate how the seasonal variations of STT can be expected to impact on the seasonal cycle of ozone at various altitudes in the troposphere, based on FLEXPART, we prescribe a mean seasonal cycle of ozone mixing ratios for the northern midlatitudes in the lower stratosphere, using monthly values of its linear relationship with PV derived from observations (Stohl et al. 2000). The decay time constant of ozone in the troposphere is highly variable and depends strongly on sunlight and humidity levels. We test the sensitivity of our results to some different decay time scales (7, 15, and 30 days, respectively), which span typical values obtained from chemical models (Liu et al. 1987). Local chemical ozone lifetimes in the wintertime polar upper troposphere may be longer than 30 days, but particles do not remain solely in the upper troposphere, and, thus, the above range is appropriate for effective ozone lifetimes, integrated along an air parcel's path in the troposphere. The total stratospheric ozone contribution at various tropospheric altitudes, integrated over all STT flows, is shown in Fig. 5. It increases strongly with an increasing decay time scale. At a decay time scale of 30 days, the stratospheric ozone contribution exceeds 30 ppb in the lower troposphere in winter, which is certainly too high, while the seasonal cycle maximum in March–April is fairly constant with altitude, at least up to 6 km (note that 8 km may not always be in the troposphere, especially in wintertime troughs). However, for the much shorter decay time scale of 7 days, the seasonal cycle shifts with altitude from a distinct winter maximum below 2 km to a March maximum in the middle troposphere. At all decay time scales, the annual minima of ozone with a strato-

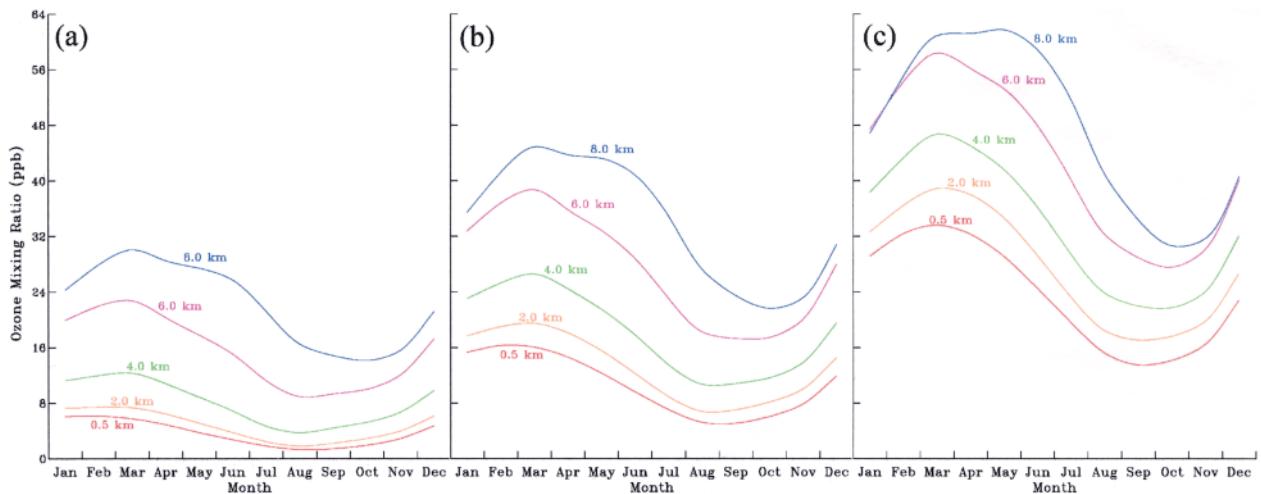


FIG. 5. Mean seasonal cycle of the mixing ratio of ozone of stratospheric origin in the troposphere in the northern midlatitudes (30° – 60° N) at altitudes of 0.5, 2, 4, 6, and 8 km MSL, respectively, based on assumptions of the ozone decay time scale of (a) 7, (b) 15, and (c) 30 days, derived from the 15-yr climatology of STT with FLEXPART.

spheric origin shift to later in the year with increasing altitude, typically from August, at low altitudes, to October, in the upper troposphere.

Although we have accounted for chemical and deposition processes in a simplified way by assuming a constant tropospheric lifetime of ozone, the calculated ozone concentrations compare well with the contribution of stratospheric ozone in full chemistry model simulations (Lelieveld and Dentener 2000; Fusco and Logan 2003). In reality, the chemical lifetime of ozone decreases from winter to spring, and, thus, the decrease from February to May of the contribution of stratospheric ozone at the surface will be even steeper than in our estimate. Thus, we conclude that ozone of stratospheric origin has a late winter maximum at the surface, and that the observed spring ozone maximum must be mainly due to photochemical production, in agreement with chemistry model studies (Lelieveld and Dentener 2000; Fusco and Logan 2003). However, without STT the spring ozone maximum would be shifted to a somewhat later date, and STT may still be the reason for the spring ozone maximum in the upper troposphere. We do not claim that our study of the effect of STT on tropospheric ozone is more accurate than studies using chemistry transport models. However, it does explain the relatively small contribution of ozone of stratospheric origin to surface ozone concentrations seen in these models from a dynamical point of view with the scarcity of deep STT events compared to the full set of STT events.

SUMMARY. A new perspective on STE has been established through complementary Lagrangian particle and trajectory experiments, based on 15 yr of

ECMWF global atmospheric reanalysis data, with the FLEXPART and LAGRANTO models, respectively. To achieve this, the importance of distinguishing between deep STE, which bring into contact air from the (potentially polluted) boundary layer and the lower stratosphere via fast ascent of tropospheric, or fast descent of stratospheric, air masses, and shallower STE, which is transient in nature and only influences layers close to the tropopause, has been demonstrated. In particular, deep exchange is seen to have different characteristics, both in terms of its preferred geographical locations and the phase and amplitude of its seasonal cycle, relative to the full set of all exchange events.

Most cross-tropopause exchange is shallow and much of the air involved returns across the tropopause within 24 h. This is true in both directions. Focussing on downward exchange, in particular, only about 5% of the mass of the troposphere is composed of stratospheric air, which was last in the stratosphere even up to 4 days earlier. Of this fraction, less than 10% is to be found at altitudes below 3 km (James et al. 2003b), indicating that deep STT, involving direct descent on synoptic time scales, is not the normal case but occurs within specific meteorological systems, mainly near the Atlantic and Pacific storm-track entrance and exit regions. However, the seasonal cycle of this deep STT is so substantially different from that of shallow STT that it begs to be taken into separate account. Deep STT has a strong winter maximum, whereas shallow STT has a weak amplitude seasonal cycle, which even shifts to a spring maximum for air originating above the LS (James et al. 2003b). The influence of this difference on the seasonal cycle of

stratospherically derived ozone in the troposphere has been demonstrated with the FLEXPART results based on empirical assumptions. Although ozone concentrations in the LS have a spring maximum, which are in turn reflected in a spring ozone maximum in the upper troposphere, ozone in the lower troposphere brought down by deep STT has at best a late winter maximum. This suggests that STT cannot be the cause of the late springtime maximum of ozone in the lower troposphere indicated by observations.

ACKNOWLEDGMENTS. This study was part of the EU project STACCATO under Contract EVK2-CT-1999-00050 (respectively, BBW 99-0582-2 for H. Wernli, and M. Sprenger). We thank the STACCATO community, particularly H. C. Davies, G. J. Roelofs, and V. Wirth, for valuable discussions, and the German and Swiss Weather Service and ECMWF for access to the reanalysis data.

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