Stratosphere-troposphere exchange: A review, and what we have learned from STACCATO

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[1] This paper provides a review of stratosphere-troposphere exchange (STE), with a focus on processes in the extratropics. It also addresses the relevance of STE for tropospheric chemistry, particularly its influence on the oxidative capacity of the troposphere. After summarizing the current state of knowledge, the objectives of the project Influence of Stratosphere-Troposphere Exchange in a Changing Climate on Atmospheric Transport and Oxidation Capacity (STACCATO), recently funded by the European Union, are outlined. Several papers in this *Journal of Geophysical Research–Atmospheres* special section present the results of this project, of which this paper gives an overview. STACCATO developed a new concept of STE in the extratropics, explored the capacities of different types of methods and models to diagnose STE, and identified their various strengths and shortcomings. Extensive measurements were made in central Europe, including the first monitoring over an extended period of time of beryllium-10 (^{10}Be), to provide a suitable database for case studies of stratospheric intrusions and for model validation. Photochemical models were used to examine the impact of STE on tropospheric ozone and the oxidizing capacity of the troposphere. Studies of the present interannual variability of STE and projections into the future were made using reanalysis data and climate models. *INDEX TERMS:* 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 3362 Meteorology and Atmospheric Dynamics: Stratosphere/troposphere interactions; *KEYWORDS:* Brewer-Dobson circulation, trajectories, Lagrangian model, reanalysis, tropopause

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1. Stratosphere-Troposphere Exchange: A Review

1.1. Tropopause

[2] The troposphere and the stratosphere have fundamentally different characteristics (e.g., static stability, chemical composition) and, thus, air mass exchange between these two regions of the atmosphere is of great interest. They are separated by the tropopause, which, according to the World Meteorological Organization, is defined as the lowest level at which the temperature lapse rate decreases to 2 K km^{-1} or less, and the lapse rate averaged between this level and any level within the next 2 km does not exceed 2 K km^{-1} [*World Meteorological Organization* (*WMO*), 1986; *Hoinka*, 1997]. In the tropics, the tropopause roughly

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coincides with the 380 K potential temperature surface at a height of 15–18 km [*Holton et al.*, 1995; *Highwood and Hoskins*, 1998; *Seidel et al.*, 2001]. It slopes downwards toward the poles, where it may be as low as 6–8 km at a potential temperature of 290–320 K.

[3] On large space and timescales, in the extratropics the WMO tropopause corresponds rather well to a surface of constant potential vorticity (PV) [e.g., *Reed and Danielsen*, 1959; *Danielsen*, 1968; *Hoerling et al.*, 1991], but there exist systematic differences on smaller scales [*Wirth*, 2000]. A PV-based dynamical definition of the tropopause is for many purposes more attractive than the conventional WMO thermal definition because PV is conserved under adiabatic and frictionless conditions [*Ertel*, 1942] and emphasizes the material-surface nature of the tropopause [*Hoskins et al.*, 1985; *Wirth*, 1995b]. Tropopause PV values used in the literature range from approximately 1.6 pvu to 3.5 pvu, with a value of 2 pvu used most often (PV units, 1 pvu = 10^{-6} m^2 s^{-1} K kg^{-1}). Therefore it can account for tropopause folds, which appear as multiple stable layers in vertical temperature profiles, although they represent the same folded discontinuity surface [*Bjerknes and Palmén*, 1937]. Another tropopause definition in use is based on ozone (O_3), which shows a sharp transition from low concentrations in the troposphere to high concentrations in the stratosphere. The O_3 tropopause is usually somewhat lower than the thermal one [*Bethan et al.*, 1996].

[4] There exist several climatologies of global tropopause heights [e.g., *Hoinka*, 1998; *Nielsen-Gammon*, 2001] and of parameters at the tropopause [*Hoinka*, 1999; *Seidel et al.*, 2001] based on different data sets and using different tropopause definitions. However, substantial difficulties remain in determining the height of the tropopause, particularly at polar latitudes [*Highwood et al.*, 2000; *Zängl and Hoinka*, 2001]. A certain degree of subjectivity as to which definition is used remains, and the associated sensitivity can be substantial in cases when the tropopause is less sharp than normal [*Wirth*, 2000; *Zängl and Hoinka*, 2001].

[5] The tropopause height is controlled by many interacting processes, such as radiation, the effect of baroclinic eddies [*Egger*, 1995] and the effect of the stratospheric mean meridional circulation. *Thuburn and Craig* [1997], using GCM simulations, demonstrated that the tropopause height is highly sensitive to changes of the temperature at the Earth's surface. Tropospheric warming may thus lead to an increase of the average tropopause height, although the mechanisms are not yet adequately understood. Indeed, *Steinbrecht et al.* [1998] have recently shown that the tropopause at Hohenpeißenberg in Germany has moved up by 150 m per decade over the last 30 years. They found that this increase is consistent with the observed trend of temperature at the 500 hPa level and is thus likely to be linked to greenhouse-gas-induced warming. However, the stratospheric cooling resulting from stratospheric O_3 losses also reduces the vertical stability of the upper troposphere/lower stratosphere (UTLS) system [*Rind and Lonergan*, 1995]. *Highwood et al.* [2000] related an observed increase in Arctic tropopause heights to stratospheric cooling.

1.2. STE Aspects of the General Circulation

[6] Figure 1, taken from the detailed review of *Holton et al.* [1995], illustrates the global aspects of STE. Viewed from a global perspective, the circulation in the upper troposphere and the stratosphere can be described by an organized upwelling from the troposphere to the stratosphere in the tropics [*Plumb*, 1996; *Mote et al.*, 1996], transport to the extratropics in the stratosphere [*Waugh*, 1996], and a downward mass flux from the stratosphere to the troposphere in middle and high latitudes [*Holton et al.*, 1995]. In the zonally averaged sense, this corresponds to the Brewer-Dobson circulation [*Brewer*, 1949] that is driven nonlocally by the Rossby and gravity wave breaking in the extratropical middle atmosphere. It acts as a "suction pump," withdrawing air from the tropical upper troposphere, pushing it poleward and finally downward. In the stationary limit, this leads to the so-called downward control principle [*Haynes et al.*, 1991]. Recently, it has become clear that the wave-breaking must extend into the subtropics to account for the tropical upwelling [*Plumb and Eluszkiewicz*, 1999].

[7] According to *Hoskins* [1991], it is useful to distinguish between different regions of the lower stratosphere: the overworld, the middleworld, and the underworld. The overworld is the region above the (approximately) 380 K isentropic surface and lies entirely in the stratosphere. Transport from the overworld to the troposphere is slow because the air must cross isentropic surfaces, which requires diabatic cooling. In the middleworld, isentropic surfaces lie partly in the troposphere (in the tropics) and partly in the (lowermost) stratosphere (at high latitudes). Therefore, relatively fast transport between the stratospheric part of the middleworld and the troposphere is enabled along the isentropes, which can occur in both directions [*Chen*, 1995]. The underworld lies entirely in the troposphere. Transport from the underworld into the stratosphere can only occur across the isentropes through diabatic heating.

[8] As isentropes intersect the tropopause in the middleworld, it has been argued [e.g., *Holton et al.*, 1995] that the tropopause itself is not necessarily the best suited control surface for measuring STE fluxes, but that the 380 K control surface is more convenient, because the transport across this surface can be described on the largest scales using the downward control principle [*Rosenlof*, 1995]. This avoids the need for detailed knowledge about the smaller-scale synoptic and mesoscale processes that are relevant at the tropopause. If, in addition, the seasonal variation of the mass of the lowermost stratosphere is estimated, this approach can be extended to calculate the monthly mean net mass flux across the extratropical tropopause [*Appenzeller et al.*, 1996b]. This net mass flux shows a distinct seasonal cycle with a maximum in late spring and a minimum in fall in the Northern Hemisphere, and a maximum in midwinter in the Southern Hemisphere (Figure 2). This is in agreement with a Northern Hemispheric spring maximum near the surface of fallout from nuclear bomb testing, which had a stratospheric reservoir during the 1950s and 1960s [*Fry et al.*, 1960; *Staley*, 1982].

[9] The net mass flux across the extratropical tropopause calculated by *Appenzeller et al.* [1996b] using global principles is an important constraint on estimates obtained with other techniques. However, global estimates have three important limitations: They are spatially averaged (usually over the hemisphere), temporally averaged (usually over a month), and they only account for the net flux. The averaging makes it impossible to diagnose spatial and temporal varia-

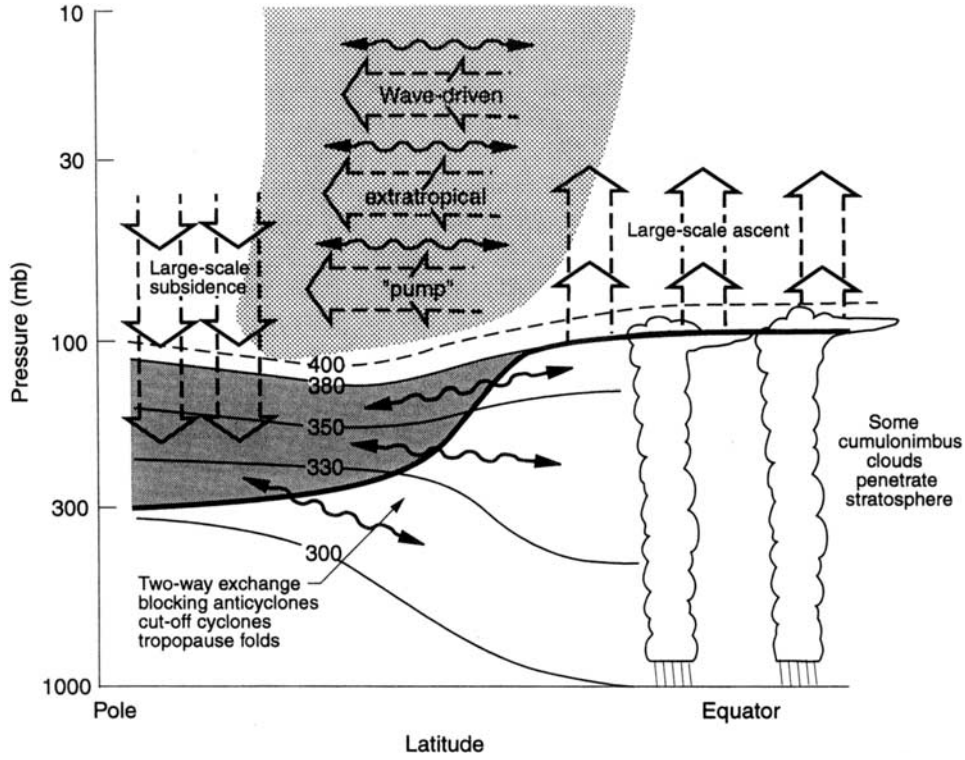

Figure 1. Dynamical aspects of stratosphere-troposphere exchange (taken from *Holton et al.* [1995]; their Figure 3). Be aware that, according to present knowledge, tropical cumulonimbus clouds do not reach into the stratosphere. See the main text for more details.

bility of STE. For instance, the lifetime of O_3, which can be transported from the stratosphere to the troposphere, varies significantly with latitude and season. To study the impact of STE on the tropospheric O_3 budget it is thus necessary to know where and when stratospheric air masses are transported into the troposphere. The fact that only net fluxes can be calculated renders global estimates completely inadequate for trace species (e.g., nitrogen oxides, O_3) that have sources (e.g., aircraft emissions, chemical processes) or sinks (e.g., chemical processes) in the lowermost stratosphere. Note in particular that for these species the seasonal cycle of the cross-tropopause flux can differ from that obtained by *Appenzeller et al.* [1996b] and seen in surface concentrations of fallout from nuclear bomb testing. We argue below that, to characterize the effects of STE more completely, it is important to consider not only the cross-tropopause fluxes, but also the tropospheric (and lower stratospheric) transport preceding or following the crossing of the tropopause. Central to the STACCATO project was the idea that there may be systematic differences between exchange events of a ''deep'' and ''shallow'' nature, i.e., between events that connect points outside the tropopause region and such that only connect points within the tropopause region.

1.3. STE in the Tropics

[10] The tropopause is highest and temperatures at the tropopause are lowest in the tropics [*Highwood and Hoskins*, 1998; *Seidel et al.*, 2001]. Air rises in the Hadley cell from the surface to the upper troposphere. Above the Hadley cell, the Brewer-Dobson circulation [*Brewer*, 1949] causes further ascent in the stratosphere. On passing through the cold regions of the tropical tropopause the air is freeze-dried and retains the saturation mixing ratio corresponding to the low temperatures. The fact that the whole stratosphere (also the extratropical one) is extremely dry testifies that most of the air enters the stratosphere via this tropical pathway. A further piece of evidence is that in the tropics the signal of the water vapor mixing ratio at the tropopause level is maintained by the air parcel as it rises through the stratosphere for up to 18 months. In this way, the seasonal variation of the water vapor mixing ratio (caused by seasonal differences in tropopause temperatures) is recorded as a layering of water vapor concentrations (the tape recorder mechanism). The layered structures travel slowly upward with time and are still observable near the 20 hPa level [*Mote et al.*, 1996; *Jackson et al.*, 1998].

[11] The details of how tropospheric air enters the stratosphere in the tropics and the scales involved are still heavily debated. The extreme dryness of the stratosphere, with water vapor mixing ratios lower than those that could be explained by the zonal mean tropical tropopause temperature, requires a mechanism that prefers pathways through the coldest regions of the tropical tropopause. However, most convective cells extend only to the level of maximum outflow of the Hadley cell at about 200 hPa, and even cumulonimbus cloud tops are normally below 140 hPa, whereas the tropical tropopause is near 100 hPa, and the coldest point in tropical temperature profiles is even higher at about 80 hPa [*Highwood and Hoskins*, 1998]. Thus, *Newell and Gould-Stewart* [1981] suggested that the air enters the stratosphere primarily by slow ascent in restricted regions, termed stratospheric fountains, where tropopause

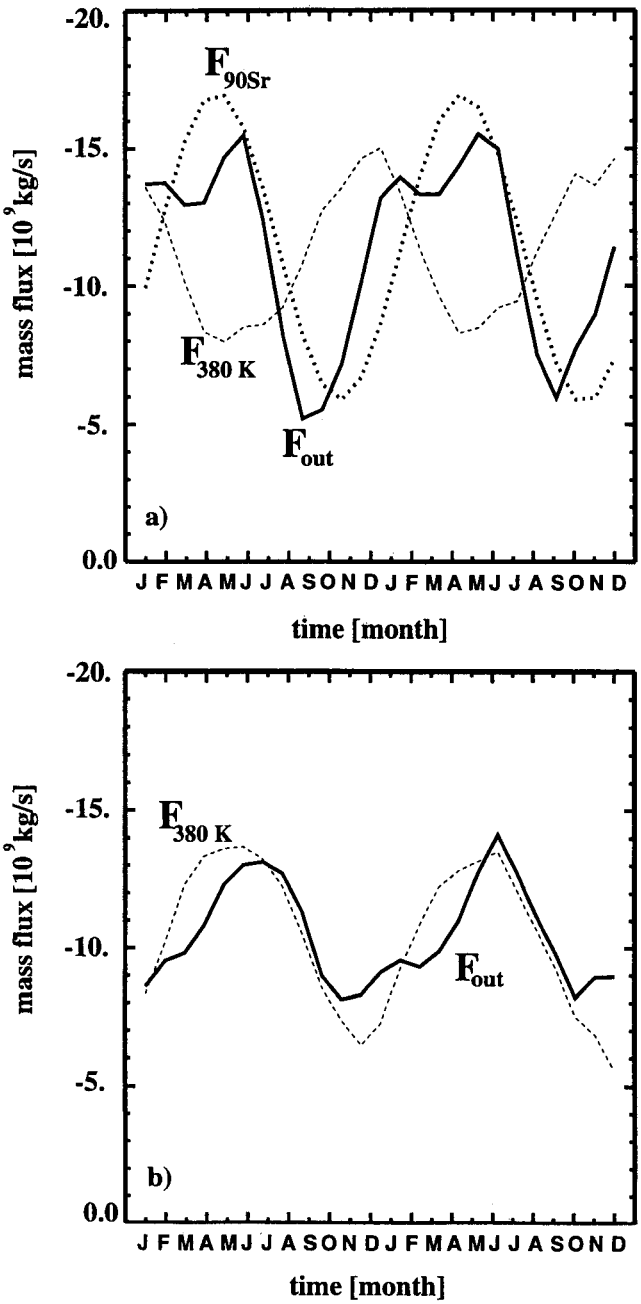

Figure 2. Annual variation of the net downward mass transport across the extratropical tropopause (F_{out}, thick plain curve), the 380 K isentropic surface (F_{380}, thin dashed curve), and mass flux according to ^{90}Sr measurements (F_{90Sr}, dotted line). Shown are the two years 1992 and 1993. (a) Northern Hemisphere. (b) Southern Hemisphere. (Values are given in 10^9 kg s^{-1}; negative values denote downward mass flux.) Taken from *Appenzeller et al.* [1996b] (their Figure 8).

temperatures are especially low, and only during specific seasons of the year. However, the entry of air into the stratosphere in restricted regions only has recently been questioned [*Dessler*, 1998], and it became clear that air can enter the stratosphere throughout the year [*Mote et al.*, 1996]. Other recent studies [*Michelsen et al.*, 2000; *Zhou et al.*, 2001], on the other hand, argue that this is not the case. Furthermore, slow ascent associated with the stratospheric fountain would likely cause large cirrus sheets in the tropics, which could not be observed for a long time [*Russell et al.*, 1993]. Note, though, that large subvisible cirrus clouds have been detected recently [*Jensen et al.*, 1996; *McFarquhar et al.*, 2000], which are difficult to observe. Also, several studies [*Gettelman et al.*, 2000; *Sherwood*, 2000] indicate that there is net subsidence at the tropopause in the stratospheric fountain region, not ascent. Alternatively, it may also be possible that only the very highest and coldest cumulonimbus clouds, that can penetrate the tropopause, and radiatively triggered turbulence in their cirrus anvils, determine the amount of water vapor transferred into the stratosphere [*Danielsen*, 1982], whereas the signal produced by lower clouds is erased by subsequently occurring higher clouds. Another possible dehydration process is the large-scale ascent in the tropical tropopause region [*Holton et al.*, 1995; *Holton and Gettelman*, 2001]. The dehydration here is likely to be realised by the formation of widespread (subvisible) cirrus clouds in the tropical upper troposphere. Yet another possible dehydration mechanism are gravity waves that trigger the formation of clouds in the stratosphere upwind of convective regions, from which ice particles can precipitate [*Potter and Holton*, 1995].

[12] *Highwood and Hoskins* [1998] favor the picture of a transition zone between the top of the convective outflow of the Hadley cell, and the stratospheric Brewer-Dobson circulation above the cold point, over a single tropopause surface. In this transition zone, substantial exchange with air from the extratropics can occur, further complicating the picture. *Jackson et al.* [2001] point out that most of the air at 150 hPa descends and remains in the troposphere, but a few air parcels at that level experience strong heating and can ascend into the stratosphere through the transition zone. According to *Sherwood and Dessler* [2001] this transition layer has properties similar to the entrainment zone at the top of the convective planetary boundary layer. The exact nature of the transfer of air through the transition zone, however, is still under debate.

[13] Although transport across the tropical tropopause is mostly from the troposphere to the stratosphere, episodically it can occur in the opposite direction, due to breaking Kelvin waves [*Fujiwara et al.*, 1998], or in tropical cyclones [*Baray et al.*, 1999]. Furthermore, Rossby wave breaking at the subtropical tropopause in the vicinity of the subtropical jet stream can transport stratospheric air into the tropical upper and middle troposphere [*Zachariasse et al.*, 2001]. Stratospheric air intrudes into the tropics preferably in the westerly ducts [*Waugh and Polvani*, 2000] and can trigger convection there.

1.4. STE in the Extratropics

[14] According to the downward control principle [*Haynes et al.*, 1991], one would expect that the transport in the extratropics is from the stratosphere to the troposphere. However, it applies to a control surface at the top of the lowermost stratosphere and is only partly relevant for the mass transport at the tropopause level [*Egger*, 1996; *Haynes et al.*, 1996], where other processes contribute to control STE, e.g. the relationship with the momentum transport in

the troposphere [*Juckes*, 1997, 1994]. As the isentropes intersect the tropopause in the extratropics, transport across the tropopause can occur in both directions [*Chen*, 1995] and on timescales of only a few days. However, the strong isentropic gradients of PV and many chemical substances at the tropopause show that there must be a dynamic resilience to transport along the isentropes, since otherwise the band of strong gradients at an isentropic surface around the tropopause would be destroyed [*Holton et al.*, 1995]. Indeed, using contour advection, *Kowol-Santen* [1998] found that the PV contours at the tropopause grow much slower than the PV contours both in the troposphere and in the stratosphere. This is in agreement with *Haynes and Shuckburgh* [2000], who applied a diagnostic introduced by *Nakamura* [1996]. They even proposed that the minimum in effective diffusivity at the tropopause level can be used for an alternative definition of the tropopause.

[15] There is confusion of notions in the STE literature. *Stohl* [2001] and *Wernli and Bourqui* [2002] use STE only for stratosphere-to-troposphere exchange (and TSE for troposphere-to-stratosphere exchange), which is also the case for many chemical studies in the troposphere, which refer to STE as a process causing O_3 enhancements, for instance. However, at the same time STE is used in a more general sense for the exchange of air between stratosphere and troposphere in both directions. To be as consistent as possible with previous nomenclature while removing ambiguities, we propose that STE should refer to exchange in both directions in the most general sense, whereas stratosphere-to-troposphere transport (STT) and troposphere-to-stratosphere transport (TST) should be used to refer specifically to one-way transport.

[16] Extratropical STE is associated with synoptic-scale and mesoscale processes. Due to the intermittency of these processes, STE is a staccato of strong exchange events, separated by more quiescent periods. Transport of stratospheric air into the troposphere has been observed to occur in tropopause folds, both in the vicinity of the polar jet [*Danielsen*, 1968; *Vaughan*, 1988; *Ebel et al.*, 1991; *Lamarque and Hess*, 1994; *Vaughan et al.*, 1994; *Langford et al.*, 1996; *Beekmann et al.*, 1997] and the subtropical jet [*Baray et al.*, 2000], cutoff lows [*Ebel et al.*, 1991; *Price and Vaughan*, 1992, 1993; *Ancellet et al.*, 1994; *Wirth*, 1995b], in mesoscale convective complexes [*Poulida et al.*, 1996] and thunderstorms [*Tremblay and Servranckx*, 1993], and due to breaking gravity waves [*Lamarque et al.*, 1996]. Tropopause folds and cutoff lows are especially important, because they are associated with great latitudinal displacements of the tropopause on isentropic surfaces, which facilitate the exchange of large amounts of air. Note that tropopause folds and cutoff lows have partly equivalent meaning: the former emphasize the vertical structure in which a stratospheric intrusion may occur, the latter the horizontal one. Often, a tropopause fold is formed during the development of a cutoff low.

[17] Once in the extratropical troposphere, stratospheric air is stirred quasi-adiabatically with the surrounding tropospheric air in large-scale cyclonic and anticyclonic disturbances [*Mahlmann*, 1997]. Typically, the intruding stratospheric air forms filamentary structures, which are observed in various contexts. For instance, they appear as laminae in ozone profiles [*Dobson*, 1973; *Reid and Vaughan*, 1991], as filamentary features on water vapor satellite images [*Appenzeller and Davies*, 1992; *Wirth et al.*, 1997], they are evident by ozone lidar measurements [*Stohl and Trickl*, 1999] and have been detected by in situ aircraft measurements [*Reid and Vaughan*, 1993; *Balluch and Haynes*, 1997; *Newell et al.*, 1999]. These filaments cascade down to ever smaller scales, through the action of ''chaotic advection'' [*Ottino*, 1989], and interleave with tropospheric air [*Appenzeller et al.*, 1996a]. They can subside deep into the troposphere [*Stohl and Trickl*, 1999], and such deep descent of stratospheric air can be associated with severe weather, particularly high wind speeds at the surface [*Browning and Reynolds*, 1994; *Goering et al.*, 2001]. Finally, these structures may be destroyed by turbulence and convection. However, little is known about the timescales of the mixing with tropospheric air (see below).

[18] Midlatitude transport from the troposphere to the lowermost stratosphere has been observed less frequently than transport in the opposite direction. Nevertheless, there is an increasing body of evidence for its occurrence [*Poulida et al.*, 1996; *Hintsa et al.*, 1998; *Vaughan and Timmis*, 1998; *Ray et al.*, 1999]. In these studies, the existence in the lowermost stratosphere of relatively moist layers containing tracers that unambiguously were transported from the Earth's surface via an extratropical pathway provides a posteriori evidence for the existence of these processes. In modeling studies, *Rood et al.* [1997] and *Zierl and Wirth* [1997] have shown that diabatic processes are very important for this. *Stohl* [2001] and *Wernli and Bourqui* [2002] emphasized the role of warm conveyor belts, i.e. airstreams ahead of cold fronts associated with extratropical cyclones in which strong diabatic heating by latent heat release occurs [*Browning*, 1990; *Wernli and Davies*, 1997], for rapid transfer of lower tropospheric air into the lower stratosphere. However, this midlatitude transport into the stratosphere must be seen against the background of the large-scale descent from the overworld [*Haynes et al.*, 1991] and the high stability of the lowermost stratosphere, which requires a large increase in potential temperature for further deep penetration into the stratosphere, which cannot be accomplished by radiative heating. This is the reason that, although material can be transported from the troposphere into the stratosphere in the extratropics, it usually does not reach high levels in the stratosphere and especially not the overworld. The lowermost stratosphere is a region with mostly stratospheric character, episodically spiced with tropospheric air. This has lead to the concept of a mixing zone just above the tropopause at higher latitudes, where upwelling tropospheric air is mixed with downwelling stratospheric air [*Fischer et al.*, 2000; *Zahn*, 2001; *Dethof et al.*, 2000]. The relative importance of these two sources varies, with transport from the overworld being strongest in spring, and transport from the troposphere being possibly strongest in fall [*Ray et al.*, 1999]. In the mixing zone chemical composition is highly variable, and even aerosols in this region can have tropospheric or stratospheric character, or both [*Murphy et al.*, 1998].

1.5. Diagnosis of Cross-Tropopause Transport

[19] Because global estimates of net cross-tropopause mass flux in the extratropics [*Appenzeller et al.*, 1996b] characterize extratropical STE insufficiently, alternative

methods have been developed to diagnose STE directly from the output of general circulation models (GCMs) or mesoscale meteorological models. Traditionally this has been done using a Eulerian diagnostic developed by *Wei* [1987]. However, numerical problems with the cancellation of large terms can render this method inaccurate [*Wirth*, 1995a; *Wirth and Egger*, 1999]. Particularly, its accuracy depends on the vertical coordinate system of the model applied, and actually the most reliable implementation requires PV as the vertical coordinate and knowledge of the PV budget [*Wirth and Egger*, 1999], which is not applicable in most models. For other implementations the net cross-tropopause fluxes, if averaged over great time or space scales, may be sufficiently accurate, but estimates for the instantaneous local STT or TST fluxes are very uncertain [*Kowol-Santen et al.*, 2000], as they are highly sensitive to intrinsic parameters [*Gettelman and Sobel*, 2000]. Recently, *Hall and Holzer* [2003] have shown that for advective-diffusive flows the "gross" STT and TST fluxes are actually infinite, if no residence time limits in the troposphere and stratosphere are specified, thus explaining the great sensitivity of these fluxes to the numerical implementation of the diagnostic. With too much spatial or temporal averaging, on the other hand, the advantage of having a better resolution than global estimates is almost lost. Furthermore, a recent summary of existing estimates for extratropical net STE revealed that even the net fluxes using the Wei formula may be biased toward too high values [*Gettelman and Sobel*, 2000], if compared with the *Appenzeller et al.* [1996b] global budget constraint. Lagrangian methods have only recently been applied to derive cross-tropopause fluxes [*Wirth and Egger*, 1999; *Kowol-Santen et al.*, 2000; *Stohl*, 2001; *Meloen et al.*, 2001; *Wernli and Bourqui*, 2002; *Seo and Bowman*, 2002]. Although difficulties were also noticed for Lagrangian estimates, they are more robust than most practical implementations of the *Wei* [1987] approach [*Wirth and Egger*, 1999].

[20] Other uncertainties in diagnosing cross-tropopause transport arise from the choice of the control surface, which can be either the thermal tropopause or a certain PV threshold for the dynamical tropopause [*Grewe and Dameris*, 1996; *Gettelman and Sobel*, 2000]. Furthermore, many studies in the past have worked in an isentropic framework [e.g., *Chen*, 1995; *Dethof et al.*, 2000; *Seo and Bowman*, 2001] and have neglected diabatic processes that certainly play an important role in STE.

1.6. Impact of STE on Tropospheric Chemistry

[21] Intrusions of stratospheric air into the troposphere are rich in O_3 [*Danielsen*, 1968] and, because of the long chemical lifetime (order of a month) of O_3 in the free troposphere [*Liu et al.*, 1987], contribute substantially to the tropospheric O_3 budget [*Fabian and Pruchniewicz*, 1977]. During pre-industrial times, when tropospheric O_3 concentrations were lower than today [*Volz and Kley*, 1988], STT likely made the major contribution to surface O_3 [*Levy et al.*, 1985]. STT of O_3 is also important for the oxidative capacity of the troposphere, because, in the presence of water vapor, photolysis of O_3 leads to the formation of hydroxyl radicals (STT of reactive nitrogen species is less relevant). It has been argued, though, that nowadays in situ photochemical formation of O_3 is much more important than the supply from the stratosphere [*Fishman et al.*, 1979].

[22] Although the impact of STT on tropospheric chemistry has been discussed for more than three decades now, there is still no consensus on its relative contribution to tropospheric O_3. For instance, a phenomenon that is still under debate is the spring O_3 maximum that is observed at many background surface measurement stations in the northern hemisphere, typically in late April or May [*Derwent et al.*, 1998; *Harris et al.*, 1998; *Monks*, 2000]. A qualitative comparison of the seasonal cycles of the surface O_3 mixing ratios with those of net cross-tropopause fluxes of O_3, which both show a spring maximum, has often led to the conclusion that surface O_3 and STT are strongly related to each other. However, other compounds without a stratospheric source also exhibit a springtime peak [*Penkett and Brice*, 1986], and O_3 may be produced from precursors that accumulate during the winter, because of less efficient removal processes. Furthermore, O_3 chemistry is approximately in balance between production and destruction in summer, whereas there is net production in winter and spring. This is because in winter O_3 net production can be positive at lower levels of nitrogen oxides than in summer [*Yienger et al.*, 1999].

1.6.1. Photochemical Model Studies

[23] *Roelofs and Lelieveld* [1997] estimated that O_3 originating from the stratosphere contributed about 40% on average to O_3 in the troposphere, and between 10% (in summer and at the equator) and 60% (in winter) at the surface. These values are much higher than those previously published by *Austin and Follows* [1991] and *Follows and Austin* [1992], who estimated that stratospheric O_3 contributed only 25% at 300 hPa and less than 5% at the surface. *Roelofs and Lelieveld* [1997] attributed this to their more detailed treatment of transports, and noted, particularly, the efficient mixing within the troposphere. *Collins et al.* [2000] recently summarized the O_3 budgets from 16 global photochemical model studies. They give a range (mean ± standard deviation) of 340–930 Tg yr^{-1} O_3 as input from the stratosphere, and 2820–4190 Tg yr^{-1} of photochemical production of O_3. In most of these models, the stratospheric source accounts for only about 10–20% of the photochemical O_3 production. However, the actual stratospheric contribution to tropospheric ozone is larger than that because STT maximizes in extratropical latitudes in winter and spring, when the lifetime of ozone is much longer than in the tropics and in summer, when photochemical ozone production maximizes. For instance, *Lelieveld and Dentener* [2000] estimated that in the year 1993 the average global tropospheric O_3 content was 278 Tg, of which almost half (131 Tg) was from the stratosphere, although the stratospheric input in their study was only 17% of the photochemical production.

[24] A particular shortcoming of most model validation studies, even those involving several models and large measurement data sets [e.g., *Law et al.*, 2000], is that they normally do not use directly measured data, but only seasonal means or regionally averaged data. This makes it impossible to determine, on a process level, how accurately the models simulate particular aspects of the O_3 budget. For instance, models can predict similar O_3 seasonal cycles for contradicting reasons: a strong springtime stratospheric

input, an enhanced photochemical production in spring, or both. Only detailed case studies can constrain the models in this respect.

1.6.2. Observations

[25] To study the impact of STE on tropospheric chemistry independently from models, analyses of observations are important. An analysis of O_3 soundings in western Europe revealed that O_3 concentrations in tropopause folds are highest in late spring and early summer, but that the frequency of tropopause folds shows little seasonal variation [*Van Haver et al.*, 1996; *Beekmann et al.*, 1997]. In contrast, stratospheric intrusions identified without requiring a folded tropopause, were about three times more frequent in summer than in winter in these soundings, and, thus, the O_3 input to the troposphere should also be greater in summer. However, this is in contradiction to the global occurrence of tropopause folds as identified from meteorological analyses, which has a summer minimum [*Elbern et al.*, 1998]. A problem when analyzing O_3 sounding data is that tropospheric air may be mixed into air originating from the stratosphere, and the tropospheric air is O_3-richer in summer than in winter.

[26] Mountain peaks are ideal locations to study stratospheric intrusions. Using beryllium-7 (^{7}Be, a tracer for stratospheric air, see below), O_3 and humidity data from the Zugspitze peak in Germany (at about 700 hPa), *Elbern et al.* [1997] found a summer minimum of the frequency of identifiable stratospheric intrusions. *Stohl et al.* [2000], using the same data and also data from other mountain peaks in central Europe, but a different detection algorithm, confirmed the summer minimum, but it was much deeper than in the study by *Elbern et al.* [1997]. Both studies agree that only a few percent of the O_3 at about 700 hPa could be clearly attributed to direct stratospheric intrusions. However, the contribution from aged stratospheric air that could not be identified as such in the measurement data was left unquantified in these studies. Stratospheric intrusions can occasionally directly influence O_3 concentrations at the surface [e.g., *Davies and Schuepbach*, 1994], but these appear to be rare events. For instance, *Reiter* [1991] who found frequent intrusions of stratospheric O_3 at the Zugspitze summit was unable to detect stratospheric O_3 below 1600 m in a valley nearby.

[27] Radionuclide measurements have often been used to study STE processes and to check the representation of STE in models [*Rehfeld and Heimann*, 1995; *Koch and Rind*, 1998]. Unfortunately, no single radionuclide is an ideal stratospheric tracer. For instance, approximately one third of the ^{7}Be, which is often used as a stratospheric tracer, is actually produced in the upper troposphere [*Koch and Mann*, 1996]. Furthermore, ^{7}Be attaches to aerosols and is removed from the atmosphere by wet scavenging [*Gerasopoulos et al.*, 2001]. Therefore, *Raisbeck et al.* [1981] suggested to use the concentration ratio of two radionuclides (^{7}Be and ^{10}Be), which are both mostly produced in the stratosphere and are both scavenged in the same way. The ratio (^{10}Be)/(^{7}Be) is thus not affected by the removal process. *Dibb et al.* [1994] applied this method and found that stratospheric input can account for a maximum of 10–15% of the surface O_3 in the Canadian Arctic in spring, and for less during the rest of the year. Unfortunately, few ^{10}Be data are available globally, because ^{10}Be measurements require accelerator mass spectrometry.

1.7. Mixing of Stratospheric and Tropospheric Air

1.7.1. Mixing Processes

[28] Extrusions from both the stratosphere and the troposphere are usually organized within small-scale filamentary structures and laminae, which appear in various tracer (e.g., PV, water vapor, O_3) fields [e.g., *Dobson*, 1973; *Reid and Vaughan*, 1991; *Newell et al.*, 1999], and are produced by shear in the large-scale wind fields. O_3 layers originating from the stratosphere can persist for 10 days in the troposphere [*Bithell et al.*, 2000], but these structures cascade down to ever-smaller scales until they are mixed with the surrounding air. During the last few decades, numerical techniques have been specifically developed to represent small-scale structures, the most popular ones being contour dynamics [e.g., *Dritschel*, 1989], contour advection [e.g., *Waugh and Plumb*, 1994], and reverse domain filling with trajectories [e.g., *Sutton*, 1994]. But while the generation of filaments is explicitly simulated (at least in an isentropic framework), their decay (if any) is completely nonphysical in these models.

[29] The lifetime of filaments may be limited by three processes: radiation, which can dissolve only the PV anomalies [*Haynes and Ward*, 1993; *Forster and Wirth*, 2000], and turbulence [*Shapiro*, 1980] and molecular diffusion, which both can destroy the filaments physically. Regarding radiation only, the lifetime of stratospheric filaments in the troposphere is a few days [*Forster and Wirth*, 2000]. However, radiation changes locally the static stability of the atmosphere. This may have important implications for the occurrence and strength of turbulence, which takes place on much shorter timescales than radiation. In general, the free troposphere and especially the stratosphere is not a turbulent region, and turbulence is thus intermittent. Turbulence can be generated by convection, breaking gravity waves, wind shear and radiation. Convection is especially important, as streamers of stratospheric air in the troposphere are associated with a PV anomaly. Ascent on the forward flank of the PV streamer can trigger convection, which disrupts the filament and destroys the associated O_3 anomaly [*Langford and Reid*, 1998]. Tropopause folds are considered a region of especially strong mixing between stratospheric and tropospheric air, due to clear air turbulence [*Gidel and Shapiro*, 1979; *Shapiro*, 1980; *Hartjenstein*, 2000]. It has become quite popular to infer information on mixing by analyzing tracer-tracer scatter plots [*Fischer et al.*, 2000; *Parrish et al.*, 2000; *Zahn*, 2001]. All these efforts, however, are in the stage of case studies, and correct quantification of mixing is still an unsolved problem.

1.7.2. Effects of Mixing on Chemistry

[30] In the context of chemical processes, it is important to distinguish between mixing and stirring [*Wiggins*, 1998], because for chemical reactions to take place requires two volumes of air with different chemical characteristics to come into direct contact. Therefore, as long as only filamentary structures are formed, the chemical regimes within and outside the filament are separated. Only molecular diffusion can mix the two regimes and create a new regime with distinct characteristics. However, wind shear, convection and turbulence all act to increase the area/volume ratio

of an air parcel and, thus, produce a cascade down to ever-smaller scales, on which air masses are finally mixed by molecular diffusion.

[31] In the presence of nonlinear chemical reactions, the average chemical composition of two volumes of air with different chemical composition after some time depends on whether they are mixed before or after chemical reactions occur, i.e. on the rate of mixing. In the process of STE, air masses of contrasting chemical composition may be located particularly close to each other [*Pierrehumbert and Yang*, 1993]. High levels of anthropogenic pollution can sometimes be found immediately adjacent to elevated O_3 originating from the stratosphere and mix with each other [*Parrish et al.*, 2000]. During mixing, the combination of high O_3 levels from the stratosphere and high water vapor concentrations from the troposphere increases the production of hydroxyl radicals [*Esler et al.*, 2001], and thus increases the oxidative capacity of the atmosphere. This, in turn, leads to a stronger degradation of carbon monoxide, methane, and higher hydrocarbons, and also to faster loss of nitrogen oxides. Insufficient model resolution or excessive numerical diffusion in Eulerian models, or inadequate parameterizations of mixing processes may therefore lead to systematic errors in chemistry model predictions.

1.8. STE in a Changing Climate

[32] Increases of greenhouse gas concentrations are likely leading to a warming in the troposphere, and a cooling in the stratosphere [*Pawson et al.*, 1998]. Climate models predict that the future warming in the tropics should be stronger in the upper troposphere than at the surface [*Rind et al.*, 1998], increasing the temperature gradient between the tropics and the polar regions and strengthening westerly winds at the jet stream level. Eddy kinetic energy in the lower stratosphere is expected to increase, too. Furthermore, in conjunction with the cooling in the middle and upper stratosphere, the stability in the lowest stratosphere would probably decrease.

[33] Indeed, *Butchart and Scaife* [2001] recently identified a positive trend in STE (3% per decade) in climate model integrations into the year 2051, that was caused by increases in extratropical planetary-wave driving. This, via downward control, led to increased extratropical downward mass flux, balanced by stronger tropical upwelling, in the model. Possible future changes in STE must also be seen against the background of already observed increases in tropopause heights [*Steinbrecht et al.*, 1998; *Highwood et al.*, 2000].

2. STACCATO Objectives and Activities

[34] STACCATO was a joint project of 13 institutes from 7 European countries that was funded by the Commission of the European Union over a two-year period within the Fifth Framework Program. The project's general idea was to bring together modelers and experimentalists from various disciplines for a joint study of STE. 7 groups had their focus on modeling activities, while 6 groups were mostly concerned with measurements. The results of STACCATO are presented in this *Journal of Geophysical Research–Atmospheres* special section and in the project's final report [*STACCATO Team*, 2002] (available from http://www.forst.tu-muenchen.de/EXT/LST/METEO/staccato/).

[35] STACCATO had six major objectives:

[36] 1. From the conceptual point of view the overarching aim of STACCATO was to develop a new three-dimensional Lagrangian perspective of STE, with a focus on processes associated with "deep" exchange events in both directions. This was envisaged as a significant extension, to both the budget approach of *Appenzeller et al.* [1996b] and the *Wei* [1987] method.

[37] 2. Measurements were done in an extended network in order to provide a data set suitable for a model-independent estimate of the strength of STT and its impact on tropospheric chemistry, and to validate the STACCATO models. At two sites, O_3 UV lidars were used to record intrusions of stratospheric air into the troposphere, based on forecasts of these events. Various measurements, including O_3, ^{7}Be and meteorological parameters, were carried out at five mountain stations in the Alps, Northern Apennines, and in Northern Greece. Furthermore, for the first time ^{10}Be was sampled at two stations continuously during the project in order to follow the ideas of *Raisbeck et al.* [1981] and *Dibb et al.* [1994] to estimate the impact of STT on surface O_3.

[38] 3. STACCATO employed 9 different models and methods to diagnose STE. Therefore, it was an important task of the project to intercompare and validate these tools, to find the strengths and weaknesses of each approach, and to identify the reasons for discrepancies.

[39] 4. As mixing of stratospheric and tropospheric air is poorly understood, both in terms of the meteorological processes that cause the mixing, and in terms of the consequences of the mixing for nonlinear atmospheric chemistry, idealized model studies were performed to investigate the mixing of stratospheric and tropospheric air.

[40] 5. A focal point of STACCATO was to examine the variability and possible trends of STE during the past few decades and under scenarios of climate change in the future. For this, two Lagrangian models were driven with consistent high-resolution global meteorological reanalysis data sets for a period of fifteen years to examine the past and present spatiotemporal variability and possible trends of STE. To investigate possible future changes in STE, simulations with three climate models, one with increased vertical resolution in the middle atmosphere, were performed. Two of them were coupled to chemistry models to understand how changes in STE impact on tropospheric chemistry.

[41] 6. Because STT is a major source of ozone for the troposphere, the relative impact of STT on factors controlling the oxidation capacity of the troposphere and lower stratosphere was studied using Eulerian and Lagrangian global chemistry models.

3. What We Have Learned From STACCATO

3.1. Climatologies of STE

[42] The new Lagrangian perspective of STE is illustrated in Figure 3. It was developed for two reasons: First, because the budget approach of *Appenzeller et al.* [1996b] to estimate net STE cannot resolve individual exchange processes, whereas the *Wei* [1987] method is rather inaccurate, especially when it comes to quantifying both STT and TST, and not only the net STE. Second, and more importantly,

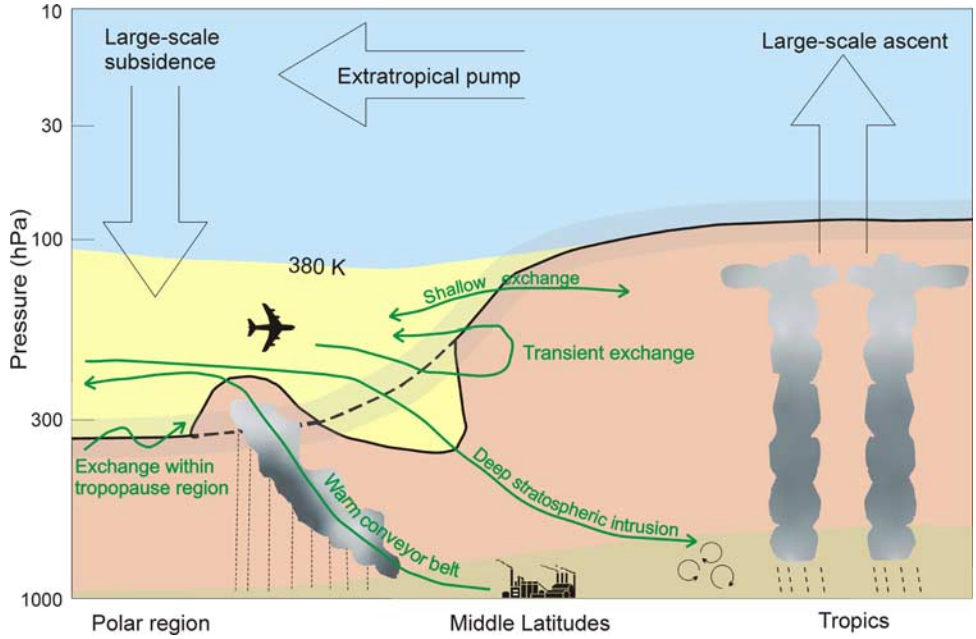

Figure 3. Global aspects of STE from *Holton et al.* [1995], with our new STE concept superimposed. The average position of the tropopause is shown by the thick black line, with shaded regions on either side representing the tropopause region. The blue region is the ''overworld,'' in which isentropes (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 atmospheric boundary layer. Broad arrows show transport by the global-scale circulation. Green trajectories illustrate our new concept, as explained in the main text. 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 line). Note that the pressure is not to scale.

even accurately calculated cross-tropopause mass fluxes do not allow one to characterize relevant aspects of STE. In particular it is of major importance to consider also STT and TST depths and residence times. These characteristics of STE can only be investigated with a Lagrangian approach (Figure 3). For instance, STT or TST that is limited to the tropopause region has moderate impact on atmospheric chemistry. The same holds for transient STT (i.e., air that returns to the stratosphere after residing only a short time in the troposphere), where stratospheric air probably has little opportunity to mix with tropospheric air (and vice versa for TST). Irreversible exchange events of air between the upper troposphere and the lower stratosphere and, in particular, deep exchange events that transport air from the potentially polluted atmospheric boundary layer upward into the lower stratosphere (see Figure 3), or vice versa, within a short time interval have greater impact. If the subset of deep exchange events has particular characteristics (geographical location, seasonal cycle), existing estimates of STT and TST, which are dominated by shallow events, do not characterize the effects of deep STT and deep TST correctly.

[43] First applications of these ideas can be found in the 1-year STACCATO studies of *Stohl* [2001] and *Wernli and Bourqui* [2002]. *Stohl* [2001] required STE particles to cross a PV region instead of a single PV surface. This effectively eliminated shallow exchange within the tropopause region, and, thus, reduces both STT and TST fluxes considerably. *Wernli and Bourqui* [2002] required an air parcel to reside 4 days in both the stratosphere and the troposphere in order to consider it a significant exchange event. In both studies, it was found that deep STT shows a pronounced summer minimum that is not present to the same extent in the full set of STT events. Furthermore, it was shown that both deep STT and deep TST events have a geographical distribution that is different from the full sets of STT and TST events.

[44] Three papers in this special issue extend the above ideas and present, for the first time, Lagrangian global multi-annual (15 years) climatologies of STE. The papers complement each other and focus on different aspects of STE. *Sprenger and Wernli* [2003], using a normal trajectory model, concentrate on timescales of a few days and present detailed geographical distributions of STT and TST and their seasonal variability. *James et al.* [2003a, 2003b], using a Lagrangian particle dispersion model, extend the timescales considered and include transport processes (convection, turbulence) that are missing in the trajectory model.

[45] According to *James et al.* [2003a], most of the STT and TST fluxes are of a rather transient nature. More than 90% of the STT (TST) air resides in the troposphere (stratosphere) for less than 6 hours, before returning to the stratosphere (troposphere). These transient fluxes are highly sensitive to certain parameters in the model (e.g., parameterizations for turbulence, interpolation of winds and tropopause heights) and the associated air parcel remains close to the tropopause. STT (TST) fluxes on timescales of more than 4 days make up only 1–2% of the total fluxes, but their contribution to the concentrations of stratospheric tracer in the troposphere (tropospheric tracer in the strato-

sphere) is much larger, and they can penetrate deeply into their destination sphere.

[46] In both climatologies [*James et al.*, 2003b; *Sprenger and Wernli*, 2003], the annual average net STE (TST-STT) mass flux in the extratropics is similar to that in the budget study of *Appenzeller et al.* [1996b], but its magnitude is lower than those obtained in most studies using the Wei approach. In the extratropics net STE is highest during winter and early spring, which agrees well with the *Appenzeller et al.* [1996b] study (although the seasonal cycle is stronger in the Lagrangian studies, and net STE is close to zero in the fall). Both climatologies show net STT only in the subtropical and middle latitudes, whereas net TST is found in the polar regions. In the particle dispersion model climatology [*James et al.*, 2003b] STE shifts from net STT to net TST at lower latitudes than in the trajectory model climatology [*Sprenger and Wernli*, 2003], possibly because of the additional convection parameterization in the former. The fact that we see net TST in polar regions is a new feature compared to previous STE climatologies, which is in agreement with quasi-geostrophic theory [*Juckes*, 1997]. Net TST in higher latitudes was also noted in a recent paper by *Seo and Bowman* [2002], albeit only seasonally.

[47] In contrast to net STE, both STT and TST maximize already in January and show a strong decrease toward spring and further into summer. The annual cycle's phase of the subset of deep STT is similar to that of total STT, but its amplitude is strongly enhanced. In contrast, there is relatively little seasonal variation of shallow STT flows that only reach the upper troposphere [*James et al.*, 2003b]. One consequence of the steep drop in the frequency of deep STT events from winter toward summer is that STT cannot be the primary reason for the maximum of surface O_3 in spring.

[48] Preferred low-tropospheric source regions for deep TST, that is associated with strong diabatic heating in the troposphere, are located in the middle latitudes near the east coasts of Asia and North America, especially in winter. This indicates that the high-emission areas Japan and eastern North America are the most important extratropical source regions for rapid transport of pollutants into the lower stratosphere. Similarly, there are preferred areas where deep STT influences the low troposphere, along the North American west coast, at the beginning of the Atlantic storm track, and (albeit weaker) in the Mediterranean region. It is in these areas where deep STT is most likely to impact directly on the surface O_3 budget [*Sprenger and Wernli*, 2003]. *Sprenger et al.* [2003] introduce a new technique to identify tropopause folds from global meteorological data and show that 50–70% of the exchange in the subtropics occurs in the vicinity of folds. In the extratropics this value amounts to about 20%.

[49] *James et al.* [2003b] report links between climate variations, such as the North Atlantic Oscillation (NAO) and El Niño/Southern Oscillation (ENSO) and STE. During positive phases of the NAO, the locations of tropopause crossings in the North Atlantic shifts toward higher latitudes and altitudes [see also *Sprenger and Wernli*, 2003]. During El Niño phases, STE in the eastern tropical Pacific shifts toward higher altitudes and intensifies.

[50] *Land and Feichter* [2003] present simulations of ^{7}Be and ^{10}Be for the time-slices 1860, 2000, and 2100 using the middle atmosphere GCM MAECHAM4 and applying the IPCC scenario IS92a. Sea surface temperatures and greenhouse gas concentrations were taken from a climate response experiment with a coupled atmosphere-ocean GCM. In the results for 2100 the zonal mean ^{10}Be/^{7}Be ratio is 10–20% lower above the tropopause (i.e., in the regions where the highest vertical gradient of ^{10}Be/^{7}Be occurs) than in 2000, which is attributed to changes in the residual mean circulation. The model also predicts enhanced STT in the extratropics in the future. However, this increase is asymmetric, with a decrease of cyclonic activity north of 30°N, and a strong increase in the middle latitudes of the southern hemisphere (by up to 45% at around 60°S). The simulated changes of ^{10}Be/^{7}Be ratios in the troposphere are also affected by changes in wet scavenging, and show an increase in the southern, but a decrease in the northern hemisphere.

3.2. STACCATO Measurements

[51] During STACCATO, measurements of O_3, humidity, ^{7}Be and, depending on the station, various other parameters, were continued at three stations in the Alps, one at Mt. Cimone in the Northern Apennines, and one in Northern Greece. *Gerasopoulos et al.* [2001] presented a climatology of the ^{7}Be data from these stations. For the first time in Europe, ^{10}Be was monitored at two stations for more than a year. A total of about 400 daily or bi-daily ^{10}Be measurements are now available, which multiplies the worldwide available measurements of ^{10}Be. These measurements are analyzed by *Zanis et al.* [2003a] following ideas presented by *Dibb et al.* [1994]. At Jungfraujoch the ^{10}Be/^{7}Be ratios range from 1.04 to 3.69 (average of 2.08), while at Zugspitze the ratios range from 0.73 to 3.54 (average of 1.82). The ratio ^{10}Be/^{7}Be shows a seasonal cycle with a clear maximum in May and June at Jungfraujoch, which is less pronounced at Zugspitze. Normally this ratio increases during stratospheric intrusion episodes, but an error propagation analysis [*Zanis et al.*, 2003a] shows that, using the *Dibb et al.* [1994] method, the stratospheric contribution to the surface O_3 can be estimated from these data with large uncertainties only.

[52] C. Land and J. Feichter [see *STACCATO*, 2002] analyzed ^{7}Be measurements from a global network. ^{7}Be concentrations have decreased at most stations (by about 3% per decade on average) during the period 1971–2000, although precipitation at these stations has decreased and the tropopause height has increased during the same time period. However, it remains unclear whether this really indicates a decrease of STT, as other sources of information indicate an increase of STT. In contrast, the Zugspitze time series shows an increase of ^{7}Be by about 50% since 1970. Furthermore, during the past decade O_3 at the Zugspitze has increased most strongly in air masses that contained high concentrations of ^{7}Be [*Scheel*, 2002].

[53] O_3 lidar measurements have been carried out in Germany and Northern Greece, which have been used to learn more about the transport processes involved with both high and low O_3 concentrations [*Galani et al.*, 2003; T. Trickl et al., Intercontinental transport and its influence on the ozone concentrations in the free troposphere over central Europe: Three case studies, submitted to *Journal of Geophysical Research,* 2003] (hereinafter referred to as Trickl et al., submitted manuscript, 2003). The measure-

ments were co-ordinated with O_3 sounding activities at other stations and were conducted specifically during STT events. Daily forecasts of these events using trajectories based on ECMWF data were carried out for the first time during STACCATO and proved to be extremely helpful for the campaign planning. It turned out that STT events over Europe occur frequently in conjunction with episodes of intercontinental transport of air pollution from North America (Trickl et al., submitted manuscript, 2003). Hence, air masses of stratospheric origin were often found in proximity to polluted air masses, indicating the possibility of mixing between the two.

3.3. Model Intercomparison and Validation

[54] Another focus of STACCATO was model intercomparison and model validation. *Meloen et al.* [2003] and *Cristofanelli et al.* [2003] report on a case study of a deep STT event for which 9 different methods and models (Wei formula, 3 trajectory models, 2 Lagrangian particle models, 1 Eulerian transport model, and 2 GCMs nudged toward analyses) were requested to simulate the transport of an idealized stratospheric tracer with a lifetime in the troposphere of 2 days. *Meloen et al.* [2003] intercompare the model results and *Cristofanelli et al.* [2003] compare them with measurements. It is found that all models captured the STT event, but the magnitudes of the stratospheric tracer fluxes in the troposphere are larger by about a factor of four for the GCMs than for the Lagrangian models. When compared to the measurements, it appears that the trajectory models sometimes fail to capture the presence of stratospheric air in the troposphere, likely because convection and turbulence schemes are missing in these models. They therefore tend to underestimate STT. On the other hand, the GCMs disperse the stratospheric tracer over too large areas and too deeply into the troposphere, which is likely an artefact from numerical diffusion, combined with low resolution and strong tracer gradients across the tropopause. They, thus, tend to overestimate STT.

[55] *Zanis et al.* [2003b] evaluated O_3 from the European Centre for Medium-Range Weather Forecasts (ECMWF) during the same event. To our knowledge, this is the first validation of a chemical tracer that is used operationally in a weather forecast model. Even though O_3 chemistry in the ECMWF model is represented in a very simplified way, very good agreement with the observations was found. This suggests that coupling chemistry and weather forecast models may significantly improve both weather and chemical forecasts in the future.

3.4. Influence of STE on the Oxidizing Capacity of the Troposphere

[56] B. Krüger [*STACCATO Team*, 2002] used an idealized chemical box model to study chemical effects of the mixing of stratospheric and tropospheric air. He found that hydroxyl radical concentrations can be increased strongly (up to a factor of 25) in mixed air masses, and that chemical losses of O_3 in stratospheric intrusions are slightly increased by the mixing with tropospheric air. This leads to a slight speed-up of the disappearance of the O_3 maximum. On a global scale, the lifetime of O_3 of stratospheric origin is mostly determined by the transport time between the stratosphere and the subtropical lower troposphere, where radiation is strong and water vapor concentrations are high. According to simulations with the chemistry-climate model ECHAM, *Kentarchos and Roelofs* [2003] report that the overall influence of stratospheric O_3 on hydroxyl radical concentrations in the northern hemisphere varies between 40% in winter and about 5% during summer.

[57] Analyzing O_3 data obtained aboard commercial airliners within the MOZAIC program, *Stohl et al.* [2001] found that even in the upper troposphere photochemical O_3 production over the continents left the strongest signal in the measured O_3 concentrations during spring and summer.

[58] Aircraft emissions play an important role in the chemistry of the upper troposphere and lower stratosphere. Including aircraft emissions in the chemistry-climate model ECHAM improves the agreement between simulated and measured nitrogen oxides in the upper troposphere and lower stratosphere over the North Atlantic flight corridor considerably, but O_3 concentrations increase only by about 3 ppb in summer, and less during the rest of the year [*Kentarchos and Roelofs*, 2002]. Higher hydrocarbon chemistry causes a 20% increase of the ozone production per NO_x molecule from aircraft compared to a background chemistry scheme [*Kentarchos and Roelofs*, 2002].

[59] Normally, aircraft emission inventories report annual emissions, and these are applied continuously in chemistry models. In reality, however, the flight tracks of commercial aircraft are adjusted to the actual meteorological situation in order to avoid head winds and turbulence. *Forster et al.* [2003] show that accounting for these shifts in the flight tracks increases the fraction of aircraft emissions released in the stratosphere from 62 to 67%, a rather small but systematic effect that is not accounted for in global chemistry models.

[60] *Collins et al.* [2003] investigate the impact of circulation changes in a changed climate on the exchange of O_3 between the stratosphere and the troposphere. Using simulations with the Lagrangian chemistry transport model STOCHEM for 4 periods of 6 years (1990–95, 2030–35, 2060–65, 2090–95), they have identified an increase in the net transport of O_3 into the troposphere in the future climate of 37%. However, assuming unchanged O_3 precursor emissions, a decreased O_3 lifetime due to a moister and warmer climate means that the overall tropospheric O_3 burden decreases. While there may be caveats from the way stratospheric O_3 was prescribed in the model, increased input from the stratosphere was also seen in other stratospheric tracers (^{7}Be and ^{10}Be), in agreement with the MAECHAM4 simulations of *Land and Feichter* [2003].

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