Stratosphere-troposphere exchange properties

by

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To my wife, my parents, and my family

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Abstract

Stratosphere-troposphere exchange (STE) and its effects on the stratospheric and tropospheric chemical compositions have been studied for the past two decades, but details on how mass is transported between the stratosphere and the troposphere are not well established. The goal of this study is to better describe global properties of cross tropopause trajectories, and to understand the processes related to transport of mass between the troposphere and the stratosphere. This understanding led us to build the simplest model which captures the most important properties of STE.

To do this, nine-day extra-tropical stratosphere-troposphere exchange trajectories covering a period of 10 years, calculated using the ERA-15 re-analysis data, are investigated. The present study shows that the fraction of trajectories that reside in the stratosphere or in the troposphere does not depend on the direction of the exchange (stratosphere-to-troposphere transport, STT, or troposphere-to-stratosphere transport, TST). Trajectories are found to reside longer in the troposphere than in the stratosphere which suggests that they are driven down by asymmetric two-way motion.

A random walk model is used to see whether this asymmetric transport is a result of a diffusive process. The transport of trajectories along isentropic coordinates is found to be compatible with a Brownian motion with higher probabilities to go downward. Since stratosphere-troposphere exchange reflects a differential motion of air masses and the tropopause, the potential temperature at the tropopause directly above or below the air mass is also investigated. The tropopause steps distributions are not stationary and they show some dynamical behaviors like the deformation of the tropopause at exchange time.

Dispersion of trajectories in the atmosphere was furthermore investigated using several methods. They gave rise to three different transport mechanisms: diffusion, sub-diffusion and super-diffusion transports. These transport processes neither depend on the direction of the exchange STT/TST nor the environment of transport (strato-sphere/troposphere).

Résumé

Les échanges entre la stratosphère et la troposphère (STE) et leurs effets sur la composition chimique de la stratosphère et de la troposphère ont été étudiés intensivement, mais les détails sur la manière dont les masses d'air sont transportés entre la stratosphère et la troposphère ne sont pas bien établis. Le but de cette étude est de décrire les propriétés globales des trajectoires qui traversent la troposphère et la stratosphère. Une fois ces détails établis, nous avons essayé de construire un modèle simple qui capture les propriétés les plus importantes des STE.

Pour ce faire, des trajectoires d'échanges entre la stratosphère et la troposphère (STE), couvrant les extra-tropiques durant une période de 10 ans ont été calculées en utilisant les données d'ERA-15. Dans cette étude on montre que la fraction de trajectoires qui résident dans la stratosphère ou dans la troposphère ne dépend pas de la direction de l'échange (transport de la stratosphère à la troposphère, STT, ou de la troposphère à la stratosphère, TST). Les trajectoires résident plus longtemps dans la troposphère que dans la stratosphère. Ceci suggère que les trajectoires sont entraînées vers le bas par un flux asymétrique.

Un modèle de marches aléatoires est utilisé pour voir si ce transport asymétrique est le résultat d'un processus de diffusion. Le transport de trajectoires à travers les surfaces de la température potentielle est compatible avec un mouvement Brownien avec une plus grande probabilité d'aller vers le bas. Puisque l'échange entre la stratosphère et la troposphère reflète un mouvement différentiel entre les masses d'air et la tropopause, la température potentielle à la tropopause directement au-dessus ou au-dessous de la masse d'air est également étudiée. Les distributions de pas de la tropopause ne sont pas stationnaires et montrent quelques effets dynamiques comme la déformation de la tropopause au temps d'échange.

De plus, différentes méthodes ont été utilisées pour étudier la dispersion des trajectoires dans l'atmosphère. Trois différents mécanismes de transport ont été identifiés: diffusion, sous-diffusion et super-diffusion. Ces processus de transport ne dépendent ni de la direction d'échange ni de l'environnement de transport (stratosphère/troposphère).

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Introduction

Structure of the atmsophere

The atmosphere is usually divided in layers with respect to the vertical structure of the temperature. The troposphere is the lowest and the most dense layer of the atmosphere; it contains 80% of the mass of the atmosphere. The atmosphere (here we focus on the troposphere and the stratosphere) is composed of 78% of N₂, 21% of O_2 and 1% of trace gases. In the troposphere the temperature decreases globally with altitude as a result of the heating of the Earth's surface by the Sun and the radiativeconvective adjustment aloft (*Thuburn and Craig*, 1997). The troposphere is therefore well mixed vertically. The stratosphere is situated between the troposphere and the mesosphere. Due to its vertically increasing temperature the stratosphere is vertically stable, therefore there is no formation of convection. Three dimensional turbulence still occurs at small scales due to the breaking of gravity waves. The stratosphere is heated at the top by the absorption of ultraviolet radiation coming from the Sun by stratospheric ozone. About 90% of atmospheric ozone is found in this layer, where concentrations reach 10 ppm, in comparison to the typical 50 ppb found in the free troposphere. The distribution of ozone is determined by a balance between photochemical reactions, and by the transport between source regions and sink regions. The stratosphere was divided into different regions based on their radiative, dynamical and chemical behavior (WMO, 2003). The structure and zonal mean circulations of the stratosphere are shown in Figures 1 and 2. The lowermost stratosphere (LMS) is the region that lies between the tropopause and the 380 K surface (~ 15 km). This is the region of the stratosphere where isentropes reach the troposphere. The lower stratosphere (LS) covers the region between 380 K and about 25 km. In the LMS and LS, odd-oxygen has a long lifetime, and hence reacts slowly to chemical changes compared to transport processes. Above the LS, between 25 km and 50 km, there is the upper stratosphere (US). In the US, lifetime of odd-oxygen is becoming shorter at higher elevation, compared to transport time scale, and then photochemistry dominates (WMO, 2003).

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The stratosphere and troposphere are separated by a boundary region called the tropopause. The WMO definition for the thermal tropopause is the lowest level at which the lapse rate decreases to 2 o C km⁻¹ or less, provided also that the average lapse rate between this level and the higher levels within 2 km does not exceed 2 ^{o}C $\rm km^{-1}$. Its height varies with the weather systems, seasons and latitude. The tropopause can be defined dynamically by iso-surfaces of potential vorticity (PV). The advantage of the dynamical definition is that PV is a conserved property for an air mass within adiabatic and frictionless conditions. This definition gives a clear physical meaning to stratospheric-tropospheric exchange (STE) processes across a well defined surface. The tropopause is a surface of reduced transverse mass flux, and is therefore a "mixing barrier" (Holton et al., 1995), leading to two environments with different properties and with significant contrasts of chemical compositions. Only in some favorable situations the stratosphere-troposphere exchange is significant, like a formation of folds, anticyclones and cut-off cyclones. The flux of chemical species across the tropopause is important and crucial in order to understand the chemical evolution of the stratosphere and the troposphere.



Figure 1: Schematic diagram showing the structure of, and transport within, the stratosphere. Adapted from (WMO, 2003).

Figure 2: Net mass flux across iso-PV surfaces. Units in 10^6 kg s⁻¹ km⁻¹. Solid lines: 1.5, 2, 3, 5, 8 PVU iso-PV surfaces. Dashed lines: Isentropes in K. Adapted from (*Bourqui*, 2001).

Stratosphere-Troposphere Exchange

The exchange of mass between the stratosphere and the troposphere was first studied using radioactive elements to trace stratospheric air in the troposphere (*Staley*, 1962; Danielson, 1968), and then using satellite and aircraft data of chemical tracers (Shapiro, 1980; Browell et al., 1987). Transport in the over-world (LS+US) is realized by large scale Brewer-Dobson circulation (Figures 1 and 2) induced by momentum deposition by planetary and gravity waves (Brewer, 1949; Dobson, 1956; Holton et al., 1995); the wave driven pump draws air upward across the troppause in the tropics, and pushes it poleward downward into the LMS at extra-tropical latitudes. Troposphereto-stratosphere transport (TST) occurs mainly in the tropics, and also at higher latitudes. Conversely, stratosphere-to-troposphere transport (STT) happens everywhere. but dominates in the mid-latitudes. The exchange of mass between the extra-tropical stratosphere and the troposphere is thought to be mainly quasi-isentropic (*Chen*, 1995), since non conservative upward transport which occurs mostly through deep convection is less important due to the stable vertical layering of the LMS. STE are dominated by synoptic scales eddies such as upper-level troughs, cut-off lows, blocking anticyclones and tropopause folds (*Bregman et al.*, 1997; *Lelieveld et al.*, 1997), that often take place in baroclinic wave breaking events. We can distinguish different processes: i) The formation of filaments or folds of stratospheric air entering the troposphere where it mixes, or vice versa (Appenzeller and Davies, 1992; Thorncroft et al., 1993). Lamargue and Hess (1994) and *Rood et al.* (1997) show that diabatic processes in cyclones and associated tropopause folds are important mechanisms for STE. ii) The formation of a cut-off of cyclonic vortex of stratospheric air in the troposphere, where it remains for a few days before returning back while it mixes with tropospheric air (*Price and Vauqhan*, 1993; Thorncroft et al., 1993). iii) The formation of a cut-off of anti-cyclonic vortex of tropospheric air in the stratosphere, where it remains for a few days before returning back while it mixes with stratospheric air (*Peters and Waugh*, 1996; *Vaughan and Timmis*, 1998).

To study such mixing processes, various methods have been used. In the case of the Eulerian diagnostic methods (*Wei*, 1987; *Searle et al.*, 1998), the cross-tropopause flux has generally been computed with one of the formulas given by *Wei* (1987); vertical coordinate can be pressure, potential temperature or potential vorticity (PV). In the Wei-formulation with the PV coordinates, the air mass per unit area across a PV surface is approximated to:

$$F = \frac{-1}{g} \frac{\partial p}{\partial PV} \frac{DPV}{Dt} \tag{1}$$

where p and g are the pressure and the acceleration due to gravity, respectively. The unit of F is kg $m^{-2} s^{-1}$. When using (re-)analysis data it is difficult to extract the material

derivative of PV needed for this formulation. Wirth and Egger (1999) reported that when using p or θ as a vertical coordinates, large relative errors in cross-tropopause flux will be expected. When using numerical models, PV is taken as vertical coordinates, PV-sources can be calculated with reasonable accuracy.

In the Eulerian description, the motion of a fluid is observed at a given position, and the Lagrangian reference frame is the frame that moves with individual fluid particles as they move through space and time. The position of these individual particles versus time gives the trajectory or the pathline of a particle. In the Lagrangian picture new and different phenomena can be seen and understood in a rather simpler manner than in the Eulerian framework. For Lagrangian methods (Wernli and Davies, 1997; Stohl et al., 2003), elementary air volumes are advected by 3-dimensional winds, and rather than estimating $\frac{DPV}{Dt}$, which is very noisy; we count the number of elementary volumes crossing the tropopause. Wirth and Egger (1999) found by comparing Lagrangian and Eulerian techniques that realistic results can be obtained with trajectory method. The contour advection technique was used to study the isentropic aspect of transport during the formation of small structures (*Norton*, 1994; Waugh and Plumb, 1994; Appenzeller et al., 1996). Potential vorticity and specific humidity fields were used as initial tracer fields, which were then advected as passive tracers. Mesoscale aspects have been also studied using limited area models (Lamarque and Hess, 1994; Wirth and Egger, 1999). The estimation of the magnitude of the average STE mass (Lamarque and Hess, 1994; Murphy and Fahey, 1994; Roelofs and Lelieveld, 1995; Beekmann et al., 1997; Tie and Hess, 1997; McLinden et al., 2000) gives very different results. Some differences are attributed to resolution and parameterization schemes, but the cause of the other differences remains unknown.

Another approach was used based on the Lagrangian technique (*Bourqui*, 2001; *Wernli and Bourqui*, 2002; *Bourqui*, 2006) that takes into account the pathways of exchanged air parcels and their residence time before and after the exchange. In these studies it is found that the cross-tropopause mass fluxes are sensitive to a residence time threshold.

Age Spectrum

The flux can be constrained using measurements of atmospheric chemical tracers, but the determination of transport properties from tracers is more difficult due to ambiguity of the sources and sinks of tracers. Recently, an increasing interest has been devoted to age spectrum analysis in the stratosphere (*Hall and Plumb*, 1994; *Holzer and Hall*, 2000; *Hall and Holzer*, 2003) to improve the interpretation of the processes that lead to the distribution of stratospheric tracers. The age spectrum, in other words distribution of transit times, was identified to a type of Green function that propagates a boundary condition on tracer mixing ratio from the tropical tropopause into the stratosphere. The application of this concept to the stratosphere results from the fact that the boundary region is easily localized, which is the tropical tropopause.

The continuity equation of a passive tracer is

$$(\partial_t + \Gamma)\chi = S \tag{2}$$

with χ is the mass mixing ratio, the transport operator Γ represents advection and diffusion, and S is the source of tracer.

If we know the solution of the equation where the source S is replaced by the Dirac function

$$(\partial_t + \Gamma)G(\vec{r}, t \mid \vec{r'}, t') = \delta(\vec{r} - \vec{r'})\delta(t - t'),$$
(3)

the solution of the Eq. 2 is

$$\chi(\vec{r},t) = \int d^3 \vec{r'} \quad G(\vec{r},t \mid \vec{r'},0)\chi(\vec{r'},0) + \int_0^t dt' \int d^3 \vec{r'} \quad G(\vec{r},t \mid \vec{r'},t')S(\vec{r'},t')$$
(4)

G is called the Green function, and the first term in Eq. 4 is the time evolved initial condition. $G(\vec{r}, t \mid \vec{r'}, t')$ is the response at position \vec{r} and time *t* to a unit mass source of tracer, $\rho^{-1}\delta(\vec{r}-\vec{r'})\delta(t-t')$, where ρ is the fluid density. Tracer fluid elements were at position $\vec{r'}$ at time *t'*.

The familiar form of the transport operator is

$$\Gamma \chi = \vec{v}.\vec{\nabla}\chi - \rho^{-1}\vec{\nabla}(\rho\kappa\vec{\nabla}\chi) \tag{5}$$

with $\vec{v}(\vec{r},t)$ is advecting velocity and $\kappa(\vec{r},t)$ is eddy-diffusivity.

Transport in the atmosphere

Although STE and its effect on stratospheric and tropospheric chemical composition have been studied for a long time, details on how mass is transported between the stratosphere and the troposphere are not well established. Understanding transport of elements has been a subject of considerable interest for theoretical and practical studies in many fields of science. The dispersion of transported quantities is usually the result

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of two different contributions: diffusion and advection. Atmospheric models take into account small scales (the unresolved scales) by representing them by turbulent diffusion. This turbulent diffusion is much larger than molecular diffusion, of course. It is important to reproduce correctly the transport and mixing processes in the stratosphere in order to provide adequate description of the impact of anthropogenic species on climate. Transport is related to the motion of a group of fluid elements from an environment to another one, and it is measured by the mean position of fluid elements. The common examples of transport phenomenas are diffusion, convection and radiation. Mixing is related to the spreading of nearly neighbors fluid particles, and it is measured by the growth of the mean square deviation of particles position with time.

A wide range of diffusive processes were interpreted using a random walk theory; *Einstein* (1905) showed that the Brownian motion or random motion of passive tracers in a homogeneous fluid is a result of random walk collisions. In a random walk theory, we put a walker in a domain to execute a series of random steps. That is repeated for several times and for long times to get converging statistics. For a standard diffusion the mean square displacement of the ensemble of particles grows linearly with time. When the spreading of particles does not grow linearly with time, the process is called anomalous diffusion, i.e. non-Brownian. When relative dispersion grows slower than diffusive process we have sub-diffusion transport. A super-diffusion transport occurs when relative dispersion grows faster than diffusive transport.

It is known that turbulence transports and mixes species much faster than diffusion processes. The two major sources of the atmospheric turbulence are wind shear and gravity wave breaking. Transport and mixing processes in the atmosphere were studied by using theoretical and numerical experiment studies (*Pierrehumbert*, 1991a,b; *Weiss*, 1991; *Bowman*, 1993; *Bowman and Mangus*, 1993; *Pierrehumbert and Yang*, 1993; *Huber et al.*, 2001). It has been shown that fluid elements can be mixed by a deterministic flow through chaotic mixing; a pair of initially close trajectories separate exponentially with time. By studying the mixing on isentropic surfaces in the troposphere, *Pierrehumbert and Yang* (1993) found that the zonal variance grows superdiffusively with time due to the systematic shear in the extra-tropical jet, and a diffusive growth has been found for the meridional variance. *Huber et al.* (2001) studied meridional turbulent mixing along the isentropic surfaces in the troposphere using the winds from the European Centre for Medium-Range Weather Forecasts (ECMWF) numerical weather prediction model. Tropical dispersion was found to be characterized by exponential growth with time, and a super-diffusivity growth was found in the extra-tropics.

Motivation and structure of the research

Over the last few years, *Hegglin et al.* (2005) studied aircraft observations of seasonal distributions of CO in the LMS. A 2-dimensional advection-diffusion model was used to simulate transport of tracers which includes diabatic descent of air from the stratosphere as well as horizontal and vertical eddy diffusion. To simplify the complexity of transport process, equivalent latitude and potential temperature coordinates were used. The observed tracer time series are fitted with the model to get diffusion coefficients for the LMS region. Simulations without vertical mixing were unable to reproduce the observed results, which suggests the importance of diabatic mixing across the isentropes. The extracted values of the vertical diffusivities corresponds to $\kappa_{\theta} \sim 0.45 - 1.1 \ m^2 s^{-1} \ (8.7-21.4 \ K^2 day^{-1})$. These values depend directly on the chosen diabatic heating.

Hall and Holzer (2003) computed the flux of an advective-diffusive flow using the concept of boundary-propagator Green functions. The gross flux was established for 1-dimensional idealized advective-diffusive model. A model schematic is shown in the small windows of Figure 3. Fluid of constant density advects around a loop at constant speed u, and diffusion occurs along the flow. The surface S divides the domain into R_1 and R_2 . The gross flux is plotted in Figure 3 as a function of residence time, for different Péclet numbers ($Pe = uL/\kappa$, where u is advective wind, L is length scale, κ is diffusion coefficient). $Pe = \frac{\text{advective time scale}}{\text{diffusive time scale}}$, with advective time scale=L/u and diffusive time scale= κ/u^2 . At low residence times ($\tau < \kappa/u^2$) a $\tau^{-1/2}$ divergence of the flux occurs. This corresponds to diffusive regime. At large Pe the diffusive time scale (κ/u^2) is smaller than the advective time scale (L/u), $\kappa/u^2 = 1/Pe L/u << L/u$, and the two regimes are separated enough to allow the flux distribution to develop a plateau, while for Pe around 1-10, no plateau was seen since the diffusive time scale and the advective one are not very separated.

The authors suggest that the high sensitivity of the computed flux to residence times is a natural feature of advective-diffusive flows; The flux is evaluated in the regime dominated by diffusion (*Hall and Holzer*, 2003).

Our current study contributes to the research efforts towards a better understanding of STE, with an emphasis on the global statistical properties of the transport of mass between the troposphere and the stratosphere. We use 10-years extra-tropical STE trajectories calculated using the ERA-15 re-analysis data.

We know from previous studies (*Bourqui*, 2001; *Wernli and Bourqui*, 2002; *Bourqui*,



Figure 3: The gross flux at different Pe established for 1-dimensional idealized advectivediffusive model as a function of residence time. Figure from (*Hall and Holzer*, 2003)

2006) that the Lagrangian framework provides an adequate perspective to identify and quantify the relevant STE properties. As an example, the residence time that is used to distinguish between significant exchange events, i.e. air volumes with a residence time τ on either side of the troppause exceeding a certain threshold τ^* , which has very important impact on chemistry and exchange events that moves rapidly between the stratosphere and the troposphere and has a weak chemical impact. We studied these residence time distributions for different seasons. Starting from the results of *Heaglin et al.* (2005) which suggested that overall effect of cross-tropopause exchanges may be represented by a simple advective-diffusive model and connecting it to the work of Hall and Holzer (2003) which attributed the differences in the computed values of STE fluxes to domination of diffusion process, our efforts will be focused on improving our knowledge about the representation of STE as a diffusive process. We study crosstropopause air mass transport problem from a global perspective. We discussed this using the random walk theory to simulate diffusion, and we will see whether crosstropopause trajectories data properties can be reproduced by a simple random walk theory. To investigate the dynamics of particles moving in the atmosphere, we have followed trajectories of air masses and calculated the distributions or the ensemble averages of some statistical variables as a function of the total number of steps executed by moving air mass.

In the second part of the analysis, we investigate the mean flow characteristics

by studying the deformation tensor using a group of trajectories. The goal of using 3dimensional tensor is to see if there is isotropy in the atmospheric transport, i.e. to look if the trajectories are dispersed with the same probability horizontally and vertically. After we investigate in more details the diffusion and dispersion of air volumes in the atmosphere using the relative dispersion technique of trajectories. In addition, characteristics of STE trajectories are analyzed with non-linear dynamics technique. Previous studies (*Artale et al.*, 1997; *Boffetta et al.*, 2001; *Lacorata et al.*, 2001) show that useful information may be obtained by using the Finite Size Lyapunov Exponent (FSLE). In the present work both proposed techniques to compute FSLE are used.

The first chapter offers a brief description of the data and the method used to compute Lagrangian trajectories. The second chapter focused on the description of the techniques and the relevant variables used for the analysis of the trajectories, and in the third chapter we apply all these techniques to ERA-15 re-analyses data from ECMWF. In conclusion we summarize the main achievements of the work.

Introduction

Chapter 1

Data and Lagrangian trajectory computation

This chapter contains a summary of the data that we will analyze and study in the next chapters. The Lagrangian method used to compute STE trajectories will be described.

1.1 Data

In this study, I analyze cross-tropopause trajectories, in the extra-tropical region, calculated using ERA-15 re-analyses data from the European Center for Medium-Range Weather Forecasts (ECMWF) (*Sprenger and Wernli*, 2003), over a 10-year period from January 1983 to December 1993. ERA-15 is the first re-analyses project and applied the optimal interpolation method to the period 1979-1994.

The horizontal and vertical wind components needed to compute trajectories are available every 6 hours. The re-analysis were accomplished with a horizontal spectral resolution of T106, corresponding to a resolution of 1.125 degrees, with 31 vertical hybrid levels from the surface up to the 10 hPa. Secondary variables like potential temperature θ and PV have been calculated on the original hybrid model levels.

1.2 Lagrangian computation method

The transport and dispersion of air mass volumes are computed with 3D LA-GRangian ANalysis TOol LAGRANTO (*Wernli and Davies*, 1997; *Wernli and Bourqui*, 2002; *Bourqui*, 2006). This technique was introduced by *Wernli and Davies* (1997) to study the dynamics of extra-tropical cyclogenesis, and after was used to quantify the exchange of mass between the stratosphere and the troposphere (*Wernli and Bourqui*, 2002; *Bourqui*, 2006). The evaluation of trajectories is undertaken with the Peterssen's kinematic method (*Petterssen*, 1956). For each time step $(t \rightarrow t+\Delta t)$, the position of the trajectory is given by the convergence for $i \geq 2$

$$\vec{r}_i(t + \Delta t) = \vec{r}_0 + \frac{\Delta t}{2} (\vec{u}(r_0, t) + \vec{u}(r_{i-1}, t + \Delta t)), \qquad (1.1)$$

where $\vec{r}_1 = \vec{r}_0 \Delta t \ \vec{u}(r_0, t)$.

The wind components \vec{u} at the integrid locations are linearly interpolated from the gridded values.

STE is studied using the notion of dynamical tropopause defined by the 2 PVU iso-surface. Starting on a regular grid the horizontal and vertical positions (longitude, latitude and pressure), potential temperature and potential vorticity of air parcels are traced for 9 days with a resolution of 6 hours. To compute the exchange trajectories:

- The time dependent flow is divided in space and time into a series of trajectories. These trajectories are started every 24 hours on a regular grid space with the grid that has increments of 80 km in the horizontal and 30 hPa in the vertical, so that each trajectory represents an air mass of $\Delta m = g^{-1}\Delta x \ge \Delta y \ge \Delta p = 157 \ge 10^9 kg$.
- Cross-tropopause events are selected when the trajectory's PV value intersects 2 PVU within 24 hours.
- The cross- tropopause trajectories are extended for four days both backward and forward in time.

These data have been used by *Sprenger and Wernli* (2003) to produce 15-year climatology of the geographical distribution of cross-tropopause transport. In this study the authors were interested only on exchange events associated with long residence times. Except for summer, the geographical distributions of STT show strong zonal variations and pronounced maxima in the northern Atlantic and Pacific storm track regions and the Mediterranean. During summer, STT is weak over oceans and strong over the Eurasian continent. Different processes could be responsible for this STT activity during summer over the continents and during the other seasons over the oceans. TST is important only near the Greenland and the Aleutian Islands. Aspects of the interannual variability of the geographical exchange distributions were studied. The authors report that STT varies significantly with the changing storm tracks during the North Atlantic Oscillation.

Chapter 2

Methodology

In order to study the transport processes involved during the exchange of mass between the stratosphere and the troposphere, air mass trajectories are analyzed using different techniques. In this chapter we describe the techniques and define the variables used for the analysis.

2.1 Residence time

The residence time of a particle in a domain S (the stratosphere or the troposphere) is the time duration that the particle will remain in this domain. For a given trajectory if t_f is the time at which a particle leaves the domain S, and t_i is the time at which a particle enters this domain, the residence time of particle in the domain S is $\tau = t_f - t_i$ (see Figure 2.1).

The fraction of trajectories that spent a time longer than a threshold τ in the stratosphere or in the troposphere is defined by

$$F(\tau) = \int_{\tau}^{+\infty} E(\tilde{\tau}) d\tilde{\tau}, \qquad (2.1)$$

where $E(\tilde{\tau})$ is the normalized stratospheric or tropospheric residence time distribution, such that $\int_0^{+\infty} E(\tilde{\tau}) d\tilde{\tau} = 1$



Figure 2.1: Schematic illustrating the notion of residence time of a trajectory that crosses the tropopause.

2.2 Random walks theory

A stochastic or random process is a collection of random variables X_t . Instead of dealing only with one possibility for the future evolution of the system, in random processes there are more than one possible realizations of the future evolution.

Different stochastic processes exist, depending on the nature of the state space, the index parameter T and the dependence on the random variables (*Karlin and Taylor*, 1975). The state space S is the space in which the values of each variable X_t lie:

- If $S = \{0, 1, 2, ..\}, X_t$ is called discrete process.
- If $S =]-\infty, +\infty[, X_t \text{ is called real-valued stochastic process.}$

The index parameter T is the space in which the time values lie:

- If $T = \{0, 1, 2, ..\}, X_t$ is called discrete time stochastic process.
- If $T = [0, +\infty[, X_t \text{ is called continuous time stochastic process.}]$

The stochastic processes are classified into different types:

- 1- Process with stationary independent increments: If the random variables X_{t_i} - $X_{t_{i-1}}$ are independent for all times $\{t_i, i = 1, .., n\}$, the $\{X_t\}$ is called a process with independent increments. If the distribution of the increments $X_{t_{i+h}}$ - X_{t_i} depends only on the length h of the interval and not on time t_i , the process is said to have stationary increments. Examples: the Brownian and the Poisson processes.
- 2- Martingales process: It is a stochastic process which has the property that the expected value of all future instant is zero. The expected value at any future moment equals the value of the process at the present instant.
- 3- Markov process: It is a process with the property that the probability of any particular future realization, when its present state is known exactly, does not depend on its past realizations. If the present state of the process is not precisely known, then the probability of some future behavior will be altered by additional information relating to the past state of the system.
- 4- Stationary process: A stochastic process X_t is said to be strictly stationary if the joint distribution functions of the families of random variables $\{X_{t_{1+h}}, X_{t_{2+h}}, ..., X_{t_{n+h}}\}$ and $\{X_{t_1}, X_{t_2}, ..., X_{t_n}\}$ are the same for all h > 0 and $t_i \in T$.

The man properties of a simple random walk process are:

- discrete Δt time, discrete Δx value process.
- independent increments.
- stationary.
- probability distributions are given by Binomial law.

These properties and other ones will be described in more details in next sections.

2.2.1 Analogy between random walk and diffusion

Mathematically, diffusion of the trace χ is described by the diffusion equation:

$$\frac{\partial \chi}{\partial t} = D \ \vec{\nabla}^2 \chi \tag{2.2}$$

Diffusion can also be described by a simple random walk. Consider a symmetric random walk, where $p_k(n)$ the probability that a particle finds itself k steps to the right of its starting point at time n. From the Chapaman-Kolmogorov relation (*Karlin and Taylor*, 1975)

$$p_k(n+1) = \frac{1}{2}p_{k+1}(n) + \frac{1}{2}p_{k-1}(n)$$
(2.3)

This equation can be written as

$$p_k(n+1) - p_k(n) = \frac{1}{2}([p_{k+1}(n) - p_k(n)] + [p_{k-1}(n) - p_k(n)])$$
(2.4)

Let Δ and η be the length of time between transitions and the length of each step, respectively. Eq. 2.4 becomes

$$\frac{p_{k\eta}((n+1)\Delta) - p_{k\eta}(n\Delta)}{\Delta} = \frac{\frac{1}{2}([p_{(k+1)\eta}(n\Delta) - p_{k\eta}(n\Delta)] - [p_{k\eta}(n\Delta) - p_{(k-1)\eta}(n\Delta)])}{\Delta}$$
(2.5)

We replace $k\eta$ by x and $n\Delta$ by t, Eq. 2.5 becomes

$$\frac{p(x,t+\Delta) - p(x,t)}{\Delta} = \frac{\frac{1}{2}([p(x+\eta,t) - p(x,t)] - [p(x,t) - p(x-\eta,t)])}{\Delta}$$
(2.6)

If Δ and η shrink to zero preserving the relationship $\delta = \eta^2$. Taylor expanded around p(x,t):

$$p(x,t+\Delta) = p(x,t) + \Delta \frac{\partial p(x,t)}{\partial t} + \dots$$
(2.7)

$$p(x+\eta,t) = p(x,t) + \eta \frac{\partial p(x,t)}{\partial x} + \frac{\eta^2}{2} \frac{\partial^2 p(x,t)}{\partial x^2} + \dots$$
(2.8)

$$p(x-\eta,t) = p(x,t) - \eta \frac{\partial p(x,t)}{\partial x} + \frac{\eta^2}{2} \frac{\partial^2 p(x,t)}{\partial x^2} + \dots$$
(2.9)

If we replace Eq. 2.7, 2.8 and 2.9 in Eq. 2.6, we get the diffusion equation

$$\frac{\partial p(x,t)}{\partial t} = \frac{1}{2} \frac{\partial^2 p(x,t)}{\partial x^2}$$
(2.10)

Diffusion equation with advection term

Einstein (1905) gave another derivation of the diffusion equation, starting from a continuous Markov process representing a Brownian motion. He considers many independent Brownian motions that take steps that are independent in time. Each particle has a transition probability $p(\delta x, \tau)$ for step length δx in the time interval τ . The concentration of particles at x and time $t + \tau$ is

$$C(x,t+\tau) = \int_{-\infty}^{+\infty} p(\delta x,\tau) C(x-\delta x,t) \, d\delta x, \qquad (2.11)$$

If we assume that the step size is very small compared to characteristic length scale for variations in C(x, t), and we do Taylor expanded of $C(x - \delta x, t)$ around C(x, t)

$$C(x - \delta x, t) = C(x, t) - \delta x \frac{\partial C(x, t)}{\partial x} + \frac{\delta x^2}{2} \frac{\partial^2 C(x, t)}{\partial x^2} + \frac{\delta x^3}{3!} \frac{\partial^3 C(x, t)}{\partial x^3} + \dots$$
(2.12)

We replace this equation in Eq. 2.11 and we put C(x,t), $\frac{\partial C(x,t)}{\partial x}$, $\frac{\partial^2 C(x,t)}{\partial x^2}$... out of the integrals, since they do not depend on δx , and we get

$$C(x,t+\tau) \approx C(x,t) \int_{-\infty}^{+\infty} p(\delta x,\tau) \ d\delta x - \frac{\partial C(x,t)}{\partial x} \int_{-\infty}^{+\infty} \delta x \ p(\delta x,\tau) \ d\delta x + \frac{\partial^2 C(x,t)}{\partial x^2} \int_{-\infty}^{+\infty} \frac{\delta x^2}{2} p(\delta x,\tau) \ d\delta x + \frac{\partial^3 C(x,t)}{\partial x^3} \int_{-\infty}^{+\infty} \frac{\delta x^3}{3!} p(\delta x,\tau) \ d\delta x + .(2.13)$$

We note that $\int_{-\infty}^{+\infty} p(\delta x, \tau) d\delta x = 1$ and $\langle \delta x^n \rangle = \int_{-\infty}^{+\infty} \delta x^n p(\delta x, \tau) d\delta x$ is the nth moment of the transition probability distribution. We replace in Eq. 2.13 and divide by τ to have

$$\frac{C(x,t+\tau) - C(x,t)}{\tau} = \frac{\partial C(x,t)}{\partial t} = -\frac{\langle \delta x \rangle}{\tau} \frac{\partial C(x,t)}{\partial x} + \frac{\langle \delta x^2 \rangle}{2\tau} \frac{\partial^2 C(x,t)}{\partial x^2} - \frac{\langle \delta x^3 \rangle}{3!\tau} \frac{\partial^3 C(x,t)}{\partial x^3} + \dots \quad (2.14)$$

If the second moment of the probability density is finite $\lim_{\tau\to 0} \langle \delta x \rangle / \tau$ and $\lim_{\tau\to 0} \langle \delta x^2 \rangle / 2\tau$ are constant and can be defined as

$$v = \lim_{\tau \to 0} \frac{\langle \delta x \rangle}{\tau}$$
 and $D = \lim_{\tau \to 0} \frac{\langle \delta x^2 \rangle}{2\tau}$ (2.15)

where v is a net drift (advection) of the distribution of walkers and D is the diffusion constant.

For smaller times $(t \sim \tau)$, the length scale based on diffusion $\delta x \sim \sqrt{2D\tau} = O(\tau^{\frac{1}{2}})$ is much greater than the characteristic scale based on advection $\delta x \sim v\tau$. It follows that $\langle \delta x^n \rangle \sim \tau^{\frac{n}{2}}$ and

$$\frac{\partial C(x,t)}{\partial t} + v \frac{\partial C(x,t)}{\partial x} = D \frac{\partial^2 C(x,t)}{\partial x^2} + O(\tau^{\frac{1}{2}})$$
(2.16)

For small times $\tau^{\frac{1}{2}} >> \tau$, so the $O(\tau^{\frac{1}{2}})$ term is significant and the fluctuations are dominant. But as $\frac{t}{\tau} \to \infty$, the fluctuations become insignificant and the $O(\tau^{\frac{1}{2}})$ term and higher orders become negligible. After neglecting the higher order terms, the diffusion equation with the drift terms is obtained.

2.2.2 Discrete sampling of a Brownian motion

Since the positions of our STE trajectories are known only every 6 h, we need to give some remarks on the discrete sampling of a stationary Brownian motion:

- increments for discrete time intervals are independent and stationary, since they are composed of infinitesimally small independent and stationary increments.
- increment distributions are Gaussian, as a direct consequence of the central limit theorem.
- these two remarks allow to consider a Brownian motion, when sampled on a discrete time axis, as a discrete time random walk with Gaussian stationary independent increments.

Furthermore, a discrete time random walk can be looked from the point of view of the number of steps, i.e. by considering the state of step indices, instead of the physical space. In that case, it becomes a simple random walk in 1-dimension, i.e. a discrete time random walk on a discrete space axis with probability to go upward given by p and to go downward given by q=1-p.

2.2.3 Auto-correlation function

One principal characteristic of random walks is the independence between successive increments, that is there is no correlation between two successive increments (or steps) of a walker. The correlation time is the time needed to obtain statistically independent steps. This time is obtained by computing the auto-correlation function at lag k for the ith term in the sequence

$$C(i,k) = \frac{\langle x_{i+k}x_i \rangle - \langle x_i \rangle^2}{\langle x_i^2 \rangle - \langle x_i \rangle^2},$$
(2.17)

 x_i is the ith step in the sequence, the mean is over all trajectories. C(i, k) is defined so that C(i, k = 0) = 1 and $C(i, k) \to 0$ if x_{i+k} and x_i are not correlated ($\langle x_{i+k} | x_i \rangle = \langle x_{i+k} \rangle \langle x_i \rangle$).

When we sum over i, we get the total auto-correlation function at lag k

$$C(k) = \frac{\sum_{i=1}^{N-k} (\langle x_{i+k} x_i \rangle - \langle x_i \rangle^2)}{\sum_{i=1}^{N-k} (\langle x_i^2 \rangle - \langle x_i \rangle^2)},$$
(2.18)

where N is the total number of steps.

2.2.4 Binomial law

We consider a walker that executes a series of random steps in a discrete vertical axis, with a probability p to go upward and q=1-p to go downward. If n_1 is the number of upward steps and n_2 is the number of downward steps, the total number of steps (N) and the distance walked (m) (in units of steps) are given by:

$$N = n_1 + n_2$$
 and $m = n_1 - n_2$. (2.19)

The probability to get n_1 upward steps and n_2 downward steps is given by the binomial distribution law :

$$P_N(n_1) = \frac{N!}{n_1!n_2!} p^{n_1} q^{n_2} = \frac{N!}{\left[\frac{N+m}{2}\right]! \left[\frac{N-m}{2}\right]!} p^{\frac{N+m}{2}} (1-p)^{\frac{N-m}{2}}$$
(2.20)

For large values of N this distribution law can be writing as a Gaussian Law:

$$P_N(m) = \frac{1}{\sqrt{2\pi\sigma^2}} e^{\frac{(m-\overline{m})^2}{2\sigma^2}}$$
 (2.21)

2.2.5 Characteristic properties of binomial law

From the binomial law we can deduce other statistic properties of random walk. The mean of upward steps is

$$\overline{n_1} = \sum_{n_1=1}^{N} P_N(n_1)n_1$$

$$= \sum_{n_1=1}^{N} {N \choose n_1} n_1 p^{n_1} q^{n_2}$$

$$= p \frac{\partial}{\partial p} \sum_{n_1=1}^{N} {N \choose n_1} p^{n_1} q^{n_2}$$

$$= p \frac{\partial}{\partial p} (p+q)^N$$

$$= p N (p+q)^{N-1} \qquad (2.22)$$

Since p+q=1, the mean of the total number of upward steps is

$$\overline{n_1} = pN. \tag{2.23}$$

Also we can derive that the mean of downward steps and the mean distance walked are

$$\overline{n_2} = qN$$
 and $\overline{m} = (p-q)N.$ (2.24)

For symmetric random walk p=q=1/2, and then the mean distance walked vanishes. For asymmetric random walk with higher probability to go downward (q > p), the slope of \overline{m} versus the total number of steps is negative.

With the same approach we can show that the variances are also linear with the total number of steps,

$$\frac{\sigma_m^2}{4} = \overline{m^2} - \overline{m}^2 = \sigma_{n_1}^2 = \sigma_{n_2}^2 = pqN.$$
(2.25)

2.3 Form tensor

To associate a flow pattern with a set of measured momentum, the simplest procedure is to construct a weighted flow tensor defined as (*Cugnon and L'Hote*, 1983):

$$T_{ij} = \sum_{\mu=1}^{M} \gamma(p^{(\mu)}) p_i^{(\mu)} p_j^{(\mu)}$$
(2.26)

with M the total number of trajectories, $\gamma(p^{(\mu)}) = 1, \frac{1}{m_{\mu}}, \frac{1}{p^{\mu}}, \frac{1}{p^{\mu^2}}$ is a weight factor and $p_i^{(\mu)}$ are Cartesian components of momentum (i, j = x, y, z).

The two variables associated with the shape of the ellipsoid are:

- The sphericity $S = \frac{3}{2}(1 \lambda_1)$.
- The coplanarity $C = \frac{\sqrt{3}}{2}(\lambda_2 \lambda_3).$

with $\lambda_1 \geq \lambda_2 \geq \lambda_3$ are the normalized eigenvalues of the flow tensor.

Figure 2.2 gives a schematic of the coplanarity versus the sphericity spectra. The red line indicates the case where the coplanarity is linear to sphericity ($\lambda_3 = 0$); In this case the form is planar. At low C and S we have a very elongated shape, and when C and S are very big we have a disc shape. For low coplanarities the shape changes from a very elongated shape to a Segar shape at intermediate S and to a spherical shape for large S.

The angle between the major axis of a flow tensor and x,y or z (see Figure 2.3) axis also can be used to study directed flows.





Figure 2.2: Coplanarity versus sphericity.

Figure 2.3: Schematic diagram showing a directed flow.

To study the evolution and dispersion of STE trajectories with time, using the form tensor, we need first to select trajectories that are associated to the initial divided flux. Figure 2.4 shows a schematic of cross-tropopause exchange fluxes. To select the same group of trajectories we use only trajectories that are located initially in the same area.

We select trajectories initially localized horizontally in a region defined by $\lambda_0 \pm \frac{\Delta \lambda}{2}$, $\phi_0 \pm \frac{\Delta \phi}{2}$ (see Figure 2.5). For each time interval $\Delta t = t_{i+1} - t_i$, we compute for each trajectory (μ) the velocity $\vec{V}_i^{(\mu)}$. After we calculate the flow tensor, and we diagnolize it to get the eigenvalues and the eigenvectors needed to compute the sphericity, coplanarity and the flow angles.

2.4 Diffusion and dispersion analysis

In this analysis we followed the motion of air trajectories, and we compute the ensemble average of the relative displacement $(r_i(t) - r_i(t = 0))$, with i = x, y, z) as a function of time.

In case of a purely diffusive transport the relative dispersion grows linearly with time as (*Weeks et al.*, 1996)

$$\sigma_x^2 = \langle x^2 \rangle - \langle x \rangle^2 = 2 \kappa t, \qquad (2.27)$$



Figure 2.4: Schematic illustrating some geographical distribution of crosstropopause exchange mass fluxes.

Figure 2.5: Schematic illustrating the calculation of the momentum tensor at time step $t_{i+1} - t_i$ for individual trajectories localized initially within the $\lambda_0 \pm \frac{\Delta\lambda}{2}, \phi_0 \pm \frac{\Delta\phi}{2}$ zone.

where the mean is the average over all trajectories. If we plot the dispersion versus time we obtain a direct estimation of the vertical diffusivity κ .

The relative dispersion of trajectories can be generalized by the formula ($Boffetta \ et \ al.$, 2001)

$$\langle x^2 \rangle \sim t^{2\nu} \tag{2.28}$$

where the brackets indicate the ensemble averaged. The value of the exponent ν defines the nature of transport involved; we can determine whether the diffusion is normal, $\nu = 0.5$, or anomalous $\nu \neq 0.5$. If $\nu > 0.5$ the spread grows with time faster than linearly. This is super-diffusion $(\lim_{t\to\infty} \kappa \approx \infty)$. If $\nu < 0.5$, then the spread grows with time slower than linearly. This is called sub-diffusion. For this regime of transport the mixing is slow and inefficient $(\lim_{t\to\infty} \kappa \approx 0)$.

2.5 Lyapunov analysis

To compute the finite size lyapunov exponent (FSLE) we select trajectories initially localized horizontally in a region $\lambda_0 \pm \frac{\Delta \lambda}{2}$, $\phi_0 \pm \frac{\Delta \phi}{2}$ (see Figure 2.5). There are two methods to calculate the FSLE. The first one use the radius of the cloud of points (*Artale et al.*, 1997; *Boffetta et al.*, 2001) and the second one uses the distance between two trajectories (*Lacorata et al.*, 2001).

For the first method, the relative dispersion is given by the mean square of the radius of the cloud at time t which is defined by

$$R^{2}(t) = \langle |\vec{r}(t) - \langle \vec{r}(t) \rangle |^{2} \rangle$$
(2.29)

where $\langle \vec{r}(t) \rangle = \frac{1}{N} \sum_{i=1}^{N} \vec{r}_i(t)$.

For the second method the relative dispersion is measured by the relative distance between two trajectories $\vec{r}^{(1)}$ and $\vec{r}^{(2)}$ at time t which is defined by

$$R^{2}(t) = |\vec{r}^{(1)}(t) - \vec{r}^{(2)}(t)|^{2}$$
(2.30)

To compute FSLE, the initial size of the set of N trajectories is given by:

$$\delta^{(0)} = \delta_0 = R(0) \tag{2.31}$$

with R(0) is done by Eq. 2.29 for the first method or by Eq. 2.30 for the second method. We define a series of scales $\delta = \{\delta_0, \delta_1, ..., \delta_n\}$, where $\delta_i = \rho \delta_{i-1} = \rho^i \delta_0$ and $\rho = \sqrt{2}$ is the doubling factor, and we compute the time T_n it takes for the size to grow from δ_n to $\delta \geq \delta_{n+1}$. The FSLE is defined by

$$\lambda(\delta_n) = \frac{1}{\langle T_n \rangle} \langle \ln(\frac{\delta}{\delta_n}) \rangle$$
(2.32)

the average is performed over each ensemble of trajectories.

The finite size lyapunov exponent at different δ reveals, some properties, the spreading of trajectories. In fact, it can be written as a power law (*Lacorata et al.*, 2001):

$$\lambda(\delta) \sim \delta^{-\alpha} \tag{2.33}$$

- For small size $\lim_{\delta \to 0} \lambda(\delta) = \lambda$, with λ is the maximum Lyapunov exponent.
- $\alpha = 2$ indicates diffusion regime.
- $\alpha < 2$ for super-diffusion regime where advection is important.
- $\alpha = 0, \lambda(\delta)$ is constant for chaotic advection.

Chapter 3

Analysis of STE trajectories

In the last chapter we described and define the methods that will be used in this chapter to study STE trajectories characteristics before, during and after the exchange. Here, we take the problem from a global perspective, with the emphasis on ensemble statistics.

3.1 Residence time

Figures 3.1 and 3.2 show $F(\tau)$ the fraction of trajectories that spent a time longer than a threshold τ (Eq. 2.1) for different seasons in case of STT and TST.

By convention, we will call $F(\tau)$ the residence time distribution in the following. We can see from these diagrams that F increases rapidly when $\tau \to 0$, and at high residence times F changes only slightly. Figure 3.1 shows that the stratospheric residence time distribution does not seem to change with seasons, whereas for the troposphere $F(\tau)$ varies slightly between winter and summer.

In Figure 3.2, residence time distributions have been rearranged to highlight differences between STT and TST (top panel) and differences between the stratosphere and the troposphere (bottom panel). From the top panels of Figure 3.2 we conclude that STT and TST events have the same stratospheric and tropospheric properties, respectively. Residence time properties in the stratosphere or in the troposphere do not depend on the direction of exchange. In bottom panels of Figure 3.2 we compare stratospheric and tropospheric distributions in summer and winter STT. It suggests



Figure 3.1: Fraction of trajectories that reside in the stratosphere (left) and the troposphere (right) a time longer than τ for both STT (top panels) and TST (bottom panels).

Figure 3.2: Fraction of trajectories that reside in the stratosphere and in the troposphere a time longer than τ . Top panels: Winter STT versus Winter TST for stratosphere (left) and troposphere (right). Bottom panels: stratospheric versus tropospheric distribution in summer (left) and in winter (right).

that the fraction of trajectories that reside in the troposphere a time longer than τ is bigger than in the stratosphere, or in other words, trajectories reside longer in the troposphere than in the stratosphere.

To understand the processes involved during the exchange of air masses between the stratosphere and the troposphere, we need first to know the nature of the transport that drives trajectories, and allow them to reside longer in the troposphere than in the stratosphere. Is this behavior a result of a slow diabatic descent superimposed to a diffusion process, or in other words, the result of a pure asymmetric diffusion?

Hall and Holzer (2003) suggested that the slope of these residence time distributions was due to the fact that the flux was computed in a regime dominated by diffusion.

3.2 Random walk theory and statistical properties of STE trajectories

The question we ask here is: can the behavior of stratospheric versus tropospheric trajectories observed in the bottom panels of Figure 3.2 be the result of purely asymmetric diffusive transport? Since we are placed in a Lagrangian perspective, the most appropriate approach is to use random walk theory.

Diffusive processes can be interpreted as a random walk. An asymmetric diffusive flow can be represented by a simple asymmetric 1-dimensional random walk, where steps in one direction occur with higher probability than steps in the opposite direction. In this section we will investigate the characteristics of trajectories. The random walk model is used to test if the computed spectra agree or disagree with diffusion process. For this purpose we will compare STE trajectory results with the formula developed in Section 2.2 of Chapter 2, or with random walk results simulated using the Monte-Carlo technique.

As a first step we will test if STE trajectories possess the basic properties of a simple random walk. Remember that our STE trajectories data set contains horizontal position, potential temperature, pressure and potential vorticity of the air parcels, traced for 9 days with time steps of 6 h. We focus only on the vertical motion of air mass trajectories in isentropic coordinates.

3.2.1 Statistical properties of STE trajectories

Auto-correlation functions

To study the memory along trajectory steps, in isentropic coordinates, the autocorrelation functions are used. The auto-correlation function at lag t (t = kx6 h) for the ith term in the sequence (Eq. 2.17 of Chapter 2) are plotted in Figure 3.3 for different values of i. The auto-correlation at i=0 represents the auto-correlation between the instant of the exchange and later times. Also the total auto-correlation functions (Eq. 2.18 of Chapter 2) are given in 3.4.

These functions take an initial value of 1, and decay rapidly to nearly 0 after 6 h. Hence, we can consider that the memory is lost for time intervals larger than 6 h. Then we have a process with independent steps every 6 h.



Figure 3.3: Auto-correlation between steps, for the ith term in the sequence at different i, versus time.

Figure 3.4: The total auto-correlation function between steps versus time.

Probability distributions

The method used to compute the relevant variables is as follows. We begin from the first extremity of the trajectory (first instant). A step is given by the difference between two successive values of potential temperature of the trajectory. For a given total number of steps that the trajectory executed we count how many times it goes upward or downward. A step is considered as upward when $\theta_{i+1} - \theta_i \ge 0$, and as downward when $\theta_{i+1} - \theta_i < 0$ during the interval of time $\Delta t = t_{i+1} - t_i = 6h$. Then, we compute the number of upward steps n_1 and the number of downward steps n_2 and the distance walked by the trajectory $m = n_1 - n_2$, as a function of the total number of steps $N = n_1 + n_2$. Note that this distance is taken in the space of step numbers, not the physical space, since step length is not considered.

Distributions of the distance walked for different total number of steps are shown in Figure 3.5. The dashed red lines indicate the results of the fit with the Gaussian law (Eq. 2.21). The fit is in good agreement with the data. We note that the spread of the distributions becomes larger as the total number of steps increases. This agrees with the fact that the deviation increases with the total number of steps, as shown in Eq. 2.25. we conclude here that the process is Gaussian.



Figure 3.5: Probability distributions of a distance walked m at different values of the total number of steps for data (histograms) and Monte-Carlo simulations (symbols). The dashed red line is the fit of data with a Gaussian law.

Other statistical properties

Figure 3.6 gives ensemble average values of total upward steps n_1 , downward steps n_2 , and their variances σ_1^2 and σ_2^2 , the distance walked m and its variance σ_m^2 versus the total number of steps N.

The first remark is that all variables are close to linear with the total number of steps. This suggests that the process is stationary in the space of the step number. From the slope of the mean of the number of upward and downward steps versus the total number of steps we can estimate upward and downward probabilities (Eqs. 2.23 and 2.24). We find that upward probability is p=0.469 and downward one is q=0.531, the sum of the two probabilities being equal to unity. The variances of n_1 and n_2 and the mean distance walked and its variance are shown in middle and bottom panels of Figure 3.6, respectively. The blue values are expected slopes when values of p=0.469 and q=0.531 are used in formulas 2.23, 2.24 and 2.25. The computed and the expected slopes are in good agreement.

Also we present in Figure 3.5 the distributions of m for Monte-Carlo simulations using the estimated values of p and q. The agreement between trajectories and simulations



Figure 3.6: Statistical properties computed for trajectories using potential temperature as vertical coordinate. Top panels: The mean number of upward steps n_1 (left) and downward steps n_2 (right) versus the total number of steps N. Middle panels: variance of upward steps σ_1^2 (left) and downward steps σ_2^2 (right) versus the total number of steps. Bottom panels: The mean distance walked m (left) and its variance σ_m^2 (right) versus the total number of steps. The red values give the slope of the fitted lines. The blue values give the expected slopes in case of asymmetric random walks with p=0.469 and q=1-p=0.531.

is good. Note that all these results reflect that trajectories have some characteristics of an asymmetric random walk, i.e. with a mean downward motion. This qualitatively agrees with residence time results of bottom panels of Figure 3.2 which suggested that trajectories reside longer in the troposphere than in the stratosphere.

3.2.2 Statistical properties of the tropopause

In the problem of characterizing STE trajectories, we are interested in the relative vertical motion between trajectories and the tropopause. We saw in the previous section that trajectories have basic characteristics of a stochastic process in θ coordinates. The question we addresses now is: what is the process that can describe the motion of the

tropopause? To answer this question for a given time and horizontal position of the trajectory we compute the vertical position of the tropopause in isentropic coordinates. We repeat the same analysis, but this time for the tropopause.

Auto-correlation functions

Figure 3.7 and Figure 3.8 show the auto-correlation function at lag t (t = kx6 h) for the ith term in the sequence (Eq. 2.17 of Chapter 2) and the total auto-correlation functions (Eq. 2.18 of Chapter 2), respectively. For the tropopause we see that there is anti-correlation equals to -0.25 at t=6 h, which becomes small at longer lag times. Although we will keep in mind this non-zero auto-correlation at t=6h, we will consider that, like trajectories, for a tropopause we have a process with independent steps every 6 h.



Figure 3.7: Auto-correlation between steps of tropopause, for the ith term in the sequence at different i, versus time.

Figure 3.8: The total auto-correlation function between steps of tropopause versus time.

Probability distributions

Distributions of the distance walked for different total number of steps are shown in Figure 3.9. The dashed red lines indicate the results of the fit with the Gaussian law (Eq. 2.21). The fit is in good agreement with the data, and again, we can consider the process to be Gaussian.



Figure 3.9: Probability distributions of a distance walked m at different values of the total number of steps for data (histograms) and Monte-Carlo simulations (symbols). The dashed red line is the fit of data with a Gaussian law.

Other statistical properties

Figure 3.10 gives ensemble average values of total upward steps n_1 , downward steps n_2 , and their variances σ_1^2 and σ_2^2 , the distance walked m and its variance σ_m^2 versus the total number of steps N.

This Figure shows that the tropopause has roughly the same characteristics as trajectories. We note however that the mean distance walked is not really linear with the total number of steps N. For high values of N the absolute value of the slope decreases. This suggests a saturation of the distance walked by the tropopause. In Figure 3.9 we plot the distributions of m for Monte-Carlo simulations using the estimated values of p and q. This illustrates the slight difference in \overline{m} and σ_m^2 between STE trajectories and Monte-Carlo simulations.

All these results suggest that tropopause has some characteristics of an asymmetric random walk, but also suggest some deviations from random walk model.



Figure 3.10: Statistical properties computed for tropopause using potential temperature as vertical coordinate. Panels and lines are same as Figure 3.6.

3.3 Stationarity of step length distributions

In order to link our simple random walk model to the Brownian motion, we need to check the stationarity of the step length distributions. To get information about the stationarity, the three first moments of the step length distributions $\Delta \theta = \theta_{i+1} - \theta_i$ will be given at different times, where t = 0 represents the exchange time between the troposphere and the stratosphere.

The mean, the standard deviation and the skewness $(\langle (x - \overline{x})^3 \rangle / \sigma^3)$ of air mass trajectories and tropopause are given in Figures 3.11 and 3.12, respectively. For air mass trajectories there is quasi-stationary distributions. The mean and standard deviation are quasi-constant. The skewness remains very close to zero. These results confirm the stationarity of the process as well as its Gaussianity. For the tropopause (Figure 3.12) the step distributions are not stationary with time. At the exchange (t=0) the tropopause moves upward for STT and downward for TST with larger step. The skewness values indicate very elongated distributions during the exchange.



Figure 3.11: The mean step $(\Delta \theta)$ of trajectories at different times before (t < 0)and after (t > 0) the exchange (open symbols). The shaded area shows the standard deviation, and the blue line gives the skewness of a distribution.

Figure 3.12: The mean step $(\Delta \theta)$ of a tropopause at different times before (t < 0) and after (t > 0) the exchange (open symbols). The shaded area shows the standard deviation, and the blue line gives the skewness of a distribution.

These results show that STE occurs in deformed regions of the tropopause. Similar results are obtained when using PV as a vertical coordinates (see Figure 3.13). In this case, since the tropopause is defined by the fixed 2PVU iso-surface, the deformation is seen in the trajectory distributions.

As a conclusion, the transport of trajectories across the isentropic surfaces is compatible with asymmetric Brownian process. Trajectories show stationarity and Gaussianity of step length distributions, and independence of steps. For the tropopause, even if it has some properties of random walk, the step distributions show some dynamical effects and non-stationarities.



Figure 3.13: The mean step (ΔPV) of trajectories at different times before (t < 0) and after (t > 0) the exchange (open symbols). The shaded area shows the standard deviation, and the blue line gives the skewness of a distribution.

3.4 Monte-Carlo and STE residence time distributions

Since we are interested on the relative motion between the tropopause and the trajectories, we need to know exactly the process that represents best the motion of the tropopause. Physically, the tropopause is not really free, so it cannot walk randomly without restrictions on the distance walked. Also the tropopause is not fixed in θ coordinates, it varies depending on latitude and time. Since we have not acquired a complete understanding of the process that the tropopause follows, we will investigate two extreme cases in order to compute the residence time distributions:

- * The tropopause as a random walker.
- * The tropopause as a fixed iso-surface.

3.4.1 The tropopause as a random walker

To compute the residence time distribution we use Monte-Carlo simulations. We need two walkers, the first one represents the trajectory and the second one represents the tropopause. The two walkers have a probability p=0.469 to go upward and a probability q=1-p=0.531 to go downward, and they are initially at position x=0. We compute the time τ (or the total number of steps) it needs for the two walkers to go to the same position. If the first step of the trajectory is bigger than the first step of the tropopause, the distribution of τ gives the residence time distribution in the stratosphere, otherwise it gives the residence time distribution in the troposphere. From the distribution of τ we get the distribution of the fraction of particles that reside a time τ or longer in the stratosphere or in the troposphere. The step length is chosen randomly between -1 and 1.





Figure 3.14: Fraction of trajectories that reside in the stratosphere/troposphere (lines) compared to distributions computed from Monte-Carlo (symbols) using values found of p and q. Black symbols show the results of symmetric random walk.

Figure 3.15: Fraction of trajectories that reside in the stratosphere/troposphere (lines) compared to distributions computed from Monte-Carlo (symbols) using values found of p and q. Black symbols show the results of symmetric random walk.

The results for both random walk theory and STE trajectories are compared in

Figure 3.14. In case of symmetric random walk (black symbols) the power of the distribution is 0.5. The simulated tropospheric and stratospheric distributions are identical to symmetric random walk results and disagree with STE trajectories residence time distributions. In a Monte-Carlo simulation, the relative motion of these two asymmetric random walkers (the trajectory and the tropopause) is a symmetric random walk.

When we compute the residence time distribution we suppose that the tropopause is a random walker which is not true because the tropopause is not free, it is localized in some altitudes (depending on latitude) and it can not go very far from these altitudes. Random walk theory assumes that there is stationarity and independence in the steps, but as seen in this chapter the tropopause fails to comply with these hypotheses.

3.4.2 The tropopause as a fixed iso-surface

In the Monte-Carlo simulations, we take an air mass (walker) initially at position $\theta = \theta_0$. We compute the time τ (or the total number of steps) it needs to the walker to return the first time to position $\theta = \theta_0$. We use p and q as estimated for STE trajectories. The results for both random walk theory and STE trajectories are compared in Figure 3.15. The distributions now agree very well.

Residence time distributions of STE are accurately simulated when the tropopause is assumed a fixed iso-surface in θ coordinates. We note that in case of symmetric random walk (black symbols in Figure 3.15) the power of the distribution is 0.5 but for asymmetric random walk the log-log spectra are not really linear, which means that the $\tau^{-1/2}$ divergence of the flux suggested by *Hall and Holzer* (2003) for diffusive processes is only manifested for symmetric transport (p=q=0.5).

Even if the residence time distributions are well simulated when we consider the tropopause as a fixed iso-surface, in reality its motion will be an intermediate process between a random walk and a fixed iso-surface. To simulate the tropopause correctly we need to investigate other processes. For example a martingale process, or just a fixed iso-surface with noise.

3.5 Form tensor and trajectories

Diffusion is fully described using two infinitesimal parameters: mean and standard deviation. However in the presence of anisotropy, dispersion can no longer be characterized so simply, but requires a tensor which fully describes mobility along each direction and correlation between these directions.

We consider a uniform spherical sample of particles (see Figure 3.16). In case where these particles are transported by a uniform flow (a), we get a same probability for particle dispersion in any direction. For a case where particles are transported by asymmetric flow (b), particles have higher probability to be dispersed in the $\vec{e_1}$ direction and a lower probability to be dispersed in the $\vec{e_2}$ direction. Differential advection can also affect the isotropy.



Figure 3.16: Redistribution of uniform particles resulting from: (a) Isotropic dispersion transport. Probability of particle dispersion is the same in every direction. (b) Anisotropic dispersion transport. Particles have a larger probability of being dispersed in the $\vec{e_1}$ (major eigenvector) direction of the ellipsoid.

The form tensor (Section 2.3 of Chapter 2) will be used to study directed flow signatures. We compute the angle between the major axis of the tensor with the

center of mass of the system (θ_{flow}) , and its angle with longitude (θ_x) and its angle with latitude (θ_y) . We use only trajectories located initially in the region $\Delta\lambda < 10^{\circ}$, $\Delta\phi < 10^{\circ}$. The distributions of these angles are shown in Figure 3.17 for different times. These distributions are enhanced at low angles for θ_{flow} and θ_x and at high angles for θ_y , i.e. the major axis of the tensor is directed towards the longitude axis and perpendicularly to the latitude axis. The coplanarity versus sphericity is linear and the tensor has a prolate form; the dispersion happens in a plane. All these results indicate that air mass volumes are dispersed horizontally with higher probability along the longitude axis. This can be the fact of the westerly wind that disperses trajectories more along the longitude axis.

Since there is large differences in length scales between the longitude and the latitude in one side, and between horizontal and vertical in other side, to get significant information about the transport process we need to investigate each direction separately.

3.6 Diffusion and dispersion analysis

The Lagrangian trajectories approach is used to characterize transport and mixing for passive tracers in the atmospheric flows. This approach has been proved to be a useful diagnostic tool to study transport and mixing processes. There is several statistical techniques to measure the separation of particles; correlation functions, relative dispersion and finite size Lyapunov exponent.

3.6.1 Relative dispersion

We investigate the ensemble average of the relative displacement $(r_i(t) - r_i(t = 0))$, with i = x, y, z of trajectories as a function of time. Our goal is to characterize stratosphere-troposphere transport as a function of zonal, meridional and vertical coordinates.

Vertical dispersion

From the analysis of Section 3.2 it is clear that the cross-tropopause trajectories are driven down by purely asymmetric diffusion in isentropic coordinates. In this case



 $\Delta \Phi < 10^{\circ}, \Delta \lambda < 10^{\circ}$

Figure 3.17: Left panel: The distributions of θ_{flow} (top), θ_x (middle) and θ_y (bottom) at different times. right panel: coplanarity versus sphericity spectra for different times.

the relative dispersion will grow linearly with time.

The relative dispersion curves in θ coordinates for STT and TST trajectories are shown in Figure 3.18. Using hydrostatic approximation we compute the dispersion spectra and the diffusivity coefficients along the z axis (Figure 3.19). The green lines are computed by fitting data using Eq. 2.27 of Chapter 2. As expected for standard diffusion, isentropic dispersion is typically well fitted by a linear growth law.

Estimated diffusivity coefficients for STT and TST are displayed in Table 3.1; these vertical diffusivities are in the same order of magnitude for the stratosphere and the troposphere and equal to $4-5 K^2 day^{-1}$. This dispersion technique to evaluate the vertical diffusivity gives approximately the same values when using balloon ozone sound-



Figure 3.18: The variance of crossisentropic displacement of trajectories in K^2 versus time for STT (left panel) and TST (right panel). Green lines give the fit results using Eq. 2.27.

Figure 3.19: The variance of latitude displacement of trajectories in km^2 versus time for STT (left panel) and TST (right panel). Green lines give the fit results using Eq. 2.27.

		Stratosphere	Troposphere
$\kappa_{ heta}$	\mathbf{STT}	4.5	3.9
$\left(K^2 day^{-1}\right)$	TST	4.6	5.3
κ_z	\mathbf{STT}	3.3	4.2
$(m^2 s^{-1})$	TST	2.6	4.7

Table 3.1: Vertical diffusivity coefficients

ings in the lower stratosphere (*Legras et al.*, 2003). The values of vertical diffusivities found by *Hegglin et al.* (2005), from seasonal distributions of CO in the LMS, are $\kappa_{\theta} \sim 0.45 - 1.1 \ m^2 s^{-1} \ (8.7-21.4 \ K^2 day^{-1}).$

Zonal dispersion

The results of calculating the relative dispersion along the longitudinal axis for STT and TST are shown in Figure 3.20. The Green lines are computed by fitting data using Eq. 2.28 of Chapter 2. Zonal dispersions are well fitted by the super-diffusion growth law since $\langle x^2 \rangle \sim t^{2\nu}$ and $\nu = 0.935 > 0.5$.

We note that the dispersion in the stratosphere is the same as in the troposphere, and STT gives the same results as TST.

Meridional dispersion

The meridional relative dispersion for STT and TST are shown in Figure 3.21. The Green lines are computed by fitting data using Eq. 2.28 of Chapter 2. The meridional dispersion of trajectories in the stratosphere is the same as in the troposphere, and STT gives the same results as TST. The dispersion behavior can be summarized as a combination of the super-diffusive growth regime at low times t < 24h ($\nu = 0.825 > 0.5$), and the sub-diffusive growth regime at high times t > 24h ($\nu = 0.28 < 0.5$).





Figure 3.20: Longitudinal relative displacement of trajectories versus time for STT (left panel) and TST (right panel). Green lines give the fit results using Eq. 2.28.

Figure 3.21: Meridional relative displacement of trajectories versus time for STT (left panel) and TST (right panel). Lines give the fit results using Eq. 2.28.

3.6.2 Finite Size Lyapunov Exponent

In this part we calculate the FSLE from STE trajectories in order to study their separation growth. For all the results presented in the following we use only trajectories located initially in the region $\Delta\lambda < 10^{\circ}$, $\Delta\phi < 10^{\circ}$.

We report the results for two initial radiuses: $\delta_0 = 200$ km and $\delta_0 = 500$ km. The FSLE analysis has been done as follows. We fixed a series of thresholds $\delta_n = \rho^n \delta_0$ $(\rho = \sqrt{2} \text{ and } n = 0, ...N)$. For the first technique we used (second technique), for each time t = 0 a new cloud of points (a new couple of trajectories) was considered with $\delta(t=0) \leq \delta_0$. The separation growth is then followed for times t > 0 and the doubling times T_n at scales δ_n are evaluated by measuring the time it takes for the size δ to grow from δ_n to $\delta \geq \delta_{n+1}$.

In Figure 3.22 we show FSLE $\lambda(\delta)$ for two values of δ_0 and using the technique of a cloud of points. Figure 3.23 shows FSLE $\lambda(\delta)$ using the separation between two trajectories. We can see that the dispersion of trajectories in the stratosphere is the same as in the troposphere, and STT gives the same results as TST. These results agree with the results of the relative dispersion. The exponent does not change a lot when we change δ_0 from 200 km to 500 km, and remain all times bigger than -2 which indicates a super-diffusive regime.



Figure 3.22: Finite size Lyapunov coefficient versus the size of the cloud for STT (left panel) and TST (right panel). The color of symbols refers to the initial cloud radius: $\delta_0 = 200$ km (red symbols), and $\delta_0 = 500$ km (blue symbols). Lines give the fit results. The blue symbols are multiplied by a factor 3.

Figure 3.23: Finite size Lyapunov coefficient versus the distance between two trajectories. Panels and lines are same as Figure 3.22.

These results agree with the results of tensor form and relative dispersion; We expect that longitudinal component of the separation between trajectories δ is the dominant and relative dispersion analysis show a super-diffusive process in this axis.

Conclusion

In this work, we studied extra-tropical stratosphere-troposphere exchange (STE) trajectories over a 10-year period. The aim of this study is to better describe global statistical properties of cross-tropopause trajectories, as well as the processes related to transport of mass between the troposphere and the stratosphere. Once these processes and statistical properties were established, we built the simplest model that captures the most important properties of STE.

Air parcel residence time distributions indicate that residence time properties in either the stratosphere or the troposphere do not depend on the direction of the exchange (stratosphere-to-troposphere transport, STT, or troposphere-to-stratosphere transport, TST). In addition, by comparing residence time properties we conclude that trajectories reside longer in the troposphere than in the stratosphere. This suggests that STT and TST trajectories are both driven by the same asymmetric flow, which in average drives down the air mass from the stratosphere to the troposphere.

By investigating the properties of trajectory motion in isentropic coordinates, we found that they agree with those of a random walker having probability p=0.469 to go upward and probability q=1-p=0.531 to go downward. In spite of the fact that some statistical properties of the tropopause agree with those of a random walker, we are not able to reproduce the residence time spectra using a simple random walk theory. This is because the tropopause step distributions are not stationary and they show some dynamical behavior like the deformation of the tropopause at exchange time. Further analysis will be needed in order to clarify the processes related to the motion of the tropopause.

Dispersion of trajectories in the atmosphere was investigated using different methods. Form tensor analysis indicates that trajectories are dispersed more horizontally with higher probabilities along the longitude axis. The technique of relative dispersion gave rise to different transport mechanisms in which the mean square displacement of air mass volumes grows linearly or non-linearly with time. The vertical dispersion along isentropic surfaces is well fitted with a linear growth law. This agrees with statistical analysis of trajectories that are compatible with diffusion processes. The extracted vertical diffusivities are equal to $4-5 K^2 day^{-1}$. Zonal dispersion analysis shows unusual transport in which the mean square displacement grows faster than a diffusion process: this is super-diffusion. Meridional relative dispersion behavior is a combination of two regimes. On short timescales (t < 24 h), dispersion is faster than diffusive processes (super-diffusion), while on long timescales (t > 24 h), relative dispersion grows more slowly than diffusion processes (sub-diffusion).

Dispersion properties are also investigated by means of the finite size Lyapunov exponent analysis. Both techniques show that dispersion grows faster than linearly with time.

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