Advective-diffusive mass flux and implications for stratosphere-troposphere exchange

Timothy M. Hall
NASA Goddard Institute for Space Studies and Columbia University, New York, New York, USA

Mark Holzer
Department of Earth and Ocean Sciences, University of British Columbia and Physics Department, Langara College, Vancouver, BC, Canada

Received 9 October 2002; revised 5 December 2002; accepted 22 December 2002; published 7 March 2003.

[1] We show that the flux of mass crossing in one direction (the "gross" flux) through any specified surface S that divides anadvective-diffusive flow in a closed domain is infinite. That is, the flux, $\mathcal{F}(\tau)$, through *S* of the fluid mass that spent at least time τ on one side of *S* diverges like $\tau^{-1/2}$ as $\tau \to 0$, in the continuum limit. The gross flux is completely dominated by fluid elements residing infinitesimally short times on one side of *S* before re-crossing to the other side. This general result puts into context the widely varying estimates of gross mass flux across the midlatitude tropopause. Such estimates are dominated by the smallest resolved scales, leading us to conclude that gross mass flux is not a useful diagnostic of stratosphere-troposphere exchange. The function $\mathcal{F}(\tau)$, however, provides important information on transport across the tropopause. *INDEX TERMS:* 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 3299 Mathematical Geophysics: General or miscellaneous; 3362 Meteorology and Atmospheric Dynamics: Stratosphere/troposphere interactions; 3399 Meteorology and Atmospheric Dynamics: General or miscellaneous. **Citation:** Hall, T. M., and M. Holzer, Advective-diffusive mass flux and implications for stratosphere-troposphere exchange, *Geophys. Res. Lett.*, *30*(5), 1222, doi:10.1029/2002GL016419, 2003.

1. Introduction

[2] Diffusion combined with advection is an appropriate description for a wide variety of fluid flows in the earth sciences [e.g., *Waugh and Hall*, 2002]. A common approach to characterizing transport in such systems is the estimation of fluid and trace constituent mass fluxes. For example, several recent studies (see below) have attempted to estimate the mass flux in one direction, the "gross" flux, across the extratropical tropopause. We deduce from general considerations, however, that the advective-diffusive gross flux of fluid mass across an arbitrary surface is infinite and, therefore, not a useful transport diagnostic. In practice estimates of gross flux are dominated by the smallest resolved scales of motion. We propose instead the estimation of the flux $\mathcal{F}(\tau)$ of fluid that resides for at least time τ on one side of the surface before re-crossing. The infinite gross flux emerges as a singularity at $\tau = 0$ while the behaviour of $\mathcal{F}(\tau)$ for finite τ allows for novel physical interpretation of the transport processes across the surface. We discuss the application to stratosphere-troposphere exchange (STE), a subject of intense current interest. More generally, our results are relevant to any advective-diffusive system in which the rate of fluid exchange between reservoirs is of interest.

[3] Considerable effort has gone into quantifying midlatitude STE [e.g., *Holton et al.*, 1995; *Appenzeller et al.*, 1996]. A number of studies conclude that the net flux is the difference of larger opposing gross fluxes [e.g., *Siegmund et al.*, 1996; *Wirth and Egger*, 1999; *Dethof et al.*, 2000; *Seo and Bowman*, 2001; *Wernli and Bourqui*, 2002]. Efforts to estimate the gross fluxes are motivated in part by the fact that the net flux of air mass is an incomplete diagnostic of tracer transport [*Gettelman and Sobel*, 2000]. Even if the net air mass flux is zero, opposing gross fluxes will cause tracer transport across the tropopause if the tracer has local gradients, for example due to chemical sources and sinks. However, estimates of the gross air mass flux vary widely. *Gettelman and Sobel* [2000] note a factor of four variation among various estimates of the ratio of downward to upward gross flux across the extratropical tropopause. Several of these estimates are based on the method of *Wei* [1987], who developed an expression for the advective flux through an arbitrary evolving surface. *Gettelman and Sobel* [2000] conclude that the estimation of gross fluxes using the *Wei* [1987] method is highly sensitive to inaccuracies in the underlying data from meteorological assimilation systems or general circulation models (GCMs). They show that space and time averaging can reduce the sensitivity, but such a procedure may suppress real transport events, because there is no clear choice of scale over which to apply the averaging.

[4] A different approach to calculate gross fluxes from meteorological data is to compute Lagrangian trajectories of particles, and to count the number of particles crossing a predefined tropopause. *Wernli and Bourqui* [2002] take this approach and find that the computed flux is highly sensitive to a "residence-time" threshold they use to include or exclude tropopause-crossing events. Particles spend a range of times ("residence times" in the nomenclature of *Wernli and Bourqui* [2002]) on one side of the tropopause before crossing back to the other side. If progressively shorter residence times are allowed in the calculation, the gross flux increases. No convergence with decreasing threshold is

0094-8276/03/2002GL016419$05.00

apparent in their analysis. *Sugata* [2000], applying a particle diagnostic to global tropospheric transport, also shows no convergence of gross flux with decreasing residence-time threshold. *Sobel et al.* [1997] apply a similar technique to transport across the stratospheric polar vortex and discuss the sensitivity to thresholds in detail.

[5] In the trajectory and *Wei* [1987] methods transport is treated as purely advective. Either one counts particle trajectories crossing the tropopause or sums velocity components normal to the tropopause, both of which vary in space and time at all scales, including the smallest that are resolved. We take an alternative approach based on the fact that cross-tropopause transport is fundamentally advective-diffusive. Although global assimilated meteorological data sets do not resolve the irreversible mixing of small-scale, three-dimensional turbulence, the smallest-scale resolved features of such data (order 1°) are still pseudo-random in character (e.g., tropopause folds, thin stratospheric filaments, cutoff cyclones). In addition, assimilated data sets contain noise from a variety of sources that leads to pseudo-random jitter in tropopause height [e.g., *Gettelman and Sobel*, 2000]. The diffusive effect of small-scale, pseudo-random processes on particle trajectories is well documented [e.g., *Bowman*, 1995]: While an individual particle experiences only advection, a cluster of particles disperses as particle trajectories diverge. A statistical description of the transport in terms of diffusion, Fickian or otherwise, becomes necessary.

[6] The high sensitivity of cross-tropopause gross-flux estimates to details of the particular analysis and underlying meteorological data is consistent with a natural feature of advective-diffusive flows that we derive here: the gross flux of fluid mass across any surface S separating an advective-diffusive fluid flow is infinite. In the continuum limit, the gross flux is completely dominated by fluid elements that cross S and reside infinitesimally short times on one side of S before re-crossing in the other direction. Relatedly, a commonly defined bulk timescale, the steady-state mass divided by the gross flux, is zero. (We use the term "turnover time" rather than "residence time" for this timescale to avoid confusion with the residence time of individual fluid elements.) Our result generalizes a similar observation made in the context of the stratospheric polar vortex by *Sobel et al.* [1997]. We conclude that gross flux and turnover time are not useful diagnostics of STE, and efforts to estimate them in hopes of better understanding the system are futile. Estimates of these quantities are dominated by the smallest resolved scales of motion and provide no information about passage of air across the tropopause of extended duration.

2. General Development

[7] We will show under general conditions that the gross flux through any surface dividing an advective-diffusive flow in a finite domain is infinite in the continuum limit. First, however, it is instructive to examine an extremely simple model that highlights the essential physics of our analysis. Because diffusion is the continuum limit of random particle motion, consider a one-dimensional lattice with spacing δx and an average particle number n per unit distance. Particles randomly move up or down by δx with equal probability 1/2 every timestep δt, and the entire lattice is translated, or "advected", upward with velocity u. The gross flux into $x = 0$ from $x = -\delta x$ is on average $F_{up} = n\delta x/(2\delta t) + nu$. In terms of the corresponding macroscopic diffusivity, $\kappa = (\delta x)^2/(2\delta t)$, this gross flux becomes $F_{up} = n\sqrt{\kappa/(2\delta t)} + nu$, or equivalently, $F_{up} = n\kappa/\delta x + nu$. Taking the continuum limit (δx, $\delta t \to 0$) such that n and κ remain constant, we see that F_{up} diverges like $\delta t^{-1/2}$, or equivalently, like δx^{-1}. Thus, at any finite resolution, F_{up} is dominated by motion at the shortest space and timescales. Meanwhile, the same argument applies to the downward flux $F_{dn} = n\delta x/(2\delta t)$, which for this simple model is purely diffusive, so that the net flux $F_{up} - F_{dn} = nu$ remains finite. We note that there is a close connection with the subject of first-passage times [*Montroll and West*, 1979], where similar singularities appear.

[8] We now consider advective-diffusive fluid flow in generality for a domain R of finite mass. R is separated by a surface S into two sub-domains R_1 and R_2. The surface S is purely a conceptual construction; it has no effect on the fluid transport, and it need not have any relationship to the flow. The fluid is imagined to consist of infinitesimal elements that maintain their identity against mixing for all timescales of interest. Diffusion is manifest as random motions of the fluid elements. Only properties averaged over the large number of fluid elements in a "parcel" can be measured.

[9] To compute the flux of fluid mass across S we make use of a hypothetical tracer with idealized sources and boundary conditions, namely, the Green's function, G. The analysis exploits the techniques of *Holzer and Hall* [2000]. The Green's function $G(\mathbf{r}, t|\mathbf{r}', t')$ is the response at position $\mathbf{r}$ and time t to a unit mass source of tracer, $\rho^{-1}\delta(t - t')\delta(\mathbf{r} - \mathbf{r}')$, where ρ is the fluid density, and $\mathbf{r}'$ is in R_2. Tracer "marks" fluid elements as having been at $\mathbf{r}'$ at t'. The quantity $G(\mathbf{r}, t|\mathbf{r}', t')\,\rho(\mathbf{r})\,d^3r$ is the mass fraction of the tracer-marked fluid elements contained in the fluid parcel of mass $\rho(\mathbf{r})\,d^3r$ at $\mathbf{r}$ and t. The marked fraction in all R_2 at elapsed time ("residence time") $\tau = t - t'$ is

$$P(t' + \tau|\mathbf{r}', t') = \int_{R_2} d^3r\rho(\mathbf{r})G(\mathbf{r}, t' + \tau|\mathbf{r}', t'). \quad (1)$$

The evolution of $P(t' + \tau|\mathbf{r}', t')$ depends on the tracer boundary conditions (BCs) on S. Consider a BC of zero mixing ratio, that is, $G(S, t' + \tau|\mathbf{r}', t') = 0$. On the other boundaries of R_2, G complies with the natural no-flux conditions of the fluid. With these BCs, tracer marking a fluid element indicates that the fluid element has not made contact with S since being at $\mathbf{r}'$. When a fluid element makes S contact its tracer marking is removed by the BC. Therefore, $P(t' + \tau|\mathbf{r}', t')$ declines from unity in elapsed time, as tracer-marked fluid elements make contact with S. $P(t' + \tau|\mathbf{r}', t')$ is the fraction of fluid mass that has not made S contact by $t' + \tau$, given that it was at $\mathbf{r}'$ at t'. Now, $d^3r'\rho(\mathbf{r}')\,P(t' + \tau|\mathbf{r}', t')/M_2$ is the fraction of fluid mass in R_2 that was in d^3r' about $\mathbf{r}'$ at t' and has not made S contact by $t' + \tau$, where M_2 is the total fluid mass in R_2. Thus, the fraction of fluid mass *anywhere* in R_2 at t' that resides at least time τ before exiting, $\mathcal{P}(t' + \tau|t')$, is obtained by integrating over R_2:

$$\mathcal{P}(t' + \tau|t') = \frac{1}{M_2}\int_{R_2} d^3r'\rho(\mathbf{r}')P(t' + \tau|\mathbf{r}', t'). \quad (2)$$

Finally, the flux $\mathcal{F}(t' + \tau|t')$ out of R_2 of the mass fraction of fluid that has resided in R_2 at least time τ is

$$\mathcal{F}(t'+\tau|t') = -\frac{\partial}{\partial\tau}\mathcal{P}(t'+\tau|t'). \quad (3)$$

[10] In order to show that $\mathcal{F}(t' + \tau|t')$ is singular at $\tau = 0$, we first rewrite it by using (1), (2), (3) and certain properties of Green's functions. In the following, $\kappa(\mathbf{r})$ is the local diffusivity, and $\mathbf{n}$ is the outward normal to S.

$$\mathcal{F} = -\frac{1}{M_2}\int_{R_2} d^3r'\rho(\mathbf{r}')\frac{\partial}{\partial\tau}\int_{R_2} d^3r\rho(\mathbf{r})G(\mathbf{r},t'+\tau|\mathbf{r}',t') \quad (4)$$

$$= -\frac{1}{M_2}\int_{R_2} d^3r'\rho(\mathbf{r}')\int_S d^2r\rho(\mathbf{r})\kappa(\mathbf{r})\mathbf{n}\cdot\nabla_r G(\mathbf{r},t'+\tau|\mathbf{r}',t') \quad (5)$$

$$= -\frac{1}{M_2}\int_{R_2} d^3r'\rho(\mathbf{r}')\int_S d^2r\rho(\mathbf{r})\kappa(\mathbf{r})\mathbf{n}\cdot\nabla_r G^{\dagger}(\mathbf{r}',t'|\mathbf{r},t'+\tau) \quad (6)$$

$$\equiv -\frac{1}{M_2}\int_S d^2r\rho(\mathbf{r})\kappa(\mathbf{r})\mathbf{n}\cdot\nabla_r P^{\dagger}(t'|\mathbf{r},t'+\tau). \quad (7)$$

In (5) the rate of change of tracer-marked mass has been replaced by the flux through S. Note that the flux is purely diffusive. Any advective component $\mathbf{v}G$ vanishes on S by construction, because of the BC chosen. In (6) we have used the reciprocity condition for Green's functions: $G(\mathbf{r}, t' + \tau|\mathbf{r}', t') = G^{\dagger}(\mathbf{r}', t'|\mathbf{r}, t' + \tau)$ [*Morse and Feschbach*, 1953], where the dagger indicates the adjoint. The reciprocity condition states that the response at $\mathbf{r}$ to a point source at $\mathbf{r}'$ in the normal time-forward flow equals the response at $\mathbf{r}'$ to a point source at $\mathbf{r}$ in the time-reversed adjoint flow (TRAF). Expression (7) defines $P^{\dagger}(t'|\mathbf{r}, t' + \tau)$, the TRAF analog of (1). $P^{\dagger}(t'|\mathbf{r}, t' + \tau)$ is the mass fraction in all R_2 at time t' of fluid marked by tracer at $\mathbf{r}$ at $t' + \tau$ in the TRAF.

[11] This final form for $\mathcal{F}(t' + \tau|t')$ can now be analyzed using an argument from *Holzer and Hall* [2000]. Consider the behavior of $P^{\dagger}(t'|\mathbf{r}, t' + \tau)$ at small τ. For any $\mathbf{r}$ not on S, $\lim_{\tau\to 0} P^{\dagger}(t'|\mathbf{r}, t' + \tau) = 1$. At early enough elapsed times the tracer released at $\mathbf{r}'$ is still localized about $\mathbf{r}'$, and thus all tracer-marked fluid elements remain in R_2. For $\mathbf{r}$ on S, however, $P^{\dagger}(t'|\mathbf{r}, t' + \tau) = 0$ by the BC. Therefore, as $\tau \to 0$ the dependence of P on $\mathbf{r}'$ becomes discontinuous at S, the normal gradient of P on S becomes infinite, and, thus, from (7), $\lim_{\tau\to 0} \mathcal{F}(t' + \tau|t') = \infty$. In other words *the flux into S of the mass fraction of fluid elements that have resided for any time in* R_2 (*the gross flux out of* R_2) *is infinite*.

[12] We can further infer that $\mathcal{F}(t' + \tau|t')$ diverges like $1/\sqrt{\tau}$, that is, as $\tau \to 0$,

$$\mathcal{F}(t'+\tau|t') \to \frac{1}{\sqrt{\tau}}\frac{1}{M_2}\int_S d^2r'\rho(\mathbf{r}')\sqrt{\kappa(\mathbf{r}')}. \quad (8)$$

For small τ tracer-marked fluid elements must start at $\mathbf{r}'$ within a correspondingly small distance ℓ from S, for $P^{\dagger}(t'|\mathbf{r}', t' + \tau)$ to differ significantly from unity; i.e., for a significant probability of loss in elapsed time τ. This is the distance over which fluid elements can be diffusively transported in time τ, namely $\ell \sim \sqrt{\kappa\tau}$. Thus, $\nabla P^{\dagger} \sim 1/\sqrt{\kappa\tau}$, and (8) follows from (7).

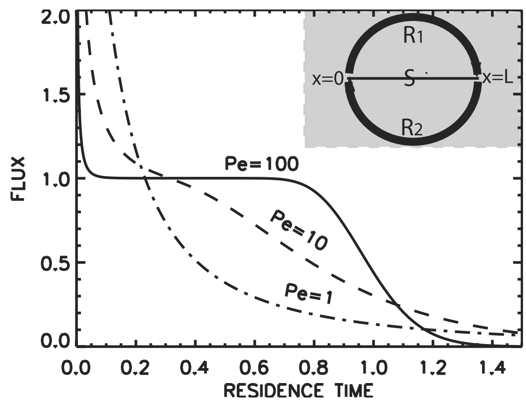

Figure 1. The gross flux $\mathcal{F}$ versus residence time, τ, for the advective-diffusive loop model at three Péclet numbers. $\mathcal{F}$ is in units of $\rho\, u/M_2$ and τ is in units of L/u. A model schematic is shown in the inset. Advection is clockwise, and diffusion occurs along the flow. S divides the domain into R_1 and R_2, and the x coordinate runs along the upper half loop.

3. Idealized Illustration

[13] We have shown the gross flux to be infinite for general advective-diffusive flows. $\mathcal{F}(t' + \tau|t')$, however, depends on the particular flow. It is instructive to examine an advective-diffusive flow that has an analytic solution for $\mathcal{F}$, so that the limiting behavior as $\tau \to 0$ can be explored. A highly idealized flow with such a solution is the "advective-diffusive loop" illustrated in Figure 1. Fluid of constant density ρ advects around a loop at constant speed u, and diffusion occurs along the flow at constant diffusivity κ. The domain is separated by the "surface" S into R_1 and R_2 with masses M_1 and M_2. The model can be imagined to represent an eddy crossing a fixed tropopause at midlatitudes on an isentropic surface.

[14] To good approximation a convenient non-dimensionalized form for the gross flux across S is

$$\tilde{\mathcal{F}}(\tilde{\tau}) \approx \frac{e^{-\tilde{\tau}/4}}{\sqrt{\pi\tilde{\tau}^3}}\int_0^{Pe}\tilde{x}\,\cosh\left(\frac{\tilde{x}}{2}\right)e^{-\tilde{x}^2/4\tilde{\tau}}d\tilde{x}, \quad (9)$$

where $Pe = uL/\kappa$ is the Péclet number, $\tilde{\tau} = \tau u^2/\kappa$, $\tilde{x} = xu/\kappa$, and $\tilde{\mathcal{F}} = \mathcal{F}M_2/(\rho u)$. (To obtain (9) we exploit the relationship derived by *Holzer and Hall* [2000] between (5) and the "age spectrum" of *Hall and Plumb* [1994], then apply standard methods to compute the age spectrum.) As $\tilde{\tau} \to 0$, the integrand is dominated by small Péclet number $\tilde{x}$, that is, the small-time limit is dominated by diffusive transport. For small $\tilde{\tau}$ we can replace the upper limit of integration with ∞ to obtain $\tilde{\mathcal{F}}(\tilde{\tau}) \to 2/\sqrt{\pi\tilde{\tau}}$.

[15] $\mathcal{F}$ is plotted in Figure 1 as a function of residence time, τ, for several Pe. The $\tau^{-1/2}$ divergence is only manifest for $\tau < \kappa/u^2$, when diffusive contact with S dominates. At large Pe, the timescale, $\kappa/u^2 = (1/Pe)(L/u)$, and the advective timescale, L/u, are well separated, allow-

ing $\mathcal{F}$ to develop a plateau. The plateau results from fluid elements penetrating far enough into R_2 to get caught up in the advective circulation around the loop. For $\tau > L/u$, $\mathcal{F}$ declines to zero. In the limit of large Pe, $\mathcal{F}$ approaches a box-car shape, but a vanishingly thin divergence at $\tau = 0$ remains. Note, however, that at $Pe = 100$, fluid elements can reside longer than L/u due to accumulated time spent moving back and forth diffusively. As Pe is lowered to 10 and 1, the timescales $(1/Pe)(L/u)$ and L/u are no longer well separated, and no advective plateau develops.

4. Stratosphere-Troposphere Exchange

[16] We have shown on general grounds that "gross" flux, the flux passing in one direction across an arbitrary surface S dividing an advective-diffusive fluid flow, is infinite in the continuum limit. That is, the flux $\mathcal{F}(t' + \tau|t')$ of mass that is on one side of S at t' and resides at least time τ on that side diverges as $1/\sqrt{\tau}$ for small τ. Relatedly, the "turnover time" (steady state mass/gross flux), is zero. This result has implications for the estimation of air-mass flux in either direction across the extratropical tropopause. Such flux can occur isentropically via synoptic and smaller-scale events [e.g., *Holton et al.*, 1995]. Although in many studies the flux has been treated as purely advective (e.g., using the method of *Wei* [1987] or particle trajectories as in *Wernli and Bourqui* [2002]), the mixing due to both resolved and unresolved motions renders the flow effectively advective-diffusive. In fact, if the tropopause is defined in a mass-following coordinate system, then the flux must be purely diffusive [*Nakamura*, 1996]. It is perhaps not surprising, therefore, that gross flux estimates vary widely. Differences in temporal resolution and time "thresholds" for including tropopause-crossing events translate to different values of τ at which to evaluate $\mathcal{F}(t' + \tau|t')$. The fact that many studies show high sensitivity of the estimated gross flux to such thresholds [e.g., *Gettelman and Sobel*, 2000; *Wernli and Bourqui*, 2002] suggests that the studies are evaluating $\mathcal{F}$ in a regime of rapid variation with τ, that is, a regime effectively dominated by diffusion.

[17] The space and time dependencies of the cross-tropopause transport are linked. The singular flux at infinitesimal residence time τ corresponds to fluid elements that have penetrated only infinitesimal distances across the tropopause. This is consistent with *Dethof et al.* [2000] and *Seo and Bowman* [2001] who find, using different analyses of meteorological data, that the distribution of penetration depth of stratosphere-troposphere exchange peaks sharply at zero.

[18] The singularity at $\tau = 0$ implies that gross flux is not a useful diagnostic of stratosphere-troposphere exchange. We advocate the estimation from meteorological data of the full function $\mathcal{F}(t' + \tau|t')$, rather than a single value at small τ. $\mathcal{F}$ may be well suited for analyzing the effects of stratosphere-troposphere exchange on the chemical environment of the upper troposphere and lower stratosphere. Given a timescale τ_C for a chemical reaction on one side of the tropopause, $\mathcal{F}(t' + \tau_C|t')$ is the fraction of air crossing the tropopause at t' that will reside at least τ_C and, therefore, undergo significant transformation. Note that $\mathcal{F}(t' + \tau|t')$ is a non-increasing function of τ. However, if Pe is sufficiently large then there may be one or more plateaus (as in Figure 1) that could be revealed by as few as two data points ($\mathcal{F}$, τ), either for a specific calendar time t' or for an ensemble average. At small τ, given the generic form of the divergence, it may be possible to infer an effective κ near the tropopause from estimates of $\mathcal{F}$. The calculation of $\mathcal{F}$ should be a relatively straightforward extension of several studies already performed. For example, when using particle crossings with residence-time thresholds, τ, [e.g., *Wernli and Bourqui*, 2002] the computed flux as a function of the threshold is, in fact, $\mathcal{F}(t' + \tau|t')$.

[19] **Acknowledgments.** We thank Adam Sobel for discussions, Birger Bergensen for pointing out the reference on first-passage times, and the anonymous reviewers for their comments. TMH acknowledges support from NASA grant 622-58-19-31.

References

Appenzeller, C., J. R. Holton, and K. H. Rosenlof, Seasonal variations of mass transport across the tropopause, *J. Geophys. Res.*, *101*, 15,071–15,078, 1996.
Bowman, K. P., Diffusive transport by breaking waves, *J. Atmos. Sci.*, *52*, 2416–2427, 1995.
Dethof, A., A. O'Neill, and J. Slingo, Quantification of the isentropic mass transport across the dynamical tropopause, *J. Geophys. Res.*, *105*, 12,279–12,293, 2000.
Gettelman, A., and A. H. Sobel, Direct diagnosis of stratosphere-troposphere exchange, *J. Atmos. Sci.*, *57*, 3–16, 2000.
Hall, T. M., and R. A. Plumb, Age as a diagnostic of stratospheric transport, *J. Geophys. Res.*, *99*, 1059–1070, 1994.
Holton, J. R., P. H. Haynes, M. E. McIntyre, A. R. Douglass, R. B. Rood, and L. Pfister, Stratosphere-troposphere exchange, *Rev. Geophys.*, *33*, 403–439, 1995.
Holzer, M., and T. M. Hall, Transit-time and tracer-age distributions in geophysical flows, *J. Atmos. Sci.*, *57*, 3539–3558, 2000.
Montroll, E. W., and B. J. West, On an enriched collection of stochastic processes, in *Studies in Statistical Mechanics*, vol. VII, *Fluctuation Phenomena*, edited by E. W. Montroll and J. L. Lebowitz, North-Holland, New York, 1979.
Morse, P. M., and H. Feschbach, *Methods of Theoretical Physics*, 997 pp., McGraw-Hill, New York, 1953.
Nakamura, N., Two-dimensional mixing, edge formation, and permeability diagnosed in an area coordinate, *J. Atmos. Sci.*, *53*, 1524–1537, 1996.
Seo, K.-H., and K. P. Bowman, A climatology of isentropic cross-tropopause exchange, *J. Geophys. Res.*, *106*, 28,159–28,172, 2001.
Siegmund, P. C., P. F. J. van Velthoven, and H. Kelder, Cross-tropopause transport in the extratropical northern winter hemisphere diagnosed from high resolution ECMWF data, *Q. J. R. Meteorol. Soc.*, *122*, 1921–1941, 1996.
Sobel, A. H., R. A. Plumb, and D. W. Waugh, Methods of calculating transport across the polar vortex edge, *J. Atmos. Sci.*, *54*, 2241–2260, 1997.
Sugata, S., Time threshold diagnostics: A mixed Lagrangian-Eulerian method for describing global tracer transport, *J. Meteorol. Soc. Jpn.*, *78*, 259–277, 2000.
Waugh, D. W., and T. M. Hall, Age of stratospheric air: Theory, observations, and models, *Rev. Geophys.*, *40*(4), 1010, doi:10.1029/2000RG000101, 2002.
Wei, M. Y., A new formulation of the exchange of mass and trace constituents between the stratosphere and troposphere, *J. Atmos. Sci.*, *44*, 3079–3086, 1987.
Wernli, H., and M. Bourqui, A Lagrangian "1-year climatology" of (deep) cross-tropopause exchange in the extratropical Nothern Hemisphere, *J. Geophys. Res.*, *107*(D2), 4041, doi:10.1029/2001JD000812, 2002.
Wirth, V., and J. Egger, Diagnosing extratropical synoptic-scale stratosphere-troposphere exchange: A case study, *Q. J. R. Meteorol. Soc.*, *125*, 635–655, 1999.

T. M. Hall, NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA. (thall@giss.nasa.gov)
M. Holzer, Department of Earth and Ocean Sciences, University of British Columbia, 6339 Stores Road, Vancouver, British Columbia, Canada V6T 1Z4. (mholzer@eos.ubc.ca)