The Transit-Time Distribution from the
Northern Hemisphere Midlatitude Surface

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ABSTRACT

The distribution of transit times from the Northern Hemisphere (NH) mid-latitude surface is a fundamental property of tropospheric transport. Here we present an analysis of the transit time distribution (TTD) since air last contacted the NH midlatitude surface layer, as simulated by the NASA Global Modeling Initiative Chemistry Transport Model. Throughout the troposphere the TTD is characterized by long flat tails. This results in mean ages (Γ) that are significantly larger than their corresponding modal ages (τ_mode), particularly in the NH, where Γ ~ 0.5 years while τ_mode < 20 days. Furthermore, the ratio of the spectral width (Δ) to the mean age decreases sharply from ~ 2 at NH high latitudes to less than ~ 0.7 in the Southern Hemisphere, which reflects large differences in the contributions of fast transport paths relative to slow eddy-diffusive recirculations. It is shown that the spatial patterns and seasonal cycles of idealized tracers exponentially decaying at rates of 5 days⁻¹ and 50 days⁻¹ reflect the behaviors of the modal age and mean age, respectively. This suggests that suites of tracers undergoing atmospheric loss at different rates may be used to constrain the TTD from observations. Overall, our results indicate that current diagnostics of tropospheric transport are not sufficient for assessing and comparing model transport, as they do not capture the broad range of timescales that connect the free troposphere to the NH midlatitude surface.
1. Introduction

The Northern Hemisphere (NH) midlatitude surface is a source of major greenhouse gases, ozone depleting substances, tropospheric ozone (and its precursors) and aerosols. Understanding the factors that control present and future distributions of species emitted from NH midlatitudes is central to predicting tropospheric air quality, stratospheric ozone depletion, and changes in the earth’s radiative balance. This is complicated, however, by the fact that species distributions reflect the complex interplay between their emissions, chemistry and transport, which are difficult to disentangle even for long-lived trace species. In particular, many aspects of tropospheric transport are still not well understood, which has limited the ability for numerical models to accurately represent carbon monoxide, tropospheric ozone, and black carbon, among other species (e.g., Shindell et al. 2008; Young 2013; Lee et al. 2013).

It has long been appreciated that tropospheric transport is captured neither by conventional Eulerian-mean diagnostics nor the Lagrangian mean (Andrews and McIntyre 1978) and residual mean circulations (Andrews and McIntyre 1976). Rather, the transport circulation, which is not directly observable but can be approximated using numerical models, reflect the combination of the slow mean diabatic circulation and rapid isentropic mixing (Plumb and Mahlman 1987). This has been shown in both Eulerian and Lagrangian approximations of the tropospheric transport climate (e.g., Holzer 1999; Bowman and Erukhimova 2004; Stohl 2006) and recent studies of extratropical mixing in the lower and middle troposphere that have expanded traditional eddy diffusivity diagnostics to account for both diabatic diffusion (Leibensperger and Plumb 2014) and near-surface isentropic flow (Chen and Plumb 2014). While these studies have contributed significantly to our current understanding of tropospheric transport, however, many fundamental aspects of the transport circulation remain poorly understood.
One aspect of transport that is particularly important for chemical species is the time that it takes for transport to occur from the NH midlatitude surface to different regions in the troposphere. Typical timescales that have been used to assess transport include gross interhemispheric exchange times (e.g., Levin and Hesshaimer 1996; Geller et al. 1997) which have been inferred from measurements of various chlorofluorocarbons and sulfur hexafluoride (SF6) and interpreted in terms of the strength and position of tropical convection (Lintner et al. 2004) and the zonally averaged Hadley circulation (Bowman and Erukhimova 2004).

More recently, Waugh et al. (2013) use surface measurements of SF6 to infer the “mean age” or the average time since the air in different regions in the troposphere last contacted the NH midlatitude surface. As the mean age is calculated for locations throughout the troposphere it provides more information on transport than the interhemispheric exchange time. In addition, while the mean age is equal to the SF6 age (Patra et al. 2009), it can be calculated in models independent of SF6 and, thus, does not reflect intermodel differences in prescribed emissions, which are often validated using observational datasets that vary between each other.

The mean age provides a convenient description of tropospheric transport in terms of a single timescale. Because of irreversible diffusive mixing, however, there is no single timescale or unique pathway for transport to occur from the surface to the interior, but rather a distribution of times and paths. In this sense, the mean age is an incomplete diagnostic by representing an average over a range of timescales. By comparison, the transit-time distribution (TTD) – the distribution of transit times since last surface contact – contains complete information about the flow, where “transit-time” refers to the elapsed time since last contact occurred at the surface (here, NH midlatitudes).

The TTD has been shown to be a far more powerful diagnostic than the mean age in understanding transport in the oceans (e.g., Primeau and Holzer 2006; Holzer and Primeau 2010) and the stratosphere (e.g., Bönisch et al. 2009), where it is referred to commonly as the age spectrum.
(Hall and Plumb 1994; Waugh and Hall 2002). Along with providing complete information about the transport, the TTD has been used to infer the distributions and evolution of chemical species that are difficult to measure directly, including total chlorine in the stratosphere (e.g., Waugh et al. 2001) and anthropogenic carbon dioxide in the oceans (e.g., Hall et al. 2002; Waugh et al. 2006; Khatiwala et al. 2009).

In the troposphere, where measurements of trace species are more widely available, the TTD has been used mainly to understand various aspects of tropospheric transport, including inter-hemispheric exchange times and transport pathways (Holzer and Boer 2001; Holzer 2009) and transpacific transport (Holzer et al. 2003, 2005; Holzer and Hall 2007). Its applications to troposphere chemistry, by comparison, have been relatively limited. Recent work by Holzer and Waugh (2015), however, shows that the TTD can be used to infer species’ path-dependent lifetimes, which unlike global lifetimes, measure the integrated loss that a tracer experiences during its transit through the troposphere to a point of measurement and, therefore, carry information about the transport paths through the loss field. Thus, the TTD may provide not only a rigorous way to assess and compare model transport but also estimates of path-dependent chemical loss that may improve constraints on the emissions of greenhouse gases and ozone depleting substances, among other applications.

Here we focus on transport from the NH midlatitude surface, which we define as latitudes spanning 30°N-50°N to approximate the region of largest emissions of greenhouse gases and ozone depleting substances. We present the transit-time distribution connecting the NH midlatitude surface to the free troposphere using the NASA Goddard Modeling Initiative Chemistry Transport Model driven with MERRA reanalysis meteorological fields. We also discuss how the TTD can be used to interpret the distributions of tracers subject to spatially uniform first-order loss and, conversely, how tracers that experience loss can be used to infer different aspects of the TTD. Fol-
Following a brief exposition of the methodology in Section 2 we present the model results in Sections 3-5, followed by conclusions in Section 6.

2. Methods

a. Model Simulations

We use the NASA Goddard Modeling Initiative (GMI) three-dimensional chemistry transport model (CTM) (Strahan et al. 2007), which has a horizontal resolution of 2 degrees latitude by 2.5 degrees longitude with 72 vertical levels spanning the surface to 0.01 hPa. We analyze one integration of the CTM driven with MERRA reanalysis meteorological fields for the years 2000-2010 (Rienecker et al. 2011), which is identical to the simulation used in Waugh et al. (2013). That study, however, only presented calculations of the mean age, not the TTD.

We examine two types of tracers that are carried in the integration: tracers that are used to infer the TTD and two idealized tracers subject to spatially uniform first-order loss. All tracers’ boundary conditions are defined over a common surface NH midlatitude region $\Omega_{\text{MID}}$, defined as the first model layer spanning latitudes between 30$^\circ$N and 50$^\circ$N. The idealized decay tracers, referred to throughout as the 5-day and 50-day tracers, are emitted uniformly over $\Omega_{\text{MID}}$ and undergo spatially uniform exponential decay at loss rates of 5 days$^{-1}$ and 50 days$^{-1}$, respectively.

We analyze the decay tracer values in the last year of the integration (2010), at which point they have reached a statistically stationary state. These tracers are identical to the idealized loss tracers that were requested in the most recent Chemistry-Climate Model Initiative (CCMI) and, therefore, provide a means for comparing the transport of the GMI CTM with other models. The tracers used to approximate the TTD are described next.
b. TTD Calculations

The TTD is a propagator of boundary conditions applied over a control surface $\Omega$ (in this case, $\Omega_{\text{MID}}$) (Hall and Plumb 1994; Holzer and Hall 2000)). Formally, the boundary propagator $\mathcal{G}(r,t|\Omega_{\text{MID}},t')$ is a Green’s function that solves the continuity equation for the mixing ratio of a conserved and passive tracer, where $r$ is the receptor region, $t'$ is the source time, or the time that tracer last had contact with $\Omega_{\text{MID}}$, and $t$ is the specific field time, or the time when tracer is sampled at $r$. The boundary propagator can also be expressed in terms of the elapsed $t-t'$ transit time $\tau$ (i.e., $\mathcal{G}(r,t|\Omega_{\text{MID}},t') \rightarrow \mathcal{G}(r,t'+\tau|\Omega_{\text{MID}},t)$).

We construct the boundary propagator using four Boundary Impulse Response (BIR tracers), each of which corresponds to a particular instance (in $t'$) of the boundary propagator $\mathcal{G}(r,t|\Omega_{\text{MID}},t')$ (Haine et al. 2008). More precisely, a pulse of a conserved and passive tracer is placed in $\Omega_{\text{MID}}$ at a specific source time $t'$ and the model’s time-evolving response $\mathcal{G}(r,t|\Omega_{\text{MID}},t')$ is the Boundary Impulse Response (BIR). Four BIRs are released at source times $t'=\text{January 1, April 1, July 1, and October 1}$ during the first year of the integration (2000) and integrated for ten years. Unlike in the stratosphere, where 20-30 year integrations are needed to guarantee convergence everywhere (Schoeberl et al. 2005; Li et al. 2012), we find that ten years is more or less sufficient to capture the full evolution of the BIR tracers in the troposphere. In order to ensure that the long term asymptotic behavior is captured, however, we extrapolate the exponential tails for transit times $\tau > 10$ years.

After shifting each BIR response about $\tau = 0$ we treat the average of the centered BIR responses as our approximation to the TTD, designated throughout as $\mathcal{G}(r,\tau|\Omega_{\text{MID}})$. (Note that after averaging we have dropped the explicit dependence on source time $t'$). The resulting quantity is then normalized so that $\int_0^{\infty} d\tau \mathcal{G}(r,\tau|\Omega_{\text{MID}}) = 1$. Physically, the normalization of $\mathcal{G}$ means that the
total air mass fraction at \( \textbf{r} \) had to last have contacted \( \Omega_{\text{MID}} \) sometime in its history (Holzer and Hall (2000)).

The pulse tracer method is direct and easy to implement, compared to the Lagrangian trajectory methods and Eulerian adjoint methods used in Schoeberl et al. (2005) and Haine et al. (2008). However, while the statistics of the TTD and the BIR are identical, the direct product of the pulse tracer method \( G(\textbf{r}, t'+\tau|\Omega, t') \) is not the TTD \( G(\textbf{r}, t|\Omega, t-\tau) \) (Haine et al. 2008). This distinction becomes important when considering seasonal variations in the TTD, which can be deduced using the simplified “BIR map” method outlined in Li et al. (2012). For the purposes of this study, however, we are only interested in approximating the annual mean TTD, in which case \( G(\textbf{r}, \tau|\Omega_{\text{MID}}) \) is adequate. More about the BIR method and distinctions with the TTD can be found in Haine et al. (2008) and Li et al. (2012).

The TTD is summarized throughout in terms of its modal age \( \tau_{\text{mode}}(\textbf{r}|\Omega_{\text{MID}}) \) and its first and second temporal moments, the mean transit time \( \Gamma(\textbf{r}|\Omega_{\text{MID}}) \equiv \int_0^\infty d\tau \tau G(\textbf{r}, \tau|\Omega_{\text{MID}}) \) and the spectral width \( \Delta(\textbf{r}|\Omega_{\text{MID}}) \equiv \sqrt{\frac{1}{2} \int_0^\infty d\tau (\Gamma(\textbf{r}|\Omega_{\text{MID}}) - \tau)^2 G(\textbf{r}, \tau|\Omega_{\text{MID}})} \). Throughout we refer to the mean transit time as the “mean age” for consistency with the presentation in Waugh et al. (2013) and needs to be distinguished from the mean age presented in other studies (Ray et al. 1999; Engel et al. 2006; Patra et al. 2009) that conceptually are similar but are defined with respect to transport from the tropical tropopause or tropical surface, not the NH midlatitudes.

To illustrate the adequacy of the BIR approximation, we compare \( \Gamma(\textbf{r}|\Omega_{\text{MID}}) \) with the mean age derived from an idealized “NH-clock” tracer that is initially set to a value of zero throughout the troposphere. Thereafter it is held to be zero over \( \Omega_{\text{MID}} \) and subject to a constant aging of 1 year per year in the rest of the model surface layer and throughout the atmosphere. The statistically stationary value of the clock tracer is equal to the mean age from the NH midlatitude surface (Waugh et al. 2013) and thus provides an independent check on \( G(\textbf{r}, \tau|\Omega_{\text{MID}}) \). Throughout we designate
the mean age calculated from the clock tracer as $\Gamma(r, t|\Omega_{\text{MID}})$. Note that, unlike $\Gamma(r|\Omega_{\text{MID}})$, the mean age calculated from the clock tracer is explicitly dependent on field time $t$ and thus provides information that otherwise would only be recoverable from the TTD-based calculation using the method outlined in Li et al. (2012).

3. Transit-Time Distribution (TTD)

Figure 1 shows the zonally averaged TTD evaluated near the surface (873 hPa) and the upper troposphere (312 hPa). In the Northern Hemisphere the TTD is skewed to young transit times and characterized by long flat tails. The distribution remains highly peaked poleward of the midlatitude surface and becomes less skewed in the upper midlatitude and subtropical troposphere. South of the equator, the air at the leading edge of the TTD becomes less important relative to air with older transit times. Signatures of fast transport (i.e. $\tau < 50$ days) altogether disappear in the Southern Hemisphere (SH) extratropics as the TTD approaches an inverse Gaussian (IG) distribution.

Different aspects of the TTD are conveniently summarized in terms of the mean age $\Gamma(r|\Omega_{\text{MID}})$ (Figure 2a), the modal age $\tau_{\text{mode}}(r|\Omega_{\text{MID}})$ (Fig. 2b) and the spectral width $\Delta(r|\Omega_{\text{MID}})$ (Fig. 2c). (For convenience, the dependencies on $r$ and $\Omega_{\text{MID}}$ are suppressed throughout, except when they add clarity to the text). Note that $\Gamma$ is overall consistent with the annually averaged mean age calculated from the idealized “NH-clock” tracer (Appendix Figure 1; see also Figure 7 in Waugh et al. (2013)). The mean age $\Gamma$ increases from $\sim 3$ months at the NH middle and high latitude surface to $\sim 1.8$ years at SH high latitudes, while the modal age, $\tau_{\text{mode}}$, remains nearly uniform throughout the NH, ranging from a few days in the extratropics to only $\sim 30$ days at the equator. The spectral width $\Delta$ increases sharply from $\sim 0.5$ over NH high latitudes to $\sim 1.1$ at the equator but varies little in the SH, despite the large gradients in the mean age that line the SH subtropical surface.
It is perhaps surprising that $\Gamma$ and $\Delta$ do not vary consistently with each other. This reflects the fact that the TTD becomes significantly less skewed moving from northern middle and high latitudes to southern extratropical latitudes. This behavior is captured by changes in the shape parameter, $\Delta/\Gamma$, a non-dimensional measure of the relative affects of advective and diffusion on transport from the surface layer. In particular, $\Delta/\Gamma$ decreases from $\sim 2.5$ at NH high latitudes to $\sim 0.7$ in the SH extratropics (Figure 2d). The large gradients in $\Delta/\Gamma$ that span the NH extratropics align roughly with the isentropic boundary between the middleworld and the underworld, where $\Delta/\Gamma < \sim 1.5$ and $\Delta/\Gamma > \sim 1.5$ respectively. (The underworld is the region of the atmosphere where isentropes lie entirely in the troposphere, while the middleworld is the region of the atmosphere where isentropes cross the tropopause (Hoskins 1991)).

Physically, gradients in $\Delta/\Gamma$ reflect changes in the contributions of fast transport pathways relative to slow eddy-diffusive recirculations. That is, poleward of the $\sim 300$ K isentrope where $\Delta/\Gamma > 1.5$, air that re-enters the NH is isentropically transported (and relabeled) back at the midlatitude surface so that the long flat tails carry little mass relative to the leading edge of the TTD. In this region, therefore, the mean and modal ages are weakly related and can vary against each other as they quantify very different aspects of the transport circulation, with $\tau_{\text{mode}}$ and $\Gamma$ respectively reflecting fast and older transport paths from the midlatitude surface. In particular, consider the Arctic where, unlike the mean age, $\tau_{\text{mode}}$ decreases away from the surface, transitioning from $\sim 35$ days in the lower troposphere to $\sim 5$ days just below the tropopause.

In regions where $\Delta/\Gamma < 1.5$ – that is, latitudes equatorward of the $\sim 300$ K isentrope – the mean age varies more-or-less in concert with the modal age. For example, in the tropical and SH subtropical upper troposphere both $\Gamma$ and $\tau_{\text{mode}}$ decrease away from the surface, consistent with the fact that transport to the SH occurs primarily via the upper troposphere (e.g., Plumb and Mahlman 1987; Prather et al. 1987; Holzer 1999; Bowman and Erukhimova 2004). This is also
evident at the SH subtropical surface, where large horizontal gradients in $\Gamma$ reflect changes in $\tau_{\text{mode}}$, which increases sharply from $\sim 30$ days at $10^\circ\text{S}$ to $\sim 300$ days at $20^\circ\text{S}$; by comparison, the spectral width $\Delta$ changes negligibly. As discussed further in Section 4, these changes in $\tau_{\text{mode}}$ reflect the disappearance of fast transport paths south of $10^\circ\text{S}$. Furthermore, the decreases in $\Delta/\Gamma$ at the SH subtropical surface are consistent with estimates of the spectral width and mean age in the SH derived from surface measurements of SF$_6$ and a range of CFCs and CFC replacement compounds (Holzer and Waugh 2015). In particular, observed values of $\Delta \sim 1.3$ vary little with SH latitude, while $\Gamma$ increases from $\sim 1.1$ years in the SH tropics to $\sim 1.4$ years at the South Pole, indicating large changes in the shape parameter $\Delta/\Gamma$ between SH tropics and high latitudes.

4. Transport Pathways

Next we interpret changes in the TTD shape parameter, $\Delta/\Gamma$, in terms of the large-scale circulation by examining the pathways that connect the NH midlatitude surface layer to different regions in the troposphere. We infer transport pathways from the responses of the individual BIR tracers, which are each normalized to unity so that the integral of $\mathcal{G}(\mathbf{r}, t | \Omega_{\text{MID}}, t')$ over transit times in the interval $\tau \in (\tau_1, \tau_2)$ corresponds to the fraction of air at $\mathbf{r}$ that left the midlatitude surface at time $t'$ sometime in the interval $(\tau_1 + d\tau, \tau_2 + d\tau)$. In this sense, the “BIR fraction” $f_{\tau_1}^{\tau_2} (\mathbf{r} | \Omega_{\text{MID}}, t') \equiv \int_{\tau_1}^{\tau_2} d\tau \mathcal{G}(\mathbf{r}, t' + \tau | \Omega_{\text{MID}}, t')$ (hereafter $f_{\tau_1}^{\tau_2}(t')$) provides a gross measure of the transport pathways that trace back to $\Omega_{\text{MID}}$ and how they depend on the time $t'$ when last contact at the midlatitude surface occurred. For convenience, we refer to air that left the midlatitude surface at $t'$ as $t'$ air (i.e. “January air” or air that was last at $\Omega_{\text{MID}}$ on January 1).

It is important to contrast $f_{\tau_1}^{\tau_2}(t')$ with the path-density diagnostic introduced in Holzer (2009), which provides the joint probability of transit times and regions through which the air at $\mathbf{r}$ passed since leaving $\Omega_{\text{MID}}$ and is a more rigorous, but also more computationally intensive, ap-
proach. Nonetheless, by comparing the spatial patterns of $f^\tau_1(t')$ evaluated over successive non-overlapping transit-time intervals we can obtain a gross sense for the paths along which $t'$ air travel until its eventual return back to the NH midlatitude surface.

a. Fast Transport Pathways

The largest spread between the BIR responses $G(r,\tau|\Omega_{\text{MID}},t')$ occurs over small transit times and in regions where large seasonal variations in the circulation project onto the fast transport paths that trace back to the midlatitude surface. (Note that by “fast” we mean occurring on timescales $\tau$ typical of the modal ages in the NH). For the BIR responses examined in this study, these regions coincide with the Arctic and the tropics (Fig. 3, columns 2-4), which we now discuss separately. By comparison, the spread between the BIR responses is relatively smaller over SH midlatitudes (Fig. 3, column 1) and negligible south of $30^\circ$S (not shown).

1) ARCTIC

The fastest transport from the midlatitude surface layer to the Arctic occurs during January, with $G(r,\tau|\Omega_{\text{MID}},\text{January})$ peaking at $\sim 5$-10 days throughout the troposphere at $80^\circ$N (Fig. 3, column 4). By comparison, during summer transport to the Arctic is relatively slower as $G(r,\tau|\Omega_{\text{MID}},\text{July})$ peaks at $\sim 10$ days in the upper troposphere (312 hPa) and $\sim 30$ days near the surface (873 hPa). This summertime delay can be interpreted by comparing the fractions $f^\tau_1(\text{January})$ and $f^\tau_1(\text{July})$ evaluated over successive 10-day transit-time bands spanning $\tau \in [1, 50]$ days (Figure 4, top panel), which show that during boreal winter $\Omega_t$-air moves isentropically from the midlatitude surface to the upper Arctic and then descends to the Arctic surface (Fig. 4, top panels).
This “up-and-over” transport path into the Arctic is consistent with previous Lagrangian studies showing that air parcels at the midlatitude surface rise adiabatically to high latitudes and descend across isentropes that bend over the polar cap and form the so-called “polar dome” (Klonecki et al. 2003; Stohl 2006; Law and Stohl 2007). Seasonal changes in the polar dome have been invoked in previous studies to interpret, for example, the relative contributions of different midlatitude source regions to pollution transport into the Arctic (Stohl 2006).

By comparison, during boreal summer the fractions $f_{t_2}^{t_1}$ (July) reveal that the fastest transport paths connect the midlatitude surface to the midlatitude and subtropical upper troposphere. Transport to the upper Arctic during summer is relatively delayed as isentropic transport to the high latitude upper troposphere weakens relative to diabatic transport associated with convection over midlatitudes that draws surface air from $\Omega_{\text{MID}}$ upward and equatorward to the subtropics. That is, successive patterns of $f_{t_2}^{t_1}$ (July) indicate that during summer air is convectively transported to the upper midlatitude and subtropical troposphere before moving poleward into the upper Arctic (Fig. 4, bottom panels). Similar transport paths have been observed in both tracers of air-mass origin (Orbe et al. 2015) and transport calculations using idealized carbon monoxide (Klonecki et al. 2003).

Interestingly, the lag between the BIR responses in the upper and lower Arctic also increases during boreal summer, as manifest by the large values of $f_{t_2}^{t_1}$ (July) that persist over the pole more than 50 days since last contacting the midlatitude surface (Figure 4, bottom panels). This indicates that residence times in the Arctic increase during boreal summer (Stohl 2006) and are most likely related to enhanced vertical transport away from the lower Arctic as surface temperatures warm and the lower troposphere becomes less stable. We also note that stronger and more persistent horizontal gradients at 60°N – that represent a barrier to horizontal transport back to the midlat-
itude surface – may also play an important role in increasing Arctic residence times by reducing the likelihood that air at the Arctic surface is relabeled at $\Omega_{\text{MID}}$.

2) TROPICS AND NH SUBTROPICS

Next we discuss seasonal variations in the fast transport paths that connect the midlatitude surface to the tropics and SH subtropics (Figure 3, columns 2-3). As discussed in Section 3, these paths extend south to $\sim 10^\circ$S where they represent an important contribution to the TTD, in addition to the older transport paths along which air recirculates between the hemispheres. Here, $\Delta/\Gamma \sim 0.9$ and the mean age $\Gamma$ and the $\tau_{\text{mode}}$ vary together. South of $\sim 10^\circ$S, by comparison, changes in the mean age reflect changes in the spectral width, and $\Delta/\Gamma$ decreases to $\sim 0.7$, reflecting the disappearance of fast transport paths that trace back to the midlatitude surface.

For the BIR responses examined in this study we find that air that is released in January takes the least time to reach the tropical and SH subtropical surface, with $\mathcal{G}(r, \tau|\Omega_{\text{MID}}, \text{January})$ peaking at $\sim 30$ days at $10^\circ$S, compared to $\sim 50$-60 days for the other pulses. The fractions $f_{\tau_1}^{\tau_2}(\text{January})$ reveal that this rapid cross-equatorial transport reflects low-level convergence and ascent over the tropics and SH subtropics (Figure 4, top panels), consistent with the fact that during boreal winter, the zonally averaged intertropical convergence zone (ITCZ) is in the SH and surface inflow represents a fast path across the equator (e.g., Prinn et al. 1992; Holzer 1999; Bowman and Erukhimova 2004).

By comparison, the fractions $f_{\tau_1}^{\tau_2}(\text{July})$ reveal that air released at the midlatitude surface during boreal summer largely avoids the deep tropics and reaches the tropical upper troposphere via the NH subtropics (Figure 4, bottom panels; Figure 6, left panels). This is consistent with the fact that trace gases of midlatitude origin are drawn into the ITCZ in the NH during boreal summer and enter the SH aloft via the upwelling branch of the Hadley cell (Holzer 1999).
Longitudinal sections of $f_{t_1}^{\tau_2}(t')$ at 10°S (Figure 5, top panels) and maps of $f_{t_1}^{\tau_2}(t')$ at 873 hPa, overlaid with the surface precipitation flux due to convection (Figure 6, left panels), more clearly illustrate the relationship between the position of the ITCZ and the rapid near-surface cross-equatorial transport during boreal winter. In particular, most of the air that arrives at 10°S within one month of leaving the midlatitude surface is concentrated over South America and the western Pacific, where the ITCZ is located furthest south and where strong surface winds prevail (Fig. 6, left panels). Over the Indian Ocean, by comparison, surface convection is stronger but located further north, ensuring that air released over $\Omega_{MID}$ is first picked up by convection before it reaches the SH subtropical surface.

While several studies have examined the strong relationship between cross-equatorial transport and the southward extent of tropical convection, invoking shifts in the ITCZ to interpret the seasonal cycles of surface SF$_6$ (Gloor et al. 2007) and mean age (Waugh et al. 2013; Holzer and Waugh 2015), fewer studies have examined the paths underlying cross-equatorial transport in the upper troposphere. The BIR fractions $f_{t_1}^{\tau_2}$(January) evaluated for transit times $\tau \in (21,50)$, show that the air that left the midlatitude surface in January enters the SH over South America and the Indian Ocean (Figure 5, top panels). While the cross-equatorial paths over South America remain more-or-less confined below 200 hPa, higher paths below the tropopause supply surface midlatitude air to the SH over the Indian Ocean, where convective detrainment is both higher and stronger (Figure 7, left panels).

Fast upper tropospheric cross-equatorial transport occurring above 200 hPa increases significantly during boreal summer, with the fastest paths (i.e. $\tau \in [1,10]$ days) passing through a narrow region over the Indian Ocean (Figure 5, bottom panels) and relatively older transport paths (i.e. $\tau < 10$ days) spanning a broader range of longitudes over the East Pacific (Figure 5). Maps of $f_{t_1}^{\tau_2}$(July) at 163 hPa (Figure 7, right panels) reveal that the rapid upper tropospheric pathway over
the Indian Ocean is consistent with strong cross-equatorial winds at the southward edge of the monsoon anticyclone that transport surface air as far south as $30^\circ$S.

While several studies have noted the important role of the Asian monsoon anticyclone in supplying boundary layer constituents to the lower stratosphere (Park et al. (2004); Randel et al. 2010; Park et al. 2013; Bergman et al. 2013), fewer studies have addressed the role of the monsoon in irreversibly transporting surface air and trace species into the SH. How this occurs – either via the meridional advection of convectively detraining air across the equator or through eddy-driven processes such as the shedding of PV along the southward edge of the anticyclone (Popovic and Plumb 2001) – is not well understood and will be explored further in future work.

\textit{b. Older Transport Pathways: Recirculation Back to the Northern Hemisphere}

In Section 3 we showed that there are interesting and subtle relationships between the modal age, the mean age and the spectral width that reflect changes in the shape parameter $\Delta/\Gamma$. It was shown that the large horizontal gradients in $\Delta/\Gamma$ in the NH mark the boundary between the middleworld and underworld, where recirculating air contributes large (small) amounts of mass to the tails of the TTD. Similar signatures of a tropical-subtropical transport barrier in the NH have been noted both in particle dispersion approximations of the tropospheric transport climate (Bowman and Erukhimova 2004) and satellite observations of upper tropospheric carbon monoxide (Bowman 2006).

To obtain a gross sense for the paths through which air recirculates between the hemispheres, we examine the BIR fractions $f^{t_2}_{t_1}(t')$, evaluated for transit times between three months and one year (Figure 8). Air that is released in January, for example, is concentrated in the NH tropical upper troposphere after three months since leaving the surface (Figure 8, top first panel), consistent with low-level convergence at the SH subtropics and ascent within the rising branch of the wintertime
Hadley cell (Figure 4, top panels). The strong horizontal gradients in $f_{t_1}^{t_2}$ (January) in the NH subtropical upper troposphere reflect the interface between upper level convective outflow and strong north-south gradients of potential vorticity at the subtropical jet stream that form a barrier to meridional transport during late winter and early spring (here, $t = March$) (Mahlman 1997). Correspondingly, $f_{t_1}^{t_2}(t')$ extends down to the NH subtropical surface as the descending branch of the Hadley Cell represents the more favorable transport path and facilitates re-entry back into the SH. Similar signatures of this path back into the SH through the Hadley Cell appear in the 7-9 month-old fraction $f_{t_1}^{t_2}$ (July) (Figure 8, bottom third panel).

During boreal summer, by comparison, the mean circulation in the tropical upper troposphere opposes transport back into the NH, which is reflected in the weak vertical gradients in $f_{t_1}^{t_2}(t')$ over the NH subtropics during the summer months (Figure 8 top third panel, bottom first panel). Nonetheless, lower stratospheric transport paths may enable re-entry back into the NH during summer, as indicated by the weak horizontal gradients in $f_{t_1}^{t_2}(t')$ that line the NH lower stratosphere. Weaker horizontal gradients during boreal summer are consistent with a summertime relaxation in the lower stratospheric tropical-extratropical mixing barrier (Chen 1995; Konopka et al. 2009), which enables air that left the midlatitude surface to re-enter the NH troposphere above the subtropical jet (Figure 8 top third panel, bottom fourth panel).

It is more difficult to infer physically meaningful pathways for $\tau > 1$ year because older air reflects more circuitous and less clearly defined paths, consistent with the diffusive nature of transport. Nonetheless, successive integrals of $G(r, \tau | \Omega_{MID})$ evaluated over older transit-time intervals (not shown) reveal that a large amount of air that leaves the midlatitude surface passes through the stratosphere before re-entering the troposphere. Although $G(r, \tau | \Omega_{MID})$ does not explicitly distinguish between tropospheric and stratospheric paths, this apparent influence of stratospheric paths on the troposphere is qualitatively consistent with Holzer (2009), who show that air at the
SH surface has nearly a 20% probability of being found in the stratosphere since leaving the NH high latitude surface.

To obtain a more quantitative sense of possible signatures of stratospheric paths on tropospheric air we fit the tails of the TTD with an exponentially decaying mode $\Psi_0(r,t)e^{-\tau/\tau_0}$, where $\tau_0$ is the eigentime of the lowest mode of the TTD and describes how fast transport alone can cause the eventual decay of the mixing ratio of a conserved tracer (Ehhalt et al. 2004). For the GMI-MERRA integration we find that $\tau_0 \sim 3$ years throughout the troposphere, where fits have been performed over transit times greater than four years. This decay rate is similar in magnitude, albeit smaller, to the decay mode of stratospheric age spectra calculated using the free-running NASA chemistry climate model GEOSCCM (Li et al. 2012), which suggests that the long flat tails of the TTD reflect air of stratospheric origin. The fact that $\tau_0$ is slightly older in the CTM, however, may indicate excessive mixing, as analyses of stratospheric age spectra calculated using other reanalysis products (ERA-Interim) have suggested (Diallo et al. 2012). Further studies, however, are needed to quantitatively separate out the stratospheric and tropospheric contributions to the tails of the TTD.

5. Idealized Decay Tracers

The interpretation of the TTD as a propagator of boundary conditions is reinforced by expressing the interior mixing ratio $\chi$ of a passive tracer at every interior point $(r,t)$ by the following integration:

$$\chi(r,t) = \int_{-\infty}^{t} dt' \mathcal{G}(r,t|\Omega,t')\chi(\Omega,t')e^{-(t-t')/\tau_c}. \quad (1)$$
where the mixing ratio over the source region, $\chi(\Omega, t')$, has been assumed to be uniform over $\Omega$ (here $\Omega_{\text{MID}}$) and where the decay term $e^{-(t-t')/\tau_c}$ represents spatially uniform first-order loss operating at a constant timescale $\tau_c$. Equation (1) underscores the fact that the distribution and evolution of species subject to chemical decay (or other loss mechanisms) are determined by the interaction between $\tau_c$ and the underlying TTD.

Equation (1) provides a way to interpret patterns of tracers that undergo loss in terms of the underlying transport, as Waugh et al. (2003) explored for species undergoing both loss and time variations in their surface boundary conditions using the related, but distinct, concept of concentration “tracer ages.” Conversely, it also suggests that tracers experiencing loss can be used to constrain information about the TTD. Both points are examined now for the 5-day and 50-day tracers.

a. Annual Mean

The annual mean 5-day and 50-day tracer concentrations $\chi_{5y}^\ast(r|\Omega_{\text{MID}})$ and $\chi_{50}^\ast(r|\Omega_{\text{MID}})$, normalized by their average concentrations over the NH midlatitude source region, $\chi_{\Omega_{\text{MID}}}$, are shown in Figure 9 (top panels). This normalization ensures that spatial patterns and amplitudes of the 5-day and 50-day tracers, expressed throughout as percentages, can be meaningfully compared. Throughout we only consider normalized concentrations and, for convenience, we refer to $\chi_{\tau_c}^\ast(r|\Omega_{\text{MID}})/\chi_{\Omega_{\text{MID}}}$ for $\tau_c = 5$ days and $\tau_c = 50$ days simply as $\chi_5^\ast$ and $\chi_{50}^\ast$, respectively. (Note that the overbar denotes the annual mean time average, while the asterisk refers to the normalization by $\chi_{\Omega_{\text{MID}}}$).

Reconstructions of $\chi_5^\ast$ and $\chi_{50}^\ast$ using the convolution in Equation (1) compare well with the explicitly integrated tracer concentrations (Fig. 9, bottom panels). Here the reconstructed concentrations have been calculated as $\chi_{\tau_c}^\ast(r)/\chi_{\Omega_{\text{MID}}}$ $\equiv \int_0^\infty d\tau \mathcal{G}(r, \tau|\Omega_{\text{MID}}) e^{-\tau/\tau_c}$, where the decay
timescale $\tau_c$ is assumed to be spatially uniform and constant in time. The close correspondence
between reconstructed and explicitly integrated tracers indicates that $\mathcal{G}(r, \tau|\Omega_{\text{MID}})$ not only cap-
tures the mean age, but also the leading edge and middle portion of the TTD, to which the 5-day
and 50-day tracers are more sensitive.

The 5-day tracer concentration, $\mathcal{X}_5^*$, is mainly confined to the NH midlatitude lower and middle
troposphere and is flanked by strong horizontal gradients poleward and equatorward of $\Omega_{\text{MID}}$. By
comparison, $\mathcal{X}_{50}^*$ extends into the tropics and SH subtropics and exhibits weak vertical gradients
over the NH high latitude troposphere. Rather, strong vertical gradients in $\mathcal{X}_{50}^*$ span the tropopause
and the NH subtropics, where they align with isentropes that slope down from the midlatitude
tropopause to the surface.

Comparing the spatial patterns of $\mathcal{X}_5^*$ and $\mathcal{X}_{50}^*$ with the TTD timescales (Figure 2) we find sim-
ilarities with the modal age $\tau_{\text{mode}}$ and the mean age $\Gamma$, respectively. These relationships are con-
ditional, however, on the underlying transport characteristics, as captured by the shape parameter
$\Delta/\Gamma$, and change in response to subtle changes in the balance between fast transport paths and
slow eddy-diffusive recirculations. For example, $\mathcal{X}_5^*$ and $\tau_{\text{mode}}$ correspond well over NH middle
and high latitudes where $\Delta/\Gamma > 2$ – that is, in regions where the TTD is highly peaked and most
of the air mass arrived after transit times near the modal age. Thus, large vertical gradients in $\mathcal{X}_5^*$
in the Arctic (Figure 9a) reflect strong vertical gradients in the modal age $\tau_{\text{mode}}$ (Figure 2b).

The 5-day tracer and modal age, however, agree less well in regions where $\Delta/\Gamma$ is smaller (i.e.
the NH middleworld and SH), reflecting the fact that the total mass of tracer is more broadly
distributed over transit times older than the modal age. For example, the large surface gradients
in $\mathcal{X}_5^*$ at $30^\circ$N do not reflect changes in the modal age, but rather changes in the spectral width $\Delta$
(Fig. 2c), as eddy-diffusive recirculations contribute more mass relative to fast transport paths (see
discussion in Section 3 and Figure 1). That is, species with short lifetimes (i.e. $\tau_c = 5$ days) do not
survive transit on the old recirculating paths that manifest in the middle world equatorward of the 300 K surface.

In a similar sense, the correspondence between $\mathcal{X}_{50}^*$ and the mean age $\Gamma$ is conditional on the shape of the underlying TTD. The agreement is best in regions where $\Delta/\Gamma < 1.5$, as the TTD is less peaked and more of the air (and tracer) mass is distributed over older transit times. This is illustrated by the strong vertical gradients in both $\mathcal{X}_{50}^*$ and $\Gamma$ that line the NH subtropical surface.

By comparison, the vertical gradients in $\mathcal{X}_{50}^*$ over the Arctic reflect vertical gradients in the modal age since recirculating transport paths back in the Arctic – that skew $\Gamma$ to $\sim 0.3$ years – contribute little mass to the 50-day tracer.

The 5-day and 50-day tracers only represent two examples of how $\tau_c$ interacts with the underlying TTD. We explore this more systematically by reconstructing the concentrations $\mathcal{X}_{\tau_c}^* (r|\Omega_{MID})$ for species subject to loss between $\tau_c = 5$ days and $\tau_c = 1$ year using Equation (1) and our approximation to the TTD, $\mathcal{G}(r, \tau|\Omega_{MID})$. First we examine latitudinal profiles of $\mathcal{X}_{\tau_c}^*$ (Figure 10a) at 712 hPa, which feature horizontal gradients poleward of the midlatitude source region for the range of species considered. However, the gradients in $\mathcal{X}_{\tau_c}^*$ are significantly stronger for loss rates $\tau_c \leq 1$ month and nearly negligible for species with chemical lifetimes of $\tau_c > 3$ months).

The sensitivity of gradients in $\mathcal{X}_{\tau_c}^*$ at NH high latitudes to the underlying loss $\tau_c$ reflects the fact that subtle increases in the modal age moving poleward from midlatitudes (Figure 1a) result in large changes in the integrated mass at the leading edge of the TTD and, correspondingly large changes in the concentrations of species that undergo rapid loss. The concentrations of longer-lived species, by comparison, are more sensitive to older transport paths and, therefore, only weakly responsive to modal age changes at high latitudes. This suggests that subtle variations in the modal age $\tau_{\text{mode}}$ (Figure 2b) related to isentropic transport from the midlatitude surface (Figure 4, top rows) are only captured by species with lifetimes $\tau_c \leq 1$ month.
Finally, the spread between the individual profiles of \( \overline{X}^+_\tau_c \) also reveals information about the shape parameter \( \Delta/\Gamma \). For example, the spread between the concentrations of species with short lifetimes (i.e. \( \tau_c < 50 \) days) increases moving poleward out of the NH subtropics coincident with a doubling of the shape parameter from 1.1 at 20°N to 2.1 at 50°N (Figure 2d). Conversely, the spread between concentrations with species with longer lifetimes decreases over the Arctic as older transport paths contribute weakly to the total integrated tracer mass. Similar changes in the spread between tracers of different lifetimes are seen in the vertical profiles of \( \overline{X}^+_\tau_c \) (Figure 10b) and reflect the strong vertical gradients in \( \Delta/\Gamma \) that occur at 40°N. In particular, the largest spread between profiles of \( \overline{X}^+_\tau_c \) at the surface occurs between species with short lifetimes, consistent with the fact that the TTD is skewed to young transit times and characterized by long flat tails (i.e. \( \Delta/\Gamma > 1.5 \)). By comparison, in the upper troposphere where the total tracer mass is increasingly dominated by older transport paths, the largest differences between \( \overline{X}^+_\tau_c \) manifest between species with loss timescales \( \tau_c \) greater than a few months.

**b. Seasonal Cycle**

The close correspondence between the 5-day and 50-day tracers and the modal age \( \tau_{mode}(r|\Omega_{MID},t') \) and the mean age \( \Gamma(r,t|\Omega_{MID}) \) provides an opportunity for interpreting \( \chi^+_5(t) \) and \( \chi^+_{50}(t) \) in terms of the large-scale circulation. This is explored by comparing the seasonal cycles of the mean and modal ages with the seasonal cycles of the 5-day and 50-day tracers, \( \chi^+_5(r,t|\Omega_{MID}) \) and \( \chi^+_{50}(r,t|\Omega_{MID}) \). (For convenience, we refer to \( \chi^+_5(r,t|\Omega_{MID}) \) for \( \tau_c = 5 \) days and \( \tau_c = 50 \) days as \( \chi^+_5(t) \) and \( \chi^+_{50}(t) \), respectively, where the overbar in the previous notations has been replaced with explicit dependence on field time \( t \). As before, the asterisk denotes normalization by \( \chi_{\Omega_{MID}} \).)
The seasonal cycle of the 5-day tracer is large in the Arctic, where \( \chi^*_5(t) \) triples from 3% at 600 hPa in July to 9% in January (Figure 11a). By comparison, gradients in \( \chi^*_5(t) \) are relatively weaker and the amplitude of the seasonal cycle at high latitudes is smaller (Figure 11b). The opposite behavior holds in the NH subtropical upper troposphere, where circulation changes project more strongly onto gradients in \( \chi^*_5(t) \), partly reflecting the fact that there is simply more 50-day tracer that survives transit to that region. The seasonal cycle of \( \chi^*_5(t) \) is also large near the tropical surface, where latitudinal shifts in the ITCZ project onto large horizontal tracer gradients. Similar shifts in tropical convection have been used to interpret the seasonality of mean age at the tropical surface (Waugh et al. 2013).

Direct comparisons of the TTD timescales and the decay tracers are shown in Figure 12. (Note that \( \tau_{\text{mode}}(r|\Omega,t') \) has been interpolated linearly in \( t' \) to a monthly interval.) The anomalously low concentrations of \( \chi^*_5(t) \) in the Arctic during boreal summer reflect the fact that direct isentropic transport from the midlatitude surface is delayed compared to during winter and is manifest as the large values of \( \tau_{\text{mode}}(r|\Omega_{\text{MID}},t') \) for \( t' = \) July (Fig. 12, left column, bottom panel). In addition, the seasonal cycles of the modal age \( \tau_{\text{mode}}(r|\Omega_{\text{MID}},t') \) and \( \chi^*_5(t) \) both weaken in the upper Arctic (Fig. 12, left columns top panel) compared to the seasonal cycle in the NH subtropics, which is much stronger. The late summer/early fall increase in \( \chi^*_5(t) \) in the NH subtropical upper troposphere (Fig. 12, left column, top panel) coincides with a decrease in the modal age (Fig. 12, second column, top panel) as air is rapidly transported from the midlatitude surface into the upper troposphere during boreal summer. Recall from Section 4 that fast transport to the NH subtropical upper troposphere during summer reflects pathways associated with the Asian monsoon.

The seasonal cycle of the 50-day tracer, \( \chi^*_{50}(t) \), by comparison, corresponds closer to the mean age \( \Gamma(r,t|\Omega_{\text{MID}}) \), where the “NH-clock” tracer-based age is used in place of \( \Gamma(r|\Omega_{\text{MID}}) \), which does not depend on \( t \) (see Section 2.2). Unlike for \( \chi^*_5(t) \), the 50-day tracer and the mean age ex-
hibit a weak seasonal cycle in the Arctic middle and lower troposphere since they do not respond strongly to the seasonally varying fast transport paths that trace isentropically back to the midlatitude surface. In the upper Arctic, by comparison, the seasonal cycle of both the 50-day tracer and the mean age is much stronger and characterized by older mean ages – and low concentrations of \( \chi_{50}^*(t) \) – during January. Here the seasonal cycle \( \chi_{50}^*(t) \) is opposite to \( \chi_{5}^*(t) \), and coincides with a lower tropopause during boreal winter that drags small (large) values of \( \chi_{50}^*(t) \) (\( \Gamma(t) \)), leading to anomalously less tracer (older ages) (Figure 12, right columns, top panel).

It is important to note that the seasonal cycle of \( \tau_{\text{mode}}(r|\Omega_{\text{MID}},t') \) is not necessarily – and most often not – the same as \( \tau_{\text{mode}}(r,t|\Omega_{\text{MID}}) \). Hence, we have only used \( \tau_{\text{mode}}(r|\Omega_{\text{MID}},t') \) to interpret transport in the NH middle and high latitudes, where the modal age is both young and the TTD is highly skewed to young transit times. This implies that the air in the Arctic most likely had recent contact at the surface rendering \( t' \sim t \). As this approximation is crude, however, we have taken care so that \( \tau_{\text{mode}}(r|\Omega_{\text{MID}},t') \) only qualitatively guides our interpretation of the idealized tracers.

6. Conclusions

An analysis of the transit-time distribution (TTD) since last contact at the NH midlatitude surface, calculated using the NASA GMI Chemistry Transport Model driven with MERRA reanalysis meteorological field, reveals that:

- Throughout the troposphere the TTD is characterized by long flat tails, resulting in mean ages (\( \Gamma \)) that are much larger than their corresponding modal ages (\( \tau_{\text{mode}} \)). Thus, while \( \tau_{\text{mode}} \) is nearly uniform in the NH, the mean age \( \Gamma \) increases from \( \sim 0.25 \) years at the high latitude surface to \( \sim 1.25 \) years in the northern subtropics.
The TTD transitions from a highly skewed distribution at NH middle and high latitudes, where $\Delta/\Gamma > 1.5$, to an inverse Gaussian in the SH extratropics, where $\Delta/\Gamma \sim 0.7$. Physically, gradients in the shape parameter $\Delta/\Gamma$ signal changes in the importance of fast transport pathways relative to slow eddy-diffusive recirculations, with large gradients at the SH subtropical and tropical surface signaling the disappearance of fast cross-equatorial paths south of 10°S. By comparison, strong gradients in $\Delta/\Gamma$ that line the isentrope bounding the NH middleworld and underworld reflect changes in the amount of air that recirculates back into the NH.

The spatial patterns and seasonal variations of idealized 5-day and 50-day tracers reflect the patterns and seasonal cycles of $\tau_{\text{mode}}$ and $\Gamma$, respectively. In the Arctic seasonal variations in the 5-day tracer and modal age reflect changes in isentropic transport away from the midlatitude surface and result in delayed transport to the Arctic surface during boreal summer compared to winter. In the tropics seasonal variations reflect shifts in the position of tropical convection and changes in upper tropospheric cross-equatorial flow associated with the Asian monsoon, which represents an important source of fast cross-equatorial transport deep into the SH.

The spread between tracers that undergo atmospheric loss provides information about the shape parameter $\Delta/\Gamma$. In the NH underworld, where $\Delta/\Gamma > 1.5$, most of the tracer variance due to the interaction of chemical loss and the underlying TTD is captured by species with lifetimes $\tau_c < 1$ month. By comparison, in the NH middleworld $\Delta/\Gamma$ decreases and the optimal range of $\tau_c$ is significantly broader.

One implication of our results is that meaningful diagnostics for comparing and assessing transport in numerical models must consider the interaction between loss and the underlying TTD. This point was made in Waugh et al. (2003) and now illustrated for the specific case of transport from the NH midlatitude surface and for idealized spatially uniform first-order loss with constant
lifetime $\tau_c$. Here we find interesting and subtle relationships in the interaction between loss and transport that reflect changes in the relative contributions of fast versus recirculating transport paths. In particular, we show that in the Arctic, where $\Delta/\Gamma > 1.5$, there are large differences in the mean gradients and seasonal cycles between species subject to loss rates of 5 days$^{-1}$ and 50 days$^{-1}$. Thus, while idealized tracers with prescribed lifetimes of 50 days and idealized variants of carbon monoxide have been used to assess black carbon transport to the Arctic (Shindell et al. 2008; Lee et al. 2013), idealized tracers with loss on the order of the modal age $\tau_{\text{mode}}$ (i.e. $\sim 5$ days) may be more appropriate at constraining the transport experienced by shorter-lived species.

Our results also show that tracers undergoing chemical loss can not only be used to test model transport, but may also be used to constrain the TTD from observations. As such, our study complements Holzer and Waugh (2015) who use a maximum-entropy based inversion technique to constrain the mean age and the spectral width from surface measurements of SF$_6$, CFCs and various CFC replacement compounds. Their analysis, however, was restricted to the SH and our model results provides some guidelines for repeating their calculations in the tropics and the NH.

In particular, our results indicate that the ability to constrain the TTD in the NH depends strongly on the tracers used and the underlying transport characteristics (Waugh et al. 2003). For the flows typical of NH middle and high latitudes (i.e. $\Delta/\Gamma > 1.5$), measurements of a range of species with chemical lifetimes less than one month are needed to best constrain the TTD, and may include, for example, the non-methane hydrocarbons butane, benzene and propane which have approximate lifetimes of 5, 8 and 10 days respectively. For flows typical of the NH tropics and subtropics (i.e. $\Delta/\Gamma < 1.5$), however, the optimal lifetime window is significantly broader and measurements of longer-lived species are also needed. These may include, for example, the broad range of CFCs and CFC replacement compounds available from surface networks (e.g., HATS, CCGG and AGAGE) and recent aircraft campaigns (e.g., HIPPO, (Wofsy 2011)).
With these guidelines in mind, and with knowledge of the mean age from observations of SF$_6$, it may be then be feasible to recover estimates of the spectral width in the NH. Such constraints would be especially useful in the NH where the TTD is not inverse Gaussian, which limits the use of traditional parametric inversion methods that have been used to infer the stratospheric age spectrum from observational data (Schoeberl et al. 2005). Our focus on uniformly emitted tracers subject to uniform loss, however, may limit the ability to extract TTD timescales from more realistic tracers undergo spatially and temporally varying loss and/or changes in their emissions and will first need to be examined systematically within a modeling framework.

One important caveat in our study is that our results represent transport for one model and our analysis will need to be repeated for other chemistry transport models and chemistry climate models. One opportunity for comparing our results includes analysis of the “NH-clock” tracer and the 5-day and 50-day tracers that were requested as model output in the most recent Chemistry-Climate Model Initiative (CCMI) and which, to the best of our knowledge, have not yet been extensively compared between participating models.

In particular, the fast cross-equatorial transport pathways inferred from the BIR pulse tracers needs to be examined more thoroughly. Connections between trace species’ variability in the SH subtropics and the El Nino-Oscillation (ENSO) have been well documented (Elkins et al. 1993; Prinn et al. 1992; Lintner et al. 2004) and possible sensitivities to ENSO phase will therefore need to examined. While preliminary investigations with GEOSCCM, forced with annually repeating ENSO-neutral sea surface temperatures and sea ice concentrations, indicate that the observed cross-equatorial transport over the Pacific and Indian ocean are qualitatively robust features of the transport, more work is needed to address this systematically.
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Throughout the TTD is approximated using $G(r, \tau|\Omega_{\text{MID}})$, the average of four Boundary Impulse Response (BIR) response tracers released at source times $t' = \text{January 1, April 1, July 1, and October 1}$ during the first year of the integration. Appendix Figure 1 (A1) shows how well this approximation recovers the mean transit time (or mean age) since last contact at the NH midlatitude surface, by comparing the first temporal moment $\Gamma(r|\Omega_{\text{MID}}) \equiv \int_0^\infty d\tau \tau G(r, \tau|\Omega_{\text{MID}})$ with the mean age calculated as the annual average of the “NH-clock” tracer evaluated during the last year of the integration.
Figure 1 The distribution of transit times (TTD) since last contact at the NH midlatitude surface layer, \( G(\mathbf{r}, \tau | \Omega_{\text{MID}}) \), approximated as the average of four one-day-long pulses released at \( \Omega_{\text{MID}} \) at source times \( t' = \) January 1, April 1, July 1 and October 1 during the first year of the integration. The red dashed and solid lines mark the modal age, \( \tau_{\text{mode}} \), and mean age \( \Gamma \), respectively, where the mean age is calculated as the first temporal moment of the TTD. The ratio of the spectral width and the mean age, \( \Delta/\Gamma \), ranges from \( \sim 0.7 \) in the SH, where the TTD is approximately inverse Gaussian, to \( \sim 2 \) over NH midlatitudes, where the distribution is skewed to young transit times and characterized by long flat tails.
Figure 2 The mean age, $\Gamma(r|\Omega_{MID})$ (a), modal age $\tau_{mode}(r|\Omega_{MID})$ (b) and spectral width $\Delta(r|\Omega_{MID})$ (c), calculated from the approximation to the TTD, $\mathcal{G}(r, \tau|\Omega_{MID})$. The shape parameter, $\Delta/\Gamma$, is shown in (d). The thick black line and thin white contours denote the annually averaged climatological mean thermal tropopause and isentropes, respectively (the 300 K, 330 K, and 360 K isentropes are shown). Note the nonlinear colorbar in (b).
Figure 3 The Boundary Impulse Responses (BIRs), $G(r,t|\Omega_{\text{MID}},t')$, released over $\Omega_{\text{MID}}$ at sources times $t' = \text{January 1 (blue), April 1 (cyan), July 1 (red), and October 1 (green)}$, plotted with respect to the elapsed transit time $\tau \equiv t - t'$. The responses are evaluated in the upper troposphere (312 hPa; top) and the lower troposphere (873 hPa; bottom). The black dashed line shows the average of the BIRs, $\bar{G}(r,\tau|\Omega_{\text{MID}})$. 

$G(r,t|\Omega_{\text{MID}},t')$ evaluated at 312 hPa (top) and 873 hPa (bottom).
Figure 4: The zonal mean BIR fraction for = January (top) and = July (bottom).

Here is the fraction of air that last contacted within the time interval and is calculated asПроизведение произведений. Transit-time bands span 1-10 days (column 1), 11-20 days (column 2), 21-30 days (column 3), 31-40 days (column 4), and 41-50 days (column 5). Note that the horizontal axis extends from 40 S to 80 N.

\[
\text{Zonal Mean (top)} = \frac{1}{40-50} \text{ days (column 1)} \quad \text{and} \quad \frac{1}{41-50} \text{ days (column 2)}, \quad \text{for (r|W,MID|t) = 0} \quad \frac{1}{40-50} \text{ days (column 3)} \quad \frac{1}{41-50} \text{ days (column 4)} \quad \frac{1}{41-50} \text{ days (column 5)}.
\]
\[ f^{\tau_2}_{\tau_1}(r|\Omega_{\text{MID}}, t') \] for \( t' = \text{January} \) (top) and \( t' = \text{July} \) (bottom)

Cross-section at 10°S (%)

\( \tau \in [1,10] \) days  \( \tau \in [11,20] \) days  \( \tau \in [21,30] \) days  \( \tau \in [31,40] \) days  \( \tau \in [41,50] \) days

**Figure 5** Same as Figure 4, except the cross-section at 10°S is shown.
Figure 6. Maps of the BIR fraction $f_2$ for source times $t_0 = January$ (left) and $t_0 = July$ (right). The reanalysis surface precipitation flux from convection is overlaid in the black contours and has been averaged over field times corresponding to each panel. The field time averaged winds at 873 hPa are also shown (white arrows). The transit times bands are identical to those shown in Figures 4 and 5 and span 1-10 days, 11-20 days, 21-30 days, and 31-40 days, moving from top to bottom. Only the last interval (i.e., $t \in [41, 50]$ days) is not shown.

\[ \text{873 hPa} \]
\[ f_{T_2}^r (r | \Omega_{MID}, t') \text{ for } t' = \text{January (left) and } t' = \text{July (right)} \% \]

Figure 7 Same as Figure 6, except now evaluated at 163 hPa. The 163 hPa field time averaged reanalysis winds are shown in the white arrows.
Transit-time bands span 3 months, 4-6 months, 7-9 months, and 10-12 months since air was released over WMI. The corresponding field times \( t_1 \) and \( t_2 \) (top) and \( t_0 \) and \( t_1 \) (bottom) are labeled above each panel. The thick black line denote the field times. The corresponding field times \( t_1 \) and \( t_2 \) (top) and \( t_0 \) and \( t_1 \) (bottom) are labeled above each panel. The thick black line denote the field times. The thick black line and thin white contours denote the field times.
Figure 9 The annually averaged concentrations $\overline{\chi}^{\ast}(r|\Omega_{\text{MID}})$ of the 5-day ($\tau_c = 5$ days) (a, top panel) and 50-day ($\tau_c = 50$ days) (b, top panel) idealized decay tracers, averaged over the last year of the integration and normalized by the $\Omega_{\text{MID}}$-averaged surface concentration for each tracer, $\chi_{\Omega_{\text{MID}}}$. The bottom panels show reconstructions of $\overline{\chi}^{\ast}(r)/\chi_{\Omega_{\text{MID}}}$ using Equation 1 and the approximation to the TTD, $\mathcal{G}(r, \tau|\Omega_{\text{MID}})$. The thick black line and thin white contours denote the annually averaged climatological mean thermal tropopause and isentropes, respectively (the 300 K, 330 K, and 360 K isentropes are shown).
The normalized concentrations have been calculated using the convolution (Equation 1) and the approximation to the TTD, equation (Equation 1). The normalized concentrations evaluated at 712 hPa (red) and 50 days (blue) for $t_c = 0.2$, 1 month (black, dashed), and 1 year (black, circles) and 1 year (black, stars). The normalized concentrations have been calculated using the convolution (Equation 1) and the approximation to the TTD, equation (Equation 1).
Figure 11 The seasonal cycle of $\chi_5^*(r,t|\Omega_{MID})$ (a) and $\chi_{50}^*(r,t|\Omega_{MID})$ (b), evaluated during the last year of the integration after a statistically stationary state has been reached. The thick black line and thin white contours denote the monthly-mean thermal tropopause and isentropes, respectively, averaged over the last year of the integration (the 300 K, 330 K, and 360 K isentropes are shown). Note that the horizontal axis spans 20°S to 90°N.
Figure 12 The seasonal cycles $c(t)$ for $t_c = 5$ days (column 1) and $t_c = 50$ days (column 3). The seasonal cycles of the modal age $t_{mode}(r, t | W_{MID})$ (column 2) and the mean age calculated from the "NH-clock" tracer $T_{av}(r, t | W_{MID})$ (column 4) are shown with respect to source time $t_0$ and field time $t$, respectively. All fields are evaluated at 312 hPa (top), 506 hPa (middle) and 873 hPa (bottom).
LIST OF FIGURES

Fig. 1. Figure 1 The distribution of transit times (TTD) since last contact at the NH midlatitude surface layer, \( \mathcal{S}(r, \tau | \Omega_{\text{MID}}) \), approximated as the average of four one-day-long pulses released at \( \Omega_{\text{MID}} \) at source times \( t' = \) January 1, April 1, July 1 and October 1 during the first year of the integration. The red dashed and solid lines mark the modal age, \( \tau_{\text{mode}} \), and mean age \( \Gamma \), respectively, where the mean age is calculated as the first temporal moment of the TTD. The ratio of the spectral width and the mean age, \( \Delta/\Gamma \), ranges from \( \sim 0.7 \) in the SH, where the TTD is approximately inverse Gaussian, to \( \sim 2 \) over NH midlatitudes, where the distribution is skewed to young transit times and characterized by long flat tails.

Fig. 2. Figure 2 The mean age, \( \Gamma(r|\Omega_{\text{MID}}) \) (a), modal age \( \tau_{\text{mode}}(r|\Omega_{\text{MID}}) \) (b) and spectral width \( \Delta(r|\Omega_{\text{MID}}) \) (c), calculated from the approximation to the TTD, \( \mathcal{S}(r, \tau | \Omega_{\text{MID}}) \). The shape parameter, \( \Delta/\Gamma \), is shown in (d). The thick black line and thin white contours denote the annually averaged climatological mean thermal tropopause and isentropes, respectively (the 300 K, 330 K, and 360 K isentropes are shown). Note the nonlinear colorbar in (b).

Fig. 3. Figure 3 The Boundary Impulse Responses (BIRs), \( \mathcal{S}(r, \tau|\Omega_{\text{MID}}, t') \), released over \( \Omega_{\text{MID}} \) at sources times \( t' = \) January 1 (blue), April 1 (cyan), July 1 (red), and October 1 (green), plotted with respect to the elapsed transit time \( \tau = t - t' \). The responses are evaluated in the upper troposphere (312 hPa; top) and the lower troposphere (873 hPa; bottom). The black dashed line shows the average of the BIRs, \( \mathcal{S}(r, \tau|\Omega_{\text{MID}}) \).

Fig. 4. Figure 4 The zonal mean BIR fraction \( f_{\Omega_{\text{MID}}}^\tau (r|\Omega_{\text{MID}}, t') \) corresponding to source times \( t' = \) January 1 (top) and \( t' = \) July 1 (bottom). Here \( f_{\Omega_{\text{MID}}}^\tau (r|\Omega_{\text{MID}}, t') \) is the fraction of air at \( r \) that last contacted \( \Omega_{\text{MID}} \) within the time interval \( (t' + \tau_1, t' + \tau_2) \) and is calculated as \( f_{\Omega_{\text{MID}}}^\tau (r|\Omega_{\text{MID}}, t') \equiv \int_{\tau_1}^{\tau_2} d\tau \mathcal{S}(r, \tau|\Omega_{\text{MID}}, t') \). Transit-time bands span 1-10 days (column 1), 11-20 days (column 2), 21-30 days (column 3), 31-40 days (column 4), and 41-50 days (column 5). Note that the horizontal axis extends from 40°S to 80°N.

Fig. 5. Figure 5 Same as Figure 4, except the cross-section at 10°S is shown.

Fig. 6. Figure 6 Maps of the BIR fraction \( f_{\Omega_{\text{MID}}}^\tau (r|\Omega_{\text{MID}}, t') \) at 873 hPa for source times \( t' = \) January (left) and \( t' = \) July (right). The reanalysis surface precipitation flux from convection is overlaid in the black contours and has been averaged over field times corresponding to each panel, \( t \in (t' + \tau_1, t' + \tau_2) \). The field time averaged winds at 873 hPa are also shown (white arrows). The transit times bands \( (\tau_1, \tau_2) \) are identical to those shown in Figures 4 and 5 and span 1-10 days, 11-20 days, 21-30 days, and 31-40 days, moving from top to bottom. Only the last interval (i.e. \( \tau \in (41 - 50) \)) is not shown.

Fig. 7. Figure 7 Same as Figure 6, except now evaluated at 163 hPa. The 163 hPa field time averaged reanalysis winds are shown in the white arrows.

Fig. 8. Figure 8 The zonal mean BIR fraction \( f_{\Omega_{\text{MID}}}^\tau (r|\Omega_{\text{MID}}, t') \) corresponding to source times \( t' = \) January 1 (top) and \( t' = \) July 1 (bottom). Transit-time bands span 3 months, 4-6 months, 7-9 months, and 10-12 months since air was released over \( \Omega_{\text{MID}} \). The corresponding field times \( (t \in (t' + \tau_1, t' + \tau_2)) \) are labeled above each panel. The thick black line and thin white contours denote the field time averaged thermal tropopause and isentropes, respectively (the 300 K, 330 K, and 360 K isentropes are shown).

Fig. 9. Figure 9 The annually averaged concentrations \( \overline{X}_s^\tau (r|\Omega_{\text{MID}}) \) of the 5-day (\( \tau_c = 5 \) days) (a, top panel) and 50-day (\( \tau_c = 50 \) days) (b, top panel) idealized decay tracers, averaged over the last year of the integration and normalized by the \( \Omega_{\text{MID}} \)-averaged surface concentration for.
each tracer, $\chi_{\Omega_{\text{MID}}}$. The bottom panels show reconstructions of $\chi_{\Omega_{\text{MID}}}^\tau (r) / \chi_{\Omega_{\text{MID}}}$ using Equation 1 and the approximation to the TTD, $\mathcal{G}(r, \tau_{\Omega_{\text{MID}}})$. The thick black line and thin white contours denote the annually averaged climatological mean thermal tropopause and isentropes, respectively (the 300 K, 330 K, and 360 K isentropes are shown).

Fig. 10. Figure 10 The normalized concentrations $\chi_{\Omega_{\text{MID}}}^\tau$ evaluated at 506 hPa (right) and 40°N (right) for $\tau_c = 5$ days (blue), $\tau_c = 1$ week (black), $\tau_c = 1$ month (black, dashed), $\tau_c = 50$ days (red), $\tau_c = 3$ months (black, circles), and $\tau_c = 1$ year (black, stars). The normalized concentrations have been calculated using the convolution (Equation 1) and the approximation to the TTD, $\mathcal{G}(r, \tau_{\Omega_{\text{MID}}})$.

Fig. 11. Figure 11 The seasonal cycle of $\chi_{\Omega_{\text{MID}}}^\tau (r, t|\Omega_{\text{MID}})$ (a) and $\chi_{\Omega_{\text{MID}}}^{50} (r, t|\Omega_{\text{MID}})$ (b), evaluated during the last year of the integration after a statistically stationary state has been reached. The thick black line and thin white contours denote the monthly-mean thermal tropopause and isentropes, respectively, averaged over the last year of the integration (the 300 K, 330 K, and 360 K isentropes are shown). Note that the horizontal axis spans 20°S to 90°N.

Fig. 12. Figure 12 The seasonal cycles $\chi_{\Omega_{\text{MID}}}^\tau (r, t|\Omega_{\text{MID}})$ for $\tau_c = 5$ days (column 1) and $\tau_c = 50$ days (column 3). The seasonal cycles of the modal age $\tau_{\text{mode}} (r|\Omega_{\text{MID}}, t')$ (column 2) and the mean age calculated from the “NH-clock” tracer, $\Gamma (r, t|\Omega_{\text{MID}})$ (column 4) are shown with respect to source time $t'$ and field time $t$, respectively. All fields are evaluated at 312 hPa (top), 506 hPa (middle) and 873 hPa (bottom). Note that the horizontal axis only spans the NH.

Fig. A1. Comparison of $\Gamma (r|\Omega_{\text{MID}})$ (black lines) and mean age derived from the “NH-clock” tracer (red dashed lines), where the clock-based tracer age has been averaged over the last year of the integration. The thick black line denotes the annually averaged climatological mean thermal tropopause.
Fig. A1: Comparison of $\Gamma(\bm{r}|\Omega_{\text{MID}})$ (black lines) and mean age derived from the “NH-clock” tracer (red dashed lines), where the clock-based tracer age has been averaged over the last year of the integration. The thick black line denotes the annually averaged climatological mean thermal tropopause.