

**“Spatial and Temporal Variability of Interhemispheric Transport Times” by Xiaokang Wu et al.**

**Response to Reviewers**

We thank the reviewers for their helpful comments. Our responses to specific reviewer’s comments are given below, in red. A track-changed version of revision of the manuscript is attached at the end of this document,

**Anonymous Referee #1**

The authors have done several new things in this study, and the paper would be improved by highlighting the novel aspects (interannual variability, robust seasonal analysis, comparison with data) of the work. Generally, more discussion of mechanisms, why the results matter, and improved framing would take this paper from being a simple advance in the examination of inter-hemispheric transport to being a more interesting and useful contribution.

We thank the reviewer for their suggestions, and as described below we have revised the manuscript to provide improved justification and more discussion of mechanisms and links to meteorology.

Introduction: The previous literature on interhemispheric transport is treated well. It is not clear from the discussion why neglecting the seasonal and interannual variability in inter-hemispheric transport matters, and why it is important to consider both shorter lived and longer lived trace gases. ... A stronger connection to the chemistry would go a long way for motivating the rest of the study.

We have including the following paragraph in the Introduction that follows the reviewer’s comments: “However, understanding the temporal variability of the transport is important for understanding and interpreting the observed temporal variations in tracer concentrations, and determining the relative role of changes in transport, emissions, sinks, and chemistry for different species. For example, observations of methyl chloroform, or other species with reaction with OH as their primary sink, can be used to infer the abundance of OH (Krol and Lelieveld 2003, Prinn et al 2005, Montzka et al 2011, Liang et al 2017), and knowledge of the seasonal and interannual variability of the transport is required to isolate similar variability in the OH abundance. Similarly, knowledge of the interannual variability of transport from the NH is required for estimates of the variability in emissions or sinks (ocean uptake) of CO<sub>2</sub> from measurements of CO<sub>2</sub> in the SH (e.g., Francey and Frederiksen 2016).”

In each section, explain what we expect to see. The ITCZ is known to drive interhemispheric transport and ENSO is known to change tropical variability in DJF, so the results are not surprising. Unfortunately, the paper currently reads as a methodical discussion of plots. In my opinion, the story of this paper would be more compelling if it were presented as “this is what we expect to see”, “let’s check–this is what we see”, and “these are interesting details”.

We have revised the discussion to address the reviewers concerns. In some places we can start with “this is what we expect”, but in other sections we feel it is better to look at general features before focusing on a particular aspect (e.g. while ENSO is a major contributor to interannual variability it is not the only issue and we prefer not to start with a discussion of ENSO).

For the ENSO discussion, an explanation of why the weaker Walker circulation would lead to the pattern of age difference would be nice. The comparison with the wind anomalies in Wang and Fiedler (2006) in Fig. 2c definitely makes Fig. 9a seem reasonable. The current discussion based on the case study comparison of one El Nino and one La Nina year is less convincing than a more general discussion would be. For example, if the variations in deep convection in the SPCZ region are the norm for La Nina years, a citation here would be helpful. For JJA interannual variability, it is not as obvious what mechanisms are at play.

We have replaced the case study of ENSO with composites, including those from two other simulations. The discussion now focuses on the general features rather individual features that may be only in a single event.

Perhaps the variability is related to changes in the monsoon and therefore the phasing of the MJO (the strong variability in age and the mean age contours are both apparently coincident with the Somali jet, and the authors discuss the ascent in this region). Since ENSO is the primary signal of interannual climate variability, I would also think that checking whether JJA age is correlated with the ONI would be worthwhile, just in case. I do not expect the authors to do any extensive calculations for these dynamical connections rather, I ask for a discussion of the transport in the context of the tropical dynamics. I think a different, longer model run would be necessary to get at the details here, and that is beyond the scope of this paper.

We agree that phasing of the MJO may be playing a role, but we only have monthly-mean output which limits our ability to analyze the MJO. We have however, include a figure and discussion of the ENSO influence during JJA.

I find the comparison to observations makes this paper more valuable; this should be emphasized in the introduction, and the discussion should be expanded (p. 7).

We have expanded the discussion on page 7 a little, but there is not a lot more we can say given the limited data, and have included the following in the Conclusions: “Trace gas observations from surface stations provide support for these model results: The SF6 age derived from tropical measurements varies seasonally with the latitude of the ITCZ in a similar manner to the simulated ideal age, and lower concentrations of tracers with NH sources are observed at the America Samoa station during El Nino years (consistent with slower transport).”

The point that is made in this conclusion is that by examining a bunch of CCM1 models, the robustness of these relationships could be tested. I would contend that we fully expect the gradients of age to be tightly coupled to convection and that Orbe’s work has shown as much—these relationships don’t really need to be tested. However, differences in these relationships between models might have interesting implications for the impacts of dynamics and convective

parameterization on model transport and chemistry and how they differ in the CCM1 models.

We have removed this paragraph as this was not central to our conclusions, and we agree with the reviewer that robustness doesn't need to be tested.

\*P4L16: Why do you use CAM SD? What advantages (and disadvantages) does using the nudged run have for this study?

We have included the following in Section 2. "As the CAM-C1SD simulation uses meteorology from reanalyses it has the advantage over free-running simulations (in which the meteorology is generated internally) in that the tracer distributions can then be directly compared with observations for the same period. However, Orbe et al (2017a) have recently shown there is large uncertainty in specified dynamics simulations due to the transport by parameterized convection."

\*P5L35: Can you discuss the cause of the zonal differences? It looks to me like the monsoon pattern of southerly winds. Also, this discussion seems better suited to the next section on the seasonal variability.

We have included the following discussion of the cause of zonal variations: "These variations in the tracers can again be related to variations in meteorology. In particular, the large seasonal variation over the tropical Indian Oceans is related to seasonal changes in convection and wind direction associated with the South Asian monsoon, i.e., there is deep convection over the equatorial Indian ocean and northerly surface winds in DJF, whereas the deep convection is over the northern subtropics and there are southerly winds in JJA."

\*P8L25-...: Why is the analysis of one El Nino and one La Nina event preferable to examining composites of high/low ONI? It seems that with a composite you could be more clear about mechanisms.

We have replaced the single event analysis with a composite analysis.

Specific edits (content ones have a \*, otherwise these are mostly grammar):

All grammatical errors corrected.

## **Anonymous Referee #2**

In this study, the authors analyze the seasonal and interannual variability of transport times from northern hemisphere midlatitudes to the southern hemisphere for 3 different idealized age tracers emitted over North Hemisphere midlatitudes (one for mean age, and two decay tracers with 5- and 50-day decay times). ... Overall, the results of the study are broadly consistent with findings of previous work regarding seasonal and interannual variability of tracer transport, and the explanations are qualitatively plausible. I do think, however, that the study could benefit from more detailed discussion of the interplay of tracer transport and dynamics and convection.

We have increased the discussion of transport and dynamics in each of the results sections.

For the discussion of interannual variability (ENSO) in particular, the authors could do much more. For example, prior work on the SPCZ-ENSO relationship points to the axis of the SPCZ “diagonal” shifting generally northeastward during El Niño and south- westward during La Niña (see, for example, Vincent et al. 2011; reference appended below). The authors may want to consider placing their results in the context of such spatial displacements of the SPCZ.

This is a good suggestion and we have included the following in the text: “The reverse age-ENSO correlation occurs in the southern tropical Pacific because of interannual variations in the SPCZ. During most winters the SPCZ is orientated diagonally in the north-west to south-east direction, but during some strong El Niño events the SPCZ is shifted north and is more zonally orientated (Vincent et al 2011). During these El Niño years there is less rapid transport of younger air from the NH and older air from the SH high latitudes, and hence older tracer ages, in the south-western tropical Pacific.”

More generally, I wonder about the relative role of changes in intensity of convection are relative to changes in its location (as discussed in a two-box model interhemispheric exchange time in Lintner et al. 2004)?

We have not done a formal analysis of intensity relative to location (this is we think beyond the scope of this study), but have included mention of this in the revised manuscript.

It may also be worth noting that the 1997-1998 El Niño event represented what Cai et al. (2012) have described as a “zonal SPCZ” event, with the SPCZ and eastern Pacific ITCZ effectively merging into a single convection zone near the equator. During other El Niño years, the SPCZ does not experience such an extreme response to ENSO forcing. (Whether zonal SPCZs occur appears to be tied to the flavor of ENSO forcing, as these events are more common during so-called “eastern Pacific El Niños” relative to “central Pacific El Niños”.)

We have replaced the single event analysis with a composite analysis, and included the following in the text: “These changes are connected to changes in location of ITCZ over eastern Pacific, the SPCZ, and convection over northern Indian Ocean / south Asia. For example, the region of convection over southern Asia is displaced to the north during El Niño, reducing transport of young northern hemisphere air into the region.”

Given the consideration of 5-day and 50-day loss tracers, it also seems that performing some analysis with respect to intraseasonal variability, especially the Madden Julian Oscillation (MJO), could be of value.

Unfortunately, we only have monthly mean data which prevents any detailed analysis of the MJO or intraseasonal variations.

P5, Last Paragraph: I think it would be worthwhile to develop a bit more in the way of mechanistic explanation for the zonal variations of age in the tropics. For example, for the

relatively high values over the northern Indian Ocean in summer, presumably this is related to the South Asian monsoon, which (relative to winter) has the “ITCZ” located far to the north and relatively strong cross-equatorial flow, particularly over the western portion of the Indian Ocean (with the Findlater/Somali jet). This does seem to be touched on later.

We have included following discussion at the end of the Section: “These variations in the tracers can again be related to variations in meteorology. In particular, the large seasonal variation over the tropical Indian Oceans is related to seasonal changes in convection and wind direction associated with the South Asian monsoon, i.e., there is deep convection over the equatorial Indian ocean and northerly surface winds in DJF, whereas the deep convection is over the northern subtropics and there are southerly winds in JJA.”

Figure 4 also seems ripe for further discussion. For example, the structure of the standard deviation for the 5-day loss tracer in subtropical to mid-latitudes of the South Hemisphere exhibits relatively high variability co-located with not only the SPCZ but also the South Atlantic Convergence Zone and the South Indian Convergence Zone. While I realize that this might be beyond the scope of the present study, I’d be curious to see how the tracers reflect observed synoptic-scale interactions in these convection zones (see, e.g., Matthews 2012 or Niznik and Lintner 2013).

We have included more discussion of tracers and SPCZ variability (see below), but, as with the MJO, we cannot examine synoptic-scale interactions with only monthly-mean fields. “Seasonality in surface convergence also contributed to the region of enhanced seasonal variability in the subtropical western south Pacific. The South Pacific Convergence Zone (SPCZ) lies within this region, and the orientation and intensity of the SPCZ varies on synoptic through to interannual time scales (e.g., Matthews 2012, Niznik and Lintner 2013). This variability in the SPCZ then results in variability in tracer ages, e.g., when the SPCZ is shifted to the north-east from its climatological there is less rapid transport from the NH and more from SH middle-latitudes, resulting in older ages.”

#### Other Comments

All other comments (e.g. grammatical errors) corrected.

# Spatial and Temporal Variability of Interhemispheric Transport Times

Xiaokang Wu<sup>1,2</sup>, Huang Yang<sup>1</sup>, Darryn W. Waugh<sup>1</sup>, Clara Orbe<sup>1,3</sup>, Simone Tilmes<sup>4</sup>, and Jean-Francois Lamarque<sup>4</sup>

<sup>1</sup>Department of Earth and Planetary Sciences, Johns Hopkins University, Baltimore, Maryland, USA

<sup>2</sup>Department of Atmospheric Sciences, Texas A&M University, College Station, Texas, USA

<sup>3</sup>Goddard Earth Science Technology and Research, NASA Goddard Space Flight Center, Greenbelt, Maryland, USA

<sup>4</sup>National Center for Atmospheric Research, Boulder, Colorado, USA

*Correspondence to:* Darryn Waugh (waugh@jhu.edu)

**Abstract.** The seasonal and interannual variability of transport times from the northern mid-latitude surface into the southern hemisphere is examined using simulations of three idealized “age” tracers: ~~A~~ An ideal age tracer that yields the mean transit time from northern mid-latitudes and two tracers with uniform 50-day and 5-day decay. For all tracers the largest seasonal and interannual variability occurs near the surface within the tropics, and is generally closely coupled to movement of the intertropical convergence zones (ITCZ). There are, however, notable differences in variability between different tracers. The largest seasonal and interannual variability in the mean age is generally confined to latitudes spanning the ITCZ, with very weak variability in the southern extratropics. In contrast, for tracers subject to spatially uniform exponential loss the peak variability tends to be south of the ITCZ, and there is a smaller contrast between tropical and extratropical variability. These differences in variability occur because the distribution of transit time from northern mid-latitudes is very broad and tracers with more rapid loss are more sensitive to changes in fast time scales than the mean age tracer. These simulations suggest that the seasonal/interannual variability in the southern extratropics of trace gases ~~;~~ with predominantly NH mid-latitude sources ~~;~~ may differ depending on the gases’ chemical lifetimes.

## 1 Introduction

Interhemispheric transport is ~~an important aspect~~ important for understanding the global distribution of tropospheric trace gases. In particular, it is important to quantify the pathways and time scales for transport from northern hemisphere (NH) middle latitudes into the southern hemisphere (SH) as anthropogenic emissions of tropospheric ozone precursors, major greenhouse gases, aerosols, and ozone depleting substances occur primarily in the NH.

~~The majority of~~ Most previous studies that have examined interhemispheric transport have used a simple two-box framework to quantify a single ~~interhemispheric~~ interhemispheric exchange time, calculated in terms of the temporal change in the difference between the southern and northern hemispherically integrated tracer mass (e.g., Levin and Hesshaimer, 1996; Geller et al., 1997; Lintner et al., 2004; Maiss et al., 1996; Denning et al., 1999). This metric is useful as it collapses all the transport into a single parameter that can be used for ~~model-data~~ model-observations or inter-model comparisons. However, it is only a gross

measure of interhemispheric transport, with no information of spatial variations in transport times. In particular, it does not distinguish between transport into the southern tropics versus transport into the southern extratropics. Tracer observations and simulations support the existence of a strong tropical-extratropical transport barrier (e.g., Bowman and Carrie, 2002; Bowman, 2006; Miyazaki et al., 2008). In fact, Bowman and Carrie (2002) suggest that it may be more appropriate to use a three box model (with northern extratropical, tropical, and southern extratropical boxes) to quantify tropospheric transport (see also Bowman and Erukhimova (2004)).

Alternatively, recent studies have used observed and simulated SF<sub>6</sub> or simulated idealized mean age tracers to estimate the mean transport time from the NH surface to locations throughout the troposphere (Holzer and Boer, 2001; Waugh et al., 2013). This approach provides a more complete description of interhemispheric transport, quantifying not only differences in transport into the tropics versus southern extratropics, but also differences in transport between the lower and upper troposphere. However, these tracers (and two-box or three-box exchange times) only provide information about the mean transport time, whereas observations and models show there is a wide range of times and paths for transport from the NH surface. More precisely, both observational-based estimates (Holzer and Waugh, 2015) and numerical simulations (Holzer and Boer, 2001; Orbe et al., 2016) of the ~~transit time distribution (TTD)~~ distributions of transit times from the NH ~~surface to the SH mid-latitude surface~~ show very broad distributions in the SH, characterized by young modes and long tails. As a result, the mean transit time to SH locations, which controls the distributions of long-lived trace gases, is much larger than the modal transit time, which is associated with the fast transport pathways that play a much more important role in controlling the distributions of chemical tracers with lifetimes of days to months.

Most of the focus in the above studies has been on the climatological mean transport, with only limited analysis of seasonal and interannual variability. ~~This is especially the case for consideration of more than the mean transport time~~ However, understanding the temporal variability of the transport is important for understanding and interpreting the observed temporal variations in tracer concentrations, and determining the relative role of changes in transport, emissions, sinks, and chemistry for different species. For example, ~~Orbe et al. (2016) performed pulse tracer releases at only four different times in a single year, which meant they could only do a limited analysis of the seasonality and could not examine any interannual variability~~ observations of methyl chloroform, or other species with reaction with OH as their primary sink, can be used to infer the abundance of OH (e.g., Krol and Lelieveld, 2003; Prinn et al., 2005; Montzka et al., 2011; Liang et al., 2017), and knowledge of the seasonal and interannual variability of the transport is required to isolate similar variability in the OH abundance. Similarly, knowledge of the interannual variability of transport from the NH is required for estimates of the variability in emissions or sinks (ocean uptake) of CO<sub>2</sub> from measurements of CO<sub>2</sub> in the SH (e.g., Francey and Frederiksen, 2016).

Here we examine ~~this issue, and examine~~ the seasonality and interannual variability of transport from the NH surface into the SH, considering not only the mean transit times but also faster transport pathways and zonal variations in the transport.

The approach taken is to examine simulations of several tracers with the same NH source region but different time dependencies (loss rates) (e.g., Waugh et al., 2003; Orbe et al., 2016). This approach does not enable the same detailed analysis of ~~TTDs as~~ pulse release simulations (unless a large number of tracers are simulated), but does enable detailed analysis of seasonal and interannual variations (see next section for more discussion). Here we examine 30 year simulations of three idealized “age”



tracers requested as part of IGAC/SPARC Chemistry-Climate Model Initiative (CCMI) (Eyring et al., 2013). One of the tracers (the NH clock or ideal age tracer) yields the mean transit time from the NH source region, while the other two tracers have 50 and 5 day loss rates and provide information on shorter transit times (that is more use for understanding the distributions of short lived trace gases). The long simulations enable an examination of interannual, as well as seasonal, variations of transport into the SH.

The tracers and simulations examined are described in the next section, and the climatological distribution of the tracers presented in Section 3. Then the seasonal and interannual variations are examined in Sections 4 and 5, respectively, with concluding remarks in Section 6.

## 2 Methods

### 2.1 Tracers

We examine interhemispheric transport using simulations of three idealized age tracers: An ideal **mean**-age tracer that yields the mean transit time from the NH source region, and two tracers with uniform decay time of 50 or 5 days.

The governing equation for the ideal **mean**-age tracer  $\Gamma(\mathbf{x}, t)$  is (Haine and Hall, 2002)

$$\frac{\partial \Gamma}{\partial t} + \mathcal{L}(\Gamma) = \Theta(t), \quad (1)$$

where  $\mathcal{L}$  is the linear transport operator and  $\Theta(t)$  is Heaviside function (zero for  $t < 0$  and one for  $t > 0$ ). The boundary condition is  $\Gamma(\Omega, t) = 0$  where  $\Omega$  is the source region, and  $\Gamma(\mathbf{x}, 0) = 0$  initially. In other words, the tracer is initially set to a value of zero throughout the atmosphere, is held to be zero over  $\Omega$ , and is subject to a constant aging of 1 year per year in the rest of the model surface layer and throughout the atmosphere. Here  $\Omega$  is the surface layer between 30°N to 50°N, and the ideal age tracer yields the mean transport time from this region. The ideal age tracer  $\Gamma$  is referred to as age of air from northern hemisphere (AOA\_NH) in CCMI (Eyring et al., 2013).

The two decay tracers have fixed concentration over  $\Omega$  and undergo spatially uniform exponential loss, i.e.,

$$\frac{\partial \chi_T}{\partial t} + \mathcal{L}(\chi_T) = -\frac{1}{T}\chi_T \quad (2)$$

where  $T$  is the constant decay time,  $\chi_T$  is the concentration of tracer with decay time  $T$ , and  $\chi_T(\Omega, t) = \chi_\Omega$  is a constant. We consider tracers with  $T = 5$  and 50 days, that we referred to as the 5-day and 50-day loss tracers (the tracers correspond to NH\_5 and NH\_50 in CCMI).

In our analysis we express the concentration of the loss tracers as an age

$$\tau_T(r, t) = -T \ln \left( \frac{\chi_T(r, t)}{\chi_\Omega} \right). \quad (3)$$

This approach is common in oceanography (e.g., Waugh et al., 2003; Deleersnijder et al., 2001), and enables easier comparison with  $\Gamma$ . The basis for the age definition (3) can be seen by considering the idealized case of steady, advective flow



with no mixing (i.e.  $\mathcal{L}(\chi) = u\partial\chi/\partial r$ , with  $u$  a constant). The tracer concentration satisfying (2) is then given by  $\chi_T(r, t) = \chi_\Omega \exp(-t_{adv}/T)$ , where  $t_{adv} = r/u$  is the advective time from the source region to the interior location, and equation (3) reduces to  $\tau_T = t_{adv}$ , i.e. for purely advective flow the tracer age (3) equals the advective time.

In the simple advective flow case the tracer age is independent of the tracer decay time  $T$ , and tracers with different decay rates yield the same age. However, this is not the case for more realistic flows with mixing, where the tracer age depends on the flow and the tracer decay  $T$ . For a steady flow with mixing the tracer age is (Waugh et al., 2003)

$$\tau_T(r) = -T \ln \int_0^\infty \mathcal{G}(r, t') e^{-t'/T} dt', \quad (4)$$

where  $\mathcal{G}(r, t) = \mathcal{G}(\nabla, \perp)$  is the distribution of transit times, or the elapsed times,  $(t - t')$  since the air at  $(r, t)$  was last at the source region at time  $t'$ , and is referred to as the transit time distribution (TTD) or age spectra) from the source region to ~~spectrum~~. Because of the exponential term inside the convolution integral in (4), tracers with different  $T$  yield different  $\tau_T$ . This is illustrated by considering a loss tracer with decay time  $T$  is much larger than the width of the TTD,  $\Delta$ . In this case (4) reduces to (Hall and Plumb, 1994)

$$\tau_T \approx \Gamma - \Delta^2/T. \quad (5)$$

From this we can see that tracers with smaller  $T$  have a younger  $\tau_T$ , and that for tracers with very slow decay the tracer age is close to the mean age ( $\tau_T \rightarrow \Gamma$  as  $T \rightarrow \infty$ ).

While the ~~above means dependence of~~ the tracer ages on the decay time  $T$  means that the ages cannot be interpreted directly as a transport time scale, it does mean examination of tracers with different decay times highlight different aspects of the distribution of transit times (i.e. analysis of multiple tracers provides information on the characteristics of the TTD). Specifically, the age of a tracer is sensitive to the fraction of transit times less than the decay time of the tracer, but insensitive to transit time much longer than the decay time, as these long transit times carry very little tracer mass.

## 2.2 Model and Analysis

~~The tracer fields examined here are~~ We focus primarily on tracer fields from a simulation with the 4th version of the Community Atmospheric Model with troposphere-stratosphere chemistry (CAM4-chem) (Tilmes et al., 2015; Lamarque et al., 2012) run in “specified dynamics” mode using meteorology from Modern-Era Retrospective Analysis for Research and Application (MERRA) (Rienecker et al., 2011). This corresponds to the CAM4-REFC1SD simulation in Tilmes et al. (2016) and the CAM-C1SD simulation in the recent CCM1 model intercomparison of Orbe et al. (2017b). Here we ~~refer to the model simply as CAM~~ use the latter notation. As the CAM-C1SD simulation uses meteorology from reanalyses it has the advantage over free-running simulations (in which the meteorology is generated internally) in that the tracer distributions can then be directly compared with observations for the same period. However, Orbe et al. (2017a) have recently shown there is large uncertainty in specified dynamics simulations due to the transport by parameterized convection.

The ~~CAM-CAM-C1SD~~ simulation has horizontal resolution of  $1.9^\circ$  latitude by  $2.5^\circ$  longitude, 56 hybrid vertical levels from the surface to 1.87 hPa. For our analysis we interpolate from the hybrid levels to a standard set of isobaric levels spanning

1000 hPa to 10 hPa. The simulation examined was run from 1979 to 2010, after being “spun up” by running 5 years with 1979 meteorology. Here, we examine the monthly averaged fields from January 1980 to December 2009.

We examine the climatological seasonal-mean of the tracer ages (i.e. 30-year average for each month), as well as the seasonal and interannual variability. The seasonal variability is quantified by calculating the standard deviation of the climatological 12 month annual cycle, and is referred to as  $\sigma_{\tau}^{\text{seas}}$  (with  $\tau = \Gamma, \tau_{50},$  or  $\tau_5$ ). The interannual variability is similarly quantified by calculating the standard deviation over 30 years. To minimize the impact of seasonality, the interannual variability is calculated for each season, i.e.,  $\tau$  is averaged over every three months and the standard deviation is calculated of these seasonal means. We focus here on interannual variability for December to February (DJF) and June to August (JJA), which we refer to as  $\sigma_{\tau}^{\text{DJF}}$  and  $\sigma_{\tau}^{\text{JJA}}$ . (For both seasonal and interannual variability, the calculations of the standard deviation are performed at individual locations, and any zonal averaging is done after these calculations.)

### 3 Climatological Distributions

We first examine the climatological seasonal-mean distributions of the tracer ages, [and the connection of these distributions with the general circulation](#). Fig. 1 shows the zonally averaged  $\Gamma, \tau_{50},$  and  $\tau_5$  for northern winter (DJF) and summer (JJA). There is a similar distribution for the different tracer ages, with the smallest values in northern mid-latitudes (close to the source region), oldest surface values at the south pole, weak meridional gradients in northern extratropics, largest meridional gradients in tropics, and relative weak vertical gradients at all latitudes (with slightly positive vertical gradients in the northern hemisphere (NH) and slightly negative gradients in the southern hemisphere (SH)). The spatial distribution of the tracer age shown in Fig. 1 [are-is](#) similar to the distribution of idealized or realistic long-lived tracers shown in previous studies (e.g., Denning et al., 1999; Holzer and Boer, 2001; Miyazaki et al., 2008; Waugh et al., 2013), and can be related to [differences in meteorology and transport between regions](#) [the general circulation \(e.g., Hadley cells and intertropical convergence zone ITCZ\)](#). There is rapid transport from the NH mid-latitude surface into the NH extratropical troposphere, through a combination of along-isentropic and convective mixing, and as a consequence there are weak age gradients in the NH extratropics. There is also rapid low-level transport from NH mid-latitudes into the tropics, but the transport into the SH is “slowed” by convection and rapid vertical mixing associated with the [intertropical convergence zone \(ITCZ\)](#) [ITCZ](#), resulting in large surface meridional age gradients near the ITCZ. The rapid vertical mixing within tropical convection results in very weak vertical tracer gradients within the tropics, and the strong meridional gradients in the tropics persistent into the middle troposphere. In the tropical upper troposphere there is increased meridional transport due to the upper branch of the Hadley Cell, and this results in weaker meridional tracer gradients.

While there is qualitative agreement in the spatial distributions of the different tracer ages, there are substantial quantitative differences. First, there are large differences in the magnitude of the ages, especially in the SH where  $\Gamma \gg \tau_{50} \gg \tau_5$  (consistent with equation (5)). Second, there are differences in the meridional gradients: the meridional gradients of  $\Gamma$  in the tropics are much larger than those in the SH (where  $\Gamma$  is nearly constant), whereas the meridional gradients of  $\tau_5$  are similar in the

tropics and SH. These differences are illustrated in Fig. 3(a,b) which shows the latitudinal variation of the tracer ages at 900 hPa, for DJF and JJA.

These quantitative differences among the tracer ages occur because the TTDs in the tropics and SH are very broad (Holzer and Waugh, 2015; Orbe et al., 2016), and the tracers are sensitive to different aspects of the TTDs. As discussed above,  $\tau_5$  is most sensitive to the shorter transit times whereas  $\Gamma$  is the mean of the TTD and is dependent on the long tail of old transit times. The differences in meridional gradients of the two ages are related to changes in the shape of the TTD with latitude. Orbe et al. (2016) showed there is a transition in shape of the TTD from north of the ITCZ to south of the ITCZ, which they attributed to a change in the relative contribution of rapid, advective pathways from northern mid-latitudes and slow eddy diffusive recirculation of “old” air into the tropics from the SH. The latter has a much larger impact on  $\Gamma$  than on  $\tau_5$ , resulting in a much larger increase in  $\Gamma$  across the ITCZ but relatively constant values in the southern extratropics. By comparison,  $\tau_5$  is most sensitive to very short transit times as it is determined more by rapid advective pathways, resulting in roughly constant meridional gradients of  $\tau_5$  throughout the SH.

The latitudinal gradients in the tracers are much larger than zonal gradients, but there are still some zonal variations. This is illustrated in Fig. 2 which shows the 900 hPa distribution of the climatological  $\Gamma$  and  $\tau_5$  for (a,c) DJF and (b,d) JJA. There are weak zonal variations in the extratropics for both tracers, but noticeable zonal variations within the tropics. For example, in DJF the mean age over the equatorial Indian Ocean is smaller than over the equator of other oceans, whereas in JJA the mean over the northern tropical Indian Ocean is larger than over the Pacific or Atlantic oceans. These variations in the tracers can again be related to variations in meteorology. In particular, the large seasonal variation over the tropical Indian Oceans is related to seasonal changes in convection and wind direction associated with the South Asian monsoon, i.e., there is deep convection over the equatorial Indian ocean and northerly surface winds in DJF, whereas the deep convection is over the northern subtropics and there are southerly winds in JJA.

#### 4 Seasonal Variability

We now examine the seasonality of the tracer ages in more detail. Previous studies have linked seasonal differences in the distributions of tracer to the seasonally-varying Hadley circulation (e.g., Bowman and Cohen, 1997; Bowman and Erukhimova, 2004), and we examine this connection for the tracer ages.

Comparison of the left and right panels of Fig 1 shows seasonal differences in ~~the tracer age distributions, which are again qualitatively similar among the tracers. For example, the all three tracer ages:~~ The location of the largest surface meridional gradients are south of the equator during DJF but north of the equator during JJA, and the near-surface tracer ages at the equator and in the southern tropics are younger in DJF than in JJA (see also Fig. 3(a,b)). There are also seasonal differences away from the surface, with older ages in DJF than in JJA in both northern and southern subtropical middle-upper troposphere.

~~These~~ As expected, these seasonal differences in the tracer ages are linked to the ~~seasonally-varying Hadley circulation (e.g., Bowman and Cohen, 1997; Bowman and Erukhimova, 2004)~~ seasonal variations is the Hadley circulation and location of the ITCZ. The largest surface age gradients occur at the ITCZ, with young ages north of the ITCZ and older ages south. The

latitude of the ITCZ moves with season and there is a corresponding north-south shift in the latitude of large meridional age gradients, i.e. largest surface meridional gradients are south of the equator during DJF but north of the equator during JJA (Fig. 1, 3(a,b)). This results, as will be shown below, in a large seasonality at locations within the seasonal range of the ITCZ, with older ages when the ITCZ is north of the location and younger ages when it is to the south.

5 The seasonality of the Hadley circulation can also explain the seasonality in the tracers in the northern subtropical middle troposphere and southern tropical upper troposphere. During DJF the northern cell is strongest (see arrows in Fig. 1a) and transports “older” ages from the equatorial upper troposphere into the northern subtropical middle troposphere (resulting in older ages in DJF than JJA), whereas during JJA the stronger southern cell (Fig. 1b) increases the transport of “young” air into the southern subtropical upper troposphere (again resulting in older ages in DJF than JJA).

10 As with the climatological distributions, there are quantitative differences in the seasonality (DJF-JJA differences) of the different tracer ages. In particular, the seasonality of near-surface  $\Gamma$  south of  $20^\circ\text{S}$  is much smaller than at the equator, whereas for near-surface  $\tau_5$  there is a smaller decrease in the seasonality from the equator to southern mid-latitudes. This can be seen clearly in Fig. 3(c) which shows the latitudinal variation in the seasonal standard deviation  $\sigma_\tau^{\text{seas}}$ .

As mentioned in the previous ~~sections~~ section, there are zonal variations in the tracer ages that vary with  
15 season. ~~These~~ Again, these zonal variations in the ages are consistent with variations in the ITCZ ~~;~~ (see surface winds (arrows) and convergence (contours) in Fig. 2). The ITCZ is close to the equator in both DJF and JJA over the Pacific, whereas there is a large seasonal variation of ITCZ over the Indian ocean: it is well north of the equator during JJA but south of the equator in DJF. Similar variations occur for the regions of largest meridional age gradients. Associated with the seasonal movement of the ITCZ there is a change in direction of the surface winds, with the largest changes again occurring in the Indian ocean  
20 sector. In particular, in the tropical western Indian ocean there is a strong southward flow during DJF, but a strong northward flow in JJA. This seasonality in wind direction results in a large seasonality in the age.

The spatial variation of the seasonality, and differences between  $\Gamma$  and  $\tau_5$ , can be seen clearly in Figs. 4 and 5 which show maps of surface and vertical cross-sections, respectively, of the seasonal standard deviation  $\sigma_\tau^{\text{seas}}$ . Consistent with the above discussion, the largest values of both  $\sigma_\Gamma^{\text{seas}}$  and  $\sigma_{\tau_5}^{\text{seas}}$  are within the tropics. However, while the peak  $\sigma_\Gamma^{\text{seas}}$  and  $\sigma_{\tau_5}^{\text{seas}}$  occur at  
25 similar latitudes over the Pacific and Atlantic Oceans, the peak  $\sigma_{\tau_5}^{\text{seas}}$  is south of the peak  $\sigma_\Gamma^{\text{seas}}$  in the Indian Ocean sector. Also,  $\sigma_\Gamma^{\text{seas}}$  in the southern extratropics is much smaller than in the tropics ( $\sigma_\Gamma^{\text{seas}}$  is as high as 180 days in the tropics but only ~~round~~ around 10 days in the southern extratropics), ~~where~~ whereas  $\sigma_{\tau_5}^{\text{seas}}$  is comparable in the tropics and southern extratropics (5-10 days). Fig. 5 also shows that large seasonality is generally only near the surface (pressures above 800 hPa). However, there is ~~moderate-to~~ larger seasonality in the northern subtropical mid-troposphere, southern subtropical upper troposphere over the  
30 Indian ocean, and near the tropopause (especially for  $\tau_5$ ).

The seasonal movement of ~~the ITCZ~~ convergence zones can explain much of the seasonality in the tracer ages. In particular, the north-south movement of the ITCZ results in a similar movement of the region of high meridional age gradients, and large seasonality of tracer age for tropical locations, i.e., ~~as the ITCZ moves from north to south of a particular~~ when the ITCZ is displaced south from its climatological location there will be ~~decrease in a~~ decrease in the tracer age, and vice-versa for a northward shift (Lintner et al., 2004). The seasonal migration of the ITCZ varies with longitude, with a much larger variation  
35

over the Indian ocean than over the ~~Eastern-eastern~~ Pacific (e.g., Waliser and Gautier, 1993; Gloor et al., 2007). This is shown by contours in Figs. 2 and 4. ~~This, and~~ results in a wider range of latitudes that the ITCZ crosses during the annual cycle, and hence larger seasonality in tracer ages over the Indian ocean than over the ~~Eastern-eastern~~ Pacific (Fig. 4a). ~~There are some differences in locations of peak seasonality of and over the Indian Ocean sector, with the peak occurring north of the equator while the peak is at or south of the equator. These differences are again related to differences in the mean meridional gradients of the tracers: Only in regions with large meridional gradients do perturbations of the circulation lead to large changes in the tracer).~~

Seasonality in surface convergence also contributed to the region of enhanced seasonal variability in the subtropical western south Pacific. The South Pacific Convergence Zone (SPCZ) lies within this region, and the orientation and intensity of the SPCZ varies on synoptic through to interannual time scales (e.g., Matthews, 2012; Niznik and Lintner, 2013). This variability in the SPCZ then results in variability in tracer ages, e.g., when the SPCZ is shifted to the north-east from its climatological there is less rapid transport from the NH and more from SH middle-latitudes, resulting in older ages.

To quantify the age-ITCZ relationship we compare the latitudinal movement of the ITCZ with the age at a fixed location. Fig. 6 shows the relationship between the simulated 900 hPa  $\Gamma$  or  $\tau_5$  with the ITCZ latitude (calculated as the latitude of maximum convergence at 900 hPa between 15°S-30°N for each longitude) for four different longitudes (corresponding to the Indian, Western Pacific, Eastern Pacific or Atlantic oceans). For both tracer ages and all locations there is a positive correlation, i.e. older age for a more northern location of the ITCZ. There are some differences in the age-ITCZ relationships between the ocean basins, with a more ~~compact~~, linear relationship over the Atlantic and Eastern Pacific than other regions. Over the Indian Ocean the age-ITCZ relationship is nonlinear, especially for  $\Gamma$ , with a more rapid change of age with latitude of the ITCZ when the ITCZ is south of 10°N than north.

Observational evidence for the above relationship between the seasonality of tracer ages and latitude of the ITCZ is found in the ~~estimates of measurements of SF<sub>6</sub> from surface stations. Waugh et al. (2013) showed that a “SF<sub>6</sub> age” from surface, which is an approximation of the ideal age, can be estimated from~~ measurements of SF<sub>6</sub>. ~~Waugh et al. (2013) showed that there~~ There are large annual cycles of SF<sub>6</sub> age derived from measurements in the tropical Indian (Mahe Island, Seychilles; 4.7S, 55.5E ) and Eastern Pacific (Christmas Island; 1.7N, 157.1W ) oceans. ~~The, and the~~ variation of the SF<sub>6</sub> age with latitude of ITCZ at these stations (Fig. 7) is similar to those for the simulated  $\Gamma$  shown in (Fig. 6a,c), including the linear relationship for the Eastern Pacific station but nonlinear relationship for the Indian Ocean station. Unfortunately, there are not stations within the tropical Western Pacific or Atlantic to test the simulated seasonality in these regions.

## 5 Interannual Variability

We now examine the interannual variability of the tracers, first for northern winter (DJF) and then northern summer (JJA).

### 5.1 Northern Winter

As for seasonal variations, Given the above relationship between seasonality variability of tropical convergence zones and tracer ages, we expect the interannual variations of the DJF ages ( $\Gamma$ ) are largest in the tropics – subtropics, and the regions of largest variability for  $\Gamma$  are south of those for  $\tau_5$  (Figure 8) tracer ages to be largest near these convergence zones. As shown in Figure 8 this is indeed the case for DJF, with largest variance generally around or south of the ITCZ (contours) and SPCZ (not marked).

5 The interannual variability is, however, weaker than the seasonality, e.g. the maximum  $\sigma_{\Gamma}^{\text{DJF}}$  is around 50 days compared to 180 days for  $\sigma_{\Gamma}^{\text{seas}}$ . There, and there are also differences in the locations of peak seasonal and interannual variability. For example, the peak  $\sigma_{\Gamma}^{\text{DJF}}$  over the Indian sector is in the central equatorial Indian Ocean, whereas the peak  $\sigma_{\Gamma}^{\text{seas}}$  is north of the equator (with two local maximum maxima). A similar difference in locations of peak values occurs between  $\sigma_{\tau_5}^{\text{DJF}}$  and  $\sigma_{\tau_5}^{\text{seas}}$ , and the tropical - extratropical difference in  $\sigma_{\tau_5}^{\text{DJF}}$  is much smaller than that for  $\sigma_{\tau_5}^{\text{seas}}$  (see also Fig. 3). Again consistent with

10 seasonal variability, the interannual variability is largest near the surface and generally small in the upper troposphere (not shown). The regions of highest  $\sigma_{\Gamma}^{\text{DJF}}$  are generally close to the location of the ITCZ or the South Pacific convergence zone (SPCZ), suggesting that the interannual variability of  $\Gamma$  is again connected to variability in the surface convergence and to the location of the strongest mean tracer gradients. Several

The El Nino - Southern Oscillation (ENSO) is the major cause of interannual variability in low latitude meteorology, and

15 previous studies have linked shown that variability in interhemispheric transport to the El Nino – Southern Oscillation (ENSO) is linked to ENSO (e.g. Elkins et al., 1993; Prinn et al., 1992; Lintner et al., 2004; Waugh et al., 2013). We examine this relationship here by calculating the correlation  $r$  and regression  $m$  coefficients between the 30-year times series of DJF  $\Gamma$  at each location with the Ocean Nino Index (ONI) (where  $\text{ONI} > 0.5$  indicates an El Nino event, while  $\text{ONI} < -0.5$  indicates a La Nina event). A 30-year time series is too short to do a detailed analysis of ENSO and transport, but it does provide some

20 guidance on possible ENSO-related variability. As shown in Fig. 9a there show maps of the regression coefficients (shading) and correlation coefficients (contours) for the  $\Gamma$ -ENSO relationship. There are coherent regions with large positive or negative ONI correlations, with both occurring either side correlations in both hemispheres, with correlations north of the equator generally the opposite sign to those south of the equator at the same longitude. There is large region with positive correlation in the southern subtropical central Pacific (near 170°E), but negative correlations are found in southern subtropical eastern Pacific

25 and Indian oceans. Thus, during an El Nino year there tends to be older ages over the southern subtropical central Pacific but younger ages over the southern subtropical eastern Pacific and Indian oceans, and the reverse for La Nina years. The age-ONI  $\Gamma$ -ENSO correlations at and north of the equator are generally the opposite sign to those south of the equator at the same longitude, i.e. there are negative correlations in northern tropical central Pacific. A similar pattern of correlations with ENSO is also found for  $\tau_5$ , with region of positive correlations in south Pacific, consistent with above analysis, the region of largest

30 correlations slightly south of that for correlation with  $\Gamma$  (not shown Fig. 9b).

The above age-ENSO correlations are illustrated in  $\Gamma$ -ENSO correlations are exemplified further by considering composites of  $\Gamma$  and meteorological fields for El Nino years (ONI greater than 1) and La Nina years (ONI less than -1). Fig. 9b-e which shows maps of DJF  $\Gamma$ , CAM-CAM-CISD precipitation (as a proxy for the intensity of tropical convection), and surface winds for a strong El Nino year (1998, ONI=2.1) and a strong La Nina year (2000, ONI=-1.6) the El Nino and La Nina composites;

35 in panels (b,c) the full fields are shown whereas in panels (d,e) the anomalies from the 30-yr climatology are shown. There is a

large difference in precipitation over the Pacific between these years: During the El Nino year there is high precipitation located south of the equator. These maps show a difference in the location of the region of high precipitation (ITCZ) over the tropical Pacific between ENSO phases: During El Nino years the ITCZ is south of its location during La Nina years (around 5°S that extend across the Pacific, while in the La Nina year the precipitation is higher on western than eastern side of the Pacific and there are two regions of high precipitation fluxes, one north (compared to around ~10°N) and the other south (~15°S) of the equator. These difference in the location of peak convection result in. This results in differences in transport to the equatorial western-central Pacific. During the El Nino year El Nino years there is rapid, direct low-level transport to the equator as the deep convection is south of the equator, consistent with younger ages. In contrast for the La Nina year, whereas for La Nina years the convection around 10°N reduces this direct transport and the  $\Gamma$  is older in the same region (consistent with the negative correlation shown in Fig. 9a). The reverse correlation occurs south of the equator because of ENSO variations in deep convection in the SPCZ region, which modify the transport of very old air from southern extratropics back into the southern tropics, i.e., during La Nina years the SPCZ convection reduces this transport resulting in younger ages in southern  $\Gamma$ . ENSO correlation occurs in the southern tropical Pacific because of interannual variations in the SPCZ. During most winters the SPCZ is orientated diagonally in the north-west to south-east direction, but during some strong El Nino events the SPCZ is shifted north and is more zonally orientated (Vincent et al., 2011). During these El Nino years there is less rapid transport of younger air from the NH and older air from the SH high latitudes, and hence older tracer ages, in the south-western tropical Pacific.

The sign of the age-ENSO correlations over the eastern Pacific - Atlantic and the Indian oceans are opposite to that over the western-central Pacific, i.e. there is positive correlation in the northern tropics over eastern Pacific but a negative correlation over the western Pacific (Fig. 9a). The cause of this is not clear, although it likely due to El Nino - La Nina differences in the subtropical surface flow over these these regions. For example, during El Nino years the 1997-1998 El Nino year the equatorial winds over the equatorial Indian ocean have a stronger than average northward component, which transport more old, southern hemisphere, air across the equator resulting in older age in the northern tropical Indian ocean.

**Observational** Some caution is needed with the simulated age-ENSO relationship as it is based only on a 30 year simulation. However, analysis of the  $\Gamma$ -ENSO relationship in two free-running CAM4-chem simulations yields very similar regression patterns, including high negative and positive correlations either side of the equator in western-central Pacific and the opposite signed correlations over the eastern Pacific and Indian Oceans, see Fig. 10. (In the "REFC1" simulation CAM4-chem is constrained by observed sea surface temperatures (SSTs) and sea ice concentrations (SICs), whereas "REFC2" is a simulation where the atmosphere is coupled to dynamic ocean and sea ice models.)

**Observational** support for the above age-ENSO correlation is found in trace gas measurements at America Samoa (14°S, 170°W). Measurements of methyl chloroform and CFCs from this station show lower concentrations (indicating slower transport from NH sources) during El Nino year-years (e.g, Elkins et al., 1993; Prinn et al., 1992). As America Samoa lies just inside the region of positive age-ONI correlation, this is consistent with the above simulated variability. The simulations indicate that the observed result of slower transport to the SH during El Nino years may hold only in the western-central Pacific, and there could be faster transport to the eastern Pacific or Indian subtropical oceans. Unfortunately similar multi-year trace gas measurements are not available from these locations to test this.



Some caution is needed with the simulated age-ENSO relationship as it is based only on a 30 year simulation, that includes only two major El Ninos (1982/83 and 1997/98). However, preliminary analysis of the age-ENSO relationship in a CAM4chem REFC2 simulation covering 1960 to 2100 yields correlation patterns similar to those shown in Fig. 9a. Although ENSO explains much of the variability in the Pacific this is not the case for other basins. In particular, the largest interannual variability of  $\Gamma$  and  $\tau_5$  occurs over the southern tropical Indian Ocean, but variability here is only weakly correlated with ENSO. The interannual of the tracers is still related to changes in location of surface convergence and direction of surface winds, but this variability surface flow is not correlated with ENSO or with the Indian Ocean Dipole (not shown); including high negative and positive correlations either side of the equator in western-central Pacific and the opposite signed correlations over the eastern Pacific and Indian Oceans. Further analysis is required to determine the causes of the interannual variability of the flow and transport in the Indian Ocean.

[

## 5.2 Northern Summer

The general characteristics of the interannual variability during northern summer (JJA) ~~is~~ are similar to that in winter, i.e., the largest variability is in the tropics and there is small variability in the SH (especially for  $\Gamma$ ), see Figure 11a,b. ~~The~~ However, the location of the peak interannual variability at the surface varies between seasons, with the peak in  $\sigma_{\tau}^{JJA}$  generally north of that for  $\sigma_{\tau}^{DJF}$ . This is consistent with the more northern location of the ITCZ in JJA, i.e. the peak standard deviation for each season is close to the climatological location of the ITCZ for that season. As in DJF, the largest  $\sigma_{\tau_5}^{JJA}$  is located south of peak  $\sigma_{\Gamma}^{JJA}$ . This is especially true in the Indian Ocean sector, where  $\sigma_{\Gamma}^{JJA}$  is largest around 20°N but  $\sigma_{\tau_5}^{JJA}$  is largest around 5°S. This difference between the tracer ages is again consistent with the differences in their meridional gradients, e.g. there are weak  $\Gamma$  gradients in the tropical Indian ocean but large  $\tau_5$  gradients in southern tropics.

Some of the interannual variability in JJA tracer ages is correlated with ENSO, with older ages over the northern central tropical Pacific and southern subtropical eastern Pacific, and younger ages over south-eastern Asia during the warm phase (El Nino), see Fig. 12. However, as in DJF, this applies mainly to the Pacific ocean and ENSO is not the dominant source of variability over the Indian and Atlantic Oceans.

While the variability of the tracer ages generally decreases with height from the surface, this is not the case for  $\sigma_{\Gamma}^{JJA}$  over the western tropical Indian ocean. Here there is very little interannual variability near the surface for this region, but as shown in Fig. 13 there is a region of high interannual variability at 650 hPa that extends from tropical Africa over the Indian Ocean. Near the surface the largest  $\sigma_{\Gamma}^{JJA}$  occurs around 30°N, but around 650 hPa the largest variability is around 5°N. The large  $\sigma_{\Gamma}^{JJA}$  near the surface can be attributed to the variations of ITCZ (surface convergence), but variability in the ITCZ does not account for the large variability near 650 hPa. A possible cause of the large  $\sigma_{\Gamma}^{JJA}$  at 650 hPa is variability in the ascent in the lower-mid troposphere over this region. During JJA there is a narrow region of strong ascent in the lower-mid troposphere over tropical Africa - Indian Ocean, between the African easterly jet and tropical easterly jet, that does not extend down to the surface but does produce large precipitation in a “tropical rainbelt” south of the surface ITCZ (e.g., Nicholson (2009)). This region of

strong ascent likely impacts meridional tracer transport, and the large  $\sigma_{\Gamma}^{\text{JJA}}$  at 650 hPa could be connected to variability in ascent. This possibility requires further examination.

## 6 Conclusions

The seasonal and interannual variability of transport times from the northern hemisphere mid-latitude surface into the tropics and southern hemisphere has been examined using simulations of idealized “age” tracers. For all tracers the largest seasonal and interannual variability occurs near the surface within the tropics, and is generally closely coupled to variability in the tropical convergence zones (ITCZ, SPCZ). The seasonal migration of the ITCZ is responsible for the majority of seasonality in the tracer ages (with younger ages when the ITCZ is further south), while a large amount of the interannual variability during DJF is due to ENSO-related variations in surface convergence and convection, especially over the Pacific Ocean. Trace gas observations from surface stations provide support for these model results: The “SF<sub>6</sub> age” derived from tropical measurements varies seasonally with the latitude of the ITCZ in a similar manner to the simulated ideal age, and lower concentrations of tracers with NH sources are observed at the America Samoa station during El Nino years (consistent with slower transport).

There are, however, notable differences in the variability of tracers with different time dependencies. The largest variability in the mean age ( $\Gamma$ ) is confined to the tropics, generally close to the location of the ITCZ (or SPCZ), and there is very weak seasonal or interannual variability in the southern extratropics (e.g., the interannual standard deviation of  $\Gamma$  in the southern extratropics is less than 1% of the climatological mean value). In contrast, for the 5-day and 50-day loss tracers the peak variability of the age of tracer tends to be south of the ITCZ, and there is a smaller contrast between tropical and extratropical variability. For example, the DJF interannual standard deviation of the age of the 5-day loss tracer ( $\tau_5$ ) is around 30-40% of the mean in both the tropics and southern mid-latitudes.

These differences in temporal variability of the tracers occur because the tracers are sensitive to different aspects of the TTD (e.g.,  $\tau_5$  is more sensitive to changes in the fast transit scales than  $\Gamma$ ), and this results in differing meridional age gradients. Orbe et al. (2016) noted that fast (advective) transport pathways make only a very small contribution to the TTD south of the ITCZ, and the TTD is dominated by slow (eddy-diffusive) pathways. Changes in these fast transport pathways south of the ITCZ can cause substantial variations in tracers with rapid loss (e.g.,  $\tau_5$ ) as these tracers are sensitive to changes in the fast time scales (and insensitive to changes in transit times much longer than a month as these carry little tracer), but have much weaker impact of the mean transit time (with which is more strongly influenced by the trail-tail of the TTD).

~~One possible concern with this analysis is that it considered only one model. However, analysis of the same tracers from several other models (those considered in (Orbe et al., 2017a)) yield similar patterns of seasonal and interannual variability, and connections to the ITCZ and SPCZ, as presented here (not shown). This suggests that the results presented here are robust, but this will need to be examined using the full suite of models participating in CCMI. An open question is the magnitude of longer term variability and trends in the transport times. This analysis here suggests this will likely be small, but this needs to be examined. The CCMI model simulations of the 21st century offer an opportunity to perform such analysis, and to also test the robustness of the age-ENSO variability presented here.~~

The differing seasonal and interannual variability of the idealized tracers ~~suggest~~ suggests that the seasonal/interannual variability in the southern extratropics of trace gases, with predominantly NH sources, may differ depending on the chemical lifetimes of the gases. For tracers with very long lifetimes (e.g., SF<sub>6</sub> and CFCs) we may expect very weak temporal variability due to transport, whereas for tracers with shorter lifetimes (e.g. non-methane hydrocarbons) there may be noticeable transport-  
5 induced seasonal or interannual variability. Conversely, our study also suggests that combinations of tracers with different lifetimes may be used to constrain the TTD from observations. This possibility requires further examination.

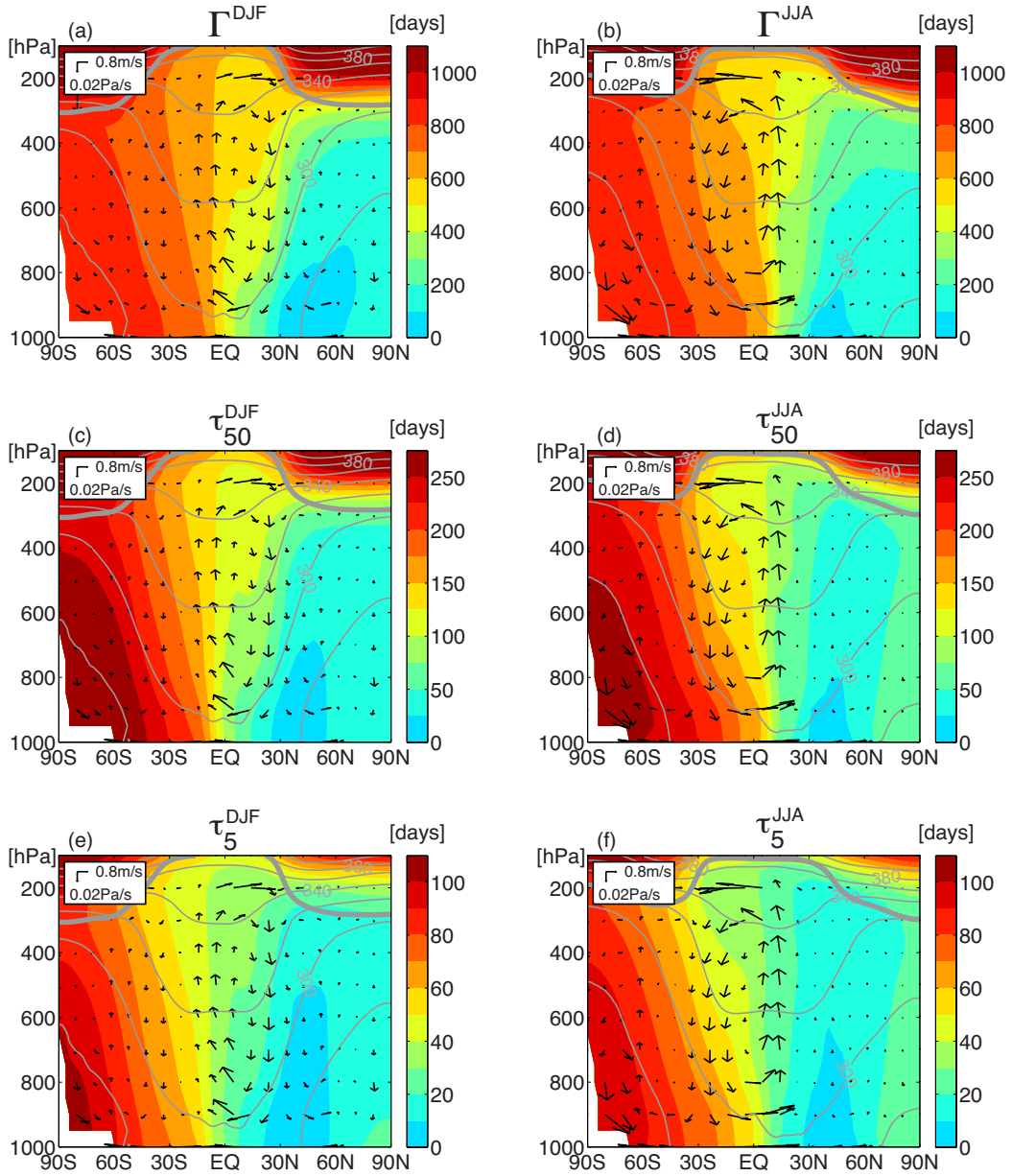
*Acknowledgements.* This work was supported by NSF Grant AGS-1403676 and NASA Grant NNX14AP58G. The CAM4chem output can be downloaded from <https://www.eathsystemgrid.org/search.html?freeText=ccmi1> and the ONI data from <http://www.cpc.ncep.noaa.gov/>.

## References

- Bowman, K. P.: Transport of carbon monoxide from the tropics to the extratropics, *Journal of Geophysical Research: Atmospheres*, 111, 2006.
- Bowman, K. P. and Carrie, G. D.: The mean-meridional transport circulation of the troposphere in an idealized GCM, *Journal of the atmospheric sciences*, 59, 1502–1514, 2002.
- Bowman, K. P. and Cohen, P. J.: Interhemispheric exchange by seasonal modulation of the Hadley circulation, *Journal of the atmospheric sciences*, 54, 2045–2059, 1997.
- Bowman, K. P. and Erukhimova, T.: Comparison of global-scale Lagrangian transport properties of the NCEP reanalysis and CCM3, *Journal of climate*, 17, 1135–1146, 2004.
- Deleersnijder, E., Delhez, E., and Beckers, J.: Some properties of generalized age-distribution equations in fluid dynamics, *SIAM Journal on Applied Mathematics*, 61, 1526–1544, 2001.
- Denning, A. S., Holzer, M., Gurney, K. R., Heimann, M., Law, R. M., Rayner, P. J., Fung, I. Y., FAN, S.-M., Taguchi, S., Friedlingstein, P., et al.: Three-dimensional transport and concentration of SF<sub>6</sub>, *Tellus B*, 51, 266–297, 1999.
- Elkins, J., Thompson, T., Swanson, T., Butler, J., Hall, B., Cummings, S., Fisher, D., and Raffo, A.: Decrease in the growth rates of atmospheric chlorofluorocarbons 11 and 12, *Nature*, 364, 780–783, 1993.
- Eyring, V., Lamarque, J., Hess, P., Arfeuille, F., Bowman, K., Chipperfield, M., Duncan, B., Fiore, A. and Gettelman, A., Giorgetta, M., and Granier, C.: Overview of IGAC/SPARC Chemistry-Climate Model Initiative (CCMI) community simulations in support of upcoming ozone and climate assessments, *SPARC newsletter*, 40, 48–66, 2013.
- Francey, R. and Frederiksen, J.: The 2009–2010 step in atmospheric CO<sub>2</sub> interhemispheric difference, *Biogeosciences*, 13, 873, 2016.
- Geller, L. S., Elkins, J. W., Lobert, J. M., Clarke, A. D., Hurst, D. F., Butler, J. H., and Myers, R. C.: Tropospheric SF<sub>6</sub>: Observed latitudinal distribution and trends, derived emissions and interhemispheric exchange time, *Geophysical Research Letters*, 24, 675–678, doi:10.1029/97GL00523, <http://dx.doi.org/10.1029/97GL00523>, 1997.
- Gloor, M., Dlugokencky, E., Brenninkmeijer, C., Horowitz, L., Hurst, D., Dutton, G., Crevoisier, C., Machida, T., and Tans, P.: Three-dimensional SF<sub>6</sub> data and tropospheric transport simulations: Signals, modeling accuracy, and implications for inverse modeling, *Journal of Geophysical Research: Atmospheres* (1984–2012), 112, 2007.
- Haine, T. W. and Hall, T. M.: A generalized transport theory: Water-mass composition and age, *Journal of physical oceanography*, 32, 1932–1946, 2002.
- Hall, T. M. and Plumb, R. A.: Age as a diagnostic of stratospheric transport, *Journal of Geophysical Research: Atmospheres*, 99, 1059–1070, 1994.
- Holzer, M. and Boer, G. J.: Simulated changes in atmospheric transport climate, *Journal of climate*, 14, 4398–4420, 2001.
- Holzer, M. and Waugh, D. W.: Interhemispheric transit time distributions and path-dependent lifetimes constrained by measurements of SF<sub>6</sub>, CFCs, and CFC replacements, *Geophysical Research Letters*, 42, 4581–4589, 2015.
- Krol, M. and Lelieveld, J.: Can the variability in tropospheric OH be deduced from measurements of 1, 1, 1-trichloroethane (methyl chloroform)?, *Journal of Geophysical Research: Atmospheres*, 108, 2003.
- Lamarque, J., Emmons, L., Hess, P., Kinnison, D. E., Tilmes, S., Vitt, F., Heald, C., Holland, E. A., Lauritzen, P., Neu, J., et al.: CAM-chem: Description and evaluation of interactive atmospheric chemistry in the Community Earth System Model, *Geosci. Model Dev*, 5, 369–411, 2012.

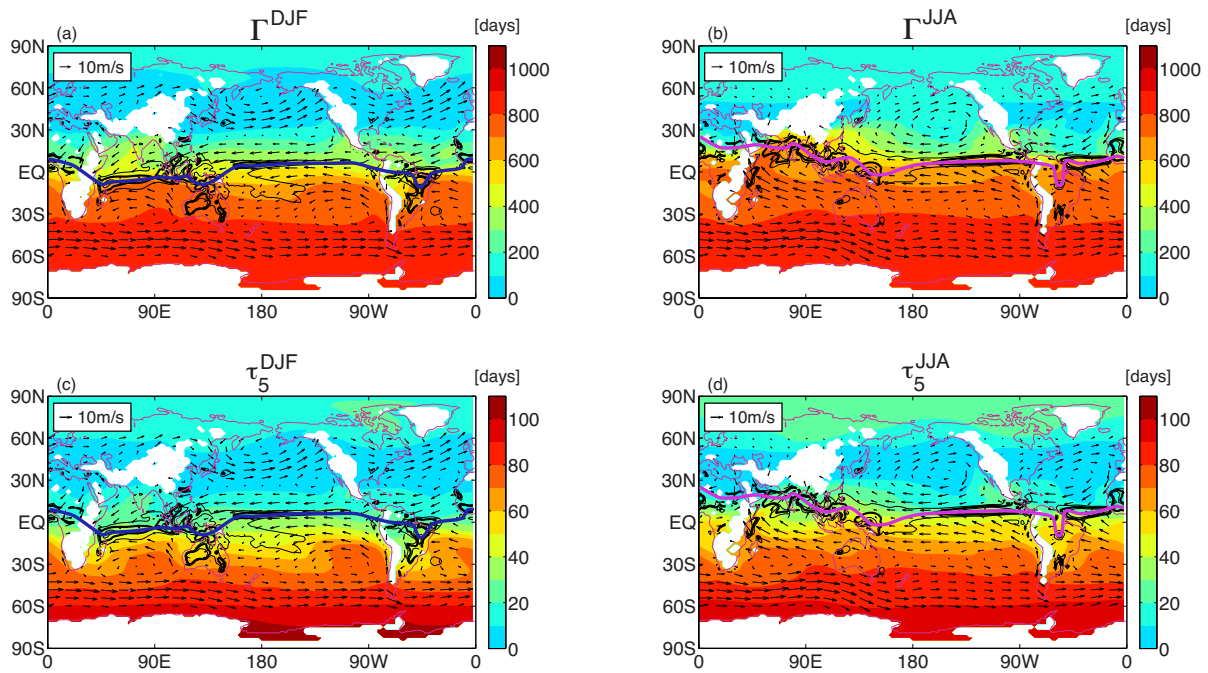
- Levin, I. and Hesshaimer, V.: Refining of atmospheric transport model entries by the globally observed passive tracer distributions of 85krypton and sulfur hexafluoride (SF6), *Journal of Geophysical Research: Atmospheres* (1984–2012), 101, 16 745–16 755, 1996.
- Liang, Q., Chipperfield, M. P., Fleming, E. L., Abraham, N. L., Braesicke, P., Burkholder, J. B., Daniel, J. S., Dhomse, S., Fraser, P. J., Hardiman, S. C., et al.: Deriving Global OH Abundance and Atmospheric Lifetimes for Long-Lived Gases: A Search for CH3CCl3 Alternatives, *Journal of Geophysical Research: Atmospheres*, 122, 2017.
- 5 Lintner, B. R., Gilliland, A. B., and Fung, I. Y.: Mechanisms of convection-induced modulation of passive tracer interhemispheric transport interannual variability, *Journal of Geophysical Research: Atmospheres* (1984–2012), 109, 2004.
- Maiss, M., Steele, L. P., Francey, R. J., Fraser, P. J., Langenfelds, R. L., Trivett, N. B., and Levin, I.: Sulfur hexafluoride—A powerful new atmospheric tracer, *Atmospheric Environment*, 30, 1621–1629, 1996.
- 10 Matthews, A. J.: A multiscale framework for the origin and variability of the South Pacific Convergence Zone, *Quarterly Journal of the Royal Meteorological Society*, 138, 1165–1178, 2012.
- Miyazaki, K., Patra, P. K., Takigawa, M., Iwasaki, T., and Nakazawa, T.: Global-scale transport of carbon dioxide in the troposphere, *Journal of Geophysical Research: Atmospheres*, 113, 2008.
- Montzka, S. A., Krol, M., Dlugokencky, E., Hall, B., Jöckel, P., and Lelieveld, J.: Small interannual variability of global atmospheric hydroxyl, *Science*, 331, 67–69, 2011.
- 15 Nicholson, S. E.: A revised picture of the structure of the “monsoon” and land ITCZ over West Africa, *Climate Dynamics*, 32, 1155–1171, 2009.
- Niznik, M. J. and Lintner, B. R.: Circulation, moisture, and precipitation relationships along the South Pacific convergence zone in reanalyses and CMIP5 models, *Journal of Climate*, 26, 10 174–10 192, 2013.
- 20 Orbe, C., Waugh, D. W., Newman, P. A., and Steenrod, S.: The transit-time distribution from the Northern Hemisphere midlatitude surface, *Journal of the Atmospheric Sciences*, 73, 3785–3802, 2016.
- Orbe, C., Waugh, D. W., Yang, H., Lamarque, J.-F., Tilmes, S., and Kinnison, D. E.: Tropospheric transport differences between models using the same large-scale meteorological fields, *Geophysical Research Letters*, 44, 1068–1078, 2017a.
- Orbe, C. et al.: Tropospheric Transport Differences Between Models Using the Same Large-Scale Meteorological Fields, *Atmos. Chem. Phys. Disc.*, p. to appear, 2017b.
- 25 Prinn, R., Cunnold, D., Simmonds, P., Alyea, F., Boldi, R., Crawford, A., Fraser, P., Gutzler, D., Hartley, D., Rosen, R., et al.: Global average concentration and trend for hydroxyl radicals deduced from ALE/GAGE trichloroethane (methyl chloroform) data for 1978–1990, *Journal of Geophysical Research: Atmospheres* (1984–2012), 97, 2445–2461, 1992.
- Prinn, R., Huang, J., Weiss, R., Cunnold, D., Fraser, P., Simmonds, P., McCulloch, A., Harth, C., Reimann, S., Salameh, P., et al.: Evidence for variability of atmospheric hydroxyl radicals over the past quarter century, *Geophysical Research Letters*, 32, 2005.
- 30 Rienecker, M. M., Suarez, M. J., Gelaro, R., Todling, R., Bacmeister, J., Liu, E., Bosilovich, M. G., Schubert, S. D., Takacs, L., Kim, G.-K., et al.: MERRA: NASA’s modern-era retrospective analysis for research and applications, *Journal of Climate*, 24, 3624–3648, 2011.
- Tilmes, S., Lamarque, J., Emmons, L., Kinnison, D. E., Ma, P.-L., Liu, X., Ghan, S., Bardeen, C., Arnold, S., Deeter, M., et al.: Description and evaluation of tropospheric chemistry and aerosols in the Community Earth System Model (CESM1. 2), *Geoscientific Model Development*, 8, 1395–1426, 2015.
- 35 Tilmes, S., Lamarque, J.-F., Emmons, L. K., Kinnison, D. E., Marsh, D., Garcia, R. R., Smith, A. K., Neely, R. R., Conley, A., Vitt, F., et al.: Representation of the Community Earth System Model (CESM1) CAM4-chem within the Chemistry-Climate Model Initiative (CCMI), *Geoscientific Model Development*, 9, 1853–1890, 2016.

- Vincent, E. M., Lengaigne, M., Menkes, C. E., Jourdain, N. C., Marchesiello, P., and Madec, G.: Interannual variability of the South Pacific Convergence Zone and implications for tropical cyclone genesis, *Climate Dynamics*, 36, 1881–1896, 2011.
- Waliser, D. E. and Gautier, C.: A satellite-derived climatology of the ITCZ, *Journal of Climate*, 6, 2162–2174, 1993.
- 5 Waugh, D., Crotwell, A., Dlugokencky, E., Dutton, G., Elkins, J., Hall, B., Hints, E., Hurst, D., Montzka, S., Mondeel, D., et al.: Tropospheric SF<sub>6</sub>: Age of air from the Northern Hemisphere midlatitude surface, *Journal of Geophysical Research: Atmospheres*, 118, 11–429, 2013.
- Waugh, D. W., Hall, T. M., and Haine, T. W.: Relationships among tracer ages, *Journal of Geophysical Research: Oceans*, 108, 2003.

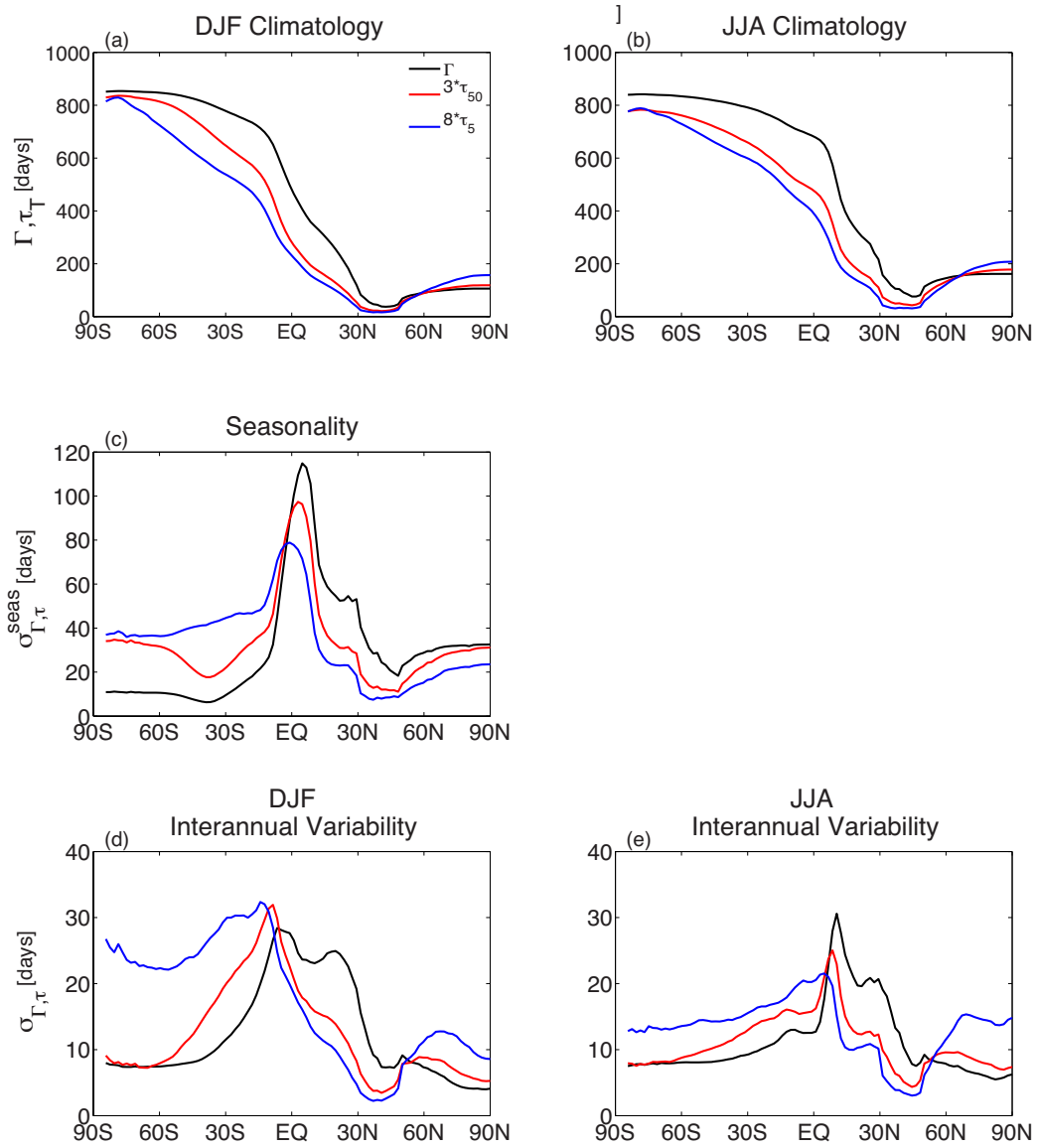


**Figure 1.** Latitude-pressure distribution of the climatological seasonal-mean zonal-mean (a,b)  $\Gamma$  (c,d)  $\tau_{50}$ , and (e,f)  $\tau_5$ , for boreal winter (DJF) and boreal summer (JJA). Arrows show the meridional circulation, thin contours show isentropes (contours every 20 K), and the thick contour shows the tropopause.

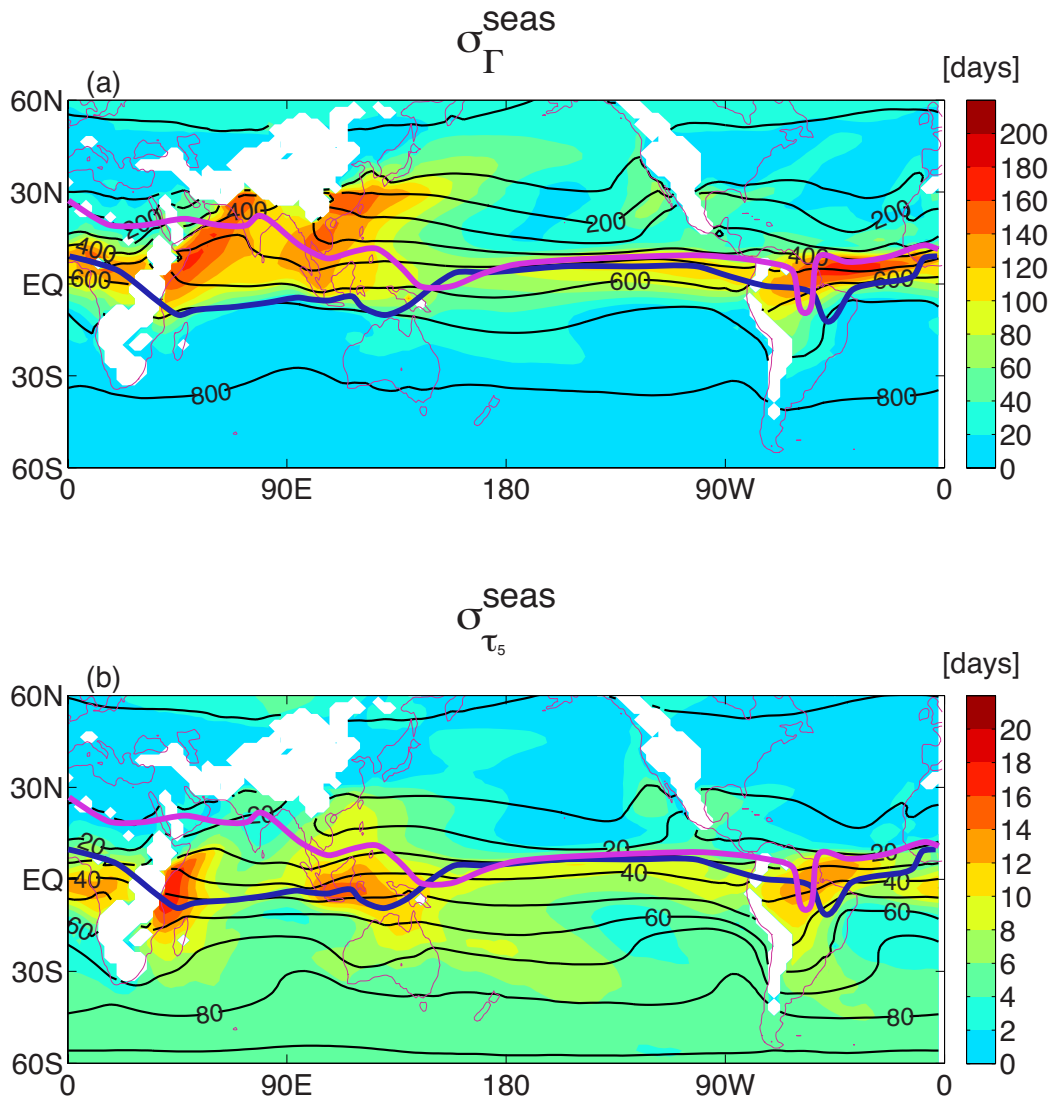




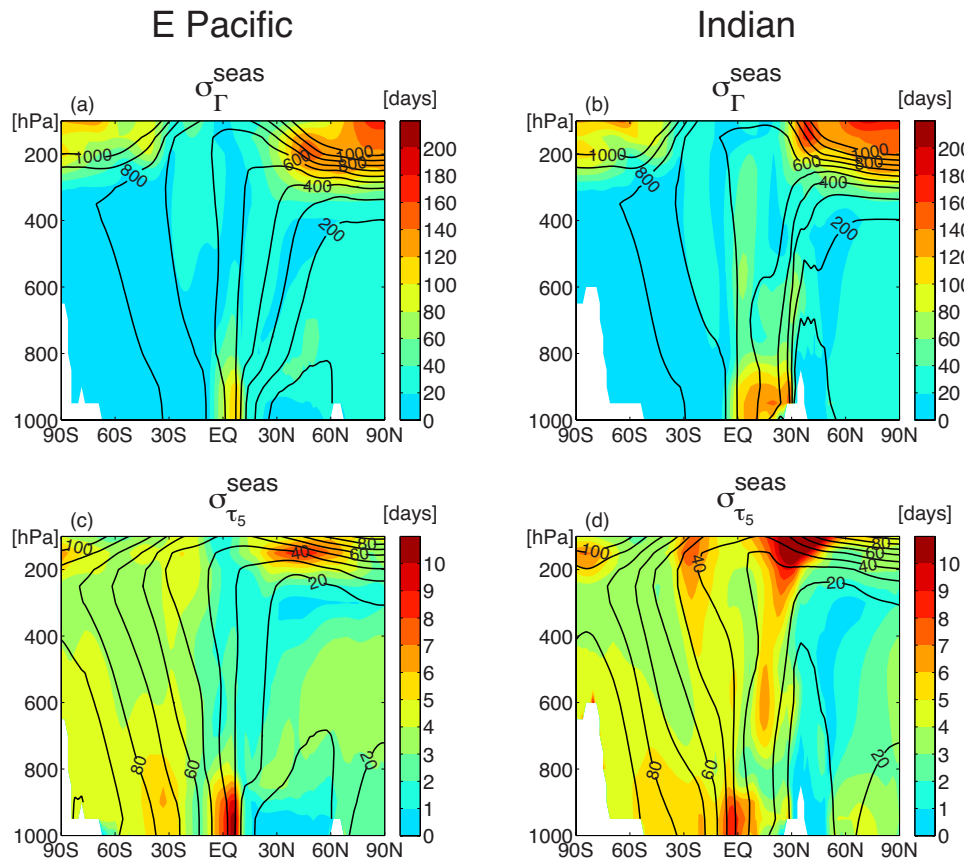
**Figure 2.** Climatological-mean  $\Gamma$  and  $\tau_5$  at 900 hPa for (a,c) DJF and (b,d) JJA. Arrows show horizontal velocity, black contours convergence at 900 hPa (contours at  $(-3,-2,-1) \times 10^6 \text{ s}^{-1}$ , with  $-2 \times 10^6 \text{ s}^{-1}$  bold), and blue/pink curves ~~approximation~~ show the approximate location of the ITCZ.



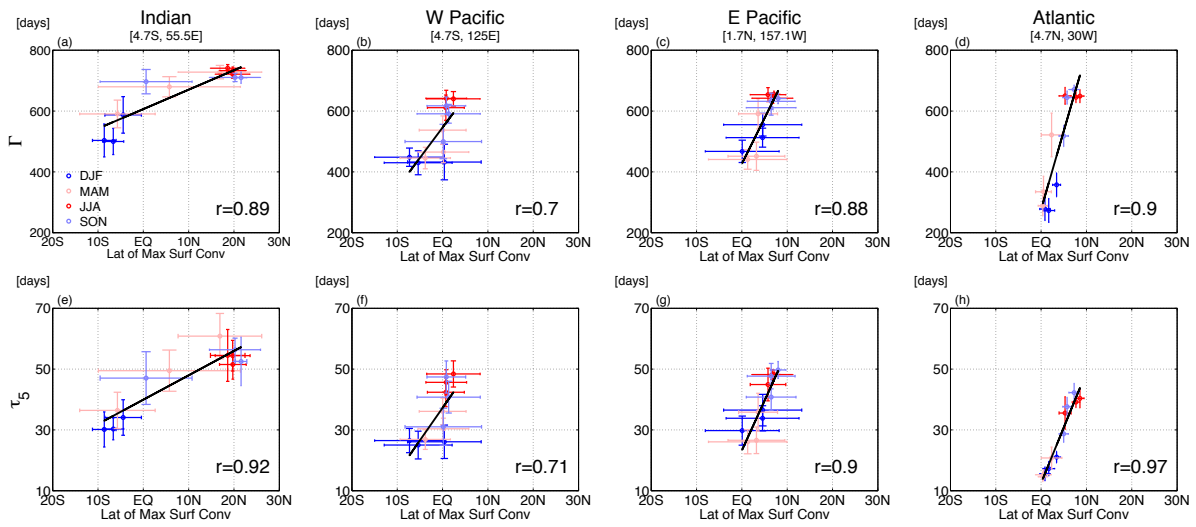
**Figure 3.** Latitudinal variation of (a) DJF climatological mean, (b) JJA climatological mean, (c) seasonal standard deviation, (d) DJF interannual standard deviation, and (e) JJA interannual standard deviation for  $\Gamma$ ,  $\tau_{50}$ , and  $\tau_5$  at 900 hPa. For all panels the quantity for  $\tau_{50}$  is multiplied by 3 and that for  $\tau_5$  multiplied by 8.



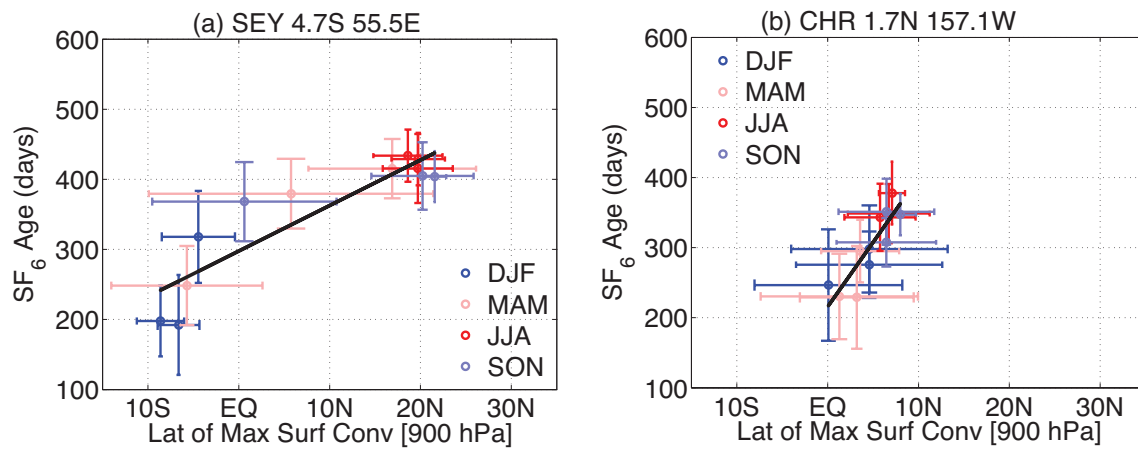
**Figure 4.** Maps of seasonal variability-variability ( $\sigma_S$ ) of (a)  $\Gamma$  and (b)  $\tau_5$  at 900 hPa. Thin contours show climatological mean tracer ages (in days), and blue and pink curves approximation location of the ITCZ in DJF and JJA, respectively.



**Figure 5.** Latitude-pressure variation of  $(\sigma_S)$  of (a,b)  $\Gamma$  and (c,d)  $\tau_5$ , for Eastern Pacific (150-120°W) and Indian Ocean (60-90°E) sections. Contours show climatological mean distributions of (a,b)  $\Gamma$  and (c,d)  $\tau_5$  (in days).



**Figure 6.** Relationship between the latitude of maximum surface convergence and tracer age at 900 Pa for locations in the (a,e) Indian, (b,f) West Pacific, (c,g) East Pacific, and (d,h) Atlantic oceans. Top row shows  $\Gamma$  and bottom row  $\tau_5$ . Coordinates of the locations are shown above panels (a)-(d). Width of the horizontal or vertical bars are twice the interannual standard deviation, and different colors represent different seasons (see panel a). Black line shows linear fit, and correlation coefficient is given within each plot.



**Figure 7.** As in Figure 6a and c except for the relationship between observed SF<sub>6</sub> age with the latitude of the ITCZ, for measurements from (a) Seychilles and (b) Christmas Island.

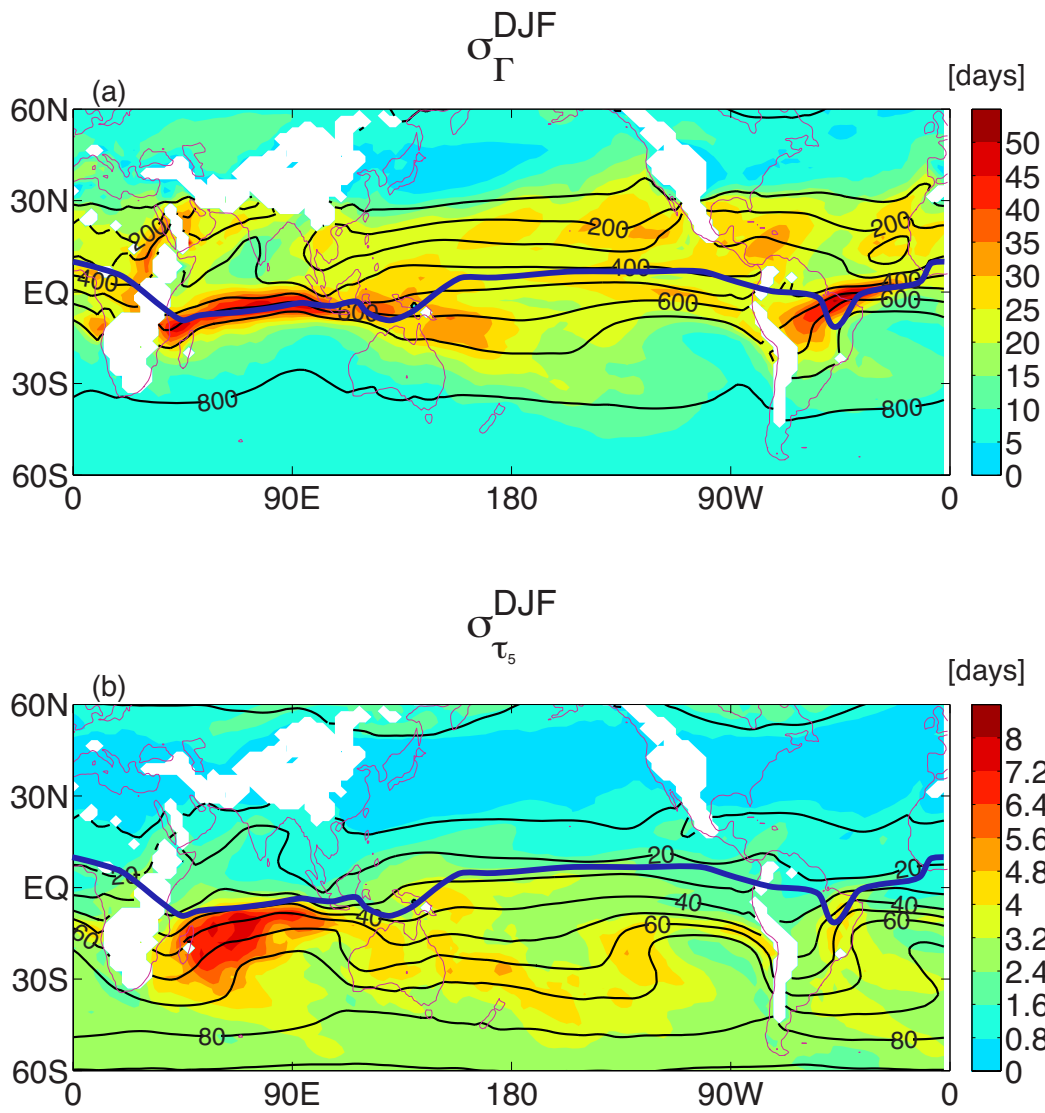
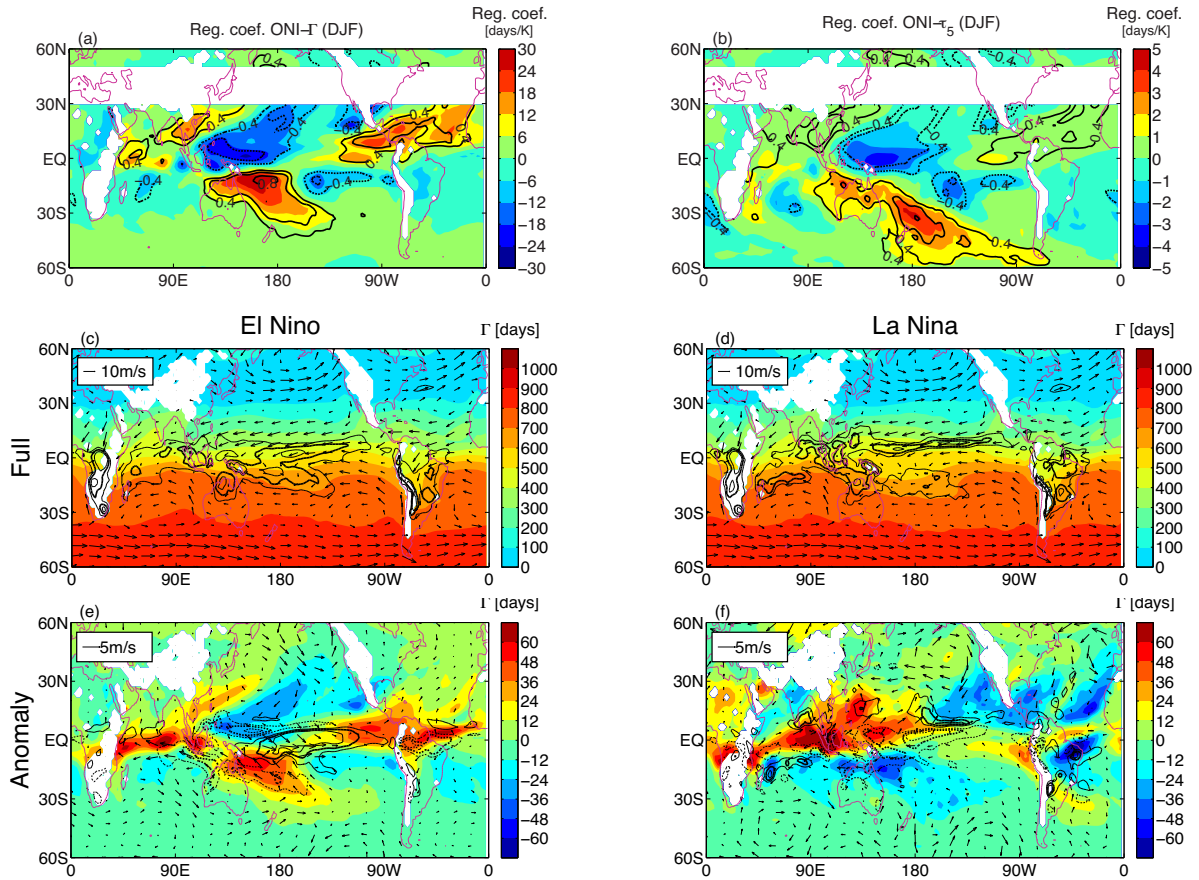


Figure 8. As in Figure 4 except for DJF Interannual standard deviation.





**Figure 9.** Relationships between tracer ages and ENSO. (a) Correlation between ONI-Regression coefficients (shading) and correlation coefficients (contours) between  $\Gamma$  at 900 hPa and ONI for DJF. Correlations with absolute value larger than 0.361 are significant at 95% confidence level. (b) as in (a) except for  $\tau_5$  at 900 hPa. (c,d)  $\Gamma$  (shading), 900 hPa horizontal winds (arrows) and precipitation (contours; (4,6,8) mm/day, with the 6 mm/day bold) for composites of (bc) DJF-1997-98 (El Niño winter) winters and (ed) DJF-1999-2000 (La Niña winter) winters. (d,e) As in (bc,ef) each anomalies from climatological DJF fields. In (de) black contours show precipitation anomalies of (4,6,8) mm/day, and gray contours show precipitation anomalies of (-8,-6,-4) mm/day; while in (ef) black contours show precipitation anomalies of (2,3,4) mm/day, and gray contours show precipitation anomalies of (-4,-3,-2) mm/day.

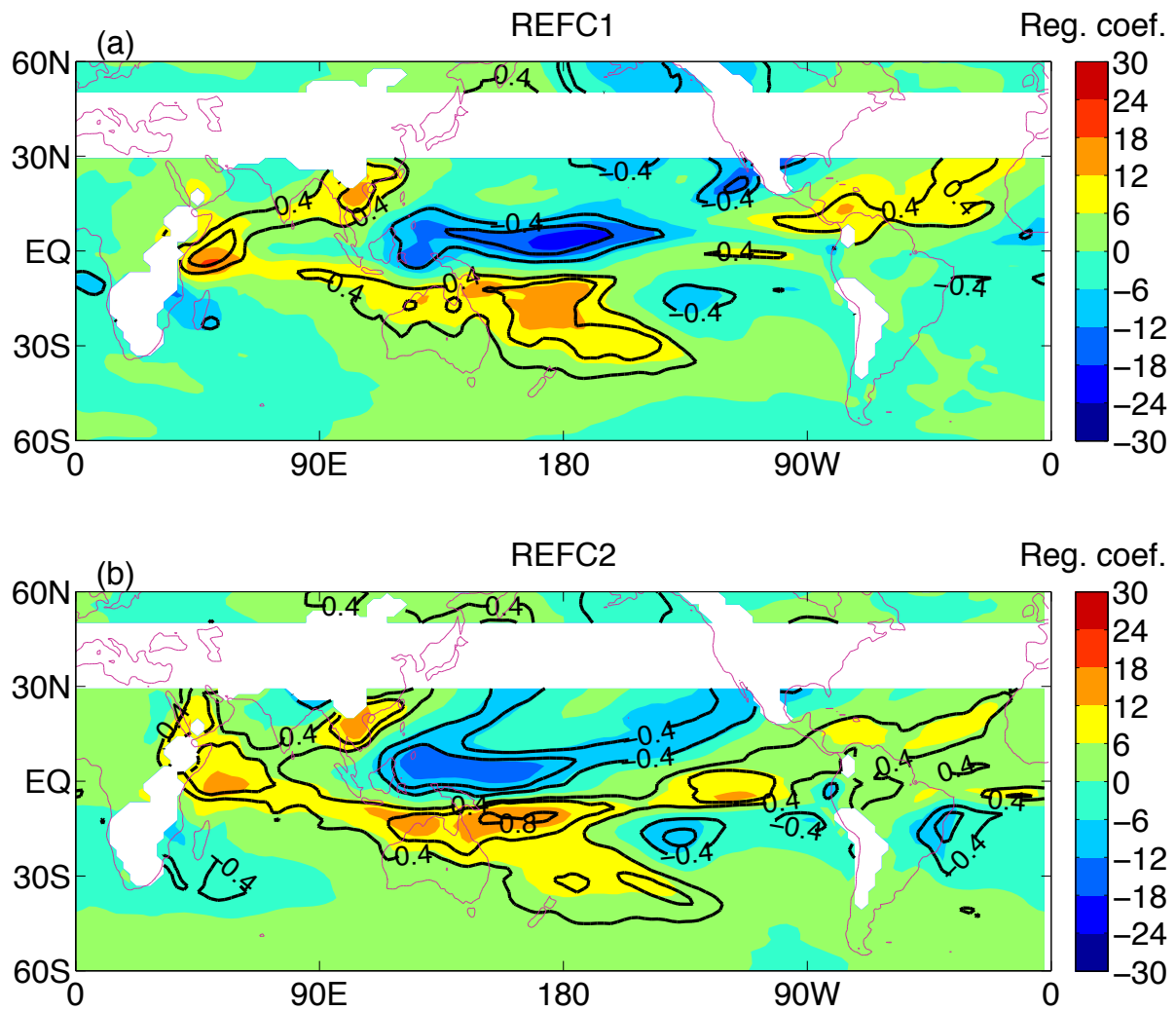


Figure 10. As in Fig. 9(a) except for CAM4Chem (a) REFC1 and (b) REFC2 simulations.

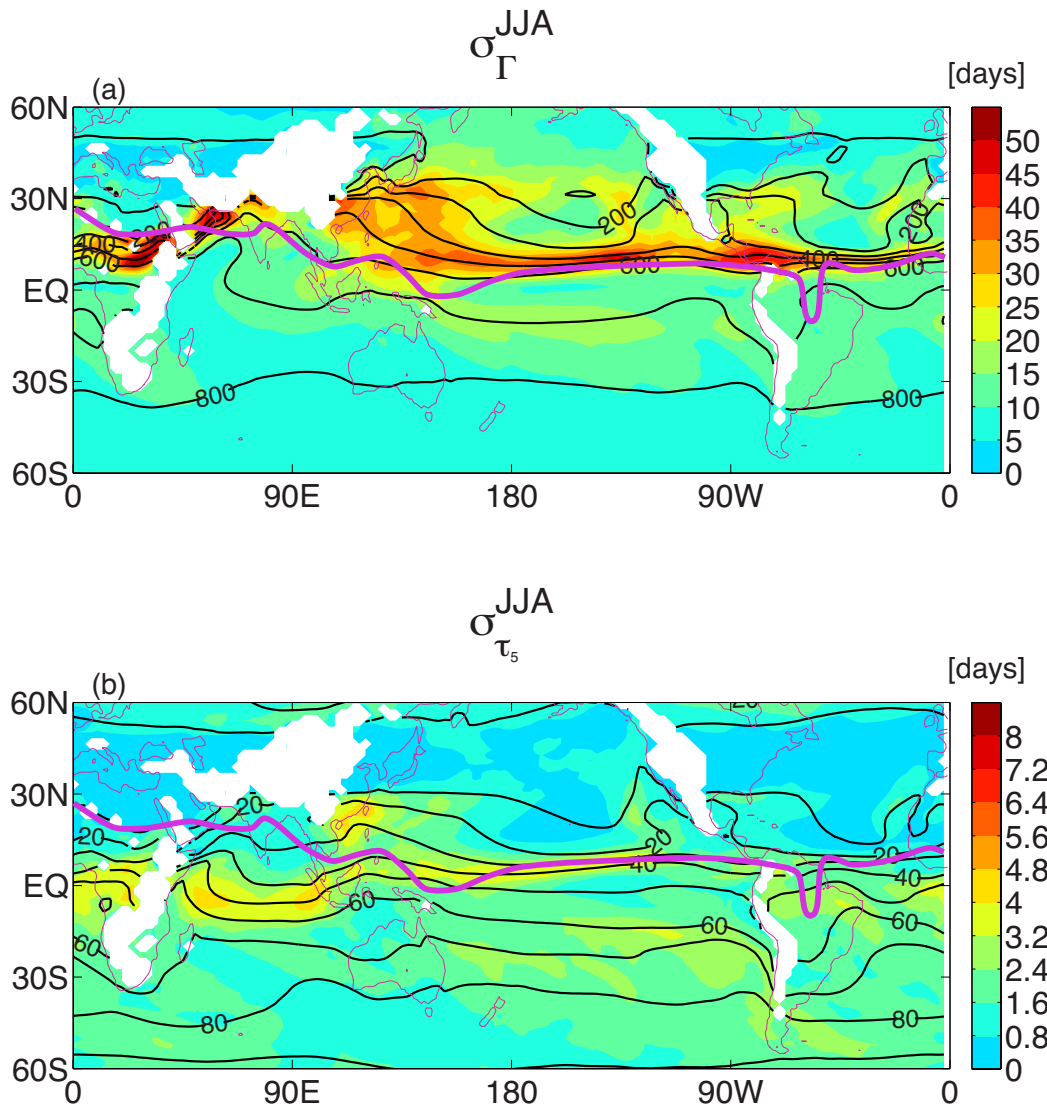


Figure 11. Same as 8 except for JJA Interannual standard deviation.

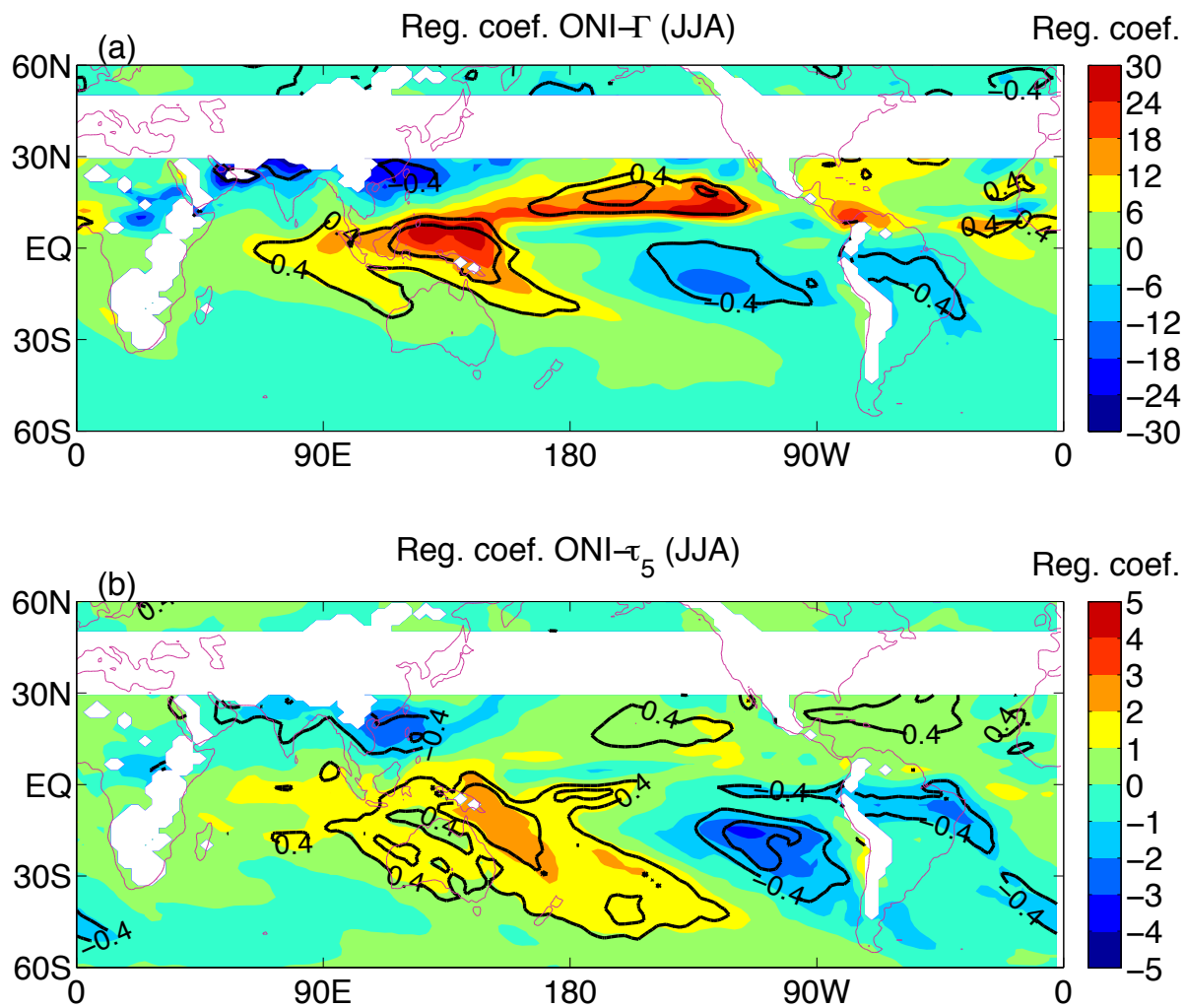


Figure 12. As in Fig. 9(a,b) except for JJA.

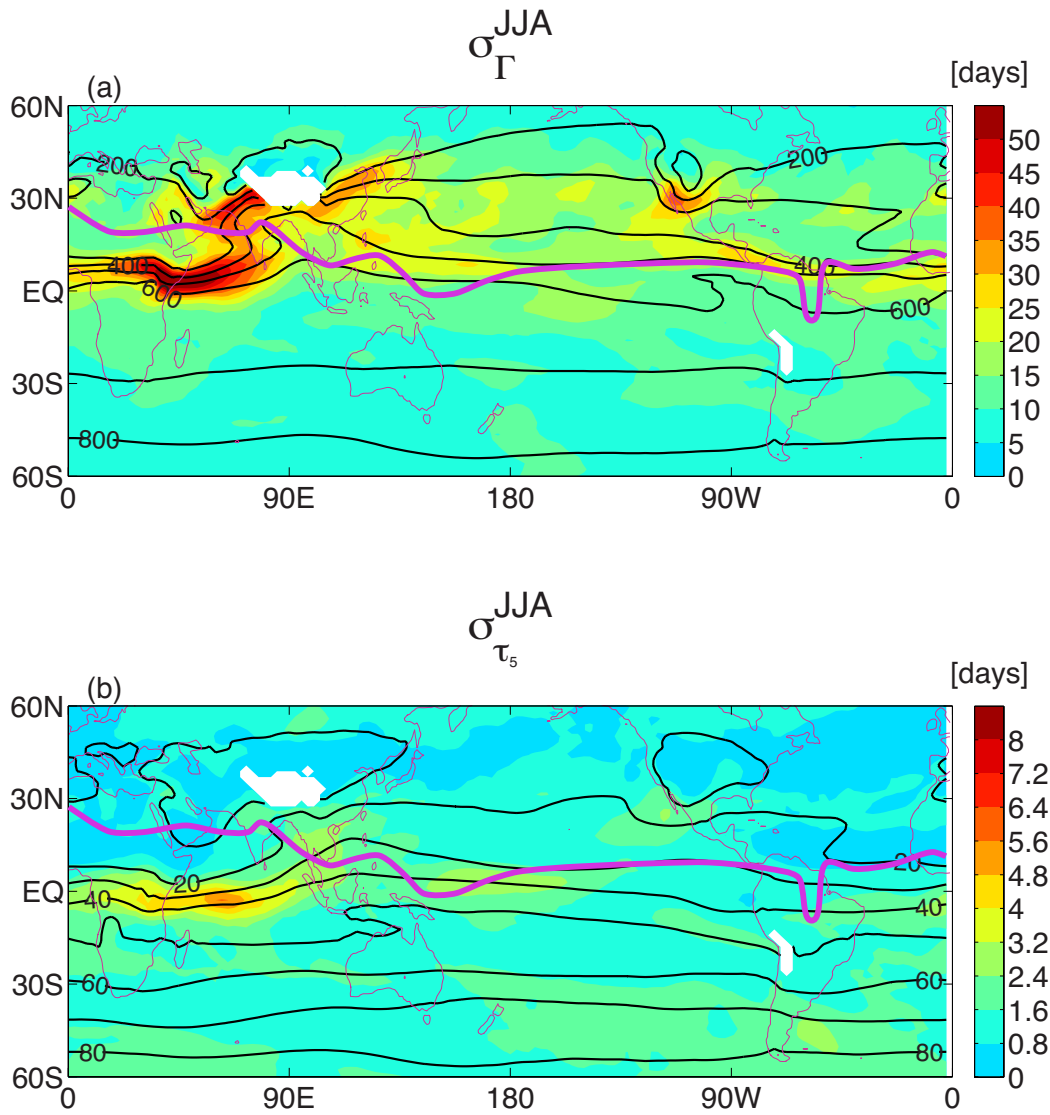


Figure 13. Same as 11 except for 650 hPa.