Estimating the Meridional Extent of Adiabatic Mixing in the Stratosphere using Age-of-Air

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November 22, 2022

Abstract

Wave-induced adiabatic mixing in the winter midlatitudes is one of the key processes impacting stratospheric transport. Understanding its strength and structure is vital to understanding the distribution of trace gases and their modulation under a changing climate. age-of-air is often used to understand stratospheric transport, and this study proposes refinements to the vertical age gradient theory of Linz et al. (2021). The theory assumes exchange of air between a well-mixed tropics and a well-mixed extratropics, separated by a transport barrier, quantifying the adiabatic mixing flux across the interface using age-based measures. These assumptions are re-evaluated and a refined framework that includes the effects of meridional tracer gradients is established to quantify the mixing flux. This is achieved, in part, by computing a circulation streamfunction in age-potential temperature coordinates to generate a complete distribution of parcel ages being mixed in the midlatitudes. The streamfunction quantifies the "true" age of parcels mixed between the tropics and the extratropics. Applying the revised theory to an idealized and a comprehensive climate model reveals that ignoring the meridional gradients in age leads to an underestimation of the wave-driven mixing flux. Stronger, and qualitatively similar fluxes are obtained in both models, especially in the lower-to-middle stratosphere. While the meridional span of adiabatic mixing in the two models exhibits some differences, they show that the deep tropical pipe, i.e. latitudes equatorward of 15\$^{\circ}\$ barely mix with older midlatitude air. The novel age-potential temperature circulation can be used to quantify additional aspects of stratospheric transport.

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¹⁴ Key Points:

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15	- The isentropic formulation of the leaky pipe stratospheric transport model (Linz $$
16	et al., 2021) is used to estimate midlatitude mixing fluxes
17	• A new metric, which quantifies the meridional range of air parcels being mixed
18	across transport barriers, is proposed to estimate mixing
19	• The deep tropical stratosphere is remarkably isolated and mixes with the extra-
20	tropics only in the uppermost stratosphere

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21 Abstract

Wave-induced adiabatic mixing in the winter midlatitudes is one of the key processes im-22 pacting stratospheric transport. Understanding its strength and structure is vital to un-23 derstanding the distribution of trace gases and their modulation under a changing cli-24 mate. age-of-air is often used to understand stratospheric transport, and this study pro-25 poses refinements to the vertical age gradient theory of Linz et al. (2021). The theory 26 assumes exchange of air between a well-mixed tropics and a well-mixed extratropics, sep-27 arated by a transport barrier, quantifying the adiabatic mixing flux across the interface 28 using age-based measures. These assumptions are re-evaluated and a refined framework 29 that includes the effects of meridional tracer gradients is established to quantify the mix-30 ing flux. This is achieved, in part, by computing a circulation streamfunction in age-potential 31 temperature coordinates to generate a complete distribution of parcel ages being mixed 32 in the midlatitudes. The streamfunction quantifies the "true" age of parcels mixed be-33 tween the tropics and the extratropics. Applying the revised theory to an idealized and 34 a comprehensive climate model reveals that ignoring the meridional gradients in age leads 35 to an underestimation of the wave-driven mixing flux. Stronger, and qualitatively sim-36 ilar fluxes are obtained in both models, especially in the lower-to-middle stratosphere. 37 While the meridional span of adiabatic mixing in the two models exhibits some differ-38 ences, they show that the deep tropical pipe, i.e. latitudes equatorward of 15° barely mix 39 with older midlatitude air. The novel age-potential temperature circulation can be used 40 to quantify additional aspects of stratospheric transport. 41

42 1 Introduction

The large-scale stratospheric circulation, known as the Brewer-Dobson Circulation 43 (BDC), plays a primary role in transporting long-lived trace gases throughout the strato-44 sphere, thereby determining their spatial distributions. The BDC brings tropospheric 45 air up into the stratosphere through the tropical tropopause and transports the air ver-46 tically and poleward. The breaking of planetary waves in the upper stratosphere and syn-47 optic waves in the lower stratosphere both drives this meridional circulation across isen-48 tropes (diabatic) and mixes air horizontally along isentropic surfaces (adiabatic). This 49 adiabatic mixing moves tracers over large spatial scales and plays an important role in 50 exchanging midlatitude air with tropical air (Plumb, 2002). From a Lagrangian perspec-51 tive, an air parcel entering the stratosphere will experience both diabatic advection and 52

adiabatic mixing over the course of its time in the stratosphere (Hall & Plumb, 1994;

54 Garny & Randel, 2016).

The trace gases we observe in the stratosphere tend to be those that are long-lived. 55 On short timescales, these gases can be treated as passive tracers advected by the back-56 ground flow. When the relative importance of the chemistry is small, the trace gas con-57 centration is dominantly determined by transport. It has been found that isopleths (sur-58 faces of constant mixing ratio) of different trace gases with varied chemistry have nearly 59 identical shapes (Mahlman et al., 1986; Plumb & Ko, 1992). For instance, trace gases 60 exhibit weak meridional gradients in the midlatitudes due to rapid horizontal mixing. 61 In contrast, observations of trace gases reveal sharp local gradients in concentration in 62 the subtropics and near the edge of the polar vortex, indicating existence of transport 63 barriers (Neu et al., 2003; Shah et al., 2020). 64

Understanding the mixing across these transport barriers is important for climate 65 and climate change: the distribution of trace gases determines their associated radiative 66 forcing, and mixing plays a key role in transporting ozone-depleting substances into the 67 polar vortex where they act as catalysts for ozone depletion (Lee et al., 2001). Intensi-68 fication in isentropic mixing has been known to increase the residence time of trace gases 69 in the stratosphere (Neu & Plumb, 1999; Garny et al., 2014), increasing their likelihood 70 of participating in ozone-destroying chemical reactions. Isentropic mixing is strongly in-71 fluenced by the mean flow and the wave-propagation conditions, especially in the win-72 ter stratosphere (Charney & Drazin, 1961; Holton et al., 1995). Moreover, changes in 73 mixing can further project on to changes in the BDC strength itself, as enhanced mix-74 ing can potentially increase the diabatic upwelling of mass and trace gases into the strato-75 sphere. Therefore, these non-linear changes will collectively play a key role in determin-76 ing transport trends throughout the stratosphere in a changing climate (Ploeger et al., 77 2012, 2015; Abalos et al., 2016). 78

In order to understand short-term changes and long-term trends in isentropic mixing in the stratosphere, it is essential to first accurately estimate the wave-induced mixing fluxes. Several past studies have attempted to investigate the spatial structure of mixing through use of both dynamical and tracer-based metrics (Konopka et al., 2009; Abalos & de la Cámara, 2020). For instance, eddy diffusivity, which depends on the eddy flux of potential vorticity and vorticity gradients, has been used to quantify mixing ex-

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clusively using dynamical quantities (Plumb & Mahlman, 1987; Schneider, 2004). This
idea has also been extended to measure eddy diffusivity of a tracer by redefining the diffusivity in terms of eddy flux and meridional gradient of the trace gas (Nakamura, 2001).
Many other studies have analyzed the seasonality and long-term trends in adiabatic mixing through use of effective diffusivity (Allen & Nakamura, 2001; Shuckburgh et al., 2001;
Chen & Plumb, 2014; Abalos et al., 2016), or other methods including Lyapunov diffusivity (Shuckburgh et al., 2009) and Lagrangian diffusivity (Curbelo et al., 2021).

While each method offers physical insights, each has its own set of limitations. For 92 instance, studies using effective diffusivity frequently employ a coordinate transforma-93 tion and work in the "equivalent latitude" space, instead of regular latitudes. This makes 94 it non-trivial to connect changes and trends in mixing in the specialized coordinates to 95 changes in true mixing; especially as the equivalent latitude is itself defined in terms of 96 tracer contours which may substantially change over time. Moreover, computing effec-97 tive diffusivity requires computing multiple spatial derivatives and integrals, which is not 98 always readily possible for limited observational data. 99

An alternative approach is to formulate the transport-dynamics coupling using the-100 oretical or simple models of stratospheric transport (Plumb, 1996; Neu & Plumb, 1999; 101 Ray et al., 2016). In order to mathematically model trace gas transport, these models 102 study advection of an idealized tracer called "age-of-air" (Hall & Plumb, 1994; Waugh 103 & Hall, 2002). Age-of-air quantifies how long an air parcel has been in the stratosphere 104 and can be defined analytically. It is an effective tool to both quantify the transport timescales 105 of trace gases in the stratosphere, and to study the transport of observed trace gases (Hall 106 & Plumb, 1994). This is because, like many trace gases, one formulation of the age-of-107 air tracer has no spatial sources/sinks in the stratosphere (Boering et al., 1996); only a 108 linear trend at the domain boundary. Thus, there exists a compact relation between the 109 idealized tracer age-of-air and other chemically active trace gases (Plumb & Ko, 1992; 110 Plumb, 2007). In fact, it is possible to directly estimate age-of-air from satellite obser-111 vations of various atmospheric trace gases (Waugh & Hall, 2002; Linz et al., 2017), and 112 age can also be readily computed in climate models either through time-lag analysis or 113 through use of an idealized "clock" tracer (Hall et al., 1999; Garcia et al., 2011; Gerber, 114 2012). A similar approach has been used by several past studies focused on using the 115 age-of-air distribution to infer dynamical properties of the stratosphere (Ray et al., 2010; 116 Garny et al., 2014; Linz et al., 2016, 2021). 117

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Of particular interest to this study are the recent works of Linz et al. (2016) and 118 Linz et al. (2021), which proposed a theoretical framework using age-of-air to estimate 119 the magnitude and vertical structure of both the diabatic circulation strength and the 120 midlatitude mixing flux in the stratosphere. They used age-of-air to assess transport in 121 the stratosphere and connect it to the large-scale circulation. Simply put, Linz et al. (2016) 122 re-formulated the tropical "leaky pipe" model of Neu and Plumb (1999) in isentropic co-123 ordinates and established an inverse relationship between the diabatic circulation strength 124 and the difference between tropical and extratropical age-of-air. Further, Linz et al. (2021) 125 proposed a vertical age gradient theory which connects the net aging of air in the trop-126 ical "pipe" to the combined effects of aging due to the slow diabatic advection within 127 the tropics and to the adiabatic mixing of air between the tropical pipe and the extra-128 tropics. 129

Both the age-of-air and the transport diagnostics proposed in the studies can be computed using satellite observations and/or climate models through straightforward integration. Therefore, this approach can be potentially used to directly and quantitatively connect observations to theory and models. This enables inference of dynamical properties of the stratosphere that are difficult to directly measure using satellite measurements of trace gases, as explore by Linz et al. (2017).

In this study, we propose a framework to improve the mixing flux estimates obtained 136 by Linz et al. (2021). To motivate this analysis, estimates of mixing rates derived by ap-137 plying their theory for two different models, one idealized (GFDL-FV3) and one com-138 prehensive (WACCM), are shown in Figure 1(a). Qualitatively similar mixing structures 139 are obtained for the two models in that the mixing maximizes in the lower stratosphere: 140 near 400 K for the idealized model (red) and near 500 K for the comprehensive model 141 (gray). Above this level, the mixing rapidly decreases to near-zero in the middle strato-142 sphere, before exhibiting a small increase again in the upper stratosphere. The level of 143 maximum mixing differs for the two models on account of a lower tropopause in the ide-144 alized model (Linz et al., 2021). 145

The models, however, show some key differences in the vertical mixing structure as well, observed in both the mixing rates (Figure 1(a)) and mixing efficiency (Figure 1(b)). Mixing efficiency, a dimensionless quantity, is a ratio of the mixing flux to the net poleward mass flux and captures the vertical mixing structure in the stratosphere. Apart

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Figure 1. (a) Derived mixing rate of air (in yr⁻¹) from the winter midlatitudes stratosphere into the tropical stratosphere inferred from (red) an idealized finite volume dynamical core (FV3) with a perpetual January climatology and (grey) a comprehensive model (WACCM) for NDJFM months. Linz et al. (2021) define the mixing rate as the ratio of the estimated mixing flux (units kg/K/s) and the horizontally integrated isentropic density (units kg/K). (b) Mixing efficiency (a proxy for mixing) derived from the inferred mixing rates in (a). Mixing efficiency (ϵ) is a dimensionless quantity and is defined as the ratio of the net mixing flux and the net poleward flux. The net poleward flux, μ_{net} , is defined as the vertical convergence of the net diabatic flux in the tropical pipe. For exact definitions see Equation (17) of Linz et al. (2021).

from the differences in mixing peaks between the idealized (400 K) and the comprehen-150 sive model (500 K), the idealized model exhibits a significantly weaker mixing over the 151 400-600 K interval. These differences cannot be attributed to differences in the tropopause 152 height alone. In fact, the mixing is weak to the extent that the estimated mixing flux 153 and efficiency, which should be non-negative by definition, assumes non-sensical nega-154 tive values around 600 K in the idealized model. Similar mixing differences and nega-155 tive mixing estimates were also found among the models in the annual mean calculations 156 of Linz et al. (2021) (their Figure 5). More pronounced negative mixing fluxes were ob-157 tained by Gupta et al. (2021), who used the theory to compare mixing fluxes among a 158 broad range of idealized models. 159

Rectifying the false negative flux estimates, reconciling the difference in mixing be-160 tween the two models, and ultimately obtaining more accurate mixing fluxes, are the goals 161 of this study. We show that "negative" mixing appears due to an incorrect assumption 162 of a perfectly-mixed tropics and midlatitudes. The Linz et al. (2021) theory assumes fast 163 horizontal mixing within each of the tropical and extratropical "pipes", so neglecting tracer 164 gradients within each region. This effectively implies the deep tropics are mixing just 165 as much as with the extratropics as air at the edge of the pipe, and that the polar air 166 is mixing just as much with the tropics as midlatitude air. This incorrent assumption 167 leads to a potential underestimation of the mixing fluxes across the mixing barrier. In 168 reality, adiabatic mixing is more localized to the winter midlatitudes with only occasional 169 intrusions into the polar vortex or subtropics. 170

It is possible to get a more accurate estimate of mixing fluxes by accounting for 171 subtropical gradients. We refine the theory of Linz et al. (2021) accordingly to account 172 for the sensitivity of the mixing estimates to strong subtropical gradients. These refine-173 ments enable us to improve the mixing flux estimates without compromising the sim-174 plicity of their model. Both the original mixing theory and our refinement to correct for 175 the strong meridional gradients within each "pipe" are discussed in Section 2. Section 176 3 presents the model setup for the idealized and comprehensive model assessed in this 177 study. We apply the proposed theory and discuss our results in Section 4. Finally, we 178 draw conclusions from the analysis and discuss potential avenues for further research in 179 Section 5. 180

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2 A refined theory to estimating the mixing flux

The isentropic formulation of Linz et al. (2016, 2021) begins with the horizontally integrated, mass-flux weighted ages Γ_u and Γ_d over respective regions of diabatic upwelling and diabatic downwelling respectively. They are defined as:

$$\Gamma_u(\theta) = \frac{\int_u \rho_\theta \dot{\theta} \Gamma dA}{\int_u \rho_\theta \dot{\theta} dA} \quad ; \quad \Gamma_d(\theta) = \frac{\int_d \rho_\theta \dot{\theta} \Gamma dA}{\int_d \rho_\theta \dot{\theta} dA} \tag{1}$$

where θ is the potential temperature, $\dot{\theta}$ is the diabatic velocity, $\rho_{\theta} = \frac{-1}{g} \left| \frac{d\theta}{dp} \right|$ is the isentropic density, Γ is the age-of-air, the integrand dA is the infinitesimal area element in the latitude-longitude space, p is the pressure, and the subscripts \int_{u} and \int_{d} respectively denote selective integration over regions of diabatic upwelling $(\dot{\theta} > 0)$ and diabatic downwelling $(\dot{\theta} < 0)$.

Linz et al. (2021) showed that for a steady circulation, in the limit of no vertical diffusion and fast horizontal mixing, the vertical age gradient in the tropical pipe is the sum of aging due to vertical advection and aging due to adiabatic mixing. Mathematically, this is expressed as :

$$\underbrace{\frac{d\Gamma_u}{d\theta}}_{\text{T1: vertical gradient}} = \underbrace{\frac{\sigma_u}{\mathcal{M}}}_{\text{T2: advection}} + \underbrace{\mu_{mix}\frac{\Delta\Gamma}{\mathcal{M}}}_{\text{T3: bulk mixing}}$$
(2)

where θ is the potential temperature, $\Delta \Gamma = \Gamma_d - \Gamma_u$ is the age difference between the 194 extratropics and the tropics, \mathcal{M} is the diabatic mass flux, σ_u (units of kg/K) is the isen-195 tropic density (ρ_{θ}) horizontally integrated over region of upwelling and μ_{mix} is the mass 196 flux per Kelvin that mixes the midlatitude air with the tropical air. The advection term 197 T2 captures the net aging of air as it is advected by the diabatic circulation up the trop-198 ical pipe: the mass of air per unit potential temperature (kg/K) divided by the flux of 199 mass (kg/s)—literally, how long it takes the air to rise one unit of potential tempera-200 ture. The bulk mixing term T3 captures the total aging of the tropical pipe by older air 201 mixed in from the extratropics: $\Delta\Gamma$ quantifies how much older this extratropical air is 202 relative to the tropics, and μ_{mix}/\mathcal{M} quantifies the relative contribution of this air per 203 unit Kelvin. 204

All the terms in the mixing equation except μ_{mix} can be computed from the winds and age-of-air. Thus, the mixing flux μ_{mix} is estimated as a residual in Equation 2. For a more detailed discussion, see Section 3 of Linz et al. (2021) and Appendix A. As we will show, computing the mixing as a residual allows errors in other terms to corrupt the implied mixing flux.

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2.1 Modification to the vertical age gradient equation

Both the original leaky pipe formulation (Neu & Plumb, 1999) and the isentropic formulation of Linz et al. (2021) assume fast horizontal mixing of trace gases within the upwelling and downwelling regions. With sufficiently fast mixing, the horizontal gradient of age-of-air within each region can be ignored, and thus the age of all parcels in each region can be simply assumed to be $\Gamma_u(\theta)$ and $\Gamma_d(\theta)$, the corresponding mass flux weighted age-of-air in the upwelling/downwelling regions. It follows that the entrainment flux trans²¹⁷ ports parcels with a fixed age Γ_u across the barrier towards higher latitudes, and the mix-²¹⁸ ing flux transports parcels with a fixed age Γ_d back to lower latitudes. The mixing term ²¹⁹ T3 in Equation 2 is therefore interpreted as the bulk mixing flux μ_{mix} which mixes the ²²⁰ meridional age difference $\Delta\Gamma$ between the two regions.

The meridional profile of age-of-air estimated from observations and models, how-221 ever, exhibits sharp tracer gradient around the sub-tropical barrier (Waugh & Hall, 2002). 222 For instance, age at 50 hPa from the benchmark tests of Gupta et al. (2020) (shown in 223 Figure 4(b) later) linearly increases from 3 Yr at the equator to 5.5 Yr around the sub-224 tropical barrier. Further poleward, in the surf-zone, the meridional gradient is much weaker 225 on account of wave-induced mixing. Therefore, assuming a perfectly-mixed tropics leads 226 to an overestimation of the actual age difference between air being mixed between the 227 tropics and the extratropics. The midlatitude mixing, which is the main driver of hor-228 izontal churning of trace gases, does not generally extend all the way to the equatorial 229 region. As will be shown in later sections, the deep tropics, especially in the middle and 230 lower stratosphere, are fairly isolated from the wave-induced mixing fluxes that originate 231 in the surf-zone. Consequently, assuming perfectly mixed tropics and extratropics leads 232 to an underestimation of the midlatitude mixing flux μ_{mix} from Equation 2. 233

Consider perturbations $\delta\Gamma_u$ and $\delta\Gamma_d$ to the mean ages Γ_u and Γ_d respectively on a given isentrope, and assume that instead of air with the tropical mean age Γ_u mixing with air with the extratropical age Γ_d , slightly older air in the tropics with age $\Gamma_u + \delta\Gamma_u$ mixes with a slightly younger air from the extratropics with age $\Gamma_d - \delta\Gamma_d$. Physically, this captures the fact that the mixing is more localised around the turnaround latitude and does not span the entirety of the two regions. Re-deriving the mixing equation with this assumption yields the *revised mixing equation* :

$$\frac{d\Gamma_u}{d\theta} = \underbrace{\frac{\sigma_u}{\mathcal{M}}}_{C1} + \underbrace{\mu_{mix}^T \frac{\Delta\Gamma_{eff}}{\mathcal{M}}}_{C2} - \underbrace{\mu_{net} \frac{\delta\Gamma_u}{\mathcal{M}}}_{C4: \text{ enterainment freshening}} (3)$$

where $\Delta\Gamma_{eff} = \Delta\Gamma - (\delta\Gamma_u + \delta\Gamma_d)$ is the effective age difference being mixed across the barrier, following the modification, and μ_{net} is the net poleward flux defined as the vertical diabatic flux convergence, i.e. $\mu_{net} = -\frac{\partial\mathcal{M}}{\partial\theta}$. Substituting $\delta\Gamma_u = \delta\Gamma_d = 0$ yields the original mixing equation of Linz et al. (2021). The detailed derivation is provided in the Appendix. The advective term C2 on the right is identical to T2 in Equation 2.

The total age difference $\Delta\Gamma$ in term T3 in Equation 2 is now replaced with the effective 246 age difference $\Delta \Gamma_{eff}$ in the true mixing term C3 in Equation 3. The age perturbations 247 $\delta \Gamma_u$ and $\delta \Gamma_d$ should be non-negative, so $\Delta \Gamma_{eff} \leq \Delta \Gamma$; the true age difference of the parcels 248 mixed between the two regions is smaller than the age difference $\Delta\Gamma$. In later sections, 249 it is shown that for the idealized dynamical core setup the perturbations $\delta \Gamma_u$ and $\delta \Gamma_d$ 250 are indeed positive. This, however, cannot be said to be generally true for the compre-251 hensive model WACCM due to complications in interpreting the annual cycle. An ad-252 ditional term C4 now appears in Equation 3: the air leaving the tropics is older than the 253 mean age Γ_u and this term corrects the net poleward age transport. For non-negative 254 perturbations, C4 should also be non-negative and provide a positive offset/correction 255 to the mixing flux estimate from Equation 2. The updated C3 and new C4 terms pro-256 vide a positive "correction" to yield the true mixing flux μ_{mix}^T . 257

In order to compute the true mixing flux using equation 3, an estimate of the per-258 turbation ages $\delta \Gamma_u$ and $\delta \Gamma_d$ is needed. To compute these, we define a $\Gamma - \theta$ circulation 259 distribution in the following subsection. The circulation distribution provides a compre-260 hensive description of the age range of parcels being transported at a given latitude and 261 isentropic height allowing us to estimate the perturbation ages $\delta \Gamma_u$ and $\delta \Gamma_d$. To differ-262 entiate between the mixing estimates obtained from Equation 2 and 3, we will hereafter 263 refer to μ_{mix} estimated from Equation 2 as the "bulk mixing flux" and μ_{mix}^{T} estimated 264 from Equation 3 as the "true mixing flux". 265

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2.2 Computing $\delta \Gamma_u$ and $\delta \Gamma_d$ with diabatic circulation as a function of age

The schematic in Figure 2(a) provides an intuitive introduction to the Γ - θ circu-268 lation, showing the meridional wind (black) and age-of-air (color) on a fixed, arbitrar-269 ily chosen, 475 K (\approx 70 hPa) isentropic surface as a function of latitude and longitude. 270 Our goal is to switch from viewing this circulation in longitude to viewing this circula-271 tion in age: at 60°N, for instance, what is the age of mass moving poleward and equa-272 torward? We see that the poleward advected air across this latitude is, on average, younger 273 than the equatorward advected air. The Γ - θ circulation provides the entire distribution 274 of this transport. 275

To compute the Γ - θ circulation, we first isolate a chosen isentropic surface with $\theta = \theta_0$. Further, we selectively consider only a chosen $\Gamma = \Gamma_0$ contour on this chosen isentrope. Finally, we compute the mean meridional mass transport exclusively associated with the chosen potential temperature and age-of-air level. This provides the net mass flux associated with the whole range of ages associated with the transported parcels on the selected θ_0 -isentrope.



Figure 2. (a) A schematic providing physical intuition behind the mathematical formulation of the Γ - θ circulation defined in Equation 4. The solid and dashed curves show the positive and negative meridional velocity on the 475 K isentropic surface, as marked by the orange arrows. The colors show the contours of age-of-air which is transported by the meridional flow across a given latitude (dashed blue). The 5.5 yr age contour is highlighted with orange borders. (b) Γ - θ circulation at 60°N on the 475 K isentrope, as a function of age. The vertical bar in dashed black marks an age of 5.5 Yr corresponding to the orange contours in (a). The age Γ in the horizontal axis corresponds to the integrand Γ' in Equation 5.

The Dirac-delta formulation of the diabatic streamfunction developed by Pauluis 282 et al. (2009), computes meridional mass transport in moist isentropic coordinates in the 283 troposphere through selective binning of mass transport into potential temperature bins 284 or levels. Pauluis et al. (2009) use the approach to estimate a joint distribution of mass 285 transport as a function of both dry and moist potential temperature by using two Dirac-286 delta functions (see Equation (1) of Pauluis et al. (2009) for details). This study follows 287 a similar approach to obtain the joint distribution of the meridional mass transport as 288 a function of both potential temperature (θ) and age-of-air (Γ) . Mathematically, this is 289 akin to using two Dirac-delta functions, one for potential temperature and one for age 290 in order to bin the meridional mass transport according to both the potential temper-291 ature and the mean age of the parcels being transported. The joint Γ - θ meridional mass 292 transport distribution, ψ , can thus be expressed as : 293

$$\psi(\phi,\theta_0,\Gamma_0) = \frac{2\pi R\cos\phi}{g} \int_0^{2\pi} \int_0^{p_s} v\delta(\theta-\theta_0)\delta(\Gamma-\Gamma_0) \, dpd\lambda \tag{4}$$

where ϕ is the latitude, θ_0 and Γ_0 are the select potential temperature and age bins at 294 which the circulation is sampled, v is the meridional velocity on model pressure levels, 295 $\delta(\cdot)$ is the Dirac-delta function, p_s is the surface pressure, dp and $d\lambda$ represent vertical 296 and zonal integration respectively, R is the radius of earth, and q is the acceleration due 297 to gravity. To compute the zonal averages in the integral on isentropic surfaces, the isen-298 tropic binning technique introduced in Yamada and Pauluis (2015) was used, as it al-299 lows us to avoid direct computation of the isentropic density. For brevity, we hereafter 300 refer to the quantity ψ as the Γ - θ circulation. In the northern hemisphere, positive val-301 ues of ψ indicate poleward transport and negative values, equatorward transport. The 302 signs are reversed for the southern hemisphere. 303

Revisiting Figure 2(a), for data originally on pressure levels, the Dirac-delta func-304 tion $\delta(\theta - \theta_0)$ isolates the isentropic surface with $\theta = \theta_0$. Moreover, the Dirac-delta 305 function $\delta(\Gamma - \Gamma_0)$ selectively considers only the $\Gamma = \Gamma_0$ contour on the chosen isen-306 tropic surface. For $\Gamma_0 = 5.5$ Yr, for instance, the two delta functions isolate the 5.5 Yr 307 age contour, highlighted in orange, allowing us to compute the meridional mass trans-308 port along that contour. Finally, computing zonal mean along a latitude circle captures 309 the net mean meridional circulation i.e., computing the net meridional transport only 310 in the regions where the orange highlighted contours meets the (say) 60°N latitude cir-311 cle (dashed blue). We compute the mass transport for a wide range of age levels from 312 0 to 15 Yr and show the mass streamfunction on 475 K and 60° N latitude in Figure 2(b). 313

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For a given latitude circle, such as that shown in dashed blue, the net mass transport is distributed over a broad range of parcel ages. In this case, the circulation moves air parcels with age 4 Yr to 5.7 Yr poleward and air parcels with age 5.7 Yr to 8 Yr equatorward. The equatorward transport is spread over a much wider range at 60°N due to sharp tracer gradients poleward of 60°N on account of the polar vortex edge barrier. This can be clearly seen in Figure 2(a).

For a chosen latitude and potential temperature, ψ provides a distribution of meridional mass transport as a function of age. Therefore, it enables estimation of the complete range of ages of the parcels being transported by the meridional circulation of the stratosphere across a given latitude and on a given isentrope. This distribution is used to compute the true age being mixed around the transport barrier for a given isentrope, which in turn allows us to compute $\delta\Gamma_u$ and $\delta\Gamma_d$ needed for the refined theory in Equation 3.

To estimate the true age of the parcels being mixed between the upwelling and downwelling regions and hence $\delta\Gamma_u$ and $\delta\Gamma_u$, we calculate the Γ - θ circulation-weighted ages separately over the intervals of poleward vs equatorward mass transport at the turnaround latitude. This yields the *effective ages* $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$ of the parcels being mixed across the turnaround latitude on this potential temperature surface. For latitudes in the northern hemisphere, the ages, expressed as a circulation-weighted average, can be expressed as:

$$\Gamma_{u,eff} = \frac{\int_{\psi>0} \psi \Gamma' \, d\Gamma'}{\int_{\psi>0} \psi \, d\Gamma'} \quad ; \quad \Gamma_{d,eff} = \frac{\int_{\psi<0} \psi \Gamma' \, d\Gamma'}{\int_{\psi<0} \psi \, d\Gamma'} \tag{5}$$

where the variable of integration, Γ' , is the age-of-air, and the integration limit $\psi > 0$ means integration only over positive values of ψ . In our computations, Γ' ranges from 0 Yr to 15 Yr, which is the maximum age-of-air in our model runs. The difference between the effective and the mass flux-weighted ages is defined as the perturbation ages $\delta\Gamma_u$ and $\delta\Gamma_d$. More precisely, the Γ - θ circulation is used to estimate $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$, and the perturbations are calculated as $\delta\Gamma_u = \Gamma_{u,eff} - \Gamma_u$ and $\delta\Gamma_d = \Gamma_d - \Gamma_{d,eff}$.

340

2.3 Eddy Diffusivity

The isentropic eddy diffusivity is an alternative method to assess the meridional profile of the mixing flux of age. Plumb and Mahlman (1987) and Schneider (2004) define eddy diffusivity as a diffusive parameterization of eddy fluxes in terms of zonal mean

quantities to obtain closure of the tracer continuity equation. Nakamura (1996), Shuckburgh 344 et al. (2001), and Abalos et al. (2016) quantify mixing using an effective diffusivity based 345 on a Lagrangian treatment of mixing and transformation of equations based on tracer-346 area coordinates. For this study, the former approach is followed on account of its di-347 rectness. The isentropic eddy diffusivity is defined as the ratio of the net eddy transport 348 of age to the mean meridional age gradient. The eddy age flux F_{eddy} is computed as the 349 difference of total meridional age flux and the mean meridional advection of age by the 350 mean flow. Mathematically, in isentropic coordinates, it is expressed as: 351

$$F_{eddy}(\phi,\theta) = \overline{\rho_{\theta}v\Gamma} - \overline{(\rho_{\theta}v)}\tilde{\Gamma}$$
(6)

where, $\tilde{\Gamma}(\phi, \theta) = \overline{\rho_{\theta}\Gamma}/\overline{\rho}_{\theta}$ is the mass weighted age in isentropic coordinates, ρ_{θ} is the isentropic density, v is the meridional velocity, and overbar denotes zonal averaging on fixed isentropes. The isentropic eddy diffusivity \mathcal{D}_{eff} (units m² s⁻¹) is then defined as the ratio of the density-scaled eddy flux and the meridional gradient of age as :

$$\mathcal{D}_{eff} = \frac{1}{\overline{\rho}_{\theta}} \frac{-F_{eddy}}{\partial_{y} \tilde{\Gamma}} \tag{7}$$

where $\partial_y = (1/R)\partial_{\phi}$ is the meridional gradient of the mean isentropic age. The eddy diffusivity is expected to peak in the stratospheric midlatitudes on account of strong planetary wave-induced eddy transport and weak meridional gradients in the surf zone. \mathcal{D}_{eff} can therefore be used to qualitatively compare the structure of midlatitude eddy mixing.

361 3 Models

The revised mixing equation is tested using the two models introduced in Figure 362 1. The first is an idealized model with a prescribed equilibrium temperature profile, a 363 simple treatment of gravity wave drag as a Rayleigh drag at the model top, and no other 364 physical parameterizations in the stratosphere. The second is a comprehensive climate 365 model with a detailed representation of chemistry and radiation. The models have sim-366 ilar numerics in that both employ a finite volume fluid dynamical solver, albeit on dif-367 ferent horizontal grids. This eliminates a key source of uncertainty that can arise due 368 to differences in model numerics itself (Gupta et al., 2020). Working with a comprehen-369 sive model and an idealized model also allows us to test how strongly the negative mix-370

ing fluxes estimated using the vertical age gradient theory are related to complex param eterized processes.

373

3.1 Idealized Model: FV3

The study uses the Free Running model setup used in Gupta et al. (2020), with 374 a finite volume dynamical core based on a cubed sphere grid. The core was developed 375 at the Geophysical Fluid Dynamics Laboratory (GFDL) and is referred to as FV3 for 376 short. The core employs finite volume schemes in both the vertical and the horizontal 377 to solve the primitive equations (Putman & Lin, 2007). FV3 was built as the core of GFDL's 378 Atmospheric Model, Version 3, AM3 (Donner et al., 2011), and a related non-hydrostatic 379 version was recently adopted as the core of the National Center for Environmental Pre-380 diction Global Forecasting System. Further details on FV3 are provided in Gupta et al. 381 (2020, Section 3 and Appendix A). Briefly, the model is driven with thermal forcings (Held 382 & Suarez, 1994; Polvani & Kushner, 2002) detailed in Section 4.1 of Gupta et al. (2020). 383 Newtonian relaxation to an analytically defined temperature state, which can be inter-384 preted as a state of radiative-convective equilibrium, generates a perpetual northern-winter 385 climatology. To quantify transport, a tracer with a concentration that is linearly increas-386 ing in time (a clock tracer) is introduced near the surface ($p \ge 700$ hPa) as detailed in 387 Gupta et al. (2020), Section 4.1. The clock tracer is used to compute the age-of-air, which 388 provides a measure of transport timescales of trace gases in the stratosphere. 389

As in Gupta et al. (2020), the model was integrated for 10,000 days and the last 330 3,300 days were used for transport analysis. The model was integrated at a $1^{\circ} \times 1^{\circ}$ horizontal resolution using a C90 horizontal grid with 80 σ -p hybrid levels in the vertical. The levels were pure- σ (terrain following) below 500 hPa, pure-pressure above 200 hPa and a linear combination of the two between 200 hPa ad 500 hPa.

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3.2 Comprehensive Climate Model: WACCM

The comprehensive climate model employed in the study is the same as that employed in Linz et al. (2021): the Community Earth System Model 1 Whole Atmosphere Community Climate Model (WACCM), an interactive chemistry-climate model (Garcia et al., 2017; Marsh et al., 2013) developed at the National Center for Atmospheric Research (NCAR). This model uses physical parameterizations to represent complex earth system processes including atmospheric chemistry and radiation, and is based on a finitevolume dynamical core (Lin, 2004) from the Community Atmosphere Model, version 4
(Neale et al., 2013). Its domain extends from the surface to 140 km, with 31 pressure
levels from 193 to 0.3 hPa. The horizontal resolution is 2.5° longitude × 1.875° latitude,
corresponding to the F19 horizontal grid.

The WACCM simulations are based on the Chemistry Climate Model Initiative REF-C1 scenario (Morgenstern et al., 2017). An ideal clock tracer, to compure the age-of-air, is included and is specified as in Garcia et al. (2011), with a uniform mixing ratio at the lower boundary that is linearly increasing in time. The model is forced with observed sea surface temperatures. The Quasi-Biennial Oscillation is nudged to observed winds, but otherwise the model evolves freely. The model was integrated from 1979 to 2014. Further details on WACCM are provided in Section 3 of Linz et al. (2021).

413 4 Results

The idealized FV3 model best allows us to contrast the original and revised mix-414 ing equation. Because it is in perpetual northern hemisphere winter, there is a contin-415 uous, strong mixing barrier associated with the Northern Hemisphere polar vortex. We 416 therefore expect the assumptions of Linz et al. (2021) to be less appropriate than in a 417 seasonally-varying context, where the annual breakdown of the vortex partially erodes 418 the barrier. This experiment provides a more stringent test of our revised mixing for-419 mulation. Following that, we discuss our findings in the context of the comprehensive 420 WACCM model. 421

422

4.1 Estimating the mixing fluxes using the refined theory

The climatological winds generated in response to the prescribed diabatic heating 423 in FV3 comprises a strong polar vortex in the northern high latitudes and a strong east-424 erly jet in the subtropics, Figure 3(a). The solid blue lines trace the turnaround latitude 425 in each hemisphere, i.e. the latitude associated with zero diabatic velocity. The "trop-426 ical pipe" enclosed between the two blue curves is characterized by slow diabatic ascent 427 of mass. Likewise, the region poleward of the blue curves is characterized by slow dia-428 batic descent of mass. We refer to these two partitions as upwelling and downwelling re-429 gions respectively. 430

The climatological mean age-of-air profile (in color), obtained as a 10-year aver-431 age, is well-stratified, monotonically increasing with height. At any given height, the youngest 432 air is found within the tropics, and the oldest air either in the winter or summer polar 433 region, depending on the level. The concave, vertically stacked contours in the equato-434 rial region are consistent with slow vertical transport in the region; fresh air from the 435 tropical tropopause is vertically advected upward. Away from the equator, the age in-436 creases monotonically and exhibits a strong gradient around the subtropical barrier. Pole-437 ward of the barrier, in the surf-zone, an abrupt flattening of the age contours is created 438 by enhanced midlatitude mixing. In the winter hemisphere most of this mixing is induced 439 by breaking planetary waves, while in the summer hemisphere it is induced by a com-440 bination of both synoptic-scale wave breaking in the lower stratosphere and slow diffu-441 sive transport elsewhere. In winter high latitudes, a steep increase in potential vortic-442 ity with latitude inhibits transport into and out of the polar vortex. Consequently, an 443 abrupt increase in age is observed near the edge of the polar vortex. 444

The age-of-air in Figure 3(a) when weighted by the diabatic mass flux and averaged over the upwelling and downwelling regions, respectively, yields $\Gamma_u(\theta)$ and $\Gamma_d(\theta)$ (defined in equation 1), shown in Figure 3(b). The downwelling/midlatitude age, Γ_d , is older than the upwelling/tropical age, Γ_u , throughout the stratosphere, with the difference $\Delta\Gamma = \Gamma_d - \Gamma_u$ maximizing near 500 K in the lower stratosphere.

The mixing equation, Equation 2, connects the net vertical aging of air in the trop-450 ical pipe to the aging by diabatic advection vs adiabatic mixing. The net aging, i.e., the 451 vertical gradient of the upwelling age (T1), is maximum near 400 K, above which it mono-452 tonically decreases with height, Figure 3(c). In the lower stratosphere, a major fraction 453 of the aging can be attributed to mixing in the lower stratosphere, as shown in Figure 454 3(d) (solid curve). The other factor contributing to the aging is the reduction of verti-455 cal motion on account of increased static stability as compared to the troposphere. At 456 400 K, the bulk mixing term (T3) accounts for an aging rate of 0.018 yr K^{-1} out of the 457 net vertical aging (T1) of 0.028 yr K^{-1} , Figure 3(c), as the synoptic-scale mixing dom-458 inates in both the hemispheres. This fraction, however, rapidly decreases with height and 459 between 550-600 K almost all the aging is accounted for by the advective term (T2). In 460 fact, our computations yield slightly negative values of the bulk mixing term in this re-461 gion, a clearly nonsensical result. In the upper stratosphere, the increase in the relative 462

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Figure 3. (a) The climatological mean age profile (in color) and zonal mean zonal wind (in black), along with the zero diabatic velocity curve (in yellow) for the FV3 idealized model. (b-d) Individual terms of Equation 2 applied to the idealized model: (b) The diabatic flux weighted upwelling and downwelling ages, Γ_u (solid) and Γ_d (dashed), (c) the vertical gradient of upwelling age, $\partial \Gamma_u / \partial \theta$, i.e., term T1 in the mixing equation, and (d) advection (dashed) and bulk mixing (solid) terms i.e., terms T2 and T3 in the equation respectively. The term T3 is computed as the difference T1-T2, leading to spurious, negative values for the bulk mixing flux μ_{mix} .

463 contribution by the bulk mixing term is due to enhanced mixing due to breaking plan-464 etary waves.

As was pointed out in Linz et al. (2021) and also shown in Figure 1, minor negative fluxes are obtained in the middle stratosphere both for the comprehensive WACCM ⁴⁶⁷ model (annual average) and for the idealized FV3 model. This "negative mixing" is not ⁴⁶⁸ a numerical artifact but rather a consequence of the assumption of fast horizontal mix-⁴⁶⁹ ing within both the upwelling and downwelling regions. While this assumption allows ⁴⁷⁰ convenient assessment of age in the two regions using characteristic values Γ_u and Γ_d , ⁴⁷¹ it ignores the meridional gradient in age around the subtropical transport barrier. The ⁴⁷² flux of older midlatitude air into the tropics is localized around the barrier itself, barely ⁴⁷³ churning the air in the deep tropics.

The bulk mixing term (T3) and bulk mixing flux μ_{mix} in Equation 2 are computed 474 as residual terms in the mixing equation. To first verify that the mixing is always pos-475 itive, we compute the meridional profile of the eddy flux of age, F_{eddy} , was computed and 476 is shown in Figure 4(a) (in color). F_{eddy} is computed from the eddy covariance in isen-477 tropic coordinates, as in Equation 6. Negative and positive fluxes in the winter and sum-478 mer hemisphere respectively indicate an equatorward transport of age by midlatitude 479 eddies. In the winter hemisphere, the eddy age flux has a two-peaked structure in the 480 vertical, with the first maximum at 425 K in the lower stratosphere (due to synoptic-481 scale mixing) and the second maximum at around 1000 K in the upper stratosphere (due 482 to planetary-scale mixing). This structure mirrors the two-peaked mixing efficiency struc-483 ture obtained by Gupta et al. (2021) and shown in Figure 1(b). Substantially weaker eddy 484 flux is found in the 550-650 K region — the region where negative μ_{mix} is obtained us-485 ing Equation 2 — but the flux is always equatorward. Weak mixing in this region is con-486 sistent with past studies (e.g. Shuckburgh et al. (2001)) which employ other metrics to 487 quantify stratospheric mixing over a shorter time period. 488

The dashed black contours in Figure 4(a) show the corresponding eddy diffusivity, \mathcal{D}_{eff} (defined in Equation 7). Focusing on the winter hemisphere, large values of the eddy diffusivity around the turnaround latitude (solid black) indicates that most of the mixing is indeed localized around the turnaround latitude. This validates the use of the turnaround latitude to compute the midlatitude mixing flux. The diffusivity is substantially lower within the tropical pipe indicating minimal mixing of midlatitude air into the deep tropics (15°S to 15°N).

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The wintertime eddy diffusivity is overlaid on to the age-of-air profile on the 550 K isentrope using a thick orange highlighting in Figure 4(b), an isentrope height associated with negative mixing (Figure 1). The bold orange curve highlights the latitudes

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Figure 4. (a) Eddy flux of age, F_{eddy} (in color), and eddy diffusivity \mathcal{D}_{eff} (in dashed), for idealized model stratosphere. The solid black curve traces the region with net zero diabatic velocity. The eddy diffusivity has units of m² s⁻¹ and is shown at contour intervals of 0.75×10^6 m² s⁻¹. (b) Zonal mean age profile on the 550 K isentropic level for the idealized model (thin orange). The solid and dashed red line in the tropics and extratropics show the weighted ages Γ_u and Γ_d respectively, and the bold orange curve in the northern extratropics highlights the region with strong eddy diffusivity, i.e., 10% and higher of the maximum diffusivity at 550 K. The vertical grey bars show the turnaround latitude, demarcating the regions of upwelling and downwelling.

associated with strong eddy diffusivity, defined as values 10% or higher than the maximum diffusivity on a given level. Most of the mixing occurs between 30°N and 60°N, exchanging parcels with age over a range of 5.5 years to 6.8 years. This range is overestimated by the mixing theory which instead assumes that the tropical parcels with age $\Gamma_u = 4$ Yr (solid red bar) mix with the extratropical parcels with age $\Gamma_d = 6.8$ Yr (dashed red bars). These ages correspond to the latitudes 15°N and 60°N respectively. The age difference $\Delta\Gamma$ between the parcels being mixed between the upwelling and downwelling regions is overestimated in the assumptions leading to Equation 2, which causes the mixing flux, μ_{mix} , to be underestimated.

We next demonstrate that a better estimation of the age difference mixed across 508 the mixing barrier is indeed crucial for quantifying the mixing flux; especially in the mid-509 dle and lower stratosphere. We quantify the actual age range of parcels being mixed near 510 the partition boundary with the Γ - θ circulation, ψ (defined in Equation 4) was computed 511 for the dry dynamical core FV3 using daily samples over a 3300-day period. The circu-512 lation ψ at 450 K for FV3 is shown in Figure 5(a). Latitude and ages with a non-zero 513 circulation strength (in color) show the latitudes that exhibit tracer transport by mid-514 latitude eddies and the age of parcels being transported at those latitudes. In the north-515 ern hemisphere, a positive ψ signifies a poleward mass transport. Figure 5(a) shows the 516 predominantly bidirectional transport of mass in the midlatitudes in FV3. For age val-517 ues less than the mean isentropic age (solid black), a positive (teal colored) circulation 518 cell means that the eddies transport younger age poleward. Likewise, for ages older than 519 the mean age, a negative (brown colored) circulation cell means that the eddies trans-520 port older age equatorward. 521

The range of age values being mixed is the highest in the winter midlatitudes, con-522 sistent with the predominant wave-mixing over a large spatial scale. Moreover, the cir-523 culation peaks in the vicinity of the turnaround latitude. The circulation in the vortex 524 region is considerably weaker, on account of the vortex being isolated from the midlat-525 itudes. The age range of the streamfunction is markedly narrower in the subtropical lat-526 itudes, which are characterized by weak adiabatic mixing, virtually going to zero at the 527 equator. The distribution in the summer midlatitudes (not shown) is qualitatively sim-528 ilar to the winter midlatitudes, except that mixing at higher altitudes occurs over a nar-529 rower age range, i.e., over a narrower range of latitudes. 530

Focusing around the turnaround latitude in the winter hemisphere (dashed black vertical bars; $\approx 37.4^{\circ}$ N) — where the mean isentropic age is 5.2 Yr (solid black) — the poleward transport of mass (teal) carries parcels with ages between 3 to 5.2 Yr from the subtropics to higher latitudes. Likewise, the equatorward flux (brown) carries parcels with ages between 5.2 Yr to 6.5 Yr from higher latitudes into the subtropics. Nearly all of the circulation near the turnaround latitude is associated with carrying parcels with age in

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the 4-6 Yr range, implying that a majority of parcels being carried poleward have age 537 between 4 and 5.2 Yr, and the majority of parcels being carried equatorward have age 538 between 5.2 to 6 Yr. This is seen more clearly in Figure 5(b) which shows a cross-section 539 of ψ along the dashed black bar in Figure 5(a) at the turnaround latitude of 37.4°N (blue 540 curve); by fixing both θ and ϕ , ψ is now a function of age alone. The asymmetric cir-541 culation distribution strongly tapers off away from the mean isentropic age of 5.2 Yr in 542 either direction. It confirms that most of the mixing at this latitude occurs among parcels 543 with age not younger than 4 Yr but also not older than 6 Yr. 544

The average age transported from the subtropics to the midlatitudes (and vice versa) 545 can be computed by calculating the ψ -weighted mean age exclusively over the region of 546 positive (negative) Γ - θ circulation. Using the terminology introduced in Section 2, this 547 weighted age-of-air actually being mixed at 450 K represented as $\Gamma_{u,eff}$ (and $\Gamma_{d,eff}$) is 548 shown using the solid (dashed) green vertical bars in Figure 5(b). Here, $\Gamma_{u,eff} = 4.2$ Yr 549 and $\Gamma_{d,eff} = 5.6$ Yr. Thus, the effective age difference mixed across the barrier is $\Delta \Gamma_{eff} =$ 550 $\Gamma_{d,eff} - \Gamma_{u,eff} = 1.4$ Yr. This age difference is approximately half of the age difference 551 $\Delta\Gamma = 2.5$ Yr between the ages $\Gamma_u = 3$ yr (solid red) and $\Gamma_d = 5.5$ yr (dashed red) as-552 sumed while deriving Equation 2. This correction implies that the bulk mixing flux μ_{mix} 553 underestimates the actual exchange flux by about 50%. Accordingly, the perturbation 554 age $\delta \Gamma_u = \Gamma_{u,eff} - \Gamma_u = 1.2$ Yr, and $\delta \Gamma_d = \Gamma_d - \Gamma_{d,eff} = -0.1$ Yr are obtained. 555

We can use the effective mixing ages to quantify the range of mixing. Referring to the mean age in subplot (a), the inferred values for $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$ suggest that the mixing predominantly occurs between 25°N and 55°N, instead of 15°N and 55°N as the red bars would indicate. Mixing is localized near the mixing barrier and the deep tropics are fairly isolated from the mixing flux originating in the midlatitudes.

Figure 5(c) shows the Γ - θ circulation at the 550 K isentropic height. At this level, $\Gamma_{u,eff} = 5.1$ Yr and $\Gamma_{d,eff} = 6.3$ Yr, and thus $\Delta\Gamma_{eff} = 1.2$ Yr. With $\Gamma_u = 4.2$ Yr and $\Gamma_d =$ 6.6 Yr, $\Delta\Gamma = 2.4$ Yr. Similar to 450 K, at 500 K the corrected age difference $\Delta\Gamma_{eff}$ is approximately half of $\Delta\Gamma$, and perturbation age $\delta\Gamma_u = 0.9$ Yr and $\delta\Gamma_d = 0.3$ Yr are obtained.

The procedure was repeated for all the isentropic levels, i.e., the Γ - θ circulation was used to compute the distribution of ages being mixed at the turnaround latitude in the winter hemisphere. The distribution was used to estimate $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$, which were ⁵⁶⁹ ultimately used to obtain the perturbation ages $\delta\Gamma_u$ and $\delta\Gamma_d$ at each vertical level. This ⁵⁷⁰ allows us to calculate all the variables except μ_{mix}^T in the refined mixing equation (Equa-⁵⁷¹ tion 3), and ultimately compute μ_{mix}^T as a residual.

The structure of the "effective" upwelling and downwelling age-of-air being exchanged 572 is shown in Figure 6(a) (black curves). The analysis using the Γ - θ circulation reveals that 573 influence of meridional gradients to the mixing estimation is the strongest in the lower 574 stratosphere in the tropics and in the middle stratosphere in the extratropics. The weighted 575 age Γ_u (solid orange) and the effective mixing age $\Gamma_{u,eff}$ (solid black) in Figure 6(a) dif-576 fer the most between 400 K and 600 K, and thus $\delta\Gamma_u$ peaks in this interval. The cor-577 responding midlatitude ages (dashed curves), however, differ the most between 500 K 578 to 900 K, and so $\delta \Gamma_d$ peaks in this interval. 579

Our analysis also shows prominent differences between the original age difference 580 $\Delta\Gamma$ and the effective mixing age difference $\Delta\Gamma_{eff}$ (Figure 6(b)). Throughout the lower 581 and middle stratosphere, a significant reduction in the age difference is observed, indi-582 cating that the difference in the mean poleward and the mean equatorward moving age 583 is indeed much lower than the assumed mixing age difference $\Delta\Gamma$. It should be noted that 584 the two measures are found to be almost indistinguishable in the upper stratosphere, but 585 elsewhere, the true age difference being mixed is overestimated by at least 50%. Note 586 that this has no implications for the diabatic circulation strength, which is still directly 587 related to $\Delta\Gamma$ (Linz et al., 2016). 588

Finally, we consider the new "entrainment freshening" term (C4) in the refined mix-589 ing equation and assess its contribution to the net aging in the tropics. The quantity, 590 shown in Figure 6(d), is consistent with a peak in $\delta\Gamma_u$, and contributes most to the ag-591 ing in the lower stratosphere. Note the negative sign in front of C4 in Equation 3, mean-592 ing that a positive C4 leads to a reduction in aging. The key is that the air being ex-593 pelled from the tropics is older than the mass flux weighted age Γ_{μ} : entrainment "fresh-594 ens" the pipe by ejection of older air. The term is positive even around 600 K, where 595 very weak (and even negative mixing) was estimated for the FV3 core (Figure 1). The 596 positive value of the term C4 in this region can explain the spurious "negative" mixing 597 observed in the bulk flux. With a lower stratospheric peak of 0.013 yr/K, this entrain-598 ment freshening term significantly reduces the overall aging. Comparing this term with 599 the net vertical aging in Figure 3(c) and the advection term in Figure 3(d) reveals that 600

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the entrainment freshening term C4 contributes a substantial fraction to the net aging in the tropics. In fact, in the 400-500 K range, the magnitude of this term is more than half as large as the net aging (comparing the green curve in Figure 6(d) and the teal curve in Figure 3(c)), reaffirming our hypothesis.

We now compare the "bulk" mixing fluxes obtained from Equation 2 with the "true" 605 mixing fluxes obtained from Equation 3. To do this, the mixing efficiency ϵ , which mea-606 sures the strength of the mixing flux relative to the net poleward flux, is used: $\epsilon = \mu_{mix}/\mu_{net}$. 607 Using the refined mixing equation to estimate the mixing fluxes leads to a striking en-608 hancement in the mixing efficiency in FV3 (Figure 7(c)). A positive age perturbation 609 $\delta \Gamma_u$ in the region leads to a positive offset in the mixing flux. Moreover, a markedly lower 610 age difference $\Delta \Gamma_{eff} < \Delta \Gamma$ results in a multiplied (magnified) mixing flux μ_{mix}^T . The 611 entrainment freshening term (C4) ensures a non-negative mixing flux throughout the strato-612 sphere, while the adjustment to $\Delta \Gamma_{eff}$ increases the amplitude of the flux. The origi-613 nal mixing equation yielded an erroneously weaker ϵ (thin dashed red) in the lower strato-614 sphere than the ϵ computed from the refined mixing equation (thick solid red). As will 615 be discussed in more detail later, the revised mixing efficiency (and thus the mixing flux) 616 bears a much stronger resemblance to the mixing efficiency obtained from the compre-617 hensive model WACCM. 618

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4.2 Estimating the meridional extent of adiabatic mixing

We now address the question that naturally follows: What then is the region over 620 which the air is truly mixed in the winter stratosphere? The estimates $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$ 621 of the true ages of the parcels mixed across the turnaround latitude can be mapped to 622 the latitudes with the corresponding mean age Γ . That is, we find the latitudes $\phi_1(\theta)$ 623 and $\phi_2(\theta)$ such that the zonal mean age $\Gamma(\phi_1(\theta), \theta) = \Gamma_{u,eff}(\theta)$ and $\Gamma(\phi_2(\theta), \theta) = \Gamma_{d,eff}(\theta)$. 624 The two latitudes obtained by the mapping provide the equatorward and poleward ex-625 tent of the midlatitude mixing. The results for FV3 from this procedure are shown in 626 Figure 8(a) (orange curves and shading). Throughout much of the stratosphere, the re-627 gion of mixing (orange shading) roughly spans from 20° N to 60° N. The most notable fea-628 ture obtained is that the mixing region is shifted poleward in the 400-500 K interval. In 629 addition, above 600 K, a steady but small widening of the mixing region is noted. 630

Comparing this with the region deduced from gross upwelling ages Γ_u and Γ_d (dashed 631 black curves and shading), we find that the most prominent differences appear in the 400-632 700 K region, where the blue curve in the subtropics aligns more strongly with the sharp 633 subtropical age gradient while the dashed black curve does not. This is readily observed 634 in the 450-500 K height range. Between 400-700 K, the Linz et al. (2021) theory assumes 635 the mixing to dominate over a wider region in the subtropics. However, the opposite is 636 suggested above 700 K. Here, Linz et al. (2021) theory assumes mixing to be restricted 637 to poleward of 25° N, presenting a contrast to a boundary of 20° N proposed by the re-638 fined theory. 639

The poleward boundary of mixing, however, is not as sensitive to the definition used. 640 In the lower stratosphere, the solid blue and dashed black curves from the two defini-641 tions tend to be nearly identical. Some small but notable differences emerge only above 642 500 K where the corrected ages suggest that the mixing is restricted to relatively lower 643 latitudes than that expected by the original Linz et al. (2021) theory. Even though both 644 estimates suggest that the mixing regions are not significantly different, it is surprising 645 how much impact even such small differences can make to the mixing flux estimates in 646 the lower stratosphere; purely on account of strong sub-tropical gradients in the age-of-647 air profile. 648

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4.3 Insights from the WACCM Comprehensive Climate Model

The analysis was repeated for WACCM, which provides a more Earth-like representation of stratospheric variability and seasonal cycle. The northern winter months of November to March were considered for a period of five years and daily samples were used to compute the age-of-air and the Γ - θ circulation. The more limited data, coupled with more internal variability, leads to more sampling uncertainty in these calculations.

We first emphasize that the zonal mean age-of-air in WACCM (shown later in Figure 8(b)) is much younger than in FV3 both on account of a faster circulation and a higher tropopause, and because the age in WACCM is computed w.r.t. the tropical stratosphere, i.e., the whole troposphere is the source. In FV3 the age-of-air is computed w.r.t the source region of 700 hPa and below, due to which the age at the tropical stratosphere has a mean value of approx. 1 year. This offset does not affect the results in this study, as the differences in age are dominated by the differences in circulation. The Γ - θ circulation in WACCM at the 450 K isentropic height is shown in Figure 5(d). Comparing subplots (a) and (d), the gross structure of ψ is quite similar between the idealized dry dynamical core and WACCM. The circulation peaks in the midlatitudes and is weaker elsewhere. Moreover, for latitudes with considerable circulation strength, parcels with age younger than the mean age (solid black) are carried poleward and parcels with age older than the mean age are carried equatorward. Little to no circulation is noted in the polar regions on this isentropic surface.

The cross section of ψ at 450 K along the turnaround latitude of 32.6°N in the win-669 ter hemisphere (dashed black bar in Figure 5(d)) is shown in Figure 5(e). Yet again, we 670 find that the gross structure of ψ is very similar between WACCM and FV3 (Figure 5(b)). 671 The Γ - θ circulation is informative of the broad range of ages of parcels being mixed around 672 the barrier. At 450 K, parcels as young as 0.5 Yr are mixed with parcels with age up to 673 2.5 Yr. A considerable portion of the mixing, however, occurs between parcels with age 674 $\Gamma_{u,eff} = 0.9$ Yr and $\Gamma_{d,eff} = 2$ Yr (solid and dashed green respectively). Computations 675 yield $\delta \Gamma_u$ =-0.05 Yr and $\delta \Gamma_d$ =0.75 Yr. 676

The most conspicuous difference between the mixing in FV3 and WACCM on 450 677 K is that in FV3, Γ_u and $\Gamma_{u,eff}$ are quite different, while Γ_d and $\Gamma_{d,eff}$ are almost iden-678 tical (Figure 5(b)). In contrast, for WACCM, it is the Γ_u and $\Gamma_{u,eff}$ which are almost 679 identical and the downwelling ages Γ_d and $\Gamma_{d,eff}$ which are quite different. So, the (rel-680 atively weaker) subtropical gradients barely impact the estimation of the tropical age 681 being mixed. Nevertheless, the Γ - θ circulation in both the models suggest that the true 682 age difference mixed around the turnaround latitude is much lower than the bulk age 683 difference $\Delta\Gamma$, with most of the difference coming from the downwelling ages. 684

⁶⁶⁵ Next, the Γ - θ circulation at 550K is shown in Figure 5(f). At this level, parcels with ⁶⁶⁶ stratospheric age as young as 2 Yr are mixed with parcels with age as old as 4 Yr. How-⁶⁸⁷ ever, the bulk of the mixing occurs between ages $\Gamma_{u,eff} = 2.4$ Yr and $\Gamma_{d,eff} = 3.4$ Yr, ⁶⁸⁸ yielding an effective mixed age difference $\Delta\Gamma_{eff} = 1$ Yr, much less than a $\Delta\Gamma = 1.4$ Yr. ⁶⁸⁹ It follows that $\delta\Gamma_u \approx 0$ and $\delta\Gamma_d = 0.4$ Yr. Similar to 450 K, a weak perturbation age ⁶⁹⁰ $\delta\Gamma_u$ on 550 K is obtained for WACCM.

Similar to FV3, the wintertime averages of all the terms in the refined mixing equation were computed for WACCM. Here, we only show the effective age difference $\Delta\Gamma_{eff}$ (Figure 6(c)) and the entrainment freshening term C4 for WACCM (violet curve in Fig-

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⁶⁹⁴ ure 6(d)); terms which are associated with the revised mixing equation. Our estimate ⁶⁹⁵ for $\Delta\Gamma_{eff}$ using the Γ - θ circulation shows a considerably smaller age difference being mixed ⁶⁹⁶ across the turnaround latitude; Therefore, for both FV3 and WACCM, $\Delta\Gamma_{eff} << \Delta\Gamma$.

The entrainment freshening term for WACCM (violet curve in 6(d)) is weaker and 697 unlike FV3, barely contributes to the net vertical aging in the lower stratosphere. This 698 is due to the fact $\Gamma_u \approx \Gamma_{u,eff}$ in WACCM; the air being entrained into the extratrop-699 ics carries the mean age with it. In fact, the quantity is nearly vanishing throughout the 700 stratosphere, indicating that entrainment freshening does not have a strong effect on the 701 mixing flux estimation in WACCM. Since C4 is weakly negative in the lower stratosphere, 702 a smaller mixing term (C3) leads to a slight reduction in the "true" mixing flux estimated 703 from Equation 3. We believe, however, that the entrainment mixing term is not suffi-704 ciently sampled due to limited data availability for WACCM. The $\Gamma - \theta$ circulation is 705 sensitive to the sampling frequency and sample size, more so than the other dynamical 706 quantities in the revised mixing equation. Sampling the streamfunction at a frequency 707 higher than the timescales associated with isentropic mixing is crucial to obtaining a smooth 708 streamfunction profile. Since the entrainment freshening term is weak and not always 709 physically consistent, we neglect this term and only consider the effective age difference 710 while computing the revised mixing efficiency for WACCM. 711

The figure shows the bulk and true mixing efficiency computed for WACCM. Both 712 the bulk and true mixing efficiency in WACCM (in dashed and bold gray respectively) 713 have similar gross structure throughout the stratosphere. The two mixing profiles dif-714 fer in that the true mixing is more enhanced in the lower stratosphere. $\Delta\Gamma_{eff}$ which is 715 smaller than $\Delta\Gamma$ results in enhancement of the mixing fluxes in lower and middle strato-716 sphere. A stronger mixing in WACCM is also consistent with a stronger wintertime cir-717 culation in WACCM. The bold grey curve suggests a mixing efficiency of 2 in the lower 718 stratosphere. This is equivalent to the true mixing flux μ_{mix} being two-thirds the strength 719 of the equatorward mass flux. 720

The same procedure as described in Section 4.2 was applied to WACCM in order to estimate the latitudinal span of wave-induced mixing. Figure 8(b) shows the mixing span estimated for WACCM using both $\Gamma_{u,eff}$, $\Gamma_{d,eff}$ (orange curves and shading) and Γ_{u} , Γ_{d} (dashed black curves and shading). The two types of ages yield regions which are different in structure. The region determined from corrected ages spans from 20°N to

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 45° N in the lowermost stratosphere (400K) and gradually decreases in width up to 700 726 K where it only spans from 15° N to 35° N. Most notable is the equatorward shift in the 727 orange curve in the subtropics, in phase with the turnaround latitude (solid black). Above 728 700 K, the region re-widens, spanning all the way from the equator on the left to 60° N 729 on the right. We note that above 1100 K the corrected tropical age $\Gamma_{u,eff}$ is younger than 730 the equatorial age and thus the mixing span has been truncated at the equator. Such 731 young ages are most likely obtained due to (i) rapid equatorward shift of the turnaround 732 latitude with height, (ii) transience in the zonal mean age at lower latitudes in WACCM, 733 and (iii) prevailing inter-hemispheric transport. 734

In contrast, the mixing span determined from Γ_u, Γ_d spans from 20°N on the equa-735 torward side to 70°N on the poleward side. This range is considerably wider than that 736 for FV3 (black shading in subplot (a)). Interestingly, for FV3, both the dashed black curves 737 lay on either side of the turnaround latitude. This is not the case for WACCM, where 738 the turnaround latitude makes a drastic equatorward turn. Since the corrected ages were 739 inferred by analyzing the Γ - θ circulation around the turnaround latitude, differences in 740 the diabatic velocity structure between FV3 and WACCM lead to major differences ob-741 tained between the orange and black shaded regions as well in WACCM. Despite these 742 differences, it is found that the deep tropics barely witness any wave-induced adiabatic 743 mixing and are fairly isolated, just as in FV3. 744

In summary, the structure of the Γ - θ circulation is similar for FV3 and WACCM, 745 and both models demonstrate an overestimation of the mixed age difference in the orig-746 inal mixing equation. Employing the refined mixing theory, which estimates the age dif-747 ference more accurately, yields similar mixing efficiency ϵ for the two models. Limited 748 sampling in WACCM may also contribute slightly to the observed differences between 749 FV3 and WACCM. Only 5 years of daily data, 150 days per year, was considered from 750 WACCM. This is significantly lower than 10 years of data, 360 days per year, consid-751 ered for the dynamical core. Moreover, the original mixing equation of Linz et al. (2021) 752 assumes a stratosphere in steady state, and for WACCM the NDJFM climatology is cou-753 pled with the effects of dynamics and transport during other seasons. The choice of per-754 petual solstice climatology in FV3 ensures that contributions from inter-seasonal vari-755 ability are eliminated in the analysis and the circulation is statistically steady. Some dif-756 ferences in mixing may also be due to the presence of a nudged QBO in WACCM vs per-757 sistent tropical easterlies in FV3. 758

⁷⁵⁹ **5** Discussion and Conclusions

Wave-induced adiabatic mixing plays a key role in mixing trace gases over large 760 spatial scales in the winter stratosphere, and in influencing their global distribution. Ac-761 curately quantifying this mixing is key to understanding transport trends in the strato-762 sphere in a changing climate. We have proposed a method to improve the estimates of 763 the adiabatic mixing flux in the stratosphere obtained from applying the vertical age gra-764 dient theory of Linz et al. (2021), and have used it to quantify the extent of mixing be-765 tween the tropics and midlatitudes. We show that the deep tropics do not participate 766 in the adiabatic mixing of tracers in most of the stratosphere. 767

The theoretical formulation of Linz et al. (2021) partitions the stratosphere into 768 regions of diabatic upwelling and diabatic downwelling. This allowed them to connect 769 the vertical gradient of tracer (age-of-air) in the upwelling region (tropics) with the quasi-770 horizontal adiabatic mixing flux between the two partitions, thus allowing computation 771 of the mixing flux using tracer-based measures. The theory assumes fast mixing within 772 both the tropics and the extratropics, and hence neglects any gradients in age-of-air pro-773 file expected near the subtropical transport barrier. We show that this assumption leads 774 to an overestimation of the meridional span of adiabatic mixing in the winter stratosphere, 775 and as a result, an underestimation of the mixing fluxes. We refined the framework in 776 order to include the effects of meridional age gradients. This allows a more comprehen-777 sive estimation of the age of the air parcels exchanged between the upwelling and down-778 welling partitions of the stratosphere. We used the refined age mixing theory to quan-779 tify the meridional span of adiabatic mixing in the winter stratosphere. 780

The correction was made possible by a novel strategy to obtain the complete age 781 range of air parcels mixed around the turnaround latitude through the Γ - θ circulation 782 streamfunction. The streamfunction is essentially a joint distribution which allows quan-783 tification of mass transport in the latitude-age-isentrope space; i.e., at a given latitude 784 and isentropic height, what fraction of the total mass transport is associated with a given 785 age? Having information of the age-of-air distribution being mixed enabled estimation 786 of the "true" ages that are mixed around the winter hemisphere turnaround latitude. We 787 calculated the true difference in the age-of-air being transported from upwelling to the 788 downwelling region and vice versa, and found that this effective age difference is signif-789 icantly smaller than the age difference assumed in the Linz et al. (2021) theory. 790

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The true ages of air being mixed was used to re-derive the mixing equation of Linz et al. (2021) (Equation 2) to yield Equation 3, which accounts for age gradients as well. The new mixing equation replaces the gross age differences $\Delta\Gamma$ of the mixed parcels with the revised age difference calculated from the Γ - θ circulation. It also contains an additional advection term (the entrainment freshening) that captures the effects of entrainment on the tropical age in the presence of subtropical gradients.

For a thorough test of the proposed theory, we considered both an idealized model, 797 a dry dynamical core and a comprehensive model. Both of these choices were made to 798 maximize the error that will be introduced into the original mixing theory, as averag-799 ing over several years of seasonally-varying circulation (as in Linz et al. (2021)) will damp 800 the effects highlighted here. Both the models exhibit similar Γ - θ circulation, qualitatively, 801 in the lower stratosphere. Further, results from both models indeed verify an overesti-802 mation of the age difference $\Delta\Gamma$ mixed between the upwelling and downwelling regions 803 by the original mixing equation. On applying the revised mixing theory to the models, 804 it was found that both the idealized and the comprehensive model qualitatively produced 805 very similar mixing efficiencies (a proxy for mixing fluxes). 806

The models show a strong agreement throughout the stratosphere. In fact, we highlight that both models also agree that the deep tropics, from the equator all the way to 15°N, is fairly isolated from adiabatic mixing and that the equatorial region almost exclusively experiences pure diabatic advection of air. This is especially true in the lower and the middle stratosphere. Our findings corroborate the findings from an independent study by the co-authors Curbelo and Linz, who used Langrangian trajectories to quantify the mixing of midlatitude air into the tropics.

We conclude that our analysis highlights both the sensitivity of the mixing flux to 814 the strong age gradients near the turnaround latitude, and their importance in the es-815 timation of the quasi-horizontal mixing flux. It also demonstrates that tracer-based mea-816 sures can be used to estimate the meridional span of adiabatic mixing in the winter mid-817 latitudes given sufficiently detailed data. The theory proposed in this study can be used 818 to quantify mixing in the observed stratosphere as well, using both satellite observations 819 and reanalyses, especially in the lower and middle stratosphere. Past studies have used 820 transport models and specified dynamics to compute and compare the mean age-of-air 821 across different reanalyses (Chabrillat et al., 2018; Ploeger et al., 2019). Our analysis could 822

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⁸²³ be further used to connect the impact of differences in adiabatic mixing to the observed
⁸²⁴ age differences, and over a longer timescale than those considered in this study, using
⁸²⁵ more than forty years of reanalyses.

Projecting the mass transport into latitude-age-isentrope space using the Γ - θ cir-826 culation presents a new way to assess the adiabatic mixing structure in the stratosphere. 827 The streamfunction couples the dynamics and transport into a single quantity, and in-828 stead of age-of-air tracer, it could be generalized to any chemical trace gas species with 829 weak sources and sinks in the stratosphere: nitrous oxide, sulphur hexafluoride and methane, 830 for example. The circulation distribution can be of value in investigating transport ex-831 changes across the wintertime polar vortex, or mixing in the upper troposphere lower 832 stratosphere (UTLS) region where the combined effects of stratosphere-troposphere mass 833 exchange, convection and jet instability, influence the ozone and water vapor reservoirs. 834

Appendix A Revised Mixing Equation

The age-of-air is an idealized tracer with no spatial sources and sinks away from the boundary. The age tracer only has a source in time, i.e., the age of a parcel in the free atmosphere ages at a rate of 1 second per second. Therefore, for a stratosphere in steady state, the tracer continuity equation for the age-of-air, following Linz et al. (2016), can be generally written as :

$$\frac{1}{\rho_{\theta}} \nabla \cdot \mathcal{F} = 1 \tag{A1}$$

where $\rho_{\theta} = \frac{-1}{g} |dp/d\theta|$ is the three-dimensional isentropic density, and \mathcal{F} is the net advectivediffusive mass flux of age. Considering a stratospheric air mass enclosed between two isentropes θ and over $\theta + \Delta \theta$ in the vertical and spanning the whole upwelling partition in the horizontal, the net age budget for the box (or layer) can be, most simply, written as :

Age Flux
$$Out = Age Flux In + Source$$
 (A2)

Mathematically, this is equivalent to multiplying Equation A1 by ρ_{θ} , and horizontally integrating both sides over the upwelling region and over the isentropes θ and θ + $\Delta\theta$ in the vertical. Assuming the contribution from diffusion to be small, all the fluxes take an advective form. Adopting the terminology introduced in Linz et al. (2021) and in Section 2, the total age flux entering the box is the sum of the upward directed diabatic flux of age entering the box from below (i.e. the θ isentrope) and the adiabatic advective flux of older age $\Gamma_d - \delta \Gamma_d$ by the mixing flux μ_{mix} from the downwelling region. Similarly, the total age flux exiting the box is the sum of the upward directed distance directed di-

abatic flux of age exiting the box from the top (i.e. the $\theta + \Delta \theta$ isentrope) and the pole-

- ward advective flux of $\Gamma_u + \delta \Gamma_u$ by the entrainment μ_{ent} into the downwelling region.
- Both μ_{mix} and μ_{ent} have units of kg/s/K (mass flux per unit isentropic height). The tracer
- continuity equation for the infinitesimal layer can thus be written as :

$$\Gamma_{u}\mathcal{M}_{u}(\theta+\Delta\theta) + \int_{\theta}^{\theta+\Delta\theta} \mu_{ent}(\Gamma_{u}+\delta\Gamma_{u}) \ d\theta = \int_{\theta}^{\theta+\Delta\theta} \mu_{mix}(\Gamma_{d}-\delta\Gamma_{d}) \ d\theta + \Gamma_{u}\mathcal{M}_{u}(\theta) + \int_{\theta}^{\theta+\Delta\theta} \rho_{\theta}d\theta$$
(A3)

We rearrange and express $\Gamma_u \mathcal{M}_u(\theta + \Delta \theta) - \Gamma_u \mathcal{M}_u(\theta)$ as $\Delta \theta \cdot d(\Gamma_u \mathcal{M}_u)/d\theta$ and apply the product rule to get the left hand side of Equation A4. In addition, we approximate the integrals by multiplying the integrand by $\Delta \theta$. Cancelling $\Delta \theta$ from each side, we get :

$$\Gamma_u \frac{d\mathcal{M}_u}{d\theta} + \mathcal{M}_u \frac{d\Gamma_u}{d\theta} = -\mu_{ent}(\Gamma_u + \delta\Gamma_u) + \mu_{mix}(\Gamma_d - \delta\Gamma_d) + \sigma_u \tag{A4}$$

where σ_u is the isentropic density horizontally integrated over the upwelling region (units kg/K). By mass continuity, $\mu_{net} = -\frac{d\mathcal{M}_u}{d\theta}$, and by definition, $\mu_{net} = \mu_{ent} - \mu_{mix}$. On substitution and rearranging, we get :

$$\frac{d\Gamma_u}{d\theta} = \frac{\sigma_u}{\mathcal{M}_u} + \frac{\mu_{mix}(\Delta\Gamma_d - \delta\Gamma_d - \delta\Gamma_u)}{\mathcal{M}_u} - \frac{\mu_{net}\delta\Gamma_u}{\mathcal{M}_u}$$
(A5)

For a steady state circulation, the upwelling mixing flux balances the downwelling mixing flux and thus, $\mathcal{M}_u = -\mathcal{M}_d = \mathcal{M}$. On substitution, this yields the revised mixing equation A6, identical to Equation 3 :

$$\frac{d\Gamma_u}{d\theta} = \frac{\sigma_u}{\mathcal{M}} + \mu_{mix} \frac{\Delta\Gamma_{eff}}{\mathcal{M}} - \mu_{net} \frac{\delta\Gamma_u}{\mathcal{M}}$$
(A6)

868 Acknowledgments

We thank Hella Garny and Thomas Birner for insightful suggestions. WACCM is a com-869 ponent of the Community Earth System Model (CESM), which is supported by the Na-870 tional Science Foundation (NSF) and the Office of Science of the U.S. Department of En-871 ergy. Computing resources were provided by NYU High Performance Computing, and 872 NCAR's Climate Simulation Laboratory, sponsored by NSF and other agencies. This re-873 search was enabled by the computational and storage resources of NCAR's Computa-874 tional and Information System Laboratory (CISL). We acknowledge support of the US 875 National Science Foundation through grant AGS-1852727 to New York University. M. 876 Linz acknowledges support from NASA New Investigator Program Award 80NSSC21K0943. 877 J. Curbelo also acknowledges the support of the U.S. NSF Grant AGS-1832842 and the 878 RyC project RYC2018-025169. Data availability: FV3 model data available on NYU high 879 performance computing cluster Greene and WACCM model data available on NCAR's 880 supercomputing storage. Zonal mean model data, in a polished form, is being prepared 881 and will be made pubicly available for peer review latest by 15 September 2022. 882

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Figure 5. The Γ-θ circulation, ψ , as defined in Equation 4 for (a) the FV3 core and (d) the WACCM model, at 450 K isentropic height. In both (a) and (b), the black curves shows the mean age-of-air at 450 K, and the solid and dashed red lines respectively mark the upwelling and downwelling ages, Γ_u and Γ_d at this height. The dashed black curves in each hemisphere mark the turnaround latitudes at 450 K. Subplots (b) and (e) show a slice of ψ at 450 K in bold blue across a fixed latitude (chosen near the turnaround latitude for FV3 and WACCM respectively, i.e., they show a cross-section of ψ along the dashed black curve in (a) and (d)). Subplots (c) and (f) are identical to subplots (b) and (e) except that they show the distribution on 550 K. In subplots (b-c,e-f), the solid and dashed red vertical bars show Γ_u and Γ_d respectively, while the solid and dashed green vertical bars show the ages $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$ which are calculated from the Γ-θ circulation by a circulation-weighted average over the positive and negative portion of ψ (in blue). The ages $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$ more accurately reflect the ages of the parcels actually being mixed at the given height and indicate an overestimation of the age difference $\Delta\Gamma$ being mixed across the transport barrier, as considered in Equation 2.



Figure 6. A comparison of the terms between the original mixing equation, Equation 2, and the revised mixing equation, Equation 3. (a) The mass-flux weighted ages Γ_u and Γ_d (in orange) and the ψ -weighted ages $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$ (in black) for the idealized FV3 model. The upwelling and downwelling ages are shown using solid and dashed curves respectively. (b) The age differences $\Delta\Gamma$ (thin green) and $\Delta\Gamma_{eff}$ (bold green) as inferred from the ages in subplot (a) for FV3. (c) Corresponding age differences $\Delta\Gamma$ (thin blue) and $\Delta\Gamma_{eff}$ (bold blue) obtained for NDJFM months in WACCM. (d) The entrainment freshening term, i.e., term C4 in Equation 3 for both the idealized FV3 model (yellow) and the comprehensive model WACCM (violet).



Figure 7. The bulk mixing efficiency and true mixing efficiency ϵ , which are defined as the ratio of the bulk/true mixing flux to the net poleward flux, i.e., bulk $\epsilon = \mu_{mix}/\mu_{net}$ and true $\epsilon = \mu_{mix}^T/\mu_{net}$. The mixing efficiency obtained by applying Equation 3 is shown using thick solid curves. The mixing efficiency obtained for both FV3 (red) and WACCM (grey) using the original mixing equation of Linz et al. (2021) is shown using thin dashed curves for reference.



Figure 8. Age inferred meridional range of adiabatic mixing in (a) the idealized FV3 core and (b) the comprehensive model WACCM. For both the plots, the zonal mean age profile is shown in color. The solid black curve shows the turnaround latitude, and the dashed black curves on either side show the latitudes with Γ_u and Γ_d as the mean age. The orange curves, and the area enclosed, demarcate the extent of the mixing region inferred from the mean ages $\Gamma_{u,eff}$ and $\Gamma_{d,eff}$. These ages were estimated as an average of the circulation distribution around the turnaround latitude. The analysis reveals little-to-no mixing of the midlatitude air into the deep tropics. The deep tropics are markedly more isolated for the FV3 core, than for the WACCM model. Both the subplots use separate colorbars.