

# Stratospheric Transit Time Distributions Derived from Satellite Water Vapor Measurements

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## Abstract

Stratospheric transit time distributions (age-of-air spectra) are estimated using satellite water vapor (H<sub>2</sub>O) measurements from the Microwave Limb Sounder over 2004-2021 assuming stationary transport. Latitude-altitude dependent spectra are derived from correlations of interannual H<sub>2</sub>O anomalies with respect to the tropical tropopause source region, fitted with an inverse Gaussian distribution function. The reconstructions accurately capture interannual H<sub>2</sub>O variability in the ‘tropical pipe’ and global lower stratosphere, regions of relatively fast transport (~1-2 years) in the Brewer-Dobson circulation. The calculations provide novel observational estimates of the corresponding ‘short transit-time’ part of the age spectrum in these regions, including the mode. However, the H<sub>2</sub>O results do not constrain the longer transit-time ‘tail’ of the age spectra, and the mean age of air and spectral widths are systematically underestimated compared to other data. We compare observational results with parallel calculations applied to the WACCM chemistry-climate model and the CLaMS chemistry-transport model, and additionally evaluate the method in CLaMS by comparing with spectra from idealized pulse tracers. Because the age spectra accurately capture H<sub>2</sub>O interannual variations originating from the tropical tropopause, they can be used to identify ‘other’ sources of variability in the lower stratosphere, and we use these calculations to quantify H<sub>2</sub>O anomalies in the Southern Hemisphere linked to the Australian New Years fires in early 2020 and the Hunga volcanic eruption in 2022.

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1  
2 **Stratospheric Transit Time Distributions Derived from Satellite Water Vapor**  
3 **Measurements**

4  
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11  
12 **Key Points:**

- 13 • Stratospheric transit time distributions (age spectra) are derived from time series of  
14 satellite water vapor measurements
- 15 • Water vapor reconstructions from age spectra accurately capture variability in the tropics  
16 up to 30 km and in the global lower stratosphere
- 17 • Age spectrum results are compared between observations and global model simulations  
18

## 19 **Abstract**

20 Stratospheric transit time distributions (age-of-air spectra) are estimated using satellite water  
21 vapor ( $\text{H}_2\text{O}$ ) measurements from the Microwave Limb Sounder over 2004-2021 assuming  
22 stationary transport. Latitude-altitude dependent spectra are derived from correlations of  
23 interannual  $\text{H}_2\text{O}$  anomalies with respect to the tropical tropopause source region, fitted with an  
24 inverse Gaussian distribution function. The reconstructions accurately capture interannual  $\text{H}_2\text{O}$   
25 variability in the ‘tropical pipe’ and global lower stratosphere, regions of relatively fast transport  
26 ( $\sim 1$ -2 years) in the Brewer-Dobson circulation. The calculations provide novel observational  
27 estimates of the corresponding ‘short transit-time’ part of the age spectrum in these regions,  
28 including the mode. However, the  $\text{H}_2\text{O}$  results do not constrain the longer transit-time ‘tail’ of  
29 the age spectra, and the mean age of air and spectral widths are systematically underestimated  
30 compared to other data. We compare observational results with parallel calculations applied to  
31 the WACCM chemistry-climate model and the CLaMS chemistry-transport model, and  
32 additionally evaluate the method in CLaMS by comparing with spectra from idealized pulse  
33 tracers. Because the age spectra accurately capture  $\text{H}_2\text{O}$  interannual variations originating from  
34 the tropical tropopause, they can be used to identify ‘other’ sources of variability in the lower  
35 stratosphere, and we use these calculations to quantify  $\text{H}_2\text{O}$  anomalies in the Southern  
36 Hemisphere linked to the Australian New Years fires in early 2020 and the Hunga volcanic  
37 eruption in 2022.

38

## 39 **Plain Language Summary**

40 Stratospheric water vapor ( $\text{H}_2\text{O}$ ) is mainly controlled by transport across the cold tropical  
41 tropopause, which sets the entry value for the global stratosphere. Interannual variations in  $\text{H}_2\text{O}$   
42 originate near the tropical tropopause and then propagate throughout the stratosphere with the  
43 global Brewer-Dobson circulation (BDC), where anomalies are lagged in time and smoothed by  
44 mixing. We use the observed time series of  $\text{H}_2\text{O}$  from the Microwave Limb Sounder during  
45 2004-2021 to calculate transit time distributions (also called age spectra) from the tropical  
46 tropopause source region, based on fits to an idealized inverse Gaussian distribution. The results  
47 accurately capture  $\text{H}_2\text{O}$  variability in the tropics up to 30 km and in the global lower  
48 stratosphere, regions of relatively fast transport ( $\sim 1$ -2 years) in the BDC. The calculations  
49 provide novel observational estimates of the short transit time part of the age spectra in these  
50 regions, although the longer transit time part of the spectra are poorly constrained. The  
51 calculations are straightforward to apply to global models, and we compare observational results  
52 with simulations from the WACCM and CLaMS models.

53

## 54 **1 Introduction**

55 The stratospheric age spectrum is the probability distribution of transit times from the  
56 stratospheric entry point (i.e., the tropical tropopause) to any location, and thus links the  
57 boundary sources of transported constituents with their global distribution (Kida, 1983; Hall and  
58 Plumb, 1994). It provides a powerful diagnostic of the effective stratospheric circulation and is

59 especially useful for evaluating model transport behavior (e.g., Hall et al, 1999; Waugh and Hall,  
60 2002). The age spectrum can be computed explicitly with a numerical model using pulse tracers  
61 (e.g., Hall and Plumb, 1994), and recent work has focused on evaluating the detailed seasonal  
62 and interannual variability of model transport (Li et al, 2012, Ploeger and Birner, 2016; Ploeger  
63 et al, 2021). Model spectra have also been deduced using a set of chemically active trace species,  
64 either in idealized set-ups (Podglajen and Ploeger, 2019; Hauck et al, 2019) or observations  
65 (Schoeberl et al, 2005; Hauck et al, 2020). Stratospheric age spectra have also been derived from  
66 trajectory calculations using three-dimensional wind fields from meteorological analyses or  
67 reanalyses (Schoeberl et al, 2003; Diallo et al, 2012), and large differences in results demonstrate  
68 the uncertainty related to reanalyses (Ploeger et al, 2019), in particular their representation of  
69 heating rates.

70 A related quantity is the mean age of air (the first moment of the age spectrum) which  
71 can be exactly diagnosed using idealized linearly increasing conservative tracers (Hall and  
72 Plumb, 1994). While the mean age is a useful metric for comparison to observations, using for  
73 example measurements of carbon dioxide ( $\text{CO}_2$ ) and sulfur hexafluoride ( $\text{SF}_6$ ) (Waugh and Hall,  
74 2002), it provides much less information than the full age spectrum.

75 While the stratospheric age spectrum is a common diagnostic for models, it cannot be  
76 measured directly from observations. Rather, observational estimates have been made using  
77 conservative tracers such as carbon dioxide ( $\text{CO}_2$ ; Andrews et al, 1999; 2001) or combinations of  
78 tracers with varying photochemical lifetimes (Schoeberl et al, 2005; Hauck et al, 2020). These  
79 calculations assume an idealized functional form for the age spectrum (often taken to be an  
80 inverse Gaussian, as discussed below) and optimize a fit to the observed constituent  
81 measurements. Johnson et al (1999) used a combination of satellite and balloon-borne  
82 measurements of water vapor ( $\text{H}_2\text{O}$ ) and methane ( $\text{CH}_4$ ) to estimate age spectra, calculating the  
83 spectra by direct Fourier inversion and singular value decomposition. Our work revisits the  
84 estimation of stratospheric age spectra using the long-term (2004-2021) global measurements of  
85  $\text{H}_2\text{O}$  from the Aura Microwave Limb Sounder (MLS) instrument. We analyze time series of  
86 observations at the tropical tropopause source region and throughout the global stratosphere, and  
87 derive age spectra from optimal correlations using an idealized inverse Gaussian functional form.  
88 In addition to observational results we perform parallel analyses on chemistry-climate model  
89 simulations to test the methodology, inform the data analysis and evaluate the model behavior.

90 Because the age spectrum reconstruction can accurately capture H<sub>2</sub>O variability tied to the  
91 tropical tropopause, differences with observations can be used to identify additional sources of  
92 H<sub>2</sub>O. We demonstrate these calculations to quantify H<sub>2</sub>O input to the Southern Hemisphere  
93 lower stratosphere from the Australian New Years fires in early 2020 and the Hunga volcanic  
94 eruption in 2022.

95

## 96 **2. Observations and model simulations**

### 97 2.1 MLS data

98 We analyze water vapor measurements from the Aura MLS instrument (Read et al, 2007)  
99 covering the period September 2004 – March 2021, using retrieval version 5.1 (Livesey et al,  
100 2020). Data are available for standard pressure levels (12 per decade) for levels 316 to above 1  
101 hPa, with a vertical resolution of ~3 km. The precision of MLS H<sub>2</sub>O over 100-1 hPa is 5-7% for  
102 a single profile and 20 times smaller for monthly zonal means. The MLS v5 H<sub>2</sub>O retrievals have  
103 corrected a small positive drift found in previous v4 results (Livesey et al, 2021). The age spectra  
104 are calculated using data prior to the Hunga volcanic eruption in January 2022, which injected a  
105 large amount of H<sub>2</sub>O into the stratosphere (Millan et al, 2022), and our results are not influenced  
106 by that event. We include some updated MLS results through early 2024 in Section 4b. Our  
107 analyses are based on monthly and zonally averaged data on a 5° latitude grid, which have been  
108 deseasonalized with respect to the long-term monthly seasonal cycle. We use deseasonalized  
109 data because we are interested in quantifying the behavior of interannual variations in H<sub>2</sub>O, but  
110 we note that very similar age spectrum results are found if the seasonal cycle is included.

111

### 112 2.2 Global models

113 We perform parallel analyses deriving age spectra from global model simulations to both  
114 inform the observational data studies and evaluate model transport. We use output from the  
115 Whole Atmosphere Community Climate Model (WACCM), which is a comprehensive chemistry  
116 climate model that spans the altitude range from the surface to ~140 km; WACCM details are  
117 described at <https://www2.acom.ucar.edu/gcm/waccm>. The results here are derived from the  
118 recently updated version 6 (WACCM6) described in Gettelman et al (2019). Our analyses use

119 standard historical simulations from WACCM spanning the years 1960-2014, using horizontal  
120 resolution 2 x 2 degrees and 70 vertical levels (vertical resolution ~1 km in the stratosphere), and  
121 the quasi-biennial oscillation (QBO) in these runs was nudged to observations. We analyze  
122 deseasonalized monthly and zonal average fields of H<sub>2</sub>O, and we note that this model includes  
123 methane (CH<sub>4</sub>) oxidation effects on stratospheric H<sub>2</sub>O.

124 We also analyze results derived from the Chemical Lagrangian Model of the Stratosphere  
125 (CLaMS), which has been used extensively for stratospheric age-of-air calculations (Ploeger and  
126 Birner, 2016; Podglajen and Ploeger, 2019). The model is driven by ERA-interim (Dee et al.,  
127 2011) wind and heating rates and features a dedicated parameterisation of small-scale mixing  
128 (Konopka et al., 2004). CLaMS also includes CH<sub>4</sub> oxidation effects on stratospheric H<sub>2</sub>O. The  
129 model set-up used here is described by Pommrich et al. (2014) and includes additional pulse  
130 tracers (Ploeger and Birner, 2016) released at the tropical tropopause with a 2-month resolution  
131 along the transit-time axis extending up to 10 years.

### 132 **3 Results**

#### 133 *3.1 Deriving age spectra from time series of H<sub>2</sub>O*

134 Aura MLS provides a long-term (more than 17 years) global observational data record  
135 of stratospheric water vapor (H<sub>2</sub>O), which has been used extensively in empirical data studies  
136 and model evaluations (e.g. Kawatani et al, 2014; Diallo et al, 2018; Yu et al, 2022). It is well-  
137 known that H<sub>2</sub>O in the lower and middle stratosphere is mainly controlled by the tropical cold  
138 point temperature, while CH<sub>4</sub> oxidation contributes to (low frequency) H<sub>2</sub>O variations in regions  
139 of ‘older’ stratospheric air (e.g. Yu et al., 2022; Tao et al., 2023). The MLS data show that  
140 stratospheric H<sub>2</sub>O anomalies originate near the tropical tropopause and propagate coherently in  
141 latitude and height throughout the stratosphere, transported by the Brewer-Dobson circulation  
142 (Randel and Park, 2019). During such transport, the H<sub>2</sub>O anomalies are lagged in time and  
143 systematically reduced in amplitude compared to variations at the tropical tropopause source  
144 region, due to advection and mixing. This is the fundamental signature of the vertical  
145 propagation of H<sub>2</sub>O over the equator, the so-called tape recorder signal (Mote et al, 1996) found  
146 both in seasonal cycle and interannual H<sub>2</sub>O anomalies. This behavior in the deseasonalized MLS  
147 H<sub>2</sub>O data in the tropics is illustrated in Fig. 1a, which shows time series of anomalies at the  
148 tropopause source region (83 hPa), and at levels 46 hPa and 18 hPa, for data averaged over 10°

149 N-S. As a note, the 83 hPa H<sub>2</sub>O anomalies closely follow corresponding variations in cold point  
 150 tropopause temperature, with correlations close to 0.9 (Randel and Park, 2019). The H<sub>2</sub>O  
 151 anomalies at upper levels are lagged in time, smoothed and reduced in amplitude compared to  
 152 the variations at 83 hPa. Similar behavior is found for H<sub>2</sub>O anomalies that propagate  
 153 meridionally to the extratropical lower stratosphere in both hemispheres (Randel and Park,  
 154 2019).

155 As reviewed by Waugh and Hall (2002), the age spectrum mathematical problem is  
 156 defined in a straightforward manner as:

$$157 \quad X_B(t) = \int X_A(t - \tau) * G(\tau) d\tau \quad (1)$$

158  
 159 Here X<sub>A</sub>(t) represents the source function of H<sub>2</sub>O for air entering the stratosphere near the  
 160 tropical tropopause (83 hPa tropical H<sub>2</sub>O anomalies in this case, the lower level curve in Fig. 1b),  
 161 X<sub>B</sub>(t) is the H<sub>2</sub>O at some ‘distant’ location in the stratosphere, and G(τ) is the age-of-air  
 162 spectrum. We note that this expression is only valid for stationary transport, where G(τ) does not  
 163 depend on time. G(τ) is often characterized by an inverse Gaussian distribution of the form:

$$164 \quad G_{\Gamma,\Delta}(\tau) = \sqrt{\frac{\Gamma^3}{4\pi\Delta^2\tau^3}} \exp\left(\frac{-\Gamma(\tau-\Gamma)^2}{4\Delta^2\tau}\right) \quad (2)$$

165  
 166 with parameters Γ=mean age and Δ=spectral width (Waugh and Hall, 2002). The mode (most  
 167 likely value) is given by:

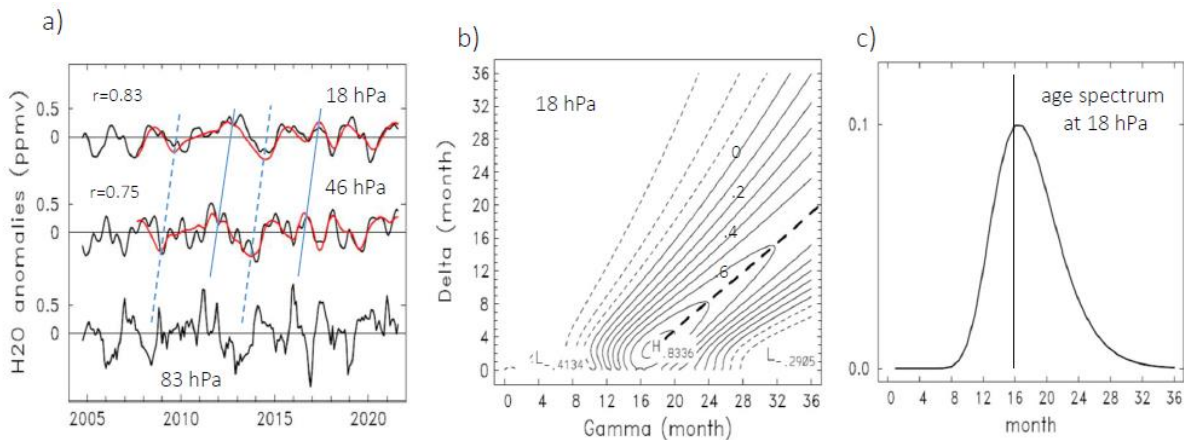
$$168 \quad M_{\Gamma,\Delta} = \Gamma \left[ \sqrt{\left(1 + 9\frac{\Delta^4}{\Gamma^4}\right)} - 3\frac{\Delta^2}{\Gamma^2} \right] \quad (3)$$

169  
 170 This G<sub>Γ,Δ</sub>(τ) is based on the solution to a 1-dimensional diffusive transport problem (Hall and  
 171 Plumb, 1994). Our calculations involve deriving empirical estimates of G<sub>Γ,Δ</sub>(τ) in Eqns. 1-2 by  
 172 finding optimal correlations between observed H<sub>2</sub>O time series and X<sub>B</sub>(t) derived from Eq. 1, to  
 173 obtain the best-fit age spectrum G<sub>Γ,Δ</sub>(τ). We note that our calculations provide a different and  
 174 complementary technique to the Fourier inversion or singular value decomposition calculations  
 175 used by Johnson et al (1999).

176 Our calculations use the monthly time series of MLS data over September 2004 – March  
 177 2021 (~16.5 years), and we derive the convolution in Eq. 1 using a G(τ) window of width 36

178 months, which reduces the length of  $X_B(t)$  to  $\sim 13.5$  years (September 2007-March 2021). The  
 179 choice of 36-month convolution width in Eq. 1 is made to balance resolution of  $G_{\Gamma,\Delta}(\tau)$  vs. the  
 180 length of the available data record, and hence focuses on the short time scale structure of the age  
 181 spectrum rather than on the tail of the distribution; this is an important detail discussed further  
 182 below. Tests using a longer window (e.g., 60 months) do not show significant differences from  
 183 the results shown here.

184 We demonstrate these calculations for the MLS  $H_2O$  time series at the equator and 18  
 185 hPa shown in Fig. 1a. Figure 1b shows the correlation between calculated  $X_B(t)$  and observed  
 186  $H_2O(t)$  as a function of the parameters  $\Gamma$  and  $\Delta$  in Eq. 2, and the results identify an absolute  
 187 maximum in the correlation for  $\Gamma=18$  months and  $\Delta=3$  months. Figure 1c shows this derived age  
 188 spectrum with a peak (modal) value of 16 months. The resulting time series of  $X_B(t)$  at 18 hPa is  
 189 shown as the red line in Fig. 1a, which has a correlation of 0.83 with the observed  $H_2O$ ;  
 190 corresponding results are also shown for the 46 hPa level in Fig. 1a. At all levels throughout the  
 191 tropics the  $H_2O$  correlations exhibit maximum values similar to Fig. 1b that are used to select  
 192 optimal  $\Gamma$  and  $\Delta$  fit parameters. However, we note that the correlation maxima in Fig. 1b are  
 193 somewhat broad in  $(\Gamma,\Delta)$  space with highest values maximizing along a line with constant mode  
 194  $M$  (heavy dashed line in Fig. 1b), and relatively strong  $H_2O$  correlations occur for very different  
 195  $(\Gamma,\Delta)$  combinations. We find that this is a general result of these parameter sweep calculations.  
 196 As a note, estimates of best fit  $\Gamma$  and  $\Delta$  based on minimum rms differences of (calculated  $X_B(t)$   
 197 minus observed  $H_2O(t)$ ) give very similar results to those for maximum correlation.



198

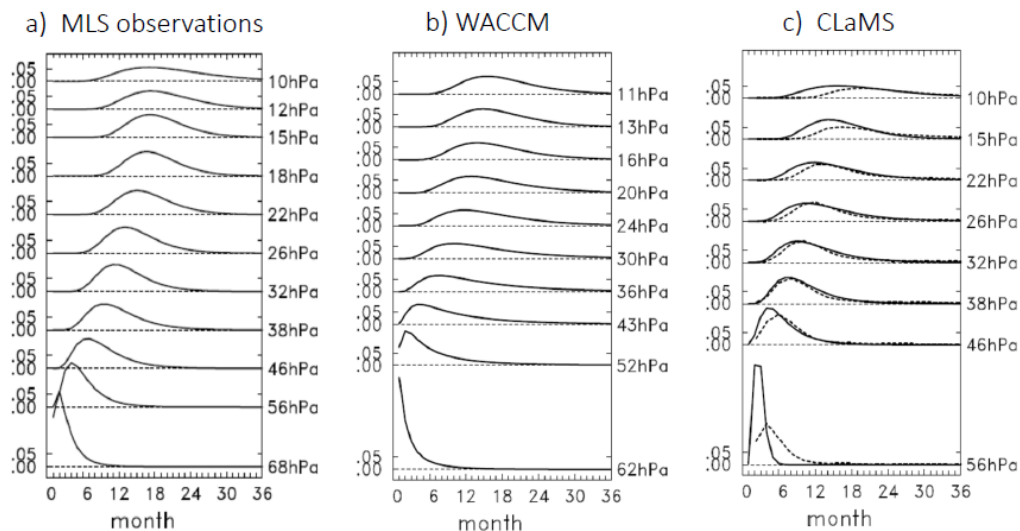
199 Figure 1. (a) Black lines show time series of tropical ( $10^\circ$  N-S) MLS  $H_2O$  deseasonalized  
 200 anomalies at 83 hPa (source region near tropical tropical tropopause), 46 and 18 hPa.



201 Red lines at 46 and 18 hPa show  $X_B(t)$  reconstructions based on convolving the 83 hPa  
 202 time series with derived age spectra. Solid and dashed blue lines trace several maxima  
 203 and minima in altitude. (b) Correlation of observed  $H_2O(t)$  at 18 hPa vs. the age  
 204 spectrum reconstruction  $X_B(t)$  as a function of the parameters ( $\Gamma$ ,  $\Delta$ ), with a maximum at  
 205 (18,3). The heavy dashed line corresponds to a constant mode  $M$  that crosses through  
 206 the maximum correlation point,  $M=16$  months in this case. (c) Resulting optimum age  
 207 spectrum at 18 hPa.

208

209 These calculations can be repeated at each latitude and height, and Fig. 2a shows the  
 210 resulting age spectra derived from MLS  $H_2O$  data in the tropics over 68-10 hPa (~19-32 km).  
 211 The modal (most likely) values trace the phase speed of vertical propagation in the  $H_2O$  tape  
 212 recorder, and this calculation provides novel quantification of this speed as a complement to  
 213 simple lag correlations e.g. Niwano et al, 2003; Glanville and Birner, 2017. The width of the age  
 214 spectra broadens with altitude in Fig. 2a, indicative of increased mixing of air between the  
 215 tropics and extratropics at higher levels.



216

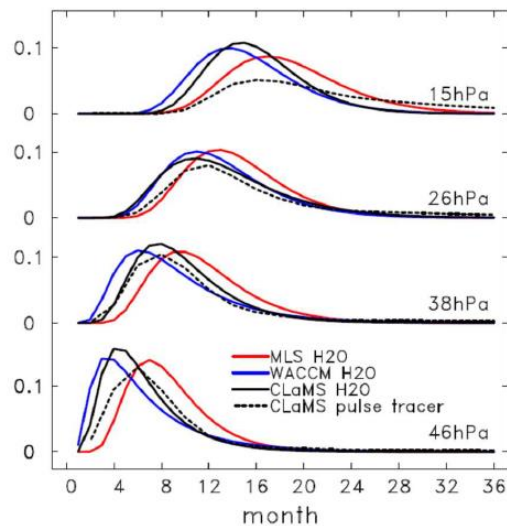
217 Figure 2. Tropical ( $10^\circ$  N-S) age spectra versus altitude (pressure level) derived from  $H_2O$  time  
 218 series, for (a) MLS observations, (b) WACCM and (c) CLaMS model. Dashed lines in  
 219 (c) show age spectra derived from the CLaMS pulse tracers.

220

### 221 3.2 Comparisons with WACCM and CLaMS model calculations

222 The calculations described above are easy to implement using standard monthly mean  
 223 output of chemistry-climate model simulations. For both the WACCM and CLaMS model

224 calculations, we identify a source time series of H<sub>2</sub>O near the model tropical tropopause, which  
 225 is at p=87 hPa in WACCM and p=82 hPa in CLaMS, and the rest of the calculations proceed as  
 226 above. Tropical age-of-air spectra derived from WACCM and CLaMS model simulations of H<sub>2</sub>O  
 227 are shown in Figs. 2b-c, showing approximate agreement with MLS results (Fig. 2a) in terms of  
 228 modal times and spectral width. This is consistent with the good performances of WACCM and  
 229 CLaMS models to simulate atmospheric water vapor variability (Gettelman et al, 2019; Yu et al.,  
 230 2022; Diallo et al., 2018; Kopopka et al., 2022; Tao et al., 2023). Direct comparison of the  
 231 calculated spectra at several levels in the tropics (10° N-S) are shown in Fig. 3. The comparisons  
 232 show approximate agreement in the shape of the spectra, and that inferred vertical propagation  
 233 times (spectral mode) are slightly faster in both models compared to MLS observations. These  
 234 WACCM6 results are consistent with the H<sub>2</sub>O tape recorder comparisons to MLS data shown in  
 235 Gettelman et al, 2019.



236

237 Figure 3. Comparison of tropical (10° N-S) age spectra derived from H<sub>2</sub>O time series at several  
 238 pressure levels, from MLS observations, WACCM and CLaMS models. Dashed black  
 239 lines show corresponding results from the CLaMS pulse tracers.

240

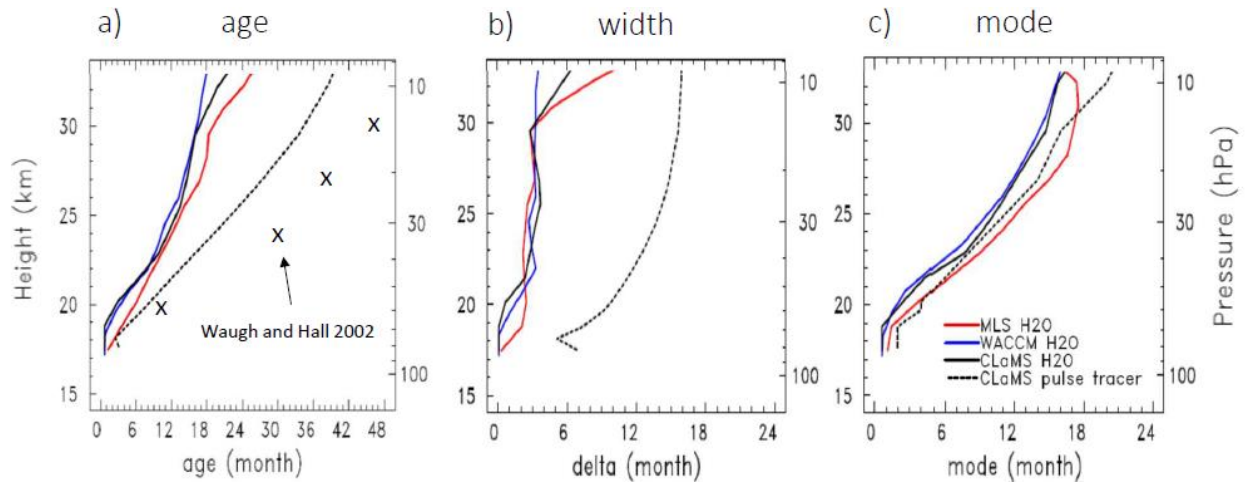
### 241 3.3 Comparing CLaMS spectra from H<sub>2</sub>O with results from pulse tracers

242 For the CLaMS simulations analyzed here we can directly compare the age-of-air  
 243 spectra derived from H<sub>2</sub>O with model calculations based on pulse tracers, as described in Ploeger  
 244 and Birner (2016). Figures 2c and 3 include direct comparisons of the tropical age spectra  
 245 between the two calculations, and Fig. 4 compares tropical vertical profiles of derived age, width

246 and mode. The spectra show reasonable agreement with the H<sub>2</sub>O fit results in the lower  
247 stratosphere (e.g. lower altitude or higher pressure levels in Fig. 3), including the mode estimate.  
248 Larger differences become evident at upper levels (15 and 10 hPa), where the pulse tracer  
249 calculations show spectra with larger components of aged air, i.e. a larger tail to the spectra.

250 Comparisons of the derived mean age and width from the pulse tracers show larger  
251 differences with the H<sub>2</sub>O estimates (Figs. 4a-b). The tropical H<sub>2</sub>O-derived ages from MLS,  
252 WACCM and CLaMS are similar in Fig. 4a, increasing with height to approximately 18-24  
253 months near 10 hPa. However, these estimates are substantially smaller than the CLaMS pulse  
254 tracer values and also only about half as large as the tropical age-of-air profile derived from CO<sub>2</sub>  
255 and SF<sub>6</sub> observations in Waugh and Hall (2002). This difference can be explained by noting that  
256 the age is the first moment of the age spectrum and is sensitive to the longer time scales (tail) of  
257 the distribution. The longer tail of the pulse tracer spectra compared to the H<sub>2</sub>O inverse Gaussian  
258 fits can be seen in Figs. 2-3. While the H<sub>2</sub>O variations in the tropical pipe are quite accurately fit  
259 by the assumed inverse Gaussian distribution (with correlations of order 0.8, as shown in Fig.  
260 1a), these fits mostly constrain the fast part of the spectrum (less than ~2 years) and less-so for  
261 the tail, which more strongly influences the age. In a similar manner, the spectral widths (second  
262 moment) of the pulse tracer spectra are much larger than the H<sub>2</sub>O inverse Gaussian fits (Fig. 4b)  
263 due to the influence of the tail. Hence while our choice of an inverse Gaussian distribution  
264 accurately reproduces the ‘fast’ transport of H<sub>2</sub>O from the tropical tropopause and gives accurate  
265 estimates of the mode (Fig. 4c), it provides a poor estimate for mean age and spectral width,  
266 which depend heavily on the tail of the spectrum.

267



268

269 Figure 4. Vertical profiles of tropical ( $10^{\circ}$  N-S) age spectrum parameters derived from the H<sub>2</sub>O  
 270 time series fits, for MLS observations, WACCM and CLaMS model results. The dashed  
 271 black lines show corresponding results from the CLaMS pulse tracers. The age results in  
 272 (a) also include values derived from CO<sub>2</sub> and SF<sub>6</sub> observations described in Waugh and  
 273 Hall (2002).

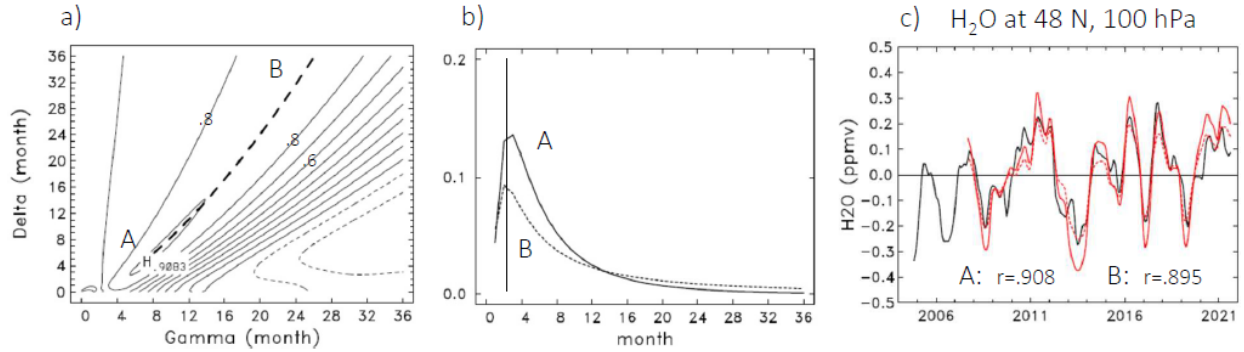
274

### 275 3.4 Transit time distribution in the extratropical lower stratosphere

276 In addition to coherent vertical propagation in the tropical pipe, the MLS H<sub>2</sub>O anomalies  
 277 exhibit coherent quasi-isentropic horizontal transport from the tropics to the extratropical lower  
 278 stratosphere of both hemispheres, with strong (lag) correlations ( $>0.7$ ) on the time scale of a few  
 279 months (e.g. Randel and Park, 2019). Our methodology can also be applied to deduce the age  
 280 spectra associated with this transport pathway. Results of these calculations show that high  
 281 H<sub>2</sub>O(t) vs. X<sub>B</sub>(t) correlations typically maximize along a broad ‘ridge’ in ( $\Gamma$ ,  $\Delta$ ) space consistent  
 282 with a constant mode, but without a strong correlation maximum as seen in the tropics (e.g. Fig.  
 283 1b). This behavior is illustrated in Fig. 5a, showing the age spectrum fit for H<sub>2</sub>O anomalies at 48°  
 284 N, 100hPa. Strong correlations are found along the ridge in ( $\Gamma$ ,  $\Delta$ ) space corresponding to a  
 285 constant mode  $M=2.5$  months (heavy dashed line in Fig. 5a), with relatively small differences in  
 286 the H<sub>2</sub>O correlations along this line. In this case the maximum correlation ( $r=.908$ ) is identified  
 287 at point A ( $\Gamma=7$ ,  $\Delta=5$ ) but nearly as high correlation ( $r=.895$ ) is found at point B ( $\Gamma=26$ ,  $\Delta=36$ ).  
 288 Hence the specific ( $\Gamma$ ,  $\Delta$ ) values and detailed shape of the spectra (Fig. 5b) are poorly constrained  
 289 by these calculations, although very high H<sub>2</sub>O anomaly correlations are derived in both cases

290 (Fig. 5c). This is typical behavior for age spectra fits in the extratropical lower stratosphere and  
 291 cautions against over-interpreting details of the derived spectra.

292



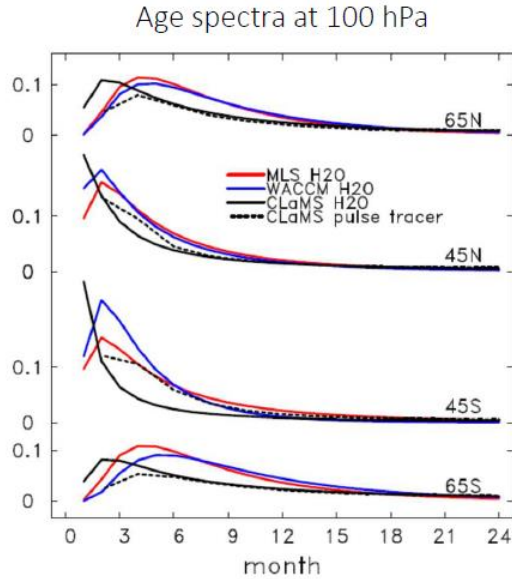
293

294 Figure 5. (a) Correlation of observed H<sub>2</sub>O anomalies at 48° N, 100 hPa with the age spectrum  
 295 reconstruction  $X_B(t)$  as a function of the parameters ( $\Gamma$ ,  $\Delta$ ). Relatively high correlations  
 296 are observed over the locus of points corresponding to a mode of 2.5 months, as noted  
 297 by the thick dotted line spanning A to B. (b) Age spectra corresponding to points A and  
 298 B. The mode for both spectra (2.5 months) are indicated by the vertical line. (c) Time  
 299 series of observed H<sub>2</sub>O anomalies (black) and  $X_B(t)$  reconstructions using the respective  
 300 A and B age spectra (solid and red dashed lines, respectively).

301

302 Figure 6 shown the calculated age spectra at 100 hPa for results at 45° and 65° N and S,  
 303 from MLS H<sub>2</sub>O along with the corresponding WACCM and CLaMS model results. The results  
 304 show broad age spectra peaking at time scales of ~2-4 months, with the spectra shifting to longer  
 305 modal times between 45° and 65° in both hemispheres. The WACCM calculations are in  
 306 reasonable agreement with MLS results, while CLaMS H<sub>2</sub>O spectra hint at systematically shorter  
 307 modal times, possibly suggesting faster quasi-horizontal transports in CLaMS.

308



309

310 Figure 6. 100 hPa age spectra derived from H<sub>2</sub>O time series at 45° and 65° N and S, showing  
 311 latitude dependent structure associated with quasi-horizontal transport from the tropics  
 312 to extratropics. Dashed black lines show corresponding results derived from the CLaMS  
 313 pulse tracers.

314

### 315 3.5 Global behavior

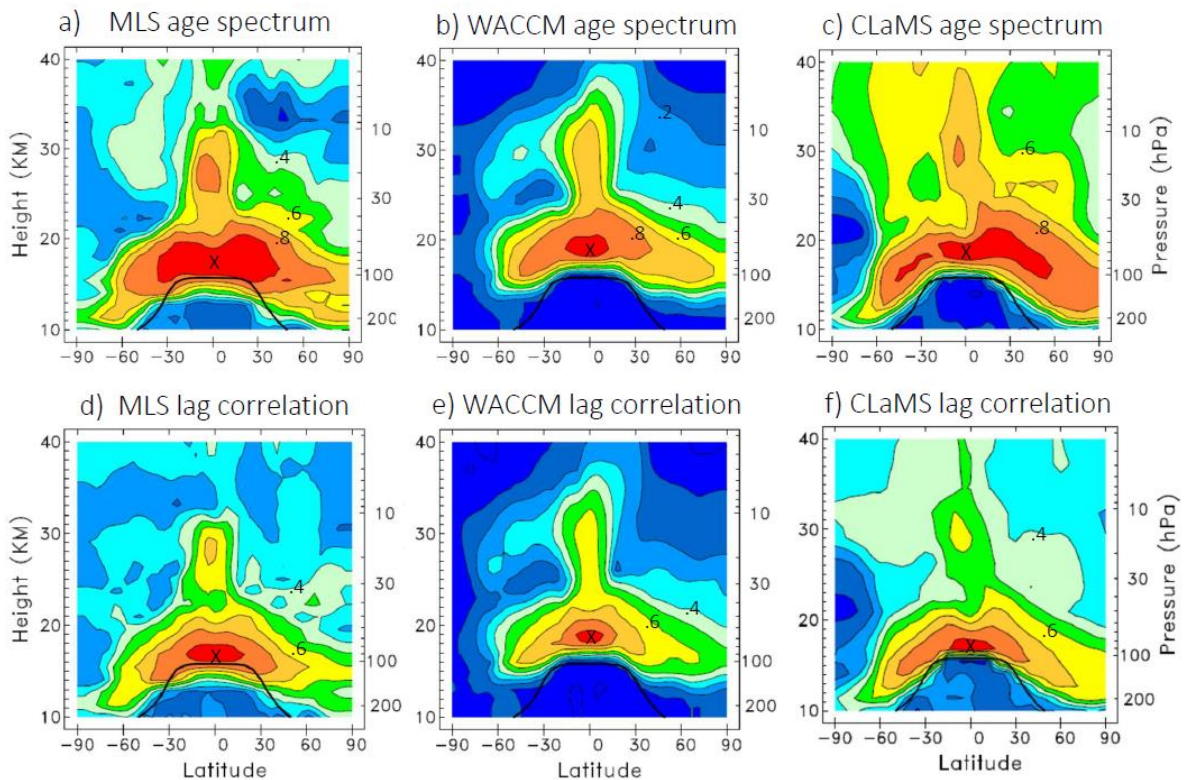
316 One measure of the goodness-of-fit of  $G_{T,\Delta}(\tau)$  is how well the calculated  $X_B(t)$  agrees  
 317 with observed H<sub>2</sub>O(t). Figure 7a shows this correlation as a function of latitude and height  
 318 derived from the MLS H<sub>2</sub>O data, showing high correlations (>0.7) extending upwards in the  
 319 tropics to ~32 km and throughout the global lower stratosphere. These are the regions of strong  
 320 interannual variability in H<sub>2</sub>O controlled by relatively rapid transport from the tropical  
 321 tropopause region. Note that strong lower stratosphere correlations extend all the way to the pole  
 322 in the NH in Fig. 7a, while they only extend to ~60° S in the SH. This is because local  
 323 dehydration in the Antarctic polar vortex in winter effectively decouples polar H<sub>2</sub>O from the  
 324 tropical tropopause. The high correlations in Fig. 7a demonstrate accurate fits of the observed  
 325 interannual variability using age spectra derived from the MLS H<sub>2</sub>O measurements. In regions of  
 326 low correlations, e.g. the extratropical middle stratosphere and above 10 hPa, the H<sub>2</sub>O data alone  
 327 are not as useful for constraining the transit time distribution.

328 Parallel results derived from WACCM and CLaMS H<sub>2</sub>O fields are shown in Fig. 7b-c.  
 329 WACCM results are similar to MLS statistics (Fig. 7a) both in terms of magnitude and spatial

330 patterns. The correlations from WACCM are smoother compared to the observational results  
 331 because we use a much longer time series from the model (~50 years) to evaluate the fits  
 332 (compared to 16.5 years of data for MLS). The CLaMS age spectrum correlations (Fig. 7c) show  
 333 patterns similar to MLS and WACCM, but the high correlations in the tropical pipe show weaker  
 334 latitudinal gradients extending into middle latitudes, i.e. the tropical pipe appears less isolated in  
 335 CLaMS compared to MLS and WACCM. This could be consistent with the faster meridional  
 336 propagation time scales for CLaMS discussed above, or perhaps with a weaker  
 337 subsidence/overturning circulation in midlatitudes.

338 For comparison, Figs. 7d-f show H<sub>2</sub>O anomaly correlation patterns using simple lag  
 339 regressions with respect to the tropical tropopause source region (picking the optimal time lag at  
 340 each latitude and height). Results show patterns like the age spectra correlations (Figs. 7a-c), but  
 341 the age spectrum systematically enhances the strongest correlations by ~0.1 to 0.2.

342



343

344 Figure 7. Correlations of observed H<sub>2</sub>O time series versus reconstruction from derived age  
 345 spectra for (a) MLS observations, (b) WACCM model and (c) CLaMS model. The ‘X’s  
 346 denote the corresponding source regions at the tropical tropopause. Panels (d-f) show the  
 347 corresponding results for simple lag correlations with respect to the tropical tropopause.

348

349 **4. Discussion**350 *4.1 Effects of CH<sub>4</sub> oxidation*

351 CH<sub>4</sub> oxidation contributes to low-frequency H<sub>2</sub>O variability in regions of ‘older’  
352 stratospheric air, i.e., the extratropics of the middle stratosphere and in the tropics above ~30 km  
353 (Yu et al., 2023; Tao et al, 2023). However, CH<sub>4</sub> oxidation contributes little to H<sub>2</sub>O fast  
354 variability in the tropical pipe and global lower stratosphere, although presumably the in-mixing  
355 of aged air (Neu and Plumb, 1999) is partly responsible for the reduction in H<sub>2</sub>O correlations  
356 with altitude in the tropical pipe (and lack of correlations above ~30 km). Tests with WACCM of  
357 parallel calculations using the conserved quantity (H<sub>2</sub>O + 2\*CH<sub>4</sub>) show only marginal increases  
358 in correlation and almost no changes in derived spectra compared to H<sub>2</sub>O alone.

359 *4.2 Identifying H<sub>2</sub>O variability not linked to the tropical tropopause*

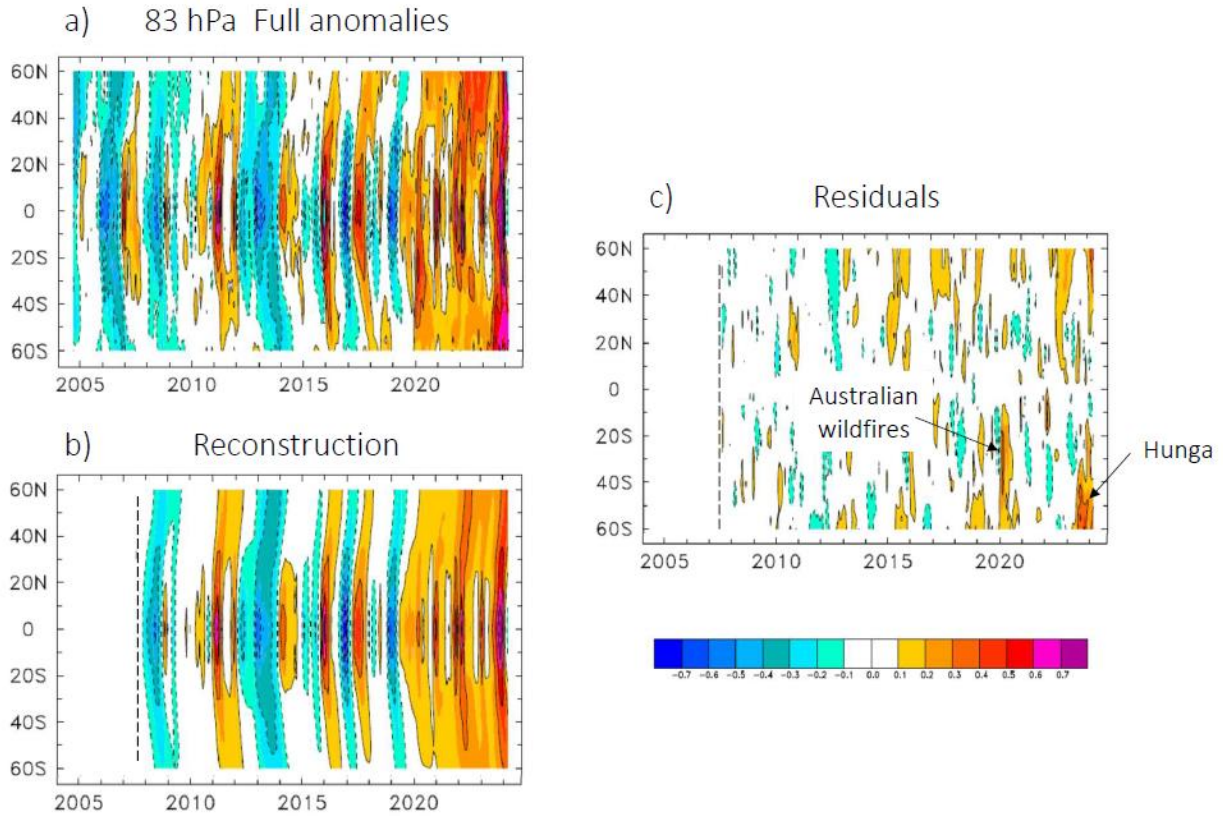
360 Because so much of the interannual variability of H<sub>2</sub>O in the lower stratosphere is  
361 captured by the age spectrum reconstruction (Fig. 7), it is interesting to examine the differences  
362 between observed vs. reconstructed anomalies to identify sources of variability not linked to  
363 transport from the tropical tropopause. Figure 8 compares the observed latitude vs. time  
364 evolution of MLS H<sub>2</sub>O anomalies at 83 hPa (Fig. 8a) with the age spectrum reconstruction (Fig.  
365 8b), and their differences (or residuals) shown in Fig. 8c. Here we have included MLS  
366 observations through March 2024, although the reconstructions are based on age spectra  
367 calculated with data prior to 2022. The H<sub>2</sub>O anomalies in Fig. 8a show episodic maxima near the  
368 equator that propagate to extratropics, and there are persistent positive anomalies after 2020  
369 linked to warm tropopause temperatures. As expected, the reconstruction (Fig. 8b) captures most  
370 of the H<sub>2</sub>O variability and the residuals (Fig. 8c) are generally small (~0.1 ppmv). There are  
371 generally positive H<sub>2</sub>O residuals over extratropics for the latter half of the record, which might  
372 reflect effects of CH<sub>4</sub> increases (in aged air) over time. However, somewhat larger residuals are  
373 observed for several specific periods in Fig. 8c. A localized maximum is seen in the SH low-to-  
374 middle latitudes during the first half of 2020, and these are likely related to H<sub>2</sub>O injected into the  
375 lower stratosphere from the extreme Australian New Year (ANY) fires (Khaykin et al., 2020; Yu  
376 et al, 2021; Peterson et al, 2021; Friberg et al, 2023). Figure 9a shows a latitude vs. height cross  
377 section of the H<sub>2</sub>O residuals for February 2020 showing positive anomalies up to 0.3 ppmv



378 covering altitudes ~15-19 km over ~10-60° S, and this region approximately overlaps the  
379 observed aerosol enhancement from the ANY fires (Rieger et al, 2021). Similar patterns of H<sub>2</sub>O  
380 residuals are evident for the first 6 months of 2020, slowly declining in magnitude over time.  
381 While these are relatively small H<sub>2</sub>O amounts (~0.1 to 0.3 ppmv), their temporal and spatial  
382 structure is strongly suggestive of links to the ANY fires.

383           Large and persistent H<sub>2</sub>O residuals are also seen in Fig. 8c over SH high latitudes in late  
384 2023, which are likely related to downward transport of H<sub>2</sub>O from the Hunga volcanic eruption  
385 in early 2022 (Millan et al, 2022). While much of the Hunga H<sub>2</sub>O plume was transported into the  
386 tropics above 25 km and upwards in the Brewer-Dobson circulation (Zhou et al, 2024), the  
387 global stratospheric circulation is expected to result in long-term downward transport over high  
388 latitudes. Figure 9b shows a cross-section of H<sub>2</sub>O residuals in October 2023, highlighting large  
389 positive anomalies in middle and high latitudes of both hemispheres, reflecting the global  
390 transport of the plume nearly 2 years after the eruption. The SH high latitude maxima at lower  
391 levels in late 2023 may reflect the climatological enhanced downward transport in the lower  
392 stratosphere during SH winter and spring, and we note that there are positive H<sub>2</sub>O residuals at  
393 high latitude in the NH during winters 2022-23 and 2023-24 in Fig. 8c that could likewise reflect  
394 the seasonal downward transport of Hunga H<sub>2</sub>O anomalies. It is likely that these high latitude  
395 effects will continue and increase over time as the Hunga H<sub>2</sub>O plume is transported downwards  
396 in the global stratospheric circulation.

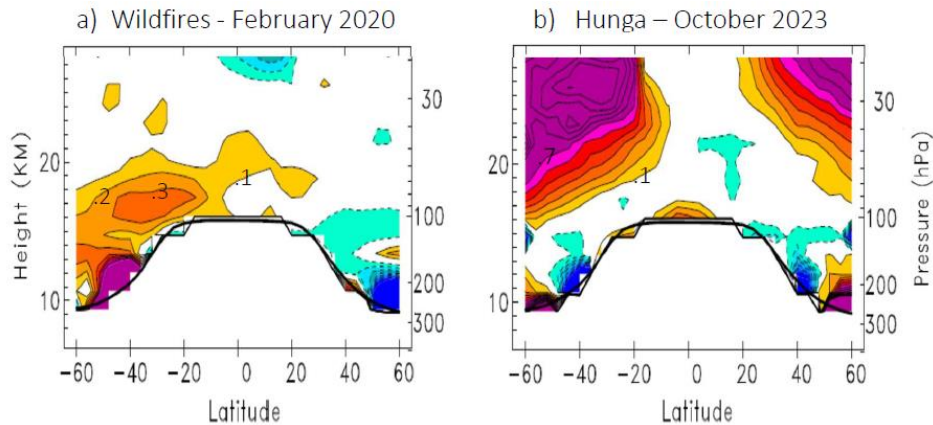
397



398

399 Figure 8. (a) Latitude vs. time section of deseasonalized MLS H<sub>2</sub>O anomalies at 83 hPa. (b)  
 400 Corresponding H<sub>2</sub>O anomalies based on age spectrum reconstruction covering the  
 401 period after September 2007. (c) Differences or residuals between observed and age  
 402 spectrum reconstruction.

403



404

405 Figure 9. Height vs. latitude structure of H<sub>2</sub>O residuals (ppmv) in (a) February 2020 and (b)  
 406 October 2023 derived from differences between observed H<sub>2</sub>O and age spectrum  
 407 reconstruction.

408

409 **5 Conclusions**

410 We have used observed interannual variations in H<sub>2</sub>O from MLS satellite observations  
411 to derive empirical stratospheric transit time distributions, based on optimal fits to an inverse  
412 Gaussian distribution function. These calculations are straightforward to apply to observational  
413 data or global model output. The results accurately capture H<sub>2</sub>O interannual variability in the  
414 tropical pipe and global lower stratosphere (with anomaly correlations > 0.8 in Fig. 7), which are  
415 regions of relatively fast transport (~1-2 years) in the Brewer-Dobson circulation. These  
416 calculations provide novel estimates of the corresponding ‘fast’ part of the transit time  
417 distribution in these regions, including the mode, and the age spectrum reconstructions capture  
418 H<sub>2</sub>O interannual variability more accurately than simple lag correlations. There is less  
419 information content for constraining the lower frequency tail of the age spectra, and the derived  
420 mean age and spectral width in the H<sub>2</sub>O calculations are too low compared to other observational  
421 data. Parallel H<sub>2</sub>O age spectra calculations from WACCM and CLaMS give similar results, and  
422 comparisons show faster upward transport in the tropics in the models compared to MLS data.  
423 Direct comparisons with spectra from CLaMS pulse tracers show reasonable agreement to the  
424 H<sub>2</sub>O calculations in the lower stratosphere, while there are larger differences in the tropics above  
425 ~25 km where the pulse tracers give flatter age spectra with a wider tail (c.f. Fig. 2c). This  
426 corresponds to more in-mixing of aged air at the top of the tropical pipe, which is not captured in  
427 the H<sub>2</sub>O results.

428 Because the age spectra reconstruction captures much of the H<sub>2</sub>O transport from the  
429 tropical tropopause in the lower stratosphere (outside of the Antarctic vortex), differences with  
430 observations can identify additional sources of H<sub>2</sub>O. We have demonstrated these calculations  
431 and identify anomalous H<sub>2</sub>O in the SH lower stratosphere in early 2020 that appears linked to the  
432 ANY fires, as suggested by space-time overlap with observed ANY aerosols. The associated  
433 ANY H<sub>2</sub>O anomalies are of order 0.1-0.3 ppmv (Fig. 9a) and persist for approximately 6 months.  
434 We additionally find persistent positive residuals over high SH latitudes in 2023 that are linked  
435 to the Hunga volcanic eruption in 2022. Quantification of these small anomalies is facilitated by  
436 the accurate fit of the background H<sub>2</sub>O variability (transport from the tropical tropopause) using  
437 the empirically derived transit time distributions.

438

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444

## 445 **Open Research**

446 MLS satellite data were obtained from the Goddard Earth Sciences Data and Information  
447 Services Center at [doi.org/10.5067/Aura/MLS/](https://doi.org/10.5067/Aura/MLS/) CESM2/WACCM6 is an open-source community  
448 model, which was developed with support primarily from the National Science Foundation. See  
449 Gettelman et al. (2019). The CLaMS code used for the simulations in this article is available on  
450 the GitLab server: <https://jugit.fz-juelich.de/clams/CLaMS> (last access: 14 May 2024). ERA-  
451 Interim reanalysis data are available from the European Centre for Medium-Range Weather  
452 Forecasts (via <https://apps.ecmwf.int/archive-catalogue/?class=ei>, last access: 15 May 2021).

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