

1 **The strength of the meridional overturning circulation of the stratosphere**

2 Marianna Linz\*

3 *Massachusetts Institute of Technology–Woods Hole Oceanographic Institution Joint Program in*  
4 *Physical Oceanography, Massachusetts Institute of Technology, Cambridge, MA, USA*

5 R. Alan Plumb

6 *Department of Earth, Atmospheric and Planetary Sciences, Massachusetts Institute of*  
7 *Technology, Cambridge, MA, USA*

8 Edwin P. Gerber

9 *Courant Institute of Mathematical Sciences, New York University, New York, NY, USA*

10 Florian J. Haenel

11 *Karlsruhe Institute of Technology, Institute of Meteorology and Climate Research, Karlsruhe,*  
12 *Germany*

13 Gabriele Stiller

14 *Karlsruhe Institute of Technology, Institute of Meteorology and Climate Research, Karlsruhe,*  
15 *Germany*

16 Douglas E. Kinnison

17 *Atmospheric Chemistry Observations and Modeling Laboratory, National Center for*  
18 *Atmospheric Research, Boulder, CO, USA*

19

Alison Ming

20

*Department of Applied Mathematics and Theoretical Physics, University of Cambridge,*

21

*Cambridge, UK*

22

Jessica L. Neu

23

*Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California, USA*

24 *\*Corresponding author address:* Marianna Linz, 54-1615, Massachusetts Institute of Technology,

25 Cambridge, MA 02139

26 E-mail: [mlinz@mit.edu](mailto:mlinz@mit.edu)

## ABSTRACT

27 The Brewer–Dobson circulation, the meridional overturning of mass in the  
28 stratosphere, is important for the distribution of gases in the stratosphere,  
29 such as ozone and water vapor, which impact surface climate. Previously, no  
30 observations-based estimate of its global strength existed. We present two  
31 such calculations of the mean strength of the meridional overturning of the  
32 stratosphere, quantified by the global diabatic circulation, between 2007–  
33 2011 from satellite data and compare these to three reanalyses and a state-  
34 of-the-art model. Using measurements of sulfur hexafluoride ( $\text{SF}_6$ ) and ni-  
35 trous oxide, we calculate the global mean diabatic overturning mass flux at  
36 all isentropic levels within the stratosphere. In the lower stratosphere, these  
37 two estimates agree, and at 460 K (about 20 km or 60 hPa in tropics), the  
38 global circulation strength is  $7.3 \pm 0.3 \times 10^9$  kg/s. In that region, the reanaly-  
39 ses broadly agree. Higher in the atmosphere, only the  $\text{SF}_6$  data-based estimate  
40 is available, and it diverges from the reanalyses and model. Interpretation of  
41 the  $\text{SF}_6$  data-based estimate is limited by the mesospheric sink of  $\text{SF}_6$ ; how-  
42 ever, the reanalyses also differ substantially from each other, implying 100%  
43 uncertainty in the mean meridional overturning circulation strength at upper  
44 levels.

45 Previous calculations of the strength of the stratospheric circulation from data have relied on  
46 indirect measures. Observational estimates of the strength of the overturning have been limited  
47 to qualitative descriptions based on tracer distributions (e.g. Stiller et al. 2012, Engel et al. 2009,  
48 Mahieu et al. 2014, Haenel et al. 2015) or quantitative measures of very limited regions, such as the  
49 vertical velocity over a narrow range in the tropics (Mote et al. 1996, Schoeberl et al. 2008, Flury  
50 et al. 2013). Free-running climate models vary widely in stratospheric circulation metrics, includ-  
51 ing the tropical upwelling mass flux at 10 hPa and 70 hPa, though the multimodel mean is relatively  
52 close to some reanalysis products (Butchart et al. 2011). Reanalyses, meanwhile, differ substan-  
53 tially in their mean tropical upwelling velocity, with the magnitude of the mismatch depending  
54 on how it is computed (Abalos et al. 2015). Here we consider the diabatic circulation of the  
55 stratosphere; because the stratosphere is stratified, vertical motion moves air across potential tem-  
56 perature surfaces and thus must be associated with warming/cooling in the ascending/descending  
57 branches. Hence the net meridional overturning of mass is tightly linked to diabatic processes.  
58 We use potential temperature as our vertical coordinate and the meridional overturning becomes  
59 explicitly the diabatic circulation in this framework.

60 In a generalization of the work by Neu and Plumb (1999), Linz et al. (2016) presented a theory  
61 to calculate the strength of the diabatic stratospheric circulation through each isentropic surface  
62 above the tropical tropopause from the idealized tracer “age of air” (Waugh and Hall 2002), which  
63 is a measure of how long a parcel of air has spent in the stratosphere. The difference between  
64 the age of the air that is upwelling and downwelling through an isentropic surface is inversely  
65 proportional to the strength of the diabatic circulation through that surface, in steady-state and  
66 neglecting diabatic diffusion.

67 In this paper, we apply the age difference theory to calculate the mean magnitude and vertical  
68 structure of the global overturning circulation of the stratosphere based on observations of sulfur

69 hexafluoride ( $\text{SF}_6$ ) and nitrous oxide ( $\text{N}_2\text{O}$ ). We demonstrate the validity of the theory and explore  
70 limitations of the tracer data with a coupled chemistry-climate model. We calculate the magnitude  
71 and vertical structure of the global overturning directly from the diabatic vertical velocity from  
72 three reanalyses to compare with the data and model results. Information on the data products,  
73 model, and reanalyses is given in Table 1.

## 74 **1. Age of air observations and model**

75 A trace gas that is linearly increasing in time in the troposphere and has no stratospheric sinks  
76 can be converted to age following the theory presented in Waugh and Hall (2002). Carbon diox-  
77 ide ( $\text{CO}_2$ ) and  $\text{SF}_6$  are both approximately linearly increasing in the troposphere and have minimal  
78 sinks in the stratosphere. We use age derived from sulfur hexafluoride ( $\text{SF}_6$ ) measurements (hence-  
79 forth  $\text{SF}_6$ -age) from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) on  
80 Envisat (Haenel et al. 2015). We interpolate  $\text{SF}_6$ -age onto isentropic surfaces using simultaneously  
81 retrieved pressure and temperature from MIPAS (von Clarmann et al. 2003, 2009). The resulting  
82  $\text{SF}_6$ -age on the 500 K surface is shown in Figure 1a. Age is young in the tropics, older in the  
83 extratropics, and oldest at the winter poles, consistent with the pattern of upwelling in the tropics  
84 and the majority of downwelling in the winter polar region. The  $\text{SF}_6$ -age at high latitudes in win-  
85 tertime is older than observations of age based on  $\text{CO}_2$  measurements (Plumb et al. 2002).  $\text{SF}_6$  is  
86 not conserved in the mesosphere, and its sink will result in a high bias in  $\text{SF}_6$ -age in areas with  
87 mesospheric influence (Hall and Waugh 1998), such as the poles and the upper stratosphere.

88 To explore the limitations of using  $\text{SF}_6$ -age, we compare  $\text{SF}_6$ -age to ideal age of air in a coupled  
89 chemistry-climate model, the Community Earth System Model 1 Whole Atmosphere Community  
90 Climate Model (WACCM). This is a fully coupled state-of-the-art interactive chemistry climate  
91 model (Marsh et al. 2013; Garcia et al. 2017). WACCM includes the physical parameterizations

92 and finite-volume dynamical core (Lin 2004) from the Community Atmosphere Model, version 4  
93 (Neale et al. 2013). The model domain extends from the Earth’s surface to the lower thermosphere  
94 (140 km). The WACCM simulations examined are based on the Chemistry Climate Model Initia-  
95 tive REF-C1 scenario (Morgenstern et al. 2017). WACCM models only one of the two sinks of  
96 SF<sub>6</sub> in the mesosphere; photolysis at Lyman-alpha wavelengths is included, but associative elec-  
97 tron attachment, which has recently been shown to be the dominant loss mechanism for SF<sub>6</sub> below  
98 105 km (Totterdill et al. 2015; Kovács et al. 2017), is not. The impact of the mesospheric sink  
99 of SF<sub>6</sub> on the stratospheric SF<sub>6</sub> will be determined by the strength of the dynamical coupling be-  
100 tween the stratosphere and the mesosphere. We calculate SF<sub>6</sub>-age following the methods of Stiller  
101 et al. (2012) to compare with the MIPAS SF<sub>6</sub>-age, for details see the Methods section. Although  
102 WACCM is missing the dominant SF<sub>6</sub> loss mechanism, the difference between SF<sub>6</sub>-age and ideal  
103 age will qualitatively illustrate the type and location of any bias introduced by using SF<sub>6</sub> as an age  
104 tracer.

105 Age on the 500 K surface between 2002 and 2012 is shown for WACCM SF<sub>6</sub>-age in Figure 1c,  
106 and for WACCM ideal age of air in Figure 1d. The close agreement between the ideal age and  
107 SF<sub>6</sub>-age on the 500 K surface suggests that SF<sub>6</sub>-age is a good proxy for ideal age. The temporal  
108 correlation at each latitude on the 500 K surface is high ( $r = 0.93$ ), and only at the poles is the  
109 SF<sub>6</sub>-age older than the ideal age by up to half a year. Where there is more mesospheric influence,  
110 the correlation is weaker and is no longer one-to-one: higher in the stratosphere and at the highest  
111 latitudes ( $r = 0.52$  and age has only 35% of the magnitude of variations of SF<sub>6</sub>-age at 1200 K at  
112 85° N). Since WACCM is missing the dominant sink of SF<sub>6</sub>, the differences shown here represent  
113 a lower bound on the bias induced by using SF<sub>6</sub>-age as a proxy for ideal age.

114 To corroborate the circulation strength calculations from SF<sub>6</sub>-age, other age tracers are desir-  
115 able. CO<sub>2</sub>, is currently not retrieved from satellites with enough accuracy and spatial coverage to

116 calculate age of air differences (Carlotti et al. 2016). Instead, we determine age from  $\text{N}_2\text{O}$ , which  
117 demonstrates a compact relationship with age, like other long-lived stratospheric tracers (Plumb  
118 and Ko 1992). We use the relationship between age of air and  $\text{N}_2\text{O}$  calculated empirically by  
119 Andrews et al. (2001), assuming that this compact relationship has not changed substantially in  
120 the interim while accounting for the linear growth in tropospheric  $\text{N}_2\text{O}$ . Following the procedure  
121 outlined in the Methods, we calculate age of air from the Global OZone Chemistry And Related  
122 trace gas Data records for the Stratosphere (GOZCARDS)  $\text{N}_2\text{O}$  data for 2004–2013 (Froidevaux  
123 et al. 2013). Because of the range of tracer values over which the empirical relationship holds,  
124 global coverage exists for a small range in potential temperature (about 450 K–500 K).

125 The age on the 500 K surface calculated from the empirical relationship of age with  $\text{N}_2\text{O}$  is  
126 shown in Figure 1b. The Southern Hemisphere winter polar coverage is poor on this level because  
127 values of  $\text{N}_2\text{O}$  are below 50 ppbv, the lower limit of the empirical fit. Age from the  $\text{N}_2\text{O}$  data is  
128 generally younger than MIPAS  $\text{SF}_6$ -age, though somewhat older than ages from WACCM. The  
129 temporal correlation of MIPAS  $\text{SF}_6$ -age and  $\text{N}_2\text{O}$ -age at every latitude on the 500 K surface is  
130 around  $r = 0.5$ , except in the Northern Hemisphere midlatitudes, where the correlation is not  
131 significant.

## 132 2. Age difference and the diabatic circulation

133 Linz et al. (2016) showed that, in steady state, the diabatic circulation ( $\mathcal{M}$ ) through an isentropic  
134 surface wholly within the stratosphere can be calculated as the ratio of the mass above the surface  
135 ( $M$ ) to the difference in the mass-flux-weighted age of downwelling and upwelling air on the  
136 surface ( $\Delta\Gamma$ , or age difference).

$$\mathcal{M} = M/\Delta\Gamma. \quad (1)$$

137  $\mathcal{M}$  is the total mass flux that is upwelling (or downwelling, as in steady-state these must be equal)  
138 through the isentropic surface. Intuitively this reflects the idea of a residence time; the age differ-  
139 ence is how long the air spent above the surface, and it is equal to the ratio of the mass above the  
140 surface to the mass flux passing through that surface.

141 The real world is not in steady-state, and so some amount of averaging is necessary for this  
142 theory to apply. The MIPAS data has five years of continuous data, and so the longest average  
143 possible for this study is five years. To test the validity of applying this steady-state theory to five-  
144 year averages of age difference, we have calculated the 2007–2011 averages of ideal age difference  
145 and the ratio of the total mass above each isentrope to the mass flux through that isentrope from  
146 WACCM output. These are shown in the blue lines (solid and dotted respectively) in Figure 2.  
147 The total overturning strength is calculated from the potential temperature tendency,  $\dot{\theta}$ , which is  
148 the total all sky radiative heating rate interpolated onto isentropic surfaces. The upwelling and  
149 downwelling regions are defined based on where  $\dot{\theta}$  is instantaneously positive or negative, and the  
150 mass fluxes through these regions are averaged to obtain the total overturning mass flux,  $\mathcal{M}$ . If  
151 the age difference theory held exactly, the two blue lines in Figure 2 would be identical. In the  
152 upper stratosphere, these two calculations agree closely; in the lower stratosphere, the ratio of the  
153 mass to the mass flux is greater than the ideal age  $\Delta\Gamma$ . This behavior is consistent with the neglect  
154 of diabatic diffusion, which is greater in the lower stratosphere (Sparling et al. 1997). Using area  
155 weighting of ideal age, since mass-flux weighting is not possible with data, results in about a 10%  
156 low bias of  $\Delta\Gamma$  compared to the mass-flux weighting shown here.

157 We calculate the five year average (2007–2011) of the difference in area-weighted age of air in  
158 the regions poleward and equatorward of  $35^\circ$  from the  $\text{SF}_6$ -age from both MIPAS and WACCM,  
159 and from the  $\text{N}_2\text{O}$ -age. The results of this are shown in Figure 2. The MIPAS  $\text{SF}_6$ -age  $\Delta\Gamma$  is  
160 notably different from the other estimates except around 450 K. At 400 K, it is much smaller, in

161 part because of young polar air at that level (not shown). Starting around 500 K, MIPAS SF<sub>6</sub> ΔΓ  
162 is much greater than the model ΔΓ using either ideal age or SF<sub>6</sub>-age. Age difference for N<sub>2</sub>O is  
163 calculated only where there is data available over the entire surface at almost all times, 450–480 K.  
164 In this limited range, the age difference from N<sub>2</sub>O-age is somewhat greater than the age difference  
165 from WACCM and agrees with the age difference calculated from MIPAS SF<sub>6</sub>-age.

166 To gain insight into the role of the mesospheric sink, we compare the ideal age ΔΓ with SF<sub>6</sub>-age  
167 ΔΓ in WACCM. The ideal age ΔΓ is the mass-flux-weighted age difference between upwelling  
168 and downwelling regions, and the SF<sub>6</sub>-age ΔΓ from WACCM is calculated in the same way as the  
169 MIPAS SF<sub>6</sub>-age ΔΓ. Because of the area-weighting, we expect the SF<sub>6</sub>-age ΔΓ to be 10% lower  
170 than the ideal age ΔΓ. This is true from 450–550 K, but above that, the SF<sub>6</sub>-age ΔΓ is either equal  
171 to or greater than the ideal age ΔΓ, and at 1200 K SF<sub>6</sub>-age ΔΓ is 50% greater. Since WACCM  
172 does not include the dominant sink of SF<sub>6</sub> for the mesosphere, the bias is certainly greater, and we  
173 cannot estimate an upper bound.

174 All three calculations of ΔΓ from the model as well as the ΔΓ from MIPAS SF<sub>6</sub>-age show a peak  
175 somewhere in the middle stratosphere. This peak indicates a relative minimum of the diabatic  
176 velocity at that level, and so this provides evidence that there are indeed two branches of the  
177 circulation (Birner and Bönisch 2011) and is a straightforward diagnostic for the separation level.

### 178 **3. Circulation from Reanalyses, Model, and Age**

179 Figure 3 shows the total overturning circulation strength calculated using the ratio of the total  
180 mass above the isentrope to ΔΓ for the MIPAS SF<sub>6</sub>-age and the N<sub>2</sub>O-age. Total mass is determined  
181 from the simultaneously retrieved pressure in the former case and from pressure from the Mod-  
182 ern Era Retrospective analysis for Research and Applications (MERRA, Rienecker et al. 2011)  
183 for N<sub>2</sub>O. Also shown is the directly calculated overturning circulation strength from the three

184 reanalysis products MERRA, Japanese 55-year Reanalysis (JRA 55, Kobayashi et al. 2015 and  
185 the ECMWF Reanalysis Interim (ERA-Interim, Dee et al. 2011), and from WACCM. The total  
186 overturning strength is calculated from the potential temperature tendency,  $\dot{\theta}$ , from the total di-  
187 abatic heating rates from JRA 55 and ERA-Interim forecast products and from total temperature  
188 tendency provided by MERRA, and then following the same procedure as above for WACCM.

189 These six estimates of the strength of the circulation are quite different, as can be seen clearly  
190 by examining the circulation at individual levels. At the lowermost levels, the reanalyses tend to  
191 agree, while the MIPAS SF<sub>6</sub>-age circulation estimate is much greater because of its very low  $\Delta\Gamma$ .  
192 In the range where we have estimates from both observational data sets, they agree closely and  
193 are flanked by the reanalyses, which vary more widely (within 35% of the mean of all estimates  
194 at that level). At 500 K and above, the MIPAS SF<sub>6</sub>-age based circulation strength has the lowest  
195 value, and at 900 K and above, it is lower by a factor of three. The circulation strength from  
196 MIPAS SF<sub>6</sub>-age  $\Delta\Gamma$  is biased low consistent with the sink of SF<sub>6</sub> in the mesosphere (Kovács et al.  
197 2017). The disagreement at 1200 K would require that the bias from using SF<sub>6</sub>-age  $\Delta\Gamma$  be nearly  
198 300% for the model and reanalyses to be correct. In addition to the disagreement of MIPAS SF<sub>6</sub>-  
199 age circulation strength with the model and reanalyses, there is significant disagreement between  
200 different reanalyses. MERRA has a distinct vertical structure, with weaker circulation in the lower  
201 stratosphere and stronger circulation in the mid stratosphere. JRA 55 and ERA Interim have a  
202 similar vertical structure; JRA 55 is stronger by around  $3 \times 10^9$  kg s<sup>-1</sup>, except above 800 K, where  
203 it decreases much more quickly than ERA-Interim so that they converge by 1200 K. The shading  
204 is the standard deviation of the annual averages that make up the five year average, and it shows  
205 the interannual variability, which is generally small.

## 206 4. Conclusions

207 In summary, we have calculated the strength of the overturning circulation of the stratosphere  
208 from observations, reanalyses, and a model. We find that at 460 K (about 60 hPa or 20 km in  
209 the tropics), the total overturning circulation of the stratosphere is  $7.3 \pm 0.3 \times 10^9$  kg/s based on  
210 two independent global satellite data products. Apart from that level, where the estimates are in  
211 relatively close agreement, substantial discrepancies exist.

212 The global SF<sub>6</sub> data have enabled this first quantitative calculation of the diabatic circulation in  
213 the middle and upper stratosphere. However, the interpretation of age from SF<sub>6</sub> is limited because  
214 we cannot quantify the impact of the mesospheric sink of SF<sub>6</sub>, which we find to be important  
215 above 500 K. We estimate that this makes the age difference a minimum of 60% too high at 1200  
216 K, which would imply a 35% low bias in the overturning strength at 1200 K, and we cannot  
217 estimate an upper bound on the bias. The reanalyses may correctly represent the true stratospheric  
218 circulation where they agree at the uppermost levels, although at those levels the data becomes  
219 more limited (e.g. Dee and Uppala 2009). Beneath 900 K, however, the reanalyses disagree with  
220 each other as well as with the circulation strength implied by the data; it is clear that the existing  
221 data are not sufficient to constrain estimates of the circulation.

222 Climate models predict an increase in the strength of the Brewer–Dobson circulation of about  
223 2% per decade (Butchart et al. 2006, Hardiman et al. 2014), which will impact stratospheric ozone,  
224 including the ozone hole recovery, and stratosphere troposphere exchange (Butchart 2014). Much  
225 effort has recently gone towards calculating trends in the stratospheric circulation based on ob-  
226 servations and reanalyses to see if such a trend can be detected (Engel et al. 2009, Seviour et al.  
227 2012, Diallo et al. 2012, Abalos et al. 2015). However, the mean diabatic circulation strength is  
228 not known except at one level. At the upper levels, the circulation is uncertain to within 100%. We

229 suggest cautious interpretation of trends that are much smaller than that uncertainty. More global  
230 age of air tracer data, in particular CO<sub>2</sub>, is necessary to provide an independent estimate of age  
231 difference necessary to calculate the strength of the diabatic stratospheric circulation.

## 232 **Methods**

233 **MIPAS SF<sub>6</sub>** For more details on validation and methods, we refer the readers to the papers on  
234 this product (Stiller et al. 2008, 2012; Haenel et al. 2015). We note that the vertical resolution  
235 is 4 to 6 km at 20 km, 7 to 10 km at 30 km, and 12 to 18 km at 40 km altitude. Noise error on  
236 individual profiles is of the order 20%, but because of the many profiles, meaningful SF<sub>6</sub> has been  
237 obtained by using monthly and zonal mean averages in 10 degree bins.

238 **N<sub>2</sub>O** Andrews et al. (2001) calculate an empirical fit between N<sub>2</sub>O and age from an exten-  
239 sive record of NASA ER-2 aircraft flights and high-altitude balloons from 1992–1998. Age is  
240 based on CO<sub>2</sub>, and for details of the conversion from CO<sub>2</sub> to age, see Andrews et al. (2001).  
241 The fit holds well for 50 ppbv < N<sub>2</sub>O < 300 ppbv and is given by the equation  $\Gamma(N_2O) =$   
242  $0.0581(313 - N_2O) - 0.000254(313 - N_2O)^2 + 4.41 \times 10^{-7}(313 - N_2O)^3$ , where 313 ppbv was  
243 the average tropospheric mixing ratio for 1992–1998. Although different tracer-tracer relation-  
244 ships are expected in the tropics and the extratropics (e.g. Strahan et al. 2011, Plumb 2007), the  
245 limited tropical data used to calculate this relationship were not treated separately. In order to ac-  
246 count for the increase in tropospheric N<sub>2</sub>O, we calculate the trend from the data product provided  
247 by the EPA Climate Indicators (US Environmental Protection Agency 2016), a combination of sta-  
248 tion measurements from Cape Grim, Australia, Mauna Loa, Hawaii, the South Pole, and Barrow,  
249 Alaska. The slope is  $0.806 \pm 0.014$  ppbv/yr. (One standard error on the slope is reported. Using  
250 only Mauna Loa, the tropical station, does not change the fit much, since N<sub>2</sub>O is quite well mixed  
251 in the troposphere.) We linearly adjust the GOZCARDS N<sub>2</sub>O data using this slope to account for

252 the growth in tropospheric N<sub>2</sub>O, although simply subtracting the mean difference in tropospheric  
253 N<sub>2</sub>O between 2009 and 1995 yielded very similar results. Then we apply the empirical relation-  
254 ship between 2004 and 2012 to obtain age estimates. Age difference is calculated only on those  
255 levels for which there are very few gaps in age. Only 460 and 470 K have no gaps at all. This  
256 method relies on several potentially problematic assumptions: the compact relationship from the  
257 1990s is assumed to be applicable over a decade later; the tropics are assumed be represented by  
258 this relationship well enough to obtain unbiased estimates of age difference; and linearly adjusting  
259 the data is assumed to sufficiently account for the changing tropospheric source.

260 **WACCM SF<sub>6</sub>** The method to calculate age from SF<sub>6</sub> in WACCM is as follows: The SF<sub>6</sub> on  
261 pressure levels is zonally averaged and then averaged in the same latitudinal bins that were used  
262 for MIPAS. That zonally averaged SF<sub>6</sub> is then converted to age following Stiller et al. (2012). The  
263 reference curve for SF<sub>6</sub> is the zonal mean value in the tropics at 100hPa just north of the equator  
264 (0.5° N) with a one year low-pass fourth order Butterworth filter applied to remove the weak  
265 seasonal cycle. Results are insensitive to the filtering provided the filter is sufficient to obtain  
266 a strictly increasing reference curve. We use the same method for correcting the age of air for  
267 the nonlinear tropospheric growth, with a Newtonian iteration (see Stiller et al. 2012 equation  
268 3). The nonlinearity correction is insensitive to the choice of constant parameter used to describe  
269 the relationship of the width of the age spectrum with the age. Once the age is determined, it  
270 is interpolated to isentropic levels using zonal mean temperatures that have also been binned by  
271 latitude according to the MIPAS grid. No attempt is made either by Haenel et al. (2015) or in this  
272 work to adjust the age for the mesospheric sink.

273 **Statistics for 460 K overturning** To calculate the average overturning circulation strength  
274 where the two data estimates agree most closely (within 5% at 460 K), we average them. The error  
275 estimate is based on the variability in the total overturning circulation strength from WACCM cal-

276 culated using  $\text{SF}_6$ -age to infer the circulation ( $M/\text{SF}_6$ -age  $\Delta\Gamma$ ). We take the average of five annual  
277 averages chosen randomly from the annual averages from 1999–2014 100,000 times. The standard  
278 deviation of the 100,000 resulting mean circulation strength estimates ( $0.14 \times 10^9$  kg/s) is taken  
279 to be half of the error. We repeated this procedure using the true overturning circulation strength  
280 ( $\mathcal{M}$ ) and found smaller variations in the standard deviation ( $0.09 \times 10^9$  kg/s). This error estimate  
281 assumes that WACCM represents the variability of the true circulation. The standard deviations  
282 of the five annual averages that were averaged for each data estimate were considerably smaller  
283 than these reported error bars. We therefore believe this is a conservative representation of the  
284 uncertainty in the diabatic circulation strength.

285 *Acknowledgments.* We thank Aditi Sheshadri and Susan Solomon for helpful discussions. Fund-  
286 ing for ML was provided by the National Defense Science and Engineering Graduate fellowship.  
287 This work was supported in part by the National Science Foundation grant AGS-1547733 to MIT  
288 and and AGS-1546585 to NYU. FH was funded by the “CAWSES” priority programme of the Ger-  
289 man Research Foundation (DFG) under project STI 210/5-3 and by the German Federal Ministry  
290 of Education and Research (BMBF) within the “ROMIC” programme under project 01LG1221B.  
291 MIPAS data processing was co-funded by the German Federal Ministry of Economics and Tech-  
292 nology (BMWV) within the “SEREMISA” project under contract number 50EE1547. AM acknowl-  
293 edges funding support from the European Research Council through the ACCI project (Grant  
294 267760) lead by John Pyle. The National Center for Atmospheric Research (NCAR) is sponsored  
295 by the U.S. National Science Foundation. Any opinions, findings, and conclusions or recommen-  
296 dations expressed in the publication are those of the author(s) and do not necessarily reflect the  
297 views of the National Science Foundation. WACCM is a component of the Community Earth Sys-  
298 tem Model (CESM), which is supported by the National Science Foundation (NSF) and the Office

299 of Science of the U.S. Department of Energy. Computing resources were provided by NCAR's  
300 Climate Simulation Laboratory, sponsored by NSF and other agencies. This research was en-  
301 abled by the computational and storage resources of NCAR's Computational and Information  
302 System Laboratory (CISL). A portion of the research was carried out at the Jet Propulsion Labo-  
303 ratory, California Institute of Technology, under a contract with the NASA Aeronautics and Space  
304 Administration. One of the datasets used for this study is from the Japanese 55-year Reanalysis  
305 (JRA-55) project carried out by the Japan Meteorological Agency (JMA). MERRA was developed  
306 by the Global Modeling and Assimilation Office and supported by the NASA Modeling, Analysis  
307 and Prediction Program. Source data files can be acquired from the Goddard Earth Science Data  
308 Information Services Center (GES DISC). ERA-Interim data provided courtesy ECMWF.

## 309 **References**

- 310 Abalos, M., B. Legras, F. Ploeger, and W. J. Randel, 2015: Evaluating the advective Brewer-  
311 Dobson circulation in three reanalyses for the period 1979–2012. *Journal of Geophysical Re-*  
312 *search, Atmospheres*, 1–21, doi:10.1002/2015JD023182, URL [http://onlinelibrary.wiley.com/](http://onlinelibrary.wiley.com/doi/10.1002/2015JD023182/full)  
313 [doi/10.1002/2015JD023182/full](http://onlinelibrary.wiley.com/doi/10.1002/2015JD023182/full).
- 314 Andrews, A. E., and Coauthors, 2001: Mean ages of stratospheric air derived from in situ obser-  
315 vations of CO, CH<sub>4</sub>, and N<sub>2</sub>O. *Journal of Geophysical Research*, **106**.
- 316 Birner, T., and H. Bönisch, 2011: Residual circulation trajectories and transit times into the ex-  
317 tratropical lowermost stratosphere. *Atmospheric Chemistry and Physics*, **11** (2), 817–827, doi:  
318 10.5194/acp-11-817-2011, URL <http://www.atmos-chem-phys.net/11/817/2011/>.
- 319 Butchart, N., 2014: The Brewer-Dobson circulation. *Reviews of Geophysics*, doi:10.1002/  
320 2013RG000448.One.

- 321 Butchart, N., and Coauthors, 2006: Simulations of anthropogenic change in the strength  
322 of the Brewer–Dobson circulation. *Climate Dynamics*, **27** (7-8), 727–741, doi:10.1007/  
323 s00382-006-0162-4, URL <http://link.springer.com/10.1007/s00382-006-0162-4>.
- 324 Butchart, N., and Coauthors, 2011: Multimodel climate and variability of the stratosphere.  
325 *Journal of Geophysical Research*, **116** (D5), D05 102, doi:10.1029/2010JD014995, URL <http://doi.wiley.com/10.1029/2010JD014995>.
- 327 Carlotti, M., B. M. Dinelli, G. Innocenti, and L. Palchetti, 2016: A strategy for the measurement of  
328 CO<sub>2</sub> distribution in the stratosphere. *Atmospheric Measurement Techniques*, **9** (12), 5853–5867,  
329 doi:10.5194/amt-9-5853-2016, URL <http://www.atmos-meas-tech.net/9/5853/2016/>.
- 330 Dee, D. P., and S. Uppala, 2009: Variational bias correction of satellite radiance data in the era-  
331 interim reanalysis. *Quarterly Journal of the Royal Meteorological Society*, **135** (644), 1830–  
332 1841, doi:10.1002/qj.493, URL <http://dx.doi.org/10.1002/qj.493>.
- 333 Dee, D. P., and Coauthors, 2011: The ERA-Interim reanalysis: configuration and performance of  
334 the data assimilation system. *Quarterly Journal of the Royal Meteorological Society*, **137** (656),  
335 553–597, doi:10.1002/qj.828, URL <http://dx.doi.org/10.1002/qj.828>.
- 336 Diallo, M., B. Legras, and a. Chédin, 2012: Age of stratospheric air in the ERA-Interim. *Atmo-*  
337 *spheric Chemistry and Physics*, **12** (24), 12 133–12 154, doi:10.5194/acp-12-12133-2012, URL  
338 <http://www.atmos-chem-phys.net/12/12133/2012/>.
- 339 Engel, A., and Coauthors, 2009: Age of stratospheric air unchanged within uncertainties over the  
340 past 30 years. *Nature Geoscience*, **2**, 28–31.

341 Flury, T., D. L. Wu, and W. G. Read, 2013: Variability in the speed of the Brewer–Dobson cir-  
342 culation as observed by Aura/MLS. *Atmospheric Chemistry and Physics*, **13** (9), 4563–4575,  
343 doi:10.5194/acp-13-4563-2013, URL <http://www.atmos-chem-phys.net/13/4563/2013/>.

344 Froidevaux, L., R. A. Fuller, A. Lambert, N. J. Livesey, P. F. Bernath, and K. A. Walker,  
345 2013: GOZCARDS Source Data for Nitrous Oxide Monthly Zonal Averages on a Geode-  
346 tic Latitude and Pressure Grid v1.01. NASA Goddard Earth Sciences Data and Informa-  
347 tion Services Center, URL <https://doi.org/10.5067/MEASURES/GOZCARDS/DATA3012>, doi:  
348 10.5067/MEASURES/GOZCARDS/DATA3012.

349 Garcia, R. R., A. K. Smith, D. E. Kinnison, Ivaro de la Cmara, and D. J. Murphy, 2017: Mod-  
350 ification of the Gravity Wave Parameterization in the Whole Atmosphere Community Cli-  
351 mate Model: Motivation and Results. *Journal of the Atmospheric Sciences*, **74** (1), 275–  
352 291, doi:10.1175/JAS-D-16-0104.1, URL <http://dx.doi.org/10.1175/JAS-D-16-0104.1>, <http://dx.doi.org/10.1175/JAS-D-16-0104.1>.

354 Haenel, F. J., and Coauthors, 2015: Reassessment of MIPAS age of air trends and variability.  
355 *Atmospheric Chemistry and Physics*, **15**, 13 161–13 176, doi:10.5194/acp-15-13161-2015, URL  
356 [www.atmos-chem-phys.net/15/13161/2015/](http://www.atmos-chem-phys.net/15/13161/2015/).

357 Hall, T. M., and D. W. Waugh, 1998: Influence of nonlocal chemistry on tracer distributions: Infer-  
358 ring the mean age of air from SF<sub>6</sub>. *Journal of Geophysical Research: Atmospheres*, **103** (D11),  
359 13 327–13 336, doi:10.1029/98JD00170, URL <http://dx.doi.org/10.1029/98JD00170>.

360 Hardiman, S. C., N. Butchart, and N. Calvo, 2014: The morphology of the Brewer-Dobson  
361 circulation and its response to climate change in CMIP5 simulations. *Quarterly Journal of*  
362 *the Royal Meteorological Society*, **140** (683), 1958–1965, doi:10.1002/qj.2258, URL <http://doi.wiley.com/10.1002/qj.2258>.

363

- 364 Kobayashi, S., and Coauthors, 2015: The JRA-55 Reanalysis: General Specifications and Basic  
365 Characteristics. *Journal of the Meteorological Society of Japan. Ser. II*, **93** (1), 5–48, doi:10.  
366 2151/jmsj.2015-001.
- 367 Kovács, T., and Coauthors, 2017: Determination of the atmospheric lifetime and global warming  
368 potential of sulfur hexafluoride using a three-dimensional model. *Atmospheric Chemistry and*  
369 *Physics*, **17** (2), 883–898, doi:10.5194/acp-17-883-2017, URL [http://www.atmos-chem-phys.](http://www.atmos-chem-phys.net/17/883/2017/)  
370 [net/17/883/2017/](http://www.atmos-chem-phys.net/17/883/2017/).
- 371 Lin, S.-J., 2004: A “Vertically Lagrangian” Finite-Volume Dynamical Core for Global Mod-  
372 els. *Monthly Weather Review*, **132** (10), 2293–2307, doi:10.1175/1520-0493(2004)132<2293:  
373 AVLFDC>2.0.CO;2, URL [http://dx.doi.org/10.1175/1520-0493\(2004\)132<2293:AVLFDC>2.0.](http://dx.doi.org/10.1175/1520-0493(2004)132<2293:AVLFDC>2.0.CO;2)  
374 [CO;2, http://dx.doi.org/10.1175/1520-0493\(2004\)132<2293:AVLFDC>2.0.CO;2.](http://dx.doi.org/10.1175/1520-0493(2004)132<2293:AVLFDC>2.0.CO;2)
- 375 Linz, M., R. A. Plumb, E. P. Gerber, and A. Sheshadri, 2016: The relationship between age of air  
376 and the diabatic circulation of the stratosphere. *Journal of the Atmospheric Sciences*, **73** (11),  
377 4507–4518, doi:10.1175/JAS-D-16-0125.1.
- 378 Mahieu, E., and Coauthors, 2014: Recent Northern Hemisphere stratospheric HCl increase due  
379 to atmospheric circulation changes. *Nature*, **515** (7525), 104–7, doi:10.1038/nature13857, URL  
380 <http://www.ncbi.nlm.nih.gov/pubmed/25373680>.
- 381 Marsh, D. R., M. J. Mills, D. E. Kinnison, J.-F. Lamarque, N. Calvo, and L. M. Polvani,  
382 2013: Climate Change from 1850 to 2005 Simulated in CESM1(WACCM). *Journal of Cli-*  
383 *mate*, **26** (19), 7372–7391, doi:10.1175/JCLI-D-12-00558.1, URL [http://dx.doi.org/10.1175/](http://dx.doi.org/10.1175/JCLI-D-12-00558.1)  
384 [JCLI-D-12-00558.1, http://dx.doi.org/10.1175/JCLI-D-12-00558.1.](http://dx.doi.org/10.1175/JCLI-D-12-00558.1)

- 385 Morgenstern, O., and Coauthors, 2017: Review of the global models used within phase 1 of the  
386 Chemistry–Climate Model Initiative (CCMI). *Geoscientific Model Development*, **10** (2), 639–  
387 671, doi:10.5194/gmd-10-639-2017, URL <http://www.geosci-model-dev.net/10/639/2017/>.
- 388 Mote, P. W., and Coauthors, 1996: An atmospheric tape recorder: The imprint of tropical  
389 tropopause temperatures on stratospheric water vapor. *Journal of Geophysical Research: At-*  
390 *mospheres*, **101** (D2), 3989–4006, doi:10.1029/95JD03422, URL [http://dx.doi.org/10.1029/](http://dx.doi.org/10.1029/95JD03422)  
391 [95JD03422](http://dx.doi.org/10.1029/95JD03422).
- 392 Neale, R. B., J. Richter, S. Park, P. H. Lauritzen, S. J. Vavrus, P. J. Rasch, and M. Zhang, 2013: The  
393 Mean Climate of the Community Atmosphere Model (CAM4) in Forced SST and Fully Coupled  
394 Experiments. *Journal of Climate*, **26** (14), 5150–5168, doi:10.1175/JCLI-D-12-00236.1, URL  
395 <http://dx.doi.org/10.1175/JCLI-D-12-00236.1>, <http://dx.doi.org/10.1175/JCLI-D-12-00236.1>.
- 396 Neu, J. L., and R. A. Plumb, 1999: Age of air in a “leaky pipe” model of stratospheric transport.  
397 *J. Geophys. Res.*, **104** (D16), 19 243–19 255, doi:10.1029/1999JD900251.
- 398 Plumb, R. A., 2007: Tracer interrelationships in the stratosphere. *Reviews of Geophysics*, 1–33,  
399 doi:10.1029/2005RG000179.1.INTRODUCTION.
- 400 Plumb, R. A., and M. K. W. Ko, 1992: Interrelationships between mixing ratios of long-lived  
401 stratospheric constituents. *Journal of Geophysical Research: Atmospheres*, **97** (D9), 10 145–  
402 10 156, doi:10.1029/92JD00450, URL <http://dx.doi.org/10.1029/92JD00450>.
- 403 Plumb, R. A., and Coauthors, 2002: Global tracer modeling during SOLVE: High-latitude de-  
404 scent and mixing. *Journal of Geophysical Research: Atmospheres*, **107** (D5), doi:10.1029/  
405 [2001JD001023](http://dx.doi.org/10.1029/2001JD001023), URL <http://dx.doi.org/10.1029/2001JD001023>, 8309.

406 Rienecker, M. M., and Coauthors, 2011: MERRA: NASA's Modern-Era Retrospective Anal-  
407 ysis for Research and Applications. *Journal of Climate*, **24 (14)**, 3624–3648, doi:10.1175/  
408 JCLI-D-11-00015.1, URL <http://dx.doi.org/10.1175/JCLI-D-11-00015.1>.

409 Schoeberl, M. R., A. R. Douglass, R. S. Stolarski, S. Pawson, S. E. Strahan, and W. Read, 2008:  
410 Comparison of lower stratospheric tropical mean vertical velocities. *Journal of Geophysical*  
411 *Research: Atmospheres*, **113 (D24)**, n/a–n/a, doi:10.1029/2008JD010221, URL <http://dx.doi.org/10.1029/2008JD010221>, d24109.

413 Seviour, W. J. M., N. Butchart, and S. C. Hardiman, 2012: The Brewer-Dobson circulation inferred  
414 from ERA-Interim. *Quarterly Journal of the Royal Meteorological Society*, **138 (665)**, 878–888,  
415 doi:10.1002/qj.966, URL <http://doi.wiley.com/10.1002/qj.966>.

416 Sparling, L. C., J. A. Kettleborough, P. H. Haynes, M. E. McIntyre, J. E. Rosenfield, M. R. Schoe-  
417 berl, and P. A. Newman, 1997: Diabatic cross-isentropic dispersion in the lower stratosphere. *J.*  
418 *Geophys. Res.*, **102 (D22)**, 25 817–25 829, doi:10.1029/97JD01968.

419 Stiller, G. P., and Coauthors, 2008: Global distribution of mean age of stratospheric air from  
420 MIPAS SF<sub>6</sub> measurements. *Atmospheric Chemistry and Physics*, **8 (3)**, 677–695, doi:10.5194/  
421 acp-8-677-2008, URL <http://www.atmos-chem-phys.net/8/677/2008/>.

422 Stiller, G. P., and Coauthors, 2012: Observed temporal evolution of global mean age of strato-  
423 spheric air for the 2002 to 2010 period. *Atmospheric Chemistry and Physics*, **12 (7)**, 3311–3331,  
424 doi:10.5194/acp-12-3311-2012, URL <http://www.atmos-chem-phys.net/12/3311/2012/>.

425 Strahan, S. E., and Coauthors, 2011: Using transport diagnostics to understand chemistry climate  
426 model ozone simulations. *Journal of Geophysical Research: Atmospheres*, **116 (D17)**, doi:10.  
427 1029/2010JD015360, URL <http://dx.doi.org/10.1029/2010JD015360>, d17302.

428 Totterdill, A., T. Kovcs, J. C. Gmez Martn, W. Feng, and J. M. C. Plane, 2015: Mesospheric  
429 Removal of Very Long-Lived Greenhouse Gases SF<sub>6</sub> and CFC-115 by Metal Reactions, Lyman-  
430  $\alpha$  Photolysis, and Electron Attachment. *The Journal of Physical Chemistry A*, **119** (10), 2016–  
431 2025, doi:10.1021/jp5123344, URL <http://dx.doi.org/10.1021/jp5123344>.

432 US Environmental Protection Agency, 2016: Climate change indicators in the United States. EPA  
433 430-R-16-004, URL <https://www.epa.gov/climate-indicators>.

434 von Clarmann, T., and Coauthors, 2003: Retrieval of temperature and tangent altitude pointing  
435 from limb emission spectra recorded from space by the Michelson Interferometer for Passive  
436 Atmospheric Sounding (MIPAS). *Journal of Geophysical Research: Atmospheres*, **108** (D23),  
437 n/a–n/a, doi:10.1029/2003JD003602, URL <http://dx.doi.org/10.1029/2003JD003602>, 4736.

438 von Clarmann, T., and Coauthors, 2009: Retrieval of temperature, H<sub>2</sub>O, O<sub>3</sub>, HNO<sub>3</sub>, CH<sub>4</sub>, N<sub>2</sub>O,  
439 ClONO<sub>2</sub> and ClO from MIPAS reduced resolution nominal mode limb emission measure-  
440 ments. *Atmospheric Measurement Techniques*, **2** (1), 159–175, doi:10.5194/amt-2-159-2009,  
441 URL <http://www.atmos-meas-tech.net/2/159/2009/>.

442 Waugh, D., and T. M. Hall, 2002: Age of stratospheric air: Theory, observations, and models.  
443 *Reviews of Geophysics*, **40** (4), 1010, doi:10.1029/2000RG000101, URL [http://doi.wiley.com/](http://doi.wiley.com/10.1029/2000RG000101)  
444 [10.1029/2000RG000101](http://doi.wiley.com/10.1029/2000RG000101).

445 **List of Tables**

446 **Table 1.** Data, reanalyses, and model output used in this study. SW is the shortwave  
447 radiation and LW is the longwave radiation. . . . . 23

Data source	Variables	Resolution	Time period	Reference
MIPAS	age from SF <sub>6</sub> ; temperature; pressure	zonal mean, 10° lat, 41 levels from 8 km to 54 km	2002–2012	Haenel et al. 2015
GOZ-CARDS	N <sub>2</sub> O	zonal mean, 10° lat, 15 pressure levels from 100 to 0.46 hPa	2004–2014	Froidevaux et al. 2015, Andrews et al. 2001
EPA Climate Indicators	tropospheric N <sub>2</sub> O	<i>in situ</i> surface	1980–2014	US EPA 2016
WACCM	SW; LW; temperature; ideal age; SF <sub>6</sub>	2.5 ° lon, 1.875 ° lat, 31 pressure levels from 193 hPa to 0.3 hPa	1979–2014	Marsh et al., 2013, Garcia et al. 2017
JRA 55	SW; LW; temperature	1.25° × 1.25°, 16 pressure levels from 225 hPa to 1 hPa	1979–2014	Kobayashi et al. 2015
MERRA	total dT/dt; temperature	1.25° × 1.25°, 17 pressure levels from 200 hPa to 0.5 hPa	1979–2014	Rienecker et al. 2011
ERA-Interim	SW; LW; temperature	1° × 1°, 26 pressure levels from 150 hPa to 0.5 hPa	1979–2014	Dee et al. 2011

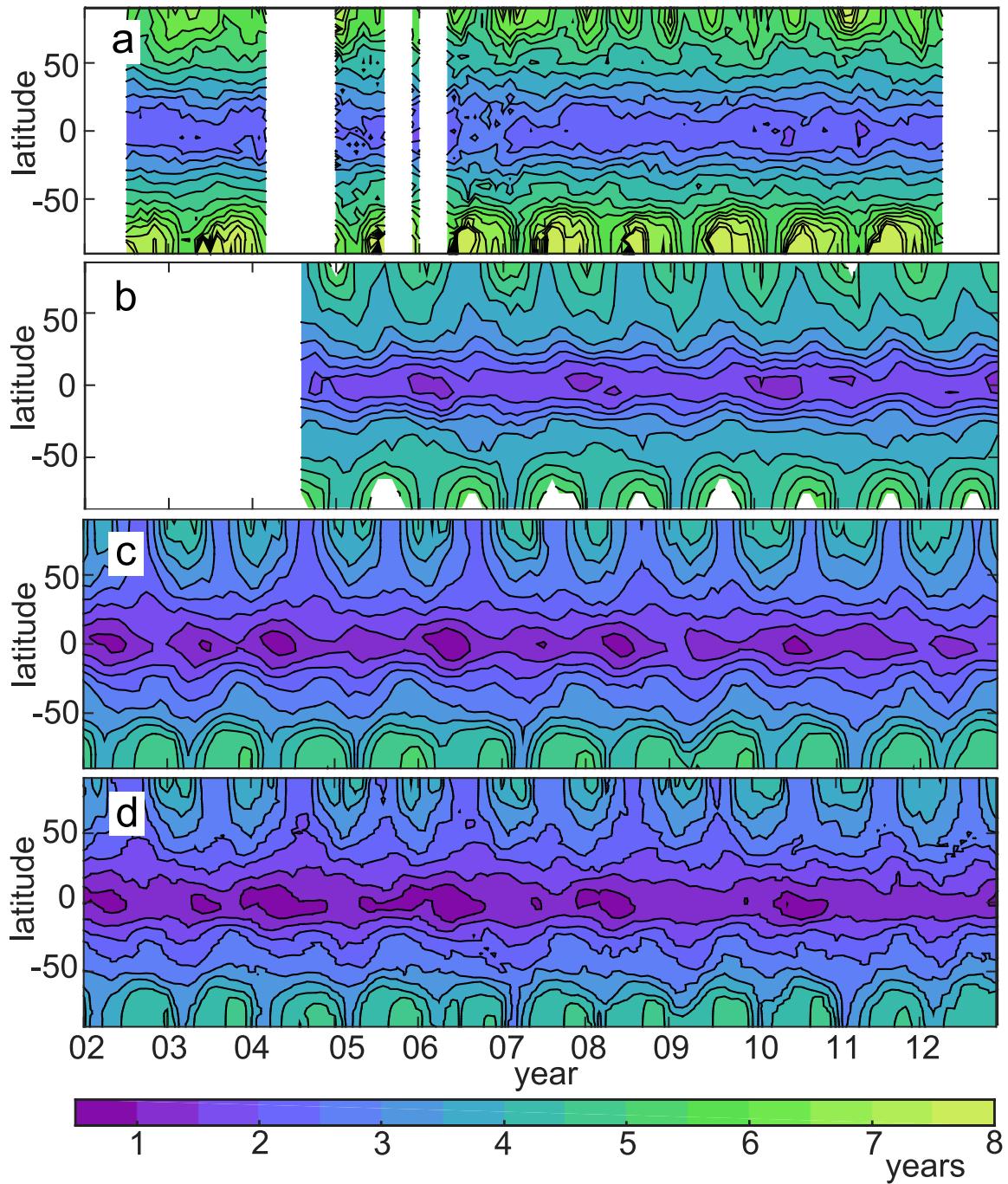
448 Table 1. Data, reanalyses, and model output used in this study. SW is the shortwave radiation and LW is the  
449 longwave radiation.

450 **List of Figures**

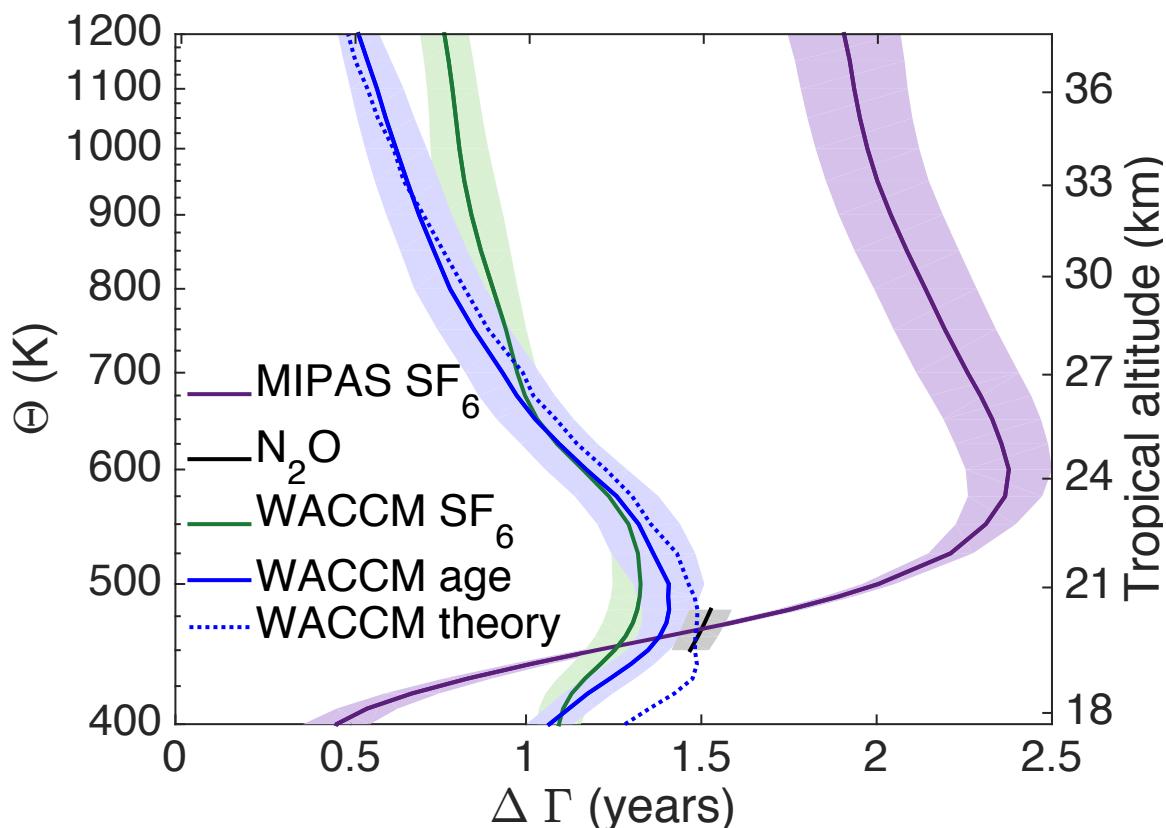
451 **Fig. 1.** Age of air on the 500 K surface. (a) SF<sub>6</sub> from MIPAS, (b) N<sub>2</sub>O from GOZCARDS, (c) SF<sub>6</sub>  
452 from WACCM, and (d) WACCM ideal age tracer. Contours are every half year, and the ages  
453 in the Southern Hemisphere winter for MIPAS get above 8 years old. . . . . 25

454 **Fig. 2.** The average age difference between downwelling and upwelling age of air on each isentrope  
455 between 2007–2011.  $\Delta\Gamma$  is plotted in solid lines: MIPAS SF<sub>6</sub>-age in purple, GOZCARDS  
456 N<sub>2</sub>O age in black, WACCM SF<sub>6</sub>-age in green, and WACCM ideal age of air in the blue.  
457 The blue dotted line shows the ratio of the total mass above each isentrope to the mass flux  
458 through the isentrope ( $M/\mathcal{M}$ ) from WACCM. The shading shows one standard deviation of  
459 the five annual averages that are averaged to get the mean. The mean height of each isentrope  
460 in the tropics (calculated from MIPAS pressure and temperature) is on the right y-axis. . . . . 26

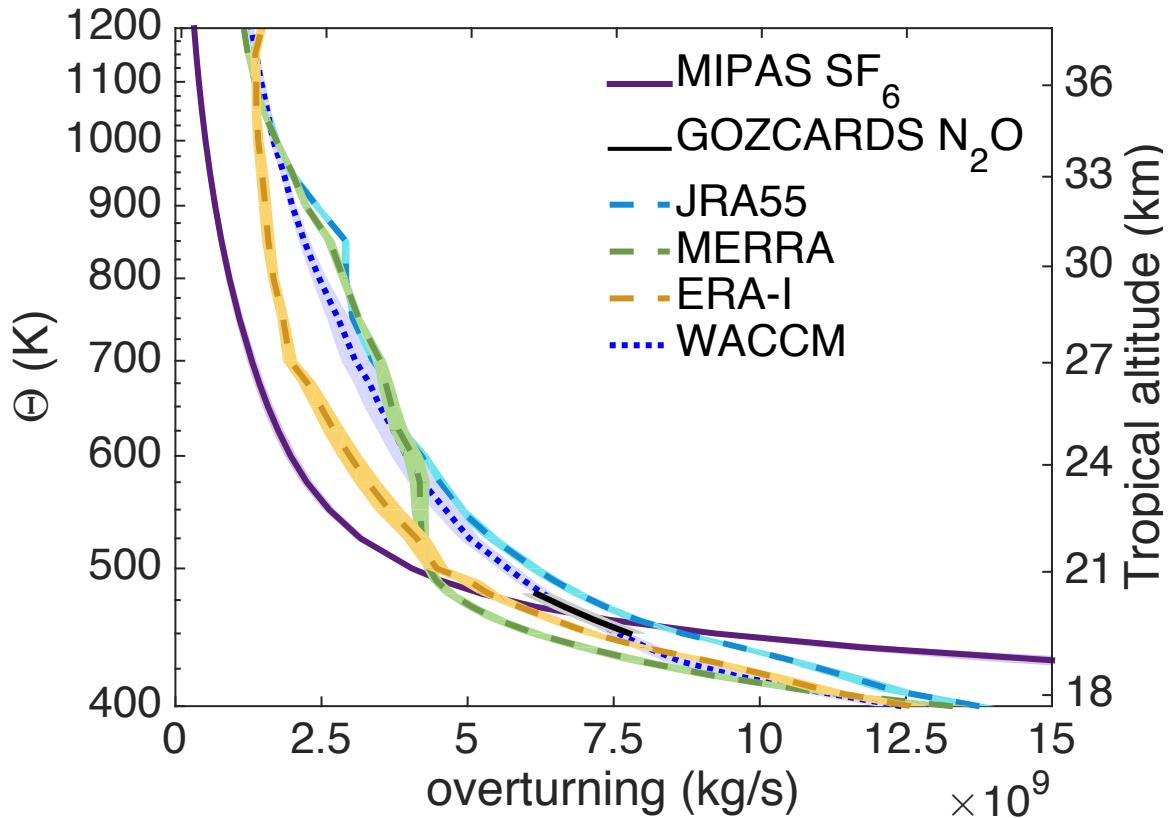
461 **Fig. 3.** The strength of the total overturning circulation through each isentrope averaged between  
462 2007–2011. The solid lines are for the data-based estimates MIPAS SF<sub>6</sub> is in purple and  
463 GOZCARDS N<sub>2</sub>O in black. Reanalyses are shown in dashed lines: JRA 55 in light blue,  
464 MERRA in green and ERA-Interim in gold. The dotted blue line is WACCM. The shading  
465 shows one standard deviation of the five annual averages. The details of the calculation for  
466 each data product, the model, and the reanalyses are described in the text. The mean height  
467 of each isentrope in the tropics (calculated from MIPAS pressure and temperature) is on the  
468 right y-axis. . . . . 27



469 Figure 1. Age of air on the 500 K surface. (a) SF<sub>6</sub> from MIPAS, (b) N<sub>2</sub>O from GOZCARDS, (c) SF<sub>6</sub>  
 470 from WACCM, and (d) WACCM ideal age tracer. Contours are every half year, and the ages in the Southern  
 471 Hemisphere winter for MIPAS get above 8 years old.



472 Figure 2. The average age difference between downwelling and upwelling age of air on each isentrope be-  
 473 tween 2007–2011.  $\Delta\Gamma$  is plotted in solid lines: MIPAS  $\text{SF}_6$ -age in purple, GOZCARDS  $\text{N}_2\text{O}$  age in black,  
 474 WACCM  $\text{SF}_6$ -age in green, and WACCM ideal age of air in the blue. The blue dotted line shows the ratio of  
 475 the total mass above each isentrope to the mass flux through the isentrope ( $M/\mathcal{M}$ ) from WACCM. The shading  
 476 shows one standard deviation of the five annual averages that are averaged to get the mean. The mean height of  
 477 each isentrope in the tropics (calculated from MIPAS pressure and temperature) is on the right y-axis.



478 Figure 3. The strength of the total overturning circulation through each isentrope averaged between 2007–  
 479 2011. The solid lines are for the data-based estimates MIPAS SF<sub>6</sub> is in purple and GOZCARDS N<sub>2</sub>O in black.  
 480 Reanalyses are shown in dashed lines: JRA 55 in light blue, MERRA in green and ERA-Interim in gold. The  
 481 dotted blue line is WACCM. The shading shows one standard deviation of the five annual averages. The details  
 482 of the calculation for each data product, the model, and the reanalyses are described in the text. The mean height  
 483 of each isentrope in the tropics (calculated from MIPAS pressure and temperature) is on the right y-axis.