

## Tracer transport in the tropical stratosphere due to vertical diffusion and horizontal mixing

Timothy M. Hall and Darryn Waugh

CRC for Southern Hemisphere Meteorology, Monash University, Clayton, Australia

### Abstract.

Observations of linearly increasing (e.g., SF<sub>6</sub>) and periodically varying (e.g., H<sub>2</sub>O+2CH<sub>4</sub>) long-lived tracers in the lower stratosphere provide independent constraints on theories of transport in the region. Taken together, these data allow separation of the roles of diffusion (with coefficient  $K_\theta$ ) and advection (at rate  $Q$ ) through isentropic surfaces, and mixing of extra-tropical air into the tropics (with relaxation time  $\tau$ ). Using a one-dimensional diffusive-advective model of the tropical stratosphere, which allows relaxation of mixing ratios to extra-tropical values, we obtain solutions for periodic and linear tracers. Fitting the solutions to observations yields  $K_\theta \approx 0.3$  K<sup>2</sup>/day ( $K_z \approx 0.01$  m<sup>2</sup>/s),  $\tau \approx 1.3$  years, and  $Q \approx 0.5$  K/day ( $\bar{w} \approx 0.3$  mm/s). These values produce profiles for CO<sub>2</sub> in reasonable agreement with aircraft observations. However, a large range of  $K_\theta$  results in equally good agreement, although  $\tau$  and  $Q$  are more tightly constrained. In the lower tropical stratosphere, vertical diffusion appears to play little role in transporting tracer.

### 1. Introduction

The rate at which lower stratospheric extra-tropical air mixes into the tropics and is lofted upwards determines the extent to which midlatitude aircraft emissions will affect the ozone layer. Although aerosol observations [Trepte and Hitchman, 1992] suggested a mixing barrier between the tropics and extratropics, an idea formalized by Plumb [1996], photochemical calculations show the tropics cannot be completely isolated [Avalone and Prather, 1996]. Recently, several studies have estimated rates of mixing and ascent using aircraft [Boering et al., 1996; Minschwaner et al., 1996; Volk et al., 1996] and satellite measurements [Mote et al., 1996; Schoeberl et al., 1997; Randel et al., 1997].

One way to estimate transport parameters is to observe the vertically propagating signal in the tropics of conserved tracers whose mixing ratio varies periodically (e.g.,  $\hat{H} = \text{H}_2\text{O}+2\text{CH}_4$  and CO<sub>2</sub>). However, the oscillation amplitude alone cannot pin down a horizon-

tal mixing timescale, as it is attenuated by both the presence of extratropical air and by vertical diffusion. Limiting bounds on the mixing rate and diffusion have been estimated from the  $\hat{H}$  amplitude; e.g., Mote et al. [1996] assumed no mixing to obtain an upper bound on the rate of diffusion, while Randel et al. [1997] assumed no diffusion to obtain an upper bound on the rate of mixing. Recently, Phil Mote (personal communications; 1997) has used H<sub>2</sub>O and CH<sub>4</sub> separately to estimate rates of diffusion and mixing simultaneously.

Hall and Waugh [1997] showed that the phase lag of a periodic signal and the mean age (the stratospheric lag time to a tracer varying linearly in the troposphere [e.g., Hall and Plumb, 1994]) are timescales characterizing different properties of transport. In this paper we show how these two timescales supplement the amplitude information, thus providing three independent constraints on the roles of horizontal mixing, vertical diffusion, and vertical advection in the tropics. We estimate these parameters using analyses of  $\hat{H}$  from the Halogen Occultation Experiment (HALOE) instrument aboard Upper Atmosphere Research Satellite (UARS) [Randel et al., 1997], and of SF<sub>6</sub> by the ACATS instrument [Elkins et al., 1996] during the Airborne Southern Hemisphere Ozone Experiment (ASHOE) and Stratospheric Tracers of Atmospheric Transport (STRAT) campaigns.

### 2. One Dimensional Model

In order to extract rates of diffusion, mixing, and vertical advection from tracer observations, we use the simplest model possible that includes these effects. We solve the one-dimensional tracer continuity equation

$$\frac{\partial \chi}{\partial t} + Q \frac{\partial \chi}{\partial \theta} - \frac{1}{\rho} K_\theta \frac{\partial}{\partial \theta} \left( \rho \frac{\partial \chi}{\partial \theta} \right) = -\frac{1}{\tau} (\chi - \chi_e) \quad (1)$$

where  $\chi$  is a tracer mixing ratio,  $K_\theta$  a coefficient of vertical diffusion,  $Q$  a vertical velocity,  $\tau$  a relaxation time to the specified extra-tropical mixing ratio  $\chi_e$ , summarizing the mixing rate of air into the tropics, and  $\rho$  the air density falling exponentially with scale height  $H_\theta$ . The vertical coordinate is potential temperature  $\theta$  above the tropical tropopause (assumed to be at 390K). In the atmosphere,  $K_\theta$ ,  $Q$ , and  $\tau$  vary with space and time, but here we consider only constant values.

We solve (1) for two boundary conditions: (i), a sinusoidal source ( $\chi(0, t) = \cos \omega t$ ), and (ii), a tracer of

linearly increasing abundance ( $\chi(0, t) = ct$ ). The periodic solution has amplitude  $A$  and phase lag  $\tau_\omega$ , while the linear solution determines the mean age  $\Gamma$ . In each case, the extra-tropical value  $\chi_e$  must be specified. For the periodic tracer, we take  $\chi_e = 0$ , as observations of  $\hat{H}$  show small amplitude in the extra-tropical stratosphere compared to the tropics [Randel *et al.*, 1997]. For the linear tracer, we take  $\chi_e = c(t - \Gamma_e(\theta))$ , where  $\Gamma_e(\theta) = m\theta + \Gamma_e(0)$  is an extra-tropical vertical age profile with a constant vertical age gradient  $m$ .

The explicit solutions are shown in Table 1. For low diffusion ( $\tau_\theta\omega \ll 1$ ,  $Q \gg K_\theta/H$ , and  $\tau \gg \tau_\theta$ ) the amplitude  $A \sim e^{-\theta/\tau Q}$  and the phase lag  $\tau_\omega \sim \theta/Q$ . In this limit, diffusion plays a negligible role in propagating the annual cycle compared to advection, and also a negligible role in attenuating the cycle compared to relaxation to the extra-tropical value.

### 3. Observations

The HALOE instrument measures stratospheric  $\text{H}_2\text{O}$  and  $\text{CH}_4$ , and the quantity  $\hat{H}$  is well conserved in the lower and middle stratosphere [Dessler *et al.*, 1994].  $\hat{H}$  varies seasonally at the tropical tropopause, and this signal is transported vertically through the tropical stratosphere [Mote *et al.*, 1995; 1996]. Randel *et al.* [1997] have fit annual and semi-annual harmonics to the HALOE  $\hat{H}$  time series from 1991 to 1996. We use their phase and peak-to-peak amplitude at 100 hPa to 31 hPa to obtain  $k_A = 0.0043 \text{ K}^{-1}$  and  $k_\omega = 0.0050 \text{ yr/K}$  for the coefficients defined in Table 1, giving a 55% attenuation and an 11 month phase lag at 31 hPa.

$\text{SF}_6$  is nearly inert and its abundance is increasing approximately linearly [Maiss *et al.*, 1996; Geller *et al.*, 1997]. Here we use  $\text{SF}_6$ -inferred age from ASHOE and STRAT data to estimate mean age [Elkins *et al.*, 1996; Waugh *et al.*, 1997; Volk *et al.*, 1997]. Using the surface mean time series from Geller *et al.* [1997] and a tropopause age of 0.8 year [Volk *et al.*, 1997], we obtain, in the tropics ( $\pm 15^\circ$ ) at 80K ( $\pm 5\text{K}$ ) above the tropopause,  $\Gamma = 0.9 \pm 0.3$  years with respect to the tropopause. To obtain an estimate of  $\Gamma_e(0)$ , we average mean ages between 0 and 20K from the height of the tropical tropopause (i.e., 390 and 410K) at latitudes between  $20^\circ$  and  $40^\circ$ , finding  $\Gamma_e(0) = 0.9 \pm 0.4$  years, again with respect to the tropical tropopause. Linear fits of the vertical age variation in the extra-tropics produce  $m = 0.025 \text{ yr/K}$ . Finally,  $H = 7 \text{ km}$  is converted to  $H_\theta = 162 \text{ K}$  for use in (1) by employing the close linear relation between  $\log p$  and  $\theta$  of ASHOE and STRAT data in the tropical lower stratosphere.

### 4. Solving for $K_\theta$ , $\tau$ , and $Q$

$A$  and  $\tau_\omega$  (Table 1) may be used to eliminate  $Q$ , thereby providing a relation between  $K_\theta$  and  $\tau$ . This “ $A$ - $\tau_\omega$  curve” is drawn as a heavy solid line in Figure

**Table 1.** Explicit Solutions

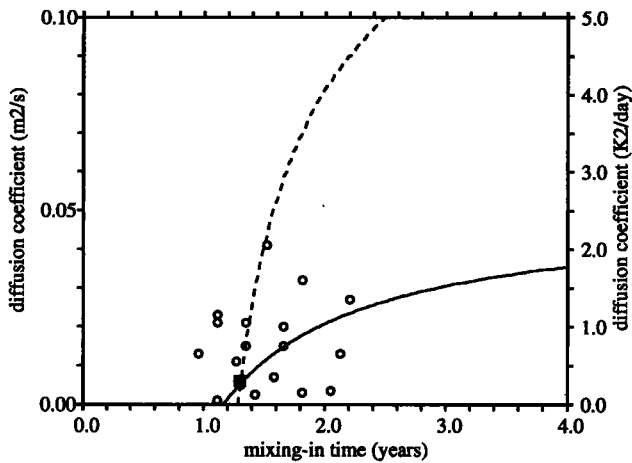
<p>periodic boundary condition <math>\chi(0, t) = \cos \omega t</math>:  <math>\chi(\theta, t) = A \cos \omega(t - \tau_\omega)</math>            where <math>A = e^{-k_A \theta}</math>, <math>\tau_\omega = k_\omega \theta</math>,  <math>k_A = (\tilde{Q}/2K_\theta) (\psi \cos(\frac{1}{2} \tan^{-1} \gamma) - 1)</math>,  <math>k_\omega = (\tilde{Q}/2K_\theta \omega) \psi \sin(\frac{1}{2} \tan^{-1} \gamma)</math>,  <math>\psi = (\tau_\theta \omega)^{1/2} (1 + \gamma^{-2})^{1/4}</math>,  <math>\gamma = \omega / (\tau_\theta^{-1} + \tau^{-1})</math>, <math>\tau_\theta = 4K_\theta / \tilde{Q}^2</math>, and  <math>\tilde{Q} = Q + K_\theta / H_\theta</math></p>
<p>linear boundary condition <math>\chi(0, t) = ct</math>:  <math>\chi(\theta, t) = c(t - \Gamma)</math>            where <math>\Gamma = (\Gamma_e(\theta) - \delta\Gamma) - (\Gamma_e(0) - \delta\Gamma)e^{-\theta/L}</math>,  <math>\Gamma_e(\theta) = m\theta + \Gamma_e(0)</math>, <math>\delta\Gamma = \tau(m\tilde{Q} - 1)</math>, and  <math>L = (2K_\theta / \tilde{Q}) (\sqrt{1 + \tau_\theta / \tau} - 1)^{-1}</math></p>

1. There are two limits to note. For  $\tau \rightarrow \infty$  (complete isolation of the tropics), vertical diffusion is solely responsible for the observed attenuation of the periodic tracer signal. This upper bound on  $K_\theta$  is about  $1.8 \text{ K}^2/\text{s}$  ( $K_z \approx 0.04 \text{ m}^2/\text{s}$ ), within the range obtained by Mote *et al.* [1996]. In the opposite limit,  $K_\theta \rightarrow 0$ , all the attenuation of the periodic signal is due to mixing of extra-tropical air into the tropics.  $\tau$  in this limit is about 1.2 years, a lower bound consistent with Randel *et al.* [1997].

A second relation between  $K_\theta$  and  $\tau$  is obtained by eliminating  $Q$  from  $\tau_\omega$  and  $\Gamma$  (Table 1), producing a “ $\Gamma$ - $\tau_\omega$  curve”. This curve is shown in Figure 1 as a heavy dashed line. The  $A$ - $\tau_\omega$  and  $\Gamma$ - $\tau_\omega$  curves intersect at  $\tau \approx 1.3$  years and  $K_\theta \approx 0.3 \text{ K}^2/\text{day}$  ( $K_z \approx 0.01 \text{ m}^2/\text{s}$ ). The simultaneous solution for the vertical velocity is  $Q \approx 0.5 \text{ K/s}$  ( $\bar{w} \approx 0.3 \text{ mm/s}$ ). This represents our best estimate for these values.

The best estimate values are in the low diffusion limit. The phase velocity  $\theta/\tau_\omega$  of the HALOE  $\hat{H}$  signal only differs from  $Q$  by 1%, the first order correction to  $Q$  being  $4K_\theta/(Q\tau)$ , which is 100 times smaller. The effect of diffusion on the amplitude is greater, the first order correction to  $k_A \sim (\tau Q)^{-1}$  being  $\omega^2 K_\theta / Q^3$ , which is about 10 times smaller: for best estimate values,  $A(80\text{K}) = 0.70$ , while for the same  $\tau$  but  $K_\theta = 0$ ,  $A(80\text{K}) = 0.73$ .

The circles of Figure 1 are solutions obtained by varying  $A$ ,  $\tau_\omega$  and  $\Gamma$  by  $\pm 10\%$  about the fits to observations. Some combinations (not plotted) shift the  $\Gamma$ - $\tau_\omega$  and  $A$ - $\tau_\omega$  curves far enough apart that there is no intersection.  $Q$  is insensitive to  $A$  and  $\Gamma$ , but varies nearly inversely with  $\tau_\omega$ . Despite these uncertainties, it is encouraging that  $\tau = 1.3$  years is within the range  $1.1 \pm 0.2$  years obtained by Volk *et al.* [1996] in an independent analysis, and  $\bar{w} \approx 0.3 \text{ mm/s}$  falls within the uncertainty range of the annual mean vertical velocity in the tropics estimated by Rosenlof [1995].



**Figure 1.** Curves of the vertical diffusion coefficient  $K_\theta$  versus the mixing-in time  $\tau$  derived from fits to satellite and aircraft observations. Solid and dashed lines represent independent techniques. The best estimate solution is indicated at the intersection point by the dark square symbol. Circles are solutions using 10% variations of observations about the fits. See text for details.

## 5. Predicted CO<sub>2</sub> Profiles

Measurements of stratospheric CO<sub>2</sub> [Boering *et al.*, 1994; 1996] are independent data testing our estimates of  $K_\theta$ ,  $\tau$ , and  $Q$ . CO<sub>2</sub> exhibits stratospheric gradients due to both a long term quasi-linear trend and an annual cycle in the troposphere. Thus, we combine solutions for oscillating and linear tracers to obtain a modeled CO<sub>2</sub> profile. As a boundary condition, we fit a linear trend and an annual and semi-annual cycle to the observed tropopause time series of Boering *et al.* [1996].

Figure 2 shows the aircraft CO<sub>2</sub> measurements on February 1996 and October 1994 over the first 100K of potential temperature from the tropical tropopause within 15° of the equator. The solid line is the model solution for the best estimate values from the HALOE and SF<sub>6</sub> data. There is good agreement in the position of the minimum and maximum. The October model profile is a remarkable fit to the measurements, while the February profile is similar in shape to the measurements but shifted by about 1 ppm. There are fewer tropical tropopause measurements over the year preceding February 1996 than during that preceding October 1994, so that the boundary condition is less accurately determined. In general, the model profiles are quite sensitive to the tropopause boundary condition. The observed February profile also show a sharp fall-off above 80K. These data were taken further from the equator than the others, and may represent a recent extra-tropical intrusion.

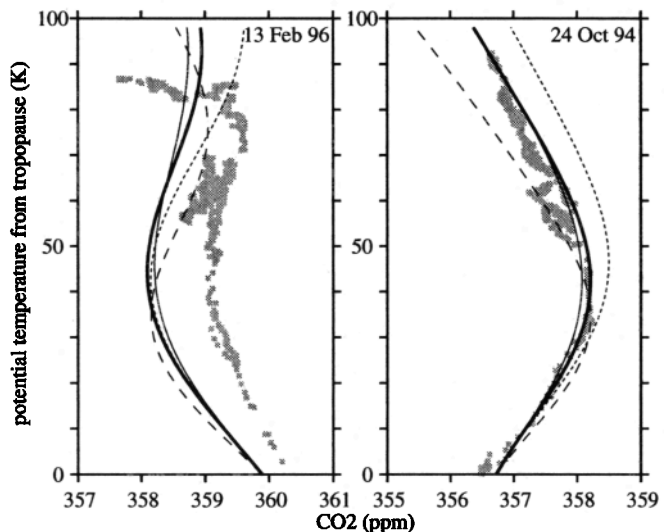
The minima and maxima of the model profiles ascend at different rates, even though there is no variation in the phase velocity, the result of adding an attenuating

sinusoid to a non-constant background [Waugh *et al.*, 1997]. Agreement in the height of profile extrema in seasons of opposite phase, despite the constant coefficients of the model, suggests that observed variations in the ascent rate of the CO<sub>2</sub> cycle [Boering *et al.*, 1996] may not be due entirely to seasonal variations in vertical velocity. For our best estimate values, the February minimum propagates upward from the tropopause about 25% faster than does the October maximum (compared to a 60% difference observed by Boering *et al.* [1996]). Reducing  $\tau$  to 1.0 year results in a 40% difference. Because the minimum versus maximum effect is of comparable magnitude to seasonal  $\bar{w}$  variations, it should be accounted for, if possible, when inferring  $\bar{w}$  from CO<sub>2</sub> observations. In principle, this can be accomplished by removing the CO<sub>2</sub> variation due to the  $\Gamma$  fall-off before tracking the cycle extrema.

To test sensitivities, we computed model CO<sub>2</sub> profiles for other values of  $K_\theta$ ,  $\tau$ , and  $Q$  (Figure 2). Doubling  $\tau$  worsens the agreement with observations, as does a 20% reduction in  $Q$ . On the other hand, tripling  $K_\theta$  has very little effect. This is consistent with the low diffusion limit. Vertical diffusion plays little role in tracer transport, and hence tracer observations provide little information about the rate of diffusion.

## 6. Conclusions

Using satellite  $\hat{H}$  measurements, aircraft SF<sub>6</sub> measurements, and a simple diffusion-advective model of the lower tropical stratosphere that allows relaxation to the extra-tropics, we have separated the roles of vertical diffusion and extra-tropical mixing on trace gas signals



**Figure 2.** Tropical profiles of CO<sub>2</sub> for February 1996 (left panel) and October 1994 (right panel). The symbols are measurements from aircraft. The heavy solid line is the model using our best estimate parameter values. Other curves use best estimates, but with  $K_\theta$  tripled (light solid),  $\tau$  doubled (dotted), and  $Q$  decreased 20% (dashed).

in the tropics. Our best estimate from this analysis is  $K_\theta \approx 0.3 \text{ K}^2/\text{day}$  ( $K_z \approx 0.01 \text{ m}^2/\text{s}$ ),  $\tau \approx 1.3$  years, and  $Q \approx 0.5 \text{ K/day}$  ( $\bar{w} \approx 0.3 \text{ mm/s}$ ). The uncertainty in the measurements, and the limits of such a simple model, imply a large range of equally possible values (at least a factor 3 for  $K_\theta$ ) about this best estimate. However, the estimates of  $\tau$  and  $\bar{w}$  agree well with independent estimates of Volk *et al.* [1996] and Rosenlof [1995], respectively. In addition, these values of  $K_\theta$ ,  $\tau$ , and  $Q$  produce model  $\text{CO}_2$  profiles in good agreement with  $\text{CO}_2$  measurements in the lower stratosphere in different seasons, despite the fact that the model employs constant coefficients.

Our analysis has implications for two- and three-dimensional models of the stratosphere. It is a great challenge for such models simultaneously to simulate accurate values of  $\Gamma$ ,  $A$ , and  $\tau_w$  in the tropics. Hall and Waugh [1997] show two GCMs with greatly different  $\Gamma$  which both significantly overattenuate the annual cycle amplitude. Preliminary analysis similar to that presented here, but applied to model data, suggests that a major problem is too much vertical diffusion in the tropics, perhaps by an order of magnitude. However, once models are in a low diffusion regime, the precise value for  $K_\theta$  is not critical. The condition for achieving this regime may be estimated from the first order correction to the low diffusion limit for the amplitude, and is  $K_\theta \ll Q^2/\omega^2\tau$ , or, for our best estimate values,  $K_\theta \ll 5 \text{ K}^2/\text{day}$  ( $K_z \ll 0.1 \text{ m}^2/\text{s}$ ).

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## References

- Avallone, L. M., and M. J. Prather, Photochemical evolution of ozone in the lower tropical stratosphere, *J. Geophys. Res.*, **101**, 1457–1461, 1996.
- Boering, K. A., et al., Tracer-tracer relationships and lower stratospheric dynamics:  $\text{CO}_2$  and  $\text{N}_2\text{O}$  correlations during SPADÉ, *Geophys. Res. Lett.*, **21**, 2567–2570, 1994.
- Boering, K. A., et al., Stratospheric transport rates and mean age distribution derived from observations of atmospheric  $\text{CO}_2$  and  $\text{N}_2\text{O}$ , *Science*, **274**, 1340–1343, 1996.
- Dessler, A. E., et al., An examination of the total hydrogen budget of the lower stratosphere, *Geophys. Res. Lett.*, **21**, 2563–2566, 1994.
- Elkins, J. W., et al., Airborne gas chromatograph for in situ measurements of long-lived species in the upper troposphere and lower stratosphere, *Geophys. Res. Lett.*, **23**, 347–350, 1996.
- Geller, L. S., et al., Recent trends and latitudinal distribution of tropospheric sulfur hexafluoride, *Geophys. Res. Lett.*, **24**, 675–678, 1997.
- Hall, T. M., and R. A. Plumb, Age as a diagnostic of stratospheric transport, *J. Geophys. Res.*, **99**, 1059–1070, 1994.
- Hall, T. M., and D. W. Waugh, Timescales for the stratospheric circulation derived from tracers, *J. Geophys. Res.*, **102**, 8891–9001, 1997.
- Maiss, M., et al., Sulfur hexafluoride: A powerful new atmospheric tracer, *Atmos. Environ.*, **30**, 1621, 1996.
- Minschwaner, K., et al., The bulk properties of isentropic mixing into the tropics in the lower stratosphere, *J. Geophys. Res.*, **101**, 9433–9439, 1996.
- Mote, P. W., et al., Seasonal variations of water vapor in the tropical lower stratosphere, *Geophys. Res. Lett.*, **22**, 1093–1096, 1995.
- Mote, P. W., et al., An atmospheric tape recorder: the imprint of tropical tropopause temperatures on stratospheric water vapor, *J. Geophys. Res.*, **101**, 3989–4006, 1996.
- Plumb, R. A., A tropical pipe model of stratospheric transport, *J. Geophys. Res.*, **101**, 3957–3972, 1996.
- Randel, W. J., et al., Seasonal cycles and QBO variations in stratospheric  $\text{CH}_4$  and  $\text{H}_2\text{O}$  observed in UARS HALOE data, *J. Atmos. Sci.*, 1997, (in press).
- Rosenlof, K. H., Seasonal cycle of the residual mean circulation in the stratosphere, *J. Geophys. Res.*, **100**, 5173–5191, 1995.
- Schoeberl, M. R., et al., An estimation of the dynamical isolation of the tropical lower stratosphere using UARS wind and trace gas observations of the quasi-biennial oscillation, *Geophys. Res. Lett.*, **24**, 53–56, 1997.
- Trepte, C. R., and M. H. Hitchman, Tropical stratospheric circulation deduced from satellite aerosol data, *Nature*, **355**, 626–628, 1992.
- Volk, C. M., et al., Quantifying transport between the tropical and mid-latitude lower stratosphere, *Science*, **272**, 1763–1768, 1996.
- Volk, C. M., et al., On the evaluation of source gas lifetimes from stratospheric observations, *J. Geophys. Res.*, 1997, (submitted).
- Waugh, D. W., et al., Three-dimensional simulations of long-lived tracers using winds from MACCM2, *J. Geophys. Res.*, 1997, (in press).

CRC for Southern Hemisphere Meteorology, Bldg 70, Monash University, Wellington Rd., Clayton Vic 3168, Australia

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