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Changes in reactive stratospheric gases due to a change in Brewer–Dobson circulation: results from a simple model

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Abstract

Amounts of source gases with stratospheric sinks (CFCs, N₂O, CH₄) are affected by changes in Brewer–Dobson circulation. Source gases and their degradation products are important for atmospheric chemistry and climate. With a simple model, we examine how amounts and lifetimes of source gases and products depend on speed of the circulation. Transient results differ from steady-state and stratospheric results differ from those for stratosphere plus troposphere. Increases in speed increase the stratospheric burden of source gases, but reduce products and reduce total burdens and lifetimes of source gases. Copyright © 2011 Royal Meteorological Society

Keywords: stratosphere; Brewer–Dobson; trend

1. Introduction

The Brewer–Dobson circulation (Brewer, 1949; Dobson, 1956), whereby tropospheric air enters the stratosphere in the tropics, moves towards the winter pole, and returns via tropopause folds, is driven by the breaking of waves against the mean zonal wind in the stratosphere and mesosphere. After entry, air takes about 5 years to reach the polar stratosphere (Boering *et al.*, 1996). The circulation speed will change with planetary and gravity wave intensities or the degree of their filtering, for example, due to increased greenhouse gases, and is predicted to increase over coming decades (Butchart *et al.*, 2006).

Some longer lived trace gases (CFCs, N₂O, CH₄) have sources near the ground, but have sinks in the stratosphere where they photo-oxidise to form reactive products (Cl, NO_x, H₂O). When returned to the troposphere, the product gases are rapidly converted to soluble products (HCl, HNO₃) and washed out by rain. Some source gases (N₂O, CH₄) also have sinks in the troposphere. Changes in speed of the Brewer–Dobson circulation will affect total lifetimes and burden of source gases (Butchart and Scaife, 2001) and their products, and because they are important to atmospheric climate and chemistry it is important to know how they will be affected.

There has been some disagreement as to how an increase in the Brewer–Dobson circulation will affect the stratospheric burdens of products. Some researchers have concluded that with a faster circulation there is greater exchange between troposphere and stratosphere so that more source gases are photo-oxidised at any one time, leading to shorter source lifetimes (Schauffler and Daniel, 1994; Butchart and Scaife, 2001). Others have argued that a slower circulation will lead to increased products because the

source gas has a longer time to photo-oxidise in the stratosphere (Fish *et al.*, 2000).

Simple models are required to obtain a better understanding of how amounts and lifetimes of trace gases are affected by changes in Brewer–Dobson circulation. General circulation models (GCMs) that include interactive chemistry are difficult to use as changes to forcing of the Brewer–Dobson circulation demand changes to initialisation that may change chemistry. Despite their group's access to a GCM, Ray *et al.* (2010) also designed a simplified model to investigate the results of changes in circulation.

Here, we show a simple model of the Brewer–Dobson circulation and trace gases. Its advantages are the few processes involved, the location and strength of the circulation are exactly defined, and there is only one source and product to consider.

2. A simple model of the Brewer–Dobson circulation

The model is shown in Figure 1. The ten stratospheric boxes can be seen as positions in a transect with a circuit of air passing through them, or as time steps in the stratospheric circuit of air. The mass flux is:

$$\text{MassFlux} = M_S/T_S \quad (1)$$

Within each stratosphere box the mass of source gas ($M_S C_{Sx}/10$) is increased each time step ($T_S/10$) by the mass flux from the previous box, and reduced by the mass flux to the next box and by photo-oxidation. A reasonably large number of boxes are required so that the time step is small enough to prevent large jumps in mixing ratio between adjacent boxes, ten being a

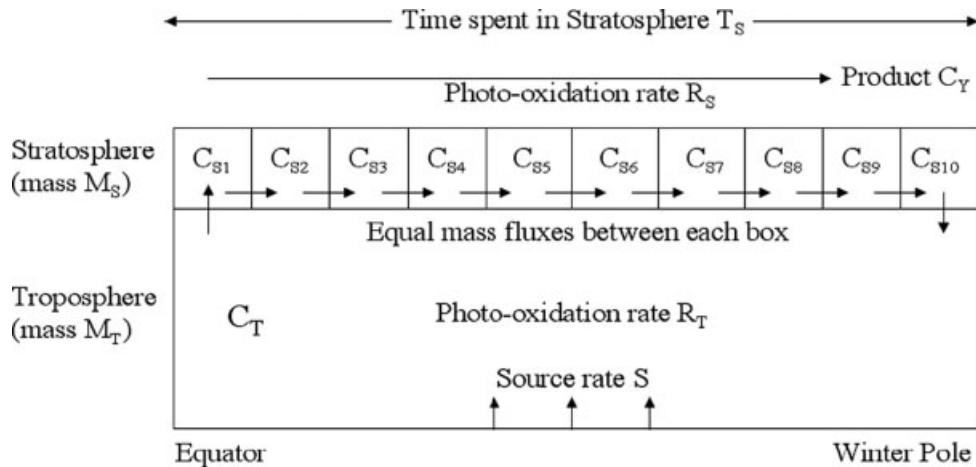


Figure 1. Our simple model of Brewer–Dobson circulation and source gas with stratosphere sinks. The tropical and winter troposphere is a single well-mixed box containing air with source gas mixing ratio C_T , source rate S and decay rate R_T . The stratosphere is represented by ten boxes, each containing air with mass $M_s/10$, with source gas mixing ratios C_{S1} to C_{S10} and decay rate R_S . Circulation is a single mass flux from troposphere to the first stratosphere box, then to each stratosphere box in turn before returning to the troposphere, spending time step $T_S/10$ in each box. The product in the last stratosphere box is C_Y . Product is lost when air re-enters the troposphere but the remaining source gas is added to the tropospheric amount. Mass mixing ratios are used to simplify the mathematics.

good compromise. The source gas is photo-oxidised at a constant rate R_S . Hence for boxes after the first:

$$\begin{aligned}\Delta(M_S C_{Sx}/10) &= (M_S/T_S)C_{Sx-1}(T_S/10) \\ &\quad - (M_S/T_S)C_{Sx}(T_S/10) - (M_S/10)C_{Sx}R_S(T_S/10)\end{aligned}$$

Which reduces to:

$$\Delta C_{Sx} = C_{Sx-1} - C_{Sx} - C_{Sx}R_S T_S/10 \quad (2)$$

The resulting mixing ratio of the product in the polar stratosphere, C_Y , is C_T minus C_{S10} . Products represent a sum whose subdivision is governed by sunlight and temperature (e.g. $\text{NO}_y = \text{N} + \text{NO} + \text{NO}_2 + \text{NO}_3 + 2\text{N}_2\text{O}_5 + \text{ClONO}_2 + \text{BrONO}_2 + \text{HO}_2\text{NO}_2 + \text{HNO}_3$).

$$C_Y = C_T - C_{S10} \quad (3)$$

The tropospheric mass of source gas ($M_T C_T$) is increased each time step by the source S , reduced by air containing the source gas entering the stratosphere (mass flux times mixing ratio), increased by air returning to the troposphere after spending T_S in the stratosphere, and reduced by photo-oxidation in the troposphere with constant rate R_T :

$$\begin{aligned}\Delta(M_T C_T) &= S(T_S/10) - (M_S/T_S)C_T(T_S/10) \\ &\quad + (M_S/T_S)C_{S10}(T_S/10) - M_T C_T R_T(T_S/10) \quad (4)\end{aligned}$$

We could include variations in photo-oxidation with latitude and season, but for annual average global or polar mixing ratios, a mean photo-oxidation rate is adequate and it greatly simplifies the model.

Schauffler and Daniel (1994) developed a similar model, but with only four boxes for the troposphere

plus stratosphere, and including both hemispheres with mass fluxes between them. Our model with 11 boxes and one-way mass flux is more realistic, and has a useful time delay in its response for studying transient effects.

3. The model in steady-state conditions

In steady state, the source S and time T_S are constant, as are source and product mixing ratios, and ΔC_{Sx} and $\Delta M_T C_T$ are zero. The equations can then be solved analytically. Equation (2) reduces to:

$$\frac{C_{Sx}}{C_{Sx-1}} = \frac{1}{1 + R_S T_S/10} \quad (5)$$

Equation (4) to $S = C_T \left[\left(\frac{M_S}{T_S} \right) \left(1 - \frac{C_{S10}}{C_T} \right) + M_T R_T \right]$ and from Equation (5):

$$\frac{C_{S10}}{C_T} = \left(\frac{1}{1 + R_S T_S/10} \right)^{10} \quad (6)$$

Hence:

$$C_T = \frac{S}{\left[\left(\frac{M_S}{T_S} \right) \left(1 - \left(\frac{1}{1 + R_S T_S/10} \right)^{10} \right) + M_T R_T \right]} \quad (7)$$

From Equations (3) and (6):

$$C_Y = C_T \left(1 - \left(\frac{1}{1 + R_S T_S/10} \right)^{10} \right) \quad (8)$$

$$\text{And: } C_S = \frac{1}{10} \left[\frac{C_T}{(1 + R_S T_S / 10)} + \frac{C_T}{(1 + R_S T_S / 10)^2} + \frac{C_T}{(1 + R_S T_S / 10)^3} + \dots + \frac{C_T}{(1 + R_S T_S / 10)^{10}} \right]$$

which is summed to:

$$C_S = \frac{C_T}{R_S T_S} \left(1 - \left(\frac{1}{1 + R_S T_S / 10} \right)^{10} \right) \quad (9)$$

Hence, changing the source produces a linear response in C_T , C_Y and C_S , but the result of changing the time spent in the stratosphere T_S (thereby changing the speed of circulation) is more complex. Increasing T_S reduces the mass flux, but increases the fraction of source photo-oxidised to stratospheric product.

While the mass flux has a linear response to changes in T_S , the fraction of the source photo-oxidised follows an exponential decay and the response is less than linear, so doubling the time spent in the stratosphere does not lead to double the source being photo-oxidised. Therefore, increasing T_S increases C_T and C_Y but reduces C_S , and C_Y has the greatest response to changes in T_S .

The total burden (B) of source is:

$$B = M_T C_T + M_S C_S \quad (10)$$

The total lifetime (L) of the source, using Equations (6), (9) and (10), is:

$$L = \frac{B}{S} = \frac{M_T C_T + M_S C_S}{S} = \frac{M_T + \left(\frac{M_S}{T_S} \right) \frac{1}{R_S} \left(1 - \left(\frac{1}{1 + R_S T_S / 10} \right)^{10} \right)}{M_T R_T + \left(\frac{M_S}{T_S} \right) \left(1 - \left(\frac{1}{1 + R_S T_S / 10} \right)^{10} \right)} \quad (11)$$

The fraction of stratospheric photo-oxidation (F_S), using Equation (9), is:

$$F_S = \frac{M_S C_S R_S}{M_T C_T R_T + M_S C_S R_S} = \frac{\frac{M_S}{T_S} \left(1 - \left(\frac{1}{1 + R_S T_S / 10} \right)^{10} \right)}{M_T R_T + \frac{M_S}{T_S} \left(1 - \left(\frac{1}{1 + R_S T_S / 10} \right)^{10} \right)} \quad (12)$$

4. Examining different trace gases

We used the above steady-state equations to find the response of three separate source gases and their products to changes in circulation, with the ratio of tropospheric to stratospheric masses set to 10 – the global mean pressure at the tropopause gives about this value, and Schauffler and Daniel (1994) use 9. Details of the gases are given in Table I.

Results in Table II show that reducing T_S (increasing the speed) reduces C_T , C_Y , B and L , but increases C_S and F_S . With a faster circulation the mixing between the troposphere and stratosphere is more efficient and C_T and C_S are more similar, and with increased C_S photo-oxidation is greater. Although products are forming faster, C_Y is reduced because the greater mass flux cleans products out of the stratosphere faster than they are formed. Hence, increasing the speed of circulation does reduce the total burdens and lifetimes of source gases (consistent with Schauffler and Daniel 1994; Butchart and Scaife, 2001), but also reduces the products including that of NOy (consistent with Fish *et al.*, 2000).

For CFC-12, which is entirely photo-oxidised in the stratosphere, C_T , C_Y , B and L all show large responses to changes in speed, C_Y has a near-linear response, but C_S is unchanged as stratospheric photo-oxidation must balance the source S . For CH₄, only partly photo-oxidised in the stratosphere, the responses of C_T , B and L are very small, but C_S , F_S and C_Y have large responses. Responses for N₂O are between those for CFC-12 and CH₄.

Table I. The observed tropospheric mixing ratios of source gases, total lifetimes, where photo-oxidised, and products.

Source gas	Tropospheric volume mixing ratios	Tropospheric mass mixing ratios ^a	Total lifetime	Where photo-oxidised	Product gas
CFC-12 (molecular weight of Cl = 71)	540 pptv (WMO, 2007)	1.33 ppbm of Cl	100 years (WMO, 2007)	All in stratosphere	Cly
N ₂ O (molecular weight of N = 28)	319 ppbv (WMO, 2007)	309 ppbm of N	120 years (WMO, 2007)	Mostly in stratosphere	NOy
CH ₄ (molecular weight of H = 4)	1777 ppbv (WMO, 2007)	246 ppbm of H	8 years (Karlsdottir and Isaksen, 2000)	Mostly in troposphere	H ₂ O

^a volume mixing ratio x molecular weight/28.9

Table II. Steady-state results for different circulation speeds (different T_S).

Trace gas	M_T	R_T	M_S	R_S	S	T_S	C_T	C_S	C_Y	B	L	F_S
CFC-12	10	0	1	0.1585	0.142	7	1.53	0.90	0.99	16.16	113.9	1
						5	1.33	0.90	0.71	14.19	100.0	1
						3	1.15	0.90	0.43	12.35	87.0	1
						40.0	14.3	0.0	39.4	13.4	13.5	0.0
N_2O	10	0.001	1	0.1043	27.7	7	337	234	170	3604	130.0	0.878
						5	309	236	123	3326	120.0	0.889
						3	282	239	75	3056	110.3	0.898
						40.0	8.9	1.1	38.6	8.2	8.2	1.1
CH_4	10	0.1227	1	0.1585	328	7	249	146	162	2632	8.02	0.070
						5	246	166	131	2626	8.00	0.080
						3	243	190	90	2619	7.98	0.092
						40.0	1.2	13.3	27.5	0.2	0.3	13.8

Defined are: masses of air in troposphere and stratosphere (M_T , M_S , arbitrary units), photo-oxidation rates (R_T , R_S , fraction/year), source (S , mass units/year), time spent in stratosphere (T_S , years). Results are: mean mass mixing ratios of source (C_T , C_S) and stratospheric product (C_Y , all ppbm of Cl, N, H), burden (B, mass units), lifetime (L, years), fraction of photo-oxidation taking place in the stratosphere (F_S), size of changes when varying T_S from 5 years (%), in bold). Photo-oxidation rates and source terms are set to give the observed lifetimes and tropospheric mixing ratios at $T_S = 5$ years.

For some source gases only a fraction of the photo-oxidised products are reactive – only 6% of N_2O loss becomes NOy , the remainder becoming N_2 . Mixing ratios and burdens of products would be reduced by the same fraction (by 0.06 for NOy). This does not affect the amount of source lost within each box as the nature of the product has no influence on the source, so mixing ratio and burden of source are unchanged. Only the amount of product needs to be reduced, and the accumulation of product in the final box is reduced by the same fraction. Relative changes in mixing ratio, and burden of reactive products between different scenarios, are unaffected. Factors other than the circulation speed can also change NOy – the trend in emission of N_2O to the troposphere, and the temperature-dependent loss rate of NOy (Rosenfeld and Douglass, 1998).

Our model necessarily excludes mixing of air parcels that have travelled different routes through the stratosphere. But because mixing is a linear process,

and because mass is conserved in our system, the results would be the same if air parcels travelling routes with different transit times, but the same average transit time, were mixed.

5. The model in changing conditions

Equations (1)–(4) can be cast in a model to be run as in Figure 1 to make transient calculations. As an example, we started the model for CFC-12 at steady state with T_S of 5 years with other parameters as in Table II.

Figure 2 shows the results. When T_S is reduced the C_S initially increase, reducing the differences between them as there is less time for photo-oxidation within each box, although changes take time to propagate through to all the boxes. Then C_T declines due to the greater mass flux, until the increase in C_{S10} increases the CFC-12 returning to the troposphere, to give a

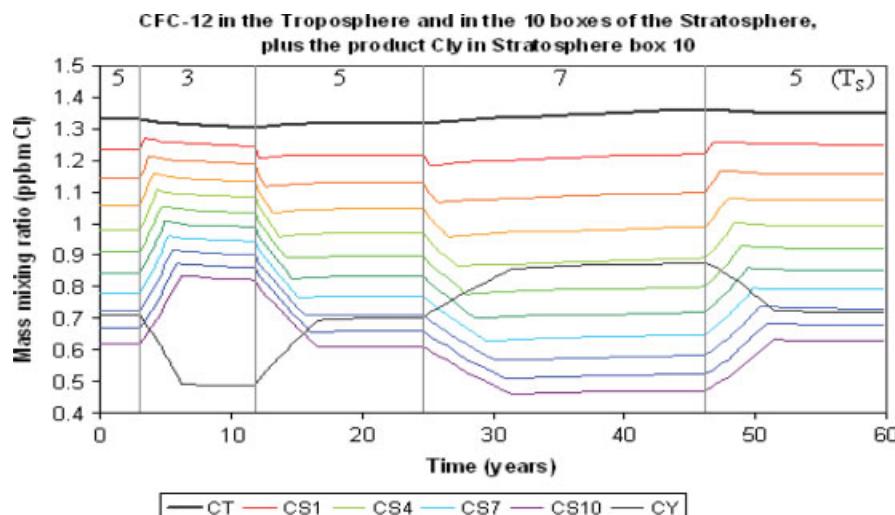


Figure 2. Mixing ratios (ppbm of Cl) of CFC-12 in the troposphere box (C_T) and in the stratosphere boxes (C_{S1} to C_{S10}), and of the product C_Y in the final stratosphere box (C_Y) during the model run. Steady-state values are given in Table II. The differences between the values in adjacent boxes show the exponential decay as CFC-12 is photo-oxidised within each stratosphere box.

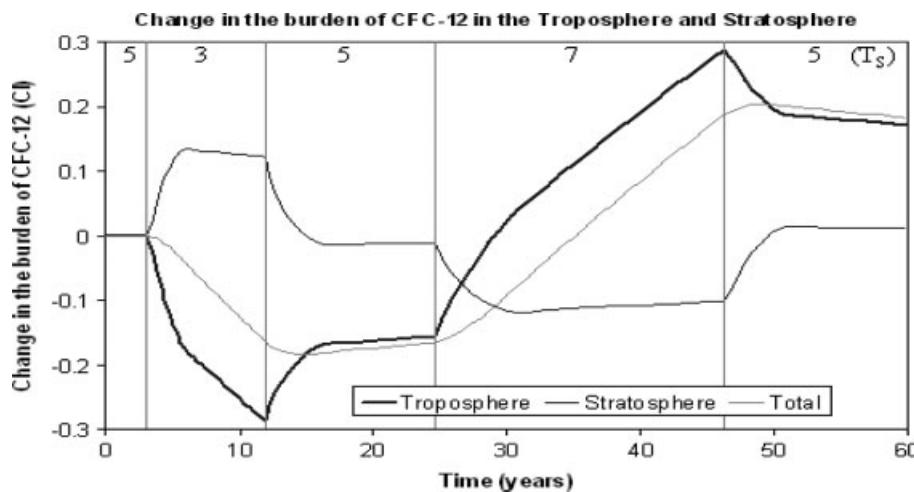


Figure 3. Changes in burdens of CFC-12 during the model run. Steady-state burdens are given in Table II.

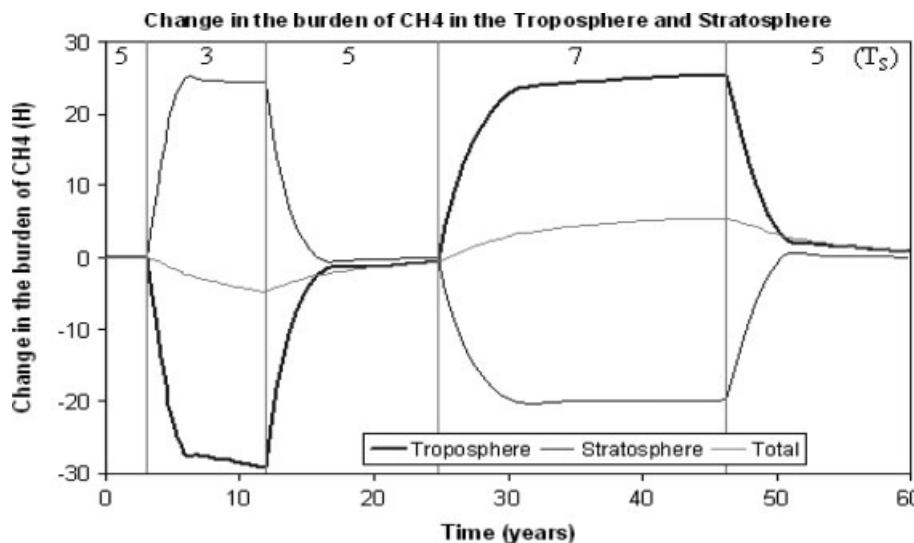


Figure 4. Changes in burdens of CH₄ during the model run. Steady-state burdens are given in Table II.

new balance with a reduced tropospheric but increased stratospheric burden of CFC-12. This also leads to slight reductions in C_{S1} to C_{S10} . There is then a slow decline in all mixing ratios as CFC-12 is now photo-oxidised at a faster rate overall. C_T would eventually reach a new steady-state value (Table II), but the response is slow due to the large mass of air in the troposphere – only 11% of the way to its final value after time T_S .

The mixing ratio of the products C_{LY} (C_Y) responds at the same rate as C_{S10} to changes in T_S . Reducing T_S results in a reduction in C_{LY} as less CFC-12 is photo-oxidised, then a slow decline as all mixing ratios and the total burden are reduced. C_{LY} would eventually reach a new steady state (Table II) but the response is much quicker than for C_T and it is 78% of the way there after time T_S . With the greater mass flux through the stratosphere more C_{LY} is being produced, but it is then cleaned out faster so that the mixing ratio is reduced. When T_S is restored there is a general reversal, and an inverted pattern is seen.

Figure 3 shows the changes in burdens. Reducing T_S first reduces tropospheric CFC-12 but increases it in the stratosphere due to the greater mass flux, then the burden slowly declines in both. CFC-12 is photo-oxidised at a faster rate overall as a larger fraction of the CFC-12 is in the stratosphere, but this is more noticeable in the troposphere as it contains most of the air. The burden would eventually reach a new steady state (Table II). Increasing T_S produces a similar but inverted pattern.

The model was also run with N₂O and CH₄. Results for N₂O, mostly photo-oxidised in the stratosphere, are very similar to those for CFC-12. Results for CH₄, mostly photo-oxidised in the troposphere, have significant differences, shown in Figure 4. The tropospheric mixing ratio of CH₄ and its total burden do not change as much as CFC-12, but with the much shorter total lifetime, the source and product mixing ratios and total burden respond more quickly to changes in speed. After time T_S , C_T is over 88% of the way to the new steady state, and C_Y is over 94% of the way.

6. Conclusions

Changes in Brewer–Dobson circulation will change total atmospheric burdens of trace gases and their lifetimes. The results from this simple model imply that, with an increased circulation, a larger fraction of source gas resides in the stratosphere, so its photo-oxidation rate is increased thereby reducing the total burden and lifetime (consistent with Schauffler and Daniel, 1994). An increased circulation leads to CFCs being removed from the atmosphere at a faster rate (consistent with Butchart and Scaife, 2001).

An increased circulation also results in the products being formed at a faster rate, but with the increased mass flux the products are cleaned out of the stratosphere at an even faster rate, so their mixing ratios and stratospheric burdens are reduced (consistent with Fish *et al.*, 2000). Hence a faster circulation results in less stratospheric NO_y (consistent with Roscoe, 2004). The stratospheric mixing ratios of the products are reduced more than the tropospheric mixing ratios of the source, and with a quicker response.

For CH₄, an increased circulation results in only a small reduction in burden and lifetime, although its short lifetime ensures rapid responses.

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