Tellus (1982), *34*, 398–405

The partitioning of excess CO_2 in a five-reservoir atmosphere–ocean model

By A. J. CRANE, *Central Electricity Research Laboratories, Kelvin Avenue, Leatherhead, Surrey KT22 7SE, England*

(Manuscript received July 14; in final form December 21, 1981)

ABSTRACT

The two-box-diffusion model of the oceans developed by Oeschger et al. (1975) is extended to include a well-mixed, deep polar reservoir in contact with the atmosphere and a split diffusive deep ocean comprising two reservoirs with characteristics akin to the Atlantic and Pacific oceans. It is shown that the division of the deep ocean has little influence on the airborne fraction of fossil fuel CO_2. The partial or complete replacement of the diffusive Atlantic reservoir with the well-mixed polar reservoir significantly reduces the airborne fraction, although it still remains above the observed value for the period 1959–78. The sensitivity of the airborne fraction to buffer factor and fossil fuel CO_2 emission rate is seen to be large enough to limit the usefulness, as predictive tools, of linear models in which these parameters are held constant.

1. Introduction

The fraction of fossil fuel CO_2 emitted during the industrial era that has remained airborne is poorly known owing chiefly to uncertainties in the biospheric CO_2 source and in the preindustrial concentration. Oeschger and Heimann (1981) conclude that for the period 1956–1978 the "theoretical" airborne fraction, i.e. the increase in atmospheric CO_2 between 1956 and 1978 due solely to fossil fuel consumption divided by the fossil fuel CO_2 input during that period, should lie somewhere in the range 38 to 67%, depending on the influence upon the observed change in atmospheric CO_2 concentration of biospheric changes during and prior to the observation period. The "apparent" airborne fraction, i.e. the observed change in atmospheric CO_2 due to all sources and sinks divided by the fossil fuel CO_2 input, was 55% at Mauna Loa and rather less than 51% at the South Pole for the period 1959–78 (Bacastow and Keeling, 1981).

When realistically calibrated, the various linear box models, box-diffusion models and vertical diffusion-advection models which have been used to estimate the oceanic uptake of fossil fuel CO_2 (e.g. Craig, 1957, Keeling, 1973, Oeschger et al., 1975, Bacastow and Björkström, 1981) predict airborne fractions considerably higher than that observed in the period 1959–78, very near the top extreme of the range quoted above. It is desirable, therefore, to investigate whether models simulating oceanic processes more realistically are capable of producing airborne fractions closer to the middle of the range.

Two unrealistic features of the simpler models are the neglect of convective mixing in high latitudes, where deep water outcrops at the surface, and the treatment of the world's deep oceans as a single reservoir. The present paper describes an adaptation of the box-diffusion model of Oeschger et al. (1975) which includes a deep polar reservoir and a split diffusive deep ocean, and examines the sensitivity of the airborne fraction principally to these features of the model. During the preparation of this paper the author learnt of the outcrop-diffusion model developed by Siegenthaler (1981) with which useful comparisons may be drawn.

2. The five-reservoir model

The component reservoirs of the model are shown in Fig. 1. It comprises a well-mixed

0040-2826/82/040398-08$02.50/0

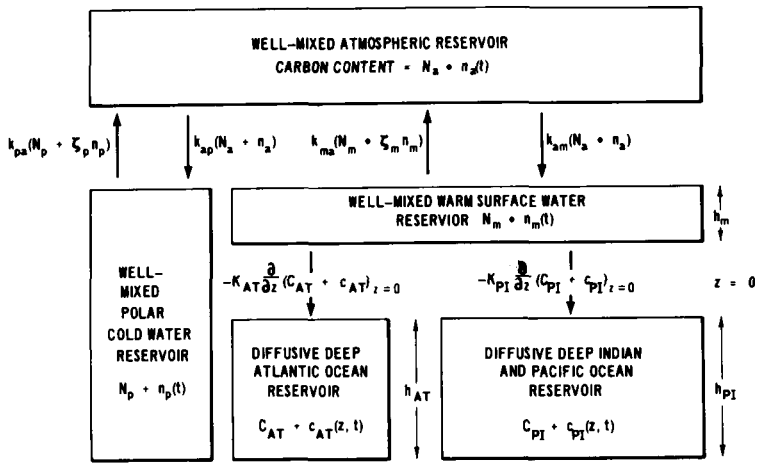

Fig. 1. Definition sketch of the five-reservoir atmosphere–ocean model. Exchanges of carbon between reservoirs indicated by arrows. Nomenclature is given in Table 1.

atmosphere interfaced with a well-mixed warm surface water reservoir, 75 m deep and covering the temperate and tropical oceans, and a polar well-mixed reservoir, of total ocean depth and variable surface area, representing regions where convective mixing with the thermocline and deep waters occurs in high latitudes. Beneath the warm surface reservoir are two parallel diffusive reservoirs representing the deep Atlantic and the deep Pacific and Indian oceans. Since interest is focused on the oceanic uptake of carbon no biospheric reservoir is included, the implication being that the biosphere is regarded as neither a sink for fossil fuel CO_2 nor an additional source of CO_2. The most unrealistic feature of the model is the lack of horizontal interaction between the oceanic reservoirs. It has long been considered that lateral mixing plays a major role in the distribution of oceanic tracers (e.g. Montgomery, 1936). The effect of its omission is considered briefly in the discussion of the results.

The equations which describe the rate of change of carbon in each reservoir are natural extensions of those presented by Oeschger et al. (1975):

$$\frac{dn_a}{dt} = \gamma_f(t) + k_{pa}(N_p + \xi_p n_p) - k_{ap}(N_a + n_a) + k_{ma}(N_m + \xi_m n_m) - k_{am}(N_a + n_a) \quad (1)$$

$$\frac{dn_p}{dt} = -k_{pa}(N_p + \xi_p n_p) + k_{ap}(N_a + n_a) \quad (2)$$

$$\frac{dn_m}{dt} = -k_{ma}(N_m + \xi_m n_m) + k_{am}(N_a + n_a) + A_{AT} K_{AT} \left.\frac{\partial c_{AT}}{\partial z}\right|_{z=0} + A_{PI} K_{PI} \left.\frac{\partial c_{PI}}{\partial z}\right|_{z=0} \quad (3)$$

$$\frac{\partial c_i}{\partial t} = K_i \frac{\partial^2 c_i}{\partial z^2}, \quad i = \text{AT, PI} \quad (4)$$

The deep sea boundary conditions are

$$C_i + c_i|_{z=0} = \frac{N_m + n_m}{A_m h_m}, \quad i = \text{AT, PI} \quad (5)$$

$$K_i \frac{\partial}{\partial z}(C_i + c_i)|_{z=h_i} = 0, \quad i = \text{AT, PI} \quad (6)$$

The notation, which conforms to the standard recommended by Bolin (1981), is explained in Table 1, which also gives numerical values of constants.

The diffusivities K_{AT} and K_{PI} were evaluated by the method of Oeschger et al. (1975). Assuming uniform pre-industrial carbon concentration, the ^{14}C balance for each deep reservoir is given by:

$$R_i(z) = R_m \frac{\cosh\left[\left(\frac{\lambda}{K_i}\right)^{1/2}(h_i - z)\right]}{\cosh\left[\left(\frac{\lambda}{K_i}\right)^{1/2} h_i\right]}, \quad i = \text{AT, PI} \quad (7)$$

Table 1. *Nomenclature and values of constants used in the five-reservoir model. Values in brackets indicate changed values for sensitivity studies (See Tables 2 and 3)*

Symbol	Meaning	Value
N_a	Pre-industrial carbon content of atmospheric reservoir	6.156×10^{17} gC
N_p	Pre-industrial carbon content of polar reservoir	variable
N_m	Pre-industrial carbon content of warm surface reservoir	variable
$n_i(t)$	Departure of carbon content of reservoir i from pre-industrial state	
C_i	Pre-industrial carbon concentration in diffusive reservoir i	$N_m/(A_m h_m)$
$c_i(z,t)$	Departure from C_i of carbon concentration in diffusive reservoir i	
h_a	Depth of complete ocean surface layer containing as much carbon as pre-industrial atmosphere	69 m
h_m	Depth of warm surface reservoir	75 m
h_{AT}, h_{PI}	Depth of deep Atlantic and Pacific reservoirs	3725 m
h_p	Depth of polar reservoir	3800 m
z	Depth beneath warm surface—deep ocean boundary	
k_{ij}	Exchange coefficient reservoir i to reservoir j	
e	Ratio of CO_2 exchange rate in polar reservoir to that in warm surface reservoir	1.0 (1.7)
ξ_m	Buffer factor for warm surface reservoir	9 (12)
ξ_p	Buffer factor for polar reservoir	14 (16)
λ	^{14}C decay constant	1/8267 a
μ	Reciprocal of fossil fuel CO_2 input time constant	1/22.5 a (1/34.5 a)
γ_f	Fossil fuel CO_2 input function	
r_i	Fraction of fossil fuel input residing in reservoir i	
R_i	Average ^{14}C content of reservoir i relative to atmosphere ($R_a = 1$)	
K_{AT}	Eddy diffusivity for deep Atlantic reservoir	12590 $m^2 a^{-1}$
K_{PI}	Eddy diffusivity for deep Pacific and Indian reservoir	2417 (3500, 2204) $m^2 a^{-1}$
A_{oc}	Total surface area of world oceans	3.62×10^{14} m^2
A_p	Surface area of polar reservoir	variable
A_m	Surface area of warm surface reservoir	$A_{oc} - A_p$
A_{PI}	Horizontal area of deep Pacific and Indian reservoir	$0.8A_{oc}$
A_{AT}	Horizontal area of deep Atlantic reservoir	$0.2A_{oc} - A_p$

Equation (7) was fitted to the Atlantic and Pacific ^{14}C data used by Oeschger et al. (1975) as shown in Fig. 2. R_m is taken as 0.95. Values of $K_{AT} = 12590$ m^2 a^{-1} and $K_{PI} = 2417$ m^2 a^{-1} were adopted. The fit to the Pacific data was intended to model the upper layers of the deep ocean since interest lies in the fossil fuel uptake over one or two centuries. The higher ^{14}C concentrations in the deeper layers suggest that the ^{14}C pathway is not solely via vertical diffusion. The above values of K_{AT} and K_{PI} yield depth-averaged ^{14}C concentrations $\overline{R_{AT}}$ and $\overline{R_{PI}}$ of 0.91 and 0.78 respectively.

The warm surface water–atmosphere exchange coefficient, k_{ma}, was evaluated, for specified values of A_{AT} and A_{PI}, from the ^{14}C balance of the warm surface reservoir:

$$\frac{d}{dt} N_m R_m = k_{am} N_a R_a - k_{ma} N_m R_m - \lambda N_m R_m + A_{AT} K_{AT} C_{AT} \left.\frac{\partial R_{AT}}{\partial z}\right|_{z=0} + A_{PI} K_{PI} C_{PI} \left.\frac{\partial R_{PI}}{\partial z}\right|_{z=0} \quad (8)$$

noting that

$$k_{am} = \frac{N_m}{N_a} k_{ma} \quad (9)$$

The remaining coefficients were determined from the steady state relation

$$k_{pa} = \frac{N_a}{N_p} k_{ap} \quad (10)$$

and the relation

$$k_{ap} = e \frac{A_p}{A_m} k_{am} \quad (11)$$

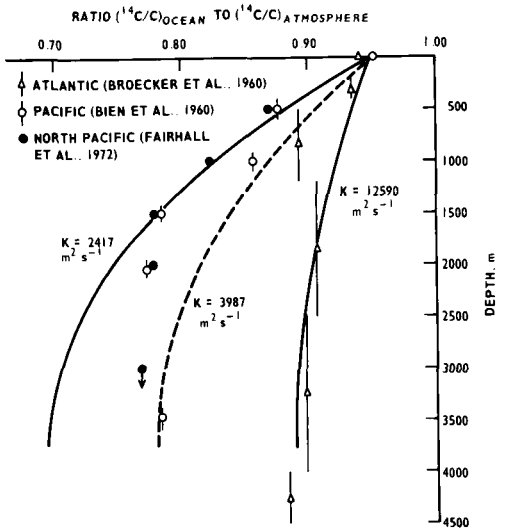

Fig. 2. Pre-bomb ^{14}C distribution in Atlantic and Pacific oceans. Solid curves are obtained from eq. (7) in text using values of K shown. Dashed line shows the curve obtained using the value of K adopted by Oeschger et al. for a single diffusive reservoir. The figure is based on that presented by Oeschger et al. (1975).

where e is the ratio of the air–sea exchange rate in polar waters to that in the warm surface waters.

GEOSECS data indicate that an annual pre-industrial CO_2 concentration of 290 p.p.m. ($N_a = 6.156 \times 10^{17}$ gC) is equivalent to the carbon content of a mean surface ocean layer of 69 m depth (h_a) (Bacastow and Björkström 1981).[1] Thus when the surface area of the polar reservoir is zero in the present model $N_m/N_a = h_m/h_a$. When the warm mixed layer occupies an area less than that of the total ocean surface, N_m takes correspondingly smaller values. In calculating N_p, the carbon concentration in the polar reservoir is assumed to be 2.26 mol m^{-3}, the value given by Broecker (1974) for North Atlantic Deep Water.

Following Oeschger et al. (1975) eqs. (1)–(6) may be reformulated in terms of the fractional change in carbon content relative to pre-industrial values. Thus

$$n'_i = n_i/N_i, \quad i = a, p, m,$$

$$c'_i = c_i/C_i, \quad i = \mathrm{AT}, \mathrm{PI}$$

An exponential fossil fuel input $\gamma_f(t)$ is expressed as

$$\gamma'_f(t) = \gamma_f(t)/N_a = \gamma'_{f0} \exp \mu t$$

whence the cumulative input at time t approximates $N_a\gamma'_f/\mu$ once γ'_f becomes much larger than γ'_{f0}. With an exponential input function, the differential equations for the fractional change in carbon content reduce to a set of algebraic equations:

$$\mu n'_{a0} = \gamma'_{f0} + k_{ap}(\xi_p n'_{p0} - n'_{a0}) + k_{am}(\xi_m n'_{m0} - n'_{a0}) \tag{12}$$

$$\mu n'_{p0} = k_{pa}(n'_{a0} - \xi_p n'_{p0}) \tag{13}$$

$$\mu n'_{m0} = k_{ma}(n'_{a0} - \xi_m n'_{m0}) - n'_{m0}(\theta_{\mathrm{AT}} + \theta_{\mathrm{PI}}) \tag{14}$$

where

$$\theta_i = \frac{A_i}{A_m h_m}(\mu K_i)^{1/2} \tanh\left[\left(\frac{\mu}{K_i}\right)^{1/2} h_i\right], \quad i = \mathrm{AT}, \mathrm{PI}$$

$$\mu c'_i(z) = K_i \frac{\partial^2 c'_i}{\partial z^2}, \; i = \mathrm{AT}, \mathrm{PI} \tag{15}$$

The airborne fraction, r_a, is given by

$$r_a = n_a/(N_a\,\gamma'_f/\mu) = n'_{a0}\,\mu/\gamma'_{f0} \tag{16}$$

From eqs. (12)–(14) and (16) are obtained the relations

$$r_a = \mu\left[\mu\left(1 + \frac{k_{ap}}{\mu + k_{pa}\,\xi_p}\right) + \frac{k_{am}(\mu + \theta_{\mathrm{AT}} + \theta_{\mathrm{PI}})}{\mu + k_{ma}\,\xi_m + \theta_{\mathrm{AT}} + \theta_{\mathrm{PI}}}\right]^{-1} \tag{17}$$

which, for convenience is written as

$$r_a = \mu[\mu(1 + X) + Y(\mu + \theta_{\mathrm{AT}} + \theta_{\mathrm{PI}})]^{-1},$$

and the fractions of the fossil fuel carbon input that reside in the polar and warm surface reservoirs:

$$r_p = Xr_a \tag{18}$$

$$r_m = Yr_a \tag{19}$$

The solution of eq. (15) yields:

$$c'_i(z) = n'_m \frac{\cosh[(\mu/K_i)^{1/2}(h_i - z)]}{\cosh[(\mu/K_i)^{1/2} h_i]}, \quad i = \mathrm{AT}, \mathrm{PI}$$

$$\simeq n'_m \exp[-(\mu/K_i)^{1/2} z] \tag{20}$$

when $(\mu/K_i)^{1/2} h_i \gg 1$, which condition holds for the

[1] Oeschger et al. (1975) used $h_a = 58$ m in error (see Siegenthaler, 1981) and obtained considerably smaller airborne fractions as a result.

values of μ and K_i used. Integrating the concentration over the depth of the deep ocean reservoirs to obtain their uptake of carbon leads to deep ocean fractions

$$r_i = \frac{A_i}{A_m h_m}\left(\frac{K_i}{\mu}\right)^{1/2} r_m \quad i = \mathrm{AT, PI} \tag{21}$$

Note that for exponential input functions, the fractions taken up by each reservoir become constant in time once the input rate greatly exceeds the initial constant ($\gamma'_f \gg \gamma'_{f0}$).

3. Results

The sensitivity of the carbon partitioning to variations in the model parameters was examined. Table 2 shows the parameter values adopted in each case. Case (c) is regarded as a standard and only where parameter values differ from those of case (c) are they included. Values for ξ_m and ξ_p of 9 and 14 respectively are consistent with the GEOSECS results presented by Sundquist et al. (1979) and were used by Siegenthaler (1981). Following Bacastow and Björkström (1981) an e-folding time of 22.5 years (μ^{-1}) is used for the fossil fuel carbon input, a rate which agrees closely with the growth since 1945 and which is therefore appropriate for a comparison of the model airborne fraction with the observed airborne fraction since 1959.

Table 3 lists the reservoir fractions for each case together with the parameter variations (from case (c)) imposed. Changes consequential upon these variations are only shown in Table 2.

Case (a) is a pure box-diffusion case and yields an airborne fraction of 67.7%. The choice of K_{PI} to fit the ^{14}C observations in the upper 1500 m of the deep Pacific means that the average ^{14}C value for the two deep reservoirs is 0.81, significantly less than the value 0.84 preferred by Bacastow and Björkström (1981) and Siegenthaler (1981) on the basis of observations throughout the whole depth of the ocean. To obtain a mean value of 0.84, K_{PI} was increased in case (b) to 3500 $m^2\ a^{-1}$. The corresponding airborne fraction was 65.8%. This agrees well with the equivalent fractions of 66.4% obtained by Bacastow and Björkström (1981) employing a slightly larger buffer factor (9.64) and 66.7% obtained by Siegenthaler (1981). The measure of agreement indicates that the separate treatment of the deep ocean reservoirs does not significantly influence the airborne fraction. Comparing cases (a) and (b) also suggests that the airborne fraction is relatively insensitive to the precise value of the deep ocean ^{14}C content in a pure box-diffusion model.

Cases (c) and (d) introduce the polar well-mixed reservoir, covering 10% and 20% of the total oceanic area respectively. Correspondingly, the area of the deep Atlantic reservoir is reduced from $0.2A_{oc}$ in cases (a) and (b) to $0.1A_{oc}$ and zero in cases (c) and (d). The airborne fractions fall to 64.3% (case (c)) and 60.5% (case (d)). It is

Table 2. *Variation of parameter values from those of standard case (c) for cases (a)–(i)*

Parameter \ Case	a	b	c	d	e	f	g	h	i
A_p/A_{oc}	0	0	0.1	0.2		0.2	0.2		
A_{AT}/A_{oc}	0.2	0.2	0.1	0		0	0		
A_m/A_{oc}	1.0	1.0	0.9	0.8		0.8	0.8		
ξ_m			9					12	
ξ_p	—		14					16	
μ^{-1} (a)			22.5						34.5
e			1.0		1.70	1.70	1.73		
k_{ma}^{-1} (a)	10.11	9.69	10.25	10.44		10.44	10.61		
k_{am}^{-1} (a)	9.30	8.91	10.48	11.52		11.52	11.71		
k_{pa}^{-1} (a)	—	—	572.27	559.15	336.63	328.91	328.91		
k_{ap}^{-1} (a)	—	—	94.35	46.09	55.50	27.11	27.11		
K_{PI} ($m^2\ a^{-1}$)		3500	2417				2204		
K_{AT} ($m^2\ a^{-1}$)			12590						

Table 3. *Fossil fuel carbon partitioning* (%) *for cases* (*a*)–(*i*) *detailed in Table 2. Imposed departures from case* (c) *are given; consequential changes in other parameters are given in Table 2*

Case	Imposed variation from case (c)	r_a	r_m	r_p	r_{AT}	r_{PI}
a	$A_p = 0$	67.7	6.6	—	9.3	16.3
b	$A_p = 0, K_{PI} = 3500$	65.8	6.3	—	9.0	18.9
c		64.3	5.7	9.9	4.5	15.7
d	$A_p = 0.2A_{oc}$	60.5	5.0	18.9	—	15.6
e	$e = 1.7$	62.1	5.5	13.0	4.3	15.2
f	$A_p = 0.2A_{oc}, e = 1.70$	56.6	4.7	24.0	—	14.6
g	$A_p = 0.2A_{oc}, K_{PI} = 2204, e = 1.73$	57.0	4.8	24.2	—	14.1
h	$\xi_p = 16, \xi_m = 12$	68.4	4.8	10.0	3.7	13.1
i	$\mu^{-1} = 34.5$	59.0	5.4	11.7	5.3	18.6

apparent that a well-mixed deep reservoir in contact with the atmosphere is a more efficient abstractor of carbon than the sub-surface diffusive reservoir which it replaces. Moreover, if the air–sea exchange rate for polar waters is reckoned to be 1.7 times faster than for the warm surface waters (Siegenthaler, 1981), the fractions fall to 62.1% and 56.6% for $A_p = 0.1A_{oc}$ and $A_p = 0.2A_{oc}$ respectively (cases (e) and (f)). These results must be interpreted with caution, however. The introduction of the polar reservoir leaves the uptake by the warm surface and the Pacific and Indian reservoirs little altered apart from the effect of small changes in the exchange coefficients consequent upon the changed configuration of the model. However, the average ^{14}C content of the mixed polar reservoir, calculated from k_{pa}, is 0.94 in cases (c) and (d) and 0.96 in cases (e) and (f) compared with 0.91 for the deep Atlantic diffusive reservoir it replaced. The average ^{14}C content of the whole deep ocean therefore increases, which suggests that the calculated fall in airborne fraction may be exaggerated. Thus in case (g) K_{PI} was reduced from 2417 to 2204 $m^2\ a^{-1}$ ($A_p = 0.2A_{oc}$) which reduces the ^{14}C content of the diffusive reservoir sufficiently to compensate exactly for the increase in the polar reservoir. Maintaining the same polar exchange coefficients (now equivalent to an exchange rate 1.73 times faster than for the warm mixed layer due to small changes in the warm water exchange coefficients consequent upon the reduction of K_{PI}) the airborne fraction is only slightly increased to 57.0%. The conclusions drawn above regarding the effectiveness of the polar reservoir thus remain qualitatively unaltered.

Siegenthaler's (1981) outcrop-diffusion model incorporated infinitely rapid horizontal mixing across the deep ocean of carbon entering via the deep sea outcrop. No separate polar reservoir was included. This enabled him to calibrate the model in a more consistent manner than adopted here, the exchange coefficients and diffusivity being determined for each value of the outcrop area such that the surface and deep ocean ^{14}C contents remained unchanged. For outcrop areas of $0.1A_{oc}$ and $0.2A_{oc}$ he obtained airborne fractions of 60.0% and 58.2%, with little further reduction for greater outcrop areas. In the present model the reduction of airborne fraction over the range $0 < A_p < 0.2A_{oc}$ is more nearly linear, and the effect of the different representations of sub-surface mixing in the two models is evident. In reality, the degree of horizontal mixing presumably lies somewhere between the extremes represented by these models. Nevertheless the qualitative agreement of the results of the two models gives confidence in the conclusions drawn.

As CO_2 concentrations rise the capacity of the oceans to take up excess CO_2 will decrease (Keeling, 1973), in the absence of any mitigating processes. Our linear model cannot simulate a time-dependent buffer factor, but it is of interest to examine the partitioning for higher values of the buffer factor. If a climatic warming occurs at high latitudes which is significantly greater than in the tropics, the approximate values for the buffer factor for an atmospheric CO_2 concentration of 600 p.p.m. are around 12 for warm surface waters and 16 for the polar reservoir (based on Fig. 5 of Sundquist and Plummer, 1981).

These values (case h) yield an airborne fraction of 68.4% compared with 64.3% in case (c). The airborne fraction is seen to be sufficiently sensitive to a moderate change in the buffer factor to suggest that the future behaviour of the carbon cycle will not be reliably predicted using a linear model of the present type.

While a fossil fuel carbon input time constant of 22.5 years is appropriate for comparison with the observational record of airborne fraction, a time constant of 34.5 years is a better fit to the complete release history (Bacastow and Björkström, 1981). Using this value the airborne fraction falls from 64.3% (case (c)) to 59.0% (case (i)). The effect is again sufficiently large to favour the option of a numerical solution of a time-dependent model employing actual release rates, especially for the recent period. According to Rotty's (1981a) latest estimates, based on fuel consumption data, the growth rate changed from 4.58% per year for the period 1950–73 (corresponding well with $\mu^{-1} = 22.5$ years) to 2.25% per year from 1973 to 1980. This abrupt change was not evident in the estimates based on fuel production data formerly used (Rotty, 1981b). The present model indicates an airborne fraction well below 60% for this lower rate of increase.

4. Conclusions

The results of the model indicate that the division of the deep ocean into two diffusive reservoirs, with the characteristics of the deep Atlantic and the deep Pacific and Indian oceans, yields an airborne fraction that is not significantly different from that obtained with a single diffusive reservoir with the same mean ^{14}C content. The replacement of a sub-surface diffusive reservoir with a deep well-mixed reservoir in contact with the atmosphere reduces the airborne fraction enough to warrant the separate treatment of high latitude convective regions in ocean uptake models. However, comparable variations in airborne fraction are likely to result from variations in the decade to century time scale of the buffer factor and the fossil fuel CO_2 emission rates, the accommodation of which requires the use of a non-linear, time-dependent model.

5. Acknowledgement

This work was carried out at the Central Electricity Research Laboratories and is published with the permission of the Central Electricity Generating Board.

REFERENCES

Bacastow, R. B. and Björkström, A. 1981. Comparison of ocean models for the carbon cycle. In *Carbon cycle modelling* SCOPE 16 (ed. B. Bolin). Chichester: J. Wiley and Sons, 29–80.

Bacastow, R. B. and Keeling, C. D. 1981. Atmospheric carbon dioxide concentration and the observed airborne fraction. In *Carbon cycle modelling* SCOPE 16 (ed. B. Bolin). Chichester: J. Wiley and Sons, 103–112.

Bien, G. S., Rakestraw, N. W. and Suess, H. E. 1960. Radiocarbon concentration in Pacific Ocean water. *Tellus 12*, 436–443.

Bolin, B. 1981. Standardisation of notations and procedures. In *Carbon cycle modelling* SCOPE 16: (ed. B. Bolin). Chichester: J. Wiley and Sons, 81–101.

Broecker, W. S., Gerard, R., Ewing, M. & Heezen, B. C. 1960. Natural radiocarbon in the Atlantic Ocean. *J. Geophys. Res. 71*, 2903–2931.

Broecker, W. S. 1974. *Chemical oceanography*. New York: Harcourt Brace Jovanovich.

Craig, H. 1957. The natural distribution of radiocarbon and the exchange time of carbon dioxide between the atmosphere and the sea. *Tellus 9*, 1–17.

Fairhall, A. W., Young, A. W. and Bradford, P. A. 1962. Radiocarbon in the sea. In *Proceedings of the 8th International Conference on Radiocarbon Dating*. Wellington, New Zealand, 226–240.

Keeling, C. D. 1973. The carbon dioxide cycle. In *Chemistry of the lower atmosphere* (ed. S. I. Rasool). New York: Plenum Press, 251–331.

Montgomery, R. B. 1936. The present evidence on the importance of lateral mixing processes in the ocean. *Bull. Amer. Meteorol. Soc. 21*, 87–94.

Oeschger, H. and Heimann, M. 1981. Uncertainties of predictions of future atmospheric CO_2 concentrations. Presented at WMO/UNEP/ICSU Meeting on Analysis and Interpretation of Atmospheric CO_2 Data, Bern, 14–18 September 1981. WCP-14, Geneva, 225–232.

Oeschger, H., Siegenthaler, U., Schotterer, U. and Gugelmann, A. 1975. A box diffusion model to study the carbon dioxide exchange in nature. *Tellus 27*, 168–192.

Rotty, R. M. 1981a. Distribution and changes in industrial carbon dioxide production. Presented at WMO/UNEP/ICSU Meeting on Analysis and In-

terpretation of Atmospheric CO_2 Data, Bern, 14–18 September 1981. WCP-14, Geneva, 123–134.

Rotty, R. M. 1981b. Data for global CO_2 production from fossil fuels and cement. In *Carbon cycle modelling* SCOPE 16 (ed. B. Bolin). Chichester: J. Wiley and Sons, 121–126.

Siegenthaler, U. 1981. Uptake of excess CO_2 calculated by an outcrop-diffusion model of the ocean. Presented at WMO/UNEP/ICSU Meeting on Analysis and Interpretation of Atmospheric CO_2 Data. Bern, 14–18 September 1981. WCP-14, Geneva, 233–240.

Sundquist, E. T. and Plummer, L. N. 1981. Carbon dioxide in the ocean surface layer: some modelling considerations. In *Carbon cycle modelling* SCOPE 16 (ed. B. Bolin). Chichester: J. Wiley and Sons, 259–270.

Sundquist, E. T., Plummer, L. N. and Wigley, T. M. L. 1979. Carbon dioxide in the ocean surface: the homogeneous buffer factor. *Science 204*, 1203–1205.