

The Use of Tracers in Modelling the Oceanic Uptake of Carbon Dioxide [and Discussion]

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The use of tracers in modelling the oceanic uptake of carbon dioxide

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Increasing levels of atmospheric carbon dioxide from the burning of fossil fuels and changes in land use pose a threat of significant global climatic change in the 21st century. Owing to uncertainty in the pre-industrial atmospheric CO_2 concentration and in CO_2 releases from land-use change, direct estimates of the airborne fraction of this man-made CO_2 are not well established. Effort has therefore been devoted to estimating the strengths of the oceanic and biospheric sinks as an alternative route to determining the airborne fraction. This paper reviews the development of oceanic CO_2 -uptake models.

One-dimensional box models, with first-order exchange kinetics or vertical diffusion to simulate CO_2 penetration into the deep ocean and calibrated against natural ^{14}C distributions, appear inadequate. Their uptake is too small to be consistent with the recent atmospheric record and most estimated histories of CO_2 release from land use change.

Models incorporating representations of specific oceanic mixing processes important in CO_2 uptake achieve somewhat larger CO_2 uptake, especially when calibrated against 'short timescale' tracers, such as radiocarbon and tritium derived from bomb tests. Despite this general conclusion, substantial differences between such models remain. A comparison between my two-dimensional advection–diffusion model and Siegenthaler's outcrop–diffusion model illustrates how the relative importance of air–sea gas exchange rate and rate of ocean mixing in limiting CO_2 uptake depends critically upon modelling assumptions.

The failure of most models calibrated with a single oceanic tracer to reproduce well the distribution of other tracers has encouraged the development of multi-box, geographically realistic, models whose circulation and mixing patterns are determined by simultaneous inverse solution of a set of conservation equations for a range of tracers. This technique, when augmented with additional dynamical constraints, probably offers the most promise for advancing CO_2 -uptake modelling while suitable three-dimensional oceanic general circulation models are being developed. The latter, atmospherically driven, models will eventually play a key role in assessing how any future climatic change may feed back on atmospheric CO_2 levels. Feedback could arise either by alteration of the mixing processes responsible for man-made CO_2 uptake, or more fundamentally if changes in the surface-ocean productivity result from changes in circulation-mediated nutrient supply.

1. INTRODUCTION

Man's activities over the past century have caused substantial releases of carbon dioxide into the atmosphere. Since 1957, when continuous accurate measurements began, the atmospheric concentration has risen by about 10%, from 315 to around 348 parts per million by volume (p.p.m.v.). Carbon dioxide, water vapour and several other trace gases in the atmosphere are strong absorbers at the wavelengths near the peak of the Earth's infrared emission spectrum. Their natural concentrations maintain the surface temperature at habitable levels. Any increase

in their abundance poses a possible threat of global-scale warming and consequent climatic change. Although estimates of the likely climatic change are highly uncertain, it is probable that an increase in these infrared-absorbing trace gases corresponding to a doubling of the pre-industrial level of CO_2 would produce significant effects. Projections of future emissions suggest that such an increase may occur by, or soon after, the middle of the 21st century.

The rate, and probably the nature, of any climatic change will clearly depend on the rate of increase of CO_2 concentration. This in turn will depend upon the rates of future emissions and upon the fraction of these cumulative emissions that will remain airborne. This airborne fraction is itself a function of CO_2 emission growth rate and thus likely to vary with time.

Scientific research is unlikely to yield improved estimates of future emission rates; these will be governed by political and economic considerations. It should, however, provide improved understanding of the way excess CO_2 emitted to the atmosphere is partitioned between the atmospheric, oceanic and biospheric carbon reservoirs, and hence estimates of the airborne fraction likely to prevail for a given emission scenario.

In as much as past values of the airborne fraction, α , are a useful guide to the future, it is instructive to review how accurately these can be determined from available knowledge of emissions and atmospheric CO_2 concentration changes. Over the period 1860–1980, the main uncertainties in the calculation lie in the pre-industrial concentration of CO_2 and the amount of CO_2 that has been released through deforestation and land-use changes, in addition to that from fossil fuels. Assuming fossil-fuel emissions to have been as reported by Rotty (1981), figure 1 shows that the mean α over this period may have lain anywhere in the range 0.32–0.60 for the indicated ranges of uncertainty in the pre-industrial CO_2 level (275 ± 10 p.p.m.v.) and the forest and soil emissions from deforestation and land-use changes (150 ± 50 Gt C). Over the period 1958–1982, for which accurate measurements of the increase in atmospheric concentration are available, Bolin (1986*a*) adopts annual forest–soil emissions of 1.6 ± 0.8 Gt C, giving α in the range 0.34–0.46.

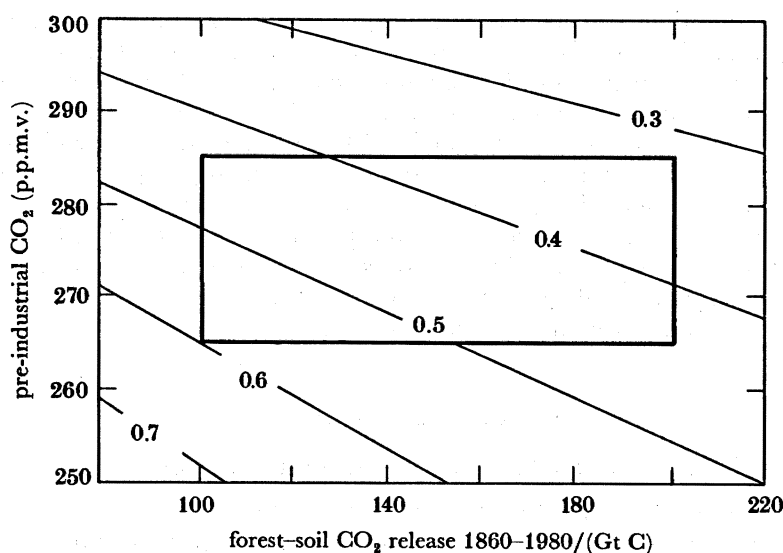


FIGURE 1. The average airborne fraction of CO_2 emissions from 1860–1980 as a function of pre-industrial atmospheric CO_2 concentration and emissions due to deforestation and changing land use, assuming fossil-fuel CO_2 emissions as given by Rotty (1981). The heavy lines indicate the uncertainty estimated by Bolin (1986*a*). (After Bolin 1986*a*.)

Because of isotopic fractionation by plants, the size of the global biosphere at any time is indicated by the ¹³C:¹²C ratio of atmospheric CO₂. On this basis estimates of past forest–soil emissions have been made by using carbon-cycle models to deconvolve records of ¹³C:¹²C ratio recorded in tree rings (see, for example, Stuiver *et al.* 1984; Peng 1985). These are subject to the uncertainty associated with climatic and growth effects on the ¹³C:¹²C ratio and with the accuracy with which carbon-cycle models simulate the carbon system. These studies have tended to calculate similar total non-fossil carbon releases over the industrial period, but with quite different distribution in time, giving much lower releases over the past 30 years than direct estimates. The same technique has been used by Druffel & Benavides (1986) employing measurements of the ¹³C:¹²C ratios in a banded Jamaican sclerosponge, which is considered a more accurate recorder of prevailing oceanic, and hence atmospheric, ¹³C:¹²C ratios. They calculate a much smaller overall forest–soil emission, but accord with Stuiver *et al.* (1984) in obtaining a value close to zero for the recent period, indicating α around 0.58.

Deconvolution of the CO₂ record from measurements of air trapped in an Antarctic ice core (Siegenthaler & Oeschger 1987) also indicates a smaller release (0–0.9 Gt C annually for the period 1959–1983) than obtained from direct estimates of deforestation and land-use change, giving α in the range 0.58–0.48. This range results from the use of two alternative carbon-cycle models. The lower value, 0.48, is obtained by using the model whose ocean is able to absorb larger amounts of carbon and that can thus permit a larger non-fossil carbon emission rate (0.9 Gt C a⁻¹) given the same atmospheric CO₂ record. This model is considered to provide an upper limit to the oceanic uptake.

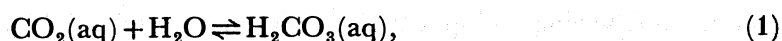
The inconsistency that exists between the carbon-isotope – model-based analyses and the direct estimates of forest–soil emissions, and the consequent wide range of uncertainty in the current airborne fraction, indicate a continuing need to reduce further the uncertainties associated with (i) direct estimates of forest–soil carbon release, (ii) the size of the oceanic sink and (iii) the numerous other minor sinks for man-made carbon that have been proposed in the face of this failure to balance the carbon budget. This paper addresses the second of these requirements, and examines the way in which oceanic tracers have played a key role in the development of oceanic carbon-uptake models.

2. FACTORS GOVERNING THE CO₂ UPTAKE BY THE OCEANS

The extent to which the oceans take up excess CO₂ is controlled primarily by three factors: the capacity for CO₂ absorption when the ocean is in equilibrium with the atmosphere; the rate of CO₂ exchange between atmosphere and surface ocean; and the rate at which surface water, having absorbed CO₂, is mixed into the deeper layers.

(a) CO₂ capacity

Dissolved inorganic carbon exists in the ocean in four forms: dissolved carbon dioxide gas, undissociated carbonic acid, carbonate ion and bicarbonate ion. Ignoring the influence of the borate system, their relative concentrations are governed by the reactions



It is essentially the presence of the CO_3^{2-} ions, which comprise about 10–15 % (by mass) of total dissolved inorganic carbon (ΣC), that give seawater its ability to absorb CO_2 in amounts substantially larger than dissolution alone would provide. CO_3^{2-} ions react with H^+ ions, derived from dissociation of H_2CO_3 , to form HCO_3^- ions. This effectively removes H^+ ions and the system responds by dissolving more CO_2 . With the present atmosphere–ocean carbon system, although dissolved CO_2 constitutes less than 1 % of ΣC , reaction with carbonate ensures that at equilibrium ΣC typically increases by 1 % for every 10 % increase in dissolved CO_2 concentration or atmospheric partial pressure of CO_2 , p_{CO_2} . This ratio of the fractional changes

$$\frac{\delta p_{\text{CO}_2}/p_{\text{CO}_2}}{\delta \Sigma \text{C}/\Sigma \text{C}}$$

is known as the Revelle factor, R . The value of 10 adopted is an average value of R , which ranges from about 9 in low latitudes to 14 in high latitudes, depending primarily on temperature. As p_{CO_2} rises, R increases, so gradually reducing the capacity of the oceans to take up CO_2 .

Many carbon-cycle models do not attempt to handle the different species of dissolved inorganic carbon and therefore rely on the use of the Revelle factor to simulate correctly the response of ΣC to changes in the atmospheric concentration.

(b) Air–sea exchange

The surface layer of the ocean influenced by wind stress is generally regarded as well mixed in carbon-cycle models. The rate at which its carbon content responds to a change in atmospheric CO_2 concentration is determined by the rate at which CO_2 can cross the air–sea interface. A review of the many theoretical, laboratory and field techniques that have been applied to evaluating air–sea gas-exchange rates is given by Liss & Merlivat (1986). For most carbon-cycle models developed to date, which are highly aggregated geographically, the most appropriate techniques are those that provide a global mean estimate of gas-exchange rates. One such relies on the distribution between atmosphere and surface ocean of natural ^{14}C . Assuming a steady state before man's perturbation of the system, the radioactive decay of ^{14}C within the oceans must have balanced the excess of input over output at the ocean surface. Thus

$$R_{\text{oc}} N_{\text{oc}} \lambda = F(R_{\text{a}} - R_{\text{s}}) A_{\text{s}}, \quad (4)$$

where F is the exchange flux, A_{s} the ocean surface area, R_{a} , R_{s} and R_{oc} the relative atmospheric, surface ocean and mean ocean ^{14}C concentrations, N_{oc} the total dissolved carbon content of the ocean and λ the radioactive decay constant for ^{14}C . Setting $R_{\text{a}} = 100\%$, $R_{\text{s}} \approx 95\%$ and $R_{\text{oc}} \approx 84\%$ as indicated by results of the GEOSecs programme, Siegenthaler (1986) obtains an exchange rate of $18 \text{ mol m}^{-2} \text{ a}^{-1}$ ($\pm 25\%$) corresponding to a transfer velocity of 17.5 cm h^{-1} for a pre-industrial concentration of 280 p.p.m.v. Given that the upper 70 m of the ocean contain about 150 mol m^{-2} , the residence time of carbon atoms in this layer with respect to transfer to the atmosphere is about 8 years. However, re-establishment of equilibrium between atmosphere and ocean following an input to the atmosphere is achieved in a fraction $1/R$ of the residence time, where R is the Revelle factor. Equilibration between atmosphere and mixed layer is thus realized in less than one year (Broecker *et al.* 1971).

The distribution of natural radiocarbon within the ocean indicates that, on average, surface

water is replaced by sub-surface ocean water very much less frequently than this, so the surface ocean can be considered close to equilibrium with the atmosphere. The rate of air–sea exchange is thus not expected to constrain the overall uptake of CO₂ by the oceans. This conclusion may not, however, be valid regionally, a point to be considered later in connection with the development of disaggregated models of the ocean.

(c) *Mixing between the surface and deep ocean*

Whereas the surface mixed layer of the ocean contains roughly the same amount of carbon as the atmosphere, the total ocean content is about 60 times this amount. With a Revelle factor of 10, this indicates that if only the surface ocean had been involved in absorbing man-made CO₂, only 9% would have been taken up. Equilibration with the whole ocean, on the other hand, would result in about 85% uptake. This calculation assumes no dissolution of calcium carbonate sediments, which would alter the alkalinity.

It is clear both from the estimates of airborne fraction discussed in the introduction and from the observed distributions of bomb tracers that the volume of ocean that has been affected by man-made CO₂ is considerably greater than the mixed layer but falls far short of the total volume. The circulation and mixing processes that achieve the transfer of CO₂-laden water into the deeper ocean are not well understood. It is these same processes that will be responsible for transferring heat away from the surface ocean during the course of a climatic warming and that will profoundly influence the rate, and perhaps also the geographical distribution, of consequent climatic changes. It will be at least a decade before three-dimensional ocean circulation models with the required resolution, and the data necessary for their validation, are developed to deal with this task adequately. Lacking such tools, a range of less sophisticated atmosphere–ocean models have been developed during the past three decades to address the carbon-uptake problem. These analogue models have adopted simple representations of the physics of mixing and circulation and have relied heavily on observed tracer distributions for their calibration and validation. Subsequent sections of this paper review the important developments that have taken place.

3. ONE-DIMENSIONAL BOX MODELS

The simplest models (see, for example, Craig 1957; Revelle & Suess 1957; Bolin & Eriksson 1959; Keeling 1973) are those in which the atmosphere, surface ocean and deep ocean are treated as three well-mixed reservoirs (figure 2). Transfer of carbon between reservoirs is by first-order kinetics, i.e. fluxes are proportional to the carbon content of the upstream reservoir. With an input to the atmospheric box from man-made emissions, and with allowance made for the Revelle factor in CO₂ exchange at the surface, a set of ordinary differential equations describes how the carbon in each reservoir changes with time.

The constants of proportionality are evaluated from a consideration of the observed steady-state natural ¹⁴C balance. For the atmospheric reservoir, for example,

$$\frac{d}{dt} {}^{14}N_a = 0 = -k_{as} {}^{14}N_a + k_{sa} {}^{14}N_s - \lambda {}^{14}N_a + {}^{14}F_a,$$

where ¹⁴N_a, ¹⁴N_s are the ¹⁴C contents of the atmosphere and surface ocean, *k*_{as} and *k*_{sa} are the atmosphere–surface ocean and surface ocean–atmosphere exchange constants, λ is the ¹⁴C

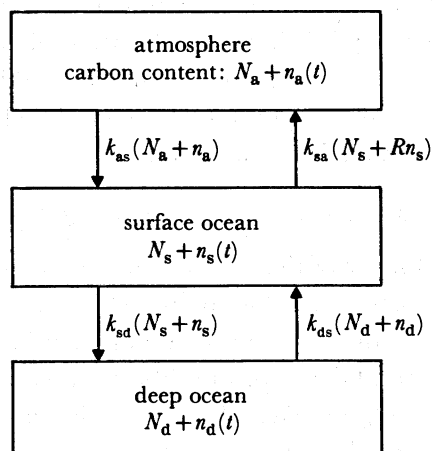


FIGURE 2. Three-box atmosphere–ocean model. N_a , N_s , N_d represent steady-state pre-industrial carbon contents of the atmosphere, surface ocean and deep ocean respectively, and n_a , n_s , n_d man-made departures therefrom. k_{xy} is the first-order exchange coefficient for the carbon flux from box x to box y . R is the Revelle factor.

decay constant and $^{14}F_a$ the atmospheric ^{14}C source from cosmic rays. Analogous equations for the ocean reservoirs and corresponding equations for the pre-industrial steady-state carbon balance allow all exchange constants to be evaluated.

It is found that if calibrated to match the observed ^{14}C distributions, this model can achieve an airborne fraction of less than 0.60 only if the surface mixed-layer reservoir is more than 500 m deep. For a more realistic depth of 75 m, an airborne fraction of over 0.8 is obtained (Bacastow & Björkström 1981). The assumption that the ocean beneath the surface layer is well mixed prevents the simulation of relatively rapid mixing between the surface ocean and the upper layers of the deep ocean as evidenced by the structure of the ^{14}C distribution.

To simulate this feature, Oeschger *et al.* (1975) developed the box-diffusion model in which the well-mixed deep-ocean reservoir is replaced with a reservoir characterized by vertical diffusion. The diffusion coefficient was chosen to match the mean vertical distribution of ^{14}C . The uptake of CO_2 is considerably enhanced relative to the three-box model but still appears to fall short of that suggested by the atmospheric record. Only a small improvement is obtained by splitting the deep ocean into an 'Atlantic' and 'Pacific' reservoir to reflect the more rapid mixing into the former as revealed in the ^{14}C observations (Crane 1982).

Although early modelling studies had explored the implications of direct CO_2 exchange between the atmosphere and the deep ocean (Craig 1963; Broecker *et al.* 1971), it was the distributions of bomb tracers that graphically illustrated the potential importance for CO_2 uptake of deep convection in high-latitude regions of the Atlantic Ocean (Östlund *et al.* 1974). Subsequent modelling studies have explored various techniques for simulating surface – deep-water exchange (Björkström 1979; Bacastow & Björkström 1981; Killough & Emanuel 1981; Hoffert *et al.* 1981; Crane 1982; Peng *et al.* 1983; Siegenthaler 1983). Generally such modifications to the two-box and box-diffusion models increase their capacity for CO_2 uptake, although the extent depends critically upon the exact structure of the downwelling and return flows adopted (Killough & Emanuel 1981).

The availability of bomb ^{14}C and tritium (3H) data from the GEOSECS surveys has encouraged recalibration of various versions of the box-diffusion model against these shorter timescale

tracers (Broecker *et al.* 1980; Viicelli *et al.* 1981). Whereas the global mean natural ¹⁴C profile best matches a vertical diffusivity, K , of $1.25 \text{ cm}^2 \text{ s}^{-1}$ (Oeschger *et al.* 1975), Broecker *et al.* (1980) calculated $K = 1.7 \text{ cm}^2 \text{ s}^{-1}$ from ³H data and $K = 1.9\text{--}3.3 \text{ cm}^2 \text{ s}^{-1}$ from bomb ¹⁴C observations. The uncertainty in the latter case arises from difficulties in disentangling bomb-produced and natural ¹⁴C in the upper layers of the oceans.

The typical extent of the influence of both the introduction of surface – deep-ocean exchange and recalibration with a bomb tracer is illustrated by Siegenthaler's (1983) outcrop–diffusion model (figure 3), which allows direct exchange between the atmosphere and deep ocean via

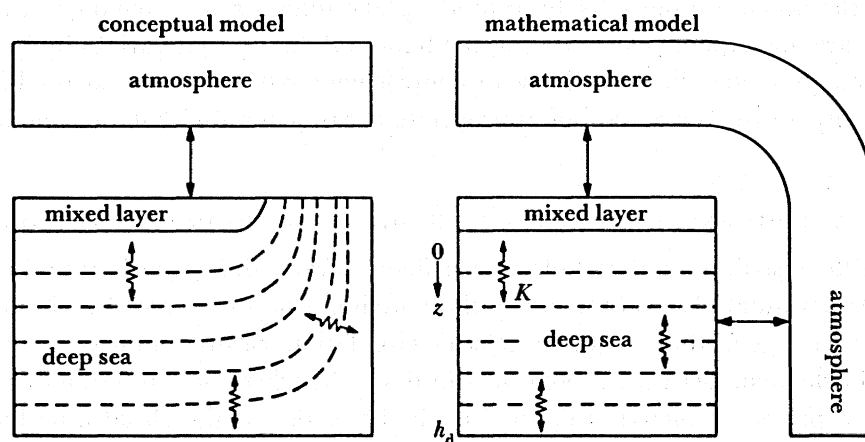


FIGURE 3. The outcrop–diffusion model developed by Siegenthaler (1983). In low latitudes, carbon is exchanged between the atmosphere and a well-mixed surface-ocean layer. At high latitudes, exchange occurs between atmosphere and outcropping isopycnal deep-ocean layers; represented by well-mixed horizontal layers in the mathematical representation. Vertical (cross-isopycnal) eddy diffusion represents the effect of other mixing processes within the ocean. (After Siegenthaler 1983).

outcropping isopycnal surfaces. Calibrated against natural ¹⁴C, a change from 0 to 10% outcrop area reduced the airborne fraction, α , from 0.67 to 0.61. When calibrated against bomb ¹⁴C, the corresponding figures for α were 0.61 and 0.53 respectively. Not surprisingly, Siegenthaler found that the use of a uniform K value reflecting the overall effects of mixing processes that have operated on the timescale of 10–20 years (bomb ¹⁴C) overestimated the rate of mixing into the deep ocean. Generally, low-resolution models calibrated with bomb tracers are unlikely to be appropriate for simulating the long-term response of the oceans to CO₂ input, just as those calibrated against natural ¹⁴C appear to underestimate uptake on the short timescale.

To achieve compatibility between short- and long-timescale mixing processes, a much more disaggregated model of the ocean would seem to be required. This in itself, however, poses further potential difficulties with regard to tracer calibration as has been discussed by Peng & Broecker (1985). None of the available tracers provides a perfect analogue for man-made CO₂. Tritium was transferred quickly to the surface ocean in rain and by vapour exchange, so its distribution is strongly influenced by the location of bomb tests. Carbon-14 differs from CO₂ in that it takes approximately 10 times longer for the surface ocean to equilibrate with atmospheric ¹⁴C (*ca.* 10 years) than with CO₂ (*ca.* 1 year). Because uptake into the thermocline is thought to occur primarily through mixing along isopycnal surfaces that outcrop at the

surface only in winter, very little ^{14}C would enter during the temporary exposure of this water at the surface. However, partial equilibration with atmospheric CO_2 would ensure a CO_2 uptake route not reflected in the bomb ^{14}C distributions. Unlike ^{14}C , ^{85}Kr and chlorofluorocarbons, being non-reactive, equilibrate rapidly between atmosphere and ocean in about one-tenth the time taken for CO_2 equilibration.

The extent to which the differences between the characteristics of CO_2 and these other tracers devalue their individual roles as suitable analogues for calibrating CO_2 -uptake models depends in part on the degree to which gas exchange rate is a limiting process. Atlantic circulation models that have recently been developed (Peng & Broecker 1985) (see §4) suggest that mixing rates within the ocean are more likely to limit uptake than air-sea exchange rate. However, these models are not capable of simulating the temporal and spatial variability or the scale of the circulation processes that are likely to control winter-time mixing so resolution of this debate probably awaits the arrival of appropriate ocean general-circulation models.

4. A TWO-DIMENSIONAL ADVECTION-DIFFUSION MODEL OF THE OCEAN

A study of the distribution of oceanic tracers demonstrates that variations in the meridional (depth against latitude) plane are significantly more important than zonal variations. Patterns of tracers further suggest that several large-scale circulation features of the ocean are probably responsible for the main features of their distributions. Broecker *et al.* (1980), for example, used the circulation pattern depicted in figure 4 as the basis for a regionalized upwelling – downwelling-diffusion model. It would seem worthwhile therefore to explore the potential of two-dimensional meridional advection-diffusion models with simple circulation patterns to simulate the uptake of man-made CO_2 emissions. Such a model has been constructed (Crane 1986). The following subsections describe the structure and calibration of the model, and the sensitivity of carbon uptake to key model parameters. Some comparisons are drawn with the model of Siegenthaler (1983).

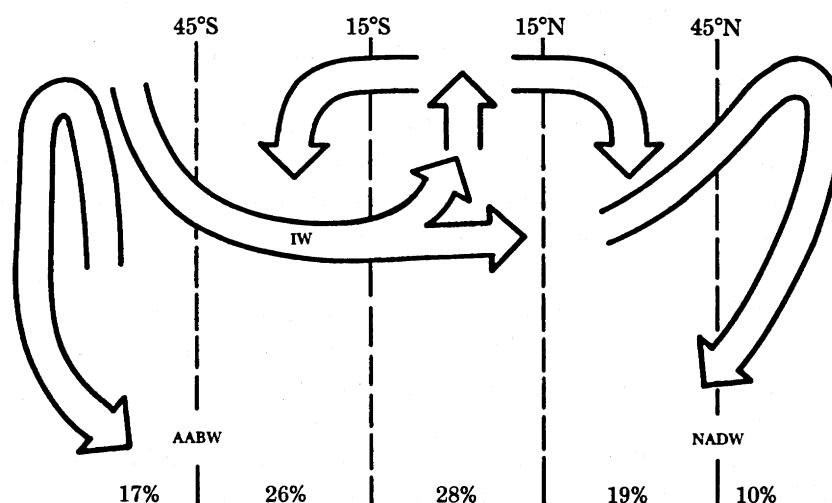


FIGURE 4. Atlantic circulation system envisaged by Broecker *et al.* (1980) as the basis for a regionalized upwelling-downwelling diffusion model, comprising Antarctic Bottom Water (AABW) and North Atlantic Deep Water (NADW) formation, and northward-penetrating Antarctic Intermediate Water (AIW). Equatorial upwelling feeds water polewards in the upper layers to sink in the subtropical gyres. (After Broecker *et al.* 1980.)

(a) *Model structure*

The two-dimensional advection–diffusion equation

$$\frac{\partial C}{\partial t} + v \frac{\partial C}{\partial y} + w \frac{\partial C}{\partial z} - \frac{\partial}{\partial y} \left[K_y \frac{\partial C}{\partial y} \right] - \frac{\partial}{\partial z} \left[K_z \frac{\partial C}{\partial z} \right] = 0$$

is solved by timestepping with an upwind differencing scheme on a finite difference grid. The grid has regular 8° latitude spacing in the horizontal but variable depth spacing ranging from 50 m in the upper ocean (0–200 m), to 200 m at intermediate depths (200–1000 m), to 500 m in the deep ocean (1000–4000 m). The depth is uniform with latitude and the boundaries are vertical. The model is representative of the Atlantic Ocean and the varying width of the ocean is taken into account when water fluxes derived from the specified stream functions are converted into horizontal and vertical velocities. The advective circulation is largely determined by the specification of the stream function for four independent circulation systems: the two sub-tropical gyres, the Antarctic Bottom Water (AABW) system and the Antarctic Intermediate – North Atlantic Deep Water (NADW) system. This idealized circulation pattern was modified during the calibration procedure to permit advective exchange between the rising branches of the AABW and NADW systems in high southern latitudes (figure 5). A simple linear interpolation of stream function to zero at the ocean boundaries and the circulation-system interfaces was adopted. Based on estimates of water mass formation and equatorial upwelling rates (W. S. Broecker & T.-H. Peng, unpublished results) the AABW and NADW circulations were set to $20 \times 10^6 \text{ m}^3 \text{ s}^{-1}$ and the subtropical gyres at $5 \times 10^6 \text{ m}^3 \text{ s}^{-1}$.

One major difficulty with the model was the choice of diffusivities. Given the uncertainty that exists over the relative role of advective and diffusive mixing processes, token values were adopted initially, and adjusted where necessary during the calibration procedure. For the same reason, no attempt was made to minimize the numerical diffusion generated by the upwind finite differencing by adopting more sophisticated schemes.

(b) *Calibration*

In view of the disaggregated nature of the model of the ocean an attempt was made to calibrate the model against both long- and short-timescale traces. Salinity was chosen as a suitable long-timescale tracer. With timesteps ranging from one thirtieth of a year in the 0–200 m layer to one half of a year in the deep ocean, the model was integrated forward in time until a steady salinity distribution was obtained. Initial conditions comprised observed surface-ocean salinities. Surface salinities were restored to these values after each timestep as a means of representing the salinity sources and sinks at the surface. Salinity was also held fixed in two other regions of the model to represent the highly saline Mediterranean outflow (0–600 m, 28–36° N) and the outflow for the Weddel Sea (200–400 m, 68–76° S). As indicated above, adjustments to the circulation rates centred on the exchange across the rising branches of the AABW and NADW circulations. Figure 6, which compares the modelled and observed salinity distributions, shows that the main features have been reproduced by the model.

The GEOSECS (1972) and TTO (1981) tritium distributions (Östlund *et al.* 1974; Östlund 1983) were used to assess the time-dependent response of the model to short-timescale tracers. The temporal and latitudinal distribution of bomb-tritium input to the surface ocean from 1952 to 1975 was calculated by using the methods outlined by Weiss & Roether (1980) and

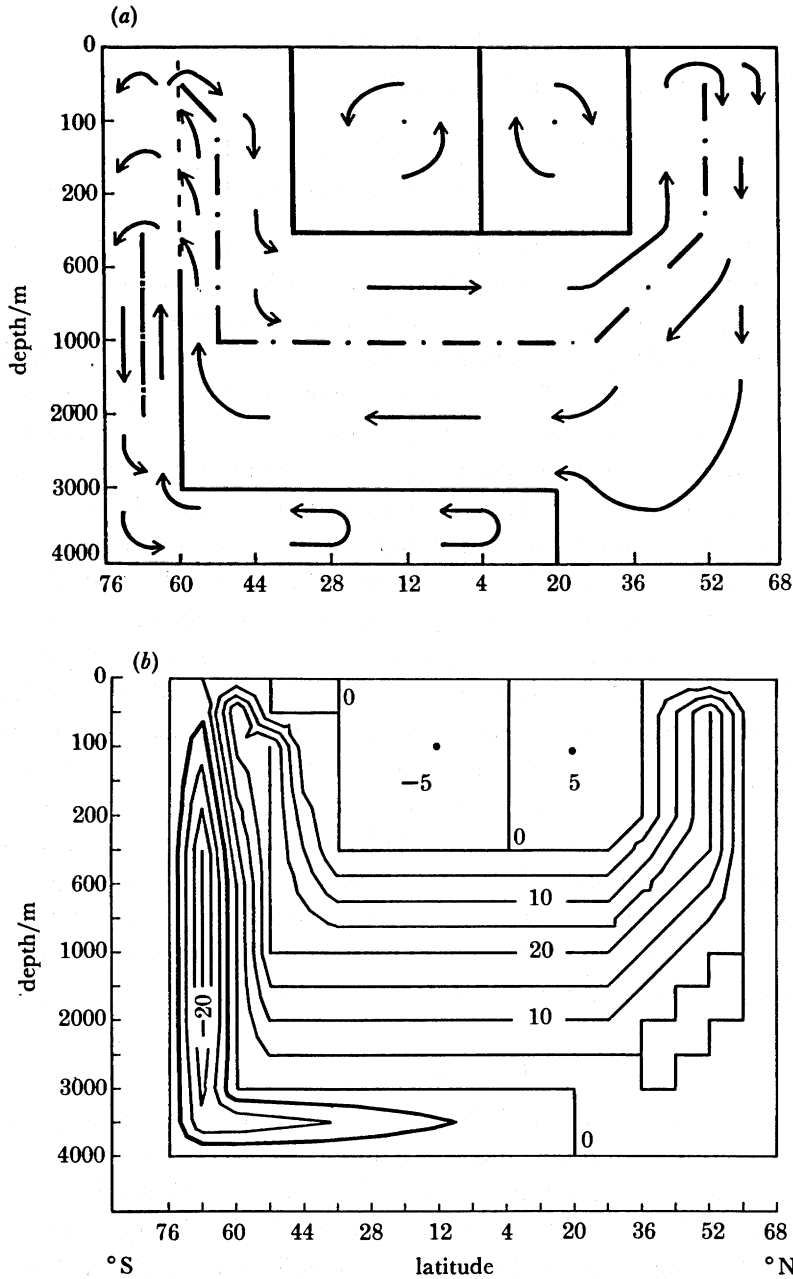


FIGURE 5. (a) Schematic of advective circulation system adopted in the advection–diffusion model of the Atlantic Ocean. (b) Stream function ($10^6 \text{ m}^3 \text{ s}^{-1}$) specified for Atlantic advective circulation depicted in (a). (See text for details.)

used as input to the model into which a tritium-decay term was introduced. In figure 7, a comparison is shown between observations and the modelled tritium distributions in 1972 and 1981, starting from zero concentration in 1952. Although the model contains too many degrees of freedom to ensure that calibration was optimized, the only change in model parameters from those used to simulate salinity that was required to significantly improve the tritium simulations was the enhancement of horizontal diffusivity in northern temperate and polar latitudes at

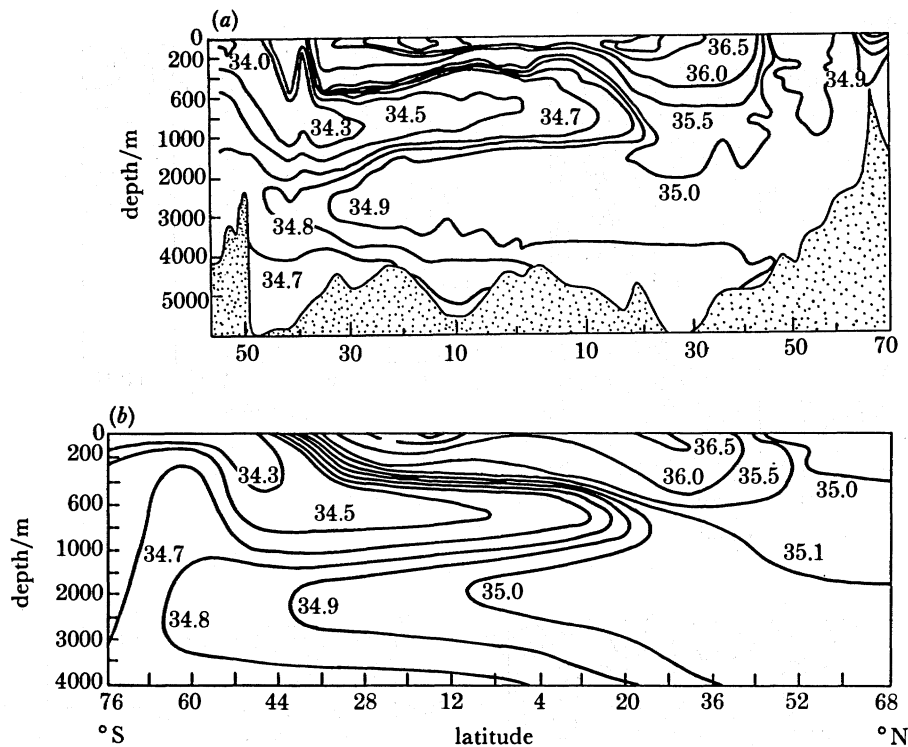


FIGURE 6. Comparison between (a) observed (GEOSECS) Atlantic salinity distribution (in units of per mille) and (b) modelled salinity in the advective-diffusion model. The latter represents the steady state achieved with a 1200 year integration of the model. Surface salinity is maintained at observed values. Constraints are imposed to represent high-salinity Mediterranean and Weddel Sea outflows. (See text for details.)

intermediate depths. No parameter variation, however, within the bounds dictated by numerical stability considerations was able to simulate the rapid penetration of tritium to the deep ocean in high latitudes.

(c) Response to atmospheric CO₂ inputs

The model was used to simulate the oceanic uptake of man-made atmospheric CO₂ inputs. To represent the world ocean two alternative scalings were adopted. In the first, the surface area of each latitude band was multiplied by 4.5, the ratio of global ocean area to Atlantic area, so the world ocean behaved as a scaled version of the Atlantic. In the second the same factor was applied, but circulation rates in the AABW and NADW systems were then reduced by a factor of 3.3 (the ratio of global to Atlantic Ocean area poleward of 36° latitude) on the basis that these circulations do not exist in the Pacific and Indian Oceans. Of these alternatives the first is likely to overestimate CO₂ uptake on account of too much deep- and bottom-water formation. The second will give rise to a considerable underestimate because it also excludes the Indian and Pacific AIW circulations due to the linked AIW – NADW system in the model.

The airborne fraction, α , over the period 1959–1979 for fossil-fuel CO₂ inputs (1860–1979) given by the model as calibrated was 0.67 from the first method of extrapolation to the world ocean and 0.76 from the second. The sensitivity of airborne fraction to variation in model parameter values was also examined in view of the imprecision of the calibration technique. With circulation rates (i) double and (ii) half the calibration values, α values of 0.60 and 0.73

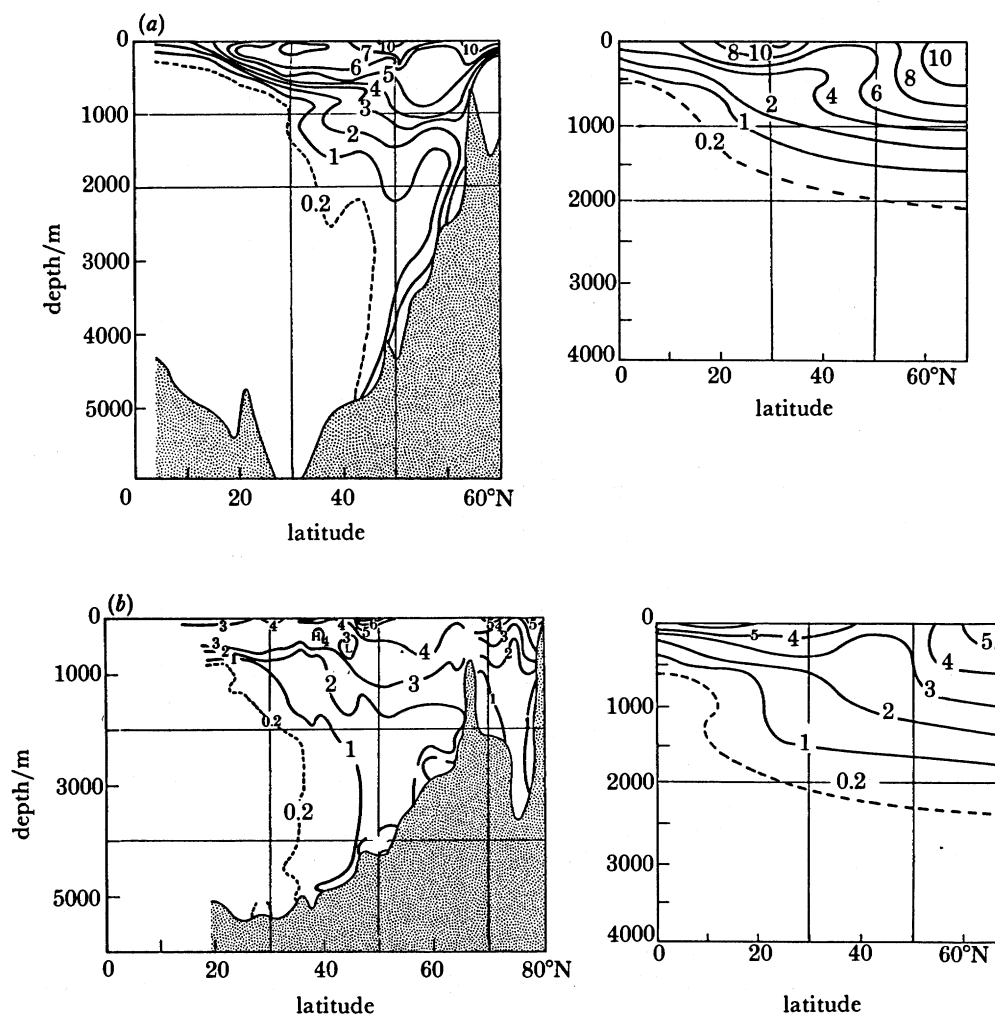


FIGURE 7. (a) Comparison between observed (GEOSecs) North Atlantic tritium distribution in 1972 (left) with modelled tritium at 1972 in the advection-diffusion model (tritium units). The latter results from a 21 year integration of the model from the tritium input history 1952–1972 derived by Weiss & Roether (1980). (b) As (a) but for 1981 (units TU81). Input continues until 1975. (Note the different depth-latitude scaling of the two figures.)

were obtained, by using the first method of extrapolation. Subsequent tritium integrations suggested that the doubled circulation rate continued to give reasonably good agreement with observations, whereas the halved rate led to excessively high surface concentrations.

Airborne fraction was found to be insensitive to realistic variations in the size and latitudinal pattern of air-sea gas exchange rates. An increase at all latitudes from 20 to 30 mol m⁻² a⁻¹ reduced α by only 0.01. It is interesting to relate this feature of the model to the discussion by Siegenthaler (1983) of the importance of air-sea exchange rate in determining the CO₂ uptake of the outcrop-diffusion (o-d) model. The p_{CO_2} of surface water in the outcrop region of the o-d model is far from equilibrium with atmospheric CO₂, with the sea-air flux of CO₂ only 0.1–0.2 times the air-sea flux. This results from the infinitely rapid lateral mixing assumed in the layer of the ocean that outcrops. In the low-latitude region of the o-d model the ratio of the exchange fluxes is about 0.8. Figure 8 shows the latitudinal distribution of the ratio of air-sea to sea-air

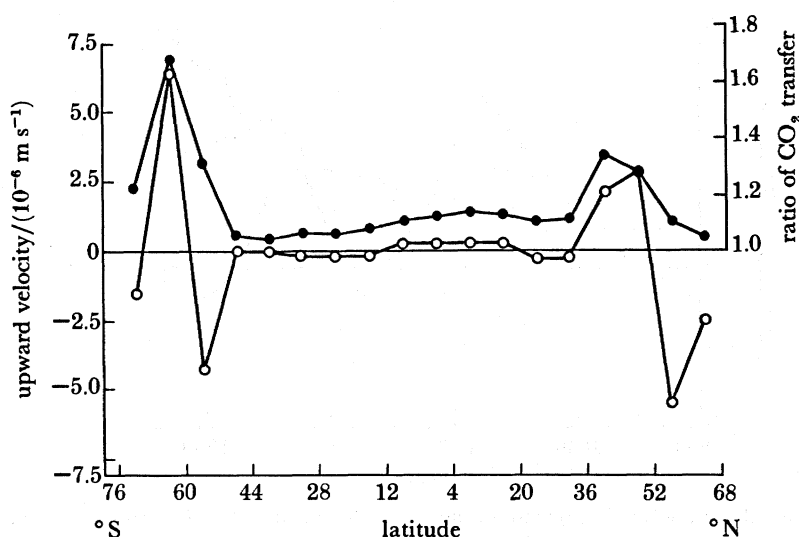


FIGURE 8. Ratio of air-sea to sea-air transfer of carbon (right-hand ordinate, ●) and vertical velocity at 50 m (10^{-6} m s $^{-1}$) (left-hand ordinate, ○) as a function of latitude in the globally scaled (version 1) Atlantic model. (See text for details.)

transfer obtained in the advection-diffusion model. It is seen that although the net uptake is greater in regions of rapid overturning, the finite rate of mixing into the sub-surface layers restricts the ratio to more modest values. Thus whereas in the o-d model air-sea exchange rate is the limiting factor for high-latitude uptake, it is the rate of circulation that limits uptake in the present model. This would appear to be an important reason why the o-d model generally simulates a rather lower airborne fraction (§3) than the advection-diffusion model.

It is worth reiterating here an important aspect of the CO₂ system which has been noted previously by other authors (Siegenthaler *et al.* 1978; Oeschger & Heimann 1983; Baes & Killough 1985; Siegenthaler 1986; Siegenthaler & Oeschger 1987). The airborne fraction during recent decades not only depends upon the releases of fossil and non-fossil CO₂ releases during this period but also upon the pattern of release in earlier decades. Table 1 illustrates the model response to four CO₂ input histories: fossil-fuel release only and fossil-fuel release combined with the biospheric-release histories calculated by Stuiver *et al.* (1984), Peng *et al.* (1983) (from ¹³C:¹²C records in tree rings) and Houghton *et al.* (1983) (from land-use statistics). These three biospheric-release histories are illustrated in figure 9. A large biospheric

TABLE 1. RESPONSE OF ADVECTION-DIFFUSION MODEL TO FOUR CO₂ INPUT FUNCTIONS

(Fossil-fuel emissions (FF) (Clark 1982), and FF plus biospheric release histories estimated by Stuiver *et al.* (1984), Peng *et al.* (1983) and Houghton *et al.* (1983). The Stuiver *et al.* (1984) release was extended from 1970 to 1979 assuming emissions constant at 1970 values. 'Observed airborne fraction' implies (observed CO₂ increase 1959-1979/CO₂ input 1959-1979).)

input function	CO ₂ input/(Gt C)		airborne fraction 1959-1979		implied 1820 CO ₂ level (p.p.m.v.)
	1820-1979	1959-1979	model	observed	
FF	160	78	0.67	0.58	292
FF + Stuiver <i>et al.</i> (1984)	258	79	0.62	0.58	273
FF + Peng <i>et al.</i> (1983)	422	105	0.53	0.43	237
FF + Houghton <i>et al.</i> (1983)	357	131	0.61	0.36	254

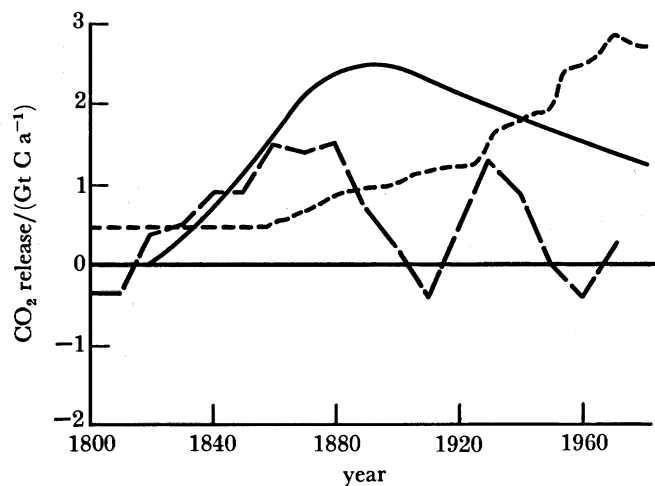


FIGURE 9. Estimates of biospheric CO₂ release to the atmosphere from ¹³C:¹²C records in tree rings (—, Peng *et al.* 1983; ———, Stuiver *et al.* 1984) and from land-use statistics (-----, Houghton *et al.* 1983).

release during the first half of this century can significantly affect the present CO₂ concentration by providing a decreasing base line while this CO₂ is slowly taken up by the oceans. This is illustrated by the model's response to the release history of Peng *et al.* (1983). Although the total release is the largest of the three, it produces the lowest 1959–1979 model airborne fraction.

It is clear from table 1 that only a biospheric-release history with a moderate pre-1959 release and a near zero post-1959 release, like that of Stuiver *et al.* (1984), allows both a model airborne fraction and a pre-industrial CO₂ concentration roughly consistent with observations. A larger historical release implies too low a pre-industrial CO₂ concentration, whereas significant current biospheric release gives rise to a substantial over estimate of the contemporary airborne fraction.

5. MULTI-TRACER MODELS AND INVERSE TECHNIQUES

The advection–diffusion model described in §4 essentially relies upon the use of a single tracer to calibrate an arbitrarily specified circulation pattern. Peng & Broecker (1985) used tritium data to calibrate five alternative 11-reservoir models of Atlantic thermocline mixing. Again, trial-and-error methods were used to determine the flow rates that gave a 'best fit' to the data. Once calibrated, Peng & Broecker (1985) subjected their models to rigorous checks on their ability to reproduce both the temporal and spatial characteristics of the tritium signal, and the time histories, distributions and ratios of other bomb tracers. In this way they were able to judge which of the five alternative mixing patterns gave an overall best fit to all the tracer data employed.

Although multiple tracers were involved in the validation of Peng & Broecker's thermocline models, only one, tritium, was used in calibration. Given the large number of reasonably well-observed oceanic tracers, a more-even allocation between the number of tracers used for calibrating models and the number used for validation would seem to be desirable.

The problem of the simultaneous use of a number of tracers in a three-box ocean model was first studied twenty years ago (Keeling & Bolin 1967, 1968), but was not taken up again until the methodology for multi-reservoir models was developed by Wunsch & Minster (1982) and Bolin *et al.* (1983).

(a) *Inverse methodology*

The objective of the inverse method is to find the patterns of advection and turbulent diffusion between model boxes that simultaneously best satisfy the known distributions of a number of tracers. When modelling the carbonate system, the rates of biological production, decomposition and dissolution of carbon, like those of advection and turbulent diffusion, may also be included as unknowns (figure 10). Generally, with M tracers, I ocean boxes and L

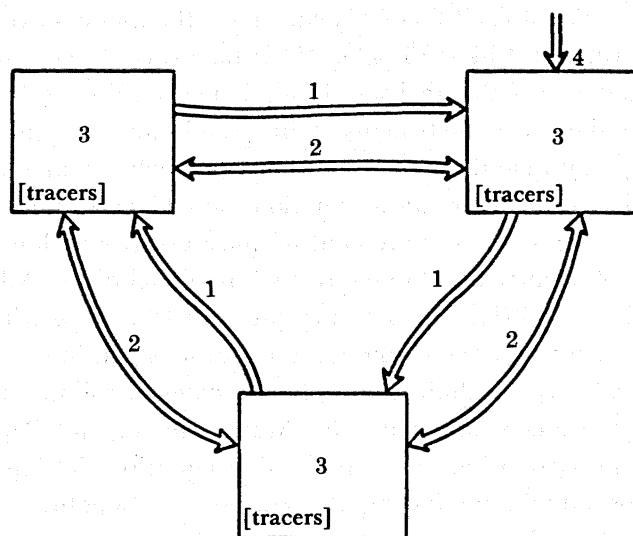


FIGURE 10. The basis of inverse methodology as applied to multi-tracer box models. Arrows represent advective or turbulent diffusive exchanges or exchange at model boundaries. 'Knowns' and 'unknowns' are listed: 1, advective flux across internal boundaries (unknown); 2, turbulent mixing across internal boundaries (unknown); 3, radioactive decay (known) and biochemical processes (unknown) within box; 4, flux across external boundaries (known). (After Moore & Björkström 1986.)

surfaces between boxes, the equation of continuity for each tracer and for water for each box yields a system of $(M+1)I$ simultaneous equations in $2L+2I$ unknowns, i.e. velocity and diffusivity at each box interface and source-sink terms for organic carbon and carbonates in each box. In their study, Bolin *et al.* (1983) derive a further $2S$ equations, where S is the number of surface boxes, by requiring that detritus formed in the surface layer decomposes (organic carbon) or dissolves (carbonate) in the layers beneath. In matrix form the problem to be solved is

$$Ax = b$$

where the vector x comprises the $2(L+I)$ unknowns, A is the $r \times s$ matrix of coefficients of the unknowns, where $r = (M+1)I + 2S$ and $s = 2(L+I)$, and the vector b contains the tracer production and decay terms.

Generally $r \neq s$. If too few tracers are available relative to the number of unknowns $r < s$ and the problem is under-determined or *indeterminate*. If more conservation equations are used than there are unknowns to solve for, $r > s$ and the problem is over-determined or *incompatible*. There is thus no one solution that satisfies all the conservation equations exactly. The methods of solution therefore seek to find a set of values for the unknowns x that either minimize the solution vector x (indeterminate case) or minimize the errors in a least squares sense in the conservation equations $Ax = b$ (incompatible case), subject to any additional constraints that

may be imposed, such as the requirement that diffusivities be non-negative. The solution may be influenced by appropriate weightings to stress the role of some tracers relative to others. Water conservation, for example, would be given a high weighting.

Bolin *et al.* (1983) describe how they adapted the solution techniques of Lawson & Hanson (1974) to their particular problem.

(b) *The 84-box Atlantic Ocean model*

Bolin *et al.* (1983) tested the inverse methodology on a 12-box world ocean model. When used to simulate the uptake of fossil-fuel CO₂ emissions, the model gave an airborne fraction over the period 1959–1980 of 0.74–0.75. The ¹⁴C uptake was likewise smaller than indicated by observation. Although reasonably realistic circulation patterns were derived, they concluded that future studies should explore the benefits of finer resolution by application to a part of the world ocean, that temperature and salinity data should be included in addition to dynamically passive tracers, and that the conservation equations should be supplemented by additional dynamical constraints, such as the requirement of quasi-geostrophic flow.

The same group are currently developing an 84-box model of the Atlantic Ocean (Moore & Björkström 1986; Bolin 1986*b*). The ocean is split into 12 regions with up to eight density layers in each region (figure 11). Data from GEOSECS stations within each region define depth profiles for total carbon, ¹⁴C, alkalinity, phosphorus, oxygen, salinity and heat. Material is exchanged with the atmosphere and with the Arctic, Indian and Pacific Oceans and the Mediterranean Sea. The system has 696 conservation equations in 540 unknowns so a least squares solution appropriate for the incompatible problem is sought.

A number of realistic circulation features are reproduced: deep-water formation in regions

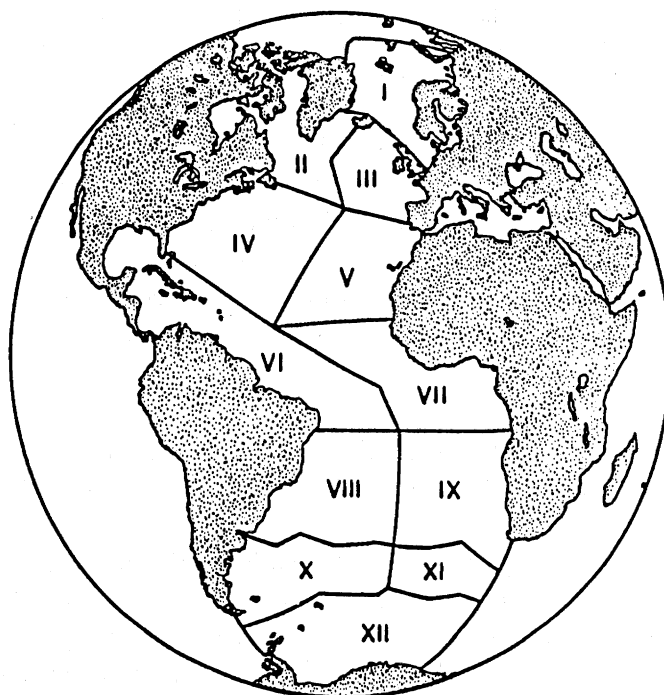


FIGURE 11. The regionalized 84-box model of the Atlantic Ocean of Moore & Björkström (1986). Within each region (depicted by Roman numerals) are up to 8 layers delineated by isopycnal surfaces.

I and III (figure 11), equatorial upwelling, and turbulent exchange between Mediterranean and Atlantic water in regions IV and V. Other features, such as the distribution of biological productivity, are less well reproduced. Preliminary results suggest that the model is more accessible to carbon uptake than the Atlantic component of the 12-box model.

Although the methodology has posed a number of technical problems, there would appear to be potential for further development. Geostrophic-balance equations have been included since the reports published in 1986 were written (A. Björkström, personal communication 1987). Other possible developments include the incorporation of surface wind stress to allow simulation of Ekman pumping. Adoption of even finer resolution may require improved resolution in oceanographic data sets together with a tendency towards under-determined systems of equations, the solution methodologies for which have been less well explored.

6. CLIMATE FEEDBACK ON THE OCEANS

Within the context of carbon-cycle research, ocean models have so far been primarily devoted to establishing the ability of the oceans to take up fossil-fuel CO₂ and hence determining the airborne fraction of man-made emissions. These models have been used also to simulate future atmospheric CO₂ concentrations from scenarios of future emissions (see, for example, Bacastow & Björkström 1981). As has been shown, the CO₂ uptake by different models varies according to the structure and the rates of mixing processes adopted. The study by Peng & Broecker (1985) nevertheless suggests that, at least for the current timescales of man-induced transients, any models that distribute tritium correctly will take up about the same amount of fossil-fuel CO₂. Moreover, in the context of the overall CO₂ issue, the uncertainty in future concentrations is governed much more strongly by uncertainty in future emissions than uncertainty in airborne fraction. Where, then, does the importance of future ocean-carbon modelling lie?

Recent analyses of ice-core CO₂ data have shown that in the past atmospheric CO₂ concentrations have undergone rapid fluctuations of several tens of parts per million by volume over periods as short as 100 years and have coincided with similar fluctuations in the ¹⁸O:¹⁶O ratio, an indicator of prevailing atmospheric temperature (Oeschger & Stauffer 1986). Any viable explanation would appear to require the redistribution of carbon and nutrients within the ocean, leading to changes in productivity in the surface water. This would alter the dissolved CO₂ concentration in the water and hence the atmospheric CO₂ concentration. Broecker *et al.* (1985) review the various theories that have been proposed on this theme. The fundamental line of reasoning is that the atmospheric CO₂ concentration is strongly affected by the surface-ocean CO₂ partial pressure. This in turn is controlled by photosynthesis, which depletes the carbon content of the upper layers. Photosynthesis is regulated by the supply of nutrients, and this is maintained by the ocean circulation, which recycles nutrients from deep layers where biogenic material decays to surface layers where they are involved in biogenic production.

The possibility of a future CO₂-induced warming operating on the ocean biology in this way has encouraged the development of models incorporating ocean productivity. (Formerly marine biological processes were often excluded on the grounds that they do not represent a significant sink for fossil-fuel CO₂ on account of their not being carbon limited.) Baes & Killough's (1986) two-dimensional model designed to explore the influence of biochemical processes on atmospheric CO₂ levels illustrates the effect of gross changes in the parameters controlling atmospheric CO₂ levels. If in the model biological processes are 'turned off' the

atmospheric CO₂ level approximately doubles, whereas a doubling of nutrient concentrations leads to a near halving of atmospheric CO₂ concentration, a result consistent with earlier estimates by Broecker (1982) and Baes (1982). Halved circulation rate reduces atmospheric CO₂ by about 10 %, whereas warming raises the CO₂ level by some 4 % K⁻¹ as expected from the temperature dependence of the distribution of CO₂ between atmosphere and ocean.

In addition to any biologically induced changes in the atmospheric CO₂ level that climate change might cause, direct effects on oceanic CO₂ uptake resulting from circulation changes might also be expected to occur.

It is clear that determining the partitioning of man-made CO₂ emissions between atmosphere and ocean is only the first of a number of increasingly complex and important questions about the atmosphere–ocean system that must be addressed.

Our state of knowledge of ocean circulation, biogeochemical cycling in the oceans, and the likely effects of climatic change on the ocean circulation is at present quite inadequate to address these complex interactions with any predictive capability. Hope of gaining even a qualitative picture of the outcome relies very heavily on continued improvement in modelling capability, which in turn requires the gathering and analysis of the data necessary for their construction and validation. The study of ocean tracers is likely to remain an essential component of this long-term effort.

The work described in §4 and the writing of this paper were carried out at the Central Electricity Research Laboratories and the paper is published with the permission of the Central Electricity Generating Board.

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Discussion

W. ROETHER (*University of Bremen, F.R.G.*). Dr Crane's two-dimensional circulation model assumed a time-averaged circulation. However, both deep-water formation and surface-water p_{CO_2} are seasonally variable to a large degree. Would Dr Crane expect this fact to affect his rates appreciably?

A. J. CRANE. The steady-state cycling of CO₂ between atmosphere and ocean is not treated in the model. The transfers of CO₂ from the atmosphere to the ocean at all latitudes must be interpreted as the departures from steady state arising from increased atmospheric CO₂ levels, namely enhancements in the air–sea fluxes and reductions in the sea–air fluxes when and wherever they occur in the steady state. The influence that seasonality has on the steady-state cycling of CO₂ between atmosphere and ocean is thus not of direct concern. However, as was mentioned in the paper, there is a problem over the extent to which tritium and other tracers are good analogues for CO₂. Because of the differences in surface equilibration rates between CO₂ and other tracers, seasonal variations in p_{CO_2} and rates of surface-water overturning could ensure rates of CO₂ transfer quite different from those estimated on the basis of tracer calibration, depending on the relative rates of uptake and mixing.

P. D. KILLWORTH (*Hooke Institute, University of Oxford, U.K.*). With regard to the part of Dr Crane's talk about box models, even an under-determined set of boxes (merely conserving tracer, say, with no dynamics) may not be able to produce a physically meaningful solution (for example, positive fluxes and diffusions) if the constraints are poor. For example, an interior extremum of tracer cannot be so modelled. Thus we need to choose the boxes with as much physics as possible, and as accurate data as possible.

A. J. CRANE. Dr Killworth is right to emphasize this aspect. The early results from inverse modelling show clearly that physical and dynamical constraints are needed if physically meaningful solutions are to be obtained and if critical oceanic circulation features are to be captured. I think it is generally recognized that progress in this field relies on combining the principles of geophysical fluid dynamics and the properties of tracer distributions, and developing the techniques for solving the increasingly complex models that result.