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1 Short-term evolution of atmospheric CO₂

We have spoken of volcanism as the *long-term* source of CO₂.

Among the other sources, the respiration flux is about 3 orders of magnitude greater than volcanism, and fossil-fuel combustion is about one and one-half orders of magnitude greater.

Whatever the source, we consider the following questions:

- How can we identify the source?
- How long does CO₂ stay in the atmosphere?

We shall answer these questions in a precise way that pertains to short, roughly decadal, time scales.

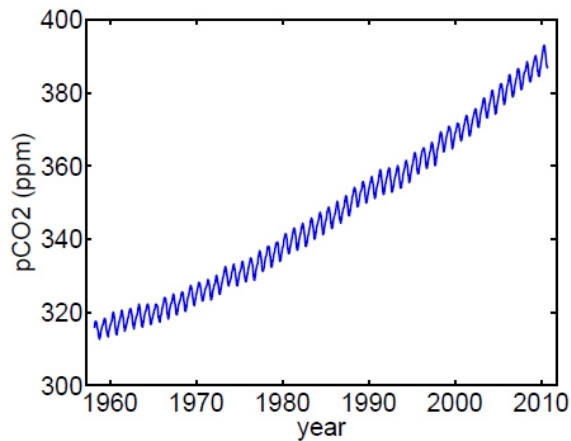
We first consider the growth of atmospheric CO₂ since the mid-20th century.

1.1 The Keeling curve

Atmospheric CO₂ concentrations have been measured monthly at Mauna Loa Observatory, Hawaii, since 1958.

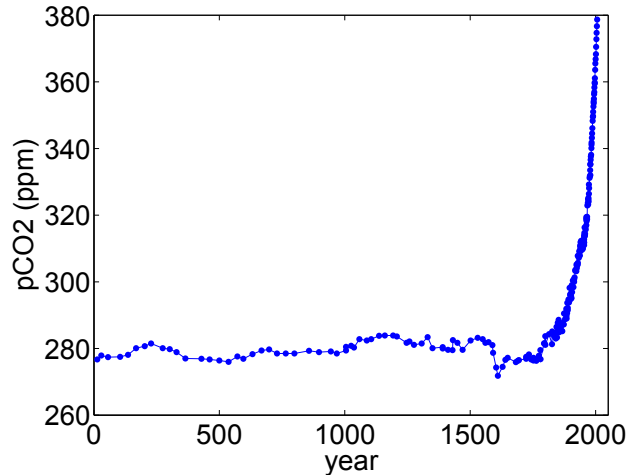
The measurements, begun by C. David Keeling, report CO₂ concentration as a dry mole fraction: the number of molecules of carbon dioxide divided by the number of molecules of dry air multiplied by one million (ppm).

The resulting plot is known as the *Keeling curve*:



Data from P. Tans, NOAA/ESRL (www.esrl.noaa.gov/gmd/ccgg/trends/).

Studies of CO₂ in ice cores show that this increase is the latest chapter in process that started about 200 years ago:



Data from the Law Dome ice core, Antarctica [1].

An obvious question arises: Why is CO₂ increasing?

To provide a precise answer, we first digress to a discussion of carbon isotopes.

1.2 Carbon isotopes

1.2.1 Natural abundance

Reference: Emerson and Hedges [2].

Every atom of carbon has $Z = 6$ protons. Z is the *atomic number*.

When there is no net charge, each atom of C has 6 electrons.

However the *mass number* A varies:

$$A = 12, 13, \text{ or } 14.$$

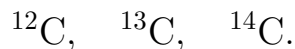
The variation in mass number derives from the variations in the *neutron number*

$$N = A - Z.$$

Each *isotope* of carbon corresponds to a specific neutron number N , and therefore mass number A . These are named according to their mass number:

carbon-12, carbon-13, carbon-14,

and are generally written symbolically as



^{14}C is radioactive, and ^{12}C and ^{13}C are stable.

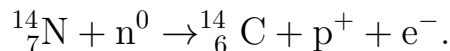
About 98.9% of Earth's carbon is ^{12}C and nearly all the rest is ^{13}C ; thus

$$\frac{^{13}\text{C}}{\text{C}} \sim 10^{-2},$$

where the denominator represents the sum of all carbon. ^{14}C is much less abundant:

$$\frac{^{14}\text{C}}{\text{C}} \lesssim 10^{-12}.$$

^{14}C is naturally produced in the upper atmosphere by cosmic rays, which can shatter a nucleus (N or O), releasing neutrons, some of which are absorbed by ^{14}N such that



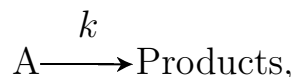
Thus ^{14}N is converted to ^{14}C , releasing a proton and an electron.

1.2.2 Radioactive decay

^{14}C , being unstable, converts back to ^{14}N .

To understand the rate at which such radioactive decay occurs, we consider briefly the following general model.

Consider the reaction



signifying the disappearance or extinction of A with rate constant k .

The meaning of the rate constant k is that, in a small interval of time $\Delta t \ll k^{-1}$,

$$\text{Probability}(\text{an arbitrary atom decays}) \simeq k\Delta t.$$

This probability applies to each atom independently, meaning that, whatever the quantity of A, a fraction $k\Delta t$ of A reacts in an interval Δt .

Thus if we have N atoms of A at time t , after a small time Δt , we have

$$N(t + \Delta t) = N(t) - k\Delta t N(t)$$

and therefore

$$\frac{N(t + \Delta t) - N(t)}{\Delta t} = -kN.$$

Letting $\Delta t \rightarrow dt$, we have the *first-order decay*

$$\frac{dN}{dt} = -kN$$

with solution

$$N(t) = N(0)e^{-kt}.$$

The above development essentially defines a first-order decay process.

Its application to radioactive decay requires merely the assumption that each atom decays with the same constant probability.

For ^{14}C , the rate constant

$$k = \frac{1}{8267 \text{ yr}}.$$

The *half-life* $t_{1/2}$ is the time it takes for one-half of A to decay. Thus

$$\frac{1}{2} = e^{-kt_{1/2}} \quad \Rightarrow \quad t_{1/2} = \frac{\ln 2}{k}$$

For ^{14}C ,

$$t_{1/2} = 5730 \text{ yr}.$$

1.2.3 Notation

Reference: Emerson and Hedges [2].

Geochemists use a very special notation for isotopic abundance.

The first component of the notation is the abundance ratio

$$R_n = \left(\frac{^n\text{C}}{^{12}\text{C}} \right)$$

where $n = 13$ or 14 .

Geochemists then compare the R obtained on a given sample to R obtained for a “standard” sample. Noting the latter ratio as R_{std} , they define the *isotopic composition*

$$\delta^n\text{C} = \left(\frac{R_n - R_{\text{std}}}{R_{\text{std}}} \right) \times 1000.$$

The factor of 1000 is included because the factor in parentheses is often of order 10^{-2} – 10^{-3} , so that when multiplied by 1000 it becomes of order 1–10. One then speaks of δ in units of “per mil”, which is equivalent to “parts per thousand,” and is written with the symbol ‰, not to be confused with the per cent sign % (parts per hundred).

Analyses of ^{14}C contain even greater notational complexity.

The problem arises from the frequent desire to compare a particular measurement of $\delta^{14}\text{C}$ (say, of atmospheric CO_2), to the value of $\delta^{14}\text{C}$ that would be obtained in the absence of anthropogenic perturbation.

Consequently geochemists define the following “cap delta” notation:

$$\Delta^{14}\text{C} = \delta^{14}\text{C} - (\delta^{14}\text{C})_0$$

The second term on the RHS is too complicated to discuss here, but it has the property that if $\delta^{14}\text{C}$ is measured from wood formed in 1850, then $\Delta^{14}\text{C} = 0$.

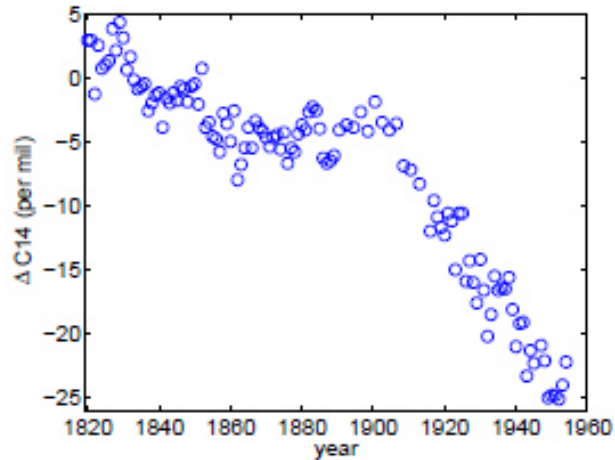
The point is that $\Delta^{14}\text{C} = 0$ represents a “typical” value that represents a balance between atmospheric ^{14}C production and decay.

1.3 The origin of the recent CO_2 rise

CO_2 levels undergo natural fluctuations. Thus it is appropriate to ask whether the recent rise in CO_2 levels are the result of natural changes rather than the burning of fossil fuels.

We can use the ^{14}C content of the atmosphere to help answer this question.

The figure below shows the ^{14}C composition of tree rings associated with the period 1820–1954 [3].



Over the last 50 years, $\Delta^{14}\text{C}$ decreases by about 20‰.

Over such a time period, natural variations in the cosmic ray flux should produce changes no greater than that of experimental error (a few per mil).

The combustion of fossil fuels, on the other hand, should create such a decrease, because the fuels, being derived from geologically ancient organic matter, are entirely free of ^{14}C .

Thus the ^{14}C content of the atmosphere is diluted by the burning of fossil fuels, so that $\Delta^{14}\text{C}$ declines.

This phenomenon, known as the *Suess effect*, is one way to trace the source of the recent CO_2 rise to human activities.

1.4 The bomb spike

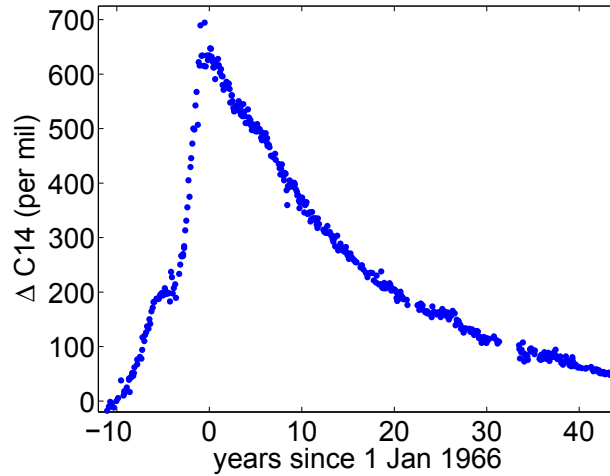
^{14}C can also be used to address the short-term residence time of atmospheric CO_2 .

Nuclear weapons tests in the 1950s and early 1960s produced large amounts of ^{14}C in the atmosphere.

The tests were essentially ended by the Nuclear Test Ban Treaty in October

1963.

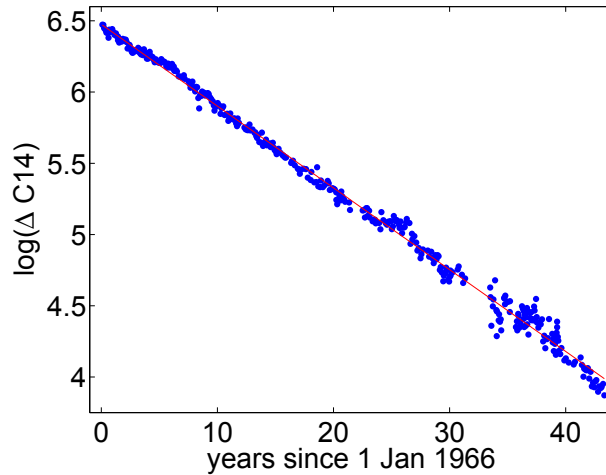
Measurements of atmospheric ^{14}C show the build-up followed by a decay [4]:



The decrease is far too fast to be associated with radioactive decay.

A log-linear plot shows that the decay is nevertheless exponential, dropping off like

$$e^{-t/\tau}, \quad \tau \simeq 17.4 \text{ yr.}$$



The decay is mostly due to “uptake” of ^{14}C in the oceans, in large part by diffusion.

1.5 Microscopic model of molecular diffusion

Reference: Berg [5].

To gain some physical insight, we first consider a microscopic model of diffusion.

1.5.1 One-dimensional random walk

Consider a (drunkard's) random walk along a line:



- Start at time $t = 0$ and position $x = 0$.
- Every τ seconds, take a random step s to the left or right.
- Assume equiprobable steps of equal size δ :

$$P(s = \delta) = P(s = -\delta) = 1/2.$$

- No memory (statistically independent jumps).

We think of this as a caricature of real diffusion (e.g., Brownian motion).

Now consider an ensemble of N independent random walks (i.e., many such drunkards, each acting with no awareness of the others).

Let $x_i(n)$ be the position of the i th walker after n steps. Then

$$x_i(n) = x_i(n - 1) + s.$$

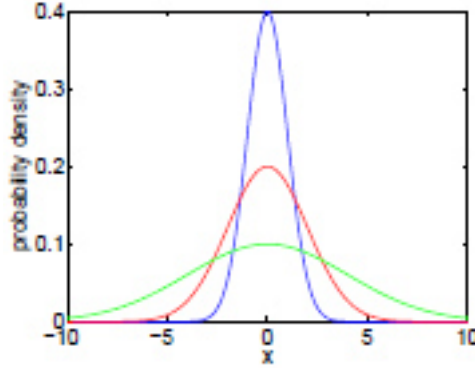
The mean position of a large ensemble of walkers after n steps is

$$\begin{aligned} \langle x(n) \rangle &= \lim_{N \rightarrow \infty} \frac{1}{N} \sum_{i=1}^N (x_i(n - 1) + s) \\ &= \langle x(n - 1) \rangle + \langle s \rangle \\ &= \langle x(n - 1) \rangle. \end{aligned}$$

Here we have used the angle brackets $\langle \cdot \rangle$ to denote the ensemble average. The result shows that the mean position is independent of n , thus retaining permanent memory of the initial condition:

$$\langle x(n) \rangle = 0.$$

Intuitively we understand that there should nevertheless be a wide spread in the probability $P(x)$ that increases with time:



We characterize this spread by the *root-mean-square displacement* $\langle x^2(n) \rangle^{1/2}$. To calculate it, first write

$$\begin{aligned} x_i^2(n) &= [x_i(n-1) + s]^2 \\ &= x_i^2(n-1) + 2sx_i(n-1) + s^2. \end{aligned}$$

Because the mean of a sum of random variables is the sum of the means, the mean-square displacement in the ensemble is

$$\begin{aligned} \langle x^2(n) \rangle &= \langle x^2(n-1) \rangle + 2\langle s x(n-1) \rangle + \langle s^2 \rangle \\ &= \langle x^2(n-1) \rangle + 2\langle s \rangle \langle x(n-1) \rangle + \langle s^2 \rangle \\ &= \langle x^2(n-1) \rangle + \delta^2. \end{aligned}$$

In the second relation, we have replaced the average of a product with the product of averages because s is uncorrelated to x . (This also may be deduced from the observation that the walk contains no memory of past steps.)

Note that our result is in the form of a recursion, which is readily put in the simpler form

$$\langle x^2(n) \rangle = n\delta^2$$

Since $t = n\tau$, we have

$$\langle x^2 \rangle = \delta^2 t / \tau = 2Dt,$$

where we have defined the *diffusion coefficient*

$$D = \frac{\delta^2}{2\tau}.$$

Thus the mean-squared displacement increases linearly with time, like $2Dt$. Consequently the root-mean-square displacement increases like the square-root of time:

$$\langle x^2 \rangle^{1/2} = (2Dt)^{1/2}.$$

Intuitively we understand that the width of a bell-shaped distribution $P(x, t)$ increases like $\sqrt{2Dt}$.

Indeed, in the plot above,

$$\langle x^2 \rangle^{1/2} = 1, 2, \text{ and } 4$$

corresponding to times t such that

$$2Dt = 1, 4, \text{ and } 16.$$

For a small molecule in water, $D \simeq 10^{-5}$ cm²/s. So imagine you're a bacterium (size $\sim 10^{-4}$ cm), and you want to know how long some molecular nutrient will take to diffuse a distance ℓ away from you. Identifying ℓ with $\langle x^2 \rangle^{1/2}$, the diffusion time τ_d is

$$\tau_d \sim \ell^2 / 2D.$$

Consider two particular cases:

ℓ (cm)	τ_d (s)
10^{-4}	5×10^{-4}
1	5×10^4

In other words, the molecule would stay within a length commensurate to a bug's size for only about a millisecond. But it would stay within 1 cm for about 14 hours!

This huge change is a consequence of the quadratic scaling $\tau_d \propto \ell^2$, a hallmark of diffusive processes.

In contrast, for a simple advective flow times scale linearly with distance.

1.5.2 Higher dimensions

Before moving on, we first argue that our little toy problem is equally valid in higher dimensions.

In, say, two dimensions, the random walker is on a plane. In our discrete approximation, this corresponds to a lattice with a “Manhattan metric,” with the drunkard originating at his corner bar and moving $\pm\delta$ in each dimension at each time step.

Because the drunk’s motion in x is independent of his motion in y ,

$$\langle x^2 \rangle = \langle y^2 \rangle = 2Dt$$

Since the mean-square distance from the origin is

$$r^2 = x^2 + y^2,$$

we have

$$\langle r^2 \rangle = 4Dt.$$

The generalization to higher dimensions is obvious. The point is that we retain the diffusive scaling $\ell^2 \propto t$.

1.5.3 Macrodynamics: the diffusion equation

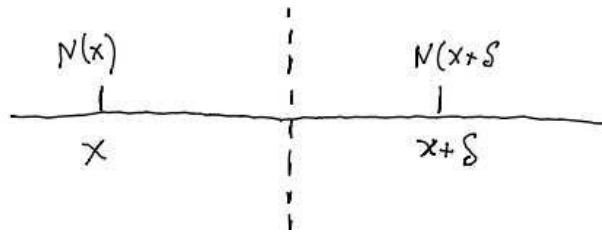
We now proceed to derive the diffusion equation from our random walk.

Suppose we have a long tube of cross-section A in which particles undergo random walks. We are interested in $N(x)$, the number of particles at x (i.e., between $x - \delta/2$ and $x + \delta/2$), along with the particle flux J_x .

How many particles pass through a unit area in unit time, from x to $x + \delta$?
And in the other direction?

In other words, what is the net flux J_x ?

We imagine a boundary between x and $x + \delta$. During one time step τ , half the particles at x cross over to the right, and half the particles at $x + \delta$ cross over to left.



The net flux (number particles per unit area per unit time) is

$$J_x = \left(\frac{N(x)}{2} - \frac{N(x + \delta)}{2} \right) \frac{1}{A\tau}$$

where the factor of $1/2$ comes from the fact that half the particles at each location move away from the boundary rather than towards it.

Rearranging and multiplying by δ^2/δ^2 ,

$$J_x = -\frac{\delta^2}{2\tau} \frac{1}{\delta} \left(\frac{N(x + \delta)}{A\delta} - \frac{N(x)}{A\delta} \right)$$

Defining the number density or concentration $C = N/A\delta$ and recalling $D = \delta^2/2\tau$, we have

$$J_x = -D \frac{C(x + \delta) - C(x)}{\delta}.$$

Letting $\delta \rightarrow 0$, we obtain

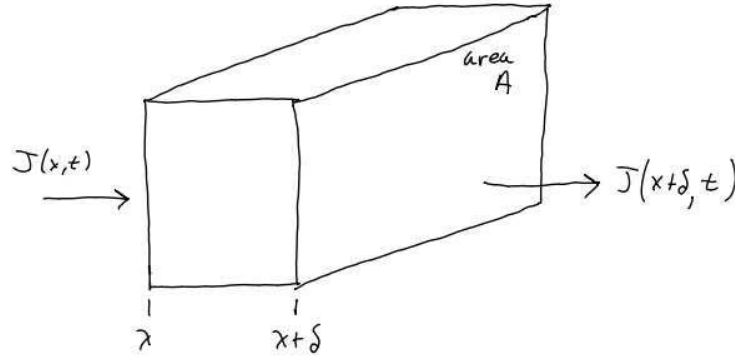
$$J_x = -D \frac{\partial C}{\partial x}.$$

This is Fick's (first) law: the concentration flux goes down the concentration gradient, at a rate given by the diffusivity D .

Fick's law is an example of a "linear-response relation." Others include Ohm's law and Hooke's law. The linearity is essentially an assumption, which follows

in our case from assuming that the two sides of the boundary through which particles flow act independently of one another.

Now consider particles flowing into and out of a box with cross-sectional area A perpendicular to and width δ parallel to the x -axis.



The concentration $C(t)$ inside the box changes with the net flux into it.

In τ units of time the concentration changes as

$$C(t + \tau) - C(t) = (J_x(x) - J_x(x + \delta)) \frac{A\tau}{A\delta}$$

The factor of $A\tau$ converts the concentration flux to the number of particles flowing through the face, and the factor of $1/A\delta$ converts that number to a concentration. Simplifying, we obtain

$$\frac{1}{\tau}(C(t + \tau) - C(t)) = -\frac{1}{\delta}(J_x(x + \delta) - J_x(x)).$$

Letting $\tau \rightarrow 0$ and $\delta \rightarrow 0$, we obtain

$$\frac{\partial C}{\partial t} = -\frac{\partial J_x}{\partial x}$$

Substituting Fick's first law for J_x then yields the *diffusion equation*:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}.$$

These developments can be derived succinctly by an alternative approach. Let

$P_n(i)$ = probability that a random walker is at site i after n steps.

Since steps to the left and right occur with equal probability, we have

$$P_n(i) = \frac{1}{2}P_{n-1}(i+1) + \frac{1}{2}P_{n-1}(i-1)$$

Now set

$$t = n\tau \quad \text{and} \quad x = i\delta$$

and consider the probability to be spread over an interval of size δ so that

$$P_n(i) = \delta \cdot p(x, t).$$

Then

$$p(x, t) = \frac{1}{2}p(x + \delta, t - \tau) + \frac{1}{2}p(x - \delta, t - \tau).$$

Multiplying both sides by $1/\tau$ and rearranging, we have

$$\frac{1}{\tau} [p(x, t) - p(x, t - \tau)] = \frac{\delta^2}{2\tau} \cdot \frac{1}{\delta^2} [p(x + \delta, t - \tau) - 2p(x, t - \tau) + p(x - \delta, t - \tau)]$$

We recognize the LHS as a finite difference in time and the RHS as a finite difference of finite differences in space.

Thus in the limit as $\tau \rightarrow 0$ and $\delta \rightarrow 0$, we have

$$\frac{\partial p}{\partial t} = D \frac{\partial^2 p}{\partial x^2}, \quad D = \frac{\delta^2}{2\tau}$$

expressing the diffusion of probability.

Reverting back to the concentration C , note that in higher dimensions, Fick's first law is

$$\vec{J} = -D\nabla C$$

and mass conservation yields

$$\frac{\partial C}{\partial t} = -\nabla \cdot \vec{J}.$$

Combining the two, we have

$$\frac{\partial C}{\partial t} = D\nabla^2 C,$$

which may be straightforwardly obtained by generalization of our random walk to higher dimensions.

Note that our expression of the diffusion equation is precisely the same as the heat equation we derived purely by continuum arguments in an earlier lecture.

By rederiving the diffusion equation via a random walk, however, we have exposed the connection of diffusion to random motion.

One might wonder if the explicit microdynamics matter. For example, do things change if there is a distribution of step sizes, or waiting times?

A more detailed analysis shows that only the coefficient D changes, not the diffusion equation itself, provided that the step size and waiting time distributions are not too wide.

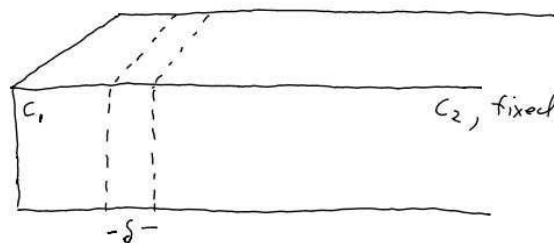
Conclusion: The simplest possible random walks are solutions to the diffusion equation. Consequently:

- We can think about diffusive processes as random walks.
- We can equally think about random walks as diffusive.

1.6 Relaxation to equilibrium

As an elementary application of what we have just learned, suppose that we have two well mixed (e.g., stirred) reservoirs of gas separated by permeable walls a distance δ apart from one another.

We assume that the concentration of CO_2 in reservoir 2 is held fixed at c_2 , and that the concentration c_1 in the first reservoir initially differs from c_2 .



In other words, we require that reservoir 1 equilibrate with reservoir 2, and take reservoir 2 to be so large compared with reservoir 1 that any changes to its concentration are negligible.

The characteristic time τ for a molecule of CO_2 to diffuse from one permeable boundary to the other is

$$\tau = \delta^2/D,$$

where D is the diffusion coefficient.

We assume that the time taken to pass through a permeable wall is much less than τ . Consequently δ is the thickness of a *diffusive boundary layer* between the two reservoirs.

Clearly CO_2 diffuses so that $c_1 \rightarrow c_2$. How, then, does c_1 evolve with time?

The flux J through the boundary layer, from reservoir 1 to 2, is obtained from Fick's law:

$$J = -D \frac{c_2 - c_1}{\delta}.$$

J has dimensions

$$[J] = \frac{L^2}{T} \frac{1}{L} \frac{M}{L^3} = \frac{M}{L^2 T},$$

i.e., mass per unit time per unit area.

Letting A represent the area of the boundary between reservoir 1 and 2 and V_1 be the (constant) volume of reservoir 1, we have

$$V_1 \frac{dc_1}{dt} = -JA = \frac{DA}{\delta}(c_2 - c_1),$$

Defining

$$\tilde{c} = c_1 - c_2,$$

we have

$$\frac{d\tilde{c}}{dt} = -\frac{DA}{V_1 \delta} \tilde{c}.$$

which has the solution

$$\tilde{c}(t) = \tilde{c}(0)e^{-t/\tau}$$

where the time constant

$$\tau = \frac{V_1 \delta}{DA}. \quad (1)$$

This simple exponential decay is a characteristic of physical problems defined by a single time scale and a linear response relation.

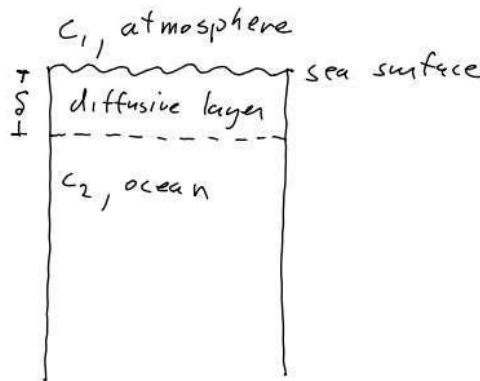
1.7 Air-sea transfer and the decay constant

Reference: Broecker and Peng [6].

We return now to the problem of the decay of the bomb spike.

In essence, we apply the model of the previous section, where reservoir 1 is the atmosphere and reservoir 2 is the ocean.

The key idea is that the transfer from air to sea occurs through a thin layer of water, of thickness δ .



Above this layer, the atmosphere is considered well mixed, with a constant concentration of ^{14}C .

Below the layer, the ocean is also considered well mixed.

Within the layer, there is a diffusion gradient—thus we say that absorption into the ocean is *diffusion-limited*.

This model of air-sea transfer is known as the *stagnant film* model.

The central problem is to determine the effective thickness δ of the diffusive boundary layer.

1.7.1 Thin-film model

To determine δ , we make use of the following:

- ^{14}C is created in the atmosphere.
- The typical residence time of CO_2 in the atmosphere is much shorter than the radiocarbon decay constant ($k^{-1} = 8267 \text{ yr.}$)
- Nearly all of the decay of ^{14}C takes place in the oceans.

Thus the atmosphere is the source, and the oceans a sink.

We therefore expect that, prior to the bomb spike, a steady state in which the production of ^{14}C in the atmosphere is balanced by its decay in the oceans.

Then the differences in the concentration of ^{14}C in the oceans and atmosphere would be due to a balance between ^{14}C absorbed into the oceans at the rate given by

$$\text{gas transfer velocity} = D/\delta$$

and the ^{14}C “destroyed” in the oceans at the decay time scale k^{-1} .

The flux per unit area of ^{14}C into the oceans is again given by

$$J = -\frac{D}{\delta}(c_2 - c_1)$$

where now

c_1 = concentration of $^{14}\text{CO}_2$ in the water at the air-sea interface

c_2 = $^{14}\text{CO}_2$ concentration just below diffusive layer, in the “mixed” layer.

The total input flux is then

$$^{14}\text{C input} = JA,$$

where A is the area of the air-sea interface.

The decay of ^{14}C in the oceans, on the other hand, is given by

$$^{14}\text{C decay} = kc_oV_o$$

where c_o is the average radiocarbon concentration in the whole ocean (essentially the deep ocean), and V_o is the volume of the oceans.

Balancing the input with the decay, we have

$$\frac{DA}{\delta}(c_1 - c_2) = kc_oV_o$$

and therefore the effective thickness of the stagnant film is

$$\delta = \frac{DA}{kV_o} \left(\frac{c_1 - c_2}{c_o} \right)$$

The ratio

$$V_o/A = \text{mean ocean depth} \simeq 3800 \text{ m.}$$

Analyses of radiocarbon in the atmosphere and oceans provide the pre-industrial, pre-bomb concentration ratio

$$\frac{c_1 - c_2}{c_o} \simeq 2.8 \times 10^{-4}.$$

The diffusivity of CO_2 gas in water is about

$$D \simeq 5 \times 10^{-2} \text{ m}^2/\text{yr.}$$

We thus find the effective thin film thickness

$$\delta = 30 \text{ } \mu\text{m}$$

and the gas transfer velocity

$$D/\delta = 1800 \text{ m/yr} \simeq 20 \text{ cm/hr.}$$

1.7.2 Decay constant of the bomb spike

We now apply the simplified model of decay of Section 1.6. There we predicted the decay time constant

$$\tau = \frac{V_1/A}{D/\delta}.$$

Here we have

$$\frac{V_1}{A} = \text{equivalent height of atmosphere} \simeq 8500 \text{ m}$$

where “equivalent height” means the height that the atmosphere would have if the atmospheric pressure everywhere were the same as at sea level.

Then

$$\tau \simeq 4.7 \text{ yr.}$$

Given the simplicity of the models, this prediction compares reasonably well to the time constant of 17.4 yr measured from the decay of the bomb spike in Section 1.4.

Since most of the ^{14}C uptake should be in the oceans, this result suggests that the predicted uptake is about 4 times faster than the observed uptake.

There are many possible sources of error. Some possibilities:

- The “well-mixed” assumption, which derives from an assumption about turbulence, does not account for the effectively thicker films due to wave action.
- The ocean is not a perfect sink for ^{14}C .

1.7.3 Relation to the residence time of atmospheric CO_2

In reality, the uptake of CO_2 into the oceans, land, and ultimately rocks occurs at many time scales, some as long as 100 Kyr.

Here we have addressed only the fastest of these time scales, as it manifests itself (roughly) in the decay of the bomb spike.

In this case, we merely observe the absorption into the oceans of a kind of passive tracer.

However, when CO_2 is injected into the atmosphere (rather than “labeling” a particular kind of CO_2 as $^{14}\text{CO}_2$), CO_2 levels in the oceans rise in response.

There is then an intermediate time scale, longer than that studied here, but shorter than the 100 Kyr time scale of the rock cycle, wherein the atmosphere equilibrates not back to its original CO₂ concentration, but that which brings it into equilibrium with the change in the oceans.

Indeed, models suggest that 20-35% of the increase in atmospheric CO₂ remains in the atmosphere at a time scale of 200–2000 yr [7,8].

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