The concept of age in marine modelling
I. Theory and preliminary model results

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Abstract

The age of a particle of a seawater constituent is defined to be the time elapsed since the particle under consideration left the region, in which its age is prescribed to be zero. An Eulerian theory of the age is presented, in which advection, diffusion, production and destruction phenomena are properly accounted for. The key hypothesis is that the mean age of a set of particles is to be evaluated as the mass-weighted average of the ages of the particles under study. The basic variable is the concentration distribution function, representing, at a given time and location, the distribution over the age of the concentration of the constituent being considered. This function satisfies a partial differential equation, which, upon appropriate integration over the age, yields the equations, in flux form, governing the evolution of the concentration and the age concentration. The ratio of the latter variable to the former is the mean age. Further theoretical developments are presented, including a thought experiment showing that mixing processes cause the ages of various constituents to be different from each other.

The potential of the age as a tool for understanding complex marine flows is briefly demonstrated by analysing the results of two numerical models. The ages of a passive tracer, a radioactive tracer and the water are computed, along with a suitably defined radio-age. First, the fate of tracers released into the English Channel at La Hague is simulated. Then, ages are computed in the World Ocean as a measure of the time that has elapsed since leaving the surface layers. A theorem is demonstrated, which specifies that the age of the radioactive tracer must be smaller than the relevant radio-age, the latter being smaller than the age of the passive tracer, which, under appropriate hypotheses, can be seen to be equivalent to the age of the water. These inequalities seem to be remarkably robust, since they are found to hold valid in most of the numerical and analytical results examined in the present study. On the other hand, a dimensionless number is highlighted, which is believed to play an important role in the scaling of the differences between ages. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction: the concept of age

Most marine flows are so complex that it may prove difficult to gain a profound insight into their functioning by means of a simple inspection of the

available values of their state variables, be they measured in situ or simulated numerically. This is why specific interpretation techniques are often needed, some of which demand that auxiliary variables be estimated. The age is one such auxiliary variable. The definitions of the age that were given so far (e.g. Bolin and Rodhe, 1973; Jenkins and Clarke, 1976; Zimmerman, 1976; Dronkers and Zimmerman, 1982; Stuiver et al., 1983; Prandle, 1984; Takeoka, 1984; Jenkins, 1987; Thiele and Sarmiento, 1990; Hall and Plumb, 1994; Hirst, 1994; England, 1995; Salomon et al., 1995; Anonymous, 1997; Boon et al., 1997; Karstensen and Tomczak, 1998; Campin et al., 1999; Delhez et al., 1999a; Hirst, 1999; Hall and Haine, 2000), though not equivalent, all view the age as a measure of an elapsed time. Herein, in accordance with Delhez et al. (1999a), the age of a particle of a constituent of seawater is regarded as "the time elapsed since the particle under consideration left the region in which its age is prescribed to be zero".

Schematically, the age was used so far for two main classes of purposes, i.e. estimating the ventilation rate of ocean basins (e.g. Jenkins and Clarke, 1976; Stuiver et al., 1983; Jenkins, 1987; Doney and Jenkins, 1988; Thiele and Sarmiento, 1990; Broecker et al., 1990; Doney and Jenkins, 1994; England, 1995; England and Rahmstorf, 1999; England and Maier-Reimer, 2001; Haine and Richards, 1995; Beining and Roether, 1996; England and Holloway, 1998; Campin et al., 1999) or inferring the horizontal circulation of shelf seas (e.g. Prandle, 1984; Salomon et al., 1995; Anonymous, 1997; Boon et al., 1997; Delhez and Deleersnijder, 2001). In the former type of applications, the age of a water parcel was considered as the time elapsed since it left the ocean surface, while in the latter, the age of a tracer particle was usually estimated as the time elapsed since it was released into the sea by a point source.

Obviously, the concept of age is intimately associated with that of water parcel, constituent particle, etc. Therefore, it is necessary that such notions be clearly defined first. Seawater is a mixture of several constituents, i.e. pure water, dissolved salts, pollutants, plankton, etc. It is pointless—and, in practice, impossible—to study in detail the motion of every molecule of every constituent, because there are too many of them. Instead, seawater is modelled as a continuous medium, an approach adopted in most fluid mechanics problems. Accordingly, the state variables of the flow under study—i.e. seawater velocity, pressure, temperature, constituent concentration, etc.—are assumed to be defined at every time and location in the domain of interest. This approach, which is usually called Eulerian, is that resorted to in the mathematical developments of the present article. However, for the sake of simplicity, several explanations will be given in Lagrangian terms. In the Lagrangian description, every constituent contained in the domain of interest is considered to be concentrated in pointwise particles, of which the properties and displacements are to be studied. In practice, for the results of a Lagrangian simulation to tend to the exact solution of the problem, the mass of any Lagrangian particle of a given constituent must be much smaller than the total mass of the constituent under consideration, but may be much larger than that of one molecule of the latter. So, as the present study is concerned only with the processes taking place at macroscopic scales, the mass of a Lagrangian particle will be considered to be arbitrarily small.

Let t denote time and $\mathbf{x}$ represent the position vector of any point in the domain of interest. At time t and location $\mathbf{x}$, the velocity of seawater, $\mathbf{u}(t,\mathbf{x})$, is a suitably defined weighted average of the velocities of the particles of the constituents of seawater. A material volume is a fictitious, moving, and deforming volume, delineated by thought only, of which the velocity of every point is equal to the velocity of seawater at the point considered. Then, a seawater— or fluid—parcel is a very small material volume: its characteristic size must be much smaller than that of the smallest length scale of the flow under study and must be much larger than the typical distance between the molecules of the seawater constituents. Because of its smallness, a fluid parcel can be identified only by the location of its centre and its volume, which is regarded as arbitrarily small from the macroscopic point of view the present study is concerned with.

Most methods for estimating the age rely on the assumption that diffusive or mixing processes may be neglected, implying that they may be outlined within a common framework. Any transport of matter proceeding at velocity $\mathbf{u}(t,\mathbf{x})$—at time t and

location $\mathbf{x}$—is viewed as advection. All other types of transport are not explicitly resolved by the model under study, and are generally parameterised as diffusive or mixing phenomena, since, in most cases, the related fluxes of matter tend to reduce the spatial contrasts of the concentration of the constituent transported. If mixing is negligible, a seawater parcel is made up at any time of the same particles (Fig. 1a). As a consequence, all the particles of all the constituents of seawater contained in a given fluid parcel share the same history, implying that they may be ascribed a unique age.

Consider the fluid parcel following path $\mathbf{r}(t)$, where $\mathbf{r}(t)$ represents the position vector of the fluid parcel under study at time t. By definition of the notion of trajectory, $\mathbf{r}(t)$ is related to the fluid velocity by the expression

$$\frac{d\mathbf{r}(t)}{dt} = \mathbf{u}[t,\mathbf{r}(t)]. \qquad (1.1)$$

As diffusive processes are neglected for the moment, time and the age $a[t,\mathbf{r}(t)]$ of the fluid parcel under study must progress at the same pace since the age is a measure of the time elapsed, implying that

$$\frac{dt}{dt} = 1 = \frac{d}{dt} a[t,\mathbf{r}(t)]. \qquad (1.2)$$

Hence,

$$a[t^2,\mathbf{r}(t^2)] = a[t^1,\mathbf{r}(t^1)] + (t^2 - t^1), \qquad (1.3)$$

where t^1 and t^2 are two arbitrarily chosen instants (Fig. 2).

Assume that the fluid parcel whose trajectory is $\mathbf{r}(t)$ contains a passive constituent, i.e. a constituent that is neither produced nor destroyed. Therefore, its concentration $C_p[t,\mathbf{r}(t)]$ within the fluid parcel remains constant as time progresses:

$$C_p[t^2,\mathbf{r}(t^2)] = C_p[t^1,\mathbf{r}(t^1)]. \qquad (1.4)$$

If a source causes the concentration of the passive constituent to vary in time, the fluid parcels passing through the source region will reflect the history of the variations of the concentration of the passive constituent in this region. Thus, as the passive constituent concentration must remain constant within a fluid parcel once it has left the source region, measuring the concentration of the passive constituent in a fluid parcel, originating from the source region, may help to estimate the age of the fluid parcel as the time that has elapsed since it left the source region—if the time variation of the concentration in the source region is known. This was taken advantage of by Salomon et al. (1995) to evaluate the age of tracers released into the English Channel by the nuclear fuel reprocessing plant of Cap de La Hague. On the other hand, as the history of the input into the World Ocean of certain tracers, such as CFCs, is rather well known, measuring the concentration of CFCs in a water sample taken at depth helps to estimate the age of this water sample, which is defined as the time that has elapsed since it left the oceanic surface layers (e.g. Haine and Richards, 1995; Beining and Roether, 1996; England and Holloway, 1998; England and Maier-Reimer, 2001).

Now, hypothesise that the fluid parcel under study also contains a radioactive isotope, the half-life of which is $T\log 2$. As is well known, the concentration
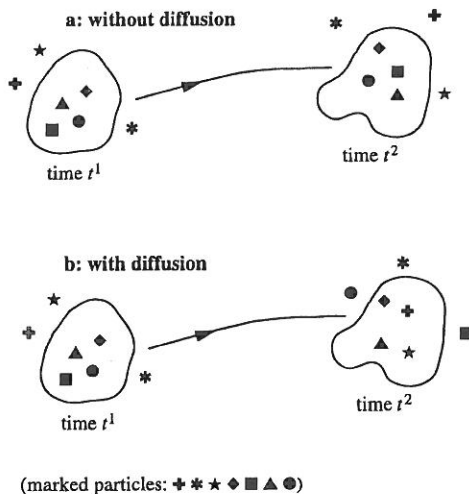
a: without diffusion

time t^1

time t^2

b: with diffusion

time t^1

time t^2

(marked particles: ✚ ✳ ★ ◆ ■ ▲ ●)

Fig. 1. Schematic illustration of the displacement of a fluid parcel, displayed at instants t^1 and t^2, along with marked constituents particles. Without diffusion (a), the fluid parcel is isolated from its environment, so that marked particles do not enter—or leave—it as time progresses. By contrast, if diffusion processes are assumed to be significant (b), the fluid parcel considered exchanges matter with its environment.
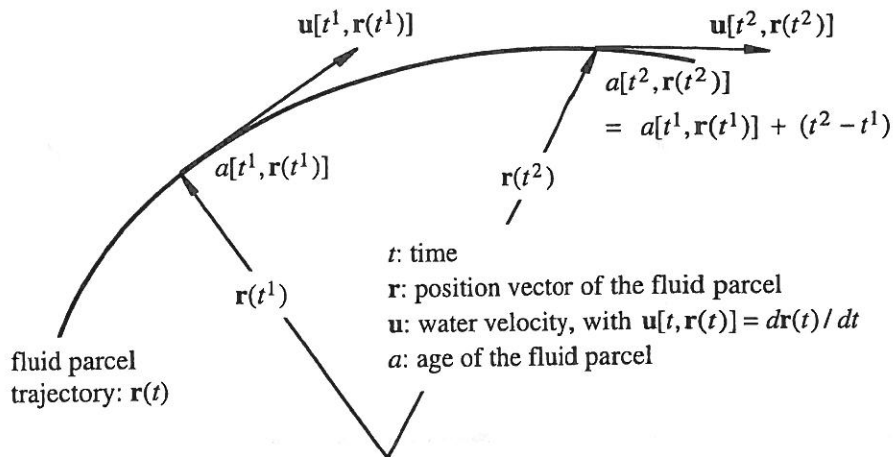
Fig. 2. Trajectory $\mathbf{r}(t)$ of a fluid parcel, with its age at instants t^1 and t^2 if diffusive processes are negligible.

$C_r[t,\mathbf{r}(t)]$ within the fluid parcel of this radioactive constituent decreases as:

$$C_r\left[t^2,\mathbf{r}(t^2)\right] = C_r\left[t^1,\mathbf{r}(t^1)\right]\exp\frac{t^1 - t^2}{T}. \qquad (1.5)$$

Then, combining Eqs. (1.3)–(1.5) yields

$$a\left[t^2,\mathbf{r}(t^2)\right] = a\left[t^1,\mathbf{r}(t^1)\right] + T\log\frac{C_r\left[t^1,\mathbf{r}(t^1)\right]}{C_r\left[t^2,\mathbf{r}(t^2)\right]}, \qquad (1.6)$$

or

$$a\left[t^2,\mathbf{r}(t^2)\right] = a\left[t^1,\mathbf{r}(t^1)\right]$$
$$+ T\log\frac{C_r\left[t^1,\mathbf{r}(t^1)\right]/C_p\left[t^1,\mathbf{r}(t^1)\right]}{C_r\left[t^2,\mathbf{r}(t^2)\right]/C_p\left[t^2,\mathbf{r}(t^2)\right]}. \qquad (1.7)$$

Thus, measuring the concentration of a radioisotope in a water sample, or the ratio of the concentration of a radioisotope to that of a stable one, may lead to an estimate of the age of this sample. For instance, ventilation rates of ocean basins were estimated with the help of measured or numerically simulated carbon-14 and carbon-12 concentrations (e.g. Shackleton et al., 1988; Broecker et al., 1990; Adkins and Boyle, 1997; England and Rahmstorf, 1999; England and Maier-Reimer, 2001). Moreover, for studying motions developing on much shorter timescales ($\approx$

1–10 years) in the ocean surface layers and the main thermocline, measurements of the concentrations of tritium and helium-3 were used (e.g. Jenkins and Clarke, 1976; Jenkins, 1987, 1988; Doney and Jenkins, 1988, 1994; Doney et al., 1997; England and Maier-Reimer, 2001). Finally, other radioactive tracers proved to be useful for evaluating the age of water masses (e.g. Sarmiento et al., 1990; England and Maier-Reimer, 2001).

The methods outlined above are capable of providing age estimates from field data. It is readily understood that they can be implemented in numerical models too. However, as was already pointed out, these techniques rest on the key assumption that diffusive processes are negligible. Since these phenomena may prove to play a significant role, even on oceanic transport at the largest possible scales, several authors realised (e.g. Jenkins and Clarke, 1976; Jenkins, 1988; Thiele and Sarmiento, 1990; Wallace et al., 1992; Hirst, 1994; England and Holloway, 1998; Karstensen and Tomczak, 1998; Delhez et al., 1999a) that mixing processes must be taken into account in an appropriate way.

When diffusive processes are significant, a fluid parcel may no longer be considered to be isolated from its environment (Fig. 1b). Instead, there are matter fluxes entering and leaving the fluid parcel under study. As the age of the particles entering the fluid parcel by diffusion is unlikely to be equal to those already present in this fluid parcel, it ensues that the history of a given particle of a seawater

constituent present in the fluid parcel under study is likely to be different from that of every other particle included in the same fluid parcel. Furthermore, production/destruction processes may also alter the distribution of the age within a fluid parcel. Therefore, to address these issues, Delhez et al. (1999a) pointed to the need for a *general theory of the age*, the main features of which are:

1. *the age of every constituent of seawater can be evaluated, as well as that of suitably defined groups of constituents;*
2. *the age of every constituent depends on time and position;*
3. *advection, mixing, and production / destruction processes are properly accounted for.*

Such a theory is outlined in Delhez et al. (1999a), and presented in detail in the present article. It is intended for mathematical and numerical models. Nonetheless, some of its aspects are believed to be useful to studies relying exclusively on field data to estimate the age.

The age of a constituent is a characteristic that a fluid parcel or constituent particle carries with itself, i.e. it is a Lagrangian concept. Lagrangian models can therefore seem to be best suited to model the age distribution of any constituent. It is indeed simple matter to implement the computation of ages in such a model; one needs just to attach a "clock" to each particle and to initialise that clock when the particle passes through the region where the age is prescribed to be zero. This approach turned to be appropriate for the study of point sources and for theoretical issues (e.g. Hall and Haine, 2000).

To model the fate of tracers with more continuous sources in space and time, to study the concentration field away from the discharge locations, or to simulate mutually interacting tracers, Eulerian transport models are however usually preferred to their Lagrangian counterparts. The obvious reasons are then the need for a huge number of Lagrangian particles required to produce synoptic maps, the associated demand for large computer time and the theoretical problems associated with the modelling of turbulence in nonhomogeneous conditions (e.g. Hunter et al., 1993; Visser, 1997).

As far as the age is concerned, adopting the Eulerian approach is further motivated by the fact that the age is often computed for investigating the functioning of the underlying hydrodynamic model itself. The age is indeed a diagnostic quantity and should therefore be evaluated by means of an algorithm as close as possible to those used in the hydrodynamic model, which is usually Eulerian. This is why it is desirable that the equations governing the age be derived and utilised in the Eulerian framework.

In Section 2, the general age theory is established. Next, in Section 3, some important consequences of the theory are examined. Then, a range of numerical experiments is carried out to investigate the impact of mixing on the age, with a particular focus on the differences between the age of the water, the age of a passive tracer, the age of a radioactive tracer, as well as a suitably-defined radio-age, i.e. a fluid parcel age estimated from the concentration of a passive and a radioactive constituent, assuming that mixing is negligible. In Section 4, timescales of the long-term transport through the English Channel and the southern North Sea are computed, essentially by modelling the age of tracers released by the La Hague nuclear fuel reprocessing plant. An idealised version of this problem is seen to admit analytical solutions, in which a dimensionless number governs the order of magnitude of the differences between various ages. In Section 5, ages computed by means of a model of the World Ocean are analysed and theoretical results related to the numerical simulations are established. Finally, in Section 6 conclusions are drawn and future work is outlined.

2. General theory of the age

As was pointed out in Section 1, to take into account diffusive and production/destruction phenomena, a general theory of the age must cope with the fact that every fluid parcel is likely to contain particles of seawater constituents having different ages. In this respect, it is first necessary to work out a method for specifying how the age is distributed in every fluid parcel. To do so, inspiration may be

found in previous works (e.g. Bolin and Rodhe, 1973; Dronkers and Zimmerman, 1982; Hall and Plumb, 1994; Holzer and Hall, 2000), who had recourse to a concentration distribution function. Such a quantity is also needed herein, and will be considered in a rather general acceptation.

2.1. Concentration distribution function

In the general mathematical developments below, the constituents of seawater are identified by the subscript "i" ($0 \le i \le I$). The latter is defined in such a way that pure water corresponds to $i = 0$. However, in certain particular applications concerned only with the fate of a passive and a radioactive tracer, the subscripts "p" and "r" will be preferred to refer to the former and the latter, respectively.

As in most marine models, the Boussinesq approximation is invoked: the variations of the density of seawater, ρ, are assumed to be negligible, implying that the divergence of the seawater velocity is considered to be zero. Then, the concentration distribution function is defined as follows (Delhez et al., 1999a):

Consider the fluid parcel of volume ΔV, that at time t has its center at location x; the total mass of the particles of the i-th constituent contained in this fluid parcel, of which the age lies in the interval $[\tau - \Delta\tau/2, \ \tau + \Delta\tau/2]$, is asymptotic to $\rho\Delta V\Delta\tau c_i(t,\mathbf{x},\tau)$, as $\Delta V \to 0$ and $\Delta\tau \to 0$, where $c_i(t,\mathbf{x},\tau)$ is the concentration distribution function of the i-th constituents; this definition holds true for any fluid parcel in the domain of interest.

Clearly, in this definition the age τ is an independent variable, which must not be mistaken for the mean age of the i-th constituent, a dependent variable which is defined below and denoted as a_i.

The mass of the i-th constituent contained in the fluid parcel under consideration, $m_i(t,\mathbf{x},\Delta V)$, satisfies the asymptotic expression

$$m_i(t,\mathbf{x},\Delta V) \sim \rho\Delta V\int_0^\infty c_i(t,\mathbf{x},\tau)\mathrm{d}\tau, \qquad \Delta V \to 0. \tag{2.1}$$

Hence, the mass $m(t,\mathbf{x},\Delta V)$ of the fluid parcel is

$$m(t,\mathbf{x},\Delta V) \sim \sum_{i=0}^{I} m_i(t,\mathbf{x},\Delta V), \qquad \Delta V \to 0. \tag{2.2}$$

In the marine science literature, several definitions of the concentration of a constituent of seawater may be found. Herein, the concentration $C_i(t,\mathbf{x})$ of the i-th constituent is taken to be the dimensionless mass fraction

$$C_i(t,\mathbf{x}) \sim \frac{m_i(t,\mathbf{x},\Delta V)}{m(t,\mathbf{x},\Delta V)}, \qquad \Delta V \to 0. \tag{2.3}$$

Combining relations (2.1)–(2.3), it is readily seen that the concentrations of the seawater constituents must obey the constraint,

$$\sum_{i=0}^{I} C_i(t,\mathbf{x}) = 1, \tag{2.4}$$

and that the concentration of every constituent is related to the corresponding concentration distribution function by the integral

$$C_i(t,\mathbf{x}) = \int_0^\infty c_i(t,\mathbf{x},\tau)\mathrm{d}\tau. \tag{2.5}$$

As the concentration of any constituent may be evaluated once the corresponding concentration distribution function is known, the issue to be addressed now is thus the following: what is the equation governing the evolution of the concentration distribution function of every seawater constituent? The answer to this question is provided by examining the budget of the mass of every constituent over an appropriate control volume.

Let x, y, and z denote Cartesian coordinates such that the position vector of any point in the domain of interest reads $\mathbf{x} = (x, y, z)$. Consider the four-dimensional volume element $\Delta\Omega$ in the four-dimensional space comprising the physical—three-dimensional—space, associated with coordinates x, y, and z, and the age dimension, associated with coordinate τ. This volume element consists of the physical volume element

$$[x - \Delta x/2, x + \Delta x/2] \times [y - \Delta y/2, y + \Delta y/2]$$
$$\times [z - \Delta z/2, z + \Delta z/2], \tag{2.6}$$

and the age interval

$$[\tau - \Delta\tau/2, \tau + \Delta\tau/2], \qquad (2.7)$$

with

$$\Delta x, \Delta y, \Delta z, \Delta\tau \to 0. \qquad (2.8)$$

The mass of the i-th constituent contained in $\Delta\Omega$ varies as a result of local production/destruction, due to processes such as chemical reactions or radioactive decay, and transport through the boundaries of $\Delta\Omega$. Therefore, the mass budget of $\Delta\Omega$ reads:

$$\rho\Delta x \Delta y \Delta z \Delta\tau \frac{\partial}{\partial t} c_i(t, \mathbf{x}, \tau)$$

$$= \rho\Delta x \Delta y \Delta z \Delta\tau (p_i - d_i)$$

$$- \rho\Delta y, \Delta z \Delta\tau \big[q_{i,x}(t, x + \Delta x/2, y, z, \tau)$$

$$- q_{i,x}(t, x - \Delta x/2, y, z, \tau) \big]$$

$$- \rho\Delta x, \Delta z \Delta\tau \big[q_{i,y}(t, x, y + \Delta y/2, z, \tau)$$

$$- q_{i,y}(t, x, y - \Delta y/2, z, \tau) \big]$$

$$- \rho\Delta x \Delta y \Delta\tau \big[q_{i,z}(t, x, y, z + \Delta z/2, \tau)$$

$$- q_{i,z}(t, x, y, z - \Delta z/2, \tau) \big]$$

$$- \rho\Delta x \Delta y \Delta z \big[q_{i,\tau}(t, x, y, z, \tau + \Delta\tau/2)$$

$$- q_{i,\tau}(t, x, y, z, \tau - \Delta\tau/2) \big], \qquad (2.9)$$

where $p_i (\geq 0)$ and $d_i (\geq 0)$ are the rates of production and destruction, respectively, i.e. the source and sink terms; $q_{i,x}$, $q_{i,y}$, $q_{i,z}$, and $q_{i,\tau}$ denote the fluxes in the x, y, z, and τ directions.

In the limit $\Delta x, \Delta y, \Delta z, \Delta\tau \to 0$, Eq. (2.9) yields

$$\frac{\partial c_i}{\partial t} = p_i - d_i - \nabla \cdot \mathbf{q}_i - \frac{\partial q_{i,\tau}}{\partial \tau}, \qquad (2.10)$$

where $\mathbf{q}_i = (q_{i,x}, q_{i,y}, q_{i,z})$ and ∇ is the nable or del vector operator in the physical space, so that $\nabla \cdot \mathbf{q}_i = \partial q_{i,x}/\partial x + \partial q_{i,y}/\partial y + \partial q_{i,z}/\partial z$.

The flux $\mathbf{q}_i$ represents the transport of mass in the physical space, a phenomenon which may be safely assumed to be independent of the age of the particles being transported. The flux $\mathbf{q}_i$ consists of an advective part, due to the fluid velocity $\mathbf{u}$, and a contribution ensuing from the transport phenomena that are not explicitly resolved, which is parameterised in a Fourier–Fick manner with the help of the eddy diffusivity tensor $\mathbf{K}$. Accordingly, $\mathbf{q}_i$ reads

$$\mathbf{q}_i = \mathbf{u} c_i - \mathbf{K} \cdot \nabla c_i. \qquad (2.11)$$

The expression of the diffusive flux, $-\mathbf{K} \cdot \nabla c_i$, is sufficiently general that it is capable of taking into account the impact of a wide variety of phenomena, ranging from the three-dimensional turbulent motions (e.g. Mellor and Yamada, 1982; Rodi, 1993), the typical scale of which does not exceed a few metres, to the synoptic-scale eddies that most ocean general circulation models cannot, yet, resolve (e.g. Redi, 1982; Cox, 1987). Convective adjustments can also be considered as a particular type of diffusive phenomenon (e.g. Marotzke, 1991). Obviously, the formulation of $\mathbf{K}$ depends on the nature of the motions that are unresolved in the model under consideration. Nonetheless, as pointed out in Appendix A, tensor $\mathbf{K}$ may be assumed to be symmetric and positive-definite: $\mathbf{K}$ satisfies

$$\mathbf{K} = \mathbf{K}^T, \qquad (2.12)$$

where $\mathbf{K}^T$ is the transposed of $\mathbf{K}$, and, for any vector $\mathbf{y} \neq 0$,

$$\mathbf{y} \cdot \mathbf{K} \cdot \mathbf{y} > 0. \qquad (2.13)$$

The flux $q_{i,\tau}$ is related to ageing, i.e. the process by which the age of every particle tends to increase by a certain amount of time as time progresses by the same amount of time. This may be viewed as advection with a unit velocity in the age direction. Hence,

$$q_{i,\tau} = c_i. \qquad (2.14)$$

Substituting relations (2.11) and (2.14) into Eq. (2.10) leads to the *equation governing the evolution of the concentration distribution function*:

$$\boxed{\frac{\partial c_i}{\partial t} = p_i - d_i - \nabla \cdot (\mathbf{u} c_i - \mathbf{K} \cdot \nabla c_i) - \frac{\partial c_i}{\partial \tau}}$$

$$(2.15)$$

The latter is the core of the present theory of the age, meaning that all the relevant equations may be derived from it.

2.2. Concentration equation

By integrating Eq. (2.15) over τ, taking into account relation (2.5) and the common-sense boundary condition

$$\lim_{\tau \to \infty} c_i(t,\mathbf{x},\tau) = 0, \qquad (2.16)$$

the equation obeyed by the i-th constituent concentration is obtained, i.e.

$$\frac{\partial C_i}{\partial t} = P_i - D_i - \nabla \cdot (\mathbf{u} C_i - \mathbf{K} \cdot \nabla C_i), \qquad (2.17)$$

with

$$P_i(t,\mathbf{x}) = c_i(t,\mathbf{x},\tau = 0) + \int_0^{\infty} p_i(t,\mathbf{x},\tau)\mathrm{d}\tau, \qquad (2.18)$$

and

$$D_i(t,\mathbf{x}) = \int_0^{\infty} d_i(t,\mathbf{x},\tau)\mathrm{d}\tau. \qquad (2.19)$$

Obviously, $c_i(t,\mathbf{x},\tau = 0)$ is zero, unless there is a source producing i-th constituent with its age prescribed to be zero at time t and location $\mathbf{x}$. In the latter case, however, $c_i(t,\mathbf{x},\tau = 0)$ is directly related to the production rate, implying that it should not be regarded as an additional unknown. On the other hand, a source at $\tau = 0$ may also be accounted for by including an appropriate Dirac function in p_i. Then, whether or not there exists a source producing i-th constituent with zero age, $c_i(t,\mathbf{x},\tau = 0)$ may be considered to be zero, but must be understood as the value at $\tau = 0^-$. The latter approach is adopted herein.

Eq. (2.17) is far from novel. However, it is worth remarking that this classical relation can be derived from the equation governing the evolution of the concentration distribution function.

2.3. Age-averaging hypothesis

In most applications, solving the equation governing the concentration distribution function is unlikely to be easy, since the latter depends on five independent variables, i.e. the time, the three space coordinates and the age itself. If no information is required about the distribution of the age in every fluid parcel, then it may be sufficient to determine the mean age of every constituent. However, before establishing the equation allowing to do so, it is necessary to specify how the mean age is related to the concentration distribution function.

Once the concentration distribution function and the concentration are defined, there seems to exist a logical and unambiguous path leading to expression (2.5), the relation between the aforementioned variables. However, it must be realised that this reasoning calls on, for writing Eqs. (2.1) and (2.2), the basic physical principle which specifies that mass is an additive quantity, i.e. the total mass of a set of particles is the sum of the masses of the particles considered. There is no such physical principle about the age, implying that the mean age may be derived in many conceivable ways from the concentration distribution function. This is why a suitable assumption is to be introduced, which will be called the "age-averaging hypothesis."

It is decided to retain the age-averaging hypothesis which reads:

The mean age of a set of particles is defined as the mass-weighted, arithmetic average of the ages of the particles considered; this is valid for particles of the same constituents or for particles of different constituents.

To illustrate the meaning of the assumption above, it is appropriate to tackle a simple problem. Consider two particles, identified by superscript "A" and "B," the masses and ages of which are m^A, m^B, and τ^A, τ^B, respectively. According to the age-averaging hypothesis, the mean age τ^{A+B} of the system made up of particle A and particle B is

$$\tau^{A+B} = \frac{m^A \tau^A + m^B \tau^B}{m^A + m^B}. \qquad (2.20)$$

Thus, the age is an intensive variable, i.e. the age of a set of two particles is not the sum of the age of the particles considered. By contrast, mass is an extensive—or additive—quantity; the mass m^{A+B} of the system, consisting of particles A and B, is the sum of the masses of the particles of which the system is made up:

$$m^{A+B} = m^A + m^B. \qquad (2.21)$$

Combining Eqs. (2.20) and (2.21) shows that there is an additive quantity associated with the age,

$$m^{A+B}\tau^{A+B} = m^A\tau^A + m^B\tau^B, \qquad (2.22)$$

which might be called the *age content*—an appellation inspired from the fact that mass is a measure of the matter content of the system under study.

To deal with a system consisting of more than two particles, straightforward extensions of formulas (2.20) to (2.22) may be worked out. Furthermore, translating these Lagrangian expressions into Eulerians ones is readily achieved. In particular, the mean age $a_i(t,\mathbf{x})$ of the i-th constituent at time t and location $\mathbf{x}$ is given by

$$a_i(t,\mathbf{x}) = \frac{\int_0^\infty \tau c_i(t,\mathbf{x},\tau)\,\mathrm{d}\tau}{\int_0^\infty c_i(t,\mathbf{x},\tau)\,\mathrm{d}\tau}, \qquad (2.23)$$

an expression which is obviously consistent with the age-averaging hypothesis.

Let the *age concentration* of the i-th constituent be defined as

$$\alpha_i(t,\mathbf{x}) = \int_0^\infty \tau c_i(t,\mathbf{x},\tau)\,\mathrm{d}\tau, \qquad (2.24)$$

then, it is readily seen that the mean age is the ratio of the age concentration to the concentration, i.e.

$$a_i(t,\mathbf{x}) = \frac{\alpha_i(t,\mathbf{x})}{C_i(t,\mathbf{x})}. \qquad (2.25)$$

On the other hand, the age concentration is closely associated with the concept of age content. Consider, for instance, the fluid parcel of volume ΔV that at time t has its center at location $\mathbf{x}$. Then, adapting relation (2.22) to the present case, the age content $A_i(t,\mathbf{x},\Delta V)$ of the i-th constituent of the fluid parcel satisfies:

$$A_i(t,\mathbf{x},\Delta V) \sim \rho\Delta V\alpha_i(t,\mathbf{x}), \qquad \Delta V \to 0. \qquad (2.26)$$

Hence, the age content being an additive quantity, the total age content of the fluid parcel under study is

$$A(t,\mathbf{x},\Delta V) \sim \sum_{i=0}^I A_i(t,\mathbf{x},\Delta V), \qquad \Delta V \to 0. \qquad (2.27)$$

Comparing relations (2.26)–(2.27) with relations (2.1)–(2.2), it appears that the role played by the age concentration in the calculation of the age content is similar to that of the concentration in the measure of the matter content—i.e. the calculation of the mass. Therefore, it is no surprise that the age concentration will be seen to obey a partial differential equation in flux form, bearing similarities with that governing the evolution of the concentration.

2.4. Age concentration and age equations

It seems reasonable to assume that the concentration distribution function satisfy the following end conditions:

$$\lim_{\tau \to 0} \tau c_i(t,\mathbf{x},\tau) = 0 \qquad (2.28)$$

and

$$\lim_{\tau \to \infty} \tau c_i(t,\mathbf{x},\tau) = 0. \qquad (2.29)$$

It must be pointed out that relations (2.16) and (2.28) are needed for ensuring the existence of integral (2.5), while integral (2.24) would not exist if relation (2.29) is not satisfied.

Multiplying Eq. (2.15) by τ, integrating over τ, taking into account the above boundary conditions, it follows that the age concentration obeys

$$\frac{\partial \alpha_i}{\partial t} = C_i + \pi_i - \delta_i - \nabla \cdot (\mathbf{u}\,\alpha_i - \mathbf{K} \cdot \nabla\alpha_i), \qquad (2.30)$$

with

$$\pi_i(t,\mathbf{x}) = \int_0^\infty \tau p_i(t,\mathbf{x},\tau)\,\mathrm{d}\tau, \qquad (2.31)$$

$$\delta_i(t,\mathbf{x}) = \int_0^\infty \tau d_i(t,\mathbf{x},\tau)\,\mathrm{d}\tau. \qquad (2.32)$$

As expected, the age concentration satisfies an equation similar to that governing the evolution of the concentration of every constituent, hence its name. As the independent variable τ, the age, will be used hardly ever from here on, it is decided, for simplicity, to call the variable a_i the "age" rather than the "mean age."

The equation governing every constituent concentration and its age concentration are derived from the same equation, i.e. the equation obeyed by the concentration distribution function. Therefore, the subgrid-scale parameterisations in the equations governing the constituent concentration and the age concentration, respectively, are consistent with each other. This is a key aspect of the present theory,

since it was previously underscored that the impact of mixing on the age must be taken into account properly.

It is now clear how the age is to be calculated in an *Eulerian numerical model* (Fig. 3):

1. *first, the concentration of every constituent is computed from Eq. (2.17);*
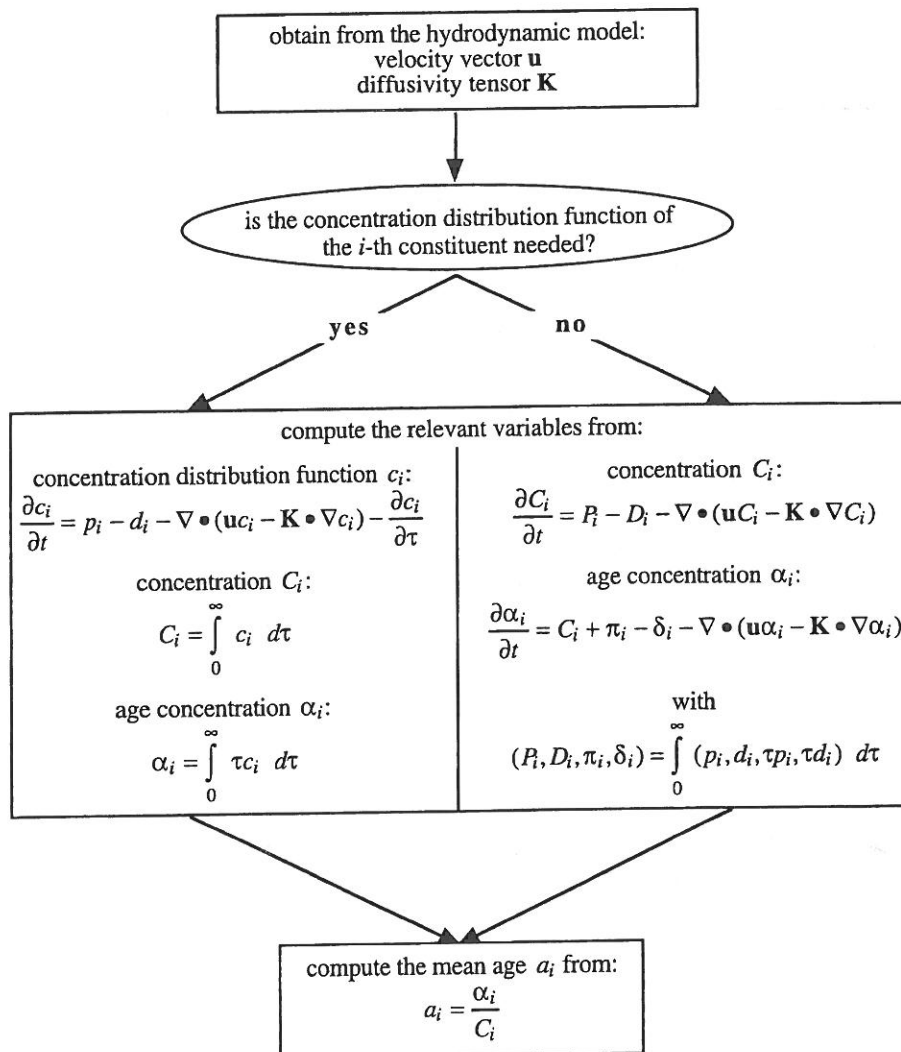
Fig. 3. Schematic illustration of the two approaches that may be adopted to estimate the mean age of every constituent in a numerical model. In the right-hand side route, with which the present study is mostly concerned, the concentration distribution function must not be calculated. This renders it less expensive in terms of CPU time, since four independent variables—i.e. time and three space coordinates—rather than five—i.e. time, three space coordinates and age—must be retained. However, ignoring the concentration distribution function implies that useful information on the flow is unavailable, as will be seen in other articles (e.g. Delhez and Deleersnijder, 2001).

2. *second, using a numerical scheme similar to that needed to obtain the constituent concentration, the age concentration is calculated from Eq. (2.30)—provided appropriate boundary conditions for the age concentration are set;*
3. *finally, according to relation (2.25), the age of every constituent is evaluated as the ratio of the age concentration to the constituent concentration.*

Thus, as the differential terms in the concentration and age concentration equations have a similar form, there is no need to develop new numerical schemes for the purpose of estimating the age. Instead, those implemented for computing the concentration can be used to obtain the age concentration. Of course, this approach is valid only if the age distribution can be ignored. Otherwise, Eq. (2.15) is to be solved first. Then, the concentration and the age may be obtained from relations (2.5) and (2.23), respectively (Fig. 3).

Since the production/destruction terms are unlikely to have a similar form in the concentration and age concentration equations, it may be objected that significant additional numerical efforts may be required for solving the age concentration equation. This argument is probably irrelevant because, as is seen below, the source/sink terms generally include no derivatives, making them rather easy to handle in numerical models. In other words, the non-differential terms may safely be ignored when assessing the problems associated with the numerical solution of the equations considered herein.

That the age may be derived from an algorithm similar to that included in the associated marine model is a guarantee of its relevance as a tool for understanding the functioning of the model. This advantage would be lost if the age were to be evaluated in a Lagrangian manner or directly from from the age equation,

$$\frac{\partial a_i}{\partial t} = 1 + \frac{\pi_i - a_i P_i}{C_i} - \frac{\delta_i - a_i D_i}{C_i}$$
$$- \nabla \cdot (\mathbf{u} a_i - \mathbf{K} \cdot \nabla a_i) + \frac{2 \nabla a_i \cdot \mathbf{K} \cdot \nabla C_i}{C_i},$$

$$(2.33)$$

which can be obtained by manipulating Eqs. (2.17), (2.25) and (2.30), and taking into account the fact that—according to the Boussinesq approximation—the velocity is divergence-free. The last term in the right-hand side of the above equation is not in flux form and has no counterpart in the equation obeyed by the concentration, implying that a specific numerical scheme should be devised to deal with it. However, Eq. (2.33) may be useful in certain analytical studies or, as will be seen below, for understanding the impact of production/destruction terms on the age.

3. Further theoretical developments

It is now appropriate to examine a few direct consequences of the general theory of the age established in Section 2.

3.1. Aggregates

The number of the constituents of seawater is so large that it is unreasonable to develop a strategy that would demand that the concentration and, possibly, the age of each of them is computed separately. It is more appropriate to deal with suitably defined groups of constituents. Herein, such a group is called an "aggregate" (e.g. Nihoul, 1993). So, seawater may be viewed as made up of a certain number of aggregates, the concentrations C_k^{ag} and age concentrations α_k^{ag} of which are defined as

$$\left(C_k^{ag}, \alpha_k^{ag} \right) = \sum_{j=0}^{I} \omega_{k,j} \left(C_j, \alpha_j \right), \qquad (3.1)$$

where $\omega_{k,j}$ is equal to 1 or 0, according to whether the j-th constituent is included in or excluded from the k-th aggregate. Applying to the Eqs. (2.17) and (2.30) a weighted sum similar to that used in definition (3.1), it is readily seen that the equations obeyed by the concentration and the age concentration of an aggregate are similar to those governing the evolution of the concentration and age concentration of any individual constituent. Finally, by virtue of the

age-averaging hypothesis, the age of the k-th aggregate must be evaluated as

$$a_k^{\mathrm{ag}} = \frac{\alpha_k^{\mathrm{ag}}}{C_k^{\mathrm{ag}}} = \frac{\sum\limits_{j=0}^{I} \omega_{k,j}\alpha_j}{\sum\limits_{j=0}^{I} \omega_{k,j}C_j}. \quad (3.2)$$

Thus, as the mathematical relations pertaining to aggregates are equivalent to those derived for the individual constituents, there is no need to keep distinguishing aggregates from individual constituents. Accordingly, for simplicity, only the term "constituent" will be used from here on, even if the constituent under consideration is in fact an aggregate.

3.2. Age of seawater

Seawater is the aggregate that comprises all constituents. By virtue of relation (2.4), its concentration is equal to unity:

$$C^{\mathrm{sw}} = 1. \quad (3.3)$$

In general, there exists not net local source or sink of mass, implying that the production and destruction rates must satisfy the constraint

$$\sum_{i=0}^{I} (P_i - D_i) = 0. \quad (3.4)$$

Therefore, the equation governing the concentration of seawater, which is obtained by taking the sum over all constituents of the concentration Eq. (2.17), reduces to

$$\nabla \cdot \mathbf{u} = 0, \quad (3.5)$$

the continuity equation to be used when the Boussinesq approximation is assumed to hold valid.

Since the major constituent of seawater is pure water, the concentration of the latter is close to unity. Thus, the age concentration of pure water, $\alpha_0 = C_0 a_0$, is almost equal to its age:

$$\alpha_0 \sim a_0, \quad C_0 \rightarrow 1. \quad (3.6)$$

Therefore, as pure water is a passive constituent—i.e. a constituent that is neither produced nor destroyed,

implying that $P_0 = 0 = D_0$—the equation governing the evolution of the age concentration of pure water reduces asymptotically to

$$\frac{\partial a_0}{\partial t} \sim 1 - \nabla \cdot (\mathbf{u}\,a_0 - \mathbf{K} \cdot \nabla a_0), \quad C_0 \rightarrow 1. \quad (3.7)$$

On the other hand, unless the age of a constituent other than pure water is found to be so large, due to particular initial or boundary conditions, that its age concentration is of the same order of magnitude as that of pure water, the age concentration of seawater, α^{sw}, tends to be equivalent to that of pure water, i.e.

$$\alpha^{\mathrm{sw}} \sim \alpha_0, \quad C_0 \rightarrow 1. \quad (3.8)$$

Hence, as $C^{\mathrm{sw}} = 1$, the age of seawater is close to that of pure water,

$$a^{\mathrm{sw}} = \frac{\alpha^{\mathrm{sw}}}{C^{\mathrm{sw}}} \sim a^0, \quad C_0 \rightarrow 1. \quad (3.9)$$

As a consequence, the age of seawater may be obtained from an equation similar to that governing the age of pure water:

$$\frac{\partial a^{\mathrm{sw}}}{\partial t} \sim 1 - \nabla \cdot (\mathbf{u}\,a^{\mathrm{sw}} - \mathbf{K} \cdot \nabla a^{\mathrm{sw}}), \quad C_0 \rightarrow 1. \quad (3.10)$$

It is noteworthy that several authors (e.g. Thiele and Sarmiento, 1990; Haidvogel and Bryan, 1992; England, 1995; Jia and Richards, 1996; England and Rahmstorf, 1999; Campin et al., 1999) evaluated the steady-state solution of the latter equation in order to estimate ventilation timescales in oceanic basins, showing that the general theory developed herein is consistent with certain previous studies involving the age.

Finally, if there is a net local source or sink of matter, the developments above remain relevant, provided the associated net production or destruction rate is sufficiently small that constraint (3.5) holds approximately valid.

3.3. Production / destruction terms

The production/destruction terms included in the age concentration and concentration equations, being

formulated as integrals over the independent age variable τ, may appear as unnecessarily complex. It is believed, however, that, in most cases, these integrals can be easily computed, leading to simple expressions.

The source/sink terms appearing in the equation governing the concentration distribution function of a passive constituent—i.e. a constituent that is neither produced nor destroyed—are obviously zero:

$$p_p = 0 = d_p. \qquad (3.11)$$

Hence, the corresponding production and destruction rates of concentration and age concentration are also zero:

$$P_p = 0 = D_p \qquad (3.12)$$

and

$$\pi_p = 0 = \delta_p. \qquad (3.13)$$

Most destruction phenomena develop irrespective of the age of the particles being destroyed, i.e. the probability for a particle of a given constituent to be transformed into a particle of another constituent does not depend of its age. If this is the case, the sink terms in the equations governing the age distribution function and the age concentration are:

$$(d_i, \delta_i) = \left(\frac{c_i}{C_i}, a_i \right) D_i. \qquad (3.14)$$

As a consequence, the term related to destruction appearing in the age equation, Eq. (2.33), is identically zero,

$$\frac{\delta_i - a_i D_i}{C_i} = 0, \qquad (3.15)$$

indicating that this type of destruction does not tend to modify the age of the constituent under study. For instance, the sink terms to be included in the evolution equations relevant to a radioactive isotope are

$$\left[d_r, D_r, \pi_r (\delta_r - a_r D_r)/C_r \right] = \frac{1}{T}(c_r, C_r, \alpha_r, 0), \qquad (3.16)$$

where $T \log 2$ denotes the half-life.

The distribution of the age of the particles of a given constituent being produced is unlikely to be similar to that of the particles of this constituent already present in the fluid, implying that parameterisations similar to Eq. (3.14) are unlikely to hold valid for source terms. At time t and location $\mathbf{x}$, the mean age of the particles of the i-th constituent being produced is given by

$$\mu_i(t,\mathbf{x}) = \frac{\int_0^\infty \tau p_i(t,\mathbf{x},\tau)\,\mathrm{d}\tau}{\int_0^\infty p_i(t,\mathbf{x},\tau)\,\mathrm{d}\tau} = \frac{\pi_i(t,\mathbf{x})}{P_i(t,\mathbf{x})}. \qquad (3.17)$$

Therefore, the term associated with production processes included in the age equation, Eq. (2.33), reads

$$\frac{\pi_i - a_i P_i}{C_i} = -\frac{P_i}{C_i}(a_i - \mu_i), \qquad (3.18)$$

so that the mean age a_i is nudged toward the mean age of the particles being produced, μ_i.

An illustrative example is that of a constituent which is released by a source into the water with a prescribed, unique age $\mu(t,\mathbf{x})$. Let "pa" and $Q_{pa}(t,\mathbf{x})$ be the subscript identifying this constituent and the mass rate of release of the source per unit of volume. The production term to be included in the equation satisfied by the age distribution function is

$$p_{pa} = \frac{Q_{pa}}{\rho}\delta(\mu - \tau), \qquad (3.19)$$

where δ is the Dirac function. Then,

$$(P_{pa}, \pi_{pa}) = \frac{Q_{pa}}{\rho}(1, \mu). \qquad (3.20)$$

Therefore, the second term in the age equation, Eq. (2.33), is

$$\frac{\pi_{pa} - a_{pa} P_{pa}}{C_{pa}} = -\frac{Q_{pa}}{\rho C_{pa}}(a_{pa} - \mu), \qquad (3.21)$$

an expression similar to relation (3.18). Hence, the source results in the age being nudged toward the age of the particles released into the flow, a process of which the importance grows as the rate of release of the source increases.

As the production/destruction rates are likely to be relatively simple expressions, applying the present theory of the age to active constituents is not just a mere speculation that would hardly ever become

reality because of insuperable difficulties in the formulation of the production/destruction rates. This does not imply, however, that parameterisations, which explicitly retain the age dimension, should never be utilised. It might be appropriate, for example, to set up ecological models in which the behaviour of living organisms would depend explicitly on their age, demanding to work in the four-dimensional space comprising the physical space and the age dimension. One of the improvements that such an approach could bring about would be the offering the opportunity of formulating the death rate as a function of the age. Nonetheless, investigating this is far beyond the scope of the present article.

3.4. Mass and age budgets

As was pointed out above, mass and age content are additive quantities. Thus, it is possible to evaluate the corresponding budgets over a given control volume. The latter is assumed to be motionless, denoted as V, and is limited by surface S, of which the outward unit normal reads $\mathbf{n}$. For the i-th constituent, the matter content, i.e. the mass, and the age content of the control volume are

$$m_i(t) = \rho \int_V C_i(t, \mathbf{x}) \, dV \tag{3.22}$$

and

$$A_i(t) = \rho \int_V \alpha_i(t, \mathbf{x}) \, dV, \tag{3.23}$$

respectively. By integrating the concentration equation, Eq. (2.17), and the age concentration equation, Eq. (2.30), over V, and using the divergence theorem, the temporal rates of variation of $m_i(t, V)$ and $A_i(t, V)$ are seen to be given by

$$\frac{1}{\rho} \frac{d}{dt} m_i(t)$$

$$= \underbrace{\int_V P_i \, dV}_{\text{production}} - \underbrace{\int_V D_i \, dV}_{\text{destruction}} - \underbrace{\int_S (\mathbf{u} C_i) \cdot \mathbf{n} \, dS}_{\text{advective transport through } S}$$

$$- \underbrace{\int_S (-\mathbf{K} \cdot \nabla C_i) \cdot \mathbf{n} \, dS}_{\text{diffusive transport through } S}, \tag{3.24}$$

and

$$\frac{1}{\rho} \frac{d}{dt} A_i(t)$$

$$= \underbrace{\int_V C_i \, dV}_{\text{ageing}} + \underbrace{\int_V \pi_i \, dV}_{\text{production}} - \underbrace{\int_V \delta_i \, dV}_{\text{destruction}}$$

$$- \underbrace{\int_S (\mathbf{u} \alpha_i) \cdot \mathbf{n} \, dS}_{\text{advective transport through } S} - \underbrace{\int_S (-\mathbf{K} \cdot \nabla \alpha_i) \cdot \mathbf{n} \, dS}_{\text{diffusive transport through } S}, \tag{3.25}$$

respectively. Obviously, the matter and age budgets depend on similar processes, with the exception of ageing. Nonetheless, the latter term may conceivably be viewed as a special type of production, leading then to a complete similarity of the two relations above. Finite-volume or conservative finite-difference schemes for solving numerically the concentration and age concentration equations, may be derived from the integral relations above (e.g. Peyret and Taylor, 1983; Hirsch, 1988).

The diffusive flux of age concentration, $-\mathbf{K} \cdot \nabla \alpha_i$, may be split into two parts,

$$-\mathbf{K} \cdot \nabla \alpha_i = a_i(-\mathbf{K} \cdot \nabla C_i) + C_i(-\mathbf{K} \cdot \nabla a_i). \tag{3.26}$$

Therefore, the diffusive transport across S is due to the spatial variations of both the constituent concentration and the age. So, for instance, if the age is homogeneous ($\nabla a_i = 0$), a mass flux of the i-th constituent may still cause the age content of V to vary. Conversely, if the concentration of the i-th constituent is homogeneous ($\nabla C_i = 0$), there is no mass flux of this constituent through S, but diffusive transport may still transport younger or older constituent particles across S, which may induce variations of the age content of V.

3.5. No-diffusion limit

Let D_t denote the material derivative operator,

$$D_t = \frac{\partial}{\partial t} + \mathbf{u} \cdot \nabla, \tag{3.27}$$

which represents the temporal rate of change evaluated by an observer "tied" to a fluid parcel, i.e. an

observer whose velocity is equal to that of the fluid. If diffusive phenomena are negligible, the diffusivity tensor may be assumed to be zero. In this case, at every time t and location $\mathbf{x}$ in the domain of interest, the ages of a passive tracer, $a_p(t, \mathbf{x})$, of a radioactive constituent, $a_r(t, \mathbf{x})$, and of seawater, $a^{sw}(t, \mathbf{x})$, all obey similar equations

$$D_t\left[a_p(t,\mathbf{x}), a_r(t,\mathbf{x}), a^{sw}(t,\mathbf{x}) \right] = (1,1,1). \quad (3.28)$$

In agreement with considerations presented in Section 1, this suggests that a unique age, $a(t,\mathbf{x})$, be ascribed to every fluid parcel, which would obey

$$D_t a(t,\mathbf{x}) = 1, \quad (3.29)$$

an equation which is the Eulerian counterpart of Lagrangian equation, Eq. (1.2).

Clearly, diffusion is one of the causes of the differences between ages. This is why the impact of diffusion on the age of various types of constituents must be assessed. This issue is addressed with the help of sophisticated numerical results in Sections 4 and 5. However, before tackling these complex prob-lems, it is appropriate to carry out a simple—per-haps even simplistic—illustrative thought experi-ment to illustrate the impact of mixing on the ages of various constituents.

3.6. A thought experiment on the impact of mixing

Radioisotope dating, such as the carbon-14 tech-nique, is a widely-used method for estimating the time elapsed since the death of a living system. Its basic principle is readily outlined. As long as the system under consideration is alive, it exchanges matter with its environment. Further assume that a stable—or passive—isotope and a radioactive iso-tope of the same element are part of the matter fluxes between the system and its environment. If, in the latter, the ratio ξ of the concentration of the radioactive isotope to that of the stable one is con-stant, then it is conceivable that, at any instant t of the lifetime of the system, the ratio of the mass $m_r(t)$ of the radioactive isotope contained in the system to the mass $m_p(t)$ of the stable isotope is approximately equal to the constant ξ (Fig. 4).

living system :
exchanges of matter
with the environment

open system

dead system :
no exchange of matter
with the environment

closed system

$t = \theta$
time of death

time : t

$t < \theta: \quad \dfrac{m_r(t)}{m_p(t)} = \xi = const.$

$\theta < t: \begin{cases} m_p(t) = M_p = const. \\ m_r(t) = M_r \exp\dfrac{-(t - \theta)}{T} \neq const. \end{cases}$

$\Rightarrow$ radio $-$ age: $\tilde{a}(t) = T \log\dfrac{m_p(t)/M_p}{m_r(t)/M_r}$
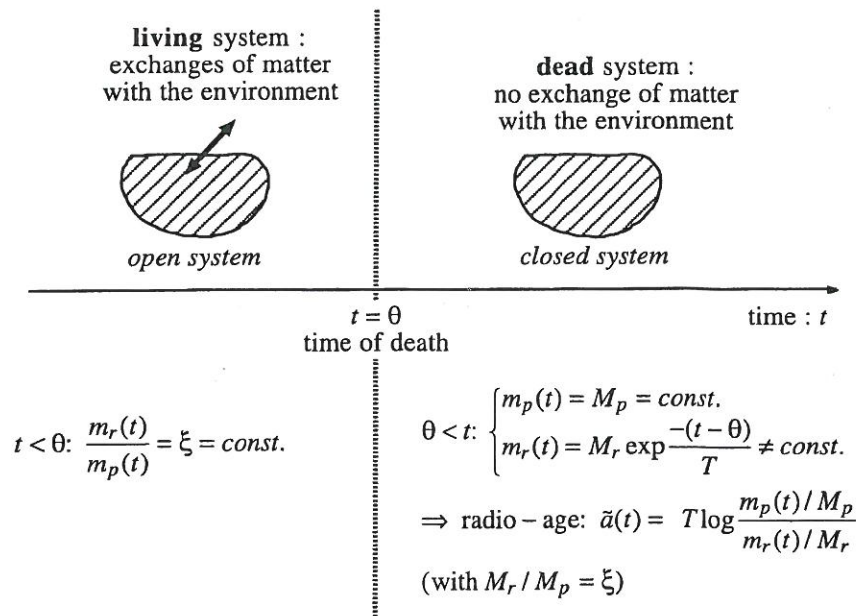
(with $M_r / M_p = \xi$)

Fig. 4. Illustration of the basic principle of the radioisotope dating technique, where $m_p(t)$ and $m_r(t)$ represent the mass of a passive/stable isotope and the mass of a radioactive isotope, respectively, contained at time t in the system under study. It is assumed that the ratio ξ of the concentrations of these two isotopes is constant in the environment of the living system. In practice, the stable and radioactive isotopes considered are often carbon-12 and carbon-14, respectively.

Table 1

Values of the stable and radioactive isotope contents of systems A, B and A + B, along with the relevant ages, as functions of time t

The dimensionless parameter ν is defined in Eq. (3.39), while T represents the timescale characterising the decay of the radioactive constituent. Prior to the mixing event, which occurs at $t = \theta^{A+B}$, all ages are equal in system A and system B, whereas the ages are different after the mixing event.

	system A	system B	system $A+B$
relevant time interval	$\theta^A \leq t < \theta^{A+B}$	$\theta^B \leq t < \theta^{A+B}$	$\theta^{A+B} < t$
passive/stable isotope mass	$m_p^A(t) = M_p$	$m_p^B(t) = M_p$	$m_p^{A+B}(t) = 2M_p$
radioactive isotope mass	$m_r^A(t) = M_r \exp\dfrac{-(t-\theta^A)}{T}$	$m_r^B(t) = M_r \exp\dfrac{-(t-\theta^B)}{T}$	$m_r^{A+B}(t) = M_r(1+\nu)\exp\dfrac{-(t-\theta^A)}{T}$
passive/stable isotope age	$a_p^A(t) = t - \theta^A$	$a_p^B(t) = t - \theta^B$	$a_p^{A+B}(t) = t - \theta^A + \dfrac{T}{2}\log\dfrac{1}{\nu}$
radioactive isotope age	$a_r^A(t) = t - \theta^A$	$a_r^B(t) = t - \theta^B$	$a_r^{A+B}(t) = t - \theta^A + \dfrac{\nu T}{1+\nu}\log\dfrac{1}{\nu}$
radio-age	$\tilde{a}^A(t) = t - \theta^A$	$\tilde{a}^B(t) = t - \theta^B$	$\tilde{a}^{A+B}(t) = t - \theta^A + T\log\dfrac{2}{1+\nu}$

(with $\theta^B < \theta^A < \theta^{A+B}$ and $0 < \nu < 1$)

Hypothesise that the system dies at instant $t = \theta$ and remains isolated from its environment since then. In other words, there is no matter flux between the system and its environment after its death. Let M_p and M_r denote the mass of the passive isotope and that of the radioactive one at $t = \theta$. Clearly,

$$M_r / M_p = \xi. \qquad (3.30)$$

Furthermore, at any instant subsequent to the death, i.e. at $t > \theta$, the following relations hold valid:

$$m_p(t) = M_p \qquad (3.31)$$

and

$$m_r(t) = M_r \exp\frac{-(t-\theta)}{T}, \qquad (3.32)$$

where $T\log 2$ denotes the half-life of the radioactive isotope. Thus, if the constant ξ is known, the time elapsed since the death of the system, $t - \theta$, may be estimated from measurements of the masses of the stable and the radioactive isotope present in the dead system from the well-known formula (Fig. 4)

$$t - \theta = T\log\left[\xi\frac{m_p(t)}{m_r(t)}\right], \qquad (3.33)$$

which is obtained by manipulating relations (3.30)–(3.32).

For the purposes of the present discussion, the time elapsed since the death of the system is regarded as the age of the system, while the timescale estimated according to relation (3.33) is termed the "radio-age," i.e. an estimate of the age of the system derived from measurements of its stable and radioactive isotopes content. Obviously, the radio-age (3.33), which is denoted $\tilde{a}(t)$ below, is consistent with relation (1.7). As long as the dead system remains isolated from its environment, it seems natural to assume that the age of the passive isotope, $a_p(t)$, that of the radioactive one, $a_r(t)$, and the radio-age, $\tilde{a}(t)$, are equal to each other, i.e.

$$a_p(t) = a_r(t) = \tilde{a}(t) = t - \theta. \qquad (3.34)$$

Now consider two systems, which are identified by superscripts "A" and "B." Their fate is somewhat similar to that of the single system studied above. They die at time θ^A and θ^B, respectively, which may be assumed to satisfy the inequality $\theta^B < \theta^A$ without any loss of generality. At the time of their
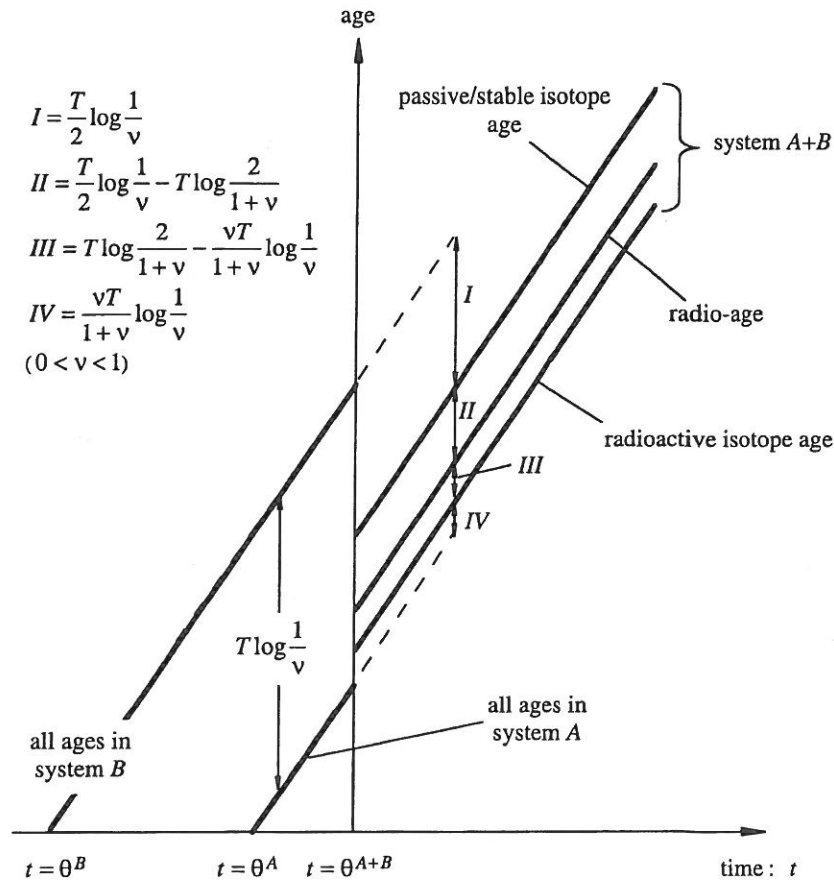
Fig. 5. Schematic representation of the ages in systems A, B and A + B as functions of time. At time $t = \theta^{A+B}$, system A and system B are mixed instantaneously, giving rise to system A + B. The half-life of the radioactive constituent is $T \log 2$. The dimensionless parameter ν is defined in Eq. (3.39). The discontinuity in the ages at time θ^{A+B} is caused by the mixing event taking place at this instant.

death, the stable and radioactive isotope contents of both systems are M_p and M_r. After their death, the systems remain isolated from their environment until time θ^{A+B}, the instant at which they are instantaneously and perfectly mixed, giving rise to a new system, identified by superscript "A + B," which does not exchange any matter with its environment from time θ^{A+B} on. In system A, from the time of its death until the instant at which it is mixed with system B, the age of the passive isotope, $a_p^A(t)$, that of the radioactive one, $a_r^A(t)$, and the radio-age, $\tilde{a}^A(t)$, may be assumed to be equal to each other (Table 1). Similar considerations hold valid for system B. In other words, before the mixing event, there is no ambiguity as to the definition of the age of a

constituent of every system, or its radio-age. After the mixing, it seems natural to consider that all ages increase as the same pace as time progresses. However, defining ages just after the mixing event is not straightforward, as several age definitions may be invoked.

The radio-age (3.33) may be adapted to system A + B as follows

$$a^{A+B}(t) = T \log \left[\xi \frac{m_p^{A+B}(t)}{m_r^{A+B}(t)} \right]. \tag{3.35}$$

On the other hand, the age of the passive constituent and that of the radioactive one may be estimated

with the help of the mass-weighted arithmetic means

$$\lim_{\varepsilon \to 0^+} \left[m_p^{A+B}(t) a_p^{A+B}(t) \right]_{t = \theta^{A+B} + \varepsilon}$$
$$= \lim_{\varepsilon \to 0^+} \left[m_p^A(t) a_p^A(t) + m_p^B(t) a_p^B(t) \right]_{t = \theta^{A+B} - \varepsilon}, \quad (3.36)$$

$$\lim_{\varepsilon \to 0^+} \left[m_r^{A+B}(t) a_r^{A+B}(t) \right]_{t = \theta^{A+B} + \varepsilon}$$
$$= \lim_{\varepsilon \to 0^+} \left[m_r^A(t) a_r^A(t) + m_r^B(t) a_r^B(t) \right]_{t = \theta^{A+B} - \varepsilon}, \quad (3.37)$$

i.e. expressions which ensue from the age-averaging hypothesis introduced in Section 2. As may be seen in Table 1, Figs. 5 and 6, the ages that may be defined in the system A + B are different from each other, and satisfy the inequalities (Appendix B)

$$a_r^{A+B}(t) < \tilde{a}^{A+B}(t) < a_p^{A+B}(t). \quad (3.38)$$

It must be underscored that the differences between the ages after the mixing event depend only on the timescale of radioactive decay T and the dimensionless parameter

$$\nu = \lim_{\varepsilon \to 0^+} \left[\frac{m_r^B(t)}{m_r^A(t)} \right]_{t = \theta^{A+B} - \varepsilon}, \quad (3.39)$$

which satisfies the inequality $0 < \nu < 1$ since $\theta^B < \theta^A$.
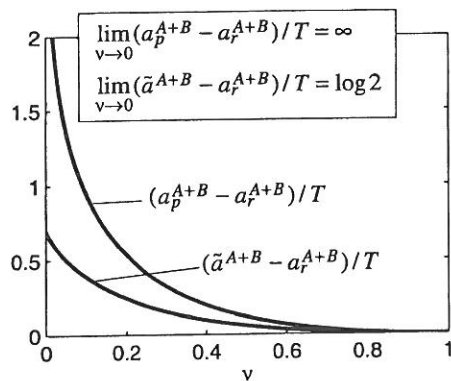
Fig. 6. Age differences in system A + B, scaled by the characteristic time of radioactive decay T, represented as functions of the dimensionless parameter ν. The latter is defined in Eq. (3.39). The expressions of the age differences are derived from equations included in Table 1, and are independent of time—at any instant $\theta^{A+B} < t$.

The above thought experiment illustrates how mixing may foul a radioactive dating process. It may be objected that this has little to do with age determination in marine applications. However, if systems A and B are interpreted as idealised fluid parcels, then this experiment may be regarded as a metaphor of the impact of mixing on the ages of various constituents of seawater, i.e. an idealised process in which diffusive phenomena take place abruptly, in a single mixing event, rather than progressively, as is the case in the seas. Nonetheless, this correctly shows that mixing is one of the causes of the differences between ages, and leads to inequalities (3.38), which will turn out to hold true in the much more realistic applications dealt with below.

4. Age in the English Channel and southern North Sea

Prandle (1984) studied the fate of the caesium-137 released into the Irish Sea by the Sellafield, UK, nuclear fuel reprocessing plant to investigate the long-term—i.e. over several years—circulation and mixing over the northwestern European shelf seas. Somewhat later, a similar approach was adopted by the European Union MAST-52 programme, which used radionuclides discharged into the English Channel by the nuclear fuel reprocessing plant of La Hague, France (Fig. 7), to "trace advection and dispersion of water masses in the 'European Coastal Current' from the English Channel to the Baltic" (Dahlgaard, 1995). Several numerical models were set up in the course of this programme, which simulated the transport of relevant tracers (Breton and Salomon, 1995; Nielsen, 1995; Salomon et al., 1995; Schönfeld, 1995). For a given radionuclide particle, a useful diagnostic variable was found to be the time that had elapsed since the particle under consideration was released into the English Channel (e.g. Salomon et al., 1995), a timescale which, in accordance with the definitions put forward herein, may be regarded as the age. Similar timescales of transport are estimated numerically in this Section, which include ages derived from the general theory of the age established in Sections 2 and 3 and a radio-age inspired by formula (1.7)—which is also considered since it is a popular way of estimating the age of a
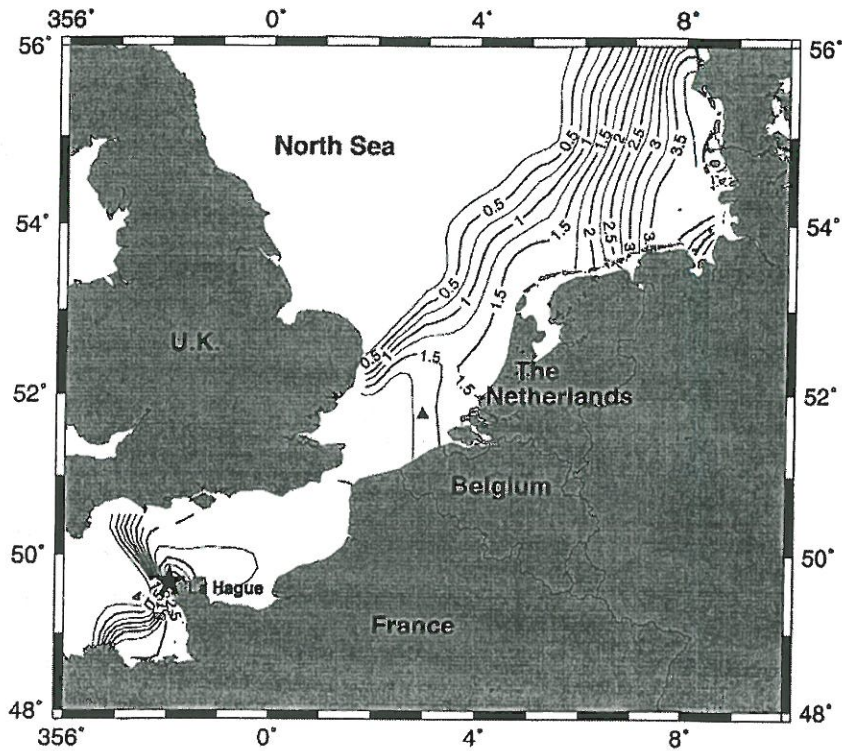
Fig. 7. Surface concentration of technetium-99 (in Bq m^{-3}) released by the La Hague nuclear fuel reprocessing plant, as simulated numerically on March 1, 1987. The contour interval is 0.25 Bq m^{-3}. The point identified by the symbol ▲ is that at which time series of various ages are displayed in Fig. 13.

tracer released by a point source (e.g. Anonymous, 1997; Boon et al., 1997; Beckers et al., 2001). Significant discrepancies between various age estimates are found. The reasons thereof are briefly investigated, highlighting the impact of diffusive processes, as well as that of the choice of boundary conditions.

The fate of the radionuclides released at La Hague is investigated here by means of the three-dimensional hydrodynamic model of the North Western European Continental Shelf, developed at the GHER (GeoHydrodynamics and Environment Research laboratory, University of Liège). This model is baroclinic, and includes a robust and versatile turbulence closure scheme (Delhez et al., 1999b). It covers the whole shelf to the east of the 200-m isobath, from 48° to 61°N, including the Skagerrak and Kattegat, with a horizontal resolution of 10′ in longitude and latitude, i.e. about 10 × 16 km, and 10 vertical

sigma-levels (Delhez and Martin, 1992). The numerical implementation is based on a finite-volume approach and uses a TVD scheme with superbee limiter for the advection of scalar properties (James, 1996). For the current study, the model is forced at its open ocean boundaries, which are located far away from the region of interest, by nine tidal constituents and the inverse barometric effect. The meteorological forcing data (six-hourly air temperature, surface pressure, relative humidity, wind speed, cloud cover; horizontal resolution 2.5 × 2.5°) are the NCEP/NCAR reanalysis of surface data from NOAA/CDC (http://www.cdc.noaa.gov/cdc/re-analysis). The simulations span a period of over 6 years starting on the first of January 1983 from adjusted climatological mean winter fields. A slightly different version of this model produced excellent results in a recent European advection/dispersion model inter-comparison exercise (Anonymous, 1997).
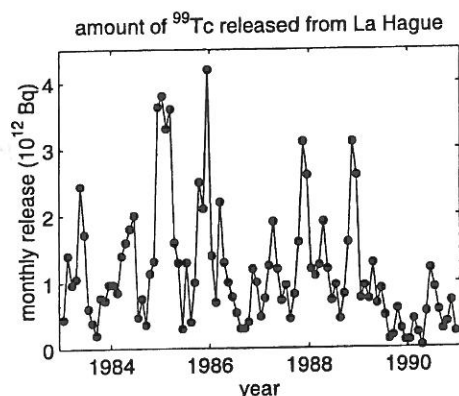
Fig. 8. Amount of technetium-99 released every month (in 10^{12} Bq) into the English Channel by the nuclear fuel reprocessing plant of La Hague. These figures, which span the period 1983–1990, were provided by the COGEMA, the company running the La Hague installations.

Therefore, it is believed that the GHER numerical model is capable of providing a rather realistic de-

scription of the large-scale hydrodynamic processes occurring in the region of interest that is well suited to the needs of the present study.

Technetium-99 is one of the tracers released into the English Channel at La Hague to which much attention was devoted in the above-mentioned MAST-52 programme (e.g. Dahlgaard et al., 1995). This element is radioactive, which is why its concentration is often expressed in Becquerel per cubic metre ($Bq\ m^{-3}$). However, its half-life, 214×10^3 years (Krane, 1988), is so much longer than the characteristic timescales of transport in the domain of interest, that it may safely be considered as passive in the present study. Under this assumption, using the release rate from the La Hague plant depicted in Fig. 8, the concentration of technetium-99 is simulated by means of the GHER model for the period from January 1983 until September 1989, starting from zero concentration (Fig. 7). In the region where the concentration of technetium-99 is significant, the vertical mixing is sufficiently intense
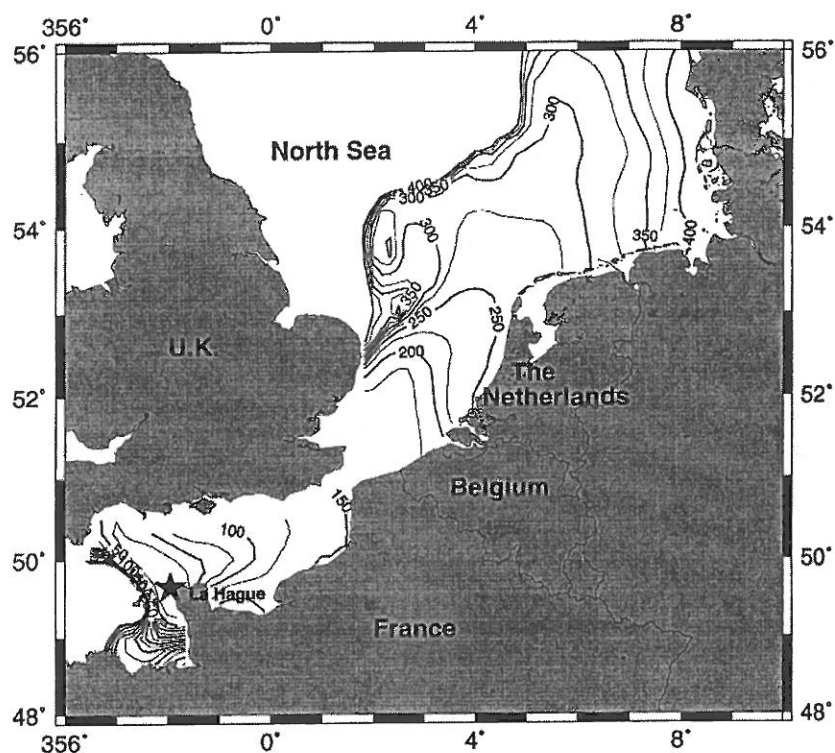

Fig. 9. Surface value of the age of the technetium-99 (in days) released by the La Hague nuclear fuel reprocessing plant, as simulated numerically on March 1, 1987. The contour interval is 25 days.
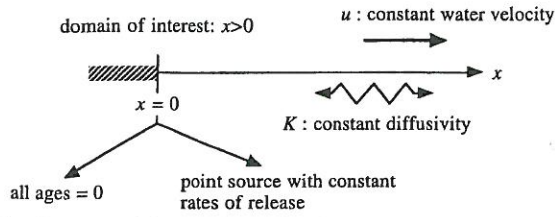
Fig. 10. Setup of the one-dimensional model for representing, in a highly idealised way, the transport of tracers released into the English Channel by the La Hague nuclear fuel reprocessing plant. The relevant equations and their solutions are collected in Table 2.

(e.g. Simpson et al., 1978; Otto et al,. 1990; Martin and Delhez, 1994) that its concentration is almost

homogeneous over the water column. Of course, the concentrations of other tracers, as well as their ages, also exhibit a near vertical homogeneity in the English Channel and southern North Sea. As a result, no crucial information is lost by displaying only surface values of concentrations or ages.

The age of technetium-99, as depicted in Fig. 9, is computed by assuming that the age of the particles of this tracer are zero at the instant they leave the outfall pipe. The technetium-99 particles present in the vicinity of La Hague are a mixture of particles that have just left the source, of which the age tends to be zero, and particles that have travelled the

Table 2

The one-dimensional, steady state model for representing, in a highly idealised way, the transport of tracers released into the English Channel by the La Hague nuclear fuel reprocessing plant

Coordinate x represents the distance to the source point. The dimensionless number λ is equal to $K/(u^2 T)$, where the positive constants K, u and T denote the diffusivity, the water velocity and the timescale of decay of the radioactive tracer, respectively. The solutions of the differential equations, which are displayed in the right-hand side column, are only valid downstream of the source, i.e. in the region where $0 \le x < \infty$ water.

water

$$C_0 \approx 1$$

$$u\frac{da_0}{dx} = 1 + K\frac{d^2 a_0}{dx^2} \qquad\qquad a_0(x) = \frac{x}{u}$$

passive tracer

$$u\frac{dC_p}{dx} = K\frac{d^2 C_p}{dx^2} \qquad\qquad C_p(x) = C_p(0)$$

$$u\frac{da_p}{dx} = 1 + K\frac{d^2 a_p}{dx^2} + \frac{2K}{C_p}\frac{dC_p}{dx}\frac{da_p}{dx} \qquad\qquad a_p(x) = \frac{x}{u}$$

radioactive tracer

$$u\frac{dC_r}{dx} = -\frac{C_r}{T} + K\frac{d^2 C_r}{dx^2} \qquad\qquad C_r(x) = C_r(0)\,\exp\frac{-u[(1+4\lambda)^{1/2}-1]x}{2K}$$

$$u\frac{da_r}{dx} = 1 + K\frac{d^2 a_r}{dx^2} + \frac{2K}{C_r}\frac{dC_r}{dx}\frac{da_r}{dx} \qquad\qquad a_r(x) = \frac{x}{u}\frac{1}{(1+4\lambda)^{1/2}}$$

radio-age

$$\tilde{a}(x) = T\,\log\frac{C_r(0)/C_p(0)}{C_r(x)/C_p(x)} \qquad\qquad \tilde{a}(x) = \frac{x}{u}\frac{(1+4\lambda)^{1/2}-1}{2\lambda}$$

domain of interest as a result of advective and diffusive transport since they were released by the reprocessing plant, implying that their age is greater than zero. Therefore, the mean age in the grid box in which technetium-99 is released, being the average of the age of all tracer particles present in this control volume, is likely to be larger than zero. For diagnostic purposes, it may be equally relevant to prescribe that the age be zero in this grid box. In this case, the age represents the time elapsed since leaving the grid box where the technetium source is located. Obviously, this alternative approach has no impact on the tracer concentration, but requires a slightly different solution procedure of the age concentration equation. However, leaving out short periods of time just after sharp reductions of the release rate of technetium, both ages are very close to each other, in accordance with the analytical results obtained by Deleersnijder et al. (2001).

The residual current is known to flow northeastward through the English Channel and along the Dutch and German coast (e.g. Lee, 1980; Otto et al., 1990; Delhez, 1996), which is reflected in the concentration and age fields of technetium-99 depicted in Figs. 7 and 9. All this is consistent with the concept of the "European Coastal Current" referred to in Dahlgaard (1995), strongly suggesting that the transport processes under study may be idealised as a one-dimensional advection–diffusion problem. Such a problem is handled under the additional simplifying hypothesis that a steady state prevails. A passive constituent and a radioactive one are discharged at a constant rate into a one-dimensional flow, where the velocity u and the eddy diffusivity K are constant. The distance to the source is measured by coordinate x, which is positive downstream of the source (Fig. 10). The age of the water, that of the passive constituent, and that of the radioactive tracer are evaluated along with a radio-age inspired by formula (1.7). All ages are prescribed to be zero at the source point, at $x = 0$. The relevant equations and their solutions in the domain $0 \leq x < \infty$ are displayed in Table 2. Upstream of the source, i.e. in the region where $-\infty < x < 0$, the tracer concentrations are not zero, because of diffusive processes. The associated ages are symmetric with respect to the source point. Investigating this counterintuitive property is beyond the scope of the present article; this is dealt with in

Beckers et al. (2001), Deleersnijder et al. (2001), and Hall and Haine (2000).

As may be seen in Table 2, all ages—including the radio-age—exhibit a similar form

$$\text{age}(x) = \frac{x}{u} f(\lambda),   (4.1)$$

where the dimensionless number

$$\lambda = \frac{K}{u^2 T},   (4.2)$$

is a measure of the impact of diffusive processes. The function f of λ depends on the type of age being considered. In any case,

$$f(\lambda) \leq 1,   (4.3)$$

implying that the upper limit of all ages is x/u, i.e. the amount of time needed to travel the distance x to the source at speed u. As may be seen in Fig. 11, the no-diffusion limit of the function f is

$$\lim_{\lambda \to 0} f(\lambda) = 1.   (4.4)$$

Therefore, if diffusion tends to be negligible, all ages tend to a unique value, the "advective age" x/u. Accordingly, in the limit $\lambda \to 0$, the ages satisfy the asymptotic relations

$$(a_0, a_p, \tilde{a}, a_r) \sim \frac{x}{u}(1,1,1-\lambda,1-2\lambda),   \lambda \to 0.   (4.5)$$

It is worth pointing out that, downstream of the source ($0 < x < \infty$), the ages satisfy inequalities simi-
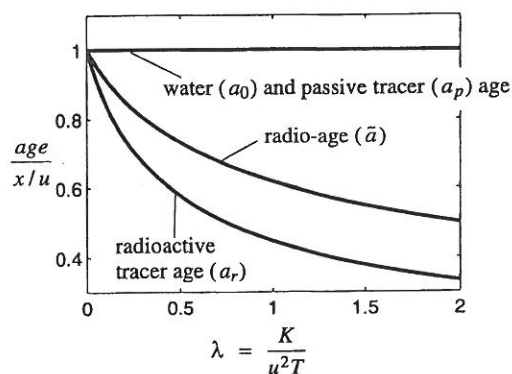

Fig. 11. Graphical representation of the ages listed in Table 2, i.e. the ages obtained in the one-dimensional, idealised English Channel model. All ages are scaled by x/u, are valid downstream of the source only ($0 \leq x < \infty$), and are represented as functions of the dimensionless parameter λ.

lar to those obtained in the idealised instantaneous mixing the problem dealt with in Section 3, i.e.

$$a_r(x) < \tilde{a}(x) < a_p(x) = a_0(x). \qquad (4.6)$$

On the other hand, the differences between the ages increase as λ increases (Fig. 11). In other words, the larger the impact of diffusion, the larger the discrepancies between the ages. Finally, if the diffusivity K is kept constant, the dimensionless number λ decreases as T increases, which reduces the difference between the ages (Fig. 11). This is readily understood: increasing T means diminishing the rate at which the radioactive tracers decays, implying that the latter becomes "more passive" so that its concentration and hence its age get closer to those of the passive one.

To investigate the impact of mixing on the ages that may be simulated by means of the GHER model of the fate of tracers discharged at La Hague into the English Channel, inspiration may be found in the analytical solutions above. Clearly, a passive tracer and a radioactive one are needed. As already men-

tioned, technetium-99 plays the role of the former, implying that a radioactive tracer now needs to be introduced. The latter is a mere numerical artefact. Its rate of release is prescribed to be equal to that of the passive tracer. Therefore, the concentrations of both tracers are approximately equal in the vicinity of La Hague, which is why a radio-age, at any time t and location $\mathbf{x}$ in the domain of interest, may be evaluated as

$$\tilde{a}(t,\mathbf{x}) = T \log \frac{C_p(t,\mathbf{x})}{C_r(t,\mathbf{x})}, \qquad (4.7)$$

a formula which is, roughly speaking, in agreement with relation (1.7). Fig. 12 represents the surface value of this radio-age, obtained by taking $T = 600$ days.

Table 2 suggests that the ages, as well as the differences between them, should be sensitive to the horizontal diffusivity. However, the ages derived from the numerical model hardly vary when this parameter is modified, indicating that horizontal dif-
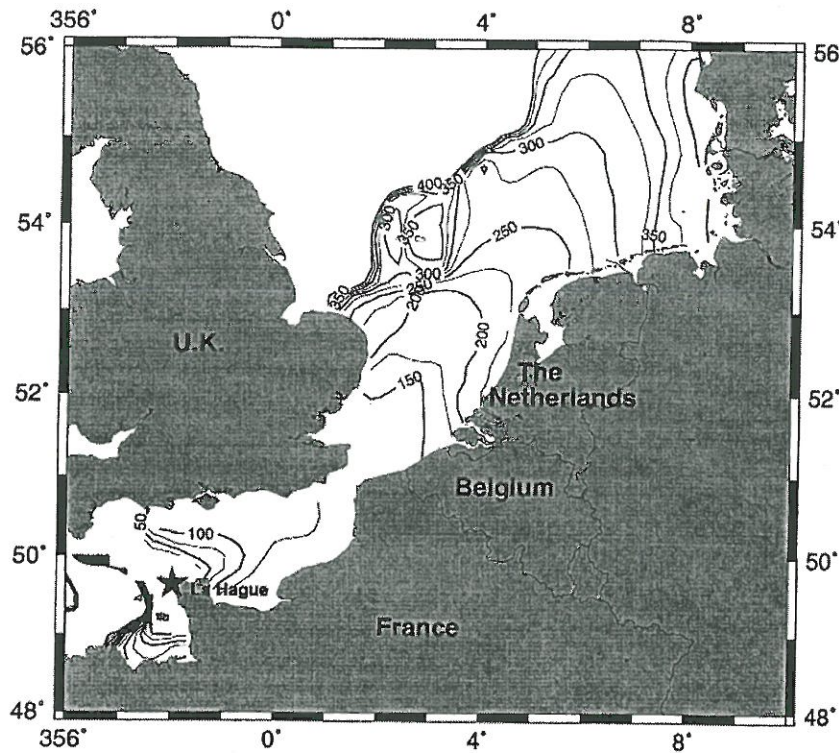

Fig. 12. Surface value of the radio-age (in days) defined by relation (4.7), as simulated numerically on March 1, 1987. The timescale of decay of the radioactive tracer, T, is taken to be 600 days. The contour interval is 25 days.

fusion per se plays a minor role in the present numerical simulations—provided the horizontal diffusivity is given physically acceptable values. The reason thereof is that more processes contribute to the horizontal spreading of the properties than the horizontal diffusivity specified in the equations of the model. While numerical diffusion is minimised by the use of a gradient-preserving TVD scheme, explicitly described mesoscale processes and short-term wind events are responsible for the largest part of the diffusion observed at the large spatial and temporal scales, in which we are interested. In particular, shear dispersion, produced by the combined effect of vertical diffusion and sheared horizontal advection by large tidal currents, accounts for a large part of the observed horizontal diffusion.

As the present article is not intended to present a detailed analysis of model results, the effects of these various processes will not be examined herein. It is sufficient to realise that the horizontal diffusion coefficient K introduced in the idealised one-dimensional model, does not correspond to the diffusion coefficient used in the three-dimensional transport model, but must be interpreted as a crude parameterisation of all the diffusion-like processes.

According to the idealised results depicted in Fig. 11, decreasing the timescale of decay of the radioactive tracer, T, should increase the difference between

the ages. This is confirmed by the numerical results represented in Fig. 13, in which, as expected, the age of the passive tracer is generally greater than the radio-age. So, the three-dimensional simulations are, to a large extent, in agreement with the results of the simple, steady-state, one-dimensional model presented in Table 2.

The fate of tracers released at La Hague is representative of the transport through the English Channel, only if mixing across the Channel is sufficiently intense. To examine the relevance of this assumption, an artificial, passive constituent, tracing the "English Channel water," is introduced. This water mass, which is put forward for the purpose of the present study, is defined as follows: its concentration is forced to be equal to unity along a north–south transect of the English Channel, of which the southern end is very close to La Hague (Fig. 14); its age is set to zero in the same section. The concentration and the age concentration of the English Channel water are simulated by solving the relevant equations, i.e. the equations governing the concentration and age concentration of a passive tracer. Then, the age of this tracer is readily calculated. The latter is generally significantly smaller than the ages associated with tracers originating from La Hague (Figs. 13 and 14). An explanation thereof may be found in Guegueniat et al. (1995). According to these authors,
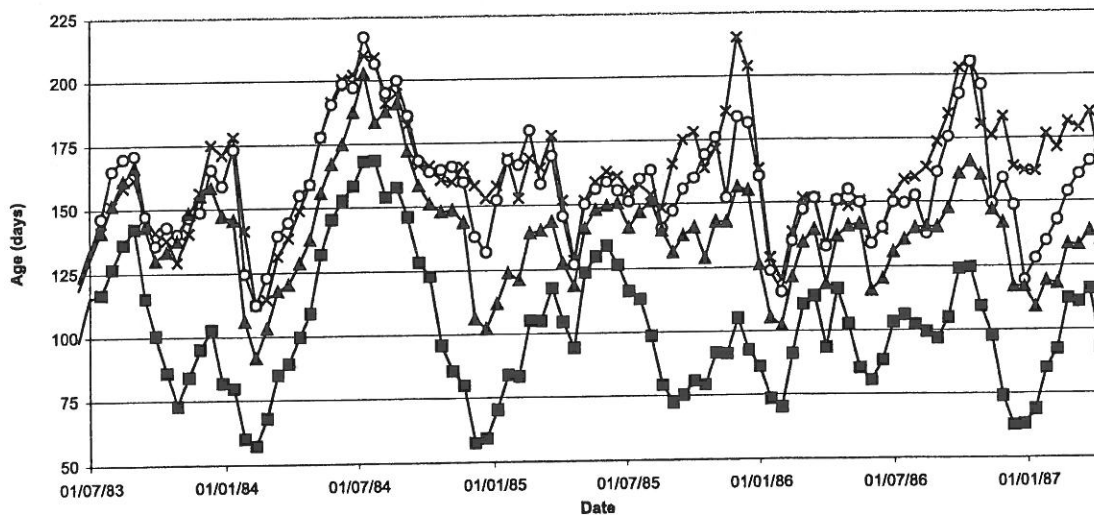

Fig. 13. Time series of various ages simulated at the top of the water column located at (51°45′N, 3°E), i.e. in the Southern bight of the North Sea. This point is marked in Fig. 7. The symbols ×, ○, ▲, and ■, identify the age of the passive tracer, the radio-age evaluated with $T = 600$ days, the radio-age evaluated with $T = 60$ days, and the age of the English Channel water, respectively.
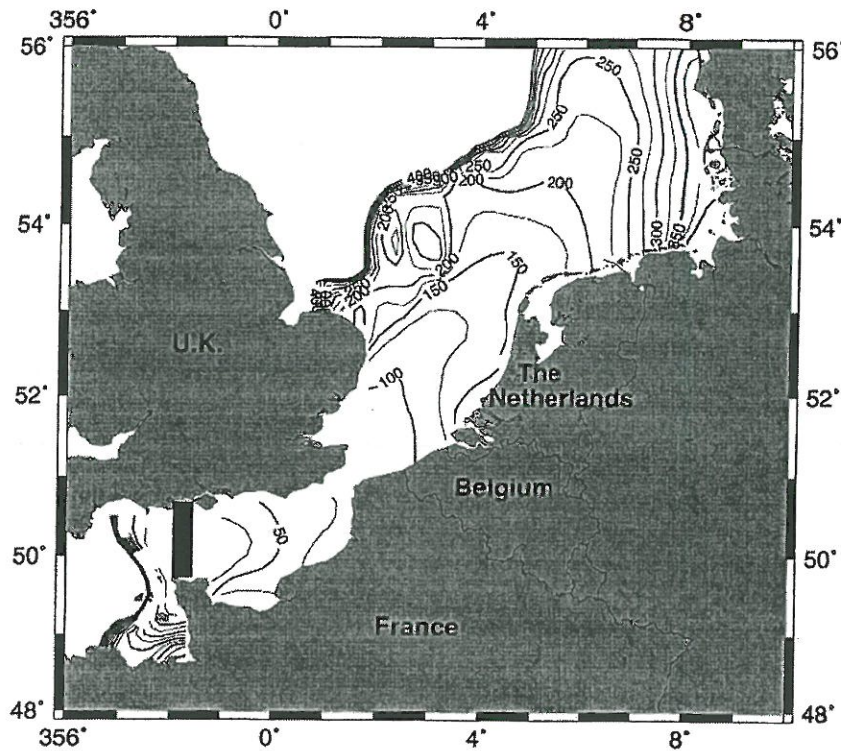
Fig. 14. Surface value of the age of the English Channel water (in days), as simulated numerically on March 1, 1987. The contour interval is 25 days. The age of the English Channel water is set to zero in the black-coloured grid boxes making up a north–south transect running through the English Channel from the English coast to the neighbourhood of La Hague.

the long-term transport through the English Channel is achieved through two routes, namely, a rapid central route and a slow coastal route. The latter, which is adjacent to the French coast, is slower because it includes a number of residual gyres. Obviously, most of the tracer particles from La Hague enter the slow route, which is why their age tends to be larger than that of the English Channel water, which mostly follows the quicker, central route. As a consequence, studying the fate of tracers released into the English Channel by the La Hague nuclear fuel reprocessing plant, may not be sufficient to fully grasp the transport processes occurring in the English Channel and, presumably, downstream of it in the southern North Sea.

In Section 5, the impact of mixing on various ages is investigated once again. The flow to be considered, namely the general circulation in the World Ocean, is believed to be more complex than

that taking place in the English Channel and the southern North Sea.

5. Age in the World Ocean

According to England (1995), the "World Ocean circulation at its largest scale can be thought of as a gradual renewal or ventilation of the deep ocean by water that was once at the sea surface." Thus, estimating the age as the time elapsed since leaving the surface layers is likely to provide useful insight into the ventilation processes of the World Ocean. This is why the age is a popular diagnostic tool in this domain of interest.

Diffusive transport is believed to be of a smaller importance than advection in the interior of the World Ocean, i.e. the whole World Ocean except the surface and bottom boundary layers, the thickness of

which rarely exceeds a few hundreds of metres. Thus, in most of the domain of interest, neglecting diffusion may be considered as a fair approximation. This is actually the key assumption of radioisotope-dating techniques, yielding radio-ages—a concept which was outlined above, in Section 1, and used in Sections 3 and 4. Such an approach is widely used in World Ocean studies. Therefore, it is appropriate to examine to what extent radio-ages are different from ages ensuing from the theory developed herein. Addressing this issue in an exhaustive manner is beyond the scope of the present article. Below, only one type of radio-age is considered in detail. The latter is designed specifically for numerical models, but bears some similarities with the radiocarbon age. All the numerical results presented in this section are obtained by the World Ocean general circulation model included in CLIO, the Coupled Large-scale Ice-Ocean model developed at the Université catholique de Louvain (Goosse et al., 2000), with the grid and forcings used by Campin et al. (1999).

The domain of interest V is the World Ocean. Its boundary S consists of two distinct parts, the ocean–atmosphere interface, S^A, and the ocean bottom, S^B. No water crosses the surface S, i.e. at any time and location on S, the water velocity $\mathbf{u}$ satisfies:

$$\mathbf{u} \cdot \mathbf{n} = 0, \tag{5.1}$$

where $\mathbf{n}$ is the outward unit normal to S. A passive tracer and a radioactive one are considered: the source/sink terms included in the equations governing the concentration and age concentration of these tracers are $(P_p, D_p, \pi_p, \delta_p) = (0, 0, 0, 0)$ and $(P_r, D_r, \pi_r, \delta_r) = (0, C_r/T, 0, \alpha_r/T)$, respectively—where $T \log 2$ is the half-life of the radioactive tracer. There is no diffusive flux of these tracers across the boundary S^B: at any time and location on S^B, the relations

$$\left(\mathbf{K} \cdot \nabla C_p \right) \cdot \mathbf{n} = 0 = \left(K \cdot \nabla C_r \right) \cdot \mathbf{n}, \tag{5.2}$$

hold true. At the ocean surface, the concentration of both tracers is prescribed to be constant; at any time and location on S^A, the tracer concentrations satisfy

$$\left(C_p, C_r \right) = \left(C_p^A, C_r^A \right), \tag{5.3}$$

where C_p^A and C_r^A are positive constants. At the initial instant, the tracer concentrations are assumed to be zero; at any location $\mathbf{x}$ in the domain of interest, the initial concentrations are

$$C_p(t = 0, \mathbf{x}) = 0 = C_r(t = 0, \mathbf{x}). \tag{5.4}$$

For this tracer transport problem, the radio-age that is in agreement with formula (1.7) is obviously

$$\tilde{a}(t, \mathbf{x}) = T \log \frac{C_r^A / C_p^A}{C_r(t, \mathbf{x}) / C_p(t, \mathbf{x})}. \tag{5.5}$$

Even if the timescale T is set equal to that of carbon-14, i.e. 8266 years (Krane, 1988), the radio-age (Eq. (5.5)), once a steady state is reached, can be viewed at best as an idealised version of that ensuing from the carbon-14 method based on in situ measurements. The first reason thereof is that the concentration at the ocean surface of carbon-12 and carbon-14 are actually not constant (e.g. Toggweiler et al., 1989; Campin et al., 1999). In addition, the carbon isotopes are involved in a wealth of biological processes, so that the production/destruction terms to be taken into account, to represent the evolution of their concentrations, should be far more complex than the simple expressions selected above. In other words, no claim is made that the concentrations C_p and C_r, which are the solutions of the partial differential problem presented above, can be realistic approximations of the in situ concentrations of carbon-12 and carbon-14, respectively. Nonetheless, it is believed that the idealised approach adopted herein is sufficiently relevant, especially for showing that ages estimated by means of different methods cannot be equal.

The partial differential problem from which the concentration of the passive tracer and that of the radioactive one may be obtained is linear. Thus, scaling the tracer concentrations by dividing them by their surface values does not imply any modification in the mathematical expressions related to these concentrations, except for the surface boundary conditions (Eq. (5.4)) and the radio-age definition (Eq. (5.5)), which must be transformed to

$$\left(C_p, C_r \right) = (1, 1) \tag{5.6}$$

and

$$\tilde{a}(t, \mathbf{x}) = T \log \frac{C_p(t, \mathbf{x})}{C_r(t, \mathbf{x})}. \tag{5.7}$$

For simplicity, the concentrations scaled in this way are used in the remainder of the present section.

Clearly, all ages are zero at the ocean surface; at any time and location on S^A, the age concentrations must satisfy

$$\alpha_p = 0 = \alpha_r. \tag{5.8}$$

As the surface S^B is impermeable, there must be no age concentration flux across this surface; at any time and location on S^B, the following boundary conditions are to be enforced

$$(\mathbf{K} \cdot \nabla \alpha_p) \cdot \mathbf{n} = 0 = (\mathbf{K} \cdot \nabla \alpha_r) \cdot \mathbf{n}. \tag{5.9}$$

On the other hand, it is assumed that all ages are zero at $t = 0$: at any location $\mathbf{x}$ in the domain of interest, the initial age concentrations are

$$\alpha_p(t = 0, \mathbf{x}) = 0 = \alpha_r(t = 0, \mathbf{x}). \tag{5.10}$$

Taking into account the initial and boundary conditions above (Fig. 15), the concentration and age concentration of the passive tracer, as well as those of the radioactive one, may be obtained by solving the relevant partial differential equations. Then, the age of the two tracers and the radio-age (Eq. (5.7)) are readily calculated. If mixing processes were neg-

Fig. 15. Vertical section in the World Ocean, V, illustrating the initial and boundary conditions to be applied to the partial differential problem that provides the age of the passive tracer a_p, that of the radioactive tracer a_r, and the radio-age $\bar{a}$. The surface boundary conditions are in agreement with the scaling of the tracer concentrations leading to Eq. (5.6).

ligible, all ages would be equal. Of course, this is wrong since the diffusivity tensor $\mathbf{K}$ is positive-definite. In fact, it will be seen that at any time $t \ (\geq 0)$ and location $\mathbf{x}$ in the domain of interest, the ages satisfy the following inequalities:

$$\boxed{0 \leq a_r(t, \mathbf{x}) \leq \tilde{a}(t, \mathbf{x}) \leq a_p(t, \mathbf{x}) \leq t.} \tag{5.11}$$

That the ages are all greater than or equal to zero is the least that can be demanded. In addition, that all ages must be smaller than or equal to the elapsed time is also obvious. However, the other inequalities included in (5.11) are more remarkable, and are similar to those prevailing in the problems dealt with in Sections 3 and 4.

To prove that inequalities (5.11) hold true, use must be made of the following lemma:

Lemma. *Assume that the function χ satisfies in the domain of interest V the partial differential equation*

$$\frac{\partial \chi}{\partial t} = P_\chi - D_\chi - \nabla \cdot (\mathbf{u}\chi - \mathbf{K} \cdot \nabla \chi), \tag{5.12}$$

where the production rate is positive, $P_\chi \geq 0$, while the sink term is of the form $D_\chi = \gamma \chi$, with $\gamma \geq 0$. The initial value of χ is zero at any point in V. At any time $t > 0$, the boundary conditions $\chi = \chi^A$, with $\chi^A \geq 0$, and $(\mathbf{K} \cdot \nabla \chi) \cdot \mathbf{n} = 0$ are satisfied on the surfaces S^A and S^B, respectively. Then, at any time $t \geq 0$ and location $\mathbf{x}$ in V, χ is positive:

$$0 \leq \chi(t, \mathbf{x}). \tag{5.13}$$

This lemma is demonstrated in Appendix C, by having recourse to a mathematical technique that is largely inspired by Lewandowski (1997).

If the production rate P_χ and the decay coefficient γ are set to zero, while the boundary value χ^A is prescribed to be equal to unity, function χ satisfies a partial differential problem that is equivalent to that governing the evolution of the concentration of the passive tracer C_p. Therefore, χ is equal to C_p at any time and location in the domain of interest. Then, the lemma above implies that C_p is positive at any time and location in the domain of interest. A similar approach may be used for a series of relevant variables. In fact, by specifying in a suitable manner the
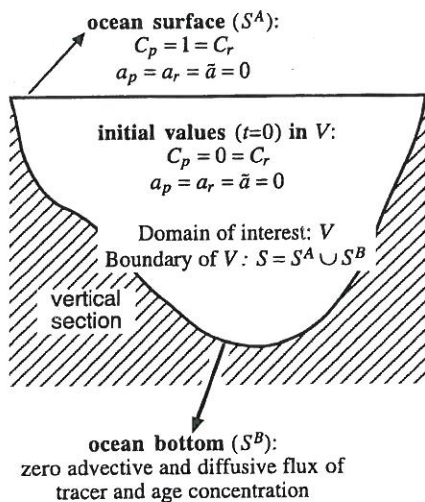
Table 3
The values successively taken by χ, which is governed by Eq. (5.12)
The corresponding production rate P_χ, the decay coefficient γ, associated with sink term $D_\chi = \gamma\chi$, the initial value at $t = 0$, the boundary value χ^A on S^A, and the diffusive flux through boundary S^B are also provided. Function ϕ is defined in relation (5.14).

χ	C_p	C_r	$C_p(t - a_p)$	$C_p(a_p - \tilde{a})$	$C_r(\tilde{a} - a_r)$	$C_r(a_r - 0)$
P_χ	0	0	0	$(T\phi)/(C_p C_r^2)$	$(T\phi)/(C_r C_p^2)$	C_r
γ	0	$1/T$	0	0	$1/T$	$1/T$
$[\chi]_{t=0}$	0	0	0	0	0	0
χ^A	1	1	t	0	0	0
$[(\mathbf{K} \cdot \nabla\chi) \cdot \mathbf{n}]_{S^B}$	0	0	0	0	0	0

initial and boundary conditions, the production rate and the decay coefficient of function χ (Table 3), the latter may successively be made to be equal to C_p, C_r, $C_p(t - a_p)$, $C_p(a_p - \tilde{a})$, $C_r(\tilde{a} - a_r)$, and $C_r(a_r - 0)$. Clearly, the initial values of these variables are zero. So are their diffusive flux through the surface S^B, $[(\mathbf{K} \cdot \nabla\chi) \cdot \mathbf{n}]_{S^B}$. Proving that the relevant values of P_χ, γ and χ^A are positive, which is required in the hypotheses of the lemma above, is trivial as is clear from Table 3, except for the production rates of $C_p(a_p - \tilde{a})$ and $C_r(\tilde{a} - a_r)$, which are proportional to the function ϕ

$$\phi = C_r^2 \nabla C_p \cdot \mathbf{K} \cdot \nabla C_p + C_p^2 \nabla C_r \cdot \mathbf{K} \cdot \nabla C_r - 2 C_p C_r \nabla C_p \cdot \mathbf{K} \cdot \nabla C_r, \tag{5.14}$$

which must be seen to be positive. Tensor $\mathbf{K}$ being symmetric and positive-definite, it can be expressed as

$$\mathbf{K} = \mathbf{D}^T \cdot \mathbf{D}, \tag{5.15}$$

where $\mathbf{D}$ is an appropriate tensor, and $\mathbf{D}^T$ is the transposed of $\mathbf{D}$. Therefore, substituting relation (5.15) into definition (5.14), it follows that function ϕ may be cast into the form

$$\phi = (C_p \mathbf{D} \cdot \nabla C_r - C_r \mathbf{D} \cdot \nabla C_p) \cdot (C_p \mathbf{D} \cdot \nabla C_r - C_r \mathbf{D} \cdot \nabla C_p), \tag{5.16}$$

which obviously is positive. Then, the lemma above implies that, at any time and location in V, the inequalities

$$C_p, C_r, C_p(t - a_p), C_p(a_p - \tilde{a}), C_r(\tilde{a} - a_r),$$
$$C_r(a_r - 0) \geq 0, \tag{5.17}$$

hold valid. As a consequence, inequalities (5.11) are correct. QED.

So far, only one type of radio-age has been considered, namely that defined in formula (5.7), which is inspired by Eq. (1.7). Another radio-age was established in Section 1, i.e. expression (1.6), in which only the concentration of a radioactive tracer is needed. This expression could be adapted to the estimation of the age in the World Ocean, leading to the alternative radio-age

$$\tilde{a}'(t,\mathbf{x}) = T \log \frac{1}{C_r(t,\mathbf{x})}. \tag{5.18}$$

As already pointed out, radio-ages (1.6) and (1.7) are equivalent in the no-diffusion limit. However, diffusive processes, though not dominant, cannot be neglected in the World Ocean. Therefore, the radio-age $\tilde{a}$, defined by relation (5.7), is unlikely to be equal to the alternative formulation $\tilde{a}'$. In fact, using the lemma above, it may be seen that, at any time t and location $\mathbf{x}$ in the domain of interest, the alternative radio-age satisfies the inequalities

$$\tilde{a}(t,\mathbf{x}) \leq \tilde{a}'(t,\mathbf{x}) \leq t. \tag{5.19}$$

Let χ be equal to $C_p(a_p - \tilde{a}')$. Then, introduce this expression in Eq. (5.12). As the corresponding production term,

$$P_\chi = \frac{\phi T}{C_p C_r^2} - \frac{T}{C_p} \nabla C_p \cdot \mathbf{K} \cdot \nabla C_p,$$

for $\chi = C_p(a_p - \tilde{a}')$, \tag{5.20}

may be positive or negative, the alternative radio-age may be greater or lesser than the passive tracer age.

Therefore, the alternative radio-age $\tilde{a}'$ is less representative of the age of the passive and radioactive
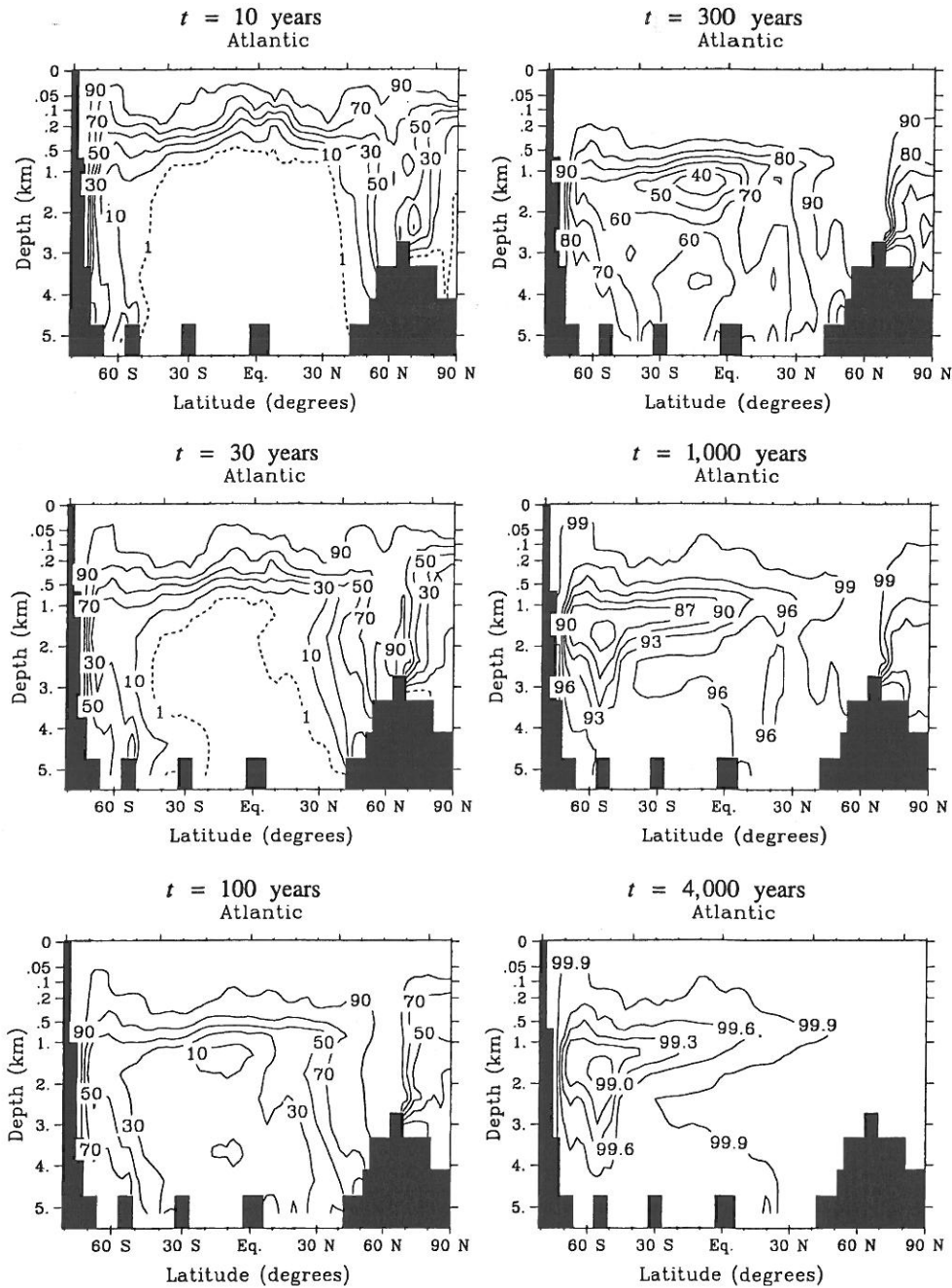

Fig. 16. Zonal average, taken over the Atlantic basin, of the passive tracer concentration C_p (in 10^{-2}), as simulated numerically after 10, 30, 100, 1000, and 4000 years. The dotted isoline corresponds to the value 10^{-2}. Owing to the initial and boundary conditions satisfied by the passive tracer, the latter may be regarded as a marker of the surface water.

tracers than the radio-age $\tilde{a}$, rendering the latter superior to the former for transient calculations.

As time progresses, the concentrations, age concentrations and ages tend toward their steady-state
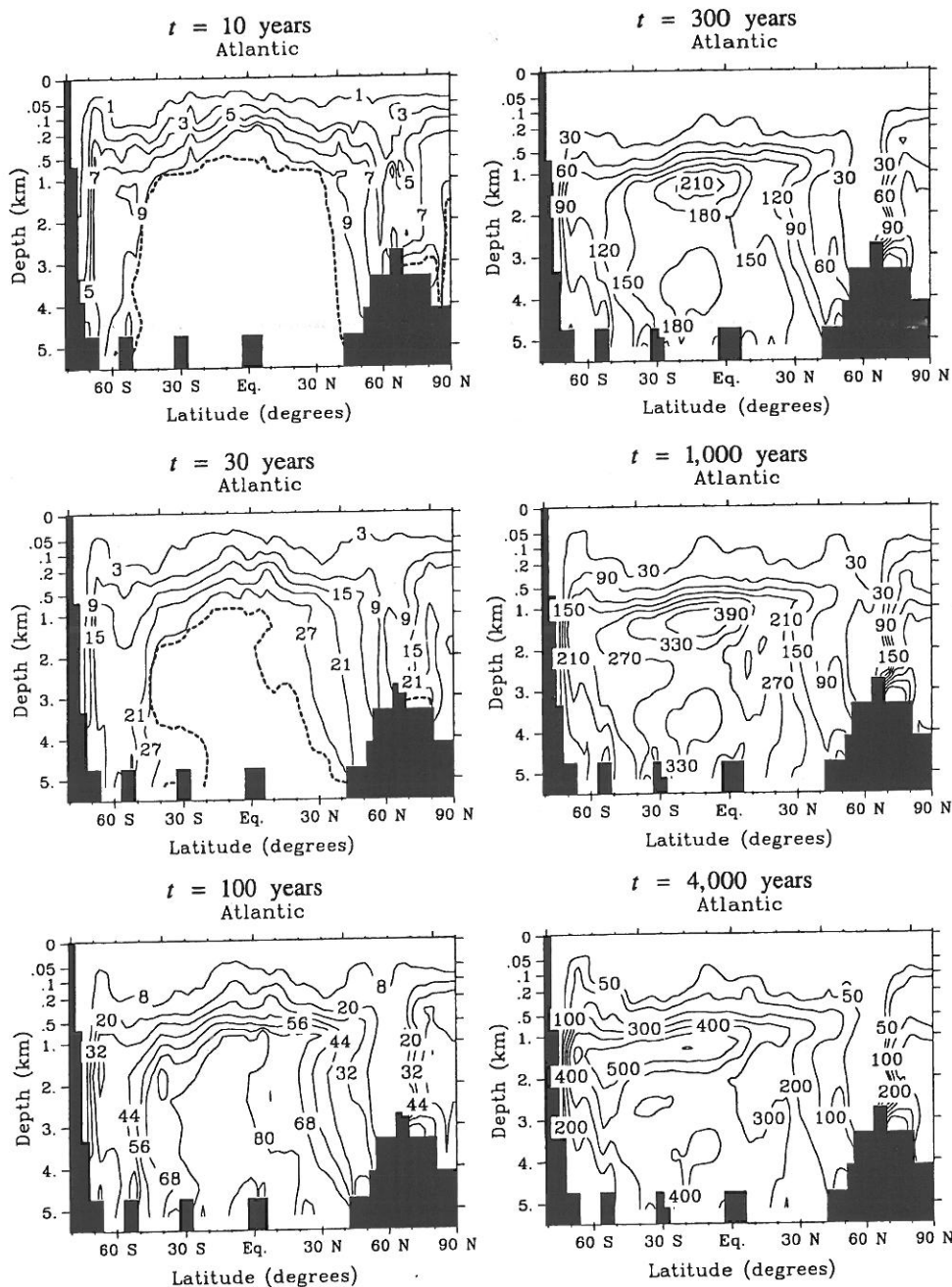
Fig. 17. Zonal average, taken over the Atlantic basin, of the passive tracer age a_p (in years), as simulated numerically after 10, 30, 100, 1000, and 4000 years. The dotted isoline is associated with $C_p = 10^{-2}$. Owing to the initial and boundary conditions satisfied by the passive tracer, the latter may be regarded as a marker of the surface water. Therefore, the age displayed above may be interpreted as that of the surface water. The age is not represented in the regions where $C_p < 10^{-2}$.

limit, as is exemplified in Figs. 16 and 17. In particular, the steady-state concentration of the passive tracer is equal to unity at any location **x** in the World Ocean, i.e.

$$\lim_{t \to \infty} C_p(t,\mathbf{x}) = 1. \tag{5.21}$$

Hence, by virtue of definitions (5.7) and (5.18), the two radio-ages introduced above exhibit the same steady-state value:

$$\lim_{t \to \infty} \tilde{a}(t,\mathbf{x}) = \lim_{t \to \infty} \tilde{a}'(t,\mathbf{x}). \tag{5.22}$$

Thus, there is no need to refer again to the alternative radio-age—as it is inferior to the radio-age $\tilde{a}$ for transients and is equivalent to the latter in the steady state limit.

Tracing a particular water mass as a passive tracer was achieved by several authors (e.g. Cox, 1989; Hirst, 1999; Goosse et al., 2001). In this respect, the concentration of the passive tracer may be interpreted as the concentration of the surface water, i.e. the water that has touched the ocean surface. As time progresses, the global average of the surface water concentration increases as the number of water parcels grows which have been in contact at least once with the atmosphere–ocean interface. Of course, in the limit $t \to \infty$, as indicated in Eq. (5.21), the concentration of the surface water is equal to unity. Then, the age of the passive tracer or, equivalently, the age of the surface water obeys the steady-state equation

$$\nabla \cdot \left(\mathbf{u} \, a_p - \mathbf{K} \cdot \nabla a_p \right) = 1, \tag{5.23}$$

which is similar to that put forward in Thiele and Sarmiento (1990) or England (1995) to estimate ventilation or water age. Clearly, this equation is equivalent to the steady-state limit of Eq. (3.10), the equation governing the age of the water. In other words, in the limit $t \to \infty$, the age of the water and the age of the surface water are equivalent. The transient behaviours of these ages, however, are completely different.

As depicted in Fig. 18, the modelled Atlantic meridional circulation is consistent with the circulation scheme widely referred to as the "ocean conveyor belt," proposed by Gordon (1986) and popularised by Broecker (1991): water generally flows northward close to the surface; North Atlantic Deep
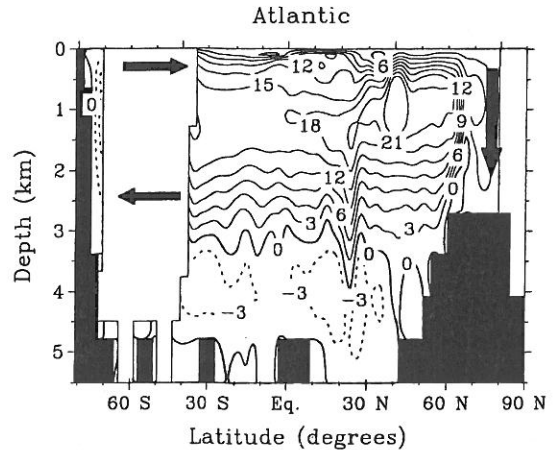

Fig. 18. Illustration of the meridional transport in the Atlantic, which results from the integral of the meridional and vertical velocity components, over the longitude from the western side of the basin to the eastern one. This transport is divergence-free. Hence, it can be viewed as deriving from the streamfunction, of which some isolines are displayed above. The contour interval is 3×10^6 m^3 s^{-1}. The arrows indicate the direction of the transport.

Water (NADW) is formed by convective processes in regions around 60°N; NADW returns southward at a depth of the order of 2–3 km, flowing on top of Antarctic Bottom Water, which was formed in the vicinity of the Antarctic. The Atlantic zonal mean of the water age (Fig. 19) is visibly consistent with the meridional circulation as displayed in Fig. 18.

In the Atlantic basin, the oldest water (age > 500 years) is located at intermediate depth, around 1.5 km, between 40° and 60°S. However, this water appears tagged by surface tracer after only 30 years of transient simulation (Fig. 16). The large difference between these two timescales is a signature of a blended water mass from different origins, some as young as 30 years, but most much older. This is consistent with the age differences displayed in this region in Figs. 19 and 20, which are associated unambiguously with mixing of different water masses.

The model results displayed in Fig. 19 are based on the assumption that the timescale of decay of the radioactive tracer is equal to that of the Carbon-14, i.e. 8266 years. In this case, the difference between the ages is of the order of a few tens of years, i.e. about 5% of the age of the passive tracer. Thus, the
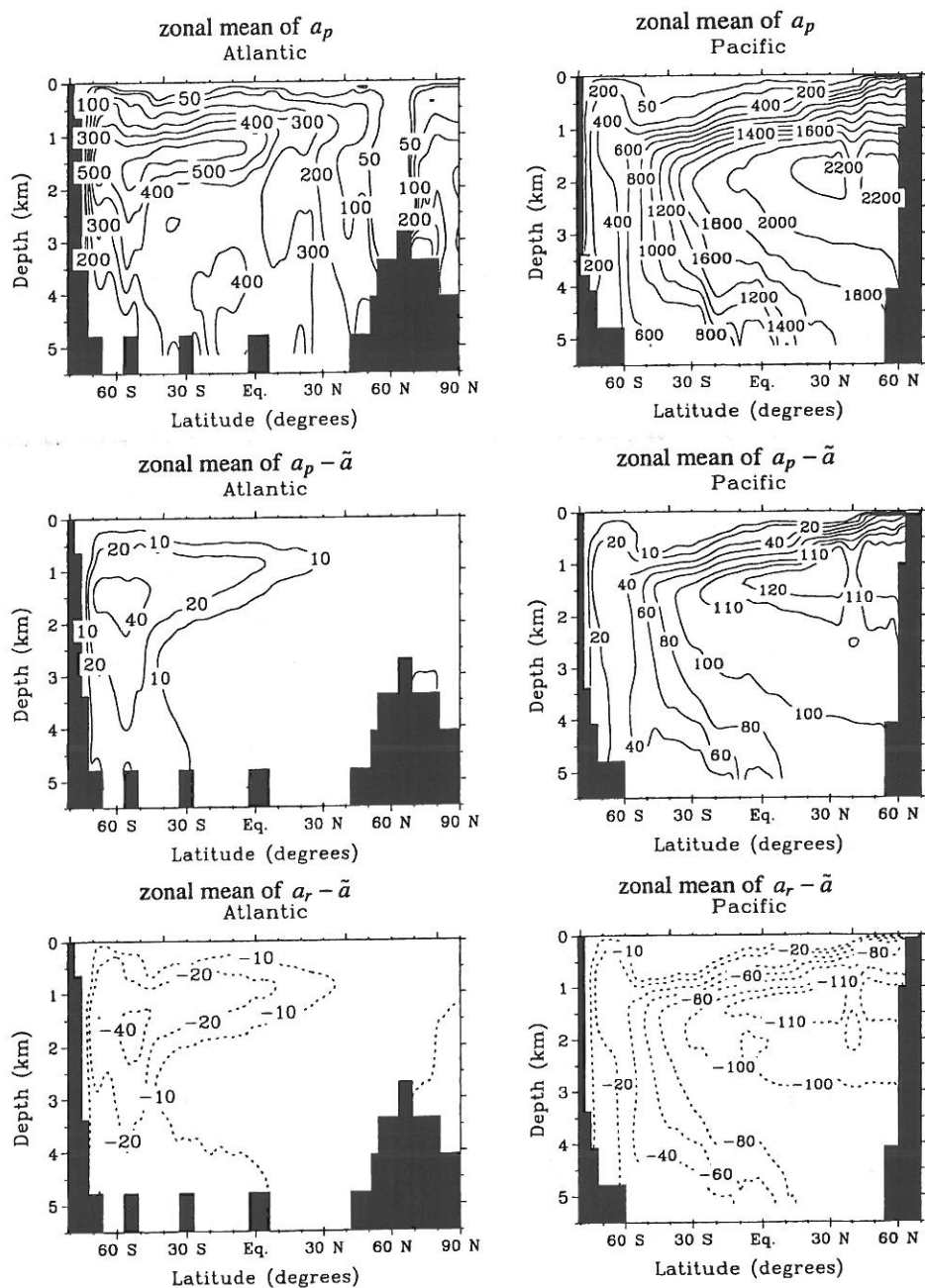
Fig. 19. Zonal average (in years) of the age of the passive tracer, a_p, the difference between the age of the passive tracer and the carbon-14-like radio-age, $a_p - \tilde{a}$, and the difference between the age of the radioactive tracer and the radio-age, $a_r - \tilde{a}$. Results from the Atlantic and the Pacific are displayed in the left-hand and the right-hand columns, respectively. Steady-state quantities are considered. The timescale of decay of the radioactive tracer is equal to that of the carbon-14, i.e. 8266 years.

radio-age based on this timescale is a good estimate of the age of the water, as well as that of the

radioactive tracer. The reason thereof is that the timescale of decay is sufficiently large that the con-

Table 4

Averages over the World Ocean—denoted by operator "$\langle \rangle$"—of the steady-state values of the radio-age, $\tilde{a}$, and of the differences $a_p - \tilde{a}$ and $\tilde{a} - a_r$, as simulated for two different values of the radioactive decay time scale T

All quantities are expressed in years.

	$T = 8266$	$T = 1000$
$\langle a_p - \tilde{a} \rangle$	51	260
$\langle \tilde{a} \rangle$	848	639
$\langle \tilde{a} - a_r \rangle$	47	151

centration variations due to the radioactive decay are sufficiently small that the associated diffusive fluxes

—which cause the ages to be different—are rather negligible. However, if the timescale of decay is decreased to a much smaller value, say 1000 years, then the difference between the ages grows up to an order of magnitude comparable with that of the age of the water, as is shown in Table 4 and Fig. 20. These results indicate that the order of magnitude of the differences between the ages might be a function of a dimensionless number similar to that introduced in Section 4, i.e. the ratio of a diffusivity scale to the product of the square of a typical velocity and the timescale of decay of the radioactive tracer. Nevertheless, the present numerical results are far from sufficient to fully support this proposition. This is
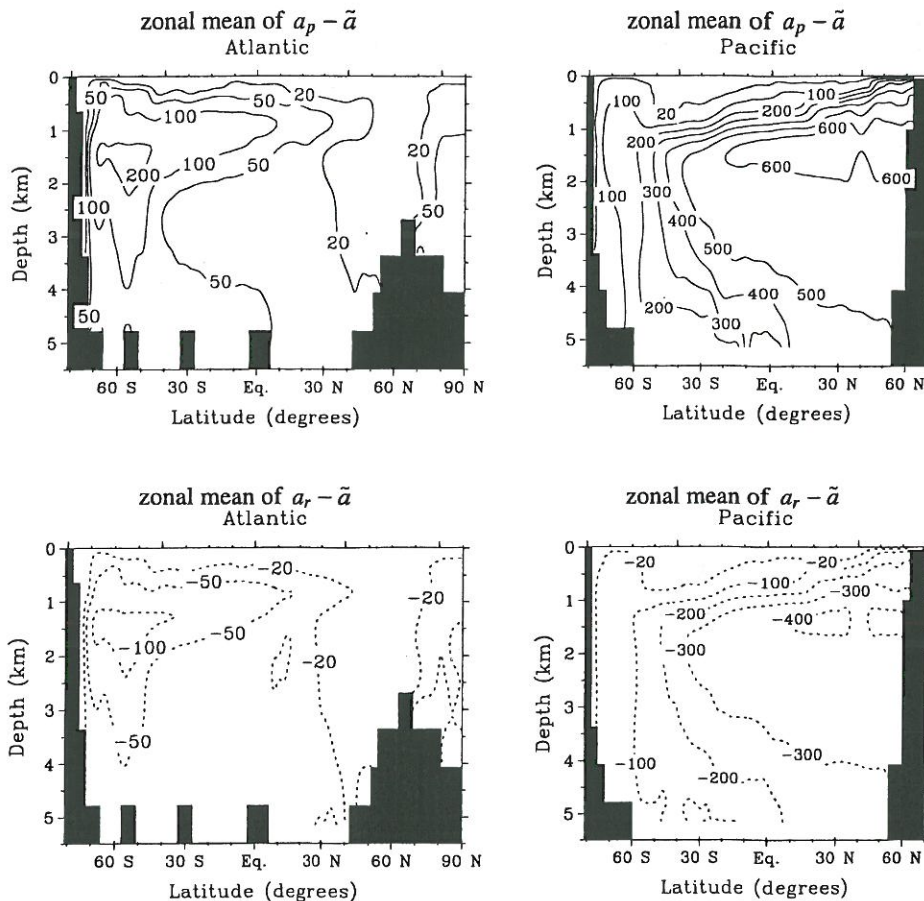

Fig. 20. Zonal average (in years) of the difference between the age of the passive tracer and the carbon-14-like radio-age, $a_p - \tilde{a}$, and the difference between the age of the radioactive tracer and the radio-age, $a_r - \tilde{a}$. Results from the Atlantic and the Pacific are displayed in the left-hand and the right-hand columns, respectively. Steady-state quantities are considered. The timescale of decay of the radioactive tracer is equal to 1000 years, a value much smaller than that of the carbon-14.

why the latter will be investigated in detail in a forthcoming study.

6. Conclusion

The partial differential equations were established from which the concentration distribution function and the age of every seawater constituent, or suitably-defined sets of constituents, may be computed. All phenomena affecting the fate of seawater constituents are taken into account properly, i.e. advection, diffusion, production and destruction. In addition, efforts were devoted to the estimation of the age of the water. Although the theory of the age was presented in the scope of marine modelling, none of the theoretical developments and assumptions that were made are really specific to marine problems. Therefore, it is believed that the general theory of the age may be applied to any fluid flow in which the Boussinesq approximation is resorted to. If this were not the case, the theory should be modified for taking into account compressibility effects, which could be achieved rather easily.

To show that the age theory may lead to powerful diagnostic tools, two flows were briefly investigated, in the English Channel and southern North Sea, and the World Ocean. In all cases, the age of a radioactive tracer and that of a passive tracer were calculated, under the hypothesis that the concentrations of these tracers obey similar initial and boundary conditions. Moreover, the age of the water or that of a particular water mass, treated as a passive tracer, was computed. Because of mixing, the ages were found to be different.

On the other hand, from the concentration of the passive and the radioactive tracer, it was possible to evaluate a radio-age. The very notion of radio-age is inconsistent with the theory developed herein, and is based on apparently conflicting hypotheses. Indeed, the expression of a radio-age, like for instance, relation (1.6) or (1.7), is obtained by assuming that mixing is negligible, while the tracer concentrations must generally be computed by taking into account diffusive processes. Surprisingly, the analytical and numerical results obtained herein lead, to a certain extent, to the rehabilitation of the radio-age concept, at least the radio-age based on formula (1.7)—which

is probably much superior to Eq. (1.6), though relations (1.6) and (1.7) are equivalent if there is no mixing at all. This is due to the fact that, in the problems studied, a remarkably robust set of inequalities was seen to hold true: the age of the radioactive tracer was smaller than the radio-age, and the latter was smaller than the age of the passive tracer, which, in some cases, was equal to the age of the water. Therefore, the radio-age may be viewed as a single approximate value of the age of several seawater constituents, as well as that of the water.

It must be realised, however, that the difference between the ages can be as large as the order of magnitude of the ages. In an idealised version of the English Channel problem, the difference between the ages was seen to be governed by the dimensionless number consisting of the ratio of the diffusivity scale to the product of the square of the characteristic velocity and the timescale of decay of the radioactive tracer: the larger this dimensionless ratio, the larger the difference between the ages. Subsequent numerical experiments carried out by means of the English Channel and southern North Sea model confirmed the importance of this dimensionless ratio, which may be interpreted as a measure of the impact of diffusion. Numerical results obtained in the World Ocean model seem to indicate that the above-mentioned dimensionless number is also relevant to this problem, though this would need to be confirmed by a comprehensive series of model runs.

Clearly, the understanding of the differences between various ages is far from complete. This is why future work will include a comprehensive analysis of the order of magnitude of the terms of the equations from which the age of a passive tracer, that of a radioactive tracer and an appropriate radio-age may be obtained. A series of dimensionless numbers will be derived from this analysis—the relevance of which will need to be assessed in analytical, idealised models—as well as numerical simulations of realistic flows. On the other hand, it will be necessary to examine the usefulness of the age of constituents having complex production and destruction rates, such as those dealt with in biogeochemical models. Finally, the concentration distribution function will be computed in idealised and realistic flows (e.g. Delhez and Deleersnijder, 2001), as the latter contains potentially much more information than the

mean age. This will help to determine whether or not it is sufficient to evaluate the mean age to gain appropriate insight into the phenomena under study.

Acknowledgements

Appendix A

Griffies (1998) reformulated the Gent–McWilliams closure scheme (Gent and McWilliams, 1990; Gent et al., 1995), obtaining a parameterisation which includes a diffusivity tensor having an anti-symmetric part. The diffusivity tensor $\mathbf{K}$, as any tensor, may be formulated as the sum of its symmetric part, $\mathbf{K}_s$, and its anti-symmetric part, $\mathbf{K}_a$, i.e.

$$\mathbf{K} = \mathbf{K}_s + \mathbf{K}_a. \tag{A.1}$$

Hence, the divergence of the diffusive flux of the i-th constituent, which appears in the equation governing the concentration of this constituent, reads

$$\nabla \cdot (\mathbf{K} \cdot \nabla C_i) = \nabla \cdot (\mathbf{K}_s \cdot \nabla C_i) + \nabla \cdot (\mathbf{K}_a \cdot \nabla C_i). \tag{A.2}$$

As $\mathbf{K}_a$ is anti-symmetric, it is always possible to reformulate the dot product $\mathbf{K}_a \cdot \nabla C_i$ as

$$\mathbf{K}_a \cdot \nabla C_i = \mathbf{v} \times \nabla C_i, \tag{A.3}$$

where $\mathbf{v}$ is an appropriate vector. Hence,

$$\nabla \cdot (\mathbf{K}_a \cdot \nabla C_i) = (\nabla \times \mathbf{v}) \cdot \nabla C_i. \tag{A.4}$$

If $\nabla \times \mathbf{v}$ is divergence-free, as in the parameterisation of Griffies (1998), then relation (A.4) may be reformulated as an advective term, i.e.

$$\nabla \cdot (\mathbf{K}_a \cdot \nabla C_i) = \nabla \cdot \left[(\nabla \times \mathbf{v}) C_i \right], \tag{A.5}$$

where $\nabla \times \mathbf{v}$ plays the role of the advective velocity (e.g. Griffies, 1998). Thus, the impact of the anti-symmetric part of $\mathbf{K}$, if any, may be viewed as additional advection. Therefore, without any loss of generality, the diffusivity tensor may be assumed to be symmetric, while bearing in mind that this symmetric tensor must also be positive-definite in order to represent truly diffusive processes, i.e. phenomena which tend, at any time and location, to homogenise the concentration of any constituent.

Appendix B

Demonstrating that inequalities (3.38) hold true is tantamount to showing that

$$\frac{\nu}{1+\nu} \log \frac{1}{\nu} < \log \frac{2}{1+\nu} \tag{B.1}$$

and

$$\log \frac{2}{1+\nu} < \frac{1}{2} \log \frac{1}{\nu} \tag{B.2}$$

are valid if ν satisfies the constraints

$$0 < \nu < 1. \tag{B.3}$$

Let function $F(\nu)$ be defined as

$$F(\nu) = (1+\nu)\log 2 - (1+\nu)\log(1+\nu)$$
$$+ \nu \log \nu. \tag{B.4}$$

Inequality (B.1) holds true if $F(\nu)$ is positive for $0 < \nu < 1$. Since

$$F(1) = 0 \tag{B.5}$$

and

$$\frac{dF}{d\nu} = \log\frac{2\nu}{1+\nu} \qquad (B.6)$$

is negative for $0 < \nu < 1$, $F(\nu)$ is obviously positive for $0 < \nu < 1$. QED.

Inequality (B.2) is equivalent to

$$\log\frac{2}{1+\nu} < \log\frac{1}{\nu^{1/2}}. \qquad (B.7)$$

Hence, it must be seen that

$$\frac{2}{1+\nu} < \frac{1}{\nu^{1/2}} \qquad (B.8)$$

holds true. The latter leads to

$$2\nu^{1/2} < 1 + \nu, \qquad (B.9)$$

or, equivalently,

$$0 < \left(1 - \nu^{1/2}\right)^2, \qquad (B.10)$$

which is obviously true for $0 < \nu < 1$. QED.

Appendix C

It is to be proven that the lemma included in Section 5 holds valid, i.e. it must be seen that function $\chi(t,\mathbf{x})$ is larger than or equal to zero at any time $t(\geq 0)$ and location $\mathbf{x}$ in the domain of interest V. Function $\chi(t,\mathbf{x})$ is governed by the partial differential equation

$$\frac{\partial\chi}{\partial t} = P_\chi - \gamma\chi - \nabla\cdot(\mathbf{u}\chi - \mathbf{K}\cdot\nabla\chi), \qquad (C.1)$$

where $P_\chi \geq 0$ and $\gamma \geq 0$; the velocity field u is divergence-free, i.e. $\nabla\cdot\mathbf{u} = 0$; the diffusivity tensor $\mathbf{K}$ is positive-definite. At the initial instant, $t = 0$, χ is zero at any point in V. The boundary S of the domain of interest is made up of two parts, S^A and S^B. The outward unit normal to surface S is denoted $\mathbf{n}$. At any time $t > 0$, the boundary conditions $\mathbf{u}\cdot\mathbf{n} = 0$, $\chi = \chi^A$, with $\chi^A \geq 0$, and $(\mathbf{K}\cdot\nabla\chi)\cdot\mathbf{n} = 0$ are satisfied on the surfaces S, S^A and S^B, respectively.

Let χ_- represent the negative part of χ, i.e.

$$\chi_- = \frac{|\chi| - \chi}{2}. \qquad (C.2)$$

Obviously, χ_- is zero if χ is positive, and is equal to $-\chi$ if χ is negative. Let V_- denote the sub-domain of V in which $\chi < 0$ or, equivalently, $\chi_- > 0$. Then, demonstrating that χ is positive is equivalent to showing that its negative part, χ_-, is zero at any time and position in the domain of interest V. This is also tantamount to showing that V_- is a vanishing volume.

The surface S_-, which is the boundary of V_-, may have an intersection with S^B, S_-^B, on which the value of χ is not prescribed. On the surface S_-^A, which is common to S^A and S_-, χ_- must be zero, since χ^A is not negative. The remainder of the surface S_-, denoted S_-^V, has no intersection with S, and hence runs through the domain of interest V. Clearly, on S_-^V, χ is zero. Needless to say, even if volume V_- exists, S_-^A or S_-^B, or both, may be vanishing surfaces. However, for the purpose of the demonstration below, S_-^A and S_-^B will be assumed to exist, even if their surface is actually zero.

Multiplying Eq. (C.1) by χ_- and integrating over V yields

$$\int_V \chi_-\frac{\partial\chi}{\partial t}dV = \int_V \chi_- P_\chi dV - \int_V \gamma\chi_-\chi dV$$
$$- \int_V \chi_-\nabla\cdot(\mathbf{u}\chi)dV$$
$$+ \int_V \chi_-\nabla\cdot(\mathbf{K}\cdot\nabla\chi)dV. \qquad (C.3)$$

Each of the integrals above is to be examined separately. Using definition (C.2), it is readily seen that

$$\int_V \chi_- P_\chi dV \geq 0 \qquad (C.4)$$

and

$$-\int_V \gamma\chi_-\chi dV \geq 0. \qquad (C.5)$$

Next, as the divergence of the velocity field $\mathbf{u}$ is zero, having recourse to the divergence theorem, the third integral in the right-hand side of Eq. (C.3) may be transformed as follows:

$$-\int_V \chi_-\nabla\cdot(\mathbf{u}\chi)dV = \frac{1}{2}\int_{V_-}\nabla\cdot(\mathbf{u}\chi^2)dV_-$$
$$= \frac{1}{2}\int_{S_-}(\mathbf{u}\cdot\mathbf{n}_-)\chi^2 dS_-, \qquad (C.6)$$

where $\mathbf{n}_-$ is the outward unit normal to surface S_-. The last integral above is zero, because, $\mathbf{u} \cdot \mathbf{n}_- = 0$ on S_-^A and S_-^B, and $\chi = 0$ on S_-^V. Finally, integrating by parts the fourth integral in the right-hand side of relation (C.3) yields

$$\int_V \chi_- \nabla \cdot (\mathbf{K} \cdot \nabla \chi) dV = \int_{V_-} \nabla \chi \cdot \mathbf{K} \cdot \nabla \chi dV_-$$
$$- \int_{V_-} \nabla \cdot (\chi \mathbf{K} \cdot \nabla \chi) dV_-.$$
(C.7)

Then, applying the divergence theorem leads to

$$\int_{V_-} \nabla(\chi \cdot \mathbf{K} \cdot \nabla \chi) dV_- = \int_{S_-} [(\mathbf{K} \cdot \nabla \chi) \cdot \mathbf{n}_-] \chi dS_-,$$
(C.8)

which is zero, since $\chi = 0$ on S_-^A and S_-^V, and the diffusive flux $(\mathbf{K} \cdot \nabla \chi) \cdot \mathbf{n}_-$ is prescribed to be zero on S_-^B. Hence,

$$\int_V \chi_- \nabla \cdot (\mathbf{K} \cdot \nabla \chi) dV = \int_{V_-} \nabla \chi \cdot \mathbf{K} \cdot \nabla \chi dV_-. \quad \text{(C.9)}$$

If V_- is non-vanishing, there must exist a region in V_- where $|\nabla \chi| \neq 0$. Therefore, the integral (C.9) is zero if V_- vanishes, and is > 0 otherwise, because tensor $\mathbf{K}$ is positive-definite. It follows that the right-hand side of relation (C.3) is zero if V_- vanishes, and is > 0 otherwise.

The left-hand side of relation (C.3) transforms to

$$\int_V \chi_- \frac{\partial \chi}{\partial t} dV = -\frac{1}{2} \int_{V_-} \frac{\partial \chi^2}{\partial t} dV_-. \quad \text{(C.10)}$$

Surfaces S_-^A and S_-^B are motionless, but S_-^V may conceivably move, causing the volume V_- to vary in time. Then, applying Reynolds' transport theorem leads to

$$\int_{V_-} \frac{\partial \chi^2}{\partial t} dV_- = \frac{d}{dt} \int_{V_-} \chi^2 dV_- - \int_{S_-} (\mathbf{v}_- \cdot \mathbf{n}_-) \chi^2 dS_-,$$
(C.11)

where $\mathbf{v}_-$ is the velocity at which S_- moves. The integral over S_- is zero, because $\mathbf{v}_-$ is zero on the

fixed surfaces S_-^A and S_-^B, and $\chi = 0$ on S_-^V. Therefore,

$$\int_V \chi_- \frac{\partial \chi}{\partial t} dV = -\frac{1}{2} \frac{d}{dt} \int_{V_-} \chi^2 dV_-. \quad \text{(C.12)}$$

Finally, by virtue of the results established above,

$$\frac{d}{dt} \int_V (\chi_-)^2 dV_- = -2 \int_{V_-} \nabla \chi \cdot \mathbf{K} \cdot \nabla \chi dV_-. \quad \text{(C.13)}$$

The right-hand side member of the above relation is < 0 if V_- exists, and is equal to zero otherwise. Thus, as $\chi_- = 0$ at any location of the domain of interest at the initial instant $t = 0$, χ_- cannot grow, and hence will remain equal to zero at any time and location. QED.

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