On a multidimensional model for the codiffusion of isotopes: localization and asymptotic behavior

Elena Comparini[a*†] and Maura Ughi[b]

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The paper deals with a system of parabolic–hyperbolic partial differential equations, which models the diffusion of N species of isotopes of the same element, possibly radioactive, in a multidimensional medium. Some qualitative properties of the solutions, such as localization property, are studied together with the asymptotic behavior for large times.

Keywords: isotopes; diffusion; parabolic–hyperbolic systems

1. Introduction

The aim of this paper is to examine a model for the diffusion of N species of radioactive isotopes of the same element in a multidimensional medium.

Let $\mathbf{C} = (c_1, \ldots, c_N)$ be the vector of the N single concentrations, $c_i(\mathbf{x}, t), \mathbf{x} \in \mathbb{R}^n,\ t \geq 0$.

The total concentration c therefore is defined as

$$c = \sum_{i=1}^{N} c_i. \tag{1.1}$$

The model is based on the assumption that the flux $\mathbf{J}_i$ of each isotope is mainly affected by the gradient of the total element concentration in a relative percentage $\frac{c_i}{c}$, that is, $\mathbf{J}_i = -\frac{c_i}{c}\nabla c$, after a suitable scaling.

In the case of radioactive isotopes, the radioactive decay law must be taken into account. In a spatially homogeneous distribution, the decay is expressed as a linear ODE system:

$$\dot{\mathbf{C}} = \Lambda \mathbf{C}, \qquad \mathbf{C} \in \mathbb{R}^N, \tag{1.2}$$

where Λ is a suitable $N \times N$ constant matrix.

Under these assumptions, the mathematical model is the following PDE system for $\mathbf{C}$:

$$c_{it} = \div \left(\frac{c_i}{c} \nabla c \right) + (\Lambda \mathbf{C})_i, \qquad i = 1, \ldots, N. \tag{1.3}$$

This system is a limit case of a model first proposed by Bremer and Cussler [1, 2] for diffusion and mass transfer in fluid systems and later considered by Pescatore [3] for the diffusion of radioactive isotopes of the same element, such as the couple (U^{234}, U^{238}).

In their model, the flux $\mathbf{J}_l$ depends on the gradient both of c_i and c, that is, after a scaling, $\mathbf{J}_i = -\left(k\nabla c_i + \frac{c_i}{c}\nabla c\right)$, with k a positive constant. Pescatore pointed out that there can be relevant physical situations where k can be taken 'small'. The main motivation for this conjecture is to understand experimental situations presenting strong oscillations of the isotopes spatial distribution for large time, which are difficult to justify with the classical Fick's law. When k is positive, the resulting PDE system has been studied by the authors in

[a] *Dipartimento di Matematica e Informatica 'Ulisse Dini', Università di Firenze, Viale Morgagni 67/A, Firenze I-50134 , Italy*
[b] *Dipartimento di Matematica e Geoscienze, Università di Trieste, V. Valerio 12/b, Trieste I-34127, Italy*
* *Correspondence to: Elena Comparini, Dipartimento di Matematica e Informatica 'Ulisse Dini', Università di Firenze, Viale Morgagni 67/A, Firenze I-50134, Italy.*
† *E-mail: elena.comparini@math.unifi.it*

[4] for $\mathbf{x} \in \mathbb{R}^n$. It has been proved there that the system of equations is a quasilinear parabolic one, together with the existence and the uniqueness of a classical solution.

System (1.3), that is, $k = 0$, has been considered only in one space dimension by the authors [4–9]. It turned out that the classical solution can have oscillations for large times. Moreover, for any positive time, there might be regions depleted of one or more isotopes, a phenomenon denoted in the literature as 'localization'. This means that if for some $i = 1...., N$ c_{i0} has compact support, then c_i has compact support for $t > 0$.

Systems similar to (1.3) have been considered in connection with problems of population dynamics, tumor growth and so on [10–14]. In all these, models the main assumption is that the diffusion of one species out of a family depends on the gradient of the density of the whole family. All these models, together with the present one (1.3), belong to the following class:

$$\begin{cases} c_{it} = \div(c_i \nabla h(c)) + g_i(\mathbf{c}), \quad i = 1, \dots, N, \\ c = \sum_{j=1}^{N} c_j, \qquad h' > 0. \end{cases}$$

In the case of system (1.3), $h(c) = \ln c$, while $h(c) = c$ in [10–14]. Therefore, the equation satisfied by the total density c is a porous medium type equation for $h(c) = c$. The porous medium equation implies finite speed of propagation, thus justifying a phenomenon as 'localization'. On the other hand, for the system (1.3), the total concentration c is determined by a strictly parabolic equation, so that 'localization' for any positive time seems to be a quite surprising phenomenon.

The aim of the present paper is to analyze whether localization and oscillations hold in the multidimensional space as well.

This turns out to be the case although the methods to be used are quite different from the one-dimensional case.

This paper is organized as follows: Section 2 illustrates the precise statement of the problem and summarizes the results on the existence and the uniqueness of a classical solution proved by the authors in a recent paper [15]. Sections 3 and 4 are focused on the localization properties and on the asymptotic behavior for large times, respectively.

2. Statement of the problem

Let Ω be a bounded compact domain of $\mathbb{R}^n$ with a regular boundary $\partial\Omega$.

Setting $c_N = c - \sum_{j=1}^{N-1} c_i$ and defining $\tilde{\mathbf{C}} = (c_1, \dots, c_{N-1}, c), \tilde{\mathbf{C}}_0 = (c_{10}, \dots, c_{N-1\,0}, c_0)$, the following homogeneous Neumann problem is to be considered:

$$\begin{cases} c_{it} = \div\left(\frac{c_i}{c}\nabla c\right) + (\tilde{\Lambda}\tilde{\mathbf{C}})_i, \quad i = 1, \dots, N-1, & \text{in } Q_T = \Omega \times (0, T), \\ c_t = \Delta c + (\tilde{\Lambda}\tilde{\mathbf{C}})_N, & \text{in } Q_T, \\ \nabla c \cdot \mathbf{n} = 0, & \text{in } \Gamma_T = \partial\Omega \times (0, T), \\ \tilde{\mathbf{C}}(\mathbf{x}, 0) = \tilde{\mathbf{C}}_0(\mathbf{x}) & \text{in } \overline{\Omega}, \end{cases} \tag{2.1}$$

where $\mathbf{n}$ is the outer normal to $\partial\Omega$ and the matrix $\tilde{\Lambda}$ is derived from the matrix Λ in (1.2):

$$\tilde{\Lambda} = \begin{pmatrix} \Lambda_{11} - \Lambda_{1N} & \dots & \Lambda_{1N} \\ \Lambda_{21} - \Lambda_{2N} & \dots & \Lambda_{2N} \\ \vdots & \ddots & \vdots \\ \sum_{m=1}^{N}(\Lambda_{m1} - \Lambda_{mN}) & \dots & \sum_{m=1}^{N} \Lambda_{mN} \end{pmatrix}. \tag{2.2}$$

The assumptions on the initial data are the following:

(H1) $c_{i0} \in C^{2+l}(\overline{\Omega}),\ l > 0, i = 1, \dots, N, 0 \le c_{i0} \le K, c_0 = \sum_{i=1}^{N} c_{i0} \ge k_0 > 0, \nabla c_0 \cdot \mathbf{n} = 0$ on $\partial\Omega$, and
(H2) positivity property of the constant matrix Λ: if $c_{i0} \ge 0$, then $c_i(t) \ge 0,\ \ i = 1, \dots, N,\ \ t > 0$.

The positivity property **H2** is equivalent to assume that the region $V = \{\mathbf{y} \in \mathbb{R}^N : y_i \ge 0,\ \ i = 1, \dots, N\}$ is invariant for the flux generated by the vector field $\Lambda\mathbf{y}$. Therefore, all the non-diagonal elements of Λ must be non-negative (i.e. $\Lambda_{ij} \ge 0\ \forall ij,\ \ i, j = 1, \dots, N$) so that $\Lambda\mathbf{y} \cdot \mathbf{n} \ge 0$ for any $\mathbf{y} \in \partial V$, where $\mathbf{n}$ is the interior normal to ∂V in $\mathbf{y}$.

In the paper [15], it has been proved that in the assumptions **H1**, and **H2**, there exists a unique classical solution of (2.1) ([15], Thm.3.1, Thm.3.2) with $c \in C^{2,1}(\overline{Q}_T),\ \ c_i \in C^{1,1}(\overline{Q}_T),\ \ i = 1, \dots, N-1$, for any T positive. Moreover, there exists a positive constant $\gamma(k_0,\ T, \Lambda)$ such that $c \ge \gamma > 0$ and $0 \le c_i \le c,\ \ 0 \le \sum_{i=1}^{N-1} c_i \le c$, for $i = 1, \dots, N-1$ in $\overline{Q}_T$.

In the same paper [15], it has also been proved that

$$c_i(\mathbf{x}, t) = r_i(\mathbf{x}, t)c(\mathbf{x}, t),\ \ i = 1, \dots, N-1, \tag{2.3}$$

where $(r_1, \dots, r_{N-1}, c) = (\mathbf{r}, c)$ is the unique classical solution of the following problem:

$$\begin{cases} r_{it} = \nabla r_i \cdot \dfrac{\nabla c}{c} + P_i(\mathbf{r}), \ \ i = 1, \dots, N-1, & \text{in } Q_T, \\ c_t = \Delta c + b(\mathbf{r})\, c, & \text{in } Q_T, \\ r_i(\mathbf{x}, 0) = \dfrac{c_{i0}(\mathbf{x})}{c_0(\mathbf{x})}, \ \ i = 1, \dots, N-1, & \text{in } \Omega, \\ c(\mathbf{x}, 0) = c_0(\mathbf{x}), & \text{in } \Omega, \\ \nabla c \cdot \mathbf{n} = 0, & \text{in } \Gamma_T, \end{cases} \tag{2.4}$$

with

$$\begin{cases} P_i(\mathbf{r}) = (\tilde{\Lambda}\tilde{\mathbf{r}})_i - r_i(\tilde{\Lambda}\tilde{\mathbf{r}})_N, \qquad \tilde{\mathbf{r}} = (r_1, \dots, r_{N-i}, 1), \\ b(\mathbf{r}) = (\tilde{\Lambda}\tilde{\mathbf{r}})_N = \tilde{\Lambda}_{NN} + \sum_{k=1}^{N-1} \Lambda_{Nk} r_k = \beta_0 + \sum_{k=1}^{N-1} \beta_k r_k. \end{cases} \tag{2.5}$$

Therefore, the total concentration c has a 'parabolic' behavior because it satisfies a uniformly parabolic equation, while the concentrations of the single isotopes may have a 'hyperbolic' behavior considering that the evolution of r_i satisfies a first-order semilinear hyperbolic system.

In the following sections, it will be proved that there may exist regions depleted of a component c_i (the so-called localization property) and that c_i may have strong oscillations also asymptotically for large time, as it was already found for $n = 1$ [5,7,9].

Some results obtained in [15] are to be recalled here to be used in the following sections. First of all, $\mathbf{r}$ can be constructed in a classical way by the method of the characteristics, and the evolution of $\mathbf{r}$ along each characteristic does not depend on c but only on the polynomial $P_i(\mathbf{r})$ and hence on the decay law Λ.

With a standard notation, let $\mathbf{X}(t; \mathbf{z}, \tau)$ be the characteristic through $(\mathbf{z}, \tau), \mathbf{z} \in \Omega, \ \tau \geq 0$, that is the solution of the ODE system

$$\frac{d\mathbf{X}}{dt} = \mathbf{f}(\mathbf{X}, t), \qquad \mathbf{f} = -\frac{\nabla c}{c}, \qquad \mathbf{X}(\tau; \mathbf{z}, \tau) = \mathbf{z}. \tag{2.6}$$

From the homogeneous Newmann boundary condition on c, it follows that no characteristic can cross the lateral boundary Γ_T. For any $\mathbf{x} \in \Omega$, the evolution of $\mathbf{r}$ along the characteristic starting in $\mathbf{x}$ for $\tau = 0$ is given by

$$R_i(\mathbf{x}, t) = \frac{Y_i(t)}{Y(t)}, \ i = 1, \dots, N-1, \tag{2.7}$$

where $\mathbf{R} = (R_1, \dots, R_{N-1})$ and $\mathbf{Y}(t) = (Y_1, \dots, Y_N)$ is the solution of the ODE system

$$\begin{cases} \dot{\mathbf{Y}} = \Lambda \mathbf{Y}, \qquad \mathbf{Y}(0) = (c_{10}(\mathbf{x}), \dots, c_{N0}(\mathbf{x})), \\ Y = \sum_{k=1}^{N} Y_i. \end{cases} \tag{2.8}$$

Therefore, $\mathbf{r}$ is given by

$$\begin{cases} \mathbf{r}(\mathbf{z}, \tau) = \mathbf{R}(\tau; \mathbf{r}_0(\mathbf{X}(0; \mathbf{z}, \tau))), \\ 0 \leq r_i \leq 1, \ \sum_{i=1}^{N-1} r_i \leq 1. \end{cases} \tag{2.9}$$

As for the total concentration c, from the estimate (2.9), one has that b, defined in (2.5), is bounded by

$$|b(\mathbf{r})| \leq B = |\beta_0| + \max_j |\beta_j|. \tag{2.10}$$

Therefore,

$$0 < w(\mathbf{x}, t)e^{-Bt} \leq c(\mathbf{x}, t) \leq w(\mathbf{x}, t)e^{Bt}, \ \ t \in [0, T], \tag{2.11}$$

where $w(\mathbf{x}, t)$ is the solution of

$$\begin{cases} w_t = \Delta w & \text{in } Q_T, \\ w(\mathbf{x}, 0) = c_0(\mathbf{x}), & \text{in } \Omega, \\ \nabla w \cdot \mathbf{n} = 0, & \text{in } \Gamma_T. \end{cases} \tag{2.12}$$

From assumption **H1** and (2.11), it follows

$$e^{-Bt} \min_{\Omega} c_0 \leq c(\mathbf{x}, t) \leq \max_{\Omega} c_0 e^{Bt}, \ \ t \in [0, T]. \tag{2.13}$$

It should be remarked that if the function $b(\mathbf{r})$ defined in (2.5) is constant (i.e. $\beta_k = 0$, $k = 1, \dots, N-1$, $b \equiv \beta_0$), then c is independent of $\mathbf{r}$, and the problem (2.4) reduces to a hyperbolic semilinear system for $\mathbf{r}$ with c given by

$$c(\mathbf{x}, t) = w(\mathbf{x}, t)e^{\beta_0 t}, \tag{2.14}$$

where w has been defined in (2.12).

The examples 1 and 2 of Section 3 illustrate some physically relevant situations with b constant.

3. Qualitative behavior and localization

This section examines the behavior of the solution of the homogeneous Neumann problem (2.1) in Q_T.

Let $\Omega^0 \subset \Omega$ be an arbitrary region and $\Omega^0(t)$ be the flux tube starting from Ω^0 at time $t = 0$:

$$\Omega^0(t) = \left\{ \mathbf{z} \in \Omega : \mathbf{z} = \mathbf{X}(t; \mathbf{x}_0, 0),\ \mathbf{x}_0 \in \Omega^0 \right\}.$$

As previously discussed, because the system for $\mathbf{r}$ is semilinear, the characteristics do not cross each other, and hence, the boundary of $\Omega^0(t)$ consists of the characteristics starting from the boundary of Ω^0 for $t = 0$.

Let the masses in $\Omega_0(t)$ be defined as

$$\begin{cases} m_i(t) = \int_{\Omega^0(t)} c_i(\mathbf{z}, t)\, d\mathbf{z}, \quad i = 1, \ldots, N, \\ m(t) = \int_{\Omega^0(t)} c(\mathbf{z}, t)\, d\mathbf{z} = \sum_{i=1}^{N} m_i(t). \end{cases} \tag{3.1}$$

By differentiating (3.1), it follows for each single mass

$$\begin{aligned} \dot{m}_i(t) &= \int_{\Omega^0(t)} c_{it}\, d\mathbf{z} + \int_{\partial\Omega^0(t)} c_i \frac{d\mathbf{X}}{dt} \cdot \mathbf{n}\, d\mathbf{s} \\ &= \int_{\Omega^0(t)} (\Lambda \mathbf{C})_i\, d\mathbf{z} + \int_{\Omega^0(t)} \operatorname{div}\left(\frac{c_i}{c} \nabla c\right) d\mathbf{z} + \int_{\partial\Omega^0(t)} -\frac{c_i}{c} \nabla c \cdot \mathbf{n}\, d\mathbf{s}. \end{aligned}$$

From the divergence Theorem and the definition of the characteristics, the equations for the masses are reduced to

$$\dot{m}_i(t) = \left(\Lambda \int_{\Omega^0(t)} \mathbf{C}\, d\underline{z} \right)_i, \quad i = 1, \ldots, N, \tag{3.2}$$

that is, the evolution of the masses along the flux tubes of the characteristics is due to the decay law, Λ, and to the initial mass, just as the physical point of view suggests.

Then $\mathbf{m}(t) = (m_1(t), \ldots, m_{N-1}(t), m(t))$ satisfies the same ODE as $\tilde{\mathbf{C}}$:

$$\dot{\mathbf{m}} = \tilde{\Lambda} \mathbf{m}, \quad \mathbf{m}_0 = \int_{\Omega^0} \tilde{\mathbf{C}}_0(\mathbf{z})\, d\mathbf{z}. \tag{3.3}$$

It is then possible to prove the following estimate of the measure of $\Omega^0(t)$ for $t > 0$:

Proposition 3.1
In hypotheses **H1** and **H2**, the following estimate holds:

$$0 < m(t; \mathbf{m}_0, 0) \frac{e^{-Bt}}{\max_\Omega c_0} \leq |\Omega^0(t)| \leq m(t; \mathbf{m}_0, 0) \frac{e^{Bt}}{\min_\Omega c_0}, \tag{3.4}$$

with B defined in (2.10) and $\mathbf{m}$ in (3.3).

Proof
From the estimate (2.13) on c, it follows for $m(t)$, (3.1),

$$e^{-Bt} \min_\Omega (c_0) |\Omega^0(t)| \leq m(t; \mathbf{m}_0, 0) \leq e^{Bt} \max_\Omega (c_0) |\Omega^0(t)|.$$

Then (3.4) follows because $m(t; \mathbf{m}_0, 0)$ is the explicit solution of (3.3). □

Proposition 3.1 yields the localization of one component in the following assumption on the matrix Λ:

(H3) The $N \times N$ constant matrix Λ is such that if $y_{i0} = 0$, then $y_i(t; \mathbf{y_0}, 0) = 0$ for some i.

Proposition 3.2 (localization properties)
In assumptions **H1**, **H2** and **H3**, if c_{i0} has compact support, then $c_i(\mathbf{x}, t)$ has compact support for any bounded interval $[0, T]$.

Proof
Let $\tilde{\Omega}_i$ be the support of c_{i0} and B_ρ a ball of radius ρ such that $\tilde{\Omega}_i \subset B_\rho \subset \Omega$.

Then $c_{i0} \equiv 0$ in $\Omega^0 = \Omega \backslash B_\rho$ and $m_{i0} = 0$ in Ω^0.

From (3.2) and **H3**, it follows that $m_i(t) = 0$ in $\Omega^0(t)$, $t \in (0, T)$.

Estimate (3.4) guarantees that the measure of $\Omega^0(t)$ is strictly positive. Moreover, the characteristics starting from the boundary of Ω, where $\nabla c \cdot \mathbf{n} = 0$, stay on the boundary of Ω, and the characteristics starting from the boundary of B_ρ, say S_ρ, stay inside Ω for $t > 0$.

Consequently, from (3.1), it follows that $c_i(\mathbf{x}, t) \equiv 0$ too in $\Omega^0(t)$ for $t > 0$. □

Remark 3.1
The result of localization obtained in the case of the homogeneous Neumann problem is valid also for other problems, for example the Cauchy problem. A further result for the Neumann problem is that the support of c_i stays separated from the boundary of Ω for any $t \in [0, T]$.

Assumption **H3** is justified by the following physically relevant examples.

- **Example 1** Each isotope is either stable or decays out of the element, that is, Λ is a diagonal matrix and the ODE is

$$\dot{c}_i = -\gamma_i c_i, \qquad i = 1, \ldots, N, \ \ \gamma_i \geq 0. \tag{3.5}$$

  This is the case, for example of the stable couple (Cl^{37}, Cl^{35}), that is, $\gamma_i = 0, \ i = 1, 2$, or of the radiative couple (U^{235}, U^{238}) for which $0 < \gamma_1 < \gamma_2$, with the γ_i very close one to the other so that they can be taken equal as a first approximation.

  For such decay law, assumption **H3** and hence a localization property hold for any i. This also means that if two isotopes are initially segregated, they are segregated any time.

  Moreover, if $\gamma_i = \gamma \geq 0, \ i = 1, ..., N$, such as for the two couples quoted in the preceding text, then the equation for c is decoupled from the system for $\mathbf{r}$, because $b \equiv -\gamma$. Therefore, $c = w(\mathbf{x}, t)e^{-\gamma t}$ (2.14) and $m(t) = m(0)e^{-\gamma t}$ (3.3).

  Hence, by the definition of $m(t)$ (3.1), it follows

$$\int_{\Omega^0(t)} w(\mathbf{z}, t) d\mathbf{z} \equiv m(0), \qquad t > 0, \tag{3.6}$$

  and the following more precise estimate on the measure of $\Omega^0(t)$

$$\frac{m(0)}{\max c_0} \leq |\Omega^0(t)| \leq \frac{m(0)}{\min c_0}, \qquad t > 0. \tag{3.7}$$

- **Example 2** A chain of N isotopes in which the i-th one decays into the $(i+1)$-th one, and the N-th one is stable, that is, the decay law is

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1 \\ \dot{c}_i = \gamma_{i-1} c_{i-1} - \gamma_i c_i, \qquad i = 2, \ldots, N-1, \\ \dot{c}_N = \gamma_{N-1} c_{N-1}, \end{cases} \tag{3.8}$$

  with $\gamma_i > 0, \ i = 1, \ldots, N$

  In this case, the localization property holds for c_1, independently of the initial value of the other isotopes while it holds for c_i, $i = 2, \ldots, N-1$, only if $c_{k0} = 0$ for all $k = 1, \ldots, i-1$.

  Again in this example, $b \equiv 0$, that is, c is a solution of the heat equation, so that (3.6) and (3.7) hold.
- **Example 3** A chain in which the i^{th} one decays into the $(i+1)$-th one, with $i = 1, \ldots, N-1$, and the N-th one decays out of the element. This applies to the couple (U^{234}, U^{238}). The decay law is

$$\begin{cases} \dot{c}_1 = -\gamma_1 c_1 \\ \dot{c}_i = \gamma_{i-1} c_{i-1} - \gamma_i c_i, \qquad i = 2, \ldots, N, \end{cases} \tag{3.9}$$

  with $\gamma_i > 0, \ i = 1, \ldots, N$.

  Under such conditions, the total mass decays, and both (3.4) and the localization property for c_1 hold, and the other isotopes have the same behavior as in the previous example.

4. Asymptotic behavior for $t \to \infty$

The asymptotic behavior of the solutions of the problem (2.1) strongly depends on the decay law Λ, as it is reasonable and clear in the one-dimensional case, dealt with in [7,9]. In general, the total concentration c does not converge uniformly, but for the decoupled case, that is, when b is equal to a constant β_0, so that $c(\mathbf{x}, t)e^{-\beta_0 t}$ converges to the mean value of $c_0(\mathbf{x})$, see (2.12). In the general case, a result on the distribution of mass can be proved. Because the purpose of this study is to analyze the properties of isotopes, which either decay or are stable, similarly to the one-dimensional case [7], it is assumed that

(H4) All the eigenvalues of Λ are real and nonpositive, and if the maximum eigenvalue is zero, then it is semisimple.

Because the eigenvalues of Λ and $\tilde{\Lambda}$, defined in Section 2, coincide, hypothesis **H4** can be extended to $\tilde{\Lambda}$ directly.
From the ODE theory (e.g., [16,17]), we have a complete description of the solutions of the ODE.
Let $\tilde{\Lambda}$ have $s \leq N$ distinct eigenvalues $\lambda_s < \ldots < \lambda_1 \leq 0$; for each λ_i, $i = 1, \ldots, s$, the following variables are defined:
$\alpha(\lambda_i)$ = algebraic multiplicity of λ_i,
$\nu(\lambda_i)$ = geometric multiplicity of λ_i,
$E(\lambda_i)$ = generalized autospace of λ_i, and
$h(\lambda_i)$ = the least integer k such that Ker $(\tilde{\Lambda} - \lambda_i I)^{k+1}$ = Ker $(\tilde{\Lambda} - \lambda_i I)^k$,
so that $E(\lambda_i)$ = Ker $(\tilde{\Lambda} - \lambda_i I)^{h(\lambda_i)}$, with $I = Id$ matrix $N \times N$.
Any solution is a linear combination of the product of exponential functions time polynomials ones. More specifically,

$$\mathbf{y}(t) = \sum_{i=1}^{s} \left[\sum_{k=0}^{h(\lambda_i)-1} (\tilde{\Lambda} - \lambda_i I)^k \frac{t^k}{k!} \right] e^{\lambda_i t} \mathbf{y}_{0,i}, \tag{4.1}$$

with $\mathbf{y}_0 = \sum_{i=1}^{s} \mathbf{y}_{0,i},\ \mathbf{y}_{0,i} \in E(\lambda_i)$.

Assumption **H4** implies that the leading term in (4.1) as $t \to +\infty$ is of the order $t^{h(\lambda_1)-1}e^{\lambda_1 t}$:

$$\lim_{t\to+\infty} t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}\mathbf{y}(t;\mathbf{y}_0) = \frac{1}{(h(\lambda_1)-1)!}(\tilde{\Lambda}-\lambda_1 I)^{h(\lambda_1)-1}\mathbf{y}_{01} = \hat{B}\mathbf{y}_0. \tag{4.2}$$

Because Λ is a constant matrix, $\hat{B}$ is a constant $N \times N$ matrix, determined by the autospaces $E(\lambda_i)$, $i = 1, \ldots, s$.

Let the vector $\mathbf{F}$ be defined as

$$\mathbf{F}(\mathbf{x}) = (F_1, \ldots, F_{N-1}, F) = \hat{B}\tilde{\mathbf{C}}_0(\mathbf{x}), \qquad F(\mathbf{x}) = (\hat{B}\tilde{\mathbf{C}}_0(\mathbf{x}))_N, \tag{4.3}$$

and for a given index i, say $i = 1$, let the single masses 'up to x_1' be

$$\tilde{m}_i(x_1,t) = \int_{\Omega(x_1)} c_i(\mathbf{z},t)\,d\mathbf{z}, \quad i = 1,\ldots,N, \qquad \tilde{m}(x_1,t) = \int_{\Omega(x_1)} c(\mathbf{z},t)\,d\mathbf{z}, \tag{4.4}$$

where $\Omega(x_1) = \Omega \cap \{\mathbf{z} \in \mathbb{R}^n : z_1 < x_1\}$. Let Ω belong to the strip $\{\mathbf{z} \in \mathbb{R}^n : 0 \le z_1 \le L_1\}$, L_1 being the diameter of Ω in direction x_1. Then the following result about the asymptotic behavior of $\tilde{m}_i(x_1,t)$ holds:

Theorem 4.1

In assumptions **H1**, **H2**, and **H4**, and for any initial data $\tilde{\mathbf{C}}_0(\mathbf{x})$ such that $F(\mathbf{x}) \ge \delta > 0$ in $\overline{\Omega}$, F defined in (4.3), it follows

$$\lim_{t\to+\infty} t^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}\tilde{m}(x_1,t) = \frac{x_1}{L_1}\int_\Omega F(\mathbf{x})\,d\mathbf{x}, \tag{4.5}$$

uniformly in $[0, L_1]$.

Proof

The function $\tilde{m}(x_1,t)$ a is solution of the problem

$$\begin{cases} \tilde{m}_t = \tilde{m}_{x_1x_1} + \tilde{f} & \text{in } Q_T^1 = (0,L_1)\times(0,T), \\ \tilde{m}(0,t) = 0, & t > 0 \\ \tilde{m}(L_1,t) = M(t), & t > 0 \\ \tilde{m}(x_1,t) = \int_{\Omega(x_1)} c_0(\mathbf{z})\,d\mathbf{z} = \tilde{m}_0(x_1), & \text{in } [0,L_1] \end{cases} \tag{4.6}$$

where

$$\tilde{f}(x_1,t) = \int_{\Omega(x_1)} c(\mathbf{z},t)\,b(\mathbf{r}(\mathbf{z},t))\,d\mathbf{z}, \tag{4.7}$$

and $M(t)$ is the total mass in Ω at time t, which is explicitly known. In fact, because of the homogeneous boundary condition on Γ_T, the characteristics starting for $t = 0$ from $\partial\Omega$ cannot enter the domain Ω so that the flux tube starting from the whole Ω coincides with Ω for any positive time, and the arguments of Section 3 can be repeated with Ω instead of $\Omega_0(t)$ in (3.1), (3.2), and (3.3).

Therefore,

$$\dot{\tilde{\mathbf{y}}} = \tilde{\Lambda}\tilde{\mathbf{y}}, \quad \tilde{\mathbf{y}}(0) = \int_\Omega \tilde{\mathbf{C}}_0(\mathbf{x})\,d\mathbf{x}, \quad M(t) = \tilde{y}_N(t). \tag{4.8}$$

In view of the asymptotic behavior of the solution of the previous ODE, the function $\tilde{m}$ can be rescaled as

$$v(x_1,t) = (1+t)^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}\tilde{m}(x_1,t), \tag{4.9}$$

so that v is a solution of the one-dimensional problem

$$\begin{cases} v_t = v_{x_1x_1} + f & \text{in } Q_T^1 = (0,L_1)\times(0,T), \\ v(0,t) = 0, & t > 0 \\ v(L_1,t) = (1+t)^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}M(t), & t > 0 \\ v(x_1,t) = \tilde{m}_0(x_1), & \text{in } [0,L_1] \end{cases} \tag{4.10}$$

with

$$f = \int_{\Omega(x_1)} \hat{b}\,u\,d\mathbf{z},\ \ \hat{b} = b - \lambda_1 - \frac{h(\lambda_1)-1}{1+t}, \qquad u(\mathbf{z},t) = (1+t)^{-(h(\lambda_1)-1)}e^{-\lambda_1 t}c(\mathbf{z},t). \tag{4.11}$$

Because $M(t)$ is given by (4.8), by assumption **H4**, its asymptotic behavior is given by (4.2). Hence, by the definition (4.3) of F, it is

$$\lim_{t\to+\infty} v(L_1,t) = \int_\Omega F(\mathbf{x})\,d\mathbf{x}. \tag{4.12}$$

Therefore Theorem 4.1 is proved by means of classical results (e.g., [18], cap.VI, Thm.1) provided that $f(x_1, t)$ tends to 0 uniformly in $[0, L_1]$ for $t \to +\infty$. This comes from the two uniform estimates $\forall \mathbf{x} \in \Omega$, $t \geq 0$:

$$|\hat{b}| \leq k(\delta)g(t), \ \ g = \frac{h(\lambda_1) - 1}{(1+t)^2} + (s-1)e^{\frac{\lambda_2 - \lambda_1}{2}t}, \tag{4.13}$$

$$\min_{\Omega} c_0 e^{-k(\delta)\sigma(t)} \leq u \leq \max_{\Omega} c_0 e^{k(\delta)\sigma(t)}, \qquad \sigma = \int_0^t g(\tau)\,d\tau, \tag{4.14}$$

with $k(\delta)$ a positive constant depending on c_0, and Λ, and δ the inferior of $F(\mathbf{x})$.

The estimate (4.13) is proved in [7], Lemma 3.1 for $n = 1$, and the proof is the same for $n > 1$ because of the assumption $F(\mathbf{x}) \geq \delta > 0$.

The estimate (4.14) is a consequence of the fact that u is the solution of

$$\begin{cases} u_t = \Delta u + \hat{b}u & \text{in } Q_T, \\ \nabla u \cdot \mathbf{n} = 0, & \text{in } \Gamma_T, \\ u(\mathbf{x}, 0) = c_0(\mathbf{x}), & \text{in } \Omega, \end{cases} \tag{4.15}$$

and of the estimate on $\hat{b}$. Because $0 \leq \sigma(t) \leq k_1$ for $t \geq 0$, where k_1 is a constant depending only on Λ, (4.14) give a uniform bound on u for any $t \geq 0$.

Therefore, from the estimates (4.13) and (4.14) and the definition of f in (4.11), it follows

$$|f(x_1, t)| \leq k_2(\delta)g(t), \qquad \forall x_1 \in [0, L_1], \ t \geq 0,$$

with k_2 a positive constant depending on $k(\delta)$, k_1, $\max_\Omega c_0$, and $|\Omega|$. Finally, $f \to 0$ as $t \to \infty$ uniformly in $[0, L_1]$, which concludes the proof of Theorem 4.1. □

From estimates (4.13) and (4.14), it is also possible to derive bounds on $|\Omega_0(t)|$, as defined in Proposition 3.1, valid also asymptotically, specifically the following:

Proposition 4.1
Under the conditions of Theorem 4.1, and if $F(\mathbf{x}) \geq \delta$, F defined by (4.3), then

$$\omega_1(t) \leq |\Omega_0(t)| \leq \omega_2(t), \qquad t \geq 0, \tag{4.16}$$

where

$$\lim_{t \to +\infty} \omega_1(t) = \frac{1}{\max c_0} e^{-\bar{k}} \int_{\Omega_0} F(\mathbf{x})\,d\mathbf{x} > 0,$$
$$\lim_{t \to +\infty} \omega_2(t) = \frac{1}{\min c_0} e^{\bar{k}} \int_{\Omega_0} F(\mathbf{x})\,d\mathbf{x} > 0,$$

$\bar{k} = k(\delta) \int_0^{+\infty} g(s)\,ds$, g given in (4.13).

Proof
In the same framework of Proposition 3.1, let the function μ be defined as

$$\mu(t) = \int_{\Omega_0(t)} u(\mathbf{x}, t)\,d\mathbf{x} = (1+t)^{-(h(\lambda_1)-1)} e^{-\lambda_1 t} \int_{\Omega_0(t)} c(\mathbf{z}, t)\,d\mathbf{z},$$

so that

$$\lim_{t \to +\infty} \mu(t) = \int_{\Omega_0} F(\mathbf{x})\,d\mathbf{x}.$$

From the estimate (4.14) on u, it follows

$$\frac{1}{\max c_0} e^{-k(\delta)\sigma(t)} \mu(t) \leq |\Omega_0(t)| \leq \frac{1}{\min c_0} e^{k(\delta)\sigma(t)} \mu(t),$$

and being $g(t)$ positive and integrable at $+\infty$,

$$\sigma(t) = \int_0^t g(s)ds < \int_0^{+\infty} g(s)ds$$

□

In the decoupled case, that is, $b = \beta_0$, a more accurate behavior for $|\Omega_0(t)|$ can be proved:

Proposition 4.2
In the assumptions of Proposition 4.1 and if $b = \beta_0$, then

$$\lim_{t\to+\infty} |\Omega_0(t)| = \frac{1}{\bar{c}_0}\int_{\Omega_0} c_0(\mathbf{x})\,d\mathbf{x}, \tag{4.17}$$

where $\bar{c}_0$ is the mean value of c_0 on the whole Ω, that is,

$$\bar{c}_0 = \frac{1}{|\Omega|}\int_{\Omega} c_0(\mathbf{x})\,d\mathbf{x}. \tag{4.18}$$

Proof
Under these conditions, the ODE (3.3) holds, and

$$\int_{\Omega_0(t)} c(\mathbf{x},t)\,d\mathbf{x} = m(t) = m(0)e^{\beta_0 t}, \quad m(0) = \int_{\Omega_0} c_0(\mathbf{x})\,d\mathbf{x}.$$

The Proposition is then demonstrated by the mean value theorem, the fact that $c = e^{\beta_0 t}w$ and w, solution of (2.12), converges uniformly to $\bar{c}_0$ as $t \to +\infty$. □

Clearly, Propositions 4.1 and 4.2 imply the asymptotic localization property (Proposition 3.2).

As mentioned in the preceding text, the behavior of $\mathbf{r} = (r_1, r_2, \ldots, r_{N-1})$ depends only on the decay law Λ on each characteristic $\mathbf{X}(t;\mathbf{x}_0, 0)$ starting from $\mathbf{x}_0$ at time zero.

From the explicit expressions (4.1), (4.2), and (4.3), it is derived on $\mathbf{X}(t;\mathbf{x}_0, 0)$,:

$$r_i = \frac{F_i(\mathbf{x}_0) + H_i e^{-\lambda_1 t}(1+t)^{-(h(\lambda_1)-1)}}{F(\mathbf{x}_0) + H_N e^{-\lambda_1 t}(1+t)^{-(h(\lambda_1)-1)}} = R_i(\mathbf{x}_0, t), \quad i = 1, \ldots, N-1, \tag{4.19}$$

where, setting $\mathbf{H}(\mathbf{x_0}) = (H_1(\mathbf{x_0}), \ldots, H_N(\mathbf{x_0}))$,

$$||\mathbf{H}|| \le k_1 \left(\frac{h(\lambda_1)-1}{1+t} + (s-1)e^{\frac{\lambda_2-\lambda_1}{2}t}\right)), \ \forall \mathbf{x} \in \Omega, \ t > 0$$

and k_1 is a positive constant depending on Λ and on $\max_\Omega ||\mathbf{c}_0||$.

It is quite clear that the asymptotic behavior of the set of isotopes changes depending on the decay law; precisely ,it depends on

$$\lim_{t\to+\infty} R_i(\mathbf{x}_0, t) = \frac{F_i(\mathbf{x}_0)}{F(\mathbf{x}_0)}, \tag{4.20}$$

which is uniform in $\mathbf{x}_0$.

Namely, if the functions $\frac{F_i}{F}$ are constant, then $\mathbf{r}$ has a uniform limit, that is, a 'parabolic asymptotic behavior', while if $\frac{F_i}{F}$ varies with $\mathbf{x}_0$, the asymptotic of $\mathbf{r}$ strongly depends on the initial spatial distribution of the isotopes, and it might show strong oscillations also for large times, a 'hyperbolic asymptotic behavior'.

In the Examples of Section 3, $\frac{F_i}{F}$ is constant with respect to $\mathbf{x_0}$ for Examples 2 and 3, while for Example 1, this is true if and only if λ_1 is semisimple. More specifically,

- **Example 2** The maximum eigenvalue is $\lambda_1 = 0$ and $\mathbf{F}(\mathbf{x}_0) = (0, \ldots, 0, c_0(\mathbf{x}))$. Then Theorem 4.1 is valid for each data satisfying **H1**, that is, **H4** always holds because $F(\mathbf{x}) = c_0(\mathbf{x}) \ge k_0 > 0$.

  Hence, $r_i \to 0$, $i = 1, \ldots N-1$, as $t \to +\infty$, and only the N-th isotope, the stable one, remains asymptotically.
- **Example 3** A detailed analysis can be found in [7], Section 4. The most interesting case from a physical point of view is when there is the so-called 'secular equilibrium' of all the set of isotopes, that is, each isotope appears in a given positive percentage of the whole element. Such is the case of the already mentioned couple (U^{234}, U^{238}) as of many others.

  From the decay law (3.9), it follows that secular equilibrium is possible if and only if all the eigenvalues $\lambda_i = -\gamma_i$ are distinct (i.e., $s = N$) and $\gamma_1 = \min_{i=1,\ldots,N} \gamma_i$.

  In this assumption, by denoting $\mathbf{v}^1$ the eigenvector corresponding to $\lambda_1 = -\gamma_1$, it follows

$$\mathbf{F} = c_{10}(\mathbf{x})\frac{\mathbf{v}^1}{v^{1,1}}, \quad F(\mathbf{x}) = c_{10}(\mathbf{x})\frac{v^{1,N}}{v^{1,1}}, \quad \frac{F_i}{F} = \frac{v^{1,i}}{v^{1,N}}, \quad i = 1, \ldots, N-1,$$
$$v^{i,1} = \prod_{j=1}^{N-1} \frac{\gamma_{j+1} - \gamma_1}{\gamma_j}, \qquad i = 1, \ldots, N-1,$$
$$v^{1,N} = 1 + \sum_{i=1}^{N-1} \prod_{j=1}^{N-1} \frac{\gamma_{j+1} - \gamma_1}{\gamma_j}.$$

Assumption **H4**, that is, $c_{10}(\mathbf{x}) \geq \delta > 0$, guarantees that the isotope 1 is initially present, that is, the start of the chain; otherwise, the time behavior of the solution is different. Still in Theorem 4.1, the $\int_\Omega F(\mathbf{x})d\mathbf{x}$ depends on the initial mass of the isotope 1 in the whole domain Ω.

- **Example 1** Here, Λ is a diagonal $N \times N$ matrix, and without a loss of generality, the diagonal elements $-\gamma_i$ can be ordered setting $-\lambda_1 = \gamma_1 \leq \gamma_2 \ldots \leq \gamma_N$, in this case, $h(\lambda_1) = 1$.

  If $\alpha(\lambda_1) = 1$, then $\mathbf{F} = c_{10}(\mathbf{x})\mathbf{e}^1$, $e^{1j} = \delta_{1j}$ so that the assumption **H4** means $c_{10}(\mathbf{x}) \geq \delta$; that is, there is initially isotope 1, and asymptotically, only this isotope is present with a total mass equal to the initial mass of c_1 in the whole Ω, in fact $\int_\Omega F = \int_\Omega c_{10}(\mathbf{x})d\mathbf{x}$ and $\frac{F_i}{F} \equiv \delta_{1i}$.

  If $1 < \alpha(\lambda_1) \leq N$, then the asymptotic distribution of $\mathbf{r}$ strongly depends on the initial data, precisely:

$$\frac{F_i}{F} = \frac{c_{i0}(\mathbf{x}_0)}{\sum_{j=1}^{\mu(\lambda_1)} c_{j0}(\mathbf{x}_0)}.$$

  In particular, for the two couples mentioned in Section 3 , $\alpha(\lambda_1) = N$ and $F(\mathbf{x}) = c_0(\mathbf{x})$, so that **H4** holds for any positive c_0 and there is localization up to large time of each isotope and possibly strong oscillations too (i.e., true hyperbolic asymptotic behavior).

Remark 4.1
The assumption **H3** that all the eigenvalues of Λ are nonpositive has been considered in order to describe the physical problem of decaying or stable isotopes, but the results of Section 4 hold also without any restriction on the sign of λ_i, with λ_1 the maximum eigenvalue.

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