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On tracer theory in geophysical systems in the steady and non-steady state
Part II. Non-steady state—theoretical introduction

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ABSTRACT

In continuation to Part I,[1] which dealt with the steady state, tracer theory is extended to time-varying systems. Most geophysical systems are recognized as time-varying when observed for a prolonged time and in sufficient detail. We distinguish between explicit and implicit system variability. The unique composition system response of the steady-state system, $h^c(\tau)$, is replaced by three time-dependent system functions which relate to input and output conditions. Time relations of the steady state are also generalized and illustrated heuristically by analogies to biological populations. Input and output average transit times are defined, as well as their ensemble or time averages. The distributions over transit time and age in the flux and volume are generalized to the non-steady state. Examples of the different categories of non-steady state in natural systems are indicated.

1. Introduction

In continuation of Part I of this article we try to formulate the tracer theory in terms of non-steady-state systems; to develop some areas of this theory; to demonstrate its application on specific examples in several areas of geoscience. We attempt, as before, to do this in a unified and consistent form, and will refer therefore to Part I as a basic form of presentation, which will be expanded as required. As this task may result often in undue formalism, which is not our objective, we will try to indicate conceptual and geophysical background, as space permits.

Negative definition of the subject (non-steady state) makes it practically intractable in all its generality, except for statements about non-validity of most theorems of steady-state theory.

In order to avert such a situation, we treat the subject by classifying the systems into various classes of variability such as: internal or external parameter variability, piecewise steady system, linear time-varying, stationary stochastic systems, etc. Here we also try to associate the mathematical form of presentation with physical structure.

The difficulty is enhanced by the fluid state and fast rate of development in the mathematical theory and numerical methods dealing with non-steady-state systems. Undoubtedly a more satisfactory presentation could be written in a few years' time. We believe, however, that the user of tracers has an immediate need for some theoretical tools and overview of the methods. Conversely, the knowledge of the unsatisfactory status of this area may induce more vigorous efforts by the system theorist.

2. List of symbols and definitions

Definitions of Part I are used whenever possible and are not repeated here. Additional ones are:

$(\,)'$—primed quantities, e.g. $(q(t_i))'$, are deviations from time average. However, primed differentials in integration formulae refer to dummy variables, e.g. τ', $d\tau'$.

[1] Reference to Part I of this paper (Nir and Lewis, *Tellus 27*, 372–383, 1975) will be indicated by I followed by the section, equation or figure number, e.g., I,2.1.2.

$q^{(k)}(t)$—kth derivative of $q(t)$.
$h^c(\tau;t_i)$—time-dependent composition response function.
$h_r^c(\tau;t_o)$—time-dependent system response function.
$h_{wi}^c(\tau;t_0)$—time-dependent input weighting function.
$h_{wo}^c(\tau;t_o)$—time-dependent output weighting function.
$\bar{F}_r^c(T;t_o-T) = q_i(t_o-T) - F_r^c(T;t_o-T)$

3. Definitions and concepts for time-varying systems

3.1. *Formal definitions*

A system is defined as time-varying if the relationship between its input and output is time-dependent (De Russo et al., 1965). Such a system is explicitly time-varying but linear if the governing parameters vary independently of the input and system variables. If parameter variation is a function of input or system variables, the system is non-linear and, hence, implicitly time-varying. In a steady state, i.e. when input and state variables have constant values, a tracer experiment will not discern this type of implicit time variability; see also Section 3.4 and Jaquez (1972) and DiStefano (1976).

Essentially all environmental systems may be shown to be time-varying when observed for a prolonged time and in sufficient detail. Real systems are not envisaged as persisting from infinite past to infinite future. Therefore considerations of time scale and precision of observation will determine whether or not the system under consideration will be described as time-varying.

3.2. *Variable parameter systems in the natural environment*

Several examples of natural processes and systems characterized by time-varying behaviour will be mentioned briefly.

(a) *Geology*. Processes comprising the geomorphological evolution of a region, e.g. tectonics, climate, erosion and sedimentation, introduce time variability in structural parameters over long time scales. The climatic factors result from periodic and stochastic processes with periods ranging from diurnal and annual to thousands and millions of years (Yevdjevich, 1971). Examples of chemical and physical processes altering the structure of natural systems within relatively short time spans include hysteresis in soil permeability as a function of water content, chemical reactions between solute and soil matrix and transport through compacting sediments.

(b) *Hydrology*. Most processes involved in mass and energy transfer in the hydrological cycle are implicitly or explicitly time-dependent. Yevdjevich (1971) discussed the periodic-stochastic nature of these processes as derived from astronomical cycles and the many sources of stochasticity and attenuation in the environment. The basic storage equation for a natural lake (Jeng and Yevdjevich, 1966) is a clear example of time-dependence in the equation governing a system:

$$\frac{dV(t)}{dt} = q_i(t) - q_o(t) = q_i(t) - k(t)V(t)^n \qquad (1a)$$

or

$$\frac{dV(t)}{dt} + k(t)V(t)^n = q_i(t) \qquad (1b)$$

where $V(t)$, the lake volume, is the system variable. The exponent n comprises the ratio of exponents of the outflow rating curve and the lake morphology curve. The coefficient $k(t)$ comprises both these exponents and the lake volume and outflow. Although n commonly varies between 0.25 and 4.0, introducing nonlinearity, it is often given the value of 1.0, defining a linear reservoir. However, in this case, explicit time-dependence as retained through $k(t)$ may be due to evaporation and water level manipulation by pumping or damming. The effect of non-steady hydrological relations on the equations of tracer concentrations in mixed lakes has been discussed by Nir (1973).

The transport and mixing patterns in a lake may be influenced implicitly by seasonal changes in inflow rates, by relative changes in the contributions of various components, e.g. streamflow, groundwater seepage, direct precipitation and evaporation. Explicit changes may be exemplified by stratification changes caused by atmospheric heat input or by wind stress, including winter turnover in monomictic lakes and, of course, man-made changes.

These selected examples serve to emphasize the need to deal with time-varying systems and to be able to incorporate the tracer theory in elucidating their structure.

3.3. *Time-dependent system response functions*

As discussed in I,2.1.2, the transit time distribution of molecules in a system can be viewed either as a system response or as a weighting function. In analogy to the steady state we find the system response by application of a unit impulse at a given time, t_i. The observation of the fraction of this input leaving the system at times $t > t_i$ will provide the system response function. The value of this function will now depend both on the input time t_i and the transit time, $\tau = t - t_i$, while in the steady state it depended on τ only. On the other hand, the weighting function will be obtained by observing the contribution of past inputs to present output, at time t_o. This will be discussed in more detail in Section 3.4. Both functions were shown to be equal to $h(\tau)$ under steady-state conditions.

Fig. 1 illustrates these time relations for a particular molecule which entered the system at

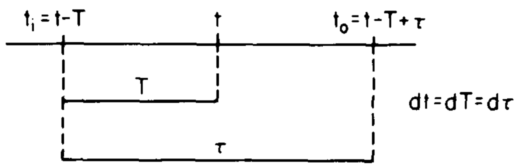

Fig. 1. Time relations.

time t_i, has the "age" T at the instant of observation t, and will leave the system at $T_o = t - T + \tau$, after a transit time τ.

In analogy to I(1b) we can write for composition response of a time-varying linear system

$$q_o(t) = \int_0^\infty q_i(t - \tau) h^c(\tau; t)\, d\tau \tag{2}$$

Here $h^c(\tau; t)$ is a generalization of $h^c(\tau)$, being dependent also on the parameter t. The time parameter may be assigned one of several meanings: the input time, t_i, output time, t_o, or running time, t. We will indicate in each case the nature of the parameter which will be selected to suit the circumstances.

We will observe the time-dependent system response by measuring the output $q_o(t)$ after application of a unit impulse at time t_i, i.e. $q_i(t) = \delta(t - t_i)$. We obtain, similarly to I(2)

$$q_o(t) = \int_0^\infty \delta(t - t_i - \tau)\, h^c(\tau; t_i)\, d\tau$$
$$= h^c(t - t_i; t_i) \tag{3a}$$

Substituting $t = t_i + \tau$

$$q_o(t_i + \tau) = h^c(\tau; t_i) \tag{3b}$$

In this representation the input time is emphasized as a parameter of $h^c(\tau; t_i)$ to indicate our interest. It is, however, a convention and other parameters may be used if preferred.

Conservation of matter requires that the cumulative output equals the input. We are therefore justified to integrate the output from t_i to infinity and set the integral equal to the input, i.e. 1.

$$\int_0^\infty q_o(t_i + \tau)\, d\tau = \int_0^\infty h^c(\tau; t_i)\, d\tau = 1. \tag{4}$$

In analogy to the steady state, the normalization of the system response function holds also in the non-steady state.

In cases in which ambiguity may arise we will assign to the system response function a subscript r, i.e. $h_r^c(\tau; t_i)$.

In Fig. 2a, the position of some discrete values of $h_r^c(\tau; t_i)$ is shown in the plane determined by t and τ coordinates. Fig. 2b shows these values in three-dimensional representation, for an arbitrary response function.

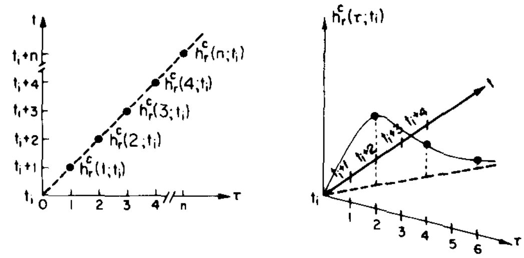

Fig. 2. (a) System response function in the t–τ plane. (b) System response function in three dimensions.

Fig. 3a and 3b show succeeding system response functions for inputs initiated at times later than t_i. As seen in Fig. 3b, the response functions may differ in their shape, and are not necessarily parallel displacements of $h_r^c(\tau; t_i)$ as would have been the case for a steady-state system. The discrete values of the functions shown in Fig. 3b are tabulated in Table 1.

3.4. *Time-dependent weighting functions*

An alternative interpretation of the transit time distribution is derived from (2), by regarding it as the input weighting function at a constant output

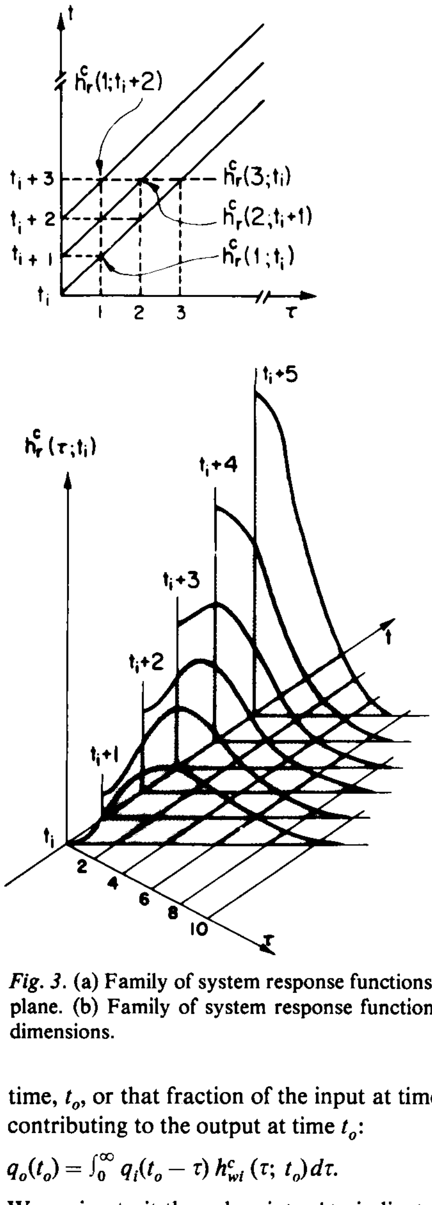

Fig. 3. (a) Family of system response functions in the t–τ plane. (b) Family of system response functions in three dimensions.

time, t_o, or that fraction of the input at time $(t_o - \tau)$ contributing to the output at time t_o:

$$q_o(t_o) = \int_0^\infty q_i(t_o - \tau)\, h_{wi}^c(\tau;\, t_o) d\tau. \tag{5}$$

We assign to it the subscript *wi* to indicate that the weighting refers to the input at time $t_o - \tau$.

We can rewrite it in terms of system response, noting that $h_{wi}^c(\tau;t_o)$ has the same physical interpretation as $h_r^c(\tau;(t_o - \tau)_i)$.

$$q_o(t_o) = \int_0^\infty q_i(t_o - \tau)\, h_r^c(\tau;(t_o - \tau)_i)\, d\tau. \tag{6}$$

Normalization of $h_r^c(\tau;(t_o - \tau)_i)$ is not ensured, as was the case before, as the fractions given by $h_r^c(\tau;(t_o - \tau)_i)$ refer here to different inputs. Subsequent examples will emphasize this point.

In Table 1a and Fig. 4 we observe that $h_{wi}^c(\tau;t_o)$ is situated along the τ coordinate, while $h_r^c(\tau;t_i)$ was on the diagonal (Fig. 2).

A third representation of $h^c(\tau;t)$ is provided by considering the fraction of the output flow $q_o(t_o)$ which is contributed by the input at time $t_o - \tau$. We designate this, the output weighting function, by $h_{wo}^c(\tau;t_o)$. This function is normalized by its definition as a fraction of the output at a given t_o

$$\int_0^\infty h_{wo}^c(\tau;t_o)\, d\tau = 1. \tag{7}$$

On grounds of physical identity we require the fraction of input $q_i(t_o - \tau)$ with transit time τ, to equal that fraction of output, $q_o(t_o)$, which entered at $t_o - \tau$. Using the definition of $h_{wo}^c(\tau;t_o)$:

$$q_i(t_o - \tau)\, h_{wi}^c(\tau;t_o) = q_o(t_o)\, h_{wo}^c(\tau;t_o) \tag{8}$$

Therefore

$$h_{wo}^c(\tau;\, t_o) = \frac{q_i(t_o - \tau)}{q_o(t_o)}\, h_{wi}^c(\tau;\, t_o) \tag{9a}$$

At steady state $q_i(t_o - \tau) = q_o(t_o) = q$; therefore $h_{wo}^c(\tau;t_o) = h_{wi}^c(\tau;t_o)$, and according to I(2) both are equal to $h^c(\tau)$. This formulation will be of use subsequently for the description of tracer behaviour in non-steady-state systems. In the discrete form (9a) becomes

$$h_{wo}^c(\tau;\, t_o) = \frac{q_i(t_o - \tau) h_{wi}^c(\tau;\, t_o)}{\sum_\tau q_i(t_o - \tau) h_{wi}^c(\tau;\, t_o)} \tag{9b}$$

Fig. 4 and the associated Table 1b compare the input and output weighting functions for time t_o with an arbitrary inflow function. Table 1 uses discrete values for simplicity of presentation.

In the probabilistic interpretation (Krambeck et al., 1969) $q_i(t_o - \tau)$ is proportional to the probability of entry of a random particle into the system at time $t_o - \tau$, while $h_{wi}^c(\tau;t_o)$ is the conditional probability of exit at t_o, provided it entered at $t_o - \tau$. $h_{wo}^c(\tau;t_o)$ has then the structure, and possible interpretation, of Bayes' probability of cause (Feller, 1957), and represents the conditional probability that a particle at the output at t_o originated at the input at $t_o - \tau$.

3.5. *Tracer response in non-steady-state formulation*

In Section 3.2 on the basis of physical identity we set the fraction of bulk flow input, $q_i(t_o - \tau)$,

Table 1a. *Discrete system response function $h^c_r(\tau;\, t_i)$ of Fig. 3b*

$h^c_r(\tau;\, t_k + 5)$

t \ τ	0	1	2	3	4	5	6	7	8	9	10	
											.02	
					.02	.05	.05	.03	.04	.05		
·				.06	.08	.07	.06	.07	.07			
·			.12	.12	.10	.10	.09	.10				
·		.30	.18	.15	.13	.13	.12					
$t_k + 5$	.50	.27	.20	.18	.16	.14						← $h^c_{wi}(\tau;\, t_k + 5)$
$t_k + 4$	.30	.23	.20	.18	.16							
$t_k + 3$	.20	.18	.17	.14								
$t_k + 2$	.17	.10	.12									
$t_k + 1$	.06	.07										
t_k	.01											

Table 1b

τ	$q_i(t_o - \tau)$	$h^c_{wi}(\tau;\, t_i + 5)$	$h^c_{wo}(\tau;\, t_i + 5)$
0	5	.50	.32
1	3	.27	.10
2	9	.20	.23
3	10	.18	.23
4	4	.16	.08
5	2	.14	.04
		$\sum_{\tau=0}^{5} = 1.45$	1.00

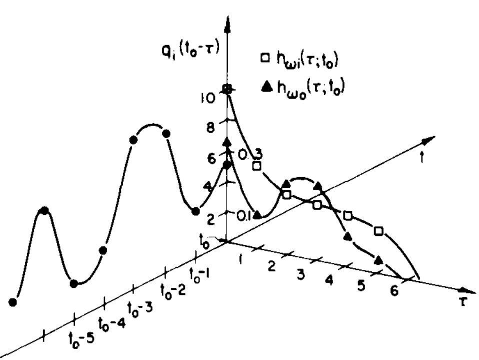

Fig. 4. Weighting functions situated on the τ coordinate.

with transit time τ equal to the fraction of bulk flow output, $q_o(t_o)$, which entered at $t_o - \tau$. Using the definitions of $h^c_{wi}(\tau;t_o)$ and $h^c_{wo}(\tau;t_o)$ we obtained eq. (8)

$$q_i(t_o - \tau)\, h^c_{wi}(\tau;t_o) = q_0(t_o)\, h^c_{wo}(\tau;t_o)$$

We want now to set up an analogous relation for the response of a system to an ideal tracer transported by the main component.

As discussed in Part I, our basic assumption has been that the composition response of the tracer is identical to that of the main component. The tracer quantity input at time $t_o - \tau$ is given by

$$q^T_i(t_o - \tau) = c_i(t_o - \tau)\, q_i(t_o - \tau) \qquad (10)$$

where $c(t)$ is tracer concentration. By the above assumption the fraction of tracer quantity input at $t_o - \tau$ with transit time τ is $q^T_i(t_o - \tau)\, h^c_{wi}(\tau;t_o)$. In other words, the fraction of tracer quantity input at $t_o - \tau$ contributing to the tracer quantity output at t_o is the same as that for the bulk flow.

However, the fraction of tracer quantity appearing in the output at t_o which derived from the tracer quantity input at $t_o - \tau$ need not be identical to the fraction of the bulk flow input. This is so since the tracer quantity and the bulk flow are generally not correlated, i.e. the tracer input concentration may not be constant.

Typical examples of such situations are: (a) inflow composed of several components with time-varying flow rate, each having different tracer concentration, (b) the causes of tracer variability being different from those of main component variability, such as tritium influx to atmospheric moisture, which originated in atmospheric nuclear explosions, and subsequent rainfall. Examples of the nature of such correlations are given by Gibbs and Slinn (1973).

In analogy to (8), we designate $h^T_{wo}(\tau;t_0)$ as the fraction of tracer quantity in the output at time t_o, $q^T_o(t_o)$, which originates from the tracer quantity which entered at time $t_o - \tau$, namely $q^T_i(t_o - \tau)$. In order to evaluate $h^T_{wo}(\tau;t_o)$ we write the tracer analogue to eq. (8):

$$q^T_o(t_o)\, h^T_{wo}(\tau;t_o) = q^T_i(t_o - \tau)\, h^c_{wi}(\tau;t_o) \tag{11}$$

Hence, using (10) and (9a)

$$h^T_{wo}(\tau;t_o) = \frac{q^T_i(t_o - \tau)}{q^T_o(t_o)}\, h^c_{wi}(\tau;t_o) = \frac{c_i(t_o - \tau)}{c_o(t_o)}\, h^c_{wo}(\tau;t_o) \tag{12}$$

With $h^T_{wo}(\tau;t_o)$ related to $h^c_{wo}(\tau;t_o)$ by eq. (12), it may be considered a higher order weighting function in the hierarchy of response functions.

Integrating eq. (11) over τ we obtain

$$q^T_o(t_o) \int_0^\infty h^T_{wo}(\tau;t_o)d\tau = \int_0^\infty q^T_i(t_o - \tau)\, h^c_{wi}(\tau;t_o)\, d\tau \tag{13}$$

Normalization of $h^T_{wo}(\tau;t_o)$ is guaranteed by its definition as an output weighting function. Hence, recalling that according to (10) $q^T_o(t) = c_o(t)q_o(t)$

$$c_o(t_o) = \int_0^\infty c_i(t_o - \tau)\, \frac{q_i(t_o - \tau)}{q_o(t_o)}\, h^c_{wi}(\tau;t_o)\, d\tau \tag{14}$$

or,

$$c_o(t_o) = \int_0^\infty c_i(t_o - \tau)\, h^c_{wo}(\tau;t_o)\, d\tau \tag{15}$$

Equation (15) demonstrates that the tracer concentration input weighting function is identical to the bulk flow output weighting function which is normalized by definition. We should note the apparent similarity to eq. (5) where the weighting function is, however, not normalized.

It is instructive to examine eq. (14) as a guide to possible time-dependence of the tracer concentration weighting function.

In the case of a system characterized by a time-independent input weighting function, we may write eq. (14) using $h^c_{wi}(\tau)$:

$$c_o(t_o) = \int_0^\infty c_i(t_o - \tau)\, \frac{q_i(t_o - \tau)}{q_o(t_o)}\, h^c_{wi}(\tau)\, d\tau \tag{16}$$

Note, however, that the tracer concentration weighting function retains time-dependence if the ratio $q_i(t_o - \tau)/q_o(t_o)$ is time-varying.

Alternatively, in cases in which $q_i(t) = q_o(t) = q$, then $h^c_{wi}(\tau;t_o)$ and $h^c_{wo}(\tau;t_o)$ may nevertheless have an explicit time-dependence due to system parameter variability. In many cases of practical interest $h^c_{wi}(\tau;t_o)$ depends on the history of past inflows (Chiu and Bittler, 1969; Nir, 1973) and therefore both terms contribute implicitly to time-dependence.

Table 2 summarizes representative types of time-dependent behaviour according to the definitions of Section 1.1. As discussed earlier, detailed study will reveal all environmental systems to be explicitly or implicitly time-dependent to some extent. The essential aspect of tracer studies stressed here is recognition of important time-dependent elements and their inclusion in models of tracer behaviour. Neglect of significant time-dependencies in tracer experiments will be shown to lead in some instances to erroneous conclusions.

Attempts to use deconvolution techniques designed for time-invariant systems, as mentioned in I,2.1.3, without accounting for time-varying behaviour, can result in oscillations and unrealistic results. These effects have been noted in the tracer and hydrologic literature, e.g. Eriksson (1963), Eagleson et al. (1966) and Blank et al. (1971); however no direct solution for time-dependence was offered.

While no general solution seems to be feasible, there are several approaches that are satisfactory under certain conditions of practical significance. These are discussed and demonstrated in part III of this study and in several other articles (Mandeville and O'Donnell, 1973; Niemi, 1973; Ur and Lissack, 1973).

4. Time relations in the non-steady state

4.1. *Introduction*

We will now re-examine the various distributions and densities introduced in Part I in order to test

Table 2. *Representative types of time-dependence*

	Type of dependence	Flow	Response functions	Examples
1	No implicit or explicit time-dependence	$q(t)$ variable	$h^m(\tau)$ $h^c(\tau)$	gravity flow through mixed, variable volume, linear vertical reservoir
2	Implicit dependence of system response on flow rate	$q(t)$ variable	$h^m(\tau)$ $h^c(\tau; q(t))$	constant volume, completely mixed compartment
3	Implicit dependence of system response on flow rate	$q(t)$ variable	$h^m(\tau; q(t))$ $h^c(\tau; q(t))$	nonlinear rainfall-runoff relations for a watershed
4	Explicit time-dependence of system response	$q_i = q_o$ constant	$h^m(\tau)$ $h^c(\tau; t)$	transport processes in reservoirs controlled by weather variables
5	Explicit time-dependence of system response and flow variability	$q(t)$ variable	$h^m(\tau; t, q(t))$ $h^c(\tau; t, q(t))$	transient water flow through unsaturated soil (incl. root activity, evaporation, evapotranspiration, hysteresis)

their validity in the non-steady state and modify them whenever required.

We will use the concepts of time as discussed in I,2.2.1. The generalization of the density $h(\tau)$ has already been discussed in Section 1 and will be used throughout the following.

4.2. *The $F(\tau; t)$ distribution*

In extending the generalization of the $h(\tau)$ density, we find it expedient to define the ensuing distributions. Analogously to I,2.2.2 we define

$$F_r^c(\tau;t_i) = q_i(t_i) \int_0^\tau h_r^c(\tau';t_i)d\tau' \tag{17a}$$

$$\frac{dF_r^c(\tau;t_i)}{d\tau} = q_i(t_i)h_r^c(\tau;t_i) \tag{17b}$$

From normalization of h_r^c follows that $F_r^c(\infty;t_i) = q_i(t_i)$.

$$\frac{F_r^c(\tau;t_i)}{q_i(t_i)} = \int_0^\tau h_r^c(\tau';t_i)\,d\tau' \tag{17c}$$

is the fraction of input at t_i that will leave the system before $t_i + \tau$.

On the other hand, the density $h_{wo}^c(\tau;t_o)$ leads to a distribution

$$F_{wo}^c(\tau;t_o) = q_o(t_o) \int_0^\tau h_{wo}^c(\tau';t_o)\,d\tau \tag{18a}$$

$$\frac{F_{wo}^c(\tau;t_o)}{q_0(t_0)} = \int_0^\tau h_{wo}^c(\tau';t_o)\,d\tau'$$

is the fraction of the output at t_o that entered the system after $t_o - \tau$.

From (5) and (7) we obtain

$$\begin{aligned} F_{wo}^c(\tau;t_o) &= \int_0^\tau q_i(t_o-\tau')\,h_{wi}^c(\tau';t_o)\,d\tau' \\ &= \int_0^\tau q_i(t_o-\tau')\,h_r^c(\tau';t_o-\tau')\,d\tau' \end{aligned} \tag{18b}$$

$F_r^c(\tau;t_i)$ is in general not equal to $F_{wo}^c(\tau;t_o)$.

Both distributions being time-dependent, we can ask about their time averages $\overline{F_r^c(\tau)}$ and $\overline{F_{wo}^c(\tau)}$. In stationary systems these should not depend on time.

$$\overline{F_r^c(\tau)} = \overline{q_i(t_i) \int_0^\tau h_r^c(\tau';t_i)\,d\tau'} \tag{19a}$$

$$\overline{F_{wo}^c(\tau)} = \overline{\int_0^\tau q_i(t_o-\tau')\,h_r^c(\tau';t_o-\tau')\,d\tau'} \tag{19b}$$

Reversing the order of time averaging and integration in (19a) and (19b), we find that these expressions are equal:

$$\overline{F_r^c(\tau)} = \overline{F_{wo}^c(\tau)} \tag{20a}$$

The physical interpretation of (20a) is exemplified by Fig. 5. Recall from Fig. 2a that $h_r^c(\tau;t_i)$ is located on the line diagonal between the τ and t axes, whereas from Fig. 4 $h_{wo}^c(\tau;t_o)$ lies on the τ axis. The parallelogram $ACFE$ is a typical area of averaging of $F_r^c(\tau)$ while the rectangle $BDFE$ is a corresponding area for $F_{wo}^c(\tau)$. Note that the areas of averaging are the same, except for the two triangular end effects. For a long averaging period and a stationary system, the end effects are

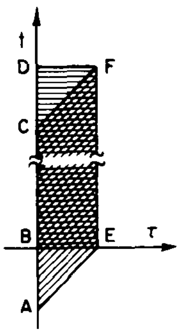

Fig. 5. Physical interpretation of time-averaged fluxes.

negligible and $\overline{F_r^c(\tau)} = \overline{F_{wo}^c(\tau)}$. The same cannot be said for a nonstationary system nor for any pair $F_r^c(\tau;t_i)$, $F_{wo}^c(\tau;t_o)$ where the instantaneous non-steady state may be influential.

Expanding $\overline{F_r^c(\tau)}$, from (19a) we obtain

$$\overline{F_r^c(\tau)} = \overline{q_i(t_i)}\ \overline{\int_0^\tau h_r^c(\tau';t_i)\,d\tau'} + \overline{q_i(t_i)'\left(\int_0^\tau h_r^c(\tau';t_i)\,d\tau'\right)'} \qquad (20b)$$

In stationary, conservative systems $q_i(t_i) = q_o(t_o) = q$ and

$$\overline{\int_0^\tau h_r^c(\tau';t_i)\,d\tau'} = \overline{\int_0^\tau h_r^c(\tau')\,d\tau'} \qquad (20c)$$

For uncorrelated $q_i(t)$ and $h_r^c(\tau;t)$ (20b) becomes

$$\overline{F_r^c(\tau)} = q\int_0^\tau \overline{h_r^c(\tau')\,d\tau'} \qquad (20d)$$

which is analogous to the steady-state formulation, I(3a).

We must bear in mind, however, that in many geophysical environments $q_i(t)$ and $h_r^c(\tau;t)$ are correlated, e.g. Nir (1973) and several examples in this work. The physical reason is that in some systems the fluctuation of the input of a major component modifies the transit time distribution in the whole system, the disturbance being propagated with appropriate phase and group velocity (e.g. Glover and Johnson, 1974).

However, for an ideal tracer we expect that fluctuations of tracer concentrations will not affect the system response. This does not allow us to assume lack of correlation between these quantities without additional analysis, as discussed in Section 3.5. They may be correlated indirectly, through correlations between bulk input and its tracer concentrations, as for example when tracer quantity is injected at a constant rate into varying input.

4.3. *The population analogy*

The population analogy presented in Part I can now be extended to non-steady-state systems, which provide a better description of the real world. The birth rate is given by the input function $q_i(t_o - \tau)$. Birth and death are meant here to include all possible entries into and exits from the system; however, entries at ages above 0 are not considered here. The population of age T at t_o (observation time) will be that which was born at $t_o - T$, i.e. $q_i(t_o - T)$, less than which died before reaching age T, i.e. less $F_r^c(T;(t_o - T))$:

$$\begin{aligned} q_i(t_o - T) - F_r^c(T;t_o - T) &= q_i(t_o - T)\left(1 - \int_0^T h_r^c(\tau;t_o - T)\,d\tau\right) \\ &= q_i(t_o - T)\int_T^\infty h_r^c(\tau;t_o - T)\,d\tau \\ &= \overline{F}_r^c(T;t_o - T) \end{aligned} \qquad (21a)$$

Analogously to the steady-state case, this age group is composed of sub-groups of all lifetimes above T. However, the birth rate $q_i(t_o - T)$ is variable—it may have trends, periods or random fluctuations. The probability of leaving the system after the age T, which may be considered as a probability of survival to that age, given by the term

$$\int_T^\infty h_r^c(\tau;t_o - T)\,d\tau$$

is also variable. Both cause variability of the age group with time.

In analogy to the relation expressing conservation of probability in biological populations (Bartlett, 1970; Gani, 1973; Rubinow, 1974) we write

$$\frac{\partial \bar{F}(T,t)}{\partial t} = -\frac{\partial \bar{F}(T,t)}{\partial T} - \lambda(T,t)\bar{F}(T,t) \qquad (21b)$$

Here the first term on the right accounts for the effect of age composition and the second accounts for the removal from the population (exit, death). $\bar{F}(T,t)$ in the "biological" notation is equivalent to $\overline{F}_r^c(T;t_o - T)$ in (21a), the "geoscience" notation.

From (21a) we obtain

$$\begin{aligned} -\frac{\partial \bar{F}}{\partial T}(T,t) = &-\frac{\partial q_i(t)}{\partial T}\int_T^\infty h_r^c(\tau;t) \\ &- q_i(t)\int_T^\infty\left(\frac{\partial}{\partial T}(h_r^c(\tau;t))\right)d\tau \\ &+ q_i(t)h_r^c(T;t) \end{aligned} \qquad (21c)$$

On the other hand the change along a life-line (diagonal) is given by differentiation with respect to T at $t_o - T =$ constant

$$\frac{d\bar{F}(T,t)}{dT} = -q_i(t)h_r^c(T;t) \tag{21d}$$

In geophysical systems a removal term due to radioactive decay may add an additional term, $-\lambda\bar{F}(T;t)$, resulting in

$$\frac{d\bar{F}(T,t)}{dT} = -q_i(t)h_r^c(T;t) - \lambda\bar{F}(T,t) \tag{21e}$$

More detailed insight into biological populations will show that the input rate depends on the population age structure and may even contain inputs at ages other than 0. This is, however, beyond the scope of our consideration, though some geophysical analogy could be conceived.

We require now three dimensions to represent the age and transit time structure of a system in non-steady state, while two dimensions were adequate to describe the steady state (see I, Figs. 3, 4 and 5). Following Skellam (1967, 1972) we represent in Fig. 6 a segment of population which originated at $t_o - T$ (a cohort), which is therefore of age T at time t_o. Each individual may be represented by a horizontal track advancing in time and age. They are arranged vertically in decreasing order of survival, so as to produce a surface over the time–age plane.

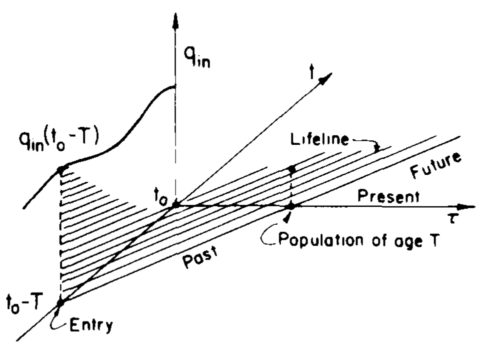

Fig. 6. Population analogy of transit time structure.

An alternative, more quantitative representation is given in Fig. 7. Each square element $dt\,d\tau$ in the t–τ plane is assigned a value $q_i(t_o - T)$ $h_r^c(\tau;(t_o - T)_i)\,dt\,d\tau$, which equals the flux of particles leaving the system at time $t_o - T + \tau$ to $t_o - T + \tau + d\tau$ which entered at time $t_o - T$ to $t_o - T + dT$. It is the complete specification of the transit time and age densities in the system, valid at time of observation t_o. Various integral representations like that of Fig. 6 may be derived from it. One should recall, however, that such representation entails knowledge of future behaviour of the system ($t > t_o$), which is applicable only for predictive deterministic systems. However, this formulation can be used for simulation and prediction of stochastic properties of future behaviour of stationary systems.

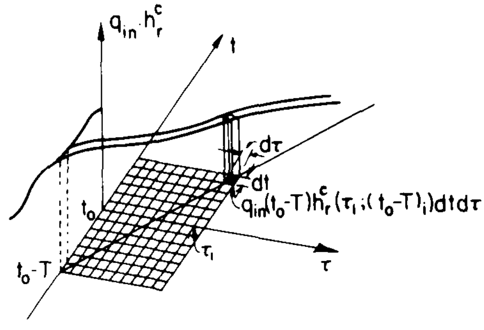

Fig. 7. Quantitative representation of population transit time structure.

4.4. *The $\phi(\tau;t)$ density and the $M(\tau;t)$ distribution*

In analogy to the steady-state theory in I,2.2.4 we define $\phi(\tau;t)d\tau$ as that fraction of the mass in the system at time t having transit time between τ and $\tau + d\tau$

$$\phi(\tau;t_o)\,d\tau = \frac{d\tau}{M(t_o)} \int_0^\tau q_i(t_o - T)h_r^c(\tau;t_o - T)\,dT \tag{22a}$$

where $M(t_o)$ is the total mass in the system at time t_o. The corresponding steady-state expression is given by I(4a).

The associated distribution $M(\tau;t_o)$ is

$$M(\tau;t_o) = M(t_o)\int_0^\tau \phi(\tau';t_o)\,d\tau'$$

$$= \int_0^\tau \int_0^{\tau'} q_i(t_o - T)\,h_r^c(\tau';t_o - T)\,dT\,d\tau' \tag{22b}$$

The corresponding steady-state expression is given by I(5). Fig. 8 shows the area of integration of

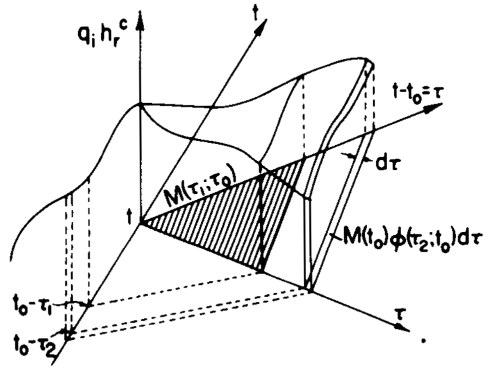

Fig. 8. System mass transit time structure.

$\phi(\tau; t_o)$ and $M(\tau; t_o)$. The total mass $M(t_o)$ is given by

$$M(t_o) = \lim_{\tau \to \infty} M(\tau; t_o)$$
$$= \int_0^\infty \int_0^{\tau'} q_i(t_o - T) h_r^c(\tau'; t_o - T)\, dT\, d\tau' \tag{23}$$

4.5. *The $\psi(T;t)$ density and the $M(T;t)$ distribution*

In analogy to the steady-state theory in I,2.2.5 we define the $\psi(T;t)$ density as that fraction of the mass in the system at time t, having ages between T and $T + dT$. Following (21a) we have

$$\psi(T; t_o)\, dT = \frac{dT}{M(t_o)} q_i(t_o - T) \times \left(1 - \int_0^T h_r^c(\tau; t_o - T)\, d\tau\right)$$
$$= \frac{dT}{M(t_o)} \left(q_i(t_o - T) - F_r^c(T; t_o - T)\right) \tag{24a}$$

based on past information, or

$$\psi(T; t_o)\, dT = \frac{dT}{M(t_o)} q_i(t_o - T) \times \int_T^\infty h_r^c(\tau; t_o - T)\, d\tau$$
$$= \frac{dT}{M(t_o)} \bar{F}_r^c(T; t_o - T) \tag{24b}$$

based on future information.

The associated distribution $M(T; t_o)$ is

$$M(T; t_o) = M(t_o) \int_0^T \psi(T'; t_o)\, dT'$$
$$= \int_0^T \left[q_i(t_o - T') \int_{T'}^\infty h_r^c(\tau; t_o - T')\, d\tau\right] dT'$$
$$= \int_0^T \bar{F}(T'; t_o - T')\, dT' \tag{25}$$

which may be based on integration of past or future information, depending on the nature of the problem.

Fig. 9 shows the area of integration of $\psi(T, t_o)$ and $M(\tau; t_o)$.

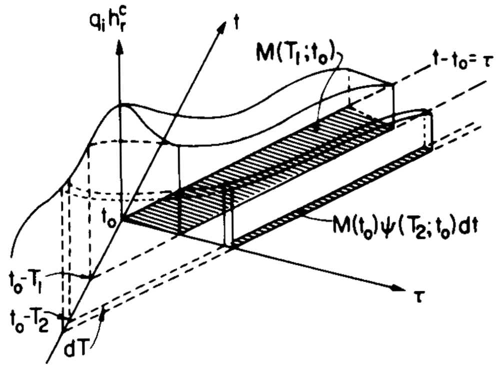

Fig. 9. System mass age structure.

The total mass is

$$M(t_o) = \lim_{T \to \infty} M(T; t_o)$$
$$= \int_0^\infty \left[q_i(t_o - T') \int_{T'}^\infty h_r^c(\tau; t_o - T')\, d\tau\right] dT' \tag{26}$$

We expect on physical grounds that (23) and (26) should converge to the same total mass $M(t_o)$. By changing the order of integration in (26) we find that the formulations are equivalent as they cover the same area of integration (Kaplan, 1952).

4.6. *Average transit time of incoming and outgoing flux. Average transit time of a system. Average age of a system*

For an incoming flux we define the average transit time similarly to I,2.2.7,

$$\bar{\tau}_i^h(t_i) = \int_0^\infty \tau h_r^c(\tau; t_i)\, d\tau \tag{27}$$

which is independent of the in- and outflow. For an outgoing flux we obtain from (5) and (9a) an expression which depends on the flows

$$\bar{\tau}_o^h(t_o) = \frac{1}{q_o(t_o)} \int_0^\infty \tau q_i(t_o - \tau) h_{wi}^c(\tau; t_o)\, d\tau$$
$$= \int_0^\infty \tau h_{wo}^c(\tau; t_o)\, d\tau \qquad (28)$$

$\bar{\tau}_i^h(t_i)$ and $\bar{\tau}_o^h(t_o)$ may therefore be different and may have different time-dependence. They do not necessarily equal any of the expressions $M(t_i)/q(t_i)$, $M(t_o)/q(t_o)$, $M(t)/q(t)$ or $M(t)/q(t)$ which are often found in literature as indicators of transit time.

We can ask about the values of the ensemble average or, for ergodic systems, the time averages of both expressions (27) and (28). As shown in Appendix 1, direct averaging over t_i and t_o respectively leads to expressions for $\bar{\bar{\tau}}_i^h$ and $\bar{\bar{\tau}}_o^h$ which are generally different. However, weighted averages, i.e. $\overline{q_i(t_i)\bar{\tau}_i^h(t_i)}$ and $\overline{q_o(t_o)\bar{\tau}_o^h(t_o)}$, are equal.

As shown in (14) the h_r^c and h_{wo}^c densities are generally unequal and the corresponding average transit times will also be so.

For the system (mass) average transit time we obtain from (22a) and analogously to I(10),

$$\bar{\tau}_\phi(t_o) = \int_0^\infty \tau \phi(\tau; t_o)\, d\tau$$
$$= \frac{1}{M(t_o)} \int_0^\infty \tau \int_0^\tau q_i(t_o - T) h_r^c(\tau; t_o - T)\, dT\, d\tau$$
$$= \frac{1}{M(t_o)} \int_0^\infty \tau F_{wo}^c(\tau; t_o)\, d\tau \qquad (29)$$

For the average age of the system (mass) we obtain from (24a), analogously to I(12)

$$\bar{T}_\phi(t_o) = \int_0^\infty T \psi(T; t_o)\, dT$$
$$= \frac{1}{M(t_o)} \int_0^\infty T q_i(t_o - T)$$
$$\times \left(1 - \int_0^T h_r^c(\tau; t_o - T)\, d\tau\right) dT$$
$$= \frac{1}{M(t_o)} \int_0^\infty T \bar{F}_r^c(T; t_o - T)\, dT \qquad (30)$$

5. Summary

In the preceding sections an attempt was made to formalize the concepts of tracer behaviour in systems which are in a non-steady state. The ensuing formalism is a direct extension of the steady-state theory presented in Part I. As before it draws on the presentation and results of other disciplines, primarily biological population dynamics for the theoretical basis while the more practical aspects benefit from the experiece of electronic and chemical engineering and physiological work with tracers.

While the results of the general theorems and formulations may seem overly abstract for the prospective user, they provide the basic formal and intuitive structure to which various simplifying assumptions can be applied. They may also serve as a framework for simulation of tracer behaviour in situations which do not allow for analytical simplifications. Some of these simplifying assumptions and examples are discussed in Part III.

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Appendix 1

Ensemble (or time) averages of average transit times

The ensemble averages, or for ergodic systems, the time averages of expressions (27) and (28) are evaluated as follows:

Averaging $\bar{\tau}_i^h(t)$ over t_i yields

$$\bar{\bar{\tau}}_i^h = \int_0^\infty \overline{\tau h_r^c(\tau; t_i)\, d\tau} \qquad A(1)$$

in analogy to the steady-state expression.

Averaging of eq. (28) leads to a different expression, more complex due to correlation between three time-dependent factors:

$$\bar{\bar{\tau}}_o^h = \int_0^\infty \tau\, \overline{\frac{q_i(t_o - \tau)}{q_o(t_o)} h_{wi}^c(\tau; t_o)}\, d\tau \qquad A(2)$$

In the singular case of constant input, $q_i = q_o = q$, and therefore permitting only explicit time-dependence, one obtains that the expressions are equal, in analogy to the argument used in proving (20a).

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О ТЕОРИИ ТРАССЕРОВ В ГЕОФИЗИЧЕСКИХ СИСТЕМАХ В СТАЦИОНАРНЫХ И НЕСТАЦИОНАРНЫХ СОСТОЯНИЯХ. ЧАСТЬ II. НЕСТАЦИОНАРНОЕ СОСТОЯНИЕ — ТЕОРЕТИЧЕСКОЕ ВВЕДЕНИЕ

В продолжение части I (Tellus, 27, 372, 1975), которая касалась стационарного состояния, теория трассеров расширяется на системы, меняющиеся со временем. Большинство геофизических систем представляются меняющимися со временем, если их наблюдать продолжительное время и в достаточных деталях. Мы различаем явную и неявную временную изменчивость систем. Единственная реакция составной системы в установившемся состоянии, $h^c(\tau)$, заменяется тремя функциями, зависящими от времени, которые связывают условия на входе и выходе системы. Временные соотношения для установившегося состояния также обобщаются и иллюстрируются эвристически путем аналогий с биологическими популяциями. Определяются входные и выходные средние времена перехода, также как их средние по времени или по ансамблю. Распределения по временам перехода и по возрасту для потоков и в объеме обобщаются на неустановившееся состояние. Указаны примеры различных категорий неустановившихся состояний в природных системах.