EUR 4636 e

COMMISSION OF THE EUROPEAN COMMUNITIES

PRINCIPLES AND METHODS FOR THE DERIVATION OF A FORMULA FOR THE DISCHARGE OF RADIOACTIVE LIQUID WASTES IN SURFACE WATERS

by

G. BRANCA, F. BREUER, A.A. CIGNA
(CNEN)
and
R. AMAVIS
(CEC)

1971

Report prepared by CNEN
Comitato Nazionale per l'Energia Nucleare
Rome — Italy

EURATOM Contract No. 044-68-12 PST I

LEGAL NOTICE

This report is on sale at the addresses listed on cover page 4

at the price of F. Fr. 6.65 B. Fr. 60.— DM 4.40 It. Lire 750 Fl. 4.30

When ordering, please quote the EUR number and the title, which are indicated on the cover of each report.

Printed by Ceuterick s.a. — Louvain
Luxembourg, April 1971

EUR 4636 e

COMMISSION OF THE EUROPEAN COMMUNITIES

PRINCIPLES AND METHODS
THE DERIVATION OF A FORMULA
FOR THE
CHARGE OF RADIOACTIVE LIQUID
WASTES IN SURFACE WATERS

EUR 4636 e

PRINCIPLES AND METHODS FOR THE DERIVATION OF A FORMULA FOR THE DISCHARGE OF RADIOACTIVE LIQUID WASTES IN SURFACE WATERS by G. BRANCA, F. BREUER, A.A. CIGNA (CNEN) and R. AMAVIS (CEC)

Commission of the European Communities
Report prepared by CNEN
Comitato Nazionale per l'Energia Nucleare, Rome (Italy)
Euratom Contract No. 044-68-12 PST I
Luxembourg, April 1971 — 44 Pages — 4 Figures — B. Fr. 60.—

In any nuclear project, it is essential to know exactly the maximum quantity of radioactive substances which may be disposed of in a specified environment. Many difficult problems are involved in the determination of a discharge formula: charac-

EUR 4636 e

PRINCIPLES AND METHODS FOR THE DERIVATION OF A FORMULA FOR THE DISCHARGE OF RADIOACTIVE LIQUID WASTES IN SURFACE WATERS by G. BRANCA, F. BREUER, A.A. CIGNA (CNEN) and R. AMAVIS (CEC)

Commission of the European Communities
Report prepared by CNEN
Comitato Nazionale per l'Energia Nucleare, Rome (Italy)
Euratom Contract No. 044-68-12 PST I
Luxembourg, April 1971 — 44 Pages — 4 Figures — B. Fr. 60.—

In any nuclear project, it is essential to know exactly the maximum quantity of radioactive substances which may be disposed of in a specified environment. Many difficult problems are involved in the determination of a discharge formula: charac-

ABSTRACT

In any nuclear project, it is essential to know exactly the maximum quantity of radioactive substances which may be disposed of in a specified environment. Many difficult problems are involved in the determination of a discharge formula: characteristics of radioactive wastes, characteristics of environmental media, various phenomena which may modify the distribution of radioactivity and all factors which influence the exposure to radiation for man.

In this report, a mathematical solution is given for the problem of the distribution of radionuclides in the environments which are associated with surface waters.

KEYWORDS

RADIOACTIVE WASTE
WASTE DISPOSAL
GROUND WATER
ENVIRONMENT
DISTRIBUTION
MATHEMATICS

PREFACE

The assessment of the possibilities of radioactive waste disposal in the environment is one of the major factors to be considered in any nuclear power project. It is essential to know exactly the maximum quantity of radioactive substances which may be disposed of in a given area during a given period, with due regard to the basic requirements of health protection, in order to safeguard the populations likely to be affected by the disposal of these wastes.

Many difficult problems are involved in the determination of this maximum quantity of waste, which may be defined as the limit radiological capacity for the receiving environment. For example, the nature and physico-chemical form of each radionuclide contained in the waste must be taken into account. The nature, composition and utilisation of the receiving hydrobiological medium must be subjected to thorough examination in both space and time.

The radiation hazard resulting from the disposal of radioactive wastes in the environment must also be determined, along with these various other factors. Moreover, it is essential that the problem should be looked at as a whole, in all its aspects.

This document uses a mathematical approach based on the compartment theory. It is essentially a simplified analytical model, but one which may be amplified for concrete, clearly defined situations. This moreover presupposes detailed knowledge of the factors of transfer between two compartments, which, unfortunately is not always present. But the characterisation of absent data may provide the necessary lead for new studies and experimental research. Moreover, a method of this kind should make it possible:

a) to interpret transfer factors more effectively by means of a detailed analysis of the phenomena which determine them and by seeking to establish what causes them to vary in particular situations;

b) to establish a more accurate calculation method for the evaluation of radiation doses sustained by individual members of a population as a result of the disposal of radioactive wastes in the environment;

c) to improve the planning of measures to be taken for the general survey of the area surrounding nuclear power plants;

d) to acquire new knowledge which will provide a better general insight into all the processes affecting the distribution of radioactivity from the moment of disposal until its intake by man.

This study is an interesting contribution to the solution of an important health physics problem, by a more accurate and more objective determination of the quantities of radioactivity likely to be disposed of in the environment.

P. Recht

CONTENTS

Si quis in hoc artem populo non novit amandi
me legat : et lecto carmine doctus amet.
Ovid, Ars amatoria, I.

1. — INTRODUCTION

The term environmental pollution in its widest meaning implies a chemical, physical, or biological change of the environment as a result of the introduction of foreign substances into it.

Therefore, defining "radioactive contamination" as that particular form of pollution due to radionuclides not present naturally in a certain environment, it is evident that any discharge, albeit a modest one, of radioactive substances can represent a definite contamination of that environment. Nevertheless, in practice, when evaluating a state of pollution (or of contamination) one cannot leave out of consideration the presence or absence of harmful effects of the above changes, meaning by harmful effects those effects which render the environment less suitable for any of those uses for which it would be suitable if it were in its natural state. With this more precise statement the conception of pollution, or of contamination) assumes a significance which is no longer strictly tied to naturalistic criteria, but rather to broadly economic criteria and to the concept of use. From this it follows that the total preservation of the natural characteristics of an environment is not the objective to pursue everywhere and always, in the struggle for the protection of it. In the case of protection from ionizing radiation the basic criterion consists, as is well known, not only in limiting the injury to persons but also in positively containing the risk of such damage within limits which are not statistically significant. This extreme caution, which is rarely encountered in the manner in which conventional pollution is considered, of course implies that all the other components of the environment are protected from possible harmful changes, considering that man is among the organisms most sensitive to radiation.

The above mentioned criterion finds its expression in the condition that no one shall receive by total irradiation (internally and/or externally) doses greater than those established as dose limits by the basic standards for the protection of health against the hazards of ionizing radiation. If therefore the nature and the amount of the radioactive substances and the rate at which they are discharged into an environment are such that the dose limits are not exceeded, then one can exclude the occurrence of "radioactive contamination" in the previously specified sense. For every environment therefore there exists a maximum quantity of radioactive substances which can be introduced in a given time, under specified conditions and in a specific manner, without there being "contamination", i.e. without producing a situation which can carry a statistically significant risk of harm to the population. The precise knowledge of such a quantity is thus indispensable in safeguarding public health; it must also be said, however, that an over-cautious estimate of the receptive capacity of an environment could cause unwarranted limitations on the discharges and therefore a limitation in the corresponding industrial or research activity, or else the adoption of unnecessary treatment or precautions, so that it is easily understood that in any case one ends with a greater economic burden. The determination of the quantity of radioactive substances which can be discharged into an environment, in such a way as to balance the fundamental requirements of public health protection with those of general economics, presupposes a knowledge of the nature of the waste and the manner of the discharge. It also presupposes knowledge of the chemical, physical and ecological characteristics of the receiving environment, and of the phenomena which affect the radionuclides after discharge and which are able to have an influence on their distribution in such an environment, and finally of the use which is made of that environment or of some parts of it.

The exact evaluation of the above mentioned factors allows one in the first place to single out from among all the particular population groups which are in some way tied to the environment that group of people most exposed to the risk of radiation injury from a given radionuclide. This group is known as the *critical group* and the pathway taken by the radionuclide which could cause such a group to suffer irradiation is known as the *critical pathway*. Once one has distinguished the critical group for a specific radionuclide one can proceed to calculate the maximum quantity of that radionuclide which can be discharged systematically in unit time (in the absence of other radionuclides) into the receiving environment concerned, without such a critical group exceeding in total the dose limits given by the basic standards; this is known as the *maximum quantity of a radionuclide for a specified environment*.

In this report, after an examination of the characteristics of the discharges and of the environment meant to receive them, we consider the various complex phenomena which may alter the distribution of the radioactivity in the environment and also the social and economic factors which influence the exposure to radiation which could follow for man. The mathematical solution is given for the problem of the distribution of a specified radionuclide in an environment. The way by which it is possible to distinguish the critical groups and finally the way to calculate the maximum quantity of that radionuclide for that environment are described.

In practice, very often more than one radionuclide is contained in the discharged wastes; in such a case it is necessary to repeat the procedure and, given the possibility that other radionuclides also reach man, in general the quantity of each one that can be introduced into the environment will prove to be less than the corresponding maximum quantity. The combined expression which results will constitute the *discharge formula* for a certain establishment and for a particular environment.

The report contains a description of the procedure which allows one to derive such a discharge formula in various cases.

The receiving environments considered in this report are those associated with surface waters, indicated below by the generic expression "bodies of water". These exclude sea water, both ocean and coastal waters. Water in underground aquifers is dealt with here only to the extent that it can interact with surface water.

The considerations and results in this study are valid generally. Nevertheless, in practice, the less important cases (clinics, university laboratories, small users etc.) for which the procedure would be unduly burdensome can apply other criteria which are simpler but naturally more restrictive*.

Obviously this work is not intended to completely exhaust the subject but only to develop certain basic concepts, since the complexity of the phenomena and the extreme diversity of the various actual situations would necessitate going into more detail which would take much more time than has been available.

Finally, it is well to observe clearly that all the experience concerning radioactive discharges into the environment and in particular the criteria which are stated in the following pages can be usefully extended to the problem of conventional chemical pollution.

2. — THE CHARACTERISTICS OF THE WASTES

When radioactive liquid wastes are discharged into a body of water, a series of phenomena or processes take place or can take place. These are of the physical, chemical, biological or mixed type, some of which can dilute or disperse the dangers connected with radioactivity, whereas

---

(*) Some of these criteria are described by L. Cigna Rossi and A.A. Cigna, *Giorn. Fis. San. Radioprot.*, **12** (4), 261-271 (1968).

others assist, to varying degrees, in the concentration of such dangers at particular points in the system.

It is necessary therefore to take into account and to balance all these processes in facing the problem of the evaluation of the maximum quantity, for a particular hydrobiological environment, in regard to a specified radionuclide. From the moment of discharge, the phenomena that involve the radionuclides depend on the characteristics of the wastes and on those of the receiving environment, to the extent to which the former interact with the latter in reciprocal and various combinations.

The properties of discharges will be reviewed in this section and the various possible forms of interactions with the environment will be briefly indicated. Naturally, given the aims of this report, we shall examine only those properties capable of determining the distribution in time and space of the radionuclides associated with the wastes, whilst all those aspects which only concern conventional pollution will be disregarded.

The significant parameters which exert an influence in one way or another on the fate of the radionuclides introduced into the environment can be classified as in the following paragraphs.

Physical parameters (Temperature and density of the liquid wastes, density and dimensions of the contaminants possibly present in a particulate form, half-life, presence of substances capable of exercising a dispersing action, etc.).

Chemical and *chemical-physical parameters* (Chemical-physical state and the nature of the substances responsible for the contamination, acidity, alkalinity, salinity, etc.).

Manner of discharge (Position and shape of the effluent outlet, rate of discharge, predilution of the wastes, etc.).

We will consider each of these properties separately.

Temperature

This parameter can influence the diffusion due to convection. If the effluent has a higher temperature than the dilutant body of water and is injected into the latter at a depth close to that of the open surface, then the vertical mixing takes place slowly. This is because the effluent is lighter and floats on the receiving water, especially if the turbulence due to the motion of the current is slight or absent. If the discharge takes place into a lake, which often has a thermal stratification, this may result in a sheet-like distribution of the effluent in which it mixes slowly with the waters of the lake; this can happen even if the injection is effected at some depth.

The temperature of the effluent can also exercise a certain influence on the phenomena of adsorption and desorption.

Density

This parameter also can influence the turbulent diffusion. Evidently, beside the temperature, already considered as a separate parameter, other causes can produce an effluent density different from that of the receiving water, for example a higher salinity or the nature of the wastes themselves (oils, fats). In the first case the liquid wastes are heavier than the external waters and if they are introduced into the receiving waters at a certain depth there may be a well-defined saline stratification. In the second case, where oils and/or fats are present, the lower density causes the wastes to float on the receiving water with the formation of a floating film of a greater or lesser thickness and area. In both cases, the complete mixing of the effluent with the receiving water may take place only slowly and with difficulty, especially in the absence of turbulent motion due subjected after discharge. These processes determine the subsequent route of that substance. The physical type of processes (except, in rare cases, the radioactive decay) do not modify the initial chemical-physical state, whilst the others generally do influence the state.

The chemical nature of the substances responsible for the contamination

Looked at in relation to the characteristics of the receiving environment, the chemical nature of substances to which the contamination is due can have a notable influence on the fate which such substances meet with after discharge.

TABLE I

The Processes to which the Radionuclides may be Subjected after Discharge

Physical-chemical state of the contamination	Type of process			
	Physical	Chemical-physical	Chemical	Biological
Ionised-molecular	Transport by water movement. Turbulent diffusion. Ionic or molecular diffusion. Radioactive decay.	Isotopic dilution. Elution. Ion exchange. Adsorption. Coprecipitation.	Chemical reaction (formation of insoluble compounds). Formation of complexes.	Isometabolic dilution. Ingestion by animals (including man). Absorption by plants. Surface adsorption by animals or plants Sedimentation of dead plants and animals. Transport by migration of animals.
Complexed	Transport by water movement. Turbulent diffusion. Ionic or molecular diffusion. Radioactive decay.	Isotopic dilution. Elution. Ion exchange. Adsorption. Coprecipitation.	Chemical reaction (formation of insoluble compounds, destruction or dissociation of the complex).	Isometabolic dilution. Ingestion by animals (including man). Absorption by plants. Surface adsorption by animals or plants. Sedimentation of dead plants and animals. Transport by migration of animals.
Particulate	Transport by water movement. Turbulent diffusion. Radioactive decay. Suspension. Sedimentation.	Isotopic dilution. Dissolution. Dispersion (sol formation).	Chemical reaction.	Ingestion by animals (including man). Surface adsorption by animals or plants. Sedimentation of dead plants and animals. Transport by migration of animals.
Colloidal	Transport by water movement. Turbulent diffusion. Radioactive decay.	Isotopic dilution. Dissolution. Adsorption. Coagulation.	Chemical reaction.	Ingestion by animals (including man). Surface adsorption by animals or plants. Sedimentation of dead plants and animals. Transport by migration of animals.

The most important chemical phenomenon is evidently that connected with possible reactions which involve the contaminating substances (formation of insoluble compounds, and subsequent precipitation of the latter, the formation of complexes, and reciprocally the dissociation or destruction of complexes initially present, chemical reaction of particulate or colloidal substances etc. with transition into solution).

One can cite, for example, the possibility that alkaline-earth elements, the rare earths etc., meet precipitating agents, such as for example fluorine, in the receiving waters.

Another example occurs if the receiving waters are in equilibrium with carbonate, they present a high degree of water hardness due to carbonates, and if for some reason (increase of temperature, aeration) the free carbon dioxide content becomes less; there is then a formation of an insoluble precipitate of calcium carbonate. If strontium and rare earths are present in the liquid wastes, then these elements will also precipitate as carbonates. In fact this is a somewhat improbable event, because the surface waters tend rather to attack such precipitates than to promote their formation.

Also important is the content of radioactive transition elements (like iron, cobalt, nickel, ruthenium, rhodium, palladium) in the effluent, on account of the relative ease with which such elements form compounds, sometimes insoluble, or complexes with ions present in the receiving waters (sulphates, chlorides, nitrates).

If the contamination is initially present in a particulate form, it can become subject to chemical reaction with the receiving water which causes its transition into solution (corrosion). Also, any colloids present can meet conditions of pH for example which cause them to become more soluble, whilst complexes can become dissociated or more rarely destroyed if suitable environmental conditions apply. For example, strontium complexes can partially dissociate if a high concentration of calcium ions is present, which upset the equilibrium by entering the molecule of the complex in place of the strontium.

The influence of the nature of the substances responsible for the contamination manifests itself also in the phenomena and processes of chemical-physical type which can take place after the discharge. Of these, isotopic dilution, ion exchange, coprecipitation, adsorption, and dissolution are of particular interest from the point of view considered here. All these phenomena are discussed in detail in section 4. Also the biological phenomena are affected by the chemical nature of the contaminating substances, as will be better seen in section 4.

Acidity and alkalinity

The influence of the pH value of the liquid wastes in relationship with that of the environment can affect the phenomena of flocculation and coprecipitation and naturally also the inverse processes. For example, if the effluent possesses an alkaline pH value and contains hydroxides of aluminium or of iron, or basic phosphates incorporating radioactive substances, the lowering of the pH value which occurs after the discharge can produce peptisation of the floc and the transition into solution of the radionuclides. As has already been pointed out, by increasing solubility and dispersion, the alkalis also influence the physical fate of the radionuclides.

Vice versa, if the liquid wastes have a high acidity and contain iron or aluminium, then after the discharge they can produce the phenomena of flocculation and coprecipitation resulting in part of the activity in the liquid wastes sinking to the bottom. Certain pH values of the liquid wastes combined with those of the environment can also influence the mechanism of ion exchange and in general the absorption of certain radionuclides by the sediments or suspended materials in the receiving water, and naturally also the reverse processes of elution and desorption.

Salinity of the liquid wastes

It has already been indicated how such a parameter can influence the density of the liquid wastes and hence also their turbulent diffusion. A high salinity can also cause the elution of the radionuclides previously fixed by ion exchange on the clays or the humic substances present in the muddy water or the sediment.

The salinity of the liquid wastes also affects the isotopic and isometabolic dilution of the radionuclides.

Shape and position of the effluent outlet; predilution of the liquid wastes

Evidently these parameters influence the mixing of the liquid wastes with the waters of the receiving body and therefore the rapidity of the process of dispersion of the radionuclides into the environment.

Having reviewed the principal parameters which are capable in some way of influencing the fate of the radionuclides introduced into a hydrobiological environment, one can pose the problem of whether or not it is possible to quantify such parameters according to the origins of the liquid wastes, that is, whether is possible to relate the characteristics of the wastes to their type of origin.

At present the possible sources of radioactive liquid wastes are principally the following.

a) Installations for the chemical treatment by lixiviation of the minerals of uranium and thorium.

b) Installations for the metallurgy of nuclear fuels.

c) Reactors.

d) Installations for the reprocessing of irradiated nuclear fuels.

e) Radiochemical laboratories, radiobiological laboratories etc.

f) Decontamination centres.

g) Nuclear research centres.

Only some of these give rise to liquid wastes which are of a more or less definite and constant composition.

As regards the concentration by chemical methods of the minerals of uranium and thorium, although there are many types of minerals which contain these elements, one can say that for all of them processes are used which involve digesting them with acid or alkaline reagents. As a consequence the liquid wastes are aqueous solutions with pH values which in different cases can drop very nearly to 1.5 or go up to about 10. The radioactive contamination consists of ^{226}Ra, ^{230}Th and ^{234}Th, partly dissolved and partly suspended. The wastes have a high non-radioactive chemical content of manganese, copper, boron, arsenic, nitrates, sulphates and fluorides. The actual metallurgical phase of the manufacture of fuel elements begins with a uranium or thorium concentrate and leads to the production of metallic uranium, uranium hexafluoride, or uranium dioxide. The liquid wastes produced in the course of these forms of treatment have a chemical and radiochemical composition somewhat similar to that already described for the liquid wastes which arise from the concentration of the minerals.

The production of fuel elements in turn gives rise to aqueous wastes containing traces of uranium in solution, and of other metallic ions such as aluminium, magnesium, zirconium and also nitrate and fluoride ions resulting from the pickling operations.

All the liquid wastes referred to so far are in general only weakly contaminated and their elimination presents no problem. The liquid wastes originating from nuclear reactors, on the contrary, have a composition which varies generally from one type of reactor to another, whereas

Treatments based on processes of the biological type (activated sludge, percolating beds etc.).

Evidently the process or the combination of processes adopted influences appreciably the characteristics of the effluents; thus the liquids subjected to sedimentation or filtration will have a rather low suspended-solids content, the liquids resulting from a process of ion exchange or of electrodialysis will have a negligible salinity, the supernatant from a calcium-iron-phosphate flocculation will have a high pH value, and so on.

These few examples demonstrate the importance of an exact knowledge of the type of source and especially the treatment of the radioactive liquid wastes when evaluating the maximum quantity that can be discharged into the receiving environment. Indeed the source and possible treatment precisely determine the characteristics of the liquid wastes and, as has been seen, it is these which influence the fate of the radionuclides.

3. — THE CHARACTERISTICS OF THE RECEIVING ENVIRONMENT

When radioactive liquid wastes are introduced into a body of water they then disperse over an area (*the receiving environment*) with boundaries which are generally somewhat difficult to define.

On the other hand the problem of the location of the area boundaries is less important than at first it may seem. The *critical area* however must be identified with great accuracy and this will be defined later. Immediately after discharge the wastes meet a water-course or a lake (aquatic environment); such an environment is defined here as the *primary receiving environment*. Later the radioactive contamination can be transferred to the land environment (plants and/or animals) which one may call the *secondary receiving environment*. Obviously, from the point of view of propagation to man, the radionuclides can come either directly from the primary or from the secondary environment, as will be explained in section 5.

Nevertheless the subdivision referred to above has advantageous results, since the interaction between the liquid wastes and the receiving environment takes place mainly in the aquatic environment.

3.1 — The Primary Receiving Environment

The characteristics of the aquatic environment can be subdivided into the following groups.

Physical parameters (flowrate, volume, temperature, density, water stratification, nature of the bottom).

Chemical and chemical-physical parameters (suspended solids content, pH values, oxygen concentration, salinity).

Biological parameters (biotic communities).

Each of the above mentioned characteristics will be briefly illustrated from the point of view of a possible interaction with the released radioactivity.

Flowrate is one of the most important parameters of water-courses and the factors on which this parameter depends are well known, in particular the meteorological factors. For the purposes of the present investigation, the knowledge of the *average annual flowrate* over a great number of years and also of the *average flowrate for each individual month* acquires a particular importance, as will be seen later.

Volume

If the receiving waters are not flowing (lakes, reservoirs etc.) then obviously one does not speak of the flowrate but rather of the volume of water relevant to the release. Such a volume need not necessarily coincide with the total volume of the whole body of water in question. Consider for example the case of a lake with thermal stratification, where the useful volume, as far as dispersion of the radionuclides is concerned, is at most the volume in which the circulatory motion occurs, i.e. that of the epilimnion (see "Water stratification").

Temperature

It is well known that this parameter depends on the absorption of solar radiation by the water and on the heat exchange with the atmosphere and with the bottom and is therefore variable within wide limits in time and space. The temperature exercises a direct influence on many of the chemical reactions which take place in the receiving water mass. In particular the rate at which the oxygen is consumed in the oxidation processes which take place in the hypolimnion (see "Water stratification") can double or triple following an increase of temperature of some ten degrees. Temperature is evidently important therefore in the processes of absorption of radioactive substances by aquatic organisms.

Density

For surface and ground waters the density values lie within fairly narrow limits, although in certain cases (brackish water) there is a substantial difference from the fresh-water values.

Water stratification

In certain types of aquatic environment water stratification phenomena can occur which are due to differences in density. The stratum which separates two strata of different density is called a *pinocline.* Where the stratification is of thermal origin, the stratum which has the most rapid drop of temperature is called the *thermocline* or the *metalimnion.* This stratum separates the water above it, known as the *epilimnion*, where there is no thermal stratification or if there is it is not important, from the water below it, which extends to the lake bottom and is known as the *hypolimnion.*

Nature of the bottom

In most cases the original bed of rock beneath the mass of water is overlaid with a stratum of sediment, the thickness of which can vary widely and the physical, chemical and biological characteristics of which are closely related to the processes of production of the sediment itself (deposition of inorganic particles transported by the water or of dead vegetable or animal organic material).

Suspended solids content

Surface waters often contain considerable quantities of finely divided particles in suspension. It is known that such particles can differ in origin and chemical composition and also in physical-chemical behaviour. The concentration of these materials in water can change within very wide limits either from one mass of water to another or within the same mass of water at different times or places.

pH *values*

The value of this parameter depends in the first place on the carbon dioxide content. In general the pH value is lower when the concentration of carbon dioxide is higher. The pH value is a particularly distinctive factor in the biological processes of an aquatic environment.

Dissolved oxygen

The oxygen balance in a mass of water involves two fundamental constituents.

a) The oxygen which is dissolved from the atmosphere (reaeration) and that evolved by the vital metabolism of aquatic plants containing chlorophyll.

b) The oxygen consumed by the aerobic biochemical activity which decomposes organic matter, and more generally the oxygen consumed by all the oxidizable substances present.

Considering that in metabolism carbon dioxide is the complementary parameter of oxygen, in the aquatic environments where biogenic phenomena occur, there is a well-defined inverse correlation between oxygen and carbon dioxide.

Salinity

The quantity and nature of substances dissolved in a body of water depend on various factors and in particular on the solvent power of the water, on the degree of solubility of the minerals of which the rocks are composed, on the duration and extent of contact with the rocks, and on the temperature. Evidently the salinity must also be considered in relation to possible domestic or industrial discharges which take place into the body of water in question. It will be shown in detail in section **4** that the dissolved substances may give rise to side-effects which interfere with the effluents.

Biotic communities

A biotic community is defined as the totality of the animals and plants which exist in a particular environment. In the present case it is necessary to consider those communities in the receiving environment which are able to intervene significantly in the transport and diffusion processes of the radioactive contamination. In practice one studies the communities which directly furnish food suitable for human consumption (fish, watercress, etc.) or else those aquatic plants etc. which although not edible can constitute an intermediate link in the food chain. In order to study the fate of discharged radionuclides it is necessary to determine both the quality and the quantity of the flora and fauna present.

3.2 — The Secondary Receiving Environment

As has been stated before, the propagation of radioactive contamination to man can also take place through the land environment (plants or animals), which is to be called the secondary receiving environment.

Its characteristics are determined by certain physical, chemical, and biological factors analogous with the case of the aquatic environment. Of these the abiotic factors (pedology, geology etc.) are however of secondary interest here in comparison with the biotic factors, in that the abiotic factors of agricultural land vary within fairly narrow limits. In consequence of the above, the principal parameter which characterizes the secondary receiving environment consists of the biotic communities (agricultural plants, cattle, etc.).

4 — PHENOMENA CAPABLE OF INFLUENCING THE DISTRIBUTION OF RADIONUCLIDES WITHIN THE RECEIVING ENVIRONMENT

The factors which have been listed and described in the previous two sections, as has already been mentioned, can enter into various reciprocal combinations, thus determining the manner of propagation of the radionuclides to man.

In this section the various phenomena which affect the radionuclides after discharge will be reviewed, mostly phenomena which occur in the receiving body of water, others in the corresponding terrestrial environment (animals or plants); they can be classified in the manner already shown in Table I.

Evidently not all the phenomena mentioned here are of equal importance everywhere, but form a spectrum of values which can be different from one case to another. Most often the principal phenomena are those connected with water movement, sedimentation and suspension of particulate substances, the absorption of radionuclides by materials present in the receiving body of water, uptake by plants and/or animals, and of course radioactive decay.

Nevertheless in principle one cannot exclude the possibility that because of some particular local situation a factor which is usually of secondary importance may become one of primary importance. This shows that it is desirable to obtain complete information on all the phenomena which can theoretically take place in the environment after the discharge; thus case by case one can fix attention on those which most influence the distribution and the subsequent routes of the radionuclides.

Another comment concerns the concrete possibility of appraising in numerical terms the phenomena of which we are speaking. It is easily seen that analytical information of such a type would allow one to proceed with the calculation of the concentrations of radionuclides in the various homogeneous components of the environment in a manner which is direct and scientifically rigorous. Nevertheless, although today there is a great mass of data and reports on the various phenomena capable of modifying the distribution of the radioactivity in the environment, the complexity and variability in time and space of such phenomena still do not allow one to make such a sophisticated calculation. But, apart from the obvious consideration that, with the development of methods of investigation and research, the direct quantitative evaluation of the above mentioned phenomena will become steadily easier, it must be said explicitly that for practical purposes, as will be shown in section 6, one can dispense with a detailed knowledge of each phenomenon and substitute for it a treatment of a more empirical type.

4.1 — The Physical Type of Phenomena

Transport and turbulent diffusion

Only transport by movement of water is considered here, that is that due to currents, excluding therefore the transport which is dependent on the movement of aquatic organisms which have ingested or adsorbed the radioactive substances. It is known that these organisms can effect a considerable migration both vertically and horizontally, influencing therefore to a certain degree the distribution of the contamination. Such an influence is not taken into consideration, however, since it is closely linked with the biological type of phenomena which will be examined later.

Transport by water movement affects radioactive substances whatever their initial chemical-physical state may be; it is generally accompanied by a turbulent diffusion which is both horizontal and vertical except in the somewhat improbable case of a laminar flow.

In the case of water-courses, turbulence almost always results from the friction with the banks, air resistance, wind action on the free surface of the water, collisions and deviations caused by the roughnesses of the river bed, by projections, direction changes etc.

In the case of lakes, the turbulent diffusion in the surface strata is associated with the movements of the water due to wave motion, to the currents, and to the eddies. It is assumed that the lakes that occur within the Community can all be considered to be of the temperate type; the waters of the surface strata are in free circulation because the movements require only small amounts of energy. In the thermocline and especially in the hypolimnion the movement of the dissolved or suspended substances is very much slower. In the epilimnion, the mixing to the horizontal diffusion and about a thousand times less to the vertical diffusion. In section 2 it has been mentioned that in general the vertical diffusion is also affected by any difference of temperature and/or density between the liquid wastes and the receiving waters.

Ionic or molecular diffusion

This effect takes place very slowly and is quite negligible compared with that of turbulent diffusion, being only a millionth or less of the latter. However, in very special cases (completely stagnant water) it could theoretically be the only factor affecting the dispersion of the radionuclides in the environment. In practice, however, it would be prohibited for a discharge to take place under such conditions.

Radioactive decay

The radionuclides contained in the liquid wastes have a physical half-life which may be anything from a few seconds to some thousands of years. Evidently, if the time that a certain radionuclide takes to travel through the pathways of propagation which bring it to man is one or more orders of magnitude greater than its half-life, then the radioactive decay can be in practice an important phenomenon and may be decisive for the purposes of the evaluation of the maximum quantity of that radionuclide for a particular receiving environment.

Sedimentation and suspension

It has been indicated that even the particulate pollutants or those associated with particles suspended in water can be transported or dispersed within a current according to the kinetic energy of the water and the size and weight of the pollutants themselves. The transporting capability of a water-course and more generally of a current is, as is well known, bigger for a greater velocity and density of water and for finer and lighter materials. If however the transporting capability of the current lessens, then the pollutants in suspension tend to form a sediment on the bed, undergoing the same fate as the rest of the detritus transported by the flow and clouding the water. In general therefore a clarification of the water is a sign of a purification as regards radioactive contamination but at the same time means a progressive accumulation of radioactive substances in the sediments, a circumstance which should not be underrated, having regard to the possible presence of detritus-eating fauna or certain aquatic flora or the use of dredged silt for land reclamation.

It must be remembered that the deposition on the bed can also be of a temporary nature in the sense that the water transporting capability, being dependent on variable factors such as flowrate and velocity, is itself variable and may therefore later be more than that needed to remove and maintain in movement the solids already settled (suspension). The final deposition of such solids will happen at a point farther downstream, depending on the conditions of the current to which the greater flowrate and velocity of the water correspond. The importance of this is obvious when one considers that the processes of sedimentation and suspension greatly influence the behaviour of the radioactive substances discharged into a body of water. In this regard however the literature* provides data which can be extremely useful.

The preceding considerations are substantially valid for moving water. For lakes, which generally behave like settling tanks, the particulate material reaches the bottom and remains there indefinitely, there being no renewal of the sediments as is brought about in rivers by the action of floods.

4.2 — The Chemical-Physical Type of Phenomena

Isotopic dilution

The presence in the receiving water of one or more stable isotopes of one of the radionuclides contained in the waste in the same chemical-physical state causes isotopic dilution and this in turn influences the biological fate of the contamination.

Ion exchange. Elution

It is well known that many natural materials possess ions or groups, mainly cationic, replaceable by other ions or groups. The natural ion exchangers are of either the organic or the inorganic type; the latter can be divided into two categories, clays and zeolites. From the point of view being considered here, the clays (montmorillonite, kaolinite, illite, chlorite) are most interesting, being very widespread in Nature and hence often present in the receiving water; but in certain geological situations, the zeolites (clinoptilolite, tufa etc.) can also have an important role.

If the radionuclides present in the wastes are cat ionic, they can exchange with the argillaceous and/or organic materials transported by moving water or otherwise present in the receiving body, for example in the sediments on the bottom or the banks. The chemical nature of the radionuclides has an influence in that the selectivity of the exchanging materials is greater for certain ions than for others, according to certain scales. For some important bivalent ions, the scale is

$$Ba^{++} > Sr^{++} > Ca^{++} > Mg^{++}$$

and for monovalent ions

$$Cs^{+} > Rb^{+} > K^{+} > Na^{+} > Li^{+}.$$

By the general laws which govern the equilibrium of exchange such scales are inverted, for solutions of suitable concentration, which in principle can allow the elution of exchanging materials and the return into solution of the radionuclides.

The characteristics of exchange that need to be known to try to forecast the variation in time and space of the contamination introduced into a particular hydrogeological environment are as usual those of the exchange capability, exchange kinetics, and the ability of any exchange material, present both in suspension and in the sediments on the bottom and on the banks, to remove from solution an ion present in trace amounts.

This last property is best expressed by means of the coefficient of distribution, defined as the ratio of the ion concentration in the solid phase (q) and that in the liquid phase (c) under equilibrium conditions. The exchange mechanism of a particular material with respect to a certain ion depends not only on the chemical nature of that ion, as previously stated, but also of course on the nature of that material, the particle size, the pH value, the concentration of other ions (especially those chemically similar to the absorbed ion) and also the time in contact. As with all absorption phenomena, ion exchange is also reversible; the absorbed ions can in theory become

---

(*) For example see E.C. Dapples, Basic Geology for Science and Engineering, John Wiley & Sons, Inc. (1959).

eluted, if for example, as was stated before, the salinity of the receiving water exceeds certain levels or if the pH value falls below certain values. In practice, however, such conditions are rarely realised except by the sudden introduction of industrial waste or water rich in fertilizers or other such cases.

Adsorption

The phenomenon of adsorption can occur between the radionuclides present in solution in the wastes and substances transported by the receiving water or contained in the sediments on the bottom or the banks.

The effect diminishes as the surface area of the material lessens and with an increase in temperature.

Carbon and silica are the substances for which the adsorption phenomena can occupy an important place in the phenomenological complex which determines the fate of the radionuclides introduced into an environment. Also metallic particles possibly discharged into the environment by metallurgical industries upstream can cause intense adsorption processes. Colloidal particles can also undergo adsorption phenomena, although in such a case the mechanism seems to be different from that for solutes.

The adsorption process is nearly always irreversible.

Coprecipitation

If the radionuclides are in a cationic form they are liable to coprecipitate as a result of flocculations in the receiving body of water, if for example this has a high iron or aluminium content and for some reason there has been a rise in the pH value. This process is very effective in regard to trivalent cations and those of higher valencies, and therefore to the alpha emitters and in particular to plutonium.

Plainly, other cases are possible: for example one can think of the possibility of effluent rich in phosphates (such as the effluent resulting from the brewing of beer) being poured into the receiving water upstream of the radioactive discharge. If the radioactive wastes originate from a water supply which is fairly hard and if they have a high pH value, then the meeting of the wastes and the receiving water precipitates the calcium as a basic phosphate. The precipitate has a flocculent consistency and acts as a carrier for many of radionuclides present in the liquid in a cationic form, but is not efficient as regards anions and radiocolloids (among the latter may be mentioned caesium, zirconium, scandium, ruthenium and plutonium). These two examples show, among other things, the importance of an accurate survey of the discharges of neighbouring industries.

Dissolution

In rare cases the radioactive waste may not be in aqueous solution but in liquids of various kinds, mainly organic, within which the radionuclides responsible for the contamination can exist in a particulate or colloidal state (or else in some way associated with material in such a chemical-physical state).

When these liquids are discharged into the external environment such particles sooner or later transfer from the medium in which they were initially contained to the receiving water, where they pass into an ionic or molecular state to the extent that they are soluble in water.

Dispersion

This phenomenon consists in the formation of sols from material in a coagulated state, and can happen spontaneously if suitable conditions occur. Such a case is more properly spoken of as peptisation. It can be supposed for instance that the presence of an electrolyte in the waste initially impedes the formation of a colloidal solution from coagulated material in the waste itself; after the discharge, the removal of the electrolyte consequent on dilution causes the dispersion, that is, the peptisation of such coagulates.

Coagulation

This phenomenon concerns the radionuclides present in the wastes in a colloidal state. It is known that it can happen spontaneously due to heating, concentration or the addition of electrolytes. A typical instance is what happens when a river runs into the sea; in such a case the electrolytic action of the sea-water facilitates the precipitation of the colloidal particles. Obviously the same phenomenon can occur if industrial effluent with a high temperature or high electrolyte content is introduced into the receiving water.

Chemical phenomena

Chemical phenomena can be of different kinds but in each they involve a chemical reaction. Thus, if the contamination is in ionic form one can have the formation of complexes with the ions in the receiving water and conversely, if the contamination is initially present in the form of a complex then this can dissociate or be destroyed. In both cases the substances responsible for the contamination, reacting with other substances present in the environment, can give rise to compounds which, if insoluble, are precipitated.

If the contamination is in a particulate or colloidal state or associated with materials which are initially in those states, then a mainly chemical reaction can take place, as a result for example of high alkalinity or acidity in the receiving environment, or the presence of large amounts of chlorides, or in some cases by an increase in electrical conductivity due to the presence of strongly ionized substances (strong acids or bases, organic or inorganic salts).

In lakes it can happen that in the deep layers where the particulate material is sedimented, very low pH values are reached as a consequence of the loss of oxygen by biological phenomena, associated with a lack of bicarbonates and therefore a lack of the buffering ability of the lake. It is also possible in lakes that, with a complete lack of oxygen in contact with the sediments, an aerobic processes assert themselves with the production of much hydrogen sulphide, a lowering of the redox potential, and proportionate dissolution of materials such as iron, manganese, etc.

4.3 — Biological Phenomena

Isometabolic dilution

As a result of the analogous behaviour displayed by certain elements when they take part in certain biological processes, a radionuclide can be diluted not only by the corresponding stable element (isotopic dilution) but also by stable elements which follow the same metabolic processes. Two well-known examples of isometabolic dilution are the dilution of the radioactive nuclides of strontium by calcium and the dilution of the radioactive nuclides of caesium by potassium. There is no need to stress the fact that the calcium-strontium couple is more important than the potassium-caesium couple from the point of view of isometabolic dilution, because of the closer analogy between the chemical behaviour of the elements concerned.

Ingestion by animals (including man)

As a result of their metabolism and feeding habits, it is known that certain kinds of animals can concentrate radionuclides in their bodies, often by many orders of magnitude.

If therefore such animals take part in a certain food chain they can furnish man with food with a high concentration of radioactivity. If not, they can exert an inhibiting action which slows the progress of the radionuclides in the environment.

Absorption by plants

The same considerations as were developed above for animals are valid for this phenomenon. It is necessary to add that root absorption is spoken of here; leaf uptake, partly because of the special characteristics of the process, is discussed in the following sub-section.

Surface adsorption by animals and plants

This form of intake can represent a significant contribution to the radioactive contamination of organisms, considered as a whole. As a rule surface adsorption is more important for plants than for animals.

In the case of irrigation by rain for example, leaf uptake can represent a critical pathway for the transfer of contamination for those radionuclides which, like ^{137}Cs, are retained by the soil or for which the time required for intake through the roots is too long compared with their short half-life (iodine-131 for example).

In the same way as has been mentioned before, this process may or may not reduce the total quantity of radionuclides available to man according as the organisms under consideration do not take part or take part in some human food chain.

Sedimentation of dead aquatic plants and animals

When aquatic animals and plants die, the radioactive contamination contained within them is transferred to the sediment on the bottom. In certain cases this process can be responsible for a considerable contribution to the accumulation of contamination within the sediments, for example when the life of the organisms is fairly short and their number high. The radionuclides in the sediments can pass back into solution and if the rate at which they do so is high enough an equilibrium can be established between the concentration in the sediment and that in the water.

None of these processes can be easily evaluated quantitatively but must be considered on the basis of the particular characteristics of each single environment.

Transport due to the migration of aquatic animals

The migration of certain animals, for example fish, can contribute, although not in great quantities, to the transport of radioactive contamination. Such a process can be disregarded in general because of the small quantities involved. Nevertheless it appears appropriate to mention it here, because in certain cases it can become the principal cause of the radioactive dispersion upstream of the discharge point or well downstream from it, even if there are considerable processes of deposition of contamination in the sediments on the bottom.

5 — MODEL OF THE RECEIVING ENVIRONMENT AND PROCESSES OF RADIONUCLIDE TRANSFER TO MAN

Figure 1 shows a receiving-environment model which has been worked out, taking into account the foregoing discussion, the necessity of having a general picture of the environmental distribution of the radioactive contamination, and also the criteria which will be better illustrated in the following section. In figure 1, it can be seen that the entire receiving environment (total system) is divided into three partial systems (or subsystems) corresponding respectively to

(i) W = the primary environment (aquatic),

(ii) P = the secondary environment (plants),

(iii) A = the secondary environment (animals).

Each of these comprises in its turn a certain number of *subdivisions*. In the sub-sections which follow, the subsystems with their respective subdivisions, and the processes on the basis of which can be derived the internal and external exposure of man to radiation, will be examined.

5.1 — The Primary Environment (Aquatic)

This consists principally of the receiving *body of water* and its sediments and also of the aquatic flora and fauna. Given then the possibility of the exchange of water between surface water and subterranean water, one considers a further subdivision represented by the adjacent aquifer(s). From the receiving body of wafer as such, man can undergo both external and internal exposure to radiation; if the water is used only for drinking or in food, then the internal hazard predominates over the external hazard, the latter being linked with an indirect use of the body of water such as navigation and recreational activities (bathing, water skiing etc.).

Another important subdivision of the primary environment consists, as has been said, of *the sediments* at the bottom of the body of water.

The risks associated with the areas in the vicinity of beaches or of materials originating from the sediments on the bottom (in dredging and filling) involve only an external exposure. The importance of the sediments is due also to the interchanges between them and the water and aquatic organisms. The transfer of the radioactive contamination from one to the other of these subdivisions can reach quite high values because of the concentration of the contamination in the sediments themselves.

The aquatic flora and fauna are the other two subdivisions taken into consideration, obviously on the hypothesis that the animals and/or plants present in the body of water are abundant and the species are edible.

Certain plants of the *aquatic flora* can be eaten directly by man (for example cress) and therefore can act as a source of internal irradiation. In any case the aquatic flora produce food for the successive trophic levels which make up the *aquatic fauna*. In their turn the edible species of these expose man to internal irradiation.

As was pointed out earlier, it cannot be claimed that the subsystems model adopted for the primary environment (aquatic) is complete in the sense of covering all cases describing every possible situation. Water can be processed to render it potable or more generally for the correction of certain of its characteristics; in such a case it is possible that one could have an accumulation of radioactivity at certain places (sludges, sand filters, ion exchange resins, etc.) with the consequent risk of external irradiation. Another example can be found in building constructions using sand or some other material from the banks or from the bottom, with a risk of exposure to radiation either for persons assigned to do the work or for persons living in an environment so constructed. The incineration of contaminated plants also can be responsible for the transfer to the atmosphere of the contamination in the form of an aerosol or fineley divided powder. A similar effect can occur in the proximity of waterfalls, caused by the dispersion of contaminated droplets.

The proposed model can, however, be applied in the majority of real cases. On the other hand it will be seen later, how, in those few cases where particular phenomena such as those just

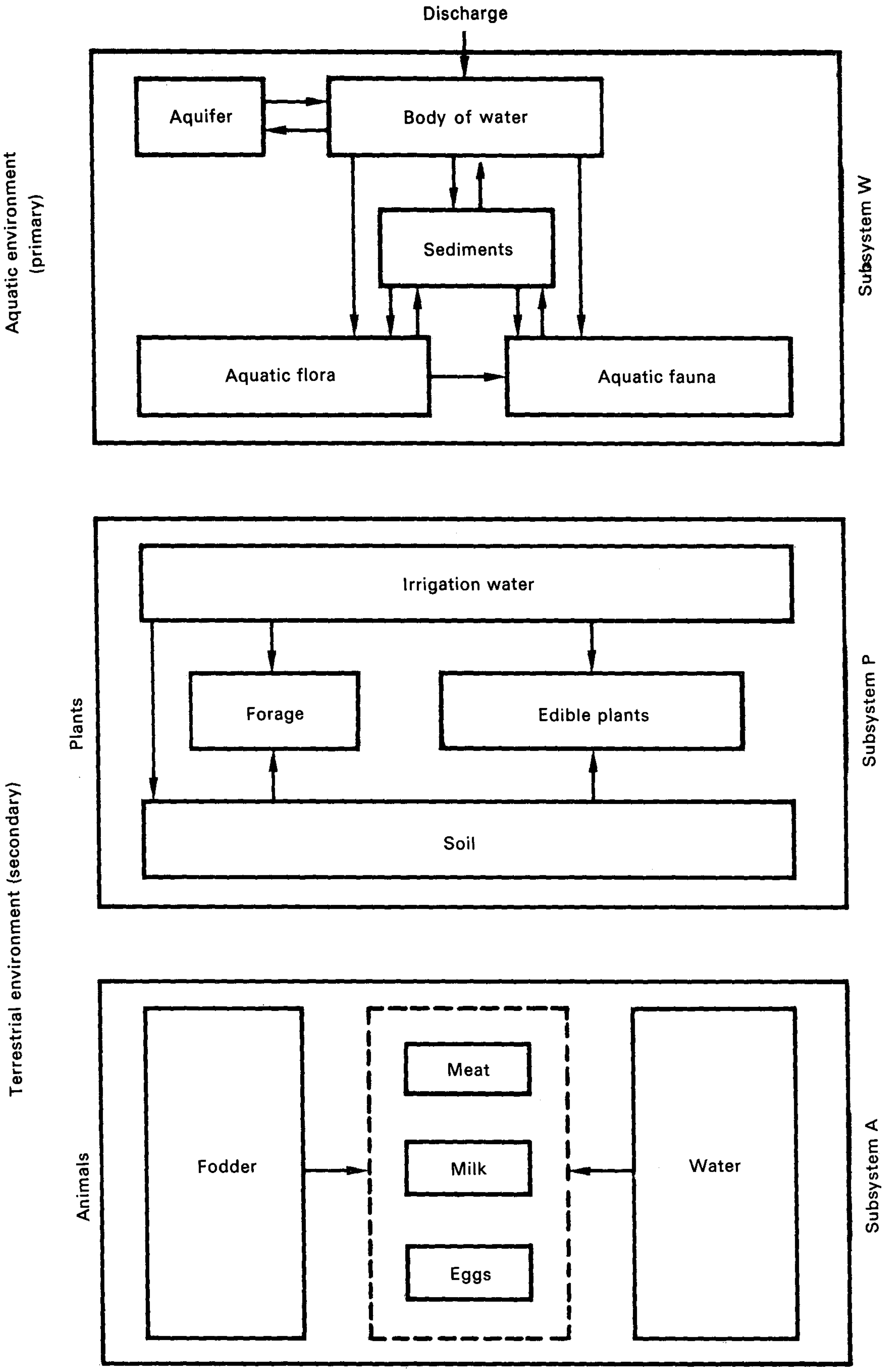


Fig. 1 — Environmental model.

6 — THE MATHEMATICAL SOLUTION OF THE PROBLEM OF RADIONUCLIDE DISTRIBUTION IN THE RECEIVING ENVIRONMENT

6.1 — Introduction

The preceding sections have given an idea of the wide variety of the phenomena which occur in the receiving environment and which affect the route of the radionuclides discharged into it. It will be understood therefore that the problem of obtaining a complete and precise quantitative knowledge of the distribution of such radionuclides in the environment cannot be without considerable complexity.

The result is that, in common practice, an extremely empirical principle is often followed; this consists of direct measurement, at the points concerned, of the concentrations of radionuclides introduced by experimental discharges into the environment or reaching the latter as a result of radioactive fall-out; or, sometimes, of the concentration of corresponding stable isotopes. Such a method is equivalent to the setting up of an empirical system of blocks, each of which describe the concentration of the considered radionuclide in a definite component of the system; however, it furnishes valid solutions only for conditions analogous to those prevailing at the moment of experimental observation and in general cannot be applied when conditions vary.

Between the strictly accurate method based on the analysis of all the phenomena which influence the exchange of the radioactive substances within the individual components of the environment, and the empirical method mentioned above, there is clearly a whole series of intermediate possibilities which furnish the better solutions the more detailed is the knowledge of the characteristics of the environment and the processes which take place in it.

In this section, the definition and mathematical solution of the problem being dealt with here are presented in terms of reasonable generalizations. It will be seen that the formulae obtained permit a numerical representation which becomes closer to reality with increasing numbers of parameters and quantities having known values. Thus one will approach the ideal limiting case in the most favourable situation, and the empirical limiting case in the most unfavourable situation.

For the mathematical treatment, we will apply the compartment system theory and it seems therefore desirable to begin with a brief summary of this theory.

6.2 — Brief Account of the Theory of compartment systems

The applications of compartment models in biology, pharmacology, nuclear medicine, radiological protection, and in other fields are well known. It is also well known that this has allowed results of great interest to be obtained, for example in the study of the metabolic functions of living organisms, dosimetric calculations etc. For a complete treatment of the subject the reader is referred to one of the numerous publications in the literature; here we shall merely review some fundamental concepts, mainly to give an idea of how a suitable mathematical model applied to the problem of the diffusion of radioactive substances in the environment can give a panoramic outline of the distribution of the radioactive material discharged from a nuclear installation.

A linear compartment model—as that utilized in the present paper—is a system which consists of ideal volumes containing specific substances which move from one volume to another according to laws of a Kinetics analogous to the Kinetics that describe diffusion phenomene. Each volume represents an ensemble of molecules which have the same probability of passing from the state in which they are into another possible state.

If understood in this way, each volume represents the equivalent of a chemical state or a localization or both. A compartment system is made up of an assembly of compartments variously connected among themselves, in which the substance which enters a certain compartment is wholly or partly released from another which is called the precursor of the considered compartment. A complete model is made up of as many compartments as there are homogeneous components of the system. In mathematical terms it is defined by a system of a correspondent number of differential equations. In practice, to simplify the mathematical development of the model and also to allow for the incomplete knowledge of the required parameters, the number of compartments is considerably reduced, provided that the simplified model may be adapted to the description and study of these phenomena concerned.

A model can be used both to describe the behaviour of extraneous substances introduced into a certain system, and to study effects involving the substances present in a system in dynamic equilibrium.

In the study of the diffusion of radioactive substances in the environment it is possible to refer both to chemical forms extraneous to the ecological system concerned, and to chemical forms already existing in it in a state of dynamic equilibrium.

It should be noted that when the environment has been contaminated by radioactivity, the high specific activity of most of the radionuclides ensures that the contamination never reaches chemical and/or physical saturation levels. The rare exceptions, represented by certain radionuclides with a low specific activity (e.g. 129 I), are of no practical interest from the point of view of health physics, and experience has shown that the long-lived radioelements often found in practice (uranium, ^{239}Pu, ^{226}Ra) can still be dealt with by means of a model using compartments.

It follows that the formulas discussed below have a general validity which is independent of the concentration of radioactive contamination.

Passing to the mathematical treatment of the problems, consider the compartment i, whose volume is V_i(*) is containing a quantity Q_i of a given substance. If this substance leaves the compartment with a velocity $K_i Q_i$, then the constant K_i which represents the fraction of the quantity Q_i of the substance eliminated in unit time is called the *constant of elimination or renewal* of compartment i. If the substance eliminated from compartment i penetrates into another compartment j, k_{ij} denotes the *transfer constant* from i to j. Evidently, if all the substance leaving compartment i passes into j then $k_{ij} = K_i$, otherwise $k_{ij} < K_i$.

When the compartment is not supplied from the outside, the variation in time of the quantity of the substance present in compartment i is described by the equation

$$\frac{dQ_i}{dt} = -K_i Q_i \tag{1}$$

for the concentration, by analogy, if the ideal distribution volume V_i remains constant, we have

$$\frac{dC_i}{dt} = -K_i \frac{Q_i}{V_i} = -K_i C_i . \tag{2}$$

Integrating equations (1) and (2) gives

$$Q_i(t) = Q_i(0)\, e^{-K_i t} \tag{3}$$

and

$$C_i(t) = \frac{Q_i(0)}{V_i}\, e^{-K_i t}. \tag{4}$$

Equations (3) and (4) describe the variation with respect to time of the quandity $Q_i(t)$ and of the

---

(*) The volume V_i represents the ideal volume of distribution. Such a value corresponds to the ratio of the quantity introduced in a short time into the isolated compartment, to the concentration of this substance extrapolated to zero time. Physically it corresponds to the actual volume in which the introduced substance is distributed, corrected if necessary for those factors which determine a particular concentration or dilution within the compartment itself.

concentration $C_i(t)$ in the compartment, isolated from the system, after the introduction into it of the quantity $Q_i(0)$ at time zero.

The function

$$e^{-K_i t}$$

is called the *characteristic retention function* of compartment *i*.

The product

$$K_i \, e^{-K_i t}$$

of the elimination constant and the retention function is called the *elimination function* of compartment *i*.

If the compartment is replenished by a precursor *h*, k_{hi} being the transfer constant from *h* to *i*, the product

$$k_{hi} \, e^{-K_i t}$$

constitutes what is called the *weighting function* from compartment *h* to compartment *i*.

Then calling *R* the quantity of substance that enters *i* in unit time, equation (1) becomes

$$\frac{dQ_i}{dt} = R - K_i Q_i \tag{5}$$

and equation (2) becomes

$$\frac{dC_i}{dt} = \frac{R}{V_i} - K_i C_i . \tag{6}$$

The mathematical formulation of a compartment model involves writing a differential equation of type (5) or (6) for each compartment, depending on the connections that the compartment has with those adjacent to it. The solution of the system of equations which results naturally depends on the knowledge of all the elimination and transfer constants which appear in the equations; this then gives the equations which describe the quantity of substance present as a function of time, or its concentration, in individual compartments.

The solution is relatively simple if the system is open (*) and if there is a continuous and constant introduction of substances into it, and if it is desired to know the distribution of the latter in the equilibrium state; in this case it is enough to equate the differential terms of the system to zero, consequently reducing it to a system of algebraic equations.

Under conditions of non-equilibrium the evaluation of the function increases in complexity with the increase in the number of compartments making up the system and the number of exchanges taking place between the compartments.

When the functions representing the compartments are linear and have a constant time-dependent behaviour, the solution of the system differential equations wich describe the model is essentially a series of successive convolutions of the weighted functions representing the separate compartments with the functions describing the time-dependent behaviour of the amount of material present in the respective precursors. In a complete ecological model the hypothesis of the linearity of the functions representing the compartments of may be accepted in most cases. The hypothesis of constant time-dependent behaviour is merely to be regarded as a working hypothesis, and the parameters should be selected with reference to the conditions which are the closest approximation to the formulated model. Similarly the choice of an equilibrium state study or a transitory state study will depend on specific phenomena. Whereas some phenomena reach a state of equilibrium within a few hours or days, others proceed more slowly and only reach a state of equilibrium after several months or years.

(*) Compartment systems are said to be open or closed according to whether or not substances can leave them.

Consequently, in the overall ecological system some phenomena can be more conveniently described with reference to equilibrium condition and others with reference to a transitory state, depending on the interval of time considered in the study.

Before passing to the application of distribution theory to the problem which is the object of this report, it is desirable to mention that in the consideration so far it has been assumed that the compartments are homogeneous, that is, characterized by retention, elimination and weighting functions which are single exponential functions. If, however, a certain component cannot be considered to be homogeneous then it can be assumed to have an empirical retention function described by a sum of exponential functions (or power functions) and such a compartment (heterogeneous) can be included in the system beside all the other homogeneous compartments, bearing in mind that the elimination functions are also sums of exponentials in which the distributions coefficients vary according the exit points from the compartment.

6.3 — Application of compartment theory to the Receiving-Environment Model

All the considerations and equations in the previous sub-section are obviously valid also in the case in which the substance in question is radioactive. The only difference between the behaviour of a radionuclide and a corresponding stable isotope that has the same chemical form is that due to radioactive decay; this can easily be corrected for by increasing the elimination constant by an amount equal to the decay constant.

It is obvious that here again the numerical solution of the system of equations which expresses the environmental model in mathematical terms is possible only if the values of all the elimination and transfer constants concerned are known, these being closely linked to and dependent on the various phenomena which occur in the environment after the discharge and which have been described in section 4. Here, without going into details which are beyond the scope of this report, it will be sufficient to say that the values of the above-mentioned constants can be obtained by different methods of enquiry. For example, since a transfer constant represents the fraction of the amount present in a compartment which undergoes a certain fate in unit time, it can be determined by measuring the variation with time of the absolute quantity of the substance or its concentration, or else by the evaluation of the exchange of particular carrier substances. Thus a knowledge of the average quantity of river water used daily in the irrigation of a field is useful in calculating the transfer constant of the radioactivity from the river water to the cultivated plants and so on.

In regard to the ecological model of the environment, to which the theory is to be applied, it is obvious that the description of the environment by means of a model is effected, as with all compartment models, by taking into account the type of information required to be obtained (annual average concentrations after continual discharge, or concentration integrals with respect to time for a pulse discharge etc.) and the extent of the study which it is intended to carry out (examination of a particular phenomenon or the whole group of phenomena). Naturally in the setting up of the model one cannot leave out of consideration the real possibilities of research into the various components of the system.

In this case, given that it is proposed to outline a general picture of the environmental distribution of the contamination, one can justify the adoption of the model (or complete system) shown in figure 1, which consists of three subsystems *W*, *P* and *A*. The first represents the primary ecological system, i.e. the aquatic environment; the second and third represent the secondary ecological system, i.e. the terrestrial environment of plants and animals respectively.

6.3.1 — *The subsystem W (the receiving, primary or aquatic environment)*

The subsystem W shown schematically in figure 2 is derived from the corresponding model in figure 1 with some slight simplifications (elimination of the pathway back to the sediments from the aquatic flora and fauna); this will be justified later on.

The physical significance and the dimensions of the various parameters used in figure 2 are described in table II, in which are also indicated the phenomena upon which such parameters depend.

In reality, the receiving environmental body of water can rarely be considered as one single compartment because of the unlikelihood of homogeneity over its whole extent. Mostly it is necessary to subdivide the receiving body into a certain number of tracts which can be held to be homogeneous and as a consequence to repeat in whole or part the application of the model of the corresponding subsystem for each tract.

The differential equation which expresses the concentration in each part of the body of water, considered by itself, i.e. without interaction with the other compartments in the system, is as follows:

$$\frac{\mathrm{d}C_1}{\mathrm{d}t} = \frac{R}{V_1} - (K_1+\lambda)\,C_1\,, \qquad (7)$$

where V_1 is the volume of the considered tract, K_1 is the elimination constant for the hydrodynamic transport effect, λ is the radioactive decay constant and the diffusion processes (*) and R represents the quantity of radioactive material introduced into the tract considered in unit time, a quantity that for the first tract corresponds obviously to the quantity discharged in unit time.

Under equilibrium conditions and in the absence of interactions with other compartments of the system, putting $\mathrm{d}C_1/\mathrm{d}t = 0$ one has as the solution of (7)

$$C_1 = \frac{R}{V_1(K_1+\lambda)}, \qquad (8)$$

from which, if $\lambda \ll K_1$, the well-known formula

$$C_1 = \frac{R}{V_1 Q_1} = \frac{R}{\text{flowrate}}$$

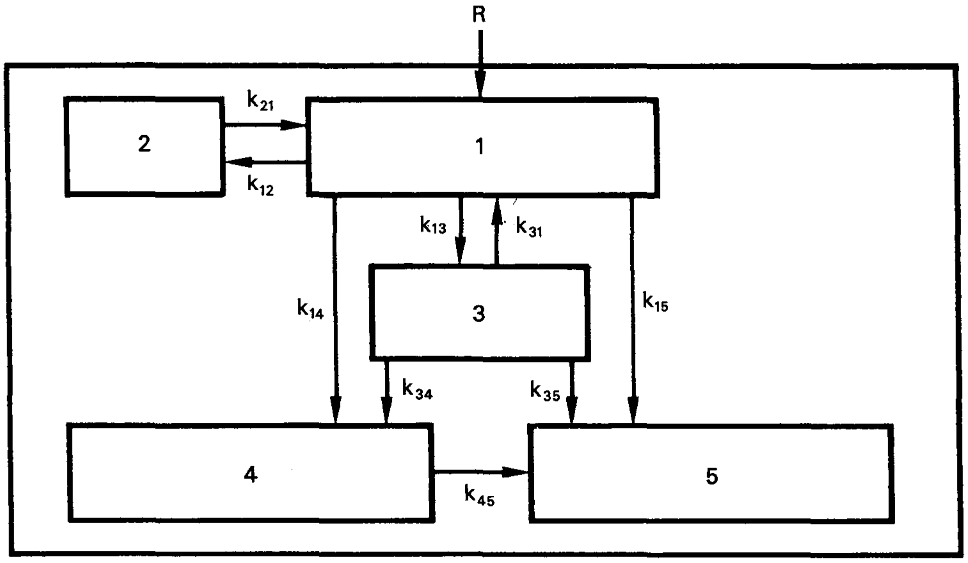
can be derived.

If however the discharge is instantaneous, and for all transitional states, the variation of the concentration as a function of time is obtained by repeatedly solving the differential equation given above, tract by tract, from the point of discharge to the tract concerned.

Turning now to the subsystem W as a whole, for the different tracts it is necessary to study first of all the assembly of compartments 1, 2 and 3, in which 2 corresponds to the aquifer, and 3 to the sediments. $k_{1,2}$ and $k_{2,1}$ are the parameters which characterize the exchange between 1 and 2, both having the dimensions of flowrate and corresponding to the volume of water which passes daily from the body of water to the aquifer and vice versa. It should be noted that such exchanges should also be taken into account when calculating K_1 and K_2. In fact, if in a certain tract the water-course receives a contribution of water from an aquifer which is not contaminated, then the radioactive material present in the water course undergoes a dilution which corresponds to an increase of the value of K_1. The parameters which characterize the exchange between 1 and 3 are k_{13} (in m^3/day), which is linked with the various processes of sedimentation, precipitation,

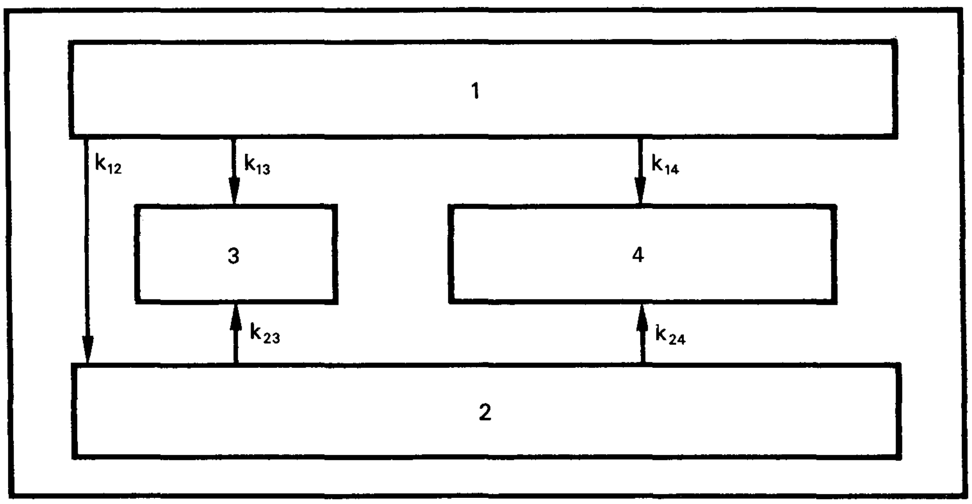
---

(*) When the most important process of renewal is dependent on the flowrate, as in water-courses, V_1, K_1 corresponds to the flowrate. In such a case, where the flowrate is known and the value of V_1 (the product of the average cross section of the water-course and the length of the part considered) is measured, the value of K_1 can be derived.



R	Discharge	3	Sediments
1	Body of water	4	Aquatic flora
2	Aquifer	5	Aquatic fauna

Fig. 2 — Diagram of the subsystem W; primary receiving environment (aquatic).



1	Irrigation water	3	Forage
2	Soil	4	Edible plants

Fig. 3 — Diagram of the subsystem P; secondary receiving environment (terrestrial, plants).

TABLE II

Characteristic parameters of the subsystem W (primary or aquatic receiving environment)

Category	Parameter	Dimension	Definition	Associated phenomena
Volumes and areas of distribution	V_1	m^3	Volume of the body of water considered.	—
	V_2	m^3	Volume of the aquifer considered.	—
	S_3	m^2	Area of the sediment below the body of water considered.	—
Transfer constants	k_{12}	m^3/day	Flowrate.	Passage of water from the body of water to the aquifer.
	k_{21}	m^3/day	Flowrate.	Passage of water from the aquifer to the body of water.
	k_{13}	m^3/day	Equivalent volume of water which contains the amount of radionuclide passing in one day into the sediment.	Sedimentation, ion exchange, adsorption, coprecipitation, coagulation, formation and precipitation of insoluble compounds.
	k_{31}	m^2/day	Equivalent area of sediment containing the amount of radionuclide passing in one day into the water.	Suspension, elution, dissolution, dispersion, chemical reaction.
	k_{14}	m^3/kg.day	Equivalent volume of water which contains the amount of radionuclide being taken up in one day by a kilogramme of aquatic plants.	Absorption and surface adsorption by aquatic plants, isotopic and/or isometabolic dilution.
	k_{15}	m^3/kg.day	Equivalent volume of water which contains the amount of radionuclide being taken up in one day by a kilogramme of aquatic animals.	Ingestion and surface adsorption by aquatic animals, isotopic and/or isometabolic dilution.
	k_{34}	m^2/kg.day	Equivalent area of sediment which contains the amount of radionuclide being taken up in one day by a kilogramme of aquatic plants.	Adsorption by roots of aquatic plants, isotopic and/or isometabolic dilution.
	k_{35}	m^2/kg.day	Equivalent area of sediment which contains the amount of radionuclide being taken up in one day by a kilogramme of aquatic animals.	Ingestion by detritus-eating fauna, isotopic and/or isometabolic dilution.
	k_{45}	day^{-1}	Mass of aquatic plants assimilated in one day by a kilogramme of aquatic animals.	Ingestion of aquatic flora by aquatic fauna, isotopic and/or isometabolic dilution.
Elimination or renewal constants	K_1	day^{-1}	Elimination constant from compartment 1.	All the phenomena associated with the transfer constants k_{13}, k_{14}, k_{15}, and k_{21} and also the hydrodynamic transport and turbulent diffusion of the body of water.
	K_2	day^{-1}	Elimination constant from compartment 2.	The passage of water from the aquifer to the body of water and also the transport and diffusion within the aquifer.
	K_3	day^{-1}	Elimination constant from compartment 3.	All the phenomena associated with the transfer constants k_{31}, k_{34} and k_{35} and also the possible removal of the sediment from the subsystem W
	K_4	day^{-1}	Elimination constant from compartment 4.	All the phenomena associated with the transfer constant k_{45} and also all the phenomena associated with the metabolism and renewal of the aquatic flora.
	K_5	day^{-1}	Elimination constant from compartment 5.	All the phenomena associated with the metabolism and renewal of the aquatic fauna.

coprecipitation, ion exchange etc. and on which the elimination constant K_1 is dependent; and k_{31} (m^2/day) which expresses the opposite processes (resuspension etc.). Other parameters which should be considered are:

a) the ideal distribution volume V_2(m^3) of the aquifer in the tract concerned,

b) the distribution area S_3(m^2) of the sediment,

c) the elimination constant K_3 (day^{-1}), which usually corresponds to k_{31}/S_3.

Then the differential equations which express the assembly of compartments 1,2 and 3 are

$$\frac{dC_1}{dt} = \frac{R + C_2 k_{21} + C_3 k_{31}}{V_1} - (K_1 + \lambda) C_1 , \tag{9}$$

$$\frac{dC_2}{dt} = \frac{C_1 k_{12}}{V_2} - (K_2 + \lambda) C_2 , \tag{10}$$

$$\frac{dC_3}{dt} = \frac{C_1 k_{13}}{S_3} - (K_3 + \lambda) C_3 ; \tag{11}$$

for equilibrium conditions:

$$\frac{dC_1}{dt} = \frac{dC_2}{dt} = \frac{dC_3}{dt} = 0 .$$

One has therefore a system of algebraic equations which are solved as follows:

$$C_1 = R \frac{V_2(K_2+\lambda)\, S_3(K_3+\lambda)}{V_1(K_1+\lambda)\, V_2(K_2+\lambda)\, S_3(K_3+\lambda) - k_{12}k_{21}\, S_3(K_3+\lambda) - k_{13}k_{31}\, V_2(K_2+\lambda)} , \tag{12}$$

$$C_2 = R \frac{k_{12}\, S_3(K_3+\lambda)}{V_1(K_1+\lambda)\, V_2(K_2+\lambda)\, S_3(K_3+\lambda) - k_{12}k_{21}\, S_3(K_3+\lambda) - k_{13}k_{31}\, V_2(K_2+\lambda)} , \tag{13}$$

$$C_3 = R \frac{k_{13}\, V_2(K_2+\lambda)}{V_1(K_1+\lambda)\, V_2(K_2+\lambda)\, S_3(K_3+\lambda) - k_{12}k_{21}\, S_3(K_3+\lambda) - k_{13}k_{31}\, V_2(K_2+\lambda)} . \tag{14}$$

For the transitional states, the solution of the system of differential equations (9)-(11) is usually much more complex. It is sufficient to be reminded for example that the function of input to a stretch of the water-course is calculated on the basis of the solution of the system effected for the stretch immediately upstream.

For this reason, unless a particular siduation lends itself to a simplified theoretical calculation, it is more convenient to give up trying to solve equations (9), (10) and (11) and instead use empirical formulae which express the relationship between the quantity and the conditions of discharge on the one hand and the change with time of the concentration in the stretches concerned on the other hand.

The choice of the numeric values of the parameters adopted in those formulae must be obviously is easily checked, by calculating the curve of change of concentration with time for a discharge of short duration and verifying the correspondence of the integral of such a curve with the ratio between the equilibrium concentration of the tract concerned and the rate of discharge (continuous and constant).

It is worth noting that an empirical function which has been well investigated for one radioelement can be adapted to an isotope of different half-life, provided that it is of the same chemical form; it is enough to multiply the function which is used to express the instantaneous discharge curve by $e^{(\lambda 1 - \lambda 2)t}$, λ_1 and λ_2 being the decay constants of the first and second isotopes respectively. It should also be noted that, because of the multiplicity of the phenomena which are involved in the propagation of the radioactive substances discharged into the environment, long-term equilibrium states can exist which are different from the short-term ones. This involves the necessity of carrying out a continuous survey of the environment and a periodic revision of the model adopted.

To complete the study of the subsystem W, the last two compartments of the model remain to be considered, i.e. aquatic flora and fauna, numbers 4 and 5 respectively. It is true that these compartments are of less importance in the contamination balance within the subsystem W but nevertheless they should be taken into account as pathways of propagation of contamination to man. In reality it is possible for the accumulation of contaminated skeletons of aquatic animals or of detritus of aquatic plants to bring about a progressive accumulation of contamination in the sediments such that subsequent resuspension may raise the equilibrium level after a long discharge period, or it may maintain a certain radioactive concentration in the body of water even after the discharge ceases. However, because such a phenomenon acts in parallel with the phenomena of sedimentation and resuspension one can avoid difficulties arising from the more complicated calculation, by taking these effects into account in the exchange between compartments 1 and 3. In studying compartments 4 and 5 it may be necessary to consider the body of water as a whole and relate the parameter C_1 to the average concentration in the water body itself.

The parameters used in the study of compartments 4 and 5 are therefore (see table II)

k_{14} (m^3/day kilogramme)
k_{34} (m^2/day kilogramme)
k_{15} (m^3/day kilogramme)
k_{35} (m^2/day kilogramme)
k_{45} (day^{-1})
K_4 (day^{-1})
K_5 (day^{-1})

If the phenomena connected with the deposition of contaminated organic material are accounted for as previously suggested in the exchanges between 1 and 3 and therefore in the transfer constants from 1 and 3 to 4 and 5, then subdivisions 4 and 5 in a state of equilibrium are described by

$$C_4 = \frac{C_1 k_{14} + C_3 k_{34}}{K_4 + \lambda}, \tag{15}$$

$$C_5 = \frac{C_1 k_{15} + C_3 k_{35} + C_4 k_{45}}{K_5 + \lambda}. \tag{16}$$

Equations (15) and (16) are obtained by solving the differential equations appropriate for compartments 4 and 5 (omitted here for simplicity) in an analogous manner to that which has been carried out for the other compartments.

It is interesting to notice that equations (15) and (16) can be put into relationship with the concentration factors of aquatic plants or animals. Since as a result of the examination of the exchanges between compartments 1, 2 and 3, for an equilibrium state one obtains

$$C_3 = \frac{C_1 k_{13}}{S_3 (K_3 + \lambda)}, \tag{17}$$

the concentration factor for a given aquatic plant (C.F.P. $= C_4/C_1$) corresponds to

$$\frac{C_4}{C_1} = \frac{k_{14}}{K_4 + \lambda} + \frac{k_{13} k_{34}}{S_3 (K_3 + \lambda)(K_4 + \lambda)} \tag{18}$$

and the concentration factor for a given aquatic animal (C.F.A $= C_5/C_1$) corresponds to

$$\frac{C_5}{C_1} = \frac{k_{15}}{K_5+\lambda} + \frac{k_{13}k_{35}}{S_3(K_3+\lambda)(K_4+\lambda)} + \frac{k_{14}k_{45}}{(K_4+\lambda)(K_5+\lambda)} + \frac{k_{13}k_{34}k_{45}}{S_3(K_3+\lambda)(K_4+\lambda)(K_5+\lambda)}. \tag{19}$$

Equations (18) and (19) immediately explain the variability of the concentration factors, which can be related to the physical-chemical conditions of the radionuclide in the body of water, to the characteristics of the sediments, and for aquatic animals to the plant species present in the body of water itself and finally to the decay constant of the radionuclide.

For the transient states, if $C_1(t)$ denotes the concentration of the radioelement in the body of water and $C_3(t)$ the concentration in the sediments, the concentrations at a time t in the components of the aquatic flora and fauna are given by the summation of the convolutions: (*)

$$C_4(t) = C_1(t)^* \, k_{14}\, \mathrm{e}^{-(K_4+\lambda)t} + C_3(t)^* \, k_{34}\, \mathrm{e}^{-(K_4+\lambda)t}, \tag{20}$$

$$C_5(t) = C_1(t)^* \, k_{15}\, \mathrm{e}^{-(K_5+\lambda)t} + C_3(t)^* \, k_{35}\, \mathrm{e}^{-(K_5+\lambda)t} + C_4(t)^* \, k_{45}\, \mathrm{e}^{-(K_5+\lambda)t}. \tag{21}$$

The formulation of these functions is, however, fairly complex because of the difficulty in writing valid functions for the concentrations $C_1(t)$ and $C_3(t)$, which refer to the mean values in the water body and the sediments respectively. Here also, therefore, it is sometimes more convenient to use empirical functions.

One has to remember, nevertheless, that the ratio of the temporal integrals of contamination between an organism and its precursor corresponds always to the value of the concentration factor.

6.3.2 — *The subsystem P (the secondary or terrestrial receiving environment: plants)*

The subsystem P outlined in figure 3 follows directly from the corresponding model in figure 1. Compartment 1 represents the point of entry into the subsystem. The function which expresses the concentration of radioactive materials in it with respect to time can easily be calculated on the basis of the variation of the concentrations in compartments 1 and 2 of the subsystem W. If for example the irrigation water is $\frac{3}{4}$ surface water and $\frac{1}{4}$ ground water and the concentration in the surface water is given by $f(t)$ and that in the ground water by $g(t)$, the concentration in the irrigation water $h(t) = 0.75 f(t) + 0.25\, g(t)$. (This calculation must also take into account the possibility of water-treatment capable of altering the radioactive concentration.)

The parameters used in the study of subsystem P (transfer and elimination constants) are listed and defined in table III.

It is to be noticed that the ratios k_{2i}/K_i represent the equilibrium transfer coefficients between a stable isotope in the plants and its surface density in the soil. The ratios $k_{2i}/(K_i+\lambda)$ are the transfer coefficients for a radioisotope with a decay constant λ.

The ratios k_{1i}/K_i and $k_{1i}/(K_i+\lambda)$ are the direct transfer coefficients for a stable isotope and for a radioisotope from water to plants.

Since compartments 3 and 4 are practically independent of each other and linked to 1 and 2 by transfer phenomena which can be considered irreversible, they can be studied independently both for equilibrium conditions and for transient conditions.

In equilibrium conditions, if the concentration in 1 is C_1, the concentration C_i in the plant i is given by

$$C_i = C_1\left(\frac{k_{12}}{K_2+\lambda} \cdot \frac{k_{2i}}{K_i+\lambda} + \frac{k_{1i}}{K_i+\lambda}\right). \tag{22}$$

(*) The convolution of $f(t)$ with $g(t)$ is

$$f(t)^* \, g(t) = \int_0^t f(\tau)\, g(t-\tau)\, \mathrm{d}\tau .$$

TABLE III

Characteristic parameters of the subsystem P (secondary or terrestrial plant receiving environment)

Category	Parameter	Dimension	Definition	Associated phenomena
Transfer constants	k_{12}	m^3/m^2 day	Volume of water used to irrigate 1 m^2 of soil per day. (*)	Deposition of radioactive contamination in the soil by irrigation water.
	k_{13}	m^3/kg.day	Equivalent volume of water which contains the amount of radionuclide being taken up in one day by a kilogramme of forage.	Direct (leaf) absorption of the radioactive contamination by forage.
	k_{14}	m^3/kg.day	Equivalent volume of water which contains the amount of radionuclide being taken up in one day by a kilogramme of edible plants.	Direct (leaf) absorption of the radoiactive contamination by edible plants.
	k_{23}	m^2/kg.day	Equivalent area of soil which contains the amount of radionuclide being taken up in one day by a kilogramme of forage.	Indirect (root) absorption of the radioactive contamination by forage.
	k_{24}	m^2/kg.day	Equivalent area of soil which contains the amount of radionuclide being taken up in one day by a kilogramme of edible plants.	Indirect (root) absorption of the radioactive contamination by edible plants.
Elimination or renewal constants	K_2	day^{-1}	Elimination constant for compartment 2.	Washing away, causing insolubility and removal of soil from the subsystem.
	K_3	day^{-1}	Elimination constant for compartment 3.	Metabolism and gathering of forage.
	K_4	day^{-1}	Elimination constant for compartment 4.	Metabolism and gathering of edible plants.

(*) Substract the equivalent volume of water whose capacity of radionuclide is absorbed by the vegetals.

These equations are also obtained by solving the differential equations for the compartments concerned.

For transient conditions, if $C_1(t)$ is the concentration in the irrigation water as a function of time, then the surface activity $C_2(t)$ in the soil at a time t is given by

$$C_2(t) = C_1(t)^* \, k_{12} \, e^{-(K_2+\lambda)t}, \tag{23}$$

and the function $C_i(t)$ which expresses with respect to time the state of the concentration of the radioactive substance as a function of time in the plant i is given by

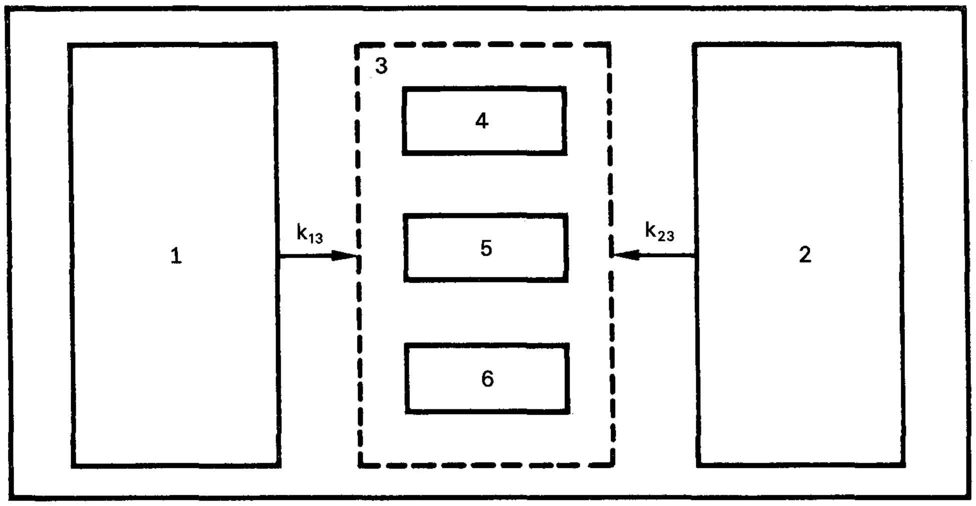
$$C_i(t) = C_1(t)^* \, k_{1i} \, e^{-(K_i+\lambda)t} + C_2(t)^* \, k_{2i} \, e^{-(K_i+\lambda)t}. \tag{24}$$

6.3.3 — *The subsystem A (the secondary or terrestrial receiving environment: animals)*

The subsystem A outlined in figure 4 likewise follows directly from the corresponding model of the ecological environment in figure 1. Compartments 1 and 2 represent the points of entry into the subsystem and are directly connected to compartment 3 of P and to 1 and 2 of W. The calculation of the functions used in the study of the subsystem A, which express the time dependence of the radioactivity concentrations in these last two compartments, is relatively simple and can be obtained with an analogous procedure to that discussed for the calculation of the concentration in compartment 1 of the subsystem P.

The parameters which need to be known for the study of A are those listed and defined in table IV. Given that here, as with the subsystem P, the exchanges between the various subdivisions are practically irreversible and there are no closed cycles, the mathematical solution of the problem does not present any particular difficulty. For example, in equilibrium conditions the concentration in meat C_4 is given by

$$C_4 = \frac{C_1 k_{13} + C_2 k_{23}}{M_4 (K_4 + \lambda)}. \tag{25}$$



1 Fodder and forage
2 Drinking water
3 Animals as a whole
4 Meat
5 Milk
6 Eggs

Fig. 4 — Diagram of the subsystem A; secondary receiving environment (terrestrial, animals).

In transient conditions one has

$$C_4(t) = C_1(t)^* \frac{k_{13}}{M_4} e^{-(K_4+\lambda)t} + C_2(t)^* \frac{k_{23}}{M_4} e^{-(K_4+\lambda)t}. \tag{26}$$

Milk and eggs have entirely analogous equations, with, instead of M_4 and K_4, in the case of milk the parameters V_5 and K_5 respectively and in the case of eggs f_6 and K_6. For eggs the results are expressed in activity per egg (*).

(*) As was explicitly pointed out earlier, the model of the receiving environment, which has up to now been described morphologically and its mathematical solution given, has a sufficiently extensive character to cover the greater part of the cases actually possible. In practice the model can be further simplified if some of the subdivisions taken into consideration here are not present in the environment or if their contribution to the solution of the system is negligible. Sometimes the opposite can happen and it may be necessary to extend the research into particular transfer phenomena, in which case there will be a need to formulate complementary models. For example, if it is intended to consider the contribution of animal manure to contamination, a model of the following type can be formulated.

1 = receiving water body
2 = soil
3 = forage
4 = animals
5 = manure

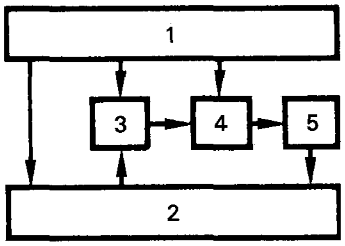


TABLE IV

Characteristic parameters of the subsystem A (secondary or terrestrial animal receiving environment)

Category	Parameter	Dimension	Definition	Associated phenomena
Volumes, masses and distribution factors	M_4	kg	Distribution mass of meat: body mass of the animal, corrected for the intestinal absorption factor and the factor for distribution in tissue.	Intestinal absorption of the contamination by animals, distribution in tissue.
	V_5	litre	Distribution volume of milk: volume the body fluid of the animal, corrected for the intestinal absorption factor and the distribution factor in milk.	Intestinal absorption of the contamination by animals, milk secretion.
	f_6	—	Fraction of ingested activity present in an egg, extrapolated to zero time.	Intestinal absorption of the contamination by animals, incorporation in eggs.
Transfer constants	k_{13}	kg/day	Mass of forage ingested per day by an animal.	Animal feeding.
	k_{23}	litre/day	Volume of water ingested per day by an animal.	Animal watering.
Elimination or renewal constants	K_4	day^{-1}	Elimination constant for compartment 4.	Attenuation in tissue as an effect of biological elimination by animals.
	K_5	day^{-1}	Elimination constant for compartment 5.	Attenuation in milk as an effect of biological elimination by animals.
	K_6	day^{-1}	Elimination constant for compartment 6.	Attenuation in eggs as an effect of biological elimination by animals.

6.3.4 — *Further considerations*

Once the compartment model has been formulated, the solution of equations such as (1) or (5) which express it can be found both for equilibrium and for transient conditions; we have seen that the solution is relatively simple in the first case, not differing substantially from that which can be obtained by the direct application of concentration and dilution factors to the successive links of the chain (*).

Nevertheless the formulation of a compartment model with the appropriate choice of the volumes and parameters is useful here also since it allows a more detailed description of the concentration and dilution factors to be obtained which in turn allows a better interpretation of the measurements, takes account of parameter variations for particular conditions and times, and allows the adaptation to the calculation for a radioelement of the results of measurements carried out on another isotope having a different decay constant.

It should be observed also that the study of the equilibrium conditions can be used advantageously to obtain useful information on the environmental contamination which results from a discharge effected over a short time. In a linear compartment model, following a unit injection at a given point in the system, the integral with respect to time of the concentration in any compartment is equal to the equilibrium concentration in the same compartment which occurs when at the same

---

(*) To facilitate the numerical solution of the problem, the procedure proposed by A. Rescigno and G. Segre (The kinetics of drugs and radioactive tracers, Boringhieri, Turin, 1961) can be very useful; it introduces the use of the operational calculus and the graph method.

point in the system the injection is continuous, constant and of unit value in unit time. Concentration and dilution factors derived for a state of equilibrium can also be correctly and usefully applied to conditions which are not those of equilibrium. Another consequence is that a continuous discharge can produce the same environmental contamination as an intermittent one, provided that the appropriate "rate" is chosen.

In particular the possibility can be considered of discharging the monthly output in a single monthly operation. This does not seem to give rise to any particular difficulties because the diffusion through the food chain tends to render the contamination more uniform in time. A flywheel effect exists which spreads in the successive links of the chain those peaks of contamination which occur at the discharge point as a result of injections of brief duration. This already happens in the receiving water body and is more marked in subsequent links of the chain. It follows that the highest peaks occur in the receiving water body in relation to the possibility of the use of the water for drinking purposes.

From a dosimetric point of view it is necessary to distinguish the longlived radioelements from those that are short-lived. For those that are long-lived, since the dose to the critical organ is reached after prolonged exposure, the effect of the ingestion of a given amount of activity does not differ substantially whether the ingestion takes place over a short time or over a fairly long period of time. The increase in the bodily accumulation can be considered to be practically the same in both cases. As a result of a discharge of short-lived radioelements effected over a short time there can be an increase in the dose rate which very unlikely will exceed three times the maximum permissible doses for occupationally exposed workers.

Being of a limited duration, such an excess may not be considered unacceptable, provided that the food reference levels take account of possible safety factors in the application of them to children.

Uranium is a special case because of its chemical toxicity.

7 — CRITICAL GROUPS; CRITICAL PATHWAYS; CRITICAL AREAS

Descriptions were given in the preceding sections of the characteristics of radioactive discharge and those of receiving environments, of the pathways of exposure to radionuclides as far as man, and of the general distribution model representative of the receiving environment. Finally, the formulae for the calculation of the concentration of every radionuclide in each of the subdivisions were derived. These formulae allow one to specify the critical elements of the environment (critical groups, critical pathways, critical areas), as will be now shown in detail.

As mentioned already in the introduction, the critical group refers to that group of the population most exposed to injury from radiation; the critical pathways are those by which this group can most easily suffer injury. Finally the critical area will be the geographical area involved in the radionuclide propagation phenomena along the critical pathways previously mentioned (*).

It is first of all necessary in each case to delimit the effective receiving environment and to adapt to it the general model described previously.

For this purpose we identify all the ecosystems (various kinds of plants cultivated, animals reared etc.) which are dependent directly or indirectly on the body of water into which the discharge is made. For this purpose a general knowledge of the overall character of the environmental situation will be sufficient.

Obviously, as already mentioned in section 6.3, not all the subdivisions of the general

---

(*) As has been pointed out in the Commission publication «Principles and general methods of establishing the limiting radiological capacity of a hydrobiological system», Brussels, October 1970, the term *critical* is used here not to introduce some idea of danger but to give a basis which will allow decisions to be taken in the field of health protection.

theoretical model will find in every case a corresponding element in the real receiving environment, so that in most cases there will be a simplification following the elimination of certain subdivisions.

Next, one seeks the numerical values to attribute to the various constants which affect the relationships listed in section 5 and relate to the interdependences between various subdivisions.

Some of these constants will be known or calculable experimentally whilst for others one will have to have recourse to values obtained on the basis of theoretical considerations or by extrapolation from analogous cases.

It should be repeated thad at present the accuracy of the quantitative knowledge of the processes which link the various subdivisions of the receiving environment is in general somewhat low. But it is likely that in future the amount of experience acquired in this field will allow an increasingly objective evaluation of the problem, thus reducing the uncertainty associated with the theoretical suppositions.

At this point the application of the formulae of the types (12)-(16) and (20)-(26) (possibly simplified to take account of the lack of one or another subdivision), in which one may put $R = 1$, will allow the calculation of the concentration of a single radionuclide in each of the subdivisions of which the system is composed, following a unit injection of each radionuclide.

The next step is to carry out a phytosociological survey in conjunction with a social-economic enquiry. A deep and analytical knowledge will be obtained both of the individual components of each ecosystem involved and of the composition and feeding and living habits of the populations. Such components can be identified in terrestrial environments by direct enquiry in situ or by using survey maps and information obtained from agricultural firms, syndicates etc. Thus a chart may be drawn of the environment involved, indicating the areas covered by various colonies and the forms of use of the body of water and the soil.

Such a survey will be concerned with the species which are present in such numbers and density as to allow practical and easy measurement of the radioactive contamination. For example, in cornfields, lucerne, various specialized cultivations, etc., the survey of the species will be limited specifically to the principal one and possibly some others (also easily evaluated) concerned from the point of view of a high concentration factor.

Besides the identification of the species, quantitative determinations (productivity) of each one will be necessary, distinguishing between the percentage destined for local consumption and that destined for markets outside the area. It will be necessary then to establish the distribution area, since, the more limited this is, the greater is the risk arising from any possible contamination, for the human population.

Analogous procedures should be followed for qualitative and quantitative surveys of livestock characteristics.

In certain cases where a component of the food chain causes a "bottleneck" in the propagation of radiocontamination, because it discriminates against the contamination itself for example, the study of the component must be particularly deep. On the other hand the opposite case must also be considered, that is, when a component of the food chain has a particularly high concentration factor. These data will be supplemented by an investigation into the prevailing consumption by the population involved and into its feeding and living habits. Qualitative and as far as possible quantitative surveys should be carried out analogously for aquatic environments on the animal and plant species present and the collective form and the interactions between the various species should be established.

Having determined the food consumption and more generally the living habits of the particular population groups related in any way to the receiving environment, the incorporations and/or exposures that result for each of such groups from a unit injection of a given radionuclide can be calculated on the basis of the previously determined concentrations of that radionuclide.

That group which turns out to be by far the highest among all the incorporations and exposures calculated is obviously the *critical* group for the radionuclide involved.

It is possible, even probable, that a value will not always be found which is clearly above all the others, or that more than one group will have a value of exposure or irradiation which is equally high, or is of the same order of magnitude. In such a case *critical groups* are referred to (for that particular radionuclide). It would be illusory to establish a sequence for values which were not clearly distinguishable one from another (that is, differing by at least an order of magnitude), having regard to the uncertainty associated with the evaluation of many of the parameters used to calculate these values. The critical group (or groups), once identified, will lead to an unequivocal determination of the corresponding *critical pathway*, i.e. that pathway, among all those possible, which contributes predominantly to the total exposure of the group. The same considerations as those developed for the identification of the critical group are valid also in this phase, and there may be more than one critical pathway, again with reference to a single radionuclide. Finally the *critical area* will be the geographical region across which each of the critical pathways mentioned above extends. From these definitions of critical pathway and area it is evident that they will be related not so much to the definition of the quantity of radioactive substances dischargeable (cf. section 8) as to the surveillance and control operations. These operations may be limited to those parts of the whole receiving environment in which the most important propagation phenomena of the radionuclides are located.

All the above refers to a single radionuclide, as has been stated repeatedly. Plainly, when it is foreseen that more than one radionuclide is to be discharged, it will be necessary to repeat the whole procedure described above for each of them. As a result of this, it could be found that different radionuclides result in critical groups which are completely different or which are partly or wholly coincident. Nevertheless, as will be seen in the following section, such differing possibilities have no effect on the derivation of the discharge formula, although they could extend the surveillance and control operations.

If neither edible flora nor edible fauna are present in the aquatic environment and if contaminated water is not used for irrigation downstream of the point at which the radioactive wastes are discharged, then it is not necessary to carry out an ecological investigation, and the maximum permissible concentrations for drinking water can be taken as the limiting factor.

8 — DISCHARGE LIMIT; DISCHARGE FORMULA

Considerations developed in the previous section have shown how in practice the critical group or groups can be identified for a particular radionuclide. Such a group has the maximum incorporation and or exposure following the injection into the environment of a unit discharge in unit time.

A simple ratio will now allow the maximum dischargeable quantity ($Q_{\max}$) to be calculated, such that the resultant incorporation and/or exposure is equal to the maximum allowed for a year by the regulations for protection from ionizing radiation:

$$Q_{\max} = \frac{\text{Annual maximum permissible incorporation}}{\text{Incorporation corresponding to } Q = 1}$$

or, in the case of external irradiation,

$$Q_{\max} = \frac{\text{Annual dose limit}}{\text{Dose corresponding to } Q = 1}.$$

The value $Q_{\max}$ is said to be the *discharge limit* for the radionuclide being considered. Clearly the condition which must be satisfied for the discharge of that single radionuclide will be

$$\frac{q}{Q_{\max}} \leqslant 1,$$

where q represents the quantity effectively discharged.

In the more general case in which there may be more than one radionuclide discharged, the above-mentioned condition obviously will be changed to

$$\sum_{r=1}^{n} \frac{q_r}{(Q_{\max})_r} = \frac{q_1}{(Q_{\max})_1} + \frac{q_2}{(Q_{\max})_2} + \ldots + \frac{q_n}{(Q_{\max})_n} \leqslant 1 .$$

This inequality represents the discharge formula for the receiving environment and the type of discharge which are being considered.

It is easily seen from the structure of this formula that this ensures the safety of each individual critical group in regard to all the radionuclides concerned in the discharge, even those for which another group may be critical. On the basis of the definition of the discharge limit, $(Q_{\max})_r$ represents the maximum quantity of a particular radionuclide which may be *continuously* discharged throughout the year. Consequently, the quantity effectively discharged q_r must also be understood to be continuously introduced into the environment. Nevertheless, on the basis of the considerations developed in section 6.3.4, it is possible to carry out instantaneous discharges of not more than one twelfth of $\sum_{r=1}^{n} q_r$ in a single monthly operation.

If several nuclear installations or centres are located within the same receiving environment, all the radionuclides associated with the waste from such installations are taken into account in the determination of the discharge formula. In this case the above mentioned formula is an overall one and for practical purposes needs to be divided into as many partial formulae as there are installations involved. To do this one must proceed to apportion the various discharge limits on the basis of the requirements of installations themselves.

Before concluding this work it is desirable to develop certain considerations on an important aspect of the whole problem. It has been seen from the beginning that the whole treatment has been carried out taking into account the fact that each radionuclide in an environment behaves in a manner which is dependent on its chemical characteristics, and therefore in general in a manner which is different from all the others. It is therefore not permissible to group chemically different radionuclides on the basis of their emission characteristics (for example, beta emitters).

Nevertheless indiscriminate measurements of radionuclides (for example, total betas) can still be usefully carried out for contamination levels which are much lower than those permissible, when all the contamination can be considered to be due to the most restrictive radionuclide. In this case, if the indiscriminate total measurement furnishes a lower result than that permitted for the most restrictive radionuclide, it will not be necessary to carry out any further measurement.

As the contamination level approximates to the maximum permitted, one must proceed to specific measurements, starting with the most restrictive nuclide and continuing in order of decreasing restrictivity.

When the contamination levels allow, it is an obvious advantage to be able to carry out indiscriminate total measurements.

NOTICE TO THE READER

All scientific and technical reports published by the Commission of the European Communities are announced in the monthly periodical **"euro-abstracts"**. For subscription (1 year : US$ 16.40, £ 6.17, Bfrs 820,–) or free specimen copies please write to :

Handelsblatt GmbH
"euro-abstracts"
D-4 Düsseldorf 1
Postfach 1102
Germany

or

Office for Official Publications
of the European Communities
P.O. Box 1003 - Luxembourg/Station
37, rue Glesener, Luxembourg

> To disseminate knowledge is to disseminate prosperity — I mean general prosperity and not individual riches — and with prosperity disappears the greater part of the evil which is our heritage from darker times.
>
> Alfred Nobel

SALES OFFICES

All reports published by the Commission of the European Communities are on sale at the offices listed below, at the prices given on the back of the front cover. When ordering, specify clearly the EUR number and the title of the report which are shown on the front cover.

OFFICE FOR OFFICIAL PUBLICATIONS OF THE EUROPEAN COMMUNITIES
P.O. Box 1003 - Luxembourg/station
37, rue Glesener, Luxembourg (Compte chèque postal Nº 191-90)

BELGIQUE — BELGIË
MONITEUR BELGE
Rue de Louvain, 40-42 - B-1000 Bruxelles
BELGISCH STAATSBLAD
Leuvenseweg 40-42 - B-1000 Brussel

DEUTSCHLAND
VERLAG BUNDESANZEIGER
Postfach 108 006 - D-5 Köln 1

FRANCE
SERVICE DE VENTE EN FRANCE
DES PUBLICATIONS DES
COMMUNAUTÉS EUROPÉENNES
rue Desaix, 26 - F-75 Paris 15e

ITALIA
LIBRERIA DELLO STATO
Piazza G. Verdi, 10 - I-00198 Roma

LUXEMBOURG
OFFICE DES
PUBLICATIONS OFFICIELLES DES
COMMUNAUTÉS EUROPÉENNES
Case Postale 1003 - Luxembourg/gare
37, rue Glesener - Luxembourg

NEDERLAND
STAATSDRUKKERIJ
en UITGEVERIJBEDRIJF
Christoffel Plantijnstraat - Den Haag

UNITED KINGDOM
H. M. STATIONERY OFFICE
P.O. Box 569 - London S.E.1

Commission of the
European Communities
D.G. XIII - C.I.D.
29, rue Aldringer
Luxembourg