

## **Assessment of doses to humans and biota from releases of radionuclides to the environment. (Lecture number EO-5).**

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### ***Abstract.***

It is an inevitable consequence of the production of electrical power from nuclear sources that there will be releases of radionuclides to the environment. Such releases are likely to arise from all stages of the nuclear fuel cycle - uranium mining, fuel manufacture, the operation of the power reactor, and the management of the spent fuel (including the reprocessing of the spent fuel and final disposal, if either is undertaken). A similar situation applies for any other activity employing radioactive materials. It is also inevitable that, if the releases are made to those sectors of the environment that are accessible to humans and the native wild biota, these will experience some (variable) degree of incremental radiation exposure. It is the primary objective of radioactive waste management that the incremental dose rates to humans from such releases should be as low as reasonably achievable and, in any event, below the levels that are prescribed in legislation. At the present time there are no internationally accepted dose rate criteria that can be applied to achieve an acceptable degree of protection for wild organisms. It is the current position, however, that if controls are applied to sufficiently protect humans, then the native biota are also likely to be adequately protected.

The generic conceptual framework for dose rate assessment and management is: radionuclide releases → environmental transfer → exposure pathways → dose rate estimation → comparison with standards → development and application of appropriate controls. This talk will focus on steps 2 through 4 of this process. For human activities, we are effectively dealing with point sources: a gaseous release to the atmosphere; a discharge of liquid effluent into surface waters; and, a surface or underground waste repository. All other factors being equal, it may be assumed that the highest dose rates, and the steepest gradients, are likely to be in the vicinity of the source. It is clear, therefore, that the dose rate assessment in the local area should be site specific and on spatial and temporal scales that relate to the human use of the site; further afield the assessment can be on a coarser scale and more generic. In the pre-operational phase of any facility employing radioactive materials (and in the consideration of potential accidental releases) it is necessary to have recourse to more or less elaborate models in order to assess the incremental radiation exposure likely to arise from the potential radionuclide releases. This is the case for both humans and the wild biota. A selection of the available models will be briefly presented to illustrate the developments that have been made. In the operational phase of a facility (and after an accidental release), it is possible to short-circuit the framework and obtain data on the actual distributions and concentrations of the radionuclides in the exposure pathways; these data have two uses: first, they can be used to validate and refine the models used to predict the environmental transfer of the radionuclides and their behaviour in the exposure pathways; second, and more importantly, they can be directly input into the dosimetry models.

It is very apparent, however, that the assessment models can be no better than our understanding of the radioecological processes governing the behaviour of the radionuclides in the environment and the uncertainty with which we are able to evaluate the parameters that define the models.

### ***Introduction.***

All human activities that utilise radioactive materials entail the possibility that there will be a production of radioactive wastes that will require management and/or disposal. In the case of a major activity, such as the production of electrical power from a nuclear-generated steam source, this possibility becomes a certainty at all stages of the fuel cycle - uranium mining, fuel manufacture, the operation of the power reactor, and the management of the spent fuel (including the reprocessing of the spent fuel and final disposal, if either is undertaken). Other activities that are also likely to generate radioactive wastes include the uses of radioactive materials in: R&D projects, medical diagnosis and therapy, the production of consumer goods, e.g., luminous watches and signs, smoke detectors, etc., etc. As is the case for any toxic agent, radioactive materials only become a health problem when the components of the biosphere become exposed to the emitted radiation, i.e., humans or the biota are

exposed to a radiation dose rate additional to that from the natural background. Depending on the management strategies adopted for the different classes of wastes, it is more or less likely that there will be releases of radioactive material to the accessible environment and that humans and the native biota will experience some (variable) incremental radiation exposure. The ICRP framework for the control of any consequential exposure arising from human activities involving radiation sources requires that the additional radiation dose rate to humans be as low as reasonably achievable and, in any event, below the levels that are prescribed in relevant legislation. At the present time, there are no internationally accepted criteria that can be applied to ensure an appropriate degree of protection for the wild flora and fauna in environments contaminated with radioactive materials. The current ICRP position is that if controls are applied to sufficiently protect humans, then the native biota are also likely to be adequately protected [ICRP, 1991].

***The management of radiation exposure.***

For the purpose of this paper, the radiation exposure is taken to arise from contamination of the environment due to the releases of radioactive material, including wastes, in the course of normal operations. If it is accepted that the radioactive waste management objective is to reduce the dose rates to a level that is as low as reasonably achievable, it is implicit that there is some balance between the efforts taken to reduce the radiation exposure, usually through the application of waste treatment procedures to reduce the releases of radionuclides, and the costs of achieving these reductions. It is also implicit that the consequential dose rate arising from the releases should be estimated so that this balance may be struck. The management framework to achieve these ends is given in Table 1. The iteration of this framework provides the basis for applying the ICRP system of radiological protection, i.e., the justification of a practice, the optimisation of protection, and the application of limits to the radiation dose and risk [ICRP, 1991]. The same general considerations would apply for an assessment of possible accidental releases, although the temporal and spatial scales would probably be different.

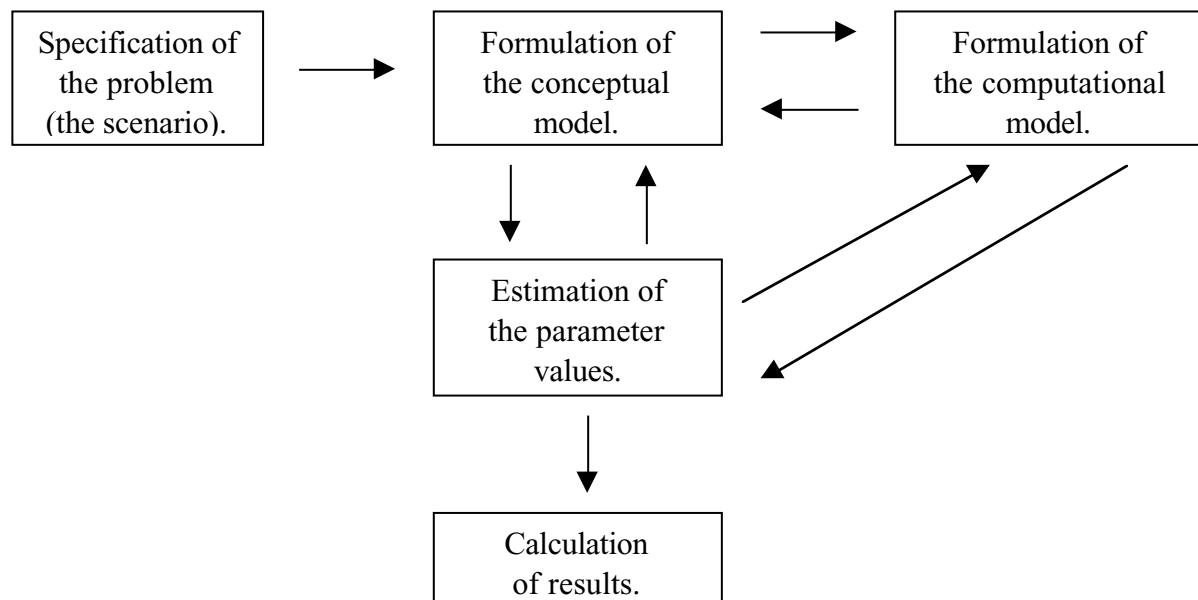
Table 1. A conceptual framework for the management of radioactive waste releases.

The management framework:	
• Source term, releases to:	atmosphere; surface waters; and, the ground.
• Environmental transfers.	
• Exposure pathways:	inhalation; ingestion; and, external exposure.
• Dose rate estimation.	
• Comparison with standards.	
• Application of appropriate controls	

Prior to the commencement of the practice, and the generation of any wastes, it is clear that the assessment of the consequential radiation exposure arising from any releases of radioactive materials to the environment will have to rely on some form of model. The designers of the facility will be able to provide information concerning the radionuclide source in terms of its physical and chemical composition, magnitude, and time dependence. These factors influence the spatial and temporal scales, and the complexity, of the environmental processes that need to be included in the models that are required for the assessment. It is clearly not sensible to include complex environmental processes in the model if there is no realistic prospect of either obtaining the relevant information for the site to parameterise the model or, more importantly, the probable effort required is not commensurate with the likely gain in the quality of the output. This is particularly the case for situations where simple, but robust and probably conservative, models indicate that the predicted dose rates are a small fraction of the relevant limits. There is also a purely scientific basis for limiting the spatial and temporal scales of an assessment model: it is important to remember that the activity of a given radionuclide is a discrete quantity, i.e., it is not infinitely divisible and that there is, therefore, a lower limit to the activity of each radionuclide that is physically meaningful. In principle, this lower limit corresponds to one atom of the radionuclide and is  $0.693/t$  Bq (where  $t$  is the radionuclide half-life in seconds). For example, this is equivalent to  $4.2 \times 10^{-9}$ ,  $7.3 \times 10^{-10}$  and  $1.0 \times 10^{-13}$  Bq of  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{239}\text{Pu}$ , respectively. Outputs that derive from activities (or activity concentrations) within primary model compartments that approach these values should be treated with some caution (see below). In practice, however, it is likely that these minimum activity values would be increased by a factor of, at least,  $10^4$  to give a 1% uncertainty in the value (on the assumption that Poisson statistics apply).

Models are required at steps 2 - 4 of the framework outlined in Table 1, and these will be the main focus of this paper. A framework for model development is outlined in Fig. 1 [IAEA, 1989], and there have been recent reviews of the status of modelling radionuclide transport and behaviour [Thiessen et al., 1999], and of the use of radioecological data in predictive models [Whicker et al., 1999].

Fig. 1. An outline framework for model development [IAEA, 1989].



### ***Environmental transfer of radionuclides.***

Human practices involving radionuclides lead, essentially, to point releases of active material to the environment. These may be gaseous releases to the atmosphere, discharges of dissolved or suspended particulate activity into a lake, river or coastal waters, and the disposal of packaged wastes into a surface or underground repository. All other factors being equal, the highest radionuclide concentration gradients in environmental materials are most likely to be in the immediate vicinity of the release point. It may be concluded, therefore, that the dose assessment in this local area should be site specific, i.e., it should take account of both the particular processes operating on the radionuclides in the area, together with their relevant parameter values, and the spatial and temporal scales of the human utilisation of the area. If there is a significant time-variability in the release profile and/or short-lived radionuclides are likely to be important contributors to the radiation exposure, then the model may need to be fully dynamic in nature rather than relying on empirical equilibrium representations of particular processes, e.g., bioaccumulation. This immediately makes for an increased demand for relevant input (radioecological) data on process rates to support the more complex models. At greater distances from the source, where concentration gradients are smaller, it is likely to be appropriate to use a more generic model to describe the dispersion of the activity, if for no other reason than that it would probably be impossible to obtain relevant process parameter values at small scales over large areas. The accumulation into the compartments leading directly to the radiation exposure, i.e., into foodstuffs for internal exposures and onto sediments and soils for external exposures, should, however, retain local site specificity. This latter, obviously, is also the case for the human habits that influence the magnitude of the exposures, i.e., the food consumption rates and occupancy rates. These considerations relate to the specification of the problem and are essential for the development of a realistic and credible assessment. In conclusion, it must be emphasised that any assessment model should be fit for purpose, i.e., it should not exclude, or poorly represent, any process that is known or suspected of having an important influence on radionuclide behaviour; equally, it should not attempt to include detail that is not relevant to either the spatial or temporal scale of the assessment.

Following the release of radionuclides into the air, surface waters or ground waters, the immediate concern is to provide a realistic description of the advection and dispersion of the activity in the local area taking due account of the processes that abstract the radionuclides from the primary transport medium. The development of the conceptual model entails the identification of all the processes that might influence the behaviour of the radionuclides, together with the corresponding environmental compartments and their interconnections. The outcome, however, is that a number of the processes will be excluded from the final model, either because detailed data do not exist, or because several processes can be subsumed within a single empirical parameter value, or because scientific understanding of the process indicates that it is of little ultimate significance in the context of the defined scenario. The conceptual models that have been used for releases to the atmosphere and to surface waters are given in

Fig. 2 and 3 [IAEA, 1982]. For a surface or underground waste repository, the release would be to groundwater; the radionuclides would then be transported by the groundwater flow with hold-up by sorption onto the rocks and/or soil, and would eventually either intersect the plant root zone, or be released into surface waters. The radionuclides would then become available at the corresponding points in Fig. 2 and 3, respectively.

The basic conceptual models for the assessment of the dose rates to the flora and fauna are very similar to those for humans (see Fig. 4 and 5) but differ in detail in the contamination and exposure pathways, but particularly so in the dosimetry models.

*Atmospheric dispersion and deposition of radionuclides.*

The radionuclides released to the atmosphere are at the mercy of the weather. This simple fact, together with our experience of weather forecasting, immediately demonstrates that it is very unlikely that it will be possible to make accurate predictions of the actual behaviour of any given short-term release more than a few days in advance. Although physically-realistic models based on solutions of the diffusion-transport equation are available, these are both data-hungry and demanding of computing power. In the medium term, the best that is likely to be achieved is to apply the historic, average wind speed and direction, atmospheric stability and rainfall data appropriate to the season of the release, and obtain an indication of the most probable behaviour. For the more usual situation of a continuous release, it is possible to make simplifying assumptions and apply these same historic meteorological data, possibly in Monte Carlo simulations, to predict the cumulative behaviour around the release point. The most widely utilised approach is the semi-empirical Gaussian plume model with parameter values that are simply derived from the historic meteorological information. In the simplest version of this model, the radionuclides are dispersed by the mean wind speed and turbulent diffusion in the atmospheric boundary layer; it has been further developed to take account of the influences of buoyant plume rise, building-induced turbulence, topographic variability, and the specific wind conditions prevalent at coastal sites (see [IAEA, 1982; EC, 1995] for more detailed descriptions). For both humans and the flora and fauna, the distribution of the dispersed radionuclides in the air column is the first source of radiation exposure. The data to determine the human exposure from the active cloud are given in [IAEA, 1982; EC, 1995] and it remains to be determined whether they can be adapted for plants and animals.

In addition to radioactive decay in transit, there are two other loss mechanisms operating on the radionuclides in the plume: wet deposition and dry deposition. These processes serve to both reduce the activity in (and the dose rate from) the plume at down-stream points, and input the activity to the vegetation and soil compartments. Wet deposition is a consequence of two processes: washout due to rain falling through the plume, and rainout due to the incorporation of the activity in the plume into cloud condensation droplets, i.e., the cloud is developing within the plume. In practice, it is difficult to separate these processes and they are combined into a single (empirical) washout coefficient that gives the proportion of the activity in the plume that is removed by rainfall in unit time. More complex models that take account of the stochastic nature of the rainfall patterns in the plume transit path have been developed [EC, 1995]. Dry deposition is a similarly complex process by which activity is removed from the underside of the plume where it interacts with the ground surface or

vegetation. Again, it is modelled by an empirical deposition velocity that relates the deposition rate ( $\text{Bq m}^{-2} \text{s}^{-1}$ ) to the activity concentration in air ( $\text{Bq m}^{-3}$ ) at the ground surface.

A limited selection of the results obtained with the basic generic model is given in [EC, 1995] and there is some discussion of the validation of the model. In general, it appears that the simple Gaussian plume model provide results that are within an order of magnitude of either the observations made at specific sites, or the results obtained with the more complex, physically-based models. For the latter, it was again concluded that it is difficult to obtain all the required input data to execute the model in a satisfactory way. It was also noted that the likely accuracy of predictions obtained using the Gaussian model improved as the duration of the release increased, with annual average values within a factor of two.

*The dispersion of radionuclides released into coastal waters.*

The earliest attempts to predict the behaviour of releases of radioactive waste into coastal waters relied on the use of dye tracer experiments to estimate the dilution of the input by tidal flows and its transport by the currents induced by the wind [Seligman, 1956]. This approach is, however, restricted to the vicinity of the discharge point and the recognized transfer of radionuclides from local to regional scales by advective processes led to the development of box models. Due to the use that has been made of the sea for the disposal of low level, liquid, radioactive effluents, there are many examples of these models in the literature [e.g., Clark *et al.*, 1980; Gurbutt *et al.*, 1988; EC, 1995 (this latter reference also includes the modelling of rivers and estuaries)]. The amount of a released radionuclide is assumed to be instantaneously and uniformly distributed throughout the receiving box and the transport processes between the boxes are simply described by coefficients that transfer a proportion of the box inventory into each of the neighbouring boxes per unit time. As the available computational resources have increased, the number of boxes included within the model domain has tended to increase [e.g., Abril and Garcia Le n, 1992]. The known uptake of radionuclides onto sediments has been included in the models with one, or more sediment boxes underlying each of the water column compartments; this uptake is usually modelled as an equilibrium system with constant distribution coefficients ( $k_d$ ). A particular problem with particle-reactive radionuclides (e.g., Pu and Am) is the significant proportion of the inventory that is associated with either suspended particulate material or settled sediments. This has led to attempts to develop more realistic (or mechanistic) descriptions of both the uptake of the radionuclides by the particles [e.g., Abril and Garcia Le n, 1993] and the movement of the contaminated sediment [e.g., Gurbutt *et al.*, 1988].

To address the lack of realism in the parameterization of the majority of box models, especially the aggregation of several processes into a single empirical coefficient, attempts have been made to develop, and apply, models that are based on a more detailed understanding of the underlying physics [e.g., Aldridge, 1998]. This includes a 2- (optionally, 3-) dimensional hydrodynamic description of the tidal- and wind-induced flows; a wind-wave model to provide the wave-induced bed stress that controls the behaviour of the suspended and settled sediments; and, a physically-based transport model to simulate the movement of both the dissolved and particle-bound radionuclides. As was the case for the implementation of the diffusion-transport equation for atmospheric releases, these more complex models are very demanding of both input data and computing resources.

In practice, the more complex, but more realistic, models have mainly been employed to simulate radionuclide behaviour on short time scales in the local to regional areas, whereas the box models have been applied to the prediction of behaviour on all scales. The most recent example of this differential application has been to the potential future releases of radionuclides from solid wastes dumped in Arctic waters [IAEA, 1998]. The use of the hydrodynamic circulation models to simulate the past behaviour of releases can utilise real wind data series to drive the models; forecasts the behaviour of future releases are, however, constrained, as in the case of the atmospheric diffusion-transport models, by the necessity to employ time-averaged, historic wind data. Where long-term data series are both available and considered to be statistically representative of the real distribution of conditions, it is possible that forecasts could be generated by probabilistic techniques [Aldridge, 1998].

Models of radionuclide dispersion in coastal waters have been calibrated with data for non-radioactive tracers, e.g., the conservation of salinity [Abril and Garcia Le n, 1992], or with data for one radionuclide and then applied to another, e.g.,  $^{137}\text{Cs}$  and  $^{239+240}\text{Pu}$ , respectively [Gurbutt *et al.*, 1988]. Comparisons of model output (usually annual averages) with relevant environmental observations of radionuclide distributions (usually obtained on a single Research Vessel cruise of limited duration) generally show broad agreement and are frequently within a factor of five [Abril and Garcia Le n, 1992; Gurbutt *et al.*, 1988; EC, 1995; Aldridge, 1998].

### ***Radionuclide exposure pathways.***

The dispersion models briefly described in the previous section provide information on the environmental distributions of radionuclides in air, water and sediments ( $\text{Bq m}^{-3}$  or  $\text{Bq kg}^{-1}$ ) and deposition densities onto soil or vegetation ( $\text{Bq m}^{-2}$ ). These values are both the source terms for the estimation of external exposures and the input terms for the modelling of pathways that lead to internal exposure - the inhalation of contaminated air, and the ingestion of contaminated water and food.

#### *External exposure from contaminated air.*

The radiation exposure from a plume of contaminated air depends on the uniformity and extent of the contaminated volume (governed by the distance from the emission point and the meteorological conditions) and the energy of the radiations. Data are given in [EC,1995] for three situations:

for  $\gamma$ -rays with energy less than 20 keV, the plume size under almost all conditions is such that it may be approximated by a semi-infinite, uniformly contaminated volume. In this situation, the absorbed energy in a small volume of air at any point is half the energy emitted in the volume.

for  $\gamma$ -rays with energy greater than 20 keV, a finite cloud model must be used that takes account of the radionuclide distribution in the cloud around the point of interest, and the absorption, scattering and energy build-up processes in operation.

for  $\beta$ -particles, the cloud is always effectively infinite, and the dose rate at the surface of the body is half that in the air.

It is immediately apparent that there is a number of, more or less complex, sub-models that are required to provide the estimates of the dose rate, and that these are subsumed in the numerical values tabulated in the report [EC, 1995].

#### *External exposure from contaminated ground.*

The EC report [EC, 1995] also includes the data required to estimate the exposure from radionuclides deposited on the ground. The model assumes that the radionuclides penetrate to a depth of 30 cm in an undisturbed soil by natural processes. The  $\gamma$ -radiation dose rate at a height of 1 m above the soil surface is then estimated for this contaminated layer taking account of the radiation attenuation and scattering, and energy build-up in both the soil and air. The  $\beta$ -particle source is assumed to be the radionuclide contamination at the soil-air interface due to the limited range of the radiation in soil. The results from the application of these sub-models are tabulated in [EC, 1995] for a number of radionuclides.

#### *Internal exposure from the inhalation of contaminated air.*



There are two possible sources of radionuclides for the inhalation pathway - those present in the contaminated plume, and those that have been deposited by wet or dry processes to the ground and are then subject to resuspension into the air by natural processes (predominantly, the wind) or human activities (e.g., ploughing). In either case, the resuspension is likely to be episodic and require information on occupancy factors to estimate the time-integrated exposure to the radionuclides in the air. The direct plume pathway is not treated further in [EC, 1995] simply because it is the outcome of the product of the one year-integrated exposure to activity in the plume, the breathing rate, and the inhalation dose coefficient provided by the ICRP [ICRP, 1996] on the basis of a model of the human respiratory tract [ICRP, 1994].

Resuspension of deposited radionuclides into the air from the soil surface is a more complex process and may result in the activation of the inhalation pathway, and lead to the contamination of crops and grazing animals (the ingestion pathway). No mechanistic model is available to describe the wind-driven resuspension process and three possible empirical approaches are considered in [EC, 1995]. The approach adopted employs a time-dependent resuspension factor ( $k \text{ m}^{-1}$ ) that relates the concentration in air due to resuspension ( $\text{Bq m}^{-3}$ ) to the surface deposit ( $\text{Bq m}^{-2}$ ). The time-dependency arises from the weathering of the deposit and the model was calibrated with data obtained from wind tunnel experiments. An alternative approach was used for the resuspension due to human activities. A dust loading of  $10 \text{ mgm m}^{-3}$  was assumed to have the same radionuclide concentration as the surface soil. Once the resuspended radionuclide concentration in the air has been determined, the assessment of the radiation exposure follows that of the contaminated plume. In practice, an analysis has shown that it is unlikely that the resuspension pathway will be significant as compared with the direct inhalation from exposure to the contaminated plume [EC, 1995].

*Internal exposure from contaminated food and water.*

Terrestrial foodstuffs.

The contamination of terrestrial plant and animal foodstuffs from the deposition of radionuclides from the air is, in fact, considerably more complex than is implied by the simplified conceptual model given in Fig. 2, i.e., the single routes for transfer may actually be multiple, there may also be some recycling between compartments, and each of the generic soil, vegetation and animal compartments includes a range of examples. There is a large number of models representing terrestrial foodchains - some are discussed in [EC, 1995] and others are available from the literature (e.g., the PATHWAY model [Whicker and Kirchner, 1987]). These models differ in the complexity with which they represent the environmental processes but the primary contrast is whether they are equilibrium or time-dependent in nature. The former use equilibrium factors to determine the relationships between the activity concentrations in the various components of the models, and are relatively simple to apply with the large supporting databases that are available [e.g., IAEA, 1982; EC 1995]. Time-averaging is implicit in these models and they are most appropriately applied to an assessment of the consequences of fairly uniform and continuous releases. Fully dynamic (time-dependent) models, most usually of the compartmental type, are necessary to make assessments either of potential accidents, or for the case of variable release or deposition rates, or to allow for seasonality in the processes governing the transfer of radionuclides in the foodchain. The expanding complexity of these dynamic models makes increasing demands for

relevant input data to parameterize the various processes that could be included. In practice, a degree of simplification is applied at two levels. First, sub-models have been developed to describe the processes influencing radionuclide behaviour in each of the three main compartments, i.e., migration in the soils, transfers to plants, and transfers to animals: this segmentation eases the computations, makes the assessments somewhat more transparent, and allows for modifications of a regional or site-specific nature to be included. Second, there is amalgamation of the wide range of foods that are consumed into a more limited set of generic types that represent the major groupings - green vegetables, grains, root crops, meat, liver, milk and eggs; the animals are: cattle, sheep, goats, pigs and chickens. Even so, the models remain complex (e.g., the metabolic models required to describe the uptake of radionuclides into meat and milk), and it has not been possible to develop fully dynamic models for all aspects of these foodchains [EC, 1995]. This reference also provides a full description of the models, the input data necessary to apply them (at least, generically), and some sample outputs.

Once the models have generated the (time-dependent) concentrations of the radionuclides in the foodstuffs, it is a simple matter to apply the relevant consumption rates and the ingestion dose coefficients [ICRP, 1996] to assess the radiation dose rate. It should be noted, however, that obtaining the consumption data relevant for particular areas likely to be contaminated by a release may not be simple particularly if the local agricultural production can be variably dispersed into the general food supply system, and that the metabolic and dosimetry models underlying the dose coefficients are anything but simple.

#### Aquatic foodstuffs.

The approach adopted for the majority of assessments of radionuclide contamination in aquatic systems is to apply equilibrium concentration factor values to the computed time-dependent radionuclide concentrations in the water to obtain the radionuclide concentrations in aquatic foodstuffs [IAEA, 1982, 1985; EC, 1995]. For freshwater systems, however, there is a growing trend towards the development of more complex time-dependent models of bioaccumulation that include the transfer of the radionuclides along the phytoplankton - zooplankton - fish foodchain [e.g., Garnier-Laplace *et al.*, 1997; IAEA, 2000]. As in the previous examples, this development substantially increases the demand for data. The driving force behind these attempts to provide mechanistic models of environmental processes is to improve the quality of the predictions. That this might not be as straightforward as it appears can be demonstrated by reference to a study of the accumulation of plutonium by the edible winkle (the marine gastropod, *Littorina littorea* L.), an important source of exposure for the discharges to sea from Sellafield [Swift and Pentreath, 1988]. Data from environmental samples had shown that the concentration factor for  $^{239+240}\text{Pu}$  in the winkle was variable whether it was based on the concentration of soluble Pu in the water (CF = 1500 - 9200) or the total Pu concentration (soluble + particle sorbed, CF = 75 - 1200). A laboratory study showed that the direct uptake from water was a minor contributor to the body burden of Pu and that intake of seaweed contaminated from water alone would only account for the lower end of the observed range at high feeding rates. It was found necessary to add a small quantity of contaminated silt to the weed in order to account for the observed environmental concentration factors at reasonable feeding rates. Given the evident complexity (and stochasticity) of the processes at work, it may be questioned whether any improvement in predictive ability could be gained. The mechanistic model approach may, however, provide a

necessary input for making the next step towards probabilistic assessments (see [IAEA, 1989]).

Due to the importance of assessments of the dose rates arising from discharges to coastal waters, a considerable effort has been devoted to developing the methodology for determining the consumption rates of marine foodstuffs (fish, crustaceans, molluscs and seaweeds) and the identification of critical groups [Preston and Jefferies, 1969; Preston *et al.*, 1974; Hunt *et al.*, 1982; Leonard and Hunt, 1985]. Together with the computed radionuclide concentrations in the foodstuffs and the ingestion dose coefficients [ICRP, 1996], these site specific habits data give the basis for an estimation of the dose rates from the contaminated pathways.

For the dose assessments of both terrestrial and aquatic pathways, the transfer between compartments employs either equilibrium, or time-dependent, transfer factors, concentration factors and distribution coefficients ( $k_d$ ). It is appropriate to raise again the point made above relating to the limiting values for radionuclide concentrations (i.e., those having real physical meaning) in environmental compartments. At these low (and possibly physically unrealistic) concentrations in the source compartment, the implicit multiplicative process can lead, at least in principle, to the apparent generation of radionuclides in the recipient compartments. This is a situation in which it would be appropriate to apply either radionuclide, activity or mass balance controls to ensure that the simulation is valid.

Water.

As compared with the assessment of the dose rates arising from contaminated foodstuffs, that relating to the intake of potable water from contaminated surface water sources is relatively straightforward. Once the radionuclide concentration in the lake or river from either direct deposition or run-off has been modelled, it only requires the application of a factor to allow for any losses during treatment, the standard consumption rate and the ingestion dose coefficient.

There is a number of additional pathways that are usually considered for completeness - water used for irrigation, aquatic plants used as soil conditioners or fertilizers, sea spray in the coastal zone, either via inhalation or by deposition onto agricultural land, and inadvertent ingestion of contaminated soil or sediment. The relative significance of these pathways in a given situation can only be determined by a habits survey.

In respect of radionuclide transfers in the environment and possible exposure pathways, continuing development in one area can be envisaged for the future, i.e., the combination of the complex, physically realistic transfer models, used in probabilistic mode, together with Geographical Information Systems (GIS), to identify radiologically vulnerable areas in the assessment of the consequences of potential accidents [Howard, in press].

### ***Dosimetry models.***

Human.

The inhalation and ingestion dose coefficients provided by the ICRP [ICRP, 1996] represent the culmination of almost 50 years of development of the underlying dosimetry models (a project that is still continuing) and it is not intended to examine them in detail here (a listing of the ICRP publications, including those directly related to the dosimetry models, may be collated from [ICRP, 1991, 1996]). It is important to note that the dosimetry models

are generic in nature, originally for a standard (later, reference) man or woman with defined anatomies and physiologies [e.g., ICRP, 1975], but now including babies (age 3 months), infants (1 and 5 years) and juveniles (10 and 15 years) [ICRP, 1996 and the supporting documents].

One of the important determinants of the dose rate from ingested radionuclides is the proportion (expressed by the parameter  $f_1$ ) that is absorbed from the gut into the body fluids and redistributed around the body. The values of  $f_1$  employed by the ICRP in their dosimetry models have been defined on the basis of both human and animal data for a variety of chemical forms. The latter may not, however, necessarily be representative of the chemical species present in contaminated foodstuffs. At a time when there were proposals to increase the value of  $1 \times 10^{-4}$  for the  $f_1$  for plutonium in food by a factor of 5 unless the lower value could be justified [NRPB, 1984], it was decided to determine a site specific value for the marine foodstuffs of concern in relation to the northeast Irish Sea [Hunt *et al.*, 1986]. This experimental determination of the  $f_1$  values made use of human volunteers consuming shellfish (edible winkles) contaminated with plutonium and americium under natural conditions in the northeast Irish Sea. The results provided evidence that the existing (lower) value of  $f_1$  for plutonium would be appropriate for this site and that the existing value for americium ( $5 \times 10^{-4}$ ) was conservative. A second study, incorporating and re-interpreting these earlier data, concluded that an  $f_1$  value of  $2 \times 10^{-4}$  would be appropriate for both elements [Hunt *et al.*, 1990]. More recent studies have shown that the  $f_1$  value for  $^{210}\text{Po}$ , an important internal source of natural background exposure from seafood consumption, could be increased to 0.8 from the ICRP-recommended value of 0.1, [Hunt and Allington, 1993], and provided data in reasonable conformity with the recommended ICRP values for the absorption of plutonium, americium, cobalt, caesium and technetium following the consumption of naturally-contaminated cockles (the bivalve mollusc, *Cerastoderma edule*) [Hunt, G.J., 1998].

#### Flora and fauna.

The radiation exposure of the flora and fauna arising from radioactive waste management practices has only recently begun to attract widespread attention. This has primarily been a consequence of the default acceptance by the regulatory authorities of the statements from the ICRP, the latest of which is [ICRP, 1991]:

*The Commission believes that the standard of environmental control needed to protect man to the degree currently thought desirable will ensure that other species are not put at risk. Occasionally, individual members of non-human species might be harmed, but not to the extent of endangering whole species or creating imbalance between species.*

Because the ICRP presented no evidence in support of these statements, they have come under pressure [e.g., Thompson, 1988], and there has been increasing interest in providing a more transparent basis for assessing the possible effects of increased radiation exposure on native populations of wild organisms, including the development of appropriate dosimetry models [IAEA, 1992].

Again, due to the use that has been made of the sea for liquid waste disposal, there has been greater progress in the development of dosimetry models for aquatic organisms [e.g., IAEA, 1976, 1979, 1988, 1992; NCRP, 1991; Pentreath and Woodhead, 1988] than has been the case for the terrestrial environment [e.g., IAEA, 1992; Amiro, 1997; Amiro and Zach,

1993]. Because there is a very wide range of habitats, plants and animals and very few could be considered in detail, the necessity, if not the requirement, to employ reference, generic organisms as the basis of the dosimetry models is even greater than is the case for humans. The primary objective has been to determine the broad range of dose rates that might be received by aquatic organisms with varying sizes, capacities to accumulate radionuclides, and habitats. A set of general selection criteria is summarized in [Woodhead, 1993], and other animals have been added as the need arises [e.g., IAEA, 1998]. Given that it is unlikely that there will be any detailed information on the internal distributions of the radionuclides in the majority of cases, it has been usual to assume that the internal activity is uniformly distributed within a simple geometry (e.g., a sphere or ellipsoid) that is taken to represent the body of the organism. The accumulation of the radionuclides by the organisms (for internal sources) and sediments (for the external exposure) have been determined by the equilibrium concentration factors and  $k_d$  using the concentrations in the seawater ( $\text{Bq m}^{-3}$ ) computed from a dispersion model as the basic input data. The details of the dose calculation methods are given in [IAEA, 1979] where the illustrative examples relate to the exposure of developing fish eggs. Where information is available to take account of the internal distributions of the radionuclides, the methods may be extended appropriately [IAEA, 1979; Pentreath and Woodhead, 1988].

In the terrestrial environment, generic organisms and exposure conditions have similarly been employed [Amiro, 1997]. In this situation, the basic input data from the dispersion or deposition model would be the radionuclide concentration in air ( $\text{Bq m}^{-3}$ ) or the deposition density ( $\text{Bq m}^{-2}$ ); for some terrestrial organisms it might be necessary to go one step further and estimate the radionuclide migration into the surface layer of the soil ( $\text{Bq kg}^{-1}$ ) as the input data for the dosimetry model.

#### *The use of the dose rate estimates.*

##### Humans

It should be emphasized that the output of any assessment model is only as good as the information on the environmental processes (including the way in which the environment is structured within the model) and the parameter values that are included. It is to be expected, therefore, that the predictions from different models are likely to be different; this is particularly the case when different modelling approaches are employed, e.g., compartment models as compared with hydrodynamic models for marine situations. Recent examples of the ranges of predicted dose rates from a variety of models assessing the same source term are included in [IAEA, 1998, 2000]. Detailed discussions of the sources of the variation are given and underline the point made in [IAEA, 1998] that any comparisons of assessment results from different models must include an analysis of the model structures, assumptions and databases in order to avoid a misinterpretation of the significance of any differences in the predicted radiation exposures. This again emphasizes that it is important that the model selected for a particular assessment should be fit for purpose. The assessment of the releases from the solid wastes dumped in the Arctic seas is notable for the inclusion of a caveat regarding the possible radiological significance of low dose rates ( $10^{-16}$  -  $10^{-19}$   $\text{Sv a}^{-1}$ ), produced as the output of assessment models, that correspond to no more than a few radionuclide decays in the human body in the normal lifetime [IAEA, 1998] (see also below).

This point may be particularly important for the assessment of collective doses for which the interpretation of small dose to large number of people remains an issue.

In the case of human exposures, the assessed dose rates per unit discharge or release can, by comparison with the relevant dose limits, provide the basis for setting regulatory limits on the total radionuclide release rate. Once the authorized releases are in progress, monitoring data may be employed to validate the different components of the overall model, provide site specific information concerning model parameters and, thus, lead to the possibility of improvements in the reliability of the model. At this time, however, an assessment of the actual dose rates from the external and ingestion pathways is most likely to be based on direct measurements of the dose rate in air in the contaminated area, and of the concentrations of radionuclides in the relevant foodstuffs. The use of site specific occupancy and food consumption data together the ICRP dose coefficients (the one remaining input from a model) then allows the estimation of the committed effective dose rate. The data on the actual release rates and the dose rate estimates allow the compliance with the regulatory standards to be assessed [e.g., MAFF/SEPA, 1999].

The validity of the dose assessment procedure employing monitoring data, consumption information from habits surveys and the ICRP dose coefficient has been investigated for the  $^{137}\text{Cs}$  present in the seafood pathway arising from the northeast Irish Sea [Hunt *et al.*, 1989]. A number of adult, mainly high rate, seafood consumers (identified from the habits survey) volunteered to undergo whole body counting at quarterly intervals over a period of about 18 months. Monitoring data on the  $^{137}\text{Cs}$  concentrations in the various seafoods from two local sources and consumption data from the habits survey, periodic logs of consumption over two week intervals and the responses to questionnaires were used as a basis of estimating the intake of the radionuclide. The body burden of  $^{137}\text{Cs}$  was estimated on the basis of the metabolic model underlying the ICRP dose coefficient; it was also estimated on the basis of the whole body measurements. A comparison of the values showed that the latter were almost always less than the former (only one value was greater) with a mean ratio of  $0.3 \pm 0.3$  (s.d.). Thus, the conventional (dose) assessment approach is shown to be conservative, and there is little evidence that the caesium metabolic model would underestimate the body content of  $^{137}\text{Cs}$  and, therefore, the dose rate.

Flora and fauna.

At the present time, there are no dose rate limits that have been accepted as providing for the protection of the environment although values have been suggested as appropriate for particular situations [e.g., IAEA, 1976, 1988, 1992; NCRP, 1991; UNSCEAR, 1996]. There are several problems in the context of the protection of the environment from radiation that need to be resolved:

For the environment, is it the individual or the population that should be the focus of protective action?

What is (are) the relevant radiation effect(s) that should be considered for native wild organisms? In the case of humans, ethical considerations point to the stochastic effects in individual persons, primarily cancer induction and heritable genetic damage, with deterministic effects being of much lesser concern except in certain occupational settings. For individual native wild organisms, or their populations, are stochastic or deterministic effects, or both, of concern?

What are the degrees of effect, in individuals or populations, that might be considered acceptable in order to define limiting dose rates?

What are the relevant targets from the point of view of dosimetry?

Of course, these problems have already been the subject of some discussion in the references noted above, and elsewhere [e.g., Pentreath, 1998], but the move towards an internationally acceptable position is only just beginning. Dose rate assessments for the environment have, therefore, had to be considered on an *ad hoc* basis.

One of the more thoroughly studied situations is the case of the plaice (*Pleuronectes platessa*, a bottom-living flatfish) in the northeast Irish Sea. Before there were any discharges to the sea from the Windscale/Sellafield site, it was recognised that the waste radionuclides would be a source of possible hazard for the fish but it was concluded that the risk of significant effects would be minimal [Dunster, 1952]. By the mid-60s, monitoring data showed that there was a significant build-up of radionuclides in the seabed in the northeast Irish Sea, and the question of possible effects on the (commercial) plaice population was again raised. Estimates of the radiation exposure of the plaice showed that it might experience dose rates approaching  $100 \mu\text{Gy h}^{-1}$ , primarily from the underlying sediment source. The assessed dose rates were sufficiently high over a large enough area that it was possible to consider an experimental determination of the actual exposure of the fish with thermoluminescent dosimeters (LiF). Nearly 3600 fish were tagged with the dosimeters and over 1000 were recaptured through normal commercial fishing operations during a three year period. The results showed that a few fish might experience long-term average exposures up to  $25 \mu\text{Gy h}^{-1}$  and that the mean dose rate for the population was of the order of  $3.5 \mu\text{Gy h}^{-1}$  [Woodhead, 1973]. These data provided reasonable confirmation of the original assessment of the radiation exposure derived on the basis of a simple dosimetry model.

A second study assessed the radiation exposure of developing plaice eggs using laboratory data on radionuclide accumulation and distribution on, and in, the developing eggs, environmental data on the concentrations of radionuclides in the water of the spawning area and a dosimetry model. For the  $\beta$ - and  $\gamma$ -emitting fission product radionuclides the exposure was assessed to be less than that from the natural  $^{40}\text{K}$  present in the seawater and in the eggs ( $8 \times 10^{-4}$  as compared with  $7 \times 10^{-3} \mu\text{Gy h}^{-1}$ ) [Woodhead, 1970]. The exposure from the  $\alpha$ -emitting  $^{239+240}\text{Pu}$  was also assessed at  $9 \times 10^{-4}$  or  $5 \times 10^{-3} \mu\text{Gy h}^{-1}$  depending on which of two uptake curves obtained in a laboratory study was applied to the environmental data. Although these values are again less than the exposure from the  $^{40}\text{K}$ , this comparison can be shown to be superficial. The estimate of  $9 \times 10^{-4} \mu\text{Gy h}^{-1}$  for the  $\alpha$ -radiation dose rate derives from a  $^{239+240}\text{Pu}$  concentration of  $0.037 \text{ Bq l}^{-1}$  in the seawater, a time-averaged concentration factor of  $2.4 \text{ l kg}^{-1}$  over the 17 day development period of the egg, and, therefore, a  $^{239+240}\text{Pu}$  concentration in the egg of  $0.089 \text{ Bq kg}^{-1}$ . The plaice egg is, however, small -  $\sim 4.3 \text{ mg}$  wet weight - and this means the amount of  $^{239+240}\text{Pu}$  per egg is  $4.1 \times 10^{-7} \text{ Bq}$ , i.e., there is one Pu atom disintegration on a plaice egg every  $2.4 \times 10^6$  seconds or  $\sim 28$  days, on average. Because the development period is  $\sim 17$  days it may be concluded that the assessed dose rate has no meaning. In fact, applying Poisson statistics, 74% of the eggs will not experience the passage of an  $\alpha$ -particle into the interior of the egg during the development period, i.e., there is no radiation dose rate from the Pu; 22% and 4% of the eggs experience the passage of 1 and 2+  $\alpha$ -particles, respectively. This again demonstrates that great care is required in the interpretation of low dose rates [Hetherington *et al.*, 1976].

The final study relates to the effects of chronic, low-level  $\gamma$ -irradiation on the plaice [Knowles, 1999]. Male fish were exposed to dose rates of 240, 500 and 1200  $\mu\text{Gy h}^{-1}$  for a period of 197 days leading up to the normal spawning time. The conditions of temperature and day length for the irradiated and control tanks paralleled those in the natural environment to encourage normal spermatogenesis. When normalised for body weight, there was a clear, dose rate dependent reduction in testis weight at the end of the irradiation period. At the lowest dose rate, histological examination of the testes showed that there had been a significant reduction in mature sperm production and non-germinal tissue; at the two higher dose rates, the numbers of spermatocytes were also reduced. Due to the small proportions of spermatogonia in the testes at sacrifice, it was not possible to detect any effects on the stem cells. Overall, it was concluded that the plaice testis had a similar radiosensitivity as had been determined for mammals. Although the clear effect at 240  $\mu\text{Gy h}^{-1}$  is at a dose rate that is greater than that which now obtains in the northeast Irish Sea, it has significantly narrowed this difference for an effect on a biological function in an individual that also has relevance at the population level. A model to investigate the possible links between small changes in the reproductive capacity of individual fish due to irradiation and the consequent response, if any, at the population level is now under study [Woodhead, unpublished]. Of course, radiation is not unique in its capacity to affect reproductive capacity, nor is it likely to be the only anthropogenic stressor present in the coastal waters near Sellafield. Equally, the plaice is only one of the species, each irradiated to a greater or lesser degree, comprising the local aquatic community. The overall environmental response to the incremental radiation exposure is, therefore, likely to be more complex than any simple model would predict.



### ***Conclusions.***

This brief, and by no means exhaustive, review of radiation dose assessment models for humans, and the native wild organisms in contaminated environments, permits a number of general conclusions to be drawn:

- the assessment models in any given instance should be fit for purpose, i.e., they should be commensurate with the scale of the problem in terms of the quantities of radionuclides likely to be released and their half-lives; and, our understanding of, and the available information concerning, the complexity of the environmental processes in the receiving environment; the correct balance also has to be struck both between generic and site specific assessments, and between equilibrium and dynamic models;
- the assessment models should include the process of radionuclide, activity or mass balancing, to ensure that the model operations are proceeding in a realistic way in accord with physical, geochemical and biological expectations;
- the overall complexity of the hierarchy of models employed in dose assessments leads to the possibility that unrecognized mistakes could occur - at critical points in the calculations, the values of estimated parameter and/or intermediate model outputs must be checked for environmental realism or validity;
- great care should be taken in the interpretation of low estimates of dose rates produced by assessment models, and their further use for risk or impact assessment;
- for prospective assessments, it is likely that future developments will be in the area of probabilistic models that will provide a more rigorous basis for estimating the uncertainties in the predictions. This will, however, make increasing demands on environmental information concerning the probability distributions of the process parameters, but it is also likely to indicate useful areas for research;
- probabilistic modelling techniques, together with GIS software, is likely to better indicate radiologically vulnerable areas for assessing the possible consequences of accidents; and,
- once an authorized release of radionuclides is in progress, the available monitoring data should be used to validate, and improve, the assessment model.

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Fig. 2. The generic conceptual model of the behaviour of the radionuclides released to the atmosphere, and the pathways leading to human radiation exposure [IAEA, 1989].

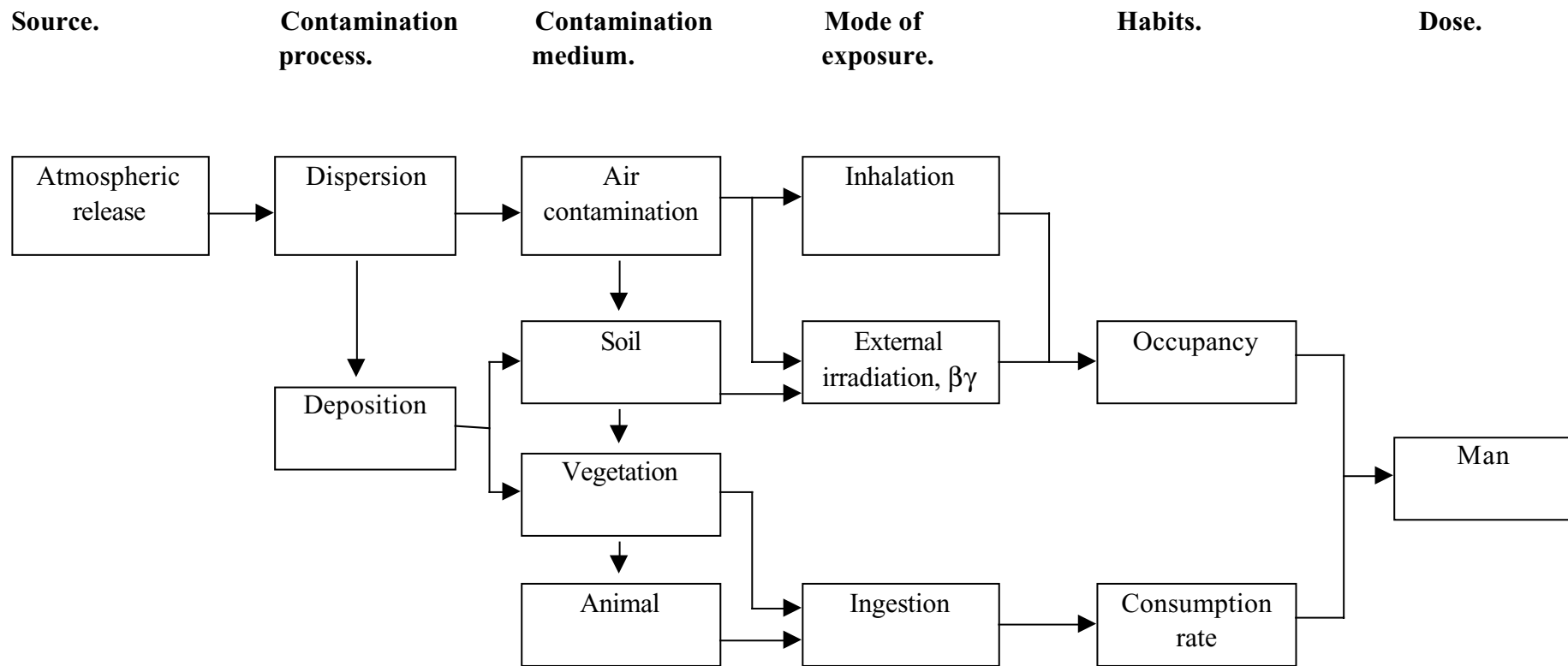


Fig. 3. The generic conceptual model of the behaviour of the radionuclides released to surface waters, and the pathways leading to human radiation exposure [IAEA, 1989].

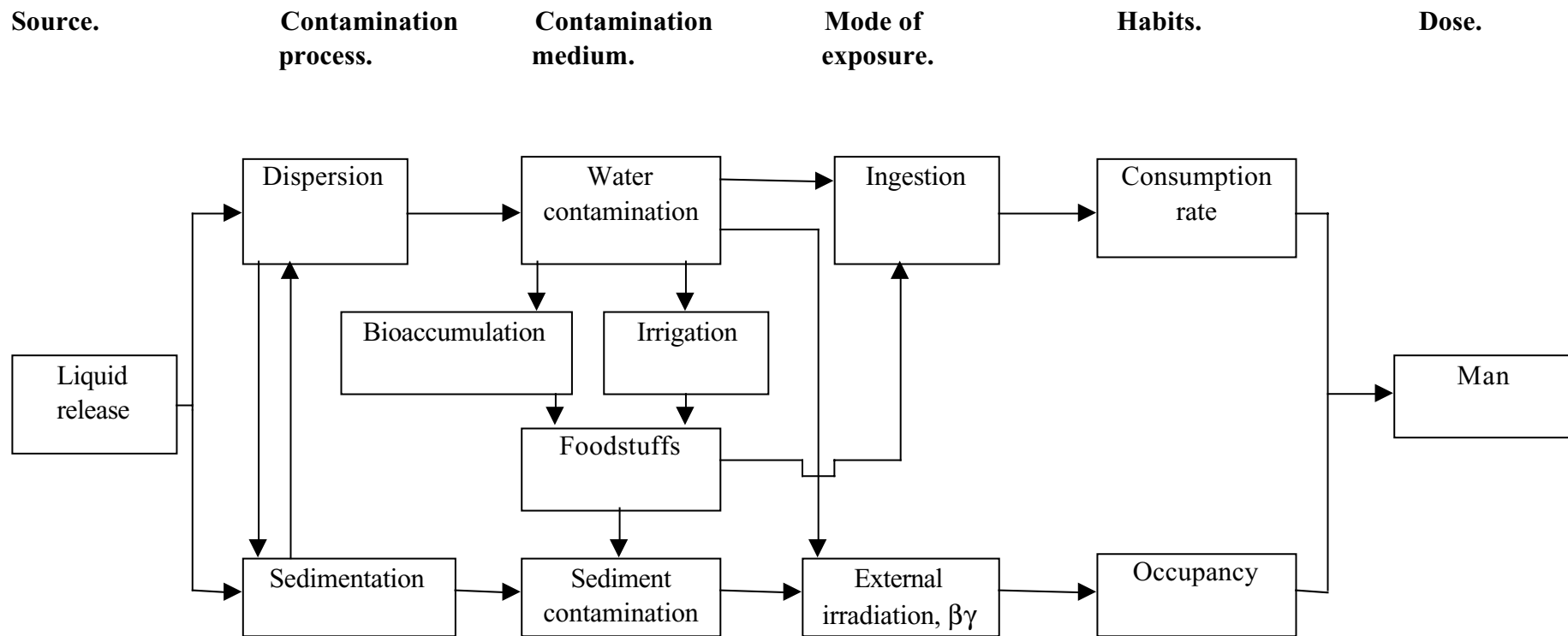


Fig. 4. The generic conceptual model of the behaviour of the radionuclides released to the atmosphere, and the pathways leading to the radiation exposure of plants and animals.

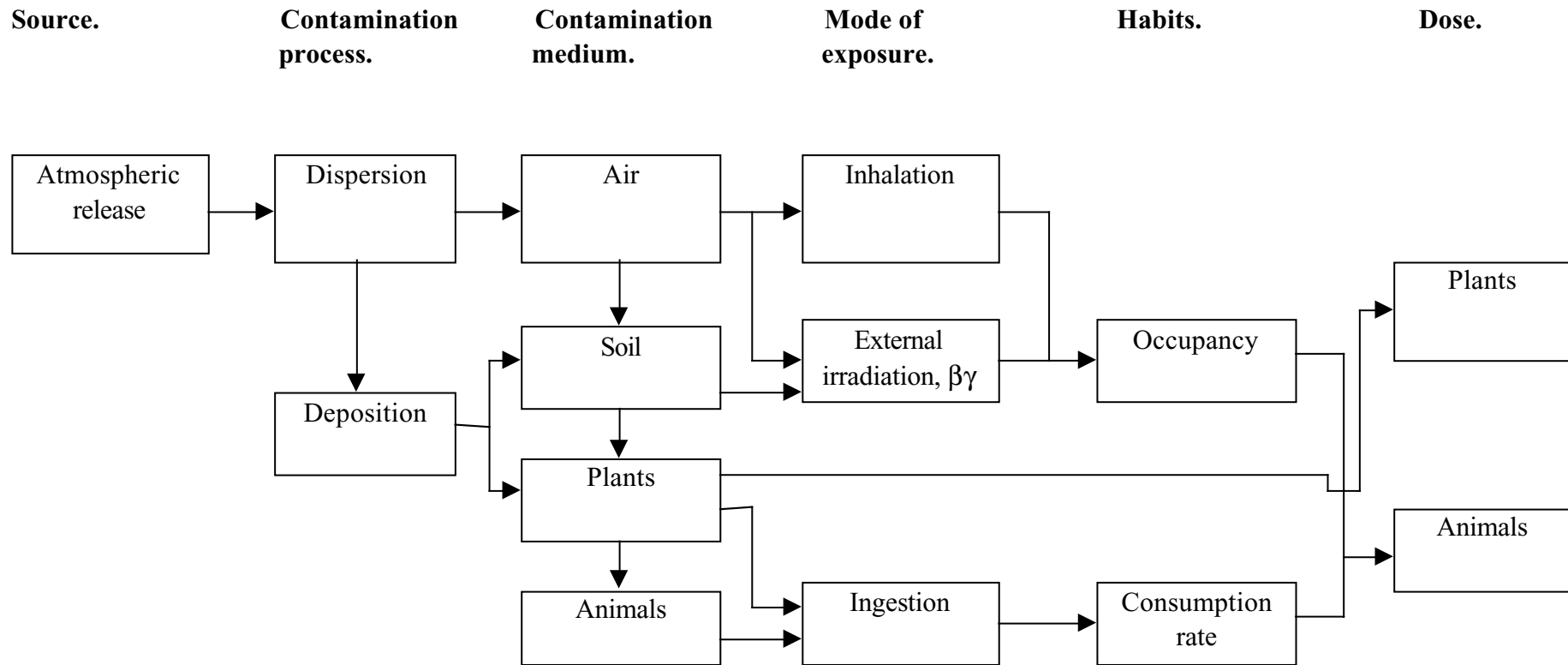


Fig. 5. The generic conceptual model of the behaviour of the radionuclides released to surface waters, and the pathways leading to the radiation exposure of the flora and fauna.

