# Environmental Dosimetry: The Current Position and the Implications for Developing a Framework for Environmental Protection

R & D Technical Report P350

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This report considers reference organisms for radiation dosimetry modelling for the purpose of developing a framework for environmental protection The information in this document is provided for information but will be used in further research projects in this field.

#### Keywords

Radioactive wastes, radiation exposure, radionuclides, wildlife impacts, reference organisms, dosimetry models, control framework.

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**R&D TECHNICAL REPORT P350** 

# FOREWORD

A previous report has shown that, in principle at least, there is a basis in current UK regulations and statutes for the development of criteria, and the application of controls, for the protection of the natural environment from any incremental radiation exposure from radioactive waste management activities [Woodhead, 1998].

The aims of this report are to:

- consider what flora and fauna could be usefully included as reference types across Europe for radiation dosimetry modelling for the purpose of developing a framework for environmental protection;
- consider to what level of complexity such models can reasonably be developed given the recognized constraints on the information that is likely to be available (or easily obtainable) concerning the radionuclide distributions in space and time both within, and external to, the organisms; and,
- propose a transparent procedure by which the dose factors required for environmental dose assessment can be developed for the identified reference flora and fauna, and up-dated as required.

This report provide a short introduction (Section 1) to the requirements of environmental radiation protection. Section 2 examines the factors that will influence the radiation exposure of native wild organisms in contaminated marine, freshwater and terrestrial environments and discusses the criteria that should be considered in identifying reference organisms to fairly represent the European region; it then goes on to suggest a range of reference organisms that might be appropriate for the each of the three environments. The details of the dosimetry models that have been employed in past environmental impact assessments are discussed in Section 3. These are further considered as to their utility as a basis for determining the radiation exposures of native wild organisms in contaminated environments that would be adopted within a framework for environmental protection. An outline is given of the future work that is necessary to realise this objective. The final Section 4 provides an overall summary and conclusions.

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# **EXECUTIVE SUMMARY**

The disposal of radioactive wastes has the probable consequence of increasing the radiation exposure of native wild organisms, both now and in the future. The magnitude of this hazard depends upon the radionuclides in the wastes (their quantities, half-lives and the radiations emitted) and their behaviour in the biosphere (their physical and chemical form). These factors govern the evolving distributions of the radionuclides, and their concomitant radiation field, in the environment. The nature, habitat preference and behaviour of the plants and animals - including their capacity for accumulating the radionuclides from their environment - in turn influence the degree of the incremental radiation exposure arising from the wastes. The magnitude of this incremental radiation exposure, from both internal and external sources, is the sole determinant of the impact of the radioactive properties of the wastes on the native plants and animals. It is apparent that any framework intended to assess and control the impact of radioactive waste disposal on the environment must include a means of estimating, in advance of any releases, the likely radiation exposure of the plants and animals.

Due to the data requirements it would not be possible to assess the exposure of individuals of each and every species in a contaminated area, nor to make an estimate of the corresponding possible impact. Recourse must be made to the use of reference (or generic) organisms that can provide a reasonable representation of the typical plants and animals present in the environment and, therefore, the range of dose rates likely to be received. The biological, physical and geochemical factors that influence the possible radiation exposure, and that must be considered in the selection of reference organisms, have been discussed. The sets of reference organisms that have been used in previous assessments of the environmental impacts of waste disposal have been reviewed and suggestions made for additions that would be relevant to the marine, freshwater and terrestrial environments of the UK and the wider European area.

In the pre-operational phase of a nuclear facility that is likely to release waste radionuclides into the environment, an assessment of the possible dose rates to the native wildlife has to rely on dosimetry models. These are simplified geometric representations of the reference plants or animals that permit the estimation of the radiation dose rate to the whole organism, or relevant internal organs or tissues (e.g., the gonads), from the radiation fields generated by internal and external sources of  $\alpha$ -,  $\beta$ - and  $\gamma$ -radiation. The available dosimetry models, mainly relating to aquatic organisms, have been reviewed, and their future development, not only for aquatic systems, but also for the terrestrial environment, has been discussed.

It has been concluded that the existing information on the behaviour of radionuclides in the aquatic and terrestrial environments is sufficient to provide a basis for the selection of reference organisms for the purposes of environmental dosimetry. The available dosimetry models are suitable for development and incorporation into a framework that would provide for the control of the incremental radiation exposure arising from the disposal of radioactive wastes and, thus, for the protection of the environment.

# 1. INTRODUCTION

From the very beginning of the nuclear programme in the UK and the attendant practice of releasing low-level radioactive wastes to the environment, in particular to coastal waters, it was recognized that there would be a concomitant increase in the radiation exposure of the native wild organisms. This led to the institution of a research programme to investigate the tolerance of fish towards ionizing radiation (MAF, 1947), and a consideration of the potential radiation effects on fish in the first radiological assessment of the discharges of liquid effluent to sea from the Windscale site (Dunster, 1952). While it cannot be said that this assessment of the potential radiation effects in fish was comprehensive, it included two essential elements: the estimation of the radiation dose rates from the contaminant radionuclides, and the comparison of these values with the dose rates that had been shown to cause harm in laboratory studies. These two elements have informed all subsequent assessments and been the subject of R&D programmes intended to improve our capacity to estimate, and ultimately control to an acceptable degree, the potential impacts of increased radiation exposure on the environment.

In any practical scheme intended to limit, or prevent, harm to the natural environment from radiation exposure, there are iterative links between the objective(s) of environmental protection, the relevant targets for dosimetry and the biological effects of radiation that are of concern. In addition, the source of the release, the range of radionuclides involved, and the receiving medium are likely to influence the development of a practical management framework.

# **1.1** The objective of environmental protection

The radionuclides released into the environment from human activities will have a range of chemical properties and speciation, and half-lives. These will interact with the physical and biogeochemical processes in the local environment to produce spatially and temporally varying radionuclide distributions. For the great majority of radionuclides, at the release rates of concern here, the gravimetric concentrations of the individual elements in the environment are extremely small, such that chemical toxicity may be neglected as a source of hazard. Their radioactive emissions, however, generate a spatially and temporally varying radiation field.

The concept of protecting the environment from any incremental radiation exposures arising from human activities may be resolved into two primary questions:

- what quality, function or attribute of the environment is to be protected? and,
- having determined the object(s) of protective action, what are the criteria by which an appropriate degree of protection may be applied and, as importantly, which may be used as a basis for a demonstration of compliance?

It may reasonably be accepted that the hazard due to the presence of waste radionuclides in the environment arises from the interactions of the ionizing radiation field with the native living organisms, i.e., from the radiation dose (rate) to the organism (this explicitly excludes the effects of radiation absorption in the abiotic, chemical and physical, components of the environment that are known to become significant only at dose rates and doses far higher than are relevant in the present context). This, however, immediately begs the question: "what is the nature of the risks from the radiation exposure?" (In this context it should be noted that the term "risks" is used in a general sense to

indicate "adverse outcomes" rather than in the more specific sense of the probability of a defined endpoint, e.g., the premature death of an individual).

A century of radiobiological research has clearly demonstrated that there is a wide range of biological consequences of irradiation, but that these are all mediated by initial damage at the biomolecular level. This is hardly surprising given that the absorption of radiation energy is via ionization - the separation of orbital electrons from atoms - and this may lead to the break-up of the biomolecules. Within the finely balanced biochemical environment of the cell, such molecular damage, particularly in the essential and non-replaceable DNA, may either:

- be repaired so that the cell can survive and function normally; or
- be misrepaired giving latent damage that may be expressed in the cell or its progeny. In the somatic cells of animals, this may lead to the initiation of cancer, and in the germ cells of any organism, to hereditary defects in offspring. In these two cases, the risk, rather than the severity of the outcome, increases in some manner (usually assumed to be in direct proportion) with the dose received, i.e., they are stochastic responses; or,
- cause the cell to die (apoptosis). In this case, the severity of the response the loss of tissue or organ function due to cell death increases rapidly with the total dose received above an effective threshold before saturating. This gives a sigmoid dose-response relationship, and effects of this type are termed deterministic.

It is apparent that the initial damage at the molecular level could, in principle, propagate to successively higher levels in the biological hierarchy, i.e., from the biomolecule to the cell, tissue, organ, individual organism, population, community, ecosystem and, ultimately, to affect biodiversity. In practice, however, there is a number of factors that may act, singly or in combination, to modify the nature and extent of the propagated damage. Misrepaired damage in a single cell may predispose an organism to the development of a cancer. If the host animal has a sufficiently long life expectancy for tumour development, the end result may be premature mortality (note that this is a specific risk for which it is possible, in principle, to define a probability). The early death of a single animal in the great majority of species would be unremarked, but that of a proportion of the population could have the immediate effect of changing the age-specific mortality rate for the present population, and, depending on the age-specific reproductive rate, may influence the future development or maintenance of the population. It is clear, therefore, that significant harm at the population level will only result from a substantial combined expression of the risk of early mortality in a proportion of the individuals in the exposed population.

If sufficiently large, the effects at the population level could have an impact at the community and higher levels of the biological hierarchy (it should be remembered that, in a natural system, all populations of organisms will receive a greater or lesser radiation exposure from the contamination and that there would be interactions between the responses of the different organisms). In the normal circumstance of a population that is being regulated by a variety of intra- and inter-species interactions (e.g., competition for food or habitat, predation, etc.) and environmental variables (e.g., the weather), it is possible that some small degree of radiation damage at the individual level could be accommodated (this is a general risk situation for which it would be very difficult to determine the probability of a defined outcome). In addition to effects on mortality, these considerations apply with equal validity to the potential effects of radiation on fertility (induction of cell death in the gonads),

fecundity (induction of embryo mortality) and hereditary mutation rate (for which selection pressure also enters the picture). These are all attributes that operate at the level of organs in individual organisms, but are important for the maintenance of a healthy population, and potentially, the community and higher levels of the biological hierarchy.

The discussion in the previous two paragraphs indicates that it is, in principle, possible that the radiation damage initially induced in the cells of individual organisms could be propagated up through the hierarchy of biological organization and produce effects at each succeeding level. The question remains, however, of whether the information concerning any possible radiation effects in individuals, at the dose rates expected in the environment from controlled waste disposal, could be used as a basis for assessing the implications at these higher levels with any certainty. Equally, it may well be asked "could any such impact at the supra-individual level be detected?". In the latter respect, it must be remembered that it has usually been very difficult to attribute observed changes in populations of native flora and fauna to anything other than major, and self-evident, factors such as deliberate culling, loss of food supply or habitat, known endemic disease and so on; the effect of the pesticide, DDT, on bird populations is a notable, and cautionary, exception.

## 1.2 Conclusions

The primary conclusion from this brief discussion is that if the radiation exposure from radionuclides in the environment produces no discernable effect in any of these (or, indeed, any other) attributes in the individual organisms, then it is inconceivable that there will be any effects at the population and higher levels of organization. This conclusion is consonant with that developed in the case of chemical contaminants (Haux and Forlin, 1988). It also clearly identifies the biological level - the individual organism - at which it is appropriate to focus attention in order to provide for protection of the environment, and the action to be taken - the restriction of the radiation exposure of the individual. It is consistent with the UK Wildlife and Countryside Act (UK-Parliament, 1981) that specifically provides for the protection of individuals of some species of plants and animals. In addition, it provides for the concerns expressed for the protection of a higher degree of protection might be justified.

# 2. THE IDENTIFICATION OF TARGETS FOR DOSIMETRY AND REFERENCE ORGANISMS

From the brief introductory discussion, it has been concluded that:

- 1. the individual organism is the appropriate focus for action to provide for environmental protection; and
- 2. any required degree of protection can be achieved by applying appropriate limits to the radiation exposures of the individual organisms.

All the native flora and fauna in the immediate vicinity of a release of radioactive materials into the environment are potentially at risk of harm from the increased radiation exposure. It is very unlikely, however, that all the necessary information would be available, or could reasonably be obtained, to undertake a detailed risk assessment for every species - this would require the estimation of the (time- and space-dependent) dose rate for individuals of each species, and the availability of species - specific dose rate/response relationships for all the effects endpoints of interest. The first step is to accept that it is not possible to consider all the native species - recourse must be had to the use of reference or generic organisms for both dosimetry and the assessment of potential radiation effects. In these circumstances, it is necessary to simplify and generalise the process of dose assessment whilst retaining sufficient realism for the results to achieve credibility.

It has also been clearly recognized that radionuclides released into the environment would become more or less widely distributed depending on their chemical nature and half-life. There is, therefore, a range of biological, physical and geochemical factors that will influence the choice of relevant targets for the purposes of dose assessment in any system aimed at providing for the protection of the environment. What, then, are the criteria that should be applied to this process?

## 2.1 Influences on the radiation exposure of native organisms

#### 2.1.1 Biological factors

Given that the radiation field will show spatial and temporal variability, it is considered essential that the chosen range of reference organisms should include sufficient examples to demonstrate the influence, on the dose rate, of differing habitat preferences, behaviour, and the innate capacity of the organisms to accumulate radionuclides. This process is also likely to result in the selection of representative species from the main trophic levels. The purpose is to encompass the range of dose rates likely to be experienced by all the native organisms within the contaminated area. This may be termed selection on the basis of "radioecological sensitivity".

The radiobiological literature is consistent in showing that there are considerable variations in radiosensitivity between species, between tissues and organs within individual organisms, and between different stages in the life cycles of many individual species. The first source of variability is reduced, but not eliminated, when comparisons of acute responses (usually mortality) are made on a consistent basis (e.g., the effects of differences in metabolic rate between the homeothermic mammals and the poikilothermic vertebrates and invertebrates are taken into account), and when the

effects of chronic low-level irradiation are examined. The second source of variation in radiosensitivity must be considered when it is known or suspected that there will be significant differential accumulation of radionuclides within the tissues of the organism; this helps to identify potential targets for dosimetry at the sub-individual level. The final source of variation becomes significant when radiation effects in the individual are considered in the context of potential impacts at the population level; i.e., it is not only the survival of the individual that is of concern, but also its capacity to contribute, through its total reproductive performance, to the maintenance of a healthy population. Again, this implies important targets for dosimetry at the sub-individual level. This variability provides a second basis for the selection of the range of reference organisms, i.e., their radiosensitivity, insofar as this can be assessed with the available information.

A final factor that might need to be taken into account relates to the concept of the ecological significance of particular types of organism, i.e., do they have an important role in the normal functioning of the community? An example might be the grasses in a meadow system. This may be termed selection on the basis of "ecological sensitivity".

Collectively, these three factors:

- radioecological sensitivity;
- radiosensitivity; and,
- ecological sensitivity

may be employed to guide the selection of an appropriate range of reference organisms, and target tissues and organs, for the purposes of dose rate assessment.

#### 2.1.2 Physical factors

Physical processes act, primarily, to disperse the radionuclides released into the environment under controlled or accidental conditions. Air and water currents transport the radionuclides away from the source, and the associated turbulent conditions cause dilution. For releases to air, dry deposition and washout by rain then lead to accumulation by vegetation and at the soil surface. In aquatic systems, depending on their chemical natures, the radioelements are partitioned to a greater or lesser extent from the soluble to the particulate phase, and sedimentation leads to accumulation of activity on the beds of lakes, rivers and the sea. Contaminated fine sediment can be resuspended by currents and wave activity (particularly during storms) and transported by the tides and residual currents to low energy areas where redeposition can occur, e.g., on salt marshes in estuaries. These factors again relate to what has been termed radioecological sensitivity.

The physical half-lives of the radionuclides in the release control the extent to which their distributions come to an effective equilibrium with environmental processes - if the rate constant of the environmental process is of the same order as the radioactive decay constant, then effective equilibrium is attainable and fluctuations in the release rate will be closely followed by the resultant changes in the environmental concentrations. All other factors being equal, this also tends to mean that nuclides with short half-lives remain relatively closer to the point of release, and that there will be a greater range (as well as greater quantities) of radionuclides in this area to make a contribution to the total radiation exposure.

The final significant physical factor relates to the nature of the radiations emitted by the different radionuclides and their typical ranges in tissue, i.e., about 50  $\mu$ m for  $\alpha$ -particles, 1 - 2 cm for  $\beta$ -particles and 0.1 - 1 m for x- and  $\gamma$ -rays. Depending on the radionuclides involved, this governs the relative importance of internal and external sources of exposure for different sizes of organism. For example, it is probably not necessary to specifically consider very small organisms, such as a bacterium, for three reasons: first, the quantity of radionuclide that can be incorporated into the organism at the concentrations that could be reasonably be expected in the external environment are so small that a nuclide decay in its "lifetime" would be improbable; second, even if a decay did occur, the great majority of the energy would be dissipated outside the cell (this factor would also apply to tissues such as fungal hyphae that are extremely fine although extended and may have significant total mass); and, third, the radionuclides outside the cell, but within the radiation range, would be a much more significant source of exposure (it could approach D<sub>∞</sub> for the radionuclide concentration in the external medium, see chapter 3). The ranges of the radiations also indicate the scale on which the distribution of the radionuclides needs to be known if reasonably accurate estimates of the dose rate to specific targets are to be made.

#### 2.1.3 Geochemical factors

The specific chemical form of a radionuclide in the effluent (if this differs from that of the corresponding element in the environment) may differentially influence its initial behaviour. However, the long-term behaviour of the majority of radionuclides in the environment is controlled by the chemical nature of the element, i.e., the radionuclides become incorporated, with some time delay, into the natural geochemical cycles of the labile fractions of the corresponding elements (this effectively excludes the greater part of the mineral fraction in the majority of cases).

For radionuclide releases to the lower atmosphere, dry deposition and washout by rainfall are relatively rapid although, as the Chernobyl accident has shown, there can be widespread dispersion. In the terrestrial environment, the major concerns relate to radionuclide retention by vegetation and in the surface layers of the soil. Here it constitutes an external source of exposure for the plants and surface-living and burrowing organisms. The soil type influences the radionuclide partitioning into the soil solution where it becomes available for uptake into plants and transfer into the foodchain - there representing an internal source of exposure.

Radionuclide transport by run-off into surface waters, leaching from soils into groundwater flows and deposition from air to the water surface represent inputs that are comparable to the direct discharges, i.e., the radionuclides are introduced into the water column. The primary concern in aquatic systems is the partitioning of the radionuclides from the soluble to the particulate phase and subsequent sedimentation to the river-, lake- or sea-bed. Even with a relatively low distribution coefficient ( $k_d$ ), e.g., a value of 350 for <sup>137</sup>Cs in the sea, this means that the underlying sediment becomes a much more significant source of external radiation exposure than the water. The residual radionuclide contamination in the water is a significant potential source for accumulation for a large proportion of the aquatic fauna, either directly, or via their foodchain.

#### 2.1.4. Conclusion

A consideration of these factors, taken together, provides a reasonable basis for selecting a suite of reference organisms that can be expected to experience the full range of radiation dose rates in a contaminated environment, and include representatives that are radiosensitive (either as individuals or in terms of specific organs or tissues), and may also be ecologically sensitive.

## 2.2. Selection of reference organisms

This general selection procedure for reference organisms has been most frequently applied in respect of the marine environment [IAEA, 1988, 1998a, b; Pentreath and Woodhead, 1988]. This has primarily been a consequence of the use that has been made of the sea as a repository for low level liquid, and solid, radioactive wastes and the *ad hoc* need to provide some assurance that the consequent radiation exposures of the native flora and fauna would not lead to significant harm. These reference organisms could be easily adapted for the freshwater environment. Application in the terrestrial environment has been much more limited and fewer reference organisms have been selected for dosimetric purposes [IAEA, 1992; Amiro and Zach, 1993; Amiro, 1997]. For contaminated areas in both the aquatic and terrestrial environments, there has been a number of specific dose rate assessments for local species of flora and fauna without any suggestion that they would necessarily be suitable as reference organisms; these models might, however, be adaptable for this purpose [e.g., NCRP, 1991; Woodhead 1970, 1986].

#### 2.2.1 Reference organisms in the marine environment

The most extensive single list of reference organisms that has been selected, with some consideration of these criteria, to represent the marine environment is [IAEA, 1988; Pentreath and Woodhead, 1988]:

- *Fish*: radiobiological studies have shown that these are probably the aquatic organisms most sensitive to the effects of chronic irradiation [IAEA, 1976, 1988; UNSCEAR, 1996]. Assessments for pelagic and benthic species allow the contribution of γ-radiation from the underlying contaminated sediment to be highlighted. Fish generally show a relatively low capacity for accumulating radionuclides;
- *Large crustaceans*: these organisms generally have higher concentration factors for radionuclides than fish, thus increasing the relative importance of the internal source as compared with fish. Again, assessments are made for pelagic and benthic species and, being smaller than fish, serve to demonstrate the relative importance of the  $\beta$  and  $\gamma$  emitters in the sediment;
- *Benthic molluscs*: these organisms generally have higher concentration factors than the previous two groups and, with smaller size, show the influence of these factors on the dose rates from internal and external sources; and,
- *Small crustaceans*: if it is assumed that the concentration factor data available for surface-living zooplankton are applicable, then these organisms would have the highest concentration factors for most elements. Also, being the smallest organisms in this selection, and considering both pelagic and benthic types, they show most clearly the

effect of these factors on the relative contributions to the dose rate from internal and external sources.

In comparison with the potential selection criteria discussed in the previous section there are, however, some obvious omissions. For the reference organisms listed, there is no provision to take account of:

- either preferential accumulation of the contaminant radionuclides into particular tissues or organs. To a great extent, this was a tacit recognition of the lack of relevant data in the majority of cases. Pentreath and Woodhead [1988] did, however, briefly consider the potential effect of this factor in a general manner;
- or differential tissue or organ radiosensitivity. This omission is, again, largely a reflection of the lack of relevant data, but there is sufficient information available to indicate that the ecologically important process of gametogenesis is relatively radiosensitive. It would seem sensible, therefore, to provide for the specific assessment of the radiation exposure of the gonads in those situations where the particular radionuclides, the source distributions and the organism behaviour indicate that these factors could increase the dose rate to these organs relative to that to the whole body;
- or different stages of the life-cycle it is implicit that adult organisms are being considered. Earlier studies [Woodhead, 1970; IAEA, 1979] had considered the problem of radiation dosimetry for fish eggs (developing embryos), and these could have been incorporated into the assessment with little difficulty.

In addition, the ecologically important phytoplankton was not included in the list of reference organisms for coastal waters considered by Pentreath and Woodhead [1988], although it has been considered in other contexts [IAEA, 1976; Woodhead, 1973a]. Marine macrophytes are also conspicuous by their absence but have been included in previous generic assessments [NRCC, 1983]. A seabird [Woodhead, 1986], a seal [Pentreath and Woodhead, 1988] and a whale [IAEA, 1998b] have also been considered in site-specific assessments. Potential candidate organisms for the marine reference set, taking due account of the factors discussed in section 2.1, are listed in Table 1.

#### 2.2.2 Reference organisms in the freshwater environment

Studies related to the freshwater environment have been more limited, and there has been little development, in terms of reference organisms, beyond the set selected in [IAEA, 1976], i.e., phytoplankton, zooplankton, a mollusc, a crustacean and a fish. Individual site-specific assessments have, however, considered a range of additional organisms including aquatic plants, insects, turtles, alligators, musk rats and ducks [NRCC, 1983; NCRP, 1991]. A notable absentee, with a life-cycle split between the aquatic (embryonic and larval phases) and the terrestrial (adult phase) environments, is a reference amphibian. In view of the generic similarities between the marine and freshwater environments in terms of the criteria discussed in section 2.1., it is to be expected that there would be some equivalence between the lists of potential reference organisms (see Table 1.).

The marine environment	The freshwater environment	The terrestrial environment
Pelagic phytoplankton	Pelagic phytoplankton	Tree
Macrophyte	Macrophyte	Shrub
Pelagic zooplankton	Pelagic zooplankton	Herb
Benthic mollusc	Benthic mollusc	Germinating seed
Small benthic crustacean	Small benthic crustacean	Fungus
Large benthic crustacean	Large benthic crustacean	Caterpillar
Pelagic fish	Pelagic fish	Social insect
Benthic fish	Benthic fish	Wood louse
Fish egg	Fish egg	Earthworm
Seal	Amphibian	Herbivorous mammal
Whale	Small aquatic mammal	Carnivorous mammal
Seabird	Duck	Small burrowing rodent
		Woodland bird

 Table 1.
 Potential reference organisms for the purpose of environmental dosimetry.

#### 2.2.3 Reference organisms in the terrestrial environment

The generic terrestrial organisms that have been included in previous assessments are: a vascular plant, soil microflora, soil invertebrates, a large herbivorous mammal, and a fruit/seed eating bird [IAEA, 1992; Amiro and Zach, 1993; Amiro, 1997]. This list of reference organisms is clearly partial, and could be extended by the application of the selection criteria outlined in section 2.1.

For the terrestrial environment there are two release scenarios to consider: a controlled or accidental release to the atmosphere in gaseous or aerosol form, and the remobilization of radionuclides from a surface or geological waste repository into the near-surface groundwater as a consequence of natural

processes. A third source that might be considered is the use of contaminated surface water for irrigation.

From the viewpoint of environmental dosimetry, a release to the atmosphere has two phases: in the near-field and at short time-scales, the contaminated cloud is a significant source of exposure, and in the longer term and at all distances, dry deposition and washout reduce the significance of the atmospheric source relative to the contamination on the plants and the soil surface. At all stages, the experience of the Chernobyl release appears to indicate that the woodland systems are more effective in intercepting and retaining the airborne activity by the process of dry deposition than meadow or pasture environments; this can, however, be substantially modified by the frequency of occurrence and quantity of rainfall which often resulted in upland sites becoming more contaminated than neighbouring lowland areas.

Contaminated groundwater, with the inevitable redistribution of the radionuclides between the soluble and particulate phases, would be comparable to the late phase of a release to the atmosphere when the greater part of the activity is in the surface layer of the soil. Irrigation with contaminated surface water would, similarly, be comparable to the washout phase of a release to the atmosphere.

Within the woodland system, the plants show a range of sensitivities to chronic irradiation - i.e., in terms of mortality, coniferous trees > deciduous trees > shrubs > herbs > fungi - but there is relatively little comparative information on the effects of such exposure on gametogenesis and reproductive capacity [UNSCEAR, 1996]. Purely biological factors that are likely to be significant for the selection of targets for dosimetry include the facts that: green plants are the primary producers; tree growth and the production of seeds arise from meristem tissue in aerial buds, whereas for grasses the meristem is at or near the ground surface; many shrubs and herbs can regenerate from sub-surface vegetative growth points; virtually all plant seeds germinate on, or just under the soil surface; and, fungi have a quite different lifestyle and are very important in breaking down and recycling biological material. These factors indicate that the meristem in a range of green plants, the fungal fruiting body, and a germinating seed, are candidates for inclusion in the list of reference organisms (see Table 1.).

There is an enormous range of invertebrate species, with contrasting lifestyles, in the terrestrial environment, and it is clearly impossible to consider them all. From the brief discussion in the previous paragraph, it may be concluded that the woodland canopy and the litter layer at the soil surface would be two environments giving external exposures at the high end of the range. The selection of potential reference organisms could include:

- a leaf-consuming insect larva (caterpillar) in the canopy;
- a nectar-feeding social insect (bee). This could double as the adult form of the caterpillar and also represent the biological transport of contaminated material pollen and nectar to the hive where it would lead to external and internal irradiation of both the current generation of workers and, through the exposure of males and queens, the next generation;
- a litter-inhabiting detritivore (wood louse); and,
- an earthworm.

This would appear to include a sufficient variety of habitats and life-styles to indicate the effects of these factors on the dose rate and, more importantly, show the range of dose rates likely to be experienced.

Radiobiological studies show that the vertebrates are the most radiosensitive of the terrestrial organisms, in terms of either mortality or reproductive capacity [UNSCEAR, 1996]. Further consideration of feeding habits and habitat occupancy lead to the selection of a reference herbivore (deer), a carnivore (fox), a small burrowing omnivore (rodent) and a bird (the European blackbird could be an appropriate choice as it spends a substantial fraction of its time on the contaminated ground, and is a consumer of earthworms and litter invertebrates). In all cases, an attempt should be made to estimate the radiation exposure of the gonads and/or the developing embryo, in addition to the whole body of the adult animal.

# 2.3 Conclusions

A brief discussion has been given of the factors that could influence the selection of reference organisms for the purpose estimating the radiation dose rates in contaminated environments. These have been considered, and the reference organisms proposed for the marine, freshwater and terrestrial environments are listed in Table 1. It is not suggested that these selections are necessarily comprehensive but they should give a fair representation of the range of dose rates likely to be experienced from both internal and external sources of contamination in these environments.

# **3.** THE DOSIMETRY MODELS

It has been noted above that the biogeochemical behaviour and the consequent distributions of waste radionuclides after release to the environment are governed by their individual chemical natures and speciation. Except for the short-lived radionuclides, the distributions will only rarely come to an effective equilibrium with the range of environmental processes in operation. Together with the fluctuations in the release rates from individual sources, this results in radiation fields that show large spatial and temporal variabilities. This variability is further compounded by the characteristic ranges of the radiations that extend from about 50  $\mu$ m ( $\alpha$ -particles in tissue) to many metres ( $\gamma$ -rays in air). This range of spatial scales also applies to the organisms of potential interest, i.e., from phytoplankton and fish to deer and trees. The temporal scale of interest relates to the generation times of the organisms and ranges from a few hours (unicellular phytoplankton) to decades (a tree).

In principle, this means that the dose rates and accumulated doses should be assessed on these spatial and temporal scales and this requires corresponding information concerning the detailed behaviour and distributions of the radionuclide. In practice, the procedure is simplified to utilize the actual detail of the information that is realistically likely to become available. The dose rates are estimated for unit radionuclide concentrations in a specific source compartment that provides a basis for estimating consequential equilibrium concentrations in other compartments to a greater or lesser degree of detail, depending on the information available. For the aquatic environment, where authorized liquid discharges are made to the water column and inputs from accidental releases are likely to be to the water column or the water surface, the unit concentration is taken to be in the water (Bq  $m^3$ ); partitioning to sediment and uptake into aquatic organisms is then determined by application of the relevant distribution coefficients  $(k_d)$  and concentration factors, respectively. The situation is less well-developed for the terrestrial environment, but as both authorized and accidental releases are likely to be to air, it is reasonable to propose that the consequential dose rates to terrestrial organisms be related to either a unit concentration in air (Bq m<sup>-3</sup>), or, more realistically, to a unit deposition density (Bq  $m^2$ ). For disposal to landfill or a sub-surface repository, the waste radionuclides are likely to be mobilised by groundwater flow; where this migrates to surface waters, the unit concentration in the receiving water body is the relevant base parameter; if the ground water intersects the soil surface in transit, then the unit concentration in the surface soil (Bq kg<sup>-1</sup>) could be used. These are cases that need to be further explored and developed. The dosimetry models provide the dose coefficients for the unit radionuclide concentration in the relevant source medium, and these can then be applied to the actual or predicted concentrations in these media.

### 3.1 Radiation dosimetry

From the earlier discussion it may be taken as given that the biological effects of radiation are the result of ionization processes in tissue. Because ionization is the separation of orbital electrons from the parent atoms, a process that requires energy, this results in the absorption of energy from the incident radiation field. This leads directly to the definition of the radiation dose as the quantity:

absorbed dose, 
$$D = \frac{d\varepsilon}{dm}$$
 (1)

where  $d\bar{\epsilon}$  is the mean energy imparted to matter of mass dm (see [ICRU, 1998] for fuller details). The quantity, absorbed dose, has units of J kg<sup>-1</sup>, and this is given the special name gray (Gy). Although this definition relates to a limiting domain, in practical radiation protection the absorbed dose is usually determined as the average value over some specified biological entity - a tissue, organ or the whole body.

At the low dose rates and low total accumulated doses characteristic of the majority of environments contaminated by authorized releases, it may well be that microdosimetric considerations become important, i.e., the distribution of absorbed energy divided by the mass of the individual cell or cell nucleus (the presumed primary targets for radiation action) becomes extremely inhomogeneous. In this case, the quantity:

specific energy, 
$$z = \frac{\varepsilon}{m}$$
, (2)

where  $\varepsilon$  is the energy imparted to the matter of mass m in the defined target, may be more relevant to the determination of the consequent radiation effects. The unit of the quantity specific energy remains the J kg<sup>-1</sup>, and this retains the special name gray (Gy). The specific energy may be due to one or more (energy deposition) events, i.e., the passage through the defined target mass m of one or more directly ionizing particle tracks. The probability that the specific energy is  $\le z$  is given by the distribution function F(z), and the probability density, f(z), is the derivative of F(z):

$$f(z) = \frac{dF(z)}{dz}.$$
(3)

Both F(z) and f(z) are dependent on the absorbed dose.

From a consideration of microdosimetric factors, DNA repair processes, experimental radiobiology and epidemiological studies of tumour induction in the atom bomb survivors, it has been concluded that low doses and low dose rates of low LET radiation are less than  $2.10^5 \,\mu\text{Gy}$  and  $6.10^3 \,\mu\text{Gy} \,h^{-1}$ , respectively [UNSCEAR, 1993]. Below these levels, it is to be expected that the response relationship for stochastic effects would be linear with dose.

#### 3.1.1 The need for dosimetry models

Due to the requirement to assess the absorbed dose rates from both external and internal sources of  $\alpha$ - and  $\beta$ -particles and  $\gamma$ - and x-rays, to small and large, sedentary and mobile, organisms in the preoperational phase of the development of a nuclear facility, it is not possible to employ instrumental methods. For all these situations, it is necessary to develop computational methods using dosimetric models. The simplification of the description of the environmental behaviour of the radionuclides has already been referred to above. A further degree of simplification must be obtained by reducing the implicitly complex morphologies of the reference wild organisms to regular geometric solids that are amenable to mathematical manipulation. For example, the numerous, morphologically complex,

extensible appendages of a crustacean (together with their radionuclide content) are simply incorporated into a body that is represented by a solid ellipsoid (see below). Provided that the implications of the underlying assumptions are recognized, it can be demonstrated that the estimates of dose rate obtained using these simplified models *do* give a reasonable indication of the radiation exposure of organisms in contaminated environments.

Physical descriptions of the processes by which energy is transferred to tissue from  $\alpha$ - and  $\beta$ particles and  $\gamma$ - and x-rays have been developed theoretically from first principles and given mathematical expression [see, for example, Johns and Laughlin, 1956; Evans 1968; Bichsel, 1968]. These expressions are, however, very complex and, due to the energy-dependent and stochastic nature of the processes involved, are not easy to apply to real situations in the environment [Roesch, 1968]. This has led to the development of simpler empirical expressions, involving energy-dependent parameters, to describe the absorbed dose distribution about point sources of  $\alpha$ - and  $\beta$ -particles and  $\gamma$ -rays [Loevinger *et al.*, 1956; Berger, 1968, 1971; Harley and Pasternack, 1972; IAEA, 1979]. These expressions can then be integrated over defined source distributions to give an estimate of the dose rate at specified points in tissue [Loevinger *et al.*, 1956]. The determination of the dose rate at different points within the target volume provides a basis for estimating the average dose rate to the tissue or organ [Brownell *et al.*, 1968; Ellett and Humes, 1971]. Although relatively simple in concept, this approach can be developed, as necessary, to accommodate more complex organism morphologies, and more detailed information on the time- and space-dependent radionuclide distributions as it becomes available.

#### 3.1.2 The biologically-effective dose rate

There is a very substantial body of experimental evidence to indicate that the absorbed dose of high linear energy transfer (LET) radiation ( $\alpha$ -particles) required to produce a given biological effect is less than that of low LET radiation ( $\beta$ -particles and  $\gamma$ -rays) - the relative biological effectiveness (RBE) phenomenon [e.g., Sinclair, 1985]. The relative biological effectiveness is defined as:

For human radiological protection practice, this phenomenon is taken into account by applying dimensionless radiation weighting factors ( $w_r$ ) to the absorbed doses from the different radiations to give a quantity called the equivalent dose, where:

equivalent dose,  $H = w_r x D$ .

The unit of the quantity, equivalent dose, remains the J  $kg^{-1}$  but it is given the special name Sievert (Sv).

In an environmental protection context, it has been suggested [Pentreath, 1999] that a specific quantity be defined, i.e.,

Dose Equivalent Flora and Fauna, DEFF = 
$$w_r \ge D$$
.

Although this quantity has the unit J kg<sup>-1</sup> and could, in principle, take the special name Sv, it was additionally suggested that it be given the special name DEFF to avoid confusion with human radiation protection practice. (It should be noted that the allocation of special names for derived units in the International System of Units (SI) is at the discretion of the Bureau International des Poids et Mesures.). For the usual case of a mixture of radiation fields in a contaminated environment, the total, biologically effective, radiation exposure would then be given by:

DEFF = 
$$w_r(\beta,\gamma) \ge D(\beta,\gamma) + w_r(\alpha) \ge D(\alpha)$$

In this manner, the equivalent doses to a tissue or organ from the different radiations may simply be summed to give a single measure for the total biologically effective radiation exposure from the radionuclides present in a contaminated environment.

The values of the radiation weighting factor (then the Quality Factor, Q) originally chosen by the ICRP [ICRP, 1977] for use in human radiological protection were broadly related to the LET of the radiations, and the default values were 1 for  $\beta/\gamma$  radiations and 20 for  $\alpha$ -particles. The current values of  $w_r$  (compatible with the Q and numerically the same) have been chosen to be representative of the RBE values determined for the induction of stochastic effects (principally cancer, but to the extent that this response is initiated by somatic mutation, it would also apply to heritable mutations) [ICRP, 1991].

A similar approach, initially based on comparisons of LET, could be employed in respect of the exposure of the flora and fauna. At the present time, therefore, it seems reasonable to propose that a provisional  $w_r$  value of 20 be applied in respect of the  $\alpha$ -radiation absorbed dose rate to the tissues of wild organisms, with the recommendation that all the available data be reconsidered from an environmental protection viewpoint (this implicitly assumes that the  $w_r(\beta,\gamma) = 1$ ). In this review, particular emphasis should be placed on the experimental RBE data that are available for: the species of organism that correspond to each of the reference (generic) organisms chosen to be representative of the different environments; the endpoints of relevance in an environmental context; and, low dose/dose rate exposures from both  $\beta/\gamma$  radiations and  $\alpha$ -particles. Although, in principle, this review could lead to proposals for a number of differing  $w_r(\alpha)$  for the variety of generic organisms and endpoints of interest, this level of sophistication is unlikely to be justified by the uncertainties in both the raw data, and the assessments of the absorbed dose rates for the contaminated environment. The more pragmatic approach of selecting a single representative value would have the twin virtues of simplicity in application and the production of comparable quantities for the total, biologically effective, doses/dose rates, i.e., the DEFF, for the flora and fauna.

### **3.2 Dosimetry models**

#### 3.2.1 The point source dose distribution functions

A dosimetry model is a basis for estimating, through computation, the radiation exposure of an organism from a source of radiation. Provisional lists of generic organisms that might be selected to represent the marine, freshwater and terrestrial environments have been given in Table 1. As noted in Chapter 2, some of these generic organisms have already been employed for practical assessments of radiation exposure in existing, or potentially, contaminated areas.

The simple geometries adopted for these examples of dosimetry models are given in Table 2. These geometries, then, become the targets for which the dose distribution is determined by the integration of the point source dose functions over the relevant radiation source distribution. For ease of computation (see Section 3.1.1. above), empirical expressions have been developed for application in the aquatic environment [IAEA, 1976; 1979].

Organism	Geometry	Dimensions cm	Mass kg <sup>a</sup>	Reference
Phytoplankton	Sphere	Diameter 5 10 <sup>-3</sup>	6.5 10 <sup>-11</sup>	IAEA, 1976
Fish eggs	Sphere	Diameters: 0.08, 0.12 and 0.2	2.7 10 <sup>-7</sup> , 9.1 10 <sup>-7</sup> and 4.2 10 <sup>-6</sup>	IAEA, 1979
Zooplankton/ small benthic crustacean/ a small insect	Ellipsoid	Major axes: 6.2 x 3.1 x 1.6	1.6 10 <sup>-5</sup>	IAEA, 1988 NCRP, 1991
Large benthic crustacean	Ellipsoid	Major axes: 3.1 x 1.6 x 0.78	2.0 10 <sup>-3</sup>	IAEA, 1988
Benthic mollusc/ a large insect	Ellipsoid	Major axes: 2.5 x 1.2 x 0.62	1.0 10 <sup>-3</sup>	IAEA, 1988 NCRP, 1991
Pelagic and benthic fish	Ellipsoid	Major axes: 45 x 8.7 x 4.9	1.0	IAEA, 1988
Seabird and duck	Ellipsoid	Major axes: Solid tissue at an average density of $0.8 \text{ g cm}^3$ :	Total - 0.6	NCRP, 1991
		15 x 11 x 7.6 Feathers at an average density of 0.33 g cm <sup>-3</sup> and overall dimensions:	0.55	
		21 x 16 x 11	0.05	
Seal	Ellipsoid	Major axes: 180 x 35 x 19	58	IAEA, 1998b
Whale	Ellipsoid	Major axes: 450 x 87 x 48	10 <sup>3</sup>	IAEA, 1998b

Table 2. The shiple geometries that have been adopted for generic of gamping.	Table 2.	The simple geometries that have been adopted for generic organisms	j.
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<sup>a</sup> Apart from the seabird/duck, the organisms have been assumed to have a uniform body density of 1 g cm<sup>-3</sup>.

For  $\alpha$ -radiation, the empirical point source dose distribution function has the form:

$$D_{\alpha}(r) = \frac{4.59 \times 10^{-2}}{\rho r^2} (A + Br^2) \qquad \mu Gy \ h^{-1} \ Bq^{-1} \tag{4}$$

where:

- $\rho$  is the density of the medium (assumed to be soft tissue, freshwater or seawater with a density of 1 g cm<sup>-3</sup>);
  - is the distance between the point source and the target point ( $\mu$ m) and is limited to r  $\leq R(E_{\alpha em})$ , the range of an  $\alpha$ -particle at the emission energy  $E_{\alpha em}$ ;

Α

r

is the stopping power of the medium (assumed to be tissue at a density of 1 g cm<sup>3</sup>) at the emission energy of the  $\alpha$ -particle:

$$A = \left(\frac{dE_{\alpha}}{dr}\right)_{E_{\alpha em}} \text{Mev } \mu \text{m}^{-1} \text{, and may be obtained from Fig.1; and,}$$
$$B = \frac{3[E_{\alpha em} - AR(E_{\alpha em})]}{R(E_{\alpha em})^3} \text{Mev } \mu \text{m}^{-3} \text{, and may be obtained from Fig. 2. The}$$

end of the nominal range of the  $\alpha$ -particles has been taken to be the point at which the stopping power falls to one half of its peak value and the value of R(E<sub>\alphaem</sub>) can be determined from Fig.3 (see [IAEA, 1979] for fuller details).



Figure 1. Stopping power as a function of **a**-particle energy for tissue, freshwater and seawater.



Figure 2. Log<sub>10</sub>B as a function of **a**-particle energy for tissue, freshwater and seawater.



Figure 3. Stopping power as a function of the distance from the source travelled in tissue, freshwater and seawater for **a**-particles of initial energy 10 MeV, and residual energies at points along the track.

The point source dose distribution function for  $\beta$ -particles, originally developed empirically by Loevinger *et al.* [1956], has been slightly modified [IAEA, 1979] to give a better fit to the scaled point source absorbed dose distributions for a wide range of radionuclides given by Berger [1971]. The modified point source dose distribution function for  $\beta$ -particles in water or soft tissue is:

$$D_{\beta}(\mathbf{r}) = \frac{k}{(\rho \nu r)^{2}} \left\{ a \left[ 1 - \frac{\rho \nu r}{c} \exp\left(1 - \frac{\rho \nu r}{c}\right) \right] + \rho \nu r \exp\left(1 - \rho \nu r\right) \right\} \ \mu Gy \ h^{-1} \ Bq^{-1}$$
(5)

where:

$$\left[1 - \frac{\rho v r}{c} exp\left(1 - \frac{\rho v r}{c}\right)\right] \equiv 0 \text{ for all } r \geq \frac{c}{\rho v}$$

$$k = \frac{4.59 x 10^{-2} \rho^2 v^3 \overline{E}_{\beta} n_{\beta}}{ac(3-e) + e} \ \mu Gy \ h^{-1} \ Bq^{-1};$$

- $\rho$  is the density of the medium (assumed to be soft tissue, freshwater or seawater with a density of 1 g cm<sup>-3</sup>);
- r is the distance between the point source and the target point (cm);
- v is the apparent absorption coefficient and has the following dependence on the maximum  $\beta^+$  or  $\beta^-$ -particle emission energy:

$$v = 15.1 \ E_{\beta max}^{-1.74} \ cm^2 \ g^{-1}$$
, for 0.0186 Mev  $\leq E_{\beta max} < 0.92$  Mev, and,  
 $v = 17.9 \ E_{\beta max}^{-1.24} \ cm^2 \ g^{-1}$ , for 0.92 Mev  $\leq E_{\beta max} \leq 2.996$  Mev;

- $n_{\beta}$  is the fractional number of  $\beta^{+/-}$ -particles of mean energy  $\overline{E}_{\beta}^{+/-}$  emitted per disintegration;
- a is a dimensionless parameter given by:  $a = 1 + 3.43 \exp(-1.41 \text{ E}_{\beta^{-}\text{max}})$ , for 0.0186 Mev  $\leq \text{ E}_{\beta^{-}\text{max}} \leq 2.996$  Mev, for  $\beta^{-}$ -particles, and, a = 1.12 for  $\beta^{+}$ -particles of all energies; and, c is a dimensionless parameter given by:  $c = 1 + 0.059 \text{ E}_{\beta^{-}\text{max}}^{-0.616}$ , for 0.0186 Mev  $\leq \text{ E}_{\beta^{-}\text{max}} \leq 2.996$  Mev, for  $\beta^{-}$ particles, and,  $c = 1.45 + 0.507 (\text{ E}_{\beta^{+}\text{max}} + 0.4)^{-3.65}$ , for  $0.324 \leq \text{ E}_{\beta^{+}\text{max}} \leq 1.88$  Mev for  $\beta^{+}$ particles (see [IAEA, 1979] for fuller details).

The situation for  $\gamma$ -radiation is more complex due to the existence of several different processes of energy absorption and the fact that scattered radiation represents a significant proportion of the radiation field incident on the target tissue. For the internal contamination of small aquatic organisms (dimensions ~1 cm) with  $\gamma$ -emitting radionuclides, it is reasonable to ignore absorption and scattering and employ the simple inverse square law to describe the radiation field from the point source, thus:

$$D_{\gamma}(r) = 4.59 \text{ x } 10^{-2} \sum_{E\gamma} \frac{\mu E_{\gamma} n_{\gamma}}{\rho r^2} \ \mu Gy \ h^{-1} \ Bq^{-1}$$
(6)

where:

 $\frac{\mu}{\rho}$  is the true mass energy absorption coefficient, at energy  $E_{r}$  of the material (unit density tissue) being irradiated. In the energy range of primary interest (~0.06 - 2.5 Mev) it may be assumed that  $\frac{\mu}{\rho} = 0.03 \text{ cm}^2 \text{ g}^{-1}$  within ±10%;  $n_{\gamma}$  is the fractional number of  $\gamma$ -rays of energy  $E_{\gamma}$  emitted per disintegration; and,

r is the target distance from the point source (cm).

This expression for  $D_{\gamma}$  relates to the positional dependence of the energy absorption from the  $\gamma$ -radiation field and not to the energy deposition in tissue which occurs along the tracks of the secondary electrons. It will, therefore, tend to overestimate the dose rate to small organisms, such as fish eggs or zooplankton, having dimensions of the order of the secondary electron range. This effect can be accommodated by the inclusion of a modifying factor as follows:

$$D_{\gamma}(r) = 4.59 \text{ x } 10^{-2} \frac{\mu n_{\gamma} E_{\gamma}}{\rho r^2} \left[ 1 - \exp\left(\frac{-2.30r}{r_e(0.3E_{\gamma})}\right) \right] \mu \text{Gy } \text{h}^{-1} \text{Bq}^{-1}$$
(7)

where:

 $r_e(0.3E_{\gamma})$  is the range of an electron with energy  $0.3E_{\gamma}$ . The required electron range values are tabulated in [Berger, 1971].

For internal contamination of the larger aquatic organisms with  $\gamma$ -emitting radionuclides, and for photon irradiation from their external environment (water and sediment), the effects of absorption and scattering have to be taken into account. In the human radiological field, this has been achieved by employing the Monte Carlo technique with realistic source and target geometries in the human body [Berger, 1968; Brownell *et al.*, 1968; Ellett and Humes, 1971]. Some of the results that have been obtained can be adapted either directly, or by extrapolation or interpolation, to the geometries of interest for organisms in contaminated environments. The results have been given in terms of the energy-dependent absorbed fraction,  $\Phi(E_{\gamma})$ :

 $\Phi(E_{\gamma}) \equiv$  photon energy absorbed by the target

photon energy emitted by the source

The mean dose rate to the target tissue volume is then:

$$D_{\gamma} = 5.76 \text{ x } 10^{-1} \sum_{E_{\gamma}} \frac{E_{\gamma} n_{\gamma} \Phi(E_{\gamma})}{m} \ \mu \text{Gy } \text{h}^{-1} \text{ Bq}^{-1}$$
(8)

where:

m is the mass of the target; and,

 $n_{\gamma}$  is the fractional number of photons of energy  $E_{\gamma}$  emitted per disintegration.

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In the particular case when the target volume and the source volume are coincidental, then:

$$D_{\gamma} = 5.76 \text{ x } 10^{-1} \text{ } \text{E}_{\gamma} n_{\gamma} \Phi(\text{E}_{\gamma}) \, \mu \text{Gy } \text{h}^{-1} \, (\text{Bq } \text{g}^{-1})^{-1}$$
(9)

The values of  $\Phi(E_{\gamma})$  have been computed for point and distributed sources with varying geometries, with and without the inclusion of a back-scattered contribution from the external environment [Brownell *et al.*, 1968; Ellett and Humes, 1971]. The reciprocal dose theorem can be applied to extend these results [Loevinger *et al.*,1956; Loevinger and Berman, 1968].

#### **3.2.2** Practical application of the point source dose distribution approach

#### For a -radiation.

To justify the use of the  $\alpha$ -particle point source dose distribution (PSDD) function, information on the distributions of the  $\alpha$ -emitting radionuclides on the scale of ~a few µm in tissue is required. In practice, it is very rare that such detailed information is available from laboratory studies, and even less so for either natural or contaminant radionuclides in the environment. In both cases, it would require the application of autoradiographic methods to generate such detailed information. The approach has been, therefore, to assume a uniform distribution of the  $\alpha$ -emitting radionuclides within either the individual tissues or, in the worst case, the whole body. In these circumstances, the absorbed dose rate is given by:

$$D_{\alpha}(\infty) = 5.76 \text{ x } 10^{-1} \sum_{E_{\alpha}} E_{\alpha} n_{\alpha} \ \mu \text{Gy } \text{h}^{-1} (\text{Bq } \text{g}^{-1})^{-1}$$
(10)

where:

nα

is the fractional number of  $\alpha$ -particles emitted with energy  $E_{\alpha}$  per disintegration.

This is the equilibrium absorbed dose rate in a uniformly contaminated medium of effectively infinite extent. For  $\alpha$ -particles with energy  $\leq \sim 10$  Mev and a range in tissue of  $\leq \sim 100 \mu m$ , this is the situation in any tissue or organ with dimensions  $\geq \sim 1$  mm and in which the radionuclide distribution on the micro-scale is unknown. It would also apply equally to internal distributions of  $\alpha$ -emitting radionuclides in either terrestrial or aquatic organisms.

Due to its practical interest and importance, the PSDD function has been applied in the case of developing fish eggs [IAEA, 1979] - a case in which there are reasonable prospects of determining the micro-scale radionuclide distributions, relevant to the natural environment, from laboratory studies. The geometries of the models adopted are given in Fig. 4 ab and the results of the calculations for  $^{239}$ Pu  $\alpha$ -particles are presented in Fig. 5 a-b [IAEA, 1979].



Figure 4. The geometries for the fish egg dosimetry models for calculating the absorbed dose rate:

(a) At a point inside the egg from radionuclides uniformly distributed over the surface; and,

(b) At a point inside the egg from radionuclides uniformly distributed throughout the volume of the egg.



Figure 5. Variation of absorbed dose rate inside the fish egg (r = 0.1 cm) for <sup>239</sup>Pu uniformly distributed:

- (a) over the surface of the egg; and,
- (b) throughout the volume of the egg.

Earlier laboratory studies had indicated that the uptake of plutonium by the developing plaice egg (*Pleuronectes platessa*) was quite low. In terms of the amount and rate of accumulation, two experiments gave differing results (concentration factors (CF) at hatching of 5.8 and 35, or mean CF over the development period of 2.4 and 14), but it was clear that the great majority of the activity (>90%) was present on the outer surface of the eggs [Hetherington *et al.*, 1976; Woodhead, 1984]. On the assumptions that the  $^{239+240}$ Pu concentration in the coastal waters of the northeast Irish Sea was 0.037 Bq I<sup>1</sup> and that a 2 mm diameter egg weighs 4.2 mg and has a surface area of 0.13 cm<sup>2</sup>, then the mean activity per egg would be 3.7 x 10<sup>-7</sup> or 2.2 x 10<sup>-6</sup> Bq (or 3.0 x 10<sup>-6</sup> or 1.7 x 10<sup>-5</sup> Bq cm<sup>-2</sup>). Application of the data in Fig. 5a then indicates that the mean dose rates in the irradiated portion of the egg would have been about 1.7 x 10<sup>-3</sup> or 9.4 x 10<sup>-3</sup> µGy h<sup>1</sup>. (The calculation in Hetherington *et al.* [1976] estimated the mean dose rates to be about 9 x 10<sup>-4</sup> and 4.7 x 10<sup>-3</sup> µGy h<sup>-1</sup> in a spherical shell with inner and outer radii of 0.094 and 0.1 cm. This shell has a width of 60 µm as compared with a width of 39 µm in the PSDD model used in [IAEA, 1979], and this difference largely accounts for the difference in the dose rate estimates.)

More importantly, however, Hetherington *et al.* [1976] pointed out that the actual amount of plutonium activity on the egg had additional implications for the dosimetry. A mean plutonium activity of  $3.7 \times 10^{-7}$  Bq on each egg for the 17 day development period would correspond to one plutonium atom disintegration every 2.7 x  $10^{6}$  seconds, or ~31 days, on average, or a mean disintegration rate per egg during embryonic development of 0.54. The Poisson distribution can, therefore, be applied to determine the proportion of eggs experiencing 0, 1, 2 and 3+ disintegrations, i.e., 0.58, 0.32, 0.09 and 0.01, respectively. Further, assuming that there is a 50% chance that any given  $\alpha$ -particle emitted by plutonium on the egg surface will penetrate the egg, the binomial distribution can be used to give the proportions of the eggs into which 0, 1 or 2  $\alpha$ -particles will penetrate, i.e., 0.76 (equivalent to receiving no  $\alpha$ -radiation dose), 0.21 and 0.03, respectively. In these circumstances, it is clear that the estimated macroscopic dose rate is quite meaningless. Indeed, it is not immediately apparent how such radiation exposures, consisting of one or a few particle tracks passing into small autonomous biological entities, should be interpreted. These considerations will apply to any small organism (particularly phyto- and zooplankton, and the soil micro-fauna), organ or tissue.

The approach, demonstrated here for fish eggs, can be adapted for the estimation of the  $\alpha$ -radiation exposure of any small organism in either the terrestrial or the aquatic environments.

#### For **b**-radiation.

The ranges, in soft tissue, of the  $\beta$ -particles emitted from natural and artificial radionuclides extend up to about 2 cm. The assessment of the dose rate resulting from the incorporation of  $\beta$ -emitters into tissue requires, therefore, information on their internal distributions on the scale of a few mm. Again, as for the  $\alpha$ -emitters, such information is rarely available and a uniform distribution must be assumed in either the tissues or the whole body. If the organ or organism has dimensions greater than ~2 cm, then the dose rate at the centre (assuming a uniform radionuclide distribution) can be approximated by:

$$D_{\beta}(\infty) = 5.76 \text{ x } 10^{-1} \sum_{\overline{E}\beta} \overline{E}_{\beta} n_{\beta} \ \mu Gy \ h^{-1} \ (Bq \ g^{-1})^{-1}$$
(11)

where:

$$n_{\beta}$$
 is the fractional number of  $\beta$ -particles emitted with mean energy  $E_{\beta}$  per disintegration.

For smaller organs and organisms, use may be made of the  $\beta$ -radiation PSDD function.

The  $\beta$ -radiation PSDD function has been applied to the case of developing fish eggs [Woodhead, 1970; IAEA, 1979]. The geometry of the models is as given in Fig. 4 a-b and the results of the calculations for a number of variants are given in Fig. 6 a-d. In Fig. 6a, the influence of the egg size and the maximum  $\beta$ -particle energy on the dose rate at the putative position of the developing embryo (0.1x the egg radius from the egg surface) can be seen for radionuclides on the egg surface. The dose rate per unit activity (Bq  $\text{cm}^2$  of egg surface) increases as the egg radius decreases due to the increased proportion of the egg surface that is within the range of the low energy  $\beta$ -particles. For the three egg sizes considered, the dose rates converge at the higher  $\beta$ -energies because essentially all of the energy is deposited at the target site by the low LET portion of the initial part of the  $\beta$ particle tracks and the increasing activity on the egg shell with increasing egg radius, although all within range of the target, is approximately (and coincidentally) counteracted by the increasing distance (0.1 x radius) of the target point from the egg surface. In Fig. 6b, the variation of the dose rate through the egg is shown for the environmentally important radionuclide pair, <sup>90</sup>Sr-<sup>90</sup>Y, for two different egg sizes. As is to be expected, the egg size has a significant influence on the radial variation of the dose rate for the low energy  ${}^{90}$ Sr  $\beta$ -particles but is of little consequence for the higher energy <sup>90</sup>Y radiation (but bear in mind that, in absolute dimensions, the curves for the smaller egg would be contracted towards the ordinate relative to those for the larger egg). Fig. 6 c and d show the corresponding dependencies for the case of the activity uniformly distributed throughout the egg. For an egg at more than a few cm from any water boundary, the dose rate at the point P inside the egg from  $\beta$ -emitters in the surrounding water is simply:

$$D_{\beta}(P) = D_{\beta}(\infty) - D_{\beta}(P)_{v} \quad \mu Gy \ h^{-1} \ (Bq \ g^{-1})^{-1}$$
(12)

where:

 $D_{\beta}(P)_{v}$  is the dose rate at the point P from activity uniformly distributed within the egg (see Fig 6. c and d) evaluated at the concentration of the radionuclide in the water.

The full expressions and greater detail are given in [IAEA, 1979].



Figure 6. (a) Variation of absorbed dose rate (at OP = 0.9r) as a function of the maximum **b**particle energy for radionuclides uniformly distributed over the surface of eggs of differing radii.

(b) Variation of **b**-particle absorbed dose rate within the egg for <sup>90</sup>Sr-<sup>90</sup>Y uniformly distributed over the surface of eggs of differing radii.


Figure 6. (c) Variation of absorbed dose rate (at OP = 0.9r) as a function of the maximum **b**-particle energy for radionuclides uniformly distributed throughout the volume of eggs of differing radii.

(d) Variation of **b**-particle absorbed dose rate within the egg for <sup>90</sup>Sr-<sup>90</sup>Y uniformly distributed throughout the volume of eggs of differing radii.

For organisms or organs with dimensions of the same order as, or less than, the ranges of the  $\beta$ -radiation in tissue (< ~ 2 cm), the dose rate at their centre will be less than  $D_{\beta}(\infty)$  evaluated at the radionuclide concentration in the organism whole body or the organ. In this case, the organism or organ is divided into a sphere and a succession of partial shells centred on the point of interest (see Fig. 7). The dose rate at that point is then simply the sum of the contributions from the sphere and the individual segments evaluated at the radionuclide concentration in the tissue using the  $\beta$ -particle PSDD. Using this approach, the dose rates at the centre of some of the reference organisms have been determined; the results, as a fraction of the corresponding  $D_{\beta}(\infty)$ , are given in Fig. 8 for the small crustacean, the mollusc and the large crustacean, as a function of the mean  $\beta$ -particle energy. In [NCRP, 1991], the small crustacean model was taken to be applicable for a small insect, and the mollusc model for a large insect. For the larger organisms, if there is no relevant information on the differential distribution of the radionuclides within the potentially important target tissues, the  $\beta$ -radiation dose rate to tissues at greater than ~ 2cm from the body surface is effectively  $D_{\beta}(\infty)$  evaluated at the mean radionuclide concentration in the whole body. Again, the  $\beta$ -particle dose rate at the centre of the organism from activity in the water is given by:

$$D_{\beta}(0) = D_{\beta}(\infty) - D_{\beta}(0)_{v} \quad \mu Gy \ h^{-1} (Bq \ g^{-1})^{-1}$$
(13)

where:

 $D_{\beta}(0)_{v}$ , the dose rate at the centre of the organism from activity uniformly distributed within the body (see Fig 8), and  $D_{\beta}(\infty)$  are evaluated at the concentration of the radionuclide in the water;

and from activity in the sediment is given by

$$D_{\beta}(0) = 0.5 \{ D_{\beta}(\infty) - D_{\beta}(0)_{v} \} \ \mu Gy \ h^{-1} \ (Bq \ g^{-1})^{-1}$$
(14)

where:

 $D_{\beta}(0)_{v}$ , the dose rate at the centre of the organism from activity uniformly distributed within the body (see Fig 8), and  $D_{\beta}(\infty)$  are evaluated at the concentration of the radionuclide in the sediment, assumed to be uniform over distances of the order of the maximum  $\beta$ -particle range (~2 cm).

The influence of differential radionuclide distributions within the body has been investigated in a general way [Pentreath and Woodhead, 1988]. Here, the cases of 16 mg and 1 g ellipsoidal target organs centrally placed in 1 and 1000 g ellipsoidal bodies, respectively, are considered (i.e., so that the data in Fig 8. may be used). It is assumed that the radionuclide concentration in the organ is either 0.1x (discrimination) or 10x (preferential accumulation) the assumed mean whole body concentration of 1 Bq g<sup>-1</sup> (this allows the calculation of the radionuclide concentration in the remainder of the body surrounding the organ), and that the distributions are, otherwise, uniform. Fig. 9 provides the results in terms of (sub-) multiples of the dose rate at the centre of the body that would have resulted from a uniform distribution of the same total quantity of radionuclide throughout the whole body. At low  $\beta$ -particle energies, where the ranges are less than, or of the same order as, the dimensions of the target organ, but at higher energies (and longer ranges) the dose rate falls below proportionality for preferential accumulation in the target (curves A and C), and increases above proportionality where there is discrimination (curves B and D). Off-centre organs at less than the  $\beta$ -

particle range from the body surface can be treated using the same approach, but this has yet to be done.



Figure 7. The geometry of the dosimetry model adopted to represent a small mollusc or large insect, and a schematic indication of the partial spherical shell method for estimating the **b**-particle absorbed dose rate at the centre of the body from a uniform distribution of radionuclides throughout the volume.



Figure 8. **b**-particle dose rate, as a fraction of  $D_b(\mathbf{X})$ , at the centre of the geometries taken to represent the bodies of aquatic organisms, from a uniform distribution of radionuclides throughout the volume, as a function of the maximum **b**-particle energy.



- Figure 9. The influence of preferential radionuclide accumulation or discrimination on the **b**-particle absorbed dose rate at the centre of a small centrally-placed organ in a larger body. The absorbed dose rate at the centre of the organ is given relative to that which would be delivered by the assumed uniform whole body concentration of 1 Bq g<sup>-1</sup>.
  - Curve A. A 1 g organ centrally-placed in a 1000 g body with preferential accumulation to a radionuclide concentration 10 x the assumed mean whole body radionuclide concentration of 1 Bq g<sup>-1</sup>.
  - Curve B. A 1 g organ centrally-placed in a 1000 g body with preferential discrimination giving a radionuclide concentration 0.1 x the assumed mean whole body radionuclide concentration of 1 Bq g<sup>-1</sup>.
  - Curve C. A 16 mg organ centrally-placed in a 1 g body with preferential accumulation to a radionuclide concentration 10 x the assumed mean whole body radionuclide concentration of 1 Bq g<sup>-1</sup>.
  - Curve D. A 16 mg organ centrally-placed in a 1 g body with preferential discrimination giving a radionuclide concentration 0.1 x the assumed mean whole body radionuclide concentration of 1 Ra g<sup>-1</sup>

The particles generated by  $\beta$ -decay have a distribution of energies ranging from zero to a variable  $E_{\beta max}$  that depends on the nuclide and its decay scheme. Auger electrons and the electrons produced by the internal conversion of  $\gamma$ -rays are, however, mono-energetic. The former are generally of such low energy that their ranges in tissue are less than the smallest dimensions of the majority of organisms and organs considered as targets for dosimetry; it is appropriate, therefore, to use  $D(\infty)$  evaluated at the Auger emission energy for the absorbed dose rate. The energies and ranges of conversion electrons can be significant, e.g., 1.82 MeV and ~ 0.9 cm, respectively, for <sup>88</sup>Y, and the dose rate at the centre of a small organism from internal sources will be less than  $D(\infty)$ . The data in [Berger, 1971] can be used to estimate the absorbed fractions for mono-energetic electrons in small organs and organisms (see Fig. 10).



# Figure 10. Monoenergetic electron dose rate, as a fraction of $D_e(\mathbf{X})$ , at the centre of the geometries taken to represent the bodies of aquatic organisms, from a uniform distribution of radionuclides throughout the volume, as a function of the monoenergetic electron energy.

The same approach is appropriate for internal sources of electrons and  $\beta$ -particles in terrestrial organisms (as noted above, the geometry of the small crustacean has been used to represent a small insect and that of the mollusc, a large insect). The estimation of the dose rate from external sources of electrons and  $\beta$ -particles in the terrestrial environment is substantially more complex due to the inhomogeneous density distribution - it is likely that the radiation flux will be incident on the organism after passing through the air and, possibly, plant material, from sources that are cm to metres distant. In these circumstances the PSDD functions are not applicable and an alternative approach remains to be developed.

#### For g-radiation.

Fish eggs are generally sufficiently small that the  $\gamma$ -ray PSDD function (Eq. 7) can be employed. The results for <sup>137</sup>Cs on the surface of, and uniformly distributed within, eggs of two different diameters are given in Fig. 11 a and b, together with those from the application of the simple inverse square law for comparison [IAEA, 1979]. As would be expected, the inclusion of the electron build-up factor reduces the dose rate as compared with the simple inverse square law.

In the more usual situation, in which it must be assumed that the contaminant  $\gamma$ -emitting radionuclides are uniformly distributed throughout organisms with dimensions greater than electron build-up range, the published data [Brownell *et al.*, 1968; Ellett and Humes, 1971] have been used (with interpolation or extrapolation) to determine the absorbed fractions ( $\Phi$ ) for a number of the generic aquatic organisms listed in Table 1. The results are given in Fig. 12 and it can be seen that the smaller the organism, the smaller is the amount of the  $\gamma$ -ray energy emitted from internal sources that is absorbed within the organism. The mean dose rate to the organism from the internal sources of  $\gamma$ -radiation can then be calculated from Eq. 9.





- (a) over the surface of the egg; and,
- (b) throughout the volume of the egg.



Figure 12. **g**-ray absorbed fractions for aquatic organisms for a uniform distribution of radionuclides throughout the whole body.

The effects of the differential accumulation of  $\gamma$ -emitting radionuclides in internal organs has been considered, in a general way, by Pentreath and Woodhead [1988]. Use was made of the published values for the absorbed dose fractions for point and distributed sources in 1 g and 100 g elongated ellipsoids [Ellett and Humes, 1971] and the reciprocity theorem [Brownell *et al.*, 1968]. This showed that, if 100% of the total body burden of a nuclide is present in a centrally-placed target organ of 1% (1 g) of the body weight, then the  $\gamma$ -ray dose rate to the organ is approximately 30x greater than the mean dose rate to the whole body (100 g) from the same total quantity of activity uniformly distributed. In a more usual case, in which 25% of the total body burden resides within the organ, the dose rate would be increased by a factor of 5; for larger organisms, the factor would be greater, and for smaller organisms, less. Similarly, it can be shown that if the radionuclide is not accumulated at all by the target organ, the  $\gamma$ -ray dose rate to the organ is little different to the mean dose rate to the whole body from the entire, otherwise uniformly distributed, body burden.

The mean  $\gamma$ -ray dose rate to the organism from activity in the water (assuming a position >~1m from the sea surface or the seabed) is given by:

$$D_{\gamma}(M) = D_{\gamma}(\infty) - D_{\gamma}(M)_{\nu} \quad \mu Gy \ h^{-1} \ (Bq \ g^{-1})^{-1}$$
(15)

where:

 $D_{\gamma}(M)_{\nu}$ , the mean dose rate to the organism from activity uniformly distributed within the body, and  $D_{\gamma}(\infty)$  are evaluated at the concentration of the radionuclide in the water;

and from activity in the sediment is given by

$$D_{\gamma}(M) = 0.5 \{ D_{\gamma}(\infty) - D_{\gamma}(M)_{v} \} \ \mu Gy \ h^{-1} \ (Bq \ g^{-1})^{-1}$$
(16)

where:

 $D_{\gamma}(M)_{v}$ , the mean dose rate to the organism from activity uniformly distributed within the body, and  $D_{\gamma}(\infty)$  are evaluated at the concentration of the radionuclide in the sediment.

Eq. 16 implicitly assumes that the  $\gamma$ -emitting radionuclides are uniformly distributed in the seabed to a depth greater than the mean free path for absorption (i.e., >~1m), hence the inclusion of the factor of 0.5 (for a uniformly contaminated, effectively semi-infinite space). In many situations, it is known that the concentrations of the anthropogenic radionuclides decline with depth (half-value depths up to 10s of cm) due to radioactive decay, limited input histories, low bioturbation and/or sedimentation rates, etc., and in these cases, a factor of 0.25 (rather than 0.5) yields more realistic estimates of the  $\gamma$ -ray dose rate from the seabed [IAEA, 1976].

The approach of using the published data to estimate the absorbed fractions could be adapted for internal sources of  $\gamma$ -rays for the geometries representing the generic terrestrial organisms. For external  $\gamma$ -ray sources, however, the absorption mean free path in air of 10s of m and the presence of density inhomogeneities, e.g., vegetation and soil surface topography, mean that an alternative approach, probably employing Monte Carlo methods for generic environmental geometries, must be developed.

The techniques discussed in this Section for estimating radiation dose rates have been applied in a number of instances for existing, or potentially, contaminated environments. Rather than give extensive Tables of the results, the references are summarized in Table 3. A number of the models listed in this summary correspond to those suggested for the reference organisms in Tables 1 and 2.

#### Organism References Pelagic phytoplankton Woodhead, 1973a; IAEA, 1976. Pelagic zooplankton, small Woodhead, 1973a; IAEA, 1976, 1988, 1998a, b; benthic crustacean, small Hoppenheit et al., 1980; OECD/NEA, 1985; insect or larva Pentreath and Woodhead, 1988; NCRP, 1991; St-Pierre et al., 1999. Fish eggs Woodhead, 1970; Hetherington et al., 1976. Benthic mollusc IAEA, 1976, 1988, 1998a, b; OECD/NEA, 1985; Pentreath and Woodhead, 1988; NCRP, 1991; St-Pierre et al., 1999; Woodhead, 1973a. Large benthic crustacean IAEA, 1976, 1988, 1998a, b; OECD/NEA, 1985; Pentreath and Woodhead, 1988; Woodhead, 1973a. Pelagic fish IAEA, 1976, 1988, 1998a, b; OECD/NEA, 1985; Pentreath and Woodhead, 1988; Woodhead, 1974; St-Pierre et al., 1999. Benthic fish IAEA, 1976, 1988; OECD/NEA, 1985; Pentreath et al., 1973; Pentreath and Woodhead, 1988; Woodhead, 1973a, b, 1974; St-Pierre et al., 1999. Pentreath and Woodhead, 1988; Calmet et al., Seal, dolphin 1992; IAEA, 1998b. Whale IAEA, 1998b. Woodhead, 1986; NCRP, 1991, IAEA, 1998b; Duck, coot, gull St-Pierre et al., 1999. Alligator NCRP, 1991. Turtle NCRP, 1991. Small polychaete worm IAEA, 1998a. Large gastropod mollusc IAEA, 1998a. Pearl oyster IAEA, 1998a.

# Table 3. A summary of references giving results from the use of environmentaldosimetry models described in Sections 3.2.1. and 3.2.2.

#### 3.2.3 The terrestrial environment

A cursory examination of the summary in Table 3 immediately shows that the aquatic environment has been the most thoroughly studied; this is quite simply a consequence of the extensive use that has been made of surface waters for the disposal of low-level liquid effluents, and the deep ocean as a repository for low-level solid radioactive wastes. Most nuclear sites do, however, make discharges to the atmosphere. These have implications for the terrestrial biosphere in terms of radiation exposure from the radionuclides, either when they are airborne or following dry/wet deposition, and there has been some development of the requisite dosimetry models.

It has been noted at several points in Sections 3.2.1 and 3.2.2 that the terrestrial system is more complex, in terms of the dosimetry of external sources, due to both the much extended ranges of the  $\beta$ - and  $\gamma$ -radiations in air and the presence of the substantial density variations between air, soil and plant and animal tissues. In the aquatic environment, it is reasonable to assume an equivalence (at the level of accuracy required for environmental impact assessment) between the surrounding water and soft tissue in terms of radiation absorption and scattering properties.

The problems of estimating the absorbed dose to terrestrial animals from external sources of  $\gamma$ -radiation have been discussed in [UNSCEAR,1996]. It was concluded that the simple derivation of the absorbed dose rate from an estimate of air kerma would not be possible because it would depend on the assumptions of photon field uniformity, secondary electron equilibrium and no photon scattering; these would be unlikely to be valid in a contaminated environment with inhomogeneous distributions of both the radionuclides and material densities.

Nevertheless, Jacobi and Paretzke [1986] have made approximate estimates of the absorbed dose rates to the leaves of deciduous trees and the needles of coniferous trees from external sources of  $\beta$ - and  $\gamma$ -radiation. They assumed that there was radiation equilibrium in air, that scattering could be neglected, and that the ratios of the mass energy absorption coefficients and electron stopping powers in air and leaf/needle tissue could be taken as unity. Under these assumptions the absorbed dose rates to the leaf/needle tissue from external  $\beta$ - and  $\gamma$ -radiations are:

$$D_{\beta} = g_{\beta} C_{a} \ \mu Gy \ h^{-1} \tag{17}$$

and

$$D_{\gamma} = g_{\gamma} K_a \ \mu Gy \ h^{-1} \tag{18}$$

where:

 $C_a\,$  and  $\,K_a$  are, respectively, the cema and kerma rates in air, in  $\mu Gy\,h^{\text{-}1}\!;$  and,

 $g_{\beta}$  and  $g_{\gamma}$  are dimensionless geometrical factors to take account of the attenuation of the incident radiations in the tissue.

The value of  $g_{\beta}$  was taken to vary between unity for high energy  $\beta$ -particles and zero for the low energy  $\beta$ -particles unable to penetrate the leaf/needle cuticle and irradiate the cell growth layer at around 0.1 mm depth; the value of  $g_{\gamma}$  was taken to be unity and independent of  $\gamma$ -ray energy. A complementary approach was used to estimate the dose rate from internal sources. It is of interest to note that the important situation of surface contamination was not addressed. A deficiency in this dosimetry model is that it implicitly assumes that the leaf or needle is an isolated entity and does not, therefore, take account of sources in other parts of the same tree or the effects of self-shielding. It might also be more relevant to estimate the dose rates to the growing buds rather than the mature leaves/needles although this does, of course, depend on the radiation effect endpoint of interest.

A similar degree of simplification was adopted in [IAEA, 1992] in estimating the absorbed dose rates to a generic plant and animal from internal and external sources. For internal sources, the D( $\infty$ ) value for the radionuclide was reduced by a geometrical factor relevant to the radiation type and energy, i.e., unity for  $\alpha$ -particles; unity for  $\beta$ -particles except in the case of <sup>32</sup>P for which a value of 0.5 was adopted; and, 0.1 for  $\gamma$ -rays. The dose rate to plant tissues from external sources of  $\gamma$ -rays deposited on the ground was estimated to be 3.3 times that for humans (available from published sources). This value of 3.3 takes account of the variations in geometry and occupancy between plants and humans. For external sources of  $\beta$ -radiation, it was concluded that, even for high energy emitters such as <sup>32</sup>P and <sup>90</sup>Y, the exposure would be less than 10% of that from the contamination on, and in, the plant. This contribution was, therefore, ignored.

A similar approach was followed for the generic animal. For internal sources, the geometrical factors for the reproductive tissues for  $\alpha$ -,  $\beta$ - and  $\gamma$ -radiation were taken to be unity, unity and 0.3, respectively. The dose to animal tissues from external sources of  $\gamma$ -radiation was assumed to be the same as that for plants.

Amiro and Zach [1993] and Amiro [1997] have estimated the dose conversion factors (DCF) for a number of generic terrestrial organisms - a plant, a mammal and a bird (in addition to pelagic and benthic freshwater fish) - from internal and external sources of radiation. The underlying dosimetry models were generalised and were made deliberately conservative to ensure that any consequent action provided the environment with the benefit of the doubt. For the radionuclides taken up into, and assumed to be uniformly distributed within, the organisms, it was assumed that all the emitted energy was absorbed within the tissue, i.e., the absorbed dose rate was equivalent to  $D_{\alpha,\beta,\gamma}(\infty)$ evaluated at the radionuclide concentration in tissue (e.g., Eq. 10). For organisms with dimensions > 2 cm, this is a reasonable assumption for  $\alpha$ - and  $\beta$ -particles; for the majority of organisms, however, it would lead to substantial over-estimates of the dose rate from the internal  $\gamma$ -emitters. (Note that in [Amiro and Zach, 1993] the dose rate to the animal thyroid from <sup>129</sup>I was increased by a factor of 10 to account for the preferential accumulation of this element in the organ; this approach was not carried over into [Amiro, 1997]). There are many potential sources of external exposure in a contaminated environment, with different radionuclide concentrations in each compartment depending on their varying biogeochemical behaviours, and significant simplifications had to be made. Although the behaviour of the individual species of animals and birds also has a significant influence on their potential radiation exposure, this was neglected and three generic situations were considered for the terrestrial environment:

*Immersion in contaminated air* - It was assumed that the target organism was situated at 1 m above a plane boundary (the soil surface) in a semi-infinite, uniformly contaminated volume of air

with a density of 1.189 kg m<sup>-3</sup>. This particular geometry was originally developed for humans [Holford, 1988, 1989] and will give conservatively high values of dose rate for organisms that predominantly live closer to the ground surface; it will, however, underestimate (by a factor < 2) the exposure of organisms, e.g., the swift, that spend a large proportion of their lives high in the air.

*Immersion in contaminated soil* - It was assumed that the target organism was situated 0.1 m below the plane surface (boundary between soil and air) of a semi-infinite, uniformly contaminated body of soil. This geometry, appropriate for plant roots, litter fauna and burrowing animals, will give conservatively high values of the exposure from the soil source to plants, and to animals that live on, or above, the soil surface.

*Immersion in contaminated vegetation* - The geometry of the model is as above for air contamination, i.e., the target organism is assumed to be 1 m above the soil surface. It was also assumed, however, that the plant yield was 1 kg m<sup>-2</sup> with a mean plant height of 1 m so that the plant density is 1 kg wet biomass m<sup>3</sup> of air. The vegetation immersion DCF value (Gy a<sup>-1</sup> Bq<sup>-1</sup> kg wet biomass) was then simply obtained as the product of the air immersion DCF (Gy a<sup>-1</sup> Bq<sup>-1</sup> m<sup>-3</sup>) and the vegetation density (1 kg wet biomass m<sup>3</sup> of air). This will give conservatively high estimates of the dose rate from the contaminated vegetation in most instances because it implicitly assumes that the vegetation (together with its associated radionuclides) is uniformly distributed, at a density of 1 kg wet biomass m<sup>-3</sup> of air, in a semi-infinite volume.

Using the data in [Holford, 1988, 1989], DCF values, derived from these models for 99 radionuclides of interest in the context of a geological waste repository, have been tabulated [Amiro 1997].

#### **3.2.4** General ranges of environmental absorbed dose rates

Although the detailed results of the application of the dosimetry models to real, or potentially, contaminated environments have not been given (see Table 3 for references), it is possible to indicate the general limits of the radiation exposures for a number of situations [summarized from UNSCEAR, 1996]. For the natural background, the absorbed dose rates are normally up to ~1  $\mu$ Gy h<sup>1</sup> but, exceptionally, may be up to  $2x10^2 \mu$ Gy h<sup>1</sup>. In all situations,  $\alpha$ -particles appear to contribute a substantial proportion of the total absorbed dose rate (the <sup>222</sup>Rn + short-lived daughters, and <sup>210</sup>Po). In environments receiving radioactive wastes, the absorbed dose rates from the contamination are generally < 10<sup>2</sup>  $\mu$ Gy h<sup>1</sup>, but may, exceptionally, rise to ~10<sup>3</sup>  $\mu$ Gy h<sup>-1</sup>. The highest environmental dose rates have followed accidental releases of radionuclides. In the southeastern Urals (1957) and at Chernobyl (1986), the initial absorbed dose rates were > 10<sup>4</sup>  $\mu$ Gy h<sup>-1</sup> (and, locally, > 10<sup>5</sup>  $\mu$ Gy h<sup>-1</sup>); these have declined to current values of < ~1.5x10<sup>2</sup> and ~10<sup>2</sup>  $\mu$ Gy h<sup>1</sup>, respectively. The significance of these ranges of dose rate is that they indicate the domain of the dose rate/response relationship over which information for the biological endpoints of interest is required.

#### **3.2.5** Future developments

Aside from the clear requirement to develop dosimetry models applicable to the terrestrial environment, there is a number of developments that can be foreseen. Once the generic organisms for the marine, freshwater and terrestrial environments have been confirmed (see section 2) and the corresponding geometric models defined, the conceptual outline of the process of absorbed dose rate calculation can be developed. In the case of the marine environment [Pentreath and Woodhead, 1988], it was concluded that the basic datum should be the concentration of the radionuclide in the seawater, and for the purpose of estimating the dose factors, this concentration was taken to be 1 Bq m<sup>3</sup>. From this, the available data on the uptake of the radionuclide into the organisms (the equilibrium concentration factor - CF ( $10^{-3}$  m<sup>3</sup> kg<sup>-1</sup> wet weight) - probably at the level of the whole body, but at the organ/tissue level if the relevant data are available) and by sediment (the equilibrium distribution coefficient - k<sub>d</sub> ( $10^{-3}$  m<sup>3</sup> kg<sup>-1</sup> on a dry weight basis)) [see IAEA, 1985] would be applied to determine the radionuclide concentrations in these compartments. Hence:

radionuclide concentration in seawater:

 $C_w = 1 \text{ Bq m}^{-3};$ 

radionuclide concentration in organism (whole body) is:  $C_{wb} = CF_{wb} \; Bq \; kg^{-1}; \; and,$ 

radionuclide concentration in sediment (wet) is:

$$C_{s} = \frac{C_{w} \left[ f'(\rho_{s} k_{d} - 1) + 1 \right]}{f'(\rho_{s} - \rho_{w}) + \rho_{w}} Bq kg^{-1} wet,$$

where: f is the fraction of solids in the sediment (taken to be 0.4 by volume); and,

 $\rho_s$  and  $\rho_w$  are the densities of the solids and water, and taken to be  $1.5 \times 10^3$  and  $1.0 \times 10^3$  kg m<sup>-3</sup>, respectively.

These radionuclide concentrations were then, together with the data in Fig. 8, 9 and 12, and the radiation emission characteristics of the radionuclides, the input data for the calculation of the dose factors tabulated in [Pentreath and Woodhead, 1988]. (Note that the dose factors in this reference are given in terms of mSv h<sup>1</sup> per Bq m<sup>3</sup> of seawater, i.e., the  $\alpha$ -particle, and the  $\beta$ -particle and  $\gamma$ -ray, components of the absorbed dose rate were multiplied by radiation weighting factors of 20 and 1, respectively, to give the dose equivalent rate. It was recognized that this procedure was open to argument and, as discussed in section 3.1.2. above, this is a question that has still to be resolved.) Pentreath and Woodhead [1988] did not consider all of the generic marine organisms suggested in Table 1, and the calculation of the dose factors for the additional organisms, if confirmed as necessary, remains to be done. Essentially the same approach would be appropriate for the freshwater environment.

In the case of the terrestrial environment, there is a number of steps to be undertaken:

- 1. Decide on the generic organisms that are broadly representative of the European region taking account of the factors discussed in section 2 (some are suggested in Table 1);
- 2. Define the geometries that will represent these generic plants and animals, and the environmental compartments that will be the sources of radiation exposure, e.g., radionuclides on and in the plant foliage; on the surface, and in the surface layers, of the soil; and, in the animals.
- 3. Use these dosimetry models to generate the terrestrial equivalents of the data presented in Fig. 8, 9 and 12.
- 4. Decide on the basic radionuclide concentration datum appropriate to the terrestrial environment as discussed at the start of this section i.e., either a unit concentration in air (Bq  $m^3$ ) or a unit deposition density (Bq  $m^2$ ); it may turn out to be necessary to use both in different circumstances. From these basic data, information on interception rates, deposition rates and transfer factors will be required to generate the radionuclide concentrations in the compartments that will give rise to the radiation exposures.
- 5. Calculate the dose factors for the generic organisms.

Thus far, an equilibrium situation has been assumed, i.e., the time dependence of the evolution of radionuclide distributions has not been included in the calculations. This is an assumption that needs to be examined to determine circumstances in which it applies, e.g., a time-averaged CF was used in the estimation of the dose rates to developing fish embryos for which the accumulation half time was of the same order as the development period [Woodhead, 1970; Hetherington *et al.*, 1976], and it would almost certainly be inappropriate for the situation of an accidental release.

In terms of realising a comprehensive set of dose factors for the range of generic organisms that is to be identified as appropriate to the European region, the ideal would be an inter-linking hierarchy of spreadsheets that:

- calculated the absorbed fractions for each generic organism geometry, the identified internal and external sources of radiation, and each radiation type (α-, β-particles, x-, γ-rays or monoenergetic electrons). It would be necessary to have individual worksheets to calculate:
- the parameter values for the point source dose distribution functions, i.e., for αparticles: A, B and R( $E_{\alpha em}$ ) in Eq. 4 - probably by interpolation from discrete values taken from Fig. 1, 2 and 3; for β-particles: v, a and c in Eq. 5, and also, for the smaller organism geometries, the proportions of the spherical shells of increasing radii centred on the target point and included within the geometry; and, for x- and γ-rays, and the smaller organisms:  $r_e(0.3E_{\gamma})$  in Eq. 7 - probably by interpolation between discrete values taken from [Berger, 1971]; for the larger organisms, the discrete values of absorbed fraction, as in Fig.12, would be used as the basis of interpolation;

- contained the radiation emission characteristics for each of the radionuclides of interest in an addressable form, i.e., the values of  $n_r$ , the fractional number of emissions of type r ( $\alpha$ -,  $\beta$ -particles, x-,  $\gamma$ -rays or monoenergetic electrons) and (mean) energy  $E_r$  MeV per disintegration in a format that is recoverable for use in calculations (note that the radionuclide emission data in [ICRP, 1983] are available in electronic form and might be used either directly, or through adaptation to the spreadsheet format);
- contained the data on radionuclide behaviour in terms of CF, k<sub>d</sub>, interception rate, deposition rate and transfer factor values; and,
- combined these data on absorbed fractions, the radiation emission characteristics for each radionuclide, and radionuclide behaviour to generate the corresponding dose factors.

The advantages of having all the models underlying the dose factor values on spreadsheets are threefold: the calculations would be relatively transparent; they could be easily updated as new or improved information on radionuclide behaviour in the environment became available and, the implicit framework would be immediately available for adaptation to additional generic (or site-specific) target organs or organisms.

## 3.3 Conclusions

It may reasonably be concluded from the information discussed in this Section that there is a substantial basis for the further development of radiation dosimetry models appropriate to native wild organisms in contaminated environments. The greatest effort probably needs to be directed at the identification of relevant generic organisms and habitats for the terrestrial environment. Once this has been completed, the remaining work outlined in section 3.2.5. could be undertaken.

It is almost certainly the case that improvements in the accuracy and precision of the estimates of the dose rates to native wild organisms from radioactive waste management practices will be limited by the availability of the relevant information on the behaviour and distribution (in time and space) of the radionuclides both external to, and within, the organisms, rather than the complexity of the dosimetry models that could be developed. The acquisition of improved input data for the dosimetry models would be a resource-expensive activity, and it is likely that this factor will enforce simplifying assumptions in environmental dosimetry for the foreseeable future.

# 4. SUMMARY AND CONCLUSIONS

## 4.1 The present position

A previous report [Woodhead, 1998] has shown that, in principle at least, the Environment Act 1995 (including transferred powers) provides a statutory basis for the establishment of criteria, and the application of controls, for the protection of the natural environment from any incremental radiation exposure arising from radioactive waste management activities. It also showed that the ability to estimate the consequent radiation exposure of native wild organisms in the contaminated environment would be an essential component of the framework to provide for environmental protection. A very brief outline was given of some existing approaches to the estimation of the radiation dose rates to a variety of wild organisms and the results summarized. Due to the practical impossibility of estimating the radiation exposure of individuals of every species of flora and fauna inhabiting a contaminated area (the numbers involved and the input data requirements), it has always been recognized that it would be necessary to limit the radiation dose rate assessments to reference (or generic) organisms considered to be broadly representative of the area. The basis for the selection of such reference organisms was not discussed although it was determined that the individual plant or animal, and/or their internal biological processes, e.g., gametogenesis, would represent the appropriate focus for measures to protect the environment.

# 4.2 Dosimetry targets and reference organisms

For a framework for environmental protection to have credibility, it is necessary that the dose rate (and risk) assessment should include a sufficient variety of organisms that the full range of both environmental dose rates, and potential sensitivities to the effects of radiation, are encompassed. Although the potential influences on the degree and significance of radiation exposure have been discussed in terms of biological, physical and geochemical factors, it was determined that the selection of the representative reference organisms would more usefully be based on considerations of:

- *Radioecological sensitivity.* The dose rates to the native flora and fauna in a contaminated environment will be influenced by habitat preference, behaviour and innate capacity to accumulate radionuclides in relation to the temporal and spatial variabilities of the radionuclide distributions. The latter are governed by the chemical nature of the radioactive elements and their consequent interactions with environmental processes, and their half-lives. This selection basis is likely to result in the choice of representative species from the main trophic levels.
- *Radiosensitivity*. There is considerable evidence that the response to irradiation varies between species, between organs and tissues within a given type of organism and between the different stages in the life cycle of many individual species. In addition to providing a basis for selecting reference organisms, this also serves to identify potentially important dosimetry targets at the sub-individual level.
- *Ecological sensitivity.* In the majority of ecosystems the native flora and fauna are grouped into communities as a consequence of the multiple interactions between the requirements of the organisms and the physical and geochemical properties of the local

environment. Within these communities there is frequently an apparent hierarchy in the roles of the organisms in the functioning of the community, e.g., the importance of primary producers; this provides a third potential basis for the selection of relevant reference organisms and is, again, likely to result in the choice of examples from each trophic level.

The application of these selection criteria led to the choice of the reference organisms set out in Table 1, although it is not suggested that these are necessarily comprehensive.

#### 4.3 Dosimetry models

A selection of the dosimetry models that have been employed to assess the radiation exposure of native organisms in contaminated environments has been described. The models have been most highly developed for the marine environment and calculated dose factors for a sub-set of the marine organisms listed in column 1 of Table 1 (small and large, pelagic and benthic crustaceans, benthic molluscs, and pelagic and benthic fish) have been published [Pentreath and Woodhead, 1988]. There has been a more limited application of dosimetry models in the terrestrial environment with dose factors being published for a plant, a mammal and a bird [Amiro, 1997]. There are two important questions that remain to be resolved in respect of the assessed radiation exposures:

- How is the known, relatively higher effectiveness of a given absorbed dose from  $\alpha$ -particle radiation (densely ionizing or high linear energy transfer (LET)) as compared with the same absorbed dose from  $\beta$ -particles and the electrons generated by  $\gamma$ -rays (sparsely ionizing or low LET) to be taken into account? For the radiation exposure of native wild organisms in contaminated environments a quantity, corresponding to the equivalent dose (= absorbed dose x radiation weighting factor,  $w_r$ ) in human radiation protection practice, is required. (Note that proposals have been made, but these need to gain acceptance in the wider scientific community).
- How are small doses/dose rates to small and/or short-lived organisms to be interpreted when there is a significant probability that the actual dose (rate) received by a proportion of the organisms theoretically exposed is, in fact, zero (see discussion in section 3.2.2. relating to  $\alpha$ -radiation)?

It has been concluded that these existing models form a substantial basis for further development and application to native wild organisms in contaminated environments. A conceptual outline of a transparent framework for making this development has been given.

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