NRPB-R300

NRPB Models for Calculating the Transfer of Radionuclides through the Environment: Verification and Validation

J R Simmonds (Editor)

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National Radiological Protection Board

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Ionising radiation quantities and units

Quantity	SI unit	In other SI units	Old unit	Conversion factor
Exposure	—	$C\ kg^{-1}$	röntgen (R)	$1\ C\ kg^{-1} \sim 3876$ R
Absorbed dose	gray (Gy)	$J\ kg^{-1}$	rad (rad)	1 Gy = 100 rad
Dose equivalent	sievert (Sv)	$J\ kg^{-1}$	rem (rem)	1 Sv = 100 rem
Activity	becquerel (Bq)	s^{-1}	curie (Ci)	$1\ Bq \sim 2.7\ 10^{-11}$ Ci

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Abstract

There is a wide range of models available at NRPB to predict the transfer of radionuclides through the environment. Such models form an essential part of assessments of the radiological impact of releases of radionuclides into the environment. These models cover: the atmosphere; the aquatic environment; the geosphere; the terrestrial environment including foodchains. It is important that the models used for radiological impact assessments are robust, reliable and suitable for the assessment being undertaken. During model development it is, therefore, important that the model is both verified and validated. Verification of a model involves ensuring that it has been implemented correctly, while validation consists of demonstrating that the model is an adequate representation of the real environment.

The extent to which a model can be verified depends on its complexity and whether similar models exist. For relatively simple models verification is straightforward, but for more complex models verification has to form part of the development, coding and testing of the model within quality assurance procedures. Validation of models should ideally consist of comparisons between the results of the models and experimental or environmental measurement data that were not used to develop the model. This is more straightforward for some models than for others depending on the quantity and type of data available. Validation becomes increasingly difficult for models which are intended to predict environmental transfer at long times or at great distances. It is, therefore, necessary to adopt qualitative validation techniques to ensure that the model is an adequate representation of the real environment.

This report summarises the models used at NRPB to predict the transfer of radionuclides through the environment as part of a radiological impact assessment. It outlines the work carried out to verify and validate the models. The majority of these models are not currently available to users outside NRPB. However, computer packages have been developed which incorporate some of these models and which are generally available. These computer packages are also briefly described in this report.

Contents

1 Introduction

Assessing the radiological impact of releases of radionuclides to the environment is carried out through a series of stages. Firstly, the effluent released is defined and characterised, eg by specifying the radionuclides involved and the quantity of each released into the environment. Secondly, the transfer of radionuclides through the environment has to be determined. Thirdly, the resulting environmental concentrations are related to the different ways that people can be exposed to radiation; this may be through external irradiation or through radionuclides being taken into the body by inhalation and ingestion. Finally, the radiation exposures have to be estimated together with the associated risks, if required. In some cases the first two stages can be omitted as the assessment is based on measurements of radionuclides in the environment. However, in a large number of cases measurements are not available and models are required to estimate the transfer of radionuclides through the environment. This may be because sufficient measurements are not available, because the actual values are below the analytical limits of detection, because radiation doses are required over wide areas or because the assessment is concerned with radiation doses in the future.

At NRPB there is a wide range of models available to estimate the transfer of radionuclides in the environment. These models cover: the atmosphere; the aquatic environment, freshwater and marine environment; the geosphere; the terrestrial environment including foodchains. The global circulation of radionuclides is also modelled when necessary. These models are used to determine the radiological impact of both routine and unplanned, accidental releases of radionuclides into the environment, together with the release into the environment of radionuclides disposed of as solid radioactive waste. It is extremely important that the models used for radiological impact assessments are robust, reliable and suitable for the assessment being undertaken. Important steps in the development and use of these models are, therefore, their verification and validation. Verification of a model involves ensuring that it has been implemented correctly, while validation consists of demonstrating that the model is an adequate representation of the real environment.

A number of different verification procedures may be adopted depending on the complexity of the model and the way in which it is implemented. This includes ensuring that any mathematical operations are carried out accurately and, where the models are implemented on a computer, that the computer coding is correct. For all models developed at NRPB appropriate hand calculations are used to check the model implementation wherever possible. In addition, formal quality assurance procedures have been established as part of the verification procedure. These include: the review of model structure and basic equations by staff other than those involved in developing the model; checking computer codes to ensure that programming is correct; comparing computed results with problem solutions obtained from other models. In addition, NRPB takes part in national and international model comparison exercises and discussions with other experts in the field.

Ideally, validation is carried out by comparing model results with sets of field observations and experimental measurements other than those which were used to develop the model. In practice, full validation of any of the models in this way is not possible as independent datasets do not exist for all the radionuclides, times and environmental conditions for which the model is likely to be applied. However, where possible the environmental transfer models or parts of them are partially validated in this way. In addition, it is possible to check in a more qualitative way that the models are adequate through the use of data on chemically similar radionuclides or stable elements, the use of data from laboratory experiments and through external peer review of the model assumptions. Again national and international model comparison exercises can be used as part of this validation process. At NRPB a

number of different validation techniques are used depending on the type of model and the available data. The decision as to whether the model is an adequate representation of the real environment is necessarily a subjective one. It depends on the intended use of the model and the degree of accuracy deemed acceptable.

However well a model is verified and validated, uncertainties inevitably remain both in the model itself and in the parameter values used in the model[1]. The quantification of such uncertainties is an important part of applying such models and work has been carried out at NRPB on the quantification of uncertainties (see, for example, Jones *et al*[2] and Mobbs *et al*[3]).

This report summarises the models used at NRPB to estimate the transfer of radionuclides through the environment as part of a radiological impact assessment. It outlines work carried out to verify and validate the models in addition to the general procedures specified above. The majority of these models are not currently available to users outside NRPB. However, NRPB, often in collaboration with other organisations, has developed computer packages which incorporate some of these models and which are generally available. These computer packages are also briefly described in this report. As stated above an important part of model verification and validation is taking part in model intercomparison exercises; Appendix A outlines some of the international exercises in which NRPB has participated.

Models for the transfer of radionuclides through the environment are only part of the process of assessing the radiological impact of releases of radionuclides to the environment. For example, information is also required on the habits of the population of interest, such as the amounts of particular foods eaten and the time people spend at particular locations. A report has been prepared detailing the data of this type used at NRPB[4]. Also an important source of exposure is external irradiation from radionuclides in the environment. A number of models exist at NRPB for estimating such exposure; these models are outside the scope of this report but will be discussed in a separate report.

2 Atmospheric dispersion and deposition

2.1 Models

Most of the models for predicting rates and patterns of atmospheric dispersion used at NRPB are based on those recommended by the UK Atmospheric Dispersion Modelling Working Group (ADMWG), and the Atmospheric Dispersion Modelling Liaison Committee[5] (ADMLC); ADMWG consisted of scientists from universities, the Meteorological Office, the nuclear industry, consultancies and NRPB. It has recommended models for a wide range of situations. Only a few of these models have been incorporated into computer programs at NRPB.

ADMWG originally recommended a model[6] for dispersion from an isolated stack for short and continuous releases and distances up to a few tens of kilometres. Subsequent models gave extensions for deposition[7], and to longer range for continuous releases[8]. ADMWG also gave a model for long-range dispersion for short releases[9], for use in a restricted set of conditions. A further report[10] gave models for dispersion for releases from buildings, for coastal situations, and for the rise of buoyant material. All these models are based on the so-called Gaussian plume model, which is widely used with various sets of parameter values.

A model for dispersion from continuous releases based on the ADMWG recommendations is incorporated into the ESCLOUD program[11] which is used as the first step in calculating doses from

routine releases, as described in Section 7.1. ESCLOUD calculates activity concentrations in air and deposition rates, using the models given by Clarke[6] and Jones[7].

Models for dispersion following short releases are included in the accident consequence assessment programs CONDOR[12] and COSYMA[13], described in Section 7.2. The model in CONDOR[12] is based on that given by Clarke[6] extended to include the effects of deposition, buildings, plume rise and changes of atmospheric conditions during the plume's travel. COSYMA[13] includes several different dispersion models appropriate for different distances and based on different assumptions[14]. Those for short distances are similar to the ADMWG models as they are also based on the Gaussian plume model. However, they use different sets of values for the variation of plume size with distance from the source. These models are based on a series of experiments undertaken at Karlsruhe in Germany and Mol in Belgium[14]. COSYMA also includes the Imperial College code MESOS[15]. This model, which calculates dispersion over long distances, is not based on the Gaussian plume model but calculates trajectories using information on atmospheric conditions over a wide area.

NRPB also possesses the ADMS code[16], developed by Cambridge Environmental Research Consultants (CERC), but pending further testing, has not yet used it to any great extent.

2.2 Verification and validation

As noted above, most of the models used at NRPB are based on the Gaussian plume model of dispersion. A considerable amount of work has been undertaken outside NRPB to validate the different forms of the Gaussian plume model. ADMWG has produced a report[17] summarising the validation work for Gaussian plume models in general. This report suggests that the maximum concentration from a release of about one hour's duration can generally be predicted to within a factor of two or three. The report also concludes that annual average concentrations within a few kilometres of the site can generally be predicted within a factor of two.

The work done elsewhere to validate the models suggested by ADMWG is summarised by Jones[17] and in the ADMWG reports describing the models. This covers in particular the simple model for short releases described by Clarke[6] and the model for building wake releases described by Jones[10]. The models for plume rise recommended by ADMWG are those derived by Briggs[18] and Moore[19]. Both these models are based on extensive observations of rising plumes, as described in the original reports.

Kretzschmar *et al*[20] have carried out a series of releases of sulphur hexafluoride over an agricultural area near Mol in Belgium. The observed values of the parameter representing plume size, σ_y and σ_z, were compared with the predictions of a number of dispersion models including the one recommended by ADMWG[6] and the Mol model which is included in COSYMA[13]. A comparison between predicted and observed concentrations was only carried out for the Mol model, for which the ratio of predicted to measured concentrations was 1.02 ± 0.65.

NRPB has undertaken a detailed evaluation of the risks of leukaemia and other cancers in Seascale[21]. As part of this work, the annual average activity concentrations in air and amounts of material deposited each year in Seascale from all sources were calculated using the ESCLOUD model, and compared with observed values. The predicted and observed values are generally within a factor of two, with the predicted value tending to be higher than the observed value. The greatest difference observed is a factor of around five[21].

3 Terrestrial foodchains

Following the release of radioactive material to the atmosphere and its subsequent deposition on to the ground, one of the principal routes of exposure is internal irradiation from the ingestion of contaminated food. NRPB has developed models to simulate the transfer of radionuclides through terrestrial foods to be used in assessments of the radiological significance of accidental and routine releases of radioactive material to the terrestrial environment[22–26].

3.1 FARMLAND

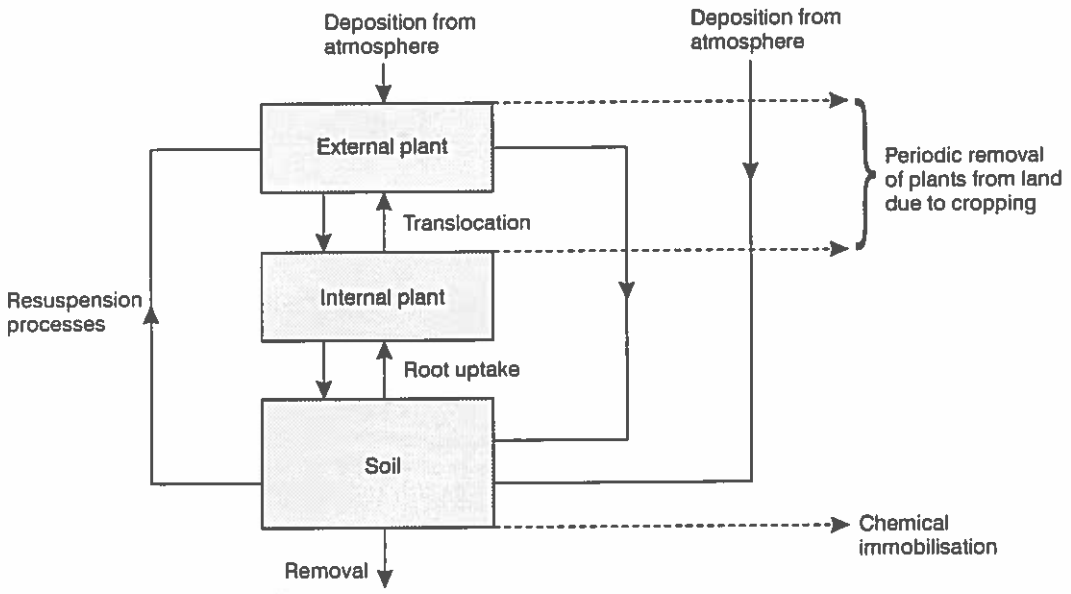
The main foodchain model is named FARMLAND (Food Activity from Radionuclide Movement on LAND) and contains a suite of submodels, each of which simulates radionuclide transfer through a different part of the foodchain. These submodels can be combined in various orders so that they can be used for different situations of radiological interest. The foods considered are: green vegetables; grain products; root vegetables and potatoes; fruit; milk, meat and offal from cattle; meat and offal from sheep. A large variety of elements can be considered, although the degree of complexity with which some are modelled is greater than that for others; isotopes of caesium, strontium and iodine are treated in greatest detail. FARMLAND is a general dynamic model with a compartmental structure. The main features of the submodels developed to describe the transfer of radionuclides to plants and to grazing animals are illustrated in Figure 1. It includes many time-dependent features; in particular, element-specific modules have been developed for animals to take into account the important biological and metabolic processes for those elements whose transfer to animal products is significant. For illustration, the compartment model structure used for the transfer of radioiodine to cattle is shown in Figure 2.

FARMLAND contains a submodel for the transfer of radionuclides through undisturbed soil, of which permanent grassland is an example. This submodel is also used in other models for the transfer of radionuclides through the urban environment[27]. The movement of radionuclides through the soil column is represented by a series of transfers between compartments of varying depth; within each compartment the radionuclides are assumed to be uniformly mixed. Many parameters influence the rate of migration, particularly the nature of the element and its chemical form, soil composition, climate and rainfall. Further details of the soil submodel are given by Brown and Simmonds[22].

FARMLAND can be used for a number of applications. It is primarily used to study the transfer of radionuclides into the foodchain following accidental or routine releases of radionuclides to the atmosphere. The way in which the model is used and the assumptions made depend on the application and these are described elsewhere[22,23]. An equilibrium version of FARMLAND is used in other models of the transfer of radionuclides in the biosphere[28] (see Section 7.3). FARMLAND has also been used in the development of a default model for application in the European Union (EU) which is intended for applications where site-specific data are not available[22,29]. The model can also be used to estimate the activity concentrations of radionuclides in terrestrial foods when the input is through irrigation[22]. A PC-based package for routine and accident applications of FARMLAND is under development.

3.1.1 Verification and validation of FARMLAND

A number of verification and validation studies have been carried out on FARMLAND during its development and since its implementation. Full details and results of these studies are described elsewhere[30]. A brief summary of the performance of FARMLAND is given here. A list of the verification studies performed using FARMLAND is given in Table 1.



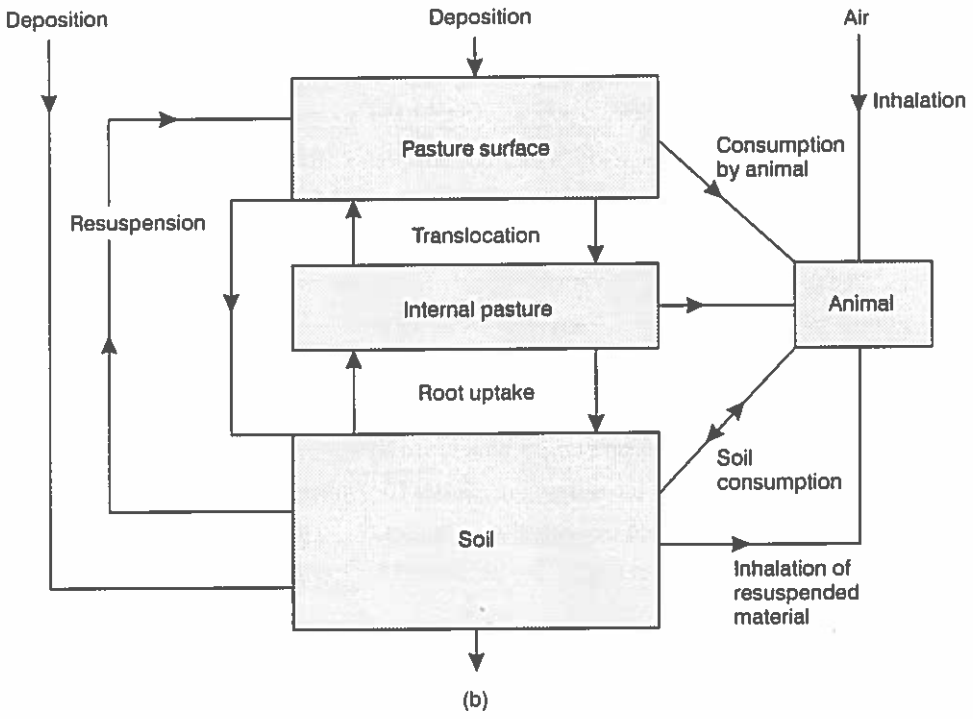
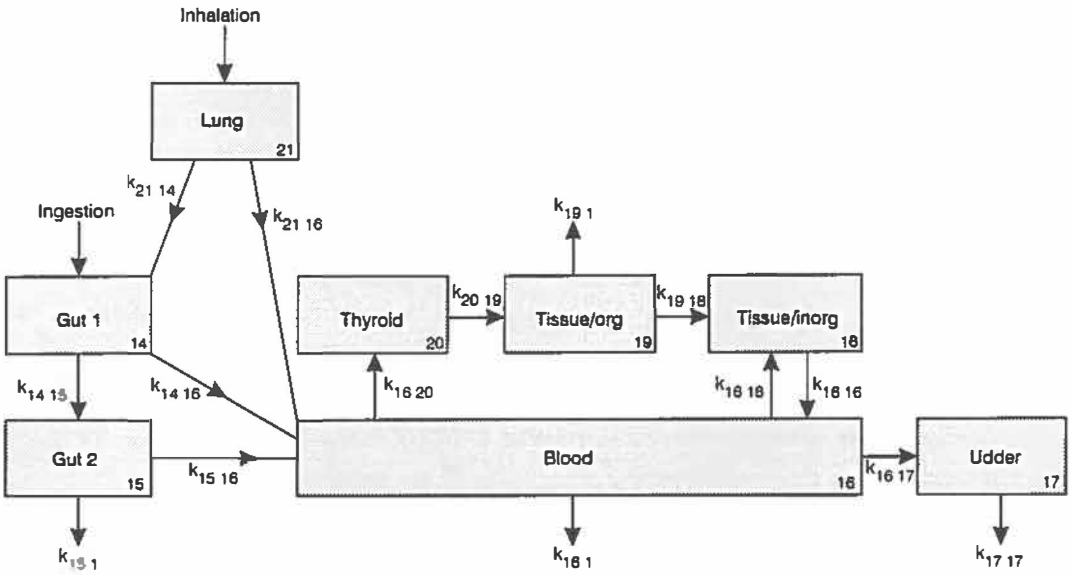


FIGURE 1 Schematic of the principal mechanisms for the transfer of radionuclides: (a) in plants and (b) in grazing animals



Notes
(a) $k_{14\ 16}$ represents the early absorption of iodine from the rumen of the cow.
(b) The storage of iodine in the soft tissues is represented by two compartments (18 and 19). The organic iodine produced in the thyroid is re-distributed throughout the soft tissues and organs of the body where it remains for some time before being broken down into inorganic iodine.
(c) $k_{19\ 1}$ represents the excretion of the organic fraction of iodine in the circulating fluids.
(d) Periodic slaughter is represented by losses from all compartments, the value of the rate constant is $4.56\ 10^{-4}\ d^{-1}$.

FIGURE 2 Compartmental model structure for iodine transfer in dairy cows

FARMLAND has been used in four extensive model intercomparison studies at various stages of its development[30].

In one of the intercomparisons, carried out under the BIOMOVS programme (see Appendix A), a scenario (B1) was designed to compare the predictions of a number of foodchain models given an average long-term concentration of iodine-131 and caesium-137 in air and known precipitation. The predictions of 16 foodchain models were compared for soil, vegetation, meat and milk. In addition, predictions of wet and dry deposition of these radionuclides were compared. The predicted values of caesium-137 in soil, pasture, milk and meat made by the models are illustrated in Table 2 where the values predicted by FARMLAND can be seen to be similar to those of most of the other models. The analysis of the comparison concluded that to a certain extent the variability of the results across models was due to the different purposes for which the models had been developed. These differences could be eliminated by normalising the results to the total deposition, and the mass interception factor was the single parameter that accounted for the major part of the variation across the models.

In the model intercomparison studies in which FARMLAND has been used, the activity concentrations in food predicted by the models were generally in reasonable agreement and, where applicable, the same pattern of time dependence was seen. Agreement was generally closest for strontium, caesium and iodine, elements which have been extensively studied and differences due to the structure of the models were usually smaller than those due to agricultural practices and the choice of parameter values. In the studies carried out FARMLAND has compared favourably with other major foodchain models in Europe and elsewhere[30].

TABLE 1 Verification and validation studies carried out with FARMLAND*

Study	Year
Verification studies	
Comparison of three UK foodchain models	1983
Comparison of FARMLAND with German model ECOSYS	1985
Comparison of FARMLAND with German model ECOSYS	1988–1989
BIOMOVS international model intercomparison: scenario B1 for continuous deposition to agricultural land	1988–1990
Validation studies – testing model predictions against:	
UK fallout from weapons testing	1980 and 1987
Field data from Cumbria, UK, for pasture-cow-milk pathway	1983
UK monitoring data from the Chernobyl accident for milk, green vegetables and lamb	1987–1989
Field data from the EU from the Chernobyl accident for pasture-cow-milk pathway	1987–1989
Field data from Cumbria, UK, from the Chernobyl accident for winter feeding of cattle	1987–1989
Feeding trials on cattle and sheep	1987–1989
Data from the Chernobyl accident for milk, beef and grain at locations in the northern hemisphere (BIOMOVS – Scenario A4)	1987–1990
Data from the Chernobyl accident from southern Finland for a number of foods and whole body burdens (VAMP scenario S)	1992–1994

*For more details, see Brown[30] and IAEA[31].

TABLE 2 Model predictions for caesium-137 in soil, pasture, milk and meat, for BIOMOVS B1 scenario[30]

Model	Soil (Bq kg^{-1} dry weight)	Pasture (Bq kg^{-1} dry weight)	Milk (Bq l^{-1})	Meat (Bq kg^{-1})
ABG	$2.3\ 10^4$	$1.9\ 10^4$	$3.0\ 10^3$	$7.6\ 10^3$
AIRDOS	$7.9\ 10^4$	$7.9\ 10^4$	$4.9\ 10^3$	$6.9\ 10^3$
BIOPATH	$7.5\ 10^3$	$3.3\ 10^4$	$3.9\ 10^3$	$2.0\ 10^4$
CHERPAC	$3.6\ 10^5$	$1.8\ 10^5$	$1.3\ 10^4$	$4.6\ 10^4$
CRRIS	$2.3\ 10^5$	$1.9\ 10^5$	$1.4\ 10^4$	$1.9\ 10^4$
DOSDIM	$2.5\ 10^4$	$2.5\ 10^4$	$2.6\ 10^3$	$1.1\ 10^4$
ENEA	$2.5\ 10^4$	$2.2\ 10^4$	$2.9\ 10^3$	$1.2\ 10^4$
FARMLAND	$\mathbf{1.0\ 10^5}$	$\mathbf{4.1\ 10^4}$	$\mathbf{3.5\ 10^3}$	$\mathbf{1.3\ 10^4}$
IAEA	$6.0\ 10^4$	$6.3\ 10^4$	$8.1\ 10^3$	$1.5\ 10^4$
NCRP	$3.5\ 10^4$	$3.6\ 10^4$	$5.8\ 10^3$	$1.3\ 10^4$
NRC	$1.1\ 10^4$	$1.1\ 10^4$	$5.5\ 10^2$	$2.2\ 10^2$
PATHWAY	$1.8\ 10^4$	$4.3\ 10^3$	$2.0\ 10^2$	$1.0\ 10^3$
RIST	–	$6.4\ 10^4$	$2.8\ 10^3$	$1.4\ 10^4$
SIRATEC	$1.4\ 10^4$	$3.0\ 10^4$	$3.0\ 10^3$	$7.6\ 10^3$
STOCHASTIC	–	$5.2\ 10^4$	$3.8\ 10^3$	$9.1\ 10^3$
TERFOC	$3.9\ 10^4$	$4.3\ 10^4$	$5.1\ 10^3$	$1.5\ 10^4$
Geometric mean		$3.8\ 10^4$	$3.3\ 10^3$	$8.6\ 10^3$
Geometric standard deviation		2.6	2.9	3.5

FARMLAND has been tested against several types of data and has been used in two international model validation studies, BIOMOVS[30] and VAMP; a description of the objectives and content of these studies is given in Appendix A. Data for validation are largely only available for the pasture-cow-milk pathway; limited data for green vegetables, grain, beef and sheep meat are available, mainly in the form of environmental monitoring data following the Chernobyl accident.

In one study, the predictions of FARMLAND were compared with measurements of caesium-137 in milk made over the winter period and into the following summer after the Chernobyl accident at two farms in the UK, one in Cumbria and the other in Berkshire. The farming practices at these two farms were very different and the winter feeding regimes were complex. The FARMLAND model predictions using default assumptions were compared with the measurements over this period (Figure 3). The time variation in measured concentrations in milk at the two farms can be seen to be different reflecting the different feeding regimes. The FARMLAND predictions generally reflected the trend in milk concentration but did not reproduce exactly the temporal changes in activity concentrations at either farm. This was expected as in its generic form the model makes general assumptions about husbandry in the UK which are not necessarily appropriate for specific farms. When the time-integrated concentrations in milk were compared for the winter period, the FARMLAND model overestimated the integrated concentration by a factor of two for the Berkshire farm and a factor of four for the Cumbrian farm. Most of these differences could be attributed to the differences in 'feed-to-milk' transfer factors seen over the winter period. This comparison shows that in its generic form FARMLAND adequately predicts the seasonal variation in activity concentrations in milk at example locations in the UK[30].

FARMLAND is expected to perform well when the model predictions are compared with measurements over a range of sites. This has been shown in one of the BIOMOVS scenarios, in which FARMLAND and 21 other models were tested against data collected after the Chernobyl accident

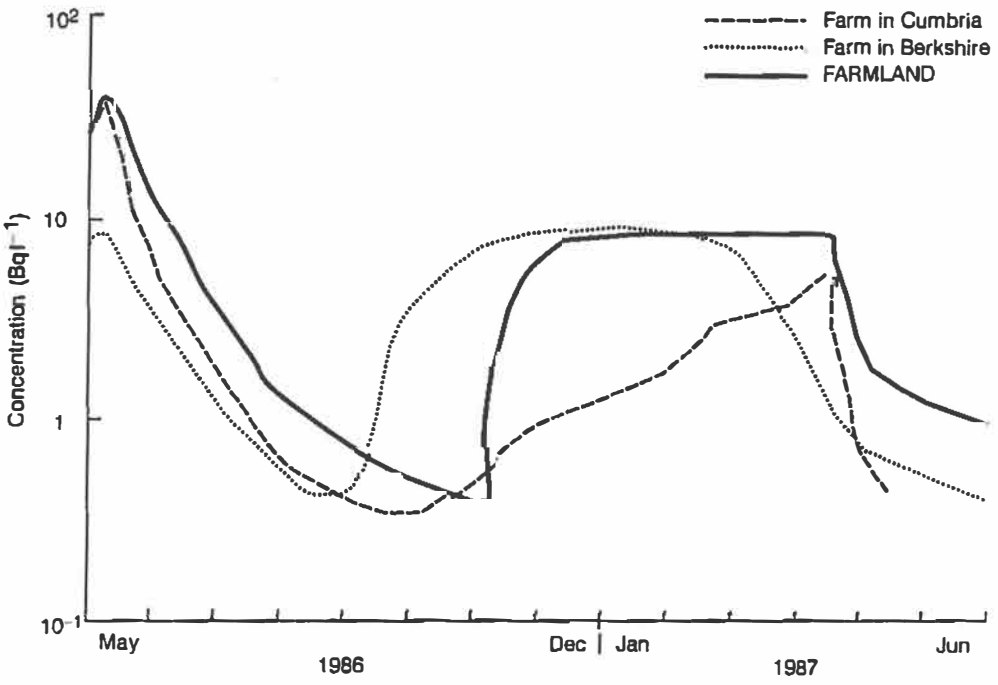


FIGURE 3 Concentration of caesium-137 in milk normalised to 100 Bq m^{-2} on grass

for a number of locations in the northern hemisphere, and also in other studies that have been carried out[30]. The comparison of FARMLAND predictions with measurement data, and particularly with post-Chernobyl measurements, has strengthened confidence in the validity of the model for use in general radiological assessments, which was the use for which it was intended.

FARMLAND is not, however, necessarily expected to perform well at any one specific site unless all the local conditions are taken into account. Where FARMLAND has been tested against site-specific data some discrepancies between measurements and model predictions have been found. These are often due to the difference between general assumptions made in the default FARMLAND model and the actual conditions. An example of this is seen from the participation of FARMLAND in a scenario of the Multiple Pathways Assessment Working Group of the IAEA VAMP programme, where predictions of activity concentrations in a range of foods for a site in southern Finland following the Chernobyl accident were compared with measured values in a blind test[31]. For some foods significant differences were seen between the predictions of FARMLAND and the measured values, particularly in the first year following deposition. In general, these differences could be explained by the application of the general model to a specific site with characteristics and farming practices very different to those assumed as default in FARMLAND.

3.2 TRIF

TRIF (Tritium TRansfer Into Food)[26] is a dynamic model of tritium movement through the foodchain which is designed to bridge the gap between applying a limited specific activity calculation and a very involved integrated dispersion and environmental transfer model. The most important practical developments are the inclusion of organically bound tritium (OBT) within a specific activity framework and the development of a semi-integrated dynamic model when the complexity of carrying out a more involved calculation cannot be justified.

TRIF is distinct from conventional representations of radionuclide movement through the environment in modelling the dispersion of tritiated water recycled from the ground assuming an area source. Some of the tritium depositing on the ground is assumed to be returned to the atmosphere directly from the ground or as part of the transpiration stream of plants. This material then disperses and recycles between the atmosphere and the ground. TRIF in its standard form treats this process explicitly while assuming that the dispersion of the original source plume can be treated separately. By default, TRIF uses long-term generic averages for water balance constraints but it is advisable to replace these with site-specific information if this is available. Site-specific information should also be used to assess the effective deposition left under different conditions by very short timescale processes (half-life of around 30 minutes) not included in TRIF[26,32].

3.2.1 Verification and validation of TRIF

Substantial amounts of the available experimental data have been used in deriving the transfer coefficients of the model, thus making an independent validation difficult. However, under conditions of dynamic equilibrium and neglecting the contribution of OBT, TRIF reproduces the results of a specific activity calculation[26]. Also in agreement with experimental data[33], TRIF predicts that grain will have a similar concentration of tritium as other crops but with 90% in the form of OBT.

The predictions of TRIF are also within a factor of two of experimental data for the concentrations in milk produced by a cow kept in an environmental enclosure and given tritiated water for 20 days[34]. This result also demonstrates the importance of the direct transfer from cow OBT to milk OBT which is included in TRIF[26].

4 Radionuclide transfer in water bodies

4.1 Models

4.1.1 Freshwater and estuaries

The freshwater environment is very diverse, with the values of major parameters, such as volumetric flow, varying over several orders of magnitude from one site to another and from one time to another. NRPB has adopted simple river and lake models in preference to more complex models which require large quantities of site-specific data, which are generally not available. The models are of the compartmental type, where each compartment represents a homogeneous freshwater body within which radionuclides are instantaneously well mixed[35]. The basic features of the models are generic but the models can be applied to specific rivers and lakes by appropriate choice of the number and volumes of compartments, and other parameters (see, for example, Cooper *et al*[36]). The structure of the river model is based on that devised by Schaeffer[37] following measurements of radionuclide distributions in the Rhône. Radionuclides in the water in each river compartment are assumed to be partitioned between a dissolved component and a component sorbed on to suspended sediment, the partitioning being determined by the freshwater sediment distribution coefficient and the suspended sediment load. The exchange of radionuclides between the river water and the bed sediment is represented by a depletion parameter which removes radionuclides from the water as a function of distance travelled downstream. The radionuclides in the river bed are also transported downstream, but at a much lower velocity than the river water.

In the lake model, a lake is represented by a single water compartment and three sediment compartments; the sediment compartments represent the top 10 cm, the underlying 1.9 m and the deep sediment of the lake bed. Radionuclides in the lake water are partitioned in the same way as in river water. The radionuclides adsorbed on to suspended sediment are transferred to the top 10 cm of the lake bed according to the sedimentation rate. Radionuclides in the upper lake bed may be remobilised by bioturbation and diffusion back to the water or they may be transported to deeper sediment by diffusion and burial. Element-dependent concentration factors are used to obtain concentrations in freshwater fish from concentrations in filtered river or lake water. Radionuclides may also be transferred to agricultural land from river or lake water or bed sediment by irrigation, dredging, flooding or changes in the size or course of the freshwater body.

At present, NRPB modelling of estuaries is normally limited to the desorption of selected elements from river-bed sediment as this sediment enters the marine environment. A multi-compartmental model does exist for one British estuary, the Severn[35,38] (see Figure 4). The estuary has been divided into four compartments and water exchanges have been derived from a model by Uncles and Radford[39]. The modelling of sediment movement takes into account observations of large quantities of bottom sediments which are remobilised by spring tides (see, for example, Kirby and Parker[40]).

4.1.2 Coastal seas

The marine dispersion model currently used at NRPB to assess the radiological consequences of radioactive discharges into the marine environment is derived from the compartmental model developed for the MARINA project of the European Commission (EC)[41].

The model, called DORIS (Dispersion Of Radionuclides In the Sea), describes the significant movements of radionuclides in European coastal waters[35]. In the model, the different areas of the European marine system are represented by compartments and the movement of radionuclides between compartments is modelled using transfer rates. The main difference between the present model and the

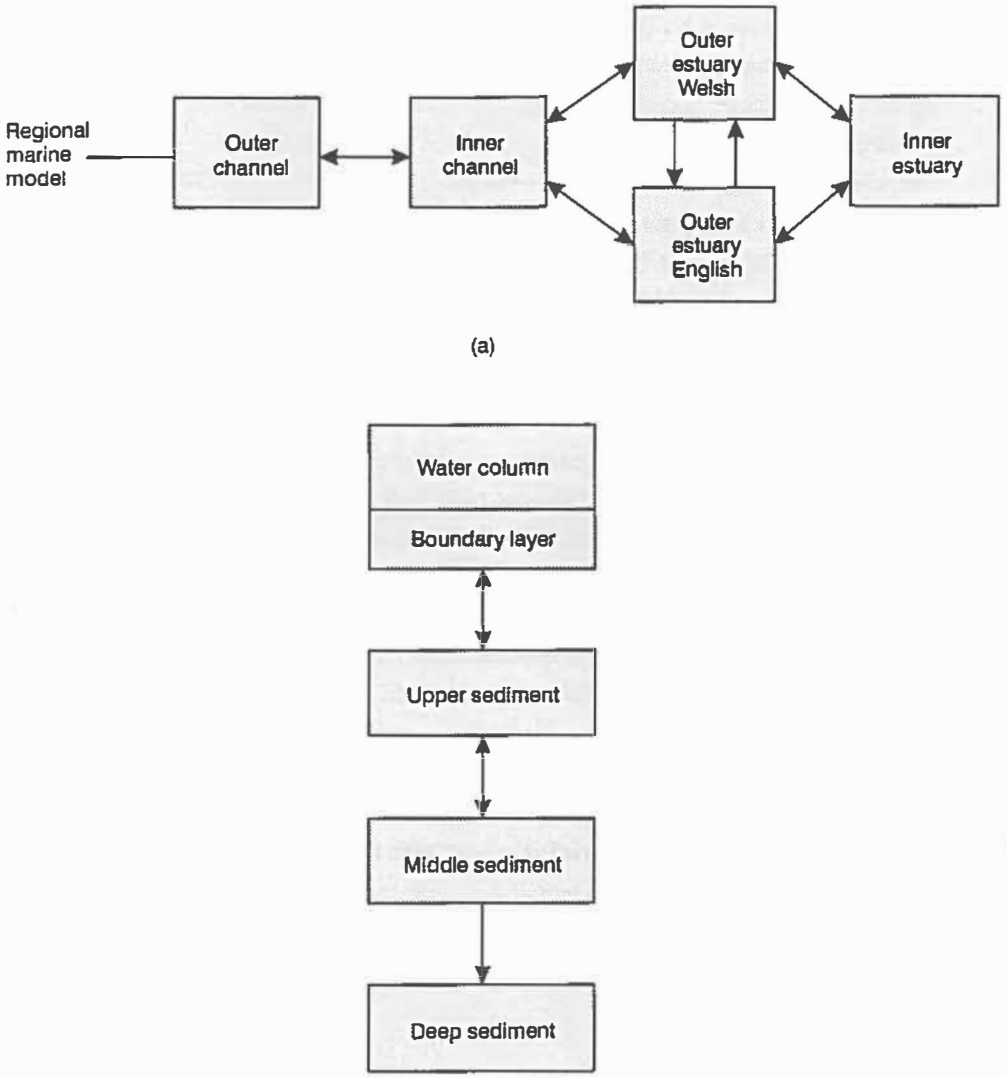


FIGURE 4 Severn estuary model: (a) water compartments and (b) sediment model for each compartment

MARINA project model lies in the more complete description of water movements in the North Sea, the English Channel, the Irish Sea, the waters around the north and west coasts of Scotland, the Bay of Biscay and the Atlantic coastal waters of Spain and Portugal.

The regional marine model is interfaced with a local compartment model, usually represented by a single compartment, into which radioactive liquid effluents are assumed to be released. The local compartment describes the local environmental conditions, which may be very important in determining the impact of any radioactive release[35].

The transport processes considered in the model are advection, diffusion and interaction with sediments. The adsorption of activity by sediments is due to both partitioning of the activity between the liquid phase and the solid phase (suspended sediments) and the removal of activity from the water column to bottom sediments. Both processes are modelled using element-dependent distribution coefficients (k_d). The removal of activity from the water column is described by a particle scavenging model. Each sea compartment has associated a seabed compartment divided into two layers. The upper

layer is 0.1 m thick and the underlying layer is 1.9 m thick. The removal of radionuclides is determined by distribution coefficients and by the rate of settling of the particulate matter. Movement between the two layers, burial, and the return of radionuclides to the water column through bioturbation and diffusion are also taken into account[35].

The model predicts radionuclide concentrations in water (filtered and unfiltered), in suspended sediments and in seabed sediments. Concentrations of radionuclides in seafood are calculated from activity concentrations in filtered seawater using equilibrium concentration factors. Activity concentrations in seaspray are, generally, calculated using model predictions in unfiltered seawater and an empirical equation developed by UKAEA Harwell[42]. Activity concentrations in sand are related to the concentrations in the top layer of the seabed. The predicted radionuclide concentrations in environmental media are then used to calculate the intakes of these radionuclides by man through ingestion or inhalation, while activity concentrations in sediments are generally used to calculate intakes through inhalation of resuspended material or dose rates for external exposure.

A detailed description of DORIS is given by Simmonds *et al*[35].

4.1.3 Deep oceans

A total of four deep ocean dispersion models are available at NRPB. These are the numerical models COMMA and MINIBOX and the analytical models GESAMP6 and GESAMP7. COMMA is a compartmental model of the world's oceans with a total of 91 water compartments and 69 sediment compartments. The compartments are defined on the basis of bottom topography and density, using data from the GEOSECS survey[43], with greater resolution in the Atlantic Ocean. The general structure of the model is shown in Figure 5. The exchanges between the compartments were derived from a review of water current measurements[44]. Scavenging of radionuclides by particles in the water column

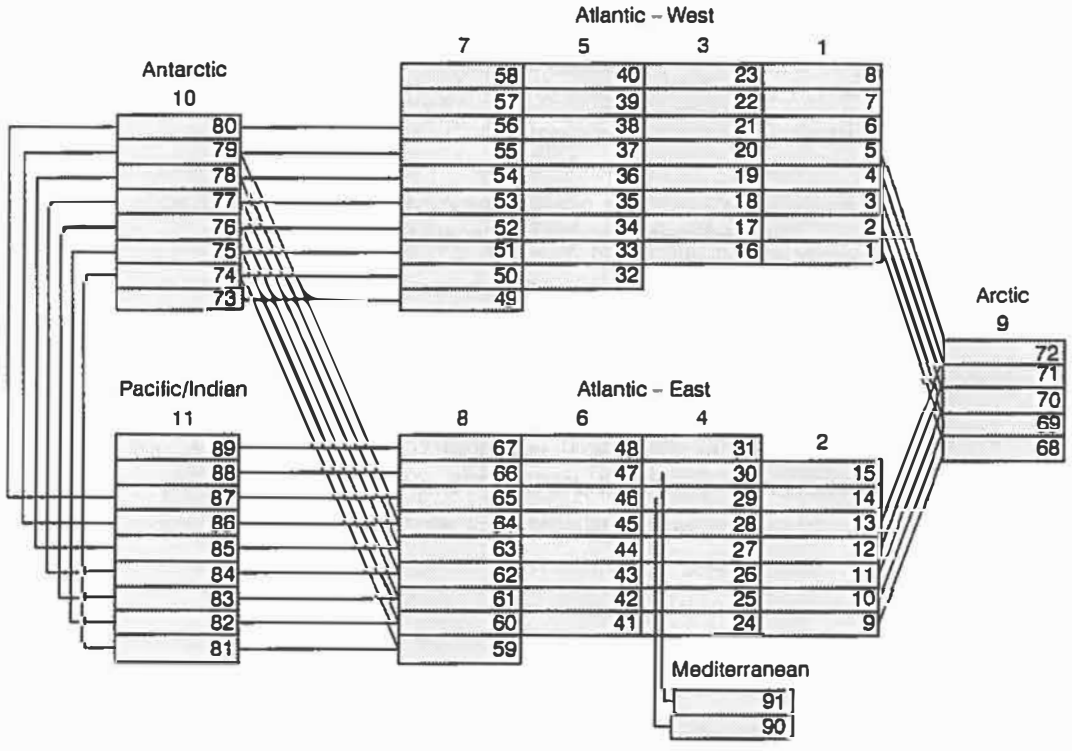


FIGURE 5 Structure of the ocean model, COMMA (the numbers identify the various compartments into which the oceans are subdivided)

is represented by two particle types falling at different velocities. Dissolution of the carbonate fraction of the particles as they fall between the carbonate saturation depth and the carbonate compensation depth is included. At the ocean floor resuspension of particles, pore water diffusion, bioturbation and burial are modelled. The model also includes a set of nested compartments around the source and these enable radionuclide concentrations in the 'near field' to be predicted more accurately. The model is described in detail elsewhere[44–48] and was developed in close collaboration with the Fisheries Directorate of MAFF (now CEFAS, the Centre for Environment, Fisheries and Aquaculture Science). Revisions to the database in the late 1980s have been incorporated in a separate version, COMMA2.

MINIBOX[49] is a compartmental model derived from COMMA with a total of 31 water compartments. It was derived by reducing the horizontal and vertical resolution and by removing the diffusive layer in the ocean floor. A revised version, MINIBOX2, corresponds to a reduction of the model COMMA2. MINIBOX was developed for use in uncertainty analysis and therefore it is possible to specify a series of input parameter values for the model. Parameters that can be varied are the oceanic diffusivity, the scavenging rate (small particle settling rate, sediment concentration, sediment k_d value and soluble fraction) and the depth of the mixed layer in the bed sediment. The flow pattern can also be scaled up or down.

GESAMP6 and GESAMP7 are implementations of the models described in a report by IAEA[50] which were developed for and are recommended by the United Nations Group of Experts on the Scientific Aspects of Marine Pollution (GESAMP). GESAMP6 is the analytical steady-state solution of the one-dimensional ocean model described in Appendix VI of the report. It is suitable for studies of dispersion from a continuous source. This model has also been used in uncertainty analyses and has the same variable parameters as MINIBOX. GESAMP7 is the analytical steady-state solution of the three-dimensional ocean model described in Appendix VII of the report. It models diffusion in the ocean, scavenging by one particle type, and bioturbation, pore water diffusion and burial at the sediment–water interface. This model is suitable for studies of concentrations close to a source. In the two numerical models the equations are solved using the computer code FACSIMILE[51].

4.2 Verification and validation

4.2.1 Freshwater and estuary models

The river model is based on measurements on the Rhône conducted by Schaeffer[37]. The model uses Schaeffer's empirically derived values for the depletion parameter used to represent the interaction between the water and the river-bed sediment. In one sense, therefore, the model is valid for the Rhône. The model has been applied to other rivers by varying the parameter values, eg Molse Nete, Belgium[52]. Measurements of activity concentration were available for points downstream of the discharge outfall. The ratio of predicted values to those observed was generally within a factor of five; results are presented for caesium-137 and americium-241 in Tables 3 and 4, respectively. The methodology report for routine releases[35] suggested the addition of a riverbank compartment to the model to account for natural deposition and dredging. These processes may give rise to enhanced activity concentrations in bank sediments close to a discharge outfall. Recent validation tests using data for the Molse Nete river have shown that there are few advantages in the addition of the riverbank compartment. The river model is included in the PC-CREAM[53] suite of codes and further validation using data for UK rivers has been proposed.

The lake model has undergone verification, by comparison with models developed independently by other organisations, within the BIOMOVS international project[54]. Two test cases involving lakes have been defined[55]: one involves a constant source of radionuclides into a

TABLE 3 Comparison of observed and predicted activity concentrations of caesium-137 in the bed sediment of the river Molse Nete

	Activity concentration (Bq kg^{-1})					
	0.9 km downstream			3.7 km downstream		
Year	Observed	Predicted	Ratio of predicted to observed	Observed	Predicted	Ratio of predicted to observed
1978	429	1430	3.33	1709	4284	2.51
1979	470	1242	2.64	4588	3666	0.80
1980	725	1072	1.47	2882	3167	1.09
1983	163	1554	9.53	544	3479	6.40

TABLE 4 Comparison of observed and predicted activity concentrations of americium-241 in the bed sediment of the river Molse Nete

	Activity concentration (Bq kg^{-1})					
	0.9 km downstream			3.7 km downstream		
Year	Observed	Predicted	Ratio of predicted to observed	Observed	Predicted	Ratio of predicted to observed
1978	270.1	109.8	0.41	329.3	250.3	0.76
1979	207.2	72.6	0.35	2190.4	215.3	0.099
1980	208.68	47.7	0.23	876.9	172.8	0.19
1983	15.54	16.72	1.08	98.42	71.7	0.73

lake with constant characteristics, while the second involves a constant source into a lake with time-varying characteristics. In the first case the comparison between the codes was based on calculated concentrations in water, sediment and fish. In the second case the comparison was based on calculated concentrations in water, fish, sediment and soils generated as the lake silts up. The results of this comparison exercise were rather inconclusive as there were substantial differences between the models and data used by different participants.

The compartmental model of the Severn estuary has been used to simulate the salinity profile along the estuary; reasonable agreement with observed salinities was obtained. The model has also been used to simulate the dispersion of caesium-137 discharged from Berkeley, Hinkley Point and Oldbury nuclear power stations; again, reasonable agreement with observed concentrations was obtained[38] (see Figure 6).

4.2.2 Coastal sea models

A calibration of the marine dispersion models developed by Grimwood[56] and Evans[57], on which the current NRPB marine model is based, was carried out by Hallstadius *et al*[58]. Activity concentrations of caesium-137 in seawater of the North Sea, Skagerrak, Kattegat and Baltic Sea predicted by the models using historical discharges from Sellafield, Cap de la Hague and Dounreay were compared with measurements taken in the same areas. Transfer rates between compartments were adjusted to give the best fit between model predictions and observations. The

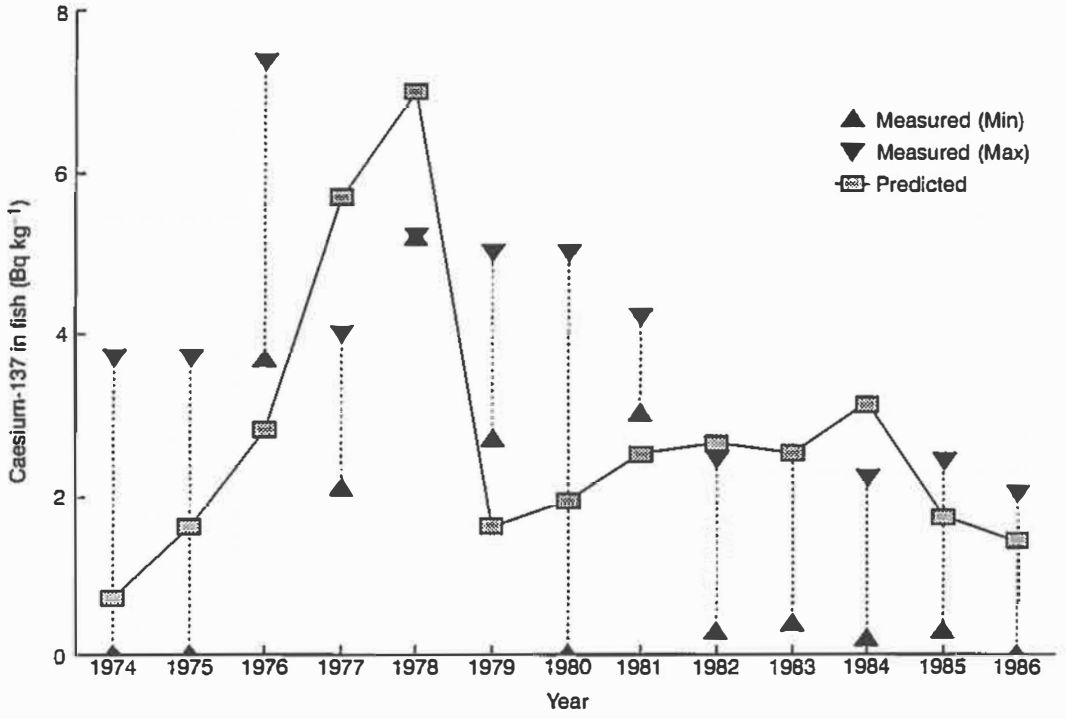


FIGURE 6 Predicted and measured concentrations of caesium-137 for the Severn estuary model

fluxes calculated in this calibration exercise were not adopted in the present marine model because the compartmental structure of the North Sea adopted in the model is different from that of the Grimwood model. However, fluxes between compartments of the North Sea, and between North Sea compartments and other areas, currently adopted are closer to those calculated by Hallstadius *et al*[58] than to those suggested by Grimwood[56].

The version of the coastal sea model known as MARIN1 was used in the EC MARINA project to determine the radiological impact on EC member states of routine discharges into north European waters[41]. As part of this study, the predictions of MARIN1 were compared with a range of environmental measurements for caesium-137, technetium-99 and plutonium-239+240. The measurements considered were concentrations in filtered seawater, fish and fucoid seaweed. Figure 7 shows the measured and calculated concentrations of caesium-137 in filtered seawater in the eastern Irish Sea, while Table 5 gives a comparison of results for plutonium-239 in filtered seawater. This exercise showed that MARIN1 described reasonably accurately the dispersion of radionuclides in European shelf seas. The predicted concentrations of the three radionuclides considered were generally within an order of magnitude of their measured values, and usually much closer. The larger differences between measured and predicted concentrations tended to be at some distance from the source of the radionuclides[41].

A validation of the Irish Sea section of the model was carried as part of a study to assess the radiological implications of radioactive contamination of west Cumbria[59]. Activity concentrations of caesium-137, plutonium (alpha) and americium-241 in seawater and seaspray predicted by the model were compared with measurements taken by MAFF in the Irish Sea and by UKAEA at Eskmeals. The results of the comparison are given by Wilkins *et al*[59] and again generally acceptable agreement was obtained.

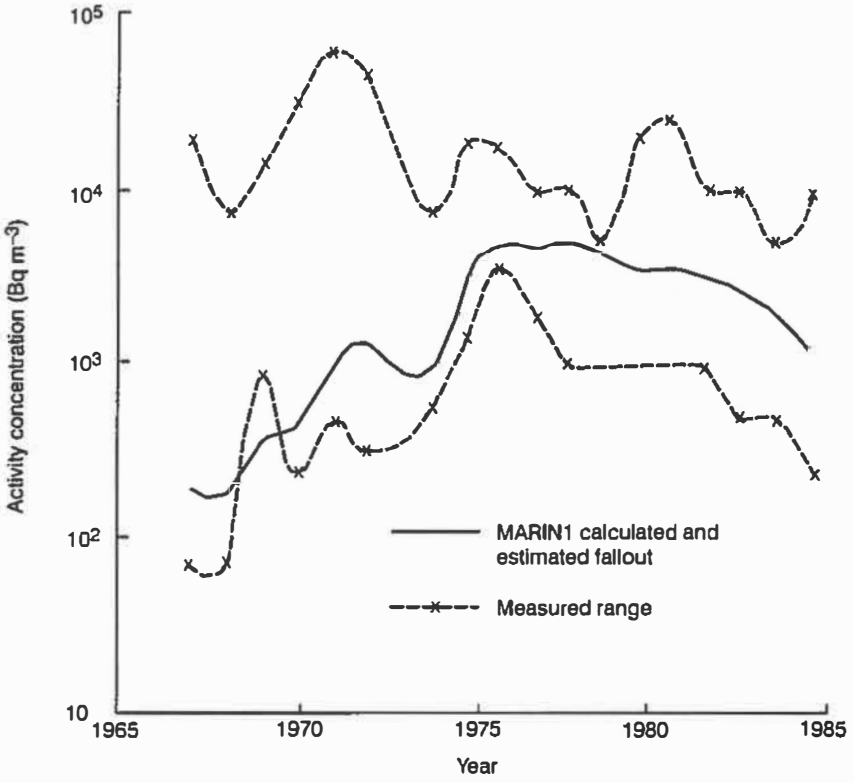


FIGURE 7 Measured and calculated filtered seawater concentrations of caesium-137 in the eastern Irish Sea

4.2.3 Deep ocean models

Verification of the ocean dispersion models has been carried out by model intercomparisons[60,61]. The most extensive intercomparison study[61] was that set up by NEA under the Coordinated Research and Environmental Surveillance Programme (CRESP) on sea disposal[62]. In this study the predictions of the model COMMA were compared with those of three other deep ocean models for two release periods and for five radionuclides. In general, allowing for differences in the ocean volumes assumed, all models gave similar results at long times. Differences, where they occurred, were understandable and were caused by differences in the databases and in the detail in which vertical water movement was modelled. COMMA agreed well with the other compartmental model in the intercomparison, MARINRAD[63]. GESAMP7 was also compared with the results in the nested compartments of COMMA and gave good agreement[61]. An intercomparison of COMMA with a compartmental model developed at CEA[64] was performed using the CRESP test cases and the two models agreed extremely well for poorly sorbed, long-lived radionuclides. The differences for other radionuclides were due to different vertical mixing rates and different exchange rates at the sediment–water interface.

At NRPB, MINIBOX has been compared with COMMA using the CRESP intercomparison cases[49] and the predictions of the two models agree well in the region close to the release point. Both models predict that the peak surface concentration occurs in the Antarctic. MINIBOX predicts higher concentrations in this compartment at times less than 1000 years, by less than a factor of ten, but the two models agree well at later times, for all the radionuclides considered except thorium-230; MINIBOX consistently predicts higher thorium-230 concentrations. MINIBOX2, COMMA2 and

TABLE 5 Measured and predicted activity concentrations of plutonium-239 in filtered seawater

	Activity concentrations (Bq m^{-3})							
	1980		1981		1982		1985	
Compartment	Measured	Predicted	Measured	Predicted	Measured	Predicted	Measured	Predicted
Scottish waters	$7.3\ 10^{-2} - 2.37\ 10^{-1}$	$3.2\ 10^{-1}$	–	–	–	–	$1.7\ 10^{-1}$	$2.8\ 10^{-1}$
North East Atlantic	$7.0\ 10^{-3}$	$1.6\ 10^{-3}$	–	–	$6.8\ 10^{-2} - 1.35\ 10^{-1}$	$2.1\ 10^{-4}$	–	–
English Channel East	$4.2\ 10^{-2}$	$4.0\ 10^{-2}$	–	–	$6.4\ 10^{-3}$	$3.3\ 10^{-2}$	–	–
North Sea South	$2.1\ 10^{-2} - 2.8\ 10^{-2}$	$5.7\ 10^{-2}$	$3.4\ 10^{-2} - 4.8\ 10^{-2}$	$6.0\ 10^{-2}$	$2.5\ 10^{-2} - 2.6\ 10^{-2}$	$5.8\ 10^{-2}$	$3.3\ 10^{-2}$	$5.5\ 10^{-2}$
North Sea Central	$9.0\ 10^{-3} - 8.9\ 10^{-2}$	$1.3\ 10^{-1}$	$3.9\ 10^{-2} - 8.1\ 10^{-2}$	$1.4\ 10^{-1}$	$9.1\ 10^{-3} - 1.05\ 10^{-1}$	$1.4\ 10^{-1}$	$3.0\ 10^{-2} - 8.4\ 10^{-2}$	$1.3\ 10^{-1}$
North Sea North	$7.0\ 10^{-3} - 8.9\ 10^{-2}$	$3.0\ 10^{-2}$	–	–	$1.2\ 10^{-2} - 7.9\ 10^{-2}$	$3.1\ 10^{-2}$	$7.7\ 10^{-3}$	$3.1\ 10^{-2}$

GESAMP6 have been compared for a number of simulated releases of radionuclides at the bottom of the sea and the results from MINIBOX2 and GESAMP6 were compared in an uncertainty analysis[49]. Peak individual doses from all three models, using the same exposure model, were in very good agreement[49]. GESAMP6 predicted that doses occurred earlier than did the two numerical models since it does not model the build-up in the ocean explicitly. Integrated collective doses from MINIBOX2 and COMMA2 were within 10%, whereas GESAMP6 gave lower values since it assumed a higher effective global sedimentation rate.

Owing to the difficulties in validating ocean models, such studies have generally taken two forms: a qualitative validation by expert review and a quantitative validation by comparison with measurements of tracers in the oceans. COMMA has been subjected to international review[44,50] and was considered suitable for studies of dispersion of radionuclides in the deep ocean. The model has also been used to predict the profiles of a few tracers in the ocean and the results have been compared with measurements[65]. Predictions of temperature and salinity were found to agree well with measured values. However, the model overestimates the concentrations of thorium-230 and the flux of radium-226 from the bottom sediment. This indicates that the model underestimates the scavenging rate and this may be due to the k_d value chosen, the particle settling rate assumed, or both. Predicted concentrations of thorium-232 and its progeny in water had the expected vertical profile but uncertainties in the input data made it difficult to comment on the predicted magnitudes. Concentrations of carbon-14 predicted for the cosmogenic component were within a factor of five of measured values.

5 Radionuclide migration in the geosphere

5.1 Models

The computer code GEOS[66] is designed for modelling the migration of radionuclides in groundwater in one dimension using a compartmental model of the geosphere. GEOS models radionuclide transport in saturated porous media using the advection-diffusion equation, which is solved for given boundary conditions. The code incorporates radionuclide-dependent retardation coefficients to account for linear equilibrium sorption of activity from solution in groundwater on to the solid medium. The code also incorporates simple leaching models to simulate the release of radionuclides from solid waste placed in a geological repository.

The differential equations defining the model are solved using FACSIMILE[51].

5.2 Verification and validation

Validation of the code over the timescales relevant for radionuclide migration in the geosphere is not feasible.

The GEOS code has been verified as part of the International Nuclide Transport Code Intercomparison Study (INTRACOIN)[67], using four of the seven level 1 test cases. Agreement between GEOS and the other participating codes was good. Any differences between the results from the various codes were usually explainable in terms of the specified boundary conditions.

In addition GEOS has been verified using the PSACOIN level E intercomparison exercise (PSAC user group)[68], for releases of iodine-129 and neptunium-237 into a two-layer geosphere. GEOS was designed for only one layer and therefore the results were compared with the analytical solution of the advection-diffusion equation at the boundary between the first and second layer. The agreement between GEOS and the analytical solution was very good for both radionuclides.

6 Global circulation of radionuclides

6.1 Models

Some radionuclides, owing to the magnitude of their half-lives and their behaviour in the environment, may become dispersed around the globe and act as a long-term source of exposure of both regional and world populations. The radionuclides that are important in this context are tritium, carbon-14, krypton-85 and iodine-129. Doses to individuals from the global circulation of these radionuclides are generally very small and the models are therefore used principally to determine collective doses.

Models are available at NRPB for all four globally circulating radionuclides, three of which were developed in-house. Briefly, they are compartmental models in which a compartment may represent the whole of a particular environmental medium, for example soil, on a global basis. The large size of compartments and the assumption that the mixing within them is instantaneous and homogeneous means that the models cannot be used for short-term, local predictions of dispersion from a point source. The models take into account releases to both atmospheric and marine environments, except in the case of krypton-85 for which the model is only relevant for atmospheric discharges. The individual models are briefly described below and summaries of their verification and validation are given in Section 6.2. For a more detailed description see Simmonds *et al*[35].

6.1.1 Tritium

The model adopted by NRPB for the global circulation of tritium is that developed by the National Council on Radiation Protection and Measurements (NCRP) in the USA[69] and is based on earlier work by Easterly and Jacobs[70]. The model, based on the hydrological cycle, assumes that the tritium is released as tritiated water and is used to calculate the specific activity of tritium in water in seven environmental compartments as a function of time following release. The underlying bases for the model are data on the volume of water in each compartment and the transfer rates between compartments, such as those associated with evaporation and precipitation.

6.1.2 Carbon-14

A new global circulation model for carbon-14 was developed by NRPB between 1991 and 1993. The model is based on the carbon cycle and is described by Titley *et al*[71]. The model was developed from a survey of the literature to identify processes and parameters likely to be important, including the possible effects of future climate change. The movement of carbon between compartments was based on estimates of mass transfer, either taken directly from the literature or derived from a combination of parameters.

The model divides the globe into two sections, terrestrial and marine, which are linked by a two-way exchange through the atmosphere and a one-way transfer via river runoff from the terrestrial to the marine environment. It has been assumed that there is no net transfer of carbon between the terrestrial and marine sections of the model. The terrestrial section of the model was taken directly from the literature, while the marine section was developed from an existing compartmental model of the oceans[72], estimates of ocean size[73] and measurements of dissolved carbon[43].

6.1.3 Krypton-85

The model used by NRPB to predict the global circulation of this radionuclide is a simple two-compartment model based on the one developed by Kelly *et al*[35,74]. Krypton-85 discharges are assumed to be uniformly and instantaneously dispersed throughout the troposphere of the northern hemisphere. Exchange takes place between the troposphere of the two hemispheres with a transfer

rate of 0.5 y^{-1}. Within a few years the krypton-85 becomes uniformly mixed throughout the whole troposphere and the only loss from the system is by radioactive decay.

6.1.4 Iodine-129

The compartmental model for the global circulation of iodine-129 currently used at NRPB was developed under an EC contract for the revision of global models of carbon-14 and iodine-129[71]. The model updates the previous NRPB iodine-129 global circulation model developed by Smith and White[75], which was based on a model devised by Kocher[76]. A detailed description of the revised model is given by Titley *et al*[71].

In the model, the compartments represent the principal sectors of the environment (atmosphere, hydrosphere, lithosphere and terrestrial biosphere) in which iodine circulates. The most important modifications to the previous NRPB model were introduced to take into account the suggestion made by Cohen[77] and Fabryka-Martin[78] that iodine in soil is the product of the weathering of sedimentary rocks as well as due to transport from the oceans through the atmosphere. The revised iodine-129 global dispersion model has the same number of compartments (eleven) as Smith and White's model but four more fluxes: three from the sedimentary rock compartment to the other lithosphere compartments and a fourth from the soil solid compartment to the ocean mixed layer compartment. Inventories at steady state of iodine in each compartment and fluxes between them were calculated using environmental measurements and mass balance conditions in each compartment. Iodine-129 released into the environment is assumed to be transported with stable iodine; specific activities of iodine-129 in total iodine in each compartment can therefore be determined by dividing activities of iodine-129 predicted by the model by inventories of stable iodine at steady state.

6.2 Verification and validation

6.2.1 Tritium

The results of the tritium model have been compared with those from two other models[79]. The NCRP model compares well with a model developed by Bergman *et al*[80], the latter predicting greater global collective doses at times greater than about ten years by a factor of about two. The model developed by Kelly *et al*[74] predicts doses about a factor of six lower than the NCRP model.

The validation potential of the tritium model is quite low given the number of sources and the complications introduced by the presence of fallout from nuclear weapons testing. However, the validation of the models using environmental concentrations of tritium from weapons testing in the northern hemisphere has been attempted[79]. This showed good agreement in general between predicted and observed values; however, the model failed to reproduce measured tritium concentrations in marine precipitation in the southern hemisphere. One reason for this is the fact that the model assumes a release into the troposphere, whereas weapons fallout originates in the stratosphere.

6.2.2 Carbon-14

The new model has been verified in three ways. Firstly, the model was run using the initial stable carbon inventories to ensure mass balance between compartments. Secondly, the results of the new model were compared with those predicted by other established models of varying degrees of complexity ranging from a simple six-compartment model[81] to more complex models with relatively detailed ocean[82] or terrestrial[83] sections. The results of the intercomparison are summarised by Titley *et al*[71]. Finally, the dose calculation part of the model was verified by comparison of the predicted collective effective doses and collective effective dose truncated at 500 years from carbon-14

unit releases to relevant compartments, with the doses predicted by various other compartmental models[81–84]. The results of this intercomparison are also summarised by Titley *et al*[71].

Validation of the model has been carried out using two approaches, one using data on natural carbon-14 production and the other using data on weapons testing. The production of natural carbon-14 in the upper atmosphere was used for several separate validation exercises of the global model. Using a natural production rate of 1.075 PBq y^{-1}, the model output was compared with measurements of specific activity concentration in the terrestrial environment and atmosphere, the relative depletion in the specific activity concentrations in the surface and deep ocean compartments compared with the atmosphere, and estimates of the total inventories of carbon-14 in the ocean, atmosphere, sediments, terrestrial biosphere and soil compartments. The distribution of carbon-14 in the environment from atmospheric weapons testing carried out between 1955 and 1970 was compared with the predictions from the model over a relatively short timescale. Using a production rate from weapons testing of 24 PBq y^{-1}, the model output was compared with measurements of the deviation from the pre-weapons-testing specific activity concentrations in the oceans as a function of depth, the total inventories of carbon-14 arising from weapons testing in various compartments and the atmospheric burden of carbon-14 as a function of time. The results of the validation exercises are summarised in Table 6 and

TABLE 6 Summary of the validation of the global circulation model for carbon-14

(a) % deviation from pre-weapons-testing levels

Year	Compartment	Carbon-14 source	% deviation from pre-weapons level	
			Observed	Predicted
1965	Atmosphere	Weapons fallout	60–80	65
1970	Atmosphere	Weapons fallout	55	73
1978	Atmosphere	Weapons fallout	32	37
1985	Atmosphere	Weapons fallout	22	27

(b) Inventory

Year	Compartment	Carbon-14 source	Inventory (PBq)	
			Observed	Predicted
1972	Atlantic	Weapons fallout	28	32
1972	All oceans	Weapons fallout	110	140
Equilibrium	Atmosphere	Natural production	150–160	170
Equilibrium	All oceans	Natural production	7800–8500	8000
Equilibrium	Total	Natural production	8300–9300	8900

described in more detail by Titley *et al*[71]. The results show good agreement between observed and predicted values. The measured percentage deviation from pre-weapons-testing carbon-14 levels in the atmosphere was compared with predicted values for years between 1965 and 1985. Agreement is good and the observed fall in the atmospheric levels following cessation of weapons testing is reproduced by the model with a slight tendency to overpredict concentrations in the later years. Observed and predicted inventories in the world's oceans from weapons fallout were compared for the year 1972. The results showed a degree of overprediction but in general by no more than about 25%. Measured and calculated equilibrium levels due to natural production also compared well.

6.2.3 Krypton-85

Krypton-85 in the atmosphere is dominated by man-made sources, particularly from nuclear fuel reprocessing. The equilibrium global inventory of this radionuclide from natural sources, ie from the spontaneous fission of uranium and the activation of atmospheric krypton-84 by cosmic ray neutrons, is around 440 GBq[85]. This equates to an annual natural production rate of about 30 GBq y^{-1}. For comparison the average annual discharge by BNFL Sellafield during the 1980s was around 40 PBq y^{-1}, ie about one million times greater than global natural production. The contribution to measured air concentrations from weapons testing is also relatively small, NCRP[85] quotes an estimated global average value of 0.02 Bq m^{-3} compared to a measured value of 1.2 Bq m^{-3} in middle latitudes of the northern hemisphere in 1994[86]. This indicates that a very large fraction of measured activity concentrations in air will be due to releases from nuclear fuel reprocessing plants, with the result that if discharges can be quantified reasonably accurately, validation of the model should be relatively straightforward.

A simple validation exercise was carried out using data which approximated to the average annual discharges from the world's major nuclear fuel reprocessing sites and annual average measured air concentrations at mid-latitudes in the northern and southern hemispheres. The predicted air concentrations in both the northern and southern hemispheres for 1994 were within ±10% of the measured data. However, it must be pointed out that the exercise was based on limited data and for a more definitive statement of the global model's accuracy further work is required.

6.2.4 Iodine-129

The predictions of the ratio of iodine-129 to stable iodine have been compared both to measured values and to results from other models. The results show that the predictions are within the range of results from other models and from measurements. The verification and validation work carried out for this model is described in more detail in Appendix B.

7 Assessment applications

7.1 Radiological consequences of routine releases

CREAM (Consequences of Releases to the Environment Assessment Methodology) is a suite of models and data for performing radiological impact assessments of routine and continuous discharges of radioactivity to the environment[35]. PC-CREAM[53*] is a software implementation of CREAM for use

*PC-CREAM is available from NRPB. For further information and an order form write to Mrs A Jones, NRPB, Chilton, Didcot, Oxfordshire OX11 0RQ.

on personal computer. The program can consider the radiological impact of discharges to atmosphere, sea or rivers depending on the location of the site and both individual and collective doses may be calculated. The PC-CREAM suite is composed of the main assessment application which uses datasets of generic model results, and a number of supporting model applications which allow the user to create alternative datasets for use with the assessment application if required. Verification of the software package itself has focused primarily on peer review and extensive software testing; however, the environmental transfer models included in the suite have been subject to their own separate verification and validation as summarised below.

7.1.1 Atmospheric dispersion and deposition

The estimation of the dispersion of radionuclides in the atmosphere is performed using an implementation of the model given by Clarke[6] appropriate for continuous releases. A description of this model and details of its validation are given in Section 2.

7.1.2 Terrestrial foodchain transfer

The estimation of the transfer of radionuclides into foodstuffs is carried out using the FARMLAND model described in Section 3.

7.1.3 Resuspension

The PC-CREAM suite includes a model to predict the activity concentrations in air due to resuspension following deposition. The model is based on a series of wind tunnel experiments supported by field measurements and uses a time-dependent resuspension factor. Its development and validation are described in more detail by Simmonds *et al*[35]. The model is semi-empirical and validation against data not used in the model's development is limited. Garger *et al*[87] described one validation study carried out using data from the Chernobyl nuclear plant accident. The results included a comparison of the annual average resuspension factor estimated by the model with that measured near Chernobyl. Agreement for the year of deposition (1986) and for the following five years was good, with about 70% of predictions within a factor of two of the measured values with a slight tendency to underpredict. However, it is likely that accuracy will decrease with time since deposition.

7.1.4 Global circulation

Collective doses resulting from the global circulation of tritium, carbon-14, krypton-85 and iodine-129 are also calculated by the package. A description of these models and details of their validation are given in Section 6.

7.1.5 Dispersion in marine and freshwater environments

Models are included in the package to predict the activity concentrations in water bodies, suspended sediment, riverbank or inter-tidal sediment, seafoods and freshwater foods. A description of these models and details of their validation are given in Section 4.

7.1.6 External doses

The package also includes models for estimating external doses from gamma and beta irradiation from both airborne and deposited radioactivity. The validation and verification of these models are outside the scope of this report and will be described elsewhere.

7.2 Radiological consequences of accidental releases to atmosphere

Two probabilistic accident consequence assessment program systems (CONDOR[12] and COSYMA[13]) are currently in use at NRPB for calculating the risks posed by potential nuclear accidents giving rise to releases of radioactive material to the atmosphere. The general features of the codes are similar, although they differ in their detailed structure and in some of the models adopted. They model the transfer of radionuclides through the environment, the subsequent dose distributions in the population, the impact of countermeasures which might be introduced to reduce the doses, the health effects in the population and the economic costs of the health effects and countermeasures. The codes can be used to calculate the radiological consequences of a release in a particular set of atmospheric conditions, but are intended primarily for use in probabilistic accident consequence assessments. In these, many sets of conditions are considered and the results for each set are weighted by the probability of occurrence of the conditions, and presented in terms of the probability distribution of consequences. This means that the calculations must be undertaken many times in a single run of the code, and so for reasons of practicality not all the models can be included directly within a code used for probabilistic calculations. Both CONDOR and COSYMA include atmospheric dispersion models directly, but use results of calculations made with dosimetric or foodchain models through data libraries.

It is not possible to validate the whole of a code such as CONDOR or COSYMA. However, validation of the models used for each stage of the calculation has been undertaken, as described in earlier sections of this report. Both CONDOR and COSYMA were included in a recent international comparison of probabilistic accident consequence codes[88]. The spread of predictions from the participating codes varied in size depending on the endpoint, being frequently of the order of two to four, with less difference being seen between the major participating codes, including COSYMA and CONDOR. This gives confidence in the results of the systems.

7.2.1 CONDOR

CONDOR[12] was developed by NRPB, SRD (now AEA Technology Consultancy Services) and Nuclear Electric's Berkeley Technology Centre*. It consists of a single computer program together with a set of appropriate data libraries. The program is modular in structure, with different modules addressing the various steps of the calculation or the presentation of results.

It includes an atmospheric dispersion model based on those recommended by ADMWG and described in Section 2. It also uses foodchain information derived from the FARMLAND model (Section 3.1) or from the Nuclear Electric model FOODWEBB[89].

CONDOR is intended for use on a mainframe computer or a workstation.

7.2.2 COSYMA

COSYMA[13] (COde SYstem from MARIA) was developed by NRPB and Forschungszentrum Karlsruhe GmbH (Germany). It consists of three major subsystems for different distance ranges and different health effects, as illustrated in Figure 8. The NE subsystem calculates early health effects and the actions taken to reduce them. The NL and FL subsystems calculate late health effects and the actions taken to reduce them. Each of these subsystems is a large code divided into separate modules which address the various parts of the calculation. COSYMA also includes a number

*CONDOR could be made available to other users. For further information, contact Dr W Nixon, AEA Technology, Thomson House, Warrington Road, Risley, Cheshire WA3 6AT.

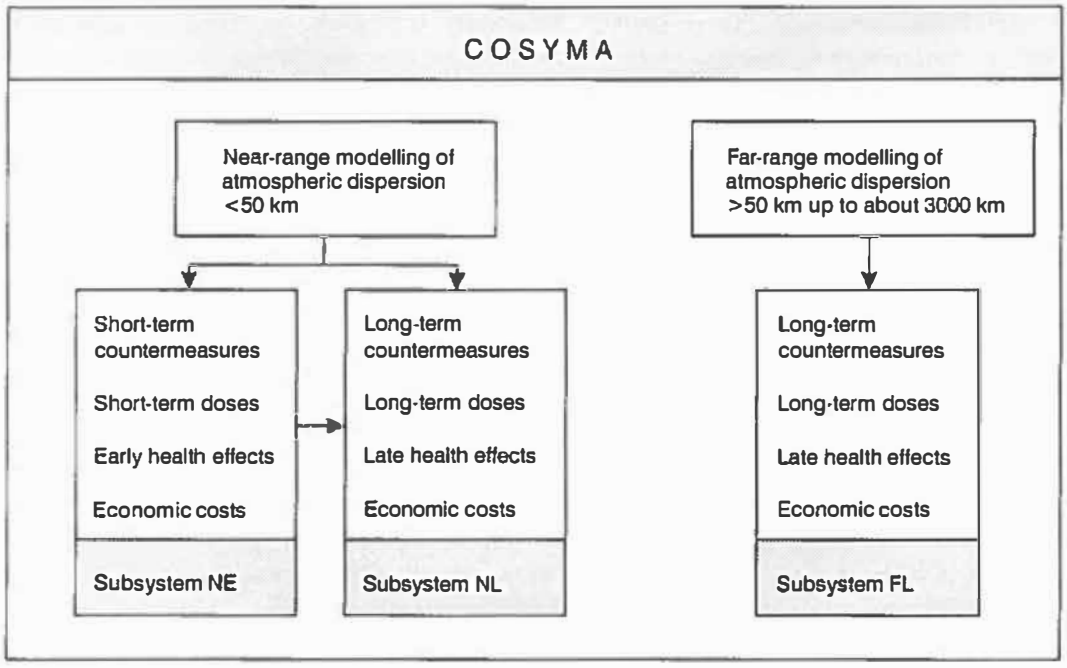


FIGURE 8 COSYMA: general structure of the program system

of separate programs to process input data for use with the system and to present the results in a variety of ways.

The split into separate systems allows different dispersion models, which are appropriate in different distance bands, to be used. The NE and NL subsystems use versions of the Gaussian plume model as described in Section 2. The FL subsystem uses the MESOS model, which is also described in Section 2.

COSYMA uses information on the concentration in foods as a function of time from a single deposit. Two sets of data libraries are available, one derived from FARMLAND (Section 3.1), and the other from the German program ECOSYS[90].

COSYMA is intended for use on a mainframe computer or a workstation. However, a version for use on a PC[91] has also been developed*. This includes many of the facilities and models incorporated in the mainframe version of COSYMA, and can be used for many of the applications for which the mainframe version is intended. The PC version includes a user-friendly interface to control the process of selecting the required endpoints, setting the input data, undertaking the appropriate set of calculations and presenting the results.

7.3 Long-term radionuclide movement in the biosphere following releases from waste repositories

7.3.1 Models

The models in use at NRPB for assessing the transport of radionuclides in the biosphere, and the associated doses to humans, arising from releases to the surface environment from solid waste

*The mainframe and PC versions of COSYMA are available from the European Commission, subject to a signed agreement between the EC and the intending user. This agreement can be obtained from Dr G N Kelly, European Commission, DGXII/F/6, Rue de la Loi 200, B-1049 Brussels, Belgium.

disposal facilities are BIOS[28,92] and MiniBIOS[93*]. Releases from waste repositories are usually only likely to occur in the long term and could be to one or more of several biosphere receptors, such as freshwater streams, soil or the marine environment. BIOS is essentially a combination of models developed in the context of each of the biosphere receptors, joined together so as to provide a complete and connected representation of the surface environment. Consideration is given to local, regional and global sections of the environment so as to permit an evaluation of higher individual doses near to the point of release and local collective doses, as well as the doses occurring from more widespread dispersion of activity (see Figure 9). MiniBIOS is a simplified version of BIOS, which uses less detailed models of farmland, sediments and the oceans, and calculates only individual doses. It was originally developed for use in stochastic analyses, where the time required to run BIOS was too great to allow the large numbers of runs needed for uncertainty or sensitivity analyses. In both BIOS and MiniBIOS, the differential equations are solved using FACSIMILE[51].

Both BIOS and MiniBIOS are compartmental models, and can be used in both generic and site-specific assessments, through suitable choice of local and regional compartments and their associated parameters. Figure 10 shows an example of the compartments and exchanges that can be used to model radionuclide transfer in the environment local to the release point. BIOS and MiniBIOS are dynamic models in the sense that results can be obtained as a function of time, but it is also possible with minor modifications to allow the use of time-dependent model parameters, eg so as to allow representation of the drying out of freshwater bed sediments, or the effects of changing sea level.

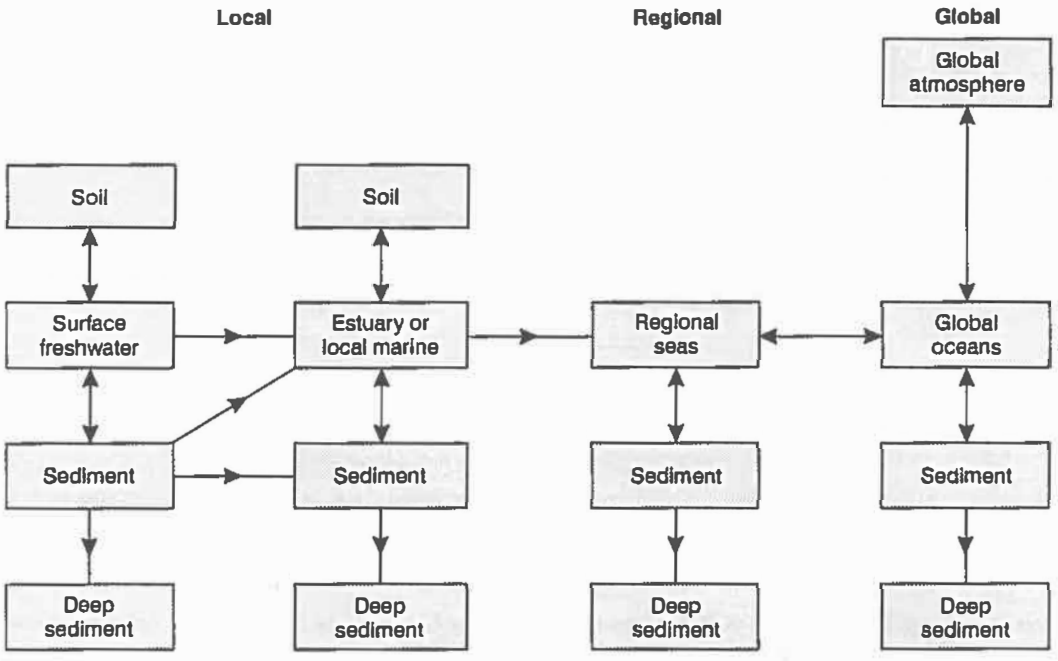


FIGURE 9 BIOS model

*BIOS and MiniBIOS can be supplied to other organisations. Where the proposed use is solely for the purpose of aiding the advancement of radiological protection studies (ie excluding use for financial gain), only a small administrative charge is made. However, potential users should be aware that they will also need to have access to the differential equation solving program FACSIMILE, which is marketed by AEA Technology.

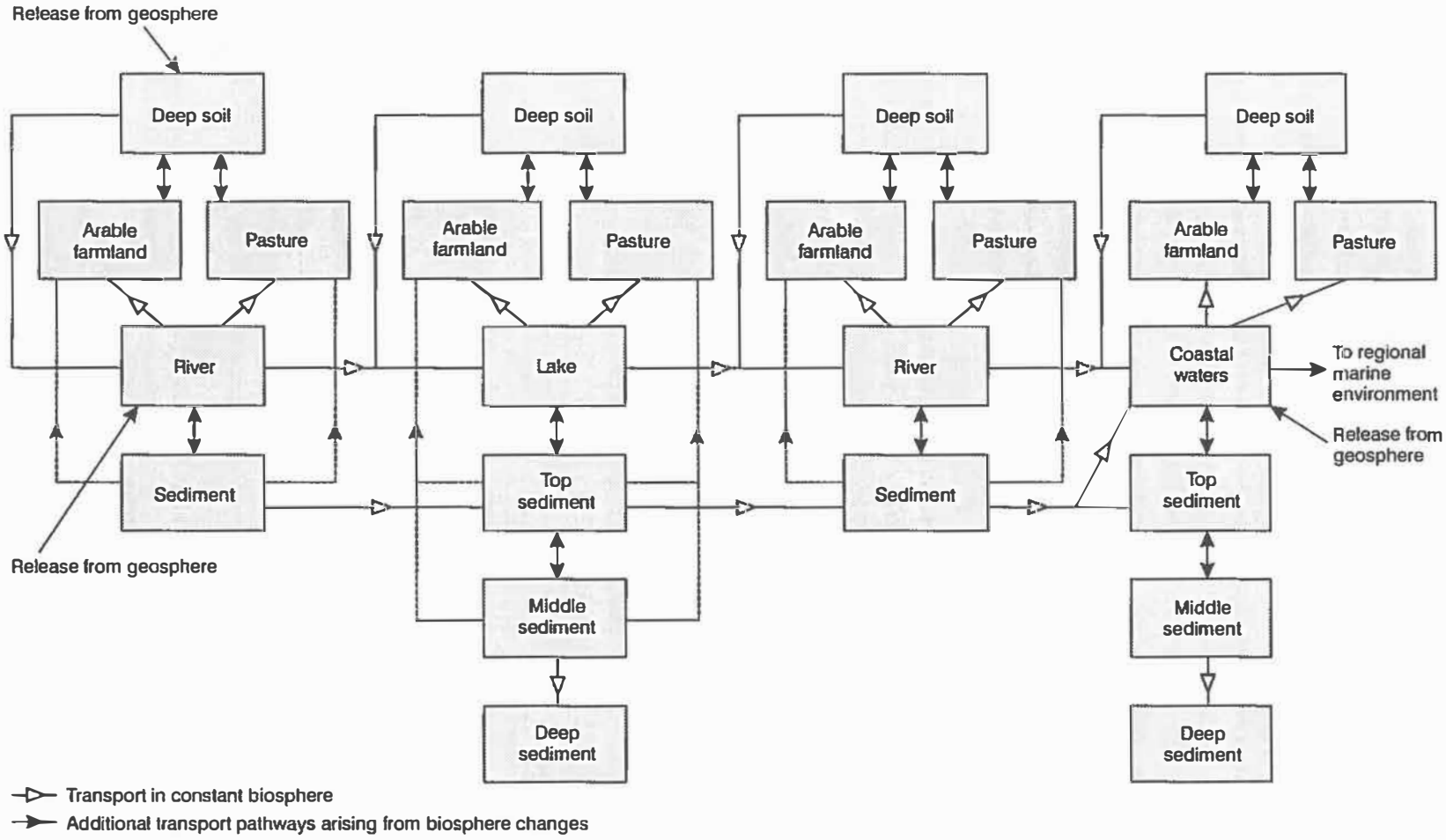


FIGURE 10 Example of a BIOS local environment model

BIOS and MiniBIOS typically cover large spatial scales and long timescales, so the approaches adopted for modelling radionuclide transfer in various parts of the environment tend to be different from those described in Sections 3 and 4. However, they are consistent with them and, where appropriate, the same parameter values are used. Hence the river, lake and associated sediment models (see Figure 10) are the same as those described in Section 4.1.1 and include transfers to arable and pasture land from irrigation, dredging, flooding and changes in the watercourse. However, the soil model is more complex than that described in Section 3.1, because of the need to represent upward migration of radionuclides in soil from sources below the soil as well as downward migration from sources above. Therefore, the BIOS/MiniBIOS soil model has the same structure as the FARMLAND model but uses upward and downward transfers between soil compartments rather than just net downward transfers. The transfers are based on the movement of radionuclides through saturated porous media and the effect of bioturbation in the soil. Erosion of the soil surface layers is also considered.

The foodchain models in BIOS and MiniBIOS are equilibrium versions of the FARMLAND models described in Section 3.1 and are overlaid on to the appropriate soil compartments.

BIOS uses the marine model DORIS to represent dispersion in the seas, whereas MiniBIOS uses a simplified version containing only four water compartments, representing local, regional, continental shelf and deep oceans, respectively.

For uncertainty analyses, BIOS or MiniBIOS is run, along with GEOS (see Section 5), as part of a suite of programs called ESP (Executive Sampling Procedure), that also handles the sampling of input parameter values for multiple model runs.

7.3.2 Verification and validation

BIOS has been verified through internal NRPB procedures, and through model–model comparisons within BIOMOVS (BIOsphere MOdel Validation Study, see Appendix A). The model was also subjected to detailed peer review as a result of its use in the Nirex Research Programme[94]. Updated versions of BIOS are benchmarked against the previous versions to ensure that improvements to particular parts of the model do not adversely affect other parts. MiniBIOS has been verified by means of detailed benchmarking against BIOS (see, for example, Mobbs *et al*[3]), and through model–model comparisons within an NEA Probabilistic System Assessment Code (PSAC) user group and BIOMOVS 2. Both models have also been provided to a number of other organisations, and this has provided further verification.

It is not possible to validate BIOS or MiniBIOS as a whole quantitatively, primarily because of the long-term nature of the calculations for which either model is most commonly used. However, most of the sub-models within BIOS and MiniBIOS are based on models described elsewhere in this report and have been quantitatively validated to some extent, even if only for a few radionuclides and for short-term predictions. Qualitative validation of BIOS and MiniBIOS has been carried out through participation in BIOMOVS and BIOMOVS 2.

8 Summary and conclusions

In this report brief descriptions have been given of a variety of models used at NRPB to represent the transfer of radionuclides through the environment. Such models form an important part of assessments of the radiological impact of releases of radionuclides to the environment. They are essential for predicting future radiation exposures both from current practices and from possible future releases. They also have a major role in supplementing the use of environmental measurements in assessing current radiation doses and in reconstructing doses received in the past. To carry out such

assessments it is important to have reliable models that have been verified and validated to the extent possible. However, it is still important to recognise the uncertainties associated with the use of environmental models and in some cases to quantify such uncertainties.

The extent to which a model can be verified depends on its complexity and whether similar models exist. For relatively simple models verification is straightforward. However, for more complex models verification is not straightforward and has to form part of the development, coding and testing as part of the quality assurance procedures. It is difficult to verify a complex computer model retrospectively.

Validation of models should ideally consist of comparisons between the results of the model and experimental or environmental measurement data that were not used to develop the model. This is more straightforward for some models than for others depending on the quantity and type of data available. For estimating the transfer of radionuclides through foodchains a large body of data is available for model validation, notably as a result of the nuclear reactor accident at Chernobyl. However, even here data are limited to a few key radionuclides and to limited timescales. Validating models becomes increasingly difficult for models which are intended to predict environmental transfer at long times or over great distances, eg the global circulation models, or those to predict the transfer of radionuclides through the geosphere. In all cases it is therefore necessary to adopt qualitative validation techniques to ensure that the model is an adequate representation of the real environment.

Validation of existing environmental transfer models will continue at NRPB as data or other sources of information become available. When new or revised models are developed the verification and validation of the models will form an integral part of their development under the quality management system used.

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APPENDIX A

Some International Verification and Validation Studies

1 VAMP

VAMP (VAlidation of environmental Model Predictions) was a programme for testing and improving biosphere models using fallout data from the Chernobyl nuclear accident, sponsored by IAEA and the EC. The VAMP study started in 1988 and was run as an IAEA coordinated research programme for six years. It focused on the validation of models for the transfer of radionuclides through the terrestrial, aquatic and urban environments. The objectives of the VAMP programme were:

- to provide a mechanism for the validation of assessment models by using the environmental data on radionuclide transfer which have resulted from the Chernobyl release,
- to acquire data from affected countries for that purpose,
- to produce reports on the current status of environmental modelling and the improvements achieved as a result of post-Chernobyl validation efforts.

Four working groups were established to cover the field of radioecological modelling; they were on terrestrial, urban, aquatic and multiple pathways assessment modelling.

Meetings of the full VAMP group have been held annually in Vienna and individual working group meetings have been held in the period between the main meetings. A series of IAEA reports in the Technical Document Series has been published on the findings and results of validation studies carried out by the working groups.

NRPB has actively participated in three of the working groups, on terrestrial, urban and multiple pathway assessment modelling, and has also been involved to a limited extent in the aquatic modelling working group. A member of staff has also acted as a consultant to the urban working group for the writing of the reports on the validation studies carried out by this group.

Further details of the programme can be found elsewhere[1].

2 BIOMOVS

BIOMOVS (BIOsphere MOdel Validation Study) is an international, cooperative effort to test those biosphere models designed to calculate the environmental transfer and bioaccumulation of radionuclides and other trace substances. The first phase of the programme, BIOMOVS 1 (1986–1990), was set up by the Swedish National Institute of Radiation Protection, partly supported by the Nordic Liaison Committee for Atomic Energy. A summary of the work carried out in BIOMOVS 1 and a list of publications are given elsewhere[2,3]. The second phase of the programme which started in 1992, BIOMOVS 2[4], is described in Section 3.

The objectives of BIOMOVS are:

- to test the accuracy of predictions of environmental transfer models for selected contaminants and exposure scenarios,
- to explain the differences in model predictions due to structural deficiencies, invalid assumptions and differences in selected input data,
- to recommend priorities for future research to improve the accuracy of model predictions.

Two approaches were adopted throughout BIOMOVS 1, which are referred to as approaches A and B. Approach A is the testing of models using the process of model validation. Where it was not possible to test the models empirically (as in approach A), an intercomparison of model predictions and associated uncertainty estimates was made for specific assessment questions (approach B). Approach B was adopted for a larger number of test questions and scenarios than approach A.

NRPB participated in a number of the test scenarios and the results of these study scenarios are given elsewhere[5–9].

3 BIOMOVS 2

BIOMOVS 2 is sponsored by the Atomic Energy Control Board of Canada, Atomic Energy of Canada Ltd, Centro de Investigaciones Energéticas, Medioambientales y Technològicas, Spain, Empresa Nacional de Residuos Radiactivos SA, Spain, and the Swedish Radiation Protection Institute.

The aims and general approach of BIOMOVS 2 are the same as those for BIOMOVS 1, with several working groups operating under four main themes:

- scenario development and model intercomparison,
- uncertainties and validation,
- reference biospheres,
- use of post-Chernobyl data for model testing.

The work of some working groups has gone beyond the conventional concepts of verification, validation and model intercomparison. For example, the working group on reference biospheres has been seeking a form of validation for biosphere models for long-term assessments of waste disposal facilities by means of an international consensus on the methodology for developing such models.

NRPB has participated in the model intercomparison working group on complementary studies using MiniBIOS (see Section 7.3 of the main text), and in the working group on reference biospheres.

The BIOMOVS 2 programme finished at the end of 1996, and the reports of the working groups are available from any of the sponsoring organisations.

4 PSAC/PSAG

The Probabilistic System Assessment Code User Group (PSAC) was set up by the Nuclear Energy Agency of the Organisation for Economic Cooperation and Development in 1985[10]. It was later renamed PSAG – the Probabilistic System Assessment Group – and was disbanded in 1994. The terms of reference included:

- exchange of codes, information and experience,
- conducting mutual peer reviews,
- contributing to code justification,
- identifying and discussing technical issues of concern.

In seeking to achieve these aims, PSAC organised a number of international code comparison exercises, known as PSACOIN exercises[10]. NRPB participated in two of these, as follows:

- level E – a study of radionuclide migration in a two-layer geosphere, involving deterministic calculations for comparison with an analytical solution, and stochastic calculations for intercomparison,
- level 1b – an intercomparison study of radionuclide transport in the biosphere, including deterministic and stochastic calculations.

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APPENDIX B

Validation of the Model for the Global Circulation of Iodine-129

1 Introduction

This appendix briefly describes the exercise carried out to validate the NRPB model for the global circulation of iodine-129. In the validation exercise, ratios of iodine-129 to stable iodine in different compartments of the model using known sources of natural iodine-129 were calculated and compared with values obtained from measurements.

2 Sources of natural iodine-129

Iodine-129 is the only naturally occurring radioisotope of iodine. Natural iodine-129 in the hydrosphere and lithosphere is produced mainly by the spontaneous fission of uranium-238; other mechanisms of production of natural iodine-129 in the terrestrial environment are the spontaneous fission of uranium-239 and neutron-induced fission of uranium-235.

The production rate of iodine-129 by the spontaneous fission of uranium-238 ($Q_{I\text{-}129}$) is given by[1]:

$$Q_{I\text{-}129} = \frac{M_{U\text{-}238}\,\lambda_{sp}\,Y_{I\text{-}129}\,\alpha_{I\text{-}129}}{\alpha_{U\text{-}238}} \qquad (1)$$

where $M_{U\text{-}238}$ is the mass of uranium-238, λ_{sp} is the spontaneous fission rate constant for uranium-238, $Y_{I\text{-}129}$ is the fractional spontaneous fission yield at mass 129, and $\alpha_{I\text{-}129}$ and $\alpha_{U\text{-}238}$ are the molecular masses of iodine-129 and uranium-238, respectively.

Iodine-129 is also produced in the upper atmosphere by the interaction of high energy particles (cosmic rays) with xenon and by spallation reactions of isotopes of different elements (mainly tellurium) contained in cosmic dust and meteorites. Kohman and Edwards[2] estimated the production rates of iodine-129 due to these two processes as 2.4 10^{19} atoms and 3.7 10^{17} atoms per year, respectively.

3 Validation exercise

Ratios of iodine-129 to stable iodine (iodine-127) in the compartments of the NRPB iodine-129 global circulation model were calculated by dividing iodine-129 inventories at equilibrium by the iodine inventories at steady state[3]. Equilibrium inventories of iodine-129 were obtained by running the NRPB global circulation model until steady -state conditions were reached using production rates of natural iodine-129 as sources for various compartments. The spontaneous fission of uranium-238, interaction of cosmic rays with xenon and nuclear reactions in cosmic dust were the processes of production of natural iodine-129 considered.

The production rates of natural iodine-129 due to the spontaneous fission of uranium-238 were calculated using equation 1. The mass of uranium-238 in compartment i ($M_{i\ U\text{-}238}$) was calculated from the equation:

$$M_{i\ U\text{-}238} = M_i\, f_{i\ U\text{-}238} \qquad (2)$$

where M_i is the mass of compartment i and $f_{i\ U\text{-}238}$ is the fractional concentration of uranium-238 in the same compartment. Values for λ_{sp} and $Y_{I\text{-}129}$ adopted in the validation exercise were 8.5 10^{-17} y^{-1} and 3 10^{-4}, respectively[1]. Fractional concentrations of uranium-238 in seawater and groundwater were taken from Fabryka-Martin[1], while values for soil and rocks were calculated using an average activity concentration of uranium in soil and rocks[4] of 40 Bq kg^{-1} and the specific activity of uranium-238 (1.245 10^4 Bq g^{-1}). Table B1 gives the iodine-129 production rates due to the spontaneous fission of uranium-238 assumed in the validation exercise; the sedimentary rocks compartment is the most important contributor to the production of natural iodine-129, accounting for more than 99% of the production rate of iodine-129.

TABLE B1 Production rates of natural iodine-129 due to spontaneous fission of uranium-238 in the compartments of the iodine-129 global circulation model

Compartment	Mass (g)	Uranium-238 fractional concentration	Iodine-129 production rate (g y^{-1})
Ocean mixed layer	2.71 10^{22}	3.0 10^{-9}	1.12 10^{-6}
Deep ocean	1.34 10^{24}	3.0 10^{-9}	5.58 10^{-5}
Ocean sediments	6.46 10^{21}	3.2 10^{-6}	2.87 10^{-4}
Shallow groundwater	4.68 10^{21}	1.5 10^{-9}	9.70 10^{-8}
Deep groundwater	3.67 10^{21}	1.5 10^{-9}	7.61 10^{-8}
Soil solid	2.04 10^{20}	3.2 10^{-6}	9.07 10^{-6}
Sedimentary rocks	2.50 10^{24}	3.2 10^{-6}	1.11 10^{-1}

The production of natural iodine-129 by the spontaneous fission of uranium-238 through igneous activity was also considered. Fabryka-Martin[1] estimated that the annual contribution of natural iodine-129 due to the spontaneous fission of uranium-238 in volcanic matter was 2.2 grams. In her model for the global circulation of iodine-129, Fabryka-Martin included explicitly a compartment for igneous activity connected to the sedimentary rocks compartment and to the atmospheric compartments. This compartment is not present in the NRPB model. To take into account the contribution of volcanic matter to the total inventory of natural iodine-129 in the model, an external source representing volcanic activity was connected to the atmospheric compartments. The assumed transfer rates from this source to the atmospheric compartments were those calculated by Fabryka-Martin; they are given in Table B2.

TABLE B2 Parameter values used to calculate production rates of natural iodine-129 due to spontaneous fission of uranium-238 through igneous activity

Parameter	Value
Transfer rate to ocean atmosphere compartment (y^{-1})	7.08 10^{-11}
Transfer rate to land atmosphere compartment (y^{-1})	2.92 10^{-11}
Mass of compartment (g)	6.0 10^{25}
Fractional concentration of uranium-238	3.2 10^{-6}
Source strength (g y^{-1})	2.66 10^{0}

The source strength was calculated using equations 1 and 2 and assuming a fractional concentration of uranium-238 in volcanic matter of $3.2\ 10^{-6}$.

The production rates of iodine-129 in the two atmospheric compartments of the model due to the interaction of cosmic rays with xenon and nuclear reactions in cosmic dust (see Table B3) were calculated using the values suggested by Kohman and Edwards[2] and were taken to be proportional to the volumes of the compartments.

TABLE B3 Production rates of natural iodine-129 due to interaction of cosmic rays and nuclear reactions in cosmic dust in the compartments of the iodine-129 global circulation model

Compartment	Volume (m^3)	Iodine-129 production rate ($g\ y^{-1}$)
Ocean atmosphere	$2.78\ 10^{18}$	$3.70\ 10^{-3}$
Land atmosphere	$1.15\ 10^{18}$	$1.53\ 10^{-3}$

4 Results and conclusions

Table B4 gives a summary of the results obtained. Predicted ratios of iodine-129 to stable iodine range from $1.8\ 10^{-13}$ to $2.8\ 10^{-13}$. These values are generally within the ranges of both theoretical calculations and observations, albeit nearer the lower limits.

Theoretical calculations of ratios of iodine-129 to stable iodine in the hydrosphere[1,5,6,7] are between $3.5\ 10^{-14}$ and $2.2\ 10^{-12}$; Burger[7], for instance, predicted ratios in the range $10^{-13} - 10^{-12}$. Ratios for the ocean region predicted by the NRPB model ($2.5\ 10^{-13}$) are slightly lower than those estimated by Fabryka-Martin[1] ($3.0\ 10^{-13} - 3.0\ 10^{-12}$) but within the range derived from a number of other studies. Measurements of iodine in recharge water taken in the Great Artesian Basin[1] give ratios in the range $4.2\ 10^{-13} - 8.3\ 10^{-13}$ for shallow groundwater and $6.4\ 10^{-13} - 7.1\ 10^{-12}$ for deep

TABLE B4 Ratios of iodine-129 to stable iodine in the compartments of the iodine-129 global circulation model calculated for the validation exercise

	Iodine-129 to stable iodine ratio		
Compartment	NRPB model predictions	Observations*	References
Ocean atmosphere	$2.7\ 10^{-13}$	–	–
Land atmosphere	$2.8\ 10^{-13}$	–	–
Ocean mixed layer	$2.5\ 10^{-13}$	$3.5\ 10^{-14} - 3.0\ 10^{-12}$	1, 5, 6, 7
Deep ocean	$2.5\ 10^{-13}$		
Ocean sediments	$2.5\ 10^{-13}$	–	–
Terrestrial biosphere	$2.3\ 10^{-13}$	–	–
Soil water	$2.3\ 10^{-13}$	–	–
Shallow groundwater	$2.1\ 10^{-13}$	$4.2\ 10^{-13} - 8.3\ 10^{-13}$	1
Deep groundwater	$1.8\ 10^{-13}$	$6.4\ 10^{-13} - 7.1\ 10^{-12}$	1
Soil solid	$2.3\ 10^{-13}$	$2.0\ 10^{-15} - 5.0\ 10^{-12}$	6, 8, 9
Sedimentary rocks	$1.8\ 10^{-13}$		

*Except in the case of the ocean compartments which are theoretical calculations by other workers.

groundwater. These ratios are slightly higher than the values predicted by the NRPB model ($1.8\ 10^{-13}$ for deep groundwater and $2.1\ 10^{-13}$ for shallow groundwater). Ratios of iodine-129 to stable iodine predicted by the model for soil ($2.3\ 10^{-13}$) and sedimentary rocks ($1.8\ 10^{-13}$) are within the wide range of measurements taken in mineral samples ($2.0\ 10^{-15} - 5.0\ 10^{-12}$)[6,8,9].

This validation exercise has shown that the NRPB model for the global circulation of iodine-129 predicts ratios of natural iodine-129 to stable iodine in the environment reasonably well. The model seems to underpredict these ratios in some compartments (groundwater) and this can be attributed to the uncertainties on some parameters used to estimate production rates of iodine-129 and to the fact that not all sources of natural iodine-129 were considered in the exercise (the spontaneous fission of uranium-239 and neutron-induced fission of uranium-235 were not included). Another factor which could have an effect on the ratios predicted by the model is the way in which the production of natural iodine-129 through igneous activity was modelled for this exercise. There are still too few data to assess the significance of the transport of iodine in volcanic matter on its global circulation. Volcanic matter is the main source of natural iodine-129 (see Tables B1–B3) and the model is quite sensitive to the values of the fluxes from the igneous activity compartment to the atmospheric compartments. These fluxes were calculated by Fabryka-Martin[1] using an estimate made by Miyake and Tsunogai[10]. The ratios of natural iodine-129 to stable iodine in some compartments increase by factors between two and three, when these fluxes are increased by a factor of ten.

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